Nonlinear narrow density resonances of excited molecules in a standing light wave

V. S. Letokhov and B. D. Pavlik
Institute of Spectroscopy, USSR Academy of Sciences
(Submitted October 6, 1972)
Zh. Eksp. Teor. Fiz. **64**, 804-812 (March 1973)

The existence of a resonant variation of the number of particles in an excited level when the frequency of a standing light wave passes through the center of a Doppler-broadened absorption line is shown. The parameters (width and amplitude) of the narrow particle-density resonance are found. The resonance is shown to differ from the Lamb dip in the nonlinear-amplification (absorption) factor. It is found that in the case of molecular transitions rotational relaxation can appreciably enhance the effect. Schemes for the observation and some applications of the narrow excited-particle-density resonances are considered.

1. INTRODUCTION

The effect whereby a narrow resonance is formed in the amplification (adsorption) of a standing wave at a Doppler-broadened transition is well known. This effect was theoretically predicted by Lamb[1] in the form of a resonant decrease in the power of a gaseous laser at the center of a Doppler line, and was experimentally observed in the investigations[2,3]. The "Lamb dip" is easily explained as follows. Two oppositely directed running light waves, waves which inevitably appear in a laser resonator, interact with two groups of atoms at a Doppler-broadened transition whose frequencies satisfy the condition $\omega_0 - \nu \pm \mathrm{kv} \lesssim \Gamma$, where Γ is the homogeneous halfwidth of the line and ω_0 and ν are the frequencies of the Doppler-line center and the light field. For $\Delta\omega_d \gg |\omega_0 - \nu| \gg \Gamma$ the two groups of atoms are completely independent, and two "Bennett holes"[4], whose depth is determined by the amplitude of each of the running waves, are produced within the Doppler contour. An exception is the case of a fine resonance $|\omega_0 - \nu) \ll \Gamma$, when both running waves interact with one and the same group of atoms, the two holes merge, and the degree of amplification saturation doubles. As a result the amplification-saturation coefficient undergoes a resonant decrease at the center of the Doppler line.

In[5] it was noted that the formation of the resonant dip does not require a standing wave, i.e., the absorption (amplification) saturation of each running wave. It is sufficient for the absorption saturation to be realized by only one running wave, while the counter wave may be a weak test wave (such a field may be called a "quasi-traveling wave"). In this case only one "hole" is burnt out in the Doppler contour, but the absorption coefficient function also has in this case a resonant dip. Moreover, in contrast to the Lamb dip, such a dip has a larger amplitude and exists at a high degree of saturation[5].

The resonant dip in the standing and quasi-traveling waves is connected with a change in the absorption of one of the waves in the presence of a necessarily intense, counter wave. We may raise the following question: does a resonant change occur in the total number of particles in the upper or lower transition levels when the frequency of the field passes through the center of the Doppler-broadened line? Strictly speaking, this effect and the Lamb dip formation effect are essentially distinct effects. Thus, the latter effect is determined by the formula for the Doppler contour in a strong field, while the former is determined by an integral over the Doppler contour. The total number of particles at the upper level is determined by the total area of the "Bennett holes" burnt out by the running counter waves (Fig. 1a), while the Lamb dip is, simply speaking, the result of the inequality of the depths of these "holes" at the center of the Doppler line in the case when the field frequency is detuned relative to the line center by an amount much larger than the homogeneous width (Fig. 1a). Therefore, it may turn out, in particular, that when the standing-wave absorption saturation is weak, the Lamb dip exists, while no dip exists in the total number of particles at the upper resonance level. If, however, the saturation is sufficiently high, then both these effects exist, but they behave essentially differently with growth of the degree of saturation. These basic differences between the two types of dips can be understood in the framework of the simplest (two-level) model for the absorbing gas (Sec. 2). Further, if the total number of particles at the upper level has a resonant dip at the center of the Doppler line, then, conversely, the total number of particles at the lower level has a peak at the center of the line (Fig. 1b), but its relative magnitude is smaller, since the lower level is usually highly populated.

