Thermal Conductivity and Theory of Inelastic Scattering of Phonons by Collective Fluctuations

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We study the intrinsic scattering of phonons by a general quantum degree of freedom, i.e. a fluctuating “field” \( Q \), which may have completely general correlations, restricted only by unitarity and translational invariance. From the induced scattering rates, we obtain the consequences on the thermal conductivity tensor of the phonons. We find that the lowest-order diagonal scattering rate, which determines the longitudinal conductivity, is controlled by two-point correlation functions of the \( Q \) field, while the off-diagonal scattering rates involve a minimum of three to four point correlation functions. We obtain general and explicit forms for these correlations which isolate the contributions to the Hall conductivity, and provide a general discussion of the implications of symmetry and equilibrium. We evaluate these two- and four-point correlation functions and hence the thermal transport for the illustrative example of an ordered two dimensional antiferromagnet. In this case the \( Q \) field is a composite of magnon operators arising from spin-lattice coupling. A numerical evaluation of the required integrals demonstrates that the results satisfy all the necessary symmetry restrictions but otherwise lead to non-vanishing scattering and Hall effects, and in particular that this mechanism leads to comparable thermal Hall conductivity for thermal currents within and normal to the plane of the antiferromagnetism.

I. INTRODUCTION

Two-point correlation functions are ubiquitous in the study of condensed matter systems. They are often the building blocks of response functions in scattering and other experiments and appear in Feynman diagrams, as well as Monte Carlo simulations. They are the central elements of linear response theory, as is evident from Kubo’s formula [1] [2]. They are often independent of the arbitrary phase choice of the wave function.

Higher order correlation functions have witnessed renewed interest recently. They arise theoretically in the measurement of chaos. A particular type of four-point correlation function, the “out-of-time-ordered” correlator, has been shown to be related to the Lyapunov exponent, which measures the rate at which the result of a measurement diverges after a weak initial perturbation [3]. Multi-point correlations also naturally describe non-linear response, e.g. in non-linear optics such as second harmonic generation, and in “multi-dimensional spectroscopy” [4]. They may also arise in scattering measurements at resonance, such as RIXS [5] [6]. From a statistical point of view, higher order correlation functions measure the non-Gaussianity of the distribution of an observable. The more strongly correlated a state is, i.e. the more it deviates from a free-particle description, the more significant the non-Gaussianity. Hence multi-point functions are essential harbingers of strong correlations.

Here we study the thermal conductivity due to phonons coupled to another degree of freedom, for example an electronic or a magnetic one, and express our results in terms of the correlations of the local observable – e.g. an order parameter – \( Q \) coupled to the phonons, e.g. with an interaction Hamiltonian density

\[
H' = \sum_{n\mathbf{k}} \left( a_{\mathbf{n}\mathbf{k}}^\dagger Q_{\mathbf{n}\mathbf{k}} + a_{\mathbf{n}\mathbf{k}} Q_{\mathbf{n}\mathbf{k}}^\dagger \right),
\]

for the simplest case of linear coupling to the strain tensor (in complete calculations described in the main text, we consider also quadratic coupling, c.f. Eq. (17)). The latter is expressed in terms of phonons, whose creation and annihilation operators in mode \( \mathbf{n}\mathbf{k} \), i.e. at momentum \( \mathbf{k} \) in polarization \( n \), are \( a_{\mathbf{n}\mathbf{k}}^\dagger \), \( a_{\mathbf{n}\mathbf{k}} \). Then we find that the leading diagonal scattering rate is

\[
D_{\mathbf{n}\mathbf{k}} = -\frac{1}{\hbar^2} \int dt e^{-i\omega_{\mathbf{n}\mathbf{k}} t} \left\langle \left( Q_{\mathbf{n}\mathbf{k}}(t), Q_{\mathbf{n}\mathbf{k}}^\dagger(0) \right) \right\rangle_{\beta} + \tilde{D}_{\mathbf{n}\mathbf{k}} ,
\]

where \( \tilde{D}_{\mathbf{n}\mathbf{k}} \) includes both higher order terms and contributions from other mechanisms such as scattering from impurities, \( \left\langle \right\rangle_{\beta} \) denotes thermal averaging, and \( \hbar \) is Planck’s constant divided by \( 2\pi \). This controls the longitudinal (dissipative) part of the thermal conductivity, which, to the same leading order, is

\[
\kappa_{\mu}^{\text{L}} = \frac{\hbar^2}{k_B T^2 V} \sum_{\mathbf{n}\mathbf{k}} \frac{v_{\mathbf{n}\mathbf{k}}^\mu \omega_{\mathbf{n}\mathbf{k}}^2}{4D_{\mathbf{n}\mathbf{k}} \sinh^2(\beta \hbar \omega_{\mathbf{n}\mathbf{k}}/2)},
\]

for \( \mu = x,y,z \), where \( V \) is the volume of the system, \( T \) the temperature, \( k_B \) Boltzmann’s constant, \( \beta = 1/(k_B T) \), and \( \omega_{\mathbf{n}\mathbf{k}} \) and \( v_{\mathbf{n}\mathbf{k}} = \nabla_{\mathbf{k}} \omega_{\mathbf{n}\mathbf{k}} \) are respectively the phonon frequency and velocity in mode \( \mathbf{n}\mathbf{k} \). By contrast, the thermal Hall conductivity is antisymmetric, and hence completely controlled by off-diagonal scattering, and of fourth order. As explained in Sec. [11] the fourth order nature of the Hall effect is guaranteed because second
order contributions to scattering are equivalent to the first Born approximation, which obys detailed balance and effectively preserves time-reversal symmetry (see also Ref. [7]). In turn, we define two four-time correlation functions,

\[
\begin{align*}
\mathbb{W}^{\sigma,++}_{nk\nu'k'}(t, t_1, t_2) &= \text{sign}(t_1) \left\langle \left\{ Q_{nk}(-t - t_1), Q_{\nu'k'}(-t + t_1) \right\} \left\{ Q_{\nu'k'}(t_2), Q_{nk}^\dagger(t_2) \right\} \right\rangle N_{uc}, \\
\mathbb{W}^{\sigma,+-}_{nk\nu'k'}(t, t_1, t_2) &= \text{sign}(t_1) \left\langle \left\{ Q_{nk}(-t - t_1), Q_{\nu'k'}(-t + t_1) \right\} \left\{ Q_{nk}^\dagger(-t_2), Q_{nk}^\dagger(t_2) \right\} \right\rangle N_{uc},
\end{align*}
\]

(4)

(where \(N_{uc}\) is the number of unit cells in the crystal) reflecting particle-particle and particle-hole type processes. Note the combination of commutator \([,\] \) and anti-commutator \(\{,\\} \) in Eq. (4), which imposes the subtle structure that extracts the part of the four-point correlations responsible for a Hall effect. We obtain this structure by isolating the “skew scattering” terms in the phonon Boltzmann equation, which are those which are appropriately anti-symmetric in \(nk \leftrightarrow n'k'\), violate detailed balance, and thereby contribute to a Hall effect. Proper Fourier transformation \(\tilde{W}(\omega) = \int_{-\infty}^{\infty} dt \ e^{i\omega t} W(t)\) converts these into rates

\[
\begin{align*}
\mathbb{W}^{\sigma,++}_{nk\nu'k'} & = \frac{2}{\hbar^2} \mathcal{M} \left[ \tilde{\mathbb{W}}^{\sigma,++}_{nk\nu'k'}(\omega_{nk} + \omega_{\nu'k'}, \omega_{nk} - \omega_{\nu'k'}, \omega_{nk} - \omega_{\nu'k'}) \right], \\
\mathbb{W}^{\sigma,+-}_{nk\nu'k'} & = \frac{2}{\hbar^2} \mathcal{M} \left[ \tilde{\mathbb{W}}^{\sigma,+-}_{nk\nu'k'}(\omega_{nk} - \omega_{\nu'k'}, \omega_{nk} + \omega_{\nu'k'}, \omega_{nk} + \omega_{\nu'k'}) \right].
\end{align*}
\]

(5)

Then the thermal Hall (antisymmetric) conductivity at leading order is expressed as

\[
\kappa_{\mu\nu}^H = \frac{\hbar^2}{k_B T^2} \sum_{nk\nu'k'} \frac{1}{2D_{nk}} \left( \left( \frac{1}{N_{uc}} \sum_{q=\pm1} (e^{\beta \hbar \omega_{nk}} - e^{\beta \hbar \omega_{\nu'k'}}) \mathbb{W}^{\sigma,+-}_{nk\nu'k'} \right) e^{\beta \hbar \omega_{nk}} / 2 - D_{nk}\right),
\]

(6)

where \(\mu, \nu = x, y, z\) and we defined the phonon current \(J^\mu_{nk} = N_{eq}^{\omega_{nk}} \hat{v}_{nk}^\mu,\) and \(N^{eq}_{nk}\) is the number of phonons in mode \(nk\) in thermal equilibrium.

Eqs. (2-6) summarize the key general results of this paper, to leading order for the simplest case of linear coupling to an order parameter to phonons. More general formulae including both linear and quadratic coupling are given in Section IIIID. These equations may be applied to obtain the phonon thermal conductivity for any system provided the correlations of the quantities \(Q\) coupling to phonons are known.

The results in this paper are derived using the Boltzmann equation [8], applying Fermi’s golden rule and the first and second Born approximations to the transition probabilities between initial and final states of the joint observable-and-phonon system. These resulting collision terms can be expressed through multi-point correlation functions of the observable. By solving the Boltzmann equation, and computing the resulting phonon thermal currents, we obtain the results quoted above and their generalizations.

In light of several experimental and theoretical studies [9][12] which highlight the major role of phonons in the thermal transport in magnetic systems, we demonstrate our formalism on a model for an ordered two-dimensional antiferromagnet, inspired by experiments on the cuprates [13]. In this case the \(Q\) fields constitute bilinears of magnon operators. In a separate publication [14], we will present a second application to a spinon Fermi surface quantum spin liquid [13] and other fermionic systems, including electronic ones.

\[\text{II. SETUP}\]

\[\text{A. Derivation}\]

The quasiparticle nature of phonons justifies treating their dynamics within the Boltzmann equation,

\[
\partial_t \mathcal{N}_{nk} + v_{nk} \cdot \nabla_r \mathcal{N}_{nk} = \mathcal{C}_{nk}\{\{\mathcal{N}_{nk}\}\},
\]

(7)

where \(\mathcal{N}_{nk}(i_p) = \langle i_{p}\rangle a^\dagger_{nk} a_{nk}|i_p\rangle\) is the number of \((n,k)\) phonons \((k)\) is the phonon momentum and \(n\) an extra phonon label, containing the band index and polarization) in the \(|i_p\rangle\) state, \(\mathcal{N}_{nk} = \sum_{i_p} \langle N_{nk}(i_p)\rangle\) is the average population, and \(v_{nk} = \nabla_k \omega_{nk},\) with \(\omega_{nk}\) the dispersion of phonons, is the group velocity of phonons. \(\mathcal{C}\) is the “collision integral,” which captures in particular the scattering of phonons with other degrees of freedom \((Q \text{ fields}) whose coupling to the phonons is given by \(H'\) in Eq. (1) or more generally by Eq. (17)). In turn, using Born’s approximation, we have the following perturbative expansion of the scattering matrix:

\[
T_{1 \rightarrow t} = T_{\pm} = \langle f|H'|\bar{i}\rangle + \sum_n \frac{\langle f|H'|n\rangle \langle n|H'|\bar{i}\rangle}{E_i - E_n + i\eta} + \cdots,
\]

(8)

where the \(|i, f, n\rangle\) states are product states in the \(Q\) (index \(s\) and phonon (index \(p\)) Hilbert space, \(|g\rangle = |g_s\rangle |g_p\rangle\) for \(g = i, f, n\), and \(E_g\) is the energy of the unperturbed

\[\text{[This section continues with more detailed equations and discussions.]}\]
Hamiltonians of the $Q$ and phonons in state $g$, $\eta \to 0^+$ is a small regularization parameter. The expression Eq. (8) can be derived from time-dependent perturbation (scattering) theory, in which $\eta$ captures causality and the regularizability of $1/(E_i - E_f)$ in the case of a continuous energy spectrum, appropriate for scattering (unbounded) states which we are interested in [10].

The rate of transitions from state $i$ to state $f$ is obtained using Fermi’s golden rule,

$$\Gamma_{i \to f} = \frac{2\pi}{\hbar} |T_{i \to f}|^2 \delta(E_i - E_f).$$

(9)

Note that $\Gamma_{i \to f}$ is a transition rate in the full combined phonon-$Q$ system. This in turn determines the collision integral through the master equation

$$C_{nk} = \sum_{i_f f_p} \tilde{\Gamma}_{i_f f_p} (N_{nk}(f_p) - N_{nk}(i_f)) p_{i_f},$$

(10)

where $p_{i_f} = \sum_{i_f} p_i$, where $p_i = \frac{1}{Z} e^{-\beta E_i}$ is the canonical probability to find the system in state $i$, and $Z$ is the partition function of the two subsystems. Here

$$\tilde{\Gamma}_{i_f f_p} = \sum_{i_f i_s} \Gamma_{i_f \to i_s} p_{i_s}$$

is the transition rate between just phonon states, with $p_{i_s} = \frac{1}{Z} e^{-\beta E_{i_s}}$.

B. Discussion

The above approach is “semiclassical” in two respects. First, it ultimately treats phonons as quasiparticles within a Boltzmann equation. This is justified whenever the scattering rate is small compared to the energy of the particles. Second, we use the Fermi’s golden rule relation, Eq. (9), to determine the scattering rates. This approximation leads to slight differences from an exact calculation of the quantum rates, but preserves all symmetries and physical processes, and we expect it to capture all the key features of a fully quantum approach. We proceed with the T-matrix approach here which has the advantage of (relative) physical transparency, as every effect can be directly identified with a scattering process.

One can understand the need for effects beyond the first Born approximation entirely through the symmetries of the T-matrix. Specifically, since the time reversal (TR) operator is anti-unitary, and requires complex conjugation, one can see from Eq. (8) that under time reversal, $TR : T \to T^\dagger$ ($\eta \to -\eta$ under complex conjugation). Since TR invariance is sufficient to enforce a vanishing Hall effect, the hermiticity of $T$ is enough to guarantee a vanishing Hall effect. From Eq. (8), $T$ is indeed always hermitian within the first Born approximation, because $H'$ itself must be hermitian.

Finally, we note that we are focusing on collisional effects, i.e. on real transitions induced by interactions, rather than Berry phase contributions, which arise from entirely virtual transitions and manifest as modifications to the semiclassical equations of motion for phonons, e.g. an anomalous velocity. Formally, real transitions are captured within the collision integral on the right hand side of the Boltzmann equation [7], while Berry phase contributions enter the left hand side and in the definition of the currents. For phonons, our focus on collisions is justified by strong phase space constraints on the Berry curvature effects which are typical to acoustic bosonic modes. Specifically, as shown in Ref. [17], the Berry phase contributions are described by an emergent vector potential which at small momenta must by symmetry be at least second order in gradients, making it a formally “irrelevant” perturbation to the phonon Lagrangian, and strongly suppressing its effects at low temperature [18].

III. FORMAL EXPRESSIONS FOR THE THERMAL CONDUCTIVITY

A. Formal expressions

To solve Eq. (7), we expand $\overline{N}_{nk} = N^{eq}_{nk} + \delta \overline{N}_{nk}$ around the equilibrium distribution $N^{eq}_{nk}$, which solves Boltzmann’s equation at $\nabla T = 0$, keep terms up to linear order in $\delta \overline{N}_{nk}$ in the collision integral and for convenience separate the diagonal $D_{nk}$ and off-diagonal $M_{nk,n'k'}$ parts, i.e. we write the collision integral

$$C_{nk} = \sum_{n'k'} (\delta_{nn'}\delta_{kk'} D_{nk} + M_{nk,n'k'})\delta \overline{N}_{n'k'} + O(\delta \overline{N}^2),$$

(12)

where by definition $M_{nk,nk} = 0$. The equation $C_{nk} \{N^{eq}_{n'k} \} = 0$ —i.e. the collision integral is zero in equilibrium— should be considered the definition of the equilibrium densities $\{N^{eq}_{n'k} \}$ of the interacting phonons (see Appendix C3).

Using Fourier’s law,

$$j = -\kappa \cdot \nabla T = V^{-1} \sum_{nk} N_{nk} v_{nk} \omega_{nk},$$

(13)

and formally inverting the collision integral leads to the following expressions for the longitudinal $\kappa_L^{\mu\mu}$, and Hall $\kappa_L^{\mu\nu}$ conductivities (along the $\mu$ direction and in the $\mu\nu$ plane, respectively):

$$\kappa^{\mu\nu}_{L/H} = \frac{\hbar^2}{k_B T^2} \frac{1}{V} \sum_{nk} J^{\mu\nu}_{nk} K^{L/H}_{nk,n'k'} J^{\mu\nu}_{n'k'},$$

(14)

where $\nu = \mu$ for $\kappa_L$. Assuming $\sum_{n'k'} M_{nk,n'k'} \ll D_{nk}$, one can effectively invert the collision integral to obtain the kernels.
\[ K^L_{nk',k'} = \frac{e^{\beta \hbar \omega_{nk}}}{D_{nk}} \delta_{nk'} \delta_{k,k'} + \frac{e^{\beta \hbar (\omega_{nk} + \omega_{n'k'})/2}}{2D_{nk} D_{n'k'}} \left( \frac{\sinh(\beta \hbar \omega_{nk}/2)}{\sinh(\beta \hbar \omega_{n'k'}/2)} M_{nk,n'k'} + (nk \leftrightarrow n'k') \right), \]
\[ K^H_{nk',k'} = \frac{e^{\beta \hbar (\omega_{nk} + \omega_{n'k'})/2}}{2D_{nk} D_{n'k'}} \left( \frac{\sinh(\beta \hbar \omega_{nk}/2)}{\sinh(\beta \hbar \omega_{n'k'}/2)} M_{nk,n'k'} - (nk \leftrightarrow n'k') \right). \]  

Here we identified the equilibrium phonon current \( j^\mu_{nk} = \nabla_n \Phi_{nk,n} \), and made the “standard” approximation \( \nabla_r \Phi_{nk,n} \approx \nabla_r N_{nk,n} \), and looked for a stationary solution \( (\partial_t N_{nk,n} = 0) \) to Boltzmann’s equation. While the sign of \( \kappa_H \) depends on the details of the system (see later), the second law of thermodynamics imposes \( \kappa_L > 0 \). Considering Eq. (15), we therefore expect \( D_{nk} > 0 \).

Clearly, only contributions to \( K^L/H_{nk,n'k'} \) which are symmetric (resp. antisymmetric) in exchanging \( (nk \leftrightarrow n'k') \) contribute to \( \kappa_L \) (resp. \( \kappa_H \)). As a special case, the term diagonal in \( nk, n'k' \), being symmetric, does not contribute to the Hall conductivity. Below we will isolate the correlation functions of the \( Q \) operators which give anti-symmetric (in \( nk \leftrightarrow n'k' \)) contributions \( \frac{\sinh(\beta \hbar \omega_{nk}/2)}{\sinh(\beta \hbar \omega_{n'k'}/2)} M_{nk,n'k'} \), and hence contribute to \( \kappa_H \). These correspond to scattering processes which violate detailed balance.

### B. Model

To describe the interaction between the phonons and another degree of freedom, we introduce general coupling terms between phonon annihilation \( (\phi_{nk} \rightarrow \phi_{n'k'}) \) and creation \( (\phi_{nk} \rightarrow \phi_{n'k'}) \) operators \( a_{nk}^{(1)} \) and general, for now unspecified, fields \( Q_{[q]}^{[n_k,k]} \) which are operators acting in their own Hilbert space. In what follows we only consider the first two terms of the expansion with respect to phonon operators, i.e. we write the interaction hamiltonian as \( H' = H'_1 + H'_2 \), where

\[ H'_1 = \sum_{nk} \sum_{q=\pm} a_{nk}^q Q_{[n_k,k]}^q, \]
\[ H'_2 = \frac{1}{\sqrt{N_{uc}}} \sum_{nk \neq n'k'} \sum_{q,q'=\pm} a_{nk}^q a_{n'k'}^{q'} Q_{[n_k,n'}^{q'q}}, \]

and in the following, we consider Eq. (17) as a perturbative expansion with respect to a small parameter \( \lambda \), such that formally \( Q_{nk} = \lambda Q_{nk}^{q=\pm} \), \( Q_{nk,n'} \sim \lambda^2 \), etc. Note we consider generalizations of this model in Appendix D.

In the above expression we used \( a_{nk}^q = a_{nk}^{q\dagger} \) and \( a_{nk} = a_{nk}^{q\dagger} \). The hermiticity of \( H' \) imposes \( Q_{[n_k,k]}^{[n_k,k]} = Q_{[q]}^{[n_k,k]} \) and \( Q_{[n_k,k]}^{[n_k,k]} = Q_{[n_k,k]}^{[n_k,k]} \), and for many-phonon terms, we have \( Q_{[q]}^{[n_k,k]} = (Q_{[n_k,k]}^{[n_k,k]}) \dagger \). The single-phonon interaction terms, which may physically be seen as single-phonon scattering off the \( Q \) degrees of freedom, correspond in particular to a coupling of the \( Q \) operators to the strain tensor \( \epsilon_{\alpha\beta}(r) \),

\[ \epsilon_{\alpha\beta}(r) = \frac{i \hbar}{\sqrt{N_{uc}}} \sum_{kn} a_{nk}^q \left( k^\alpha \varepsilon_{\alpha\beta} + k^\beta \varepsilon_{\alpha\beta} \right) n_{kn} - a_{nk}^{q\dagger} n_{kn}, \]

where \( M_{uc} \) is the unit cell mass and \( \varepsilon_{nk} \) is the polarization vector of the \( |nk\rangle \) phonon. The two-phonon terms capture quadratic coupling of the lattice displacements to the electrons/spins, as is often considered for example in treatments of Raman scattering \[19,20]. A priori, the quadratic terms are much smaller than the linear ones, but the former may be important if they give rise to distinct effects or contribute at a lower order in perturbation theory than the linear ones.

### C. Scattering rates

#### 1. T-matrix elements

The transition matrix elements are \( T_{i\rightarrow \ell} = \sum_{j} T^{|i| \cdots |\ell|}_{i\rightarrow \ell} \), (the \( l_i \) represent which \( H[i] \) appear successively in \( T \), so that the number of \( l_i \) appearing in \( T^{|i| \cdots |\ell|}_{i\rightarrow \ell} \) is the order of the Born approximation used for that term), where

\[ T^{|1|}_{i\rightarrow \ell} = \sum_{nq} \sqrt{N^{|1|}_{nk,n'k'}} \langle f_s | Q^{|1|}_{n_k,k'} | i_s \rangle \, I(i_p \quad q \rangle f_p), \]
\[ T^{|2|}_{i\rightarrow \ell} = \frac{1}{\sqrt{N_{uc}}} \sum_{nq} \sqrt{N^{|2|}_{nk,n'k'}} \langle f_s | Q^{|2|}_{n_k,k'} | i_s \rangle \, I(i_p \quad q \rangle f_p), \]
\[ T^{|1|1|}_{i\rightarrow \ell} = \sum_{nk,n'k',q',q} \sqrt{N^{|1|1|}_{nk,n'k'}} \langle f_s | Q^{|1|1|}_{n_k,n'k'} | i_s \rangle \langle m_s | Q^{|1|1|}_{n_k,n'k'} | i_s \rangle \, I(i_p \quad q \rangle f_p), \]
\[ T^{|1|2|}_{i\rightarrow \ell} = \sum_{nk,n'k',q',q} \langle f_s | Q^{|1|2|}_{n_k,n'k'} | i_s \rangle \langle m_s | Q^{|1|2|}_{n_k,n'k'} | i_s \rangle \, I(i_p \quad q \rangle f_p), \]

and \( T^{|1|2|}_{i\rightarrow \ell} \) and \( T^{|1|1|1|}_{i\rightarrow \ell} \) are given in Appendices C.4 and C.5 respectively. Here, \( I(i_p \quad q \rangle f_p) \) (resp. \( I(i_p \quad q \rangle q \rangle f_p) \)
\( \rho \)) is a large product of delta functions which enforce \( N^{f}\_{n'k'} = N^{i}\_{n'k'} \), \(\forall n''k'' \neq n'k'\) (resp. \(\forall n''k'' \neq (nk,n'k')\)), and \(N^{f}_{nk} = N^{i}_{nk} + q\) (resp. \(N^{f}_{nk} = N^{i}_{nk} + q, N^{f}_{n'k'} = N^{i}_{n'k'} + q')\). Note that the cases where \(nk = n'k'\) require a formal correction. However, at any given order in the \(\lambda\) expansion, such terms are smaller than all others by a factor \(1/N_{uc}\), where \(N_{uc}\) is the number of unit cells, and therefore vanish in the thermodynamic limit. In what follows we thus use \(\sum_{nk,n'k'}\) and \(\sum_{nk,n'k'}\) exchangeably, unless we specify otherwise.

The scattering rate as given by Eq. (9), involves the squares of the elements of the total transition matrix (see Appendices C4, C5 for computational details). Its full expression to perturbative order \(\lambda^2\) is

\[
\Gamma_{i\rightarrow f} = \Gamma_{i\rightarrow f}^{\text{sc}} + \Gamma_{i\rightarrow f}^{q1} + \Gamma_{i\rightarrow f}^{q2},
\]

where

\[
\begin{align*}
\left[ \Gamma_{i\rightarrow f}^{\text{sc}} ; \Gamma_{i\rightarrow f}^{q2} ; \Gamma_{i\rightarrow f}^{q1} \right] = & \frac{2\pi}{\hbar} \delta(E_i - E_f) \\
\times & \left[ |T_{i\rightarrow f}^{(1)}|^2 + |T_{i\rightarrow f}^{(1,1)}|^2 + |T_{i\rightarrow f}^{(2)}|^2 ;
2\Re \left\{ (T_{i\rightarrow f}^{(1,1)})^\ast T_{i\rightarrow f}^{(2)} \right\} ;
2\Re \left\{ (T_{i\rightarrow f}^{(1,1)})^\ast T_{i\rightarrow f}^{(1)} + (T_{i\rightarrow f}^{(1,1,1)})^\ast T_{i\rightarrow f}^{(1)} \right\} \right].
\end{align*}
\]

This decomposition into three terms is discussed in Sec. IIIID3.

2. Collision matrix elements

Following Eq. (10), the scattering rates \(\Gamma_{i\rightarrow f}\) give access to the collision integral, i.e. to \(M_{nk,n'k'}\) and \(D_{nk}\). We decompose the latter as \(D_{nk} = D_{nk}^{(1)} + D_{nk}^{(2)} + D_{nk}\), where \(D^{(1)}\) and \(D^{(2)}\) are obtained in our perturbative expansion at orders \(\lambda^2\) and \(\lambda^4\), respectively, and \(D_{nk}\) encompasses contributions due to other scattering processes as well as higher-order terms of the expansion. In the following, we also use the “\([l]_i;[l']_f\)” superscripts to denote a term obtained from the product of \(T_{i\rightarrow f}^{[l]_i}\) and \(T_{i\rightarrow f}^{[l']_f}\) within \(|T_{i\rightarrow f}^{[l]}|^2\). For instance, at order \(\lambda^2\), we have \(D_{nk}^{(1)} = D_{nk}^{[1];[1]}\); details of the derivation are given in Appendix C2. At order \(\lambda^4\), the diagonal and off-diagonal contributions to the collision integral take the forms

\[
D_{nk}^{(2)} = -\frac{1}{N_{uc}} \sum_{n'k'} \sum_{q,q'} q \left( N^{eq}_{n'k'} + \frac{q+1}{2} \right) \left[ M_{nk,n'k'}^{qq'} \right],
\]

and

\[
M_{nk,n'k'} = \frac{1}{N_{uc}} \sum_{q,q'=\pm} q \left( N^{eq}_{nk} + \frac{q+1}{2} \right) \left[ M_{nk,n'k'}^{qq'} \right],
\]

respectively, where \(M_{nk,n'k'}^{qq'}\) is an off-diagonal scattering rate which involves two different phonon states \(|nk\) and \(|n'k'\). More precisely, \(M_{nk,n'k'}^{+,+}\) (resp. \(M_{nk,n'k'}^{-,-}\)) corresponds to scattering processes where two phonons are emitted (resp. absorbed), and \(M_{nk,n'k'}^{+,+}\) (resp. \(M_{nk,n'k'}^{-,-}\)) to processes where one phonon is emitted and one is absorbed. \(D_{nk}\) is the diagonal scattering rate, i.e. it is associated with variations in \(\delta N_{nk}\) only.

We will now decompose the \(M_{nk,n'k'}^{qq'}\) scattering rates into

\[
M_{nk,n'k'}^{qq'} = M_{nk,n'k'}^{\ominus;\ominus} + M_{nk,n'k'}^{\oplus;\ominus},
\]

where \(M_{nk,n'k'}^{\ominus;\ominus}/M_{nk,n'k'}^{\oplus;\ominus}\) satisfy detailed (\(\sigma = 1\)) or “anti-detailed” (\(\sigma = -1\)) balance equations

\[
M_{nk,n'k'}^{\ominus;\ominus} = \sigma e^{-\beta(q_{nk} + q_{n'k'})} M_{nk,n'k'}^{\ominus;\ominus}, \quad \sigma = \pm \ominus, \oplus.
\]

Physically, Eq. (27) expresses “microscopic” thermodynamic equilibrium between the process which takes \(\{N_{nk} \rightarrow N_{nk} + q, N_{n'k'} \rightarrow N_{n'k'} + q'\}\) to the “conjugate” process taking \(\{N_{nk} \rightarrow N_{nk} - q, N_{n'k'} \rightarrow N_{n'k'} - q'\}\), with \(q,q' = \pm 1\), leaving \(N_{nk} + N_{n'k'}\) unchanged for \(n'k' \notin \{nk,n'k'\}\). Note that this is different from time-reversal symmetry which provides a relation between the processes acting on \(\{ni,ki\}\) phonons to the same processes acting on \(\{ni,-ki\}\) phonons.

