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Stabilisation of a laser to the calculated quantum transition frequency

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***Abstract*.** **A method to stabilise the frequency of a He – Ne laser with an intracavity nonlinear absorption cell to the calculated frequency of the 7 → 6 transition of $F_2^{(2)}P(7)\nu_3$ methane line ($\lambda = 3.39$ μm) is proposed and realised. The long-term frequency stability and reproducibility are measured for a He – Ne/CH_4 laser with a telescopic cavity.**

Keywords: *laser, stabilisation, transition frequency, recoil effect, methane.*

1. Introduction

Frequency standards are characterised, as a rule, by the radiation linewidth, long-term frequency stability and reproducibility [1]. The simplest method for generating a narrow radiation line (high short-term stability) is based on the use of an intense resonance providing a large slope of a frequency discriminator against a low noise background in the control bandwidth. Such a system can provide the required gain in the control loop at high frequencies, which is required to compensate for fast perturbations. As frequency references, along with resonances at quantum transitions in particles [2], the transmission bands of high-Q interferometers are used [3].

To obtain a high long-term stability and reproducibility of a laser frequency, it is necessary to use the narrowest optical references with Q factors achieving $10^{12} - 10^{14}$ (see, for example, [4 – 7]). The maximum slope of a frequency discriminator is achieved by detecting the first harmonic of the resonance in laser radiation. However, the long-term frequency stability and reproducibility are limited by the influence of the parasitic amplitude modulation (also caused by radiation scattering from extracavity optical elements [8]), the slope of a background signal due to the non-coincidence of the centres of the gain and absorption lines, and by other factors. In the case of uncontrollable variations in system parameters, this impairs the frequency stability.

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Thus, to obtain a high long-term frequency stability and reproducibility, it is preferable to use the third harmonic of resonance in laser radiation, which considerably reduces the influence of the above-mentioned factors on the frequency stability.

At the same time, the stabilised frequency is subjected to the parasitic amplitude modulation at the third harmonic, whose value is proportional, as a rule, to the frequency deviation. A decrease in the frequency deviation leads to the quadratic reduction of the signal-to-noise ratio in the frequency control loop, which also adversely affects the metrological parameters of the laser. In addition, upon stabilisation by the zero of the odd harmonic signal, the drift of the integrator zero and variation in the resonance amplitude will deteriorate the laser frequency stability.

Due to the recent development of digital equipment and computers, it became possible to stabilise frequency not only to the zero of odd harmonics but also to the extremum of a spectral line whose position is independent of the zero drift of a detection system. For frequency stabilisation over saturated dispersion resonances, the odd harmonics were used, and by recording the entire shape of the saturated dispersion line, the frequency was locked to the calculated 'unperturbed' transition frequency [9].

In this paper, we stabilised a He – Ne laser by saturated absorption resonances in methane by using as a reference the transition frequency in methane determined from the best fit of the experimental shape of the resonance by its calculated shape. The results of a direct comparison of the calculated frequencies of the 7 → 6 transition of $F_2^{(2)}P(7)\nu_3$ methane line ($\lambda = 3.39$ μm) obtained by using two identical lasers are presented.

2. Experimental setup

To achieve a high short-term and long-term stability (reproducibility), a frequency standard is constructed by using three He – Ne/CH_4 lasers [10] (Fig. 1). Reference laser 1 provides a narrow radiation line and high short-term frequency stability, laser 3 is used to obtain ultranarrow optical resonances applied as references for achieving a high long-term frequency stability and reproducibility. Heterodyne laser 2 has no frequency modulation, which is convenient for using its radiation as the output signal of the frequency standard.

