Radiative force for an optically polarized atom

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The radiative force in the field of two counterpropagating waves is examined for an atom with a nonuniform initial Zeeman sublevel distribution. The study is carried out for an atom with a preliminary optical orientation induced by a circularly polarized optical pump wave, and for an atom prealigned by a linearly polarized light wave. Here the frequency of the counterpropagating light waves and that of the optical pump, of either circular or linear polarization, are resonant with adjacent atomic transitions of the Λ level configuration. It is shown that the radiative force for an optically polarized atom possesses vector properties and other quantitative characteristics that are fundamentally new as compared to the previously studied radiative forces for atoms with a uniform initial Zeeman sublevel distribution. The main features of the new radiative force are the absence of spatial oscillations with a half-wavelength period, and the possibility of controlling the signs of the even and odd (in the atomic velocity) parts of a given force. The parameters involved can be chosen such that the new radiative force appears as either a damping or an accelerating force.

In problems of laser cooling of atoms and localizing atoms in electromagnetic fields, radiative forces in the field of one or more light waves have been widely studied in recent years (see Refs. 1–3 and references therein). Particular attention has been paid to the rectified radiative force, which varies smoothly over a wavelength. For a two-level atomic model that is nondegenerate with respect to the projection of the angular momentum, the rectification of the radiative force is achieved either by the interference of two or more light waves of equal frequency[4,5] or in a bichromatic light field.[4,6,7] For this purpose, three-level Λ-configuration atoms with no degeneracy have also been considered, for which the rectified radiative force is obtained by mixing light waves of different frequencies.[8–11] In a special atomic model and $1/2 \rightarrow 1/2$ and $1/2 \rightarrow 3/2$ angular momentum transitions, radiative force rectification was achieved by averaging the force over a half wavelength.[12–15] Inclusion of degeneracy in the above problems leads to considerable difficulties in radiative force calculations, and complicates the physical interpretation of the effects involved.[16–18]

In all previous theoretical and experimental studies, radiative force analyses have been carried out for an atom which, prior to entering the field of one or more light waves, is in a degenerate state uniformly distributed over the projection of the angular momentum (Zeeman sublevels), or alternatively in the zero-momentum ground state. However, it is known that prior optical polarization of the atom, which leads to a nonuniform distribution over the Zeeman sublevels, has a significant influence on the subsequent optical phenomena arising from the atom's interaction with a resonant light field. This has been observed in spontaneous emission,[19] resonant fluorescence,[20] in the photon echo effect,[21] two-pulse free induction,[22] light scattering,[23] and probe pulse spectroscopy.[24] It is natural to assume that a nonuniform initial Zeeman sublevel distribution will change the properties of the radiative force and will influence the methods by which it is rectified. However, these changes are possible only for an atom with nonvanishing momentum in its resonant states, so the inclusion of degeneracy in this problem is necessary in principle.

In the present work, we investigate the radiative force theoretically for an optically polarized atom of arbitrary momentum which, prior to entering the field of two counterpropagating waves, has a nonuniform Zeeman level population due to preliminary optical pumping. To calculate this force we employ an atom with a Λ configuration of levels $E_c < E_a < E_b$, and with the $E_c \rightarrow E_a$ transition forbidden. Owing to the interaction of the atom with the optical pump light wave, which is resonant with the $E_c \rightarrow E_b$ transition, and owing to the spontaneous decay of the excited E_b state, the Zeeman sublevels of the metastable level E_a become nonuniformly populated. This sort of optical pumping, involving the transition of the atom to the metastable level E_a, gives rise to an optical polarization of the atom, with a definite orientation and alignment in the long-lived E_a state. The optically polarized atom subsequently enters the field of two counterpropagating light waves which are resonant with the adjacent $E_a \rightarrow E_b$ transition, and exchanges photons with those light waves, the system having a different symmetry than that studied earlier in Refs. 1–18 for an atom with an initially equiprobable Zeeman sublevel distribution and for an atom in a zero-momentum initial state. In an optically polarized atom moving in the field of counterpropagating light waves, two symmetry types are possible. One corresponds to optical pumping by a circular wave, and the other appears after optical pumping by a linearly polarized wave.

If the optical pumping is by a circular wave with wave vector $\mathbf{k}_0$ and circular polarization $s = \pm 1$, the crucial factor is the preliminary orientation of the atom, characterized by the direction of $\mathbf{k}_0$ and by the right-handed ($s=1$) or left-handed ($s=-1$) rotation of the optical electric field, i.e, by a right- or left-handed screw. For an atom with prior optical

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orientation moving in the field of two counterpropagating circular waves with wave vectors $\mathbf{k}_0$ and $\mathbf{k}_2=-\mathbf{k}_1$ collinear with $\mathbf{k}_0$ and with the same senses of circular polarization $\lambda_1=\lambda_2=\pm 1$, the radiative force contains independent contributions from the counterpropagating waves. Therefore the force does not depend on interference effects, and does not contain spatial oscillations with a period equal to half the wavelength π/k_1. Consequently, the force does not require a rectification procedure, which sets it quite apart from radiative forces previously examined in Refs. 1–18.

For counterpropagating light waves of equal amplitude, certain terms of the calculated force cancel, and the remainder divide the force into two parts, even and odd with respect to the atomic velocity $\mathbf{v}$. The odd part, for a negative offset of the counterpropagating waves from resonance ($\Delta<0$), plays the role of a frictional force. The even part changes sign under the substitution $\lambda_1\rightarrow-\lambda_1$, and also under the substitution $\mathbf{k}_0\rightarrow-\mathbf{k}_0$ for fixed s or under the substitution $s\rightarrow-s$ for fixed $\mathbf{k}_0$. Thus, one can control the signs of the even and odd parts in an experiment. This makes it possible to choose the parameters Δ, λ_1, S, and $\mathbf{k}_0$ such that the radiative force in question plays the role of either a damping or an accelerating force. For $\Delta=0$ the odd part vanishes, whereas the even part is nonvanishing. In particular, for highly collimated atoms with $\Delta \neq 0$ moving with low longitudinal velocities (relative to $\mathbf{k}_1$) such that $|\mathbf{k}_1 v|\ll\gamma_b$, the ratio of the odd part to the even is $|\mathbf{k}_1 v|\gamma_b^{-1}$, where γ_b^{-1} is the relaxation time of the excited state of energy E_b. In this case the radiative force assumes a characteristic form with resonances at $\Delta=\pm\gamma_b/2$.

If the optical pumping is produced by a linearly polarized wave with wave vector $\mathbf{k}_0$ and polarization vector $\mathbf{l}_0$, then the crucial factor is the preliminary alignment of the atom, characterized by the direction of the vectors $\mathbf{k}_0$ and $\mathbf{l}_0$. In this case, the interaction of the atom with linearly polarized counterpropagating waves with wave vectors $\mathbf{k}_1$ and $\mathbf{k}_2=-\mathbf{k}_1$ collinear with $\mathbf{k}_0$, and with polarization vectors $\mathbf{l}_1$ and $\mathbf{l}_2$ forming angles φ_1 and φ_2 with $\mathbf{l}_0$ is of particular interest. Under these conditions, the radiative force at $\Delta=0$ contains independent contributions from the counterpropagating waves, with arbitrary amplitudes R_1 and R_2. It is therefore unaffected by interference and does not contain spatial oscillations with period π/k_1, i.e., it is a rectified force. For $R_1=R_2$ some of the terms of the force cancel; those that remain are due solely to the alignment of the atom and are even functions of the velocity $\mathbf{v}$. This even radiative force (in $\mathbf{v}$) changes sign under the substitution $\mathbf{k}_1\rightarrow-\mathbf{k}_1(\mathbf{k}_2\rightarrow-\mathbf{k}_2)$ if the polarization planes of the counterpropagating waves remain unchanged, and under the substitution $\varphi_1\leftrightarrow\varphi_2$, with constant $\mathbf{k}_1$. Depending on φ_1 and φ_2, the force under study therefore appears as either a damping or accelerating force. Owing to the atomic alignment, one can choose the angles φ_1 and φ_2 such that for $R_1=R_2$ and $\Delta \neq 0$, the radiative force also contains independent contributions from each of the two counterpropagating waves. This is achieved by setting $\varphi_1=0$ and $\varphi_2\pm\pi/2$, or $\varphi_2\pm\pi/2$ and $\varphi_1=0$. In this case the radiative force splits into two parts, even and odd in the velocity $\mathbf{v}$, which exhibit no spatial oscillations with period π/k_1. The angles φ_1 and φ_2 and the offset Δ can be chosen such that a given radiative force plays the role of either a damping or an accelerating force. For low longitudinal velocities (along $\mathbf{k}_1$) such that $|\mathbf{k}_1\mathbf{v}|\ll\gamma_b$, the ratio of the odd part to the even is $|\mathbf{k}_1\mathbf{v}|\gamma_b^{-1}$, and the radiative force has resonances at $\Delta=\pm\gamma_b/2$. In all cases the signs of the odd and even parts are easy to control, which is very important in practical applications.

In the present study we employ an ultrashort optical pump of arbitrary intensity, and the radiative force calculation is carried out in second-order perturbation theory in the (weak) field of counterpropagating light waves over the time interval $0\leq t\ll\tau_p$, where τ_p is the optical pumping time in the counterpropagating waves. This makes it possible to examine the radiative force for arbitrary degeneracy of the resonant levels, and to compare with an initially uniform Zeeman sublevel distribution both in the unsteady regime for $0\leq t\lesssim\gamma_b/2$, and in the steady-state regime at $\gamma_b/2\ll t\ll\tau_p$. It is found that owing to the orientation and alignment of the atom, the absorption of a photon from one light wave and the emission of a photon toward the other occur in a substantially different way compared with an initial equilibrium Zeeman sublevel population. The vector properties and quantitative characteristics of the radiative force in the field of two counterpropagating light waves change so markedly that the force due to radiation pressure on an optically polarized atom may be regarded as a new radiative force. The latter may find application in the confinement of an atom using light waves and in the laser cooling of atoms. This is especially attractive for atoms with a hyperfine level structure, because for these the required Λ configuration of the hyperfine structure components can be easily achieved.

1. OPTICAL PUMPING IN THE Λ LEVEL CONFIGURATION

Consider an atom with no nuclear spin and with a Λ level configuration $E_c<E_a<E_B$, where the $E_c\rightarrow E_a$ transition is forbidden. Prior to the interaction with the external force field, the ground level E_c has a uniform Zeeman sublevel distribution, while the metastable E_a and excited E_b levels are unpopulated. Apart from the energy, the state of the atom is characterized by the quantum numbers J_c, J_a, and J_b of the angular momenta $\mathbf{J}_c$, $\mathbf{J}_a$, and $\mathbf{J}_b$, and by the projections M_c, M_a, and M_b of these momenta on the quantization axis. At some instant of time, the atom reaches the electric field of a traveling light wave

$$\mathbf{E}_0=l_0R_0\exp\{i[\mathbf{k}_0\mathbf{r}-\omega_0t-\alpha_0]\}+\text{c.c.},\tag{1}$$

where $\mathbf{l}_0$ is the unit polarization vector, R_0 the amplitude, $\mathbf{k}_0$ the wave vector, α_0 a constant phase shift, and ω_0 is a frequency close to the transition frequency $\omega_{bc}=(E_b-E_c)\hbar^{-1}$. Compared to the exponential $\exp[i(\mathbf{k}_0\mathbf{r}-\omega_0t)]$, the amplitude R_0 is a slow function of time if one takes into account the retardation of the light, and it is also a slow piecewise continuous function of coordinates in the plane perpendicular to the wave vector $\mathbf{k}_0$.

