Photoinduced angular motion of states and field splitting of levels

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(Submitted 10 January 1983)

Pis'ma Zh. Eksp. Teor. Fiz. **37**, No. 4, 184–187 (20 February 1983)

Wave equations are derived for the probability amplitudes for the angular motion of degenerate states of a resonant transition induced by elliptically polarized light. The spectrum of the field-induced splitting of levels is found by quantizing the azimuthal motion of quasiparticles described by these equations.

PACS numbers: 03.65.Ge, 42.50. + q

1. In resonant radiative processes, the sum rules for dipole transitions regulate not only the optical but also the angular degrees of freedom. The level degeneracy along the direction of the angular momentum, which is associated with these degrees of freedom, is lifted by a field-induced splitting that occurs in nonlinear spectroscopy.[1]

In linearly and circularly polarized light, the Hamiltonian of the resonant interaction in the JM angular-momentum projection basis can be diagonalized by virtue of the axial symmetry. For elliptically polarized light, however, in the case without a symmetry axis, the field-induced splitting spectrum can be found by solving the secular equation. The difficulties encountered in solving this equation increase with increasing J. In this letter we wish to propose a new approach, which is particularly useful in the semiclassical limit, $J \gg 1$. This approach is based on wave equations for the probability amplitudes for the angular motion in the basis of coherent states of the SU(2) rotation group.[2,3]

The photoinduced normal waves of the angular-motion probability amplitudes correspond to quasiparticles whose classical analog is the angular momentum. The angular momenta of these quasiparticles trace out paths which serve as characteristics of the wave equations.

As an example, we might cite the quasiparticles which were introduced in a description of a nonlinear-optics magnetic resonance involving a transition in an atom interacting with linearly polarized light.[3]

In this letter we quantize the energy of the quasiparticles excited by an elliptically polarized light wave. The spectrum of field splitting of levels is determined by the energy spectrum of the quasiparticles whose wave functions satisfy the requirement of a cyclic behavior during complete revolutions.

2. We consider a dipole-allowed transition between levels with identical values of the total angular momentum (a J–J transition in atoms; the Q branch in molecules). We represent the dipole operators of the Hamiltonian of the electrodipole interaction and the state vectors in a basis of coherent states of the SU(2) rotation group:

$$|J\xi> = \sum_{M=-J}^{J} (C_{2J}^{J-M})^{1/2} \exp(iM\xi^*)|JM> . \quad (1)$$

Here C_{2J}^{J-M} is a binomial coefficient, and $\xi^* = \phi + i \ln \mathrm{tg}(\theta/2)$ is a complex coordinate in the band $0 < \mathrm{Re}\xi < 2\pi$, determined by the azimuthal and polar angles ϕ and θ on the angular-momentum direction sphere. According to the Wigner-Eckart theorem, the dipole momentum operators are proportional to irreducible tensor operators, which are the same, in case of a J–J transition, as the infinitesimal operators of the rotation group. In basis (1) the Cartesian components of the irreducible tensor operators are

$$\hat{T}_z = i\partial_\xi , \quad \hat{T}_x = J\cos\xi - \sin\xi\,\partial_\xi, \quad \hat{T}_y = -J\sin\xi - \cos\xi\,\partial_\xi . \quad (2)$$

We orient the coordinate system in such a manner that the y axis coincides with the propagation direction of the elliptically polarized light wave, while the z axis coincides with the major semiaxis of the polarization ellipse. The standard Schrödinger equations for the wave functions $\Psi(\xi) = \langle J\xi\,|\Psi\rangle$ of the angular motion of the resonant-transition states $|n\rangle$ and $|m\rangle$ are

$$(E - \Omega)m(\xi) = (\hat{T}_z + ik\hat{T}_x)n(\xi),$$
$$(E + \Omega)n(\xi) = (\hat{T}_z - ik\hat{T}_x)m(\xi). \quad (3)$$

Here E and 2Ω are the energy of the states and the difference between the transition frequency and the light frequency, expressed in units of G_z; $k = G_x/G_z$ is the eccentricity, which varies from 0 to 1; $G_{z(x)} = d_{mn}\mathscr{E}_{z(x)}[J(J+1)(2J+1)]^{-1/2}$, d_{mn} is a reduced transition matrix element; and $\mathscr{E}_{z(x)}$ is the electric field of the light wave.

