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OPTICAL FREQUENCY STANDARDS

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I. INTRODUCTION

During the last decade, in connection with the development of quantum radiophysics, much progress was made in the accuracy of measuring of one of the most fundamental physical quantities—the time interval. The accuracy of time measurement has increased from several times 10^{-8} to several times 10^{-12}. This progress was attained as a result of the use of atomic resonances to stabilize the frequency of coherent electromagnetic oscillations of quartz oscillators (quantum frequency standards). Both active frequency standards, in which microwave coherent radiation is generated as a result of stimulated emission of molecules or atoms[1-5], and passive frequency standards, in which the frequency of microwave generators is stabilized against the aborption lines of atoms[6-8], have been developed and are now in use. The most advanced devices are: a quantum generator using a hydrogen-atom beam[3] (stability on the order of 10^{-4}), a generator using a molecular ammonia beam (stability on the order of 10^{-11})[4], a generator stabilized by a rubidium gas cell (stability on the order of 10^{-11}), and a generator stabilized against the absorption line of a beam of cesium atoms in a resonator (stability on the order of 10^{-11}). Certain standards have a high relative frequency stability when the position of the generation frequency relative to the peak of the spectral line is not known but remains constant with a high degree of accuracy for the given standard (for example, rubidium standard). There exist, however, standards with high absolute frequency stability, in which the generation frequency coincides with or can be tuned to the "peak" of the spectral line* with a high degree of accuracy (for example, cesium and hydrogen standards). In 1964, the International Committee on Measures and Weights adopted as a standard for the physical measurement of time an instrument based on the Cs^{143} spectral line (transition $F = 4$, $m_F = 0 \rightarrow F = 3$, $m_F = 0$ between the levels of the hyperfine structure of the $2F_{1/2}$ ground state of Cs^{133} in the absence of external fields). The frequency of this standard has been assigned a value 9 192 631 770 Hz †.

In view of recent development of methods for generating coherent radiation in the visible and infrared bands[12-16], it became feasible in principle to develop frequency standards in the 10^{13}–10^{15} Hz band (optical frequency standard). It is obvious that even if the frequency stability already attained in the microwave band can be achieved in optical generators, the region of their physical applications will greatly increase. An essential feature of optical-band frequency standards is the appreciable reduction of the time of frequency measurement with a given accuracy, since the accuracy of frequency measured within a finite time interval is inversely proportional to the frequency. Therefore, at a frequency 10^{14} Hz, measurements with accuracy 10^{-14} can be performed within a time on the order of 1 second, but at the frequency of the hydrogen maser (ν = 1420.405 MHz) this would require a time on the order of a day.

To produce a frequency standard in the optical band, it is necessary to solve two rather complicated problems. First, it is necessary to produce an optical-oscillation generator with a highly stabilized emission frequency, and second it is necessary to produce a "meter" for the frequency (and not for the wavelength) of the optical oscillations. The solution of the first problem makes it possible to produce a length standard. To this end, it suffices to use the radiation from an optical generator of stable frequency in interferometric length-measuring devices.*

The solution of the second problem makes it possible to develop a frequency or time standard. Independent measurements of the frequency and of the wavelength make it possible to measure the speed of light with the accuracy with which the time and the distance are determined. In other words, this means that it is possible to dispense with separate standards for time and length.[19,20]

Most papers on optical frequency standards deal with the solution of this problem. We shall therefore naturally pay principal attention to this question (Ch. II–V). The problem of absolute measurement of the frequency of optical oscillations is dealt with in only a few papers (see Ch. VI). In the last section of Ch. VII we shall discuss briefly the fundamental physical experiments that can be realized with the aid of optical frequency standards. This discussion is more of an illustration of the possibility of the frequency standards, and therefore does not claim to be complete.

1. Possible Methods

The frequency ω of a quantum generator depends on the frequency ω_0 of the center of the spectral line and

*Owing to the hyperfine structure of the line, the position of the generation frequency relative to the vertex of the spectral line may not be known, but can be reproducible with a high degree of accuracy in independent samples of the standard.

† More detailed information on atomic frequency standards can be found in the monographs [9,149] and in the special review [10,11,139,140].

*At the present time, the primary wave standard is the wavelength of the $2p_{10}$–$5d_5$ spectral line of Kr^{86}. The international meter spans 1 650 763.73 such wavelengths (resolution of the International Committee on Measures and Weights, 1960) [18].

on the natural frequency ω_r of the resonator in the following manner[2,21]:

$$\frac{\omega-\omega_r}{\omega_0-\omega}=\frac{\Delta\omega_r}{\Delta\omega_a}, \tag{1.1}$$

where $\Delta\omega_r$ is the bandwidth of the resonator and ω_a is the width of the spectral line. The natural frequency ω_r of the resonator is inversely proportional to its length and is therefore known to be unstable. The influence of this instability on the generation frequency can be eliminated in two ways.

First, it is possible to exclude in practice the dependence of the generation frequency on the resonator frequency ω_r by making the bandwidth $\Delta\omega_r$ of the resonator larger by several orders of magnitude than the bandwidth $\Delta\omega_a$ of the spectral line of the active medium:

$$S=\frac{\Delta\omega_r}{\Delta\omega_a}\gg 1. \tag{1.2}$$

Fairly high values of the factor S 10^3–10^5 were attained in the radio band, by using a number of effective methods of obtaining narrow spectral lines (the molecular-beam method[1,2], the Dicke method of buffer gas[22], the Ramsey method of separated resonators[40], the Ramsey storage-tube method[3]). In the optical band, the value of S usually does not greatly exceed unity. However, recently, a number of methods has been proposed for obtaining high values of the stability factor in the optical band, by narrowing down the spectral line or by broadening the resonator bandwidth. Two methods of obtaining narrow spectral amplification lines are considered in[23,24]. These methods make it possible to obtain in principle a spectral line width $\Delta\omega_a \cong 10^3$–$10^5$ Hz, determined only by the finite time of interaction between the atoms and the light field. An effective method of broadening the resonator band by completely destroying the resonant properties of the feedback (method of numbers in feedback) is proposed in[25].

Second, the instability of the natural frequency of the resonator can be eliminated by automatically tuning the generation frequency either to the peak of the spectral line employed in the laser, or to the peak of the spectral line of an external resonant element. This method is the basis of numerous schemes for stabilizing the frequency of gas lasers. A review of these schemes is contained in[30,31], so that these are considered in the present article only very briefly (Ch. V). We shall consider in greater detail only the further development of this method, namely the use of a beam of atoms or molecules as an external resonant element[32], and self-tuning of the generation frequency to the center of the spectral line without an external feedback loop (the method of autoresonant feedback[33]).

In the ideal case, when the influence of the frequency drift of the generator is completely eliminated, the stability of the generation frequency is determined by the stability of the center of the spectral line.

2. Frequency Stability of Optical Spectral Lines

The shape and width of spectral lines in the optical band are determined by the joint action of three principal effects: 1) radiative broadening, 2) Doppler broadening, 3) broadening due to particle collisions. Usually the Doppler widths $\Delta\omega_D$ of the lines used in lasers exceed greatly the radiative and the collision widths, so that a Doppler line shape is obtained in practice. The Doppler broadening is inhomogeneous: the position of the spectral line of each particle $S_0(\omega)$ depends on its velocity **v**, and the spectral line of the entire aggregate of moving particles $S(\omega)$ consists of the spectral lines of the individual particles in accordance with their velocity distribution. Thus, the spectral line of the absorption of the traveling light wave $E\cos(\omega t - \mathbf{k}\cdot\mathbf{r})$ is of the form

$$S(\omega, \mathbf{k})=\int S_0(\omega-\mathbf{kv})\,W(\mathbf{v})\,dv, \tag{1.3}$$

where $S_0(\omega)$ is the spectral line of one particle and $W(\mathbf{v})$ is the particle velocity distribution. If the distribution $W(\mathbf{v})$ is isotropic, then only broadening of the spectral line takes place. However, in the case of an anisotropic velocity distribution, for example, as a result of particle drift in the gas covering the beam, a shift of the spectral line, which depends on the direction of the traveling wave, is possible.

A feature of an inhomogeneously broadened Doppler line is the formation of a "hole" in the spectral line under the influence of the intense light wave $E\cos(\omega t - \mathbf{k}\cdot\mathbf{r})$ causing saturation of the transition. The hole is produced at the frequency ω of the light wave, and its width equals the homogeneous width of the spectral line $\Delta\omega_{hom}$. In the optical range, the homogeneous width consists of a radiative width and of a width determined by the collisions. At an appreciable intensity of the light wave, it is necessary also to take into account the broadening due to the saturation effect[58]. This question is discussed in greater detail in Sec. 2 of Ch. III.

The symmetry of the spectral line depends on the hyperfine structure of the levels and on the particle collisions. The hyperfine structure of the levels can lead also to asymmetry of the spectral line, which depends on the random external electric and magnetic fields. Particle collisions not only lead to a shift of the peak of the spectral line, but can also lead to asymmetry of the line contour, for example, as a result of correlation of the broadening due to the interaction and the Doppler broadening following the collisions of the particle[51].

Methods of obtaining narrow spectral lines, proposed in[23,24], are based on separating in the inhomogeneously broadened line, with the aid of the saturation effect*, a homogeneous line with a maximum located in the ideal case in the center of the Doppler line. Therefore the limiting width of the lines is determined by the homogeneous width. For allowed transitions, the radiative part of the homogeneous width in the optical range usually amounts to 10^7–10^8 Hz. Therefore, to obtain narrow and ultranarrow lines, it is necessary to use

*One more method of separating a narrow spectral line in the Doppler contour is considered in [147]. It is based on the effect of "dragging" of the atoms in an intense standing light wave without using the saturation effect. The limiting width of the spectral line is determined in this case also by the finite time of the interaction of the atoms with the field.

forbidden transitions with a probability $A_{12} \lesssim 10^3-10^5$ sec^{-1}. In this case the radiative width can amount only to $\gamma = 10^2-10^4$ Hz or even less. The broadening due to the collisions is determined by the expression[131]

$$\Gamma = 2N\langle\sigma' v\rangle, \tag{1.4}$$

where N and v are the density and velocity of the particles, and σ' is the effective broadening cross section. In broadening by neutral particles, the cross section σ' usually exceeds the gas-kinetic cross section, and for rough estimates we can use $\sigma' \cong 10^{-15}$ cm^2. At an average particle velocity $v \cong 10^5$ cm/sec, the broadening due to the collisions amounts to 1 MHz/torr. In individual cases, for example in collisions with resonant excitation transfer or in collisions with small-angle scattering, when σ' exceeds the gas-kinetic cross section by several orders of magnitude, the broadening is much larger[131]. To obtain spectral lines with a width on the order of 10^3-10^4 Hz, the pressure of the gas should not exceed $10^{-4}-10^{-3}$ torr. At small homogeneous widths on the order of 10^3-10^4 Hz, it is necessary to take into account broadening effects that are negligibly small in ordinary cases: namely, broadening due to collisions with the vessel walls and broadening due to the finite time of coherent interaction of the particles with the light beam. As will be shown later (Ch. II and III), the broadening due to saturation and broadening due to the finite time of interaction determine the maximum principally attainable value of the width of the spectral line. We note that it is precisely these two effects which determines the limiting width of the spectral lines in the radio band. Thus, in the optical band, it is possible to obtain in principle ultranarrow spectral lines with a width close to the line widths in the radio band.

To produce a frequency standard it is necessary to have high stability of the center ω_0 of the spectral line. The greatest contribution to the frequency stability of the spectral line is made by the line shape due to the collisions of the particles with one another. The magnitude of the shift is determined with the expression[131]

$$\Delta = N\langle\sigma'' v\rangle. \tag{1.5}$$

The magnitude of the shift Δ due to the collisions is usually of the same order as the broadening Γ due to the collision[131], it amounts to several MHz per torr of gas pressure. In presently existing lasers, the pressure of the gas mixture reaches several torr and the dependence of the generation frequency on the pressure is fully observable in the experiment[26,27]. To obtain a frequency stability better than 10^{-11} it is necessary to maintain the pressure constant within $10^{-3}-10^{-4}$ torr, which is not feasible in practice. A natural way of getting around this difficulty is to use active media with very low pressure. This is perfectly feasible in principle, since the pressure of those active "atoms" that are at the two working levels is quite small, and in an He-Ne laser, for example, it does not exceed 10^{-6} torr. Of course, it is quite difficult to obtain inverted population in a low-pressure gas by standard methods (gas discharge in the gas mixture). It is more realistic to obtain population inversion by optical pumping*[35,12,29,80,81]. To this end it is necessary to choose atoms or molecules that absorb resonantly the radiation of well known gas lasers. This is perfectly attainable both using atomic or molecular beams[23,28] or by using low-pressure vapor in a laser with nonresonant feedback[29]. The situation is even simpler in the method of a nonlinear absorbing gas cell[24], where there is no need for inverted level population. The gas pressure in an absorbing cell can be less than 10^{-3} torr.

The position of the spectral line is influenced by external magnetic and electric fields. For example, the linear Zeeman splitting of the level is determined by the relation[131]

$$\Delta\nu = g\frac{e}{2mc}H, \tag{1.6}$$

where g is the g-factor of the level. Usually $g \cong 1$ and the splitting amounts to 10^7 Hz/Oe. The linear Zeeman effect does not shift the center of gravity of the line, but owing to the polarization of the radiation, to the hyperfine structure of the levels, and to the collisions, an asymmetry unavoidably takes place, and we can expect a generation-frequency shift by an amount on the order of several MHz/Oe. Experimentally, this was recently observed in an He-Ne laser[26]. The generation frequency shift amounted to several tenths of a MHz/Oe. The Stark splitting is quadratic in the field and amounts to several MHz in a field of intensity of several V/cm. It is clear that in order to obtain high frequency stability, random external magnetic and electric fields should be quite small, and the operating regime should be chosen such as to make the dependence of the frequency on the external fields sufficiently small. From this point of view, there is a certain danger in the use of a gas discharge to produce inverted population, all the more since a shift of the spectral line is possible in discharge tubes, owing to the effects of the ion and electron drift and of the corresponding motion of the emitting or absorbing ions. Effects of this kind were observed with a krypton length standard[141]. However, these difficulties are eliminated when optical pumping is used.

In addition, there are also relativistic effects of the shift of spectral lines: the transverse Doppler effect and the Doppler shift due to the recoil effect. The Doppler effect of second order produces a line shift in the red direction

$$\frac{\Delta\nu}{\nu} = -2\frac{v^2}{c^2}, \tag{1.7}$$

which at thermal velocities amounts to $10^{-11}-10^{-12}$. It contributes to the frequency instability if the average thermal particle velocity is unstable. The recoil effect leads to a shift of the spectral emission line relative to the absorption line by an amount

$$\frac{\Delta\nu}{\nu} = \frac{h\nu^2}{Mc^2}, \tag{1.8}$$

where M is the particle mass. In the visible and in the infrared bands, for particles with atomic weight

*In this case the generation frequency may depend on the pump frequency. Methods of suppressing this effect are considered below (Sec. 1 of Ch. II and Sec. 6 of Ch. IV).

$A \cong 10^2$, this shift amounts to 10^{-11}–10^{-12}. The recoil effect makes no contribution to the frequency instability of the spectral line, but is capable of limiting the ultimate spectral width in a laser in which emission and absorption are simultaneously used, particularly in a laser with coherent excitation[23].

Thus, in principle, optical spectral lines of atoms and molecules make it possible to attain a high long-time stability. Short-time frequency stability is determined by the spectral width of the generating radiation.

