Phonon Hall effect in four-terminal nano-junctions

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Abstract. Using an exact nonequilibrium Green's function formulation, the phonon Hall effect (PHE) for paramagnetic dielectrics is studied in a nanoscale four-terminal device setting. The temperature difference in the transverse direction of the heat current is calculated for two-dimensional models with the magnetic field perpendicular to the plane. We find that there is a PHE in nanoscale paramagnetic dielectrics, the magnitude of which is comparable to millimeter scale experiments. If the dynamic matrix of the system satisfies mirror reflection symmetry, the PHE disappears. The Hall temperature difference changes sign if the magnetic field is sufficiently large or if the size increases.

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1. Introduction

In parallel to the study of electronics, phononics has been a very active and hot topic recently [1]. Several conceptual phononic (thermal) devices such as thermal rectifier/diodes [2], transistors [3], logic gates [4] and even thermal memory [5] have been proposed to control phonons and process information with phonons. Most strikingly, the thermal rectifier has been realized experimentally in nanostructures [6]. The magnetic field is another degree of freedom, which could be potentially used to control thermal transport [7]. Indeed, a novel phenomenon—the phonon Hall effect (PHE)—has been discovered experimentally by Strohm et al [8], which is an analogue of the electrical Hall effect for the heat flow in dielectrics. The authors found a temperature difference up to 200 $\mu$K between the sample edges in the direction perpendicular to both the heat flow and the magnetic field. This effect has been confirmed in [9]. The electronic Hall effect is well understood in terms of the Lorentz force. However, since no charges are involved and phonons cannot couple to the magnetic field directly, it is thus not straightforward at all to understand the PHE. The magnetic field can polarize the paramagnetic ions; the subsystem of isolated ions carrying magnetic moment $M$ couples to phonons, and this spin–phonon interaction (SPI) determines the PHE [10, 11].

In order to understand the physics underlying the experiments [8, 9], theoretical models for PHE have been proposed in [10, 11], in which phonons are treated ballistically. However, according to Strohm et al [8], the mean free path (1 $\mu$m) is far less than the system size (15.7 mm); therefore, it is not appropriate to treat the diffusive PHE with a ballistic theory. Moreover, in all previous theoretical work, the SPI were considered by a perturbation theory, and they are not consistent with each other.

As we know, the thermal transport in nanoscale structures can be regarded as ballistic because the size is far less than the mean free path; therefore we can use a ballistic theory to study the PHE in nanosystems. In this paper, we address two questions: (1) whether the PHE exists in nanoscale systems; (2) whether there is an exact non-perturbative theory for PHE in the ballistic region. The first question is in fact not trivial, since many physical laws valid at the macroscopic scale are not necessarily true at the nanoscale. For example, Fourier’s law of heat conduction is broken down at the nanoscale [12]. As for the second question, we will take the nonequilibrium Green’s function (NEGF) approach [13]–[15]. The NEGF is an elegant and powerful method to treat nonequilibrium and interacting systems in a rigorous way. The NEGF is widely applied to electronic and thermal transport, and is successful for the study of the spin Hall effect in junctions [16].

2. Method and formulae

To develop a nonperturbative theory for PHE in nanoscale four-terminal junctions, we consider a model shown in figure 1 by taking into account the actual measuring process. The thermal conductance of the system can be calculated by the NEGF method. As we shall see later, our model systems can produce features similar to experiments, even though our systems are at the nanometer scale while the experimental systems are of millimeter scale and are in the diffusive regime.

We consider the same Hamiltonian of SPI as in [10, 11], which can be expressed in the form $H_I = g \sum_n \vec{S}_n \cdot (\vec{U}_n \times \vec{P}_n)$. Here, $\vec{U}_n$ and $\vec{P}_n$ are the vectors of displacement and momentum of the $n$th lattice site. In the presence of a magnetic field $\vec{B}$, each lattice site has a
magnetization $\vec{M}$. For isotropic SPI, the isospin $\vec{s}_n$ is parallel to $\vec{M}_n$, and the ensemble average of the isospin is proportional to the magnetization, that is $\langle \vec{s}_n \rangle = c \vec{M}$. Therefore, under the mean-field approximation, the SPI can be represented as

$$H_I = \sum_n \tilde{\Lambda} \cdot \vec{U}_n \times \vec{P}_n,$$

where $\tilde{\Lambda} = gc \vec{M}$ has units of frequency. According to Sheng et al [10], $\Lambda$ is estimated to be $0.1 \text{ cm}^{-1} \approx 3 \times 10^9 \text{ Hz}$ at $B = 1 \text{ T}$ and $T = 5.45 \text{ K}$, which is within the possible range of the coupling strength in ionic insulators [17, 18]. In our calculation, we will use this relation to map $\Lambda$ to the magnetic field.