The frequency ν_1 of reference laser 1 with an intracavity absorption cell was locked to a narrow (~ 50 kHz) intense (~ 1 mW) resonance in methane by the zero of the first

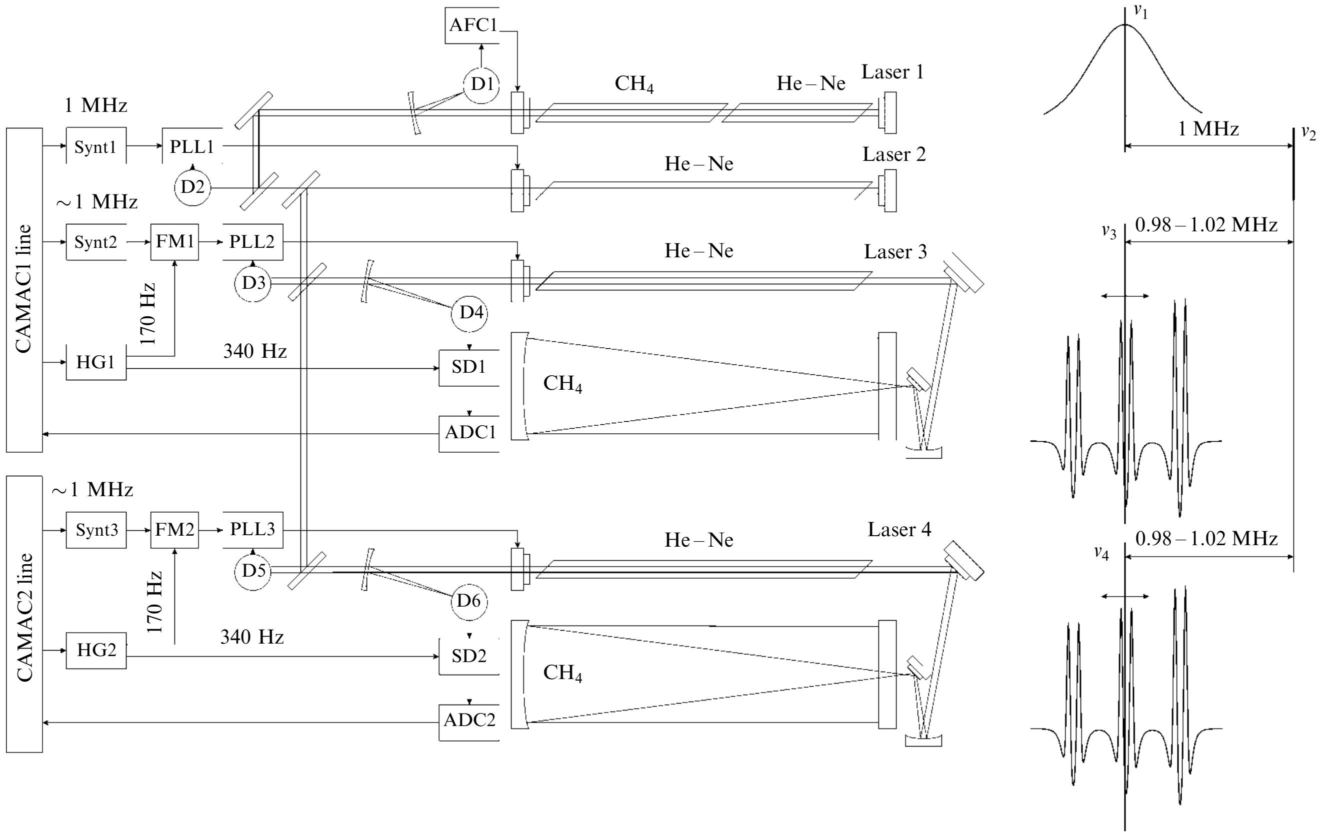

Figure 1. Scheme of the experimental setup.

harmonic of the absorption signal. The laser radiation was focused by a semi-transparent mirror on a liquid-nitrogen-cooled InSb photoresistor D1 whose output signal was fed to the input of an automatic frequency control (AFC) system. The error signal was obtained by modulating the laser frequency at a frequency of 15 kHz with deviation amplitude of $\sim$ 20 kHz and by lock-in detection of the first harmonic signal, the sign and value of this signal being proportional to the detuning from the absorption resonance centre. When the laser frequency shifts from the resonance maximum, the error signal appears at the AFC output, which was fed to a piezoelectric transducer PZT (not shown in Fig. 1). The corresponding change in the PZT length compensates the perturbation of the optical length of the cavity and, hence, the frequency shift. The control bandwidth $\sim$ 3 kHz provided the reference laser linewidth of 1 Hz.

