

Alignment-orientation conversion by quadratic Zeeman effect: Analysis and observation for Te₂

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This paper reports the observation of the molecular fluorescence circularity under irradiation with linear polarized light. The phenomenon arises as a result of partial transformation from alignment to orientation of the ensemble of molecular angular momenta into orientation due to quadratic correction to Zeeman effect. Circularity rate up to 0.05 at magnetic field 0.4 T was registered in $B^3\Sigma_u^-(1_u^-) \rightarrow X^3\Sigma_g^-(1_g^-)$ fluorescence of ¹³⁰Te₂ molecules at angle $\pi/4$ with respect to \hat{E} vector of linear polarized exciting light. Quadratic magnetic energy terms are associated with magnetic field induced $\Delta J = \pm 1$ e/f mixing between $1_u^- \sim 1_u^+$ and $1_u^- \sim 0_u^+$ states. Circularity data fitting shows that the electronic part of Landé factor caused by $1_u^- \sim 0_u^+$ interaction is equal to $G_{\pm} \cong g_l \langle 0_u | \mathbf{J}_{a\pm} | 1_u \rangle + (g_s - g_l) \langle 0_u | \mathbf{S}_{\pm} | 1_u \rangle \cong 2.72$.

I. INTRODUCTION

In the preceding paper¹ a theoretical consideration was presented allowing to predict the appearance of orientation of angular momenta under excitation by broadband linear polarized light through the action of external perturbation causing asymmetric splitting between coherent $M, M \pm 1$ magnetic sublevels when the corresponding magnetic splitting frequencies $\omega_{MM \pm 1} \neq \omega_{-M \mp 1 - M}$. In this situation one is inclined to speak about anisotropic destruction of coherence, and the external field may play the same role as anisotropic collisions, which, as was first forecast in Refs. 2 and 3, are able to produce partial transformation from alignment to orientation. Significant interest in such a phenomenon of alignment-orientation conversion is connected with changing the symmetry type of angular momenta distribution, namely, with breaking the symmetry in respect to the reflection in the plane perpendicular to the axis of alignment. Nevertheless, as far as we know, there exist up to now only a few direct experimental observations, cf. Refs. 4–7, demonstrating the above mentioned effect of alignment-orientation transformation, which have been performed on atoms under anisotropic collisions. As is, in principle, clear,¹ any kind of external field action may cause the effect, if necessary asymmetric magnetic sublevel splitting is produced. In Ref. 8 it was proposed to achieve effective J -selective angular momenta orientation of aligned linear molecules moving in a beam with a fixed velocity by means of quadratic Stark effect.

An interesting possibility appears to be the use of the quadratic correction to the Zeeman effect in diatomic molecules. In particular, we direct now our attention on external magnetic field \mathcal{B} induced interaction between the levels with $\Delta J = \pm 1$, leading to quadratic Zeeman energy dependence on field strength. We have chosen the Te₂ molecule as a convenient object for analysis and experimental observation of the effect. Indeed, sufficiently enough data are known about the energetic and radiative properties of

low-lying excited electronic states of Te₂, among them the closely positioned $B0_u^+$ and 1_u^{\pm} states (cf. Refs. 9–13 and works cited therein). A number of data about g factors of these states have been obtained previously by measuring Hanle effect in laser induced fluorescence^{14–16} supposing that only a linear Zeeman effect is taking place. In fact, as recently established by deperturbation analysis in Ref. 13, the 1_u^{\pm} state mentioned above is, most likely, predominantly a $\Omega = 1$ component of the $B^3\Sigma_u^-$ state, and not of the $A^3\Pi_u$ state, as previously supposed,^{9,10} hence the notation $B^3\Sigma_u^-(1_u)$ will be used herein for this state. It is worth mentioning that the 1_u^- component can be considered, with fair approximation, as unperturbed by heterogeneous electron-rotation $\Delta J = 0$ interaction with 0_u^+ state. For this reason the 1_u^- component of Te₂ molecule was chosen here in order to demonstrate the appearance of alignment-orientation transformation induced by quadratic Zeeman effect.

II. ZEEMAN ENERGY CALCULATION

The Zeeman operator \mathcal{H}_{mag} for Hund's (c) coupling case can be written in the following form:¹⁷

$$\mathcal{H}_{\text{mag}} = -\mu_B \mathcal{B} [g_l \mathbf{J}_{az} + (g_s - g_l) \mathbf{S}]. \quad (1)$$

We suppose here that the space-fixed z axis of quantization is directed along the external magnetic field \mathcal{B} , $g_l = 1$ is the orbital electronic g factor, $g_s \cong 2.0023$ is the spin electronic g factor, $\mathbf{J}_a = \mathbf{L} + \mathbf{S}$ is an overall electronic orbital and electronic spin angular momentum of the molecule, μ_B is the Bohr magneton.