3. Spectral Width of Laser Emission

In an ideal laser with resonant feedback (with a Fabry-Perot interferometer), the spectral emission widths in one oscillation mode is determined only by the spontaneous noise fluctuation[12] and by the thermal (Brownian) motion of the resonator mirrors[143]. The line width $\Delta\nu_{sp}$ due to the spontaneous-radiation noise in one oscillation mode is determined by the relation

$$\Delta\nu_{sp} = \frac{8\pi\hbar\omega}{P}\left(\frac{\Delta\nu_a\Delta\nu_r}{\Delta\nu_a+\Delta\nu_r}\right)^2, \tag{1.9}$$

where $\Delta\nu_a$ is the width of the spectral line of the active medium, $\Delta\nu_r$ is the line width of the resonator, P is the generation power. At $\Delta\nu_r = 1$ MHz, $\Delta\nu_a = 100$ MHz, a generation power $P = 10^{-3}$ W, and a wavelength $\lambda = 1\ \mu$, which are typical of ordinary He-Ne lasers, the emission spectrum width is $\Delta\nu_{sp} \cong 5 \times 10^{-3}$ Hz, or $\Delta\nu_{sp}/\nu \cong 10^{-17}$. In optical frequency standards with narrow spectral lines, $\Delta\nu_a \cong 10^3$–10^5 Hz, the spectral emission widths due to the spontaneous noise should be even smaller. The emission line widths connected with thermal fluctuation motion of the mirrors is determined by the relation

$$\Delta\nu_T = \nu\frac{\Delta\nu_a}{\Delta\nu_a+\Delta\nu_r}\frac{\delta L}{L}, \tag{1.10}$$

where L is the resonator length and δL is the mean-square fluctuation variation of the resonator length. If the mirrors are securely fastened on a solid rod having a volume V, a Young's modulus Y, and a temperature T, then, according to Javan[143], the thermal excitation of the lowest mode of the rod yields the following value of δL:

$$\frac{\delta L}{L} = \sqrt{\frac{2kT}{YV}}. \tag{1.11}$$

For a metallic rod ($Y \cong 2 \times 10^{11}$ dyne/cm^2) with volume $V = 10^3$ cm^3, at a temperature $T = 300$°K, the relative fluctuation change of length $\delta L/L \cong 2 \times 10^{-14}$. For ordinary gas lasers, when $\Delta\nu_r \ll \Delta\nu_a$, the corresponding "thermal" width of spectrum is $\Delta\nu_T/\nu \cong 2 \times 10^{-14}$, i.e., higher by three orders of magnitude than the width due to the spontaneous noise. However, for an optical frequency standard with $\Delta\nu_r/\Delta\nu_a \gg 1$, the "thermal" width is much smaller. For example, at $S = 10^3$ the broadening of the spectrum due to the thermal fluctuations of the mirrors is of the same order as the width due to the spontaneous noise, and amounts to 10^{-17}. Thus, we can expect a rather high limiting short-time stability for optical frequency standards. In a laser with nonresonant feedback, owing to the fluctuations inherent in this type of laser, the short-time frequency stability is apparently much lower. A detailed discussion of this question is contained in Sec. 6 of Ch. IV.

II. METHOD OF COHERENT EXCITATION OF A BEAM OF ATOMS OR MOLECULES

1. Idea of Method

The use of beams to reduce the Doppler width of emission lines in the optical spectrum is well known[34]. In the case of observation in a direction perpendicular to the beam, it is possible to reduce the Doppler width of the line by one or two orders of magnitude.* For example, a beam of mercury atoms excited by a resonant-radiation beam emits a 2536.7 Å line with width 10^{-5} Å, determined by the radiative damping of the level[39]. By various methods (see, for example,[35-37]) it is possible to produce inverted population of the levels of the atoms in the beam and then obtain coherent generation. However, such a use of a beam of active atoms is not effective in a laser, since the line is quite broad.

A method of obtaining very narrow amplification lines (with widths 10^4–10^5 times smaller than the Doppler width), based on coherent excitation of atoms or molecules in a light beam, was proposed in[23]. The beam of atoms intersects, parallel to the wave front, the coherent-light beam whose frequency ω coincides with the transition frequency ω_0 of the atoms in an excited long-lived state. Since there is no relaxation, the atom oscillates in the resonant field between two levels with a frequency Ω proportional to the intensity of the light field. If the time τ_0 that the atoms stay in the light beam coincides with the half-period π/Ω of the oscillations, then the initially unexcited atoms turns out to be excited after the passage of the light beam. The small dimension of the light wave λ compared with the beam diameter a leads to a unique shape of the atom-beam line after passage of the light beam. As will be shown later, the only inverted atoms are those in the narrow spectral interval with center at a frequency ω and with a width

$$\Delta\omega_{tf} = \frac{2}{\tau_0} = \omega\frac{v_0}{c}\frac{\lambda}{\pi a}, \tag{2.1}$$

determined by the time of flight of the atoms through the light beam $\tau_0 = a/v_0$ (v_0—average velocity of the atoms in the atom beam). The amplification line width is smaller by a factor $\pi a/\lambda$ than the Doppler line width of the emission of the atoms. For example, at $a = 1$ cm and $\lambda = 1\ \mu$, the amplification line is narrower than the Doppler line by a factor 2×10^4.

The frequency of the center of the amplification line is determined by the frequency ω of the exciting light beam, and therefore the generation frequency stability cannot be better than the stability of the pump frequency ω. This difficulty can be circumvented by using as the exciting light beam the radiation of the beam laser itself. To compensate for the inevitable radiation losses in the laser, it is necessary to pre-

*A beam laser for the submillimeter band with a Fabry-Perot resonator was proposed in [14].

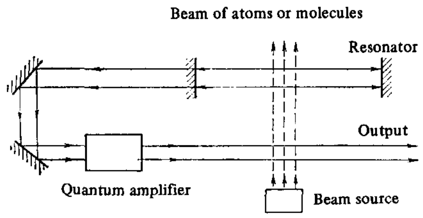


FIG. 1. Diagram of beam laser with coherent pumping by its own radiation [23].

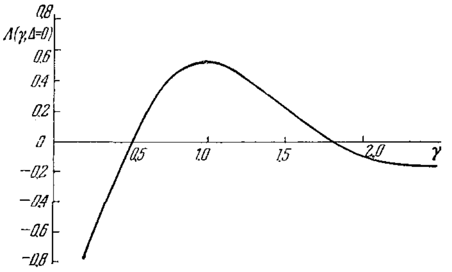


FIG. 2. Dependence of the inversion Λ of the atoms in the beam after passage of the exciting light beam on the parameter $\gamma = \tau_0/\tau_{in}$ at zero detuning ($\Delta = 0$) [28].

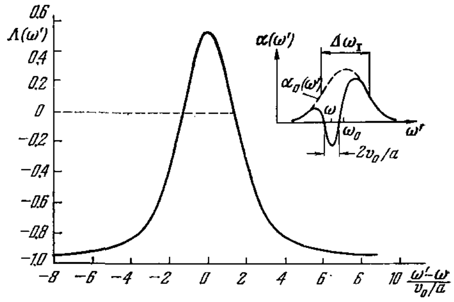


FIG. 3. Dependence of inversion Λ of atoms in a beam after passage of an exciting light beam on the detuning Δ of the transition frequency relative to the field frequency at $\gamma = 1$ [28].

amplify the laser emission. As a result, we obtain a beam laser with coherent pumping by its own radiation, as shown in Fig. 1. The most important feature of this scheme is the high stability of the emission frequency.

2. Line Shape of Excited Beam

By considering the resonant interaction of an atom with a light beam during the time of flight of the atom through the light beam $\tau = a/v$, we can calculate the probability amplitudes a_1 and a_2 for finding the atoms at the ground level 1 and the excited level 2. The expression for the inversion of one atom is

$$|a_2|^2-|a_1|^2=-1+\left(2\,\frac{p_{12}\mathscr{E}}{\hbar\Omega}\right)^2(1-\cos\Omega\tau), \tag{2.2}$$

where

$$\Omega=\left[(\omega-\omega_0+\mathbf{kv})^2+\left(2\,\frac{p_{12}\mathscr{E}}{\hbar}\right)^2\right]^{1/2} \tag{2.3}$$

and p_{12} is the matrix element of the dipole moment, and $\mathscr{E}$ is the intensity of the field in the beam. The Doppler frequency shift $\mathbf{k}\cdot\mathbf{v}$ is due to the fact that the atom velocity $\mathbf{v}$ is not parallel to the wave front of the light beam ($\mathbf{k}$ is the wave vector of the beam). If φ is the angle of deviation of the velocity from the wave front of the light beam, then $\mathbf{k}\cdot\mathbf{v} = kv\varphi$ ($\varphi \ll 1$). The inverted population per atom Λ is determined by the relation (2.2) averaged over the velocity distribution of the atoms:

$$\Lambda=\langle|a_2|^2-|a_1|^2\rangle. \tag{2.4}$$

The average inversion Λ of the atom depends, obviously, on two parameters: on the ratio γ of the average time of flight τ_0 of the atoms through the light beam to the time of the total inversion at exact resonance ($\omega = \omega_0$), $\tau_{in} = \pi/\Omega = \pi h/2p_{12}\mathscr{E}$, and the detuning Δ of the field frequency ω relative to the absorption frequency $\omega_0 - \mathbf{k}\cdot\mathbf{v}$. It is physically clear that the inversion Λ is maximal at $\gamma = 1$. The exact $\Lambda(\gamma, \Delta = 0)$ dependence obtained by numerical averaging at $\Delta = 0$ is shown is Fig. 2.

In the real case of a beam of atoms with angular divergence $\varphi_0 \gg \lambda/\pi a$, the Doppler width of the beam $\Delta\omega_D \cong \varphi_0 k v_0$ greatly exceeds the line width $\Delta\omega_{lim} = 2/\tau_0$ due to the finite time of flight of the atoms through the light beam. In this case the absorption line is inhomogeneously broadened, and it is natural to expect that only the atoms whose Doppler frequency shift lies within the limits of the homogeneous widths $\Delta\omega_{lim}$ relative to the field frequency ω will be inverted. The dependence of the inversion of the atoms $\Lambda(\gamma = 1, \Delta)$ on the detuning of the field frequency relative to the absorption frequency $\omega = \omega_0 - \mathbf{k}\cdot\mathbf{v}$, obtained by numerical averaging at optimal $\gamma = 1$, is shown in Fig. 3. In the upper part of Fig. 3 is shown the absorption line shape of the atom beam after passage of the light beam $\alpha(\omega') = \Lambda(\omega')\alpha_0(\omega')$, where $\alpha_0(\omega')$ is the initial absorption line shape of the atom beam. The line width of the negative absorption is determined only by the average time of flight of the atoms through the light beam*.

3. Generation Regime

A beam laser with coherent excitation by its own radiation should have a number of properties that greatly distinguish it from ordinary lasers[28,38].

When a beam of atoms falls into a resonator after being acted on by the exciting light beam, absorbing atoms for which $\Lambda < 0$ enter into the resonator together with the amplifying atoms for which $\Lambda > 0$. If the wave fronts of the field in the resonator and in the exciting light beam are strictly parallel, then at a sufficiently prolonged time of flight of the atoms through the resonator, only the amplifying atoms will interact effectively with the field. The contribution of the ab-

*It is interesting to note that level population inversion of atoms can be obtained also in an incoherent light field. In this case, besides the requirement with respect to the field intensity ($\gamma = 1$), it is necessary to satisfy the following conditions:

$$\tau_0 \leqslant \tau_{coh} \quad a \leqslant r_{coh}$$

where τ_{coh} and r_{coh} are the coherence time and the dimension of the coherence area of the incoherent field.

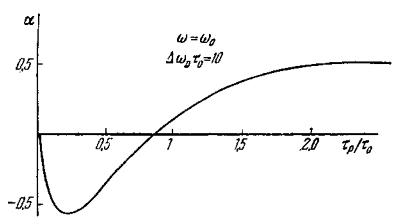


FIG. 4. Gain of atom beam in resonator vs. ratio of the time of flight τ_r of the atoms through the resonator to the time of flight τ_0 through the light beam, for the simplest model of the laser-beam absorption line shape $\alpha(\omega')$ shown in the top of Fig. 3. [38]

sorbing atoms will be quite small, since the resonance condition is not satisfied for them. Figure 4 shows the dependence of the gain of the laser beam in the resonator on the ratio of the time of flight τ_r of the atom through the resonator to the time of flight τ_0 through the light beam for the absorption line after the excitation, as shown in Fig. 3. The role of the absorbing atoms becomes negligibly small already in the case when $\tau_r/\tau_0 = 2$. It is shown in[38] that there exist a "sharp" dependence of the gain of the laser beam in the resonator on the detuning of the field frequency ω relative to the absorption frequency ω_0 of the atoms traveling parallel to the wave front of the beam. The laser beam has positive gain if $|\omega - \omega_0| \lesssim 1/\tau_0$, which is maximal when $\omega = \omega_0$. Physically this is explained by the fact that, in the presence of a detuning $\omega_0 - \omega$, the inverted atoms are those having a transverse velocity $u = (\omega_0 - \omega)/k$. One of the traveling waves propagating in the direction of the exciting beam interacts resonantly with the amplifying atoms (with $\Lambda > 0$) in the resonator, and the other, propagating in the opposite direction, interacts resonantly with the absorbing atoms (with $\Lambda < 0$), which have a transverse velocity component $u = -(\omega_0 - \omega)/k$.

Another feature of the laser consists in the fact that the gain of the atoms entering the resonator is determined by the amplitude of the generated field $\mathscr{E}$:

$$\alpha(t) \sim \Lambda\left[\frac{2p_{12}}{\pi\hbar}\int_0^{\tau} k_0\mathscr{E}(t-\tau-t')\,dt'\right], \tag{2.6}$$

where $\Lambda(\gamma)$ is the excitation function shown in Fig. 2, τ is the time of flight from the pump light to the resonator (delay time), k_0 is the ratio of the field amplitude in the resonator and in the exciting light beam ($k^2 = \eta_0 k$, η_0 is the transmission coefficient of the output mirror of the laser, and k is the gain of the optical quantum amplifier). Since $\Lambda < 0$ for $\gamma = 0$, the laser has a "hard" self-excitation mode. This means that to obtain generation it is necessary to have an initial field with a threshold amplitude. The frequency ω of the exciting radiation should satisfy the condition $(\omega - \omega_0) \lesssim 1/\tau_0$, and the intensity at the amplifier output should exceed the threshold intensity given by the expression

$$I_{\text{thr}} \simeq \frac{\pi^3}{6}\,\frac{\hbar\omega}{A\lambda^2\tau_0^2}, \tag{2.7}$$

where A is the transition probability. A scheme for obtaining a "soft" self-excitation regime will be given below in Chapter V. In[142] it is shown that the amplification of the "molecular ringing" can lead to a "soft" self-excitation regime.

The threshold gain of the amplifier k_{thr} is given by

$$k_{\text{thr}} = \frac{\eta}{\eta_0\varkappa_0}\,\frac{2-\Lambda_m+\frac{\eta}{\varkappa_0}}{\Lambda_m-\frac{\eta}{\varkappa_0}}, \tag{2.8}$$

where η is the coefficient of radiation loss in the resonator, including the transmission loss: $\Lambda_m = \Lambda$ ($\gamma = 1, \Delta = 0$); κ_0 is the coefficient of absorption of radiation by the beam of atoms. Expression (2.8) pertains to the case $\eta, \eta_0, \kappa_0 \ll 1$. Physically such a laser is realizable only when $\eta < \kappa_0\Lambda_m$. When the threshold is exceeded, there are two stationary values of the field amplitude, $\mathscr{E}_1$ and $\mathscr{E}_2$. Only the second stationary state $\mathscr{E}_2 > \mathscr{E}_1$ is stable. The presence of the delay τ leads to an upper limit of the region of stable values of $\mathscr{E}_2$. This limitation is significant only at small values of the delay τ_0 compared with the time of flight τ_0 through the light beam.

4. Frequency Stability

Let us consider first an ideal case when 1) the Doppler line of the molecule beam has no hyperfine structure; 2) the center of the amplification line $\omega_{0\text{amp}}$ coincides exactly with the generation frequency ω; 3) the average direction of the molecule beam is parallel to the wave front of the standing wave in the resonator, and 4) the distribution of the gain in the transverse direction inside the resonator is homogeneous. Then the stationary value of the generation frequency is determined by the expression

$$\omega = \omega_0 + \frac{\Delta\omega_{\text{lim}}}{\Delta\omega_r}\left(1+\frac{\Delta\omega_{\text{lim}}}{\Delta\omega_r}+\frac{\Delta\omega_{\text{lim}}}{\Delta\omega_D}\right)^{-1}(\omega_r-\omega_0), \tag{2.9}$$

where ω_r and $\Delta\omega_r$ are the natural frequency and the bandwidth of the resonator, and $\Delta\omega_D$ is the Doppler line width of the beam. It is possible to obtain realistically a stability factor $S = \Delta\omega_r/\Delta\omega_{\text{lim}} \cong 103$, particularly when $\Delta\omega_{\text{lim}} \cong 105\ \text{sec}^{-1}$ and $\Delta\omega_r \cong 10^8\ \text{sec}^{-1}$. Then, to obtain a frequency stability on the order of 10^{-12} it is necessary to stabilize the resonator length accurate to 10^{-9}. However, in practice the frequency stability is influenced by the effects 1)–4), which are not accounted for by the relation (2.9).

