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Quantum theory of inverse magneto-optical effects in rare earth compounds

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Inverse magneto-optical effects arising from the action of short laser pulses on rare-earth ions in various materials are investigated. Primary attention is paid to direct allowed electric dipole $f-d$ and $f-g$ transitions. A theoretical model describing the interaction of ions with the electric field of a laser pulse wave is developed. Expressions for calculating the states of rare-earth ions in the field of an electromagnetic wave are derived. The results show that direct $f-d$ transitions decisively shape the dynamics of magnetic excitations in the material, which is confirmed by numerical calculations for dysprosium orthoferrite.

Keywords: inverse magneto-optical effects, rare earth ions, $f-d$ and $f-g$ transitions, short laser pulses, dysprosium orthoferrite.

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1. Introduction

Research of inverse magneto-optic effects, despite its long history, remains of great scientific importance. Among these effects the most interesting is inverse Faraday effect, which consists in induction of magnetic torque in a magnetic ion with the ion exposure to the circularly polarized light. Development of high power lasers made it possible to study the inverse magneto-optic effects experimentally. Thus, in paper [1] they studied the inverse Faraday effect in $Eu^{2+}:CaF_2$ using a powerful laser with pulse duration of $\tau \approx 30$ ns. The effect arose for the duration of the pulse and practically coincided with it in duration.

Theoretical analysis of the inverse Faraday effect conducted in paper [2] was based on the use of the effective Hamiltonian obtained from analysis of the ion exposure to the infinitely lasting monochromatic electromagnetic wave. Besides, an objective arose to find the internal values, and the solution thereto resulted in the conclusion that the ion had a time-constant magnetic torque.

This approach seems to be rather simplified, especially in connection with the fact that recently the lasers with rather short pulses with duration of $\tau \sim 10$ fs and below have become widely used in the experimental practice [3–5]. Therefore, the theoretical analysis of the magnetic ions exposure to such short pulses is becoming highly relevant.

Among the magnetic materials an important role is played by compounds, whose unique magnetic, magnetic elastic and magnetooptical properties depend on the presence of rare-earth ions in their composition [6].

This paper completed the theoretical analysis of inverse magnetooptical phenomena arising under the exposure of rare-earth ions to the short laser pulses and caused by direct permitted electrodipole $f-d$ and $f-g$ transitions ($4f^N \to 4f^{N-1}5d$ and $4f^N \to 4f^{N-1}5g$).

2. Non-stationary perturbation theory

Let us consider mixing of the states of the rare-earth ion in the wave field of the laser pulse using the approach presented in the paper by Pershan et al. [2] and based on the production of the effective Hamiltonian. In case of radiation in the infrared, visible and ultraviolet ranges, the interaction of the ion with the wave electric field is relevant.

Let us imagine the perturbation Hamiltonian as

$$V(t) = v(t)e^{i\omega t} + v^*(t)e^{-i\omega t}. \quad (1)$$

For the pulse with the envelope of Gaussian shape,

$$v(t) = -\left(\mathbf{dE}_0 + \mathbf{mH}_0\right)e^{-t^2/\tau^2}, \quad (2)$$

where $\mathbf{d} = -e\sum_{i=1}^{N}\mathbf{r}_i$ — operator of electric dipole moment of a rare-earth ion with N electrons in its f-shell, $\mathbf{m} = -\mu_B g_J \mathbf{J}$ — operator of magnetic moment of ion, $\mathbf{E}_0$ and $\mathbf{H}_0$ — amplitudes of accordingly the electric and magnetic fields of wave in the center of the pulse (in a general case, complex values), τ — parameter that defines the pulse duration.

Let us imagine the nonstationary Schrödinger equation as

$$i\hbar\frac{\partial\psi}{\partial t}=(\mathcal{H}_0+\mathcal{V}(t))\,\psi,$$

where $\mathcal{H}_0$ — unperturbed Hamiltonian, energy levels E_k and internal functions φ_k of which are deemed to be known. Wave functions φ_k do not depend on time We will search for the solution to the nonstationary Schrödinger equation as

$$\psi_g(t)=\sum_k a_{kg}(t)e^{-i\omega_k t}\varphi_k$$
$$=\varphi_g e^{-i\omega_g t}+\psi_g^{(1)}(t)+\psi_g^{(2)}(t)+\ldots, \qquad (3)$$

where

$$\psi_g^{(1)}(t)=\sum_e a_{eg}^{(1)}(t)e^{-i\omega_e t}\varphi_e$$

and

$$\psi_g^{(2)}(t)=\sum_n a_{ng}^{(2)}(t)e^{-i\omega_n t}\varphi_n, \qquad (4)$$

and time-dependent coefficients of expansion $a_{eg}^{(1)}(t)$ and $a_{ng}^{(2)}(t)$ are expressed via the matrix elements $V_{eg}(t)=\langle\varphi_e|\mathcal{V}(t)|\varphi_g\rangle=\langle e|\mathcal{V}(t)|g\rangle$ of the perturbation operator $\mathcal{V}(t)$ by states φ_k as follows:

$$a_{eg}^{(1)}(t)=\frac{1}{i\hbar}\int_{-\infty}^{t}e^{i\omega_{eg}t'}V_{eg}(t')dt', \qquad (5)$$

$$a_{ng}^{(2)}(t)=\frac{1}{(i\hbar)^2}\sum_e\int_{-\infty}^{t}V_{ne}(t')e^{i\omega_{ne}t'}\int_{-\infty}^{t'}V_{eg}(t'')e^{i\omega_{eg}t''}dt'dt''.$$

