Measurements of the absolute frequency of a He-Ne/CH_4 laser

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The results of absolute measurements of the methane transition frequency $F_2^{(2)}$ carried out in experiments separated by a time interval of 2 yr are reported. The difference between the $F_2^{(2)}$ and E transition frequencies of methane has also been measured.

1. The most accurate measurements of absolute frequencies of vibrational-rotational transitions of molecules which have been carried out to date have been obtained by several groups using He-Ne/CH_4 lasers at the wavelength $\lambda = 3.39\ \mu m$ (Table I). Domnin *et al.*[1] recently reported that the values measured in 1985 for the frequencies of the $F_2^{(2)}$ and E transitions in the absorption of methane ($\nu_F^{85} = 88\,376\,181\,595.7 \pm 0.5$ kHz and $\nu_E^{85} = 88\,373\,149\,029 \pm 0.07$ kHz) differ from values found in 1981: $\nu_F^{81} = 88\,376\,181\,600 \pm 0.5$ kHz (Ref. 2)[1)] and $\nu_E^{81} = 88\,373\,149\,033.3 \pm 1.7$ kHz (Ref. 3). The frequency of the methane E line was found by subtracting the independently measured difference between the frequencies of the $F_2^{(2)}$ and E absorption lines[3] ($\Delta\nu_{F-E}^{81} = 3\,032\,570.4 \pm 1.4$ kHz) from the measured frequency of the $F_2^{(2)}$ ($\nu_E^{81} = 88\,376\,181\,603.4 \pm 1.4$ kHz; Ref. 2).[2)]

Domnin *et al.*[1] suggested that the observed frequency shift might have occurred because the fundamental constants changed over the elapsed time and led to different changes in the frequencies of electronic and vibrational-rotational transitions. A conclusion of such a fundamental nature on the basis of a measurement of the frequency drift of one laser requires verification. In the present letter we report the results of a more detailed analysis of our absolute measurements of the frequencies of the methane

TABLE I.

Methane transition	ν_{CH_4}, kHz	Year	Reference
$F_2^{(2)}$ line	88376181618 ± 13.8	1980	7
	616 ± 3.0	1980	8
	603.0 ± 3.0	1981	4
	612 ± 11.0	1981	9
	603.4 ± 1.4	1981	2
	602.9 ± 1.2	1983	5
	600.0 ± 3.4	1985	10
	603.4 ± 7.0	1985	11

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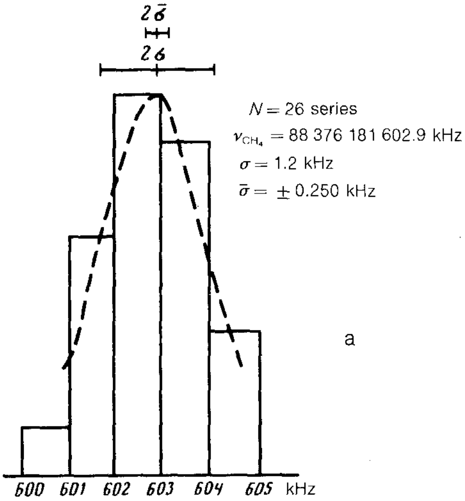
$F_2^{(2)}$ transitions, which were carried out in 1981 and 1983 (Refs. 4 and 5). As in Ref. 2, we carried out the analysis not only for the rms measurement error σ but also for the rms deviation of the mean value of the frequency, $\bar{\sigma}$. We also report here measurements of the frequency difference between the $F_2^{(2)}$ and E absorption lines of methane.



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Series 1
          Variance        rms deviation      Frequency
          141.8114           11.908          83506.57

Optical frequency F 88 376 181 603 075.3 Hz
Number of measurements deviating from range 35
Total number of measurements 100
Width of diff. corridor 10.0 Hz
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FIG. 1. a—Histogram of the results of absolute measurements of the frequency of He-Ne/CH_4 laser in 1983 by series; b—typical histogram of the results of one series of measurements of the frequency of a He-Ne/CH_4 laser.

In addition to the independent scientific and practical importance of these measurements, there is the possibility of using them to test independent measurements of the frequencies of the $F_2^{(2)}$ and E lines, since the frequency difference between these lines should not vary over time.

