THEORY OF RESONANCE SCATTERING OF LIGHT BY A GAS IN THE PRESENCE OF A MAGNETIC FIELD

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Formulas for the intensity of the light scattered by a gas in a magnetic field are derived for arbitrary total angular momenta of the normal and excited states. A number of resonance effects of the double-resonance type are treated for arbitrary angular momenta, and the relaxation times that appear in these effects are determined.

1. In recent years there have been detailed studies, both experimental and theoretical, of the resonance effects that appear when light is scattered by the atoms of a gas subjected to a magnetic field. Examples of such experiments are those on double resonance,[1,2] parametric resonance,[3] the intersection of levels,[4] and so on. In all such experiments one observes an increase of the intensity of the scattered light when certain conditions are satisfied (for example, when the Larmor precession frequency of the magnetic moment of the atom is equal to the frequency of an alternating magnetic field[1] or to the frequency of modulation of the incident light[5]). For low gas pressures the width of the resonance curve is determined by the natural lifetime of the excited state. When the pressure is increased the width at first decreases owing to diffusion of the radiation, and thereafter increases owing to binary collisions of excited atoms with normal atoms, accompanied by transfer of excitation. These processes have been studied theoretically (see[6,7]). In these papers it is shown that there are two relaxation times γ_1^{-1} and γ_2^{-1}—the respective relaxation times of circular and linear polarizations—and explicit expressions for these times are derived.

The theoretical treatment of effects of the double-resonance type has usually been confined to the case in which the total angular momentum of the excited level of the atom is equal to unity, and that of the lower level is zero. The purpose of the present paper is to derive formulas for the intensity and polarization of the light scattered by a gas in a magnetic field for arbitrary total angular momenta of the upper and lower levels (Secs. 3 and 4). In Secs. 5—8 we consider certain resonance effects for arbitrary angular momenta, and also indicate in what sort of experiments the relaxation times γ_1^{-1} and γ_2^{-1} can be observed.

2. Let a gas be placed in a uniform magnetic field and illuminated with light of the resonance line. We shall assume that the spectral composition of the light is much broader than the Zeeman splitting of the excited level. We denote by j_1 and j_0 the respective total angular momenta of the atom in the excited and ground states. We shall number the Zeeman sublevels of the excited state with an index m, and those of the ground state with an index μ. The intensity $I_{\mathbf{k}\lambda}$ of the light scattered in the direction **k** with the polarization λ (λ = 1, 2) is

$$I_{\mathbf{k}\lambda} = I_0' \sum_{mm'\mu} f_{mm'} (\mathbf{e}_{\mathbf{k}\lambda} \mathbf{d}_{m'\mu}) (\mathbf{e}_{\mathbf{k}\lambda} \mathbf{d}_{m\mu})^*, \tag{1}$$

where I_0' is a proportionality constant, $f_{mm'}$ is the density matrix of the excited atoms, $\mathbf{e}_{\mathbf{k}\lambda}$ is the unit vector of the polarization, and $\mathbf{d}_{m\mu}$ is the matrix element of the dipole-moment operator of the atom.

In the presence of a magnetic field the density matrix $f_{mm'}$ satisfies the equation

$$df_{mm'}/dt = -i[V, f]_{mm'} - \sum_{m_1m_1'} \Gamma_{mm'}^{m_1m_1'} f_{m_1m_1'} + \mathscr{F}_{mm'}. \tag{2}$$

Here $V = \mu_0 g\mathbf{H}\cdot\mathbf{J}$ is the energy operator of the interaction between the atom and the external magnetic field **H**, μ_0 is the Bohr magneton, g is the g-factor, and **J** is the operator of the total angular momentum of the atom. The matrix Γ describes the relaxation processes, and the matrix $\mathscr{F}$ describes the excitation of the atoms by the external light source. If the ground-state sublevels are equally populated, then, as has been shown in a paper by Konstantinov and Perel',[8]

$$\mathscr{F}_{mm'} = F_0' \sum_{\mu} (\mathbf{d}_{m\mu}\mathbf{e}) (\mathbf{d}_{m'\mu}\mathbf{e})^*, \tag{3}$$

where F_0' is a proportionality constant and **e** is a unit vector proportional to the complex amplitude of the electric field in the exciting light.

