Laser frequency standards

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This paper gives a brief review of the present status of the problem of developing optical frequency standards. The basic principles of laser-frequency stabilization based on narrow nonlinear optical resonances are presented. The basic physical factors determining the shape and the shift of narrow resonances are discussed. The design of the modern laser frequency standard, based on the use of supernarrow resonances with a relative width of 10^{-11} and smaller as the reference, is studied. The currently used techniques for stabilizing the frequency of lasers for different ranges and the methods used for measuring the characteristics of the frequency stability are described. Specific achievements in the development of lasers with a narrow radiation line and high long-term frequency stability and reproducibility are presented. Special attention is devoted to the He–Ne laser. For this laser a radiation line width of less than 0.1 Hz and a long-term stability of 10^{-14}–10^{-15} have been obtained, and the effect of different factors on the shifts of the stabilized frequency are studied in detail. Results achieved in the development of the first optical clocks in the world are presented. The progress achieved in the absolute measurement of submillimeter, IR, and visible range laser frequencies is examined. Some applications of laser frequency standards and the prospects for their further development are described briefly.

CONTENTS

1. INTRODUCTION

One of the most significant achievements of microwave quantum electronics was the development of quantum frequency standards,[1,2] based on which the atomic time scale was constructed.[3] The period of the oscillations of a cesium standard, whose frequency is locked to the center of the hyperfine transition in the cesium atom, has now been adopted as the time standard and is equal to 1/9192631770.0 seconds. The development of coherent sources in the optical range and their scientific and practical applications stimulated work on the stabilization of laser frequencies. Just as in the microwave range, laser-frequency stabilization is based on narrow optical lines, to whose center the frequency is locked. All known methods for obtaining narrow spectral lines in the microwave range—the molecular beam method,[1,2] the bulb storage method (used in the H maser),[4] and the method of separated fields[5]—have not found applications in the optical region. This is attributable primarily to the effect of Doppler broadening, which can be neglected in the microwave range. For this reason, in the optical range it was necessary to develop fundamentally new methods for obtaining narrow spectral lines. During the last few years lasers with long-term frequency stability and frequency reproducibility at least as good as for masers have been developed. The short-term stability of lasers substantially exceeds

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that of the best masers. The realization of the division of the frequency of a stable laser into the radio frequency range has made possible the construction of a time scale based on the period of the optical oscillations.[6] The basic solution of this problem has stimulated attempts to achieve extremely high laser stability, based on which it will be possible to create new optical time and frequency standards.

Work on frequency stabilization can be divided into several stages, each of which has its own characteristic problems, methods, and results. The first stage encompasses work from the very beginning of the development of lasers (1960–1961) up to 1968. The high spectral purity of laser radiation was demonstrated in the very first studies of the properties of lasers.[7] During this period laser-frequency stabilization was realized primarily based on the maximum of the Doppler contour of the gain line and the Lamb dip in the radiation power of the laser. The relatively low degree of narrowness of the reference (10^{-6}–10^{-7}) did not permit achieving high short-term frequency stability. It was essentially determined by the perturbations of the laser cavity. The comparatively high gas pressure in the active medium and the concomitant collisional shift of the line prevented obtaining high long-term frequency stability and reproducibility. The relative laser-frequency stability achieved fell into the range 10^{-8}–10^{-9}. A complete description of the different methods of stabilization and of the results obtained is given in Ref. 8, which covers work up to 1968.

In the following years there was a sharp jump in the development of lasers with high frequency stability. This jump is linked to the use of the saturated absorption method,[90] which enabled obtaining intense narrow resonances with a relative width of 10^{-10}–10^{-11}. Frequency stabilization with the help of this method was first realized in Ref. 10. Lasers with a relative frequency stability of 10^{-14}–10^{-15}, frequency reproducibility of 10^{-13}–10^{-14}, and a line width of 0.07 Hz have now been developed.[11] Work on the synthesis and measurement of submillimeter, IR, and visible range frequencies began concurrently with the development of optical frequency standards.[12]

The new stage (from the end of the 1970s up to the present) is characterized by the development of new methods for obtaining extremely narrow nonlinear resonances (with a width of 10–100 Hz) by increasing the interaction time of the particles with the field: the method of separated optical fields, two-photon resonances, and resonances in the absorption of trapped particles. The methods for cooling particles and for selecting particles according to their velocities currently under development make it possible to decrease markedly the influence of the second-order Doppler effect. All this promises the achievement of frequency stability of the order of 10^{-16}–10^{-17}.

The detailed analysis of the properties of frequency-stabilized lasers and of the different technical aspects of stabilization falls outside the scope of this review. We shall confine our attention here to the basic principles of the construction of laser frequency standards, the description of the best results on the frequency stabilization of IR and visible range lasers, and a brief analysis of some applications of frequency-stabilized lasers.

2. PRINCIPLES OF FREQUENCY STABILIZATION

Even the narrowest gain lines of gas lasers are much wider than the width of the transmission band of the cavity, so that in the overwhelming majority of the cases the laser frequency is determined by the frequency of the cavity itself. The problem of frequency-stabilizing a laser is directly linked to the necessity of stabilizing the length of the laser cavity itself. External thermal, mechanical, and acoustic perturbations of the structural elements of the laser, as well as fluctuations of the coefficient of refraction of the gas-discharge plasma, make the largest contribution to the instability of the optical length of the cavity. The shift in the laser frequency $\delta\omega$ is related by a simple relation with the fluctuation of the cavity length L:

$$\delta\omega = \frac{\delta L}{L}\,\omega, \tag{1}$$

where ω is the laser frequency and δL is the change in the cavity length. For example, the thermal drift of a quartz cavity accompanying a temperature change of 1° gives $\delta\omega = 10^8 - 10^9$ Hz. The mechanical vibrations of the cavity caused by acoustic noise give rise to rapid fluctuations of the radiation frequency. The frequency spectrum of these perturbations depends strongly on the conditions of the experiment and lie in the range up to 10^4 Hz. For example, in dye jet lasers the perturbation spectrum is determined by the fluctuations of the pumping amplitude and the oscillations of the optical density of the jet and falls in the range up to 1 MHz.

The frequency of a laser is locked to the center of a spectral line with the help of an electronic automatic frequency control system (AFC). The relative accuracy of the tuning when recording the first harmonic in the radiation power of the laser is determined by the relation

$$\frac{\delta\omega}{\omega} = \frac{U_{\text{sn}}\,(\Delta f)^{1/2}}{2QK\eta\Delta I}, \tag{2}$$

where $Q = \omega/\gamma$ is the Q factor of the resonance, γ is the half-width of the resonance, $K = \Delta\Omega/\gamma$, $\Delta\Omega$ is the amplitude of the deviation of the lasing frequency by the probing modulation signal, ΔI is the intensity of the resonance, η is the sensitivity of the photodetector in V/W, U_{sn} is the spectral density of the noise at the output of the photodetector, and Δf is the stabilization band of the system.

The narrowest spectral lines are obtained in gases. The most important factor determining their width is the Doppler broadening $\Delta\omega_{\text{D}} \sim (v_0/c)\omega \sim 10^9$–$10^{10}$ Hz, where v_0 is the average thermal velocity of the particles. Narrow resonances with Doppler-free broadening in the optical range are obtained by the methods of nonlinear laser spectroscopy. Their width is directly related to the homogeneous width and can be 10^6 times smaller than the Doppler width. The narrower the resonance the more accurately the frequency can be tuned to its center. In achieving short-term frequency stability, however, the narrowness of the resonance is not determining, the intensity of the resonance is just as important (in other words, the signal-to-noise ratio in the AFC system). The position of the center of the resonance determines the long-term frequency stability and reproducibility. To achieve high long-term frequency stability and especially reproducibility the basic trend is still toward obtaining very narrow resonances, because their use makes it possible to reduce to a minimum the influence of different physical and technical factors on the shift of the stabilized laser frequency.

We shall give the basic standard definitions for describing the characteristics of frequency standards.[13] The accuracy of the frequency is the degree to which the frequency of the generator coincides with the frequency of the unperturbed (obtained under ideal conditions) quantum transition. The reproducibility of the frequency is the degree to which a generator of a given type will reproduce one and the same frequency from one firing of the generator to another and from sample to sample. The frequency stability is the degree to which the frequency of the generator remains constant over the period of time during which it operates continuously. When determining the value of the stability it is necessary to indicate the time interval τ over which the measurement is performed.

The properties of frequency-stabilized lasers can be described by the standard concepts used in the microwave range. The relative frequency stability over the time τ — $(\delta\omega/\omega_\tau)$ can be evaluated by measuring the variance of the fluctuations of the frequency difference $\Delta\omega = \omega_2 - \omega_1$ of two statistically independent generators:

$$\left(\frac{\delta\omega}{\omega}\right)_\tau = \frac{1}{\omega}\left[\frac{1}{2}\sigma^2(N, \tau, T)\right]^{1/2},$$

$$\sigma^2(N, \tau, T) = \frac{1}{N-1}\sum_{k=1}^{N}\left(\Delta\omega_k(\tau) - \frac{1}{N}\sum_{j=1}^{N}\Delta\omega_j(\tau)\right)^2, \quad (3)$$

where $\sigma^2(N, \tau, T)$ is the average value of the variance of the frequency fluctuations over N measurements, T is the period with which separate measurements are repeated, and τ is the time for one measurement (averaging time). Estimates of the variances based on the formula (3) depend on N and T, so that it is difficult to compare them in different experiments. A more convenient quantity is the two-sample "Allan variance"[14]:

$$\sigma^2(2, \tau) = \sigma^2(N = 2, \tau, T = \tau) = \frac{1}{2}\langle(\Delta\omega_{k+1}(\tau) - \Delta\omega_k(\tau))^2\rangle,$$

where $\langle ... \rangle$ is the averaging operator. This characteristic is now most widely used as a measure of the stability of the frequency of generators in the time domain. Although Allan's variance does not give complete information about the statistical properties of the radiation and it is difficult to change over to other characteristics of the radiation, its main advantages—the simplicity of the method for obtaining it and the possibility of classifying the perturbation noise with power-law spectral densities—explain its widespread use, which makes it possible to make a general comparison of the quality of generators of electromagnetic radiation in different ranges and of different types.

A more complete description of the statistical properties of laser radiation can be obtained by measuring the spectral characteristics: the form of the radiation line (line width) and the spectral density of the intensity (SDI) of frequency fluctuations. Analysis of the Fourier transformations in the frequency domain plays an important role both on a theoretical level and for applied purposes from the viewpoint of the power distribution in the frequency domain, so that the concept of SDI is widely used for describing the stability of generators, while the estimation of the SDI of relative deviations of the frequency is the basic indicator of frequency stability. It should be noted that for microwave generators the phase fluctuations determine the relative frequency stability in the range 10^{-12}–10^{-13}. In the optical range, as a rule, we are dealing with frequency fluctuations; phase fluctuations become important, just as in the microwave range, only when a frequency stability of 10^{-15}–10^{-16} is achieved. Therefore, in the general case, the optical radiation spectrum has a complicated form, consisting of a narrow part determined by frequency fluctuations and a wide plateau determined by the amplitude and phase fluctuations. The radiation spectrum of frequency-stabilized generators is analyzed in detail in Refs. 15–17.

3. OPTICAL REFERENCES

3.1. Methods for obtaining nonlinear resonances

There are a large number of methods for obtaining narrow nonlinear resonances without Doppler broadening.[9] For frequency standards the methods which make it possible to obtain resonances at the center of the spectral line are important. This makes it possible to relate the radiation frequency directly with the center of the transition. At the present time three physically different methods are of greatest interest for frequency standards: the saturated absorption method, two-photon absorption resonances in the field of a standing wave, and the method of separated optical fields.

The saturated absorption method is one of the most widely used methods in ultrahigh resolution spectroscopy. The absorption resonances appear in the nonlinear interaction of counterpropagating waves with a gas. Because of saturation effects two dips appear in the distribution of the population difference of the particle levels near the velocities $v_z = \pm\Omega/k$, where $\Omega = \omega - \omega_0$ is the detuning of the lasing frequency ω from the enter of the line ω_0, and c is the wave number. The dips intersect at the center of the spectral line, which intensifies the effects of saturation and gives rise to the appearance of a resonance dip in the absorption line with a homogeneous width. The basic physical idea of this method was developed in connection with the theory of the gas laser.[18] However, it was difficult to use an amplifying medium to obtain very narrow resonances because of the large homogeneous width of the gain line. An extremely small homogeneous width can be obtained for the absorption line because of the low gas pressure and by the selection of appropriate transitions. In the first experiments absorption-line resonances were observed with the help of a laser with an internal absorbing cell.[19,20] Here a decrease in the absorption at the center of the line gave rise to the appearance of a narrow peak in the radiation power when the lasing frequency of the laser was varied. The internal absorbing cell, as was shown theoretically in Ref. 21, also has a self-stabilizing effect on the lasing frequency within the homogeneous width of the line. All basic results on frequency stabilization have been obtained with the help of the saturated absorption method, so that the properties of nonlinear resonances and the results obtained will be studied in detail below.

The two-photon resonance method[22] is based on the elimination of the Doppler shift accompanying the absorption of two counterpropagating photons. The two-photon resonance has properties that are important for applications: the recoil effect is absent and all atoms, irrespective of their velocity, contribute to the resonance.

