Resonant reflection of intense optical radiation from a low-density gaseous medium

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(Submitted 13 August 1984)
Zh. Eksp. Teor. Fiz. **88**, 1147–1152 (April 1985)

The nonlinear optical phenomena accompanying the reflection of light from a low-density gaseous medium are analyzed in the resonance approximation. The spatial dispersion due to the Doppler effect is taken into account. A narrow peak arises against the background of a broad Doppler profile in the spectrum of the reflection coefficient. The width of this new peak is determined in a weak field by homogeneous broadening, while in a strong field it depends on the intensity of the incident light. The specific shape of this peak and its nonlinear properties are intimately related to the nature of the accommodation of the gas atoms at the reflecting surface.

1. The reflection of light at the interface between a transparent dielectric and a resonant gas is highly selective.[1] If the optical density is not too high, the reflection coefficient near an isolated, homogeneously broadened absorption line duplicates the spectrum of the anomalous dispersion.[2] A totally unexpected result is that in cases in which the Doppler mechanism is the governing broadening mechanism the changes do not reduce to simply an averaging of the light reflection coefficient over the equilibrium distribution of the velocities of the gas atoms, as in the case several volume properties. Under the conditions described above, a narrow peak arises in the spectrum of the reflection coefficient against the background of the broad Doppler profile. The width of this new peak is determined by the homogeneous broadening of the resonant transition. An explanation of this phenomenon requires a more careful consideration of the thermal motion of the atoms, which gives rise to a spatial dispersion,[3] and of the collisions of these atoms with the confining surface.[4]

The theoretical[5] and experimental[6–9] results which are presently available deal exclusively with the linear reflection regime, in which the resonant medium remains unsaturated. Our purpose in the present paper is to analyze the nonlinear optical phenomena which arise upon reflection of intense light. This topic is of fundamental interest, and it is obviously of practical interest also, since a narrow peak in reflection can be exploited to narrow down laser output lines,[10] and the sensitivity of this peak to the details of the dynamics of the collisions of excited atoms with the surface makes it possible to extract new information about this topic, which has received little study.

2. We consider the reflection of a monochromatic electromagnetic plane wave

$$E(x,\ t)={}^1/_2E_0\exp i(nkx-\omega t)+\text{c.c.},$$

in a transparent dielectric with a refractive index n as it is incident normally on an interface with a low-density resonant gas. Placing the $x=0$ plane at this interface, we can express the reflection coefficient R in terms of the surface admittance of the gas $M=[dE/dx]_{x=0}/ik\,[E]_{x=0}$

$$R=\left|\frac{n-M}{n+M}\right|^2. \tag{1}$$

Since we would have $M=1$ in vacuum, in a low-density gas we can assume $|M-1|\ll 1$ and then write

$$R=R_0\left[1-\frac{4n}{n^2-1}\operatorname{Re}(M-1)\right],\quad R_0=\frac{(n-1)^2}{(n+1)^2}, \tag{2}$$

where R_0 is the reflection coefficient of the boundary of the dielectric in the absence of the gas.

Working in the resonance approximation, we consider the interaction of the light with only two levels, $|1\rangle$ and $|2\rangle$ ($|\omega_{12}-\omega|\ll\omega$), and we ignore the generation of harmonics. In this case the field and the polarization in the gas (in half-space $x>0$) are

$$E(x,\ t)={}^1/_2E(x)e^{-i\omega t}+\text{c.c.},\quad P(x,\ t)={}^1/_2P(x)e^{-i\omega t}+\text{c.c.}, \tag{3}$$

and their spatial parts satisfy the equation

$$d^2E(x)/dx^2+k^2E(x)=-4\pi k^2P(x). \tag{4}$$

In turn, the polarization is expressed in terms of an off-diagonal element of the density matrix,

$$\rho_{12}(x,\ t,\ V)=\rho_{12}(x,\ V)e^{-i\omega t}$$

as follows:

$${}^1/_2P(x)=N\langle d_{21}\rho_{12}(x,\ V)\rangle, \tag{5}$$

where d_{21} is the transition dipole moment, N is the number density of particles, V is the x projection of the velocity, and the angle brackets mean an average over the ensemble of moving atoms.

