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Simple atomic systems in resonant laser fields

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Contents

1. **Introduction** **1195**
2. **Two-level atom in the field of a monochromatic wave** **1198**
   2.1 Resonance approximation and nonstationary wave functions; 2.2 Adiabatic wave functions in the resonance approximation; 2.3 Quasi-energy, atomic level shifts, and the adiabatic inversion of populations; 2.4 First-order processes in a weak field and radiative decay of adiabatic wave functions; 2.5 Processes of light scattering and absorption when the interaction is suddenly switched on; 2.6 Examination of first-order processes with regard for radiation decay in the steady-state mode
3. **Behaviour of a two-level atom in the pulse light field** **1211**
   3.1 Relation between adiabatic and nonstationary functions; 3.2 Exact solutions for particular pulse shapes; 3.3 'Quasi-classical' approximation in temporal space; 3.4 Adiabatic interaction of light pulse with a two-level system
4. **Interaction of intense radiation with an atom in the presence of two adjacent resonance transitions** **1217**
   4.1 Classification of different interaction schemes and the adiabatic wave functions of three-level atoms in the resonance approximation; 4.2 Self-induced resonance and self-induced adiabatic inversion of a population; 4.3 Non-stationary wave functions and two-photon absorption; 4.4 Permittivity of a three-level atomic gas; 4.5 Interaction of resonance radiation with a three-level system with regard for radiative relaxation processes (degenerate pumping)
5. **Electron Raman scattering with resonant amplification** **1229**
   5.1 Illustrative consideration of the theory of electron Raman scattering; 5.2 Theory of resonant ERS in the monochromatic pumping approximation with regard for saturation effects and Stark shifts; 5.3 ERS in two intense fields; 5.4 Radiative processes in $\Phi_i \leftrightarrow \Phi_j$ transitions; 5.5 Coherent population transfer in the field of two pulses with a time delay; 5.6 Scheme V
6. **Conclusions** **1234**
7. **Appendix** **1235**

References **1235**

Abstract. The interaction of simple, two- and three-level atomic systems with monochromatic and nonmonochromatic laser radiation near resonance transitions is discussed based on the Schrödinger equation with radiation decay and no collisional relaxation, outside the framework of the usual perturbation theory. The mathematical schemes employed include the resonance approximation, exact methods, the quasi-energy method, the adiabatic approximation, etc. A large number of physical phenomena, such as oscillating and inverted populations, self-induced resonances, processes of first order in the weak field, Raman scattering, coherent population transfer, etc. are considered. The review is intended for a broad readership.

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1. Introduction

This review is dedicated to the interaction of simple atomic systems with laser radiation near resonance transitions. In the last few years this field of nonlinear optics has made significant progress thanks to advances and enhancements in laser facilities. Here individual atomic systems are considered in laser fields apart from laser radiation propagation through macroscopic nonlinear media, which until recently has been the major subject of nonlinear optics. Thus, current nonlinear optics can be already divided into two parts: microscopic (the nonlinear optics of individual atoms and molecules) and macroscopic. In this review we discuss some issues of microscopic nonlinear optics.

Note that we do not review works, in which phenomenological decay constants are built into relaxation processes. The radiative decay of atoms in resonant fields is considered only when it can be introduced correctly. Thus, the results that are obtained for times shorter than times of radiative decay can be extended to quasi-stationary systems.

Unfortunately, I have to omit many interesting results on 'nonclassical light' in this review. On the other hand this makes it possible to use only the classical description of electromagnetic fields and the relevant wave function of an atom in resonant fields ('atom + field')† in almost all sections of the review. As a consequence our discussion should be understandable to a broad circle of readers.

In the present review we consider numerous mathematical models since in the region close to resonance interactions the conventional perturbation theory (without summation of an infinite number of Feynman diagrams) used in quantum electrodynamics becomes inapplicable.

Advances in experimental laser facilities achieved in recent years (a new generation of high-yield lasers, the Doppler-free spectroscopy, atomic 'cooling', etc.) have enabled the required experiments to be conducted.

However, as in all problems of this kind, there are common physical points and results, the discussion of which would be very helpful even without going into the mathematical side of the problem. Therefore, prior to the strict consideration of the problem we shall discuss, on the basis of perturbation theory, some common issues of the behaviour of atoms in varying fields using the example of the two-level system.

The atomic wave function in the presence of a radiation field can be found from the Schrödinger equation

$$\mathrm{i}\hbar\,\frac{\partial\Phi}{\partial t} = (\hat{H}_0 + \hat{H}')\Phi\,, \tag{1.1}$$

where $\hat{H}_0$ is an unperturbed atomic Hamiltonian, and in the dipole approximation the interaction potential $\hat{H}'$ for the atom and an external field has the form

$$\hat{H}' = -\hat{\mathbf{d}}\cdot\hat{\mathbf{E}}\,, \tag{1.2}$$

where, in the case of the classical description of the field, it follows

$$\mathbf{E}(\mathbf{r},t) = \vec{\mathcal{E}}\exp(\mathrm{i}\mathbf{k}\cdot\mathbf{r} - \mathrm{i}\omega t) + \text{c.c.}\,, \qquad \vec{\mathcal{E}} = \mathcal{E}\mathbf{e}\,, \tag{1.3}$$

and $\hat{\mathbf{d}}$ is an Hermitian operator of the dipole moment. The field $\mathbf{E}(\mathbf{r},t)$ describes a monochromatic wave with propagation vector $\mathbf{k}$, frequency ω, and polarization vector $\mathbf{e}$. Since the field $\mathbf{E}(\mathbf{r},t)$ is time-dependent, the atom and field can constantly exchange energy and, as a consequence, the atom cannot possess any certain energy. For $\hat{H}' = 0$, the solution of Eqn (1.1) has the form

$$\Psi_m = U_m\exp(-\mathrm{i}E_m t)\,, \tag{1.4}$$

where E_m are the energies (frequencies) of the relevant levels (in s^{-1}), and U_m is the complete system of wave functions of Hermitian operator $\hat{H}_0$.

To integrate Eqn (1.1) with respect to time, we use the initial condition

$$\Phi_n = \Psi_n \quad \text{for} \quad t = -T\,, \tag{1.5}$$

which is physically equivalent to a 'sudden switch-on' of the field $\mathbf{E}$ at $t = -T$.

†The term 'dressed atom' will be used to describe the complete system of 'atom plus quantized field'.

If the conventional procedure of perturbation theory is applied, then the wave function in the first approximation of the perturbation theory can be presented in the form (see, for example, Ref. [1])

$$\begin{aligned}\Phi_n^{(1)} = \Psi_n &+ \sum_{m\neq n}\frac{(\mathbf{d}_{mn}\cdot\vec{\mathcal{E}}^*)\Psi_m}{\hbar(E_{mn}+\omega)}\Big\{\exp\big[\mathrm{i}(E_{mn}+\omega)t\big] \\ &- \exp\big[-\mathrm{i}(E_{mn}+\omega)T\big]\Big\} \\ &+ \sum_{m\neq n}\frac{(\mathbf{d}_{mn}\cdot\vec{\mathcal{E}})\Psi_m}{\hbar(E_{mn}-\omega)}\Big\{\exp\big[\mathrm{i}(E_{mn}-\omega)t\big] \\ &- \exp\big[-\mathrm{i}(E_{mn}-\omega)T\big]\Big\}\,,\end{aligned} \tag{1.6}$$

where the notation

$$E_{mn} = E_m - E_n \tag{1.6$'$}$$

is used for the difference between the energy levels of an atom. Formula (1.6) can be considered as an expansion of the wave function of an atom in the field (1.3) in terms of the complete set of functions of the operator $\hat{H}_0$. This means that the squared absolute value of the coefficient of a function U_m determines the probability that the atom finds itself in the state U_m. As is seen from (1.6), the coefficients of wave functions U_m depend essentially on time and this automatically leads to oscillations of the electron density (population) at the atomic levels with time. Functions of this type will be called nonstationary functions. The dipole moment of the transition $m \to n$ entering (1.6) can be determined by means of the formula

$$\mathbf{d}_{nm} = \int U_n^*\hat{\mathbf{d}}U_m\,\mathrm{d}V\,, \qquad \mathbf{d}_{nm}^+ = \mathbf{d}_{mn}^*\,. \tag{1.7}$$

The average dipole moment is zero for the nondegenerate atomic state, $\mathbf{d}_{mm} = 0$. The last conclusion follows automatically from definition (1.7) and the law of conservation of parity.

In the further discussion we restrict ourselves to the consideration of resonance phenomena, for which the frequency of the electromagnetic field is close to the atomic transition energy (1.6$'$). In this case the condition

$$\frac{|\Delta_{mn}|}{\omega} \ll 1 \tag{1.8}$$

is satisfied for a certain mn. Here Δ_{mn} stands for the resonance detuning

$$\Delta_{mn} = E_{mn} - \omega\,. \tag{1.8$'$}$$

In this case the first sum in (1.6) can be neglected and only the term with the resonance denominator is left in the second sum. We shall number the energy levels of the resonance transition by $m = 2$, $n = 1$ and omit the indices

$$\mathbf{d}_{21} \to \mathbf{d}\,, \qquad \Delta_{21} \to \Delta\,. \tag{1.7$'$}$$

As is seen from formula (1.6), the admixture of 'foreign' states is given by the squared quantity

$$\alpha = \frac{2|\mathbf{d}\cdot\vec{\mathcal{E}}|}{\hbar|\Delta|}\,, \tag{1.9}$$

which is the expansion parameter under resonance conditions. For $\alpha^2 \ll 1$, the ordinary perturbation theory is applicable and this admixture is small. Since the relationship

$$\frac{|\mathbf{d}|}{\hbar|E_{21}|} \sim |\mathcal{E}_{\text{atom}}|^{-1}$$

holds true, in the region far from resonance, i.e. when $\Delta \sim E_{21}$, the parameter α^2 is simply the ratio of the square of the incident field intensity to the atomic field strength squared. This parameter is commonly used in problems of nonlinear (nonresonant) optics. Near the resonance, $|\Delta| \ll E_{21}$, the denominator in (1.9) comes into play and this leads to a significant rise in α^2. The parameter α^2 can be increased in two ways: either by increasing the wave intensity $\mathbf{E}$ or by approaching the exact resonance, $\omega \to E_{21}$. In the first case (and in the absence of resonance) the parameter α^2 is about unity when the electric field of the wave is comparable with atomic fields in its strength. This type of the problem is hard to be solved because all the wave functions of the atom and all the energy levels are to be severely distorted in the field of the wave. In the second case when the resonance occurs, the perturbing field can be considered as being much less than the atomic fields in strength and only two levels, which are in resonance with the external field, will be intensively mixed. In so doing the increase in the parameter α^2 will depend entirely on the accuracy with which the resonance can be approached. Table 1 lists some values of the parameter α (1.9) for various intensities of laser radiation and for various detunings $\tilde{\Delta}$ when $d = 5 \times 10^{-18}$ CGSE. Note that the fourth row of the table presents the value of the quantity

$$2\left|\frac{\mathbf{d}\cdot\vec{\mathcal{E}}}{\hbar}\right| = 2|V|$$

in inverse centimetres. In the scientific literature this quantity is sometimes called the Rabi frequency and represents the frequency for which an electron jumps from level to level back and forth under the action of a strong field $\mathbf{E}$ (see, for example, Ref. [1]). For the purpose of comparison with experimental data it is convenient to express the detuning $\tilde{\Delta} = \Delta/2\pi c$ in inverse centimetres. This quantity $\tilde{\Delta} = \tilde{\nu}_{21} - \tilde{\nu}$, where the atomic transition frequency $\tilde{\nu}_{21} = E_{21}/2\pi c$ and the excitation frequency $\tilde{\nu} = \omega/2\pi c$ are also expressed in inverse centimetres. Inside the area enclosed by the bold line $\alpha > 1$ and the ordinary perturbation theory is inapplicable.

The absolute value squared of the coefficient of Ψ_m gives the probability of the transition W_{mn} from the state Ψ_n to the state Ψ_m in a time $t + T$. For $m = 2$, $n = 1$, we have

$$W' = \alpha^2 \sin^2\left[\frac{\Delta(t+T)}{2}\right] \tag{1.10}$$

(indices are omitted).

In the derivation of (1.10) only the resonant term has been taken into account. The probability that the transition will occur in all the period of time during which the perturbation $\hat{H}'$ takes effect is an oscillating function of time. If the time $t + T$ complies with the condition

$$t + T \gg \frac{2}{|\Delta|}\,, \tag{1.11}$$

the fast-oscillating sinusoidal term can be replaced with its mean value. Then the probability of transition is

$$W' = \frac{\alpha^2}{2}\,. \tag{1.12}$$

If the time interval satisfies the inverse condition to (1.11), the probability of transition in a time $t + T$ is proportional to the time squared:

$$W' = \frac{\alpha^2\Delta^2(t+T)^2}{4}\,. \tag{1.13}$$

Upon dividing by $t + T \to \infty$, expression (1.10) can be transformed to take the usual form for the probability of light absorption by an atom in a unit of time

$$W = \frac{W'}{t+T} = \frac{|\mathbf{d}\cdot\vec{\mathcal{E}}|^2}{\hbar^2}\,2\pi\delta(E_{21} - \omega)\,, \tag{1.14}$$

whence it follows that within the framework of perturbation theory an atom can absorb an energy proportional to the time of the perturbative action at the exact resonance only. The resultant values of W' should always be less than unity since our calculations are based on perturbation theory. However, the probability of transition in a time $t + T$ can become greater than unity for $\alpha^2 > 1$. This indicates that perturbation theory is inapplicable here.

In order to find out how switching the electromagnetic radiation on and off affects the behaviour of an atom, we should give up the monochromatic wave approximation. In studying the resonant interaction between light and atom in a cell, electromagnetic pulses are usually used. The duration of a pulse and, hence, the time of interaction are varied over a very wide range from several femtoseconds (i.e. of the order of 10^{-14} s) to unlimited times pertinent to continuous sources. Currently another design of the experiment is widely used for studying interaction of atomic beams when they pass with a velocity $\mathbf{v}$ through a laser radiation field of length l. In this case the period of interaction $\tau \sim l/v$ can also vary over a wide range. In subsequent sections we present an in-depth discussion of these issues beyond the scope of perturbation theory. However, the results of perturbation theory can be used even in this situation to illustrate the physical nature of the phenomenon and to calculate the probability of atomic transition under the action of a pulse field. For this purpose,

Table 1. Values of the parameter α (1.9).

$I_{\omega i}\delta\omega$, W cm^{-2}	1	10^3	10^6	10^9	10^{12}
$\lvert\mathbf{E}\rvert$, W cm^{-1}	13.5	4.3×10^2	1.35×10^4	4.3×10^5	1.35×10^7
$\tilde{\Delta} = 10^{-3}$ cm^{-1}	2.25	72.4	2.25×10^3	7.24×10^4	2.25×10^6
$\tilde{\Delta} = 1$ cm^{-1}	2.25×10^{-3}	7.24×10^{-2}	2.25	72.4	2.25×10^3
$\tilde{\Delta} = 10$ cm^{-1}	2.25×10^{-4}	7.24×10^{-3}	2.25×10^{-1}	7.24	2.25×10^2

instead of expression (1.3) for a monochromatic field $\mathbf{E}$ we shall use the expression

$$\mathbf{E} = \vec{\mathcal{E}}(t)\exp\left[\mathrm{i}(\mathbf{k}\cdot\mathbf{r} - \omega t)\right] + \text{c.c.}\,, \tag{1.15}$$

where the quantity $\vec{\mathcal{E}}(t)$ defines the pulse shape and varies in time more slowly than $\exp(-\mathrm{i}\omega t)$. If the expression

$$\mathcal{E}(t) = \mathcal{E}_0\exp(-\zeta^2 t^2) \tag{1.16}$$

is used for $\mathcal{E}(t)$, then the probability of atomic transition from a state Ψ_1 to a state Ψ_2 in a time $t \to \infty$ is given as

$$W'(t\to\infty) = \frac{\pi}{\zeta^2}\left|\frac{\mathbf{d}\cdot\vec{\mathcal{E}}_0}{\hbar}\right|^2 \exp\left(-\frac{\Delta^2}{2\zeta^2}\right). \tag{1.17}$$

For

$$2\zeta^2 \gg \Delta^2\,, \tag{1.18}$$

the transition probability is proportional to

$$\left|\frac{\mathbf{d}\cdot\vec{\mathcal{E}}_0}{\hbar}\right|\frac{1}{\zeta^2}\,.$$

The probability of atomic transition is exponentially small when the inverse inequality is true.

It can be shown that for any continuous (together with their derivatives) functions $\mathcal{E}(t)$ the dependence of the atomic transition probability will adhere to an exponential law of the type of (1.17). However, for discontinuous functions the dependence will have a power-law form. For example, if we choose $\mathcal{E}(t) = \mathcal{E}_0\exp\left(-\zeta|t|\right)$, then the transition probability is to be proportional to $|\zeta/\Delta|^2$.

If $\tau \sim 1/\zeta$ determines the typical time of a pulse change, then for

$$|\Delta|\tau \ll 1 \tag{1.19}$$

the atomic transition $1 \to 2$ will actually occur and the upper level will be populated. For the inverse inequality, the probability of such processes is exponentially small.

Thus, even a simple analysis based on perturbation theory brings us to a conclusion that the duration of interaction and how the electromagnetic field is switched on and off are extremely important for considering the interaction between light and an atom near resonance. These issues acquire an even greater meaning beyond the scope of perturbation theory (see, for example, Ref. [2]).

2. Two-level atom in the field of a monochromatic wave

2.1 Resonance approximation and nonstationary wave functions

After the above illustrative consideration based on ordinary perturbation theory we shall come to more rigorous treatment of the problem. Let the frequency of the external electromagnetic field (1.3) be close to the energy difference between two atomic levels, in our case these are the upper level $m = 2$ and the lower level $n = 1$, viz.

$$\frac{|E_{21} - \omega|}{\omega} = \frac{|\Delta|}{\omega} \ll 1\,. \tag{2.1}$$

We shall abstract ourselves for a while from the influence of other atomic levels on the process we are considering. Then the problem is reduced to the interaction between an electromagnetic field and a two-level system. The consideration of such a problem will help to reveal the major features of the resonance interaction between light and an atom.

Due to the influence of the resonance interaction the wave functions of two levels will be mixed. Since in this problem they form a complete set of functions, the exact solution can be sought as a superposition of two atomic states, which are described by the wave functions (1.4):

$$\Phi = a_1(t)\Psi_1 + a_2(t)\Psi_2\,, \qquad |a_1|^2 + |a_2|^2 = 1\,. \tag{2.2}$$

The functions of the atom, Ψ_1 and Ψ_2, which are present in all the subsequent calculations, will match the specific wave functions of the atom, while indices '1' and '2' will comprise the full set of indices for the atomic functions.

The solution of the Schrödinger equation (1.1) will be sought in the form of (2.2). The interaction operator (1.2) is taken as

$$\hat{H}'(r,t) = -\frac{\hat{\mathbf{d}}\cdot\mathbf{E}}{\hbar}\,. \tag{2.3}$$

In matrix elements of interaction we shall leave only slow-oscillating terms

$$H'_{21} = -\frac{\mathbf{d}_{21}\cdot\vec{\mathcal{E}}}{\hbar}\exp(\mathrm{i}\Delta t) = H'^{*}_{12}\,. \tag{2.4}$$

The time-independent part of the interaction operator will be denoted as

$$V = -\frac{\mathbf{d}_{21}\cdot\vec{\mathcal{E}}}{\hbar} = |V|\exp(\mathrm{i}\varphi_0 + \mathrm{i}\pi)\,, \qquad \varphi_0 = \varphi + \varphi_1 - \varphi_2\,, \tag{2.5}$$

where φ_1 and φ_2 are the arbitrary phases of wave functions Ψ_1 and Ψ_2, φ is the constant phase of the field $\mathbf{E}$. Note that $\mathbf{d}_{21}$ enters the formula together with $\mathcal{E}$, i.e. the atomic transition $1 \to 2$ is accompanied by photon absorption; the dipole moment $\mathbf{d}_{12}$ is associated with the amplitude of the field $\mathcal{E}^*$, which describes the photon creation. We recall that in quantum electrodynamics the classical amplitudes $\mathcal{E}^*$ and $\mathcal{E}$ of the field are replaced by the creation and annihilation operators, respectively. In the resonance approximation the Schrödinger equation (1.1) takes the form

$$\mathrm{i}\dot{a}_1 = a_2 V^*\exp(-\mathrm{i}\Delta t)\,, \qquad \mathrm{i}\dot{a}_2 = a_1 V\exp(\mathrm{i}\Delta t)\,. \tag{2.6}$$

To derive (2.6), we have neglected the fast-oscillating terms of the form $\exp\left[\mathrm{i}(E_{21} + \omega)t\right]$ in the right-hand side of (2.3) since they vanish upon averaging over the 'fast' time $t \sim 1/\omega \sim 1/E_{21}$. This procedure can be justified mathematically [3–5]. The resonance approximation will be used repeatedly below. Previously it was used in the magnetic resonance theory [6–9] (the rotating field approximation) and in calculations for laser amplifiers [10, 11]. For a two-level system in the optical frequency range the 'rotating field approximation' was first applied in Ref. [12]. Monograph [13] considers how this method can be applied to solve a number of problems for a two-level atom. In our opinion if the number of levels exceeds two, then the change to a rotating

coordinate system is not efficient and it is more appropriate to use the resonance approximation without such a change (see, for example, Refs [5, 14]). The solution of Eqn (2.6) will be sought in the form

$$a_1 = C\exp(-\mathrm{i}\lambda t)\,, \qquad a_2 = C\,\frac{\lambda}{V^*}\exp\left[-\mathrm{i}(\lambda-\Delta)t\right]. \quad (2.7)$$

For the quantity λ we obtain the quadratic equation

$$\lambda^2 - \lambda\Delta - |V|^2 = 0\,, \quad (2.8)$$

whose solution will be written in the following way

$$\lambda_{1,2} = \frac{\Delta}{2} \mp \sqrt{\frac{\Delta^2}{4} + |V|^2} = \frac{\Delta \mp \Omega}{2}\,. \quad (2.9)$$

In (2.9) we have introduced a new quantity

$$\Omega = \sqrt{\Delta^2 + 4|V|^2}\,, \quad (2.10)$$

and we will use it repeatedly in the subsequent text. The quantities α, $|V|$, Δ, and $\lambda_{1,2}$ are related by the expressions

$$\alpha = 2\left|\frac{V}{\Delta}\right|, \qquad \lambda_1 + \lambda_2 = \Delta\,, \qquad \lambda_1\lambda_2 = -|V|^2\,. \quad (2.11)$$

In what follows we shall frequently say that the interaction is 'switched off'. This means that the solutions are considered in the limit of small $|V|$, or more exactly, for

$$|V|^2 \ll \left|\frac{\Delta}{2}\right|^2. \quad (2.12)$$

This procedure should be taken conventionally since the resultant solutions are valid for constant V. In fact, to consider the particular behaviour of a solution when the interaction is switched off, it is required to solve the problem of the behaviour of a two-level system in a field with a varying $V(t)$. Although the subsequent solutions refer to the behaviour of an atom in the presence of a wave with a constant amplitude we shall, however, switch off the field and compare the resultant solutions with those for an unperturbed system, i.e. as $|V| \to 0$. This procedure makes it possible to establish a correspondence between the functions for an unperturbed atom and those for an atom in the field of a wave. The exact resonance should be approached with a special care because in this case the system 'atom plus electromagnetic field' is degenerate in terms of energy. Figure 1 shows dependences of $\lambda_{1,2}$ as functions of Δ.

The general solution of the Schrödinger equation (1.1) will be an arbitrary combination of the two aforementioned solutions to Eqn (2.6):

$$\Phi = D_1\exp(-\mathrm{i}\lambda_1 t)\left[\Psi_1 + \Psi_2\,\frac{\lambda_1}{V^*}\exp(\mathrm{i}\Delta t)\right] + D_2\exp(-\mathrm{i}\lambda_2 t)\left[\Psi_1 + \Psi_2\,\frac{\lambda_2}{V^*}\exp(\mathrm{i}\Delta t)\right]. \quad (2.13)$$

Two linearly independent orthonormal functions, which are particular solutions of Eqn (1.1), can be extracted from expression (2.13).

Assume that a 'monochromatic' pulse with a rectangular leading edge acts on the atom starting from $t = 0$.

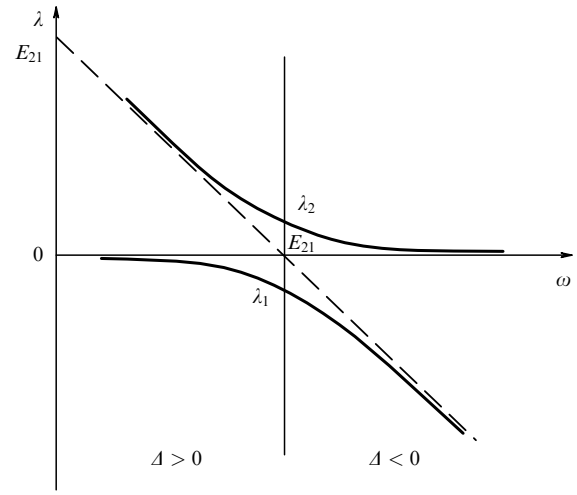

Figure 1. Quasi-energies λ_1 and λ_2 as functions of the electromagnetic field frequency. The asymptotes $\lambda = 0$ and $\lambda = \Delta$ represent the unperturbed states Ψ_1 and Ψ_2. If for $t \to -\infty$ the atomic system is in the left lower branch of λ_1, then it goes from the state Ψ_1 to the state Ψ_2 as the frequency is scanned from $\Delta > 0$ to $\Delta < 0$ and the resonant field is switched off [see Eqn (2.9)].

In fact, as we shall see below, for the case in question the condition $\tau|\Delta| \ll 1$ should be satisfied (here τ is the build-up time of the pulse). In this case the coefficients D_1 and D_2 are determined from the following initial condition

$$\Phi = \Psi_1 \quad \text{for} \quad t = 0\,. \quad (2.14)$$

The wave function has the form (see Ref. [1], where, unfortunately, several errata have crept into the text):

$$\Phi_1' = \Psi_1\exp\left(-\mathrm{i}\,\frac{\Delta}{2}\,t\right)\left(\cos\frac{\Omega t}{2} + \frac{\mathrm{i}\Delta}{\Omega}\sin\frac{\Omega t}{2}\right) - \Psi_2\exp\left(\mathrm{i}\,\frac{\Delta}{2}\,t\right)\frac{2\mathrm{i}V}{\Omega}\sin\frac{\Omega t}{2}\,. \quad (2.15)$$

The second function can be extracted from (2.13) with the use of the initial condition

$$\Phi = \Psi_2 \quad \text{for} \quad t = 0\,. \quad (2.16)$$

Then, one obtains

$$\Phi_2' = -\Psi_1\exp\left(-\mathrm{i}\,\frac{\Delta}{2}\,t\right)\frac{2\mathrm{i}V^*}{\Omega}\sin\frac{\Omega t}{2} + \Psi_2\exp\left(\mathrm{i}\,\frac{\Delta}{2}\,t\right)\left(\cos\frac{\Omega t}{2} - \frac{\mathrm{i}\Delta}{\Omega}\sin\frac{\Omega t}{2}\right). \quad (2.17)$$

It is easy to verify that the functions Φ_1' and Φ_2' form a complete orthonormal set of functions for a two-level atom at each instance of time, i.e. they are subject to the conditions

$$\int \Phi_i^{*\prime}\Phi_k'\,\mathrm{d}V = \delta_{ik}\,. \quad (2.18)$$

Notice that when the interaction is switched off, i.e. for $\alpha \to 0$, it follows

$$\Phi_1' \to \Psi_1\,, \qquad \Phi_2' \to \Psi_2 \quad (2.19)$$

independently of the sign of detuning Δ. The arbitrary constant phase of the wave function Φ_1' is the same as the arbitrary phase of Ψ_1, while the phase of Φ_2' matches the phase of Ψ_2. The states of (2.16) are represented by oscillations of the probability density of the electron between the two states of an unperturbed atom. For example, the probabilities of the electron residence at the upper or lower level in the state described by functions (2.15) are

$$n_2(t) = \left|a_2(t)\right|^2 = \frac{\alpha^2}{1+\alpha^2}\sin^2\frac{\Omega t}{2}\,,$$
$$n_1(t) = \left|a_1(t)\right|^2 = 1 - n_2(t)\,. \qquad (2.20)$$

It should be emphasized that by interchanging the indices $1 \to 2$ and $2 \to 1$ the same expressions will give the populations of the lower and upper levels for the second function Φ_2' (2.17).

Now we shall turn our attention to optical experiments on the observation of population oscillations. In the first optical experiment, G Hocker and C Tang [15] observed oscillations of a population with the Rabi frequency $2|V|$ at the exact single-photon resonance $\Delta = 0$. A decade later M Bassini et al. [16] studied transition processes at the two-photon resonance for Na in the 3S $\to$ 4D transition. These processes are illustrated in Fig. 2. The interaction was switched on for $t_2 = 14$ ns which is less than the 4D-level lifetime of about 50 ns. Thus, the principal conditions of a relaxationless approximation were satisfied. Two-photon absorption occurred in the presence of two opposing waves with mutually orthogonal polarizations and in this way the Doppler effect was suppressed. The two-photon detuning $\Delta = 2\omega - (E_{\rm D} - E_{\rm S})$ varied from zero to 30 MHz. Thus, nonstationary energy functions may justifiably be used even for the maximal detuning $|\tau\Delta| \approx 0.42 < 1$. The effect was observed for spontaneously emitted photons of the 4D $\to$ 3P transition. Since for the fields used in the experiment the quantity α was small, the population of the upper level 4D would oscillate with the frequency $\Omega = \Delta$ according to formula (2.20).

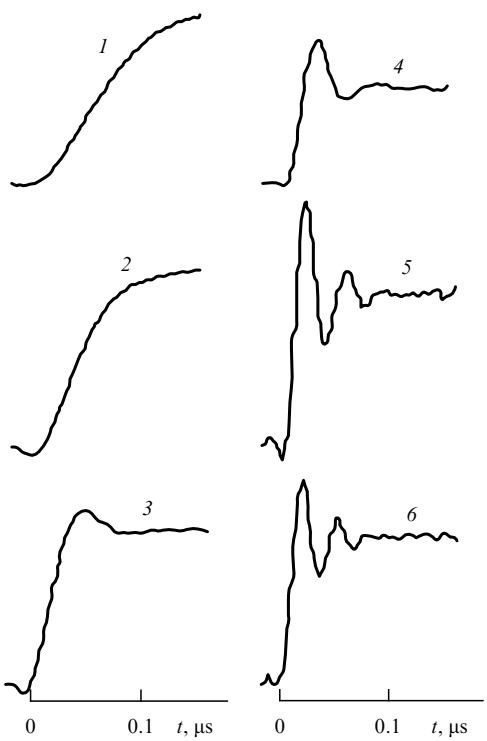

Figure 2. Oscillations of the upper level population as a function of time. The time is measured in microseconds. The population is in relative units. Different curves represent various resonance detunings from $\Delta = 0$ (curve *1*) to $\Delta = 30$ MHz (curve *6*). The experiment was conducted for small field intensities such that $\Omega \approx \bar{\Delta}$, where $\bar{\Delta}$ is the effective value of Δ. The oscillation amplitude decreases due to the spontaneous decay of the upper level.

2.2 Adiabatic wave functions in the resonance approximation

Here we shall consider the behaviour of an atom in the presence of an ideal (i.e. infinite in space and in time) plane monochromatic wave. In this case we cannot set up the state of the atom for $t = t_0$ as we wish. The wave function for $t = t_0$ would result by itself from the solution of the Schrödinger equation without switching the interaction on or off. The last item should be understood in the sense that any switching of the interaction would occur slowly (or adiabatically, as is customary said) so that any stationary state of the atomic system in the absence of a field could be matched with a specific quasi-stationary state of the atomic system in the electromagnetic field. The adiabatic switching lets us exclude transitions between different nondegenerate levels and to consider processes at a fixed atomic level perturbed by the action of a wave.

In problems on the scattering of a monochromatic wave of light, perturbation theory assumes [17] that the inequality

$$(E_{mn} \pm \omega)T \gg 1\,, \qquad (2.21)$$

always holds, i.e. all the periods T characterizing the times of the field switching on and off along with the duration of pulse should be significantly greater than the inverse value of the resonance detuning. In fact, for a monochromatic wave it is required that $T \to \infty$ and, hence, inequality (2.21) holds good automatically. In a real situation T is limited from above due to various effects (the nonmonochromaticity of the pulse, the relaxation and decay of atomic levels, collisional and other relaxation processes). Therefore it is necessary that condition (2.21) be satisfied for the least possible T. Only in this case will the statement of the problem match the scattering of light from an atom in a specified state. Otherwise the problem should be solved for a particular pulse but this makes the consideration less general. Consequently, the fast-oscillating terms can be omitted in (1.6) when the condition (2.21) is satisfied. And this formula can be rewritten as

$$\Phi_n^{(1)} = U_n \exp(-{\rm i}E_n t) + \exp(-{\rm i}E_n t)\sum_{m\neq n}\left[\frac{\mathbf{d}_{mn}\cdot\vec{\mathcal{E}}^*}{\hbar(E_{mn}+\omega)}\exp({\rm i}\omega t) + \frac{\mathbf{d}_{mn}\cdot\vec{\mathcal{E}}}{\hbar(E_{mn}-\omega)}\exp(-{\rm i}\omega t)\right]U_m\,. \qquad (2.22)$$

However, near resonance the first term in the sum (2.22) can be neglected and only the resonant term is left in the second one. In this case it follows from (2.22) that the admixture of the mth state in the nth wave function, which is represented by the square of the absolute value of the coefficient of the function U_m, is time independent. Consequently, unlike (1.6) oscillations of level populations should not occur here with time. Kramers and Heisenberg used just these wave functions in their paper on the scattering of light

by an atom [17]. When the interaction is switched off, i.e. when $\mathbf{E} \to 0$, the function in (2.22) goes over to the unperturbed function of the atom Ψ_n, independently of the sign of Δ. Functions with a similar structure as that of (2.22) will be taken to describe the behaviour of atoms in the monochromatic radiation field in the resonance approximation. To determine these functions, we shall consider the structure of the general solution (2.13). We assume that if the interaction is adiabatically switched off, i.e. when $\alpha \to 0$ or $\mathbf{E} \to 0$, the desired solution should go over either to Ψ_1 or Ψ_2. There are two arbitrary coefficients D_1 and D_2 at our disposal, whose choice determines two linearly independent solutions. We shall choose these coefficients from the following condition. Assume that one of the functions, Φ_1, goes into Ψ_1 and the other goes into Ψ_2 for $\alpha \to 0$ and $\Delta > 0$. Taking into account the limiting values of $\lambda_{1,2}$ for $\alpha \ll 1$ we can easily see that there are two pairs of solutions for the coefficients D_1 and D_2 as $\alpha \to 0$: $D_1 = 1$, $D_2 \to 0$ (it approaches zero faster than α), and $D_1' = 0$, $D_2' = (\mathbf{E}^* \cdot \mathbf{d})/\hbar\Delta$. This choice implies that we use two solutions (2.13): one with λ_1, the other with λ_2. For $\Delta < 0$ and $\alpha \to 0$ we have another pattern: $\Phi_1 \to \Psi_2$, and $\Phi_2 \to \Psi_1$. Thus, we obtain two functions from (2.13) and they are orthonormal. As follows from quantum-electrodynamic considerations these functions are eigenfunctions of the Hermitian operator $\hat{H}$, which describes the system 'atom plus quantized field'. In this case, the complete system 'atom plus quantized field', which has been called a 'dressed atom' (see, for example, Ref. [18]), will possess a specific energy and the above functions will describe the stationary state of the complete system in the resonance approximation. The coefficients chosen, which we will designate as $D_1 = C_1$, $D_2' = C_2$, must satisfy the normalization requirement. This gives rise to the following results (valid for arbitrary α and Δ):

$$|C_1| = \left(1 + \frac{\lambda_1^2}{|V|^2}\right)^{-1/2} = \frac{1}{\sqrt{2}}\left(1 + \frac{\Delta}{\Omega}\right)^{1/2}, \tag{2.23}$$

$$|C_2| = \left(1 + \frac{\lambda_2^2}{|V|^2}\right)^{-1/2} = \frac{1}{\sqrt{2}}\left(1 - \frac{\Delta}{\Omega}\right)^{1/2}. \tag{2.24}$$

From (2.13), we obtain two functions $\Phi_{1,2}$ [4, 5] and in the subsequent discussion we will call them adiabatic functions

$$\Phi_1 = C_1 \exp\left[-\mathrm{i}(E_1 + \lambda_1)t\right]\left\{U_1 + \frac{\lambda_1}{V^*} U_2 \exp(-\mathrm{i}\omega t)\right\}, \tag{2.25}$$

$$\Phi_2 = C_2 \exp\left[-\mathrm{i}(E_1 + \lambda_2)t\right]\left\{U_1 + \frac{\lambda_2}{V^*} U_2 \exp(-\mathrm{i}\omega t)\right\}. \tag{2.26}$$

As all wave functions in quantum mechanics, functions Φ_1 and Φ_2 are accurate to arbitrary phases. Relationships (2.23) and (2.24) specify the absolute values of coefficients C_1 and C_2. Phases of C_1 and C_2 remain unknown. Without any loss in generality C_1 can be considered a real quantity. This can be done because wave functions Ψ_1 and Ψ_2 are also determined to within arbitrary phases, which introduce an additional uncertainty into the process of phase selection for $\Phi_{1,2}$. However, expression (2.25) has a specific structure and arbitrary phases $\varphi_{1,2}$ enter the coefficient of Ψ_2 so that the phase φ_1 can be factored out by braces and combined with the arbitrary phase of the coefficient C_1. Therefore, if C_1 is selected to be real, then the arbitrary phase of Φ_1 will be the same as that of Ψ_1 (for $\Delta > 0$).

