Fundamental relations for anomalous thermoelectric transport coefficients in the nonlinear regime

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In a series of recent papers, anomalous Hall and Nernst effects have been theoretically discussed in the nonlinear regime and have seen some early success in experiments. In this paper, by utilizing the role of Berry curvature dipole, we derive the fundamental mathematical relations between the anomalous electric and thermoelectric transport coefficients in the nonlinear regime. The formulas we derive replace the celebrated Wiedemann-Franz law and Mott relation of anomalous thermoelectric transport coefficients defined in the linear response regime. In addition to fundamental and testable new formulas, an important by-product of this work is the prediction of nonlinear anomalous thermal Hall effect which can be observed in experiments.

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Introduction. Onsager's reciprocity relations mandate that the Hall effect in linear response has to vanish in a timereversal invariant system, whereas the nonlinear Hall effect has no such restriction [1]. The generalized Onsager's relation appropriate for nonlinear current response indicates that in order to get a nonzero DC nonlinear conductivity, the current response requires dissipation and should be proportional to the relaxation time τ [2]. Unlike the anomalous Hall effect in the linear-response regime [3-15], the nonlinear anomalous Hall effect (NLAHE) does not require broken time-reversal symmetry (TRS) but needs inversion symmetry (IS) breaking. The Berry curvature dipole (BCD), which is defined as the first-order moment of the Berry curvature over the occupied states, is found to be responsible for NLAHE [16-21]. The importance of electron-electron interactions for the external magnetic field dependence of the nonlinear conductivities has been pointed out [22,23]. Motivated by the idea of NLAHE, another second-order response function, nonlinear anomalous Nernst effect (NLANE), has been predicted in transitionmetal dichalcogenides (TMDCs) [24-26]. Interestingly, these nonlinear responses could manifest distinctive behaviors and have become promising tools for understanding novel materials with low crystalline symmetry in experiments. In this Rapid Communication, by utilizing the role of the Berry curvature dipole, we derive the fundamental mathematical formulas among the anomalous electric and thermoelectric transport coefficients in the nonlinear regime, replacing the celebrated Wiedemann-Franz law and the Mott relation [27] which are valid in the linear response regime.

In this Rapid Communication, we begin with the derivation of a new nonlinear response function, namely, the nonlinear anomalous thermal Hall effect (NLATHE), which can be directly observed in experiments. NLATHE refers to the appearance of a transverse thermal gradient as a second-order response to an applied longitudinal heat current (Fig. 1). Armed with these calculations, we then address the question of fundamental relations among the anomalous transport coefficients in the nonlinear regime. In linear response theory, the relations among electric, thermoelectric, and thermal transport coefficients of metals are encapsulated by the celebrated Wiedemann-Franz law and Mott formula [27]. These formulas in the context of linear anomalous transport coefficients have been studied in topologically trivial and nontrivial materials in theory as well as experiments [4,28-35]. While according to the Wiedemann-Franz law, the electric and thermal conductivities (regular or anomalous) are directly proportional to each other, the Mott formula predicts that the Nernst coefficient is proportional to the derivative of the Hall coefficient with respect to the chemical potential [see Eqs. (7) and (8)]. Interestingly, our analytical calculations for all three anomalous transport coefficients allow us to predict fundamentally new relations among the transport coefficients in the nonlinear regime. The principal result of this work is the remarkable prediction that in the nonlinear regime the anomalous Hall and Nernst coefficients are directly proportional to each other [Eq. (16)], while they are related through a derivative in the linear response regime [Mott relation, Eq. (8)]. Moreover, the derivative appears in the formula relating the electric and thermal conductivities in the nonlinear regime [Eq. (14)], while the Wiedemann-Franz law [Eq. (7)] in the linear response regime has no such derivative. The role of the derivative is thus interchanged in the nonlinear regime with respect to its linear response counterpart. These results should be tested in experiments as confirmation of the intrinsic nonlinearity, rather than a more conventional departure from the Wiedemann-Franz law and the Mott formula. We check the validity of our analytical results by full numerical

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FIG. 1. Schematic experimental setup for measuring the nonlinear anomalous thermal Hall effect. A transverse thermal gradient (ΔT) can be measured as a second-order response of the longitudinal heat current even in the absence of external magnetic field. The sample breaks inversion but respects time-reversal symmetry, so the linear response anomalous thermal Hall current is known to vanish by symmetry.

evaluation of the relevant quantities for MoS₂, a TR invariant but inversion-symmetry-broken TMDC that has been intensively studied in experiments recently.

