RAYLEIGH AND RAMAN SCATTERING IN THE FIELD OF AN INTENSE WAVE

M. L. TER-MIKAELYAN and A. O. MELIKYAN

Institute of Physical Research, Armenian Academy of Sciences

Submitted July 4, 1969

Zh. Eksp. Teor. Fiz. 58, 281–290 (January, 1970)

We consider the scattering of a light wave by an atom without assuming that the wave being scattered can be treated by perturbation theory. The frequency of the scattered light can be close to the frequency ω_{21} that corresponds to the energy difference of two levels of the atom. The relaxation of the system is neglected and it is assumed that the field strength of the wave being scattered is much smaller than characteristic values of the field strength in the atom. It is shown that the deviation from the usual perturbation-theory results is determined by a parameter α, Eq. (11). We first treat the scattering of light by a two-level system near resonance; we show that besides the spontaneous undisplaced scattering, Eq. (23), which includes a saturation effect, there is a Raman induced scattering at the frequency given by Eq. (24), with the cross section given by Eq. (30). Moreover, the two-level system in an intense field can have an absorption line at the frequency (25) with the cross section (31). These results are generalized to the case of a many-level system. The transition $1 \rightleftarrows 2$ is examined with intermediate levels and nonresonance terms taken into account. It is found that there is an additional coherent spontaneous scattering at twice the frequency with the cross section (38). Besides this there is noncoherent induced Raman scattering of two more frequencies, Eqs. (40) and (42), with the cross sections (41) and (43). The system will possess absorption at other new frequencies (45) and (42′), with the cross sections (46) and (47). The formulas correctly describe the passage to the limit of the perturbation-theory formulas.

LET us consider the scattering of a light wave whose frequency ω can be close to the frequency ω_{21},

$$\hbar\omega_{21} = E_2 - E_1, \tag{1}$$

where E_2 and E_1 are the energies of the excited and ground states of the system. We shall describe an intense electromagnetic wave incident on the atom by the classical vector potential

$$\mathbf{A}(\mathbf{r}, t) = a_{\mathbf{k}}\mathbf{e}\exp(i\mathbf{kr} - i\omega t) + \text{c.c.}, \tag{2}$$

In what follows we shall assume that the time intervals in which we are interested are so short that the relaxation of the system during the time of the process can be neglected. Actually this will mean that the characteristic frequency $\alpha\epsilon$ which appears in our exposition [see Eqs. (10) and (11)] is much larger than the widths of the levels. It is obvious that the spectral width of the radiation (2) being scattered must be much smaller than the characteristic frequencies. In this case we can regard the wave as a monochromatic wave propagated in the direction of $\mathbf{k} = \omega\mathbf{n}/c$.

We begin our treatment with the resonance scattering, when the lack of matching of the frequencies is much smaller than ω:

$$\varepsilon = \omega_{21} - \omega \ll \omega. \tag{3}$$

We ignore for the time being the interaction of the wave with electrons that are in energy levels not involved in resonance transitions. In this case our problem reduces to the scattering of light by a two-level system. In the first nonvanishing approximation of perturbation theory (with respect to both the incident and the scattered waves) the scattering of the light by the unperturbed state of the two-level atom consists only of (undisplaced) Rayleigh scattering. This is easily verified if one applies the formula derived by Kramers and Heisenberg[1] to the two-level system. In this case, assuming that the mean dipole moments vanish, $d_{11} = d_{22} = 0$, and that the energy levels are nondegenerate, the formula can be rewritten in the following form:

$$d\sigma_{\text{K.H.}} = \left| \frac{(\mathbf{d}_{12}\mathbf{e}'^*)(\mathbf{d}_{21}\mathbf{e})}{\omega_{21} - \omega} + \frac{(\mathbf{d}_{12}\mathbf{e})(\mathbf{d}_{21}\mathbf{e}'^*)}{\omega_{21} + \omega} \right|^2 \frac{\omega^4 d\sigma'}{\hbar^2 c^4} \tag{4}$$

where $\mathbf{d}_{12}$ and $\mathbf{d}_{21}$ are the dipole matrix elements [see Eqs. (8), (9)], and $\mathbf{e}$ and $\mathbf{e}'$ are the polarization vectors of the incident photon and a photon scattered into the solid angle $d\sigma'$. Near resonance the main contribution to the scattering is that of the first term. To avoid a divergence at exact resonance, we can follow Weisskopf[2] and replace E_2 by $E_2 - i\Gamma_2/2$ in the denominator in Eq. (4), in order to take into account the finite lifetime in the upper level.