The state of the atom in the field (1) is described by the quantum-mechanical equations for the components of the density matrix $\rho=\rho(t)$ in the JM representation,

$$\left(\frac{\partial}{\partial t}+\mathbf{v}\nabla+\gamma_b\right)\rho_{M_bM_b'}=\frac{i}{\hbar}(\mathbf{E}_0\mathbf{d}_{M_bM_c}\rho_{M_cM_b'}-\rho_{M_bM_c}\mathbf{E}_0\mathbf{d}_{M_cM_b'}), \quad (2)$$

$$\left(\frac{\partial}{\partial t}+\mathbf{v}\nabla\right)\rho_{M_cM_c'}=\frac{\gamma'(2J_b+1)}{|d_{bc}|^2}\mathbf{d}_{M_cM_b}\rho_{M_bM_b'}\mathbf{d}_{M_b'M_c'}+\frac{i}{\hbar}(\mathbf{E}_0\mathbf{d}_{M_cM_b}\rho_{M_bM_c'}-\rho_{M_cM_b}\mathbf{E}\mathbf{d}_{M_bM_c'}), \quad (3)$$

$$\left(\frac{\partial}{\partial t}+\mathbf{v}\nabla+\frac{\gamma_b}{2}\right)\rho_{M_bM_c}=\frac{i}{\hbar}(\mathbf{E}_0\mathbf{d}_{M_bM_c'}\rho_{M_c'M_c}-\rho_{M_bM_b'}\mathbf{E}_0\mathbf{d}_{M_b'M_c}), \quad (4)$$

where

$$\gamma_b=\gamma+\gamma', \quad \gamma=\frac{4|d_{ba}|^2\omega_{ba}^3}{3\hbar c^3(2J_b+1)},$$

$$\gamma'=\frac{4|d_{bc}|^2\omega_{bc}^3}{3\hbar c^3(2J_b+1)},$$

$\mathbf{v}$ is the velocity of the atom, $\mathbf{d}_{M_bM_c}$ is the matrix element of the electric dipole moment operator $\mathbf{d}$, d_{ba} and d_{bc} are the reduced dipole moments,[25] γ and γ' are the spontaneous emission probabilities of the photons $\hbar\omega_{ba}$ and $\hbar\omega_{bc}$, and c the speed of light *in vacuo*; summation over repeated matrix indices is understood.

Suppose the atom reaches the field (1) at point r_0 at time t_0 and, as time goes on $(t_0\leqslant t)$, moves through the field in a straight line, $\mathbf{r}=\mathbf{r}_0+\mathbf{v}(t-t_0)$. Then in Eqs (2)–(4) we may write

$$\frac{\partial}{\partial t}+\mathbf{v}\nabla=\frac{d}{dt}. \quad (5)$$

The role of the boundary conditions for Eqs. (2)–(4) is played by the density matrix elements,

$$\rho_{M_bM_b'}(t_0)=\rho_{M_bM_c}(t_0)=0, \quad \rho_{M_cM_c'}(t_0)=\delta_{M_cM_c'}/(2J_c+1), \quad (6)$$

which describe the equilibrium state of the atom before entering the field (1).

Equations (2)–(4), with (5) and (6), are solved in the resonance approximation, when the offset $\Delta_0=\omega_0-\omega_{bc}$ is small, $|\Delta_0|\ll\omega_0$. For a weak light field (1), which during the time period $0\leqslant t-t_0$ satisfies the inequality

$$|R_0d_{bc}|^2\gamma_b(t-t_0)\ll 2\hbar^2[(\Delta_0-\mathbf{k}_0\mathbf{v})^2+(\gamma_b/2)^2],$$

the method of successive approximations used in perturbation theory is applicable. In this case the required density matrix quadratic in the field (1) is

$$\rho_{M_bM_b'}(t)=\frac{(\mathbf{l}_0\mathbf{d}_{M_bM_c})(\mathbf{l}_0^*\mathbf{d}_{M_cM_b'})}{\hbar^2(2J_c+1)}\exp[-\gamma_b(t-t_0)]\times\int_0^{t-t_0}d\tau_2R_0^*(\tau_2)\exp\{[\gamma_b-\gamma_{ba}+i(\Delta_0-\mathbf{k}_0\mathbf{v})]\tau_2\}\int_0^{\tau_2}d\tau_1R_0(\tau_1)\exp\{[\gamma_{ba}-i(\Delta_0-\mathbf{k}_0\mathbf{v})]\tau_1\}+\text{h.c.}\,. \quad (7)$$

For a circular wave (1), the dependence of the polarization vector $l_0=l_{k_0s}$ on k_0 and on the parameter s, which defines the direction of rotation of E_0, will be written in explicit form independent of the choice of coordinate system, namely

$$\mathbf{l}_{\mathbf{k}_0s}=2^{-1/2}(s\mathbf{l}_{\mathbf{k}_0}^{(1)}+i\mathbf{l}_{\mathbf{k}_0}^{(2)}), \quad (8)$$

where $s=1$ $(s=-1$ for right-handed (left-handed) circular polarization. The vectors in (8) satisfy the relations

$$\mathbf{k}_0\mathbf{l}_{\mathbf{k}_0}^{(1)}=\mathbf{k}_0\mathbf{l}_{\mathbf{k}_0}^{(2)}=\mathbf{l}_{\mathbf{k}_0}^{(1)}\mathbf{l}_{\mathbf{k}_0}^{(2)}=0, \quad \mathbf{l}_{-\mathbf{k}_0}^{(1)}=\mathbf{l}_{\mathbf{k}_0}^{(1)}, \quad \mathbf{l}_{-\mathbf{k}_0}^{(2)}=-\mathbf{l}_{\mathbf{k}_0}^{(2)},$$

$$[\mathbf{l}_{\mathbf{k}_0}^{(1)}\mathbf{l}_{\mathbf{k}_0}^{(2)}]=\beta\frac{\mathbf{k}_0}{k_0}, \quad \beta=\frac{\mathbf{k}_0}{k_0}[\mathbf{l}_{\mathbf{k}_0}^{(1)}\mathbf{l}_{\mathbf{k}_0}^{(2)}],$$

$$\mathbf{l}_{-\mathbf{k}_0s}=\mathbf{l}_{\mathbf{k}_0s}^*, \quad \mathbf{l}_{\mathbf{k}_0s}\mathbf{l}_{\mathbf{k}_0s'}^*=\delta_{ss'},$$

where β is a unit pseudoscalar. Under the substitution $k_0\to -k_0$, the polarization vector (8) transforms as

$$\mathbf{l}_{\mathbf{k}_0s}\to -\mathbf{l}_{\mathbf{k}_0,-s},$$

which for (1) implies the substitutions $s\to -s$ and $\alpha_0\to\alpha_0+\pi$. Under the inversion, the polarization vector (8) transforms in a different way,

$$\mathbf{l}_{\mathbf{k}_0s}\to\mathbf{l}_{\mathbf{k}_0,-s}, \quad (9)$$

implying the substitution $s\to -s$ for (1).

We now employ a Cartesian coordinate system xyz with basis unit vectors $\mathbf{l}_x,\mathbf{l}_y$, and $\mathbf{l}_z$. We choose the quantization axis (z) to be collinear with $\mathbf{k}_0$ and direct the x axis along the unit vector $\mathbf{l}_{\mathbf{k}_0}^{(1)}$, which enters into the polarization vector (8). The density matrix (7) then becomes diagonal,

$$\rho_{M_bM_b'}(t,s)=\rho_{M_b}(t,s)\delta_{M_bM_b'}, \quad s=\pm 1. \quad (10)$$

Coordinate inversion leads in (7) and (10) to the substitutions (9) and

$$M_b=\mathbf{l}_z\mathbf{J}_b\to -M_b, \quad M_b'\to -M_b'.$$

From this it follows that under inversion, we have $\rho_{-M_b,-M_b'}(t,-s)=\rho_{M_bM_b'}(t,s)$ or

$$\rho_{-M_b}(t,-s)=\rho_{M_b}(t,s). \quad (11)$$

Equations (10) and (11) are a consequence of the symmetry of the interaction of the atom with the circular wave (1), where the quantization axis is collinear with $\mathbf{k}_0$ and the x axis is parallel to $\mathbf{l}_{\mathbf{k}_0}^{(1)}=\mathbf{l}_x$. Therefore, the equations are valid to any order in perturbation theory as well as outside of its range of applicability.

Equations (2)–(4) are simplest to solve nonperturbatively when the interaction time τ of the atom with an arbitrarily intense light wave (1) is extremely short,

$$\gamma_b\tau \ll 1. \tag{12}$$

If an atom with velocity $v_\perp$ crosses a laser beam (1) of diameter D, then the inequality (12) holds for $D\gamma_b \ll |\mathbf{v}_\perp|$, where $v_\perp$ is the component of $\mathbf{v}$ perpendicular to $\mathbf{k}_0$. The inequality (12) also holds for an ultrashort light pulse of duration τ which at t_0 impinges upon an atom located at point $\mathbf{r}_0$. For velocities $v \ll c$, the atomic motion along $\mathbf{k}_0$ may be neglected during the passage of the ultrashort light pulse. In both cases quoted, the amplitude in (1) is some real function of the time, $R_0 = R_0(t-t_0)$.

In order to solve Eqs. (2)–(4) at $t \geq t_0$ for an ultrashort interaction (12), we generalize the calculations of Ref. 21 to a single atom with velocity $\mathbf{v}$. As a result, for some offset $\Delta_0 = \omega_0 - \omega_{bc}$ and velocity $\mathbf{v}$ satisfying

$$|\Delta_0 - \mathbf{k}_0\mathbf{v}|\tau \ll 1, \tag{13}$$

the solution to Eqs. (2)–(4) for a circular wave takes the same form as (10) with a characteristic factor

$$\rho_{M_b}(t,s) = \frac{1}{2J_c+1}[B^{(s)}_{M_b}(t-t_0)]^2 \exp[-\gamma_b(t-t_0)], \tag{14}$$

where

$$B^{(s)}_{M_b}(t-t_0) = \sin\left[\Lambda^{(s)}_{M_b}\int_0^{t-t_0} R_0(\xi)d\xi\right], \tag{15}$$

$$\Lambda^{(s)}_{M_b} = (-1)^{J_b-M_b+s}\begin{pmatrix} J_b & J_c & 1 \\ M_b & s-M_b & -s \end{pmatrix}\frac{|d_{bc}|^2}{\hbar^2}, \tag{16}$$

and the $3j$ symbol $(\begin{smallmatrix} a & b & c \\ d & e & h \end{smallmatrix})$ is defined in Ref. 25. If the field (1) is weak, Eqs. (10) and (14) turn into (7) by virtue of (8) and (12).

