Resonance interaction of intense light with atoms

N. B. Delone and V. P. Kraĭnov

P. N. Lebedev Physics Institute, Academy of Sciences of the USSR
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Some typical resonance nonlinear-optics phenomena taking place when light interacts with atoms are considered. The multitude of such phenomena reduces to scattering of the perturbing light, perturbation of the atomic spectrum, excitation of an atom and its ionization. In all these cases the investigations are carried out using the simplest model—an isolated atom in a monochromatic external field. Such a model enables one to give an exact description of many elementary processes. On the other hand it is sufficiently realistic. A detailed investigation is carried out of resonance fluorescence, spontaneous Raman scattering, multiphoton excitation of atoms, resonance shifts and splitting of atomic levels and resonance ionication of atoms. The nature of these processes is investigated as a function of the intensity of the external field, its frequency, a specific atomic spectrum, the nature of polarization of the perturbing field, and the degree of its nonlinearity. The role played by the field of the electromagnetic vacuum is analyzed. Theoretical predictions are compared with the results of many experiments describing resonance processes of the interaction of an intense light field with atoms.

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CONTENTS

1. INTRODUCTION

In this review we consider the overall picture of resonance nonlinear-optics phenomena occurring when light interacts with atoms.

Throughout we shall utilize the simplest model—an isolated atom in a monochromatic classical external field. Such a model, on the one hand, enables us to give an exact description of many elementary processes, and, on the other hand, is a sufficiently realistic one. At the present time it is possible in practice to obtain at any frequency in the visible range sufficiently intense radiation with spectral width of the order of the natural width of atomic levels or even with a smaller width. The use of the method of intersecting beams—a light beam and an atomic beam—makes it possible not to have to take the Doppler effect into account. The high intensity of laser radiation, and also the high sensitivity of phonon and ion detectors enable one to utilize sufficiently rarified targets and thereby to ignore secondary processes. One should also keep in mind that these particular ideal conditions are optimal ones for a wide range of practically important problems associated with selective action of intense laser radiation on an atomic medium.

First of all we specify more precisely the content which we ascribe to the concept of resonance interaction.

When reference is made to a resonance between the frequency of an external field ω (or of its higher harmonics) and the transition frequency ω_{mn} between definite bound states of an electron in an atom $E_m^{(0)}$ and $E_n^{(0)}$, then it is always assumed that these frequencies are close to each other. In the simplest case of a phonon resonance with which we will deal at the outset the condition of closeness takes the form $\omega \approx \omega_{mn} = E_m^{(0)} - E_n^{(0)}$, where $E_{m,n}^{(0)}$ are energies of bound states in an unperturbed spectrum,[1)] It is well known in practice that the actual condition for the observation of resonance phenomena is one imposed on the magnitude of the deviation from resonance $\Delta = \omega_{mn} - \omega \leqslant \gamma_{m,n}$, where $\gamma_{m,n}$ is the reduced width of the resonance. In what follows we also shall be basically interested in small deviations from resonance or, as is often said, in "exact resonance", the condition for the attainment of which will differ from the one stated above only by the presence of the dependence of the resonance on the intensity of the external field. Such an approach follows naturally from the assumption of high intensity of the external field. However, in actual fact the attainment of resonance is associated with the selection from among the finite number of bound electron states $E_i^{(0)}$ of two specific ones, $E_{m,n}^{(0)}$, and therefore the rigorous definition of the resonance process turns out to be much less restrictive:

$$\Delta = \omega_{mn} - \omega \ll \omega_{mn}. \tag{1}$$

This condition is well known from the old, "prelaser," physics. However in cases of interest to us when we are dealing with processes occurring in a strong light field it is necessary to take into account the perturbation of the atomic spectrum by the external field, so that condition (1) turns out to be a necessary but not a sufficient one. It must be complemented by the con-

[1)] We shall everywhere use the system of units $e = \hbar = m_e = 1$.

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dition

$$|\mathbf{d}_{mn}\vec{\mathcal{E}}| \ll \omega_{mn}, \tag{2}$$

which is in actual fact the condition for the weakness of the perturbation of the resonance states m,n by the external field $\vec{\mathcal{E}}$ (d_{mn} is the dipole moment of the two-level system m,n). We note that the ratio $\mathbf{d}_{mn}\vec{\mathcal{E}}/\Delta$ can take on arbitrary values.

A generalization of conditions (1) and (2) to the case of multiphoton resonance is not trivial; it is carried out in section 5.

We now turn our attention to the principal parameters characterizing the light field.

First of all we note that, as is well known, if the condition $n_{k\alpha} \gg 1$ is satisfied, then the field can be described as a classical one (where $n_{k\alpha}$ is the number of quanta of a given oscillation mode, i.e., of an oscillation characterized by the propagation vector k and polarization α). The condition stated above is equivalent to the following condition on the intensity of the field of the wave: $\mathcal{E} \gg (\omega^3 \Delta\omega/c^3)^{1/2}$ which in the visible range gives a numerical estimate of $\mathcal{E} \gg 1$ V/cm. Obviously this condition is in fact satisfied in all cases of interest to us. Therefore in what follows the field will everywhere be described classically, and the interaction of light with atoms (in the dipole approximation which we will utilize in the greater part of this review) has the form $V = \mathbf{d}\vec{\mathcal{E}}\cos\omega t$. The terms "quantum" or "photon" will be utilized either to preserve the traditional terminology, or to characterize the frequency of the classical field in language usual in the physics of an atom which is, as is well known, in principle a quantum object.

One of the basic parameters appearing in problems of interest to us is the time T during which the external field $\vec{\mathcal{E}}$ acts on an atoms. It is obvious that the characteristic time with which it makes sense to compare the time of action T is the lifetime of an atom τ in a particular state. This can be either the natural lifetime, or a lifetime determined by an induced transition of an electron from this state. If for the sake of definiteness we refer to natural lifetimes, then their values are $\tau_i \gtrsim 10^{-8}$ sec, while the magnitudes of the level widths are respectively $\gamma_i \lesssim 10^{-3}$ cm^{-1}. It is obvious that in the case of induced transitions the lifetimes can be smaller by many orders of magnitude. When lasers are used as sources of an intense light field, then, as a rule, we are dealing with a so-called regime of modulation of the resonator factor[2)] when the laser is operating in a pulsed regime emitting (in the case of a single mode generation regime) a smooth bellshaped pulse with a front duration of the order of the half-width $T \approx 10^{-8}$ sec. But in certain cases one could also utilize radiation from lasers operating in the regime of continuous generation. Thus, if one speaks of spontaneous transitions, then typical experimental conditions correspond to the case $\gamma_i T \gtrsim 1$.

All the conditions for bringing about resonance interaction which were discussed above were stated under the assumption of a monochromatic external field. This assumption is well realised in practice since in realising a single-mode (i.e., one with three fixed indexes) generation regime of the laser one can have a very narrow spectrum of emitted light, so that $\Delta\omega/\omega \sim 10^{-8}$, while in the worst case of a multimode generation regime of an active medium characterized by a broad luminescence line one has $\Delta\omega/\omega \sim 10^{-4}$. It is also necessary to keep in mind that the monochromatic nature of laser radiation in the case of a single-mode generation regime is closely associated with the duration T of the radiation pulse: $\Delta\omega = 1/T$ (in a multimode generation regime one always has $\Delta\omega \gg 1/T$). In pulses of nanosecond duration $\Delta\omega \gg \gamma_i$, where γ_i is a typical natural width of atomic levels. In pulses of picosecond duration, which are obtained from lasers in the regime of synchronized phases of the modes, $\Delta\omega$ attains values of $\sim 10^1 - 10^2$ cm^{-1}.

We now turn to the intensity of the light field and specify the terms "intense light" and intense light field". It is quite natural that these terms are defined both by the field and by the atom and are characterized by the interaction of the field with the atom. We shall use these terms when the interaction exceeds the natural width of the resonance determined by spontaneous decay. Applied to the single-photon resonance in a two-levels system which we discussed above the criterion for an intense external field $\vec{\mathcal{E}}$ has the obvious form

$$|\mathbf{d}_{mn}\vec{\mathcal{E}}| \gtrsim \gamma_{m,n}. \tag{3}$$

We give a numerical estimate assuming $d_{mn} \approx 1$ Debye, while $\gamma_{m,n} \sim 10^8$ sec We obtain the condition $\mathcal{E} \gtrsim 50-100$ V/cm. We note that this is a lower limit, since the estimate of d_{mn} is an upper estimate, and moreover the resonance is assumed to be exact. In all other cases, including the case of multiphoton resonance, the critical value of the intensity of the field exceeds the value indicated above by several orders of magnitude. The corresponding criterion is given in Section 5. Comparing numerical estimates of critical intensities for an intense field and for a classical field it can be seen that the intense field of interest to us is always a classical one.

In the discussion carried out above we have restricted the range of field intensities also from above by the sufficiently obvious condition

$$\mathcal{E} \ll \mathcal{E}_{at}, \tag{4}$$

where $\mathcal{E}_{at} = 5 \times 10^9$ V/cm is the atomic field intensity. From a theoretical point of view this enables us always to assume that the ratio $\mathcal{E}/\mathcal{E}_{at}$ is a small parameter. From the point of view of the physical essence of the phenomena under consideration this enables us to leave out of consideration the tunnel effect since it is well known that in the frequency range of visible light the condition stated above is equivalent to the condition $\gamma \gg 1$, where the adiabatic parameter $\gamma = \tau_{tun}/T = \omega(2E_i^{(0)})^{1/2}/\mathcal{E}$.

Finally, the intensity of the field is restricted by the condition (2) of the smallness of the perturbation com-

[2)] Such a regime is often referred to as a "giant pulse regime".

pared with the distance between the levels. Since the quantity ω_{mn} varies sharply as the principal quantum number characterizing the states m and n is varied, the criteria (2) and (4) cannot be reduced to a single one. Specifically, while for states of low excitation these criteria are close to one another, for highly excited states the criterion (2) turns out to be the more restrictive one.

From the point of view of the physics of the interaction of an intense field with an atom the specific situation consists of the perturbation of the spectrum of bound electron states and of the occurrence of multiphoton transitions with a probability comparable to the probability of single-photon transitions. From the theoretical point of view the specific situation consists of the inapplicability of perturbation theory of the first order with respect to $\mathcal{E}$ to the description of the effects taking place; in the language of Feynman diagrams this means that it is necessary to take into account diagrams of second and higher orders in $\mathcal{E}$. From a practical point of view an intense field is characterized by the nonlinear nature of the observed phenomena with respect to the intensity of the exciting light.

The critical intensity of the field, which in the optimal case must be regarded as intense, is quite small not only in comparison with the atomic field intensity, but even with the intensity of the field of laser radiation and is at the upper limit of intensities which can be realised utilizing noncoherent light sources. However, detailed experimental data on elementary resonance nonlinear-optics phenomena were obtained only in recent years. The principal difficulty consisted of carrying out experiments with a spectral resolution better than the natural line width. Two circumstances enable one to carry out such experiments. The first is the use of lasers operating in the regime of single-mode generation with a spectral width of the order of 10^{-4} cm^{-1} and with an absolute generation frequency variable over wide limits. The second is the use as targets of atomic beams of such low divergence that in the case of transverse propagation of laser radiation the Doppler effect does not exceed the value indicated above. The choice of a specific transition in an atomic spectrum also presents a certain difficulty, since the contribution of third levels, i.e., of nonresonance interaction, must be small. At the same time it is evident that the fine structure of the spectrum must also be taken into account.

We now turn to the general methods of solving the problems indicated above. The theoretical description of resonance phenomena is based on using the resonance approximation.[3)1,2] The essence of this method consists of an approximate solution of the system of quantum mechanical equations for the atom which is acted upon by a monochromatic light perturbation $V \sim \cos \omega t$: in this perturbation only the exponential $e^{-i\omega t}$ (or $e^{i\omega t}$) which leads to a small energy denominator in the solution is taken into account and the exponent for which the denominator is not anomalously small is neglected. The resonance approximation is correct when conditions (1) and (2) are satisfied.