The kinetic equation for the density matrix $\rho(x,V)$ simplifies substantially if we can ignore collisions involving a change in velocity. In this case the velocity distribution function

$$N(V)=[\rho_{11}(x,\ V)+\rho_{22}(x,\ V)]N$$

does not depend on x, so that we can write $N\hat{\rho}$ in the

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form $N(V)\hat{f}$, where the density matrix $\hat{f}$, normalized by the condition

$$f_{11}(x, V)+f_{22}(x, V)=1,$$

satisfies the equations

$$V\frac{\partial f_{12}}{\partial x}+[\gamma_\perp+i(\omega_{12}-\omega)]f_{12}=-\frac{i}{2\hbar}d_{12}E(x)D,$$
$$V\frac{\partial D}{\partial x}+\gamma_\parallel(D-D^0)=\frac{i}{\hbar}[d_{12}E(x)f_{12}^*-f_{12}d_{21}E^*(x)]. \quad (6)$$

Here $D(x,V)=f_{11}(x,V)-f_{22}(x,V)$; D^0 is the equilibrium value of the population difference; and $\gamma_\parallel$, $\gamma_\perp$ are the longitudinal and transverse relaxation times in the volume.[1)]

Equations (4)–(6) constitute a closed system from whose solution, with specified boundary conditions, we can find the field in the gas. This system of equations is rather complicated, but the complete system need not be studied here, since in our case of a low-density gas the right side of (4) can be treated as a small perturbation, and we can set $E(x)=E_1\exp(ikx)$ in (6). The substitution

$$f_{12}=(u+iv)e^{ikx},\quad D=-2w$$

converts (6) into an inhomogeneous system of three linear equations with constant coefficients,

$$u'+\Gamma_2u-\Omega v=0,\quad v'+\Gamma_2v+\Omega u-\mathscr{E}w=0,$$
$$w'+\Gamma_1w+\mathscr{E}v=\Gamma_1w^0, \quad (7)$$

which are formally Bloch equations describing the time evolution of a two-level system in a radiation field.[11] In contrast with the actual Bloch equations, the prime here means a differentiation with respect to the coordinate x, rather than with respect to the time, and the frequency difference $\Omega=(\omega_{12}-\omega+kV)/V$ incorporates the Doppler shift:

$$\Gamma_1=\gamma_\parallel/V,\quad \Gamma_2=\gamma_\perp/V,\quad \mathscr{E}=d_{12}E_1/\hbar V,\quad w^0=-{}^1/_2D^0.$$

The general solution of system (7) is

$$\begin{pmatrix}u\\v\\w\end{pmatrix}=\begin{pmatrix}u_{ss}\\v_{ss}\\w_{ss}\end{pmatrix}+\sum_{i=1}^{3}C_i\exp(\mu_ix)\begin{pmatrix}u_i\\v_i\\w_i\end{pmatrix}, \quad (8)$$

where the first term is a particular solution of the inhomogeneous system which does not depend on x; the x dependence of the general solution of the homogeneous system of equations is determined by the characteristic exponents μ_i, since the corresponding eigenvectors (u_i,v_i,w_i) do not depend on x. The constants C_i are fixed by the boundary conditions. Since the sign of $\mathrm{Re}\mu_i$ is opposite that of Γ_i and Γ_2, the condition that the solution be bounded as $x\to+\infty$ at $V>0$ gives us $C_i=0$. For particles which are moving into the interior of the medium after a collision with the surface ($V>0$), the boundary conditions must be imposed at the reflecting surface itself, i.e., at $x=0$. Without specifying these boundary conditions at this point, we will use (8) to evaluate the right side of (4); treating this right side as a perturbation, we then find the first-order correction of $E(x)$. The surface admittance of the gas can thus be written in the form

$$\mathrm{Re}(M-1)=I_1+I_2,\quad I_1=\frac{4\pi d_{21}}{E_1}\int_{-\infty}^{0}u_{ss}(V)N(V)dV, \quad (9)$$

$$I_2=\frac{4\pi d_{21}}{E_1}\int_0^\infty\left[u_{ss}(V)+\mathrm{Re}\sum_{i=1}^{3}\frac{C_i(u_i+iv_i)}{1-i\mu_i/2k}\right]N(V)dV.$$

