Ultrahigh-resolution laser spectroscopy with cold particles

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The first results on the behavior of a nonlinear resonance in methane ($\lambda = 3.39$ μm) in the transit region as a function of the intensity of the saturating field are reported. The experimental results show that in weak fields the size of the derivative of the resonance, $\tilde{\gamma}$, is determined exclusively by the homogeneous half-width Γ ($\tilde{\gamma} = 1.4\ \Gamma$). The resonance forms primarily because of "cold" particles, with velocities well below the average thermal velocity. It thus becomes possible to distinguish transit broadening from the quadratic Doppler effect.

1. Producing ultranarrow nonlinear resonances with a relative width $\sim 10^{-13}$ and correspondingly increasing the resolution of optical spectroscopy require methods for increasing the duration of the interaction of the particles with the field. Cooling the particles makes this possible without substantially increasing the dimensions of the fields. An important aspect of the use of "cold" particles is the decrease in the influence of the quadratic Doppler effect, which broadens and shifts resonances. In the present letter we report the first spectroscopic study of saturated-absorption resonances by means of "cold" particles with an effective temperature $\sim 10^{-1}$ K, which is 10^3 times lower than the temperature of the working gas. The width of the nonlinear resonances is related to the effective selection of the cold particles, for which the interaction time is determined by the homogeneous width.

2. Under the condition $\Gamma \ll kv_0$ (where Γ is the collisional line half-width, k is the wave number, and v_0 is the average thermal velocity of the particles), the Lamb dip results from a saturation of the particles for which the velocity projection onto the field axis is $v_z \simeq 0$. The selection of the transverse velocity of the particles, v_r, occurs by virtue of saturation effects. In the transit region, where $\Gamma\tau_0 \ll 1$ (τ_0 is the transit time of a particle with the average thermal velocity across a field of radius a), we run into a new mechanism of inhomogeneous saturation. The saturation depends on $\tau = a/v_r$, the duration of the interaction of the particles with the field. For particles with a velocity of $v_r < a\Gamma$, the saturation parameter may be assumed to remain constant, independent of the velocity, at $\kappa = 4d^2E^2/\hbar^2\Gamma^2$, where d is the dipole matrix element of the transition, and $2E$ is the field amplitude. At velocities $v_r > a\Gamma$ the saturation parameter depends on the transverse velocity: $\kappa(v_r) = 4d^2E^2/\hbar^2)(a^2/v_r^2)$. It increases as this velocity decreases. The dip in a medium is a set of dips with various intensities and widths, in accordance with the velocity v_r. A fundamental point, which led us to carry out a detailed study, is that the slope of the derivative of the resonance with respect to the frequency is determined under the condition $\kappa \lesssim 1$ by the homogeneous width, while its maximum lies a distance $\tilde{\gamma} = 1.4\ \Gamma$ from the center of the line.[1]

Particles with velocities $v_r \sim a\Gamma$ make the major contribution here. The corresponding temperature of the particles is $T_{\rm eff} \sim (\Gamma\tau_0)^2 T_0$, where T_0 is the gas temperature.

If structures with a half-width $\sim\Gamma$ are to be produced under the condition $\Gamma\tau_0 \ll 1$, the saturation parameter must satisfy $\kappa \lesssim 1$. For a Gaussian beam, this requirement corresponds to a saturating power $P \lesssim (\Gamma\tau_0)^2 c(\hbar v_0/4d)^2$ of one traveling wave, where c is the velocity of light. An aspect of the saturation in the transit region which is of importance for an experiment is the sharp difference between the saturation for the interacting particles and that for the medium. The reason for this difference is that particles with all velocities contribute to the linear absorption coefficient, while the saturation resonance is determined by a small fraction $\sim(\Gamma\tau_0)^2$ of these particles. In the microwave range the saturation line is observed against the background of a linear lineshape with a width $\sim 1/\tau_0$, so that it is difficult to measure the narrow saturation line. In the optical range, the saturation line is observed in its "pure" form against the background of a linear lineshape with a Doppler width. The saturation in a medium is $\kappa_{\rm med} \sim (\Gamma\tau_0)^2\kappa = (4d^2E^2/\hbar^2)\tau_0^2$ [or, more precisely,[1] $\kappa_{\rm med} = \kappa(\Gamma\tau_0)^2 \ln(1/\Gamma\tau_0)$]. At a low saturation ($\kappa_{\rm med} \ll 1$) and a low absorption, the intensity of the resonance becomes very low. In the early work, the study of resonances was restricted to the region of relatively large saturation values[2] and to a qualitative observation of the contraction of the line.[3,4]

3. For a study of the resonance and its derivative, special spectrometers were developed on the basis of a He-Ne laser with output at $\lambda = 3.39$ μm with an internal methane absorption cell [$F_2^{(2)}$ line of the $P(7)$, ν_3 transition of methane]. These spectrometers are described in detail in Ref. 5. Experiments were carried out over a broad pressure range with weak fields inside the resonator.

