Atom in magnetic and resonant optical fields

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(Submitted January 29, 1975)
Zh. Eksp. Teor. Fiz. **68**, 2010–2021 (June 1975)

A general solution is obtained for the problem of finding the quasi-energy spectrum and wave functions of an atom in a magnetic field and in the field of a light wave of arbitrary polarization under conditions when the interaction with the wave field is of the same order of or larger than the Zeeman splitting of the atomic levels. The case of resonating 1/2–1/2 and 0–1 levels is analyzed in detail. The atomic parameters that characterize the magnetization and the emf in a beam of atoms interacting with the light (the inverse Faraday and Kerr effects) are calculated. General formulas are obtained for the correlation and polarization parameters of the spontaneous emission excited by strong light; these formulas are the basis of the theory of the Hanle effect in a strong optical field.

PACS numbers: 32.10.Dk

1. INTRODUCTION

Diligent research is being carried out of late on both theoretical and experimental nonlinear-optical phenomena in atomic gases. In particular, interesting results were obtained concerning the polarization phenomena in a resonant atomic medium. Arutyunyan et al.[1] obtained the spectrum of quasistationary states of an atom in a partially polarized field that is at resonance with the transition frequency between the levels with angular momenta $1/2$ and $3/2$, considered the variation of the polarization characteristics of radiation propagating in such a medium, and calculated the cross sections of certain four-photon processes. An experimental investigation of these phenomena is the subject of[2]. A number of results pertaining to this question were obtained by Rautian et al.[3] and are contained in the lectures of Ter-Mikaelyan[4].

In this paper we consider the influence of an external magnetic field on the polarization phenomena that are produced when an elliptically-polarized ray passes through a resonant atomic medium, As is well known, in a linearly polarized light field with electric vector **E** directed along a constant magnetic field **B**, this direction is singled out and the projections of the angular momenta of the atomic states on this direction are conserved. Each pair of the Zeeman sublevels having the same magnetic quantum number forms a two-level system, different systems in the isolated atom do not interact with one another, and transitions between them occur only as a result of interatomic collisions or incoherent radiative corrections[5]. A similar situation obtains also for the circularly-polarized field propagating along **B**. The only difference is that the two-level systems are made up of states with magnetic quantum numbers that differ by unity. In all other cases there is no preferred direction, the magnetic quantum numbers are not conserved, and for a resonant optical field whose interaction with the atom is of the order of or larger than the Zeeman splitting it is necessary to solve the Schrödinger equation for a system of n nondegenerate levels ($n > 2$). In this paper this solution is constructed by a general method proposed by one of us and Katsnel'son[6].

The entire exposition is in the language of the quasienergy states of a quantum system in a periodic external field[7], in which it is possible to ascribe natural and illustrative quantum numbers to each energy state of the atom in the field. The construction of the quasienergy states and the determination of the quasienergy spectrum are carried out in Sec. 2. Section 3 is devoted to the calculation of the alternating components of the magnetic dipole and electric quadrupole moments of the atom in a strong optical field. These parameters are determined by the RF radiation of the medium into which the light beam is focused[8]. In Sec. 4 are calculated the polarization and correlation characteristics of the spontaneous emission of the atom in the magnetic field when the excitation of the atom is produced by light of high intensity. The results are thus the main theory of the Hanle effect in a strong optical field.

2. QUASIENERGY STATES OF THE ATOM

The Schrödinger equation for an atom in a constant magnetic field **B** directed along the z axis and a monochromatic optical field with electric vector **E**(t) = Re{**F**$e^{-i\omega t}$} is of the form[1)]

$$i\frac{\partial\psi}{\partial t}=[H_0-\mu\mathbf{B}-\mathbf{dE}(t)]\psi(t). \quad (1)$$

Here μ and **d** are the magnetic and electric dipole moments of the atom, and H_0 is the unperturbed Hamiltonian. The interaction of the atom with the optical field is described in the dipole approximation.

Assume that the atom has two states with total angular momenta j and J ($|J - j| \leq 1$) and energies ϵ_1 and ϵ_2, so that $\epsilon_2 - \epsilon_1 \approx \omega$ (see Fig. 1). In this case, if the atom was in one of the states prior to turning on the light field, we can use the $2(J + j + 1)$-level approximation and seek the solution of (1), following[6], in the form

$$\psi(t)=\sum_m a_m(t)\exp\{-i(\varepsilon_1+\delta/2)t\}|njm\rangle+\sum_M b_M(t)\exp\{-i(\varepsilon_2-\delta/2)t\}\times|NJM\rangle,\quad \delta=\varepsilon_2-\varepsilon_1-\omega. \quad (2)$$

The equations for a_m and b_M take in the resonance approximation the form

$$i\dot{a}_m=\left(-i\gamma_m-\frac{\delta}{2}+\mu_0 m g_1 B\right)a_m-\frac{1}{2}\sum_M\langle njm|\mathbf{dF}^*|NJM\rangle b_M,$$
$$i\dot{b}_M=\left(-i\Gamma_M+\frac{\delta}{2}+\mu_0 M g_2 B\right)b_M-\frac{1}{2}\sum_m\langle NJM|\mathbf{dF}|njm\rangle a_m. \quad (3)$$

Here μ_0 is the Bohr magneton, $g_{1,2}$ are the gyromagnetic factors, and γ_m and Γ_M are the decay widths of the levels $|njm\rangle$ and $|NJM\rangle$.

