Resonance fluorescence in a strong monochromatic field

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The problem of resonant scattering of a monochromatic electromagnetic wave by an atom in the ground state is solved without assuming that the incident field is weak. The cross sections for coherent and incoherent scattering are obtained. To obtain these cross sections it is essential in principle to take into account the relaxation of the excited level even in the case of a very strong field. The Raman scattering produced upon relaxation of an excited state via an intermediate state is considered. The results are generalized to include the case of collisions that lead to renormalization of the transition widths.

The theory developed by Weisskopf for quantum fluorescence presupposes that the incident photons are scattered by the atom independently. As noted by Heitler [2] the resonant fluorescence is a single-photon coherent process (the emitted wave is coherent with the primary wave), if the atom is not externally perturbed during the lifetime $1/\gamma$ of the excited state. The collision of the atom with some particle during this time randomizes the phase of the radiating dipole, and this leads to the appearance of an incoherent part of the scattering, which has a finite line width. A similar situation arises also when the intensity of the scattered light is increased, i.e., when the density of the incident photons is increased. Scattering of one photon can no longer be regarded as independent of the remaining ones if the probability of the collision of the atom with other photons becomes noticeable after a time $1/\gamma$. It becomes necessary to consider two-photon collisions which lead to non-monochromaticity of the scattered radiation. An analysis of the two-photon collisions was carried out by Rubin and Sokolovskiĭ [3]. When the intensity of the incident field is increased, it is necessary to take into account the multiphoton process.

The characteristic parameter of the problem, the smallness of which makes it possible to regard the incident field as weak, can be obtained by stipulating that the number of protons colliding with the atom in a time $1/\gamma$ be much less than unity, i.e., $\sigma J/\gamma \ll 1$, where σ is the resonance-fluorescence cross section ($\sigma \sim \lambda^2$, λ is the radiation wavelength) and J is the photon-flux density. This condition is equivalent to

$$(dE/\hbar\gamma)^2 \ll 1,$$

where d is the dipole moment of the transition and E is the incident-wave electric-field intensity.

This paper is devoted to resonant scattering of light by atoms in the ground state, without restriction on the intensity of the incident field relative to the parameter $(dE/\hbar\gamma)^2$. [1)] Allowance for the relaxation of the excited level plays the principal role even in the case of high intensities. The cross section of the unshifted scattering in a strong field breaks up into two parts, coherent and incoherent, which are determined in this paper. The sum of these parts can be obtained also without taking into account the level relaxation constant [4].

1. COHERENT-SCATTERING CROSS SECTION

Assume that an electromagnetic wave of frequency ω close to the frequency ω_{10} of the transition between the excited state 1 and the ground state 0 (Fig. 1) is incident on an immobile atom (molecule). The wave is

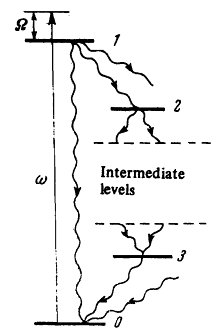

FIG. 1. Level scheme. The wavy lines correspond to the scattered photons.

linearly polarized, and the z axis is directed along the electric field

$$E(t) = Ee^{-i\omega t} + \text{c.c.}$$

The state 1 has a total angular momentum J = 1 and a z-projection of the momentum m = 0. The equations for the density matrix of the atom are

$$\begin{aligned} &\partial\rho_{11}/\partial t+\gamma\rho_{11}=Ge^{-i\omega t}\rho_{10}^*+G^*e^{i\omega t}\rho_{10},\\ &\partial\rho_{00}/\partial t=-(Ge^{-i\omega t}\rho_{10}^*+G^*e^{i\omega t}\rho_{10})+\gamma\rho_{11},\\ &\partial\rho_{10}/\partial t+(i\omega_{10}+\Gamma)\rho_{10}=Ge^{-i\omega t}(\rho_{00}-\rho_{11}). \end{aligned} \quad (1)$$