Now we turn to the derivation of the general formulae for the temperature difference in the system as illustrated in figure 1, where a two-dimensional (2D) lattice sample, which can be honeycomb lattices or square lattices, is connected with four ideal semi-infinite leads. We denote the lattice as $N_R \times N_C$ where $N_R$, $N_C$ correspond to the number of rows and columns. We adjust temperatures of the upper and lower probes $T_3$ and $T_4$ such that the heat currents from these two leads vanish, namely, $I_3 = I_4 = 0$. Then we get the relative Hall temperature difference as $R = (T_3 - T_4)/(T_1 - T_2)$.

The total Hamiltonian is assumed to be

$$H = \sum_{\alpha=0}^{4} H_{\alpha} + \sum_{\beta=1}^{4} U_{\beta}^T V P_{\beta} U_0 + U_0^T A P_0,$$

where $H_{\alpha} = \frac{1}{2} P_{\alpha}^T P_{\alpha} + U_{\alpha}^T K_{\alpha} U_{\alpha}$, and $A$ is an antisymmetric, block diagonal matrix with the diagonal elements $(-\Lambda, 0)$. Here, the integers 0 to 4 are associated with the center region, left, right, upper and lower leads, respectively. $U_{\alpha}$ ($P_{\alpha}$) are column vectors consisting of all the displacement (momentum) variables in region $\alpha$. $K_{\alpha}$ is the spring constant matrix and

Figure 1. The four-terminal PHE setup used for calculating the thermal conductance and the temperature difference $T_3 - T_4$. (a) The left and right leads have temperatures $T_1$ and $T_2$; the upper and lower probe-leads have temperatures $T_3$ and $T_4$. The center part can be different lattices, such as square lattices (b) or honeycomb lattices (c).
\( V_{\beta,0} = (V_{0,\beta})^T \) is the coupling matrix between the \( \beta \) lead and the central region. The dynamic matrix of the full linear system without SPI is

\[
K = \begin{pmatrix}
K_1 & 0 & V_{1,0} & 0 & 0 \\
0 & K_3 & V_{3,0} & 0 & 0 \\
V_{0,1} & V_{0,3} & K_0 & V_{0,4} & V_{0,2} \\
0 & 0 & V_{4,0} & K_4 & 0 \\
0 & 0 & V_{2,0} & 0 & K_2
\end{pmatrix}.
\]  

(3)

We obtain the equation for \( U_0 \) and \( P_0 \) as

\[
\frac{\partial U_0(\tau)}{\partial \tau} = P_0(\tau) - AU_0(\tau); \\
\frac{\partial P_0(\tau)}{\partial \tau} = -K_0U_0(\tau) - \sum_{\beta=1}^{4} V_{0,\beta}U_\beta(\tau) - AP_0(\tau).
\]

(4)

(5)

The energy flux to the central region from the lead \( \alpha \) is,

\[
I_\alpha = -\langle \hat{H}_\alpha \rangle = \frac{i}{\hbar} \langle [H_\alpha, H] \rangle, \quad \alpha = 1, 2, 3, 4.
\]

(6)

We define the contour-ordered Green’s function as \( G^{\alpha\beta}(\tau, \tau') \equiv -\frac{1}{\hbar} \langle T_c U_a(\tau)U_\beta(\tau')^T \rangle \), where \( \alpha \) and \( \beta \) refer to the region that the coordinates belong to and \( T_c \) is the contour-ordering operator. Then the equations of motion of the contour ordered Green’s function can be derived. In particular, the retarded Green’s function for the central region in frequency domain is \( G^r[\omega] = (\omega + i\eta)^2 - K_0 - \Sigma'[\omega] - A^2 + 2i\omega A)^{-1} \). Here, \( \Sigma' = \sum_{\alpha=1}^{4} \Sigma_{\alpha}^r \), and \( \Sigma_\alpha = V_{0,\alpha}g_\alpha V_{\alpha,0} \) is the self-energy due to interaction with the heat bath, \( g_\alpha = [(\omega + i\eta)^2 - K_\alpha]^{-1} \). The lesser Green’s function is obtained through \( G^< = G^r \Sigma^< G^a \) in the usual way. We thus can calculate the heat flux by the following formula,

\[
I_\alpha = -\frac{1}{2\pi} \int_{-\infty}^{\infty} d\omega \hbar \omega \text{Re}[\text{Tr}(G^r \Sigma^{<}_\alpha + G^< \Sigma^a_\alpha)].
\]

(7)

