Light-induced rotation of atoms and magnetic resonance

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A universal nonlinear optical phenomenon of the rotation of moments of resonant states of atoms (molecules) in an electromagnetic wave is established. The structure of the nonlinear resonance in a magnetic field, owing to the rotation of atoms, is described.

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According to classical ideas, the total moments of atoms precess in a magnetic field around the direction of the field. For low light intensities, the precession causes splitting of the spectral lines (Zeeman effect). On the other hand, in nonlinear spectroscopy, which involves intense radiation, so-called field-splitting of spectral lines occurs. In analogy to the Zeeman effect, we can say that field splitting is related to the motion of moments of the combining states in the electromagnetic wave. This motion occurs for any radiative transition and is a result of strict conservation laws and selection rules.

Just as the action of light on the translational motion of an atom follows from the conservation laws for energy and momentum in absorption and emission, the motion of moments of states follows from the law of conservation of angular momentum. The form of the classical trajectories of moments of degenerate atomic states, excited by light, is determined by the polarization and spectral composition of this light.

In what follows, we shall examine the classical motion of the moments of a J–J transition in linearly polarized monochromatic light. Linearly polarized light gives rise to rotation of moments with an angular velocity determined by its intensity. This occurs as a result of the stimulated emission of a quantum of one circular polarization and absorption of another. Since the intensity of the circular components of linear polarization is equal, rotation is possible in both directions around the electric vector **E** of the light wave.

In a magnetic field, Larmor procession is superimposed on the light-induced rotation moments. As a result, the moments begin to precess around the resultant vectors, consisting of the vector **E** of the light wave and of the intensity vector of the external magnetic field.

Light-induced rotation of atoms can be realized by using the fruitful concepts of coherent states of the rotation group for quasiclassical representation of tensor operators.[1] In this representation, Schrödinger's equation for the probability amplitudes of the states m and n involved in the transition at an exact resonance ($\omega = \omega_{mn}$) in an electromagnetic field have the form

$$i\left(\frac{\gamma}{2}+\frac{\partial}{\partial t}\right)\begin{pmatrix} m \\ n \end{pmatrix}=\begin{pmatrix} \Delta \hat{T}_z^{JJ} & G\hat{T}_x^{JJ} \\ G\hat{T}_x^{JJ} & \Delta T_z^{JJ} \end{pmatrix}\begin{pmatrix} m \\ n \end{pmatrix}, \tag{1}$$

$$\Delta = \mu_0 H, \quad G = \mathcal{E} d_{mn}/(J(J+1)(2J+1))^{1/2},$$

where H is the external magnetic field, oriented along the z axis; μ_0 is the magnetic moment of the mJ and nJ shells; ϵ is the electric component of the light propagating along the z axis and polarized along the x axis; d_{mn} is the reduced dipole-moment matrix element for the transition. The tensor part of the electric dipole and magnetic interactions is given by the operators

$$\hat{T}_z^{JJ} = J - \mu^* \partial/\partial\mu^*, \quad \hat{T}_x^{JJ} = J\mu^* + 0{,}5\,(1-\mu^{*2})\,\partial/\partial\mu^*;$$

$\mu^* = \tan(\theta/2)\exp(i\psi)$ is the complex coordinate in a space which is a stereographic projection from the directional sphere.[2] The initial conditions, corresponding to absorption, are

$$\begin{pmatrix} m \\ n \end{pmatrix}_{t=0} = \sum_{m=-J}^{J}\begin{pmatrix} J-M \\ 2J \end{pmatrix}^{1/2} \exp(i a_M)\,\mu^{*J-M}\begin{pmatrix} 0 \\ 1 \end{pmatrix}, \tag{2}$$

where $\binom{J-M}{2J}$ is a binomial coefficient, and a_M is a random phase.