Here γ is the reciprocal lifetime of the level 1; $\Gamma = \gamma/2$ [2)]; $G = idE/\hbar$; $d = \langle 0 | d_z | 1\rangle$ is the matrix element of the projection of the dipole-moment operator on the z axis. The term $\gamma\rho_{11}$ in the second equation takes into account the arrival of the atom at the ground state. The presence of this term does not make it possible to use the Schrödinger equation to solve the problem. If we are interested only in the stationary solution (1), then, taking into account the normalization $\rho_{11} + \rho_{00} = 1$, we can easily obtain

$$\rho_{11}=\frac{\varkappa}{2}\frac{\Gamma^2}{\Omega^2+\Gamma_0^2},\quad \rho_{00}=1-\rho_{11},\quad \rho_{00}-\rho_{11}=\frac{\Omega^2+\Gamma^2}{\Omega^2+\Gamma_0^2}, \quad (2)$$

$$\rho_{10}=\frac{\Gamma+i\Omega}{\Omega^2+\Gamma_0^2}Ge^{-i\omega t}, \quad (3)$$

where

$$\varkappa=4|G|^2/\gamma\Gamma,\quad \Gamma_0^2=\Gamma^2(1+\varkappa),\quad \Omega=\omega-\omega_{10}.$$

The average value of the dipole moment of the atom is

$$\bar{d}_z=d\rho_{10}+\text{к.с.}\quad =\alpha e^{-i\omega t}E+\text{c.c.},$$

where

$$\alpha=\frac{|d|^2}{\hbar}\frac{\Omega+i\Gamma}{\Omega^2+\Gamma_0^2}.$$

The cross section of the unshifted coherent scattering is expressed in terms of the polarizability α in the following manner:

$$d\sigma=(\omega/c)^4|\alpha|^2\sin^2\theta do,$$

where $do=\sin\theta d\theta d\varphi$, while θ and φ are the polar and azimuthal angles in a spherical coordinate system with polar axis along z (see [6]). Consequently

$$d\sigma=\frac{d^4\omega^4}{c^4\hbar^2}\frac{\Omega^2+(\gamma/2)^2}{[\Omega^2+(\gamma/2)^2(1+\varkappa)]^2}\sin^2\theta\, do. \tag{4}$$

At $\kappa \ll 1$ we obtain the known resonance-fluorescence cross section [1,6]:

$$d\sigma=\frac{d^4\omega^4}{c^4\hbar^2}\frac{1}{\Omega^2+(\gamma/2)^2}\sin^2\theta\, do.$$

for $\kappa \gg 1$ we have

$$d\sigma=\frac{d^4\omega^4}{c^4\hbar^2}\frac{\Omega^2}{(\Omega^2+2|G|^2)^2}\sin^2\theta\, do. \tag{5}$$

As already noted in the Introduction, the parameter κ is a characteristic of our problem. When $\kappa \ll 1$, the field is weak (the cross section does not depend on the field intensity). In the general case (4) the resonance-curve width, which is connected with the change of the frequency ω, is determined by the width of the level and by the field—an additional "broadening by the field" takes place. At the maximum, i.e., at $\Omega=0$, the cross section for the coherent scattering decreases by a factor $(1+\kappa)^{-2}$ in comparison with the weak field. If we separate the total cross section of the coherent scattering σ_c, then (4) can be rewritten in the form

$$d\sigma=\sigma_c\frac{3}{8\pi}\sin^2\theta\, do,$$
$$\sigma_c=\frac{8\pi}{3}\frac{d^4\omega^4}{c^4\hbar^2}\frac{\Omega^2+\Gamma^2}{[\Omega^2+\Gamma^2(1+\varkappa)]^2}=\lambda^2\frac{3}{2\pi}\left(\frac{\gamma_{10}}{2}\right)^2\frac{\Omega^2+\Gamma^2}{[\Omega^2+\Gamma^2(1+\varkappa)]^2}, \tag{6}$$

where $\gamma_{10}=4d^2\omega^3/3c^3\hbar$ is the probability of the radiated transition $1\to 0$, with $\gamma_0\le\gamma$, since the transition can proceed also via intermediate levels (Fig. 1).

2. INCOHERENT PART OF THE SCATTERING

The cross section for the incoherent scattering can be obtained by jointly considering the system comprising the atom and the scattered photons. The Hamiltonian of the system is expressed in the form

$$H=\hbar\omega_{10}\sigma_3/2+\hbar\sum_k\omega_k c_k^+c_k-i\hbar\sum_k q(c_k\sigma_+-c_k^+\sigma_-)+i\hbar(Ge^{-i\omega t}\sigma_+-G^*e^{i\omega t}\sigma_-). \tag{7}$$

Here c_k^+ and c_k are the operators for the production and annihilation of a photon with wave vector $\mathbf{k}$, polarized linearly in the plane of the vectors E and k; $\omega_k=c|\mathbf{k}|$; $q=(2\omega/\hbar V)^{1/2}d\sin\theta$; θ is the angle between the z axis and k; V is the volume:

$$\sigma_3=\begin{pmatrix}1&0\\0&-1\end{pmatrix},\quad \sigma_+=\begin{pmatrix}0&1\\0&0\end{pmatrix},\quad \sigma_-=\begin{pmatrix}0&0\\1&0\end{pmatrix};$$

