Nonlinear and parametric effects in atomic rf spectroscopy

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Usp. Fiz. Nauk **125**, 449–488 (July 1978)

Nonlinear phenomena occurring during interaction between an atom and a strong rf field of resonant or nonresonant frequency are reviewed. The account is given in both semiclassical and consistently quantum-mechanical languages. The concept of an atom "dressed by the radiation field" in which not only the optical-frequency field but also the radio-frequency field is quantized is presented in detail. The results are summarized of theoretical and experimental studies of multiphoton transitions, radiative shifts of normal magnetic resonance lines, parametric resonance, and coherent resonances under the conditions of optical pumping. The influence of nonresonance rf fields on magnetic properties and the rate of magnetic relaxation of atoms is discussed.

PACS numbers: 32.30.Bv, 43.50. + q, 33.25. − j, 42.65. − k

CONTENTS

1. INTRODUCTION

Radio-frequency spectroscopy is a branch of atomic physics dealing with transitions between the energy levels of an atomic system, induced by electromagnetic fields in the radio and microwave frequency bands. The range of application of radio-frequency spectroscopic methods is exceedingly broad, not only because the frequency band covered by it extends from a few hertz to many thousands of megahertz, but also because of the great variety of resonance phenomena observed by these methods. Having originally been developed as a powerful tool for investigating energy levels in experiments with atomic and molecular beams, radio-frequency spectroscopic methods were subsequently successfully extended to media in liquid and solid phases (NMR and ESR spectroscopy). The discovery of optical methods for the "preparation" of atomic systems and for the detection of their state (double rf-optical resonance[1] and optical orientation of atoms[2]) has substantially extended the subject matter of radio-frequency spectroscopy. It has led to the application of the method of magnetic resonance to the study of the ground and excited states of atoms in the gaseous phase of very low pressures (10^{-6}–10^{-3} Torr) when these atoms exhibit either nuclear or electronic paramagnetism. The "optical pumping" technique is very flexible in relation to the methods that can be used to prepare the atomic system, the range of frequencies employed, and the possibility of recording a number of observable variables, and has enriched radio-frequency spectroscopy by the discovery of new physical effects (multiphoton transitions,[3] parametric resonance,[4,5] coherence resonances[6] or "off-diagonal" resonances,[7–9] and so on) connected with different aspects of the interaction of the electromagnetic field with matter.

Extensive literature has emerged in recent years on the theoretical and experimental analysis of the interaction between atomic systems and one or more strong rf fields. All this work can be assembled under the general heading of "nonlinear radio-frequency spectroscopy" because all the effects under consideration are characterized by a nonlinear response of an atomic system to the application of a strong rf electromagnetic field. In this sense, nonlinear atomic rf spectroscopy has the same relation to ordinary radiospectroscopy as nonlinear laser spectroscopy[10,11] has to ordinary linear

 0038-5670/78/070589-22$01.10

optical spectroscopy.[1)] This analogy is not purely formal because, in general, the same phenomenon is involved in both cases, namely, the nonlinear interaction between the electromagnetic field and matter. The results obtained in the radio band thus naturally supplement those available in the optical band. The flexibility of the methods used in radio-frequency spectroscopy is such that the researcher is now presented with very extensive possibilities for the experimental verification of many theoretical predictions obtained within the framework of nonlinear optics.

Our aim is to review the basic theoretical and experimental work on nonlinear effects of rf spectroscopy with particular emphasis on the physics of the phenomena. Most published work on the experimental verification of particular theoretical predictions has been carried out with optically oriented atomic systems. It will be assumed that the reader is adequately familiar with the optical methods employed in rf spectroscopy. These have been reviewed frequently in both Soviet[12,13,20] and foreign[14-16] literature.

There are several methods for the theoretical description of nonlinear phenomena in atomic systems. They differ by the approach to the solution of the problems, by the mathematical methods employed, by the degree of physical completeness, and by ease of interpretation. Nevertheless, in most cases, they lead to identical or almost identical results. It has not been our intention to compare these methods in detail and we do not regard any of them as definitely to be preferred. Nevertheless, we shall devote considerable space to one of these methods, namely, the idea of an atom "dressed" by the field.

In contrast to other methods (vector, phenomenological, or "neoclassical"[74]), this method is the only one that is consistently quantum-mechanical. It enables us to describe the entire range of phenomena accompanying the interaction of both optical and radio fields with atomic systems from a unified point of view. This simple, elegant, and beautiful method was developed and widely used by the French school of spectroscopy headed by A. Kastler, whose work will be summarized in some detail in this review. Nevertheless, it is not our intention to contrast their work with the work of other authors. On the contrary, in many cases, explanations given in the language of an atom "dressed" by the field are accompanied by the more graphic explanations given in the language of the vector model.

There is a large number of published theoretical and experimental papers on nonlinear effects in optical and rf spectroscopy. Different aspects of this problem have frequently been discussed on the pages of this journal[11,12,19,70] and in special monographs. Our review will be largely devoted to problems of nonlinear rf spectroscopy.

The references given at the end are not intended to be complete. They are simply a relatively easily accessible source of additional information extending the range of this review.

2. METHODS FOR THE THEORETICAL DESCRIPTION OF NONLINEAR EFFECTS IN RF SPECTROSCOPY

The great majority of papers published at the time of writing are based on the approach in which the mathematical formalism used to interpret nonlinear effects employs the semiclassical theory of interaction between an rf field and matter, in which the field is described classically and the atomic system quantum-mechanically. This approach is natural in view of the fact that the average number of photons per rf mode is exceedingly large. This means that one can neglect the spontaneous emission of rf photons and retain only the phenomena of absorption and stimulated emission. Moreover, since the phase of the rf field is determined with a high degree of precision, it would appear that the choice of the classical approach to the description of the field is dictated by its coherence. In fact, comparison of the results of calculations based on the semiclassical and quantum-mechanical approaches demonstrates that the two are completely equivalent whenever the calculations can be performed by either method up to terms of the same order of small quantities.

It is well known that the quantum-mechanical formalism which makes use of the concepts of absorption and emission of rf photons is often very much simpler. For example, in multiphoton transitions, simple application of the laws of conservation of energy and angular momentum leads much more rapidly and conveniently to the prediction of observable resonances as compared with calculations based on classical theory. On the other hand, many of the phenomena known in rf spectroscopy as "coherence resonances"[4,6,7] can be easily and simply interpreted within the framework of the semiclassical formalism. The analysis of the role of photons in these phenomena is then found to encounter considerable difficulties. Thus, coherence resonances cannot be explained from the standpoint of real transitions between the energy levels of an atomic system, accompanied by absorption and emission of rf quanta, because these phenemena are observed under the conditions of "transverse" pumping when a population difference is not produced between the sublevels of the system. Moreover, such transitions cannot ensure the conservation of total energy and total angular momentum of the system. Thus, for example, if the rf field is polarized along the direction of the constant magnetic field (π polarization), each photon has a zero angular momentum component along $\mathbf{H}_0$ and absorption and emission of any number of such photons cannot communicate to the atomic system the angular momentum necessary for the $\Delta m = \pm 1$ transition.

The above examples emphasize the necessity for developing a quantum theory capable of providing an adequate unified description of all the physical effects that arise during the interaction between an arbitrary polar-

[1)] The only nonlinear reaction of a spin system to an external disturbance is saturation of the radio-frequency transition. This was taken into account already in early work on $+f$ spectroscopy.

ized rf field and matter. This aim has been achieved in the rigorous quantum theory of the "dressed" atom.[17] On the other hand, a semiclassical theory of magnetic resonance in strong rf fields has recently been developed,[18,19,21] and its application to the analysis of coherent phenomena in atomic systems has led to results that agree with the predictions of the theory of the "dressed" atom and with experimental data. We shall extensively employ these two equivalent approaches to the theoretical description of nonlinear phenomena without attempting to contrast them. In each case, we shall tend to use the particular approach that is the simplest mathematically and yields results that are simpler to interpret.

A. Semiclassical theory of magnetic resonance in strong rf fields

The semiclassical model of the interaction between atoms and an rf field has led not only to the development of a linear theory of magnetic resonance[22] but also to the description of many of the observed nonlinear effects. In particular, it has been shown[23] that the center of a resonance line corresponding to a transition in an ensemble of spin $\frac{1}{2}$ atoms interacting with even a weak rf field $2\mathbf{H}_1\cos\omega t$ perpendicular to the constant magnetic field $\mathbf{H}_0$ will shift by the amount $\delta = -(\gamma H_1)^2/4\omega$ (this is the Bloch-Siegert shift[24]). A further increase in the amplitude of the rf field is accompanied by multiphoton transitions which can also be described within the framework of the semiclassical theory.[25-27 2)]

1) *Formulation of the problem.* Consider a paramagnetic atom with magnetic moment $\mu = \gamma \mathbf{J}$ ($\hbar = 1$) interacting with a constant magnetic field $\mathbf{H}_0$ and an alternating field $\mathbf{H}_1(t)$ which can have arbitrary polarization. In the semiclassical approach, the atomic system is described quantum-mechanically, whereas the electromagnetic field is regarded as classical. All the variables characterizing the field are represented by ordinary algebraic variables whilst dynamic variables describing the atom are looked upon as operators. The behavior of the atom in a field of this kind then reduces to the solution of the Schrödinger equation with the Hamiltonian

$$\hat{\mathcal{H}}(t) = -\hat{\mu}\mathbf{H}(t) = -\hat{\mu}(\mathbf{H}_0 + \mathbf{H}_1(t)). \tag{1}$$

In most cases that are of interest in practice, the field $\mathbf{H}_1(t)$ either rotates about a given axis or oscillates along this axis. Because of the different symmetry properties of the field $\mathbf{H}_1(t)$, these two cases have to be considered separately.

The exact solution of the problem can be found in the two special cases where $\mathbf{H}_1(t)$ rotates in the plane perpendicular to the direction of the constant field $\mathbf{H}_0$ and when it oscillates along this field.

The case of $\mathbf{H}_1(t)$ rotating around $\mathbf{H}_0$, i.e.,

$$\mathbf{H}_1(t) = H_0\mathbf{e}_z + H_1(\mathbf{e}_x\cos\omega t + \mathbf{e}_y\sin\omega t), \tag{2}$$

(where $\mathbf{e}_x, \mathbf{e}_y, \mathbf{e}_z$ are unit vectors along the coordinate axes) is a standard case in the theory of magnetic resonance. The unitary transformation

$$\hat{S}(t) = \exp(+i\hat{J}_z\omega t) \tag{3}$$

may be looked upon as a transformation to a coordinate system rotating about the axis $OZ \| \mathbf{H}_0$ and enables us to write the transformed Hamiltonian in the form

$$\hat{\mathcal{H}}' = \hat{\mathcal{H}}_0 + \hat{\mathcal{H}}_1' - \omega_0\hat{J}_z, \tag{4}$$

where

$$\hat{\mathcal{H}}_1' = \hat{S}\hat{\mathcal{H}}_1\hat{S}^{-1}.$$

The Schrödinger equation can now be written in the form

$$i\frac{\partial}{\partial t}|'t\rangle = \hat{\mathcal{H}}_1'|'t\rangle, \tag{5}$$

where $|'t\rangle$ is the state vector of the atomic system in the transformed coordinate frame at time t. The Hamiltonian then no longer depends on time, and (5) is easily integrated to yield

$$|'t\rangle = \exp[-i\hat{\mathcal{H}}_1'(t-t_0)]|'t_0\rangle. \tag{6}$$

If there exists an operator $\hat{T}$ that diagonalizes the transformed Hamiltonian $\hat{\mathcal{H}}_1'$, the state vector $|t\rangle$ in the laboratory frame at time t can be written in the form

$$|t\rangle = \hat{U}(t, t_0)|t_0\rangle, \tag{7}$$

where the time-shift operator is given by

$$\hat{U}(t, t_0) = e^{-i\hat{J}_z\omega_0 t}\hat{T}^{-1}e^{-i\hat{T}\hat{\mathcal{H}}_1'\hat{T}^{-1}(t-t_0)}\hat{T}e^{it\hat{J}_z\omega_0 t}. \tag{8}$$

2) *Solution of the problem in special cases.* The solution obtained by the above method for a system of spin $\frac{1}{2}$ particles is given by Series.[28]

When the field $\mathbf{H}_1(t)$ oscillates along $\mathbf{H}_0$, the Hamiltonian has the form

$$\hat{\mathcal{H}} = -\hat{\mu}\mathbf{e}_z(H_0 + H_1\cos\omega t) = \gamma\hat{J}_z(H_0 + H_1\cos\omega t). \tag{9}$$

The choice of the OZ axis as the quantization axis enables us to perform the integration of the Schrödinger equation without the use of an additional transformation of the basis:

$$|t\rangle = \exp\left(-i\int_{t_0}^{t}\hat{\mathcal{H}}(t')\,dt'\right)|t_0\rangle = \hat{U}(t, t_0)|t_0\rangle, \tag{10}$$

where

$$\int_{t_0}^{t}\hat{\mathcal{H}}(t')\,dt' = \hat{J}_z\left[\gamma H_0(t-t_0) + \frac{\gamma H_1}{\omega}(\sin\omega t - \sin\omega t_0)\right]. \tag{11}$$

Since it is well known that

$$e^{-ia\hat{J}_z\sin\omega t} = \sum_{n=-\infty}^{n=+\infty} J_n(a\hat{J}_z)e^{-in\omega t}, \tag{12}$$

where J_n is the Bessel function of the first kind, we can write (10) in the form

$$|m, t\rangle = e^{-im\omega_0(t-t_0)}\left[\sum_{n=-\infty}^{+\infty} J_n(am)e^{-in\omega t}\sum_{l=-\infty}^{+\infty} J_l(am)e^{-il\omega t_0}\right]|m, t_0\rangle, \tag{13}$$

where $\omega_0 = \gamma H_0$, $a = \gamma H_1/\omega$, and m is the magnetic quantum number. The expression given by (13) can be rearranged to read

$$|m, t\rangle = \left[\sum_{n, l} J_n(am)J_l(am)e^{-i(n-l)\omega t}e^{-i(m\omega_0+l\omega)(t-t_0)}\right]|m, t_0\rangle. \tag{14}$$

This expression contains two groups of terms that are

2) The possibilities and range of validity of the semiclassical theory of interaction between a two-level system and radiation are discussed in the excellent paper by Mandel.[14]

periodic functions of time, namely, terms with frequencies $(n-l)\omega$, whose phase is uniquely determined by the phase of the field $H_1(t)$, and terms with frequencies $m\omega_0 + l\omega$, whose phase is determined by the time interval $t - t_0$.

