Classical description of angular-momentum motion due to optical pumping

K. A. Nasyrov
Institute of Automation and Electrometry, Novosibirsk, 630090, Russia
(Received 12 April 2000; published 7 March 2001)

The Wigner representation of angular-momentum orientation in the classical limit on $J \gg 1$ has been used to derive an equation of angular-momentum motion in the case of a closed two-level system interacting with an arbitrary polarized radiation. This equation enables us to carry out a theoretical analysis of angular-momentum motion and consider final states of the quantum system for all types of dipole transitions. We have found that in the case of a zero magnetic field in the final state of the system with $J \rightarrow J$ or $J \rightarrow J+1$ optical transition, the angular momentum is directed along or opposite to the wave vector of the radiation depending on the sign of the polarization ellipsity, while for a linear polarization the angular momentum is isotropically distributed on the plane orthogonal to the polarization vector. For the $J \rightarrow J-1$ transition two directions of angular momentum are possible in the final state. These directions are determined by the ellipsity of polarization. The spatial size of the final distribution of the angular momentum has been defined by the quantum uncertainty, $1/\sqrt{J}$, of the angular-momentum orientation. In the case of the $J \rightarrow J-1$ or $J \rightarrow J$, transition particles come into ''dark'' states while for the $J \rightarrow J+1$ transition they occupy the ''brightest'' state. In the presence of a nonzero magnetic field, particles with $J \rightarrow J$ transition have in the final state only one angular-momentum orientation, i.e., along or opposite to the direction of the magnetic field depending on the polarization ellipsity sign. For the $J \rightarrow J+1$ transition both directions are possible, i.e., along and opposite to the magnetic field. In the case of the $J \rightarrow J-1$ transition the directions of the angular momentum form a cone around the magnetic field.

DOI: 10.1103/PhysRevA.63.043406 PACS number(s): 32.80.$-$t, 03.65.Sq, 33.80.Be

I. INTRODUCTION

Spatial orientation of the atomic angular momentum or optical pumping between ground magnetic sublevels is a well-known phenomenon [1,2]. The angular-momentum motion is a result of the repeated cycles of the resonant absorption of the polarized light followed by the spontaneous emission. Finally, after a long time of interaction with light, atoms come into their final stationary states. If the relaxation is only caused by spontaneous emission, then for some types of optical transitions ($J \rightarrow J-1, J \rightarrow J$) the final states coincide with so-called atomic stationary coherent states [3]. In these states the atom does not interact with polarized radiation (''dark'' states). For an elliptical polarization of light and an arbitrary value of J, exact solutions for the stationary coherent states have been found in Refs. [3,4]. However, this theory only analyses final atomic states and cannot describe angular-momentum motion. The analytical description of the optical pumping process of magnetic sublevels is only possible for a small J, such as $J=0,1/2,1$ [5].

In recent work [6] the process of the interaction of an open two-level quantum system having a large value of angular momentum ($J \approx 10$) with elliptically polarized light has been investigated by numerical methods. The open system means that a spontaneously emitted photon does not bring the system back to its original ground state but populates other levels that are not affected by radiation. This case is typical for molecules. The result is that this interaction produces an anisotropic distribution of angular-momentum orientation in the ground state. However this anisotropy is an effect of the population depletion of the open system caused by an anisotropy of the interaction potential, rather than an actual effect of angular-momentum motion. For the closed two-level system the population depletion of the ground state does not exist, so that in this case we have got a pure effect of angular-momentum motion.

In the present paper we consider a process of the orientation of the closed two-level system with a large value of J, which interacts with elliptically polarized light. The condition $J \gg 1$ enables us to use the classical description of the angular-momentum motion based on the Wigner representation of rotational motion [7,8]. This approach significantly simplifies the solution of the problem in comparison with a quantum-mechanical description.

The closed two-level model is valid rather more for atoms than for molecules. For example for alkali atoms, part of the optical transitions of the D_2 line are closed. Unfortunately, almost all alkali atoms in the ground state have a small value of total atomic momentum $\mathbf{F}=\mathbf{I}+\mathbf{J}$, here I is nuclear momentum and J is total electronic momentum. Nevertheless the approximation $F \gg 1$ can be applied for a few atoms like Fr_{208} and Fr_{210}, having sufficiently high value of nuclear momenta: $I=7$ and $I=6$, respectively. Note that francium isotopes are the best candidates for parity nonconservation experiments [9]. Also we assume that there are a few atoms interacting with light and radiation trapping is negligible. In particular, this case is typical for a francium magneto-optic trap [10].

Here it is worth noticing that the ''classical'' approach in Ref. [6] does not correspond to a correct classical description of angular-momentum motion because the equations used in Ref. [6] for the density matrix in the coherent-state representation [7,11] are only valid in the limit of $J \rightarrow \infty$. However it is easy to show that in this limit there is no angular-momentum motion caused by a resonant interaction with light. Indeed, for each cycle of the resonant photon absorption followed by the spontaneous emission, the orientation of the angular momentum is changed by the angle $\Delta\theta$

$\sim\Delta M/J\sim 1/J$ (where $|\Delta M|\leqslant 1$ for the dipole optical transition). In the limit of $J\to\infty$ we have $\Delta\theta=0$, which means there is no angular-momentum motion. Therefore, in the classical limit, for a correct description of angular-momentum motion we have to assume a large but finite value of J. Formally it means that in the equation for the density matrix we have to take into account terms at least of order $1/J$. These additional terms have a differential form and the procedure to obtain them is presented in Refs. [7,8].

Here after we show that angular-momentum motion depends strongly on the polarization of light and the type of the optical transition. Due to the motion the system comes into dark states ($J\to J-1, J\to J$) or into the brightest state ($J\to J+1$) for an arbitrary polarization of light. Final orientations of the angular momentum have been found for all types of optical transitions and results have been generalized for the case of a nonzero magnetic field.

II. EQUATIONS FOR ANGULAR-MOMENTUM MOTION

Consider a model system of the two-level system with angular momenta J_n and J_m for the ground n and excited m states, respectively, which degenerated on the projections of angular momenta M_n, M_m. The resonant electromagnetic field induces the transition between the levels and populates the level m. Let us assume that the system has one only relaxation channel, i.e., the excited level m spontaneously decays back to the ground level n.

