ABSORPTION OF AN INTENSE MONOCHROMATIC WAVE AND DISPERSION IN RESONANT MEDIA WITH INHOMOGENEOUSLY BROADENED LINES

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The passage of an intense monochromatic wave through a resonant medium with an inhomogeneously broadened line is studied. Equations are derived for the variation of the wave phase and its absorption in such a medium, and solutions of the equations are found. It is shown that absorption ($\Delta_0 > 0$) always occurs in the case of exact resonance; the absorption cross sections strongly grows with increasing intensity. In the case of deviations from resonance and sufficiently large intensities, the wave may either be absorbed or amplified, depending on the input value of the intensity. A wave is then asymptotically formed, and its amplitude has definite discrete values.

THE problems involved in the passage of intense radiation through a system with a homogeneously-broadened line have been considered in sufficient detail[1]. Similar problems for systems with an inhomogeneously broadened line have recently become the subject of studies. Among the publications on this question, notice should be taken of[2-4], where the formation of stationary π pulses following the passage of an intense ultrashort wave through a resonant medium is considered. Since only asymptotic solutions for the field have been obtained in[2-4] (stationary pulses), the question of the influence of the initial conditions on the process of formation and on the form of the stationary pulses remains unclear. This question arises in connection with the fact that the form of the asymptotic pulse, obtained in[2-4], is not unique (see[5-6]).

In this paper we investigate the influence of the intensity of a monochromatic wave on its absorption and on the dispersion characteristics of a resonant medium. It turns out that in this case it is possible to obtain an exact analytic solution of the problem. It is shown that although the incident radiation is monochromatic, self-transparency of the medium can occur at certain values of the input intensity and deviation from resonance, just as in[2-4].

We shall take into account only the spread of the energy levels, and neglect the radiative damping. Although the results are applicable to systems with an arbitrary energy spread, principal attention will be paid to a Gaussian distribution, to be able to use the results later for gases with a Doppler line contour.

We shall start with the quasiclassical equations for the radiation field and a system of two-level atoms[1]

$$\frac{\partial a}{\partial x}+\frac{1}{c}\frac{\partial a}{\partial t}=\frac{2\pi i}{\omega}M\int w(\varepsilon)\rho d\varepsilon, \quad (1)$$

$$\frac{\partial \rho}{\partial t}+i\varepsilon\rho=-\frac{i}{c\hbar}M^*\Delta a, \quad (2)$$

$$\frac{\partial \Delta}{\partial t}=\frac{2i}{c\hbar}(M^*a\rho^*-Ma^*\rho). \quad (3)$$

Here a is the amplitude of the vector potential, ρ the current of the transition, Δ the density of the excess population of the levels, M the matrix element of the transition, ϵ the deviation from resonance, and $w(\epsilon)$ the distribution function of the atomic frequencies.

In Eq. (2) we have neglected the homogeneous broadening of the line, thereby imposing definite limitations on the pulse duration. Our analysis is valid for pulses with duration $T \ll T_{hom}$, where T_{hom} is the time of irreversible relaxation. Such a situation takes place, for example, when laser pulses pass through rarefied gases. The monochromatic-wave approximation, which is employed here, is a certain idealization, since the spectral width of the incident radiation is $\Delta\omega \gg T_{hom}^{-1}$. From this point of view, the relation between the parameters $\Delta\omega$ and ω is more important. It will be shown below that when $\Delta\omega \ll \epsilon$ it is possible to neglect the nonmonochromaticity of the incident radiation.

It is impossible to change over directly in formulas (1)–(3) to the case of a monochromatic wave, since the interaction between the radiation and the atoms must be turned on adiabatically. To this end, we note that it follows from (2) and (3) that

$$\frac{\partial \Delta^2}{\partial t}+4\frac{\partial|\rho|^2}{\partial t}=0. \quad (4)$$

Let the interaction be turned on at the instant $t \to +\infty$. It is then necessary to assume that $\rho \to 0$ and $\Delta \to \Delta_0$ when $t \to -\infty$, where Δ_0 is the initial value of the excess-population density. It follows therefore that

$$\Delta=(\Delta_0^2-4|\rho|^2)^{1/2}. \quad (5)$$

Substituting (5) in (2), we get

$$\frac{\partial \rho}{\partial t}+i\varepsilon\rho=-\frac{i}{c\hbar}M^*a(\Delta_0^2-4|\rho|^2)^{1/2}. \quad (6)$$

