Phonon Hall thermal conductivity from the Green-Kubo formula

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We derive an exact 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 nonzero only if the dynamic matrix of the phonon system breaks mirror-reflection symmetry. The results are obtained without perturbative treatment of 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.[1–3] 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 esoteric and not well understood. Electrons couple directly to the magnetic field through the Lorentz force. However, there is no obvious coupling between phonons and magnetic field. In 2005, Strohm *et al.*[4] reported such an effect in a paramagnetic dielectric garnet $Tb_3Ga_5O_{12}$ and confirmed also in Ref. 5. 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 in Ref. 7 appeared 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 and the other on Boltzmann-type kinetic equation, curiously, the final results for the off-diagonal thermal-conductivity tensor are similar.

In this Brief Report, 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 model in Refs. 6 and 7 but with an exact treatment. The perturbative expansion with respect to the spin-phonon interaction breaks down near the Γ 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 such as the two-dimensional square or honeycomb lattices, the off-diagonal thermal conductivity is in fact finite. In the following, we introduce the model and outline a derivation of the thermal conductivity using Green-Kubo formula, present numerical results, and give some comparison with experiments.

We consider a harmonic periodic lattice with the extra Raman (or spin-orbit) interaction at each lattice site proportional to $\mathbf{s}\cdot(\mathbf{r}\times\mathbf{p})$. Here $\mathbf{s}$ is a (pseudo) spin representing the Kramer doublet; $\mathbf{r}$ and $\mathbf{p}$ are displacement and conjugate momentum. We will 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

$$H = \frac{1}{2}p^T p + \frac{1}{2}u^T K u + u^T A p, \tag{1}$$

where u is a column vector of displacements away from lattice equilibrium positions for all the degrees of freedom, multiplied by the square root of mass, p is the associated conjugate momentum vector, and K is the force-constant matrix. The superscript T stands for matrix transpose. The Raman term, $u^T A p$, is on site, A is an antisymmetric real matrix, and $A^T=-A$ is block diagonal with diagonal elements (in two dimensions)

$$\begin{pmatrix} 0 & +h \\ -h & 0 \end{pmatrix}. \tag{2}$$

We will call h magnetic field though h is only proportional to the real magnetic field in a paramagnet. It has a dimension of frequency. Since the interaction term depends on the momentum, the velocity and momentum are not the same but related through $\dot{u}=p-Au$. This is the same model studied in Refs. 6 and 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.[8–10] We also note that Hamiltonian (1) is identical in form to the model of ionic crystals in a uniform magnetic field.[11]

Equation (1) is quadratic in the dynamic variables u and p, thus is amenable for an exact solution. Our calculation procedure is as follows. We first obtain the eigenmodes of the system. Using the eigenmodes, 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 $\boldsymbol{\epsilon}$ satisfies

$$[(-i\omega+A)^2+D]\epsilon=0, \tag{3}$$

where $D(\mathbf{k})=\Sigma_{l'}K_{l,l'}e^{i(\mathbf{R}_{l'}-\mathbf{R}_l)\cdot\mathbf{k}}$ is the dynamic matrix. $K_{l,l'}$ is the submatrix between unit cells l and l' in the full spring-constant matrix K; $\mathbf{R}_l$ is the real-space lattice vector. This equation does not define a standard eigenvalue problem. It is numerically more advantageous to consider both the coordinates and momenta and to solve an eigenvalue problem:

$$i\omega x=\begin{pmatrix} A & D \\ -I & A \end{pmatrix}x, \tag{4}$$

where $x=(\mu,\epsilon)^T$ 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 find both the right and left eigenvectors. Because of the special form of Eq. (4), the left and the right eigenvectors are not really independent. It is possible to choose the left eigenvectors $\tilde{x}=(\tilde{\mu},\tilde{\epsilon})=(\epsilon^\dagger, -\mu^\dagger)$. The orthonormal condition then holds between the left and right eigenvectors. In particular, the eigenmodes can be normalized according to

$$\epsilon^\dagger\epsilon+\frac{i}{\omega}\epsilon^\dagger A\epsilon=1. \tag{5}$$

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

$$u_l=\sum_k \epsilon_k e^{i\mathbf{R}_l\cdot\mathbf{k}}\sqrt{\frac{\hbar}{2\omega_k N}}a_k+\text{H.c.}, \tag{6}$$

$$p_l=\sum_k \mu_k e^{i\mathbf{R}_l\cdot\mathbf{k}}\sqrt{\frac{\hbar}{2\omega_k N}}a_k+\text{H.c.}, \tag{7}$$

where $k=(\mathbf{k},\sigma)$ specifies the wave vector as well as the 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 $H=\Sigma_k\hbar\omega_k(a_k^\dagger a_k+1/2)$.

