

RESONANT RADIATIVE PROCESSES

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The frequency correlation properties of the radiation from an atom in a strong field in resonance with neighboring transitions are considered. It is shown that the difference in frequency correlation in two-photon and stepwise processes decreases with increase of the external field. The spectral compositions of the Doppler-broadened resonance scattering and fluorescence are analyzed. It is shown that in these cases the Doppler line width is anisotropic.

1. INTRODUCTION

Thermal motion of radiating atoms leads, owing to the Doppler effect, to an isotropic broadening of the luminescence line. At the same time, the line width of Rayleigh scattering depends on the direction^[1]

$$\Delta\omega = \omega \frac{\bar{v}}{c} \cdot 2 \sin \frac{\theta}{2}, \quad \bar{v} = \sqrt{2kT/m},$$

and vanishes in the case of forward scattering ($\theta = 0$). This difference in the manifestation of the Doppler effect is due entirely to the difference between the frequency-correlation properties of the indicated processes^[2]. On the other hand, the frequency-correlation properties are strongly pronounced also in other two-photon processes (two-quantum absorption and luminescence, or Raman scattering). It is therefore natural to expect the existence of anisotropy of the Doppler line width in this case, too. This question is discussed in Sec. 3.

We note now that an analysis of radiative processes within the framework of second order perturbation-theory leads to a delimitation of two-photon processes proper from stepwise (or cascade) processes, for example two-photon luminescence and cascade emission of two photons with a real intermediate state. Such a delimitation is based essentially on the frequency-correlation properties. On the other hand, if the energy of interaction of the atom with the field is larger than the level width, then the frequency-correlation properties of the radiative processes experience a strong metamorphosis (Sec. 2). In particular, two-photon and stepwise processes turn out to be physically indistinguishable. As a consequence, in strong fields the manifestation of the Doppler broadening also changes strongly. The resultant phenomena are traced for the resonant-scattering doublet and resonant-fluorescence triplet (Secs. 4 and 5).

2. FREQUENCY-CORRELATION PROPERTIES OF RADIATIVE PROCESSES

We shall consider the radiation of an atom situated in an external field with two monochromatic components of frequencies ω and ω_μ and amplitudes E and E_μ . We assume that each field component interacts only with one transition (resonance approximation). In the scheme corresponding to Raman scattering (Fig. 1), a photon

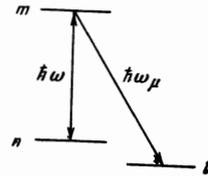


FIG. 1. Term scheme.

$\hbar\omega$ is absorbed and a photon $\hbar\omega_\mu$ is emitted. The probability amplitudes a_i of the states $i = m, n, l$ satisfy the system of equations

$$\begin{aligned} \dot{a}_m + \gamma_m a_m &= iG e^{-i\Omega t} a_n + iG_\mu e^{-i\Omega_\mu t} a_l, \\ \dot{a}_n + \gamma_n a_n &= iG^* e^{i\Omega t} a_m, \quad \dot{a}_l + \gamma_l a_l = iG_\mu^* e^{i\Omega_\mu t} a_m, \end{aligned} \quad (2.1)$$

$$\Omega = \omega - \omega_{mn}, \quad \Omega_\mu = \omega_\mu - \omega_{ml}, \quad G = d_{mn}E / 2\hbar, \quad G_\mu = d_{ml}E_\mu / 2\hbar,$$

where d_{ij} are the matrix elements of the dipole moment.

We shall henceforth regard $Ge^{-i\Omega t}$ as a strong perturbation and $G_\mu \exp(-i\Omega_\mu t)$ as a weak perturbation. We are interested in the probability w_μ of emission of the photon $\hbar\omega_\mu$. The solution of the system (2.1) can be obtained by successive approximations in the parameter G_μ ^[3-5]: we consider the system of equations

$$\dot{a}_m + \gamma_m a_m = iG e^{-i\Omega t} a_n, \quad \dot{a}_n + \gamma_n a_n = iG^* e^{i\Omega t} a_m, \quad (2.2)$$

in the zeroth approximation and introduce its exact solution into the right-hand side of the equation for $a_l(t)$ in (2.1). Integration of this equation yields the first approximation G_μ for $a_l(t)$, with the aid of which we calculate