The arbitrary phase of the wave function Φ_2 will be specified by the arbitrary phase of Ψ_2 if the arbitrary phase of the constant C_2 is expressed through the arbitrary phase φ_0 as follows:

$$C_2 = |C_2| \exp(-\mathrm{i}\varphi_0 + \mathrm{i}\pi)\,. \tag{2.27}$$

In this case

$$C_2 = -\frac{\lambda_1}{V} C_1\,, \tag{2.27$'$}$$

and formula (2.26) can be rewritten in the following form

$$\Phi_2 = C_1 \exp\left[-\mathrm{i}(E_2 - \lambda_1)t\right]\left[U_2 - \frac{\lambda_1}{V} U_1 \exp(\mathrm{i}\omega t)\right]. \tag{2.26$'$}$$

Using the limiting values of $\lambda_{1,2}$, when the field is switched off ($E \to 0$), we obtain from (2.25) and (2.26$'$) that

$$\begin{aligned} &\Phi_1 \to \Psi_1\,, \quad \Phi_2 \to \Psi_2 \quad \text{for} \quad \Delta > 0\,, \\ &\Phi_1 \to \Psi_2 \exp(\mathrm{i}\varphi_0)\,, \quad \Phi_2 \to \Psi_1 \exp(-\mathrm{i}\varphi_0) \quad \text{for} \quad \Delta < 0\,. \end{aligned} \tag{2.28}$$

In the limit of perturbation theory condition (2.28) presents some problems in the 'passage' through the point $\Delta = \pm 0$ due to the absence of one-to-one correspondence between subscripts of Φ_i and Ψ_j. These problems are due to the fact that the populations are adiabatically inverted in the passage through the point $\Delta = 0$ as we have seen in Section 2.3. In this case, the system 'atom plus field' is degenerate in terms of energy and correct basis wave functions are obtained in the zero-order approximation from (2.25) and (2.26$'$) by setting $\Delta = 0$:

$$\begin{aligned} \Phi_1 &= \frac{\exp(\mathrm{i}|V|t)}{\sqrt{2}}\left[\Psi_1 - \Psi_2 \exp(\mathrm{i}\varphi_0)\right], \\ \Phi_2 &= \frac{\exp(-\mathrm{i}|V|t)}{\sqrt{2}}\left[\Psi_2 + \Psi_1 \exp(-\mathrm{i}\varphi_0)\right]. \end{aligned} \tag{2.29}$$

For $\mathbf{E} \to 0$ and $\varphi_0 = 0$, functions (2.29) go over to symmetric and asymmetric combinations of the functions $\Psi_{1,2}$. These functions remain valid upon substitution $|V|t \to \int |V|\,\mathrm{d}t$ for arbitrary dependence of V on t.

One can easily verify by direct calculation that functions Φ_1 and Φ_2 form a complete orthonormal set of functions for a two-level atom. Functions (2.25) and (2.26$'$) are applicable when the light is essentially monochromatic and the phase φ does not in a typical time for the problem in question. The states which adiabatic functions (2.25)–(2.26$'$) describe, will be called quasi-energy states of the atom (see Section 2.3) in the resonance approximation.

The admixture of functions Ψ_1 and Ψ_2, which comprise the state Φ_1 (and, respectively, Φ_2) is determined by the absolute value of the square of the relevant coefficient in the expansion of Φ_1 in terms of the functions Ψ_1 and Ψ_2. For example, the 'populations' of levels Ψ_1 and Ψ_2 for the function Φ_1 are given by the expressions

$$n_1 = \frac{1}{2}\left(1 + \frac{\Delta}{\Omega}\right), \qquad n_2 = 1 - n_1\,. \tag{2.30}$$

Clearly, the admixture of the function Ψ_2 to the state Φ_2 is given by the expression for n_1, and the admixture of the function Ψ_1 to the state Φ_2 is given by the expression for n_2.

D Grishkovsky [19] conducted an experiment to determine the populations n_1 and n_2 using the optical double-resonance technique. Instead of a monochromatic field, a time-limited pulse was used. With some restrictions, formula (2.30) is, however, applicable in this case as well. A tube with a length of 100 cm was filled with a vapor of rubidium and two pulses with circular polarizations were passed through the tube. The frequency of the first pulse was off-resonant by $\tilde{\Delta} = +1$ cm^{-1} for the atomic transition $5S_{1/2} \to 5P_{1/2}$ ($2\pi\lambda = 7948$ A) in rubidium. The second weak field was either tuned to resonance with the transition $5P_{1/2} \to 6D_{3/2}$ ($2\pi\lambda = 6206$ A), or to the exact two-photon resonance for the two-photon transition $5S_{1/2} \to 6D_{3/2}$. In the first case, the probe field aided in measuring the actual population of the level $5P_{1/2}$, which is due to various relaxation processes and to the absence of monochromatism; in the second case, the 'coherent population', which formulae (2.30) specify, was measured. Tuning to resonance was performed in two ways: firstly, the frequencies of two dye lasers were changed and, secondly, they were finely tuned with the use of a magnetic field. The intensity of the strong laser field as a function of time was measured over the course of the passage through the vapor of rubidium and it reached 500 W cm^{-2} for the time interval 4 ns and linewidth 0.005 cm^{-1}. For the second continuous probe laser the linewidth was narrower than 0.003 cm^{-1}. For the atomic transition $5S_{1/2} \to 5P_{1/2}$, the linewidth was dictated by the spontaneous lifetime $\tau_{sp} = 28$ ns and by the collision time $1/\tau_{sp} = 0.8 \times 10^{-7}\pi N$ ($N = 6 \times 10^{13}$ for 140 °C). The probe pulse did not affect the level populations and its intensity was only 15 mW cm^{-2} in the continuous mode. In experiment [19], the pulse duration was about $\tau \sim 4$ ns and the amplitude changed slowly with time. Therefore, all the relevant phenomena can be described by means of the above formulae, in which the quantity $\alpha^2 \propto \mathcal{E}^2(t)$ is time-dependent. For example, formula (2.30) gives the number of particles at the upper level for $\alpha^2 \ll 1$:

$$n_2 = \frac{\alpha^2}{4}\,. \tag{2.30$'$}$$

This formula defines the coherent excitation of an atom. As is seen from formula (2.25), the 'energy' of such an excitation is $E_1 + \omega$. In this case, the change in the atomic polarization (i.e. the admixture of $5P_{1/2}$ state) will strictly follow the pulse and vanish once the pulse has passed. The number of coherently excited atoms was determined through the absorption of the probe laser, the frequency of which was adjusted so that the exact two-photon resonance occurred. However, besides the coherent 'population' there is a noncoherent population, which is related to the nonmonochromaticity of the pulse as well as to relaxation. The probability of absorption due to the pulse nonmonochromaticity was small. The experiment was performed for values of α of about 0.1 and, hence, nonlinear phenomena were not available for observation (these phenomena manifest themselves for $\alpha > 1$). The necessary calculations were also performed in Ref. [19] using the 'rotating field' method. The results are equivalent to the resonance approximation for $\alpha < 1$, which was employed previously.

2.3 Quasi-energy, atomic level shifts, and the adiabatic inversion of populations

Now we shall introduce the notion of the quasi-energy of an atom in a periodic field [20, 21]. By analogy with Bloch functions for electrons in a space-periodic field in Refs [20, 21], we shall introduce quasi-energy wave functions of an atom in a time-dependent periodic field in the form $\Phi_m = \exp(-\mathrm{i}E_m t)\varphi_m(t, r)$, where $\varphi_m(t, r)$ is a time-periodic function, the period of which is the same as that of the external excitation. By expanding $\varphi_m(t, r)$ into a Fourier series and substituting the series into Φ_m we have

$$\Phi_m = \sum_{s=-\infty}^{+\infty} \Psi_{sm}(\mathbf{r}) \exp(-\mathrm{i}E_m t - \mathrm{i}s\omega t)\,, \tag{2.31}$$

i.e. the same wave function as in Refs [20, 21]. In the scientific literature abroad these states received the name Floquet's states. In a periodic field the quasi-momentum of an electron is specified to within $(2\pi/a)s$ (where a is the lattice constant, and s is an integer) and, in the same way, in a varying electromagnetic field of a frequency ω the energy is specified to within $s\omega$ and is called the quasi-energy. V Ritus [20a] considered the interaction of radiation and an atom in the absence of a resonance. The next step was made in Ref. [4], where resonance interactions were treated. In the resonance approximation that we use, the quasi-energy wave functions become extremely simple and the quasi-energy consideration becomes a very efficient method to solve problems of nonlinear resonant optics (see, for example, Ref. [22]).

Now we want to call the reader's attention to the time dependence of the functions in (2.25) and (2.26$'$). The expressions of interest include terms like $\exp(\pm\mathrm{i}\omega t)$. If we digress from factors proportional to $\exp(\pm\mathrm{i}\omega t)$, then the exponential time dependence for these functions is governed by the factors $\exp\left[-\mathrm{i}(E_1 + \lambda_1)t\right]$ and $\exp\left[-\mathrm{i}(E_2 - \lambda_1)t\right]$, and this corresponds to the structure of stationary states in quantum mechanics. Hence, the quantities $E_1 + \lambda_1$ and $E_2 - \lambda_1$ play the role of 'energy' in a varying field.

Let us consider the case of a positive detuning, $\Delta > 0$. When the field is switched off, the wave functions Φ_1 and Φ_2 go over to the nonperturbed atomic functions Ψ_1 and Ψ_2, respectively, and the quasi-energy levels $E_1 + \lambda_1$ and $E_2 - \lambda_1$ go into unperturbed atomic levels E_1 and E_2. Hence, we can introduce the quantities E_1' and E_2', which play the roles of the 'former' energies E_1 and E_2. In the first function

$$E_1' = E_1 + \lambda_1\,, \tag{2.32}$$

and in the second function

$$E_2' = E_2 - \lambda_1\,. \tag{2.32$'$}$$

In what follows we shall call E_1' and E_2' the reduced quasi-energies of the atom in the lower and upper states or, simply, the 'quasi-energies'. The case $\Delta < 0$ is considered similarly. In this case [since $\Phi_1(\alpha \to 0) \to \Psi_2$, $\Phi_2(\alpha \to 0) \to \Psi_1$]

$$E_1' = E_1 + \lambda_2\,, \tag{2.33}$$

$$E_2' = E_2 - \lambda_2\,. \tag{2.33$'$}$$

Again as in the case of $\Delta > 0$, E_2' is the exponent in front of the function U_2 in the function Φ_1, and E_1' is the exponent in front of the function U_1 in the function Φ_2.

Figure 3 shows energy 'levels' of an atom in the field as a function of Δ, and Fig. 4 presents them as a function of α. The plots show that the system goes away from resonance as α increases†.

† Similar plots for quasi-energies when the interaction is suddenly switched on can be found in many references. According to (3.3) they present a combination of plots shown in Figs 3, 4.

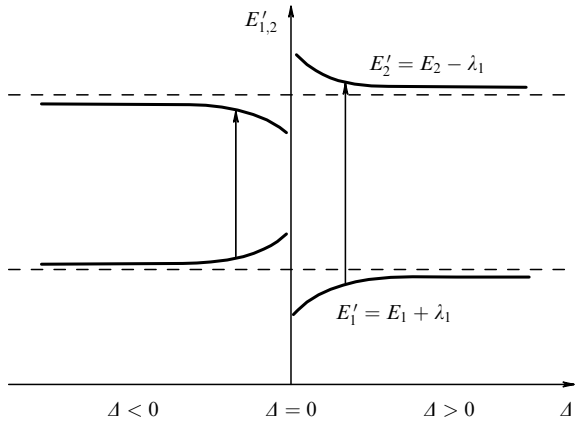

Figure 3. Quasi-energy levels $E'_{1,2}$ of an atom as functions of the detuning Δ. The lower curve represents the quasi-energy of the ground atomic state in the field of a wave and the upper curve represents the quasi-energy of the excited state. The arrows indicate transitions between the atomic 'levels' when absorption or emission occurs. As $\Delta \to \pm 0$, the quasi-energy levels E'_1 and E'_2 split into two sublevels, the spacing between which is $2|V|$.

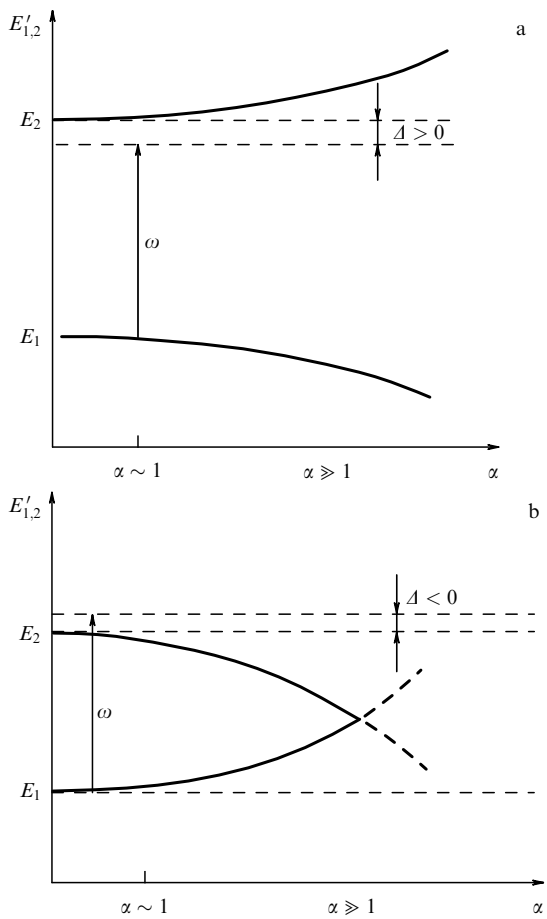

Figure 4. Quasi-energies of the lower E'_1 and upper E'_2 states of an atom as functions of the quantity α for positive (a) and negative (b) detunings. (a) The quasi-energy levels approach each other as the laser field intensity increases, i.e. the resonance detuning grows and the levels become inverted for very large α (dashed line). (b) The quasi-energy levels separate as α increases, i.e. resonance detuning occurs in this case as well.

For the difference $E'_2 - E'_1$ between quasi-energies we have

$$E'_2 - E'_1 = \begin{cases} \omega + \Omega\,, & \Delta > 0\,, \\ \omega - \Omega\,, & \Delta < 0\,. \end{cases} \tag{2.34}$$

It follows from (2.34) and Figs 2 and 3 that, depending on the sign of Δ, the atomic system absorbs radiation of various frequencies, when placed in a resonant field.

The quasi-energies E'_1 and E'_2 (2.32)–(2.33′) become discontinuous as $\Delta \to \pm 0$. At the exact resonance, $\Delta = 0$, two absorption lines and two emission lines, which are different in frequency, will be observed depending on whether we approach the resonance in the frequency ω from below ($\Delta \to +0$) or from above ($\Delta \to -0$). The experimental results (discussed below) support this statement. Clearly, if we consider relaxation, then the splitting of the absorption line into two lines will occur not exactly at $\Delta = 0$ but within the relaxation frequency range from the exact resonance. The cited effect can be interpreted as follows. For example, two radio circuits initially oscillate at the same frequency and then are detuned due to their interaction. In this case we have two close frequencies instead of one. It is well known that two hydrogen atoms are combined in a hydrogen molecule due to the splitting of energy levels of two identical atoms as a result of an exchange interaction. Similar phenomena take place in the example under consideration as well. A two-level system with an electromagnetic radiation at the frequency ω (photon) will correspond to two oscillating circuits (or two hydrogen atoms). In this case we also have a 'coupled' system 'atom plus field' and the accompanying phenomena are similar to those which occur in the process of formation of a molecule. If one wants to delve into the problem, then the resonant field should be quantized and the energy levels of the full system of 'atom plus quantized field' ('dressed atom') (see Fig. 5) should be considered. At this point, there is an analogy with the problem of how a particle can be tunnelled through a barrier between two symmetric potential wells. Studies on this problem can be found in Refs [1, 23] (see also references cited therein).

Level shifts in the field of a wave (high-frequency Stark effect) were established experimentally in the radio-frequency range by French scientists from the group under the supervision of A Kastler for the ground state of a mercury atom [24]. In the optical region level shifts were investigated in potassium vapor by the group under the direction of A M Bonch-Bruevich for absorption lines [25, 26], by the group under the direction of M E Movsesyan for so-called three-photon scattering lines [27], and by the group under the direction of S G Rautian [28]. We consider the results of the second group in Section 2.6.

A M Bonch-Bruevich and his collaborators were the first to reveal and explain the splitting of the resonance absorption line into two components, symmetric about E_{21} for $\Delta \to \pm 0$ in accordance with (2.34). This phenomenon is called sometimes the Autler–Townes effect [29].

The approach or separation of energy levels depicted in Figs 3 and 4 occurs while the resonance approximation holds true, i.e. for $\lambda_1 \ll \omega$ and $\lambda_1 \ll E_{21}$. However, the resultant solutions remain valid and the quasi-energy levels separate arbitrarily far from one another for $\Delta > 0$ and approach for $\Delta < 0$ (and, consequently, intersect and are even inverted) in the case of a magnetic dipole interaction when the resonance approximation is strict. This may occur in the case of a dipole

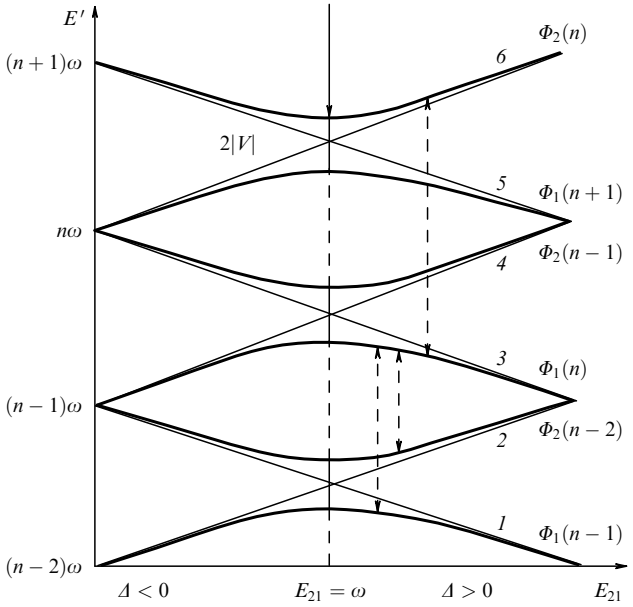

Figure 5. Energy levels of a 'dressed' atom as functions of E_{21}. The solid intersecting straight lines *1–6* represent the energies of the interacting systems $E_1^0(n) = -E_{21}/2 + n\omega$, $E_2^0(n) = E_{21}/2 + n\omega$. For the straight lines *1*, *3*, *5* the atom is 'at the bottom', and for the straight lines *2*, *4*, *6* it is 'at the top'. The curves represent the energy levels of the complete system with regard for their interaction. The shortest distance between two adjacent lines is $2|V|$. The dashed vertical lines match the following processes that we consider in Section 2.4 (from right to left): absorption or emission line $\omega'' = E_{21} - 2\lambda_1$; three-photon emission or absorption line $\omega' = 2\omega - E_{21} + 2\lambda_1$; single-photon emission or absorption at the frequency ω. The energy of the atom is marked from the level midpoint: $E_{21}/2$ when the atom is in the upper state; $-E_{21}/2$ when the atom is in the ground state.

interaction of one definite circularly polarized wave (either σ^- or σ^+) and a two-level system with $m = \pm 1/2$ wherein nonresonance transitions vanish automatically due to the polarization conditions. In the radio-frequency range this phenomenon was observed by physicists from Leningrad and set forth in Refs [25, 26].

Later several papers [30–32] were published in which level shifts were measured in the presence of an intensive resonant wave with better accuracy. P Liao and J Bjorkholm [32] measured the $3P_{3/2}$-level shift for a sodium atom in the field of a pulse laser with wavelength $2\pi\lambda \approx 569$ nm. The pulse had a duration $\tau \approx 5$ ns, a spectral width $\delta\nu \sim 3-50$ GHz and was passed into a cell of length 1 m filled with sodium vapor. The peak intensity reached 5–10 MW cm^{-2} and the sodium atomic number density was 2×10^{12} cm^{-3} (10^{-4} Torr). The laser radiation of 569 nm was close to the resonance transition $3P_{3/2} \to 4D_{5/2}$ and the detuning comprised $\Delta/2\pi = 15$ GHz. The $3P_{3/2}$-level shift was measured by the radiation absorption in the transition with the use of another continuous dye laser of wavelength $2\pi\lambda = 569$ nm with an intensity lower than 0.3 W cm^{-2}. The second laser was tuned to the unperturbed difference between the energy levels $3S_{1/2} \to 3P_{3/2}$ with an accuracy of 11 GHz and 4 GHz. By changing the detuning the authors recorded the $3P_{3/2}$-level shift dependence on the laser intensity. This dependence is proportional to the square of the strong field strength for small intensities and it becomes linear for large intensities in accordance with the theoretical dependence of λ_1 on $|V|^2$.

An interesting phenomenon occurs with an atom in a field when the resonance detuning changes slowly and passes through the point $\Delta = 0$. The passage through the resonance requires a careful consideration. As is seen from formulae (2.32)–(2.34) and from Fig. 3, the quasi-energies E_1' and E_2' ('tied' to the relevant atomic levels) become discontinuous as $\Delta \to \pm 0$, when the field is switched off. On the other hand, if Δ is changed gradually, then the wave functions in (2.25) and (2.26), as well as all the relevant quantities entering them, including λ_1 and λ_2, would change continuously. The procedure of switching-off can be traced in Fig. 1 by moving along continuous curves $\lambda_{1,2}$ which will approach their limiting values $\lambda = 0$ or $\lambda = \Delta$ as $E \to 0$. This means that for the process to be continuous the atom in the field must transit from one state to another by absorbing or emitting a photon, as resonance detuning passes through the point $\Delta = 0$. This optical phenomenon is analogous to the well-known '180° spin direction inversion' at the magnetic resonance [9]. At first we shall consider an initial state which is characterized by the wave function Ψ_1 for the ground state of an atom when $\lambda_1 \to 0$ and $\Delta > 0$. The detuning Δ will be decreased so that it passes through the point $\Delta = 0$ in the direction of negative Δ. If the interaction is switched off once the point $\Delta = 0$ is left behind, then for negative $\Delta < 0$ the wave function Φ_1 is the same as the function Ψ_2 for the excited atomic state up to an arbitrary phase. Hence, if the point is passed adiabatically and then the field is switched off, the atom can be transferred from the ground state to an excited state. The associated experiment was first conducted in the optical region in Refs [33, 34].

M Loy [34] received clear experimental data on population inversion for an NH_3 molecule as a result of its interaction with a resonant light pulse. The wave frequency remained unchanged, and 'the passage through the resonance' was conducted by changing the atomic transition frequency in the NH_3 molecule using the Stark effect [35]. Population inversion can be observed if the upper level decay time T_1 is substantially greater than the time for which the system passes through the resonance. In addition, yet another condition should be satisfied for the process to be adiabatic, i.e.

$$|\mathbf{d} \cdot \vec{\mathcal{E}}|^2 \gg \hbar^2 \left| \frac{\mathrm{d}\Delta}{\mathrm{d}t} \right| .$$

At the magnetic resonance this passage through the resonance is called adiabatic rapid passage (ARP). In Ref. [34], a 10.35 μm CO_2-laser line was used. This line differs by 2.98 GHz from that of the rotational band in an NH_3 molecule. The electric pulse, which is responsible for the Stark effect and shifts the atomic transition frequency, was synchronized with the laser pulse peak. The amplitude of the electric field producing the Stark effect, matched the atomic frequency shift of 800 MHz. The tuning to the resonance started from 500 MHz below the resonance frequency for a specific rotational transition and its rate of change was 4 MHz ns^{-1}. The laser pulse lasted for 5 μs. In the cell, the laser intensity comprised 300 W cm^{-2}, and the transition matrix element was $d \approx 0.24$ D. M Loy [34] clearly observed an inversion of the state in the course of the passage through the resonance depending on the Stark shift magnitude. The laser radiation absorption became negative with time, and this indicated that the state was inverted, namely, the atom went into an excited state. He was also able to measure what

time T_1 it takes for the inverted state to decay and how this time depends on the gas density. The time T_1 was about 1 μs at pressure up to 40 mTorr. The density-independent part ($T_1 = 0.75$ μs) was determined by the transit time for molecules of NH_3 passing through the laser beam section. Hence, $T_1 \Delta \gg 1$ for efficient values of Δ, and adiabatic wave functions could be used to a good accuracy.

In Fig. 3, the cited experiment on passage through the point $\Delta = 0$ corresponds to the following 'motion' along the plotted curves. Assume that for $\Delta < 0$ the starting point is on the left-hand branch of the lower bold curve. As the frequency increases, $\Delta \to -0$, the point will continue moving to the right along this curve. Since $E_2'(+0) - E_1'(-0) = \hbar\omega$, on passage through $\Delta = 0$ from left to the right the system rises, by absorbing a photon of resonance radiation, to the upper right-hand branch of the quasi-energies (see Fig. 3), along which it proceeds as ω decreases for $\Delta > 0$. Thus, the system has jumped from the lower level to the upper level, i.e. a complete inversion has taken place. Certainly, a nonadiabatic passage through the resonance is also possible, when in Fig. 3 the motion proceeds only along the lower discontinuous curve, which represents the lower state of the atom (or along the upper discontinuous curve, which represents the upper state of the atom). Nonadiabatic passage through the resonance can be observed when the laser field frequency (or the atomic transition frequency) changes nonadiabatically so that it compensates for the discontinuity of quasi-energies at the point $\Delta = \pm 0$.

Adiabatic population inversion is thoroughly studied in the case of a two-level system in the optical region as well as in the microwave band. It can be observed in a two-level system either by scanning the pulse frequency or by changing the atomic transition frequency. This makes the experimental design very difficult. As will be shown below, a multilevel system is subject to the same phenomenon. However, unlike a two-level system, a three-level system can adjust itself for specific values of parameters and this leads to a self-induced population inversion.

2.4 First-order processes in a weak field and radiative decay of adiabatic wave functions

In all the above considerations we have implicitly assumed that the energy levels of a separate atom are ideally narrow. In fact they are broadened due to various natural causes. First of all is the so-called radiation width, which all excited states of an atom possess. The radiation width is usually about $10^8\ \mathrm{s}^{-1}$ in the optical region for allowed electric dipole transitions and increases approximately in proportion to the square of frequency, up to the X-ray region. This assertion follows from the formula for the probability of dipole emission of 'atomic oscillators' in the $n \to l$ transition

$$W = \frac{2}{3}\frac{e^2}{\hbar c^3 E_{ln}}|\ddot{x}_{ln}|^2 \approx \frac{1}{3}\frac{e^2}{c^3}\frac{E_{ln}^2}{m}\,, \tag{2.35}$$

in which the uncertainty relation

$$x_{ln} \sim \frac{\hbar}{p_{ln}} \sim \sqrt{\frac{\hbar}{2mE_{ln}}} \tag{2.35$'$}$$

is used to evaluate the matrix elements x_{ln} of the transition. Due to the level de-excitation the system 'atom plus field' can no longer be described by a wave function with a specific quasi-energy. If we restrict ourselves to pure radiative decay, then besides the photons of the intensive resonance field of frequency ω, which have a specific direction and a specific polarization, other photons with wave vectors $\mathbf{k}'$ and polarization $\mathbf{e}'$ have to be considered. These photons appear as a result of the spontaneous decay of excited atomic levels and their intensity is 'weak'. To describe these processes we should go over to a quantized electromagnetic field and adopt the apparatus of quantum electrodynamics since the theory in which the electromagnetic field is described by a classical vector potential does not have a means, without introducing special new rules, to calculate spontaneous emission. The problem which we shall discuss below differs from the common problem of calculating level widths for separate atoms. In fact, the quasi-energy level widths of an atom in a resonance field have to be calculated, i.e. the decay of states Φ_1 and Φ_2 has to be determined. Up to this point the assumption has been made that the atom interacts with the wave for a shorter time lapse than the relaxation times of atomic levels, i.e. we have neglected the decay of functions Ψ_1 and Ψ_2. The mechanisms of relaxation processes are diverse, but they are as yet imperfectly understood in varying external fields. We shall restrict ourselves to consideration only of the radiation width in the presence of an intense field. For $\gamma t > 1$, where γ is the natural width of an excited level of a separate atom, the functions Φ_1 and Φ_2 are no longer eigenstates of the Hamiltonian since they attenuate with a time, $\Gamma^{-1} \sim \gamma^{-1}$. Photons $\omega_{\mathbf{k}'\mathbf{e}'}$, other than photons of the intense resonance field, can be emitted and absorbed due to various processes and will cause decay of the quasi-energy functions. For the purpose of calculation we shall use a common perturbation theory in terms of a 'weak' field (see Fig. 5). First of all is the process of scattering of a resonance photon $\omega_{\mathbf{k}'\mathbf{e}'}$, in which only direction of the wave vector $\mathbf{k}$ changes. This is the so-called unshifted-in-frequency or Rayleigh scattering whose probability can be easily calculated using the standard technique of perturbation theory and wave functions (2.25)–(2.26$'$). It is given by the expression [4]

$$\mathrm{d}W_{\mathrm{R}} = \frac{\alpha^2}{4(1+\alpha^2)}\,\mathrm{d}W_{\mathrm{sp}}\,, \tag{2.36}$$

where $\mathrm{d}W_{\mathrm{sp}}$ is the probability of spontaneous emission for a free atom

$$\mathrm{d}W_{\mathrm{sp}} = \frac{\omega_{\mathbf{k}'}^3}{2\pi\hbar c^3}|\mathbf{e}'^{*}\cdot\mathbf{d}^{-}|^2\,\mathrm{d}O'\,. \tag{2.37}$$

The probability in (2.36) differs from the ordinary quantum-mechanical expression for the spontaneous emission of a dipole with moment $\mathbf{d}^-$ in that the negatively-frequency part of the dipole moment $\mathbf{D}_{11}^-$ of a two-level atom in the resonance field is used in (2.36) instead of the negatively-frequency value of the dipole moment $\mathbf{d}^-$ of a free atom. It can easily be calculated using quasi-energy wave functions (2.25)–(2.26$'$):

$$\begin{aligned}\mathbf{D}_{11} &= \int \Phi_1^* e\mathbf{r}\Phi_1\,\mathrm{d}V \\ &= \mathbf{D}_{11}^-\exp(-\mathrm{i}\omega t) + \mathbf{D}_{11}^+\exp(\mathrm{i}\omega t) = -\mathbf{D}_{22}\,, \\ \mathbf{D}_{11}^- &= -\frac{V}{\Omega}\,\mathbf{d}^*\,.\end{aligned} \tag{2.38}$$

It follows from expression (2.38) that the probability of Rayleigh scattering should be the same for the lower and upper states of the 'atom plus field' system.

J Carlsten et al. [36] measured the Rayleigh scattering intensity as a function of the pumping intensity for various Δ. The authors revealed the saturation effect (for $\alpha^2 > 1$) near the resonance ($\Delta = \pm 0.17$ nm, $\alpha \sim 3$) in accord with expression (2.36). Rayleigh radiation was observed at a right angle to the direction in which the laser beam propagated. The experiment was conducted in a vapor of atomic strontium ($10^{-4} - 10^{-1}$ Torr) using a tunable pulse (5 ns) dye laser, the frequency of which was tuned to the transition $5s^2\,{}^1S_0 \to 5s5p\,{}^1P_1$ (460.73 nm). The results of these experiments are presented in Fig. 6.

Formula (2.36) can be rewritten in a more compact form by integrating over all directions of radiation and by summing over polarizations:

$$\Gamma_{11} = \sum_{\mathbf{e}'} \int \mathrm{d}W_{\mathrm{R}} = \frac{\omega^3 |d|^2}{3\hbar c^3} \frac{\alpha^2}{1+\alpha^2} \,. \tag{2.39}$$

If we introduce the total probability of the spontaneous emission of a free atom for the $\Psi_2 \to \Psi_1$ transition

$$\gamma = \sum_{\mathbf{e}'} \int \mathrm{d}W_{\mathrm{sp}} = \frac{4}{3} \frac{\omega^3}{\hbar c^3} |d|^2 \,, \tag{2.40}$$

then formula (2.39) will take the form

$$\Gamma_{11} = \frac{\gamma}{4} \frac{\alpha^2}{1+\alpha^2} \,. \tag{2.41}$$

It is useful to compare the probability in (2.36), in which the intense field is taken into account, with the common cross section of resonance fluorescence (without consideration for the intense field but considering the decay of the upper level) [37]

$$\mathrm{d}\sigma_0 = \frac{\left|(\mathbf{e}'^* \cdot \mathbf{d}^*)(\mathbf{e} \cdot \mathbf{d})\right|^2}{(E_{21} - \omega)^2 + \gamma^2/4} \frac{\omega^4 \,\mathrm{d}O'}{c^4 h^2} \,. \tag{2.42}$$

The comparison of both expressions shows that the approximation we use in this review is valid when the decay of the upper level γ is negligible compared with the quantity Ω. In other words, multiple oscillations of populations occur between levels for the lifetime of the atom in the upper state $1/\gamma$. For $\alpha \sim 1$, $\omega \sim 10^{15}$, and $\Delta/\omega \sim 10^{-5}$, the quantity Ω is of order $|\alpha\Delta| \sim 10^{10}$ s^{-1}, while the spontaneous width γ is usually of the order of 10^8 s^{-1}.

The spontaneous scattering of light by different atoms is a coherent process because expression (2.38) is independent of the random phases of wave functions of different atoms. Therefore, the total number of scattered quanta will be proportional to the square of the number of atoms located in a volume of the order of λ^3. In volumes whose linear dimensions exceed λ, the interference between different regions in the scattering volume should be taken into account. As a result the electric fields of waves, which have been scattered by different regions, are mutually compensated. Therefore, the Rayleigh scattering becomes zero in a homogeneous medium in volumes much greater than λ^3 (see, however, Section 2.5, where we show that the Rayleigh radiation has noncoherent part when it is suddenly switched on).

We shall now come to Raman scattering from the two-level system 'atom plus field'. For this purpose we shall consider the process in which the $\Phi_1 \to \Phi_2$ transition occurs. The $1 \to 2$ transition is defined by the dipole moment

$$\mathbf{D}_{21} = e \int \Phi_2^* \mathbf{r} \Phi_1 \,\mathrm{d}V \,. \tag{2.43}$$

Using the quasi-energy functions we have

$$\mathbf{D}_{21} = \mathbf{D}_{21}^- \exp\left[-\mathrm{i}(\omega - \Omega)t\right] + \mathbf{D}_{21}^+ \exp\left[\mathrm{i}(\omega + \Omega)t\right] \,. \tag{2.44}$$

The $\Phi_2 \to \Phi_1$ transition is defined by the complex-conjugate quantity

$$\mathbf{D}_{12} = \mathbf{D}_{21}^* \,. \tag{2.45}$$

The quantities $\mathbf{D}_{21}^*$ and $\mathbf{D}_{21}^-$, which enter (2.44), are equal to

$$\mathbf{D}_{21}^+ = \frac{1}{2}\left(1 + \frac{\Delta}{\Omega}\right)\mathbf{d} \,, \tag{2.46}$$

$$\mathbf{D}_{21}^- = -\frac{1}{2}\left(1 - \frac{\Delta}{\Omega}\right)\mathbf{d}^* \exp(2\mathrm{i}\varphi_0) \,, \tag{2.47}$$

where φ_0 is given by expression (2.5). It follows from expression (2.44) that the $\Phi_1 \to \Phi_2$ transition is accompanied by the emission of a photon, the frequency of which is

$$\omega' = \omega - \Omega = 2\omega - E_{21} + 2\lambda_1 \,. \tag{2.48}$$

As is seen from (2.48) for $\Delta > 0$ and $\alpha^2 \ll 1$, a photon with frequency $\omega' \approx 2\omega - E_{21}$ is emitted. The Raman scattering, in the process of which emission with the frequency defined by

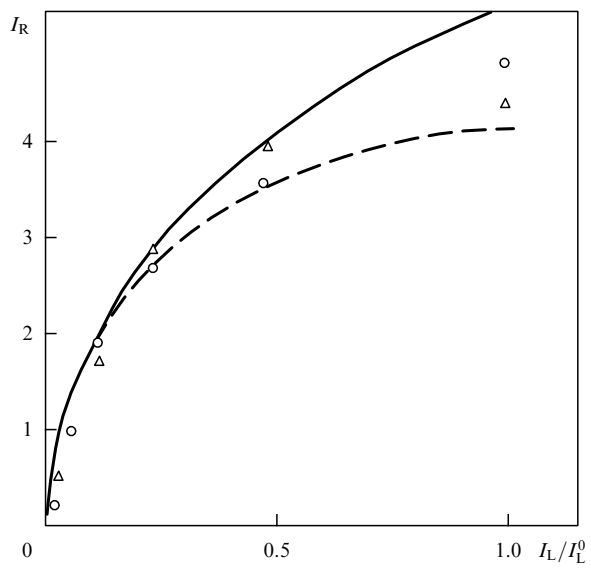

Figure 6. The Rayleigh scattering intensity I_{R} in relative units against the normalized laser intensity $I_{\mathrm{L}}/I_{\mathrm{L}}^0$. Triangles mark the experimental results for a high-frequency detuning of 8 cm^{-1} (0.17 nm) from the resonance transition; circles mark those for a low-frequency detuning by the same amount. The dashed line is the theoretical curve for the assumption that the laser radiation has a rectangular space profile; the solid curve is calculated for the assumption that the space profile has a special shape, which is better suited to describe the actual distribution of the laser radiation intensity in a cell filled with atomic strontium vapor. As is clearly seen, the Rayleigh scattering intensity becomes saturated as α^2 increases [36].

(2.48) occurs for $\Delta > 0$, will be called three-photon radiation process and the relevant frequency will be called the three-photon frequency for $\Delta > 0$. The photon with resonance frequency $\omega'' \approx E_{21}$ is emitted for $\Delta < 0$ and $\alpha^2 \ll 1$ (the $\Psi_2 \to \Psi_1$ transition!). This process is equivalent to the process $\Phi_2 \to \Phi_1$ for $\Delta > 0$. The $\Phi_1 \to \Phi_2$ process is accompanied by the absorption of a photon, the frequency of which is

$$\omega_1'' = \omega + \Omega = E_{21} - 2\lambda_1 \,. \tag{2.49}$$

This frequency is close to the resonance frequency for $\Delta > 0$ and $\alpha^2 \to 0$, and to the three-photon frequency for $\Delta < 0$ and $\alpha^2 \to 0$.

Unlike coherent Rayleigh scattering, the absorption and emission processes occur at different frequencies. These frequencies are symmetric about the frequency ω of the resonant field and, consequently, the stimulated emission and absorption processes cannot compensate each other. Therefore, if photons of frequency ω' are to be found in space, then in the case of the $\Phi_1 \to \Phi_2$ transition, in addition to spontaneous radiation at this frequency, a stimulated radiation arises proportional to the intensity of photons with frequency ω'.