The first integral describes the contribution of particles moving toward the surface. These particles have a Maxwellian velocity distribution,

$$N(V<0)=N_M(V)=\frac{N}{\pi^{1/2}V_T}\exp\left(-\frac{V^2}{V_T^2}\right)$$

(V_T is determined by the gas temperature), a steady-state polarization, and a steady-state population difference. The second integral describes the contribution from particles which are moving away from the surface. It is this transient process, in which the polarization and population difference of these particles are established, which determines the basic features of the phenomena in which we are interested here. Inelastic processes accompanying the collision of the atoms with the surface can cause the velocity distribution $N(V>0)$ to deviate from a Maxwellian distribution and thereby affect the spectrum of the selective reflection. We turn now to several examples in which we can see the relationship between the spectral features of the reflection and the dynamics of the collisions of excited atoms with the surface.

3. We first consider the contribution to the reflection coefficient made by atoms moving toward the surface. Substituting the steady-state part of solution (8) into (9), we find

$$I_1=m\int_{-\infty}^{0}e^{-y^2}\frac{(\Delta+y)dy}{(\Delta+y)^2+(\gamma_\perp/\gamma_\parallel)r^2+(\gamma_\perp/kV_T)^2}$$
$$\Delta=(\omega_{12}-\omega)/kV_T,\quad m=2\pi^{1/2}d_{21}^2N/\hbar kV_T,\quad r=d_{21}E/\hbar kV_T. \quad (10)$$

The integral in (10) can be expressed in terms of special functions for arbitrary values of the parameters in it. However, it is more useful here to consider various limiting cases. If the field is so weak that it does not cause the saturation of even the resonant atoms, $(d_{21}E/\hbar)^2\ll\gamma_\perp\gamma_\parallel$, we can ignore the field-induced broadening and derive the results of the linear theory.[5] In the case $(\gamma_\perp/\gamma_\parallel)r^2\gg1$, in contrast, the field-induced broadening exceeds the Doppler frequency spread, and expression (10) reduces to a simple dispersive line shape under conditions of pronounced saturation:

$$I_1=\frac{m}{2}\pi^{1/2}\frac{\Delta}{\Delta^2+(\gamma_\perp/\gamma_\parallel)r^2}. \quad (11)$$

We point out that under conditions such that the transverse relaxation is collision-governed the linewidth is proportional to the square root of the pressure. Of greatest interest is the intermediate case, $(\gamma_\perp/kV_T)^2 \ll (\gamma_\perp/\gamma_\parallel)r^2 \ll 1$, in which the field causes saturation of only those atoms which (when the Doppler shift and the field-induced broadening are taken into account) are at resonance with the field, while the greatest part of the Doppler profile remains unsaturated. The profile of the reflection coefficient in this case is analogous to that derived in the linear theory, but the width and height of the peak depend on the field intensity. At the peak, $\Delta^2 \ll 1$, we have

$$I_1 = {}^1/_2 m[\mathrm{C} + \ln(\Delta^2 + (\gamma_\perp/\gamma_\parallel)r^2)], \tag{12}$$

where $\mathrm{C} = 0.577...$ is the Euler constant.[12] The reflection coefficient at the peak thus becomes dependent on the intensity even in a relatively weak field, $d_{21}E/\hbar \sim (\gamma_\perp\gamma_\parallel)^{1/2} \ll kV_T$, while the greater peak of the Doppler profile $\Delta^2 \gg (\gamma_\perp/\gamma_\parallel)r^2$, remains unsaturated at such an intensity and is described by

$$I_1 = {}^1/_2 m[\exp(-\Delta^2)\,\mathrm{Ei}(\Delta^2) + \pi\,\mathrm{Im}\,w(\Delta)], \tag{13}$$

where Ei is the integral exponential function, and w is the probability integral of complex argument—the ordinary convolution of the Doppler and dispersive profiles. Expressions (12) and (13) are joined in the region $(\gamma_\perp/\gamma_\parallel)r^2 \ll \Delta^2 \ll 1$ (Figs. 1 and 2a).

