Polarization moments in a state with high angular momentum

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(Received 19 April 1985)
Opt. Spektrosk. 60, 406–410 (February 1986)

This paper is devoted to a graphic treatment of polarization moments (PM) and the concept of coherence for a classical ensemble of particles which can be described using the probability density (PD).

To describe the interaction of light with atoms and molecules the coefficients of the expansion of the quantum density matrix over the irreducible tensor operators, i.e., the PM are often used.[1,2] However, when transitions with high angular momentum and interactions nonlinear with respect to the light field are involved, one cannot always succeed in finally solving the equations of motion of the PM.

As experiment shows[3,4] these difficulties can be in definite measure overcome by using the asymptotic equations of motion of the PM for high values of the angular momentum

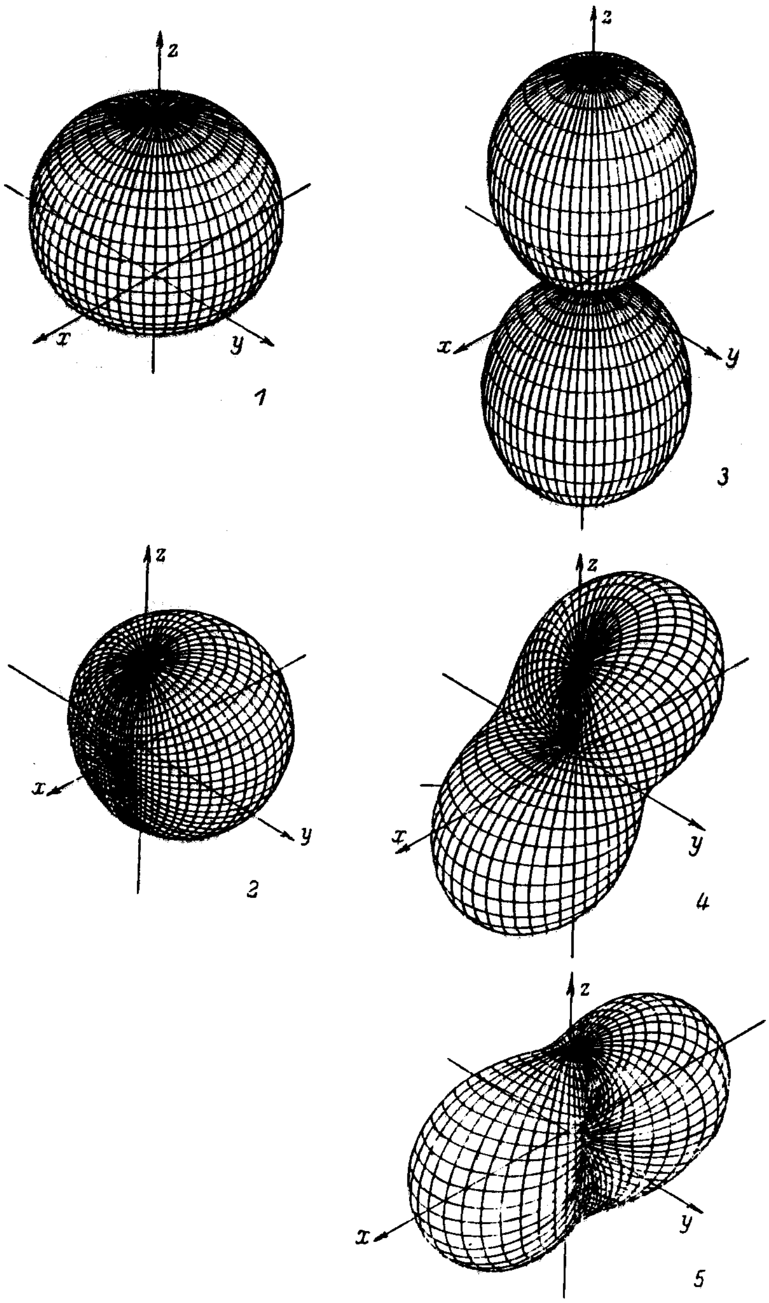

FIG. 1. Probability density for various numerical values of the polarization moments. 1—$\rho_0^0 = 1, \rho_0^1 = 0.3$; 2—$\rho_0^0 = 1, \rho_1^1 = -\rho^1_{-1} = 0.15$; 3—$\rho_0^0 = 1,\ \rho_0^2 = 0.3$; 4—$\rho_0^0 = 1,\ \rho_1^2 = -\rho^2_{-1} = 0.15$; 5—$\rho_0^0 = 1,\ \rho_2^2 = \rho^2_{-2} = 0.15$