We now discuss which quantum mechanical equations must be solved. In the most general case, when the complete Hamiltonian includes the atomic Hamiltonian $\hat{\mathcal{H}}_0$, the intense classical external field V and the quantum field of the electromagnetic vacuum, the problem of finding the complete wave function is a very complicated one. Indeed, in intense fields during times of their action T which are not too small the condition $\gamma T \gg 1$ is satisfied, i.e., the number of spontaneously emitted quanta $\sim \gamma T$ turns out to be quite large. However, complete information containing all that needs to be known concerning the photons mentioned above is not at present available experimentally; only the probabilities of emitting one spontaneous quantum of a definite frequency are measured. From the mathematical point of view this means that the necessity does not arise to obtain the complete wave function, but one needs only average values of the probabilities over the photon variables.

As is well known,[5] such an approach is realized by introducing the atomic density matrix ρ. The equations for it have the form

$$\frac{\partial \rho}{\partial t} = i\,[\rho, \hat{\mathcal{H}}] - \hat{\gamma}\rho, \tag{5}$$

where $\hat{\mathcal{H}} = \hat{\mathcal{H}}_0 + V$, while the matrix $\hat{\gamma}$ describes the relaxation of the elements of the density matrix.[4)]

Without reference to a specific phenomenon one can note the following general character of the solutions of equation (5) in different time regimes.

For times $T \ll 1/\gamma$ radiation damping is unimportant. In this case the elements of the density matrix break up into products of amplitudes: $\rho_{ij} = a_i^* a_j$. The equations for these amplitudes are the usual Schrödinger equations with the Hamiltonian $\hat{\mathcal{H}} = \hat{\mathcal{H}}_0 + V$. In the resonance approximation they reduce to a system of equations with constant coefficients. The solutions of this system of first order differential equations are in principle obtained by a direct method. They represent a set of states orthogonal to each other which are referred to as quasienergy or "dressed states".[3,6] The corresponding atomic system in resonance external fields is often referred to as a "dressed atom". One of the formulations of such problems consists of transitions between quasienergy states under the action either of a weak test field or of the electromagnetic vacuum field, etc.

Each of the quasienergy states represents a combination of stationary states. But the energies of these states differ from the energies of the initial atomic states unperturbed by the field. They are called "quasienergies".[6] From the Floquet[7] theorem it follows that the energies of the stationary states, a combination of which makes up the fixed quasienergy state, differ from each other by integral multiples of ω.

[3)] In foreign literature the equivalent term "rotating wave approximation" is utilized. [3] The "method of averaging" has the same meaning.[4]

[4)] Equations (5) are often called the "optical Bloch equations".

We note that in order for the Floquet theorem to be applicable it is not necessary that the resonance approximation be satisfied, but only periodicity of the perturbation is required. But in the general case quasienergy states consist of a large number of stationary states and therefore their use is impractical. In the resonance approximation an essential contribution to the quasienergy states is made by a small number of stationary states and they become convenient for analytic investigations.

If we turn to times $T \sim 1/\gamma$, then a transitional regime occurs which is characterized by a very complicated form of solutions.

Finally for $T \gg 1/\gamma$ a stationary regime occurs which is characterized by constant occupation numbers of atomic levels. A distinguishing feature of the stationary regime is the fact that the solutions do not depend on the initial conditions, and, in particular, on the method of switching on the external field.

Taking spontaneous effects into account in the stationary regime turns out to be considerably simpler compared to solving the system (5) when a transition is being investigated between two states between which no strong resonance field is acting. For example, we may be dealing with a three-level system in which resonance with the strong field occurs between the first two levels while spontaneous emission occurs as a result of the transition of an electron to the third level (resonance spontaneous Raman scattering; section 4). Then taking the interaction with the field of the electromagnetic vacuum into account reduces to the Breit-Wigner procedure.[8] It consists of replacing in all the formulas the level energies $E_j^{(0)}$ by the quantities $E_j^{(0)} - \frac{1}{2} i\gamma_j$. It should be noted that such a replacement is of a very general nature in the sense that the widths of the levels are not necessarily radiative widths, but represent the sum of all possible widths.

Without reference to whether the Breit-Wigner procedure is applicable or not the specific features of the strong field consist of the fact that it leads to transitions between different quasienergy states and also numerically alters the values of the spectral widths of such transitions compared to the case of a weak field. We emphasize that in strong fields these widths may be altered also in the direction of being reduced.

2. TWO-LEVEL SYSTEM

The simplest of all quantum-mechanical systems which can be used to investigate the resonance phenomenon is a two-level system. We denote by n the lower state with energy $E_n^{(0)}$, and by m the upper state with energy $E_m^{(0)}$, and by γ_m the spontaneous width of the upper level with respect to a transition to the lower state. Resonance occurs in an external field when conditions (1), (2) are satisfied.

As has been noted in the Introduction, phenomena arising in the case of resonance are of a different nature depending on the time that the field is acting. We first consider the short times when relaxation can be neglected. The solution of the problem of the behavior of

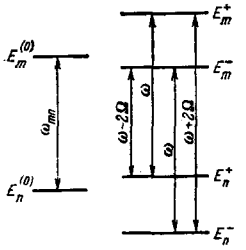

FIG. 1. Quasienergies of a two-level system m, n in a resonance field $\mathcal{E}$, ω.

a two-level system in a resonance external field is well known,[1,2] so that we shall not reproduce it here.[5)] We recall that one obtains two quasienergy states orthogonal to each other, the wave function represents a superposition of them, the coefficients of which depend on the initial conditions imposed. Since each of the quasienergy states arises from two stationary states the wave function in the general case represents a superposition of four stationary states, the quasienergies of which have the form (Fig. 1)

$$E_n^{\pm} = E_n^0 \pm \Omega + \frac{\Delta}{2}, \quad E_m^{\pm} = E_m^{(0)} \pm \Omega - \frac{\Delta}{2}. \tag{6}$$

The quantity

$$\Omega = \frac{1}{2}\sqrt{\Delta^2 + |\mathbf{d}_{mn}\vec{\mathcal{E}}|^2}, \tag{7}$$

appearing in (6) is known as the Rabi frequency.[6] In the case of an exact resonance, i.e., when $\Delta = 0$ we obtain from (7) the so-called Rabi resonance frequency $\Omega = |\mathbf{d}_{mn}\vec{\mathcal{E}}|/2$.

As can be seen from (6) twice the Rabi frequency is just the splitting of each of the levels m and n. From (3) and (7) it follows that such splitting can be observed only in a strong field. A characteristic feature of this splitting is its linear dependence on the amplitude of the field intensity $\mathcal{E}$ (of course, for not too great deviations from resonance, and specifically when $\Delta \leq \mathbf{d}_{mn}\vec{\mathcal{E}}$).

The dynamic development of the two-level system in time in the relaxationless regime indicated above ($\gamma_m T \ll 1$) depends significantly on the initial conditions. Depending on the time of growth of the amplitude of the field δT one can select two limiting cases: the instantaneous and the adiabatic switching-on of the perturbation. In the case of the adiabatic switching-on the initial unperturbed state dynamically develops into one of the quasienergy states, and for this reason no splitting is observed. In contrast to this in the case of the instantaneous switching-on superposition of both quasienergy states occurs and splitting is observable. It turns out[9] that if the relation $\Delta\delta T \ll 1$ is satisfied then the regime of switching on the external field is instantaneous, and when the inequality is reversed, it becomes adiabatic. This relation can be quantitatively understood from the point of view of the indeterminacy principle: mixing of the quasienergy states occurs when the time of mixing $1/\Delta$ is sufficiently great.

From what has been said above one can conclude that in the non-resonance case the adiabatic regime of switching-on is always realized. In the resonance case

[5)] The problem under consideration is mathematically equivalent to the problem of a particle of spin $\frac{1}{2}$ in a magnetic field.[3]

for characteristic values of deviation from resonance $\Delta \sim \gamma_m$ the instantaneous regime of switching-on is realized since $\delta T \lesssim T$, which is typical for pulsed lasers.

Until now we have been interested only in the splitting of levels. We now turn to the question of level shifts. From (7) it may be seen that in the resonance approximation the nonresonance shift[6] is absent. In actual fact such a shift does occur. Its amplitude can be calculated if in the lowest order of perturbation theory one takes into account effects arising from the exponential $e^{i\omega t}$ which does not lead to a small energy denominator. Taking nonresonance terms into account within the framework of the two-level system leads to shifts of the levels m and n which are small, quadratic in the field, equal in magnitude and opposite in direction. In this case the position of the resonance is shifted from the point $\Delta = 0$ by an amount $|\mathbf{d}_{mn}\vec{\mathcal{E}}|^2/4\omega_{mn}$, which is referred to as the Bloch-Siegert shift (cf., Refs. 10, 11). It is small since the denominator of the ratio indicated above contains the distance between the levels ω_{mn} which has a relatively large value.

Thus, when resonance occurs, nonresonance shifts of levels are small compared with the resonance splitting of the levels.

In conclusion we note that the relaxationless regime is of no practical interest since when the conditions $\gamma_m T \ll 1$ are satisfied a very high field intensity is required for the development of the quasienergy states, such that the resonance Rabi frequency would be very great:

$$\Omega = \tfrac{1}{2}|\mathbf{d}_{mn}\vec{\mathcal{E}}| \gg \frac{1}{T} \gg \gamma_m.$$

We have in mind a field intensity which is by several orders of magnitude greater than the critical intensity which follows from the inequality (3).

We now turn to the more realistic case of times T which are not small compared to $1/\gamma_m$. Then spontaneous relaxation should be taken into account. In particular, the quasienergies (6) become complex with widths of the order γ_m. If we now concern ourselves with a transition from the state m under the action of an infinitely weak test field, which does not perturb the system under consideration, to some third state (we are not interested here in its nature) then we can select two cases: in a weak field, $\mathbf{d}_{mn}\vec{\mathcal{E}} < \gamma_m$, the levels $E_m^{\pm}$ overlap and splitting is not observed; in the opposite case, in a strong field, $\mathbf{d}_{mn}\vec{\mathcal{E}} > \gamma_m$, two maxima must be clearly observed in the absorption spectrum of the test radiation the distance between which depends linearly on the intensity of the field $\vec{\mathcal{E}}$.

Such an effect, which was first observed experimentally in the perturbation of molecular spectra by an electromagnetic field in the UHF range,[12] has been recently observed also in the case of perturbation of atomic spectra in a light field.[7] Starting with experiments on observing the splitting of the D-line in a potassium atom under the action of intense resonance light[13] a similar phenomenon was also observed in many other experimental situations.[14] The progress in experimental techniques of which we have spoken in the introduction has made it possible in recent times to obtain in a number of cases sufficiently accurate quantitative data characterizing resonance splitting in the optical frequency range.[15,16] As an example we refer to the results of the experiment in which the intense field of radiation from an infrared laser at a wavelength of 3.51 μm was utilized for the resonance perturbation of the transition $6P_{5/2} - 5D_{7/2}$ in the spectrum of a xenon atom. A test field of 4.54 μm wavelength was used to measure the population of the $5D_{3/2}$ state as a function of the deviation of the frequeccy of the test field from resonance with the $6P_{5/2}$ state. Figure 2 shows two maxima in the absorption spectrum of the test radiation corresponding to the quasienergy states of the $6P_{5/2}$ level. The distance between the maxima depended linearly on the intensity of the strong field.