Let us now consider the analogous problem without use of perturbation theory for the interaction of the intense monochromatic wave described by the potential (2) with the two-level system. The interaction between the scattered wave and the atom will be dealt with by perturbation theory as before. The wave function of the two-level atom can be put in the form

$$\Phi = b_1\psi_1(r)e^{-iE_1t/\hbar} + b_2\psi_2(r)e^{-iE_2t/\hbar}. \tag{5}$$

The coefficients b_1 and b_2 are to be determined from the Schrödinger equation

$$i\hbar\frac{\partial\Phi}{\partial t} = \left(H_0 - \frac{e}{mc}\mathbf{Ap} + \frac{e^2}{2mc^2}A^2\right)\Phi. \tag{6}$$

The action of the field will be expressed in the formation of bound states of the type shown in (14).

Because it is so intense the field $\mathbf{A}(\mathbf{r}, t)$ can be treated classically (see below), but the scattered field must

be treated on the basis of the quantum theory of radiation.

The solution of the problem of the behavior of a two-level system in the field of a classical electromagnetic wave near resonance is well known.[3,4] In this case we have from Eqs. (5) and (6)

$$i\hbar\frac{\partial b_1}{\partial t}=-b_2V_{12}a_{\mathbf{k}}^*e^{-iet},\qquad i\hbar\frac{\partial b_2}{\partial t}=-b_1V_{21}a_{\mathbf{k}}e^{-iet} \tag{7}$$

where $V_{12}=V_{21}^*$ is the matrix element for the transition with emission of a photon,

$$V_{12}=\frac{e}{mc}\int\psi_1^*(\mathbf{e}^*\mathbf{p})\psi_2e^{-i\mathbf{kr}}d\mathbf{r}. \tag{8}$$

In the derivation of (7) we have dropped terms containing rapidly oscillating functions of the time (nonresonance terms). We shall take them into account later. In the dipole approximation we must make the replacement

$$V_{12}a_{\mathbf{k}}^*\to E_{\mathbf{k}}^*(\mathbf{e}^*\mathbf{d}_{12}). \tag{9}$$

in the formulas that follow. The equations (7) have solutions of the form $\exp(i\lambda_{1,2}t)$, where

$$\lambda_{1,2}=\tfrac{1}{2}\varepsilon(-1\pm\sqrt{1+\alpha^2}). \tag{10}$$

The characteristic parameter which appears in (10),

$$\alpha=\frac{2}{\varepsilon\hbar}|V_{12}a_{\mathbf{k}}|, \tag{11}$$

and which in the dipole approximation takes the value

$$\alpha=|E_{\mathbf{k}}(\mathbf{ed}_{12})|/\varepsilon\hbar, \tag{11'}$$

will determine the deviation from the well known results of perturbation theory. The expression $\mathbf{E_k}=i\omega\mathbf{a_k}/c$ denotes the electric field strength that causes the scattering of the electromagnetic wave.

As will be seen in what follows, for values of α that satisfy the condition

$$\alpha\ll1,\quad \lambda_1\approx\varepsilon\alpha^2/4,\quad \lambda_2\approx-\varepsilon, \tag{12}$$

we arrive at the formulas of perturbation theory. If the opposite inequality

$$\alpha\gg1,\quad \lambda_{1,2}\approx\varepsilon|\alpha|/2 \tag{13}$$

holds, the cross sections for scattering of the light by the two-level system take an essentially different form. We shall look for the solution of (7) under initial conditions that correspond to an infinitely slow turning on of the field at $t=-\infty$. The turning on of the interaction at $t=0$ has to be specially analyzed, since in the higher orders of perturbation theory the instantaneous turning on of an interaction leads to spurious effects (cf., e.g.,[5]). The desired orthonormalized system of wave functions of Eq. (7) is