$$w_\mu = 2\gamma_l \int_0^\infty |a_l(t)|^2 dt.$$

The solution of the system (2.2) can be represented in the form

$$a_n(t) = [A_1 e^{-\alpha_1 t} + A_2 e^{-\alpha_2 t}] e^{i\Omega t}, \quad a_m(t) = iG \left[\frac{A_1 e^{-\alpha_1 t}}{\gamma_m - \alpha_1} + \frac{A_2 e^{-\alpha_2 t}}{\gamma_m - \alpha_2} \right], \quad (2.3)$$

$$\alpha_{1,2} = \frac{\Gamma + i\Omega}{2} \pm i \sqrt{G^2 + \left(\frac{\Omega - i\gamma}{2}\right)^2}, \quad \Gamma = \gamma_m + \gamma_n, \quad \gamma = \gamma_n - \gamma_m, \quad (2.4)$$

where $A_{1,2}$ are the integration constants. In the case of interest to us, $a_n(0) = 1$, we have

$$A_{1,2} = \mp \frac{\gamma_m - \alpha_{1,2}}{\alpha_1 - \alpha_2}, \quad a_m(t) = \frac{iG}{\alpha_1 - \alpha_2} [e^{-\alpha_1 t} - e^{-\alpha_2 t}], \quad (2.5)$$

$$w_\mu = \frac{2|GG_\mu|^2}{|\alpha_1 - \alpha_2|^2} \text{Re} \left\{ \frac{(\alpha_1 + \alpha_1^*)^{-1} - (\alpha_2 + \alpha_1^*)^{-1}}{\gamma_l + \alpha_1^* + i\Omega_\mu} \right\}$$

$$\times \frac{(\alpha_2 + \alpha_2^*)^{-1} - (\alpha_1 + \alpha_2^*)^{-1}}{\gamma_l + \alpha_2^* + i\Omega_\mu} \}. \quad (2.6)$$

The frequency-correlation properties consist in the fact that the frequencies Ω_μ at which there is maximum probability of emitting the photon $\hbar\omega_\mu$ ($\Omega_{\mu 1} = \text{Im } \alpha_1$, $\Omega_{\mu 2} = \text{Im } \alpha_2$) turn out to depend on Ω , i.e., on the frequency of the absorbed photon. In the case of small G , we have in place of (2.4) and (2.6)

$$\alpha_1 = \gamma_m, \quad \alpha_2 = \gamma_n + i\Omega, \quad G \ll |\Omega - i\gamma|, \quad (2.7)$$

$$w_\mu = \frac{|GG_\mu|^2}{|\gamma_n - \gamma_m + i\Omega|^2}$$

$$\times \text{Re} \left\{ \frac{\gamma_m^{-1} - 2(\gamma_m + \gamma_n + i\Omega)^{-1}}{\gamma_l + \gamma_m + i\Omega_\mu} + \frac{\gamma_n^{-1} - 2(\gamma_m + \gamma_n - i\Omega)^{-1}}{\gamma_l + \gamma_n + i(\Omega_\mu - \Omega)} \right\}$$

The second term in the expression for w_μ reaches a maximum at a frequency $\omega_\mu = \omega - \omega_{ln}$ (Raman scattering). It can therefore be said that in the second stage of the two-process the atom "remembers" which quantum was absorbed during the first stage. The Raman-scattering line width $\gamma_l + \gamma_n$ also "remembers" from which level the atom arrived at the first stage. These indeed are the properties of frequency correlation. To the contrary, the first term in (2.7) gives resonance at $\Omega_\mu = \omega_\mu - \omega_{ml} = 0$, i.e., at the frequency of the transition between the intermediate and final states of the atom. The width of the corresponding line is also determined by the levels m and l . This term describes the cascade or stepwise transition $n \rightarrow m \rightarrow l$, and there is no correlation in it at all between the absorption and emission acts.

The correlation properties of the emission processes are closely connected with the type of evolution of the amplitude $a_m(t)$ of the intermediate state. Let $|\Omega| \gg \gamma_m, \gamma_n$; then the rapidly oscillating term $\exp[-(i\Omega + \gamma_n)t]$ (virtual state) carries information concerning the initial state (γ_n) and the absorbed quantum (Ω), and causes the appearance of a scattering line, i.e., the second term in (2.7) (the terms $2(\gamma_m + \gamma_n \pm i)^{-1}$ can be discarded). The time dependence of the second term, $\exp(-\gamma_m t)$, contains no attributes of the absorption act and does not differ from the case when the state m is the initial state (i.e., $a_m(0) = 1$). We can therefore say with respect to this term and with respect to the corresponding unshifted line at the transition $m \rightarrow l$ that the intermediate state is a real state of the atom, having a finite lifetime $(2\gamma_m)^{-1}$.