 0030-400X/86/020248-03$05.00

J.[5] On the other hand, such an ensemble of particles can be described using the classical analogue of the density matrix—the probability density $\rho(\theta,\varphi,t)$. The physical meaning of the value $\rho(\theta,\varphi,t)\sin\theta\, d\theta\, d\varphi$ is the probability that at the time t the angular momentum vector **J** is located between angles θ and $\theta + d\theta$; φ and $\varphi + d\varphi$. It is not surprising that in a definite way expanding $\rho(\theta,\varphi,t)$ over the spherical harmonics $Y_{KQ}(\theta,\varphi)$,[6] which are a particular example of the components of the irreducible tensors, we obtain the equations of motion of the coefficients of the expansion coinciding with the corresponding asymptotic equations for the PM. The expansion

$$\rho(\theta, \varphi, t) = (4\pi)^{1/2} \sum_{k=0}^{\infty} (2K+1)^{1/2} \sum_{Q=-K}^{K} \rho_Q^K(t)\, Y_{KQ}^{*}(\theta, \varphi)$$

was performed in such a way that the coefficients ρ_Q^K and the spherical harmonics Y_{KQ} turn out to be covariant. Then the value of ρ_Q^K is proportional to the average value $\langle Y_{KQ}\rangle$ in the state examined. The fact stated permits regarding ρ_Q^K as the classical analogue of the PM.

It is well known that the PM of rank $K = 0$ characterizes the population of the state and in fact is the normalization factor for the remaining PM. In the classical limit the PM with rank $K \neq 0$ describe the redistribution of the probability of detection of the angular momentum over the directions in space. Since the function $\rho(\theta,\varphi,t)$ is real, the PM are connected by the relation $\rho_Q^K = (-1)^Q \rho_{-Q}^{K*}$ and at $Q \neq 0$ can appear only in pairs.[1,2]