Equations (1) are diagonalized for the states $\varphi_\pm = m \pm n$. Quasiparticles, corresponding to the states, are distinguished by the fact that their moments rotate around different axes. The orientation of the axes and the rate of precession are determined from the classical equations of motion of the quasiparticle moments

$$d\mu^*/dt = j_\pm(\mu^*),\ j_\pm(\mu^*) = i\,(-\Delta\mu^* \pm G(1-\mu^{*2})/2), \tag{3}$$

which are the equations of the characteristics (streamlines) of the wave equations for the state amplitudes:

$$(\gamma/2 + \partial/\partial t)\,\varphi_\pm = -j_\pm(\mu^*)\,\partial\varphi_\pm/\partial\mu^* + J(\partial j_\pm(\mu^*)/\partial\mu^*)\,\varphi_\pm. \tag{4}$$

Equations (3) in the μ plane are equivalent to Bloch's equations $\dot{\mathbf{n}} = [\vec{\nu}_\pm \mathbf{n}]$ for the unit vector $\mathbf{n}$, oriented along the average moment and rotating around the axes $\vec{\nu}_\pm$ with frequency $\nu = \sqrt{\Delta^2 + G^2}$. The precession axes $\nu_\pm$ of the quasiparticle moments are collinear with the resultant of vectors $\mathbf{H}$ and $\pm\mathbf{E}$. It is easy to find the solution of Eqs. (4) and initial conditions (2)

$$\varphi_\pm(\mu^*, t) = \pm\sum_{M=-J}^{J}\begin{pmatrix} J-M \\ 2J \end{pmatrix}^{1/2} (-1-|a|^2)^{-J}\,(1 \pm a\,\mu^*)^{J-M}(\mu^* \mp a^*)^{J+M}\,e^{i a_M - \gamma t/2}$$

$$a = -\Delta/G + i\,(\nu/G)\,\mathrm{ctg}(\nu t/2),\ \nu = \sqrt{\Delta^2+G^2}, \tag{5}$$

and from these, the state amplitudes $m(\mu^*, t)$ and $n(\mu^*, t)$. Averaging the product of the wave functions over an ensemble of random phases a_M, it is possible to calculate the elements of the density matrix: population distribution ρ_{nn} and ρ_{mm} and directional

polarization ρ_{mn}. The magnitude of the absorbed power is determined by the induced polarization

$$P(t) = \hbar\omega G \operatorname{Im} \operatorname{Sp}\left\{\left(\frac{\partial}{\partial\mu^*} + \frac{\partial}{\partial\mu}\right)\rho_{mn}(\mu\mu^*)\right\} = \frac{4\hbar\omega J(J+1)G}{\pi} e^{-\gamma t}$$

$$\times \int \frac{d\operatorname{Re}\mu\, d\operatorname{Im}\mu}{(1+|\mu|^2)^2} \operatorname{Im}\left\{\frac{(|a|^2-1)\operatorname{Re}\mu + 2i\operatorname{Im}a^*}{(|a|^2+1)(1+|\mu|^2)}\left[\frac{|a|^2-1}{|a|^2+1} + \frac{4i\operatorname{Im}(a^*\mu)}{(|a|^2+1)(1+|\mu|^2)}\right]^{2J-1}\right\}. \tag{6}$$

The function in the integrand, which characterizes the directional distribution of absorbed power, for large J, has two sharp maxima (Fig. 1), traversing the circle $x^2+y^2=1$ in time $\tau = \nu^{-1}$. Without the magnetic field, the packets are localized near the points $x=\pm 1$ (light linearly polarized along the x axis induces the x component of the transition polarization). The slope of the localized packets increases with increasing J. The profile of the magnetic resonance $P(\Delta) = \int_0^\infty dt P(t)$ can be calculated in the M representation by choosing the axis of quantization z along $\mathscr{E}$. Then the state amplitudes have the form

$$\begin{pmatrix} m \\ n \end{pmatrix} = \sum_{M=-J}^{J} 0.5 \begin{pmatrix} J-M \\ 2J \end{pmatrix}^{1/2} \left[(-1)^M D^J_{MM'}(\psi,\theta,\psi)\begin{pmatrix} 1 \\ 1 \end{pmatrix}\right.$$

$$\left. + (-1)^{-M'} D^J_{M'M}(-\psi,-\theta,-\psi)\begin{pmatrix} -1 \\ 1 \end{pmatrix}\right] e^{i a_M - \gamma t/2} \tag{7}$$

