Coherent dragging of atomic populations in problems of resonant light pressure

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We investigate the influence of coherent dragging of populations on the light pressure exerted on a three-level atom in a two-frequency light field. The Λ , V, and cascade configuration schemes of the atomic levels are considered. It is shown that in the case of a Λ atom the influence of the coherent dragging manifests itself differently in comoving and contermoving light waves. In the former case, at exact resonance between the waves and the transitions, the light pressure is zero regardless of the atom velocity. In the latter case the influence of the coherent dragging is substantial only at some definite atom velocity. For the V and cascade configurations, the light-pressure force differs from zero at all atom velocities.

I. INTRODUCTION

The pressure of resonant optical radiation on a twolevel atom has been recently investigated in detail many times.¹ At the same time, experimental progress in the control of atom motion by laser-radiation pressure has demonstrated the advantages of exciting atoms by multifrequency radiation that is at resonance with several atomic transitions. In particular, the use of multifrequency radiation is effective in laser cooling of atoms.² An advantage of the multifrequency excitation is that it permits prolonged resonant interaction of the atoms with the radiation. The latter, in turn, permits an appreciable light pressure on the atom to be maintained for a long time.

It is known that excitation of a multilevel atom by multi-frequency radiation has a number of features not possessed in the case of a two-level atom. Principal among them is coherent dragging of atomic populations.³⁻⁵ This dragging forbids in many cases an atom to be excited from sublevels of the ground states. This means that an atom situated in a resonant-radiation field may possibly not be acted upon by the light pressure.

The simplest multilevel scheme in which atomic-coherence effects are significant is a three-level atom. The question of resonant-radiation pressure on a three-level atom was considered earlier^{6,7} in connection with an analysis of methods of laser cooling of sodium atoms. The three-level atom is the model of a real multilevel atom that has a hyperfine structure in the ground and excited states. No account was taken in Refs. 6 and 7, however, of the atomic-states coherence, which is immaterial in the presence of a large number of closely-spaced levels.

We report here an analysis of the features of resonant light pressure for a nondegenerate three-level atomic system excited by two-frequency optical radiation. We consider three possible schemes of exciting the three-level atom by two monochromatic light waves and indicate for each scheme the dependences of the light-pressure force and of the diffusion tensor on the atom velocity. We emphasize that the manifestation of the effect of atomic coherence in problems dealing with resonant light pressure differs from its manifestation in spectroscopic problems of atom excitation, for in the former case we include the atom motion into consideration. In spectroscopic problems, the effectiveness of excitation of an immobile atom is determined only by the conditions of the resonance between the immobile atom and the frequencies of the atomic transitions. In resonant-light-pressure problems the Doppler effect causes in addition a dependence of the excitation on the atom velocity and on the exciting-wave-vector directions.

2. QUALITATIVE ANALYSIS OF ATOMIC COHERENCE

Consider in succession the atomic-level schemes shown in Fig. 1. Assume that a Λ -configuration atom (Fig. 1a) is excited by two waves, both propagating in the positive z direction. The effectiveness of exciting an atom having a velocity projection v_z depends on the conditions of the resonance between the atomic transitions and the radiation. Let the wave of frequency ω_2 be at resonance with the transition $|2\rangle \rightarrow |3\rangle$, and the wave of frequency ω_1 at resonance with $|1\rangle \rightarrow |3\rangle$. In view of the Doppler effect, the frequencies of the exciting waves in the atom's rest system are respectively $\omega_1 + kv_z$ and $\omega_2 + kv_z$. Coherent dragging of the population occurs in the Λ configuration when the difference of the





wave frequencies, considered in the atom's rest system, coincides with the frequency interval between levels $|1\rangle$ and $|2\rangle$ of the ground state $^{3-5}$:

$$(\omega_1 - \omega_{31} + kv_z) - (\omega_2 - \omega_{32} + kv_z) = (\omega_1 - \omega_2) - \omega_{21} = 0.$$
(1)

Regardless of the velocity, the atom is not excited by the light field in this case. In other words, when the Λ atom is in a resonant field, it it not subject to the action of the light pressure under condition (1). The situation is different for a Λ configuration of atomic levels in the case of opposing light waves. Assume that a wave of frequency ω_1 propagates in the negative z direction, and a wave with frequency ω_2 in the positive. In this situation, the coherent-dragging condition takes in the rest system of the atom the form

$$(\omega_1 - \omega_{31} - kv_z) - (\omega_2 - \omega_{32} + kv_z) = (\omega_1 - \omega_2) - \omega_{21} - 2kv_z = 0.$$
 (2)

It follows hence that if the waves propagate counter to each other an atom with a Λ level configuration is not excited by the radiation only at one velocity that satisfies the condition (2).