The dependence of $\mathbf{D}_{21}^{\pm}$ on the 'strong' field $\mathbf{E}$ should be given special attention. For $\alpha \ll 1$, the term D_{21}^{+}, which is responsible for the absorption at the frequency ω'', for $\Delta > 0$ is independent of the external field $\mathbf{E}$, while the term D_{21}^{-}, which is responsible for the emission at the frequency ω', for $\Delta > 0$ is proportional to E^2. This means that the cited process appears only in the second-order (conventional) perturbation theory in terms of the resonant field $\mathbf{E}$ (Fig. 7a).

A simple calculation based on the quasi-classical theory of radiation leads to an expression for the probability of three-photon emission of a photon of frequency ω' [4]

$$\begin{aligned} \mathrm{d}W' &= \frac{\omega'^3 |\mathbf{e}'^* \cdot \mathbf{d}^*|^2}{8\pi\hbar c^3 (1+\alpha^2)} \left(2 + \alpha^2 - 2\sqrt{1+\alpha^2}\,\mathrm{sgn}\,\Delta\right) \\ &\quad \times \left(1 + \frac{8\pi^3 c^2}{h\omega'^3} I'\right) \mathrm{d}O' \\ &= \mathrm{d}W_{\mathrm{sp}} \frac{2 + \alpha^2 - 2\sqrt{1+\alpha^2}\,\mathrm{sgn}\,\Delta}{4(1+\alpha^2)} (n_{\mathbf{k}'\mathbf{e}'} + 1)\,, \end{aligned} \tag{2.50}$$

where $I' = I(\mathbf{k}', \mathbf{e}')$ is the spectral-angular density of the intensity of the 'weak' radiation at the frequency ω', and $\mathrm{d}O'$ is either the solid angle for spontaneously emitted photons, or the solid angle in the angular distribution I' for the term with $n_{\mathbf{k}'\mathbf{e}'}$. Formula (2.50) can very easily be derived using the apparatus of quantum electrodynamics and the standard technique of calculations when selecting particular basis functions (2.24)–(2.26′).

For large radiation intensities $\alpha \gg 1$, the probability of emission of a photon for the 'weak field' seeks a saturation and the frequency is given by the expression $\omega' = \omega - 2|V|$. For $\alpha \gg 1$, the probability of three-photon emission becomes of the order of $\mathrm{d}W_{\mathrm{sp}}(n'_{\mathbf{k}'\mathbf{e}'} + 1)/4$, i.e. this quantity is of the order of the probability of emission for a free atom when $n_{\mathbf{k}'\mathbf{e}'} = 0$.

In the optical region, the stimulated radiation of the frequency ω' (2.48) was first observed by the group under the supervision of M E Movsesyan [38]. This process was called a three-photon process because three photons take part in it (in the first nonvanishing order in terms of the strong field strength): two photons are absorbed from the incident beam and one photon is emitted at the frequency ω'. In this process the atom goes from the lower state to the upper state, or more exactly, from the state Φ_1 to the state Φ_2. The authors [38] also measured the dependence of the frequency shift of the three-photon line (2.48) on the radiation intensity (see Fig. 7b). The experiment was conducted for $\alpha \approx 1$ when $|\tau\Delta| > 1$, where τ is the pulse duration. Hence, the adiabatic approximation could be used. The relaxation of levels took about τ and therefore did not perceptibly affect the results.

Now we come to the calculation of the radiation absorption for an atom in a strong resonant field. In the $\Phi_1 \to \Phi_2$ transition, the probability of absorption of a quantum $\mathbf{k}''$, $\mathbf{e}''$ (2.49) from the external beam is given by means of the formula [4]

$$\begin{aligned} \mathrm{d}W'' &= \frac{\pi^2 |\mathbf{e}'' \cdot \mathbf{d}|^2}{c^3 \hbar^2 (1+\alpha^2)} \left(2 + \alpha^2 + 2\sqrt{1+\alpha^2}\,\mathrm{sgn}\,\Delta\right) I'' \,\mathrm{d}O'' \\ &= \mathrm{d}W_{\mathrm{sp}} \frac{2 + \alpha^2 + 2\sqrt{1+\alpha^2}\,\mathrm{sgn}\,\Delta}{4(1+\alpha^2)} n_{\mathbf{k}''\mathbf{e}''}\,, \end{aligned} \tag{2.51}$$

where $I'' = I(\mathbf{k}'', \mathbf{e}'')$ is the spectral-angular density of the probe field intensity. For $\alpha \to 0$ and $\Delta > 0$, expression (2.51) goes into the ordinary coefficient of resonant absorption of a photon $\omega'' \approx E_{21}, \mathbf{k}'', \mathbf{e}''$. This result can easily be understood if we turn our attention to the fact that in essence expression (2.51) describes the absorption coefficient of a two-level system in the intense wave field. Therefore, expression (2.51) should give the ordinary probability of absorption of a photon for the weak field in the approximation of the perturbation theory when the intensity of the wave approaches zero, $\alpha \to 0$. For $\alpha \gg 1$, the absorption frequency shifts, as in the case of the radiation emission frequency, in proportion to the field strength. For large α, the system 'atom

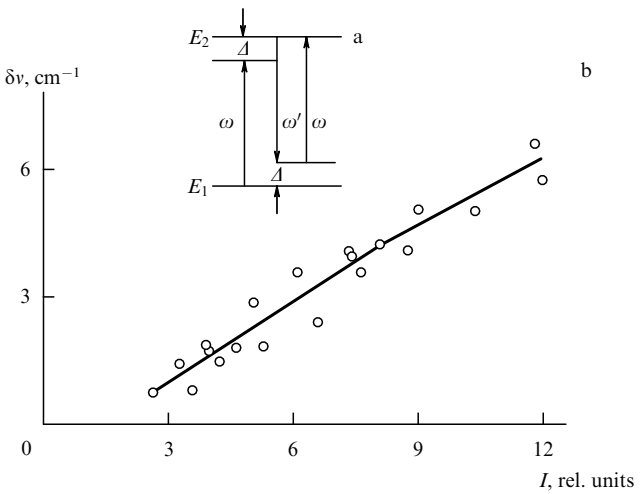

Figure 7. (a) Scheme of the three-photon Raman scattering in the first nonvanishing approximation of the perturbation theory with respect to the field E can be presented as a sequence of three processes: absorption of two quanta ω from the external field, emission at the 'three-photon' frequency ω', and transition of the system from the initial state E_1 to the excited state E_3. (b) The lower figure shows the shift of the three-photon stimulated scattering line $\delta\nu = \lambda_1/\pi$ vs. the exciting radiation intensity. The quantity $\delta\nu$ is plotted against the intensity in relative units. The experiment was conducted for $\Delta \sim 21$ cm^{-1}, $\delta\nu \sim 6$ cm^{-1}, and $\alpha^2 \sim 0.6$, which means that the intensity is $I \sim 30$ MW cm^{-2}. The curve should go through the origin as $I \to 0$. It is not quite clear why the experimental results were different [38].

plus field' absorbs photons at frequencies $\omega'' = \omega + 2|V|$, and for $\alpha^2 \ll 1$ it absorbs at $\omega'' \approx E_{21}$.

All the above peculiarities apply to the case of light scattering from the system 'atom plus field' in the $\Phi_1 \to \Phi_2$ transition. Similar results can be obtained for scattering of light by an atom in the state Φ_2. In this case the process is symmetric, i.e. emission substitutes for absorption and vice versa. The absorption of a photon with frequency ω' would be governed by expression (2.50) with $n_{\mathbf{k}'\mathbf{e}'}$ in place of the factor $n_{\mathbf{k}'\mathbf{e}'} + 1$. The emission of a photon with frequency ω'' would be governed by expression (2.51) with $n_{\mathbf{k}''\mathbf{e}''} + 1$ in place of $n_{\mathbf{k}''\mathbf{e}''}$.

Thus, it follows from (2.50) for $\Delta > 0$ that the total probability of emission of a photon with the frequency ω' given by (2.48) in the $\Phi_1 \to \Phi_2$ transition is

$$\Gamma_{21} = \sum_{\mathbf{e}'} \int \frac{\mathrm{d}W_{\mathrm{sp}}}{4} \frac{\alpha^2}{1+\alpha^2} \frac{\sqrt{1+\alpha^2}-1}{\sqrt{1+\alpha^2}+1} (n_{\mathbf{k}'\mathbf{e}'} + 1) . \quad (2.52)$$

The process of absorption of a photon with frequency ω_1'' (2.49) and with probability $\mathrm{d}W''$ (2.51) will lead to the 'stimulated' width

$$\Gamma_{21} = \sum_{\mathbf{e}'} \int \frac{\mathrm{d}W_{\mathrm{sp}}}{4} \frac{\alpha^2}{1+\alpha^2} \frac{\sqrt{1+\alpha^2}+1}{\sqrt{1+\alpha^2}-1} n_{\mathbf{k}''\mathbf{e}''} . \quad (2.53)$$

If we restrict ourselves to the case $n_{\mathbf{k}'\mathbf{e}'} = n_{\mathbf{k}''\mathbf{e}''} = 0$, then the total width of the quasi-level, which the function Φ_1 represents, depends solely on the spontaneous processes:

$$\Gamma_1 = \Gamma_{11} + \Gamma_{21} = n_2 \gamma , \quad (2.54)$$

where n_2 is the probability that the electron is in the excited state Ψ_2 for the wave function Φ_1. This is given by the second formula in (2.30). The width of the quasi-level, which the function Φ_2 represents, can be obtained in a similar way:

$$\Gamma_2 = n_2' \gamma , \quad (2.54')$$

where n_2' is given by the first formula in (2.30). These two results are very clear. The wave functions $\Phi_1 \exp(-\Gamma_1 t/2)$ and $\Phi_2 \exp(-\Gamma_2 t/2)$ decay as a consequence of the excited Ψ_2-state decay, this state being present in the states Φ_1 and Φ_2 with the probabilities n_2 and n_2', respectively. Hence, the spontaneous lifetime of the state $\Phi_{1,2}$ is of the order of the duration of the allowed transition $2 \to 1$.

As a result of the decay of quasi-energy functions, the quasi-energy wave function, which describes the two-level atom in an intense resonant field, differ for $t > \gamma^{-1}$ from the initial quasi-energy wave function. Over times much greater than the time of spontaneous decay of a separate atom, a statistical radiative equilibrium will be established between the functions $\Phi_{1,2}$, and it will be possible to describe this system by combined quasi-energy functions. Thus, we can also use the quasi-energy functions $\Phi_{1,2}$ over times greater than times of $\Phi_{1,2}$ relaxation [39a]. The first-order processes we have considered here in terms of the 'weak' field are presented in Fig. 5.

The second-order processes in terms of the 'weak' field are considered in Ref. [39b] †. They also cause decay of the quasi-energy wave functions.

† It should be noted that although the second-order processes involving a single atom are much less probable than the above first-order processes, when propagating in a medium they can be more important due to the coherent combining (appeared as parametric processes).

2.5 Processes of light scattering and absorption when the interaction is suddenly switched on

If we start from the solutions given by (2.15) and (2.17) and calculate the atomic polarization and then the probabilities of radiation emission and absorption for an atom in the field under the same assumptions, we obtain different results. Physically this fact can easily be explained: populations start to oscillate when the interaction is suddenly switched on, and therefore all calculations involve transitions between linear combinations of functions rather than pure functions $\Phi_{1,2}$ (see Section 3.1).

Simple calculations yield the following expression for the atomic dipole moment in the state Φ_1':

$$\mathbf{D}_{11} = \mathbf{d}^* \frac{(\vec{\mathcal{E}} \cdot \mathbf{d})}{\hbar\Omega} \left\{ \frac{\Delta}{\Omega} \exp(-\mathrm{i}\omega t) - \frac{1}{2}\left(\frac{\Delta}{\Omega} - 1\right) \exp\left[-\mathrm{i}(\omega - \Omega)t\right] - \frac{1}{2}\left(\frac{\Delta}{\Omega} + 1\right) \exp\left[-\mathrm{i}(\omega + \Omega)t\right] \right\} + \text{c.c.} \quad (2.55)$$

The expressions for the probabilities of spontaneous emission at frequencies ω, $\omega - \Omega$, and $\omega + \Omega$ have the form

$$\mathrm{d}W_{\omega',\mathbf{e}'} = \frac{\omega^3}{8\pi\hbar c^3} \left|\mathbf{d}^* \cdot \mathbf{e}'^*\right|^2 \frac{\alpha^2}{(1+\alpha^2)^2} \, \mathrm{d}O' , \quad (2.56)$$

$$\mathrm{d}W_{\omega-\Omega,\mathbf{e}'} = \frac{(\omega-\Omega)^3}{32\pi\hbar c^3} \left|\mathbf{d}^* \cdot \mathbf{e}'^*\right|^2 \frac{\alpha^2}{(1+\alpha^2)^2} \times \left[\operatorname{sgn}\Delta - \sqrt{1+\alpha^2}\right]^2 \mathrm{d}O' , \quad (2.57)$$

$$\mathrm{d}W_{\omega+\Omega,\mathbf{e}'} = \frac{(\omega+\Omega)^3}{32\pi\hbar c^3} \left|\mathbf{d}^* \cdot \mathbf{e}'^*\right|^2 \frac{\alpha^2}{(1+\alpha^2)^2} \times \left[\operatorname{sgn}\Delta + \sqrt{1+\alpha^2}\right]^2 \mathrm{d}O' . \quad (2.58)$$

Formula (2.56) differs from the similar formula (2.36), which was obtained using quasi-energy functions, by an additional factor $1 + \alpha^2$ in the denominator. For $\alpha \geqslant 1$, this factor is responsible for the faster suppression of Rayleigh scattering.

The above probabilities of radiation emission (2.56)–(2.58) correspond to coherent processes. In these transitions the state of the system 'atom plus field' does not change and upon transition it is described by the same nonstationary wave function Φ_1'. This means that if photons are emitted by different atoms at a specific frequency (ω or $\omega \pm \Omega$), they can interfere with each other.

Besides the aforementioned coherent processes, photons can be noncoherently emitted with frequencies ω and $\omega \pm \Omega$. This will occur when in the resonant field the atom goes from the initial state Φ_1' to the final state Φ_2'. In so doing the transition dipole moment arises and in the case of nonstationary wave functions the moment takes the form

$$\begin{aligned} \mathbf{D}_{21}' = {} & 2\left(\frac{\vec{\mathcal{E}} \cdot \mathbf{d}}{\hbar\Omega}\right)^2 \left[\mathbf{d}^* \exp(-\mathrm{i}\omega t) + \mathbf{d}^* \exp(\mathrm{i}\omega t)\right] \\ & - \left(\frac{\vec{\mathcal{E}} \cdot \mathbf{d}}{\hbar\Omega}\right)^2 \left\{\exp\left[-\mathrm{i}(\omega+\Omega)t\right] + \exp\left[-\mathrm{i}(\omega-\Omega)t\right]\right\} \mathbf{d}^* \\ & + \frac{\mathbf{d}}{4}\left(1 + \frac{\Delta}{\Omega}\right)^2 \exp\left[\mathrm{i}(\omega+\Omega)t\right] \\ & + \frac{\mathbf{d}}{4}\left(1 - \frac{\Delta}{\Omega}\right)^2 \exp\left[\mathrm{i}(\omega-\Omega)t\right] . \end{aligned} \quad (2.59)$$

The first two terms in expression (2.59) bring about spontaneous noncoherent Rayleigh scattering, the probability of which is

$$\mathrm{d}W_{\omega,\mathbf{e}'} = \frac{\omega^3}{8\pi\hbar c^3}\left|\mathbf{d}^*\cdot\mathbf{e}'^*\right|^2\frac{\alpha^4}{(1+\alpha^2)^2}\,\mathrm{d}O'. \tag{2.60}$$

Notice that noncoherent Rayleigh scattering has to be observed in a homogeneous medium as well [39c]. Adding the coherent Rayleigh scattering from (2.56) we can see that the resultant expression is the same as (2.36), which we obtained using quasi-energy wave functions.

The emission and absorption of photons with frequencies $\omega \pm \Omega$ are governed by the subsequent terms in expression (2.59). For example, the fourth term is responsible for radiation emission, while the sixth term is responsible for the absorption of a photon with the frequency $\omega - \Omega$. Upon their summation the radiation emitted in the $1 \to 2$ transition at the frequency $\omega - \Omega$ will be given by the expression

$$\mathrm{d}W_{\omega-\Omega,\mathbf{e}'} = \frac{(\omega-\Omega)^3}{32\pi\hbar c^3}\frac{\left|\mathbf{d}^*\cdot\mathbf{e}'^*\right|^2}{(1+\alpha^2)^2} \times\left[\alpha^4(n_{\mathbf{k}'\mathbf{e}'}+1)-\left(\operatorname{sgn}\Delta-\sqrt{1+\alpha^2}\right)^4 n_{\mathbf{k}'\mathbf{e}'}\right]\mathrm{d}O'. \tag{2.61}$$

The term in the square brackets with $n_{\mathbf{k}'\mathbf{e}'}+1$ allows for the stimulated and spontaneous emission and that with the factor $n_{\mathbf{k}'\mathbf{e}'}$ determines the stimulated absorption. Similarly the expression for noncoherent emission of photons in the $1 \to 2$ transition at the frequency $\omega + \Omega$ is

$$\mathrm{d}W_{\omega+\Omega,\mathbf{e}'} = \frac{(\omega+\Omega)^3}{32\pi\hbar c^3}\frac{\left|\mathbf{d}^*\cdot\mathbf{e}'^*\right|^2}{(1+\alpha^2)^2} \times\left[\alpha^4(n_{\mathbf{k}'\mathbf{e}'}+1)-\left(\operatorname{sgn}\Delta+\sqrt{1+\alpha^2}\right)^4 n_{\mathbf{k}'\mathbf{e}'}\right]\mathrm{d}O'. \tag{2.62}$$

The total probability of emission of a photon with the frequency $\omega - \Omega$ can be obtained by adding the coherent and noncoherent parts, i.e. by summing formulae (2.57) and (2.61):

$$\mathrm{d}W_{\omega-\Omega,\mathbf{e}'} = \frac{(\omega-\Omega)^3}{8\pi\hbar c^3}\frac{\left|\mathbf{d}^*\cdot\mathbf{e}'^*\right|^2}{(1+\alpha^2)^{3/2}} \times\left\{\left[-2(1+\alpha^2)^{1/2}+(2+\alpha^2)\operatorname{sgn}\Delta\right]n_{\mathbf{k}'\mathbf{e}'} +\frac{\alpha^2}{2}\left(\sqrt{1+\alpha^2}-\operatorname{sgn}\Delta\right)\right\}\mathrm{d}O'. \tag{2.63}$$

In a similar manner, the total emission of photons with the frequency $\omega + \Omega$ is determined as

$$\mathrm{d}W_{\omega+\Omega,\mathbf{e}'} = \frac{(\omega+\Omega)^3}{8\pi\hbar c^3}\frac{\left|\mathbf{d}^*\cdot\mathbf{e}'^*\right|^2}{(1+\alpha^2)^{3/2}} \times\left\{\left[-2(1+\alpha^2)^{1/2}-(2+\alpha^2)\operatorname{sgn}\Delta\right]n_{\mathbf{k}'\mathbf{e}'} +\frac{\alpha^2}{2}\left(\sqrt{1+\alpha^2}+\operatorname{sgn}\Delta\right)\right\}\mathrm{d}O'. \tag{2.64}$$

Note that for $\alpha^2 \ll 1$ the probability of stimulated emission of a photon with the frequency $\omega - \Omega$ for $\Delta < 0$ and, which is the same, with $\omega + \Omega$ for $\Delta > 0$ becomes negative. This means that absorption occurs at the atomic frequency. At the exact resonance, $\Delta \to \pm 0$, the absorption line splits into two absorption lines at $\omega \pm 2|V|$ (see Fig. 3). The probability of absorption for $\alpha \ll 1$ is the same as the probability of absorption calculated for a free atom in the weak field. It is important to note that the probabilities of stimulated processes of emission are of different signs for the frequencies $\omega - \Omega$ and $\omega + \Omega$. This means, for example, that for $\Delta > 0$ and $\alpha^2 \ll 1$ the stimulated emission of a photon occurs at the frequency $\omega - \Omega \approx 2\omega - E_{21}$ and the absorption of a photon occurs at the frequency $\omega + \Omega \approx E_{21}$. For $\Delta < 0$, the pattern changes symmetrically. The third terms in formulae (2.63) and (2.64) are responsible for spontaneous emission of quanta with the frequencies $\omega \pm \Omega$. Naturally both terms are positive.

We shall see in Section 2.6 that numerous experiments conducted by different authors generally support the above results of calculations in the case when the interaction is suddenly switched on. Firstly, there is a three-spike structure for spontaneous emission at frequencies ω and $\omega \pm \Omega$ (resonance fluorescence) with the equal probabilities of emission at the frequencies $\omega \pm \Omega$. Secondly, the probe field is absorbed by the system 'atom plus field' at the frequencies which are symmetric about ω, and at the exact resonance, $\Delta \to \pm 0$, the absorption and emission lines split into two lines each (see Fig. 8 and Appendix).

2.6 Examination of first-order processes with regard for radiation decay in the steady-state mode

Over the last decade numerous theoretical and experimental works have been conducted on the measurement of the weak field gain by atoms in the resonant field and on the examination of light scattering near the resonance in a two-level system (resonance fluorescence). These two, at first sight, different phenomena are, as we have seen in Sections 2.4 and 2.5, closely related to each other. To elucidate their relationship we want to call the reader's attention to the fact that the formulae for the probabilities of emission of photons with the frequencies ω and $\omega \pm \Omega$ [see, for instance, Eqns (2.63), (2.64)], presented in the previous sections, include terms whose physical meaning is different. Some terms are proportional to the intensities of created or absorbed photons (stimulated processes), while other terms are independent of the intensities of emitted or absorbed photons (spontaneous processes). The first type of experiments should be designed to measure the characteristics of stimulated processes for an atom in the resonant field or, more exactly, to measure the amplification and decay of the probe signal. The second type of experiments should be designed to study spontaneous processes of atomic decay in the resonant field, i.e. to study resonance fluorescence.

All the experiments we know were generally conducted in conditions where the influence of the radiative decay of the upper level could not be neglected [except the experiments cited in Section (2.4)]. In this situation the expressions we have derived in the previous sections cannot be directly apply to the experimental data.

Resonance fluorescence and absorption of a probe field by an atom in the resonant field under a stationary behaviour, i.e. in the case of the interaction, the duration of which is much longer that the lifetime of the upper level, were studied

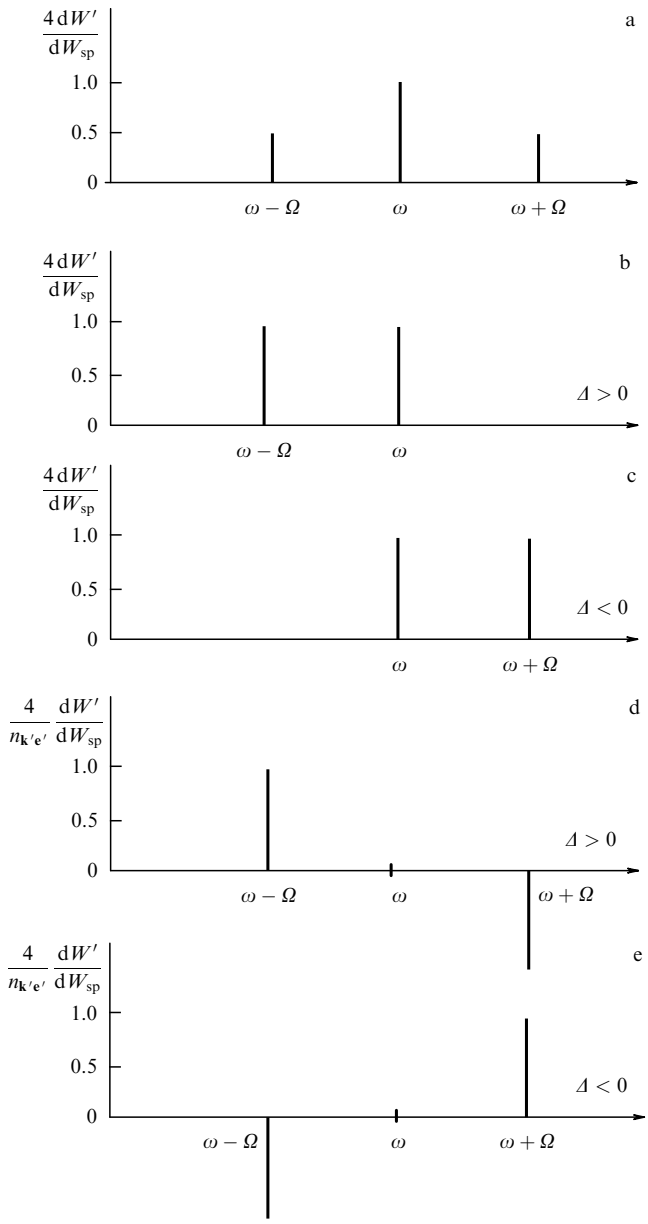

Figure 8. The probabilities of resonance fluorescence in the units of the probability of spontaneous emission of a free atom when the interaction is switched on suddently (a) or adiabatically (b, c). Plot (a) presents also the results from Ref. [40], which have been integrated with respect to the fluorescence line widths. The probabilities of absorption of the 'probe' field (the lines below the abscissa axis) and that of stimulated emission of the 'probe' field (the lines above the abscissa axis) for $\Delta > 0$ (d) and $\Delta < 0$ (e) are given at $\alpha^2 \gg 1$, when the interaction between a strong resonant field and an atom is suddenly switched on. On the abscissa, the probe field frequency is plotted while on the ordinate emission and absorption probabilities W' are plotted in units of $dW_{sp} n_{\mathbf{k}'\mathbf{e}'}/4\alpha$. The results from Ref. [43a] in Fig. 9 have been integrated with respect to the spectrum and in this form they are qualitatively agree with the patterns in Fig. 8. The ordinate values in two lower plots should be multiplied by α when the interaction is switched on adiabatically.

in numerous theoretical and experimental works [39 – 58] and their results are incorporated to reviews and textbooks. Therefore, we shall not discuss these issues in detail here.

However, from the methodological standpoint it is useful to examine whether the quasi-energy adiabatic functions can be applied as a simple way to describe resonance fluorescence and accompanying processes in the stationary regime. Relaxation processes are one of the factors restricting the use of quasi-energy states for $t > 1/\gamma$. If we consider only one separate atom in a strong field, then the atom would drop back, due to spontaneous emission, from the initial quasi-energy state to the ground state in a time of the order of the natural lifetime $1/\gamma$ of a free atom, as described in Section 2.4. Clearly, reverse transitions up to the initial quasi-energy state are also possible and they are accompanied by absorption of a photon with the relevant frequency. One may expect that the system 'atom plus classical strong field' will ultimately go into a state of dynamic radiative equilibrium, which is described by a noncoherent mixture of quasi-energy states. There is sense in talking of quasi-energy features pertinent to this equilibrium state only when the levels do not overlap (remember that they are broadened due to radiative processes). In other words, typical gaps between quasi-energy levels must be significantly wider than the level widths. In this case the quasi-energy approach can still be used. This project was realized in Ref. [39a], in which the familiar results of B Mollow [40] were obtained in a simple way and the problem of scattering for the nonmonochromatic pulse was also considered.

Here we shall cite several experiments on the interaction of a strong field with a two-level system in the steady-state mode. Interesting results in this line were obtained by a group of physicists from Leningrad in Ref. [41], where the authors measured the shape of the absorption line for a two-level system in a strong radio-frequency field. They checked their results against the calculations in Ref. [42]. The results of these experiments are in good accord with the positions and shifts of lines of three-photon emission and of the absorption line. The authors of Ref. [41] clearly observed how absorption lines split for $\Delta = 0$.

Optical experiments on the measurement of the absorption coefficient for an 'atom in field' and on resonance fluorescence were conducted by the group under the supervision of S Ezekiel [43 – 45] for beams of sodium atoms in the optical region. Figure 9 shows experimental results from Ref. [43a]. These results fit well with the calculations in Ref. [40]. The experiment was conducted using beams of sodium atoms; the $3\,^2S_{1/2}(F=2) \to 3\,^2P_{1/2}(F'=3)$ transition was studied. The two-level system was specially 'prepared' by pumping over atoms of sodium to a given sublevel ($F = 2$, $m_F = 2$) by means of circularly polarized light. Then the sodium atoms got into the region of interaction with a strong circularly polarized field, which affected only two specific levels $F = 2$, $m_F = 2$ and $F = 3$, $m_F = 3$. Simultaneously, the attenuation or amplification of the weak probe field was recorded. Note that the frequency of this field was scanned, and thus the contours of absorption and emission lines were obtained. The light beams of the probe and intense resonant fields were mutually orthogonal and, in addition, they were orthogonal to the direction of the atomic beam. Typical values of the parameters were 3 μW for the probe beam intensity, up to 560 mW for the resonant radiation intensity, $\gamma/2\pi = 11$ MHz for the width of the resonance transition line, and 0 to $-\Delta/2\pi \sim 80$ MHz for the probe field detuning. For the peak detuning, $\Delta \gg \gamma$, the Rabi frequency was $\Omega_R = 2\pi \cdot 60$ MHz. Hence, the parameter α^2 may be much greater than unity. The velocity of sodium atoms was 10^5 cm s^{-1}, and the interaction length was about a millimetre. As a result, the interaction time (i.e. the time it takes to transit through the region of intense radiation) was of order

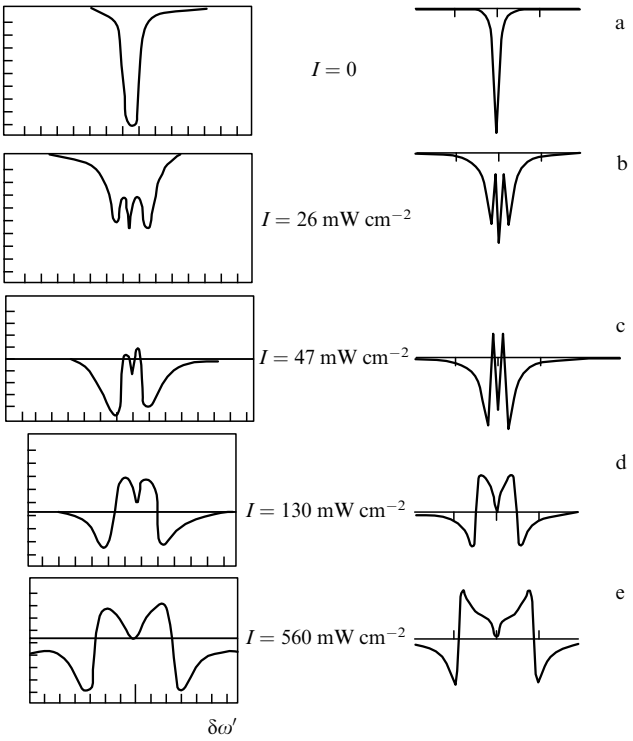

Figure 9. Absorption line contours for a weak signal with frequency ω' in an 'atom plus resonant field' system depending on $\delta\omega' = \omega' - E_{21} = -\Delta$ (the plots are taken from Ref. [43a]). The curves represent the exact resonance, $\Delta = 0$, and different pumping intensities from $I = 0$ to $I = 560$ mW cm^{-2}. The left graphs pertain to the experiment, while the right ones are concerned with the theory.

10^{-6} s. This means that the condition $\tau\gamma \geqslant 1$ of stationary behaviour was satisfied in the experiments with beams.

Research of the resonance fluorescence of the system 'atom plus strong field' was initiated by F Shuda et al. [46]. Since then many experiments have been conducted to measure the spectrum of resonance fluorescence. In Ref. [44], sodium atoms were excited by radiation resonant to the $^2S_{3/2}(F = 2) \to {}^2P_{3/2}(F' = 3)$ transition. The typical values of α met the requirement $\alpha^2 \gg 1$. Therefore, as we noted earlier, the resonance fluorescence spectrum is to be symmetric about the central frequency and the areas under the curves for photon emission at the frequency $\omega \pm \Omega$ must be about one-half the intensity of the Rayleigh scattering without frequency shift. The width of the nonshifted Rayleigh scattering is one-half the width of the scattering with frequency shift, as follows from the relevant theory [40, 39]. H Walther et al. [47] presented a number of experimental curves for the dependence of the resonance fluorescence spectrum on the power in the case when the intense field is near the $^2S_{1/2}(F = 2) \to {}^2P_{3/2}(F' = 3)$ transition line in sodium. They also presented the dependence of the fluorescence spectrum on Δ. The resonant field intensity reached 1.8 W cm^{-2} at a power of 35 mW and for a beam diameter of 3 mm. The curves for the resonance fluorescence in Ref. [47] indicate that the side lines $\omega \pm \Omega$ appear and disappear symmetrically when α changes.

There has been a long series of theoretical works on these issues [48–61]. In Refs [51, 52, 60b, c], the correlation between the first-order processes in terms of the 'weak field' was considered. In Refs [56–59], the existence of bunching and antibunching of photons, and also of sub-Poissonian statistics was revealed for resonance fluorescence.

Absorption of radiation by the system 'atom plus field' is usually measured by a so-called 'pumping and probe field' method. However, A Bonch-Bruevich et al. [62] have already obtained the first results on the absorption of probe field radiation by the system 'atom plus field' when the probe field intensity was of the order of the pumping intensity. N Manson et al. [63a] obtained new results on light absorption and dispersion in the case of two strong fields. These last results differ radically from the results of measurements in the case of a weak probe field. Finally, E Manykin et al. [63b] studied the interaction of a two-level atom with three coherent pulses and examined the parametric amplification process.

3. Behaviour of a two-level atom in the pulse light field

3.1 Relation between adiabatic and nonstationary functions

In the previous sections we have considered wave functions of a two-level atom in the field of an ideal monochromatic wave and also in the field of a 'monochromatic' wave with an extremely sharp leading edge. A monochromatic wave provides a mathematical idealization of an actual field. Actual fields are, as a rule, nonmonochromatic to some extent. In nonlinear optics the degree of nonmonochromaticity varies over the very wide range from $\delta\omega/\omega \approx 10^{-2}$ (femtosecond pulses) to $\delta\omega/\omega \approx 10^{-12} - 10^{-14}$ (highly-stabilized gas lasers). Nonmonochromaticity of a light pulse occurs when the electromagnetic field is 'switched on' one way or another for a time of order $1/\Gamma$ or the light pulse in use lasts for a time τ, because it is related to Γ and τ as follows

$$\delta\omega \approx \Gamma\,, \qquad \delta\omega \approx \frac{1}{\tau}\,. \tag{3.1}$$

To this end it is useful to consider the behaviour of a two-level system in the field of a nonmonochromatic wave

$$\mathbf{E} = \vec{\mathcal{E}}(t)\exp\{\mathrm{i}(\mathbf{k}_0 \cdot \mathbf{r} - \omega_0 t)\} + \text{c.c.} \tag{3.2}$$

Clearly, the nonmonochromatic nature of the wave brings about mixing of the two states Φ_1 and Φ_2, which expressions (2.25)–(2.26′) define. The degree of such mixing will depend on how 'smooth' the pulse (3.2) is. In the limiting case of an extremely fast switching-on (a rectangular pulse) the wave functions will be specified by the nonstationary wave functions (2.15) and (2.17).

Now we shall determine the degree of mixing for the wave functions Φ_1 and Φ_2, which enter the nonstationary functions Φ_1' and Φ_2'. For this purpose the nonstationary functions, which formulae (2.15) and (2.17) define, should be expanded into a series in terms of the adiabatic functions Φ_1 and Φ_2. One can easily verify that these series are defined by the expressions

$$\Phi_1' = C_1\Phi_1 + C_2^*\Phi_2\,, \qquad \Phi_2' = -C_2\Phi_1 + C_1\Phi_2\,, \tag{3.3}$$

where $C_{1,2}$ are given by formulae (2.23) and (2.24). It follows from relations (3.3), (2.23), and (2.24) that for $\alpha \ll 1$ the nonstationary and quasi-energy functions differ only slightly from each other. For $\alpha \gg 1$, the use of nonstationary functions will lead to strong mixing of the quasi-energy functions Φ_1 and Φ_2.

3.2 Exact solutions for particular pulse shapes

Exact methods of solution to the problems on the behaviour of an atom in the field of a nonmonochromatic wave and the related issues of a correct description of how the interaction is switched on and off are of great importance in physics since they can help to understand how radiation interacts with matter. Unfortunately, this program was a success for rather limited class of concrete light pulses. The pulse we consider in this section closely matches, in our opinion, a real pulse and in this case the problem of the behaviour of an atom in the field of a wave is amenable to a strict analysis. By investigating these problems we shall find conditions in which adiabatic and nonstationary functions are applicable.