Equations (3)−(5) use designations $\omega_k=E_k/\hbar$ and $\omega_{eg}=(E_e-E_g)/\hbar$. In the case when the perturbation is determined by formula (1), where the value v does not depend on time, the expressions for $a_{kn}^{(1)}(t)$ and $a_{kn}^{(2)}(t)$ may be recorded in a simpler form:

$$a_{eg}^{(1)}(t)=-\frac{1}{\hbar}\left(v_{eg}\frac{e^{i(\omega_{eg}+\omega)t}}{\omega_{eg}+\omega}+v_{eg}^*\frac{e^{i(\omega_{eg}-\omega)t}}{\omega_{eg}-\omega}\right),$$

$$a_{ng}^{(2)}(t)=\frac{1}{\hbar^2}\sum_e\left(\frac{v_{ne}^*v_{eg}}{\omega_{eg}+\omega}+\frac{v_{ne}v_{eg}^*}{\omega_{eg}-\omega}\right)\frac{e^{i\omega_{ng}t}}{\omega_{ng}}.$$

For the Gaussian shape (2) of the pulse the integration of expressions (5) results in the following results:

$$a_{eg}^{(1)}(t)=\frac{i\tau\sqrt{\pi}}{2\hbar}\langle e|\mathbf{dE}_0|g\rangle\big(f_+(\omega_{eg},t)+f_-(\omega_{eg},t)\big),$$

$$f_\pm(\omega_{eg},t)=\exp\left(-\frac{\tau^2(\omega_{eg}\pm\omega)^2}{4}\right)\operatorname{erfc}\big(z_\pm(\omega_{eg},t)\big),$$

$$z_\pm(\omega_{eg},t)=\frac{i\tau(\omega_{eg}\pm\omega)}{2}-\frac{t}{\tau}, \qquad (6)$$

$$a_{ng}^{(2)}(t)=-\frac{\tau^2\sqrt{\pi}}{2\hbar^2}\sum_e\langle n|\mathbf{dE}_0^*|e\rangle\langle e|\mathbf{dE}_0|g\rangle F_+(t)$$
$$+\langle n|\mathbf{dE}_0|e\rangle\langle e|\mathbf{dE}_0^*|g\rangle F_-(t),$$

$$F_\pm(t)=\frac{1}{\tau}\int_{-\infty}^{t}\phi_\pm(t')dt',$$

$$\phi_\pm(t)=\exp\left(i\omega_{ng}t-\frac{2t^2}{\tau^2}\right)$$
$$\times\exp\big(z_\pm^2(\omega_{eg},t)\big)\operatorname{erfc}\big(z_\pm(\omega_{eg},t)\big). \qquad (7)$$

The $\operatorname{erfc}x$ symbol defines the additional function of errors determined by the ratio

$$\operatorname{erfc}x=\frac{2}{\sqrt{\pi}}\int_x^\infty e^{-t^2}dt.$$

Expression (6) provides the description of direct magnetooptical phenomena. In its turn, the inverse magnetooptical phenomena that are of interest for us are determined by ratio (7).

Note that in expressions (6) and (7) the states φ_g and φ_n belong to the main $4f^N$ configuration (orbital quantum number $l=3$), and perturbed states φ_e, having the opposite parity, belong to $4f^{N-1}5d$ $(l'=l-1=2)$ or $4f^{N-1}5g$ $(l'=l+1=4)$ electronic configurations. Further we will ignore the splitting of the levels of perturbed configurations (Judd−Ofelt approximation) and will substitute in equations (6) and (7) all the values ω_{eg} for the value $\omega_{l'}=E_{l'}/\hbar$ (here $l'=l\pm1$), where $E_{l'}$ — average energy of the states of electronic configurations $4f^{N-1}5d$ $(l'=2)$ or $4f^{N-1}5g$ $(l'=4)$. Typical values $E_{l'}$ make $10^4-10^5\,\mathrm{cm}^{-1}$ [10].

Let us introduce operators $W_{\alpha\beta}=d_\alpha P_{l'}d_\beta$ (indices $\alpha,\beta=x,y,z$), where the projection operator is $P_{l'}=\sum_{e_{l'}}|e_{l'}\rangle\langle e_{l'}|$, and expand them into a symmetric $W_{\alpha\beta}^S(l')$ and antisymmetric $W_{\alpha\beta}^A(l')$ components: $W_{\alpha\beta}(l')=W_{\alpha\beta}^S(l')+W_{\alpha\beta}^A(l')$, where $W_{\alpha\beta}^{S,A}=(W_{\alpha\beta}\pm W_{\beta\alpha})/2$, and find that expression (7) may be presented as

$$a_{ng}^{(2)}=-\frac{\tau^2\sqrt{\pi}}{2\hbar^2}\sum_{l'=l\pm1}\langle n|q_{l'}^S(\mathbf{E})(F_++F_-)$$
$$+q_{l'}^A(\mathbf{E})(F_+-F_-)|g\rangle, \qquad (8)$$

where

$$q_{l'}^S(\mathbf{E})=\sum_\alpha|E_\alpha|^2W_{\alpha\alpha}(l')+(E_x^*E_y+E_xE_y^*)W_{xy}^S(l')$$
$$+(E_x^*E_z+E_xE_z^*)W_{xz}^S(l')+(E_y^*E_z+E_yE_z^*)W_{yz}^S(l'), \qquad (9)$$

$$q_{l'}^A(\mathbf{E})=(E_x^*E_y-E_xE_y^*)W_{xy}^S(l')+(E_x^*E_z-E_xE_z^*)W_{xz}^S(l')$$
$$+(E_y^*E_z-E_yE_z^*)W_{yz}^S(l'). \qquad (10)$$