We now consider the stationary regime in detail. In this case one should turn to the system of equations (5). Since in a stationary regime $\partial\rho/\partial t = 0$, this system is an algebraic one and can be easily solved. In particular, the probability of finding the electron in the upper level turns out to be equal to

$$\rho_{mm}(\infty) = \frac{|\mathbf{d}_{mn}\vec{\mathcal{E}}|^2}{2|\mathbf{d}_{mn}\vec{\mathcal{E}}|^2 + 4\Delta^2 + \gamma_m^2}. \qquad (8)$$

In accordance with the general remarks made in the introduction expression (8) does not depend on the initial conditions, and in particular, on the regime of switching on the perturbation. We note that always $\rho_{mm}(\infty) \leq \frac{1}{2}$.

Expression (8) enables us to obtain easily the total probability of spontaneous radiation per unit time:

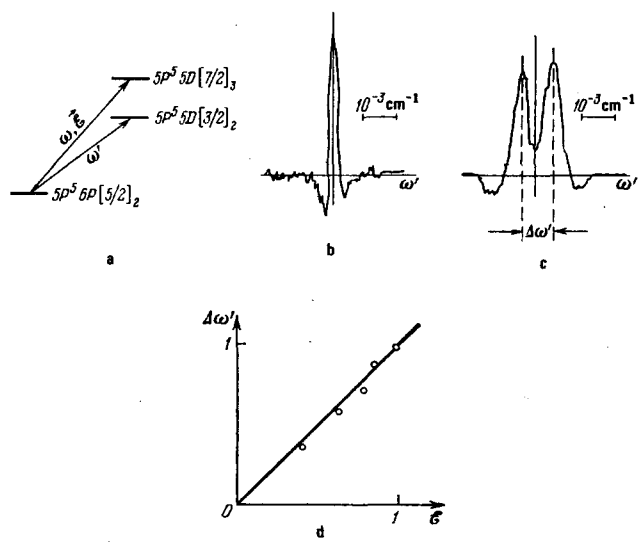

FIG. 2. Level splitting as a function of the intensity of a strong resonance field $\vec{\mathcal{E}}$, ω measured by the method of absorption of the test radiation ω'. a) Level scheme for the xenon atom (ω' is a weak test field); b) population of the upper level in the case of a weak resonance field ω; c) population in the case of a strong resonance field ω; d) dependence of the amplitude of the splitting $\Delta\omega'$ on the intensity of the strong field $\vec{\mathcal{E}}, \omega$.

[6] Sometimes the equivalent term "quadratic Stark effect in an alternating field" is utilized.

[7] The phenomenon under consideration is also called the "linear dynamic Stark effect", and also the "Autler-Townes effect".

$$w = \gamma_m \rho_{mm}(\infty). \quad (9)$$

On the other hand, we evaluate the total probability of unshifted radiation (elastic scattering) per unit time. The unshifted radiation is produced by the average dipole moment $\overline{\mathbf{d}(t)} = \rho_{mn}\mathbf{d}_{nm} + \text{compl. conj.}$, which oscillates harmonically with the frequency of the forcing field ω. It is well known from classical mechanics that the dipole will radiate just at this frequency. Thus, the total probability of unshifted radiation per unit time has the form[17]

$$w^{el} = \gamma_m |\rho_{mn}(\infty)|^2 = \frac{4\Delta^2 + \gamma_m^2}{4\Delta^2 + \gamma_m^2 + 2|\mathbf{d}_{mn}\vec{\mathcal{E}}|^2} w, \quad (10)$$

where w is determined by (9). In particular, in a weak field ($\mathbf{d}_{mn}\vec{\mathcal{E}} \ll \gamma_m$) formula (10) goes over into the well-known expression[18] obtained within the framework of the perturbation theory for resonance fluorescence:

$$w^{el} = \frac{(1/4)|\mathbf{d}_{mn}\vec{\mathcal{E}}|^2}{(\omega_{mn}-\omega)^2 + (1/4)\gamma_m^2} \gamma_m, \quad (11)$$

which is nothing other than the well-known Breit-Wigner formula for elastic resonance scattering by a quasi-discrete level.[19] Formula (11) describes a two-proton process in which a quantum of the external field is absorbed and a spontaneous quantum is emitted. This process is characterized by the Feynman graph shown in Fig. 3a. As a result of the law of conservation of energy the frequency ν of the emitted quantum is rigorously equal to the frequency ω of the absorbed quantum. It is just for this reason that the scattering is said to be "unshifted" (or "Rayleigh").

In a strong field the elastic scattering is described by expression (10). All the Feynman graphs corresponding to relation (10) contain only one emitted spontaneous quantum. A typical graph is shown in Fig. 3b. Again as a consequence of the law of conservation of energy the frequency ν of the emitted quantum is rigorously equal to ω.

From (10) we see that $w^{el} < w$. The remaining part of the scattering $w - w_{el}$ represents shifted (or inelastic) scattering. Feynman grapsh corresponding to inelastic scattering contain at least two emitted spontaneous quanta. A typical graph is shown in Fig. 3c. As can be seen from Fig. 3c, in accordance with the law of conservation of energy $\nu_1 + \nu_2 = 2\omega$, and the frequencies of the emitted quanta themselves ν_1 and ν_2 can differ from ω. From a physical point of view the shifted scattering represents spontaneous transitions between quasienergy states of a two-level system. In section 3 we shall consider the spectral distribution of the shifted scattering.

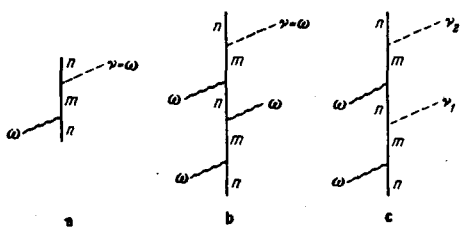

FIG. 3. Feynman graphs illustrating the process of resonance fluorescence. a) Unshifted scsttering in a weak field; b) unshifted scattering in a strong field; c) shifted scattering in a strong field.

It was assumed above that the strong field is strictly monochromatic. In actual fact in any real experiment this is not so. Therefore formula (10) should be multiplied by a factor which takes into account the line shape of the spectrum of the exciting light. Let $\Delta\omega$ be the width of this line. Then, obviously, the width of the spectrum of emitted quanta (the fluorescence spectrum) is determined by the smaller of the two widths $\Delta\omega$ or $(\gamma_m^2 + 2|\mathbf{d}_{mn}\vec{\mathcal{E}}|^2)^{1/2}$.

In a weak field the second width reduces to γ_m and the fact mentioned above is well known.[18] Numerous experiments in which resonance fluorescence has been observed were carried out under conditons when $\Delta\omega \gg \gamma_m$. This is the usual situation when the spectral width of the radiation of the exciting source is greater than the natural line width. In this case a fluorescence spectrum of width γ_m is observed. But the experiment can be carried out under conditions when the above inequality has the opposite sign. An example of such an experiment is given in Ref. 20. The barium atom was excited from the ground state into the excited state $^1P_1$ which has a natural width $\gamma_m \sim 5 \times 10^{-4}$ cm^{-1} by laser radiation with an effective width of the spectrum $\Delta\omega$ which is smaller than γ_m by a factor of severalfold. In this case the fluorescence spectrum was observed to be approximately twice as narrow as the natural width of this transition (Fig. 4).

3. RESONANCE FLUORESCENCE SPECTRA

The fluorescence phenomenon consists of scattering of light by an atom in the course of which the initial state of the atom is not altered. In the case when the frequency of the incident light is close to one of the atomic frequencies ω_{mn} the probability of fluorescence increases sharply and we are dealing with resonance fluorescence. Limiting ourselves to only such a case we consider the problem of the spectral distribution with respect to the frequencies of the emitted spontaneous photons in a two-level system which is acted upon by a monochromatic external field $\vec{\mathcal{E}}\cos\omega t$.

The total probabilities per unit time of the unshifted scattering w^{el} and of the shifted scattering $w - w^{el}$ are determined by the formulas of section 2. From the physical point of view the shifted scattering is determined by transitions between quasienergy levels. As can be seen from section 2 it is not prominent in weak

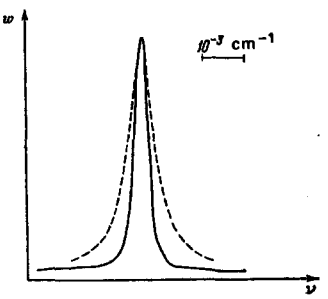

FIG. 4. The resonance fluorescence spectrum (solid line) in the case of a narrow spectrum of exciting radiation measured in Ref. 20. Dashed line is the Lorentz contour of the line in the case of spontaneous decay of the same state.

fields when these levels merge due to the spontaneous width, and conversely, is pronounced in strong fields. In a strong field one cannot consider the scattering process as a reemission of a single photon absorbed by the atom since in the course of a lifetime of the atom in an excited state other photons are scattered by it.[21]

In the case of unshifted scattering the spectral distribution has a trivial appearance: $dw^{el}(\nu)=w^{el}\delta(\nu-\omega)d\nu$, where w^{el} is determined by formula (10). As can be seen from Fig. 1 the spectral distribution of the shifted scattering must have maxima at frequencies corresponding to differences between quasienergy levels. The widths of these maxima are of the order of γ_m. There are three such maxima with the central one corresponding to scattering at the frequency ω, while the satellites are separated from it by twice the Rabi frequency. The aim of the present section is a quantitative description of the shapes of spectral distributions and their comparison with experiment.

According to the general principles of the theory of electromagnetic radiation in the dipole approximation the probability $dw(\nu)\sim\langle|\hat{d}_\nu|^2\rangle$, where $\hat{d}_\nu$ is the Fourier component of the operator for the dipole moment $\hat{d}(t)$, i.e., $\hat{d}_\nu=\int_0^T\hat{d}(t)e^{-i\nu t}dt$, while the brackets $\langle\cdots\rangle$ denote quantum mechanical averaging over the initial state of the system with one electron; T is the time during which the field acts.

The probability of elastic scattering is determined by the square of the average value of the dipole moment, i.e., $dw^{el}(\nu)\sim|\langle\hat{d}_\nu\rangle|^2$. Consequently, the inelastic scattering is determined by the difference $\langle|\Delta\hat{d}_\nu|^2\rangle=\langle|\hat{d}_\nu|^2\rangle-|\langle\hat{d}\rangle|^2$, i.e., by the quantum fluctuation of the dipole moment. Thus, while elastic scattering can be described within the framework of the classical radiation theory, inelastic scattering represents an essentially quantum effect.

For this reason it is necessary to introduce quantum operators for the creation and annilhilation of particles: a_i^+ and a_j. In this case, since in future we shall not be interested in different instants of time, it is convenient to take these operators in the Heisenberg representation i.e., depending on the time.[22]

In order to calculate averages of which we spoke above we introduce the operator for the atomic density matrix $\hat{\rho}=\sum_{i,j}a_i^+a_j\rho_{ij}$. Of course, in the general case the operator for the total density matrix of the whole system must depend also on the operators for the photons of the electromagnetic field. But if one assumes that the changes occurring in the atom have only a small effect on the states of the vacuum field then the operator for the total density matrix can be represented in the form of a product of two operators, with one factor depending on the operators for the atomic particle, and the other one depending on the operators for the photons. This is the "Markov (or factorization) approximation". This is violated, for example, for close levels when $\gamma_m\sim\omega_{mn}$. In such a case the time for the emission of a quantum $1/\gamma_m$ is comparable with the time of transition of the system

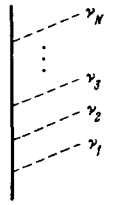

FIG. 5. Feynman graph for the emission of N quanta by a quasienergy state (solid line).

from one state to the other one $\sim 1/\omega_{mn}$ which follows from the energy-time indeterminacy relation. This leads to the appearance of retardation.

Averaging the operator for the total density matrix over the states of the photons according to the indicated Markov approximation we arrive at the operator introduced above for the atomic density matrix. This operator enables us to predict changes in atomic states without analyzing what happens at the same time to the state of the electromagnetic field.

