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SPECTRAL OPTICAL-TRANSITION LINE SPLITTING INDUCED IN GASES BY RESONANT EXTERNAL FIELDS

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An analysis is presented of the effect of inhomogeneous broadening on spectral line splitting induced by an additional field which is resonant with respect to the adjacent transition. It is demonstrated that the manifestation of the effect depends strongly on the relation between the frequencies and directions of propagation of the emitted and auxiliary fields. Even in case of strongly inhomogeneous broadening and under certain other conditions, the effect may arise when the interaction energies between the atom and auxiliary field are comparable with the widths of the energy levels.

1. Under the influence of a field that is in resonance with an adjacent optical transition, the spectral emission line of an atom can split into two components. To this end it is necessary that the energy of interaction between the atom and the external field be comparable with the characteristic widths of the energy levels. This phenomenon can be interpreted as a lifting of the degeneracy between the state of the atom-plus-field system, when there are l photons in the field and the atom is in a state n, and the state in which there are $l - 1$ photons in the field and the atom is in the state m ($E_m > E_n$). The splitting of the spectral lines can be interpreted as the splitting of the overall energy level of the interacting transitions into quasienergy sublevels. This effect has analogs, for example, in the modification of the dispersion laws of resonantly-interacting quasiparticles.

For optical transitions, the widths of the energy levels are smallest in gases. In gases, however, the broadening of the spectral lines is as a rule inhomogeneous. It would seem natural that under these conditions the effect of splitting of the spectral line of an individual atom should be leveled out as a result of the presence of the Maxwellian distribution of the atoms with respect to velocities, and that in order for the splitting effect to become manifest it would be necessary to have an external field of intensity high enough to make the interaction energy comparable with the Doppler width of the line (in units of h)[1-4]. As will be shown below, the dynamics of the interaction of two traveling waves of a field with a gas of moving atoms is such that the appearance of the splitting effect depends strongly on the ratio of the moduli and the directions of the wave vectors. It turns out that at a definite ratio of these quantities the effect of splitting of the line of an individual atom becomes manifest at interaction energies comparable with the widths of the levels, even under conditions when the Doppler broadening is much larger than the impact broadening.

2. Let us examine the power of emission (absorption) of a weak field at the transition ml ($E_m > E_l$) in the presence of an additional field at the transition mn ($E_m > E_n$). The action of the additional field reduces only to the splitting effect if the population difference $N_m - N_n$ at the transition mn is equal to zero. In this case the formula for the emission power (if $N_m - N_l > 0$) or absorption power (if $N_m - N_l < 0$) at the frequency ω_μ is given by

$$w_{ml} = 2\hbar\omega_{ml}|G_\mu|^2 \frac{N_m - N_e}{\sqrt{\pi} k_\mu \bar{v}} \times \operatorname{Re} \int_{-\infty}^{\infty} \frac{\exp[-(\mathbf{v}/\bar{v})^2]\, d(\mathbf{k}_\mu \mathbf{v})}{\Gamma_{ml} - i\Omega_\mu' + |G|^2[\Gamma_{ln} + i(\Omega' - \Omega_\mu')]^{-1}}. \tag{1}$$

Here $|G_\mu|^2 = |\mathbf{E}_\mu \mathrm{d}_{lm}/2\hbar|^2$, $|G|^2 = |\mathbf{E}\mathrm{d}_{nm}/2\hbar|^2$, Ω_μ' and Ω' are the yields of the atomic resonances for the corresponding fields with allowance for the Doppler shift

$$\Omega_\mu' = \omega_\mu - \omega_{ml} - \mathbf{k}_\mu \mathbf{v} = \Omega_\mu - \mathbf{k}_\mu \mathbf{v};$$

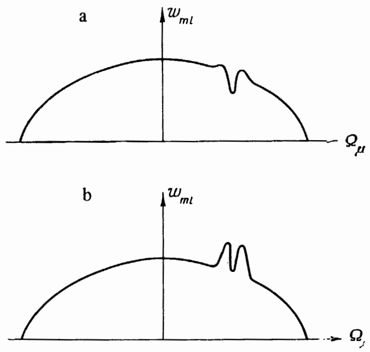
$\Omega' = \omega - \omega_{mn} - \mathbf{kv} = \Omega - \mathbf{kv}$, ω_{ij} and Γ_{ij} are the frequencies and the Lorentz half-widths for the corresponding transitions; ω_μ, $\mathbf{k}_\mu$ and ω, $\mathbf{k}$, are the frequencies and wave vectors for the analyzed and supplementary field, and $\bar{v}$ is the half-width of the Maxwellian velocity distribution. Putting $N_l = 0$ in (1) and replacing $|G_\mu|^2$ by the corresponding quantity for the vacuum oscillations, we can use the expression for the analysis of the spectral properties of spontaneous emission in an external field. From (1) we see that at $G \neq 0$ the denominator of the integrand as a function of Ω_μ' has two roots. Consequently, the formula for w_{ml} can be represented in the form of a sum of two spectral components with different amplitudes and positions of the maxima. This circumstance is a reflection of the effect of splitting of the spectral line of the individual atom.