$$\Phi_1=C_1\exp(i\lambda_1t-iE_1t/\hbar)(\psi_1+B_1\psi_2e^{-i\omega t}), \tag{14}$$

$$\Phi_2=C_2\exp(i\lambda_2t-iE_1t/\hbar)(\psi_1+B_2\psi_2e^{-i\omega t}), \tag{15}$$

where

$$B_1=\hbar\lambda_1/V_{12}a_{\mathbf{k}}^*,\quad B_2=\hbar\lambda_2/V_{12}a_{\mathbf{k}}^*, \tag{16}$$

and the coefficients C_1 and C_2 are determined up to a phase factor by the normalization condition

$$C_{1,2}=[1+|B_{1,2}|^2]^{-1/2}. \tag{17}$$

Light scattering in which the state Φ_1 (or Φ_2) is not changed will be coherent; that is, the photons scattered by different atoms can interfere with each other. When the condition (12) holds the functions (14) and (15) go over into the perturbation-theory functions used by Kramers and Heisenberg. When the strong field is turned off the function Φ_1 corresponds to the ground state ψ_1 of the atom, while Φ_2 goes over into the function ψ_2 that describes the excited state of the atom. Turning on the strong field causes an admixture of the state ψ_2 to appear in the function Φ_1, and of ψ_1 in the function Φ_2. The admixtures of "other states" are determined by the coefficients B_1 and B_2, and for large α the electron has equal probabilities for being in the upper and lower levels. This is the saturation effect or induced transparency of the medium. For small values of α the coefficients $B_{1,2}$ correspond to perturbation-theory calculations with the condition (3). At first sight the functions (14) and (15) look nonstationary, but after the electromagnetic field is quantized these functions describe stationary states of the closed system made up of the two-level atom and the quantized electromagnetic field with a single definite propagation vector $\mathbf{k}$ and polarization $\mathbf{e}$.

Let us now proceed to the calculation of the scattered radiation with the wave vector $\mathbf{k}'$ and the polarization $\mathbf{e}'=\mathbf{e_{k'}}$. For the calculations to be made with perturbation theory it is necessary that

$$\alpha'=\left|\frac{E_{k'}(\mathbf{e}'\mathbf{d})}{\hbar(\omega_{21}-\omega')}\right|\ll1. \tag{12'}$$

We shall assume that there may be quanta present in space with the wave vector $\mathbf{k}'$; consequently we shall be including both spontaneous and stimulated scattering in what follows. It follows from the foregoing arguments that the scattering problem reduces to the problem of the radiation from atomic electrons whose wave functions are given by (14) and (15). The system "atom + classical field + quantized field" $\mathbf{A}'(\mathbf{rt})$,

$$\mathbf{A}'(\mathbf{r}t)=\sqrt{\frac{2\pi\hbar c^2}{v}}\sum_{\mathbf{k}'\neq\mathbf{k}}\frac{1}{\sqrt{\omega_{\mathbf{k}'}}}[C_{\mathbf{k}'}\mathbf{e}_{\mathbf{k}'}e^{i[\mathbf{k}'\mathbf{r}-\omega't]}+\text{h.c.}], \tag{18}$$

is described by the function

$$\varphi=b_1(t)\Phi_1+b_2(t)\Phi_2, \tag{19}$$

where $b_1(t)$ and $b_2(t)$ describe the quantized electromagnetic field. At $t=-\infty$

$$\varphi(-\infty)=\Phi_1|0\ldots n_k\ldots0\rangle. \tag{20}$$

The choice of the initial condition in (20) means that we are considering the scattering of the intense electromagnetic wave by an atom which is initially in the state Φ_1.

The change of the wave function (19) under the influence of the perturbation

$$V'=-\frac{e}{mc}\mathbf{A}'\mathbf{p} \tag{21}$$

is found by perturbation theory. To calculate the emission or absorption it is necessary to calculate matrix elements of the following form:

$$S\sim\langle0\ldots n_k\pm1\ldots\left|\int\Phi_i^*\left(-\frac{e}{mc}\mathbf{A}'\mathbf{p}\right)\Phi_jdx^4\right|0\ldots n_k\ldots0\rangle. \tag{22}$$

We take $j=1$, which will correspond to the scattering of the light by an atom which is in the state Φ_1.