Under the conditions of stationary rf interaction, the phase of the latter terms will, in general, be a random quantity. However, when

$$(m - m')\omega_0 = -(l - l')\omega \tag{15}$$

this random phase is eliminated from the density matrix, so that the element (m, m') of this matrix can be written in the form

$$\sigma_{mm'} = \sum_{\substack{n, l; \\ n', l'}} J_n(am) J_l(am) J_{n'}(am') J_{l'}(am') e^{-i(n-l-n'+l')\omega t}. \tag{16}$$

The condition given by (15) is essentially the condition for the degeneracy of certain atomic energy levels (level crossing[12b]). The expressions given by (15) and (16) describe the effect known as parametric resonance,[4,5] which is observed at frequencies ω that are multiples of ω_0. Its characteristic feature is the absence of radiative broadening and radiative line shift.

The semiclassical formalism has recently been used[18] to analyze the radiative shift of the magnetic resonance line for spin $\frac{1}{2}$ particles when the radiofrequency field $\mathbf{H}_1(t)$ acting on the system oscillates in the plane perpendicular to the field $\mathbf{H}_0$ (this is the Bloch-Siegert effect). The recent interest in this apparently well-known question is not accidental. Experimental studies have shown[29,30] that the dependence of the observed shift on the amplitude of the field $H_1(t)$ follows the previously established quadratic law[24] only for small values of H_1. The first attempt to explain the discepancy on the basis of quantum electrodynamics[31] was unsuccessful. The theoretical predictions were in poor agreement with experimental results. It is only in more recent papers[18,32,33] in which both the quantum and semiclassical approaches were employed that a successful solution was achieved and higher-order corrections were obtained, so that the Bloch-Siegert effect could be satisfactorily described over a broad range of variation of the rf field amplitude.

Pegg[18a] has used a formalism analogous to that described above, in which the semiclassical Hamiltonian describing the interaction between spin $\frac{1}{2}$ particles and the field

$$\mathbf{H}(t) = H_0 \mathbf{e}_z + 2H_1 \cos \omega t \cdot e_x \tag{17}$$

assumes the form

$$\hat{\mathcal{H}}(t) = -\hat{\mu}H(t) = \omega_0 \hat{J}_z + 4b \cos \omega t \cdot \hat{J}_x, \tag{18}$$

where $\omega_0 = -\gamma H_0$ and $b = -\gamma H_1/2$.

The usual transformation to the reference frame rotating with frequency ω, which is accomplished with the aid of the operator (3), is inadequate because, when the amplitude of the field H_1 is large, one can no longer neglect the components of the oscillating field rotating in the opposite direction. Better results are obtained with the aid of the unitary transformation

$$\hat{S}(t) = \exp[i\hat{J}_z(a \sin 2\omega t + (p+1)2\omega t)]\, \hat{R}^{-1}(\theta) \exp(-i\hat{J}_z \cdot 2\omega t), \tag{19}$$

where $a = b\sin\theta/\omega$, $\theta = \arctan[2b/(\omega + \omega_0)]$, and

$$\hat{R}(\theta) = \begin{pmatrix} \cos\frac{\theta}{2} & -\sin\frac{\theta}{2} \\ \sin\frac{\theta}{2} & \cos\frac{\theta}{2} \end{pmatrix}$$

is the rotation operator for the OZ axis. The transformation defined by (19) is, in fact, a transformation to the system of coordinates in which the component of the field $\mathbf{H}_1(t)$ rotating in the opposite direction is a constant, and the system is then rotated through the angle θ so that the new OZ' axis coincides with the direction of the new constant field.

Diagonalization of the transformed Hamiltonian then yields the eigenvalues in the new coordinate frame and, consequently, the frequency ω corresponding to the center of the magnetic resonance line ($p=0$) for $\omega_0 = \text{const}$:

$$\omega = \omega_0 + \frac{b^2}{\omega_0} + \frac{1}{4}\frac{b^4}{\omega_0^3} - \frac{35}{32}\frac{b^6}{\omega_0^5} + \ldots .$$

This expression[32] describes the Bloch-Siegert shift and is identical with the expansion obtained by Shirley[34] and deduced by Cohen-Tannoudji *et al.*[33] on the basis of the quantum-mechanical "dressed" model of the atom.

The transformation given by (19) can also be performed when $p \neq 0$, i.e., when there are multiphoton transitions connected with the presence of a component of the rf field rotating in the opposite direction. Calculations reported in Ref. 18 show that resonance effects should be observed whenever

$$(2p+1)\omega = \omega_0 + \frac{(2p+1)b^2}{p(p+1)\omega} \qquad (p > 0).$$

This condition was also reported by Winter,[25] who investigated multiphoton transitions both theoretically and experimentally.

The semiclassical interpretation of the Bloch-Siegert shift and the analysis of multiphoton transitions were given in a series of papers by Stenholm,[21] in which the eigenvalues of the Hamiltonian with higher-order corrections were obtained in the form of continued fractions. Stenholm's results are in good agreement with those discussed above. The semiclassical formalism has resulted in a completely satisfactory interpretation of other nonlinear effects as well (coherence resonances under transverse optical pumping,[35,56] mixing of quasidegenerate states by a strong nonresonant field,[21] change in the g-factor of an atom in the presence of an oscillating rf field,[37] and so on). These will be considered below.

The phenomenological Bloch equations written for the macroscopic magnetization of the spin system[35,37] can also be successfully used for the description of a broad range of nonlinear effects. This is hardly surprising since the Bloch equations are strictly valid for a system of $\frac{1}{2}$ spins, i.e., they are valid for all the cases mentioned above. Despite the appearance of certain mathematical difficulties, the application of the phenomenological equations is frequently more convenient because the final results are more readily interpreted. This is so because the observed physical variables are often directly related to the longitudinal

and transverse components of macroscopic magnetization.

B. Quantum theory of nonlinear effects

In the semiclassical theory, the effect of an rf electromagnetic field on atoms is regarded as a perturbation. The atomic system is regarded as a quantum-mechanical system, whereas the effect of the field on electrical charges and currents in the atom are described classically. Second quantization of the rf field, i.e., the introduction of rf quanta, enables us to remove this inconsistency. The evolution of the complete system consisting of the atom and the field is described by a time-independent Hamiltonian, and the analysis of the system is, at least in principle, simpler than the solution of the Schrödinger equation with a time-dependent Hamiltonian. Moreover, as we shall see, quantization of the field enables us to generalize the results to the case of arbitrary polarization of the field. It should be noted in this connection, that the formal analogy between field quantization and transformations of the form (19), which are designed to remove the time dependence from the Hamiltonian $\mathscr{H}(t)$, was noted in the early paper by Shirley.[34]

Within the framework of the quantum theory, we consider the physical system consisting of the atom and the radio-frequency photons that are absorbed and emitted by the atom in either real or virtual processes. The clear analogy between this formulation of the problem and the formulation used in quantum field theory, which takes vacuum fluctuations into account, enables us to borrow the essential terminology from the many-body problem and this, in turn, has led to the concept of the "dressed" atom[43] for the complete atom + photons system. Correspondingly the rf field interacting with the atom has been referred to as the "dressing" field.

1) *The concept of an atom "dressed" by the field.* The Hamiltonian $\hat{\mathscr{H}}$, describing a spin J atom and the quantized electromagnetic field interacting with it, is given by the sum

$$\hat{\mathscr{H}} = \hat{\mathscr{H}}_{at} + \hat{\mathscr{H}}_{rf} + V, \tag{20}$$

where $\hat{\mathscr{H}}_{at}$ is the Hamiltonian for the free atom, $\hat{\mathscr{H}}_{rf}$ is the Hamiltonian for the radiation field, and $\hat{V}$ is the operator representing the energy of interaction between the atoms and the radiation field.

When a constant magnetic field H_0 parallel to the OZ axis is present, the Hamiltonian $\hat{\mathscr{H}}_{at}$ can be represented in the case of interest to us by its Zeeman part:

$$\hat{\mathscr{H}}_{at} = \omega_0 \hat{J}_z, \tag{21}$$

where $\omega_0 = -\gamma H_0$ is the Larmor frequency, $\gamma = g\mu_B$, g is the Landé factor for the atom, and μ_B is the Bohr magneton. There are thus $2J+1$ states $|m\rangle$ of the atom in the field H_0 (m is the magnetic quantum number), the energy levels of which are separated by ω_0.

If the radiation field contains only one oscillation mode of frequency ω and polarization ε, the radiation-field Hamiltonian can be written in the form

$$\hat{\mathscr{H}}_{rf} = \omega \hat{a}^* \hat{a},$$

where $\hat{a}^+$ and $\hat{a}$ are the creation and annihilaton operators for the rf photons corresponding to this mode. It is well known that the effect of the application of these operators to the state $|n\rangle$ is as follows:

$$\hat{a}^+ |n\rangle = \sqrt{n+1}\,|n+1\rangle, \quad \hat{a}\,|n\rangle = \sqrt{n}\,|n-1\rangle.$$

Radio-frequency spectroscopy is mostly concerned with magnetic dipole transitions, so that we shall be interested in the operator for the magnetic rf field $\hat{\mathbf{H}}_1(t)$. It is well known[44] that, if the field $\mathbf{H}_1(t)$ is uniform within the limits of the specimen, then, in the dipole approximation, the operator $\hat{\mathbf{H}}_1$ is given by

$$\hat{\mathbf{H}}_1 = \mu (\hat{a}\boldsymbol{\varepsilon} + \hat{a}^+ \boldsymbol{\varepsilon}^*),$$

where μ is a constant having the sense of the "magnetic field of the photon" and equal to roughly 10^{-14} G.[17]

Real rf fields have a fully determined phase and, by virtue of the uncertainty principle, cannot be represented by the state $|n\rangle$ corresponding to a definite number of photons. They are, in fact, the coherent superposition of states $|\alpha\rangle$ with a different number of photons. Such states $|\alpha\rangle$ (coherent states) were introduced[45,56] as the eigenstates of the annihilation operator

$$\hat{a}\,|\alpha\rangle = \alpha\,|\alpha\rangle,$$

and can be written in the form

$$|\alpha(t)\rangle = \sum C_n(\alpha)\, e^{-in\omega t}\,|n\rangle,$$

where

$$|C_n(\alpha)|^2 = (\alpha^2)^n \frac{e^{-\alpha^2}}{n!}$$

is the possibility of finding n photons in the radiation field. It can be shown[45] that the mean number N of photons of the quantized field is related to the classical amplitude H_1 by

$$\alpha^2 = N = \frac{H_1^2}{4\mu^2}.$$

If we use the above value of μ, we can easily show that roughly 10^{28} rf photons per cm^3 are necessary to produce a field with an amplitude of 1 G. Of course, one can then speak of the exact value of the phase of the field. Since the field contains a large number of photons, we can neglect the change in this number when we consider transitions in the atomic system.

In view of the foregoing, the operator for the energy of interaction can be written in the form

$$\hat{V} = \lambda \hat{\mathbf{J}} (\hat{a}\boldsymbol{\varepsilon} + \hat{a}^+ \boldsymbol{\varepsilon}^*), \tag{22}$$

where

$$\lambda = -\frac{\gamma H_1}{2\sqrt{n}} = \frac{\omega_1(n)}{2\sqrt{n}}. \tag{23}$$

The structure of the operator $\hat{V}$ is very simple: it describes either the absorption or emission of an rf photon with given polarization.

The total Hamiltonian for the "dressed" atom can thus be written in the form

$$\hat{\mathscr{H}} = \omega_0 \hat{J}_z + \omega \hat{a}^* \hat{a} + \lambda \hat{\mathbf{J}} (\hat{a}\boldsymbol{\varepsilon} + \hat{a}^+ \boldsymbol{\varepsilon}^*). \tag{24}$$

The existence of a time-independent Hamiltonian en-

ables us to determine the energy levels of the physical system under investigation, which cannot be accomplished by means of the classical formalism. Each of these levels corresponds to the stationary state of the complete system, consisting of the atom and the rf field, and the dependence of the energy of the complete system on the magnetic field H_0 enables us to construct a level diagram which can then be used to interpret all the resonance phenomena in terms of level crossing and anticrossing.[38]

The analysis of the role of rf photons in processes involving their interaction with atoms is substantially simplified in this approach. In fact, anticrossings can exist only if states of the complete atom + field system, which have equal energies, are coupled, and this is accompanied by real absorption or emission of rf photons with the conservation of total energy and total angular momentum of the system. Conversely, level crossing suggests that the states of the complete system are not coupled and, consequently, there are no real transitions at the crossing point, so that the observed resonant phenomena arise as a result of virtual absorption or emission of rf photons.

The advantages of the quantum-mechanical formalism also become apparent in the course of the analysis of nonlinear phenomena associated with the interaction between the atomic system and a strong rf field. Thus, the solution of the equations of motion is usually accomplished within the framework of the semiclassical theory in the form of an expansion in powers of the perturbation, and this leads to expressions in the form of series whose convergence is not obvious; the quantum-mechanical theory enables us to perform a systematic study of the variation of the energy-level diagram for the complete system with increasing number of interacting photons. Radiative broadening effects and resonance line shifts are then amenable to a relatively simple treatment and the results are more easily interpreted.

Finally, it is important to note that the application of the quantum formalism has led to the prediction of a number of new effects which, because of the complexity of the mathematical formalism, appear to have previously escaped the attention of researchers using the semiclassical theory. In particular, predictions include the variation (and even the vanishing) of the atomic Landé factor for atoms interacting with strong rf fields,[39,40] a change in the hyperfine spectrum in the presence of the rf perturbation,[42] a transfer of coherence during spin exchange between different atoms in a strong rf field,[41] a change in the relaxation parameters of the atomic system when a nonresonant field is applied,[17] and many other physical phenomena.

2) *Atom "dressed" by a field. The case of spin* $J=\frac{1}{2}$. In this case, the next stage of the analysis becomes much simpler. Let us segregate in the Hamiltonian (24) the component corresponding to the absence of interaction

$$\hat{\mathcal{H}}=\hat{\mathcal{H}}_0+\hat{V}, \tag{25}$$

where

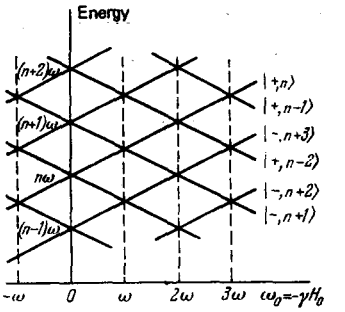

FIG. 1. Energy levels of an atom + photons system in the absence of interaction ($J=\frac{1}{2}$).

$$\hat{\mathcal{H}}_0=\omega_0\hat{J}_z+\omega\hat{a}^+\hat{a}. \tag{26}$$

The eigenstates of the Hamiltonian $\hat{\mathcal{H}}_0$ are the states $|m,n\rangle$ ($m=\pm\frac{1}{2}$) with energy $E_{m,n}=m\omega_0+n$, which can be represented by the state vectors $|\pm,n\rangle$. The corresponding energy-level diagram (Fig. 1) is a superposition of Zeeman diagrams for spin $J=\frac{1}{2}$ separated by an interval equal to ω, i.e., the energy of one photon.

It is clear from the diagram that there is an infinite number of level crossings for values of the field H_0 corresponding to $\omega_0=p\omega$, where $p\geq 0$.