We analyze the above statement in terms of Feynman graphs. The exact approach consists of obtaining the amplitude for the emission of a large number of quanta from the quasienergy states. Figure 5 shows a typical graph. The heavy lines in this diagram denote the propagation function for quasienergy states in contrast to the thin lines corresponding to the unperturbed atomic states.[23] The total amplitude is represented by the sum of all graphs of similar type. A transition to the description with the aid of the atomic density matrix corresponds to averaging over the frequencies of all the spontaneously emitted quanta with the exception of the one which is recorded by the measuring instrument.

The average value I of any operator $\hat{I}$ is calculated by means of the formula $I=\langle\hat{I}\rangle=\mathrm{Tr}(\hat{I}\hat{\rho})$. The operator for the dipole moment has the form $\hat{d}(t)=\sum_{i,j}d_{ij}a_i^+a_j$ where d_{ij} are the matrix elements of the dipole moment. In the expression

$$\langle|\hat{d}_\nu|^2\rangle=\int_0^T\int_0^T\langle\hat{d}^*(t)\,\hat{d}(t')\rangle\,e^{i\nu(t-t')}\,dt\,dt'$$

we go over to the variable $\tau=t'-t$ and let $T\to\infty$. We then obtain[24]

$$\langle|\hat{d}_\nu|^2\rangle=T\int_{-\infty}^{\infty}|d_{mn}|^2\,\overline{\langle\hat{\rho}_{nm}(t)\,\hat{\rho}_{mn}(t+\tau)\rangle}\,e^{-i\nu\tau}\,d\tau+\text{compl. conj.};\qquad(12)$$

here the line denotes averaging with respect to time t, and $\hat{\rho}_{nm}\equiv a_m^+a_n$. Formula (12) has meaning for the stationary regime when all the times are great compared to $1/\gamma_m$.

We now turn to the calculation of the averages. For simultaneous operators this is relatively uncomplicated. For example,

$$\langle\hat{\rho}_{mn}\rangle=\sum_{ij}\mathrm{Tr}(a_n^+a_ma_i^+a_j)\rho_{ij}=\sum_{ij}\delta_{mi}\delta_{nj}\rho_{ij}=\rho_{mn},$$

which clarifies the definition of $\hat{\rho}_{mn}$. In the derivation commutation relations for the operators a_m and a_i^+ have been utilized here, and also the fact that the state over which the averaging is taken contains one particle so that the action of two sequentially situated operators a_ma_j yields zero.

Formula (12) contains averages of the products of the operators $\hat{\rho}_{mn}$ at different instants of time. For the evaluation of such averages it is necessary to bring them to the same time, since only then do the Heisenberg operators have the usual commutation properties.[22] With this aim in mind we formally expand the operator $\hat{\rho}_{mn}$ at the time instant $t+\tau$ in terms of the complete system of operators $\hat{\rho}_{ij}$ at the time instant t:

$$\hat{\rho}_{mn}(t+\tau)=\sum_{ij}\rho_{mn}^{ij}(t,\tau)\,\rho_{ij}(t). \tag{13}$$

In view of the linear dependence of the operators $\hat{\rho}_{mn}(t+\tau)$ and the new quantities $\rho_{nm}^{ij}(t,\tau)$ regarded as functions of τ the equations for these new quantities formally coincide with the equations (5) for the density matrix ρ. Indeed, averaging relation (13) quantum mechanically we find that it is valid not only for the operators, but also for their averages, i.e., for the elements of the density matrix ρ_{ij}. The only new different aspect are the new initial conditions. They follow from (13) if in that relation we set $\tau=0$: $\rho_{kl}^{ij}(t,0)=\delta_{ki}\delta_{lj}$.

Substituting (13) into (12) we obtain

$$(|\hat{d}_\nu|^2)=T\,|d_{mn}|^2\sum_{ijkl}\mathrm{Tr}\,(a_m^+a_na_j^+a_ia_k^+a_l)\int_0^\infty\overline{\rho_{kl}(t)\,\rho_{mn}^{ij}(t,\tau)}\,e^{-i\nu\tau}\,d\tau+\text{compl. conj.}$$

We note that in this formula the lower limit of integration is equal to zero, and not to $-\infty$, as in the general case (12). This is associated with the fact that the operator $a(t+\tau)$ can annihilate a particle only after it is present at the time instant t, i.e., for $\tau\geq 0$.

Evaluating the trace of the product of six operators a^+, a in the same manner as was done above for the product of four operators, and going over from $\langle|\hat{d}_\nu|^2\rangle$ to the probability for the emission of a photon $dw(\nu)$ per unit time we obtain

$$dw(\nu)=\gamma_m\,2\,\mathrm{Re}\int_0^\infty[\rho_{mm}(t)\,\rho_{mn}^{mn}(t,\tau)+\rho_{nm}(t)\,\rho_{mn}^{nn}(t,\tau)]\,e^{-i\nu\tau}\,d\tau\,\frac{d\nu}{2\pi}. \tag{14}$$

Averaging over t in (14) is superfluous since in the stationary regime there is no dependence on t. This can be easily verified utilizing for the quantities appearing in (14) the stationary solution (8) of the system (5).

If one integrates expression (14) over ν and takes into account the initial conditions for ρ_{mn}^{kn} then, as should have been expected, we obtain the total probability of scattering (9).

Further, if in (14) we consider the case $\tau\to\infty$, then $\rho_{mn}^{mn}\to 0$, while $\rho_{mn}^{nn}\to\rho_{mn}(t+\tau)$. Substituting these values from (14) we obtain, as should have been expected, the probability of elastic scattering (10).

The solution of equations (5) for the quantities $\rho_{kl}^{ij}(t,\tau)$ in the general case is quite awkward due to the Cardano formulas for the characteristic values.[17] In different limiting cases it takes on a more tractable form. In what follows we shall consider only the case of zero deviation from resonance: $\Delta=0$. The roots of the characteristic cubic equation of the system (5) in this case have the simple form:

$$s_0=-\frac{1}{2}\gamma_m,\quad s_\pm=-\frac{3}{4}\gamma_m\pm\sqrt{\left(\frac{1}{4}\gamma_m\right)^2-|\mathbf{d}_{mn}\vec{\mathscr{E}}|^2}.$$

while any solution of (5) can be represented in the form

$$\rho\sim a+be^{s_0t}+ce^{s_+t}+de^{s_-t},$$

where the coefficients a, b, c, d are determined by the initial conditions.

For a weak field, in particular for $|\mathbf{d}_{mn}\vec{\mathscr{E}}|<\gamma_m/4$, all the roots mentioned above are real. In this case in addition to coherent scattering determined by the constant a there appears also inelastically scattered light of frequency ν near ω which represents a superposition of three resonances with the same position but of different widths. All these widths, as has been noted already, are of order γ_m.

But if on the other hand the field is strong, specifically $|\mathbf{d}_{mn}\vec{\mathscr{E}}|>\gamma_m/4$ (cf., criterion (3)), then we see that in addition to the central peak at a frequency ω of width γ_m there appear two symmetrically situated satellites of widths $3\gamma_m/2$ at a distance $(|\mathbf{d}_{mn}\vec{\mathscr{E}}|^2-(\gamma_m/4)^2)^{1/2}$ from the central peak. In particular, for a very strong field the satellites are separated from the central peak by an amount of twice the resonance Rabi frequency $|\mathbf{d}_{mn}\vec{\mathscr{E}}|$, as has already been mentioned above.

Calculation using formula (14) in this last case leads[17] to the following result for $dw(\nu)\simeq dw^{\text{inel}}(\nu)$ (as has been noted above the elastic scattering is negligibly small):

$$dw(\nu)=\left\{\frac{1/3}{[4(\nu-\omega-|\mathbf{d}_{mn}\vec{\mathscr{E}}|)/3\gamma_m]^2+1}+\frac{1}{[2(\nu-\omega)/\gamma_m]^2+1}+\frac{1/3}{[4(\nu-\omega+|\mathbf{d}_{mn}\vec{\mathscr{E}}|)/3\gamma_m]^2+1}\right\}\frac{d\nu}{2\pi}. \tag{15}$$

As should have been expected after integration of this expression over ν we obtain $w=\gamma_m/2$ which corresponds to spontaneous emission from the level m the probability of finding an electron in which is equal to $\frac{1}{2}$.

The result (15) could also have been obtained by a considerably simpler method without turning to the general expression (14) and the functions ρ_{kl}^{ij} depending on two different times. From Fig. 1 it is clear that transitions between them can occur as a result of emission of four photons: one of frequency $\omega-|\mathbf{d}_{mn}\vec{\mathscr{E}}|$, one of frequency $\omega+|\mathbf{d}_{mn}\vec{\mathscr{E}}|$ and two of frequency ω. In the case of a very strong field for $\Delta=0$ the probability of finding the electron in these quasilevels is the same, so that the total probabilities per unit time for the emission of each of the photons indicated above are equal to $(1/8)\gamma_m$. Taking into account the fact that the spectral widths for the emission of each of these quanta are equal, as we have seen, respectively to $(3/2)\gamma_m$ and γ_m (this result does not require the introduction of functions depending on two different times), we obtain formula (15) directly. Naturally, such a simple approach is not suitable for the case of nonzero deviations from resonance or for weaker fields.

Experiments on observing the resonance fluores-

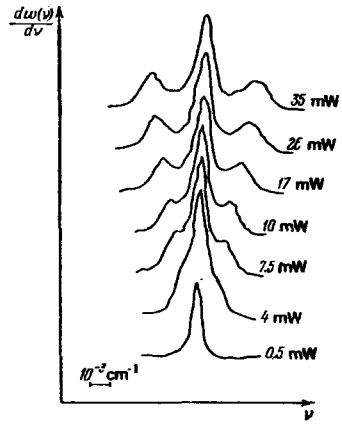

FIG. 6. Resonance fluoresence spectrum as a function of the power of the exciting radiation (the parameter is stated on the curves).[20]

cence spectrum in a strong field[20,25] were carried out by means of exciting the transition between components of the hyperfine structure in a sodium atom $3S_{1/2}(F'=2) - 3P_{3/2}(F'=3)$.

The fluorescence spectrum observed in Ref. 20 for different values of the intensity of the exciting field and different values of the deviation from resonance is shown in Fig. 6. It can be clearly seen that as the intensity of the resonance field is increased satellites appear in addition to scattering at the undisplaced frequency. This occurs for values of fields satisfying condition (3). The amplitude of the central maximum in a very strong field is greater than the amplitudes of the satellites by approximately a factor of three, while the width of the satellites is one and a half times greater than that of the central peak; the frequencies of the satellites differ from the undisplaced frequency by an amount of twice the resonance Rabi frequency. Thus, both qualitatively and quantitatively relation (15) gives a good description of the experimental data (Fig. 7).

The dependence on the deviation from resonance is illustrated by Fig. 8. In complete agreement with theory we note that the satellites cease to be observed when the deviation from resonance is of the order of the width of the central maximum. Results obtained in other papers[25] are of a similar nature.

Summarizing one can assert that the experimental data on the resonance fluorescence spectra in a strong

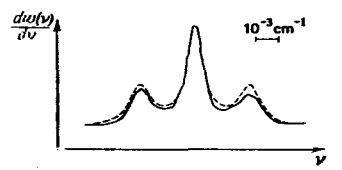

FIG. 7. Resonance fluorescence spectrum in a strong field calculated on the basis of the data of Ref. 17 (dashed line) and measured in Ref. 20 (solid line).

