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Shifts of the nonlinear methane resonance at 3.39 μm

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Shifts of the nonlinear resonance in methane at the $F_2^{(2)}$ line due to various physical factors have been investigated, using frequency stabilized He-Ne lasers with methane absorption cells. The measured shifts of the resonance on varying the field intensity in the resonator, the methane pressure in the cell, and the strength of an applied longitudinal magnetic field are in agreement with theoretical calculations. The shifts depend nonlinearly on the uniform width 2Γ of the resonance. When $\Gamma \sim 100$ kHz the observed field shift is large and is due to differences in the field broadening of the magnetic hyperfine structure components and to the effect of crossed resonances. When $\Gamma \sim 10$–20 kHz the field shift is small and is due mainly to relative changes in the intensities of the principal hyperfine structure components resulting from saturation. The nonlinear pressure dependence of the resonance shift is due to the effect of the magnetic hyperfine structure and to particle collisions. At methane pressures above 1 mTorr the shift is due mainly to collisions and amounts to ~ 400 Hz/mTorr; at pressures below 1 mTorr the impact shift of the resonance is compensated by a shift due to the effect of the magnetic hyperfine structure, and the resultant shift is small.

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INTRODUCTION

Narrow nonlinear optical resonances have made it possible to make precise measurements of the shifts of spectral lines of gases under the influence of various physical factors.[1] This has proved to be especially important in studying lines due to molecular vibrational-rotational (VR) transitions. The collisional shift of molecular VR lines amounts to ~100 kHz at ~1 Torr, and this is some 10^3 times smaller than the Doppler width of the line. It is virtually impossible to study such small line shifts by the traditional methods of linear spectroscopy. The lines are substantially broader at high pressures and the individual VR components of the molecular transitions overlap; this also makes it difficult to analyze the profile of an isolated spectrum line. We note that for complex molecules the VR lines overlap even within the limits of their Doppler widths. The shifts of such lines can be studied only with the aid of nonlinear resonances whose widths are 10^3–10^4 times smaller than the Doppler width.

In this paper we report detailed studies of the shifts of the methane $F_2^{(2)}$ line at $\lambda = 3.39$ μm under the action of various physical factors. This line was chosen for study for a number of reasons. The narrowest resonances in the optical range have been obtained from this methane transition and have been used to achieve high stability and reproducibility at a level of 10^{-13}–10^{-14} of the frequency of a He-Ne laser working at $\lambda = 3.39$ μm.[2,3] The use of lasers whose frequency can be adjusted with high accuracy to the peak of the resonance makes it possible to measure shifts of the methane resonance with an accuracy of the order of 10 Hz. The studies proved to be important for an understanding of the physical nature of the shifts of the $F_2^{(2)}$ line and shed light on the problem of achieving high frequency reproducibility in a He–Ne/CH_4 laser.

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The choice of the methane absorption line and the research techniques are discussed in Sec. 1; the experimental setup, the modes, and the technique of measuring the resonance shifts are described in Sec. 2; the principal results of the research are presented in Sec. 3; Sec. 4 is devoted to a discussion of the results and their relation to other studies.

§1. CHOICE OF THE METHANE ABSORPTION LINE

Methane has been proposed as a nonlinear absorber for He-Ne lasers to obtain narrow resonances[4] and for automatic frequency stabilization.[5] The nonlinear resonance in methane was first observed by Barger and Hall.[6] Since then the He-Ne laser with a methane absorption cell has constantly attracted attention. The simplicity of the He-Ne/CH_4 laser, the fairly large absorption coefficient of methane (~0.2 cm^{-1}/Torr), which makes it possible to use very low methane pressures (10^3–10^{-5} Torr), and the coincidence of the centers of the neon working lines and the $F_2^{(2)}$ methane absorption line make this system very suitable for research. In the usual He-Ne/CH_4 laser with a resonator ~1 m long, the resonance is some 100–300 kHz wide. Increasing the length of the resonator to ~5 m and the diameter of the light beam to ~1 cm made it possible to obtain resonances 30–50 kHz wide with an intensity of ~1 mW.[2,3] The use of external[7] and internal[8] telescopic beam broadeners made it possible to achieve resonances in methane only ~1 kHz wide because of the increase in the time during which the particles interact with the field.

Many frequency-stabilization studies have been carried through with He-Ne/CH_4 lasers.[6,9-13,2,3,14-17] The construction of a frequency stabilized He-Ne/CH_4 laser with an emission line width of 0.4 Hz has been recently reported[18] as well as the frequency stabilization of an He-Ne laser against ultranarrow methane resonances with a relative width of ~10^{-11}.[19] Methane has been the subject of many physical experiments. The nonlinear pressure dependences of the broadening and shift,[2,20] the temperature shift of the resonance due to the quadratic Doppler effect,[3] the normal[21] and anomalous[22] Zeeman effects, the Stark effect,[23] the magnetic hyperfine structure (MHFS) of the VR transition,[7,8] and the recoil effect[22,24] have all been observed. The shift of the peak of the nonlinear resonance in methane due to the curvature of the wave front of the light beam has been detected.[25,26] The frequency of the working transition in methane has been measured[27] with an accuracy of ~10^{-10}.