Qualitatively new effect in these studies in the transit region are the field-induced broadening of the resonance and its derivative. Figure 1 shows the half-width $\tilde{\gamma}$ of the derivative of the resonance of one component of the magnetic hyperfine structure of

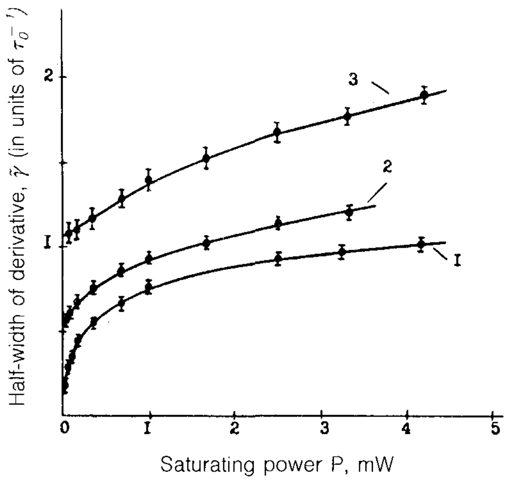

FIG. 1. Representative experimental curves of the half-width of the derivative of the resonance, $\tilde{\gamma}$ (in units of τ_0^{-1}), versus the saturating power P for one of the traveling waves ($a = 0.08$ cm, $\tau_0 = 1.45 \times 10^{-6}$ s). 1—$\Gamma\tau_0 = 0.13$; 2—$\Gamma\tau_0 = 0.54$; 3—$\Gamma\tau_0 = 1.3$.

the $F_2^{(2)}$ absorption line of methane versus the saturating power P for various values of $\Gamma\tau_0$. At $\Gamma\tau_0 > 1$, the curves have their usual shape, $\tilde{\gamma} \sim \tilde{\gamma}_0(1+\kappa)^{1/2}$, where γ_0 is the half-width in the limit $\kappa \to 0$. At $\Gamma\tau_0 \ll 1$, we can distinguish three characteristic regions on the curve of $\tilde{\gamma}(P)$. The linear initial region corresponds to a saturating power $P \lesssim 20\ \mu$W, with $\kappa \lesssim 1$. At a saturating power $P = 15\ \mu$W and $\Gamma\tau_0 = 0.1$ ($a = 0.08$ cm), we have $\tilde{\gamma} \approx 14$ kHz experimentally. The values of the widths found through a linear extrapolation of $\tilde{\gamma}(P)$ and $\gamma(P)$ to zero power were used to plot $\tilde{\gamma}_0$ and γ_0 versus the gas density. In the second region ($P \sim 100\ \mu$W) we have $\Gamma \ll dE/\hbar \ll 1/\tau_0$, and the role of the homogeneous linewidth is played by the Rabi frequency $dE/\hbar$. We thus have $\tilde{\gamma} \sim dE/\hbar$ and[6] $\gamma \sim 1/\tau_0[(dE/\hbar)\tau_0]^{1/2}$. In relatively strong fields ($P \sim 1$ mW), with $dE/\hbar \sim 1/\tau_0$ (the third region), the contribution of slow particles is suppressed by the pronounced saturation. Here the width of the resonance, determined primarily by the particles with the average thermal velocity, can be described as a function of the power by a square-root law:

$$\gamma \sim 1/\tau_0[1 + (4d^2E^2/\hbar^2)\tau_0^2]^{1/2} = 1/\tau_0[1 + (P/P_0)]^{1/2},$$

where $P_0 = c(\hbar v_0/4d)^2$ is the saturation parameter, expressed in milliwatts. The calculated value $P_0 \approx 1$ mW agrees with the value found in our experiments[2] and in Ref. 2. An extrapolation of γ from this region to zero power yields a value $\gamma \sim (1/\tau_0)^2$.

