Intradoppler spectroscopy of a three-level gas in the field of a frequency modulated traveling wave

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(Submitted 26 October 1983)
Pis'ma Zh. Eksp. Teor. Fiz. **38**, No. 11, 532–536 (10 December 1983)

A new method of frequency-modulation spectroscopy of a three-level gas, whose resolution is virtually not restricted by transit effects and by the technical fluctuations of the radiation frequency, is proposed and realized experimentally. Using a medium with absorption of 3×10^{-4} cm^{-1} and a length of 15 cm, a resolution of 3×10^{9} was achieved.

PACS numbers: 07.65.Eh, 51.70. + f

1. A new high-sensitivity method of laser spectroscopy—frequency modulation saturation absorption spectroscopy—was demonstrated in Refs. 1–3. The method consists of recording the amplitude modulation signal, arising as a result of the interaction of the frequency-modulated radiation with a two-level gas, in which a countermoving saturating beam propagates. Since the modulation frequency can be quite high (up to the microwave range), the minimum recordable signal is determined only by photon noise. However, the resolution of the method,[1–3] as also the resolution of traditional methods of nonlinear laser spectroscopy, is limited ultimately by transit broadening of the investigated line and broadening of the laser radiation spectrum, associated primarily with technical fluctuations of its frequency.

In this work we proposed and experimentally realized a new method of frequency-modulation spectroscopy of a three-level gas, whose resolution is essentially not limited by transit broadening and technical fluctuations of the radiation frequency.

We shall examine the interaction of a frequency-modulated traveling wave $\epsilon(z,t)=\epsilon_0\exp\times[i2\pi(t-z/c)\nu-i\Delta\cos 2\pi ft]$ + c.c. with a three-level system and, in addition, we shall assume that the frequency spacing ν_{12} between levels 1 and 2 is much smaller than the Doppler width of the allowed optical transitions 1–3 and 2–3. For simplicity, we shall restrict ourselves to the case $\Delta \ll 1$. The field $\epsilon(z,t)$ can then be assumed to be a superposition of three traveling waves: a strongly saturating wave with amplitude ϵ_0 and frequency ν and two weak probing waves with frequencies $\nu\pm f$ with amplitudes $\epsilon_+=\epsilon_-=-i\frac{\Delta}{2}\epsilon_0$. The variable part of the wave power P, on passing through a weakly absorbing medium with length L, is

$$\Delta P/P=\delta_{\rm disp}\cos 2\pi ft+\delta_{\rm abs}\sin 2\pi ft, \qquad (1)$$

where $\delta_{\rm disp}=4\pi^2(\nu/c)L\Delta(\kappa'_+ +\kappa'_- -2\kappa'_0)$, $\delta_{\rm abs}=4\pi^2(\nu/c)L\Delta(\kappa''_+ -\kappa''_-)$, $\kappa_\pm=\kappa'_\pm+i\kappa''_\pm$ is the polarizability of the medium for the weak wave $\epsilon_\pm$ in the presence of the strong ϵ_0 and κ_0 is the real part of the polarizability of the medium in the field ϵ_0. A calculation of κ with the help of the well-known technique for solving the equations for the density matrix in the field of three waves ϵ_0 and $\epsilon_\pm$ (see, for example, Ref. 4) shows that with unequal dipole moments of the transitions 1–3 and 2–3 $d_{13}^2 \neq d_{23}^2$, the quantities κ_+ and κ_- are different and, therefore, in passing through the medium, the frequency-modulated wave becomes an amplitude-modulated wave ($\delta_{abs}, \delta_{disp} \neq 0$). Without going into details of the analysis, which will be presented in a more detailed publication, we shall only give the final result in the two most interesting cases. For vibrational-rotational transitions in low-pressure molecular gases, when the relaxation of the density matrix may be described sufficiently accurately with a single relaxation constant γ, we have in the Doppler limit and with $\gamma \ll \omega_{12}$:

$$\delta_{abs} + i\delta_{disp} = \alpha_0 L \frac{\Delta}{2}\left\{L_{12}\left(\frac{f-\nu_{12}}{\gamma}\right) - L_{21}\left(\frac{f-\nu_{12}}{\gamma}\right)\right\}, \tag{2}$$

where

$$L_{lj}(x) = a_l\left(1 - \frac{1}{\sqrt{1+a_j I}}\right) \Big/ \left(1 + i\frac{2x}{1+\sqrt{1+a_j I}}\right), \quad a_l = \frac{d_{l3}^2}{d_{13}^2 + d_{23}^2},$$

$$l, j = 1, 2;$$

α_0 is the unsaturated absorption coefficient, $I = I_1 + I_2$, $I_j = d_{j3}^2 |\epsilon_0|^2/\hbar^2\gamma^2$ is the saturation parameter of the transition j-3. In the case of electronic transitions from the ground or metastable level to the upper short-lived level, for $\delta_{abs} + i\delta_{disp}$ we have (2), where γ is the relaxation constant of the nondiagonal element ρ_{12} of the density matrix.

As is evident from (2), the amplitude modulation signal varies in a resonant manner upon variation of f for ν_{12}. The antiphase component δ_{abs} represents the difference between two field-broadened Lorentzian contours with different amplitudes. The resulting contour with weak absorption has a nearly Lorentzian shape with a peak at $f = \nu_{12}$ and width $\sim\gamma$. Analogously, δ_{disp} has a nearly dispersion shape with a zero at $f = \nu_{12}$ and width $\sim\gamma$. We note that with the help of synchronous detection, it is possible to record δ_{disp} and δ_{abs} separately.

Expression (2) was obtained for the case of a monochromatic plane wave. Inclusion of drift effects and fluctuations of the carrier frequency ν requires a special analysis, which will also be performed in a future publication. Here, however, we note that as is evident from (2), the amplitude modulation signal with weak saturation $I \ll 1$ arises only in second order with respect to the intensity of the saturating field. This indicates that the transit effects do not lead to broadening of the resonance (2), since due to the anomalous contribution of slow molecules to the polarization of the medium, the nonlinear second- and higher-order resonances are not subject to transit broadening.[5] The effect of the fluctuations of the laser radiation frequency on the width of the resonance (2) is likewise greatly weakened, since the frequency ν does not enter into (2). The contribution of fluctuations of the freuqncy ν to the width of the resonance (2) depends on the shape of their spectra. Thus, for example, slow technical fluctuations under the condition $\gamma\tau \gg 1$, where τ is the correlation time of the frequency fluctuations, have virtually no effect on the width of the resonance (2).

2. To realize the experimental method, we used the Zeeman effect in the $F_2^{(2)}$ component of the vibrational-rotational transition of the methane line ($\lambda = 3.39\ \mu$m). In the presence of a magnetic field, the transition J_1-J_2 represents a superposition of a series of three-level systems: for example, two magnetic sublevels $m + 1$ and $m - 1$ of the lower state J_1 and the magnetic sublevel m of the upper state J_2, etc. The difference between the dipole moments of the transitions $\Delta m = +1$ and $\Delta m = -1$, which is required for the existence of the signal in the frequency-modulation spectroscopy, can be obtained by using elliptically polarized laser beam. Solving the system of equations for the density matrix of molecules in the field of the plane waves ϵ_0 and $\epsilon_\pm$ resonant with the transition J_1-J_2, in the presence of an axial magnetic field in fifth order with respect to the field ϵ_0 and first order with respect to $\epsilon_\pm$, the signal $\delta_{abs} + i\delta_{disp}$ with $J_1, J_2 \gg 1$ can be represented sufficiently accurately in the form

$$\delta_{abs} + i\delta_{disp} \propto \alpha_0 L \cdot \Delta \cdot I^2 \frac{\epsilon(1-\epsilon^2)^2}{(1+\epsilon^2)^3}$$

$$\times \left[\frac{2}{1 + i\dfrac{2\Omega - f}{2\gamma}} + \frac{1}{\left(1 + i\dfrac{2\Omega - f}{2\gamma}\right)^2} - \frac{2}{1 - i\dfrac{2\Omega + f}{2\gamma}} - \frac{1}{\left(1 + i\dfrac{2\Omega + f}{2\gamma}\right)^2} \right], \tag{3}$$

where ϵ is the ratio of the principal axes of the polarization ellipse of the laser field, and Ω is the Zeeman splitting.