If during an ultrashort time interval $0 \leq t-t_0 \leq \tau$ the amplitude R_0 is constant, then for an arbitrary offset $\Delta_0 = \omega_0 - \omega_{bc}$ and any velocity $\mathbf{v}$, the quantity (15) in Eq. (14) should be replaced by

$$B^{(s)}_{M_b}(t-t_0) = \frac{R_0\Lambda^{(s)}_{M_b}}{\Omega^{(s)}_{M_b}}\sin[\Omega^{(s)}_{M_b}(t-t_0)], \tag{17}$$

where

$$\Omega^{(s)}_{M_b} = \left[\frac{1}{4}(\Delta_0 - \mathbf{k}_0\mathbf{v})^2 + R_0^2(\Lambda^{(s)}_{M_b})^2\right]^{1/2}, \tag{18}$$

and after the interaction with the field (1) the quantity (17) for $t_0+\tau \leq t$ maintains a constant value with an argument $t-t_0=\tau$.

If the atom interacts with a linearly polarized wave (1) of arbitrary intensity, we direct the quantization axis ($\bar{z}$ axis) along the real vector $\mathbf{l}_0$. Then in the Cartesian coordinate system [**strut**]$\bar{x}\bar{y}\bar{z}$, of the three contravariant unit vectors $\mathbf{l}^{(-s)}$ $(s=0,\pm 1)$ of the form

$$\mathbf{l}^{(-1)} = 2^{-1/2}(\mathbf{l}_{\bar{x}} + i\mathbf{l}_{\bar{y}}), \quad \mathbf{l}^{(1)} = 2^{-1/2}(-\mathbf{l}_{\bar{x}} + i\mathbf{l}_{\bar{y}}),$$

$$\mathbf{l}^{(0)} = \mathbf{l}_{\bar{z}}, \tag{19}$$

we choose $\mathbf{l}_0 = \mathbf{l}^{(-s)}$ with $s=0$ as the polarization vector. In this case it is convenient to take a Cartesian system with the $\bar{z}$ axis along $\mathbf{l}_0$ and the $\bar{x}$ axis antiparallel to $\mathbf{k}_0$, in which the atomic E_b state is described, according to Eq. (7), by the density matrix

$$\bar{\rho}_{\bar{M}_b\bar{M}'_b}(t,s) = \bar{\rho}_{\bar{M}_b}(t,s)\delta_{\bar{M}_b\bar{M}'_b}, \quad s=0, \tag{20}$$

where the factor preceding the δ function satisfies

$$\bar{\rho}_{-\bar{M}_b}(t,0) = \bar{\rho}_{\bar{M}_b}(t,0). \tag{21}$$

Equations (20) and (21) are a consequence of the symmetry exhibited in the interaction of the atom with a linearly polarized wave when the quantization axis $\bar{z}$ is chosen along the polarization vector $\mathbf{l}_0$ and the $\bar{x}$ axis is antiparallel to $\mathbf{k}_0$. They therefore retain their validity no matter what the method of calculation.

In an ultrashort interaction (12) with a linearly polarized wave (1), the solution of Eqs. (2)–(4) in the coordinate system $\bar{x}\bar{y}\bar{z}$ for $t_0 \leq t$ takes the form (20), in which the characteristic factor $\bar{\rho}_{\bar{M}_b}(t,s)$ with $s=0$ is given by Eqs. (14)–(18) with $M_b \to \bar{M}_b, M'_b \to \bar{M}'_b$, and $s \to 0$.

We now go from the $\bar{x}\bar{y}\bar{z}$ coordinate system to the xyz system discussed earlier, with the quantization axis z collinear with $\mathbf{k}_0$, and with the x axis along the prescribed linear polarization vector $\mathbf{l}_x = \mathbf{l}_0$, by executing three successive rotations through Euler angles α, β, and γ, as discussed in Ref. 25. To this end we first rotate about the $\bar{z}$ axis through an angle $\alpha=0$, to transform to a new coordinate system $x_1y_1z_1$. This is followed by a rotation about the new y_1 axis through an angle $\beta=-\pi/2$, transforming to another new coordinate system, $x_2y_2z_2$. Finally, a rotation about the new axis z_2 through an angle $\gamma=0$ takes us to the desired xyz system. Using the density matrix transformation law under rotations of a Cartesian coordinate systems through Euler angles α, β, and γ (see Ref. 25), we obtain the density matrix $\rho_{M_bM'_b}(t,0)$ in the xyz system, with the quantization axis z collinear with $\mathbf{k}_0$ and the x axis along $\mathbf{l}_0$:

$$\rho_{M_bM'_b}(t,0) = \sum_{\bar{M}_b}\bar{\rho}_{\bar{M}_b}(t-t_0,0)\sum_{\varkappa q}(2\varkappa+1)\,(-1)^{J_b-\bar{M}_b}\begin{pmatrix} J_b & J_b & \varkappa \\ \bar{M}_b & -\bar{M}_b & 0 \end{pmatrix}(-1)^{J_b-M'_b}\begin{pmatrix} J_b & J_b & \varkappa \\ M_b & -M'_b & q \end{pmatrix}d^{\varkappa}_{0q}\left(-\frac{\pi}{2}\right), \tag{22}$$

where the quantity $d^{\varkappa}_{0q}(-\pi/2)$ is a special case of the real factor $d^J_{MM'}(\beta)$ that enters into the Wigner D function

$$\mathsf{D}^J_{MM'}(\alpha,\beta,\gamma) = e^{-iM\alpha}d^J_{MM'}(\beta)e^{-iM'\gamma}.$$

The spontaneous decay of the excited state, which is described by the density matrices (10) and (22), gives rise to a nonuniform population in the Zeeman sublevels of the metastable level E_a. This sort of optical pumping, in which an atom is transferred from the ground state E_c to a long-lived state E_a by a circularly polarized $s=\pm 1$ wave or a linearly polarized $s=0$ wave, is described by a density matrix $\rho_{M_aM_a'}(t,s)$ that satisfies

$$\frac{d}{dt}\rho_{M_aM_a'}(t,s)=\frac{\gamma(2J_b+1)}{|d_{ba}|^2}\mathbf{d}_{M_aM_b}\rho_{M_bM_b'}(t,s)\mathbf{d}_{M_b'M_a'}, \quad (23)$$

where the width of the metastable level is taken to be zero and the initial condition at $t=t_0$ is $\rho_{M_aM_a'}(t_0,s)=0$ $(s=0,\pm 1)$. Here $\rho_{M_bM_b'}(t,s)$ is defined in Eqs. (10) and (22).

Let the optical pump duration be arbitrary and equal to τ. Then at some much later time

$$t-t_0-\tau\ll\gamma_b^{-1}, \quad (24)$$

the density matrix in the excited state E_b relaxes to zero [$\rho_{M_bM_b'}(t,s)=0$], whereas in the metastable level E_a it assumes a constant value $\rho_{M_aM_a'}(t,s)=\rho_{M_bM_b'}(s)$ determined by the solution of Eq. (23):

$$\rho_{M_aM_a'}(s)=\frac{\gamma(2J_b+1)}{\gamma_b|d_{ba}|^2}\left[\gamma_b\int_{t_0}^{t_0+\tau}\mathbf{d}_{M_aM_b}\rho_{M_bM_b'}(\xi,s)\right.$$
$$\left.\times\mathbf{d}_{M_b'M_a'}d\xi+\mathbf{d}_{M_aM_b}\rho_{M_bM_b'}(t_0+\tau,s)\mathbf{d}_{M_b'M_a'}\right], \quad (25)$$

where the first (second) term inside the brackets describes the population of the metastable level E_a during (after) optical pumping.

For an arbitrary duration τ and any amplitude R_0, the density matrix (25) for a circularly polarized optical pump wave is diagonal by virtue of Eq. (10), and in view of Eq. (11) it satisfies

$$\rho_{-M_a,-M_a'}(-s)=\rho_{M_aM_a'}(s), \quad s=\pm 1. \quad (26)$$

The optical polarization of an atom in a long-lived state of energy E_a is characterized by multipole moments $\rho_q^{(\varkappa)}$ $\times(J_a,s)$ which enter into the series expansion of the density matrix (25) in $3j$ symbols,

$$\rho_{M_aM_a'}(s)=(-1)^{J_a-M_a'}$$
$$\times\sum_{\varkappa q}\frac{2\varkappa+1}{\sqrt{2J_a+1}}\begin{pmatrix}J_a & J_a & \varkappa\\ M_a & -M_a' & q\end{pmatrix}\rho_q^{(\varkappa)}(J_a,s), \quad (27)$$

where $0\leqslant\varkappa\leqslant 2J_a$ and $-\varkappa\leqslant q\leqslant\varkappa$. Here the multipole moments of order $\varkappa=0,1,2$ describe the atomic population, orientation, and alignment, respectively.

Owing to the symmetry taken into account in Eqs. (10), (11), and (26) we obtain for a circularly polarized optical pump wave

$$(-1)^{\varkappa}\rho_{-q}^{\varkappa}(J_a,-s)=\rho_q^{(\varkappa)}(J_a,s), \quad s=\pm 1, q=0. \quad (28)$$

In optical pumping by a linearly polarized wave, from the symmetry taken into account in Eqs. (20)–(22) it is found that the multipole moments in (27) with $s=0$ are of order $\varkappa=2n$ $(n=0,1,2,...)$ and that $0\leqslant|q|$.

To calculate (25) to second order in a weak field (1), Eq. (7) is useful. If the circularly polarized optical pump wave is ultrashort (see Eq. (12)) and arbitrarily intense, then in order to calculate the density matrix (25) and the multipole moments in (27), one should employ Eqs. (10) and (14)–(18) with $s=\pm 1$. For a linearly polarized optical pump wave, Eqs. (22) are needed.

As a result, for ultrashort pumping by a circularly polarized optical pump wave (1) with $s=\pm 1$, we obtain

$$\rho_q^{(\varkappa)}(J_a,s)=(-1)^{J_a+J_b+\varkappa+1}\frac{\gamma(2J_a+1)^{1/2}(2J_b+1)}{\gamma_b(2J_c+1)}$$
$$\times\begin{Bmatrix}J_a & \varkappa & J_a\\ J_b & 1 & J_b\end{Bmatrix}$$
$$\times\sum_{M_b}(-1)^{J_b-M_b}\begin{pmatrix}J_b & J_b & \varkappa\\ M_b & -M_b & 0\end{pmatrix}$$
$$\times\left\{[B_{M_b}^{(s)}(\tau)]^2+\gamma_b\int_0^{\tau}[B_{M_b}^{(s)}(\xi)]^2d\xi\right\}\delta_{0q}, \quad (29)$$

where the $6j$ symbol $\{a\ b\ cd\ e\ h\}$ is defined in Ref. 25.