Moreover, since, by construction, the two-phonon scattering rates satisfy

\[
M_{nk,n'k'}^{\sigma;\sigma} = M_{n'k',nk}^{\sigma;\sigma},
\]

the following relations also hold:

\[
M_{nk,n'k'}^{\sigma;\sigma} = \sigma e^{\beta(\omega_{nk} - \omega_{n'k'})} M_{nk,n'k'}^{\sigma;\sigma},
\]

Together, these imply that there are only four independent such scattering rates between the \(|n,k\) and \(|n',k'\) phonons, namely \(M_{nk,n'k'}^{\ominus;\ominus}\) and \(M_{nk,n'k'}^{\ominus;\ominus}\) with \(\sigma = \ominus, \oplus\).

As discussed at length, the first Born approximation alone does not lead to a nonzero thermal Hall effect, neither do those scattering rates which satisfy detailed balance as the latter imposes thermal equilibrium between “left” and “right” scattering. We find the kernels \(K_{I/H}^{L}\) defined in Eqs. (15,16) in terms of the \(M\) scattering rates.
Incorporating the expression for $D$ in the denominators of $K_\text{L,H}$ provides an expansion up to $O(\Lambda^4)$ of the latter. We recover, as mentioned before, that the terms in $\mathbb{M}_s^\ominus$ do not contribute to $K_H^\text{D}$ (they satisfy detailed-balance). The “anti-detailed-balance” relations satisfied by the $\mathbb{M}_s^\ominus$ terms do not however prohibit their contribution to $K_L$. See Sec. IVB for a discussion. Inserting Eq. (31) into Eq. (32), and after some algebra, one obtains the result Eq. (6) for $\kappa_{\text{L,H}}$.

D. The collision integral as correlation functions

1. Terms at $O(\Lambda^2)$

The diagonal scattering rate $D_{nk}^{(1)}$, obtained by inserting $T_{\text{L}}^{[1]}$ into Eqs. (9–11), may now be cast into the form of a correlation function of $Q$ operators. To do so, we first enforce the energy conservation $\delta(E_f - E_i)$ by writing the latter as a time integral, i.e. use $\int_{-\infty}^{+\infty} dte^{\pm i\omega t} = 2\pi \delta(\omega)$; we then identify $A(t) = e^{+iHt}Ae^{-iHt}$ and use the identity $1 = \sum_f |f_i\rangle\langle f_i|$ with the Qs in the initial state to be in thermal equilibrium $p_{i_s} = Z_s^{-1}e^{-\beta E_{i_s}}$, summing over $|i_s\rangle$, identifying $\langle A\rangle_\beta = Z_s^{-1}\text{Tr}(e^{-\beta H}A)$, summing over final phononic states $f_p$ and taking the average over initial phononic states $i_p$, we obtain

$$D_{nk}^{(1)} = -\frac{1}{\hbar^2} \int dt e^{-i\omega_{nk}t} \langle [Q_{nk}(t), Q_{nk}(0)]_\beta \rangle.$$  

We now apply the same method to higher orders of the perturbative expansion.

2. Terms at $O(\Lambda^4)$

We use the following time integral representation for the denominators appearing at second and higher Born orders (using a regularized definition of the sign function, i.e. $\lim_{\eta \to 0} \text{sign}(t)e^{-\eta|t|} \to \text{sign}(t)$),

$$\frac{1}{x \mp i\eta} = \frac{1}{x} \mp i\pi \delta(x)$$

Using Eqs. (19) and Eq. (23), we find the explicit expressions for the semiclassical scattering rates as correlation functions of the $Q$ operators,

$$\mathbb{M}_{nk}^{\ominus,[2]:[2];qq'} = \frac{2}{\hbar^4} \int_{t_1,t_2} \langle Q_{nk,k'}(-t)Q_{nk,k'}(0),$$

$$\mathbb{M}_{nk}^{\ominus,[1,1]:[1,1];qq'} = \frac{2}{\hbar^4} N_{\text{ac}} \Re \int_{t_1,t_2} \langle \{Q_{nk}(-t-t_2),Q_{nk,k'}(-t-t_2)\},Q_{nk,k'}(t_1)\rangle,$$

$$\mathbb{M}_{nk}^{\ominus,[1,1]:[1,1];qq'} = \frac{1}{\hbar^4} N_{\text{ac}} \int_{t_1,t_2} \langle \{\cdot,\cdot\}\{\cdot,\cdot\} - \{\cdot,\{\cdot,\cdot\}\},$$

where we use the shorthand notation

$$[A(t_a), B(t_b)] = \text{sign}(t_b - t_a)|A(t_a), B(t_b)|,$$

and $\{f_{\text{L}}(t_j), j = 1, \ldots, l$, denotes the set of $1 + l$ Fourier transforms evaluated once at $\sum_k\omega_{nk} + q'\omega_{nk'}$ and $l$ times at $\Delta_{nk}^{n'k'} = \sum_k\omega_{nk} - q'\omega_{nk'}$, i.e.

$$\int_{t_{\text{L}}} d\hat{t}_1, d\hat{t}_2 e^{i\Delta_{nk}^{n'k'}(t_1+\ldots+t_l)}.$$
The above terms capture all contributions to the collision integral arising from the Born expansion of the transition amplitude, up to perturbative order $\lambda$. This gives, correspondingly, physical processes in the collision integral which contribute up to $O(\lambda^4)$. 

In Eq. (22), while $\Gamma_{1\rightarrow f}^{\Omega}$ and $\Gamma_{1\rightarrow f}^{Q}$ are “two-phonon” terms, the contribution from $\Gamma_{2\rightarrow f}^{\Omega}$ is a “one-phonon” term, i.e. one where the initial $i$ and final $f$ states differ by only one phonon $|nk\rangle$. Physically, this contributes to processes which create or annihilate a single phonon, in contrast with the $O(\lambda^4)$ processes described so far, which create/annihilate two phonons with different quantum numbers. Because the single phonon process is physically distinct from the two-phonon ones, we expect that it is independent from the latter in the sense that the set of all the $O(\lambda^4)$ single-phonon processes satisfies independently all physical constraints such as symmetries and conservation laws. Hence omitting these contributions is a “conserving approximation” in the traditional sense [21], and we will proceed with this omission for the most part in the following. We however include formal expressions for these terms in the appendices.

The remaining contributions in Eq. (22) are “two-phonon” terms, i.e. terms in which the initial $i$ and final $f$ states differ by two phonons $|nk\rangle, |n'k'\rangle$. The two-phonon, $O(\lambda^4)$, contributions to the $\mathcal{W}$ scattering rates thus read

\[
\mathcal{W}_{nk, n'k'}^{\Omega, qq'} = \mathcal{W}_{nk, n'k'}^{\Omega, [1,1];[2,qq']} + \mathcal{W}_{nk, n'k'}^{\Omega, [1,1];[1,qq']},
\]

\[
\mathcal{W}_{nk, n'k'}^{Q, qq'} = \mathcal{W}_{nk, n'k'}^{Q, [2,2];[2,qq']} + \mathcal{W}_{nk, n'k'}^{Q, [1,1];[2,qq']} + \mathcal{W}_{nk, n'k'}^{Q, [1,1];[1,qq']},
\]

Another physical distinction between the contributions in Eq. (22) can be made according to the “quantum” or semiclassical, nature of the terms. The one-phonon $\Gamma_{1\rightarrow f}^{\Omega}$ and two-phonon $\Gamma_{2\rightarrow f}^{Q}$ terms in Eq. (22) are “quantum” in the sense that the physical process corresponding to each contribution therein is an interference term between distinct scattering channels. In particular, in a “quantum” term, the number of scattering events in the two channels are different. On the contrary, each contribution in $\Gamma_{1\rightarrow f}^{\Omega}$ is the probability amplitude of one given scattering channel, corresponding physically to the probability amplitude of a given scattering process, and in this respect is truly semiclassical. As a semiclassical approximation, we will neglect “quantum” contributions in the following; formal expressions for these terms are nonetheless included in the appendices. The only “semiclassical” contributions, up to $O(\lambda^4)$, to the collision integral are from the scattering rates shown in Eq. (34).

Upon applying our results to the case of a staggered antiferromagnet in Sec. V we focus on the lowest-order contributions to $K_{nk, n'k'}^{\Omega}$ and $K_{nk, n'k'}^{Q}$, which come from $D_{nk}^{(1)}$ and $D_{nk}^{(2);qq'}$, respectively. Therefore, in Sec. V we consider only the lowest-order semiclassical contributions $D_{nk} \approx D_{nk}^{(1)} + \mathcal{W}_{nk, n'k'}^{\Omega, qq'}$ and $\mathcal{W}_{nk, n'k'}^{Q, qq'} \approx \mathcal{W}_{nk, n'k'}^{[1,1];[1,1],qq'}$.

To leading order, the longitudinal conductivity is controlled by the diagonal scattering rate, whose main contribution occurs at order $\lambda^2$. The latter is given as the first term in Eq. (2), and is shown again in Eq. (22). It is related to the Fourier transform of the commutator of two $Q_{nk}$ operators at unequal times. The commutator structure identifies the phonon scattering rate $D_{nk}^{(1)}$ with the spectral function of the $Q_{nk}$ field at energy $\omega_{nk}$, i.e. it captures the proportion of the energy density contained in the $Q_{nk}$ field located at $\omega_{nk}$, as expected from (lowest-order) linear response [22, 23].

As mentioned above, the first Born order transition matrices are hermitian. At second Born’s order, the advanced/retarded Green’s function, $1/(E_i/\hbar - E_n \pm i\eta)$, appearing in $T_{1\rightarrow f}$, splits into on-shell and off-shell contributions, so that the scattering rate $\propto |T_{1\rightarrow f}|^2$ then involves the product of two on-shell or two off-shell contributions, as well as the products of one on-shell and one off-shell one. Because of complex conjugation of one term upon taking the square modulus of the $T$ matrix, the scattering rates which involve either two on-shell or two off-shell contributions are blind to the sign of $\pm i\eta$, i.e. to the advanced or retarded nature of the process, and enforce a detailed-balance relation, Eq. (27) with $\sigma = \oplus$. Therefore, the only scattering rates which can contribute to the Hall conductivity are those involving one on-shell (imaginary part) and one off-shell (real part) scattering event, which translates here into the product of a commutator and an anticommutator, Eq. (35).

IV. RELATIONS AND SYMMETRIES

In this section, we explore in more detail some physical relations verified by the scattering rates defined above, and their possible consequences on the longitudinal and Hall conductivities.

A. Time-reversal symmetry: reversal of the momenta

We investigate the implications of time-reversal (TR) invariance on our results. In particular, we check explicitly that the Hall conductivity vanishes in a TR-symmetric system. It is important to note that, in a time-reversal invariant system, the scattering rates are a priori not time-reversal invariant themselves.

We denote with $\tilde{Q}$ and $\tilde{a}$ the time-reversed of operator $Q$ and of state $|n\rangle$, respectively. Then, because of the antiunitarity of the time-reversal operator, for any states $n, m$ and any operator $Q$, we have $(\tilde{a}^\dagger Q^\dagger |\tilde{m}\rangle = |m\rangle Q |n\rangle$. Moreover, it is possible to choose a polarization index $n$ invariant under TR, whence $a^\dagger_{nk} = a^\dagger_{n,-k}$.
| Q operators | $Q^P_{n-k} = Q^n_{n-k}$ |
|------------|------------------------|
| scattering rates | $Q^q q'_{n-k'} = Q^q q'_{n-k,n'-k'}$ |
| conjugate process | $D^1_{n,k} = D^{(1)}_{n-k}$ |
| kernels | $\begin{array}{c}
\mathbb{M}^{q q'}_{n-k,n'-k'} = \sigma \mathbb{M}^{q q'}_{n-k,n'-k'} \\
\mathbb{M}^{q q'}_{n-k,n'-k'} = \sigma \mathbb{M}^{q q'}_{n-k,n'-k'}
\end{array}$ |
| conductivities | $\kappa_H = 0$ |

TABLE I: Relations which hold true in the presence of time-reversal symmetry. The phonon operator relation $a^\dagger_{n,k} = a^q_{n,-k}$ holds true even when no time-reversal symmetry is present. See text for definitions and justifications.

Let us now consider what happens in a time-reversal-invariant system. In that case, the hamiltonian $H[1] = \sum_{n,k} Q^q q'_{n-k}$ must be TR-invariant, so that $Q^q q'_{n-k} = Q^q q'_{n-k}$. Similarly, TR-invariance of $H[2]$ (defined in Eq. (17)) entails $Q^q q'_{n-k,n'-k'} = Q^q q'_{n-k,n'-k'}$.

1. Consequences for the scattering rates.

Following the same steps as those sketched in Sec. [D.1] and using the fact that $E_{\bar{n}} = E_{\bar{n}}$ for any state $\bar{m}$ of a TR-symmetric system, we can show explicitly that, in a time-reversal-invariant system, the following relations for the scattering rates exist:

$$
D^{(1)}_{n,k} = D^{(1)}_{n-k} \\
\mathbb{M}^{q q'}_{n-k,n'-k'} = \sigma \mathbb{M}^{q q'}_{n-k,n'-k'} \\
\mathbb{M}^{q q'}_{n-k,n'-k'} = \sigma \mathbb{M}^{q q'}_{n-k,n'-k'}
$$

(39)

(40)

The $\sigma$ sign in the second relation can be understood as arising from two facts: (1) schematically, $\mathbb{M}^\sigma \sim E_{\bar{m}} \mathbb{M}^\sigma$ (resp. $\mathbb{M}^\sigma$) expressed as an integral, Eqs. [34–36], contains an even (resp. odd) number of sign functions; and (2) an effect of time-reversal on the $T$-matrix is to exchange denominators $E_{\bar{m}} + i\eta \rightarrow E_{\bar{m}} - i\eta$ (see Sec. [D.4] for an interpretation of the $+i\eta$ regularization).

2. Relation to detailed balance.

The decomposition of the scattering rate $\mathbb{M}^{q q'}_{n-k,n'-k'} = \sum \mathbb{M}^{q q'}_{n-k,n'-k'}$ into odd and even terms under the “conjugation” (in the sense of detailed balance, i.e. thermodynamic equilibrium) of the associated scattering processes, is also that of its decomposition into terms, odd and even under the inversion of momentum, in the presence of time-reversal symmetry. Indeed, if a scattering process $S = (\frac{q,n}{q',n'})$ transfers an energy $\delta E_{\bar{m}}(S) = q\omega_{n,k} + q'\omega_{n',k'}$ to the phonon system, (anti-)detailed balance with the “conjugate process” $pc[S] = (\frac{-q,n}{q',n'})$ reads $\mathbb{M}^\sigma pc[S] = e^{-\delta E_{\bar{m}}(S)} \mathbb{M}^\sigma(S)$. Meanwhile, in the presence of time-reversal symmetry, the momentum-reversal symmetry reads $\mathbb{M}^\sigma mr[S] = \sigma \mathbb{M}^\sigma(S)$, for the “momentum-reversed” process $mr[S] = (\frac{q,n}{q',n'})$. In other words, in a time-reversal invariant system, the scattering rate associated with the process “conjugate” of a given process $S$ coincides (up to a Boltzmann weight) with that of its momentum-reversed one:

$$
\frac{\mathbb{M}^\sigma(mr[S])}{\mathbb{M}^\sigma(S)} = e^{-\delta E_{\bar{m}}(S)} \frac{\mathbb{M}^\sigma(pc[S])}{\mathbb{M}^\sigma(S)} = \sigma.
$$

(41)

Hence, while $\sigma$ was defined as signature of the behavior of the scattering rates under “process conjugation,” it is also that of momentum reversal in a time-reversal invariant system.

3. Consequences for the kernels.

How is this reflected in the kernels $K^L, K^H$? Because the relation $v_{n,k} = \nabla_{\bar{k}} \omega_{n,k} = -v_{n,-k}$ holds, only that component of $K^L/H$ which is even upon reversal of the momenta, $k^{(i)} \leftrightarrow -k^{(i)}$, has a non-vanishing contribution to the sum Eq. (14). A first consequence of this is that, in a TR-invariant system, the identity

$$
K^H_{n-k,n'-k'} = -K^H_{n-k,n'-k'}
$$

(42)

entails $\kappa_H = 0$ — as per Onsager’s reciprocity relations stating that $\kappa_H$ is TR-odd. Note that $K^L$ in Eq. (30) involves both $\mathbb{M}^\odot$ and $\mathbb{M}^\otimes$. Therefore, there is no analog to Eq. (12) for $K^L$. However, in a TR-invariant system, the $\mathbb{M}^\odot$ term in $K^L$ does not contribute to $\kappa_L$ — this is consistent with the Onsager-Casimir relations which state that $\kappa_L$ is TR-even.

This indeed reflects the previous discussion as follows: when time reversal is preserved, TR-even $\kappa_L$ gets contributions solely from “detailed-balance-even” and TR-even $\mathbb{M}^\odot$. On the other hand TR-odd $\kappa_H$ gets contributions solely from “detailed-balance-odd” and TR-odd $\mathbb{M}^\otimes$. Since the system is actually TR-even, $\kappa_H$ vanishes.

B. Point-group symmetries

Here we provide some sufficient (but non-necessary) conditions on $K^H_{n-k,n'}$ under which the Hall conductivity vanishes.
1. Onsager relations

From Fourier’s law \( j^\mu = -\kappa^\mu v \nabla_u T \), the Onsager relations provide general constraints on the \( \kappa^\mu v \) coefficients, and in turn on its Hall component \( \kappa^\mu v \). In Table II we look at the \( D_{4h} = D_4 \times Z_2 \) point group—the largest tetragonal point group—with associated axes aligned with the orthogonal basis \((\mu, \nu, \rho)\) (\( \mu \nu \) is the basal plane and \( \rho \) the transverse direction. We can see that if the system is invariant under any one of the transformations \( g \in D_{4h} \) which are odd under the \( A_{2g} \) representation (i.e. \( C_2', C_2'' \), \( \sigma_v \), \( \sigma_d \)), the Hall conductivity must vanish.

| \( A_{2g} \) | 1 | 1 | 1 | -1 | -1 | 1 | 1 | 1 | -1 | -1 |
|---|---|---|---|---|---|---|---|---|---|---|
| \( \mu \) | \( \mu \) | \( \nu \) | -\( \mu \) | ±\( \mu \) | ±\( \nu \) | -\( \nu \) | \( \mu \) | ±\( \nu \) | ±\( \mu \) | ±\( \nu \) |
| \( \nu \) | \( \nu \) | -\( \nu \) | ±\( \nu \) | ±\( \mu \) | -\( \mu \) | ±\( \nu \) | -\( \mu \) | ±\( \nu \) | ±\( \nu \) | ±\( \mu \) |
| \( \rho \) | \( \rho \) | \( \rho \) | -\( \rho \) | -\( \rho \) | -\( \rho \) | -\( \rho \) | -\( \rho \) | -\( \rho \) | -\( \rho \) | -\( \rho \) |
| \( k^\mu v \) | \( k^\mu v \) | -\( k^\mu v \) | \( k^\mu v \) | -\( k^\mu v \) | \( k^\mu v \) | -\( k^\mu v \) | \( k^\mu v \) | -\( k^\mu v \) | \( k^\mu v \) | -\( k^\mu v \) |
| \( K^\mu v H \) | \( K^\mu v H \) | -\( K^\mu v H \) | -\( K^\mu v H \) | \( K^\mu v H \) | \( K^\mu v H \) | \( K^\mu v H \) | \( K^\mu v H \) | -\( K^\mu v H \) | -\( K^\mu v H \) |
| \( \text{cat} \) | (a) | (d) | (a) | (b) | (c) | (a) | (a) | (b) | (a) | (b) | (c) |

TABLE II: Elements of the \( D_{4h} \) point group aligned along the \((\mu, \nu, \rho)\) basis (with \( \mu \nu \) the basal plane), their characters in the \( A_{2g} \) irrep (also labeled \( \Gamma_+ \)), and transformations of \( \mu, \nu, \rho, k^\mu v, k^\mu v H \). The lines for \( k^\mu v \) and \( k^\mu v H \) hold true when the system is invariant under the corresponding \( D_{4h} \) operation (aligned with the \( \mu \nu \rho \) basis). The last line is the “category” (\( \text{cat} \)) to which the operation belongs, as defined in Sec. [IVB2]. Here Id is the identity; \( C_4 \) is the 4/2 rotation around the \( \rho \) axis; \( C_2, C_2' \) and \( C_2'' \) are \( \pi \) rotations through the \( \rho \) axis, \( \mu \) axis or \( \nu \) axis, and in-plane directions bisecting the \( \mu \nu \) plane, respectively; inv is inversion, \( S_4 \) are \( \pi/2 \) rotations around the \( \rho \) axis followed by a reflection through the basal \( \mu \nu \) plane; \( \sigma_h \), \( \sigma_v \), \( \sigma_d \) are reflections through the \( \mu \nu \) plane, while a plane containing \( \mu \) or \( \nu \) and the \( \rho \) direction, and through a plane containing the \( \rho \) direction and one bisecting the \( \mu \nu \) directions, respectively.

2. Symmetry relations on \( K^H \)

We now turn to relations specific to the scattering situation, i.e. we analyze under which conditions on \( K^H_{n k v k' c} \), it befalls that \( k^\mu v H = 0 \). We start with the expression of \( k^\mu v H \) as a momentum integral, Eq. (14), i.e. \( k^\mu v H = \sum_{n k v k' c} J^H_{n k v k' c} K^H_{n k v k' c} \), and recall \( J^H_{n k} = N_{n k} \omega_{n k} \partial_{\omega_{n k}} \omega_{n k} \).

If the phonon is invariant under a unitary transformation \( g \), then \( \omega_{n k} \) is also invariant under this transformation. In turn only \( \mu \) in \( J^H_{n k} \) transforms nontrivially under \( g \). Therefore:

- If the phonon system is invariant under an operation \( g \in D_{4h} \) which leaves the \( \mu, \nu \) axes invariant, i.e. \( g = C_2, C_2', \sigma_h, \sigma_v \), and if one of the two following conditions, \((a)\) under \( g \) the \( \mu \nu \) product is even (i.e. \( g = C_2, \sigma_h \) and \( K^H_{n k v k' c} \) is odd, \(b\) under \( g \) the \( \mu \nu \) product is odd (i.e. \( g = C_2', \sigma_v \) and \( K^H_{n k v k' c} \) is even), is satisfied, then it follows that \( k^\mu v H \neq 0 \).

- Besides, recalling that by construction \( K^H_{n k v k' c} = -K^H_{k' k n c} \), if the system is invariant under an operation \( g \in D_{4h} \) which exchanges the \( \mu, \nu \) axes, i.e. \( g = C_4, C_2'' \), \( \sigma_d \), and if one of the two following conditions, \((c)\) under \( g \) the \( \mu \nu \) product is even (i.e. \( g = C_2', \sigma_d \) and \( K^H_{n k v k' c} \) is even, \(d\) under \( g \) the \( \mu \nu \) product is odd (i.e. \( g = C_4, S_4 \) and \( K^H_{n k v k' c} \) is odd, is satisfied, then it follows that \( k^\mu v H = 0 \).

In terms of the behavior of \( K^H_{n k v k' c} \), this analysis reduces to: if \( g \in D_{4h} \) is a symmetry of the phonon system, and if \( g : K^H_{n k v k' c} \rightarrow -\chi_{A_{2g}}(g) K^H_{n k v k' c} \), where \( \chi_{A_{2g}}(g) \) is the character of \( g \) in the \( A_{2g} \) representation of the \( D_{4h} \) point group, then \( k^\mu v H = 0 \). We emphasize that this analysis holds if the transformation \( g \) is a symmetry of the phonon system, and whether or not \( g \) is a symmetry of the whole system. For example, we will show explicitly in Sec. [VC4] that there are cases where, under TR or \( \sigma_d \) the system is not invariant, but the kernel \( K^H_{n k v k' c} \) and the phonon system are, and so \( \kappa_H = 0 \).

Finally, note that the above analysis goes beyond the general predictions from Onsager, which tell us that \( \kappa_H \) vanishes in the presence of some symmetries of the whole system, namely \( C_2, C_2', \sigma_v \) or \( \sigma_d \) (as well as time-reversal discussed in the previous subsection). Here, not only do we establish relations for the other symmetries in \( D_{4h} \) (as symmetries of the phonon subsystem only), we also show in which way \( \kappa_H \) vanishes, by inspecting the behavior of the kernels \( K^H_{n k v k' c} \) under those symmetry transformations. In turn, this may for example allow to gather information about the system—about \( K^H_{n k v k' c} \)—from the (non-)cancellation of \( \kappa_H \).

V. APPLICATION TO AN ORDERED MAGNET

We now turn to an application of these general results. Here, we keep only the lowest-order terms in the expressions derived above, as described in Sec. [III D4]. We consider an ordered magnetic system, which we take to be a spin-orbit coupled Néel antiferromagnet with tetragonal symmetry. For concreteness, we treat the magnetism as purely two-dimensional, i.e. the full spin+phonon system is described by a stack of two dimensional antiferromagnets embedded into the three-dimension solid, so that in particular, we take, when going from the lattice to the
continuum limit
\[ \sum_{\mathbf{r}} \rightarrow \frac{1}{\mathbf{a}^2} \sum_{\mathbf{z}} \int d^2 x, \quad \sum_{\mathbf{k}} \rightarrow (\frac{2\pi}{\mathbf{a}})^2 \sum_{k_z} \int d^2 k, \] (43)
where \( \mathbf{a} \) is the in-plane lattice spacing.

### A. Magnon dynamics

#### 1. Low-energy field-theoretical description

We consider a Néel antiferromagnet with a two-site magnetic unit cell, more precisely a bipartite lattice of spins such that the classical ground state is ordered in an antiferromagnetic configuration, with a local moment \( \mu_0 \) oriented in the direction \( \mathbf{n} \), i.e. \( \mathbf{n} \) is the Néel vector which has unit length in the ordered state at zero field. Within standard spin-wave theory, \( \mu_0 = S \) with \( S \) the spin value. For concreteness, we will choose the ordering axis at zero field to be aligned along the \( \mathbf{u}_x \) axis (the set \( \{ \mathbf{u}_x, \mathbf{u}_y, \mathbf{u}_z \} \) is an orthonormal cartesian basis)—the results of this subsection hold regardless of this choice.

A general low energy spin configuration is described by two continuum fields: the aforementioned Néel vector \( \mathbf{n}(\mathbf{r}) \) and a uniform magnetization density \( \mathbf{m}(\mathbf{r}) \), such that

\[ S_r = (-1)^r \mu_0 \mathbf{n}(\mathbf{r}) + \mathbf{a}^2 \mathbf{m}(\mathbf{r}). \] (44)

where \( (-1)^r \) is a sign which alternates between neighboring sites (recall we are considering a Néel antiferromagnet), and both continuum fields are assumed to be slowly-varying relative to the lattice spacing. Here \( \mathbf{a} \) is the 2d lattice spacing. We will assume the non-linear sigma model constraint that the spin length is fixed to \( \mu_0 \), which implies that

\[ |\mathbf{n}|^2 + \frac{\mathbf{a}^2}{\mu_0^2} |\mathbf{m}|^2 = 1, \quad \mathbf{m} \cdot \mathbf{n} = 0. \] (45)

The spin wave expansion consists of expanding these fields around the zero field ordered state, i.e. \( \mathbf{n}_{\text{ord}} = \mathbf{u}_x, \mathbf{m}_{\text{ord}} = 0 \). To linear order around this state, we take \( \mathbf{n} = \mathbf{u}_x + \mathbf{n} \) and \( \mathbf{m} = \mathbf{m} \), where \( n_x = m_x = 0 \), leaving the remaining degrees of freedom \( n_y, n_z, m_y, m_z \). In terms of the spins, this gives

\[ S_r = (-1)^r \mu_0 \mathbf{u}_x + \sum_{a=y,z} ((-1)^r \mu_0 n_a(\mathbf{r}) + \mathbf{a}^2 m_a(\mathbf{r})) \mathbf{u}_a. \] (46)

Because the local moment along the \( \mathbf{u}_x \) axis is non-zero, the low energy fields satisfy the commutation relations

\[ [m_y(\mathbf{r}), n_z(\mathbf{r}')] = -[m_z(\mathbf{r}), n_y(\mathbf{r}')] = -i\delta(\mathbf{r} - \mathbf{r}'). \]

The low energy continuum Hamiltonian density for these fields is

\[ \mathcal{H}_{\text{NLS}} = \frac{\rho}{2} \left( |\nabla m_y|^2 + |\nabla n_z|^2 \right) + \frac{1}{2\chi} (m_y^2 + m_z^2) + \sum_{a,b=y,z} \frac{\Gamma_{ab}}{2} n_a n_b, \] (47)

where \( \rho \) is the spin stiffness constant, \( \chi \) is the spin susceptibility, \( \nabla = (\partial_x, \partial_y) \) denotes the in-plane gradient, and the \( \Gamma_{ab} \) are anisotropy coefficients which open a small spin wave gap (see App. F3). For an approximately Heisenberg system with isotropic exchange constant \( J \), we have within spin wave theory that \( \chi^{-1} \approx 4J\mathbf{a}^2, \rho \approx 2J\mu_0^2 \), while \( \Gamma_{ab} \) are determined by exchange anisotropies. The choice to normalize \( \mathbf{m} \) as a density while keeping \( \mathbf{n} \) dimensionless ensures that \( m_y, z \) fields are just the canonical momenta conjugate to the \( n, y, z \) fields, and hence Eq. (47) is just a Hamiltonian density of two free scalar boson fields.

The above description is appropriate to describe the ordered phase of the antiferromagnet, for any value of the spin, provided temperature is low compared to the Néel temperature and any applied magnetic fields are small compared to the saturation field. These conditions are well-satisfied in practice in experiments on many antiferromagnets. Specifically we will be interested in the case with an applied magnetic field perpendicular to the axis of the Néel vector (e.g. along \( z \) or \( y \), given the choice in Eq. (46)). In general the field induces a non-zero uniform magnetization along its direction, e.g. for a \( z \)-axis field \( \langle m_z \rangle \neq 0 \). Such a “spin flip” configuration is favorable for an antiferromagnet in a field.

