Oriented gas of diatomic molecules in a magnetic field

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We consider the loss of magnetization in an external transverse magnetic field of an ensemble of diatomic molecules optically oriented by "depopulation" in the gas phase. We use the polarization moment formalism and the asymptotic limit for large values of the angular moment. The superimposed signals from the ground and excited states are modelled numerically as functions of the degree of circularity of the fluorescence and the effect of polarization moments of different order is studied. We discuss the presence of an additional narrow peak due to polarization moments of order $\chi > 3$ being manifested through the octupole moment ($\chi = 3$) of the ground state. We consider an experimental geometry which gives a signal of the dispersive type, whose shape changes qualitatively when the signs of the magnetic moments of the ground and excited states change, and therefore can be used to determine these signs.

When circularly polarized resonant light interacts with atoms, a considerable optical orientation of the ground state often occurs thereby creating a magnetization of the atomic gas. For example, in orientation by "population" through fluorescence,[1] a significant fraction of the alkali atoms can be oriented. The method has been used in several applications, particularly in quantum magnetometers and gyroscopes.[2,3] The orientation is usually detected by changes in absorption or fluorescence, and also by the method of magnetic resonance. Because if the vibrational–rotational structure of the electronic transitions in diatomic molecules, another type of orientation is possible: through depopulation of the lower level (Fig. 1) by absorption of circularly polarized laser radiation.[4–6] In particular[5] for moderate absorption rates X → B in Na_2 for light with λ = 632.8 nm from a 30 mW helium–neon laser, an average degree of orientation $V = \langle M\rangle/\sqrt{J(J+1)} \approx 0.2$ was produced ($\langle M\rangle$ is the average value of the magnetic quantum number of the angular moment J) in the state ($v'' = 2$, $J'' = 45$) $X^1\Sigma_g^+$.

The orientation was detected by measuring the change in the intensity and degree of circularity of the fluorescence. This, however, is not always sufficient to determine the magnitude and sign of the magnetic moment acquired by the ensemble because the Landé factors in the diamagnetic ground state are unknown for the great majority of molecules. At the same time, the smallness of the Landé factor (10^{-4} to 10^{-6}) and the comparatively large orientation relaxation rate ($\sim 10^6$ sec^{-1}) make the method of magnetic resonance between the M-sublevels inapplicable because rf magnetic fields of very large amplitude are required. In this case a useful method of determining the magnetization of the ground state is to measure its decay in an external magnetic field (level crossing in the ground state).[7] A similar effect in the alignment of the ground state by linearly polarized light (the Hanle effect) was observed and studied[8–10] in Na_2, K_2, and Te_2; however, an overall magnetization is not created in this case.

The purpose of the present paper is to treat the phenomenon of optical orientation by circularly polarized light of the ground state of diatomic molecules with large angular moments $J \gg 1$ using the polarization moment formalism[7,11] and also to model numerically the crossing-level signals in an external magnetic field, which leads to a relaxation of the ensemble magnetization. The signals are discussed in terms of the conventionally observed quantity: the degree of circular polarization of fluoresence in the b → c transition (Fig. 1). The effect of the polarization moments of the ground state of different orders was studied, as well as their capability to induce additional structure in the signal. An experimental geometry is suggested in which the signal is of the dispersive type so that both the magnitude of the magnetic moment (provided that the relaxation rate is known) and its sign can be determined.

EQUATIONS OF MOTION OF THE POLARIZATION MOMENTS

In many cases it is convenient to transform from the density matrix to an expansion of the density matrix in terms of irreducible tensor operators.[7,11] The expansion coefficients (the polarization moments) contain all the necessary information for a system of weakly interacting particles. Equations for the polarization moments for the optical pumping conditions of Fig. 1 are known[12,13] for arbitrary values of J; however their solution is a difficult problem. There is a classical approach to the treatment of the orientation states of the angular moments,[14,15] however in this case the effect of separate polarization moments cannot be treated. These difficulties can be eliminated if we take the asymptotic limit of large J in the summation coefficients of the moments, thereby keeping the advantage of a quantum treatment.[16,17] The system of equations for the polarization moments of the excited f_Q^K and the ground $\varphi_q^\varkappa$ states in this case has the form