The projection operator $P_{l'}$ is invariant relative to the spatial rotations $\mathbb{R}_3$, therefore, operators $W^{(0)}(l') = \frac{1}{3}\sum_\alpha W_{\alpha\alpha}(l')$, $W^{(1)}_{\alpha\beta}(l') = W^A_{\alpha\beta}(l')$ and $W^{(2)}_{\alpha\beta}(l') = W^S_{\alpha\beta}(l') - W^{(0)}(l')\delta_{\alpha\beta}$ are converted accordingly by D_0, D_1 and D_2 irreducible representations of group $\mathbb{R}_3$. Note also that the operators $W^{(1)}_{\alpha\beta}(l')$ and $L_{\alpha\beta} = L_\alpha L_\beta - L_\beta L_\alpha = i\sum_\gamma \varepsilon_{\alpha\beta\gamma}L_\gamma$ (here $\mathbf{L}$ — operator of the orbital angular moment of the ion) in the operations of the rotation group $\mathbb{R}_3$ are converted using the same representation D_1, therefore, according to the Wigner−Eckart theorem, the matrix elements of these operators in the wave functions $L-S$ of the ion therm are proportionate to each other: $W^{(1)}_{\alpha\beta}(l') = ic_1\sum_\gamma \varepsilon_{\alpha\beta\gamma}L_\gamma$. Similarly, operators $W^{(2)}_{\alpha\beta}(l')$ in the operations of group $\mathbb{R}_3$ are converted in the same manner as the operators of the quadrupole moment $Q_{\alpha\beta}(\mathbf{L})$, therefore in the wave functions $L-S$ of the therm ion $W^{(2)}_{\alpha\beta} = c_2 Q_{\alpha\beta}$. However, for more convenient calculations, it makes more sense to express operators $W^{(2)}_{\alpha\beta}$ and, respectively, operators $q^S(\mathbf{E})$ and $q^A(\mathbf{E})$, via the irreducible tensor operators C^k_q, widely used in the crystal field theory. In this case the corrections to the wave functions of the second order $\psi^{(2)}_g(t)$, introduced by equation (4), that determine the inverse magneto-optic effects, may be presented as, see annex (34),

$$\psi^{(2)}_g(t) = -\frac{\sqrt{\pi}}{2}\left(\frac{\tau}{\hbar}\right)^2 \sum_n \Big(c_1\langle n|q^S(\mathbf{E}_0)|g\rangle + c_2(2-g_J)i[\mathbf{E}^*_0\mathbf{E}_0]\langle n|\mathbf{J}|g\rangle\Big)e^{-i\omega_{n}t}|n\rangle, \quad (11)$$

where $q^S(\mathbf{E})$, c_1 and c_2 are determined accordingly by formulas (25) and (33). The second summand in this expression determines the inverse Faraday effect caused by the presence of the effective magnetic field perturbed by the electric field of the circularly polarized laser pulse:

$$\mathbf{H}_{\text{eff}} \sim i[\mathbf{E}^*\mathbf{E}].$$

The first summand at the ratio (11) determines the even inverse magneto-optic effects, radiation-induced anisotropy etc.

3. Dynamic magnetic structures

In this section, using a developed general theory, we will consider the dynamic magnetic structures of the subsystem of rare-earth ions perturbed by the laser pulse in dysprosium orthoferrite.

Recently some experimental and theoretical studies were conducted on the inverse magneto-optic effects in rare-earth materials, in particular, in dysprosium orthoferrite $DyFeO_3$ [7]. In orthoferrites the rare-earth ions are placed in four nonequivalent nodes, the environmental symmetry of which is described with a point group C_S (reflection in ab-plane) and contains no center of inversion, which causes occurrence of complicated magnetic structures when exposed to a light pulse [7]. The crystalline field splits the multiplets of the rare-earth ions into singlets in case of non-Kramers ions and into doublets in case of Kramers ions.

The choice of dysprosium orthoferrite for analysis (from „the theoretical" point of view) was substantiated by the fact that ion Dy^{3+} in $DyFeO_3$ has simple and well-studied low-lying states [6,8,9]. The crystalline field splits the main multiplet $^6H_{15/2}$ of ion Dy^{3+} into doublets with energies $E_0 = 0\,\text{cm}^{-1}$, $E_1 = 52\,\text{cm}^{-1}$, $E_2 = 147\,\text{cm}^{-1}$ etc., see paper [9]. Besides, the main and first perturbed doublets are Ising and are described accordingly with functions $|\pm 15/2\rangle$ and $|\pm 13/2\rangle$ in the systems of coordinates with axes z, lying in ab-plane at angles $\delta = \pm 60°$ to a-axis. Coordinates of the local axes of symmetry $\mathbf{e}^\alpha_k$ ($\alpha = x, y, z$, and $k = 1, 2, 3, 4$) of nonequivalent positions of rare-earth ions in dysprosium orthoferrite are shown in the table.