The frequency ν_2 of heterodyne laser 2 was synchronised with that of reference laser 1 by using a phase-locked loop PLL1. Radiations of lasers 1 and 2 were mixed on a beamsplitter and the beat signal was detected by photodetector D2. The laser 2 frequency detuning specified by a quartz synthesiser SYNT1 was positive and equal to 1 MHz, i.e. $\nu_2 = \nu_1 + 1$ MHz.

Lasers 3 and 4 had a telescopic cavity with an intracavity absorption cell of diameter $\sim$ 50 cm and absorption length 8.5 m. To decrease the influence of the Earth's magnetic field, the absorbing cells had permalloy jackets with a field reduction coefficient of more than 10. Thus, the magnetic field inside the cells was less than 0.5 Oe.

By using PLL2 and computer-controlled SYNT2 synthesiser, the frequency of laser 3 was synchronised with the heterodyne laser frequency with detuning of $\sim$ 1 MHz to the red. By changing the frequency of the reference signal fed to the PLL of laser 3, we could tune slowly the frequency ν_3 of this laser and record a change in the radiation power caused by nonlinear absorption in methane.

Weak absorption signals were recorded by the method of lock-in detection. By using a frequency modulation oscillator FM1, the PLL2 reference signal frequency (and, correspondingly, the frequency of laser 3) was additionally modulated by a low-frequency signal ($f_m \sim 170$ Hz) produced by a harmonic generator HG1. A signal from the second output of this generator at the frequency $2f_m$ was used as a reference signal for a synchronous detector SD1. The synchronous detector separated the second harmonic from the absorption resonance in the laser output power detected with an InSb photodiode D4. An analogue-to-digital converter ADC1 was used to record signals in a PC.

Laser 4 was similar to laser 3. Its frequency ν_4 was also mixed with the frequency of heterodyne laser 2 and a beat signal was recorded with a photodetector D5. By using the PLL3, the beat frequency was set equal to the reference frequency supplied from the output of a computer-controlled SYNT3 synthesiser. By using a frequency modulation oscillator FM2, the PLL3 reference frequency was additionally modulated by a low-frequency signal ($f_m \sim 170$ Hz) produced by a harmonic generator HG2. The second harmonic of the saturated absorption resonance in the output power of laser 4 detected with an InSb photodiode D6 was recorded by using lock-in detection with the help of SD2 and ADC2 by scanning the SYNT3 frequency.

3. Stabilisation by the unperturbed quantum transition frequency

The first estimates of the stability and reproducibility of frequency standards based on the components of the recoil doublet were obtained in paper [11]. The authors of [11] showed the advantage of laser frequency locking to the maximum of the high-frequency recoil doublet component whose second-order Doppler frequency shift is compensated with an accuracy of ± 2 Hz by the shift due to the influence of the wing of the neighbouring resonance at the low-frequency component.

Note that the advantage of using the high-frequency component of the recoil doublet for frequency stabilisation can be realised only when the deviation amplitude is small compared to the homogeneous width Γ of the resonance ($A_{\rm dev} < 0.5\Gamma$). At the same time, for small deviation amplitude, the second-harmonic signal is weak, which results in the deterioration of the stability and reproducibility of the frequency standard. As the deviation amplitude increases, the frequency shifts of the high- and low-frequency components with changing the time-of-flight parameter become comparable. To achieve the high long-term frequency stability and reproducibility, it is also necessary to determine preliminarily the parameters of the system for which the shifts of the stabilised frequency with changing conditions will be minimal. At the same time, by locking the laser frequency to the calculated transition frequency, it is possible to vary the parameters of the system for obtaining the best signal-to-noise ratio. In addition, in this method of frequency stabilisation, the standards using the same transition as a frequency reference should give the same value of the stabilised frequency, which simplifies a comparison of standards.

