Dips in emission lines of preliminarily polarized molecular (atomic) beams

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The conditions of appearance of a narrow dip in the center of a homogeneously broadened line are investigated theoretically and experimentally. It is shown that the physical pattern of formation of the dip is different for the cases of single-component and multi-component lines. The method may be employed not only for obtaining narrow lines, but also for resolving the structure of homogeneously broadend lines.

It is well known that a beam of previously polarized molecules spontaneously emits a highly monochromatic signal on subsequent passage through a resonator [1]. The signal frequency coincides with the frequency of the radiation that induces the polarization. In the present study we have investigated the line shape of the spontaneous-emission signal of a previously polarized molecular (atomic) beam as a function of the deviation between the frequency ω of the signal that induces the polarization and the natural frequency ω_L of the molecular transition.1) It turns out that under definite conditions the emission line shape has a minimum (a "dip") at $\omega = \omega_L$. The mechanism whereby this dip is produced differs radically from the mechanism of the "Lamb" dip in gas lasers [2] and consists in the fact that the intensity of the coherent spontaneous emission in the resonator of a previously polarized beam is proportional to the square of the amplitude of the polarization $|P|^2 \sim (P')^2 + (P'')^2$, where P' and P'' are the real and imaginary components of the polarization, respectively. However, at the emission frequency $\omega = \omega_L$, the real part of the polarization $P' = 0$. And while P'' has a maximum at this point, the decrease of P''^2 with the detuning $\omega - \omega_L$ near ω_L, under conditions that will be described below, is less abrupt than the increase of P'^2. It is this which causes the existence of a radiation minimum ("dip") at $\omega = \omega_L$.

We have investigated the dependence of the parameters of this dip, namely its depth and width, on the power of the polarizing radiation, on the structure of the homogeneously broadened line employed, on the average velocity of the molecular beam, and on the distance between resonators.

On passing through the first resonator with a field intensity $E_1(\omega)$, the molecule beam acquires a polarization[1]

$$P_1(\tau_1) = |\mu|^2 E_1[\delta(\cos\gamma\tau_1 - 1) + i\gamma\sin\gamma\tau_1] / \hbar\gamma^2, \tag{1}$$

where $\gamma = (\delta^2 + |\mu|^2 E_1^2/\hbar^2)^{1/2}$, $\delta = \omega - \omega_L$, μ is the matrix element of the molecule dipole moment, $\tau_1 = l_1/v$, l_1 is the length of the first resonator, and v is the molecule velocity. The beam of molecules then traverses the space between the two resonators and enters the second resonator, where energy is radiated as a result of the polarization acquired in the first resonator. This radiation is called "molecular ringing."

The amplitude of the field E_2 in the second resonator, in accord with [3], is determined by the relation

$$E_2 = 4\pi N D_0 Z_{\text{eff}}^{-1} \left\langle \frac{\exp\{-i\delta\tau_2\} - 1}{-i\delta\tau_2} e^{-i\delta T} P_1(\tau_1) \right\rangle_v, \tag{2}$$

when the effective resistance of the resonator, when tuned to the frequency ω, is equal at a low regeneration level to $Z_{\text{eff}} = -i/Q$, where Q is the figure of merit of the second resonator, $\tau_2 = l_2/v$, l_2 is the length of the second resonator, $T = L/v$, L is the distance between resonators, D_0 characterizes the difference between the populations of the working levels of the molecules on entering the first resonator, N is the density of the active molecules in the second resonator, and the symbol $\langle\ \rangle_v$ denotes averaging over the velocities.

Calculation of the dependence of the spontaneous-emission power in the second resonator, which is proportional to $|E_2|^2$, determines the shape of the radiated line and the sought parameters of the "dip."

We consider first a model of a single-component line for a monochromatic molecule beam. Then $|E_2|^2 \sim |P_1(\tau_1)|^2$. As follows from (1), the polarization $P_1(\tau_1)$ induced by the field has an oscillating character as a function of the amplitude of the field E_1 and the detuning δ. At small values of E_1, it turns out that when the detuning is increased the quantity $|E_2|^2$ decreases, i.e., the power of the spontaneous emission will be maximal at the line center. However, one can choose a value of E_1 such that when $|\delta|$ is increased the value of $|E_2|^2$ increases in the vicinity of the zero value. The molecule radiation power in the second resonator will then also increase following detuning from the line center. In other words, a narrow dip should appear at the center of the line.

