Anomalous Hall Effect for the Phonon Heat Conductivity in Paramagnetic Dielectrics

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(Received 12 July 2007; published 11 April 2008)

The theory of the anomalous Hall effect for the heat transfer in a parmagnetic dielectric, discovered by Strohm, Rikken, and Wyder [Phys. Rev. Lett. **95**, 155901 (2005)], is developed. The appearance of the phonon heat flux normal to both the temperature gradient and the magnetic field is connected with the interaction of magnetic ions with the crystal field oscillations. In crystals with an arbitrary phonon spectrum this interaction creates the elliptical polarization of phonons. The kinetics related to phonon scattering induced by the spin-phonon interaction determines an origin of the off-diagonal phonon density matrix. The combination of both factors is decisive for the phenomenon under consideration.

DOI: 10.1103/PhysRevLett.100.145902 PACS numbers: 66.70.−f, 72.15.Gd, 72.20.Pa

A novel interesting phenomenon has been found experimentally in a recent Letter [1]. It is a matter of an analog of the anomalous Hall effect (AHE) for the heat conductivity of an ionic paramagnetic dielectric. In fact, applying a magnetic field $\vec{B}$ in the direction normal to heat flow $\vec{j}$, the authors discovered the appearance of the heat transfer in the direction normal to $\vec{B}$ and $\vec{j}$. For the complete lack of free charged carriers and negligible role of the spin-spin coupling at the parameters concerned, the transverse flow is naturally associated with the evolution of the phonon system. However, the magnetic field does not directly act on phonons and only polarizes paramagnetic ions. It is the coupling between phonons and the subsystem of isolated ions carrying magnetic moment $\vec{M}$ that determines the formation of the picture observed. Note in this aspect that the phenomenon concerned is an analog of the AHE in the paramagnetic phase of a ferromagnetic above the Curie point, discovered long time ago [2]. The existence of the effect found in [1] has been confirmed in [3]. In both [1,3] the choice of terbium gallium garnet (TbGG) was not accidental. In this compound for $T < 10$ K the heat resistivity due to spin-phonon interaction (SPI) proves to be about two orders of the magnitude larger as compared with, e.g., gadolinium garnet of the identical structure [4].

As is known, in most of ion dielectrics the spin-phonon relaxation is determined by the two-phonon processes, see, e.g., [5,6]. Usually this supposes the presence of the Kramers doublet in the ground state and the transitions via virtual excitation of higher Kramers doublets. The trivalent ion Tb^{3+} has an even number of f electrons and the Kramers degeneration is absent. However, for rare-earth ions with the even number of f-electrons, appearance of the quasidoublet structure is typical for splitting the multiplet in the crystalline field with the level spacing ε_{12} of about several kelvins in the ground state. TbGG is an example of a similar compound, see, e.g., [7]. From the general analysis [8–10] one can conclude that the SPI under conditions concerned reduces to the interaction of ion magnetic moment and the orbital moments of oscillating surrounding ions. At low temperatures when the long wave acoustic branches alone are excited, the displacement of all atoms in the elementary cell for each mode is the same. One can show in this case that the total orbital moment of atoms in the elementary cell equals to the angular moment of the cell center of gravity. We will assume that the magnetic ordering temperature $T_c \ll T$. This makes it possible to analyze the phenomenon adequately, considering a simple model system with one magnetic atom in the elementary cell and supposing high symmetry of SPI. Assuming that the spacing from the ground quasidoublet to higher levels $\Delta\varepsilon \gg T$, the Hamiltonian of SPI can be represented in the form (see, e.g., [8])

$$H_1 = g\sum_n \vec{s}_n(\vec{U}_n \times \vec{P}_n). \tag{1}$$

Here $\vec{U}_n$ and $\vec{P}_n$ are the vectors of displacement and momentum of the center of gravity in the nth elementary cell, and $\vec{s}_n$ is the isospin for the lowest quasidoublet. For the isotropic SPI $\vec{s}_n \parallel \vec{M}_n$. In the approximation linear in H_1 we can replace $\vec{M}_n$ and, correspondingly, $\vec{s}_n$, with the quantities $\vec{M}$ and $\vec{s}$ averaged over the crystal.