Equations (2.6) specify coefficients $a_{1,2}$ for the wave functions of an atom in the field of a wave and they retain their form in the case when the amplitude of the wave (1.3) is time-dependent and has the form of (3.2). Using the same method as that to derive Eqn (2.6) we obtain

$$\mathrm{i}\dot{a}_1 = a_2 V^*(t)\exp(-\mathrm{i}\Delta t)\,,$$
$$\mathrm{i}\dot{a}_2 = a_1 V(t)\exp(\mathrm{i}\Delta t)\,, \tag{3.4}$$

where

$$V(t) = -\frac{\mathbf{d}\cdot\vec{\mathcal{E}}(t)}{\hbar}\,. \tag{3.4$'$}$$

We shall illustrate the influence of the way in which the interaction is switched on using an example for which an exact solution exists. This example was first proposed in Ref. [64] (see also Ref. [65]). Let the amplitude $\vec{\mathcal{E}}(t)$ vary slowly with time compared with the exponent:

$$\vec{\mathcal{E}}(t) = \frac{\vec{\mathcal{E}}_0}{\exp(-\Gamma t)+1}\,. \tag{3.5}$$

If we introduce

$$b = a_2\exp(-\mathrm{i}\Delta t)\,, \tag{3.6}$$

instead of a_2, then the equations for the coefficients $a_1(t)$ and $b(t)$ of the desired function Φ take the form

$$\mathrm{i}\dot{a}_1 = \frac{V_0}{\exp(-\Gamma t)+1}\,b\,, \qquad \mathrm{i}\dot{b} = \Delta a_1 + \frac{V_0}{\exp(-\Gamma t)+1}\,a_1\,, \tag{3.7}$$

where

$$V_0 = -\frac{\mathbf{d}\cdot\vec{\mathcal{E}}_0}{\hbar}\,. \tag{3.8}$$

For $\Gamma\to 0$ we have the limiting case of adiabatic field activation and for $\Gamma\to\infty$ the field is suddenly switched on when $t=0$. The calculation shows that in fact the width Γ should be compared to the resonance detuning so that the inequalities $\Gamma \ll |\Delta|$ and $\Gamma \gg \Delta$ correspond to the cited limiting cases. To solve Eqn (3.7), we introduce a new quantity

$$\chi = -\exp(\Gamma t)\,. \tag{3.9}$$

Eliminating the amplitude b from the system of equations we obtain an equation for a_1 which can be solved exactly. Under the initial conditions

$$a_1 = 1\,, \qquad a_2 = 0 \qquad \text{for} \qquad t\to-\infty \tag{3.10}$$

the solution takes the form

$$a_1 = (1-\chi)^{-\mathrm{i}V_0/\Gamma}F(\alpha,\beta,\gamma,\chi)\,, \tag{3.11}$$

where $F(\alpha,\beta,\gamma,\chi)$ is a hypergeometric function. Arguments of the hypergeometric function are expressed via constants Γ, Δ, and V_0 as follows:

$$\alpha = \frac{\mathrm{i}}{2\Gamma}\left(\Delta + 2V_0 - \sqrt{\Delta^2 + 4|V_0|^2}\right),$$
$$\beta = \frac{\mathrm{i}}{2\Gamma}\left(\Delta + 2V_0 + \sqrt{\Delta^2 + 4|V_0|^2}\right), \tag{3.12}$$
$$\gamma = \frac{\mathrm{i}\Delta}{\Gamma}\,. \tag{3.13}$$

The formula

$$F(\alpha,\beta,\gamma,\chi) = \frac{\Gamma(\gamma)\Gamma(\beta-\alpha)}{\Gamma(\beta)\Gamma(\gamma-\beta)}(-\chi)^{-\alpha}F(\alpha,1-\gamma+\alpha,1-\beta+\alpha,\chi^{-1}) + \frac{\Gamma(\gamma)\Gamma(\alpha-\beta)}{\Gamma(\alpha)\Gamma(\gamma-\beta)}(-\chi)^{-\beta}F(\beta,1-\gamma+\beta,1-\alpha+\beta,\chi^{-1})\,, \tag{3.14}$$

where $\Gamma(x)$ is a gamma function, is very useful for the analysis of limiting transitions.

At first we shall consider the case of adiabatic field activation. Here it is quite natural to consider a steady-state behaviour for $t \gg \Gamma^{-1}$. Then

$$a_1 = \frac{\Gamma(\gamma)\Gamma(\beta-\alpha)}{\Gamma(\beta)\Gamma(\gamma-\alpha)}\exp(-\mathrm{i}\lambda_1 t) + \frac{\Gamma(\gamma)\Gamma(\alpha-\beta)}{\Gamma(\alpha)\Gamma(\gamma-\beta)}\exp(-\mathrm{i}\lambda_2 t)\,, \tag{3.15}$$

where

$$\lambda_{1,2} = \frac{1}{2}\left(\Delta \mp \sqrt{\Delta^2 + 4|V_0|^2}\right) \tag{3.16}$$

is given by formula (2.16) with $|V_0|$ in place of $|V|$.

For the sake of simplicity we shall assume that $\Delta > 0$. For $\Gamma \ll \Delta$, all indices α, β, and γ become large, except, maybe, α in the case when $|V_0| \ll |\Delta|$. But even in the last case the arguments of all gamma functions in front of the first exponent in (3.15) are large and the familiar formula

$$\left|\Gamma(\mathrm{i}z)\right|^2 = \Gamma(\mathrm{i}z)\Gamma(-\mathrm{i}z) = \frac{\pi}{z\sin(\pi z)} \to \frac{2\pi}{|z|}\exp\bigl(-\pi|z|\bigr) \tag{3.17}$$

from the theory of gamma functions can be used to evaluate the absolute value of the pre-exponential factor. After simple manipulations we can see that the factor in front of the exponent $\exp(-\mathrm{i}\lambda_1 t)$ in (3.15) is precisely equal to the absolute value of the coefficient of Ψ_1 in the wave function Φ_1 (2.25) whereas the factor in front of $\exp(-\mathrm{i}\lambda_2 t)$ is exponentially small. Thus, if the atom was in the ground state until the field is switched on, then for $\Gamma \ll \Delta$ a quasi-energy state with the wave function Φ_1 is established.

In another limiting case, $\Gamma \gg \Delta$, the indices of the function $F(\alpha, \beta, \gamma, \chi)$ become small and again we come to the formulation (3.15). Now the arguments of the gamma functions are small and we should use the relation

$$\Gamma(z) = \frac{\Gamma(1+z)}{z} . \tag{3.17$'$}$$

As a result, the amplitude a_1 is precisely the same as the coefficient of Ψ_1' in the wave function (2.15). This last function is the solution of the Schrödinger equation when the interaction is suddenly switched on.

Equation (3.4) can be solved by several mathematical methods. In the case of real $V(t)$, upon elimination of a_2, the system is reduced to one second-order equation

$$\ddot{a}_1 + \left(\mathrm{i}\Delta - \frac{\dot{V}}{V}\right)\dot{a}_1 + V^2 a_1 = 0 . \tag{3.18}$$

The first solution of (3.18) for $\Delta \neq 0$ and $V \neq \text{const}$ has been yet found in Ref. [66] for a bell-shaped pulse $V \approx \operatorname{sech}(\Gamma t/2)$. A Bambini and P Berman [67] presented a solution in terms of hypergeometric functions for the two-parametric family of envelopes

$$V(z) = \frac{\sqrt{z(1-z)}}{\lambda z + \mu} ,$$

where the variable z is related to t by means of the formula

$$\exp(t) = \frac{z^{\mu}}{(1-z)^{\lambda+\mu}} \tag{3.19}$$

(here λ and μ are real parameters). This family of solutions includes the previous result for $\mu = 1/2$, $\lambda = 0$.

The second two-parametric class $V(t)$, for which Eqn (3.18) can also be solved in terms of hypergeometric functions, was found in Ref. [68]:

$$V(z) = \frac{1}{1+z^2}\frac{\mathrm{d}z}{\mathrm{d}t} , \qquad t = \lambda \arctan z + \mu \ln(1+z^2) \tag{3.20}$$

(λ and μ are arbitrary).

The above integrable system (3.4), when $E(t)$ has the form as in (3.5), corresponds to the dependence $V(t) = \lambda\left[1 + \tanh(\mu t)\right]$ for arbitrary λ and μ.

All the above integrable solutions are obtained when the transformation of dependent and independent variables is simultaneously applied to (3.18):

$$a(z) = \frac{U(z)}{\varphi(z)} , \qquad \frac{\mathrm{d}z}{\mathrm{d}t} = \rho(z) , \tag{3.21}$$

where φ and ρ are some functions [68]. This transformation is the most general procedure by which a second-order equation can be transformed into another second-order equation.

If we require that the resultant equation be coincident with some common equation with a well-studied solution (for example, with a hypergeometric solution) upon application of transformation (3.21) to Eqn (3.18):

$$U_{zz} + f(z)U_z + g(z)U = 0 \tag{3.22}$$

(the subscript z means differentiation with respect to z), then we arrive at a system of equations for the functions $\varphi(z)$, $\rho(z)$, and $U(z)$, which depend on $f(z)$ and $g(z)$. This system allows about twenty explicit solutions [68] resulting in twenty two- and three-parametric classes of envelopes $V(t)$, in which (3.18) can be integrated in terms of special functions, and one two-parametric class, in which (3.20) can be solved in quadratures. However, these mathematical methods have not been applied to solve any particular problems.

In Ref. [69a], R Unanyan used the method of quantum supersymmetry to find relations between the coefficients in the asymptotic expansion for wave functions as $x \to \pm\infty$, when $V(t)$ is arbitrary. In Ref. [69b], this method was employed to obtain the analytic solution for $V = V_0(\cosh t)^{-1} + W_0 \tanh t$, where V_0 and W_0 are arbitrary constants.

A series of works on the mathematical methods of resonant interaction was conducted in relation to problems of multiphoton ionization and the results were set forth in monograph [70].

There is a wide circle of problems on the behaviour of a two-level atom in the presence of two monochromatic fields with arbitrary amplitudes and near-resonance frequencies [71–77].

Though simple analytic solutions of this problem are lacking. The system of equations has an exact solution only when the two pulses are symmetric about the resonance, i.e. when $\Delta_2 = -\Delta_1$. In this case, for $V_1 = V_2 = V$, the solution can be written in the form (for $t = 0$, $\Phi_1 = \Psi_1$, $\Phi_2 = \Psi_2$):

$$\Phi_1 = \cos\left(\frac{V}{\Delta}\sin\frac{\Delta t}{2}\right)U_1 + \mathrm{i}\sin\left(\frac{V}{\Delta}\sin\frac{\Delta t}{2}\right)U_2 \exp\left[-\mathrm{i}(E_2 - E_1)t\right] , \tag{3.23}$$

$$\Phi_2 = -\mathrm{i}\sin\left(\frac{V}{\Delta}\sin\frac{\Delta t}{2}\right)U_1 + \cos\left(\frac{V}{\Delta}\sin\frac{\Delta t}{2}\right)U_2 \exp\left[-\mathrm{i}(E_2 - E_1)t\right] . \tag{3.24}$$

The solutions for a bichromatic field are closely related to the solutions for the behaviour of an electron and atoms in the field of a standing wave. This issue was studied in two reviews [78].

The case of two fields provides the simplest example of an interaction of atomic systems with a nonmonochromatic field. Note that the interaction will depend on the statistics of the radiation by which the atom is perturbed. These issues have been studied in a variety of papers (see, for example, Refs [79, 80]).

Another wide circle of problems deals with behaviour of a two-level atom in the field of a monochromatic wave without the use of a resonance approximation. A Melikyan [81] considered the well-known linear Hill equation with periodic coefficients. The frequency of the incident wave is arbitrary. In the case of the one-photon resonance, for $\alpha \gg 1$ and as $\Delta \to -0$, the quantities λ_1 and λ_2 in formulae (2.7) and (2.9) are given by the expression

$$\lambda_{1,2} = \pm\frac{2\omega}{\pi}\arcsin\left(\frac{\pi}{2}\sqrt{r_1}\right) , \tag{3.25}$$

where r_1 is calculated from the convergent series in terms of the variable $\alpha\Delta/\omega$:

$$r_1 = \left(\frac{\alpha\Delta}{2\omega}\right)^2 \left[1 - \left(\frac{\alpha\Delta}{4\omega}\right)^2 \left(\frac{\pi^2}{3} + 1\right) + \ldots\right]. \tag{3.26}$$

If only the first term in the series (3.26) is inserted to (3.25), then the latter expression coincides with expression (2.9) which we have used. In the case of a multiphoton resonance, i.e. when $\omega \approx E_{21}/n$, where $n = 3, 5, 7$, formula (3.25) retains its form. Only the quantity r_1 in (3.26) should be replaced with the quantity r_n equal to

$$r_n = \left(\frac{\alpha\Delta}{2\omega}\right)^4 \frac{n^2}{n^2 - 1} \left[1 - \left(\frac{\alpha\Delta}{\omega}\right)^2 \frac{3n^2 + 1}{2(n^2 - 1)} + \ldots\right]. \tag{3.27}$$

Further results can be obtained by differentiating the quasi-energy with respect to the parameters on which it depends [81, 82a]. V Krainov and Z Mulyukov [82b] considered the problem of harmonic generation in a two-level system without the use of the resonance approximation.

3.3 'Quasi-classical' approximation in temporal space

The equations of interaction (3.4) between a nonmonochromatic electromagnetic pulse and an atom can be examined in detail near resonance. To this end we shall use a widely-known quasi-classical approximation of quantum mechanics to study problems in the space of time rather than in x, y, z space. The field amplitude that enters the expression for the nonmonochromatic field (3.4) has the form

$$\vec{\mathcal{E}}(x, t) = \left|\vec{\mathcal{E}}(x, t)\right| \exp\left[\mathrm{i}\varphi(x, t)\right]. \tag{3.28}$$

A solution of the Schrödinger equation (1.1) will be sought as an expansion in terms of the complete set of functions of an unperturbed atom (2.2). Equations (3.4) for the a_1 and a_2 expansion coefficients of the desired wave function Φ for a two-level system can be rewritten in the form

$$\mathrm{i}\dot{a}_1 = W^* a_2, \qquad \mathrm{i}\dot{a}_2 = W a_1, \tag{3.29}$$

where

$$W = V\exp(\mathrm{i}\Delta t) = \left|\frac{\mathbf{d}\cdot\vec{\mathcal{E}}}{\hbar}\right| \exp(\mathrm{i}\Delta t + \mathrm{i}\varphi + \mathrm{i}\varphi_1 - \mathrm{i}\varphi_2 + \mathrm{i}\pi)$$
$$= \left|\frac{\mathbf{d}\cdot\vec{\mathcal{E}}}{\hbar}\right| \exp\left[\mathrm{i}\varphi_0(x, t)\right], \tag{3.30}$$

here φ_1 and φ_2 are random phases of wave functions of the atom in the lower and upper states, and in addition

$$\varphi_0(x, t) = \Delta t + \varphi + \varphi_1 - \varphi_2 + \pi. \tag{3.31}$$

Equation (3.29) is followed by the equation for a_1:

$$\ddot{a}_1 - \dot{a}_1 \frac{\mathrm{d}}{\mathrm{d}t} \ln W^* + |W|^2 a_1 = 0. \tag{3.32}$$

With W in place of W^*, Eqn (3.22) gives the equation for a_2. The orthonormality of functions $\Phi_{1,2}$ yields the condition

$$|a_1|^2 + |a_2|^2 = 1. \tag{3.33}$$

A solution of Eqn (3.32) will be sought in the form

$$a_1 = \exp\left(\frac{\mathrm{i}}{\hbar} S\right) = \exp\left[\frac{\mathrm{i}}{\hbar}(S_0 + \hbar S_1 + \hbar^2 S_2 + \ldots)\right]. \tag{3.34}$$

Substituting (3.34) into (3.32) we have

$$\dot{S}_0 = \pm\hbar|W|, \tag{3.35}$$

$$\dot{S}_1 = -\frac{\dot{\varphi}_0}{2}. \tag{3.36}$$

The general solution of the Schrödinger equation can be presented in the following form [83] (under the condition that $|\dot{\varphi}_0| \ll 2|W|$):

$$\Phi = C_1 \exp\left\{\mathrm{i}\int_{-\infty}^{t} \left[|W| + \frac{1}{2}\dot{\varphi}_0(t)\right] \mathrm{d}t\right\} \left[\Psi_1 - \exp(\mathrm{i}\varphi_0)\Psi_2\right]$$
$$+ C_2 \exp\left\{-\mathrm{i}\int_{-\infty}^{t} \left[|W| - \frac{1}{2}\dot{\varphi}_0(t)\right] \mathrm{d}t\right\} \left[\Psi_1 + \exp(\mathrm{i}\varphi_0)\Psi_2\right]. \tag{3.37}$$

We shall select two functions $\Phi_{1,2}$ from the general solution such that the initial conditions $\Phi_1 \to \Psi_1$ and $\Phi_2 \to \Psi_2$ as $t \to -\infty$ are satisfied. The result for $\varphi_0(-\infty) = 0$ can be represented as

$$\Phi_1 = \exp\left(-\mathrm{i}\,\frac{\varphi_0}{2}\right) \Psi_1 \cos\left(\int_{-\infty}^{t} |W|\,\mathrm{d}t\right)$$
$$- \mathrm{i}\exp\left(\mathrm{i}\,\frac{\varphi_0}{2}\right) \Psi_2 \sin\left(\int_{-\infty}^{t} |W|\,\mathrm{d}t\right),$$
$$\Phi_2 = -\mathrm{i}\exp\left(-\mathrm{i}\,\frac{\varphi_0}{2}\right) \Psi_1 \sin\left(\int_{-\infty}^{t} |W|\,\mathrm{d}t\right)$$
$$+ \exp\left(\mathrm{i}\,\frac{\varphi_0}{2}\right) \Psi_2 \cos\left(\int_{-\infty}^{t} |W|\,\mathrm{d}t\right). \tag{3.38}$$

One can easily verify that functions (3.38) are orthonormalized. Thus, the applicability condition $\hbar S_1 \ll S_0$ for the quasi-classical approach is reduced to the condition

$$\left|\int_{-\infty}^{t} \frac{\dot{\varphi}_0}{2}\,\mathrm{d}t\right| \ll \int_{-\infty}^{t} |W|\,\mathrm{d}t. \tag{3.39}$$

With the use of (3.31) it follows from the latter inequality that if the pulse lasts for $-t_{\mathrm{eff}} < t < t_{\mathrm{eff}}$, then (3.39) takes the form

$$\left|\frac{1}{2}\int_{-t_{\mathrm{eff}}}^{t_{\mathrm{eff}}} \left(\Delta + \frac{\partial\varphi}{\partial t}\right) \mathrm{d}t\right| \ll \int_{-t_{\mathrm{eff}}}^{t_{\mathrm{eff}}} |W|\,\mathrm{d}t. \tag{3.40}$$

Consequently, the quantity

$$\int_{-t_{\mathrm{eff}}}^{t_{\mathrm{eff}}} |W|\,\mathrm{d}t,$$

which is proportional to the effective Rabi frequency, will be much greater, when multiplied by the pulse duration $2t_{\mathrm{eff}} \sim \tau$, than the total phase change due to the field amplitude phase φ as well as due to the detuning Δ.

In addition to condition (3.40) it is necessary to require that the 'accumulation' effect due to S_1 over the time the pulse lasts is unessential:

$$S_1(t) - S_1(-t_{\mathrm{eff}}) \ll \pi. \tag{3.41}$$

Condition (3.41) limits the applicability of solutions (3.37) and generally it depends on the specific behaviour of the pulse amplitude phase with time. For example, in the case of $\varphi = \text{const}$ the condition (3.41) will imply that 'accumulation' effects can be neglected provided that $\tau \ll \pi/\Delta$. In addition, since $\Delta \ll \omega$, functions (3.38) can be used to consider processes, which last longer than several periods of the laser wave, i.e. for much greater lengths than atomic and even interatomic distances. In the case when the exact resonance occurs and the amplitude of the wave possesses a time-independent phase ($\Delta = 0$, $\varphi = \text{const}$), the wave functions (3.38) give exact solutions to Eqn (3.29) and can be used in any time intervals.

In the case when $W(t)$ is suddenly switched on at $t = t_0$ (in so doing the lower limit of integration should be replaced with t_0 and the following quantities can be taken as being constant for $t > t_0$: $|W| = \text{const}$ and $\dot{\varphi}_0 = \Delta + \dot{\varphi} = \text{const}$), functions (3.37) and (3.38) go over to expressions (2.15) and (2.17) provided that $|\Delta| < |V|$ and $\dot{\varphi} = 0$. As this takes place corrections of the order of S_1 should also be retained in the pre-exponential factors in expressions (3.37) and (3.38); however, when $|W|$ and $\dot{\varphi}_0$ depend on time, pre-exponential corrections must be omitted for the normalization condition to be satisfied.

It is useful to compare Eqn (3.32) with the Schrödinger equation for which the quasi-classical method of solution is thoroughly developed [1, 84, 85]. To this end we shall introduce the variable b_1:

$$b_1(t) = a_1(t)\exp\left[\int^t f(t)\,\mathrm{d}t\right] = a_1(t)W^*, \qquad f(t) = \frac{\mathrm{d}}{\mathrm{d}t}\ln W^* \tag{3.42}$$

instead of a_1. Then Eqn (3.32) takes the form

$$\dot{b} + b\left(-\frac{\dot{f}}{2} - \frac{f^2}{4} + |W|^2\right) = 0\,. \tag{3.43}$$

Functions (3.37) and (3.38), especially when the field phase depends linearly on time, can find application in various problems on the interaction between light pulses and simple atomic systems near resonance transitions.

It should be emphasized that functions (3.38) require further investigation at 'turning points' [1, 84, 85].

We shall now come to a consideration of the opposite case when the pulse nonmonochromaticity and resonance detuning are such that the characteristic quantities of the problem are slow-varying time-dependent functions.

3.4 Adiabatic interaction of light pulse with a two-level system

The method of adiabatic perturbations [86] was used in a variety of papers [5, 87, 88] to study wave functions of an atom in an arbitrary nonmonochromatic field. We shall assume that the amplitude $\vec{\mathcal{E}}(t)$ in expression (3.2) is a slowly varying (as compared with the exponent) function of time. Then the expansion of $\vec{\mathcal{E}}(t)$ in terms of Fourier integrals taken over frequencies

$$\vec{\mathcal{E}}(t) = \int_{-\infty}^{\infty} \vec{\mathcal{E}}(\omega)\exp(-\mathrm{i}\omega t)\,\mathrm{d}\omega \tag{3.44}$$

would not contain the Fourier components $\mathcal{E}(\omega')$ for frequencies comparable to ω_{eff} or higher. Consequently, one obtains

$$\vec{\mathcal{E}}(\omega') = 0 \quad \text{for} \quad \omega' > \omega_{\text{eff}}\,,$$
$$\vec{\mathcal{E}}(\omega') \neq 0 \quad \text{for} \quad \omega' < \omega_{\text{eff}}\,, \tag{3.45}$$

where $\omega_{\text{eff}} \approx 1/\tau_{\text{eff}} \ll \omega_0$, and τ_{eff} is the pulse duration. In particular, if there is a set of monochromatic oscillations with frequencies $\omega_0 + \omega_i$ and phases φ_i, then the expression for $\vec{\mathcal{E}}(\omega')$ has the form

$$\vec{\mathcal{E}}(\omega') = \sum_i \vec{\mathcal{E}}_i \exp(\mathrm{i}\varphi_i)\delta(\omega' - \omega_i)\,, \tag{3.46}$$

where all frequencies $\omega_i \ll \omega_{\text{eff}}$. The phases φ_i can be either random or related (with each other) numbers. In the first case we shall deal with independent monochromatic oscillations; in the second case they will be correlated. Thus, the function $\mathcal{E}(t)$ defines the characteristics of nonmonochromatic light. Given $\mathcal{E}(t)$, all the statistical properties of the light are defined, including the polarization properties described by the density matrix, i.e. the 'fast'-time-averaged quantity

$$R_{ij} = \overline{\mathcal{E}_i(t)\mathcal{E}_j^*(t)} \tag{3.47}$$

(the mean is taken over time of the order of the oscillation period $1/\omega_0$). The behaviour of an atom in the field of a quasi-monochromatic wave, when the wave vectors are almost parallel, can be considered similarly. To this end $\mathcal{E}(t)$ should be also a slowly varying function of ρ, where ρ are the coordinates in the plane perpendicular to the x axis.

We shall consider now the adiabatic interaction of a plane nonmonochromatic electromagnetic wave travelling along the x axis, with simple atoms. We shall select the two-level model of an atom as an example. The wave functions of the lower and upper states of an unperturbed atom are given by formulae (1.4). The field $\mathbf{E}$ is described by the expression

$$\mathbf{E}(x,t) = \vec{\mathcal{E}}(x,t)\exp(\mathrm{i}k_0 x - \mathrm{i}\omega_0 t) + \text{c.c.}\,, \tag{3.48}$$

where

$$k_0 = \frac{\omega_0}{c}\,, \qquad \vec{\mathcal{E}}(x,t) = \mathbf{e}\mathcal{E}(x,t)\,. \tag{3.48$'$}$$

The slowly varying amplitude $\vec{\mathcal{E}}(x,t)$ possesses polarization components in the plane perpendicular to the x axis. If the envelope of the pulse $\vec{\mathcal{E}}(x,t)$ depends also on the coordinate $\rho(y,z)$, then in (3.48) $\vec{\mathcal{E}}(x,t)$ should be replaced with $\vec{\mathcal{E}}(\rho,x,t)$. If the pulse has a transverse structure, then on the integral expansion in terms of ρ some components with $\mathbf{k}_\rho$ will appear, for which the condition $k_\rho \ll k_0$ is to be satisfied. In this case we are concerned with a set of plane waves travelling at a small 'angle' to the x axis. These waves depend on the transverse structure of $\vec{\mathcal{E}}(\rho,x,t)$, and this result was used in Ref. [88] to explain the conical radiation.

The simplest way to effect a two-level system is to select levels with different parity since they interact differently with circularly polarized waves. Then the interaction potential in the Schrödinger equation can be replaced with the expression

$$\mathbf{d}\cdot\mathbf{E} = d_+E_+ + d_-E_-$$
$$= (d_+\mathcal{E}_+ + d_-\mathcal{E}_-)\exp(\mathrm{i}k_0 x - \mathrm{i}\omega_0 t) + \text{c.c.}\,, \tag{3.49}$$

where $d_\pm = d_y + \mathrm{i}d_z$. With expansion (3.44) in place of the amplitude $\vec{\mathcal{E}}(x,t)$, expression (3.48) can be rewritten in the form

$$\mathbf{E}(x,t) = \int_{-\infty}^{\infty} \vec{\mathcal{E}}_\omega \exp\left[\mathrm{i}k_0 x - \mathrm{i}(\omega_0 + \omega)t\right] \mathrm{d}\omega + \text{c.c.} \quad (3.50)$$

Since $\mathcal{E}(x,t)$ varies slowly compared to $\exp(\mathrm{i}\omega_0 t)$, the frequency range $-\omega_{\text{eff}} < \omega < \omega_{\text{eff}}$ makes a major contribution to the integral over ω. When $|\omega_{\text{eff}}| < \omega_0$, expression (3.50) can be considered, by analogy with quantum electrodynamics, as the expansion of the electromagnetic pulse field in terms of positive frequencies $(\omega_0 \pm \omega)$, at which photons are absorbed, and in terms of negative frequencies $(-\omega_0 \pm \omega)$, at which emission occurs.

The solutions of the equation can be presented as an expansion in terms of the complete orthonormal set of functions of the unperturbed atom. The equations for the desired coefficients $a_{1,2}$ have the form of (3.4).

When deriving Eqn (3.4) we have omitted the so-called nonresonance terms on the right-hand side. These terms include exponents of the form $\exp(E_{21} + \omega_0)$ and, therefore, the condition

$$|\Delta| \ll E_{21} + \omega_0 \quad (3.51)$$

should be satisfied.

We shall seek the solution in the form

$$a_1 = C(t) \exp\left(-\mathrm{i}\int^t \lambda \, \mathrm{d}t\right). \quad (3.52)$$

It follows from the first equation in (3.4) that

$$a_2 = \frac{\lambda}{V^*} \exp(\mathrm{i}\Delta t) a_1 . \quad (3.53)$$

In deriving (3.53) we have neglected terms with $\dot{C}$. The relevant error will be estimated below. Substituting (3.52) and (3.53) into (3.4) gives the stationary Schrödinger equation

$$\lambda a_1 = V^* \exp(-\mathrm{i}\Delta t) a_2 ,$$

$$(\lambda - \Delta) a_2 = V \exp(\mathrm{i}\Delta t) a_1 . \quad (3.54)$$

When obtaining (3.54) we have neglected the slow dependence of λ/V^* on t (see error estimates below).

The existence condition for nonzero solutions of Eqn (3.54) yields the roots

$$\bar{\lambda}_{1,2} = \frac{\Delta}{2}\left(1 \mp \sqrt{1+\alpha^2}\right). \quad (3.55)$$

The roots $\bar{\lambda}_{1,2}$ in the form (3.55) are very handy to expound the issues we consider in this section. This selection does not violate the one-to-one correspondence between the functions $\Phi_{1,2}$ and unperturbed wave functions of an atom. Actually, when the field is switched off, $\Phi_{1,2} \to \Psi_{1,2}$ independently of the sign of Δ. The fact that the roots $\bar{\lambda}_{1,2}$ are discontinuous as $\Delta \to \pm 0$, is unessential since all the media we consider are transparent and an exact resonance cannot occur. In linear optics the medium transparency means that two conditions must be satisfied:

$$|\Delta| \gg \gamma , \quad (3.56)$$

where γ is the upper level width, and

$$\delta\omega \ll |\Delta| , \quad (3.57)$$

where $\delta\omega$ is the bandwidth of radiation traversing the medium.

Condition (3.56) changes somewhat when nonlinear effects are taken into account. This is because Δ should be replaced by $\Delta' = E'_{21} - \omega$, where E'_{21} is the energy level difference with regard for the Stark shifts. In the adiabatic approximation $|\Delta'| > |\Delta|$, i.e. the system 'goes away' from the resonance. With regard for the nonlinear scattering effects, we considered in Section 2.4, condition (3.57) also changes due to spectral line broadening when the radiation passes through the medium. This circumstance will restrict the linear dimensions of the system.

In a nonlinear medium the true absorption should be considered in addition to the coherent processes like Rayleigh scattering, we considered in Section 2.4. This absorption is related to the transition of the atom to the excited state Φ_2 and to the emission of radiation of a three-photon frequency $\omega' = \omega - \Delta\sqrt{1+\alpha^2}$.

This process is incoherent in nature and brings about the real absorption of radiation when it passes through the nonlinear medium. As this takes place, absorption is accompanied by the $\Phi_1 \to \Phi_2$ transition and the three-photon emission at the frequency ω. For arbitrary α^2, the probability of the process is given by the formula

$$\mathrm{d}W = \frac{\mathrm{d}W_{\text{sp}}}{4}\left(1 - \frac{1}{\sqrt{1+\alpha^2}}\right)^2 , \quad (3.58)$$

where $\mathrm{d}W_{\text{sp}}$ is the probability of the spontaneous emission of a free atom.

Since condition (3.56) is satisfied, this absorption can be neglected even for large α^2 as

$$\gamma = \int \mathrm{d}W_{\text{sp}} .$$

In solving Eqn (3.4) we have neglected terms proportional to $\dot{C}$. They can be omitted provided that

$$|\dot{C}| \ll |\bar{\lambda} C| . \quad (3.59)$$

It can easily be rewritten in the form

$$\left|\frac{1}{|\mathcal{E}|}\frac{\partial|\mathcal{E}|}{\partial t} + \frac{\partial\varphi}{\partial t}\right| \ll \Delta(1+\alpha^2) . \quad (3.60)$$

The last condition follows from (3.57).

Consideration of the line broadening as a result of the phase self-modulation when radiation propagates through the medium, sets the restriction for the linear dimensions of the medium [88]:

$$l \ll l_{\text{coh}} = \frac{1}{qk_0}\,\frac{1+\alpha^2}{\alpha^2}\,\frac{|\Delta|}{\delta\omega_0} , \quad (3.61)$$

where

$$q = \frac{2\pi N|d|^2}{\hbar\Delta(1+\alpha^2)^{1/2}} , \quad (3.61')$$

$\delta\omega_0$ is the spectral width of the radiation on entry into the medium.

Using (3.52), (3.53), and (3.55) we obtain the complete orthonormal set of wave functions in the ground and excited states in the framework of adiabatic approximation [88]:

$$\Phi_1 = C\exp\left(-\mathrm{i}\int^t \bar{\lambda}_1\,\mathrm{d}t - \mathrm{i}E_1 t\right) \times \left\{U_1 + \frac{\bar{\lambda}_1}{V^*}\exp(\mathrm{i}k_0 x - \mathrm{i}\omega_0 t)U_2\right\}, \tag{3.62}$$

$$\Phi_2 = C\exp\left(\mathrm{i}\int^t \bar{\lambda}_1\,\mathrm{d}t - \mathrm{i}E_2 t\right) \times \left\{U_2 - \frac{\bar{\lambda}_1}{V^*}\exp(-\mathrm{i}k_0 x + \mathrm{i}\omega_0 t)U_1\right\}, \tag{3.63}$$

where the coefficient C is defined by the normalization condition

$$C = \frac{1}{\sqrt{2}}\left(1 + \frac{1}{\sqrt{1+\alpha^2}}\right)^{1/2}. \tag{3.64}$$

The adiabatic functions (3.62) and (3.63) were used to consider many issues on propagation of radiation through a medium of two-level atoms (see, for example, Ref. [88]). In conclusion of our presentation on the subject of the interaction of a resonant interaction with a two-level system we want to note that numerous problems are left beyond the scope of our review. They are set forth in monographs [89–98].

4. Interaction of intense radiation with an atom in the presence of two adjacent resonance transitions

4.1 Classification of different interaction schemes and the adiabatic wave functions of three-level atoms in the resonance approximation

In this section we shall consider two resonance transitions connected to each other through one common level. We shall call them adjacent resonances. The exciting radiation may consist of one or two monochromatic waves depending on whether adjacent resonances are closely located (quasi-equidistant levels) or widely separated in frequency. In the first case pumping is referred to as degenerate, in the second it is nondegenerate. The systems that Fig. 10 presents schematically are usually referred to as three-level systems. In the theory of paramagnetic amplification, the three-level system was considered by N Basov and A Prokhorov [99]. In the optical region in the stationary case (with regard for relaxation) it was considered by A Javan [100a] (the scheme is presented in Fig. 10b) in calculating the combination-type laser and some other authors as well [100b, c]. After that the three-level system was considered in a variety of original papers, reviews, and monographs, the results of many of which were set forth in Refs [5, 89–98, 101–161]. In recent years interest in the three-level system has been rekindled because new experimental opportunities have opened (see Section 5). However, the study of adjacent resonances is far from being complete. The physical processes which occur in the course of the interaction between the two resonant fields and a three-level system are much more diverse and complicated than the similar processes we studied in the previous systems using the example of a two-level system.

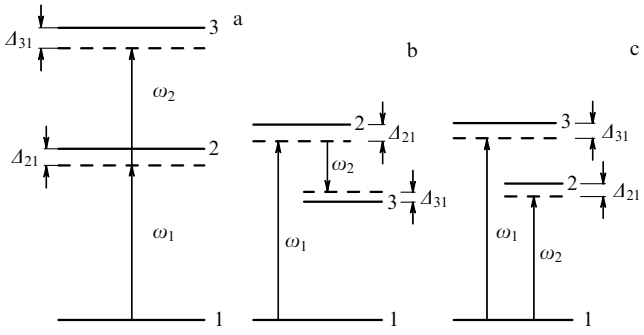

Figure 10. Three-level systems in the field of two resonance waves with adjacent transitions: (a) for $E_{31} = \omega_1 + \omega_2$, the scheme represents the two-photon absorption process; (b) at the exact resonance, $\Delta_{31} = 0$, this scheme describes the electron Raman scattering process, which is considered in detail in the next section; (c) simultaneous excitation of the ground level by two electromagnetic waves with resonance transitions to two different levels. In modern literature the first scheme is called a ladder-type scheme (the Ξ-type), the second scheme is called a Λ-type scheme, and the third scheme as the V-type scheme.

Our concern is with the behaviour of a three-level system when exact resonances do not necessarily occur and actual step transitions may be lacking. In this case with addition of a fourth level the schemes in Fig. 10 transform to four-level schemes. They can be considered using techniques we set forth below (see, for example, Ref. [117]). Here again, as in Section 2, the level population is understood to be the admixture of excited states to the ground state in the presence of resonant fields.

In optics, electric dipole transitions play a predominant role and we shall usually cite results of calculations just for these transitions. With a simple substitution we can go directly to the proper formulae for higher multipolarity. The following propositions and formulae are applied to molecules and atoms alike but in the last case the prohibitions that the law of conservation of parity introduces are to be considered.

Let two adjacent resonances occur in the system. We shall consider scheme in Fig. 10a as an example. With a simple renaming of variables the same expressions can be applied to schemes in Figs 10b and 10c:

$$|E_{21} - \omega_1| \ll E_{21}\,, \qquad |E_{32} - \omega_2| \ll E_{32}\,. \tag{4.1}$$

Notice that the occurrence of two adjacent resonances automatically brings about the third two-photon resonance:

$$|E_{31} - \omega_1 - \omega_2| \ll E_{31}\,. \tag{4.2}$$

The wave function of a three-level atom in the resonant field ('atom plus field') can be thought as an expansion in terms of three mutually orthogonal normalized atomic functions of an unperturbed atom:

$$\Phi = a_1\Psi_1 + a_2\Psi_2 + a_3\Psi_3\,. \tag{4.3}$$

The interaction operator for the field $\mathbf{E}(\mathbf{r}, t)$ has the form

$$\hat{V} = -\frac{\mathbf{d}\cdot\mathbf{E}(\mathbf{r},t)}{\hbar} = \hat{V}^{(-)}\exp(-\mathrm{i}\omega_1 t) + \hat{V}^{(+)}\exp(-\mathrm{i}\omega_1 t) + \hat{W}^{(-)}\exp(-\mathrm{i}\omega_2 t) + \hat{W}^{(+)}\exp(\mathrm{i}\omega_2 t)\,, \tag{4.4}$$

where

$$\mathbf{E}(\mathbf{r},t) = \vec{\mathcal{E}}_1 \exp(\mathrm{i}\mathbf{k}_1 \cdot \mathbf{r} - \mathrm{i}\omega_1 t) + \vec{\mathcal{E}}_2 \exp(\mathrm{i}\mathbf{k}_2 \cdot \mathbf{r} - \mathrm{i}\omega_2 t) + \text{c.c.} \tag{4.5}$$

The negatively-frequency $\hat{V}^{(-)}$, $\hat{W}^{(-)}$ and positively-frequency $\hat{V}^{(+)}$, $\hat{W}^{(+)}$ components of the interaction operator $\hat{V}$ describe, respectively, photon absorption and photon emission at the frequencies ω_1 and ω_2. Substituting (4.3) into the Schrödinger equation (1.1) and assuming that the field $\vec{\mathcal{E}}_1$ interacts only with the $1 \leftrightarrow 2$ transition and that the field $\vec{\mathcal{E}}_2$ interacts solely with the $2 \leftrightarrow 3$ transition, we arrive at three equations for the coefficients $a_i(t)$. In the resonance approximation, the equations take the form

$$\begin{aligned} \mathrm{i}\dot{a}_1 &= a_2 V_{12}^{(+)} \exp(-\mathrm{i}\Delta_{21} t)\,, \\ \mathrm{i}\dot{a}_2 &= a_1 V_{21}^{(-)} \exp(\mathrm{i}\Delta_{21} t) + a_3 W_{23}^{(+)} \exp(-\mathrm{i}\Delta_{32} t)\,, \\ \mathrm{i}\dot{a}_3 &= a_2 W_{32}^{(-)} \exp(\mathrm{i}\Delta_{32} t)\,, \end{aligned} \tag{4.6}$$

where

$$\Delta_{21} = E_{21} - \omega_1\,, \qquad \Delta_{32} = E_{32} - \omega_2 \tag{4.7}$$

are the resonance detunings, and $V_{ik}^{(\pm)}$, $W_{ik}^{(+)}$ stand for the matrix elements of the interaction operator. In the dipole approximation we have

$$V_{21}^{(-)} = -\frac{\mathbf{d}_{21} \cdot \vec{\mathcal{E}}_1}{\hbar}\,, \qquad W_{32}^{(-)} = -\frac{\mathbf{d}_{32} \cdot \vec{\mathcal{E}}_2}{\hbar}\,. \tag{4.8}$$

The operators $\hat{V}$, $\hat{W}$ are Hermitian and, hence, it follows

$$V_{nm}^{(+)} = \left[V_{mn}^{(-)}\right]^*, \qquad W_{nm}^{(+)} = \left[W_{mn}^{(-)}\right]^*. \tag{4.9}$$

To simplify the notation we denote

$$\begin{aligned} V_{21}^{(-)} &= V_{21}\,, \qquad V_{12}^{(+)} = V_{12}^*\,, \\ W_{32}^{(-)} &= V_{32}\,, \qquad W_{23}^{(+)} = V_{23}^*\,. \end{aligned} \tag{4.10}$$

Note that the matrix elements of the $\hat{V}$ operator and those of the $\hat{V}^{(-)}$ operator with negatively-frequency component are denoted by the same symbol V_{nm}. This fact should not cause misunderstanding since either matrix elements of the negatively-frequency component of the $\hat{V}$ operator or those of the positively-frequency component of the same operator are involved in all subsequent calculations. In addition, in this particular scheme from Fig. 10a the $1 \to 2 \to 3$ transitions are always related to the ω_1 and ω_2 photon absorption, i.e. to matrix elements V_{21} and W_{32} with the $(-)$ superscript. Using the notation of (4.10), the superscripts $(\pm)$ can be thereafter omitted and W_{32} can be renamed as V_{32}. A solution is sought in the form

$$\begin{aligned} a_1 &= b_1 \exp(-\mathrm{i}\lambda t)\,, \\ a_2 &= b_2 \exp\left[-\mathrm{i}(\lambda - \Delta_{21})t\right]\,, \\ a_3 &= b_3 \exp\left[-\mathrm{i}(\lambda - \Delta_{31})t\right] \end{aligned} \tag{4.11}$$

and for slowly varying amplitudes b_i we have the system of algebraic equations

$$\begin{aligned} -\lambda b_1 + V_{21}^* b_2 &= 0\,, \\ V_{21} b_1 - (\lambda - \Delta_{21}) b_2 + V_{32}^* b_3 &= 0\,, \\ V_{32} b_2 - (\lambda - \Delta_{31}) b_3 &= 0\,, \end{aligned} \tag{4.12}$$

where

$$\Delta_{31} = \Delta_{21} + \Delta_{32} = E_{31} - \omega_1 - \omega_2 \ll E_{31} \tag{4.13}$$

is the two-photon resonance detuning.