The density of the transition current ρ, determined from this equation, is characterized by rapid time variations with frequency ϵ as well as by small variations determined by the potential a. The modulation of the current with the frequency ϵ, due to the presence of the time derivative in (6), leads to emission of multiphoton combination lines, which are shifted relative to the incident line by an amount $k\epsilon$ ($k = \pm 1, \pm 2, \ldots$). Since we do not take into account the spontaneous transitions, stimulated emission of these lines is possible only if the incident radiation has nonzero spectral components at the indicated frequencies. If $\Delta\omega \ll \epsilon$, then the probability of the appearance of combination frequencies is small, and in the limit of monochromatic radiation it is equal to zero. Therefore the quantity $\partial\rho/\partial t$ in (6) can be neglected when $\Delta\omega \ll \omega$. A rigorous mathematical proof of the possibility of neglecting $\partial\rho/\partial t$ is based on the transition to the spectral components a and ρ in Eqs. (1) and (6). We shall demonstrate this, for example, for the linear case without allowance for the scatter of the levels. In this case

$$a(t,x)=\int_{-\infty}^{+\infty} a_0(\omega')\exp\left[-i(\omega'-\omega)\left(t-\frac{x}{c}\right)+\frac{ipx}{\varepsilon-(\omega'-\omega)}\right]d\omega' \quad (7)$$

$$p=-\frac{2\pi|M|^2}{\omega c\hbar}\Delta_0, \quad (8)$$

where ω is the carrier frequency of the incident radiation, and $a_0(\omega')$ is the spectrum incident on the boundary $x = 0$. If $a_0(\omega') = A\delta(\omega' - \omega)$, then $a(t, x) = Ae^{ipx/\epsilon}$, which coincides exactly with the solution of Eqs. (1) and (6) neglecting $\partial\rho/\partial t$. On the other hand, if the spectral width is small but nonvanishing, $\Delta\omega \ll \epsilon$, then, putting $|\omega' - \omega| \ll \epsilon$ in (7) and confining ourselves to the first two terms of the expansion, we get

$$a(t,x)=a_0\left(t-\frac{x}{c}-\frac{px}{\varepsilon^2}\right)e^{ipx/\varepsilon}.$$

This result follows also from formulas (1) and (6), if we regard $\partial\rho/\partial t$ in (6) as a small perturbation. The nonmonochromaticity, as seen from the last formula, leads to a group delay of the pulses, an important factor when $x \sim x_0 = T\epsilon^2 p^{-1}$. The next terms of the expansion in (7) are significant when $x \sim x_0\epsilon/\Delta\omega \gg x_0$. For rarefied gases with allowed transitions, and for $\epsilon \sim 10^{11}$–10^{12} sec^{-1}, the characteristic length x_0 is of the order of several meters. On the other hand, effects of bleaching of the medium, which are considered below, occur much earlier, and therefore, with high accuracy, the nonmonochromaticity of the wave can be neglected. An analysis of the nonlinear case changes these results insignificantly. It can be shown that in this case the spectrum of a monochromatic wave does not become any richer. In the case of a nonmonochromatic wave, a group delay also appears, but it is already dependent on the intensity. This leads to a distortion of the shape of the pulse (since different sections of the pulse move with different velocities), but these effects also takes place when $x > x_0$ and are significant.

We proceed to the case of a monochromatic wave and discard the time derivatives in formulas (1) and (6). We then get from (6)

$$\rho=-\frac{M^*\Delta_0}{c\hbar}\frac{a}{\sqrt{\varepsilon^2+q^2|a|^2}} \quad (9)$$

where $q = 2|M|/c\hbar$. In (6) it is necessary to require that the sign of the root at $\epsilon \gg q/|a|$ coincide with the sign of ϵ. Substituting now (9) in (1), we obtain

$$\frac{\partial a}{\partial x}=ipa\int\frac{w(\varepsilon)\,d\varepsilon}{\sqrt{\varepsilon^2+q^2|a|^2}}. \quad (10)$$

When no account is taken of the scatter of the levels, Eq. (10) leads only to a change in the phase of the wave, which is connected with the deviation of the refractive index of the medium n from unity:

$$n=1+\frac{cp}{\omega\sqrt{\varepsilon^2+q^2|a|^2}}. \quad (11)$$

When $q|a| \ll \epsilon$, we obtain the well known dispersion law near resonance. We shall not discuss here the nonlinear effects that result from the given dependence of the refractive index on the intensity. This was done in a separate publication.