The failure to excite the atom under conditions (1) and (2) is physically due to the onset of a coherent superposition of the lower states $|1\rangle$ and $|2\rangle$. This superposition of the atomic state is not coupled optically with the upper state $|3\rangle$, so that the atom remains at the lower levels under conditions (1) and (2).

If the atomic levels are in a V configuration (Fig. 1b), the atom is excited into the upper states $|1\rangle$ and $|2\rangle$ at any velocity. The absence of coherent dragging in the case of a V atom is due to radiative decay of the atomic coherence between the upper levels. Accordingly the radiation pressure on the atom does not vanish at any atom velocity. For the same reason, coherent dragging is likewise not decisive in a cascade configuration of atomic levels (Fig. 1c).

It follows thus from the foregoing qualitative considerations that coherent dragging can influence strongly the light pressure only in the case of a Λ configuration of the atomic levels. We consider below a kinetic equation for Λ atoms that interact with two light waves.

3. BASIC EQUATIONS

We assume that the light field is a superposition of two Gaussian wave with polarization unit vectors \mathbf{e}_1 and \mathbf{e}_2 , frequencies ω_1 and ω_2 , and wave vectors \mathbf{k}_1 and \mathbf{k}_2 ;

$$E(\mathbf{r}, t) = \mathbf{e}_{1}E_{1} \exp \left[-(x^{2}+y^{2})/2\rho_{0}^{2}\right] \cos(\omega_{1}t \mp \mathbf{k}_{1}\mathbf{z}) + \mathbf{e}_{2}E_{2} \exp \left[-(x^{2}+y^{2})/2\rho_{0}^{2}\right] \cos(\omega_{2}t - \mathbf{k}_{2}\mathbf{z}).$$
(3)

We have introduced here a Cartesian coordinate frame whose z axis coincides with the propagation direction of the ω_2 wave. The quantity ρ_0 determines the scale of the field of the beams in the XY plane. Each beam in (3) is a laser beam with TEM_{00a} fundamental mode.

We assume for the sake of argument that the waves of frequency ω_1 and ω_2 excite respectively transitions $|1\rangle \rightarrow |3\rangle$ and $|2\rangle \rightarrow |3\rangle$ transitions. The $|1\rangle \rightarrow |2\rangle$ transition is assumed to be dipole-forbidden. The level $|3\rangle$ decays with a total

spontaneous-emission probability $\Gamma = 4\gamma$, while the partial probabilities of the transitions $|3\rangle \rightarrow |2\rangle$ and $|3\rangle \rightarrow |1\rangle$ are assumed equal.

To describe the atomic motion it is convenient to use an initial density matrix in the Wigner representation. Without dwelling on the well known equations for the Wiger density matrix,⁸ we note that for the functions that determine the relative probability of spontaneous photon emission in a definite direction we assume the spherical-symmetry approximation.9

Next at times $t \gg \gamma^{-1}$ and under conditions $\hbar k_i^2 / k_i^2$ $2M \ll \hbar \gamma$ (i = 1, 2), which are satisfied for dipole transitions of atom, the system of equations for the atomic density matrix can be reduced to a single kinetic equation for a classical distribution function $w(\mathbf{r}, \mathbf{p}, t)$.^{10,11} To this end it is necessary to expand the Wigner density-matrix elements in powers of the photon momenta and use the rotating-wave approximation and the rate-equations approximation. These transformations yield a system of equations that is conveniently written in terms of Bloch variables^{6,7}:

$$\frac{dw}{dt} = -\hbar k V \frac{\partial}{\partial p_z} (f \pm s) - \sum_{i=x,y} \hbar \frac{r_i}{\rho_0^2} \frac{\partial}{\partial p_i} V(g+q) \\
+ \frac{1}{3} \hbar^2 k^2 \gamma \sum_{i=x,y,z} \frac{d^2}{dp_i^2} (w-Q-R), \quad (4a)$$

