Oscillations in nonlinear transmission of a rarefied resonant gas—a manifestation of optical nutation under steady-state conditions

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A new nonlinear optical effect is predicted. This effect involves a modulation of the intensity of a stationary monochromatic plane wave which propagates from the boundary surface into a rarefied resonant gas. The amplitude and spatial modulation period depend on the intensity and frequency of light.

The optical properties of gases near the absorption line acquire, if Doppler broadening is dominant, several unexpected features which are attributable to a strong spatial dispersion.[1] Thus, for example, a sub-Doppler structure,[2,3] which undergoes characteristic changes as the intensity of light is increased, appears in the spectrum of a reflection from the boundary of such a gas even in a weak field.[4] In this letter we show that a three-dimensional structure of a field that enters a medium also changes significantly when an intense light strikes the boundary of the medium.

Let us consider the following situation. A monochromatic plane wave strikes at right angles the boundary of the gas that occupies a half-space $x>0$ of the region $x<0$, $(1/2)E_0 \exp i(kx-\omega t)+$ c.c. Restricting the analysis to a resonance approximation, we will consider the interaction of the light with only two discrete levels $|1\rangle$ and $|2\rangle$ ($|\omega_{12}-\omega| \ll \omega$) and disregard the generation of harmonics. In this case the field in the gas is given by $E(x,t)=(1/2)E(x)\exp(i\omega t)+$ c.c., where the three-dimensional part of $E(x)$ can be determined by simultaneously solving Maxwell's equation and a kinetic equation for the density matrix $\rho(x, v)$ (v is the velocity of an atom). In the case of a rarefied gas, in which collisions can be ignored, the velocity-distribution function $N(v)=N[\rho_{11}(x, v)+\rho_{22}(x, v)]$ (N is the total particle density) does not depend on x, which greatly simplifies the kinetic equation. To further simplify the mathematical structure of the equations, we restrict the discussion to the case in which the optical density of the gas is small. Disregarding in first approximation the reaction of the medium to the field, we solve the kinetic equation for the density matrix by substituting into it the unperturbed $E(x)=E_0 \exp(ikx)$ and we then use the solution to calculate the polarization of the medium. We should note that the contribution to the polarization from the atoms that move toward the gas boundary (a transparent insulator may be the boundary) is different from the contribution from the atoms that move away from the boundary not only because the atoms have different Doppler shifts but also because the initial conditions under which they are excited are completely different. The former atoms remain in the field a long time and therefore are stable, whereas the latter undergo an optical nutation due to the instantaneous application of the field at the moment of recoil.[5,6] To complete the calculations, we must define concretely the conditions for the scattering of atoms from a reflecting surface. A simple and suffi-

ciently realistic assumption is that the atoms that leave the surface are in a complete thermodynamic equilibrium with it. This means that these atoms have a Maxwellian velocity distribution (they are in the electronic ground state) and that the macroscopic polarization of the incident particles is completely extinguished. Knowing the polarization that the groups of atoms produce collectively, we can find, by integrating over the Maxwellian velocity distribution, the macroscopic polarization of the medium, and proceeding in the spirit of optical-density perturbation theory, we find the correction to the field. As a result, we can represent the field in the medium as $E(x) = E_0 \times \exp(i\tilde{k}x)[1 + I(x)]$, where $\tilde{k}$ is the wave vector which was renormalized with allowance for the polarization of the medium, and $I(x)$ is a correction for the optical nutation. If the field is resonantly tuned to a certain group of atoms that are emitted into the gas, $\omega > \omega_{12}$, and if it is strong enough to saturate the resonant transition, $dE_0 \gg \hbar\gamma$ (d and γ are the dipole moment and the uniform resonant-transition width), but at the same time not strong enough to saturate the entire Doppler contour, $dE_0 \ll \hbar|\omega_{12} - \omega| \lesssim \hbar k v_T$ (v_T is the characteristic thermal velocity of the gas atoms), then we will have

$$I(x) = 2\sqrt{\pi}\,\frac{Nd(\omega-\omega_{12})}{E_0 k v_T}\exp\left[-\left(\frac{\omega-\omega_{12}}{k v_T}\right)^2\right] F\left(\frac{x}{x_0}\right), \tag{1}$$

$$x_0 = \lambda\,\frac{\hbar(\omega-\omega_{12})}{dE_0}; \qquad F(X) = 1 - \int_0^X J_0(\alpha)\,d\alpha,$$

where J_0 is the Bessel function. The plot of the F function for $x \sim x_0$ is shown in Fig. 1. For $x/x_0 \gg 1$, F rapidly reaches the asymptotic form:

$$F\left(\frac{x}{x_0}\right) = \sqrt{\frac{2x_0}{\pi x}}\cos\left(\frac{x}{x_0} + \frac{\pi}{4}\right). \tag{2}$$

We see from Fig. 1 and from asymptotic expression (2) that the oscillations of the amplitude of the field in the gas become slowly damped. The period $2\pi x_0$ of these oscillations in three-dimensional space depends on the intensity and frequency of light. Under the conditions considered by us, a considerable number of oscillations can fit

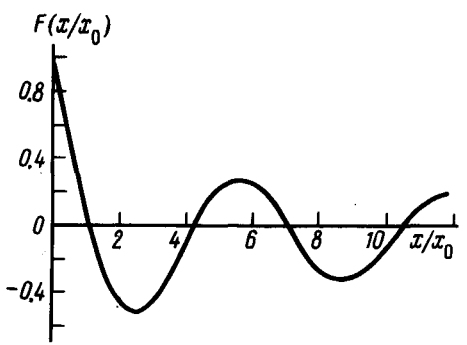

FIG. 1.

into the region in which (1) is applicable: $x/x_0 \ll \min\{dE_0/\hbar\gamma; \hbar(\omega - \omega_{12})/dE_0\}$. The period of these oscillations is much longer than their wavelength.

The modulation depth is $\hbar\gamma/dE_0$ in order of magnitude. The most obvious effect of modulation of the field amplitude is the oscillation of the nonlinear transmissivity of a thin layer of rarefied gas as a function of the thickness of the layer and as a function of the frequency and intensity of the emitted light. Another feature of this phenomenon is the spatial modulation of the population difference of the resonant levels. This modulation is the same as that of the field amplitude in every respect but the phase, which is opposite. The polarization of the medium, like the indicated quantities, oscillates in space due to the quarter-period shift, giving rise to spatial modulation of the scattered light.

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