The method of separated fields makes it possible to increase efficiently the interaction time of the particles interacting with the field and to obtain extremely narrow resonances. This method was developed in the microwave range[5] and is the basis for the cesium standard. Because of the Doppler effect, however, for a long time it appeared that this method could not be used in the optical range. The method of separated optical fields in which the Doppler effect is eliminated by phenomena of the echo type was proposed in 1976 in Ref. 23. The last two methods are important for obtaining extremely narrow resonances with a relative width of 10^{-13}—10^{-14}. The potential possibilities of these methods have already been demonstrated in a number of studies.[24–33] However, they are not widely used and results of practical importance have not been obtained because of the difficulties in obtaining stable as well as frequency-tunable radiation and in recording very low absorption.

3.2. Saturated absorption resonances and their properties

A saturated absorption resonance can be recorded both by recording the change in the properties of the radiation passing through an absorbing medium (resonances of the radiation intensity, resonances of the refraction coefficient,[34–36] resonances of the absorption of a weak counterpropagating wave,[37] and the polarization method[38]) and by directly detecting the absorbed energy (fluorescence resonances[39,40] and optothermal and optoacoustical detectors[41,42]). It is difficult to give preference to any one method *a priori*. Extremely high sensitivity can be achieved with selective photoionization of excited particles.

The coefficient of absorption in the field of a standing wave in the presence of weak saturation has the form[18]

$$\alpha=\alpha_0\left[1-\frac{\varkappa}{2}\left(1+\frac{\Gamma^2}{\Gamma^2+\Omega^2}\right)\right]e^{-\Omega^2/\Delta\omega_D^2},\quad \varkappa\ll 1, \qquad (4)$$

where $\alpha = 4\pi^{3/2}d^2\Delta N/\hbar v_0$ is the unsaturated absorption coefficient, $\Omega = \omega - \omega_0$ is the detuning of the frequency of the field ω from the center of the line ω_0, $\Delta\omega_D$ is the Doppler line width, $\varkappa = 4d^2E^2\hbar^2\Gamma^2$ is the absorption saturation parameter, d is the dipole moment of the absorbing transition, ΔN is the difference of the populations of the working levels in the absence of a field, and $2E$ is the amplitude of the field. It is evident from (4) that the dip in the Doppler contour at the center of the absorption line has a homogeneous width 2Γ and an amplitude $-\alpha_0\varkappa/2$, proportional to the intensity of the field. The form of the saturated absorption resonance in the strong field of a standing wave is studied in Refs. 43–45.

The intensity of the nonlinear resonance accompanying saturation of the absorption in the field of a standing wave can be written in the form

$$\Delta I = q\alpha_0\left(\frac{cE^2}{8\pi}\right)lS, \qquad (5)$$

where l is the absorption length, S is the cross section of the light beam, and the coefficient q determines the relative depth of the Lamb dip. The optimal value of the field for obtaining the deepest dip (maximum intensity of the resonance ΔI_M) is achieved when the saturation parameter $\varkappa\simeq 1.4$, which corresponds to $q = 0.13$.[9] Taking into account the fact that $\alpha_0 = 4\pi^{3/2}d^2\Delta N/\hbar v_0$, $E^2 = 1.4\hbar^2\Gamma^2/d^2$, $\Gamma = (\partial\Gamma/\partial p)p$ (p is the gas pressure) and introducing the required normalization factors we obtain from (5) an expression for the absolute intensity of the resonance

$$\Delta I_M[L\mathrm{w}] = 10^2\frac{(\partial\Gamma/\partial p)^2[\mathrm{MHz/torr}]\,p^3[\mathrm{torr}]\,l\,[\mathrm{cm}]\,S\,[\mathrm{cm}^2]\,\xi}{(T\,[K]/m(\mathrm{amu})^{1/2}}, \qquad (6)$$

where ξ is the relative fraction of the particles in the absorbing level. It is evident from (6) that when the gas pressure is lowered the intensity of the resonance rapidly drops proportionally to the cube of the pressure (or $\sim\Gamma^3$). For example, for methane ($\lambda = 3.39\ \mu$m) with $\Gamma\sim 1$ kHz, $l\sim 1$ m, $S\sim 1$ cm^2, $T = 300$ K, $\xi = 0.01$ we have $\Delta I_M \sim 10^{-12}$ W, which corresponds to the average detection capability of photodetectors. When an internal absorbing cell is used the intensity of the resonance depends substantially on the ratio of the saturation parameters of the amplifying and absorbing media and the operating states of the laser.

The first experiments on the use of saturated absorption resonances for frequency stabilization were carried out in an He-Ne laser at $\lambda = 0.63\ \mu$m with a Ne absorbing cell.[10] The advantage of the saturated absorption method lies in the fact that very low pressures of the absorbing gas, when the homogeneous absorption line width and therefore the width of the power peak can be small, can be used. An especially important idea[10,19,21] was the idea of using for the nonlinear absorber the vibrational–rotational transitions (VRT) of molecules and, in particular, the $F_2^{(2)}$ absorption line of methane ($P(7)$, ν_3), whose position coincides with the gain line of the He-Ne laser at $\lambda = 3.39\ \mu$m. Frequency stabilization of the He-Ne laser to the nonlinear resonance in methane was first realized in Ref. 46. The long lifetime and the high coefficient of absorption in VRT of molecules in the ground state made it possible to obtain resonances with a relative width of 10^{-10}–10^{-11} and to achieve great progress in developing lasers with high frequency stability and reproducibility.[9] The narrowest resonances with an absolute line width of less than 1 kHz were obtained in methane on the $F_2^{(2)}$ line of the $P(7)$ transition in the ν_3($\lambda = 3.39\ \mu$m) band in an He-Ne/CH$_4$ laser with an external[47] and internal[48] telescopic beam expander.

We shall study briefly the main physical factors determining the width and shift of nonlinear resonances, which is important for building optical frequency standards. In a low-pressure molecular gas, when the mean-free path length of the particles is of the order of the transverse dimensions of the light beam, the width of a resonance is determined primarily by collisions and transit effects.

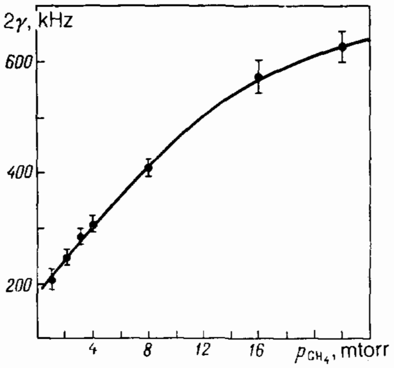

FIG. 1. The width 2γ of a resonance in methane at $\lambda = 3.39$ μm as a function of the methane pressure.[49]

3.2.1. Collision broadening and shift of the resonance

At high pressures of the absorbing gas the broadening and shift of the spectral lines for the electronic and vibrational-rotational transitions depend linearly on the particle density and are of the order of ~ 10 MHz/torr and ~ 1 MHz/torr, respectively. The width of a narrow saturated absorption resonance at low pressure varies nonlinearly as a function of the gas density,[49-54] and the shift decreases sharply at low pressures.[55] Figure 1 shows the experimentally observed pressure dependence of the width of the resonance in methane on the $F_2^{(2)}$ line.[49] In the pressure range 1–5 mtorr the slope of the curve is equal to 30 ± 2 MHz/torr. As the pressure is raised the slope decreases, and at a pressure of ≈ 20 mtorr it is equal to 10 ± 5 MHz/torr. At these pressures the broadening of the resonance coincides with the Doppler broadening of the contour.[56] The qualitative picture of the observed phenomenon is determined by the elastic scattering of particles by small angles without phase interruption.[49]

Figure 2 shows the characteristic nonlinear dependence of the collisional shift of the resonance to methane in the E line (the $P(7)$, ν_3 transition).[57] For gas pressures in the range 6–8 mtorr the magnitude of the shift is ≈ 300 Hz/mtorr. The shift decreases rapidly with the gas density. At a pressure of ≈ 1 mtorr the slope of the shift is equal to ≈ 30 Hz/mtorr. The rapid drop in the shift of the resonance at low pressures makes it possible to achieve high frequency reproducibility. For example, at a pressure of ≈ 1 mtorr it is sufficient to maintain the pressure constant to within 10% in order to ensure a frequency reproducibility at a level of $3 \cdot 10^{-14}$. The theory of resonance broadening and shift in a gas at low pressure[58,59] is in agreement with experiment.

3.2.2. Transit effects

In low-pressure absorbing gases the coherent interaction time of the particles interacting with the field is largely determined by the transit time of the particles through the light beam. When the interaction of the field with the gas is nonlinear, the contribution of the particles to the formation of the resonance depends on their transverse velocity v. As the pressure is lowered and therefore the homogeneous line width 2Γ is decreased, the contribution of the particles with low velocities to the saturation of the absorption will increase. The magnitude of this effect is determined by the parameter $\beta = \Gamma\tau$, which characterizes the ratio between the collisional width 2Γ and the width determined by the finite transit time of particles with a thermal velocity v_0 through the light beam $\tau = a/v_0$ (a is the beam radius). For $\beta \ll 1$ (transit region) the slow particles will determine the form of the resonance, which decreases its width. The first observation of narrowing of a resonance in the transit region was made in methane on the $F_2^{(2)}$ line.[60] The theoretical questions regarding the influence of transit effects on the form of the saturated absorption resonance are studied in Refs. 61–64.

A detailed analysis of the behavior of the width of the resonance and its derivative[1] for a light beam with a Gaussian profile was carried out in Ref. 62. In the region $\beta \gg 1$ the half-width of the resonance γ is determined primarily by the homogeneous half-width of the line Γ:

$$\gamma = \Gamma\left(1 + \frac{2.5}{\beta^2}\right). \tag{7}$$

For $0.5 \leqslant \beta \leqslant 2$ we have $\gamma = \Gamma + 0.58/\tau$. The correction $0.58/\tau$ to the homogeneous half-width of the line determines the contribution of transit effects to the broadening of the resonance. In the transit region, when $\beta \ll 1$, the half-width of the resonance is described by the expression

$$\gamma = 1.51\,\frac{\beta^{1/2}}{\tau}. \tag{8}$$

When the pressure (i.e., β) is lowered the resonance narrows. The half-width $\tilde{\gamma}$ of the derivative of the resonance with respect to frequency ($\tilde{\gamma}$ is equal to the distance from zero to the maximum of the derivative) when $\beta \ll 1$ is determined solely by the collisional half-width Γ ($\tilde{\gamma} = 1.4\,\Gamma$), and transit effects are manifested only in the change in the slope of the broadening of the resonance as a function of the pressure. Figure 3 shows the results of an experiment[65] studying the behavior of the width of the resonance and its derivative as a function of the pressure in a wide range of variation of the parameter β. The experiments were performed in methane at $\lambda = 3.39$ μm with the help of the He-Ne/CH_4 laser. It is evident that the experimental data are in good agreement with the results of the numerical calculation.[62]

Field-induced resonance broadening in the transit re-

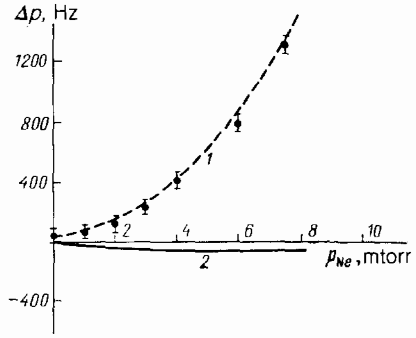

FIG. 2. The shift in the peak of the resonance Δ, in methane on the E line [$P(7)$, ν_3] versus the neon pressure.[57] The methane pressure is equal to 2 mtorr. The broken curve 1 shows the shift of the resonance in methane on the E line, and curve 2 shows the predicted shift of the peak of the resonance produced by the second-order Doppler effect.

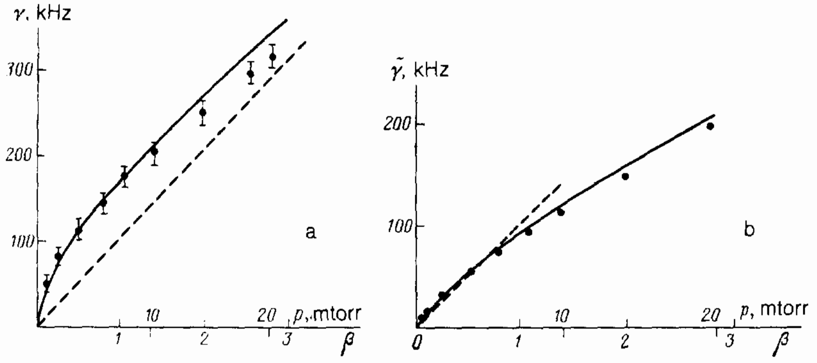

FIG. 3. The half-width of the resonance γ and its derivative $\tilde{\gamma}$ versus the parameter $\beta = \Gamma\tau$.[65] The dots are the experimental values; the solid curve was computed theoretically; the broken curve corresponds to purely collisional broadening, $a = 0.08$ cm.

gion was studied in Ref. 66. The behavior of the resonance in the saturated coefficient of refraction was studied in Ref. 67. As in the case of the derivative of the absorption resonance, when $\beta \ll 1$ the half-width of the resonance in the coefficient of refraction is proportional to Γ.