Boltzmann theory and anomalous thermal Hall effect in nonlinear regime. The phenomenological Boltzmann transport equation can be written as

$$\{\partial_t + \dot{\boldsymbol{r}} \nabla_{\boldsymbol{r}} + \boldsymbol{k} \nabla_{\boldsymbol{k}}\} f(\boldsymbol{k}, \boldsymbol{r}, t) = I_{\text{coll}}\{f(\boldsymbol{k}, \boldsymbol{r}, t)\}, \qquad (1)$$

where the collision integral I_{coll} {f(k, r, t)} incorporates the effects of electron correlations (inelastic scattering) and elastic scattering from impurities. For the sake of simplicity, we here focus only on the impurity scattering. Invoking the relaxation time approximation, the steady-state solutions to the Boltzmann equation is given by

$$\{\dot{\boldsymbol{r}}\boldsymbol{\nabla}_{\boldsymbol{r}}+\dot{\boldsymbol{k}}\boldsymbol{\nabla}_{\boldsymbol{k}}\}f(\boldsymbol{k})=-\frac{g_{\boldsymbol{k}}}{\tau},$$
(2)

where $g_k = f(k) - f_0$ is the difference between the perturbed Fermi-Dirac distribution f_k and equilibrium Fermi-Dirac function f_0 . Considering the homogeneous uniform fields, we have dropped the r dependence of f(k, r, t). Here, τ is the average scattering time between two successive collisions. For simplicity, we ignore the momentum dependence of the scattering time τ and assume it to be a constant for this work.

To find the nonlinear anomalous thermal Hall coefficient in the absence of the external fields, we expand g_k as $g_k = g_k^1 + g_k^2 + \cdots$, where g_k^n is understood as the *n*th-order response to the applied thermal gradient, i.e., $g_k^n \propto (\nabla T)^n$. Substituting $f(k) = f_0 + g_k$ into the steady-state Boltzmann equation given in Eq. (2), we could find the distribution function at the first and second orders in the thermal gradient as

$$g_{k}^{1} = \tau \boldsymbol{v}_{k} \frac{(\varepsilon_{k} - \mu)}{T} \frac{\partial f_{0}}{\partial \varepsilon_{k}} \nabla T,$$

$$g_{k}^{2} = \tau \boldsymbol{v}_{k} \frac{(\varepsilon_{k} - \mu)}{T} \frac{\partial g_{k}^{1}}{\partial \varepsilon_{k}} \nabla T,$$
(3)

where μ is the chemical potential, $v_k = \hbar^{-1} \nabla_k \varepsilon_k$ is group velocity with ε_k the energy dispersion. In principle, expansions with higher orders (∇T) around the equilibrium distribution function can be derived by iteration. However, in this paper

we restrict the expansions only up to the quadratic order and neglect the small higher order ($\propto O(\tau^n)$, $n \ge 3$) terms.

After accounting for both the normal and anomalous contributions, the total thermal current j_{tot}^Q is given by $j_{tot}^Q = j_N^Q + j_E^Q + j_T^Q$, where j_N^Q is the standard contribution to thermal current coming from the conventional velocity v_k of the carriers and j_E^Q is the anomalous thermal current mediated by the Berry curvature Ω_k in the presence of electric field E [28]. In this paper, we are interested in the last term j_T^Q given by [36]

$$j_{T}^{Q} = -\frac{k_{B}^{2}T}{\hbar} \nabla T \int [d\mathbf{k}] \sum_{n} \mathbf{\Omega}_{\mathbf{k}}^{n} \bigg[\beta^{2} \big(\varepsilon_{\mathbf{k}}^{n} - \mu \big)^{2} f_{0} + \frac{\pi^{2}}{3} - \ln^{2} (1 - f_{0}) - 2 \operatorname{Li}_{2} (1 - f_{0}) \bigg], \qquad (4)$$

which describes the transverse thermal response to the applied thermal gradient $-\nabla T$ in the presence of a nontrivial Berry curvature Ω_k .