2. TWO-LEVEL TRANSITION

Let us consider an ensemble of two-level particles interacting with a strong monochromatic field in the form of a plane standing light wave:

$$E(t, x) = \mathscr{E} \cos kx \cos \nu t. \tag{2.1}$$

We shall restrict ourselves to practically the most important case of a rapid transverse relaxation T_2 (the polarization relaxation) as compared to the longitudinal relaxation T_1 (population relaxation):

$$T_2 \ll T_1,$$

where $2/T_2$ is the homogeneous transition width, which

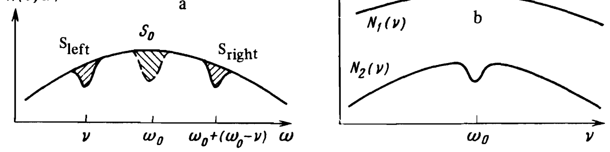

FIG. 1. Formation of narrow, particle-density resonances at the levels: a—decrease in the area of the Bennett holes at the line center when they merge; b—resonant dip in the excited-particle density $N_2(\nu)$ and the resonant peak in the density $N_1(\nu)$ of the particles at the ground level (ν is the field frequency and $N_i(\nu) = N_i(\nu, \omega)d\omega$).

is equal to the total homogeneous width of the levels and $1/T_1$ is the total rate of loss of population by the two levels. In this case we can neglect the coherent interlevel particle oscillations and the spatial inhomogeneity in the burning out of the level populations$^{1)}$. In this approximation, called the velocity-equations approximation, the variation of the population of the particles with a definite velocity component in the direction of the wave is described by the simple equations:

$$\begin{aligned}\frac{d}{dt}n_2(v)&=\frac{1}{\tau_2}[n_2{}^0(v)-n_2(v)]-w[n_2(v)-n_1(v)],\\ \frac{d}{dt}n_1(v)&=\frac{1}{\tau_1}[n_1{}^0(v)-n_1(v)]+w[n_2(v)-n_1(v)],\end{aligned}\tag{2.2}$$

where w is the probability of an induced transition of a particle with velocity v in the field of the standing wave, determined by the expression

$$w={}^1/{}_2\sigma_0P[\mathscr{L}(\Omega-kv)+\mathscr{L}(\Omega+kv)],\tag{2.3}$$

where $\sigma_0 = 4\pi|p_{12}|^2\omega_0T_2/\hbar c$ is the transition cross section at the peak for a particle that is in exact resonance with one of the waves; P [photons/cm^2-sec] is the radiation flux density; $\mathscr{L}(\xi) = (1+\xi^2)^{-1}$ is the Lorentz contour; $\xi = T_2(\Omega \pm kv)$ is the dimensionless frequency; $\Omega = \nu - \omega_0$ is the detuning of the field frequency relative to the line center ω_0; τ_i is the relaxation time for the i-th level population scaled by the equilibrium value $n_i^0 = N_i^0W(v)$, where N_i^0 is the total equilibrium population of the i-th level and W(v) is the distribution of the component of the particle velocity in the direction of the wave.

Knowing the steady-state solution of (2.2), we can find an expression for the density of the excited particles

$$N_2=\int\limits_{-\infty}^{\infty}n_2(v)\,dv.$$

We have

$$N_2-N_2{}^0=(N_1{}^0-N_2{}^0)\left\langle\frac{w\tau_2}{1+w(\tau_1+\tau_2)}\right\rangle=\frac{\tau_2}{\tau_1+\tau_2}\beta I(\beta,\Omega),\tag{2.4}$$

where $\beta = \sigma_0(\tau_1 + \tau_2)P$ is the degree of saturation of the transition and $I(\beta, \Omega)$ is a function defined by the expression

$$I(\beta,\Omega)=\frac{1}{2}\int\limits_{-\infty}^{\infty}dv\,W(v)\frac{\mathscr{L}(\Omega+kv)+\mathscr{L}(\Omega-kv)}{1+{}^1/{}_2\beta[\mathscr{L}(\Omega+kv)+\mathscr{L}(\Omega-kv)]},\tag{2.5}$$

where $W(v) = (1/\sqrt{\pi}u)\exp(-v^2/u^2)$ is a Maxwellian distribution of the velocity component and $u = (2kT/M)^{1/2}$ is the mean thermal velocity. The resonance effect in the excited-particle density is contained in the integral $I(\beta, \Omega)$. Of practical interest is the case $ku \gg \Gamma\sqrt{1+\beta}$, when the homogeneous width is substantially smaller than the Doppler width. Let us consider two limiting cases$^{2)}$.