Let us first examine processes in which the state of the system "atom + intense classical field" does not change. Here it is necessary to take i = 1. Obviously the frequency ω' of the scattered light is exactly equal to the frequency ω. In this case the cross section for Rayleigh spontaneous scattering is of the form (hereafter we shall write the formulas in the dipole approximation)

$$d\sigma_0 = \frac{|(\mathbf{e}'^*\mathbf{d}_{12})(\mathbf{e}\mathbf{d}_{21})|^2}{\varepsilon^2(1+\alpha^2)} \frac{\omega^4 do'}{c^4\hbar^2}. \quad (23)$$

For large $\alpha \gg 1$ the characteristic effect of saturation appears in the cross section (23). The number of scattered photons ceases to depend on the density of the incident radiation. This effect is physically identical to the effect considered by Schwinger and Karplus in[4] and developed in other papers, for example in[6]. The cross sections for "stimulated scattering" will have the additional factor $(n_{\mathbf{k}'} + 1)$ for the emission of a quantum with wave vector $\mathbf{k}'$ and polarization $\mathbf{e}'$, and a factor $n_{\mathbf{k}'}$ for the cross section for absorption of this same quantum.

Consequently, at the frequency $\omega' = \omega$ and in directions different from the direction of $\mathbf{k}$ there appears only spontaneous scattering, since the processes of stimulated scattering with emission and with absorption cancel each other. But the process of spontaneous scattering is coherent.

Let us now proceed to the calculation of scattering in which the system "atom + intense field" makes a transition from the state j = 1 to the state i = 2 (Raman scattering). It is easy to see that the processes of Raman stimulated scattering with absorption and with emission of a quantum $\mathbf{k}'$, $\mathbf{e}'$ occur at different frequencies. Accordingly, in contrast with the case of coherent scattering, in Raman scattering there is stimulated appearance of an additional quantum $\mathbf{k}'$ whose frequency is given by

$$\omega' = \omega - (\lambda_1 - \lambda_2). \quad (24)$$

Along with this the scattering process can lead to the absorption from a beam of quanta $\mathbf{k}''$ of a quantum $\mathbf{k}''$ whose frequency is given by

$$\omega'' = \omega + (\lambda_1 - \lambda_2). \quad (25)$$

It must be pointed out that the characteristic frequency $\lambda_{1,2}$ of the atomic electron in the strong field must be much smaller than the atomic frequencies. Otherwise it is necessary to take into account the shifts of the levels and the changes of the functions ψ_1 and ψ_2 in the field of the intense wave. These questions, and also the associated problems of finding the wave functions and estimating the corresponding approximations, have been treated in a number of papers.[7-11] For small intensities of the radiation scattered by the atom [cf. the condition (12)] we have

$$\omega' \approx 2\omega - \omega_{21} - \varepsilon\alpha^2/2, \quad (26)$$

$$\omega'' \approx \omega_{21} + \varepsilon\alpha^2/2; \quad (27)$$

and for large intensities, i.e., when the condition (13) holds, we have

$$\omega' \approx \omega - \varepsilon|\alpha|, \quad (28)$$

$$\omega'' \approx \omega + \varepsilon|\alpha|. \quad (29)$$

The cross section for Raman scattering is

$$d\sigma_1 = \frac{|(\mathbf{e}'^*\mathbf{d}_{12})(\mathbf{e}\mathbf{d}_{21})|^2}{\varepsilon^2(1+\alpha^2)} \left|\frac{1-\sqrt{1+\alpha^2}}{1+\sqrt{1+\alpha^2}}\right| \left(\frac{8\pi^3c^2}{\omega'^2} I_{\mathbf{k}'\mathbf{e}} + 1\right) \frac{\omega\omega'^3}{c^4\hbar^2} do'. \quad (30)$$

The analogous quantity with stimulated absorption of a quantum $\mathbf{k}''$ is

$$d\sigma_2 = \frac{|(\mathbf{e}'^*\mathbf{d}_{12})(\mathbf{e}\mathbf{d}_{21})|^2}{\varepsilon^2(1+\alpha^2)} \left|\frac{1+\sqrt{1+\alpha^2}}{1-\sqrt{1+\alpha^2}}\right| \frac{8\pi^3\omega''^2}{c^2\hbar^2} I_{\mathbf{k}''\mathbf{e}''} do'. \quad (31)$$

