

The exact steady-state solution of the problem of optical pumping in an elliptically polarized field for closed atomic transitions $j_g = j \rightarrow j_e = j$ with j a half-integer

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The exact steady-state solution of the problem of optical pumping of the transitions $j_g = j \rightarrow j_e = j$ where j is a half-integer, where and j_g and j_e are the angular momenta of the ground (g) and excited (e) states, is found for light of arbitrary intensity and ellipticity. The properties of the obtained solution are investigated. Finally, the conditions for weak and strong saturation of the transition are established as functions of the ellipticity of the field. © 1995 American Institute of Physics.

1. INTRODUCTION

Problems related to resonant interaction of polarized light and atoms occupy a central place in atomic physics. Generally, this interaction involves exchange of energy, momentum, and angular momentum between field and atoms. We can usually ignore momentum transfer from light to atoms, however, and focus only on the redistribution of atoms among the Zeeman sublevels of degenerate energy levels caused by light-induced and spontaneous transitions. Among the various problems concerned with optical pumping of Zeeman sublevels the most interesting situation occurs when one level is the ground level. In this case light-induced anisotropy is long-lived and makes it possible to gather information about very weak interactions, which is important for many applications, such as high-resolution spectroscopy, magneto-optics, and laser cooling of atoms. Since the problem of optical pumping of the degenerate ground state requires simultaneous study of light-induced and spontaneous processes, the generalized optical Bloch equations (quantum transport equations) for the atomic density matrix are used to describe the medium.¹ Depending on the light intensity and the time it takes the atoms to interact with the field, two limiting cases can be specified:

1. The intensity and interaction time are so small that the redistribution of atoms among the sublevels can be taken into account by perturbation techniques. Earlier Akul'shin *et al.*⁴ applied the method of irreducible tensor operators^{2,3} to this case to obtain the solution of the corresponding Bloch equations for a field of arbitrary ellipticity and for arbitrary Zeeman and hyperfine structures of the levels interacting with the field.

2. The interaction time is so long that perturbation theory becomes inapplicable and one must find the steady-state solution of the Bloch equations corresponding to the interaction of the atoms with an elliptically polarized plane wave. The usual restriction is to consider only two degenerate atomic levels whose total population is conserved (a closed optical transition). The present paper is devoted to this case. Finding the steady-state solution of Bloch equations in analytical form for a field of arbitrary ellipticity and for arbitrary

angular momenta of the ground and excited states is a complex mathematical problem because of the large number of equations.

At present the exact steady-state solution is known for two types of closed optical transitions: $j_g = j \rightarrow j_e = j - 1$ and $j_g = j' \rightarrow j_e = j'$ with j' an integer, where j_g and j_e are the angular momenta of the ground (g) and excited (e) states. For such transitions, as follows from Ref. 5, in the process of optical pumping the atoms accumulate in the so-called coherent population-trapping states $|\psi_0\rangle$, which do not interact with the field. These are superpositions of the wave functions of the Zeeman sublevels of the ground state and satisfy the equation $\hat{V}_{E-D}|\psi_0\rangle = 0$, where $(\hat{V}_{E-D} = -(\hat{\mathbf{d}}\mathbf{E}))$ is the operator of the interaction of light and atoms in the electro-dipole approximation ($\hat{\mathbf{d}}$ is the dipole-moment operator). The steady-state density matrix is constructed from such states $|\psi_0\rangle$.

The steady-state solution of the optical Bloch equations for other types of optical transitions, $j_g = j \rightarrow j_e = j + 1$ and $j_g = j'' \rightarrow j_e = j''$ with j'' a half-integer, has been found for the particular cases of linear and circular polarizations of the field.⁶⁻⁸ The steady-state solution for light of arbitrary ellipticity has been found only for cases with moderate values of j_g and j_e (see Refs. 7 and 9–11).

In this paper we find the steady-state solution of the problem of optical pumping of the closed transition $j_g = j \rightarrow j_e = j$, where j is a half-integer, for light of arbitrary intensity and ellipticity. The solution has a number of interesting properties:

1. The atoms are distributed isotropically among the Zeeman sublevels of the excited state, i.e., the populations of all the sublevels are the same and there is no coherence between them at any intensity of the pump field. The anisotropic properties of the medium are determined here by the distribution of atoms among the Zeeman sublevels of the ground state, which are populated nonuniformly, and the coherence is nonzero.