The determinant of Eqn (4.12) must be zero for nonzero solutions to exist. The equation meeting this condition is written as

$$\lambda(\lambda - \Delta_{21})(\lambda - \Delta_{31}) - (\lambda - \Delta_{31})|V_{21}|^2 - \lambda|V_{32}|^2 = 0\,. \tag{4.14}$$

Equation (4.12) has an Hermitian matrix and, hence, Eqn (4.14) defines three real roots λ_1, λ_2, and λ_3. Substituting the desired solutions $b_1^{(s)}$, $b_2^{(s)}$, $b_3^{(s)}$ for each λ_s into (4.3) we obtain three mutually orthogonal quasi-energy wave functions Φ_s, $s = 1, 2, 3$. These functions can readily be written out [5, 101, 102]:

$$\Phi_s = C_s \exp\left[-\mathrm{i}(E_1 + \lambda_s)t\right] \left\{ U_1 + \frac{\lambda_s}{V_{21}^*} U_2 \exp(-\mathrm{i}\omega_1 t) + \frac{V_{32}\lambda_s}{V_{21}^*(\lambda_s - \Delta_{31})} U_3 \exp\left[-\mathrm{i}(\omega_1 + \omega_2)t\right] \right\}. \tag{4.15}$$

The coefficient C_s is determined from the normalization condition up to an arbitrary phase:

$$C_s = \left\{ 1 + \frac{\lambda_s^2}{|V_{21}|^2} + \frac{\lambda_s^2}{(\lambda_s - \Delta_{31})^2} \frac{|V_{32}|^2}{|V_{21}|^2} \right\}^{-1/2}. \tag{4.16}$$

The choice of functions (4.15) can be substantiated by replacing the classical field $\mathbf{E}(\mathbf{r},t)$ with a quantized field and by considering the quantum-electrodynamic problem on the eigenvalues of energy and on eigenfunctions of a 'three-level atom plus quantized electromagnetic field' system, which is typically called a 'dressed' atom [118].

Apart from the time-dependent factors in braces, functions (4.15) are similar to stationary wave functions, where the quantity $E_s' = E_1 + \lambda_s$ plays the role of energy. However, because of the factors, which depend exponentially on ω_1 and ω_2, the quasi-energies become ambiguous and are determined up to an integer multiple of ω_1 and ω_2. This ambiguity can be used to establish the relationship between the quasi-energies and the energies of atomic states. We shall require the 'quasi-energies' to be the same as the relevant energies of atomic levels when the field $\mathbf{E}(\mathbf{r},t)$ is switched on and each function Φ_s goes into one of the atomic functions Ψ_i. For this purpose, the quasi-energies must be redefined by adding an integer multiple of ω_1 and ω_2.

Clearly the quasi-energies can be rewritten in the form

$$\begin{aligned} E_1' &= E_1 + \lambda_1\,, \qquad E_2' = E_1 + \lambda_2 + \omega_1\,, \\ E_3' &= E_1 + \lambda_3 + \omega_2 + \omega_1\,. \end{aligned} \tag{4.17}$$

Whence it follows that if $\lambda_1 \to 0$, $\lambda_2 \to \Delta_{21}$, and $\lambda_3 \to \Delta_{31}$, then E_s' approaches, respectively, E_1, E_2, and E_3. But if $\lambda_1 \to \Delta_{21}$, $\lambda_2 \to 0$, and $\lambda_3 \to \Delta_{31}$, then $E_1' \to E_2$, $E_2' \to E_1$,

and $E_3' \to E_3$. In three-level systems, as well as in multilevel systems it is convenient to operate directly with λ_s, which we shall also call quasi-energies. Since $\lambda_i \neq \lambda_j$ for $i \neq j$, as in the case of a two-level system, crossing of 'quasi-energies' is impossible in a three-level system.

Given

$$|V_{21}| \ll |\Delta_{21}|\,, \qquad |V_{32}| \ll |\Delta_{32}|\,, \tag{4.18}$$

the λ_s roots of the cubic equation (4.14) in the perturbation theory limit and in the resonance approximation have the form

$$\begin{aligned} \lambda_1 &= \delta_1\,, \\ \lambda_3 &= \Delta_{21} - \delta_1 + \delta_2\,, \\ \lambda_2 &= \Delta_{31} - \delta_2\,, \end{aligned} \tag{4.19}$$

where

$$\delta_1 = -\frac{|V_{21}|^2}{\Delta_{21}}\,, \qquad \delta_2 = -\frac{|V_{32}|^2}{\Delta_{32}}\,. \tag{4.19$'$}$$

In the framework of approximation described by (4.18) the wave functions (4.15) go over to wave functions for second-order perturbation theory, with regard only for resonant terms, and the 'quasi-energy' levels E_s' (4.17) and (4.19) are identical to the ordinary expressions for energy levels in second-order perturbation theory.

In the opposite limiting case of strong resonances all detunings can be set equal to zero. Then it follows from Eqn (4.14) (the numeration is taken according to the rule $\lambda_1 < \lambda_2 < \lambda_3$):

$$\lambda_1 = -\sqrt{|V_{21}|^2 + |V_{32}|^2}\,, \quad \lambda_2 = 0\,, \quad \lambda_3 = \sqrt{|V_{21}|^2 + |V_{32}|^2}\,. \tag{4.20}$$

The roots of Eqn (4.14) are continuous and single-valued functions of the frequencies ω_1 and ω_2 and of the resonant field strengths. Since the roots do not intersect anywhere, the condition $\lambda_1 < \lambda_2 < \lambda_3$ is satisfied over the whole range of their values. The numeration of roots (4.20) must be consistent with the numeration of roots (4.19). In this case roots (4.19) must be arranged in increasing order of magnitude. This arrangement depends on the signs of Δ_{ik} as well as on the ratios of quantities Δ_{ik} and each case should be examined separately.

Analysis of the roots of the cubic equation can easily be made using graphical methods. We shall consider the scheme in Fig. 10a with degenerate pumping, $\omega_1 = \omega_2$ and $\mathcal{E}_1 = \mathcal{E}_2$. In this case the λ_s roots of Eqn (4.14) are functions of two variables only: the frequency and amplitude of the wave field strength, i.e. they represent surfaces in three-dimensional space. At first we consider the dependence of quasi-energies on the electromagnetic field frequency at a certain field strength $\mathbf{E} \neq 0$. Figures 11a and 11b show these dependences in two possible cases: $E_{21} < E_{32}$ ($E_{31} < 2E_{32}$) and $E_{21} > E_{32}$ ($E_{31} > 2E_{32}$). It follows from the plot in Fig. 11a that the roots $\lambda_{1,2,3}$ are bounded by three asymptotes, one of which coincides with the horizontal axis, the two other straight lines intersecting the horizontal axis at the points $\omega = E_{31}$ and $\omega = E_{21}$. The roots λ_s approach the asymptotes at the points of two-photon and one-photon resonances when the field is switched off, $E \to 0$, i.e. when condition (4.18) is satisfied.

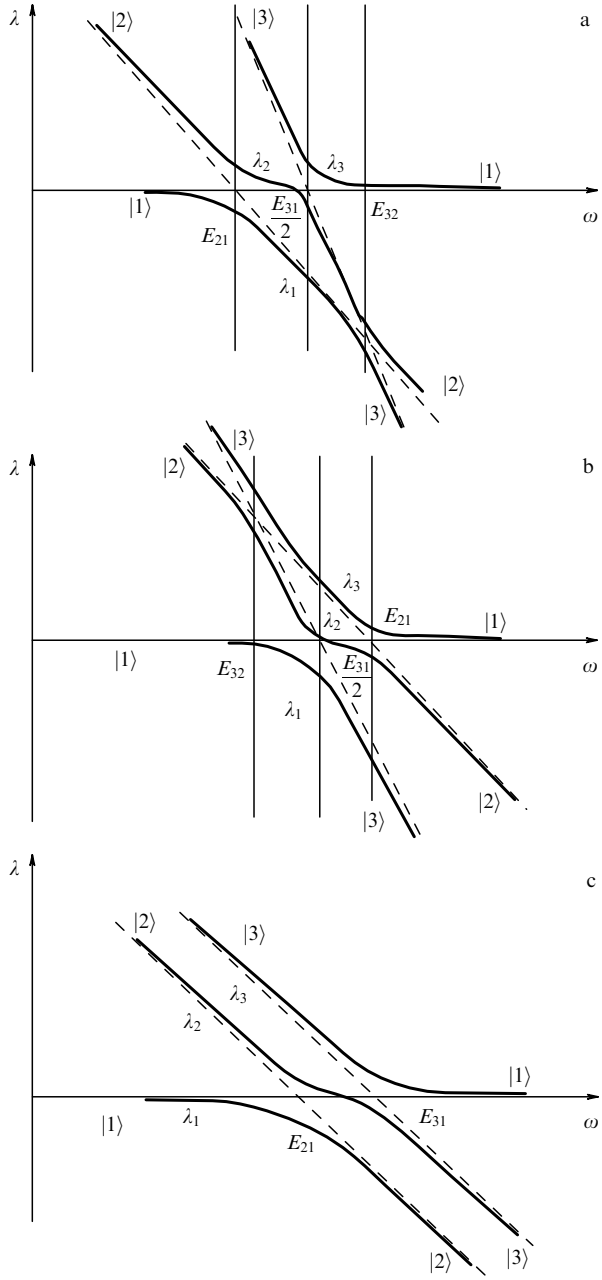

Figure 11. Roots of the cubic equation (4.14) as functions of the electromagnetic field frequency, when (a) $E_{21} < E_{32}$ and (b) $E_{21} > E_{32}$. The quantities are numbered according to the inequality $\lambda_1 < \lambda_2 < \lambda_3$. The values of the λ_i root are limited by the asymptotes, described by equation (4.20$'$). Two asymptotes are shown by dashed lines, and the third asymptote is the horizontal axis. The asymptotes intersect the λ axis at the points $\lambda = 0$, $\lambda = E_{21}$, and $\lambda = E_{31}/2$. The asymptotes (i.e. quasi-energies) λ_1, λ_2, and λ_3 intersect at the resonance points $\Delta_{21} = 0$ and $\Delta_{31} = 0$; λ_2 intersects λ_3 at the point Δ_{32}. The crosspoints are eliminated when the interaction V_{ik} is switched on. Figure 11c plots the roots of the cubic equation against ω for the system from Fig. 10c, when this system interacts with a single field.

The equations for the asymptotes are given by the following expressions

$$\lambda_i = 0\,, \qquad \lambda_j = \Delta_{21}\,, \qquad \lambda_k = \Delta_{31}\,. \tag{4.20$'$}$$

For definiteness we shall use λ_1 to denote the smallest root of Eqn (4.14), i.e. the lower branch; λ_3 to denote the largest

root, i.e. the upper branch; and, hence, λ_2 to denote the intermediate branch†.

Assume that the system finds itself in a specific quasi-energy state Φ_s, for example, when $\lambda_s = \lambda_1$. The plot in Fig. 11a shows that when the field is switched off the atomic system can be found in any state Ψ_i depending on the field frequency. If $\omega < E_{21}$, then $\Phi_1(\lambda_1) \to \Psi_1$; $\Phi_1(\lambda_1) \to \Psi_2$ for $E_{21} < \omega < E_{32}$; and $\Phi_1(\lambda_1) \to \Psi_3$ for $\omega > E_{32}$. Similarly, $\Phi_2(\lambda_2) \to \Psi_2$ for $\omega < E_{21}$; $\Phi_2(\lambda_2) \to \Psi_1$ for $E_{21} < \omega < E_{31}/2$; $\Phi_2(\lambda_2) \to \Psi_3$ for $E_{31}/2 < \omega < E_{32}$; and again $\Phi_2(\lambda_2) \to \Psi_2$ for $\omega > E_{32}$. For the state $\Phi_3(\lambda_3)$ we have $\Phi_3(\lambda_3) \to \Psi_3$ for $\omega < E_{31}/2$ and $\Phi_3(\lambda_3) \to \Psi_1$ for $\omega > E_{31}/2$. In a similar manner the correspondence can be established between the unperturbed states Ψ_i and quasi-energy states Φ_s for the system, for which the order of levels is such that $E_{32} < E_{21}$ $(E_{31}/2 < E_{21})$. In this case the plot in Fig. 11b should be used.

With adiabatic changes in parameters the system develops over the specific quasi-energy surface, on which it lands from the initial state when the field is switched on. However, the system does not necessary return to the initial state when the field is switched off. This can occur if one detuning reverses its sign under the frequency change. Thus, with an adiabatic passage of resonance and subsequent deactivation of the field, a transition to another state takes place. In particular, if the system is initially in the ground state, it can be fully inverted in this way to the excited states Ψ_2 and Ψ_3. A similar effect was discussed in Section 2.3 in the case of a two-level system.

A three-level system, which interacts with resonant fields, exhibits a number of new effects, which are absent in the two-level case. In particular, the above analysis of the plots in Fig. 11a, b yields an interesting conclusion: in its consecutive passage through all the resonances from left to right, i.e. with motion along one of the curves λ_1 in Fig. 11a, the system is excited to the state Ψ_3 when the field is switched off. Certainly we have assumed that the system was in the unperturbed ground state Ψ_1 prior to activation of the field.

Similar plots can also be drawn for the other three-level systems in Fig. 10, when they interact with one field. For example, such plots are presented in Fig. 11c for the system from Fig. 10c. The figure shows that in its passage through all the resonances from left to right (or from right to left) the system is excited upon deactivation of the field to the state, whose resonance was passed first. The required equation for the roots is obtained from (4.14) by re-indexing.

The above features can be used for selective excitation of atoms and molecules [5, 103a]. The selectiveness of the adiabatic inversion method exceeds that of other methods.

The plots in Fig. 11 correspond to the intersections of quasi-energy surfaces with a plane parallel to $\lambda\omega$. The sections of quasi-energy surfaces in the plane which is parallel to $\lambda\mathcal{E}$ and intersects the ω axis at the point $\omega = \omega_1$ can be obtained similarly (see Figs 12 and 13). The resultant curves will describe the dependence of quasi-energies on the field strength for a specific frequency of the field or, which is the same, at specific initial detunings from the resonances.

† This method of numbering may be occasionally inconvenient. In general the numbering of roots should be selected so that the index of the quasi-energy function is the same as that of the atomic function Ψ_i, when the fields are switched off. To this end $\bar{\lambda}_i$ have to be numbered so that $\bar{\lambda}_1 \to 0$, $\bar{\lambda}_2 \to \Delta_{21}$, and $\bar{\lambda}_3 \to \Delta_{31}$ as $E \to 0$. In this case, upon comparison with (4.19) we have $\lambda_1 = \bar{\lambda}_1$, $\lambda_3 = \bar{\lambda}_2$, and $\lambda_2 = \bar{\lambda}_3$ for $\Delta_{21} > 0$, and $\Delta_{32} > 0$. However, as frequency-dependent functions, the roots $\bar{\lambda}_i$ are discontinuous at the points $\Delta_{21} = 0$ and $\Delta_{32} = 0$.

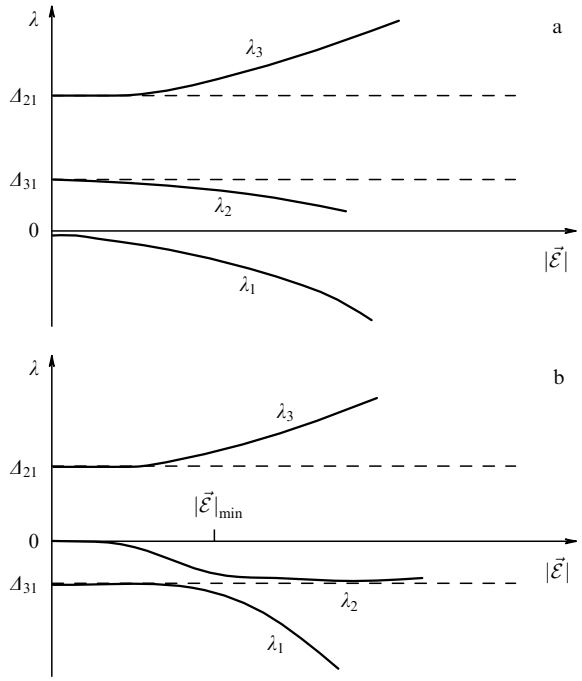

Figure 12. Quasi-energies λ_s as functions of the field intensity amplitude $|\vec{\mathcal{E}}|$ under conditions (4.21) and (4.22) in the case of a single field, when $|d_{21}| \gg |d_{32}|$ and $\Delta_{21} > 0$. The quasi-levels approach each other when $\Delta_{21}\Delta_{31} < 0$. The symbol $|\vec{\mathcal{E}}|_{\min}$ stands for the intensity value, for which the quasi-levels approach to the shortest distance [see formula (4.30)]. Note that $\lambda_1 \sim -\delta_2$ for small $|\vec{\mathcal{E}}|$ in the conditions analyzed in Fig. 12a, while $\lambda_1 \to \Delta_{31}$ in Fig. 12b.

In the case of a three-level system there are many plots due to a large number of parameters and their combinations, and consideration of them is of great interest for the purpose of interpreting laser experiments with pulse fields, the strength of which can vary. In the subsequent sections we shall dwell on some examples in greater detail.

4.2 Self-induced resonance and self-induced adiabatic inversion of a population

One of the interesting limiting cases when relatively simple expressions can be derived for quasi-energies is the two-photon resonance

$$|\Delta_{31}| \ll |\Delta_{21}|\,, \qquad \Delta_{21} \approx -\Delta_{32}\,. \tag{4.21}$$

If in addition to (4.21) we require

$$|V_{21}| \ll |\Delta_{21}|\,, \qquad |V_{32}| \ll |\Delta_{21}|\,, \tag{4.22}$$

which means that the perturbation theory can be applied to consideration of one-photon resonances, then the roots of the cubic equation (4.14) take the form

$$\lambda_{1,2} = \frac{1}{2}\left(\Delta_{31} + \delta_1 - \delta_2 \mp \sqrt{\delta^2 - 4\delta_1\delta_2}\right), \tag{4.23}$$

$$\lambda_3 = \Delta_{21} + \delta_2 - \delta_1\,, \tag{4.24}$$

where we have introduced the notation

$$\delta = \Delta_{31} - \delta_1 - \delta_2\,. \tag{4.24$'$}$$

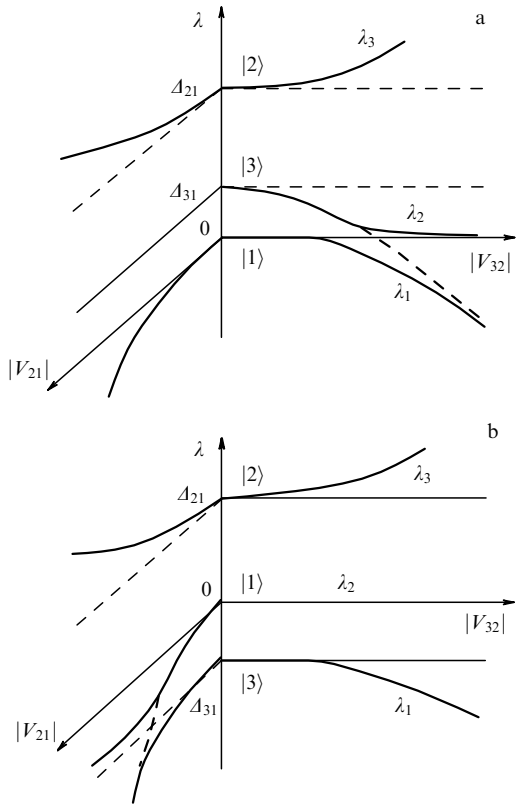

Figure 13. Quasi-energies as functions of $|V_{21}|$ and $|V_{32}|$ in the case of two fields $\mathcal{E}_1$ and $\mathcal{E}_2$. They are projected onto the planes $|V_{32}| = 0$ and $|V_{21}| = 0$ for $\Delta_{21} > 0$ and $|\Delta_{21}| \gg |\Delta_{31}|$. (a) Quasi-energies in the case of a two-photon resonance for $\Delta_{31} > 0$. The intersection of the λ_2 and λ_1 quasi-levels (the dashed line) at the point $|V_{32}|_{\rm cr}$ is replaced by their repulsion. The quasi-levels start to approach when $|V_{32}| \gg |V_{21}|$. The limiting values of the quasi-energies 0, Δ_{21}, and Δ_{31}, when both fields are switched on, match the unperturbed atomic states $0 \to |1\rangle$, $\Delta_{21} \to |2\rangle$, and $\Delta_{31} \to |3\rangle$. (b) Quasi-energies under the condition $\Delta_{31} < 0$. They approach each other when $|V_{21}| \gg |V_{32}|$ [cf. (4.31)].

The roots $\lambda_{1,2}$ are numbered in increasing order of magnitude for positive Δ_{21} and for positive Δ_{31}. When $\Delta_{31} < 0$, the roots $\lambda_{1,2}$ change places.

The dependences of quasi-energies on the field strength are plotted in Fig. 12 in the case of degenerate pumping both for positive and negative Δ_{31}, when $|d_{21}| > |d_{32}|$.

If the last term under the radical sign can be neglected, expressions (4.23) go over to the appropriate formulae (4.19) for the perturbation theory, when we take into account condition (4.21).

It is useful to write down the quasi-energies (4.23), when the additional condition

$$|\Delta_{31}| \ll |\delta_1 + \delta_2| \tag{4.25}$$

is satisfied.

Physically this condition means that the difference of Stark shifts for the first and third atomic levels exceeds the two-photon detuning. With the appropriate expansion of the square root we have, to within the terms proportional to Δ_{31}:

$$\lambda_1 = \delta_1 - \delta_2 + \frac{\delta_2}{\delta_2 - \delta_1}\,\Delta_{31}\,, \tag{4.26}$$

$$\lambda_2 = \frac{\delta_1}{\delta_1 - \delta_2}\,\Delta_{31}\,. \tag{4.27}$$

Comparison of expressions (4.26) and (4.27) with the plots in Fig. 12 shows that for $\Delta_{31} > 0$ (when $\Delta_{21} > 0$ and $|d_{21}| > |d_{32}|$) the quasi-energy λ_1 corresponds to the ground quasi-energy state, i.e. to the state, to which the system goes from the unperturbed state Ψ_1 when the field is switched on adiabatically, whereas λ_2 corresponds to the state Ψ_3, i.e. $\Phi_2 \to \Psi_3$. But if $\Delta_{31} < 0$ (for $|d_{21}| > |d_{32}|$ and $\Delta_{21} > 0$), then $\Phi_1 \to \Psi_3$ and $\Phi_2 \to \Psi_1$. Thus, depending on the sign of the two-photon detuning the wave function of the quasi-energy state Φ_1 goes over to either Ψ_1 or Ψ_3 when the field is switched off, $\mathbf{E}(\mathbf{r}, t) \to 0$.

As noted earlier, in a three-level system the quasi-energy levels cannot cross. However, unlike a two-level system, in which the levels always 'go away' from resonance, in a three-level system the levels can approach the resonance with an appropriate choice of the signs of detunings Δ_{21} and Δ_{31}, as well as the ratios of dipole moments $\mathbf{d}_{21}$, $\mathbf{d}_{32}$, and the field strengths (Fig. 12b). This phenomenon will be called self-induced resonance. In fact, using, for example, expression (4.23) we have

$$|\lambda_1 - \lambda_2| = \sqrt{\left[\Delta_{31} + \frac{|d_{21}|^2 - |d_{32}|^2}{\Delta_{21}}\,|\mathcal{E}|^2\right]^2 + \frac{4|d_{21}d_{32}|^2}{\Delta_{21}^2}\,|\mathcal{E}|^4} \tag{4.28}$$

in the case of one exciting field, where d_{21} and d_{32} are projections of the dipole moments onto the directions of the corresponding fields. For $|\mathcal{E}| = 0$, the spacing between quasi-energy levels is equal to the initial detuning $|\Delta_{31}|$. With an increase in intensity, the levels can approach as well as separate. Here we shall establish in which conditions self-induced resonance is possible for the $1 \to 3$ transition. It follows from the condition

$$\frac{\mathrm{d}}{\mathrm{d}|\mathcal{E}|^2}\,|\lambda_1 - \lambda_2| = 0 \tag{4.29}$$

that

$$|\lambda_1 - \lambda_2|_{\min} = \frac{2|d_{21}d_{32}|}{|d_{21}|^2 + |d_{32}|^2}\,|\Delta_{31}| \tag{4.30}$$

for the field intensity

$$|\mathcal{E}|^2_{\min} = \frac{|d_{32}|^2 - |d_{21}|^2}{\left[|d_{21}|^2 + |d_{32}|^2\right]^2}\,\Delta_{21}\Delta_{31}\,. \tag{4.31}$$

As is seen from (4.30), the minimum spacing between quasi-levels is proportional to Δ_{31} and depends on the projections of the matrix elements of transitions onto the direction of field polarization. The condition for which 'self-induced resonance' becomes possible, has the form

$$\left[|d_{32}|^2 - |d_{21}|^2\right]\Delta_{21}\Delta_{31} > 0\,. \tag{4.31$'$}$$

If $|d_{21}| > |d_{32}|$, then a 'self-induced resonance' occurs for $\Delta_{21}\Delta_{31} < 0$; but if $|d_{21}| < |d_{32}|$, then it is required that $\Delta_{21}\Delta_{31} > 0$. As is seen from (4.30), if the matrix elements d_{21} and d_{32} are of the same order, then the system levels cannot approach very close and $|\lambda_1 - \lambda_2|_{\min} \cong |\Delta_{31}|$. If the matrix elements differ radically from one another, then the quasi-levels can approach each other to a much smaller separation than the initial two-photon detuning $|\Delta_{31}|$.

In the case of nondegenerate pumping the results change somewhat. Based upon a minimization of roots (4.23), the two conditions

$$\frac{\partial}{\partial|\mathcal{E}_1|^2}|\lambda_1-\lambda_2|=0\,, \tag{4.32}$$

$$\frac{\partial}{\partial|\mathcal{E}_2|^2}|\lambda_1-\lambda_2|=0 \tag{4.33}$$

have to be satisfied. They yield

$$\varDelta_{31}-\delta_1+\delta_2=0 \quad (\varDelta_{31}\varDelta_{21}>0)\,, \tag{4.34}$$

$$\varDelta_{31}+\delta_1-\delta_2=0 \quad (\varDelta_{31}\varDelta_{21}<0)\,. \tag{4.35}$$

In this case the spacing between quasi-levels is

$$|\lambda_1-\lambda_2|=\sqrt{4\delta_2(\delta_2-\delta_1)} \tag{4.34$'$}$$

or

$$|\lambda_1-\lambda_2|=\sqrt{4\delta_1(\delta_1-\delta_2)}\,. \tag{4.35$'$}$$

The above expressions determine two different conditions linking $|\mathcal{E}_1|^2$ with $|\mathcal{E}_2|^2$ (with other variables fixed), for which expressions of the type of (4.28) reach their conditional minima in the case of two fields. Note that conditions (4.34) and (4.35) go over to each other with a change either in the sign of $\varDelta_{21}$ or in the sign of $\varDelta_{31}$. In the case of two fields, the expression $|\lambda_1-\lambda_2|$ does not have an absolute minimum since equalities (4.34) and (4.35) cannot be satisfied simultaneously. The quasi-levels get nearer to each other when one of these conditions is met.

It is easily seen from expression (4.30) that the minimum spacing between quasi-levels can be much smaller than the initial detuning $|\varDelta_{31}|$ and be comparable with the level widths or the exciting radiation bandwidth. In this region, i.e. for

$$|\lambda_1-\lambda_2|_{\min}\leqslant\varGamma_{1,3}\,,\qquad |\lambda_1-\lambda_2|_{\min}\leqslant\frac{1}{\tau}\,, \tag{4.36}$$

an actual crossing of quasi-levels takes place, i.e. the exact resonance occurs and the states Φ_1 and Φ_2 are mixed. This can result in the atom remaining in the excited state Ψ_3 when the pulse has passed through. This situation requires special consideration.

Figure 13 show the dependences of the quasi-energies λ_s on the external field intensities and on the electrical dipole moments of the $1\to2$ and $2\to3$ transitions, i.e. on $|V_{21}|$ and $|V_{32}|$. The projections onto the planes $|V_{21}|=0$ and $|V_{32}|=0$ are presented. Let us consider the curve in Fig. 13a. In the plane $|V_{21}|=0$ only the atomic transition $3\to2$ interacts with the resonant field, and the energy of the first level does not change. We shall change the interaction between the atom and field, i.e. the quantity $|V_{32}|$. Then for a critical value $|V_{32}|_{\rm cr}$ the curve λ_2 can intersect the straight line λ_1 or, which is the same, the $E_2'-E_1'$ difference of quasi-energies becomes equal to an integer multiple of ω. This means that the 'atom plus field' system becomes degenerate in terms of energy. In this case for very small $|V_{21}|$ the atomic levels 1 and 3 begin to interact intensely and the functions Ψ_3 and Ψ_1 intermix ultimately. Because of this interaction the crossing levels 'repulse' and, instead of an actual crossing, the levels approach each other (i.e. anti-crossing occurs). Physically this means that if the system was in the state Ψ_1 ahead of the critical point (when the resonant field is switched off), then, behind the point, it goes to the branch which the state Ψ_3 represents. Here the situation is the same as in the case of a rapid adiabatic inversion of levels which we considered in detail in Section 2.3, where at the exact resonance the energy levels of the 'atom plus field' system split apart and the 'atom plus field' system goes from the initial state Ψ_1 to the state Ψ_2, absorbing a resonant photon upon passage through the resonance point. The difference is that in the two-level case the 'passage' through resonance can be achieved by changing (scanning) the external field frequency or atomic transition frequency in the course of the interaction, whereas in the three-level case the 'passage' through resonance can be achieved by changing the laser field intensity. This phenomenon will be referred to as 'self-induced passage through resonance'. In addition, if in the three-level case the system goes continuously into the initial state Ψ_1 when the external field is switched off, as is seen from Fig. 13a, then in the two-level case it stays in the state Ψ_2. A similar situation can also take place in the plane $|V_{32}|=0$, when the level E_3' gets into an exact double resonance with the level E_1'. In both cases the wave function of the third atomic level mixes intensely with the ground-level wave function.

In the region of maximal proximity of quasi-levels the interaction ceases to be adiabatic and in general the system does not develop along a specific branch (surface). The adiabaticity criterion for interaction between a laser pulse and a three-level system under the self-induced resonance passage can be obtained by calculating the probability of transition from one branch (surface) of quasi-energy to another branch (surface).

We shall consider the wave function of the quasi-energy state Φ_1 near the two-photon resonance. The admixture of the second and third atomic functions to the wave function Φ_1 will result in the following 'populations' of atomic levels

$$\begin{aligned} n_1&=C_1^2\,,\\ n_2&=\left|a_2^{(1)}\right|^2=C_1^2\,\frac{\lambda_1^2}{|V_{21}|^2}\,,\\ n_3&=\left|a_3^{(1)}\right|^2=C_1^2\,\frac{\lambda_1^2}{|\lambda_1-\varDelta_{31}|^2}\left|\frac{V_{32}}{V_{21}}\right|^2. \end{aligned} \tag{4.37}$$

When the field is switched off, $\lambda_1\to0$ (for $\varDelta_{31}>0$) and, therefore, Φ_1 goes over to the ground state of the atom [see (4.23) and Fig. 12a]. If λ_1 remains finite for finite $|\mathcal{E}|$ as $\varDelta_{31}\to+0$, then the ratio of the population of the third atomic level to that of the first level is

$$\frac{n_3}{n_1}=\left|\frac{V_{32}}{V_{21}}\right|^2 \quad \text{when} \quad \varDelta_{31}\to+0\,. \tag{4.38}$$

As $\varDelta_{31}\to-0$ (for $\varDelta_{21}>0$ and $|d_{21}|>|d_{32}|$), according to the above analysis and to Fig. 12b, the wave function Φ_2 will correspond to the ground atomic state. Consequently, the quasi-energy of the ground state is λ_2 and, according to (4.27), it also vanishes as $\varDelta_{31}\to-0$. In the state Φ_1, the populations are given by the expression (4.32) with the quasi-energy λ_2 of the ground state in place of the λ_1. Using (4.27) we have

$$\frac{n_3}{n_1}=\left|\frac{V_{21}}{V_{32}}\right|^2 \quad \text{when} \quad \varDelta_{31}\to-0\,. \tag{4.39}$$

Thus, with any ratio of interaction parameters V_{21} and V_{32}, except when they are equal, the populations can be inverted between the first and third atomic levels, $n_3 > n_1$, by appropriate choices of the sign of the $\Delta_{21}\Delta_{31}$ product and the ratio of the dipole moments, provided that condition (4.31′) is satisfied.

Close results are also obtained for self-induced inversion of levels in the case of a nondegenerate pumping.

Conditions for level populations (4.37) retain their forms in the case of nondegenerate pumping. However, the analysis is much more complicated than in the case of degenerate pumping, because there are two free parameters $\mathbf{E}_1(\mathbf{r}, t)$ and $\mathbf{E}_2(\mathbf{r}, t)$. However, a condition can be derived such that $n_3 \geqslant n_1$, i.e. self-induced overpopulation takes place.

It follows from Eqn (4.37) that self-induced overpopulation, $n_3 \geqslant n_1$, occurs when

$$\lambda_1^2 \geqslant (\lambda_1 - \Delta_{31})^2 \left|\frac{V_{21}}{V_{32}}\right|^2 . \tag{4.40}$$

Condition (4.40) is necessary and sufficient for self-induced overpopulation to occur.

Figure 14 shows the populations of the levels for the system in the state Φ_1 in the case of degenerate pumping.

M Ter-Mikaelyan [5] was the first to indicate that the populations of a three-level system can be inverted with an adiabatic pulse. He presented expressions for the relative overpopulations n_3/n_1 in the case of a strong field as $\Delta_{31} \to \pm 0$. The case of a two-photon resonance inversion was also considered in Refs [101, 104, 105]. M Grishkovsky and M Loy [104] used the term 'self-induced adiabatic rapid passage' by analogy with the adiabatic inversion at the magnetic resonance. In subsequent papers N Kroll, K Watson, and A Lau [106–108] studied whether self-induced inversion is possible depending on the pulse shape and on the time delay between two pulses [103a].

In Ref. [121a], self-induced resonance was first used to investigate nonstationary phenomena. M Loy observed optical nutation for molecules NH_3, i.e. population oscillation and free-polarization damping in the $(0^-, 4, 4) \to (2^-, 4, 4)$ transition, the energy difference of which is close to the total energy of two laser photons for the P_{34} and P_{18} lines of a CO_2-generator. When measuring the absorption coefficient for weak radiation ω_1 passing through vapors, sharp oscillations of the absorption coefficient were observed, depending on the time it takes for the power pulse of the second laser to pass through. This phenomenon can be explained as follows. When the leading edge of a power pulse passes through the system, the two-photon resonance sets in and, as a result, the interaction loses its adiabatic nature and the oscillations of the level populations manifest themselves under weak field absorption — optical nutation goes continuously (smoothly) into free polarization damping. A similar signal also appears at the trailing edge of the pulse. This experiment is analogous to the experiment on the observation of population oscillations that we mentioned in Section 2. However, self-induced adiabatic inversion has not yet been observed.

The influence of self-induced resonance on the excitation and ionization of a three-level system was considered in Refs [109–112]. In Refs [113–115], the influence of self-induced resonance was discussed in relation to the possibility of selective excitation of atoms. In Ref. [116], self-induced resonance was used to calculate the magnetization of alkali-metal atoms and in Refs [117, 118] it was used to study a four-level atom and to generate the third harmonic. Self-induced resonance plays an important role in studying the dielectric constant of the three-level system [119]. Most of the aforementioned papers were conducted either for monochromatic pumping fields or for laser pulses with a given activation law.