In ultrashort optical pumping by a linearly polarized wave (1) with $s=0$, we find

$$\rho_q^{(\varkappa)}(J_a,0)=(-1)^{J_a+J_b+1}\frac{\gamma(2J_a+1)^{1/2}(2J_b+1)}{\gamma_b(2J_c+1)}$$
$$\times\begin{Bmatrix}J_a & \varkappa & J_a\\ J_b & 1 & J_b\end{Bmatrix}$$
$$\times\sum_{M_b}(-1)^{J_b-M_b}\begin{pmatrix}J_b & J_b & \varkappa\\ M_b & -M_b & 0\end{pmatrix}$$
$$\times\left\{[B_{M_b}^{(0)}(\tau)]^2+\gamma_b\int_0^{\tau}[B_{M_b}^{(0)}(\xi)]^2d\xi\right\}d_{0q}^{(2)}$$
$$\times\left(-\frac{\pi}{2}\right), \quad (30)$$

where $\varkappa=2n(n=0,1,2,...)$.

From the solution of Eqs. (2)–(4) and (23) for an arbitrary instant of time $t_0+\tau\leqslant t$, after optical pumping of any duration τ, it follows that the level population probabilities W_b, W_c, and W_a will be determined by the spontaneous decay of the excited state E_b and

$$W_b(t-t_0)=W_b(\tau)\exp[-\gamma_b(t-t_0)], \quad (31)$$

$$W_c(t-t_0)=W_c(\tau)+\frac{\gamma'}{\gamma_b}W_b(\tau)\{1-\exp[-\gamma_b(t-t_0)]\}, \quad (32)$$

$$W_a(t-t_0)=W_a(\tau)+\frac{\gamma}{\gamma_b}W_b(\tau)\{1-\exp[-\gamma_b(t-t_0)]\}, \quad (33)$$

$$W_b(t-t_0)+W_c(t-t_0)+W_a(t-t_0)=W_b(\tau)+W_c(\tau)$$
$$+W_a(\tau)=1, \quad (34)$$

where $W_b(\tau), W_c(\tau)$, and $W_a(\tau)$ are the population probabilities of the levels E_b, E_c, and E_a at time $t=t_0+\tau$, when the optical pump is turned off. For sufficiently large τ in Eqs. (31)–(34) we must write

$$W_b(\tau)=W_c(\tau)=0, \quad W_a(\tau)=1.$$

For an ultrashort optical pump, the probability $W_b(\tau)$ can be calculated using Eqs. (10) and (22), and for $W_c(\tau)$ a similar argument applies. The probability $W_a(\tau)$ is determined by using the first terms in brackets in Eq. (25).

2. RADIATIVE FORCE FOR AN OPTICALLY POLARIZED ATOM

Consider two collinear laser beams. The electric field of one is defined in Eq. (1). The second is a superposition of two counter- or copropagating traveling light waves

$$\mathbf{E}=[\mathbf{l}_1 R_1 \exp(i\phi_1)+\mathbf{l}_2 R_2 \exp(i\phi_2)]\exp(-i\omega t)+\text{c.c.}, \quad (35)$$

where

$$\phi_1=\mathbf{k}_1\mathbf{r}-\alpha_1, \quad \phi_2=\mathbf{k}_2\mathbf{r}-\alpha_2,$$

$\mathbf{l}_1$ and $\mathbf{l}_2$ are unit polarization vectors, R_1 and R_2 are constant real amplitudes, α_1 and α_2 are constant phase shifts, and the frequencies ω_1 and ω_2 are equal, $\omega_1=\omega_2=\omega$, and close to the frequency $\omega_{ba}=(E_b-E_a)\hbar^{-1}$ for the transition from the metastable level E_a to the excited level E_b. The wave vectors $\mathbf{k}_0, \mathbf{k}_1$, and $\mathbf{k}_2$ are collinear, and the atomic velocity $\mathbf{v}$ and wave vectors $\mathbf{k}_0, \mathbf{k}_1$, and $\mathbf{k}_2$ all lie in the same plane.

The atom moves in the plane of the beams in such a way that its transverse velocity $\mathbf{v}_\perp$ is directed from the first beam, which is responsible for the optical pumping, to the second one, which exerts on the atom the radiative force we wish to determine. The beam separation l and the transverse atomic velocity are chosen such that the time $l/v_\perp$ the atom spends between the beams obeys the inequality (24). This requires that $v_\perp \ll l\gamma_b$.

An optically polarized atom with a nonuniform Zeeman sublevel population (25), with $s=0,\pm 1$, enters the field (35) at point $\mathbf{r}_0'=0$ at time $t_0'=0$, and for $0\leqslant t$ it moves through the field in a straight line, $\mathbf{r}=\mathbf{v}t$. The interaction of the atom and the field (35) in the Cartesian system xyz with quantization axis z collinear with $\mathbf{k}_0$ is described by a density matrix $\rho=\rho(t,s)$ in the JM representation, which by virtue of (5) satisfies the equations

$$\left(\frac{d}{dt}+i\omega_{ba}+\frac{\gamma_B}{2}\right)\rho_{M_bM_a}=\frac{i}{\hbar}(\mathbf{Ed}_{M_bM_a'}\rho_{M_a'M_a}-\rho_{M_bM_b'}\mathbf{Ed}_{M_b'M_a}), \quad (36)$$

$$\left(\frac{d}{dt}+\gamma_b\right)\rho_{M_bM_b'}=\frac{i}{\hbar}(\mathbf{Ed}_{M_bM_a}\rho_{M_aM_b'}-\rho_{M_bM_a}\mathbf{Ed}_{M_aM_b'}), \quad (37)$$

$$\frac{d}{dt}\rho_{M_aM_a'}=\frac{\gamma(2J_b+1)}{|d_{ba}|^2}\mathbf{d}_{M_aM_b}\rho_{M_bM_b'}\mathbf{d}_{M_b'M_a'}+\frac{i}{\hbar}(\mathbf{Ed}_{M_aM_b}\rho_{M_bM_a'}-\rho_{M_aM_b}\mathbf{Ed}_{M_bM_a'}), \quad (38)$$

with initial conditions at $t=0$ in the form

$$\rho_{M_bM_a}(0,s)=\rho_{M_bM_b'}(0,s)=0, \quad \rho_{M_aM_a'}(0,s)=\rho_{M_aM_a'}(s), \quad (39)$$

where the density matrix $\rho_{M_aM_a'}(s)$ is defined for the general case in Eq. (25).

We use the familiar formula for the radiative force (see, e.g., Refs. 1–3, 16, 18)

$$\mathbf{F}=Sp[\rho\nabla(\mathbf{dE})] \quad (40)$$

and calculate perturbatively to second order in the weak field (35). We drop terms in (40) with double the frequency ω, which is equivalent to averaging over time from t to $t+2\pi/\omega$.

In solving Eqs. (36)–(38) by perturbation theory, the small parameter in the series expansion of $\rho=\rho(t,s)$ in the field (35) is the greater of the two parameters

$$\frac{R_n^2|d_{ba}|^2\gamma_b t}{2\hbar^2[(\Delta-k_nv)^2+(\gamma_b/2)^2]}\ll 1, \quad n=1,2, \quad (41)$$

where $\Delta=\omega-\omega_{ba}$. In Eq. (41) we have omitted an unimportant factor preceding t, which depends on J_a and J_b and does not exceed unity.

As a result, the desired density matrix at $t\geqslant 0$ and bounded at large t by (41) takes the form

$$\rho_{M_bM_a}(t,s)=\frac{2i}{\hbar\gamma_b}\mathbf{Ad}_{M_bM_a'}\rho_{M_a'M_a}(s)\exp(-i\omega t), \quad s=0,\pm 1, \quad (42)$$

where

$$\mathbf{A}=\mathbf{l}_1R_1I(\mathbf{k}_1)\exp(i\phi_1)+\mathbf{l}_2R_2I(\mathbf{k}_2)\exp(i\phi_2), \quad (43)$$

$$I(\mathbf{k}_n)=\frac{\gamma_b[\gamma_b/2+i(\Delta-\mathbf{k}_n\mathbf{v})]}{2[(\Delta-\mathbf{k}_n\mathbf{v})^2+(\gamma_b/2)^2]}\times\left\{1-\exp\left[-\left(\frac{\gamma_b}{2}-i(\Delta-\mathbf{k}_n\mathbf{v})\right)t\right]\right\}, \quad n=1,2. \quad (44)$$

It is clear that for a weak field

$$R_n^2|d_{ba}|^2\ll\hbar^2[(\Delta-\mathbf{k}_n\mathbf{v})^2+(\gamma_b/2)^2]$$

the density matrix (42) may be used both in the unsteady regime $t\lesssim\gamma_b/2$, and in the steady-state regime $\gamma_b/2\ll t$, which sets in in the region (41) after a long period of time compared with the relaxation time $2/\gamma_b$. For any t in the region (41), the radiative force (40) can be written as

$$F=\frac{2k_1|d_{ba}|^2}{\hbar\gamma_b}(Q+\text{c.c.}), \quad (45)$$

where

$$Q=\frac{1}{|d_{ba}|^2}(\mathbf{Ad}_{M_bM_a'})\rho_{M_a'M_a}(s)(\mathbf{Bd}_{M_aM_b}), \quad (46)$$

$$\mathbf{B}=\mathbf{l}_1^*R_1\exp(-i\phi_1)-\mathbf{l}_2^*R_2\exp(-i\phi_2). \quad (47)$$

The vectors $\mathbf{A}$ and $\mathbf{B}$ are a convenient notation for counter- or copropagating light waves (35) of arbitrary amplitude and polarization. We can expand these vectors in terms of the contravariant unit vectors (19) defined in the Cartesian coordinate system xyz,

$$\mathbf{A}=\sum_{q_1} A_{q_1}\mathbf{l}^{q_1},\quad \mathbf{B}=\sum_{q_2} B_{q_2}\mathbf{l}^{q_2}.$$

Then the quantity (46), after summing over the matrix indices, using (27) and the $3j$ sum rules,[25], takes the form

$$Q=\frac{(-1)^{J_a+J_b}}{\sqrt{2J_a+1}}\sum_{\varkappa q_1 q_2}(2\varkappa+1)A_{q_1}B_{q_2}\rho^{(\varkappa)}_{-q_1-q_2}(J_a,s)\times\begin{pmatrix}1 & 1 & \varkappa\\ q_2 & q_1 & -q_1-q_2\end{pmatrix}\begin{Bmatrix}1 & 1 & \varkappa\\ J_a & J_a & J_b\end{Bmatrix}, \tag{48}$$

where the summation indices q_1 and q_2 take on the values ±1 because the vectors (43) and (47) are orthogonal to the quantization axis z.

The force expression (45), with (48), remains valid for a long time (24) after passage of an optical pump wave (1) of arbitrary duration τ, which may have either circular ($s=\pm1$) or linear ($s=0$) polarization. In an ultrashort optical pump wave (12), calculation of the multipole moments in (48) requires that we make use of Eqs. (29) and (30).