#### 2. Symmetry considerations

Two symmetries clarify the calculations and provide physical insight. The first is the macroscopic time-reversal symmetry of the zero field state, which is what makes it an anti-ferromagnet. Specifically, the system in zero field is invariant under the combination of time-reversal symmetry \( \mathcal{T} = \mathcal{TR} \) and a translation \( T \). Under this operation, we see that the continuum fields transform according to

\[ \mathcal{T} = \mathcal{TR} \times T : \quad m \rightarrow -m, \quad n \rightarrow n. \] (48)

The presence of a staggered magnetization (with any orientation) does not break this symmetry, but a uniform magnetization does. Note that the effective quadratic low energy Hamiltonian, Eq. (47), is invariant under this symmetry. This is true even at non-zero fields, because the low energy Hamiltonian is quadratic. Thus effects of time-reversal symmetry breaking will become evident in terms beyond this form, notably in anharmonic corrections, and in the spin-lattice coupling itself. Specifically, we see that time-reversal symmetry will be effectively broken only by terms involving an odd number of powers of the \( m_a \) fields.

The second important symmetry is one which may be preserved not only by the underlying exchange Hamiltonian and crystal structure, but also by the applied field and the spontaneous ordered moments. In particular, the latter breaks the original translational symmetry of the square lattice by a single lattice spacing. However, a symmetry may be retained under such a simultaneous
translation composed with a $C_2$ spin rotation around the field axis. In the presence of spin-orbit coupling, generically the spin rotation must be accompanied by a spatial rotation, and the full combined operation is in fact nothing but a $C_2$ rotation about an axis passing through the midpoint of a bond of the square lattice. This of course requires the $C_2$ rotation in question to be part of the lattice point group. In our problem, this is true when the field is along $z$ or $y$ (but not for a general orientation in the $yz$ plane).

This odd symmetry is important for simplifying the magnon interactions. In particular, if the field axis is along $z$, then we see that $m_z$ and $n_y$ are both even under this operation, while $m_y$ and $n_z$ are odd under it (and vice versa if the field is along $y$). Note that the fields within a canonically conjugate pair transform the same way under this symmetry. We take advantage of these facts in the following. In particular, only $\Gamma_0 = \Gamma_{yy}$ and $\Gamma_1 = \Gamma_{zz}$ do not vanish a priori, which ensures that the two valleys ($\ell = 0, 1$) are exactly decoupled.

3. External magnetic field

At the lowest order, an applied external magnetic field $\mathbf{h}$ couples solely to the $\mathbf{m}$ field; this is already taken into account in Eq. (47) where the $\mathbf{m}$ fields can acquire a (static) nonzero expectation value due to the spin alignment with the field.

Meanwhile, at higher orders the magnetic field also couples to the $\mathbf{n}$ field; the main contribution comes from the square, isotropic coupling $(\mathbf{n} \cdot \mathbf{h})^2$. Due to the $C_2$ symmetry around the field axis ($y$ or $z$), and since first-order terms of the form $h_n n_b$ are forbidden by translational symmetry, this results in an additional term

$$ H_{\text{field}} = \frac{\chi}{2} \sum_{a=y,z} h_a^2 n_a^2. \tag{49} $$

Note this form is valid only when the field is along the $y$ or $z$ axis, not at other angles in the $y-z$ plane (which would violate the $C_2$ symmetry). The prefactor $\chi/2$ is fixed to match the results obtained from microscopic calculations in Ref. [25], and we provide an alternative derivation in App. [F.3] as well as a more detailed derivation of the full form of the gap from a microscopic XXZ exchange model plus a Zeeman coupling to the field in App. [F.3]

4. Diagonalization

We proceed to diagonalize Eq. (17), supplemented by Eq. (49) following the discussion in Sec. [VA3] by introducing creation and annihilation operators in the standard way for free fields. We use the Fourier convention $\phi_k = \frac{1}{\sqrt{V}} \int d\mathbf{x} \phi(\mathbf{x})e^{-i\mathbf{k}\cdot\mathbf{x}}$ for any continuum field $\phi$, where $V$ is the volume of the system. Then

$$ m_k^y = \frac{\sqrt{\Omega_k^0}}{2} (b_{-k,0} + b_{k,0}^\dagger), $$
$$ n_k^z = \frac{1}{\sqrt{2\chi_{1k,0}}} (b_{-k,0} - b_{k,0}^\dagger), $$
$$ m_k^z = \frac{\sqrt{\Omega_k^1}}{2} (b_{-k,1} + b_{k,1}^\dagger), $$
$$ n_k^y = -\frac{1}{\sqrt{2\chi_{1k,1}}} (b_{-k,1} - b_{k,1}^\dagger), \tag{50} $$

where

$$ \Omega_{k,\ell} = \sqrt{v_m^2 k^2 + \Delta_\ell^2}, \tag{51} $$

with $v_m = \sqrt{\rho/\chi}$. The magnon gaps depend on the applied (transverse) magnetic field in the form

$$ \Delta_\ell = \sqrt{\Gamma_\ell/\chi + h_\ell^2}, \tag{52} $$

with valley index $\ell = 0, 1$ and where we set $h_0 = h_y$ and $h_1 = h_z$. This reflects the explicit breaking of $O(3)$ rotational symmetry of the order parameter $\mathbf{n}$ by the transverse field. With these definitions, we obtain

$$ H_{\text{NLS}} + H_{\text{field}} = \sum_{\ell} \sum_{k} \Omega_{k,\ell} b_{k,\ell}^\dagger b_{k,\ell}. \tag{53} $$

The $b, b^\dagger$ fields with index $\ell = 0$ have opposite $C_2$ eigenvalue to those with $\ell = 1$. This guarantees that all terms preserving $C_2$ symmetry must conserve the two boson flavors modulo 2.

B. Formal couplings

1. Definitions

In general we can expand the operator $Q_{nk}$, which couples to a single phonon, in powers of the magnon operators,

$$ Q_{nk}^q = \sum_{\ell,q,z} A_{k \ell,q,z} e^{ik_z z} b_{\ell,1,k,z}^q + \sum_{\ell,q,z} \frac{1}{\sqrt{N_{\text{uc}}}} \sum_{p,\ell',q'} B_{k,p,\ell,q,z} e^{ip_{\ell',z} z} b_{\ell',1,p,z}^{q'} b_{\ell,1,k,z}^{q}. \tag{54} $$

Note that while the phonons are three-dimensional excitations, and hence have a three-dimensional momentum $\mathbf{k}$, the spin operators (and hence magnons) only have two dimensional momenta. We will make use of the following: $\mathbf{k} = k + k_z \mathbf{\hat{z}}$, where $k$ is the projection of $\mathbf{k}$ onto the $k_z = 0$ plane and $\mathbf{\hat{z}}$ is the unit vector along $z$. A phonon is coupled to the sum of spin operators in all layers—we have here introduced the explicit label $z$ for the layer. Because the spins in different layers are completely uncorrelated, there are however no cross-terms involving $b$.
operators from different layers, and in correlation functions the sums over \( z \) will collapse to independent correlators within each layer, which are all identical to one another. When possible, we will therefore take \( z = 0 \) and suppress this index.

The naïve leading term in Eq. (54) is the single magnon one \( A \), linear in \( b_{\ell,k} \) and \( b_{\ell,k}^\dagger \) operators (notations defined below). This results in a quadratic mixing term in the Hamiltonian, hybridizing phonons and magnons. Being quadratic, it is trivially diagonalized, and has been considered by several authors. Generally, such coupling has little effect except when it is resonant, i.e. near a crossing point of the decoupled magnon and phonon bands. Since such a crossing is highly constrained by momentum and energy matching, it occurs in a narrow region of phase space, if at all, and is likely to be unimportant for transport. It in any case does not give rise to scattering, the focus of this work. We therefore henceforth neglect the \( A \) contribution.

Non-trivial scattering processes arise from the second order term in the magnon field expansion of \( Q_{nk} \), parametrized by \( B \). Here as elsewhere we introduce particle-hole indices \( q_1, q_2 \in \pm \), such that in particular

\[
 b_{\ell,p,z}^+ = b_{\ell,-p,z}^-, \quad b_{\ell,-p,z}^- = b_{\ell,p,z}^+ .
\]  

(55)

Note the minus sign in the momentum in the second relation. This means generally that

\[
 \left( b_{\ell,p,z}^\dagger \right) = b_{\ell,-p,z}^- .
\]  

(56)

To make the coefficients unambiguous, we choose the symmetrized form

\[
 B_{k,p}^{n,\ell,1} = B_{k,-p}^{n,\ell,-1} .
\]  

(57)

Demanding that \( Q_{nk}^+ = (Q_{nk}^-)^\dagger \) implies that

\[
 B_{k,p}^{n,\ell,1 | q_1 q_2} = B_{k,-p}^{n,\ell,-1 | q_2 q_1} .
\]  

(58)

If the phonon mode \( n \) which \( Q_{nk}^q \) is coupled to is \( C_2 \) invariant, then only terms with \( \ell_1 = \ell_2 \) are non-zero. In Sec. [V C T 1], we will introduce a concrete and general model of spin-lattice couplings, and see that within this model, almost all interactions obey this selection rule. In particular, off-diagonal terms with \( \ell_1 \neq \ell_2 \) arise only from the \( A_{b_{\ell,\ell}}^{(l)} \) couplings defined in Eq. (69), which are furthermore smaller in magnitude than other couplings as they are related to magnetic anisotropy.

2. Diagonal scattering rate

Contributions to the first-order longitudinal scattering rate, Eq. (2) or Eq. (24), can be computed exactly using Wick’s theorem. To do so we use the free particle two point function, which in the notation of Eq. (55) is

\[
 \left\langle b_{\ell_1,p_1,z_1}^\dagger (t_1) b_{\ell_2,p_2,z_2} (t_2) \right\rangle = \delta_{\ell_1,\ell_2} \delta_{z_1,z_2} \delta_{q_1,-q_2} \delta_{p_1+p_2,0} \times f_{\ell_2}(\Omega_{\ell_1,p_1}) e^{-i\eta\Omega_{\ell_2,p_2}(t_1-t_2)} .
\]  

(59)

Here \( f_{\ell}(\Omega) = (1+g)/2 + n_{\Omega}(\Omega) \), where \( n_{\Omega}(\Omega) \) is the Bose distribution, and we used the fact that the magnon dispersions are even functions of momentum. One obtains two contributions, \( D_{nk}^{(1)+} = \sum_{s=\pm} D_{nk}^{(1)s} \), where \( D_{nk}^{(1)+} \) corresponds to the emission of two magnons and \( D_{nk}^{(1)-} \) corresponds to the scattering of a magnon from one state to another:

\[
 D_{nk}^{(1)+} = \frac{2\pi}{\hbar^2} \frac{1}{N_{\text{uc}}^2} \sum_{p,\ell,\ell'} \frac{1}{\sinh(\frac{\beta}{2} \hbar \omega_{nk})} \left[ \sinh(\frac{\beta}{2} \hbar \Omega_{\ell,p} + \frac{\beta}{2} \hbar \Omega_{\ell',p} - \frac{\beta}{2} \hbar \Omega_{\ell',p}) \right] \times \delta(\omega_{nk} - \Omega_{\ell,p} + \frac{\beta}{2} \hbar \Omega_{\ell',p} - \frac{\beta}{2} \hbar \Omega_{\ell',p}) \right|_{B_{k,p}^{n,\ell,+}+\ell_-}^2 ,
\]  

(60)

and

\[
 D_{nk}^{(1)-} = \frac{4\pi}{\hbar^2} \frac{1}{N_{\text{uc}}^2} \sum_{p,\ell,\ell'} \frac{1}{\sinh(\frac{\beta}{2} \hbar \omega_{nk})} \left[ \sinh(\frac{\beta}{2} \hbar \Omega_{\ell,p} + \frac{\beta}{2} \hbar \Omega_{\ell',p} - \frac{\beta}{2} \hbar \Omega_{\ell',p}) \right] \times \delta(\omega_{nk} - \Omega_{\ell,p} + \frac{\beta}{2} \hbar \Omega_{\ell',p} - \frac{\beta}{2} \hbar \Omega_{\ell',p}) \right|_{B_{k,p}^{n,\ell,-}+\ell_-}^2 .
\]  

(61)

Note that the prefactor involves just the number of two-dimensional unit cells in a single layer, \( N_{\text{uc}} = N_{\text{uc}}/N_{\text{layers}} \), which results because a single sum over \( z \) gives a factor of the number of layers \( N_{\text{layers}} \), converting the \( N_{\text{uc}} \) to \( N_{\text{uc}}^2 \). One can compare the expressions in Eq. (60) and Eq. (61), and observe a difference of a factor 2 in the prefactor, the sign of the second \( \Omega \) frequency in the delta function, and that of the second to last index in \( B \). The squared modulus \( | \cdots |^2 \) can be traced back to Fermi’s golden rule, and the thermal \( \sinh(\cdots) \) factors, which originate from Bose factors, fall off exponentially at large momenta. Energy conservation imposed by the delta functions strongly constrain these scattering rates. Specifically, if all magnons have the same velocity \( v_m \) and the phonons have an isotropic velocity \( v_{ph} \), then we find that

\[
 \text{supp} \left( D_{nk}^{(1)+} \right) \subseteq \left\{ (k, k_z) | (v_{ph}^2 - v_m^2) |k|^2 + v_{ph}^2 k_z^2 > 4\Delta^2 \right\} \cup \left\{ (k, k_z) | (v_{ph}^2 - v_m^2) |k|^2 + v_{ph}^2 k_z^2 < 0 \right\} ,
\]  

(62)

where \( \Delta = \min(\Delta_0, \Delta_1) \) and \( \text{supp}(D) \) is the support of \( D \). It follows that if \( v_m > v_{ph} \), \( D_{nk}^{(1)+} \) is non-zero in two regions of large \( |k_z| \) bounded by hyperboloid surfaces tangent to the \( \left\{ k_z = \pm \sqrt{v_m^2 - v_{ph}^2} \right\} \), while \( D_{nk}^{(1)-} \) is non-zero in the region outside the said cone, containing large \( |k| \). The two regions are mutually exclusive, i.e. for any given \( k \) at most one of the two rates is non-zero. For \( v_m < v_{ph} \), the constraints are even stronger, and
\( D^{(1)\uparrow}_ {\text{nk}} = 0 \) strictly vanishes, while \( D^{(1)\downarrow}_ {\text{nk}} \) is non-zero within an ellipsoid region containing \( k = 0 \). The first and second scenarios are realized in La\(_2\)CuO\(_4\) [20], and in, e.g., FeCl\(_2\) [27], respectively.

\[
\begin{align*}
\mathbb{M}^{q,q'}_{nk,n'k'} & = \frac{64\pi^2}{\hbar^2} \frac{1}{N_{\text{uc}}} \sum_{p} \sum_{\{\xi,\eta\}} \delta\left(\Sigma^{q,q'}_{nk,n'k'} + q_{1}\Omega_{\xi,1,p} + q_{2}\Omega_{\xi,2,p+q+k+q'k'}\right) \delta\left(\Delta^{q,q'}_{nk,n'k'} + 2q_{3}\Omega_{\xi,3,p+q+k+q'k'} - q_{1}\Omega_{\xi,1,p} + q_{2}\Omega_{\xi,2,p+q+k+q'k'}\right) \\
& \times q_{4} \left(2n_{B}(\Omega_{\xi,3,p+q+k'k'}) + 1\right) \left(2n_{B}(\Omega_{\xi,1,p}) + 1\right) \left(2n_{B}(\Omega_{\xi,2,p+q+k+q'k'}) + 1\right) \text{Im}\left\{ \mathbb{E}^{\ell_{1}\ell_{4}\ell_{3}\ell_{2} q_{2} q_{4} q_{1} q_{q'q'}}_{k,p+q+k+q'k'} \right\},
\end{align*}
\]

where we defined \( \Sigma^{q,q'}_{nk,n'k'} = q_{\omega_{nk}} + q'_{\omega_{n'k'}} \), \( \Delta^{q,q'}_{nk,n'k'} = q_{\omega_{nk}} - q'_{\omega_{n'k'}} \). Note that while we described and will use below a continuum formulation of the spin wave theory in Sec. V A, the result in Eq. (63) is actually valid at the lattice level, i.e., when the full periodic band structure of the magnons is included, as it relies only upon the canonical commutation relations of the magnon operators, and their dispersions and couplings are taken completely arbitrary at this stage. Therefore this formula could be applied directly in many other circumstances.

We may understand the terms in Eq. (63) as follows: the second energy conservation delta function comes from Fermi’s golden rule; the first delta function, and the denominator in the third line, come from \( \sum_{\xi,\eta} \mathbb{E}^{\ell_{1}\ell_{4}\ell_{3}\ell_{2} q_{2} q_{4} q_{1} q_{q'q'}}_{k,p+q+k+q'k'} \); while the Bose factors appear when evaluating the thermal averages of magnon population numbers, and their product falls off exponentially at large momenta. \( \mathbb{M}^{q,q'}_{nk,n'k'} \) may display divergences when the denominator vanishes. One can explicitly check that the detailed balance relation, Eq. (27), holds, using the properties of the \( \mathbb{B} \) coefficients, as well as \( \mathbb{M}^{q,q'}_{nk,n'k'} = \mathbb{M}^{q',q}_{n'k',nk} \).

### C. Phenomenological coupling Hamiltonian

We now propose a symmetry-based phonon-magnon coupling Hamiltonian, Eq. (65), for the low-temperature ordered phase of a Néel antiferromagnet on lattice made of layers of square lattices, and, as above, we consider the layers to be magnetically decoupled. Moreover, for concreteness, we take the classical ground state to be Néel antiferromagnetic along the \( \hat{u}_z \) axis, so that all the point-group symmetries of the crystal are preserved by the magnetic structure, up to a translation of half a magnetic unit cell. [28]

#### 1. Interaction Hamiltonian density

We consider the most general coupling between \( (1) \) the strain tensor, \( \mathcal{E}^{\alpha\beta} = \frac{1}{2}(\partial^\nu u^\nu + \partial^\nu u^\nu) \), where \( u^\nu \) is the lattice displacement field, and \( (2) \) spin bilinears in terms of the \( m, n \) fields, allowed by the symmetries of our tetragonal crystal in its paramagnetic phase, which has the largest symmetry group provided by the crystal structure (generated by mirror symmetries \( \hat{S}_{x}, \hat{S}_{y}, \hat{S}_{z} \), fourfold rotational symmetry \( C_{4y}^\ast \), translation and time-reversal). Since we treat the magnetism as two dimensional, the coupling Hamiltonian is a sum over layers and an integral over two dimensional space,

\[
H'_{\text{tetra}}(r) = \sum_{z} d^2 x \mathcal{H}'_{\text{tetra}}(r). \tag{64}
\]

We use \( r = (x, z) \) to denote the three-dimensional coordinate. The corresponding local hamiltonian density reads, with all fields expressed in real space:

\[
\mathcal{H}'_{\text{tetra}}(r) = \sum_{\alpha,\beta} \mathcal{E}_{\alpha} \left( \lambda^{(n)\alpha\beta}_{\alpha} n_{\alpha} n_{\beta} + \frac{1}{n_{0}} \mathcal{L}_{\alpha\beta} \right)_{x,z}, \tag{65}
\]

where \( n_{0} = \mu_{0}/a^2 \) is the ordered moment density. Here each \( \lambda^{(n)}_{\alpha\beta} \) tensor, which we define to be symmetric in both \( ab \) and \( \alpha\beta \) variables, has seven independent coefficients,
which we call
\[
\begin{align*}
\Lambda_1 & = \Lambda^{(\xi,xx)} = \Lambda^{(\xi,yy)}, \\
\Lambda_2 & = \Lambda^{(\xi,xx)} = \Lambda^{(\xi,yy)}, \\
\Lambda_3 & = \Lambda^{(\xi,xx)} = \Lambda^{(\xi,yy)}, \\
\Lambda_4 & = \Lambda^{(\xi,zz)} = \Lambda^{(\xi,zz)}, \\
\Lambda_5 & = \Lambda^{(\xi,zz)}, \\
\Lambda_6 & = \Lambda^{(\xi,xx)} = \Lambda^{(\xi,yy)} = \Lambda^{(\xi,yy)} = \Lambda^{(\xi,yy)}, \\
\Lambda_7 & = \Lambda^{(\xi,zz)} = \Lambda^{(\xi,zz)} = \Lambda^{(\xi,zz)} = \Lambda^{(\xi,zz)}, \\
& = \Lambda^{(\xi,zz)} = \Lambda^{(\xi,zz)} = \Lambda^{(\xi,zz)} = \Lambda^{(\xi,zz)},
\end{align*}
\]
and all other \(\Lambda_{ab}^{(\xi,\alpha\beta)}\) are zero. In Appendix F1, we provide a microscopic derivation of these \(\Lambda_{ab}^{(\xi,\alpha\beta)}\) coefficients in terms of \(\eta^2 \partial^2_{\eta^2} J^{ab}\), i.e. in terms of the spatial derivatives of the magnetic exchange \(J^{ab}\). Within this microscopic approach \(\Lambda_{ab}^{(\xi)}\) are related to the spatial derivatives of symmetric off-diagonal exchange \(J^{xy}, J^{yx}, J^{yz}\), while \(\Lambda_{ab}^{(\xi,xx)} = \Lambda_{ab}^{(\xi,yy)}\) and \(\Lambda_{ab}^{(\xi,xx)} = \Lambda_{ab}^{(\xi,yy)}\) are associated with the spatial derivatives of XXZ exchange anisotropy \(J^{xx,yy} = J^{yy}\).

Finally, note that in Eq. \((66)\) bilinears of the \(n_a m_b\) kind, arising from e.g. alternating DM interactions \(J^{ij}\) i.e. such that \(J^{rr,xx} = -J^{rr,yy}\) with \(a = x, y\), could also contribute to the thermal Hall conductivity \(\mathcal{H}_{\text{Hall}}\), but are not allowed in the single-site (paramagnetic) Bravais lattice we consider here.

2. Expansion

We now carry out an expansion of the \(m, n\) fields in two steps. First we expand around the zero-field, zero-net-magnetization Néel-ordered configuration \((n_{\text{ord}} = 1, m_{\text{ord}} = 0)\), assuming deviations are small and satisfy Eq. \((46)\). One thereby expresses \(n_x\) and \(m_x\) in terms of the free fields \(n_{y/z}, n_{y/z}\) as, in real space:

\[
\begin{align*}
n_x &= 1 - \frac{1}{2} \sum_{b=y,z} \left( n_b^2 + \frac{1}{n_0^2} m_b^2 \right), \\
m_x &= - \sum_{b=y,z} m_b n_b,
\end{align*}
\]

which are correct to second order in the free fields (this constitutes a non-linear correction to Eq. \((46)\)). In a second step, we include a net magnetization and expand \(\mathbf{m}\) around it, i.e. write \(m^\alpha = m_0^\alpha + m^\alpha\) where \(m_0^\alpha\) is the sum of both a possible spontaneous magnetization and response to the external magnetic field. This two-step expansion physically assumes \(m \ll m_0 \ll n_0\). Using these forms in Eq. \((66)\), we obtain the spin-lattice coupling to second order in the free field fluctuations:

\[
H'_{\text{teta}}(r) \approx \sum_{\alpha\beta} c_{\alpha\beta}^{(r)} \sum_{a,b=y,z} \lambda_{ab}^{(\xi,\alpha\beta)} n_0^{-\xi} \xi_{a\xi} \xi_{b\xi}.
\]

3. In terms of the eigenbosons, \(b, b^\dagger\)

We now seek to identify the \(\mathcal{B}\) coefficients as defined in Eq. \((64)\) (with the convention Eq. \((65)\)). To do so, we use the Eq. \((48)\) representation of the \(n_a, n_b\) fields in terms of the \(b\) bosons, which diagonalize the pure magnetic Hamiltonian, and plug in their expressions into Eq. \((68)\). For computational convenience, we carry this out using an explicit formula, which associates \(\xi = n \leftrightarrow \xi = 0\) and \(\xi = m \leftrightarrow \xi = 1\), and \(a = y \leftrightarrow a = 0\) and \(a = z \leftrightarrow a = 1\). Then Eq. \((59)\) can be written in the following compact form

\[
\xi_{nk} = \frac{1}{\sqrt{2}} (-1)^{d_{\alpha\xi}} \xi_{\ell} \xi^{\ell}/2 \Omega_{\alpha\xi,k}^{\ell/2} \sum_{q=\pm 1} (-1)^{(1+q)/2} \delta_{k,\alpha\xi}^{\ell} \xi_{\ell},
\]

where \(\xi = 1 - \xi\), i.e. \(\xi = 1\), \(\bar{\xi} = 0\), and \(\bar{\xi} = 2 - 1 = 1\). When writing Eq. \((70)\), we used relation for the valley \(\ell = \delta_{\alpha\xi}\), and conversely \(a = \xi \ell + \bar{\xi}\).

We similarly express the local strain in terms of its constituent Fourier modes, which are proportional to the phonon creation/annihilation operators, as discussed in detail in Appendix A. Putting in these two ingredients, some algebra (shown also in Appendix A) finally yields
where $\xi,\xi' \in (0,1)$ and $\eta,\eta' \in (-1,1)$.

\[
B_{\xi,\xi'}^{\eta,\eta'} = \frac{i}{4} \sqrt{\frac{h}{2M_{nc}}} \sum_{\xi',\xi''=m,n} \chi(\xi+\xi'/2) \chi(\xi-\xi'/2) \frac{\chi'(\xi'+\xi')/2}{n_0} \frac{\chi'(\xi'-\xi')/2}{n_0} \frac{\chi'(\xi''+\xi'')/2}{n_0} \frac{\chi'(\xi''-\xi'')/2}{n_0} \Gamma_{\xi',\xi''}^{\xi,\xi'} \Gamma_{\xi',\xi''}^{\xi,\xi'} \Omega_{\xi',\xi''}^{\eta,\eta'} \Omega_{\xi',\xi''}^{\eta,\eta'}.
\] (71)

Which terms in Eq. (68) are compatible with an effective time-reversal symmetry breaking? By direct inspection of Eq. (72), one finds that $i\mathcal{L}_{n,p}^{\xi,\xi'} = (i\mathcal{L}_{n,p}^{\xi,\xi'})^*$. Thus, only those terms in Eq. (71) with $i\mathcal{L}_{n,p}^{\xi,\xi'} = i$ may satisfy $B_{\xi,\xi'}^{\eta,\eta'} = (B_{\xi,\xi'}^{\eta,\eta'})^*$. All others are such that $B_{\xi,\xi'}^{\eta,\eta'} = (B_{\xi,\xi'}^{\eta,\eta'})^*$.

The breaking of effective time-reversal in the phonon system thus relies upon the presence of spin-phonon couplings where $\xi' = \bar{\xi}$, i.e. $\lambda_{n,m}$ coefficients; this is consistent with the argument in Sec. IV C.2 based on macroscopic time-reversal $T$, Eq. (48). Moreover, going back to Sec. IV B 2 we see that if $m_{\bar{k}} \neq 0$ but $\lambda_{\bar{k}} = 0$, then the kernel $K_{n,k}^{\eta,\eta'}$ is invariant under momentum reversal; and so $\kappa_H = 0$, even though the system breaks $T$.

(ii). $\sigma_d$ operation. Here we briefly study the $\sigma_d$ operation, i.e. a mirror transformation through the plane containing the $\hat{z}$ and $\hat{\xi}$ axes. The system, having antiferromagnetic ordering along the $x$ axis as well as possibly $m_{\bar{k}} 
eq 0$, explicitly breaks this symmetry. However, if $\lambda_{\bar{k}} = 0$ and $\lambda_{\bar{k}} = 0$, then $\sigma_d$ is preserved at the level of the kernel $K_{n,k}^{\eta,\eta'}$, whence $\kappa_H = 0$. This illustrates the importance of knowing the action of $D_{\bar{k}}$ operations upon the kernels $K_{n,k}^{\eta,\eta'}$, because some symmetries which are explicitly broken globally might fail to be effectively broken in phonon scattering.

4. Effective breaking of symmetries

(i). Time reversal. We now briefly comment on the relation between the “effective” time-reversal of the spin system $T$ and the transport properties of the phonon system. Indeed, it is obvious from Eqs. (60) and Eq. (63) that if all the $B$ coefficients satisfy

\[
B_{\xi,\xi'}^{\eta,\eta'} = (B_{\xi,\xi'}^{\eta,\eta'})^*,
\] (74)

then $D_{n-k}^{(1)} = D_{n-k}^{(1)}$ and $2\mathcal{M}_{n,k-n,k'}^{\xi,\xi'} = -2\mathcal{M}_{n,k-n,k'}^{\xi,\xi'}$, i.e. the phonon collision integral is effectively time-reversal symmetry preserving, as discussed in Sec. IV A. Therefore, no phonon Hall effect follows if the spin-phonon coupling satisfies Eq. (74).

D. Solutions of the delta functions

Each contribution to the scattering rate involves a momentum integral over an integrand which contains either a single delta function or a product of two delta functions. These express energy conservation constraints, which must be solved to carry out the integration. The argument of each delta function, which must be set to zero, is of the form

\[
\varpi - \Omega_{\ell,p} - s\Omega_{\ell,p-k} = 0,
\] (75)

where $s = \pm 1$. Using the continuum form of the magnon dispersion, $\Omega_{\ell,p} = \sqrt{\nu_m |p|^2 + \Delta_\ell^2} = \nu_m \sqrt{|p|^2 + \delta_\ell^2}$, $\nu_m \Omega_{\ell,p}$, with $\Delta_\ell = \Delta_\ell/v_m$, we can rewrite this as

\[
\Delta_\ell^2 + \delta_\ell^2 + s\sqrt{|p|^2 - k^2 + \delta_\ell^2} = a,
\] (76)

where $a = \varpi/v_m$ and $s = \pm 1$. The existence and type of solutions depend on the value of $a^2 - k^2$, where $k^2 = k_x^2 + k_y^2$. When they exist, the solutions are conics, as is summarized in Table III.
\begin{tabular}{|c|c|c|c|}
\hline
$s = +$ & $a^2 - k^2 < 0$ & $0 < a^2 - k^2 < 4\delta_2^2$ & $4\delta_2^2 < a^2 - k^2$ \\
\hline
$s = -$ & no solutions & no solutions & ellipse \\
\hline
\end{tabular}

TABLE III: Solutions to a single delta function of the form \( \delta(\omega - \Omega_{\ell,p} - s\Omega_{\ell,p - k}) \), with \( s = \pm,1 \), as a function of the value of \( a^2 - k^2 \), where \( a = \omega/v_m \), \( k^2 = k_x^2 + k_y^2 \), and \( \Omega_{\ell,p} = v_m \sqrt{k_x^2 + \delta_2^2} \). The necessary existence conditions described in this Table are captured by the equation \( s(a^2 - k^2) > 4\delta_2(s + 1)/2 \).