$$\dot{f}_Q^K = \Gamma_p\Big(\sum_{\varkappa q} {}^K_Q D_q^\varkappa \varphi_q^\varkappa - \sum_{K'Q'} {}^K_Q D_{Q'}^{K'} f_{Q'}^{K'}\Big) - (\Gamma_K - iQ\omega_b) f_Q^K, \tag{1a}$$

$$\dot{\varphi}_q^\varkappa = \Gamma_p\Big(\sum_{KQ} {}^\varkappa_q D_Q^K f_Q^K - \sum_{\varkappa' q'} {}^\varkappa_q D_{q'}^{\varkappa'} \varphi_{q'}^{\varkappa'}\Big) - (\gamma_\varkappa - iq\omega_a)\varphi_q^\varkappa$$
$$+ \Gamma_{J'J''} f_Q^K \delta_{K\varkappa}\delta_{Qq} + \lambda_q^\varkappa \delta_{\varkappa 0}\delta_{q0}. \tag{1b}$$

Here we let

$${}^\varkappa_q D_{q'}^{\varkappa'} = (-1)^\Delta \sqrt{\frac{2\varkappa'+1}{2\varkappa+1}} \sum_X \sqrt{2X+1}\, C_{1\Delta\,1-\Delta}^{X0} C_{X0\,\varkappa 0}^{\varkappa' 0} C_{X\,q-q'}^{\varkappa q} \Phi_{q-q'}^X, \tag{2}$$

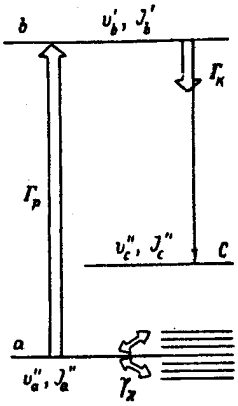

FIG. 1. Optical pumping scheme of diatomic molecules.

where the C^{ab}_{cdef} are the Clebsch–Gordan coefficients, $\Delta = J'_b - J''_a$ (Fig. 1) and the D'yakonov tensor[11] Φ^X_ξ is expressed in terms of the circular components e_{q_1} and e_{q_2} of the polarization vector e of the light

$$\Phi^X_\xi = \frac{1}{\sqrt{2X+1}} \sum_{q_1 q_2} (-1)^{q_1} e_{q_1} (e_{q_2})^* C^{X\xi}_{1q_1\, 1-q_2}. \tag{3}$$

The quantity Γ_p is the absorption rate $a \to b$ (Fig. 1), Γ_K, $\gamma_\varkappa$ are the decay rates of f^K_Q and $\varphi^\varkappa_q$, and the term $\lambda^\varkappa_q \delta_{\varkappa 0} \delta_{q0}$ is the number of repopulations in encounters with the v", J" levels of state a. The magnetic splitting frequencies are $\omega_{a,b} = g_{a,b}(\mu_0 H/\hbar)$, where the $g_{a,b}$ are the Landé factors, μ_0 is the Bohr magneton, and H is the magnitude of the external magnetic field. In (1a) and (1b), induced transitions are taken into account, as well as spontaneous transitions $b \to a$ with rate $\Gamma_{J'J''}$. In the normalization used here, polarization moments with K = 0 and $\varkappa = 0$ reduce to the densities.

The intensity I(e') of the fluorescence $b \to c$ with polarization e' can be written in terms of the f^K_Q as

$$I(e') \sim (-1)^{\Delta'} \sum_K \sqrt{2K+1}\, C^{K0}_{1-\Delta',\, 1\Delta'} \sum_Q (-1)^Q \Phi^K_{-Q}(e') f^K_Q, \tag{4}$$

where $\Delta' = J'_b - J'_c$.

Equations (1a) and (1b) were solved numerically, putting $\varphi^{\varkappa \geq 8} = 0$. It has been shown[17] that in this case the solution for the polarization moments $f^{K \leq 2}_Q$ contributing to I(e') converges rapidly.