The energy levels of the "dressed" atom can be obtained from the diagram by including the interaction $\hat{V}$ between the atom and the field. It is clear that it is sufficient to consider only part of the diagram near the energy $n\omega$ because the diagram remains practically invariant under a shift by the energy $p\omega$ if $p/n\ll 1$. The problem of finding the energy levels of the "dressed" atom is thus reduced to the diagonalization of the Hamiltonian (24).

In the special cases when the rf field rotates in the plane perpendicular to the constant field $\mathbf{H}_0$ ($\sigma^{\pm}$ polarization), or is polarized along its direction (π polarization), we have the possibility of exact diagonalization of the Hamiltonian $\hat{\mathcal{H}}$.

Thus, if the field $\mathbf{H}_1(t)$ has the σ^+ polarization, the Hamiltonian can be written in the form

$$\hat{\mathcal{H}}=\hat{\mathcal{H}}_0+\hat{V}_{\sigma+}=\omega_0\hat{J}_z+\omega\hat{a}^+\hat{a}+\lambda(\hat{J}_+\hat{a}+J_-\hat{a}^+). \tag{27}$$

In accordance with the selection rule $\Delta m+\Delta n=0$, the operator $\hat{V}_{\sigma+}$ can couple in pairs the states $|+,n\rangle$ and $|-,n+1\rangle$ of the unperturbed Hamiltonian $\hat{\mathcal{H}}_0$. The eigenstates $|+\rangle_n$ and $|-\rangle_n$ of the complete Hamiltonian $\hat{\mathcal{H}}$ are superpositions of the unperturbed state $|+,n\rangle$ and $|-,n+1\rangle$ of the form

$$\begin{aligned}|+\rangle_n&=-\sin\frac{\theta_n}{2}|+,\ n\rangle+\cos\frac{\theta_n}{2}|-,\ n+1\rangle,\\ |-\rangle_n&=\cos\frac{\theta_n}{2}|+,\ n\rangle+\sin\frac{\theta_n}{2}|-,\ n+1\rangle,\end{aligned} \tag{28}$$

where the angle θ_n is given by

$$\operatorname{tg}\theta_n=\frac{2\lambda\sqrt{n+1}}{\omega_0-\omega}=\frac{\omega_1(n)}{\omega_0-\omega}. \tag{29}$$

The position of the energy levels corresponding to the states $|\pm\rangle_n$ is given by

$$E_{\pm n}=\left(n+\frac{1}{2}\right)\omega\pm\frac{1}{2}\sqrt{(\omega_0-\omega)^2+\omega_1^2(n)}. \tag{30}$$

It is clear that they form hyperbolas which asymptotically approach the levels of the unperturbed Hamiltonian $\hat{\mathscr{H}}_0$. All crossings at the point $\omega=\omega_0$ that occur in the absence of interaction then transform into anticrossings, due to the resonant coupling between the states $|+,n\rangle$ and $|-,n+1\rangle$, whereas the remaining crossings $\omega_0=p\omega$ ($p\neq 1$) remain the crossings of the levels of the complete Hamiltonian $\hat{\mathscr{H}}$, shifted relative to their original position.

The anticrossings $\omega=\omega_0$ in the case of weak coupling $\omega_1\ll\omega$ occur for real absorption of one rf photon and may be looked upon as the ordinary magnetic resonance stimulated by the rotating rf field. The characteristic feature of this phenomenon is the absence of the radiative shift (the point of maximum approach of levels does not change its position as ω_1 increases) and the presence of radiative broadening of the resonance line (minimum distance between anticrossing levels proportional to ω_1).

The crossings $\omega_0=p\omega$ ($p\neq 1$) exhibit the radiative shift due to virtual absorption and emission of rf photons by the atom, which, according to the usual perturbation theory, is proportional to a quantity of the second order in $\hat{V}_{\sigma^+}$, i.e., ω_1^2/ω. The eigenstates of the dressed atom are then found to be very close to the states $|+,n\rangle$, $|-,n+p\rangle$ of the unperturbed Hamiltonian $\hat{\mathscr{H}}_0$, but the nonresonant interaction with the rf field changes their energy. As the amplitude ω_1 increases, the crossing points $\omega_0\approx 0$ and $\omega_0\approx 2\omega$ (points 2 and 3 in Fig. 2) are found to shift in the direction indicated by the arrows. They coalesce at $\omega=\omega_0$ for a certain definite value of ω_1 and then vanish. It is easily seen that they correspond to the coherence resonances[6] or the "off-diagonal" resonances.[7]

The second special case in which exact diagonalization of the Hamiltonian $\hat{\mathscr{H}}$ is possible corresponds to the rf field $\mathbf{H}_1(t)$ that is linearly polarized along $\mathbf{H}_0$ (π polarization). The Hamiltonian can then be written in the form

$$\hat{\mathscr{H}}=\omega_0\hat{J}_z+\omega\hat{a}^+\hat{a}+\lambda\hat{J}_z(\hat{a}+\hat{a}^+)=\hat{\mathscr{H}}_0+\hat{V}_\pi. \tag{31}$$

It is clear that the operator $\hat{V}_\pi$ couples only the states $|m,n\rangle$ with the states $|m,n+1\rangle$ and $|m,n-1\rangle$ with the same quantum number m. In fact, the π-polarized photon has no angular momentum and, consequently, its absorption or emission cannot modify the angular momentum of the atom. Hence, it follows that the energy eigenvalues of the dressed atom remain the same as the eigenvalues of the unperturbed Hamiltonian $\hat{\mathscr{H}}_0$ (Fig. 1), and all the level crossings shown in Fig. 1 remain in force independently of the amplitude of the rf field:

$$E_{\pm n}=n\omega\pm\frac{\omega_0}{2}. \tag{32}$$

The eigenvalues $|\pm\rangle_n$ of the Hamiltonian $\hat{\mathscr{H}}$ can be obtained in the form of the tensor product of the eigenvalues of the free atom $|\pm\rangle$ by the state $|\bar{n}_\pm\rangle$ of the rf field:

$$|\pm\rangle_n=|\pm\rangle|\bar{n}_\pm\rangle=|\pm\rangle\exp\left(\mp\lambda\frac{\hat{a}^+-\hat{a}}{2\omega}\right)|n\rangle=\sum_{n'}J_{n-n'}\left(\frac{\pm\omega_1}{2\omega}\right)|\pm,n'\rangle, \tag{33}$$

where $J_{n-n'}$ is the Bessel function of the first kind of order $n-n'$. This indicates that the eigenstates of the "dressed" atom are coherent superpositions of the states of the unperturbed Hamiltonian $\hat{\mathscr{H}}_0$ resulting from the virtual absorption or emission of rf photons by the atom. It is easily seen that the level crossing corresponds, in this case, to the phenomenon known as "parametric" resonance.[4,5] Its characteristic feature is the absence of radiative shifts or resonance line broadening.

In the general case of arbitrary polarization of the rf field, the diagonalization of the Hamiltonian $\hat{\mathscr{H}}$ becomes a much more complicated problem, whose solution can only be obtained by the method of successive approximations, or by numerical computation. However, in many cases that are of practical importance, it is possible to obtain approximate results that are in good agreement with experimental data. We shall return to this question later.

The "dressed"-atom method enables us not only to obtain all the results of the theory of optical orientation of atoms,[42,47] but also to achieve a quantitative analysis of the case where the optical pumping of the atoms is produced by high-intensity light. Depending on the particular features of the atomic system, the spectral composition of the radiation field, its polarization, and certain other illumination conditions, different methods have to be used to modify the thermodynamic equilibrium state of the system. A review of the pumping methods available can be found in the literature.[12b,16]

3) *Atom "dressed" by an optical frequency field.* In the case discussed above, we assumed that the atoms were in the ground state. We may now suppose that the operator $\hat{\mathscr{H}}_{at}$ is constructed so that it takes into account the finite lifetime $1/\Gamma$ of an atom in an excited state:

$$\hat{\mathscr{H}}=\hat{\mathscr{H}}_{at}+\hat{\mathscr{H}}_{rad}+\hat{\mathscr{H}}_1=\hat{\mathscr{H}}_0+\hat{\mathscr{H}}_1. \tag{34}$$

Here $\hat{\mathscr{H}}_{at}$ and, consequently, $\hat{\mathscr{H}}$ as well, are non-Hermitian operators.

Suppose that there are only two Zeeman sublevels in the ground state of the atom and in the excited state coupled to it by the optical-frequency field. The eigenstates of $\hat{\mathscr{H}}_0$ will then be $|\pm m,n\rangle$ and $|\pm l,n\rangle$. In the absence of the external magnetic field, all the levels are doubly degenerate. Level crossing occurs for $\omega_0=p\omega$ ($p=1,2,\ldots$). It is clear from Fig. 3 that single-photon transitions correspond to crossings at $p=1$.

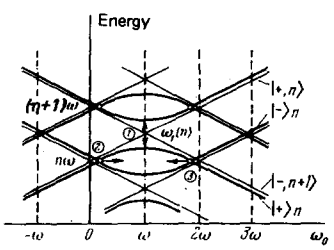

FIG. 2. Energy levels of an atom "dressed" by an rf field with σ^+ polarization ($J=\frac{1}{2}$).

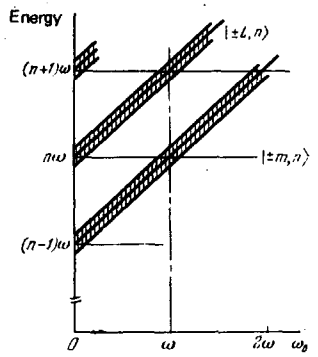

FIG. 3. Energy levels corresponding to the Hamiltonian $\hat{\mathscr{H}}_0$ in the absence of interaction with an optical field. The natural level width is conventionally indicated by the shaded band.

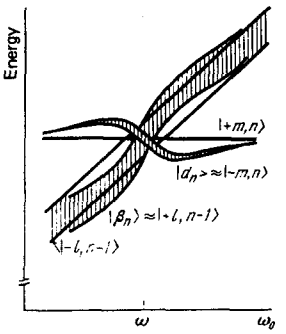

FIG. 4. Energy-level diagram for the Hamiltonian $\hat{\mathscr{H}}$ near the points $\omega=\omega_0$ for a "dressing" optical field of low intensity. The ground state $|-m,n\rangle$ is shifted and broadened.

As an example, consider the change in the energy spectrum of atoms under conditions of optical pumping. If this pumping is produced by a beam of circularly (σ^+) polarized rays of frequency ω, the interaction Hamiltonian $\hat{\mathscr{H}}_1$ couples the levels $|-m,n\rangle$ and $|+l,n-1\rangle$, and the corresponding transition matrix elements can be written in the form

$$\langle +l,\ n-1|\hat{\mathscr{H}}_1|-m,\ n\rangle = iv\sqrt{n}. \tag{35}$$

The levels of the states $|+m,n\rangle$ and $|-l,n\rangle$ are not coupled to other levels and are the eigenstates of the complete Hamiltonian $\hat{\mathscr{H}}$. It can be shown[44] that the eigenstates can also be linear combinations of $|-m,n\rangle$ and $|+l,n-1\rangle$ of the form

$$\begin{aligned}|\alpha_n\rangle &= \cos\frac{\theta}{2}|-m,\ n\rangle + i\sin\frac{\theta}{2}|+l,\ n-1\rangle,\\ |\beta_n\rangle &= i\sin\frac{\theta}{2}|-m,\ n\rangle + \cos\frac{\theta}{2}|+l,\ n-1\rangle,\end{aligned} \tag{36}$$

where θ is given by

$$\operatorname{tg}\theta = \frac{2v\sqrt{n}}{\omega-\omega_0+i(\Gamma/2)}. \tag{37}$$

We note that, since $\hat{\mathscr{H}}$ is not-Hermitian, the states $|\alpha_n\rangle$ and $|\beta_n\rangle$ are not orthogonal. The corresponding eigenvalues are given by

$$\begin{aligned}E_n^\alpha - i\frac{\Gamma_n^\alpha}{2} &= (n-1)\omega + \frac{\omega+\omega_0}{2} - i\frac{\Gamma}{4} + \sqrt{\left(\frac{\omega-\omega_0}{2}+i\frac{\Gamma}{4}\right)^2+nv^2},\\ E_n^\beta - i\frac{\Gamma_n^\beta}{2} &= (n-1)\omega + \frac{\omega+\omega_0}{2} - i\frac{\Gamma}{4} - \sqrt{\left(\frac{\omega-\omega_0}{2}+i\frac{\Gamma}{4}\right)^2+nv^2}.\end{aligned} \tag{38}$$

If the intensity of the optical beam is low, i.e., $nv^2 \ll (\Gamma/4)^2$, it follows from (38) that

$$\begin{aligned}E_n^\alpha &= n\omega + nv^2\frac{\omega-\omega_0}{(\omega-\omega_0)^2+(\Gamma/2)^2}, & \frac{\Gamma_n^\alpha}{2} &= nv^2\frac{\Gamma/2}{(\omega-\omega_0)^2+(\Gamma/2)^2},\\ E_n^\beta &= (n-1)\omega+\omega_0 - nv^2\frac{\omega-\omega_0}{(\omega-\omega_0)^2+(\Gamma/2)^2}, & \frac{\Gamma_n^\beta}{2} &= \frac{\Gamma}{2} - nv^2\frac{\Gamma/2}{(\omega-\omega_0)^2+(\Gamma/2)^2},\end{aligned} \tag{39}$$

and

$$\begin{aligned}|\alpha_n\rangle &= |-m,\ n\rangle + i\frac{v\sqrt{n}}{\omega-\omega_0+i(\Gamma/2)}|+l,\ n-1\rangle,\\ |\beta_n\rangle &= |+l,\ n-1\rangle + i\frac{v\sqrt{n}}{\omega-\omega_0+i(\Gamma/2)}|-m,\ n\rangle.\end{aligned} \tag{40}$$

It is clear from (39) and (40) that, at low optical-field intensities, the eigenstates of the "dressed" atom, i.e., the states $|\alpha_n\rangle$ and $|\beta_n\rangle$, are not very different from the unperturbed states $|-m,n\rangle$ and $|+l,n-1\rangle$. The energy spectrum of the atom "dressed" by the weak optical field is shown in Fig. 4. It is clear that the levels continue to cross even in the presence of perturbation. Although, according to (39) and (40), the energy corrections and the level shifts are small, they are easily observed in the ground state because the intrinsic level width in this state is small. It is shown in the literature[42,47] that the removal of level degeneracy, and the broadening of the levels by some tens of hertz, can be easily observed in the ground state of the atoms for large relaxation times.