In contrast to intramolecular perturbations the Zeeman operator has nonzero off-diagonal ($\Delta J = \pm 1$) matrix elements,^{18,19} hence, a magnetic field is able to induce interaction between levels with different J in $0_u^+ \sim 1_u^{\pm}$ complex of $B^3\Sigma_u^-$ state. As follows from the rigorously valid selection rule, total parity \oplus or \ominus is conserved, cf. Fig. 1. In terms of e/f labeling,¹⁸ this means that allowed interactions with $\Delta J = \pm 1$ are $1_u^- \sim 1_u^+$ and $1_u^- \sim 0_u^+$, thus leading to magnetic field induced $e \sim f$ mixing. At the same time, the 1_u^- state can be considered as nonperturbed by

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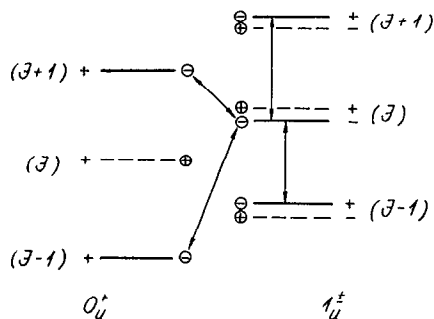


FIG. 1. Magnetic field induced mixing between 1_u^- and 1_u^+ , 0_u^+ state levels with $\Delta J = \pm 1$. For an even isotopic $^{130}\text{Te}_2$ molecule J possesses an even integer value and the levels shown with dashed lines are not realised due to symmetry considerations. \oplus, \ominus is the total parity, whilst $+, -$ is the electronic parity corresponding to e, f states, respectively.

both intramolecular electron rotation $1_u^- \sim 0_u^+$ as well as by diagonal over J ($\Delta J = 0$) magnetic $1_u^- \sim 1_u^+$, $1_u^- \sim 0_u^+$ interactions; it is the 1_u^- state which is of interest to study in order to analyze the role of quadratic Zeeman effect. Indeed, neglecting diamagnetic terms^{19,20} the quadratic over \mathcal{B} contribution ΔE_M to magnetic energy can be written for 1_u^- state as

$$\Delta E_M(1_u^-, v_1, J) = \frac{\langle 1_u^-(J) | \mathcal{H}_{\text{mag}} | 1_u^+(J \pm 1) \rangle^2}{E_{1_u^-}^{v_1 J} - E_{1_u^+}^{v_1 J \pm 1}} + \frac{\langle 1_u^-(J) | \mathcal{H}_{\text{mag}} | 0_u^+(J \pm 1) \rangle^2}{E_{1_u^-}^{v_1 J} - E_{0_u^+}^{v_0 J \pm 1}}. \quad (2)$$

Here v_0 and v_1 denote the vibrational levels of 0_u^+ and 1_u^- , 1_u^+ states, respectively.

We further neglect the heterogeneous $0_u^+ \sim 1_u^+$ interaction effect on rovibronic level energies of 0_u^+ and 1_u^+ states and their wave functions. Then, assuming that the electronic part of the Zeeman operator \mathcal{H}_{mag} is independent of internuclear distance, the total magnetic energy of a certain rovibronic level (v_1, J) takes the form

$$E_M(1_u^-, v_1, J) = \frac{G_z}{J(J+1)} \mu_B \mathcal{B} + G_z^2 A(M, J) \mu_B^2 \mathcal{B}^2 + G_\pm^2 B(M, J) \mu_B^2 \mathcal{B}^2, \quad (3)$$

where the first term corresponds to linear Zeeman effect. G_z is the electronic part of the molecular g factor in the $\Omega = 1$ state, which is equal to^{17,18}

$$G_z = [g\Omega + (g_s - g_l) \langle \Omega | S_z | \Omega \rangle] \Omega. \quad (4)$$

The second term of Eq. (3), in agreement with Eq. (2), is emerging due to $1_u^+ \sim 1_u^-$, $\Delta J = \pm 1$ interaction. The “geometric” factor $A(M, J)$ in Eq. (3) can be found by application of direct cosine matrix elements^{18,21} α_{11}

$$A(M, J) = \frac{\alpha_{11}^2(M, J; M, J+1)}{E_{1_u^-}^{v_1 J} - E_{1_u^+}^{v_1 J+1}} + \frac{\alpha_{11}^2(M, J; M, J-1)}{E_{1_u^-}^{v_1 J} - E_{1_u^+}^{v_1 J-1}},$$