Local axes of symmetry of rare-earth ions in dysprosium orthoferrite

k	$\mathbf{e}^x_k$	$\mathbf{e}^y_k$	$\mathbf{e}^z_k$
1	$(0;0;-1)$	$(-\sin\delta;+\cos\delta;0)$	$(+\cos\delta;+\sin\delta;0)$
2	$(0;0;+1)$	$(-\sin\delta;-\cos\delta;0)$	$(+\cos\delta;-\sin\delta;0)$
3	$(0;0;-1)$	$(+\sin\delta;-\cos\delta;0)$	$(-\cos\delta;-\sin\delta;0)$
4	$(0;0;+1)$	$(+\sin\delta;+\cos\delta;0)$	$(-\cos\delta;+\sin\delta;0)$

Besides, the energy of quantum radiation of the laser with wavelength $\lambda = 0.8\,\mu$m is quite close to the difference in the energies of multiplets $^6H_{15/2}$ and $^6F_{5/2}$ of the dysprosium ions. This circumstance initiated the theoretical research of the accounting for the states of the perturbed multiplet $^6F_{5/2}$ of the dysprosium ion as intermediate ones to consider the perturbations of the magnetic system of dysprosium orthoferrite by the light wave field [7].

However, according to the results of paper [7], the closeness of the laser quantum radiation energy to the difference of energies of $^6H_{15/2}$ and $^6F_{5/2}$ multiplets in case of short pulses does not cause drastic resonant increase of the amplitude of magnetic moment oscillations. Therefore, it seems quite relevant to account for direct transitions ($f-d$ and $f-g$) regarding the generation of magnetic perturbations in dysprosium orthoferrite, which was not done in paper [7] and which is considered below as the example of using the general theory of this paper.

The wave function $\psi_g(t)$ of the rare-earth ion in the field of Gaussian-shaped pulse according to equation (3) at $\omega_g = 0$ shall be presented as

$$\psi_g(t) = \varphi_g + \psi^{(2)}_g(t).$$

In case of dysprosium ion, in dysprosium orthoferrite we have

$$\psi_g(t) = |\pm g\rangle = \left|\pm\frac{15}{2}\right\rangle + C^{(2)}_\pm(\mathbf{E},t)\left|\pm\frac{13}{2}\right\rangle, \quad (12)$$

where the following functions are introduced

$$C_{\pm}^{(2)}(\mathbf{E},t) = -\frac{\sqrt{\pi}(er_{fd})^2}{2\sqrt{7}\hbar^2}\left\{\left\langle \pm\frac{13}{2}\middle|q_S\middle|\pm\frac{15}{2}\right\rangle\big(F_+(\omega_1,t) + F_-(\omega,t)\big) + \left\langle \pm\frac{13}{2}\middle|q_A\middle|\pm\frac{15}{2}\right\rangle \times \big(F_+(\omega_1,t) - F_-(\omega,t)\big)\right\}e^{-i\omega_1 t}, \tag{13}$$

where $\omega_1 = E_1/\hbar = 9.9\cdot 10^{12}\,\mathrm{s}^{-1}$, and operators q_S and q_A are given as formulas (9).

The electromagnetic wave of the laser pulse induces the time-dependent magnetic torque in the dysprosium ion, whose components at low temperatures $T \ll E_1/k_\mathrm{B} \sim 75\,\mathrm{K}$ in local axes

$$M_\alpha = \langle +g|\hat{\mu}_\alpha|+g\rangle + \langle -g|\hat{\mu}_\alpha|-g\rangle, \tag{14}$$

where states $|\pm g\rangle$ are determined by ratio (12), and the operator of magnetic torque $\hat{\mu}_\alpha = -\mu_\mathrm{B} g_J J_\alpha$, Landé factor $g_J = 4/3$.

Let the light spread along the crystallographic b-axis. Components of the vector $\mathbf{E}$ in local axes

$$E_x^{(k)} = E_c\big(\mathbf{c}\mathbf{e}_k^x\big), \quad E_y^{(k)} = E_a\big(\mathbf{a}\mathbf{e}_k^y\big), \quad E_z^{(k)} = E_a\big(\mathbf{a}\mathbf{e}_k^z\big),$$

where $\mathbf{e}_k^\alpha$ are given in the table, and E_a and E_c — projections of the vector $\mathbf{E}$ on axis a and c, the directions of which are set by single vectors $\mathbf{a}$ and $\mathbf{c}$ respectively.

Let us consider the incident Gaussian-shaped pulse with the linear and circular polarization. For linear polarized light

$$M_a^{(k)} = A\mu_B g_J(\sin 2\delta)E_{0a}E_{0c}\Phi_1(F_\pm,t)(-1)^{k+1},$$

$$M_b^{(k)} = 2A\mu_B g_J(\cos^2\delta)E_{0a}E_{0c}\Phi_1(F_\pm,t),$$

$$M_c^{(k)} = A\mu_B g_J(\sin 2\delta)E_{0a}^2\Phi_1(F_\pm,t)(-1)^k. \tag{15}$$

For circular polarized light with the left direction of polarization

$$M_a^{(k)} = \frac{1}{2}B\mu_B g_J(\sin 2\delta)E_0^2\Phi_2(F_\pm,t)(-1)^k,$$

$$M_b^{(k)} = B\mu_B g_J(\cos^2\delta)E_0^2\Phi_2(F_\pm,t),$$

$$M_c^{(k)} = \frac{1}{2}B\mu_B g_J(\sin 2\delta)E_0^2\Phi_2(F_\pm,t)(-1)^k. \tag{16}$$

In case of the right direction of circular polarization the values $M_a^{(k)}$ and $M_b^{(k)}$ in formulas (16) must be written with a minus sign. The following designations were used to record expressions (15) and (16):

$$A = \frac{\sqrt{\pi}\tau^2}{4\hbar^2}\,27\sqrt{5\cdot 7}\alpha_2(er_{fd})^2,$$

$$B = \frac{\sqrt{\pi}\tau^2}{12\hbar^2}\,5\sqrt{3}(2-g_J)(er_{fd})^2,$$

where $\alpha_2 = -2/(5\cdot 7\cdot 9)$ — Stevens parameter.