The transition to low pressures is accompanied, however, by a rapid drop in the intensity of the resonance, which becomes the main factor limiting the use of the saturated absorption method for building optical frequency standards. The behavior of the intensity of the resonance in the transit region with weak saturation is described by the simple expression[65]

$$\Delta I = \alpha_0 l \frac{\varkappa\beta^2}{2} \ln\frac{1}{\beta} \frac{cE^2}{2\pi} \pi a^2. \tag{9}$$

It is evident from (9) that as the gas pressure (i.e., β) is lowered the intensity of the resonance rapidly drops proportionately to p^5. The rapid drop in the intensity of the resonance in the transit region makes it virtually impossible to use the resonance narrowing effect for achieving very low line widths of ~ 1 kHz. The intensity can be increased and the width of the resonances in a low pressure gas can be decreased by using telescopic beam expanders.[47,48]

3.2.3. Other physical factors

For very small relative widths of resonances (10^{-11}–10^{-12}) the form of the resonance will be appreciably affected by physical effects such as the second-order Doppler effect and the recoil effect.

a) *The second-order Doppler effect* is the principal limitation on the width of supernarrow resonances. Because of the spread of the particles over absolute velocities, each molecule will have a resonant frequency which depends on its absolute velocity. For this reason, the line of an ensemble of particles will be additionally broadened by the amount[68]:

$$\Delta_D \approx \frac{1}{2}\left(\frac{v_0}{c}\right)^2 \omega_0 = \frac{kT}{mc^2}\omega_0. \tag{10}$$

For example, the methane molecules at a temperature of $T = 300$ K ($v_0 = 5\cdot 10^4$ cm/s) the broadening of the resonance is equal to 150 Hz ($\Delta_D/\omega_0 \approx 10^{-12}$). When the homogeneous line width 2Γ is less than Δ_D, the inhomogeneous broadening of the line occurs because of the second-order Doppler effect.

The second-order Doppler effect is the most important factor limiting the reproducibility and accuracy of the frequency of optical frequency standards. Because of this effect resonance shifts accompanying a change in the gas pressure and the intensity of the field in the cavity will be observed in the transit region, where the contribution of atoms (molecules) to the intensity of the resonance will depend on their velocity. The general form of the dependence of the shift in the peak of the resonance (for the case of weak absorption) can be written as

$$\Omega_{\max} = \frac{kT}{mc^2}\omega_0 (F_1(\beta) + F_2(\beta)\varkappa). \tag{11}$$

The functions $F_1(\beta)$ and $F_2(\beta)$ characterize the shift as a function of the pressure and the slope of the field-induced shift, respectively, and depend nonlinearly on the value of the parameter β (Fig. 4).[63,66] The shifts are substantially lower for small values of β. Estimates of shifts for methane ($\lambda = 3.39\,\mu$m) with $\Gamma \sim 10^3$ Hz, $\beta \sim 1$ and $\varkappa \sim 1$ give a relative value of 10^{-14} when the field intensity and the gas pressure are varied by a factor of two. To decrease the influence of the second-order Doppler effect heavy molecules must be used, the gas must be cooled, or molecules must be selected according to their absolute velocities.[69]

b) *The recoil effect* splits the nonlinear resonance by $2\delta(\delta = \hbar k^2/2m)$.[70] When the half-width of the resonance γ is greater than δ and splitting does not occur, the recoil effect is manifested as an asymmetry of the resonance contour, determined by the difference in the lifetime of the upper and lower levels of the transition. The field-induced shift of the center owing to the recoil effect appears in a strong field, when coherent effects are important.[71] The magnitude of the

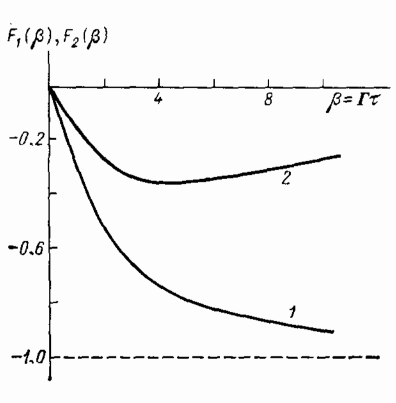

FIG. 4. The shift of the peak of the resonance $\Omega_{\max}/\Delta_D = F_1(\beta)$ (1) and the slope of the field-induced shift $(1/\Delta_D)\partial\Omega_{\max}/\partial\varkappa = F_2(\beta)$ (2) versus the gas pressure owing to the second-order Doppler effect[63] $\Delta_D = (kT/mc^2)\omega_0$.

TABLE I. Effect of different physical factors on the shifts of the resonance in methane on the $F_2^{(2)}$ line.

Parameter	Physical factor	Frequency shift, Hz ($a = 7$ cm, $\varkappa = 0.1$)	
		$p = 10^{-4}$ torr	$p = 10^{-5}$ torr
Pressure (change by a factor of two)	Collisions	3	0.3
	Recoil effect	10	0.3
	Second-order Doppler effect	5	1
	Magnetic hyperfine structure (MHFS)	10	<1
Intensity (change by a factor of two)	Recoil effect	1	0.2
	Second-order Doppler effect	8	0.08
	MHFS	10	0.03
Temperature (change by 1°)	Second-order Doppler effect	0.1	0.01
	Collisions	0.1	<0.01

shift also depends on the relaxation constants of the levels. For vibrational–rotational transitions of molecules the relaxation constants of the levels are determined by the total elastic scattering cross section and are virtually equal to one another. In this case the shifts owing to the recoil effect are small and do not exceed 10^{-15}.

For comparison Table I shows the theoretical and experimental data on the effect of different physical factors on the shifts of a narrow nonlinear resonance for an He-Ne/CH_4 laser with a telescopic light-beam expander.[48] It is evident that at pressures in the range $\sim 10^{-5}$ torr a frequency reproducibility of 10^{-14} is achieved for this laser.

4. THE MODERN OPTICAL FREQUENCY STANDARDS

To obtain maximum long-term frequency stability and reproducibility, supernarrow resonances with a width of ~ 1 kHz and smaller must be used.

The fundamental layout of a modern optical frequency standard (OFS), making use of a supernarrow resonance as the frequency reference, is shown in Fig. 5.[48,72] It includes a frequency-stabilized laser 1 with a narrow radiation line, a tunable laser 2, and a system for obtaining a supernarrow resonance (SNR). The frequency of laser 2 is locked with the help of a phase-locked loop (PLL) to the frequency of the stable laser 1 with a frequency offset determined by the frequency f_{RO} of the radio oscillator. The width of the radiation line of the tunable laser is in this case equal to the width of the radiation line of laser 1. The radiation of laser 2 is used to obtain supernarrow resonances. Smooth tuning of the frequency to the peak of such a resonance is implemented with the help of an extremal automatic frequency control system. To obtain the error signal in the AFC system the frequency of the radio oscillator and therefore the frequency of laser 2 are modulated with the help of the probing signal generator (AG). When the frequency deviates from the center of the resonance an error signal is fed from the output of the AFC into the radio oscillator; this signal tunes the frequency of the radio oscillator (and the frequency of the laser 2) so that the frequency of the laser would coincide with the peak of the resonance. An important advantage of such an OFS system is the possibility of obtaining simultaneously high values of the short- and long-term frequency stability and frequency reproducibility.

5. TECHNIQUES FOR LASER-FREQUENCY STABILIZATION

5.1. Fast frequency-stabilization systems

The frequency of a laser is tuned to the center of a narrow resonance and is held there with the help of an extremal system for automatic frequency control (AFC). The simplest scheme for stabilizing the frequency of a laser is shown in Fig. 6. It includes a frequency reference, with whose help the signal controlling the frequency of the laser is formed; a modulating element, required for obtaining the error signal; the AFC system; and, a controlling element which tunes the frequency of the laser. If the frequency is shifted relative to the maximum of the resonance, then an alternating signal appears in the emitted power at the modulation frequency (the fundamental or first-harmonic signal[2)]). The phase-sensitive detector of the automatic frequency control system, detecting the signal at the first harmonic, generates a

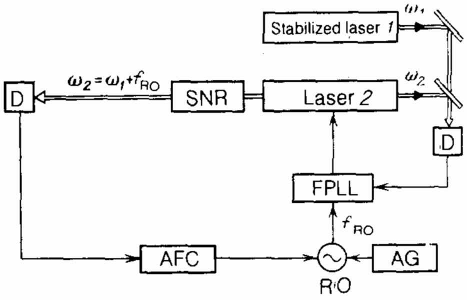

FIG. 5. Scheme of a modern optical frequency standard. D is a photodetector; SNR is the system for obtaining a supernarrow resonance, FPLL is the electronic frequency-phase-locked loop; RO is a radio frequency oscillator; AG is an acoustic generator; AFC is an extremal automatic frequency control system.

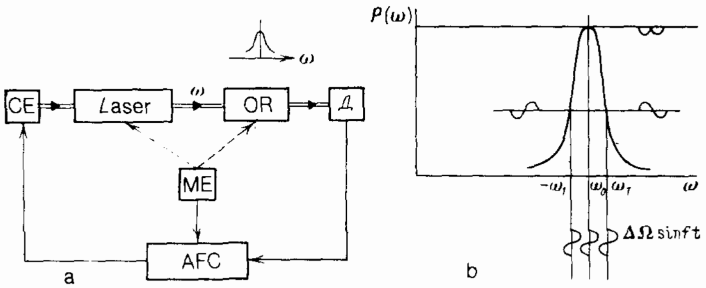

FIG. 6. a) Scheme for stabilizing the frequency of a laser; OR is the optical reference; ME is a modulating element; CE is a controlling element. b) Priciple for obtaining the error signal.

voltage with definite polarity depending on the direction into which the maximum of the frequency is shifted. This voltage is fed into the controlling element of the laser, with whose help the frequency is set at the center of the resonance (the zero of the first-harmonic signal).

The error signal in the AFC system is obtained by the modulation method. The modulation of the laser frequency $\tilde{\omega} = \omega + \Delta\Omega \sin ft$ (f is the modulation frequency and $\Delta\Omega$ is the amplitude of the deviation of the lasing frequency) is most often carried out by a sinusoidal probing signal, which is fed into the controlling element with a mirror. For large widths of the resonance $\sim 10^5$–10^7 Hz, when the modulation index can be large ($m = \Delta\Omega/f \gg 1$), broadening of the radiation spectrum of the laser occurs owing to frequency modulation. In the case when narrow resonances ($\gamma \sim 10^2$–10^3 Hz) are used, the modulation index can be easily made to be small ($m < 1$), which makes it possible to realize the phase-modulation state. This state does not lead to broadening of the radiation line. Another method for obtaining the error signal is based on scanning the resonance absorption contour itself, for example, with the help of magnetic[73] and electric[74] fields. When an external absorption cell is used, phase modulation with the help of a special modulator, placed between the laser and the cell, can also be used.

The laser frequency is tuned with a controlling element by varying the optical length of the cavity by the piezoelectric effect, magnetostriction, the electromagnetic effect, etc.

To achieve high short-term stability the feedback system must have a wide transmission band, substantially exceeding the characteristic range of the frequency perturbations, and a high gain for suppressing rapid fluctuations of the cavity length. For example, in order to obtain a narrow radiation line in dye lasers with a wide perturbation spectrum the bandwidth of the AFC must be equal to ~ 1 MHz. In gas lasers the characteristic width of the spectrum of acoustic perturbations is of the order of 10^2–10^4 Hz. Effective suppression of frequency fluctuations caused by these perturbations requires a feedback bandwidth of 10^4–10^5 Hz. The fundamental limitation on the fast-action of the AFC system is the resonance width 2γ. It determines the magnitude of the modulation frequency $f \lesssim \gamma$ and, therefore, the band of the stabilization system:

$$\Delta f_{\rm p} < f_{\rm AFC} < f \lesssim \gamma,$$

where $f_{\rm AFC}$ is the frequency at which the AFC system has unit gain, and Δf_p is the width of the perturbation spectrum.

Elimination of large thermal drifts of the laser frequency, which is important for obtaining high long-term laser stability, must be achieved with a static gain of the feedback loop

$$K_{\rm st} \geqslant \frac{\delta\omega_{\rm T}}{\delta\omega},$$

where $\delta\omega_T$ is the magnitude of the temperature drift and $\delta\omega$ is the required stability. For example, to obtain a long-term stability of 10^{-14} it is necessary to have $K_{\rm st} \approx 10^9$.

