Manifestation of the sixth-order polarization moment in the Hanle-effect signal of the electronic ground state of dimers

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(Received 28 September 1982)
Opt. Spektrosk. 55, 1105–1108 (December 1983)

1. Efficient optical absorption of laser light from a vibration–rotational level (v'', J'') of the electronic ground state of dimers can give rise to polarization moments[1] $\phi_q^{\varkappa}$ of various orders $\varkappa < 2J$ in this state. Destruction of the moments $\phi_q^{\varkappa}$ by a magnetic field H has been observed in laser-induced fluorescence as a manifestation of the ground-state Hanle effect,[2] and it has been observed in the case of a sinusoidally modulated excitation as a resonance of quantum beats.[3] These are apparently the only methods presently available for determining the Lande factor for an individual level (v'', J'') of the electronic ground state and the relaxation rates of the polarization moments.

There is the very interesting possibility of observing narrower resonances with $\varkappa > 2$, associated with the coherence of magnetic sublevels $|\Delta M_J| > 2$. All that has been reported so far is the detection of a hexadecapole resonance $(\varkappa = 4)$, in particular, of Ne atoms with $j = 2$.[4] It was also suggested in Refs. 2 and 6 on the basis of the calculations of Ref. 5 and the K_2 Hanle effect $(v'' = 1, J'' = 72)$ that the narrow additional peak which was detected in Refs. 2 and 6 at $H = 0$ is caused by a relaxation of the hexadecapole moment. In this paper we show that this signal structure is a manifestation of a sixth-order moment $(\varkappa = 6)$. In the calculation below we outline a simple method for numerically solving the system of kinetic equations for the polarization moments for large angular momenta J.

2. Although the kinetic equations of the polarization moments describing the nonlinear phenomena in the Hanle effect have been formulated for arbitrary J,[7] the classical approximation of Ref. 5 has heretofore been used in calculations corresponding to experiments on the optical pumping of dimers with $J \gg 1$. This approximation has the advantage of simplicity, but in it we cannot introduce various relaxation rates $\gamma_{\varkappa}$ for the polarization moments of order $\varkappa$. In this paper, this deficiency can be remedied by writing kinetic equations for the polarization moments of the excited state f_Q^K and the ground state $\phi_q^{\varkappa}$ (K and $\varkappa$ are the ranks of the tensor, and Q and q are its projections) but assuming $J \to \infty$ in the equations for combining the angular momenta.

We write the system of equations as[3]

$$\dot{f}_Q^K = \Gamma_p \sum_{X\varkappa}^{K} F^{X\varkappa} \{\Phi^{(X)} \otimes \varphi^{(\varkappa)}\}_Q^K - (\Gamma_K - iQ\Omega) f_Q^K, \tag{1a}$$

$$\dot{\varphi}_q^{\varkappa} = -\Gamma_p \sum_{X\varkappa'}^{\varkappa} A^{X\varkappa'} \{\Phi^{(X)} \otimes \varphi^{(\varkappa')}\}_q^{\varkappa} + \lambda_q^{\varkappa} \delta_{\varkappa 0} \delta_{q0} - (\gamma_{\varkappa} - iq\omega) \varphi_q^{\varkappa}. \tag{1b}$$

Here $\{\Phi^{(X)} \otimes \varphi^{(\varkappa')}\}_q^{\varkappa} = \sum_{\xi q'} C_{X\xi\varkappa' q'}^{\varkappa q} \Phi_{\xi}^{X}(\mathbf{e}) \xi_{q'}^{\varkappa'}$ is an irreducible tensor product, $\Phi_{\xi}^{X}(\mathbf{e})$ is a function introduced by Dyakonov,[1] $\mathbf{e}$ is the polarization vector of the exciting light, and $C_{X\xi\varkappa' q'}^{\varkappa q}$ is a Clebsch–Gordan coefficient. The term $\lambda_q^{\varkappa} \delta_{\varkappa 0} \delta_{q0}$ is the number of events in which the population φ_0^0 is restored per unit time per unit volume, Γ_K and $\gamma_{\varkappa}$ are the relaxation rates of the moments f_Q^K and $\varphi_q^{\varkappa}$, and Γ_p is the absorption rate. The last terms in Eqs. (1a) and (1b) characterize the effect of the external magnetic field H; $\Omega = g'\mu_0 H/\hbar$ and $\omega = g''\mu_0 H/\hbar$ are the frequencies of the magnetic splitting of the excited and ground levels, which contain the Lande factors g' and g'', respectively. The coefficients ${}^{K}F^{X\varkappa}$ and ${}^{\varkappa}A^{X\varkappa'}$ are given by[3]