MHFS measurements have shown that the $F_2^{(2)}$ line of methane has three strong components that differ little from one another in intensity, and two weak components. The main components are separated by ~11 kHz,[7,8] and the intensities of the nonlinear resonances at the MHFS components bear the ratios 0.9:1:1.2 to one another.[8,22] The differences between the intensities of the components, even when they are resolved, leads to their being saturated differently by the action of the light field and, as a consequence, to a dependence of the position and shape of the resonance on the field intensity in the resonator and the gas pressure in the absorption cell. This accounts for the previously observed[6,13] field-intensity dependence of the methane resonance shift. Under the conditions described in Refs. 6 and 13, the field shift was large (~1 kHz), whereas in Refs. 2 and 3 no appreciable shift of the stabilized frequency was observed on altering the methane pressure and the field intensity, so that high reproducibility of the He-Ne/CH_4 laser frequency could be achieved. The contradiction between the results of Refs. 2 and 3 and those of Refs. 6 and 13 made it necessary to pursue additional theoretical and experimental studies. Baklanov and Titov[28] carried through theoretical studies of the effect of the MHFS on the shift of the resonance. The effects on the position of the methane resonance of the quadratic Doppler effect, the recoil effect, and a magnetic field were investigated by Baklanov and Dubetskiĭ,[29] Baklanov,[30] and Baklanov and Belyaev,[31] respectively. The experimental studies presented below, as well as theorectial calculations, have shown that the shifts due to the MHFS depend on the impact width of an individual hyperfine-structure component and on the frequency stabilization conditions.

§2. EXPERIMENTAL SETUP

In the work reported here, the main attention was devoted to investigating the shifts of the methane resonance under conditions in which the MHFS components are not resolved. Unlike previous investigators, we measured the absolute shift of the resonance with respect to the central MHFS component of the $F_2^{(2)}$ line. The research technique was as follows: The frequency of an He-Ne/CH_4 laser was stabilized to the peak of the methane resonance, and the shift of this frequency was measured with respect to a laser having a telescopic beam broadener (TBB), whose frequency was stabilized to the peak of the central MHFS component.[19]

The investigations were conducted with the setup shown schematically in Fig. 1. The apparatus included two identical He-Ne lasers whose frequencies were in-

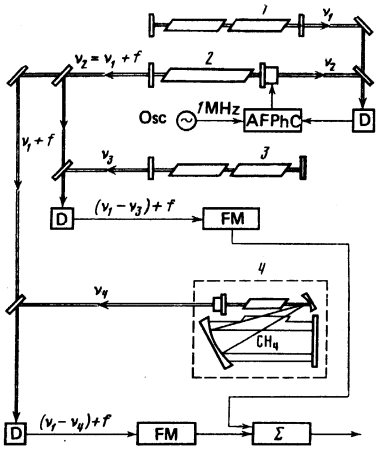

FIG. 1. Schematic diagram of the apparatus: 1, 3—stabilized lasers, 2—heterodyne laser, 4—laser with telescopic beam spreader, AFPhC—electronic automatic frequency and phase control system, Osc—rf oscillator, FM—frequency meter, D—photodetector.

dependently stabilized and which contained methane absorption cells (lasers 1 and 3), an auxiliary heterodyne laser (laser 2), and a laser containing a TBB (laser 4). The use of the heterodyne laser 2 made it possible to eliminate the effect of one of the investigated lasers on the frequency of the other and to make measurements in the region in which the difference between the frequencies of lasers 1 and 3 is not zero. The emission frequency of laser 2 was stabilized to that of laser 1 by means of an automatic frequency and phase control (AFPhC) system with a frequency shift of 1 MHz. Because of the high accuracy with which the laser frequencies were locked by the AFPhC system ($\sim 10^{-15}$), the auxiliary laser 2 had the same emission line width and frequency stability as laser 1. The recording system provided for simultaneous measurement of the frequency difference ($\nu_2 - \nu_3$) between the stabilized laser 3 and the auxiliary laser 2, and the beat frequency ($\nu_2 - \nu_4$) between laser 2 and laser 4, using electronic frequency meters. The absolute value of the difference frequency ($\nu_3 - \nu_4$) was recorded after the subtraction unit.

The He-Ne lasers used in the investigation were provided with long absorption cells so as to obtain strong methane resonances. This made it possible to achieve good absorption at low methane pressures. Increasing the diameter of the light beam with the simple resonator geometry made it possible to match the saturation parameters in the amplifying and absorbing cells and to increase the emission power. Each of the lasers had a 5-m long resonator and a 3-m long absorption cell. The laser resonators were formed by plane and spherical mirrors, the radii of curvature of the latter being 50 m. The light beam was ~1 cm in diameter. The transmission factors of the mirrors were so chosen as to maximize the power. A dc discharge was maintained in the amplifying tubes, which were of special construction,[17] in order to reduce the discharge noise. With a methane pressure of $\sim 10^{-3}$ Torr in the absorption cell we obtained resonances in the laser emission power some 30–50 kHz wide with a contrast above 70% and an intensity of ~1 mW. The pressure of the He-Ne mixture in the discharge tubes (~5.3 Torr) was so chosen as to ensure coincidence of the centers of the amplification and absorption lines within ~1 mHz. The lasers operated in a single-frequency generation mode with a power yield of 1–2 mW. Laser 3 was mounted within a solenoid in order to investigate the effect of a magnetic field on the methane absorption resonance. The longitudinal magnetic field could be varied from zero to 50 Oe.

