Narrowing of nonlinear resonances in low-pressure gases

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We report the first observation of the effect of narrowing of nonlinear power resonance under the influence of slow atoms. This effect has made it possible to resolve the magnetic hyperfine structure of the $\lambda = 3.39\ \mu$ vibrational-rotational transition in methane ($F_2^{(2)}$ component of the $P(7)$ line of ν_3 band) in an He-Ne laser in which the transverse dimension of the light beam is ~ 4 mm.

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1. Observation of absorption-saturation resonances of width ~1 kHz in nethane at $\lambda = 3.39\ \mu$ was reported in[1,2]. These narrow resonances were obained at pressures $\sim 10^{-5}$ Torr with a telescopic beam expander[1] and inside he resonator[2] with a beam diameter ~20 and ~10 cm, respectively. Further ncrease of the laser-beam diameter for the purpose of increasing the time of nteraction of the particles with the field entails great technical difficulties. Other methods were recently proposed for obtaining narrow resonances, based on the use of extended particle beams with two-photon absorption[3] and on the Ramsey nonlinear optical resonance.[4] A simple method that increases the time of the coherent interaction of the atom with the field involves selection of the atoms in velocity. It is known that the Lamb dip is caused by saturation of the atoms whose velocity projection on the direction of the radiation propagation is close to zero. At very low pressures, the particle mean free path becomes much larger than the transverse beam dimension. In this case the slow atoms nteract with the field for a longer time than the atoms with average thermal elocity. The result, at low saturations, is that the slow atoms make the prinipal contribution to the Lamb dip, and its width is determined by the time of flight of these atoms through the laser beam. Nonlinear-resonance narrowing nder the influence of slow atoms was first pointed out long ago in[5], but has ot been observed so far. We report here the first observation of this phenomnon, which may be of importance for the production of ultranarrow resonances.

2. The narrowing of the power resonance at low gas pressures was observed n an He-Ne laser with a methane absorber. The laser resonator was made up f mirrors with curvature radii $R_1 = 10$ m and $R_2 = \infty$. The laser resonator length as ≈ 5 m. The light-beam diameter at the caustic was ≈ 4.5 mm.

The form of the power resonance of the investigated He-Ne laser was studied ith the aid of a reference laser whose frequency was stabilized against the nethane absorption line. The resonance was registered by means of a signal roduced by the second harmonic of the laser power as the laser emission freency was modulated, and this made it possible to increase the resolution of he measurement setup. The second-harmonic signal was recorded as a function of the change of the frequency investigated laser relative to the reference laser with an x–y recorder.

Figures 1(a) and 1(b) show the experimental results. At a methane pressure $\approx 360\ \mu$Torr, the shape of the observed power resonances is close to Lorentzian, and the magnetic hyperfine structure (MHFS) at the working transition of the methane ($F_2^{(2)}$ of the $P(7)$ transition of the ν_3 band) is not resolved. When the pressure decreased to 60 μTorr, it was possible to observe distinctly the hyperfine structure of the methane. Figure 2(b) shows three maxima corresponding to the centers of the lines of the individual MHFS components. The distance between the MHFS components was ≈ 11 kHz, in agreement with the calculated[6] and experimental data.[1,2,7]

3. A detailed analysis of the behavior of the width of the resonance in the region of low pressures for a Gaussian field profile was carried out in[8]. Figure 2 shows the calculated dependence of the homogeneous line width on the parameter $\Gamma\tau$ (Γ is the collision frequency and 2τ is the time of flight of the atoms through the beam with average thermal velocity).[8] At a given value of τ in the region $0.5 < \Gamma < 2$, the dependence of the resonance half-width γ on $\Gamma\tau$ (that is, on the impact width) can be represented in the form $\gamma = 0.58/\tau + \Gamma$. The increment $0.58/\tau$ to the impact half-width Γ is the cause of the contribution of the transit effects to the line width and determines the transit half-width