$$\frac{d}{dt} Q = -2Vs - Vf + 2\gamma (w-Q-R) + \frac{\hbar k}{2} V \frac{\partial}{\partial p_z} f + \dots, \\
\frac{d}{dt} R = -Vs - 2Vf + 2\gamma (w-Q-R) \pm \frac{\hbar k}{2} V \frac{\partial}{\partial p_z} s + \dots, \\
\frac{d}{dt} S = \alpha q + 2VQ + Vu - \gamma s \mp \frac{1}{3} \hbar k V \frac{\partial}{\partial p_z} (2w-2R+Q) \\
- \frac{1}{2} \hbar k V \frac{\partial}{\partial p_z} u + \dots, \\
\frac{d}{dt} q = -\alpha s - \gamma q - Vn + \frac{1}{2} \hbar k V \frac{\partial}{\partial p_z} n + \dots, \\
\frac{d}{dt} f = \xi g + 2VR + Vu - \gamma f - \frac{1}{3} \hbar k V \frac{\partial}{\partial p_z} (2w-2Q+R) \\
= \frac{1}{2} \hbar k V \frac{\partial}{\partial p_z} u + \dots, \quad (4b)$$

$$\frac{d}{dt}g = -\xi f - \gamma g + Vn \mp \frac{1}{2}\hbar kV \frac{\partial}{\partial p_z}n + \dots,$$

$$\frac{d}{dt}u = -\Omega n - Vf - Vs - \frac{1}{2}\hbar kV \frac{\partial}{\partial p_z}(s\pm f) + \dots,$$

$$\frac{d}{dt}n = \Omega u - Vg + Vq + \frac{1}{2}\hbar kV \frac{\partial}{\partial p_z}(q\mp g) + \dots,$$

where

 $Q = \rho_{11} - \rho_{33}, \quad R = \rho_{22} - \rho_{33}, \quad is = \rho_{31} - \rho_{13},$ $w = \rho_{11} + \rho_{22} + \rho_{33},$ $in = \rho_{21} - \rho_{12}, \quad q = \rho_{31} + \rho_{13},$ $if = \rho_{32} - \rho_{23}$ $g = \rho_{32} + \rho_{23}$ $u = \rho_{21} + \rho_{12}$.

We have introduced here equations for the detunings

(4b)

$$\Omega_{31} = \omega_1 - \omega_{31}, \quad \alpha = \Omega_{31} \mp k v_z,$$

$$\Omega_{32} = \omega_2 - \omega_{32}, \quad \xi = \Omega_{32} - k v_z, \quad \Omega = \alpha - \xi$$

and for the Rabi frequencies, which we assume to be equal to

$$V_{ij} = -\langle i | \mathbf{de} | j \rangle E/2\hbar.$$

The upper and lower signs in the system (4) correspond to light waves propagating with and counter to the beams, respectively.

To obtain from the microscopic equations (4) a kinetic equation for the atomic distribution function $w(\mathbf{r}, \mathbf{p}, t)$ we can apply to the system (4) the Bogolyubov analysis used earlier in Ref. 10. Such a derivation of the kinetic equation is premised on the treatment of the functions $h(\mathbf{r}, \mathbf{p}, t) = Q, R, s$, f, q, g, u, n as functionals of the distribution function

$$h(\mathbf{r}, \mathbf{p}, t) = h[\mathbf{r}, \mathbf{p}; w(\mathbf{r}, \mathbf{p}, t)].$$
(5)

Using this method, which was described earlier in Ref. 10, we can write an equation for w in second order in the photon momenta:

$$\frac{\partial}{\partial t}w = -\frac{\partial}{\partial p_z}(F_z w) - \sum_{i=x,y}\frac{\partial}{\partial p_i}(F_i w) + \sum_{i=x,y,z}\frac{\partial^2}{\partial p_i^2}(D_{ii} w).$$
(6)

In the case of light beams propagating in the same direction [the upper sign in (4)] the components of the radiation force acting on a Λ atom are equal to (i = x, y)

$$F_z = \hbar k \gamma \cdot 4G \Delta^2 L^{-1}, \tag{7a}$$

$$F_i = \hbar \gamma r_i / \rho_0^2 \cdot 2G\Delta \left(\Delta_2^2 - \Delta_1^2 \right) L^{-1}.$$
(7b)