A separate examination of the transitions through the virtual and real states signifies that only squares of the moduli of the first and second terms remain in the expression for $|a_l(t)|^2$. The crossing term lead to the appearance of $2(\gamma_m + \gamma_n \pm i\Omega)^{-1}$ in w_μ , and it is legitimate to neglect them if $|\Omega| \gg \gamma_m, \gamma_n$. In the case when $|\Omega| \sim \gamma_m, \gamma_n$, the crossing terms, which reflect the interference between the real and virtual states, are significant and cannot be discarded. However, even here w_μ can be represented in the form of a sum of terms with and without "memory," and allowance for the interference only changes the coefficient preceding these terms.

The only physical basis for contrasting the stepwise and two-photon processes is the difference between

their frequency-correlation properties, which are uniquely connected with the singularities of the evolutions of the individual terms of the amplitude $a_m(t)$ of the intermediate state. On the other hand, formulas (2.7) are valid within the framework of second-order perturbation theory, and are no longer valid at sufficiently large G^2 . In the general case, a_m contains two exponential terms (formula (2.5)), which are formally analogous to the "virtual" and "real" states. However, both α_1 and α_2 depend on the parameters of the field (G^2, Ω) and of the two combining levels (γ_m, γ_n). With respect to w_μ , this means that in both resonances the atom "remembers" which quantum was observed during the first stage of the process. In the limiting case of a very strong field we have

$$\alpha_{1,2} \cong [\gamma_m + \gamma_n + i\Omega]/2 \pm iG, \quad G \gg |\Omega - i\gamma|, \quad (2.8)$$

i.e., the differences in the temporal properties of the two exponentials in (2.5) have disappeared completely. At the same time, the differences in the frequency-correlation properties of the corresponding lines have also disappeared, and there are no grounds for distinguishing between the two. The concepts of stepwise and two-photon transitions or of the virtual and real states are likewise physically indistinguishable. It is clear from the foregoing that these concepts are inseparably linked with perturbation theory and lose physical meaning outside the region of its applicability.

The external field levels out the differences between α_1 and α_2 both with respect to the yield of the resonance (Ω) and with respect to the difference in the damping of the states m, n . Let us consider in greater detail the case $\gamma_m = \gamma_n = \Gamma$, when the leveling function of the field simplifies:

$$\alpha_{1,2} = \Gamma + i\alpha_{1,2}'', \quad \alpha_{1,2}'' = 1/2[\Omega \pm \sqrt{\Omega^2 + 4G^2}]. \quad (2.9)$$

Figure 2 shows plots of $\alpha_{1,2}''$ as functions of Ω . The asymptotic approach of the plots to the abscissa axis and to the dashed line $\alpha'' = \Omega$ corresponds respectively to real and virtual states. As Ω changes from positive to negative values, we have for α_1'' , for example, a smooth transition from the properties of the virtual state to the properties of the real state, and for α_2'' the inverse sequence. In the region $|\Omega| < G$, the states α_1 and α_2 differ little.

As the measure of the "memory" of the absorbed quantum we can choose the quantity

$$M_{1,2} \equiv \frac{d\alpha_{1,2}''}{d\Omega} = \frac{1}{2} \left[1 \pm \frac{\Omega}{\sqrt{\Omega^2 + 4G^2}} \right], \quad (2.10)$$

which varies from 0 to 1. The value $M = 0$ means complete absence of memory (stepwise transition), while

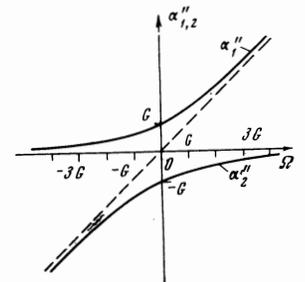


FIG. 2. Plot of $\alpha''_{1,2}$ against Ω .

$M = 1$ corresponds to total correlation between the frequencies of the absorbed and emitted photons (two-photon transition). Since $\alpha_1 + \alpha_2 = \Gamma + i\Omega$, it follows that $M_1 + M_2 = 1$; this fact can be interpreted as follows: the intermediate state as a whole, without subdivision into two states, retains the entire information on the absorbed photon. In the case $|\Omega| \ll G$, we have $M_{1,2} \approx 1/2$, i.e., the "memory" is equally divided between the two terms in $a_m(t)$.