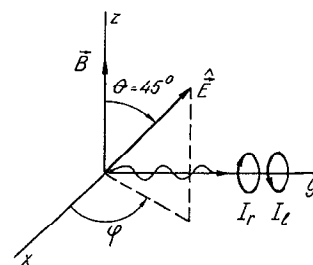


FIG. 2. Excitation and observation geometry.

$$\alpha_{11}^2(M, J; M, J+1) = J(J+2) f(J, M),$$

$$\alpha_{11}^2(M, J; M, J-1) = (J^2 - 1) h(J, M),$$

$$f(J, M) = \frac{1}{(J+1)^2} \left[\frac{(J+1)^2 - M^2}{4(J+1)^2 - 1} \right],$$

$$h(J, M) = \frac{1}{J^2} \left(\frac{J^2 - M^2}{4J^2 - 1} \right). \quad (5)$$

We are neglecting here the interaction between different vibronic 1_u^- state levels, supposing that the overlap integrals $\langle v_1^j | v_1^{j \pm 1} \rangle \cong 0$ for $v_1 \neq v_1$, where v_Ω , v_Ω are different 1_u^- state vibrational quantum numbers. Then, neglecting the centrifugal distortion terms in expressions for energies entering Eq. (5), we get

$$A(M, J) \cong \frac{1}{2B_v} \left[\frac{\alpha_{11}^2(M, J; M, J-1)}{J} - \frac{\alpha_{11}^2(M, J; M, J+1)}{J+1} \right]. \quad (6)$$

The second quadratic term in Eq. (3) describes the magnetic field induced $1_u^- \sim 0_u^+$, $\Delta J = \pm 1$ interaction, and G_\pm is the component of the electronic part of the g factor caused by heterogeneous ($\Omega' - \Omega = \Delta\Omega = 1$) interaction^{17,18}

$$G_\pm = g_l \langle \Omega | J_{a \pm} | \Omega' \rangle + (g_s - g_l) \langle \Omega | S_\pm | \Omega' \rangle. \quad (7)$$

The geometric factor $B(M, J)$ then appears as

$$B(M, J) = \alpha_{10}^2(M, J; M, J+1) S_1 + \alpha_{10}^2(M, J; M, J-1) S_2,$$

$$\alpha_{10}^2(M, J; M, J+1) = \frac{J(J+1)}{2} f(J, M),$$

$$\alpha_{10}^2(M, J; M, J-1) = \frac{J(J+1)}{2} h(J, M), \quad (8)$$

where

$$S_1 = \sum_{v_0} \frac{\langle v_1^j | v_0^{j+1} \rangle^2}{E_{1_u^-}^{v_1 J} - E_{0_u^+}^{v_0 J+1}}, \quad S_2 = \sum_{v_0} \frac{\langle v_1^j | v_0^{j-1} \rangle^2}{E_{1_u^-}^{v_1 J} - E_{0_u^+}^{v_0 J-1}}. \quad (9)$$

The direction cosine matrix elements²¹ α_{11} and α_{10} in Eqs. (6) and (8) are arising in the transformation from the molecule-fixed coordinate system to a space-fixed one. The overlap integrals and energy differences entering into Eq. (9) can be calculated for given v, J values using molecular constant set.^{9,10}

Thus the problem of getting Zeeman energy $E_M(1_u^-, v_1, J)$ values accounted for quadratic terms due to magnetic field induced interaction, cf. Eq. (3), is reduced to a minimum number of adjustable parameters, namely, G_z and G_{\pm} .

III. CALCULATION OF FLUORESCENCE CIRCULARITY

The general requirements imposed on the Zeeman sub-level splitting matrix $\omega_{MM'} = (E_M - E_{M'})/\hbar$ which are necessary to observe the occurrence of circularity under linear polarized excitation were examined in Ref. 1. According to

this, the nonzero even rank X coefficient ω_0^X value has to appear in $\omega_{MM'}$ expansion over irreducible tensor operators of the rotation group. This is equivalent to the condition

$$\omega_{MM+1} \neq \omega_{-M-1-M}, \quad (10)$$

which is fulfilled for the case described by Eq. (3) together with Eqs. (6) and (8) due to the M^2 dependence of α_{11} , α_{10} . By substitution the analytical expressions for the α_{11} and α_{10} values into Eqs. (6), (8) and then into Eq. (3), we get the explicit form for E_M