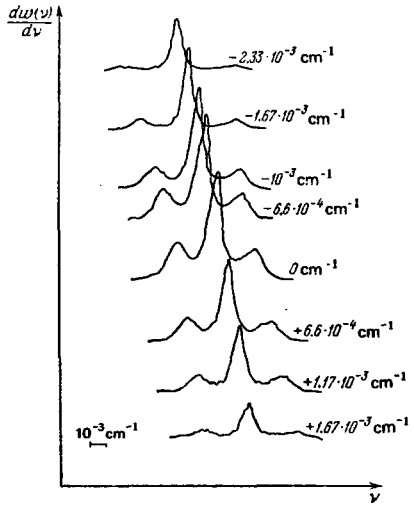

FIG. 8. Resonance fluorescence spectrum as a function of the deviation from resonance (the parameter is shown on the curve) between the frequency of the strong external field and the transition frequency (results of experiment of Ref. 20).

field are well described by theory both qualitatively and quantitatively.

4. THREE-LEVEL SYSTEM IN A RESONANCE FIELD

In the preceding section a description was given of scattering of light in the course of which the atom remains in the initial state. We next consider the process when the atom after resonance scattering of light goes over into a new state. In this case, obviously, it is necessary to take into account at least three atomic levels.

We first investigate the simplest situation shown in Fig. 9a: an electron situated in the initial state n makes a transition after the strong field $\vec{\mathcal{E}}\cos\omega t$ of frequency ω close to ω_{mn} has been instantaneously switched on at the instant of time $t=0$ to the state m from which it makes a further transition to the state l accompanied by the emission of a spontaneous quantum. Just as before, we are interested in the solution for large times $t \gg 1/\gamma_m$.

The specific feature of the strong field consists of

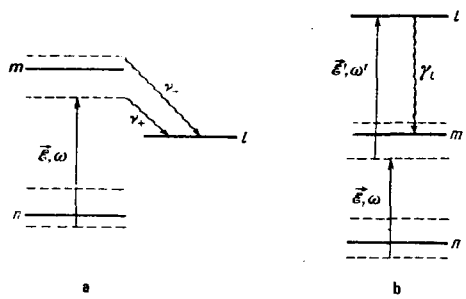

FIG. 9. Diagrams of processes occurring in a three-level system. a) Spontaneous Raman scattering; b) case of two external fields $\vec{\mathcal{E}}, \omega$ and $\vec{\mathcal{E}}', \omega'$.

the fact that the electron can oscillate many times between the states m and n and only subsequently pass from the state m to the state l. As was noted in section 2, such oscillations can be described using the language of quasienergy states. It is obvious that in this problem we are interested in quasienergy states of the level m. In contrast to the situation considered in section 2, in the present case it is necessary to take into account the spontaneous decay of quasienergy states to the third final state l. This can be accomplished by applying the Breit-Wigner procedure to the state m. If this is done for the quasienergies (6) then in an arbitrary case the expression for the splitting of the quasilevels has a fairly awkward appearance. Therefore we here restrict ourselves to the case of zero deviation from the m,n resonance: $\Delta=0$. Then the frequencies of the emitted spontaneous quanta are concentrated near the values $\nu_{+}=\omega_{ml}+\frac{1}{2}(|\mathbf{d}_{mn}\vec{\mathcal{E}}|^2 - (\frac{1}{4})\gamma_m^2)^{1/2}$. In the limiting case $\gamma_m=0$ the frequencies of the emitted quanta are determined as before by expressions (6). We see that splitting is observed only in strong fields $|\mathbf{d}_{mn}\vec{\mathcal{E}}|>\gamma_m/2$. We note that in a very strong field ($|\mathbf{d}_{mn}\vec{\mathcal{E}}|\gg\gamma_m$) the resonance splitting is smaller by a factor of two than in the case of resonance fluorescence, this being explained by the absence of splitting of the level l in the present case. We note that for $\Delta\neq0$, if $\Delta\gg\gamma_m$, then in accordance with (6) splitting can be observed in arbitrary fields.

We now turn to the question of the probability of emission of spontaneous radiation with the particle making a transition to the level l as a function of the frequency of the emitted photon ν. In contrast to the case of fluorescence we are here dealing with an absolute probability, and not with probability per unit time. It is obvious that for $t\gg1/\gamma_m$ the particle completely goes over to the state l, i.e.,

$$w_l=\gamma_m\int_0^\infty|a_m(t)|^2dt=1;$$

here $a_m(t)$ is the amplitude for finding the electron in the level m at the instant of time t. The method of obtaining this quantity in the resonance approximation for a two-level system m,n was described in section 2. It now needs only to be modified by means of applying the Breit-Wigner procedure to the state m.[26] It is based on the fact that in accordance with our assumption the total spontaneous width γ_m corresponds to a transition to the level l, while the spontaneous width of the corresponding transition to the state m is negligibly small. Setting

$$w_l=\int_{-\infty}^{\infty}dw_l(\nu)=\gamma_m\int_{-\infty}^{\infty}|a_m(\nu)|^2\frac{d\nu}{2\pi}=1,$$

one can obtain the probability for the emission of a photon $dw_l(\nu)$ in the frequency range $[\nu,\nu+d\nu]$; here

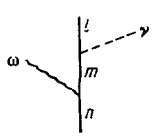

FIG. 10. Feynman graph for processes of spontaneous Raman scattering.

$a_m(\nu)$ is the Fourier-component of the quantity $a_m(t)$.

The general expression for $dw_l(\nu)$ is quite awkward. We restrict ourselves to the case $\Delta=0$ and to very strong fields $|\mathbf{d}_{mn}\vec{\mathcal{E}}|\gg\gamma_m$. We then obtain

$$dw_l(\nu)=\left[\frac{\gamma_m/4}{(\nu-\omega_{ml}-(1/2)|\mathbf{d}_{mn}\vec{\mathcal{E}}|)^2+(1/16)\gamma_m^2}+\frac{\gamma_m/4}{(\nu-\omega_{ml}+(1/2)|\mathbf{d}_{mn}\vec{\mathcal{E}}|)^2+(1/16)\gamma_m^2}\right]\frac{d\nu}{2\pi}. \tag{16}$$

We see that the spectrum consists of two peaks with widths which are by a factor of two smaller than the wdith γ_m, while the probabilities of transitions from both quasienergy states are the same. As the field is decreased the splitting of the quasienergy levels is diminished and in the limit of a weak field both resonances merge into one of width γ_m.

If we pass on to deviations from resonance different from zero, then although, as we have mentioned earlier, the splitting of the state m may be observed also in a weak field, nevertheless the intensity will in fact be great for a transition of frequency corresponding to the law of conservation of energy being satisfied. In this case the expression for the probability of scattering of light is well known[14]:

$$dw_l(\nu)=\frac{(1/4)|\mathbf{d}_{mn}\vec{\mathcal{E}}|^2\gamma_m}{(\omega_{mn}-\omega)^2+(1/4)\gamma_m^2}\delta(\omega-\nu-\omega_{ln})\,d\nu. \tag{17}$$

It is described by the Feynman diagram shown in Fig. 10. It may be seen that the relation (17) is similar to the relation (11) for the probability of resonance fluorescence. This is quite natural, since both processes are qualitatively similar—induced transition, spontaneous transition. Correspondingly the probabilities of both processes are also of the same order of magnitude. The process of Raman scattering of light in a weak external field can also be characterized by a quantity which does not depend on the intensity of the field, —the scattering cross section. It is obtained from the expression for the probability (17) by the standard method—division by the factor $c\vec{\mathcal{E}}^2/8\pi\omega$. We note that the relation (17) can be obtained from the well known Kramers-Heisenberg formula,[27] if in the latter one neglects the nonresonance terms compared with the resonance ones. The quantitative criteria for the applicability of (17) were stated in the introduction—they follow from the condition of the absence of mixing in the two-level system m,n.

If the lower level n also has a spontaneous width γ_n, then the expression (16) should be modified by the replacement in the denominator of the width: $\gamma_m\to(\gamma_m+\gamma_a)$.[26] This result can be obtained also from the general Breit-Wigner theory. Then the total probability is given, as it should be, by $w_l=\gamma_m/(\gamma_m+\gamma_n)<1$.

Finally, if the probability of a spontaneous transition from the state m to the state n is not small, then, as has been noted above, the Breit-Wigner formalism is inapplicable, and one must utilize the density matrix method[28] for the solution of the problem. As a result of this the structure of expression (16) is qualitatively preserved, but the resonance widths entering into this

expression will be complicated functions of different partial widths.

Experimentally the process shown in Fig. 9a, referred to as spontaneous Raman scattering of light by an atom, has been observed both under conditions of exact resonance (cf., for example, Ref. 29), and in the case of a large deviation from resonance (cf., for example, Ref. 30). But for a number of reasons all the experimental data known until now have been obtained under such conditions that they did not give interesting information concerning the spectrum of the scattered photons. First of all these data were all obtained under conditions when the Doppler effect played a significant role, and therefore resolution as to frequency was insufficient. Secondly, in the majority of experiments the process of induced Raman scattering was observed—in order to increase the yield of the scattered light an extended atomic target was utilized in which the forcing field at a frequency close to the frequency of the transition ω_{ml} was produced as a result of the large number of atoms in the target and consequently as a result of the large number of scattered quanta.

From the theoretical point of view the description of induced scattering is close to the description of spontaneous scattering given above. We consider in (16) a certain very narrow frequency range $[\nu_0, \nu_0+d\nu]$ in the neighborhood of one of the resonances. Then $\gamma_m d\nu$ is a quantity proportional to the power of the spontaneous radiation in this frequency range. If in the same frequency range we apply to the system a weak field $\vec{\mathscr{E}}'\cos\nu t$, such that $|\mathbf{d}_{ml}\vec{\mathscr{E}}'| \ll \gamma_m$, then the probability of the transition m,l can be calculated by means of perturbation theory: $w_{ml} = 2\pi(\mathbf{d}_{ml}\vec{\mathscr{E}}'/2)^2\delta(\nu-\nu_0)$. Multiplying this by $d\nu$ we obtain a quantity proportional to the power of the induced radiation: $(\pi/2)|\mathbf{d}_{ml}\vec{\mathscr{E}}'|^2$. When the condition $|\mathbf{d}_{ml}\mathscr{E}'|^2 \gg \gamma_m d\nu$ is satisfied induced scattering will be considerably more prominent than spontaneous scattering. In this case in formula (16) in the term contained therein corresponding to the frequency interval $d\nu$ indicated above one should replace the power of spontaneous emission $\sim\gamma_m d\nu$ appearing in the numerator by the power of induced emission $\sim(\pi/2)|\mathbf{d}_{ml}\vec{\mathscr{E}}'|^2$; as a result of this we obtain the probability of induced emission in the form

$$w_l^{\mathrm{el}}(\nu) = \frac{dw_l(\nu)}{d\nu}\frac{\pi}{2\gamma_m}|\mathbf{d}_{ml}\vec{\mathscr{E}}'|^2 \qquad (18)$$

(we recall that the ratio of the power of induced scattering to the power of spontaneous scattering is characterized by the Einstein coefficient). Since as a result of the weakness of the field $\vec{\mathscr{E}}'$ this probability is small compared to unity it does not affect the condition $\int dw_l(\nu)d\nu = 1$.

In a number of experiments[31] quantitative measurements were made which enabled one to obtain information on the total probability of scattered light. These data obtained under conditions both of a weak and also of a strong field are well described by the theory given above. This is quite natural, since the total probability of scattering does not depend strongly on the spectral characteristics of the resonance process.