The experimental arrangement is shown in Fig. 1. A single-mode He-Ne laser 1 with output power of 250 μW was used. The radius of the beam at the neck of the caustic was 0.1 cm. The frequency of the laser radiation was modulated by applying a sine-wave voltage with frequency $f = 200$ kHz to the piezoceramic, to which one of the cavity mirrors is attached. To eliminate the parasitic signal of amplitude modulation resulting from the difference between the frequency of the laser radiation and the top of the gain line, the laser frequency 1 was fixed with the help of the automatic-frequen-

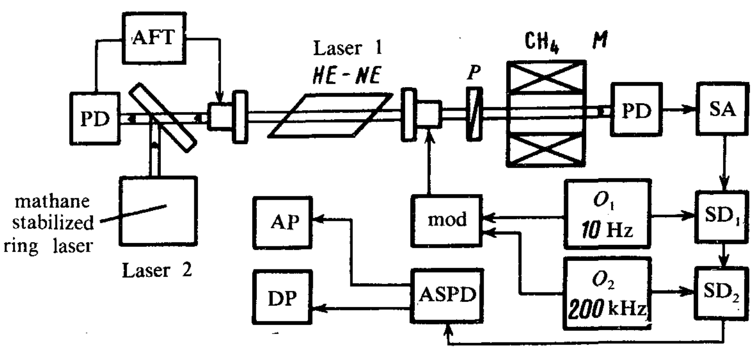

Fig. 1. Diagram of the experimental setup. AFT, automatic frequency tuning; PD, photodetector; P, quarter-wave plate; M, solenoid; CH_4 methane cell; SA, selective amplifier; SD synchronous detector; O, audio oscillator; ASPD, automatic system for storing and processing data; AP, automatic plotter; DP, digital printing setup; MOD, modulator.

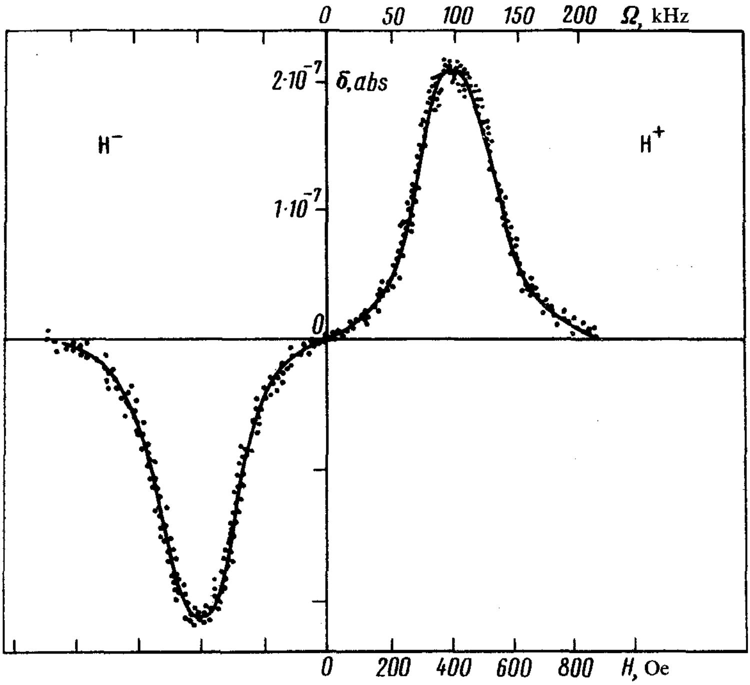

Fig. 2. Amplitude modulation signal δ_{abs} as a function of the magnetic field. The orientation of the magnetic field reversed when the level $H=0$ was crossed. The solid curve represents the dependence constructed using Eq. (3) with $\gamma = 41$ kHz.