$$E_M(1_u^-, v_1, J) = \frac{G_z M \mu_B \mathcal{B}}{J(J+1)} + \frac{G_z^2 \mu_B^2 \mathcal{B}^2}{2B_v} \left[\frac{(J^2-1)(J^2-M^2)}{J^3(4J^2-1)} - \frac{J(J+2)[(J+1)^2-M^2]}{(J+1)^3[4(J+1)^2-1]} \right] \\ + \frac{G_{\pm}^2 \mu_B^2 \mathcal{B}^2}{2} \left[\frac{J[(J+1)^2-M^2]}{(J+1)[4(J+1)^2-1]} S_1 + \frac{(J+1)(J^2-M^2)}{J(4J^2-1)} S_2 \right]. \quad (11)$$

This leads to

$$\omega_{MM+1} = \omega_L [-1 - (a_J + b_J) \omega_L (2M+1)], \\ \omega_{-M-1-M} = \omega_L [-1 + (a_J + b_J) \omega_L (2M+1)], \quad (12)$$

where

$$\omega_L = \frac{G_z}{J(J+1)} \mu_B \frac{\mathcal{B}}{\hbar}, \quad (13)$$

and a_J and b_J are M -independent factors arising from $A(M, J)$ and $B(M, J)$, respectively. As is evident from Eqs. (11)–(13), condition (10) holds and hence the appearance of circularity $I_r - I_l$ is expected, I_r, I_l being fluorescence intensities in the cycle $J'' \rightarrow J' \rightarrow J'_1$ possessing right-handed and left-handed circular polarization, respectively. Let us discuss the geometry of excitation and observation, cf. Fig. 2. The \hat{E} -vector of linear polarized excitation is directed at spherical angles θ, φ , whilst the direction of observation is chosen along y axis. Expressions for $I_r - I_l$ and $I_r + I_l$ at arbitrary θ, φ are given in Ref. 1. The optimal θ value equal to $\pi/4$, will be supposed in further treatment, and we obtain

$$I_r - I_l \propto \sum_M \frac{\Gamma_p (\Gamma \sin \varphi + \omega_{MM+1} \cos \varphi)}{\Gamma^2 + \omega_{MM+1}^2} \\ \times (C_{J''M+11-1}^{J'M} C_{J''M+110}^{J'M+1} - C_{J''M10}^{J'M} C_{J''M11}^{J'M+1}) \\ \times (C_{J''M10}^{J'M} C_{J''M11}^{J'M+1} + C_{J''M+11-1}^{J'M} C_{J''M+110}^{J'M+1}). \quad (14)$$

where $C_{\alpha\beta\gamma}^{\alpha\beta\gamma}$ are Clebsch–Gordan coefficients, Γ is the excited state relaxation rate and Γ_p is the pumping (excitation) rate.

Figure 3 demonstrates the results of numeric calculation of the expected value of circularity rate $\mathcal{C} = (I_r - I_l)/(I_r + I_l)$ as dependent on a dimensionless parameter ω_L/Γ

$\Gamma \propto \mathcal{B}$, cf. Eq. (13). The $I_r + I_l$ value has been calculated according to Eq. (15) from Ref. 1. Fig. 3(a) demonstrates the case when $\varphi = \pi/2$ which is fulfilled when the exciting beam is perpendicular to the direction of observation, cf. Fig. 2. Figure 3(b) refer to the geometry when $\varphi = 0$, which can be achieved when fluorescence is observed along the exciting beam direction. Parameters involved in the calculation refer to the $B^3 \Sigma_u^-(1_u^-)$ state of $^{130}\text{Te}_2$ with $v_1 = 2 (J=96)$, as studied in Refs. 13, 14, and 16. The $J-1 \rightarrow J \rightarrow J+1$ cycle was considered. We used here $G_z = -1.86$ and $\Gamma = \tau^{-1} = 8.55 \times 10^6 \text{ s}^{-1}$ obtained from Hanle effect and lifetime measurements. Deperturbed molecular constants were taken from Refs. 13 and 22.