If there is no magnetic field and the decay widths can be neglected, then the solutions of (1) can contain stationary-state superpositions whose energies remain unchanged in the resonance approximation. Consider, for

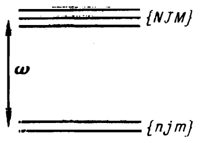

FIG. 1

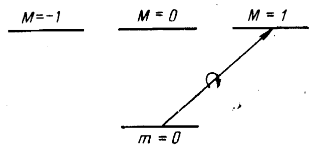

FIG. 2

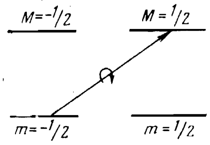

FIG. 3

example, the case $J = j + 1$. Then there exist solutions of the system (3)

$$a_m = 0, \quad b_M = b_M^{(0)} e^{-i\delta t/2},$$

where the constants $b_M^{(0)}$ satisfy the system of algebraic equations

$$\sum_M \langle njm|\mathbf{dF}^*|NJM\rangle b_M^{(0)} = 0, \quad m = -j, \ldots, j. \tag{4}$$

Since the number of equations is here $2j + 1$ and the number of unknowns is $2j + 3$, there always exist at least two sets of two constants $b_M^{(0)}$ satisfying Eqs. (4) at any polarization of the field.

In the case of circular polarization, the existence of such solutions is obvious from Fig. 2 with $j = 0$, $J = 1$ as an example: a right-polarized field does not resonate with the sublevels $M = 1$ and -1 of the upper level. In a field with linear polarization, if the quantization axis is chosen along the electric field intensity, the stationary sublevels in this case will be $M = \pm 1$. Nonetheless, the existence of unperturbed states at arbitrary field polarization is in some sense unexpected.

At $j = J + 1$ there exists a nontrivial solution

$$a_m = a_m^{(0)} e^{i\delta t/2}, \quad b_M = 0,$$

and there are in the general case two linearly independent sets of constants $a_m^{(0)}$ satisfying an algebraic system of equations of the type (4).

Of course, by making a definite choice of the polarization we can obtain more than two independent solutions corresponding to the stationary energies of the atom at $|j - J| = 1$, and also obtain nontrivial solutions at $j = J$. In a circularly-polarized field the solutions can in the latter case be easily constructed from physical considerations. Figure 3 shows the simplest example $j = J = \frac{1}{2}$: the levels $|N\frac{1}{2} - \frac{1}{2}\rangle$ and $|n\frac{1}{2}\frac{1}{2}\rangle$ are not perturbed by a right-polarized field.

For further transformation of the system (3), we use the Wigner-Eckart theorem

$$\langle NJM|\mathbf{dF}|njm\rangle = \sum_k C_{jm1k}^{JM} (-1)^k \langle NJ\|d\|nj\rangle F_{-k},$$

$$\langle njm|\mathbf{dF}^*|NJM\rangle = \sum_k C_{jm1k}^{JM} \langle NJ\|d\|nj\rangle^* F_k^*,$$

with the spherical projections of the vector $\mathbf{F}$ defined in the following manner:

$$F_0 = F_z, \quad F_{\pm 1} = \mp 2^{-1/2}(F_x \pm iF_y).$$

Putting $\langle NJ \| d \| nj\rangle \equiv d$, we obtain

$$\begin{aligned} i\dot{a}_m &= \left(-i\gamma_m - \frac{\delta}{2} + \mu_0 m g_1 B\right) a_m - \frac{d^*}{2} \sum_{kM} C_{jm1k}^{JM} F_k^* b_M, \\ i\dot{b}_M &= \left(-i\Gamma_M + \frac{\delta}{2} + \mu_0 M g_2 B\right) b_M - \frac{d}{2} \sum_{km} (-1)^k C_{jm1k}^{JM} F_{-k} a_m. \end{aligned} \tag{5}$$

