Optical spectroscopy free of quadratic Doppler effect

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The first direct observation of the influence of the quadratic Doppler *effect on the shape of the saturated-absorption line is reported. The attainment of narrowest optical resonances with a half-width* ~ 100 Hz *is also reported. The possibilities of an optical spectroscopy free of the quadratic Doppler effect are analyzed.*

1. The methods of nonlinear laser spectroscopy, as is well known, are capable of producing resonances with a uniform linewidth 2Γ and with maxima which correspond to a transition frequency ω_0. In the production of very narrow resonances with a relative width less than 10^{-11}, the quadratic Doppler effect becomes important. A moving particle with a transverse velocity v radiates (or absorbs) at a frequency which is shifted by an amount $\Delta = -1/2(v/c)^2\omega_0$ from the transition frequency. If $\Delta_0 \gg \Gamma$ (Δ_0 is the shift for a particle with an average thermal velocity v_0), the resonance in the saturated absorption consists of a set of resonances, whose frequency shifts depend on the velocity of the particle. Here we are encountering a new mechanism for inhomogeneous broadening of a resonance, whose width and shift will be on the order of Δ_0. The production of resonances with a homogeneous width 2Γ in this case requires eliminating the influence of the quadratic Doppler effect and the inhomogeneous broadening which it causes. There are two ways to do this. This first is to use deep radiative cooling of particles.[1] The second is to use optical selection of particles in a gas.[2] As was shown experimentally in Ref. 2, the width of the dispersive resonance under transit conditions ($\Gamma\tau_0 \ll 1$, where $\tau_0 = a/v_0$ is the time required for a particle with a velocity v_0 to traverse a beam of radius a) is determined by the homogeneous width 2Γ. Under these conditions, the resonance is dominated by "cold" particles with an effective temperature $T_{ef} = (\Gamma\tau_0)^2\ T_0$, where T_0 is the gas temperature. The shift of the resonance, δ, is correspondingly $\delta = (\Gamma\tau_0)^2\Delta_0$, and it becomes very small under the condition $\Gamma\tau_0 \ll 1$.

In this letter we are reporting the first direct observations of the influence of the quadratic Doppler effect on the shape of nonlinear resonances. We have achieved the narrowest optical resonances through the elimination of the influence of the quadratic Doppler effect. The experiments were carried out with a specially developed laser spectrometer, designed for observing resonances with a relative width of 10^{-13}–10^{-14}.

2. The production of resonances in the case $\Gamma \ll \Delta_0$ is possible only at very low pressures of the absorbing gas, $< 10^{-6}$ torr. Because of the low density of particles, the velocity selection of particles, and the low saturation, the intensity of the resonance decreases sharply. In an effort to overcome this difficulty, the laser spectrometer has a telescopic beam expanded inside the resonator. This approach not only achieves an effective selection of particles but also raises the intensity of the resonances.

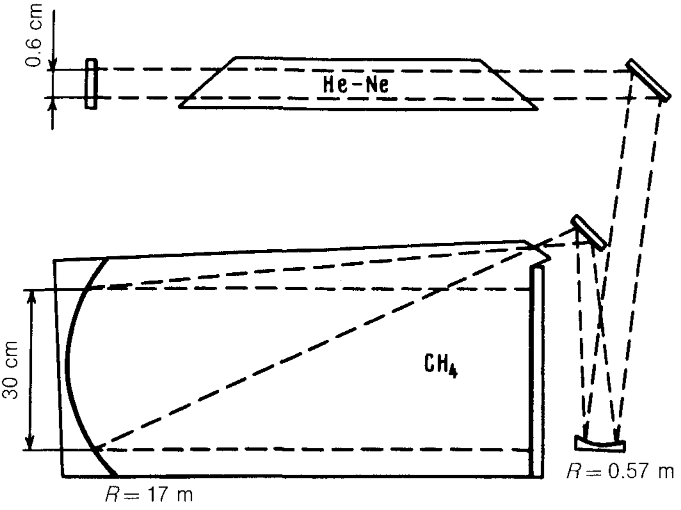

FIG. 1. Layout of the He–Ne/CH_4 laser with an internal telescopic beam expander.

The first experiments were carried out with a He–Ne laser with a wavelength $\lambda = 3.39\ \mu m$. The absorption resonances were observed at the individual components of the magnetic hyperfine structure of the $F_2^{(2)}$ line of methane. The laser resonator was formed by a system of six mirrors (Fig. 1). The beam diameter in the cell was 30 cm; the length of the cell was 800 cm. The spectrometer included a heterodyne laser and a stable-frequency laser. The use of phase matching of the laser frequencies resulted in a narrow output line in the test laser, ~ 1 Hz, and a long-term frequency stability

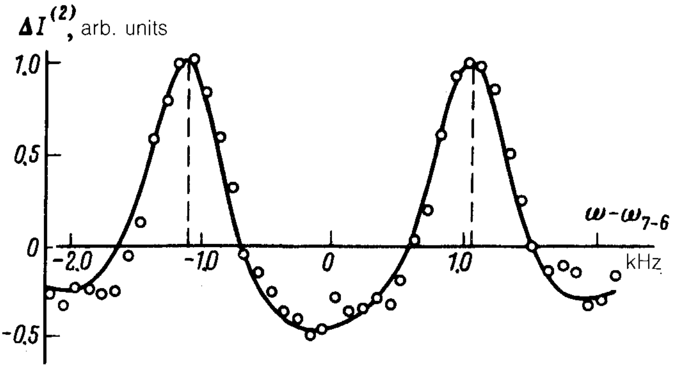

FIG. 2. Recording of the recoil doublet of the 7–6 component of the hyperfine structure of the $F_2^{(2)}$ line of the $P(7)$ transition of the $v(3)$ band of methane at a pressure of 10^{-5} torr and a gas temperature $T_0 = 300$ K. The points are experimental data for the signal representing the second harmonic in the laser output power. The modulation frequency was 230 Hz, the amplitude of the deviation was 200 Hz, and the averaging time was 4 s. The solid line was calculated from expression (1).