We now consider the case when the third level l lies higher than both resonating levels m,n (cf., Fig. 9b). Then obviously the state l cannot be populated as a result of spontaneous transitions (in contrast to the case of Fig. 9a). If in the case of the transition m,l one switches on a weak field $\vec{\mathscr{E}}'\cos\nu t$ of frequency ν close to ω_{lm}, then this field will lead to induced transitions to the level l. We determine the probability w_l of finding the particle in the state l by considering the field $\vec{\mathscr{E}}'$ within the framework of the first order of the theory of nonstationary perturbations, i.e., by assuming that $|\mathbf{d}_{lm}\vec{\mathscr{E}}'| \ll \gamma_l$. We assume for the sake of simplicity that spontaneous emission with the width γ_l occurs only from the level l to the level m, while spontaneous emission from m to n is negligibly small. As before, we restrict ourselves to the stationary regime. Then solution of the equations for the quantum mechanical amplitudes is relatively uncomplicated.[32] In this case the Breit-Wigner procedure is utilized for the state l. For the case of zero deviation from resonance $\Delta = \omega_{mn} - \omega = 0$ and of a very strong field ($|\mathbf{d}_{mn}\vec{\mathscr{E}}| \gg \gamma_l$) the probability of scattering as a function of the frequency of the weak field ν has the form

$$w_l = \left[\frac{1}{(\nu-\omega_{lm}-(1/2)|\mathbf{d}_{mn}\vec{\mathscr{E}}|)^2+(1/4)\gamma_l^2} + \frac{1}{(\nu-\omega_{lm}+(1/2)|\mathbf{d}_{mn}\vec{\mathscr{E}}|)^2+(1/4)\gamma_l^2}\right]\left|\frac{\mathbf{d}_{lm}\vec{\mathscr{E}}'}{4}\right|^2. \qquad (19)$$

As was expected expressions (16) and (19) have a similar structure. Again, the width γ_m of the transition from the state m to the state n is not small, then one must solve the system of equations (5) for the density matrix. The result has a form analogous to (19), but with a width which is a combination of γ_l and γ_m: $\gamma_l \to \gamma_l + \frac{1}{2}\gamma_m$.[32]

Experimental results obtained in Refs. 16 for the case of exact resonance demonstrate the validity of equation (19). In similar experiments two lasers were used which emit in the visible spectrum. The strong radiation field from one of the lasers was tuned to be in resonance with a transition from the ground state of the atom to an excited state. The weak field of the radiation from the second laser with variable frequency of generation ν was utilized for measuring the dependence of the probability of finding the electron in the excited state $5^2S(F=2)$ as a function of the frequency ν. This probability was recorded by means of observing the spontaneous decay of the third state. Figure 11 shows the dependence of the occupancy of the excited state on the frequency ν and on the intensity of the strong field. Both in this and in other cases that have been investigated two maxima which follow from (19) are clearly observed.

Until now the whole investigation of the three-level system was carried out for the case when one of the external fields is equal to zero or is small. Investigation of the general case when the second field $\vec{\mathscr{E}}'$ is also strong ($|\mathbf{d}_{lm}\vec{\mathscr{E}}'|^2 \gtrsim \gamma_l$) represents a much more complicated problem. We note that no experimental investigations of the spectrum of the scattered light in the

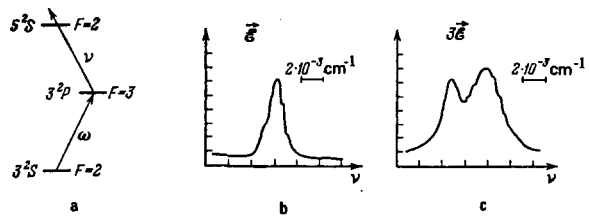

FIG. 11. Intermediate resonance in a three-level system. a) Level scheme for the sodium atom; b) population of the upper state in the case of a weak resonance field $\vec{\mathcal{E}}, \omega$; c) the same in the case of a strong field (data from experiment of Ref. 16).

case of two strong fields have been carried out. However such a situation can easily arise in the case of induced Raman scattering of light when the intensity of the field $\vec{\mathcal{E}}'$ is great. Just as in the case of the two-level system we consider only two limiting cases: the relaxationless and the stationary regimes.

Thus we first consider the case when the times are so short that spontaneous widths are not important. The resonance fields $\vec{\mathcal{E}}$ and $\vec{\mathcal{E}}'$ acting respectively on the transitions n, m and m, l, lead to oscillations of the probabilities of finding the particle in each of the three levels under consideration. In the resonance approximation the problem can be solved relatively simply[33] and one can obtain the probabilities averaged over time. We shall present a number of results of such solutions (for the system of Fig. 9b). It is well known that in a two-level system no choice of the deviation from resonance and of the field will lead to the probability of finding the particle in the upper level being greater than the probability of finding the particle in the lower level. In contrast to this in a three-level system it is possible to choose the fields $\vec{\mathcal{E}}$ and $\vec{\mathcal{E}}'$ with zero deviations from resonance in both cases in such a manner that the average probability of finding the particle in the upper level l would be greater than either of the probabilities of finding the particle in level m and n. But even in the optimal case the inversion of the population of the state l with respect to n does not exceed 3% while the average probability itself of finding the particle in the level l amounts to 35%.

Still more interesting results are obtained in the stationary regime.[34] In this case the problem should be solved by the density matrix method. From the solution it follows that for zero deviation from both resonances and under the condition $|\mathbf{d}_{mn}\vec{\mathcal{E}}| \gg |\mathbf{d}_{lm}\vec{\mathcal{E}}'| \gg \gamma_m \gg \gamma_l$ the stationary probability of finding the particle in the upper level tends to 100%.

Each of the levels of the three-level system situated in two strong resonance fields is split into three quasi-energy levels, so that in the general case for each of the two transitions —m, n and l, m—the spectral line consists of seven resonance peaks. In the case of zero deviation from both resonances the number of resonances reduces to five. In Ref. 34 the theory of correlation functions involving two times described in section 3 was utilized for numerical calculations of the line shape.

The situation is simplest in the case of very strong fields and zero deviation from resonance. In this case along with the central peak at the undisplaced frequency $\nu = \omega_{mn}$ there exist two satellites on each side whose distances from the central peak amount respectively to $\pm\frac{1}{2}(|\mathbf{d}_{mn}\vec{\mathcal{E}}|^2 + |\mathbf{d}_{lm}\vec{\mathcal{E}}'|^2)^{1/2}$ and $\pm(|\mathbf{d}_{mn}\vec{\mathcal{E}}|^2 + |\mathbf{d}_{lm}\vec{\mathcal{E}}'|^2)^{1/2}$.[33] A similar picture is observed near the central peak at the undisplaced frequency $\nu = \omega_{lm}$. The widths of all the peaks are combinations of the widths γ_m and γ_l.

5. MULTIPHOTON EXCITATION

As is the case for multiphoton process, multiphoton excitation of atoms is not a threshold process with respect to the intensity of the external field. However, in order that the the probability of multiphoton excitation of an atom should be sufficiently great for observation, the intensity of the exciting field must also be sufficiently great. In a strong external field the definition of the process of multiphoton excitation is that it is an induced multiphoton transition from the lower state to the upper one the lifetime of which is determined by the process of its spontaneous decay. In other words, this is multiphoton spontaneous Raman scattering. Obviously competition of induced transitions from the upper state to other bound states and into the continuous spectrum sharply narrows the possibilities of realizing multiphoton excitation. For this reason in the case of atoms and of the frequency range of visible light one can practically be concerned only with relatively few-photon excitation processes.

The probability of a K-photon transition from the state n to the state m under the action of an external field of intensity $\vec{\mathcal{E}}$ and frequency ω is described within the framework of the nonstationary perturbation theory of the K-th order by means of replacing in the formula for the single photon probability the quantity $\mathbf{d}_{mn}\vec{\mathcal{E}}/2$ by $V_{mn}^{(K)}\mathcal{E}^K$. The quantity $V_{mn}^{(K)}$ is referred to as the multiphoton matrix element and has the well known form

$$V_{mn}^{(K)} = \sum_{a,b,\ldots,p} \frac{1}{2^K} \frac{(\mathbf{ed}_{ma})(\mathbf{ed}_{ab})\ \ldots\ (\mathbf{ed}_{pn})}{[\omega_{an}-(K-1)\omega][\omega_{bn}-(K-2)\omega]\ \ldots\ [\omega_{pn}-\omega]}. \tag{20}$$

It is described by the Feynman diagram shown in Fig. 12. This compound matrix element takes into account virtual transitions both into the bound states and into the states of the continuous spectrum.

From (20) it can be seen that the magnitude of the multiphoton matrix element depends sharply on the frequency of the external field, and that as the frequency or its higher harmonics approach the frequency of some intermediate transition in the spectrum of the atom the

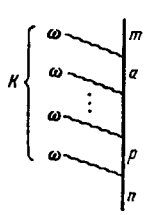

FIG. 12. Feynman graph illustrating multiphoton excitation.

matrix element increases without limit within the framework of the given approximation. Correspondingly, just as in other similar cases, perturbation theory is applicable only to such frequencies for which the denominators in (20) are larger than the spontaneous widths of the intermediate states. If this is not so, then the problem appears to be relatively simple only for weak fields when $V_{sn}^{(K')}\mathcal{E}^{K'} \ll \gamma_s$, where $K' = \omega_{sn}/\omega$ is the order of the intermediate resonance. Then in accordance with the Breit-Wigner procedure one should replace in (20) $E_s^{(0)}$ by $E_s^{(0)} - \frac{1}{2}i\gamma_s$.

In accordance with what has been said above we note the changes and modifications that must be made in the formulas describing different resonance phenomena in the preceding sections. In going over from the single photon to the K-photon resonance in a two-level system (cf., section 2) the deviation from resonance Δ is replaced by the multiphoton deviation from resonance $\Delta_K = \omega_{mn} - K\omega + \delta E_{mn}$, while the Rabi frequency Ω is replaced by the multiphoton Rabi frequency $\Omega_K = \frac{1}{2}(\Delta_K^2 + 4|V_{mn}^{(K)}|^2\mathcal{E}^{2K})^{1/2}$, where δE_{mn} is the difference between the nonresonance shifts of the levels m and n.[35] From the form of the Rabi frequency it follows that in order to calculate it correctly in the case of K-photon resonance it is necessary to know δE_{mn} with an accuracy up to terms of order $K-1$.

We discuss the criteria for the applicability of the resonance approximation to K-photon resonance. It is obvious that the criterion (2) goes over into the criterion

$$|V_{mn}^{(K)}|\mathcal{E}^K \ll \omega_{mn}. \tag{21}$$

Criterion (1) is replaced by one which is considerably more demanding:

$$\Delta_K \ll \omega_{mn}\left(\frac{\mathcal{E}}{\mathcal{E}_{at}}\right)^{K-1}. \tag{22}$$

Indeed, if $\Delta_K \gtrsim \omega_{mn}(\mathcal{E}/\mathcal{E}_{at})^{K-1}$, then the amplitude of the multiphoton resonance transition is of order $V_{mn}^{(K)}\mathcal{E}^K/\omega_{mn}(\mathcal{E}/\mathcal{E}_{at})^{K-1} \sim d_{mn}\mathcal{E}/\omega_{mn}$, i.e., it is of the same order of magnitude as the nonresonance amplitude of the transition m,n in the first (nonvanishing) order of perturbation theory.

In addition to the resonance criteria in the case of multiphoton transitions the criterion for the field strength $\vec{\mathcal{E}}$ is also altered. In place of the criterion (3) the field should now be considered strong if the condition $V_{mn}^{(K)}\mathcal{E}^K \gtrsim \gamma_m$ is satisfied. The critical intensity satisfying this condition is considerably higher than in the single photon case.

We now turn to the probability of multiphoton excitation. If we adopt the scheme of Fig. 9a, then the probability of multiphoton excitation from the state n to the state m with a subsequent spontaneous transition to the state l has for weak fields the form

$$w_l^{(K)} = \frac{\gamma_m}{(\omega_{mn} - K\omega)^2 + (1/4)\gamma_m^2}|V_{ml}^{(K)}|^2\mathcal{E}^{2K}; \tag{23}$$

here γ_m is the spontaneous width for a transition from the state m to the state l. As everywhere else, this formula refers to the stationary regime. We note that for $K=1$ formula (23) agrees with (17). It is not useful to consider the case of strong multiphoton excitations in the sense indicated above since the fields corresponding to it rapidly approach atomic fields with increasing K.