2. At intensities so high that the transition becomes saturated the distribution of the populations of the ground state also becomes isotropic. However, the value of the saturation intensity strongly depends on the ellipticity of the light field. For instance, when the light is circularly polarized, coherent

trapping of populations occurs and saturation never sets in, no matter how high the intensity.

2. STATEMENT OF THE PROBLEM

Let us examine the resonant interaction of atoms whose ground and excited states form a closed optical transition, $j_g = j \rightarrow j_e = j$, with j a half-integer, and an elliptically polarized plane wave

$$\mathbf{E} = E_0 \mathbf{e} \exp[-i(\omega t - \mathbf{k}\mathbf{r})] + \text{c.c.}, \quad \mathbf{e} = \sum_{q=0,\pm 1} e^q \mathbf{e}_q, \quad (1)$$

where \mathbf{e} is the unit complex-valued field-polarization vector, and the e^q are its components in the cyclic basis $\{\mathbf{e}_q; q=0,\pm 1\}$. The quantum transport equation describing the evolution of the density matrix of atoms in the external field (1) in the Wigner representation without recoil effects taken into account has the general form

$$\left(\frac{\partial}{\partial t} + \mathbf{v}\nabla\right)\hat{\rho} + \hat{\Gamma}\{\hat{\rho}\} = -\frac{i}{\hbar}[\hat{H}_0, \hat{\rho}] - \frac{i}{\hbar}[\hat{V}_{E-D}, \hat{\rho}], \quad (2)$$

where \mathbf{v} is the velocity of an atom, \hat{H}_0 is the Hamiltonian of a free atom in the center-of-mass reference frame, and the operator $\hat{\Gamma}\{\hat{\rho}\}$ describes the radiative relation of atoms. In the basis of the Zeeman wave functions $\{|g\mu\rangle\}$ and $\{|e,\mu\rangle\}$ of, respectively, the ground and excited states ($\mu = -j, -j+1, \dots, j$), the density matrix $\hat{\rho}$ can be partitioned into four $(2j+1)$ -by- $(2j+1)$ matrix blocks $\hat{\rho}^{gg}$, $\hat{\rho}^{ee}$, $\hat{\rho}^{eg}$, and $\hat{\rho}^{ge}$:

$$\begin{aligned} \rho_{\mu\mu'}^{gg} &= \langle g, \mu | \hat{\rho} | g, \mu' \rangle, & \rho_{\mu\mu'}^{ee} &= \langle e, \mu | \hat{\rho} | e, \mu' \rangle, \\ \rho_{\mu\mu'}^{eg} &= \langle e, \mu | \hat{\rho} | g, \mu' \rangle, & \rho_{\mu\mu'}^{ge} &= \langle g, \mu | \hat{\rho} | e, \mu' \rangle. \end{aligned}$$

Here $\hat{\rho}^{gg}$ and $\hat{\rho}^{ee}$ have the meaning of the density matrices of the ground and excited states, respectively. The off-diagonal elements $\hat{\rho}^{eg}$ and $\hat{\rho}^{ge}$ describe the optical coherence between the ground and excited states. Separating the rapid and slow dependence on time and position in the components $\hat{\rho}^{eg}$ and $\hat{\rho}^{ge}$,

$$\begin{aligned} \hat{\rho}^{eg} &= \exp[-i(\omega t - \mathbf{k}\mathbf{r})] \hat{\rho}^{\hat{eg}}, \\ \hat{\rho}^{ge} &= \exp[i(\omega t - \mathbf{k}\mathbf{r})] \hat{\rho}^{\hat{ge}}, \end{aligned} \quad (3)$$

and using the rotating wave approximation, we obtain from Eq. (2) the following system of generalized Bloch equations for the slow components of the density matrix:

$$\left(\frac{\partial}{\partial t} + \frac{\gamma}{2} - i\delta\right)\hat{\rho}^{\hat{eg}} = -i\Omega[\hat{V}\hat{\rho}^{\hat{eg}} - \hat{\rho}^{\hat{ee}}\hat{V}], \quad (4)$$

$$\left(\frac{\partial}{\partial t} + \frac{\gamma}{2} + i\delta\right)\hat{\rho}^{\hat{ge}} = -i\Omega^*[\hat{V}^\dagger\hat{\rho}^{\hat{ge}} - \hat{\rho}^{\hat{gg}}\hat{V}^\dagger], \quad (5)$$