The present Letter is devoted to determining the heat conductivity tensor $\varkappa^{ab}_{odd}(\vec{B})$ antisymmetric and odd in magnetic field $\vec{B}$ [11]. The heat conductivity problem is considered at low temperatures when the heat is carried with long wave acoustic phonons. As is shown below the interaction (1) induces the elleptic renormalization of phonon polarization vectors leaving the phonon spectrum in the linear approximation invariable. The transverse heat flux arrives in these conditions in presence of the correlation between phonons of different branches with the same wave vector $\vec{k}$. Found a solution of the kinetic equation for the heat conductivity problem, at the scattering of phonons on magnetic ions, demonstrates the existence of such correlation. The both factors together, inherent in a crystal with arbitrary phonon spectrum, play a key role in the formation of the phenomenon under consideration.

In the recent paper [12] a variant of the theoretical description for the phenomenon observed in. [1] is presented. Introducing the spin-phonon coupling in the form analogous to (1), the authors take an artificial model for the phonon spectrum in a crystal, assuming the existence of two degenerate transverse modes for an arbitrary wave vector. Within the framework of the model the authors expected to reveal a drift character of the phenomenon for phonons by the analogy with the electron drift in a metal. Not considering obvious doubts in this analogy, note that the authors make a formal mistake in the starting formulas in [12] and, as result, in the following analysis. Authors use the relation (3) (see below) for the velocity $\vec{V}_n$ in the expression for the Hardy energy flux [Eq. (5) in [12]]. Deriving two terms they put incorrectly $\vec{P}_n = m_0 \frac{\partial}{\partial t}\vec{U}_n$ in the first one. Destroying thus the relation (3), they start to consider the second term [Eq. (7) in [12]] as a real flux, which is proportional explicitly to the parameter g and responsible for the phenomenon. In fact, the velocity $\vec{V}_n$ in the initial expression [Eq. (5) in [12]] is the time derivative of atom displacement ($\vec{V}_n = \frac{\partial}{\partial t}\vec{U}_n$). Provided that the relation (3) is strictly kept, the additional heat flux proportional to g does not appear.

Treating the phonon system in the harmonic approximation, we write the general Hamiltonian of the system as

$$H = H_0 + H_1, \qquad H_0 = \sum_n \frac{1}{2m_0}\vec{P}_n^2 - \frac{m_0}{2}\sum_{nn'} D^{ab}_{nn'} U^a_n U^b_{n'}. \tag{2}$$

Here m_0 is the total mass of the elementary cell. Let us determine the Hamilton's equations. For velocity V^a_n, one has, straightforwardly,

$$V^a_n = \dot{U}^a_n = P^a_n/m_0 + g e_{\mathrm{abc}} s^b U^c_n, \tag{3}$$

where e_{abc} is the antisymmetric third-rank unit tensor. Using explicit expression (3), the equation of motion can be transformed to the form

$$-\ddot{U}^a_n = \sum_{n'} D^{ab}_{nn'}(U^b_n - U^b_{n'}) + 2g e_{abc}\dot{U}^b_n s^c. \tag{4}$$

Assuming further consideration to the linear approximation in the SPI, we replaced P^b_n with $m_0\dot{U}^b_n$ in the last term. A usual Fourier-transformation of linear Eq. (4) results in the dispersion equation

$$\omega^2 U^a_{\vec{k}} = D^{ab}_{\vec{k}} U^b_{\vec{k}} - 2ig\omega e_{\mathrm{abc}} U^b_{\vec{k}} s^c, \tag{5}$$

where

$$D^{ab}_{\vec{k}} = \sum_{\vec{R}} D^{ab}_{\vec{R}}(1 - e^{i\vec{k}\vec{R}}), \qquad \sum_{\vec{R}} D^{ab}_{\vec{R}} = 0. \tag{6}$$

