Phonon Hall thermal conductivity from Green-Kubo formula

Jian-Sheng Wang and Lifa Zhang
Department of Physics and Center for Computational Science and Engineering, National University of Singapore, Singapore 117542, Republic of Singapore
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We derive a formula for the thermal conductivity tensor of a ballistic phonon Hall model. It is found that, although the diagonal elements of the conductivity tensor diverge to infinite, the off-diagonal elements are finite, antisymmetric, and odd in magnetic field. The off-diagonal elements are non-zero only if the dynamic matrix of the phonon system breaks mirror reflection symmetry. The results are obtained without perturbative assumptions about the spin-phonon interactions.

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The Hall effect of electronic conduction is well-known and has many applications. The analogous effects for the transport of gas molecules, spins, and photons also exit \[1,2,3\]. The phonon Hall effect, that is, the appearance of a transverse thermal current when a magnetic field is applied perpendicular to the direction of temperature gradient, is isoteric and not well understood. Electrons couple directly to the magnetic field through the Lorentz force. There is no obvious coupling between phonons and magnetic field. In 2005, Strohm et al. reported such an effect in a paramagnetic dielectric garnet \(\text{TB}_3\text{Ga}_5\text{O}_{12}\) \[4\] and confirmed also in Ref. \[5\], and called it phonon Hall effect. Two theoretical papers followed \[6,7\]. Both of them considered a similar model of the spin-phonon interaction, and both of them treated the interaction perturbatively. The work of Ref. \[6\] appears to imply that ballistic systems cannot produce a phonon Hall effect, and the authors evoked further high order spin-phonon interaction terms to demonstrate the existence of the effect. Although the two approaches are quite different, one based on Green-Kubo formula, the other on Boltzmann-type kinetic equation, curiously, the final results for the off-diagonal thermal conductivity tensor are similar.

In this paper, we address the following issues. (1) Is a ballistic system capable of producing the phonon Hall effect? Our answer to this question is affirmative. Although the effect will be smaller as the linear size \(L\) of the system becomes larger (scaled as \(1/L\)). (2) What is the role of symmetry? We found that break of a mirror reflection symmetry is essential to observe the phonon Hall effect. If the system looks the same inside a mirror, we should not observe such effect on very general ground. We use the same model as that of Refs. \[6,7\] but without the perturbative assumption. The perturbative expansion with respect to the spin-phonon interaction breaks down near the \(\Gamma\)-point of the phonon dispersion. This complicates the behavior of thermal conductivity at very low temperatures. Since the model is ballistic, the thermal conductivity in general should diverge with the system sizes. But for isotropic systems like the two-dimensional square or honeycomb lattices, the off-diagonal thermal conductivity is in fact finite. In the following, we introduce the model, outline a derivation of the thermal conductivity using Green-Kubo formula, and present numerical results and give some comparison with experiments.

We consider a harmonic periodic lattice with the extra Rahman (or spin-orbit) interaction at each lattice site proportional to \(\mathbf{s} \cdot (\mathbf{r} \times \mathbf{p})\). Here \(\mathbf{s}\) is the (pseudo-) spin representing the Kramer doublet; \(\mathbf{r}\) and \(\mathbf{p}\) are displacement and conjugate momentum. We’ll replace \(\mathbf{s}\) by an average magnetization of the system and choose the vector to be in \(z\) direction. The explicit spin degrees of freedom drop out of the problem. The Hamiltonian of the system can be written in a compact form

\[
H = \frac{1}{2} \mathbf{p}^T \mathbf{p} + \frac{1}{2} \mathbf{u}^T \mathbf{A} \mathbf{u} + \mathbf{u}^T \mathbf{A} \mathbf{p},
\]

where \(\mathbf{u}\) is a column vector of displacements away from lattice equilibrium positions for all the degrees of freedom, multiplied by \(\sqrt{m}\), \(\mathbf{p}\) is the associated conjugate momenta. The Rahman term, \(\mathbf{u}^T \mathbf{A} \mathbf{p}\), is onsite; the matrix \(\mathbf{A}\) is an antisymmetric real matrix, \(\mathbf{A}^T = -\mathbf{A}\), and is block diagonal with diagonal elements (in two dimensions)

\[
\begin{pmatrix}
0 & +h \\
-h & 0
\end{pmatrix}.
\]

We’ll call \(h\) magnetic field although \(h\) is only proportional to the real magnetic field in a paramagnet. It has the dimension of frequency. Since the interaction term depends on momentum, the velocity and momentum are not the same but related through \(\dot{\mathbf{u}} = \mathbf{p} - \mathbf{A} \mathbf{u}\). This is the same model studied in Refs. \[6,7\] except a slightly different notation. It has been proposed (in a more general form) based on quantum theory and fundamental symmetries long time ago to study spin-phonon interactions \[8,9,10\].