The experimental equipment was mounted on a massive metal plate equipped with shock absorbers and coated with sound absorbing material. Under these conditions the emission line of a laser operating in the free generation mode was some 10 kHz wide. The characteristic frequencies of the disturbances lay in the region 20–30 Hz. The noise amplitude at the detector output did not exceed 4×10^{-8} W/Hz$^{1/2}$ and was determined by the photodetector noise. The laser with the TBB had a four-mirror resonator with an internal methane absorption cell. The construction of this laser is described in detail in Ref. 22. Its frequency was stabilized to the 2–3 kHz wide resonance at the 6→7 transition of the methane $F_2^{(2)}$ line.[19] The long-term frequency stability of this laser was 10^{-14} for an averaging time τ of 10 sec; this was adequate for measuring the frequency shifts of the investigated lasers.

The frequencies of lasers 1 and 2 were stabilized to peak generation power with the aid of electronic automatic frequency control (AFC) systems, which are described in detail in Ref. 17. The resonance shifts were measured for two stabilization modes: with the modulation frequency f comparable with the half width Γ of the resonance ($f = 15$ kHz), and with $f \ll \Gamma$ ($f = 340$ Hz). In both cases the deviations were smaller than 1 kHz.

At $f = 15$ kHz there was phase modulation of the resonator frequency (the modulation index $\beta = \Delta f/f$ being much smaller than unity). In this stabilization mode there are two weak sideband components with frequencies $\omega \pm f$ in the emission spectrum of the laser (ω is the laser emission frequency). In this case the stabilized frequency corresponds to the position at which the side components are equally absorbed and there is no amplitude modulation. When the absorption line is asymmetric the stabilized frequency does not coincide with the resonance peak. The phase modulation also made it possible to eliminate the effects of modulation on the width of the laser emission spectrum and on the results of measuring the stability of the frequency. In addition, when the modulation index was increased there appeared additional shifts of the stabilized frequency due to the effect of the MHFS on the methane working transition. Under the conditions of our experiment ($\beta \sim 0.1$) the shifts of the stabilized frequency were no greater than a few hertz when the modulation index was changed by a factor of two or three.

In the stabilization mode with $f = 340$ Hz ($f \ll \Gamma$) the stabilized frequency does not coincide with that of the peak of the resonance. In this case if all the necessary MHFS and methane-resonance parameters are known we should expect the calculated and experimental frequency dependences of the shift on the amplitude of the deviation to coincide. That dependence was calculated for the conditions of our experiment.[1)] The observed shift coincided with the calculated shift,[2)] indicating that there are no systematic frequency shifts due to the effects of various technical factors.

When the shifts were being measured, the long-term frequency stability of the investigated He-Ne/CH_4 lasers was better than 10^{-14} (with an averaging time τ of 1 sec) and the emission line was ~50 Hz wide. When the lasers were operating in the optimal modes for obtaining high short-term frequency stability, the width of the emission line was $\sim 4 \times 10^{-15}$ and the short-term stability was 10^{-13}–10^{-14} for an averaging time of 10^{-3}–10^{-1} sec (see Ref. 18).

In investigating the shifts of the stabilized frequency much attention was given to eliminating the effects of various technical factors on the accuracy with which the frequency was adjusted to the resonance. As in our earlier studies, the main difficulties were encountered

in eliminating the additional modulation of the laser power at the scanning frequency. The modulation of the laser emission power was mainly associated with angular motions of the piezoelectric element to which the mirror was fastened. The modulation of the power increased considerably, especially at high scanning frequencies above 1 kHz, when methane was introduced into the cell. This phenomenon is apparently associated with the difference between the refractive indices of methane at the side bands of the frequency modulated field in the resonator, and their different dispersions. The modulation signal strength depended nonlinearly on the emission power and the amplitude and frequency of the modulation; it was monitored with a synchronous detector detuned from the center of the resonance by ±5 MHz. The effect of amplitude modulation was reduced by specially fastening piezoceramic elements to the mirrors, carefully tuning the resonator, and using low amplitudes of the frequency swing and low power levels.

While the shifts were being measured we monitored the field strength in the resonator, the methane pressure, the modulation index, and the phase of the error signal in the AFC system. The output power of the laser was used to monitor the field strength in the resonator. The relation between the output power level and the value of the saturation parameter $\varkappa$ was established as follows. The laser was tuned to the peak of the resonance and the second harmonic of the modulation frequency was registered in the laser emission. For each investigated methane pressure we determined the dependence of the strength of the second harmonic signal on the power level and compared it with the calculated dependence of the amplitude of the second harmonic on the saturation parameter $\varkappa$. The calculation was carried through under the assumption that the field in the resonator had a Gaussian profile. The maximum of the second harmonic signal at the center of the resonance corresponded to $\varkappa \approx 1$. The methane pressure in the cell was monitored with the aid of specially calibrated manometric lamps. We also measured the modulation index during the experiments. To do this the beats between laser 2 and the investigated laser 3 were brought to an analog frequency-to-voltage converter (not shown on Fig. 1), whose output voltage was proportional to the frequency deviation. After appropriate calibration one could determine the scanning amplitude.

