Polarization moments of higher rank in the fluorescence noise spectrum

L. I. Plimak and E. S. Polzik

G. V. Plekhanov Mining Institute, Leningrad

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The fluorescence noise spectrum is analyzed theoretically for atoms in a magnetic field which are excited from the ground state by intense polarized light. Experimental conditions are described under which the only effect of the excitation is to broaden the lines in the noise spectrum, although an oppositely polarized field of equal intensity would alter the noise spectrum in a very complicated way. Under these conditions the individual lines in the noise spectrum can be assigned to specific atomic polarization moments. The detailed fluorescence noise spectrum for alkali metals is calculated numerically, and it is shown that the separate contributions from the octupole and hexadecapole atomic moments can be identified experimentally.

INTRODUCTION

The fluorescence spectra of atoms in a magnetic field typically contain lines at multiples of the Zeeman frequencies.[1] Although the mechanisms by which these lines form can be very diverse,[2–4] in all cases quantum beating between the Zeeman sublevels is involved. In other words, these lines correspond to the free precession frequencies for the atomic polarization moments $\rho_q^\varkappa$. For example, the line structure recorded in Ref. 2 was caused by orientation relaxation in the resonant levels ($\varkappa = 2$), while in Ref. 3 damping of the atomic orientation in the ground state ($\varkappa = 1$) was responsible.

In this paper we discuss how atomic polarization moments of high rank ($\varkappa = 3, 4$) might be observed for atoms in the ground state; these moments are difficult to identify by conventional methods.[5] Although the possibility of observing the higher-rank moments was mentioned in Ref. 4, where fluorescence noise spectra from atomic vapor was studied, estimates indicate that it will not be feasible to observe the higher-order polarization moments when the latter precess freely, i.e., when the system is only weakly excited, as assumed in Ref. 4. Intense light sources will therefore be required.

The nearly perfect spherical symmetry of the atoms simplifies the calculations for weakly excited systems; however, the atoms become distorted when strongly excited by polarized light (in particular, optical pumping occurs). The field-induced distortion "mixes up" the individual polarization moments, and their contributions to the noise spectrum can be separated only with great difficulty. However, we have found that by suitably choosing the polarization of the excitation light, one can ensure that the individual moments $\rho_q^\varkappa$ remain separated (independent) at field intensities which would cause complete overlapping for light of the opposite polarization. In terms of its effects on the atomic coherence dynamics, light with this polarization is equivalent to spherically symmetric irradiation, and we will refer to this type of excitation as nondeforming. It occurs in its simplest form for spherically symmetric irradiation; however, this configuration is unsuitable for our purposes, because in this case the noise spectrum contains no moments of rank higher than 2. We will therefore be interested in examples of nonspherical, nondeforming excitation, e.g., in excitation by polarized light. This would make it possible to observe moments with $\varkappa > 2$ in the noise spectrum while at the same time permitting the noise spectrum to be calculated analytically.

For nondeforming excitation, the fluorescence noise spectrum consists of individual lines (polarization resonances). If the experimental configuration is suitable, these resonances may be attributed to the relaxation of certain $\rho_q^\varkappa$ in one of the hyperfine sublevels of the ground state, and the only effect of strong excitation is to broaden these resonances. We stress that this simple behavior occurs only for nondeforming excitation—a field of equal intensity but opposite polarization would change the noise spectrum in a much more complicated way. For nondeforming excitation the calculations proceed in two steps—one first determines the (field) widths of the resonances and then calculates their intensities. We have derived general formulas for the line widths and intensities for arbitrary moments $\rho_q^\varkappa$ and calculated the noise spectrum numerically in detail for the alkali metals. Among other things, the results demonstrate that excitation by an intense light field should in fact permit the observation of the octupole and hexadecapole atomic moments.