To construct the general solution (5), we make up the vectors

$$\chi = (a_{-j} \ldots a_j b_{-J} \ldots b_J)^T$$

(T is the transpose symbol), after which Eq. (5) can be written in matrix form

$$i\dot{\chi} = Q\chi, \tag{6}$$

where Q is a matrix made up of the coefficients of the system (5):

$$\begin{gathered} Q_{mm'} = (-i\gamma_m - \delta/2 + \mu_0 m g_1 B)\delta_{mm'}, \quad Q_{MM'} = (-i\Gamma_M + \delta/2 + \mu_0 M g_2 B)\delta_{MM'}, \\ Q_{mM} = Q_{Mm}^* = -\frac{d^*}{2} \sum_k C_{jm1k}^{JM} F_k^*. \end{gathered} \tag{7}$$

We introduce $2(j + J + 1)$ linearly-independent normalized eigenvectors of this matrix:

$$Q\mathbf{f}^{(\alpha)} = q_\alpha \mathbf{f}^{(\alpha)}, \quad \alpha = 1, 2, \ldots, 2(j+J+1). \tag{8}$$

Some of the numbers q_α may coincide. Neglecting the probabilities of the spontaneous decays of the levels, the matrix (7) is self-adjoint and the vectors $\mathbf{f}^{(\alpha)}$ satisfy the completeness and orthogonality conditions:

$$\sum_{\mu = m, M} f_\mu^{(\alpha)} f_\mu^{(\alpha')*} = \delta_{\alpha\alpha'}, \quad \sum_\alpha f_\mu^{(\alpha)} f_{\mu'}^{(\alpha)*} = \delta_{\mu\mu'}. \tag{9}$$

Using (8), we write the general solution of (6) in the form

$$\chi(t) = \sum_\alpha C_\alpha \mathbf{f}^{(\alpha)} \exp(-iq_\alpha t),$$

where C_α are constants determined from the initial conditions. Substituting this formula in (2) we obtain

$$\begin{aligned} \psi(t) &= \sum_\alpha C_\alpha \exp[-i(\varepsilon_1 + q_\alpha + \delta/2)t]\,\Phi_\alpha(t), \\ \Phi_\alpha(t) &= \sum_{m=-j}^{j} f_m^{(\alpha)} |njm\rangle + e^{-i\omega t} \sum_{M=-J}^{J} f_M^{(\alpha)} |NJM\rangle. \end{aligned} \tag{10}$$

For example, if at the instant $t = 0$ the atom was in the state $|njm_0\rangle$, then the constants C_α, on the basis of (9), are given by $C_\alpha = f_{m0}(\alpha)^*$.

The quantities $\epsilon_1 + \delta/2 + q_\alpha$ determine the quasienergy spectrum of the atom in a periodic field, while Φ_α are the corresponding quasienergy wave functions. These functions contain with large weights, owing to the proposed resonance, not only the zeroth but also the first quasienergetic harmonics, in accordance with the physical meaning of the quasienergetic solutions of the Schrödinger equation (cf.[6,9]). This becomes spectroscopically manifest in the appearance of $2(j + J + 1)$ spectral states in the region of each of the unperturbed Zeeman multiplets, with the exception of the possible degeneracy cases, such as the ones referred to above.

The components of the vector $\mathbf{f}^{(\alpha)}$ are the weights of the unperturbed atomic states in the α-th quasienergy state. In particular, the constant component of the z-projection of the magnetic field of the atom in the state α is

$$M_z^{(\alpha)} = -\mu_0 \left[g_1 \sum_{m=-j}^{j} m|f_m^{(\alpha)}|^2 + g_2 \sum_{M=-J}^{J} M|f_M^{(\alpha)}|^2 \right]. \tag{11}$$

We consider now several examples, assuming for simplicity that $\gamma_m = \Gamma_M = 0$.

a) $j = J = 1/2$. The secular equation for the matrix **Q** is given by

$$\begin{vmatrix} -\frac{\delta}{2}-\frac{\mu_0 g_1 B}{2}-q & 0 & \frac{d^*}{2\sqrt{3}}F_0^* & \frac{d^*}{\sqrt{6}}F_1^* \\ 0 & -\frac{\delta}{2}+\frac{\mu_0 g_1 B}{2}-q & -\frac{d^*}{\sqrt{6}}F_{-1}^* & -\frac{d^*}{2\sqrt{3}}F_0^* \\ \frac{d}{2\sqrt{3}}F_0 & \frac{d}{\sqrt{6}}F_1 & \frac{\delta}{2}-\frac{\mu_0 g_2 B}{2}-q & 0 \\ -\frac{d}{\sqrt{6}}F_{-1} & -\frac{d}{2\sqrt{3}}F_0 & 0 & \frac{\delta}{2}+\frac{\mu_0 g_2 B}{2}-q \end{vmatrix} = 0, \tag{12}$$

where the rows and columns of the matrix are arranged in the order $m = -1/2$, $m = 1/2$, $M = -1/2$, $M = 1/2$. In a linearly polarized field vector **F** parallel to **B**, two noninteracting two-level systems appear, each of which is made up of states with identical magnetic quantum numbers. At $m = M = 1/2$ we have

$$q_{1,2} = -\frac{\mu_0 B}{4}(g_1+g_2) \pm \frac{1}{2}\left[\left(\delta+\frac{\mu_0 B}{2}(g_1-g_2)\right)^2+\frac{|d|^2}{3}I\right]^{1/2},$$

where $I = \mathbf{F}\cdot\mathbf{F}^*$ is the intensity of the wave; at $m = M = -1/2$, the roots differ from those presented above by reversal of the sign of B.