Calculation of the multiphoton matrix elements (20) requires overcoming typical difficulties associated with the choice of the optimal wave function for the optical electron in a complex atom, and also with the necessity of carrying out infinite summations over intermediate states. The nature of the first of these difficulties is one of principle. The methods of overcoming this difficulty are analogous in the case of bound-bound and bound-free transitions; they have been discussed in sufficiently great detail in reviews devoted to the multiphoton ionization of atoms[36] and to the nonresonance perturbation of atomic levels.[37] Here we merely note that the number of concrete calculations of multiphoton matrix elements for bound-bound transitions is very limited[38] so that at present there is no possibility of reaching any conclusions concerning the optimal methods of calculation.

In calculating the multiphoton matrix element (20) it is necessary to satisfy the selection rules for multiphoton transitions; in a typical case when one can restrict oneself to the dipole approximation the selection rules for the orbital quantum number have for a K-photon transition[39] the form $\Delta l = l_m - l_n = -K, -K+1, \ldots, K-1$, K independently of the degree of ellipticity of the external field. The selection rules with respect to the magnetic quantum number depend on the ellipticity of the external field. In the case of linear polarization $\Delta M = 0$, for circular polarization $\Delta M = +1$, for the general case of elliptical polarization the values $\Delta M = -K, -K+1, \ldots, K-1, K$ are allowed. We note that in the often encountered case of circularly polarized light and an initial S-state in the case of $\Delta M = K$ we have $\Delta l = K$, since the angular momentum cannot be less than its component.

In the case of linear and circular polarizations it is not necessary to take into account degeneracy with respect to the magnetic quantum number since there is no mixing of degenerate substates. In the case of elliptical polarization each state is characterized by a superposition of substates with different M. This superposition may, in principle, be found by means of solving the corresponding secular equation.[37] However such a procedure is required only for the final state and for the case of a strong field. For intermediate states, and also for a final state in a weak field the choice of the basis does not affect the result as a consequence of summing over all the substates of the degenerate level.

In a hydrogen atom and in the case of hydrogenlike atoms there is no point in finding the probability of transitions under the action of a strong field to states with a definite angular momentum since states with different angular momenta are mixed in an elliptically polarized field.[40] A similar mixing occurs also between states of an atomic multiplet when the criterion $V_{nn'}^{(2)}\mathcal{E}^2 \gtrsim \omega_{nn'}$ is satisfied, where n,n' are different states of

the multiplet, while the matrix element describes their nonresonance perturbation in an external field.[41]

If we now turn to experiment, then the first point that must be noted is the absence of systematic experimental data on multiphoton matrix elements for bound-bound transitions. However, on the other hand, a large number of experiments have been carried out in which basically two-photon excitation was realized (as a rule, of the first excited states in alkali atoms). The results of these experiments related to multiphoton excitation are well described by theory. It should be noted that in these experiments one had the simplest situation for their theoretical interpretation—there were only a few intermediate states so that one had to take into account a small number of terms in the multiphoton matrix element. We do not give here references to these experiments since data on multiphoton excitation can be extracted from them only in an indirect manner.

We note only one experiment in which there was observed a completely obvious but practically important role of the presence of an intermediate quasiresonance state, i.e., of a state with an extremely small deviation from resonance.[42] As can well be seen from (20), a decrease in one of the denominators of the expression for the multiphoton matrix element can increase by many orders of magnitude the probability of multiphoton excitation. Figure 13 shows the result of the experimental mentioned above in which observations were made of the dependence of the probability of two-photon excitation of the $4D$ state in the sodium atom on the deviation from resonance in the case of an intermediate single photon resonance with the $3P$ state.

In investigating multiphoton bound-bound transitions it is necessary to keep in mind that it is not always possible to restrict oneself to the dipole approximation. We have in mind here such frequencies of the external field for which resonance quadrupole transitions can compete with nonresonance dipole transitions. The effectiveness of the competition by the quadrupole transitions is due to the fact that the relatively smaller numerator (matrix element) is compensated by the relatively smaller (resonance) denominator, so that the probability of the transition can turn out to be comparable with the probability of a nonresonance dipole transition. Experimentally in a number of papers[43,44] single photon resonance transitions were observed between quadrupole coupled states in different atoms. Data from an experiment[43] in which in the same experimental arrangement both dipole and quadrupole transitions were observed made it possible to determine with good accuracy the magnitude of the matrix elements for the quadrupole transitions. In this experiment (Fig. 14) a sodium atom was excited under resonance conditions by relatively weak light from one laser from the ground state into the $3P$ state. Under the action of relatively strong light from a second laser the frequency of generation of which could be varied, the electron, on absorbing two photons, made a transition into the continuous spectrum. Depending on the frequency of this second laser the two-photon transition could take place through different intermediate states, among them the $4D$ state ($\Delta l = 1$, dipole transition) and the $4F$ state ($\Delta l = 2$, quadrupole transition). The ions were recorded and probabilities of transitions via the indicated states were compared: the probability of the dipole transition turned out to be greater by a factor of approximately 10^5. From this ratio and from the well-known magnitude of the corresponding dipole matrix element it was possible to obtain the magnitude of the quadrupole matrix element $|\langle 3P|r^2|4F\rangle| \approx 10^{-15}\,\text{cm}^2$. This quantity is well described by calculations,[45] as well as the ratio between the matrix elements for the dipole and quadrupole transitions proportional to n^2, where n is the principal quantum number.

We now turn to the question of violating the conditions for realizing multiphoton excitation due to the appearance of induced transitions from the excited state. It is obvious that the experimental criterion for the appearance of induced transitions is the deviation of the dependence of the probability of multiphoton excitation as a function of the intensity of the exciting field from the corresponding power law. Such deviations were observed in a number of experiments on two-photon ex-

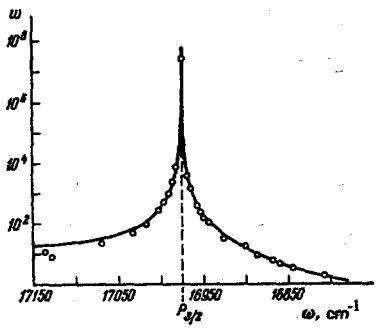

FIG. 13. Probability w of two-photon excitation of a sodium atom in the $4D_{5/2}$ state as a function of the deviation from intermediate resonance between the frequency of the external field ω and the energy of the transition to the $3P_{3/2}$ state (experiment of Ref. 42).

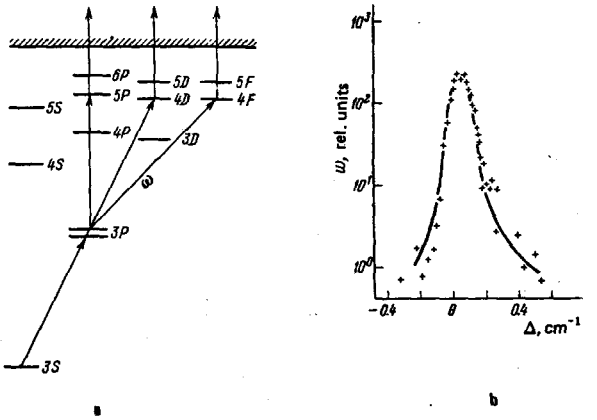

FIG. 14. a) Resonance transitions in the spectrum of a sodium atom – dipole ($3P - 4D$) and quadrupole ($3P - 5P$ and $3P - 4F$); b) probability w of ionization of a sodium atom as a function of the deviation Δ from resonance between the frequency of an external field ω and the energy of the quadrupole transition $3P - 4F$ (result of the experiment of Ref. 43).

citation,[46] but these experiments did not give the answer as to what competing process was taking place.[47] The frequently realized case when competition is provided by ionization from excited states—the resonance process of multiphoton ionization—is discussed in the next section. As regards multiphoton mixing of resonance states, so far no experiments are known in which this interesting process is observed. We note that in a real atom as a result of the criteria on the intensity of the field indicated above ($V_{mn}^{(K)}\mathcal{E}^K \geq \gamma_m$) we can be dealing only with two-photon mixing.

Finally, we make a remark concerning multiphoton emission. It is obvious that this process is qualitatively different from multiphoton excitation, since one of the virtual transitions can have a spontaneous nature. The presence in addition to spontaneous transitions also of an induced transition results in a greater probability of such a process compared to the probability of spontaneous multiphoton emission which, as is well known, is very small.[48] However, for the process of induced multiphoton emission to be realized it must successfully compete with the process of single-photon or cascade spontaneous emission. It is just for this reason that the induced emission so far has been observed only under fairly exotic conditions—such as the decay of the metastable 2S-state of deuterium situated in the field of radiation from a laser using neodymium glass.[49] With the energy of excitation of this state being approximately 10 eV and with the energy of the quantum of the external field being approximately 1 eV photons were observed with an energy of approximately 9 eV. Thus, a simple two-photon decay of the metastable state was occurring in which the process of emission of a single photon of energy in the neighborhood of 1 eV was of an induced nature. The magnitude of the cross section of the process indicated above measured in this experiment is well described by calculations.[49]

In conclusion we note that when we are speaking of multiphoton transitions the presence of an intermediate quasiresonance bound state must, obviously, increase the probability of induced multiphoton emission by many orders of magnitude.[50]

6. RESONANCE IONIZATION OF ATOMS

It is customary to say that the process of multiphoton ionization of atoms is a resonance one if the energy of one or several quanta of the external field is close to the energy of the transition from the ground to some excited bound state. We first consider a particular simplest process of two-photon resonance ionization (Fig. 15) and assume that it takes place under the action of two fields of different frequencies ω and ω' and of intensities $\vec{\mathcal{E}}$ and $\vec{\mathcal{E}}'$.

In the case of a weak resonance field $\vec{\mathcal{E}}(|d_{mn}\vec{\mathcal{E}}| \ll \gamma_m)$ and of a weak ionizing field $\mathcal{E}'(w_{mE} \ll \gamma_m)$ the probability of resonance ionization is described by the obvious relation within the framework of perturbation theory:

$$w = \frac{\frac{1}{4}|d_{mn}\vec{\mathcal{E}}|^2}{(\omega_{mn}-\omega)^2+(1/4)\gamma_m^2} w_{mE} \sim \mathcal{E}^2\mathcal{E}'^2, \qquad (24)$$

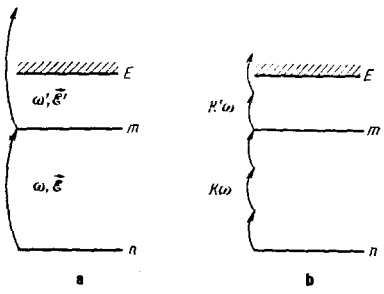

FIG. 15. Diagram for the process of resonance ionization of an atom. a) Two-photon ionization in two fields $\vec{\mathcal{E}}, \omega$ and $\vec{\mathcal{E}}', \omega'$; b) multiphoton ionization in a single field $\vec{\mathcal{E}}, \omega$.

where $w_{mE} \sim \mathcal{E}'^2$ is the probability of ionization from the state m per unit time, while $(\omega_{mn} - \omega)$ is the deviation from resonance.[8)]

Formula (24) can be easily generalized to the multiphoton case. It follows from the results of section 5 that for this it is necessary to carry out the replacements $\frac{1}{2}d_{mn}\mathcal{E} \to V_{mn}^{(K)}\mathcal{E}^K$; $\Delta \to \Delta_K = \omega_{mn} - K\omega$; $w_{mE} \to w_{mE}^{(K')}$. At the same place in section 5 criteria are stated for the weakness of the field which are valid in the case of multiphoton transitions.