The dynamic of the quantum system is described by the equation for the density matrix $\hat{\rho}$, which in the JM representation has the form [12]

$$\left(\frac{d}{dt}+\Gamma_m\right)\rho(J_mM_m|J_mM_m')=-\frac{i}{\hbar}\sum_{M_n}[V(J_mM_m|J_nM_n)\rho(J_nM_n|J_mM_m')-\rho(J_mM_m|J_nM_n)V(J_nM_n|J_mM_m')],$$

$$\frac{d}{dt}\rho(J_nM_n|J_nM_n')=-\frac{i}{\hbar}\sum_{M_m}[V(J_nM_n|J_mM_m)\rho(J_mM_m|J_nM_n')-\rho(J_nM_n|J_mM_m)V(J_mM_m|J_nM_n')]$$
$$+\Gamma_m\sum_{M_m,M_m',\sigma}C^{J_m,M_m}_{J_n,M_n;1,\sigma}C^{J_m,M_m'}_{J_n,M_n';1,\sigma}\rho(J_mM_m|J_mM_m'), \quad (1)$$
$$\left(\frac{d}{dt}+\frac{1}{2}\Gamma_m+i\omega_{mn}\right)\rho(J_mM_m|J_nM_n)=-\frac{i}{\hbar}\left[\sum_{M_n'}V(J_mM_m|J_nM_n')\rho(J_nM_n'|J_nM_n)-\sum_{M_m'}\rho(J_mM_m|J_mM_m')V(J_mM_m'|J_nM_n)\right].$$

Here ω_{mn} is the frequency of the optical transition $n-m$ and Γ_m is the rate of the spontaneous decay. $V(J_mM_m|J_nM_n)$ is the matrix element of the electrodipole interaction $V=-\mathbf{d}\boldsymbol{\mathcal{E}}(t)$, $\mathbf{d}$ is the dipole moment, and $\boldsymbol{\mathcal{E}}(t)$ is the monochromatic electric field

$$\boldsymbol{\mathcal{E}}(t)=\mathbf{E}e^{-i\omega t}+\mathbf{E}^*e^{i\omega t}, \quad (2)$$

where $\mathbf{E}$ is a complex vector of the amplitude of the electrical field. The last term in the right-hand side of Eq. (1) for density-matrix elements of the ground state, describes the contribution into the ground state, related to the spontaneous decay of the m level. $C^{J_m,M_m}_{J_n,M_n;1,\sigma}$ is the Clebsh-Gordan coefficient [13].

Further, instead of the JM representation we introduce the Wigner representation of rotational motion. For this specific problem it is more convenient to use the Wigner representation of the angular-momentum orientation [7] or $\phi\theta\alpha$ representation [8]. The transition from JM to $\phi\theta\alpha$ representation is given by the transformation [8]

$$\rho_{J_1J_2}(\phi,\theta,\alpha)=\sum_{\kappa,q,M_1,M_2}\frac{\sqrt{2\kappa+1}}{\sqrt{J_1+J_2+1}}(-1)^{J_2-M_2}\times C^{\kappa,q}_{J_1,M_1;J_2,-M_2}D^{\kappa*}_{q,J_1-J_2}(\phi,\theta,\alpha)\times\rho(J_1M_1|J_2M_2), \quad (3)$$

where $D^{\kappa*}_{q,J_1-J_2}(\phi,\theta,\alpha)$ is the Wigner D matrix [13], $\rho_{J_1J_2}(\phi,\theta,\alpha)$ is the Wigner function in the $\phi\theta\alpha$ representation, ϕ, θ are azimuthal and polar angles of the angular-momentum orientation, and the angle α defines the position of the rotator axis in the plane orthogonal to the angular momentum. Note, if $J_1=J_2=J$, according to Eq. (3), the Wigner function is independent of the angle α so that $\rho_{JJ}(\phi,\theta)$ means a spatial distribution of angular momentum.

Equations (1) in the $\phi\theta\alpha$ representation take the form [8]

$$\left(\frac{d}{dt}+\Gamma_m\right)\rho_{mm}=-ie^{i\hat{w}/2J}G\rho_{nm}+ie^{-\hat{w}/2J}G^*\rho_{mn},$$

$$\frac{d}{dt}\rho_{nn}=ie^{-i\hat{w}/2J}G\rho_{nm}-ie^{i\hat{w}/2J}G^*\rho_{mn}+\hat{\Gamma}_m\rho_{mm},$$

$$\left(\frac{d}{dt}+\frac{\Gamma_m}{2}-i(\omega-\omega_{nm})\right)\rho_{mn}=-ie^{i\hat{w}/2J}G\rho_{nn}+ie^{-i\hat{w}/2J}G\rho_{mm},$$

$$\rho_{nm}=\rho_{mn}^*,\quad 2J=J_m+J_n+1. \tag{4}$$

Here we use notations

$$\rho_{kl}\equiv\rho_{J_kJ_l}(\phi,\theta,\alpha),$$

$$G(\phi,\theta,\alpha)=\frac{V_{J_mJ_n}(\phi,\theta,\alpha)}{\hbar}e^{i\omega t}$$

$$=\sum_\sigma G_\sigma D^1_{\sigma,J_n-J_m}(\phi,\theta,\alpha)=\tilde{G}(\phi,\theta)e^{i(J_m-J_n)\alpha},$$

$$\tilde{G}(\phi,\theta)=\sum_\sigma G_\sigma D^1_{\sigma,J_n-J_m}(\phi,\theta,0),$$

$$G_\sigma=(-1)^{J_n-J_m-1}\frac{d_{mn}}{\hbar\sqrt{2J+1}}E_\sigma, \tag{5}$$

where d_{mn} is the reduced matrix element of the dipole moment, and $E_\sigma(\sigma=-1,0,1)$ are circular components of the electric field vector **E**. The term $\hat{\Gamma}_m\rho_{mm}$ describes the income to the ground state due to the spontaneous decay of the excited atoms in the $\phi\theta\alpha$ representation. The explicit form of the operator $\hat{\Gamma}_m$ is given in Appendix A.