If the wave has nonzero components $F_{\pm1}$, then the magnetic quantum numbers are not conserved. For example, for a wave propagating along the magnetic field we have $F_0 = 0$ and the roots of Eq. (12) are

$$q_{1,2} = \frac{\mu_0 B}{4}(g_2-g_1) \pm \frac{1}{2}\left[\left[\delta+\frac{\mu_0 B}{2}(g_1+g_2)\right]^2+\frac{|d|^2}{3}I(1+A)\right]^{1/2}.$$

The eigenfunctions corresponding to these roots are, apart from the normalization,

$$|n\,{}^1/_2\,{-}^1/_2\rangle + e^{-i\omega t}\frac{dF_{-1}}{\sqrt{6}(\delta/2+\mu_0 g_2 B/2-q_{1,2})}|N\,{}^1/_2\,{}^1/_2\rangle.$$

Here $A = i(F_x F_y^* - F_y F_x^*)/I$ is the degree of circular polarization of the wave[10]: $A = 1$ and $A = -1$ correspond to right- and left-hand circular polarization, while $A = 0$ corresponds to linear polarization.

It should be noted that the incident field is assumed to be fully polarized, for in the case of partial polarization it is necessary to average over the realizations of the field of not the quasienergy spectrum, but of directly measurable quantities. This can give rise to correlation functions of second, third, etc. orders. Nonetheless, description of fully polarized radiation in terms of the Stokes parameters is quite convenient. We recall the corresponding definitions[10]:

$$|F_x|^2-|F_y|^2 = Il\cos 2\varphi, \quad F_xF_y^*+F_yF_x^* = Il\sin 2\varphi;$$

l is the degree of linear polarization, φ is the angle between the major axis of the polarization ellipse and the x axis, and $l^2 + A^2 = 1$.

The remaining roots and eigenfunctions of the matrix **Q** differ in the considered case from those given here by the substitutions

$$B\to -B, \quad A\to -A, \quad F_{-1}\to F_1,$$
$$|n\,{}^1/_2\,{-}^1/_2\rangle \to |n\,{}^1/_2\,{}^1/_2\rangle, \quad |N\,{}^1/_2\,{}^1/_2\rangle \to |N\,{}^1/_2\,{-}^1/_2\rangle.$$

b) $j = 0$, $J = 1$. The secular equation is in this case

$$\begin{vmatrix} -\delta/2-q & -d^*F_{-1}^*/2 & -d^*F_0^*/2 & -d^*F_1^*/2 \\ dF_1/2 & \delta/2-\mu_0 g_2 B-q & 0 & 0 \\ -dF_0/2 & 0 & \delta/2-q & 0 \\ dF_{-1}/2 & 0 & 0 & \delta/2+\mu_0 g_2 B-q \end{vmatrix} = 0. \tag{13}$$

The rows and columns of the matrix in (13) are in the sequence $m = 0$, $M = -1$, $M = 0$, $M = 1$.

The case of an external field that is linearly polarized along the z axis is trivial. The electric field mixes only sublevels with zero magnetic quantum numbers:

$$q_{1,2} = \pm 1/2(\delta^2+|dF_0|^2)^{1/2}.$$

The levels $M = \pm1$ remain unperturbed.

Of greater interest is the case when the wave propagates along the magnetic field: $F_0 = 0$. One root of (13), $q = \delta/2$, corresponds to the state $M = 0$, which is not perturbed now by the field. To find the remaining roots we transform (13) in the following manner

$$\begin{vmatrix} -\delta/2-q & |d|^2I(1-A)/8 & |d|^2I(1+A)/8 \\ 1 & \delta/2-\mu_0 g_2 B-q & 0 \\ 1 & 0 & \delta/2+\mu_0 g_2 B-q \end{vmatrix} = 0. \tag{14}$$

We consider now some particular cases in which Eq. (14) admits of simple solutions. At $B = 0$ the second root $q = \delta/2$ corresponds to the wave function

$$e^{-i\omega t}[l|N11\rangle + e^{2i\varphi}(1+A)|N1-1\rangle].$$

The two remaining roots $q = \pm 1/2(\delta^2 + |d|^2I)^{1/2}$ correspond to the wave functions

$$2F_1^*l(\delta-2q)|n00\rangle + dI(1+A)e^{-i\omega t}[l|N11\rangle + (A-1)|N1-1\rangle].$$

We note that from (14) we can obtain only the eigenvalues, and to find the eigenfunctions it is necessary to use the transformed matrix (13).

