Effect of radiation pressure on the nonlinear susceptibility of resonant atoms

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The ponderomotive effect of light on resonant atoms with strong transitions over sufficiently long interaction times causes substantial changes in not only the magnitude of the nonlinear response of a system but also its parity as a function of the deviation from the resonant frequency.

The nonlinear response of a gas of resonant atoms is usually attributed to a perturbation of internal degrees of freedom. Under the influence of the field, Bennett peaks and holes arise in the distributions of particles in working levels, while the overall velocity distribution of the atoms remains an equilibrium distribution.

In the present letter we examine another source of nonlinearity: the effect of radiation pressure on the translational motion of the particles, which leads to a perturbation of the velocity distribution of the atoms. The influence of the recoil effect on the optical characteristics of atoms was first studied by Kol'chenko *et al.*[1] in the case of narrow resonances. When the recoil energy exceeds the line width $\epsilon_r = \hbar k^2/2m \gg \gamma$, the Lamb dip splits into two symmetric dips. This subtle nonlinear effect, which requires a high resolution ($\gamma/\omega \sim 10^{-10}$), has been observed experimentally.[2,3]

We are interested here in atoms with strong transitions, $\gamma \gg \epsilon_r$ (e.g., $\epsilon_r/\gamma \sim 10^{-3}$ for sodium), in which case the recoil effect is manifested in the kinetic stage of the interaction of the resonant gas with the field. The resonant dependence of the radiation-pressure force $f(v)$ on the velocity leads to a redistribution (bunching) of particles in velocity space.[4,5] This bunching strongly affects the nonlinear response if the change in the velocity of the resonant particle, $\delta v \sim \tau f/m$, reaches a value on the order of the width of the resonance, γ/k. For the spontaneous radiation pressure, $f \sim \hbar k \gamma$, this condition is satisfied at $\epsilon_r \tau \gtrsim 1$; i.e., the thickness of the light beam must be quite large: $a \gtrsim v_0/\epsilon_r \sim 0.1$ (v_0 is the thermal velocity). Since the number of spontaneous transitions, $\gamma\tau$, is large, the lower working level of the atoms must be the ground level.

To find the susceptibility $\chi(|E|^2)$ of a gas in a resonant field $E(x)\exp(-i\Delta t)$ (Δ is a small frequency deviation from resonance), we work from the standard Bloch equations for the density matrix of an atom and from the kinetic equation for the velocity distribution function $F(v,t)$:

$$\frac{\partial F}{\partial t} + \frac{\partial}{\partial v}\left[\frac{f(v)}{m} F\right] = 0. \qquad (1)$$

This equation reflects only the drift motion (along the light beam) under the influence of the radiation-pressure force $f(v)$; the velocity diffusion of the particles is ignored. This simplification is legitimate if the interaction time is not too long[5]: $\epsilon_r \tau \ll (\gamma/\epsilon_r)^{3/5}$.

We restrict the analysis to the case of a slight distortion of the equilibrium distribution function: $F(v,t) = F_0(v) + F_1(v,t)$, where F_1 is found by perturbation theory,

$$F_1(v,t) = -\frac{\tau}{m}\frac{\partial}{\partial v}\left[f(v)F_0(v)\right], \tag{2}$$

and τ is an effective time of transit of the atoms through the light beam.

1. In the case of the traveling wave $E = E_0 \exp(ikx - i\Delta t)$, we have[6] $f(v) = \hbar k\gamma w(\Delta - kv)$, where $w(\Delta) = (1/2)G[4\Delta^2 + \gamma^2 + G]^{-1}$ is the population of the upper level, and $G = 8(dE_0/\hbar\gamma)^2$ is the saturation parameter. The condition $F_1 \ll F_0$ holds if $\epsilon_r \tau w \ll 1$. In the case of a slight saturation ($w \ll 1$), expression (2) can also be used at $\epsilon_r \tau \gtrsim 1$.

Using an equilibrium distribution, $n(\sqrt{\pi}v_0)^{-1}\exp[-(v/v_0)^2]$, as F_0 (n is the density of atoms), we find the following expression for the nonlinear susceptibility $\chi = \chi' + i\chi''$ at $|\Delta| \lesssim \Delta_0 = kv_0$:

$$\chi'_{\rm non} = \chi_0(\Delta)G\left[\frac{\gamma\Delta}{\Delta_0^2}(1+\sqrt{1+G})^{-1} + \frac{1}{2}\epsilon_r\tau(1+G)^{-3/2}\right],$$

$$\chi''_{\rm non} = \chi_0(\Delta)\frac{G}{\sqrt{1+G}}\left[-(1+\sqrt{1+G})^{-1} + \frac{\gamma\Delta}{2\Delta_0^2}\epsilon_r\tau(1+G)^{-1}\right], \tag{3}$$

