

An atom in an elliptically polarized resonant field: the exact steady-state solution for closed $j_g=j \rightarrow j_e=j+1$ transitions

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We suggest an analytical method for finding the exact steady-state solution of the problem of the resonant interaction of atoms and a monochromatic field for closed transitions $j_g=j \rightarrow j_e=j+1$, where j_g and j_e are the angular momenta of the ground (g) and excited (e) states, for light of arbitrary intensity and ellipticity. The solution for $j=0, 1/2, 1, \dots, 5$ is given in explicit form. We study the properties of the resulting solution and prove its uniqueness. As an application we determine the dependence of the absorption coefficient on the ellipticity and calculate the gradient force acting on slow atoms. © 1996 American Institute of Physics. [S1063-7761(96)01211-5]

1. INTRODUCTION

Many problems of the resonant interaction of atoms and polarized light, such as nonlinear polarization spectroscopy and the mechanical action of light on atoms, require knowing the exact steady-state solution of the optical Bloch equations for the atomic density matrix with allowance for the Zeeman structure of the energy levels involved. Finding such a solution in analytical form for closed optical transitions (where the ground level is the lower level and the total population is conserved) in the general case of arbitrary ellipticity and intensity of the field is extremely difficult. The main mathematical difficulties stem from the fact that the number of coupled equations for the elements of the density matrix is large. In addition, even when the transition is not saturated, a perturbation-theory expansion in powers of the atom-field coupling constant does not simplify the problem much due to the inhomogeneous distribution of atoms over the magnetic sublevels and the appearance of Zeeman coherences in the ground state (optical order effects).

Earlier (see Refs. 1 and 2) we found an exact steady-state solution in general form for two groups of transitions, $j_g=j \rightarrow j_e=j-1$ and $j_g=j \rightarrow j_e=j$, where j_g and j_e are the total angular momenta of the ground (g) and excited (e) states (j is arbitrary), belonging to the class of dipole-allowed transitions ($\Delta j=j_e-j_g=0, \pm 1$). The $j_g=j \rightarrow j_e=j+1$ transitions were examined in Refs. 3–5, where an exact solution for the particular cases of linear and circular polarization of the field was obtained. For light of arbitrary ellipticity a steady-state solution in analytical form was obtained^{6,7} only for transitions involving small values of total angular momentum ($j_g=0 \rightarrow j_e=1$ and $j_g=1/2 \rightarrow j_e=3/2$).

In this paper we study the resonant interaction of atoms with energy levels degenerate in the projection of the total angular momentum and a polarized monochromatic field. We propose an analytical method for finding the exact steady-state solution of the problem, with allowance for radiative relaxation, for closed transitions $j_g=j \rightarrow j_e=j+1$ and arbitrary ellipticity and intensity of the light. Our method is based on a theorem according to which in the case of purely radiative relaxation of the atoms the steady-state den-

sity matrix can be written in an invariant form as a polynomial of the operator of the resonant atom-field interaction. The expansion coefficients can be found from recurrence relations solvable for all values of j . We list the coefficients for $j=0, 1/2, 1, \dots, 5$ explicitly. In the general case of arbitrary ellipticity the validity of the theorem is verified by direct substitution for $j=0, 1/2, 1, \dots, 10$. The theorem is proved for any value of j in the case where the field polarization is close to circular. Note that this approach can also be applied to $j_g=j \rightarrow j_e=j$ transitions, with the steady-state solutions coinciding with the results of Refs. 1 and 2 obtained by other methods.

We find that just as for the $j_g=j \rightarrow j_e=j-1$ and $j_g=j \rightarrow j_e=j$ transitions,^{1,2} the solution possesses the following property: the anisotropy of the density matrices of the excited state ($\hat{\rho}^{ee}$) and the off-diagonal elements ($\hat{\rho}^{eg}$ and $\hat{\rho}^{ge}$) is determined entirely by the ellipticity of the light, while the intensity and the detuning from resonance enter only into the corresponding scalar factors (see (22)–(24)). We also find the conditions for strong and weak saturation of a transition.

Finally, to illustrate the possible applications we study the ellipticity dependence of the absorption coefficient and calculate the velocity-independent component of the gradient force.

2. STATEMENT OF THE PROBLEM

Basically the statement of the problem is similar to that of Ref. 2. We examine the interaction of atoms whose ground and excited states form a closed optical transition $j_g=j \rightarrow j_e=j+1$ and an elliptically polarized resonant plane wave

$$\mathbf{E} = E_0 \mathbf{e} \exp[-i(\omega t - \mathbf{k} \cdot \mathbf{r})] + \text{c.c.}, \quad (1)$$

where

$$\mathbf{e} = \sum_{q=0, \pm 1} e^q \mathbf{e}_q$$

is the unit complex-valued polarization vector of the field, and the e^q are its components in a cyclic basis $\{\mathbf{e}_0 = \mathbf{e}_z,$

$\mathbf{e}_{\pm 1} = \mp(\mathbf{e}_x \pm i\mathbf{e}_y)/\sqrt{2}$. In particular, if we chose the quantization z axis directed along the wave vector \mathbf{k} and the x axis directed along the major semiaxis of the polarization ellipse, we can write

$$\begin{aligned} \mathbf{e} &= \cos \varepsilon \mathbf{e}_x + i \sin \varepsilon \mathbf{e}_y \\ &= -\cos(\varepsilon - \pi/4)\mathbf{e}_{+1} - \sin(\varepsilon - \pi/4)\mathbf{e}_{-1}, \\ -\pi/4 &\leq \varepsilon \leq \pi/4, \end{aligned} \quad (2)$$

where ε is the ellipticity angle ($\tan \varepsilon$ is the ratio of the minor semiaxis of the polarization ellipse to the major semiaxis, and the sign of ε depends on the sense of rotation).

When dealing with a low-density gas, we can completely ignore interatomic collisions. We also restrict our discussion to the zeroth order in the small recoil parameter $\hbar k/\Delta p \ll 1$ (here $\hbar k$ is the photon momentum, and Δp is the dispersion of the atomic momentum), i.e., we examine the motion of the center of mass in the classical setting. Then the quantum kinetic equation describing the evolution of the density matrix of atoms in the external field (1) assumes the form

$$\left(\frac{\partial}{\partial t} + \mathbf{v} \cdot \nabla\right) \hat{\rho} + \hat{\Gamma}\{\hat{\rho}\} = -\frac{i}{\hbar}[\hat{H}_0, \hat{\rho}] - \frac{i}{\hbar}[\hat{V}_{E-D}, \hat{\rho}]. \quad (3)$$

Here \mathbf{v} is the atomic velocity, \hat{H}_0 is the Hamiltonian of a free atom in the center-of-mass reference frame, and in the case of optical transitions we can limit ourselves to the electric dipole approximation for the operator $\hat{V}_{E-D} = -(\hat{\mathbf{d}} \cdot \mathbf{E})$ describing the atom-field interaction ($\hat{\mathbf{d}}$ is the dipole moment operator), and the operator $\hat{\Gamma}\{\hat{\rho}\}$ describes radiative relaxation.

We split the density matrix into four matrix blocks $\hat{\rho}^{gg}$, $\hat{\rho}^{ee}$, $\hat{\rho}^{eg}$, and $\hat{\rho}^{ge}$ as follows:

$$\begin{aligned} \rho_{\mu_g \mu'_g}^{gg} &= \langle g, \mu_g | \hat{\rho} | g, \mu'_g \rangle, & \rho_{\mu_e \mu'_e}^{ee} &= \langle e, \mu_e | \hat{\rho} | e, \mu'_e \rangle, \\ \rho_{\mu_e \mu'_g}^{eg} &= \langle e, \mu_e | \hat{\rho} | g, \mu'_g \rangle, & \rho_{\mu_g \mu'_e}^{ge} &= \langle g, \mu_g | \hat{\rho} | e, \mu'_e \rangle, \end{aligned} \quad (4)$$

where $\{|g, \mu_g\rangle\}$ and $\{|e, \mu_e\rangle\}$ are the Zeeman wave functions of the ground and excited states. In the resonant approximation we can express the fast dependence on time and coordinates in $\hat{\rho}^{eg}$ and $\hat{\rho}^{ge}$ explicitly,

$$\begin{aligned} \hat{\rho}^{eg} &= \exp[-i(\omega t - \mathbf{k} \cdot \mathbf{r})] \hat{\rho}^{eg}, \\ \hat{\rho}^{ge} &= \exp[i(\omega t - \mathbf{k} \cdot \mathbf{r})] \hat{\rho}^{ge}, \end{aligned} \quad (5)$$

and write the following system of equations for the components that are spatially homogeneously and slowly vary in time:

$$\left(\frac{\partial}{\partial t} + \frac{\gamma}{2} - i\delta\right) \hat{\rho}^{eg} = -i\Omega[\hat{V}\hat{\rho}^{gg} - \hat{\rho}^{ee}\hat{V}], \quad (6)$$

$$\left(\frac{\partial}{\partial t} + \frac{\gamma}{2} + i\delta\right) \hat{\rho}^{ge} = -i\Omega^*[\hat{V}^\dagger \hat{\rho}^{ee} - \hat{\rho}^{gg}\hat{V}^\dagger], \quad (7)$$

$$\left(\frac{\partial}{\partial t} + \gamma\right) \hat{\rho}^{ee} = -i[\Omega\hat{V}\hat{\rho}^{ge} - \Omega^*\hat{\rho}^{eg}\hat{V}^\dagger], \quad (8)$$

$$\frac{\partial}{\partial t} \hat{\rho}^{gg} - \hat{\gamma}\{\hat{\rho}^{gg}\} = -i[\Omega^*\hat{V}^\dagger \hat{\rho}^{eg} - \Omega\hat{\rho}^{ge}\hat{V}], \quad (9)$$

$$\text{Tr}\{\hat{\rho}^{gg}\} + \text{Tr}\{\hat{\rho}^{ee}\} = 1. \quad (10)$$

Here $\delta = (\omega - \omega_{eg} - \mathbf{k} \cdot \mathbf{v})$ is the detuning from resonance with allowance for the Doppler shift, $\omega_{eg} = (E_e - E_g)/\hbar$ is the transition frequency, γ is the spontaneous relaxation rate, and $\Omega = -E_0 \langle e || d || g \rangle / \hbar$ is the effective Rabi frequency, with $\langle e || d || g \rangle$ the reduced matrix element of the dipole moment. The matrix elements of \hat{V} can be expressed in terms of $3jm$ -symbols in accordance with the Wigner-Eckart theorem.⁸

$$V_{\mu_e \mu'_g} = \sum_{q=0, \pm 1} (-1)^{j_e - \mu_e} \begin{pmatrix} j_e & 1 & j_g \\ -\mu_e & q & \mu'_g \end{pmatrix} e^q. \quad (11)$$