3. To solve the equations in (2) and find the in-

tensity (1) of the scattered light we use the expansion of the density matrix in terms of irreducible tensor operators[9]:

$$\hat{f} = \sum_{\varkappa=0}^{2j_1} \sum_{q=-\varkappa}^{\varkappa} (-1)^q f_q^{\varkappa} \hat{T}_{-q}^{\varkappa}. \quad (4)$$

The operators $\hat{T}_q^{\kappa}$ are normalized so that

$$(T_q^{\varkappa})_{mm'} = (-1)^{j_1-m'} \frac{2\varkappa+1}{(2j_1+1)^{1/2}} \begin{pmatrix} j_1 & \varkappa & j_1 \\ -m & q & m' \end{pmatrix}. \quad (5)$$

Here we have introduced the Wigner 3j symbol. Then, in view of the orthogonality of the 3j symbols,

$$\sum_{mm'} (T_q^{\varkappa})_{mm'} (T_{q_1}^{\varkappa_1})_{mm'} = \frac{2\varkappa+1}{2j_1+1} \delta_{qq_1} \delta_{\varkappa\varkappa_1} \quad (6)$$

By means of the orthogonality relation (6) it is easy to express the f_q^{κ} in terms of the $f_{mm'}$:

$$f_q^{\varkappa} = (-1)^q \frac{2j_1+1}{2\varkappa+1} \sum_{mm'} f_{mm'} (T_{-q}^{\varkappa})_{mm'}. \quad (7)$$

The change to the quantities f_q^{κ} is very advantageous, since they have a more direct physical meaning than the matrix elements $f_{mm'}$. For example, $f_0^0 = \mathrm{Sp}\hat{f}$ is the population of the excited level, and f_q^1 is equal apart from a constant factor to the average value of the qth circular component of the total angular momentum of the excited atoms. In fact, if we introduce the circular components of a vector by the rule

$$A_1 = -(A_x + iA_y)/\sqrt{2}, \quad A_0 = A_z, \quad A_{-1} = (A_x - iA_y)/\sqrt{2},$$

then by the Wigner-Eckart theorem[9] we have

$$(J_q)_{mm'} = (-1)^{j_1-m} [(2j_1+1)(j_1+1)j_1]^{1/2} \begin{pmatrix} j_1 & 1 & j_1 \\ -m & q & m' \end{pmatrix}. \quad (8)$$

Then, using (4), (5) and (8), we get

$$\langle J_q \rangle = \mathrm{Sp}\hat{f}\hat{J}_q = (-1)^q [j_1(j_1+1)]^{1/2} f_q^1.$$

As will be seen later, the change to the representation (κ, q) allows us to distinguish the dependences of the scattered light intensity (1) on the angular momenta j_0 and j_1, and also on the directions and polarizations of the exciting and scattered beams.

By means of (4)—(8) we can easily get from (2) the equation for the quantities f_q^{κ}:

$$df_q^{\varkappa}/dt = i\mu_0 g \{[(\varkappa+q)(\varkappa-q+1)/2]^{1/2} H_1 f_{q-1}^{\varkappa} + qH_0 f_q^{\varkappa}$$
$$-[(\varkappa-q)(\varkappa+q+1)/2]^{1/2} H_{-1} f_{q+1}^{\varkappa}\} - \sum_{\varkappa_1 q_1} \Gamma_{qq_1}^{\varkappa\varkappa_1} f_{q_1}^{\varkappa_1} + \mathcal{F}_q^{\varkappa}. \quad (9)$$

Here

$$\Gamma_{qq_1}^{\varkappa\varkappa_1} = (2\varkappa_1+1) \sum_{\substack{mm' \\ m_1m_1'}} (-1)^{m-m_1} \begin{pmatrix} j_1 & j_1 & \varkappa \\ m & -m' & q \end{pmatrix} \times \Gamma_{mm'}^{m_1m_1'} \begin{pmatrix} j_1 & j_1 & \varkappa_1 \\ m_1 & -m_1' & q_1 \end{pmatrix},$$

$$\mathcal{F}_q^{\varkappa} = (2j_1+1)^{1/2} \sum_{mm'} (-1)^{m-j_1} \begin{pmatrix} j_1 & j_1 & \varkappa \\ m & -m' & q \end{pmatrix} \mathcal{F}_{mm'}. \quad (10)$$

Without its relaxation and excitation terms Eq. (9) is a special case of the equation derived by Fano[10] for an inhomogeneous field. The operators $\hat{T}_q^{\kappa}$ differ from the operators chosen by Fano by the factor $(2j_1+1)^{1/2}/(2\kappa+1)^{1/2}$ and replacement of q by $-q$.