The process of resonance ionization in a weak external field has wide practical application both for the spectroscopy of highly excited atomic states,[51] and also for a wide range of problems associated with selective action of laser radiation on an atomic medium—separation of isotopes,[52] obtaining polarized electrons[53] (cf., also the end of this section) and polarized nuclei.[54]

In a strong resonance field $\vec{\mathcal{E}}$ splitting of the state m takes place into two quasilevels the energies of which are determined by formulas (6). (Obviously splitting of the ground state is of no interest in the process under consideration.) We assume, as always, that the time during which this field acts is great, $|d_{mn}\vec{\mathcal{E}}|T \gg 1$. Since $w_{mE} \sim \mathcal{E}'^2$, while the resonance Rabi frequency $\Omega = |d_{mn}\mathcal{E}| \sim \mathcal{E}$, then at a not very high intensity of the ionizing field, when $\mathcal{E}' \sim \mathcal{E}$, the inequality $|d_{mn}\mathcal{E}| \gg w_{mE}$ is satisfied. In this case at first the filling of both quasilevels of the state m takes place, and then ionization occurs from these states. If the condition $w_{mE}T \ll 1$ is satisfied then the probability of resonance ionization is determined by the relation[55]

$$w = \frac{(1/2)|d_{mn}\vec{\mathcal{E}}|^2}{(\omega_{mn}-\omega)^2+|d_{mn}\vec{\mathcal{E}}|^2} w_{mE}, \qquad (25)$$

the general form of which is analogous to formula (24). However in the present case the width of the resonance is determined by the quantity $|d_{mn}\vec{\mathcal{E}}| \gg \gamma_m$.[9)] It is obvious

[8)] Under the conditions for the applicability of formula (24) the process of resonance ionization is sometimes called "cascade ionization"; this term reflects the specific features of such a process which can also be regarded as a transition $n \to m \to E$.

[9)] Sometimes in scientific literature one speaks of "field broadening of the resonance".

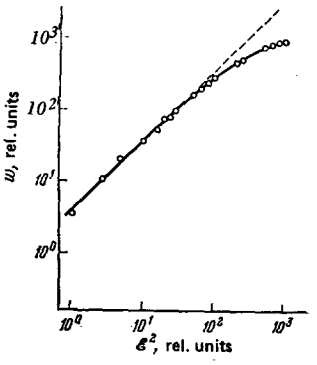

FIG. 16. Probability w of two-photon resonance ionization as a function of the intensity $\vec{\mathcal{E}}$ of the resonance field (results of Ref. 53).

that the transition from (24) to (25) associated with an increase in the intensity of the resonance field $\vec{\mathcal{E}}$ must manifest itself in the reduction of the rate of growth of w compared to a power law. Experimentally such a slowing down was observed[53] in the case of two-photon ionization of a cesium atom by two fields in accordance with the scheme $S_{1/2}(F=4) \to P_{3/2} \to E$ (F is the hyperfine structure quantum number). The deviation begins at an intensity of the resonance field $\mathcal{E} \sim 50$ V/cm (Fig. 16). This value agrees well with the estimate of the conditions for resonance mixing in a two-level system given in the Introduction.

If the ionizing field is so great that the condition $w_{mE} T \gg 1$ is satisfied, then instead of the probability per unit time (25) we go over to the absolute probability of ionization which is determined by the first factor in (25). In the case of exact resonace with $\Delta \lesssim |d_{mn}\vec{\mathcal{E}}|$ the probability of ionization ~1 is attained during atomic times.

We now consider the case when the transitions nm and mE are of multiphoton nature and are brought about by a single field (for the sake of simplicity), with $K > 2K'$ (cf., Fig. 15). In such a case it is evident that $V_{mn}^{(K)}\mathcal{E}^K \ll w_{mE}^{(K')}$; the probability of finding the electron in the state m remains small at all times, and the process of ionization from this state can be described within the framework of perturbation theory. The probability of resonance ionization is described by the relationship[56]:

$$w = \frac{|V_{mn}^{(K)}|^2 \mathcal{E}^{2K}}{\Delta(\vec{\mathcal{E}}) + (1/4)(w_{mE}^{(K')})^2 + (1/4)\gamma_m^2} w_{mE}^{(K')}, \qquad (26)$$

where $\Delta(\vec{\mathcal{E}}) = \omega_{mn} - K\omega + \delta E_{mn}(\vec{\mathcal{E}})$, $\delta E_{mn}(\vec{\mathcal{E}})$ is the nonresonance change in the energy of the transition mn in the spectrum of the atom under the action of an external field (dynamical polarizability of these states). The appearance of the quantities $w_{mE}^{(K')}$ and γ_m in the denominator of the expression (26) is a consequence of applying the Breit-Wigner procedure to the state m. Formula (26) is valid for times $T \gg 1/w_{mE}^{(K')}$, $1/\gamma_m$, with $w \ll w_{mE}^{(K')}$. It is obvious that for a sufficiently high intensity of the field $\vec{\mathcal{E}}'$ one can neglect the spontaneous width γ_m compared with the ionization width $w_{mE}^{(K')}$ which determines the resonance width.

Experimentally the process of resonance multiphoton ionization of atoms under conditions when $K > 2K'$ has been observed for different values of K and, as a rule, for $K' = 1$.[36] It should be noted that the process of ionization at a degree of nonlinearity $K > 3$ is observed with sufficiently great efficiency only in cases of an appreciable field intensity $\mathcal{E} \gtrsim 10^6$ V/cm,[36] when the nonresonance shifts of atomic levels including the resonance state are great, and exceed not only the spontaneous width of the levels but also the ionization width. Therefore resonance in the case of ionization under such conditions is, as a rule, observed with perturbed atomic levels under conditions when the energy of the transitions from the ground state to the resonance state differs from the corresponding energy in the unperturbed spectrum of the atom. In the case of a very high degree of nonlinearity and, correspondingly, of a very high intensity of the external field the change in energy becomes comparable with the distance to the nearest levels.[57,58] As long as the change in energy is not great it is well described by calculations of the dynamical polarizability of resonance state carried out by perturbation theory methods.[37] Thus, observation of the process of resonance ionization in an intense field is one of the methods for investigating the nonresonance perturbation of atomic levels.[37,59] In the case of strong perturgation the question arises of the classification of resonance states which may be new states arising in the system atom plus light field.[60] The widths of the observed resonances (Fig. 17) are described with satisfactory accuracy by the probability of ionization from resonance states.

The general solution of the problem of resonance ionization of atoms[61] enables us to obtain data both concerning the intermediate case when $|V_{mn}^{(K)}\mathcal{E}^K| \sim w_{mE}^{(K')}$ and also concerning cases when the external field acts for only a short time.

We now turn to the angular distribution and the degree of polarization of electrons originating in the resonance process of multiphoton ionization of atoms. We start with the natural assumption that the ground state is not oriented, i.e., all values of the magnetic quantum number in this state can be realized with equal

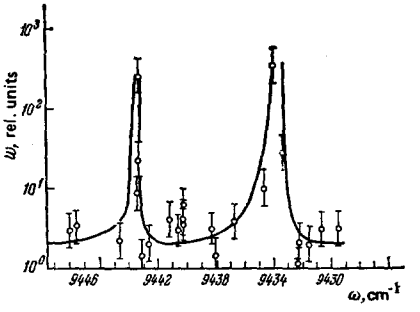

FIG. 17. Resonance dependence of the probability w of eleven-photon ionization of a krypton atom on the frequency of radiation from a single-mode laser ω. The resonances occur as a result of absorption of 10 quanta, so that their true width is ten times greater (result of an experiment in Ref. 58).

probability. In the general case intermediate states characterized by different magnetic quantum numbers are realized with different probabilities. In the case of a linearly polarized external field this is due to a difference in the values of the matrix elements for transitions between different states, in the case of circularly polarized field this is due to appropriate selection rules. Thus, not only the initial state, but also the intermediate state determines in a significant manner the properties of the emerging electrons. Both the angular distribution and the degree of polarization (i.e., the greater probability of realizing final states with magnetic quantum numbers having a definite sign) of the emerging electrons depends in the case of intermediate resonance on the specific characteristics of the resonance transition and on the degree of ellipticity of the perturbing field.

As calculations have shown,[62] by choosing appropriate parameters characterizing the atom and the external field (or external fields) one can obtain completely polarized electrons. Experiments[53] have confirmed the validity of theoretical predictions concerning the possibility of attaining a high degree of polarization together with a high efficiency of obtaining electrons.

The whole above discussion assumed that there is no relaxation of the intermediate state. If one assumes the existence of significant relaxation, then the angular distribution is characterized only by the transition from the intermediate state, and polarization cannot be significant. Three causes can lead to mixing of intermediate states characterized by different magnetic quantum numbers—ionization broadening of the resonance state, mixing of the ground and the resonance states, and also a short duration of the perturbing pulse T. Finally, one other cause is possible due to which the intermediate state can be uniformly populated—it is the selection rules. An example is the frequently encountered case of the transition $S_{1/2}, P_{1/2}$, occurring under the action of linearly polarized light. Among different experiments in which the angular distribution of electrons formed as a result of resonance ionization was observed the case mentioned above was realized in Ref. 63. Resonance ionization of sodium atoms was observed (the ground state is $3S_{1/2}$) in the presence of intermediate resonance with the $3P_{1/2}$ state. In accordance with theoretical predictions the angular distribution of electrons was observed which is well described by the relation $w = a + b\cos^2\theta$ typical of single-photon ionization.[64]

7. CONCLUSIONS

In the preceding sections we have discussed certain typical elementary resonance nonlinear-optics phenomena without attempting to describe all possible variants of the interaction of intense light with atoms (these questions have been covered in greater detail in the monograph of Ref. 2). From our point of view the problem consisted of describing the principal phenomena from a unified point of view.

It is necessary to note once again that the framework of the model which we have adopted—the interaction of intense monochromatic light with isolated atoms—unifies a wide range of problems associated with selective action of laser radiation on an atomic medium. One should at the same time be reminded that if one is interested in multiphoton transitions then the required selectivity can be realized also in a gaseous target (and not in a beam) by utilizing the method of colliding light beams.[37] The transition from a target in the form of a beam to a target in the form of a rarified gas enables one to increase the density of neutral atoms by many orders of magnitude. When dense gaseous targets are used in which during the time of action of the intense field collisions occur within the interaction volume between electrons, ions and atoms and the atoms of the target, the most significant effects are those associated with the presence of the field. These are an increase in the energy of the electrons as a result of inverse bremsstrahlung[65] and the radiative collisions of atoms.[66] It is specifically these effects, and not the gas-kinetic collision broadening,[67] that determine the optimal experimental conditions which depend in this case on three parameters—the density of the atomic target, the intensity of the light field and the duration of its action on the target. The possibility of practical utilization of nonlinear resonance effects in a number of cases is associated with the necessity of realizing a high field intensity over a large interaction volume, which is sometimes difficult or impossible to attain with a laser operating in a single-mode generation regime. If one utilizes the multimode regime of radiation which enables one to obtain a considerably greater energy in the generated pulse it is necessary to have in mind the space-time fluctuations of the intensity of the radiation due to the fluctuations in the amplitudes and the phases of the modes being generated. The presence of fluctuations leads to a number of specific effects which can significantly distort the resonance distributions.[68]

It is obvious that a detailed presentation of questions associated both with collisions and with the nonmonochromatic nature of the exciting radiation falls outside the framework of the subject under discussion. To the same extent the problem of describing the interaction of intense light with molecules also falls outside the framework of this discussion. The principal reason for this is well known—a molecular spectrum is considerably richer due to the vibrational and rotational degrees of freedom the resonance frequencies corresponding to which lie outside the frequency range of visible light. A description of the interaction of an intense electromagnetic field with molecules represents a separate topic.

Finally, we note that even within the framework of this review we did not aim to produce a complete bibliography, encompassing all the published data and reflecting the history of the investigations. In actual fact the number of publications devoted to the subject of this review is several times greater than the number cited by us. We gave preference to those sources in which the necessary material is presented in the most general form, for example in Ref. 69.

Numerous discussions of different questions touched upon in this review with our friend V. A. Khodov were exceptionally useful for us, and his sudden death was an irreplacable loss.

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