If $\alpha_1' - \alpha_2'$ is sufficiently small, the interference of the states α_1 and α_2 is quite significant. A separate analysis of the transitions through these intermediate states is meaningful if the distances between the resonances exceed their widths by w_μ :

$$|\alpha_1' - \alpha_2'| \gg \alpha_1' + \alpha_2' + 2\gamma_l.$$

Under these conditions, the emission spectrum for the transition $m \rightarrow l$ has the form of a well resolved doublet, the appearance of which can be interpreted as the splitting of the intermediate-state level into two sublevels^[3-9]. In accordance with the foregoing, it is meaningless to attribute the components of this doublet to stepwise or two-photon transitions. One can only speak of the resonance-scattering doublet as a unit. We shall henceforth use this term.

For concreteness, we have referred throughout to processes of the type of Raman scattering. All the physical conclusions pertain also to other processes in which two-photons take part, such as two-quantum luminescence, two-quantum absorption, and Raman scattering via a lower intermediate level. It is only necessary to reverse the signs of Ω and Ω_μ in all the formulas, depending on whether the corresponding quantum is emitted or absorbed.

3. DOPPLER BROADENING OF RAMAN SCATTERING LINE

Allowance for the motion of the atom in the case of traveling waves reduces, as is well known, to the substitutions $\Omega \rightarrow \Omega - \mathbf{k} \cdot \mathbf{v}$ and $\Omega_\mu \rightarrow \Omega_\mu - \mathbf{k}_\mu \cdot \mathbf{v}$, where \mathbf{k} and \mathbf{k}_μ are the wave vectors of the waves. This means that the amplitudes A_1 and A_2 also depend on the velocity of the atom. Within the framework of the second approximation of perturbation theory, this pertains only to the virtual sublevel. This case apparently has not been discussed in the literature, and will be considered in the present section. Averaging over the velocities will be carried with a Maxwellian distribution:

$$\langle \pi \bar{v} \rangle^{-1/2} \exp\{-v^2/\bar{v}^2\}. \quad (3.1)$$

Let the deviation from resonance be larger not only than the natural width but also of the Doppler width ($|\Omega| \gg k\bar{v}$). Under this condition we can neglect the interference between the real and virtual states, and we can easily obtain from (2.7)

$$\langle w_\mu \rangle = -\frac{|GG_\mu|^2}{\Omega^2} \operatorname{Re} \left\{ \frac{\sqrt{\pi}}{\gamma_m k_\mu \bar{v}} e^{p_1} [1 - \Phi(p_1)] + \frac{\sqrt{\pi}}{\gamma_n q \bar{v}} e^{p_2} [1 + \Phi(p_2)] \right\},$$

$$p_1 = [\gamma_l + \gamma_m + i\Omega_\mu] / k_\mu \bar{v}, \quad p_2 = [\gamma_l + \gamma_n + i(\Omega_\mu - \Omega)] / q \bar{v},$$

$$q = |\mathbf{k}_\mu - \mathbf{k}| = \sqrt{(k_\mu - k)^2 + 4k_\mu k \sin^2(\theta/2)}, \quad (3.2)$$

where $\Phi(z)$ is the probability integral and θ is the angle between \mathbf{k} and \mathbf{k}_μ . If the Doppler broadening dominates over the natural broadening, $k_\mu \bar{v} \gg \gamma_l + \gamma_m$, $q \bar{v} \gg \gamma_l$

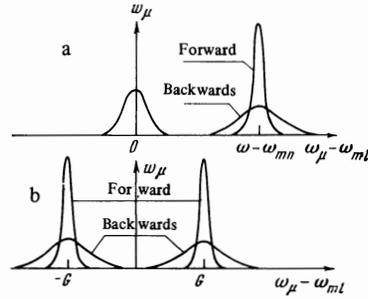


FIG. 3

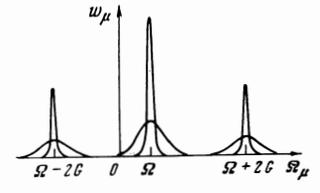


FIG. 4

FIG. 3. Spectrum of Raman scattering and stepwise transition (a) and spectrum of Doppler-broadened resonant scattering (b).

FIG. 4. Resonance fluorescence triplet.