If the temperature differences among the leads are very small, we can treat the system in the linear response regime, \( T_\alpha = T + \Delta_\alpha \). The linearized heat flux from each heat bath can be written as

\[
I_\alpha = \sum_{\beta=1}^{4} \sigma_{\alpha\beta}(\Delta_\alpha - \Delta_\beta).
\]

(8)

The conductance from heat bath \( \alpha \) to \( \beta \) is defined as

\[
\sigma_{\alpha\beta} = \int_0^{\infty} \frac{d\omega}{2\pi} \hbar \omega T_{\beta\alpha}[\omega] \frac{\partial f}{\partial T},
\]

(9)

where \( T_{\beta\alpha}[\omega] = \text{Tr}(G^r \Gamma_\beta G^a \Gamma_\alpha) \), \( f = (e^{\hbar\omega/k_B T} - 1)^{-1} \), and \( \Gamma_\alpha = i(\Sigma^a_\alpha[\omega] - \Sigma^{<}_\alpha[\omega]) \). Therefore, if we set the heat flux of lead 3 and lead 4 to zero, we can get the relative Hall temperature difference as

\[
R = (\Delta_3 - \Delta_4)/(\Delta_1 - \Delta_2) = \frac{\sigma_{13}\sigma_{24} - \sigma_{23}\sigma_{14}}{(\sigma_{13} + \sigma_{23} + \sigma_{43})(\sigma_{14} + \sigma_{24} + \sigma_{34}) - (\sigma_{43}\sigma_{34})}.
\]

(10)
Figure 2. Hall temperature difference $R$ versus magnetic field $B$ at temperature $T = 5.45$ K. The hexagons and squares correspond to central regions for the honeycomb and square lattices with a nearest-neighbor coupling. The red dotted line corresponds to a linear fit from 0 to 40 T. The size of the center region for honeycomb lattices is $9 \times 6$, the same as the inset (c) in figure 1.

3. Results

In the following calculation, we assume a lattice constant $a = 2.465 \text{ Å}$, and the force constant $K_L = 0.02394 \text{ eV (amu Å}^2)^{-1}$, $K_T = K_L/4$. The ratio of the longitudinal and transverse sound speed to be $\delta = v_L/v_T \approx \sqrt{K_L/K_T} = 2$. Then the speed of sound for longitudinal acoustic phonons is about $4000 \text{ m s}^{-1}$. As mentioned above, $\Lambda$ is estimated to be about $3 \times 10^9 \text{ Hz} \approx 2.0 \times 10^{-6} \text{ eV at } B = 1 \text{ T}$. We set all the couplings between the leads and central region the same, and all the leads and central region have the same spring constants for simplicity.

From equation (10), we find that the relative Hall temperature difference $R$ is an odd function of magnetic field. The Onsager relation, $\sigma_{\alpha\beta}(\Lambda) = \sigma_{\beta\alpha}(-\Lambda)$, always holds due to the definition of the conductance. Furthermore, if there is a symmetry operation $S$ such that

$$SKS^{-1} = K, \quad SAS^{-1} = -A,$$

then $\sigma_{a\beta}(\Lambda) = \sigma_{a\beta}(-\Lambda)$. If this relation is true, then there is no PHE in the system. These symmetry considerations are consistent with a different treatment for bulk systems based on the Green–Kubo formula [22].

We discuss numerical results in the following. Figure 2 shows the temperature difference changing with magnetic field at temperature $T = 5.45$ K for the honeycomb and square lattices with nearest-neighbor couplings. For the honeycomb case, the Hall temperature is odd and linear in the magnetic field between 0 and 40 T, in that range the slope of the curve is $3 \times 10^{-3} \text{ K T}^{-1}$, comparable to the experimental data in [8]. When the magnetic field is extremely large, it
Figure 3. Thermal conductance versus the magnetic field at temperature $T = 5.45$ K for the honeycomb lattice. Panel (a) shows the conductance between two longitudinal leads $\sigma_{12}$. Panel (b) shows the conductance between one longitudinal lead and one transverse probe-lead. The circles and triangles correspond to $\sigma_{13}$ and $\sigma_{14}$, respectively. The size of the center region is $9 \times 6$.

will decrease. From our calculation, we find that the triangular lattice has a similar behavior. However, for a square lattice with the nearest-neighbor coupling, there is no PHE. The spring constant matrix between every nearest coupling site is diagonal for the square lattice. This matrix and also the full matrix $K$ are invariant with respect to a reflection in the $x$- or $y$-direction, thus satisfying equation (11). If we consider next-neighbor couplings of the lattice, the dynamic matrix $K$ will not have mirror reflection symmetry, and the PHE can emerge.

