SPATIAL EFFECTS IN SATURATION OF RESONANCE ABSORPTION OF A GAS IN A LIGHT FIELD

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The dependence of absorption saturation in a gas at low pressure on the configuration of the light field is considered. In particular, absorption saturation of a gas in the field of two standing waves is considered as a function of the angle between the waves; absorption saturation is also analyzed for the field of a traveling spherical wave as a function of the curvature. For nonaxial modes the degree of absorption saturation is much lower than for axial modes. It is suggested that this effect may be employed for self-selection of the lowest transverse mode of a laser by means of a nonlinear absorbing gas cell in the cavity. It is shown that the degree of absorption saturation for a spherical wave decreases with increase of curvature of the wave front.

1. INTRODUCTION

IT is known that resonance absorption of gas in a light field is decreased by saturation[1]. If the homogeneous width of the resonance transition is much smaller than the Doppler width, then the wave $E\cos(\nu t - \mathbf{k}\cdot\mathbf{r})$ interacts effectively with atoms for which the projection of the velocity v on the direction of the wave vector k is equal to $v \approx (\nu - \omega)/k$, accurate to $\Delta v = \gamma/k$, where ω is the frequency of the center of the Doppler line and γ is the homogeneous line width. In an intense field, therefore, "hole burning" occurs in the Doppler line at a frequency ν, with a width γ.[2] If the light field is a standing wave (for example the fundamental mode in a laser resonator), then the interaction of the atoms with each of the oppositely traveling waves gives rise to two "holes" that are symmetrical with respect to ω[2].

Saturation of the absorption of the gas is quite sensitive to the deviation of the frequency of the standing wave relative to the center of the Doppler line. If the field frequency ν coincides with the center of the line ($|\nu - \omega| \ll \gamma$), then the degree of saturation is larger than when $|\nu - \omega| \gg \gamma$. Physically this is due to the fact that in the case of small frequency deviation the atoms interacting with the field feel both traveling waves and the degree of saturation doubles. An increase of the degree of saturation at the center of the line can be interpreted also as the consequence of a superposition of two holes at $\nu = \omega$[3]. In practice this effect is manifest by the fact that when the frequency of the standing wave is scanned along the Doppler line the absorption coefficient (gain) has a dip at the center of the line (Lamb dip)[3]. In the case of a small degree of saturation ($\beta E^2 \ll 1$), the standing-wave absorption coefficient is given by[3]

$$\alpha(\nu) = \alpha_0(\nu)\left[1 - \beta E^2\left(1 + \frac{\gamma^2}{\gamma^2 + (\omega - \nu)^2}\right)\right] \qquad (1)$$

where $\alpha_0(\nu)$ is the linear absorption coefficient. Experimentally, the dip at the center of the amplification line was observed in an He-Ne laser in[4,5].

The purpose of the present article is to show that absorption saturation of a gas of sufficiently low pressure is quite sensitive to the configuration of the light field. Physically this is due to the fact that if the atoms interacting with the field traverse without relaxation or collision a distance comparable with the transverse dimension of the beam, then the medium feels the field in a certain microscopic region. As a result, the degree of absorption saturation turns out to be critically sensitive to the form of the light field. As possible field configurations, we consider the modes of an open resonator with flat mirrors and a spherical light wave. It is shown in sections 2 and 3 that the degree of absorption saturation of non-axial modes is much lower than that of axial modes. In the case of a sufficiently small ratio γ/ku (ku—Doppler line width) this phenomenon can be used for a very effective self-selection of the lower transverse mode with the aid of a nonlinear absorbing gas cell in the laser[6]. It is shown in section 4 that the degree of absorption saturation of a spherical wave greatly depends on its radius, which is also of interest for the control of the light-field configuration.

2. QUALITATIVE ANALYSIS FOR A TRANSVERSE MODE

A plane standing wave of frequency ν interacts effectively with atoms whose velocities lie in the regions shown shaded in Fig. 1a. It is assumed in Fig. 1 that the x axis coincides with the axis of the wave, ω is the frequency of the center of the Doppler line, and $u = \sqrt{2kT/m}$ characterizes the average thermal velocity of the atoms, γ is the homogeneous line width, and $k = \omega/c$. When $|\nu - \omega| \gg \gamma$ the field interacts with two non-overlapping groups of atoms in velocity space. When the frequency of the field approaches the center of line, the shaded regions begin to overlap, i.e., the atoms begin to interact with both traveling waves and the degree of saturation increases. This corresponds to an overlap of the holes in the Doppler contour, but we have used a two-dimensional picture, since it is more convenient on going over to more complicated field configurations.