+ γ_n , then we can assume that $\Phi(p_{1,2}) = 0$ and (3.2) contains two terms of Gaussian form

$$\langle w_\mu \rangle = \frac{|GG_\mu|^2}{\Omega^2} \left\{ \frac{\sqrt{\pi}}{\gamma_m k_\mu \bar{v}} \exp\left[-\frac{\Omega_\mu^2}{(k_\mu \bar{v})^2}\right] + \frac{\sqrt{\pi}}{\gamma_n q \bar{v}} \exp\left[-\frac{(\Omega_\mu - \Omega)^2}{(q \bar{v})^2}\right] \right\}. \quad (3.3)$$

The first terms in (3.2) or (3.3) (stepwise-transition line) has a Doppler width $k_\mu \bar{v}$, which does not depend on θ . On the other hand, the Doppler width $q \bar{v}$ of the Raman-scattering line depends strongly on the observation direction, changing from $|\mathbf{k}_\mu - \mathbf{k}| \bar{v}$ to $(\mathbf{k}_\mu + \mathbf{k}) \bar{v}$ when θ changes from zero to π (Fig. 3a). If $|\mathbf{k}_\mu - \mathbf{k}| \bar{v} \ll \gamma_l + \gamma_n$, then the Raman scattering has a Lorentz shape in the angle interval $|\theta| \lesssim (\gamma_l + \gamma_n)/k_\mu \bar{v}$

$$\frac{(\gamma_l + \gamma_n)/\gamma_n}{(\gamma_l + \gamma_n)^2 + (\Omega_\mu - \Omega)^2}$$

and its width is determined by the natural damping of the initial and final states. On the other hand, for the direction $\theta = \pi$, the width ($2k \bar{v}$) is twice the width of the stepwise-transition line.

It is easy to show that the doublet-component intensities integrated with respect to Ω_μ do not depend on θ . Consequently, the width anisotropy means also an angular dependence of the ratio of the intensities at the maxima of the lines in the range from $k_\mu \gamma_m / (k_\mu + k) \gamma_n$ to $k_\mu \gamma_m / |k_\mu - k| \gamma_n$.

Formula (3.2) for $q \bar{v}$ is analogous to the expression for the line width of the Rayleigh scattering in a gas, $2k \bar{v} \sin(\theta/2)$ ^[1], which is obtained from (3.2) when $k = k_\mu$. Just as in the case of Rayleigh scattering, formulas (3.2) and (3.3) admit of a simple interpretation, if we consider Raman scattering as emission of a classical oscillator moving with velocity \mathbf{v} . A change-over to the c.m.s. of the oscillator changes the frequency ω of the external field by $\omega - \mathbf{k} \cdot \mathbf{v}$. The forced oscillations induced by the field also have a frequency $\omega - \mathbf{k} \cdot \mathbf{v}$. The internal motion in the atom (or in the molecule) with natural frequency ω_{ln} modulates the forced oscillation and leads to the appearance in the emission spectrum of a component of frequency $\omega - \mathbf{k} \cdot \mathbf{v} - \omega_{ln}$. Finally, for the wave emitted in the \mathbf{k}_μ direction, the inverse transition to the stationary system of coordinates yields a frequency $\omega - \omega_{ln} - (\mathbf{k}_\mu - \mathbf{k}) \cdot \mathbf{v}$, and averaging over \mathbf{v} leads to a Doppler width $|\mathbf{k}_\mu - \mathbf{k}| \bar{v} = q \bar{v}$.

4. DOPPLER BROADENING OF RESONANT-SCATTERING DOUBLET

Let us turn now to the strong-field problem and assume that $G \gg k\bar{v}$. Here, too, we can neglect the "interference" terms $(\alpha_1 + \alpha_2^*)^{-1}$ and $(\alpha_2 + \alpha_1^*)^{-1}$ in the expression (2.6) for w_μ , and the Doppler shifts of $\alpha_{1,2}$ can be taken into account in the first nonvanishing approximation:

$$\alpha_j = \alpha_{j0} - iM_j k\bar{v}, \quad j = 1, 2, \quad (4.1)$$

where α_{j0} are the values of α_j at $\mathbf{k} \cdot \mathbf{v} = 0$ and M_j is the "memory" factor determined by formula (2.10). Neglecting the difference between α_j and α_{j0} everywhere except in the resonance denominators, we get from (2.6)

$$\langle w_\mu \rangle = \frac{|GG_\mu|^2}{\Omega^2 + 4G^2} \operatorname{Re} \left\langle \frac{\gamma_m^{-1}}{\gamma_l + \alpha_{10}^* + i[\Omega_\mu - (\mathbf{k}_\mu - M_1 \mathbf{k}) \cdot \mathbf{v}]} \times \frac{\gamma_n^{-1}}{\gamma_l + \alpha_{20}^* + i[\Omega_\mu - (\mathbf{k}_\mu - M_2 \mathbf{k}) \cdot \mathbf{v}]} \right\rangle. \quad (4.2)$$

