Enhanced Thermal Hall Effect in Nearly Ferroelectric Insulators

Jing-Yuan Chen,[1] Steven A. Kivelson,[2] and Xiao-Qi Sun[2,3]

[1]*Stanford Institute for Theoretical Physics, Stanford University, Stanford, California 94305, USA*
[2]*Department of Physics, Stanford University, Stanford, California 94305, USA*
[3]*Stanford Center for Topological Quantum Physics, Stanford University, Stanford, California 94305, USA*

(Received 13 November 2019; accepted 7 April 2020; published 21 April 2020)

In the context of recent experimental observations of an unexpectedly large thermal Hall conductivity, κ_H, in insulating La_2CuO_4 (LCO) and $SrTiO_3$ (STO), we theoretically explore conditions under which acoustic phonons can give rise to such a large κ_H. Both the intrinsic and extrinsic contributions to κ_H are large in proportion to the dielectric constant, ϵ, and the "flexoelectric" coupling, F. While the intrinsic contribution is still orders of magnitude smaller than the observed effect, an extrinsic contribution proportional to the phonon mean-free path appears likely to account for the observations, at least in STO. We predict a larger intrinsic κ_H in certain insulating perovskites.

DOI: 10.1103/PhysRevLett.124.167601

Introduction.—While it is well known that "neutral" excitations in solids, including phonons and other collective modes, induce some charge motion, it is intuitively clear that this is in some sense a "small" effect. In particular, this suggests that the coupling of such modes to magnetic fields is generically weak, and consequently that their contribution to the Hall component of the thermal conductivity tensor, κ_H, is relatively small. This argument is assumed implicitly when the ratio of κ_H to the Hall conductivity is used to test the Weidemann-Franz law in metals. It is also why the recent observation of a large κ_H in La_2CuO_4 (LCO) [1], an insulating cuprate, generated so much interest [2–5]. Moreover, an anomalous contribution to κ_H of smaller but still comparable magnitude has been identified in the doped material, $La_{2-x}Sr_xCuO_4$ (LSCO), for a range of $0 \leq x < 0.19$ comprising much of the "high temperature" superconducting range of doping. Still more recently, a comparably large κ_H has been found in the nearly ferroelectric insulator $SrTiO_3$ (STO) [6].

In this Letter we analyze the contribution of phonons to κ_H at temperatures small compared to the Debye temperature, so as to identify conditions under which it can be larger than expected on the basis of dimensional analysis. Naively, κ_H is small compared to the longitudinal response, κ_L, for two reasons: (i) κ_H is small in proportion to B/B_0 where B is the applied field, and $B_0 \equiv \phi_0/a^2 \sim 10^4$ T is the magnetic field corresponding to one quantum of flux ($\phi_0 = 2\pi\hbar c/e$) per unit cell cross-sectional area, a^2. (ii) κ_L is large in proportion to ℓ/a, the ratio of the phonon mean free path to the lattice constant. However, especially in the context of STO, we show that κ_H is enhanced by a factor proportional to the dielectric constant, ϵ, times the flexoelectric coupling F (defined below). Moreover, we find that there is an extrinsic contribution to κ_H that—in common with κ_L—is proportional to ℓ. A combination of these effects is the likely explanation of the large κ_H observed in STO; we speculate that they are responsible for the anomalous thermal Hall response in the cuprates, as well.

To be explicit, at low temperatures in an insulator, the dominant heat carriers are the acoustic phonons. We can express the thermal conductivity tensor in terms of the thermal diffusivity D as

$$\kappa^{ij} = C_v D^{ij} \tag{1}$$

where $i, j = x, y$, $C_v = (2\pi^2/5)(T/\hbar v_1)^3$ is the specific heat per unit volume, v_1 is an appropriate average of the sound speed over polarizations and directions of propagation, and we will use units in which $k_B = 1$. As is well known, the longitudinal piece of $\mathbf{D}$ is $D_L \equiv D^{xx} = D^{yy} = (1/3)v_2\ell$ where v_2 is a slightly different average of the sound speed [7]. In the following, we will focus on $D_H \equiv (1/2)(D^{xy} - D^{yx})$ [8]. Here we have assumed $\mathbf{B} = B\hat{\mathbf{z}}$ and assumed rotational symmetry at low energies for simplicity; our results will be qualitatively unchanged in lower symmetry situations.