Figure 7 shows a block diagram of a wide-band AFC system for stabilizing the frequency of an He-Ne/CH_4 laser at $\lambda = 3.39\,\mu$m using intense resonances (≈ 1 mW) in methane with a width of 30–50 kHz.[48,75,76] The laser frequency is tuned to the peak of the resonance according to the zero of

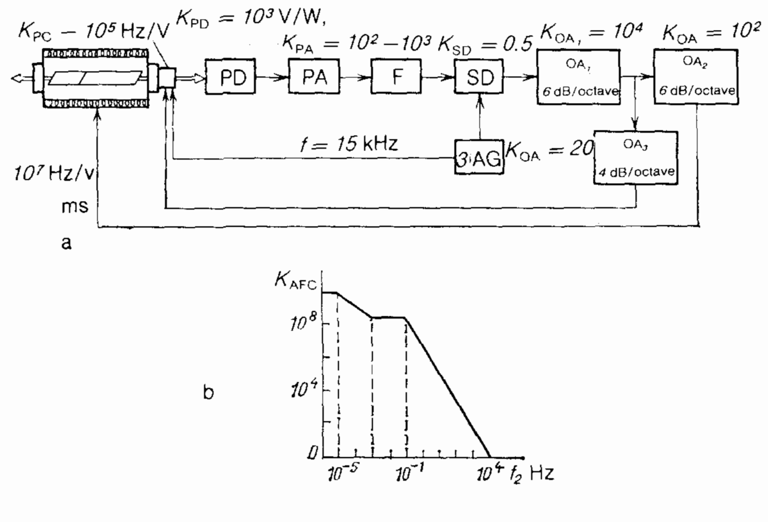

FIG. 7. a) Block diagram of a fast automatic frequency control system (AFC) for an He-Ne/CH_4 laser[48]; PD is a photodetector; PA is a preamplifier; F is a filter; SD is a synchronous detector; OA is an operational amplifier; PC is piezoelectric ceramic; MS is magnetostriction. b) The amplitude-frequency characteristic of the AFC system[48] ($K_{{\rm OA}_2} = 10^2$, $K_{{\rm OA}_3} = 20$).

the fundamental in the emitted power. The modulation frequency f was chosen to be of the order of the half-width of the resonance and in different experiments was equal to $f = 15$–20 kHz. The AFC system had two regulating loops. The slow loop eliminated thermal drift of the cavity and controlled the frequency of the laser with the help of magnetostriction elements in the cavity. The static gain in the loop was equal to $2\cdot 10^{10}$, which made it possible to compensate the thermal drift to a value not less than 10^{-15}. The fast loop of the AFC system was closed on a piezoceramic element with a mirror. The amplitude-frequency characteristic of the AFC is shown in Fig. 7b. Unit gain of the fast loop occurred at a frequency of ~ 10 kHz, and the slope of the amplitude-frequency characteristic for frequencies ranging from 1 to 10 kHz was equal to 32 dB per decade.

5.2. Phase-locking of lasers

In recent years a method for laser-frequency stabilization based on the synchronization of the laser radiation has been developed in the optical range. This method permits transferring frequency characteristics from one laser to another without losses. Once one standard laser is available it is possible by means of successive phase-locking to create an entire series of highly stable lasers in different ranges.[77] In addition, the radiation spectrum of such lasers does not have the frequency modulation which is used to stabilize the frequency of the standard laser. The method of phase-locking is used for making absolute measurements of laser frequencies.

It should be noted that the use of a phase-locking system imposes certain requirements on the noise characteristics of lasers. The phase-locking of two lasers can be realized if the variance of the residual fluctuations of the phase σ_φ^2 of the beat signal of the lasers satisfies the condition

$$\sigma_\varphi^2 = 2\int_0^\infty \frac{S(f)\,\mathrm{d}f}{f^2(1+K(f))} < \frac{\pi}{2}, \qquad (12)$$

where $S(f)$ is the spectral density of the fluctuations of the frequency of the beat signal of the two lasers, f is the frequency of the fluctuations, and $K(f)$ is the gain of the phase-locked loop. An important drawback of the phase-locking system is the absence of automatic locking of the frequencies of the lasers after phase-locking breaks down, for example, because of rapid perturbations of the cavity length. This creates certain difficulties in using phase systems. The use of coupled phase and frequency regulation loops has made it possible to eliminate this restriction and to create a fast electronic frequency-phase-locked loop system[78] (FPLL). The frequency locking system realizes frequency locking of lasers in the presence of perturbations of the laser frequencies that are larger in magnitude than the locking band and holding for the phase regulation loop, and provides automatic phase-locking of the difference frequency of the lasers owing to a decrease in the average intensity of the perturbations. The phase loop must operate as fast as possible and the frequency of the reference radio oscillator must be highly stable. The regulation band of the FPLL system, achieved in Refs. 48, 76, 77, is equal to ≈20 kHz. The precision of phase locking of two lasers achieved in the experiments of Refs. 48 and 76 was no worse than 10^{-15} for averaging times of $\tau \gtrsim 10^{-2}$ s.

5.3. Methods for measuring the frequency stability

The frequency-stability characteristics of lasers are usually measured by recording the difference frequency of two independently stabilized lasers. In the case when the lasers are stabilized to the same quantum references, the average value of their frequency difference is close to zero. This makes it difficult to measure the long-term frequency stability and reproducibility. In addition, because of the scattering of light by the optical elements of the system, a strong interaction of the emissions from the lasers can occur in the region of zero difference frequency. It is therefore necessary to shift the frequency of one laser relative to the other. In Ref. 79 an acoustic modulator, which permitted shifting the frequency of one of the lasers by ≈30 MHz was used for this purpose.

A more universal method is the method of phase-locking of laser radiation which yields virtually any relative shift of the laser frequencies. Figure 8 shows the currently used scheme for measuring the characteristics of laser stability.[48,80] It includes two independently stabilized lasers and an auxiliary laser. With the help of the frequency-phase-locking system the frequency of the auxiliary laser is locked to the frequency of one of the stable lasers. At the same time the difference of the frequencies of the lasers $\omega_2 - \omega_1$ is set equal to the frequency f_{RO} of the reference radio oscillator. The frequency difference $\omega_3 - (\omega_1 + f_{\mathrm{RG}})$ between the other stable laser and the auxiliary laser is measured.

Different apparatus is used to measure the characteris-

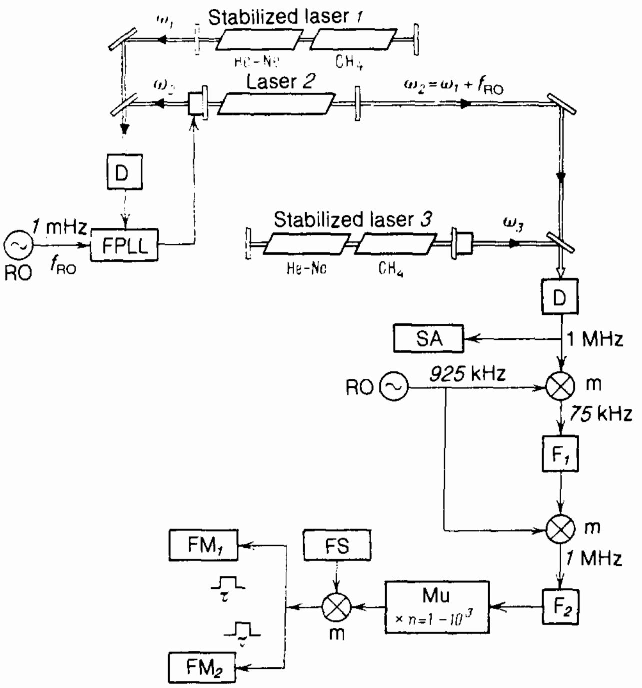

FIG. 8. Scheme for measuring the short-term frequency stability.[76] M is a mixer; MU is a frequency multiplier; FM is a frequency meter; FS is a frequency synthesizer; SA is a spectrum analyzer.

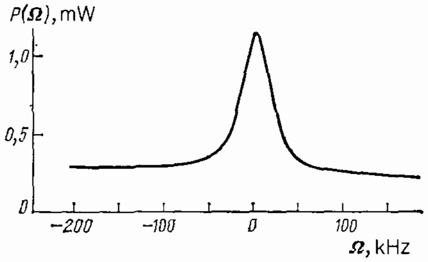

FIG. 9. Typical tracing of the power resonance of an He-Ne/CH_4 laser with a long absorbing cell.[48] The methane pressure is equal to 10^{-3} torr.

tics of the stability of laser frequencies. A simple and the most widely used method for evaluating the stability is the use of an electronic frequency meter. For example, Allan's variance (see Sec. 2) is measured with the help of two frequency meters, which are connected in series with a delay, equal to the averaging time τ. When high short-term stability of $\sim 10^{-14}$ is achieved, however, this method cannot be used for obtaining a reliable estimate of the stability when $\tau < 1$ s. Figure 8 shows the scheme used in Ref. 76 to measure the short-term stability with a relative accuracy of 10^{-14}–10^{15}. The beat frequency of the two lasers, equal to 1 MHz, was fed into a mixer, and then the frequency of the radio oscillator (925 kHz) was subtracted from this frequency. The resulting frequency was passed through a filter with a 3.5-kHz band, tuned to a frequency of 75 kHz. The side components of the frequency-modulated spectrum of the laser were not transmitted into the input of the second mixer. Then the frequency of the radio oscillator (925 kHz) was added to the second mixer. The frequency obtained (1 MHz) was fed through the filter into the frequency multiplier ($n = 1 - 10^3$). After multiplication and subtraction of the frequency of the auxiliary frequency synthesizer, the signal was measured by two serially connected electronic frequency meters for different averaging times $\tau = 1$–10^{-3} s. The delay time was equal to the averaging time τ (counting time).

The "Luch" 16-channel automated meter, which has an output to a display, is a universal instrument for studying the characteristics of frequency stability.[81] The operational principle of this device is based on the statistical accumulation and processing of information on the behavior of the laser frequency. This device has three operating modes. In the first mode the average value of the difference frequency of the lasers is measured, i.e., the apparatus operates as a frequency meter. The second mode is used to obtain on an electronic graphical display histograms of the distribution of the average value of the difference frequency of the lasers with different averaging times ranging from 10^{-4} to 10 s. The third mode is used to obtain histograms of the distribution of the rate of change of the difference frequency of the lasers. The device has a resolution of 32 Hz.

A new step in the development of high-precision methods for measuring the characteristics of frequency stability is the use of microprocessor-based digital automated systems,[82] which make it possible to record the signal studied as a function of time, to process the signal on a computer, and to determine all the necessary statistical characteristics of the laser radiation.

The form of the radiation line is usually recorded with the help of a spectrum analyzer. When a radiation line width of less than 1 Hz is achieved, a convenient method for recording it is to record directly the zero beats of the frequencies of two lasers and to reconstruct the signal spectrum by means of Fourier processing. The spectral density of the fluctuations of the laser frequency is usually measured by standard methods and apparatus using a square-law frequency detector.[83]

6. LASERS WITH A NARROW RADIATION LINE

The widely used methods for obtaining a narrow radiation line are based on locking the frequency of the laser to a passively stabilized interferometer. This technique is successfully used to obtain line widths of ~ 100–1000 Hz.[84,85] Attempts to use this method to obtain narrower radiation lines encounter great technical difficulties, associated with the stabilization of the length of the interferometer itself. For this reason, to obtain narrow lines with a width of ~ 1 Hz it is preferable to use intense and narrow nonlinear resonances.

The best results in this direction have been obtained with an He-Ne laser at $\lambda = 3.39\ \mu$m with an internal methane absorption cell. The technical simplicity of the laser makes it very accessible and convenient for widespread use. Since the absolute frequency of this laser is known with great precision, it can be used as an independent secondary frequency reference for measuring absolute frequencies both in the visible and IR ranges.

The optimum resonance for short-term frequency stability is a resonance with a width of ≈ 50 kHz. An He-Ne laser with a long absorbing cell was used in Refs. 55 and 76 to obtain intense resonances in methane on the $F_2^{(2)}$ line with this width. This made it possible to increase the absorption with low methane pressure. Increasing the diameter of the light beam with a simple cavity geometry made it possible to match the saturation parameters in amplifying and absorbing cells and to increase the radiation power. At a methane pressure of $\sim 10^{-3}$ torr resonances in the laser radiation with a width of 30–50 kHz, a contrast exceeding 70%, and an intensity of ≈ 1 mW were obtained (Fig.9). Figure 10

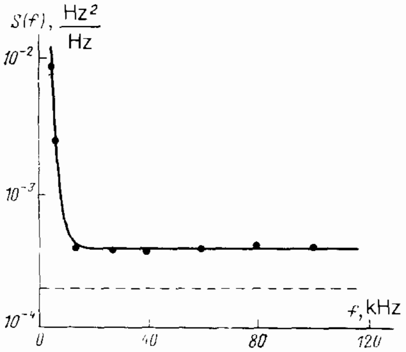

FIG. 10. The spectral density $S(f)$ of the fluctuations of the frequency of an He-Ne/CH_4 laser.[18] The broken line is the computed value of the photon noise of the laser radiation.