The amplitude of the dip becomes maximal when $|P_1(\tau_1)|^2 = 0$. On the basis of formula (1) we have

$$|P_1(\tau_1)|^2 \sim \frac{2\delta^2(1 - \cos\gamma\tau_1) + \gamma_0^2(1 - \cos 2\gamma\tau_1)/2}{(\delta^2 + \gamma_0^2)^2}, \tag{3}$$

where $\gamma_0^2 = |\mu|^2 E_1^2/\hbar^2$. It follows therefore that at $\delta = 0$ the dip will be maximal (equal in amplitude to the "molecular ringing" line itself) at $\gamma_0\tau_1 = \pi n$ ($n = 1, 2, 3 \ldots$). So large a value of the saturation γ_0 makes it possible to calculate analytically the half-width of the dip and the distance between the two maxima of the "molecular ringing" line, for when $\gamma_0 \geq \delta$ and $n = 1$ the trigonometric functions in (3) admit of the obvious transformations of the type $\cos\gamma\tau_1 = \cos(\gamma_0\tau_1 + \delta^2\tau_1/2\gamma_0) = -\cos(\delta^2\tau_1/2\gamma_0)$. Then, expanding the trigonometric functions obtained via such transformations in powers of the parameter $\delta^2\tau_1/2\gamma_0 < 1$, we obtain

$$|P_1(\tau_1)|^2 \sim \frac{4\delta^2 + \delta^4\tau_1^2/4 - \delta^6\tau_1^2/4\gamma_0^2}{(\delta^2 + \gamma_0^2)^2}. \tag{4}$$

From the condition $\partial|P_1(\tau_1)|^2/\partial\delta = 0$ we obtain the half-

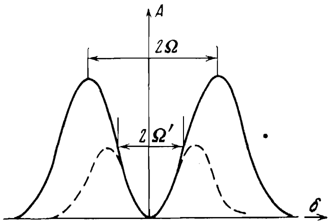

FIG. 1. Line shape of molecular ringing: solid curve—for a monochromatic beam, dashed–dashed—for a monochromatic beam.

distance $\Omega \approx 2/\tau_1$ between the maxima of the "molecular ringing" line. After determining the value of $|P_1(\tau_1)|^2_m$ at the maximum point and substituting $|P_1(\tau_1)|^2_m/2$ in formula (4), we obtain the half-width of the dip $\Omega' \approx 1.2/\tau_1$.

It is of interest to trace the variation of the parameters of the dip following introduction of averaging over the velocities, inasmuch as in practice one usually deals with beams that are not monochromatic. In this case, when calculating the dip parameters, it is necessary to take into account the phase factors $[\exp(-i\delta\tau_2) - 1]\exp(-i\delta T)/(-i\delta\tau_2)$. It is convenient to use in the calculations a distribution of the type $F(v) \sim v^{-3}\exp(-\bar{v}/v)dv$ ($\bar{v}$ is the average velocity of the molecules in the beam). Although it differs somewhat from a Maxwellian distribution, it permits, on the other hand, averaging over the velocities in analytic form and determining how allowance for the velocity distribution affects the parameters of the dip in principle. The calculations lead to the following results: at $T \gg \tau_1, \tau_2$ the condition of maximal dip depth remains unchanged, the half-distance between the maxima of the line is $\Omega \approx 1.13/T$, the half-width of the dip is $\Omega \approx 0.62/T$, and the half-width of the entire "molecular ringing" line is $\delta' \approx 1.45/T$. Thus as a result of addition of the radiation phases of the individual molecules in a nonmonochromatic beam in the second resonator, the frequency characteristics of the "molecular ringing" and of the dip in it become proportional to T^{-1} and not to τ_1^{-1}. The influence of the molecule velocity distribution leads to a change in the "molecular ringing" line shape, as shown in Fig. 1. One must not think, however, that the shape of the dip is strongly altered. The case of a beam that is monochromatic in velocity is represented by the solid line, and that of a nonmonochromatic beam by the dashed line.

We recall once more that in a single-component homogeneous line the condition under which the dip has a maximum is a large value of the saturation in the first resonator, $\gamma_0 = \pi/\tau_1$. The corresponding line broadening exceeds the transit half-width $\Delta\omega = 1/\tau_1$ for a monochromatic beam and is equal to the transit half-width $\Delta\omega = \pi/\tau_1$ for a beam with a Maxwellian distribution of the molecule velocities.