In Fig. 1 isometric projections of the function $\rho(\theta,\varphi)$

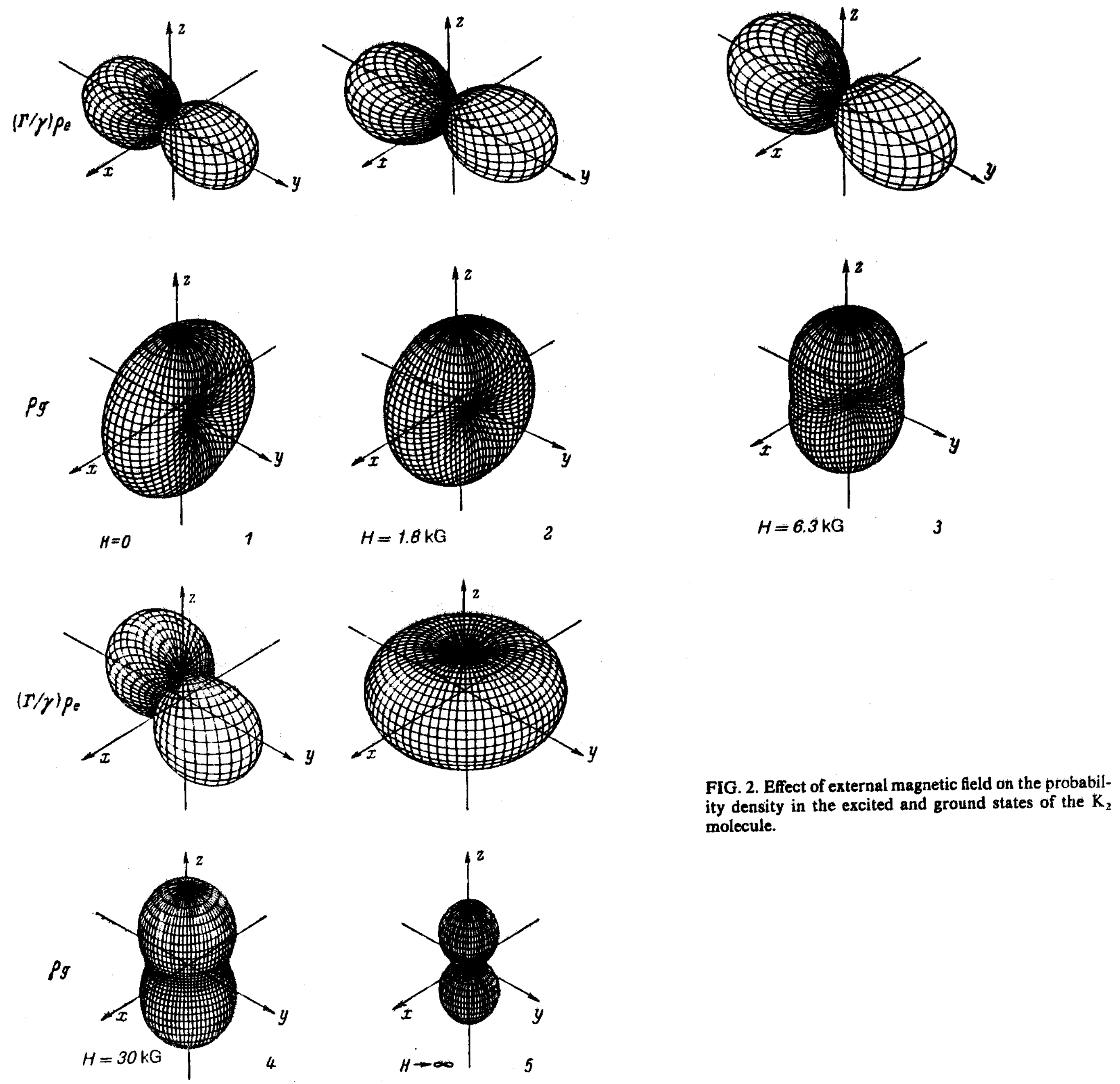

FIG. 2. Effect of external magnetic field on the probability density in the excited and ground states of the K_2 molecule.

modeled on a computer are given for several cases where the ensemble is characterized by the minimum number of PM of the lower ranks. These distributions do not correspond to any concrete process of excitation. They are employed to illustrate graphically the physical meaning of the PM in the classical limit. 1 and 3 in Fig. 1 correspond to the situation where it is accepted to state that there is no coherence in the ensemble, i.e., $Q=0$. These PD have full rotational symmetry in relation to the z axis. In the remaining cases coherence is created in the ensemble, $Q\neq 0$ (in a quantum examination $Q=\Delta M_J$ characterizes between which magnetic sublevels M_J coherence is created). In these figures z is the axis of rotational symmetry of order Q, where Q characterizes the components of the PM depicted. Such a result signifies that in the case of a classical examination of the ensemble of particles coherence can be treated from the viewpoint of symmetry of the PD.

The given approach aids interpreting the experimental results and calculations performed in the PM apparatus. For example, for experiments on the free quantum oscillations in the ground electronic state of diatomic molecules[8] the effect of an external magnetic field on the transverse alignment $\rho^2_{\pm 2}$ can be regarded as precession under the influence of this field $\rho(\theta,\varphi)$ [Fig. 1, (5)] around the z axis, which coincides with the direction of the vector of the field. However, one must always remember that the PD characterizes an ensemble of particles and the effect of a magnetic field under definite conditions; for example, the linear and nonlinear Hanle effect,[2,3] can be manifested in a more complex way than simple precession of the distribution.

As an example let us examine the effect of an external magnetic field on the functions of the PD of the ground ρ_g and excited ρ_e states for the case of excitation of the transition $(v''=1, J''=72)\,^1\Sigma_g^+\,(v'=8, J'=72)\,^1\Pi_u$ in the K_2 molecule. Such a transition is effectively excited by the 632.8-nm line of a He–Ne laser and is often used in experimental investigations (see, for example, Ref. 9). We assume that the laser beam is propagated along the x axis and is polarized along y. The external magnetic field is directed along z.

To find the PM of the ground and excited states necessary for construction of PD from Eq. (1) depending on the magnetic field intensity H the system of equations given in Ref. 5 was solved. Here it was assumed that the rate Γ_p of absorption of the laser light is related to the relaxation rate γ of the ground state of the transition as 10 to 3. The remaining parameters of the transition are as in Ref. 3. In particular the Lande factor of the ground state $g''=1.18\times 10^{-5}$ and of the excited $g'=1.9\times 10^{-4}$ Bohr magnetons. The relaxation rate Γ of the excited state is ~ 100 times greater than the absorption rate of the laser light. Then in Fig. 2 one can see that linearly polarized light induces in consequence of nonlinear absorption an anisotropic distribution of the angular momenta in the ground and excited states in the absence of a magnetic field (1). On switching on and increasing the magnetic field intensity, at first its effect on the ground state is more noticeably manifested. Gradual averaging of the angular momenta in the xy plane, situated perpendicular to the external magnetic field, occurs. The distribution of the excited state at the same time increase a bit in the volume, which corresponds to an increase in the total population of the level. This distribution is also extended along the y axis (2, 3). Such an extension corresponds to an increase in the degree of polarization of the radiation from the excited level on observation along the z axis. On a further increase in the field the PM of the excited state (4) also noticeably swings around. At very high values of the magnetic field intensity (5) complete averaging occurs in the xy plane of both the PD for the ground and the excited states. This corresponds to total depolarization of the fluorescence from the excited level on observation along the z axis.

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