Owing to the hyperfine structure of the levels within the limits of the Doppler line of the molecule beam, the excitation may give rise to several narrow amplification lines with widths $\Delta\omega_{\text{lim}}$ at the center of each component of the line structure. The generation is realized at the most intense component, and the remaining pull the generation frequency. The pulling is small if the distance between the components of the hyperfine structure is $\Delta\omega_{\text{str}} \gg \Delta\omega_{\text{lim}}$. However, when $\Delta\omega_{\text{str}} \cong \Delta\omega_{\text{lim}}$ the pulling can reach a value $\sim\Delta\omega_{\text{lim}}$. The presence of random external electric and magnetic fields changes the position of the components of the structure, and by the same token the generation frequency. We can expect that in the worst case the instability of the generation frequency will amount to several times 10% of the width $\Delta\omega_{\text{lim}}$. When $\Delta\omega_{\text{lim}} \cong 10^5\ \text{sec}^{-1}$ and $\omega = 2\times10^{15}\ \text{sec}^{-1}$, this yields an instability on the order of 10^{-11}. This, however, is an upper bound, and we can expect a better stability when a suitable transition is chosen.

The instability of the center of the amplifier line ω_{0amp} leads to a shift of the generation frequency by an amount on the order of $\Delta\omega_{lim}/\Delta\omega_{amp}(\omega_{0amp} - \omega)$, where $\Delta\omega_{amp}$ is the amplification bandwidth. The width of the amplification band of a gas optical amplifier is due to the Doppler effect, and therefore the quantity $\Delta\omega_{lim}/\Delta\omega_{amp}$ amounts to several times 10^{-5}. Consequently, at an amplifier frequency stability 10^{-8}, the contribution of this effect to the instability of the generation frequency will amount to several times 10^{-13}.

The deviation $\delta\varphi$ from parallelism between the average beam direction and the wave front in the resonator leads to a generation frequency shift amounting to about $\Delta\omega_{lim}\delta\varphi/\varphi_0$, where φ_0 is the angular spread of the beam. If $\Delta\omega_{lim}/\omega \cong 10^{-10}$, then to obtain a frequency stability of 10^{-12} it is necessary to adjust the beam direction parallel to the wave front with an accuracy of the order of 1% of the angular spread of the beam.

There is also a generation frequency shift due to the decrease of the inverted population of the laser beam on passing through the field in the resonator, similar to the shift due to the traveling-wave effect in a molecular generator[42,9]. In other words, when the laser beam passes through the resonator, a transverse inhomogeneity of the gain in the resonator takes place; this leads to a shift in the generation frequency[133]. Since the transverse inhomogeneity depends on the field amplitude (saturation effect), this leads to a dependence of the generation frequency on the field amplitude. The stability of the field amplitude depends on a number of parameters: the gain and the frequency of the amplifier, the laser beam intensity, the natural frequency of the resonator, etc. When $S \cong 10^3$, this effect can produce a frequency shift in the 11th decimal point.

There is a possibility of obtaining very narrow amplification lines with width $\Delta\omega_{lim} = 10^3$–$10^4$ sec^{-1}, by increasing the time of coherent interaction of the atoms with the pump ray and the field in the resonator. Figure 5 shows the diagram of a beam laser with coherent pumping, in which the time of interaction can reach 10^{-3}–10^{-4} sec at an atom velocity $v_0 \cong 10^4$–10^5 cm/sec. In this laser, atoms crossing all the exciting rays parallel to the wave front are inverted. Besides the additional narrowing of the amplification line in such a scheme, an increase takes place in the coefficient of absorption of the pump radiation by the atom beam, and accordingly the threshold gain of the amplifier decreases. The limiting width $\Delta\omega_{lim}$ is bounded both by technical factors (bending of the wave front of the pump ray in the resonator, non-parallelism of individual rays), and by the principal effect, namely the recoil effect. Owing to the recoil effect, the frequency of the center of the absorption line in the excited ray does not coincide with the frequency of the center of the emission line in the resonator. Since the pumping is produced by the laser's own radiation, the line shift due to the recoil, determined by relation (1.8), should be smaller than the line width $\Delta\omega_{lim}$. This imposes a limitation on the width of the spectral line:

$$\Delta\omega_{lim} \leqslant \frac{\hbar\omega^2}{2Mc^2}\,.$$

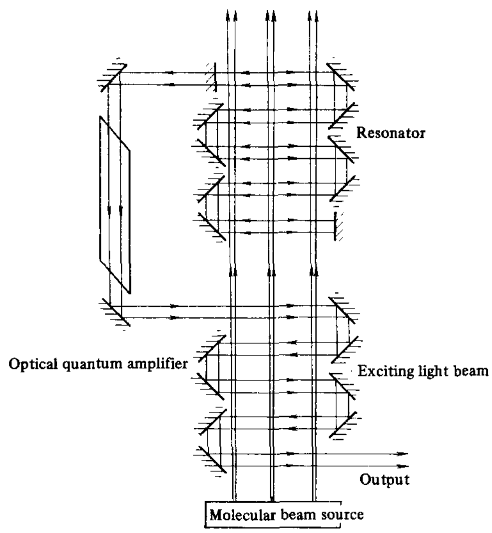


FIG. 5. Diagram of beam laser with coherent pumping by its own radiation, having a large time of coherent interaction between the atoms and the exciting light beam and the field in the resonator.

However, with the aid of such a method it is apparently possible to obtain a stability factor $S \cong 10^5$ and to increase the frequency stability by one or two more orders of magnitude.

5. Choice of Atoms and Molecules

When choosing atoms and molecules as the active media of a beam laser, it is necessary to satisfy three fundamental conditions:

1) The probability of the radiated decay of the excited particles should lie in the interval $1\ \text{sec}^{-1} \lesssim A \lesssim (\tau_0 + \tau_r + \tau)^{-1}$. The upper limit is due to the conditions that the spontaneous decay of the excited particles be small during the flight from the pump ray to the exit from the resonator. The lower limit is connected with the need of having moderate particle fluxes from the source. When $A < 1$ sec^{-1} the threshold laser-beam intensity is too high. To be sure, the lower limit is not definite. For example, it can be larger by one or two orders of magnitude for molecules than for atoms, owing to the presence of the rotational structure of the levels.

2) The lower level should either be the ground level or be within a distance $\lesssim kT$ from the ground level (T—temperature in the beam source).

3) Resonance condition: the transition frequency should be in resonance with the amplification frequency of the quantum amplifier, with a gain exceeding the threshold gain k_{thr}.

A number of forbidden transitions in the optical band between the lower levels of atoms satisfying conditions 1) and 2) was selected in[23]. Semiconductor amplifiers are promising from the point of view of satisfying condition 3), since they have a high gain[134] and the amplification band can be tuned in a rather broad range with the aid of triple coincidences[41]. The geometry of the existing semiconductor amplifiers makes this difficult. The situation will change, however, if injection amplifiers with appreciable gain in a direction perpendicular to the p-n junction are developed (similar to the laser described in[145]).

In the infrared region it is possible to use transitions between vibrational levels of molecules. For example, conditional), 2), and 3) are satisfied for the following two molecules (at the lower limit): 1) the absorption line of the CH_4 molecule, 2947.906 cm^{-1}, coincides within 0.003 cm^{-1} with the emission (amplification) line λ = 3.3913 μ of an He-Ne laser at an absorption coefficient 0.17 cm^{-1} torr[43,44]; 2) the 2850.608 cm^{-1} absorption line of the H_2CO molecule, coincides within 0.007 cm^{-1} with the emission (amplification line) λ = 3.5070 μ of an He-Ne laser at an absorption coefficient $\gtrsim$ 0.1 cm^{-1} torr[45]. Further development of radiation-amplification methods will undoubtedly facilitate the choice of the atoms and molecules most suitable for frequency standards.

III. METHOD OF NONLINEARLY ABSORBING GAS CELL

1. Idea of Method

In a gas laser with a standing light wave in the resonator, the field interacts resonantly with atoms whose velocity satisfy the condition[46,47]

$$\omega \pm ku = \omega_0, \tag{3.1}$$

where ω is the frequency of the light field, ω_0 the frequency of the center of the amplification line of the gas, and $\pm$u the projection of the velocity of the atom on the direction of propagation of each of the traveling waves forming the standing wave. If the amplitude of the field is sufficient to change the level population (to saturate), then the saturation will take place for atoms with $u = \pm(\omega_0 - \omega)/k$. As a result, two "holes" will appear on the plot of the gain against the frequency, in mirror-symmetry positions relative to the center of the line, at frequencies $\omega' = \omega$ and $\omega' = 2\omega_0 - \omega$, while a "dip" at a frequency $\omega = \omega_0$ will appear on the plot of the gain of the standing wave against its frequency. This "dip" can be interpreted as a consequence of the coincidence of the two holes at ku = 0. The occurrence of the dip in the center of the amplification line follows from intuitive physical considerations. If $\omega \neq \omega_0$, then the atom can interact resonantly with only one of the traveling waves. However, when $\omega = \omega_0$ both traveling waves change the population difference (gain) of the atom with ku = 0. Consequently, the degree of saturation is twice as large in this case. The dip in the center of the amplification line was theoretically investigated in detail by Lamb[47] and is called the Lamb dip. The dip was first observed experimentally in[48,59], an investigations of the dependence of the emission power of the gas laser on the field frequency. Figure 6 shows the experimental dependence of the output power of an He-Ne laser at the line λ = 1.15 μ when the distance between mirrors is changed by amount $\lambda/2$[48]. It was subsequently found that the plot of the output power against the frequency has an asymmetry[50], which is attributed to collisions

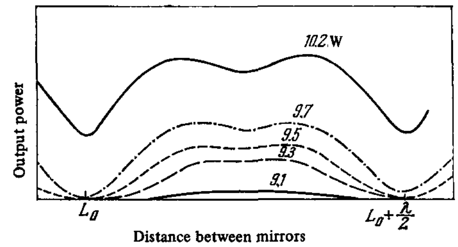


FIG. 6. Experimental dependence of the output power of an He-Ne laser at the λ= 1.15μ line when the distance between mirrors, L_0 = 50 cm, is changed by an amount $\lambda/2$, for difference levels of the high frequency excitation power [48].

of the atoms and to the correlation of the line broadening due to the interaction and the Doppler effect[51,52]. The width of the dip is equal to the homogeneous line width, which is usually determined by the radiative damping of the levels and by the collision broadening.

If a dip in the amplification line leads to a minimum of the gain, then an analogous dip in the absorption line leads to a maximum of transmission. This maximum can be quite narrow, since the gas pressure and the radiative damping of the transition can in principle be much smaller for an absorbing medium than for an amplifying medium. Physically this is connected with the fact that the absorption may occur at transitions from the ground state into the excited long-lived state, and the populations of all the remaining levels can be negligible. Essentially, in this case it is possible to obtain the lowest limit of the dip width, determined only by the finite transit time of the atoms through the light wave, i.e., values on the order of 10^3--10^5 Hz.

It was shown in[24] that if a nonlinear absorbing gas cell is placed inside a resonator (Fig. 7a), then the produced narrow dip is capable of stabilizing the laser emission frequency. This effect, which can be called "frequency self-stabilization," can be understood from Fig. 7b, which shows the line shapes of the amplifying and absorbing media in the resonator and the shape of the summary line. The amplification peak, equivalent

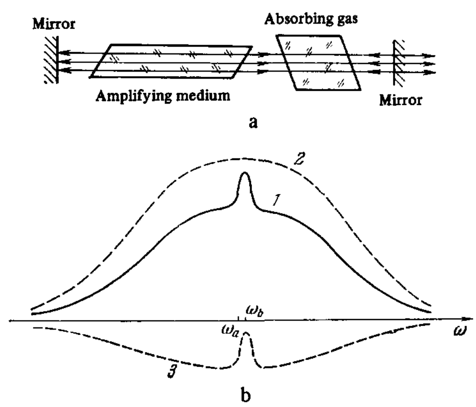


FIG. 7. Laser with nonlinearly absorbing gas cell in the resonator. a) Scheme, B) summary line shape of the amplifying and absorbing medium (1), line shape of amplifying medium (2), and line shape of absorbing medium (3) [24].

to the narrow amplifying line, ensures, in accordance with (2.1) a high value of the stabilization factor. This is the qualitative picture of the phenomenon.

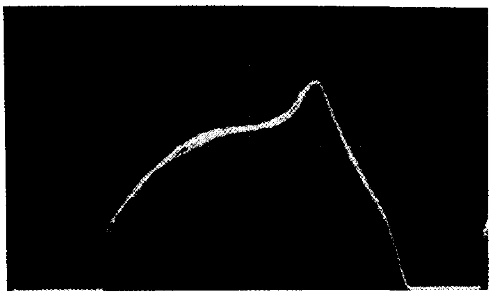


FIG. 8. Dependence of the ourpur power of an He-Ne laser ($\lambda = 6328$Å) with a neon absorbing cell in the resonator on the generation frequency, Horizontal scale–45 ± 5 MHz/division. The frequency increases from left to right [53].

Experiments on a gas laser with an absorbing cell in the resonator were performed independently in[53,54]. Figure 8 shows the dependence of the output power of an He-Ne laser at $\lambda = 6328$ Å, with a neon absorbing cell in the resonator, on the generation frequency, obtained in[63]. In these papers, notice was taken of the possibility of absolute stabilization of the laser frequency against the output-power peak, and also the possibility of measuring the homogeneous line, the broadening, and the line shift due to the pressure. In[54] it was also proposed to use absorbing molecules in the cell, and measurements were made of the dependence of the broadening and of the shift of the peak in Ne on the pressure in the absorbing cell. The region of stability of the stationary regime of an He-Ne laser with Ne absorbing cell, at the $\lambda = 6328$ Å line, was investigated in[148].

2. Conditions for Self-stabilization of the Frequency

The conditions for self-stabilization of the oscillation frequency were obtained in[24] (a more detailed analysis is presented in[55]). Let us consider them briefly.

Let a gas laser with an absorbing gas cell in a resonator with flat mirrors act on a single axial mode. Following Lamb[47] and calculating the polarization of the gas medium with accuracy to third order of perturbation theory in the field, we can obtain the following equation for the amplitude E, the frequency ν, and the phase φ of the standing wave:

$$\nu+\dot{\varphi}-\Omega=\varkappa_a c\,\frac{\omega_a-\nu}{ku_a}\left[1-\frac{a}{8}E^2\frac{\Delta\omega_a ku_a}{\Delta\omega_a^2+(\omega_a-\nu)^2}\right]-\varkappa_b c\,\frac{\omega_b-\nu}{ku_b}\left[1-\frac{b}{8}E^2\frac{\Delta\omega_b ku_b}{\Delta\omega_b^2+(\omega_b-\nu)^2}\right], \quad (3.2)$$

$$\dot{E}+\frac{\Delta\omega_r}{2}E=\frac{\varkappa_a c}{2}E\left[1-\frac{a}{4}E^2\left(1+\frac{\Delta\omega_a^2}{\Delta\omega_a^2+(\omega_a-\nu)^2}\right)\right]-\frac{\varkappa_b c}{2}E\left[1-\frac{b}{4}E^2\left(1+\frac{\Delta\omega_b^2}{\Delta\omega_b^2+(\omega_b-\nu)^2}\right)\right], \quad (3.3)$$

where the index a pertains to the amplifying component and the index b to the absorbing component, and where we have introduced the following notation: Ω (and $\Delta\omega_r$)—natural frequency (and line width) of the resonator, κ_a and κ_b—coefficients of amplification and absorption of the weak field per unit length at the generation frequency; ω_a and ω_b—centers of the amplification and absorption lines; aE^2 and bE^2—parameters of amplification and absorption saturation, defined by the expression

$$\alpha E^2=\frac{p_\alpha^2}{2\hbar^2\Delta\omega_\alpha\delta\omega_\alpha}E^2 \qquad (\alpha=a,\ b), \quad (3.4)$$

where p_α is the matrix element of the dipole moment of the transition, $\Delta\omega_\alpha$—homogeneous width corresponding to a transverse relaxation time T_2, and $\delta\omega_\alpha$—width corresponding to longitudinal relaxation time T_1[56,57]. The homogeneous width is made up of the radiative width $\gamma_a = \gamma_{\alpha 1} + \gamma_{\alpha 2}$ ($\gamma_{\alpha 1}$, $\gamma_{\alpha 2}$—radiative widths of the upper and lower levels), the width Γ_α due to the collisions (we consider the simplest model of Lorentz line broadening), and the width $\tau_\alpha^{-1} = u_\alpha/d_\alpha$, due to the finite time of stay of the atom in the light field τ_α (d_α—transverse dimension of the light beam):

$$\Delta\omega_\alpha=\gamma_\alpha+\Gamma_\alpha+\tau_\alpha^{-1}. \quad (3.5)$$

The width $\delta\omega_\alpha$ consists of the radiative width γ_α and the "flight" width τ_α^{-1}:

$$\delta\omega_\alpha=\gamma_\alpha+\tau_\alpha^{-1}. \quad (3.6)$$

Relations (3.5) and (3.6) take qualitative account of the line broadening due to the collisions and of the finite time of stay of the atoms in the light beam.

