Nonlinear shift of the resonance at the *E* line of the *P*(7) transition of the ν_3 band in methane

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The first observation is reported on nonlinear behavior in the impact shift of the resonance at the E line in methane as a function of the gas pressure; the shift falls off abruptly in the low-pressure region (~ 1 mTorr).

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1. The first observation of nonlinear behavior in the shift of the resonance at the $F_2^{(2)}$ line [the $P(7)$ transition of the ν_3 band] in methane as a function of the gas density was reported in Ref. 1 for pressures in the $\sim$1-mTorr region. The nonlinear character of the shift was attributed in that paper[1] to the influence of elastic scattering of excited particles at small angles, where the scattering-related Doppler shift $kv\theta$ (kv is the Doppler width, and θ is the characteristic scattering angle of the particles) is comparable to the homogeneous half-width Γ of the line. The pronounced nonlinear behavior of the shift had not plausible explanation in the existing theories of the broadening and shifting of spectral lines, which for a binary collision model always gave a linear dependence of the impact broadening and shift of the lines on the pressures of the gas. In the first theoretical papers[2,3] on the width and shift of resonances at low pressures with allowance for elastic scattering it was shown that under certain conditions the shift of the resonance actually depends on the homogeneous linewidth, and this circumstance, in turn, leads to a nonlinear dependence on the gas density. Unfortunately, the presence of magnetic hyperfine structure at the $F_2^{(2)}$ transitions of methane[4–6] leads to additional impact and field shifts, which complicate comparison of the theoretical and experimental results.

An interesting object for spectroscopic studies is the E line of the $P(7)$ transition of the ν_3 band of methane, which is a single line without hyperfine structure. Resonances at this line were first observed by Luntz and co-workers.[7,8] Detailed studies of the resonances and their application to frequency stabilization are reported in Refs. 9–11.

In the present study we have made the first reported observation of nonlinear behavior in the impact shift of the resonance at the E line of methane as a function of the gas pressure, which features a sharp drop in the collisional shift at low pressures that could prove important for developing optical frequency standards.

2. The experimental layout is shown in Fig. 1. The collisional shift of the resonance at the E line in methane was measured by the following technique. The frequency of He–Ne laser 1 with an internal methane cell was tuned to the maximum of the power resonance, and the shift of this frequency from that of the stable laser 3 was measured as the pressure of the gas in the cell was changed. The E line of methane lies at a distance of $\cong 3$ GHz on the red side of the center of the gain line of the He–Ne laser, at a distance of $\cong 3$ GHz. To shift the gain line we used a transverse magnetic

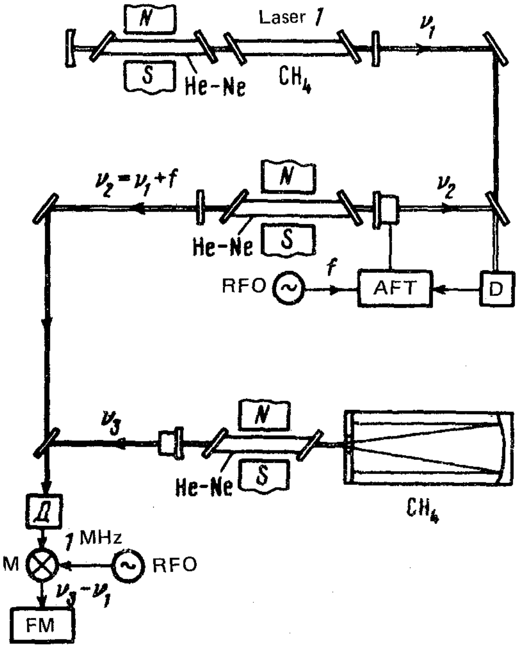

FIG. 1. Schematic of experimental apparatus. D—Photodetector, AFT—electronic automatic frequency-phase fine tuner, RFO—rf oscillator, M—mixer, FM—frequency meter.

field with a strength of $\cong 1800$ Oe. The resonator of laser 1 was 200 cm long, and the absorbing methane cell was 120 cm long. The light beam was 0.5 cm in diameter. At a methane pressure 2×10^{-3} Torr the power resonance had a width of 100 kHz, an intensity of 0.1 mW, and a contrast of 10%. The frequency was stabilized according to the maximum of this resonance with the aid of an automatic electronic fine-tuning system. The frequency stability was $\sim 10^{-14}$ at an averaging time $\tau = 1$ sec. Laser 3 was a He–Ne/CH_4 laser with a telescopic beam expander (see Fig. 1); the frequency of this laser was stabilized by the maximum of the ultranarrow (width 5 kHz) resonance at the E line of methane. The long-term stability of this laser, 6×10^{-15}, provided the necessary accuracy of measurement.

