Optical-pressure force in the cases of Raman and two-photon resonances

A. I. Alekseev and G. Yu. Tikhonov

Moscow Engineering Physics Institute, 115409 Moscow, Russia
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Perturbation theory in the resonance approximation is used to calculate new optical-pressure forces in the case of two-photon atomic transitions corresponding to Raman or two-photon resonance for two pairs of light waves with specially chosen polarizations moving in opposite directions. The forces are due to four-photon scattering of light by the atom and therefore have the fourth order in the electric field of the light waves. The new forces are rectified radiation forces and are stronger than the optical-pressure forces of second order in the electric field of light waves not in resonance with electric dipole transitions. This means that the new optical-pressure forces are the principal forces for the considered conditions when for the two pairs of opposite light waves only Raman resonance or two-photon resonance is realized. The new optical-pressure forces are odd functions of the velocity of the atom and for a definite sign of the detuning of the Raman or two-photon resonance act as friction forces. The dependence of these forces on the atom's velocity, the detuning from resonance, the relaxation, the angular momenta of degenerate levels, and the polarizations of the light waves is investigated.

1. INTRODUCTION

The optical-pressure force for an atom in the electric field of monodirectional or oppositely directed light waves in resonance with an electric dipole transition has been studied extensively for several years (see Refs. 1–3 and the references cited there). Since this force is widely used to localize atoms by light fields and in laser cooling of atoms, particular attention is devoted to obtaining a rectified optical-pressure force that is constant or varies weakly over the wavelength of the light wave. To enhance the rectification effect of the radiation force, light waves of the same frequency[4–6] or bichromatic light waves[4,7,8] are used in the case of an electric dipole transition; a special model of an atom having an electric dipole transition with change $1/2\to3/2$ or $1/2\to1/2$ of the angular momentum, for which a rectified radiation force appears after averaging over half the wavelength, has also been considered.[9–13] To this end, consideration is also given to three-level atoms with Λ configuration of nondegenerate levels (Λ atoms) that interact with two or several light waves in resonance with adjacent electric dipole transitions of a Λ atom.[14–21] The methods developed for Λ atoms are also used for atoms with K configuration of nondegenerate levels to calculate the optical-pressure force and its application for laser cooling of atoms.[22,23] However, in Refs. 24 and 25 it is shown that allowance for degeneracy of the levels in the determination of the rectified radiation force leads to appreciable computational difficulties. At the same time, allowance for level degeneracy with respect to the projection of the angular momenta is fundamentally necessary if, in addition to other characteristics, one is studying the vector properties of the optical-pressure force.[26] This circumstance is particularly important in the cases where the atom has optical polarization before entering the field of the light waves.[27]

In contrast to previous investigations of the optical-pressure forces associated with resonance electric dipole transitions in which the parity of the atomic state changes,[1–27] in this paper we consider resonance two-photon transitions without change in the parity of the atomic state. We calculate the optical-pressure force for an atom in Raman or two-photon resonance with two light waves, and also with two pairs of oppositely directed light waves. The calculations are based on the standard expression[1–3] for the optical-pressure force

$$\mathbf{F}=\mathrm{Tr}[\rho\nabla(\mathbf{dE})],$$

which was applied earlier to resonance electric dipole transitions of an atom in the electric field $\mathbf{E}$ of light waves under conditions where the state of the atom is described by a density matrix ρ and its electric polarization is determined by averaging the operator $\mathbf{d}$ of the dipole moment. Application of this expression to an atom in the presence of only Raman resonance or only two-photon resonance revealed a number of interesting properties of the optical-pressure force $\mathbf{F}$ due to two-photon transitions from the ground level E_b to an excited level E_c without change in the parity of the atomic state. The solution to the problem was obtained in the resonance approximation by taking into account the level degeneracy with respect to the projections of the angular momenta of the atom onto the quantization axis.

In the domain of applicability of perturbation theory for two light waves with total electric field $\mathbf{E}$, the nonresonance optical-pressure force $\mathbf{F}^{(2)}$ of second order in the field $\mathbf{E}$ is stronger than the optical-pressure force $\mathbf{F}^{(4)}$ of fourth order in the field $\mathbf{E}$, despite the fact that the force $\mathbf{F}^{(4)}$ is due to Raman or two-photon resonances. At the same time, each of the forces $\mathbf{F}^{(2)}$ and $\mathbf{F}^{(4)}$ can be decomposed into two essentially different parts, of which the first is an even and the second an odd function of the velocity $\mathbf{v}$ of the atom. Moreover, the even part is much greater than the odd part in the case of $\mathbf{F}^{(2)}$, whereas for $\mathbf{F}^{(4)}$ the two parts have the same order when

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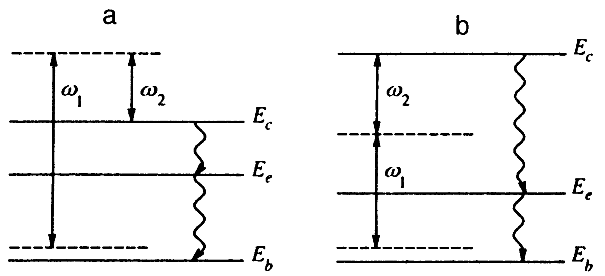

FIG. 1. Level scheme for Raman (a) and two-photon (b) resonances. The solid vertical lines characterize two-photon transitions of the atom under the influence of light waves with frequencies ω_1 and ω_2. The wavy lines describe adjacent electric dipole transitions $E_c \to E_e$ and $E_e \to E_b$ between the excited levels E_c and E_e and the ground level E_b.

$$|(\mathbf{k}_1 \pm \mathbf{k}_2)\mathbf{v}\Delta_{12}| \sim \gamma_{cb}^2,$$

where $\mathbf{k}_1$ and $\mathbf{k}_2$ are the wave vectors, γ_{cb} is the damping rate of the optical coherence of the two-photon transition $E_b \to E_c$, and Δ_{12} is the detuning from the Raman or two-photon resonance. If two pairs of oppositely directed light waves are used, then under certain conditions the sum of the terms even in $\mathbf{v}$ vanishes for the force $\mathbf{F}^{(2)}$ and the force $\mathbf{F}^{(4)}$. The remaining sum of the $\mathbf{v}$-odd terms in $\mathbf{F}^{(2)}$ is negligibly small compared with the $\mathbf{v}$-odd terms in $\mathbf{F}^{(4)}$.

Thus, in the presence of only Raman resonance or only two-photon resonance in the field of two pairs of oppositely directed light waves, a new optical-pressure force $\mathbf{F}^{(4)}$ arises due to Raman scattering of light by an individual atom–the only force that acts on the atom. This optical-pressure force is a rectified radiation force and, depending on the sign of the detuning from the Raman or two-photon resonance, can act as an accelerating or decelerating force for a moving atom.

2. INDUCED ELECTRIC DIPOLE MOMENT OF THE ATOM

We consider an atom with zero spin of its nucleus and possessing a two-photon transition of frequency $\omega_{cb} = (E_c - E_b)\hbar^{-1}$. Here E_b and E_c are the energies of the ground state and an excited state, between which there is an intermediate level E_e that occurs in adjacent electric dipole transitions $E_c \to E_e$ and $E_e \to E_b$, as indicated in Fig. 1. In an arbitrary state, the atom is characterized by, in addition to the energy E_g, the quantum number J_g of the angular momentum $\mathbf{J}_g$ and its projection $\hbar M_g$ onto the quantization axis (the index g is used to label all levels of the discrete spectrum). This atom moves in the field of a finite number n_0 of monochromatic light waves:

$$\mathbf{E} = \sum_{n=1}^{n_0} \mathbf{a}_n \exp[i(\mathbf{k}_n \mathbf{r} - \omega_n t)] + \text{c.c.}, \tag{1}$$

where the complex amplitude $\mathbf{a}_n$ determines the intensity and polarization of the nth light wave. The direction of the wave vector $\mathbf{k}_n$ is arbitrary. The frequencies ω_1 and ω_2 ($\omega_1 > \omega_2$) satisfy the conditions of Raman resonance (Fig. 1a)

$$|\omega_1 - \omega_2 - \omega_{cb}| \lesssim \gamma_{cb} \tag{2}$$

or two-photon resonance (Fig. 1b)

$$|\omega_1 + \omega_2 - \omega_{cb}| \lesssim \gamma_{cb}, \tag{3}$$

whereas each frequency ω_1 and ω_2 separately is not in resonance and is not equal to $\omega_3, \dots, \omega_{n_0}$. In addition, the frequencies $\omega_3, \dots, \omega_{n_0}$, and also their differences and sums, are also not in resonance. The doubled frequencies $2\omega_n$ of all the light waves (1) with $n = 1, \dots, n_0$ are nonresonant, i.e., $|2\omega_n - \omega_{cb}| \ll \gamma_{cb}$, and therefore two-photon absorption of photons of the same frequency will not occur without the presence of another wave.

The interaction of the moving atom with the light waves (1) can be described by means of the equation for the matrix elements $\rho_{M_f M_g}$ of the density matrix ρ in the JM representation:

$$\left(\frac{\partial}{\partial t} + \mathbf{v}\nabla + i\omega_{fg} + \gamma_{fg}\right)\rho_{M_f M_g'} = \frac{i}{\hbar}\sum_k (\rho_{M_f M_k} V_{M_k M_g'} - V_{M_f M_k}\rho_{M_k M_g'}) + \sum_{s(s>g)} \frac{\gamma_{s\to g}(2J_s+1)\delta_{fg}}{|d_{sg}|^2} \mathbf{d}_{M_f M_s}\rho_{M_s M_s'}\mathbf{d}_{M_s' M_g'}, \tag{4}$$

where

$$V_{M_f M_g} = -\sum_{n=1}^{n_0} \{\mathbf{a}_n \mathbf{d}_{M_f M_g} \exp[i(\mathbf{k}_n\mathbf{r} - \omega_n t)] + \mathbf{a}_n^* \mathbf{d}_{M_f M_g} \exp[-i(\mathbf{k}_n\mathbf{r} - \omega_n t)]\},$$

$$\gamma_{s\to g} = 4|d_{sg}|^2\omega_{sg}^3/3\hbar c^3(2J_s+1),$$

$$\omega_{fg} = (E_f - E_g)/\hbar, \quad \gamma_{fg} = (\gamma_f + \gamma_g)/2, \quad \gamma_{gg} = \gamma_g,$$

$\mathbf{v}$ is the constant velocity of the atom, $V_{M_f M_g}$ and $d_{M_f M_g}$ are the matrix elements of the operators of the interaction $V = -\mathbf{Ed}$ and of the electric dipole moment $\mathbf{d}$, d_{sg} is the reduced dipole moment (Ref. 28), γ_{fg} is the half-width of the spectral line of the atomic transition $E_f \to E_g$, $\hbar\gamma_f$ and $\hbar\gamma_g$ are the homogeneous widths of the levels E_f and E_g, and $\gamma_{s\to g}$ is the probability of spontaneous emission of a photon $\hbar\omega_{sg}$ by the isolated atom for the electric dipole transition from the upper level E_s to the lower level E_g. The indices f, g, k, and s label all possible levels of the discrete spectrum of the atom. Summation over repeated matrix indices is assumed everywhere. The final term on the right-hand side of Eq. (4) describes the arrival of the atom at the lower level E_g due to spontaneous emission of a photon $\hbar\omega_{sg}$ at an upper level $E_s > E_g$. It is nonzero only for transitions $E_s \to E_g$ in which the parity of the atomic state changes.

The atom enters the field (1) at the point $\mathbf{r}_0 = 0$ at the time $t_0 = 0$ and with the passage of time $0 \leqslant t$ moves in this field rectilinearly $\mathbf{r} = \mathbf{v}t$ with constant velocity $\mathbf{v}$. This enables us to set in Eq. (4) and in the interaction operator V

$$\frac{\partial}{\partial t} + \mathbf{v}\nabla = \frac{d}{dt}, \quad \mathbf{k}_n\mathbf{r} - \omega_n t = -(\omega_n - \mathbf{k}_n\mathbf{v})t. \tag{5}$$

Before entering the field (1), the free atom was in the ground state with energy E_b and was described by the constant density matrix

$$\rho^{(0)}_{M_fM'_g}=\frac{\delta_{fb}\delta_{bg}}{2J_b+1}\,\delta_{M_gM'_g}. \tag{6}$$

This density matrix is the initial condition at $t_0=0$ if Eq. (4) is solved in the region $0\leqslant t$.