Although no account was taken of radiative damping in (10), to obtain real absorption it is necessary to choose in a suitable manner the integration contour. In the linear case it is sufficient to assume that ϵ has an infinitesimally small negative imaginary component. For correct allowance of the absorption in the nonlinear case, we shall expand the root in (10) in powers of $q|a|/\epsilon$. Then

$$\frac{\partial a}{\partial x}=ipa\sum_{n=0}^{\infty}\frac{(-1)^n\Gamma(n+1/2)}{n!\,\Gamma(1/2)}(q^2|a|^2)^n\int_{-\infty}^{+\infty}\frac{w(\varepsilon)\,d\varepsilon}{(\varepsilon-i0)^{2n+1}}. \quad (12)$$

We use further the well known formula

$$\frac{1}{(\varepsilon-i0)^{n+1}}=\frac{P}{\varepsilon^{n+1}}+i\pi\frac{(-1)^n}{n!}\delta^{(n)}(\varepsilon).$$

After taking into account the singularity at $\epsilon = 0$, we can sum (12) again and obtain the following results:

$$\frac{\partial a}{\partial x}=ipa\int_0^{\infty}\frac{w(\varepsilon)-w(-\varepsilon)}{\sqrt{\varepsilon^2+q^2|a|^2}}d\varepsilon + \frac{\sigma\Delta_0 a}{2\pi w(0)}\int_0^1\frac{w(iq|a|\varepsilon)+w(-iq|a|\varepsilon)}{\sqrt{1-\varepsilon^2}}d\varepsilon. \quad (13)$$

Here $\sigma = 4\pi^2|M|^2 w(0)/\omega c\hbar$ is the resonant absorption cross section.

Let us examine in greater detail the following case:

$$w(\varepsilon)=\frac{e^{-(\varepsilon-\bar{\varepsilon})^2/\gamma^2}}{\gamma\sqrt{\pi}}, \quad (14)$$

where $\bar{\epsilon} = \omega - \omega_0$ (ω—frequency of the incident radiation, ω_0—center of the atomic line). In particular, for a Doppler contour $\gamma = (\omega/c)(2kT/m)^{1/2}$. Substituting (14) in (13), we get

$$\frac{\partial a}{\partial x}=\frac{2ip}{\gamma\sqrt{\pi}}e^{-\bar{\varepsilon}^2/\gamma^2}a\int_0^{\infty}\frac{e^{-\eta^2z^2}\,\mathrm{sh}(2\bar{\varepsilon}\eta z/\gamma)}{\sqrt{1+z^2}}dz + \frac{\sigma\Delta_0}{\pi}a\int_0^1\frac{e^{\eta^2z^2}\cos(2\bar{\varepsilon}\eta z/\gamma)}{\sqrt{1-z^2}}dz, \quad (15)$$

where $\eta = q|a|/\gamma$. The first term in (15) gives the change of the phase of the wave, and the second gives the absorption (if $\Delta_0 < 0$). If we neglect the absorption, then formula (15) enables us to calculate the refractive index of the medium in the nonlinear case

$$n(\bar{\varepsilon})=1+\frac{2cp}{\omega\gamma\sqrt{\pi}}e^{-\bar{\varepsilon}^2/\gamma^2}\int_0^{\infty}\frac{e^{-\eta^2z^2}\,\mathrm{sh}(2\bar{\varepsilon}\eta z/\gamma)}{\sqrt{1+z^2}}dz. \quad (16)$$

It is seen from (16) that the refractive index $n \to 1$ when $\bar{\epsilon} \to 0$ and (16) goes over into (11) when $\bar{\epsilon} \gg \gamma$. Thus, the dependence of the refractive index on the frequency has the same form as in the linear case, but the position and the absolute value of the extrema change. In particular, when $\eta \gg 1$ and $\bar{\epsilon} \lesssim \gamma$ we have

$$n(\bar{\varepsilon})=1+\frac{cp}{\omega\gamma\eta}\Phi\left(\frac{\bar{\varepsilon}}{\gamma}\right), \quad (17)$$

where Φ is the probability integral. It is seen from (17) that when $\eta \to \infty$ the refractive index $n \to 1$, i.e., the medium behaves like vacuum at high intensities.

