Polarization of optical echo in Pr^{3+}:LaF_3

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Echo signals are calculated with allowance for the mechanism for electric dipole transitions, the local symmetry of the ion site in the crystal, and the crystal structure. The echo polarization is governed by simple rules selecting optical transitions. It is equal to the sum of three vectors which are directed along three crystallographic axes, respectively. The relative magnitude of each vector depends on only the areas under the laser pulses and the orientation of the pulse polarization with respect to the corresponding axis.

The Pr^{3+}:LaF_3 crystal is a classic model, which has been studied by numerous methods. Polarization aspects of the generation of echo responses in this system have received little attention. The only study of this topic was the partial study by Zuikov *et al.*,[1] whose results look questionable. They observed experimentally that the intensity of the stimulated echo vanished when the polarizations of two pulses were the same, while that of the third pulse was normal to the first two.[1] On the face of it, it would seem that the polarization of an echo should not depend on the orientations of the polarizations of the exciting pulses, since the Pr^{3+} levels in LaF_3 are simple singlets. However, as the study we are reporting here has shown, the existence of three sites for the Pr^{3+} ions in LaF_3, differing in the direction of the local symmetry axis in the unit cell of the crystal, combines with the simple selection rules imposed by the mechanism for electric dipole transitions to give rise to an angular dependence which furnishes a qualitative explanation of the experimental results of Ref. 1.

In this letter we analyze the polarization of the primary, stimulated, and long-lived stimulated echoes, taking account of the mechanism for the electric dipole transitions in this system, the local symmetry of the rare-earth ion, and the crystal structure.

Weber[2] compared theoretical probabilities for spontaneous transitions with experimental probabilities for Pr^{3+}:LaF_3 and found that optical electronic transitions within the $4f^2$ ground configuration of the Pr^{3+}ion—transitions which are forbidden in the first approximation since they are transitions between states of identical parity—are electric dipole transitions and are caused by the Judd-Ofelt mechanism. At low temperatures this mechanism can be outlined as follows. If the site of the rare-earth ion in the crystal is not a symmetry center, the odd static component of the crystal field adds a state of the opposite parity to the ion wave function, thereby removing the restriction against electric dipole transitions.

The odd static component of the electric field has the dipole symmetry of the rare earth ion, so there are selection rules[3] in terms of the crystal quantum number for electric dipole transitions. These rules depend on the symmetry. In the case of a C_2

symmetry, which is the local symmetry of the Pr^{3+} ions in LaF_3 (Refs. 4 and 5), these rules can be written as follows:

$$\langle m_f,\ f(\Gamma_1)|\mathbf{P}(\alpha)|m_g,\ g(\Gamma_1)\rangle = \mathbf{e}_\alpha P_{fg}\langle m_f|m_g\rangle,\quad \alpha=\alpha,\beta,\gamma, \tag{1}$$

where g and f specify the ground state and the excited state,

$$|m_g,\ g(\Gamma_1)\rangle = |m_g\rangle|g(\Gamma_1)\rangle,\quad |m_f,\ f(\Gamma_1)\rangle = |m_f\rangle|f(\Gamma_1)\rangle;$$

$|m_g\rangle$ and $|m_f\rangle$ are pseudoquadrupole nuclear eigenstates in the ground $[|g(\Gamma_1)\rangle]$ and excited $[|f(\Gamma_1)\rangle]$ electronic states, $\mathbf{P}$ and P_{fg} are the operator and effective matrix element, respectively, of the electric dipole moment, and $\mathbf{e}_\alpha$ is a unit vector along crystallographic axis α. The derivation of (1) allowed for the circumstance that the Pr $^{3+}$ ions in the LaF_3 unit cell can replace La^{3+} ions in three distinct types of sites (α,β,γ),[4,5] whose local C_2-symmetry axes are directed along the crystallographic axes (α,β,γ), which make an angle of $2\pi/3$ in the plane normal to the C_3 axis (the optic axis of the crystal). The selection rules for the α, β, and γ ions are therefore different. The derivation also allowed for the circumstance that the Pr^{3+} multiplets in the C_2-symmetry field in LaF_3 are split into simple nondegenerate levels, whose states are transformed by representations Γ_1 and Γ_2. The ground states of the multiplets are Γ_1 states.[6] Yet another point incorporated in the derivation is that the optical echo is observed between the ground states of only two multiplets, ${}^3H_4(\Gamma_1)-{}^3P_0(\Gamma_1)$ and ${}^3H_4(\Gamma_1)-{}^1D_2(\Gamma_1)$.