3. By virtue of the rotational symmetry of the angular-motion equations, we can distinguish a class of wave functions which meet the requirement of a cyclic behavior, and we can find the spectrum (E) of the field splitting of the states of the m–n transition which are degenerate in the projection of the angular momentum. An important symmetry of the equations is the invariance with respect to the combined transformation consisting of a rotation R_y through an angle π around the y axis (the wave vector of the light) and a simultaneous change of the sign of the energy E, Ω to $-E, -\Omega$. In addition, R_y invariance is exhibited by the equation satisfied by the wave function of the state $|m\rangle$ or $|n\rangle$:

$$(E^2 - \Omega^2)\, m(\xi) = (\hat{T}_z^2 + k^2\hat{T}_x^2 + k\hat{T}_y)\, m(\xi). \quad (4)$$

The reason for this enhanced symmetry is that Eq. (4) simultaneously describes two types of quasiparticles—with energies E and $-E$—and the transformation R_y transforms the wave function of one quasiparticle into that of the other. For the eigenfunctions of Eq. (4), the invariance with respect to the rotation R_y allows us to introduce the quantum number $r = \pm 1$, which is the eigenvalue of the operator R_y: $R_y m_E(\xi) = r m_E(\xi)$. Since R_y operates by the rule $R_y m(\xi) = (-1)^J m(\pi - \xi)$, we find

$$(-1)^J m_E(\pi - \xi) = r\, m_E(\xi). \quad (5)$$

Condition (5) can hold only for definite discrete values of the energy. These quantized quasiparticle energies can be found by solving Eq. (4) on the real axis:

$$(1 - k^2 \sin^2 \xi)\, \partial_\xi^2 m + [(2J - 1) k \sin \xi + 1] k \cos \xi\, \partial_\xi m + [\epsilon^2 - k^2 J^2 \times (\cos^2 \xi + J^{-1} \sin^2 \xi) + kJ \sin \xi\,] m = 0, \qquad \epsilon^2 = E^2 - \Omega^2. \tag{6}$$

The energy spectrum of the quasiparticles is easily found in the semiclassical limit $J \gg 1$ within $(\epsilon/J)^2 \sim O(1/J)$, through the use of the approximate equation

$$(1 - k^2 \sin^2 \xi)\, \partial_\xi^2 m + 2Jk^2 \sin \xi \cos \xi\, \partial_\xi m + J^2 [(\epsilon/J)^2 - k^2 \cos^2 \xi] m = 0. \tag{7}$$

The substitution $m(\xi) = (1 - k^2 \sin^2 \xi)^{J/2} \mathscr{M}(\xi)$ converts Eq. (7) to a Schrödinger equation of the form

$$\partial_\xi^2 \mathscr{M} + J^2 p^2(\xi) \mathscr{M} = 0, \tag{8}$$

where

$$p^2(\xi) = [(\epsilon/J)^2 - 1]\, \frac{\delta - k^2 \sin^2 \xi}{(1 - k^2 \sin^2 \xi)^2}, \quad \delta = \frac{(\epsilon/J)^2 - k^2}{(\epsilon/J)^2 - 1}$$

Condition (5) is extended to the function $\mathscr{M}(\xi)$. By virtue of the periodicity of the potential, we can restrict the discussion to the band $0 \leqslant \mathrm{Re}\xi \leqslant \pi$ in solving Eq. (8). Two linearly independent solutions are written in the semiclassical approximation as

$$\mathscr{M}_\pm(\xi) = [J p(\xi)]^{-1/2} \exp\left\{ \pm iJ \int_{\xi_p}^{\xi} d\eta\, p(\eta) \right\}, \tag{9}$$

where ξ_p is the coordinate of the turning point $p(\xi) = 0$.