In the other limiting case of a high-intensity optical-pumping field, $nv^2 \gg (\Gamma/4)^2$, we obtain the following expressions for the eigenvalues corresponding to the states of the "dressed" atom:

$$\left.\begin{aligned}E_n^\alpha &= (n-1)\omega + \frac{\omega+\omega_0}{2} + \sqrt{\left(\frac{\omega-\omega_0}{2}\right)^2+nv^2},\\ \frac{\Gamma_n^\alpha}{2} &= \frac{\Gamma}{4}\left(1-\frac{\omega-\omega_0}{\sqrt{(\omega-\omega_0)^2+4nv^2}}\right),\\ E_n^\beta &= (n-1)\omega + \frac{\omega+\omega_0}{2} - \sqrt{\left(\frac{\omega-\omega_0}{2}\right)^2+nv^2},\\ \frac{\Gamma_n^\beta}{2} &= \frac{\Gamma}{4}\left(1+\frac{\omega-\omega_0}{\sqrt{(\omega-\omega_0)^2+4nv^2}}\right).\end{aligned}\right\} \tag{41}$$

The energy spectrum of the "dressed" atom is shown in Fig. 5. In this case, i.e., in a strong "dressing" field, we have the anticrossing of levels. Moreover, the eigenstates of the "dressed" atom correspond to strong mixing of the unperturbed states $|-m,n\rangle$ and $|+l,n-1\rangle$:

$$\begin{aligned}|\alpha_n\rangle &= \cos\frac{\theta'}{2}|-m,\ n\rangle + i\sin\frac{\theta'}{2}|+l,\ n-1\rangle\\ |\beta_n\rangle &= i\sin\frac{\theta'}{2}|-m,\ n\rangle + \cos\frac{\theta'}{2}|+l,\ n-1\rangle,\end{aligned} \tag{42}$$

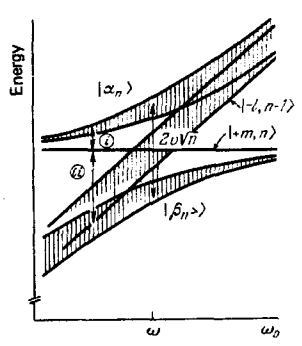

FIG. 5. Energy-level diagram for the Hamiltonian $\hat{\mathscr{H}}$ near $\omega=\omega_0$ for a high-intensity "dressing" optical field.

where

$$\operatorname{tg}\theta' = \frac{2v\sqrt{n}}{\omega - \omega_0}, \quad 0 \leqslant \theta' \leqslant \pi. \tag{43}$$

The expressions given by (42) and (43) will readily yield the expression for the amplitude of the states $|m,n\rangle$ as a function of $|\alpha_n\rangle$ and $|\beta_n\rangle$. Since the state $|m,n\rangle$ is the eigenstate of the complete Hamiltonian $\hat{\mathscr{H}}$, we can simultaneously define the amplitudes for the states $|-m,n\rangle$ and $|+m,n\rangle$. However, for a complete determination of the relationship between the amplitudes for these states and, consequently, in order to determine the orientation of the atoms in the ground state, we must, at the same time, take into account the relaxation mechanisms.

The change in the energy spectrum of the "dressed" atom as compared with the spectrum of the "bare" atom leads to a particular shape of the magnetic-resonance spectrum that is observed for atoms "dressed" by a strong field (Fig. 6). The transitions indicated by *i* in Fig. 5 occur between two ground-state levels, one of which is perturbed by the optical-frequency field. These are the transitions between the states $|+m,n\rangle$ and $|\alpha_n\rangle \sim |-m,n\rangle$. The resonance line is narrow and the transition frequency is equal to the shift of the $|-m,n\rangle$ level in the "dressing" field. For the *ii* transition, the linewidth is roughly Γ. This transition takes place between the $|+m,n\rangle$ and $|\beta_n\rangle \approx |+l,n-1\rangle$ states, i.e., it is a two-photon transition. The atom absorbs an optical photon of frequency ω and, at the same time, emits an rf photon of frequency $\Omega \approx |\omega - \omega_0|$.

The "dressed"-atom model is capable of providing a physical interpretation and a quantitative analysis of all the resonance effects observed so far during the interaction between atoms and radiofrequency fields of arbitrary configuration. Whether or not a particular signal described by the above formalism can be observed will largely depend on the conditions under which the atomic system is "prepared" for measurement, and by the conditions under which the observables are recorded. Many of the phenomena described below have been observed in experiments performed by the usual methods of rf spectroscopy (ESR and NMR spectroscopy), i.e., with the aid of radio-frequency detection

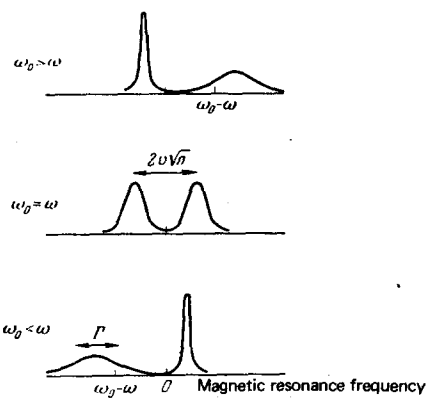

FIG. 6. Magnetic resonance spectrum for an atom interacting with an optical field. Resonances corresponding to the transition *i* and *ii* shown in Fig. 5.

of the signals, with the angular momenta of the atoms oriented by the application of a constant magnetic field. Other phenomena became accessible to observation only through the application of optical methods of detection of magnetic resonance under conditions of optical pumping. The application of optical pumping methods, and the flexibility of these methods, have ensured that the entire range of resonance phenomena and nonlinear effects has not been observed. We shall therefore largely confine our attention to experiments performed under conditions under which the atomic system is prepared by an optical method and an optical method is used to observe the quantities under consideration.

Without going into details of the theoretical analysis of the interaction between an atomic system and resonance optical radiation,[46,47] we draw the attention of the reader to some of the features of the optical methods of rf spectroscopy that are essential for the understanding of the ensuing review.

It is well known[12] that circularly polarized resonance radiation of optical frequency can be used to *orient* the angular momenta of atoms in a system. If this radiation is directed along the constant magnetic field $\mathbf{H}_0$[3)] (longitudinal optical pumping), the atomic system will have a stationary distribution of level *populations* after a certain time, which will be different from the equilibrium distribution. If, on the other hand, the pumping beam is perpendicular to the magnetic field $\mathbf{H}_0$ (transverse pumping), then, although the level-population distribution remains, as before, close to the equilibrium distribution, Zeeman coherence is continuously introduced into the system (nonzero off-diagonal elements of the density matrix appear), i.e., transverse orientation of the atomic angular momenta is produced.[47] By recording the degree of absorption of circularly polarized resonance light by the atoms, it is possible to determine the component of the angular momentum of the atoms in the direction of propagation of this radiation, averaged over the entire ensemble.

The crossing of the levels of the "dressed" atom mentioned above can be observed[17] under conditions of transverse optical orientation (coherent excitation of crossing levels), whereas the corresponding anticrossing can be detected only in the presence of longitudinal optical pumping (a population difference must be set up between the anticrossing levels).

3. NONLINEAR INTERACTION BETWEEN AN ATOM AND A ROTATING RF FIELD

The interaction between an atomic system and a rotating rf field has been examined both theoretically and experimentally in situations differing by the method used to prepare the atomic system (longitudinal and transverse optical pumping) and by the orientation of the plane of rotation of the rf field relative to the constant field $\mathbf{H}_0$. In all the cases described below, the

[3)] As before, we are assuming that the direction of the field $\mathbf{H}_0$ is the direction of the quantization axis.

magnetic-resonance signal can be recorded either by optical detection of the pumping radiation transmitted by the atomic system, the intensity of which may, in general, contain alternating components with frequencies equal to the frequency of $\mathbf{H}_1(t)$ or its multiples or by measuring the total intensity of the fluorescence radiation emitted in a previously chosen direction.

The simplest cases correspond to the configurations shown in Fig. 7. It follows from the discussion given in the last section that the energy levels of the atom "dressed" by the field $\mathbf{H}_1(t)$ rotating around the direction of $\mathbf{H}_0$ (see Fig. 2) exhibit an infinite set of anticrossings at $\omega = \omega_0$ and crossings at $\omega = p\omega_0$ ($p \neq 1$). Under the conditions of longitudinal pumping, one observes only the anticrossings corresponding to the ordinary magnetic resonance stimulated by the rotating field if the sense of this rotation is the same as that of the Larmor precession of the atoms around the field $\mathbf{H}_0$. The longitudinal pump (Fig. 7a) continuously populates one of the levels of the atom $|\pm\rangle$ (depending on the polarization σ^+ or σ^- of the pump radiation), which is equivalent to preparing the atom + field system either in the $|+,n\rangle$ state or the $|-,n\rangle$ state. Expressions (28)–(30) can readily be used to calculate the probability of finding the system in a state differing from the original state. To be specific, let us suppose that the atom is continuously prepared to be in the state $|-,n\rangle$, and that its lifetime in this state due to relaxation processes is $1/\Gamma$. The mean probability of transitions in the atom from the state $|-\frac{1}{2}\rangle$ to the state $|+\frac{1}{2}\rangle$ is

$$\bar{P}_{-,\,n;\,+,\,n-1} = \frac{\omega_1^2}{(\omega_0-\omega)^2+\omega_1^2+\Gamma^2}. \tag{44}$$

This expression describes the magnetic resonance line shape in the rotating field. It is characterized by the absence of a shift for any values of the amplitude ω_1 and, in the linear approximation ($\omega_1 \ll \Gamma$), the signal is proportional to the square of the amplitude, ω_1^2. The interaction with the radio-frequency field becomes manifestly nonlinear only when $\omega_1 \gtrsim \Gamma$, and is seen as a broadening of the resonance line by the strong rf field (saturation effect). These well-known properties of ordinary magnetic resonance are observed in ESR or NMR experiments.

A completely different situation occurs in the case of transverse pumping (Fig. 7b). This can no longer lead to a population difference between the Zeeman sublevels of an atom of spin $\frac{1}{2}$, but can maintain the atomic system in the state $|\psi_0\rangle$ in the form of a superposition of the states $|+\frac{1}{2}\rangle \equiv |\pm\rangle$ and $|-\frac{1}{2}\rangle \equiv |-\rangle$:

$$|\psi_0\rangle = C_+ |+\rangle + C_- |-\rangle,$$

i.e., it produces in it transverse components of angular momentum, precessing around the field $\mathbf{H}_0$.

The energy-level diagram of the "dressed" atom remains the same as before (Fig. 2), but the transverse character of the pump implies that it is not possible to observe the resonance corresponding to the anticrossing of the levels. Conversely, coherence resonances corresponding to crossings become observable.

Resonance phenomena of this kind were first described by Aleksandrov and Sokolov,[7] who referred to them as "off-diagonal" resonances. They were later examined by Le Dourneuf *et al.*,[48,49] who called them coherence resonances. The phenomenological theory of these phenomena was developed by Malyshev and Novikov[8] and Tsukada *et al.*[9]

It follows from the level diagram (Fig. 2) that these resonances correspond to the crossing of the levels of the "dressed" atom designated by the numbers 2 and 3. When the amplitude of the rf field is small ($\omega_1 \ll \omega$), one observes two resonances ($\omega_0 \approx 2\omega$ and $\omega_0 \approx 0$). As the amplitude of ω_1 increases, the resonances shift, approach one another, and merge into a single line which can be observed for $\omega_0 = \omega$ when $\omega_1 = \omega$. Figure 8 shows the coherence resonances observed in a system of Hg^{199} atoms ($I = \frac{1}{2}$) for different values of the ratio ω_1/ω.[48] The theoretical position of the resonances[17] is given by

$$\omega_0 = \omega \pm \sqrt{\omega^2 - \omega_1^2}. \tag{45}$$

This dependence is represented by the semicircle centered on $\omega_0 = \omega$ in Fig. 9. The resonances are fun-

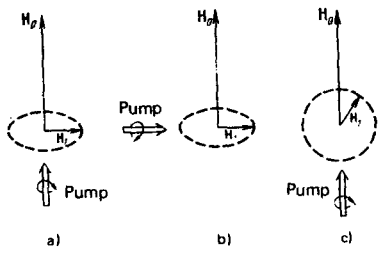

FIG. 7. Possible mutual dispositions of the fields $\mathbf{H}_0, \mathbf{H}_1(t)$, and the optical pump beam.

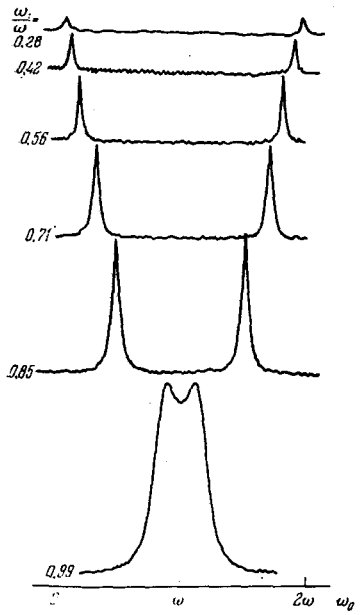

FIG. 8. Coherence resonances in Hg^{199} atoms "dressed" by a field $\mathbf{H}_1(t)$ rotating at right-angles to the direction of $\mathbf{H}_0$.[48]

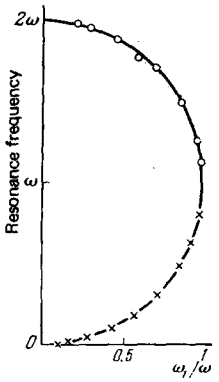

FIG. 9. Position of coherence resonances as a function of the amplitude of the radio-frequency field. Circles show experimental results for Hg[199].[48]

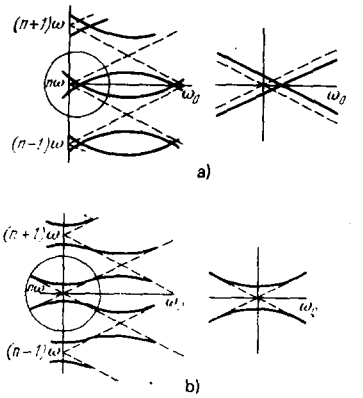

FIG. 10. Energy-level diagram for an atom "dressed" by an rf field rotating (a) in the plane perpendicular to $\mathbf{H}_0$ and (b) about the direction perpendicular to $\mathbf{H}_0$ near $H_0 \approx 0$.

damentally different from the usual magnetic resonances both by their physical origin and by their properties. For example, the resonance observed near $\omega_0 = 2\omega$ is in no way related to real absorption or emission of rf photons by the atom because it is then impossible to satisfy simultaneously the conservation of energy and of angular momentum. Nevertheless, it is quite clear that the resonance is due to the interaction between the atom and the rf field. Detailed analysis shows[17] that the existence of coherence resonances in the above situation can be explained only by virtual absorption and emission by the atoms of photons of the $\mathbf{H}_1(t)$, which leads to nonresonant coupling between the states $|-, n+1\rangle$ and $|+, n+1\rangle$, on the one hand, and the states $|+, n\rangle$ and $|-, n\rangle$, which contain an equal number of photons, on the other hand.