Curves 1 in Figs. 3(a) and 3(b) correspond to the situation when we suppose $G_{\pm} = 0$ in Eqs. (3) and (11) leading to $b_J = 0$ in Eq. (12). Thus we are neglecting the magnetic field induced $1_u^- \sim 0_u^+$ interaction. This assumption means that the G_z is the only parameter describing the magnetic energy, and the quadratic term arises due to $1_u^- \sim 1_u^+$, $\Delta J = \pm 1$ interaction. As may be seen, curve 1 in Fig. 3(a) is of dispersion type, being an odd ω_L/Γ function with a maximum at $\omega_L/\Gamma \sim 1.5$, and the appearing circularity does not exceed 0.1. The orientation signal $\mathcal{C}(\omega_L/\Gamma)$ in Fig. 3(b), cf. curve 1, is an even ω_L/Γ function and shows a more subtle form changing its sign in the vicinity of $\omega_L/\Gamma = 1$. The fact that curves in Fig. 3(a) are odd by reversing the magnetic field whilst those in Fig. 3(b) are even has a simple geometric interpretation. The orientation is always created perpendicular to the plane defined by the light \hat{E} vector and the direction of perturbation causing alignment-orientation conversion.^{1,8} In the case of Figs. 3(a) and 3(b) the created orientation is perpendicular to the \hat{E}, \mathcal{B} plane. The appeared orientation starts to precess in the magnetic field \mathcal{B} . For the geometry of Fig. 3(a) the orientation is created perpendicular to the direction of observation. Hence, the fluorescence circularity depends strictly on the direction of precession (defined by the sign

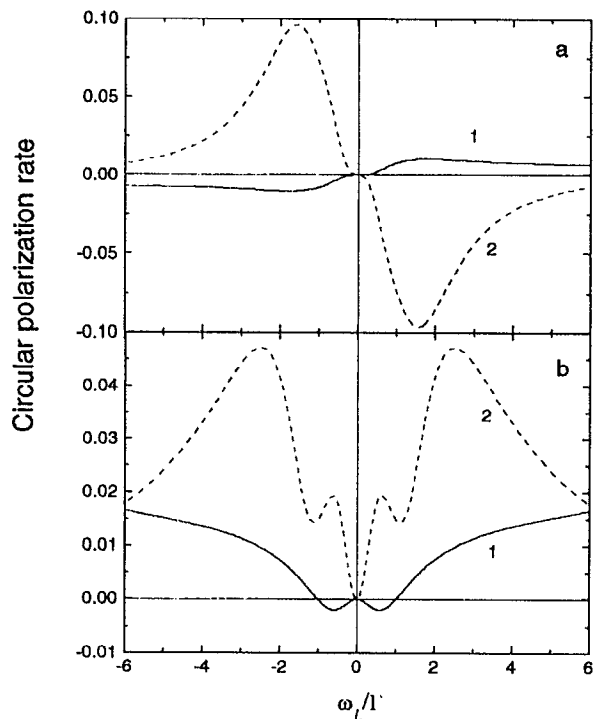


FIG. 3. Calculated circularity rate \mathcal{C} as dependent on parameter ω_L/Γ , which is proportional to magnetic field strength \mathcal{B} . (a) For E vector directed at $\theta=\pi/4$, $\varphi=\pi/2$ (cf. Fig. 2), (b) $\theta=\pi/4$, $\varphi=0$. Curves 1 correspond to $G_{\pm}=0$. Curves 2 correspond to $G_{\pm}=2.9$.

of magnetic field) and can be described by the odd type $\mathcal{C}(\mathcal{B})$ function. In the case of geometry of Fig. 3(b) the orientation is created in the direction of observation. It means that the observed fluorescence circularity is independent on the direction of precession of orientation and can be described by the even type $\mathcal{C}(\mathcal{B})$ function.

Let us now include magnetic field induced $1_u^- \sim 0_u^+$ mixing which follows the selection rule $\Delta J = \pm 1$. Using molecular constant sets¹³ for 1_u^- and 0_u^+ states we get for $v_1=2(J=96)$ the following values for the sums entering Eq. (11): $S_1 = -0.019$ 1/cm⁻¹, $S_2 = -0.029$ 1/cm⁻¹. If the G_z value is known, G_{\pm} remains the only adjustable parameter. Curves 2 in Fig. 3(a) and 3(b) demonstrate the total circularity signal which appears owing to both $1_u^- \sim 1_u^+$ and $1_u^- \sim 0_u^+$ Zeeman mixing. The concrete $G_{\pm} = 2.9$ value was used as established in Ref. 13 by “global” deperturbation analysis from simultaneous processing of magnetic and radiative data. As is seen, the circularity caused owing to $1_u^- \sim 0_u^+$ interaction dominates in this case. Circularity signals, as shown by curves 1 and 2 for the more simple dependence in Fig. 3(a), are of opposite sign due to the opposite sign of a_j and b_j in Eq. (12). Figure 4 demonstrates high sensitivity of the appeared orientation signal to G_{\pm} changes. Hence, it is promising to use the alignment-orientation conversion effect in order to determine G_{\pm} values and thus to pass to the matrix element of the heterogeneous electronic interaction.