We seek a solution of Eq. (4) in the region $0\leqslant t$ by successive approximation in the form of a perturbation series:

$$\rho(t)=\rho^{(0)}(0)+\rho^{(1)}(t)+\rho^{(2)}(t)+\rho^{(3)}(t)+\ldots, \tag{7}$$

where the first term describes the state of the atom before it enters the field (1) and is determined by (6). The other terms of the series (7) are small corrections that describe the state of the atom in the field (1) for $0\leqslant t$ in, respectively, the linear, quadratic, cubic, and higher approximations in the field; at the initial time $t_0=0$, all the small corrections are zero. In addition, it is assumed that the field (1) is sufficiently weak that each successive term of the series (7) is less than the previous one. Therefore, the terms of higher order in the field (1) in the series (7) are omitted as small quantities that are not used in this problem.

To solve Eq. (4) in the first approximation in the field (1), it is necessary in its right-hand side to use the density matrix (6) in the terms containing the field (1). In the considered first approximation, the term that describes the arrival of atoms at a lower level due to spontaneous emission of a photon at an upper level does not contribute to the required solution $\rho^{(1)}(t)$, since in the given approximation the field (1) gives rise to transitions $E_f\rightarrow E_g$ for $f\neq g$ with a change in the parity of the atomic state. If we take into account the relation (5) and the zero initial condition for $\rho^{(1)}(t)$, we can write the solution in the first approximation in the field (1) for $0\leqslant t$ as

$$\rho^{(1)}_{M_fM_g}=\frac{(\delta_{fb}-\delta_{bg})\mathbf{d}_{M_fM_g}}{(2J_b+1)\hbar}\times\sum_{n=1}^{n_0}\left(\frac{\mathbf{a}_n\{\exp[i(\mathbf{k}_n\mathbf{r}-\omega_nt)]-\exp[-(\gamma_{fg}+i\omega_{fg})t]\}}{\omega_n-\mathbf{k}_n\mathbf{v}-\omega_{fg}+i\gamma_{fg}}-\frac{\mathbf{a}_n^*\{\exp[-i(\mathbf{k}_n\mathbf{r}-\omega_nt)]-\exp[-(\gamma_{fg}+i\omega_{fg})t]\}}{\omega_n-\mathbf{k}_n\mathbf{v}+\omega_{fg}-i\gamma_{fg}}\right).$$

After a sufficiently long time has elapsed ($1\ll\gamma_{fg}t$), this solution reaches a stationary regime:

$$\rho^{(1)}_{M_fM_g}=\frac{(\delta_{fb}-\delta_{bg})\mathbf{d}_{M_fM_g}}{(2J_b+1)\hbar}\sum_{n=1}^{n_0}\left(\frac{\mathbf{a}_n\exp[i(\mathbf{k}_n\mathbf{r}-\omega_nt)]}{\omega_n-\mathbf{k}_n\mathbf{v}-\omega_{fg}+i\gamma_{fg}}-\frac{\mathbf{a}_n^*\exp[-i(\mathbf{k}_n\mathbf{r}-\omega_nt)]}{\omega_n-\mathbf{k}_n\mathbf{v}+\omega_{fg}-i\gamma_{fg}}\right). \tag{8}$$

This enables us to determine the linear electric dipole moment $\mathbf{p}^{(1)}=\mathrm{Tr}(\rho^{(1)}\mathbf{d})$ of the atom induced by the field (1) through nonresonance electric dipole transitions:

$$\mathbf{P}^{(1)}=\sum_{n=1}^{n_0}\mathbf{p}_n^{(1)}\exp[i(\mathbf{k}_n\mathbf{r}-\omega_nt)]+\text{c.c.}, \tag{9}$$

where

$$\mathbf{p}_n^{(1)}=\frac{\mathbf{a}_n}{3(2J_b+1)\hbar}\sum_g|d_{bg}|^2\left(\frac{1}{\omega_n-\mathbf{k}_n\mathbf{v}-\omega_{bg}+i\gamma_{bg}}-\frac{1}{\omega_n-\mathbf{k}_n\mathbf{v}+\omega_{bg}+i\gamma_{bg}}\right).$$

In accordance with the method of successive approximation, we substitute the density matrix found in the first approximation in the terms on the right-hand side of Eq. (4) containing the field (1). After this procedure, we determine the density matrix $\rho^{(2)}(t)$ in the second approximation in the field (1) by solving the resulting simplified equation. We assume that the Raman resonance (2) holds. Therefore, in the solution of the simplified equation in the stationary regime for $1\ll\gamma_{cb}t$ in the second approximation in the field (1) we retain only the resonance terms. Then the term in the simplified equation with factor $\gamma_{s\rightarrow g}$ does not contribute, since it is not a resonance term. The upshot is that in the stationary regime we obtain $\rho^{(2)}_{M_fM_g}=0$ for all indices f and g, except for the values of the indices that correspond to the case of resonance:

$$\rho^{(2)}_{M_cM_b}=\frac{-1}{(2J_b+1)\hbar^2[\omega_1-\omega_2-(\mathbf{k}_1-\mathbf{k}_2)\mathbf{v}-\omega_{cb}+i\gamma_{cb}]}\times\sum_g\left(\frac{(\mathbf{a}_1\mathbf{d}_{M_cM_g})(\mathbf{a}_2^*\mathbf{d}_{M_gM_b})}{\omega_1-\omega_{cg}}-\frac{(\mathbf{a}_2^*\mathbf{d}_{M_cM_g})(\mathbf{a}_1\mathbf{d}_{M_gM_b})}{\omega_1-\omega_{gb}}\right)\times\exp\{i[(\mathbf{k}_1-\mathbf{k}_2)\mathbf{r}-(\omega_1-\omega_2)t]\},$$

$$\rho^{(2)}_{M_bM_c}=(\rho^{(2)}_{M_cM_b})^*. \tag{10}$$

In the nonresonance denominators in (10), the detuning $\omega_1-\omega_2-\omega_{cb}$ from resonance, the Doppler shifts of the frequencies of the light waves, and the relaxation constant $\pm i\gamma_{gb}$ are omitted as small quantities compared with $|\omega_1-\omega_{cg}|$ and $|\omega_1-\omega_{gb}|$. This measure made it possible to use the equations

$$\omega_2+\omega_{gb}=\omega_1-\omega_{cg},\quad \omega_2+\omega_{cg}=\omega_1-\omega_{gb}. \tag{11}$$

The density matrix (10) which we found characterizes the optical coherence in the two-photon transition $E_b\rightarrow E_c$, and therefore it cannot describe the emission or absorption of a photon by the atom, which is forbidden by the parity conservation law. The nonlinear process of absorption of two photons of the field (1) and emission of a different photon is described by the density matrix $\rho^{(3)}(t)$ of the cubic approximation in the field (1). To determine $\rho^{(3)}(t)$ by means of the solution of Eq. (4) in the stationary regime in the third approximation in the field (1), we introduce the convenient notation

$$\rho^{(2)}_{M_fM_g}=r_{M_fM_g}\exp(-i\omega_{fg}t), \tag{12}$$

where the matrix $r_{M_fM_g}$ is equal to zero for all possible f and g except $f=c$ and $g=b$, and also $f=b$ and $g=c$. In addition, it can be seen from comparison of (12) with (10) for $f=c$ and $g=b$ that the matrix $r_{M_fM_g}$ is a slow function of the time, since it is proportional to $\exp[i(\omega_{cb}-\omega_1+\omega_2)t]$ with small detuning from resonance: $|\omega_1-\omega_2-\omega_{cb}|\lesssim\gamma_{cb}$. By virtue of the notation (12), we can sum over the indices f and g in the complete range of their variation when calculating the density matrix in the third approximation in the field (1) by solving the equation

$$\left(\frac{d}{dt}+i\omega_{fg}+\gamma_{fg}\right)\rho^{(3)}_{M_fM_g}=\frac{i}{\hbar}\sum_k (\rho^{(2)}_{M_fM_k}V_{M_kM_g}-V_{M_fM_k}\rho^{(2)}_{M_kM_g}),$$

where $f\neq g$. By virtue of (12), the solution of this equation can be represented in the compact form

$$\rho^{(3)}_{M_fM_g}=\sum_k (\rho^{(2)}_{M_fM_k}q_{M_kM_g}-q_{M_fM_k}\rho^{(2)}_{M_kM_g}),$$

where we have used the convenient notation

$$q_{M_kM_g}=-\frac{1}{\hbar}\sum_{n=1}^{n_0}\left(\frac{\mathbf{a}_n\mathbf{d}_{M_kM_g}\exp[i(\mathbf{k}_n\mathbf{r}-\omega_n t)]}{\omega_{kg}-\omega_n}+\frac{\mathbf{a}^*_n\mathbf{d}_{M_kM_g}\exp[-i(\mathbf{k}_n\mathbf{r}-\omega_n t)]}{\omega_{kg}+\omega_n}\right),$$

$$q^*_{M_kM_g}=-q_{M_gM_k}.$$

Here we have omitted in the nonresonance denominators the detuning $\omega_1-\omega_2-\omega_{cb}$ from resonance, the Doppler shifts of the frequencies of the light waves, and $i\gamma_{fg}$, since they are small compared with $|\omega_{kg}\pm\omega_n|$.

The required induced electric dipole moment of the atom in the cubic approximation in the field (1) is

$$\mathbf{P}^{(3)}=\sum_{fg}\rho^{(3)}_{M_fM_g}\mathbf{d}_{M_gM_f}=\sum_{fkg}(\rho^{(2)}_{M_fM_k}q_{M_kM_g}\mathbf{d}_{M_gM_f}-q_{M_fM_k}\rho^{(2)}_{M_kM_g}\mathbf{d}_{M_gM_f}).$$

In the first term on the right-hand side of the last equation, we redenote the summation indices in such a way as to factor out the common factor as follows:

$$\mathbf{P}^{(3)}=-\sum_{kgf}\rho^{(2)}_{M_kM_g}(\mathbf{d}_{M_gM_f}q_{M_fM_k}-q_{M_gM_f}\mathbf{d}_{M_fM_k}). \quad (13)$$

We can now take advantage of the fact that in accordance with (10), the density matrix $\rho^{(2)}_{M_kM_g}$ is nonzero only for $k=c$ and $g=b$ or $k=b$ and $g=c$. This enables us to represent the vector (13) in a different manner:

$$\mathbf{P}^{(3)}=-\rho^{(2)}_{M_cM_b}\sum_f(\mathbf{d}_{M_bM_f}q_{M_fM_c}-q_{M_bM_f}\mathbf{d}_{M_fM_c})-\rho^{(2)}_{M_bM_c}\sum_f(\mathbf{d}_{M_cM_f}q_{M_fM_b}-q_{M_cM_f}\mathbf{d}_{M_fM_b})=\rho^{(2)}_{M_cM_b}\mathbf{G}_{M_bM_c}+\text{c.c.}, \quad (14)$$

where

$$\mathbf{G}_{M_bM_c}=\frac{1}{\hbar}\sum_g\sum_{n=1}^{n_0}\left\{\left(\frac{(\mathbf{d}_{M_bM_g}\mathbf{a}_n)\mathbf{d}_{M_gM_c}}{\omega_{gb}+\omega_n}+\frac{\mathbf{d}_{M_bM_g}(\mathbf{a}_n\mathbf{d}_{M_gM_c})}{\omega_{gc}-\omega_n}\right)\exp[i(\mathbf{k}_n\mathbf{r}-\omega_n t)]+\left(\frac{(\mathbf{d}_{M_bM_g}\mathbf{a}^*_n)\mathbf{d}_{M_gM_c}}{\omega_{gb}-\omega_n}+\frac{\mathbf{d}_{M_bM_g}(\mathbf{a}^*_n\mathbf{d}_{M_gM_c})}{\omega_{gc}+\omega_n}\right)\times\exp[-i(\mathbf{k}_n\mathbf{r}-\omega_n t)]\right\}. \quad (15)$$