3. RADIATIVE FORCE FOR AN ATOM WITH CIRCULAR OPTICAL POLARIZATION

We next consider an atom whose optical polarization is due to a circularly polarized wave (1) with $s=\pm1$. According to Eq. (28), the lower index of the multipole moment in (48) is zero in this case, and the sum (48) becomes

$$Q=\frac{1}{2}\{(\mathbf{AB})[G(1,s)+G(-1,s)]+i[\mathbf{AB}]_z[G(1,s)-G(-1,s)]\}, \tag{49}$$

where

$$G(q,s)=(-1)^{J_a+J_b+1}\sum_{\varkappa}\frac{2\varkappa+1}{\sqrt{2J_a+1}}\begin{pmatrix}1 & 1 & \varkappa\\ q & -q & 0\end{pmatrix}\times\begin{Bmatrix}1 & 1 & \varkappa\\ J_a & J_a & J_b\end{Bmatrix}\rho^{(\varkappa)}_0(J_a,s),\quad q=\pm1,\quad s=\pm1. \tag{50}$$

Because of the symmetry property (28), the sum $G(1,s)+G(-1,s)$ in Eq. (49) is an even function of s, whereas the difference $G(1,s)-G(-1,s)$ is odd. This means that under inversion and the substitutions $\mathbf{k}_0\rightarrow-\mathbf{k}_0$ or $s\rightarrow-s$, the sum $G(1,s)+G(-1,s)$ is unchanged, whereas the difference $G(1,s)-G(-1,s)$ transforms according to

$$G(1,s)-G(-1,s)=\frac{\mathbf{l}_z\mathbf{k}_0}{k_0}s\beta G(1,1),\quad \beta=[\mathbf{l}_x\mathbf{l}_y]\mathbf{l}_z,$$

where the unit vector $\mathbf{l}_z$ characterizes the direction of the quantization axis.

As a result, the radiative force (45) at $t\geqslant0$ for an atom with a circular polarization (25) and $s=\pm1$ takes the form

$$\mathbf{F}=\frac{2\mathbf{k}_1|d_{ba}|^2}{\hbar\gamma_b}\left[(D^{(0)}_1+D^{(2)}_1)(\mathbf{AB})+iD^{(1)}_1\frac{(\mathbf{l}_z\mathbf{k}_0)}{k_0}s\beta[\mathbf{AB}]_z\right]+\text{c.c.}, \tag{51}$$

where the factors $d^{(\varkappa)}_1$ ($\varkappa=0,1,2$) are special cases of the general expression

$$D^{(\varkappa)}_s=(-1)^{J_a+J_b+1}\frac{2\varkappa+1}{\sqrt{2J_a+1}}\begin{pmatrix}1 & 1 & \varkappa\\ 1 & -1 & 0\end{pmatrix}\times\begin{Bmatrix}1 & 1 & \varkappa\\ J_a & J_a & J_b\end{Bmatrix}\rho^{(\varkappa)}_0\times(J_a,s),\quad \varkappa=0,1,2,\quad s=0,\pm1. \tag{52}$$

Consider first the force (51) for counterpropagating circularly polarized waves (35) with $\mathbf{k}_1=-\mathbf{k}_2$ and polarization vectors $\mathbf{l}_n=\mathbf{l}_{\mathbf{k}_n\lambda_n}$ of the form

$$\mathbf{l}_{\mathbf{k}_n\lambda_n}=2^{-1/2}(\lambda_n\mathbf{l}_x+i\sigma_n\mathbf{l}_y),\quad n=1,2, \tag{53}$$

where the z axis is collinear with k_n, and where

$$\sigma_n=(\mathbf{l}_z\mathbf{k}_n)/k_n;$$

$\lambda_n=1$ for right-handed and $\lambda_n=-1$ for left-handed polarization. The unit vector $\mathbf{l}_x$ coincides with the unit vector $\mathbf{l}^{(1)}_{\mathbf{k}_0}$ of Eq. (8).

For the same sense of circular polarization ($\lambda_1=\lambda_2=\pm1$) of the counterpropagating waves (35), the force (51) is

$$\mathbf{F}=\frac{4|d_{ba}|^2}{\hbar\gamma_b}\sum_{n=1}^{2}\mathbf{k}_n\times\left(D^{(0)}_1+D^{(2)}_1+\frac{(\mathbf{k}_n\mathbf{k}_0)}{k_nk_0}\lambda_n sD^{(1)}_1\right)R^2_nI'(\mathbf{k}_n), \tag{54}$$

where for the quantity (44) we have introduced the notation

$$I(\mathbf{k}_n)=I'(\mathbf{k}_n)+iI''(\mathbf{k}_n).$$

Here $I'(\mathbf{k}_n)$ and $I''(\mathbf{k}_n)$ are the real and imaginary parts of the quantity (44):

$$I'(\mathbf{k}_n)=\frac{\gamma_b/2}{(\Delta-\mathbf{k}_n\mathbf{v})^2+(\gamma_b/2)^2}\left\{\frac{\gamma_b}{2}\left[1-\exp\left(-\frac{\gamma_b t}{2}\right)\cos[(\Delta-\mathbf{k}_n\mathbf{v})t]\right]+(\Delta-\mathbf{k}_n\mathbf{v})\exp\left(-\frac{\gamma_b t}{2}\right)\sin[(\Delta-\mathbf{k}_n\mathbf{v})t]\right\},$$

$$I''(\mathbf{k}_n)=\frac{\gamma_b/2}{(\Delta-k_nv)^2+(\gamma_b/2)^2}\left\{(\Delta-\mathbf{k}_n\mathbf{v})\left[1-\exp\left(-\frac{\gamma_b t}{2}\right)\cos[(\Delta-\mathbf{k}_n\mathbf{v})t]\right]-\frac{\gamma_b}{2}\exp\left(-\frac{\gamma_b t}{2}\right)\sin[(\Delta-\mathbf{k}_n\mathbf{v})t]\right\}.$$

The force (54) contains only independent contributions from either of the counterpropagating waves (35), and no interference terms with spatially oscillating factors $\sin(2\mathbf{k}_1\mathbf{r})$ or $\cos(2\mathbf{k}_1\mathbf{r})$. The absence of these rapid spatial oscillations, with a period equal to the half wavelength π/k_1, is a basic feature of the radiative force (54) in the field of two counterpropagating circular waves (35) with the same sense of polarization, $\lambda_1=\lambda_2=\pm 1$.

The terms involving $D_1^{(1)}$ and $D_1^{(2)}$ in (54) are proportional to multipole moments of order $\varkappa=1$ and $\varkappa=2$ describing the orientation and alignment of the atom. However, of fundamental importance here is the atomic orientation induced by the circularly polarized optical pump wave (1).

The terms in $D_1^{(0)}$ and $D_1^{(2)}$ in (54) are always directed along $\mathbf{k}_n$. In contrast, the term involving $D_1^{(1)}>0$ is parallel to $\mathbf{k}_n$ only when $\mathbf{k}_n/k_n$ and λ_n are both either equal or unequal to the specified quantities $\mathbf{k}_0/k_0$ and s. This has to do with the fact that the atomic orientation induced by optical pumping is invariant under the simultaneous substitutions $\mathbf{k}_0\to-\mathbf{k}_0$ and $s\to-s$. This means that the atomic orientation can be characterized both by the direction of the wave vector $\mathbf{k}_0$ and by the sense of rotation of the electrical field of the optical pump wave, that is, by its left- or right-handedness. If only $\mathbf{k}_n/k_n$ and $\mathbf{k}_0/k_0$ or λ_n and s coincide, then the term involving $D_1^{(1)}>0$ $(D_1^{(1)}<0)$ will be antiparallel (parallel) to $\mathbf{k}_n$.

Since $D_1^{(0)}+D_1^{(2)}>|D_1^{(1)}|$, the radiative force of the nth light wave will be directed along $\mathbf{k}_n (n=1,2)$. However, for counterpropagating waves (35) with $R_1=R_2=R$, terms containing the sum $D_1^{(0)}+D_1^{(2)}$ cancel, and the direction of the force (54) depends heavily on the prior optical orientation of the atom,

$$\mathbf{F}=\frac{4\mathbf{k}_1\hbar f^2}{\gamma_b}\left\{(D_1^{(0)}+D_1^{(2)})[I'(\mathbf{k}_1)-I'(-\mathbf{k}_1)]+\frac{(\mathbf{k}_1\mathbf{k}_0)}{k_1k_0}\lambda_1 sD_1^{(1)}[I'(\mathbf{k}_1)+I'(-\mathbf{k}_1)]\right\}, \quad (55)$$

where $f=|d_{ba}|R\hbar^{-1}$ is the Rabi frequency and the sum $I'(\mathbf{k}_1)+I'(-\mathbf{k}_1)$ and difference $I'(\mathbf{k}_1)-I'(-\mathbf{k}_1)$ are respectively even and odd functions of the velocity $\mathbf{v}$.

According to Eq. (41), Eq. (55) holds for

$$0\leqslant t\ll\tau_p,$$

where τ_p is the optical pumping time in the counterpropagating light waves (35),

$$\tau_p=\frac{2[(\Delta-\mathbf{k}_1\mathbf{v})^2+(\gamma_b/2)^2]}{f^2\gamma_b}.$$

For low-intensity light waves (35), there exists a long time interval

$$2/\gamma_b\ll t\ll\tau_p, \quad (56)$$

over which the steady-state regime is established, and the force (55) takes the form

$$\mathbf{F}=\frac{\mathbf{F}_0}{\gamma_b^2}\left\{(D_1^{(0)}+D_1^{(2)})\Delta(k_1v)+\frac{(k_1k_0)}{k_1k_0}\lambda_1 sD_1^{(1)}\times\left[\Delta^2+(k_1v)^2+\frac{\gamma_b^2}{4}\right]\right\}, \quad (57)$$

where the vector $\mathbf{F}_0$ is an even function of the velocity v,

$$\mathbf{F}_0=\frac{4\mathbf{k}_1\hbar f^2\gamma_b^3}{[(\Delta+\mathbf{k}_1\mathbf{v})^2+(\gamma_b/2)^2][(\Delta-\mathbf{k}_1\mathbf{v})^2+(\gamma_b/2)^2]}. \quad (58)$$

The force (57) is unchanged by the substitution $\mathbf{k}_1\to-\mathbf{k}_1$ with fixed λ_1. Therefore with a proper choice of $\Delta,\mathbf{k}_0,s$, and $D_1^{(1)}$, it can be either a damping or an accelerating force. The odd (in $\mathbf{v}$) part of the force (57) plays the role of a frictional force for $\Delta<0$. The even (in $\mathbf{v}$) part of the force (57) changes sign under the substitution $\lambda_1\to-\lambda_1$ and under the substitution $\mathbf{k}_0\to-\mathbf{k}_0$ with the index held s fixed, and under $s\to-s$ with $\mathbf{k}_0$ held fixed. From the transformation law for the polarization vector (8) under such substitutions, it follows that the even part of the force (57) changes sign if the right-handed circular polarization in the optical pump wave (1) is replaced by a left-handed polarization or *vice versa*. Hence, one can control the signs of the even and odd (in $\mathbf{v}$) parts of the force (57) separately during an experiment.