At a circular polarization $A = \pm1$, the field does not perturb the level $|N1 \mp 1\rangle$, which corresponds to the usual Zeeman energy value $\delta/2 \mp \mu_0 g_2 B$. Two other roots and the corresponding wave functions are

$$q = 1/2[A\mu_0 g_2 B \pm ((\delta + A\mu_0 g_2 B)^2 + |d|^2 I)^{1/2}],$$
$$\Phi = d^*I|n00\rangle + (\delta+2q)F_{-A}e^{-i\omega t}|N1A\rangle.$$

One more energy value, which does not depend on the electric field intensity, is produced when $\delta/\mu_0 g_2 B = A$. In this case the actions of the sublevels $M = n$ and $M = -1$ on the level $|n00\rangle$ cancel each other and the corresponding quasienergy coincides with the eigenvalue of the unperturbed atom Hamiltonian: $q = -\delta/2$. The corresponding wave function is of the form

$$|n00\rangle + \frac{1}{2}e^{-i\omega t}\left[\frac{dF_1}{\mu_0 g_2 B-\delta}|N1-1\rangle - \frac{dF_{-1}}{\mu_0 g_2 B+\delta}|N11\rangle\right].$$

The other roots of the secular equation and the wave functions can also be easily obtained at this ratio between the magnetic field intensity, the polarization, and the frequency deviation:

$$q = 1/2[\delta \pm (4(\mu_0 g_2 B)^2 + |d|^2 I)^{1/2}],$$
$$\Phi = |n00\rangle + e^{-i\omega t}\left[\frac{dF_1}{2\mu_0 g_2 B+2q-\delta}|N1-1\rangle - \frac{dF_{-1}}{2\mu_0 g_2 B-2q+\delta}|N11\rangle\right].$$

Figure 4 shows the quasienergies $E = \epsilon_1 + q + \delta/2$ for the case $F_0 = 0$ and $A = 1/2$, numerically obtained as functions of the electromagnetic wave intensity, $\Delta = \mu_0 g_2 B$ is the Zeeman splitting of the magnetic sub-

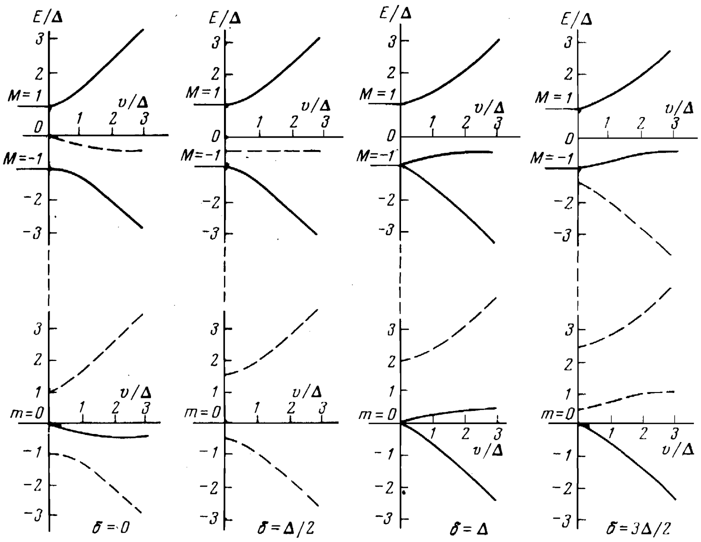

FIG. 4. Quasienergy levels of atomic states with angular momenta 0–1 for different frequency deviations at A = ½.

levels of the upper level in the absence of the wave, and $v = (|d|^2 I)^{1/2}$. The solid curves show the zeroth quasienergy harmonics, the weights of which in the quasienergy wave function tends to unity when the alternating field is adiabatically turned off, while the dashed curves show the first positive and negative harmonics, the weights of which tend in this case to zero. The position of the level M = 0 does not change when the field is turned on, and this level is not shown in the figure.

At $\delta = \Delta/2$, the condition $\delta/\mu_0 g_2 B = A$ is satisfied and the action of the magnetic sublevels M = ±1 on the level $|n00\rangle$ is mutually cancelled out, as a result of which the position of this level remains unchanged when the fields are turned on, and the corresponding zeroth quasienergy harmonic coincides with the abscissa axis. At $\delta = \Delta$, exact degeneracy of the two levels takes place in a weak field, and therefore both levels are shown by solid lines. It would be possible to establish the quantum numbers of these levels by going to the weak-field limit and introducing their widths, for then the complex energies would not coincide. We shall not dwell here on this problem, however.