LEVEL-CROSSING SIGNALS

The effect of an external magnetic field H on the ensemble orientation induced by circularly polarized light, when H is not parallel to the direction of the orienting beam, can be interpreted as destruction of coherence by the field,[7] which takes place because of the crossing of the magnetic sublevels when H = 0. Let the directions of excitation and observation be along the X axis (Fig. 2) and let H be perpendicular to the X axis. Equations (1) through (3) can be used to calculate the magnetic field dependence of the degree of circularity of the fluorescence in the cycle $a \to b \to c$ (Fig. 1)

$$C = (I_R - I_L)/(I_R + I_L), \tag{5}$$

where I_R and I_L are the fluorescence intensities polarized in the same sense as the exciting light (right-hand) and in the opposite sense (left-hand). The values I_R and I_L were calculated according to (1)-(3) for $R\uparrow$, $R\downarrow$ of the transition ($J'_b = J''_a + 1$, $J''_c = J''_a$). The dynamical constants were given by $\gamma_\varkappa = \gamma_0 = 3 \cdot 10^5$ sec^{-1}, $\Gamma_p/\gamma_0 = 100/3$, $\Gamma_K = \Gamma_0 = 10\,\Gamma_p$, and the ratio of the crossing signal widths $g_a/\gamma_\varkappa$ and g_b/Γ_K was set equal to 18. This value is typical for the $X \to B \to X$ and $X \to A \to X$ transitions in K_2, Te_2, Se_2, and others.[9,10] Under these conditions, induced transitions $b \to a$ are negligible, although they were formally taken into account. Spontaneous $b \to a$ transitions were neglected since their contribution is small because of the large number of allowed transitions from the state $v'_b J'_b$ into the different v", J" levels.

The signal shape is shown in Fig. 2, curve 1. The value C(H = 0) = 0.2802 corresponds to the orientation parameter V = −0.5545, which is determined by the moment of order $\varkappa = 1$. The increase of C with increasing H is caused by the effect of the level-crossing signal of the ground state a, i.e., the destruction of its polarization moments $\varphi^\varkappa_q$ [see (1b)] by the light field coupled with the polarization moments of the excited state f^K_Q, so that it becomes observable in the fluorescence. We note that there is an additional fine structure near zero, which will be discussed below. For large magnetic fields, level crossing in the excited state becomes important the destruction by the field of the polarization moments f^K_Q with increasing ω_b/Γ_K leads to a "disorientation" so that C decreases to zero. In the case of excitation by a very weak light source, there is no orientation of the ground state (more precisely $\varphi^\varkappa_q = 0$ for $\varkappa \neq 0$), and C varies from 5/7 to 0 (see Fig. 2, curve 2). In this case we have from (1) through (5)

$$I_R - I_L \sim \sqrt{2}\,\mathrm{Re}\, f^1_1 = \frac{1}{6} \frac{\Gamma_p \Gamma_1}{\Gamma_1^2 + \omega_b^2} \varphi^0_0, \tag{6}$$

$$I_R + I_L \sim \frac{1}{6}\left(4f^0_0 - f^2_0 + \sqrt{6}\,\mathrm{Re}\, f^2_2\right) = \frac{\Gamma_p}{360}\left(\frac{80}{\Gamma_0} + \frac{1}{\Gamma_2} + \frac{3\Gamma_2}{\Gamma_2^2 + 4\omega_b^2}\right)\varphi^0_0. \tag{7}$$

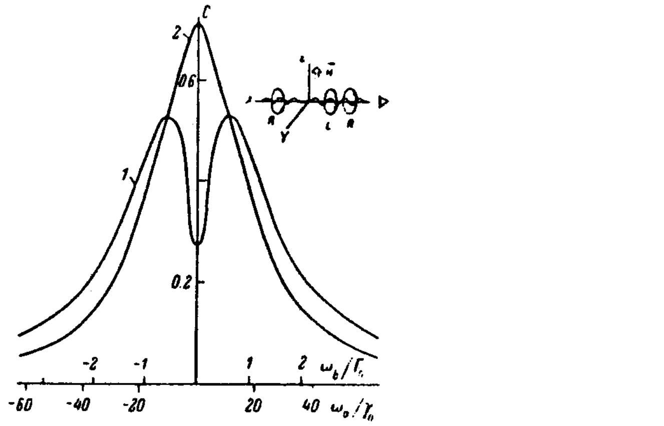

FIG. 2. Level-crossing signals with a circularly polarized excitation for the case of effective pumping $\Gamma_p/\gamma_0 = 100/3$ (curve 1) and in the limit of weak excitation $\Gamma_p/\gamma_0 \to 0$ (curve 2). In the upper right portion of the diagram, the directions of excitation, observation, and magnetic field are shown.