The components of the momentum-diffusion tensor are

$$D_{xx} = D_{yy} = \hbar^2 k^2 \gamma D_c, \quad D_{zz} = \hbar^2 k^2 \gamma [D_c + D_d (1+d)], \\ D_c = 2G \Delta^2 L^{-1}, \quad D_d = 2G \Delta^2 L^{-1},$$
(8a)

and the nonadiabatic increment to the diffusion tensor¹⁰ is defined as

$$d = [2\Delta^{3}(\Delta_{1} + \Delta_{2}) (G - \Delta^{2}) (4\Delta_{2}^{2} + 4 - G) -8\Delta^{3}(\Delta_{1} + \Delta_{2}) (\Delta_{2}\Delta - G)G +4G\Delta^{4}(\Delta_{2}^{2} - 1) + 2G^{2}\Delta^{2}(3\Delta_{1}^{2} + 5\Delta_{2}^{2}) + 4G^{3}\Delta(3\Delta_{2} + \Delta_{1}) +2G^{2}(\Delta^{2} - 2G)^{2} + 4G\Delta^{2}(G - \Delta^{2}) (1 + G)]L^{-2}.$$
(8b)

For opposing light beams (lower sign in (4) the radiation-force components F_x and F_y and diffusion coefficients D_c and D_d are also determined by expressions (7b) and (8a). In this case, however, the light-pressure force is zero $F_z = 0$ at any atom velocity and the expression for the nonadiabatic increment to the diffusion tensor takes the form

$$d = 2(\Delta_1^2 - \Delta_2^2) (\Delta^2 - G) L^{-1}.$$
(9)

We have introduced here the notation

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 $\Delta_1 = \alpha/\gamma, \ \Delta_2 = \xi/\gamma, \ \Delta = \Omega/\gamma, \ G = 2V^2/\gamma^2,$ $L=2(\Delta_1^2+\Delta_2^2+2)\Delta^2-G\Delta^2+4G^2.$

We emphasize that the kinetic equation (6) with the coefficients (7)-(9) is meaningful in the rate-equations approximation. These equations, generally speaking, limit the permissible light-wave intensity in accordance with the relation $(\gamma^2 + 2V^2) \ll \omega_{21}.$

4. RADIATION PRESSURE ON Λ ATOM

Consider the behavior of the light-pressure force and of the momentum-diffusion tensor in the case of a Λ atom in light beams having the same direction. The light pressure force F_z and the diffusion tensor D_{ii} depend substantially, according to (7a) and (8a), on the resonance between the light waves and the atomic transitions. We can distinguish here between two basic cases.⁷ The first is resonance, when the light-pressure force is close to zero at any atom velocity. This case is realized at light-field frequencies that are close to those of the atomic transitions

$$|\Omega| = |\Omega_{31} - \Omega_{32}| \ll \gamma (1+G)^{\frac{1}{2}}.$$
 (10)

The second case is that of appreciable detuning from exact resonance:

$$|\Omega| = |\Omega_{31} - \Omega_{32}| \gg \gamma (1+G)^{\frac{1}{2}}.$$
(11)

We discuss first the behavior of the light-pressure force F_{r} (Fig. 2a). If condition (10) is satisfied the effect of coherent dragging of the population decreases radically the lightpressure force (7a). At exact resonance $|\Omega| = 0$ the light pressure force vanishes. With increasing detuning from the exact resonance the light-force pressure F_z first increases in absolute value because of the decreased contribution of the effect of coherent dragging, and reaches a maximum value (for a fixed saturation parameter G) at $|\Omega| \sim \gamma (1+G)^{1/2}$ (Fig. 2a, curve 3). With further increase of the detuning (curves 4 and 5) the light-pressure force is decreased by optical pumping of one of the lower levels. The components F_x and F_y of the gradient force behave similarly (Fig. 2b). The components F_x and F_y , however decrease much less abruptly than the force F_z , since the gradient force is governed by the polarizability of the atom and is not greatly influenced by optical pumping.



FIG. 2. Dependence of the radiation-force components F_z (a) and F_x (b) on the atom velocity at different detunings of the frequency ω_2 of the second wave from resonance with the transition $|2\rangle - |3\rangle$ for $G = 10^3$, $\Omega_{31}/\gamma = 270$. The numbers at the curves correspond respectively to detunings $\Omega_{32}/\gamma = 250, 240, 230, 200, 170.$



FIG. 3. Gradient-force component F_x vs the atom velocity in the case of opposing saves at $G = 10^3$ and $\Omega_{32}/\gamma = -270$. The numbers on the curves correspond respectively to detunings $\Omega_{31}/\gamma = 265$, 130, and 30.