Effective field theory.—The low energy dynamics in a nearly ferroelectric insulator is described by the continuum Lagrangian density $L = L_0 + L_{\rm int} + L_P$ in terms of the vector fields $\mathbf{u}$ and $\mathbf{P}$, which represent the local acoustic displacement and the dipole density respectively.

L_0 is the bare Lagrangian of the acoustic modes

$$L_0 = \frac{\rho}{2}\dot{\mathbf{u}}^2 - \frac{K_1}{2}(\nabla\mathbf{u})^2 - \frac{K_2}{2}(\nabla\cdot\mathbf{u})^2 + \cdots \tag{2}$$

where ρ is the mass density, and K_a the elastic moduli. Here and below "$\cdots$" refers to higher derivative terms.

Near ferroelectric quantum criticality [9], while there is no net dipole density $\langle\mathbf{P}\rangle$, it is essential to include a fluctuating dipole order parameter $\mathbf{P}$ which arises from a combination of all the infrared active phonon modes. To leading order

$$L_P = -\frac{\mathbf{P}^2}{2\chi\epsilon_0} + \cdots \tag{3}$$

where $\chi \equiv \epsilon/\epsilon_0 - 1$ is the static electric susceptibility. Important subleading dynamical terms are discussed later with Eq. (10).

Finally, the terms that couple the acoustic displacement to the dipole density take the form

$$L_{\rm int} = \frac{\mathbf{B}}{c}\cdot(\mathbf{P}\times\dot{\mathbf{u}}) + F_1\mathbf{P}\cdot\nabla^2\mathbf{u} + F_2\mathbf{P}\cdot\nabla(\nabla\cdot\mathbf{u}) + \cdots. \tag{4}$$

These terms are the lowest order terms (in powers of fields and derivatives) allowed by symmetry. The couplings F_a are known as flexoelectric couplings [10]. Their meaning is clear from the corresponding "adiabatic" equations of motion for $\mathbf{P}$, valid so long as the acoustic displacements are slowly varying compared to the energy scale of the lowest optical modes [see Eq. (10)]

$$\frac{\mathbf{P}}{\chi\epsilon_0} = \dot{\mathbf{u}}\times\frac{\mathbf{B}}{c} + F_1\nabla^2\mathbf{u} + F_2\nabla(\nabla\cdot\mathbf{u}) + \cdots. \tag{5}$$

The first term is the attractive force between the two poles of a moving dipole in magnetic field. The two flexoelectric terms represent the dipole density induced by a strain gradient.

Intrinsic thermal Hall conductivity.—While in the absence of scattering, κ_L diverges, there is a well-defined intrinsic thermal Hall conductivity [11]. Here, we integrate out $\mathbf{P}$ using Eq. (5) for an effective Lagrangian $L_{\rm eff} = L_0 + L_B$, which (to linear order in B) modifies the dynamics of the acoustic mode by

$$L_B = \chi\epsilon_0\frac{\mathbf{B}}{c}\cdot[F_1\nabla^2\mathbf{u} + F_2\nabla(\nabla\cdot\mathbf{u})]\times\dot{\mathbf{u}} + \cdots. \tag{6}$$

Although the induced L_B is "small," both due to the B dependence and the higher derivatives on $\mathbf{u}$, it is nevertheless the leading time-reversal odd term and therefore important. In particular, it introduces a time-reversal odd contribution to the Berry curvature $\mathbf{\Omega}_\alpha(\mathbf{k})$, where α labels the phonon mode and $\mathbf{k}$ its momentum. The resulting anomalous motion of the phonons generates an intrinsic thermal Hall conductivity [11]:

$$\kappa_H^{\rm in} = T\sum_\alpha\int\frac{d^3k}{(2\pi\hbar)^3}\Omega_\alpha^z(\mathbf{k})\int_{E_\alpha(\mathbf{k})/T}^\infty d\xi\xi^2\frac{\partial f_{\rm BE}(\xi)}{\partial\xi} \tag{7}$$

where $f_{\rm BE}(\xi)$ is the Bose-Einstein distribution at energy $E = \xi T$.