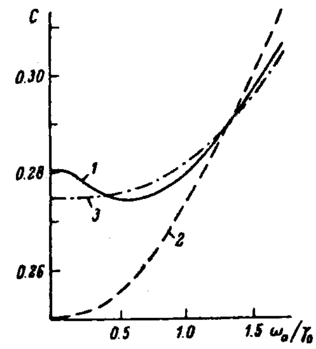

FIG. 3. Additional structure in the degree of circularity C for small values of ω_a/γ_0.

$$C = \frac{60\Gamma_1}{(\Gamma_1^2 + \omega_b^2)\left(80\Gamma_0^{-1} + \Gamma_2^{-1} + \frac{3\Gamma_2}{\Gamma_2^2 + 4\omega_b^2}\right)}. \tag{8}$$

EFFECT OF MOMENTS OF HIGHER ORDER

It is of interest to explain the origin of the additional structure occurring in level-crossing signals near H = 0 (Fig. 3, curve 1). One expects that this is caused by the effect of the polarization moments of the lower state of order higher than two. A similar additional narrow peak occurs in the classical model of the Hanle signal[14] for an optical pumping scheme of the type shown in Fig. 1 with $J \gg 1$. This narrow peak was erroneously thought to be due to the moment φ_q^4 in Ref. 14. A similar structure was detected experimentally[9] for the Hanle effect in K_2 and was explained[18] as the effect of the polarization moment of the ground state of order six φ_q^6.

For circularly polarized excitation, the manifestation of polarization moments of the ground state of odd order with $\varkappa > 1$ is possible, the first of these being $\varkappa = 3$. This is the octupole moment, which to the best of our knowledge has not been observed. In order to study the effect of the separate polarization moments $\varphi_q^\varkappa$ we (following Ref. 18) single out the moment of a certain rank $\varkappa$ by forcing it to relax with a rate $\gamma_\varkappa = 0.3 \cdot 10^9$ sec^{-1}, which is three orders of magnitude larger than $\gamma_\varkappa = \gamma_0 = 0.3 \cdot 10^6$ sec^{-1} for curve 1 of Fig. 3, which was calculated for the same conditions as curve 1 of Fig. 2 and differs only in scale. Let this moment be the octupole moment so that $\gamma_3 = 0.3 \cdot 10^9$ sec^{-1}, and let the other rates $\gamma_\varkappa$ in (1) for the $\varphi_q^\varkappa$ be equal to $0.3 \cdot 10^6$ sec^{-1}. Then the additional maximum at H = 0 vanishes (curve 2 of Fig. 3). A similar effect occurs if we isolate the effect of all moments of rank $\varkappa > 3$ (curve 3 of Fig. 3). This shows that the presence of an octupole moment φ_q^3 in the system is a necessary but not sufficient condition for the appearance of the additional structure. If we add the hexadecapole moment ($\varkappa = 4$), putting $\gamma_{\varkappa \le 4} = 0.3 \cdot 10^6$ sec^{-1}, $\gamma_{\varkappa > 4} = 0.3 \cdot 10^9$ sec^{-1}, then the additional peak again appears and its amplitude and shape are little changed from curve 1 of Fig. 3. If the moment with $\varkappa = 4$ is selected but the moments with $\varkappa = 5$ and $\varkappa = 6$ are not, the amplitude of the peak decreases by several times, and if only one of the moments with $\varkappa = 5$ or $\varkappa = 6$ is kept, the amplitude falls by two orders of magnitude.

From the above discussion, we see that the additional narrow peak in the signal $C(\omega_a/\gamma_0)$ (curve 1 of Fig. 3) is due to higher-order moments being manifested through the octupole moment ($\varkappa = 3$) of the ground state; of these higher-order moments the dominant contribution apparently comes from the moment of order $\varkappa = 4$. The moments can be observed separately using the techniques of nonlinear beat resonances.[13,18]

In Fig. 4 we show the results for the amplitude of the additional peak as a function of the ratio of dynamical constants in the system (1). From curve 1 of Fig. 4 we see that the amplitude of the effect ΔC rapidly decreases with decreasing values of the parameter Γ_p/γ_0, which characterizes the effectiveness of the pumping. For example, when Γ_p/γ_0 is decreased by a factor of three from the value 100/3 assumed for curve 1 of Fig. 2 and Fig. 3, the effect practically vanishes. Curve 2 of Fig. 4 shows the effect of the level-crossing signal of the upper state b, whose width is characterized by the ratio ω_b/Γ_0 (taking $\Gamma_K = \Gamma_0$). We see that a decrease of the ratio of the characteristic widths of the level-crossing signals of states a and b (equal to $\omega_a\Gamma_0/\omega_b\gamma_0$) significantly decreases the amplitude of the additional peak. The other parameters of curve 2 of Fig. 4 correspond to those for Fig. 2, curves 1 and Fig. 3, curve 1.