Here $\vec{R} = \vec{r}_n - \vec{r}_{n'}$, $\vec{r}_n$ is a position of the center of gravity in the nth cell. In zeroth order in the SPI the solution of a set of Eqs. (5) determines the dispersion law $\omega_s(\vec{k})$ for three acoustic branches and, correspondingly, three orthonormal polarization vectors $e^{(0)a}_s(\vec{k})$ in the long wave limit. The latter ones can be chosen as real. Assuming that the phonon branches are not degenerated, in the approximation linear in the SPI one can conclude from (5) that the phonon spectrum remains unchanged and the polarization vectors are only renormalized. Let us introduce a relation

$$U^a_s(\vec{k}) = e^a_s(\vec{k}) = e^{(0)a}_s(\vec{k}) + \delta e^a_s(\vec{k}). \tag{7}$$

Then from Eq. (5) one finds the small rotation of the polarization vector for $s = 1$ branch

$$\delta\vec{e}_1 = -2ig\omega_1\left[\frac{[(\vec{e}_2 \times \vec{e}_1)\vec{s}]}{(\omega_1^2 - \omega_2^2)}\vec{e}_2 + \frac{[(\vec{e}_3 \times \vec{e}_1)\vec{s}]}{(\omega_1^2 - \omega_3^2)}\vec{e}_3\right]. \tag{8}$$

Accordingly, for $s = 2$ and 3. In the linear approximation in SPI the condition of orthonormality holds for as $e^{a*}_s e^a_{s'} = \delta_{ss'}$.

To find the phonon energy flow j^c with the presence of SPI, one can employ the general results obtained in [13]

$$j^c = \frac{1}{2V} m_0 \sum_{nn'} R^c_{nn'} D^{ab}_{nn'} U^a_n V^b_{n'}. \tag{9}$$

Hereafter the volume is $V = 1$. Let us expand vectors U^a_n into the normal modes in the representation of secondary quantization ($\hbar = 1$)

$$U^a_n = \sum_{ks}\sqrt{\frac{1}{2m_0 N\omega_{ks}}}\exp(i\vec{k}\vec{r}_n)[e^a_s(\vec{k})a_{ks} + e^{a*}_s(-\vec{k})a^+_{-ks}]. \tag{10}$$

Using this expression and the relation (6), we find for the averaged operator of the energy flow

$$\langle j^c\rangle = \frac{1}{4}\sum_{kss'}\left\{\left(\sqrt{\frac{\omega_{ks}}{\omega_{ks'}}} + \sqrt{\frac{\omega_{ks'}}{\omega_{ks}}}\right)(\nabla^c_k D^{ab}_k)e^{a*}_{ks} e^b_{ks'}\right\}\rho_{ss'}(\vec{k}), \tag{11}$$

where $\rho_{ss'}(\vec{k}) = \langle a^+_{ks} a_{ks'}\rangle$. Keeping the approximation linear in SPI, we neglect anomalous averages $\langle a_{-ks} a_{ks'}\rangle$ and $\langle a^+_{ks} a^+_{-ks'}\rangle$ in (11). For zero order approximation in SPI only the symmetric and real component of density matrix gives nonzero contribution to (11). Assuming $\rho_{ss'}(\vec{k}) = n_s(\vec{k})\delta_{ss'}$ and the relation

$$(\nabla^c_k D^{ab}_k)e^a_{ks} e^b_{ks} = \partial\omega^2_{ks}/\partial k^c = 2\omega_{ks} C^c_{ks}, \tag{12}$$

where $\vec{C}_s = \partial\omega_s/\partial\vec{k}$ is the sound velocity, from (11) we find an ordinary expression for the heat flow

$$\langle j^c_0\rangle = \sum_{ks}\omega_{ks} C^c_{ks} n_s(\vec{k}). \tag{13}$$

The contribution linear in the SPI appears in (11) via the renormalization of the polarization vectors (7). Using relations (8), we find that the contribution diagonal in modes vanishes. The off-diagonal term with $ks = 1$ and $ks' = 2$ equals