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

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

$$\text{Tr}\{\hat{\rho}^{\hat{gg}}\} + \text{Tr}\{\hat{\rho}^{\hat{ee}}\} = 1. \quad (8)$$

Here $\delta = (\omega - \omega_{eg} - \mathbf{k}\cdot\mathbf{v})$ is the displacement from resonance allowing for the Doppler shift, $\omega_{eg} = (E_e - E_g)/\hbar$ is the transition frequency, γ^{-1} is the radiative lifetime of the excited state, $\Omega = -E_0\langle e||d||g\rangle/\hbar$ is the effective Rabi frequency, and $\langle e||d||g\rangle$ is the reduced matrix element of the dipole moment. According to the Wigner-Eckart theorem,¹² the matrix elements of \hat{V} are expressed in terms of the $3jm$ -symbols as follows:

$$\begin{aligned} V_{\mu\mu'} &= \frac{1}{\langle e||d||g\rangle} \langle e, \mu | \hat{\mathbf{d}}\mathbf{e} | g, \mu' \rangle \\ &= \sum_{q=0,\pm 1} (-1)^{j-\mu} \begin{pmatrix} j & 1 & j \\ -\mu & q & \mu' \end{pmatrix} e^q. \end{aligned} \quad (9)$$

For closed optical transitions of the form $j_g = j \rightarrow j_e = j$ the operator for the atoms to reach the ground state owing to spontaneous emission, $\hat{\gamma}\{\hat{\rho}^{ee}\}$, has the standard form (see, e.g., Ref. 1)

$$\begin{aligned} \gamma_{\mu\mu'}\{\hat{\rho}^{ee}\} &= \gamma(2j+1) \sum_{q,\mu_1,\mu_2} (-1)^{j-\mu_1} \begin{pmatrix} j & 1 & j \\ -\mu_1 & q & \mu \end{pmatrix} \rho_{\mu_1\mu_2}^{ee} \\ &\quad \times (-1)^{j-\mu_2} \begin{pmatrix} j & 1 & j \\ -\mu_2 & q & \mu' \end{pmatrix}. \end{aligned} \quad (10)$$

The steady-state solution of the system of equations (4)–(8) can be found from the condition

$$\frac{\partial}{\partial t}\hat{\rho}^{gg} = \frac{\partial}{\partial t}\hat{\rho}^{ee} = \frac{\partial}{\partial t}\hat{\rho}^{eg} = \frac{\partial}{\partial t}\hat{\rho}^{ge} = 0.$$

Equations (4) and (5) make it possible to express the off-diagonal elements of $\hat{\rho}^{eg}$ and $\hat{\rho}^{ge}$ in terms of the density matrices $\hat{\rho}^{gg}$ and $\hat{\rho}^{ee}$:

$$\begin{aligned} \hat{\rho}^{\hat{eg}} &= -\frac{i\Omega}{\gamma/2 - i\delta} [\hat{V}\hat{\rho}^{gg} - \hat{\rho}^{ee}\hat{V}], \\ \hat{\rho}^{\hat{ge}} &= -\frac{i\Omega^*}{\gamma/2 + i\delta} [\hat{V}^\dagger\hat{\rho}^{ee} - \hat{\rho}^{gg}\hat{V}^\dagger]. \end{aligned} \quad (11)$$

Substituting these equations into (6) and (7), we arrive at a closed system of matrix equations for the steady-state $\hat{\rho}^{gg}$ and $\hat{\rho}^{ee}$:

$$\begin{aligned} \gamma\hat{\rho}^{ee} &= \gamma S\hat{V}\hat{\rho}^{gg}\hat{V}^\dagger - \frac{\gamma}{2} S\{\hat{V}\hat{V}^\dagger\hat{\rho}^{ee} + \hat{\rho}^{ee}\hat{V}\hat{V}^\dagger\} \\ &\quad + i\delta S\{\hat{V}\hat{V}^\dagger\hat{\rho}^{ee} - \hat{\rho}^{ee}\hat{V}\hat{V}^\dagger\}, \\ \hat{\gamma}\{\hat{\rho}^{ee}\} &= -\gamma S\hat{V}^\dagger\hat{\rho}^{ee}\hat{V} + \frac{\gamma}{2} S\{\hat{V}^\dagger\hat{V}\hat{\rho}^{gg} + \hat{\rho}^{gg}\hat{V}^\dagger\hat{V}\} \\ &\quad + i\delta S\{\hat{V}^\dagger\hat{V}\hat{\rho}^{gg} - \hat{\rho}^{gg}\hat{V}^\dagger\hat{V}\}, \end{aligned} \quad (12)$$

where

$$S = \frac{|\Omega|^2}{\gamma^2/4 + \delta^2} \quad (13)$$

is the saturation parameter.