In the case of a fine resonance ($|\nu - \omega_0| \ll \Gamma$), the integral (2.5) can be computed in the Doppler limit:

$$I(\beta,0)=I_0(1+\beta)^{-1/2},\quad I_0=\sqrt{\pi}\Gamma/ku.\tag{2.6}$$

Far from the resonance, but inside the Doppler contour ($ku \gg |\nu - \omega_0| \gg \Gamma$), the integral (2.5) can be split into a sum of two integrals, in each of which the dominant contribution is made by the particles with velocities v satisfying the resonance condition: $|\nu - \omega_0| = \pm kv$. In consequence we obtain

$$I(\beta,|\Omega|\gg\Gamma)=I_0(1+\beta/2)^{-1/2}.\tag{2.7}$$

Comparing the expressions (2.6) and (2.7), we see that the total number of particles in the upper level resonantly decreases when the frequency of the field passes through the center of the Doppler line. The resonant change in the excited-particle density can be observed from the change in the total intensity of the spontaneous emission of the particles in the upper level. This possibility was first noted by Basov and one of the present authors[8], and the corresponding method of observation of the narrow resonances was called the "nonlinear-fluorescence cell method." The observation of the narrow resonances in the spontaneous emission is an extremely effective method for gases with a low absorption coefficient, which is natural, since it is always more suitable to use fluorescence methods to detect small variations in absorption. In particular, as was noted in[8], this method is suitable for observing narrow resonances within the Doppler line of an atomic or molecular beam, when absorption is extremely small.

The nonlinear-fluorescence cell idea was put forward independently by Freed and Javan[9], and the first successful experiment was realized by them. In the experiment, they effected saturation of a low-pressure molecular (CO_2) absorption cell with an absorption coefficient of 10^{-6} cm^{-1} at room temperature. An important distinctive feature in the case of the molecular cell is the fact that the molecules in all the collisionally coupled rotational levels of an excited vibrational state contribute to the spontaneous-emission resonance. The above-considered two-level model is applicable only when the rotational relaxation is considerably slower than the vibrational at, for example, very low pressures of the gas in the cell. Thus, the next step is to analyze the effect, taking account of collisions that lead to rotational relaxation.

3. ROTATIONAL-VIBRATIONAL TRANSITIONS

Now let a light field of the form (2.1) interact with only one out of the large group of rotational-vibrational transitions interconnected by rotational relaxation. Let us ascertain what the nature of the excited-molecule density resonance in this case is and what role collisions leading to rotational relaxation then play.

To elucidate the distinctive features of the effect under conditions of rotational relaxation, we restrict ourselves to the simplest approximation, when the values of the velocity v and the total angular momentum J of the motion after a collision do not depend on the values v′ and J′ before the collision. Furthermore, we shall assume that both the particle velocity v and the quantum number J change simultaneously and independently in a collision, the equilibrium v and J distributions being established in collisions over the same period of time τ_r. In this approximation, the equations for the population of two rotational sublevels resonantly interacting with the field have the form

$$\begin{aligned}\frac{d}{dt}n_2(v)&=\frac{1}{\tau_v}[n_2{}^0-n_2]+\frac{1}{\tau_r}[n_2{}^{0r}-n_2]-w(n_2-n_1),\\ \frac{d}{dt}n_1(v)&=\frac{1}{\tau_v}[n_1{}^0-n_1]+\frac{1}{\tau_r}[n_1{}^{0r}-n_1]+w(n_2-n_1),\end{aligned}\tag{3.1}$$