The quantity $I_{\mathbf{ke}}$ is connected with the number of quanta in the volume v by the formula

$$n_{ke} = \frac{8\pi^3c^2}{\hbar\omega^2} I_{\mathbf{ke}}. \quad (32)$$

For large intensities or for exact resonance, when the condition (13) is satisfied, the saturation effect, determined by the coefficient $1/\alpha^2$, appears in the cross sections (30) and (31). Consequently, for $\alpha \gg 1$ the cross section for Raman scattering is inversely proportional to the intensity of the radiation causing the scattering. In the opposite case of small intensities [cf. the condition (12)], the cross section for Raman scattering, Eq. (30), is proportional to $\alpha^2/4$, and consequently the number of scattered Raman quanta is proportional to the square of the intensity of the radiation incident on the atom. This result was to be expected, since, as was noted above, the two-level system has no Raman scattering in first-order perturbation theory. In the cross section (30) all orders of perturbation theory with respect to the parameter α^2 are effectively summed up.

The first experimental data on three-photon processes of the type of Eq. (30) in gaseous media were published in[12,13]. These papers also made a detailed theoretical analysis of the experiment on the basis of the equations of quasi-classical theory, which take into account effects of accumulation in the propagation of light in the medium.

For $\alpha \to 0$ the cross section (31) increases without bound in proportion to $4/\alpha^2$. It is easy to explain this result by noting that the cross section (31) essentially characterizes the absorption of the two-level system in the field of the intense wave. Therefore, if we let the intensity of the wave go to zero, $\alpha \to 0$, the product of the cross section (31) and the flux density I_ω of quanta incident on the atom must give the usual expression for the probability of absorption in the perturbation-theory approximation. Accordingly, the cross section (31) characterizes the probability of absorption per unit time at the frequency (25).

All of the results given above are for the case of scattering of intense light by an atom which was "originally" in the lower level, i.e., for $a_{\mathbf{k}} \to 0$, $\Phi_1 \to \psi_1$. Analogous results can be derived for scattering of the light by an atom which was in the state Φ_2. Furthermore, as can be seen easily from (22), the use of the function Φ_2 instead of Φ_1 to determine the Rayleigh (unshifted) scattering does not change the final result, and the cross section for scattering by the state Φ_2 is again given by Eq. (23). As for the Raman scattering, the pattern is changed symmetrically, i.e., absorption is replaced by emission, and conversely.

Let us transfer these results to a many-level system. The radiation incident on the atom is in resonance with the transition $1 \rightleftarrows 2$.

We note again that in finding the complete system of wave functions of the two-level system we dropped the

"nonresonance" terms, and also did not take into account the possibility of the transition $1 \rightleftarrows 2$ through other intermediate levels. The corrections to the functions (14) and (15) from the nonresonance terms, which were omitted in (7), and also from transitions with other intermediate states, are easily calculated by using the standard technique of perturbation theory.

The result for Φ_1 is

$$\Phi_1 = C_1 e^{i\lambda_1 t - iE_1 t/\hbar}\{\psi_1(1 + a_1 e^{-2i\omega t}) + B_1\psi_2 e^{-i\omega t}(1 + a_2 e^{2i\omega t}) + \sum_{m\neq 1;2}\psi_m(a_3 e^{-i\omega t} + a_4 e^{i\omega t} + a_5 e^{-2i\omega t} + a_6)\}. \quad (33)$$