4. We now consider the contribution made to the reflection coefficient by particles moving away from the surface. The simplest assumption, but still quite realistic, is that the atoms moving away from the surface are at a total thermodynamic equilibrium. This assumption means that these particles have a Maxwellian velocity distribution, that they are in the electronic ground state, and that the macroscopic polarization of the incident particles has been completely quenched (we assume that the surface temperature is the same as the gas temperature). It has been shown in the linear theory that in this case the contribution of the particles moving away from the surface is equal to that of the particles approaching the surface. We restrict the derivation of the linear theory to the strong-field limit, $(d_{21}E/\hbar)^2 \gg \gamma_\perp\gamma_\parallel$, in which the characteristic exponents μ_i—the roots of a third-degree equation—can easily be expressed in terms of the parameters of the problem. Ignoring small terms $\sim\gamma_\perp/kV_T$ we find the following integral, which is to be understood in the principal-value sense:

$$I_2 = -m\int_0^\infty e^{-y^2}\frac{(y+\Delta/3)\,dy}{y^2 - 2\Delta y/3 - (\Delta^2+r^2)/3}. \tag{14}$$

For $r \ll 1$, simple expressions for I_2 can be derived in two limiting cases: For $\Delta \ll 1$ we have

$$I_2 = \frac{m}{2}\left[\mathrm{C} + \ln\frac{\Delta^2+r^2}{3} + \frac{4\Delta}{(4\Delta^2+3r^2)^{1/2}}\,\mathrm{arcth}\,\frac{\Delta}{(4\Delta^2+3r^2)^{1/2}}\right], \tag{15}$$

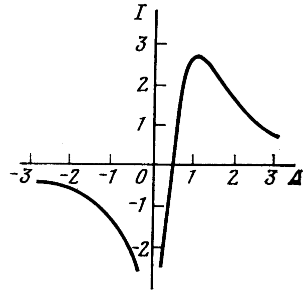

FIG. 1. Spectrum of the resonant part of the reflection coefficient at frequency deviations exceeding the field-induced and homogeneous broadening, $r^2 \ll \Delta^2$, $r^2 \ll 1$, $I = (I_1 + I_2)/m$.

and for $\Delta \gg r$ the integral I_2 is the same as I_1 in (13) (Fig. 2b). Expressions (15) and (13) agree in their common range of applicability, $r^2 \ll \Delta^2 \ll 1$. Consequently, as for incident particles, in the case $r \ll 1$ only a narrow central peak, $\Delta^2 \lesssim r^2$, experiences nonlinear distortions, while the greater part of the profile does not depend on the intensity. Comparing (12) and (15), we see that in the latter case the spectrum has a different functional dependence on the frequency deviation, so that the contributions of the incoming and outgoing particles can be distinguished experimentally.

If the field is so strong that the condition $r \gg 1$ holds, we have

$$I_2 = {}^1/_2 m\pi^{1/2}\Delta/(\Delta^2 + r^2). \tag{16}$$

Comparing (11) and (16), we see that the contributions to the reflection coefficient of the incoming and outgoing particles reach saturation at different radiation intensities, and their spectral lines have different widths.

5. If we drop the assumption of a total thermodynamic equilibrium of the flux of atoms from the reflecting surface, as assumed above, we need to examine in more detail the dynamics of the collisions of the excited and unexcited atoms with the surface. This question is exceedingly complicated and has received little study. In the crudest approximation, the scattered flux may be thought of as consisting of two components: a "diffuse" component comprising a thermodynamically equilibrium re-emission of adsorbed atoms from the scattering surface, and a "specular" component due to elastic reflection. The magnitudes of the scattered fluxes are also affected by adsorption and by quenching of the excited atoms at the surface. It was shown above that the incoming atoms and the diffusely scattered atoms contribute differently to the nonlinear reflection coefficient, so that a study of the nonlinear reflection spectrum could in principle tell us about the relative importance of the diffusely scattered particles. We now show that the specularly reflected particles also make a characteristic contribution to the spectrum of the nonlinear optical reflection coefficient. Denoting the fraction of this component of the scattered flux by η, we find the following expression for the increment to I_2 in the limit of a strong field, $d_{21}E/\hbar \gg \gamma_\perp\gamma_\parallel$:

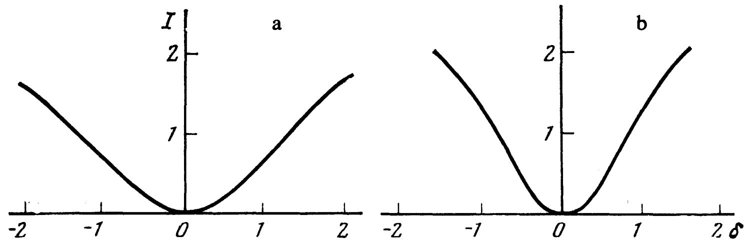

FIG. 2. The same as in Fig. 1, for frequency deviations small in comparison with the Doppler broadening, $\Delta^2 \ll 1$, $(\gamma_\perp \gamma_\parallel)^{1/2} \ll d_{21}E_1/\hbar \ll kV_T$. a) Contribution of incoming particles, $I = 2I_1/m - \ln(r^2\gamma_\perp/\gamma_\parallel) - \mathbf{C}$; b) contribution of diffusely scattered particles, $I = 2I_2/m - \ln(r^2/3) - \mathbf{C}$, $\delta = \Delta/r$.

$$\Delta I_2=\eta m r^2 \frac{\gamma_\perp}{\gamma_\parallel} \times \int_0^\infty e^{-y^2} \frac{(y+\Delta/3)\,dy}{[y^2-2\Delta y/3-(\Delta^2+r^2)/3][(\Delta-y)^2+r^2\gamma_\perp/\gamma_\parallel]}. \qquad (17)$$

It is not difficult to see that the increment is large only if $\Delta^2 \lesssim r^2\gamma_\perp/\gamma_\parallel$, and in the limit $\Delta^2 \ll r^2\gamma_\perp/\gamma_\parallel$ it takes the relatively simple form

$$\Delta I_2=\eta \frac{m}{2} \frac{3\gamma_\perp/\gamma_\parallel}{(1+3\gamma_\perp/\gamma_\parallel)^2} \times \left[\ln\frac{3\gamma_\perp}{\gamma_\parallel}+\frac{\gamma_\perp}{\gamma_\parallel}\ln\frac{27\gamma_\perp}{\gamma_\parallel}-\frac{4\pi}{3}\left(\frac{\gamma_\parallel}{\gamma_\perp}\right)^{1/2}\frac{\Delta}{r}\right]. \qquad (18)$$

The part of the component of specularly scattered particles which is odd in Δ can be described by the simple expression

$$\Delta I_2^{\text{odd}}=\eta 2\pi m\left(\frac{\gamma_\perp}{\gamma_\parallel}\right)^{1/2} \frac{\Delta/r}{16(\gamma_\perp/\gamma_\parallel)(\Delta/r)^2+(1+3\gamma_\perp/\gamma_\parallel)^2}$$

over a broader region, $\Delta \ll 1$.

A characteristic feature of this result is that the contribution of the specularly scattered particles to the optical reflection spectrum is asymmetric with respect to the frequency of the resonant transition, $\Delta = 0$, even in the case $\Delta \ll 1$. This circumstance should result in a shift of the peak in the reflection coefficient; the magnitude of the shift will depend on the fraction of the particles which are scattered specularly.

6. The results presented above obviously do not cover all the situations that might arise during the reflection of intense radiation from a low-density resonant medium. We wish to emphasize two aspects of this phenomenon. First, nonlinear effects can be see in the reflection at relatively low incident-radiation intensities, at which the greater part of the ensemble of moving atoms is unsaturated. Second, the spectrum of the nonlinear optical reflection coefficient of the interface between a gas and a transparent dielectric is extremely sensitive to the detailed behavior of the gas atoms as they collide with the surface of the dielectric. When some model is adopted for the dynamics of the scattering of gas atoms by the reflecting surface, we can use the data from nonlinear reflection spectroscopy to test and refine the model.

I wish to thank V. V. Khromov for suggesting the topic and for support, S. G. Przhibel'skiĭ for useful comments, and A. M. Bonch-Bruevich and the participants of his seminar for a discussion of these results.

[1)]Only radiative relaxation takes place in a fully collisionless gas and one must put $\gamma_\parallel = 2\gamma_\perp$. We have retained here both relaxation constants, inasmuch as in situations typical of optical transitions the cross section for collision with change of velocity is much less than the broadening-collision cross sections. A pressure interval exists in this case, in which Eqs. (6), which do not take the velocity mixing into account, are still valid, but $2\gamma_\perp > \gamma_\parallel$.

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Translated by Dave Parsons