1. STATEMENT OF THE PROBLEM

We consider an experiment in which a collisionless atomic beam in a magnetic field is excited by a transverse polarized light ray; the excitation occurs from a hyperfine sublevel of the ground state with moment F_0 to a hyperfine sublevel of an excited state with moment F. The equipment is designed to select the fluorescence at the wavelength corresponding to this transition (there are no cascade transitions). The exciting light is assumed strong enough to excite an atom to the upper level several times while the atom crosses the light ray, and the lifetime of the atom in the upper level is assumed to be negligibly short. In other words, we assume that the relation

$$\gamma_0 \lessdot \Gamma \ll \gamma \tag{1}$$

holds; here γ_0 is the inverse of the time required to cross the ray, Γ is the probability that the field will excite an atom per

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unit time, and γ is the inverse lifetime in the excited state. The fluoresence will then remain linear even if $\gamma_0 \ll \Gamma$, i.e., we may neglect induced transitions in favor of spontaneous ones. For simplicity we also assume that the Zeeman frequecies Ω_0 and Ω of the two sublevels are small compared to γ: $\Omega_0, \Omega \ll \gamma$. The statistical properties of the irradiation must be chosen to ensure that the excess noise in the frequency band of interest is much less than the shot noise, as otherwise the extraneous noise in the excitation beam would obscure the fluorescence noise.

We consider the above beam experiment only because we want to discuss the excitation of a specific hyperfine component of the spectral line. For elements lacking a hyperfine structure, our calculations are also valid for the experimental situation when metal vapors are used.

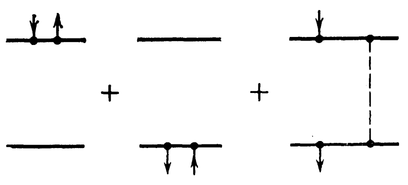
2. NONDEFORMING EXCITATION

When condition (1) holds, the polarization moments of the hyperfine sublevel of the ground state from which the transition occurs obey a closed equation of the form

$$\dot\rho_q^\varkappa = -(i\Omega_0 q+\gamma_0)\rho_q^\varkappa + \sum_{\varkappa' q'} D_{\varkappa q}^{\varkappa' q'} \rho_{q'}^{\varkappa'}. \qquad (2)$$

This equation is valid if there is no collisional transport of coherence between the hyperfine sublevels of the ground state. In terms of Perel'-Konstantinov diagrams,[6]

$$D_{\varkappa q}^{\varkappa' q'} =$$



$. \qquad (3)$

Here the heavy lines are the atomic propagators, while the dashed line is a photon propagator; the incoming and outgoing arrows correspond to the analytic signal of the exciting light and to its complex conjugate. The statistical averaging in (3) can be carried out directly if the excitation contains no correlations with times comparable to γ_0^{-1} and Ω_0^{-1}. The pumping light will therefore be assumed to be either monochromatic or broad-band gaussian in what follows. In the latter case, the spectral width need only be large compared with γ_0 and Ω_0 (it does not have to be large compared to γ).

Because the exciting light deforms the atoms, the kernel $D_{\varkappa q}^{\varkappa' q'}$ in (3) becomes nondiagonal in $\varkappa$ and q. The problem consists in finding conditions under which no deformation occurs and (3) is diagonalized.

Diagonalization with respect to q follows automatically if we assume that

$$\Omega_0 \gg \Gamma. \qquad (4)$$

The problem is then physically equivalent to a system of weakly coupled oscillators with different natural frequencies $q\Omega_0$. In this case, oscillations of one oscillator do not contribute significantly to oscillations in the other, and the coupling can be neglected.