The eigenfunctions of the states 0 and 1 are

$$|0\rangle=\begin{pmatrix}0\\1\end{pmatrix},\quad |1\rangle=\begin{pmatrix}1\\0\end{pmatrix}.$$

We are interested in the power scattered by the atom,

$$P=-\mathrm{Sp}\,\{\rho j_z\hat{E}_z\},$$

where

$$j_z=-i\omega_{10}d(\sigma_--\sigma_+),\qquad \hat{E}_z=i\sum_k q(c_k-c_k^+),$$

and ρ is the density matrix. The expression for the power can be rewritten in the form

$$P=2\omega_{10}\sum_k \mathrm{Re}\{q\rho_{1,0k}\},\qquad \rho_{1,0k}=\langle|\langle 1|\rho|0\rangle|k\rangle. \tag{8}$$

Writing down the equation $i\hbar d\rho/dt=[H\rho]$ in matrix form and omitting the matrix elements ρ containing states with two photons $\mathbf{k}$, we obtain in the stationary case [3]

$$\begin{gathered}\left(\frac{d}{dt}-i\omega_k+\gamma\right)\rho_{1,1k}=Ge^{-i\omega t}\rho_{0,1k}+G^*e^{i\omega t}\rho_{1,0k},\\ \left(\frac{d}{dt}-i\omega_k+\delta\right)\rho_{0,0k}=q\rho_{10}^*-(Ge^{-i\omega t}\rho_{0,1k}+G^*e^{i\omega t}\rho_{1,0k})+\gamma\rho_{1,1k},\\ \left(\frac{d}{dt}-i\omega_k+i\omega_{10}+\Gamma\right)\rho_{1,0k}=q\rho_{11}+Ge^{-i\omega t}(\rho_{0,0k}-\rho_{1,1k}),\\ \left(\frac{d}{dt}-i\omega_k-i\omega_{10}+\Gamma\right)\rho_{0,1k}=G^*e^{i\omega t}(\rho_{0,0k}-\rho_{1,1k}).\end{gathered} \tag{9}$$

Here $\rho_{0,0\mathbf{k}}=\langle|\langle 0|\rho|0\rangle\mathbf{k}\rangle$, etc.; ρ_{11} and ρ_{10} are given by formulas (2) and (3). The infinitesimally small damping δ was introduced to determine the manner of circling around the pole in the integration with respect to frequency in (8). Solving the foregoing equations with allowance for the fact that

$$\rho_{0,0k}\sim e^{i\omega t},\quad \rho_{1,1k}\sim e^{i\omega t},\quad \rho_{0,1k}\sim e^{2i\omega t},$$

and that $\rho_{1,0k}$ does not depend on the time, we obtain

$$\rho_{1,0k}=\pi q\rho_{11}\varphi(\nu), \tag{10}$$
$$\varphi(\nu)=\frac{1}{\pi}\left\{\frac{1}{[\Gamma-i(\nu+\Omega)]}-\frac{2|G|^2[\Gamma-i(\nu-\Omega)]}{(\gamma-i\nu)[\Gamma-i(\nu+\Omega)](\Omega^2+\beta^2)}+i\frac{\gamma}{2\Gamma}\frac{(\Gamma-i\Omega)[\Gamma-i(\nu-\Omega)]}{(\nu+i\delta)(\Omega^2+\beta^2)}\right\}, \tag{11}$$

where

$$\nu=\omega_k-\omega,\quad \beta^2=(\Gamma-i\nu)[\Gamma-i\nu+\Gamma\varkappa\gamma/(\gamma-i\nu)].$$

Substituting (10) in (8) and dividing by the incident-wave energy flux $J=E^2c/2\pi$, we obtain the scattering cross section in the frequency interval $d\nu$:

$$\sigma(\theta,\nu)\,do\,d\nu=\sigma I(\nu)\frac{3}{8\pi}\sin^2\theta\, do\, d\nu, \tag{12}$$

$$\sigma=\gamma_{10}\rho_{11}\frac{\hbar\omega}{J}=\lambda^2\frac{3}{2\pi}\frac{2\Gamma}{\gamma}\left(\frac{\gamma_{10}}{2}\right)^2\frac{1}{\Omega^2+\Gamma^2(1+\varkappa)}. \tag{13}$$

The function $I(\nu)=\mathrm{Re}\,\varphi(\nu)$ (normalized to unity) determines the spectral distribution of the scattering intensity; σ is the total scattering cross section. Expression (13) has a simple meaning. Under the influence of an external field, the atom is thrown on to level 1 (and radiates spontaneously $\gamma_{10}\rho_{11}$ photons per unit time). To obtain the cross section it is necessary to divide $\gamma_{10}\rho_{11}$ by the incident-photon flux density.