Since the physical processes underlying coherence resonances and ordinary magnetic resonance are different, the main characteristics of these processes will also be different. In fact, coherence resonances can be observed only under coherent excitation of crossing levels of the "dressed" atom (transverse pump), whereas noncoherent excitation (longitudinal) is necessary for the observation of ordinary resonance. Moreover, the intensity of the signal corresponding to a coherence resonance in the "dressed" atom in the approximation of order $2p$ in the parameter ω_1/ω is proportional to the ratio ω_1/ω, whereas, in the case of ordinary resonance, the corresponding quantity, calculated in the same approximation, is proportional to $\omega_1^{2p}/\Gamma^2\omega^{2p-2}$. In other words, the intensity of the coherence-resonance signal is smaller by a factor of Γ^2/ω^2 as compared with the intensity of the ordinary-resonance signal.

The width of coherence resonances is determined only by the natural widths and the relative slope of the crossing levels, whereas the existence of real transitions in the case of ordinary resonance reduces the lifetime of the atomic state and leads to radiative line broadening. The absence of saturation in coherence resonances leads to the fact that they can be observed for rf field amplitudes for which ordinary resonance is completely saturated.

Equally interesting results have been obtained by analyzing the resonances observed for low values of the constant field ($H_0 \sim 0$) in the presence of a rotating rf field. It will be useful to consider the two experimental situations illustrated in Figs. 7b and 7c. The corresponding segments of the level diagram of the "dressed" atom are shown in Figs. 10a and 10b.

As already noted, in the first case (Fig. 10a), the level crossing persists but shifts away from $\omega_0 \sim 0$. In the second case (Fig. 10b), the polarization of the rf field is a coherent superposition of σ^+, σ^-, and π polarization, so that the interaction $\hat{V}$ can couple any states of the unperturbed Hamiltonian $\hat{\mathscr{H}}_0$ and, consequently, all the crossings of the levels of the Hamiltonian $\hat{\mathscr{H}}_0$ must transform into anticrossings of the levels of the "dressed" atom.

It is not difficult to notice the analogy between the above diagrams and the behavior of the energy levels of the free atom with spin 1/2 in the presence of the field $\mathbf{H}_0$ and an additional constant field $\mathbf{H}_f$ either parallel (Fig. 10a) or perpendicular (Fig. 10b) to $\mathbf{H}_0$. This analogy enables us to describe the influence of the rotating rf field by introducing a fictitious constant field $\mathbf{H}_f$ perpendicular to the plane of rotation of the field $\mathbf{H}_1(t)$. We note that this is only valid for values of $\mathbf{H}_0$ approaching zero. It is readily shown[17] that, if $\omega_1 \ll \omega$, the fictitious field is given by

$$H_f = \frac{\gamma H_1^2}{2\omega}. \tag{46}$$

Experimental results[49] confirm this analogy. In fact, the coherence resonance observed near $H_0 = 0$ in the presence of an rf field rotating about $\mathbf{H}_0$ exhibits an appreciable shift when the amplitude of the field H_1 is increased (Fig. 8). If the signal is detected by measuring the static component of the magnetization of the spin system (for example, by measuring the fluorescence intensity), it can be interpreted as the Hanle-effect signal due to the free atom, shifted in the presence of the fictitious field $\mathbf{H}_f$ parallel to $\mathbf{H}_0$. Figure 11a shows a series of signals of this kind obtained for different values of the amplitude of the rotating field H_1. It is clear from Fig. 11b that, for small amplitudes, $\omega_1/\omega \ll 1$, the Hanle-effect shift is satisfactorily described by (46).

The situation corresponding to the mutual disposition of the H_0 and $H_1(t)$ fields shown in Fig. 7c has also been

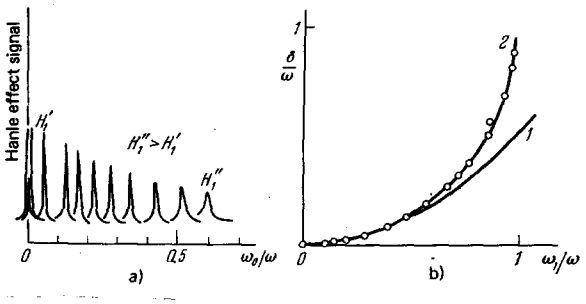

FIG. 11. (a) Hanle-effect signal in the presence of the field $H_1(t)$ $H_1(t)$ rotating around the direction of H_0, obtained for different values of H_1; (b) relative shift of the Hanle effect, δ/ω, as a function of the amplitude of the rotating field [curve 1 corresponds to (46); curve 2 is experimental[49]].

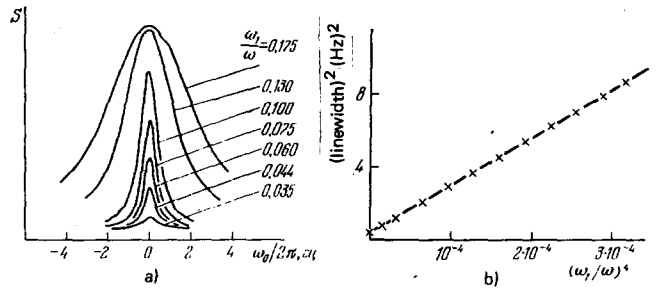

FIG. 12. Second-order magnetic resonance in zero magnetic field during the interaction of an atom with a rotating field $H_1(t)$[50]: (a) resonance curves for different amplitudes of rotating field; (b) square of resonance linewidth as a function of the fourth power of the amplitude of the field $H_1(t)$.

investigated.[48,50] Here, one observes a new and interesting type of resonance accompanying the anticrossing of the levels of the "dressed" atom in the field $H_0=0$. The rf field $H_1(t)$ can be written as a coherent superposition of photons with $\sigma^{\pm}$ and π polarizations, so that the level diagram of the "dressed" atom near $H_0=0$ shows an anticrossing because the field $H_1(t)$ produces an *a priori* coupling of the states $|+,n\rangle$ and $|-,n\rangle$. Since the states $|-\frac{1}{2}\rangle$ and $|+\frac{1}{2}\rangle$ of the atom in zero field have the same energy, conservation of energy of the complete system demands that the number of absorbed rf photons be equal to the number of emitted photons in a cycle of interaction between the atom and the field $H_1(t)$. It is easily seen that the transition $|-\frac{1}{2}\rangle - |+\frac{1}{2}\rangle$ can be executed with nonzero probability in the lowest-order approximation if there is (a) absorption of a σ^+ photon followed by the emission of a π photon, (b) absorption of a π photon with the subsequent emission of a σ^- photon, (c) emission of a π photon with the subsequent absorption of a σ^+ photon, and (d) emission of a σ^- photon with the subsequent absorption of a π photon. The angular momentum is conserved in all these cases. All the transitions are resonance transitions for $H_0=0$. Since, for a given polarization of the field $H_1(t)$, the $\sigma^{\pm}$ and π photons are coherent, the above four processes can interfere, giving a nonzero probability amplitude for the transition $|-\frac{1}{2}\rangle - |+\frac{1}{2}\rangle$, which is proportional to H_1^2 (second-order processes). Thus, independently of the frequency ω of the field $H_1(t)$, one observes a resonance at $H_0=0$, whose magnitude is proportional to H_1^4 and whose width is proportional to H_1^2. The second-order processes producing this resonance do not influence the total energy of the rf field, but do change its polarization.

Figure 12 shows a series of resonance curves in zero field, obtained in an experiment with Hg^{199} atoms ($I=\frac{1}{2}$) for different values of the rf field amplitude.[50] It is clear that there is complete agreement between the experimental results and the theoretical predictions.

It is interesting to emphasize that the above second-order resonance can also be interpreted by introducing the fictitious constant field H_f, mentioned above. The fictitious field is perpendicular to the field H_0, which is scanned across its zero value and has "zero" frequency. It couples the states $|+\frac{1}{2}\rangle$ and $|-\frac{1}{2}\rangle$ and induces transitions between them, whose probability is a maximum when $H_0=0$. According to (46), the direction of the fictitious field H_f depends on the sign of the gyromagnetic ratio of the atom. This has been used successfully to investigate the transfer of coherence between hyperfine levels of the ground state of alkali-metal atoms.[17]

4. NONLINEAR INTERACTION BETWEEN AN ATOMIC SYSTEM AND AN OSCILLATING RF FIELD

The phenomena which we are considering depend on the mutual orientation of the oscillating rf field $\mathbf{H}_1(t)$ and the constant field $\mathbf{H}_0$. We note particularly two different experimental situations: (a) the field $\mathbf{H}_1(t)$ is parallel to $\mathbf{H}_0$, and (b) $\mathbf{H}_1(t)$ is perpendicular to H_0.

A. $\mathbf{H}_1(t)$ parallel to $\mathbf{H}_0$

This case corresponds to the π polarization of the rf field and admits of an exact diagonalization of the Hamiltonian $\hat{\mathcal{H}}$ [Eq. (31)]. The qualitative theoretical analysis given above (Secs. 2–1) has shown that the level diagram for the "dressed" atom has the form indicated in Fig. 1. The observed resonance phenomena corresponding to the level crossing are frequently combined under the title "parametric resonance"[4] and have been investigated in detail in the literature.[4,5] A detailed theoretical analysis of the parametric resonance can also be found in the work of Polonsky and Cohen-Tannoudji[51] and Polonsky.[52] We shall only reproduce the final expression obtained within the framework of the "dressed"-atom formalism[17] and corresponding to the detection of the mean value $\langle S_x\rangle$ of the component of the spin angular momentum. We recall that the fields $\mathbf{H}_0$ and $\mathbf{H}_1(t)$ are, in this case, directed along the z axis of the laboratory system of coordinates, and the circularly polarized light of the pump propagates along the x axis (transverse pump). The expression for the optical signal proportional to $\langle S_x\rangle$ is

$$\langle S_x\rangle=\langle S_x\rangle_0\left\{\left[J_n\left(\frac{\omega_1}{\omega}\right)J_{n+n'}\left(\frac{\omega_1}{\omega}\right)+J_n\left(\frac{\omega_1}{\omega}\right)J_{n-n'}\left(\frac{\omega_1}{\omega}\right)\right]\frac{\Gamma^2\cos n'\omega t}{\Gamma^2+(n\omega+\omega_0)^2}\right.$$
$$\left.-\left[J_n\left(\frac{\omega_1}{\omega}\right)J_{n+n'}\left(\frac{\omega_1}{\omega}\right)-J_n\left(\frac{\omega_1}{\omega}\right)J_{n-n'}\left(\frac{\omega_1}{\omega}\right)\right]\frac{\Gamma(\omega_0+n\omega)\sin n'\omega t}{\Gamma^2+(n\omega+\omega_0)^2}\right\}, \quad (47)$$

where $\langle S_x\rangle_0$ is the mean value of the component of the angular momentum of the atomic system in the absence of the external field, and Γ is the effective width of the intersecting levels taking optical relaxation into account. It is clear from (47) that the detected signal is propor-

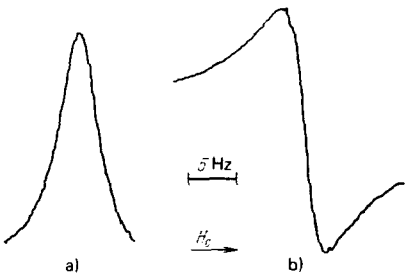

FIG. 13. Parametric resonance in Hg^{199} atoms in phase with the rf field and for a 90° phase shift.[52]

tional to the light flux transmitted by the system and modulated at the frequencies $n'\omega$. The modulation amplitude exhibits resonance behavior for $\omega_0 = -n\omega$. A particular feature of these (parametric) resonances is the absence of radiative shift and broadening for any value of the amplitude of the rf field $H_1(t)$.

Figure 13 shows the signals corresponding to the parametric resonance $\omega_0 = \omega$, obtained by recording the modulation of transmitted light at frequency ω in phase with the rf field and for a 90° phase difference.

Depending on the phase of the modulation, the resonance signal takes the shape of the absorption curve or the dispersion curve, in accordance with (47). Figure 14a shows the amplitude of the different harmonics of the signal as a function of the ratio ω_1/ω. The theoretical curves correspond to expressions of the form

$$J_n\left(\frac{\omega_1}{\omega}\right) J_{n+n'}\left(\frac{\omega_1}{\omega}\right) + J_n\left(\frac{\omega_1}{\omega}\right) J_{n-n'}\left(\frac{\omega_1}{\omega}\right),$$

appearing in (47), where $n = -1$ and $n' = 1$, 3, and 5. The dependence on the parametric-resonance linewidth on the amplitude of the rf field, shown in Fig. 14b, clearly indicates the absence of radiative broadening, We note for comparison that the width of the ordinary resonance, stimulated by the field $\mathbf{H}_1(t)$ rotating around $\mathbf{H}_0$ for the value of the amplitude indicated by the arrow in Fig. 14b, is about 4000 Hz. This confirms the connection mentioned above between parametric resonance and virtual rf transitions in the system, stimulated by the interaction with the field $\mathbf{H}_1(t)$.

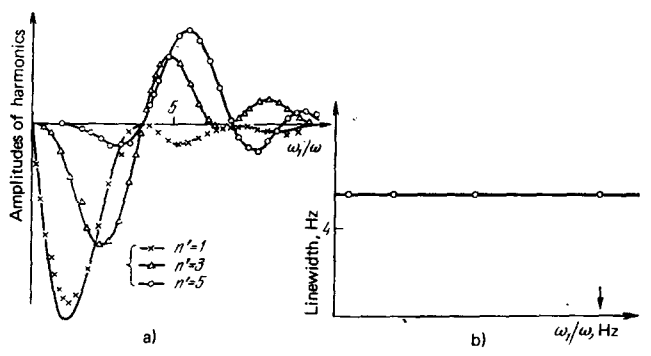

FIG. 14. (a) Amplitude of harmonics of the parametric-resonance signal as a function of the amplitude of the field $H_1(t)$ (solid curves—theory; open circles—experiment); (b) parametric resonance linewidth as a function of the amplitude of the field $H_1(t)$.[51]

B. $\mathbf{H}_1(t)$ perpendicular to $\mathbf{H}_0$

An rf field oscillating in the direction perpendicular to the field $\mathbf{H}_0$ (σ polarization) can be written as a coherent superposition of σ^+ and σ^- photons. The interaction Hamiltonian $\hat{V}_\sigma$ for this case can be written in the form

$$\hat{V}_\sigma = \lambda \hat{J}_x (\hat{a} + \hat{a}^+) = \frac{\lambda}{2}[\hat{a}\hat{J}_+ + \hat{a}^+\hat{J}_-] + \frac{\lambda}{2}[\hat{a}\hat{J}_- + \hat{a}^+\hat{J}_+]. \quad (48)$$

The perturbation $\hat{V}_\sigma$ is, therefore, the sum of two terms corresponding to the two rotating components of the rf field. Moreover, the state $|m,n\rangle$ of the unperturbed Hamiltonian $\hat{\mathcal{H}}_0$ is coupled to the states $|m+1, n-1\rangle$ and $|m-1, n+1\rangle$, because of the presence of the σ^+ component of the field, and to the states $|m+1, n+1\rangle$ and $|m-1, n-1\rangle$, because of the presence of the σ^- component. Since, in general, only those matrix elements of the operator $\hat{V}_\sigma$ which satisfy the selection rules $\Delta n = \pm 1$ and $\Delta m = \pm 1$ are nonzero, any pair of states will be coupled when Δn and Δm have the same parity. Thus, for spin $\frac{1}{2}$, the state $|+,n\rangle$ will be coupled to the states $|+, n \pm 2p\rangle$ and the states $|-, n \pm (2p+1)\rangle$, and the diagonalization of the complete Hamiltonian can be carried out only approximately.