In order to sum over the projections of the angular momenta in (14) with allowance for (10) and (15), we use the Wigner–Eckart theorem and the rules for contracting $3j$ symbols,[28,29] and we also use the method of calculation developed in Ref. 30. This enables us to obtain for arbitrary vectors $\mathbf{A}_1$, $\mathbf{A}_2$, and $\mathbf{A}_3$ the general expressions

$$(\mathbf{A}_1\mathbf{d}_{M_cM_f})(\mathbf{A}_2\mathbf{d}_{M_fM_b})(\mathbf{A}_3\mathbf{d}_{M_bM_g})\mathbf{d}_{M_gM_c}=\frac{1}{2}(D_1+D_2)\mathbf{A}_1(\mathbf{A}_2\mathbf{A}_3)+\frac{1}{2}(D_2-D_1)\mathbf{A}_2(\mathbf{A}_3\mathbf{A}_1)+\frac{1}{3}(D_0-D_2)\mathbf{A}_3(\mathbf{A}_1\mathbf{A}_2), \quad (16)$$

$$(\mathbf{A}_1\mathbf{d}_{M_cM_f})(\mathbf{A}_2\mathbf{d}_{M_fM_b})\mathbf{d}_{M_bM_g}(\mathbf{A}_3\mathbf{d}_{M_gM_c})=\frac{1}{2}(L_1+L_2)\mathbf{A}_1(\mathbf{A}_2\mathbf{A}_3)+\frac{1}{2}(L_2-L_1)\mathbf{A}_2(\mathbf{A}_3\mathbf{A}_1)+\frac{1}{3}(L_0-L_2)\mathbf{A}_3(\mathbf{A}_1\mathbf{A}_2), \quad (17)$$

where

$$D_\kappa=d_{cf}d_{fb}d^*_{cg}d^*_{gb}\begin{Bmatrix}1 & \kappa & 1\\ J_c & J_f & J_b\end{Bmatrix}\begin{Bmatrix}1 & \kappa & 1\\ J_c & J_g & J_b\end{Bmatrix},$$

$$L_\kappa=(-1)^\kappa D_\kappa,\quad \kappa=0,1,2.$$

The $6j$ symbol $\begin{Bmatrix}abc\\ deh\end{Bmatrix}$ used here is defined in Refs. 28 and 29.

By means of the general expressions (16) and (17) we can calculate the induced electric dipole moment (14) of the atom in the cubic approximation in the field (1). Omitting the simple but laborious calculations, we find

$$\mathbf{P}^{(3)}=\sum_{n=1}^{n_0}\{\mathbf{P}^{(+)}_{n12}(\omega^{(+)}_{n12})\exp[i(\mathbf{k}^{(+)}_{n12}\mathbf{r}-\omega^{(+)}_{n12}t)]+\mathbf{P}^{(-)}_{n12}\times(\omega^{(-)}_{n12})\exp[i(\mathbf{k}^{(-)}_{n12}\mathbf{r}-\omega^{(-)}_{n12}t)]\}+\text{c.c.}, \quad (18)$$

where

$$\omega^{(+)}_{n12}=\omega_n+\omega_1-\omega_2,\quad \mathbf{k}^{(+)}_{n12}=\mathbf{k}_n+\mathbf{k}_1-\mathbf{k}_2,$$

$$\omega^{(-)}_{n12}=\omega_n-\omega_1+\omega_2,\quad \mathbf{k}^{(-)}_{n12}=\mathbf{k}_n-\mathbf{k}_1+\mathbf{k}_2,$$

$$\mathbf{P}^{(+)}_{n12}(\omega^{(+)}_{n12})=\chi^{(+)}_1(\omega^{(+)}_{n12})\mathbf{a}_1(\mathbf{a}^*_2\mathbf{a}_n)+\chi^{(+)}_2\times(\omega^{(+)}_{n12})\mathbf{a}^*_2(\mathbf{a}_n\mathbf{a}_1)+\chi^{(+)}_3(\omega^{(+)}_{n12})\mathbf{a}_n(\mathbf{a}_1\mathbf{a}^*_2), \quad (19)$$

$$\mathbf{P}^{(-)}_{n12}(\omega^{(-)}_{n12})=\chi^{(-)}_1(\omega^{(-)}_{n12})\mathbf{a}^*_1(\mathbf{a}_2\mathbf{a}_n)+\chi^{(-)}_2 \times(\omega^{(-)}_{n12})\mathbf{a}_2(\mathbf{a}_n\mathbf{a}^*_1)+\chi^{(-)}_3(\omega^{(-)}_{n12})\mathbf{a}_n(\mathbf{a}^*_1\mathbf{a}_2), \tag{20}$$

$$\chi^{(+)}_1(\omega)=\frac{1}{2}[B_1(\omega)+B_2(\omega)],$$

$$\chi^{(+)}_2(\omega)=\frac{1}{2}[B_2(\omega)-B_1(\omega)],$$

$$\chi^{(+)}_3(\omega)=\frac{1}{3}[B_0(\omega)-B_2(\omega)], \tag{21}$$

$$\chi^{(-)}_1(\omega)=\frac{1}{2}[B^*_1(-\omega)+B^*_2(-\omega)],$$

$$\chi^{(-)}_2(\omega)=\frac{1}{2}[B^*_2(-\omega)-B^*_1(-\omega)],$$

$$\chi^{(-)}_3(\omega)=\frac{1}{3}[B^*_0(-\omega)-B^*_2(-\omega)], \tag{22}$$

$$B_\kappa(\omega)=\frac{-1}{(2J_b+1)\hbar}\Pi^*_\kappa(\omega)\Pi_\kappa(\omega_1) \times\frac{1}{\omega_1-\omega_2-(\mathbf{k}_1-\mathbf{k}_2)\mathbf{v}-\omega_{cb}+i\gamma_{cb}}, \tag{23}$$

$$\Pi^*_\kappa(\omega)=\frac{1}{\hbar}\sum_g d^*_{cg}d^*_{gb}\begin{Bmatrix}1 & \kappa & 1\\ J_c & J_g & J_b\end{Bmatrix}\left(\frac{1}{\omega-\omega_{cg}}-\frac{(-1)^\kappa}{\omega-\omega_{gb}}\right), \tag{24}$$

$$\Pi_\kappa(\omega_1)=\frac{1}{\hbar}\sum_g d_{cg}d_{gb}\begin{Bmatrix}1 & \kappa & 1\\ J_c & J_g & J_b\end{Bmatrix}\left(\frac{1}{\omega_1-\omega_{cg}}-\frac{(-1)^\kappa}{\omega_1-\omega_{gb}}\right),$$

$$\kappa=0,1,2. \tag{25}$$

In the nonresonance denominators in (24) and (25), we have omitted the detuning from resonance, the Doppler shift of the frequency, and the relaxation constant, whereas in the resonance denominator in (23) we cannot omit these quantities. The expressions (18)–(25) are valid for all directions of the vectors $\mathbf{k}_1,\dots,\mathbf{k}_{n_0}$.

For the two-photon resonance (3), the arguments can be made similarly, but in each step of the calculations we encounter expressions that differ from the case of Raman resonance in some details. The upshot is that the induced electric dipole moment of the atom in the cubic approximation in a field (1) satisfying the requirement of two-photon resonance (3) takes the form

$$\bar{\mathbf{P}}^{(3)}=\sum_{n=1}^{n_0}\{\bar{\mathbf{P}}^{(+)}_{n12}(\bar{\omega}^{(+)}_{n12})\exp[i(\bar{\mathbf{k}}^{(+)}_{n12}\mathbf{r}-\bar{\omega}^{(+)}_{n12}t)]+\bar{\mathbf{P}}^{(-)}_{n12} \times(\bar{\omega}^{(-)}_{n12})\exp[i(\bar{\mathbf{k}}^{(-)}_{n12}\mathbf{r}-\bar{\omega}^{(-)}_{n12}t)]\}+\text{c.c.}, \tag{26}$$

where

$$\bar{\omega}^{(+)}_{n12}=\omega_n+\omega_1+\omega_2,\quad \bar{\mathbf{k}}^{(+)}_{n12}=\mathbf{k}_n+\mathbf{k}_1+\mathbf{k}_2,$$

$$\bar{\omega}^{(-)}_{n12}=\omega_n-\omega_1-\omega_2,\quad \bar{\mathbf{k}}^{(-)}_{n12}=\mathbf{k}_n-\mathbf{k}_1-\mathbf{k}_2,$$

$$\bar{\mathbf{P}}^{(+)}_{n12}(\bar{\omega}^{(+)}_{n12})=\bar{\chi}^{(+)}_1(\bar{\omega}^{(+)}_{n12})\mathbf{a}_1(\mathbf{a}_2\mathbf{a}_n)+\bar{\chi}^{(+)}_2(\bar{\omega}^{(+)}_{n12})\mathbf{a}_2(\mathbf{a}_n\mathbf{a}_1)+\bar{\chi}^{(+)}_3(\bar{\omega}^{(+)}_{n12})\mathbf{a}_n(\mathbf{a}_1\mathbf{a}_2), \tag{27}$$

$$\bar{\mathbf{P}}^{(-)}_{n12}(\bar{\omega}^{(-)}_{n12})=\bar{\chi}^{(-)}_1(\bar{\omega}^{(-)}_{n12})\mathbf{a}^*_1(\mathbf{a}^*_2\mathbf{a}_n)+\bar{\chi}^{(-)}_2(\bar{\omega}^{(-)}_{n12})\mathbf{a}^*_2(\mathbf{a}_n\mathbf{a}^*_1)+\bar{\chi}^{(-)}_3(\bar{\omega}^{(-)}_{n12})\mathbf{a}_n(\mathbf{a}^*_1\mathbf{a}^*_2). \tag{28}$$

Here $\bar{\chi}^{(+)}_m(\omega)$ and $\bar{\chi}^{(-)}_m$ with $m=1,2,3$ are equal to the quantities given in (21) and (22) after the replacement in them of $B_\kappa(\omega)$ with $\kappa=0,1,2$ by

$$\bar{B}_\kappa(\omega)=\frac{-1}{(2J_b+1)\hbar}\Pi^*_\kappa(\omega)\Pi_\kappa(\omega_1) \times\frac{1}{\omega_1+\omega_2-(\mathbf{k}_1+\mathbf{k}_2)\mathbf{v}-\omega_{cb}+i\gamma_{cb}}, \tag{29}$$

where in writing down the nonresonance denominators for $\Pi^*_\kappa(\omega)$ and $\Pi_\kappa(\omega_1)$ in (29) we have used in place of (11) the equations

$$\omega_{gb}-\omega_2=\omega_1-\omega_{cg},\quad \omega_2-\omega_{cg}=\omega_{gb}-\omega_1. \tag{30}$$

It can be seen that the transition from the Raman resonance (2) with the expressions (18)–(25) to the two-photon resonance (3) described by the expressions (26)–(30) can be made by means of the substitutions

$$\omega_2\to-\omega_2,\quad \mathbf{k}_2\to-\mathbf{k}_2,\quad \mathbf{a}_2\to\mathbf{a}^*_2,\quad \mathbf{a}^*_2\to\mathbf{a}_2. \tag{31}$$

The obtained induced electric dipole moments of the atom (18) and (26) for the Raman resonance (2) and two-photon resonance (3) are identical in the case $n_0=3$ to the corresponding vectors that describe the density of the dielectric polarization of an atomic gas obtained in the study of Raman scattering in Ref. 30 if in them we omit the atomic collisions and use the obvious equations

$$\Pi^\kappa_{cb}(\omega)=\Pi^*_\kappa(\omega),\quad \Pi_\kappa(\omega_1,\omega_2)=\Pi_\kappa(\omega_1),$$

which contain the quantities (24) and (25). In addition, the densities N_b and N_c of the resonance atoms in the levels E_b and E_c in Ref. 30 must be replaced by $1/(2J_b+1)$ and 0. At the same time, it is necessary to choose $f(v')=\delta(\mathbf{v}'-\mathbf{v})$ as the distribution function of these atoms with respect to the velocities $\mathbf{v}'$ in Ref. 30. The identity that we have noted exists, despite the differences in the equations for the density matrix and in the subsidiary conditions.