For closed optical transitions the operator $\hat{\gamma}\{\hat{\rho}^{ee}\}$ of the arrival of atoms at the ground state due to spontaneous emission has the following standard form (see, e.g., Ref. 9):

$$\begin{aligned} \gamma_{\mu_g \mu'_g} \{\hat{\rho}^{ee}\} &= \gamma(2j_e + 1) \sum_{q, \mu_e, \mu'_e} (-1)^{j_e - \mu_e} \begin{pmatrix} j_e & 1 & j_g \\ -\mu_e & q & \mu'_g \end{pmatrix} \\ &\times \rho_{\mu_e \mu'_e}^{ee} (-1)^{j_e - \mu'_e} \begin{pmatrix} j_e & 1 & j_g \\ -\mu'_e & q & \mu'_g \end{pmatrix}. \end{aligned} \quad (12)$$

Setting all time derivatives in Eqs. (6)–(10) to zero,

$$\frac{\partial}{\partial t} \hat{\rho}^{gg} = \frac{\partial}{\partial t} \hat{\rho}^{ee} = \frac{\partial}{\partial t} \hat{\rho}^{eg} = \frac{\partial}{\partial t} \hat{\rho}^{ge} = 0, \quad (13)$$

and expressing the off-diagonal elements $\hat{\rho}^{eg}$ and $\hat{\rho}^{ge}$ in terms of the density matrices $\hat{\rho}^{gg}$ and $\hat{\rho}^{ee}$,

$$\begin{aligned} \hat{\rho}^{eg} &= -\frac{i\Omega}{\gamma/2 - i\delta} [\hat{V}\hat{\rho}^{gg} - \hat{\rho}^{ee}\hat{V}], \\ \hat{\rho}^{ge} &= -\frac{i\Omega^*}{\gamma/2 + i\delta} [\hat{V}^\dagger \hat{\rho}^{ee} - \hat{\rho}^{gg}\hat{V}^\dagger], \end{aligned} \quad (14)$$

we arrive at the following closed system of matrix equations for the steady-state $\hat{\rho}^{gg}$ and $\hat{\rho}^{ee}$:

$$\begin{aligned} \gamma \hat{\rho}^{ee} &= \gamma S \hat{V} \hat{\rho}^{gg} \hat{V}^\dagger - \frac{\gamma}{2} S \{ \hat{V} \hat{V}^\dagger \hat{\rho}^{ee} + \hat{\rho}^{ee} \hat{V} \hat{V}^\dagger \} \\ &+ i \delta S \{ \hat{V} \hat{V}^\dagger \hat{\rho}^{ee} - \hat{\rho}^{ee} \hat{V} \hat{V}^\dagger \}, \\ \hat{\gamma}\{\hat{\rho}^{gg}\} &= -\gamma S \hat{V}^\dagger \hat{\rho}^{ee} \hat{V} + \frac{\gamma}{2} S \{ \hat{V}^\dagger \hat{V} \hat{\rho}^{gg} + \hat{\rho}^{gg} \hat{V}^\dagger \hat{V} \} \\ &+ i \delta S \{ \hat{V}^\dagger \hat{V} \hat{\rho}^{gg} - \hat{\rho}^{gg} \hat{V}^\dagger \hat{V} \}, \end{aligned} \quad (15)$$

where

$$S = \frac{|\Omega|^2}{\gamma^2/4 + \delta^2} \quad (16)$$

is the saturation parameter.

Note that generally Eqs. (6)–(10) describe damped Rabi oscillations, and the condition for a steady-state regime to set in for any value of S can be written as follows:

$$\gamma S t \gg 1, \quad \gamma t \gg 1,$$

where t is the time over which the atom interacts with the field.

3. THE EXACT STEADY-STATE SOLUTION

Our method is based on a theorem formulated below.

Theorem. For arbitrary j_g and j_e in the case of purely radiative relaxation, when the steady-state Bloch equations have the form (15) with the arrival operator (12), the density matrices $\hat{\rho}^{gg}$ and $\hat{\rho}^{ee}$ commute with the Hermitian matrices $\hat{V}^\dagger\hat{V}$ and $\hat{V}\hat{V}^\dagger$, respectively:

$$[\hat{V}^\dagger\hat{V}, \hat{\rho}^{gg}] = 0, \quad [\hat{V}\hat{V}^\dagger, \hat{\rho}^{ee}] = 0. \quad (17)$$

As is known from algebra, this means that the solution of system (17) is diagonal in the basis of the eigenvectors of the operators $\hat{V}^\dagger\hat{V}$ and $\hat{V}\hat{V}^\dagger$. In addition, since by (17) the factors of $i\delta$ in (15) vanish, the matrices $\hat{\rho}^{gg}$ and $\hat{\rho}^{ee}$ depend only on even powers of δ .

The validity of Theorem (17) for the $j_g = j \rightarrow j_e = j-1$ and $j_g = j \rightarrow j_e = j$ transitions follows directly from the results of Refs. 1 and 2.

In the present paper we assume that Theorem (17) remains valid for $j_g = j \rightarrow j_e = j+1$ transitions. This makes it possible to develop an analytical procedure for finding steady-state solutions for any j .

Before we begin to look for the solution of the system of equations (15) in general form, let us examine the weak-saturation limit ($S \ll 1$).

3.1. A method for finding the solution in the case of weak saturation

For small $S \ll 1$, keeping in (15) the first-order terms in S , we get

$$\gamma \hat{\rho}_1^{ee} = \gamma S \hat{V} \hat{\rho}_0^{gg} \hat{V}^\dagger,$$

$$\begin{aligned} \gamma \{\hat{\rho}_1^{ee}\} &= \frac{\gamma}{2} S \{ \hat{V}^\dagger \hat{V} \hat{\rho}_0^{gg} + \hat{\rho}_0^{gg} \hat{V}^\dagger \hat{V} \} \\ &+ i\delta S \{ \hat{V}^\dagger \hat{V} \hat{\rho}_0^{gg} - \hat{\rho}_0^{gg} \hat{V}^\dagger \hat{V} \}. \end{aligned} \quad (18)$$

This readily leads to a closed equation for $\hat{\rho}_0^{gg}$ without the saturation parameter S :

$$\begin{aligned} \gamma \{ \hat{V} \hat{\rho}_0^{gg} \hat{V}^\dagger \} &= \frac{\gamma}{2} \{ \hat{V}^\dagger \hat{V} \hat{\rho}_0^{gg} + \hat{\rho}_0^{gg} \hat{V}^\dagger \hat{V} \} \\ &+ i\delta \{ \hat{V}^\dagger \hat{V} \hat{\rho}_0^{gg} - \hat{\rho}_0^{gg} \hat{V}^\dagger \hat{V} \}. \end{aligned} \quad (19)$$

If we employ the hypothesis of Theorem (17), the solution of Eq. (19) for the $j_g = j \rightarrow j_e = j+1$ transitions can be represented in the form of a polynomial in the matrix $\hat{V}^\dagger\hat{V}$:

$$\hat{\rho}_0^{gg} = \beta_0 \sum_{n=0}^{2j} C_n(\varepsilon) (\hat{V}^\dagger\hat{V})^n, \quad (20)$$

where β_0 is a normalization constant. Indeed, since the eigenvalues of $\hat{V}^\dagger\hat{V}$ are analytic functions of ε and at $\varepsilon = \pm \pi/4$ (circularly polarized light) all these eigenvalues are distinct,

$$\lambda_\mu = \frac{(j+1+\mu)(j+2+\mu)}{2(j+1)(2j+1)(2j+3)}, \quad \mu = -j, -j+1, \dots, j,$$

according to the well-known theorems of the theory of analytic functions, the eigenvalues may become degenerate only

at discrete points in the analyticity interval $-\pi/4 \leq \varepsilon \leq \pi/4$ (say, for linearly polarized light ($\varepsilon=0$) we have twofold degeneracy). This suggests that the expansion (20) determines the solution of Eq. (19) unambiguously as an analytic function of ε , provided that Theorem (17) is valid.

Plugging (20) into (19) yields a system of equations for the coefficients $C_n(\varepsilon)$:

$$\frac{1}{\gamma} \sum_{n=0}^{2j} C_n(\varepsilon) \gamma \{ (\hat{V}\hat{V}^\dagger)^{n+1} \} = \sum_{n=0}^{2j} C_n(\varepsilon) (\hat{V}^\dagger\hat{V})^{n+1}. \quad (21)$$

The number of equations in (21) is $(2j+1)^2$, but the system is highly degenerate, and its rank is equal to $2j$. Hence one of the coefficients can be chosen in an arbitrary manner (for instance, below we put $C_{2j}(\varepsilon)=1$). To determine the $C_n(\varepsilon)$ we can take any $2j$ linearly independent equations in (21). Note that the coefficients $C_n(\varepsilon)$ depend only on the ellipticity and are invariant, i.e., do not depend on the choice of the coordinate system.