An important advantage of the representation (κ, q) is the fact that in this representation the matrix Γ is diagonal in important cases and does not depend on q and q_1. This occurs in the diffusion of radiation in the case of complete capture, and also in relaxation owing to binary collisions of excited atoms with normal atoms.[7] Therefore in what follows we set

$$\Gamma_{qq_1}^{\varkappa\varkappa_1} = \gamma_{\varkappa} \delta_{\varkappa\varkappa_1} \delta_{qq_1}$$

The equation (9) then takes the form

$$df_q^{\varkappa}/dt = i\mu_0 g \{[(\varkappa+q)(\varkappa-q+1)/2]^{1/2} H_1 f_{q-1}^{\varkappa} + qH_0 f_q^{\varkappa}$$
$$-[(\varkappa-q)(\varkappa+q+1)/2]^{1/2} H_{-1} f_{q+1}^{\varkappa}\} - \gamma_{\varkappa} f_q^{\varkappa} + \mathcal{F}_q^{\varkappa}. \quad (11)$$

It is remarkable that the equations with different κ can be separated. Therefore the number of coupled equations for a given κ is $2\kappa+1$ and does not depend on j_1 and j_0. This fact makes it easy to examine the general case of arbitrary j_1 and j_0.

4. Let us substitute the value of $\mathcal{F}_{mm'}$ given by (3) into the formula (10). Then, when we use the expression for the matrix elements of the dipole moment operator in terms of 3j symbols and the reduced matrix elements $(j_1 \| d \| j_0)$,

$$(d_q)_{m\mu} = (-1)^{j_1-m} \begin{pmatrix} j_1 & 1 & j_0 \\ -m & q & \mu \end{pmatrix} (j_1 \| d \| j_0),$$

after some manipulations we get

$$\mathcal{F}_q^{\varkappa} = (-1)^{j_1+j_0} F_0 \begin{Bmatrix} 1 & 1 & \varkappa \\ j_1 & j_1 & j_0 \end{Bmatrix} \Phi_q^{\varkappa}(\mathbf{e}). \quad (12)$$

Here $F_0 = (2j_1+1)^{1/2} |(j_1 \| d \| j_0)|^2 F_0'$ is a new constant, and the expression in curly brackets is a 6j symbol. Also we have introduced the function

$$\Phi_q^{\varkappa}(\mathbf{e}) = \sum_{q_1q_2} (-1)^{q_2} e_{q_1} (e_{q_2})^* \begin{pmatrix} 1 & 1 & \varkappa \\ q_1 & -q_2 & -q \end{pmatrix}, \quad (13)$$

where e_q are the circular components of the polarization vector $\mathbf{e}$ of the exciting light. In the derivation of the expression (12) we have used the symmetry properties of the 3j symbols,[9] and also the formula

$$\sum_{m'}(-1)^{j'+m'}\begin{pmatrix} j_1 & j_2 & j' \\ m_1 & m_2 & m' \end{pmatrix}\begin{pmatrix} j_3 & j_4 & j' \\ m_3 & m_4 & -m' \end{pmatrix}$$
$$= \sum_{jm}(-1)^{2j_4+j+m}(2j+1)\begin{Bmatrix} j_1 & j_2 & j' \\ j_3 & j_4 & j \end{Bmatrix}$$
$$\times \begin{pmatrix} j_3 & j_2 & j \\ m_3 & m_2 & m \end{pmatrix}\begin{pmatrix} j_1 & j_4 & j \\ m_1 & m_4 & -m \end{pmatrix}.$$

Let us now express the intensity $I_{k\lambda}$ of the scattered light in terms of the quantities f_q^κ. By means of Eqs. (1), (4), (5) we find

$$I_{k\lambda} = (-1)^{j_1+j_0} I_0 \sum_\varkappa (2\varkappa+1)\begin{Bmatrix} 1 & 1 & \varkappa \\ j_1 & j_1 & j_0 \end{Bmatrix}\sum_q (-1)^q f_q^\varkappa \Phi_{-q}^\varkappa(\mathbf{e}_{k\lambda}), \tag{14}$$