Substituting Eq. (3) into the thermal Hall term in Eq. (4) [with f_0 replaced by $f_k = f_0 + g_k$], the nonlinear anomalous thermal Hall current flowing along the direction *a* (second order of $-\nabla T$) can be written as

$$\left(\boldsymbol{j}_{T}^{Q}\right)_{a}^{\prime} = \epsilon_{abc} \frac{\tau \nabla_{b} T \nabla_{d} T}{\hbar^{2}} \int [d\boldsymbol{k}] \sum_{n} \Omega_{\boldsymbol{k},c}^{n} \frac{\left(\varepsilon_{\boldsymbol{k}}^{n} - \mu\right)^{3}}{T^{2}} \frac{\partial f_{0}}{\partial k_{d}}, \quad (5)$$

where the prime on $(j_T^Q)'$ indicates the nonlinear response and a, b, c, d represent the components x, y, z, and n is the band index. In this paper, we focus on this Berry curvaturedependent anomalous contribution to $(j_T^Q)'_a$ which is nonzero in TRS invariant systems.

From Eq. (5), the nonlinear anomalous thermal Hall coefficient can be written as $[(j_T^Q)'_a = \epsilon_{abc} l'_{cd} (\nabla_b T \nabla_d T)],$

$$l'_{cd} = \frac{\tau T}{\hbar^2} \int [dk] \sum_n \Omega^n_{k,c} \frac{\left(\varepsilon^n_k - \mu\right)^3}{T^3} \frac{\partial f_0}{\partial k_d}.$$
 (6)

This is one of the main results of this paper. We find that NLATHE, which is linearly proportional to the scattering time, appears due to the Berry curvature from the states near the Fermi surface. Under TR symmetry, we know $\Omega_k = -\Omega_{-k}$, $\varepsilon_k = \varepsilon_{-k}$, and $\partial f_0 / \partial k_d = -\partial f_0 / \partial (-k_d)$. Therefore, it is clear from the Eq. (6) that NLATHE can survive even in the time-reversal invariant systems.

Analog of Wiedemann-Franz law and Mott relation in the nonlinear regime. We now investigate the celebrated Wiedemann-Franz law and Mott relation in the nonlinear regime at low temperatures. In the linear response regime, the Wiedemann-Franz law, which gives the ratio between thermal conductivity (κ_{ab}) and electrical conductivity (σ_{ab}), is given by [37]

$$\frac{\kappa_{ab}}{\sigma_{ab}} = LT \tag{7}$$

with $L = \pi^2 k_B^2 / 3e^2$ being the Lorentz number. On the other hand, the Mott relation can be written as [27]

$$\alpha_{ab} = eLT \frac{\partial \sigma_{ab}}{\partial \mu},\tag{8}$$

where α_{ab} is the thermoelectric conductivity. To derive the analog of these formulas in the nonlinear regime, we first consider the nonlinear anomalous Hall effect. The BCD-induced NLAHE at a finite temperature can be written as [17]

$$\chi_{abc} = \epsilon_{abd} \frac{e^3 \tau}{2\hbar^2} D_{cd}, \qquad (9)$$

where D_{cd} , the Berry curvature dipole, is defined as

$$D_{cd} = \sum_{n} \int [d\mathbf{k}] \frac{\partial \mathbf{\Omega}_{\mathbf{k},c}}{\partial k_d} f_{\mathbf{k}} = -\sum_{n} \int [d\mathbf{k}] \mathbf{\Omega}_{\mathbf{k},c} \frac{\partial f_{\mathbf{k}}}{\partial k_d}.$$
 (10)