It is then best to introduce coordinates \( p_{\|}, p_{\perp} \) which are along the principal axes of the hyperbola/ellipse:
\[
p = p_{\|} \hat{k} + p_{\perp} \hat{z} \times \hat{k},
\]
and we can define the major \( \bar{a} \) and minor \( \bar{b} \) semi-axes, or conversely, of the conics:
\[
\bar{a} = \frac{|a|}{2} \sqrt{1 - \frac{4\delta_2^2}{a^2 - k^2}}, \quad \bar{b} = \frac{1}{2} \sqrt{|a^2 - k^2 - \delta_2^2|}. \quad (78)
\]

An immediate consequence is that, in the case of the ellipse \(-\bar{a} \leq p_{\|} - k / \bar{b} \leq \bar{a} \) and \(-\bar{b} \leq p_{\perp} \leq \bar{b} \), while in the case of the half-hyperbola: \( p_{\|} \geq \bar{k} + \frac{\bar{b}}{2} \).

Both Eq. (79) and a pair of such equations may be solved analytically, but the solutions are analytically complicated. We provide their details in Appendix E1 and give here only the final results.

### 1. Diagonal scattering rate

We have, in particular, using the following compact form for \( D_{nk}^{(1)s} \), with \( s = \pm, \)
\[
D_{nk}^{(1)s} = \frac{(3 - s)a^2 \sinh(\frac{\beta}{2} h\omega_{nk})}{4\pi v_m h^2} \int_{-\infty}^{+\infty} dy \sum_{\eta} f_{\eta}^s(y) J_\eta^s(y) \sum_{\ell} \frac{\sinh(\frac{\beta}{2} h\Omega_{\ell,p_{\|}})}{\sinh(\frac{\beta}{2} h\Omega_{\ell,p_{\perp}}) \sinh(\frac{\beta}{2} h\Omega_{\ell,p_{\perp} - k})}, \quad (80)
\]
where \( \Omega = \Omega/v_m \), and
\[
\begin{cases}
  f_{\eta=1}^s(y) = \Theta(\bar{b} - |y|) \Theta(a^2 - k^2 - 4\delta_2^2), \\
  f_{\eta=-1}^s(y) = \delta_{\eta,1} \Theta(\bar{a}^2 - a^2)
\end{cases}, \quad (81)
\]
where \( a = \omega_{nk}/v_m, \eta = \pm,1 \) and
\[
J_\eta^s(y) = \left[ \frac{2^{1-s}/c_\eta(y)}{\sqrt{2^{1-s}c_\eta(y)^2 + y^2 + \delta_2^2}} - \frac{s c_{-\eta}(y)}{\sqrt{-c_{-\eta}(y)^2 + y^2 + \delta_2^2}} \right]^{-1} \quad (82)
\]
\[
c_\eta(y) = \frac{1}{2} \left( k + \eta a \sqrt{1 - \frac{4\delta_2^2 + y^2}{a^2 - k^2}} \right) \quad (83)
\]
and
\[
P_{\ell,p_{\|}}^{(s)}(y) = c_\eta(y) \hat{k} + y \hat{z} \times \hat{k}. \quad (84)
\]
At this point it may be comforting to check dimensions. Noting that \( y \) has dimensions of momentum, i.e. inverse length, and \( B \) has dimensions of energy, i.e. inverse time, one can indeed see that \( D \) in Eq. (80) has proper dimensions of a rate.

### 2. Off-diagonal scattering rate

In this case, we must solve a pair of conic equations simultaneously, which take the form:
\[
\begin{cases}
  \omega_1 - \Omega_{\ell,p} - s_1 \Omega_{\ell,p - k_1} = 0 \\
  \omega_2 - \Omega_{\ell,p} - s_2 \Omega_{\ell,p - k_2} = 0
\end{cases}, \quad (85)
\]
i.e.
\[
\begin{cases}
  \sqrt{p_{\|}^2 + \delta_2^2 + s_1 \sqrt{p_{\|}^2 + k_1^2 + \delta_2^2}} = a_1 \\
  \sqrt{p_{\|}^2 + \delta_2^2 + s_2 \sqrt{p_{\|}^2 + k_2^2 + \delta_2^2}} = a_2
\end{cases}, \quad (86)
\]
where \( a_i = \omega_i/v_m \). Indeed, the integrals which occur in the second order scattering rates involve pairs of delta functions, whose arguments are of the form considered above, with in Eq. (85), \( \omega_1 = -q_1 \Sigma_{nk/p_{\|}k_1}; \omega_2 = -q_1 q' \Sigma_{nk/p_{\|}k';} \), \( s_1 = q_1 q_2, \ s_2 = -q_1 q_3, k_1 = -q_k - q'k, \ k_2 = -q'k', \delta_1 = \Delta/v_m \). In this case, each of the two delta function constraints defines a half-hyperbola or an ellipse in the \( p \) plane, and the integrand is confined to the intersections of these two curves. Consequently, the integral will be collapsed to a discrete set of points. It is
straightforward to see geometrically that the intersection of two curves of these types is, except for the degenerate cases in which the two curves are identical, a set of at most four points. The two simultaneous equations can be solved analytically, but the solutions are algebraically complicated and we give here only the results and leave details to the Appendices.

Collapsing the delta functions as explained in Appendix [E1], we can write:

$$\mathcal{W}^{\omega,q,q'}_{n,k,n',k'} = \frac{4\eta^2}{m^3 h^4} \sum_j \sum_{\ell, (q_s)} \tilde{j}(p_j) \mathcal{J}^{\ell,\ell,\ell}_{p_j, q, q'}$$

where

$$\tilde{j}(p_j) = \frac{\mathcal{J}^{\ell,\ell,\ell}_{k,p_j, \frac{1}{2} q_k + q' k'}}{\mathcal{J}^{\ell,\ell,\ell}_{k,p_j, \frac{1}{2} q_k + q' k'}} PP \left[ \begin{array}{c}
\mathcal{J}^{\ell,\ell,\ell}_{k,p_j, \frac{1}{2} q_k} & -q_4 \mathcal{J}^{\ell,\ell,\ell}_{k,p_j, \frac{1}{2} q_k + q' k'} & \mathcal{J}^{\ell,\ell,\ell}_{k,p_j, \frac{1}{2} q_k - q_4 q' k'} \\
\mathcal{J}^{\ell,\ell,\ell}_{k,p_j, \frac{1}{2} q_k + q' k'} & \mathcal{J}^{\ell,\ell,\ell}_{k,p_j, \frac{1}{2} q_k} & -q_4 \mathcal{J}^{\ell,\ell,\ell}_{k,p_j, \frac{1}{2} q_k + q' k'} \\
\mathcal{J}^{\ell,\ell,\ell}_{k,p_j, \frac{1}{2} q_k - q_4 q' k'} & \mathcal{J}^{\ell,\ell,\ell}_{k,p_j, \frac{1}{2} q_k + q' k'} & \mathcal{J}^{\ell,\ell,\ell}_{k,p_j, \frac{1}{2} q_k}
\end{array} \right]$$

is a product of thermal factors and where, when they exist, the solutions, $j = 0, \ldots, 3$ take the form

$$p_j = t_{[j/2]} V_{[j/2]} + u_{[j/2]} W_{[j/2]},$$

where, for $i = 0, 1$ $v_i = a_2 k_1 + (-1)^i a_1 k_2$, $w_i = \hat{z} \times v_i$, $t_i$ and $u_i^{(\pm)}$ are given in Appendix [E1] (Also recall we defined $0 = -1$, $1 = 1$, $x [2] = x \mod 2$, and $[x]$ denotes the floor of $x$), and

$$\tilde{j}(p_j) = \left[ \begin{array}{c}
v_1 & \frac{k_1 \wedge p_j}{\Omega_{\ell,p_j} \Omega_{\ell,p_j} - k_1} + v_2 \frac{p_j \wedge k_2}{\Omega_{\ell,p_j} \Omega_{\ell,p_j} - k_2} + v_3 \frac{-k_1 \wedge k_2 + p_j \wedge k_2 - p_j \wedge k_1}{\Omega_{\ell,p_j} \Omega_{\ell,p_j} - k_2} \left. \right|^{-1},
\end{array} \right.$$}

where $V_1 \wedge V_2 = V_x V_y - V_y V_x$ for any in-plane vectors $V_{1,2}$. Coefficients $t_{0,1}$ are always well defined, but for each $i$, $u_i^{(\pm)}$ are the solutions to a quadratic equation which has zero, one or two solutions, whether the discriminant $d_{a,i}$ thereof is negative, zero, or positive.

Necessary (but not sufficient) conditions of existence of solutions are: (i) the existence of both conics, cf. Table [III] (ii) $d_{a,0} \geq 0$ and/or $d_{a,1} \geq 0$, (iii) when $s_1$ and/or $s_2$ is negative, the $p_j$ must lie on the $\eta_{1,2} = 1$ branch of the 1 and/or 2 hyperbola. Even with these constraints, spurious solutions exist, so that one must check that the solutions Eq. ([88] also satisfy the equations for the given values of $a_1, a_2, k_1, k_2, q, q', q_t$.

E. Scaling and orders of magnitude

In this subsection, we discuss the temperature dependence and magnitude of the magnonic contributions to the different phonon scattering rates, which determine the phonon thermal conductivity and thermal diffusivity tensors. Since we consider a low-energy continuum theory (without a momentum cutoff) in which the dispersion of the phonons is linear, these hold only in the low-temperature limit. In Table [IV] we summarize some of the relations derived in this section.

First we consider the leading magnonic contributions to the longitudinal scattering rate, $D^{(1)}_{skw}$. The typical magnitude of this quantity for $|k| \sim k_B T/v_{ph}$ sets the basic rate $1/\tau$. This rate has been studied previously in classic work on the phonon-magnon coupling in antiferromagnets. Reference [31] finds that $1/\tau \sim T^5$ (for the moment we give only the $T$ dependence under the above condition, and do not give the prefactor), for a model of exchange-striction in a Heisenberg antiferromagnet in three dimensions. This should be recovered from our formalism.

1. Longitudinal scattering rate: Role of anisotropies and scaling exponent

In this subsection, we discuss the temperature dependence and magnitude of the magnonic contributions to the different phonon scattering rates, which determine the phonon thermal conductivity and thermal diffusivity tensors. Since we consider a low-energy continuum theory (without a momentum cutoff) in which the dispersion of the phonons is linear, these hold only in the low-temperature limit. In Table [IV] we summarize some of the relations derived in this section.
A general estimate can be obtained from Eqs. (60, 61). To evaluate it requires, in addition to the dispersion relations, the phonon-magnon couplings, which are given in Eq. (71). At the level of temperature scaling for typical thermal momenta, for momenta well above the magnon gap, $v_{ph} k \gg \Delta$, we may replace $k \sim k_B T / v_{ph}$, $\omega \sim v_{ph} k \sim k_B T$ and $\Omega \sim k_B T$ (the latter is true if the ratio between $v_m$ and $v_{ph}$ is order one). Noting that $\xi$ and $\xi'$ in Eq. (71) equal $\pm 1$, we see that a general phonon-magnon coupling is a sum of three contributions,

$$B \sim \left( \frac{k_B T}{M_{ph}^2} \right)^{\frac{1}{2}} n_0^{-1} \left( \lambda_{mn} \frac{\lambda k_B T}{n_0} + \lambda_{mm} + \lambda_{nm} \frac{n_0}{\chi k_B T} \right).$$  

(90)

Depending upon which of these terms is dominant, the temperature dependence of $B \sim T^{1/2+\varepsilon}$, with $\varepsilon = -1, 0, 1$ corresponding to the $\lambda_{mm}$, $\lambda_{mn}$ and $\lambda_{nm}$ terms, respectively. We can then estimate the scattering rate by converting the momentum sum over $p$ to a $d$-dimensional integral ($d$ is the spin-exchange dimensionality) and recalling $|p| \sim T$. We see therefore that

$$\frac{1}{\tau} \sim T^{d-1} |B|^2 \sim T^{d+2\varepsilon}.$$  

(91)

A priori, the dominant contributions would arise from terms with $x = -1$, which have the smallest power of temperature, which would give $1/\tau \sim T^{d-2} \sim T$ in $d = 3$ dimensions. This does not agree with Ref. [31]. Instead, one notices that what one might expect to be the subdominant contribution from $x = +1$, which gives $1/\tau \sim T^{d+2}$ in general dimensions, does agree with the classic theory for $d = 3$.

Why is this the case? The resolution lies in the fact that Ref. [31] assumes isotropic Heisenberg interactions, and is carried out in zero magnetic field. As a consequence, the Hamiltonian has SU(2) symmetry, and Goldstone’s theorem protects the gaplessness of the magnon modes even in the presence of strain. In particular, because even an arbitrarily strained lattice must preserve the gapless magnons in this case, the spin-lattice coupling, Eq. (65), must be spin-rotationally invariant, and moreover its quadratic expansion, Eq. (68), must vanish for a magnon configuration which is a small rotation of the Néel order, which corresponds to either $n_y$ or $n_z$ non-zero and spatially constant. This means that the non-zero terms in Eq. (68) involve only $\xi, \xi' = m$ and not $n$ (in a treatment including higher order terms, spatial gradients $\nabla n$ would appear, but these scale in the same manner as $m$). One can indeed check in Eq. (69) that when the interactions $\Lambda^{(m/n),\alpha\beta}$ are isotropic ($\propto \delta_{ab}$), $\lambda_{nn}$ vanishes, and $\lambda_{mn}$ vanishes at zero field if when the uniform magnetization $m_0^a = 0$. Taking the $\lambda_{mm}$ contribution in Eq. (90) gives $x = +1$ in Eq. (91) as needed for agreement with earlier work.

What is the physics of the different values of $x$? We see that stronger effects (smaller powers of temperature) arise from coupling to $n$ than to $m$. This is a fundamental property of antiferromagnets: fluctuations of the order parameter $n$ are stronger and more long-ranged than those of the uniform magnetization $m$, which is naturally suppressed when antiferromagnetic interactions dominate. Thus larger effects would be expected from coupling of strain to the staggered magnetization than to the uniform one, as the formula indeed shows.

How is this reflected in $\kappa_L(T)$? The last step from the scattering time $\tau$ to the longitudinal conductivity $\kappa_L$ is a standard one [20, 32]. The sum over phonon momentum $k$ in Eq. (9) is converted to a three-dimensional integral (the magnon momentum integral was $d$-dimensional, with $d = 2$ in the case of a layered antiferromagnet).

For temperatures $k_B T \gg \Delta$, the scaling for the temperature dependence of the longitudinal conductivity is

$$\kappa_L \sim T^{3-d-2\varepsilon}.$$  

(92)

As can be seen from Eq. (90), a crossover between the low-temperature $x = -1$ and the high-temperature $x = +1$ behaviors occurs at $T^*_L$,

$$k_B T_L^* \sim \frac{n_0}{\chi} \sqrt{\frac{\lambda_{nn}}{\lambda_{mm}}}.$$  

(93)

Eq. (93) assumes that the intermediate behavior $x = 0$, due to the $\lambda_{mn}$ coupling which is proportional to both anisotropic exchanges and the net magnetization, is negligible; this is consistent with our numerical results shown in Sec. [VF5]. The above results, Eqs. (92, 93), also assume that $D^{(1)}_n$ is the dominant scattering rate contributing to the longitudinal inverse scattering time $D^{(1)}_{nk}$. The role of $D^{(1)}_n$ is considered in more detail in Sec. [VF5]. However, many more scattering processes, such as boundary or impurity scattering, which in Eq. (2) are encompassed as $D^{(1)}_{nk}$, must contribute (through Matthiessen’s rule) to the phonon relaxation. Thus, $\kappa_L$ should be considered a probe of the full $D^{(1)}_{nk}$.

2. Longitudinal scattering rate: Role of the gap and magnetic field dependence

Since we have seen that the assumption of isotropic interactions supresses the coupling to the staggered magnetization, this discussion suggests that breaking of spin-rotation symmetry should greatly enhance phonon scattering. While this may indeed be the case, we should note a subtlety: although spin anisotropy indeed allows such coupling, it also allows the formation of a magnon gap —enlarged by the presence of an external magnetic field, $\Delta_\ell = \sqrt{\Gamma_\xi / \chi + h^2}$, which behavior should be expected from the combination of these two effects?

Regardless of the form of coupling (scaling exponent $x$), if $k_B T \ll \Delta$, magnon-phonon scattering will become energetically unavailable. More precisely, $D^{(1)+}$, corresponding to the process whereby a phonon excites two magnons, is exponentially suppressed due to the required rest energy $2\Delta$, while $D^{(1)-}$, corresponding to
the process whereby a phonon scatters a magnon, is exponentially suppressed due to the exponential decrease of all magnon populations at temperatures below the gap. Therefore \( D^{(1)} \) as a whole is exponentially suppressed if \( k_B T \ll \Delta \); we check this behavior numerically in Sec. [VF.4]

Thus, a crossover in the behavior of \( \kappa_L (T) \) occurs at temperature \( T_{\Delta} \sim \Delta/k_B \). Below \( T_{\Delta} \), the phonon thermal conductivity is mostly due to other scattering effects, which are captured by \( D_{nk} \) in this work. For constant \( D_{nk} \), this yields \( \kappa_L \sim T^d \). Above \( T_{\Delta} \), phonon-magnon scattering becomes available, and is enhanced by anisotropic coupling; provided this is the dominant effect, the resulting thermal conductivity behavior is \( \kappa_L \sim T^{d-2x} \) with \( x = -1 \) which, for \( d = 2 \) (two-dimensional magnons), is the same power of temperature as that obtained with only constant \( D_{nk} \). However, the proportionality constant is larger with phonon-magnon scattering than without, which, for sufficiently strong anisotropic couplings (i.e. sufficiently large \( \lambda_{nm} \)), may lead to a "bump" in \( \kappa_L (T) \), as we indeed numerically see in Sec. [VF.5]

Remarkably, this effect depends on the external magnetic field through the width of the magnon gap (recall the latter is field dependent), and may be an important feature of \( \kappa_L (h, T) - \kappa_L (0, T) \). For the sake of completeness, we note that types of dependences on the magnetic field may arise at temperatures where the scaling exponent \( x = 0 \) plays a role, because the \( \lambda_{nm} \) coupling depends explicitly on the net magnetization \( m_B \) in (see Eq. (69)). It is however not clear how this contribution could become non-negligible in any range of temperatures, and the gap dependence \( \Delta (h) \) is the main culprit as regards the dependence on \( h \) of the longitudinal conductivity.

3. Transverse scattering: scaling exponent

We can now apply similar reasoning to the transverse/Hall scattering rate \( \mathcal{W}^{\ominus} \) from Eq. (63). Obviously if temperature is sufficiently low, i.e. below magnon gaps, the result will be exponentially suppressed. Of greater interest is the energy regime above the magnon gaps, in which we may assume acoustic linearly dispersing magnons (and phonons). We proceed by counting the obvious factors of momentum and energy, and by assuming the relevant momentum scales are set by dimensional analysis, i.e. \( k, k' \sim k_B T/v_m \) etc. Inspection of Eq. (63) shows one sum over magnon momentum \( p \), which converts to an integration in the thermodynamic limit, two energy delta functions, and one energy denominator, which, using the aforementioned momentum scaling implies that

\[
\mathcal{W}^{\ominus} \sim T^{d-3} B^4. 
\]  

Here we considered the magnon momentum integration as \( d \)-dimensional, as in the previous discussion of longitudinal scattering rates.

Now to proceed we must estimate the contribution of the four \( B \) factors. To do so, we need to consider the effective time-reversal symmetry \( \mathcal{T} \). This symmetry must be broken to obtain a non-zero effective skew-cattering rate, \( \mathcal{W}_{nk,v'k'}^{\ominus,\text{eff},qf'} \), which in particular is odd under \( \mathcal{T} \). As discussed in Secs. [VC.2] and [VC.3] under \( \mathcal{T} \) the \( \lambda_{nm} \) and \( \lambda_{nn} \) couplings are even while only the \( \lambda_{mm} \) couplings are odd; therefore \( \mathcal{W}^{\ominus,\text{eff}} \) must contain an odd number of factors of \( \lambda_{mm} \). Furthermore, in the low field regime we consider here, \( \mathcal{T} \) symmetry breaking happens through the development of a small uniform magnon density, hence \( \lambda_{mn} \propto m_0 \), which in turn is linearly proportional to the applied field (see Eq. (69)). Consequently, to obtain the linear-in-field Hall scattering rate, we should keep just one (and not three, the other available odd number) factors of \( \lambda_{mm} \). Therefore, we may use Eq. (90) to estimate

\[
\mathcal{W}^{\ominus,\text{eff}} \sim T^{d-1} \lambda_{mm} (\lambda_{mn} T + \lambda_{nn} T^{-1})^3 \sim T^{d-1+3x}. 
\]  

Here, as in Sec. [VF.3] \( x = +1 \) obtains in a large parameter region where \( \lambda_{mm} \ll \lambda_{nn}(\sim m_0/k_B T)^2 \), while \( x = -1 \) results if \( \lambda_{mm} \) is non-zero and dominant in a low-temperature regime where the magnon gap remains negligible.

It is by no means clear how the latter regime would be achieved, and if we assume that the \( x = +1 \) case dominates, then it is interesting to see that \( \mathcal{W}^{\ominus,\text{eff}} \) in Eq. (95) scales like \( T^{d+2} \) which is the same power of temperature as the magnon contribution to the longitudinal scattering rate in Eq. (91).

This scaling is a bit surprising, as we should expect that the transverse is smaller than the longitudinal scattering, since it comes from a higher order term. To resolve this, we should consider more carefully the relationship of \( \mathcal{W}^{\ominus,\text{eff}} \) to a "skew scattering rate". In particular, one should note that \( \mathcal{W}_{nk,v'k'}^{\ominus,\text{skew}} \) enters the collision term via a \( \delta \) function over the \( k' \) in the thermodynamic limit. Therefore the measure of this integral, which is expected to be dominated by \( |k'| \sim k_B T/v_m, k_B T/v_{ph} \), contributes an additional factor of \( T^3 \) (since phonons are always three-dimensional). Thus it would be more correct to estimate the skew scattering rate as

\[
\frac{1}{\tau_{\text{skew}}} \sim T^3 \mathcal{W}^{\ominus,\text{eff}} \sim T^{d+2+3x}. 
\]  

For \( x = 1 \) and \( d = 2 \), this scales as \( T^7 \) which is indeed small compared to the \( T^8 \) predicted in the same regime for the longitudinal scattering.

Additionally, we highlight in Sec. [VF.6] through numerical evaluations, the strong momentum-orientation dependence of \( \mathcal{W}^{\ominus} \).

4. Transverse scattering: thermal Hall resistivity

We would like to emphasize that within any scattering mechanism of phonon thermal Hall effect, the skew
scattering rate is a more fundamental measure of chirality of the phonons than the thermal Hall conductivity. This is because the Hall conductivity inevitably involves the combination of the skew and longitudinal scattering rates (in the form $\tau^2/\tau_{\text{skew}}$), and the longitudinal scattering rate of phonons has many other contributions that do not probe chirality, and may have complex dependence on temperature and other parameters that obscure the skew scattering. The scaling of the temperature dependence of $1/\tau_{\text{skew}}$, given above is a much more reliable prediction than any corresponding one made for $\kappa_H$ for this reason, and we do not quote the latter here. Instead, to extract the skew scattering rate, one should look at the thermal Hall resistivity, $\varrho_H$, which is simply proportional to $1/\tau_{\text{skew}}$, at least in the simplest view where the angle-dependence of the longitudinal scattering does not spoil its cancellation.

We define the thermal Hall resistivity tensor as usual by the matrix inverse, $\varrho = \kappa^{-1}$. In particular, considering the simplest case of isotropic $\kappa^{\mu\nu} \to \kappa_L$ and $\kappa_L \gg \kappa_{\mu\neq\nu}$, one thus has

$$\varrho^\mu_\nu = \frac{\varrho_{\mu\nu} - \varrho_{\nu\mu}}{2} \approx \frac{-\kappa_{\mu\nu} + \kappa_{\nu\mu}}{2\kappa_L} = -\frac{\tau^\mu_\nu}{\kappa_L}. \quad (97)$$

The quantity $\varrho^\mu_\nu$ is independent of the scale of the longitudinal scattering, in the sense that under a rescaling $D_{nk} \to \zeta D_{nk}$, then $\varrho^\mu_\nu$ is unchanged (see indeed Eqs. (96) for an explicit check at leading perturbative order).

As explained before, let us further assume that $D_{nk} = 1/\tau$ is $(n,k)$-independent, e.g. as if the case if dominated by some extrinsic effects. In that case, we can extract the longitudinal dependence from the transverse conductivity kernel, and redefine $\bar{K}^H_{n,kn',k'} = \tau^{-2}K^H_{n,kn',k'}$, which is now independent of the longitudinal scattering rate $\tau^{-1}$. Besides, to leading order one has simply $\bar{K}^H_{n,kn',k'} = \tau\delta^n_{n'}\delta_{k}^{k'}\omega_{nk}$, from which, assuming $\omega_{nk} = \nu_{ph}|k|$ and $N_{\text{eq}} = n_B$, we have simply

$$\kappa_L = \frac{\hbar^2}{k_B T^2 V} \sum_{nk} e^{\delta_{nk}}(J^\alpha_{nk})^2 = \tau c_v \frac{\nu_{ph}^2}{3}, \quad (98)$$

where by construction the result does not depend on the chosen direction $\alpha$ of the current (for instance $\alpha = x, y, z$). This is the well-known relation between the thermal conductivity $\kappa_L$ and the thermal capacity $c_v = \frac{\partial}{\partial T} \left[ \frac{1}{V} \sum_{nk} N_{\text{eq}} h \omega_{nk} \right] = k_B \frac{2\pi^2}{5} \left( \frac{k_B T}{\hbar \nu_{ph}} \right)^3 \quad (99)$

of the phonon gas. Consequently, Eq. (97) evaluates to

$$\varrho^\mu_\nu = -k_B^{-1} \left( \frac{15\nu_{ph}}{2\pi^2} \right)^2 \left( \frac{\hbar}{k_B T} \right)^8 \times \frac{V}{(2\pi)^6} \sum_{nk'k''} J^\mu_{nk} \bar{K}^H_{nk'k''} J^\nu_{n'k''}. \quad (100)$$

This expression does not depend on $\tau$, which justifies studying $\varrho_H$ instead of $\kappa_H$. From it and Eq. (31), one can readily derive the scaling relation

$$\varrho_H \sim 2\varrho_{\text{scaling}} \sim T^{d-1+3x}, \quad (101)$$

which is verified numerically in Sec. V F 7.

**F. Numerical results**

1. **Implementation**

Details about the numerical implementation are given in Appendix E.3. In short, we use C together with (i) the Cubature library to perform the one-dimensional momentum integrals (appearing in the definitions of $D^{(1)}_{nk}$, Eq. (80)), (ii) the Cuba library to perform multi-dimensional integrals (in $\kappa^\mu_\nu$, Eq. 3, and in $\varrho^\mu_\nu$, Eq. 100).

2. **Choice of parameters**

(i). **Polarization vectors** In Eq. (72), $\mathcal{L}$ is the trace over the product of the coupling matrix $\lambda$, with matrix elements $\lambda^{\alpha\beta}$, and that, $\mathcal{S}$, which determines the structure of the strain tensor and has matrix elements

$$S^{\alpha\beta}_{nk} = \frac{k^{\alpha}(\varepsilon^{\beta}_{nk})^q + k^{\beta}(\varepsilon^{\alpha}_{nk})^q}{\sqrt{\omega_{nk}}}. \quad (102)$$

Values of $(n,k)$ such that this factor vanishes correspond to phonons which are not coupled to the magnons, and whose longitudinal conductivity is solely driven by $D_{nk}$, i.e. other scattering effects. While this may indeed happen in practice, to highlight the effects of phonon-magnon scattering we choose a basis of polarization vectors $(\varepsilon_0,k,\varepsilon_1,k,\varepsilon_2,k)$ such that this is never the case (at least for $\alpha = \beta$, as with $\Lambda_{1,5}$ which are much larger than $\Lambda_{0,7}$).

These polarization vectors enforce $\varepsilon_{n,-k} = \varepsilon^{*}_{nk} = -\varepsilon_{nk}$ (so that $S^{\alpha\beta}_{n,k} = S^{\alpha\beta}_{nk} = -S^{\alpha\beta}_{-nk}$) as well as the tetragonal symmetry of the crystal, as required by the general theory of elasticity [34]; explicit expressions are given in App. E 2b.

(ii). **Extrinsic phonon scattering rate** For similar reasons, the extrinsic phonon scattering rate is taken to be $D_{nk} \to \gamma_{\text{ext}}$, a constant independent of $(n,k)$ and small compared with the typical $D_{nk}$ as soon as $T > T_3$ (see Sec. V E 2). In very clean monocrystals and in the absence of any other phonon scattering events, $\gamma_{\text{ext}} \sim \nu_{ph}/L$ reduces to the rate at which phonons bounce off the boundaries of the sample (of size $L$).