From (3.3) follow the self-excitation condition

$$\varkappa_a>\varkappa_b+\frac{\Delta\omega_r}{c} \quad (3.7)$$

and the condition for stability of the regime of the stationary generation*

$$\varkappa_a\left[1+\frac{\Delta\omega_a^2}{\Delta\omega_a^2+(\omega_a-\nu)^2}\right]>\varkappa_b\left[1+\frac{\Delta\omega_b^2}{\Delta\omega_b^2+(\omega_b-\nu)^2}\right]. \quad (3.8)$$

The generation frequency ν is determined from (3.2) with $\dot{\varphi} = 0$ and with a stationary field amplitude. The exact expression for ν is cumbersome, so that we first present a qualitative discussion. As seen from (3.2), in a weak field (at the threshold) the generation frequency is attracted to the center of the amplification line $[(\nu - \Omega)/(\omega_a - \nu) = \kappa_a c/ku_a]$ and is repelled from the center of the absorption line $[(\nu - \Omega)/(\omega_b - \nu) = -\kappa_b c/ku_b]$. When the field amplitude is increased, the attraction to the center of the amplification line gives way to a nonlinear repulsion (when $aE^2 > 8\Delta\omega_a/ku_a$ and $|\omega_a - \nu| < \Delta\omega_a$), while the repulsion from the center of the absorption line gives way to attraction (when $bE^2 > 8\Delta\omega_b/ku_b$ and $|\omega_b - \nu| < \Delta\omega_b$). The latter effect is quite appreciable, since the position of the center of the absorption line can be much more stable than the frequency of the center of the amplification line (at low gas pressure and small radiative width).

In order for the nonlinear attraction of the generation frequency to the center of the absorption line (self-

*This condition is confirmed qualitatively by experiments performed with an He-Ne laser with an Ne absorbing cell [148].

stabilization) to be predominant, it is necessary to satisfy several conditions. First, the generation frequency should lie in the self-stabilization region, i.e., within the limits of the homogeneous width of the absorption line

$$|\omega_b - \nu| < \Delta\omega_b. \tag{3.9}$$

Second, the effect of nonlinear attraction to the center of the absorption line should be appreciable, or, according to (3.2),

$$S_1 = \frac{8}{bE^2}\frac{\Delta\omega_b}{\varkappa_b c} \ll 1. \tag{3.10}$$

Finally, this effect should prevail over both the linear attraction and the nonlinear repulsion from the center of the amplification line. To this end, according to (3.2), it is necessary to satisfy the following two conditions:

$$S_2 = \frac{8}{bE^2}\frac{\varkappa_a}{\varkappa_b}\frac{\Delta\omega_b}{\Delta\omega_a} \ll 1, \tag{3.11}$$

$$S_3 = \frac{a}{b}\frac{\varkappa_a}{\varkappa_b}\frac{\Delta\omega_b}{\Delta\omega_a} \ll 1. \tag{3.12}$$

The conditions (3.9)–(3.12) guarantee high stability of the generation frequency in the vicinity of the center of the absorption line. This can be shown by direct calculation of the generation frequency. If the self-stabilization conditions (3.9)–(3.12) are satisfied, then the generation frequency is given by

$$\nu = \omega_b + S_1(\Omega - \omega_b) + (S_2 - S_3)(\omega_a - \omega_b), \tag{3.13}$$

where it is assumed that

$$(\varkappa_a c/ku_a) - (\varkappa_b c/ku_b) \cong \Delta\omega_r/ku_a \ll 1.$$

It is seen hence that when S_1, S_2, $S_3 \ll 1$, the generation frequency is stabilized in the region of the center of the absorption line ω_b.

Simultaneous satisfaction of conditions (3.9)–(3.12) is possible at a sufficiently small homogeneous width of the absorption line and a noticeable degree of saturation of the absorption. At an absorbing-gas pressure 10^{-3}–10^{-2} torr, it is possible to obtain realistically a dip width $\Delta\omega_b \cong 10^4$–$10^5$ Hz. The homogeneous amplification line width usually is $\Delta\omega_a \cong 10^7$–$10^8$ Hz, and the Doppler width is $ku_a \cong 10^8$–10^9 Hz. At $\varkappa_b c \cong 10^7$–10^8 Hz and at an absorption-saturation factor $bE^2 \cong 0.3$ (which still agrees with the considered approximation $bE^2 \ll 1$), it is perfectly realistic to obtain values $S_1 \cong 10^{-2}$ and $S_2 \cong 10^{-3}$*. Although the foregoing relations pertain to the case of weak saturation, the self-stabilization effect exists also in the case of strong saturation. However, a number of significant singularities arise here. First, the width of the dip in the absorption line increases in a strong field, since the homogeneous width is increased by the saturation[58]. The width of the dip in a strong monochromatic field $\Delta\omega_b'$ is determined by the expression[59]

$$\Delta\omega_b' = \Delta\omega_b(1 + bE^2)^{1/2}. \tag{3.14}$$

*The influence of the instability of the center of the amplification length can be greatly weakened by working under conditions when the nonlinear repulsion from the center of the amplification lines is compensated by a linear attraction. To this end it is necessary to have $S_2 = S_3$, or $aE^2 ku_a/8\Delta\omega_a = 1$.

From the point of view of the frequency stability, the optimal region is that of saturation $bE^2 \cong 1$. In this case, the attraction to the center of the absorption line is maximal, and the width is practically minimal. Second, saturation of the gain or of the absorption at the center of the line is determined by a more general expression[47,59], which is valid for all values of the saturation parameter, and the equation for the generation amplitude takes the form

$$\dot{E} + \frac{\Delta\omega_p}{2}E = E\left(\frac{\varkappa_a}{\sqrt{1+aE^2}} - \frac{\varkappa_b}{\sqrt{1+bE^2}}\right), \tag{3.15}$$

where $|\omega_a - \nu| \ll \Delta\omega_a$. We see therefore that when $b > a$ stationary values of the amplitude exist even when the self-excitation condition (3.7) is not satisfied. This corresponds to a hard self-excitation regime, which is not unexpected for a laser with a saturating absorber (see, for example,[60]). In this case the smaller of the stationary values of the amplitude is unstable. It corresponds to a threshold value of the amplitude $E_{thr} > 0$, which is necessary for the occurrence of self-oscillations. In addition, in such a laser pulsations of the radiation intensity are possible, if the self-excitation condition is satisfied, but the stability condition (3.8) is not. When $b\kappa_b \gg a\kappa_a$, the intensity pulsations have the same character as the pulsations occurring in the case of automatic laser Q switching[61,62].

3. Ultranarrow "Dips" in the Absorption Line

To obtain maximum frequency stability it is necessary to decrease the width of the absorption-line dip (3.5), which at a specified absorption coefficient per unit length

$$\varkappa_b = \frac{\lambda^2}{4}\frac{\gamma_b}{ku_b}N_b$$

is determined by the expression

$$\Delta\omega_b = \gamma_b + 4\frac{\varkappa_b}{\lambda^2}\frac{ku_b}{\gamma_b}\langle\sigma_b' u_b\rangle\frac{N_0}{N_b} + \tau_b^{-1}, \tag{3.16}$$

where σ_b' is the cross section of line broadening due to the collisions in the absorbing cell, N_0 is the density of all the molecules in the absorbing cell, $N_b = N_{1b} - N_{2b}$ is the density of the population difference of the two levels of the absorbing molecules, and it is assumed that the molecule mean free path is smaller than the cell dimension, and that the radiative width γ_b is due only to the working transition. It is easy to see that for a definite radiative width

$$\gamma_b^{\min} = \frac{2}{\lambda}\left(\varkappa_b ku_b\langle\sigma_b' u_b\rangle\frac{N_0}{N_b}\right)^{1/2} \tag{3.17}$$

a minimum of the homogeneous width $\Delta\omega_b$ equal to $2\gamma_d^{\min}$, is obtained. For example, at the wavelength $\lambda = 3\mu$, at $\kappa_b \cong 10^{-3}$ cm^{-1} and with the usual gas parameters $ku_b \cong 10^9$ sec^{-1}, $\langle\sigma_b' u_b\rangle \cong 2\times 10^{-10}$ cm^{-3} sec^{-1}, and $N_0/N_b \cong 10$ (for example, owing to the rotational structure of the molecules), the minimal homogeneous width due to the radiative damping and collisions amounts to $\Delta\omega_b = 2\gamma_b^{\min} = 6\times 10^2$ sec^{-1}.

In order to reach a minimal dip width, the time of interaction of the molecules traveling parallel to the wave front of the light ray should be $\tau_b \gtrsim (2\gamma_b^{\min})^{-1}$, i.e., the required effective interaction length is l_{eff}

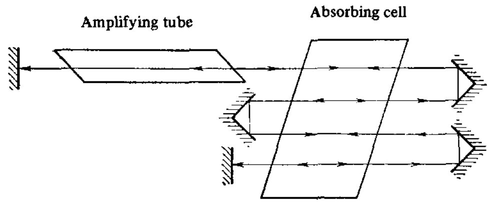


FIG. 9. Diagram of optical frequency standard with multiple passage through a nonlinearly-absorbing gas cell inside the resonator [55].

$\gtrsim u_b/2\gamma_b^{min}$. For the numerical example concerned above we have $\tau_b \gtrsim 1.6 \times 10^{-3}$ sec and $l_{eff} \gtrsim 70$ cm (at $u_b \cong 4 \times 10^4$ cm/sec). In principle, it is quite feasible to ensure such an interaction time, for example, by multiple parallel passage of the light ray through the absorbing cell (Fig. 9). In this case the largest saturation is experienced by molecules that cross several rays parallel to their wave front*. As a result, a dip appears in the absorption line, with a width (u_b/D), where D is the total path of the molecule from the first to the last ray. To be sure, in such a scheme the narrow dip is produced only by those molecules traveling parallel to the wave front, which intersect all the rays. This leads to an increase of γ_b^{min} by a factor of $\sqrt{D/d_b}$, where d_b is the diameter of one ray. In such a scheme, apparently, it is possible to attain a total dip width of several hundred Hz. If we exclude collisions, then the limiting width of the dip and the frequency stability in such a laser are the same as in a beam laser with coherent excitation. For example, at a low gas pressure in the absorbing cell (10^{-3}—10^{-4} torr) and at a large cell dimension, the stability factors S_1, S_2, and S_3 will amount to $S_1 \cong 10^{-4}$ and S_2 and $S_3 \cong 10^{-5}$. It is quite realistic to stabilize the center of the amplification line with accuracy 10^{-8}, and the center of the resonator line with accuracy 10^{-9}. In this case the relative frequency stability will be not worse than 10^{-3}. The absolute stability can be several times worse, owing to the hyperfine structure of the levels, to the shift of the center of the absorption line upon collision of the molecules with the walls, to the asymmetry of the line, to the shift of the dip as a result of the correlation of the Doppler broadening and the broadening due to the interaction[51,52], to the possible shift of the line in the light wave, etc. However, an absolute stability better than 10^{-11} is perfectly realistic.

We note that formation of an ultranarrow dip in the center of the gas absorption line under the influence of a standing light wave can be a very effective method of investigating the structure of the line inside the Doppler width[63]. If the absorption line is produced as a result of the overlap of several lines, and the distance between them exceeds the dip width $\Delta\omega_b$, then, by scanning the frequency of the standing light wave, it is possible to obtain minimum absorption in the center of each line. Apparently the limiting resolution of such a method is 10^{11}—10^{12}.

*In the case of strong saturation even an inversion of the populations of the absorbing molecules is possible, similar to the inversion of the molecules in the beam [26]. Then the dip in the absorption line becomes amplifying.

4. Pairs of Amplifying and Absorbing Media

To realize an optical frequency standard with a nonlinear absorbing gas cell, it is necessary to choose atoms or molecules having absorption at the generation frequency of known cw lasers. The simplest way is to use the same atoms or molecules as in the amplifying medium, but under conditions when there is positive absorption. In this case, the condition for the coincidence of the frequencies is automatically satisfied. Thus, in[53,54] the amplifying medium was an Ne-H gas mixture, and the absorbing medium was Ne (λ = 6328 Å). The pressure in the absorbing cell can be smaller by one or two orders of magnitude than in the amplifying cell. For the CO_2-N_2-He laser ($\lambda = 10.6\ \mu$) it is possible to use in the absorbing cell CO_2 at low pressure, for the He-Xe laser ($\lambda = 3.507\ \mu$) it is possible to use Xe at low pressure, etc. However, such pairs are certainly not optimal, since the absorption occurs at transitions between excited levels, the lower level being of necessity short-lived. This makes it impossible to reduce the gas pressure and to obtain narrow dips ($\Delta\omega_b < 10^5$ He). Therefore pairs with different amplifying and absorbing molecules are more promising.

The requirements with respect to the choice of the amplifying and absorbing molecules are here practically the same as in the case of a beam laser with coherent pumping (Sec. 2 of Ch. V). The pairs Ne-CH_4 and Xe-H_2CO, which were indicated in Sec. 2 of Ch. V, are therefore convenient. We can add to them the pair CO_2-SF_6 (the $\lambda = 10.59\ \mu$ line of the CO_2-N_2-He laser coincides with the strong 940 cm^{-1} absorption line of the SF_6 molecule[61]). Undoubtedly it is possible to choose a suitable absorbing molecule for the wavelengths of the continuous radiation of a H_2O submillimeter laser[64,65]. The indicated pairs do not make it possible to obtain the narrowest dips in the absorption line, since the probabilities of the radiative transitions are smaller by two orders of magnitude than the optimal values determined by relation (3.17), and therefore the necessary pressure in the absorbing cell is 10^{-2}—10^{-3} torr. The most convenient are atoms and molecules with a radiative transition probability $\gamma_b \cong 10^2$—10^3 sec^{-1}. Further developments of gas lasers will undoubtedly reveal optimal pairs and will make it possible to produce optical frequency standards in the bands from the visible to the submillimeter region.

IV. METHOD OF NONRESONANT FEEDBACK

1. Idea of Method

In quantum generators of the radio and optical bands, the positive feedback is resonant.[1,2,12,14] This is consequence of the use of a resonator (cavity in the radio band and Fabry-Perot in the optical band), which have minimum losses of electromagnetic energy in relatively narrow frequency intervals. Therefore quantum generators with resonant feedback emit one or several modes, which usually interact weakly with one another and can be regarded as isolated.

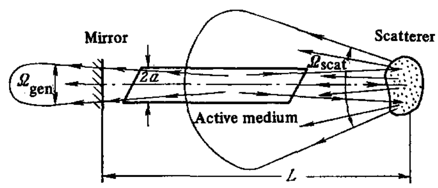


FIG. 10. Diagram of laser with nonresonant feedback [25].