A different picture of dip formation (namely, in the absence of saturation) can occur in a multicomponent line, in which the distance $\Delta_{1,2}$ between the components is smaller than its homogeneous width. Let us consider the case of two components of equal intensity. Then the moduli of the polarizations of both components will be the same at the center of the line: $|P_1| = |P_2| = |P|$. The summary polarization at the line center is then expressed in the form

$$\mathscr{P} = |P|\exp\{i[\varphi_1 + \alpha_1 - \Delta_{1,2}T/2 - \operatorname{arctg}(\Delta_{1,2}\tau_2/4)]\} \cdot (1+\exp\{i[\Delta_{1,2}T+\varphi_2+\alpha_2-\varphi_1-\alpha_1+2\operatorname{arctg}(\Delta_{1,2}\tau_2/4)]\}],$$

where φ_1 and φ_2 are the phases of the polarizations of the components on entry into the first resonator and α_1 and α_2 are the phase shifts of the polarizations of the two components due to regeneration of the first resonator by the beam of active molecules. The influence of the regeneration can be taken into account approximately by regarding the effective amplitude of the field in the expression (1) for the polarization as equal to KE_1, where K is the complex gain of the active medium. Following the results of Vuylsteke [4], we express the gain in the form

$$K = \left\{-\frac{1}{|Q_M|} + i\frac{1}{|Q_M|}\frac{\delta\tau_1}{2}\right\} \Big/ \left\{\left(\frac{2}{Q_1} - \frac{1}{|Q_M|}\right) + i\frac{1}{|Q_M|}\frac{\delta\tau_1}{2}\right\}$$

where Q_M is the equivalent Q of the inverted beam and Q_1 is the figure of merit of the first resonator. We estimate $|Q_M|$ by putting $\delta = 0$. We then obtain $|Q_M| = (k_0 + 1)Q_1/2k_0$ at $|K(0)| = k_0$. Thus, $\alpha = \pi - \tan^{-1}[(k_0 + 1)\delta\tau_1/2]$.

According to (1), the phase shifts φ of the polarization are determined from the equation $\tan\varphi = -\gamma\delta^{-1}\cot(\gamma\tau_1/2)$. It is therefore impossible to obtain the phase shifts in general form. However, if $\delta \gg \gamma_0$, then this equation admits of the solution $\varphi = \pi/2 + \delta\tau_1/2$. We have $\mathscr{P} = 0$ when $T = [(2n+1)\pi + \varphi_1 + \alpha_1 - \varphi_2 - \alpha_2 - 2\tan^{-1}(\Delta_{1,2}\tau_2/4)]/\Delta_{1,2}$ (n = 0, 1, 2). Consequently, the dip becomes maximal at

$$T = \frac{1}{\Delta_{1,2}}\left[(2n+1)\pi + \frac{\Delta_{1,2}\tau_1}{2} - 2\operatorname{arctg}\left(\frac{k_0+1}{4}\Delta_{1,2}\tau_1\right) - 2\operatorname{arctg}\left(\frac{\Delta_{1,2}\tau_2}{4}\right)\right]. \tag{5}$$

Thus, the interference of the phase shifts of the polarizations of the two components leads to formation of a dip at the center of a two-component homogeneously broadened line even in the absence of saturation in the first resonator. This dip will have a maximum depth in the case $\Delta_{1,2} \ll 1/\tau_1, 1/\tau_2$ at a distance between resonators

$$L_{min} = \bar{v}[(2n+1)\pi - k_0\Delta_{1,2}\tau_1/2 - \Delta_{1,2}\tau_2/2]/\Delta_{1,2}, \tag{6}$$

i.e., the amplitude of the dip is periodic with change of distance. Knowing the value of L_{min} we can determine the distance $\Delta_{1,2}$ between the line components. Allowance for the velocity distribution of the molecules, if it has a symmetrical form, should not change the final result in principle.

Unfortunately, it is impossible to determine analytically the dependence of the width of the dip and the width of the "molecular ringing" line on the distance between the resonators for a two-component line. However, a qualitative picture can be presented. We recall that a narrowing of the "molecular ringing" line occurs in a single-component line, owing to the interference between the phase shifts of the polarizations of the individual molecules. The presence of one more component leads to the need for taking into account the phase shifts of the polarizations of its molecules. Let us examine the line wings. In the wings, owing to the fact that $\delta_2 > \delta_1$ (or $\delta_2 < \delta_1$ for the second line wing), the polarization phase shifts of the second component are more smeared out in magnitude and therefore, upon averaging, their influence is smaller the closer they are to the line center. Consequently, the central region of the "molecular ringing" line "settles down," as it were. This should lead to an increase in the width of the dip and of the width of the "molecular ringing" line itself with increasing distance, but only up to a certain limit. The indicated values of the widths should be periodic with distance, as is also the amplitude of the dip.