The functions $\Phi_1(F_\pm,t)$ and $\Phi_2(F_\pm,t)$ are defined according to the ratios

$$\Phi_1(F_\pm,t) = \mathrm{Im}\,(F_+ + F_-)\cos\omega_1 t - \mathrm{Re}\,(F_+ + F_-)\sin\omega_1 t = C_1(t)\cos\big(\omega_1 t + \varphi_1(t)\big), \tag{17}$$

$$\Phi_2(F_\pm,t) = \mathrm{Im}\,(F_+ - F_-)\sin\omega_1 t + \mathrm{Re}\,(F_+ - F_-)\cos\omega_1 t = C_2(t)\cos\big(\omega_1 t + \varphi_2(t)\big), \tag{18}$$

where $C_{1,2}(t)$ and $\varphi_{1,2}(t)$ — respectively modules and arguments $(-\pi \leqslant \varphi \leqslant \pi)$ of complex valued functions $F_+ \pm F_-$:

$$C_{1,2}(t) = \sqrt{\mathrm{Re}^2(F_+ \pm F_-) + \mathrm{Im}^2(F_+ \pm F_-)},$$

$$\varphi_{1,2}(t) = \mathrm{sign}\left(\frac{\mathrm{Re}\,(F_+ \pm F_-)}{C_{1,2}(t)}\right)\cdot \arccos\left(\frac{\mathrm{Im}\,(F_+ \pm F_-)}{C_{1,2}(t)}\right). \tag{19}$$

For the wavelength $\lambda = 0.8\,\mu$m of the pulse (circular frequency $\omega = 2.35\cdot 10^{15}\,\mathrm{s}^{-1}$), pulse duration $\tau = 40$ fs, circular frequency $\omega_0 = 2\cdot 10^{16}\,\mathrm{s}^{-1}$ the dependence charts $C_{1,2}(t)$ and $\varphi_{1,2}(t)$ at $-2\tau \leqslant t \leqslant 2\tau$ are shown in the figure.

Therefore, it may be concluded that in case of linear polarization of light the oscillations of all components of the magnetic torque happen according to the law $\cos\omega_1 t$, and in case of the circularly polarized light for the components M_a and M_b according to the law $\sin\omega_1 t$, and for the component M_c according to the law $\cos\omega_1 t$.

Comparison of the values of contributions to the amplitude of oscillations (magnetic modes) of magnetic torques of dysprosium ions from the direct $f-d$ transitions using equations (15)–(18) and resonance ${}^6H_{15/2}$–${}^6F_{5/2}$ transitions [7] indicaes that the contribution of direct $f-d$ transitions by a factor of 3–4 exceeds the contribution of ${}^6H_{15/2}$–${}^6F_{5/2}$ transitions.

Note also that for the crystals with the nonequivalent positions of rare-earth ions, which corresponds to an odd crystalline field, effects arise that are linear by the intensity of the electric field and are described by perturbation

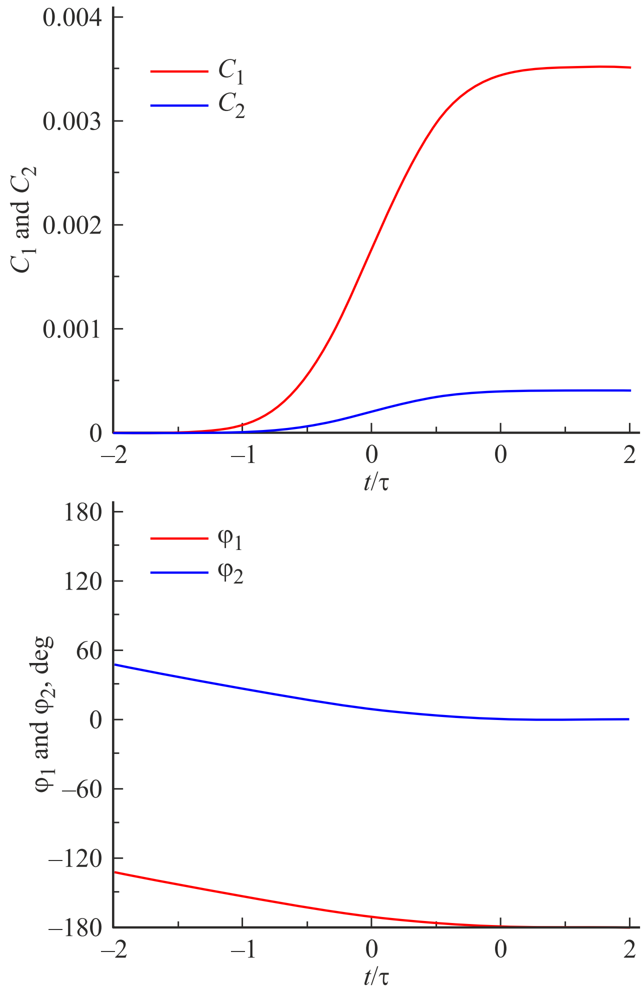
$$V_{odd} = -\mathbf{dE} + V_{CF}^{odd}.$$