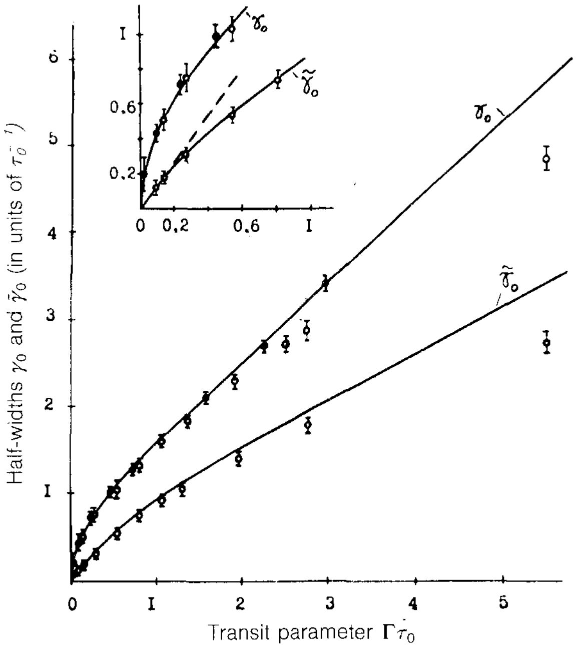

FIG. 2. Half-width of the resonance, γ_0, in methane (curve 1) and its derivative $\tilde{\gamma}_0$ (curve 2), in units of τ_0^{-1}, versus the pressure of the gas (CH_4, He), in units of $\Gamma\tau_0$. For CH_4, $\Gamma = 15$[kHz/mtorr]. P_{CH_4} [mtorr]; for He, $\Gamma = 8$[kHz/mtorr]$\cdot P_{He}$ [mtorr]. Filled circles—$a = 0.25$ cm, $\tau_0 = 4.5 \times 10^{-6}$ s; open circles—$a = 0.08$ cm, $\tau_0 = 1.45 \times 10^{-6}$ s. Solid curves—calculated from the tables on p. 19 of Ref. 5; dashed line—$\tilde{\gamma}\tau_0 = 1.4\Gamma\tau_0$.

Figure 2 shows the resulting curves of γ_0 and $\tilde{\gamma}_0$ versus the gas pressure. There is good agreement between the experimental data and the theoretical curves. There is some discrepancy at resonance widths above 500 kHz (see the data shown by the open circles at Fig. 2 for $\Gamma\tau_0 > 2$); we attribute it to a nonlinear dependence of Γ on the methane pressure.[7] In contrast with $\tilde{\gamma}_0$, the half-width γ_0 at $\Gamma\tau_0 \ll 1$ depends on the average thermal velocity. According to the theory of Refs. 1, 8, and 9, we would have $\gamma_0 \sim 1/\tau_0(\Gamma\tau_0)^{1/2}$, and this prediction is supported by the experiments. Here the width is determined primarily by particles which pass through the field without undergoing collisions, with a characteristic velocity $v \sim v_0(\Gamma\tau_0)^{1/2}$. The experimental results do not support the theoretical conclusions of[1)] Refs. 10–12 and do not agree with the experimental data of Refs. 12 and 13, where it was asserted that as the pressure goes to zero, $p \to 0$, the value of γ tends toward a constant value $\sim 1/\tau_0$, while we have $\tilde{\gamma} \sim (\Gamma\tau_0)^{1/2}$.

4. The minimum value of $\Gamma\tau_0$ at which we studied the shape of the resonance was $\Gamma\tau_0 \sim 2 \times 10^{-2}$; this value corresponds to $T_{\text{eff}} \sim 10^{-1}$ K. The quadratic Doppler shift of the resonance due to the cold particles is[6] $\Delta \sim (\Gamma\tau_0)^2\Delta_0$, where Δ_0 is the shift which corresponds to the gas temperature. A preliminary cooling of the gas to $T = 78$ K with $\Gamma\tau_0 = 10^{-2}$ makes it possible to reach effective temperatures $T_{\text{eff}} = 10^{-2}$ K. The corresponding quadratic Doppler shift is 10^{-2}–10^{-3} Hz. The primary difficulty in using this method for producing cold particles is the sharp decrease in the intensity of the resonance.[5] A telescopic beam expander, combined with other methods, would make it possible to increase the intensity of the resonance. In the visible region, the sensitivity of the detection of resonances in the absorption of cold particles could be raised by making use of fluorescence. It would thus become possible to immediately take up the development of a new generation of relatively simple lasers with a frequency reproducibility better than 10^{-16}.

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[1)]Dubetski[14] has shown that a serious physical error was made in Refs. 10–12 and affected all the results and conclusions there.

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