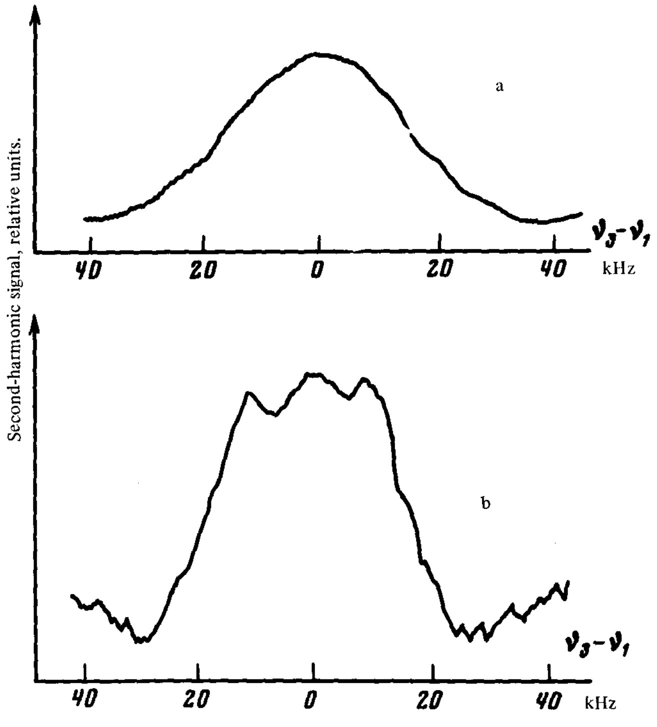

FIG. 1. Plot of power resonance (second harmonic) in He–Ne laser with CH_4 absorber at various methane pressures. Diameter of laser beam 4.5 mm, CH_4 pressure: a) 360 μTorr, b) μTorr.

of the resonance (Γ_{tr}). The decrease of the resonance width 2γ becomes noticeable at $\Gamma\tau < 0.5$, and the shape of the resonance differs significantly from Lorentzian. The half-width of the resonance at half height is now sufficiently well described by the expression $\gamma \approx (1.52/\tau)\sqrt{\Gamma\tau}$, from which we see that when $\Gamma\tau$ is decreased the width of the resonance becomes much smaller than the transit width due to the mean-thermal atoms. The difficulties in the experimental observation of the power-resonance narrowing due to slow atoms are caused by the very low intensity of the resonances in the transit region.

Under the conditions of the experiment described above, at a methane pressure 360 μTorr, the parameter $\Gamma\tau$ amounted to ≈ 0.15. At this value of the parameter $\Gamma\tau$, as follows from Fig. 2, the line half-width of each MHFS component is ≈ 22 kHz, which agrees with experiment. The impact half-width of the resonance is ≈ 5.4 kHz at 360 μTorr (the impact broadening of the methane absorption line is ≈ 30 MHz/Torr[9]), and the transit half-width is $\Gamma_{tr} \approx 20$ kHz. Consequently, the main contribution to the width of the observed resonance is made by the transit effects.

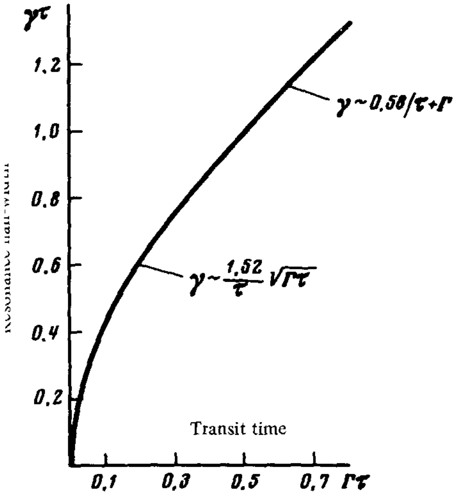

FIG. 2. Dependence of the half-width γ of the Lamb dip on the parameter $\Gamma\tau$.

At a methane pressure 60 μTorr, the half-width of the power resonance decreases to ≈ 900 Hz, and the broadening due to the finite interaction time of the mean-thermal molecules with the field remains the same (≈ 20 kHz). Under these conditions, a change in the resonance half-width by only ≈ 5 kHz should not lead to an appreciable change in its shape and to resolution of the hyperfine structure. Therefore the observed resonance-narrowing effect can be due only to the influence of the slow atoms. Indeed, at $P_{CH_2} = 60$ μTorr the parameter $\Gamma\tau$ decreases to $\approx 2.5 \times 10^{-2}$. The homogeneous line half-width of each individual MHFS component, corresponding to this value of $\Gamma\tau$, is $\gamma \approx 9$ kHz (see Fig. 2), which is much less than the transit half width $\Gamma_{tr} \approx 20$ kHz. For $\gamma \approx 9$ kHz, the hyperfine structure of the methane is resolved, as was indeed observed in experiment.

Thus, narrowing of the nonlinear resonances by slow atoms at low gas pressures greatly extends the capabilities of nonlinear spectroscopy without using telescopic systems.

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[1)] Second-harmonic signal, relative units.

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