Let the light field be a superposition of two plane

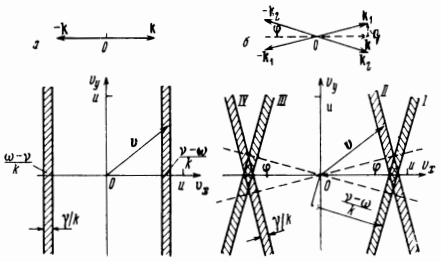

FIG. 1. Space of the projections of the atom velocities v on the xy plane. The shaded regions contain the velocities of the atoms that interact resonantly with a plane standing wave with wave vector k and frequency ν (a) and two plane standing waves with vectors $\mathbf{k}_1$ and $\mathbf{k}_2$ and with frequency ν (b).

standing waves of equal frequency, the wave vectors of which, $\mathbf{k}_1$ and $\mathbf{k}_2$, lie in the plane (x, y) and make angles $\pm\varphi$ with the x axis (Fig. 1b). Such a field configuration arises, for example, upon excitation of a mode that axis is non-axial in the y direction in a plane-parallel resonator, the axis of which coincides with the x axis, i.e., TEM_{0n} modes[7]. The transverse component of the wave vector is determined by the dimension of the mirror 2a, and by the transverse index of the mode n:

$$q \approx \pi(n+1)/a \tag{2}$$

Usually $a \gg \lambda$, and therefore the angle between the wave vectors, which equals

$$2\varphi = 2q/k \approx (n+1)\lambda/a, \tag{3}$$

is quite small[1]. Such a light field interacts effectively with atoms whose velocity projection on the (x, y) plane lies in the regions shaded in Fig. 1b. When $|\omega - \nu| \gg \gamma$ and $\varphi \gg \gamma/ku$, there exists in velocity space four little-overlapping groups of atoms interacting with the field. When the angle between the standing waves decreases, the degree of overlap of regions I and II (respectively III and IV) increases and when

$$\varphi \leqslant \gamma/ku \tag{4}$$

practically all the atoms in regions I and II (III and IV) overlap. This corresponds physically to the fact that the atoms feel the field of the traveling waves in both standing waves. As a result, the degree of absorption saturation is doubled. If $\omega = \nu$, then the degree of saturation increases by another factor of two as a result of coalescence of the regions I–II and III–IV. In this case the atoms feel the field of all four traveling waves.

Condition (4) with allowance for (3) can be represented in the form

$$u/\gamma > a/\pi(n+1). \tag{5}$$

The left side of the inequality (5) is the average distance of the coherent interaction between the atom and the field, and the right side is the characteristic dimension of the transverse inhomogeneity of the field. Thus, the degree of saturation is sensitive to the field configuration in the case when the atom is capable of feeling the inhomogeneity in the transverse field distribution during the course of its coherent interaction with the field.

3. ABSORPTION SATURATION IN THE FIELD OF TWO STANDING WAVES

Let the light field be a superposition of two standing waves in the form

$$E(\mathbf{r}, t) = E(\sin \mathbf{k}_1\mathbf{r} \pm \sin \mathbf{k}_2\mathbf{r}) \cos \nu t, \tag{6}$$

where $\mathbf{k}_1 = k\mathbf{e}_x + q\mathbf{e}_y$, and $\mathbf{k}_2 = k\mathbf{e}_x - q\mathbf{e}_y$; $\mathbf{e}_x$ and $\mathbf{e}_y$ are unit vectors along the axes x and y; the plus sign corresponds to modes that are even in y, and the minus sign corresponds to odd modes.