It can be seen from (1) that the direction of the dipole matrix element vector is independent of m_f and m_g, i.e., independent of the particular levels of the ground and excited states between which the transition occurs. This circumstance is an important distinction between Pr^{3+}:LaF_3 and other systems. For this reason, the description of the experiment in Ref. 1 with the help of a phenomenological three-level system, with the direction of the echo polarization depending on pseudoquadrupole nuclear states of the ground level, is incorrect. It also follows from (1) that the polarization properties and modulation properties of the echo are factorized, since the modulation properties are determined by nuclear states. It furthermore follows from (1) that the polarizations of the long-lived and ordinary stimulated echoes are the same, since the long-term memory is determined by the lifetime of the nuclear states of the electronic ground level. If the Pr^{3+} ions occupied sites of only one type in the LaF_3 crystal, the echo polarization would be independent of the polarizations of the exciting laser pulses, according to (1). This dependence arises from the presence of three distinct sites (α,β,γ) for the Pr^{3+} ions in the LaF_3 crystal.

To derive an expression for the echo response, we use a technique which has been used to calculate the polarization of an echo in a gas[7] and the modulation of the stimulated echo.[8] We impose selection rules (1). Assuming that the nuclear levels of the ${}^3H_4(\Gamma_1)$ state are equally populated, and that the polarizations of the pulses are linear, we easily derive the following expression for the average dipole moment of the system:

$$\langle\mathbf{P}\rangle = \mathrm{Im}[M\exp(i\omega t)]P_{fg}\mathbf{Q}, \tag{2}$$

where $\omega/2\pi$ is the frequency of the laser field, which is the same for all pulses, and M is a known[8] modulation factor, which incorporates the phases of the pulses and the relaxation and modulation of the echo as a function of the time intervals between pulses. The factor M is different for the primary, stimulated, and long-lived stimulated echoes, and for the forward and backward echoes, but it affects only the amplitude of the signal, not its polarization. We will therefore not reproduce the expression for M here. The expression for $\mathbf{Q}$ is

$$\mathbf{Q}_2 = \sum_{\alpha-\alpha,\beta,\gamma} \mathbf{e}_\alpha \sin[\theta_1(\mathbf{e}_\alpha \mathbf{e}_1)] \sin^2[(\theta_2/2)(\mathbf{e}_\alpha \mathbf{e}_2)] \tag{3}$$

for the forward echo and

$$\mathbf{Q}_3 = \sum_{\alpha-\alpha,\beta,\gamma} \mathbf{e}_\alpha \sin[\theta_1(\mathbf{e}_\alpha \mathbf{e}_1)] \sin[\theta_2(\mathbf{e}_\alpha \mathbf{e}_2)] \sin[\theta_3(\mathbf{e}_\alpha \mathbf{e}_3)] \tag{4}$$

for the stimulated echo. In the case of the backward primary echo, in which case the second pulse is a standing wave, we should replace $\sin^2[(\theta_2/2)(\mathbf{e}_\alpha \mathbf{e}_2)]$ by $1 - J_0[\theta_2(\mathbf{e}_\alpha \mathbf{e}_2)]$ in expression (3). The expression for the backward stimulated echo, in which case pulses 2 and 3 are standing waves, is the same as (4). Here $\theta_i = E_i \Delta t_i P_{fg}$ is the area under pulse i, Δt_i is the length of this pulse, and E_i and $\mathbf{e}_i$ are the amplitude and polarization of the electric field of the laser pulse.