3.2. Solution in the general case

Let us return to the system of equations (15). If we know the nontrivial solution of the system (21), the steady-state density matrices $\hat{\rho}^{ee}$ and $\hat{\rho}^{gg}$ for an arbitrary S have the form of expansions in powers of the operators $\hat{V}\hat{V}^\dagger$ and $\hat{V}^\dagger\hat{V}$:

$$\begin{aligned} \hat{\rho}^{ee} &= \beta S \sum_{n=0}^{2j} C_n(\varepsilon) (\hat{V}\hat{V}^\dagger)^{n+1}, \\ \hat{\rho}^{gg} &= \beta \left[\sum_{n=0}^{2j} C_n(\varepsilon) (\hat{V}^\dagger\hat{V})^n + S \sum_{n=0}^{2j} C_n(\varepsilon) (\hat{V}^\dagger\hat{V})^{n+1} \right]. \end{aligned} \quad (22)$$

Direct substitution with allowance for (21) clearly shows that (22) satisfies the system of equations (15) identically. The constant β can be found from the normalization condition (10):

$$\beta = [\alpha_0 + 2S\alpha_1]^{-1}, \quad (23)$$

$$\alpha_0 = \text{Tr} \left\{ \sum_{n=0}^{2j} C_n(\varepsilon) (\hat{V}^\dagger\hat{V})^n \right\},$$

$$\alpha_1 = \text{Tr} \left\{ \sum_{n=0}^{2j} C_n(\varepsilon) (\hat{V}^\dagger\hat{V})^{n+1} \right\}.$$

Here we have used the obvious fact that $\text{Tr}\{(\hat{V}^\dagger\hat{V})^n\} = \text{Tr}\{(\hat{V}\hat{V}^\dagger)^n\}$.

Thus, according to the theorem (17), the solution of the system (15) consisting of $(2j+1)^2 + (2j+3)^2$ equations can be written in an analytically invariant form by determining only $2j+1$ coefficients ($2j$ coefficients $C_n(\varepsilon)$, since one of these can be selected in an arbitrary manner, and the normalization constant β).

Plugging (22) into (14), we arrive at an expression for $\hat{\rho}^{eg}$ and $\hat{\rho}^{ge}$:

$$\hat{\rho}^{eg} = (\hat{\rho}^{ge})^\dagger = -\frac{i\beta\Omega}{\gamma/2 - i\delta} \hat{V} \sum_{n=0}^{2j} C_n(\varepsilon) (\hat{V}^\dagger\hat{V})^n. \quad (24)$$

The expressions (22)–(24) show that the anisotropy of the steady-state matrices $\hat{\rho}^{ee}$, $\hat{\rho}^{eg}$, and $\hat{\rho}^{ge}$ is determined en-

tirely by the unit polarization vector \mathbf{e} of the light, and the amplitude and detuning of the field enters only into the respective scalar factors. At the same time, the anisotropy of the ground state strongly depends on the saturation parameter S , since the second term in the expression for $\hat{\rho}^{gg}$ (see (22)) can be obtained from the first via multiplication by the matrix $S(\hat{V}^\dagger\hat{V})$. The invariant operator form of the solutions (22)–(24) makes it possible to calculate both the density matrix elements (the jm -representation) and the multipole moments of the density matrix (the κq -representation) in an arbitrary basis. Note that in contrast to a linearly or circularly polarized field, an elliptically polarized field induces a non-zero Zeeman coherence in any system of coordinates.

Let us show that the analytical solution (22)–(24) is unique. Indeed, since the normalization (10) is conserved, the system of equations (15) for closed optical transitions is singular, and one of the linearly dependent equations in (15) can be replaced by (10). The resulting system of equations for the components of the matrices $\hat{\rho}^{ee}$ and $\hat{\rho}^{gg}$ is inhomogeneous due to the presence of the right-hand side of Eq. (10). In addition, it is obvious that the determinant $\Delta(\varepsilon)$ of this system is an analytic function of the ellipticity angle ε (see Eq. (2)). Taking now the value $\varepsilon = \pm\pi/4$ (circular polarization), we can easily show that because of optical pumping the problem is reduced to the well-known two-level model, where the uppermost and lowermost Zeeman states $|e, j+1\rangle$ and $|g, j\rangle$ act as the levels. And the two-level model has a unique steady-state solution, which implies $\Delta(\pi/4) \neq 0$. Then, basing our reasoning on the most general properties of analytic functions, we can say that $\Delta(\varepsilon)$ vanishes only at discrete points in the interval $-\pi/4 \leq \varepsilon \leq \pi/4$, and the solution of the system of equations (15) and (10), an analytic function of ε , is unique, which is what we set out to prove. Note that such reasoning can be employed in proving the uniqueness of the analytical solutions found in Refs. 1 and 2 for the $j_g = j \rightarrow j_e = j$ transitions, since in the case of circular polarization the solution is obviously unique, too.

As (22) and (23) imply, the ratio of the total populations of the ground and excited states is

$$\frac{\text{Tr}\{\hat{\rho}^{ee}\}}{\text{Tr}\{\hat{\rho}^{gg}\}} = \left(\frac{\alpha_0}{S\alpha_1} + 1 \right)^{-1}. \quad (25)$$

This readily leads to the conditions for weak and strong saturations of a transition. For instance, weak saturation corresponds to

$$S \frac{\alpha_1}{\alpha_0} \ll 1. \quad (26)$$

Here

$$\text{Tr}\{\hat{\rho}^{ee}\} \ll \text{Tr}\{\hat{\rho}^{gg}\}, \quad \beta \approx \frac{1}{\alpha_0},$$

and in (22) we can ignore the second term in the expression for $\hat{\rho}^{gg}$:

$$\hat{\rho}^{ee} \approx \frac{S}{\alpha_0} \sum_{n=0}^{2j} C_n(\varepsilon) (\hat{V}^\dagger\hat{V})^{n+1}, \quad (27)$$

$$\hat{\rho}^{gg} \approx \frac{1}{\alpha_0} \sum_{n=0}^{2j} C_n(\varepsilon) (\hat{V}^\dagger\hat{V})^n.$$

The condition that is the opposite of (26),

$$S \frac{\alpha_1}{\alpha_0} \gg 1, \quad (28)$$

corresponds to strong saturation of the transition. Here the total populations of the ground and excited states are roughly the same,

$$\text{Tr}\{\hat{\rho}^{ee}\} \approx \text{Tr}\{\hat{\rho}^{gg}\}, \quad \beta \approx \frac{1}{2S\alpha_1},$$

and in (22) we can ignore the first term in the expression for $\hat{\rho}^{gg}$:

$$\begin{aligned} \hat{\rho}^{ee} &\approx \frac{1}{2\alpha_1} \sum_{n=0}^{2j} C_n(\varepsilon) (\hat{V}^\dagger\hat{V})^{n+1}, \\ \hat{\rho}^{gg} &\approx \frac{1}{2\alpha_1} \sum_{n=0}^{2j} C_n(\varepsilon) (\hat{V}^\dagger\hat{V})^{n+1}. \end{aligned} \quad (29)$$

Since for the $j_g = j \rightarrow j_e = j+1$ transitions the matrix $\hat{V}^\dagger\hat{V}$ acting in the space of the ground-state Zeeman wave functions $\{|g, \mu_g\rangle\}$ is always nonsingular (according to Ref. 1, only the matrix $\hat{V}\hat{V}^\dagger$ acting in the space $\{|e, \mu_e\rangle\}$ is singular), the condition (28) for strong saturation holds for a field of arbitrary ellipticity. This constitutes the main difference of this case from $j_g = j' \rightarrow j_e = j'$ transitions (j' is a half-integer), where, as Ref. 2 implies, the condition for strong saturation is highly dependent on the ellipticity of the field. For instance, in a circularly polarized field there is no intensity at which the $j_g = j' \rightarrow j_e = j'$ transitions (j' is a half-integer) become saturated (the effect of coherent trapping of populations).

3.3. Calculating the coefficients $C_n(\varepsilon)$

The solutions (22)–(24) show that to fully determine the steady-state solution we must find the coefficients $C_n(\varepsilon)$, which constitute the solution of $2j$ linearly independent equations in (21). Since the $C_n(\varepsilon)$ are invariants, we can use an arbitrary system of coordinates for their calculation. We select the one suggested in Ref. 10. As is known, an arbitrary ellipse is a curve along which a cylinder and a plane intersect, so that to each elliptic polarization vector \mathbf{e} we can assign a cylinder (generally there are two such cylinders) whose section is the given ellipse \mathbf{e} . We direct the quantization z axis along the axis of the cylinder and the y axis along the minor semiaxis of the polarization ellipse (Fig. 1). Then, as Ref. 10 implies, the elliptic polarization \mathbf{e} is a linear combination of the linear and one of the circular components:

$$\mathbf{e} = \sqrt{\cos(2\varepsilon)}\mathbf{e}_0 + \sqrt{2} \sin \varepsilon \mathbf{e}_{\pm 1}. \quad (30)$$

For the sake of definiteness we take the decomposition

$$\mathbf{e} = \sqrt{\cos(2\varepsilon)}\mathbf{e}_0 + \sqrt{2} \sin \varepsilon \mathbf{e}_{+1}. \quad (31)$$

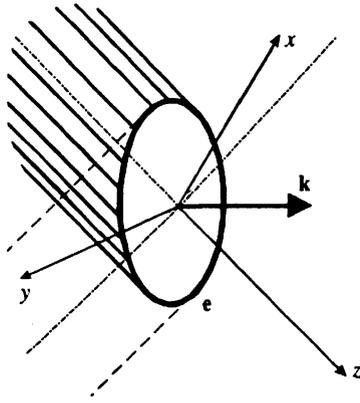


FIG. 1. The coordinate system suggested in Ref. 10 in which the z axis (the quantization axis) is directed along the axis of one of the cylinders built on the polarization ellipse e (the dashed lines stand for the second cylinder) and the y axis is directed along the minor semiaxis of the polarization ellipse. In this base the vector e is a superposition of the linear component and one of the circular components (see Eq. (30)).