cy-tuning system to the frequency of the ring laser 2 stabilized on the methane line, described in Ref. 6. The required ellipticity of the radiation was fixed by appropriately positioning a quarter-wave plate. The methane absorbing cell with a length of 15 cm was placed in an axial magnetic field. The signal from the output of the photodetector, after synchronous detection, entered the automatic system for storing and processing data. To increase the signal-to-noise ratio, the high-frequency modulation was interrupted with a 10-Hz frequency followed by synchronous detection.

Figure 2 shows a typical amplitude modulation signal δ_{abs} as a function of the intensity of the magnetic field. Each point was obtained by averaging 100 measurements. The methane pressure was $P_{CH_4} = 2.8$ Torr and the ellipticity of the beam was $\epsilon = 0.3$. The minimum recordable amplitude of the resonances (with a signal-to-noise ratio equal to 2) was 5×10^{-8}. This amplitude was determined by the residual amplitude modulation of the output radiation. Resonances with amplitude exceeding 5×10^{-8} were observed in the range of methane pressures from 1.7 to 3.3 mTorr; the maximum amplitude 3×10^{-7} was achieved at 2.5 mTorr. The decrease in the amplitude with $P_{CH_4} > 2.5$ mTorr is due to broadening of the line and decrease in the saturation parameter $I \propto (P_{CH_4})^{-2}$, while the drop at low pressures is associated with the decrease in the linear absorption $\alpha_0 \propto P_{CH_4}$ and the number of slow molecules $\propto (P_{CH_4})^2$ contributing to the resonance.

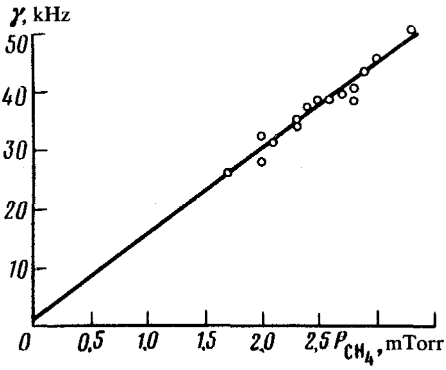

FIG. 3. Dependence of the homogeneous line width on the methane pressure. The solid line represents the straight line $\gamma = (2.5 + 14.2\ P_{CH_4})$ kHz.

The signal $\delta_{abs}(H)$ represents a resonance, whose peak, according to (3), fixes the Zeeman splitting $\Omega = g\mu_N(H)$, which is equal to $f/2$. This ratio gives a g-factor for the $F_2^{(2)}$ components $g = 0.31 \pm 0.01$, consistent with the data of Ref. 7. Figure 3 shows the dependence of the homogeneous line width γ on the methane pressure, obtained by analyzing experimental results using Eq. (3) by the method of least squares. The dependence is described well by the expression $\gamma = \gamma_0 + kP_{CH_4}$ with a coefficient of proportionality $k \simeq 14$ kHz/mTorr, which coincides with the well-known collisional broadening constant for the methane line[4] and with the constant $\gamma_0 \simeq 2.5$ kHz. This dependence $\gamma(P_{CH_4})$ indicates that the broadening of the resonance is determined by collisional broadening, while transit effects, in spite of the large transit broadening (~ 100 kHz) and fluctuations of the laser frequency (width of the spectrum ~ 10 kHz) have essentially no effect on resonance broadening.

Thus, in this work we proposed and realized a new method of frequency-modulation spectroscopy, which has high sensitivity and resolution. Using an extracavity absorbing cell with linear absorption 3×10^{-4} cm^{-1} and length 15 cm, a resolution of 3×10^9 was achieved without broadening the laser beam.

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Translated by M. Alferieff
Edited by S. J. Amoretty