When the counterpropagating waves (35) with $R_1=R_2=R$ have different senses of circular polarization $(\lambda_1=-\lambda_2=\pm 1)$, instead of (55) we obtain

$$\mathbf{F}=\frac{4\mathbf{k}_1\hbar f^2}{\gamma_b}\left(D_1^{(0)}+D_1^{(2)}+\frac{(\mathbf{k}_1\mathbf{k}_0)}{k_1k_0}\lambda_1 sD_1^{(1)}\right)\times\{[I'(\mathbf{k}_1)-I'(-\mathbf{k}_1)](1+\cos\phi)-[I''(\mathbf{k}_1)+I''(-\mathbf{k}_1)]\sin\phi\}, \quad (59)$$

where $\phi=2\mathbf{k}_1\mathbf{r}-\alpha_1+\alpha_2$, and the sum $I''(\mathbf{k}_1)+I''(-\mathbf{k}_1)$ is an odd function of the velocity $\mathbf{v}$. In the steady state (56), the force (59) takes the form

$$\mathbf{F}=\frac{\mathbf{F}_0\Delta}{\gamma_b^3}\left(D_1^{(0)}+D_1^{(2)}+\frac{(\mathbf{k}_1\mathbf{k}_0)}{k_1k_0}\lambda_1 sD_1^{(1)}\right)\times\left\{\gamma_b(\mathbf{k}_1\mathbf{v})(1+\cos\phi)-\left[\Delta^2-(\mathbf{k}_1\mathbf{v})^2+\frac{\gamma_b^2}{4}\right]\sin\phi\right\}. \quad (60)$$

The vector (58), parallel to $\mathbf{k}_1$, has been introduced to simplify Eqs. (57) and (60). Recalling the equality $\mathbf{k}_1(\mathbf{k}_1\mathbf{k}_0)=\mathbf{k}_0\mathbf{k}_1^2$ for collinear vectors $\mathbf{k}_1$ and $\mathbf{k}_0$, it is found that those terms in the forces (55), (57), (59), and (60) involving a factor $D_1^{(1)}$ are parallel or antiparallel to $\mathbf{k}_0$, depending on the signs of the parameters λ_1, s, and D_1^1.

If the counterpropagating waves (35) are linearly polarized, their polarization vectors will be

$$\mathbf{l}_n=\mathbf{l}_x\cos\varphi_n+\sigma_1\mathbf{l}_y\sin\varphi_n, \quad n=1,2, \quad (61)$$

where positive angles φ_n are reckoned clockwise from the x axis (looking along $\mathbf{l}_z$), regardless of the direction of $\mathbf{k}_1(\mathbf{k}_2=-\mathbf{k}_1)$.

The radiative force for an atom with circular optical polarization (51) moving through a field of counterpropagating linearly polarized waves (35) with $R_1=R_2=R$ is

$$\mathbf{F}=\frac{4\mathbf{k}_1\hbar f^2}{\gamma_b}\left\{\left[(D_1^{(0)}+D_1^{(2)})(1-\cos\varphi\cos\phi)-\frac{(\mathbf{l}_z\mathbf{k}_0)}{k_0}sD_1^{(1)}\sin\varphi\cos\phi\right][I'(\mathbf{k}_1)-I'(-\mathbf{k}_1)]+\left[(D_1^{(0)}+D_1^{(2)})\cos\varphi\sin\phi-\frac{(\mathbf{l}_z\mathbf{k}_0)}{k_0}sD_1^{(1)}\sin\varphi\cos\phi\right][I''(k_1)+I''(-k_1)]\right\}, \quad (62)$$

where

$$\varphi=\sigma_1\varphi_1+\sigma_2\varphi_2.$$

Here φ is the angle between the polarization planes of the counterpropagating waves (35). In the steady-state regime (56), the force (62) becomes

$$\mathbf{F}=\frac{\mathbf{F}_0\Delta}{\gamma_b^3}\left\{\left[(D_1^{(0)}+D_1^{(2)})(1-\cos\varphi\cos\phi)-\frac{(\mathbf{l}_z\mathbf{k}_0)}{k_0}sD_1^{(1)}\sin\varphi\sin\phi\right]\gamma_b(\mathbf{k}_1\mathbf{v})+\left[(D_1^{(0)}+D_1^{(2)})\cos\varphi\sin\phi-\frac{(\mathbf{l}_z\mathbf{k}_0}{k_0}sD_1^{(1)}\sin\varphi\cos\phi\right]\times\left[\Delta^2-(\mathbf{k}_1\mathbf{v})^2+\frac{\gamma_b^2}{4}\right]\right\}. \quad (63)$$

As functions of the coordinates, the forces (59), (60), (62), and (63) undergo spatial oscillations with a period of half the wavelength, $\pi/\mathbf{k}_1$, whereas the forces (55) and (57) do not exhibit such oscillations. Moreover, all of these forces contain terms that are even or odd in the velocity $\mathbf{v}$. At resonance ($\Delta=0$), all of the odd terms vanish, as do the forces (59), (60), (62), and (63).

For highly collimated atoms moving with low longitudinal velocities v_z (relative to $\mathbf{k}_1$),

$$|\mathbf{k}_1\mathbf{v}|=k_1|v_z|\ll\gamma_b, \quad (64)$$

the odd (in $\mathbf{v}$) terms in the forces are small compared to the even terms in proportion $|\mathbf{k}_1\mathbf{v}|\gamma_b^{-1}$. For low longitudinal velocities (64), the most interesting force, Eq. (57), assumes the characteristic form

$$\mathbf{F}=\frac{4\hbar f^2\gamma_b}{\Delta^2+(\gamma_b/2)^2}\left[(D_1^{(0)}+D_1^{(2)})\frac{\mathbf{k}_1\Delta(\mathbf{k}_1\mathbf{v})}{\Delta^2+(\gamma_b/2)^2}+\frac{\mathbf{k}_0k_1}{k_0}\lambda_1 sD_1^{(1)}\right]. \quad (65)$$

Clearly, for $\Delta=0$ the force (65) is nonzero, whereas the forces (60) and (63) vanish. Under the resonance conditions $\Delta=\pm\gamma_b/2$, the absolute value of the ratio of the first and second terms in brackets in (65) is

$$\left|\frac{(D_1^{(0)}+D_1^{(2)})k_1v_z}{D_1^{(1)}\gamma_b}\right|.$$

Therefore for sufficiently low velocities v_z, the main contribution to the force (65) derives from the prior optical orientation of the atom. The quantities $\Delta,\mathbf{k}_0,\lambda_1,s$, and $D_1^{(1)}$ may be chosen such that all terms in (65) play the role of either damping or accelerating forces.

To obtain an order-of-magnitude estimate of the forces (57), (60), (63), and (65), consider the factors $D_1^{(\varkappa)},\varkappa=0,1,2$ for an ultrashort optical pump (12). Making use of Eqs. (52) and (29) we obtain

$$D_1^{(\varkappa)}=\frac{\gamma(2\varkappa+1)(2J_b+1)}{\gamma_b(2J_c+1)}\begin{pmatrix}1 & \varkappa & 1\\ 1 & 0 & -1\end{pmatrix}\times\begin{Bmatrix}1 & 1 & \varkappa\\ J_a & J_a & J_b\end{Bmatrix}\begin{Bmatrix}J_a & \varkappa & J_a\\ J_b & 1 & J_b\end{Bmatrix}\times\sum_{M_b}(-1)^{J_b-M_b}\begin{pmatrix}J_b & J_b & \varkappa\\ M_b & -M_b & 0\end{pmatrix}\times\left\{[B_{M_b}^{(1)}(\tau)]^2+\gamma_b\int_0^\tau[B_{M_b}^{(1)}(\xi)]^2d\xi\right\}. \quad (66)$$

After simplifying the mathematics, the area θ of an ultrashort optical pump pulse becomes

$$\theta=\frac{|d_{bc}|}{\hbar}\int_0^\tau R_0(\xi)d\xi. \quad (67)$$

For $J_c=0$ and $s=1$, according to (16) we have $J_b=M_b=1$. Therefore, for a large area θ such that

$$\frac{1}{\sqrt{3}}\theta=\frac{n\pi}{2}, \quad n=1,2,\ldots, \quad (68)$$

from Eq. (66) for atomic transitions between states with momenta $J_c=0$ and $J_a=J_b=1$ we find

$$D_1^{(0)}=\gamma/9\gamma_b, \quad D_1^{(1)}=-\gamma/24\gamma_b, \quad D_1^{(2)}=\gamma/72\gamma_b.$$

For $J_c=1/2,s=1$ and a large area θ satisfying

$$\begin{pmatrix}J_b & 1/2 & 1\\ 1/2 & 1/2 & -1\end{pmatrix}\theta=\frac{n\pi}{2}, \quad n=\pm1,\pm2,\ldots$$

with the help of Eq. (66) we obtain

$$D_1^{(\varkappa)}=\frac{\gamma(2\varkappa+1)(2J_b+1)}{2\gamma_b}\begin{pmatrix}1 & \varkappa & 1\\ 1 & 0 & -1\end{pmatrix}(-1)^{J_b-1/2}\times\begin{pmatrix}J_b & J_b & \varkappa\\ 1/2 & -1/2 & 0\end{pmatrix}\begin{Bmatrix}1 & 1 & \varkappa\\ J_a & J_a & J_b\end{Bmatrix}\begin{Bmatrix}J_a & \varkappa & J_a\\ J_b & 1 & J_b\end{Bmatrix}, \quad (69)$$

$$J_a=1/2,3/2, \quad J_b=1/2,3/2.$$

In particular, for $J_c=J_a=1/2$ and $J_b=3/2$, Eq. (69) yields

$$D_1^{(0)}=\gamma/12\gamma_b,\quad D_1^{(1)}=\gamma/72\gamma_b,\quad D_1^{(2)}=0.$$

For $J_c=J_a=J_b=1/2$, from Eq. (69) we find

$$D_1^{(0)}=\gamma/12\gamma_b,\quad D_1^{(1)}=\gamma/36\gamma_b,\quad D_1^{(2)}=0.$$

In a weak optical pump field, which enables us to replace the sine in Eq. (15) for $t-t_0=\tau$ with its argument, Eq. (66) becomes

$$D_1^{(\varkappa)}=(-1)^{J_c+J_b+1}\frac{\gamma(2\varkappa+1)(2J_b+1)}{\gamma_b(2J_c+1)}\begin{pmatrix}1 & 1 & \varkappa\\ 1 & -1 & 0\end{pmatrix}^2 \times\begin{Bmatrix}1 & 1 & \varkappa\\ J_a & J_a & J_b\end{Bmatrix}\begin{Bmatrix}1 & 1 & \varkappa\\ J_b & J_b & J_c\end{Bmatrix}\begin{Bmatrix}J_a & \varkappa & J_a\\ J_b & 1 & J_b\end{Bmatrix}\theta^2.$$

For small angular momenta we therefore obtain the following values:

(1) for $J_c=0$ and $J_a=J_b=1$, $D_1^{(0)}=\gamma\phi^2/45\gamma_b$, $D_1^{(1)}=-\gamma\phi^2/72\gamma_b$, $D_1^{(2)}=\gamma\phi^2/216\gamma_b$;
(2) for $J_c=J_a=1/2$ and $J_b=3/2$, $D_1^{(0)}=\gamma\phi^2/36\gamma_b$, $D_1^{(1)}=5\gamma\phi^2/432\gamma_b$, $D_1^{(2)}=0$;
(3) for $J_c=J_a=J_b=1/2$, $D_1^{(0)}=\gamma\phi^2/36\gamma_b$, $D_1^{(1)}=\gamma\phi^2/108\gamma_b$, $D_1^{(2)}=0$.