where

$$I_0 = (2j_1+1)^{-1/2}|(j_1\|d\|j_0)|^2 I_0'$$

As is well known,[9] the 6j symbol

$$\begin{Bmatrix} j_1 & j_2 & j_3 \\ l_1 & l_2 & l_3 \end{Bmatrix}$$

is different from zero only if the sets of three numbers (j_1, j_2, j_3), (j_1, l_2, l_3), (j_2, l_1, l_3), and (l_1, l_2, j_3) satisfy the triangle condition. Thus the 6j symbol that appears in (12) and (14) is different from zero only if the following inequalities are satisfied:

$$|j_0 - 1| \leqslant j_1 \leqslant j_0 + 1, \quad 0 \leqslant \varkappa \leqslant 2j_1, \quad 0 \leqslant \varkappa \leqslant 2.$$

The first inequality is the selection rule for a dipole transition. The second is automatically satisfied in the expansion (4). Finally, the third inequality shows that for excitation by light the quantities $\mathcal{F}_q$ exist only for $\kappa = 0, 1, 2$, and that there are contributions to the intensity $I_{k\lambda}$ of the scattered light only from the quantities

$$f_0^0, \ f_q^1 \ (q = 0, \pm 1) \ \text{and} \ f_q^2 \ (q = 0, \pm 1, \pm 2).$$

From the fact that the density matrix $f_{mm'}$ is Hermitian it follows that

$$f_{-q}^\varkappa = (-1)^q (f_q^\varkappa)^* \tag{15}$$

and therefore to find the intensity $I_{k\lambda}$ it is sufficient to know only six quantities f_q^κ ($\kappa = 0, 1, 2$; $q \geq 0$), which can be determined from (11). We note that, according to the definition (13), the function $\Phi_q^\kappa(\mathbf{e})$ has a property analogous to (15):

$$\Phi_{-q}^\varkappa(\mathbf{e}) = (-1)^q[\Phi_q^\varkappa(\mathbf{e})]^*. \tag{16}$$

The function Φ_q^κ which appears in (12) and (14) determines which of the $\mathcal{F}_q$ occur for excitation by light incident in a given direction and with a given polarization, and which of the f_q^κ contribute to the intensity $I_{k\lambda}$ of the scattered light.

If the exciting or observed light is linearly polarized in the direction with the polar angles (θ, φ), then by using the explicit values of the appropriate 3j symbols we find from the definition (13)

$$\Phi_0^0 = -1/\sqrt{3}, \quad \Phi_q^1 = 0, \quad \Phi_0^2 = (3\cos^2\theta - 1)/\sqrt{30},$$
$$\Phi_1^2 = \sin\theta\cos\theta \, e^{i\varphi}/\sqrt{5}, \quad \Phi_2^2 = \sin^2\theta \, e^{2i\varphi}/2\sqrt{5}. \tag{17}$$

For $q < 0$ the values of the functions Φ_q^κ can be obtained from (17) by means of the relations (16).

For light propagated in the direction defined by the polar angles (θ_1, φ_1) and with right-circular polarization, we have

$$e_1 = {}^1/_2(1 - \cos\theta_1)e^{i\varphi_1}, \quad e_0 = -2^{-1/2}\sin\theta_1,$$
$$e_{-1} = {}^1/_2(1+\cos\theta_1)e^{-i\varphi_1}. \tag{18}$$

Substituting these components in (13), we get

$$\Phi_0^0 = -\frac{1}{\sqrt{3}}, \quad \Phi_0^1 = \frac{\cos\theta_1}{\sqrt{6}}, \quad \Phi_1^1 = \frac{\sin\theta_1}{2\sqrt{3}}e^{i\varphi_1},$$
$$\Phi_0^2 = -\frac{3\cos^2\theta_1 - 1}{2\sqrt{30}}, \quad \Phi_1^2 = -\frac{\sin\theta_1\cos\theta_1}{2\sqrt{5}}e^{i\varphi_1},$$
$$\Phi_2^2 = -\frac{\sin^2\theta_1}{4\sqrt{5}}e^{2i\varphi_1}. \tag{19}$$

Accordingly Φ_0^0 is different from zero for any polarization and any direction of propagation. The functions Φ_q^1 ($q = 0, \pm 1$) are different from zero only for circular polarization. As can be seen from (17) and (19), in both cases the functions Φ_q^κ are proportional to the spherical functions Y_q^κ. The proportionality factors, however, depend on κ and q.

