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Second-Order Doppler-Free Spectroscopy

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Abstract. The observation of second-order Doppler-free optical resonances with a width of 50 Hz are reported for the first time. It was achieved due to the use of optical selection of cold particles from an absorbing gas. The experiments have been carried out by using a new laser spectrometer, supposed to obtain the saturated absorption resonances with a relative width 10^{-13}–10^{-14}. The results of experimental and theoretical studies of second-order Doppler-free effect influence on the shape of nonlinear optical resonances in transit-time conditions are considered.

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Recent years witnessed the development of methods of nonlinear laser spectroscopy (saturated absorption, two-photon spectroscopy, method of separated fields and other), which allow to eliminate the linear Doppler effect and to obtain a resonance with the homogeneous half-width of about 1 kHz (the relative width is 10^{-11}–10^{-12}). These methods underlay the Doppler-free super-high-resolution spectroscopy. When obtaining very narrow lines in a gas with the relative width of 10^{-13}–10^{-14}, we deal with a new mechanism of inhomogeneous broadening and shift of optical resonances due to the Second-Order Doppler effect (SODE). A particle moving with the velocity u radiates (absorbs) a line with the homogeneous halfwidth Γ shifted by the value $\Delta = -(1/2)(u/c)^2\omega_0$ with respect to the transition frequency ω_0. If $\Delta_0 \gg \Gamma$ (Δ_0 is the SODE shift for a particle with the thermal velocity v_0), then an inhomogeneously broadened line in a gas has a width and a shift of Δ_0.

Therefore, obtaining of narrow lines with a homogeneous width and without shift is possible only by eliminating the SODE influence. At least, there are two ways to eliminate the SODE influence. The first one is connected with a deep radiative cooling of particles [1]. The second one is in the optical selection of cold particles in a gas by means of the inhomogeneous saturation in a standing light wave [2]. Under transit-time conditions ($\Gamma\tau_0 \ll 1$, $\tau_0 = a/v_0$ is the transit-time for a particle with the thermal velocity v_0 crossing the light beam of the radius a) the width of a dispersion resonance is experimentally shown to be determined by the homogeneous width of 2Γ in [2]. In this case, the main contribution to the saturation resonance is determined by slow particles with the effective temperature $T_{\text{eff}} \sim (\Gamma\tau_0)^2 T_0$, with T_0 being the gas temperature. Therefore, the SODE shift of a resonance is small $\delta \approx (\Gamma\tau_0)^2\Delta_0$ at $\Gamma\tau_0 \ll 1$. So, the saturation lineshape approaches the one for a single stationary particle.

In this paper we report on the results of the direct observation of the SODE influence on a spectral line shape in a gas and of the obtaining a super-narrow optical resonance with the halfwidth of 50 Hz, when eliminating the SODE influence [3]. The experiments have been carried out using the specially developed laser spectrometer supposed to observe a resonance with the relative width of 10^{-13}–10^{-14}. Possibilities of the second-order Doppler-free optical spectroscopy have been analysed.

1. The Influence of The Second-Order Doppler Effect on a Nonlinear Resonance

Under transit-time conditions the influence of SODE on the resonance shape has been studied for the case $\Gamma \gg \Delta_0$ [4, 5]. Main attention has been paid to the resonance shift caused by SODE. Results [4, 5] can also be applied to the case $\Gamma < \Delta_0$. The exact expression for the resonance shape under the transit-time conditions with allowance for SODE in a Gaussian field had a complicated form [3]:

$$\alpha(\Omega) = \frac{\alpha_0 \kappa \beta^2}{4} \iint_0^\infty \times \frac{d\xi d\eta e^{-\xi-\eta}\left(A\cos\frac{\Omega}{\Gamma}\xi - \frac{\Delta}{\Gamma}\beta^2 \sin\frac{\Omega}{\Gamma}\xi\right)}{A^2 + \left(\frac{\Delta}{\Gamma}\beta^2\xi\right)^2} \tag{1}$$

with $A^2=(\xi/2)^2+(\xi/2+\eta)^2+\beta^2$, $\beta=\Gamma\tau_0$, $\Omega=\omega-\omega_0$.