The light-induced transitions corresponding to (31) are depicted in Fig. 2. In this case the matrices $\hat{V}^\dagger \hat{V}$ and $\hat{V} \hat{V}^\dagger$ are real and symmetric and have three nonzero diagonals (denoted by *):

$$\hat{V}^\dagger \hat{V} = \begin{pmatrix} * & * & 0 & \cdot & \cdot & \cdot & 0 \\ * & * & * & 0 & \cdot & \cdot & \cdot \\ 0 & * & * & a_{(\mu_g-1)\mu_g} & 0 & \cdot & \cdot \\ \cdot & 0 & * & a_{\mu_g\mu_g} & * & 0 & \cdot \\ \cdot & \cdot & 0 & a_{(\mu_g+1)\mu_g} & * & * & 0 \\ \cdot & \cdot & \cdot & 0 & * & * & * \\ 0 & \cdot & \cdot & \cdot & 0 & * & * \end{pmatrix},$$

$$\mu_g = -j, -j+1, \dots, j, \quad (32)$$

$$\hat{V} \hat{V}^\dagger = \begin{pmatrix} 0 & 0 & 0 & \cdot & \cdot & \cdot & 0 \\ 0 & * & * & 0 & \cdot & \cdot & \cdot \\ 0 & * & * & b_{(\mu_e-1)\mu_e} & 0 & \cdot & \cdot \\ \cdot & 0 & * & b_{\mu_e\mu_e} & * & 0 & \cdot \\ \cdot & \cdot & 0 & b_{(\mu_e+1)\mu_e} & * & * & 0 \\ \cdot & \cdot & \cdot & 0 & * & * & * \\ 0 & \cdot & \cdot & \cdot & 0 & * & * \end{pmatrix},$$

$$\mu_e = -j-1, -j, \dots, j+1,$$

where, in accordance with (11) with (31), the matrix elements have the form

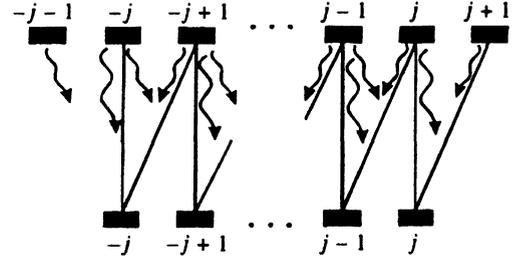


FIG. 2. The diagram representing light-induced (solid lines) and spontaneous (wavy lines) transitions in the system of coordinates depicted in Fig. 1 with the vector e specified by (31).

$$a_{\mu_g\mu_g} = \frac{[(j+1)^2 - \mu_g^2] \cos(2\varepsilon) + (j+1 + \mu_g)(j+2 + \mu_g) \sin^2 \varepsilon}{(2j+3)(2j+1)(j+1)},$$

$$a_{\mu_g(\mu_g-1)} = a_{(\mu_g-1)\mu_g} = \frac{(j+1 + \mu_g) \sqrt{(j+\mu_g)(j+1-\mu_g)}}{(2j+3)(2j+1)(j+1)} \times \sqrt{\cos(2\varepsilon)} \sin \varepsilon,$$

$$b_{\mu_e\mu_e} = \frac{[(j+1)^2 - \mu_e^2] \cos(2\varepsilon) + (j+\mu_e)(j+1 + \mu_e) \sin^2 \varepsilon}{(2j+3)(2j+1)(j+1)},$$

$$b_{\mu_e(\mu_e-1)} = b_{(\mu_e-1)\mu_e} = \frac{(j+\mu_e) \sqrt{(j+1+\mu_e)(j+2-\mu_e)}}{(2j+3)(2j+1)(j+1)} \times \sqrt{\cos(2\varepsilon)} \sin \varepsilon.$$

For the $2j$ linear independent equations for determining $C_n(\varepsilon)$ we select those obtained from (21) by taking the off-diagonal matrix elements between the states $|g, \mu\rangle$ and $|g, j\rangle$, where μ runs through $2j$ values:

$$\left\langle g, \mu \left| \left[\frac{1}{\gamma} \sum_{n=0}^{2j} C_n(\varepsilon) \hat{\gamma} \{ (\hat{V} \hat{V}^\dagger)^{n+1} \} - \sum_{n=0}^{2j} C_n(\varepsilon) (\hat{V}^\dagger \hat{V})^{n+1} \right] \right| g, j \right\rangle = 0, \quad (33)$$

with $\mu = -j, -j+1, \dots, j-1$. This system of equations is of triangular form. Indeed, by direct multiplication we can show that the matrices $(\hat{V}^\dagger \hat{V})^k$ and $(\hat{V} \hat{V}^\dagger)^k$ in the base (31) are real and symmetric and contain $2k+1$ nonzero diagonals for $0 \leq k \leq 2j$. In addition, the matrices $(\hat{V}^\dagger \hat{V})^{2j}$ and $(\hat{V} \hat{V}^\dagger)^{2j+1}$ do not have zero diagonals, while in the matrix $(\hat{V} \hat{V}^\dagger)^{2j+1}$ only the first column and the first row consist of zeros. Hence for a fixed μ only terms with $n=2j, 2j-1, \dots, j-\mu-1$ contribute to (33). The terms with $n < j-\mu-1$ contribute nothing because, first,

$$\langle g, \mu | (\hat{V}^\dagger \hat{V})^{n+1} | g, j \rangle = 0, \quad (n=0, 1, \dots, j-\mu-2)$$

and, second, since the arrival operator $\hat{\gamma}\{\dots\}$ (see (12)) does not provide additional coherence, we have

$$\langle g, \mu | \hat{\gamma}\{(\hat{V}\hat{V}^\dagger)^{n+1}\} | g, j \rangle = 0, \quad (n=0, 1, \dots, j-\mu-2).$$

Thus, at $\mu = -j$ Eq. (33) couples only two coefficients, $C_{2j}(\varepsilon)$ and $C_{2j-1}(\varepsilon)$; at $\mu = -j+1$ this equation couples three coefficients, $C_{2j}(\varepsilon)$, $C_{2j-1}(\varepsilon)$, and $C_{2j-2}(\varepsilon)$; etc.:

$$\begin{aligned} 0 &= A_{2j}^{2j-1} C_{2j}(\varepsilon) + A_{2j-1}^{2j-1} C_{2j-1}(\varepsilon), \\ 0 &= A_{2j}^{2j-2} C_{2j}(\varepsilon) + A_{2j-1}^{2j-2} C_{2j-1}(\varepsilon) \\ &\quad + A_{2j-2}^{2j-2} C_{2j-2}(\varepsilon), \\ &\vdots \\ 0 &= A_{2j}^k C_{2j}(\varepsilon) + \dots + A_{2j-1}^k C_{2j-1}(\varepsilon) + \dots + A_{2j-k}^k C_{2j-k}(\varepsilon), \\ &\vdots \\ 0 &= A_{2j}^0 C_{2j}(\varepsilon) + A_{2j-1}^0 C_{2j-1}(\varepsilon) + \dots + A_0^0 C_0(\varepsilon), \end{aligned} \quad (34)$$

where

$$\begin{aligned} A_l^k &= \left\langle g, j-k-1 \left| \left[\frac{1}{\gamma} \hat{\gamma}\{(\hat{V}\hat{V}^\dagger)^{l+1}\} - (\hat{V}^\dagger\hat{V})^{l+1} \right] \right| g, j \right\rangle, \\ k &= 2j-1, 2j-2, \dots, 0, \quad l = 2j, 2j-1, \dots, k. \end{aligned} \quad (35)$$

We can show that all the diagonal coefficients satisfy $A_k^k \neq 0$, i.e., (34) is indeed a system of linearly independent equations. The triangular form of the system (34) makes it possible to write the recurrence formula for the coefficients $C_n(\varepsilon)$ explicitly:

$$C_{2j}(\varepsilon) = 1, \quad C_n(\varepsilon) = -\frac{1}{A_n^n} \sum_{q=n+1}^{2j} A_q^n C_q(\varepsilon), \quad (36)$$

where $n = 2j-1, 2j-2, \dots, 0$. Thus, by selecting the basis (31) and the off-diagonal matrix elements in (31) in a specific form we solve the problem of determining the $C_n(\varepsilon)$ by Gauss's method.

It would seem that the system of equations for the coefficients $C_n(\varepsilon)$ acquires a triangular form similar to (34) if we take the off-diagonal matrix elements between the states $\langle g, -j |$ and $| g, \mu \rangle$ ($\mu = j, j-1, \dots, -j+1$):

$$\begin{aligned} 0 &= \widetilde{A}_{2j}^{2j-1} C_{2j}(\varepsilon) + \widetilde{A}_{2j-1}^{2j-1} C_{2j-1}(\varepsilon), \\ 0 &= \widetilde{A}_{2j}^{2j-2} C_{2j}(\varepsilon) + \widetilde{A}_{2j-1}^{2j-2} C_{2j-1}(\varepsilon) + \widetilde{A}_{2j-2}^{2j-2} C_{2j-2}(\varepsilon), \\ &\vdots \\ 0 &= \widetilde{A}_{2j}^0 C_{2j}(\varepsilon) + \widetilde{A}_{2j-1}^0 C_{2j-1}(\varepsilon) + \dots + \widetilde{A}_0^0 C_0(\varepsilon). \end{aligned}$$

However, for all values of j the coefficient $\widetilde{A}_{2j-2}^{2j-2}$ is zero, and the first two equations are linearly dependent.

Below we give the results of calculations of the coefficients $C_n(\varepsilon)$ ($n = 2j, 2j-1, \dots, 0$) by Eqs. (35) and (36) for eleven transitions $j_g = j \rightarrow j_e = j+1$ at $j = 0, 1/2, 1, \dots, 5$. We also give the coefficients $\alpha_0(\varepsilon)$ and $\alpha_1(\varepsilon)$, which determine the normalization constant β (see (23)).