In this case

$$a_{ng}^{(2)} \sim \int \langle n|\mathbf{Ed}|e\rangle\langle e|V_{odd}|g\rangle e^{i\omega t} + \langle n|\mathbf{E}^*\mathbf{d}|e\rangle\langle e|V_{odd}|g\rangle e^{-i\omega t} + \langle n|V_{odd}|e\rangle\langle e|\mathbf{Ed}|g\rangle e^{i\omega t} + \langle n|V_{odd}|e\rangle\langle e|\mathbf{E}^*\mathbf{d}|g\rangle e^{-i\omega t}dt,$$

in the specific frequency range of order ω.

The second-order corrections that determine the inverse magneto-optic effects, the effect of the electric field of the



Dependence of amplitudes $C_{1,2}$ and phases $\varphi_{1,2}$ on time t/τ.

wave, $f-d$ transitions:

$$\psi_g^{(2)}(t) = -\frac{\sqrt{\pi}}{2}\left(\frac{\tau}{\hbar}\right)^2 \frac{(er_{fd})^2}{\sqrt{7}} \times \sum_f \Big\{ \big(F_+(\omega_{fg}, t) + F_-(\omega_{fg}, t)\big) \langle f|q_S|g\rangle + \big(F_+(\omega_{fg}, t) - F_-(\omega_{fg}, t)\big) \langle f|q_A|g\rangle \Big\} e^{-i\omega_f t}|f\rangle, \quad (20)$$

where in the Judd–Ofelt approximation

$$F_\pm(\omega_{fg}, t) = \frac{1}{\tau}\int_{-\infty}^{t} \varphi_\pm(\omega_{fg}, t')dt',$$

$$\varphi_\pm(\omega_{fg}, t) = \exp\left(-\frac{2t^2}{\tau^2} + i\omega_{fg}t\right) e^{z_\pm^2(\omega_0, t)} \operatorname{erfc}\big(z_\pm(\omega_0, t)\big),$$

$$z_\pm(\omega_0, t) = \frac{i\tau}{2}(\omega_0 \pm \omega) - \frac{t}{\tau},$$

$\omega_0 \sim 2 \cdot 10^{16}\,\mathrm{s}^{-1}$ — frequency of $f-d$ transition. Operators q_S and q_A are expressed as follows:

$$q_S = \frac{3}{\sqrt{5}}\left(|E_{0z}|^2 - \frac{1}{3}\sum_\alpha |E_{0\alpha}|^2\right) C_0^2 + \frac{3}{2}\sqrt{\frac{6}{5}}\big(\mathrm{Re}\,(E_{0x}^* E_{0z})(C_{-1}^2 - C_1^2) + i\,\mathrm{Re}\,(E_{0y}^* E_{0z})(C_{-1}^2 + C_1^2) + i\,\mathrm{Re}\,(E_{0x}^* E_{0y})(C_{-2}^2 - C_2^2) + \frac{1}{2}\left(|E_{0x}|^2 - |E_{0y}|^2\right)(C_{-2}^2 + C_2^2)\big), \quad (21)$$

$$q_A = \frac{2 - g_J}{3\sqrt{3}}(\mathbf{J}\mathbf{H}_v), \text{ where } \mathbf{H}_v = i[\mathbf{E}_0^* \times \mathbf{E}_0].$$

The formulas are quite simple if the pulse is presented in the form of a sinusoidal train with duration of τ $(-\tau/2 \leqslant t \leqslant \tau/2)$:

$$\psi_g^{(2)}(t) = -\frac{(er_{fd})^2}{4\sqrt{7}\hbar^2}\sum_f \Big\{ \big(G_+(\omega_{fg}, t) + G_-(\omega_{fg}, t)\big)\langle f|q_S|g\rangle + \big(G_+(\omega_{fg}, t) - G_-(\omega_{fg}, t)\big)\langle f|q_A|g\rangle \Big\}.$$

The functions $G_\pm$ at the same time are as follows

$$G_\pm(\omega_{fg}, t) = \begin{cases} \dfrac{1}{(\omega_0 \pm \omega)\omega_{fg}}\left(e^{-i\omega_{fg}\tau/2} - e^{i\omega_{fg}t}\right) \text{ by } -\dfrac{\tau}{2} \leqslant t \leqslant \dfrac{\tau}{2}, \\ \dfrac{\tau}{i(\omega_0 \pm \omega)} \operatorname{sinc}\left(\dfrac{\omega_{fg}\tau}{2}\right) \text{ by } t > \dfrac{\tau}{2}, \\ 0 \text{ by } t < -\dfrac{\tau}{2}. \end{cases}$$

4. Conclusion

The presented paper conducted the theoretical studies of inverse magneto-optic effects arising under exposure of rare-earth ions in various materials to the short laser pulses. The main focus is on the analysis of direct permitted electrodipole $f-d$ and $f-g$ transitions, which have considerable effect on the dynamics of the magnetic perturbations in the material.

A detailed theoretical model was developed that takes into account the interaction of ions with the electric field of the laser pulse wave. This model makes it possible to calculate the mixing of the ion states and to determine the induced magnetic torques that arise under the exposure to the laser radiation. The important result of the model is the detection of the fact that the direct $f-d$ transitions influence substantially the processes of forming the magnetic perturbations in the material, which was demonstrated using the example of dysprosium orthoferrite.