It follows from (3) and (4) that if the polarizations of the two exciting pulses are normal to the β and γ crystallographic axes, respectively, then only α ions are excited, and the polarization is directed along the α axis, regardless of the polarization of the third pulse in the case of the stimulated echo. This selective degeneracy may triple the data storage density. It follows from (4) that if the polarization of each laser pulse is normal to each of the three crystallographic axes, respectively, the intensity of the stimulated echo will vanish, since no ion is excited. The echo intensity also vanishes if the polarizations of the pulses make an angle of $\pi/3$ in the plane normal to the optic axis, and their areas are equal, as follows from (4). In this case, all the ions are excited, but their polarizations cancel out. For an arbitrary orientation of the polarizations of the exciting laser pulses, in the case in which each pulse can simultaneously excite ions at all three types of sites (α,β,γ), the polarization and amplitude of the echo responses depend on the amplitudes and orientations of the polarizations of the pulses with respect to the crystallographic axes. This circumstance can be exploited to determine these axes. For example, if the polarizations of all the pulses are the same $(\mathbf{e}_1 \| \mathbf{e}_2 \| \mathbf{e}_3)$ and are along one of the crystallographic axes, then the echo intensity increases 40% when the polarization of the pulses makes an angle of $\pi/6$ with the given axis in the plane normal to the optic axis, if the relations $\theta_1 = \theta_2 = \theta_3 = \pi/2$ hold. The dependence of the echo polarization and intensity on the orientation of the polarizations of the pulses with respect to the crystallographic axes disappears at small pulse areas. Replacing the sines in (4) by their arguments, and using

$$\sum_{m=1}^{m=3} \exp(i \cdot 2\pi m/3 + i\varphi) = 0$$

(the sum of the roots of one is zero), we easily find

$$\mathbf{Q}_3 = \tfrac{3}{8}\,\theta_1\theta_2\theta_3(q^x\mathbf{e}_x + q^y\mathbf{e}_y) = \tfrac{3}{8}\,\theta_1\theta_2\theta_3\mathbf{q}, \tag{5}$$

$$q_x = \cos(-\varphi_1+\varphi_2+\varphi_3) + \cos(+\varphi_1-\varphi_2+\varphi_3) + \cos(+\varphi_1+\varphi_2-\varphi_3),$$

$$q_y = \sin(-\varphi_1+\varphi_2+\varphi_3) + \sin(+\varphi_1-\varphi_2+\varphi_3) + \sin(+\varphi_1+\varphi_2-\varphi_3),$$

where the (X, Y) plane is normal to the optic axis, along which the laser pulses are propagating, φ_i are the angles between the polarizations of the pulses and the X axis, and the unit vectors $\mathbf{e}_x$ and $\mathbf{e}_y$ are directed along the X and Y axes, respectively. For the case which was realized in Ref. 1, in which the polarizations of the two pulses are the same, e.g., $\varphi_2 = \varphi_3$, we find from (5)

$$\mathbf{q} = \mathbf{e}_1 + 2\mathbf{e}_2(\mathbf{e}_2\mathbf{e}_1), \quad \mathbf{e}_2 \| \ \mathbf{e}_3, \theta_i \ll 1. \tag{6}$$

It follows that the intensity of the stimulated echo decreases by a factor of 9 (but does not vanish, as in Ref. 1) when the polarization of the third pulse is normal to the polarizations of the two other pulses. For finite areas, the echo intensity decreases to a lesser extent, according to estimates. We believe that the reason for the disagreement with Ref. 1 lies in the experimental error, which was not stated in Ref. 1. It also follows from (6) that at small pulse areas, in which case the echo intensity decreases to the greatest extent, the echo intensity, as a function of the angles, does not depend on the areas. Accordingly, we believe that the experimental determination of the effective matrix element of the electric dipole moment in Ref. 1, from this angular dependence, is incorrect.

From the physical standpoint, the simplicity of these results is a consequence of the simple levels and the mechanism for the electric dipole transitions of the rare-earth ion in this matrix.

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