3. ALTERNATING INVERSE FARADAY AND KERR EFFECTS

A light wave propagating in a resonant medium alters coherently the dipole moments of the atoms of the medium. The frequency of these changes coincides approximately with the frequency of the light, and a small difference leads to the so-called self-modulation broadening of the transmitted radiation[11]. No low-frequency component of the dipole moment appears in this case, since neither the ground nor the excited bound states of quantum systems with inversion centers have a constant dipole moment (except for the hydrogen atom, owing to the random degeneracy of its energy spectrum).

The medium behaves differently with respect to magnetization. As is well known, owing to the inverse Faraday effect, high-power radiation with nonzero degree of circular polarization produces in a medium a constant magnetization in the direction of its propagation[12]. For gases under ordinary conditions, however, this magnetization, according to (11), cannot exceed several oersteds and is therefore of no great interest.

In a resonant medium, on the other hand, an interesting singularity appears in the inverse Faraday effect, owing to the appearance of an alternating component of the magnetization. Its source, as will be seen later on, is closely connected with the presence in the atoms of the medium of constant magnetic moments, and also with the interference of different quasienergy states when the atoms are coherently excited by the incident radiation. The frequency of the variation of the magnetization coincides precisely with the frequency of the beats in the transitions between the interfering states.

Obviously, this alternating magnetization leads to a magnetic-dipole RF emission[8], the frequency of which can be varied by the intensity, polarization, and frequency of the optical wave, and also by the intensity of the external magnetic field in a sufficiently wide range.

Similarly, if even one of the resonating atomic levels has an angular momentum ≥1, then this level has a nonzero quadrupole electric moment. In a gas of such atoms, the optical radiation produces both a dc and ac emf—the inverse Kerr effect. We note that the dc emf produced, for example, in the case of ferromagnetic resonance in solids, was observed experimentally (see[13], where references to earlier research can also be found).

We start the analysis with the inverse Faraday effect. If the state of the atoms of the medium is described by the density matrix $\rho_{\alpha\alpha'}$ of the quasienergy levels, then the average value of the s-projection of the magnetic moment is

$$M_s(t)=\sum_{\alpha\alpha'}\rho_{\alpha\alpha'}\langle\Phi_{\alpha'}|\mu_s|\Phi_\alpha\rangle\exp\{i(q_{\alpha'}-q_\alpha)t\}, \quad (15)$$

Substituting (10) in (15), we obtain

$$M_s(t)=-\mu_0[g_1(j(j+1))^{1/2}P_{j1s}(t)+g_2(J(J+1))^{1/2}P_{J1s}(t)], \quad (16)$$

where the time-dependent polarization parameters P are defined as follows:

$$P_{IL\tau}(t)=\sum_{\substack{\alpha\alpha'\\ \mu\mu'}}\rho_{\alpha\alpha'}\exp\{i(q_{\alpha'}-q_\alpha)t\}C^{I\mu'}_{I\mu L\tau}f_{\mu'}^{(\alpha')}f_\mu^{(\alpha)}; \quad (17)$$

where $(I\mu\mu') = (jmm')$ or (JMM').

It is possible to calculate in the same manner also the quadrupole moment of the atom. The quadrupole-moment operator, as is well known, is of the form

$$\hat{Q}_{ik}=\frac{3Q_I}{2I(2I-1)}\left(\hat{I}_i\hat{I}_k+\hat{I}_k\hat{I}_i-\frac{2}{3}\hat{I}^2\delta_{ik}\right),$$

where I is the total angular momentum operator and Q_I is the quadrupole moment, defined as the average value of the operator $\hat{Q}_{zz}$ in a state $M_I = I$ ($I \geq 1$). In this case it is more convenient to calculate not the Cartesian but the spherical projections of the quadrupole moment $\hat{Q}_{ps}$, p, s = 0, ±1. The connection between the two coordinate systems can be easily established. For example

$$\hat{Q}_{xx}={}^1/_2(\hat{Q}_{11}+\hat{Q}_{-1-1})-\hat{Q}_{1-1},\quad \hat{Q}_{xy}=-{}^1/_2 i(\hat{Q}_{11}-\hat{Q}_{-1-1})$$

etc. Standard calculations yield

$$Q_{ps}=\left\{\frac{j+1}{2j-1}Q_j[(-1)^s\delta_{p,-s}-\delta_{ps}]P_{j00}(t)\right.$$
$$\left.+(-1)^{p+s}\frac{3(j+1)}{2j-1}[5(2j+1)]^{1/2}Q_jC^{2p+s}_{1p1s}W(j1j1;j2)P_{j2p+s}(t)\right\} \quad (18)$$
$$+\{\text{the same with the substitution } j\to J\}.$$