where $n_i^0(v)$ and $n_i^{or}(v)$ are respectively the equilibrium and quasiequilibrium population densities of a rotational sublevel of the i-th vibrational level. The equilibrium density $n_i^0(v)$ is established in the vibrational-relaxation characteristic time τ_v for the population of the vibrational levels, while the quasi-equilibrium density $n_i^{or}(v)$ is established in the rotational-relaxation characteristic time for the population of the rotational sublevels. Naturally, the introduction in addition to vibrational relaxation of rotational relaxation, which is inevitably accompanied by a change in the rotational quantum number J, makes sense if $\tau_r \ll \tau_v$. The quasi-equilibrium population $n_i^{0r}(v)$ is connected with the total population N_i of the i-th vibrational level by the relation

$$n_i^{0r}(v) = q_i(J)\,W(v)\,N_i, \tag{3.2}$$

where $q_i(J)$ is the Boltzmann distribution of the molecules over the rotational sublevels with different J values, and the rest of the notation is the same as in (2.2). The equations for the population of the remaining rotational sublevels are exactly the same, except that the terms with w are absent, since the field does not directly react with them.

Summing $n_i(J, v)$ in Eqs. (3.1) over J and averaging over the velocities, we obtain the following system of equations for the total population of the vibrational levels:

$$\begin{aligned}\frac{dN_2}{dt} &= \frac{1}{\tau_v}(N_2^0 - N_2) - \langle w(n_2 - n_1)\rangle_v,\\ \frac{dN_1}{dt} &= \frac{1}{\tau_v}(N_1^0 - N_1) + \langle w(n_2 - n_1)\rangle_v,\end{aligned} \tag{3.3}$$

where $n_1 - n_2$ is the difference between the populations of the operating sublevels, which is equal in the steady-state case to

$$n_1 - n_2 = W(v)\left[1 + 2w\left(\frac{1}{\tau_v} + \frac{1}{\tau_r}\right)^{-1}\right]^{-1}\left[\Delta_0 - \frac{\tau_v}{\tau_v + \tau_r}(\Delta - \Delta_0)\right], \tag{3.4}$$

where $\Delta = q_1N_1 - q_2N_2$ and $\Delta_0 = q_1N_1^0 - q_2N_2^0$ are the differences between the total populations of the operating rotational sublevels. After averaging over the velocities in the steady-state case, we obtain the following system of equations for the total populations of the vibrational levels:

$$\begin{aligned}(N_2 - N_2^0)(1 + \theta q_2\Lambda) + (N_1 - N_1^0)(-\theta q_1\Lambda) &= (1 + \theta)\Lambda\Delta_0,\\ (N_2 - N_2^0)(-\theta q_2\Lambda) + (N_1 - N_1^0)(1 + \theta q_1\Lambda) &= -(1 + \theta)\Lambda\Delta_0,\end{aligned} \tag{3.5}$$

where

$$\Lambda = \frac{1}{2}\beta I(\beta, \Omega), \quad \beta = 2\sigma_0 P\left(\frac{1}{\tau_v} + \frac{1}{\tau_r}\right)^{-1}, \quad \theta = \frac{\tau_v}{\tau_r}, \tag{3.6}$$

and the function $I(\beta, \Omega)$ is determined by the previous integral (2.5).

The total steady-state populations of the levels in the field of a standing wave are equal to

$$(N_2 - N_2^0) = -(N_1 - N_1^0) = \Delta_0(1 + \theta)\frac{{}^1\!/_2\,\beta I(\beta, \Omega)}{1 + {}^1\!/_2\,\theta(q_1 + q_2)\beta I(\beta, \Omega)}. \tag{3.7}$$

The parameter β determines the degree of saturation of the operating rotational-vibrational transition $0, J_1 \to 1, J_2$ by a strong field under the conditions of rotational relaxation, i.e., it is equal to the ratio of the induced-transition probability $2\sigma_0 P$ to the sublevel-population relaxation probability $(1/\tau_v + 1/\tau_r)$. The denominator of the expression (3.7) contains another nonlinearity parameter:

$$S = {}^1\!/_2\,\theta(q_1 + q_2)\beta I(\beta, \Omega) = {}^1\!/_2(q_1 + q_2)\beta_v I(\beta, \Omega). \tag{3.8}$$