A phonon effective Lagrangian of the present form has been studied in Ref. [11]. The computation of the Berry curvature is reproduced in the Supplemental Material [12]. Its time-reversal odd components are found to be of order

$$\Omega(\mathbf{k}) \sim \frac{\chi\epsilon_0 FBvk}{Kc\hbar}\frac{\hbar}{k^2} \tag{8}$$

($k \equiv |\mathbf{k}|$, and $v \equiv \sqrt{K/\rho}$ is the sound velocity) where K and F are characteristic values of the couplings K_1, K_2 and F_1, F_2 respectively. The parameter dependence of Ω is physically intuitive: The first, dimensionless, factor is the typical ratio between L_B and L_0 (in the spirit of perturbation theory in B), where we have estimated $\partial/\partial t$ as $vk/\hbar$; the second factor is because Ω is defined with two k derivatives times $\hbar$. The same kind of reasoning yields the estimate

$$\frac{\kappa_H^{\rm in}}{\kappa_L} \sim \frac{\chi\epsilon_0 FBT}{Kc\hbar}\frac{v\hbar}{T\ell}, \tag{9}$$

where the second dimensionless factor is because κ_L, given below Eq. (1), scales with ℓ. This analysis also implies $\kappa_H^{\rm in} \propto T^3$ as κ_L does [11].

In a nearly ferroelectric material, there is a softened optical phonon branch $\mathbf{w}$ that contributes dominantly to $\mathbf{P}$ [9]. At temperatures comparable to or above the optical phonon gap, the dipole density $\mathbf{P} = \rho_e\mathbf{w} + \cdots$, where ρ_e is roughly the charge on the positive ions per unit cell, and "…" includes contributions from the flexoelectric effect Eq. (5) as well as possibly higher optical phonons. In this case, one must include the heat carried by $\mathbf{w}$. The effective Lagrangian should include the extra terms

$$-\frac{\rho'}{2\hbar^2}\Delta_{\rm TO}^2\mathbf{w}^2 + \frac{\rho'}{2}\dot{\mathbf{w}}^2 - \frac{K_1'}{2}(\nabla\mathbf{w})^2 - \frac{K_2'}{2}(\nabla\cdot\mathbf{w})^2 + \cdots + \frac{\mathbf{B}}{c}\cdot(\rho_e\mathbf{w}\times\dot{\mathbf{u}}) + \frac{\mathbf{B}}{c}\cdot(\rho_e'\mathbf{w}\times\dot{\mathbf{w}}) + \cdots \tag{10}$$

where $\Delta_{\rm TO}$ is the transverse optical phonon energy and the "…" includes the long-range Coulomb interaction [13,14] that causes an upward shift of the longitudinal optical phonon energy. Compared to the acoustic phonons, while the thermally excited $\mathbf{w}$ phonons are fewer in number due to the gap, their couplings to $\mathbf{B}$ are larger in the sense that they involve fewer derivatives than in L_B. Therefore, at finite temperatures, corrections to $\kappa_H^{\rm in}$ due to $\mathbf{w}$ should be considered. The details of both the $\mathbf{u}$ and the $\mathbf{w}$ contributions to $\kappa_H^{\rm in}$ are presented in the Supplemental Material [12].

While the intrinsic thermal Hall effect has been concretely studied, it is most often negligibly small in real materials. As we will see later, in STO, even after an enhancement by the low temperature electric susceptibility $\chi \approx 2\times10^4$ [15], $\kappa_H^{\rm in}$ is still 10^{-4} smaller than the observed value. We will however predict candidate materials in which this intrinsic effect might be sufficiently large for observation.

Extrinsic thermal Hall conductivity.—Skew scattering plays an important role in the Hall conductivity σ_H, most notably for its linear in ℓ contribution [16–19]. Now we show there is a parallel effect in the phonon thermal Hall conductivity κ_H, under appropriate, but unparallel, conditions.

There are two origins of time-reversal oddness during a scattering event. The first is directly associated with the defect off which the particle scatters, and second the Berry curvature effects on the kinetics of the particle itself. The former is the mechanism responsible for the ℓ linear contribution to the electronic σ_H [19]. Here for phonon κ_H, we focus on the latter, i.e., Berry curvature induced skew scattering, primarily because in STO there is no evidence of magnetic defects.