As seen from (16)—(18), the frequency of the variation of the magnetic-dipole and electric-quadrupole moments of the atom is determined by the difference of the quasienergies. The frequency ~10 GHz is reached at a quasienergy splitting ~1 cm^{-1}, which sets in even in relatively weak fields ~10^4 V/cm. It should be noted, however, that the thermal motion of the gas atoms gives rise to a Doppler distribution of the frequency deviations from resonance in individual atoms, and the width of the Doppler distribution, which is usually much larger than the energy of the interaction with the wave field, will determine the splitting of the quasienergies. The formulas given above are valid only for an atomary beam in the case of propagation of an electromagnetic wave.

A simple analysis based, for example, on the particular cases considered in Sec. 2, shows in particular that the alternating magnetization of the atoms is produced both in the absence of a magnetic field at a nonzero degree of circular polarization of the radiation (as in the usual inverse Faraday effect[12]), and in a field that is linearly polarized but in the presence of a constant external magnetic field whose direction does not coincide with the wave-polarization direction. Certain numerical estimates for the intensity of the magnetic dipole radio emission of a laser focus are given in[8].

4. CORRELATION AND POLARIZATION CHARACTERISTICS OF SPONTANEOUS EMISSION

We consider spontaneous electromagnetic transitions of an atom from a quasienergy state α into a certain unperturbed state $|\nu I\mu\rangle$. We assume first that the eigenvalue q_α is not degenerate and the magnetic field differs from zero, so that the Zeeman sublevels $|\nu I\mu\rangle$ have different energies, and the detector resolution is sufficient for the observation of the individual line components. In this case the amplitude of the probability of registering a photon with momentum **k** and polarization $\mathbf{e}_s$ (s = ±1) is proportional to the matrix element

$$\langle \nu I\mu|\mathbf{j}\mathbf{e}_s^* e^{-i\mathbf{kr}}|\Phi_\alpha\rangle,$$

where **j** is the current-density operator. The plane wave that enters here is conveniently represented in the form

$$\mathbf{e}_s e^{i\mathbf{kr}} = -s\sum_{\Lambda l t} i^l[4\pi(2l+1)]^{1/2} j_l(kr) C^{\Lambda s}_{l01s} D_{ts}{}^{\Lambda}(\mathbf{k})\mathbf{Y}_{\Lambda l t}(\mathbf{r}),$$

where j_l are spherical Bessel functions, D(**k**) is a finite-rotation matrix that rotates the coordinate system through the angles of the vector **k** that correspond to the Euler angles (φ, θ, 0) relative to a fixed coordinate system with z axis directed along **B**; **Y** are spherical vectors[14].

Since interest attaches only to electric dipole transitions, it is necessary to retain in the foregoing formula only the term with l = 0. The operator **j** is then proportional to the dipole-moment operator **d**. We assume for the sake of argument that the considered transition to the levels $|\nu I\rangle$ is allowed only from the levels $|NJ\rangle$. The polarization density matrix of the spontaneous emission is then, apart from normalization,

$$\rho_{ss'}(\alpha,\mu;\mathbf{k}) = ss'\sum_{tt'}\langle \nu I\mu|\mathbf{d}\mathbf{Y}^*_{10t'}|\Phi_\alpha\rangle\langle\Phi_\alpha|\mathbf{d}\mathbf{Y}_{10t}|\nu I\mu\rangle D^{1*}_{t's'}(\mathbf{k})D_{ts}{}^1(\mathbf{k})$$
$$= ss'\sum_{L\tau\sigma}|\langle NJ\|d\|\nu I\rangle|^2 C^{L\sigma}_{1-s'1s} D_{\tau\sigma}{}^L(\mathbf{k})F_{L\tau}(\alpha,\mu); \tag{19}$$

$$F_{L\tau}(\alpha,\mu) = -\sum_{MM'tt'}(-1)^{t'}C^{L\tau}_{1-t'1t}C^{JM'}_{I\mu1t'}C^{JM}_{I\mu1t}f^{(\alpha)}_{M'}f^{(\alpha)*}_{M}.$$

The angular distribution of the radiation is determined by the trace of this matrix. Leaving out the angle-independent factors, we obtain

$$W(\mathbf{k}) = F_{00} + (2\pi/5)^{1/2}\sum_\tau F_{2\tau}Y_{2\tau}(\mathbf{k}). \tag{20}$$

