Influence of dynamic Stark effect on the absorption of a test field

L. S. Vasilenko, N. M. Dyuba, N. N. Rubtsova, and V. P. Chebotaev
Institute of Thermal Physics, Siberian Branch, Academy of Sciences of the USSR

(Submitted 25 February 1988)
Pis'ma Zh. Eksp. Teor. Fiz. **47**, No. 7, 332–334 (10 April 1988)

Experiments show that the absorption of the weak test field tends toward zero with increasing intensity of the strong wave if the waves are propagating in the same direction. In contrast, the absorption remains on the order of the unsaturated absorption if the waves are propagating in opposite directions. The results are explained on the basis of a manifestation of a dynamic Stark effect of energy levels.

As the intensity of a field which is interacting resonantly with particles is increased, we know that the populations of the levels become equalized, and the absorption of the field tends toward zero (saturation effects). In a gas, the Doppler effect has the consequence that the difference between populations is erased only near the velocities of those particles which are interacting resonantly with the field. It is natural to expect that the absorption of a weak wave which is interacting with the same particles will also tend toward zero with increasing intensity of the saturating field. This letter contains the first report of an unusual behavior of a counterpropagating weak test wave, whose absorption tends toward a constant value, equal in order of magnitude to the unsaturated value, with increasing strength of the saturating field.[1,2] The absorption of a test wave which is propagating in the same direction as the strong wave tends toward zero. This behavior is attributed to a manifestation of a high-frequency Stark effect in inhomogeneously broadened systems. This Stark effect causes a qualitative change in the condition for a resonant interaction of the particles with the test field. The qualitative features seen in this study open up some new possibilities for spectroscopic studies of condensed and gaseous media.

An experiment was carried out to detect resonances in the saturated absorption under strong-saturation conditions in the gas $^{15}NH_3$ (74% enrichment in the isotope ^{15}N) on the vibrational-rotational transition $asQ(5, 4)$ of the ν_2 mode, which is reso-

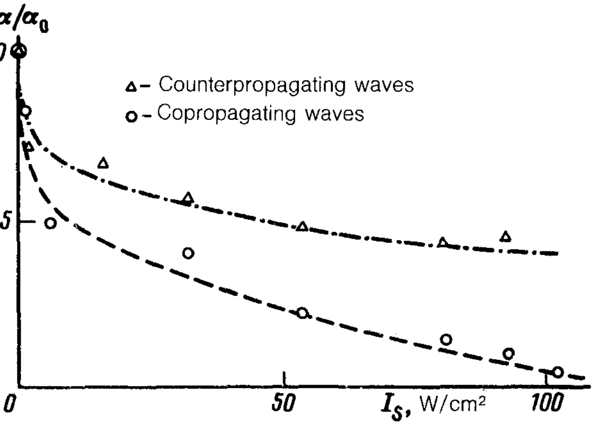

FIG. 1. Relative absorption coefficient of the test field, α/α_0, versus the intensity of the saturating light, I_S, for copropagating and counterpropagating waves in the gas $^{15}NH_3$.

nant with the $^{13}CO_2$ laser line $R(18)_I$. The beam from the $^{13}CO_2$ saturating laser was frequency-stabilized within ≈ 100 kHz on the basis of the resonance of the saturated absorption on the $asQ(5, 4)$ transition of $^{15}NH_3$. A linearly polarized test beam from a cw $^{13}CO_2$ waveguide laser, with the same line, $R(18)_I$, with a tuning interval ± 200 MHz could propagate either parallel or antiparallel to the saturating beam I_S at an angle $\theta \approx 4 \times 10^{-2}$ rad in a working cell 40 cm long and 6 mm in diameter. The power density of the saturating beam ranged up to 100 W/cm^2 and could be adjusted with the help of a polarizer-analyzer system.