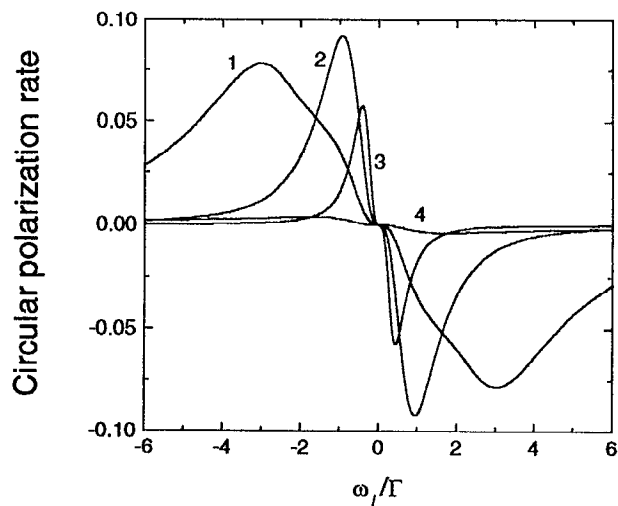


FIG. 4. The same dependence as shown by curve 2 in Fig. 3(a) for different G_{\pm} values: (1) $G_{\pm}=8$, (2) $G_{\pm}=4$, (3) $G_{\pm}=2$, (4) $G_{\pm}=1$.

IV. EXPERIMENTAL RESULTS AND DISCUSSION

Molecular fluorescence from ¹³⁰Te₂ vapor was induced by a linear polarized line 514.5 nm from an Ar⁺ laser operating in a multimode regime. The tellurium isotope was held in a fused silica cell at a temperature $T=650$ K and connected with a vacuum system through a dry valve. The R-type molecular transition ($X1_g^-, v''=4, J''=95$) \rightarrow ($B1_u^-, v'=2, J'=96$) was excited.⁹

The degree of circularity was measured at the geometry shown in Fig. 2 when $\theta=\pi/4$, $\varphi=\pi/2$. The external magnetic field was varied between $\mathcal{B} = -0.4$ and $+0.4$ T. The results for a fluorescence transition ($B1_u^-, v'=2, J'=96$) \rightarrow ($X1_g^-, v''=8, J''=97$) are presented in Fig. 5, showing the appearance of circularity up to $\mathcal{C} = 0.05$. The solid line was calculated in the same way as the curve 1 in Fig. 3(a) using G_{\pm} as the only nonlinear fitting parameter, and the value $G_{\pm} = 2.72$ yielding by fitting is in excellent agreement with the results obtained in a

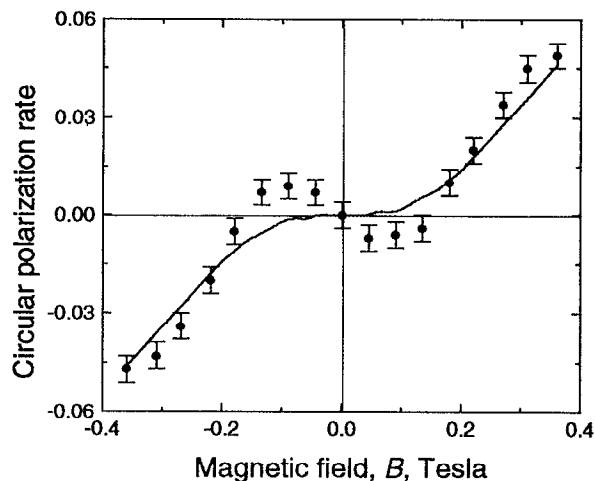


FIG. 5. Measured circularity signal for $v_1=2(J=96)$ level of $B^3\Sigma_u^-(1_u^-)$ state in ¹³⁰Te₂. Full line is obtained by fitting according to Eq. (17).

different way in Ref. 13. As it may be seen from Fig. 5, the general behavior of the calculated dependence reflects, to a major extent, the main behavior of the measured circularity values. It seems, however, that one can notice some distinction which is beyond the statistical error limit. The discrepancy in the small \mathcal{B} region can not be explained by accounting for the simultaneous $1_u^-, 0_u^+$, and 1_u^+ interaction, which gives an additional contribution to the quadratic Zeeman term in Eqs. (3) and (11) and is able to change only slightly the G_{\pm} value yielded by fitting, without changing the shape of the signal at a small magnetic field. The higher order magnetic terms not involved in our description are also unable to produce the structure. Thus, up to now we can only suppose that the discrepancy may be due to either some systematic experimental errors, or else, because the broad excitation line approximation is not valid with sufficient precision.