3.3.1. The $j_g = 0 \rightarrow j_e = 1$ transition

$$C_0 = 1, \quad \alpha_0 = 1, \quad \alpha_1 = \frac{1}{3}.$$

3.3.2. The $j_g = 1/2 \rightarrow j_e = 3/2$ transition

$$C_1 = 1, \quad C_0 = -\frac{1}{12}, \quad \alpha_0 = \frac{1}{6}, \quad \alpha_1 = \frac{5 - \cos(4\varepsilon)}{144}.$$

3.3.3. The $j_g = 1 \rightarrow j_e = 2$ transition

$$\begin{aligned} C_2 = 1, \quad C_1 &= -\frac{2}{15}, \quad C_0 = \frac{7 + 2 \cos(4\varepsilon)}{1500}, \\ \alpha_0 &= \frac{21 - 4 \cos(4\varepsilon)}{1500}, \quad \alpha_1 = \frac{7 - 3 \cos(4\varepsilon)}{3000}. \end{aligned}$$

3.3.4. The $j_g = 3/2 \rightarrow j_e = 5/2$ transition

$$\begin{aligned} C_3 = 1, \quad C_2 &= -\frac{1}{6}, \quad C_1 = \frac{155 + 29 \cos(4\varepsilon)}{16\,800}, \\ C_0 &= -\frac{27 + 13 \cos(4\varepsilon)}{168\,000}, \quad \alpha_0 = \frac{69 - 29 \cos(4\varepsilon)}{84\,000}, \\ \alpha_1 &= \frac{169 - 108 \cos(4\varepsilon) + 3 \cos(8\varepsilon)}{1\,440\,000}. \end{aligned}$$

3.3.5. The $j_g = 2 \rightarrow j_e = 3$ transition

$$\begin{aligned} C_4 = 1, \quad C_3 &= -\frac{4}{21}, \quad C_2 = \frac{440 + 59 \cos(4\varepsilon)}{33\,075}, \\ C_1 &= -\frac{1361 + 497 \cos(4\varepsilon)}{3\,472\,875}, \\ C_0 &= \frac{2[1083 + 737 \cos(4\varepsilon) + 32 \cos(8\varepsilon)]}{510\,512\,625}, \\ \alpha_0 &= \frac{2[9615 - 6094 \cos(4\varepsilon) + 167 \cos(8\varepsilon)]}{510\,512\,625}, \\ \alpha_1 &= \frac{269 - 220 \cos(4\varepsilon) + 15 \cos(8\varepsilon)}{56\,723\,625}. \end{aligned}$$

3.3.6. The $j_g = 5/2 \rightarrow j_e = 7/2$ transition

$$\begin{aligned} C_5 = 1, \quad C_4 &= -\frac{5}{24}, \quad C_3 = \frac{1495 + 153 \cos(4\varepsilon)}{88\,704}, \\ C_2 &= -\frac{22\,589 + 6507 \cos(4\varepsilon)}{34\,771\,968}, \\ C_1 &= \frac{5[4713 + 2596 \cos(4\varepsilon) + 83 \cos(8\varepsilon)]}{1\,947\,230\,208}, \\ C_0 &= -\frac{25[1167 + 988 \cos(4\varepsilon) + 85 \cos(8\varepsilon)]}{327\,134\,674\,944}, \\ \alpha_0 &= \frac{25[4659 - 3796 \cos(4\varepsilon) + 257 \cos(8\varepsilon)]}{81\,783\,668\,736}, \\ \alpha_1 &= \frac{25[3562 - 3435 \cos(4\varepsilon) + 390 \cos(8\varepsilon) - 5 \cos(12\varepsilon)]}{555\,137\,630\,208}. \end{aligned}$$

3.3.7. The $j_g=3 \rightarrow j_e=4$ transition

$$C_6=1, \quad C_5=-\frac{2}{9}, \quad C_4=\frac{1175+96 \cos(4\varepsilon)}{58\,968}, \quad C_3=-\frac{15\,799+3709 \cos(4\varepsilon)}{17\,336\,592},$$

$$C_2=\frac{2892\,489+1318\,732 \cos(4\varepsilon)+32\,705 \cos(8\varepsilon)}{128\,152\,088\,064},$$

$$C_1=-\frac{5[313\,723+226\,384 \cos(4\varepsilon)+15\,621 \cos(8\varepsilon)]}{5\,382\,387\,698\,688},$$

$$C_0=\frac{25[24\,295+23\,964 \cos(4\varepsilon)+3165 \cos(8\varepsilon)+64 \cos(12\varepsilon)]}{387\,531\,914\,305\,536},$$

$$\alpha_0=\frac{25[710\,311-683\,706 \cos(4\varepsilon)+77\,325 \cos(8\varepsilon)-986 \cos(12\varepsilon)]}{387\,531\,914\,305\,536},$$

$$\alpha_1=\frac{25[6070-6573 \cos(4\varepsilon)+1050 \cos(8\varepsilon)-35 \cos(12\varepsilon)]}{32\,520\,160\,641\,024}.$$

3.3.8. The $j_g=7/2 \rightarrow j_e=9/2$ transition

$$C_7=1, \quad C_6=-\frac{7}{30}, \quad C_5=\frac{7[697+47 \cos(4\varepsilon)]}{216\,000}, \quad C_4=-\frac{6797+1335 \cos(4\varepsilon)}{5\,832\,000},$$

$$C_3=\frac{101\,098\,369+38\,951\,668 \cos(4\varepsilon)+779\,019 \cos(8\varepsilon)}{2\,911\,334\,400\,000},$$

$$C_2=-\frac{52\,718\,937+32\,730\,548 \cos(4\varepsilon)+1861\,491 \cos(8\varepsilon)}{87\,340\,032\,000\,000},$$

$$C_1=\frac{7[2048\,696\,150+1788\,745\,935 \cos(4\varepsilon)+197\,248\,026 \cos(8\varepsilon)+3144\,641 \cos(12\varepsilon)]}{2\,490\,238\,992\,384\,000\,000},$$

$$C_0=-\frac{49[2400\,366+2642\,251 \cos(4\varepsilon)+468\,418 \cos(8\varepsilon)+20\,613 \cos(12\varepsilon)]}{4\,980\,477\,984\,768\,000\,000},$$

$$\alpha_0=\frac{49[65\,149\,338-70\,477\,367 \cos(4\varepsilon)+11\,232\,886 \cos(8\varepsilon)-373\,209 \cos(12\varepsilon)]}{2\,490\,238\,992\,384\,000\,000},$$

$$\alpha_1=\frac{49[338\,377-398\,888 \cos(4\varepsilon)+81\,620 \cos(8\varepsilon)-4760 \cos(12\varepsilon)+35 \cos(16\varepsilon)]}{139\,314\,069\,504\,000\,000}.$$

3.3.9. The $j_g=4 \rightarrow j_e=5$ transition

$$C_8=1, \quad C_7=-\frac{8}{33}, \quad C_6=\frac{14[2470+141 \cos(4\varepsilon)]}{1\,388\,475}, \quad C_5=-\frac{2[1450\,183+243\,387 \cos(4\varepsilon)]}{2\,061\,885\,375},$$

$$C_4=\frac{10\,851\,455+3597\,104 \cos(4\varepsilon)+59\,763 \cos(8\varepsilon)}{226\,807\,391\,250},$$

$$C_3=-\frac{56\,724\,509+30\,650\,862 \cos(4\varepsilon)+1469\,889 \cos(8\varepsilon)}{56\,134\,829\,334\,375},$$

$$C_2=\frac{3153\,457\,345+2442\,690\,541 \cos(4\varepsilon)+229\,168\,019 \cos(8\varepsilon)+2977\,919 \cos(12\varepsilon)}{240\,818\,417\,844\,468\,750},$$

$$C_1=-\frac{7[547\,223\,774+548\,709\,937 \cos(4\varepsilon)+83\,763\,746 \cos(8\varepsilon)+3071\,183 \cos(12\varepsilon)]}{397\,350\,389\,443\,37343\,750},$$

$$C_0 = \frac{98[38\,190\,410 + 45\,612\,597 \cos(4\varepsilon) + 10\,071\,846 \cos(8\varepsilon) + 713\,051 \cos(12\varepsilon) + 8192 \cos(16\varepsilon)]}{12\,019\,849\,280\,662\,046\,484\,375},$$

$$\alpha_0 = \frac{98}{12\,019\,849\,280\,662\,046\,484\,375} [3886\,404\,435 - 4578\,763\,678 \cos(4\varepsilon) + 935\,599\,096 \cos(8\varepsilon) - 54451074 \cos(12\varepsilon) + 399317 \cos(16\varepsilon)],$$

$$\alpha_1 = \frac{49[599\,569 - 753\,768 \cos(4\varepsilon) + 186\,228 \cos(8\varepsilon) - 15\,960 \cos(12\varepsilon) + 315 \cos(16\varepsilon)]}{10\,877\,691\,656\,707\,734\,375}.$$

3.3.10. The $j_g=9/2 \rightarrow j_e=11/2$ transition

$$C_9 = 1, \quad C_8 = -\frac{1}{4}, \quad C_7 = \frac{6755 + 333 \cos(4\varepsilon)}{250\,800}, \quad C_6 = -\frac{810\,761 + 118287 \cos(4\varepsilon)}{496\,584\,000},$$

$$C_5 = \frac{304\,103\,347 + 88\,056\,060 \cos(4\varepsilon) + 1243\,857 \cos(8\varepsilon)}{4952\,597\,760\,000},$$

$$C_4 = -\frac{53\,684\,221\,357 + 25\,535\,542\,692 \cos(4\varepsilon) + 1052\,208\,687 \cos(8\varepsilon)}{35\,955\,859\,737\,600\,000},$$

$$C_3 = \frac{187\,307\,519\,134 + 129\,416\,693\,787 \cos(4\varepsilon) + 10\,500\,196\,946 \cos(8\varepsilon) + 114\,166\,005 \cos(12\varepsilon)}{7910\,289\,142\,272\,000\,000},$$

$$C_2 = -\frac{4886\,243\,962 + 4455\,970\,441 \cos(4\varepsilon) + 592\,969\,494 \cos(8\varepsilon) + 18\,474\,983 \cos(12\varepsilon)}{20\,473\,689\,544\,704\,000\,000},$$

$$C_1 = \frac{7}{16\,590\,513\,094\,391\,808\,000\,000} [3315\,809\,101 + 3685\,574\,600 \cos(4\varepsilon) + 717\,658\,724 \cos(8\varepsilon) + 43\,558\,904 \cos(12\varepsilon) + 412303 \cos(16\varepsilon)],$$

$$C_0 = -\frac{49}{243\,327\,525\,384\,413\,184\,000\,000} [18\,106\,229 + 23\,015\,384 \cos(4\varepsilon) + 6026\,372 \cos(8\varepsilon) + 595\,560 \cos(12\varepsilon) + 15\,815 \cos(16\varepsilon)],$$

$$\alpha_0 = \frac{49}{60\,831\,881\,346\,103\,296\,000\,000} [870\,013\,975 - 1093\,383\,576 \cos(4\varepsilon) + 269\,893\,228 \cos(8\varepsilon) - 23\,099\,048 \cos(12\varepsilon) + 455\,101 \cos(16\varepsilon)],$$

$$\alpha_1 = \frac{49}{7649\,257\,763\,930\,112\,000\,000} [8613\,290 - 11\,386\,350 \cos(4\varepsilon) + 3263\,400 \cos(8\varepsilon) - 372\,435 \cos(12\varepsilon) + 13\,230 \cos(16\varepsilon) - 63 \cos(20\varepsilon)].$$