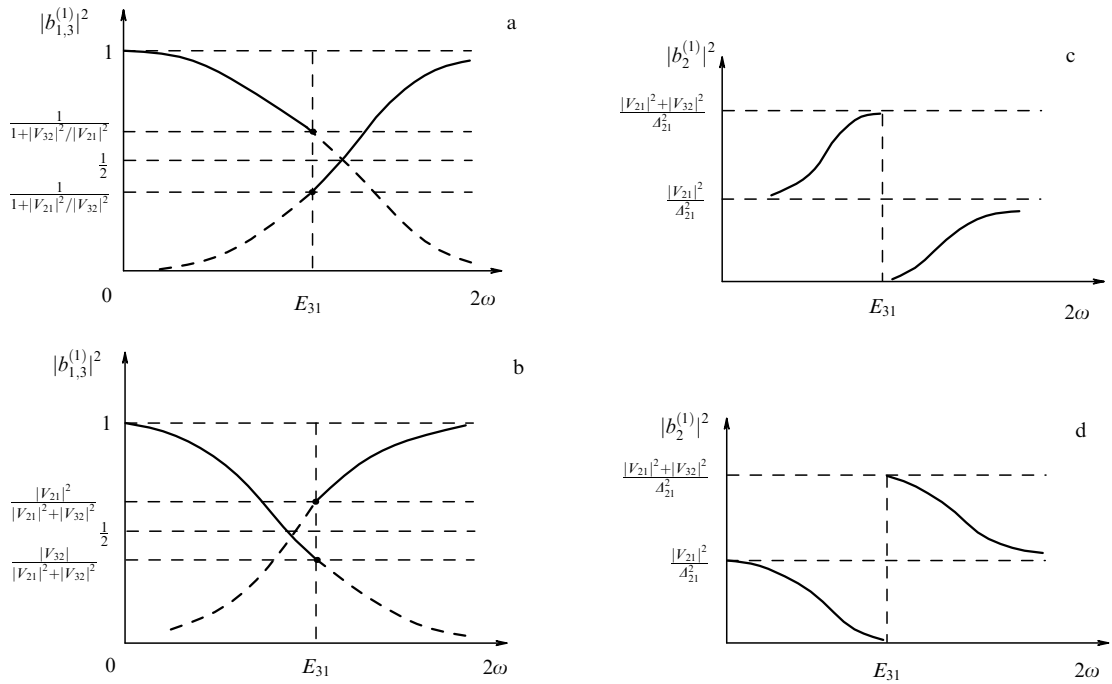

Figure 14. Populations of atomic levels for the system in the state Φ_1 as functions of 2ω (degenerate pumping) in the case when $|V_{12}| > |V_{23}|$. The curves represent the populations for the first (solid lines) and third (dashed lines) levels in the case when $\Delta_{21} > 0$ (a) and $\Delta_{21} < 0$ (b). The curves represent the populations for the second level in the case when $\Delta_{21} > 0$ (c) and $\Delta_{21} < 0$ (d).

M Sarkisyan and M Ter-Mikaelyan [103a] showed that the interaction between two time-separated laser pulses and a three-level ladder Ξ-type system brings about a selective excitation under the conditions of the self-induced resonance.

Currently a great interest was created to these problems (see, for example, [103b] and Section 5). In Ref. [103c], the selective population transfer was first observed in the adiabatic passage of two chirped laser pulses used to tune to the resonance. The work was conducted with Rb vapors by the scheme shown in Fig. 10a.

4.3 Nonstationary wave functions and two-photon absorption

The wave functions Φ_s in (4.15) form a complete orthonormal set of functions for a three-level atom in resonant monochromatic fields. Functions (4.15) are not the stationary solutions of the Schrödinger equation for an atom in a classical field since an atom in a varying field does not have a particular energy. However, in the case of a more general system with regard for a quantized electromagnetic field functions of the type (4.15) become, upon electromagnetic field quantization, stationary functions of a more complete quantum-mechanical 'atom plus quantized field' system ('dressed' atom) [18].

The wave functions (4.15) can be used when the interaction is switched on slowly. If the pulse duration is designated as τ (we shall use the quantity τ to describe the time it takes for the pulse to rise and decay), then it is necessary (but not always sufficient) for the interaction to be switched on adiabatically that the inequality

$$\tau^{-1} < \Delta_{ij} \tag{4.41}$$

holds.

In the case when the field is switched on abruptly, i.e. when conditions (4.41) are not satisfied, the wave function of the system can generally be represented as a superposition of the basis wave functions (4.15):

$$\Phi_l' = \sum R_s^{(l)} \Phi_s \,, \tag{4.42}$$

where the coefficients $R_s^{(l)}$ are determined from the initial conditions. Let the atom be in the ground state prior to switching on the field, i.e.

$$\Phi_l'(t=0) = \Psi_1 \,. \tag{4.42$'$}$$

Then the coefficients $R_s^{(1)}$ can be determined from (4.15) and (4.42$'$) by means of the formulae

$$\sum R_s^{(1)} = 1 \,, \qquad \sum R_s^{(1)} \lambda_s = 0 \,, \qquad \sum \frac{\lambda_s}{\lambda_s - \Delta_{31}} R_s^{(1)} = 0 \,. \tag{4.43}$$

The C_s coefficients of functions (4.15) enter the definition of R_s. Similarly, the coefficients $R_s^{(2,3)}$ can be obtained for the wave functions $\Phi_{2,3}'$, which go over to Ψ_2 and Ψ_3 at $t = 0$.

A solution of the type of (4.42) corresponds to the following statement of the problem: find the wave function of an atom in the field of travelling 'monochromatic' pulses with a steep leading edge perturbing the atom starting from $t = 0$. The fast switching-on also means that all the frequencies $\omega \leqslant \tau^{-1}$, where τ is the time interval in which the amplitudes of 'rectangular' pulses build up and decay to a finite value, are present in the radiation spectrum. The presence of frequencies of the order of $\omega_1 \pm \Delta_{21}$ and $\omega_2 \pm \Delta_{32}$ in the pulse spectrum causes actual transitions and oscillations of level population density to occur.

If condition (4.18) is satisfied, then the three-level system in question can be reduced to a generalized two-level system with one intermediate level provided that condition (4.2) is satisfied as well. Therefore, all the results set forth in the preceding sections can be carried over to a three-level system when the one-photon resonance-related effects can be considered within the scope of perturbation theory. Simple solutions can be obtained in other limiting cases as well.

We shall consider, for example, the exact two-photon resonance, when $\Delta_{31} = 0$. Using expansion (4.42) for the wave function of an atom, which resides in the ground state Ψ_1 till the interaction is switched on, we have (private communication, M Sarkisyan, 1984)

$$\begin{aligned}
\Phi_1' = {} & \left[1 + |q|^2\right]^{-1} \\
& \times \left\{ |q|^2 + \left[\cos\frac{\Omega_1 t}{2} + \frac{\mathrm{i}\Delta}{\Omega} \sin\frac{\Omega_1 t}{2}\right] \exp\left(-\mathrm{i}\,\frac{\Delta t}{2}\right) \right\} U_1 \\
& - \frac{\mathrm{i}\Omega_1}{2V_{21}^*} \left[1 - \frac{\Delta^2}{\Omega_1^2}\right] \sin\frac{\Omega_1 t}{2} \exp\left[-\mathrm{i}\left(\omega_1 + \frac{\Delta}{2}\right) t\right] U_2 \\
& + q \left\{ -1 + \left[\cos\frac{\Omega_1 t}{2} + \frac{\mathrm{i}\Delta}{\Omega_1} \sin\frac{\Omega_1 t}{2}\right] \exp\left(-\mathrm{i}\,\frac{\Delta t}{2}\right) \right\} \\
& \times \exp\left[-\mathrm{i}(\omega_1 + \omega_2) t\right] U_3 \,,
\end{aligned} \tag{4.44}$$

where

$$\begin{aligned}
& \Omega_1 = 2\sqrt{\frac{\Delta^2}{4} + |V_{21}|^2 + |V_{32}|^2} \,, \\
& \Delta = \Delta_{21} \,, \qquad q = \frac{V_{32}^*}{V_{21}^*} \,.
\end{aligned} \tag{4.45}$$

It follows from (4.44) for populations of atomic levels

$$\begin{aligned}
n_1(t) = {} & \frac{1}{\left(1 + |q|^2\right)^2} \left\{ |q|^4 + \cos^2\frac{\Omega_1 t}{2} + \frac{\Delta^2}{\Omega_1^2} \sin^2\frac{\Omega_1 t}{2} \right. \\
& \left. + 2|q|^2 \left[\cos\frac{\Omega_1 t}{2} \cos\frac{\Delta t}{2} + \frac{\Delta^2}{\Omega_1^2} \sin\frac{\Omega_1 t}{2} \sin\frac{\Delta t}{2}\right] \right\} , \\
n_2(t) = {} & \frac{4|V_{21}|^2}{\Omega_1^2} \sin^2\frac{\Omega_1 t}{2} \,, \\
n_3(t) = {} & \frac{|q|^2}{\left(1 + |q|^2\right)^2} \left\{ 1 + \cos^2\frac{\Omega_1 t}{2} + \frac{\Delta^2}{\Omega_1^2} \sin^2\frac{\Omega_1 t}{2} \right. \\
& \left. - 2\left[\cos\frac{\Omega_1 t}{2} \cos\frac{\Delta t}{2} + \frac{\Delta^2}{\Omega_1^2} \sin\frac{\Omega_1 t}{2} \sin\frac{\Delta t}{2}\right] \right\} .
\end{aligned} \tag{4.46}$$

At the exact one-photon resonance when $\Delta_{21} = 0$ as well, the expressions for populations of levels are greatly simplified:

$$\begin{aligned}
& n_1(t) = \left(1 + |q|^2\right)^{-2} \left(|q|^2 + \cos\frac{\Omega_0 t}{2}\right)^2 , \\
& n_2(t) = \left(1 + |q|^2\right)^{-1} \sin^2\frac{\Omega_0 t}{2} \,, \\
& n_3(t) = |q|^2 \left(1 + |q|^2\right)^{-2} \left(1 - \cos\frac{\Omega_0 t}{2}\right)^2 ,
\end{aligned} \tag{4.47}$$

where

$$\Omega_0 = 2\sqrt{|V_{21}|^2 + |V_{32}|^2}\,. \tag{4.48}$$

This case corresponds to the cascade excitation of levels.

In the opposite limiting case when the intermediate detuning is large, i.e. when the intensity parameter $\Omega_0/|\Delta| \ll 1$, we have for the population of the third level from (4.44):

$$n_3(t) = \frac{4|q|^2}{\left(1+|q|^2\right)^2}\sin\frac{\Omega_0^2 t}{4\Delta}\,. \tag{4.49}$$

Here we have a pure two-photon excitation, which was first considered in Refs [121b – d]. In this case the population $n_2(t)$ of the second level is low and the valent electron oscillates primarily between the first and third levels at a relatively low frequency Ω_0/Δ. It should be noted that the amplification phenomena which E Manykin and A Afanas'ev [121b] considered near two-photon resonances, have recently drawn much attention from researchers and they are now checked in experiments (see, for example, Ref. [121d]). We propose to set forth these issues in the review being in works for the *Progress in Optics*.

An examination of the expressions for populations (4.46) readily yields the conditions in which the three-level system is not excited during its interaction with a finite laser pulse, i.e. the atom remains in the ground state when the pulse has passed. These are so-called self-induced transparency pulses, or SIT-pulses (see, for example, Ref. [120]). The conditions in which atomic levels are maximally excited can also be easily obtained.

In the case of cascade excitation when both the $1 \to 2$ and $2 \to 3$ transitions are in exact resonance with the field, the problem is solved for an arbitrary shape of the envelope $\mathcal{E}(t)$ of both the pulses ($\mathcal{E}(t)$ is the projection of $\vec{\mathcal{E}}(t)$ onto $\mathbf{d}$) (private communication by M Sarkisyan, 1984).

It follows from Eqn (4.6) for $\Delta_{21} = \Delta_{31} = 0$ and for the initial condition $a_1 \to 1$, $a_2 \to 0$, $a_3 \to 0$ as $t_0 \to -\infty$ that the probability of the system's being in a particular level at an arbitrary instant of time t is

$$\begin{aligned} n_1(t) &= \left(1+|q|^2\right)^{-2}\left\{|q|^2 + \cos\theta(t)\right\}^2, \\ n_2(t) &= \left(1+|q|^2\right)^{-1}\sin^2\theta(t)\,, \\ n_3(t) &= \left(1+|q|^2\right)^{-2}\left[1-\cos\theta(t)\right]^2, \end{aligned} \tag{4.50}$$

where

$$\theta(t) = |d_{21}|\left(1+|q|^2\right)^{1/2}\int_{-\infty}^{t}\mathcal{E}(t')\,\mathrm{d}t'\,. \tag{4.51}$$

These expressions generalize formulae (4.47). As is seen from formulae (4.50), once the pulse has passed ($t \to \infty$), the system remains in the initial state provided that $\theta(\infty) = 2\pi n$. Therefore, in particular, the peak population of the second level is $\left(1+|q|^2\right)^{-1}$ and tends to unity as $|q| \to 0$ (a two-level system). The population of the third level peaks at a given $|q|$ when $\theta(\infty) = (2n+1)\pi$ and is equal to $4|q|^2\left(1+|q|^2\right)^{-2}$; moreover, the third level is fully populated when $|q| = 1$.

With a nonzero intermediate detuning Δ_{21}, the system of equations (4.6) does not have analytical solutions for arbitrary $\mathbf{E}(t)$. However, the basic behaviour can be established by choosing a particular function $\mathcal{E}(t)$, for which the system of equations (4.6) allows an exact solution. The benefits of this approach in the event of a two-level system are shown in Section 3.2.

Population oscillations in the case of a two-photon excitation were studied in Ref. [121].

Paper [122a] gave impetus to a large number of experimental works on the Doppler-free two-photon absorption. Even in the first experiments [123 – 125] with a sodium vapor, an extremely high resolution was achieved. These experiments can be treated on the basis of a generalized two-level system. Two-photon absorption was also studied in Refs [126 – 128]. As opposed to the aforementioned works, in the latter experiments exact intermediate resonance occurred and, thus, the processes proceeded with a much more probability and the three-level model with adjacent resonances had to be invoked for their interpretation. In the first work [126], the experiment was conducted with the use of two lasers, the frequencies of which were tuned to the $3S_{1/2} \to 3P_{1/2}$ ($2\pi n = 5895{,}9$ A) and $3P_{1/2} \to 4D_{3/2}$ ($2\pi n = 5682$ A) transitions. The authors used the approximate formulae of perturbation theory notwithstanding the fact that the intermediate level gets at the exact resonance. The comparison of the experimental data with the results of calculation from perturbation theory made it possible to determine the hyperfine structure of levels. In the second work [127], the $3S \to 4F$ transition through the intermediate resonance level $3P_{1/2}$ was studied. The electrical dipole transition $3P_{1/2} \to 4F$ is prohibited by the parity law. Therefore, the ratio of the probability of two-photon absorption for the transition $3S \to 4F$ to the relevant probability in the allowed electrical dipole transition $3S \to 4D$ was 10^{-7}. The authors measured the hyperfine structure of the 4F state.

In the next paper [128] these authors used a similar calculation technique to that we described above. The accord with the experimental data was remarkable. In the cited paper a limiting case was realized when the energy shift of levels greatly exceeded the hyperfine structure splitting of the intermediate energy level at the one-photon resonance in the field of a wave. Note that the presence of an intermediate resonance level, according to Ref. [128], increases the probability of two-photon absorption by a factor of 10 in comparison with the probability of two-photon absorption in the absence of an intermediate resonant level. The unique capabilities of Doppler-free spectroscopy were used to observe the two-photon transition $1 \to 2$ in hydrogen and deuterium [129, 130]. The authors were able to measure the isotopic shift of the transition and the Lamb shift of the ground 1S-state with an accuracy of 10^{-4}; they established that the former was 670.933 ± 0.056 GHz, and the latter was 8.20 ± 0.010 GHz for H and 8.25 ± 0.11 GHz for D. These results are in good accord with current theoretical values.

4.4 Permittivity of a three-level atomic gas

In this section we consider only the real part of the dielectric constant. As to the imaginary part this issue has not yet been resolved. If we neglect the influence of collisional widths and the motion of atoms, then the imaginary part of the dielectric constant related to radiative processes requires the probabilities of all noncoherent processes $\Phi_i \to \Phi_j$ ($i \neq j$) to be calculated since these processes affect the decay of the initial wave function (cf. Sections 2.4 and 4.5).

Notice that although the energy levels are degenerate in the direction of the total moment in an actual atom, the three-

level system can be realized by an appropriate choice of light polarization.

To calculate the real part of the permittivity of a rarefied gas involving three-level atoms it is necessary to determine the polarization of an atom in the quasi-energy state Φ_s, i.e. to determine the quantity

$$\mathbf{D}_{ss} = \left\langle \Phi_s^* | \mathbf{d} | \Phi_s \right\rangle . \tag{4.52}$$

At the two-photon resonance, the function Φ_s is given by formula (4.15). With regard for

$$\mathbf{D}_{ss}(\omega_1, \omega_2) = \chi_1^{(s)}(\omega_1)\mathbf{E}_1 + \chi_2^{(s)}(\omega_2)\mathbf{E}_2 , \tag{4.53}$$

the expressions $\chi_1^{(s)}\,(\omega = \omega_1)$ and $\chi_2^{(s)}\,(\omega = \omega_2)$ for the nonlinear polarization in the s state follow from (4.52).

The expressions for the nonlinear polarizations include quasi-energies which are the roots of a cubic equation. The use of the Cardano formulae renders these expressions too cumbersome, therefore in the general case it is simpler to use numerical calculations. However, in the above cases when the quasi-energies λ_s are given by simple analytical expressions, the polarizations $\chi_{1,2}^{(s)}$ can be expressed by means of relatively simple formulae.

Assume that the two-photon resonance condition (4.21) is satisfied for the system in question and that the one-photon resonances are weak. Then, using the results of Section 4.3 we have [119]

$$\chi_1^{(1,3)} = -\frac{|d_{12}|^2}{2\hbar\Delta_{21}}\left\{1 \pm \frac{\operatorname{sgn}(\Delta_{31}\Delta_{21})\left[\Delta_{21}\Delta_{31} + |V_{12}|^2 + |V_{32}|^2\right]}{\sqrt{(\Delta_{21}\delta)^2 + |2V_{21}V_{32}|^2}}\right\}, \tag{4.54}$$

$$\chi_2^{(1,3)} = -\frac{|d_{23}|^2}{2\hbar\Delta_{21}}\left\{1 \pm \frac{\operatorname{sgn}(\Delta_{31}\Delta_{21})\left[\Delta_{21}\Delta_{31} - |V_{12}|^2 - |V_{32}|^2\right]}{\sqrt{(\Delta_{21}\delta)^2 + |2V_{21}V_{32}|^2}}\right\}. \tag{4.55}$$

In the event of degenerate pumping the atomic polarizability $\chi(\omega)$ is equal to the sum $\chi_1 + \chi_2$ (in which it is required to set $E_1 = E_2 = E$, and $\omega_1 = \omega_2 = \omega$) and it is the same as the relevant expression in Ref. [131].

If all the atoms in a medium are in a particular quasi-energy state, then under degenerate pumping the dielectric constant for the medium is $\varepsilon = 1 + 4\pi N\chi^{(s)}$, where N is the number of atoms in the state s.

In the event of nondegenerate pumping the medium is characterized by two dielectric constants $\varepsilon_1(\omega = \omega_1) = 1 + 4\pi N\chi_1^{(s)}$ and $\varepsilon_2(\omega = \omega_2) = 1 + 4\pi N\chi_2^{(s)}$.

Note that in monochromatic fields when conditions (4.34) and (4.35) are satisfied, $\chi_1(\omega_1)$ or $\chi_2(\omega_2)$ (depending on the sign of the product $\Delta_{21}\Delta_{31}$) become linear with respect to the field and the relevant dielectric constants for the frequency ω_1 or ω_2 are independent of the field intensity.

Thus, by changing the field intensity in one of the transitions, the medium can be made transparent in the adjacent transition.

Figure 15 shows the dielectric constants ε_1 and ε_2 as functions of $\omega_1 + \omega_2$ in the vicinity of the point $\omega_1 + \omega_2 = E_{31}$. In this frequency range the one-photon detuning Δ_{21} changes only slightly and can be considered to be constant. For both quantities ε_1 and ε_2, an anomalous dispersion takes place over a certain frequency range. This range is limited by the points $|\Delta_{21}\Delta_{31}| = |V_{12}|^2 + |V_{32}|^2$ (the points of conditional minima (4.34) and (4.35) for the spacing between quasi-energies) on one side and by the two-photon resonance point on the other side.

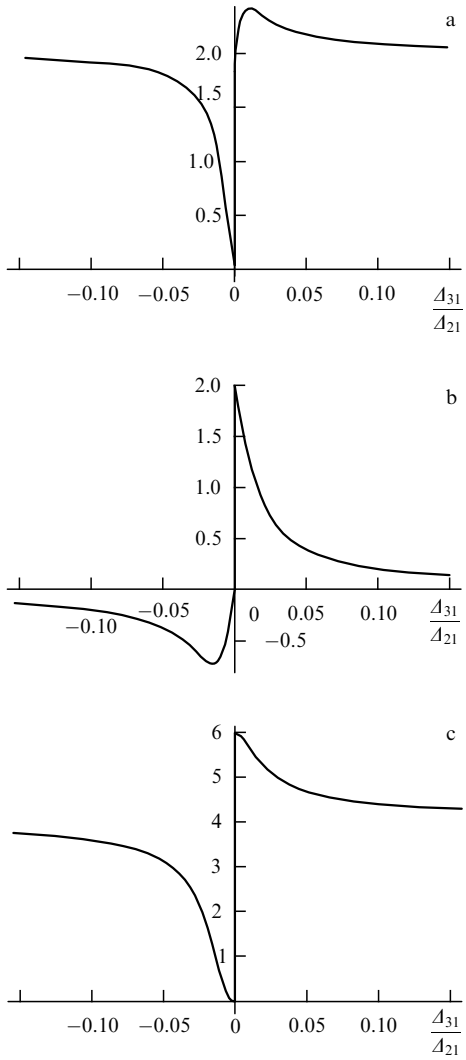

Figure 15. Dielectric constant of a three-level atomic gas as a function of the dimensionless parameter Δ_{31}/Δ_{21} for the following values of parameters: $\Delta_{21} > 0$, $|V_{12}|^2/\Delta_{21}^2 = |V_{32}|^2/\Delta_{21}^2 = 0.05$. On the vertical axes (a) $\frac{\varepsilon(\omega_1) - 1}{2\pi}\frac{\hbar\Delta_{21}}{|d_{32}|^2}$, (b) $\frac{\varepsilon(\omega_2) - 1}{2\pi}\frac{\hbar\Delta_{21}}{|d_{32}|^2}$, (c) $\frac{\varepsilon(\omega) - 1}{2\pi}\frac{\hbar\Delta_{21}}{|d_{12}|^2 + |d_{32}|^2}$ are plotted.

An explanation for the anomalous dispersion in this frequency range can be offered by analyzing the dynamics of atomic level populations [119]. In a weak field, the population of the first level decreases with an increase in E_1, i.e. the system rises to an excited state (becomes polarized) by virtually absorbing photons from the external field. However, with a further increase in the field intensity the reverse process is triggered once $|V_{12}|^2 = |\Delta_{21}\Delta_{31}| - |V_{32}|^2$, i.e. the system drops back to the lower level. Thus, the populations of atomic levels depend nonmonotonically on the field intensity and there is a region where an increase in the field intensity results in a decrease in the polarization of the medium. This nontrivial behaviour of atomic level populations is inherent solely to multilevel systems and has no analogy in the case of an ordinary two-level system. Another limiting case of strong resonances, when $|\Delta_{31}| \ll |V_{ik}|$ and Δ_{21} can take arbitrary values, was also considered in Ref. [119].

4.5 Interaction of resonance radiation with a three-level system with regard for radiative relaxation processes (degenerate pumping)

Up to this point we have considered the interaction between an atom and a laser field over times smaller than the relaxation time. It is interesting to find out how the level populations of a three-level atom change when radiative relaxation processes are taken into account [132].

The wave function of a three-level system in the field of a resonant wave with regard for the spontaneous transitions $\Phi_i \leftrightarrow \Phi_j$ can be expanded into a series in terms of the complete set of wave functions of a three-level atom:

$$\Psi(t) = \sum_{m=1}^{3} A_m(t)\Phi_m\,, \tag{4.56}$$

where $A_m(t)$ are the population amplitudes for the adiabatic 'atom plus field' functions. We shall consider the case when the resonance detunings $|\Delta_{21}|$, $|\Delta_{31}|$, and $|\Delta_{32}|$ are much greater than the radiative widths of the atomic levels γ_2 and γ_3 (the γ_1 width of the ground state is zero). In this event the direct absorption of photons from the external pumping field can be neglected and the changes in populations $A_m(t)$ for quasi-energy states will depend solely on the incoherent processes of spontaneous emission in the transitions $\Phi_i \leftrightarrow \Phi_j$.

For the three-level system under discussion the equations for level populations take the form

$$\begin{aligned}
\dot{N}_1 &= -(W_{21} + W_{31})N_1 + W_{12}N_2 + W_{13}N_3\,,\\
\dot{N}_2 &= W_{21}N_1 - (W_{12} + W_{32})N_2 + W_{23}N_3\,,\\
\dot{N}_3 &= W_{31}N_1 + W_{32}N_2 - (W_{13} + W_{32})N_3\,,
\end{aligned} \tag{4.57}$$

where W_{ij} are the time-averaged probabilities of the $\Phi_i \to \Phi_j$ spontaneous transitions. The $i = j$ transitions do not change populations and they have not been taken into consideration to write down the balance equations.

Now we shall find the physical meaning of the populations N_m when the field is switched off adiabatically fast. Since the typical time of N_m change is of the order of $\gamma_{2,3}^{-1}$, the wave functions Φ_m go over to their unperturbed functions $U_m \exp(-\mathrm{i}E_m t)$ when the field is fast ($\delta t \ll \gamma_{2,3}^{-1}$) but adiabatically switched off. In this case N_m give the level populations of a free atom.

In the resonance approximation ($i, j = 1, 2, 3$)

$$W_{ji} = \gamma_2 \left|b_2^{(i)} b_1^{*(j)}\right|^2 + \gamma_3 \left|b_3^{(i)} b_2^{*(j)}\right|^2\,, \tag{4.58}$$

where $b_i^{(m)}$ are the amplitudes of the atomic wave functions U_i in the quasi-energy state Φ_m [see (4.15)]. The transitions $\Phi_i \to \Phi_j$ are accompanied by photon emission at the frequencies $\omega' = \omega + \lambda_i - \lambda_j$. The scattering patterns are presented in Fig. 16 in the lowest order of the perturbation theory.

The mean populations of atomic levels in the field of a wave can be easily calculated using the expressions for populations and the values of $b_i^{(m)}$.

To this end functions (4.58) should be substituted into (4.57) and the squares of the absolute values of the coefficients of the functions U_i should be averaged over the time interval $t \gg (\lambda_m - \lambda_{m'})^{-1}$. For the mean values of the populations $\bar{n}_i$ of

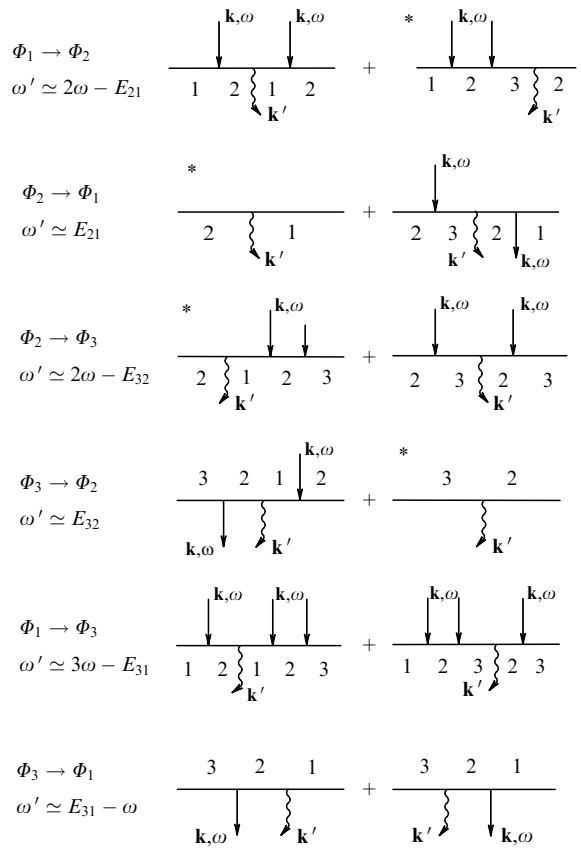

Figure 16. Scattering processes are shown diagramatically as they occur in transitions between different quasi-energy states. These processes cause the adiabatic functions to decay. Wavy lines represent photons of the scattered field.

atomic levels we have

$$\bar{n}_i = \sum_{m=1}^{3} N_m \left|b_i^{(m)}\right|^2\,. \tag{4.59}$$

The quantity $\bar{n}_i$ describes the probability that the free atom resides in a state with the wave function $U_i \exp(-\mathrm{i}E_i t)$, when the field is nonadiabatically fast (suddenly) switched off.

The expression for the total probability of emission per unit time for the 'three-level atom plus pumping field' system is

$$W = \gamma_2 \sum_{m=1}^{3} N_m \left|b_2^{(m)}\right|^2 + \gamma_3 \sum_{m=1}^{3} N_m \left|b_3^{(m)}\right|\,, \tag{4.60}$$

in which the first term represents the probability of emission for the $2 \to 1$ transition, and the second term represents the same quantity for the $3 \to 2$ transition. In other words, W is the total probability of a spontaneous emission by the pumping field incident upon a three-level atom.

Now we shall consider the stationary solutions of Eqn (4.57). Clearly they are generally cumbersome and non-informative since the dependences on the interaction parameters (detunings, field strength and matrix elements) are expressed through quasi-energies, which are roots of the cubic equation (4.14). Therefore, we shall consider two particular cases when the expressions for W_{ij} and, hence, for N_m can be greatly simplified.

(a) Consider firstly the case of a strong field (or the case of small detunings) when the conditions $|V_{21}|, |V_{32}| \gg |\Delta_{21}|, |\Delta_{31}|$ are satisfied. Using the appropriate values of $b_i^{(m)}$ for the probabilities of the $\Phi_i \to \Phi_j$ spontaneous transitions for $\Delta_{21}\Delta_{31} > 0$ we arrive at

$$W_{21} = W_{12} = \frac{\gamma_2^2 + \gamma_3^2}{4(\gamma_2 + \gamma_3)}\,,$$

$$W_{32} = W_{23} = W_{31} = W_{13} = \frac{\gamma_2\gamma_3}{2(\gamma_2 + \gamma_3)}\,. \tag{4.61}$$

Solving Eqn (4.41) together with the normalization condition, we obtain the expressions for the stationary populations in the quasi-energy states (the result is the same when $\Delta_{21}\Delta_{31} < 0$):

$$N_1 = N_2 = N_3 = \frac{1}{3}\,, \tag{4.62}$$

that is in the event of a strong interaction the atoms are equiprobably distributed over all the three quasi-energy states. Notice that the mean populations $\bar{n}_i$ of atomic levels are equal to each other. Then the total probability of scattering is

$$W = \frac{\gamma_2 + \gamma_3}{3}\,. \tag{4.63}$$

(b) Now consider the more interesting example of two-photon resonance, when $|\Delta_{31}|, |V_{21}|, |V_{32}| \ll |\Delta_{21}|$. In the resonance approximation the field mixes the first and third levels and the probabilities of transitions W_{ji} have the form (the corresponding patterns are marked by asterisks in Fig. 16)

$$W_{21} = \gamma_3 |b_3^{(1)}|^2\,, \qquad W_{32} = \gamma_2 |b_1^{(3)}|^2\,,$$

$$W_{12} = \gamma_2 |b_1^{(1)}|^2\,, \qquad W_{23} = \gamma_3 |b_3^{(3)}|^2\,,$$

$$W_{31} = W_{13} = 0\,, \tag{4.64}$$

where $|b_1^{(1)}|^2 = |b_3^{(3)}|^2 = n_1$ and $|b_3^{(1)}|^2 = |b_1^{(3)}|^2 = n_3$ are given by expressions (4.37).

In the steady-state mode it follows from (4.57) that

$$N_1 = \frac{n_1^2}{1 + (q+2)n_1 n_2}\,, \qquad N_2 = \frac{q n_1 n_2}{1 + (q+2)n_1 n_3}\,,$$

$$N_3 = \frac{n_3^2}{1 + (q+2)n_1 n_3}\,, \tag{4.65}$$

where

$$q = \frac{\gamma_3}{\gamma_2}\,. \tag{4.66}$$

It can readily be shown that the inversion can be realized by the appropriate choice of the interaction parameters for any transition between quasi-energy states and even for the $2 \to 1$ or $3 \to 2$ transitions when the field is switched off.

Using expressions (4.65) we shall calculate the mean values of the populations of atomic levels in the field of a wave. From (4.59) we have

$$\bar{n}_1 = \frac{1 - 3n_1 n_3}{1 + (q+2)n_1 n_3}\,, \qquad \bar{n}_2 = \frac{q n_1 n_3}{1 + (q+2)n_1 n_3}\,,$$

$$\bar{n}_3 = \frac{n_1 n_3}{1 + (q+2)n_1 n_3}\,. \tag{4.67}$$

For arbitrary relationships between the interaction parameters the populations N_m and $\bar{n}_i$ can be determined by numerical methods.

We shall calculate the total probability of scattering per unit time using the expressions for $\bar{n}_i$ from (4.67) and the values of $n_{1,3}$ from (4.37) [132]. The computation yields

$$W = \frac{2\gamma_3 d^2 |\mathcal{E}|^4}{\left(1 + g|\mathcal{E}|^2\right)^2 + (q+2)d^2|\mathcal{E}|^4}\,, \tag{4.68}$$

where

$$g = \frac{d_{21}^2 - d_{32}^2}{\hbar^2 \Delta_{21}\Delta_{31}}\,, \qquad d = \frac{d_{21}d_{32}}{h^2 \Delta_{21}\Delta_{31}}\,. \tag{4.69}$$

Note at once that the probability of scattering depends essentially on the sign of g. The probability of scattering peaks at a critical point for $g < 0$; it is determined from the equation $1 + g|\mathcal{E}_0|^2 = 0$ and equal to

$$W_{\max} = \frac{2\gamma_2\gamma_3}{2\gamma_2 + \gamma_3}\,, \tag{4.70}$$

i.e. $W_{\max}$ is limited by the least rate of decay. With an increase in the field intensity the probability of scattering decreases and approaches saturation when $|g||\mathcal{E}_0|^2 \gg 1$:

$$W_{\text{sat}} = \frac{2\gamma_2\gamma_3^2}{\gamma_2^2 + 2\gamma_3^2}\,. \tag{4.71}$$

Figure 17 shows the probability of scattering as a function of the pumping intensity. The probability of scattering for $g > 0$ is always lower than that for $g < 0$. The asymmetry with respect to the sign of g (or the sign of Δ_{31}) can be defined through the radiation selectivity [132]:

$$S = \frac{W(g<0)}{W(g>0)} = 1 + \frac{4|g||\mathcal{E}|^2}{\left(1 - |g||\mathcal{E}|^2\right)^2 + (q+2)d^2|\mathcal{E}|^4}\,. \tag{4.72}$$

The abrupt increase in the probability of scattering at the critical point is directly related to the nontrivial behaviour of populations of a three-level system in the presence of a resonance radiation, namely, to the self-induced resonance

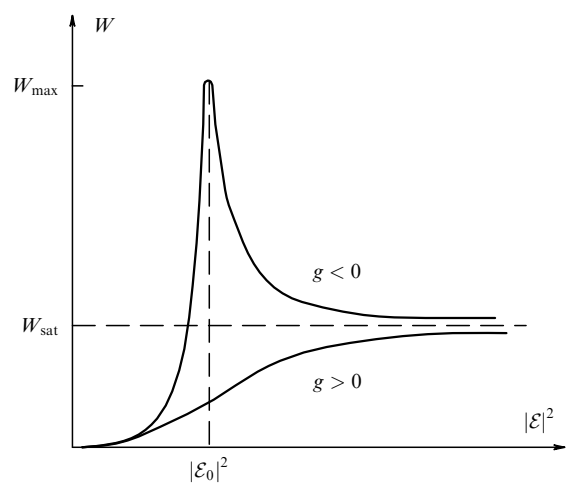

Figure 17. Total probability of scattering as a function of the pumping intensity, when $g < 0$ and $g > 0$.

phenomenon. At the critical point $|\mathcal{E}_0|^2$, the quasi-energies approach to a minimal spacing and the populations n_1 and n_3 level off with one another for $g < 0$.

The phenomenon considered can be used for selective action on atoms. The asymmetry with respect to the sign of g or, which is the same, to the sign of the two-photon detuning Δ_{31} makes it possible to act selectively upon atoms, for which $g < 0$, when the pumping frequency is chosen so that the frequency 2ω falls into the isotope shift interval. These results can readily be extended to three-level systems (see Fig. 10) which interact with two different resonance fields.

5. Electron Raman scattering with resonant amplification

5.1 Illustrative consideration of the theory of electron Raman scattering

With the advent of the first lasers, Raman scattering of light became the subject of intense experimental research. A new area of interest, so-called 'stimulated Raman scattering' (SRS), appeared and was set forth in a number of monographs and reviews [94–96, 98]. Here we shall not touch these traditional issues. Instead we shall dwell on a new subject in Raman scattering, namely, on Raman scattering involving electronic transitions. The impetus to the exploration of this area was given by the first works which Soviet and American scientists conducted independently of each other [134–136]. Here they revealed stimulated Raman scattering involving electronic transitions (SERS) in atoms.

This discovery necessitated the revision of the basic theoretical principles in this area, which had remained practically intact since the famous paper by Kramers and Heisenberg [17]. Starting from simple illustrative concepts we shall examine what changes should be introduced into the theory of Raman scattering of light [17]. Within the scope of perturbation theory, the wave functions of a three-level system in a resonant field, which acts over the $1 \to 2$ transition with the detuning $\Delta_{21} = \Delta$, have the form (see Section 4.1 and Fig. 10b):

$$\begin{aligned} \Phi_1 &\approx U_1 \exp(-\mathrm{i}E_1 t) - C_2^* U_2 \exp\left[-\mathrm{i}(E_1 + \omega)t\right] , \\ \Phi_2 &\approx U_2 \exp(-\mathrm{i}E_2 t) + C_2 U_1 \exp\left[-\mathrm{i}(E_2 - \omega)t\right] , \\ \Phi_3 &\approx \Psi_3 = U_3 \exp(-\mathrm{i}E_3 t) . \end{aligned} \tag{5.1}$$

For

$$|C_2| = \left| \frac{\mathbf{d} \cdot \vec{\mathcal{E}}}{\hbar\Delta} \right| \ll 1$$

functions (5.1) lead to the familiar expression for the probability of Raman scattering of light from Ref. [17], in which only the main resonant term is left. However, the parameter C_2 can be of the order of unity or higher when the system is close to the resonance or the fields are strong. This means that perturbation theory is herein inapplicable. In addition, the high-frequency Stark effect has to be taken into account in laser fields. Consequently, the ERS theory has to be extended to this case as well [5].