Averaging of this expression leads to a formula of the type (3.2), in which the quantities $p_{1,2}$ should be replaced by

$$p_j = [\gamma_l + \alpha_{j0}^* + i\Omega_\mu] / q_j \bar{v}, \quad q_j = \sqrt{(k_\mu - M_j k)^2 + 4M_j k k_\mu \sin^2(\theta/2)}, \quad j = 1, 2, \quad (4.3)$$

and the widths $k_\mu \bar{v}$ and $q \bar{v}$ should be replaced by $q_1 \bar{v}$ and $q_2 \bar{v}$. Thus, the widths and the positions of both lines in the $m \rightarrow l$ transition depend on the frequency and direction of propagation of the absorbed photon, and its role is determined by the "memory" factor $M_{1,2}$. In the case of exact resonance we have $M_1 = M_2 = 1/2$ and $\alpha_{10}'' = -\alpha_{20}'' = J$, i.e., both lines are symmetrical relative to the frequency ω_{ml} and have an identical angular dependence of the width (Fig. 3b). It is interesting that when the condition $k_\mu = M_1 k$ or $k_\mu = M_2 k$ is satisfied, one of the lines has a natural width in the case of observation along $\theta = 0$ (see the discussion of formulas (3.2) and (3.3)).

Thus, variation of Ω leads to the following changes in the spectrum. When $|\Omega| \gg G$, one component of the doublet is near ω_{ml} and the other near $\omega - \omega_{ln}$. With decreasing $|\Omega|$, the shifted component moves towards the unshifted one, the latter shifts in the same direction, and the rate of motion ($M_{1,2}$) is larger for the component that is farthest from ω_{ml} . The distance between the components is $\sqrt{\Omega^2 + 4G^2}$. When $\Omega = 0$, the splitting is symmetrical, and the distance between components is minimal ($2G$). Further change of Ω brings the previously-shifted component closer to ω_{ml} , and moves the previously-unshifted component at an increased rate. In addition to the shift of the lines, their Doppler widths also change in accordance with the memory factors $M_{1,2}$ and the observation direction. The widths have minimal values $|k_\mu - M_{1,2} k| \bar{v}$ along the direction $\theta = 0$, and a maximal value $(k_\mu + M_{1,2} k) \bar{v}$ in the opposite direction.

We recall that the use of the obtained results for an analysis of other two-photon processes implies a reversal of the signs of Ω and Ω_μ in accordance with whether a particular photon is absorbed or emitted. In the case of two-quantum luminescence and absorption, the quantities $\pm(\Omega + \Omega_\mu)$ are involved. Therefore, unlike Raman

scattering, the minimum of the Doppler width will be reached at $\theta = \pi$. The magnitude of the narrowing will be the same as before.

5. DOPPLER BROADENING OF RESONANCE-FLUORESCENCE LINE ON EXCITED LEVELS

The results of Sec. 2 allow us to explain the correlation and frequency properties of the radiation also in the case of a transition between the levels that interact with the strong field ($m \rightarrow n$). Within the framework of second-order perturbation theory, the radiation power w_μ is determined directly by formula (3.2) in which we put $\mathbf{k} = \mathbf{k}_\mu$ and $\gamma_l = \gamma_n$. In the angle interval $\theta \lesssim \gamma_n/k\bar{v}$, the second term will have a dispersion form with width $2\gamma_n$. Thus, the main conclusions of Sec. 3 concerning the width anisotropy, the line shift, etc. apply also to resonance fluorescence¹⁾.