For sublevels with large J values, we can assume $q \approx q_2 = q$. The population factor of one rotational sublevel of a molecule usually lies in the range $q \approx 10^{-2}$–10^{-3} and decreases with increasing number of atoms in the molecule. For example, for the simple CO_2 molecule, which was used in the experiment in[9], we have

$$q(J) = (2J + 1)\frac{B}{kT}\exp\left[-\frac{B(J + 1)J}{kT}\right]; \tag{3.9}$$

at the P(20) transition at T = 300°K, the quantity q = 0.03. The parameter β_v is the absorption saturation parameter for the whole vibrational transition and is connected with the saturation parameter β for the operating rotational-vibrational transition (3.6) by the relation

$$\beta_v = \beta\theta = (2\sigma_0 P)\tau_v(1 + \theta)^{-1}. \tag{3.10}$$

The quantity $I(\beta, \Omega)$ is, in order of magnitude, equal to $\Gamma/\sqrt{\pi}ku$, the ratio of the homogeneous width to the Doppler width, and, in the case of narrow resonances being considered, it is much smaller than unity (10^{-2}–10^{-3}). Thus, even when the absorption of the vibrational transition is strongly saturated, the nonlinearity parameter $S \ll 1$ and can be neglected in the relation (3.7).

Thus, the resonances in the variation of the density of molecules in an excited vibrational level that arise owing to saturation of the absorption of the standing wave, and also as a result of collisions leading to rotational relaxation, can be represented in the form

$$(N_2 - N_2^0) = {}^1\!/_2(N_1^0 - N_2^0)(1 + \theta)\beta I(\beta, \Omega). \tag{3.11}$$

The formula for $N_2 - N_2^0$ written in this form allows us to easily follow how the rotational relaxation influences the parameters of the resonant dip in the excited-molecule density.

In the case of a weak rotational relaxation, when the rotational sublevels are so weakly coupled to each other that $\theta \ll 1$, the relation (3.11) assumes the form

$$(N_2 - N_2^0) = {}^1\!/_2(N_1^0 - N_2^0)q\beta I(\beta, \Omega) \tag{3.12}$$

with $\beta = 2\sigma_0 P\tau_v$. The relation (3.12) coincides with the formula (2.4) for a two-level system if account is taken of the fact that in the present case the difference between the populations of the operating levels is $(N_1^0 - N_2^0)q$ and not $N_1^0 - N_2^0$, and that the level-population relaxation times are equal, i.e., $\tau_1 = \tau_2 = \tau_v$.

When the rotational relaxation rate is increased the following changes occur. First, the number of molecules transferable by the field from a lower to a higher vibrational level increases $1 + \theta$ times. This is due to an additional "influx" of molecules with resonant velocities $kv = \nu + \omega_0$ to the lower operating sublevel from other sublevels owing to rotational relaxation. Second, by the same factor decreases the degree of absorption saturation, or increases the power necessary for saturating the transition. This is natural, since the resonance sublevels decay $1 + \theta$ times more rapidly. The parameters of the resonant dip in the excited-particle density (width and depth) vary in the following manner.

The broadening of the dip by the field decreases with increasing θ:

$$\Delta\omega = 2\Gamma(1 + \beta)^{1/2} = 2\Gamma(1 + \beta_v/\theta)^{1/2}. \tag{3.13}$$

The broadening of the dip is insignificant when the whole vibrational transition is strongly saturated ($\beta_v \approx 1$). Only when $\beta_v \gtrsim \theta \gg 1$, i.e., when the induced-transition probability becomes comparable with the rotational-relaxation probability, does the broadening of the dip by the field become appreciable.