The second change which the Kramers–Heisenberg theory needs is that the interaction with a monochromatic wave has to be replaced by the interaction with a quasi-monochromatic pulse of finite extent. As noted in Section 4.1, it is difficult to consider beyond the scope of perturbation theory how the ERS is affected when the interaction between an atom and an electromagnetic wave is switched on. Functions (5.1) are the same as in Ref. [17] and they are equivalent to the adiabatic wave functions for $\alpha \ll 1$, which we examined in detail in Section 4.1.

However, in all experiments on ERS the interaction is switched on at a time $t = t_0$, i.e. when the laser pulse with finite duration reaches the atom or when the travelling atomic beam enters the region of influence of the laser field. To consider this effect, the wave functions of an atom in the field of a laser pulse should be written down under initial conditions. For example, in the perturbation theory limit for $t > t_0$ the desired wave functions have the form

$$\begin{aligned} \Phi_1 &= \Psi_1 - C_2^* \Psi_2 \left[\exp(\mathrm{i}t\Delta) - \exp(\mathrm{i}t_0\Delta)\right] , \\ \Phi_2 &= \Psi_2 + C_2 \Psi_1 \left[\exp(-\mathrm{i}t\Delta) - \exp(-\mathrm{i}t_0\Delta)\right] , \\ \Phi_3 &= \Psi_3 , \end{aligned} \tag{5.2}$$

when the field is switched on infinitely fast at $t = t_0$. For $t \leqslant t_0$, the wave functions Φ_i are specified by the unperturbed atomic functions Ψ_i and by the initial conditions. As $(t - t_0)|\Delta| \to \infty$, the fast-oscillating terms can be omitted and the system of functions (5.2) goes over to functions (5.1). Expressions (5.2) show that the quantity $\left|(t_0 - t)\Delta\right|$ is the characteristic parameter through which the wave functions depend on initial conditions. In the case of a pulse of finite duration the interaction lasts for a time τ and this time interval represents the pulse width. Thus, the quantity $|\Delta\tau|$ is the parameter by which adiabatic (5.1) and nonstationary (5.2) functions are differentiated. Consequently, the Kramers–Heisenberg theory has to be extended to consider the initial activation of the interaction and the pulse shape (see, for example, Refs [5, 137–141]). This problem is closely related to the system relaxation since if the effective time intervals are much longer than the relaxation times then the system 'forgets' the initial conditions.

Given $\tau|\Delta| \gg 1$, we can use functions (5.1) and calculate the electric dipole moment of an atom in the electromagnetic field for the $1 \to 3$ transition:

$$\mathbf{D}_{31} = \int \Psi_3^* e\mathbf{r}\Phi_1 \, \mathrm{d}V = -C_2^* \mathbf{d}_{32} \exp\left[-\mathrm{i}(E_3 - E_1 - \omega)t\right] . \tag{5.3}$$

It follows from (5.3) that the atom in the field has a varying moment and emits photons at the frequency

$$\omega_\mathrm{s} = \omega - E_{31} . \tag{5.4}$$

Similar discussions for functions (5.2) show that for $|\tau\Delta| < 1$ the atom will also emit photons, the frequency of which is

$$\omega_\mathrm{p} = E_2 - E_3 \tag{5.5}$$

in addition to photons at the frequency given by (5.4), i.e. the Raman scattering spectrum should have the resonance line relevant to the $2 \to 3$ transition. As $\Delta \to 0$, these two different lines merge. The resonance line appears because the upper level is actually occupied when the interaction is switched on.

In addition to the above changes, the ERS theory has to be improved in the case when a second strong field acts over the

$2 \to 3$ transition and the atom is in the fields of two resonant waves [142]. Even more important from the practical point of view is the case when both pulses are separated in time [103a, 143].

5.2 Theory of resonance ERS in the monochromatic pumping approximation with regard for saturation effects and Stark shifts

An atom becomes polarized under the action of a pumping field. The dipole moment of an atom in the field is given by the expression

$$\mathbf{D}_{3k} = e \int \Psi_3^* \mathbf{r} \Phi_k \, \mathrm{d}V \qquad (k = 1, 2) \,. \tag{5.6}$$

With $\mathbf{d}_{31} = 0$ and using the adiabatic wave functions of an atom in the laser field exerted on the $1 \to 2$ transition we calculate the dipole moment as

$$\mathbf{D}_{31}^{-} = \left| \frac{\sqrt{1+\alpha^2} - 1}{2\sqrt{1+\alpha^2}} \right|^{1/2} \exp\left[-\mathrm{i}(\omega - E_{31} + \lambda_1)t\right] \mathbf{d}_{32} \,, \tag{5.7}$$

where α and λ_1 are determined by means of the formulae from Section 2.3. In what follows for the sake of simplicity, we shall use functions such that the functions $\Phi_{1,2}$ map onto the wave functions of an atom $\Psi_{1,2}$ when the field is switched off, $\Phi_{1,2} \to \Psi_{1,2}$, i.e. the discontinuous $\overline{\lambda}_i$ ($\overline{\lambda}_i = \lambda_i$ for $\Delta > 0$ and $\lambda_{1,2} \to \lambda_{2,1}$ for $\Delta < 0$) are used.

The dipole moment $\mathbf{D}_{31}^{-}$ brings about emission at the combined frequency ω_s:

$$\omega_\mathrm{s} = \omega - E_{31} + \lambda_1 \,. \tag{5.8}$$

The quantities α and λ_1 in expression (5.7) for a monochromatic wave are time independent. In experiments, time-limited pulses are usually used. We shall assume that the pulse width is smaller than all relaxation times Γ^{-1}:

$$\tau\Gamma \ll 1 \,. \tag{5.8$'$}$$

The time-limited pulse results in the wave actually being quasi-monochromatic. If the spectral width of a pulse $\delta\omega \sim 1/\tau$ is essentially smaller than the resonance detuning Δ, then expression (5.7) can be used for a time-limited 'table-shaped' light pulse. The only difference from an ideal monochromatic wave is that the pulse width τ should be considered to calculate the scattered energy, i.e. to allow for a nonzero polarization amplitude in (5.7) over the time interval τ. Here we neglect any effects associated with the leading and tailing edges of the pulse, i.e. we assume that these effects make a negligible contribution to the scattered radiation intensity. The justification of these assumptions was given in the aforementioned papers. In particular, how the pulse shape affects the ERS processes is considered in Refs [138–141]. The calculations show [138–141] that consideration of the pulse shape brings about a number of interesting phenomena, which have not yet been experimentally verified.

The probability of dipole emission of a photon at frequency ω_s with polarization $\mathbf{e}_\mathrm{s}$ is usually calculated by means of the regular formula for the dipole moment $\mathbf{D}_{31}$ of emission and given by the expression [5]

$$\mathrm{d}W_\mathrm{s} = \frac{\omega_\mathrm{s}^3 \left(\sqrt{1+\alpha^2} - 1\right)}{4\pi\hbar c^3 \sqrt{1+\alpha^2}} \left|\mathbf{e}_\mathrm{s}^* \cdot \mathbf{d}_{23}\right|^2 (1 + n_\mathrm{s}) \, \mathrm{d}O \,. \tag{5.9}$$

The amplification factor

$$1 + n_\mathrm{s} = 1 + \frac{8\pi^3 c^2}{\hbar\omega_\mathrm{s}^3} I_\mathrm{s}(\omega_\mathrm{s}, O_\mathrm{s})$$

considers the spontaneous and stimulated Raman scattering. Depending on the type of scattering $\mathrm{d}O_\mathrm{s}$ is understood to be either the solid angle of a spontaneously scattered photon or the solid angle of the angular distribution of the driving radiation. In (5.9), the quantity $I_\mathrm{s}(\omega_\mathrm{s}, O_\mathrm{s})$ is the spectral-angular intensity density in the direction of the solid angle $\mathrm{d}O$. When $\alpha \ll 1$, expression (5.9) goes over to the expression that Kramers and Heisenberg [17] obtained for the three-level atom.

When $\alpha \gg 1$, the probability of Raman scattering (5.9) takes the form

$$\mathrm{d}W_\mathrm{s} = \frac{\omega_\mathrm{s}^3}{4\pi\hbar c^3} \left|\mathbf{e}_\mathrm{s}^* \cdot \mathbf{d}_{23}\right|^2 (n_\mathrm{s} + 1) \, \mathrm{d}O_\mathrm{s} \,, \qquad \alpha \gg 1 \,. \tag{5.9$'$}$$

On integrating with respect to the solid angle and summing over polarizations, expression (5.9$'$) can be easily presented in the form $W_\mathrm{s} = \gamma_{23} |C_2|^2$ for $n_\mathrm{s} = 0$. Here γ_{23} is the probability of a spontaneous transition $2 \to 3$ (for $\alpha = 0$), and C_2 is the coefficient of the wave function Φ_1 for $\Delta > 0$. Hence, $W_\mathrm{s} = \gamma_{23}/2$ when the $1 \to 2$ transition is saturated and $|C_2|^2 = 1/2$. Unfortunately, expression (5.9$'$) has not been verified experimentally so far.

Formula (5.9$'$) describes the saturation effect in Raman scattering. In this limiting case the second level has the same population as the first level and the number of scattered quanta does not grow upon a further increase in the intensity of radiation incident on the atom.

If the intensity of the scattered radiation becomes comparable with that of the exciting radiation, then perturbation theory becomes inapplicable with respect to the scattered field. Therefore, it is interesting to study the behaviour of an atom in two intense resonant fields without the use of the perturbation theory with respect to both fields [142] (see also the review [137]).

5.3 ERS in two intense fields

In the previous section we have assumed that Raman scattering is caused by the 'pumping' field which is in resonance with the $1 \to 2$ transition. We have neglected the influence of the electromagnetic field of combined-frequency photons on the atomic system. This section is dedicated to the consideration of this effect. The point is that the 'atom plus pumping field plus field of combined-frequency photons' system is degenerate in terms of energy since the electron residing at the ground level moves by absorbing a 'pumping' photon ω_1 and emitting a combined-frequency photon ω_s to the steady-state third level. This process is governed by the following law of conservation:

$$E_1 + \omega_1 - \omega_\mathrm{s} = E_3 \,, \quad \Delta_{31} = 0 \,, \quad \Delta_{21} = -\Delta_{32} = \Delta \,. \tag{5.10}$$

These states are described by two wave functions of the complete 'atom plus pumping field plus field of combined-frequency photons' system with the same energy. Strictly speaking the wave functions are incorrect if they do not account for the degeneracy. In the zero-order approximation the correct functions must be linear combinations of the expressions we wrote out for $\Phi_{1,2}$ and Φ_3 [142].

If we know the wave functions (4.15) for the scheme in Fig. 10a, then wave functions can also be found to describe

the Raman scattering process in the field of two waves for the scheme in Fig. 10b. To this end $\mathcal{E}_2$ should be replaced by $\mathcal{E}_2^*$ in the expression for V_{32} in formulae (4.15) since absorption by scheme in Fig. 10a corresponds to emission by scheme in Fig. 10b; and the sign of ω_2 should be changed for the opposite one. In this section it is convenient to use another numbering rather than that we adopted in Eqn (4.15). To this end the change $2 \leftrightarrow 3$ is to be made in all subscripts for quasi-energies and wave functions in Section (4.2). In this case $\Phi_1 \to \Psi_1$, $\Phi_2 \to \Psi_2$, $\Phi_3 \to \Psi_3$ when the field is switched off ($\Delta > 0$), and Eqn (4.14) has the solutions

$$\lambda_3 = 0\,,$$
$$\lambda_{2,1} = \frac{\Delta}{2} \pm \sqrt{\frac{\Delta^2}{4} + |V_{21}|^2 + |V_{32}|^2} \tag{5.11}$$

for $\Delta_{31} = 0$. In the quasi-energy function Φ_3 in (4.15), the coefficient of the Ψ_3 function is indefinite as $\lambda_3 \to 0$ and $\Delta_{31} \to 0$. To eliminate the ambiguity, the cubic equation (4.14) has to be invoked. As is seen, Eqn (4.14) is followed by the equation

$$\frac{\lambda}{\lambda - \Delta_{31}} = \frac{\lambda(\lambda - \Delta_{21}) - |V_{21}|^2}{|V_{32}|^2}\,.$$

In turn, this last equation leads to the following equality

$$\frac{\lambda_3}{\lambda_3 - \Delta_{31}} = -\left|\frac{V_{12}}{V_{23}}\right|^2 \tag{5.12}$$

for $\Delta_{31} = 0$ and $\lambda = \lambda_3 = 0$. Consequently, the adiabatic functions in scheme in Fig. 10b have the form [142]

$$\Phi_{1,2} = \frac{\alpha_1}{\sqrt{2p(p \mp 1)}}\left\{\Psi_1 + \frac{\mathbf{d}_{32}\cdot\vec{\mathcal{E}}_2^*}{\mathbf{d}_{12}\cdot\vec{\mathcal{E}}_1^*}\,\Psi_3 + \frac{\hbar\Delta(1 \mp p)}{2(\mathbf{d}_{12}\cdot\vec{\mathcal{E}}_1^*)}\,\Psi_2 \exp(\mathrm{i}\Delta t)\right\}\exp\left[\mathrm{i}\frac{\Delta}{2}(\pm p - 1)t\right], \tag{5.13}$$

$$\Phi_3 = \frac{\alpha_2}{\sqrt{\alpha_1^2 + \alpha_2^2}}\left\{\Psi_1 - \frac{\mathbf{d}_{21}\cdot\vec{\mathcal{E}}_1}{\mathbf{d}_{23}\cdot\vec{\mathcal{E}}_2}\,\Psi_3\right\}, \tag{5.14}$$

where the notation

$$\alpha_1 = \frac{2}{\hbar}\left|\frac{\mathbf{d}_{21}\cdot\vec{\mathcal{E}}_1}{\Delta}\right|, \qquad \alpha_2 = \frac{2}{\hbar}\left|\frac{\mathbf{d}_{23}\cdot\vec{\mathcal{E}}_2}{\Delta}\right|, \tag{5.15}$$

$$p = (1 + \alpha_1^2 + \alpha_2^2)^{1/2}\,. \tag{5.16}$$

is used.

The wave functions (5.13) and (5.14) are chosen so that the functions $\Phi_{1,2,3}$ should go over to the functions $\Psi_{1,2,3}$, respectively, when the fields are switched off (first $\vec{\mathcal{E}}_2$ and then $\vec{\mathcal{E}}_1$) and when $\Delta > 0$. For negative detunings, the passages to the limits $\Phi_{1,2} \to \Psi_{2,1}$, $\Phi_3 \to \Psi_3$ take place when the fields are switched off. An in-depth analysis of solutions to the cubic equation shows that the correspondence between the wave functions Φ_1, Φ_2, Φ_3 and the three states 1, 2, 3 of a free atom depends on the order in which the fields are switched off. For example, if first field is switched off, $\vec{\mathcal{E}}_1 \to 0$, and next $\vec{\mathcal{E}}_2 \to 0$, then $\Phi_1 \to \Psi_3$, $\Phi_2 \to \Psi_2$, $\Phi_3 \to \Psi_1$ for $\Delta > 0$.

Note that the wave functions (5.13) and (5.14) are written out in the event when the electromagnetic fields $\mathcal{E}_1$ and $\mathcal{E}_2$ are defined as classical and, therefore, different summands in the braces in these expressions correspond to various energies of the atom. However, if we rewrite these functions for the quantized fields $\vec{\mathcal{E}}_1$ and $\vec{\mathcal{E}}_2$, then the expression

$$\Psi_1 - \frac{\mathbf{d}_{21}\cdot\vec{\mathcal{E}}_1}{\mathbf{d}_{23}\cdot\vec{\mathcal{E}}_2^*}\,\Psi_3 \tag{5.17}$$

goes over to the expression

$$\Psi_1|n_1, n_2, \ldots\rangle - \frac{\mathbf{d}_{21}\cdot\vec{\mathcal{E}}_1}{\mathbf{d}_{23}\cdot\vec{\mathcal{E}}_2}\,\Psi_3|n_1 - 1,\, n_2 + 1,\, \ldots\rangle \tag{5.17$'$}$$

with the same energy in both summands.

The fact that the two states are degenerate in terms of energy means that in any calculations both states should be considered in the wave functions $\Phi_{1,2}$ and Φ_3 as superpositions, and this can be readily seen from the structure of formulae (5.13) and (5.14). Hence, the form of these functions makes it possible to ascertain that the total wave function of the 'atom plus field' system has a particular energy once the wave fields $\vec{\mathcal{E}}_1$ and $\vec{\mathcal{E}}_2$ were quantized, and from the start we are dealing with the states $\Phi_{1,2}$ and Φ_3, in which the degeneration in terms of the total energy is taken into account.

Note that the wave functions (5.13)–(5.15) are obtained under the assumption that the atom interacts with the external field adiabatically. Therefore, these functions are correctly applicable only when the external field has a rather large intensity and thus the spacing between any quasi-energy terms is sufficiently larger than any width, which is due to the widths of atomic levels as well as to the nonmonochromatic nature of the fields. In particular, the conditions

$$|\lambda_i - \lambda_j| \gg \delta\omega_{1,2} \quad \text{for} \quad i \neq j\,, \tag{5.18}$$

where $\delta\omega_{1,2}$ are the relevant widths, have to be satisfied. Only in this case can the interaction be considered adiabatic and the nonadiabatic transitions between the i and j states be neglected. When one of the fields is switched on (off), the other field should be intense enough for condition (5.18) to be fulfilled and, hence, for the interaction to be adiabatic at this stage. In this event the functions Φ_j, which describe the state of the 'atom in the field of two waves' system, are uniquely mapped to the wave functions of the 'atom in the field of a wave' system.

Currently functions (5.13) and (5.14) are widely used. However, they are usually obtained with the aid of the so-called 'rotating wave approximation' (RWA) in other notation. To compare the results of works [137, 142] with those of Refs [144–147], let us introduce the angular variables θ and φ. Using formulae

$$\sin\theta = \frac{\alpha_1}{\sqrt{\alpha_1^2 + \alpha_2^2}}\,, \quad \sin\varphi = \sqrt{\frac{p+1}{2p}}\,, \tag{5.15$'$}$$

(5.13) and (5.14) in the new notation can be written in a simpler form

$$\Phi_1 = \exp(-\mathrm{i}\lambda_1 t)\Big[\sin\varphi\big(\Psi_1\sin\theta + \Psi_3\cos\theta\exp(\mathrm{i}\varphi_0')\big) - \Psi_2\cos\varphi\exp\big[\mathrm{i}(\varphi_0'' + \Delta t)\big]\Big]\,,$$
$$\Phi_2 = \exp(-\mathrm{i}\lambda_1 t)\Big[\cos\varphi\big(\Psi_1\sin\theta - \Psi_3\cos\theta\exp(\mathrm{i}\varphi_0')\big) + \Psi_2\sin\varphi\exp\big[\mathrm{i}(\varphi_0'' + \Delta t)\big]\operatorname{sgn}\Delta\Big]\,, \tag{5.13$'$}$$

$$\Phi_3 = \Psi_1\cos\theta - \Psi_3\sin\theta\exp(\mathrm{i}\varphi_0')\,, \tag{5.14$'$}$$

where

$$\varphi_0' = \varphi_1 - \varphi_3 + \varphi' - \varphi'' , \quad \varphi_0'' = \varphi_1 - \varphi_2 + \varphi' , \qquad (5.14'')$$

here φ' and φ'' are the constant phases of the fields $\mathcal{E}_1$ and $\mathcal{E}_2$, $\varphi_{1,2,3}$ are the random phases of the atomic wave functions $\Psi_{1,2,3}$ (2.3). They must be retained when considering coherent effects. Adiabatic functions (5.13) agree with functions (2.25), (2.26) for $\mathcal{E}_2 \to 0$ and $\Delta \to 0$, i.e. they are represent the symmetric and antisymmetric combinations of Ψ_1 and Ψ_2. It should be emphasized that the functions Ψ_1 and Ψ_3 enter the formulae (5.13) and (5.14) in certain combinations, being the consequence of the 'three-level atom plus radiation'-system degeneration in energy.

Currently the states (5.14′) introduced in Refs [142, 146, 147] and called the 'trapping states' are being intensively studied and widely used in various physical processes†. For example, they are employed for inversion-free‡ lasers [148, 149], laser cooling of atoms [150–152] and coherent transfer of atomic populations [143–145, 152–156a] and atomic interferometry [156b, c]. The cited problems are set forth in reviews [157, 158] and in the special issue of *JOSA* **36** (1989). Therefore, we shall not dwell on them any more and come to issues which have inadequate presentation in the scientific literature.

5.4 Radiative processes in $\Phi_i \leftrightarrow \Phi_j$ transitions

Now we shall consider the emission and absorption processes in the atomic system in transitions between the different states Φ_1, Φ_2, and Φ_3. Using wave functions (5.13) and (5.14), the spontaneous dipole moment in the $\Phi_1 \to \Phi_3$ transition can be represented as

$$\mathbf{D}_{31} = C_1 C_2^* \exp(-\mathrm{i}\lambda_1 t)\left[\mathbf{d}_{12}\frac{\lambda_1}{V_{12}}\exp(-\mathrm{i}\omega_1 t) - \frac{\lambda_1 \mathbf{d}_{31}}{V_{23}^*}\exp\left[\mathrm{i}(E_{31}-\omega_1)t\right]\right] . \qquad (5.19)$$

As is seen from formula (5.19), the system emits photons with frequencies

$$\omega_1' = \omega_1 + \lambda , \qquad (5.20)$$

$$\omega_2' = \omega_2 + \lambda \qquad (5.21)$$

in the course of $\Phi_1 \to \Phi_3$ transition.

† The physical nature of the 'trapping' states can be easily interpreted with the use of Eqn (4.6) in the case of an exact resonance, $\Delta_{31} = 0$ ($\Delta_{21} = -\Delta_{32}$). Assuming for simplicity that $V_{21} = W_{32} = V$ and introducing the combinations $b_{1,3} = (a_1 \pm a_3)/\sqrt{2}$ intstead of the amplitudes a_1 and b_3, Eqn (4.6) can be rewritten in the form: $i\dot{b}_1 = \sqrt{2}V^* a_2$, $i\dot{b}_3 = 0$, $i\dot{a}_2 = \sqrt{2}Vb_1$. It follows from the equations that the state of the system, which the amplitude b_3 represents, does not change during the atom-field interaction. The pertinent wave functions of the system can also be easily found using, for example, the technique developed in Section 3.3.

‡ The issue of light amplification without level population inversion has a long history [5, 27, 43b, 48, 50b, 51]. In Ref. [27], amplification of the so-called three-photon radiation was experimentally investigated when the radiation propagates through the two-level atomic gas in a strong resonant field. G Kryuchkov et al. [39b] showed that the cause of light amplification in the absence of level population inversion is a two-photon emission, induced by one photon field and spontaneous with respect to the other field. Recently this issue was again discussed by P Sellin et al. [161]. It seems likely that the authors were unacquainted with the aforementioned references since they put forward a similar interpretation of the amplification processes in the absence of inversion.

The probabilities that photons are emitted at the frequencies ω_1' and ω_2', are determined by the expressions [142]

$$\mathrm{d}W_1' = \frac{\omega_1'^3 \alpha_2^2 (p-1)}{4\pi\hbar c^3(\alpha_1^2+\alpha_2^2)p}\,|\mathbf{e}_1'^* \cdot \mathbf{d}_{12}|^2 (n_1'+1)\,\mathrm{d}O_1' , \qquad (5.22)$$

$$\mathrm{d}W_2' = \frac{\omega_2'^3 \alpha_1^2 (p-1)}{4\pi\hbar c^3(\alpha_1^2+\alpha_2^2)p}\,|\mathbf{e}_2'^* \cdot \mathbf{d}_{32}|^2 (n_2'+1)\,\mathrm{d}O_2' . \qquad (5.23)$$

The radiation with the frequency ω_2', emitted with a probability defined by (5.23), corresponds to the Stokes components of Raman scattering in the strong fields $\mathcal{E}_1$ and $\mathcal{E}_2$, when the ω_1 photon is absorbed from the field $\mathcal{E}_1$, the ω_2' photon is emitted by the atomic system, and the system goes to the state Φ_3. When $\alpha_2^2 \ll 1$, the probability defined by (5.23) goes over to expression (5.9) from the previous section and describes the ERS accounting for the saturation effect.

The emission of photons with the frequency ω_1' (5.20) and a probability defined by (5.22) is somewhat unusual and interrupts as $|\mathcal{E}_2| \to 0$. As a result of the interaction with the fields $\mathcal{E}_1$ and $\mathcal{E}_2$ (the photon absorption at the frequency ω_1 and the photon emission at the frequency ω_2') the atomic functions Ψ_1 and Ψ_3 get highly mixed and form new stationary states $\Phi_{1,2}$ and Φ_3. Therefore, in the $\Phi_1 \to \Phi_3$ transition the system absorbs a photon ω_2 from the field $\mathcal{E}_2$ and emits a photon ω_1' since the third level is actually populated (due to the above process).

In the $3 \to 1$ transition, the photons are emitted and absorbed at frequencies shifted by the quantity $(p-1)\Delta$ relative to (5.20) and (5.21). As a consequence, the emission and absorption processes occur at different frequencies and do not compensate for one another.

The other transition probabilities we considered in the previous section are generalized in a similar manner. However, in the case of two intense fields scattering can develop through other new channels. In particular, the wave functions (5.13) and (5.14) bring about the coherent scattering of photons with the frequencies ω_1 and ω_2. First it is necessary to calculate the polarization in the state Φ_1. As is easily seen, the dipole moment in the state Φ_1 is

$$\mathbf{D}_{11} = C_1^2\left[\frac{\lambda_1}{V_{12}}\mathbf{d}_{12}\exp(-\mathrm{i}\omega_1 t) + \frac{\lambda_1 V_{32}}{|V_{12}|^2}\mathbf{d}_{32}\exp(-\mathrm{i}\omega_2 t) + \mathrm{c.c}\right], \qquad (5.24)$$

where

$$C_1^2 = \frac{|V_{12}|^2}{|V_{12}|^2 + \lambda_1^2 + |V_{32}|^2} . \qquad (5.25)$$

The first term in the dipole moment $\mathbf{D}_{11}$ is responsible for photon emission at the frequency ω_1, while the second is responsible for photon emission at the frequency ω_2. The complex conjugate terms in the expression are answerable for the absorption of just the same photons with the same probability. Therefore, only the spontaneous processes of photon emission at ω_1 and ω_2 occur and they match the Rayleigh-type photon scattering at ω_1 and ω_2. The above coherent light scattering by a three-level system in the field of two waves with the frequencies ω_1 and ω_2 have not been examined in detail and have not been investigated experimentally.

It should be emphasized in conclusion that, depending on the relationships between the field intensities $\mathcal{E}_1$ and $\mathcal{E}_2$ and

the matrix elements $\mathbf{d}_{21}$ and $\mathbf{d}_{23}$, the mapping between the indices of the quasi-energy functions Φ_i and those of the atomic wave functions Ψ_i is different for various detunings. This means that the atomic wave functions develop along various reaction channels but they can be easily classified for all the transitions $\Phi_i \to \Phi_j$, $i = 1, 2$, $j = 1, 2$.

5.5 Coherent population transfer in the field of two pulses with a time delay

Currently the populations of levels in a three-level system in the wave fields is attracting considerable attention of many researchers. Earlier we mentioned the papers on the ladder-type schemes Ξ. In this section we consider the Λ-system when the exact one-photon resonance, $\Delta \to 0$, occurs. Then the wave functions (5.13′) and (5.14′) can be written in the following form

$$\Phi_1(t) = \frac{1}{\sqrt{2}}(\Psi_1 \sin\theta + \Psi_2 + \Psi_3 \cos\theta)\exp(-\mathrm{i}\lambda_1 t)\,, \tag{5.26}$$

$$\Phi_2(t) = \frac{1}{\sqrt{2}}(\Psi_1 \sin\theta - \Psi_2 + \Psi_3 \cos\theta)\exp(-\mathrm{i}\lambda_2 t)\,, \tag{5.27}$$

$$\Phi_3(t) = \Psi_1 \cos\theta - \Psi_3 \sin\theta\,. \tag{5.28}$$

As noted above, the states described by (5.28) have remarkable properties and are known in the literature as 'trapping states'. Using these states, the atomic populations can be transferred coherently and efficiently from the atomic state Ψ_1 to Ψ_3. In fact, if the atom resides in the state Ψ_1 up to the instant when the laser fields are switched on and if the interaction with the Stokes field $\mathcal{E}_2 = \mathcal{E}_\mathrm{s}$ is switched on and off prior to the pumping field $\mathcal{E}_1 = \mathcal{E}_\mathrm{p}$, then, as is seen from (5.15′), θ varies from 0 to $\pi/2$. Clearly, in this case the atom goes from the state Ψ_1 to the state Ψ_3. For this process to occur it is required that the atom be in this 'trapping state' over the entire course of the interaction. This is possible when the following condition

$$\dot{\theta} \ll \sqrt{\alpha_1^2 + \alpha_1^2} \tag{5.29}$$

is satisfied.

This phenomenon is known as stimulated Raman adiabatic passage (STIRAP). It was first predicted in Ref. [143] and experimentally revealed in Ref. [144a]. Figure 18, taken from Ref. [153], shows the results of experiments on the population transfer from the ground state of an Na_2 molecule to the excited state. As is seen from Fig. 18a, efficient population transfer for $\Delta = 0$ occurs only if the Stokes component is switched on and off prior to the others. In Fig. 18b, the experiment was conducted for a large intermediate detuning Δ, i.e. when $\alpha_1\alpha_2/\Delta \leqslant 1$. In this case the Λ-system in question corresponds to a two-level system in which the Stark shifts are allowed for [103a, 104]. Therefore, the order in which the fields are switched on and off is not critical since the process is adiabatic when there is a detuning Δ. It can be seen from Fig. 18b that there appears a second peak with respect to the excitation of Na_2 molecules even if the field is switched on earlier.

Therefore, it is interesting to consider the opposite situation when the atomic system is exposed first to the pumping $\mathcal{E}_\mathrm{p}$ and then to the $\mathcal{E}_\mathrm{s}$ field under the adiabaticity condition (5.29). Condition (5.29) can be written in the explicit form as

$$\sqrt{\mathcal{E}_\mathrm{p}^2 d_{21}^2 + \mathcal{E}_\mathrm{s}^2 d_{32}^2}\,T \gg 1\,, \tag{5.29′}$$

where T is the duration of the interaction between the light pulses and the atom, and the time delay is $\tau \leqslant T$.

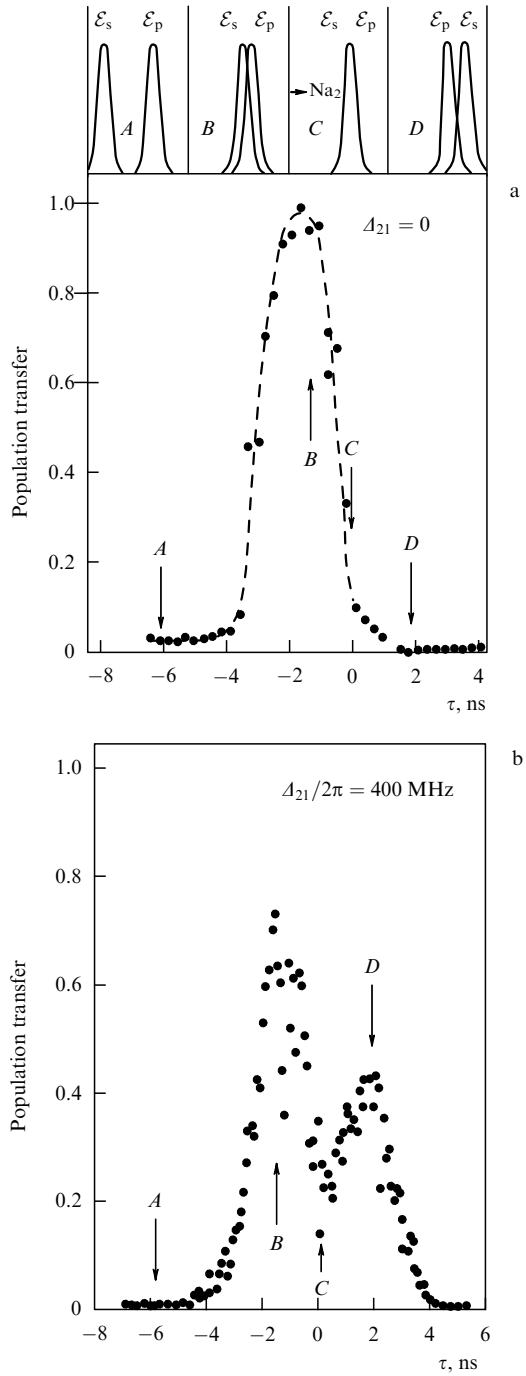

Figure 18. Results of experiments on the population transfer from the ground level to the excited level depending on the time delay between the fields $\mathcal{E}_\mathrm{s}$ and $\mathcal{E}_\mathrm{p}$ [153]. As is seen from Fig. 18a, efficient population transfer occurs only when $\mathcal{E}_\mathrm{s}$ is switched on and off prior to $\mathcal{E}_\mathrm{p}$. The experiment was conducted with molecular beams of Na_2, which interacted with spatially separated continuous laser pulses at the exact resonance $\Delta_{21} = 0$. Figure 18b shows the results of the experiment in the case of a large intermediate detuning.

In Ref. [159], the exact solutions were obtained for the interaction between two laser pulses with a time delay 2τ and the three-level Λ-system when the pulses are at the exact resonances with the $1 \to 2$ and $2 \to 3$ transitions. For $\tau > 0$, where τ is the time delay between pulses (see Fig. 19), i.e. when the field $\mathcal{E}_s$ is switched on prior to $\mathcal{E}_p$, the results are the same as those in Ref. [144a]. In the case when $\tau < 0$, a simple analytical solution was obtained such that the excited level populations broke into oscillations. The oscillation frequency depended on the effective areas of two pulses and on the time delay τ between them. After the interaction, i.e. as $t \to \infty$, the atomic populations are

$$n_1 = 0\,, \quad n_2 = \sin^2\beta\,, \quad n_3 = \cos^2\beta\,, \tag{5.30}$$

where

$$\beta = \frac{1}{2}\int_{-\infty}^{\infty} \left[\alpha_1^2 + \alpha_2^2\right]^{1/2} \mathrm{d}t\,. \tag{5.31}$$

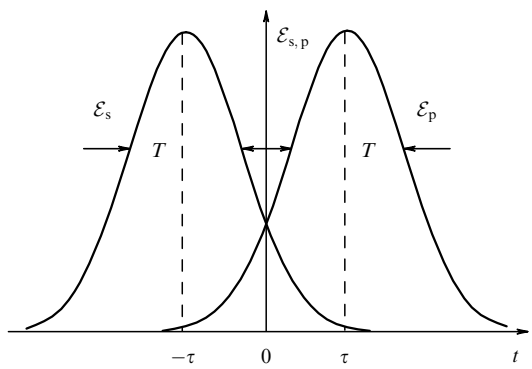

Figure 19. Temporal distributions and time delays for $\mathcal{E}_p$ and $\mathcal{E}_s$.

The quantity β depends on the pulse intensity and the time delay between them. For $\beta = 2\pi n$, the atom goes to the state Ψ_3, and for $\beta = \pi(n + 1/2)$ it goes to the state Ψ_2, i.e. we shall observe oscillations of populations for the second and third states depending on τ. Since spontaneous decay was not considered, solution (5.30) is valid only for $T \ll t_{sp}$. Therefore, in addition to (5.29) the condition $T \ll t_{sp}$ has to be satisfied for this effect to be examined experimentally. For instance, if $T \leqslant 10^{-9}$ s, then $\mathcal{E} \geqslant 10^3$ W cm^{-1}. Oscillations of populations in the state Ψ_3 were experimentally revealed in Ref. [145a]. However, thorough measurements are required to check expression (5.30).

5.6 Scheme *V*

The scheme in Fig. 10c (the V-system) does not require a separate consideration. The wave functions for the 'atom of the V-type in two fields which are close to resonances with the $1 \to 2$ and $1 \to 3$ transitions' system are obtained from the expression for Φ_s (4.15) by reindexing $1 \leftrightarrow 2$ and by replacing $\omega_1 \to -\omega_1$. Then, instead of (4.15) we have [160]

$$\Phi_s = C_s' \exp\left[-\mathrm{i}(E_1 + \lambda_s + \Delta_{21})t\right] \times \left\{U_1 + \frac{V_{21}}{\lambda_s}\exp(-\mathrm{i}\omega_1 t)U_2 + \frac{V_{31}}{\lambda_s - \Delta_{32}}\exp(-\mathrm{i}\omega_2 t)U_3\right\} \tag{5.32}$$

with the normalization factor C_s' being as

$$C_s' = \left\{1 + \frac{|V_{21}|^2}{\lambda_s^2} + \frac{|V_{31}|^2}{(\lambda_s - \Delta_{32}^2)}\right\}^{-1/2}. \tag{5.33}$$

The quantities λ_s obey Eqn (4.14) in which all the above replacements are made. Then (4.14) takes the form

$$\lambda(\lambda + \Delta_{21})(\lambda - \Delta_{32}) - (\lambda - \Delta_{32})|V_{21}|^2 - \lambda|V_{31}|^2 = 0\,, \tag{5.34}$$

where $\Delta_{32} = E_3 - E_2 + \omega_1 - \omega_2$, and $\Delta_{21} = E_2 - E_1 + \omega_1$.