In the phase modulation mode ($f \approx \Gamma, \beta \ll 1$) the phase of the error signal at the output of the AFC synchronous detector depended on the field intensity in the resonator and on the methane pressure, because of the effect of the nonlinear absorption of the medium. When measuring the shifts of the stabilized frequency, the phase difference between the error and reference signals was adjusted to zero within ~1° for each of the investigated pressures and field strengths. A method was developed that enabled us to monitor the phase difference between the signals during the course of the experiment. The length of the laser resonator was varied at a low frequency in such a manner that the detuning of the generation frequency from the peak of the resonance did not exceed 1 kHz. The phase of the error signal with respect to the reference signal was observed at the output of the AFC key synchronous detector. By correcting the phase of the test signal fed to the control element of the laser, we adjusted the phase difference between the error and reference signals to zero. Measurements showed that with accurate adjustment of the phase within the range 1°–3°, the shift of the stabilized frequency did not exceed 10 Hz. The amplitude modulation of the output emission of the laser with the orthogonal phase was monitored in the stabilization mode with the aid of an additional synchronous detector.

§3. RESULTS

We measured the shifts of the stabilized frequency (f= 15 kHz) and the resonance peak (f= 340 Hz) as the field intensity in the resonator, the methane pressure in the cell, and the strength of the longitudinal magnetic field were varied.

The results of measuring the shifts on altering the field intensity within the resonator are presented in Figs. 2 and 3. The shifts were measured for various methane pressures in the range 4×10^{-4}–4×10^{-3} Torr while varying the saturation parameter $\varkappa$ from 0 to 1. On the basis of the experimental results we determined for each pressure the slope of the field shift, $\partial\Omega_{st}/\partial\varkappa$, in the region of low saturations. The half width Γ of the resonance for an individual MHFS component was determined for each pressure with allowance for the finite size of the light beam in the resonator in accordance with the results of Ref. 33, using the formula $\Gamma = 0.58/2\pi\tau + (\partial\Gamma/\partial p)P_{CH_4}$ (τ is the transit time of a molecule through the beam of radius a). In our experiment $\Gamma = 10(\text{kHz}) + 15(\text{kHz/mTorr}) \cdot P_{CH_4}(\text{mTorr})$. The value $\partial\Gamma/\partial p = 15$ kHz/mTorr was chosen in accordance with the results of Ref. 20. Supplementary measurements of the resonance width at various pressures showed good agreement with the calculated values. The slope of the field shift of the resonance peak is shown as a function of the methane pressure in Fig. 2, a (curve 1). The shift is small at methane pressures near 4×10^{-4}

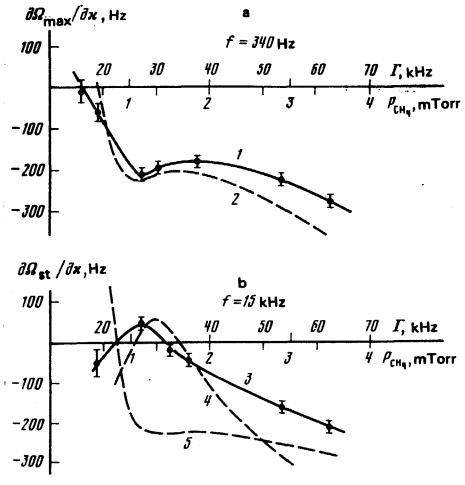

FIG. 2. Slope of the field shift of the resonance peak (a) and the stabilized frequency (b) versus methane pressure. Curves 1 and 3 represent the experimental results, and curves 2, 4, and 5 were calculated.

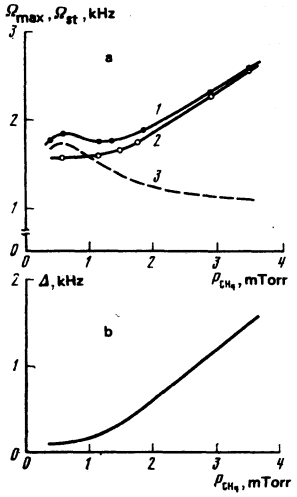

FIG. 3. a) Shift of the resonance peak (curve 1) and shift of the stabilized frequency (curve 2) of an He-Ne/CH_4 laser versus methane pressure. Curve 3 shows the calculated shift of the resonance peak due to the effect of the MHFS. b) Collisional shift of the resonance peak in methane.

Torr ($\Gamma \approx 16$ kHz), but at $P_{CH_4} = 3.5 \times 10^{-3}$ Torr ($\Gamma \approx 60$ kHz) it amounted to ≈300 Hz when $\varkappa$ was varied from 0 to 1. Figure 2, b (curve 3) shows the dependence of the slope of the field shift of the stabilized laser frequency at $f = 15$ kHz. It is evident that in the pressure region 1–2 mTorr the field shift of the stabilized frequency is considerably smaller than the shift of the resonance peak. In this pressure region ($\Gamma = 20$–40 kHz) changing the saturation parameter from 0 to 1 leads to shifts of the stabilized frequency no greater than ±50 Hz, and the slope of the field shift passes twice through zero. The shift increases strongly with increasing pressure.