5. Double resonance.[1,2] The constant magnetic field $\mathcal{H}_0$ is directed along the z axis. In addition, an alternating magnetic field $\mathcal{H}_1$ is applied in a perpendicular direction. Near resonance only one of the circular components of the magnetic field is important. Therefore we shall, as usual, regard the field $\mathcal{H}_1$ as rotating in the xy plane:

$$\mathcal{H}_{1x} = \mathcal{H}_1\cos\omega t, \quad \mathcal{H}_{1y} = \mathcal{H}_1\sin\omega t.$$

Let the excitation be produced by light of constant intensity polarized, for example, along the z axis. Then from (12) and (17) we find that only the quantities $\mathcal{F}_0^0$ and $\mathcal{F}_0^2$ are different from zero

$$\mathcal{F}_0^0 = (-1)^{j_1+j_0+1}F_0\frac{1}{\sqrt{3}}\begin{Bmatrix} 1 & 1 & 0 \\ j_1 & j_1 & j_0 \end{Bmatrix},$$
$$\mathcal{F}_0^2 = (-1)^{j_1+j_0}F_0\frac{2}{\sqrt{30}}\begin{Bmatrix} 1 & 1 & 2 \\ j_1 & j_1 & j_0 \end{Bmatrix}. \tag{20}$$

It is now easy to get the solution of the equations (11) that corresponds to the stationary condition. Substituting in (11) the circular components of the magnetic field

$$H_1 = -\mathcal{H}_1 e^{i\omega t}/2, \quad H_0 = \mathcal{H}_0, \quad H_{-1} = \mathcal{H}_1 e^{-i\omega t}/\sqrt{2}$$

and setting $f_q^\kappa \sim e^{iq\omega t}$, we solve the resulting system of algebraic equations. The final result is

$$f_0^0 = \mathcal{F}_0^0/\gamma_0, \quad f_q^1 = 0,$$

$$f_0^2 = \frac{\mathcal{F}_0^2}{\gamma_2} - \frac{\mathcal{F}_0^2}{\gamma_2}\frac{3\Omega_1^2(4\delta^2+\Omega_1^2+\gamma_2^2)}{(\delta^2+\gamma_2^2+\Omega_1^2)(4\delta^2+\gamma_2^2+4\Omega_1^2)},$$

$$f_1^2 = -\frac{\mathcal{F}_0^2}{\gamma_2}\sqrt{\frac{3}{2}} \times \Omega_1\frac{(2\delta-i\gamma_2)[(\delta+i\gamma_2)(2\delta+i\gamma_2)-\Omega_1^2]}{(\delta^2+\gamma_2^2+\Omega_1^2)(4\delta^2+\gamma_2^2+4\Omega_1^2)}e^{i\omega t},$$

$$f_2^2 = \frac{\mathcal{F}_0^2}{\gamma_2}\sqrt{\frac{3}{2}}\Omega_1^2\frac{(\delta+i\gamma_2)(2\delta+i\gamma_2)-\Omega_1^2}{(\delta^2+\gamma_2^2+\Omega_1^2)(4\delta^2+\gamma_2^2+4\Omega_1^2)}e^{2i\omega t} \tag{21}$$

Here we have introduced the notations $\Omega = \mu_0 g \mathcal{H}_0$, $\Omega_1 = \mu_0 g \mathcal{H}_1$, $\delta = \omega - \Omega$.

As can be seen from (21), the quantities f_q^2 and the associated components of the intensity of the scattered radiation have resonance maxima at $\delta = 0$. The intensity associated with f_0^0 and with the first term in f_0^2 is a constant background. Substituging the quantities (21) in the expression (14), we can easily get the intensity $I_{k\lambda}$ of the scattered light. In the case $j_1 = 1$, $j_0 = 0$ the formula for the intensity has been derived by Dodd and Series.[2]

We present the ratio of the signal of the unmodulated component in the resonance (the second term in f_0^2) to the background, for the case of observation of light polarized along the z axis. For simplicity we take $\Omega_1 \ll \gamma_2$ (absence of saturation) and $\gamma_2 = \gamma_0 = \gamma$ (low pressures). Then the ratio in question is

$$3\left[1+\frac{40(2j_1-1)j_1(j_1+1)(2j_1+3)}{\{3X(X-1)-8j_1(j_1+1)\}^2}\right]^{-1}\frac{\Omega_1^2}{\gamma^2},$$

where $X = (j_1 - j_0)(j_1 + j_0 + 1) + 2$. Here we have used the values of the 6j symbols given by Edmonds.[9] This ratio decreases rapidly with increase of j_1 and j_0.