3. When $|G|^2/\Gamma_{ln}\Gamma_{lm} \ll 1$ we obtain, accurate to $|G|^2$,

$$w_{ml} = 2\hbar\omega_{ml}|G_\mu|^2 \frac{N_m - N_l}{\sqrt{\pi} k_\mu \bar{v}} \int_{-\infty}^{\infty} d(\mathbf{k}_\mu \mathbf{v}) \exp\left[-\left(\frac{\mathbf{v}}{\bar{v}}\right)^2\right] \left\{ \frac{\Gamma_{lm}}{\Gamma_{lm}^2 + (\Omega_\mu - \mathbf{k}_\mu \mathbf{v})^2} - \operatorname{Re} \frac{|G|^2}{[\Gamma_{lm} - i(\Omega_\mu - \mathbf{k}_\mu \mathbf{v})]^2 \, [\Gamma_{ln} + i(\Omega - \Omega_\mu) - i(\mathbf{k} - \mathbf{k}_\mu)\mathbf{v}} \right\}.$$

It is easy to see that the poles of the last term as a function of $\mathbf{k}_\mu \cdot \mathbf{v}$ always lie in the same half-plane, with the exception of the case $\mathbf{k}_\mu \cdot \mathbf{k} > 0$ and $k_\mu < k$. Thus, for the case of parallel and antiparallel waves, we obtain if $\Gamma_{lm}, \Gamma_{ln} \ll k_\mu \bar{v}$

$$w_{ml} = 2\hbar\omega_{ml}|G_\mu|^2 (N_m + N_l) \frac{\sqrt{\pi}}{k_\mu \bar{v}} \exp\left[-\frac{\Omega_\mu}{k_\mu \bar{v}}\right] \times \left\{1 - \Theta(\mathbf{k}\mathbf{k}_\mu - k_\mu^2) \frac{k - k_\mu}{k} \frac{k_\mu}{k} \frac{\Gamma_+^2 - z^2}{(\Gamma_+^2 + z^2)^2} |G|^2 \right\}, \tag{2}$$



Effect of splitting of the spectral line for the case of parallel waves: a–$N_m - N_n = 0$, 6–$N_m - N_n = -(N_m - N_l)\, k_\mu - k/k\ \Gamma_m/\Gamma_+$.

where

$$\Theta(x)=\begin{cases}0, & x<0\\ 1, & x>0\end{cases};$$

$$\Gamma_+ = \frac{k_\mu}{k}\Gamma_{ln} + \left(1-\frac{k_\mu}{k}\right)\Gamma_{lm}; \quad z=\Omega_\mu - \frac{k_\mu}{k}\Omega.$$

It follows from (2) that even when $k_\mu \bar{v} \gg \Gamma_{lm}$ the effect of splitting can appear in relatively weak fields $|G|^2 \sim \Gamma_+^2$, if $\mathbf{k}_\mu \cdot \mathbf{k} > 0$ and $k_\mu < k$. It gives rise to a singularity at the frequency $\Omega_\mu = (k_\mu/k)\Omega$ against the background of the Doppler curve. As a function of Ω_μ, this contribution of the strong field reverses sign, so that the singularity has the form of a "dip on a pedestal" (see Fig. a). In the case of antiparallel waves, and for arbitrary propagation directions, when $k_\mu > k$, the dip can appear only at $|G|^2 \sim (k_\mu \bar{v})^2$ [1]. It can be shown that in the general case $N_m - N_n \neq 0$ the form of the singularity at $k < k$ and $\mathbf{k}_\mu \cdot \mathbf{k} > 0$ (see Fig. b) is described by the formula

$$I = -\frac{k_\mu}{k}\frac{|G|^2}{\Gamma_m\Gamma_+}\left[(N_m - N_n)\frac{\Gamma_+^2}{\Gamma_+^2+z^2} + (N_m - N_l)\frac{k-k_\mu}{k}\frac{\Gamma_m\Gamma_+(\Gamma_+^2 - z^2)}{(\Gamma_+^2+z^2)^2}\right].$$

When

$$|N_m - N_l| \gg (|G|^2/\Gamma^2_{min})\,|N_m - N_n|,\ \Gamma_{min} = \min\{\Gamma_{ik}\},\ i, k = \{l, m, n\}$$

a similar singularity arises also in the spectrum of generation at the transition ml in the presence of an additional field at the transition mn. The splitting effect does not appear when $(1 - k_\mu/k) \ll 1$. Thus, besides spectroscopic applications, this effect can be used to stabilize the generation frequency in arbitrary sections of the Doppler curve.

At the first All-Union Symposium on Gas-Laser Physics (Novosibirsk, July 1969), M. S. Feld told the author that he obtained analogous results theoretically[5], and P. Toschek reported an experimental observation of this effect in neon at the transition $2s_2 - 2p_4$, when the transition of the supplementary field was $3_2 - 2p_4$[6].

In conclusion, I am grateful to S. G. Rautian for useful discussions of the present results.

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