The results of measuring the shifts of the resonance peak and the stabilized frequency as functions of the methane pressure in the cell are presented in Fig. 3. The frequency for each pressure was determined by extrapolating the field shift to zero field strength in the resonator. The observed dependence of the shift on the pressure is nonlinear. At pressures near ~1 mTorr the laser frequency shift was very small and lay between 10 and 20 Hz/mTorr. As the pressure was raised the shift increased sharply, and at pressures of 3–4 mTorr the slope of the shift amounted to ~400 Hz/mTorr.

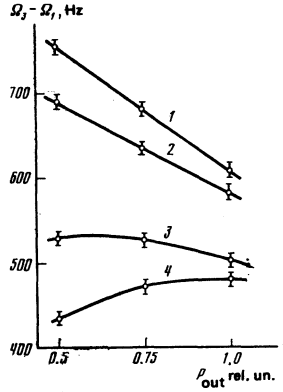

FIG. 4. Field shift of the stabilized frequency of the He-Ne/CH_4 laser for various values of the magnetic field strength H: 1 – $H = 0$, 2 – $H = 9$ Oe, 3 – $H = 27$ Oe, 4 – $H = 45$ Oe.

Figures 4 and 5 show our results on the effect of the external magnetic field on the stabilized laser frequency. The effect of the magnetic field was determined from the shift of the frequency of the investigated laser 3 with respect to that of the reference laser 1 when the solenoid was turned on. The shift of the stabilized laser frequency was measured as a function of the intensity of the light field in the resonator at various magnetic field strengths. These investigations were conducted in the region in which the half width Γ of the methane resonance was 70 kHz, where the field shift is fairly large because of the MHFS. As is evident from Fig. 4, the frequency shift resulting from a change in the light field intensity decreases as the magnetic field is increased and changes sign at a certain magnetic field strength ($H = 45$ Oe).

Figure 5 shows the dependence of the laser frequency shift on the strength of the magnetic field. The shift for each magnetic field was found by extrapolating the relationships found above to zero intensity of the field in the resonator. An appreciable dependence of the frequency on the magnetic field strength was observed (curve 1), the slope being 8 Hz/Oe; this slope increased (curve 2) when the width of the resonance was reduced (to $\Gamma = 20$ kHz).

§4. DISCUSSION

Various factors affect the position of a nonlinear resonance: collisional shift, the MHFS of the transition, the magnetic field, the quadratic Doppler effect, the recoil effect, and the curvature of the light-beam wave front. Under actual conditions the effects of these factors manifest themselves when changes take place in the gas pressure in the absorption cell, in the field intensity in the resonator, in the geometry of the resonator, in the external fields, etc.

The influence of the recoil effect on the position of the resonance as the gas pressure and field intensity are changed is due to coherent effects and depends on the ratio of the relaxation constants of the levels.[30] The resonance shifts resulting from the quadratic Doppler effect when the gas pressure and field intensity are changed will be observed in the transit region, where the contributions of the atoms to the intensity of the resonance will depend on their velocity. As was shown in Refs. 29 and 30, the influence of the recoil and quadratic Doppler effects is small (10^{-13}–10^{-14}).

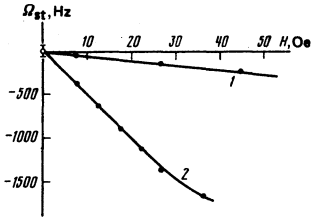

FIG. 5. Shift of the stabilized frequency versus the magnetic field strength for $\Gamma = 70$ kHz (1) and $\Gamma = 20$ kHz (2).

Under the experimental conditions described above it is extremely difficult to distinguish these effects because of the presence of other more important factors such as the MHFS and collisional shift. Here we shall therefore analyze only the effects of these latter factors, which are the principal ones under the conditions of our experiment.

As follows from our experiments (see Figs. 2 and 3) and the results of other studies,[16,28,34] when the resonance exhibits asymmetry associated with the MHFS, the position of the stabilized frequency will depend on the modulation frequency f. The stabilized frequency coincides with the resonance peak when $f \ll \Gamma$.