$$e_1^{a*}e_2^b \to 2ig\frac{(\vec{e}_2\times\vec{e}_1)\vec{s}}{(\omega_1^2-\omega_2^2)}(\omega_1 e_2^{(0)a}e_2^{(0)b}-\omega_2 e_1^{(0)a}e_1^{(0)b})-2ig\frac{(\vec{e}_3\times\vec{e}_2)\vec{s}}{(\omega_2^2-\omega_3^2)}\omega_2 e_1^{(0)a}e_3^{(0)b}+2ig\frac{(\vec{e}_3\times\vec{e}_1)\vec{s}}{(\omega_1^2-\omega_3^2)}\omega_1 e_3^{(0)a}e_2^{(0)b}. \quad (14)$$

With substitution (14) into (11) the expression in the figure brackets, proves to be antisymmetric over indices 1 and 2. The contribution linear in SPI in (11) does not vanish if the off-diagonal terms of density matrix are nonzero. In the expression (14) the first term plays a key role. To avoid the cumbersome expressions, we will find the approximate value for the flow (11), keeping the first term alone. Taking into account (12) we find

$$\langle j_{\mathrm{SO}}^a\rangle = \langle j_{12}^a\rangle + \langle j_{23}^a\rangle + \langle j_{31}^a\rangle,$$
$$\langle j_{12}^a\rangle = ig\sum_k\left(\sqrt{\frac{\omega_1}{\omega_2}}+\sqrt{\frac{\omega_2}{\omega_1}}\right)\frac{2\omega_1\omega_2(\vec{e}_2\times\vec{e}_1)\vec{s}}{(\omega_1^2-\omega_2^2)} \times (C_2^a - C_1^a)\rho_{12}. \quad (15)$$

To determine the nonequilibrium off-diagonal density matrix ρ_{12}, we employ a conventional procedure to derive the kinetic equation, see, e.g., [14]. The general equation of evolution has the form

$$-i\partial_t\rho_{kk'} = \langle[a_k^+a_{k'}, H_0 + H']\rangle. \quad (16)$$

Here H' is the Hamiltonian for the phonon scattering and H_0 is the starting Hamiltonian of noninteracting phonons (2). To simplify notations, we introduce a generalized index for the mode $k \equiv \vec{k}$, s. In the frequency representation

$$(\omega + \omega_k - \omega_{k'})\rho_{kk'} = I_{kk'}, \qquad I_{kk'} = \langle[a_k^+a_{k'}, H']\rangle. \quad (17)$$

The formal stationary solution ($\omega_k \neq \omega_{k'}$) reads

$$\rho_{kk'}|_{\omega\to 0} = \frac{I_{kk'}}{\omega_k - \omega_{k'}}. \quad (18)$$

At low temperatures the main mechanism of scattering is connected with the coupling between phonons and magnetic ions. The splitting of rare-earth ion multiplet with the crystalline field V_{cr} and the reduction of the symmetry for its oscillating part make decisive the Raman two-phonon scattering [5,6]. For ions like Tb^{3+}, the ground state is a quasidoublet with the small spacing of a few kelvins between the doublet components. Considering the region of low temperatures we assume that the kinetics is determined by the scattering of phonons at this quasidoublet. The higher levels lie at $\varepsilon_i \geq 50$ K and we neglect rescattering via them. In this case Hamiltonian H' can be represented in the general form as

$$H' = \frac{1}{N}\sum_{\vec{r}_n}\sum_{fg\alpha} e^{i(\vec{g}-\vec{f})\vec{r}_n}A_{fg}^{\alpha\alpha}\xi_n^\alpha a_f^+ a_g. \quad (19)$$