(iii). **Phonon dispersion** The phonon dispersion relation is chosen linear, $n$-independent and isotropic, $\omega_{nk} = \nu_{ph}|k|$, so that the different regimes of scaling exponents $x$ appear clearly.
We express our numerical results in units where $\Lambda^\text{code} = 1$, $v_B^\text{code} = 1$, $v_{ph}^\text{code} = 1$ and with unit lattice spacing $a^\text{code}$. Then, the mass of the unit cell $M_{uc}$ is expressed in units of $M_0 = \frac{a}{v_{ph} a}$ and is typically large—of the order $M_{uc} \sim 10^4 M_0$. $T$ is expressed in units of $T_0 = \frac{v_{ph}}{k_B a}$ and should verify $T/T_0 \lesssim 1$ so that the assumption of linearly dispersing phonons is correct. Correspondingly, we can define an energy $\epsilon_0 = k_B T_0$, and the isotropic part of the exchange $J$ is expressed in units of $\epsilon_0$.

The magnon velocity is fixed according to linear spin wave theory, which gives $v_m = 2\sqrt{\gamma J a}/\hbar$, with $J$ the isotropic magnetic exchange constant. We take $d = 2$ and $S = 1/2$: moreover, it is known that for $S = 1/2$ there is a renormalization factor $Z \approx 1.2$ enhancing the velocity, so that $v_m/v_{ph} \approx \sqrt{2\gamma J/\epsilon_0} \approx 1.71/\epsilon_0$ in our units. Since, for isotropic exchange, $\chi = \frac{\Lambda}{4\pi^2}$, we also take $\chi_0 = 4/(J/\epsilon_0)$.

Spin-phonon couplings $\Lambda_{1,7}$ are expressed in units of $\epsilon_0/a = \hbar v_{ph} a^2$. We describe a possible microscopic mechanism for spin-strain coupling in App. F1 where we show that $\Lambda_{1,5}$ typically arise as derivatives of the isotropic exchange constants. Since the latter ultimately arises from the overlap of atomic wavefunctions, which vary over distances of the order of $a_B$ the Bohr radius, we expect $\Lambda_{1,5} \sim J/a_B$. Meanwhile $\Lambda_{6,7}$ come from anisotropic exchanges and are thus expected to be considerably smaller.

Since the differences $\Lambda_{1,2} - \Lambda_3$ and $\Lambda_4 - \Lambda_5$ are due to anisotropic exchanges, they are chosen a fraction of a $\Lambda^{(i)}$. Since these magnetoelastic couplings typically arise as derivatives of magnetic exchange, we also take $\Lambda_i^{(m)} \approx -\Lambda_i^{(n)}$ for $i = 1, 7$; see App. F1 for a detailed derivation.

Scattering rates $D_{nk}$ and $\gamma_{\text{ext}}$ are expressed in units of $\gamma_0 = v_{ph}/a$, and we assume $\gamma_{\text{ext}}$ to be small, of the order of $1/L$ with $L$ the size of the sample—typically $\gamma_{\text{ext}} \sim 10^{-5} v_{ph}/a$. Finally, thermal conductivities are expressed in units of $\kappa_0 = k_B v_{ph}/a^2$.

For numerical calculations, we kept most dimensionless materials parameters (e.g. the ratio of $v_m$ and $v_{ph}$) fixed and constant, with the values expressed in Table VI. Those parameters for which we explore a given range of values are given in the captions of the figures in the following subsections. The fixed values are loosely inspired by Copper Deuterofomate Tetradueterate (CFTD), a square lattice $S=1/2$ antiferromagnet which has been extensively studied via neutron scattering [55, 57] due to its convenient scale of exchange which suits such measurements. For our purposes, CFTD has the desirable attribute that the magnon and phonon velocities are comparable (based on an estimate of the sound velocity from the corresponding hydrate [55]), which creates a significant phase space for magnon-phonon scattering. By contrast, in Li$_2$CuO$_4$, $v_m$ is much larger than $v_{ph}$.

### 3. Units and numerical values

| $v_m$ | $v_{ph}$ | $\gamma$ | $M_{uc}$ | $M_0$ | $T_0$ | $\epsilon_0$ | $\chi_0$ | $\kappa_0$ |
|------|----------|---------|---------|-------|-------|-----------|---------|---------|
| 0.05 | 0.05 | 0.05 | 0.05 | 0.05 | 0.05 | 0.05 | 0.05 | 0.05 |

**TABLE VI: Numerical values of the fixed parameters used in all numerical evaluations, expressed in the units given in Table VII**. The upper and lower entries for $m_0$ and $m_5$ correspond to the two cases for calculating $\theta_H$ and $\theta_H^2$, respectively.

Finally, note that the following scaling relations for $\kappa_L$,

$$\kappa_L \left( \{\Lambda_{1,7}\}, \gamma_{\text{ext}} \right) = \zeta^2_0 \kappa_L \left( \{\zeta_0 \Lambda_{1,7}\}, \zeta_0^2 \gamma_{\text{ext}} \right), \quad (103)$$

and for $\theta_H$,

$$\theta_H \left( \{\Lambda_{1,5}\}, \{\Lambda_{6,7}\}, m_0^{y,z} \right) = \zeta_1^{-3} \zeta_2^{-1} \zeta_3^{-1} \theta_H \left( \{\zeta_1 \Lambda_{1,5}\}, \{\zeta_2 \Lambda_{6,7}\}, \zeta_0 m_0^{y,z} \right) + O \left( \left(m_0^{y,z}\right)^3 (\Lambda_{6,7})^3 \right), \quad (104)$$

hold for any rescaling factors $\zeta_{0,..,3}$. Eqs. (103)-(104) make it possible to extrapolate results from our calculations for values of the parameters which are not explicitly explored in Table VII and Figs. 1(a), 1(b), 2(a), and 2(b).

### 4. Results for $D_{nk}$

Fig. 1(a) shows the angular dependence of $D_{nk}$. Throughout this section, we use $k = k(\cos \phi \uhat_x + \sin \phi \uhat_y)$, with $\phi \in [0, 2\pi]$, and $k_z = k \cos \psi$ with $\psi \in [0, \pi]$. Note that, in turn, $k^2 = k^2 \sin^2 \psi$. Also, since all the results are invariant under $k_z \to -k_z$, i.e. $\psi \to -\psi$, we plot results for $\psi \in [0, \pi/2]$ only.

(i) In-plane $\phi(k)$ angular dependence. We see from Fig. 1(a) that phonon-magnon scattering is typically larger for values of $\phi(k)$ associated with high-symmetry axes of the system, i.e. $\phi = 0 \mod \pi/2$. This is investigated from the structure of $\mathcal{L} = \text{Tr} [\mathbf{H}^T \mathbf{S}]$ in Eq. (72), which enters the magnetoelastic coupling, Eq. (65). The latter is by definition invariant under all symmetries of the
crystal, so that components of the strain tensor couple to functions of the magnetization fields \( m, n \) with the same symmetries.

Now, while the symmetry group of the crystal structure is tetragonal, the \( C_4 \) symmetry is spontaneously broken by the antiferromagnetic order along the \( z \) axis, while the \( C_2 \) and mirror symmetries are preserved when the magnetic field is along the \( z \) axis. More precisely, how does the \( C_4 \) symmetry (as acting on the \( \alpha, \beta \) indices) break? Since the \( S \) factor in Eq. \((72)\) has the same structure as the strain tensor itself, it preserves \( C_4 \); therefore the latter can only be broken in the \( \Lambda \) factor. Let us focus on the \( \lambda_{mm}, \lambda_{nn} \) cases, since these coefficients can be nonzero in the absence of a net magnetization \( m_0 \). A broken \( C_4 \) symmetry then means that \( 0 \neq \lambda'_{\xi,\alpha} := \lambda_{\xi,\xi} - \lambda_{\xi',\xi'} \). By inspection of Eqs. \((66)\) and \((69)\), one sees that there are two ways the latter can be nonzero: (1) in the \( \xi = \eta \) channel, \( \lambda'_{\xi,\alpha} \) is proportional to anisotropic exchanges; and (2) in the \( \xi = \mu \) channel, \( \lambda'_{\mu,\alpha} \) contains both isotropic and anisotropic exchange constants, and is consequently much larger than \( \lambda'_{\nu,\alpha} \). From this analysis, it follows that the deviation from \( C_4 \) symmetry as captured in \( D_{nk}^{(1)} \) by \( \lambda'_{\xi,\alpha} \) is largest for values of \( |k| \) where the \( \lambda_{mm} \) contributions dominate over the \( \lambda_{nn} \) ones, i.e. at large \( |k| \) (recall Eq. \((90)\)). One can check that this is indeed the case, as is shown in Appendix \( F.4 \).

(ii). Out-of-plane \( \psi(k) \) angular dependence. The out-of-plane angular dependence illustrates quite clearly the dynamical constraints satisfied by \( D_{nk} \), as outlined in Sec. \( V.B.2 \)

By inspection of Eq. \((62)\), we define

\[
\begin{align*}
\psi_- &= \arctan \left( \frac{v^{-1}_{ph} \sqrt{v^2_{ph} - v^2_m}}{v_{m}^{-1} \sqrt{v^2_{ph} - 4\Delta'/|k|^2}} \right), \\
\psi^{(1)}_+ (|k|) &= \arcsin \left( \frac{v_{m}^{-1} \sqrt{v^2_{ph} - 4\Delta'/|k|^2}}{v_{ph}^{-1} \sqrt{v^2_{ph} - 4\Delta'/|k|^2}} \right), \\
\psi^{(2)}_+ (|k|) &= \arcsin \left( \frac{v_{m}^{-1} \sqrt{v^2_{ph} - 4\Delta'/|k|^2}}{v_{ph}^{-1} \sqrt{v^2_{ph} - 4\Delta'/|k|^2}} \right),
\end{align*}
\]

where \( \Delta' = \max(\Delta_0, \Delta_1) \). Note that outside the domain of definition of \( \cdots \), by continuity one fixes \( \psi^{(1,2)}_+ := 0 \). The figure Fig. \( 1(a) \) can then be divided in four areas as follows:

- The vertical black band at angles \( \psi(k) \in [\psi^{(1)}_+ (|k|), \psi_-] \) corresponds to values of \( (k_z, |k|) \) such that energy and momentum conservation cannot be satisfied simultaneously because of the magnon gap \( \Delta \); therefore \( D_{nk}^+ = 0 = D_{nk}^- \).
- For angles \( \psi(k) > \psi_- \), scattering of the “ph+m \to m” type becomes possible, i.e. \( D^+ > 0 \). Meanwhile, following Eq. \((62)\), \( D^- = 0 \).
- Conversely, for \( \psi(k) < \psi^{(2)}_+ (|k|) \), scattering of the “ph \to m+m” type becomes possible, i.e. \( D^+ > 0 \), while \( D^- = 0 \).
- For \( \psi \in [\psi^{(2)}_+, \psi^{(1)}_+] \), scattering of the “ph \to m+m” type is possible only in the valley with the smallest gap, while in the other no scattering can happen; therefore, in that region \( D^+ > 0 \) but its value drops (without vanishing a priori) at the interface \( \psi(k) = \psi^{(2)}_+ (|k|) \).
(iii). Dependence on $|k|$. In Fig. 3(b), we show the dependence of $D_{nk}$ as a function of the norm $|k|$ and the out-of-plane angle $\psi$. This plot displays divergences near the singular lines $\psi^{(1,2)}$, $\psi_-$, which can be attributed to the thresholds for magnon scattering just above the gaps.

The angular width $\delta\psi(k)$ of the two black and darker regions bounded from the right by $\psi_-$, where scattering is forbidden in at least one of the two valleys, varies with $|k|$. From Eq. (1), we see that this width scales like $\delta\psi \sim k^2/|k|^2$ in our units. These regions extend down to $|k| = 0$, reflecting the fact that phonons with too little energy are unable to excite magnon pairs. The momentum magnitude thresholds for the excitation of magnon pairs are naturally given by $k_1 = 2\Delta/v_{ph}$ and $k_2 = 2\Delta'/v_{ph}$.

5. Results for $\kappa_L$

Numerical results are displayed in Figs. 2(a), 2(b).

Fig. 2(a) shows plots of $\kappa_L(T)$ for several values of extrinsic scattering $\gamma_{ext}$. This figure exhibits all the behaviors described in Secs. 5 and 5.1, with the extra feature that here there are two crossovers $T_{\Delta,0}$ and $T_{\Delta,1}$ defined by the two different magnon gaps $\Delta_0, \Delta_1$ whose values we give in Tab. VI. These are more clearly visible in Fig. 2(b), where we show $\kappa_L(T)$ in a small window of low temperatures and for smaller values of $\gamma_{ext}$.

Four scaling regimes can then be identified:

(i). For $T \lesssim T_{\Delta,1}^*$, only extrinsic scattering contributes to the full phonon scattering rate, and $\kappa_L \propto T^3/\gamma_{ext}$.

(ii). For $T_{\Delta,1}^* \lesssim T \lesssim T_{\Delta,0}^*$, both the extrinsic and the $x = -1$ phonon-magnon (only in the $\ell = 1$ valley) scattering rates contribute with the same scaling exponent, yielding $\kappa_L \propto T^3$ with a smaller proportionality coefficient than in the first regime.

(iii). For $T_{\Delta,0}^* \lesssim T \lesssim T_{\Delta}^*$, both the extrinsic and the $x = -1$ phonon-magnon (now in both valleys) scattering rates contribute with the same scaling exponent, yielding $\kappa_L \propto T^3$ with yet a smaller proportionality coefficient.

(iv). For $T > T_{\Delta}^*$, the $x = +1$ phonon-magnon scattering rate is dominant and yields $\kappa_L \propto T^{-1}$. Note that $T_{\Delta}^*$ is defined in Eq. (93) in the $D_{nk} = 0$ case; here by $T_{\Delta}^*$ we mean the more general crossover temperature in the presence of a finite $D_{nk} = \gamma_{ext}$.

The exponents quoted above are found with very good accuracy from a log-log scale plot (see inset of Fig. 2(a) and Appendices), regardless of the value of $\gamma_{ext}$; in that sense these exponents are universal. The influence of (non-universal) $\gamma_{ext}$ on the results of Fig. 2(a) is essentially threefold:

- Since the full phonon scattering rate is $D_{nk} = \gamma_{ext} + D_{nk}^{(1)}$, unsurprisingly $\kappa_L(T)$ is always a decreasing function of $\gamma_{ext}$.

- The “bumps” at $T \sim T_{\Delta,\ell}^*$ come from the fact that the $x = -1$ phonon-magnon scattering rate is much larger than $\gamma_{ext}$ as soon as the gap permits this scattering process; therefore, for large enough $\gamma_{ext}$, this feature disappears. More precisely, one should compare $\gamma_{ext}$ with $D_{nn,\ell} := \eta f^2 \Delta_\ell/M_{\text{uc}}$, where the dimensionless parameters $\eta, f$ are defined by $\lambda_{nn} \simeq \eta \lambda_{mm}$ and $\Lambda_{1,5} \simeq f/J/a$. The first bump is noticeable if $\gamma_{ext} \lesssim D_{nn,1}$, and the second bump is noticeable if $\max(\gamma_{ext}, D_{nn,1}) \lesssim D_{nn,0}$.

- Since $\gamma_{ext}$ and the $\lambda_{nn}$ coupling lead to the same scaling exponent, the $T \sim T_{\Delta}^*$ crossover results from a competition between $\lambda_{mm}$ on the one hand and $(\gamma_{ext}, \lambda_{nn})$ on the other; thus the larger $\gamma_{ext}$, the greater the dependence of $T_{\Delta}^*$ on $\gamma_{ext}$, and $T_{\Delta}^*(\gamma_{ext})$ is an increasing function of $\gamma_{ext}$.

6. Results for $2\mathfrak{M}_{nk/on/k'}$

Although the angular dependences of the $2\mathfrak{M}_{nk/on/k'}$ skew-scattering rates are more intricate than those of $D_{nk}$, a few general remarks can be made. In particular, in Fig. 3 where we plot $2\mathfrak{M}_{on/to}$ as a function of $\psi$ and $\theta$ at fixed $|k'| = 0.8/a$, $k_x = 0.2/a$, $k_z = 0.1/a$ (and temperature $T = 0.5T_0$), we have:

- Although $k_z \neq 0$, we can still take advantage of the $k' \leftrightarrow -k'_x$ symmetry, and it is sufficient to consider $\psi(k') \in [0, \pi/2]$. This comes from the fact that, for purely planar magnons, the phonon momenta $k_z, k'_z$ are not coupled. Meanwhile there is a priori no $\theta(k, k') \leftrightarrow -\theta(k, k')$ symmetry except when $k$ is along one of the high-symmetry axes of the crystal, as is the case here (cf. $k_y = 0$).

- The vertical black line at $\psi(k') = \psi_-$ can still be identified, and corresponds to magnons being gapped as in $D_{nk}^{(1)}$. However, in $2\mathfrak{M}$, the width and position of the gapped (black) zone now depend also on $\theta(k, k')$, due to the second energy conservation constraint in $2\mathfrak{M}$ (a feature absent in $D^{(1)}$ where there is only one energy constraint).

- In Appendix 4 we explore other orientations of in-plane $(k_z, k_y)$, and show that the features of $2\mathfrak{M}_{nk/on/k'}$ quoted above still hold. This is consistent with the above observations being consequences of the energy conservation constraints, which depend only of the relative angle $\phi(k) - \phi(k')$ since both phonon and magnon dispersions are isotropic in the $xy$ plane.

- In Fig. 3 $2\mathfrak{M}_{nk/\ell}$, also seems to vanish along certain special lines, especially those located at $\theta(k, k') = 0, \pi/2, 3\pi/2, 2\pi$. These features are not independent of the orientation $\phi(k)$; in fact they are salient features of the in-plane momenta.
FIG. 2: Longitudinal thermal conductivity $\kappa_L$ (in units of $\kappa_0 = k_B v_{ph}/a^2$) with respect to temperature $T$ (in units of $T_0 = \hbar v_{ph}/(ak_B)$), for four different values of $\gamma_{ext}$. (a) $\gamma_{ext} = 1 \times 10^{-3} (v_{ph}/a)$, $z \in [4,7]$, from darker ($z = 4$) to lighter ($z = 7$) shade. The dashed gray line indicates the evolution of the crossover temperature $T^*_\Delta$ as a function of $\gamma_{ext}$. Inset: log-log plot; the scaling behaviors are consistent with the analysis presented in the text. The inset is reproduced in App. F4. (b) $\gamma_{ext} = 1 \times 10^{-3} (v_{ph}/a)$, $z \in [6,9]$, from darker ($z = 6$) to lighter ($z = 9$) shade. The two crossover temperatures $T^*_{\Delta,1}$ and $T^*_{\Delta,0}$ are defined in the text up to a prefactor; here we identify the corresponding features in $\kappa_L$ but do not indicate specific values of $T$. See App. F4 for a log-log plot.

FIG. 3: Skew-scattering rate $\mathcal{W}_{\Delta}^{\phi,qq'}/\gamma_0$ as a function of $\psi(k') \in [0,\pi/2]$ (horizontal axis) and $\phi(k') = \phi(k') - \phi(k)$ (vertical axis) for fixed $|k'| = 0.8/a$, $k_x = 0.2/a$, $k_y = 0$, $k_z = 0.1/a$, $m_0 = 0.05 \hat{z}$ and temperature $T = 0.5 T_0$. Other parameter values are explored in App. F4. Note that the colorbar is not scaled linearly.

Finally, we point out that the values of $\mathcal{W}_{\Delta}^{\phi,qq'}$ in Fig. 3 are small compared to the values of $D^{(1)}$ obtained for similar values of momenta. This can be understood from the combination of (1) the anti-detailed-balance structure of $\mathcal{W}_{\Delta}^{\phi,qq'}$, from which it follows that $\mathcal{W}_{\Delta}^{\phi,qq'} + \mathcal{W}_{\Delta}^{\phi,qq'} = O(m_0)$ as shown in Sec. [IV A] and (2) the $C_2$ symmetry of the system around the $\hat{z}$ axis, which (since for planar magnons $k_z \leftrightarrow -k_z$ is a symmetry) entails $\mathcal{W}_{\Delta,qq''} = \mathcal{W}_{\Delta,qq''}$. Thus $\mathcal{W}_{\Delta,qq''} = O(m_0)$ itself. This, together with the analysis given in Sec. [VC] showing that terms which are odd in $m_0$ are also proportional to anisotropic couplings, implies that $\mathcal{W}_{\Delta,qq''}$ is indeed typically much smaller than $D^{(1)}_{nk}$.

7. Results for $\varrho_H$

We evaluated numerically $\varrho_{H,\mu\nu}$ for both $\mu\nu = xy$ and $xz$, in both cases with a net magnetization $m_0$ oriented along $\rho$, the axis perpendicular to the Hall plane $\mu\nu$. Results are presented in Fig. 4.

The observed scaling, $\varrho_H \propto T^4$, is consistent with
magnons are explicitly two-dimensional, carrying energy in all three directions, and that $\rho_{xy}$ and $\rho_{xz}$ are of the same order of magnitude. This is remarkable; not investigated here.

We emphasize that the numerical values of $\rho_{xy}$ and $\rho_{xz}$ are of the same order of magnitude. This is remarkable in a layered system which has entirely different magnon dynamics in the $xy$ and $xz$ planes, in this case where magnons are explicitly two-dimensional, carrying energy only within $xy$ layers. It can be understood from the fact that here phonons are isotropic, carrying energy in all three directions, and that including $T$-odd scattering exists in all directions, therefore allowing a Hall effect in both the $xy$ and $xz$ directions.

It should finally be noted that in the present model of an antiferromagnetic order along axis $\hat{x}$ and with linearized spin waves, $\theta_H^{xy}$ remains equal to zero.

G. Discussion of the results in absolute scales

Here we discuss the absolute scales of $\kappa_L$, $\theta_H$ and $\kappa_H$ we obtain using the parameter values from Table VI and those in the figure captions. First it is instructive to estimate the basic scales for thermal conductivity and temperature derived from phonons, which define the scales for our numerical plots. Using the phonon velocity for CFTD, $v_{\text{ph}}^{\text{CFTD}} = 4 \cdot 10^3$ m-s$^{-1}$ and its in-plane lattice parameter $a^{\text{CFTD}} = 5.7 \cdot 10^{-10}$ m, we find (see Table VI,

- $\kappa_0^{\text{CFTD}} = 0.17$ W-K$^{-1}$-m$^{-1}$,
- $T_0^{\text{CFTD}} = 54$ K,
- $\gamma_0 = 7.0 \cdot 10^{12}$ Hz.

Note that these scales do not vary greatly for many materials. For example, in La$_2$CuO$_4$, we find $\kappa_0^{\text{LCO}} = 0.38$ W-K$^{-1}$-m$^{-1}$ and $T_0^{\text{LCO}} = 80$ K. Importantly, the scale $\kappa_0$ is order one in SI units, which allows a roughly direct comparison with most data.

Next we can use the actual computed values to see what this mechanism predicts for the “test” material CFTD. We have at $T \approx 0.5 T_0 \approx 27$ K.

- $\kappa_L^{\text{CFTD}} \approx 75 \kappa_0 \approx 13$ W-K$^{-1}$-m$^{-1}$ for any of the $\gamma_0$ values presented in Fig. 2(a),
- for $\gamma_0 = 10^{-4} \cdot 10^{0}$, $T_0^{\text{LCO}} \approx 0.3 T_0 \approx 16$ K,
- for $\gamma_0 = 10^{-7} \cdot 10^{0}$, $T_0^{\text{LCO}} \approx 0.1 T_0 \approx 5.4$ K,
- $\theta_H^{\text{CFTD}} \approx 5 \cdot 10^{-6} \gamma_0 \approx 2.9 \cdot 10^{-5}$ K-m-W$^{-1}$,
- $|\theta_H^{\text{CFTD}}| \approx 3.8 \cdot 10^{-4}$,
- $|\kappa_H^{\text{CFTD}}| \approx 1.1 \cdot 10^{-2}$ W-K$^{-1}$-m$^{-1}$.

Note that $\kappa_L$, $\kappa_H$ and $\theta_H$ all depend on the choice of values for $\gamma_0$.

VI. CONCLUSIONS

A. Summary of results and method

In this paper, we studied the problem of scattering of phonons due to a weak intrinsic (i.e. without disorder) coupling to a fluctuating field $Q$, which is itself a quantum mechanical degree of freedom. Using the T-matrix formalism, we derived the scattering rates of phonons up to fourth order in coupling. The result is expressed generally, without any assumptions on the nature of the fluctuating field (i.e. it can be highly non-Gaussian), in terms of correlation functions of $Q$. Using these scattering rates in the Boltzmann equation leads to general expressions for the thermal conductivity tensor, and, when symmetry allows, a non-vanishing thermal Hall effect. A central result is that the skew scattering of phonons (which we define sharply as a scattering component which obeys an anti-detailed balance relation), and hence the thermal...
Hall conductivity, is proportional to a four-point correlation function of $Q$, which we give explicitly. We highlight throughout the various constraints due to symmetry (both exact and approximate), unitarity, and thermal equilibrium.

As an illustration of the method, we applied these results to the case where the fluctuating field $Q$ arises from spin wave (magnon) excitations of an ordered two-sublattice antiferromagnet. We model the latter via standard spin wave theory, for which phase space constraints imply that the dominant contribution arises from bilayers in the creation/annihilation operators of the spin waves. We obtain a general formula for the second order and fourth order scattering rates in terms of the dispersion of phonons and magnons, and the spin-lattice coupling constants. To obtain concrete results, we focus in particular on the limit in which the relevant magnons are acoustic, and we assume tetragonal symmetry and two-dimensionality of the magnons (but we retain the three dimensionality of the phonons). Under these assumptions we obtain all the (seven) symmetry-allowed spin-lattice coupling interactions, and calculate the second order and fourth order scattering rates, and thereby the thermal conductivity, including a phenomenological parallel scattering rate of phonons due to other mechanisms, e.g. boundary and impurity scattering. The final formulae are evaluated via numerical integration for representative model parameters. We observe a number of distinct scattering regimes, which we identify with features in the longitudinal thermal conductivity. We obtain a non-vanishing thermal Hall effect, in agreement with general symmetry arguments. Please see Sec. V for details.

B. Relation to other work

While we are not aware of any general results on the intrinsic phonon Hall conductivity due to scattering, there are a number of complementary theoretical papers as well as some prior work which overlap a small part of our results. The specific problem of phonons scattering from magnons was studied long ago to the leading second order in the coupling by Cottam [31]. That work, which assumed the isotropic SU(2) invariant limit, agrees with our calculations when these assumptions are imposed. The complementary mechanism of intrinsic phonon Hall effect due to phonon Berry curvature was studied by many authors [17, 39, 41], including how the phonon Berry curvature is induced by spin-lattice coupling in Ref. [18]. The majority of recent theoretical work has concentrated on extrinsic effects due to scattering of phonons by defects [32, 45]. The pioneering paper of Mori et al. [7] in particular recognized the importance of higher order contributions to scattering for the Hall effect, and is in some ways a predecessor to our work.

C. General observations

While often times scattering is regarded as a process which destroys coherence and suppresses interesting dynamical phenomena, our work reveals that higher order scattering probes highly non-trivial structure of correlations. Due to the constraints of detailed balance, the skew scattering, appropriately defined, contains only contributions of $O(Q^4)$ and no terms of lower order in $Q$, and so can in principle directly reveal subtle structures in the quantum correlations, without a need for subtraction. Measurements of such skew scattering of phonons—which a priori include but are not limited to the thermal Hall effect—might therefore be considered a probe of the quantum material hosting those phonons. Taking advantage of this potential opportunity is a challenge to experiment, as well as to theory, which should interpret the results and predict systems to maximize the effects.

We would like to comment on the analysis of thermal Hall effect experiments in quantum materials. As is well-known, thermal Hall conductivity is generally a small effect. In particular, the dimensionless measure of the Hall angle, $\theta_H = \tan^{-1}(\kappa_H^y / \kappa_H^x)$ is always much less than $\pi/2$ by two or more orders of magnitude, even in systems where thermal Hall effect is lauded as “huge”. (An actually large thermal Hall angle ($\theta = O(1)$) is obtained only the quantum thermal Hall regime when phonons are ballistic and edge states dominate over the bulk phonon contributions, which is extraordinarily difficult to achieve.) For small $\theta_H$, the skew scattering contributions are perturbative to the thermal conductivity, i.e. proportional to the latter rate $1/\tau_{skew}$. Dimensional reasoning implies that therefore $\kappa_H \sim \tau^2 / \tau_{skew}$, where $\tau$ is the standard, non-skew scattering time. This means that the thermal Hall conductivity has a very strong dependence on $\tau$, which is often sample-dependent and of course grows with sample quality, implying that the thermal Hall conductivity is larger in cleaner samples.

This dependence also means that $\kappa_H$ itself, as well as the dimensionless Hall angle $\theta_H \sim \kappa_H/\kappa_L$ depend not only on the skew scattering but also the ordinary scattering. Since the latter receives contributions from many different mechanisms, which may themselves have strong temperature and field dependence, neither $\kappa_H$ itself nor $\kappa_H/\kappa_L$ are ideal quantities to examine to probe the physics of skew scattering. Instead, we suggest that the thermal Hall resistivity, $\rho_H \equiv -\kappa_H^y / \kappa_L^x$, is the quantity which is most easily interpreted physically. This quantity is independent of the non-skew scattering, at least when the latter is largely momentum-independent, and is always independent of the overall scale of non-skew scattering. The temperature and field dependence of $\rho_H$ is generally expected to be simpler than that of the other quantities, at least when phonon skew scattering is the dominant mechanism for the Hall effect. This expectation is true not only when the skew scattering is intrinsic, as studied here, but also for extrinsic skew scattering due to defects.
D. Future directions

Our general formalism can be applied very broadly. In particular, because it does not require any assumptions on the nature of the $Q$ correlations, it may be applied directly to exotic states, to quantum or classical critical points, or to situations in which the $Q$ field is a composite operator. We present an application to a spinon Fermi surface spin liquid in an upcoming paper. Apart from other specific applications which may be easily imagined, it would also be interesting to explore further how general properties of four-point correlations of $Q$ may be detected via phonon skew scattering. In particular, the correlations which enter the scattering rates are not obviously time-ordered, and we wonder if these might contain some information on many-body chaos (Ref. [3]).