Naturally, formulas (11)–(13) contain the results of the preceding section. Equation (12) can be easily divided into coherent and incoherent parts by using the formula

$$\sigma(\theta,\nu)\,do\,d\nu=[\sigma_c\delta(\nu)+\sigma_{inc}I_{inc}(\nu)]\cdot\frac{3}{8\pi}\sin^2\theta\, do\, d\nu; \tag{14}$$

σ_c is given by (16), $\sigma_{inc}=\sigma-\sigma_c$ is the total incoherent-scattering cross section,

$$I_{inc}(\nu)=\frac{1}{\pi(1-\sigma_c/\sigma)}\mathrm{Re}\left\{\frac{1}{\Gamma-i(\nu+\Omega)}-\frac{2|G|^2[\Gamma-i(\nu-\Omega)]}{(\gamma-i\nu)[\Gamma-i(\nu+\Omega)](\Omega^2+\beta^2)}+i\frac{\gamma}{2\Gamma}\frac{(\Gamma-i\Omega)[\Gamma-i(\nu-\Omega)]}{\nu(\Omega^2+\beta^2)}\right\}. \tag{15}$$

Figures 2a and 2b show plots of the spectral distribution $I_{inc}(\nu)$ of the incoherent part of the scattering (normalized to unity). The ratio

$$\sigma_{inc}/\sigma_c=\varkappa/[1+(\Omega/\Gamma)^2]$$

shows that the fraction of the incoherent part increases with increasing intensity.

At $\kappa \ll 1$ we have

$$I_{\text{inc}}(\nu)=\frac{2\Gamma(\Omega^2+\Gamma^2)}{\pi[(\nu+\Omega)^2+\Gamma^2][(\nu-\Omega)^2+\Gamma^2]}, \tag{16}$$

which coincides with the result of Rubin and Soklovskiĭ[3]. Expression (15) is also simplified in the case of $\kappa \gg 1$:

$$I_{\text{inc}}(\nu)=\frac{1}{\pi}\frac{\Omega^2+2G^2}{\Omega^2+4G^2}\left\{\frac{2G^2}{\Omega^2+2G^2}\frac{\tilde\Gamma}{\nu^2+\tilde\Gamma^2}+\frac{1}{2}\frac{b}{(\nu-C)^2+b^2}+\frac{1}{2}\frac{b}{(\nu+C)^2+b^2}\right\},$$

where

$$C=\sqrt{\Omega^2+4G^2},\quad \tilde\Gamma=\gamma\frac{\Omega^2+2G^2}{\Omega^2+4G^2},\quad b=\gamma\frac{\Omega^2+6G^2}{2\Omega^2+8G^2} \tag{17}$$

3. RAMAN SCATTERING

As already noted in Sec. 1, the transition $1\to 0$ can proceed via intermediate levels, and be accompanied by emission of spectral lines of frequency less than ω. For example, a transition $1\to 2$ with emission of a frequency ω_{12} can occur; this corresponds to Raman scattering of a strong field of frequency ω (Fig. 1). The total cross section for Raman scattering in the transition $1\to 2$ can be written down, in analogy with (13) in the form

$$\sigma_{1\to 2}=\gamma_{12}\rho_{11}\hbar\omega/J, \tag{18}$$

where γ_{12} is the radiative probability of the transition $1\to 2$. To find the spectral distribution of the intensity in this transition we use the method of Sec. 2, the only difference being that the scattered photons have frequencies close to ω_{12} (the strong field is at resonant, as before, with the transition $1\to 0$). The radiated power is given by formula (8) with the substitutions

$$\omega_{10}\to\omega_{12},\quad \rho_{1,0\mathbf{k}}\to\rho_{1,2\mathbf{k}},\quad \rho_{1,2\mathbf{k}}=\langle 1\langle 1|\rho|2\rangle|\mathbf{k}\rangle,\quad |2\rangle=\begin{pmatrix}0\\1\end{pmatrix}.$$

To find $\rho_{1,2\mathbf{k}}$, we have the equations

$$(d/dt-i\Delta+\Gamma_{12})\rho_{1,2\mathbf{k}}=q\rho_{11}+Ge^{-i\omega t}\rho_{0,2\mathbf{k}},$$
$$(d/dt-i\Delta-i\omega_{10}+\Gamma_{20})\rho_{0,2\mathbf{k}}=q\rho_{10}^{*}-G^{*}e^{i\omega t}\rho_{1,2\mathbf{k}}.$$