Diagonalization with respect to $\varkappa$ can be achieved if the polarization of the exciting light is suitably chosen. One may argue as follows. The kernel (3) is certainly diagonal for the case of unpolarized, spherically symmetric excitation. Since condition (4) suffices to make $D_{\varkappa q}^{\varkappa' q'}$ diagonal in q, this suggests that strict spherical symmetry may not be necessary for complete diagonalization. In order to derive an explicit condition on the polarization, we examine how (3) becomes diagonalized for spherically symmetric irradiation. The quantity $D_{\varkappa q}^{q'}$ is a linear combination of the components $\langle E_\alpha E_\beta^* \rangle$ of the polarization tensor of the exciting light, and the selection rules for the magnetic quantum number imply that q-$q' = \alpha - \beta$. Because the off-diagonal components of the polarization tensor vanish for spherically symmetric excitation, (3) is diagonal in q. The quantities $D_{\varkappa q}^{\varkappa' q'}$ depend only on the diagonal components of the polarization tensor and become diagonalized with respect to $\varkappa$ because

$$\langle |E_1|^2 \rangle = \langle |E_0|^2 \rangle = \langle |E_{-1}|^2 \rangle = {}^1\!/_3 \langle |\mathbf{E}|^2 \rangle \qquad (5)$$

for spherically symmetric excitation. Condition (5) is the desired constraint on the polarization; indeed, (4) then ensures that the kernel (3) is diagonal in q independently of the off-diagonal components of the polarization tensor, which may therefore be arbitrary.

Condition (5) does not require that the irradiation be spherically symmetric, and it holds for several other pure and mixed polarizations. As simple examples we may mention linearly polarized light whose polarization vector makes an angle arccos $(1/\sqrt{3}) \approx 57°$ with the magnetic field vector,[1)] or unpolarized light with wave vector **k** making the same angle with **H**. There are no circular polarizations that satisfy (5) for all directions of irradiation, and apart from the linear polarization noted above, all other pure polarizations of type (5) are elliptical. We stress that the polarization condition (5) alone does not suffice for nondeforming excitation–condition (4) is also required.

The diagram in (3) can be calculated directly for nondeforming excitation; the result is

$$D_{\varkappa q}^{\varkappa' q} = -\Gamma_\varkappa \delta_{\varkappa\varkappa'}, \qquad \Gamma_\varkappa = \Gamma(1-\varepsilon r_\varkappa), \qquad (6)$$

where

$$\Gamma = \frac{4\pi\gamma \langle F_0 \| d \| F \rangle^2}{3(2F_0+1)} \int dk\, I_k \left[\frac{1}{4}\gamma^2 + (k-k_0)^2 \right]^{-1}$$

is the atomic excitation probability per unit time for spherically symmetric light with the same spectral intensity I_k as for the nondeforming excitation radiation actually used; k_0 is the transition frequency. There is a wide range of excitation frequencies for which Γ is given by the standard formula for the induced transition probability.[8] The factor ε characterizes the role of the other lower hyperfine sublevels–it gives the relative probability for deexcitation via the "observed channel,"[8]

$$\varepsilon = \gamma^{-1}\gamma_{F\to F_0} = (2j+1)(2F_0+1) \begin{Bmatrix} F_0 & F & 1 \\ j & j_0 & I \end{Bmatrix}^2,$$

where j_0 and j are the electron moments of the levels and I is the nuclear moment. We also have

$$r_\varkappa = \left[1 - \frac{\varkappa(\varkappa+1)}{2F(F+1)} \right]^2, \qquad F_0 = F,$$

$$r_\varkappa = \frac{(4f^2-\varkappa^2)[4f^2-(\varkappa+1)^2]}{4f^2(4f^2-1)}, \qquad F_0 = F \pm 1,$$

where $2j = F_0 + F + 1$.

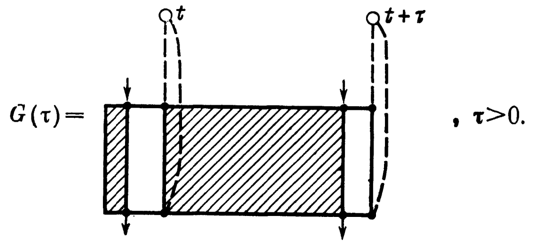
For atoms pumped by nondeforming light, the only effect of the excitation is thus to modify the relaxation constants for the ground state: $\gamma \to \gamma^{\varkappa} = \gamma_0 + \Gamma_{\varkappa}$. There is no frequency shift, because Γ is real regardless of the mismatch from resonance. It is possible to have $\Gamma_{\varkappa} \ll \Gamma$ for elements without a hyperfine structure; in all other cases, $\Gamma_{\varkappa}$ is close to Γ.