$\sim 10^{-14}$. The resonance was detected on the basis of the second-harmonic signal in the optical power during frequency modulation of the laser output. Figure 2 shows a signal recorded at a methane pressure $\approx 10^{-5}$ torr. Shown in the same figure is the theoretical shape of the signal representing the second derivative of the resonance under transit conditions; the quadratic Doppler effect has been taken into account here.[3] We see a satisfactory agreement between theory and experiment. The quadratic Doppler effect is manifested in the onset of an asymmetry of the wings of the resonances, which is particularly accentuated by the presence of a recoil doublet. At resonance widths on the order of 1 kHz the asymmetry is manifested in a difference in the intensities of the doublet components, which is a consequence of the different lifetimes of the levels.[4,5] The significant difference, $\sim 14\%$, between the component intensities which Alekseev *et al.*[6] observed and attributed to the quadratic Doppler effect is not confirmed by the results of the theoretical analysis, by the experimental results of Refs. 4 and 5, or by the results of the present study.

The line with a half-width ~ 100 Hz which was produced in the first experiments is the narrowest in the IR and optical ranges. The shift of the resonance due to the quadratic Doppler effect here was ~ 1 Hz. Achieving smaller widths was prevented by the noise due to acoustic and mechanical factors, which caused an angular detuning of the resonator and a noise modulation of the laser output. The amplitude noise in the output exceeded the receiver noise by a factor of more than 100. Removing this noise through automatic adjustment of mirrors will make it possible to detect resonances with a width ~ 10 Hz.

3. The effect of the quadratic Doppler effect on the shape of a resonance under transient conditions in the case $\Gamma \gg \Delta_0$ was studied theoretically in Ref. 7. The results of that study can be extended to the case $\Gamma \ll \Delta_0$. In general, the shape of a resonance with a quadratic Doppler effect is described by a very complicated expression. We will accordingly restrict the discussion here to the simple approximation

$$\alpha_s = \alpha_0 \left[1 - \kappa (\Gamma\tau_0)^2 \Gamma^2 /4 \int_0^\infty \frac{W(v)\, dv}{[\Omega + (v/c)^2 \omega /2]^2 + (\Gamma + v/a)^2}\right], \tag{1}$$

where $W(v)$ is a Maxwellian transverse-velocity distribution of the particles, α_0 and α_s are the unsaturated and saturated absorption coefficients, $\kappa = [2dE/\hbar\Gamma]^2$ is the saturation parameter for cold particles, $2E$ is the field amplitude, and d is the dipole-moment matrix element of the working transition. The physical meaning of expression (1) is obvious: The shape of the resonance can be represented as a set of resonances, each lying at a frequency $\omega = \omega_0 - (v/c)^2\omega_0/2$ and having a half-width $\gamma = \Gamma + v/a$. Analysis shows that expression (1) describes well qualitatively and quantitatively the shape of the resonance in the transit region.

Figure 3 shows the results of a numerical simulation based on (1) without consideration of the radiative width, for the actual operating conditions of the apparatus described above, for an absorbing-gas pressure of 2×10^{-7} torr and a gas temperature $T_0 = 77$ K. Curves 1 and 2 show the shape of the resonance in methane in the homogeneous case ($\tau_0 \to \infty$) and for $\Gamma\tau_0 = 10^{-2}$, respectively. Curve 3 shows the shape of the resonance of the second derivative of the absorption line. Its half-width is 4 Hz (and

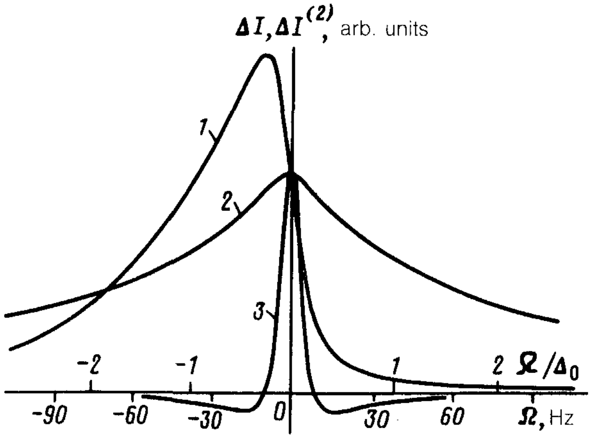

FIG. 3. Shape of the resonances in methane for an individual component of the recoil doublet for $\Delta_0 = 40$ Hz. The curves are normalized to the value of the signal at $\Omega = 0$.

its relative half-width 4×10^{-14}). The effective temperature of the cold molecules, which determines the shape of the resonance, is $T_{ef} = 0.01$ K, and the shift due to the quadratic Doppler effect is $\delta = 0.006$ Hz.

We believe that further progress will result from the use of forbidden transitions between ground and metastable states of inert gases. Here there is the hope of achieving resonances with relative widths of 10^{-15}. The primary difficulty will be in developing tunable light sources with very narrow lines in the UV part of the spectrum.

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