The action of the operator $\hat{w}$ is defined by the rule [8]

$$\hat{w}PQ=\left(\frac{\partial}{\partial\phi}-\cos\theta\frac{\partial}{\partial\alpha}\right)P\frac{\partial}{\partial\cos\theta}Q-\frac{\partial}{\partial\cos\theta}P\left(\frac{\partial}{\partial\phi}-\cos\theta\frac{\partial}{\partial\alpha}\right)Q. \tag{6}$$

For simplicity, consider the case of the exact resonance $\omega=\omega_{mn}$ and the limit of the weak intensity $G\ll\Gamma$. This limit means that the population of the excited level is always much less than one for the ground level so that we can neglect ρ_{mm} everywhere in the comparison with ρ_{nn} and omit the time derivatives in the equations for ρ_{mm},ρ_{mn}. Summing up the equations on ρ_{mm} and ρ_{nn} in this approximation we come to the equation

$$\frac{\partial}{\partial t}\rho_{nn}=2\sin\left(\frac{\hat{w}}{2J}\right)(G\rho_{nm}+G^*\rho_{mn})+(\hat{\Gamma}_m-\Gamma_m)\rho_{mm}. \tag{7}$$

This equation describes the dynamics of angular momentum. In the limit $J\to\infty$ we get $\hat{\Gamma}_m=\Gamma_m$ (see Appendix A) and after that Eq. (7) yields

$$\frac{\partial}{\partial t}\rho_{nn}=0.$$

It means that there is no motion of angular momentum in the limit $J\to\infty$ as it was mentioned above. Only the terms of order $1/J$ in Eq. (7) give the correct classical limit of the angular-momentum motion. We leave in Eq. (7) terms with accuracy up to $1/J^2$, because they play the main role in the determining of the space width of the final distribution of angular momentum. Therefore, the operator $\hat{\Gamma}_m-\Gamma_m$ can be written in the form

$$\frac{\hat{\Gamma}_m-\Gamma_m}{\Gamma_m}=\frac{J_m-J_n}{J}\left(1+\frac{J_m-J_n}{2J}\right)+\frac{1}{sJ^2}\Delta, \tag{8}$$

where Δ is angular Laplacian, $s=2$ for $J_m=J_n$, and $s=4$ for $J_m=J_n\pm1$ (Appendix A), and it suffices to find ρ_{mn},ρ_{mm} from Eq. (4) with accuracy $1/J$

$$\rho_{mm}=-\frac{i}{\Gamma_m}(G\rho_{nm}-G^*\rho_{mn})+\frac{\hat{w}}{2J\Gamma_m}(G\rho_{nm}+G^*\rho_{mn}),$$

$$\rho_{mn}=-\frac{2i}{\Gamma_m}G\rho_{nn}+\frac{\hat{w}}{J\Gamma_m}G\rho_{nn}, \tag{9}$$

$$\rho_{nm}=\frac{2i}{\Gamma_m}G^*\rho_{nn}+\frac{\hat{w}}{J\Gamma_m}G^*\rho_{nn}.$$

First of all we derive an equation for angular-momentum motion in the classical limit holding in Eq. (7) the terms of order $1/J$ only. From Eqs. (7), (8), (9) we obtain

$$\frac{d}{dt}\rho_{nn}+\frac{\partial}{\partial\cos\theta}u_{\cos\theta}\rho_{nn}+\frac{\partial}{\partial\phi}u_\phi\rho_{nn}=0,$$

$$u_{\cos\theta}=\frac{2}{J\Gamma_m}\left[i\left(\frac{\partial\tilde{G}}{\partial\phi}\tilde{G}^*-\tilde{G}\frac{\partial\tilde{G}^*}{\partial\phi}\right)-2(J_m-J_n)|G|^2\cos\theta\right],$$

$$u_\phi=\frac{2i}{J\Gamma_m}\left[\frac{\partial\tilde{G}}{\partial\cos\theta}\tilde{G}^*-\tilde{G}\frac{\partial\tilde{G}^*}{\partial\cos\theta}\right]. \tag{10}$$

Equation (10) looks like an ordinary equation of continuity, which means the conservation law of the particle number. According to this equation the redistribution of particles upon their angular-momentum orientation (ϕ, θ) occurs due to fluxes $\rho_{nn}u_{\cos\theta}$ and $\rho_{nn}u_\phi$ along coordinates $\cos\theta$ and ϕ, respectively. In the approximation used, the population of the excited level is related to one of ground level by the equation

$$\rho_{mm}=\frac{4|G|^2}{\Gamma_m^2}\rho_{nn}. \tag{11}$$

From Eq. (10) it follows that the characteristic scale of the velocity of angular-momentum motion is $u\sim|G|^2/J\Gamma_m$. This is quite clear because every spontaneously emitted photon can change the angular-momentum projection by $|\Delta M|\leqslant1$ and, subsequently, the angle of momentum orientation by $\Delta\theta \sim \Delta M/J \sim 1/J$. The number of photons spontaneously emitted per unit of time is $\Gamma_m \rho_{mm}$. Thus, Eq. (11) enables us to estimate $u \sim \Delta\theta \Gamma_m \rho_{mm} \sim |G|^2/J\Gamma_m$.

In the system of coordinates, where the z axis is directed along the wave vector and the x axis coincides with the major axis of the ellipse of the wave polarization, the values of velocities $u_{\cos\theta}$ and u_ϕ for all possible dipole transitions are

$$J_m = J_n + 1:$$

$$u_{\cos gv} = \frac{\sin^2\theta}{J\Gamma_m}[|G_{-1}|^2(1+\cos\theta) - |G_{+1}|^2(1-\cos\theta) + 2|G_{+1}G_{-1}|\cos\theta\cos 2\phi], \tag{12}$$

$$u_\phi = \frac{2|G_{+1}G_{-1}|}{J\Gamma_m}\sin 2\phi,$$

$$J_m = J_n:$$

$$u_{\cos\theta} = \frac{2\sin^2\theta}{J\Gamma_m}[|G_{-1}|^2 - |G_{+1}|^2], \tag{13}$$

$$u_\phi = 0,$$

$$J_m = J_n - 1:$$

$$u_{\cos\theta} = \frac{\sin^2\theta}{J\Gamma_m}[|G_{-1}|^2(1-\cos\theta) - |G_{+1}|^2(1+\cos\theta) - 2|G_{+1}G_{-1}|\cos\theta\cos 2\phi], \tag{14}$$

$$u_\phi = -\frac{2|G_{+1}G_{-1}|}{J\Gamma_m}\sin 2\phi.$$

Note that in this particular coordinate system $G_0 = 0$.

