Narrowing of two-photon resonance due to elastic collisions

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The two-photon absorption line shape is examined in the field of a standing wave with elastic collisions. A sharp decrease is observed in the transit broadening of the two-photon resonance in vibrational-rotational transition of molecules due to an increase in the interaction time of particles with the electromagnetic field as a result of elastic collisions. This effect permits obtaining supernarrow optical two-photon absorption resonances with high intensity in a low-pressure gas.

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It was shown in Ref. 1 that the nonlinear particle-density dependence of the width of the unsaturated absorption resonance on vibrational-rotational transitions of mole-

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cules is due both to elastic scattering of particles at small angles and the small contribution to broadening by collisions which interrupt the phase of the dipole moment of the particle. The small cross section for collisions without phase interruption can greatly facilitate obtaining very narrow optical lines. However, the width of the saturated-absorption resonance at low gas pressure is determined by the total elastic scattering cross section, irrespective of whether the phase of the dipole moment of the particle is interrupted during the collision (in the latter case, collisions are accompanied by particles leaving resonance due to the large Doppler shift in the frequency with a change in the velocity of the atom). Narrowing accompanying collisions of the Doppler contour is possible only with sufficiently high gas pressures, when the free path length of particles is of the order of or less than the wavelength of the electromagnetic field. For a wavelength of 1 μm and thermal velocity $\sim 10^5$ cm/s, pressures of $\sim$ 1–10 atm are necessary, but at such pressures, in spite of the decrease in the width of the resonance compared to the Doppler width, the width, limited by inelastic scattering and scattering with phase interruption, remains large ($\sim$ 1 GHz; see, for example, Ref. 2).

A completely different situation occurs in a low-pressure gas for a two-photon resonance in the field of a standing wave.[3] As is well known, elimination of Doppler broadening of the two-photon resonance is related to the fact that the process of absorption of two countermoving photons does not depend on the velocity of the particles, so that elastic collisions, in contrast to the saturation absorption resonance, do not contribute to broadening. The width of the two-photon absorption resonance (TPA) in the absence of collisions is determined by the transit time of the atom across the light beam. In this paper, we shall demonstrate the possibility of significant narrowing of the transit contour of the resonance due to elastic collisions for vibrational-rotational transitions of molecules. The effect occurs when the free path of the atom is less than the diameter of the light beam, which leads to an increase in the interaction time of particles with the field and narrowing of the resonance. The collisional broadening of the line is small because the cross section of inelastic and phase-interrupting collisions is small.

1. Qualitative analysis. We shall assume that the phase of the oscillator is not interrupted by elastic collisions. As a result of such collisions, the duration of the interaction of the particle with the field effectively increases and the TPA resonance has a width determined by the diffusion time of the molecule in the light beam. If at time t the particle is located, for example, on the axis of the light beam, then at a later time t' the probability density to observe the particle at a distance R is proportional to $\exp[-R^2/4D(t'-t)]$, where D is the diffusion coefficient. The diffusion time in the light beam with transverse dimension is

$$\tau_g = a^2/4D = \nu_g \tau_0^2 / 2,$$

where $\nu_g = v_0/2D$ is the diffusional collision frequency ($v_0 = \sqrt{2kT/M}$ is the thermal velocity, T is the temperature, M is the mass of the molecule), and $\tau_0 = a/v_0$ is the transit time. The width of the two-photon resonance $\gamma \sim \tau_g^{-1} + \Gamma_{21}$ where Γ_{21} is the homogeneous width of the transition 1→2, including the radiative decay of levels 1 and 2, as well as inelastic collisions. Since τ_g and Γ_{21} are proportional to the gas pressure P,