The difference of the probabilities of the populations of the two levels 1 and 2 of atoms having a velocity v is equal to[3]

$$\rho_{11}(\mathbf{v}) - \rho_{22}(\mathbf{v}) = [\rho_{11}^0(\mathbf{v}) - \rho_{22}^0(\mathbf{v})]/[1 + R(\mathbf{v}, \mathbf{k}_1, \mathbf{k}_2)]. \tag{7}$$

Here $\rho_{11}^0(\mathbf{v})$ and $\rho_{22}^0(\mathbf{v})$ are the probabilities of the populations of levels 1 and 2 in the absence of a field, and the saturation parameter $R(\mathbf{v}, \mathbf{k}_1, \mathbf{k}_2)$ is determined by the expression

$$R = \frac{p^2}{4\hbar\gamma_1\gamma_2} E^2 [\mathscr{L}(\varepsilon + \mathbf{k}_1\mathbf{v}) + \mathscr{L}(\varepsilon - \mathbf{k}_1\mathbf{v}) + \mathscr{L}(\varepsilon + \mathbf{k}_2\mathbf{v}) + \mathscr{L}(\varepsilon - \mathbf{k}_2\mathbf{v})], \tag{8}$$

where p is the matrix element of the dipole moment of the transition, $\mathscr{L}(\omega) = \gamma^2/(\omega^2 + \gamma^2)$ describes the Lorentz form of homogeneous broadening, γ_1 and γ_2 are the damping constants of the levels, $\gamma = (\gamma_1 + \gamma_2)/2$, $\epsilon = \nu - \omega$.

The absorption coefficient of a weak trial traveling wave with frequency ω_0 and wave vector k is given by

$$\alpha(\omega_0, \mathbf{k}) = \int d\mathbf{v}\, N(\mathbf{v})[\rho_{11}(\mathbf{v}) - \rho_{22}(\mathbf{v})]\sigma(\omega_0 - \omega + \mathbf{k}\mathbf{v}), \tag{9}$$

where N(v) is the velocity distribution of the atoms, $\sigma(\omega' - \omega) = \sigma_0\mathscr{L}(\omega' - \omega)$—cross section of the radiative transition at the frequency ω', σ_0—cross section of radiative transition at exact resonance ($\omega' = \omega$). The absorption coefficient of the two standing waves causing the absorption saturation of the gas, obviously, is given by

$$\alpha(\nu) = \int d\mathbf{v}\, N(\mathbf{v})[\rho_{11}(\mathbf{v}) - \rho_{22}(\mathbf{v})] \cdot \times [\sigma(\varepsilon + \mathbf{k}_1\mathbf{v}) + \sigma(\varepsilon - \mathbf{k}_1\mathbf{v}) + \sigma(\varepsilon + \mathbf{k}_2\mathbf{v}) + \sigma(\varepsilon - \mathbf{k}_2\mathbf{v})]. \tag{10}$$

We shall calculate (10) for a low degree of saturation ($R \ll 1$). This assumption has in fact already been made in the description of the saturation with the aid of (7), in which no account is taken of the line broadening in the field. In addition, the initial distribution over the velocities and over the levels will be assumed isotropic:

$$N(\mathbf{v}) = \frac{N_0}{(\sqrt{\pi}u)^3} \exp\left(-\frac{v^2}{u^2}\right), \qquad \rho_{11}^0(\mathbf{v}) - \rho_{22}^0(\mathbf{v}) = \text{const},$$

which certainly is valid for the case of an equilibrium gas.

After rather cumbersome calculations, expression (10) can be reduced to the form

$$\alpha(\nu) = \alpha_0(\nu)[1 - \beta F(\varepsilon/\gamma, qu/\gamma)], \tag{11}$$

where $\alpha_0(\nu)$ is the absorption coefficient for a weak field, $\beta = p^2E^2/8\hbar^2\gamma_1\gamma_2$ is the saturation parameter, and the function F describes the dependence of the degree of

[1] Since each wave has an angular divergence $\delta\varphi \approx \lambda/a$, a distinct separation of the propogation directions takes place for modes with sufficiently large transverse index n.