3. POLARIZATION RESONANCES IN THE NOISE SPECTRUM

In terms of diagrams, quantum electrodynamics leads to the expression

$$G_{\omega} = \int d\tau \exp(i\omega\tau) G(\tau) \tag{7}$$

for the fluorescence noise spectrum when (1) is satisfied; here G is given by the diagram



The hatched rectangles indicate the parts of the diagram which require an exact treatment of the effects of the intense field. The right- and left-hand rectangles correspond to the Green's function for Eq. (2) and to the stationary density matrix for the ground-state atoms, respectively.

The Green's function completely determines the time dependence in (7) when (1) holds. When (4) holds, relaxation eigenfrequencies $q\Omega_0$ are present which determine the time dependence $G(\tau)$. The corresponding noise spectrum then consists of individual lines at the frequencies $q\Omega_0$ with $q \leqslant 4$ [when (4) is satisfied, the stationary density matrix contains only components with $q = 0$, and even if reradiation occurs, $|\Delta q| \leqslant 4$]. The relaxation moments $\rho_q^{\varkappa}$ with $q \leqslant \varkappa \leqslant 2F_0$ contribute to each of these lines. When (4) and (5) hold, $G(\tau)$ is a sum of terms of the form $\exp(-\gamma^{\varkappa} + i\Omega_0 q)\tau$; each term corresponds to a line at frequency $q\Omega_0$ of width $\gamma^{\varkappa}$ in the noise spectrum (the detailed line shape depends on the motion of the atoms; our calculations give a Lorentzian because we introduce the constant γ_0 to treat the motion phenomenologically. Since the stationary density matrix is now spherically symmetric (just as for a weak field), the only difference in the noise spectrum (7) compared with the weak-field case is the field-induced broadening of the lines: $\gamma^{\varkappa} = \gamma_0 + \Gamma_{\varkappa}$ (all the lines have the same width γ_0 in a weak field). In particular, as in the case of a weak field the noise spectrum cannot contain any moments with $\varkappa > 4$ for a nondeforming pump.

Thus, for nondeforming excitation the noise spectrum remains the same as for a weak field, except that the widths of the polarization resonances are changed as described by the constant $\gamma^{\varkappa}$. The spectrum consists of five peaks at the frequecies $q\Omega_0$, $q = 0, 1, 2, 3, 4$; each peak is formed by the superposition of $5 - q$ elementary lines of characteristic widths $\gamma^{\varkappa}$. The peak at $q\Omega_0$ is composed of lines corresponding to moments $\varkappa \geqslant q$, i.e., the dynamics of the hexadecapole moment show up at the frequency $4\Omega_0$, while the peak at $3\Omega_0$ reflects the combined dynamics of the hexadecapole and octupole moments, etc.

The quantity $|G_{\omega}|^2$ (7) can be conveniently characterized by dividing it by the "white" shot component of the noise in the photorecorders.[9] For the $q\Omega_0$ line, a calculation gives

$$\eta_q = \sum_{\varkappa} \eta_{\varkappa q} = \sum_{\varkappa} \alpha\varepsilon\beta \frac{\Phi}{4\pi} \frac{\Gamma}{\gamma^{\varkappa}} S^{\varkappa q}, \tag{8}$$

where β is the quantum yield of the photoreceiver, Φ is the solid angle from which the fluorescence is collected, and the dimensionless parameters α and $S^{\varkappa q}$ are defined in the Appendix (since $S^{00} = 1$, $S^{\varkappa q}$ gives the relative intensity of the lines).