III. ANGULAR-MOMENTUM MOTION AND THE FINAL STATES

We start our analysis with the consideration of the optical transition $J_m = J_n$. According to Eq. (13) there is no flux of angular momentum along ϕ. A motion exists only along $\cos\theta$. We can see that $u_{\cos\theta} > 0$ for any θ if $|G_{-1}| > |G_{+1}|$. It means that for any initial orientation of angular momentum it moves towards the direction of the wave vector ($\cos\theta \to 1$). At the final point of the motion all particles have angular momenta oriented along the wave vector ($\theta = 0$). The interaction strength is defined by the value $|G|^2$, which is

$$|G|^2 = \frac{\sin^2\theta}{2}(|G_{-1}|^2 + |G_{+1}|^2 + 2|G_{-1}G_{+1}|\cos 2\phi), \tag{15}$$

for the $J_m = J_n$ transition. For the final state ($\theta = 0$) we obtain $|G|^2 = 0$. The result means that the particles come to the dark state where they do not interact with light. Obviously, for the light polarization $|G_{-1}| < |G_{+1}|$, angular momenta of the particles are oriented in the opposite direction of the wave vector.

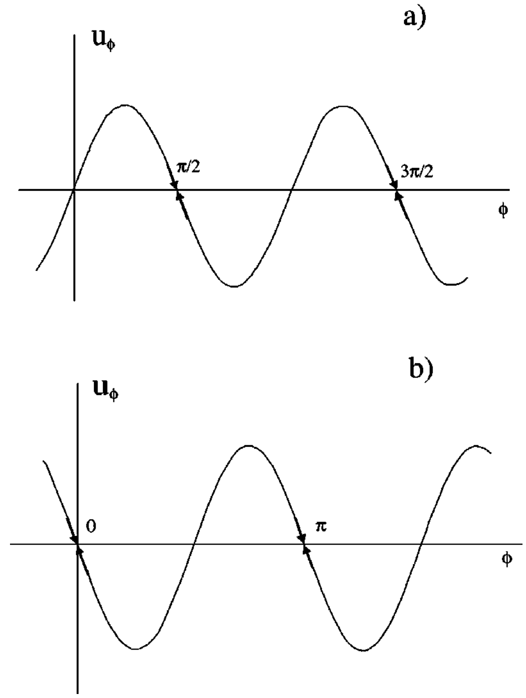

FIG. 1. The velocity u_ϕ as a function of angle ϕ for (a) $J_m = J_n + 1$ and (b) $J_m = J_n - 1$ transitions. Arrows show the points of convergence.

Due to our approximation, the Eq. (10) gives a δ-like distribution of particle angular momentum at the final state ($\theta = 0$). To find the real spatial distribution of angular momentum we have to take into account in Eqs. (7), (8), and (9) the next terms of order $1/J^2$. Moreover, it is easy to see that for the linear polarization ($|G_{-1}| = |G_{+1}|$) the velocity $u_{\cos\theta}$ [Eq. (13)] vanishes so that for this particular light polarization the equation with terms of $1/J^2$ should be used in order to describe correctly angular-momentum motion. We show later on that for this polarization, angular momenta in the final state are always oriented along the plane orthogonal to the wave vector. It is important to notice that at $J \gg 1$ the small deviation of the light polarization from a linear one that could be measured by value $\| G_{-1}| - |G_{+1}\| / (|G_{-1}| + |G_{+1}|) \geq 1/J$, forces angular momentum to take an orientation along or opposite to the z axis.

Now consider angular-momentum motion for the case of the optical transition $J_m = J_n + 1$, Eq. (12). In contrast with the transition $J_m = J_n$, in this case the velocity u_ϕ is nonzero and $u_{\cos\theta}$ is a variable quantity of angle ϕ. This makes the picture of angular-momentum motion more complicated. However the remarkable fact that u_ϕ does not depend on angle θ enables us to consider the motion only along angle ϕ. The velocity u_ϕ as a function of angle ϕ is shown in Fig. 1(a). It is clear that all particles move with the angle ϕ towards the $\pi/2$ or $3\pi/2$ angle for any θ. The velocity $u_{\cos\theta}$ for the points of the convergence $\phi^* = \pi/2, 3\pi/2$ is so

$$u_{\cos\theta} = \frac{\sin^2\theta}{2J\Gamma}(|G_{-1}| - |G_{+1}|)[|G_{-1}| + |G_{+1}| + (|G_{-1}| - |G_{+1}|)\cos\theta]. \tag{16}$$

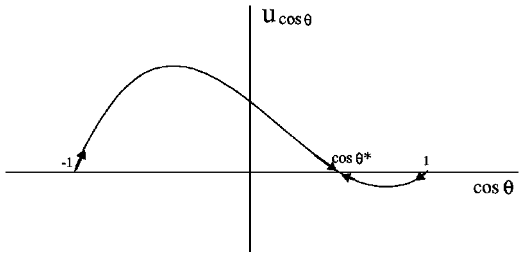

FIG. 2. The velocity $u_{\cos\theta}$ [Eq. (18)] as a function of cos θ for the $J_m = J_n - 1$ transition.

Thus we obtain that $u_{\cos\theta} > 0$ for the light polarization $|G_{-1}| > |G_{+1}|$ but $u_{\cos\theta} < 0$ if $|G_{-1}| < |G_{+1}|$, for the arbitrary angle θ. Again angular momentum moves towards the z-axis direction (for certainty we assume $|G_{-1}| > |G_{+1}|$), and in the final state all angular momenta are aligned along the z axis (i.e., along the wave vector) $\theta = 0$. For the $J_m = J_n + 1$ transition the interaction strength is

$$|G|^2 = |G_{-1}|^2 \frac{(1+\cos\theta)^2}{4} + |G_{+1}|^2 \frac{(1-\cos\theta)^2}{4} + \frac{\sin^2\theta}{2} |G_{-1} G_{+1}| \cos 2\phi. \tag{17}$$

It reaches its maximal value $|G|^2_{\max} = |G_{-1}|^2$ for the final state and it means that this state is the brightest state.