We assume dilute defects that scatter phonons strongly. To understand the importance of this assumption, first recall the opposite case of weak scattering. In this case, the disorder averaged $\overline{(\Delta H)^2}$ is a small parameter, where ΔH is the modification of Hamiltonian density by the disorder. As a consequence of Fermi's golden rule, the typical scattered angle at each event, $\Delta\hat{\mathbf{k}}$, and the inverse of the mean free path due to accumulated mild events, $1/\ell$, are both of smallness $\overline{(\Delta H)^2}$. Since Fermi's golden rule is manifestly time-reversal even, skew scattering only happens at higher orders in ΔH, which would be $\overline{(\Delta H)^4} \sim 1/\ell^2$ if the time-reversal oddness is solely due to the particle but not the defect. This is smaller than the nonskew scatterings by $1/\ell$; as a consequence, in electronic systems [19], the contribution of Berry curvature induced skew scattering to σ_H does not scale with ℓ. [20] On the other hand, for strong scattering, Fermi's golden rule does not apply. The typical scattered angle $\Delta\hat{\mathbf{k}}$ at an event is of order π, and the mean free path ℓ is about the actual spacing between the defects; neither scales with the indefinitely large ΔH. Thus, the Berry curvature induced skew scattering scales as $1/\ell$ along with nonskew scattering. For phonons specifically, a smoking gun for strong scattering is that ℓ approaches a finite constant as $T \to 0$. (This is "boundarylike" scattering [7,21–23], which we take to be strong and approximately independent of the phonon energy at $B = 0$.) This will be associated with STO phenomenology later.

With this physical picture in mind, we can write down a linearized Boltzmann equation describing phonon transport in the presence of a static temperature gradient, including an ansatz for the Berry curvature induced skew scattering with dimensionless strength A:

$$v\hat{\mathbf{k}}\cdot\nabla\delta T\frac{\partial f_{\mathrm{BE}}(\xi)}{\partial T} = -\frac{\delta f(\mathbf{k}) + \delta' f(\mathbf{k})}{\tau} + \int_{k'=k} d^2\hat{k}'\frac{A}{\hbar\tau}\mathbf{\Omega}(k)\cdot(\mathbf{k}\times\mathbf{k}')\cdot\delta f(\mathbf{k}') \times [1 + 2f_{\mathrm{BE}}(\xi)]. \tag{11}$$

Here we separated the distribution $f = f_{\mathrm{BE}} + \delta f + \delta' f$, where δf is of smallness $\nabla\delta T$, and $\delta' f$ of smallness $B\nabla\delta T$, and kept $\nabla\delta T$ and B each to linear order; $\tau = \ell/v$ is the relaxation time, and $\xi \equiv vk/T$. In writing the skew scattering ansatz, for simplicity we have assumed it is dominated by elastic single phonon scattering, whose full collision kernel is

$$\int d^3k'\delta(vk - vk')\{W_{\mathbf{k}'\to\mathbf{k}}f(\mathbf{k}')[1 + f(\mathbf{k})] - W_{\mathbf{k}\to\mathbf{k}'}f(\mathbf{k})[1 + f(\mathbf{k}')]\}. \tag{12}$$

Time reversal transformation requires $W_{\mathbf{k}'\to\mathbf{k}}(\mathbf{B}) = W_{-\mathbf{k}\to-\mathbf{k}'}(-\mathbf{B})$ in the scattering probability. The zeroth order in B nonskew scattering contributes to the relaxation time approximation (among other multiphonon processes), while the linear in B, Berry curvature induced, skew scattering is approximated by our ansatz in Eq. (11). Importantly, the dimensionless parameter A approaches a constant at small k, as we will justify later.

The Boltzmann equation Eq. (11) is easily solved by matching orders in B:

$$\delta f(\mathbf{k}) = \ell\hat{\mathbf{k}}\cdot\nabla\delta T\frac{\xi}{T}\frac{\partial f_{\mathrm{BE}}(\xi)}{\partial\xi},$$
$$\delta' f(\mathbf{k}) = \ell A\frac{\chi\epsilon_0 F v}{K\hbar}\frac{\mathbf{B}}{c}\cdot\frac{\mathbf{k}\times\nabla\delta T}{3}\cdot\frac{\xi}{T}\frac{\partial f_{\mathrm{BE}}(\xi)}{\partial\xi}[1 + 2f_{\mathrm{BE}}(\xi)], \tag{13}$$