We measured the shifts of the frequency of laser 1 when neon was added to the absorbing cell. The methane pressure was 2 mTorr. The studies were made at a low density of the field in the resonator (the saturation parameter was $\kappa \sim 0.1$). This enabled us to eliminate shifts of the resonance due to changes in the intensity of the field in the resonator (see Ref. 12). Figure 2 shows the measured shift of the resonance in methane as a function of the neon pressure. The shift is seen to be nonlinear. For pressures in the region ~ 1 mTorr the shift is small, amounting to $\cong 30$ Hz/mTorr. As the pressure is increased, however, the shift grows rapidly. At a neon pressure of 6–8 mTorr the slope of the shift is $\cong 300$ Hz/mTorr.

3. Various physical and technical factors can cause additional shifts in the resonance and in the stabilized frequency upon changes in the gas density and can conse-

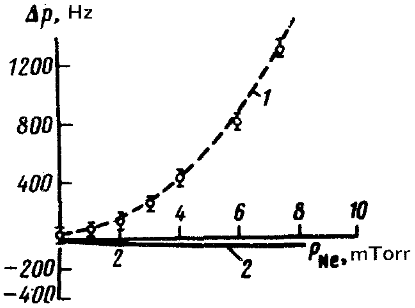

FIG. 2. Shift in the resonance peak at the E line in methane as a function of the neon pressure. The methane pressure is 2 mTorr. The dashed curve 2 is the calculated behavior of the shift in the resonance peak in methane due to the quadratic Doppler effect.

quently distort the character of the collisional shift.

Significant difficulties arose in attempts to eliminate additional modulation of the radiation power at the scanning frequency of the resonator length. This modulation arises because of angular displacements of the piezoelectric elements on which the mirrors are mounted and because of self-focusing of the radiation in the absorbing cell. The presence of an additional shift in the automatic fine-tuning system leads to a shift in the frequency with respect to the maximum of the resonance, by an amount that depends nonlinearly on the gas pressure. Measurements showed that with careful tuning of the resonator, the shift of the stabilized frequency due to the presence of additional modulation of the radiation power was not more than 200 Hz as the neon pressure was changed from zero to 8 mTorr.

The effect of the curvature of the wave front of the Gaussian beam on the shift of the nonlinear absorption resonance was examined theoretically and experimentally by Bordé, Hall, and co-workers[13,14] for the case of a weak traveling wave in the field of a strong oncoming wave. For a laser with an internal absorption cell the shifts of the resonance are proportional to the difference in the intensities of the oppositely directed waves forming the standing wave.[15] A numerical calculation for methane shows that under the conditions of the experiment described above, the shift does not exceed 10 Hz in the investigated region of gas pressures in the cell.[1)]

In the region of low methane pressure (~1 mTorr), where transit effects due to the slow-velocity selection of the molecules begin to play an important role, additional shifts in the resonance appear upon changes in the pressure of the absorbing gas because of the recoil and quadratic Doppler effects. The influence of the recoil effect in the transit region on the position of the resonance peaks in methane was considered by Titov,[16] who showed that the relative size of the shift is not more than 10^{-13}–10^{-14}. Figure 2 shows the calculated behavior (curve 2) of the quadratic Doppler shift of the resonance in methane as obtained under the conditions of our experiment and according to the results of experiment and according to the results of Baklanov and Dubetskiĭ.[17] The shift, which is toward the red, is small and does not alter the nonlinear character of the measured shift as a function of pressure. The dashed curve in Fig. 2 gives the collisional shift in the resonance at the E line in methane.

A qualitative explanation of the observed nonlinearity of the impact shift of the resonance was given in Ref. 1. According to the theory of Alekseev *et al.*,[2] the difference in the slopes of the shift of the nonlinear resonance in the low-pressure (Δ_1) and high-pressure (Δ_2) regions for a c/r^6 potential is given as $(\Delta_1/\Delta_2) \sim (\sigma_i/\sigma_e)^{3/2}$, where σ_i and σ_e are the total inelastic and elastic scattering cross sections, respectively. According to the data of a previous paper,[14] the ratio of the elastic to inelastic scattering cross sections is ~ 4, and so $\Delta_2/\Delta_1 \sim 8$, in good agreement with the results of the experiment described above.

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[1)] We are indebted to E. A. Titov for the numerical calculations.

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