2. An optical-clock system was used to measure the frequency of the He-Ne laser, stabilized on the basis of the methane $F_2^{(2)}$ absorption line [a He-Ne/CH_4 ($F_2^{(2)}$) laser]. The measurement procedure and the parameters of the system are described in detail in Ref. 6. Twenty-eight series of measurements were carried out in 1981, and twenty-seven in 1983. Each series consisted of 50–100 measurements. The measurements were made with respect to the frequency of a rubidium standard, which was certified by the portable cesium standard of the Siberian State Scientific-Research Institute of Metrology.

In the 1981 experiments, the rms error of the measurements was $\sigma = \pm 3$ kHz, and the value $\bar{\sigma} = \pm 566$ Hz was found. Figure 1a is a histogram of the results of the absolute measurements of the frequency of the He-Ne/$CH_4F_2^{(2)}$ laser. The distribution of results is seen to be nearly a normal distribution (the dashed line). The values of σ and $\bar{\sigma}$ were ± 1.2 kHz and ± 250 Hz, respectively. In each individual series, the rms error in the mean value of the frequency did not exceed 10 Hz (Fig. 1b). Additional measurements showed that the indicated random measurement errors were a consequence of the frequency-instability characteristics of the rubidium standards that were used. From these results we conclude that the measurements of the $F_2^{(2)}$ transition frequencies carried out in methane in 1981 ($\nu_{CH_4}^{81} = 88\ 376\ 181\ 603.0 \pm 0.566$ kHz) agree within the measurement errors with those carried out in 1983 ($\nu_{CH_4}^{83} = 88\ 376\ 181\ 602.9 \pm 0.25$ Hz). This conclusion is at odds with the conclusions of Ref. 1. It follows from the new absolute frequencies of the $F_2^{(2)}$ and E absorption lines of methane which were obtained in 1985, and reported in Ref. 1, that the value of the difference between these frequencies, $\Delta\nu_{F-E} = 3\ 032\ 566 \pm 0.5$ kHz, differs from the value published in Ref. 3. It is thus pertinent to report our measurements of the difference between the frequencies of the $F_2^{(2)}$ and E components of the methane absorption line[12]:

$$\Delta\nu_{F-E} = 3032571670 \pm 30\ \text{Hz}.$$

This value is close to the results of Ref. 3 and agrees with the value found for $\Delta\nu_{F-E}$ from the results of our independent measurements of the absolute frequencies of the $F_2^{(2)}$ and E methane absorption line.[5]

3. Domnin *et al.*[1] did not compare the results of their measurements with the results of measurements of the frequencies of He-Ne/CH_4 lasers carried out by other groups. Table I lists the absolute frequencies found for the He-Ne/CH_4 laser over the past 5 yr from the data of Ref. 13 and the results of Ref. 11. Comparison of the results of the measurements by different groups does not support the conclusion that the frequency of the He-Ne/$CH_4F_2^{(2)}$ laser has varied over time. The most accurate absolute measurements of the frequency of this laser, which have been carried out since 1980, yield a mean value[13] $\bar{\nu}_{CH_4} = 88\ 376\ 181\ 602.3 \pm 0.8$ kHz.

At this point, it cannot be concluded from an analysis of the absolute measurements of the frequency of the He-Ne/CH_4 laser, which have been carried out to date, that relative temporal shifts are being observed in the frequencies of electronic and vibrational-rotational transitions. In our opinion, the large shifts of observed in Ref. 1 were consequences of technical factors involving excursions of the frequencies in the standards that were used. A final resolution of this question will require the use of standards with higher accuracy characteristics (10^{-13}). Such measurements will apparently become possible in the very near future. In addition, it would be worthwhile to analyze comparisons of the frequencies of lasers stabilized on the basis of electronic and vibrational-rotational transitions, whose frequencies lie in the same part of the spectrum. Such a study would not require the use of unique and very complicated systems such as those used in Ref. 1 to observe temporal shifts in transition frequencies.

1) A different value for the frequency of the methane $F_2^{(2)}$ transition was reported in Ref. 2: $\nu_F^{81} = 88\,376\,181\,603.4 \pm 1.4$ kHz.

2) Strictly speaking, when this method is used to measure the frequency of the E line, an observed temporal excursion of this frequency cannot be regarded as an independent measurement with respect to the shift of the frequency of the $F_2^{(2)}$ line.

3) The frequency at the peak of the $F_2^{(2)}$ absorption line is shifted 1.8 kHz from the peak of the central component of the magnetic hyperfine structure of this line.[14]

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