Increase of the cross section and narrowing of the line of two-photon absorption as a result of the Dicke effect

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(Submitted October 8, 1976)
Pis'ma Zh. Eksp. Teor. Fiz. **24**, No. 10, 555–557 (20 November 1976)

We demonstrate the possibility of complete elimination of the uncompensated Doppler broadening in two-photon absorption. Narrowing down to the natural line width is connected with the Dicke effect and is accompanied by an abrupt increase of the absorption. The use of single-photon resonance for this purpose increases additionally the two-photon absorption cross section.

PACS numbers: 42.65.Bp

1. The angular anisotropy of the Doppler width of the absorption line of a weak field in the presence of a strong field at an adjacent transition was investigated in[1,2] In the succeeding years, the scheme of two-photon absorption of equal photons with opposite momenta has gained wide application. The corresponding method of nonlinear spectroscopy has been named non-Doppler two-photon spectroscopy (for references see[3]). The absorption of two equal photons under conditions of two-photon resonance is possible as a rule only at appreciable deviations from the intermediate single-photon resonance, a situation accompanied by the corresponding decrease of absorption. For this reason, schemes with two unequal photons were used subsequently for the purpose of coming closer to the intermediate resonance. This, however, has resulted in an incomplete cancellation of the Doppler width of the two-photon transition. The latter limits the spectroscopic application of the method and lowers the possible increase in the absorption cross section.

2. We propose to use elastic collisions with change of velocity for a complete cancellation of the Doppler broadening in a scheme with intermediate resonance and photons of different frequency. The proposed method is a modification of that considered in[4]. The Dicke effect, however, is used to eliminate the uncompensated Doppler width $\Delta k\bar{v}$ of the IR or optical two-photon transitions, and not the total inhomogeneous width of the real microwave transition as in[4]. This results in a narrowing of the Doppler line and an increase of the two-photon absorption by a factor $\Delta k\bar{v}/\Gamma_{ln}$, where Γ_{ln} is the homogeneous width of the two-photon transition. In addition, an appreciable increase of the absorption cross section is made possible by the approach to the single-photon intermediate resonance.

3. Consider the interaction of two monochromatic waves with opposite wave vectors k and k_μ with a three-level system $(E_l > E_m > E_n)$, n being the ground level. The frequency ω of the strong field E is close to the frequency of the one-photon transition mn. We analyze the line shape of the absorption of the weak field E_μ with frequency ω_μ close to the frequency of the single-photon transition ω_{lm}, with $\omega_\mu + \omega \approx \omega_{ln}$. The absorption cross section of this field is

proportional to the imaginary part of the density matrix element ρ_{ln}. The equation for the density matrix $\hat{\rho}$ is

$$\left(\frac{\partial}{\partial t} + v\vec{\nabla}\right)\hat{\rho} = -i[\hat{V}, \hat{\rho}] + R + \left(\frac{d\rho}{dt}\right)^{\mathrm{coll}}. \tag{1}$$

Here $\mathbf{v}$ is the velocity of the atom (molecule). $\hat{V}$ is the Hamiltonian of the interaction with the fields E and E_μ, R describes the spontaneous relaxation, and $(d\rho/dt)^{\mathrm{coll}}$ describes the change of the atom velocity as a result of elastic collisions.

To illustrate the main features of the phenomenon, it suffices to retain the corresponding term $(d\rho/dt)^{\mathrm{coll}}$ only in the equation for ρ_{ln}. We confine ourselves here to the model of strong collisions, when the change of the velocity $\mathbf{v}$ in one collision is of the order of thermal velocity $\bar{v}$

$$\left(\frac{d\rho}{dt}\right)^{\mathrm{coll}}_{ln} = -\nu\rho_{ln}(v) + \tilde{\nu}\int W(v')\rho_{ln}(v')\,dv'. \tag{2}$$