We now compare (8) with the calculations in Ref. 4, where the noise spectrum was found to lowest nonvanishing order in Γ/γ_0 without treating the hyperfine structure–instead of the single constant γ_0 introduced above, a set of constants $\tilde{\gamma}^{\varkappa}$ was introduced to treat the collisions phenomenologically. Since weak excitation is clearly nondeforming, (8) should agree with the result in Ref. 4 for $I = 0$ and $\Gamma/\gamma_0 \ll 1$ if we replace $\tilde{\gamma}^{\varkappa}$ by γ_0, and this is readily verified (cf. the Appendix). A tenfold increase in the signal/noise ratio can thus be achieved by using intense nondeforming excitation (in the latter case $\Gamma/\gamma^{\varkappa} \sim 1$, while in a weak field $\Gamma/\gamma^{\varkappa} \ll 1$). We observe also that since the quantities $S^{\varkappa q}$ depend only on F_0 and F, the relative noise line intensities are independent of field strength and are the same whether or not a hyperfine structure is present.

4. OBSERVING THE OCTUPOLE AND HEXADECAPOLE MOMENTS

If the amount of extraneous noise in the pumping light is small, the signal/noise ratio for observing the structure (7) is given by $\eta(\Delta\omega T)^{1/2}$, where η is the signal amplitude divided by the background, $\Delta\omega$ is the frequency band in which the observations are made ($\Delta\omega \lesssim \Gamma$), and T is the time during which the signal is stored.[10] If we take $\Delta\omega = 10^5\ \mathrm{s}^{-1}$ (Ref. 3) and $T = 10^3$ s, we find that $\eta \sim 10^{-4}$ at the detection threshold, at which the signal/noise ratio is equal to 1. The actual value of η_q depends on the states between which the transition occurs (on the parameters α, ε, and $S^{\varkappa q}$), on the experimental configuration (α, $S^{\varkappa q}$, Φ), and on the quantum yield β of the receiver.

To increase the solid angle Φ over which the fluorescence is recorded it is advantageous to observe the system using linearly polarized light, which makes it possible to collect radiation from the "equatorial strip" normal to the polarization vector. However, the parity properties of $S^{\varkappa q}$ (see the Appendix) imply that the noise spectrum contains no odd-order moments if the system is irradiated by polarized light (as assumed in our specific calculations) and then observed using linearly polarized light. We therefore assume that circularly polarized light is used to observe the octupole moment ($\varkappa = 3$) and set $\Phi/4\pi = 0.01$. Linearly polarized

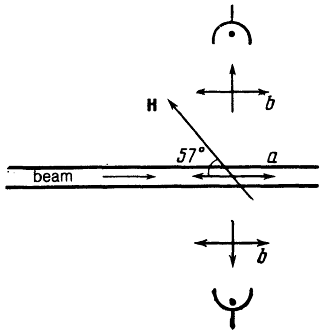


FIG. 1. Experimental configuration for observing the hexadecapole moment: a) polarization of the irradiating light; b) polarization of the observation light. In this case the two polarizations are the same, and the direction of irradiation is normal to the beam but otherwise arbitrary. The fluorescence signal is collected from a strip in the plane perpendicular to the beam.

light can be employed to observe the hexadecapole moment ($\varkappa = 4$), and we will assume that $\Phi/4\pi = 0.1$ in this case.

The numerical calculations were carried out for the configurations shown in Fig. 1 ($\varkappa = 4$) and Fig. 2 ($\varkappa = 3$). The results showed that the quantities S^{3q} and S^{4q} of interest are small for hyperfine transitions for which $F_0 = F \pm 1$, and we will henceforth consider only the case $F_0 = F$. This situation occurs, e.g., in the hyperfine structure of the fundamental transition for the alkali metals, to which our treatment will be confined. The electron moments for the upper and lower levels are $j_0 = j = 1/2$, and we have

$$\varepsilon = {}^1/_3\left(1 \pm \frac{1}{I + {}^1/_2}\right)$$

for $F_0 = I \pm 1/2$, respectively. We therefore choose the hyperfine transition for which $F_0 = F = I + 1/2$. However, $\varepsilon \sim 1/3$ when $I > 1$, and we may neglect the dependence of η_q on the nuclear moment (i.e., the signal will be roughly the same for the $F_0 = F = 3$ transition in Rb^{85} and Cs^{133}, say). Table I lists the calculated relative intensities of the $S^{\varkappa q}$ lines for Na^{23}, Rb^{85}, and Cs^{133} for $F_0 = F = 2$, 3, and 4, respectively; the values η_{00} determining the absolute line intensities are also given. The quantum yield of the receiver was assumed equal to 0.1.