We shall use the simple approximation:

$$\alpha(\Omega)=\frac{\alpha_0\kappa\beta^2}{4}\Gamma^2\int_0^\infty\frac{W(u)du}{\left[\Omega+\frac{1}{2}\left(\frac{u}{c}\right)^2\omega_0\right]^2+\gamma^2}, \quad (2)$$

where $W(u)=(u/v_0^2)\exp(-u^2/v_0^2)$ is the Maxwellian distribution of particles over the transverse velocities u, $\kappa=(2dE/\hbar\Gamma)^2$ is the saturation parameter, $2E$ is the field amplitude, d is the dipole matrix element of the absorbing transition, $\gamma=\Gamma+u/a$, α_0 is the unsaturated absorption coefficient. The physical sense of (2) is obvious, i.e. the resulting resonance shape consists of a combination of separate resonances located at the frequencies $\omega=\omega_0-(1/2)(u/c)^2\omega_0$ and having the halfwidths of $\gamma=\Gamma+u/a$. Both ω and γ depend on the velocity u. Additional computer analysis of (2) in comparison with (1) shows that the resonance shape under transit-time conditions with $\Gamma<\Delta_0$ is well enough described by (2). The results of the resonance-shape calculations are shown by Fig. 1a and b. Let us begin the SODE analysis for the homogeneous case ($\Gamma\gg\tau_0^{-1}$) when the Lamb-dip width is the same for all particles ($\gamma=\Gamma$). The resonances for $\Gamma/\Delta_0=0.025$ and $\Gamma/\Delta_0=0.1$ are shown by the curves 1 and 2, respectively, in Fig. 1a. One can see here that the resonance shape is strongly asymmetrical. It consists of two characteristic parts. The first one has the homogeneous halfwidth Γ with the centre near $\Omega=0$. The second (low-frequency) part of the resonance ($|\Omega|>\Gamma$) is determined by its inhomogeneous broadening and shift caused by SODE and has the width of about Δ_0.

In a Gaussian light beam when transit effects are considerable ($\Gamma<\tau_0^{-1}$) the widths of resonances for particles with different velocities u are also different ($\gamma=\Gamma+u/a$). Consequently, the resulting resonance shape is altered greatly (curve 3). The low-frequency part of a resonance corresponds to fast particles. The velocities of the particles interacting resonantly with the field are determined by $\Omega=-(1/2)(u/c)^2\omega_0$. So, for $|\Omega|>\Gamma$ the halfwidth of each separate resonance increases greatly due to the transit-broadening contribution $\tau^{-1}=u/a$, at the same time the intensity is decreased. Hence, the role of the SODE inhomogeneous broadening decreases. This mechanism of decreasing the SODE influence in a transit region is enhanced due to effective selection of cold particles in the gas.

Figure 1b shows the resonance shape of a second derivative of the saturated absorption line at $\Gamma\tau_0=0.1$, and $\Gamma/\Delta_0=0.1$. As it has already been noted in [2] that this resonance width is determined by the homogeneous width, because the main contribution in it is made by slow particles with the velocities $u<a$. Due to that, the SODE influence on the resonance broadening and shift can be effectively eliminated for $\Gamma\ll\Delta_0$. Its width is 0.6Γ and the maximum shift is of $\delta\sim10^{-2}\Delta_0$.

Thus, the absorption-saturation method permits one to eliminate the influence of the linear Doppler effect and to drastically reduce the influence of the second-order Doppler effect.