Despite the generality of our formulation, it is still specialized in several ways. We consider only scattering contributions to the phonon Boltzmann equation. In general the interactions with fields $Q$ will both induce scattering and modify the dynamics of the phonons in a non-dissipative way, e.g. induce phonon Berry phases [18]. While we believe it is usually the case that scattering is dominant, a more complete treatment including both effects would be of interest. Furthermore, in this paper we fully “integrate out” the electronic degrees of freedom, and follow the distribution function of the phonons only. More generally, there are coupled modes of phonons and electronic states, and one can consider the distributions for these coupled modes. One expects such effects are important largely when there are resonances between phonons and electronic excitations. All these problems could be addressed via a Keldysh treatment of coupled quantum kinetic equations, which is an interesting subject for future work.

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Appendix A: Strain tensor

In Sec. VC we employ a continuum model of the spin-phonon system. The phonons themselves correspondingly derive from the theory of continuum elasticity, which has the Hamiltonian density

$$H_{el} = \frac{1}{2\rho} \Pi^2 + \frac{1}{2} C_{\alpha\beta\gamma\delta} \varepsilon^{\alpha\beta} \varepsilon^{\gamma\delta}. \quad (A1)$$

Here $\rho$ is the mass density and $C_{\alpha\beta\gamma\delta}$ is a rank four tensor of elastic constants, which can be taken to satisfy $C_{\alpha\beta\gamma\delta} = C_{\beta\alpha\gamma\delta} = C_{\alpha\beta\delta\gamma} = C_{\gamma\delta\alpha\beta}$. The canonical variables of this classical field theory are the displacement field $u_\mu$ and its canonically conjugate momentum $\Pi_\mu$. Due to translational and rotational symmetry, the Hookian potential energy is expressed solely through the strain tensor,

$$\varepsilon^{\alpha\beta}(\mathbf{R}) = \frac{1}{2} (\partial_\alpha u_\beta + \partial_\beta u_\alpha). \quad (A2)$$
By construction the strain is a symmetric tensor in its two indices, i.e. \( \mathcal{E}^T = \mathcal{E} \). Define the Fourier transforms

\[
u_{\mu}(x) = \frac{1}{\sqrt{V}} \sum_k e^{i k \cdot x} u_{\mu,k}, \quad \Pi_{\mu}(x) = \frac{1}{\sqrt{V}} \sum_k e^{i k \cdot x} \Pi_{\mu,k}.
\]

(A3)

Here since \( u_{\mu}(x) \) and \( \Pi_{\mu}(x) \) are real fields, we have \( u_{\mu,-k} = u_{\mu,k}^\dagger \) and \( \Pi_{\mu,-k} = \Pi_{\mu,k}^\dagger \). The Fourier space fields satisfy the commutation relations

\[
[\Pi_{\mu,k}, u_{\nu,k'}] = i \delta_{\mu \nu} \delta_{k+k',0}.
\]

(A4)

We obtain

\[
H_{el} = \sum_k \left\{ \frac{1}{2\rho} \Pi_{\mu,-k} \Pi_{\mu,k} + \frac{1}{2} \mathcal{K}_{\alpha \beta}(k) u_{\alpha,-k} u_{\beta,k} \right\},
\]

with

\[
\mathcal{K}_{\alpha \beta}(k) = C_{\alpha \beta \gamma \delta} k_{\gamma} k_{\delta}.
\]

(A6)

The matrix \( \mathcal{K}_{\alpha \beta} \) is by construction real and symmetric, and hence has real eigenvalues \( \mathcal{K}_n \), which additionally must be positive for stability. We define the eigenvalues and eigenvectors \( \varepsilon_n^\alpha \) via

\[
\mathcal{K}_{\alpha \beta}(k) \varepsilon_n^\beta(k) = \mathcal{K}_n(k) \varepsilon_n^\alpha(k),
\]

(A7)

with \( \varepsilon_n^\alpha(-k) = (\varepsilon_n^\alpha(k))^* \) and the standard normalization \( \sum_n (\varepsilon_n^\alpha(k))^* \varepsilon_n^\alpha(k) = \delta_{nn'} \). Now we make the change of basis

\[
u_{\mu,k} = \sum_n \varepsilon_n^{\mu\dagger}(k) u_{n,k}, \quad \Pi_{\mu,k} = \sum_n \varepsilon_n^{\mu}(k) \Pi_{n,k},
\]

(A8)

which gives

\[
[\Pi_{nk}, u_{n'k'}] = i \delta_{nn'} \delta_{k+k',0},
\]

(A9)

and

\[
H_{el} = \sum_{n,k} \left\{ \frac{1}{2\rho} \Pi_{\mu,-k} \Pi_{\mu,k} + \frac{1}{2} \mathcal{K}_n(k) u_{n,-k} u_{n,k} \right\}.
\]

(A10)

Now we can finally define creation/annihilation operators

\[
u_{nk} = \frac{1}{\sqrt{2}} (a_{nk} + a_{n,-k}^\dagger),
\]

\[
\Pi_{nk} = \frac{1}{\sqrt{2}} (a_{nk} + a_{n,-k}^\dagger),
\]

(A11)

with canonical boson operators

\[
[a_{nk}, a_{n'k'}^\dagger] = \delta_{nn'} \delta_{kk'},
\]

(A12)

and the Hamiltonian

\[
H_{el} = \sum_{nk} \omega_{nk} a_{nk} a_{nk}^\dagger.
\]

(A13)

and

\[
\omega_{nk} = \sqrt{\frac{\mathcal{K}_n(k)}{\rho}}.
\]

(A14)

Having finally arrived at the canonical phonon operators, we recombine the several steps of the above procedure to obtain the expression for the displacement field,

\[
u_{\mu}(x) = \frac{1}{\sqrt{V}} \sum_{nk} \frac{1}{2\rho \omega_{nk}} \left( a_{nk} + a_{n,-k}^\dagger \right) \varepsilon_{nk} \varepsilon^{\mu\dagger} e^{ik \cdot x}.
\]

(A15)

Now we can use the definition in Eq. (B6) of the strain to obtain

\[
\varepsilon_{\mu \nu}(x) = \frac{1}{\sqrt{V}} \sum_{nk} \frac{1}{2\rho \omega_{nk}} \left( a_{nk} + a_{n,-k}^\dagger \right) i/2 \left( k^\mu \varepsilon_{\nu nk} + k^\nu \varepsilon_{\mu nk} \right) e^{ik \cdot x}.
\]

(A16)

Now let us consider the coupling of the strain to the continuum spin fluctuations, Eq. (68) of the main text. The full spin-lattice coupling in three dimensions is written as

\[
H_{s-1} = \sum_z \int dx dy z_{s \alpha} \mathcal{E}^{\alpha \beta}(x) \lambda_{s,\xi \xi', n_0} \xi_{s,\alpha} \xi_{s,\beta}.
\]

(A18)

Note the sum over discrete 2d layers. We now insert the Fourier expansion of the strain from Eq. (A16) and the corresponding Fourier expansion of the magnetic fluctuations, which we repeat here:

\[
\xi_{s,\alpha} = \frac{1}{\sqrt{A_{2d}}} \sum_{q} \xi_{s,\alpha q} e^{iq \cdot z}.
\]

(A19)

In this equation, and in the rest of this section, we are careful to denote two-dimensional vectors with an underline. Since magnetic fluctuations in different layers are taken as independent, we do not introduce a z-component of the wavevector for the magnons, and simply leave z explicitly as a layer index for these fields. Note also the prefactor Eq. (A19) therefore involves the square root of the two dimensional area of a single plane, \( A_{2d} \).

With this in mind, we obtain from Eq. (A18)

\[
H_{s-1} = \frac{1}{\sqrt{V}} \sum_{z} \sum_{k,p} \lambda_{s,\xi \xi', n_0} \xi_{s,\alpha} \xi_{s,\beta} \left( a_{nk} + a_{n,-k}^\dagger \right) \xi_{s,\alpha} p - \frac{1}{2} k_z \xi_{s,\beta} p - \frac{1}{2} k_z \xi_{s,\beta}.
\]

(A20)

From here we can see that

\[
Q_{nk} = \frac{i}{2\sqrt{V}} \left( k^\mu \varepsilon_{\mu nk} + k^\nu \varepsilon_{\nu nk} \right) \sum_{\xi \xi', n_0} \lambda_{s,\xi,\xi', n_0} \xi_{s,\alpha} \xi_{s,\beta} \sum_{k,p} \xi_{s,\alpha} p - \frac{1}{2} k_z \xi_{s,\beta} p - \frac{1}{2} k_z \xi_{s,\beta}.
\]

(A21)
Next we use Eq. (70) to express this in terms of bosonons:

\[
Q_{nk} = \frac{i (k^\nu_{\ell k} + k^\nu_{nk})}{4\sqrt{2V} \rho \omega_{nk}} \sum_{P} \sum_{qq'} \sum_{\xi \xi'_{ab}} \lambda_{\xi \xi', n_0 - \xi - \xi'} e^{ik_{z'}z} (-1)^{\xi (\delta_{\tau,2} + \frac{1}{2} \tau') + \xi' (\delta_{\tau,1} + \frac{1}{2} \tau')} \frac{\xi}{\xi'} \times (\chi \Omega_{\delta_{\tau,2} - \frac{1}{2} \xi} \hat{\xi}'{2} (\chi \Omega_{\delta_{\tau,1} - \frac{1}{2} \xi'}) {2} b^q \cdot (\frac{1}{2} k, \delta_{\tau,c}, z) p - \frac{1}{2} k, \delta_{\tau,c}, z'.
\]

We now define \( \ell_1 = \delta_{\tau \xi} = 0,1 \) and \( \ell_2 = \delta_{\tau \xi} \), which is inverted by \( a = \xi \ell_1 + \xi \) and \( b = \xi' \ell_2 + \xi' \). This gives

\[
Q_{nk} = \frac{i (k^\nu_{\ell k} + k^\nu_{nk})}{4\sqrt{2V} \rho \omega_{nk}} \sum_{P} \sum_{qq'} \sum_{\xi \xi'_{ab}} \lambda_{\xi \xi', n_0 - \xi - \xi'} e^{ik_{z'}z} (-1)^{\xi (\delta_{\tau,2} + \frac{1}{2} \tau') + \xi' (\delta_{\tau,1} + \frac{1}{2} \tau')} \frac{\xi}{\xi'} \times (\chi \Omega_{\ell_1,1 - \frac{1}{2} \xi} \hat{\xi}'{2} (\chi \Omega_{\ell_2,1 - \frac{1}{2} \xi'}) {2} b^q \cdot (\frac{1}{2} k, \ell_1, z) p - \frac{1}{2} k, \ell_2, z'.
\]

From here, we recognize that \( Q_{nk} = \mathcal{Q}_{nk} \) in Eq. (54), and thereby extract \( \mathcal{B} \). We use \( V \rho = N_{uc} M_{uc} \).

**Appendix B: General hydrodynamics of phonons**

Our goal is to derive the thermal current carried by the phonons,

\[
j^\mu = \frac{1}{V} \sum_{nk} \overline{\mathcal{N}}_{nk} \phi_{nk} \omega_{nk}, \tag{B1}
\]

in order to extract the thermal conductivity tensor. This requires knowledge of the average phonon populations \( \overline{\mathcal{N}}_{nk} \), which, in presence of a gradient of temperature, differ from their equilibrium values. These populations can be obtained by solving Boltzmann’s equation

\[
\partial_t \overline{\mathcal{N}}_{nk} + \mathcal{V}_{nk} \cdot \nabla \overline{\mathcal{N}}_{nk} = \mathcal{C}_{nk} [\{ \overline{\mathcal{N}}_{n'k'} \}], \tag{B2}
\]

where the collision integral \( \mathcal{C}_{nk} [\{ \overline{\mathcal{N}}_{n'k'} \}] \) depends on the populations in all \( (n', k') \) states. To solve this equation, we expand the out-of-equilibrium populations around their equilibrium value as \( \overline{\mathcal{N}}_{nk} = N_{nk} + \delta \overline{\mathcal{N}}_{nk} \). Within linear response, the perturbations can be considered small and we may expand the collision integral

\[
\mathcal{C}_{nk} [\{ \overline{\mathcal{N}}_{n'k'} \}] = \mathcal{O}_{nk} + \sum_{n'k'} \mathcal{C}_{nk'n'} \delta \overline{\mathcal{N}}_{n'k'} + O(\delta \overline{\mathcal{N}}^2),
\]

around its value \( \mathcal{O}_{nk} \) in equilibrium. Since the thermal current must vanish in equilibrium, \( \mathcal{O}_{nk} \) must be zero (we go back to this statement in Sec. C2c). In Eq. (B3), the “collision matrix” \( \mathcal{C}_{nk'n'} \) is defined as the first-order Taylor coefficient, and one neglects the quadratic order in the perturbation. Formally inverting the collision matrix in the stationary Boltzmann equation (i.e. Eq. (B1) with \( \partial_t \overline{\mathcal{N}} = 0 \) leads to

\[
\delta \overline{\mathcal{N}}_{nk} = \mathcal{C}_{nk'n'}^{-1} \frac{\omega_{nk'n'}}{k_B T^2} (N_{nk'}^{eq})^2 \phi_{nk} \mathcal{V}_{nk} T. \tag{B4}
\]

From Eq. (B4) and Fourier’s law, we can identify the components of the thermal conductivity tensor:

\[
\kappa^{\mu\nu} = \kappa^{\nu\mu} = -\frac{1}{k_B T^2} \frac{1}{V} \sum_{nk'k} \frac{\omega_{nk'nk'} \phi_{nk} \phi_{nk'}}{D_{nk'}} \left( C_{nk'nk'} e^{\beta \omega_{nk'nk'}} (N_{nk'}^{eq})^2 \pm (nk \leftrightarrow n'k') \right). \tag{B5}
\]

This expression shows that a nonzero phonon Hall conductivity requires the factor in the second line to be non-zero, which is equivalent to

\[
C_{nk'nk'} e^{\beta \omega_{nk'nk'}} (N_{nk'}^{eq})^2 \neq C_{nk'nk'} e^{\beta \omega_{nk'nk'}} (N_{nk'}^{eq})^2 \tag{B6}
\]

where the constraint is now on \( C_{nk'nk'} \) instead of its inverse. In other words, only the antisymmetric in \( nk \leftrightarrow n'k' \) part of \( C_{nk'nk'} e^{\beta \omega_{nk'nk'}} (N_{nk'}^{eq})^2 \) contributes to the Hall conductivity.

In order to proceed further analytically, and invert the scattering matrix, we separate the diagonal from the off-diagonal parts in \( C_{nk'nk'} = -\delta_{n'k'} \delta_{nk} D_{nk} + M_{nk'nk'} \), and assume that \( D_{nk} \gg \sum_{nk'} M_{nk'nk'} \). This ought to be the case whenever the interactions are small, and/or if other damping processes are large. Then, \( C_{nk'nk'} \approx -\delta_{n'k'} \delta_{nk} D_{nk}^{-1} - M_{nk'nk'} D_{nk}^{-1} D_{nk'}^{-1} \). The antisymmetry in \( nk \leftrightarrow n'k' \) condition for the Hall conductivity mentioned above leads to the fact that the diagonal term contributes to the longitudinal conductivity, but not to the Hall part, and translates into

\[
\frac{\kappa^{\mu\nu} - \kappa^{\nu\mu}}{2} = -\frac{1}{k_B T^2} \frac{1}{V} \sum_{nk'k} \frac{\omega_{nk'nk'} \phi_{nk} \phi_{nk'}}{D_{nk'}} \left( D_{nk}^{-1} - M_{nk'nk'} \right) e^{\beta \omega_{nk'nk'}} (N_{nk'}^{eq})^2 - (nk \leftrightarrow n'k'). \tag{B7}
\]

The longitudinal conductivity is

\[
\kappa^{\mu\nu} = \frac{1}{k_B T^2} \frac{1}{V} \sum_{nk'k} \frac{\omega_{nk'nk'} \phi_{nk} \phi_{nk'}}{D_{nk'}} \left( \delta_{n'k'} M_{nk'nk'} \right) e^{\beta \omega_{nk'nk'}} (N_{nk'}^{eq})^2. \tag{B8}
\]

Note that we will include all other (diagonal) scattering processes not taken into account here (e.g. boundary
scattering, scattering by impurities, phonon-phonon scattering etc.) by adding a phenomenological relaxation rate $\tilde{D}_{nk}$ to the diagonal of the scattering matrix.

Appendix C: From interaction terms to the collision integral

1. General method and definitions

We now aim at deriving an expression for the collision integral of Boltzmann’s equation using kinetic theory methods. The probability for the system to be found in a given quantum state $|i\rangle = |i_p\rangle |i_s\rangle$ is governed by the master equation

$$\partial_t p_{i_p i_s} = \sum_{f_p f_s} \left[ \Gamma_{i_p i_s \rightarrow f_p f_s} p_{f_p f_s} - \Gamma_{i_p i_s \rightarrow f_p f_s} p_{i_p i_s} \right],$$  

(C1)

where we will compute the transition rates $\Gamma_{i_p i_s \rightarrow f_p f_s}$ using scattering theory. The probability of a phonon state $|i_p\rangle$ is then obtained by summing over all possible spin configurations of the system, $p_{i_p} = \sum_{i_s} p_{i_p i_s}$. Assuming the phonon and spin probabilities are independent, i.e. $p_{i_p i_s} = p_{i_p} p_{i_s}$, and defining the transition rates $\Gamma_{i_p \rightarrow f_p} = \sum_{i_s} \Gamma_{i_p i_s \rightarrow f_p f_s} p_{f_s}$ between phonon states only, we obtain the master equation for the probabilities of phonon states. We may in turn express the collision integral in the RHS of Boltzmann’s equation, which is given by the time evolution of the populations in each phonon state $|i_p\rangle$ through the definition

$$C_{nk} [\{N_{n'k'}\}] = \sum_{i_p} \sum_{f_p} N_{nk}(i_p) \partial_t p_{i_p},$$  

(C2)

where $N_{nk}(i_p) = \langle i_p | a_{nk}^\dagger a_{nk} | i_p \rangle$ is the number of $(n,k)$ phonons in the $|i_p\rangle$ state and $N_{nk} = \sum_{i_p} N_{nk}(i_p)$ is the average population. The only phonon states involved in the sums are those whose populations of $(n,k)$ phonons are different. Now, in order to obtain the transition rates between spin-phonon states, we use Fermi’s golden rule

$$\Gamma_{i_p i_s \rightarrow f_p f_s} = 2\pi |T_{i_p i_s \rightarrow f_p f_s}|^2 \delta (E_{i_p i_s} - E_{f_p f_s}),$$  

(C3)

where the factor $N_{uc}$ ensures that $\Gamma_{i_p i_s \rightarrow f_p f_s}$ is a finite quantity in the thermodynamic limit, consistent with the choice of $H'$ as a hamiltonian density. We use Born’s expansion of the scattering matrix

$$T_{i_p i_s \rightarrow f_p f_s} = \langle f_p f_s | H' | i_p i_s \rangle$$  

(C4)

where $H'$ is the (perturbative) interaction hamiltonian between the phonons and the $Q$ fields, and the $\eta \rightarrow 0^+$ appearing in the denominator of the second-order term ensures causality, which will prove crucial in the following.

To describe the interaction between phonon and spin degrees of freedom, we introduce general coupling terms between phonon creation-annihilation operators $a^\dagger_{nk}$ and general, for now unspecified, fields $Q_{\{n_k\}}$ which depend on the spin structure: denoting $\lambda^+_{nk} \equiv a_{nk}^\dagger$ and $\lambda^+_{nk} = a_{nk}$, and similarly for the $Q$ operators, $Q_{\{n_k\}}^\dagger \equiv Q_{\{n_k\}}$ and $Q_{\{n_k\}} \equiv Q_{\{n_k\}}$, we write the couplings

$$H'[1] = \sum_{nk} \sum_{q=\pm} a_{nk}^q Q_{n_k}^q,$$  

(C5)

$$H'[2] = \frac{1}{\sqrt{N_{uc}}} \sum_{n_k, n_{k'}' \pm q, q'} a_{nk}^q a_{n_k'q'}^q Q_{n_k n_k'}^{qq},$$  

(C6)

where $Q_{n_k n_k'}^{qq} = Q_{n_k n_k'}^{q-q'}$ ensures the hermiticity of $H'[2]$. Here and throughout the manuscript, a square bracket index, e.g. $[p]$ denotes the number of interacting phonons.

By definition the term $H'[1]$ involves $p$ phonon creation-annihilation operators, and as such typically arises from microscopic models as the $p$th spatial derivative of orbital overlaps. Consequently, we assume $H'[2]$ to be of the same order of magnitude as the square of $H'[1]$, that is to say, $Q_{n_k n_k'}^q \sim \lambda$, $Q_{n_k n_k'}^{qq} \sim \lambda^2$ with $\lambda$ a small parameter. In this paper, we keep only the first two terms of the expansion (i.e. we take $H' = H'[1] + H'[2]$).

2. Computation at first Born order

In this subsection we consider only the first term of Born’s expansion. The transition rates associated with $H' = H'[1] + H'[2]$ at this order are simply the matrix elements:

$$T^{[1]}_{1-2} = \sum_{nkq} \frac{\sqrt{N_{n_k}}} \sum_{q=\pm} \frac{q+1}{2} \langle f_s | Q_{n_k}^q | i_s \rangle \mathbb{I}(i_p \rightarrow f_p),$$  

(C7)

$$T^{[2]}_{1-2} = \frac{1}{\sqrt{N_{uc}}} \sum_{n_k, n_{k'}' \pm q, q'}$$  

$$\cdot \langle f_s | Q_{n_k n_k'}^{qq} | i_s \rangle \mathbb{I}(i_p \rightarrow f_p),$$  

(C8)

where $\mathbb{I}(i_p \rightarrow f_p)$ means that the only difference between $|i_p\rangle$ and $|f_p\rangle$ is that there is $q = \pm 1$ more phonon of species $(n,k)$ in the final state. Note that the cases where $n_k = n_{k'}'$ require a formal correction. However, at any given order in the $\lambda$ expansion, such terms are smaller than all others by a factor $1/N_{uc}$, where $N_{uc}$ is the number of unit cells, and therefore vanish in the thermodynamic limit. In this article we thus take $\sum_{n_k, n_{k'}'}$ and $\sum_{n_k \neq n_{k'}'}$ exchangeably, unless we specify otherwise.
We then compute the squared matrix element. "Cross terms" such as $\langle 1|H_{[2]}|f\rangle\langle f|H_{[1]}|1\rangle$ (which are of order $\lambda^3$) vanish because $\langle ip|\hat{A}|ip\rangle = 0$ for any operator $\hat{A}$ containing an odd number of phonon creation-annihilation operators. At order $\lambda^2$, there thus remains only $\langle 1|H_{[1]}|f\rangle\langle f|H_{[1]}|1\rangle$, and at order $\lambda^4$, only $\langle 1|H_{[2]}|f\rangle\langle f|H_{[2]}|1\rangle$.

\[ a. \] Terms at $O(\lambda^2)$

At order $\lambda^2$, we have therefore

\[ \left| T_{[1]}^{[1]} \right|^2 = \sum_{nkq} (N_{nkq} + \frac{q+1}{2}) \mathbb{I}(ip\frac{q+1}{2} f_p) \langle is|Q_{nk}^q|fs\rangle\langle fs|Q_{nk}^q|is\rangle. \] (C9)

We then enforce the energy conservation $\delta(E_f - E_i) = \delta(q\omega_{nk} + E_f - E_i)$ by writing the latter as a time integral, i.e. $\int_{-\infty}^{\infty} dt e^{i\omega t} = 2\delta(\omega)$, identify $A(t) = e^{+i\beta H t}e^{-i\beta H t}$, use the identity $1 = \sum f_{s}\langle fs|\langle fs\rangle$, and take the spins in the initial state to be in thermal equilibrium $p_{is} = Z^{-1}e^{-\beta E_{is}}$. Finally summing over $\langle is\rangle$ and identifying $\langle A\rangle_\beta = Z^{-1}\text{Tr}(e^{-\beta H A})$, we find

\[ W_{nkq}^{[1]} = 2\pi \sum_{fs,i_s} \langle is|Q_{nk}^q|fs\rangle\langle fs|Q_{nk}^q|is\rangle p_{is} \times \delta(q\omega_{nk} + E_f - E_i) \]
\[ = \int_{-\infty}^{\infty} dt e^{-\beta\omega_{nk}t} \langle Q_{nk}^q(t)\rangle\langle Q_{nk}^q(0)\rangle_\beta. \] (C10)

Note that this calculation, in a time-reversal symmetric system, leads to the extra symmetry $W_{nkq}^{[1]} = W_{nk-q}^{[1]}$.

The scattering rate between phonon states, for the one-phonon interaction term at first Born's order, then reads

\[ \Gamma_{[1]}^{[1]}_{ip\rightarrow fp} = \sum_{nkq} (N_{nkq} + \frac{q+1}{2}) W_{nkq}^{[1]} \mathbb{I}(ip\frac{q+1}{2} f_p). \] (C11)

To arrive at the collision integral, the final step involves summing over final phononic states $f_p$ and taking the average over initial phononic states $i_p$. We find, the contributions to $\mathcal{C}$ at order $\lambda^2$ to be:

\[ O_{nkq}^{[1]} = \sum_{q=\pm} q W_{nk,q}^{[1]} (N_{nkq} + \frac{q+1}{2}), \] (C12)

\[ -D_{nkq}^{[1]} = \sum_{q=\pm} q W_{nk,q}^{[1]} . \] (C13)

We will address the constant term $O_{nkq}^{[1]}$ (expected to be zero) in more detail in Sec. C2c. The collision matrix is clearly diagonal, i.e. $M_{nkq,k'q'}^{[1]} = 0$. Therefore this $\lambda^2$ contribution to $\mathcal{C}$ may contribute to the longitudinal conductivity, but not to the Hall conductivity.

\[ b. \] Terms at $O(\lambda^4)$

We address the $O(\lambda^4)$ term in a similar fashion. There, the energy conservation reads $\delta(E_f - E_i) = \delta(q\omega_{nk} + q\omega_{n'k'} + E_f - E_i)$, and we find

\[ \Gamma_{[2]}^{[2]}_{ip\rightarrow fp} = \frac{1}{2N_{nkq,k'q'\pm q=\pm}} \sum_{\pm q=\pm} W_{nkq,k'q'}^{[2]} \mathbb{I}(ip\frac{q+1}{2} f_p) \cdot \left( N_{nkq,k'} + \frac{q+1}{2} \right), \] (C14)

where

\[ W_{nkq,k'q'}^{[2]} = 2\int_{-\infty}^{\infty} dt e^{-i(q\omega_{nk} + q\omega_{n'k'})t} \] (C15)

and $W_{nkq,k'q'}^{[2]} = W_{nk-q,k'q'}^{[2]}$ by definition. The resulting collision integral, up to linear order in the perturbed populations $\delta N$ contains the following contributions:

\[ O_{nkq,k'q'}^{[2]} = \frac{1}{N_{nkq,k'q'}} \sum_{q=\pm} q W_{nkq,k'q'}^{[2]} \]
\[ \cdot \left( N_{nkq,k'} + \frac{q+1}{2} \right), \] (C16)

\[ -D_{nkq,k'q'}^{[2]} = \frac{1}{N_{nkq,k'q'}} \sum_{q=\pm} q \left( N_{nkq,k'} + \frac{q+1}{2} \right) W_{nkq,k'q'}^{[2]}, \] (C17)

\[ M_{nkq,k'q'}^{[2]} = \frac{1}{N_{nkq,k'q'}} \sum_{q=\pm} q \left( N_{nkq,k'} + \frac{q+1}{2} \right) W_{nkq,k'q'}^{[2]}. \] (C18)

As above, we will address the constant term in Eq. (C26).

The diagonal contribution is of order $\lambda^4$, and we therefore expect it to be subdominant compared with the $\lambda^2$ contribution from the previous section. Finally, the off-diagonal contribution is nonzero. However, we will show that its contribution to $C_{nkq,k'q'}(\beta\omega_{nk}) (N_{nkq,k'q'}^{eq})^2$ is purely symmetric under $nk\leftrightarrow n'k'$ and therefore contributes only to the symmetric off-diagonal conductivity but not to the Hall one — see Eq. (B6).

\[ c. \] Detailed balance

First, we notice that a change of variables $i_s \leftrightarrow f_s$ in Eq. (C10) leads to the detailed-balance relation

\[ W_{nkq}^{[1]} = W_{nk-q}^{[1]} e^{-\beta\omega_{nk}}. \] (C19)

An immediate consequence is that $O_{nkq}^{[1]} = 0$ if we take the equilibrium phonon population $N_{nk}^{eq}$ to be Bose-Einstein's distribution, as was physically required. Similarly, for the two-phonon interactions at first order, we find the detailed-balance relation

\[ W_{nkq,k'q'}^{[2]} = e^{-\beta(q\omega_{nk} + q\omega_{n'k'})} W_{nk-q,n'k'-q}^{[2]}. \] (C20)
Again, taking $N_{n'k'}^{eq}$ to be Bose-Einstein’s distribution implies $O_{nk}^{[2];[2]} = 0$. Moreover, the detailed-balance relation also implies

$$M_{nk,n'k'} e^{\beta \omega_{n'k'}} (N_{n'k'}^{eq})^2 = (n_k \leftrightarrow n'_{k'}) , \quad (C21)$$

i.e. there are no antisymmetric contributions, and hence no thermal Hall effect at first Born’s order. While we proved this explicitly for the one-phonon and two-phonon cases, this is true in general (along with $O_{nk} = 0$) for any number of phonon creation-annihilation operators at first order in Born’s expansion (see Sec. D2c).

### d. Extra structure

Independently, by writing

$$Q_{n'k'}(t)Q_{nk}(0) = \frac{1}{2} \{Q_{n'k'}(t), Q_{nk}(0)\} + \frac{1}{2} \{Q_{n'k'}(t), Q_{nk}(0)\} (C22)$$

it is straightforward to show that only the commutator term contributes to $W^{[2]}_{n'k',q} - W^{[1]}_{n'k',q}$. In turn, the final expression for the diagonal of the collision matrix Eq. \[C12\] takes the form of the spectral function:

$$D_{nk}^{[1];[1]} = -\int_{-\infty}^{+\infty} dt e^{-i\omega_{n'k'}t} \langle [Q_{n'k'}^{-}(t), Q_{nk}^{+}(0)] \rangle_{\beta} . \quad (C23)$$

In the two-phonon case, a commutator structure does not naturally appear, so that one is left with

$$D_{nk}^{[2];[2]} = -\frac{2}{N_{nk}} \sum_{n'k'} \sum_{q,q'} q \left( N_{n'k'}^{eq} + q' + \frac{1}{2} \right) \quad (C24)$$

$$\times \int dt e^{-i(\omega_{n'k'} + q' \omega_{n'k'})t} \langle Q_{n'k'}^{-q}(t)Q_{nk}^{q'}(0) \rangle_{\beta} ,$$

at order $\lambda^4$ and first Born’s order.