Here $\alpha = 1, 2$ numerates the doublet levels and $\xi_n^\alpha = 1, 0$. Restricting ourself to the Born approximation in the mean-field approach we find at $\omega \to 0$

$$I_{kk'} = -i\pi\frac{1}{N}\sum_{g\alpha}P_\alpha(1-P_\alpha)A_{k'g}^{\alpha\alpha}A_{gk}^{\alpha\alpha}\{\delta(\omega_k-\omega_g)(n_k-n_g) + \delta(\omega_{k'}-\omega_g)(n_{k'}-n_g)\}, \quad (20)$$

where $P_\alpha = \langle\xi_n^\alpha\rangle$ is the occupation value of level α. From (20) we can conclude that $I_{kk'} = I_{k'k}$. Hence it follows that off-diagonal matrix $\boldsymbol{\rho}_{kk'}$ (18) is antisymmetric. For the equilibrium distribution, $I_{kk'} = 0$. Nonzero result appears only for nonequilibrium distribution $f_k = n_k - N_k^{(0)}$ due to temperature gradient. In the τ-approximation the expression (20) can be represented as

$$I_{kk'} = -i\tfrac{1}{2}(\Omega_{kk'}f_k + \Omega_{k'k}f_{k'}), \quad (21)$$

$$\Omega_{kk'} = 2\pi\frac{1}{N}\sum_{g\alpha}P_\alpha(1-P_\alpha)(A_{k'g}^{\alpha\alpha}A_{gk}^{\alpha\alpha})\delta(\omega_k-\omega_g). \quad (22)$$

The standard solution of the problem for the longitudinal heat conduction gives

$$f_k = -(\omega_k/\Omega_{kk}T^2)N_k^{(0)}(1+N_k^{(0)})\vec{C}_k\nabla T. \quad (23)$$

The substitution of the off-diagonal density matrix (18) into (15) with using (21)–(23) solves the problem for the transverse heat flux due to SPI. The relaxation frequencies (22) depend on a product of the transition amplitudes with $k \neq k'$ unlike usual square of the modulus of the transition amplitude when all quantities are determined by the diagonal density matrix. This typical feature is inherent in any scattering mechanism, in particular, due to phonon anharmonicity.

In the long wave limit the off-diagonal elements of the dynamical matrix for the crystals of high symmetry read $D_k^{ab} \sim k^a k^b$ (see, e.g., [15]). Let us rewrite the dispersion equation (5) for zero order approximation in SPI in the form $(\omega_{ks}^2 - D_k^{aa})e_{ks}^a = \sum_{b\neq a}D_k^{ab}e_{ks}^b$. With varying the sign of the separate projection of wave vector, D_k^{aa} and ω_{ks}^2 remain unchanged. Then it follows from the given equation that the projection of polarization vector e_{ks}^a changes its sign when k^a reverses sign (the sign of the other projections is conserved). This property can be expressed as

$$e_{ks}^a = \tilde{e}_s^a(\vec{k})\mathrm{sgn}k^\alpha. \quad (24)$$

Here $\tilde{e}_s^a(\vec{k})$ is a unit vector remaining unchanged with varying the sign of an arbitrary projection of wave vector $\vec{k}$.

Let the magnetic field and magnetic moment $\vec{M}$ are directed along the z axis, while the longitudinal ∇T is directed along the x axis. Then, restoring $\hbar$, k_B, and volume V, for the transverse component of the heat conduction tensor $\varkappa^{yx}$ we find finally

$$\varkappa^{yx} = \varkappa^{yx}_{12} + \varkappa^{yx}_{23} + \varkappa^{yx}_{31},$$

where

$$\varkappa^{yx}_{12} \simeq k_B \frac{g\hbar^2}{V} \sum_k s^z (\tilde{e}_1 \times \tilde{e}_2)^z \left(\sqrt{\frac{\omega_1}{\omega_2}} + \sqrt{\frac{\omega_2}{\omega_1}}\right) \times \frac{2\omega_1\omega_2 \mathrm{sgn}k^x \mathrm{sgn}k^y}{(\omega_1^2 - \omega_2^2)(\omega_1 - \omega_2)} (C_1^y - C_2^y)(F_{12}c_1^x + F_{21}c_2^x),$$

$$F_{12} = -\frac{1}{2}\omega_1 \frac{\Omega_{12}}{\Omega_{11}} \frac{1}{(k_B T)^2} N_1^{(0)}(1 + N_1^{(0)}). \quad (25)$$