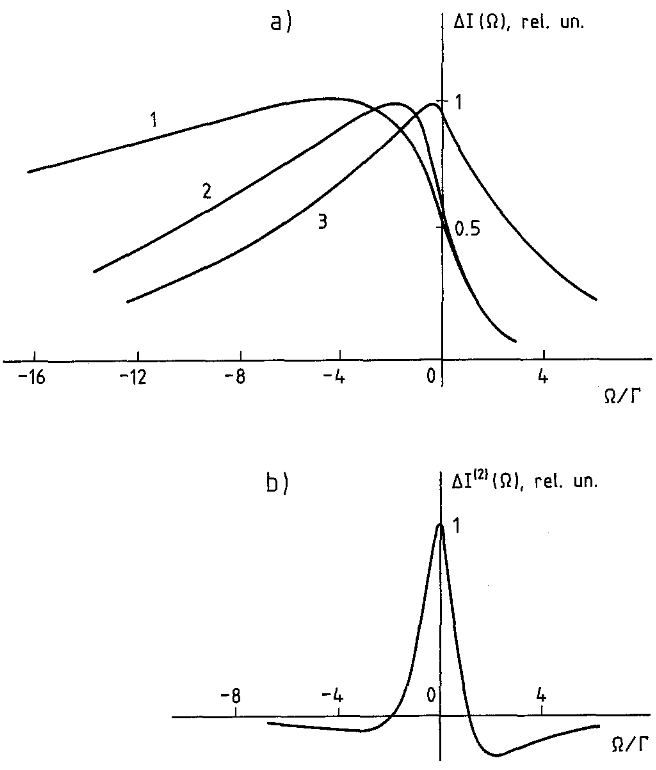

Fig. 1a, b. Calculated shape of the saturated absorption resonance in methane taking into account the influence of the second-order Doppler effect according to (2): **a** resonance shape for $\Gamma/\Delta_0=0.025$, $\Gamma\gg\tau_0^{-1}$ (curve *1*), $\Gamma/\Delta_0=0.1$, $\Gamma\gg\tau_0^{-1}$ (curve 2), and $\Gamma/\Delta_0=0.1$, $\Gamma\tau_0=0.1$ (curve *3*); **b** second derivative signal with $\Gamma\tau_0=0.1$, $\Gamma/\Delta_0=0.1$

2. Experiment and Results

To obtain the resonance with the halfwidth of $\Gamma\ll\Delta_0$, it is necessary to use an absorbing gas at the very low pressure of 10^{-6}–10^{-7} Torr. A small fraction of the cold particles in a gas and a low saturation power lead to the rapid decrease of the resonance intensity. To overcome this difficulty, a special laser spectrometer based on the use of a laser with a telescopic beam expander (TBE) inside the cavity has been developed. This allowed one to record the resonance at a pressure of 10^{-6} Torr and lower, and to make an effective selection of cold particles. The first experiments have been carried out in methane on $F^{(2)}P(7)\nu_3$-absorption line ($\lambda=3.39$ μm). A scheme of the new spectrometer is shown by Fig. 2. It includes three lasers, i.e. the frequency stable He–Ne/CH_4 laser 1 with the narrow radiation line width of about 1 Hz, the auxiliary He–Ne laser 2, and the investigated He–Ne/CH_4 laser 3 with TBE.

The 8 m cavity of the stable laser 1 is formed by the flat and spherical ($R=70$ m) mirrors. The absorption cell length is 4.5 m. The light-beam diameter in the cavity is about 1 cm, that made it possible to observe the intensive (1–2 mW) CH_4-resonances with the halfwidth of about 30 kHz. Using of these resonances, a radiation linewidth narrower than 1 Hz and the long-term stability of about 10^{-14} were obtained. The He–Ne-laser 2 with 10 mW power was used as an optical heterodyne. The cavity of the

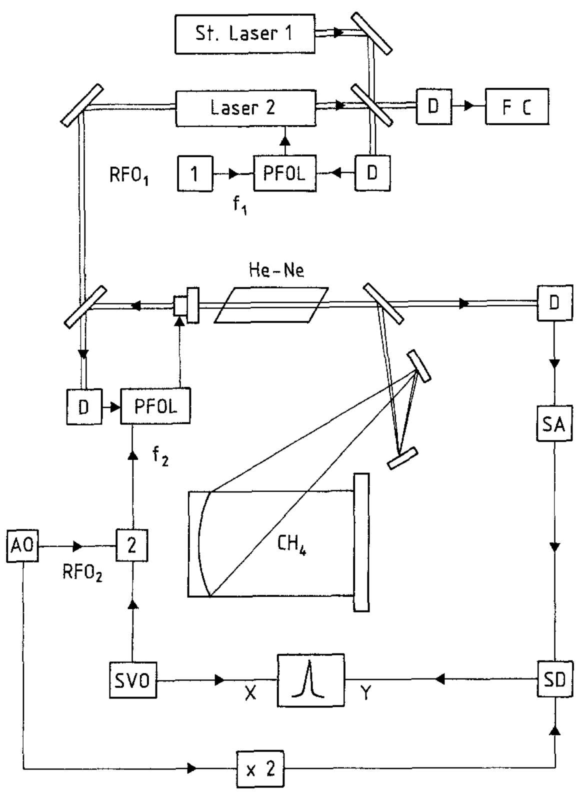

Fig. 2. Schematic representation of a new laser spectrometer at $\lambda = 3.39\,\mu m$ (D: photodetector; PFOL: electron system of phase frequency offset-lock; SA: selective amplifier; SD: synchronous detector; AO: audio-oscillator; SVO: sawtooth voltage oscillator; F.C.: frequency control, RFO: radio-frequency oscillator)