Let us also consider the question of the possibility of observing the relaxation time γ_1^{-1} in experiments on double resonance. This relaxation time is manifested if components associated with f_q^1 are observed in the scattered light. It follows from (12), (14), and (19) that this is possible only if circular polarization is used both in the excitation and in the observation. Let us consider the case in which the exciting light is propagated along the constant magnetic field (the z axis) and is circularly polarized in the xy plane. Observation of the light circularly polarized in the yz plane is made along the x axis. Equations (12) and (19) show that the only ones of the quantities $\mathcal{F}_q$ that are different from zero are the following:

$$\mathcal{F}_0^0 = (-1)^{j_1+j_0+1}F_0\frac{1}{\sqrt{3}}\begin{Bmatrix}1 & 1 & 0\\ j_1 & j_1 & j_0\end{Bmatrix},$$

$$\mathcal{F}_0^1 = (-1)^{j_0+j_1}F_0\frac{1}{\sqrt{6}}\begin{Bmatrix}1 & 1 & 1\\ j_1 & j_1 & j_0\end{Bmatrix},$$

$$\mathcal{F}_0^2 = (-1)^{j_0+j_1+1}F_0\frac{1}{\sqrt{30}}\begin{Bmatrix}1 & 1 & 2\\ j_1 & j_1 & j_0\end{Bmatrix}. \tag{22}$$

The equations (11) for $\kappa = 1$ are essentially the Bloch equations for the magnetic moment of the atom. The time γ_1^{-1} is here the time of both longitudinal and transverse relaxation. From (11) we find

$$f_0^1 = \frac{\mathcal{F}_0^1}{\gamma_1} - \frac{\mathcal{F}_0^1}{\gamma_1}\frac{\Omega_1^2}{\delta^2+\gamma_1^2+\Omega_1^2},$$

$$f_1^1 = -\frac{\mathcal{F}_0^1}{\gamma_1}\frac{\Omega_1}{\sqrt{2}}\frac{\delta+i\gamma_1}{\delta^2+\gamma_1^2+\Omega_1^2}e^{i\omega t}. \tag{23}$$

The quantities f_0^0 and f_q^2 are given as before by the expressions (21).

With the indicated type of observation the only contributions to the intensity of the scattered light are those from f_0^0, $f_{\pm 1}^1$, f_0^2, and $f_{\pm 2}^2$. Then the intensity is

$$I = I_0\frac{F_0}{\nu_0}(A+B+C+D),$$

where

$$A = \frac{1}{3}\begin{Bmatrix}1 & 1 & 0\\ j_1 & j_1 & j_0\end{Bmatrix}^2 - \frac{1}{2}\begin{Bmatrix}1 & 1 & 2\\ j_1 & j_1 & j_0\end{Bmatrix}^2\frac{\gamma_0}{\gamma_2}$$

$$B = \frac{5}{4}\begin{Bmatrix}1 & 1 & 2\\ j_1 & j_1 & j_0\end{Bmatrix}^2\frac{\gamma_0}{\gamma_2}\frac{\Omega_1^2(4\delta^2+\Omega_1^2+\gamma_2^2)}{(\delta^2+\gamma_2^2+\Omega_1^2)(4\delta^2+\gamma_2^2+4\Omega_1^2)},$$

$$C = \frac{1}{2}\begin{Bmatrix}1 & 1 & 1\\ j_1 & j_1 & j_0\end{Bmatrix}^2\frac{\gamma_0}{\gamma_1}\Omega_1\left[\frac{\gamma_1}{\delta^2+\gamma_1^2+\Omega_1^2}\sin\omega t - \frac{\delta}{\delta^2+\gamma_1^2+\Omega_1^2}\cos\omega t\right],$$

$$D = \frac{1}{4}\begin{Bmatrix}1 & 1 & 2\\ j_1 & j_1 & j_0\end{Bmatrix}^2 \times\frac{\gamma_0}{\gamma_2}\Omega_1^2\frac{3\delta\gamma_2\sin 2\omega t-(2\delta^2-\gamma_2^2-\Omega_1^2)\cos 2\omega t}{(\delta^2+\gamma_2^2+\Omega_1^2)(4\delta^2+\gamma_2^2+4\Omega_1^2)} \tag{24}$$

Here A is a constant background, and B and D are the unmodulated and modulated (at frequency 2ω) resonance components with resonance width γ_2 that ordinarily appear in double resonance. The resonance component C is peculiar to circular polarization; it is modulated with the frequency ω of the radiofrequency magnetic field. When unsaturated the line is of the Lorentz shape with width γ_1. With increase of j_1 and j_0 the intensity of component C decreases more slowly than those of components B and D.