saturation on the detuning ϵ and on the transverse component of the wave vectors q. The function F can be calculated assuming that the homogeneous width is small compared with the Doppler width:

$$\gamma \ll ku \tag{12}$$

and that the angle between the standing waves is small:

$$q \ll k. \tag{13}$$

The condition (12) is necessary for the occurrence of the effect under consideration, and the condition (13) limits only the region of large angles, which are of no interest for the problem under consideration. The final expression for $F(\epsilon/\gamma, qu/\gamma)$ is given by

$$F\left(\frac{\varepsilon}{\gamma}, \frac{qu}{\gamma}\right) = \left[1 + \frac{1}{1+(\varepsilon/\gamma)^2}\right] + \sqrt{\pi}\frac{\gamma}{qu}\left[1 - \Phi\left(\frac{\gamma}{qu}\right)\right] e^{(\gamma/qu)^2} + \frac{1}{\sqrt{\pi}}\int_{-\infty}^{\infty} \frac{e^{-\xi^2}\,d\xi}{1+(qu\xi/\gamma + \varepsilon/\gamma)^2} \tag{14}$$

where

$$\Phi(x) = \frac{2}{\sqrt{\pi}}\int_0^x e^{-\xi^2}\,d\xi$$

is the error integral.

The first term in (14) describes the frequency dependence of the degree of saturation, the second term describes the angular dependence, and the third the frequency-angular dependence. In the particular case of standing waves with identical directions (q = 0), the second term is equal to unity, and the third coincides with the first without unity. As a result, expression (14) goes over into the expression obtained by Lamb[3]:

$$F\left(\frac{\varepsilon}{\gamma}, 0\right) = 2\left[1 + \frac{1}{1+(\varepsilon/\gamma)^2}\right]. \tag{15}$$

In the case of exact resonance (ϵ = 0), the third term coincides with the second and expression (14) reduces to

$$F(0, qu/\gamma) = 2\left\{1 + \sqrt{\pi}\frac{\gamma}{qu}\left[1 - \Phi\left(\frac{\gamma}{qu}\right)\right] e^{(\gamma/qu)^2}\right\}. \tag{16}$$

Far from the center of the line ($|\epsilon| \gg \gamma$) the third term is negligibly small, and the function F is given by

$$F(\infty, qu/\gamma) = 1 + \sqrt{\pi}\frac{\gamma}{qu}\left[1 - \Phi\left(\frac{\gamma}{qu}\right)\right] e^{(\gamma/qu)^2}, \tag{17}$$

i.e., $F(0, qu/\gamma) = 2F(\infty, qu/\gamma)$.

The dependence of the degree of saturation on the transverse component of the wave vector for two limiting values of the field frequency detuning (ϵ = 0 and $|\epsilon| \gg \gamma$) is shown in Fig. 2. The decrease of the degree of saturation upon detuning relative to the center of the line is described approximately by the Lamb factor (15). One can assume approximately

$$F\left(\frac{\varepsilon}{\gamma}\,\frac{qu}{\gamma}\right) \approx \left[1 + \frac{1}{1+(\varepsilon/\gamma)^2}\right]\left\{1 + \sqrt{\pi}\frac{\gamma}{qu}\left[1 - \Phi\left(\frac{\gamma}{qu}\right)\right] e^{(\gamma/qu)^2}\right\} \tag{18}$$

It follows from (11) and (18) that the degree of absorption saturation in a field of two standing waves is sensitive not only to the detuning of the field frequency relative to the center of the line, but also to the angle between the waves. It is important that the angular dependence exists at any value of ϵ, i.e., at any point of the Doppler line.

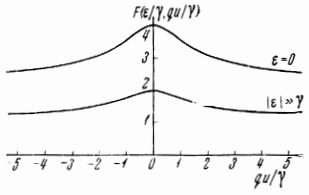

FIG. 2. Dependence of the degree of absorption saturation on the transverse components q of the wave vectors of two standing waves (inside the Lamb dip (ϵ = 0) and outside this dip ($|\epsilon| \gg \gamma$))

4. QUALITATIVE ANALYSIS FOR A SPHERICAL WAVE

Let us consider now the case of absorption saturation of a low-pressure gas in the field of a spherical wave, $\cos(\nu t \pm kr)$ with a center at the point **r** = 0. We consider a wave region sufficiently far from its center (**r** ≫ a, a—dimension of the region). At the point **r**, the wave interacts resonantly with atoms whose velocities **v** satisfy the condition

$$|\mathbf{k}(\mathbf{r})\mathbf{v} \pm (\omega - \nu)| \lesssim \gamma, \tag{19}$$

where $\mathbf{k}(\mathbf{r}) = k\mathbf{r}/r$ is the wave vector of the field at the point **r**, and the plus and minus signs correspond to converging and diverging waves.