The hexadecapole can be observed most clearly in Cs^{133}, because its contribution is greatest at the frequency $3\Omega_0$ and there is no other contribution from the other orders $\varkappa$. The signal/background ratio η_3 for this line is $\approx 3\cdot 10^{-4}$, which corresponds to a signal/noise ratio of ~ 3 under the conditions indicated above. The nuclear alignment signal at Ω_0 and $2\Omega_0$ is an order of magnitude more intense and may be helpful initially in locating the $3\Omega_0$ line.

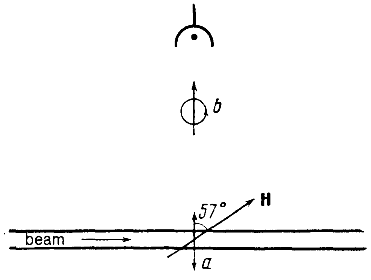


FIG. 2. Experimental configuration for observing the octupole moment: a) irradiation polarization; b) observation polarization. The direction of irradiation is normal to the plane of the figure.

The octupole moment is harder to observe; in this case it is advisable to use a beam of Na^{23} with $F_0 = F = 2$ in the experiment. There is no frequency at which a pure octupole contribution appears. Although the octupole contribution is greatest at the frequency $2\Omega_0$, it is only twice as intense as the alignment signal at the same frequency. A single/noise ratio of ~ 3 can be achieved by recording the fluorescence signal for ≈ 3 h. This signal is negative, which according to Ref. 4 reflects the corpuscular properties of light. Another possibility would be to record the octupole contribution at $3\Omega_0$ in the same experiment; at this frequency, the octupole contribution is solely responsible for the dip in the spectrum.

CONCLUSIONS

Our calculations show that nondeforming pumping is useful for studying unexcited materials in a magnetic field by noise spectroscopy. This makes it possible to see how a strong field affects the experimental results in terms of simple parameters such as the line widths. The advantages of nondeforming excitation should also be evident whenever there is no need to measure the coherence of a single hyperfine sublevel, as in the case, e.g., when analyzing fluoresence from atomic vapor. Even if no collisions occur, Doppler line broadening makes it impossible to select a unique hyperfine transition (such selection was assumed in the above calculations). This difficulty may also arise if the source spectrum is not narrow enough at the transition frequency.

However, the computational modifications needed to accommodate this difficulty are straightforward. For nondeforming excitation, the system of equations for the coherence factors of the ground-state levels splits into independent groups, each containing $2I + 1$ equations ($2j_0 + 1$ if $I > j_0$). (Coherence between different hyperfine sublevels can be ignored if the hyperfine splitting is large compared to the Zeeman splitting.) For the alkali metals, this gives two coupled equations for each value of $\varkappa$ and q, and the coherence dynamics problem can be solved analytically as before. The line intensities can be calculated from the general diagram expression (7).

We are grateful to Yu. M. Golubev for a discussion of these results and for advice concerning the exposition.

APPENDIX

Polarization dependence of the fluorescence noise spectrum

For the experimental configurations considered above, the fluorescence noise spectrum is of the form

$$G_\omega \infty \sum_{\varkappa q} S^{\varkappa q} L_\varkappa(\omega - q\Omega_0),$$

where $|q| \leqslant \varkappa \leqslant 4$ and the $L_\varkappa$ are symmetric lineshapes normalized to unit area (their detailed form depends on how the atoms move). The $S^{\varkappa q}$ can be expressed in terms of the vector addition coefficients[11] as

TABLE I.