Equation (1) is quadratic in the dynamic variables \(\mathbf{u}\) and \(\mathbf{p}\), thus is amenable for an exact solution. Our calculation procedure is as follows. We first obtain the eigen modes of the system. Using the eigen modes, we give an expression for the energy current. We then apply the
Green-Kubo formula to compute the thermal conductivity tensor. Since the system is periodic, we can apply the Bloch theorem. The polarization vector \( \epsilon \) then satisfies

\[
[(-i\omega + A)^2 + D] \epsilon = 0,
\]

where \( D(k) = \sum_i K_{i,i'} e^{i(k_i - k_{i'})} \) is the dynamic matrix. \( K_{i,i'} \) is the submatrix between unit cell \( i \) and \( i' \) in the full spring constant matrix \( K \); \( \mathbf{R}_i \) is the real-space lattice vector. This equation is not a standard eigenvalue problem. It is numerically more advantageous to consider both the right and left eigen vectors. Because of anti-hermitian, there is no guarantee that the frequencies \( \omega \) are not really independent. It is possible to consider both the right and left eigen vectors. Because of the special form of Eq. (4), the left eigen vectors and right eigen vectors are not really independent. It is possible to choose the left eigenvectors \( \hat{x} = (\mu, \overline{\epsilon})^T \) and \( I \) is an identity matrix. Contrary to the usual lattice dynamic problems, the polarization vectors are not orthogonal to each other. We need to consider both the right and left eigen vectors. The orthonormal condition then holds between the left and right eigen vectors. In particular, the eigen modes can be normalized according to

\[
\epsilon^\dagger \epsilon + \frac{i}{\omega} \epsilon^\dagger A \epsilon = 1.
\]

Since the matrix on the right-hand side of Eq. (4) is not anti-hermitian, there is no guarantee that the frequencies \( \omega \) will be real, but the eigenvalues always come in pairs, \( \pm \omega \). We take only \( \omega > 0 \) modes. With these choices of the eigen modes, displacement and momentum operators can be taken in the standard second quantization form,

\[
\begin{align*}
\mathbf{u}_l &= \sum_k \epsilon_k e^{i \mathbf{R}_k \cdot \mathbf{k}} \frac{\hbar}{2\omega_k N} a_k + \text{h.c.}, \\
\mathbf{p}_l &= \sum_k \mu_k e^{i \mathbf{R}_k \cdot \mathbf{k}} \frac{\hbar}{2\omega_k N} a_k + \text{h.c.},
\end{align*}
\]

where \( k = (\mathbf{k}, \sigma) \) specifies the wavevector as well as phonon branch, \( a_k \) is the annihilation operator, and h.c. stands for hermitian conjugate. The momentum and displacement polarization vectors are related by, e.g., \( \mu = -i \omega \epsilon + A \epsilon \). We can verify that the canonical commutation relations are satisfied, \( [u_l, p_{l'}^T] = i\hbar \delta_{l,l'} I \), and \( H = \sum_k \hbar \omega_k (a_k^\dagger a_k + 1/2) \).

Based on a definition of the local energy density and the continuity equation for energy conservation, an energy current density can be defined as \cite{6,11},

\[
J^c = \frac{1}{2V} \sum_{l,i'} (R_{i'}^\dagger - R_{i'}^\ddagger) u_l^T K_{l,i'} u_{l'}^T,
\]

where the index \( c = x, y, \) or \( z \) labels the cartesian axis, \( V \) is the total volume of \( N \) unit cells. The components of the current density vector can be expressed in terms of the creation/annihilation operators. Ignoring the \( a a \) and \( a^\dagger a^\dagger \) terms which vary rapidly with time, one obtains

\[
J^c = \frac{\hbar}{4V} \sum_{k,k'} \left( \sqrt{\frac{\omega_k}{\omega_{k'}}} + \sqrt{\frac{\omega_{k'}}{\omega_k}} \right) \epsilon_k^\dagger \frac{\partial D(k)}{\partial k'} \epsilon_{k'}^\dagger a_{k'}^\dagger \delta_{k,k'}.
\]

The thermal conductivity tensor can be obtained from the Green-Kubo formula \cite{12},

\[
\kappa_{\sigma\sigma'} = \frac{V}{T} \int_0^{\beta} d\lambda \int_0^\infty dt \langle J^a(-i\lambda) J^b(t) \rangle_{\text{eq}},
\]

where \( \beta = 1/(k_B T) \), the average is over the equilibrium ensemble with Hamiltonian \( H \). The time dependence of the annihilation operator is trivially given by \( a_k(t) = a_k e^{-i\omega_k t} \). This is also true if \( t \) is imaginary. Substituting the expression \( J^c \) into Eq. (10), using the result