The identity of the vector (18) for $n_0=3$ with the vector of the dielectric polarization of the atomic gas in the case of Raman scattering in Ref. 30 indicates that the vector (18) describes four-photon Raman scattering of light by a single atom, which reduces to absorption of photons $\hbar\omega_1$ and $\hbar\omega_2$ of the two pumping waves in the presence of a test wave with frequency ω_n and to the emission of photons $\hbar\omega^{(+)}_{n12}$ or $\hbar\omega^{(-)}_{n12}$, respectively, of anti-Stokes and Stokes waves. This four-photon nonlinear process develops in the two-photon transition $E_b\to E_c$ if the condition of Raman resonance (2) is satisfied. Similarly, the identity of the vector (26) for $n_0=3$ with the vector of the dielectric polarization of the atomic gas in the case of four-wave scattering of light in Ref. 30 shows that the vector (26) describes four-photon scattering of light by a single atom. This scattering takes the form of the absorption of photons $\hbar\omega_1$ and $\hbar\omega_2$ of the two pumping waves in the presence of a test wave with frequency ω_n and emission of a photon $\hbar\bar{\omega}_{n12}^{(+)}$ or $\hbar\bar{\omega}_{n12}^{(-)}$, respectively, of anti-Stokes and Stokes waves. The given four-photon nonlinear process is possible only in the two-photon transition $E_b\rightarrow E_c$ in the presence of the two-photon resonance (3).

Note that in a gas with given density of active atoms Raman scattering is a coherent process and, in accordance with Maxwell's equations, is optimal in the direction in which the dispersion relation holds:

$$(\omega^{(\pm)})^2\varepsilon(\omega^{(\pm)})=(k^{(\pm)}c)^2,$$

where $\varepsilon(\omega^{(\pm)})$ is the real permittivity of the given gas. In contrast, the emission of one atom in four-photon scattering of light takes place with a definite probability in all directions and depends on the rate of change of the vectors (18) and (26) as a function of time, with allowance for $\mathbf{k}_{n12}^{(\pm)}\mathbf{r}=\mathbf{k}_{n12}^{(\pm)}\mathbf{v}t=\bar{\mathbf{k}}_{n12}^{(\pm)}\mathbf{r}=\bar{\mathbf{k}}_{n12}^{(\pm)}\mathbf{v}t$, which describe the Doppler shifts of the frequencies of the light waves.

3. OPTICAL-PRESSURE FORCE IN THE FIELD OF TWO LIGHT WAVES

If an atom is in the electric field $\mathbf{E}$ of the light waves (1), then in accordance with the well-known expression[1-3] it is acted upon by the optical-pressure force

$$\mathbf{F}=\mathrm{Tr}[\rho\nabla(\mathbf{dE})]=\sum_{n=1}^{n_0} i\mathbf{k}_n\{(\mathbf{Pa}_n)\exp[i(\mathbf{k}_n\mathbf{r}-\omega_n t)] - (\mathbf{Pa}_n^*)\exp[-i(\mathbf{k}_n\mathbf{r}-\omega_n t)]\}, \quad (32)$$

where the electric dipole moment $\mathbf{P}=\mathrm{Tr}(\rho\mathbf{d})$ of the atom is induced by the same field $\mathbf{E}$. In this problem, the density matrix ρ in the field $\mathbf{E}$ is calculated perturbatively with allowance for four terms of the series (7), but the vector $\mathbf{P}$ is equal to the sum of two terms describing the emission or absorption of photons:

$$\mathbf{P}=\mathrm{Tr}(\rho^{(1)}\mathbf{d})+\mathrm{Tr}(\rho^{(3)}\mathbf{d})=\mathbf{P}^{(1)}+\mathbf{P}^{(3)},$$

where in accordance with (9) the vector $\mathbf{P}^{(1)}$ describes the process of absorption and emission of one photon of the field (1) under nonresonance conditions with respect to the electric dipole transitions $E_b\rightarrow E_g$. In contrast, the vector $\mathbf{P}^{(3)}$ describes a complicated four-photon nonlinear process of scattering of the light by a single atom in the presence of Raman resonance (2) or two-photon resonance (3) for a two-photon transition.

We consider first of all the simplest case of two light waves (1) with $n_0=2$ that satisfy the requirement (2) or (3). In the calculation of the optical-pressure force $\mathbf{F}^{(2)}$ in the second order in the electric field $\mathbf{E}$ of the nonresonance light waves (1) we then find that the double sum over the indices of the light waves (1) in (32) with $\mathbf{P}=\mathbf{P}^{(1)}$ contains by virtue of (9) exponentials with frequencies $2\omega_1$, $\omega_1\pm\omega_2$, and $2\omega_2$, and also two terms that do not depend on $\mathbf{r}$ and t. To eliminate the rapid oscillations with the frequencies $2\omega_1$, $\omega_1\pm\omega_2$, and $2\omega_2$, we average the required force over the time as follows:

$$\langle\mathbf{F}^{(2)}\rangle=\frac{1}{t_2-t_1}\int_{t_1}^{t_2}\mathbf{F}^{(2)}dt, \quad (33)$$

where the time interval t_2-t_1 is determined by the inequalities

$$|(\mathbf{k}_1-\mathbf{k}_2)\mathbf{v}|^{-1}, \quad \gamma_{cb}^{-1}\gg t_2-t_1\gg(\omega_1-\omega_2)^{-1}.$$

Here $\omega_1-\omega_2$ is the lowest of the frequencies that occur in the considered double sum in (32). In this case, the terms of the force (32) containing the harmonic function with frequency difference $\omega_1-\omega_2$ acquire after the averaging (33) the small factor

$$[(\omega_1-\omega_2)(t_2-t_1)]^{-1}\ll 1$$

and will be omitted. For the same reason, we omit the terms of the force (32) that contain harmonic functions with frequencies greater than $\omega_1-\omega_2$. As a result of the averaging over the time (33), only the two terms that do not depend on $\mathbf{r}$ and t are unchanged. Thus, after the averaging over the time (33) the force (32) will be a rectified radiation force in the second order in the electric field $\mathbf{E}$ of the nonresonance light fields (1); specifically,

$$\mathbf{F}^{(2)}=\mathbf{F}_e^{(2)}+\mathbf{F}_o^{(2)}, \quad (34)$$

where the brackets ⟨...⟩ are omitted and we have adopted the notation

$$\mathbf{F}_e^{(2)}=-\sum_{n=1}^{2}\sum_g\frac{8\mathbf{k}_n\gamma_{bg}|\mathbf{a}_n|^2|d_{bg}|^2\omega_n\omega_{bg}}{3(2J_b+1)\hbar(\omega_n^2-\omega_{bg}^2)^2},$$

$$\mathbf{F}_o^{(2)}=\sum_{n=1}^{2}\sum_g\frac{4\mathbf{k}_n(\mathbf{k}_n\mathbf{v})\gamma_{bg}|\mathbf{a}_n|^2|d_{bg}|^2}{3(2J_b+1)\hbar}\left(\frac{1}{(\omega_n+\omega_{bg})^3}-\frac{1}{(\omega_n-\omega_{bg})^3}\right).$$

Here $\mathbf{F}_e^{(2)}$ and $\mathbf{F}_o^{(2)}$ are even and odd functions of the velocity $\mathbf{v}$ of the atom. The force (34) takes into account the effect of the light waves (1) on all the nonresonance electric dipole transitions in the second order in the electric field of these light waves.

We calculate the optical-pressure force (32) in the fourth order in the field (1) in the presence of Raman resonance (2), for which the vector $\mathbf{P}=\mathbf{P}^{(3)}$ is determined in (18). In the case $n_0=2$, the role of the test wave for Raman scattering by the atom is played by one of the two given light waves with index $n=1$ or $n=2$, and therefore $\mathbf{P}^{(3)}$ contains exponentials with four different frequencies:

$$\omega_{112}^{(+)}=2\omega_1-\omega_2, \quad \omega_{112}^{(-)}=\omega_2 \quad \omega_{212}^{(+)}=\omega_1,$$
$$\omega_{212}^{(-)}=2\omega_2-\omega_1. \quad (35)$$

In this case, it is convenient to transform expression (32) with $\mathbf{P}=\mathbf{P}^{(3)}$ to a different form that contains imaginary parts of the complex quantities:

$$\mathbf{F}^{(4)} = -2\sum_{n,n'=1}^{2} \mathbf{k}_{n'} \,\mathrm{Im}\{[\mathbf{P}_{n12}^{(+)}(\omega_{n12}^{(+)})\exp[i(\mathbf{k}_{n12}^{(+)}\mathbf{r} - \omega_{n12}^{(t)}t)] + \mathbf{P}_{n12}^{(-)}(\omega_{n12}^{(-)})\exp[i(\mathbf{k}_{n12}^{(-)}\mathbf{r} - \omega_{n12}^{(-)}t)]] \times [\mathbf{a}_{n'} \exp[i(\mathbf{k}_{n'}\mathbf{r} - \omega_{n'}t)] - \mathbf{a}_{n'}^{*} \exp[-(i\mathbf{k}_{n'}\mathbf{r} - \omega_{n'}t)]]\}, \quad (36)$$

where the scalar products of the vectors exist only in the terms within the curly brackets. The double sum in (36) contains two terms that do not depend on $\mathbf{r}$ and t, and also rapidly oscillating terms having the characteristic frequencies

$$\omega_1 \pm \omega_2, \quad 2(\omega_1 - \omega_2), \quad 2\omega_1, \quad 2\omega_2, \quad 3\omega_1 - \omega_2, \quad 3\omega_2 - \omega_1. \quad (37)$$

In this connection, we average the force (36) over the time in the same manner as (33) as in the calculation of the force (34). After the averaging of the force (36) only the constant terms, which do not depend on $\mathbf{r}$ and t, will then remain unchanged, and therefore the force (36) is transformed into the rectified radiation force

$$\mathbf{F}^{(4)} = 2[\mathbf{k}_1 \,\mathrm{Im}(\mathbf{P}_{212}^{(+)}(\omega_1)\mathbf{a}_1^{*}) + \mathbf{k}_2 \,\mathrm{Im}(\mathbf{P}_{112}^{(-)}(\omega_2)\mathbf{a}_2^{*})], \quad (38)$$

in which the vectors $\mathbf{P}_{212}^{(+)}(\omega_1)$ and $\mathbf{P}_{112}^{(-)}(\omega_2)$ are determined in (19) and (20), and the indices of these vectors are equal to the indices of the corresponding frequencies in (35).

To simplify the following calculations, we use Eqs. (11). We then obtain the useful relation

$$\Pi_\kappa(-\omega_2) = (-1)^\kappa \Pi_\kappa(\omega_1).$$

Further, for the complex amplitude $\mathbf{a}_n$ of all the considered light waves (1) we introduce the notation

$$\mathbf{a}_n = \mathbf{l}_{\mathbf{k}_n\lambda_n} R_n \exp(-i\alpha_n), \quad n = 1,\ldots,n_0, \quad (39)$$

where $\lambda_n = \pm 1$, $\mathbf{l}_{\mathbf{k}_n\lambda_n}$ is a unit complex polarization vector, R_n is a constant real amplitude, and α_n is a constant phase shift. In addition, we assume that the wave vectors $\mathbf{k}_1,\ldots,\mathbf{k}_{n_0}$ are collinear with the Cartesian axis z. For circular waves (1) the polarization vectors will then take the form

$$\mathbf{l}_{\mathbf{k}_n\lambda_n} = \frac{1}{\sqrt{2}}(\lambda_n \mathbf{l}_x + i\sigma_n \mathbf{l}_y), \quad n = 1,\ldots,n_0, \quad (40)$$

where

$$\sigma_n = (\mathbf{l}_z \mathbf{k}_n)/k_n, \quad \mathbf{l}_{-\mathbf{k}_n\lambda_n} = \mathbf{l}_{\mathbf{k}_n\lambda_n}^{*},$$

in which $\mathbf{l}_x$, $\mathbf{l}_y$, and $\mathbf{l}_z$ are unit vectors along the Cartesian axes x, y, and z. At the same time, $\lambda_n = 1$ and $\lambda_n = -1$, respectively, for right- and left-circular polarization. The polarization vector of a linearly polarized wave in (1) with amplitude (39) can also be written in a different form:

$$\mathbf{l}_n = \mathbf{l}_x \cos\varphi_n + \mathbf{l}_y \sin\varphi_n, \quad n = 1,\ldots,n_0, \quad (41)$$

where φ_n is the angle measured from the x axis clockwise if we look along the z axis.