Here $\Gamma_{12}=(\gamma+\gamma_2)/2$, $\Gamma_{20}=\gamma_2/2$, γ_2 is the reciprocal lifetime of the level 2, and $\Delta=\omega_{\mathbf{k}}-\omega_{12}$ is the emitted-phonon frequency reckoned from the transition frequency (the scattered-field frequency is reckoned in Secs. 1 and 2 from the frequency ω). Recognizing that $\rho_{0,2\mathbf{k}}\sim e^{i\omega t}$, we easily obtain

$$\rho_{1,2\mathbf{k}}=\pi q\rho_{11}\varphi(\Delta),$$
$$\varphi(\Delta)=\frac{1}{\pi}(-i\Delta+\Gamma+\Gamma_{20})\{(-i\Delta+\Gamma_{12})[-i(\Delta-\Omega)+\Gamma_{20}]+|G|^2\}^{-1}. \tag{19}$$

for the differential Raman-scattering cross section in the frequency interval $d\Delta$ we have in analogy with (12)

$$\sigma(\theta,\Delta)\,do\,d\Delta=\gamma_{12}\rho_{11}\frac{\hbar\omega}{J}I_{2\to 1}(\Delta)\cdot\frac{3}{8\pi}\sin^2\theta\,do\,d\Delta,$$

where the function $I_{2\to 1}(\Delta)=\operatorname{Re}\varphi(\Delta)$ is normalized to unity; this function is shown in Fig. 4. For weak fields, $|G|^2\ll(\gamma_2+\gamma)\gamma_2/4$, we have for the spectral distribution of the Raman-scattering intensity

$$I_{2\to 1}(\Delta)=\frac{\gamma_2/2}{\pi[(\Delta-\Omega)^2+(\gamma_2/2)^2]}-\frac{[(\gamma_2/2)^2-(\Delta-\Omega)^2](\gamma+\gamma_2)/2-\Delta(\Delta-\Omega)\gamma_2}{\pi[\Delta^2+(\gamma+\gamma_2)^2/4][(\Delta-\Omega)^2+(\gamma_2/2)^2]^2}|G|^2. \tag{20}$$

In the case of the scattering of a field of higher intensity, $|G|^2\gg(\gamma+\gamma_2)\gamma_2/4$, we have

$$I_{2\to 1}(\Delta)=\frac{1}{\pi(\nu_++\nu_-)}\left[\nu_+\frac{b_+}{(\Delta-\nu_+)^2+b_+^2}+\nu_-\frac{b_-}{(\Delta+\nu_-)^2+b_-^2}\right],$$
$$\nu_\pm=\frac{1}{2}(\sqrt{\Omega^2+4|G|^2}\pm\Omega),\quad b_\pm=\frac{\Gamma}{2}\left(1+\frac{\gamma_2}{\Gamma}\mp\frac{\Omega}{\sqrt{\Omega^2+4|G|^2}}\right). \tag{21}$$

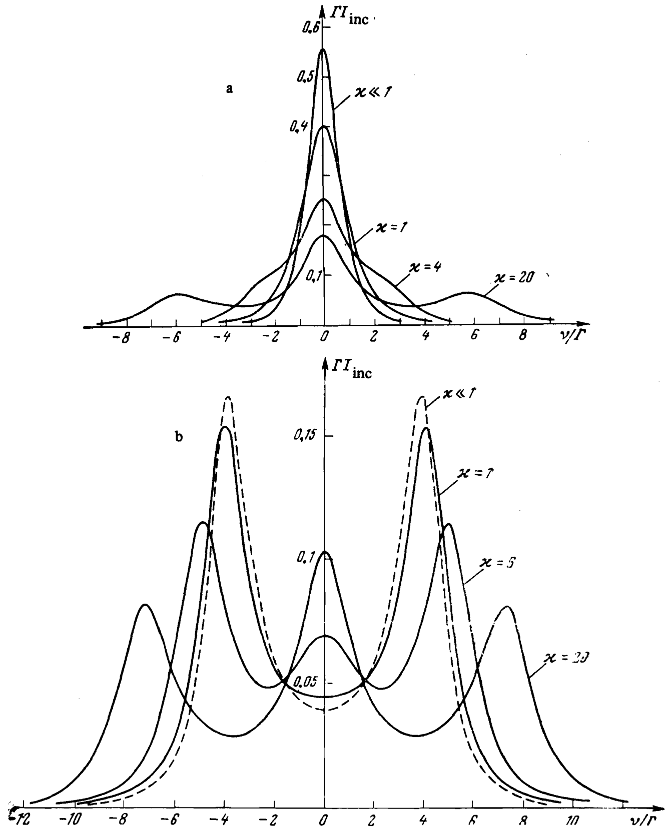

FIG. 2. Spectral distribution of incoherent scattering in the transition $1\to 0$: a) $\Omega=0$, b) $\Omega=4$. At $\kappa\geqslant 50$ it is possible to use in practice the asymptotic formula (17).