Note that in reality atoms reach the steady state if the following conditions for the field-atom interaction time are met:

$$\gamma S t \gg 1; \quad \gamma t \gg 1.$$

3. THE EXACT STEADY-STATE SOLUTION

3.1 The solution in the general case

Before finding the solution of the system of equations (12), we note an important property of the incoming operator $\hat{\gamma}\{\hat{\rho}^{ee}\}$. If we put the unit matrix $\hat{I} = \|\delta_{\mu\mu'}\|$ instead of $\hat{\rho}^{ee}$ in (10), the well-known rules of summation for 3jm-symbols (see, e.g., Ref. 12) yield

$$\begin{aligned} \gamma_{\mu\mu'}\{\hat{I}\} &= \gamma(2j+1) \sum_{q,\mu_1} \begin{pmatrix} j & 1 & j \\ -\mu_1 & q & \mu \end{pmatrix} \begin{pmatrix} j & 1 & j \\ -\mu_1 & q & \mu' \end{pmatrix} \\ &= \gamma \delta_{\mu\mu'}. \end{aligned} \quad (14)$$

Thus we find

$$\hat{\gamma}\{\hat{I}\} = \gamma \hat{I}, \quad (15)$$

which is true for all closed transitions of the form $j_g = j \rightarrow j_e = j$. From the physical point of view this property follows from the fact that relaxation processes are isotropic. Note that for transitions of type $j_g = j \rightarrow j_e = j \pm 1$ this property (15) is modified in the following way:

$$\hat{\gamma}\{\hat{I}^e\} = \frac{2j_e + 1}{2j_g + 1} \gamma \hat{I}^e,$$

where \hat{I}^g and \hat{I}^e are, respectively, $(2j_g + 1)$ -by- $(2j_g + 1)$ and $(2j_e + 1)$ -by- $(2j_e + 1)$ unit matrices.

We can now write the exact solution of the system of equations (12):

$$\begin{aligned} \hat{\rho}^{ee} &= \beta \hat{I}, \quad \hat{\rho}^{gg} = \beta \left[\frac{1}{S} (\hat{V}^\dagger \hat{V})^{-1} + \hat{I} \right] \\ &= \beta \left[\frac{1}{S} (\hat{V})^{-1} (\hat{V}^\dagger)^{-1} + \hat{I} \right]. \end{aligned} \quad (16)$$

Direct substitution into (12) with allowance for (15) clearly shows that (16) satisfies the system (12) identically. The constant β can be found from the normalization condition (8):

$$\beta = \left[2(2j+1) + \frac{1}{S} \text{Tr}\{(\hat{V}^\dagger \hat{V})^{-1}\} \right]^{-1}. \quad (17)$$

Substitution of (16) into (11) yields the following expressions for $\hat{\rho}^{eg}$ and $\hat{\rho}^{ge}$:

$$\hat{\rho}^{eg} = (\hat{\rho}^{ge})^\dagger = -i\beta \left(\frac{\gamma}{2} + i\delta \right) (\Omega^* \hat{V}^\dagger)^{-1}. \quad (18)$$

The solution (16) implies that the density matrix of the excited state, $\hat{\rho}^{ee}$ is isotropic ($\hat{\rho}^{ee} \propto \hat{I}$) for all field intensities and polarizations, which is quite unexpected. Here the Zeeman sublevels of the ground state are populated uniformly and the coherence between them is generally finite. Note that for the transitions $j_g = j \rightarrow j_e = j \pm 1$ and $j_g = j \rightarrow j_e = j$ with j an integer, the matrix \hat{V} is singular, so that the inverses