Using the Sommerfeld expansion [27], the BCD term (D_{cd}) of NLAHE at low temperature can be written as

$$D_{cd}(T,\mu) = G_{cd}(\mu) + \frac{\pi^2}{6} (k_B T)^2 G_{cd}^{(2)}(\mu) + O(T^4), \quad (11)$$

where

$$G_{cd}(\varepsilon) = \int [d\mathbf{k}] \delta(\varepsilon - \varepsilon_k) \mathbf{\Omega}_{\mathbf{k},c} \frac{\partial \varepsilon_k}{\partial k_d}$$
(12)

and $G_{cd}^{(n)}(\mu) = \partial^n G_{cd}(\mu)/\partial \mu^n$. Here, the first term $G_{cd}(\mu)$ is the zero-temperature BCD at Fermi energy μ whereas the second term shows a T^2 temperature dependence of the NLAHE, which agrees well with previous experimental results [18]. Similarly, the NLATHE at low temperature can be written as

$$l'_{cd}(T,\mu) = -\frac{7\tau\pi^4 k_B^4}{15\hbar^2} T^2 G_{cd}^{(1)}(\mu) + O(T^4)$$
(13)

with the higher order derivatives $G_{cd}^{(n)}(\mu)$ (odd number $n \ge 3$) included in $O(T^4)$.

Now, based on Eqs. (11) and (13), we can write the Wiedemann-Franz law in nonlinear regime as

$$l_{cd}' = -\frac{14}{15}eL_0^2 T^2 \frac{\partial \chi_0(\mu)}{\partial \mu},$$
 (14)

where $\chi_0(\mu) = e^3 \tau G_{cd}(\mu)/2\hbar^2$ denotes the zero-temperature NLAHE coefficient given by Eq. (9), and $L_0 = k_B^2 \pi^2/e^2$. Clearly, unlike the linear response regime, where the thermal Hall coefficient and charge Hall coefficient are directly proportional to each other [see Eq. (7)], the analog of the Wiedemann-Franz law in the nonlinear regime given by Eq. (14) shows that the anomalous thermal Hall coefficient is proportional to the *first-order derivative* of the anomalous Hall coefficient with respect to the chemical potential. Also, in contrast to the linear regime, the proportionality factor depends on T^2 , rather then T as in conventional Wiedemann-Franz law. The results in Eq. (14) should be taken as a result of the intrinsic nonlinearity, rather than a conventional departure from the Wiedemann-Franz law [38–44].

We could also derive the analog of the Mott formula in the nonlinear regime by first writing down the NLANE coefficient [25,26] as

$$\alpha_{cd}'(T,\mu) = \frac{e\tau}{\hbar^2} \left\{ \frac{\pi^2 k_B^2}{3} G_{cd}(\mu) + \frac{7\pi^4 k_B^4}{60} T^2 G_{cd}^{(2)}(\mu) + O(T^4) \right\}.$$
(15)

Based on Eqs. (11) and (15), the relation between the coefficients of NLANE and NLAHE can be written as

$$\alpha_{cd}'(\mu) = \frac{2}{3} L_0 \chi_0(\mu), \tag{16}$$

where we have considered only the first term for α'_{cd} [where the higher order terms are smaller by the successive higher order directive of the zero temperature BCD; see Eq. (11)]. Equation (16) is the Mott relation in the nonlinear regime which shows a finite value for the NLANE (α') even at zero temperature. In contrast to the linear regime, where the Nernst coefficient is proportional to the *derivative* of the Hall coefficient [see Eq. (8)], in the nonlinear regime, the corresponding anomalous coefficients are directly proportional to each other. Therefore, we find that, remarkably, the intrinsic nonlinearity introduces a derivative in the Wiedemann-Franz law while it removes the same from the Mott relation. These formulas can be directly tested in experiments in TR-invariant but inversion-broken systems where the anomalous coefficients are zero in the linear regime by symmetry.

Nonlinear transport coefficients for 2D massive Dirac fermions. We consider a model Hamiltonian of tilted 2D Dirac cones [45,46], which captures the low-energy properties of various Dirac materials, such as the surface of topological crystalline insulators and strained transition-metal dichalcogenides. The corresponding model Hamiltonian can be written as

$$H_s = s\alpha k_y \tau_0 + v_F \hbar(k_x \tau_y - sk_y \tau_x) + \Delta \tau_z.$$
(17)

Here, v_F is the Fermi velocity, Δ is the energy band gap opened at the $\pm \mathbf{K}$ valley, α is the tilting parameter, and $\tau_{x,y,z,0}$ represent Pauli matrices. The wave vector \mathbf{k} is measured from the valley center $\pm \mathbf{K}$ with index $s = \pm 1$ (which also indicates the opposite chirality of the Dirac fermions). Note that the Hamiltonian in Eq. (17) is TR invariant and the two massive Dirac cones $H_{s=\pm 1}$ are mapped to each other by the TR symmetry.

