Observation of anomalous Zeeman effect in the $F_2^{(2)}$ line of methane

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The first observation of anomalous Zeeman effect in the components of the magnetic hyperfine structure (MHFS) of the $F_2^{(2)}$ line of methane ($\lambda = 3.39$ μm), asymmetry of nonlinear resonance in a magnetic field due to the recoil effect, and measurement of the shifts of the individual components of MHFS in weak magnetic fields are reported.

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1. The Zeeman effect in the $F_2^{(2)}$ line of methane [the $P(7)$ transition of the ν_3 band] was first observed in Refs. 1 and 2 by using the method of nonlinear laser spectroscopy. The rotational magnetic moment of a methane molecule for the ground and the excited vibrational levels was measured in Ref. 1. The g factors and their sign were determined more accurately in Ref. 2 within the error limits of both levels. The nonlinear resonance width in Refs. 1 and 2 was equal to $\sim 10^5$ Hz, which made it possible to perform experiments at magnetic fields of $\sim$100 Oe and observe only a normal Zeeman effect. The magnetic hyperfine structure (MHFS) of a methane line was resolved in Ref. 3, using a telescopic beam expander (TBE) outside of the cavity. Relatively strong resonances with a width of $\sim$1 kHz were obtained by using a TBE inside a cavity.[4] This made it possible to study the anomalous Zeeman effect in weak magnetic fields of 1–10 Oe when the Zeeman level splitting is smaller than the MHFS intervals.[1)]

In this paper we report the results of the first investigation of the anomalous Zeeman effect in the $F^{(2)}$ line of methane, the observation of a new effect–a nonlinear resonance asymmetry in a magnetic field due to the recoil effect, and the measurement of the shifts of the MHFS components of methane in weak magnetic fields.

2. The experiments were performed using He-Ne/CH_4 laser with $\lambda = 3.39$ μm and and a telescopic beam expander inside a cavity, whose design and main parameters were described in detail elsewhere.[5] The light beam in the absorption cell had a diameter of $\sim$14 cm, which made it possible to obtain $\sim$1-kHz–wide resonances in methane at a pressure of 10^{-5} Torr and an MHFS in the $F^{(2)}$ line of methane. The longitudinal magnetic field with an intensity of 0 to 5 Oe was produced in the absorption cell by means of solenoid. The light in the cavity was linearly polarized. The MHFS components of methane were determined by recording the first-harmonic signal of the radiation on a recording instrument while varying the laser frequency with use of the TBE, which was measured relative to the reference, frequency-stabilized, He-Ne/CH_4 laser (see Ref. 5). Figure 1 shows traces of MHFS of the $F^{(2)}$ line of methane without a magnetic field (Fig. 1a) and in a 5–Oe, longitudinal, mag-

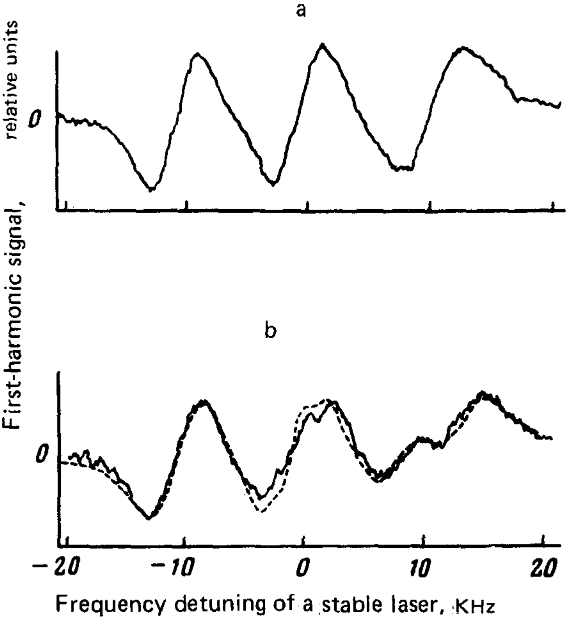

FIG. 1. Trace of a magnetic hyperfine structure of the $F_2^{(2)}$ line of methane (a) at $H = 0$ and (b) at $H = 5$ Oe. The methane pressure is 3×10^{-5} Torr, the modulation frequency is 800 Hz, the deviation amplitude is 300 Hz, the integration constant is 1 sec, and the recording time is 5 min. The dashed curve represents a calculation of the saturated absorption of methane, $\Gamma = 2$ kHz, and $H = 7$ Oe.

netic field (Fig. 1b). The shape of the hyperfine-structure components can be changed by applying a magnetic field to the absorption cell. The high-frequency component (the $F_1 \to F_2 = 8 \to 7$ transition, where F_1 and F_2 are the total moments of the lower and upper levels) is split into ~3.5 kHz and the central component ($7 \to 6$), which is broadened by 1.5 kHz, becomes asymmetric. A significant variation of the resonance was not observed in the $6 \to 5$ transition.

Because of the relatively high intensity of the nonlinear resonances in the MHFS components, we were able to stabilize the frequency of the laser with the TBE with respect to the maximum of each resonance and measure the shifts of the individual MHFS components in a magnetic field with a relative accuracy of $\sim 10^{-13}$. We measured the shifts at a methane pressure of 6×10^{-5} Torr. Within the limits of the measurement error, a quadratic frequency shift in the magnetic field was observed in all three main components of the hyperfine structure. The resonance peak in the 6–5 transition was shifted toward the blue region by the amount 14 ± 3 Hz/Oe^2 and the resonances in the 7–6 and 8–7 transitions were shifted in the opposite direction by the amount 19 ± 4 and 25 ± 15 Hz/Oe^2, respectively. The large error in the shift of the rf component of the MHFS is attributable to its relatively large splitting and broadening in a magnetic field.