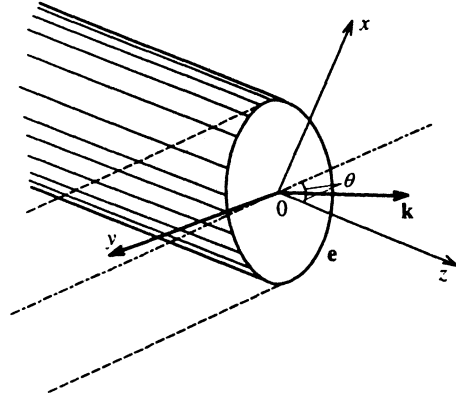


FIG. 1. The coordinate system suggested in Ref. 13 in which the z axis (the quantization axis) is directed along the axis of one of the cylinders built on the polarization ellipse e (the dashed lines stand for the second cylinder) and the y axis is directed along the minor semiaxis of the polarization ellipse. In this basis the vector e is a superposition of the linear component and one of the circular components [see Eq. (19)].

\hat{V}^{-1} and $(\hat{V}^\dagger)^{-1}$ do not exist. Consequently, the solution (16)–(18) exists only for the transition $j_g = j \rightarrow j_e = j$, where j is a half-integer.

As the solution (16)–(18) shows, the matrix elements of $\hat{\rho}$ can be expressed in terms of the matrix elements of the inverse matrices \hat{V}^{-1} , $(\hat{V}^\dagger)^{-1}$, and $(\hat{V}^\dagger \hat{V})^{-1} = \hat{V}^{-1} (\hat{V}^\dagger)^{-1}$. To calculate these we select the system of coordinates suggested in Ref. 13. As is well known, an arbitrary ellipse is the intersection of a cylinder and a plane, so that with each elliptical polarization vector e there can be associated a cylinder (generally there are two), and the given ellipse e is the intersection of this cylinder. We direct the quantization axis (the z axis) along the axis of this cylinder and the y axis along the minor semiaxis of the polarization ellipse (Fig. 1). The angle θ between the z axis and the wave vector k satisfies the relationship

$$\cos \theta = \pm \tan \varepsilon, \quad -\frac{\pi}{4} \leq \varepsilon \leq \frac{\pi}{4},$$

where ε is the ellipticity angle, whose value is defined so that $|\tan \varepsilon|$ is equal to the ratio of the minor semiaxis of the polarization ellipse to the major semiaxis. Then, as the results of Ref. 13 imply, the elliptical polarization e is the superposition of the linear component and one circular component:

$$e = \sqrt{\cos(2\varepsilon)} e_0 + \sqrt{2} \sin \varepsilon e_{\pm 1}. \quad (19)$$

For the sake of convenience we select

$$e = \sqrt{\cos(2\varepsilon)} e_0 + \sqrt{2} \sin \varepsilon e_{+1}. \quad (20)$$

The light-induced transitions corresponding to (20) are

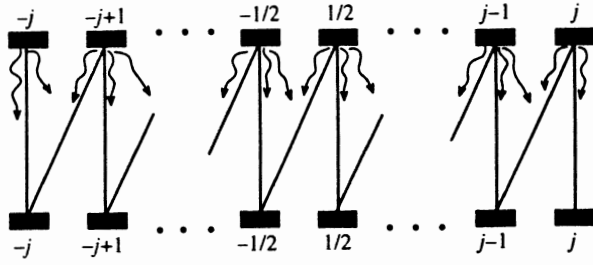


FIG. 2. The diagram representing light-induced (solid lines) and spontaneous (wavy lines) transitions in the system of coordinates depicted in Fig. 1 with the vector \mathbf{e} specified by (20).

depicted in Fig. 2. In this case the matrix \hat{V} is real and has lower triangular form with two nonzero diagonals:

$$\hat{V} = \begin{pmatrix} \cdot & & & & & & & & 0 \\ \cdot & & & & & & & & \\ \cdot & & & & & & & & \\ \cdot & & & & & & & & \\ \cdot & & & & & & & & \\ & & & & & V_{\mu(\mu-1)} & V_{\mu\mu} & & \\ & & & & & & \cdot & & \\ & & & & & & & \cdot & \\ & & & & & & & & \cdot & \\ & & & & & & & & & \cdot & \\ & & & & & & & & & & 0 \end{pmatrix}, \quad (21)$$

where, in accordance with (9) and (20),

$$V_{\mu\mu} = \frac{\sqrt{\cos(2\varepsilon)}}{\sqrt{j(j+1)(2j+1)}} \mu, \quad (22)$$

$$V_{\mu(\mu-1)} = -\frac{\sin \varepsilon}{\sqrt{j(j+1)(2j+1)}} \sqrt{(j+\mu)(j-\mu+1)}.$$