A laser in which the positive feedback is nonresonant is also possible[25]. This means that the lifetime of the photon inside the generator does not depend on the frequency. The simplest method of obtaining nonresonant feedback is to use a scatterer in lieu of one of the mirrors[25,66]. The feedback, i.e., the return of part of the energy to the generator, is by backward scattering of the radiation (Fig. 10). The scattering leads to a strong interaction and to radiative damping of a large number of modes with different wave-vector directions, and as a result there are no resonant properties in the "mirror-scatterer" system. Feedback with the aid of scattering is essentially an energy feedback, for unlike feedback with the aid of a Fabry-Perot resonator, the phase relations in scattering are random. In other words, what is produced inside the laser are not the usual steady-state standing waves, but a spatially-random light field, which interacts with the active medium as a whole.

The absence of resonant properties in the feedback causes the spectrum of the generated radiation to become continuous, i.e., it contains no discrete components at selected resonant frequencies. The only resonant element in the laser is the resonant amplification line of the active medium. Therefore, after the threshold is reached, the generation spectrum "contracts" continuously towards the center of the amplification line of the medium, and in the ideal case the limiting width of the spectrum is determined by the fluctuations. The average frequency of the generated radiation does not depend on the dimensions of the laser and is determined only by the frequency of the center of the amplification line. If this frequency is sufficiently stable, then the laser emission has a stable frequency. It was therefore proposed in[25,66] to use the method of nonresonant feedback to produce an optical frequency standard.

2. Conditions of Nonresonant Feedback

The "scatterer-mirror" system is the simplest example of a "stochastic resonator," meaning an arbitrary cavity with a large number of modes (waves of different directions) which are strongly coupled by the scattering and have large radiation losses. The large radiation losses and the strong interaction of the mode leads to a complete overlap of their frequency spectrum. The "mode" concept loses in this case the usual meaning, and the spectrum becomes continuous. If the number of interacting "modes" N is sufficiently large, then the feedback becomes nonresonant[25,66]. For example, if the mirror and the scatterer have a diameter D and are separated a distance L, then the number N of the mode coupled by the scattering is given by

$$N \simeq \frac{\Omega_{gen}}{\Omega_{dif}}, \tag{4.1}$$

where Ω_{gen} is the solid angle of the generated radiation, $\Omega_{dif} \cong (\lambda/D)^2$ is the diffraction solid angle. The spectral mode density P_ω and the average distance between them $\delta\omega = P_\omega^{-1}$, according to the Rayleigh-Jeans formulas, are equal to

$$P_\omega = N\frac{2L}{c}, \quad \delta\omega = \frac{c}{2L}\frac{1}{N}. \tag{4.2}$$

Scattering causes the radiation to be transferred from one mode to the remaining N − 1 or to the open space. The transfer of the radiation to the modes of the "stochastic resonator" does not cause loss of the energy in the system as a whole, but leads to mode interaction. The rate of radiation loss Γ in a mode as a result of the transfer of the photons to the other N − 1 modes is given by

$$\Gamma = \frac{c}{2L}\ln\left(\frac{\Omega_{gen}}{\Omega_{dif}}\right) = \frac{c}{2L}\ln N. \tag{4.3}$$

The transfer of radiation to the open space and absorption by the mirror and by the scatterer determine the radiative loss of the system. The resultant rate of radiation loss is

$$\gamma = \frac{c}{2L}\ln\left(\frac{1}{\alpha r}\frac{\Omega_{scat}}{\Omega_{gen}}\right), \tag{4.4}$$

where α is the albedo of the scatterer, r the reflection coefficient of the mirror, and Ω_{scat}—solid angle of the back scattering, it being assumed that $\Omega_{scat} \gg \Omega_{gen}$.

With the aid of the introduced attenuation constants it is possible to write down the condition under which the feedback is nonresonant in the form:

$$\Gamma, \gamma \gg \delta\omega. \tag{4.5}$$

The necessary condition for this is $N \gg 1$. In the lasers described in[25,66], $N \cong 10^5$ and the feedback can be regarded as nonresonant. The larger the number of interaction modes, the more effective the "destruction" of the resonant properties. From this point of view, the most convenient laser is one having a maximum generation solid angle ($\Omega_{gen} = 4\pi$), considered in[67,68]. It represents an aggregate of scattering particles having negative absorption, or immersed in a medium with negative absorption. At a generating-region diameter $D = 1$ cm and $\lambda = 10^{-4}$ cm, the number of interacting modes in the field reaches 10^8–10^9.

At small generation angles, when N is small (for example at $\Omega_{gen} \cong (\lambda/D)^2$), the mode overlap is small and the appearance of random "resonance" at certain frequencies is possible[69]. Such resonances were experimentally observed in[70], in which a diffuse mirror was employed, at $N \cong 1$. In such cases, the concept of nonresonant feedback, of course, becomes meaningless.

3. Self-excitation Condition

The condition for the self-excitation condition of a laser with nonresonant feedback has the usual form: the rate of radiation loss equals the rate of growth of the radiation as a result of the quantum amplification. A distinguishing feature is that the radiation loss must be taken to mean only the loss connected with the escape of the radiation to the open space. For a laser with a "stochastic resonator" in the form of a scatterer and a mirror (see Fig. 10) the rate of growth of radiation by amplification $\gamma_{amp} = c/L \ln K$ (K is the gain per pass), and the self-excitation condition $\gamma_{amp} = \gamma$ takes the form[25,66].

$$K^2 \alpha r \frac{\Omega_{gen}}{\Omega_{scat}} = 1. \quad (4.6)$$

In the laser in the form of an aggregate of scattering particles with negative absorption[67,68], the loss is due to leakage of the photons by diffusion in the generation region (it is assumed that the mean free path of the photon due to scattering is much smaller than the dimensions of the system). These losses are offset by multiplication of the photons due to the negative absorption. The generation threshold in such a laser is perfectly analogous to the condition for the criticality of neutron multiplication in a homogeneous nuclear reactor[71]. At a specified negative absorption cross section per particle Q_a and a photon mean free path due to scattering Λ_s, there exists a critical dimension of the generation region. For a spherical distribution of the aggregate of the scattering particles, the critical radius R is determined by the expression

$$R = \pi \sqrt{\frac{\Lambda_s}{3N_0Q_a}}, \quad (4.7)$$

where N_0 is the number of scattering particles per unit volume.

4. Emission Spectrum

In a laser with a Fabry-Perot resonator, generation occurs after the threshold is reached at frequencies corresponding to the spatial resonances of the electromagnetic field in the resonator. Generation begins with amplification of the spontaneous noise, but the Fabry-Perot generator ensures an effective development of only the standing waves corresponding to the resonator natural modes. Therefore narrowing down of the radiation spectrum in lasers with resonant feedback is quite rapid. The narrowing of the radiation spectrum in a laser with a nonresonant feedback is of an entirely different character. The absence of spatial resonances of the electromagnetic field causes in this case the narrowing of the spectrum to occur only as a result of the resonant character of the gain of the active medium. The instantaneous width of the emission spectrum at half-maximum at a time t after the start of the generation, is given by[72]

$$\Delta\omega(t) = \frac{\Delta\omega_0}{\sqrt{\alpha_0 ct/\ln 2}}, \quad (4.8)$$

where $\Delta\omega_0$ is the amplification line width at half-maximum, and α_0 is the stationary gain per unit length in the center of the line ($\alpha_0 = N_0Q_a$ for a laser in the form of an aggregate of scattering particles). The process of the narrowing of the radiation in a laser with nonresonant feedback is quite slow. For example, in the case of a ruby active medium ($\alpha_0 \cong 0.1$ cm^{-1}, $\Delta\omega_0 = 5 \times 10^{11}$ Hz), within a time $t = 10^{-3}$ sec the line narrows down only to 4×10^8 Hz (by a factor 10^3). Formula (4.8) for the width of the spectrum at the instant of time t after the start of the generation coincides with the formula for the width of the spectrum of the signal at the output of a traveling wave amplifier[12,73,74] with length l = ct and with the same active medium. For example, the indicated narrowing by a factor 10^3 can be obtained in an amplifier of length l = 300 km.

The narrowing of the emission spectrum of a laser with nonresonant feedback was investigated experimentally in[72] with a pulse ruby laser. Within a time on the order of 100–300 μsec, a narrowing of the emission spectrum by a factor as much as 400 was observed. The time variation of the narrowing agrees with the theoretical relation (4.8). Further narrowing can be obtained by using continuous generation.

The limiting width of the spectrum in the continuous generation regime can be estimated by using the following considerations. The generation frequency coincides with the center of the line, in the worse case, with accuracy to the average distance $\delta\omega$ between modes, determined by expression (4.2). In the case of quasi-static variations of the "distance" between the scatterer and the mirror, when the Doppler effect is insignificant, the "modes" replace one another in the vicinity of the line peak with width of the order of $\delta\omega$. This determines the width of the spectrum in the continuous generation regime. For example, in a continuously operating Xe-He laser with a diffuse mirror[75], $N \cong 10$ and the width of the emission spectrum is $\Delta\omega \cong 10$ MHz, in agreement with (4.2). Therefore extremely narrow lines can be obtained only in a laser with a very large number of interacting modes N.

If N is sufficiently large, then the narrowing of the spectrum continues to a limiting value determined by the fluctuations. In practice, apparently, the greatest role is played by fluctuations of the velocity of motion of the feedback elements (unlike the fluctuations of the position of the feedback elements in ordinary lasers), which leads to a random change (wandering) of the photon frequency as a result of the Doppler effect. For example, if the average random frequency shift in each scattering act is $\Delta\omega_{f1} \ll \Delta\omega_0$, then the limiting width $\Delta\omega_{min}$ of the laser spectrum at the half-maximum is given by

$$\Delta\omega_{min} = \left(\frac{2\ln 2\Delta\omega_0\delta\omega_0}{\sqrt{\alpha_0 L}}\right)^{1/2}. \quad (4.9)$$

The fluctuation width of the spectrum has not yet been reached experimentally.

5. Emission Coherence

The coherent or statistical properties of the emission of a laser with nonresonant feedback were investigated in[66,76,77]. It was shown that the statistical properties of such a radiation greatly differ from the statistics of emission of ordinary lasers. Moreover, the emission of a laser with nonresonant feedback is very close to the radiation of an unusually bright black body in a narrow spectral interval.

First of all, the radiation is spatially incoherent. This has been established from the absence of interference of the radiation passing through the slits[66]. Physically this means that the emission of a laser with nonresonant feedback, in analogy with incoherent equilibrium radiation, is a superposition of a large number of waves of different directions with random phases. However, the analogy with the equilibrium radiation is even deeper. In[76,77] it was shown theoretically and experimentally that the statistics of the fluctuations of intense radiation in a very narrow solid angle, corresponding to one radiation mode, coincide with the statistics of equilibrium radiation in one quantum state (one cell of phase space)[78,79]. In particular, the distribution function of the number of photons in one mode is described by

$$P(n) = \frac{1}{\bar{n}} \exp\left(-\frac{n}{\bar{n}}\right) \qquad (\bar{n} \gg 1), \tag{4.10}$$

where n is the average number of photons in the mode. Figure 11 shows the distribution of the fluctuations of the radiation intensity in a narrow solid angle (solid line—theoretical distribution (4.7)). When the number of modes N_0 in which the radiation is registered is increased, the amplitude of the fluctuations of the intensity decreases like $1/\sqrt{N_0}$, i.e., the same as for equilibrium radiation. However, the deviation from the statistics of the fluctuations of the equilibrium radiation takes place if one observes the laser emission in the entire solid angle. The amplitude of the fluctuations of the intensity of this radiation is much smaller than that of equilibrium emission with the same number of modes. Physically this is due to the fact that the saturation in the laser stabilizes the intensity of the entire radiation, permitting deep correlated fluctuations of the intensity in the individual mode.

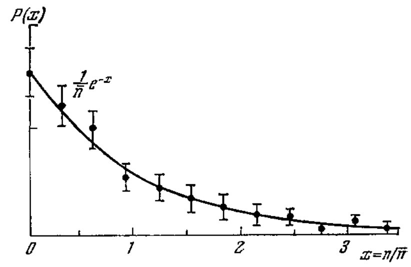


FIG. 11. Probability distribution of the fluctuations of the radiation intensity of a laser with nonresonant feedback in a narrow solid angle. Solid line—theoretical distribution [76].

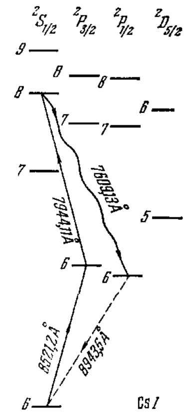


FIG. 12. Scheme of two-stage optical excitation of Cs vapor. Solid lines—exciting transitions, wavy line—generating transition, dashed line—relaxation transition [29].

6. Frequency Stability

The short-time frequency stability of the emission of a laser with nonresonant feedback is determined by the width of the spectrum $\Delta\omega$. Emission lines with a spectral width $\sim 10^3$ Hz and a short-time stability 10^{-11} are realistic. The long-time stability can be much higher. It is determined by the long-time stability of the center ω_0 of the amplification line of the active medium. To obtain high stability of the amplification resonance, it is necessary to use gaseous active media with very low pressure. Achievement of population inversion with a considerable amplification in gas at a pressure 10^{-3}—10^{-5} torr is apparently most probable when optical excitation is used. The main difficulty in optical excitation of atoms lies in the fact that it is necessary to choose coincident emission lines of the pump source and absorption lines of the active medium. There are very few such coincidences, and so far only one gas laser with optical excitation was realized, namely Cs vapor excited by the λ = 3880 Å line of a helium lamp[80,81]. However, the creation of sources of coherent radiation with variable frequency (for example, triple-coincidence semiconductor laser[41] or parametric generator[82]) and the use of two-stage optical excitation[29] can greatly change the situation.

By way of an example, let us consider the possible scheme of two-stage optical excitation of Cs vapor, the level scheme of which is shown in Fig. 12. The level $6^2P_{3/2}$ is populated by optical pumping with the λ_1 = 8521.2 Å resonance line of a cesium lamp. The population of this level can reach several percent of the population of the ground state. Radiation with wavelength λ_2 = 7944.11 Å then populates the $8^2S_{1/2}$ level. The source of such radiation may be a $Ga(As_{1-x}P_x)$ semiconductor laser diode[41], the emission frequency of which is adjusted to the same absorption line. The population of the $8^2S_{1/2}$ state can be of the same order as of the $6^2P_{1/2}$ state, since the pump radiation temperature is quite high. If the 8943.6-Å line of the cesium lamp is suppressed to such an extent that the population of the $6^2P_{1/2}$ state is small, then population inversion is produced between the $8^2S_{1/2}$ and $6^2P_{1/2}$ levels, and generation at the wavelength λ = 7609.13 Å is possible. At a Cs vapor pressure 10^{-4}—10^{-5} torr, we can expect a gain on the order of 0.1 cm^{-1}. To avoid a dependence of the central amplification line frequency on the pump-laser frequency fluctuations, the laser emission should be ideally isotropic (nondirectional) in the active medium. In this case, if the pump frequency drifts, only the gain changes, but not the frequency of the maximum gain. This optical-excitation scheme can be used for many atoms (Hg, Rb, and others) and makes it possible to produce in principle active media with very low pressure. The absolute frequency stability of the amplification line center in such media should be quite high.

V. METHODS OF STABILIZING THE FREQUENCY OF A GAS LASER

1. Principle of the Methods

In the methods considered above, the generation frequency stability was reached by obtaining a very large value of the stabilization factor $S = \Delta\omega_r/\Delta\omega_a$. These methods offer great possibilities, but much research is still necessary to obtain a stability better than 10^{-11}–10^{-12}. At the same time, the stability of ordinary gas lasers (for which usually $S \ll 1$) is only 10^{-7}. It is determined by the instability of the optical length of the resonator (the temperature instability changes the distance between mirrors, the instability of the pressure, temperature, and humidity change the refractive index of the air in the resonator, mechanical instability changes the inclination of the mirrors, the optical path length, and the optical path length of the Brewster windows of the discharge tube, etc.[83]). It is clear that the laser frequency stability can be greatly increased, even with a poor stability factor, by compensating for the instability of the optical length of the resonator. Such methods of frequency stabilization are being intensely developed recently, and they have made it possible to raise the frequency stability of gas lasers to a value on the order of 10^{-10} over several minutes and of the order of 10^{-9} over several hours (see the reviews[30,31]).