To investigate the frequency stability and reproducibility of the laser locked to the calculated transition frequency, the shapes of recoil doublet components at $7 \to 6$ transition of the $F_2^{(2)}P(7)v_3$ methane line were simultaneously registered in both lasers with telescopic beam expanders. The experimental line shapes were fitted by the method of least squares with a theoretical curve derived in the small-saturation-parameter approximation [12]

$$L(\Omega) = A_1 F(\Omega - \Omega_1) + A_2 F(\Omega - \Omega_2),$$

$$F(\Omega) = \alpha_0 \mathrm{Re}\left\{1 - \frac{\chi}{4}\int_0^\infty \int_0^\infty \mathrm{d}x\mathrm{d}y\, \mathrm{e}^{-x-y} J_2(\xi y) \times \left[K_0 + \frac{\exp(\mathrm{i}\Omega y)}{K_0^{-1} - \mathrm{i}\tilde{\omega} y}\right]\right\}, \quad (1)$$

where

$$K_0^{-1} = 1 + \frac{x^2 + (x+y)^2}{2\beta^2}; \quad \tilde{\omega} = \frac{v_0^2 \omega_{21}}{2c^2\Gamma}; \quad \xi = \frac{A_{\rm dev}}{\Gamma};$$

$$\beta = \frac{\Gamma}{\Gamma_{\rm tr}}; \quad \Gamma_{\rm tr} = \frac{v_0}{2\pi a}; \quad \Omega = \frac{\omega - \omega_{21}}{\Gamma};$$

x and y are the integration variables; J_2 is the second-order Bessel function; $\tilde{\omega}$ is the quadratic Doppler frequency shift; ξ is the deviation amplitude $A_{\rm dev}$ divided by the resonance half-width Γ; $A_{1,2}$ is the amplitude of recoil doublet components; β is the time-of-flight parameter; $\Gamma_{\rm tr}$ is the time-of-flight broadening for a particle moving through a Gaussian beam of radius a with the mean thermal velocity v_0; Ω is the normalised frequency detuning from the transition frequency ω_{21}; c is the speed of light; and α_0 is the unsaturated absorption coefficient. This model, obtained in the linear approximation in the saturation parameter χ, takes into account the influence of time-of-flight effects related to a finite time of interaction of particles with the field of a standing light wave, the second-order Doppler effect, the splitting caused by the recoil effect, and modulation in the approximation of a small modulation frequency ($f_{\rm m} \ll \Gamma$).

The theoretical curves were fitted to the experimental data by assuming that the absorbing gas temperature, the light beam radius in the absorption cell, and the deviation amplitude were the known parameters. The gas temperature during experiments was controlled with an accuracy of 1°. To determine the light beam radius in the absorption cell, we studied the dependences of the beam radius at the output mirror and the threshold current related to intracavity diffraction losses on the distance between mirrors forming a telescopic light beam expander. By using the theory of resonators and matrix optics, the parameters of the laser beam at the output mirror and in the cell were calculated. By comparing the theoretical and experimental beam parameters, we determined the centre of the stability region of the cavity and the beam radius in the absorption cell ($a = 12.4$ cm).

The least-square fit was done by varying the amplitudes, frequencies, and half-widths of resonances, which were assumed the same for the high- and low-frequency components of the recoil doublet. The unperturbed frequency was assumed equal to the half-sum of the frequencies of these components. The high resolution achieved in our lasers with a telescopic cavity allowed us to neglect the influence of the neighbouring $6 \to 5$ and $8 \to 7$ transitions on the calculated frequency of the transition under study.