The inverse of the matrix \hat{V} also has lower triangular form and is real. Direct calculation of its matrix elements yields

$$[\hat{V}^{-1}]_{\mu\mu'} = \frac{(-1)^{\mu-\mu'}}{V_{\mu'\mu'}} \prod_{\alpha=\mu'+1}^{\mu} \frac{V_{\alpha(\alpha-1)}}{V_{\alpha\alpha}}$$

$$= \frac{\sqrt{j(j+1)(2j+1)}}{\sqrt{\cos(2\varepsilon)}} \left(\frac{\sin \varepsilon}{\sqrt{\cos(2\varepsilon)}} \right)^{\mu-\mu'} \frac{1}{\mu'}$$

$$\times \prod_{\alpha=\mu'+1}^{\mu} \frac{\sqrt{(j+\alpha)(j-\alpha+1)}}{\alpha}. \quad (23)$$

Here we have introduced the following notation:

$$\prod_{\alpha=\mu'+1}^{\mu} f_{\alpha} \equiv 1; \quad \prod_{\alpha=\mu'+1}^{\mu} f_{\alpha} \equiv 0, \quad \mu' > \mu.$$

Since in the chosen basis \hat{V} is real, $(\hat{V}^{\dagger})^{-1}$ is obtained from \hat{V}^{-1} by transposition, i.e., $[(\hat{V}^{\dagger})^{-1}]_{\mu\mu'} = [\hat{V}^{-1}]_{\mu'\mu}$. Now we can easily write the matrix elements of $(\hat{V}^{\dagger}\hat{V})^{-1}$:

$$[(\hat{V}^{\dagger}\hat{V})^{-1}]_{\mu\mu'}$$

$$= (-1)^{\mu-\mu'} \sum_{\nu=-j}^j \frac{1}{V_{\nu\nu}^2} \left(\prod_{\alpha=\nu+1}^{\mu} \frac{V_{\alpha(\alpha-1)}}{V_{\alpha\alpha}} \right)$$

$$\times \left(\prod_{\alpha'=\nu+1}^{\mu'} \frac{V_{\alpha'(\alpha'-1)}}{V_{\alpha'\alpha'}} \right)$$

$$= \frac{j(j+1)(2j+1)}{\cos(2\varepsilon)} \sum_{\nu=-j}^j \left(\frac{\sin \varepsilon}{\sqrt{\cos(2\varepsilon)}} \right)^{\mu+\mu'-2\nu} \frac{1}{\nu^2}$$

$$\times \left(\prod_{\alpha=\nu+1}^{\mu} \frac{\sqrt{(j+\alpha)(j-\alpha+1)}}{\alpha} \right)$$

$$\times \left(\prod_{\alpha'=\nu+1}^{\mu'} \frac{\sqrt{(j+\alpha')(j-\alpha'+1)}}{\alpha'} \right). \quad (24)$$

To calculate the constant β in (17) we must find the trace of $(\hat{V}^{\dagger}\hat{V})^{-1}$, which value, as (24) implies, is

$$\text{Tr}\{(\hat{V}^{\dagger}\hat{V})^{-1}\}$$

$$= \frac{j(j+1)(2j+1)}{\cos(2\varepsilon)} \sum_{\mu=-j}^j \sum_{\nu=-j}^{\mu} \left(\frac{\sin^2 \varepsilon}{\cos(2\varepsilon)} \right)^{\mu-\nu} \frac{1}{\nu^2}$$

$$\times \prod_{\alpha=\nu+1}^{\mu} \frac{(j+\alpha)(j-\alpha+1)}{\alpha^2} = \frac{j(j+1)(2j+1)}{\cos(2\varepsilon)}$$

$$\times \left[A_0 + A_1 \left(\frac{\sin^2 \varepsilon}{\cos(2\varepsilon)} \right) + \dots + A_{2j} \left(\frac{\sin^2 \varepsilon}{\cos(2\varepsilon)} \right)^{2j} \right], \quad (25)$$

$$A_0 = 2 \sum_{\mu=1/2}^j \frac{1}{\mu^2},$$

$$\dots$$

$$A_n = \sum_{\nu=-j}^{j-n} \frac{1}{\nu^2} \prod_{\alpha=\nu+1}^{\nu+n} \frac{(j+\alpha)(j-\alpha+1)}{\alpha^2},$$

$$\dots$$

$$A_{2j} = 4^{2j+1} \frac{[(2j-1)!!!]^2}{[(2j)!!!]^2}.$$

Here, as usual, $!!!$ stands for the product of all even (odd) numbers up to l inclusive, with $0!!! \equiv 1$. Note that $\text{Tr}\{(\hat{V}^{\dagger}\hat{V})^{-1}\}$ is an invariant, i.e., its value does not depend on the choice of the system of coordinates.