3. The obtained experimental results can be qualitatively accounted for by the

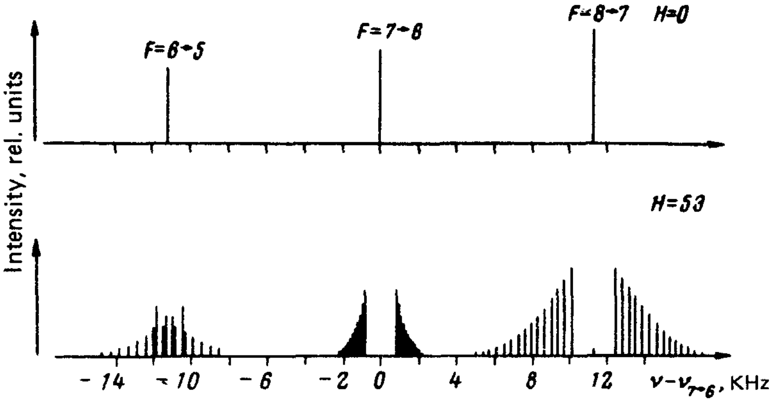

FIG. 2. Location and intensities of $\sigma_{\pm}$ components of the lines in the hyperfine-structure transitions of methane in a magnetic field when the recoil is disregarded (linear approximation).

splitting of lines into $\sigma_{\pm}$ components in the MHFS transitions. An anomalous Zeeman effect can be observed in a weak magnetic field (~0–5 Oe). The MHFS lines can be split in different ways, since the g factors of the upper and lower levels are different in the MHFS transitions and depend on the total moments F_1 and F_2. Figure 2 illustrates the splitting of the main MHFS components of the $F_2^{(2)}$ line of methane in a magnetic field. The transitions, in which the projection M of the total moment has been changed by the amount $\Delta M = \pm 1$, are resolved for a linearly polarized light and axial magnetic field. Each hyperfine-structure transition is split into $2F_2 + 1$ component. Since the frequency interval between the M components is much smaller than the experimentally obtained resonance half-width (~2 kHz) in methane, these components cannot be resolved. We can see in Fig. 2 that the calculated splitting into σ_+ and σ_- components for the lines in the MHFS transitions is in good agreement with that observed in the experiment.

The asymmetry of the line in the $7 \to 6$ transition is a new physical effect observed in the experiment. This phenomenon, which is attributable to the influence of recoil in a magnetic field, has been observed both in a normal and anomalous Zeeman effect. A qualitative picture of the asymmetry is as follows. In addition to the resonances of the σ_+ and σ_- components ($\Delta M = \pm 1$), additional resonances with common upper or lower levels (cross resonances) occur in the transitions as a result of resonance nonlinear interaction of a linearly polarized, standing, electromagnetic wave with a gas in a longitudinal magnetic field.$^{2)}$ Because of the recoil, each resonance of the σ components is split into $2\delta(\delta = \hbar k^2/2m$, where k is the wave number and and m is the mass of a molecule), the frequencies of the cross resonances with common lower levels are shifted by the amount δ toward the blue region relative to the transition frequency and the frequencies of the resonances with common upper levels are shifted by the amount $-\delta$ (Fig. 3). Since the number of transitions with common upper levels is different from the number of transitions with common lower levels, the total intensities of the indicated cross resonances differ from each other. The recoil effect produces an asymmetry of nonlinear resonances in a magnetic field.

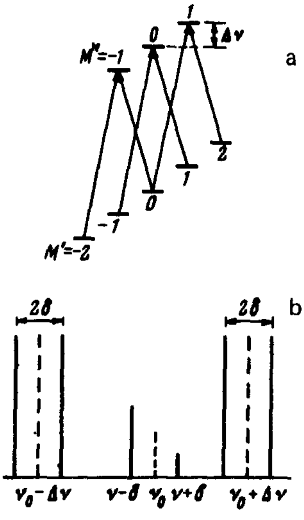

FIG. 3. A qualitative picture of the asymmetry of nonlinear resonance in a magnetic field due to the recoil when the g factors of the levels are equal. (a) Transition scheme, (b) location and relative intensities of the resonances of the σ components and of the cross resonances without the recoil effect.

We have calculated the nonlinear absorption coefficient for an anomalous Zeeman effect in a longitudinal magnetic field, with due regard for the recoil effect. A good agreement between the experimental curve and the theoretical first derivative of the saturated-absorption coefficient of the $F_2^{(2)}$ line of methane (Fig. 1b) was obtained for the calculated value of homogeneous half-width $\Gamma = 2$ kHz and magnetic-field intensity $H = 7$ Oe (Fig. 1b). At the same time, the longitudinal magnetic field in the experiment was 5 Oe. This can be attributed to the fact that in addition to the field produced by the solenoid, the magnetic field of the earth and of other external sources with an intensity of ~1–2 Oe (magnetic shielding of the absorption cell was missing) influenced the absorbing gas in the experiment.

The experimentally observed shifts of the MHFS components of methane in a magnetic field are in agreement within the limits of the measurement error with the calculated values obtained by us with use of the results of Ref. 6 and allowance for the recoil and influence of the wings of the neighboring components.

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1) The anomalous Zeeman effect in methane was first observed by Bagayev *et al.*[5]

2) The cross resonances occur in a three-level system, which is the simplest case of the occurence of such resonances, if two bound transitions are overlapped by Doppler broadening.

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