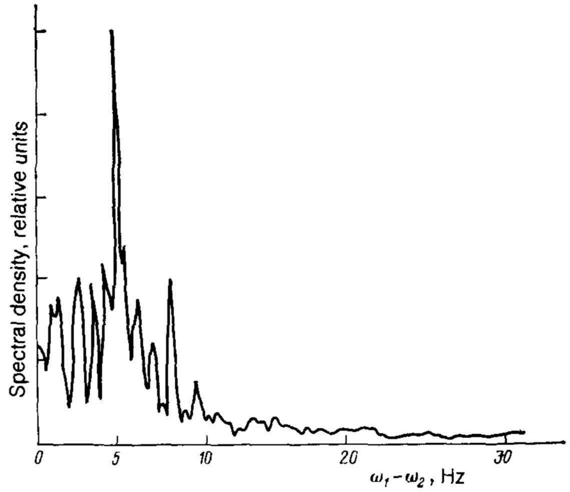

FIG. 11. Beat spectrum of the frequencies of two independently stabilized He-Ne/CH_4 lasers.[11]

shows the spectrum of the fluctuations of the frequency of this laser. It is evident that at frequencies above 5 kHz there are no acoustic and mechanical perturbations of the frequency. The level achieved for a spectral density of $\approx 10^{-4}$ Hz^2/Hz is determined solely by the noise of the photodetector and the fluctuation noise of the photons in the laser radiation. The frequency of the laser was stabilized to the peak of the resonance in methane with the help of fast-acting electronic AFC systems (see Fig. 7).

Figure 11 shows the characteristic spectrum of the beat frequencies of two independently stabilized He-Ne/CH_4 lasers. It consists of a narrow part and a wide plateau, which is associated with the effect of amplitude and phase fluctuations. The width of the radiation line of one laser was equal to 0.07 Hz (the relative width is $7\cdot10^{-16}$). The frequency stability versus the averaging time τ is shown in Fig. 12. For short averaging times $\tau = 1$–100 ms the frequency stability is equal to 10^{-13}–10^{-14} and improves as τ is increased. For $\tau = 1$–100 s the relative value of Allan's parameter is equal to $4\cdot10^{-15}$. For comparison Fig. 12 shows the frequency stability characteristics of a number of microwave standards according to the data in Ref. 86. For short averaging times (1–100 ms) the frequency stability of the He-Ne/CH_4 laser is more than two orders of magnitude higher than the frequency stability of the best masers, and for long averaging times the characteristics of the laser described are comparable to that of the hydrogen maser. Thus the He-Ne/CH_4 laser is currently the most monochromatic source of coherent radiation. Radiation from two such lasers permits observing an interference pattern which varies slowly in time (Fig. 13).[11]

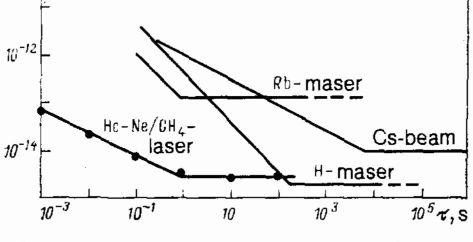

FIG. 12. Allan's parameter $\sigma(\tau)/\omega$ versus the averaging time τ.[76]

It was proposed in Ref. 87 that the counterpropagating waves in a ring laser with nonlinear absorption be used to reduce the width of power resonances. Basov *et al.*[87,88] obtained competing resonances in methane with a width of 30 kHz, which is ten times smaller than the homogeneous width. The use of these power resonances of an He-Ne/CH_4 ring laser as a frequency reference enables obtaining a short-term frequency stability of $5\cdot10^{-14}$ ($\tau = 10$ s). Because of the complexity of the competing lasing states, however, it is difficult to ensure high reproducibility of the frequency of a ring laser.[89]

A narrow radiation line with a width of about 10 Hz for a two-mode He-Ne/CH_4 laser was obtained in Ref. 36 with the use of a resonance in the coefficient of refraction as the frequency reference. Frequency resonances in a single-mode He-Ne laser with a methane cell were first observed and used for frequency stabilization in Refs. 34 and 35. The main advantage of the use of a two-mode lasing state is that an auxiliary laser heterodyne is not required in order to separate out the frequency resonance.

We note that in the widely used single-frequency He-Ne/CH_4 lasers with a cavity length of ~1 m, when the width of the resonance is equal to ~200–400 kHz while the contrast is ~1%, the achieved short-term frequency stability usually does not exceed 10^{-11}–10^{-12}. Figure 14 shows an external view of a small transferable He-Ne/CH_4 laser, developed in Ref. 90, with a relative emission line width no worse than 10^{-12}; this laser was demonstrated at the Exhibition of the Achievements of the National Economy of the USSR.

Using the method of phase-locking of laser radiation and the technique of nonlinear frequency conversion the high frequency stability of the He-Ne/CH_4 laser can be transferred to other wavelength ranges. A CO_2 laser with a relative line width of 10^{-14}, which was achieved by means of frequency-phase-locking of the third harmonic of its radiation to a He-Ne/CH_4 laser, was reported in Ref. 91.

To obtain supernarrow resonances in the visible region of the spectrum by the methods of separated optical fields, two-photon resonances, and particle confinement it is necessary to have tunable lasers with a narrow radiation line and high frequency stability. In this connection the stabilization of continuous dye lasers is of interest. The technique of lock-

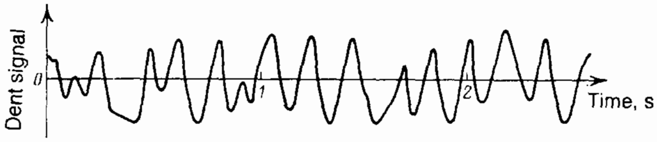

FIG. 13. Characteristic tracing of the zero beats of the frequencies of two independently stabilized He-Ne/CH_4 lasers.[11]

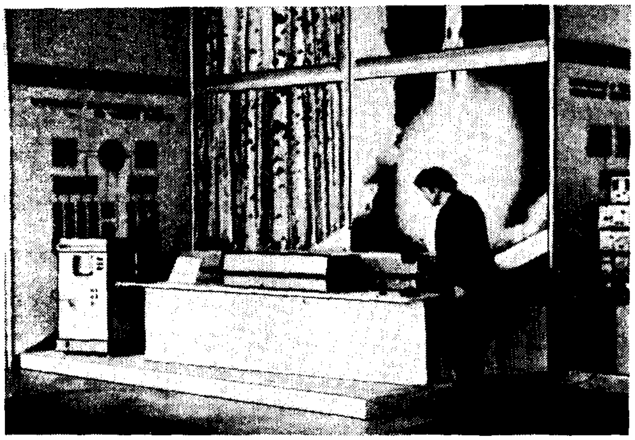

FIG. 14. External view of a transferable frequency-stabilized He-Ne/CH_4 laser with a narrow emission line.[90]

ing the frequencies of lasers to a reference calibration interferometer, based on heterodyning of the side components of the FM signal passing through the standard, is widely used for obtaining narrow radiation lines in tunable lasers.[92–94] The modulation frequency, in this case, must be higher than the characteristic transmission band of the interferometer and the characteristic width of the spectrum of amplitude fluctuations of the laser radiation, while the modulation index must be small. The 30-Hz width of the radiation line of a dye laser obtained in Ref. 94 by this phase method is five orders of magnitude smaller than the starting width of the line and is limited by the instability of the reference standard itself.

7. LONG-TERM FREQUENCY STABILITY AND REPRODUCIBILITY

Different physical factors, such as the collisional shift, the second-order Doppler effect, the recoil effect, external electric and magnetic fields, and the curvature of the wave front, affect the shifts of the frequency of the center of the nonlinear resonance and determine the long-term stability and reproducibility of the frequency of gas lasers. Under real conditions the effect of these factors is manifested when the gas pressure in the absorbing cell, the intensity of the field in the cavity, the geometry of the cavity, the external fields, and so on are varied. A large number of experiments on studying the shifts of saturated-absorption resonances and on obtaining high values of the long-term frequency stability of IR and visible range lasers has now been performed. Here we shall study only the basic results. Special attention is devoted to the He-Ne/CH_4 laser. The highest values of the long-term frequency stability[76] have been obtained for this laser, and detailed studies of the effect of different factors on the shifts of the resonances in methane have been performed.[95]

7.1. He-Ne laser with a CH_4 cell

He-Ne/CH_4 lasers stabilized to the $F_2^{(2)}$ and E absorption lines of methane were studied. It is preferable to use the narrow resonances on the E line to obtain high values of the frequency reproducibility, because the E line is a single line, unlike the $F_2^{(2)}$ line which has a hyperfine structure. The advantage of the E line is gained by using comparatively broad resonances (~ 100 kHz) in simple laser structures. To obtain narrow resonances ($\gamma \sim 1$ kHz) in telescopic systems, when the magnetic hyperfine structure (MHFS) of the $F_2^{(2)}$ line is completely resolved, both lines can be used equally for obtaining high reproducibility.

7.1.1. Use of resonances with a relative width of 10^{-9}–10^{-10}

The $F_2^{(2)}$ absorption line of methane is most widely used for frequency stabilization. It is much simpler to obtain resonances in methane on this line than on the E line, because it is not necessary to use an additional magnetic field in order to shift the gain line. When using resonances with a width of ~ 10–100 kHz, the main factor determining the reproducibility of the frequency of the He-Ne/CH_4 laser is MHFS. The $F_2^{(2)}$ line has three strong components of the MHFS with slightly different intensities and two weak components. The interval between the main components is equal to ≈ 11 kHz.[48,84] When the homogeneous width of the line is large, the components of the hyperfine structure are not resolved and the total contour of the resonance is asymmetrical. When the density of the field in the cavity and the gas pressure in the cell are varied, the relative intensities and widths of the MHFS components also vary, which gives rise to a shift in the maximum of the resonance. For resonance widths of ~ 100 kHz the MHFS limits the reproducibility of the frequency to a level of 10^{-11}.[46,96] Detailed theoretical[97,98] and experimental[95] studies have shown, however, that under certain conditions when the half-width of the resonance (10–30 kHz) is comparable to the distance between the components of the MHFS in methane, its effect on the shift of the resonance is small. Figures 15–17 show the measurements[95] of the shifts of the peak of the resonance in methane and of the stabilized frequency as a function of the field intensity, the methane pressure, and the external magnetic field. It should be noted that when measuring shifts of the stabilized frequency substantial difficulties arise in eliminating the additional modulation of the radiation power at the frequency of scanning of the cavity length. This modulation appears because of the angular displacements of the piezoelectric element with the mirror and because of self-focusing of the radiation in the absorption cell.[95]

a) Field-induced shift. The dependence of the slope of the field-induced shift of the resonance peak has a nonlinear character (Fig. 15a). At low methane pressures ≈ 0.5 mtorr ($\gamma \approx 16$ kHz) the shift is small, and a reproducibility of the frequency of the He-Ne/CH_4 laser better than 10^{-13} can be obtained. When the pressure is raised, the field-induced shift increases substantially. At high methane pressures ($\gamma > 40$ kHz) there appears an additional shift of the peak into the red region, owing to the effect of cross resonances, which appear because of the presence of weak components of MHFS in $6 \rightarrow 6$ and $7 \rightarrow 7$ transitions. In this pressure range the magnitude of the field-induced shift coincides with the data of Refs. 96 and 99. The experimentally observed dependence is in good agreement with the predicted dependence (broken curve).

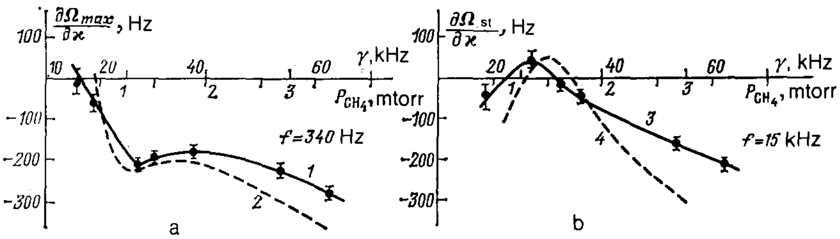

FIG. 15. The slope of the field-induced shift of the peak of the resonance in methane on the $F_2^{(2)}$ line $\partial\Omega_{max}/\partial\varkappa$ (a) and of a frequency-stabilized He-Ne/CH$_4$ laser $\partial\Omega_{st}/\partial\varkappa$ (b) versus the methane pressure.[95] Curves 1 and 3 are the experimental curves; 2, 4) are the computed curves; γ is the half-width of the resonance of a separate component of the MHFS (γ = 10 kHz + 15 [kHz/mtorr] P_{CH_4} [mtorr]).