### 3. Energy shift of the phonons

We now address the constant term $O_{nk}$ appearing in the collision integral, which must vanish because there is no current in equilibrium. Its cancellation is equivalent to a redefinition of the energies of the phonons, due to their interaction with the $Q$ degrees of freedom. This energy shift corresponds to the real part of the associated self-energy. Consequently, the equilibrium phonon populations $N_{n'k'}^{eq}$ are a priori not equal to $N_{n'k'}^{BE}$, the Bose-Einstein populations for the unperturbed phonon energies.

In this subsection, we show that the energy shift, although a priori nonzero, does not alter the results which we obtained for the thermal conductivities, up to the order $\lambda^4$ in our perturbative expansion. To understand this, we decompose

$$O_{nk}[N_{n'k'}^{eq}] = O_{nk}^{(1)}[N_{n'k'}^{eq}] + O_{nk}^{(2)}[N_{n'k'}^{eq}] + O(\lambda^6) \quad (C25)$$

where, as for $D_{nk}$ elsewhere in this paper, the upper index $O(\rho)$ indicates a term of order $\lambda^{2\rho}$.

We have shown in Sec. C2c that $O_{nk}^{(1)}[N_{n'k'}^{BE}] = 0$. However, $O_{nk}^{(2)}[N_{n'k'}^{BE}] \neq 0$ a priori, so that an energy shift is actually required to cancel the equilibrium current. We thus consider the physical requirement, $O_{nk}[N_{n'k'}^{eq}] = 0$, to be an equation on the unknown $N_{n'k'}^{eq}$.

Now expanding $N_{n'k'}^{eq} = N_{n'k'}^{BE} + \delta N_{n'k'}^{eq}$ (with $\delta N_{n'k'}^{eq}$ at least of order $\lambda^2$), this equation becomes

$$0 = O_{nk}^{(1)}[N_{n'k'}^{BE}] + \sum_{n'k'} \delta N_{n'k'}^{eq} \partial_{n'k'} O_{nk}^{(1)}[N_{n'k'}^{BE}]$$

$$+ O_{nk}^{(2)}[N_{n'k'}^{BE}] + O(\lambda^6) \quad (C26)$$

At order $\lambda^2$, one recovers $O_{nk}^{(1)}[N_{n'k'}^{BE}] = 0$, as is required by detailed balance (see Sec. D2b).

At order $\lambda^4$, formally inverting this linear equation, one obtains

$$\delta N_{nk}^{eq} = -\sum_{n'k'} \left( \partial_{n'k'} O_{nk}^{(1)}[N_{n'k'}^{BE}] \right)^{-1} \bigg|_{n'k'} \times O_{nk}^{(2)}[N_{n'k'}^{BE}] + O(\lambda^4) . \quad (C27)$$

This correction Eq. \[C18\] to the phonon equilibrium populations is of order $\lambda^2$.

This ensures that using the approximate populations $N_{n'k'}^{eq} = N_{n'k'}^{BE}$ leads to a correct estimation of $D_{nk}^{(1)}$, the lowest-order contribution to $D_{nk}$, of order $\lambda^2$. However, the next-order contribution $D_{nk}^{(2)} \sim \lambda^4$ can only be estimated correctly if one adds to it the correction that $\delta N_{nk}^{eq}$ brings to $D_{nk}^{(1)}$. Similarly, using the approximate populations $N_{n'k'}^{eq} = N_{n'k'}^{BE}$ leads to a correct estimation of the lowest-order contribution, of order $\lambda^4$, to $M_{nk'n'k''}$ as expressed in the main text. Corrections of order $\lambda^6$, not considered in the present work, would require that the population corrections $\delta N_{nk}^{eq}$ be taken into account.

### 4. Computation at second Born order

As discussed at length, the first Born approximation alone does not lead to a nonzero thermal Hall effect. Here we compute that which appears when the Born expansion is taken up to the second Born order. More precisely, we consider all possible terms up to second Born order that lead to an off-diagonal scattering rate of order at most $\lambda^4$. This includes terms like $\langle f | H_{[1]}^{n} | n \rangle H_{[1]}^{n'k'} | 1 \rangle$ as well as $\langle f | H_{[1]}^{n} | n \rangle H_{[2]}^{n'k'} | 1 \rangle$, but not $\langle f | H_{[2]}^{n} | n \rangle H_{[2]}^{n'k'} | 1 \rangle$ since this term is already of order $\lambda^4$ (thus contributes to $|T_{1-r}|^2$ at order $\lambda^5$ at least).
a. Term with one-phonon interactions only

The first of these terms reads

\[
T_{1\to 11}^{[11]} = \sum_{n,k,n',k'} \sum_{q,q' = \pm} \sqrt{N_{nk}^i + \frac{1+q}{2}} \sqrt{N_{n'k'}^i + \frac{1-q'}{2}} \cdot \sum_m \langle f_s | Q_{n'k'}^q | m_s \rangle \langle m_s | Q_{n^qk}^{q'} | i_s \rangle E_{i_s} - E_{m_s} - q_\omega k_n + i\eta \langle i_p | q_{nk}^{q''} | f_p \rangle,
\]

(C28)

where the upper index indicates that within Born’s expansion, \( T^{[i,j]} \approx \frac{\langle f | H'_l | m \rangle \langle m | H'_r | f \rangle}{E_i - E_m + i\eta} \).

The squared T-matrix elements now include cross-terms between the first and second orders of Born’s expansion (although we keep only terms of order \( \lambda^2 \) at most). Here we give details of the calculation of one term, the square of Eq. (C28), \( |T_{1\to 11}^{[11]}|^2 \).

In the numerator, the matrix elements of the \( Q \) operators can combine themselves in two different ways, which we denote in the following as (a): \( \langle i_s | Q_{n^qk}^q | m_s \rangle \langle m_s | Q_{n^qk}^q | f_s \rangle \langle f_s | Q_{n^qk}^q | m_s \rangle \langle m_s | Q_{n^qk}^q | i_s \rangle \), and (b): \( \langle i_s | Q_{n^qk}^q | m_s \rangle \langle m_s | Q_{n^qk}^q | f_s \rangle \langle f_s | Q_{n^qk}^q | m_s \rangle \langle m_s | Q_{n^qk}^q | i_s \rangle \).

We use the following time integral representation of each of the denominators (using a regularized definition of the sign function),

\[
\frac{1}{x \pm i\eta} = PP \frac{1}{x} \mp i\pi \delta(x)
\]

(C29)

and introduce a third time integral to enforce the energy conservation \( E_f - E_i = \omega \nu k_n + q \omega \nu k + E_{f_s} - E_{i_s} \). The product of the denominators (cf. Eq. (C29)) leads to four terms, which can be labeled by two signs \( s, s' = \pm \), and we define, for convenience,

\[
\Theta_{ss'}(t_1, t_2) := [-\text{sign}(t_1)]^{1-s} [\text{sign}(t_2)]^{1-s'}.
\]

(C30)

Then, the transition rate coming from this part of the total squared matrix element can be written as a sum of eight terms:

\[
\Gamma_{i_p \to f_p}^{[11],[11]} = \sum_{n,k,n',k'} \sum_{q,q'} \left( N_{nk}^i + \frac{q+1}{2} \right) \left( N_{n'k'}^i + \frac{q'-1}{2} \right) \cdot \sum_{s,s' = \pm} \sum_{a,b} W_{nkq,n'k'q'}^{[11],[11],[i],(a),ss'} \langle i_p | q_{nk}^{q''} | f_p \rangle \cdot \frac{1}{2i} \int_{-\infty}^{+\infty} dt_1 e^{i t_1 x} \Theta_{ss'}(t_1, t_2) \left( N_{nk}^e + \frac{q+1}{2} \right) \left( N_{n'k'}^e + \frac{q'-1}{2} \right) \cdot \frac{1}{2i} \int_{-\infty}^{+\infty} dt_2 e^{i t_2 x} \Theta_{ss'}(t_1, t_2)
\]

(C31)

where we defined (notice the order of the two operators in the correlator and the sign \( t_1 \pm t_2 \) in the exponential):

\[
W_{nkq,n'k'q'}^{[11],[11],[i],(a),ss'} = \int dt_1 dt_2 \Theta_{ss'}(t_1, t_2) e^{i(\omega \nu k_n + q \omega \nu k + \omega \nu k')} e^{i(t_1 + t_2)} (q \omega \nu k_n - q \omega \nu k')
\]

\[
\cdot \langle Q_{n^qk}^q(-t - t_2) Q_{n^qk}^q(-t - t_2) Q_{n^qk}^q(-t - t_2) Q_{n^qk}^q(-t - t_2) \rangle \beta
\]

(C32)

\[
W_{nkq,n'k'q'}^{[11],[11],[i],(b),ss'} = \int dt_1 dt_2 \Theta_{ss'}(t_1, t_2) e^{i(\omega \nu k_n + q \omega \nu k + \omega \nu k')} e^{i(t_1 + t_2)} (q \omega \nu k_n - q \omega \nu k')
\]

\[
\cdot \langle Q_{n^qk}^q(-t - t_2) Q_{n^qk}^q(-t - t_2) Q_{n^qk}^q(-t - t_2) Q_{n^qk}^q(-t - t_2) \rangle \beta
\]

(C33)

We will investigate the symmetries of these terms in Sec. C4c and show that only some combinations contribute to the thermal Hall conductivity. In fact the eight terms from Eq. (C31) can be rewritten as products of (anti-)commutators. Meanwhile, defining the symmetrized in \( (nkq \leftrightarrow n'k'q') \) collision rate,

\[
W_{nkq,n'k'q'}^{[11],[11],[i],ss'} = \sum_{i=a,b} W_{nkq,n'k'q'}^{[11],[11],[i],(ss'),(nkq \leftrightarrow n'k'q')},
\]

we obtain components of the part of the collision matrix due to \( |T_{1\to 11}^{[11]}|^2 \):

\[
O_{n,k}^{[11],[11]} = \sum_{n',k'} \sum_{q,q'} \sum_{s,s'} W_{nkq,n'k'q'}^{[11],[11],[i],ss'} \left( N_{nk}^e + \frac{q+1}{2} \right) \left( N_{n'k'}^e + \frac{q'+1}{2} \right).
\]

(C34)

b. Commutators and anticommutators

Similarly to the above, writing \( [A, B]_{\pm} = AB \pm BA \), one can show that only \( [Q_{n^qk}^q(-t - t_2), Q_{n^qk}^q(-t + t_2)]_{ss'} \) contributes to \( W_{nkq,n'k'q'}^{[11],[11],[i],ss'} + W_{nkq,n'k'q'}^{[11],[11],[i],(ss')},(nkq \leftrightarrow n'k'q') \), and similarly only \( [Q_{n^qk}^q(-t_1), Q_{n^qk}^q(t_1)]_{ss'} \) contributes to
We show the following ("anti")detailed-balance relations reflecting the role of causality. From this, the same holds for the symmetrized in scattering rate:

\[ W^{[1,1],[1,1],ss'}_{n \kappa q,n' \kappa' q'} = \frac{n \leftrightarrow n' \kappa' q'}. \]  

Thus the only nonzero contributions to Eq. (C34) take the form

\[ W^{[1,1],[1,1],ss'}_{n \kappa q,n' \kappa' q'} = \int dt dt_2 \Theta_{ss'}(t_1, t_2) e^{i(q \omega_{nk} + q' \omega'_n \kappa' \kappa)} t e^{i(t_1 + t_2)(q \omega_{nk} - q' \omega'_n \kappa' \kappa)} \left\langle \left[ Q^\kappa_{n k}(-t - t_2), Q^\kappa'_{n' k'}(-t^2 + t_2) \right]_s \right\rangle \beta \]  

(38)

(39)

where \( s, s' = + \) corresponds to an energy conservation constraint, i.e. to on-shell scattering event, while \( s, s' = - \) corresponds to a PP\( (E_1 - E_2)^{-1} \) term, i.e. off-shell scattering (with \( i, n, f \) the initial, intermediate, and final states in the second-order process).

Note that, in a time-reversal symmetric system, these satisfy the symmetry

\[ W^{[1,1],[1,1],ss'}_{n \kappa q,n' \kappa' q'} = ss' W^{[1,1],[1,1],s's}_s. \]  

(C40)

reflecting the role of \( \pm i \eta \) in the denominators in terms of causality:

\[ c. \text{ Detailed balance} \]

Using the method of the previous subsection Sec. C2e, we show the following ("anti")detailed-balance relations

\[ W^{[1,1],[1,1],(a),ss'}_{n \kappa q,n' \kappa' q'} = ss' W^{[1,1],[1,1],(a),s's}_s e^{-\beta(q \omega_{nk} + q' \omega'_n \kappa' \kappa)}, \]  

(C41)

\[ W^{[1,1],[1,1],(b),ss'}_{n \kappa q,n' \kappa' q'} = ss' W^{[1,1],[1,1],(b),s's}_s e^{-\beta(q \omega_{nk} + q' \omega'_n \kappa' \kappa)}. \]  

(C42)

From this, the same holds for the symmetrized in \( n \kappa q \leftrightarrow n' \kappa' q' \) scattering rate:

\[ W^{[1,1],[1,1],ss'}_{n \kappa q,n' \kappa' q'} = ss' e^{-\beta(q \omega_{nk} + q' \omega'_n \kappa' \kappa)} W^{[1,1],[1,1],ss'}_{n \kappa q,n' \kappa' q'}. \]  

(C43)

One contribution comes from the "cross-term" \( 2 \Re \{ (T^{[2]}_1)_{1 \leftrightarrow 1} T^{[1,1]}_1 \} \), which contributes to the scattering rates in the form of

\[ \mathcal{M}^{[\Sigma,[1,1],[2],qq']}_{n \kappa q,n' \kappa'} = \frac{2N_{1c}}{h^4} \Im \int_{t, t_1} \left\langle Q_{n \kappa q,n' \kappa'}(-t) \{ Q^q_{n' k'}(-t_1), Q^{q'}_{n k}(t_1) \} \right\rangle \]  

(C47)

\[ \mathcal{M}^{[\Sigma,[1,1],[2],qq']}_{n \kappa q,n' \kappa'} = -\frac{2N_{1c}}{h^4} \Im \int_{t, t_1} \left\langle Q_{n \kappa q,n' \kappa'}(-t) \{ Q_{n' k'}(-t_1), Q^{q'}_{n k}(t_1) \} \right\rangle, \]  

(C48)

The last contribution comes from considering the second Born’s order matrix element

\[ T^{[2]}_1 = \frac{1}{\sqrt{N_{uc}}} \sum_{n \kappa q,n' \kappa'} \sum_{q,q'=\pm} \left( N^i_{n \kappa q} + 1 + q \right) \sqrt{N^i_{n \kappa q} + 1 + q} \left[ (f_{i_p} \text{Im} \{ Q^q_{n \kappa q} \}_{n \kappa q,n' \kappa'}| i_{s} \} + (f_{i_p} \text{Im} \{ Q^{q'}_{n' k'} \}_{n' k',n \kappa q}| i_{s} \} \right] \]  

(C49)
which contains two-phonon operators, of order $\lambda^2$. At order $\lambda^4$, it is thus involved in the “cross-term” $2\Re T_{1\rightarrow t}^1 T_{1\rightarrow t}^{2,1}$, which contributes to scattering rates in the form of $\mathcal{M}^{[2,1];[1],q_0}_{nk',k'} = \sum_{s = \pm} \mathcal{M}^{[2,1];[1],q_0}_{nk,k's}$, where

$$
\mathcal{M}^{[2,1];[1],q_0}_{nk,k's} = \frac{N_{uc}^{1/2}}{\hbar^4} \text{Im} \int_{t,t_1} \left\langle Q_{nk}(-t) | \text{sign}(t_1) \right|^{1/2} \left\langle Q_{nk'}(-t_1), Q_{nk'}^{q_0}(0) \right\rangle_{s'},
$$

and we recall the shorthand $[A,B]_{\pm} = AB \pm BA$.

The first two terms enforce the usual, “two-phonon”, (anti-)detailed balance relations

$$
\mathcal{M}^{[2,2];[1],q_0}_{nk',nk'} = e^{-\beta (q_0 \omega_k + q_0' \omega_{k'})} \mathcal{M}^{[2,2];[1],q_0}_{nk',nk'},
$$

with $\sigma = \oplus, \ominus$. Meanwhile, the last term satisfies “one-phonon” (anti-)detailed balance,

$$
\mathcal{M}^{[2,1];[1],q_0}_{nk,k's} = e^{-q_0 \omega_k} \mathcal{M}^{[2,1];[1],-q_0}_{nk,k's},
$$

where we used a different notation ($s = \pm$ as a lower index) to emphasize the difference with the other terms derived hereabove.

5. Computation at third Born’s order

The only third-order element of $T_{3\rightarrow t}$ which can appear in a term of order $\lambda^3$ in $|T_{3\rightarrow t}|^2$ is

$$
T_{3\rightarrow t}^{[1,1,1]} = \sum_{nk',nk'q'} \sqrt{N_{nk}^i + \frac{1+q}{2}} \left( N_{nk'}^i + \frac{1+q}{2} \right) \Re \left\langle i p | \frac{Q_{nk}}{f_p} \right| \sum_{m,s} \left\{ \frac{(f_s | Q_{nk}^{q_0} | m_s') (m_s' | Q_{nk}^{q_0} | m_s)}{(\bar{E}_{is} - E_{ms} - q_0 \omega_k + i\eta)(\bar{E}_{is} - E_{ms} - q_0' \omega_{k'} + i\eta)} + \frac{(f_s | Q_{nk}^{q_0} | m_s') (m_s' | Q_{nk}^{q_0} | m_s)}{(\bar{E}_{is} - E_{ms} - q_0 \omega_k + i\eta)(\bar{E}_{is} - E_{ms} - q_0' \omega_{k'} + i\eta)} \right\},
$$

which is involved in the scattering rate

$$
\mathcal{M}^{[1,1,1];[1],q_0}_{nk,k's} = 2\Re \left( T_{3\rightarrow t}^{[1,1,1]} | T_{3\rightarrow t}^{[1,1,1]} \right) \sum_{s,s' = \pm} \mathcal{M}^{[1,1,1];[1],q_0}_{nk,k's},
$$

where we denote

$$
\mathcal{M}^{[1,1,1];[1],q_0}_{nk,k's} = \frac{-1}{2\hbar^4} \Re \int_{t,t_1,t_2} \left\langle Q_{nk}^{q_0}(-t) | Q_{nk'}^{q_0}(-t_1), Q_{nk'}^{q_0}(0), Q_{nk'}^{q_0}(t_2) \right|_{s,s'},
$$

which are operators acting in their own Hilbert space, i.e. we write $H' = \sum_i H'_i$, with

$$
H'_i = \frac{1}{\sqrt{N_{uc}}} \sum_{\{n,k\}} \sum_{q_i = \pm} \left( \prod_{j=1}^m a_{q_i | n_j k_j} \right) Q_{\{n_j k_j\}}^{q_i},
$$

In this expression, $m$ is the number of phonon creation-annihilation operators coupled to $Q_{\{n_j k_j\}}^{q_i}$. In terms of the perturbative expansion introduced in the main text and the other appendices, this means $Q_{\{n_j k_j\}}^{q_i} \sim \lambda^i$; note that since the perturbative expansion is considered (formally) up to infinite order in this appendix, we make this specification only for the sake of clarity. To avoid ambiguities, we assume that all the $n_j k_j$ indices involved in a given term of $H'_i$ are distinct; this is correct in the thermodynamic limit. Note also that, for the sake of clarity in the following developments, the normalization factors

Appendix D: Generalizations

1. Generalized model and higher perturbative orders

To describe the interaction between phonon and another degree of freedom, we introduce general coupling terms between phonon annihilation (creation) operators $a_{nk}^{(i)}$ and general, for now unspecified, fields $Q_{\{q_i\}}^{\{n_j k_j\}}$.
of $N_{\text{ac}}$ are not defined following the same convention as in the rest of the paper.

In what follows, we take special notations for the first two indices: $n_1 k_1 \equiv n k, n_2 k_2 \equiv n' k'$. Using the model Eq. (64) and following the general procedure described in Sec. [C] and in the main text, one can then (at least formally) derive the collision integral which always takes the form

$$C_{nk} = \sum_{p=1}^{\infty} \frac{1}{N_{\text{ac}}} \sum_{\{n, k\}_1, \ldots, \{n, k\}_p} \sum_{q_1} q_1 \left( \prod_{l=1}^{p} \left( N_{n, k}_l + \frac{q_1 + 1}{2} \right) \right) W^{(p)},q_1 \{n, k\},$$

where the index $(p)$ denotes a term of order $\lambda^{2p}$. The scattering rate $W^{(p)},q_1 = W^{(p),q_1}_{\{n, k\}_1, \ldots, \{n, k\}_p}$ is the sum of all the scattering rates of the $[l_1, \ldots, l_m]; [l'_1, \ldots, l'_m]$ kind (according to the nomenclature introduced in the main text) such that $\sum_{m=1}^{l} l_i + \sum_{m=1}^{l'} l'_j = 2p$. In terms of physical process, each of these terms corresponds to the interference between two scattering channels, $[l_1, \ldots, l_m]$ and $[l'_1, \ldots, l'_m]$, such that in all, $2p$ phonon creations or annihilations occur between the initial and final states. Note that in the present paper, we compute explicitly this expansion up to $p = 2$.

We then expand the phonon average populations as $\overline{N}_{n, k} = N_{n, k}^{\text{eq}} + \delta N_{n, k}$. Following Eq (22), the diagonal scattering rate is obtained as $D_{nk} = -\partial_{N_{nk}} C_{nk} \bigg|_{N_{eq}}$. It can be decomposed as $D_{nk} = \sum_{p=1}^{\infty} D^{(p)}_{nk}$, where

$$D^{(p)}_{nk} = -\frac{1}{N_{\text{ac}}} \sum_{\{n, k\}_1, \ldots, \{n, k\}_p} \sum_{q_1} q_1 \left( \prod_{l=1}^{p} \left( N_{n, k}^{\text{eq}} + \frac{q_1 + 1}{2} \right) \right) W^{(p)},q_1 \{n, k\}.$$ 

Similarly, the off-diagonal scattering rate is obtained

$$M_{nk,n'k'} = \frac{\partial_{N_{nk}} C_{nk}}{N_{eq}}. \quad \text{It can be decomposed as} \quad M^{(p)}_{nk,n'k'} = \sum_{p=2}^{\infty} M^{(p)}_{nk,n'k'},$$

where

$$M^{(p)}_{nk,n'k'} = \frac{1}{N_{\text{ac}}} \sum_{\{n, k\}_1, \ldots, \{n, k\}_p} \sum_{q_1} q_1 \left( \prod_{l=1}^{p} \left( N_{n, k}^{\text{eq}} + \frac{q_1 + 1}{2} \right) \right) W^{(p)},q_1 \{n, k\}.$$

Like in the equations for $p = 1, 2$ derived explicitly in Appendix [C], $q_1$ always factorizes in the collision integral, as the change in number of $nk$ phonons due to the scattering event.

2. Special properties of first Born's order

a. Definitions and basic results

At first order of the Born expansion, all contributions to the collision integral are "semiclassical", in the sense defined in Sec. [III D 3] i.e. an operator $Q_{n_1 k_1 \ldots n_k k_i}$ does only appear in the collision integral as $[Q_{n_1 k_1 \ldots n_k k_i}]^2$.

To make this statement more precise, we rewrite

$$H'_{l} = \frac{1}{\sqrt{N_{\text{ac}}} \sum_{l=0}^{l-1} \sum_{j=1}^{l} \sum_{j=1}^{l} \left( \prod_{j=1}^{l} a_{n k_j}^+ \right) \left( \prod_{j=1}^{l} a_{n k_j}^- \right)} \times \frac{1}{\sqrt{l! (l-r)!}} Q_{n_1 k_1 \ldots n_r k_r \ldots n_{l-r+1} k_{l-r+1} \ldots n_k k_i}^r$$

where the upper indices of $Q$ are $r$ times ‘+$’ and $l-r$ times ‘$-$’, and $Q$ is by definition symmetric under permutation of its lower indices in the two blocks $\{n_k k_i\}_{1, \ldots, r}$ and $\{n_k k_i\}_{r+1, \ldots, l}$ separately. Hermiticity is guaranteed by $Q^{+\ldots -\ldots} \ldots \ldots$.

Note that at first Born's order, distinct scattering channels $l$ and $l'$ do not interfere for $l \neq l'$; one can thus study independently the contribution of each $H'_{l}$ to the collision integral.

The contribution to the squared T-matrix obtained from $H'_{l}$ at first Born's order is

$$|T_{l+1}^{l}|^2 = \frac{1}{N_{\text{ac}}} \sum_{r=0}^{l} \frac{1}{r! (l-r)!} \sum_{\{n, k\}^r} \left( \prod_{j=1}^{r} \left( N_{n_j k_j}^2 + 1 \right) \right) \left( \prod_{j=r+1}^{l} \left( N_{n_j k_j}^2 + 1 \right) \right) \sum_{j=1}^{l} \left( \prod_{j=1}^{l} \left( N_{n_j k_j}^2 + 1 \right) \right)$$

$$\times \mathbb{I}(f_{p}^{+} \{Q_{n_j k_j}^2 \}^r \{n_k k_i\}^r \{n_k k_i\}^r \{n_k k_i\}^r \{n_k k_i\}^r | i_{r+1}^{l+1}) \langle f_{p}^{+} | \langle Q_{n_j k_j}^2 \}^r \{n_k k_i\}^r \{n_k k_i\}^r \{n_k k_i\}^r \{n_k k_i\}^r | i_{r+1}^{l+1} \rangle.$$
where

\[ W^{[\ell;\ell]'}_{\{n_j, k_j\}}|_{\{n_j, k_j\}'_{r+1}} = \int \text{d}t e^{-i \left( \sum_{j=1}^{r'} \omega_{n_j, k_j} t - \sum_{j=r+1}^{l'} \omega_{n_j, k_j} t \right)} \left\langle Q^{+, ..., +}_{\{n_j, k_j\}}|_{r+1} \left( t \right) Q^{+, ..., +}_{\{n_j, k_j\}}|_{\{n_j, k_j\}'_{r+1}} \right\rangle. \]  

(D8)

Following the same steps as in Sec. C2c it is easy to see that __always_ enforces detailed-balance, namely

\[ W^{[\ell;\ell]'}_{\{n_j, k_j\}}|_{\{n_j, k_j\}'_{r+1}} = e^{-\beta \left( \sum_{j=1}^{r'} \omega_{n_j, k_j} - \sum_{j=r+1}^{l'} \omega_{n_j, k_j} \right)} \times W^{[\ell;\ell]'}_{\{n_j, k_j\}}|_{\{n_j, k_j\}'_{r+1}}. \]  

(D9)

We now prove two important properties of the collision integral, as obtained from first Born’s order, which derive therefrom.

\[ M^{1B}_{n_k, n'k'} = \sum_{l=1}^{\infty} \frac{1}{N^{1B}} \sum_{r=0}^{l} \sum_{j=1}^{l} \prod_{j=1}^{l} (N^{eq}_{n_j, k_j} + 1) \prod_{j=r+1}^{l} (N^{eq}_{n_j, k_j}) W^{[\ell;\ell]'}_{\{n_j, k_j\}}|_{\{n_j, k_j\}'_{r+1}} \times \frac{1}{r! (l-r)!} \left( r \delta_{n,n_1} \delta_{k,k_1} \left( (r-1) \delta_{n_2,n_2} \delta_{k_2,k_2} + (l-r) \delta_{n_1,n_1} \delta_{k_1,k_1} + (l-r-1) \delta_{n_1,n_2} \delta_{k_1,k_2} \right) \right) \]  

(D11)

After some algebra, following essentially the same steps as outlined hereabove, it is possible to show that

\[ M^{1B}_{nk,n'k'} e^{\beta \omega_{n'k'}} (N^{eq}_{n'k'})^2 = M^{1B}_{n'k', nk} e^{\beta \omega_{nk}} (N^{eq}_{nk})^2. \]  

(D12)

This, as was illustrated several times in the main text and the appendices, entails that __does not contribute to __ always see Eq. 1B6. We have thus shown that no contribution to the thermal Hall conductivity can possibly come from first Born’s order, regardless of the number of phonon operators in the Hamiltonian and of the nature of the operators Q to which they are coupled.