where we have used Eq. (8) for $\mathbf{\Omega}$, with possible order 1 factors absorbed into A. We can then substitute these solutions into the heat current $\mathbf{J} = \int[d^3k/(2\pi\hbar)^3] \times v\hat{\mathbf{k}}vk[\delta f(\mathbf{k}) + \delta' f(\mathbf{k})]$ and match with $\mathbf{J} = \kappa_L(-\nabla\delta T) + \kappa_H(-\nabla\delta T)\times\hat{\mathbf{z}}$ to identify κ_L and κ_H, which are associated with δf and $\delta' f$, respectively. At this point, we shall restore the fact that there are three acoustic modes, hence a multiplicity of 3 for κ_L and a multiplicity of 9 for κ_H [the extra 3 comes from $\sum_{\alpha'}\delta f_{\alpha'}(\mathbf{k}')$ in the collision]. This leads to κ_L given below Eq. (1), and

$$\kappa_H^{\mathrm{ex}} \sim \ell A\left(\frac{T}{\hbar v}\right)^3 T\frac{\chi\epsilon_0 F B v}{K c\hbar} \times \frac{4\pi}{(2\pi)^3}\int_0^\infty d\xi\xi^5\frac{-\partial f_{\mathrm{BE}}(\xi)}{\partial\xi}[1 + 2f_{\mathrm{BE}}(\xi)]. \tag{14}$$

The integral can be readily performed, and the result is most conveniently presented as the ratio

$$\frac{\kappa_H^{\mathrm{ex}}}{\kappa_L} \sim 5A\frac{\chi\epsilon_0 F B T}{K c\hbar}. \tag{15}$$

This is our principal result for the extrinsic thermal Hall effect. Since $\kappa_L \propto T^3$, we predict $\kappa_H^{\mathrm{ex}} \propto T^4$. Compared to the intrinsic effect Eq. (9), we have an enhancement by $5AT\ell/v\hbar$. We will see this linear in ℓ enhancement (with a phenomenological parameter A of order 1), together with the large χ, reproduces the observed large κ_H in STO.

It remains to justify the claim that A approaches a constant at low energies, which is crucial for the T^4 scaling of κ_H^{ex}. Note that the dimensionless factor $A\mathbf{\Omega}\cdot(\mathbf{k}\times\mathbf{k}')/\hbar$ in our ansatz Eq. (11) is the time reversal odd modification to the scattering probability from $\hat{\mathbf{k}}'$ to $\hat{\mathbf{k}}$. It is controlled by the typical ratio between L_B and L_0, with the extra $\partial/\partial t$ in L_B estimated as $vk/\hbar$. Importantly, this $\partial/\partial t$ should not be estimated as $\Delta H/\hbar$ even though we are considering it during the course of a scattering event, because as we explained before, in strong scattering events no scattering angle would scale with the indefinitely large ΔH. Hence $A\mathbf{\Omega}\cdot(\mathbf{k}\times\mathbf{k}')/\hbar$ scales as k, and therefore, combined with Eq. (8), A approaches a constant, whose value is determined by but does not scale with ΔH.

Finally, there is another extrinsic contribution to Hall physics, that a particle's center of wave packet experiences a "side jump" by $\Delta\mathbf{r}\sim\mathbf{\Omega}\times\Delta\mathbf{k}$ during the course of scattering [19]. This extrinsic effect, unlike the skew scattering, does not scale with ℓ and hence is of the same order as κ_H^{in}. The reason is intuitive: while the shifted distance due to a modified (skew) scattering angle increases with propagation time, the shift that happened at the instant of scattering does not.

Discussion.—The behavior of κ_H in STO reported in Ref. [6] has several important features, in addition to the surprisingly large magnitude. First, κ_H is peaked at approximately the same temperature, $T_{\mathrm{peak}}\approx 20$ K, as κ_L—which certainly suggests [6] that they both reflect heat transport by the same set of excitations; in particular, above T_{peak}, acoustic phonons start to lose momentum by Umklapp scattering [7,22,23]. Moreover, the recently measured κ_L scales as T^3 at low temperatures $\lesssim 10$ K [24], and κ_H scales as T^4B in the same temperature range [6]. The T^3 scaling of κ_L implies the phonon mean free path ℓ is roughly T independent at low temperatures, and is extracted according to Eq. (1) to be ~ 1 μm [25]. The temperature independent ℓ has been interpreted [6,24] as the scattering of phonons off the twin boundaries between tetragonal domains in STO [26]. Importantly, the scattering must not be soft refraction and reflection, but rather some strong interaction process with the localized degrees of freedom on the twin boundaries (such as the localized dipoles [27]), in order to produce a temperature independent ℓ [7,21–23] comparable to the twin boundary spacing. While a detailed scattering mechanism has yet to be established, these known qualitative aspects all support the applicability of our theory of extrinsic thermal Hall effect.