The diffusion-tensor components D_{ii} have in the case of waves propagating in the same direction a smooth bellshaped dependence on the velocity projection v_z . The incluence of the population dragging and of the optical pumping on the diffusion tensor are similar to their influence on the light-pressure force F_z .

We consider now the case of opposing waves. The interesting effect here is the vanishing of the light-pressure force F_z at any A-atom velocity. The reason is the equality of the relaxation constants in the transitions $|3\rangle \rightarrow |1\rangle$ and $|3\rangle \rightarrow |2\rangle$ and of the moduli of the wave vectors of the opposing waves, $|k_1| = |k_2| = \omega_{31}/c = \omega_{32}/c$. The gradient force has in the case of opposing waves a dip at a velocity satisfying the condition (2). The general character of the velocity dependence of the gradient force is shown for different detunings in Fig. 3. Besides the contribution of the coherent-dragging effect, the gradient force depends also on the resonance between the Λ atom and the radiation. A curious feature of this interaction scheme is the vanishing of the gradient force when the detunings Ω_{31} and Ω_{32} are equal in absolute value. This effect is due to the mutual cancellation of the atom polarizabilities at adjacent transitions.

The general behavior of the diffusion-tensor components for the case of opposing waves is the same as that of the gradient force (Fig. 4). The asymmetric form of the D_{zz} component is due to the nonadiabatic increment (9).

We note that since there is no coherent population dragging in the V configuration, no anomalies whatever appear in the light-pressure force and in the diffusion tensor. The light-pressure force and the momentum-diffusion tensor are



FIG. 4. Diffusion-tensor component D_{zz} vs velocity in the case of opposing waves at $G = 10^3$ and $\Omega_{32}/\gamma = -270$. The numbers on the curves correspond respectively to detunings $\Omega_{31}/\gamma = 270$, 230, and 130.



FIG. 5. Longitudinal velocity distribution of an atom beam before (1) and after (2) interacting with two opposing light beams.

smooth functions of the atom velocity at any detuning and saturation parameter. In the case of a cascade configuration of the atomic levels, the effect of the coherent dragging of the populations can cause some irregularities of the light pressure and of the diffusion tensor in accordance with (2). These changes, however, are not as radical as for the Λ scheme.^{12,13}

5. CONCLUSION

The foregoing analysis reveals thus the presence of a number of irregularities in the radiation pressure exerted on multilevel atoms. These irregularities must be taken into account in experiments aimed at controlling the motion of atoms. It can be noted, in particular, that for Λ configurations of atomic levels, under conditions when two waves having the same direction are exactly at resonance with the atom, the light pressure vanishes, whereas in the case of a two-level atom the light pressure is a maximum at exact resonance with the atomic transition. Another interesting feature is the vanishing of the light-pressure force acting on a Λ atom in two opposing waves. Of interest is also the vanishing of the gradient force when the Λ atom has a resonant velocity satisfying the condition (2). The last feature can be used to select atoms that have a definite velocity projection v_r . Thus, if an atomic beam propagating along the z axis is irradiated by two opposing light waves, the atoms having a velocity other than (2) will be acted upon by a nonzero gradient force. Under the condition $|\Omega_{32}| < |\Omega_{31}|$ this gradient force pushes the atoms out of the beam. This means that if the interaction time of the Λ atom with the waves is long enough, there will be left in the beam atoms whose velocities are close to the resonant value (Fig. 5) $v_z = (\Omega_{32} - \Omega_{31})/k$. An important feature of this method of selecting atoms by velocity is the possibility of varying the atom velocity. A change of several hertz in the difference between frequencies of the two waves can change the velocity of the selected atoms by an amount equal to the average thermal velocity. The interval in which selection by velocity is possible is determined by the homogeneous broadening of the adjacent atomic transitions. For example, the width of a single-velocity beam of sodium atoms at $\gamma = 10$ MHz and $G \sim 1$ is of the order of $\Delta v = \gamma /$ $k \approx 5 \cdot 10^2$ cm/s. This velocity interval corresponds to an effective single-velocity atomic-beam effective temperature of order 0.1 K.

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