Faraday effect produced in the residual-ray region by the magnetic moment of an optical phonon in an ionic crystal

Yu. T. Rebane

A. F. Ioffe Physicotechnical Institute, USSR Academy of Sciences

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Faraday rotation due to interaction of the magnetic moment of a circularly polarized optical phonon with an external constant field is considered. It is shown that this mechanism makes the main contribution to the Faraday effect at frequencies near and below the frequency of the transverse optical lattice vibrations in a number of crystals (LiF, NaCl, and others). The Faraday effect can therefore be used for experimental observation of the magnetic moment of a phonon and to estimate the effective ion charges associated with it.

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1. INTRODUCTION

Interest in gyromagnetic phenomena due to molecule vibrations in crystals has increased in recent years. The theory of molecule magnetic moments produced following excitation of circularly polarized oscillations is the subject of Refs. 1–7. In 1961 Vonsovskiĭ and Svirskii pointed out that a circularly polarized phonon in a crystal carries a quantized mechanical moment.[8] It follows therefore that in principle it is possible to produce vibrational magnetization of a solid by irradiating it with circularly polarized light.[9] In this paper we investigate the Faraday effect due to the magnetic moment of the phonon in an ionic crystal.

Optical vibrations of an ionic crystal cause different ions in the unit cell to be differently shifted from their equilibrium positions. This produces in the unit cell an alternating electric dipole moment. This dipole moment plays an important role in different optical and kinetic phenomena in such crystals, since it causes the optical phonons to interact both with the electric field of the light wave and with the charge carriers. In addition, if the lattice vibration is circularly polarized, the electric dipole moment rotates and this in turn leads to the appearance of a magnetic dipole moment. Owing to the large mass of the vibrating ions compared with the electron mass, the magnetic moment of the phonon is quite small. Nevertheless, as will be shown below, the interaction of this magnetic moment with an external constant magnetic field makes the main contribution to the Faraday effect at optical frequencies near and lower than the optical vibrations of the lattice in a number of ionic crystals (LiF, NaCl, and others). By measuring the Faraday-rotation angle it is possible to determine the experimental value of the magnetic moment of the phonon, which is in turn connected with the effective charges of the ions.

2. MAGNETIC MOMENT OF CIRCULARLY POLARIZED OPTICAL PHONON

We consider a diatomic cubic ionic crystal. In long-wave transverse optical vibrations, ions of like sign are displaced in such a crystal as a whole relative to the ions of opposite sign, and the center of gravity of each lattice cell remains in place. In first-order approximation such oscillations can be described by the single-oscillator model.[10] If the crystal is cubic, the oscillator must be isotropic from symmetry considerations. We denote by m_+, m_- and u_+, u_- the masses and displacements from the equilibrium position of the positive and negative ions, respectively. The equations of motion of the lattice take then the form

$$\ddot{\mathbf{u}}=\omega_0{}^2\mathbf{u},\qquad \mathbf{u}=\mathbf{u}_+-\mathbf{u}_-,\qquad \mathbf{u}_+m_-=-\mathbf{u}_-m_+,\tag{1}$$

where ω_0 is the frequency of the transverse optical vibrations. The mechanical angular momentum **L** connected with the lattice vibrations is then

$$L=N\frac{m_+m_-}{m_++m_-}[\mathbf{u}\times\dot{\mathbf{u}}],\tag{2}$$

where N is the number of unit cells in the crystal. At the same time the magnetic moment **M** connected with the lattice vibration is

$$\mathbf{M}=N\frac{e^*}{2c}\frac{m_--m_+}{m_++m_-}[\mathbf{u}\times\dot{\mathbf{u}}],\tag{3}$$

where e^* is the effective charge of the cation and c is the speed of light. It is important that this magnetic moment is proportional to the mass difference of the anion and cation.