3.3.11. The $j_g=5 \rightarrow j_e=6$ transition

$$C_{10} = 1, \quad C_9 = -\frac{10}{39}, \quad C_8 = \frac{4485 + 194 \cos(4\varepsilon)}{156\,156}, \quad C_7 = -\frac{185\,201 + 23\,823 \cos(4\varepsilon)}{100\,486\,386},$$

$$C_6 = \frac{140\,482\,351 + 35\,991\,420 \cos(4\varepsilon) + 440\,331 \cos(8\varepsilon)}{1872\,147\,502\,368},$$

$$C_5 = -\frac{41\,899\,041\,259 + 17\,728\,511\,636 \cos(4\varepsilon) + 637\,577\,869 \cos(8\varepsilon)}{20\,614\,216\,148\,574\,048},$$

$$C_4 = \frac{1}{1804\,073\,740\,458\,606\,384\,768} [67\,366\,047\,034\,556 + 41\,790\,617\,770\,797 \cos(4\varepsilon) + 2973\,545\,622\,054 \cos(8\varepsilon) + 27\,624\,757\,535 \cos(12\varepsilon)],$$

$$C_3 = -\frac{5}{773\,947\,634\,656\,742\,139\,065\,472} [71\,725\,965\,608\,861 + 59\,574\,531\,611\,847 \cos(4\varepsilon) + 6990\,847\,337\,307 \cos(8\varepsilon) + 188\,213\,423\,921 \cos(12\varepsilon)],$$

$$C_2 = \frac{25}{442\,698\,047\,023\,656\,503\,545\,449\,984} [66\,345\,264\,612\,487 + 68\,407\,666\,859\,323 \cos(4\varepsilon) + 11\,839\,482\,235\,349 \cos(8\varepsilon) + 625\,064\,156\,281 \cos(12\varepsilon) + 4987\,772\,048 \cos(16\varepsilon)],$$

$$C_1 = -\frac{125}{6029\,125\,783\,274\,560\,000\,666\,604\,544} [863\,488\,388\,287 + 1037\,909\,611\,284 \cos(4\varepsilon) + 244\,316\,706\,564 \cos(8\varepsilon) + 21\,148\,019\,308 \cos(12\varepsilon) + 480\,836\,541 \cos(16\varepsilon)],$$

$$C_0 = \frac{4375}{226\,427\,168\,305\,200\,142\,247\,256\,926\,208} [1986\,067\,779 + 2650\,850\,296 \cos(4\varepsilon) + 795\,453\,436 \cos(8\varepsilon) + 101\,244\,360 \cos(12\varepsilon) + 4466\,945 \cos(16\varepsilon) + 32\,768 \cos(20\varepsilon)],$$

$$\alpha_0 = \frac{4375}{113\,213\,584\,152\,600\,071\,123\,628\,463\,104} [362\,584\,767\,369 - 479\,211\,795\,182 \cos(4\varepsilon) + 137\,263\,299\,604 \cos(8\varepsilon) - 15\,650\,817\,399 \cos(12\varepsilon) + 555\,293\,347 \cos(16\varepsilon) - 2640\,347 \cos(20\varepsilon)],$$

$$\alpha_1 = \frac{30\,625}{467\,341\,936\,646\,439\,922\,078\,961\,664} [156\,337\,790 - 21\,515\,450 \cos(4\varepsilon) + 6952\,440 \cos(8\varepsilon) - 992\,145 \cos(12\varepsilon) + 53\,130 \cos(16\varepsilon) - 693 \cos(20\varepsilon)].$$

Interestingly, the coefficients $C_n(\varepsilon)$ and $\alpha_i(\varepsilon)$ are linear combinations of the functions $\cos(4m\varepsilon)$:

$$C_n(\varepsilon) = \sum_{m=0}^{[j-n/2]} X_m^n \cos(4m\varepsilon),$$

$$\alpha_0(\varepsilon) = \sum_{m=0}^{[j]} Y_m \cos(4m\varepsilon),$$

$$\alpha_1(\varepsilon) = \sum_{m=0}^{[j+1/2]} Z_m \cos(4m\varepsilon),$$

where the symbol $[p]$ in the upper limit of the sums stands, as usual, for the integral part of p . This fact considerably simplifies the process of finding the coefficients $C_n(\varepsilon)$ for large j , since now the numerical coefficients X_m^n can be determined by numerically solving the recurrence relations (36) at certain points $\{\varepsilon_r\}$. To determine all the coefficients X_m^n for a given j , when the number of harmonics $\cos(4m\varepsilon)$ is $[j+1]$, we must take the same number of points differing in absolute value.

3.4. Verification of the results and the applicability range

As noted earlier, the steady-state solution has the form (22)–(24) if the theorem (17) is valid. Since we do not have the proof of (17) in the general case, for the

$j_g = j \rightarrow j_e = j+1$ transitions ($j=0, 1/2, 1, \dots, 5$) we found by direct substitution that (22)–(24) constitute the solution of the initial system of equations (14) and (15). For $j=11/2, 6, \dots, 10$ the validity of (17) was established numerically. Finally, we were able to establish the validity of (17) for arbitrary j in the case where the field polarization is close to circular (see the Appendix).

We also compared our results with those known for the particular cases of circular and linear polarizations. Note that when the field is circularly polarized, the atoms are pumped to the outermost Zeeman sublevels $|e, j+1\rangle$ and $|g, j\rangle$. i.e., the problem is reduced to the well-known two-level model, and the steady-state solution has the form

$$\rho_{\mu_e \mu_e'}^{ee} = \delta_{\mu_e \mu_e'} \delta_{\mu_e (j+1)} \frac{S/(2j+3)}{1+2S/(2j+3)}, \quad (37)$$

$$\rho_{\mu_g \mu_g'}^{gg} = \delta_{\mu_g \mu_g'} \delta_{\mu_g j} \frac{1+S/(2j+3)}{1+2S/(2j+3)}.$$

Smirnov⁴ was the first to obtain a steady-state solution in the case of linear polarization for the $j_g = j \rightarrow j_e = j+1$ transitions (with j and S arbitrary):

$$\rho_{\mu_e \mu'_e}^{ee} = \tilde{\beta} \delta_{\mu_e \mu'_e} \frac{S[(j+1)^2 - \mu_e^2]}{(j+1)(2j+1)(2j+3)[(j+1-\mu_e)!(j+1+\mu_e)!]^2},$$

$$\rho_{\mu_g \mu'_g}^{gg} = \tilde{\beta} \delta_{\mu_g \mu'_g} \frac{(j+1)(2j+1)(2j+3) + S[(j+1)^2 - \mu_g^2]}{(j+1)(2j+1)(2j+3)[(j+1-\mu_g)!(j+1+\mu_g)!]^2},$$

where

$$\tilde{\beta} = \frac{[(2j+2)!]^4}{(4j+4)! - 2[(2j+2)!]^2 + S(4j+4)! 4(j+1)/[(2j+3)(4j+3)]}$$

is a normalization constant. Our results (22) and (23) become (37) at $\varepsilon = \pi/4$ and (38) at $\varepsilon = 0$.

Note that (17) is generally valid only for steady-state solutions, since for arbitrary ellipticity the arrival operator (12) induces coherence between the eigenstates of the matrix $\hat{V}^\dagger \hat{V}$ (see Ref. 9), with the result that the time-dependent solution (6)–(10) does not satisfy (17). We also note that for an isotropic arrival operator $\hat{\gamma}\{\hat{\rho}^{ee}\}$ of general form with arbitrary rates γ_κ of transfer of multipole moments of rank $\kappa \geq 1$ from the excited state to the ground state,

$$\gamma_{\mu_g \mu'_g} \{\hat{\rho}^{ee}\} = \sum_{\kappa, q, \mu_e, \mu'_e} \gamma_\kappa (-1)^{j_g - \mu_g} \begin{pmatrix} j_g & \kappa & j_g \\ -\mu_g & q & \mu'_g \end{pmatrix} \times (-1)^{j_e - \mu'_e + q} \begin{pmatrix} j_e & \kappa & j_e \\ -\mu'_e & -q & \mu_e \end{pmatrix} \rho_{\mu_e \mu'_e}^{ee},$$

$$\gamma_0 = \gamma \sqrt{\frac{2j_e + 1}{2j_g + 1}},$$

(17) does not hold. We checked this directly for the $j_g = 1 \rightarrow j_e = 2$ transition in an elliptically polarized field. Thus, the range within which (17) operates is, apparently, limited to isotropic arrival operators of the form (12), which corresponds to (40) at

$$\gamma_\kappa = \gamma(2j_e + 1)(2\kappa + 1)(-1)^{j_g + j_e + \kappa + 1} \begin{pmatrix} j_g & j_e & 1 \\ j_e & j_g & \kappa \end{pmatrix}.$$

We discovered that the commutation relation (17) is violated if isotropic depolarizing collisions¹¹ are taken into account.