Based on a definition of the local energy density and the continuity equation for energy conservation, an energy current density can be defined as[6,7,12]

$$J^c=\frac{1}{2V}\sum_{l,l'}(R_l^c-R_{l'}^c)u_l^T K_{l,l'}\dot{u}_{l'}, \tag{8}$$

where the index $c=x$, y, or z labels the cartesian axis and 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 aa and $a^\dagger a^\dagger$ terms which vary rapidly with time, one obtains[7]

$$J^c=\frac{\hbar}{4V}\sum_{k,k'}\left(\sqrt{\frac{\omega_k}{\omega_{k'}}}+\sqrt{\frac{\omega_{k'}}{\omega_k}}\right)\epsilon_k^\dagger\frac{\partial D(\mathbf{k})}{\partial k^c}\epsilon_{k'}a_k^\dagger a_{k'}\delta_{\mathbf{k},\mathbf{k}'}. \tag{9}$$

The thermal-conductivity tensor can be obtained from the Green-Kubo formula[13]

$$\kappa_{ab}=\frac{V}{\hbar T}\int_0^{\beta\hbar}d\lambda\int_0^\infty dt\langle J^a(-i\lambda)J^b(t)\rangle_{\text{eq}}, \tag{10}$$

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

$$\langle a_i^\dagger a_j a_k^\dagger a_l\rangle_{\text{eq}}=f_if_k\delta_{ij}\delta_{kl}+f_i(f_j+1)\delta_{il}\delta_{jk}, \tag{11}$$

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

$$\kappa_{ab}=\frac{\hbar}{16VT}\sum_{\mathbf{k},\sigma,\sigma'}\frac{e^{\hbar(\omega'-\omega)\beta}-1}{\omega'-\omega}\frac{1}{\eta-i(\omega-\omega')}\times F^a_{\sigma'\sigma}(\mathbf{k})F^b_{\sigma\sigma'}(\mathbf{k})f(\omega')[f(\omega)+1], \tag{12}$$

where the F function is defined as

$$F^a_{\sigma\sigma'}(\mathbf{k})=\left(\sqrt{\frac{\omega}{\omega'}}+\sqrt{\frac{\omega'}{\omega}}\right)\epsilon^\dagger\frac{\partial D(\mathbf{k})}{\partial k^a}\epsilon'. \tag{13}$$

To simplify notations, we have suppressed indices, e.g., $\omega=\omega_\sigma(\mathbf{k})$ and $\epsilon'=\epsilon_{\sigma'}(\mathbf{k})$. 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, $F^a_{\sigma\sigma}(\mathbf{k})=2\partial\omega_k^2/\partial k^a$. The off-diagonal elements are, in general, not zero. The first term in Eq. (11) factors into two independent summations which do not contribute to κ_{ab} due to the symmetry of $\omega_\sigma(\mathbf{k})$ with respect to the wave vector $\mathbf{k}$. Equation (12), together with the definition (13), is the main result of this Brief Report.

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 η happens to be zero. The diagonal elements κ_{aa} 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 κ_{ab} is independent of the choice of the coordinate axis. In this 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 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 $\mathbf{k}$ such that $SDS^T=D$ and $SAS^T=-A$, 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 $\kappa_{ab}(D,A)$

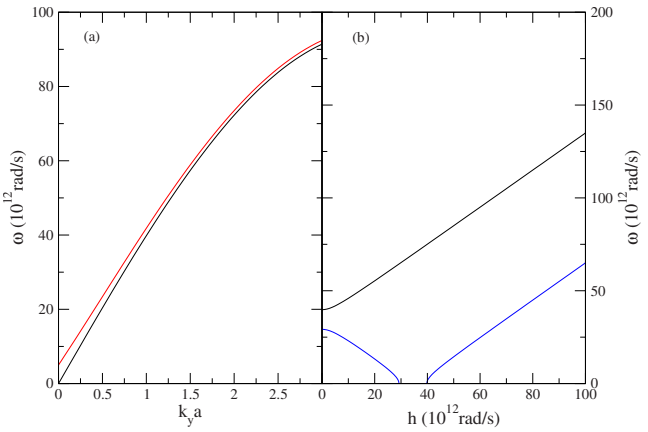

FIG. 1. (Color online) Phonon-dispersion relation of a triangular lattice. (a) The angular frequency of longitudinal mode as a function of $k_y a$ with $k_x=0$. The bottom curve is $h=0$ and the top curve is $h=5\times10^{12}$ rad s^{-1}. (b) The frequency as a function of h at a fixed wave vector $\mathbf{k}a=(0,1)$. The top curve is the longitudinal mode and the bottom (broken) curve is the transverse mode.