It can be seen from a comparision of (2) and (3) that the magnetic and mechanical moments that accompany the lattice vibration are connected by the relation $\mathbf{M} = g\mathbf{L}$, where the gyromagnetic ratio g is equal to

$$g=\frac{e^*}{2m^*c},\qquad \frac{1}{m^*}=\frac{1}{m_+}-\frac{1}{m_-}.\tag{4}$$

In the calculation of the gyromagnetic ratio the lattice vibrations were treated purely classically. On going to the quantum-mechanical description, the quantities **M** and **L** must be replaced by the corresponding operators, the connection between which is determined as before by the gyromagnetic ratio (4). We now use the fact that the mechanical angular momentum of the circularly polarized lattice vibration is quantized and equal to ph, where p is the number of phonons.[8] It is then easy to find from (4) the magnetic moment of the circularly polarized optical phonon

$$\mu=e^*\hbar/2m^*c.\tag{5}$$

In the derivation of (5) we have regarded the moving ions as

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pointlike particles with effective charges $\pm e^*$ and with corresponding masses. Departure from this model calls for a detailed examination of the current induced in the electron shells by the crystal vibrations, as is done for molecules.[11]

3. FARADAY EFFECT IN THE REGION OF RESIDUAL RAYS

The optical properties of a cubic ionic crystal are fully determined by the frequency-dependent dielectric tensor $\varepsilon_{ik}(\omega)$. In the presence of an external constant magnetic field H_0 the dielectric tensor depends also on H_0. In the approximation linear $\varepsilon_{ik}(\omega, \mathbf{H}_0)$, the following formula is valid:

$$\varepsilon_{ik}(\omega, \mathbf{H}_0) = \varepsilon_{ik}(\omega) + \sum_{j=x,y,z} F_{ikj}(\omega) H_j^0. \tag{6}$$

By virture of the cubic symmetry we have $\varepsilon_{ik}(\omega) = \varepsilon(\omega)\delta_{ik}$ and $F_{ikj}(\omega) = F(\omega)\eta_{ikj}$, where η_{ikj} is a fully antisymmetric tensor. The propagation of right- and left-polarized light waves of frequency ω along the magnetic field is described by the dielectric constants $\varepsilon_+(\omega)$ and $\varepsilon_-(\omega)$:

$$\varepsilon_\pm(\omega) = \varepsilon(\omega) \pm iF(\omega) H_0. \tag{7}$$

The Faraday-rotation angle θ, i.e., the angle of rotation of the polarization plane of linearly polarized light as it negotiates a distance d along the magnetic field, is expressed in terms of ε_+ and ε_-:

$$\theta = (\omega d/2c)\,\mathrm{Re}\,(n_+(\omega) - n_-(\omega)), \qquad n_\pm(\omega) = [\varepsilon_\pm(\omega)]^{1/2}, \tag{8}$$

where $n_\pm$ is the complex refractive index for right- and left-polarized light.

We proceed now directly to the calculation of ε_+ and ε_- in a diatomic ionic crystal in the region of the residual rays. To this end, we consider the lattice vibrations in the presence of an external constant magnetic field H_0 and of the light-wave field. We describe the lattice vibrations by the model of one oscillator with damping.[10] In this approximation the equations of motion are

$$\begin{gathered} \ddot{\mathbf{w}} = b_{11}\mathbf{w} + b_{12}\mathbf{E} + b_{13}[\dot{\mathbf{w}} \times \mathbf{H}_0] - \Gamma\dot{\mathbf{w}}, \\ \mathbf{P} = b_{21}\mathbf{w} + b_{22}\mathbf{E}, \\ \mathrm{div}\,\mathbf{D} = 0, \qquad \mathrm{div}\,\mathbf{H} = 0, \\ \mathrm{rot}\,\mathbf{E} = -\frac{1}{c}H, \qquad \mathrm{rot}\,\mathbf{H} = \frac{1}{c}\mathbf{D}, \end{gathered} \tag{9}$$

where $\mathbf{w} = \mathbf{u}[m_+m_-/(m_+ + m_-)]^{1/2}$; $\mathbf{u}$ is the vector of the relative displacement of the two sublattices; $\mathbf{E}$, $\mathbf{P}$, $\mathbf{D}$, and $\mathbf{H}$ are the variables connected with the light wave, viz., the electric field, the polarization of the medium, the electric induction, and the magnetic field; Γ is the damping rate of the free lattice vibrations.