Here v is the projection of the velocity on the wave-propagation direction; W is a Maxwellian distribution, and ν and $\tilde{\nu}$ are the collision frequencies with change of velocity. According to[4], we obtain, accurate to $|E|^2$, the following expression for the absorption cross section at the frequency ω_μ:

$$\sigma(\omega_\mu) \sim \left[\frac{2\Gamma_{lm}\Gamma_{mn}}{\Gamma_m|(\omega_\mu - \omega_{lm})(\omega - \omega_{mn})|} + \mathrm{Re}\,\frac{z(\omega_\mu)}{1 - \tilde{\nu}Z(\omega_\mu)}\right]\frac{|G|^2}{|(\omega_\mu - \omega_{lm})(\omega - \omega_{mn})|}. \tag{3}$$

Here Γ_{lm} and Γ_{mn} are the homogeneous widths of the transitions, Γ_m is the width of the level m, $G = -\mathrm{E}d_{mn}/2\hbar$, d_{mn} is the matrix element of the electrodipole transition, $|\omega_\mu - \omega_{lm}| \gg k_\mu\bar{v}$, $|\omega - \omega_{mn}| \gg k\bar{v}$, and

$$Z(\omega_\mu) = \pi^{-1/2}\int_{-\infty}^{\infty}\frac{\exp\{-(v/\bar{v})^2\}\,dv/\bar{v}}{\Gamma_{ln} + \nu - i(\omega_\mu + \omega - \omega_{ln} - \Delta k v)}. \tag{4}$$

Here $\Delta k = |\omega_\mu - \omega|/c$ and $\Gamma_{ln} = \Gamma_l/2$ is the width of the ln transition and is determined by the relaxation without change of velocity (radiative and collision). The first term in (3) corresponds to a stepwise transition, while the second corresponds to a two-photon transition. We consider the following limiting cases:

$$1)\ \mathrm{Re}\,Z = \Gamma_{ln}/[\Gamma_{ln}^2 + (\omega_\mu + \omega - \omega_{ln})^2] \quad \text{at} \quad \nu = \tilde{\nu} = 0, \quad \Delta k = 0. \tag{5}$$

This case corresponds to $k_\mu = k$ and to complete cancellation of the Doppler broadening

$$2)\ \mathrm{Re}\,Z = (\sqrt{\pi}/\Delta k\bar{v})\exp\{-[(\omega_\mu - \omega + \omega_{ln})/\Delta k\bar{v}]^2\}, \quad \text{at} \quad \nu = \tilde{\nu} = 0, \quad \Delta k\bar{v} >> \Gamma_{ln} \tag{6}$$

corresponding to the appearance of uncompensated Doppler broadening.

$$3)\quad \text{at}\ \ \nu >> \Delta k\bar{v}\begin{cases} Z = [\Gamma_{ln} + \nu - i(\omega_{\mu} - \omega + \omega_{ln})]^{-1} \\ \mathrm{Re}\,Z[1 - \tilde{\nu}Z]^{-1} = (\Gamma_{ln} + \nu - \tilde{\nu})/[(\Gamma_{ln} + \nu - \tilde{\nu})^2 + (\omega_{\mu} + \omega - \omega_{ln})^2]. \end{cases} \quad (7)$$

Thus, if the frequency of the collisions with change of velocity is $\nu \gg \Delta k\bar{v}$ (i.e., the mean free path of the atoms is $l = \bar{v}/\nu \ll \Delta k^{-1}$) and the change of the velocity is not accompanied by a substantial loss of phase matching of the atomic oscillator ($\nu - \tilde{\nu} \ll \Delta k\bar{v}$), and the uncompensated Doppler broadening of the two-photon transition (two-photon Dicke effect) is eliminated. Consequently, the use of the Dicke effect makes it possible, while retaining all the advantages of the intermediate resonance, to narrow down the width of the two-photon resonance. The narrowing is accompanied by an increase of the absorption at the maximum by a factor $\Delta k\bar{v}/(\Gamma_{ln} + \nu - \tilde{\nu})$.

After this paper was sent to press, an article appeared,[5] in which a similar narrowing was observed in the molecular gas NH_3 in two-photon absorption of the emission of two CO_2 lasers generating at different vibrational-rotational transitions. The change in the velocity of the NH_3 molecules with a small loss of phase matching was effected by adding the inert gas Ne as a buffer.

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