In the case of a strong field G , a singularity of the transition is the need for taking into account the field disturbance of both equations—both the upper and the lower. As a result, the formula for w_μ , in the case of a strong field, is more complicated than (2.6). For our purposes it suffices, however, to use the general conclusions of Sec. 2. From formula (2.3) it is easy to conclude that w_μ will consist of four transitions between two sublevels of the upper state and two sublevels of the lower state. Each of these transitions contributes its own resonant term:

$$[2\alpha_1' + i(\Omega_\mu - \Omega - q\bar{v})]^{-1}, \quad [\Gamma + i(\Omega_\mu - \mathbf{k}_\mu \mathbf{v} - 2\alpha_1'')]^{-1}, \quad (5.1)$$

$$[2\alpha_2' + i(\Omega_\mu - \Omega - q\bar{v})]^{-1}, \quad [\Gamma + i(\Omega_\mu - \mathbf{k}_\mu \mathbf{v} - 2\alpha_2'')]^{-1}.$$

In the general case these terms differ in the positions of their maxima (as functions of Ω_μ), in their widths, and in the coefficients with which they enter in w_μ . We consider the simplest and most striking case when $G \gg |\Omega|$, $k\bar{v}$, Γ , γ_l . The amplitudes of all the substates are the same here, and the fact (5.1) enter in w_μ with equal weights. Further, formula (2.8) is valid for α_1 and α_2 , and consequently $\langle w_\mu \rangle$ is given by

$$\langle w_\mu \rangle \propto \langle [\Gamma + i(\Omega_\mu - \Omega - 2G - q\bar{v})]^{-1} + 2[\Gamma + i(\Omega_\mu - \Omega - q\bar{v})]^{-1} \times [\Gamma + i(\Omega_\mu - \Omega + 2G - q\bar{v})]^{-1} \rangle, \quad (5.2)$$

$$q = k_\mu - k, \quad q = 2k \sin(\theta/2).$$

The position of the maximum of one of the terms coincides with the frequency ω of the strong field, and the two others are shifted to the points $\omega \pm 2G$. The widths of all the components of the triplet have identical angular characteristics. At large observation angles, the contours of the lines have a Gaussian form with width $q\bar{v}$. Inside the cone $\theta < \Gamma/k\bar{v}$, the lines have a dispersion form with natural width Γ .

It is of interest to trace the connection between the components of the triplet (5.2) of the resonant fluorescence with the lines of the stepwise transition or Rayleigh scattering. If we successively increase the deviation from resonance, say in the direction of positive Ω , then the component in (5.2) of frequency $\Omega - 2G$ will shift towards ω_{mn} and change into a stepwise-

¹⁾ Resonance fluorescence is usually considered for the case when the lower level is the ground level ($\gamma_n = 0$), and transitions from m are allowed only to n . Neither premise is satisfied in our problem.

transition line. The frequency of the central component will increase and coincide at all time with ω , yielding a Rayleigh-scattering line. Finally, the third component will move away from ω_{mn} at a still larger length, and its amplitude will become of the order of G^4/Ω^4 (i.e., it disappears in the second approximation of perturbation theory). When Ω changes in the opposite direction, the unshifted component, as before, remains at the frequency of the external field, and the roles of the shifted components are interchanged.

Thus, the changes of the frequency-correlation properties due to an external field become manifest in the Doppler broadening of the resonance fluorescence lines to the same degree as in Raman scattering. In addition, there appears one more line that does not fit in the classification of second-order perturbation theory.

¹I. M. Fabelinskiĭ, *Molekulyarnoe rasseyanie sveta* (Molecular Scattering of Light), Nauka, 1965 [Consul-

tants Bureau, 1968].

²W. Heitler, *The Quantum Theory of Radiation*, Oxford, 1954.

³S. G. Rautian and I. I. Sobel'man, *Zh. Eksp. Teor. Fiz.* 44, 934 (1963) [*Sov. Phys.-JETP* 17, 635 (1963)].

⁴G. E. Notkin, S. G. Rautian, and A. A. Feoktistov, *ibid.* 52, 1673 (1967) [25, 1112 (1967)].

⁵S. G. Rautian, *Trudy FIAN* 43, 3 (1968).

⁶S. H. Autler and C. H. Townes, *Phys. Rev.* 100, 703 (1955).

⁷V. M. Kontorovich and A. M. Prokhorov, *Zh. Eksp. Teor. Fiz.* 33, 1428 (1957) [*Sov. Phys.-JETP* 6, 1100 (1958)].

⁸S. G. Rautian and I. I. Sobel'man, *ibid.* 41, 456 (1961) [14, 328 (1962)].

⁹A. M. Bonch-Bruевич and V. A. Khodovoi, *Usp. Fiz. Nauk* 93, 71 (1967) [*Sov. Phys.-Uspekhi* 10, 637 (1968)].

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