The absolute magnitude of the resonant dip (Fig. 2) is determined by the relation

$$h = N_2(|\Omega| \gg \Gamma) - N_2(\Omega = 0) \tag{3.14}$$

and is equal to

$$h = h_0\left[1 - \left(\frac{1+\beta/2}{1+\beta}\right)^{1/2}\right], \tag{3.15}$$

where h_0 plays the role of a background against which the dip is observed. Its magnitude is

$$h_0 = \frac{q}{2}(N_1^0 - N_2^0)\sqrt{\pi}\frac{\Gamma}{ku}(1+\theta)\frac{\beta}{(1+\beta/2)^{1/2}} \tag{3.16}$$

In the case of weak saturation of the rotational-vibrational transition, when $\beta \ll 1$ (this is possible not only when $\beta_V \ll 1$, but when $\beta_V \gg 1$ as well),

$$h = h_0\beta/4, \quad h_0 \propto \beta, \tag{3.17}$$

i.e., the absolute magnitude of the dip decreases in proportion to β^2, while its relative magnitude (contrast) h/h_0 decreases in proportion to β. In the case of strong saturation, when $\beta \gg 1$, we have for the magnitude of the resonant dip

$$h = h_0(1 - 1/\sqrt{2}), \quad h_0 \propto \sqrt{\beta}. \tag{3.18}$$

In this limiting case the absolute magnitude of the dip increases in proportion to $\sqrt{\beta}$, while the relative magnitude tends to the constant value $1 - 1/\sqrt{2} \approx 0.3$. The function $N_2(\Omega)$ in these limiting cases is shown in Fig. 2.

4. DISCUSSION

Thus, there exists a narrow resonance in the density of excited particles in the field of a standing wave with a tunable frequency. The resonance appears at the center of the Doppler contour and the best conditions for its observation are realized when the transition is strongly saturated ($\beta \gg 1$). In this case one should observe in $N_2(\Omega)$ a contrasting (~30% of the background) and fairly narrow dip, whose absolute magnitude grows in proportion to $\sqrt{\beta}$, i.e., in proportion to the amplitude of the saturating field. In the case of molecular rotational-vibrational transitions, not only does rotational relaxation not inhibit the effect, but, on the contrary, it contributes to its enhancement. In this case in the formation of the narrow resonance participate not only the molecules in the resonance sublevels $(0, J_1)$ and $(1, J_2)$, but also the molecules in the remaining rotational sublevels coupled to each other by rotational relaxation. As a result, the absolute value of the effect increases $\sqrt{1+\theta}$ times. The optimum value of the saturation parameter in this case is $\beta \approx 1$, or $\beta_V \approx \theta$ (for $\theta \gg 1$); when the broadening of the dip by the field is small, the contrast is fairly large, and the absolute magnitude is roughly $\sqrt{\beta}$ times larger than in the absence of rotational relaxation.

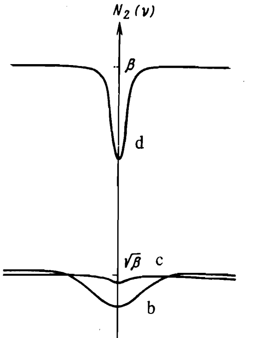

FIG. 2. Resonant dip in the excited-molecule density (in units of $\frac{1}{2}\sqrt{\pi}(N_1^0 - N_2^0)\Gamma/ku$) in diverse cases: a—weak saturation ($\beta \ll 1$ and $\beta_V \ll 1$); b—weak saturation of the vibrational-rotational transition ($\beta \ll 1$) when the saturation in the vibrational band is strong ($\beta_V \gg 1$); c—strong saturation ($\beta, \beta_V \gg 1$) when the rotational relaxation is weak ($\tau_V \ll \tau_r$); d—strong saturation ($\beta, \beta_V \gg 1$) when the rotational relaxation is strong ($\tau_V \gg \tau_r$).

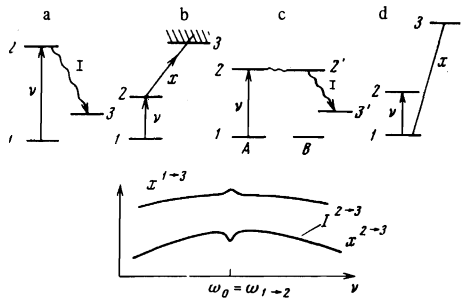

FIG. 3. Different methods of detection of the resonant variation of the total population of the upper and lower levels: a—observation of the intensity of the total spontaneous emission; b—observation of absorption into higher-lying states, including states in the continuous spectrum; c—observation of the intensity of the spontaneous emission of an admixed molecular gas: d—observation of absorption by molecules in the lower level.