If the coupling constant is small ($2\lambda\sqrt{n} = \omega_1(n) \ll \omega$), the interaction $\hat{V}_\sigma$ may be looked upon as a perturbation. Part of the level diagram of the "dressed" atom of spin $\frac{1}{2}$, obtained within the framework of this assumption, is shown in Fig. 15. It is clear that, in this case, the coupling between the $|+,n\rangle$ and $|-, n+2p+1\rangle$ states, which corresponds to the absorption of $2p+1$ photons by the atom, transforms odd-level crossings corresponding to the Hamiltonian $\hat{\mathcal{H}}_0$ into the anitcrossing of the levels of the "dressed" atom. Conversely, the states $|+,n\rangle$ cannot be coupled by the rf field to the states $|-, n+2p\rangle$ because the differences $\Delta m = 1$ and $\Delta n = 2p$ have different parities. It follows that *even* crossings of the levels of the Hamiltonian remain as crossings of the levels of the "dressed" atom. Both crossings and anticrossings may experience radiative shifts connected with nonresonance processes involving virtual absorption and emission of rf photons. Only the crossing in zero field will not exhibit a shift because of the symmetry of the perturbing Hamiltonian.

1) *Multiphoton transitions and the Bloch Siegert effect.* As before, resonance phenomena near the anticrossing of levels may be interpreted as ordinary magnetic resonance, whereas phenomena near level cros-

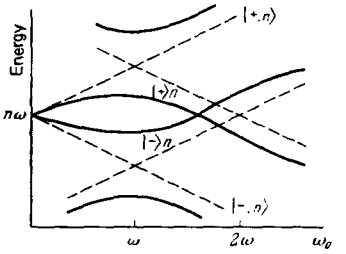

FIG. 15. Energy levels of the atom ($J = \frac{1}{2}$) "dressed" by a field with σ polarization.

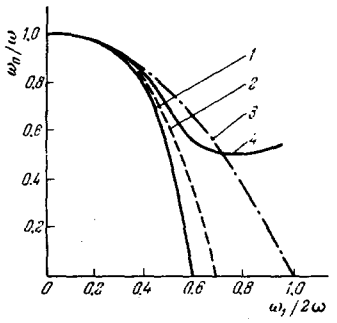

FIG. 16. Bloch-Siegert effect[32]: position of resonance corresponding to a single-photon transition as a function of the amplitude of the rf field. The curves are theoretical: (1)[21,32]; (2)[33]; (3)[24]; (4)[31].

sings may be looked upon as coherence resonances.

It follows from Fig. 15 that anticrossings, observed under the conditions of longitudinal optical pumping, form an "odd" spectrum, whereas the resonance condition is $\omega_0 = (2p+1)\omega$, where $p = 0, 1, 2, \ldots$. Thus, in addition to the "ordinary" magnetic resonance $\omega_0 = \omega$, we have the resonances $\omega_0 = 3\omega, 5\omega, \ldots$, which are associated with multiphoton transitions accompanied by the absorption of several rf photons. Calculations of the probability of k-photon transitions[25] show that the intensity of the corresponding resonance is proportional to $\omega_1^{2k}/\omega^{2k-2}$, and its width for sufficiently large rf field amplitudes is proportional to ω_1^k/ω^{k-1}.

Moreover, it is also clear from Fig. 15 that the position of the resonance, i.e., the center of the anticrossing, undergoes a shift proportional to ω_1^2/ω and amounts to the well-known Bloch-Siegert shift. There has been increasing interest in the well-known Bloch-Siegert shift in recent times because optical methods for the detection of magnetic resonance have led to a sharp increase in precision over a broad range of variation of the amplitude of the rf field. New theoretical[18,21,32] and experimental[29,30] data on the magnitude of this shift have been published. In particular, it has been shown that both the semiclassical[18] and quantum[33] theories lead to practically identical corrections of higher order in the amplitude, and this has refuted the reported conclusion[31] that the semiclassical interpretation of the Bloch-Siegert effect was invalid.

Figure 16 shows the dependence of ω_0/ω on the relative rf field amplitude, which is a measure of the position of the single-photon resonance. Curve (3) corresponds to the well-known formula for the Bloch-Siegert shift in the first-order approximation[24 4)]

$$\omega_0 - \omega \approx -\frac{\omega_1^2}{4\omega}. \tag{49}$$

The quantum theory of the "dressed" atom has been used by Cohen-Tannoudji, Dupont-Roc, and Fabre[33] to deduce the following expression containing higher-order corrections[33] (curve 2):

$$\omega_0 - \omega \approx -\frac{\omega_1^2}{4\omega} - \frac{5}{4}\frac{\omega_1^4}{2^4\omega^3} - \frac{61}{32}\frac{\omega_1^6}{2^6\omega^5} - \ldots. \tag{50}$$

A still more accurate expression has been reported[21,32] and is shown by curve (1) in Fig. 16. It is clear that, for small values of $\omega_1/2$, all theories give practically the same result but, for $\omega_1/2\omega > 0.2$, there is a substantial discrepancy. In particular, curve (4) differs qualitatively from all others.[31]

Measurements of the Bloch-Siegert shift over a broad range of values of ω_1 have been carried out independently in ESR experiments involving conduction electrons in metallic lithium[53] and optically oriented Hg^{199} atoms.[29] The results are in good agreement with theoretical predictions,[21,32,33] with the exception of the region of large amplitudes for ω_1 for which the precision of the measurements is relatively low.

2) *Coherence resonances.* It is clear from Fig. 15 that the level energies of the atom "dressed" by the radio-frequency field of σ polarization exhibit crossings near $\omega_0 = 2p\omega$, where $p = 0, 1, 2, \ldots$. The set of resonances observed when this condition is satisfied has also been referred to as "coherence resonances." If the rf field amplitude is not too large, the position of the coherence resonances can be described by the truncated series:

$$\omega_0 \approx 2p\omega - \delta_{2p}\frac{\omega_1^2}{\omega}, \tag{51}$$

where δ_{2p} is a numerical coefficient representing the shift of the level crossings, which can be calculated by perturbation theory.[17]

The existence of resonances of this kind was first demonstrated experimentally by Haroche.[54] In contrast to the above multiphoton resonances of the ordinary type, which correspond to the anticrossing of the levels of the "dressed" atom, coherence resonances are observed only under the conditions of transverse pumping, when circularly polarized resonance optical radiation produces no population difference between the atomic sublevels $|+\frac{1}{2}\rangle$ and $|-\frac{1}{2}\rangle$. If, at the same time, one records the intensity of the pump radiation transmitted by the atomic system, which is proportional to the transverse component of the atomic orientation, the resultant signal contains only the even harmonics of the rf field. In accordance with the general properties of level crossing, the observed resonances should not exhibit appreciable broadening when the rf field amplitude increases, but should show a radiative shift in the direction of decreasing field H_0 [see (51)]. The above properties of resonances of this type are well illustrated by the experimental curves[54] shown in Fig. 17.

Figure 17a shows a series of recordings of the resonance $\omega_0 \approx 2\omega$, obtained by successively increasing the amplitude of the rf field. It is clearly seen that: (1) there is no radiative line broadening and (2) the tops of the resonance curves lie on a common straight line. The latter follows immediately from the fact that the size of the resonance and its radiative shift are proportional to ω_1^2. A similar set of curves is shown in Fig. 17b for the resonance $\omega_0 \approx 4\omega$. In this case, the tops of the resonance curves recorded for different

4) An exact solution for the Bloch-Siegert shift in all orders has been reported by Swain.[75]

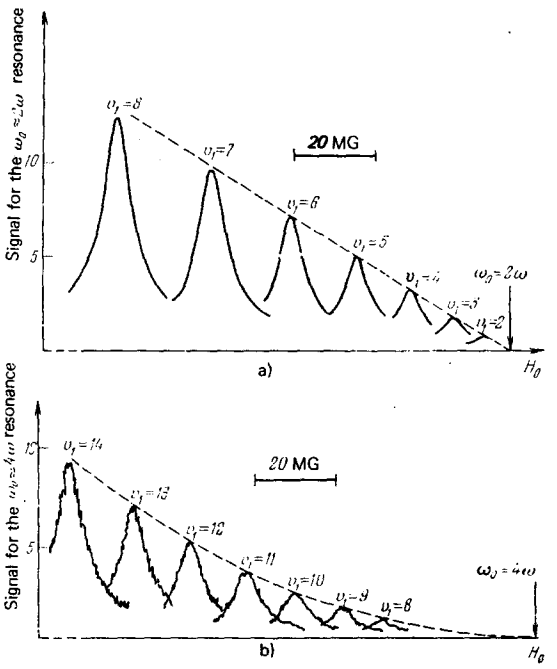

FIG. 17. Coherence resonances observed in a σ-polarized rf field[54]: (a) shift of the $\omega_0 \approx 2\omega$ resonance with increasing amplitude of the rf field; (b) shift of the $\omega_0 \approx 4\omega$ resonance with increasing amplitude of the rf field.

amplitudes of the rf field lie on a parabola because the intensity of this resonance is proportional to ω_1^4, whereas the shift is proportional to ω_1^2, as before.

C. Magnetic properties of a "dressed" atom in a weak magnetic field

We have examined the resonance phenomena observed in the neighborhood of the crossing and anticrossing of levels of the atom + rf field system when the constant magnetic rield H_0 cannot be regarded as weak. At the same time, it is clear from Figs. 1, 2, and 15 that, for fields near $H_0 = 0$, there is also a region in which the energy levels exhibit crossing or anticrossing. Qualitative analysis shows that, when H_0 is very small, the energy-level diagram for the "dressed" atom is determined by the polarization of the rf field. When this field is linearly polarized, there is always a level crossing near $H_0 = 0$, independently of the mutual orientation of $\mathbf{H}_0$ and $\mathbf{H}_1(t)$. If, on the other hand, the field $\mathbf{H}_1(t)$ is circularly polarized, the level degeneracy at $\omega_0 = 0$ is removed because the level-crossing point is shifted, or $\omega_0 = 0$ because of the appearance of anticrossing.

The interaction between the atom and the linearly polarized rf field leads to a substantial change in the magnetic properties of the atom and, in particular, to a change in the Landé factor of the "dressed" atom. This effect is observed for $\omega_0 \ll \omega$, i.e., in the region of the magnetic field in which the dependence of energy on H_0 can still be regarded as linear. When this inequality is satisfied, the frequency ω of the field $H_1(t)$ lies well away from the resonance value, so that the effect of the change in the g-factor is essentially nonresonant, and is due to processes involving virtual absorption and emission of rf photons by the atom. This is associated with deep changes in the paramagnetic properties of the "dressed" atom, including a change in the frequency of Larmor precession and in the width of the Hanle-effect curve, an equalization of the precession frequencies of the atoms of different elements, a reduction in the inhomogeneous broadening of the magnetic resonance line, and other effects.

The interaction between a system of atoms of spin $\frac{1}{2}$ with the linearly polarized rf field $\mathbf{H}_1(t)$ at an arbitrary angle relative to $\mathbf{H}_0$ has been examined theoretically by many workers in the approximation $\omega_0 \ll \omega$. Some authors use a purely classical approach[18,37] whilst others have solved this problem semiclassically[21] or have consistently used a quantum formalism.[17] As expected, all these methods yield the same results which can be simply described in terms of the "dressed"-atom formalism[17].

If $\omega_0 = 0$, the Hamiltonian for the "dressed" atom can be written in the form[5)]

$$\hat{\mathcal{H}} = \omega \hat{a}^+ \hat{a} + \lambda \hat{J}_z (\hat{a} + \hat{a}^+). \tag{52}$$

When the magnetic field $\mathbf{H}_0$ is present and $\omega_0 < \omega$, the Zeeman part of the Hamiltonian is

$$\hat{\mathcal{H}}_{\text{Zeem}} = -g_\alpha \mu_B \hat{\mathbf{J}} \cdot \mathbf{H}_0 \tag{53}$$

and may be looked upon as a perturbation. In the space of states of the "dressed" atom, the perturbation operator can be written in the form

$${}^{nn}\hat{\mathcal{H}}_{\text{Zeem}} = -g_\alpha \mu_B H_0 \cdot {}^{nn}\hat{\mathbf{J}}, \tag{54}$$

where

$${}^{nn}\hat{J}_z = {}^{nn}\hat{j}_z, \quad {}^{nn}\hat{J}_\pm = J_0\left(\frac{\omega_1}{\omega}\right) {}^{nn}\hat{j}_\pm, \tag{55}$$

and $\hat{\mathbf{j}}$ is the "fictitious" angular momentum of the "dressed" atom. Therefore,

$${}^{nn}\hat{\mathcal{H}}_{\text{Zeem}} = -g_\alpha \mu_B \left[H_{0z}\, {}^{nn}\hat{j}_z + H_{0x}\, {}^{nn}\hat{j}_x J_0\left(\frac{\omega_1}{\omega}\right) + H_{0y}\, {}^{nn}\hat{j}_y J_0\left(\frac{\omega_1}{\omega}\right) \right]. \tag{56}$$

The energy of the Zeeman levels is linearly related to the field H_0, and this enables us to introduce the Landé factor for the dressed atom, $\bar{g}$. It is easily seen that the factor $\bar{g}$ is anisotropic because it is equal to the Landé factor g_α for the free atom when $\mathbf{H}_0 \| Oz \| \mathbf{H}_1(t)$, and differs from it ($\bar{g} = g_\alpha J_0(\omega_1/\omega)$) when $\mathbf{H}_0 \perp \mathbf{H}_1(t)$. It is therefore possible to define a Landé tensor $\bar{g}$ for the "dressed" atom that is diagonal in the coordinate frame $Oxyz$ and has rotational symmetry relative to the z axis[6)]:

$$\bar{g} = g_\alpha \begin{pmatrix} J_0\left(\frac{\omega_1}{\omega}\right) & 0 & 0 \\ 0 & J_0\left(\frac{\omega_1}{\omega}\right) & 0 \\ 0 & 0 & 1 \end{pmatrix}. \tag{57}$$

When the field $\mathbf{H}_0$ is at an arbitrary angle ψ to the z axis, the Landé factor in this field is

$$|\bar{g}_\psi| = g_\alpha \sqrt{\cos^2\psi + J_0^2\left(\frac{\omega_1}{\omega}\right) \sin^2\psi}. \tag{58}$$

---

5) The quantization axis is taken to lie along the "dressing" oscillating field $\mathbf{H}_1(t)$.