After this procedure, the rectified optical-pressure force (38) for Raman resonance (2) takes the form

$$\mathbf{F}^{(4)} = \frac{(\mathbf{k}_1 - \mathbf{k}_2)\hbar\gamma_{cb}^3 C_{12}}{[\Delta_{12} - (\mathbf{k}_1 - \mathbf{k}_2)\mathbf{v}]^2 + \gamma_{cb}^2}, \quad (42)$$

where

$$C_{12} = \frac{1}{2J_b + 1}\left(\frac{R_1 R_2}{\hbar\gamma_{cb}}\right)^2 \left[\left|\Pi_1(\omega_1)\right|^2 + \left|\Pi_2(\omega_1)\right|^2 + (|\Pi_2(\omega_1)|^2 - |\Pi_1(\omega_1)|^2)(\mathbf{l}_{\mathbf{k}_1\lambda_1}\mathbf{l}_{\mathbf{k}_2\lambda_2})(\mathbf{l}_{\mathbf{k}_1\lambda_1}^{*}\mathbf{l}_{\mathbf{k}_2\lambda_2}^{*}) + \frac{2}{3}(|\Pi_0(\omega_1)|^2 - |\Pi_2(\omega_1)|^2)(\mathbf{l}_{\mathbf{k}_1\lambda_1}\mathbf{l}_{\mathbf{k}_2\lambda_2}^{*}) \times (\mathbf{l}_{\mathbf{k}_1\lambda_1}^{*}\mathbf{l}_{\mathbf{k}_2\lambda_2})\right],$$

$$\Delta_{12} = \omega_1 - \omega_2 - \omega_{cb}. \quad (43)$$

In its physical nature, the force (42) is associated with four-photon Raman scattering by the atom, and it therefore has different properties compared with the force (34). The dimensionless quantity C_{12} depends essentially on the angular momenta of the degenerate levels and the polarizations of the light waves. Depending on the directions of $\mathbf{k}_1$ and $\mathbf{k}_2$, it contains $|\Pi_\kappa(\omega_1)|^2$ with different κ. For example, for parallel $\mathbf{k}_1$ and $\mathbf{k}_2$ and different circular polarizations (40), we have $\mathbf{l}_{\mathbf{k}_1\lambda_1}\mathbf{l}_{\mathbf{k}_2\lambda_2} = 1$ and $\mathbf{l}_{\mathbf{k}_1\lambda_1}\mathbf{l}_{\mathbf{k}_2\lambda_2}^{*} = 0$, and therefore (43) contains only $|\Pi_2(\omega_1)|^2$. At the same time, for orthogonal linear polarizations in (43) we must set

$$\mathbf{l}_{\mathbf{k}_1\lambda_1} \to \mathbf{l}_1, \quad \mathbf{l}_{\mathbf{k}_2\lambda_2} \to \mathbf{l}_2, \quad \mathbf{l}_1\mathbf{l}_2 = 0,$$

and then the dimensionless C_{12} contains only the sum $|\Pi_1(\omega)|^2 + |\Pi_2(\omega_1)|^2$, and the moduli of the quantities $\Pi_\kappa(\omega_1)$ with $\kappa = 0,1,2$ or their ratios can be determined from Raman scattering in both the stationary[30,31] and nonstationary[32–34] regimes. Experimental methods of determining these quantities are presented in Ref. 31. The theoretical calculation of $\Pi_\kappa(\omega_1)$ is a laborious problem, because it must deal with the reduced dipole moments of the atom.

The force (42) can be decomposed into two terms:

$$\mathbf{F}^{(4)} = \mathbf{F}_e^{(4)} + \mathbf{F}_o^{(4)},$$

where

$$\mathbf{F}_e^{(4)} = \frac{(\mathbf{k}_1 - \mathbf{k}_2)\hbar\gamma_{cb}^3 C_{12}[\Delta_{12}^2 + ((\mathbf{k}_1 - \mathbf{k}_2)\mathbf{v})^2 + \gamma_{cb}^2]}{\{[\Delta_{12} + (\mathbf{k}_1 - \mathbf{k}_2)\mathbf{v}]^2 + \gamma_{cb}^2\}\{[\Delta_{12} - (\mathbf{k}_1 - \mathbf{k}_2)\mathbf{v}]^2 + \gamma_{cb}^2\}}, \quad (44)$$

$$\mathbf{F}_o^{(4)} = \frac{2(\mathbf{k}_1 - \mathbf{k}_2)\hbar\gamma_{cb}^3 C_{12}\Delta_{12}((\mathbf{k}_1 - \mathbf{k}_2)\mathbf{v})}{\{[\Delta_{12} + (\mathbf{k}_1 - \mathbf{k}_2)\mathbf{v}]^2 + \gamma_{cb}^2\}\{[\Delta_{12} - (\mathbf{k}_1 - \mathbf{k}_2)\mathbf{v}]^2 + \gamma_{cb}^2\}}. \quad (45)$$

The first, (44), and second, (45), terms of the force $\mathbf{F}^{(4)}$ are even and odd functions of the velocity $\mathbf{v}$ of the atom. The direction of the first term (44) is determined by the vector

$\mathbf{k}_1-\mathbf{k}_2$. In contrast, the second term (45) is parallel or antiparallel to $\mathbf{k}_1-\mathbf{k}_2$, depending on the signs of Δ_{12} and the projection of the velocity $\mathbf{v}$ onto the direction $\mathbf{k}_1-\mathbf{k}_2$. Thus, the second vector term (45) is collinear to $\mathbf{k}_1-\mathbf{k}_2$ and, depending on the sign of Δ_{12}, can be either a decelerating or accelerating force for an atom moving with velocity longitudinal with respect to $\mathbf{k}_1-\mathbf{k}_2$:

$$v_z=\mathbf{v}(\mathbf{k}_1-\mathbf{k}_2)|k_1-k_2|^{-1}.$$

In the case of the two-photon resonance (3), the general expressions (32), (33), and (39)–(41) remain, and for $\mathbf{P}^{(3)}$ we can use the expression (26) with $n_0=2$. Repeating the arguments as for Raman resonance, we obtain the optical-pressure force (32) in the presence of two-photon resonance (3) in the form

$$\bar{\mathbf{P}}^{(4)}=-2\sum_{n,n'=1}^{2}\mathbf{k}_{n'}\ \mathrm{Im}\{[\bar{\mathbf{P}}_{n12}^{(+)}(\bar{\omega}_{n12}^{(+)})\exp[i(\bar{\mathbf{k}}_{n12}^{(+)}\mathbf{r}-\bar{\omega}_{n12}^{(t)}t)]+\bar{\mathbf{P}}_{n12}^{(-)}(\bar{\omega}_{n12}^{(-)})\exp[i(\bar{\mathbf{k}}_{n12}^{(-)}\mathbf{r}-\bar{\omega}_{n12}^{(-)}t)]]\times[\mathbf{a}_{n'}\exp[i(\mathbf{k}_{n'}\mathbf{r}-\omega_{n'}t)]-\mathbf{a}_{n'}^{*}\exp[-i(\mathbf{k}_{n'}\mathbf{r}-\omega_{n'}t)]]\}. \tag{46}$$

Since the atom is acted on by the two light waves (1) with indices $n=1$ and $n=2$, the vector (26) contains exponentials with four different frequencies:

$$\bar{\omega}_{112}^{(+)}=2\omega_1+\omega_2,\quad \bar{\omega}_{112}^{(-)}=-\omega_2,$$
$$\bar{\omega}_{212}^{(+)}=\omega_1+2\omega_2,\quad \bar{\omega}_{212}^{(-)}=-\omega_1. \tag{47}$$

Therefore, the double sum in (46) over the indices n and n' contains two constant terms, which do not depend on $\mathbf{r}$ and t, and several rapidly oscillating terms, for which the lowest frequency is $\omega_1-\omega_2$. This makes it possible to average the double sum (46) over the time, (33), as for Raman resonance. As a result, the force (46) is reduced to the sum of two terms:

$$\bar{\mathbf{F}}^{(4)}=-2[\mathbf{k}_1\ \mathrm{Im}(\bar{\mathbf{P}}_{212}^{(-)}(-\omega_1)\mathbf{a}_1)+\mathbf{k}_2\ \mathrm{Im}(\bar{\mathbf{P}}_{112}^{(-)}(-\omega_2)\mathbf{a}_2)], \tag{48}$$

where the vectors $\bar{\mathbf{P}}_{212}^{(-)}(-\omega_1)$ and $\bar{\mathbf{P}}_{112}^{(-)}(-\omega_2)$ are determined by (28) with $n=1$, 2, and the indices of these vectors are equal to the indices of the corresponding frequencies in (47).

For the two-photon resonance (3), we have by virtue of Eqs. (30) the relation

$$\Pi_\kappa(\omega_2)=(-1)^\kappa\Pi_\kappa(\omega_1),$$

which makes it possible to represent the rectified optical-pressure force (48) in the compact form

$$\bar{\mathbf{F}}^{(4)}=\frac{(\mathbf{k}_1+\mathbf{k}_2)\hbar\gamma_{cb}^3\bar{C}_{12}}{[\bar{\Delta}_{12}-(\mathbf{k}_1+\mathbf{k}_2)\mathbf{v}]^2+\gamma_{cb}^2}, \tag{49}$$

where

$$\bar{C}_{12}=\frac{1}{2J_b+1}\left(\frac{R_1R_2}{\hbar\gamma_{cb}}\right)^2\Big[|\Pi_1(\omega_1)|^2+|\Pi_2(\omega_1)|^2+(|\Pi_2(\omega_1)|^2-|\Pi_1(\omega_1)|^2)(\mathbf{l}_{\mathbf{k}_1\lambda_1}\mathbf{l}_{\mathbf{k}_2\lambda_2}^{*})(\mathbf{l}_{k_1\lambda_1}^{*}\mathbf{l}_{\mathbf{k}_2\lambda_2})+\frac{2}{3}(|\Pi_0(\omega_1)|^2-|\Pi_2(\omega_1)|^2)(\mathbf{l}_{\mathbf{k}_1\lambda_1}\mathbf{l}_{\mathbf{k}_2\lambda_2})(\mathbf{l}_{\mathbf{k}_1\lambda_1}^{*}\mathbf{l}_{\mathbf{k}_2\lambda_2}^{*})\Big],$$
$$\bar{\Delta}_{12}=\omega_1+\omega_2-\omega_{cb}. \tag{50}$$

The rectified optical-pressure force (49) is due to four-photon scattering of the light waves by the atom in the presence of the two-photon resonance (3). It can also be decomposed into two terms:

$$\bar{\mathbf{F}}^{(4)}=\bar{\mathbf{F}}_e^{(4)}+\bar{\mathbf{F}}_o^{(4)},$$

where

$$\bar{\mathbf{F}}_e^{(4)}=\frac{(\mathbf{k}_1+\mathbf{k}_2)\hbar\gamma_{cb}^3\bar{C}_{12}[\bar{\Delta}_{12}^2+((\mathbf{k}_1+\mathbf{k}_2)\mathbf{v})^2+\gamma_{cb}^2]}{[(\bar{\Delta}_{12}+(\mathbf{k}_1+\mathbf{k}_2)\mathbf{v})^2+\gamma_{cb}^2][(\bar{\Delta}_{12}-(\mathbf{k}_1+\mathbf{k}_2)\mathbf{v})^2+\gamma_{cb}^2]}, \tag{51}$$

$$\mathbf{F}_o^{(4)}=\frac{2(\mathbf{k}_1+\mathbf{k}_2)\hbar\gamma_{cb}^3\bar{C}_{12}\bar{\Delta}_{12}((\mathbf{k}_1+\mathbf{k}_2)\mathbf{v})}{[(\bar{\Delta}_{12}+(\mathbf{k}_1+\mathbf{k}_2)\mathbf{v})^2+\gamma_{cb}^2][(\bar{\Delta}_{12}-(\mathbf{k}_1+\mathbf{k}_2)\mathbf{v})^2+\gamma_{cb}^2]}. \tag{52}$$

The term (51) is an even function of $\mathbf{v}$, and the other term (52) is an odd function of the velocity $\mathbf{v}$. It can be seen that the forces (44) and (45) for Raman resonance (2) go over into the forces (51) and (52) for two-photon resonance (3) after the substitutions

$$\mathbf{k}_{12}\to-\mathbf{k}_2,\quad \Delta_{12}\to\bar{\Delta}_{12},\quad C_{12}\to\bar{C}_{12}. \tag{53}$$