Numerical estimates performed for this material confirm the conclusions of the theoretical model and demonstrate that the accounting for direct $f-d$ transitions is necessary for the proper description of the observed magneto-optic effects. These results highlight the importance of further research in the field of light interaction with the magnetic materials and open new prospects for the development of innovative optomagnetic devices.

5. Appendix

Imagine $\mathbf{dE}$ as the product of cyclic coordinates $\mathbf{d}_\mu$ and $\mathbf{E}_\mu$

$$\mathbf{dE} = \sum_{\mu=0,\pm1} (-1)^\mu d_\mu E_{-\mu},$$

where $d_{\pm1} = \mp(d_x \pm i d_y)/\sqrt{2}$, $d_0 = d_z$ and $E_{\pm1} = \mp(E_x \pm iE_y)/\sqrt{2}$, $E_0 = E_z$. Let us use the genealogical scheme to build the wave functions $|g\rangle$ of the main $4f^N$ and wave functions $|e\rangle$ of the excited $4f^{N-1}5d$ and $4f^{N-1}5g$ configurations [11], according to which

$$|l^N SLM_S M_L\rangle = \sum_{S_1 L_1 m\mu} \sum_{M_{S_1} M_{L_1} \xi} G^{SL}_{S_1 L_1} C^{LM_L}_{L_1 M_{L_1} lm} \times \Psi_{S_1 L_1 M_{S_1} M_{L_1}} (-1)^{N-\xi} \psi_{lm1/2\mu}(\xi),$$

$$|e_{l'}\rangle = \frac{1}{\sqrt{N}} \sum_{\xi} (-1)^{N-\xi} \Psi_{S_1 L_1 M_{S_1} M_{L_1}} \psi_{l'm'1/2\mu'}(\xi), \quad (22)$$

where $C^{jm}_{j_1 m_1 j_2 m_2}$ — Clebsch–Gordan coefficients, $G^{SL}_{S_1 L_1}$ — genealogical coefficients [11,12], L_1, S_1, M_{L_1}, M_{S_1} — quantum numbers of the initial therm. We use formula $d_\mu = -er \sum_{\xi=1}^{N} C^1_\mu(\xi)$ and find that the symmetrical part of the operator $\mathbf{dE}^* |e_{l'}\rangle\langle e_{l'}| \mathbf{dE}$, see (9), will look as follows

$$q^S_{l'}(\mathbf{E}) = \sum_{\mu\tau mn} (-1)^{\mu+\tau} E^*_{-\mu} E_{-\tau} A(ll'1n) C^{nm}_{1\mu 1\tau} C^n_m, \quad (23)$$

where

$$A(ll'1n) = (er_{ll'})^2 \frac{C^{l0}_{l'010} C^{l'0}_{l010}}{C^{l0}_{l0n0}} \begin{Bmatrix} 1 & 1 & n \\ l & l & l' \end{Bmatrix} \sqrt{(2l'+1)(2l+1)}.$$

In formula (23) the curly brackets indicate $6j$-symbol [12], and $r_{ll'} = \langle l|r|l'\rangle = \langle 4f|r|5d\rangle$ and $r_{ll'} = \langle l|r|l'\rangle = \langle 4f|r|5g\rangle$ — radial integrals. The summation in formula (23) yields

$$q^S_{l'}(\mathbf{E}) = \frac{(2l-1)(er_{ll'})^2 q^S(\mathbf{E})}{(2l'+1)\sqrt{2l+1}}, \quad (24)$$

where

$$q^S(\mathbf{E}) = \frac{3}{\sqrt{5}} \left(|E_z|^2 - \frac{1}{3} \sum_\alpha |E_\alpha|^2 \right) C^2_0 + \frac{3}{2}\sqrt{\frac{6}{5}} \left(\frac{E^*_x E_z + E^*_z E_x}{2} (C^2_{-1} - C^2_1) + \frac{E^*_y E_z + E^*_z E_y}{2} i (C^2_{-1} + C^2_1) + \frac{E^*_x E_y + E^*_y E_x}{2} i (C^2_{-2} - C^2_2) + \frac{|E_x|^2 - |E_y|^2}{2} (C^2_{-2} + C^2_2) \right). \quad (25)$$

Now let us find the relationship between operators $W^A_{\alpha\beta}(l')$ and operator L_γ. For this purpose let us calculate the matrix elements $\langle l^N SLM_S M_L | W^A_{xy}(l') | l^N SLM_S M_L\rangle$ and $\langle l^N SLM_S M_L | L_z | l^N SLM_S M_L\rangle$, and then compare them to each other. For the first matrix element we have

$$\langle l^N SLM_S M_L | W^A_{xy}(l') | l^N SLM_S M_L\rangle = \frac{1}{2} \sum_{e_{l'}} \big(\langle LM_L | d_x | e_{l'}\rangle \langle e_{l'} | d_y | LM_L\rangle - \langle LM_L | d_y | e_{l'}\rangle \langle e_{l'} | d_x | LM_L\rangle \big) = \frac{i}{2} \sum_{e_{l'}} \big(\langle LM_L | d_{-1} | e_{l'}\rangle \langle e_{l'} | d_{+1} | LM_L\rangle - \langle LM_L | d_{+1} | e_{l'}\rangle \langle e_{l'} | d_{-1} | LM_L\rangle \big). \quad (26)$$