The coefficients a_i that appear in (33) are given by

$$a_1 = -\frac{E_k(\mathbf{d}_{12}\mathbf{e})}{E_k^*(\mathbf{d}_{12}\mathbf{e}^*)}\frac{\lambda_1}{(2\omega-\lambda_1)}, \quad a_4 = \frac{E_k(\mathbf{de}^*)_{m1}}{\hbar(\omega_{m1}+\lambda_1+\omega)} \quad (34)$$

$$a_2 = -\frac{[E_k^*(\mathbf{d}_{12}\mathbf{e}^*)]^2}{\hbar\lambda_1(\omega_{21}+\omega+\lambda_1)}, \quad a_5 = \frac{E_k}{E_k^*}\frac{(\mathbf{de})_{m1}}{(\mathbf{de})_{12}}\frac{\lambda_1}{(\omega_{m1}+\lambda-2\omega)}$$

$$a_3 = \frac{E_k(\mathbf{de})_{m1}}{\hbar(\omega_{m1}+\lambda_1+\omega)} \quad a_6 = \frac{(\mathbf{de}^*)_{m1}\lambda_1}{(\mathbf{de}^*)_{12}(\omega_{m1}+\lambda_1)}$$

In the dipole approximation the coefficients $B_{1,2}$ are given by

$$B_{1,2} = \hbar\lambda_{1,2}/E_k^*(\mathbf{d}_{12}\mathbf{e}^*). \quad (35)$$

The function Φ_2 with correction terms included is obtained from (33) and (34) if we replace C_1, λ_1, B_1, and a_i by C_2, λ_2, B_2, and a_i'. In our subsequent equations $a_1', a_2', \ldots, a_6'$ will denote the coefficients of the corresponding time factors in the function Φ_2.

By using Eqs. (33)–(35) and (22) it is easy to calculate the corrections to the Rayleigh scattering cross section (23) owing to nonresonance transitions and transitions through intermediate states. The Rayleigh scattering cross section for scattering at frequency $\omega' = \omega$ into the solid angle do' is of the form

$$d\sigma_3 = \frac{d\sigma_0}{|B_1(\mathbf{d}_{12}\mathbf{e}'^*)|^2}\Big| B_1(\mathbf{d}_{12}\mathbf{e}'^*) + B_1^*(\mathbf{d}_{21}\mathbf{e}'^*)(a_1 + a_2^*) + \sum_{m\neq 1;2}[a_3(\mathbf{d}_{1m}\mathbf{e}'^*) + a_4^*(\mathbf{d}_{m1}\mathbf{e}'^*) + a_5 B_1^*(\mathbf{d}_{2m}\mathbf{e}'^*) + a_6^* B_1(\mathbf{d}_{m2}\mathbf{e}'^*)]\Big|^2. \quad (36)$$

As can be seen from the formulas (34) (if there is no accidental two-photon resonance with one of the intermediate levels, $\omega_{m_1} = 2\omega$, and also no accidental resonance $\omega_{m_1} = \omega$) the coefficients a_i are of the orders of magnitude

$$a_1 \sim a_5 \sim a_6 \sim \lambda_1/\omega,\ a_3 \sim a_4 \sim \alpha\varepsilon/\omega,\ a_2 \sim \alpha^2\varepsilon^2/\omega\lambda_1. \quad (37)$$

Consequently, if the field causing the scattering is much smaller than the atomic fields and is in resonance with only one of the levels of the atom, there will indeed be little change owing to the coefficients a_i which we have calculated above. For small values of α it is necessary to keep the coefficients $a_3 \approx a_4$ and a_2, if we want to get the correct transition to the Kramers-Heisenberg formula for Raman scattering in the case when the condition (3) is not satisfied, i.e., when the scattering is not necessarily of the resonant type. In this case the coefficients a_1, a_5, and a_6 are smaller by a factor α than the coefficients B_1, a_2, a_3, and a_4. For $\alpha \gg 1$ all of the correction coefficients are of the order of $\alpha\epsilon/\omega \approx E_k/E_{at} \ll 1$, while $B \approx 1$.

In the perturbation-theory approximation with respect to the strong field, i.e., for $\alpha \ll 1$, the scattering cross section (36) agrees with the well known Kramers-Heisenberg formula for Rayleigh scattering.

Besides the scattering (36) without change of frequency, there is the possibility of scattering in which the frequency of the scattered photon is $\omega' = 2\omega$. The cross section for this process is given by the expression

$$d\sigma_4 = \frac{|\mathbf{e}_k\mathbf{d}_{12}|^2\,\omega\omega'\,\alpha^2\,do'}{4(1+\alpha^2)e^4\hbar^2\lambda_1^2}\Big|\sum_{m\neq 1;2} a_5(\mathbf{d}_{1m}\mathbf{e}'^*) + B_1 a_4^*(\mathbf{d}_{m2}\mathbf{e}'^*)\Big|^2. \quad (38)$$

As can be seen, $d\sigma_4$ is of the order of $(\alpha\epsilon/\omega)^2 d\sigma_0$. The scattering given by the cross sections (36) and (38) is coherent.