To begin with, the observed $\kappa_H\sim T^4$ scaling is consistent with that predicted in Eq. (15). Our theory reproduces the observed magnitude of κ_H with the dimensionless parameter A of order 1. STO has a large $\chi\approx 2\times 10^4$ at low temperatures [15]. On the other hand, the flexocouplings take "normal" values $F\approx e/4\pi\epsilon_0 a$ [28] (where $a=3.9$ Å), according to experiments [29,30] and numerics [31]. The elastic moduli components take the usual values $K\approx 1$ eV/Å^3 [32]. Thus, at $B/c=10$ T $=1.5\times 10^{-4}$ $\hbar/e$Å^2 and $T=10$ K $=8.6\times 10^{-4}$ eV, our theory Eq. (15) yields $\kappa_H^{\mathrm{ex}}/\kappa_L\approx 5A\times 5\times 10^{-5}$. This matches the observed $\kappa_H/\kappa_L\sim -10^{-3}$ at this magnetic field and temperature, [33] if we set $A\approx 4$ [34]. (The sign is inconclusive because the componentwise values and signs of F remain unsettled [10].)

On the other hand, the intrinsic thermal Hall effect Eq. (9) gives a ratio $\kappa_H^{\mathrm{in}}/\kappa_L\approx 2\times 10^{-7}$ for $\ell\sim 1$ μm, much smaller than the observed value. We estimate that the contribution from the soft optical phonon $\mathbf{w}$ (with $\Delta_{\mathrm{TO}}\approx 24$ K [9]) at $T=10$ K is less than 0.1 of that from acoustic phonon, see Supplemental Material [12].

According to our theory, a change in the mean free path is not expected to dramatically change κ_H/κ_L. However, if a single crystal domain is formed (e.g., by cooling under strain [6]), although l becomes the system size, skew scattering ceases to happen during the transport, and hence κ_H should drop to the order of κ_H^{in}. This is consistent with the negligible κ_H in $KTaO_3$ (KTO) [6]. Note that ℓ in KTO is determined by the system size below 1 K and by soft impurity scattering at higher temperatures [35], and indeed neither case produces an ℓ linear κ_H^{ex} according to our theory.

While the intrinsic κ_H^{in} is negligible in STO even with the large $\chi\approx 2\times 10^4$, we predict κ_H^{in} to be observable in other systems, in particular a large class of perovskites [10], including $Ba_{1-x}Sr_xTiO_3$ (BSTO) and PMN-PT, which not only have similarly large χ but also large flexocouplings F hundreds of times of the "normal" value $e/4\pi\epsilon_0 a$. Furthermore, if some of these materials form structural domains at low temperatures, their κ_H^{ex} should be significantly larger than in STO.

Finally, we turn to the observations in LCO and LSCO. In Ref. [1], after subtracting off a quasiparticle contribution (inferred from σ_H using the Weidemann-Franz law), the remaining "anomalous" contribution is found to decrease smoothly with hole doping x, extending to values of x greater than the "optimal doping" that maximizes T_c; indeed, it is suggested it vanishes only for $x>p^\star$, an independently determined (material dependent) crossover concentration that is roughly $p^\star\approx 0.19$ in LSCO. In the range $0\le x<p^\star$, the system evolves from an antiferromagnetically ordered insulator through an insulating spin-glass phase and over much of the superconducting dome. The only low energy excitations that exist over this entire range of doping are the acoustic phonons.