6) A more general expression has also been given in the literature,[87] of which (57) is the quasiclassical limit.

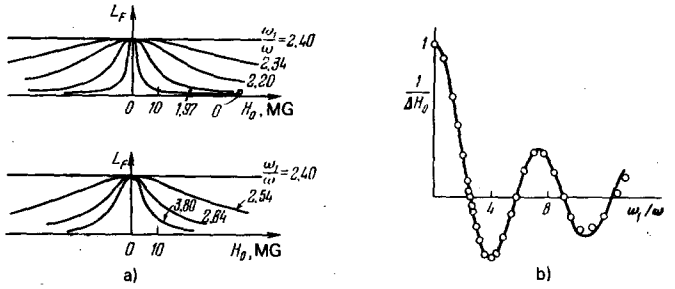

FIG. 18. Hanle effect for a "dressed" atom[39]: (a) Hanle effect lines for Hg^{199} atoms for different values of the amplitude of the "dressing" field $H_1(t)$; (b) reciprocal of the Hanle linewidth as a function of ω_1/ω.

The above properties of the "dressed" atom has led to a simple and graphic interpretation of many new effects discovered in recent years.

Direct experimental confirmation of the validity of (57) was first reported by Cohen-Tannoudji and Haroche,[39] who observed a variation in the Hanle linewidth in the ground state of Hg^{199} atoms in the presence of an rf field $\mathbf{H}_1(t) \perp \mathbf{H}_0$ ($\psi = \pi/2$). The results of this experiment are shown in Fig. 18, which gives a family of the Hanle-effect curves obtained for different values of the rf field amplitude (Fig. 18a), and a theoretical functional dependence $J_0(\omega_1/\omega)$ on which the points show the experimental values of the reciprocal linewidth.

Similar data were subsequently obtained for H^1 atoms, in a hydrogen maser,[41b] and then for protons in water, F^{19} nuclei in teflon, and Na^{23} in NaCl in ordinary NMR experiments.[55a] Kunimoto and Hashif[55b] have reported that a change in the Landé factor of Na^{23} and Cl^{35} in a single crystal with the aid of a nonresonant rf field has enabled them to change the energy of these nuclei in the rotating set of coordinates in an experiment on nuclear double resonance.

The anisotropy of the Landé factor has been confirmed by Landré *et al.*,[40] who investigated the free precession of "dressed" Hg^{199} atoms under the conditions of optical pumping with a variable angle between $\mathbf{H}_0$ and $\mathbf{H}_1(t)$. These results are shown in Fig. 19 in polar coordinates, where ψ is the angle between $\mathbf{H}_0$ and $\mathbf{H}_1(t)$, and the length of the radius vector is proportional to the Landé factor in this particular direction. The curves correspond to the theoretical formula given by (58) for different values of ω_1/ω, whereas the points represent the experimental results. As can be seen, there is excellent agreement between theory and experiment.

Haroche and Cohen-Tannoudji[41a] have described an original experiment in which observations were made of the transfer of Zeeman coherence between Rb^{87} and Cs^{133} atoms, whose Landé factors were equalized during the interaction between the atoms and the nonresonant rf field $\mathbf{H}_1(t)$. As is well known, in a nonzero field $\mathbf{H}_0$ the angular-momentum transfer efficiency for collisions between different atoms is very different for longitudinal and transverse components. Because of the difference between the Larmor frequencies $\omega_{0\alpha_1} = -g_{\alpha 1}\mu_B H_0$ and $\omega_{0\alpha_2} = -g_{\alpha 2}\mu_B H_0$ of the two atoms, the condition

$$|\omega_{0\alpha_1} - \omega_{0\alpha_2}| T_{\alpha_1\alpha_2} \ll 1, \tag{59}$$

is not satisfied, where $T_{\alpha_1\alpha_2}$ is the time between successive collisions necessary for the effective transfer of coherence. It follows that the transfer of transverse angular momentum components between the atoms is much less effective than the transfer of longitudinal components.[57] However, by applying a linearly polarized nonresonant rf field to a system consisting of two types of atoms, it is possible to ensure that (59) is satisfied by "equalizing" the corresponding factors $\bar{g}_{\alpha i}$:

$$\bar{g}_{\alpha 1} = g_{\alpha 1} J_0\left(\frac{g_{\alpha 1}\mu_B H_1}{\omega}\right), \quad \bar{g}_{\alpha 2} = g_{\alpha 2} J_0\left(\frac{g_{\alpha 2}\mu_B H_1}{\omega}\right). \tag{60}$$

Figure 20 shows curves constructed with the aid of (60) and representing the resonance frequencies $\bar{\omega}_{0\mathrm{Rb}} = -\bar{g}_{\alpha\mathrm{Rb}}\mu_B H_0$ and $\bar{\omega}_{0\mathrm{Cs}} = -\bar{g}_{\alpha\mathrm{Cs}}\mu_B H_0$ of the Rb^{87} and Cs^{133} atoms, "dresssed" by the same field $H_1(t)$, as functions of the amplitude H_1 of this field. It is clear that there is a whole range of values of H_1 for which $\bar{\omega}_{0\mathrm{Rb}} = \bar{\omega}_{0\mathrm{Cs}}$ (the points A, B, C, etc.), so that the coherence-transfer efficiency during spin exchange between Rb^{87} and Cs^{133} atoms should increase sharply in the neighborhood of these points. The theory of this effect is discussed in detail by Haroche,[17] and there is no point in reproducing it here. However, we shall briefly review experimental demonstrations of the existence of this effect.[41a]

Figure 21 shows the block diagram of the experiment and the sequence of operations along the time scale which result in the appearance of the signal indicating the transfer of coherence. The working cell contains a mixture of saturated vapors of Rb^{87} and Cs^{133}, where the density of the Cs vapor is greater by a factor of 10 than the density of the Rb vapor. This facilitates an in-

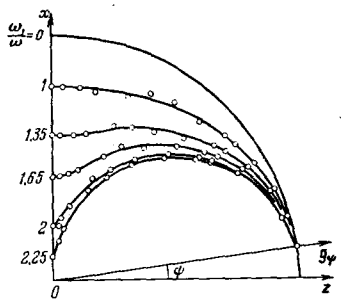

FIG. 19. Graphical representation of the anisotropic Landé factor of a "dressed" atom in polar coordinates. Solid curves—theoretical (58); open circles—experimental.[40]

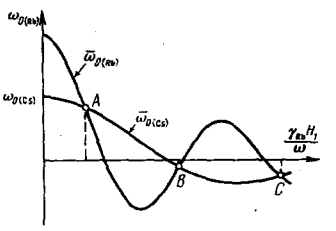

FIG. 20. Resonance frequency (H_0 = const) for "dressed" Rb^{87} and Cs^{133} atoms as a function of the amplitude of the "dressing" rf field.

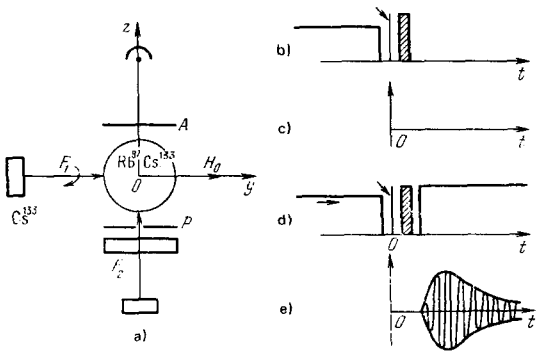

FIG. 21. Experimental observation of coherence transfer between Rb^{87} and Cs^{133} atoms[41a]: (a) experimental arrangement; (b) sequence of rf pulses producing coherence in the Cs^{133} system; (c) no signal at the photomultiplier output after the series of pulses; (d) sequence of rf pulses necessary for transfer of coherence to the Rb^{87} atoms; (e) damped-precession signal due to the Rb^{87} atoms appears at the photomultiplier output.

crease in the probability of collisions accompanied by spin exchange. The circularly polarized (σ^+) beam of light F_1 from a cesium lamp orients the angular momenta of Cs^{133} in the direction of the y axis, which is parallel to $\mathbf{H}_0$. The Rb^{87} atoms are also polarized in this direction as a result of spin exchange, but the rf field $\mathbf{h}_{Rb}(t) \perp \mathbf{H}_0$ disturbs this orientation without affecting the Cs^{133} atoms. It follows that only the Cs^{133} atoms have a longitudinal angular momentum component during the initial stage of the experiment. At $t=0$ in Figs. 21b–e, the field $\mathbf{h}_{Rb}(t)$ is turned off, and the pump beam F_1 is interrupted. A 90° pulse of the resonant rf field producing coherence in the set of Cs^{133} atoms is then applied to the system. The light beam F_2 from a lamp containing the isotope Rb^{85} was used to observe the presence of coherence in the system of Rb^{87} atoms. Since this was nonresonant radiation, it produced no perturbation but, at the same time, ensured the possibility of detection of the signal proportional to the transverse component of the angular momentum of the Rb^{87} atoms. If the Rb-Cs system is not subjected to the "dressing" field $H_1(t)$ after the 90° pulse, the precession frequencies of Rb^{87} and Cs^{133} are very different, and the absence of a signal at the output of the photomultiplier (Figs. 21b and c) indicates that no coherence is introduced into the system of Rb^{87} atoms. If, on the other hand, the field $H_1(t)$ is imposed on the cell immediately after the 90° pulse (Fig. 21d), and the amplitude of this field corresponds to point A in Fig. 20 (intersection of the two Bessel functions), coherence is transferred from the Cs^{133} system to the Rb^{87} atoms because of the equalization of the Landé factors, and the signal shown in Fig. 21e appears at the photomultiplier output.

These experiments and their analysis[17] provided qualitative and quantitative confirmation of all the predictions of the theory and once again demonstrated the value of the idea of the "dressed" atom.

The dependence of the factor $\bar{g}$ on the amplitude of the field $H_1(t)$ has led to the development of a new type of parametric resonance,[58] which appears when the frequency of the harmonic modulation of the Landé factor of the "dressed" atom [modulation of the amplitude of the field $H_1(t)$] is equal to the resonance frequency $\bar{\omega}_0$.

D. Magnetic relaxation in the presence of a strong rf field

We showed above that the interaction between an atom in a sufficiently strong nonresonant rf field will modify the magnetic properties of the atom and, in particular, its Landé factor. Since the Landé factor determines the rate of magnetic relaxation in the spin system, one would expect that the relaxation parameters would exhibit an appreciable dependence on both the intensity and the frequency of the rf field.

A similar problem of spin relaxation in the presence of an rf field has been examined in the literature[22,59] earlier on the basis of the semiclassical theory. A systematic quantum-mechanical theory of relaxation due to inhomogeneity of the constant field $\mathbf{H}_0$ has also been developed[17] for atoms interacting with a strong rf field $\mathbf{H}_1(t) \| \mathbf{H}_0$. The main predictions of this theory and the experimental results obtained are summarized below.

It is well known[60] that, when a nonuniform constant magnetic field $\mathbf{H}_0$ is present, the atoms of the specimen, which are in constant thermal motion, experience the effect of the random field $\mathbf{h}(t)$ that is capable of stimulating real transitions between the sublevels and is responsible for magnetic relaxation. Simple calculations show that the probability of relaxation transitions between sublevels satisfying $\Delta m = \pm 1$ and separated by an energy interval ω_0, that are stimulated by a field of this kind, can be written in the form

$$W_{m\pm1,\,m} = \frac{\gamma_\alpha^2}{4} |\langle \alpha J_{m\pm1} | \hat{J}_\pm | \alpha J_m \rangle|^2 f(\omega_0), \tag{61}$$

where

$$f(\omega_0) = \int_0^\infty g(\tau) e^{-i\omega_0\tau} d\tau \tag{62}$$

is the spectral density of the perturbation characterized by the correlation function

$$g(\tau) = \overline{h_-(t) h_+(t-\tau)}, \tag{63}$$

and h_- and h_+ are the transverse components of the rotating random field $h(t)$. The total relaxation probability is given by

$$\Gamma_{\alpha Jm} = W_{m+1,\,m} + W_{m-1,\,m} = \gamma_\alpha^2 A_{Jm} f(\omega_0), \tag{64}$$

where $A_{Jm} = [J(J+1) - m^2]/2$.

The above formulas describe the relaxation of the atom in the absence of an additional rf field $\mathbf{H}_1(t)$. If a linearly polarized field $\mathbf{H}_1(t) \| \mathbf{H}_0$ is present, the state vectors $|\alpha Jm\rangle$ in the expression for the magnetic relaxation probability must be replaced by the state vectors $|\overline{\alpha Jm, n}\rangle$ for the "dressed" atom, which are given by a formula similar to (33). The matrix elements in this formula are

$$\langle \overline{\alpha Jm',\, n'} | \gamma_\alpha \hat{J}_\pm | \overline{\alpha Jm,\, n} \rangle = \gamma_\alpha \delta_{m',\,m\pm1} \langle \alpha Jm' | \hat{J}_\pm | \alpha Jm \rangle J_{n'-n}\left(\pm\frac{\omega_1}{\omega}\right), \tag{65}$$

and the relaxation transition probability in the field H_1

is

$$W_{\substack{m\pm1,\,n';\,m,\,n}}=\frac{\gamma_\alpha^2}{4}[J(J+1)-m(m\pm1)]J_{n'-n}^2\left(\frac{\omega_1}{\omega}\right)f[\mp\omega_0+(n-n')\omega]. \tag{66}$$

After some rearrangement, taking into account the parity of the function f, it is possible to show that the relaxation probability for the "dressed" atom is

$$\Gamma_{\substack{\alpha Jmn}}=\gamma_\alpha^2A_{Jm}\sum_r J_r^2\left(\frac{\omega_1}{\omega}\right)f(\omega_0+r\omega). \tag{67}$$

It is easily seen that (67) becomes identical with (64) for an atom in the absence of $H_1(t)$ when $\omega_1=0$.

Qualitative analysis of (67) shows that, as ω_1 increases, the term describing direct relaxation with the lattice without the participation of the rf photons ($r=0$) tends to decrease, but the total change in the probability $\Gamma_{\alpha Jmn}$ depends on the relationship between ω_0 and the correlation time τ_c, which, in the case of optical pumping, is equal to the time of flight of the atom between the walls of the cell containing the atoms being investigated ($\tau_c\sim10^{-3}-10^{-4}$ sec). When $\omega_1\tau_c\ll1$, the influence of the "dressing" field on the relaxation processes can be neglected, and an appreciable effect is observed only for $\omega_1\tau_c\gtrsim1$.