The behavior of the slope of the field-induced shift of the stabilized frequency (f = 15 kHz, $\Delta\Omega$ = 1 kHz) is shown in Fig. 15b. In the pressure range 1–2 mtorr (γ = 20–40 kHz) variation of the saturation parameter from 0 to 1 gives rise to shifts in the range ± 30 Hz, and the slope of the field-induced shift passes twice through zero. For a saturation parameter of $\varkappa \approx 0.1$ a change of the field intensity by a factor of two gives rise to a frequency shift in the range ± (2–3) Hz. When the width of the resonance is increased ($\gamma > 50$ kHz) the field-induced shift grows substantially and is largely determined by the cross resonances.

b) *Pressure-induced shift.* The nonlinear behavior of the shift in the peak of the resonance in methane accompanying a change in the gas density (see Fig. 16) is determined by the combined effect of the magnetic hyperfine structure and the collisional shift. Figure 16a shows the curve of the predicted shift of the resonance peak owing to MHFS versus the methane pressure. The difference between the experimental and predicted curves shows the dependence of the collisional shift on the methane pressure (see Fig. 16b). The behavior of this shift is in good agreement with the results of direct measurements of the collisional shift in methane for the E line (see Sec. 3). At low pressures of ≈ 1 mtorr the shift in the resonance peak is small and constitutes ≈ 10–20 Hz/mtorr. In this pressure range the collisional shift is compensated by a displacement of the resonance peak owing to the effect of MHFS, which gives rise to a weak pressure dependence of the resulting shift. Therefore, for pressures in the range of ~ 1 mtorr the magnetic hyperfine structure not only does not prevent obtaining high-frequency reproducibility, but actually makes it much easier to obtain high reproducibility by reducing the effective shift of the resonance as a function of the pressure. The absolute shift in the frequency of the stabilized He-Ne/CH$_4$ laser with a long absorption cell, measured in Ref. 95, relative to the central component of MHFS of the $F_2^{(2)}$ line of methane at pressures in the range ≈ 1 mtorr is equal to 1600 ± 40 Hz and is in good agreement with the calculations.

c) *A magnetic field* has an appreciable effect on the shift of the stabilized frequency of a He-Ne/CH$_4$ laser. Figure 17 shows the results of measurements for the half-width of the resonance in methane γ = 70 kHz, when the field-induced shift owing to MHFS is large. A longitudinal magnetic field with intensity H = 0–50 Oe was applied to the absorption cell. When the intensity of the magnetic field was increased the slope of the field-induced shift of the frequency decreased, and for some value of the magnetic field (H = 45 Oe) the sign of the shift changed (see Fig. 17). The observed picture is linked with the presence of MHFS on the working transition of methane. We note that the effect of the magnetic field depends on the magnitude of the saturating light field in the cavity. Conditions under which a change in the magnetic field does not give rise to appreciable frequency shifts are possible in the region of high saturations.[96] The phenomenon discovered in Ref. 95 is important for improving the reproducibility of the frequency of the He-Ne/CH$_4$ laser under conditions when the width of the resonance in methane is large ($\gamma \sim 100$ kHz). The theoretical calculation of the behavior of the shift of a nonlinear resonance in a magnetic field in the presence of MHFS is in agreement with experiment.[100]

d) *A shift in the nonlinear resonance owing to the curvature of the wave front* appears if the counterpropagating waves forming the standing wave have different intensities.[64,101] It depends on the value of the parameter $2z/b$, where z is the longitudinal coordinate of the point at which the shift is calculated and b is the confocal parameter. The shift of the resonance peak owing to the curvature of the wave front was investigated in Ref. 101 for the case of an external absorption cell, where the magnitude of the shift can be quite high ($\sim 10^{-11}$). For a laser with an internal absorption cell, the shift of the resonance peak appears if the intensities of the counterpropagating waves J_+ and J_- are nearly equal $\Delta J/J = (J_+ - J_-)/J \ll 1$. Estimates of the shifts based on the results of Refs. 64 and 101 showed that under the conditions of the experiment described in Ref. 95, when $\Delta J/J \approx 0.1$; $2z/b \approx 0.2$; b = 30 m; $\gamma = 2 \cdot 10^4$ Hz, and the light beam diameter $2a$ = 1 cm, the resonance peak shifts by an amount ≈ 5 Hz when the pressure is varied by 50%. Therefore, by selecting the configuration of the cavity and

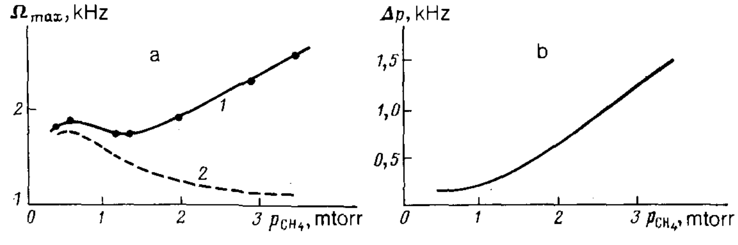

FIG. 16. a) Pressure-induced shift of the peak of the resonance Ω_{max} in methane on the $F_2^{(2)}$ line relative to the central component of MHFS $\omega_{7\to6}$ ($\Omega_{max} = \omega_{7-6} - \omega_{7\to6}$); curve 2 is the computed shift of the maximum of the resonance owing to the effect of MHFS. b) Collisional shift of the resonance in methane.[95]

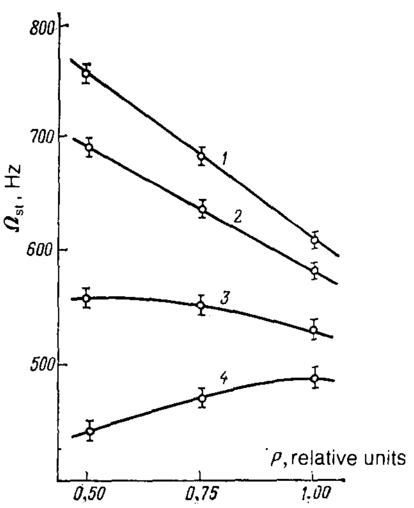

FIG. 17. Dependence of the field-induced shift of a frequency-stabilized He-Ne/CH_4 laser on the intensity of the magnetic field H.[95] H(Oe) = 0 (1), 9 (2), 27 (3), and 45 (4).

the transmission of the mirrors it is possible virtually to eliminate the effect of the curvature of the wave front on the position of the resonance.

Thus with a simple construction an He-Ne/CH_4 laser with a long absorption cell[55,95] makes it possible to obtain high short- and long-term frequency stability of 10^{-14} and higher. This laser is convenient for different spectroscopic studies and for making absolute measurements of laser frequencies.

We should say a few words about the accuracy of the frequency of the He-Ne/CH_4 laser stabilized to the $F_2^{(2)}$ line of methane. For a laser with a simple construction the accuracy of the frequency is usually limited to $\sim 10^{-11}$ because of the effect of MHFS. In the case when the shift of the laser frequency relative to the center of the $F_2^{(2)}$ line of methane is measured with high accuracy,[95] the accuracy with which the frequency is determined is raised substantially. At the present time additional experiments must be performed to resolve the question of the accuracy of the frequency of the He-Ne/CH_4 laser. Factors such as the second-order Doppler effect, the magnetic field, and the divergence of the light beam have virtually no effect on the reproducibility of the frequency, but they can appreciably affect the accuracy, giving a constant shift of the resonance.

7.1.2. Frequency-stabilization to supernarrow resonances

Stabilization of the frequency ω of the He-Ne/CH_4 laser to resonances of width $\sim$ 1 kHz was first realized in Refs. 48 and 72. The scheme for stabilizing the frequency of this laser is shown in Fig. 18. The auxiliary stable laser was a He-Ne laser with a long methane absorption cell, whose radiation line width was less than 1 Hz. To obtain ultranarrow resonances in methane on the $F_2^{(2)}$ line with high enough intensity, a four-mirror He-Ne/CH_4 laser with an intracavity telescopic beam expander (TBE) was used. Frequency stabilization of the laser with the TBE was realized using the peak of the central component (7→6) of the MHFS of the $F_2^{(2)}$ line. Figure 19 shows the typical tracing of the hyperfine structure of the $F_2^{(2)}$ line in the state used for frequency stabilization. The width of the resonance of each component of the structure was equal to 2–3 kHz. Long-term frequency stability of 10^{-14} was obtained with an averaging time of $\tau = 10$ s. The width of the radiation line of the laser was less than 1 Hz.

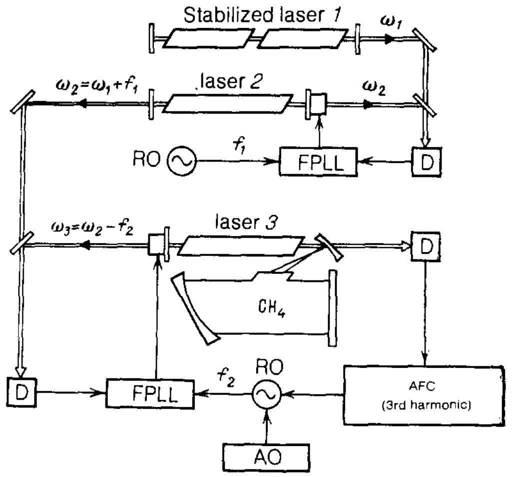

FIG. 18. Scheme of frequency stabilization based on supernarrow resonances.[72] The basic parameters of the He-Ne/CH_4 laser with the telescopic beam expander (TBE) are as follows: the beam diameter is equal to 14 cm, the absorbing cell is 5 m long, and the methane pressure is equal to 60 μtorr.

The shifts of the stabilized frequency accompanying a change in the methane pressure and the field intensity in the cavity for a methane pressure of 60 μtorr were measured in Ref. 72. A twofold change in the field intensity in the region of low saturation ($\varkappa = 0.2 - 0.4$) gave rise to shifts of 1–3 Hz. The observed nature of the shifts is in agreement with the results of the theoretical calculations[102] and is determined by the second-order Doppler effect. In Refs. 48 and 72 it was established that the frequency reproducibility of the He-Ne/CH_4 laser with the TBE is equal to 10^{-14}, which is achieved in a wide range of variation of the methane pressure

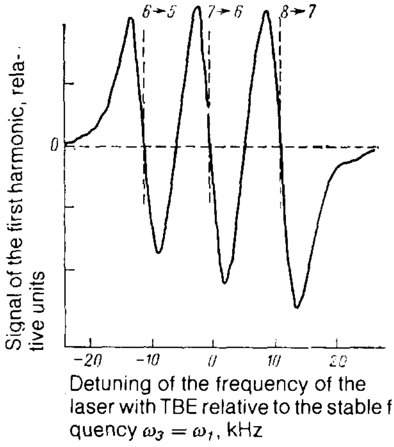

FIG. 19. Typical tracing of the MHFS of the $F_2^{(2)}$ line of methane at a pressure of 60 μtorr.[72] The modulation frequency is equal to 750 Hz, the frequency deviation is equal to 300 Hz, the temperature is equal to 300 K, the integration constant is equal to 2 s, and the recording time is 20 min.

and field in the cavity. It is important to note that the tuning of the frequency of the laser with TBE to the peak of the central component corresponds to tuning to the center of the $F_2^{(2)}$ line of methane, which provides high accuracy of the frequency equal to $\approx 10^{-13}$.

The highly stable telescopic laser developed in Refs. 48 and 72 is a cumbersome and complex system. It is of interest to obtain resonances of width 5–10 kHz in order to build simpler He-Ne lasers with high frequency reproducibility, which can be used efficiently for setting up a number of precision experiments. The single E line of the $P(7)$ transition of the ν_3 band of methane, with no hyperfine structure, is very convenient in this case for frequency stabilization. Resonances on this line were first observed in Refs. 103 and 104. In Refs. 105–107 resonances with a width of ~ 100 kHz were studied in detail and were used for frequency stabilization. Supernarrow resonances with a width of $\sim 10^{-11}$ on the E line in methane were first observed and used as a basis for a highly stable He-Ne/CH_4 laser in Ref. 11. A laser with a telescopic three-mirror beam expander was used to obtain the supernarrow resonances in methane on the E line in Ref. 11 (Fig. 20). The E line of methane is shifted from the center of the gain line of the He-Ne laser by 3 GHz into the red region, so that a transverse magnetic field with an intensity of ≈ 1800 Oe was used to shift the gain line. At a pressure of 100 μtorr the resonances had a width of 5 kHz and an intensity of 500 μW. The use of such resonances for frequency stabilization made it possible to obtain a frequency stability of $6\cdot 10^{-15}$ with averaging times of $\tau = 1$–10 s. Measurements of the shifts of the stabilized frequency accompanying a change in the methane pressure, the field intensity in the cavity, and the magnetic field determined the frequency reproducibility of this laser to be 10^{-14}. The basic factor limiting the frequency reproducibility and the accuracy of the frequency of the He-Ne/CH_4 laser with a TBE is the second-order Doppler effect. The influence of the second-order Doppler effect can be reduced by using heavy molecules, by cooling the particles, and by developing methods of selection based on absolute velocities.

A direct comparison of the frequency stability of He-Ne/CH_4 lasers independently stabilized to the $F_2^{(2)}$ and E absorption lines of methane, was made in Ref. 108. The frequency reproducibilities of these lasers, obtained as a result of the comparison, constituted $3\cdot 10^{-13}$. This made it possible to measure with high accuracy the absolute value of the difference of the frequencies of the $F_2^{(2)}$ and E absorption lines in methane. The result is

$$\omega_{F_2^{(2)}} - \omega_E = 3\,032\,571\,672 \pm 30 \text{ Hz}.$$

In Ref. 109 the frequency of a two-mode telescopic He-Ne/CH_4 laser with orthogonally polarized modes was stabilized and the frequency reproducibility was studied. The two-mode state was created with the help of a Fabry-Perot resonator, containing two phase plates close to $\lambda/4$. The distance between the neighboring orthogonal modes was regulated by the angle between the optic axis of the plate in the range 0.2 MHz and higher. Stabilization was achieved with the use of frequency resonances. The absorption cell was cooled with liquid nitrogen. The magnetic hyperfine structure of the $F_2^{(2)}$ lines of methane was resolved and frequency stabilization of the radiation to separate components was realized with comparatively small dimensions of the laser (cavity length of 1.8 m, cell length of 0.9 m, and beam diameter of 3 cm). A long-term frequency stability of 10^{-14} was achieved (the averaging time was equal to 100 s). The frequency reproducibility was estimated from the measurements of the shifts to be $\sim 10^{-14}$.