4. OPTICAL-PRESSURE FORCE IN A FIELD OF FOUR LIGHT WAVES

In accordance with the perturbative expansion (7), the forces (42) and (49) of fourth order in the field (1) are less than the forces (34) of second order in the field (1) in the ratio

$$\frac{|\mathbf{F}_e^{(4)}|}{|\mathbf{F}_e^{(2)}|}\sim\left(\frac{R_1|d_{bg}|}{\hbar\gamma_{cb}}\right)^2\ll 1, \tag{54}$$

where for an estimate of the quantities we have assumed $R_1\sim R_2$, $\omega_1-\omega_{bg}\sim\omega_1$, $\omega_2\lesssim\omega_1$, and $\gamma_{bg}\sim\gamma_{cb}$. The inequality (54) is due to the term $\mathbf{F}_e^{(2)}$ of the force (34), which is even with respect to the velocity $\mathbf{v}$ of the atom, since for nonresonance light waves it exceeds the odd term $\mathbf{F}_o^{(2)}$ in the ratio

$$\frac{|\mathbf{F}_e^{(2)}|}{|\mathbf{F}_e^{(2)}|}\sim\frac{\omega_1}{|\mathbf{k}_l\mathbf{v}|}\gg 1.$$

However, the terms even in $\mathbf{v}$ in the forces (34), (42), and (49) are undesirable in problems of the laser cooling of atoms and the localization of atoms by light fields. These terms can be eliminated by using four light waves (1) with $n_0=4=$ which form two pairs of oppositely directed waves as follows:

$$\omega_1=\omega_3,\quad R_1=R_3,\quad \mathbf{k}_1=-\mathbf{k}_3,\quad \mathbf{l}_{\mathbf{k}_1\lambda_1}=\mathbf{l}^*_{\mathbf{k}_3\lambda_3},$$

$$\lambda_1=\lambda_3=\pm 1, \tag{55}$$

$$\omega_2=\omega_4,\quad R_2=R_4,\quad \mathbf{k}_2=-\mathbf{k}_4,\quad \mathbf{l}_{\mathbf{k}_2\lambda_2}=\mathbf{l}^*_{\mathbf{k}_4\lambda_4},$$

$$\lambda_2=\lambda_4=\pm 1, \tag{56}$$

where the difference $\omega_1-\omega_2$ and the sum $\omega_1+\omega_2$ satisfy the conditions of Raman (2) or two-photon (3) resonances, whereas the frequencies ω_n and $2\omega_n$ with $n=1,2,3,4$ are nonresonant with respect to all the atomic transitions between levels of the discrete spectrum of the atom. The phase shifts α_n with $n=1,2,3,4$ of the given light waves are arbitrary. In accordance with (40), each pair of oppositely directed circular waves (55) and (56) has the same circular polarizations, right-circular for $\lambda_1=\lambda_2=\lambda_3=\lambda_4=1$ and left-circular for $\lambda_1=\lambda_2=\lambda_3=\lambda_4=-1$. These polarizations of the oppositely directed circular waves are chosen to make the sum of the $\mathbf{v}$-even terms in the force $\mathbf{F}^{(2)}$ of second order in the field (1) with $n_0=4$ vanish. For this purpose, one can also use the orthogonal linear polarizations (41) for two pairs of oppositely directed waves with the same frequency: $\mathbf{l}_1\cdot\mathbf{l}_3=\mathbf{l}_2\cdot\mathbf{l}_4=0$.

When the requirements (55) and (56) are satisfied in the presence of Raman resonance (2), a solution of Eq. (4) can be obtained by the method presented in Sec. 2. The previously obtained density matrix (8) is unchanged, and the resonance component of the density matrix $\rho^{(2)}(t)$ has the form

$$\rho^{(2)}_{M_cM_b}=\rho^{(2)}_{M_cM_b}(1,2)+\rho^{(2)}_{M_cM_b}(3,4)+\rho^{(2)}_{M_cM_b}(1,4)+\rho^{(2)}_{M_cM_b}(3,2),$$

where the numbers 1, 2, 3, and 4 are equal to the indices n and m of the amplitudes and frequencies in the general expression

$$\rho^{(2)}_{M_cM_b}(n,m)=\frac{-1}{(2J_b+1)\hbar^2[\omega_n-\omega_m-(\mathbf{k}_n-\mathbf{k}_m)\mathbf{v}-\omega_{cb}+i\gamma_{cb}]}\times\sum_g\left(\frac{(\mathbf{a}_n\mathbf{d}_{M_cM_g})(\mathbf{a}^*_m\mathbf{d}_{M_gM_b})}{\omega_n-\omega_{cg}}-\frac{(\mathbf{a}^*_m\mathbf{d}_{M_cM_g})(\mathbf{a}_n\mathbf{d}_{M_gM_b})}{\omega_n-\omega_{gb}}\right)\times\exp\{i[(\mathbf{k}_n-\mathbf{k}_m)\mathbf{r}-(\omega_n-\omega_m)t]\}.$$

Further calculations in the presence of the Raman resonance (2) lead to the following induced electric dipole moment of the atom in the third order in the field (1):

$$\begin{aligned}\mathbf{P}^{(3)}=\sum_{n=1}^{4}\{&\mathbf{P}^{(+)}_{n12}(\omega^{(+)}_{n12})\exp[i(\mathbf{k}^{(+)}_{n12}\mathbf{r}-\omega^{(+)}_{n12}t)]+\mathbf{P}^{(-)}_{n12}\\&\times(\omega^{(-)}_{n12})\exp[i(\mathbf{k}^{(-)}_{n12}\mathbf{r}-\omega^{(-)}_{n12}t)]+\mathbf{P}^{(+)}_{n34}\\&\times(\omega^{(+)}_{n34})\exp[i(\mathbf{k}^{(+)}_{n34}\mathbf{r}-\omega^{(+)}_{n34}t)]+\mathbf{P}^{(-)}_{n34}\\&\times(\omega^{(-)}_{n34})\exp[i(\mathbf{k}^{(-)}_{n34}\mathbf{r}-\omega^{(-)}_{n34}t)]+\mathbf{P}^{(+)}_{n14}\\&\times(\omega^{(+)}_{n14})\exp[i(\mathbf{k}^{(+)}_{n14}\mathbf{r}-\omega^{(+)}_{n14}t)]+\mathbf{P}^{(-)}_{n14}\\&\times(\omega^{(-)}_{n14})\exp[i(\mathbf{k}^{(-)}_{n14}\mathbf{r}-\omega^{(-)}_{n14}t)]+\mathbf{P}^{(+)}_{n32}\\&\times(\omega^{(+)}_{n32})\exp[i(\mathbf{k}^{(+)}_{n32}\mathbf{r}-\omega^{(+)}_{n32}t)]+\mathbf{P}^{(-)}_{n32}\\&\times(\omega^{(-)}_{n32})\exp[i(\mathbf{k}^{(-)}_{n32}\mathbf{r}-\omega^{(-)}_{n32}t)]\}+\text{c.c.},\end{aligned} \tag{57}$$

where the first two terms in (57) are determined by the expressions (18)–(25) with $n_0=4$. The remaining six terms in (57) can be obtained from the first two with allowance for the expressions (18)–(25) if in the vectors $\mathbf{P}^{(\pm)}_{n12}$, $\mathbf{k}^{(\pm)}_{n12}$, $\mathbf{a}_1$, $\mathbf{a}_2$, $\mathbf{a}^*_1$, $\mathbf{a}^*_2$ and the frequencies $\omega^{(\pm)}_{n12}$ we make the following three index substitutions: 1) $1\to 3$ and $2\to 4$, 2) $1\to 1$ and $2\to 4$, 3) $1\to 3$ and $2\to 2$.

In accordance with the general expression (32) with $n_0=4$ and $\mathbf{P}=\mathbf{P}^{(3)}$ (57), the required force $\mathbf{F}^{(4)}$ of fourth order in the field (1) with $n_0=4$ contains numerous terms of a double sum over the indices n and n' similar to the terms in (36). These terms can be divided into three essentially different groups. The first group contains constant terms that do not depend on $\mathbf{r}$ and t that were obtained by multiplying the exponentials in (57) with the characteristic frequencies

$$\omega^{(+)}_{212}=\omega_1,\quad \omega^{(-)}_{112}=\omega_2,\quad \omega^{(+)}_{434}=\omega_3,\quad \omega^{(-)}_{334}=\omega_4,$$

$$\omega^{(+)}_{414}=\omega_1,\quad \omega^{(-)}_{114}=\omega_4,\quad \omega^{(+)}_{232}=\omega_3,\quad \omega^{(-)}_{332}=\omega_2 \tag{58}$$

by the corresponding exponentials of the light waves (1) with $n_0=4$ having the form

$$\mathbf{a}^*_n\exp[-i(\mathbf{k}_n\mathbf{r}-\omega_nt)],\quad n=1,2,3,4. \tag{59}$$

The second group of terms was formed by multiplying the exponentials in (57) with characteristic frequencies

$$\omega^{(+)}_{412}=\omega_1,\quad \omega^{(-)}_{312}=\omega_2,\quad \omega^{(+)}_{234}=\omega_3,\quad \omega^{(-)}_{134}=\omega_4,$$

$$\omega^{(+)}_{214}=\omega_1,\quad \omega^{(-)}_{314}=\omega_4,\quad \omega^{(+)}_{432}=\omega_3,\quad \omega^{(-)}_{132}=\omega_2 \tag{60}$$

by the exponentials (59) of the same light waves (1). As a result of this multiplication, there appear in the force $\mathbf{F}^{(4)}$ terms which do not contain the frequencies (60) multiplied by t but which depend on the coordinates through the factors $\exp(\pm 2i\mathbf{k}_1\mathbf{r})$ and $\exp(\pm 2i\mathbf{k}_2\mathbf{r})$. This second group of terms is undesirable, since it hinders rectification of the radiation force $\mathbf{F}^{(4)}$. Fortunately, they can be eliminated if the polarizations of the circular waves are, with allowance for (55) and (56), chosen as follows:

$$\mathbf{l}_{\mathbf{k}_1\lambda_1}=\frac{1}{\sqrt{2}}(\lambda_1\mathbf{l}_x+i\mathbf{l}_y),\quad \mathbf{l}_{\mathbf{k}_2,-\lambda_1}=\frac{1}{\sqrt{2}}(-\lambda_1\mathbf{l}_x+i\mathbf{l}_y),$$

$$\mathbf{l}_{\mathbf{k}_3\lambda_1}=\frac{1}{\sqrt{2}}(\lambda_1\mathbf{l}_x-i\mathbf{l}_y),\quad \mathbf{l}_{\mathbf{k}_4,-\lambda_1}=\frac{1}{\sqrt{2}}(-\lambda_1\mathbf{l}_x-i\mathbf{l}_y),$$

$$\lambda_1=\pm 1. \tag{61}$$

In the case of linearly polarized waves, we choose the polarization vectors [Eq. (41)] with the same aim such that

$$\mathbf{l}_1=\mathbf{l}_2,\quad \mathbf{l}_3=\mathbf{l}_4,\quad \mathbf{l}_1\cdot\mathbf{l}_3=\mathbf{l}_2\cdot\mathbf{l}_4=0. \tag{62}$$

For the polarization vectors (61) or (62), the terms of the second group vanish by virtue of the equations

$$\begin{aligned}\mathbf{P}^{(+)}_{412}(\omega_1)\mathbf{a}_1^*&=\mathbf{P}^{(-)}_{312}(\omega_2)\mathbf{a}_2^*=\mathbf{P}^{(+)}_{234}(\omega_3)\mathbf{a}_3^*=\mathbf{P}^{(-)}_{134}(\omega_4)\mathbf{a}_4^*\\&=\mathbf{P}^{(+)}_{214}(\omega_1)\mathbf{a}_1^*=\mathbf{P}^{(-)}_{314}(\omega_4)\mathbf{a}_4^*=\mathbf{P}^{(+)}_{432}(\omega_3)\mathbf{a}_3^*\\&=\mathbf{P}^{(-)}_{132}(\omega_2)\mathbf{a}_2^*=0.\end{aligned}$$