The boundaries of the energy spectrum, $0 \leqslant (\epsilon/J)^2 \leqslant 1$, are given by the classical estimate of the Hamiltonian of Eq. (4). On the real axis, $\mathrm{Re}\xi = \phi$, the semiclassical momentum $p(\xi)$ determines classically allowed and forbidden bands of angular motions, depending on the values of $(\epsilon/J)^2$ and k^2. For quasiparticle energies in the upper sector, from k^2 to 1, all the azimuthal directions are classically allowed [$p^2(\xi) > 0$ in the interval $0 < \phi \leqslant 2\pi$]. The pattern of azimuthal motions of the quasiparticles in the upper sector corresponds to an above-barrier reflection with two turning points, at the lattitudes $\theta_+ = \mathrm{arctg}\sqrt{-k^2/\delta}$ and $\pi - \theta_+$. These points lie on the imaginary axis $\mathrm{IM}\xi$, passing through points with the azimuthal angles $\phi = 0, \pi$. We construct a general solution from functions (9); imposing condition (5), we find the quantization condition

$$J \int_0^\pi d\eta\, p(\eta) = \pi M, \tag{10}$$

where M is the number of nodes of the wave function in the band $0 \leqslant \phi \leqslant \pi$.

In the lower sector, $0 \leqslant (\epsilon/J)^2 < k^2$, there are two turning points on the real axis, at $\xi_0 = \arcsin\sqrt{\delta/k^2}$ arcsin and $\pi - \xi_0$. In the bands $0 \leqslant \xi < \xi_0$ and $\pi - \xi_0 < \xi < \pi$ there is an above-barrier, classically forbidden motion [$p^2(\xi) < 0$], while in the band $\xi_0 \leqslant \xi \leqslant \pi - \xi_0$ there is motion in a well. The quantization of the azimuthal motions is determined by the well, whose dimensions depend on the quasiparticle energy $(\epsilon/J)^2$ and the elliptical parameters of the light. If the polarization of the light tends towards circular, $k \to 1$, the turning point ξ_0 tends toward $\pi/2$, and the well collapses, leaving a singularity of the quasimomentum $p(\xi) \propto |\cos^{-1}\xi|$ at the point $\xi = \pi/2$. The contraction of the well is accompanied by a simultaneous increase in its depth, in such a manner that the condition for a semiclassical behavior of the action S on the paths between the turning points is not disrupted ($S \gg 1$). The path-quantization condition is

$$J \int_{\xi_0}^{\pi - \xi_0} d\eta\, p(\eta) = \pi M, \tag{11}$$

where M is the number of nodes of the wave function of the angular motions in the well. In the limit $k = 1$ we must take into account the nodes of the function $\cos^J \xi$, which are related to the substitution $m(\xi) = \cos^J \xi \mathscr{M}(\xi)$. Condition (11) can then be written

$$\lim_{k \to 1} \left\{ \int_{\xi_0}^{\pi - \xi_0} d\eta\, p(\eta) \right\} = \pi\,(1 + M/J)\,. \tag{12}$$

It follows from the asymptotic solutions of Eq. (7) in the limit $|\xi| \to \infty$ that the maximum number of nodes (M) of the wave functions of states with angular momentum J is limited by the value of J.

In conclusion, we wish to point out an interesting analogy between the motion of the quasiparticles and the rotation of tops. It may be concluded from Eq. (4) that the quadratic spectrum ϵ^2 of the quasiparticles or the field-splitting spectrum is the same as the spectrum of an asymmetric top rotating in a gravitational field. The moments of inertia of this top and the moments of the gravitational forces are of an especially resonant nature. By varying the eccentricity of the resonant light, we can produce tops with various symmetries. Axisymmetric tops are formed by linearly and circularly polarized light. Elliptically polarized light produces an axisymmetric top.[2]

I wish to thank G. I. Surdutovich for a useful comment and A. V. Gaĭner and S. G. Rautian for useful discussions.

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Translated by Dave Parsons
Edited by S. J. Amoretty