In the case of the exact resonances, $\Delta_{21} = \Delta_{32} = 0$, functions (5.32) can be written in the following form

$$\Phi_3 = \Psi_2 \cos\theta - \Psi_3 \sin\theta\,, \tag{5.35}$$

$$\Phi_{1,2} = \frac{\exp(\pm\mathrm{i}\lambda t)}{\sqrt{2}}\left[\Psi_2 \sin\theta \pm \Psi_1 + \Psi_3 \cos\theta\right], \tag{5.36}$$

where

$$\lambda = \frac{2}{\hbar}\sqrt{|\mathbf{d}_{12}\cdot\vec{\mathcal{E}_1}|^2 + |\mathbf{d}_{13}\cdot\vec{\mathcal{E}_2}|^2}\,, \tag{5.37}$$

$$\operatorname{tg}\theta = \left|\frac{\mathbf{d}_{12}\cdot\vec{\mathcal{E}_1}}{\mathbf{d}_{13}\cdot\vec{\mathcal{E}_2}}\right|. \tag{5.38}$$

Thus, most of the results obtained for the Λ-systems can easily be extended to the V-systems. However, the coherence is violated due to the radiative decay because for V-systems the 'trapping' state (5.35) is represented by a combination of two atomic functions, which are at upper levels. As a consequence, the so-called Zeno's paradox can be explained through this violation of coherence [162, 163].

6. Conclusions

This review is written for the attention of physicists who work actively on the problem of the interaction between radiation and an atom as well as those who are just now entering this interesting and promising field. Over the whole review I have striven to show that the study of 'pure' phenomena (neglecting collisions, atomic motions, and other relaxation phenomena) near the resonance transitions can be of paramount importance for fundamental and applied problems. The recent advances in atomic 'cooling', and experiments with a 'gas' of impurity atoms in solids at ultralow temperatures open up possibilities for experiments, in which the influence of relaxation processes can be neglected, and thus to advance further on the path to new physical results. The only relaxation which cannot be eliminated is the radiative decay itself which will always be present in experiments on the interaction between radiation and matter. Because of this we have given considerable attention to so-called 'first-order processes with respect to a weak field' since these processes govern decay and relaxation of wave functions for 'atoms in resonant intense fields'.

I also want to note that with the enormous growth of information each year and with the high degree of specialization of scientists it is hard to keep track of all the relevant publications. To write this review I, for example, have had to associate a series of recent publications with results which were obtained 15 – 20 years ago. In this connection I want to thank all the participants of the traditional meeting in Ashtarak on the subject of 'The Resonance Interaction between Laser Radiation and Matter', who at different

times since the 70th have taken part in discussions of the issues I set forth in this review.

Unfortunately, many issues have not been covered in this review. First of all, these are the issues in which a quantized electromagnetic field must be used. As to the issues in which the classical representation of a field can be used, we have not considered: four-level systems in resonant fields; the 'atom plus field' system in external constant fields; polarization phenomena; transitions involving continuous spectra, ionization processes, etc. I hope to cover this gap in a subsequent paper.

A reach variety of new results have been obtained during the past year in the microscopic nonlinear optics. I want to list below the most essential achievements which can be a help for readers in orienting themselves to the tendency toward the advancement of this field of science.

(1) Experiments on resonance fluorescence are conducted with participation of separate ions trapped and cooled down to ultralow temperatures. In Ref. [164], the measurement accuracy of order a few hertz was achieved by using heterodyne spectral method. Classical and quantum (antigrouping) characteristics of resonance fluorescence were separated and studied.

(2) Works on creation of one-atom laser have gained wide acceptance. In a series of publications (see, for instance, Ref. [165] and references cited therein) the results of experimental and theoretical investigations in this field have been presented.

(3) The study along a fresh direction which came into being at the interface between quantum optics and information theory (quantum computers) is proceeding vigorously. A particularly great amount of works was published recently (see, for example, Ref. [166] and references cited therein). Practically important results, which seem now to be illusive, may be achieved in this field of research.

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7. Appendix

When considering the first-order processes in Sections 2.5, 2.6 we have neglected the upper-level decay, i.e. we have assumed that the condition $2|\mathbf{d}\cdot\vec{\mathcal{E}}| \gg \hbar\Gamma$ is satisfied. Therefore, the results from Refs [40, 43a] should be considered in the limit $\Gamma \to 0$ and $\alpha^2 \gg 1$ before they can be compared with the results from Sections 2.5, 2.6. At first we compare the expressions for resonance fluorescence.

Using the formula

$$\lim \frac{\Gamma}{(\omega'-\omega)^2-\Gamma^2} = \pi\delta(\omega'-\omega)\,,$$

we obtain the expression for the spectral distribution of resonance fluorescence from Ref. [40] for $\alpha^2 \gg 1$:

$$g(\omega') = \frac{\pi}{2\alpha^2}\delta(\omega'-\omega) + \frac{\pi}{2}\delta(\omega'-\omega) + \frac{\pi}{4}\delta(\omega'-\omega+\Omega) + \frac{\pi}{4}\delta(\omega'-\omega-\Omega)\,. \qquad (A.1)$$

The first two terms in the above expression correspond to the coherent and incoherent parts of the unshifted-in-frequency (Rayleigh) scattering. In this limit the relevant emission probabilities from [40] are exactly the same as those in (2.56), (2.60) for the sudden activation of interaction. The unshifted-in-frequency, common scattering is fully coherent when the interaction is switched on adiabatically, as opposed to the case of a sudden activation when it is fully incoherent for $\alpha^2 \gg 1$ and its probability is equal to the sum of probabilities from expressions (2.56), (2.60). This can be understood from the fact that $\mathbf{D}_{11} = -\mathbf{D}_{22}$ and

$$\Phi' = \frac{1}{\sqrt{2}}(\Psi_1 + \Psi_2)$$

for $\alpha \gg 1$. The next two terms in the expression (A.1) for the spectral distribution over the side frequencies $\omega \pm \Omega$ and the relevant expressions from Ref. [40] are exactly the same as (2.61) and (2.62) for $\Gamma \to 0$ and $\alpha^2 > 1$ (provided that the spontaneous components are extracted, i.e. $n_{\mathbf{k}'\mathbf{e}'} = 0$ is set). In the resonance fluorescence a single side line appears on the left or on the right from the strong resonant-field frequency ($\omega' = \omega - \Omega$ for $\Delta > 0$, and $\omega' = \omega + \Omega$ for $\Delta < 0$), when the interaction is switched on adiabatically.

In a similar manner the results from Ref. [43a] can be compared against expressions for the stimulated absorption and emission processes for the system 'atom plus resonance field' in Sections 2.4 and 2.5. For $\alpha^2 \gg 1$, (2.61) and (2.62) are followed by the expressions for the stimulated emission ($\mathrm{d}W > 0$) and stimulated absorption ($\mathrm{d}W < 0$) at frequencies $\omega \pm \Omega$, respectively:

$$\mathrm{d}W_{\omega\mp\Omega} = \frac{\pm(\omega\mp\Omega)^2}{8\pi\hbar c^3\alpha}|\mathbf{d}\cdot\mathbf{e}'^*|n_{\mathbf{k}'\mathbf{e}'}\,\mathrm{sgn}\,\Delta\,\mathrm{d}O'\,. \qquad (A.2)$$

The expression (A.2) should be divided by α when the interaction is switched on adiabatically.

References

1. Landau L D, Lifshitz E M *Kvantovaya Mekhanika* (Quantum Mechanics) (Moscow: Nauka, 1980) [Translated into English (Oxford: Pergamon Press, 1977)
2. Schiff L I *Quantum Mechanics* 2nd ed. (New York: McGraw-Hill, 1955) [Translated into Russian (Moscow: Inostrannaya Literatura, 1959)]
3. (a) Bogolyubov N N, Mitropol'skiĭ Yu A *Asimptoticheskie Metody v Teorii Nelineinykh Kolebanii* (Asymptotic Methods in the Theory of Nonlinear Oscillations) (Moscow: Fizmatgiz, 1958) [Translated into English (Delhi, NY: Gordon Breach Sci. Publ., 1961)]
(b) Sen-Gupta N D *Indian J. Phys.* **8** 127 (1970); *J. Phys. A: Gen. Phys.* **3** 618 (1970); *Rev. Roum. Phys.* (Rumania) **15** 1003 (1970)
4. Ter-Mikaelyan M L, Melikyan A O *Zh. Eksp. Teor. Fiz.* **58** 281 (1970) [*Sov. Phys. JETP* **31** 153 (1970)]
5. Ter-Mikaelyan M L *Nelineinaya Rezonansnaya Optika* (Nonlinear Resonance Optics) Preprint IFI Akad. Nauk Armenii No. 74 (Erevan: IFI, 1974)
6. Rabi I I *Phys. Rev.* **51** 652 (1937)
7. Schwinger J *Phys. Rev.* **51** 648 (1937)
8. Bloch F, Siegert A *Phys. Rev.* **57** 522 (1940)
9. Abragam A *The Principles of Nuclear Magnetism* (Oxford: Clarendon Press, 1961) [Translated into Russian (Moscow: Inostrannaya Literatura, 1963)]
10. Mikaelyan A L, Ter-Mikaelyan M L *Uch. Zap. EGU, Ser. Fiz.* **2** 1 (1966); Mikaelyan A L, Ter-Mikaelyan M L, in *Prog. Opt.* (Ed. E Wolf) (London: North Holland, 1969) **VII** p. 1
11. Mikaelyan A L, Ter-Mikaelyan M L, Turkov Yu G *Opticheskie Generatory na Tverdom Tele* (Solid-State Optical Generators) (Moscow: Sovetskoe Radio, 1967)

12. Feynman R, Vernon F, Hellwarth R *J. Appl. Phys.* **28** 49 (1957)
13. Allen L, Eberly J *Optical Resonance and Two-Level Atoms* (New York: Wiley, 1975) [Translated into Russian (Moscow: Mir, 1978)]
14. Delone N B, Krainov V P *Atom v Sil'nom Svetovom Pole* (Atom in a Strong Light Field) (Moscow: Energoatomizdat, 1984) [Translated into English (Berlin, New York: Springer-Verlag, 1985)]
15. Hocker G B, Tang C L *Phys. Rev. Lett.* **21** 591 (1968)
16. Bassini M et al. *Opt. Commun.* **21** 263 (1977)
17. Kramers H A, Heisenberg W *Z. Phys.* **31** 681 (1925)
18. Cohen-Tannoudji C *Optical Pumping and Interaction of Atoms with the Electromagnetic Field* (Ecole d'ate de physique theorique de Cargese, France, 1967)
19. Grischkowsky D *Phys. Rev. A* **14** 802 (1976)
20. (a) Ritus V I *Zh. Eksp. Teor. Fiz.* **51** 1544 (1966) [*Sov. Phys. JETP* **24** 104 (1967)];
(b) Nikishov A I, Ritus V I *Zh. Eksp. Teor. Fiz.* **46** 776 (1964) [*Sov. Phys. JETP* **19** 529 (1964)]
21. Zel'dovich Ya B *Usp. Fiz. Nauk* **110** 139 (1973) [*Sov. Phys. Usp.* **16** 427 (1973)]; *Zh. Eksp. Teor. Fiz.* **51** 1492 (1966) [*Sov. Phys. JETP* **24** 1006 (1967)]
22. Melikyan A O, Ter-Mikaelyan M L *Izv. Akad. Nauk SSSR, Ser. Fiz.* **46** 1004 (1982) [*Bull. Acad. Sci. USSR. Ser. Phys.* **46** 168 (1982)]
23. Bavli R, Metiu H *Phys. Rev. Lett.* **69** 1986 (1992)
24. Kastler A *Phys. Today* **20** (9) 34 (1967); *JOSA* **47** 460 (1957)
25. Aleksandrov E B et al. *Pis'ma Zh. Eksp. Teor. Fiz.* **5** 53 (1966) [*JETP Lett.* **3** 53 (1966)]
26. Bonch-Bruevich A M, Khodovoi V A, Chigir' N A *Zh. Eksp. Teor. Fiz.* **67** 2069 (1974) [*Sov. Phys. JETP* **40** 1027 (1975)]
27. Arutyunyan V M et al. *Zh. Eksp. Teor. Fiz.* **60** 62 (1971) [*Sov. Phys. JETP* **33** 34 (1971)]
28. Kirin Yu M et al. *Pis'ma Zh. Eksp. Teor. Fiz.* **9** 7 (1969) [*JETP Lett.* **9** 3 (1969)]
29. Autler S H, Townes C H *Phys. Rev.* **100** 703 (1955)
30. (a) Schabert A, Keil R, Toschek P *Opt. Commun.* **13** 265 (1975)
(b) Bradley D J, Gale G M, Smith P D *J. Phys. B* **3** 11 (1970)
31. Liao P F, Bjorkholm J E *Phys. Rev. Lett.* **34** 1540 (1975)
32. Liao P F, Bjorkholm J E *Opt. Commun.* **16** 392 (1976)
33. Treacy E, De-Maria A J *Phys. Lett. A* **29** 369 (1969)
34. Loy M M T *Phys. Rev. Lett.* **32** 814 (1974); **41** 473 (1978)
35. Shoemaker R L, Brewer R G *Phys. Rev. Lett.* **28** 1430 (1972)
36. Carlsten J L, Szoke A, Raymer M G *Phys. Rev. A* **15** 1029 (1977)
37. Weisskopf V *Z. Phys.* **85** 451 (1933)
38. Badalyan N N, Iradyan V A, Movsesyan M E *Pis'ma Zh. Eksp. Teor. Fiz.* **8** 518 (1968) [*JETP Lett.* **8** 316 (1968)]
39. (a) Kryzhanovsky B V, Melikyan A O *Opt. Commun.* **29** 164 (1979); Kryzhanovskiĭ B V, Melikyan A O *Kvantovaya Elektron.* **11** 126 (1984) [*Sov. J. Quantum Electron.* **14** 81 (1984)]; Kryzhanovskiĭ B V, Melikyan A O *Zh. Eksp. Teor. Fiz.* **79** 2063 (1980) [*Sov. Phys. JETP* **52** 2000 (1980)];
(b) Kryuchkov G Yu et al. *Zh. Eksp. Teor. Fiz.* **88** 30 (1985) [*Sov. Phys. JETP* **61** 17 (1985)]
(c) Ter-Mikaelyan M L *Izv. Akad. Nauk Armenii, Ser. Fiz.* **33** 2 (1997); Ter-Mikaelian M L *J. Contemp. Phys. Armenia* **33** 15 (1997)
40. Mollow B R *Phys. Rev.* **188** 1969, 1976 (1969); *Phys. Rev. A* **12** 1919 (1975)
41. Aleksandrov E B, Bonch-Bruevich A M, Khodovoĭ V A, Chigir' N A *Pis'ma Zh. Eksp. Teor. Fiz.* **18** 102 (1973) [*JETP Lett.* **18** 58 (1973)]
42. Apanasevich P A *Dokl. Akad. Nauk BSSR* **12** 878 (1968); *Opt. Spektrosk.* **16** 709 (1964) [*Opt. Spectrosc.* **16** 387 (1964)]
43. (a) Wu F Y et al. *Phys. Rev. Lett.* **38** 1077 (1977)
(b) Mollow B R *Phys. Rev. A* **5** 2217 (1972)
44. Grove R E, Wu F Y, Ezekiel S *Phys. Rev. A* **15** 227 (1977)
45. Wu F Y, Grove R E, Ezekiel S *Phys. Lett.* **35** 1426 (1975)
46. Shuda F, Stroud C R, Hercher M *J. Phys. B* **7** L 198 (1974)
47. Hartig W et al. *Z. Phys. A* **278** 205 (1976)
48. Rautian S G, Sobel'man I I *Zh. Eksp. Teor. Fiz.* **41** 456 (1961) [*Sov. Phys. JETP* **14** 328 (1962)]
49. Morozov V A *Opt. Spektrosk.* **26** 9 (1969) [*Opt. Spectrosc.* **26** 4 (1969)]
50. (a) Stroud C R *Phys. Rev. A* **3** 1044 (1971)
(b) Haroche S, Hartmann F *Phys. Rev. A* **6** 1280 (1972)
51. Cohen-Tannoudji C, Reynaud S *J. Phys. B* **10** 345, 365 (1977)
52. Aspect A et al. *Phys. Rev. Lett.* **45** 617 (1980)
53. Baklanov E V *Zh. Eksp. Teor. Fiz.* **65** 2203 (1973) [*Sov. Phys. JETP* **38** 1100 (1974)]
54. Rubin P L, Sokolovskiĭ R I *Zh. Eksp. Teor. Fiz.* **56** 362 (1969) [*Sov. Phys. JETP* **29** 200 (1969)]
55. Rautian S G *Tr. Fiz. Inst. Akad. Nauk SSSR* **43** 3 (1968)
56. (a) Mandel L *Opt. Lett.* **4** 205 (1979); *JOSA* **69** 1038 (1979)
(b) Carmichael H, Walls D *J. Phys. B* **9** L 43 (1976)
(c) Kimble H, Mandel L *Phys. Rev. A* **13** 2123 (1976); **15** 689 (1977)
57. Huang C, Mandel L *Phys. Rev. A* **18** 644 (1978)
58. Dagenais M, Mandel L *Phys. Rev. A* **18** 2217 (1978)
59. (a) Kimble H, Dagenais M, Mandel L *Phys. Rev. A* **18** 201 (1978)
(b) Diedrich F, Walther H *Phys. Rev. Lett.* **58** 203 (1987)
(c) Short R, Mandel L *Phys. Rev. Lett.* **51** 384 (1983)
(d) Schubert M et al. *Phys. Rev. Lett.* **68** 3016 (1992)
60. (a) Kryzhanovskiĭ B V, Melikyan A O *Kvantovaya Elektron.* **11** 126 (1984) [*Sov. J. Quantum Electron.* **14** 81 (1984)]
(b) Kryuchkov G Yu, Ter-Mikaelyan M L *Dokl. Akad. Nauk SSSR* **286** (3) 621 (1986) [*Sov. Phys. Dokl.* **31** 64 (1986)]
(c) Apanasevich P A, Kilin S Ja *J. Phys. B* **12** L83 (1979)
61. Goreslavskiĭ S P, Krainov V P *Opt. Spektrosk.* **47** 825 (1979) [*Opt. Spectrosc.* **47** 457 (1979)]
62. (a) Bonch-Bruevich A M, Vartanyan T A, Chigir' N A *Zh. Eksp. Teor. Fiz.* **77** 1899 (1979) [*Sov. Phys. JETP* **50** 901 (1979)]
63. (a) Manson N B, Wei C, Martin J P D *Phys. Rev. Lett.* **76** 3943 (1996)
(b) Manykin E A et al. *Pis'ma Zh. Eksp. Teor. Fiz.* **56** (2) 74 (1992) [*JETP Lett.* **56** 74 (1992)]
64. Melikyan A O *Dokl. Akad. Nauk Arm. SSR* **51** 214 (1970)
65. Goreslavskiĭ S P, Yakovlev V P *Izv. Akad. Nauk SSSR, Ser. Fiz.* **37** 2211 (1973) [*Bull. Acad. Sci. USSR, Ser. Phys.*]
66. Rosen N, Zener C *Phys. Rev.* **40** 502 (1932)
67. Bambini A, Berman P R *Phys. Rev. A* **23** 2496 (1981)
68. Carroll C E, Hioe F T *J. Phys. A: Math. Gen.* **19** 3579 (1986)
69. (a) Unanyan R *Zh. Eksp. Teor. Fiz.* **101** 1463 (1992) [*Sov. Phys. JETP* **74** 781 (1992)]
(b) Kelner S R et al. *Laser Phys.* **3** 623 (1993)
70. Rapoport L P, Zon B A, Manakov N L *Teoriya Mnogofotonnykh Protsessov v Atomakh* (Theory of Multiphoton Processes in Atoms) (Moscow: Atomizdat, 1978)
71. Bakaev D S et al. *Zh. Eksp. Teor. Fiz.* **83** 1297 (1982) [*Sov. Phys. JETP* **56** 743 (1982)]
72. Chang C S, Stehle P *Phys. Rev. A* **4** 641 (1971)
73. Goreslavskiĭ S P, Krainov V P *Zh. Eksp. Teor. Fiz.* **76** 26 (1979) [*Sov. Phys. JETP* **49** 13 (1979)]
74. Swain S *J. Phys. A* (GB) **5** 1587 (1972)
75. Guccione-Gush R *Phys. Rev. A* **10** 1474 (1974)
76. Proznits D, Wildman D W, George E V *Phys. Rev. A* **16** 1165 (1977)
77. (a) Agarwal G S, Nayak N *JOSA B* **1** 164 (1984)
(b) Agarwal G S et al. *JOSA B* **8** 1163 (1991)
(c) Jakov M et al. *Phys. Rev. Lett.* **78** 1 (1997)
(d) Stalgies Y et al. *Europhys. Lett.* **35** (4) 259 (1996)
78. (a) Kazantsev A P *Usp. Fiz. Nauk* **124** 113 (1978) [*Phys. Usp.* **21** 58 (1978)]
(b) Balykin V I, Letokhov V S *Phys. Today* **42** (4) 23 (1989)
79. Przhibel'skiĭ S G *Opt. Spektrosk.* **35** 715 (1973); **42** 17 (1977) [*Opt. Spectrosc.* **35** 415 (1973); **42** 8 (1977)]
80. Kryuchkov G Yu *Zh. Eksp. Teor. Fiz.* **83** 1992 (1982) [*Sov. Phys. JETP* **56** 1153 (1982)]
81. Melikyan A O *Zh. Eksp. Teor. Fiz.* **68** 1228 (1975) [*Sov. Phys. JETP* **41** 610 (1975)]
82. (a) Melikyan A O, Saakyan S G *Dokl. Akad. Nauk Arm SSR* **59** 241 (1974)
(b) Krainov V P, Mulyukov Z S *Laser Phys.* **4** 544 (1994)
83. Ter-Mikaelyan M L *Izv. Akad. Nauk Armenii, Ser. Fiz.* **32** 125 (1997); Ter-Mikaelyan M L *J. Contemp. Phys. Armenia* **32** (3) 5 (1997)
84. Kovarskiĭ V A et al. *Neadiabaticheskie Perekhody v Sil'nom Elektromagnitnom Pole* (Nonadiabatic Transitions in Strong Electromagnetic Field) (Kishinev: Shtiintsa, 1980)
85. (a) Kovarskiĭ V A, Perel'man N F, Averbukh I Sh *Mnogokvantovye Protsessy* (Multiquantum Processes) (Moscow: Energoatomizdat, 1985)

(b) Dykhne A M *Zh. Eksp. Teor. Fiz.* **41** 1324 (1961) [*Sov. Phys. JETP* **14** 941 (1962)]
86. Born M, Fock V *Z. Phys.* **51** 165 (1928)
87. Melikyan A O, Saakyan S P Preprint IFI Akad. Nauk Arm. No. 76-16 (Erevan: IFI, 1977)
88. Ter-Mikaelyan M L, Grigoryan G G *Laser Phys.* **5** 1171 (1995); *Opt. Commun.* **119** 56 (1995)
89. Apanasevich P A *Osnovy Teorii Vzaimodeistviya Sveta s Veshchestvom* (Fundamentals of Theory of Light-Matter Interaction) (Minsk: Nauka i Tekhnika, 1977)
90. Bakhramov A S, Tartakovskiĭ R Kh, Khabibulaev P K *Nelineĭnye Rezonansnye Protsessy i Preobrazovaniya Chastoty v Gazakh* (Nonlinear Resonant Processes and Frequency Transformations in Gases) (Tashkent: Fan, 1981)
91. Aleksandrov E B, Khvostenko G I, Chaika M P *Interferentsiya Atomnykh Sostoyaniĭ* (Interference of Atomic States) (Moscow: Nauka, 1991) [Translated into English (Berlin: Springer-Verlag, 1988)]
92. (a) Kilin S Ya *Kvantovaya Optika, Polya i ikh Detektirovanie* (Quantum Optics, Fields, and their Detection) (Minsk: Nauka i Tekhnika, 1990) [Translated into English (New York: Gordon Breach, 1988)]
(b) Klyshko D N *Fotony i Nelineinaya Optika* (Photons and Nonlinear Optics) (Moscow: Nauka, 1980)
93. (a) Letokhov V S, Chebotaev V P *Printsipy Nelineĭnoĭ Lasernoĭ Spektroskopii* (Principles of Nonlinear Laser Spectroscopy) (Moscow: Nauka, 1975)
(b) Rautian S G, Smirnov G I, Shalagin A N *Nelineinye Rezonansy v Spektrakh Atomov i Molekul* (Nonlinear Resonances in Atomic and Molecular Spectra) (Novosibirsk: Nauka, 1979)
(c) Popov A K *Vvedenie v Nelineinuyu Spektroskopiyu* (Introduction to Nonlinear Spectroscopy) (Novosibirsk: Nauka, 1979)
(d) Manykin E A, Samartsev V V *Opticheskaya Ekho-Spektroskopiya* (Optical Echo Spectroscopy) (Moscow: Nauka, 1984)
(e) Shore B W *The Theory of Coherent Atomic Excitation* (New York: Wiley, 1990)
(f) Cohen-Tannoudji C, Dupon-Roc J, Grynberg G *Atom-Photon Interaction* (New York: Wiley, 1992)
94. Shen I R *Printsipy Nelineĭnoĭ Optiki* (Principles of Nonlinear Optics) (Moscow: Nauka, 1989)
95. Bloembergen N *Usp. Fiz. Nauk* **97** 307 (1969) [*Amer J. Phys.* **35** (11) 989 (1967)]; *Nonlinear Optics. A Lecture Note* (New York: Wiley, 1965) [Translated into Russian (Moscow: Mir, 1969)]
96. Akhmanov S A, Khokhlov R V *Problemy Nelineĭnoĭ Optiki* (Problems in Nonlinear Optics) (Moscow: Inst. Nauch. Inform., 1964)
97. (a) Butylkin V S et al. *Rezonansnye Vzaimodeĭstviya Sveta s Veshchestvom* (Resonant Light-Matter Interaction) (Moscow: Nauka, 1977)
(b) Akulin V M, Karlov N V *Intensivnye Rezonansnye Vzaimodeĭstviya v Kvantovoĭ Elektronike* (Intensive Resonant Interactions in Quantum Electronics) (Moscow: Nauka, 1987)
(c) Koroteev N I *Usp. Fiz. Nauk* **152** 493 (1987) [*Sov. Phys. Usp.* **30** 628 (1987)]
(d) Akhmanov S A, Vysloukh V A, Chirkin A S *Optika Femtosekundnykh Impul'sov* (Optics of Femtosecond Pulses) (Moscow: Nauka, 1988) [Translated into English (New York: AIP, 1992)]
98. Pantell R, Puthoff H *Fundamentals of Quantum Electronics* (New York: Plenum Press, 1969) [Translated into Russian (Moscow: Mir, 1972)]
99. Basov N G, Prokhorov A M *Zh. Eksp. Teor. Fiz.* **28** 249 (1955) [*Sov. Phys. JETP* **1** 184 (1955)]
100. (a) Javan A, in *Proc. Int. School of Physics Enrico Fermi Course XXX** (New York: Plenum, 1964) p. 284; *Phys. Rev.* **107** 1579 (1957)
(b) Hänsch T, Toschek P E *Z. Phys.* **236** 213 (1970)
(c) Beterov I M, Chebotaev V P *Pis'ma Zh. Eksp. Teor. Fiz.* **9** 216 (1969) [*JETP Lett.* **9** 127 (1969)]
101. Ter-Mikaelyan M L, Sarkisyan M A Preprint IFI Akad. Nauk Arm. No. 75-26 (Erevan: IFI, 1975)
102. Glushko B A, Chaltykyan V O Preprint IFI Akad. Nauk Arm. No. 75-25 (Erevan: IFI, 1975); *Izv. Akad. Nauk Arm. SSR, Ser. Fiz.* **13** 260 (1978)
103. (a) Sarkisyan M A, Ter-Mikaelyan M L *Izv. Akad. Nauk SSSR, Ser. Fiz.* **42** 2574 (1978) [*Bull. Acad. Sci. USSR, Ser. Phys.* **42** 93 (1978)]
(b) Band Y B, Magnes O *Phys. Rev. A* **50** 584 (1994)
(c) Broers B et al. *Phys. Rev. Lett.* **69** 2062 (1992)
104. Grishckowsky D, Loy M M T *Phys. Rev. A* **12** 1117, 2514 (1975)
105. Sarkisyan M A *Dokl. Akad. Nauk Arm. SSR* **65** (1) 23 (1977)
106. Kroll N M, Watson K M *Phys. Rev. A* **13** 1018 (1976)
107. Lau A M F *Phys. Rev. A* **13** 139 (1976)
108. Lau A M F *Phys. Rev. A* **14** 279 (1976)
109. Zon B A *Zh. Eksp. Teor. Fiz.* **75** 834 (1978) [*Sov. Phys. JETP* **48** 422 (1978)]
110. (a) Fedorov M V *Zh. Eksp. Teor. Fiz.* **77** 2200 (1979) [*Sov. Phys. JETP* **50** 1056 (1979)]
(b) Gazazyan A D, Sarkisyan M A, Preprint IFI Akad. Nauk Arm. SSR No. 80-93 (Ashtarak: 1980)
111. Gazazian A D, Sarkissian M A *Phys. Lett. A* **78** 133 (1980)
112. Zon B A, Katsnel'son B G *Zh. Eksp. Teor. Fiz.* **80** 474 (1981) [*Sov. Phys. JETP* **53** 241 (1981)]
113. Kuz'min M V, Sazonov V N *Zh. Eksp. Teor. Fiz.* **79** 1759 (1980) [*Sov. Phys. JETP* **52** 889 (1980)]
114. Kuz'min M V, Sazonov V N *Zh. Eksp. Teor. Fiz.* **83** 50 (1982) [*Sov. Phys. JETP* **56** 27 (1982)]
115. Kuz'min M V *Kvantovaya Elektron.* **8** 20 (1981) [*Sov. J. Quantum Electron.* **11** 9 (1981)]
116. Melikyan A O, Saakyan S G, Sarkisyan M A *Kvantovaya Elektron.* **12** 1446 (1985) [*Sov. J. Quantum Electron.* **15** 955 (1985)]
117. Ter-Mikaelian M L, Sarkissian M A, in *Proc. 6th Int. Conf. on Atomic Physics*: *Riga, USSR, 17-22 Aug. 1978* (New York: Plenum Press, 1979) p. 462; Ter-Mikaelyan M L, Sarkisyan M A, Preprint IFI Akad. Nauk Arm. SSR No. 78-75 (Erevan: IFI, 1978)
118. Melikyan A O, Saakyan S G *Zh. Eksp. Teor. Fiz.* **76** 1530 (1979) [*Sov. Phys. JETP* **49** 776 (1979)]
119. Grigoryan G G, Ter-Mikaelyan M L, Melikdzhanov D Yu *Opt. Spektrosk.* **67** 998 (1989)
120. Belenov E M, Poluéktov I A *Zh. Eksp. Teor. Fiz.* **56** 1407 (1969) [*Sov. Phys. JETP* **29** 754 (1969)]
121. (a) Loy M M T *Phys. Rev. Lett.* **36** 1454 (1976)
(b) Manykin E A, Afanas'ev A M *Zh. Eksp. Teor. Fiz.* **48** 931 (1965) [*Sov. Phys. JETP* **21** 619 (1965)]
(c) Manykin E A, in *Prokhozhdenie Izlucheniya cherez Veshchestvo* (Radiation Passage through Matter) (Moscow: Atomizdat, 1968), p. 72
(d) Manykin E A, Afanas'ev A M *Zh. Eksp. Teor. Fiz.* **52** 1246 (1967) [*Sov. Phys. JETP* **25** 828 (1967)]
(e) Chen Ce, Yi Yian Yin, Elliott D S *Phys. Rev. Lett.* **64** 507 (1990)
122. (a) Vasilenko L S, Chebotaev V P, Shishaev A V *Pis'ma Zh. Eksp. Teor. Fiz.* **12** 161 (1970) [*JETP Lett.* **12** 113 (1970)]
(b) Beterov I M, Chebotaev V P *Progr. Quantum Electronics* **3** 1 (1974)
123. Biraben F, Cagnac B, Grynberg G *Phys. Rev. Lett.* **32** 643 (1974)
124. Levenson M D, Bloembergen N *Phys. Rev. Lett.* **32** 645 (1974)
125. Hänsch T W et al. *Opt. Commun.* **11** 50 (1974)
126. Bjorkholm J E, Liao P F *Phys. Rev. Lett.* **33** 128 (1974)
127. Bjorkholm J E , Liao P F *Phys. Rev. A* **14** 751 (1976)
128. Bjorkholm J E, Liao P F *Phys. Rev. Lett.* **36** 1543 (1976); *Opt. Commun.* **21** 132 (1977)
129. Hänsch T W et al. *Phys. Rev. Lett.* **34** 307 (1975)
130. Lee S A, Wallenstein R, Hänsch T W *Phys. Rev. Lett.* **35** 1262 (1975)
131. Grischkowsky D, Loy M M T, Liao P F *Phys. Rev. A* **12** 2514 (1975)
132. Sarkissian M A, in *Proc. 2nd Int. Conf. on Trends in Quant. Electr.* (Bucharest, 1985); *Rev. Roum. Phys.* **32** 189 (1987)
133. Anikin V I et al. *Kvantovaya Elektron.* **3** 330 (1976) [*Sov. J. Quantum Electron.* **6** 174 (1976)]
134. Movsesyan M E, Badalyan N N, Iradyan V A *Pis'ma Zh. Eksp. Teor. Fiz.* **6** 631 (1967) [*JETP Lett.* **6** 127 (1967)]
135. Rokni M, Yatsiv S *Phys. Lett. A* **24** 277 (1967)
136. (a) Sorokin P P et al. *Appl. Phys. Lett.* **10** 44 (1967)
(b) Gray H R, Whitley R M, Stroud C R *Opt. Lett.* **3** 218 (1978)
(c) Gray H R, Stroud C R *Opt. Commun.* **25** 359 (1978)
(d) Sapondzhyan S O, Sarkisyan D G *Kvantovaya Elektron.* **10** 1614 (1983) [*Sov. J. Quantum Electron.* **13** 1062 (1983)]
(e) Sapondzhyan S O, Sarkisyan D G, Torosyan G A *Kvantovaya Elektron.* **12** 963 (1985) [*Sov. J. Quantum Electron.* **15** 634 (1985)]

137. Ter-Mikaelyan M L Preprint IFI Akad. Nauk Arm. SSR No. 82-102 (Erevan: IFI, 1982); Ter-Mikaelyan M L, in *Proc. Int. Conf. and School 'Lasers and Applications'* (Bucharest, 1982) p. 453
138. Kryzhanovskiĭ B V *Opt. Spektrosk.* **55** 1024 (1983) [*Opt. Spectrosc.* **55** 621 (1983)]
139. Kryzhanovskiĭ B V, Sulkhanyan R M, Ter-Mikaelyan M L *Izv. Akad. Nauk Arm. SSR* **18** 83 (1983)
140. Kryzhanovskiĭ B V Melikyan A O *Kvantovaya Elektron.* **11** 126 (1984) [*Sov. J. Quantum Electron.* **14** 81 (1984)]
141. Kryzhanovskiĭ B V Melikyan A O *Kvantovaya Elektron.* **13** 734 (1986) [*Sov. J. Quantum Electron.* **16** 477 (1986)]
142. Ter-Mikaelian M L, Movsesyan R E *Kombinatsionnoe Rasseyanie* (Raman Scattering) Preprint IFI Akad. Nauk Arm. SSR No. 23-75 (Erevan: IFI, 1975)
143. Oreg J, Hioe F T, Eberly J H *Phys. Rev. A* **29** 690 (1984)
144. (a) Goubatz U et al. *Chem. Phys. Lett.* **149** 463 (1988)
(b) Kuklinski J R et al. *Phys. Rev. A* **40** 6741 (1989)
145. (a) Schiemann S et al. *Phys. Rev. Lett.* **71** 3637 (1993)
(b) Glushko B, Kryzhanovsky B *Phys. Rev. A* **46** 2823 (1992)
(c) Vitanov N V, Stenholm S *Phys. Rev. A* **55** 648 (1997)
146. (a) Alzetta G et al. *Nuovo Cimento B* **36** 5 (1976)
(b) Arimondo E, Orriols G *Lett. Nuovo Cimento* **17** 333 (1976)
147. Gray H R, Whitley R M, Stroud C R *Opt. Lett.* **3** 218 (1978)
148. Kocharovskaya O A, Khanin A Ya *Pis'ma Zh. Eksp. Teor. Fiz.* **48** 581 (1988) [*JETP Lett.* **48** 630 (1988)]
149. Scully M O, Zhu S Y, Gavrielides A *Phys. Rev. Lett.* **62** 2813 (1989)
150. (a) Aspect A et al. *Phys. Rev. Lett.* **61** 826 (1988)
(b) Lawall J et al. *Phys. Rev. Lett.* **73** 1915 (1994)
151. Esslinger T et al. *Phys. Rev. Lett.* **76** 2432 (1996)
152. Gazazyan A D, Unanyan R G *Zh. Eksp. Teor. Fiz.* **108** 1917 (1995) [*Phys. JETP* **81** 1047 (1995)]
153. Gaubatz U et al. *J. Chem. Phys.* **92** 5363 (1990)
154. Laine T A, Stenholm S *Phys. Rev. A* **53** 2501 (1996)
155. Bergmann K, Martin J, Shore B W, in *Coherence and Quantum Optics VII* (Eds J H Eberly, L Mandel, E Wolf) (New York: Plenum, 1995)
156. (a) Shore B W et al. *Phys. Rev. A* **52** 566, 583 (1995)
(b) Marte P, Zoller P, Hall J L *Phys. Rev. A* **44** 4118 (1991)
(c) Pillet P et al. *Phys. Rev. A* **48** 845 (1993)
157. Aspect A et al. *JOSA B* **6** 2112 (1989)
158. Agap'ev B D et al. *Usp. Fiz. Nauk* **163** (9) 1 (1993) [*Phys. Usp.* **36** 763 (1993)]
159. Ter-Mikaelyan M L, Unanyan R G *Laser Phys.* **6** 953 (1996)
160. Movsesyan R E, Chaltykyan V O *Dokl. Akad. Nauk Arm. SSR* **52** (1) 33 (1971)
161. Sellin P B et al. *Phys. Rev. Lett.* **78** 1432 (1997)
162. Itano W M et al. *Phys. Rev. A* **41** 2295 (1990)
163. Khalfin L A *Usp. Fiz. Nauk* **160** (10) 185 (1990) [*Sov. Phys. Usp.* **36** 868 (1990)]
164. Hoeffs J T et al. *Opt. Commun.* **133** 179 (1997)
165. Meyer G M, Loeffer M, Walther H *Phys. Rev. A* **56** 56 (1997)
166. Maitre X, Haroche S *Phys. Rev. Lett.* **79** 769 (1997)