The third group of terms contains rapidly oscillating harmonic functions with frequencies (37). Performing the time averaging (33) of the required force in the same way as in the calculation of the force (34), we see that the terms of the third group can be ignored. Thus, a contribution to the force $\mathbf{F}^{(4)}$ is made by only the constant terms of the first group that do not depend on $\mathbf{r}$ and t; taken together, these terms constitute the following rectified radiation force:

$$\begin{aligned}\mathbf{F}^{(4)}=2\,\mathrm{Im}[&\mathbf{k}_1(\mathbf{P}^{(+)}_{212}(\omega_1)\mathbf{a}_1^*)+\mathbf{k}_2(\mathbf{P}^{(-)}_{112}(\omega_2)\mathbf{a}_2^*)\\&+\mathbf{k}_3(\mathbf{P}^{(+)}_{434}(\omega_3)\mathbf{a}_3^*)+\mathbf{k}_4(\mathbf{P}^{(-)}_{334}(\omega_4)\mathbf{a}_4^*)\\&+\mathbf{k}_1(\mathbf{P}^{(+)}_{414}(\omega_1)\mathbf{a}_1^*)+\mathbf{k}_4(\mathbf{P}^{(-)}_{114}(\omega_4)\mathbf{a}_4^*)\\&+\mathbf{k}_3(\mathbf{P}^{(+)}_{232}(\omega_3)\mathbf{a}_3^*)+\mathbf{k}_2(\mathbf{P}^{(-)}_{332}(\omega_2)\mathbf{a}_2^*)],\end{aligned} \tag{63}$$

where the indices of the vectors $\mathbf{P}^{(\pm)}$ are equal to the indices of the characteristic frequencies (58). The force (63) can be decomposed into parts even and odd with respect to the velocity $\mathbf{v}$. However, by virtue of the choice of the light waves (55) and (56) the sum of the $\mathbf{v}$-even terms vanishes, and there remain only the $\mathbf{v}$-odd terms, which form a radiation force that plays the role of a decelerating or accelerating force, depending on the sign of the detuning from Raman resonance. For the chosen polarization vectors (61) and (62), it has the form

$$\begin{aligned}\mathbf{F}_0^{(4)}=&\frac{4(\mathbf{k}_1-\mathbf{k}_2)\hbar\gamma_{cb}^3C_{12}\Delta_{12}((\mathbf{k}_1-\mathbf{k}_2)\mathbf{v})}{\{[\Delta_{12}+(\mathbf{k}_1-\mathbf{k}_2)\mathbf{v}]^2+\gamma_{cb}^2\}\{[\Delta_{12}-(\mathbf{k}_1-\mathbf{k}_2)\mathbf{v}]^2+\gamma_{cb}^2\}}\\&+\frac{4(\mathbf{k}_1+\mathbf{k}_2)\hbar\gamma_{cb}^3C_{14}\Delta_{12}((\mathbf{k}_1+\mathbf{k}_2)\mathbf{v})}{\{[\Delta_{12}+(\mathbf{k}_1+\mathbf{k}_2)\mathbf{v}]^2+\gamma_{cb}^2\}\{[\Delta_{12}-(\mathbf{k}_1+\mathbf{k}_2)\mathbf{v}]^2+\gamma_{cb}^2\}},\end{aligned} \tag{64}$$

where the dimensionless quantity C_{14} is determined by the expression (43), in which we have made the substitution

$$\mathbf{l}_{\mathbf{k}_2\lambda_2}\to\mathbf{l}^*_{\mathbf{k}_2,-\lambda_1},\quad \mathbf{l}^*_{\mathbf{k}_2\lambda_2}\to\mathbf{l}_{\mathbf{k}_2,-\lambda_1}. \tag{65}$$

For circular waves with the polarization vectors (61), we obtain

$$C_{12}=\frac{2}{2J_b+1}\left(\frac{R_1R_2}{\hbar\gamma_{cb}}\right)^2|\Pi_2(\omega_1)|^2, \tag{66}$$

$$C_{14}=\frac{2}{2J_b+1}\left(\frac{R_1R_2}{\hbar\gamma_{cb}}\right)^2\left(\frac{2}{3}|\Pi_0(\omega_1)|^2+|\Pi_1(\omega_1)|^2+\frac{1}{3}|\Pi_2(\omega_1)|^2\right), \tag{67}$$

whereas for the linear polarizations (62) we find

$$C_{12}=\frac{2}{3(2J_b+1)}\left(\frac{R_1R_2}{\hbar\gamma_{cb}}\right)^2(|\Pi_0(\omega_1)|^2+2|\Pi_2(\omega_1)|^2), \tag{68}$$

$$C_{14}=\frac{1}{2J_b+1}\left(\frac{R_1R_2}{\hbar\gamma_{cb}}\right)^2(|\Pi_1(\omega_1)|^2+|\Pi_2(\omega_1)|^2). \tag{69}$$

For small longitudinal velocities, i.e., for

$$|(\mathbf{k}_1\pm\mathbf{k}_2)\mathbf{v}|\ll\gamma_{cb}, \tag{70}$$

the radiation force (64) takes the form

$$\mathbf{F}_0^{(4)}=\frac{4[(\mathbf{k}_1-\mathbf{k}_2)((\mathbf{k}_1-\mathbf{k}_2)\mathbf{v})C_{12}+(\mathbf{k}_1+\mathbf{k}_2)((\mathbf{k}_1+\mathbf{k}_2)\mathbf{v})C_{14}]\Delta_{12}\hbar\gamma_{cb}^3}{(\Delta_{12}^3+\gamma_{cb}^2)^2}. \tag{71}$$

In the case of four light waves (55) and (56), which satisfy the two-photon resonance (3), the arguments are the same as for Raman resonance. As a result, we obtain an optical-pressure force in the fourth order in the field (1) with $n_0=4$ which contains only terms odd with respect to $\mathbf{v}$ and which has the form

$$\begin{aligned}\bar{\mathbf{F}}_0^{(4)}=&\frac{4(\mathbf{k}_1+\mathbf{k}_2)\hbar\gamma_{cb}^3\bar{C}_{12}\bar{\Delta}_{12}((\mathbf{k}_1+\mathbf{k}_2)\mathbf{v})}{\{[\bar{\Delta}_{12}+(\mathbf{k}_1+\mathbf{k}_2)\mathbf{v}]^2+\gamma_{cb}^2\}\{[\bar{\Delta}_{12}-(\mathbf{k}_1+\mathbf{k}_2)\mathbf{v}]^2+\gamma_{cb}^2\}}\\&+\frac{4(\mathbf{k}_1-\mathbf{k}_2)\hbar\gamma_{cb}^3\bar{C}_{14}\bar{\Delta}_{12}((\mathbf{k}_1-\mathbf{k}_2)\mathbf{v})}{\{[\bar{\Delta}_{12}+(\mathbf{k}_1-\mathbf{k}_2)\mathbf{v}]^2+\gamma_{cb}^2\}\{[\bar{\Delta}_{12}-(\mathbf{k}_1-\mathbf{k}_2)\mathbf{v}]^2+\gamma_{cb}^2\}},\end{aligned} \tag{72}$$

where the dimensionless quantity $\bar{C}_{14}$ is given by the expression (50), in which the substitution (65) is made. For the chosen circular polarizations (61), we obtain

$$\bar{C}_{12}=C_{14},\quad \bar{C}_{14}=C_{12}. \tag{73}$$

At the same time, for the linear polarizations (62) we have

$$\bar{C}_{12}=C_{12},\quad \bar{C}_{14}=C_{14}. \tag{74}$$

For low longitudinal velocities (70), the force (72) is

$$\mathbf{F}_0^{(4)}=\frac{4[(\mathbf{k}_1+\mathbf{k}_2)((\mathbf{k}_1+\mathbf{k}_2)\mathbf{v})\bar{C}_{12}+(\mathbf{k}_1-\mathbf{k}_2)((\mathbf{k}_1-\mathbf{k}_2)\mathbf{v})\bar{C}_{14}]\bar{\Delta}_{12}\hbar\gamma_{cb}^3}{(\bar{\Delta}_{12}^2+\gamma_{cb}^2)^2}. \tag{75}$$

The forces (64) and (71) transform into the forces (72) and (75) under the substitutions

$$\mathbf{k}_2\rightarrow-\mathbf{k}_2,\quad \Delta_{12}\rightarrow\bar{\Delta}_{12},\quad C_{12}\rightarrow\bar{C}_{12},\quad C_{14}\rightarrow\bar{C}_{14},$$

which largely repeat (53).

The first term in (72) describes a sum of identical contributions of two oppositely directed pairs of light waves with indices $n=1$, 2 and $n=3$, 4, for which $\omega_1+\omega_2=\omega_3+\omega_4\approx\omega_{cb}$ and $\mathbf{k}_1+\mathbf{k}_2=-\mathbf{k}_3-\mathbf{k}_4$. Therefore, the first term in (72) is due to the emission and absorption by the atom of two photons with total frequency $\omega_1+\omega_2-(\mathbf{k}_1+\mathbf{k}_2)\cdot\mathbf{v}$ or two photons with total frequency $\omega_3+\omega_4-(\mathbf{k}_3+\mathbf{k}_4)\cdot\mathbf{v}$ in the coordinate system attached to the moving atom. Similarly, the second term in (72) is the sum of identical contributions of two oppositely directly pairs of light waves with indices $n=1$, 4 and $n=3$, 2, for which $\omega_1+\omega_4=\omega_3+\omega_2\approx\omega_{cb}$ and $\mathbf{k}_1+\mathbf{k}_4=-\mathbf{k}_3-\mathbf{k}_2=\mathbf{k}_1-\mathbf{k}_2$. It can be seen that the second term in (72) is due to the emission and absorption by the atom of two photons with total frequency $\omega_1+\omega_4-(\mathbf{k}_1+\mathbf{k}_4)\cdot\mathbf{v}$ or two photons with total frequency $\omega_3+\omega_2-(\mathbf{k}_3+\mathbf{k}_2)\cdot\mathbf{v}$, where $\mathbf{k}_4=-\mathbf{k}_2$ and $\mathbf{k}_3=-\mathbf{k}_1$. The same interpretation of the terms can be given for the optical-pressure force (64) in the case of the Raman resonance (2).

Since for the chosen light waves (55) and (56) the optical-pressure forces of second, $\mathbf{F}_o^{(2)}$, and fourth, $\mathbf{F}_o^{(4)}$, orders in the field $\mathbf{E}$ contain only parts that are odd with respect to $\mathbf{v}$, their ratio is given in order of magnitude by the inequality

$$\frac{|\mathbf{F}_o^{(2)}|}{|\mathbf{F}_o^{(4)}|}\sim\left(\frac{\hbar\gamma_{cb}}{R_1|d_{bg}|}\right)^2\frac{\gamma_{cb}^2}{\omega_1|\Delta_{12}|}\ll 1,$$

where $|\Delta_{12}|\gtrsim\gamma_{cb}$.

This means that for the four light waves (55) and (56) the nonresonance optical-pressure forces $\mathbf{F}^{(2)}$ of second order in the field (1) are unimportant and are to be omitted. Then the principal optical-pressure force in the case of Raman, (2), or two-photon, (3), resonance is the $\mathbf{v}$-odd rectified radiation force of fourth order in the field (1) given by the expressions (64) and (72).