$${}^{K}F^{X\varkappa} = \frac{\Pi_{J'J''\varkappa}^{3}}{\Pi_{J''K}} (-1)^{X+1} \begin{Bmatrix} K & J' & J' \\ X & 1 & 1 \\ \varkappa & J'' & J'' \end{Bmatrix}, \tag{2}$$

$${}^{\varkappa}A^{X\varkappa'} = \frac{[1 + (-1)^{\varkappa+X+\varkappa'}]\,\Pi_{J'X\varkappa'}^{2}}{2\Pi_{\varkappa}} (-1)^{J'-J''+\varkappa'} \begin{Bmatrix} \varkappa & X & \varkappa' \\ J'' & J'' & J'' \end{Bmatrix} \begin{Bmatrix} 1 & 1 & X \\ J'' & J'' & J' \end{Bmatrix}, \tag{3}$$

where $\Pi_{abc\ldots} = \sqrt{(2a+1)(2b+1)(2c+1)\ldots}$, and J'' and J' are the angular momenta of the ground and excited states, respectively.

Using the expansion of the $9j$ symbols in terms of $6j$ symbols and also the asymptotic expressions for the $6j$ symbols for $J', J'' \to \infty$,[8] we can show that the optical pumping becomes symmetric and that the coefficients ${}^{K}F^{X\varkappa}$ and ${}^{\varkappa}A^{X\varkappa'}$ agree and simplify to

$${}^{\varkappa}A^{X\varkappa'} = (-1)^{\Delta} \frac{\Pi_{X\varkappa'}}{\Pi_{\varkappa}} C_{1\Delta\,1-\Delta}^{X0} C_{\varkappa 0\,X0}^{\varkappa' 0}, \tag{4}$$

where $\Delta = J' - J''$. Equations (1a) and (1b) are written for an open system with the inverse spontaneous and stimulated

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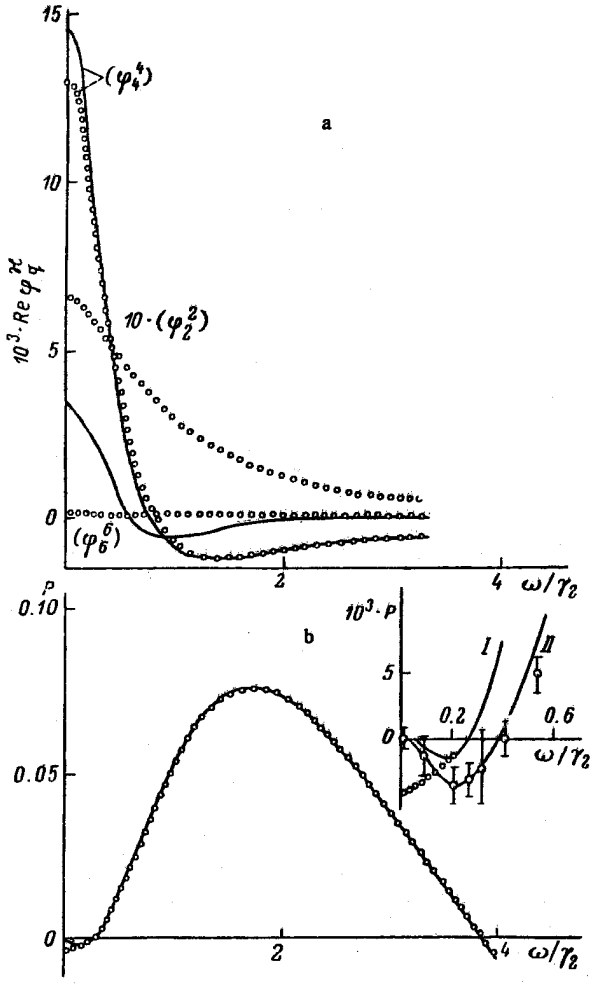