$$\chi_0 = \frac{\pi d^2}{\hbar k}F_0(\Delta/k).$$

The first terms in square brackets here stem from the ordinary saturation effect, while the terms proportional to $\epsilon_r\tau$ stem from the velocity bunching of the particles. We see that the radiation pressure changes the parity of the susceptibility as a function of the frequency deviation from resonance: A component which is even in Δ appears in $\chi'_{\rm non}$, while an odd component appears in χ''_{non}. In the absorption, this component is small, proportional to $\gamma\Delta/\Delta_0^2$, while in the real part it is, in contrast, large, in proportion to this parameter. This circumstance can be understood easily by noting that $\chi'_{\rm non}$ is determined by the derivative dF/dv at $v = \Delta/k$. For this reason, although the change in the distribution function is slight ($F_1 \ll F_0$), the derivative of this function changes significantly because of the resonant dependence of the force on the velocity. In the interval $\gamma\Delta/\Delta_0^2 < G\epsilon_r\tau$ of frequency deviations from resonance, the component stemming from the recoil effect is greater than the linear part $\chi'_{\rm lin}$.

2. Structural features stemming from the recoil effect can be observed in the response by another method also, the method of a probling field, from the absorption and phase shift of a weak (oppositely directed) signal $E_1 e^{-ikx - i\Delta_1 t}$ in the presence of a strong field $E_0 e^{ikx - i\Delta t}$. The linear susceptibility of a weak field, $\chi'_1 + i\chi''_1$, is given in first order in the saturation parameter for the strong field G by

$$\chi_1' = \frac{1}{2}\chi_0(-\Delta_1)\frac{G}{1+\delta^2}\left[\delta + \epsilon_r\tau\frac{\delta^2-1}{\delta^2+1}\right], \tag{4}$$

$$\chi_1'' = -\frac{1}{2}\chi_0(-\Delta_1)\frac{G}{1+\delta^2}\left[1 + 2\epsilon_r\tau\frac{\delta}{\delta^2+1}\right], \quad \delta = \frac{\Delta+\Delta_1}{\gamma}.$$

This linear susceptibility changes substantially because of the radiation pressure of the strong field if $\epsilon_r\tau \gtrsim 1$ and $|\delta| \lesssim 1$. At large values of δ, this component falls off rapidly.

3. Finally, the nonlinear susceptibility in the field of a standing wave $E_0 \cos kx\, e^{-i\Delta t}$, near the center of the line ($\Delta \ll \Delta_0$), for a weak saturation, is

$$\chi'_{\rm non} = \chi_0(0)\left(\frac{dE_0}{\hbar\gamma}\right)^2 \frac{\delta}{1+\delta^2}\left(1 + \epsilon_r\tau\delta\frac{\delta^2+3}{\delta^2+1}\right), \tag{5}$$

$$\chi''_{\rm non} = -\chi_0(0)\left(\frac{dE_0}{\hbar\gamma}\right)^2\left[1 + \frac{1}{1+\delta^2} + \epsilon_r\tau\frac{2\delta}{(1+\delta^2)^2}\right], \quad \delta = 2\Delta/\gamma.$$

In a standing wave, the friction force $f(v)$ accelerates atoms if $\delta > 0$. The fraction of particles that are at resonance decreases in this case, causing an additional brightening of the system. At $\delta < 0$, the particles are slowed, and the fraction of slow particles increases. For this reason, the component stemming from the radiation pressure has the sign opposite that of the saturation effect. In particular, under the conditions $\delta < 0$ and $\epsilon_r\tau \gtrsim 1$, the sign of $\chi_{\rm non}$ changes. The effect of radiation pressure on the nonlinear absorption of a standing wave was discussed in Refs. 7 and 8.

In summary, the effect of radiation pressure on the nonlinear optical properties of atoms with strong transitions becomes important at $\epsilon_r\tau \gtrsim 1$. Here there are changes in not only the magnitude of the susceptibility $\chi_{\rm non}$ but also its parity as a function of the frequency deviation from resonance. Observations of these effects in the optical frequency range will require light beams with a thickness $\gtrsim 0.1$–1 cm. A further requirement, of course, is that there be no processes of the optical-pumping type, which would disrupt the cyclic nature of the interaction of the atoms at the working transition with the field. Since these conditions are not unusual for nonlinear spectroscopy, it may be that this radiation-pressure effect has been observed experimentally. For example, a slight frequency asymmetry was detected by Akulshin *et al.*[9] in the structure of nonlinear resonances in the vapor of alkali metals. This asymmetry may possibly be a manifestation of radiation pressure, since both of the conditions listed above—broad beams and a closed tranistion—were satisfied.

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