The scattered field consists in this case of two lines with frequency difference $\nu_++\nu_-$.

On going over to the ground state 0 from some intermediate level, say 3, the spectral distribution of the intensity in this transition depends on the incident strong field. The total scattering cross section is

$$\sigma_{(3\to 0)}=\gamma_{30}\rho_{33}\hbar\omega/J, \tag{22}$$

where ρ_{33} is the population of the level and can be determined in principle if the Einstein coefficients are known for all the transitions that bring the atom to the ground state, and γ_{30} is the probability of the $3\to 0$ transition. The quantity $\gamma\rho_{3,0\mathbf{k}}=\langle 1\langle 3|\rho|0\rangle|\mathbf{k}\rangle$ we need is obtained from the equations

$$(d/dt-i\Delta+\Gamma_{30})\rho_{3,0\mathbf{k}}=q\rho_{33}-Ge^{-i\omega t}\rho_{3,1\mathbf{k}},$$
$$(d/dt-i\Delta-i\omega_{10}+\Gamma_{31})\rho_{3,1\mathbf{k}}=G^{*}e^{i\omega t}\rho_{3,0\mathbf{k}},$$

where $\Gamma_{30}=\gamma_3/2$, $\Gamma_{31}=(\gamma_3+\gamma)/2$, $\Delta=\omega_{\mathbf{k}}-\omega_{30}$. Recognizing that $\rho_{3,1\mathbf{k}}\sim e^{i\omega t}$, we have

$$\rho_{3,0\mathbf{k}}=\pi q\rho_{33}\varphi_{3\to 0}(\Delta),$$
$$\varphi_{3\to 0}(\Delta)=\frac{1}{\pi}[-i(\Delta-\Omega)+\Gamma_{31}]\{(-i\Delta+\Gamma_{30})[-i(\Delta-\Omega)+\Gamma_{31}]+|G|^2\}. \tag{23}$$

The spectral distribution of the intensity $I_{3\to 0}(\Delta)=\operatorname{Re}\varphi_{3\to 0}(\Delta)$ is given for the case of a weak field $|G|^2\ll(\gamma+\gamma_3)\gamma_3/4$ by

$$I_{3\to 0}(\Delta)=\frac{\gamma_3/2}{\pi[\Delta^2+(\gamma_3/2)^2]}-\frac{[(\gamma_3/2)^2-\Delta^2](\gamma+\gamma_3)/2-\Delta(\Delta-\Omega)\gamma_3}{\pi[\Delta^2+(\gamma_3/2)^2]^2[(\Delta-\Omega)^2+(\gamma_3+\gamma)^2/4]^2}|G|^2.$$

For a strong field, when $|G|^2\gg(\gamma+\gamma_3)\gamma_3/4$,

$$I_{3\to 0}(\Delta)=\frac{1}{\pi(\nu_++\nu_-)}\left[(\nu_+-\Omega)\frac{c_+}{(\Delta-\nu_+)^2+c_+^2}+(\nu_--\Omega)\frac{c_-}{(\Delta-\nu_-)^2+c_-^2}\right]$$

where

$$c_{\pm}=\frac{\Gamma}{2}\left(1+\frac{\gamma_3}{\Gamma}\pm\frac{\Omega}{\sqrt{\Omega^2+4G^2}}\right).$$

For transitions not connected with the levels 0 and 1, the spectral lines have, as usual, a Lorentz shape.

4. DISCUSSION

Our analysis makes it possible to separate two qualitatively different field-intensity regions. In the region $\kappa \ll 1$, the field is assumed to be weak and the expansion (13) with respect to this parameter corresponds to the single-photon, two-photon, etc., scattering processes.

At $\kappa \sim 1$ it is necessary to take into account processes of all orders, a meaningless procedure in practice. The strong-field region $\kappa \gg 1$, however, admits also of an interpretation wherein the problem of the interaction of a monochromatic field with a two-level system without allowance for the relaxation constants is solved exactly (see [7], problem following Sec. 40).