Figure 2 presents the second harmonic of the $7 \to 6$ absorption resonance fitted by the method of least squares by the theoretical curve calculated by expression (1). The experimental parameters were as follows: the absorbing gas pressure was 3×10^{-5} Torr, the saturation parameter $\chi \sim 1$ the modulation frequency $f_{\rm m} = 170$ Hz, the deviation amplitude $A_{\rm dev} = 800$ Hz, the signal accumulation time 2 s, and the total recording time of a resonance was ~ 200 s. The same parameters were used upon frequency stabilisation by the calculated unperturbed transition frequency. One can see that the theoretical curve well coincides with the experimental shape of the nonlinear absorption resonance at the components of the recoil doublet.

The calculated frequencies of the $7 \to 6$ transition of the $F_2^{(2)}P(7)v_3$ methane line obtained in lasers with telescopic cavities were compared in the following way: the second harmonic of the absorption resonance in the laser radiation power was recorded in each of lasers 3 and 4. Then the calculated curves were fitted to experimental data to determine the parameters of resonances and the calculated transition frequencies v with respect to the frequency of heterodyne laser 2, and the difference of the calculated transition frequencies $v_{7\to 6}$ obtained for lasers 3 and 4 was found. For the frequency standard, the calculated value should be used as the offset frequency between reference laser 1 and heterodyne laser 2. In this case, the radiation

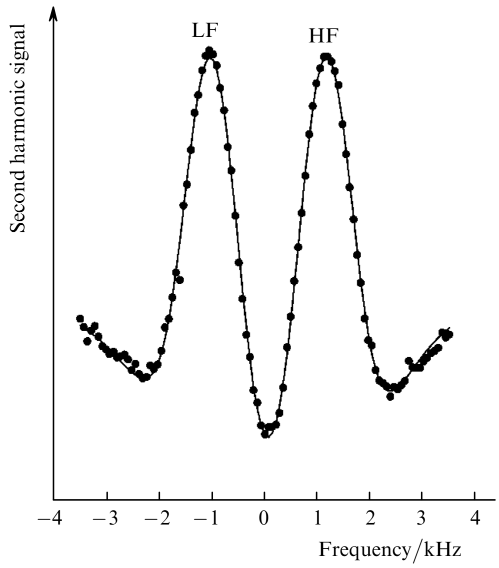

Figure 2. Second harmonic signal of the $7 \to 6$ transition of the $F_2^{(2)}P(7)\nu_3$ methane line (circles are experimental data, the solid curve is the best fit by the method of least squares).

frequency of the heterodyne laser will be located at a distance of 1 MHz from the calculated transition frequency determined in the laser with a telescopic cavity and will have simultaneously the short-term frequency stability provided by reference laser 1.

Figure 3 presents the results of measurements of the frequency stability performed during one of the days. On the abscissa the time is plotted in minutes, and on the ordinate the frequency of heterodyne laser 2 determined with respect to the calculated $7 \to 6$ transition of the $F_2^{(2)}P(7)\nu_3$ methane line is plotted. Figure 3a demonstrates the characteristic frequency drift of the reference laser with respect to the $7 \to 6$ transition frequency. Figure 3b shows the time dependence of the difference frequency $\nu_3 - \nu_4$. Because lasers 3 and 4 used same reference laser 1 and heterodyne laser 2, the long-term frequency stability and reproducibility provided by lasers with telescopic cavities were measured in the simplest way by eliminating the influence of the frequency instability of reference laser 1.