DETERMINATION OF THE SIGN OF THE MAGNETIC MOMENTS

Equation (1) can be used to calculate the signal for an arbitrary experimental geometry. The case where the directions of the circularly polarized excitation, the observation point, and the magnetic field are mutually orthogonal is particularly useful (Fig. 5). It corresponds in some degree of the case where the observation direction is at an angle of 45° for a linearly polarized excitation (Fig. 6), since this case also gives rise to a dependence of the signal on H in the form of an odd function, i.e., to a dispersion-type crossing signal sensitive to the sign of the magnetic moments.

Curve 1 of Fig. 5 shows the simultaneous effect of level-crossing signals of states a and b in the magnitude of the degree of circularity C', given as before by (4) and (5), but for the geometry of Fig. 5. Here we take $\Gamma_p/\gamma_0 = 10/3$, and the values of the other quantities are the same as for curve 1 of Fig. 2 and Fig. 3. The narrower dis-

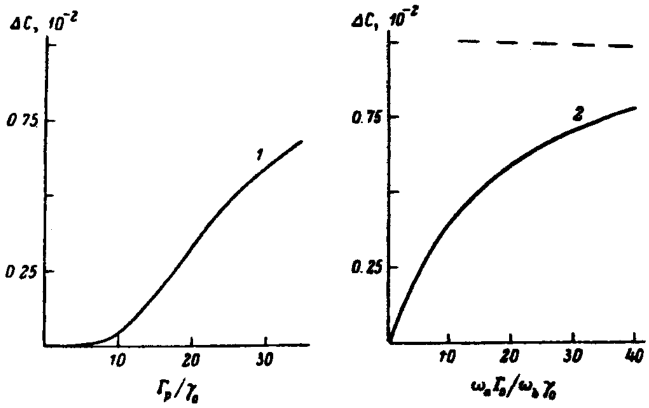

FIG. 4. Dependence of the amplitude ΔC of the additional peak on the pumping efficiency Γ_p/γ_0 (curve 1) and on the parameter $\omega_a\Gamma_0/\omega_b\gamma_0$ (curve 2). The dashed line shows the limiting value of ΔC in the absence of magnetism of the upper state b.

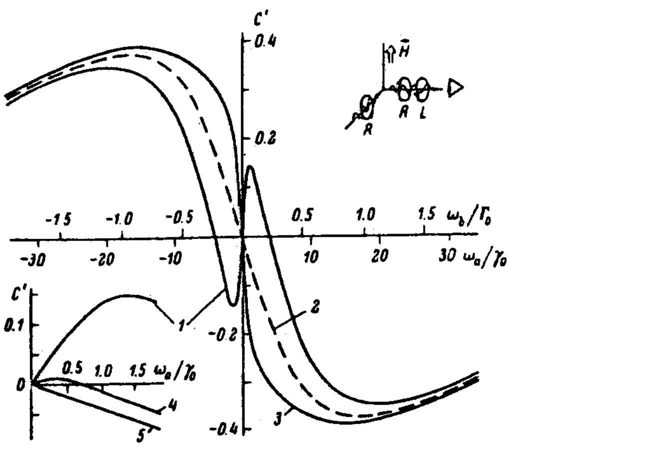

FIG. 5. Level-crossing signals for the case where the directions of the circularly polarized excitation, observation, and magnetic field are mutually orthogonal (as shown schematically in the upper right portion of the figure).