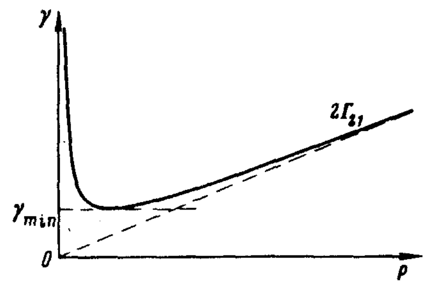

FIG. 1. Width γ of a two-photon absorption resonance as a function of the pressure P in the presence of elastic collisions.

$$\gamma \sim \alpha / P + \beta P. \tag{1}$$

The curve of the dependence $\gamma(P)$ is shown qualitatively in Fig. 1. In accordance with (1), the minimum width is

$$\gamma_{\min} \sim \sqrt{\alpha\beta} = \sqrt{\Gamma_{21}/\tau_g}\ .$$

We note that the additional increase in the coherent interaction time of particles with the field can be attained when the phase of the oscillator is not interrupted by collisions with the walls.

In essence, in the case that we have examined, the line is observed to narrow when the free path of the particles Λ is much longer than the wavelength λ, while for single-photon transitions, line narrowing occurs under the condition $\lambda \ll \Lambda$.[4]

2. *Line shape.* The two-photon absorption line shape in the field of a standing wave in the presence of elastic collisions was found by using the method of the quantum kinetic equation for the density matrix[5] and the approximations of strong and weak collisions. For example, for weak collisions and equal scattering amplitudes in levels 1 and 2, the TPA line shape is

$$L(\delta) = \frac{1}{\pi} \mathrm{Re} \int_0^\infty \frac{\exp[-(\Gamma_{21} + i\delta)t]\,dt}{1 + 2(\nu_g \tau_0)^{-2}(\nu_g t - 1 + e^{-\nu_g t})}\,, \tag{2}$$

where δ is the detuning of the doubled frequency of the field 2ω relative to the transition frequency ω_{21}, including the shift due to inelastic collisions. If the diffusion time τ_g is greater than the time Γ_{21}^{-1}, then $L(\delta)$ is the Lorentz contour with width $\gamma = 2\Gamma_{21} \propto P$ at half-height. The case $\tau_0 \ll \tau_g \ll \Gamma_{21}^{-1}$ corresponds to narrowing of the TPA resonance due to elastic collisions. The width of the line, which is much less than the transit width, is

$$\gamma = \gamma_{\min} = 1{,}5\ \sqrt{\Gamma_{21}/\tau_g}\ .$$

At very low pressures $(\nu_g \ll \tau_0^{-1})$ $L(\delta)$ is the usual transit contour with width $\gamma = 1.4\tau_0^{-1}$ (see also Ref. 6).

The strong collision model gives $L(\delta)$ in a form differing from (2), but in limiting cases, analogous to those examined here, the results coincide. This again indicates that the change in the velocity of the particle with collisions does not affect the TPA resonance; only the time in which the molecule leaves the light beam is important.

3. The phenomenon of narrowing of the two-photon resonance examined here can be observed, for example, in vibrational-rotational transitions of molecular hydrogen. Estimates, which we made using Ref. 2, for the transitions $v_1 = 0, J \rightarrow v_2 = 1, J$ (v, J are the vibrational and rotational quantum numbers of the molecules in the ground state) show that due to such narrowing, TPA resonances can be obtained with widths $\gamma = 7$–10 kHz at pressures $0.01 < P < 0.1$ Torr (the width decreases by a factor ~ 10 compared to the transit width). The TPA resonance has high intensity because of the increase in the interaction time of the molecule with the field in the presence of elastic collisions and high particle density ($\sim 10^{15}$ cm^{-3}). For the number of particles excited into level 2 per unit time and per unit length of the working cell, we have

$$dN_2/dt = 8 \times 10^9 \text{ s}^{-1} \text{ cm}^{-1}$$

(the transition $v_1 = 0, J = 1 \rightarrow v_1 = 1, J = 1$; field intensity $I = 1$ W/cm^2; beam size $a = 1$ cm; temperature $T = 300$ K).

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