Concerning the resolving power of this method, it should be noted in view of the foregoing that the interference of phase shifts having opposite signs, and consequently leading to the formation of the dip, occurs only at frequencies that lie between the frequencies of the components. Therefore the width of the dip should correspond approximately to the distance $\Delta_{1,2}$ between the components. Thus, the smaller $\Delta_{1,2}$, the narrower should be the dip, but the dip naturally becomes maximal at larger distances $L_{\min}$.

An experiment was performed to verify the main conclusions. A maser was used as a spectroscope with two resonators in tandem. In one case, the maser operated on a beam of ammonia ($N^{15}H_3$) molecules, since its principal transition lines have negligible hyperfine structure (within several tens of Hz [5]), so that these lines can be regarded as single-component. In the second, it operated with a beam of $N^{14}H_3$ molecules. In both cases, the resonators were tuned to the transition $J = K = 3$, the principal line of which for $N^{14}H_3$ consists in general of three components. Since the intensity of one of them is always small [6], this can be regarded as a two-component line. The line widths were determined mainly by the transit broadening.

The field was produced in the first resonator by using the harmonic of a quartz oscillator, the power level of which was specially monitored. The resonators had $Q_1 \approx 2000$ and $Q_2 \approx 5000$, and their lengths were $l_1 = 6$ cm and $l_2 = 8$ cm. The distance between them could be varied continuously from 3 to 12 cm. The small regeneration level in the second resonator was monitored by means of a movable diaphragm placed in the region between the resonators. Then, by smoothly blocking off the molecule beams, the absence of regeneration could be verified by watching the behavior of the line shape of the molecule emission in the second resonator.

The theoretical relations were calculated on the basis of formula (2). In the qualitative discussion above we used a distribution of the type $F(v) \sim v^{-3} \exp(-\bar{v}/v)dv$ in averaging over the velocities; this distribution enabled us to obtain an analytic result. In fact, however, the molecule velocity distribution in the beam is different. In this case it is determined by the parameters of the molecule-beam source and by the sorting system. A special investigation yielded the following expression for the distribution in the case of outflow from a source having the form of a hole in a thin wall:

$$F(v) \sim v^4 \exp(-v^2/\bar{v}^2)\,dv$$

where $0.5\bar{v} \lesssim v \lesssim 2\bar{v}$ (this velocity interval is governed by the influence of the sorting system [7]). It is impossible, however, to average expressions (1) and (2) over the velocities with such a distribution in general form. In the present study this was done by expanding the trigonometric and exponential functions of formulas (1) and (2) in power series.

Figure 2 shows the theoretical and experimental plots of the relative depth β of the dip in a single-component line (a) against the degree of saturation, and in a two-component line (b) against the distance L between resonators. We see that in the single-component line the dip starts to form at $\gamma_0\tau_1 > 1.7$ and reaches 100% at $\gamma_0\tau_1 \approx 3.1$, which, incidentally, agrees well with the calculations for a monochromatic beam. The experimental dependence was obtained for a source of the channel type. This explains a certain deviation from the theoretical curve. The maximum of the dip was not reached, since the power of the quartz-oscillator harmonic was not high enough. No dependence of the depth of the dip on the distance between resonators was observed in the experiments, in agreement with the theory.

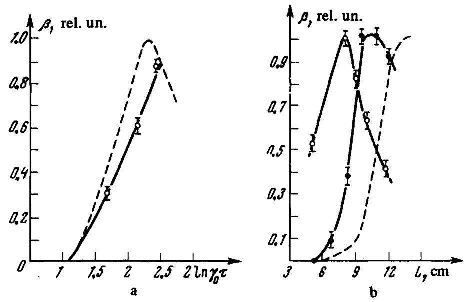

FIG. 2. Dependence of the relative depth of the dip: a—on the saturation for a single-component line; b—on the distance between the resonators for a two-component line (curve 1 (○)—for a channel-type source, curve 2 (●)—for a source in the form of a hole in a thin wall); solid lines—experiment; dashed—theory.