\[
\langle a_k^\dagger a_{k'}^\dagger a_l a_j \rangle_{\text{eq}} = f_i f_j \delta_{i,j} \delta_{k,l} + f_i (f_j + 1) \delta_{i,j} \delta_{k,l},
\]

where \( f_i = (e^{\beta \hbar \omega_i} - 1)^{-1} \) is the Bose distribution function, we obtain

\[
\begin{align*}
\kappa_{\sigma\sigma'} &= \frac{\hbar}{16VT} \sum_{k,\sigma,\sigma'} \frac{e^{k(\omega - \omega') \beta} - 1}{\omega - \omega'} \frac{1}{\eta - i(\omega - \omega')} \times
\end{align*}

\[
F_{\sigma',\sigma}^a(k) F_{\sigma',\sigma}^b(k) f(\omega') (f(\omega) + 1),
\]

where the \( F \) function is defined as

\[
F_{\sigma,\sigma'}^a(k) = \left( \sqrt{\frac{\omega}{\omega'}} + \sqrt{\frac{\omega'}{\omega}} \right) \epsilon^\dagger \frac{\partial D(k)}{\partial k^a} \epsilon'.
\]

To simplify notations, we have suppressed indices, e.g., \( \omega = \omega_\sigma(k) \), \( \epsilon' = \epsilon_{\sigma'}(k) \). We have added a damping term \( e^{-\eta t} \) when integrating the oscillatory factor. The diagonal element of \( F \) is related to the group velocity, \( F_{\sigma,\sigma}^a(k) = 2 \partial \omega_k^2 / \partial k^a \). The off-diagonal elements are in general not zero. The first term in Eq. (11) factors into two independent summations which does not contribute to \( \kappa_{\sigma\sigma} \) due to symmetry of \( \omega_{\sigma}(k) \) with respect to the wavevector \( \mathbf{k} \). Equation (12), together with the definition (13), is the main result of this paper.

We make some general comments on Eq. (12). The first and last factors inside the summation sign can be combined, \( (e^{\beta \hbar (\omega - \omega')} - 1) f'(f + 1) = f - f' \). Written in this way, the equation resembles the Landauer formula for ballistic transport. The second factor makes the conductivity diverge in the form \( 1/\eta \) unless the leading term in an expansion in the damping factor \( \eta \) happens to be zero. The diagonal elements \( \kappa_{\sigma\sigma} \) indeed diverge to infinite. This is expected, as the system is ballistic consisting of independent oscillating modes. There is no intrinsic scattering mechanism in the system.
The off-diagonal elements do not diverge if the system is isotropic in the sense that $\kappa_{ab}$ is independent of the choice of the coordinate axis. In this special case, the off-diagonal elements are antisymmetric and odd in the magnetic field $h$, $\kappa_{ab}(h) = -\kappa_{ba}(h) = \kappa_{ba}(-h)$, consistent with the Onsager relation. This property does not hold for arbitrary anisotropic systems. We argue that in the isotropic case, Eq. (12) is physical and is the correct prediction for the Hall thermal conductivity.

Even in the isotropic case, the off-diagonal term is zero unless reflection symmetry is broken. More precisely, if there exists an orthogonal transformation independent of $k$ such that $SDS^T = D$, $SAS^T = -A$, then $\kappa_{ab} = 0$ for $a \neq b$. The physical meaning of this symmetry is clear. If we look the system in a mirror, since $D$ is the same and $A$ flips a sign, but the physics must be invariant, we should have $\kappa_{ab}(D, A) = \kappa_{ab}(D, -A)$. But $\kappa_{ab}(D, A)$ must be an odd function in $A$. So we must have $\kappa_{ab} = 0$, $a \neq b$. This property should be quite general, independent of the models used. As an example of systems with vanishing off-diagonal thermal conductivity, we can cite a square lattice (or cubic lattice) with only the nearest neighbor coupling with a dynamic matrix which is diagonal.

In the following, we present numerical results based on Eq. (12). But first, we discuss some interesting features of the phonon dispersion when the Raman interaction term is turned on. In Fig. 1, we show results for a triangular lattice with only the nearest neighbor couplings. The coupling matrix between two sites is such that the longitudinal spring constant is $K_L = 0.144 \text{eV}/(\text{uÅ}^2)$ and the transverse spring constant $K_T$ is 4 times smaller. The unit cell lattice vectors are $(a, 0)$ and $(a/2, a\sqrt{3}/2)$ with $a = 1 \text{Å}$. This choice gives longitudinal and transverse sound velocity $3981 \text{m/s}$ and $1921 \text{m/s}$, respectively, comparable to typical experimental values. At the $\Gamma$-point, the effect of the interaction is to shift the frequencies from $\omega_0$ to $\omega_0 \pm h$, for both the acoustic modes and optical modes (if any). In particular, the acoustic modes develop a gap from 0. Away from the $\Gamma$-point, the corrections are of order $h^2$. Due to the interaction, some modes have imaginary frequencies and are no longer stable. This is very pronounced for the transverse modes for large $h$, see Fig. 1(b). The system can be stabilized, at least for small $h$, by adding a small onsite potential (which, of course, breaks the translational invariance of the lattice). If we change the model to use velocity $\dot{u}$ instead of the conjugate momentum $p$ in the interaction term, an onsite term of magnitude $\tilde{h}^2$ is generated naturally. In such a model, these instabilities do not appear. However, there is no good reason to use $u^T A u$ instead of $u^T A p$ other than the above observation.