$=\kappa_{ab}(D,-A)$. But $\kappa_{ab}(D,A)$ must be an odd function in A. So we must have $\kappa_{ab}=0$ for $a\neq b$. This property should be quite general, independent of 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 present. 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 $K_L=0.144$ eV/(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 $a=1$ Å. This choice gives longitudinal and transverse sound velocities of 3981 and 1921 m/s, respectively, comparable to typical experimental values. At the Γ point, the effect of the interaction is to shift the frequencies from ω_0 to $\omega_0\pm h$, for both the acoustic and optical modes (if any). In particular, the acoustic modes develop gaps from zero. Away from the Γ 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 with large h, see Fig. 1(b). The system can be stabilized, at least for small h, by adding a small on-site potential (which, of course, breaks the translational invariance of the lattice). This instability perhaps will not appear in the ionic-crystal model[11] because on-site terms are supplied by the Lorentz force itself.

In Fig. 2, we give the off-diagonal thermal conductivity κ_{xy} of the triangular lattice (assuming 1 Å thick) as a function of h for two different temperatures, $T=5$ and 100 K. For small h, the dependence of κ_{xy} on h is linear. For large h, the growth becomes weaker than linear. For very large h (not shown), owing to the instability, κ_{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

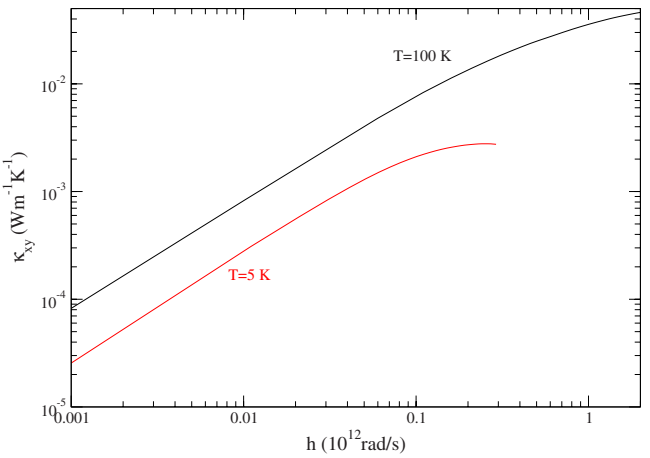

FIG. 2. (Color online) Thermal Hall conductivity as a function of the coupling h for fixed temperatures $T=5$ and 100 K, respectively.

added a small on-site value of order $10^{-6}K_L$. The results are sensitive for this on-site value only for large h but are nearly independent of the on-site value for small h.

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

We comment on experimental data[4,5] in comparison with our numerical results. We have not taken into account the specific lattice structure and atomic details used in experiments. A quantitative comparison is not possible. However, the phonon model parameters are comparable to real systems by matching the sound velocities. The most uncertainty in a comparison is the coupling h. The experimental value for κ_{xy} at $T=5.13$ K and magnetic field $H=3$ T is 2.0×10^{-5} W m^{-1} K^{-1}.[5] This is consistent with a very small value of $h=10^{-3}$ rad THz. The diagonal element $\kappa_{xx}(=\kappa_{yy})$ diverges to infinite in our theory, which implies an infinite mean-free path. We can, however, choose a finite η in Eq.

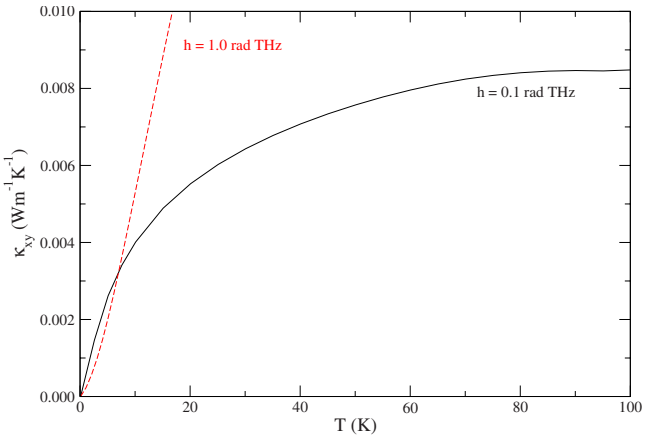

FIG. 3. (Color online) Thermal Hall conductivity κ_{xy} as a function of the temperature T for fixed coupling $h=10^{11}$ rad/s (solid line) and 10^{12} rad/s (dotted line).

(12) to mimic a finite phonon life time. By matching the experimental value of order 0.5 $W\ m^{-1}K^{-1}$, we can give an order-of-magnitude estimate of the mean-free path $\ell = c/\eta \approx 10^3$ Å for the experimental sample (where c is the speed of sound), 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. 14. 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 is 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 behaviors, perhaps of a reflection of the wave nature, are not found here. The present theory is more suitable for comparison with experiments which were done on samples of millimeter 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. Since the Hamiltonian is quadratic in the dynamic variables, a perturbative treatment is not necessary. In fact, it fails near the Γ point. We find that the phonon polarization vectors are not orthonormal. A computationally efficient method is presented to deal with it. 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. When more elaborate models are known (e.g., from a first-principles calculation), the current theory can be applied to more realistic systems. Our theory can also be used for the thermal transport in ionic crystals in a strong magnetic field.

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