The low-energy dispersion and the corresponding Berry curvature of the Hamiltonian are given as

$$\mathcal{E}_{k}^{n,s} = s\alpha k_{y} + (-1)^{n-1} \sqrt{\Delta^{2} + (v_{F}\hbar)^{2} k^{2}},$$

$$\Omega_{k}^{n,s} = (-1)^{n-1} \frac{s(v_{F}\hbar)^{2} \Delta}{2(\Delta^{2} + (v_{F}\hbar)^{2} k^{2})^{3/2}}.$$
(18)

It is clear that $\Omega_k = -\Omega_{-k}$ is satisfied for Ω_k in Eq. (18). The tilting parameter α is required to produce a nonzero Berry curvature dipole contribution which can produce NLAHE, NLANE, and NLATHE. In what follows, we use parameters relevant to MoS₂, a TR-invariant TMDC, to compute the anomalous transport coefficients.

For a system tilted along the k_y axis, only the *x*-direction mirror symmetry (\mathcal{M}_x) that takes $k_x \to -k_x$ is preserved. As shown as in Fig. 2(a), the Berry curvature Ω_k is azimuthally symmetric in the k_x - k_y plane, whereas in Fig. 2(b) the modulated Berry curvature $\beta^3 (\mathcal{E}_k - \mu)^3 \Omega_k$ is only symmetric with respect to k_x . Because of the shift of the Fermi surface [black dashed line in Figs. 2(a) and 2(b) or the ring in Figs. 2(c) and 2(d)] along k_y , the net integrals of Ω_k in Fig. 2(a) and $\beta^3 (\mathcal{E}_k - \mu)^3 \Omega_k$ in Fig. 2(b) over the Fermi surface are nonzero. This explicitly renders the NLATHE a Fermi surface property. Figure 2 shows that the only nonzero component for NLATHE given in Eq. (6) is l'_{zy} where c = z, d = y represent $\Omega_{k,z}$, $\partial_y f_0$ respectively.



FIG. 2. (a) Berry curvature $\Omega_k^{n,s}$ and (b) modulated Berry curvature $\beta^3 (\mathcal{E}_k - \mu)^3 \Omega_k^{n,s}$ projected on the *k* space for Hamiltonian given in Eq. (17). The black dashed lines indicate the Fermi surface at $\mu =$ 1.5Δ . Panels (c) and (d) show the derivative of Fermi distribution function at Fermi energy $\mu = 1.5\Delta$ for $\partial_x f_k$ and $\partial_y f_k$ respectively. The parameters used here are n = 1, s = 1, t = 1.1 eV, a =3.19 Å, v = at, $\alpha = 0.1v$, $\Delta = 1.8$ eV, $k_{x,y} \in [-0.5\pi, 0.5\pi]$, $\beta =$ 1 (eV)⁻¹ is considered for panels (a) and (b) and temperature T =100 K is applied for panels (c) and (d).