It is interesting to compare the information available from magnetic field induced alignment-orientation conversion with that yielded by conventional Landé factor measurements supposing the linear Zeeman effect conditions. Landé factors $g_{B0_u^+}$ of a diamagnetic ($\Omega=0$) $B0_u^+$ state of $^{130}\text{Te}_2$ were obtained in Ref. 15. The $g_{B0_u^+}$ values are determined by electron-rotation $0_u^+ \sim 1_u^+$ interaction with $\Delta J=0$. The $g_{B0_u^+}$ data interpretation needs, however, the notion of the electronic matrix element $\eta = \langle \Omega | J_{a\pm} | \Omega' \rangle$ for heterogeneous electron-rotation state mixing $|\Omega' - \Omega| = 1$ of the coupled 0_u^+ and 1_u^+ states. For weakly coupled levels we get^{13,15,22}

$$g_{B0_u^+} = 2G_{\pm} \eta \sum_{v_1'} \frac{\langle v_0 | B(R) | v_1' \rangle \langle v_0 | v_1' \rangle}{E_{0_u^+}^{v_0'} - E_{1_u^+}^{v_1'}}. \quad (15)$$

Hence, the $g_{B0_u^+}$ factor data yield only the product $G_{\pm} \eta$ and does not allow to determine G_{\pm} in a direct way. The η value for $0_u^+ \sim 1_u^+$ mixing in Te₂ was determined¹³ from energetic and radiative data as $\eta \cong 1.43$. This leads, for instance, to $G_{\pm} \cong 2.9$ for the $B0_u^+$ state level $v_0=0$ ($J=179$). This result is in a good agreement with the G_{\pm} values obtained in a direct and independent way in the present work. Such an agreement confirms once more that the G_{\pm} value is very close to 2η , and thus we have an additional reason for the conclusion¹³ that the Te₂(1_u) state studied here is in main a $^3\Sigma_1$ component of the $B^3\Sigma_u^-$ state. Indeed, as follows from Eqs. (7) and (15), G_{\pm} and η are not independent, and $G_{\pm} - \eta = \langle \Omega | S_{\pm} | \Omega' \rangle$. Passing to Hund's case (a) basis set, we get $G_{\pm} \cong g_s \langle ^3\Sigma_0 | S_{\pm} | ^3\Sigma_1 \rangle = 2\eta$. If the 1_u state is a $^3\Pi_1$ component of the $A^3\Pi_u$ state according to the point of view of the authors of Refs. 9 and 10, it should be $G_{\pm} \cong g_l \langle ^3\Sigma_0 | L_{\pm} | ^3\Pi_1 \rangle = \eta$.

It is worth mentioning that the alignment-orientation conversion induced by nonlinear Zeeman mixing gives the possibility to study the weak intramolecular interaction effects via magnetic characteristics of not only diamagnetic but paramagnetic states as well. For instance, the 1_u^- state of Te₂ studied here was always treated as unperturbed by $1_u^- \sim 0_u^+$ interaction and its magnetic properties were described by the G_z value only.^{13,14,16}