Let the effective mean free path of the atom relative to the change of the state of the atom due to collisions, radiative damping, etc. be l_p (obviously, $l_p \approx u/\gamma$). In order for the atom to interact resonantly with the spherical wave during the entire free path time $\tau_p = l_p/u = \gamma^{-1}$, it is necessary to satisfy the condition (19) for all **r** from the point of the start of the path $\mathbf{r}_0$ to the final point $\mathbf{r}_0 + \mathbf{v}\tau_p$. The angle between the wave vectors at the beginning and at the end of the path is $\varphi \approx l_p/\langle r\rangle$, where $\langle r\rangle$ is the average radius of curvature of the wave front, and it is assumed for simplicity that $\nu = \omega$. Then the total change of $\mathbf{k}(\mathbf{r})\cdot\mathbf{v}$ during the free path time of the atom amounts to $\frac{1}{2}\varphi ku$, and condition (19) can be represented in the form

$$\langle r\rangle \gtrsim \frac{1}{2}\frac{ku}{\gamma} l_p = \frac{k}{2} l_p^2 \tag{20}$$

If condition (20) is not satisfied, then the atom interacts effectively with the field at a distance $l_{eff} \approx \sqrt{2\langle r\rangle/k}$, with $l_{eff} < l_p$. This means that the width of the resonance can be represented in the form

$$\gamma_{eff} \approx \gamma + u/l_{eff} = \gamma + ku(2k\langle r\rangle)^{-1/2}. \tag{21}$$

The broadening of the resonance of the interaction leads to a corresponding increase of the absorption saturation power as the result of the decrease of the saturation parameter. Consequently, the degree of absorption saturation depends critically on the radius of the spherical wave. The critical radius, at which the degree of saturation begins to decrease, is determined by the expression

$$r_{cr} \approx \frac{1}{2k}\left(\frac{ku}{\gamma}\right)^2. \tag{22}$$

If the width of the interaction resonance without allowance for the curvature of the wave front is determined only by the finite time of flight of the atoms through the beam, i.e., $\gamma \approx u/a$, where a is the diameter of the beam, then the critical radius is equal to

$$r_{cr} \approx \tfrac{1}{2}ka^2. \tag{23}$$

Such a wave-front radius corresponds to the diffraction divergence of the light beam. Consequently, in a low-pressure gas the degree of absorption saturation is extremely sensitive to the curvature of the wave front.

5. POSSIBLE APPLICATIONS

The dependence of the degree of absorption saturation on the angle between two standing waves can be used for self-selection of the lower transverse mode of the laser, by placing a nonlinearly absorbing low-pressure gas cell inside the resonator[2)]. In accordance with the foregoing, effective self-selection of the lower transverse mode should occur when the molecule mean free path in the cell, without a change of state as the result of collisions and radiative relaxation, exceeds the transverse dimension of the beam. Of course, a detailed analysis of the process of self-selection of transverse modes in such a laser calls for an additional analysis. By way of examples of the possible pairs of lasers and gas absorbers we can present: 1) an argon-ion laser at wavelength $\lambda = 5145$ Å and the molecule I_2 (radiative-relaxation time $\sim 10^{-5}$ sec[11]); 2) laser with CO_2-N_2-He mixture and SF_6 molecule having strong absorption at the wavelength $\lambda = 10.5915\ \mu$[12], etc. In all these cases we can satisfy the condition of self selection of the lower transverse mode.

The effect of the dependence of the degree of absorption saturation on the curvature of the wave front of the light wave can be used for self-correction of the wave front of the beam inside the laser, by placing inside the resonator a low-pressure gas cell, or else outside the laser, by passing through the cell the emerging ray. The same lasers and absorbing molecules are suitable in this case, too.

Thus, the considered spatial effects which occur upon absorption saturation of a low-pressure gas in a light field can be used for effective control of the configuration of the light field of the laser.

In conclusion, the author is deeply grateful to academician N. G. Basov for a discussion and support of the present work.

2) Gas lasers with absorbing low-pressure cells were proposed in [8,9]. An He-Ne laser with a neon absorbing cell was considered in [10].

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