We use the ratio (10) and will receive

$$\langle LSM_L M_S | d_\tau | e_{l'}\rangle = -er_{ll'}\sqrt{N} \sum G^{SL}_{S_1 L_1} C^{LM_L}_{L_1 M_{L_1} lm_l} C^{SM_S}_{S_1 M_{S_1} 1/2\mu} C^{lm_l}_{l'm_{l'} 1\tau} \frac{\langle l||C_1||l'\rangle}{\sqrt{2l+1}},$$

$$\langle e_{l'} | d_{\tau'} | SLM_S M_L\rangle = -er_{ll'}\sqrt{N} \sum G^{SL}_{S_1 L_1} C^{LM_L}_{L_1 M_{L_1} lm_l} C^{SM_S}_{S_1 M_{S_1} 1/2\mu} C^{l'm_{l'}}_{lm_l 1\tau'} \frac{\langle l'||C_1||l\rangle}{\sqrt{2l'+1}}. \quad (27)$$

We use the ratio (27) at $\tau = \mp1$ and $\tau' = \pm1$ and will find that equation (26) will look as follows

$$\langle SLM_S M_L | W^A_{xy}(l') | SLM_S M_L\rangle = \frac{i}{2} \sum \left(C^{LM_L}_{L_1 M_1 lm_l} \right)^2 \left(G^{SL}_{S_1 L_1} \right)^2 \times \sqrt{\frac{2l'+1}{2l+1}} \Big(\big(C^{lm_l}_{l'm_{l'}1-1}\big)^2 - \big(C^{lm_l}_{l'm_{l'}11}\big)^2 \Big) N (er_{ll'})^2 \times \frac{\langle l||C^1||l'\rangle \langle l'||C^1||l\rangle}{\sqrt{(2l+1)(2l'+1)}} = -\frac{i}{2} N (er_{ll'})^2 \frac{\langle l||C^1||l'\rangle \langle l'||C^1||l\rangle}{l(2l+1)} \times \sum_{S_1 L_1 M_{L_1}} \big(G^{SL}_{S_1 L_1}\big)^2 \big(C^{LM_L}_{L_1 M_{L_1} lm_l}\big)^2 m_l. \quad (28)$$

Further, the matrix element of the operator $L_z = \sum_i l_z(i)$

$$\langle LSM_L M_S | L_z | LSM_S M_L\rangle = N \sum \big(G^{SL}_{S_1 L_1}\big)^2 \big(C^{LM_L}_{L_1 M_{L_1} lm_l}\big)^2 m_l. \quad (29)$$

We compare (28) and (29) and receive that

$$W^A_{xy}(l') = -i (er_{ll'})^2 \frac{\langle l||C_1||l'\rangle \langle l'||C_1||l\rangle}{2l(2l+1)} L_z. \quad (30)$$

Since $\langle l||C_1||l'\rangle = \sqrt{2l'+1} C^{l0}_{l'010}$, then

$$\langle l||C_1||l'\rangle \langle l'||C_1||l\rangle = \begin{cases} -l \text{ by } l' = l-1, \\ -(l+1) \text{ by } l' = l+1 \end{cases}.$$

We use (30) and find that the operator $q^A_{l'}(\mathbf{E})$, according to the definition (10),

$$q^A_{l'}(\mathbf{E}) = \frac{(er_{ll'})^2}{4l(2l+1)}\,(l+l'+1)i\big([\mathbf{E}^*\mathbf{E}]\mathbf{L}\big). \tag{31}$$

We substitute (25) and (31) to the equation (8) and will receive

$$a^{(2)}_{ng} = -\frac{\sqrt{\pi}}{2}\left(\frac{\tau}{\hbar}\right)^2 \big(c_1\langle n|q^S(\mathbf{E})|g\rangle + c_2 i[\mathbf{E}^*\mathbf{E}]\langle n|\mathbf{L}|g\rangle\big)\,. \tag{32}$$

Here the values c_1 and c_2 are expressed as follows:

$$c_1 = \sum_{l'} \frac{(er_{ll'})^2(2l-1)}{(2l'+1)\sqrt{2l+1}}\,\big(F^{l'}_+ + F^{l'}_-\big),$$

$$c_2 = \sum_{l'} \frac{(er_{ll'})^2(l+l'+1)}{4l(2l+1)}\,\big(F^{l'}_+ - F^{l'}_-\big), \tag{33}$$

where the values $F^{l'}_\pm$ are defined by formulas (7), in which frequencies ω_{eg} are replaced with $\omega_{l'}$.

In the vast majority of cases, when you consider rare-earth ions, it is enough to limit yourself to the accounting of the states of the main multiplet. In this approximation in the equation (32) the operator $\mathbf{L}$ may be replaced with operator $(2-g_J)\mathbf{J}$, where g_J — Landé factor of the multiplet. Therefore, mixing of the multiplet states by the electric field of the wave is defined by the expression

$$\psi^{(2)}_g(t) = -\frac{\sqrt{\pi}}{2}\left(\frac{\tau}{\hbar}\right)^2 \sum_n \big(c_1\langle n|q^S(\mathbf{E})|g\rangle + c_2(2-g_J)i[\mathbf{E}^*\mathbf{E}]\langle n|\mathbf{J}|g\rangle\big)e^{-i\omega_n t}|n\rangle. \tag{34}$$

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Conflict of interest

The authors declare that they have no conflict of interest.

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