5. RESONANCE STRUCTURE OF THE RECTIFIED RADIATION FORCE

In the case of the two-photon resonance (3), we set

$$\mathbf{k}_1+\mathbf{k}_2=\mathbf{l}_z(k_1+k_2),\quad \omega_1=3\omega_2,\quad v_z=\mathbf{l}_z\mathbf{v}.$$

Using (73), (74), and (66)–(69), we can therefore represent the optical-pressure force (72) in the dimensionless form:

$$F_z=xy\left\{\frac{4}{[1+(4x+y)^2][1+(4x-y)^2]}+\frac{z}{[1+(2x+y)^2][1+(2x-y)^2]}\right\}, \tag{76}$$

where

$$F_z=\mathbf{l}_z\mathbf{F}_0^{(4)}/F_{12},\quad F_{12}=16\hbar k_2\gamma_{cb}\bar{C}_{12},$$

$$x=k_2v_z/\gamma_{cb},\quad y=\bar{\Delta}_{12}/\gamma_{cb},\quad z=\bar{C}_{14}/\bar{C}_{12}.$$

For the circular polarizations (61), we obtain

$$z=\left|\frac{\Pi_0(\omega_1)}{\Pi_2(\omega_1)}\right|^2\left(\frac{1}{3}+\frac{1}{2}\left|\frac{\Pi_1(\omega_1)}{\Pi_0(\omega_1)}\right|^2+\frac{1}{6}\left|\frac{\Pi_2(\omega_1)}{\Pi_0(\omega_1)}\right|^2\right),$$

whereas for the linear polarizations (62)

$$z=\frac{3}{2}\left(\left|\frac{\Pi_1(\omega_1)}{\Pi_0(\omega_1)}\right|^2+\left|\frac{\Pi_2(\omega_1)}{\Pi_0(\omega_1)}\right|^2\right)\left(1+2\left|\frac{\Pi_2(\omega_1)}{\Pi_0(\omega_1)}\right|^2\right)^{-1},$$

where the parameters $|\Pi_\kappa(\omega_1)/\Pi_0(\omega_1)|$ with $\kappa=1$, 2 can be found from experiments on Raman scattering, as was done in Ref. 32 with allowance for the equation $\Pi_\kappa(\omega_1,\omega_2)=\Pi_\kappa(\omega_1)$ for Raman resonance and $\Pi_\kappa(\omega_1,-\omega_2)=\Pi_\kappa(\omega_1)$ for two-photon resonance. In the experiment of Ref. 33, the ratio $|\Pi_1(\omega)/\Pi_2(\omega)|$ was determined from the depth of the beats of the nonstationary Raman scattering.

Figures 2 and 3 show the dependences of the optical-pressure force (76) on the velocity v_z for some values of the parameters y and z. These dependences can be used to investigate the contribution of two-photon resonances. It can be seen from the figures that the radiation force contains four two-photon resonances: one for each pair of light waves with indices $n=1,2$, $n=3,4$, $n=1,4$, and $n=3,2$. In the rest frame of the atom, the frequencies of the light waves are $\omega_n-k_nv_z$ for $n=1,2$ and $\omega_n+k_nv_z$ for $n=3,4$. If we take into account the adopted equations $k_1=3k_2$, $k_3=3k_4$, $k_2=k_4$, and $\bar{\Delta}_{12}<0$, then in accordance with the energy conservation law the four following equations correspond to the four two-phonon resonances in Figs. 2 and 3:

$$(k_1+k_2)v_z-\bar{\Delta}_{12}=0,\quad (k_3+k_4)v_z+\bar{\Delta}_{12}=0, \tag{77}$$

$$(k_1-k_4)v_z-\bar{\Delta}_{12}=0,\quad (k_3-k_2)v_z+\bar{\Delta}_{12}=0, \tag{78}$$

where $v_z=\pm|v_z|$. Each of these equations characterizes a process of absorption (or emission) of two different pairs of photons. The two outer peaks in Figs. 2 and 3 describe the behavior of the optical-pressure force in the neighborhood of the resonances (78), while the two inner peaks characterize this force in the neighborhood of the resonances (77). At large detunings from two-photon resonance, $|\bar{\Delta}_{12}|\gg\gamma_{cb}$, the height of the inner peaks in Fig. 2 is almost independent of z, whereas the height of the outer peaks is proportional to $z/2$.

6. CONCLUSIONS

The obtained optical-pressure forces (42), (49), (64), and (72) for Raman, (2), and two-photon, (3), resonance depend strongly on the relaxation constant γ_{cb} of the two-photon transition $E_c\rightarrow E_b$. For a single atom in the weak field (1), it is due to radiative processes or the time of flight of the atom

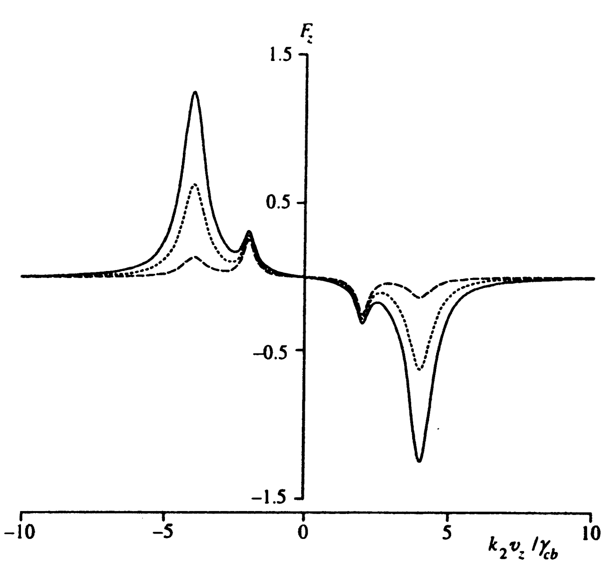

FIG. 2. Optical-pressure force (76) as function of the velocity v_z for $\bar{\Delta}_{12}=-8\gamma_{cb}$. The unit for the ordinate is F_{12}. The dashed, dotted, and solid curves correspond to $z=1$, $z=5$, and $z=10$.

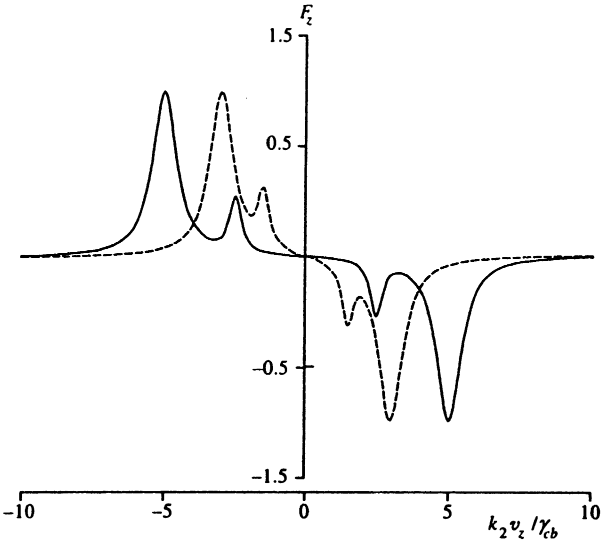

FIG. 3. Optical-pressure force (76) as function of the velocity v_z for $z=6$. The unit for the ordinate is F_{12}. The dashed and solid curves correspond to $\bar{\Delta}_{12}=-6\gamma_{cb}$ and $\bar{\Delta}_{12}=-10\gamma_{cb}$.

through this field. In the case of the stationary regime leading to the expressions (42), (49), (64), and (72), the relaxation constant γ_{cb} has a radiative origin. At the same time, for the ground level E_b in the weak field (1) we obtain $\gamma_b=0$, and therefore the relaxation constant $\gamma_{cb}=\gamma_c/2$ attains a rather high value if between the levels E_b and E_c there is at least one level E_e in adjacent electric dipole transitions $E_c \to E_e$ and $E_e \to E_b$, which increase the value of γ_c (Figs. 1a and 1b). In accordance with Ref. 35, we find such examples in the calcium atom ${}^{48}_{20}\mathrm{Ca}$ with levels $E_b(4^1S_0)$, $E_e(4^1P_1^0)$, and $E_c(5^3S_1)$, where $E_c=3.91$ eV and $\gamma_c=10^8\ \mathrm{s}^{-1}$, and also in the barium atom ${}^{138}_{56}\mathrm{Ba}$ with levels $E_b(6^1S_0)$, $E_e(6^3D_3^0)$, and $E_c(7^1D_2)$, where $E_c=4.64$ eV and $\gamma_c=5\times10^7\ \mathrm{s}^{-1}$.

In this paper we have considered one pair of oppositely directed waves with frequencies (2) or (3) and also two pairs of oppositely directed waves with frequencies and polarizations (55) and (56). Since the doubled frequencies $2\omega_n$ with $n=1$, 2, 3, and 4 are nonresonant, the atom does not absorb two photons of the same frequency without the presence of a test wave with a different frequency, and therefore a corresponding contribution to the obtained optical-pressure force is absent. In the case of one pair of oppositely directed waves in the presence of Raman resonance (2), the optical-pressure force (36) contains interference terms, which depend on the coordinates of the atom by virtue of phases of the form $(\omega_1 \pm \omega_2)t-(\mathbf{k}_1 \pm \mathbf{k}_2)\cdot\mathbf{r}$ corresponding to the frequencies (37). However, after the averaging over the time (33) these terms acquire a small factor that enables us to omit these terms. As a result, we obtain the rectified radiation force (42). For the two-photon resonance (3), the arguments are similar. In the case of two pairs of oppositely directed waves (55) and (56), the situation is more complicated, since the terms of the optical-pressure force can be decomposed into three different groups. The first group contains terms for which the phases cancel each other in the arguments of the exponentials. This group of terms leads in the case of the Raman resonance (2) to the rectified radiation force (63). The second group of terms contains factors $\exp(\pm 2i\mathbf{k}_1\cdot\mathbf{r})$ and $\exp(\pm 2i\mathbf{k}_2\cdot\mathbf{r})$ without the light frequencies in the arguments of the exponentials. However, the coefficients of these exponentials vanish if the polarizations of the waves are chosen in the form (61) or (62). The third group contains interference terms which depend on the coordinates of the atom because of the phases of the light waves that correspond to the frequencies (37). After the averaging over the time (33), the third group makes a negligible contribution which can be omitted. If after this procedure we choose the amplitudes of the oppositely directed waves to be the same, $R_1=R_2$ and $R_3=R_4$, then we obtain the rectified radiation force (64), which is odd with respect to the velocity of the atom. A similar situation occurs in the approximation of perturbation theory quadratic in the field (1) for two oppositely directed waves with $R_1=R_2$, the same circular polarizations, and frequencies $\omega_1=\omega_2$ in resonance with an electric dipole transition.[27]

In the presence of only one resonance of two-photon absorption, i.e., for

$$|2\omega_n-\omega_{cb}| \lesssim \gamma_{cb}, \quad n=1,2, \tag{79}$$

we can use two oppositely directed waves with $\mathbf{k}_1=-\mathbf{k}_2$, $R_1=R_2$, and specially chosen polarizations. The optical-pressure force for the given two-photon transition will then be the principal force acting on the atom. In this case, the calculations differ from those considered in Secs. 2–4 and constitute a separate problem with the following feature. As was shown in Ref. 36 (see also Ref. 2), in the case of satsifaction of the condition (79) in the field of a standing wave the atom can absorb two photons from the first or second wave and also one photon from each of the oppositely directed waves separately. Since in the nonrelativistic region in the center-of-mass system the frequencies of the oppositely directed waves are $\omega_1-\mathbf{k}_1\cdot\mathbf{v}$ and $\omega_2-\mathbf{k}_2\cdot\mathbf{v}$, the sum of these frequencies $\omega_1+\omega_2$ for $\mathbf{k}_1+\mathbf{k}_2=0$ is in the resonance (79), regardless of the velocity $\mathbf{v}$ of the atom. Therefore, the profile of the two-photon absorption line in a gas has a narrow peak that does not depend on Doppler broadening. However, this absorption without Doppler effect in the field of the standing wave does not contribute to the optical-pressure force, since the atom simultaneously absorbs momentum $\hbar \mathbf{k}_1$ from one wave and momentum $\hbar \mathbf{k}_2 = -\hbar \mathbf{k}_1$ from the other wave, which leads to a vanishing optical-pressure force in the case of absorption without Doppler effect. In this case, a nonvanishing contribution to the rectified optical-pressure force is made by each of the oppositely directed waves separately when the polarizations of these waves are specially chosen. Such terms do not occur in the optical-pressure force (49), since the two oppositely directed waves which we considered did not satisfy the two-photon absorption resonance (79).

It would be interesting to apply the optical-pressure force (64) in Raman resonance (2) to molecules with vibrational–rotational transitions, which have been used already in the investigation of Raman scattering in molecular gases.[37,31] Similarly, the optical-pressure force (72) in the case of the two-photon resonance (3) can be measured for molecules with vibrational–rotational or electronic–vibrational–rotational transitions, which have been studied in the spectroscopy of two-photon absorption.[2,31]

The widest selection of necessary level configurations corresponding to the requirements (2), or (3), or (79) exists among atoms with hyperfine structure of the levels due to interaction of the electron subsystem with the nuclear spin. However, the calculation of optical-pressure forces for an atom with hyperfine structure of its levels in the indicated cases contains various features that require separate study.

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