While in LCO, which is far from ferroelectricity, the electric susceptibility $\chi\approx 30$ [36] is much smaller than in nearly ferroelectric materials such as STO, the flexocouplings F are unknown and could potentially be larger as in BSTO. Moreover, in LSCO, there is a $T=0$ structural transition (from orthorhombic to tetragonal) at $x\approx p^\star$, which might be significant in the context of a phonon skew scattering mechanism. The skew scattering might also originate from magnetic defects, the existence of which is plausible given that LCO is an antiferromagnet, and that spin-glass order persists up to $x \approx p^{\star}$ [37] (albeit at lower T and larger B than those in Ref. [1]). On the other hand, in LCO at the lowest T probed so far, κ_L (as well as κ_H) drops roughly linearly in T; this is not the behavior expected in the transport regime we have explored, proving that the present analysis is not directly applicable. Nonetheless, the fact that a phonon mechanism can produce a thermal Hall response of the requisite size in STO leads us to conjecture that the same physical considerations are at play in LCO as well.

We thank Kamran Behnia and Xiaokang Li for communications of their experimental results, Tao Qin and Junren Shi for discussions on phonon Berry curvature, John Tranquada, Louis Taillefer, and Jonathan Ruhman for helpful comments. J.-Y. C. is supported by the Gordon and Betty Moore Foundation's EPiQS Initiative through Grant GBMF4302. S. A. K. is supported in part by the U.S. Department of Energy (DOE) Office of Basic Energy Science, Division of Materials Science and Engineering at Stanford under Contract No. DE-AC02-76SF00515. X.-Q. S. is supported by the DOE Office of Science, Office of High Energy Physics, under the Contract No. DE-SC0019380.

Note added.—Recently, Ref. [38] reported a comparably large thermal Hall effect in LCO when the thermal current is oriented perpendicular to the Cu-O planes. As noted by the authors, this observation of isotropy establishes beyond reasonable doubt that the thermal Hall current in LCO is carried by acoustic phonons.

[1] G. Grissonnanche, A. Legros, S. Badoux, E. Lefrançois, V. Zatko, M. Lizaire, F. Laliberté, A. Gourgout, J.-S. Zhou, S. Pyon *et al.*, Nature (London) **571**, 376 (2019).
[2] R. Samajdar, S. Chatterjee, S. Sachdev, and M. S. Scheurer, Phys. Rev. B **99**, 165126 (2019).
[3] R. Samajdar, M. S. Scheurer, S. Chatterjee, H. Guo, C. Xu, and S. Sachdev, Nat. Phys. **15**, 1290 (2019).
[4] J. H. Han, J.-H. Park, and P. A. Lee, Phys. Rev. B **99**, 205157 (2019).
[5] Z.-X. Li and D.-H. Lee, arXiv:1905.04248.
[6] X. Li, B. Fauqué, Z. Zhu, and K. Behnia, Phys. Rev. Lett. **124**, 105901 (2020).
[7] H. Beck, P. Meier, and A. Thellung, Phys. Status Solidi (A) **24**, 11 (1974).
[8] Here κ_H is defined as the thermal Hall conductance per unit thickness in the $\hat{\mathbf{z}}$ direction. For κ_H, in contrast with the electronic σ_H, the macroscopically averaged value and the bulk local value are drastically different, as there is a substantial cancellation between the heat current carried in the bulk and near the edge [11,39,40].
[9] S. Rowley, L. Spalek, R. Smith, M. Dean, M. Itoh, J. Scott, G. Lonzarich, and S. Saxena, Nat. Phys. **10**, 367 (2014).
[10] P. Zubko, G. Catalan, and A. K. Tagantsev, Annu. Rev. Mater. Res. **43**, 387 (2013).
[11] T. Qin, J. Zhou, and J. Shi, Phys. Rev. B **86**, 104305 (2012).
[12] See Supplemental Material at http://link.aps.org/supplemental/10.1103/PhysRevLett.124.167601 for a comprehensive discussion for the phonons' Berry curvature and intrinsic thermal Hall effect.
[13] R. H. Lyddane, R. G. Sachs, and E. Teller, Phys. Rev. **59**, 673 (1941).
[14] J. Ruhman and P. A. Lee, Phys. Rev. B **94**, 224515 (2016).
[15] J. Dec, W. Kleemann, and B. Westwanski, J. Phys. Condens. Matter **11**, L379 (1999).
[16] J. Smit, Physica **21**, 877 (1955).
[17] J. Smit, Physica **24**, 39 (1958).
[18] A. Majumdar and L. Berger, Phys. Rev. B **7**, 4203 (1973).
[19] N. Nagaosa, J. Sinova, S. Onoda, A. H. MacDonald, and N. P. Ong, Rev. Mod. Phys. **82**, 1539 (2010).
[20] If $\overline{\Delta H} \neq 0$ in electron scattering (e.g., when disorders are mostly repulsive [41]), then the leading Berry curvature induced skew scattering occurs at $\overline{(\Delta H)^3} \sim 1/\ell^{3/2}$, hence its contribution to σ_H is $1/\ell^{1/2}$ smaller than σ_L.
[21] H. B. G. Casimir, Physica **5**, 495 (1938).
[22] R. A. Guyer and J. Krumhansl, Phys. Rev. **148**, 766 (1966).
[23] R. Guyer and J. Krumhansl, Phys. Rev. **148**, 778 (1966).
[24] V. Martelli, J. L. Jiménez, M. Continentino, E. Baggio-Saitovitch, and K. Behnia, Phys. Rev. Lett. **120**, 125901 (2018).
[25] Between 10 K and 20 K the phenomenon of Poiseuille flow [22,23] is observed [24], where κ_L increases faster than T^3 with T. This is because the onset of phonon-phonon interactions decreases the true diffusion velocity compared to the ballistic phonon velocity, and hence increases the relaxation time. Similar increase is observed in κ_H in the same temperature range [6], in support of our interpretation of κ_H as an extrinsic effect.
[26] A. Buckley, J.-P. Rivera, and E. K. Salje, J. Appl. Phys. **86**, 1653 (1999).
[27] J. F. Scott, E. K. H. Salje, and M. A. Carpenter, Phys. Rev. Lett. **109**, 187601 (2012).
[28] S. M. Kogan, Sov. Phys. Solid State **5**, 2069 (1964).
[29] P. Zubko, G. Catalan, P. R. L. Welche, A. Buckley, and J. F. Scott, Phys. Rev. Lett. **99**, 167601 (2007).
[30] P. Zubko, G. Catalan, P. R. L. Welche, A. Buckley, and J. F. Scott, Phys. Rev. Lett. **100**, 199906(E) (2008).
[31] J. Hong, G. Catalan, J. Scott, and E. Artacho, J. Phys. Condens. Matter **22**, 112201 (2010).
[32] J. Scott and H. Ledbetter, Z. Phys. B **104**, 635 (1997).
[33] The measured values of κ_H appear to vary by of order a factor of two for different samples, and even for the same sample if a thermal cycle is performed across the tetragonal transition temperature at 105 K. This is taken as evidence that κ_H is due extrinsically to twin boundaries [6], although why the dependence on thermal history is larger in κ_H than in κ_L remains unclear without a modeling of the scattering process.
[34] We have used the bulk value of $\chi\epsilon_0 F$. Near the twin boundaries where the scatterings occur, the local value is