6. Excitation with modulated light. In experi-

ments done by Aleksandrov[5] and also by Corney and Series[11] a gas was placed in a constant magnetic field and the exciting light was modulated. In this case Eq. (11) takes the form

$$df_q^\varkappa / dt = (iq\Omega - \gamma_\varkappa) f_q^\varkappa + \mathscr{F}_q^\varkappa (1 - \varepsilon \sin \omega t), \quad (25)$$

where ϵ is the depth of modulation. It can be seen that in this case there is separation not only of equations with different κ, but also of those with different q. Therefore there are contributions to the scattered light only from the κ and q values which are generated by the exciting light. The solution of (25) for $t \gg \gamma_K^{-1}$ is

$$f_q^\varkappa = \mathscr{F}_q^\varkappa \left\{ \frac{\gamma_\varkappa + iq\Omega}{\gamma_\varkappa^2 + q^2\Omega^2} - \varepsilon \frac{[(\gamma_\varkappa + iq\Omega)^2 + \omega^2][(\gamma_\varkappa - iq\Omega)\sin\omega t - \omega\cos\omega t]}{[\gamma_\varkappa^2 + (\omega - q\Omega)^2][\gamma_\varkappa^2 + (\omega + q\Omega)^2]} \right\}. \quad (26)$$

From this we have

$$f_0^\varkappa = \mathscr{F}_0^\varkappa \left\{ \frac{1}{\gamma_\varkappa} - \varepsilon \frac{\gamma_\varkappa \sin\omega t - \omega\cos\omega t}{\gamma_\varkappa^2 + \omega^2} \right\}. \quad (27)$$

The second term in (27) describes the demodulation of the corresponding component of the scattered light when ω becomes larger than γ_K.

For $q > 0$ we assume $\Omega \gg \gamma_K$ (strong magnetic field), and we shall consider a modulation frequency of the exciting light close to the resonance frequency. Then

$$f_q^\varkappa = \mathscr{F}_q^\varkappa \frac{\varepsilon}{2} \frac{\omega - q\Omega + i\gamma_\varkappa}{\gamma_\varkappa^2 + (\omega - q\Omega)^2} e^{i\omega t}. \quad (28)$$

If the excitation is produced by plane-polarized light propagated along the magnetic field, then only $\mathscr{F}_0^0$, $\mathscr{F}_0^2$, and $\mathscr{F}_{\pm 2}^2$ are excited. Accordingly resonance is observed at the frequency $\omega = 2\Omega$. Resonance at the frequency $\omega = \Omega$ can be observed in plane-polarized light if, for example, the excitation and observation are arranged at right angles with the magnetic field and with each other. If with these same directions of excitation and observation one uses circular polarization, there are contributions to the scattered light from f_0^0, $f_{\pm 1}^1$, f_0^2 and $f_{\pm 2}^2$. In the components of the scattered light associated with $f_{\pm 1}^1$ the line width will be determined by the quantity γ_1, and the resonance will be reached at the frequency $\omega = \Omega$. The component with its resonance maximum at the frequency $\omega = 2\Omega$ (associated with $f_{\pm 2}^2$) will have a resonance width given by γ_2. Formulas for the intensity of the scattered light can be derived easily for the various cases, in analogy with the way this is done in Sec. 5, by means of Eqs. (12)—(19), (27), and (28).