In Figure 2b, curve 1 (○) was plotted for a source of the channel type ($k_0 \approx 2$), and curve 2 (●) for a source in the form of a hole in a thin wall ($k_0 \approx 1.2$–1.5). The dip for a source of the channel type therefore reaches maximum depth at a smaller distance between the resonators for a net type source (see formula (6)). Both curves were plotted at fields much lower than saturating. The theoretical curve corresponds to $k_0 = 1$ and therefore lies to the right of curve 2. The periodicity of the depth of the dip as a function of the distance L is well illustrated by curve 1. Since the molecule velocity distribution in the case of a channel-type source is unknown, no theoretical relation corresponding to curve 1 is given.

Using formula (6) and knowing the values of $L_{\min}$ and k_0, we determined the distance between the two components of the homogeneously broadened line $J = K = 3$ of $N^{14}H_3$. It turned out to be 1500 ± 300 Hz, in good agreement with measurements made by other procedures [8]. Solving the transcendental equation (5), we can determine $\Delta_{1,2}$ with much higher accuracy.

The different physical pictures of dip formation in single-components and two-component lines lead to different dependences of the "molecular ringing" line width and of the width of the dip in the line on the distance L between resonators (Fig. 3). As follows from Fig. 3a (single-component line), the widths of the line and of the dip decrease with increasing L, i.e., approximately like T^{-1} and in sufficiently good agreement with the theoretical calculations. The saturation $\gamma_0\tau_1$ was calibrated against the condition for the dip in a single-component line at $\gamma_0\tau_1 \approx 1.7$.

At the same time, it is seen from Fig. 3b that at a small field amplitude both the width of the entire "molecular ringing" line and the width of the dip in it have an oscillating character as functions of L, which we have already explained above. On comparison of Figs. 3a and 3b, it can be noted that at high degrees of saturation the principal role in the formation of the dip in a two-component line is obviously assumed precisely by the mechanism of saturation in the first resonator. The values of the saturation in Fig. 3b are given in arbitrary units, inasmuch as there was no point against which to carry out the calibration in this case. The saturation value $x = 0.4$ is tantamount to absence of saturation, as monitored against the line shape in the

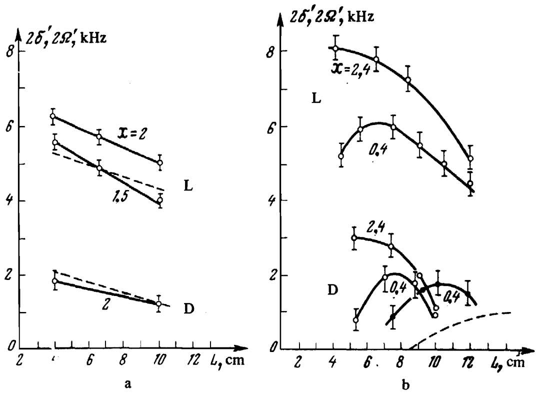

FIG. 3. Plots of the molecular-ringing line width (L) and of the dip in it (D) against the distance between the resonators: a—for a single-component line; b—for a two-component line; solid lines—experiment (○—for a channel-type source, ●—for a source in the form of an opening in a thin wall) at different x, dashed lines—theory for a: $\gamma_0\tau = 2.2$, for b: $\gamma_0\tau = 0.5$.

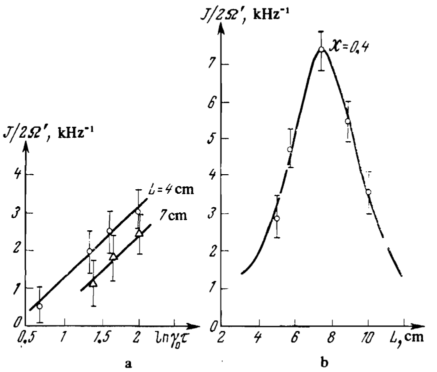

FIG. 4. Plots of slope of the dip: a—against the saturation for a single-component line and b—against the distance between the two resonators for a two-component line.

first and second resonators. The value $x = 2.4$ is given relative to $x = 0.4$. At this value of the parameter x, saturation was certainly attained. The curves drawn through the open circles were obtained for a channel-type source, and the curves drawn through the dark circles as well as the theoretical (dashed) curves were drawn for a net-type source.