In Fig. 2, we give the off-diagonal thermal conductivity $\kappa_{xy}$ of the triangular lattice (assuming 1 Å thick) as a function of $h$ for two different temperatures, $T = 5 \text{K}$ and $100 \text{K}$. It is clear that, for small $h$, the dependence of $\kappa_{xy}$ on $h$ is linear. For large $h$, the growth becomes weaker than linear. For very large $h$ (not shown), due to the instability, $\kappa_{xy}$ becomes rather singular, and can even become negative. This range of parameters is not physical. In computing the results of Fig. 2, we have added a small onsite value of order $10^{-6} K_L$. The results are sensitive for this onsite value only for large $h$, but are nearly independent of the onsite value for small $h$.

In Fig. 3, we display the temperature dependence of the off-diagonal thermal conductivity $\kappa_{xy}$. It is seen that $\kappa_{xy}$ saturates at about $100 \text{K}$ at $h = 0.1 \text{rad THz}$. At low temperatures, $\kappa_{xy}$ decreases with temperature approximately linearly. However, due to a complicated effect of $h$ to the dispersion relation, it appears that $\kappa_{xy}$ has a faster fall off than linear.

We comment on experimental data [4, 5] in compar-
duce a finite life time for the phonons, rendering a finite phonon-phonon and spin-phonon interactions will provide experiments which were done on samples of mm scale. The present theory is more suitable for comparison with experiments. It presents a reflection of the wave nature, is not found here. The in practice. Some of the oscillatory behavior, perhaps of order 0.5 Wm$^{-1}$K$^{-1}$. This has the advantage of stabilizing the sound velocities. The most uncertainty in a comparison is the coupling $h$. The experimental value for $\kappa_{xy}$ at $T = 5.13$ K and magnetic field $H = 3$ T is $2.0 \times 10^{-5}$ Wm$^{-1}$K$^{-1}$ [3]. This is consistent with a very small value of $h = 10^{-3}$ rad THz. Although the diagonal element $\kappa_{xx} (= \kappa_{yy})$ diverges to infinite in our theory, we can choose a finite $\eta$ in Eq. (12) to mimic a finite phonon life time. We find a very weak dependence of $\kappa_{xx}$ on $h$. On the scale of $h \sim 1$ rad THz, $\kappa_{xx}$ is nearly a constant. By matching the experimental value of order 0.5 Wm$^{-1}$K$^{-1}$, we can infer a mean free path $\ell = c/\eta \approx 10^3$ Å (where $c$ is sound velocity), which appears a bit too small given the very low temperatures in experiments.

It is interesting to compare the present treatment with that of nonequilibrium Green’s function (NEGF) approach in Ref. [13]. The qualitative features are in agreement, such as the vanishing phonon Hall effect on square lattice. In NEGF approach, the leads are modeled explicitly. It was assumed that leads do not have the spin-phonon interaction. This has the advantage of stabilizing the system, even though the spin-phonon system represented by the Hamiltonian $H$, Eq. (1), may be unstable as a bulk system. NEGF deals with very small systems in practice. Some of the oscillatory behavior, perhaps of a reflection of the wave nature, is not found here. The present theory is more suitable for comparison with experiments which were done on samples of nm scale.

Another point is the role of nonlinear interactions. The phonon-phonon and spin-phonon interactions will produce a finite life time for the phonons, rendering a finite thermal conductivity tensor for all components. We expect that, if there is a systematic expansion in terms of the phonon life time or in terms of the interaction strength, our main result, Eq. (12), should be the leading contribution. The interaction should give only small corrections.

In summary, we have presented a theory of phonon Hall effect based on a ballistic lattice dynamic model. It is shown that the phonon Hall effect can be present, provided that the system does not possess a reflection symmetry. This is different from Ref. [3], which does not suggest this subtlety. Since the Hamiltonian is quadratic in the dynamic variables, a perturbative treatment is not necessary. In fact, it fails near the $\Gamma$-point. We have given numerical results on a simple two-dimensional triangular lattice and the qualitative features are the same for all lattices in two and three dimensions. In particular, it is not necessary that the system has degenerate phonon modes when $h = 0$. When more elaborate model is known (e.g., from a first-principles calculation), the current theory can be applied to more realistic systems.

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