investigated laser 3 was formed by the system of six mirrors (Fig. 2). Such a cavity geometry practically allowed us to eliminate the influence of astigmatism on the shape of a light-beam in the cell. The absorption-cell length and the light-beam diameter in it were 850 cm and 30 cm, respectively. The absorption cell was protected from the magnetic field by a multilayer permalloy screen, and operated at $T_0 = 300$ K and $T_0 = 77$ K gas temperature. A special system for gas cooling by liquid nitrogen was put inside the cell. The cell was placed in a solenoid to investigate the influence of a weak magnetic field on the narrow resonance shape. All lasers are mounted in the same frame. The installation was placed on a 20-ton concrete plate lying on air dampers.

The laser 2 was phase locked to the stable laser 1. The frequency difference of these lasers was equal to the frequency $f_1 = 1$ MHz of the reference Radio-Frequency Oscillator (RFO$_1$) ($\nu_2 - \nu_1 = f_1$) with the frequency stability of 10^{-7}. In this case, the radiation linewidth and the long-term frequency stability of the laser 2 become the same as those of the laser 1. The frequency of the investigated laser 3 with TBE was locked to that of the laser 2 by the phase-frequency offset-lock system with the shift $-f_1(f_2 \approx f_1)$ determined by the Radio-Frequency Oscillator (RFO$_2$) frequency ($\nu_2 - \nu_3 = f_2$). Hence, a narrow radiation line and a high long-term frequency stability of the telescopic laser 3 have been provided.

The resonance in methane was registered by the $X-Y$ recorder using the method of a synchronous detection of the second harmonic signal in the radiation power of the laser 3 with the use of the laser frequency modulation. The

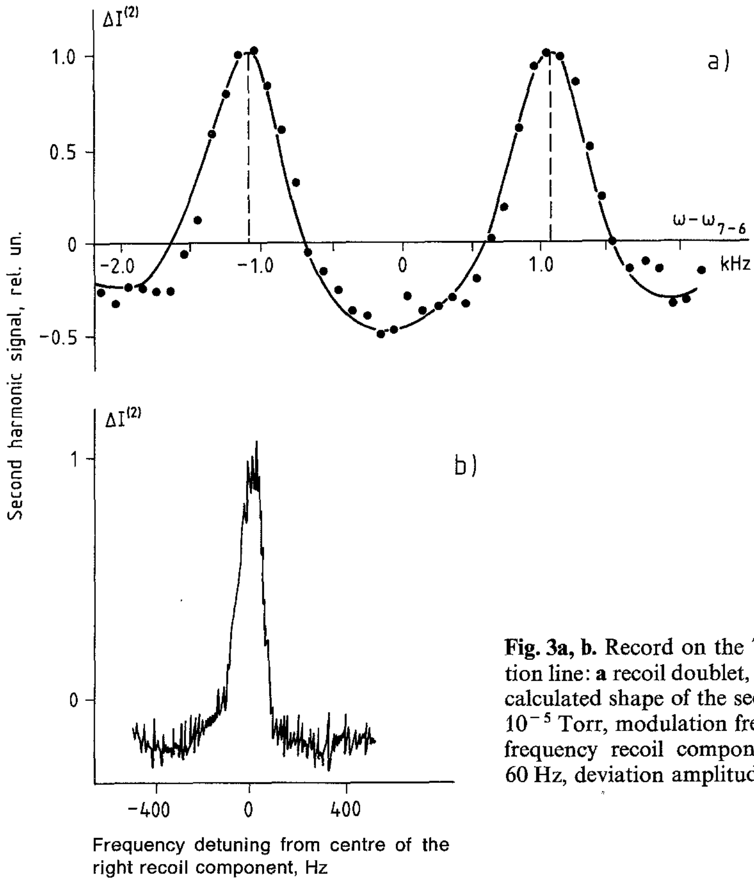

Fig. 3a, b. Record on the 7→6 component of MHFS of the $F_2^{(2)}P(7)\nu_3$-methane absorption line: **a** recoil doublet, points represent experimental data, the continuous curve is the calculated shape of the second derivative of CH$_4$-resonance using (2), methane pressure 10^{-5} Torr, modulation frequency $f = 230$ Hz, deviation amplitude $f = 200$ Hz; **b** higher frequency recoil component methane pressure 5×10^{-6} Torr, modulation frequency 60 Hz, deviation amplitude 100 Hz, record time 20 min