The resonant variation of the total number of particles in the upper level can also be detected by other methods, besides the registration of the spontaneous emission of the excited particles (Fig. 3a), a fact which has already been pointed out in[10]. For example, we can measure the absorption coefficient at the coupled transition between the upper level of the saturated transition and higher states of the molecules, including transitions into the continuous spectrum (Figs. 3b and 3d). We can also add another gas which has a luminescence level with a short lifetime close to the excited level of a coupled transition (Fig. 3c). In this case the narrow resonance in the total number of molecules at the upper level of the absorbing gas can be observed from the change in the intensity of luminescence of the admixed molecules. This method is useful for absorber molecules which have a low luminescence efficiency.

The excited-particle density resonant variation effect can be used to increase the selectivity of chemical processes occurring in the gaseous phase with the participation of particles excited by laser radiation. With the aid of this effect we can eliminate the limitation imposed on the selectivity by Doppler broadening[11].

The nonlinear, narrow, fluorescence resonances that develop owing to the excited-particle density resonance are an extremely effective spectroscopic method within the Doppler line of weakly absorbing molecular transitions. To such transitions pertain rotational-vibrational molecular transitions between excited levels, transitions between the vibrational harmonics, vibrational quadrupole transitions of homonuclear molecules, which are forbidden in the dipole approximation, and others. In this consists the significant advantage of the nonlinear-fluorescence cell method[8,9] over the nonlinear-absorption cell method[12-14].

[1)] It is shown in [6] that the contribution of these effects, which appear in a strong saturation, is small and only quantitative in nature. In particular, there occurs a small (20–30%) change in the depth of the Lamb dip.

[2)] By integrating (2.6) by parts, we can express it in terms of the plasma function [7].

[1] W. E. Lamb, Jr., Phys. Rev. A134, 1429 (1964).

[2] R. A. Mcfarlane, W. R. Bennett, Jr., and W. E. Lamb,

Jr., Appl. Phys. Lett. **2**, 189 (1963).

[3] A. Szoke and A. Javan, Phys. Rev. Lett. **10**, 521 (1963).

[4] W. R. Bennett, Jr., Phys. Rev. **126**, 580 (1962).

[5] V. S. Letokhov and V. P. Chebotaev, ZhETF Pis. Red. **9**, 364 (1969) [JETP Lett. **9**, 215 (1969)].

[6] B. J. Feldman and M. S. Feld, Phys. Rev., **A1**, 1375 (1970).

[7] S. Stenholm and W. E. Lamb, Jr., Phys. Rev. **181**, 618 (1969).

[8] N. G. Basov and V. S. Letokhov, Proceedings URSI Conference on Laser Measurements, Warsaw, Poland (1968); Electron Technology **2**, 2/3, 15 (1969).

[9] C. Freed and A. Javan, Appl. Phys. Lett., **17**, 53 (1970).

[10] V. S. Letokhov, Comments Atomic and Molecular Physics **2**, 181 (1971).

[11] R. V. Ambartzumian and V. S. Letokhov, Appl. Optics **11**, 354 (1972).

[12] V. S. Letokhov, ZhETF Pis. Red. **6**, 597 (1967) [JETP Lett. **6**, 101 (1967)]; Zh. Eksp. Teor. Fiz. **54**, 1244 (1968) [Sov. Phys.-JETP **27**, 665 (1968)].

[13] V. N. Lisitsyn and V. P. Chebotaev, Zh. Eksp. Teor. Fiz. **54**, 419 (1968) [Sov. Phys.-JETP **27**, 227 (1968)].

[14] P. H. Lee and M. L. Skolnick, Appl. Phys. Lett. **10**, 303 (1967).

Translated by A. K. Agyei
89