For the linear polarization of light the velocity $u_{\cos\theta} = 0$ [Eq. (16)]. This case requires separate consideration that is given below.

So, it is easy to see that the final orientation of the angular momentum is identical for both $J_m = J_n$ and $J_m = J_n + 1$ transitions. The differences are in the path which the angular momentum uses to come into the final state. For the $J_m = J_n$ transition the angular momentum goes directly to the z-axis direction whereas for the $J_m = J_n + 1$ transition, the angular-momentum orientation moves on a more complicated path. At first it comes to the convergent points of angle ϕ and then moves towards the z axis. Another difference is that for the $J_m = J_n$ transition the final state is a dark one but it is the brightest state for the $J_m = J_n + 1$ transition.

At last, consider angular-momentum motion of particles with the $J_m = J_n - 1$ optical transition. According to Eq. (14) the motion over angle ϕ is independent of the angle θ and particles are gathered near convergent points $\phi^* = 0, \pi$ [Fig. 1(b)]. For these angle ϕ^* the velocity $u_{\cos\theta}$ is

$$u_{\cos\theta} = \frac{\sin^2\theta}{2J\Gamma} (|G_{-1}| + |G_{+1}|)[|G_{-1}| - |G_{+1}| - (|G_{-1}| + |G_{+1}|)\cos\theta]. \tag{18}$$

This velocity as a function of cos θ is shown in Fig. 2. From this picture it is clear that particles must be gathered near an angle θ^*, which is defined by the equation

$$\cos\theta^* = \frac{|G_{-1}| - |G_{+1}|}{|G_{-1}| + |G_{+1}|}. \tag{19}$$

Thus, for a quantum system with the $J_m = J_n - 1$ transition, there are two possible final directions of angular momentum, i.e., $\theta^*, \phi^* = 0, \pi$. This conclusion is in agreement with the exact solution for coherent stationary states [14] for the given type of the optical transition. In the particular case of linear polarization $|G_{-1}| = |G_{+1}|$, we have $\theta^* = \pi/2$, and it means that the final angular momentum is directed along or opposite to the polarization vector. Note that if the initial distribution of angular momentum is isotropic then particles are gathered near these directions of the angular momentum with the same number [Fig. 3(b)]. What final direction of the angular momentum will be occupied by a particle depends only on an initial azimuthal angle ϕ. For example, if the initial value of angle ϕ is within the region $\pi/2 < \phi < 3\pi/2$ then the final direction of the angular momentum must be θ^*, $\phi^* = \pi$. It is easy to find by direct calculation that $|G(\theta^*, \phi^*)|^2 = 0$ for the $J_m = J_n - 1$ transition and, consequently, the final state is a dark one.

As mentioned above Eq. (10) gives a δ-like distribution of angular momentum for final states, because this equation was derived within accuracy of $1/J$. To describe the final distributions in more detail we have to take into account the terms of order $1/J^2$. This equation within acceptable accuracy can be obtained on the base of Eqs. (7), (8), and (9). Here, for simplicity, we consider the final distributions of angular momentum only in two cases of a circularly and linearly polarized radiation.

For instance consider the circular polarization $|G_{+1}| = 0$, $|G_{-1}| \neq 0$ and the quantum system with the optical transition $J_m = J_n$. In this case the final distribution of angular momentum satisfies the stationary equation

$$-2J|G|^2 \rho_{nn} + |G|^2 \frac{\partial}{\partial\cos\theta} \rho_{nn} + \sin^2\theta \frac{\partial}{\partial\cos\theta} |G|^2 \rho_{nn} = 0. \tag{20}$$

Since $|G|^2 \propto \sin^2\theta$ for the given light polarization and the transition type, it is easy to obtain the solution of Eq. (20)

$$\rho_{nn} = \frac{J}{\pi} \left[\frac{\sqrt{2} + \cos\theta}{\sqrt{2} + 1} \right]^{J/\sqrt{2} - 1} \left[\frac{\sqrt{2} - 1}{\sqrt{2} - \cos\theta} \right]^{J/\sqrt{2} + 1} \approx \frac{J}{\pi} e^{-J\theta^2}, \tag{21}$$

for normalization condition

$$\int \rho_{nn} \sin\theta d\theta d\phi = 1. \tag{22}$$

Thus, we see that in the final state the angular momentum should be distributed within the angle width $\Delta\theta = 1/\sqrt{J}$ near the direction $\theta = 0$ [Fig. 3(a)]. Note that the value $1/\sqrt{J}$ characterizes a scale of the quantum uncertainty of the angular-momentum orientation [7]. We can find that the same distribution of angular momentum is valid for the circular polarization of light and $J_m = J_n \pm 1$ optical transitions.

Now consider the case of a linear polarization of light. We choose a coordinate system with the z axis directed along the polarization vector and, therefore, there is one only non-

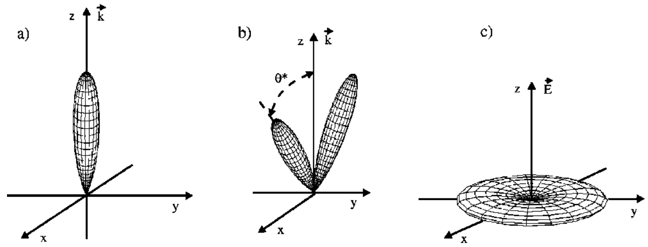

FIG. 3. The final distributions of angular momentum for (a) $J_m=J_n+1$, $J_m=J_n$ and (b) $J_m=J_n-1$ transitions for the elliptical polarization of light, and (c) for the linear polarization in the case of $J_m=J_n+1$, $J_m=J_n$ transitions.

zero circular component E_0 of the electric-field vector. For the $J_m=J_n$ transition the final distribution of angular momentum satisfies the equation

$$\Delta|G|^2\rho_{nn}=0.$$

Obviously, the solution of this equation is $|G|^2\rho_{nn}=\text{const}$. Because of $\rho_{mm}\propto|G|^2\rho_{nn}$ [see Eq. (11)] this fact means that in the final state the excited particles have an isotropic distribution of angular momentum. This conclusion is in agreement with Ref. [4]. Since for the $J_m=J_n$ transition and the linear polarization $|G|^2\propto\cos^2\theta$, the particles in the ground state have the distribution

$$\rho_{nn}\propto\cos^{-2}\theta, \tag{23}$$

and ρ_{nn} is formally divergent at $\theta=\pi/2$. This divergence indicates that for the correct description of angular momentum in this particular case we have to treat the equation for ρ_{nn} (7) with more accuracy than $1/J^2$. Nevertheless it is clear from Eq. (23) that in the final state the angular momentum is distributed isotropically on angle ϕ in the plane orthogonal to the polarization vector [Fig. 3(c)].