Let us discuss first the weak-field region. The resonance-fluorescence scattering cross section in the weak-field region is given by formulas (14) and (16). The spectral distribution of the incoherently scattered photons has two maxima at the frequencies $\omega \pm \Omega$, i.e., there are two broadened side components in addition to the coherently-scattered monochromatic component at the frequency ω.

An examination of the Feynman diagram for the indicated processes shows that the scattering of the two photons can be symbolically represented as shown in Fig. 3. The solid line corresponds to an absorbed external-field photon of frequency ω, and the dashed line to an emitted photon. Since the system is in the ground state before and after the scattering, the energy conservation law should be satisfied exactly: $2\omega = \omega_1' + \omega_2'$, but this admits of a possibility of a spectral distribution of ω_1' and ω_2'. One of the incident photons can be regarded as a perturbation that leads to the incoherent part of the scattering, a fact that agrees with the statements made in the Introduction.

In a strong field, $\kappa \gg 1$, the scattered field consists of the three lines (14) and (17). The result is conveniently interpreted as a consequence of the level splitting in a rapidly-oscillating field [7]. Assume that in the absence of the field the energies of the excited and ground states are respectively E_1 and E_0. If the atom is acted upon by a monochromatic field, then each of the levels is split into two:

$$E_1^{\pm}=E_1+\Omega/2+\sqrt{(\Omega/2)^2+|G|^2},\quad E_0^{\pm}=E_0-\Omega/2\mp\sqrt{(\Omega/2)^2+|G|^2}.$$

This splitting makes the possible frequencies of the transitions equal to ω_{10}, $\omega_{10} \pm \sqrt{\Omega^2 + 4|G|^2}$ in accordance with (17).

A principal factor in our problem is the allowance

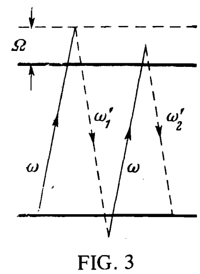

FIG. 3

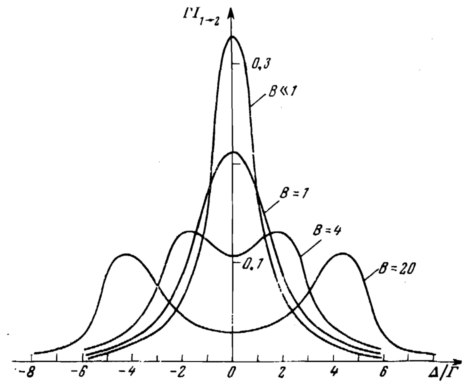

FIG. 4. Spectral distribution of Raman scattering on the $1 \to 2$ transition: $\Omega = 0$, $\Gamma = \gamma_2/2 = \gamma/2$, $B = |G|^2/\Gamma^2$.

for the relaxation of the level 1. This makes it possible to find the widths and intensities of the scattered lines at the indicated frequencies, and, most importantly, separate the coherent and incoherent parts of the scattering at the incident-field frequency. The unshifted scattering cross section σ_{sc} at $\kappa \gg 1$ is the sum of σ_c (6) and of that part of the total cross section which precedes $\tilde{\Gamma}/\pi(\nu^2+\tilde{\Gamma}^2)$ in (17):

$$\sigma_{sc}=\frac{d^4\omega^4}{c^4\hbar^2}\frac{1}{\Omega^2+4|G|^2}$$

σ_{sc} coincides with the regularly-scattering cross section obtained in [4] (formulas (23)) without taking into account the level relaxation constant. The possibility of experimentally separating the coherent incoherent parts depends on the measurement time. No matter how small the line width, the coherent part of the scattering can be separated within the measurement time $T \gg 1/\gamma$.

A warning must be sounded against a simplified solution of the strong-field scattering problem. The problem of the two-level system in a strong field was solved in [7], i.e., the induced dipoles at the indicated three frequencies were actually obtained.

Investigating the formula for the dipole radiation, it is easy to obtain the necessary cross sections. Such an analysis is valid only for a time $T \ll 1/\gamma$, during which the phase difference φ between the incident field and the induced dipole is preserved. Owing to the relaxation, the phase of the dipole becomes a random function with a correlation time on the order of $1/\gamma$, and this leads to fluctuation of the scattered-field intensity, since the absorbed power is proportional to $\cos^2\varphi$.

The first term of (20) determines the spectral intensity of the weak-field Raman scattering. As should be the case, the width γ of the intermediate level does not enter the width of the Raman-scattering line [8].