Common to all these methods is a resonant element to which the laser frequency is tuned. The resonant element in many methods is the resonance of the gain of the active medium of the laser itself, and in others it is resonance of absorption by an external cell or resonance of the interferometer transmission. Since the widths of the resonances are sufficiently large (usually 10^{8}–10^{9} Hz), the sensitivity of such methods does not make it possible to reach a stability much better than 10^{-10} (equality of the laser frequency to the maximum of the resonant element is reached with an accuracy 10^{-3}–10^{-4}, and in individual cases up to 10^{-5} of the resonance width). The stability of such a scheme cannot be better than the frequency stability of the resonant element. On the other hand, the frequency of the resonant element is influenced by pressure, temperature, etc. In addition, the stability is greatly influenced by the ratio of the bandwidth of the control system and the width of the spectrum of the short-time frequency fluctuations. We shall consider briefly methods of stabilizing the frequency of gas lasers and further possibilities of these methods.

2. Frequency Stabilization Against the Resonance of the Active Medium of the Laser

A number of stabilization methods are based on the dependence of the output power of the laser on the frequency difference between the generation and the center of the amplification line. This question was considered theoretically by Lamb[47], and studied experimentally in[48,49]; Fig. 6 shows the experimental result[48]. In a weak field, the output power is maximal at the center of the line, and with increasing field amplitude the line peak becomes flat, and a dip appears in the center of the line when the field is strong. All three cases (line peak, flat top, and dip in line center) are used in methods of stabilization by means of the atomic resonance of the laser.

In[84] the frequency of an He-Ne laser at the λ = 6328 Å line was stabilized against the peak of the output power. To obtain an error signal, one of the mirrors was oscillated with a small amplitude. The absolute stability of this laser was measured by an interferometer method, by comparison with the line of a krypton (Kr^{86}) wavelength standard. A reproducibility of about 2×10^{-8} over several months was attained.

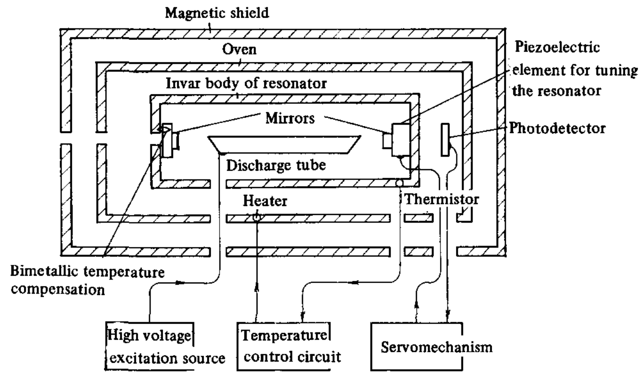


FIG. 13. Diagram of He-Ne laser (λ = 6328Å), stabilized against the dip in the center of the amplification line (Model 119 of the "Spectra Physics" firm). The diagram was taken from [31].

In[85,86], the frequency of an He-Ne laser at the λ = 1.15 μ line was stabilized against the "flat top." The error signals were obtained by sinusoidally modulating, at a small amplitude, the distance between mirrors. The fundamental, second, and third harmonics of the modulation frequency of the output power were used to correct the angular position of the mirrors, the excitation level of the discharge tube, and the distance between mirrors. The beat frequency of two lasers operating with the isotope Ne^{20} and Ne^{22}, equal to an isotopic shift of 260 MHz, was maintained constant within ± 1 MHz for several weeks, corresponding to a stability of 3×10^{-9}. An increase of the generation frequency with increasing gas pressure (+4.1 MHz torr) was observed.

A long-time stability of 2×10^{-9} in a day and a reproducibility of 10^{-8} were achieved by stabilizing the frequency of an He-Ne laser (λ = 6328 Å) against the dip. The diagram of such a laser, which is produced commercially (Model 119 of the "Spectra Physics" firm), is shown in Fig. 13. This diagram and the data are taken from the review[31]. Provision is made in this laser for temperature stabilization to prevent large changes of the length, as well as for magnetic screening to protect against stray magnetic fields. A dependence of the frequency shift on the pressure was observed with the aid of two lasers stabilized in this manner[27]. It is important that this shift depends on the output power of the laser.

A stabilization method based on the dispersion characteristics of the medium at the generation frequency was developed in[87]. This method is based on the dependence of the generation frequency on the field amplitude. If the gain of the medium (for example, the density of the inverted population is modulated, then frequency modulation of the radiation takes place when the generation frequency does not coincide with the center of the line. The amplitude of the frequency deviation is equal to zero only when the generation frequency coincides with the center of the line. In[87], the frequency of an He-Ne laser ($\lambda = 3.39\ \mu$) was stabilized by this method. A frequency stability of about 10^{-10} over eight hours was obtained. In[88] this method yielded a frequency stability of 10^{-9} over one hour. In the latter investigation, a dependence of the frequency on the pump power was observed, $\Delta\omega/\omega = 5 \times 10^{-7}\Delta p/p$. This dependence suffices to explain short-duration frequency fluctuations (2×10^{-9}) and long-time drift fluctuations (10^{-9}).

Another method of stabilizing the frequency was developed in[89]. The method is based on splitting the generation frequency of a single-mode gas laser in an axial magnetic field into two components that are circularly right- and left-hand polarized. These waves have equal intensity only when the splitting is symmetrical relative to the line center. It is clear that this effect can be used for frequency stabilization. The advantage of the method is that no laser parameter needs to be modulated. The theoretical sensitivity of the method is higher by two orders of magnitude than that of other methods, and makes it possible to reach a stability 2×10^{-12}. It must be borne in mind, however, that high sensitivity still does not mean high stability, since the line center itself has a long-range stability which is much worse than 10^{-12} (line shifts are produced by pressure, pump level, etc.). Experimentally, this method yielded a stability of 2.5×10^{-11} over eight minutes[90].

A sensitive method of frequency stabilization with the aid of the Zeeman effect in a laser was proposed and realized in[91]. This method uses the effect of "intersection" of the intensities of two axial modes of the laser in an axial magnetic field generating at two circularly and oppositely polarized components of an

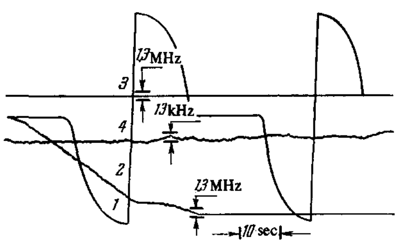


FIG. 14. Frequency drift of Xe-laser ($\lambda = 2.6\mu$). Curve 1—output signal discriminator vs. tuning of resonator. Curves 2 and 4 represent the frequency drift with and without the stabilization circuit. Curve 4—curve 2 with a frequency scale magnified 100 times. The Zeeman splitting is 685 MHz, and the distance between axial modes is 726 MHz [91].

atomic line. The distance between the axial modes is chosen approximately equal to the Zeeman splitting of the lines. The intensities of the mode radiation coincide only when the modes are symmetrical with respect to the center of the atomic line. A feature of this method is the possibility of generation in a wide range of frequencies off the center of the atomic transition. This scheme was used successfully to stabilize the frequencies of the 6328 Å and 1.153 μ lines of an He-Ne laser and the 2.65 μ line of an Xe laser. The best results were obtained for the Xe-laser, for which the stability was 10^{-10} for 100 seconds[91]. Figure 14 shows the frequency drift of this laser with and without the stabilization scheme, and also the characteristic of a frequency discriminator based on this method.

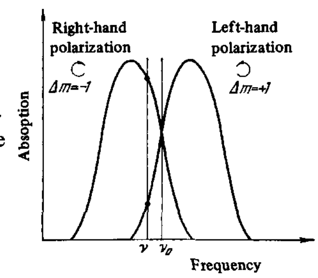
In[92] they proposed a laser frequency stabilization method based on phase modulation of the radiation in the resonator, at a frequency equal to the frequency interval between the axial modes of the laser. As a result of such a phase perturbation, the initial modes of the laser vanish and are replaced by a carrier and sidebands. The laser emission becomes free of the characteristic noise of the multimode laser. However, small beats remain in the radiation, with frequencies that are multiples of the phase modulation frequency. The beat level at the fundamental harmonic is very sensitive to the position of the carrier frequency relative to the center of the Doppler line. This effect can be used to tune the generation frequency to the center of the atomic resonance. In[93] this method was used to obtain long-range stability of 10^{-8} in an He-Ne laser.

3. Frequency Stabilization Against an External Resonant Element

An external resonant element (absorbing or amplifying cell, optical resonator) is used for stabilization in a number of methods. Thus, in[94] the resonant element is an absorbing neon cell, through which the beam of an He-Ne laser (λ = 6328 Å) is passed. In the presence of a magnetic field, the medium in the cell becomes dichroic for circularly polarized light. The difference between the absorption of left- and right-polarized light is determined by the deviation of the radiation frequency from the frequency of the center ω_0 of the absorption line (in the absence of a magnetic field) (Fig. 15). In a magnetic field of 350 G, the line splits by 1.2 GHz. By passing left- and right-circularly polarized light through the cell it is possible to obtain the frequency ω_0 of the center of the line. Two independent lasers stabilized by this method had a stability of 10^{-9}. Two such lasers were used to investigate the dependence of the frequency on the pressure, the discharge current, and the magnetic field[26]. The stability can be improved by using in lieu of the absorbing cell an amplifying cell with Zeeman splitting of the amplification line[95].

A high-Q Fabry-Perot interferometer can be used as the external resonant element.[96] An optical resonator has poor absolute long-range stability. However, it can be used to stabilize several lasers and to obtain a high relative stability. In[97], two lasers were stabilized against an external interferometer with a vibrating

FIG. 15. Shape of λ = 6328Å absorption line of He-Ne discharge tube in the presence of an axial magnetic field [94].



mirror, and a relative stability of 2×10^{-10} was obtained. It is possible to replace the external interferometer by an interferometer placed inside the optical resonator. In this case single-mode generation is obtained for a long time if the resonator is relatively long[98]. Two independent lasers stabilized by this method has a frequency stability 5×10^{-8}.

An interesting combination of two resonant elements with atomic resonance and with resonance of a Fabry-Perot interferometer for frequency stabilization was proposed in[99]. The atomic resonant element has good long-time stability, but the resonance width is quite

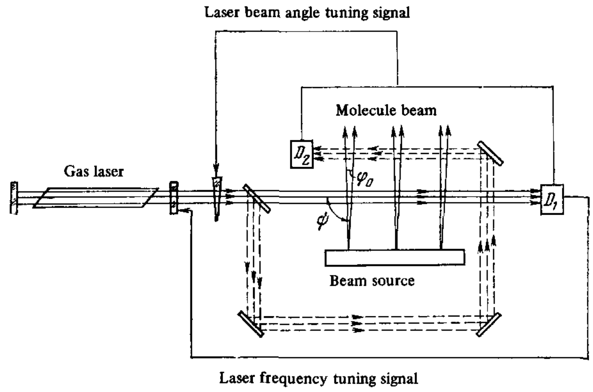


FIG. 16. Laser frequency stabilization against the absorption line of a molecule beam [33].

large (on the order of 1 GHz in the optical band). Tuning to the center of the resonance requires a relatively long time, but in this case it is impossible to compensate for the shoft-time fluctuations of the frequency. On the other hand, a resonant element in the form of a high-Q Fabry-Perot interferometer has a rather narrow resonance (to 10 MHz), good short-time stability, but insufficient long-time frequency stability. The advantages of the two resonance elements can be combined by stabilizing the laser frequency against the resonance of an external interferometer, and stabilize the resonance of the interferometer against an atomic resonant element. The first feedback link (laser—interferometer) has a large bandwidth and eliminates the short-time frequency fluctuations. The second feedback link (interferometer—atomic resonant element) has a narrow bandwidth and eliminates the slow drift of the interferometer resonance. Preliminary measurements of the frequency stability of two lasers stabilized by this method have shown that the stability is better than 3×10^{-10} in a second and 2×10^{-9} in 10^3 sec, and the reproducibility is 2×10^{-9}[100].

In[32] it was proposed to use as the external resonant element a beam of absorbing molecules. In this case, high sensitivity and high absolute stability of the resonance frequency are attained simultaneously. Laser frequency stabilization by means of a molecule beam is illustrated in Fig. 16. The real angular spread of the molecule beam (in the plane of the molecule and photon propagation directions) is $\varphi_0 \gg \lambda/a$, where λ is the radiation wavelength and a is the diameter of the laser beam. Therefore the width of the beam absorption line is $\Delta\nu/\nu_0 \cong \varphi_0 v_0/c$, where v_0 is the average molecule velocity. The divergence of the beam in the perpendicular plane is insignificant. The laser frequency ν_1 can be tuned to center ν_0 of the beam absorption line by standard means. For example, when the laser frequency is modulated, $\nu(t) = \nu_0 + \delta\nu \cos \Omega t$, intensity-modulated radiation reaches the detector D_1. The amplitude of the modulation with frequency Ω is proportional to the deviation $\nu_1 - \nu_0$, and can be used to tune the laser frequency. The accuracy of the tuning in modern schemes is $(\nu_1 - \nu_0) \lesssim 10^{-3}\Delta\nu$. When $\Delta\nu/\nu_0 \cong 10^{-8}$, the laser frequency stability is better than 10^{-11}.

This method, however, has the following feature. The frequency ν_1 of the center of the absorption of the light beam by the molecule beam depends on the angle ψ between the direction of the laser beam and the average direction of the molecule beam

$$\nu_1 = \nu_0 \left(1 - \cos\psi \frac{v_0}{c}\right). \tag{5.1}$$

To obtain $|(\nu_1 - \nu_0)/\nu_0| \lesssim 10^{-12}$ it is necessary to have $|(\pi/2) - \psi| \lesssim 10^{-4}$—$10^{-5}$ rad. This is a rather stringent requirement. However, it is possible to maintain the perpendicular orientation of the laser and molecule beams with such an accuracy automatically by the following method. A fraction of the laser beam is diverted and passed through the molecule beam in strictly the opposite direction (shown dashed in Fig. 16). The frequency of the center of the absorption line of the second laser beam is determined by the expression

$$\nu_2 = \nu_0 \left(1 + \cos\psi \frac{v_0}{c}\right). \tag{5.2}$$

It is clear the the modulation of the signal of the second detector D_2 has the same character as that of the signal of the first detector only when $\nu_1 = \nu_2$, i.e., when $\cos\psi = 0$. It is assumed that the laser radiation intensity is insufficient to saturate the absorption, and therefore there is no cross modulation of the second beam as a result of the first. By comparing the signals of the two detectors it is possible to set automatically the angle of the laser beam relative to the molecule beam. This stabilization scheme makes it possible apparently to reach a laser frequency stability 10^{-11}—10^{-12}. Of course, this method is applicable to lasers for which the absorbing atoms or molecules can be chosen. In addition, it is necessary that the beam have a noticeable absorption at reasonable molecule fluxes. Several suitable pairs were mentioned above in Sec. 5 of Ch. II and Sec. 5 of Ch. III.

We note that the stability can be increased further by a unique combination of a laser stabilized against a molecule beam with the coherent-pumping beam laser described in Ch. II. A possible scheme of such a combination of lasers is shown in Fig. 17. The emission of the stabilized gas laser is amplified with a quantum amplifier to a value sufficient for pulsed inversion of the molecules as they cross the beam. The radiation intensity needed at the output of the amplifier is determined by expression (2.7). After excitation, the beam has an extremely narrow amplification line, the width of which is determined only by the time of flight τ_0 of the molecules through the exciting laser beam (10^4—10^5 Hz). The beam of transformed atoms enters

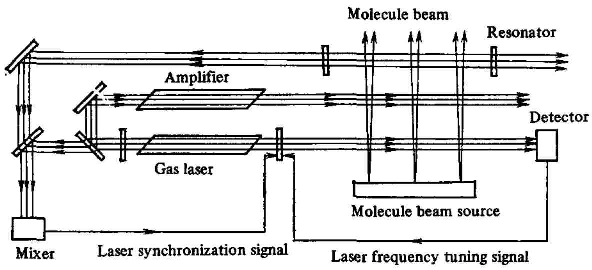


FIG. 17. Diagram of possible combination of a gas laser stabilized against a molecule beam and a coherent-pumping beam laser, having a common molecule beam [36].

the resonator, and the beam generated by the beam laser is synchronized in frequency with the beam from the stabilized gas laser. The stability of such a scheme is high as a result of the narrowness of the amplification line of the molecule beam. In addition, in such a combined scheme the self-excitation regime is "soft."