It has been shown in Ref. [26] that the nonlinear anomalous Nernst coefficient has a dependence on the chemical potential similar to that of the nonlinear anomalous Hall coefficient studied in the experiments of Refs. [18,19]. This is consistent with the analog of the Mott formula valid in the nonlinear regime given in Eq. (16). To verify the relations between the coefficients of anomalous Hall and thermal Hall effects [namely the Wiedemann-Franz law in the nonlinear regime given by Eq. (14)], we compare the results for l'_{y} (index z for $\Omega_{k,z}$ is suppressed in a 2D system) based on Sommerfeld expansion in Eq. (13) with that from numerical calculations based on Eq. (6). As shown in Fig. 3, the analytical results (red dotted line) from Eqs. (13) and (14), coincide with the numerical results (the rest of the data besides the red dotted line) at low temperatures (T = 5-50 K). The numerical results and the prediction from the modified Wiedemann-Franz law differ from each other at higher temperatures (T = 100-300 K). To verify the quadratic temperature dependence in Eqs. (13) and (14), we plot the NLATHE coefficient l'_{v} as a function of $(k_BT)^2$ at different chemical potentials in Fig. 4. At low temperatures ($T \leq 50$ K), the numerical (circles) and analytical (black lines) results are consistent with each other, while they start deviating from each other around T = 60K with $\mu = 1.05\Delta$ (blue circles). The deviations in Figs. 3 and 4 are due to the omission of the higher orders terms in temperature $[O(T^4)]$ in Eq. (13). These contributions to l'_{ν} can be ignored in the regime of low temperatures. We have checked that our results for NLATHE are robust against all the monotonous modulation of the band gap Δ such as tuning effect by external



FIG. 3. Nonlinear anomalous thermal Hall coefficient $l'_y/(k_BT)^2$ vs chemical potential μ at different temperatures *T*. The red dotted line represents the analytical results based on the nonlinear Wiedemann-Franz law [Eqs. (13) and (14)]. The rest of the data points are results of numerical calculations based on Eq. (6). The inset is an enlargement of the plot around $\mu = \Delta$ (black dash line). For $\mu < \Delta$, the numerical results deviate from the modified Wiedemann-Franz law valid in the nonlinear regime because of the absence of higher orders temperature contributions [see Eq. (13)]. Here, the unit for the *y* axes is $\tau k_B^2/\hbar^2$; the other parameters are the same as in Fig. 2.

field [47], finite-temperature effects such as electron-phonon coupling [48], doping effect through the mixing of chalcogens in MoX₂ (X = S, Se, or Te) [49], etc., as well as strength of the tilting parameter due to uniaxial strain [50–53].



FIG. 4. Nonlinear thermal Hall coefficient l'_y plotted as a function of $(k_B T)^2$ for different values of the chemical potential. The circles are from numerical calculations based on Eq. (6), while the black lines corresponding to each chemical potential are the analytical results based on Eqs. (13) and (14). Here the units for y axes are $\tau k_B^2/\hbar^2$, the applied temperature $T \in [5K, 100K]$ with a unit step (1K), and all other parameters are the same as in Fig. 2.

Discussion. We derive the fundamental relations among the anomalous transport coefficients which replace the celebrated Wiedemann-Franz law and Mott relation in the nonlinear regime. Important by-products of these calculations include the prediction of nonlinear anomalous thermal Hall effect [Eq. (5)] and the persistence of the nonlinear anomalous Nernst coefficient in the zero temperature limit [Eq. (15)]. Our analytical results are confirmed by numerical calculations on MoS₂, a TR invariant TMDC that has been intensively studied in recent experiments. The nonlinear Wiedemann-Franz law and Mott relation derived in this work are valid for topologically nontrivial conductors with nonzero BCD.

Along with the BCD-induced nonlinear thermal Hall current $(j_T^Q)'_a$ given in Eq. (5), there exist other second-order contributions such as disorder-mediated contributions [20,54] (nonlinear side jump and skew-scattering contributions), scattering time-independent contributions [20,55] and Berry-curvature-independent contributions [17]. The BCD-induced contributions to the nonlinear response dfunctions

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discussed in this work are dominant in TR-invariant systems in which the Berry-curvature-independent contribution, which is nonzero only in the absence of both TRS and IS, discussed in Ref. [24] vanishes. Moreover, the Berry-curvature-induced contribution independent of scattering time which requires the breaking of TRS to be nonzero also vanishes in TR symmetric systems where the BCD-induced contributions are dominant. In addition, experimentally, the external, disorder-mediated, side-jump, and skew-scattering contributions to the nonlinear response functions can be separated from the BCD-induced contributions using a scaling formula as shown in Ref. [54]. The Wiedemann-Franz law and Mott relation derived in this paper thus apply only to the BCD-induced anomalous part of the nonlinear response functions which are nonzero in TR symmetric systems and leave out the contributions that take nonzero values only in systems with broken TRS.

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