Formula (19) makes it also possible to calculate the Stokes parameters ξ_i of the spontaneous radiation. In spherical unit vectors, the normalized density matrix of the radiation is expressed in terms of the Stokes parameters in the following manner:

$$\rho_{ss'} = \frac{1}{2}\begin{pmatrix} 1-\xi_2 & -\xi_3-i\xi_1 \\ -\xi_3+i\xi_1 & 1+\xi_2\end{pmatrix}.$$

Using (19), we obtain

$$\xi_1 = \frac{i\sqrt{3}}{2W(\mathbf{k})}\sum_\tau F_{2\tau}[D^2_{\tau-2}(\mathbf{k}) - D_{\tau 2}{}^2(\mathbf{k})],$$
$$\xi_2 = \frac{\sqrt{2\pi}}{W(\mathbf{k})}\sum_\tau F_{1\tau}Y_{1\tau}(\mathbf{k}), \tag{21}$$
$$\xi_3 = \frac{\sqrt{3}}{2W(\mathbf{k})}\sum_\tau F_{2\tau}[D_{\tau 2}{}^2(\mathbf{k}) + D^2_{\tau-2}(\mathbf{k})].$$

The quantity ξ_2 determines, in particular, the degree of circular polarization of the spontaneous emission[10].

We consider now the case of a weak magnetic field, when the Zeeman components $|\nu I\mu\rangle$ are not resolved by the detector. It is now necessary to sum in the obtained formulas (19)—(21) over μ, and this, as is readily seen, reduces to summation over μ of only the parameters $F_{L\tau}(\alpha,\mu)$. With the aid of known formulas we easily obtain

$$F_{L\tau}(\alpha) \equiv \sum_\mu F_{L\tau}(\alpha,\mu) = (-1)^L[(2L+1)(2J+1)]^{1/2}W(L1JI;1J)P^{(\alpha)}_{L\tau}$$
$$P^{(\alpha)}_{L\tau} = \sum_{MM'} C^{JM'}_{JML\tau} f^{(\alpha)}_M f^{(\alpha)*}_{M'}. \tag{22}$$

It is precisely these values of $F_{l\tau}$ that must be substituted in (20) and (21) when **B** is small. The parameters $P^{(\alpha)}_{L\tau}$ are the polarization moments of the atom[14] in the quasienergy state α in accordance with the previously-discussed physical meaning of the coefficients $f^{(\alpha)}_M$.

Finally, a possible case, most frequently encountered in experiment, is that of a weak pumping field, when the line components corresponding to emission at different quasienergy states are not resolved. The spontaneous-emission polarization density matrix is then determined by the density matrix $\rho_{\alpha\alpha'}$ of the quasienergy levels of the atoms:

$$\rho_{ss'}(\mathbf{k}) = ss'\sum_{\alpha\alpha'\mu tt'}\langle \nu I\mu|\mathbf{d}\mathbf{Y}^*_{10t'}|\Phi_\alpha\rangle\rho_{\alpha\alpha'}\langle\Phi_{\alpha'}|\mathbf{d}\mathbf{Y}_{10t}|\nu I\mu\rangle D^{1*}_{t's'}(\mathbf{k})D_{ts}{}^1(\mathbf{k}).$$

For the angular distribution and polarization of the radiation we obtain the same formulas (20)—(22), but the polarization parameters of the atom are determined now by the relation (17).

In the case of equal population of the quasienergy states, at a random relative phase

$$\rho_{\alpha\alpha'} = \delta_{\alpha\alpha'}/2(J+j+1) \tag{23}$$

the radiation, as can be easily verified, is isotropic and fully unpolarized. Formula (23) can occur in stationary regimes, when the level $|nj\rangle$ is a ground or metastable level, and the pumping acts for a prolonged period. During that time, the coherence present in the atomic ensemble is lost because of collision or incoherent radiative processes. A necessary condition is also the mixing of all the sublevels $|njm\rangle$ and $|NJM\rangle$, without exception, by the fields B and E. For example, in the cases shown in Figs. 2 and 3, this condition is not satisfield.

At the present time, no spontaneous scattering of high-power electromagnetic radiation by atoms has been observed experimentally, since the employed lasers operate in the pulsed regime. Under these conditions, the spontaneous emission has too low an intensity to be registered[4]. However, the formulas obtained in this section may prove to be useful also in the investigation of stimulated emission, since they make it possible to calculate the direction and polarization of the stimulated scattering at arbitrary polarization of the incident radiation and at arbitrary orientation of this radiation relative to the external magnetic field. Namely, if the laser radiation is focused into a sphere, then the lowest threshold of the stimulated scattering will be in that direction in which the function $W(\mathbf{k})$ [Eq. (20)] has a maximum value. The polarization of this radiation will be right-handed or left-handed, depending on the sign of ξ_2.

[1)]We use here a system of units in which $c = \hbar = 1$.

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Translated by J. G. Adashko
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