We now change over in (15) to the intensity

$$\frac{\partial\eta}{\partial x} = \frac{\sigma\Delta_0}{2}\eta f(\eta), \quad (18)$$

where

$$f(\eta) = \frac{2}{\pi}\int_0 \frac{e^{\eta^2 z^2}\cos(2\bar{\varepsilon}\eta z/\gamma)}{\sqrt{1-z^2}}dz. \quad (19)$$

Since we neglect the radiative damping, our analysis is valid for $\bar{\epsilon} \lesssim \gamma$ (the center of the absorption line). The factor f in (18), which depends on the intensity, takes into account the nonlinear character of the absorption. When $\eta \ll 1$ we have $f \approx 1$ and we get the usual linear absorption law. In the other limiting case, $\eta \gg 1$, we can obtain from (19) the following asymptotic form

$$f(\eta) = (\eta\sqrt{\pi})^{-1}e^{\eta^2}\cos(2\bar{\varepsilon}\eta/\gamma). \quad (20)$$

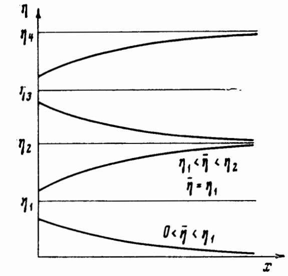
We note also that in the case of exact resonance ($\bar{\epsilon} = 0$) we have

$$f(\eta) = e^{\eta^2/2} I_0\left(\frac{\eta^2}{2}\right), \quad (21)$$

where I_0 is a Bessel function of imaginary argument (formula (21) is valid only for a monochromatic wave, for in the case $\bar{\epsilon} = 0$ the condition $\Delta\omega \ll \epsilon$ is violated). The character of the absorption will depend strongly on the input value of the parameter η, which we denote by $\bar{\eta}$. If $\bar{\epsilon} = 0$, then $f > 0$, and we always have absorption ($\Delta_0 < 0$). With increasing intensity, the absorption coefficient increases exponentially. Therefore when $\bar{\eta} \gg 1$ the wave is strongly absorbed at the beginning of the pass, and then, when η becomes smaller than unity, the usual linear absorption law sets in.

Let us examine now the case $\bar{\epsilon} \neq 0$. As seen from (20), the absorption coefficient becomes in this case an alternating-sign function of the intensity. We denote the roots of f by $\eta_1, \ldots, \eta_k, \ldots,$. Then as $x \to \infty$ we get $\eta \to 0$ if $0 < \bar{\eta} < \eta_1$ and $\eta \to \eta_{2k}$ if $\eta_{2k-1} < \bar{\eta} < \eta_{2k}$ or $\eta_{2k} < \bar{\eta} < \eta_{2k+1}$. We then get amplification in the region $\eta_{2k-1} < \bar{\eta} < \eta_{2k}$ and absorption in the region $\eta_{2k} < \eta < \eta_{2k+1}$. When $\bar{\eta} = \eta_k$, the wave passes through the medium without changing. The values of η_k in the case $\eta_k \gg 1$ can be readily obtained from (20): $\eta_k = (k + 1/2)\pi\gamma/2\bar{\epsilon}$. The character of variation of η at different values of η is shown schematically in the figure. Thus, a wave with a definite value of the amplitude is formed in the medium over a distance on the order of several absorption lengths.

As seen from (19), not only the atoms resonant with the field, but also all other atoms take part in the absorption. This is connected with the nonlinear character of the absorption. For example, besides the single-photon absorption, there can occur also the so-called three-photon interaction, wherein two incident quanta are absorbed, a quantum of frequency $2\omega - \omega_0$ is emitted, and the atom goes over to an excited state. Decay of two photons in accordance with the scheme $2\omega = \omega_1$



$+ \omega_2$ can occur. There is an infinite number of such processes, and their probabilities increase rapidly with increasing intensity. This can explain the increase of the absorption coefficient with increasing intensity in formulas (20) and (21). However, starting with certain values of η it is necessary to take into account not only the drift of photons away from the beam but also their return to the beam.

The increase of the wave amplitude with increasing depth of penetration into a medium with negative excess population, shown in the figure, seems strange at first glance, since a resonant medium with $\Delta_0 < 0$ usually absorbs the radiation. In the case of a real (time-limited) pulse, according to (20), some sections of the pulse will become amplified and others will be absorbed, so that actually only a redistribution of the number of quanta within the pulse itself will take place (see, for example,[1]). For the same reasons, a rearrangement of the angular spectrum of the radiation can occur.

The quantitative results can be obtained by integrating (18). The dependence of the intensity on the length can be written in the form of the following implicit function:

$$\int_{\bar{\eta}}^{\eta} \frac{dz}{zf(z)} = \frac{\sigma\Delta_0}{2}x. \quad (22)$$

in the general case the integral in (22) can be calculated only numerically.

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