In weak fields H_0, when $\omega_0\tau_c\ll1$ (condition for the utmost narrowing of the line), we have $f(\omega_0)\approx f(0)$, and the total probability of relaxation of the atom in the absence of the field $H_1(t)$ is

$$\Gamma_{\alpha Jm}=A_{Jm}\gamma_\alpha^2f(0). \tag{68}$$

For the "dressed" atom, we have under analogous conditions

$$\Gamma_{\substack{\alpha Jmn}}=A_{Jm}\gamma_\alpha^2\sum_r J_r^2\left(\frac{\omega_1}{\omega}\right)f(r\omega)\approx A_{Jm}\gamma_\alpha^2J_0^2\left(\frac{\omega_1}{\omega}\right)f(0). \tag{69}$$

It follows from this result that, as the amplitude of the rf field ω_1/γ_α increases, the width of the observed resonance line should decrease and, in particular, at the points at which the Bessel function vanishes ($\omega_1=2.4\omega$, 5.6ω, and so on), the broadening of the resonance line due to the inhomogeneity of the magnetic field should be zero.

Experiments[17] performed with Rb^{87} atoms have completely confirmed the above theoretical results. The inhomogeneous broadening of the $m_F=0-m_{F'}=0$ hyperfine-structure line was investigated in the presence of an inhomogeneous constant field. In a weak field $H_0(\omega_0\tau_c<1)$, magnetic relaxation appreciably broadens the resonance line even for field gradients of the order

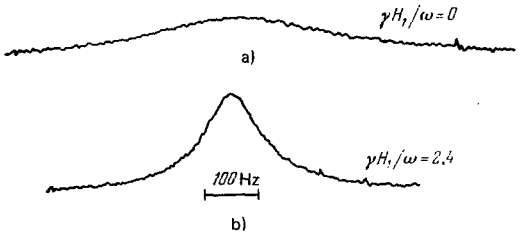

FIG. 22 Record of the $m_F=0-m_{F'}=0$ hyperfine-structure Rb^{87} line in an inhomogeneous magnetic field[17]: (a) $H_1=0$; (b) "dressing" field turned on ($\omega/2\pi=7500$ Hz).

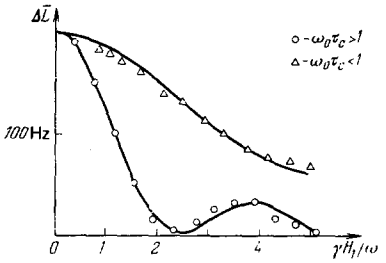

FIG. 23. Inhomogeneous broadening of the resonance line for Rb^{87} atoms as a function of $\gamma H_1/\omega$ for $\omega_0\tau_c<1$ and $\omega_0\tau_c>1$ (solid curves are theoretical).

of 0.2 mOe/cm (Fig. 22a). It is clear from Fig. 22b that the application of the rf field $\mathbf{H}_1(t)\|\mathbf{H}_0$ leads to a substantial narrowing of the line since, in accordance with (69), the inhomogeneous broadening is then zero. The experimental dependence of this broadening on the ratio $\gamma H_1/\omega$ is shown in Fig. 23, and is in full agreement with the theoretical predictions.

E. Interaction between an atom and several rf fields

The simultaneous application of two or more electromagnetic fields to an atomic system is one of the basic problems considered in the theory of nonlinear phenomena in atoms. The problem has been solved by several workers (largely within the framework of nonlinear optics) by very different models and methods. Interest in this problem has increased considerably since the advent of laser technology. New physical phenomena have been discovered, including multiphoton processes involving the participation of different types of photons,[61,66] stimulated and ordinary Raman effects, parametric frequency conversion, and parametric amplification.[62,63] All these problems can be successfully treated in terms of the concept of a "dressed" atom, for example, by considering the scattering of weak-field (probe radiation) by the "atom + strong 'dressing' field" system. This approach substantially simplifies the theoretical analysis and provides graphic descriptions of the effects under consideration.[67]

Lack of space prevents us from describing all the aspects of the interaction of the atom with several fields,[7)] and we shall therefore confine our attention to a brief description of only two phenomena, namely, the change in the Zeeman spectrum of the "dressed" atom and parametric frequency conversion. As regards other questions, including details of the theory, we refer the reader to the review paper by Haroche,[17] who gives a detailed and clear account of many of the aspects of the probelm that is only briefly touched upon in the present paper.

1) *Zeeman spectrum of a "dressed" atom in a weak magnetic field.* Before we proceed to the description of

[7)] We must also mention the important work[71,72,76-78] on the change in the absorption spectrum and dispersion of a system of two-level atoms in a quasiresonant monochromatic radiation field.

the spectrum itself, let us consider possible transitions between the energy levels of the "dressed" atom far from their anticrossing. For simplicity, we shall confine our attention to a system of atoms with spin angular momenta equal to $\frac{1}{2}$ in both the upper and lower states, subject to the condition that the gyromagnetic ratio in the lower state is much smaller than in the upper (Fig. 24a). If we analyze the energy-level diagram of the atom "dressed" by a rotating radio-frequency field (Fig. 24b), we can easily see that a probe field with $\sigma^{\pm}$ polarization can produce the following transitions: (a) the transition $|-,n\rangle \to |\overline{+,n}\rangle$ (arrow 1), which is not accompanied by a change in the number of photons of the "dressing" field and is stimulated by the simple absorption of one probe photon with polarization σ^+ and (b) the transition $|+,n\rangle \to |\overline{-,n}\rangle$, corresponding to simple absorption of one probe photon with polarization σ^- (transition 3). It is clear from the diagram that these "ordinary" transitions are characterized by resonance-frequency shifts due to the interaction of the atom with the "dressing" field.

In addition to the foregoing, there are also transitions of a totally different type between levels of the "dressed" atom. Thus, the $|+,n\rangle \to |\overline{+,n-1}\rangle$ transition (arrow 4) is accompanied by absorption of one probe photon with polarization σ^- and one photon of the "dressing field" with polarization σ^+, with the result that the angular momentum of the system is conserved ($\Delta m = 0$). The probability of this process is proportional to the square of the amplitude of the "dressing" field ω_1^2 and to the square of the amplitude of the probe field h_1^2, whilst the resonance line experiences a radiation shift proportional to $(\omega_1/\omega)^2$. Finally, the $|-,n\rangle \to |\overline{-,n+1}\rangle$ transition (arrow 2) is accompanied by absorption of a σ^+ probe photon and the stimulated emission of a σ^+ photon of the "dressing" field. It is easily seen that this process has all the characteristics of the inverse Raman effect in which the "dressing" rf field plays the role usually played by the laser field. We also note that the magnitude of the effect is proportional to ω_1^2, whereas the radiative shift of the resonance line is proportional to $(\omega_1/\omega)^2$.

In the processes described above, the photons of the "dressing" field play a twofold role. Firstly, they can undergo virtual absorption or emission by the atom, the result of which is a frequency shift and a change in the probability of transitions stimulated by the probe field. Secondly, the atom may undergo real absorption or emission of one or more photons of the "dressing" field with the simultaneous absorption of a photon of the probe field, which leads to the appearance of new lines in the absorption spectrum. In the general case of arbitrary polarization of the "dressing" field, the new lines are sufficiently numerous because they correspond to transitions with the participation of several photons.

Figure 25 shows the spectra corresponding to transitions between hyperfine-structure states in a system of Rb^{87} atoms ($0 \to 0$), stimulated by a microwave field with π and σ polarization in the presence of a "dressing" field of frequency $\omega/2\pi = 2700$ Hz, polarized in different ways.[17] It is readily seen that the spectrum contains numerous new lines, connected with the absorption of several photons of the two fields, and with Raman-type transitions. The nature of the spectrum depends to a large extent on the polarization of the rf fields. Without going into details, we merely note that the observed picture can be completely interpreted in terms of the theory given by Haroche,[17] taking into account the conservation of the total angular momentum of the system consisting of the atom, the "dressing" field, and the probe field. Analysis of the intensity of the individual spectral lines as a function of the ratio ω_1/ω demonstrates complete agreement between the theoretical and experimental curves, thus confirming once again the value of the concept of a "dressed" atom.

The change in the energy spectrum of an atom in the presence of a strong "dressing" field must be taken into account when the interaction between the "dressed" atom and external fields that are much weaker than the "dressing" field is analyzed. It is precisely in this situation, in which the atomic system experiences several electromagnetic fields (one of which substan-

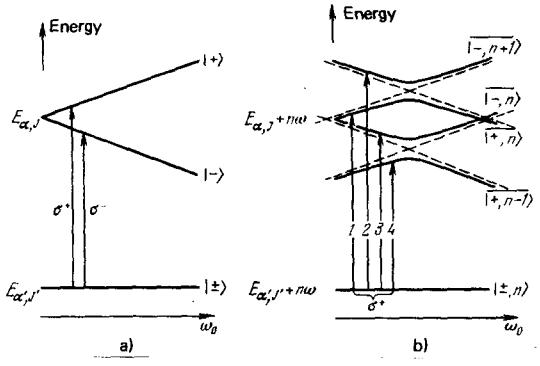

FIG. 24. Transitions stimulated by a rotating probe field (a) between levels of a free atom ($\sigma^{\pm}$) and (b) between levels of a dressed atom (σ^{+}).

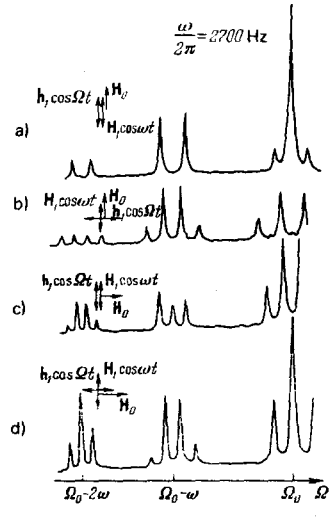

FIG. 25. Spectrum of hyperfine transitions in Rb^{87} atoms in the presence of two fields: a probe field of frequency 6834.683 MHz ($0 \to 0$ transition) and a "dressing" field of 2700 Hz. The spectra correspond to the following polarizations of the rf fields (the subscript s indicates the polarization of the probe field), (a) π, π_s; (b) π, σ_s; (c) σ, σ_s; (d) σ, π_s.

tially modifies the energy spectrum of the system and the others can be looked upon as a perturbation) that the concept of the atom "dressed" by the field is particularly fruitful. The important point is that no fundamental restrictions are imposed on the frequency of the "dressing" field: the field can be in the rf, microwave, or optical range.

An example of the situation where the atomic system experiences simultaneously several fields is parametric resonance, observed in vapors of optically oriented atoms "dressed" by a radio-frequency field of the form $H_1\cos\omega t$, which is excited by another rf magnetic field $H_2\cos\omega t$.[67,79] Theoretical analysis of parametric resonance of this kind, based on the theory of the "dressed" atom and its experimental investigation in optically oriented alkali-metal vapors in the ground state, have led to the development of a quantum optical magnetometer for the simultaneous determination of the three components of an ultraweak constant magnetic field H_0 with a precision better than 10^{-9} Oe.[64]

The method of the "dressed" atom has also been used in the analysis of the optical pumping process itself,[68,69] in the case where the intensity of the resonant pump beam is high, i.e., when the matrix elements of the interaction operator between the atoms and the field in (22) exceed the natural width Γ of the excited states. It is clear that, in this case, the absorption spectrum of the atom in the presence of the radiation field may undergo an appreciable change, and a discussion of such a process of perturbing an atom "dressed" by a field is completely natural.

2) *Parametric frequency conversion.* We have considered the absorption spectrum of a "dressed" atom. It is clear that an analogous approach can be adopted to investigate the spectral distribution of photons scattered by an atom interacting with the "dressing" rf field. We also note that, in principle, there is nothing to stop us from using as the probing field the optical harmonics that stimulate transitions between the ground and excited atomic states, especially since the cross sections for the scattering of the optical photons are much greater than the cross sections for rf photons. This naturally leads us to the problem of scattering of optical photons by an atom interacting with the "dressing" rf field. Depending on the number and polarization of photons of the optical and rf fields, one would expect to observe different multiphoton processes of a "mixed" type. We shall not analyze the entire range of possible situations, and will confine our attention to one effect, known as parametric frequency conversion.

Let Ω_k be the frequency of a harmonic of the optical field incident on the atom interacting with the rf ("dressing") field of frequency ω. When the optical photon is singly scattered, it may happen that the frequency $\Omega_{k'}$ of the scattered optical radiation will differ from Ω_k. For example, if the atom during its time in the excited state takes part in virtual absorption of one, and the emission of two, rf photons, energy conservation dictates that

$$\Omega_{k'}=\Omega_k+\omega-2\omega=\Omega_k-\omega, \tag{70}$$

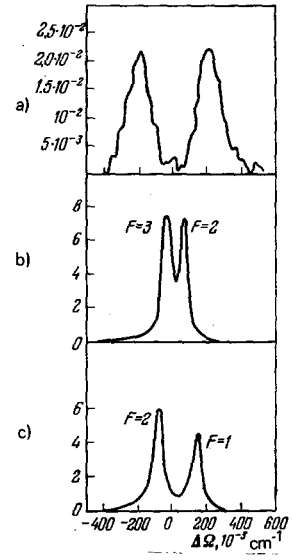

FIG. 26. Parametric optical frequency conversion[65]: (a) spectral profile of radiation scattered by the atoms; (b) D_1 emission line of Rb^{85} atoms in a discharge; (c) D_1 emission line of Rb^{87} atoms in a discharge.

from which it is clear that the scattering of the optical photon was accompanied by a conversion of its frequency $(\Omega_k-\Omega_{k'})$. In terms of the terminology employed in radiophysics, it may be said that the spectrum of the scattered radiation acquires "side bands" separated from the frequency of the optical "carrier" by an amount proportional to the frequency ω of the rf field.

The "side bands" of the scattered optical spectrum were first observed experimentally by Tang and Happer,[65] who studied the change in the emission spectrum of Rb^{85} atoms after transmission through a set of Rb^{87} atoms interacting with a microwave field ($\nu=\omega/2\pi$ = 6.835 Hz). The spectrum was recorded with a Fabry-Perot interferometer, and effective suppression of the "carrier" optical frequency was achieved by placing the cell containing the Rb^{87} atoms between two crossed polaroids. To increase the conversion efficiency, the frequency of the microwave field was chosen equal to the frequency of transitions between the hyperfine-structure levels of the Rb^{87} atoms in the ground state. Figure 26a shows a typical scattered spectrum together with the emission spectra of the Rb^{85} and Rb^{87} atoms (Figs. 26b and c) in a discharge. It is readily seen that the position of the peaks in Fig. 26a is not the same as the position of the spectral components of Rb^{85} or Rb^{87}, but is in accordance with the expected position of the "side bands" due to parametric frequency conversion. Additional control experiments confirmed the validity of this interpretation. The estimated frequency conversion efficiency was 10^{-3}.

The above experiment was performed with an ordinary spectral lamp as the source of the optical radiation. Undoubtedly, the use of a tunable laser would yield more interesting results.[73]

The authors are indebted to E. B. Aleksandrov for his detailed examination of the manuscript of this review and for valuable suggestions.

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Translated by S. Chomet