4. RADIATIVE FORCE FOR AN ATOM WITH LINEAR OPTICAL PUMPING

If an atom with linear optical polarization (25), with $s=0$, enters the field of light waves (35), the radiative force is described by Eq. (45) with the quantity Q in the form of the sum (48) containing terms with $s=0$, $\varkappa=0,2$, and $q_1+q_2=0,\pm2$. In terms with $\varkappa=2$ and $q_1+q_2=\pm2$, we must use the multipole moment relation

$$\rho_{\pm2}^{(2)}(J_a,0)=-\sqrt{\frac{3}{2}}\,\rho_0^{(2)}(J_a,0),\tag{70}$$

which follows from the properties of the Wigner D functions,

$$D_{0,\pm2}^2\left(0,-\frac{\pi}{2},0\right)=-\sqrt{\frac{3}{2}}\,D_{00}^2\left(0,-\frac{\pi}{2},0\right),\tag{71}$$

and from Eqs. (22), (25), and (27), all for $s=0$. This makes it possible to represent (48) as a sum of two parts, which are proportional to **AB** and to A_xB_x. Here A_x and B_x are the projections of the vectors (43) and (47) on the x axis, which is parallel to the polarization vector $\mathbf{l}_0$ of the optical pump wave (1). As a result, the force (45) for an atom with a linear optical polarization (25), with $s=0$, moving in the field of the light waves (35) may be written, by (70) and (71), as

$$\mathbf{F}=\frac{2\mathbf{k}_1|d_{ba}|^2}{\hbar\gamma_b}\left[(D_0^{(0)}-2D_0^{(2)})(\mathbf{AB})+6D_0^{(2)}A_xB_x\right]+\text{c.c.},\tag{72}$$

where the quantity $D_0^{(\varkappa)},(\varkappa=0,2)$, is given by Eq. (52) with $s=0$, and Eq. (72) holds for light waves (35) of arbitrary polarization and with $\mathbf{k}_1=\mathbf{k}_2$ or $\mathbf{k}_1=-\mathbf{k}_2$.

For circularly polarized counterpropagating waves (35) with $\mathbf{k}_1=-\mathbf{k}_2, R_1=R_2=R$, and the same sense of circular polarization ($\lambda_1=\lambda_2=\pm1$), the radiative force (72) takes the form

$$\mathbf{F}=\frac{4\mathbf{k}_1\hbar f^2}{\gamma_b}\{[I'(\mathbf{k}_1)-I'(-\mathbf{k}_1)][D_0^{(0)}+D_0^{(2)}(1-3\cos\phi)] +3D_0^{(2)}[I''(\mathbf{k}_1)+I''(-\mathbf{k}_1)]\sin\phi\},\tag{73}$$

and for the steady-state regime (56) it can be written

$$\mathbf{F}=\frac{\mathbf{F}_0\Delta}{\gamma_b^3}\left\{[D_0^{(0)}+D_0^{(2)}(1-\cos\phi)]\gamma_b(\mathbf{k}_1\mathbf{v})+3D_0^{(2)} \times\left[\Delta^2-(\mathbf{k}_1\mathbf{v})^2+\frac{\gamma_b^2}{4}\right]\sin\phi\right\}.\tag{74}$$

For circularly polarized counterpropagating waves (35) with opposite senses of polarization ($\lambda_1=-\lambda_2=\pm1$), instead of (73) we obtain

$$\mathbf{F}=\frac{4\mathbf{k}_1\hbar f^2}{\gamma_b}(D_0^{(0)}+D_0^{(2)})\{[I'(\mathbf{k}_1)-I'(-\mathbf{k}_1)] \times(1+\cos\phi)-[I''(\mathbf{k}_1)+I''(-\mathbf{k}_1)]\sin\phi\},\tag{75}$$

and in the steady-state regime (56) the force (75) is

$$\mathbf{F}=\frac{\mathbf{F}_0\Delta}{\gamma_b^3}(D_0^{(0)}+D_0^{(2)})\left\{\gamma_b(\mathbf{k}_1\mathbf{v})(1+\cos\phi) -\left[\Delta^2-(\mathbf{k}_1\mathbf{v})^2+\frac{\gamma_b^2}{4}\right]\sin\phi\right\}.\tag{76}$$

If the counterpropagating waves (35) are polarized linearly and have polarization vectors (61), then the radiative force (72) for $R_1=R_2=R$ is of particular interest:

$$\mathbf{F}=\frac{4\mathbf{k}_1\hbar f^2}{\gamma_b}\{3D_0^{(2)}(\cos^2\varphi_1-\cos^2\varphi_2)[I'(\mathbf{k}_1)+I'(-\mathbf{k}_1)]+[D_0^{(0)}+(3\cos^2\varphi_1+3\cos^2\varphi_2-2)D_0^{(2)}] \times[I'(\mathbf{k}_1)-I'(-\mathbf{k}_1)]+[(D_0^{(0)}-2D_0^{(2)})\cos\varphi +6D_0^{(2)}\cos\varphi_1\cos\varphi_2][[I''(\mathbf{k}_1)+I''(-\mathbf{k}_1)]\sin\phi -[I'(\mathbf{k}_1)-I'(-\mathbf{k}_1)]\cos\phi]\}.\tag{77}$$

For the steady-state regime (56), the force (77) is

$$\mathbf{F}=\frac{\mathbf{F}_0}{\gamma_b^3}\left\{[D_0^{(0)}+(3\cos^2\varphi_1+3\cos^2\varphi_2-2)D_0^{(2)}]\Delta\gamma_b(\mathbf{k}_1\mathbf{v}) +\frac{3}{2}D_0^{(2)}(\cos^2\varphi_1-\cos^2\varphi_2)\gamma_b\left(\Delta^2+(\mathbf{k}_1\mathbf{v})^2+\frac{\gamma_b^2}{4}\right) -\Delta[(D_0^{(0)}-2D_0^{(2)})\cos\varphi+6D_0^{(2)}\cos\varphi_1\cos\varphi_2] \times\left[\gamma_b(\mathbf{k}_1\mathbf{v})\cos\phi-\left(\Delta^2-(\mathbf{k}_1\mathbf{v})^2+\frac{\gamma_b^2}{4}\right)\sin\phi\right]\right\}.\tag{78}$$

Radiative forces for an atom with prior linear polarization, Eqs. (73)–(78), contain terms that are even or odd in **v**. Outside resonance, $\Delta\neq0$, all of the forces (73)–(78) have interference terms with factors $\sin(2\mathbf{k}_1\mathbf{r})$ and $\cos(2\mathbf{k}_1\mathbf{r})$, and thus exhibit spatial oscillations with a period of half a wavelength, π/k_1.

In the resonant case $\Delta=0$, the forces (73)–(76) vanish, whereas the forces (77) and (78) do not. For $\Delta=0$ and R_1

$\neq R_2$, the force (77) does not exhibit spatial oscillations and contains independent contributions from the individual light waves:

$$\mathbf{F}=\frac{4|d_{ba}|^2}{\hbar\gamma_b}\sum_{n=1}^{2}\mathbf{k}_n[D_0^{(0)}-2D_0^{(2)}\times(1-3\cos^2\varphi_n)]R_n^2I'(\mathbf{k}_n). \quad (79)$$

Given that $D_0^{(0)}>2D_0^{(2)}$, the force (79) for either light wave with index n is directed along the wave vector $\mathbf{k}_n$. For counterpropagating waves with $R_1=R_2,=R$, the terms $D_0^{(0)}-2D_0^{(2)}$ drop out and the remaining terms are different from zero only in the presence of alignment,

$$\mathbf{F}=\frac{6\mathbf{k}_1\hbar f^2\gamma_b(\cos^2\varphi_1-\cos^2\varphi_2)}{(\mathbf{k}_1\mathbf{v})^2+(\gamma_b/2)^2}D_0^{(2)}. \quad (80)$$

The force (80) is even in $\mathbf{v}$ and changes sign under the substitution $\mathbf{k}_1\rightarrow-\mathbf{k}_1(\mathbf{k}_2\rightarrow-\mathbf{k}_2)$ if one fixes the polarization planes of the counterpropagating waves. Moreover, the force (80) changes sign if one fixes the direction $\mathbf{k}_1(\mathbf{k}_2)$ and interchanges the polarization planes of the counterpropagating waves, which leads in Eq. (80) to the interchange $\varphi_1\leftrightarrow\varphi_2$. In particular, for $\varphi_1=\varphi_2$ the force (80) vanishes. Here, positive angles φ_1 and φ_2 are reckoned clockwise from the x axis looking along the z axis, and the x and z axes characterize the atomic alignment. Depending on φ_1 and φ_2, Eq. (80) is either a damping force or an accelerating force.

Owing to the atomic alignment, one can choose angles φ_1 and φ_2 such that for $\Delta\neq0$, the forces (77) and (78) contain only independent contributions from the two light waves, with no interference terms. To this end we must set $\varphi_1=0$ and $\varphi_2=\pm\pi/2$ or $\varphi_1=\pm\pi/2$ and $\varphi_2=0$. The forces (77) and (78) then contain both an odd (in $\mathbf{v}$) frictional part, and a part even in $\mathbf{v}$. For example, in the steady-state regime (56) for $\varphi_2=0$ and $\varphi_2=\pm\pi/2$, from (78) we find

$$\mathbf{F}=\frac{\mathbf{F}_0}{\gamma_b^2}\left\{(D_0^{(0)}+D_0^{(2)})\Delta(\mathbf{k}_1\mathbf{v})+\frac{3}{2}D_0^{(2)}\times\left[\Delta^2+(\mathbf{k}_1\mathbf{v})^2+\frac{\gamma_b^2}{4}\right]\right\}. \quad (81)$$

The force (81) does not exhibit spatial oscillations, which is a fundamental property of this force. For low longitudinal velocities (64), under the resonance conditions $\Delta=\pm\gamma_b/2$ the absolute value of the ratio of the first and second terms in brackets in (81) is

$$\left|\frac{2(D_0^{(0)}+D_0^{(2)})k_1v_z}{3D_0^{(2)}\gamma_b}\right|.$$

If one sets $\varphi_1=\pm\pi/2$ and $\varphi_2=0$, the sign of the factor 3/2 in (81) is negated. Hence, in an experiment one can control the signs of the even and odd (in $\mathbf{v}$) parts of the force (81). The parameters can be chosen such that (81) is either a damping or accelerating force.