We show the conductances among different leads in figure 3. Because of the symmetry of the system, we have additional relations, $\sigma_{13} = \sigma_{32} = \sigma_{24} = \sigma_{41}$, and $\sigma_{14} = \sigma_{42} = \sigma_{23} = \sigma_{31}$. We find that the conductances between two longitudinal leads or two transverse probe-leads are even in the magnetic field, which can be seen in figure 3(a); $\sigma_{34}$ has the same property. However, for a honeycomb lattice the conductance between one longitudinal lead and one transverse probe-lead is not an even function of magnetic field (figure 3(b)), which gives a contribution to the Hall temperature difference. Therefore, for a honeycomb lattice, the temperature difference is not zero. But for square lattices, $\sigma_{13}$ is an even function of magnetic field, and the same is true for other components; no PHE exists in such systems.

We show the numerical results for the ratio $R$ at $T = 5.45$ K for honeycomb lattices in figure 4(a). The temperature difference will not be linear when the magnetic field is larger than 40 T, after about 110 T it will decrease, and about $B_c = 230$ T, $R$ changes sign to negative. It is the same critical point for the difference of conductances $\sigma_{13} - \sigma_{14}$, which is consistent with equation (10). Here, the magnetic field is very high, which is just a numerical analysis at the
Figure 4. The relative Hall temperature difference $R$ versus the large magnetic field $B$ (a) and high equilibrium temperature $T$ for the honeycomb lattice (b). (a) Squares show $R$ changing with the magnetic field (left scale); the red solid line shows the conductance difference $\sigma_{13} - \sigma_{14}$ versus magnetic field (right scale). (b) $R$ versus equilibrium temperature at $B = 1$ T.

parameter of SPI: $\hbar \Lambda = 2.0 \times 10^{-6}$ eV; if we can find a material with SPI ten times larger (at the nanoscale it may be possible), the magnetic field can be ten times smaller. In figure 4(b), we show $R$ versus temperature at $B = 1$ T. When the temperature increases, $R$ will increase almost linearly. After some value, it decreases, and then increases again. Finally, it tends to a constant. This behavior is due to the competition of the numerator and denominator in equation (10). When the temperature is very high, all conductances tend to constants due to the ballistic thermal transport.

In [5], it is shown that $R$ decreases with increasing ratio of the longitudinal and transverse sound speed $\delta = v_L/v_T$ and changes sign when $\delta$ becomes larger than 5. However, we find that when the ratio ($\delta > 1$) becomes large, $R$ increases, see figure 5(a). At exactly $\delta = 1$, when the longitudinal speed equals the transverse speed, there is no PHE, which verifies our condition, equation (11), for the absence of the PHE. All the spring constant matrices between the nearest-neighbors become diagonal at $\delta = 1$; the condition (11) holds for a mirror reflection operation. If $\delta < 1$, $R$ increases again with the decreasing of $\delta$. Although the ratio $R$ does not change sign with $\delta$, due to the ballistic nature of a small system, it is sensitive to the geometric details, which is shown in figure 5(b): the magnitude and the sign of $R$ change as the size increases.

4. Conclusion

In conclusion, a theory for the PHE in nanoscale paramagnetic dielectrics by the NEGF approach is developed. The results are consistent with the essential experimental features of the PHE, such as the magnitude and linear magnetic field dependence of the observed transverse temperature difference. We find that there is no PHE if the lattice satisfies a certain symmetry. The symmetry of the dynamic matrix $K$ is the key point for the existence of the PHE. The Hall
Figure 5. (a) The relative Hall temperature difference versus magnetic field for different ratio of the longitudinal and transverse sound speed $\delta = v_L/v_T$. The inset shows $R$ versus $\delta$ at $B = 1$ T. The data are for $9 \times 6$ honeycomb lattices at $T = 5.45$ K. (b) The relative Hall temperature difference versus the number of rows of atoms for fixed aspect ratio $N_R:N_C = 1:2$ at $B = 1$ T and $T = 5.45$ K.

temperature difference changes with the equilibrium temperature and tends to a constant at high temperatures. It does not change sign with the ratio of the longitudinal and transverse sound speed in the range of $\delta \in (0.1, 10)$, but it does change sign as the system size increases.

Most of our results should be verified by experiments on nanoscale paramagnetic dielectrics, which can have potential applications in controlling nanoscale phonon transport. For most paramagnetic dielectric materials, because of the complexity of the coupling (such as next or next–next neighboring interaction), the dynamic matrix does not have the mirror reflection symmetry, and the PHE can be present. The Hall temperature difference behavior with magnetic field can be measured in a strong magnetic field. From our study, the PHE can be measured in nanoscale systems at a relatively high temperature ($\sim 100$ K).

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