4. DEPENDENCE OF ABSORPTION ON THE LIGHT'S ELLIPTICITY

Let us examine the dependence of absorption on the ellipticity when a plane-polarized wave propagates along the z axis in a gas of atoms with the $j_g = j \rightarrow j_e = j + 1$ transition. As is well known, in conditions of a steady state interaction the variation of the intensity I of a monochromatic wave is determined by the equation

$$\frac{\partial I}{\partial z} = -2\omega \operatorname{Im}\{E_0(\mathbf{e} \cdot \mathbf{P}^*)\},$$

where \mathbf{P} is the positive-frequency component of the medium's polarization vector (the average dipole moment). In our case Eq. (24) yields

$$E_0(\mathbf{e} \cdot \mathbf{P}^*) = i\hbar |\Omega|^2 \left\langle \frac{\beta}{\gamma/2 + i\delta} \right\rangle_v \operatorname{Tr} \left\{ \sum_{n=0}^{2j} C_n(\varepsilon) (\hat{V}^\dagger \hat{V})^{n+1} \right\},$$

where the symbol $\langle \dots \rangle_v$ stands for averaging over the velocity distribution of the atoms normalized to the concentration. Plugging (42) into (41) and combining the result with (22) yields

$$\frac{\partial I}{\partial z} = -\hbar \omega \gamma \langle \operatorname{Tr}\{\hat{\rho}^{ee}\} \rangle_v = -\hbar \omega \gamma \left\langle \frac{S\alpha_1(\varepsilon)/\alpha_0(\varepsilon)}{1 + 2S\alpha_1(\varepsilon)/\alpha_0(\varepsilon)} \right\rangle_v.$$

Thus, the nonlinear coefficient of steady absorption at a fixed field intensity is proportional to the total population of the excited state. At low intensities, in the limit $S \ll 1$, the dependence of the absorption coefficient on the ellipticity angle ε is determined by the ratio $\alpha_1(\varepsilon)/\alpha_0(\varepsilon)$, as Eq. (43) implies. The diagrams of this dependence at $j=0, 1/2, 1, \dots, 5$ are depicted in Fig. 3, which shows that the absorption is at its maximum for circular polarization ($\varepsilon = \pm \pi/4$) and at its minimum for linear polarization ($\varepsilon = 0$) (with the exception of the $j_g = 0 \rightarrow j_e = 1$ transition, in which case the absorption coefficient is independent of the ellipticity of the light). For all values of j the ratios $\alpha_1(\pm \pi/4)/\alpha_0(\pm \pi/4)$ and $\alpha_1(0)/\alpha_0(0)$ can be found from (37)–(39):

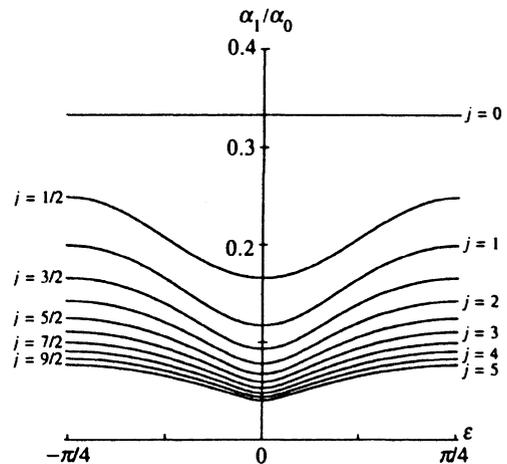


FIG. 3. The ratio $\alpha_1(\varepsilon)/\alpha_0(\varepsilon)$ as a function of ε at $j=0, 1/2, 1, \dots, 5$. As Eq. (43) implies, when saturation is weak ($S \ll 1$), the absorption coefficient is proportional to $\alpha_1(\varepsilon)/\alpha_0(\varepsilon)$.

$$\frac{\alpha_1(\pm\pi/4)}{\alpha_0(\pm\pi/4)} = \frac{1}{2j+3},$$

$$\frac{\alpha_1(0)}{\alpha_0(0)} = \frac{(4j+4)! 2(j+1)}{(2j+3)(4j+3)\{(4j+4)! - 2[(2j+2)!]^2\}}. \quad (44)$$

This implies that the ratio of the absorption coefficients at $\varepsilon = \pm\pi/4$ and $\varepsilon = 0$ is

$$\frac{\alpha_1(\pm\pi/4)/\alpha_0(\pm\pi/4)}{\alpha_1(0)/\alpha_0(0)} = \frac{(4j+3)\{(4j+4)! - 2[(2j+2)!]^2\}}{(4j+4)! 2(j+1)}. \quad (45)$$

For $j=1/2$ the ratio is $3/2$; it increases monotonically with j and tends to 2 as $j \rightarrow \infty$. Thus, with the $j_g = j \rightarrow j_e = j+1$ ($j \geq 1/2$) transitions in the case of weak saturation ($S \ll 1$), the absorption coefficient for circularly polarized light exceeds that for linearly polarized light by a factor of 1.5 to 2. This fact must be taken into account in interpreting the data in nonlinear polarization spectroscopy.

As the field intensity grows, the maximum (minimum) of absorption is still determined by the maximum (minimum) of the ratio $\alpha_1(\varepsilon)/\alpha_0(\varepsilon)$. But the dependence of the absorption coefficient on ε becomes weaker and vanishes for $S \gg 1$, when $\text{Tr}\{\hat{\rho}^{ee}\} \approx 1/2$.

Note that for the $j_g = j \rightarrow j_e = j$ transitions (j is a half-integer), as Ref. 2 implies, the situation is reversed: for any intensity the absorption is at its maximum in the case of linear polarization and at its minimum (vanishes) in the case of circular polarization.

5. THE RADIATION FORCE FOR SLOW ATOMS

Let us study the force acting on slow atoms with a resonant transition $j_g = j \rightarrow j_e = j+1$ in the inhomogeneous monochromatic field

$$\mathbf{E}(\mathbf{r}, t) = \mathbf{E}(\mathbf{r})\mathbf{e}(\mathbf{r})e^{-i\omega t} + \text{c.c.},$$

where $E(\mathbf{r})$ is the complex-valued amplitude, and $\mathbf{e}(\mathbf{r})$ is complex-valued unit polarization vector satisfying the condition $\text{Im}(\mathbf{e}\mathbf{e}) = 0$, which fixes the phase of the function $E(\mathbf{r})$. The atomic velocity is assumed to be low:

$$k\mathbf{v} \ll \gamma, \quad k\mathbf{v} \ll \gamma S,$$

so that at each point in space the internal state of atoms adiabatically follows the field vector $E(\mathbf{r})\mathbf{e}(\mathbf{r})$. In these conditions, to determine the velocity-independent component of the force we use the solution (22)–(24) in which we must put $\mathbf{v} = 0$ and introduce the coordinate functions $\Omega(\mathbf{r})$, $\varepsilon(\mathbf{r})$, and $\mathbf{e}(\mathbf{r})$ in accordance with the variation in the amplitude and the field polarization.

Proceeding from the well-known general formula for the dipole force,

$$\mathbf{F}(\mathbf{r}) = -\text{Tr}\{\hat{\rho}(\mathbf{r})\nabla\hat{V}_{E-D}\},$$

we arrive at the following expression for the force for $j_g = j \rightarrow j_e = j+1$ transitions:

$$\mathbf{F} = -\frac{i\hbar\beta}{\gamma/2+i\delta} \text{Tr}\left\{\sum_{n=0}^{2j} C_n(\varepsilon)(\hat{V}^\dagger\hat{V})^n(\Omega^*\hat{V}^\dagger)\nabla(\Omega\hat{V})\right\} + \text{c.c.}$$

$$= \mathbf{F}_1 + \mathbf{F}_2 + \mathbf{F}_3 + \mathbf{F}_4,$$

$$\mathbf{F}_1 = -\frac{i\hbar\gamma S\alpha_1}{2(\alpha_0+2S\alpha_1)} \nabla \ln \frac{\Omega}{\Omega^*},$$

$$\mathbf{F}_2 = -\frac{i\hbar\gamma S}{2(\alpha_0+2S\alpha_1)} \times \left[\sum_{n=0}^{2j} C_n(\varepsilon) \text{Tr}\{(\hat{V}^\dagger\hat{V})^n(\hat{V}^\dagger\nabla\hat{V})\} - \text{c.c.} \right], \quad (46)$$

$$\mathbf{F}_3 = -\frac{\hbar\delta\alpha_1}{\alpha_0+2S\alpha_1} \nabla S,$$

$$\mathbf{F}_4 = -\frac{\hbar\delta S}{\alpha_0+2S\alpha_1} \sum_{n=0}^{2j} \frac{C_n(\varepsilon)}{n+1} \nabla [\text{Tr}\{(\hat{V}^\dagger\hat{V})^{n+1}\}].$$

The fact that \mathbf{F} can be split into four terms corresponds to the classification of Ref. 12: \mathbf{F}_1 and \mathbf{F}_2 are the spontaneous radiation forces, which are related to processes of induced absorption and subsequent spontaneous emission, with \mathbf{F}_1 determined by the gradient of the field's phase and \mathbf{F}_2 by the gradient of the orientation of the polarization ellipse $\mathbf{e}(\mathbf{r})$, and \mathbf{F}_3 and \mathbf{F}_4 are the stimulated radiation forces, which are related to processes of coherent re-emission of photons from one mode to another, with \mathbf{F}_3 determined by the gradient of the field's intensity and \mathbf{F}_4 by the gradient of the ellipticity.

As an example, we consider four characteristic cases in which plane waves propagate along the z axis.