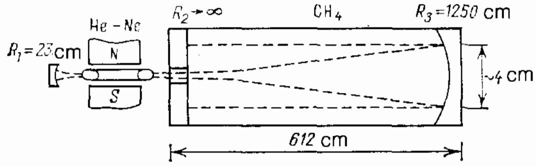

FIG. 20. Diagram of a laser with a three-mirror telescopic beam expander.[11]

7.2. He-Ne laser with an I_2 cell

He-Ne lasers at $\lambda = 0.633\,\mu$m and $\lambda = 0.612\,\mu$m, stabilized to saturated absorption resonances in $^{127}I_2$ and $^{129}I_2$ vapor on the components of the hyperfine structure of the electronic transitions, are now widely used as optical wavelength standards for metrological measurements and spectroscopic studies.[110–122]

Saturated absorption resonances in $^{127}I_2$ on the $R(127)$ 11-5 line[123,124] were first used for stabilizing the frequency of an He-Ne laser at $\lambda = 0.63\,\mu$m with an internal absorbing cell in Refs. 111 and 115. With an I_2 pressure of 0.1 torr the width of the power peak was equal to ≈ 4.5 MHz, and the contrast was equal to 0.1%. The low amplitude of the power resonance and the large slope of the Doppler contour of the gain line at the position of the resonances made it necessary to use the third-harmonic signal for stabilizing the frequency.[125] Frequency stabilization of an He-Ne/$^{127}I_2$ laser at a level of 10^{-11} and a frequency reproducibility of $4\cdot 10^{-11}$ were obtained by this method in Refs. 111 and 115. Detailed investigations of the factors determining the width and shifts of narrow nonlinear resonances in an He-Ne/$^{129}I_2$ laser were performed in Ref. 110. The measured value of the iodine-pressure broadening of the resonance was equal to 13 MHz/torr, and the slope of the shift was equal to 1 MHz/torr. The frequency stability obtained in this work for the He-Ne/$^{129}I_2$ laser was equal to $2\cdot 10^{-12}$ ($\tau = 10$ s) and the frequency reproducibility was equal to 10^{-10}. The highest values of the frequency stability of a $\lambda = 0.63\,\mu$m He-Ne/$^{127}I_2$ laser were obtained in Ref. 126, where a value of $1.9\cdot 10^{-13}$ ($\tau = 270$ s) was obtained. A reproducibility of $8\cdot 10^{-12}$ has been achieved for this laser.[127]

Frequency stabilization of the $\lambda = 0.612\,\mu$m He-Ne laser with an internal $^{127}I_2$ cell (the line $R(47)9$-2) was realized in Refs. 128 and 129. The long-term frequency stability obtained was equal to $2\cdot 10^{-13}$ ($\tau = 1000$ s).[129] An external cell was used in Ref. 130 to obtain absorption resonances in the $R(47)9$-12 line of $^{127}I_2$. To intensify the signal the I_2 cell was placed inside a high-Q Fabry-Perot resonator, whose frequency was locked to the frequency of the laser. The use of an interferometer made it possible to lower the pressure in the cell to 3 mtorr, owing to the increase in the contrast of the resonance to obtain resonances with a width of 200 kHz with a high signal-to-noise ratio. Direct comparisons of two such He-Ne/$^{127}I_2$ lasers gave the following results: a long-term stability of $2\cdot10^{-13}$ ($\tau = 100$ s) and a frequency reproducibility of $6\cdot10^{-13}$ [131]

7.3. Frequency-stabilized CO_2-N_2-He laser

The first work on the stabilization of the frequency of a CO_2 laser lasing at $\lambda = 10.6\ \mu m$ was performed with an internal CO_2 nonlinear-absorption cell.[132] A frequency stability of $4\cdot10^{-11}$ was achieved with an averaging time of $\tau = 100$ s, and the frequency reproducibility was equal to $2\cdot10^{-10}$.

The most significant results on the stabilization of the frequency of a CO_2 laser were obtained with the use of narrow resonances of saturated absorption in the vibrational-rotational transitions of SF_6[133,134] and OSO_4[135,136] molecules. The high absorption coefficient at the transitions of these molecules for a number of lines of the CO_2 laser made it possible to use ion absorbing-gas pressure of 10^{-2}–10^{-3} torr and obtain resonances with a width of ~100 kHz. Frequency stabilization of the $\lambda = 10.6\ \mu m$ (the P (18) line) CO_2 laser to the narrow resonance in SF_6 was first realized in Refs. 137 and 138 by the external nonlinear-absorption cell method with a weak counterpropagating wave.[3] A frequency stability of $5\cdot10^{-14}$ with an averaging time of 10 s[139] and a frequency reproducibility of $3\cdot10^{-11}$ [138] were achieved for the CO_2/SF_6 system.

It is of interest to use the methane-like molecule OSO_4. This molecule does not have a hyperfine structure. The second-order Doppler effect is small owing to the large mass of the molecule, which makes it possible to achieve high long-term frequency stability and reproducibility. The first experiments on the stabilization of the frequency of a CO_2 laser with an external OSO_4 cell were performed in Refs. 136 and 140, where a long-term frequency stability of $\approx 10^{-12}$ and a frequency reproducibility of 10^{-11} were obtained. In Ref. 141 a narrow resonance on the $A\,^2_2P\,(14)$ transition in $^{192}OSO_4$ with a width of 30 kHz and a contrast of ~1% was used for frequency stabilization. A frequency stability of $\approx 10^{-13}$ over a time of $\tau = 10$ s was achieved.

The method of saturated fluorescence is also widely used for stabilizing the frequency of a CO_2 laser.[39,40] The advantages of this method are its simplicity and the possibility of stabilizing the frequency on many lines of CO_2. The best results were obtained in Ref. 142, where a frequency stability of 10^{-12} over a time $\tau = 50$ s was reported.

7.4. Ar^+ laser with an I_2 cell

Stabilization of the frequency of powerful ion lasers is of interest for the development of tunable lasers based on dyes and color centers in crystals. In Ref. 143 it was pointed out that the radiation line of the Ar^+ laser at $\lambda = 5145$ Å coincides with the hyperfine structure of the P (13) and P (15) line of the transition ($v = 0$, $X^1\Sigma$) and ($v = 43$, $B^3\Pi$) in $^{127}I_2$. Narrow saturated-absorption resonances in $^{127}I_2$ were first observed with the help of an Ar^+ laser in Ref. 144. This system was studied in detail in Refs. 145 and 146. In Ref. 146 saturated-fluorescence resonances with a width of ~100 kHz were obtained at low pressure, and a stability of $5\cdot10^{-14}$ with $\tau = 100$ s and a reproducibility of $1.5\cdot10^{-12}$ were achieved. In Ref. 147 a molecular beam was used for stabilizing the frequency of an Ar^+ laser. A long-term frequency stability of $7\cdot10^{-14}$ ($\tau = 10^3$ s) was obtained in this work. The frequency reproducibility of $1.5\cdot10^{-12}$ in Ref. 147 was limited by the accuracy with which the laser beam could be set orthogonally to the molecular beam.

7.5. Frequency stabilization of a dye laser

An intensive search for efficient resonant absorbers for frequency-stabilization of continuous tunable dye lasers has been conducted in recent years. In Ref. 148 a dye laser based on rhodamine 6G was stabilized and a frequency stability of $6\cdot10^{-13}$ over a time of $\tau = 25$ s was obtained using the components of the hyperfine structure of the lines P(13) and R(15) $^{127}I_2$ and the molecular beam technique. The frequency reproducibility for two lasers, independently stabilized to the hypefine "O" components of the line P(62) 17-1 $^{127}I_2$ at $\lambda = 576$ nm, was studied in Ref. 149. In Ref. 149 the frequency shifts versus the iodine pressure in the cell, degree of modulation, and the intensity of the field in the cavity were measured. The frequency reproducibility was equal to $\approx 2\cdot10^{-11}$. The possibility of using the intercombination transition $^1S_0 - {}^3P_1$ in Ca at $\lambda = 6573$ Å to obtain narrow saturated-absorption resonances and frequency stabilization was investigated in Ref. 150.

8. OPTICAL CLOCKS

The development of lasers with long-term frequency stability no worse than for masers and a short-term stability better than that of masers has made it possible to solve the problem of constructing optical clocks, i.e., using the period of the optical oscillation of a highly stable laser as a time scale.[151] The construction of an optical time scale also required solving the problem of transferring the frequency characteristics of lasers into the microwave range without loss of precision. As a result the unit of the time scale, the second, can be determined directly from the number of highly stable optical oscillations.

Figure 21 shows a block diagram of the first optical clock.[6,151] The scheme includes a reference highly stable He-Ne/CH_4 laser at $\lambda = 3.39\ \mu m$ (see Sec. 6), a system of specially selected and phase-locked IR ($\lambda = 3.39$, 10.2 and 10.07 μm) and submillimeter ($\lambda = 70$ and 418 μm) lasers and microwave oscillators (klystrons, Gunn oscillators), which are used to divide the frequency of the He-Ne/CH_4 laser into the radio range with output at the standard frequencies of 1 and 5 MHz. Successive phase-locking of one oscillator to another made it possible to transfer the high frequency stability without losses. Point diodes of the metal-oxide-metal (MOM-diode) type with a time constant of $\sim 10^{-14}$ s were used as fast-acting nonlinear elements for converting the frequencies of the lasers and generating harmonics of high order. The measurement-computational

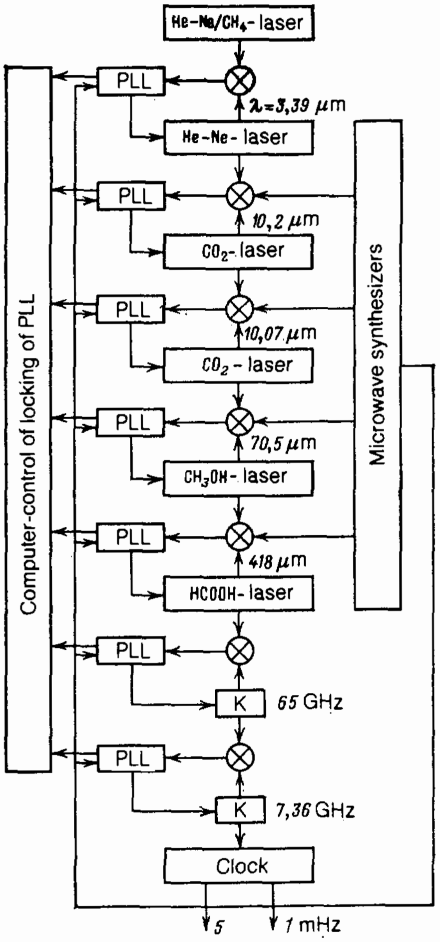

FIG. 21. Diagram of an optical time standard.[6,151] K is a klystron.

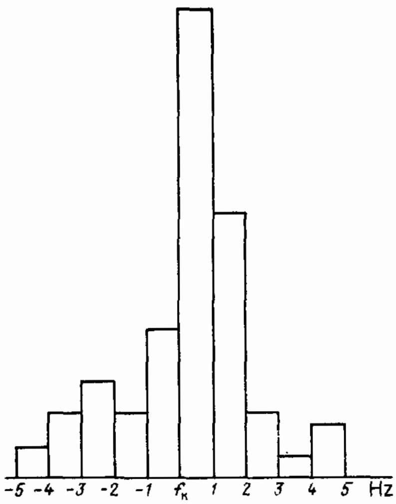

FIG. 22. Histogram of the beat signal between the klystron synchronized with a He-Ne/CH_4 laser and a klystron synchronized with a Rb standard.[151] The klystron frequency is equal to $f_k = 65.1$ GHz and the averaging time is equal to $\tau = 1$ s.

complex monitored and measured the characteristics of the optical clocks.

The development of highly stable microwave oscillations, synchronized with the He-Ne/CH_4 laser, has made it possible to make the first direct comparison of the characteristics of the frequency stability of a rubidium standard and of an He-Ne/CH_4 laser ($\lambda = 3.39$ μm). Figure 22 shows a histogram of the measurements with an averaging time of $\tau = 1$ s. The width of the histogram was determined by the stability of the rubidium standard. The frequency stability of the He-Ne/CH_4 laser was much higher.

Interest in raising the stability and reproducibility of laser frequencies has been stimulated by the development of an optical time scale. It is therefore desirable to present the basic comparative properties of lasers and masers as frequency and time standards.

1. Physical experiments involving the use of lasers for frequency measurements require much shorter times, since the absolute frequency of the lasers is 10^4–10^5 times higher.

2. The absolute intensity and width of the resonances used as references for stabilizing the frequency are 10^5–10^6 times higher in the optical range than in the microwave range with one and the same relative width. This makes it possible to construct optical standards with a short-term frequency stability that is significantly higher than for microwave standards.

3. When the frequency of an optical standard is divided into the radio frequency range, the relative width of the emission line remains virtually unchanged. In the case when a radio-range standard is used, the fluctuation spectrum of its signal is substantially broadened when the frequency is multiplied by a factor of 10^5–10^6.

4. The second-order Doppler effect, as the basic physical factor limiting the long-term frequency stability and reproducibility, plays the same role for lasers and masers. Because of the small absolute contribution to the form of the line, however, its observation and study in the microwave range presents great difficulties against the background of other factors causing the line shift.