7. Parametric resonance. Let the high-frequency magnetic field $\mathscr{H}_1$ be applied parallel to the constant magnetic field. The intensity of the exciting light is constant. Then (11) takes the form

$$df_q^\varkappa / dt = iq(\Omega + \Omega_1 \sin\omega t) f_q^\varkappa - \gamma_\varkappa f_q^\varkappa + \mathscr{F}_q^\varkappa. \quad (29)$$

As in the preceding case, equations with different κ and with different q are separated. The solution of (29) can be obtained in the form of an infinite series of terms modulated with frequencies which are multiples of ω.[3] For simplicity we shall regard the field $\mathscr{H}_1$ as weak and confine ourselves to the first approximation with respect to Ω_1/ω. Then it is not hard to get the solution ($t \gg \gamma_K^{-1}$)

$$f_q^\varkappa = \mathscr{F}_q^\varkappa \left\{ \frac{\gamma_\varkappa + iq\Omega}{\gamma_\varkappa^2 + q^2\Omega^2} + \frac{iq\Omega_1}{\gamma_\varkappa - iq\Omega} \times \frac{[(\gamma_\varkappa + iq\Omega)^2 + \omega^2][(\gamma_\varkappa - iq\Omega)\sin\omega t - \omega\cos\omega t]}{[\gamma_\varkappa^2 + (\omega - q\Omega)^2][\gamma_\varkappa^2 + (\omega + q\Omega)^2]} \right\}$$

from which we have $f_0^K = \mathscr{F}_0^K/\gamma_K$. We shall again take the constant magnetic field to be strong, and the frequency ω as close to resonance. Then the quantities f_q^K for $q > 0$ are given by (28) with the substitution $\epsilon \to \Omega_1/\omega$. The arguments given in Sec. 6 hold also in this case.

8. Finally, we note the possibility of observing the effects which have been described in unpolarized light. In this case in Eqs. (12) and (14) we must replace the functions Φ_q^K with the expression

$$\Psi_q^\varkappa(\mathbf{k}) = \sum_\lambda \Phi_q^\varkappa(\mathbf{e}_{\mathbf{k}\lambda}) = \sum_{q_1 q_2} (-1)^{q_2} \begin{pmatrix} 1 & 1 & \varkappa \\ q_1 & -q_2 & -q \end{pmatrix} [\delta_{q_1 q_2} - k_{q_1}(k_{q_2})^*],$$

where $\mathbf{k}$ is the unit vector in the direction of excitation (or observation) with the polar angles (θ_2, φ_2). Simple calculations give

$$\Psi_0^0 = -2/\sqrt{3}, \quad \Psi_q^1 = 0, \quad \Psi_0^2 = -(3\cos^2\theta_2 - 1)/\sqrt{30},$$
$$\Psi_1^2 = -\sin\theta_2 \cos\theta_2 e^{i\varphi_2}/\sqrt{5}, \quad \Psi_2^2 = -\sin^2\theta_2 e^{2i\varphi_2}/2\sqrt{5}.$$

Thus, for example, in an experiment on double resonance we can direct the unpolarized exciting light along the magnetic field (the z axis) and register the total intensity of the light scattered in the direction of the x or y axis. We will then observe an unmodulated resonance component and a resonance component modulated with the frequency 2ω. In the experiment on parametric resonance and in experiments with modulated exciting light one can observe resonance effects in unpolarized light if, for example, the directions of excitation, of observation, and of the constant magnetic field are all mutually perpendicular. Of course for unpolarized light the widths of all resonance curves are determined by the quantity γ_2.

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[1] J. Brossel and F. Bitter, Phys. Rev. **86**, 308 (1952).

[2] J. N. Dodd and G. W. Series, Proc. Roy. Soc. **A263**, 353 (1961).

[3] Aleksandrov, Konstantinov, Perel', and Khodovoĭ, JETP **45**, 503 (1963), Soviet Phys. JETP **18**, 346 (1964).

[4] P. A. Franken, Phys. Rev. **121**, 508 (1961).

[5] E. B. Aleksandrov, Optika i spektroskopiya **14**, 436 (1963).

[6] J. P. Barrat, J. phys. radium **20**, 541, 633, 657 (1959).

[7] M. I. D'yakonov and V. I. Perel', JETP **47**, 1483 (1964) and **48**, 345 (1965), Soviet Phys. JETP **20**, 997 (1965) and **21**, in press (1965).

[8] O. B. Konstantinov and V. I. Perel', JETP **45**, 279 (1963), Soviet Phys. JETP **18**, 195 (1964).

[9] A. Edmonds, CERN 55-26, Geneva, 1955, Russ. Transl. in Collection: Deformatsiya atomnykh yader (The Deformation of Atomic Nuclei), IIL, 1958, page 305.

[10] U. Fano, Phys. Rev. **133**, B828 (1964).

[11] A. Corney and G. W. Series, Proc. Phys. Soc. **83**, 207, 213 (1964).

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