Let us proceed to the calculation of formulas which improve our results (30) and (31) for Raman scattering. In this case, because the phases of the wave functions Φ_1 and Φ_2 of different atoms are arbitrary, the scattering is incoherent. The results of the calculations are as follows.

There are three combination frequencies which can appear in the scattering. One of them is given by the expression (24); its cross section is of the following form:

$$d\sigma_5 = d\sigma_1\frac{1}{|B_1(\mathbf{d}_{12}\mathbf{e}'^*)|^2}\Big| B_1(\mathbf{d}_{12}\mathbf{e}'^*) + B_2^*(a_1 + a_2'^*)(\mathbf{d}_{21}\mathbf{e}'^*) + \sum_{m\neq 1,2}[a_3(\mathbf{d}_{1m}\mathbf{e}'^*) + a_4'^*(\mathbf{d}_{m1}\mathbf{e}'^*) + B_2^* a_5(\mathbf{d}_{2m}\mathbf{e}'^*) + B a_6'^*(\mathbf{d}_{m2}\mathbf{e}'^*)]\Big|^2 \quad (39)$$

Over the entire range of variation of α the corrections to the cross section $d\sigma_1$ are of the order of magnitude of

$$d\sigma_1\varepsilon/\omega\alpha \ll d\sigma_1.$$

Another combination frequency is given by

$$\omega' = 2\omega - (\lambda_1 - \lambda_2). \quad (40)$$

Its intensity is determined by the cross section

$$d\sigma_6 = d\sigma_1\frac{1}{|B_1(\mathbf{d}_{12}\mathbf{e}'^*)|^2}\Big|\sum_{m\neq 1;2} a_5(\mathbf{d}_{1m}\mathbf{e}'^*) + a_4'^* B_1(\mathbf{d}_{m2}\mathbf{e}'^*)\Big|^2. \quad (41)$$

Over the entire range of variation of α the cross section (41) is of the order of magnitude of $d\sigma_1(\epsilon\alpha/\omega)^2$.

Finally, scattering is possible at the combination frequency (for $\epsilon < 0$)

$$\omega' = \lambda_2 - \lambda_1 = -\varepsilon\sqrt{1+\alpha^2}. \quad (42)$$

The cross section for this process is given by the expression

$$d\sigma_7 = \frac{d\sigma_1}{|B(\mathbf{d}_{12}\mathbf{e}'^*)|^2}\Big|\sum_{m\neq 1;2} B_2^* a_3(\mathbf{d}_{2m}\mathbf{e}'^*) + a_3'^* B(\mathbf{d}_{m2}\mathbf{e}'^*) + a_6(\mathbf{d}_{1m}\mathbf{e}'^*) + a_6'^*(\mathbf{de}'^*)_{m1}\Big|^2. \quad (43)$$

It is easy to see that for $\alpha \ll 1$ the cross section (43) goes over into the usual Kramers-Heisenberg formula for the Stokes component. We readily note that for values of α that satisfy the condition $\alpha \ll \epsilon/\omega$ the cross section $d\sigma_7$ is large, $d\sigma_7 \gg d\sigma_1$. For $\alpha > \epsilon/\omega$ three-photon scattering becomes predominant. For $\alpha \gg 1$ all of the cross sections considered above show saturation.