The properties enumerated above, in our opinion, certainly indicate the advantages of using lasers as frequency and time standards. Their realization, however, will require some time in order to overcome technical difficulties associated with both the problems of optical frequency standards themselves and the problem of dividing optical frequencies. The system for dividing laser frequencies based on phase-locking is still very cumbersome. The system must be simplified substantially in order for laser-based time standards to become competitive with masers in practice.

9. ABSOLUTE MEASUREMENT OF LASER FREQUENCIES

To measure optical frequencies the known frequency of the standard in the radio range must be multiplied by a factor of 10^4–10^5 or the measured frequency of the laser must be divided by the same factor.

Absolute measurements of laser frequencies were for a long time performed in stages. First, the frequencies of far-infrared lasers were determined by comparing the multiplied signal from a microwave standard with the laser frequency. Then, the known frequency of the laser was once again multiplied and compared with the frequency of the new laser. The frequency synthesis scheme at each stage of the measurement is expressed by the general formula

$$\omega_i = n\omega_{i-1} \pm f_{\text{in}},$$

where ω is the synthesized frequency, ω_{i-1} is the known frequency, and f_{in} is the measured intermediate frequency. For a known frequency multiplication factor n we obtain the absolute value of ω_i.

The first measurements of laser frequencies were performed in Refs. 152 and 153, where the absolute frequencies of the rotational transitions of submillimeter lasers were determined. The lasers were stabilized to the Doppler-broadened lines of the transitions, and the measurement accuracy was equal to $\sim 10^{-8}$. Later the frequencies of the H_2O laser,[154,155] CO_2 laser,[156–158] and the He-Ne laser at $\lambda = 3.39$ μm[156–160] were also measured. The absolute frequencies of the measurements were recently extended into the visible range thanks to measurements of the frequencies of the He-Ne laser at $\lambda = 1.5$ μm and 1.15 μm.[161,162] Using the second harmonic of this laser it will be possible to measure the frequency of an electronic transition in the iodine molecule.[163]

The development of optical clocks has made it possible to measure the absolute frequencies of lasers with extremely high precision. In Refs. 6, 151, and 164 this method was used to perform precise measurements of the frequencies of the He-Ne/CH_4 laser stabilized to the $F_2^{(2)}$ and E absorption lines of methane. In the measurements the eleventh harmonic of the frequency of a microwave oscillator at 65 GHz (see Fig. 21), synchronized with the frequency of the Rb standard, was compared with the frequency of the HCOOH laser (717 GHz), whose radiation was synchronized with the radiation of the He-Ne/CH_4 standard.

Existing setups for measuring the frequencies of infrared and visible range lasers[159,156,160,164,165] are too complicated for extensive applications. The process of measuring optical frequencies can be substantially simplified by using for this purpose a highly stable laser with a precisely measured frequency, which is converted by nonlinear means into the region of the spectrum where the frequency measurements are performed. Such a system will make it possible to perform absolute frequency measurements without using complex setups.

At the present time the frequency of the He-Ne/CH_4 laser at $\lambda = 3.39$ μm has been measured with the highest accuracy. This laser has a high frequency reproducibility and occupies a convenient intermediate position between two regions of the spectrum: the submillimeter and IR regions on one side and the near IR and visible regions on the other. The results of the measurements of the frequencies of the He-Ne/CH_4 laser stabilized to the $F_2^{(2)}$ and E lines of methane are presented in Table II. Unfortunately, the results of the latest precise measurements for the $F_2^{(2)}$ line differ appreciably. The reasons for this disagreement have not yet been fully established. One reason is the magnetic hyperfine structure of the $F_2^{(2)}$ line, which lowers the reproducibility of the laser frequency. The presence of MHFS in the $F_2^{(2)}$ transition of methane gives rise to shifts of the resonance whose magnitude depends substantially on the frequency stabilization mode of the He-Ne/CH_4 laser. For substantially different states the difference in the positions of the laser frequencies can reach ~ 1 kHz (see Sec. 7, Fig. 15). The influence of MHFs is eliminated with the use of complicated setups with a telescopic beam expander, enabling frequency stabilization based on one component of the MHFS[48] or it is performed by careful selection of the operating states of ordinary lasers.[95] In this connection, it is of interest to perform precision measurements of the frequency of a single line, which the E absorption line of methane (the transition P (7) in the ν_3 band) is.

10. APPLICATIONS OF OPTICAL FREQUENCY STANDARDS

Optical frequency standards are now widely used in ultrahigh resolution spectroscopy and in precision physical experiments, and they have practical applications in metrology, radar, geophysics, communications, machine building, space studies, and other areas. In this review it is impossible to consider all applications in detail, so that here we shall merely list some of them.

Increasing the resolution of optical spectroscopy is directly linked with obtaining supernarrow resonances. At the present time the saturated absorption method is the basic and best developed method of ultrahigh resolution spectroscopy. Lasers with a narrow radiation line of width ~ 1 Hz are required in order to record resonances with a relative width of 10^{-11} and smaller. In addition, at low absorbing gas pressures the intensity of the resonance decreases substantially and the resonance must be recorded slowly to accumu-

TABLE II. Results of absolute measurements of the frequencies of an He-Ne/CH_4 laser at $\lambda = 3.39$ μm.

Laser	Absorption line	Measured frequency, kHz	Accuracy of the measurement ± kHz	References
He—Ne/CH_4	$F_2^{(2)}$	88 376 181 625	50	156
		88 376 181 586	10	158
		88 376 181 616	3	159
		88 376 181 603,0	3	6
		88 376 181 603,4	1,4	160
		88 376 181 602,9	1,2	164
	E	88 373 149 031,2	1,2	164

late the signal. For this reason, in order to perform studies with supernarrow resonances, special laser spectrometers with a resolution of 10^{11}–10^{12} must be developed.[48,84,166] A number of physical effects have been observed in the optical range for the first time with the help of such spectrometers: recoil accompanying resonant absorption and emission of a photon,[47,48] magnetic hyperfine structure[48,84–166] and the anomalous Zeeman effect[167] in vibrational-rotational transitions in molecules, elastic scattering of excited particles at small angles,[168] nonlinear dependence of the broadening and shift of narrow resonances in a low-pressure gas,[49,55] and others.

Progress in stabilizing laser frequencies and in the absolute measurements of laser frequencies opens up new possibilities for carrying out a number of precision physical experiments.[9,69,169] The precision of the measurements of one of the basic physical constants, the velocity of light, has now reached $\approx 10^{-9}$.[170] The value of the velocity of light $c = 299\ 792\ 458$ mls ($\delta c/c = \pm 4 \cdot 10^{-9}$), now adopted by the Consulting Committee on the Definition of the Meter (CCDM), is based on the results of these measurements. Experiments have been performed on the measurement of the Rydberg constant ($R_\infty = me^4/2\hbar^2$) with a precision of 10^{-8},[171] based on the precise determination of the center of a separate component of the fine structure of the H_α (6563 Å) line, using the saturated-absorption technique under the action of radiation from a pulsed tunable dye laser. The possibility of measuring the Rydberg constant with a higher accuracy of $\approx 10^{-10}$, based on the precision measurement of the frequency of the 1S-2S transition in the hydrogen atom, was examined in Ref. 69. The second-order Doppler effect in a gas was measured in Refs. 172 and 173. New applications of frequency standards include the measurement of extremely small displacements and the development of laser gravitational-wave detectors.[174] Studies carried out in Ref. 174 with an He-Ne/CH_4 laser at $\lambda = 3.39\,\mu$m made it possible to measure a displacement of $6 \cdot 10^{-6}$ Å on a 5 m baseline (relative sensitivity of $\approx 10^{-16}$). A displacement of $6 \cdot 10^{-15}$ cm on a baseline of 85 cm (relative sensitivity of $\approx 10^{-16}$) was measured in Ref. 175 with the help of narrow competing resonances in an He-Ne/CH_4 ring laser. The use of two-photon resonances and high power (~ 1 W) will make it possible to raise the sensitivity of the measurements by four to five orders of magnitude.

Frequency-stabilized lasers are now widely used in scientific research and in engineering, both as reference and standard sources of light for interferometric measurements of lengths and radiation wavelengths. In recent years a large number of precision measurements of wavelengths of the ^{3}He-^{20}Ne lasers, stabilized to the saturated-absorption resonances in $^{127}I_2$-, $^{129}I_2$-, and CH_4-, as well as of Ar^+/I_2 lasers relative to a ^{86}Kr standard have been carried out in different laboratories around the world and direct comparisons of these lasers have been made.[110,114,118,176–180] On the basis of these results, CCDM recommended in 1973 and 1979 that the following values of laser wavelengths be used as standards:

$$\begin{aligned}\lambda_{\mathrm{He-Ne}}\,(I_2) &= 632\ 991.399 \cdot 10^{-12}\ \mathrm{m},\\ &= 611\ 970{,}771 \cdot 10^{-12}\ \mathrm{m},\\ \lambda_{\mathrm{Ar}^+}\,(I_2) &= 514\ 673.476 \cdot 10^{-12}\ \mathrm{m},\\ \lambda\,(CH_4) &= 3\ 392\ 231.40 \cdot 10^{-12}\ \mathrm{m},\end{aligned}$$

The relative error of all these quantities was determined primarily by the krypton length standard.

An important practical application of stable lasers is for the measurement of small displacements in precision geophysics, seismology, geodesy, machine building, navigation, and in other areas. The development of laser systems enabling measurements over long baselines (> 1 km) in the real atmosphere is of special interest. Figure 23a shows an

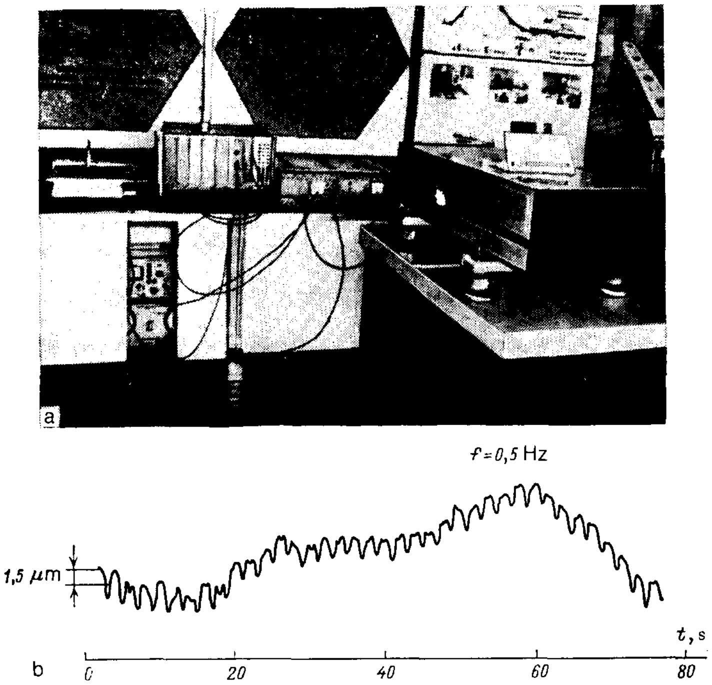

FIG. 23. a) External view of a laser meter for measuring small displacements along long baselines. b) Tracing of the periodic signal of a displacement on a 2 km path in the real atmosphere (from Ref. 181).

external view of a highly sensitive sensor for measuring small deformations of the earth's crust and recording seismic oscillations.[181] The device has a relative sensitivity of $\approx 10^{-9}$ for a baseline of 1–3 km. The first tracings of periodic displacements with an amplitude of $1\ \mu$m were obtained in the open atmosphere over a path length of ~ 2 km (Fig. 23b) with the help of such a laser deformograph.

11. CONCLUSIONS

In spite of the serious progress made in the development of laser frequency standards, a qualitatively new jump in this direction must be made in the years immediately ahead.

First, the possibilities of the saturated absorption method have not yet been completely exhausted, and we are still far from the limiting values of frequency stability which this method makes it possible to realize. Second, the expected progress is linked with the use of fundamentally new methods for stabilizing laser frequencies, based on the use of separated optical fields, two-photon resonances, and particle confinement. Together with the methods of cooling particles currently under development, they will permit obtaining extremely narrow resonances with relative widths of $\sim 10^{-14}$ and, which is very important, decreasing the influence of the second-order Doppler effect. This will enable getting down to the limit 10^{-16}–10^{-17}. Frequency-stabilization of tunable lasers remains important from the technical viewpoint. Simplification of the frequency synthesis scheme will permit developing optical time standards which will be able to compete in practice with the best microwave time and frequency standards. The accuracy of frequency measurements in the optical range must reach values of $\sim 10^{-11}$, and the measurement technique itself must be simple and widely available.

[1)] The study of the behavior of the form of the derivative of the resonance with respect to the frequency is important for both frequency stabilization problems and for spectroscopic experiments.

[2)] In many cases, when, for example, the amplitude of the resonance is small and it is necessary to exclude the effect of the slope of the Doppler contour of the gain line on the position of the resonance, the third-harmonic signal is used for frequency stabilization.

[3)] When stabilizing the frequency of high-power lasers it is desirable to use an external cell because of the strong saturation of absorption.

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