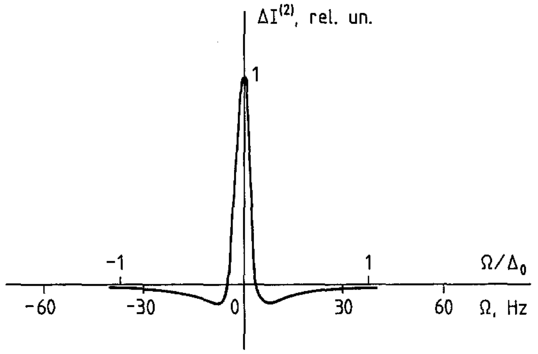

Fig. 4. Resonance shape in methane (second derivative) for one recoil component: pressure 2×10^{-7} Torr, $a = 15$ cm, $T_0 = 77$ K

telescopic laser frequency was tuned near the methane absorption line centre by a sawtooth voltage oscillator operating with the RFO_2 frequency. The frequency differences $(\nu_3 - \nu_2)$ and $(\nu_2 - \nu_1)$ were measured simultaneously during the resonance recording.

First measurements were made at room temperature of an absorbing gas $T_0 = 300$ K. We have observed CH_4 resonances at $7 \rightarrow 6$ component of the Magnetic Hyperfine Structure (MHFS) of the $F_2^{(2)}$ line. For the small transit parameter $\Gamma\tau_0 < 10^{-1}$ we had to operate with a saturating power less than 10 μW.

Figure 3a shows the record of the recoil doublet in methane. The experimental data are marked with dots. The continuous line is the shape of the second resonance derivative with allowance for the second-order Doppler effect, calculated by use of (2). The methane pressure in a cell was 10 μTorr. The resonance halfwidth was 280 Hz. Here is a good agreement between the theory and the experiment. The asymmetry of the recoil doublet wings near the centre is due to SODE. Note, if the resonance width is of the order of 1 kHz, the asymmetry is observed due to an amplitude difference of the recoil components, that may be the result of different lifetimes of the levels [6, 7]. The large difference of component intensities (14%) observed in [8] and interpreted as SODE is not confirmed by the theoretical analysis and experiments [6, 7] and by the present work. Figure 3b shows the record of a single recoil-doublet component for the $7 \rightarrow 6$ transition at the pressure 5×10^{-6} Torr. The resonance halfwidth is about 50 Hz. The shift of resonance maximum with respect to the centre of this recoil component is less than 1 Hz due to SODE. The resonance halfwidth 50 Hz obtained in our experiments is the narrowest in the optical region.

Due to acoustical and mechanical noise we could not observe narrower resonances. Noise resulted in an angular detuning of the cavity and, correspondingly, in the noise modulation of the radiation power. The amplitude of this fluctuations was higher than the photodetector noise. An elimination of this amplitude noise by an automatic system of angular tuning of the cavity mirrors enabled us to operate at the methane pressure of 10^{-6}–10^{-7} Torr and to obtain resonances with a width narrower than 10 Hz.

Using (2) and neglecting the radiation width we calculated the resonance shape (second derivative) which corresponds to parameters of our set-up with the methane pressure of 2×10^{-7} Torr and a gas temperature $T = 77$ K (Fig. 4). The resonance in the second frequency derivative of the saturated absorption line is shown by Fig. 4. Its width is about of 4 Hz (the relative width is 4×10^{-14}). The effective temperature of cold methane molecules giving rise to the resonance is $T_{eff} = 8$ mK. The respective second-order Doppler shift of the resonance is $\delta \approx 6$ mHz.

A resonance with the relative width of 10^{-13}–10^{-14} gives new possibilities for making precision physical experiments and suggesting a new generation of laser frequency standards based on cold particles and with a reproducibility of 10^{-16}.

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