The experimental results on the shift of the resonance peak agree with the results of calculations presented in Ref. 34, and for the region $\Gamma \gg \Delta$, in Ref. 35. The measurements of the slope $\partial\Omega_{max}/\partial\varkappa$ of the field shift of the resonance peak as a function of pressure agree well with the theoretical predictions of Ref. 34 (curve 2 on Fig. 2). The calculations of Ref. 34 were carried through for the three main MHFS components with allowance for degeneracy of the levels in the magnetic quantum number, the Gaussian profile of the resonator field, and the influence of transit effects and the crossing of resonances. For convenience in comparing the calculations with the experimental results, the slope of curve 2 was calibrated in units of the saturation parameter as determined in the experiment (see Sec. 2). It is evident from Fig. 2, a that, as a function of pressure, the field shift of the peak changes sign in the region $\Gamma \approx 15$ kHz. The physical meaning of this result is as follows. In the region $\Gamma \gg \Delta$ the shift is due mainly to differences in the field broadening of the MHFS components and is in the direction of the higher frequencies. When $\Gamma \ll \Delta$ the relative changes in the intensities of the components as a result of saturation play the dominant part in the shift of the peak, and in this case the shift is toward the lower frequencies. As a result of competition between these two factors, the field shift vanishes in the region $\Gamma \sim \Delta$. At low pressures (~0.5 mTorr) the measured shift amounted to ~20 Hz when the saturation parameter was varied from 0 to 1. In this region of resonance widths, high reproducibility of the frequency of the He-Ne/CH_4 laser at a level of 10^{-13} or better can be secured. The field shift increases considerably as the pressure is raised. At high methane pressure ($\Gamma > 40$ kHz) there appears an additional red shift of the peak due to the effect of resonance crossings that result from the transitions $F = 6 \to 6$, $7 \to 7$.

The observed shifts of the stabilized frequency were compared with the results of the calculations of Ref. 34. The degeneracy of the levels, the effect of crossing resonances, and the Gaussian profile of the field in the resonator were taken into account in the calculations in the fifth order of perturbation theory. As was noted above, in the phase modulation mode the phase of the first harmonic signal in the emission power is not the same as the phase of the test signal but depends on the field intensity in the resonator and on the methane pressure. The field shifts of the stabilized frequency were calculated for two cases. In the first case the adjustment to the peak of the resonance corresponded to the vanishing of the in-phase component of the error signal in the AFC system. In the second case the position of the stabilized frequency was determined from the condition that the phase difference between the error and reference signals in the AFC system vanish.[3)] The resulting dependences of the slope of the field shift of the stabilized frequency on the methane pressure for the first and second cases are presented in Fig. 2, b (curves 4 and 5, respectively). The experimental results are in qualitative agreement with the calcuations. Quantitative agreement is made difficult to achieve by the fact that transit effects and the difference between the dispersions of the 4M side components of the emission spectrum were not taken into account in the calculations. For a resonance of width $\Gamma = 20$–30 kHz the frequency shift is determined by the three strong MHFS components. For a wide resonance ($\Gamma > 60$ kHz) the field shift is due mainly to level crossings.

Analysis of the observed behavior of the slope of the field shift of the stabilized frequency ($f = 15$ kHz) as a function of the methane pressure in the cell made it possible to determine conditions under which high reproducibility of the frequency of the He-Ne/CH_4 laser on changing the field intensity in the resonator could be achieved. For a narrow resonance ($\Gamma = 20$–35 kHz) with a working value of the saturation parameter of $\varkappa \approx 0.1$, a change in the field intensity by a factor of two results in a shift of no more than $\pm(2$–$3)$ Hz in the stabilized frequency; this explains the frequency reproducibility of $\sim 3 \times 10^{-14}$ achieved in Refs. 1 and 2.

The field shift was measured earlier in Ref. 13, where a linear shift of the frequency of a stabilized He-Ne/CH_4 laser of the order of 3 kHz was observed when the field was changed by a factor of 10 (the saturation parameter was varied from 1 to 10). In Ref. 32 the experimental data were compared with the results of a calculation of the field shift of the Lamb dip in the methane absorption line, resulting from the MHFS of the transition. The shift to the red was observed to be nonlinear. For the uniform halfwidth $\Gamma = 75$ kHz of the line measured in Ref. 32 the frequency shift of the He-Ne/CH_4 laser amounted to 400 ± 40 Hz when the saturation parameter $I_{8,7}$ calculated for the $F = 8 \to 7$ transition was varied from 0 to 1. In our experiments, when $\Gamma = 62$ kHz we observed a field shift of the resonance peak by 290 ± 20 Hz (see Fig. 3, a) when the saturation parameter $\varkappa$ was varied from 0 to 1. The agreement between these results must be regarded as good, in view of the differences between the calibration of the saturation parameter in our experiments and in those of Ref. 32, where the Gaussian distribution of the field in the light beam was not taken into account.

The experimental results on the pressure dependences of the shifts of the methane resonance peak and the stabilized frequency of the He-Ne/CH_4 laser (Fig. 3) are of interest. The observed nonlinear shifts of the resonance peak and the stabilized frequency are due to the MHFS and to the collisional shift in methane. Curve 3 of Fig. 3, a shows the calculated[34] methane-pressure

dependence of the shift of the resonance peak due to the MHFS. At low methane pressures (~1 mTorr) the experimental trend of the pressure dependence of the shift of the resonance peak agrees with the calculated trend, but the trends are considerably different at higher pressures. Experimentally, one observes a shift toward the higher frequencies, while the MHFS causes the resonance peak to shift toward the lower frequencies.