The absorption coefficient for the test field, α, at the center of the line was varied over recordings of resonances of the saturated absorption. Results of measurements of α/α_0 as a function of the intensity of the saturating light, I_S, in the copropagating and counterpropagating cases are shown in Fig. 1 for a $^{15}NH_3$ pressure ≈ 10 mtorr. In the case of the copropagating waves, we observe a complete bleaching for the $^{15}NH_3$ pressure interval studied at the maximum power density I_S. The ratio α/α_0 approaches zero with increasing I_S. For the counterpropagating waves, the dependence of α/α_0 on I_S reaches a nonzero level as I_S is increased. This behavior of the absorption for the test field in the copropagating and counterpropagating cases can be explained on the basis of a model of a dynamic Stark effect[1-3] in a gas of two-level atoms with a Doppler-broadened absorption line. Specifically, the strong resonant electromagnetic field of amplitude E acts on a gas of two-level atoms and causes a splitting of each level into two sublevels. As a result, the absorption at the center of the line is dominated by groups of particles with velocities[1] $(\overline{R}v)_{1,2} = \pm dE/\hbar\sqrt{3}^1$, and the absorption coefficient for the counterpropagating wave tends toward the value

$$\alpha/\alpha_0 = \frac{2\gamma/\Gamma}{3+\gamma/\Gamma}, \tag{1}$$

where $2/\gamma = 1/\gamma_1 + 1/\gamma_2$, and Γ is the homogeneous linewidth of the transition. Consequently, in counterpropagating beams an increase in the strong field is accompanied by an increase in both the level splitting $dE/\hbar$ and the width of the region of resonant atomic velocities, i.e., the width of the "Bennett hole," $\Gamma_B = \Gamma\sqrt{1+G}$, where $G \equiv (dE)^2/\hbar^2\gamma\Gamma$. As a result, the absorption of the counterpropagating wave tends toward a constant value which depends on γ/Γ. Looking at our experimental results on the counterpropagating case (Fig. 1), we find the limiting value for α/α_0 to be 0.45. We can use expression (1) and the limiting value of α/α_0 to find $\gamma/\Gamma = 0.87$. On the other hand, the relaxation rates γ_1 and γ_2 of the vibrational-rotational levels of $^{15}NH_3$, with quantum numbers in the ground and vibrationally excited states which are close to $J = 5$ and $K = 4$, are known[4]: $\gamma_2 = 7.4$ MHz/torr, $\gamma_1 = 34$ MHz/torr. We then calculated $\gamma \approx 12$ MHz/torr. The value of Γ was measured in Ref. 5 for the qR-(0, 0) transition: the result was $\Gamma \approx 13.3$ MHz/torr. We thus find $\gamma/\Gamma = 0.92$. This value agrees reasonably well with the value found from the experimental data.

The absorption coefficient for a copropagating test wave was calculated in Ref. 3 without any restriction on the field for a gas of two-level atoms. Level degeneracy was ignored. In the strong-saturation limit, the absorption α for a copropagating test wave, at the center of the line, approaches zero as $1/\sqrt{G}$. In contrast with the case of counterpropagating waves, the resonant interaction occurs at the frequency of the strong field, so the dynamic Stark effect leads to a situation in which there are no resonant atoms if the difference (Δ) between the frequencies of the saturating and test fields satisfies $\Delta < dE/\hbar$. The shape of the absorption line for the test field becomes a square dip with a width $2dE/\hbar$, within $1/\sqrt{G}$. The vanishing of the absorption for the test field in the case of copropagating waves in our experiments agrees qualitatively with the theoretical arguments of Refs. 1–3.

[1] V. P. Baklanov and V. P. Chebotaev, Zh. Eksp. Teor. Fiz. **61**, 551 (1971) [Sov. Phys. JETP **34**, 293 (1971)].
[2] S. Haroche and F. Hartmann, Phys. Rev. **6A**, 1280 (1972).
[3] E.V. Baklanov and V. P. Chebotaev, Zh. Eksp. Teor. Fiz. **61**, 922 (1971) [Sov. Phys. JETP **34**, 490 (1971)].
[4] T. Schimizu, N. Morita, T. Kasuga *et al.*, Appl. Phys. **21**, 29 (1980).
[5] T. Shimizu, F. Shimizu, R. Turner, and T. Oka, J. Chem. Phys. **55**, 2882 (1971).

Translated by Dave Parsons