3.2. The particular cases of linear and circular polarizations

Let us now examine two particular cases most often encountered in nature: linear and circular polarization of the field.

1. *Linear polarization* ($\varepsilon=0$). Substituting $\varepsilon=0$ into (24) and (25) yields

$$\rho_{\mu\mu'}^{ee} = \frac{S \delta_{\mu\mu'}}{(2j+1)[2S+j(j+1)A_0]},$$

$$\rho_{\mu\mu'}^{gg} = \frac{\delta_{\mu\mu'}}{(2j+1)[2S+j(j+1)A_0]} \left[\frac{j(j+1)(2j+1)}{\mu^2} + S \right], \quad (26)$$

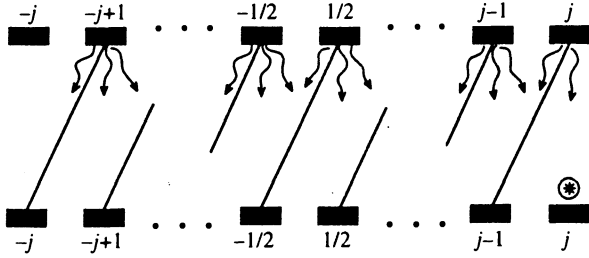


FIG. 3. Illustration of the steady-state solution for the case of circular polarization $\mathbf{e} = \mathbf{e}_{+1}$, when the atoms accumulate in states of the field marked by an * (such states do not interact with the field).

$$\begin{aligned} \rho_{\mu\mu'}^{eg} &= (\rho_{\mu'\mu}^{ge})^* \\ &= \frac{\Omega \delta_{\mu\mu'}}{(\delta + i\gamma/2)(2j+1)[2S + j(j+1)A_0]} \\ &\quad \times \frac{\sqrt{j(j+1)(2j+1)}}{\mu}. \end{aligned}$$

2. *Circular polarization* ($|\varepsilon| = \frac{1}{4}\pi$). In this case $\beta = 0$ and the inverse matrices \hat{V}^{-1} , $(\hat{V}^\dagger)^{-1}$, and $(\hat{V}^\dagger \hat{V})^{-1}$ do not exist. However, the atomic density matrix can be determined by passing to the limit $\varepsilon \rightarrow \pm \pi/4$:

$$\rho_{\mu\mu}^{ee} = 0, \quad \rho_{\mu\mu'}^{gg} = \delta_{\mu j} \delta_{\mu' j}, \quad \rho_{\mu\mu'}^{eg} = (\rho_{\mu'\mu}^{ge})^* = 0. \quad (27)$$

Thus, because of optical pumping by circularly polarized atoms of arbitrary intensity the atoms accumulate in the coherent population-trapping state $|g, j\rangle$ and cease to interact with the field (Fig. 3).

Note that the results of Sec. 3.2 coincide with those of Refs. 6–8 and are given here for the sake of completeness.

3.3. The conditions for weak and strong saturation

We now find the conditions for weak and strong saturation of the transition as functions of the field intensity and ellipticity.

1. When the saturation of the transition is weak, the ratio of the total population of the excited state to that of the ground state is much smaller than unity, i.e.,

$$\frac{\text{Tr}\{\hat{\rho}^{ee}\}}{\text{Tr}\{\hat{\rho}^{gg}\}} = \frac{2j+1}{S^{-1} \text{Tr}\{(\hat{V}^\dagger \hat{V})^{-1}\} + (2j+1)} \ll 1,$$

which is equivalent to the condition

$$\frac{S(2j+1)}{\text{Tr}\{(\hat{V}^\dagger \hat{V})^{-1}\}} \ll 1. \quad (28)$$