Appendix E: Application : further technical details

1. Solving the delta functions

In order to solve the two simultaneous delta functions, we use the following rewriting of \[M;\]
\[ 2m_{n,k,n',k'}^{\langle \varphi, q \rangle} = \frac{64n^2}{h^4N_{dc}^2} \sum_p \sum_{\ell, q_i} \varepsilon_{\ell, \ell', \ell''} [q_1, q_2, q_3, q_4] \Im \left\{ \mathcal{B}_{k, p + \frac{1}{2} \hat{q} k + \hat{q}' k'}^{n, \ell, \ell', \ell''} \mathcal{B}_{k', p + \frac{1}{2} \hat{q} k + \hat{q}' k'}^{n, \ell, \ell', \ell''} \right\} \]

\[ \delta \left\{ v_m q_1 \left[ q_1 q_2 \delta \left( n, q, k, q_4 \right) \right] + \hat{\Omega}_{\ell, p} + q_1 q_3 \hat{\Omega}_{\ell, p} + q_2 \right\} \delta \left\{ -2v_m q_1 \left[ \frac{q_1 q_4 \omega_{n' k'}}{v_m} + \hat{\Omega}_{\ell, p} - q_1 q_3 \hat{\Omega}_{\ell, p} + q_2 \right] \right\}, \]

where \( \hat{\Omega}_{\ell, p} = \Omega_{\ell, p}/v_m \) and

\[ \varepsilon_{\ell, \ell', \ell''} [q_1, q_2, q_3, q_4] = q_4 (2n_B(\Omega_{\ell_3, p} + q_1 k') + 1) (2n_B(\Omega_{\ell_2, p} + q_1 k') + 2 + 1) \]

is a product of thermal factors. Now collapsing the delta functions, we can write:

\[ 2m_{n,k,n',k'}^{\langle \varphi, q \rangle} = \frac{8\pi^2}{h^4N_{dc}^2} \sum_{\ell, q_i} \Im \left\{ \mathcal{B}_{k, p + \frac{1}{2} \hat{q} k + \hat{q}' k'}^{n, \ell, \ell', \ell''} \mathcal{B}_{k', p + \frac{1}{2} \hat{q} k + \hat{q}' k'}^{n, \ell, \ell', \ell''} \right\} \]

where, when they exist, the solutions, \( j = 0, \ldots, 3 \) take the form

\[ P_j = t_j [j/2] v_{[j/2]} + u_{[j/2]} w_{[j/2]}, \]

where, for \( i = 0, 1 \)

\[ v_i = a_2 k_1 + (-1)^i a_1 k_2, \]

\[ t_i = \frac{a_2 k_1^2 + (-1)^i a_1 k_2^2 - a_1 a_2 (a_1 + (-1)^i a_2)}{2v_i^2}, \]

\[ A_i = 4a_{i+1}^2 (v_i^2 - (k_1 \wedge k_2)^2), \]

\[ B_i = (-1)^i 4a_{i+1} (k_1 \wedge k_2) (a_{i+1}^2 - k_1^2) + 2(v_i \cdot k_{i+1}) t_i, \]

\[ C_i = - (a_{i+1} a_{i+1} - 2\delta_i - k_{i+1}^2) (a_{i+1} a_{i+1} + 2\delta_i - k_{i+1}^2) - 4(a_{i+1}^2 - k_{i+1}^2)(v_i \cdot k_{i+1}) t_i + 4(a_{i+1}^2 v_i^2 - (v_i \cdot k_{i+1})^2) t_i^2, \]

and \( \hat{J}(p_j) \) is given in the main text.

### 2. Choice of polarization vectors

Below, we enumerate possible explicit choices for a basis of polarization vectors \( (\varepsilon_{0,k}, \varepsilon_{1,k}, \varepsilon_{2,k}) \). In the numerical calculations, we use choice 2.

#### a. Choice 1

A simple choice is that of momentum-independent polarization vectors, which can be, for example: \( \varepsilon_{0,k} = \hat{x}, \varepsilon_{1,k} = \hat{y}, \varepsilon_{2,k} = \hat{z} \).

#### b. Choice 2

Below, we describe the choice of polarization vectors used in the numerical implementation. Its polarization vectors \( \varepsilon_{n,k} \) form an orthonormal basis in which \( k \) points along the \([1, 1, 1] \) axis, so that \( k \cdot \varepsilon_{n,k} = |k| \sqrt{3} \forall n \). This, as explained in the main text, ensures that the structure factor \( \mathcal{S}^{\ell,\ell'} \) does not vanish for \( \alpha = \beta \), corresponding to the largest coupling constants \( \Lambda_{1,5} \) (as opposed to anisotropic \( \Lambda_{6,7} \) for which \( \alpha \neq \beta \)).

The starting point is the orthonormal basis made of three vectors \( e_n, n = 0, 1, 2 \), defined as

\[ e_0 = \frac{1}{\sqrt{3}} (\sqrt{2} \hat{x} + \hat{z}) \]

\[ e_1 = \frac{1}{\sqrt{6}} (-\hat{x} + \sqrt{3} \hat{y} + \sqrt{2} \hat{z}) \]

\[ e_2 = \frac{1}{\sqrt{6}} (-\hat{x} - \sqrt{3} \hat{y} + \sqrt{2} \hat{z}) \]

in this basis, \( \hat{z} = [1, 1, 1] \). To rotate the \( \hat{z} \) axis into \( \hat{k} \)’s direction, we define

\[ \theta = \arccos(k_z/|k|), \]

\[ \phi = \arg(k_x + ik_y), \]
so that a good choice for the three polarization vectors is

\[
\varepsilon_{n,k} = i R_2(\phi) \cdot R_3(\theta) \cdot \text{diag}[s(k), 1, 1] \cdot e_n
\]  

(E11)

for \( n = 0, 1, 2 \). In the hereabove, we used the “sign” function

\[
s(k) = \begin{cases} +1 & \text{if } k \in \mathcal{D}_+ \\ -1 & \text{if } k \in \mathcal{D}_- \end{cases}
\]

(E12)

with respect to two domains \( \mathcal{D}_\pm \) defined by

\[
\mathcal{D}_+ = \{ k \mid k_z > 0 \text{ or } (k_y > 0 \text{ and } k_x > 0) \},
\]

(E13)

\[
\mathcal{D}_- = \{ k \mid k_z < 0 \text{ or } (k_y > 0 \text{ and } k_x < 0) \}
\]

(E14)

such that \( \mathbb{R}^3 = \mathcal{D}_+ \cup \mathcal{D}_- \cup \{ 0 \} \). The role of this \( s(k) \) function is to help ensure that this choice of polarizations enforces \( \varepsilon_n(-k) = \varepsilon_n(k)^* \), as well as all the tetragonal symmetry group of the crystal. This last statement means that under a symmetry operation \( g \) belonging to \( D_{4h} \) the symmetry group of the crystal, they transform as

\[
\varepsilon_n(g \cdot k) = \sum_{n'} c^g_{nn'}(k) g \cdot \varepsilon_{n'}(k)
\]

(E15)

where \( g \cdot \rho \) denotes the action of \( g \) on a vector, and most importantly the \( c^g_{nn'} \) coefficient either is \( \delta_{nn'} \) or exchanges the \( n = 1 \) and \( n = 2 \) polarizations, depending on whether \( k \) is in a high-symmetry position. Indeed \( n = 1, 2 \) are constructed degenerate (as eigenvectors of the dynamical matrix) at the high-symmetry planes and axes of the Brillouin zone. See [24] for details and further explanations on the behaviour of polarization vectors under symmetry operations.

\section{Choice 3}

One may also use the Hall-plane-dependent basis for the polarization vectors and label \( \varepsilon_{n,k} \), assuming \( \mu \nu \) is the Hall plane, \( \rho \) is the direction transverse to the plane, and \( \mu \nu \rho \) forms a direct orthonormal basis:

\[
\varepsilon_{0,k} = i k / |k| \quad \text{(longitudinal)},
\]

\[
\varepsilon_{1,k} = i \hat{u}_\rho \times \frac{k}{|\hat{u}_\rho \times k|} \quad \text{(transverse, in Hall plane)},
\]

\[
\varepsilon_{2,k} = \varepsilon_{0,k} \times \varepsilon_{1,k} = \frac{k^\rho k - k^2 \hat{u}_\rho}{|k| |\hat{u}_\rho \times k|} \quad \text{(transverse)}.
\]

(E16)

Then, if \( \alpha, \beta, \mu, \nu, \rho \in \{ x, y, z \} \), we can write:

\[
S^g_{\alpha \beta} = \frac{1}{\sqrt{\omega_{\alpha \beta}(k)}} k^\alpha k^\beta,
\]

(E17)

so that \( L_0 \propto k \cdot \lambda \cdot k \), \( L_1 \propto |k \times (\lambda \cdot k)|^2 \), \( L_2 \propto |k^2(\lambda \cdot k) - (\lambda \cdot k)k|^2 \).

For this choice of polarization vectors, the phonon-magnon coupling constants can be decomposed in such a way that their behavior under \( M_{\beta \mu}M_{\nu \rho} \) and \( M_{\nu \rho}, C_{\lambda \mu}^\alpha \) becomes transparent, in other words under the basis harmonics of the group generated by the latter operations. Note that, because the magnetic space group of the system is a priori independent of the symmetries associated with the choice of Hall “geometry,” the coefficients of the harmonics need not be independent. (In the the square lattice case discussed here, some of the symmetries of the system coincide with those of the Hall geometry, so that these coefficients are not entirely independent. Note that this causes additional constraints for the existence of a nonzero Hall effect.)

\section{Numerical implementation}

We define \( \hat{\lambda}^{\ell_1, \ell_2; \alpha \beta}_{\xi, \xi'} = \lambda^{\alpha \beta}_{\ell_1, \ell_2; \xi, \xi'} \), so that, in particular,

\[
\hat{\lambda}^{\ell_1, \ell_2; \alpha \beta}_{m, m} = \lambda^{\alpha \beta}_{m, m, \ell_1, \ell_2},
\]

\[
\hat{\lambda}^{\ell_1, \ell_2; \alpha \beta}_{n, n} = \lambda^{\alpha \beta}_{n, n, \ell_1, \ell_2},
\]

\[
\hat{\lambda}^{\ell_1, \ell_2; \alpha \beta}_{m, n} = \lambda^{\alpha \beta}_{m, n, \ell_1, \ell_2}.
\]

(E18)

(note the bars) and

\[
L^g_{\ell_1, \ell_2; \xi, \xi'} = \text{Tr} \left[ \hat{\lambda}^{\ell_1, \ell_2; \alpha \beta}_{\xi, \xi'} T \cdot S^g_{\alpha \beta} \right] = \sum_{\alpha, \beta = x, y, z} \hat{\lambda}^{\ell_1, \ell_2; \alpha \beta}_{\xi, \xi'} S^g_{\alpha \beta}.
\]

(E19)

Moreover, given \( (i) \) our choice of isotropic elasticity, \( (ii) \) a given Hall plane \( \mu \nu \) and perpendicular Hall axis, \( \rho \), \( (iii) \) \( \ell_1 = \ell_2 = \ell \), \( \hat{\lambda} \) is a function of \( \Lambda_{i, \ldots, 7}^{(\ell)} \) contains 72 values, which can be parametrized by a single index \( i = 0, \ldots, 71 \) through, e.g. \( i = 36 \xi + 18 \xi' + 9 \ell + 3 \alpha + \beta \) if we identify \((x, y, z)\) with \((0, 1, 2)\) for \( \alpha \) and \( \beta \), \( S \) is a complex function of \( n, \rho, k, q, \ell, \alpha, \beta \).
Appendix F: Application: further physical details

1. Microscopic derivation of the coupling constants

We consider the most general coupling between the strain tensor and bilinears of the $m, n$ fields, exhibiting all the symmetries allowed by the crystal symmetry group in the paramagnetic phase: the $D_{4h}$ tetragonal point group generated by mirror symmetries $S_x, S_y, S_z$, four-fold rotational symmetry $C_4^h$; translations of one unit cell — which forbids interactions of the $m, n$ type; and time-reversal. The corresponding Hamiltonian density reads:

$$H'_{\text{teta}} = \Lambda_1^{(m)} m_x m_x \epsilon^{xx} + \Lambda_2^{(m)} m_y m_y \epsilon^{yy} + \Lambda_3^{(m)} m_z m_z \epsilon^{zz} + \Lambda_1^{(n)} n_x n_x \epsilon^{xx} + \Lambda_2^{(n)} n_y n_y \epsilon^{yy} + \Lambda_3^{(n)} n_z n_z \epsilon^{zz} +$$

$$\left(\Lambda_4^{(m)} m_x m_x \epsilon^{xy} + \Lambda_4^{(n)} n_x n_x \epsilon^{xy} + \Lambda_5^{(m)} m_y m_y \epsilon^{yz} + \Lambda_5^{(n)} n_y n_y \epsilon^{yz}\right)$$

and

$$\Lambda_6^{(m)} m_x m_y \epsilon^{xy} + \Lambda_6^{(n)} n_x n_y \epsilon^{xy} + \Lambda_7^{(m)} m_y m_z \epsilon^{yz} + \Lambda_7^{(n)} n_y n_z \epsilon^{yz}$$

We now propose a microscopic origin to the $\Lambda_1^{(Z)}$ coefficients appearing in it.

We start from a generic spin exchange Hamiltonian of the form

$$H_{\text{ex}} = \sum_{\mathbf{R}, \mathbf{R}', a, b} S_a^{\mathbf{R}} S_b^{\mathbf{R}'} \sum_{\alpha, \beta} J_{\alpha \beta} \eta_{\alpha \beta} \epsilon_{\eta_{\alpha \beta}}$$

where $\mathbf{R}, \mathbf{R}'$ indicate the actual locations of the sites in the distorted lattice, and each sum spans the whole distorted lattice.

We then express $\mathbf{R} = \mathbf{r} + \mathbf{u}_r$, where $\mathbf{r}$ belongs to the undistorted lattice and $\mathbf{u}_r$ is the displacement field at site $\mathbf{r}$. Taylor-expanding the coefficients $J_{\alpha \beta} \epsilon_{\eta_{\alpha \beta}}$ with respect to the displacement field (and identifying $S_a^{\mathbf{R}} = S_a^{\mathbf{R}'}$), we thus obtain $H_{\text{ex}} = H_{\text{ex}}^0 + H_{\text{ex}}^1 + O(u^2)$, where $H_{\text{ex}}^0 = H_{\text{ex}} |_{\mathbf{R} = \mathbf{R}'}$ and

$$H_{\text{ex}}^1 = \sum_{\mathbf{r}, \mathbf{r}' a, b, r} S_a^{\mathbf{r}} \left( u_{r'} - u_{r'} \right) \partial_{r'} J_{\alpha \beta} \eta_{\alpha \beta} \epsilon_{\eta_{\alpha \beta}}$$

Then, identifying $\epsilon_{\alpha \beta} = \frac{1}{2} (\partial_{\alpha} u_{\beta} + \partial_{\beta} u_{\alpha})$ as the symmetric rank-2 elasticity tensor (i.e. strain tensor), we identify $H_{\text{ex}}^1 = H_{\text{ex}}^1 + \ldots$, where

$$H_{\text{ex}}^1 = \sum_{\mathbf{r}, \mathbf{r'}, a, b, r, \eta} S_a^{\mathbf{r}} S_b^{\mathbf{r'}} \left[ \eta_{\alpha} \partial_{\beta} \eta_{\beta} + \eta_{\beta} \partial_{\alpha} \eta_{\alpha} \right] J_{\alpha \beta} \eta_{\alpha \beta} \epsilon_{\eta_{\alpha \beta}} + \ldots$$

where $a, b = x, y, z$ is a spin axis index, $\alpha, \beta = x, y, z$ is a spatial index, and “+$\ldots$” encompasses terms featuring $\omega_{\alpha \beta}$ the anti-symmetric rank-2 elasticity tensor, as well as higher-order derivatives of the displacement field.

Finally, we take the particular case of a square lattice with tetragonal symmetry, and describe the spins in terms of $m, n$ fields as in the main text, namely $S_r = (1)^{y} \mu_0 (m(r)) + \alpha^2 (m(r))$. We identify $H_{\text{ex}}^1 = \sum_{\mathbf{r}, \mathbf{r'}, a, b, r} H'_{\text{teta}}(r) + \ldots$ where “+$\ldots$” is made of rapidly oscillating (time-reversal breaking) terms, and $H'_{\text{teta}}$ is as displayed in Eq. (F1), with identification

$$\Lambda_{ab}^{(m), \alpha \beta} = \frac{1}{2} \sum_{\eta} (\eta_{\alpha} \partial_{\beta} + \eta_{\beta} \partial_{\alpha}) J_{\alpha \beta} \eta_{\eta_{\alpha \beta}}$$

and

$$\Lambda_{ab}^{(n), \alpha \beta} = \frac{1}{2} \sum_{\eta} e^{i \pi \eta} (\eta_{\alpha} \partial_{\beta} + \eta_{\beta} \partial_{\alpha}) J_{\alpha \beta} \eta_{\eta_{\alpha \beta}}$$

where the sum over $\eta$ spans the whole direct (two-dimensional square) lattice, and $\pi = \left( \frac{\pi}{a}, \frac{\pi}{a} \right)$ with $a$ the square lattice parameter.

2. Contributions to intervalley couplings

In the main text, the magnon valleys are identified as:

$$\ell = 0 : \quad n_y, m_z$$

$$\ell = 1 : \quad n_z, m_y$$

Therefore, intervalley couplings are of the form $\Lambda_{\ell, \ell'}$ with $\delta_{\ell \ell'} \delta_{ab} = 1$. More explicitly, using Eq. (F9), they are:

$$\Lambda_{n_y, m_z} = \Lambda_{m_z, n_y} = \Lambda_{y_z, x_z}$$

$$\Lambda_{n_z, m_y} = \Lambda_{m_y, n_z} = \Lambda_{x_z, y_z}$$

Also recall from Eq. (F11) that

$$\Lambda_{\ell, \ell} = \Lambda_{\ell, \ell}$$

and all other values of $\alpha, \beta$ yield 0 for this set of lower indices. From this, it is clear that the $\Lambda_{\ell}^{(Z)}$ couplings always mix valleys, regardless of $m, n$, and contribute $\Lambda_{\ell, \ell'}$
term. This intervalley coupling is a small contribution which does not contribute to $T$ breaking. Meanwhile, the $T$-odd $\lambda_{n_{y}m_{y}}$ and $\lambda_{n_{z}m_{z}}$ intervalley couplings both contain contributions from both $A_{T}^{(E)}m_{0}^{y}$ and $A_{T}^{(E)}m_{0}^{y}$.

3. Derivation of the gaps from a sigma model

Here we provide a heuristic microscopic argument for expressing the gaps in terms of spin-spin couplings. We assume the addition of a term of the XXZ anisotropy form:

$$H_{XXZ} = gJ \sum_{(ij)} \left( 2S_{i}^{y}S_{j}^{z} - S_{i}^{z}S_{j}^{z} - S_{i}^{y}S_{j}^{y} \right).$$  \hspace{1cm} (F16)

This is to be added to the isotropic Heisenberg model, along with a Zeeman coupling to the transverse field.

Expansion around the $x$-ordered state, using that $m_{x} = -m_{y}n_{y} - m_{z}n_{z}$ and $n_{x} = 1 - n_{y}^{2} + n_{z}^{2}$, yields

$$H_{NLS}^{g,h} = \frac{1}{2\chi} \left( m_{y}^{2} + m_{z}^{2} \right) + 2gJ\alpha^{2} \left( 2m_{y}^{2} - m_{y}^{2} \right) - 2gJ\frac{\mu_{0}}{\alpha^{2}} \left( 2n_{z}^{2} - n_{y}^{2} \right) - h_{y}m_{y} - h_{z}m_{z}$$

$$+ \left( \frac{1}{2\chi} - 2gJ\alpha^{2} \right) \left( m_{y}^{2}n_{y}^{2} + m_{z}^{2}n_{z}^{2} + 2m_{y}n_{y}n_{z} \right) + 2gJ\frac{\mu_{0}}{\alpha^{2}} \left[ 1 - \frac{1}{2} \left( n_{y}^{2} + n_{z}^{2} + \frac{1}{n_{0}^{2}}(m_{y}^{2} + m_{z}^{2}) \right) \right]^{2}. \hspace{1cm} (F18)

Note that the first term on the second line is of the form $(\mathbf{m} \cdot \mathbf{n})^{2}$, where the $\perp$ indicates the components of the vectors normal to the ordering direction. Since we in the next step shift the magnetization by its value induced by the field, this is proportional to $(\mathbf{h} \cdot \mathbf{n})^{2}$, as is postulated in the main text on symmetry grounds.

We now show this explicitly. We shift the definition $m_{a} = m_{a} + \chi_{a}h_{a}$ for $a = y, z$, and expand the result to quadratic order in $m, n$. Here $\chi_{z} = (1/\chi + 4gJ\alpha^{2})^{-1}$ and $\chi_{y} = (1/\chi - 2gJ\alpha^{2})^{-1}$.

This gives

$$H_{NLS}^{g,h} = \frac{1}{2\chi} \left( m_{y}^{2} + m_{z}^{2} \right) + 2gJ\alpha^{2} \left( 2m_{y}^{2} - m_{y}^{2} \right) - 2gJ\frac{\mu_{0}}{\alpha^{2}} \left( 2n_{z}^{2} - n_{y}^{2} \right)$$

$$+ \left( \frac{1}{2\chi} - 2gJ\alpha^{2} \right) \left( \chi_{y}h_{y}^{2}n_{y}^{2} + \chi_{z}h_{z}^{2}n_{z}^{2} + 2\chi_{y}\chi_{z}h_{y}h_{z}n_{y}n_{z} \right) - 2gJ\frac{\mu_{0}}{\alpha^{2}} \left[ n_{y}^{2} + n_{z}^{2} + \cdots \right], \hspace{1cm} (F19)

where the ‘$\cdots$’ in the last brackets account for terms higher order in field, magnetization fluctuations, etc.

The anisotropy coefficients, denoted by $\Gamma_{ab}$ in the text, can now be extracted. The terms in $H_{NLS}^{g,h}$ which are quadratic in the $n_{y}, n_{z}$ fields read

$$H_{nn} = \chi_{y}h_{y}^{2} \left( \frac{1}{2\chi} - 2gJ\alpha^{2} \right) n_{y}^{2} + \left( \chi_{z}h_{z}^{2} \left( \frac{1}{2\chi} - 2gJ\alpha^{2} \right) - 6gJ\frac{\mu_{0}}{\alpha^{2}} \right) n_{z}^{2} + 2\chi_{y}\chi_{z}h_{y}h_{z} \left( \frac{1}{2\chi} - 2gJ\alpha^{2} \right) n_{y}n_{z}. \hspace{1cm} (F20)

Note that the two terms proportional to $n_{y}^{2}$ from the right-most contributions on each line of Eq. (F19) above canceled. That means the the coefficient of $n_{y}^{2}$ in Eq. (F20) vanishes if $h_{y} = 0$. This occurs because of Goldstone’s theorem and the assumed XXZ form of the anisotropy: if the field is purely along the $z$ direction,
XY symmetry of the Hamiltonian under rotations about the z axis is preserved, and this makes one of the spin wave modes remain gapless. Conversely, for a field along the y direction, and in the presence of anisotropy, both modes are generally gapped.

We can simplify the above expression if we assume $|g| \ll 1$, which means $\chi_y^{-1} \approx \chi_z^{-1} \approx \chi^{-1} = 4a^2 J$ and therefore $1/\chi \gg gJa^2$; hence

$$H_{nn} \approx \frac{\chi h_y^2}{2} n_y^2 + \left[ \frac{\chi h_z^2}{2} - 6gJ\mu_0^2 a^2 \right] n_z^2 + \chi h_y h_z n_y n_z. \quad (F21)$$

The above shows that if $h_z$ is small or zero, stability requires $g < 0$. This can be understood from the fact that, if the field is along $y$, then $H_{XXZ}$ is the only term, in the pure spin Hamiltonian, breaking explicitly the $O(2)$ symmetry in the $x-z$ plane. It should therefore favor antiferromagnetic alignment along the $x$ axis, which is the initial assumption of this derivation. It also proves the $\frac{1}{2}$ prefactor used in the main text.

The coefficients in Eq. (F21) give contributions to $\Gamma_{yy}$, $\Gamma_{zz}$, and $\Gamma_{yz}$, respectively. In this Appendix, as opposed to the more general expressions given in the main text, we assume they are the only contribution.

Since taking the magnetic field purely along one of the two axes $y,z$ guarantees that $\Gamma_{yz} = 0$, so that (as explained in the main text) the two magnon valleys are independent, let us assume that the field is along the $y$ axis. Then one gap is $\Delta_1 = |h_y|$, the Zeeman energy associated with the field along $y$. The other gap gets contributions both from the anisotropy and the Zeeman energy associated with the field along $z$.

Note that the anisotropy-induced gap involves the square root of the anisotropy, i.e. $\Delta_0 |_{h_z=0} = 4\sqrt{3}|g|J\mu_0$, which is not necessarily very small for reasonably small values of $g$.

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### 4. Supplementary figures

Here we present further calculations of scattering rates and (diagonal) thermal conductivity for the model of Sec. V as supplemental figures.
FIG. 5: Diagonal scattering rate $D_{nk}$ with respect to $\psi(k) \in [0, \pi/2]$ (horizontal axis) and $\phi(k) \in [0, 2\pi]$ (vertical axis) for fixed temperature $T = 0.5 T_0$, polarization $n = 0$, and momentum (1) $|k| = 0.0625/a$, (2) $|k| = 0.125/a$, (3) $|k| = 0.25/a$, (4) $|k| = 0.5/a$, (5) $|k| = 1.0/a$, (6) $|k| = 2.0/a$. Colormaps are different for the six subfigures. Note that the $C_4$ symmetry is approximately preserved for small $|k|$ but broken at large $|k|$, as stated in the main text. Also note how scattering processes at $\psi(k) < \psi_-$ become allowed for $\omega_{nk} \geq 2\Delta, 2\Delta'$, then dominant at large $|k|$.

FIG. 6: Diagonal scattering rate $D_{nk}$ with respect to $\psi(k) \in [0, \pi/2]$ (horizontal axis) and $\phi(k) \in [0, 2\pi]$ (vertical axis) for fixed temperature $T = 0.5 T_0$, momentum $|k| = 0.5/a$, and polarizations (1) $n = 0$, (2) $n = 1$, (3) $n = 2$. Colormaps are different between (1) and (2,3). Subfigure (1) is reproduced from the main text. Note that with our choice of polarization vectors $\varepsilon_{n,k}$, results for $n = 1$ and $n = 2$ are simply related by the mirror symmetry $\phi \mapsto \pi - \phi$. 
\(\phi(k) = 0; n = 1\)

\(\phi(k) = \pi/2; n = 0\)

\(\phi(k) = \pi/2; n = 1\)

FIG. 7: Diagonal scattering rate \(D_{nk}\) with respect to \(\psi(k) \in [0, \pi/2]\) (horizontal axis) and \(|k|a\) (vertical axis) for fixed temperature \(T = 0.5T_0\) and (1) \(\phi(k) = 0\) and \(n = 1\), (2) \(\phi(k) = \pi/2\) and \(n = 0\), (3) \(\phi(k) = \pi/2\) and \(n = 1\). Colormaps are different for the six subfigures. The \(\phi(k) = 0\) and \(n = 0\) case is displayed in the main text. Note that polarizations \(n = 1\) and \(n = 2\) yield the same results here. Note also that the general features are the same for polarizations \(n = 1, 2\) as for \(n = 0\): although the scattering rates of \(n = 1, 2\) polarizations for energies \(\omega_{nk} \gtrsim 2\Delta\) are not as clearly visible as they are for \(n = 0\), they are finite (of order \(10^{-4}\) in our units) and are only parametrically smaller than those for the \(n = 0\) polarization, due to purely geometrical factors (\(S_{nk}^{\alpha,\beta}\) in the main text).

FIG. 8: Longitudinal thermal conductivity \(\kappa_L\) with respect to temperature \(T\), in log-log scale, (left) for four different values of \(\gamma_{ext} = 1 \cdot 10^{-z}(v_{ph}/a)\), \(z \in [4, 7]\), from darker \((z = 4)\) to lighter \((z = 7)\) shade, (right) for four different values \(\gamma_{ext} = 1 \cdot 10^{-z}(v_{ph}/a)\), \(z \in [6, 9]\), from darker \((z = 6)\) to lighter \((z = 9)\) shade. Note that the two “bumps” come from the competition between \(\gamma_{ext}\) and \(D_{nn,\ell}\) for valley index \(\ell = 0, 1\), as explained in the main text.
\((q, q') = (-, -) \); \(\phi(k) = 0\)

\((q, q') = (-, +) \); \(\phi(k) = \pi/2\)

\((q, q') = (-, -) \); \(\phi(k) = \pi/2\)

\(\phi(k) = 0\) \((q, q') = (-, +)\) \(\phi(k) = \pi/2\) \((q, q') = (-, -)\) \(\phi(k) = \pi/2\)

\(m_0 = 0.05 \hat{z}\), temperature \(T = 0.5T_0\), momentum \(|k'| = 0.8/a\), \(k_z = 0.1/a\), and \((1)\) \(k_x = 0.2/a\), \(k_y = 0\), \((2,3)\) \(k_x = 0\), \(k_y = 0.2/a\), \(k_y = 0\) is in the main text. The colorbars are different for each figure and not linearly scaled. Note that thanks to anti-detailed-balance, angular dependences of \(\mathcal{W}_{nkn'/k'}^{\ominus,--}\), \(\mathcal{W}_{nkn'/k'}^{\ominus,+}\), \(\mathcal{W}_{nkn'/k'}^{\ominus,-+}\), \(\mathcal{W}_{nkn'/k'}^{\ominus,+}\) are identical to those of \(\mathcal{W}_{nkn'/k'}^{\ominus,--}\), \(\mathcal{W}_{nkn'/k'}^{\ominus,+}\), respectively, for an isotropic phonon dispersion.

FIG. 9: Skew-scattering rates (2) \(\mathcal{W}_{nkn'/k'}^{\ominus,--}\) and (1,3) \(\mathcal{W}_{nkn'/k'}^{\ominus,+}\), with respect to \(\psi(k') \in [0, \pi/2]\) (horizontal axis) and \(\phi(k') - \phi(k)\) (vertical axis), for fixed magnetization \(m_0 = 0.05 \hat{z}\), temperature \(T = 0.5T_0\), momentum \(|k'| = 0.8/a\), \(k_z = 0.1/a\), and \((1)\) \(k_x = 0.2/a\), \(k_y = 0\), \((2,3)\) \(k_x = 0\), \(k_y = 0.2/a\). The case \(\mathcal{W}_{nkn'/k'}^{\ominus,--}\), \(k_x = 0.2/a\), \(k_y = 0\) is in the main text. The colorbars are different for each figure and not linearly scaled. Note that thanks to anti-detailed-balance, angular dependences of \(\mathcal{W}_{nkn'/k'}^{\ominus,++}\), \(\mathcal{W}_{nkn'/k'}^{\ominus,++}\) are identical to those of \(\mathcal{W}_{nkn'/k'}^{\ominus,--}\), \(\mathcal{W}_{nkn'/k'}^{\ominus,--}\), respectively, for an isotropic phonon dispersion.