For a quantum system with the $J_m=J_n+1$ transition and for the linear light polarization we can derive the equation for the final distribution ρ_{nn}

$$-2J|G|^2\cos\theta\rho_{nn}+|G|^2\cos^2\theta\frac{\partial}{\partial\cos\theta}\rho_{nn} + \sin^2\theta\frac{\partial}{\partial\cos\theta}|G|^2\rho_{nn}=0, \tag{24}$$

and the solution of this equation is ($|G|^2\propto\sin^2\theta$)

$$\rho_{nn}\sim(1+\cos^2\theta)^{-2J}\approx e^{-2J(\theta-\pi/2)^2}. \tag{25}$$

Thus in the case of the $J_m=J_n+1$ transition the angular momentum is distributed isotropically within the plane orthogonal to the polarization vector. The spatial width of this distribution again is defined by the quantum uncertainty $1/\sqrt{J}$ of the momentum direction.

The case of the $J_m=J_n-1$ transition can be easily analyzed by the replacement $J\to-J$ in Eq. (24) so that instead of Eq. (25) the final angular-momentum distribution has the form

$$\rho_{nn}\sim(1+\cos^2\theta)^{2J}. \tag{26}$$

The equation shows the distribution with two possible maximums: at $\theta=0$ and $\theta=\pi$. For instance, for the angle θ close to $\theta=0$, this equation can be approximated as

$$\rho_{nn}\sim e^{-J\theta^2}, \tag{27}$$

that describes the particle angular momentum directed along the polarization vector and distributed within the quantum uncertainty angle $1/\sqrt{J}$.

IV. ANGULAR-MOMENTUM MOTION IN THE CASE OF A NONZERO MAGNETIC FIELD

Here we consider angular momentum motion for a nonzero magnetic field **H**. In the system of coordinates with the z axis directed along magnetic field, Eq. (10) can be easily generalized by the substitution

$$\frac{\partial}{\partial t}\to\frac{\partial}{\partial t}+\omega_H\frac{\partial}{\partial\phi}, \tag{28}$$

where $\omega_H=\mu_B gH$ is the Larmor frequency of momentum precession, μ_B is the Bhor magniton, and g is the Lande factor for the ground state. We assume that the direction of wave vector and the light polarization are arbitrary. We consider the most interesting case when Larmor frequency is much less than the spontaneous decay rate $\omega_H\ll\Gamma_m$, but it is much greater than the velocity of angular-momentum motion due to the interaction with light, $\omega_H\gg|G|^2/J\Gamma_m$. In this case it is clear that Wigner function $\overline{\rho_{nn}}$, averaged over the time period larger than $1/\omega_H$, does not depend on the angle ϕ. From Eq. (10) it is easy to derive the equation for $\overline{\rho_{nn}}$.

$$\frac{\partial}{\partial t}\overline{\rho_{nn}}+\frac{\partial}{\partial\cos\theta}(\overline{u_{\cos\theta}}\overline{\rho_{nn}})=0, \tag{29}$$

where

$$\overline{u_{\cos\theta}}=\frac{1}{2\pi}\int_0^{2\pi}u_{\cos\theta}\,d\phi. \tag{30}$$

In particular for the $J_m=J_n$ optical transition we obtain

$$\overline{u_{\cos\theta}}=\frac{\sin^2\theta}{J\Gamma}(|G_{-1}|^2-|G_{+1}|^2), \tag{31}$$

so that if $|G_{-1}|>|G_{+1}|$, the value of the velocity $\overline{u_{\cos\theta}}$ is positive everywhere and it means that the angular momentum moves towards the z axis direction. Thus in the final state the angular momentum of particles must be oriented along the magnetic field [Fig. 5(a)]. It is important that the velocity $\overline{u_{\cos\theta}}$ [Eq. (31)] does not depend on the E_0 component of the vector polarization and its value is exactly determined by the projection of the electrical-field vector on the

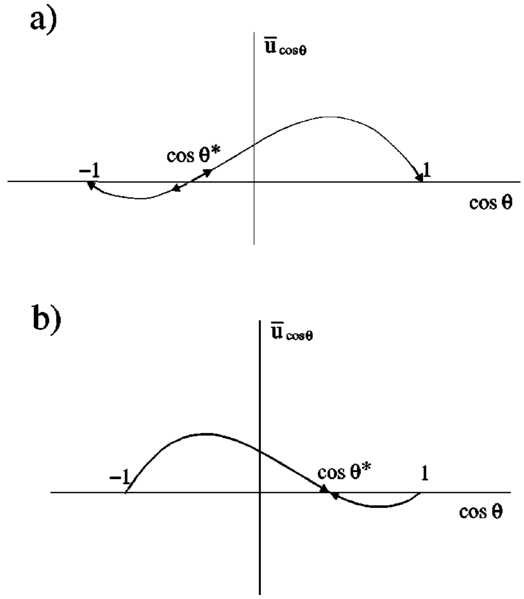

FIG. 4. The velocity $\overline{u}_{\cos\theta}$ as a function of $\cos\theta$ for (a) $J_m = J_n + 1$ and (b) $J_m = J_n - 1$ transitions.

plane orthogonal to the magnetic field only. This conclusion is valid for other types of optical transitions.