In this case we can ignore the unit matrix \hat{I} in the expression (16) for $\hat{\rho}^{gg}$ and write

$$\beta \approx \frac{S}{\text{Tr}\{(\hat{V}^\dagger \hat{V})^{-1}\}} \ll 1, \quad \hat{\rho}^{gg} \approx \frac{(\hat{V}^\dagger \hat{V})^{-1}}{\text{Tr}\{(\hat{V}^\dagger \hat{V})^{-1}\}}. \quad (29)$$

As Eq. (25) shows, in the limiting case where the polarization of the linear is close to linear ($|\varepsilon| = 0$ and $|\sin \varepsilon| \ll 1$) the condition (28) assumes the form

$$\frac{S}{j(j+1)A_0} \ll 1. \quad (30)$$

In the other limiting case where the polarization of the field is close to circular ($|\varepsilon| = \pi/4$ and $\cos(2\varepsilon) \ll 1$) we have

$$\frac{S[2 \cos(2\varepsilon)]^{2j+1}}{2j(j+1)A_{2j}} \ll 1. \quad (31)$$

2. Strong saturation corresponds to the total populations of the excited and ground states becoming practically equal, $\text{Tr}\{\hat{\rho}^{ee}\} / \text{Tr}\{\hat{\rho}^{gg}\} = 1$, and the condition becomes

$$\frac{S(2j+1)}{\text{Tr}\{(\hat{V}^\dagger \hat{V})^{-1}\}} \gg 1. \quad (32)$$

In this case the term $(\hat{V}^\dagger \hat{V})^{-1} S^{-1}$ in (16) can be ignored, i.e.,

$$\hat{\rho}^{gg} \approx \hat{\rho}^{ee} \approx \frac{1}{2(2j+1)} \hat{I}. \quad (33)$$

Thus, in the strong-saturation region specified by (32) the atomic density matrix becomes completely isotropic and the populations of the ground and excited states become practically the same. In the limiting case where the polarization of the field is close to linear ($|\varepsilon| = 0$ and $|\sin \varepsilon| \ll 1$), the condition (32) assumes the form

$$\frac{S}{j(j+1)A_0} \gg 1. \quad (34)$$

In the other limiting case where the polarization of the field is close to circular ($|\varepsilon| = \pi/4$ and $\cos(2\varepsilon) \ll 1$) we have

$$\frac{S[2 \cos(2\varepsilon)]^{2j+1}}{2j(j+1)A_{2j}} \gg 1. \quad (35)$$

We see that when the polarization of the field is close to circular, strong saturation sets in only for extremely high field intensities, since $\cos(2\varepsilon) \ll 1$. In the case of circular polarization ($\varepsilon = \pm \pi/4$ and $\cos(2\varepsilon) = 0$), where the matrix \hat{V} is singular, the condition (35) cannot be met no matter how high the intensity of the field.

4. CONCLUSION

We have found the exact steady-state solution of the problem of optical pumping of transitions $j_g = j \rightarrow j_e = j$ with j a half-integer, which, as any exact solution of a quantum mechanical problem, is of interest from the fundamental point of view. It can also be used in various applications related to the interaction of atoms with polarized radiation. Below we give examples of such applications:

1. Averaging the velocity distribution of the atomic ensemble, we can easily use (18) to build the nonlinear dielectric susceptibility tensor and study the propagation of an elliptically polarized plane wave in a gaseous medium.

2. The solution (16) can be employed in polarization spectroscopy in a situation where the pump field is strong and the probe field weak.

3. If we assume that the atomic velocity \mathbf{v} is zero and introduce a coordinate dependence in (1) for the amplitude $E_0(\mathbf{r})$ and the polarization vector $\mathbf{e}(\mathbf{r})$, the solution (16) also becomes spatially nonuniform, with both S and \hat{V} depending on \mathbf{r} . The solution can then be used to calculate the gradient force, the force of friction, and the diffusion coefficient in the quasiclassical description of the translational motion of slow atoms in a nonuniform monochromatic field. Note that Alekseev¹⁴ used a perturbation-theory technique to obtain an expression for the radiative force that allows for the nonuniformity in the initial distribution of atoms among the Zeeman sublevels.

There are several other applications of the steady-state solution (16).

Combining the results of the present work and those of Ref. 5, we can claim to have found the exact steady-state solutions for closed $j_g = j \rightarrow j_e = j - 1$ and $j_g = j \rightarrow j_e = j$ optical transitions for arbitrary values of j . Hence in general only the exact solution of transitions of type $j_g = j \rightarrow j_e = j + 1$ remains unknown.

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