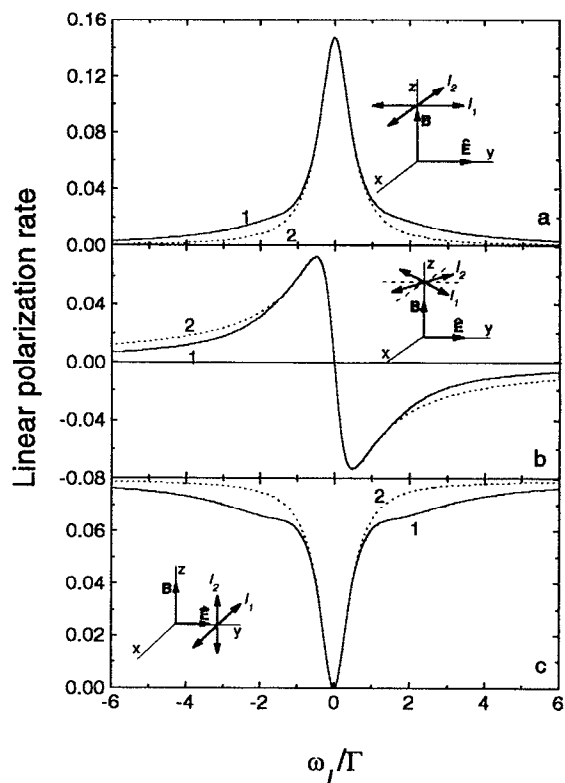


FIG. 6. Calculated linear polarization rate $\mathcal{P} = (I_1 - I_2)/(I_1 + I_2)$ as dependent on ω_L/Γ for various observation-excitation schemes. Curve 1—with accounting on quadratic Zeeman term in Eq. (11), curve 2—the ordinary Hanle effect signal.

Let us now follow the influence of quadratic Zeeman effect terms in Eqs. (3) and (11) on the Hanle effect signal measured in linear polarization of laser induced fluorescence. The results of calculations of linear polarization rate $(I_1 - I_2)/(I_1 + I_2)$ of fluorescence as dependent on magnetic field strength \mathcal{B} are presented in Fig. 6 for three different excitation-observation schemes for the same molecular transitions and parameters as in Fig. 5. As one may see, the dependencies accounting for nonlinear Zeeman term (curves 1) do not differ much from the signal caused by linear Zeeman effect (curves 2). The “traditional” Hanle signal of Lorentz shape, cf. Fig. 6(a), was the one used in the experiments^{14,16} in order to determine G_z for $v_1=2$ ($J=96$) of the $B^3\Sigma_u^-(1_u^-)$ state in $^{130}\text{Te}_2$. The uncertainty caused in this case by using the Lorentzian dependence does not considerably exceed the experimental error value. The most favorable is the situation in the case of dispersion shape signal, cf. Fig. 6(b), when one can determine G_z value from the positions of signal maxima without any influence of the quadratic Zeeman term. By contrast, the geometry presented in Fig. 6(c) leads to the largest changes due to the quadratic effect. Nevertheless, this geometry is very convenient technically and, besides, has its advantages allowing to measure both circularity rate and linear polarization rate in one and the same experimental scheme when the exciting light beam is directed orthogonally to both magnetic field and observation directions. One needs only to turn \mathbf{E} vector of exciting beam to obtain

$\theta = \pi/4$ (cf. Fig. 2) and to remove the $\lambda/4$ plate. Thus at least at certain parameters of the studied molecular state it is possible to determine G_z from the linear polarization rate signal and G_{\pm} by measuring the circularity appeared at linear polarized excitation as a result of magnetic e/f mixing.

V. CONCLUDING REMARKS

We demonstrated and analyzed the transformation of alignment of molecular angular momenta into their orientation under the influence of nonlinear Zeeman effect. This effect of breaking the reflection symmetry manifests itself in the occurrence of fluorescence circularity under linear polarized excitation. Owing to the presence of a linear term of Zeeman energy the dispersion shape magnetic field dependence of the circularity signal can be observed if fluorescence is viewed in the plane containing the exciting light \mathbf{E} vector and the magnetic field vector \mathcal{B} , the signal being zero in the case of pure quadratic external perturbation as in the case of Stark effect.¹ Sufficiently effective alignment-orientation conversion was registered in a 1_u^- component of $B^3\Sigma_u^-$ state of Te₂ caused predominantly by magnetic field induced $1_u^- \sim 0_u^+$ mixing with $\Delta J = \pm 1$. It is important to mention that since the linear Zeeman effect is not able to cause orientation, the registration of circularity at linear polarized excitation makes it possible to separate this effect from the own 1_u^- -state paramagnetism. Hence, the measurement of magnetic properties can be used to study intramolecular interaction not only in diamagnetic ($\Lambda = 0$ or $\Omega = 0$) states which appeared to be an extremely sensitive test of weak perturbations^{3,15,18,23,24} but can be applied also to paramagnetic states. In doing so, it is possible by combining the circularity measurement with the traditional (linear) Hanle effect measurement to determine both matrix elements G_z and G_{\pm} of the electronic Landé factor, in one and the same experiment. Generally speaking, the emergence of circularity may serve as a test of any kind of external perturbation causing asymmetric magnetic sublevel splitting, including hyperfine structure effects. Finally, since the quite noticeable degree of orientation can be maintained, alignment-orientation conversion may be considered as an additional possibility to achieve orientation of a molecular ensemble.

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