For the $J_m = J_n + 1$ optical transition we obtain

$$\overline{u}_{\cos\theta} = \frac{\sin^2\theta}{2J\Gamma}[|G_{-1}|^2(1+\cos\theta) - |G_{+1}|^2(1-\cos\theta)], \tag{32}$$

and $\overline{u}_{\cos\theta}$ as a function of $\cos\theta$ is shown in Fig. 4(a). This velocity is equal to zero at

$$\cos\theta^* = \frac{|G_{+1}|^2 - |G_{-1}|^2}{|G_{-1}|^2 + |G_{+1}|^2}. \tag{33}$$

This point is divergent because angular momentum moves towards $\theta \to 0$ for the initial state with $\theta < \theta^*$ and $\theta \to \pi$ if $\theta > \theta^*$. Thus angular momenta of the particles are oriented along or opposite the magnetic-field direction depending on what side of θ^* it was in the beginning [Fig. 5(b)].

At last consider the angular-momentum motion of the quantum system with the $J_m = J_n - 1$ optical transition. In this case

$$\overline{u}_{\cos\theta} = \frac{\sin^2\theta}{2J\Gamma}(|G_{-1}|^2(1-\cos\theta) - |G_{+1}|^2(1+\cos\theta)). \tag{34}$$

We can see [Fig. 4(b)] that the angle defined by the equation

$$\cos\theta^* = \frac{|G_{-1}|^2 - |G_{+1}|^2}{|G_{-1}|^2 + |G_{+1}|^2}, \tag{35}$$

where the velocity $\overline{u}_{\cos\theta}$ is equal to zero, is a convergent point and the angular momentum moves towards this angle

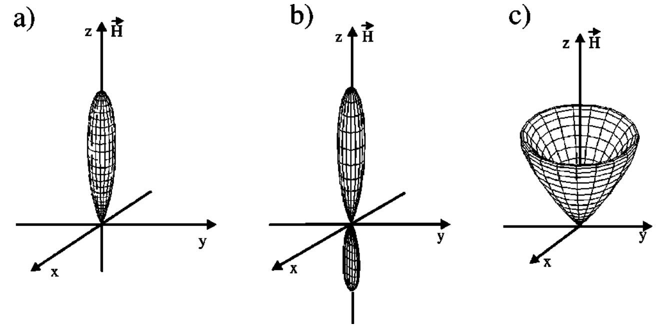

FIG. 5. The final distributions of angular momentum for (a) $J_m = J_n$, (b) $J_m = J_n$, and (c) $J_m = J_n - 1$ transitions for the elliptical polarization of light in the case of nonzero magnetic field.

$\theta \to \theta^*$ independent of its initial direction. Thus the final distribution has the form of a cone with angle θ^* at the cone top [Fig. 5(c)]. Note, the angle θ^* defined by Eq. (35) does not coincide with one defined by Eq. (19). In the particular case, for example $|G_{+1}| = 0$, this cone of angular-momentum orientations is degenerated into one direction along the magnetic field. The picture is in agreement with the exact quantum-mechanical solution [14] for this special case of mutual orientations of the magnetic field and the polarization of light.

V. CONCLUSION

We have derived an equation that describes a classical motion of the angular momentum of the closed two-level system that interacts with a resonant radiation. The equation has enabled us to describe the behavior of the angular-momentum orientation on the basis of introduced velocities, $u_{\cos\theta}$ and u_ϕ. As the result of this motion the system comes into a final state, that is the dark one, for $J_m = J_n$, $J_m = J_n - 1$ transitions, but the brightest one for the $J_m = J_n + 1$ transition and the arbitrary polarization of light. The results have been generalized for the arbitrary mutual orientation of the polarization vector and the direction of a nonzero magnetic field.

In the present analysis we have used the approximation of the weak light intensity $|G| \ll \Gamma_m$, however the proposed approach might easily be applied in the analysis of angular-momentum motion in the case of the high intensity limit $|G| \gg \Gamma_m$. The results of this consideration will be presented elsewhere.

ACKNOWLEDGMENTS

We extend our sincere thanks to S. Rautian, A. Shalagin, A. Tumaykin and S. Tchernov for many stimulating discussions. This work was supported by the Russian Foundation for Basic Research (Grant No. 98-02-17924).

APPENDIX

The term of the contribution into the ground state n due to the spontaneous decay of the excited state m has the form [12]

$$\Gamma_m \sum_{M_m, M'_m, \sigma} G^{J_m, M_m}_{J_n, M_n; 1,\sigma} C^{J_m, M'_m}_{J_n, M'_n; 1,\sigma} \rho(J_m, M_m | J_m, M'_m).$$

Due to the transformation (3) we can get this term in the following form

$$\hat{\Gamma}_m \rho_{mm}(\theta\phi) = \Gamma_m \sum_{M_m, M'_m, \sigma} \sum_{\kappa, q} \frac{\sqrt{2\kappa+1}}{\sqrt{2J_n+1}} (-1)^{J_n - M'_n} D^{\kappa *}_{q,0}(\phi, \theta, 0) C^{\kappa, q}_{J_n, M_n; J_n, -M'_n} C^{J_m, M_m}_{J_n, M_n; 1,\sigma} C^{J_m, M'_m}_{J_n, M'_n; 1,\sigma} \rho(J_m M_m | J_m M'_m). \tag{A1}$$

By summing over σ in Eq. (A1), we come to

$$\hat{\Gamma}_m \rho_{mm}(\theta\phi) = \Gamma_m \sum_{M_m, M'_m, \sigma} \sum_{\kappa, q} \frac{(2J_m+1)\sqrt{2\kappa+1}}{\sqrt{2J_n+1}} (-1)^{J_n - M'_n} D^{\kappa *}_{q,0}(\phi, \theta, 0) C^{\kappa, q}_{J_m, M_m; J_m, -M'_m} (-1)^{1+J_n+J_m+\kappa}$$
$$\times \begin{Bmatrix} J_n & 1 & J_m \\ J_m & \kappa & J_n \end{Bmatrix} \rho(J_m, M_m | J_m, M'_m)$$

or, using the relation between $6j$ symbols and the V function [13], to

$$\hat{\Gamma}_m \rho_{mm}(\theta\phi) = \Gamma_m \sum_{M_m, M'_m} \sum_{\kappa, q} \frac{(2J_m+1)\sqrt{2\kappa+1}}{\sqrt{2J_n+1}} (-1)^{J_m - M'_m} D^{\kappa *}_{q,0}(\phi, \theta, 0)$$
$$\times C^{\kappa, q}_{J_m, M_m; J_m, -M'_m} \left[\frac{(2J_n-\kappa)!(2J_m-\kappa)!}{(2J_n+\kappa+1)!(2J_m+\kappa+1)!} \right]^{1/2} V_\kappa(J_n, \ 1, \ J_m) \rho(J_m, \ M_m | J_m, \ M'_m). \tag{A2}$$