Figure 4 shows the experimental dependence of the slope of the dip $J/2\Omega'$ (ratio of the signal/noise ratio for the dip to its width) for a single-component line (a) and for a two-component line (b). Such plots are of interest from the point of view of technical applications of narrow lines for the stabilization of lasers [9]. It can be seen from Fig. 4 that this ratio is larger for a two-component line. In principle, one should expect the dip to become much narrower and the slope of the dip much larger at a smaller distance between components. In concluding the analysis of the experimental data, we note that our experiments were not aimed at obtaining record-breaking results on the width of the dip. It served only as a clear illustration of the physical picture of dip formation in single- and two-component homogeneously-broadened lines.

If the line consists of two components, each of different intensity, then to determine the distance between them it is necessary to know, from some other considerations, the ratio between their intensities. On the other hand, if the number of components is more than two, then by measuring the values of L_{min} corresponding to pairwise interference of the components, and knowing the ratio of their intensities, it is also possible to calculate the distances between the components. Similar calculations differ only negligibly from calculations for the two components. From the number of dips, each of which has its own period of appearance, it is possible to assess the number of components of the homogeneously broadened line.

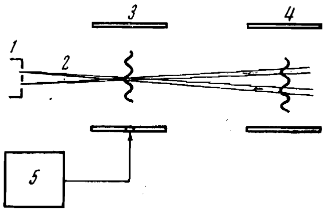

FIG. 5. Experimental setup for obtaining "molecular ringing" lines in the case of an inhomogeneously broadened line: 1—source of molecular beams; 2—molecule trajectories; 3, 4—resonators; 5—field source.

In addition to resolving the structure of the homogeneously broadened lines and determining the distance between the components of these lines, interest attaches to the following practical utilization of the dip in a two-component line. If the line corresponding to the emission or absorption of electromagnetic fields is single-component, and if application of, say, a magnetic field causes this line to split into two components, then when a system with two separate fields is placed in a constant magnetic field we are dealing in fact with a two-component line. In this case the width of the dip, which is approximately equal to $\Delta_{1,2}$, and the distance L_{min} are determined only by the applied magnetic field. It is convenient to stabilize the frequency of a laser against such a supernarrow dip.

For inhomogeneously broadened lines, one can advance arguments similar to those presented above. Let the molecule-beam direction be perpendicular to the axes of both resonators (Fig. 5). Then that fraction of molecules which experiences perturbation with one and the same phase during passage through the region of the first field acquires a polarization in this field. Consequently, these molecules radiate in the second resonator and produce "molecular ringing." According to the physical meaning of the "molecular ringing" effect, the corresponding line has a transit-governed width, since the averaging of the emission phases of the molecules polarized at frequencies that differ from the central line frequency by more than the transit width causes these molecules, in fact, not to produce a field in the second resonator. On going to higher frequencies, it is necessary to use open resonators, which have a large number of oscillation modes. It must be borne in mind, however, that all the described effects develop within the limits of the homogeneous width of the line at small detuning from its top. Therefore the molecules that participate in the formation of the dip will interact only with the one mode that is closest in frequency to the top of the line.

Thus, in the optical band it is possible in practice to resolve the structure not only of inhomogeneously broadened lines with the aid of the "Lamb" dip, which has a homogeneous width, but also to resolve the structure of homogeneously broadened lines. In addition, by using ultranarrow dips in homogeneously broadened lines and in the optical band, it is possible to stabilize the frequency of a laser. Dip widths on the order of 1 kHz or even smaller are attainable in this case.

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[1] The corresponding line width is homogeneous.

[1] A. N. Oraevskiĭ, Molekulyarnye generatory (Masers), Nauka, 1964.
[2] W. E. Lamb, J. Phys. Rev. **134A**, 1429 (1964).
[3] N. G. Basov and A. N. Oraevskiĭ, Zh. Eksp. Teor. Fiz. **42**, 1529 (1962) [Sov. Phys.-JETP **15**, 1062 (1962)].
[4] A. Buylsteke, Elements of Maser Theory, Van Nostrand, 1960.
[5] V. V. Nikitin and A. N. Oraevskiĭ, Radiotekhnika i êlektronika **7**, 859 (1962).
[6] K. Shimoda, J. Phys. Soc. Japan, **12**, 1006 (1957).
[7] K. Takcuhi, K. Shimoda, Y. Ogawara, Japan J. Appl. Phys. **5**, 608 (1966).
[8] A. F. Krupnov, V. A. Skvortsov, and L. A. Sinegubko, Izv. VUZov, Radiofizika **10**, 142 (1967).
[9] A. S. Bashkin, Kvantovaya êlektronika No. 5, 3 (1971) [Sov. J. Quant. Electr. **1**, 437 (1972)].

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95