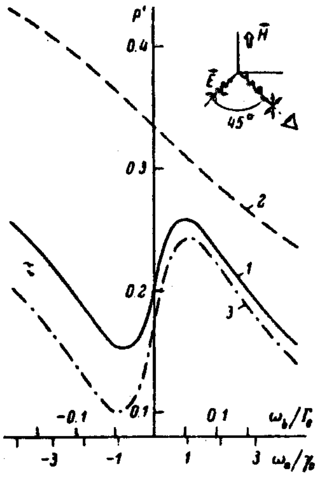

FIG. 6. Level-crossing signals for a linear polarized excitation and an observat on direction of 45° (shown schematically in the inset).

persion form (curve 1 of Fig. 5) corresponds to the level-crossing signal of the ground state, which vanishes in the limit of excitation by a weak light source $\Gamma_p/\gamma_0 \to 0$ (curve 2 of Fig. 5). In this limit the analytical expressions analogous to (6) through (8) have the form

$$I'_R - I'_L \sim \sqrt{2}\,\mathrm{Im}\, f^1_1 = \frac{1}{6}\frac{\Gamma_p\Gamma_1}{\Gamma_1^2+\omega_b^2}, \tag{9}$$

$$I'_R + I'_L \sim \frac{1}{6}\left(4f^0_0 - f^2_0 - \sqrt{6}\,\mathrm{Re}\, f^2_2\right)$$
$$= \frac{\Gamma_p}{3}\left[\frac{2}{3}\Gamma_0^{-1} + \frac{1}{120}\Gamma_2^{-1} - \frac{\Gamma_2}{40(\Gamma_2^2+4\omega_b^2)}\right], \tag{10}$$

$$C' = \frac{20\omega_b}{(\Gamma^2+\omega_b^2)\left(27\Gamma^{-1} - \frac{\Gamma}{\Gamma^2+4\omega_b^2}\right)}. \tag{11}$$

In (11) it was assumed that all $\Gamma_K = \Gamma$. The parameters of curve 3 differ from those of curve 1 (Fig. 5) only by a change in the sign of the Landé factor. In the case when g_a and g_b have different signs, the structure characteristic of the crossing signal of the ground state levels disappears. The significant difference between curves 1 and 3 of Fig. 5 means that we can at once establish a relation between the signs of g_a and g_b, and also the signs of each factor separately if the magnetic nature of one of the combining states is known. Finally, knowing the sign of the external field and the direction of circular polarization, we can determine the signs of g_a and g_b without the use of a priori information.

It is interesting that in the level-crossing signal of a state a the additional peak from the polarization moments $\varphi^\varkappa_q$ of higher rank does not show up for the geometry of Fig. 5, even for large values of Γ_p/γ_0. The lower left part of Fig. 5 (in expanded scale) shows to what degree the form of curve 1 is determined by the orientation of the ground state φ^1_q. Here curve 4 is obtained by discriminating only the orientation and putting $\gamma_1 = 10^4\gamma_{\varkappa\neq 1}$ and curve 5 is obtained by discriminating all moments of order $\varkappa \neq 0$ $(\gamma_{\varkappa\neq 0} = 10^4\gamma_0)$, i.e., as in curve 2 of Fig. 5, there is only the population density in the lower state.

In Fig. 6 we show the level-crossing curves (including the ground state) for the case where the observation direction makes a 45° angle with the direction of the linearly-polarized excitation, a case not considered previously by other authors. The degree of linear polarization $Q\uparrow$, $Q\downarrow$ of the transition $(J'_h = J''_a = J''_c)$, $P' = (I_2 - I_1)/(I_2 + I_1)$, where I_1 is the intensity of the fluorescence polarized along the magnetic field vector H and I_2 corresponds to the orthogonal direction. The parameters for curve 1 of Fig. 6 are the same as those for curve 1 of Fig. 5, i.e., the signs of g_a and g_b are the same. Curve 2 of Fig. 6 shows that the dispersion shape of the signal is destroyed by the field aligning the upper level b, since it was calculated in the limit $\Gamma_p/\gamma_0 \to 0$. Curve 3 of Fig. 6 was calculated for $\gamma_{\varkappa\geq 4} = 10^4\gamma_{\varkappa<4}$ and shows how the moments φ^4_q and φ^6_q effect the shape of the signal. We see that the shape is practically unchanged and only a shift of the curve is observed. If we discriminate only the sixth-order moment φ^6_q, then within the accuracy of the calculation, the points are the same as for curve 1. The results for large Γ_p/γ_0 show that in a dispersion-type signal the additional fine structure does not appear.

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