In our opinion the observed difference is due to the effect of impact shift in methane. The difference between the experimental and calculated curves reveals the pressure dependence of the collisional shift in methane (Fig. 3, b). The slope of the shift is about 10 times greater at high pressures (3–4 mTorr) than at low pressures (~1 mTorr). If we assume, in accordance with the results of Ref. 20, that the ratio of the cross sections for elastic and inelastic processes in methane is 4, then in accordance with theoretical results[36] we must conclude that for a $-1/r^6$ potential the difference between the slopes of the shifts for broad and narrow resonances will be ~12. At a methane pressure of ~1 mTorr the impact shift compensates the shift of the resonance peak due to MHFS effects and therefore leads to a very weak pressure dependence of the resultant shift. At pressures above 2 mTorr the pressure shift dominates the shift due to the MHFS. At pressures near ~1 Torr, therefore, the MHFS not only does not hinder the achievement of high frequency reproducibility, but even facilitates it to a considerable extent by reducing the effective pressure shift of the resonance. We note that the absolute frequency shift of the stabilized He-Ne/CH_4 laser with respect to the central MHFS component of the methane $F_2^{(2)}$ line agrees well with the calculated shift at pressures near ~1 mTorr.

The frequency shifts of an He-Ne/CH_4 laser resulting from changes in the methane pressure have been previously investigated.[6,2,37] The shift of the methane resonance peak was estimated in Ref. 6 as 75 ± 150 Hz/mTorr, which is in agreement with our results. The shift of the resonance peak observed in Ref. 37 on increasing the pressure, unlike the corresponding shift observed in the present work, was toward the lower frequencies. The authors of Ref. 37 attributed their observed red shift to the MHFS of the transition, but they did not take into account the shift in methane due to collisions.

The specific effect of a magnetic field on the stabilized frequency described in the present work is also associated with the MHFS of the working transition. In weak magnetic fields, when the Zeeman splitting is considerably smaller than the characteristic MHFS intervals, the shift due to the magnetic field is associated with two factors: the shift of each component in the magnetic field, and the Zeeman splitting of each line. Because of the differences among the g factors of the levels involved in the transitions under consideration, the MHFS components will be differently split and hence will have different effective widths. This phenomenon was recently observed experimentally[22] in a study of the anomalous Zeeman effect in the $F_2^{(2)}$ line of methane. It was found[22] that the strongest component of the $F_2^{(2)}$ transition ($F = 8 - 7$) is split more strongly than the others and is therefore broader. From this it follows that applying a magnetic field to the methane absorption cell will result in an additional shift of the resonance. The saturations of the MHFS components will become equalized in a magnetic field of a certain strength, and then the shift of the peak of the over-all methane resonance due to a change in the field intensity in the resonator will be substantially smaller (Fig. 4).

We note that the effect of the magnetic field depends on the saturating light field in the resonator. At high saturations, conditions may arise in which changes in the magnetic field will lead to no appreciable frequency shifts. The phenomenon that we have found is important for the purpose of improving the frequency reproducibility of an He-Ne/CH_4 laser under conditions in which the methane resonance is broad ($\Gamma \approx 70$ kHz). The shift of a nonlinear resonance in a magnetic field in the presence of MHFS has been treated theoretically.[31] The results of the calculation are in qualitative agreement with experiment. We note that a study of the effect of a magnetic field on the frequency of a stabilized He-Ne/CH_4 laser was reported earlier.[13] In that study no appreciable frequency shift was observed in a magnetic field of 10 Oe. In our experiments, in which the conditions were similar to those reported in Ref. 13 ($\Gamma \approx 70$ kHz) we observed a frequency shift of ~100 Hz on changing the magnetic field strength by 10 Oe.

The shift of a nonlinear resonance as a result of curvature of the wave front of the light beam in the resonator was discussed in Refs. 25 and 26. The shift occurs when the opposing waves that form a standing wave have different intensities and it depends on the parameter $2z/b$, where z is the longitudinal coordinate of the point at which the shift is calculated and b is a conformal parameter. In those studies the shift of the resonance peak due to curvature of the wave front was investigated for the case of an external absorption cell; in that case the shift may be fairly large (~10^{-11}).

The conclusions of Ref. 25 have been extended[34] to the case of a laser with an internal absorption cell. The shift of the resonance peak was determined for the case in which the intensities J_+ and J_- of the opposing waves differed little from one another ($\Delta J/J = (J_+ - J_-)/J \ll 1$). Estimates of the shifts based on the results of Refs. 25 and 34 showed that under the conditions of our experiment, in which $\Delta J/J \sim 0.1$, $2z/b \approx 0.2$, $b = 30$m, $\Gamma = 2 \times 10^4$ Hz, and the light-beam diameter $2a$ was 1 cm, a 50% change in the pressure would result in a ~5 Hz shift of the resonance peak. Thus, by properly selecting the resonator configuration and the transparency of the mirrors one can virtually eliminate the effect of wave-front curvature on the position of the resonance.

The studies of the shifts of the methane resonance discussed above show that when the width of the resonance is such that the MHFS components are not resolved, the main contributions to the resonance shift come from the MHFS and from particle collisions. The complicated effects of the MHFS are associated with

differences in the saturations of the individual components. The investigations have made it possible to explain the differences between the results of a number of studies of the frequency shifts and reproducibility of an He-Ne/CH_4 laser. The resonance shifts depend substantially on the widths of individual MHFS components. With half widths in the range 10–30 kHz the shifts are small and the MHFS does not hinder the achievement of high frequency reproducibility.