The coefficients in (9) are equal to

$$\begin{gathered} b_{11} = -\omega_0^2, \qquad b_{12} = b_{21} = [(\varepsilon_0 - \varepsilon_\infty)/4\pi]^{1/2}\omega_0, \\ b_{22} = (\varepsilon_\infty - 1)/4\pi, \qquad b_{13} = (e^*/c)(1/m_+ - 1/m_-), \end{gathered} \tag{10}$$

where ε_0 and ε_∞ are the static and high-frequency dielectric constants of the crystal, and ω_0 is the frequency of the transverse lattice vibrations. We seek the solution of the system (9) in the form of a circularly polarized plane wave propagating along the z axis which is parallel to the field H_0. Assuming then that $\mathbf{w}$, $\mathbf{P}$, $\mathbf{E}$, $\mathbf{D}$, and $\mathbf{H}$ are proportional to $(\mathbf{e}_x \pm i\mathbf{e}_y) \times \exp(i\mathbf{kr} - i\omega t)$, we obtain for the amplitudes of these quantities the expressions

$$\begin{gathered} -\omega^2 w = b_{11}w + b_{12}E \pm b_{13}\omega H_0 w + i\Gamma w\omega \qquad P = b_{21}w + b_{22}E, \\ kE = \frac{\omega}{c}H, \qquad kH = -\frac{\omega}{c}(E + 4\pi P) = -\frac{\omega}{c}D. \end{gathered} \tag{11}$$

On the other hand, the connection between D and E is determined by the relation

$$D = \varepsilon_\pm(\omega) E. \tag{12}$$

Comparing (11) and (12) we find that $\varepsilon_\pm(\omega)$ is equal to

$$\varepsilon_\pm(\omega) = \varepsilon_\infty + \frac{(\varepsilon_0 - \varepsilon_\infty)\omega_0^2}{\omega_0^2 - \omega(\omega \pm \omega_{ci} + i\Gamma)}, \tag{13}$$

where $\omega_{ci} = 2\mu H_0$ is the cyclotron frequency for the ion pair, and the phonon magnetic moment μ is defined in (5).

The calculate the angle θ it is necessary to substitute (13) in (8). While the corresponding calculations are straightforward, they are quite cumbersome. We present therefore simple analytic equations for the Faraday-rotation angle θ at high and low frequencies, as well as near resonance.

In the high-frequency region, i.e., at $\omega \gg \omega_0$,

$$\theta = d(\varepsilon_0 - \varepsilon_\infty)\omega_0^2\omega_{ci}/2c\varepsilon_\infty^{1/2}\omega^2. \tag{14}$$

In the low frequency region, i.e., at $\omega \ll \omega_0$

$$\theta = d(\varepsilon_0 - \varepsilon_\infty)\omega^2\omega_{ci}/2c\varepsilon_0^{1/2}\omega_0^2. \tag{15}$$

Near resonance, i.e., in the region $\omega_0^2 - \omega^2 \ll \omega\Gamma$, and if the resonance is narrow enough, namely at $\Gamma \ll (\varepsilon_0 - \varepsilon_\infty)\omega_0/\varepsilon_\infty$, we have

$$\theta = -\frac{\omega d\omega_{ci}}{2c\Gamma}\left[\frac{(\varepsilon_0 - \varepsilon_\infty)\omega_0}{2\Gamma}\right]^{1/2}. \tag{16}$$

What hinders the measurement of the Faraday effect due to the phonon magnetic moment is the strong infrared absorption near the frequency of the transverse optical vibrations. An important characteristic is therefore the value of the Faraday-rotation angle $\theta(\alpha^{-1})$ when the light covers the distance α^{-1}, where α is the light-absorption coefficient. This coefficient is equal to

$$\alpha = 2(\omega/c)\,\mathrm{Im}\,n(\omega). \tag{17}$$

To calculate the complex refractive index $n(\omega)$ in (17) we can use Eq. (13). Since $\omega_{ci} \ll \omega_0$ and $\omega_{ci} \ll \omega$ for experimentally attainable magnetic fields, we have

$$\varepsilon_+(\omega) \approx \varepsilon_-(\omega) = \varepsilon(\omega) = n^2(\omega). \tag{18}$$

Simple calculations show that at high and low frequencies

$$\theta(\alpha^{-1}) = \omega_{ci}/2\Gamma. \tag{19}$$

Near resonance, however,

$$\theta(\alpha^{-1}) = \omega_{ci}/4\Gamma. \tag{20}$$

It can be seen from a comparison of these equations that it is easier to register in experiment the Faraday effect at high or