likely different (probably enhanced) due to the localized dipoles and structures [27,29,42].

[35] B. Salce, J. Gravil, and L. Boatner, J. Phys. Condens. Matter **6**, 4077 (1994).

[36] C. Y. Chen, R. J. Birgeneau, M. A. Kastner, N. W. Preyer, and T. Thio, Phys. Rev. B **43**, 392 (1991).

[37] M. Frachet, I. Vinograd, R. Zhou, S. Benhabib, S. Wu, H. Mayaffre, S. Krämer, S. K. Ramakrishna, A. Reyes, J. Debray, T. Kurosawa, N. Momono, M. Oda, S. Komiya, S. Ono, M. Horio, J. Chang, C. Proust, D. LeBoeuf, and M.-H. Julien, arXiv:1909.10258.

[38] G. Grissonnanche, S. Thériault, A. Gourgout, M.-E. Boulanger, E. Lefrançois, A. Ataei, F. Laliberté, M. Dion, J.-S. Zhou, S. Pyon *et al.*, arXiv:2003.00111.

[39] N. R. Cooper, B. I. Halperin, and I. M. Ruzin, Phys. Rev. B **55**, 2344 (1997).

[40] T. Qin, Q. Niu, and J. Shi, Phys. Rev. Lett. **107**, 236601 (2011).

[41] H. Ishizuka and N. Nagaosa, Phys. Rev. B **96**, 165202 (2017).

[42] A. Morozovska, E. Eliseev, M. D. Glinchuk, L. Q. Chen, S. Kalinin, and V. Gopalan, Ferroelectrics **438**, 32 (2012).