4. Method of Autoresonant Feedback

In the frequency stabilization methods considered above, the laser frequency is connected with the aid of an external feedback loop. It is possible to forego this loop by introducing into the laser an element that automatically tunes the generation frequency to the amplification-line center. This can be done with the method of autoresonant feedback proposed in[33]. By autoresonant feedback is meant feedback produced when the light wave is reflected from a spatial phase grating produced in the medium under the influence of the standing light wave of the laser itself. An "autoresonator" in the form of a mirror and a nonlinear spatial grating has the ability of tuning itself to the center of the resonance of the laser active medium.

A diagram of a laser with autoresonant feedback is shown in Fig. 18. The resonator is made up of a mirror and a spatial phase grating. The phase grating is a medium in which the refractive index varies periodically in space, and is produced in a nonlinear medium under the influence of a standing light wave A cos kz. If the refractive index of the medium depends quadratically on the intensity of the light wave, then the phase grating is of the form

$$n(z) = n_0 + \delta n(A^2)\cos^2 kz. \qquad (5.3)$$

Changes in the refractive index in the light field are caused by various nonlinearity mechanisms: thermal, striction, electrooptical, etc. (see, for example, the review[101]). The phase grating (5.3) reflects resonantly, in the backward direction, a light wave with a wave vector k. The reflection coefficient R in the region $R \ll 1$ is determined by the expression

$$R = \pi \frac{l}{\lambda}\delta n(A^2), \qquad (5.4)$$

where l is the thickness of the nonlinear medium. If it is recognized that in the given case the amplitude A of the standing waves is connected with the amplitude A_0 of the traveling wave by the relation $A^2 = \sqrt{R}A_0^2$, we finally get for the reflection coefficient

$$R = \left[\pi \frac{l}{\lambda}\delta n(A_0^2)\right]^2, \qquad (5.5)$$

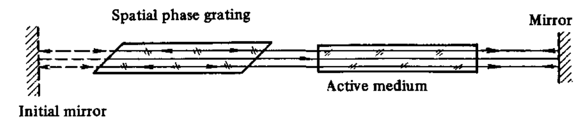


FIG. 18. Diagram of laser with autoresonant feedback. The gas line represents the mirror which is "turned off" after the spatial phase rating is produced [33].

The generation threshold is determined by the usual relation $rR(A_0^2)K^2 = 1$, where r is the mirror reflection coefficient and K is the gain of the active medium per pass. A laser with autoresonant feedback is an oscillating system with a hard self-excitation regime, since $R(A_0 = 0) = 0$. Its self-excitation calls for an initial light field with amplitude A_0 sufficient for the formation of a phase grating with a reflection coefficient $R(A_0^2) \geq 1/K^2r$. To this end it suffices, in principle, to obtain first laser generation with a resonator made up of two mirrors (see Fig. 18), and then eliminate one of the mirrors.

The dynamic properties and the frequency stability in the case of fluctuation motions of the mirror are investigated in[102]. It is shown that the natural frequency of the resonator is determined by the frequency of the generated radiation, and the generation frequency in the stationary regime coincides with the center of the amplification line. The frequency stability is determined essentially by the relation between the time constant τ (inertia) of the phase grating and the maximum frequency Ω_{max} of the fluctuation motions of the mirror. If $\Omega_{max}\tau \ll 1$, then the stability equals

$$\frac{\Delta\nu}{\nu} = \frac{\delta L}{L}\Omega_{max}\tau\frac{\Delta\omega_a}{\Delta\omega_r}, \qquad (5.6)$$

where $\delta L/L$ is the relative average magnitude of the displacements of the mirror relative to the phase grating, and $\Delta\omega_a$ and $\Delta\omega_r$ are the amplification and resonator line widths, respectively.

Thus, an appreciable gain in frequency stability can be obtained when $\Omega_{max}\tau\Delta\omega_a/\Delta\omega_r \ll 1$. For a thermal phase grating, the time constant is given by[102]

$$\tau \cong \frac{c\rho}{\varkappa(2k)^2}, \qquad (5.7)$$

where c, ρ, and κ are the specific heat, density, and heat conductivity of the medium of the phase grating, respectively. Realistically one can obtain $\tau \cong 10^{-5}$–10^{-6} sec. When the distance between the grating and the mirror fluctuates with frequencies up to 10^3–10^4 sec^{-1}, autoresonant feedback can produce a gain in the frequency stability by two–three orders of magnitude. The main difficulties in the experimental realization of such a scheme lie apparently in obtaining a sufficiently large gain per path at a power density up to 1 W/cm^2 and preventing self-excitation of the generator as a result of other types of feedback.

5. Prospects

By now, highly sensitive methods nave been developed to stabilize the generation frequency at the line center. For example, in a gas laser in a magnetic field it is possible to determine the center of the atomic line with accuracy of 10^{-5} of the line width[89]. It is therefore possible in principle to obtain good stability also at a poor value of the stability factor S. However, stabilization methods produce only a sufficiently high short-time stability. The long-time frequency stability in all the existing stabilized lasers is not better than 10^{-9}. This apparently can be naturally attributed to the fact that stabilized gas lasers and resonant cells have a pressure of several torr and are excited by the discharge in the gas. A noticeable dependence of the center of the atomic line on the pressure was observed (from several MHz to several dozen MHz per torr[26,27,28]), on the discharge current (several MHz per mA[26]), or on the pump power p ($5 \times 10^{-7}\,\Delta p/p$)[88]. To obtain a long-time frequency stability of 10^{-11} in the existing stabilized lasers, it would be necessary to have an exceedingly high stability of the pressure (down to 0.1%), excitation power, etc., which apparently is not attainable in practice.

Good long-time stability can be obtained by stabilizing the frequency of gas lasers with a low gas pressure. An example of such a laser is a cesium laser with optical pumping (gas pressure of approximately 10^{-2} torr)[80 81]. Therefore the progress of stabilized lasers will be determined by the success in developing gaseous active media with very low pressure. One of the possibilities of this kind was considered above in Sec. 6 of Ch. IV[29]. An analogous requirement must be satisfied also by the external resonant element. The most effective, although not always convenient in practice, is the use of a beam of absorbing molecules[32]. In this case we can expect high values of the short- and long-time stability.

VI. ABSOLUTE MEASUREMENT OF THE FREQUENCY OF OPTICAL OSCILLATIONS

1. Measurement of the Wavelength and Frequency of Electromagnetic Oscillations

In the radio band, up to the microwave region, it is easy to measure the frequency of electromagnetic oscillations, since detectors are available with time constants much shorter than the period of the oscillation. With the aid of radio interferometers it is possible to measure the wavelength of oscillations, but the accuracy with which the wavelength is measured is much lower than that of the frequency. In the optical band the situation is just the opposite. The duration of the period of the light oscillations is of the same order as the duration of the period of the oscillations of electrons in atoms and molecules, so that the frequency of the optical oscillations cannot be measured. On the other hand, the large ratio of the interferometer base to the light wavelength makes it possible to measure accurately (to 10^{-8}) the length of the light wave. The absolute accuracy with which the light wavelength is measured is determined in principle by the accuracy of the krypton length standard[18]. The frequency of the optical oscillations is determined from wavelength data and from the speed of light, which is known accurate to 10^{-6}:

$$c = (2.997925 \pm 0.000004)\cdot 10^{10}\ \text{cm/sec}^{103}.$$

In this sense, there is a difference in principle between the methods of the ratio band and the optical band. As a result of the recent progress in quantum electronics, particularly in the development of methods of generating coherent oscillations in the optical band, this gap has been greatly reduced. However, the frequency of the optical oscillations has not yet been measured. Further development of methods of quantum electronics and nonlinear optics will apparently enable us to solve this problem, too.

Thus, for example, recently Javan and co-workers succeeded in measuring the absolute frequency of four laser transitions in the submillimeter band[104,105]. In[104] they measured the frequencies of the 337 μ and 311 μ lines of a CN-gas cw laser[106] by mixing these lines with the 12th and 13th harmonics of a klystron with frequency 75 GHz. The measurement accuracy was several parts in 10^7. The obtained generation frequencies were

$$\nu_1 = 890.7595\ \text{GHz}, \quad \nu_2 = 964.3123\ \text{GHz}.$$

In[105] they succeeded in measuring the frequencies of the 190 and 194 μ lines of D_2O and C_2N_2 lasers[107]. The measurement was made by mixing with the 22nd and 23rd harmonics of a 70-GHz klystron. The obtained generation frequencies were:

$$\nu_1 = 1578.279\ \text{GHz and } \nu_2 = 1539.756\ \text{GHz}.$$

The measurement error was 1.5 MHz and was determined by the stability of the center of the laser lines. These experiments are the first stage in the path towards further measuring the frequencies of the submillimeter, infrared, and optical bands. Further progress in the measurement of frequency in the short-wave band can be made by converting the submillimeter frequencies into optical frequencies (successive coherent frequency multiplication) or by converting the optical oscillations into submillimeter ones (successive coherent frequency division). We shall discuss below the possibilities of realizing these methods.

2. Methods of Coherent Frequency Multiplication

The method of coherent frequency multiplication* for the purpose of measuring the frequency of light oscillations was proposed already by Townes[19]. The gist of this method reduces to the following: Oscillation of frequency ν_1 acts on a nonlinear element placed in the resonator of the quantum generator, whose generation frequency ν_2 is close to the second harmonic $2\nu_1$ (Fig. 19a). The nonlinear element (frequency doubler) can be placed also outside the resonator and a weak second-harmonic signal can be used to act on the quantum generator (Fig. 19b). The weak signal at the frequency $2\nu_1$ "captures" the generation frequency ν_2, and the result is intense oscillations at frequency $2\nu_1$, coherent with the oscillations at frequency ν_1. This procedure is then repeated several times. In order to cover the frequency band from Ω to $\omega \gg \Omega$, it is necessary to make $N = 3.3 \log(\omega/\Omega)$ successive frequency doublings. For example, to cover the range from 100 to 1 μ it is necessary to have seven frequency doublings.

*Frequency multiplication in the radio band is considered in detail in the book [146].

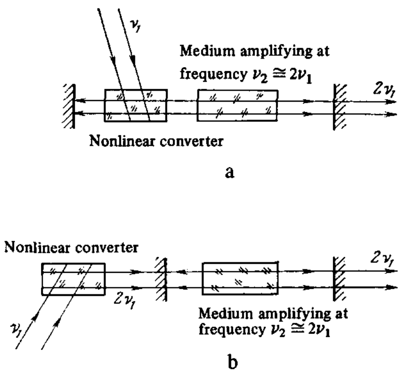


FIG. 19. Coherent frequency doubling with internal (a) and external (b) nonlinear frequency converters.

To realize the frequency locking regime, the frequency ν_2 should be sufficiently close to the frequency $2\nu_1$. If η is the ratio of the intensity of the synchronizing signal at the second harmonic frequency $2\nu_1$ to the intensity of the synchronized radiation, then the locking band is determined by the relation[108]

$$|\nu_2-2\nu_1|\cong\eta\frac{c}{L}=\nu_2\eta\frac{\lambda_2}{L}, \qquad (6.1)$$

where L is the resonator length. With the aid of cw lasers rated 1–10 W it is possible to obtain in the nonlinear element a conversion coefficient of the order of 10^{-4} [109,110]. At such values of η, the relative locking band is quite small ($\Delta\nu/\nu \cong 10^{-8}$–$10^{-11}$ at $\lambda/L \cong 10^{-4}$–10^{-5}), and consequently the laser generation must be stabilized also at the intermediate frequencies ν, 2ν, 4ν, etc.

Another serious difficulty is the choice of active media for the sequence of multiple frequencies ν, 2ν, 4ν, etc. If we are dealing with atomic and molecular laser lines, then coincidence of the lines is quite rare even for a single pair (ν, 2ν). For example, in[111], in an analysis of 880 laser transitions, only 11 pairs of wavelengths ν_1 and ν_2 coinciding within ±1 Å could be chosen. The pairs of transitions found in that investigation are listed in the table. The probability of coincidence turned out to be approximately 10^{-2}. Consequently, the probability of selecting a chain of five successive pairs ($\nu \rightarrow 2\nu \rightarrow 64\,\nu$) is 10^{-10}.

Pairs of laser transitions at multiple frequencies (fundamental and second harmonic; accuracy of wave coincidences $\Delta\lambda$ = ± 1Å)[111]

Fundamental frequency		Second harmonic	
gas	in Å units	gas	in Å units
Xenon	4954.10	Xenon	2477.18
Chlorine	6094.74	Argon	3047.0
»	6094.74	Oxygen	3047.15
Iodine	6127.0	»	3063.46
Carbon monoxide	6611.5	Xenon	3305.92
Ditto	6613.5	»	3306.4
Nitrogen	8886.5	Krypton	4443.28
»	10449.3	Mercury	5225.0
Neon	11180.6	Carbon monoxide	5590.6
Krypton	21165.0	Mercury	10583.0
Iodine	32360.0	Argon	16180.0

It is simplest to satisfy the condition for the agreement of the multiple frequencies in semiconductor active media[112]. The amplification line widths of semiconductors are quite large (on the order of kT[112]), and the position of the amplification line can be varied in a wide range with the aid of ternary semiconducting compounds[41], pressure[113], and a magnetic field[114]. By way of illustration, Fig. 20 shows the regions of the generation frequencies of semiconducting injection lasers according to data as of the end of 1967. We see that semiconductors cover practically the entire range

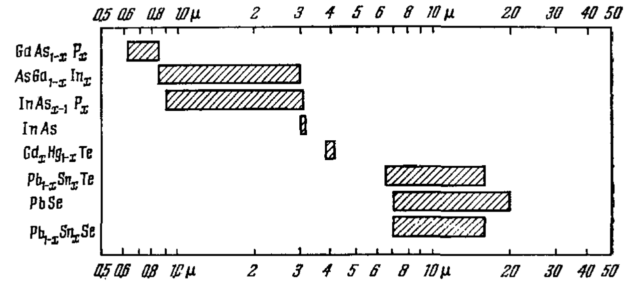


FIG. 20. Tuning range of the generation frequency of various semiconductor injection lasers. InAs lasers can be tuned with a magnetic field of 9 kG in a range 100Å [114], PbSe lasers can be tuned by a pressure of 20 kbar up to 21μ [113], and the remaining lasers can be tuned by varying the composition of the semiconducting compound [41].

from the far infrared to the visible. In the case of semiconductors, there are two mitigating circumstances. First, the ratio λ/L, and consequently the frequency locking band can be higher by 3–4 orders of magnitude than for gas lasers. For example, in[115] an ultrathin (2–4 μ) CdSe laser (λ = 6894 Å) was realized, with optical excitation by means of an injection laser. Second, in injection lasers it is possible to have internal generation of the second harmonic in the active medium itself, with a conversion coefficient up to 10^{-4} [116,117]. Nonetheless, the realization of a chain of coherent frequency multiplications, with the aid of semiconductors calls for an appreciable improvement of the parameters of the semiconducting lasers, particularly the development of single-mode semiconductor cw lasers operating at a specified stabilized generation frequency, and of semiconductor optical amplifiers.