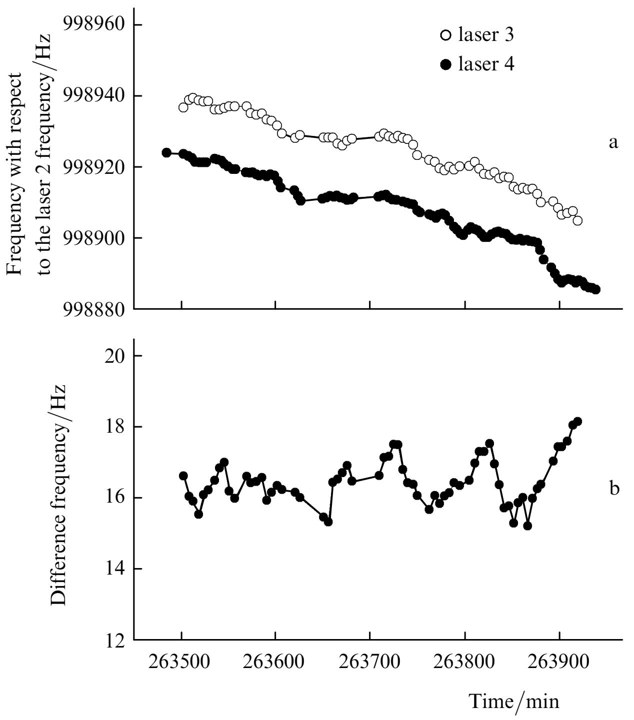

Figure 3. Time dependences of the calculated transition frequency determined in lasers 3 and 4 with respect to laser 2 (a) and the difference frequency $\nu_3 - \nu_4$ for lasers 3 and 4 (b).

Figure 4 shows the difference frequency for lasers 3 and 4 measured during six days. Based on these results, the long-term frequency stability and reproducibility from switching to switching of the standard was estimated as ~ 1 Hz (1.1×10^{-14} rel. units). The frequency reproducibility from standard to standard at this stage of investigations was estimated as ~ 8 Hz (9×10^{-14} rel. units). Note that the difference frequency $\nu_3 - \nu_4$ equal to 15 Hz was reproduced with an accuracy of ± 1 Hz for two years of observations.

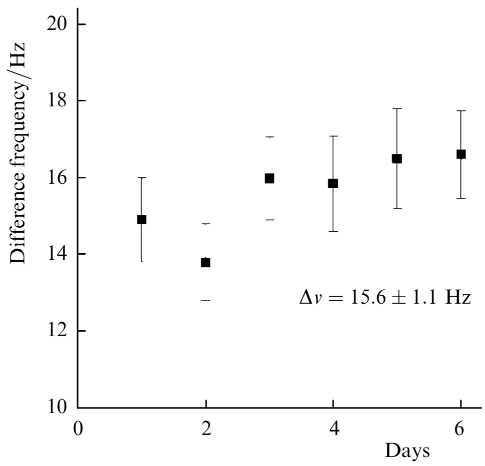

Figure 4. Variations in the difference frequency from switching to switching.

4. Conclusions

We have measured the frequency stability and reproducibility of a $He-Ne/CH_4$ laser stabilised to the calculated transition frequency. The main advantage of this frequency stabilisation method is that the frequency standards using the same transition as a reference but operating under different parameters, will give the same stabilised frequency coinciding with the unperturbed frequency of the transition used. In addition, this stabilisation method allows one to vary parameters of the standard for obtaining the best signal-to-noise ratio, while upon stabilisation to the resonance maximum, the main criterion for the choice of the operating parameters is the minimisation of the stabilised frequency shifts.

One of the main factors limiting the frequency stability of our standard is, in our opinion, the collision frequency shift. Our previous studies have shown that a change in pressure by 1 μTorr during experiments caused the frequency shift of 0.6 Hz [13]. On the other hand, the frequency difference for lasers 3 and 4 (~ 15 Hz) obtained in our experiments is caused by the difference in the size and structure of optical fields in absorption cells of these lasers. Measurements performed by the Foucault test have shown that the profiles of spherical mirrors of telescopic cavities are substantially different from ideal profiles. As a result, beam diameters in the absorption cells of telescopic lasers do not coincide, which leads to considerable errors in the determination of the transition frequency. It is obvious that this will also affect the accuracy of measuring the absolute transition frequency.

The metrological parameters of our frequency standards on the components of the recoil doublet can be further improved by manufacturing resonator mirrors more accurately and providing a precision control of the absorbing gas pressure, which can be achieved by controlling the homogeneous width of resonances. The consideration of the frequency modulation used in model calculations [14] also will increase the reproducibility of the frequency standard.

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