5.1. An elliptically polarized traveling wave

Here the saturation parameter S and the polarization vector \mathbf{e} do not depend on coordinates, and $\Omega(z) = \Omega_0 \exp(ikz)$. Then the force is spatially homogeneous and is determined by the number of spontaneously scattered photons per unit time ($\gamma \text{Tr}\{\hat{\rho}^{ee}\}$):

$$F = F_1 = \hbar k \gamma \frac{S\alpha_1}{\alpha_0+2S\alpha_1}. \quad (47)$$

5.2. Oppositely propagating waves $\sigma_+ - \sigma_-$

Such a configuration is a linear combination of two oppositely propagating plane waves with orthogonal circular polarizations \mathbf{e}_{+1} and \mathbf{e}_{-1} and intensities I_+ and I_- . In this case the phase and the saturation parameter are coordinate-independent, and the polarization vector $\mathbf{e}(z)$ is an ellipse rotating in space about the z axis, with the ellipticity constant ε determined by the following relationship:

$$\sin(2\varepsilon) = \frac{I_+ - I_-}{I_+ + I_-}.$$

The force is spatially homogeneous and proportional to the rate of variation of the z -component of the angular momentum caused by spontaneous emission:

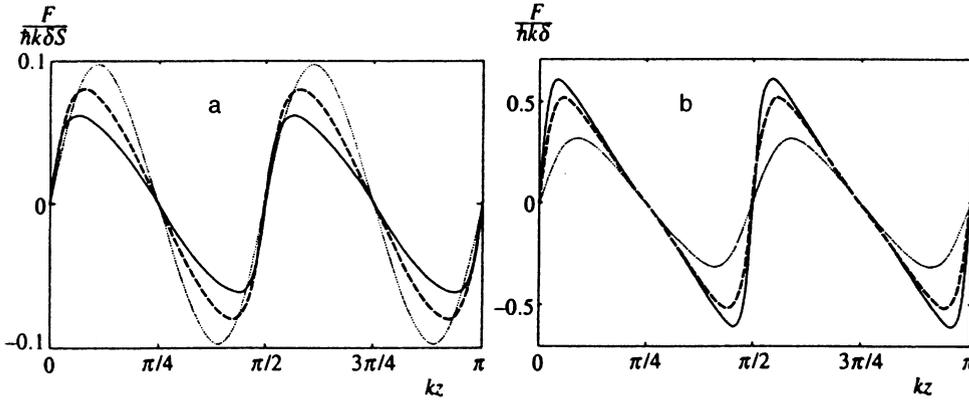


FIG. 4. The spatial dependence of the $lin \perp lin$ field configuration for three transitions with $j = 1$ (dotted curves), 3 (dashed curves), and 5 (solid curves) for (a) weak saturation ($S \ll 1$; calculations via (50)), and (b) strong saturation ($S \gg 1$; calculations via (51)).

$$\begin{aligned}
 F = F_2 &= \hbar k [\gamma \text{Tr}\{\hat{J}_z \hat{\rho}^{ee}\} - \text{Tr}\{\hat{J}_z \hat{\gamma}\{\hat{\rho}^{ee}\}\}] \\
 &= \hbar k \gamma \frac{S}{\alpha_0 + 2S\alpha_1} \sum_{n=0}^{2j} C_n(\varepsilon) [\text{Tr}\{\hat{J}_z (\hat{V} \hat{V}^\dagger)^{n+1}\} \\
 &\quad - \text{Tr}\{\hat{J}_z (\hat{V}^\dagger \hat{V})^{n+1}\}]. \quad (48)
 \end{aligned}$$

5.3. An elliptically polarized standing wave

In this case the phase and polarization are coordinate-independent, and $S(z) = S_0[1 + \cos(2kz)]$, where S_0 is the saturation parameter averaged over the period. For this force we have

$$\begin{aligned}
 F(z) = F_3(z) &= -\frac{\partial}{\partial z} U(z), \\
 U(z) &= \frac{\hbar \delta}{2} \ln \left[1 + \frac{2\alpha_1}{\alpha_0} S(z) \right]. \quad (49)
 \end{aligned}$$

Clearly, the expression for the optical potential coincides with the one in the theory of a two-level atom¹³ with a renormalized saturation parameter $\tilde{S} = 2\alpha_1 S/\alpha_0$, which is now ellipticity-dependent.

5.4. The $lin \perp lin$ field configuration

The $lin \perp lin$ field configuration is a linear combination of two oppositely propagating plane waves with orthogonal linear polarizations and equal intensities. For it the gradients of the phase and intensity vanish, the orientation of the ellipse axes is constant, and the ellipticity angle is spatially dependent: $\varepsilon(z) = kz$.

In this case, we have $F(z) = F_4(z)$. For $S \ll 1$,

$$F_4(z) \approx \hbar k \delta S \frac{1}{\alpha_0} \sum_{n=0}^{2j} \frac{C_n(\varepsilon)}{n+1} \frac{\partial}{\partial \varepsilon} [\text{Tr}\{(\hat{V}^\dagger \hat{V})^{n+1}\}], \quad (50)$$

while for $S \gg 1$,

$$F_4(z) \approx \hbar k \delta \frac{1}{2\alpha_1} \sum_{n=0}^{2j} \frac{C_n(\varepsilon)}{n+1} \frac{\partial}{\partial \varepsilon} [\text{Tr}\{(\hat{V}^\dagger \hat{V})^{n+1}\}]. \quad (51)$$

The results of calculations of the force by Eq. (5) for three transitions $j_g = j \rightarrow j_e = j+1$ ($j = 1, 3, 5$) are shown in Fig. 4. We see that the F vs z dependence becomes more jagged as j grows.

Note that Alekseev,¹⁴ using perturbation theory techniques with allowance for the finiteness of the atom-field interaction time, arrived at an expression for the radiation force for an arbitrary initial distribution of the atoms over the Zeeman sublevels of the ground state.

6. CONCLUSION

The obtained exact steady-state solution of the problem of optically pumping the $j_g = j \rightarrow j_e = j+1$ transition is of interest from the fundamental viewpoint, just as any exact solution of a quantum mechanical problem is. In addition, this solution can be used in various approximations related to the interaction of atoms and polarized radiation. For instance, in Secs. 5 and 6 we examined the dependence of the absorption coefficient on the ellipticity and calculated the gradient force. Other possible applications are the nonlinear self-consistent problem of propagation of an elliptically polarized wave in a medium of resonant atoms and the polarization spectroscopy with a strong pump field and a weak probe wave. Note that the closed transitions $j_g = j \rightarrow j_e = j+1$ are always present in the $D2$ line of alkali metals and are often used in experiments in spectroscopy and laser cooling (in the presence of hyperfine splitting, j_g and j_e are the total atomic angular momenta F_g and F_e , respectively).

Combining the results of the present work with those of Refs. 1 and 2, we may assume that we have found all steady-state solutions of the problem of the resonant interaction of atoms and elliptically polarized light that allow for radiative relaxation for all closed dipole transitions (including inter-Raman and magnetic-dipole). The only unsolved problem is Theorem (17), which has not been proved in the general case for $j_g = j \rightarrow j_e = j+1$ transitions. However, the results of the present investigation suggest that there are no physical grounds to doubt the validity of (17) for all values of j and ε .

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APPENDIX

As Secs. 3.1 and 3.2 clearly show, the theorem (17) is valid if and only if we can prove that the steady-state solution of Eq. (19) for $\hat{\rho}_0^{gg}$ commutes with $\hat{V}^\dagger \hat{V}$:

$$[\hat{V}^\dagger \hat{V}, \hat{\rho}_0^{gg}] = 0, \quad (\text{A1})$$

i.e., the steady-state density matrix of the ground state for $S \ll 1$ is independent of the detuning δ . Let us prove this for any value of j in the case of a field polarization close to circular, where $\varepsilon = \pi/4 - \xi$ ($\xi \ll 1$) and the unit vector (2) can be represented by the following expansion:

$$\mathbf{e} = -\mathbf{e}_{+1} + \xi \mathbf{e}_{-1} + O(\xi^2). \quad (\text{A2})$$

We write $\hat{\rho}_0^{gg}$ in the form of a power series in ξ :

$$\hat{\rho}_0^{gg} = \hat{\sigma}^{(0)} + \xi \hat{\sigma}^{(1)} + O(\xi^2). \quad (\text{A3})$$

To zeroth order in ξ , where the field's polarization is exactly circular, the solution of Eq. (19) has the form

$$\sigma_{\mu_g, \mu_g'}^{(0)} = \delta_{\mu_g, \mu_g'} \delta_{\mu_g, j}, \quad (\text{A4})$$

and the property (A1) holds in the same approximation.

Setting the factors of the first order in ξ equal to zero in (19) and allowing for (A4), we arrive at the following expression for the matrix element $\sigma_{j, j-2}^{(1)}$:

$$\begin{aligned} & \left\{ \gamma(2j+3)(V_{j+1, j}^{+1})^2 (V_{j-1, j-2}^{+1})^2 - \left(\frac{\gamma}{2} + i\delta \right) (V_{j+1, j}^{+1})^2 \right. \\ & \left. - \left(\frac{\gamma}{2} - i\delta \right) (V_{j-1, j-2}^{+1})^2 \right\} \sigma_{j, j-2}^{(1)} \\ & = \gamma(2j+3)(V_{j+1, j}^{+1})^2 V_{j-1, j}^{-1} V_{j-1, j-2}^{+1} \\ & \quad - \left(\frac{\gamma}{2} - i\delta \right) V_{j-1, j}^{-1} V_{j-1, j-2}^{+1}, \end{aligned} \quad (\text{A5})$$

where

$$V_{\mu_e, \mu_g}^q = (-1)^{j_e - \mu_e} \begin{pmatrix} j_e & 1 & j_g \\ -\mu_e & q & \mu_g \end{pmatrix}.$$

Writing the $3jm$ -symbols in (A5) explicitly and allowing for the hermiticity of $\hat{\sigma}^{(1)}$ yields

$$\sigma_{j, j-2}^{(1)} = \sigma_{j-2, j}^{(1)} = -\frac{\sqrt{(2j-1)j}}{4j+1}. \quad (\text{A6})$$

For the other matrix elements of $\hat{\sigma}^{(1)}$ in (19) we obtain a homogeneous system of equations, which has only a trivial solution. Thus, to first order in ξ , the steady-state solution of Eq. (19) for $\hat{\rho}_0^{gg}$ does not depend on δ , which means that the condition (A1) is met to within the same approximation.

A complete proof of (17) requires extending this procedure to all powers of ξ .

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