The explicit form of the V function for $J_m = J_n$ and $J_m = J_n \pm 1$ transitions is [13]:

$$V_\kappa(J,1,J) = 2[2J(J+1) - \kappa(\kappa+1)] \frac{(2J-1)!(2J+\kappa+1)!}{(2J+2)!(2J-\kappa)!},$$

$$V_\kappa(J,1,J+1) = \frac{(2J)!(2J+\kappa+3)!}{(2J+3)!(2J-\kappa)!},$$

$$V_\kappa(J,1,J-1) = \frac{(2J-2)!(2J+\kappa+1)!}{(2J+1)!(2J-\kappa-2)!}.$$

In particular, for the case of $J_m = J_n$, we get

$$\hat{\Gamma}_m \rho_{mm}(\theta\phi) = \Gamma \sum_{M_m, M'_m} \sum_{\kappa, q} \frac{\sqrt{2\kappa+1}}{\sqrt{2J_n+1}} (-1)^{J_m - M'_m}$$
$$\times D^{\kappa *}_{q,0}(\phi, \theta, 0) C^{\kappa, q}_{J_m, M_m; J_n, -M'_m}$$
$$\times \left[1 - \frac{\kappa(\kappa+1)}{2J_n(J_n+1)} \right] \rho(J_n, M_m | J_n, M'_m).$$

By using the fact that the equation

$$\Delta D^{\kappa *}_{q,0}(\phi, \theta, 0) = -\kappa(\kappa+1) D^{\kappa *}_{q,0}(\phi, \theta, 0),$$

is valid and due to the transformation (3), we finally obtain

$$\hat{\Gamma}_m \rho_{mm}(\theta\phi) = \Gamma_m \left[1 + \frac{\Delta}{2J_n(J_n+1)} \right] \rho_{mm}(\theta\phi).$$

In the case $J_m = J_n + 1$, Eq. (A2) takes the form

$$\hat{\Gamma}_m \rho_{mm}(\theta\phi) = \Gamma_m \sum_{M_m, M'_m} \sum_{\kappa, q} \frac{\sqrt{2\kappa+1}\sqrt{2J_m+1}}{2J_n+1}$$
$$\times (-1)^{J_m - M'_m} D^{\kappa *}_{q,0}(\phi, \theta, 0) C^{\kappa, q}_{J_m, M_m; J_m, -M'_m}$$
$$\times \left[\left(1 - \frac{\kappa(\kappa+1)}{2(2J_m-1)J_m} \right) \right.$$
$$\left. \times \left(1 - \frac{\kappa(\kappa+1)}{2(2J_m+1)J_m} \right) \right]^{1/2}$$
$$\times \rho(J_m, M_m | J_m, M'_m). \tag{A3}$$

Assuming that $J_n \gg 1$, we can transform Eq. (A3) to

$$\hat{\Gamma}_m \rho_{mm}(\theta\phi) \approx \frac{2J_m+1}{2J_n+1} \Gamma_m \left[1 + \frac{\Delta}{4J_m^2} \right] \rho_{mm}(\theta\phi).$$

In the same way for $J_m = J_n - 1$ we can derive

$$\hat{\Gamma}_m \rho_{mm}(\theta\phi) \approx \frac{2J_m+1}{2J_n+1} \Gamma_m \left[1 + \frac{\Delta}{4J_n^2} \right] \rho_{mm}(\theta\phi).$$

[1] A. Kastler, J. Phys. Radium **11**, 55 (1950).
[2] W. Happer, Rev. Mod. Phys. **44**, 169 (1972).
[3] V. S. Smirnov, A. M. Tumaikin, and V. I. Yudin, Zh. Eksp. Teor. Fiz. **96**, 1613 (1989) [Sov. Phys. JETP **69**, 913 (1989)].
[4] A. B. Taichenachev, A. M. Tumaikin, V. I. Yudin, and G. Nienhuis, Zh. Eksp. Teor. Fiz. **108**, 415 (1995) [Sov. Phys. JETP **81**, 224 (1995)].
[5] J. Dalibard and C. Cohen-Tannoudji, J. Opt. Soc. Am. B **6**, 2023 (1989).
[6] V. Milner, B. M. Chernobrod, and Y. Prior, Phys. Rev. A **60**, 1293 (1999).
[7] K. A. Nasyrov and A. M. Shalagin, Zh. Eksp. Teor. Fiz. **81**, 1649 (1981) [Sov. Phys. JETP **54**, 877 (1981)].
[8] K. A. Nasyrov, J. Phys. A **32**, 6663 (1999).
[9] G. D. Sprouse and L. A. Orosco, Annu. Rev. Nucl. Part. Sci. **47**, 429 (1997).
[10] J. E. Simsarian, A. Chost, G. Gwinner, L. A. Orosco, D. D. Sprouse, and P. A. Voytas, Phys. Rev. Lett. **76**, 3522 (1996).
[11] M. Ducloy, J. Phys. (Paris) **36**, 927 (1975).
[12] S. G. Rautian and A. M. Shalagin, *Kinetic Problems of Nonlinear Spectroscopy* (Elsevier, Amsterdam, 1991).
[13] D. A. Varshalovich, A. N. Moskalev, and A. N. Khersonskii, *Quantum Theory of Angular Momentum* (Nauka, Leningrad, 1975).
[14] A. M. Tumaikin and V. I. Yudin, Zh. Eksp. Teor. Fiz. **98**, 81 (1990) [Sov. Phys. JETP **71**, 43 (1990)].