Recall that $s^z \equiv \langle s^z \rangle$. The expression obtained demonstrates the existence of the anomalous phonon Hall effect under comparatively general conditions. It is interesting that the kinetics, reflecting phonon scattering character, enters (25) only via a ratio Ω_{12}/Ω_{11}. This holds for any dominant mechanism of scattering. Under these conditions the transverse heat transfer is determined mainly by the spectral properties of the phonon system. Note, that as it follows from (25), the quantitative enhancement of the effect takes place when the region in which branches ω_{ks} come close is noticeable. We will give a quantitative estimate of the effect, assuming that in the general case the role of these regions in the integral determining (25) is limited.

For the estimate, one should determine the magnitude of parameter g introduced formally. The Hamiltonian (1) due to two-phonon processes accompanying vibrations of the crystalline field has the same origin as H' (19). The Hamiltonian of single-phonon interaction is written usually for a mode $(\vec{k}s)$ in the simplified form $V'(U_{ks}/a)(ka)$, where V' is close to the magnitude of the static crystalline field (see, e.g., [6]). Factor (ka) appears with regard to the relative motion of magnetic ion and surrounding atoms. In the second order in this interaction, the standard canonical transformation results in the spin-orbit interaction as (see, e.g., [8])

$$s^z V'^2 U_{ks} V_{ks'} \frac{\hbar}{\bar{C}^2} \frac{\omega_{ks}^2}{\varepsilon_{12}^2 - \hbar^2\omega_{ks}^2}.$$

To simplify, we introduce here an averaged value for sound velocity $\bar{C}$ and take $\omega_{ks} \simeq \omega_{ks'}$. Choosing two-mode contribution to SPI, written in the form (1), we have $s^z g m_0 U_{ks} V_{ks'}$. Comparing the expressions presented and assuming the temperature $T > \varepsilon_{12}$, we find $g \simeq V'^2/(\hbar m_0 \bar{C}^2)$.

The integral in (25) can be presented in the form $J \sim (\hbar\bar{C}/\pi T)^2 \int dk k^2 \ldots$. The remaining part of the integrand, where $C^a_{1,2}$ is replaced by $C^a_{1,2}/\bar{C}$, proves to be dimensionless. Then, supposing that $\mu_{\mathrm{eff}} B/k_B T < 1$, we find within the accuracy of a numerical factor

$$\varkappa^{yx} \simeq k_B g (k_B \Theta_D/\hbar\bar{C})(T/\Theta_D)(\mu_{\mathrm{eff}} B/k_B T). \quad (26)$$

Taking $V' \simeq 50$ K, $\bar{C} \simeq 4 \times 10^5$ cm/s, $\Theta_D \simeq 400$ K, we arrive at $\varkappa^{yx} \sim 10^{-7}(\mu_{\mathrm{eff}} B/k_B T)$(W/cm K). Using the experimental value for $\varkappa^{xx}$ [1,4], at $\mu_{\mathrm{eff}} B/k_B T \sim 1$ we find for the Hall angle

$$\eta = \varkappa^{yx}/\varkappa^{xx} \sim 2 \times 10^{-5}. \quad (27)$$

In reality, this ratio may be larger since the numerical coefficient omitted in (26) can take the value significantly larger than unity due to the structure of integrand in (25).

Thus, in the general case the anomalous phonon Hall effect originates from a combination of two important factors. The first of them is associated with the appearance of elliptic renormalization for phonon modes as a result of the interaction between phonons and paramagnetic ions. The second factor is connected with the demonstration of the existence of nonzero off-diagonal phonon density matrix in the kinetics accompanying longitudinal heat transfer.

The model used in the Letter is undoubtedly simplified. That is why, the fact that the estimate obtained for the Hall angle (27), with using the real parameters, proves to be comparable with the experimental results [1,3], lying within interval 10^{-4}–10^{-5}, seems very encouraging.

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