Let us briefly consider the accuracy of the frequency of an He-Ne/CH_4 laser. The accuracy of the frequency is defined as the degree to which the frequency of the unperturbed transition. Various physical and technical factors shift the laser frequency from the nominal frequency of the transition. Because of MHFS effects, the accuracy of the frequency of an He-Ne/CH_4 laser is usually limited to ~10^{-11}. In our experiments, in which the shift of the laser frequency with respect to the center of the methane $F_2^{(2)}$ line was measured very precisely, the accuracy in determining the frequency was substantially higher. At present, more experimental work on the accuracy of the frequency of an He-Ne/CH_4 laser is necessary. Such factors as the quadratic Doppler effect, the magnetic field strength, and the spread of the light beam have little effect on the frequency reproducibility, but they may substantially affect the accuracy of the frequency by giving rise to a constant shift of the resonance.

Lasers with high long-term frequency stability and reproducibility in the 10^{13}–10^{14} range make it possible to investigate the shifts of ultranarrow resonances with relative widths of 10^{-11}–10^{-12} and to conduct a number of high-precision physical experiments. The high frequency stability and reproducibility of an He-Ne/CH_4 laser can be carried over to other wavelength ranges by making use of methods of nonlinear optics. The construction of a CO_2 laser with a frequency stability of ~10^{-14} has been reported[38]; this was achieved by locking the frequency and phase of the third harmonic of the CO_2 laser to those of the He-Ne/CH_4 laser described above.

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[2)]The frequency shift as a function of the deviation amplitude, given in Ref. 32, does not agree with the calculation.

[3)]These calculations were made by E. A. Titov and V. M. Semibalamut, to whom the authors are indebted for permission to use their results here.

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Quantum statistics of multimode lasing and noise in intracavity laser spectroscopy

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The statistics of a multimode laser with equidistant modes is investigated during all stages of lasing evolution. The distribution functions of the photon numbers in individual modes and of the total photon number are obtained. The distribution of the photons in an individual mode agrees in all lasing stages with the thermal distribution, but the time of correlation of the number of photons in the laser mode is much longer than the corresponding time for thermal emission. The total photon distribution function is obtained also in the dynamic stage of the lasing and in the stationary regime. The limitations of the intracavity laser spectroscopy method, due to the statistical character of the multimode laser radiation, are considered. The sensitivity threshold of this method is obtained.

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INTRODUCTION

In a number of situations connected with the operation or application of a multimode laser, the description of the laser radiation in terms of the mean values of the photon occupation numbers or fields is insufficient. In these cases it is necessary to take into account the statistical properties of the laser radiation. Thus, e.g., the onset of an individual ultrashort pulse in the multimode radiation is not a determined process and requires a probabilistic description. This, as is well known, is due to the statistical character of the radiation during the initial stage of lasing.[1] Allowance for the statistics of multimode radiation is important also in the method of intracavity laser spectroscopy (ILS). The fluctuations of the photon occupation numbers about the mean values produce noise (shot noise due to the emission and absorption of the photons), which will be shown below to limit the capabilities of this method.

In the first example, an important role in the description of the system is played by the phase relations between the fields of the individual modes. In the photon-occupation-number representation this means allowance for the off-diagonal elements of the density matrix. In the second case, however, interest attaches usually to the spectroscopic aspect of the problem, i.e., to the distribution of the photons over the laser modes. To describe the statistical properties of the field, it suffices here to know the photon distribution function, i.e., the diagonal part of the density matrix of the photon sub-system. Since the spectrum is usually recorded over times that are long compared with the duration of the initial (linear) lasing stage, it is of interest to investigate the properties of the photon distribution function not only during the initial lasing stage, but also during all the succeeding ones. There is at present no such published analysis of the statistics of multimode laser emission. From among the papers closest in scope, we note that of Letokhov *et al.*[2,3] who obtained, for a special model of a laser with identical modes, control equations for the distribution functions of the number of photons in an individual mode and of the total number of photons, and obtained their stationary solutions.

In the present paper we consider a model of laser with unlike modes and with an inhomogeneous gain contour, a model more appropriate for standard multimode lasers. Inasmuch as in this laser the inversion depends only on the total number N of the photons in the resonator, the gain saturation leads to narrowing of only the distribution N, whereas the photon numbers in the individual modes turn out to have a broad Rayleigh (thermal) distribution. The absence of factors that stabilize the number of photons in an individual mode causes the fluctuations of the number of photons to be long-lived, and the correlation time to be long compared with the case of a single-mode laser. The results of the investigation can be used to analyze the role of fluctuations in ILS. The threshold of the sensitivity of the ILS method is obtained.

§1. CONTROL EQUATION FOR THE PHOTON SUBSYSTEM AND ITS SOLUTION FOR THE INITIAL LASING STAGE

1. We consider the interaction of two-level atoms of an active medium with an assembly of laser modes. We assume that the luminescence-line contour of the medium is homogeneous and the transverse relaxation time $T_2 \ll T_1$, where T_1 is the longitudinal-relaxation

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