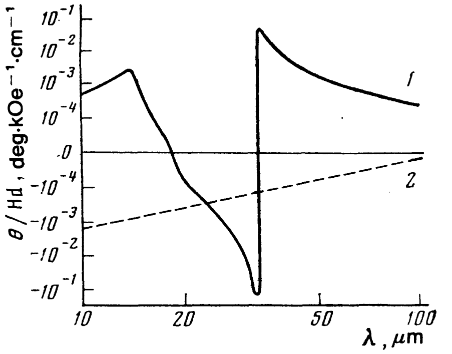

FIG. 1. Spectral dependence of Verdet constant in LiF. 1—rotation due to the phonon magnetic moment; 1—rotation due to virtual interband transitions.

low frequencies than near resonance.

A competing mechanism is Faraday rotation due to virtual transitions of the electrons from the filled valence band to the conduction band. The rotation angle θ_{ib} due to interband transitions can be estimated by using the Becquerel formula:

$$\theta_{\mathrm{ib}}=d(\varepsilon_\infty-1)\,\omega^2\omega_{ce}/2c\varepsilon_\infty^{1/2}\ \omega_{\mathrm{ib}}^2, \tag{21}$$

where $\hbar\omega_{\mathrm{ib}}$ is the characteristic energy of the interband transitions (it is of the order of 10 eV for most ionic crystals); $\omega_{ce} = -|e|H_0/mc$ is the cyclotron frequency of the free electrons.

Figure 1 shows the dependence of the Verdet constant $V=\theta/Hd$ on the light wavelength in LiF, calculated from Eqs. (8) and (13). The following parameter values were used in the calculations[10]: $\omega_0 = 5.78\cdot10^{13}$ sec^{-1}, $m_+ = 7m_p$, $m_- = 19m_p$ (m_p is the proton mass) $\Gamma = 0.05\,\omega_0$, $\varepsilon_0 = 9.27$, $\varepsilon_\infty = 1.92$. The ion charges were assumed equal to the electron charge $\pm e$. Figure 1 shows also an estimate of the Faraday rotation due to interband transitions. The contributions to the Faraday rotation due to the magnetic moment of the phonon and to the interband transitions are of opposite sign at high and low frequencies. The reason is that the signs of the cyclotron frequencies ω_{ci} and ω_{ce} are opposite. It can be seen from the figure that at low frequencies, as well as near resonance, the rotation due to the magnetic moment of the phonon makes the main contribution. It is interesting also to note that allowance for this rotation leads to reversal of the sign of the low-frequency Faraday effect.

As already noted, besides the Verdet constant an important characteristic is also the Faraday-rotation angle over the light-absorption length $\theta(\alpha^{-1})$. For LiF at the magnetic field intensity $H_0 = 100$ kOe this angle equals 0.001° according to (19). Thus, even though the rotation due to the phonon magnetic moment makes the main contribution to the Faraday effect, its observation calls for a highly accurate measurement of the angle of rotation of the light-polarization plane.

We chose the LiF for the particular study because it has a relatively low mass $m^*\approx 11m_p$ and has therefore a relatively large phonon magnetic moment. In addition, as can be seen from (16) and (21), the rotation due to the magnetic moment of the phonon makes the main contribution to the Faraday effect near resonance not only in LiF, but in a large number of other crystals, e.g., NaCl, NaI, KI, BN, and others.

We note in conclusion that in the derivation of the expressions for the dielectric constant (6) and (13) we have neglected effects connected with the fact that the wave vector of light is not zero. Allowance for a finite wave vector of light splits the degenerate transverse vibrations for the asymmetric directions in the crystal. If this splitting is larger than the splitting of the right- and left- polarized vibrations due to the interaction of the magnetic moment of the phonon with the constant magnetic field, the Faraday effect becomes anisotropic and our formulas are valid only for symmetric directions in the crystal. At an opposite inequality, namely at sufficiently strong magnetic fields

$$H_0>\omega_0^3a^2m^*/e^*c, \tag{22}$$

where a is the lattice constant,the Faraday effect will be isotropic and the theory developed here is fully applicable. The inequality (22) the numerical values are substituted for real crystals, requries that the field H_0 exceed several dozen oersteds. This inequality will apparently be always satisfied in practice, for otherwise, according to (19), the Faraday rotation angle will be too small to observe.

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