Here, $D^J_{MM'}(\psi,\theta,\psi)$ is Wigner's matrix, $\psi = \pi - \arctan[(\nu/G)\cot(\nu t/2)]$, and $\theta = \arccos[(G^2+\Delta^2\cos\nu t)/\nu^2]$. This magnetic interaction leads to transitions between neighboring M sublevels of degenerate levels, thereby causing "frequency migration" of the light-induced polarization. In classical language, this process corresponds to phase modulation of polarization oscillations.

Coherence of the levels $\rho_{gg}(M,M')$ and the transition $\rho_{mn}(M,M')$ are also represented by the D matrices, but with different arguments. The absorbed power is concisely expressed in terms of the characters of the rotation group:

$$P(t) = \frac{\hbar\omega G^2}{\nu}\frac{\sin(\nu t)}{\sin(\omega/2)} e^{-\gamma t}\frac{d\chi^J(\omega)}{d\omega}, \quad \chi^J(\omega) = \frac{\sin((2J+1)\omega/2)}{\sin(\omega/2)},$$

$$\cos(\omega/2) = -(\Delta^2 + G^2\cos(\nu t)/\nu^2. \tag{8}$$

Finally, the profile of the magnetic resonance is given by the integral

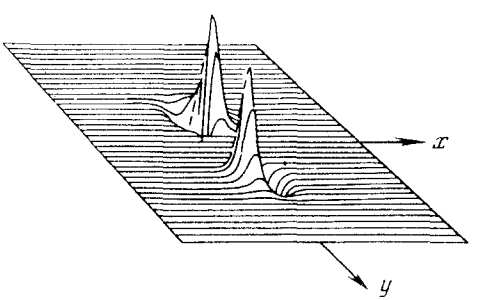

FIG. 1. Distribution of absorbed power in a J-J transition: $J=10$, $\Delta = G$, $t = \nu^{-1}/2$.

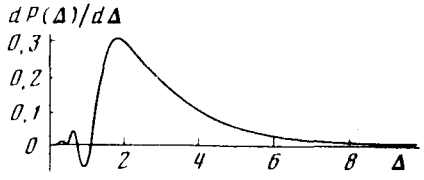

FIG. 2. Derivative of the magnetic resonance in the transition with moment $J = 6$ as a function of the splitting Δ, measured in units of γ.

$$P(\Delta) = \frac{\hbar\,\omega G^2}{\nu} \int_0^\infty dt\, e^{-\gamma t} \sin(\nu t)\, \frac{J \sin(J+1)\omega - (J+1)\sin J\,\omega}{\sin^3(\omega/2)} .$$

Figure 2 presents a calculation[1)] of the quantity $dP/d\Delta$ at saturation $(G/\gamma)^2 = 0.05$. The difference between this line profile and the dispersion profile increases with increasing saturation. The fine structure of the saturated absorption profile is explained by the amplitude and phase modulation of the absorbed power, owing to the light-induced rotation of the moments of the atomic states.

Similar structures were recently observed in experimental work,[3] which motivated the present paper.

Thus, the idea of light-induced rotation of atoms is of fundamental significance for nonlinear spectroscopy and, in particular, in the theory of Zeeman structures of nonlinear resonances for calculations and interpretation of nonlinear absorption line profiles.[4,5]

The existence of exact results for the problem being examined stems from the fact that the dynamic symmetry group of the interaction coincides with the rotation group SU(2). For this reason, the solutions (7), rotation of the initial state, are formed from a finite-dimensional irreducible representation of the dynamic group SU(2). For an inexact resonance or a transition with a change in J, the dynamic group no longer reduces to simple rotations. However, the Schrödinger equation can be solved in a basis of coherent states of the rotation group without dynamic symmetry. The classical trajectories of the moments in these cases are also characteristics.

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