Element	Exp. config. (Figs. 1, 2)	η_{00}	S^{10}	S^{20}	S^{30}	S^{40}	S^{11}	S^{21}	S^{31}	S^{41}	S^{22}	S^{32}	S^{42}	S^{33}	S^{43}	S^{44}
Na^{23},	1	0.017	0	0	0	0.005	0	0.40	0	0,001	0,20	0	0,004	0	0.008	0.002
$F_0=F=2$	2	0.0007	0.067	0	−0.047	0.009	0.067	−0.078	−0.018	0.002	−0.039	−0.089	0.008	−0.030	0.013	0.003
Rb^{85},	1	0.016	0	0	0	0.013	0	0.49	0	0.002	0,23	0	0.010	0	0,018	0.004
$F_0=F=3$	2	0,0006	0.047	0	−0.034	0.025	0.047	−0.014	−0,013	0.005	−0,007	−0,064	0.020	−0.021	0,035	0.009
Cs^{133},	1	0.015	0	0	0	0.016	0	0.50	0	0,003	0,25	0	0.013	0	0,023	0.006
$F_0=F=4$	2	0.0005	0.032	0	−0.024	0.033	0,032	0,008	−0,009	0,007	0.004	−0,044	0,027	−0.015	0,047	0.012

$$S^{\varkappa q}=\mathrm{Re}\, Z^{\varkappa q}(\mathbf{e}_0, \mathbf{e})\,[Z^{\varkappa q}(\mathbf{e}, \mathbf{e}_0)]^*, \qquad Z^{\varkappa q}=\zeta^{\varkappa q}/\zeta,$$

$$\zeta^{\varkappa q}=(-1)^{\varkappa+F_0+F+1}(2F_0+1)^{1/2}\sum_{\varkappa'\varkappa''}[(2\varkappa'+1)(2\varkappa''+1)]^{1/2}\cdot$$

$$\times\begin{Bmatrix} 1 & 1 & \varkappa'' \\ F_0 & F_0 & F \end{Bmatrix}\begin{Bmatrix} \varkappa & \varkappa' & \varkappa'' \\ F_0 & 1 & F \\ F_0 & 1 & F \end{Bmatrix}\sum_{q'q''} C^{\varkappa q}_{\varkappa' q' \varkappa'' q''} P_0^{\varkappa' q'} P^{\varkappa'' q''}, \quad \zeta=\zeta^{00}.$$

Here $P_0^{\varkappa q}$ is the polarization tensor of the pumping light in the $\varkappa q$ representation,

$$P_0^{\varkappa q}=-\sum_{\alpha\beta} C^{\varkappa q}_{1\alpha 1\beta}\,\overline{e_0^{\alpha}\, e_0^{*\beta}},$$

e_0 is the corresponding polarization vector, and overbars denote statistical averaging. The expression for the polarization tensor $P^{\varkappa q}$ of the light used to observe the system is similar, except that the averaging is replaced by a summation over the observation polarizations. The total intensity for the structure (8) is given by the coefficients $a = 18(2F+1)\zeta$. The quantities ζ and $S^{\varkappa q}$ are the same as in Ref. 4 [we note that Eq. (6) for $S^{\varkappa q}$ in Ref. 4 contains a misprint—the factor $(2\varkappa+1)^{1/2}$ should appear in the numerator; the numerical factor $a/\zeta = 18(2F+1)$ was also omitted in Ref. 4].

To demonstrate that the odd-rank contributions vanish in (7) for linear pumping and observation polarizations, we note that the symmetry properties of the 9*j*-symbols[11] imply that only the terms for which $\varkappa+\varkappa'+\varkappa''$ is even can be nonzero. Since $P^{1q}=0$ for the linear polarization, in this case only the $S^{\varkappa q}$ with even $\varkappa$ can be nonzero.

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1)This polarization may also be equivalent to spherically symmetric irradiation in conventional optics. For example, the excited fluorescence in this case is unpolarized for intense magnetic fields $\Omega \gg \gamma$ (Ref. 7).

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