The appearance of two lines in the strong-field Raman scattering (21) is the consequence of the splitting of the E_1 level. Since the E_2 level is not split, the radiation on the transition $1 \to 2$ occurs at the frequencies $E_1^{\pm} - E_2$:

$$\omega_{\pm}=\omega_{12}+{}^1/_2(\Omega\pm\sqrt{\Omega^2+4|G|^2}),$$

which agrees with (21).

5. INFLUENCE OF COLLISIONS

As indicated in the introduction, the collision between the radiating atom and extraneous particles (atoms in a gas, phonons or other excitations in a solid) leads to a coherent scattering component. We shall take into account the collisions in the simplest case when their influence reduces to renormalization of the radiative transition widths, i.e., to introduction of collision frequencies β_{ij} on the corresponding transitions:

$$\Gamma=\frac{\gamma}{2}+\beta_{10},\quad \Gamma_{20}=\frac{\gamma_2}{2}+\beta_{20},\quad \Gamma_{12}=\frac{\gamma+\gamma_2}{2}+\beta_{12},$$

$$\Gamma_{30}=\frac{\gamma_3}{2}+\beta_{30},\quad \Gamma_{31}=\frac{\gamma_3+\gamma}{2}+\beta_{31}.$$

For the resonant and Raman scattering formulas (10)–(15), (19), and (23) remain in force, provided that Γ, Γ_{20}, Γ_{12}, Γ_{30}, and Γ_{31} are suitably defined.

In a weak field, $\varkappa \ll 1$, in the case of resonance fluorescence we have from (11) for the spectral scattered-radiation intensity distribution

$$I(\omega_k)=\left(1-\frac{\beta_{10}}{\Gamma}\right)\delta(\omega_k-\omega)+\frac{\beta_{10}}{\pi\Gamma}\frac{\Gamma}{(\omega_k-\omega_{10})^2+\Gamma^2}. \quad (24)$$

The collisions lead to the appearance of spontaneous radiation, which is due to the appearance of incoherent pumping to the level 1. The fraction of the coherent part of the scattering decreases correspondingly. When $\gamma \ll \beta_{10}$, the light is incoherently scattered.

For Raman scattering we obtain from (19) at $|G| \ll \Gamma, \Gamma_{20}, \Gamma_{12}$

$$I(\Delta)=\frac{\Gamma_{20}}{\pi[(\Delta-\Omega)^2+\Gamma_{20}^2]}+\frac{\beta_{10}+\beta_{20}-\beta_{10}}{\pi\Gamma_{20}}\frac{\Gamma_{12}\Gamma_{20}^2-\Delta(\Delta-\Omega)\Gamma_{20}}{(\Delta^2+\Gamma_{12}^2)[(\Delta-\Omega)^2+\Gamma_{20}^2]}. \quad (25)$$

The first term describes Raman scattering with the line broadened by collisions. The second term contains the resonance denominators corresponding both to Raman scattering and to spontaneous emission on the $1\to 2$ transition. The appearance of a spontaneous-decay channel is due to the appearance of incoherent pumping. The non-Lorentz line shape is due to interference between the spontaneous and Raman excitation channel [4)]. In the case of a detuning $\Omega \gg \Gamma_{11}, \Gamma_{20}$ the second term in (25) decreases in proportion to Ω^{-2}, i.e., we have the usual case of Raman scattering.

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[1)]When both levels have finite lifetimes, the problem must be differently formulated–it is necessary to take into account the excitation of the atom on the upper and lower levels. Rautian [5] considered in this formulation the spontaneous emission of an atom in the presence of a strong field.

[2)]We have introduced the transition width Γ to simplify the generalization of the results to include the case of collisions (see Sec. 5). For the same reason, we retain in formulas (2), (11), (13), and (15) the factor $2\Gamma/\gamma$, which is equal to unity.

[3)]The relaxation of level 1 is taken into account in (9) by introducing the constant γ, although it is more consistent to use only the Hamiltonian (7) without additional assumptions. It can be shown that the radiative lifetime remains unchanged in resonant interaction between the strong field and the atom. This enables us, in the derivation of (1) and (9), to take into account the level of relaxation as a complex increment to the energy ($E_1 \to E_1 - i\gamma/2$), and the arrival at the state 0 can be added phenomenologically by using the balance equation. Equations (9) can be obtained also directly from (7), but the required manipulations are quite cumbersome.

[4)]For the case when the state 0 is not the ground state, the influence of the interference of the indicated channel on the Raman-scattering cross section was analyzed in [8].

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