Analogous results can be obtained for the cross sections for stimulated absorption of an atom which is in the field of an electromagnetic wave. For absorption at the frequency ω'' given by (25) we have

$$d\sigma_8 = \frac{d\sigma_2}{|B_2(\mathbf{d}_{21}\mathbf{e}'^*)|^2}\Big[B_2(\mathbf{d}_{21}\mathbf{e}'^*) + (a_1'^* + a_2)B(\mathbf{d}_{12}\mathbf{e}'^*) + \sum a_3'^*(\mathbf{d}_{m1}\mathbf{e}'^*) + a_4(\mathbf{d}_{1m}\mathbf{e}'^*) + a_5'^* B(\mathbf{d}_{m2}\mathbf{e}'^*) + a_6 B_2'^*(\mathbf{d}_{2m}\mathbf{e}'^*) \Big]^2. \quad (44)$$

The absorption at the frequency

$$\omega'' = 2\omega + (\lambda_1 - \lambda_2) \quad (45)$$

is determined by the cross section

$$d\sigma_9 = \frac{d\sigma_2}{|B_2(\mathbf{d}_{21}\mathbf{e}')|^2}\Big| \sum_{m\neq 1,2} a_5'^*(\mathbf{d}_{m1}\mathbf{e}^*) + a_4 B_2^*(\mathbf{d}_{2m}\mathbf{e}'^*) \Big|^2 \quad (46)$$

Finally, for $\epsilon > 0$ there is absorption at the frequency

$$\omega' = \lambda_1 - \lambda_2 = \varepsilon\sqrt{1+\alpha^2}. \quad (42')$$

In this case the absorption cross section is given by

$$d\sigma_{10} = \frac{d\sigma_2}{|B_2(\mathbf{d}_{21}\mathbf{e}')|^2}\Big| \sum_{m\neq 1,2} a_3 B_2^*(\mathbf{d}_{2m}\mathbf{e}'^*) + a_3'^* B_1(\mathbf{d}_{m2}\mathbf{e}'^* + a_6(\mathbf{d}_{1m}\mathbf{e}'^*) + a_6'^*(\mathbf{d}_{m1}\mathbf{e}'^*) \Big|^2. \quad (47)$$

An analysis of these formulas can be carried out in analogy with that given above, and therefore we omit it here.

The calculations we have given can be easily generalized to a many-level system for which the incident radiation is in resonance with one of the "intermediate" levels.

Effects of the spectral width of the incident radiation, and also of the widths of the levels of the system, will be considered in a separate paper.

[1] H. A. Kramers and W. Heisenberg, Z. Physik 31, 681 (1925).

[2] V. Weisskopf, Z. Physik 85, 451 (1933); Ann. Physik 9, 23 (1931).

[3] L. D. Landau and E. M. Lifshitz, Quantum Mechanics: Non-relativistic Theory, Addison-Wesley, Reading, Mass., 1958, 1965.

[4] R. Karplus and J. Schwinger, Phys. Rev. 73, 1020 (1948).

[5] L. Schiff, Quantum Mechanics, New York, McGraw-Hill Book Co., 1955, 1968.

[6] A. L. Mikaelyan, M. L. Ter-Mikaelyan, and Yu. Turkov, Opticheskie generatory na tverdom tele (Solid-State Lasers), Izd. Radio, 1967.

[7] L. V. Keldysh, Zh. Eksp. Teor. Fiz. 47, 1945 (1964) [Sov. Phys.-JETP 20, 1307 (1965)].

[8] V. Ritus, Zh. Eksp. Teor. Fiz. 51, 1544 (1966) [Sov. Phys.-JETP 24, 1041 (1967)].

[9] A. I. Nikishov and V. I. Ritus, Zh. Eksp. Teor. Fiz. 46, 776 (1964) [Sov. Phys.-JETP 19, 259 (1964)].

[10] L. P. Kotova and M. V. Terent'ev, Zh. Eksp. Teor. Fiz. 52, 732 (1967) [Sov. Phys.-JETP 25, 481 (1967)].

[11] V. M. Galitskiĭ, S. P. Goreslavskiĭ, and V. F. Elesin, Zh. Eksp. Teor. Fiz. 57, 207 (1969) [Sov. Phys.-JETP 30, 117 (1970)].

[12] N. N. Badalyan, V. A. Iradyan, and M. E. Movsesyan, ZhÉTF Pis. Red. 8, 518 (1968) [JETP Lett. 8, 518 (1968)].

[13] V. M. Arutyunyan, N. N. Badalyan, V. A. Iradyan, and M. E. Movsesyan, Zh. Eksp. Teor. Fiz. 58, 37 (1970) [this issue, p. 22].

Translated by W. H. Furry
35