For an ultrashort optical pump (12), the quantities $D_0^{(\varkappa)},(\varkappa=0,2)$ in Eqs. (72)–(81) can be calculated using (52) and (30) to give

$$D_0^{(\varkappa)}=-\frac{\gamma(2\varkappa+1)(2J_b+1)}{2\gamma_b(2J_c+1)}\begin{pmatrix}1 & 1 & \varkappa\\ 1 & -1 & 0\end{pmatrix}\times\begin{Bmatrix}1 & 1 & \varkappa\\ J_a & J_a & J_b\end{Bmatrix}\begin{Bmatrix}J_a & \varkappa & J_a\\ J_b & 1 & J_b\end{Bmatrix}\times\sum_{M_b}(-1)^{J_b-M_b}\begin{pmatrix}J_b & J_b & \varkappa\\ M_b & -M_b & 0\end{pmatrix}\times\left\{[B_{M_b}^{(0)}(\tau)]^2+\gamma_b\int_0^\tau[B_{M_b}^{(0)}(\xi)]^2d\xi\right\}. \quad (82)$$

If $J_c=0$ and $s=0$, according to Eq. (16) we have $J_b=1$ and $M_b=0$. Then in the case when the area (67) is large [satisfying Eq. (68)], for atomic transitions between the states with momenta $J_c=0$ and $J_a=J_b=1$, using (82) we obtain

$$D_0^{(0)}=-\gamma/18\gamma_b,\quad D_0^{(2)}=\gamma/72\gamma_b.$$

For the weak field ($\theta\ll3^{1/2}$) of an ultrashort optical pump obeying Eq. (12), Eq. (82) yields

$$D_0^{(0)}=-\gamma\theta^2/54\gamma_b,\quad D_0^{(2)}=\gamma\theta^2/216\gamma_b.$$

5. RADIATIVE FORCE FOR AN ATOM WITHOUT OPTICAL POLARIZATION

To determine changes in the radiative force (40) due to the prior optical polarization (25) with $s=0,\pm1$, consider an atom with a velocity $\mathbf{v}$ which, prior to entering the field of the counterpropagating waves (35) at point $\mathbf{r}_0'=0$ at an initial time $t_0'=0$, has a uniform Zeeman sublevel distribution at the metastable E_a level. This initial state of the atom is described by a density matrix $\rho=\rho(t)$ in the JM representation at $t=0$:

$$\rho_{M_bM_a}(0)=\rho_{M_bM_b'}(0)=0,\quad \rho_{M_aM_a'}(0)=\frac{1}{2J_a+1}\delta_{M_aM_a'}. \quad (83)$$

The initial conditions (83) for Eqs. (36)–(38) then satisfy all the requirements used in calculating the radiative forces (55), (59), (62), and (72)–(78). Therefore the force (40) for an atom with no prior optical polarization can be described by Eqs. (55), (59), (62), and (72)–(78) in which, according to (83), we must put

$$D_s^{(0)}=\frac{1}{3(2J_a+1)},\quad D_s^{(\varkappa)}=0,\quad \varkappa=1,2,\quad s=0,\pm1. \quad (84)$$

Thus in the steady-state regime (56) with $R_1=R_2=R$, the force (40) for an atom with no prior optical polarization moving in the field of counterpropagating circular waves (35) with the same sense of circular polarization ($\lambda_1=\lambda_2=\pm1$) takes the form

$$\mathbf{F}=\frac{\mathbf{F}_0\Delta(\mathbf{k}_1\mathbf{v})}{3(2J_a+1)\gamma_b^2}. \quad (85)$$

For opposite senses of circular polarization ($\lambda_1=-\lambda_2=\pm1$), the force (40) has a different value,

$$\mathbf{F}=\frac{\mathbf{F}_0\Delta}{3(2J_a+1)\gamma_b^3}\left\{\gamma_b(\mathbf{k}_1\mathbf{v})(1+\cos\phi)-\left[\Delta^2-(\mathbf{k}_1\mathbf{v})^2+\frac{\gamma_b^2}{4}\right]\sin\phi\right\}. \quad (86)$$

If an atom with no prior optical polarization moves in a field due to linearly polarized counterpropagating light waves (35) with polarization vectors (61), the radiative force (40) is

$$\mathbf{F}=\frac{\mathbf{F}_0\Delta}{3(2J_a+1)\gamma_b^3}\left\{\gamma_b(\mathbf{k}_1\mathbf{v})(1-\cos\varphi\cos\phi)+\left[\Delta^2-(\mathbf{k}_1\mathbf{v})^2+\frac{\gamma_b^2}{4}\right]\cos\varphi\sin\phi\right\}. \quad (87)$$

For an orthogonal velocity $\mathbf{k}_1\mathbf{v}=0$, the forces (85)–(87) are identical to those in Ref. 18 if in the latter the elliptical polarizations are replaced by circular and linear polarizations, and if the magnetic field is omitted. The forces (85)–(87) at $\Delta=0$ vanish, in contrast to those for an optically polarized atom. For $\Delta\neq 0$, the forces (86) and (87) as functions of the coordinates exhibit spatial oscillations with a period π/k_1. These oscillations disappear for the force (87) when $\varphi=\pm\pi/2$ but reappear in the next order of perturbation theory. To eliminate the spatial oscillations, various methods for rectifying the radiative force (40) exist. For example, Refs. 4–7 make use of the interference phenomena arising from addition of two or more light waves, while Refs. 12–15 average the radiative force (40) over a half wavelength, π/k_1.

In low-intensity counterpropagating light waves (35), such averaging makes the $\sin(2\mathbf{k}_1 r)$ and $\cos(2\mathbf{k}_1 r)$ terms vanish; the averaged terms that survive do not oscillate spatially but they are quadratic functions of the perturbation parameter. In contrast, radiative forces for an optically polarized atom, Eqs. (57), (65), (80), and (81), do not require either a rectification procedure or an averaging over half a wavelength π/k_1. These forces are fundamentally new as far as their vector properties and other quantitative characteristics are concerned. In particular, for low longitudinal velocities (64) under the resonance conditions $\Delta=\pm\gamma_b/2$, the absolute ratio of the wavelength-averaged forces (85)–(87) to the radiative force for an atom with circular polarization (65) is

$$\frac{|\mathbf{k}_1\mathbf{v}|}{3\gamma_b(2J_a+1)|D_1^{(1)}|},$$

whereas for a linearly polarized atom subject to the force (81), this ratio can be written

$$\frac{2|\mathbf{k}_1\mathbf{v}|}{9\gamma_b(2J_a+1)|D_0^{(2)}|}.$$

In low-intensity light waves (35) at low longitudinal velocities (64), the force (40) was calculated in Refs. 12–15 for a special atomic model and under the assumptions that the population of the two lower-state sublevels, at $\tau_p\ll t$, changes adiabatically under the influence of the light waves, and that the upper-state sublevel populations may be ignored. These calculations are beyond the perturbation theory region $0\leqslant t\ll\tau_p$ and are only valid for the $J_a=1/2\rightarrow J_b=1/2,3/2$ atomic transitions. To rectify the force, Refs. 12–15 take a half-wavelength average. As we have already shown, the use of optically polarized atoms serves as an alternative method for obtaining a rectified force for arbitrary momenta J_a and J_b.

6. DISCUSSION

To facilitate the calculations and simplify the mathematics, we have examined atoms in a spinless-nucleus Λ configuration atoms. Examples include the $E_c(6^1S_0)$,$E_a(5^3D_1)$, and $E_b(6^1P_1^0)$ atomic levels of barium ^{138}Ba, and the $E_c[6p^2(1/2,1/2)_0]$,$E_a[6p^2(3/2,1/2)_1]$, and $E_b[6p7s(1/2,1/2)_1^0]$ levels of lead ^{208}Pb.

However, radiative force experiments are usually carried out on atoms with nonzero nuclear spin, and under conditions where the nuclear spin cannot be neglected. In the presence of nuclear spin, the state of the atom is described by the energy E_F and by the quantum numbers J,I,F, and M_F, which specify the angular momentum **J**, nuclear spin **I**, the total momentum **F**, and the projection of **F** on the quantization axis. The level E for $J\geqslant I(J<I)$ is split by the hyperfine interaction into $2I+1(2J+1)$ components with energies E_F; the range of F depends on J and I.

Let the ground state E_0 of an atom with a given angular momentum J_0 split into two hyperfine-structure components E_{F_c} and E_{F_a} corresponding to total momenta F_c and F_a, where $|J_0-I|\leqslant F_c,F_a\leqslant J_0+I$, and $E_{F_c}<E_{F_a}$. The excited level E_b for an atomic state with angular momentum J_b splits into hyperfine components with energies E_{F_b}, where $|J_b-I|\leqslant F_b\leqslant J_b+I$. If the frequency ω_0 of the optical pump wave (1) is close to the transition frequency $\omega_{F_bF_c}=(E_{F_b}-E_{F_c})\hbar^{-1}$, and if the frequency ω of the counterpropagating waves (35) is close to the transition frequency $\omega_{F_bF_a}=(E_{F_b}-E_{F_a})\hbar^{-1}$, then the hyperfine structure components E_{F_c},E_{F_a}, and E_{F_b} form the Λ sublevel configuration required. The radiative force analyses carried out for an optically polarized atom will then also apply in the presence of nuclear spin if in all of the formulas obtained one makes the substitutions

$$E_c\rightarrow E_{F_c},E_a\rightarrow E_{F_a},E_b\rightarrow E_{F_b},J_c\rightarrow F_c,J_a\rightarrow F_a,J_b\rightarrow F_b,$$
$$M_c\rightarrow M_{F_c},M_a\rightarrow M_{F_a},M_b\rightarrow M_{F_b},d_{bc}\rightarrow d_{F_bF_c},d_{ba}\rightarrow d_{F_bF_a},$$

where $d_{F_bF_c}$ is the reduced dipole moment for the transition $F_b\rightarrow F_c$, which alters the total momentum. Examples are the $3^2S_{1/2}$, and 3^2P^0 terms of sodium ^{23}Na, $5^2S_{1/2}$ and 5^2P^0 of rubidium ^{85}Rb, and $6^2S_{1/2}$ and 6^2P^0 of cesium ^{133}Cs. It should be kept in mind that in the equilibrium state prior to entering the optical pump wave field (1), the E_{F_c} and E_{F_a} levels are equally populated, which halves the coefficients $D_1^{(1)}$ and $D_0^{(2)}$.

These arguments also hold for a four-level configuration $E_c<E_a<E_b<E_g$, with transitions $E_c\rightarrow E_a$ and $E_b\rightarrow E_g$ forbidden, provided the optical pump wave (1) is resonant with the allowed transition $E_c\rightarrow E_b$ and the counterpropagating light waves (35) are resonant with the allowed $E_a\rightarrow E_g$ transition. One must then make the substitutions $d_{ba}\rightarrow d_{ga}$, $\gamma_b\rightarrow\gamma_g$, and $\Delta\rightarrow\omega-(E_g-E_a)\hbar^{-1}$ in the radiative force formulas for an optically polarized atom. The generalization to hyperfine structure components can be carried out in a similar manner.

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