The foregoing scheme of coherent frequency multiplication by successive doubling is based on known experimental data. In particular, it is assumed that there exists only a "weak" optical nonlinearity, i.e., a nonlinearity that leads to a very small change of the wave parameters over the wavelength. In this case, appreciable distortion of the wave occurs when the distortions accumulate over considerable distances and under conditions of spatial synchronism of the interaction[109,110]. In principle, there can exist also a "strong" optical nonlinearity, when the appreciable distortion of the wave occurs over distances comparable with the wavelength. It is difficult at present to indicate concrete mechanisms of a "strong" optical nonlinearity. However, one can attempt to discuss one of the effects which can serve as a possible candidate for this role, owing to the very low inertia (on the order of the period of the light). We have in mind the tunneling of electrons through a very thin barrier[118], particularly the p-n junction of a tunnel diode[119] in the field of a light wave. The scheme of the energy bands in a p-n junction is shown in Fig. 21a. The electrons from the conduction band can enter the valence band without loss of energy by tunneling through the barrier of the p-n junction. An external voltage changes only the probability of tunneling from the conduction to the valence band and back, and as a result current is produced through the p-n junction. The current-voltage characteristic of a tunnel diode is shown in Fig. 21b[120]. An essential feature of the tunnel-diode p-n junction is its small thickness L (100--200 Å) at an impurity concentration 10^{18}—10^{20} cm^{-3} [120]. As a result, the characteristic time of flight of the electron through the junction $\tau = L/v_e$ (v_e—electron velocity) can be quite small. For example, at L = 100 Å and $v_e = 10^8$ cm/sec, this time amounts to 10^{-14} sec, i.e., it is of the same order as the period of the optical oscillations. Assume that an electromagnetic polarized in a plane perpendicular to the p-n junction, and having a frequency $\nu \ll 1/\tau$, is incident on the p-n junction of a tunnel diode. Owing to the bend of the tunnel-diode characteristic at the origin, the current through the junction will be different in the positive and in the negative half-cycles. Consequently, the spectrum of the current will contain the second and higher harmonics, the amplitudes of which depend on the intensity of the radiation. To increase the second-harmonic amplitude it is possible to employ a so-called "inverted" tunnel diode, in which the semiconductor is degenerate only on one side of the p-n junction, for in this case the bend of the characteristic near the origin is particularly sharp[121]. The higher current harmonics can be obtained by applying simultaneously to the diode a constant bias voltage, such that the operating point is shifted to the maximum of the characteristic (point U_p in Fig. 21b). In all these cases the spectrum of the current contains harmonics, whose maximum number is in principle equal to $N \cong \frac{1}{2}\nu$. Their relative intensity is determined by the nonlinearity of the characteristic in the working band of the field amplitudes. Of course, it is hardly possible to use directly such high-frequency current oscillations, since they become essentially smoothed out inside the semiconductor. However, in the vicinity of the p-n junction, in a region with a depth on the order of the dimension of the skin layer, the current oscillations should lead to radiation of electromagnetic waves to the outside, with spectral components corresponding to the spectrum of the current through the p-n junction. The intensity of such an emission is apparently low, particularly at higher harmonics.

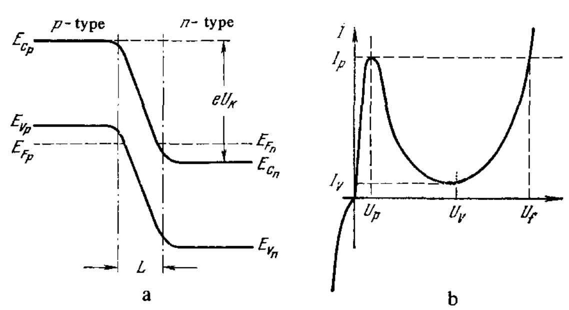


FIG. 21. Energy band scheme in a p-n junction (a) and current-voltage characteristic (b) of a tunnel diode.

3. Methods of Coherent Frequency Division

Coherent frequency division of light oscillations can be realized by parametric interaction of waves in optical parametric generators. Parametric generators of light waves were proposed in[122,123] and were recently realized in a number of laboratories (see[82]). Without stopping to discuss the operating principle and the construction of such generators, which are considered in detail in the substantial review[82], we note that in a parametric generator an intense light wave of frequency ω is coherently converted into waves with frequencies ω_1 and ω_2 satisfying the condition

$$\omega = \omega_1 + \omega_2. \tag{6.2}$$

In a degenerate parametric generator, exact division of the pump frequency takes place: $\omega_1 = \omega_2 = \omega/2$. By successively producing coherent frequency division in a chain of N parametric generators it is possible to obtain a frequency $\Omega = \omega/2^N$, which can be measured by radio devices. It is also possible to measure the frequency of optical oscillations with the aid of a chain of nondegenerate parametric generators. The use of nondegenerate generators greatly facilitates the choice of optical amplifiers at the intermediate frequencies. The smallest number of parametric generators necessary in the case when the degeneracy is so small that the difference frequency of the optical oscillations $\Omega_1 = \omega_2 - \omega_1$ can be measured by radio means. A system for measuring frequency with the aid of a chain of parametric generators with small degeneracy is shown in Fig. 22. The frequency of the initial oscillation experiencing N divisions is given by the relation

$$\omega = 2^N (\Omega_N + \Omega_{N+1}) + 2^{N-1}\Omega_{N-1} + 2^{N-2}\Omega_{N-2} + \ldots + \Omega_1, \tag{6.3}$$

where $\Omega_1, \Omega_2, \ldots, \Omega_{N+1}$ are the frequencies measured by radio means.

We note that it may be advantageous to combine simultaneous frequency division and multiplication to cover the entire range from 1 to 100 μ. For example, the frequency of a high-power CO_2 laser[124] $\lambda = 10.6\ \mu$ or $\lambda = 9.6\ \mu$ can be divided 3—4 times with the aid of parametric generators, covering thereby the 10—100 μ band. On the other hand, it is feasible to double the

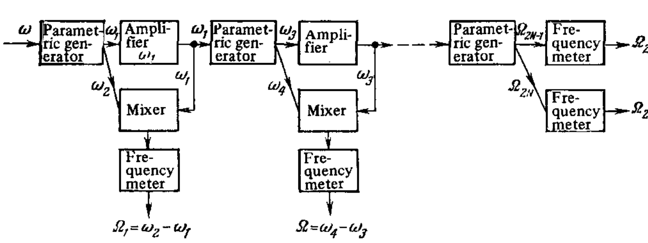


FIG. 22. System for measuring optical frequencies with a chain of parametric generators with small degeneracy.

frequency of this high-power laser 3–4 times and thus cover the range of approximately 1–10 μ.

In conclusion we note that progress in the absolute measurement of the frequency of optical oscillations is determined essentially by further progress in nonlinear optics.

4. Accuracy of Absolute Measurement of Light Frequency

It was noted in the introduction that in the optical band it is possible in principle to attain a higher frequency-measurement accuracy, for a given observation time, than in the radio band. However, this advantage cannot be directly realized, owing to the lack of direct methods of measuring optical frequencies. Indeed, let the optical frequency ω be measured by comparison with the n-th harmonic $(n \gg 1)$ of the radio-frequency oscillation Ω:

$$\omega = n\Omega + \Omega', \tag{6.4}$$

where Ω' is the measured difference of the optical frequency and of the n-th harmonic. If the radio frequencies Ω and Ω' are measured with accuracies $\Delta\Omega$ and $\Delta\Omega'$ respectively, then the relative accuracy of measurement of the optical frequency is

$$\frac{\Delta\omega}{\omega} = \frac{\Delta\Omega}{\Omega} + \frac{\Delta\Omega'}{\omega} \simeq \frac{\Delta\Omega}{\Omega}. \tag{6.5}$$

Thus, the relative accuracy of measurement of the optical frequency does not exceed the accuracy $\Delta\Omega/\Omega$ with which the radio frequency is measured. To obtain accuracies on the order of 10^{-14} it is necessary to have exceedingly large measurement times.

This difficulty can be circumvented by measuring the optical frequency ω by comparison with the frequency ω_0 of an optical heterodyne having a high frequency stability during the time T necessary for the absolute measurement of its frequency with a specified accuracy. Let the frequency of the optical heterodyne $\omega_0(t)$ be measured within a time T with the aid of coherent frequency division ω_0 to a frequency Ω or multiplication of the frequency Ω to ω_0, and let the maximum deviation of the heterodyne frequency during the time T amount to $\delta\omega_T$. Then the error in the measurement of the frequency ω is determined by the relation

$$\Delta\omega = \Delta\Omega' + \frac{\omega_0}{\Omega}\frac{1}{T} + \delta\omega_T, \tag{6.6}$$

where $\Delta\Omega'$ is the error in the measurement of the difference radio frequency of the heterodyne and the frequency $\omega(\Delta\Omega' \cong 1/\tau$—time of measurement of the difference frequency), $\omega_0/\Omega T$ is the error of the absolute measurement of the frequency of the heterodyne during the prolonged time T. The relative accuracy of frequency measurement with the aid of such a scheme is

$$\frac{\Delta\omega}{\omega} = \frac{1}{\tau\omega} + \frac{1}{T\Omega} + \frac{\delta\omega_T}{\omega}. \tag{6.7}$$

Thus, the measurement accuracy is determined essentially only by the frequency stability of the optical heterodyne during the time T of measurement of its absolute frequency. If an optical frequency standard is used as the heterodyne, then it is apparently realistic to expect a measurement accuracy $\Delta\omega/\omega \cong 10^{-13}$–$10^{-14}$ over relatively short time intervals, on the order of one second.

VII. CERTAIN POSSIBLE PHYSICAL EXPERIMENTS WITH THE AID OF OPTICAL FREQUENCY STANDARDS

1. Measurement of the Speed of Light

At present, the speed of light, one of the basic physical constants, is measured with an accuracy 10^{-6}. This accuracy can be greatly increased with the aid of an optical frequency standard and a system for the absolute measurement of the frequency of light. The idea of the experiment is simple[19]. Simultaneous high-accuracy measurement of the frequency and wavelength of the optical oscillations gives the speed of light with the same accuracy as is attained in the measurement of the wavelength and the frequency. The accuracy of this method is apparently limited more readily by the accuracy of the wavelength measurement than the frequency measurement. For example, an accuracy $\Delta\lambda/\lambda \cong 10^{-10}$ can be attained with the aid of an interferometer with a base $L = 10^3$ cm, at a measurement accuracy $\lambda/1000$, and at a wavelength $\lambda = 1\ \mu$. Consequently, it is perfectly sufficient to measure the frequency of the optical oscillations and to have an optical frequency standard with stability on the order of several times 10^{-11}. In this case the speed of light will be measured with an accuracy 10^{-10}. This is higher by two orders of magnitude than the accuracy of the present length standard[18]. Therefore, having such an exact value of the speed of light and having a time standard with accuracy better than 10^{-10}, we can forego an independent length standard[19].

The success of such an experiment depends mainly on the progress in the absolute measurement of the oscillation frequency in the visible and infrared bands.

2. Verification of the Constancy of the Universal Constants

Following an analysis of the dimensionless combinations of fundamental physical constants, including the radius and age of the universe, Dirac advanced the hypothesis[125] (see also[126,127]) that the physical constants can change with time as a result of the expansion of the universe.

This hypothesis can be approached from a different point of view[128]. The elementary particles make their own contribution to the mass and energy density, and thus influence, in accordance with general relativity theory, the curvature of the universe. Does the curvature of the universe react on the elementary particle? If the curvature changes, is this reflected in the masses of the particles or in other properties?

There is still no experimental proof whatever of the validity of this hypothesis. Moreover, as noted by Dirac[125], this point of view contradicts both the general and the special relativity theories. Several objections against this point of view were advanced by Ya. B. Zel'dovich[129].

However, the possibility of experimentally verifying the constancy of the universal constants is discussed to this day. Thus, Dicke analyzed many possibilities of verifying this hypothesis[17,126]. One of them is connected with the use of quantum generators as time standards. Optical frequency standards are capable of expanding the possibility of experiments of this kind.

If this hypothesis is valid, then $\alpha = e^2/\hbar c$ and other dimensionless ratios of physical quantities change with time as the universe expands. The reciprocal Hubble constant is equal to 1.4×10^{10} years, i.e., in 1.4 years the constant increases by 10^{-10}. During this time, certain dimensionless physical constants can experience changes, in accordance with Dirac's hypothesis, which do not differ greatly in order of magnitude from 10^{-10}. In this case, for example, two standard meters ($L_1 = n_1a_0$—length standard in the form of a rule, $L_2 = n_2\hbar c/Ry$—optical length standard, where $a_0 = \hbar^2/me^2$ is the Bohr radius, $Ry = me^4/2\hbar^2$ is the Rydberg constant, and n_1 and n_2 are integers), which differ by the dimensionless factor $\hbar c/e^2 = 137.0366 \pm 0.0005$, will no longer coincide. For experimental verification of the constancy of $\hbar c/e^2$ it is necessary to compare two such length standards with a sufficiently high degree of accuracy.*

Another possible experimental verification was indicated by Dicke[17] and consists of a comparison of the frequencies of two highly stable quantum generators operating at quantum transitions of different nature, i.e., which depend differently on the universal constant. Optical frequency standards may make feasible the formulation of experiments in accordance with such a scheme. A number of such experiments are discussed below.

Assume then we have a radio-frequency standard operating on the transition between the levels of the hyperfine structure of an atom (for example, a hydrogen maser at $\lambda = 21$ cm[3]) and a frequency standard in the submillimeter band, based on the transition between the levels of the rotational structure (for example, laser using the radicals CN or OH[64,65,106,107], but with very high frequency stability). What can we obtain from a comparison of the frequencies of these two quantum generators? The frequency of the hydrogen maser is determined by the relation[131]

$$\omega_{\text{hfs}} = \frac{4}{3}\alpha^3 g_I \frac{m}{M}\frac{c}{a_0}, \tag{7.1}$$

where a_0 is the Bohr radius and g_I is the gyromagnetic ratio of the proton.

The frequency of the rotational transition, for example for the singlet term of a diatomic molecule, is given by[132]

$$\omega_{\text{rot}} = \frac{\hbar}{I}(K+1), \tag{7.2}$$

where I is the moment of inertia of the molecule and K is the rotational quantum number. The moment of inertia of the molecule is $I \sim Ma_0^2$, and consequently the ratio of the frequencies of the hyperfine and rotational transitions are given by the relation

$$\frac{\omega_{\text{hfs}}}{\omega_{\text{rot}}} \sim \alpha^2 g_I. \tag{7.3}$$

It follows therefore that the exact comparison of the frequency of the hydrogen maser and of the rotational-transition laser, carried out over a sufficiently long time, can yield information on the time variation of the quantity $\alpha^2 g_I$, i.e., the fine structure constant and the gyromagnetic ratio.

A similar experiment can be carried out with two frequency standards operating on the transitions between the rotational and vibrational levels of molecules. The frequency of the vibrational transition of the molecule is given by[132]

$$\omega_{\text{vib}} = \sqrt{\frac{K_0}{M}}, \tag{7.4}$$

where K_0 is the elastic constant and M is the molecule mass. The elastic constant can be expressed in terms of the known constants, namely $K_0R^0 \sim E_e$, where R is the amplitude of the normal oscillation ($R \cong a_0$) and E_0 is the electron energy ($E_e \cong Ry$). As a result, we obtain for the ratio of the frequencies of the rotational and vibrational transition

$$\frac{\omega_{\text{rot}}}{\omega_{\text{vib}}} \sim \sqrt{\frac{m}{M}}. \tag{7.5}$$

An exact comparison of the frequencies of two such standards for a prolonged time can yield information concerning the constancy of another dimensionless quantity—the ratio of the masses of the electron and the nucleon. Similar information can be obtained from an experimental comparison of the frequencies of the vibrational transition of the molecule and the electronic transition of the atom.

To carry out the described experiments, it is sufficient to create frequency standards in the submillimeter and infrared bands, with a stability better than 10^{-11} in one month.

Comparison of the frequency of the submillimeter standard with the hydrogen maser with the aid of existing experimental methods is perfectly feasible[104,105]. To compare the frequency of standards on vibrational and rotational transitions, it is necessary to develop methods for the absolute measurement of the frequency of optical oscillations.

The development of highly stable optical frequency standards can apparently greatly increase the accuracy of interferometric measurements, and by the same token contribute to progress in the experimental observation of gravitational waves. Indeed, all the methods of observing gravitational waves (see the review[135]) are based on the measurement of very small mechanical displacements of bodies[136] or very small changes in the length of the optical path of a ray[137]. The accuracy with which small mechanical displacements are measured by non-optical methods at the present time is several parts in 10^{16} [138], i.e., much higher than the accuracy of optical methods. With the aid of optical frequency standards it is apparently possible to increase greatly the accuracy of the optical methods.

VIII. CONCLUSION

Optical frequency standards constitute a relatively new and promising trend in quantum radiophysics. Considerable progress can be expected in this region

*An attempt was made recently to observe astronomically the "constancy" of the fine-structure constant by observing the absorption spectrum of a quasistellar source [130].

in the next few years. Undoubtedly, new methods will be found for obtaining narrow and ultranarrow optical spectral lines, active media with very low pressure will be produced, effective methods will be developed for the absolute measurement of the frequency of optical oscillations, etc. In principle there are no limitations whatever to hinder to creation of highly stable frequency standards in the frequency range from the submillimeter band to the visible band.

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