FIG. 1. Real parts of the polarization moments of various orders (a) and the corresponding Hanle lines (b).

transitions ignored. This case is completely realistic for the laser optical pumping of dimers when the rate ($\Gamma_{J'J''}$) of the return from the (v', J') state to the (v'', J'') original state is much lower than the Γ_K, and, furthermore, we have $\Gamma_p \ll \Gamma_K$. To incorporate spontaneous transitions we would have to add a term $\Gamma_{J'J''}\delta_{K\varkappa}\delta_{0q} f_Q^K$, to Eq. (1b), and to incorporate stimulated transitions we would have to add a term $\times\Gamma_p \Sigma_{XK'}^K A^{XK'}\{\Phi^{(X)} \otimes f^{(K')}\}_Q^K$ to Eq. (1a) and a term $\Gamma_p \Sigma_{XK}^\varkappa F^{XK}\{\Phi^{(X)} \otimes f^{(K)}\}_q^\varkappa$ to Eq. (1b).

3. System (1a), (1b) has been solved for $\dot f_Q^K = \dot\varphi_q^\varkappa = 0$. Ignoring the decrease in the absolute values of the moments with increasing order, we can set, for example, $f_0^K = \varphi_q^\varkappa = 0$ for $K, \varkappa > 8$. We assume that the exciting beam and its vector E are directed perpendicular to the field H. The system of equations has been solved numerically for the experimental parameters of Refs. 2 and 6 for K_2 with excitation by the beam from a He–Ne laser (632.8 nm) of the transition $(v'' = 1, J'' = 72)X^1\Sigma_g^+ \to (v' = 8, J' = 72)B^1\Pi_u$. Specifically, we adopt the parameters $\Gamma_p = 10^6$ sec^{-1}, $\Gamma_K = 8.62\times10^7$ sec^{-1}, $g' = 1.90\times10^{-4}$, and $g'' = 1.18\times10^{-5}$. Figure 1(a) shows the real parts of the moments of the lower level ($\varphi_{\pm2}^2$, $\varphi_{\pm4}^4$, and $\varphi_{\pm6}^6$) for identical values of $\gamma_\varkappa = 0.3\times10^6$ sec^{-1} (the solid curves) and for $\gamma_{\varkappa<6} = 0.3\times10^6$ sec^{-1} and $\gamma_6 = 0.3\times10^8$ sec^{-1} (dotted curves). We see that in the second case the sixth moment essentially vanishes because of the more rapid relaxation, while the other moments change only slightly.

4. Figure 1(b) shows the corresponding Hanle-effect signal in a quantity which is measured experimentally: the degree of polarization $P = (I^\pi - I^\sigma)/(I^\pi + I^\sigma)$ of the laser-induced fluorescence, observed along the vector E of the exciting beam. The intensity of the laser-induced fluorescence with polarization e' is[1]

$$I(e') \sim (-1)^\Delta \sum_K C_{1\Delta\,1-\Delta}^{K0} \sum_Q (-1)^Q f_Q^K \Phi_{-Q}^K. \qquad (5)$$

In this geometry we have $P(0) = 0$; the increase in P with increasing H is a manifestation of the Hanle effect from the lower level (v'', J''), while a decrease is a manifestation of the effect from the excited level (v', J'). We see that, in addition to the sixth moment, the additional peak near the origin disappears. This peak must, therefore, be interpreted as a result of the destruction by the field H of the moment beginning with $\varkappa = 6$, not $\varkappa = 4$. The experimental results of Refs. 2 and 6, which confirm the presence of this peak in K_2 (1.72) $X^1\Sigma_g^+$, are shown in the inset in Fig. 1(b), where we have $\Gamma_p = 10^6$ sec^{-1} for curve I and 2×10^6 sec^{-1} for curve II.

We are indebted to A. I. Okunevich for useful discussions.

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