Energy Magnetization and the Thermal Hall Effect

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We obtain a set of general formulas for determining magnetizations, including the usual electromagnetic magnetization as well as the gravitomagnetic energy magnetization. The magnetization corrections to the thermal transport coefficients are explicitly demonstrated. Our theory provides a systematic approach for properly evaluating the thermal transport coefficients of magnetic systems, eliminating the unphysical divergence from the direct application of the Kubo formula. For a noninteracting anomalous Hall system, the corrected thermal Hall conductivity obeys the Wiedemann-Franz law.

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The thermal Hall effect, or Righi-Leduc effect, is the thermal analogue of the Hall effect [1]. It gives rise to a transverse heat flow when a temperature gradient is applied. Recently there has been surging experimental interest in studying the thermal Hall effect in various systems, such as the phonon Hall effect [2,3], magnon Hall effect [4], and so on. There are also many theoretical efforts to study these phenomena [5–8]. However, most of these theoretical studies face a fundamental issue: direct application of the Kubo formula, when done correctly without questionable ''tricks''[9], always yields unphysical divergence at zero temperature [10,11]. Such an issue is actually a major obstacle in theoretical studies of the thermal Hall effect.

The underlying cause of this issue has previously been identified [12,13]: in a system breaking the time-reversal symmetry, either by applying an external magnetic field or due to the spontaneous magnetization, the temperature gradient not only drives the transport (heat) current but also drives the circulating (heat) current that is not observable in the transport experiment. Both contributions are present in the microscopic current density calculated by the standard linear response theory, and a proper subtraction of the nonobservable circulating component is necessary. For the electric transport, such subtraction involves the electromagnetic orbital magnetization density, while the subtraction of the energy current will involve the gravitomagnetic energy magnetization density [14], which characterizes the circulating energy flow. However, the previous theoretical discussions do not clarify what the transport current and the magnetizations are and how the magnetizations can be evaluated for a general extended system. The issue becomes more fundamental because the magnetizations are gauge-dependent quantities [15], and it is not *a priori* clear what the proper gauges of the magnetizations should be when calculating the transport coefficients.

In this Letter, we attempt to build the theory of thermal transport of magnetic systems on a firmer basis. We obtain a set of general formulas for determining the magnetizations, including the usual electromagnetic orbital magnetization as well as the gravitomagnetic energy magnetization [Eqs. (7)–(10)]. We further show that these magnetizations naturally emerge as corrections to the thermal transport coefficients, recovering the Onsager relations and Einstein relations [Eq. (20)], and eliminating the unphysical divergence. The result is a complete set of general formulas for calculating the transport thermal Hall conductivity, as well as the other thermal-electric responses such as Nernst effect and Ettingshausen effect [16]. The formulas also clarify what the gravitomagnetic energy magnetization is and how it can be calculated, and its thermodynamics is determined. We test our theory by calculating the thermal Hall coefficient of a noninteracting anomalous Hall system and observe the emergence of the Wiedemann-Franz law, consistent with the recent experimental observation [17].

Preliminaries.—To make our discussion specific, we consider a general electronic system. We should note that the formulas we will develop are general, applicable to the other systems such as the phonon and spin systems.

We assume that the total Hamiltonian of the unperturbed system can be written as $\hat{H} = \int d\boldsymbol{r}\hat{h}(\boldsymbol{r})$, where $\boldsymbol{r}$ denotes the spatial coordinate and $\hat{h}(\boldsymbol{r})$ is the local energy density operator. To study the electric and thermal responses, we introduce the external mechanic fields: the potential $\phi(\boldsymbol{r})$ and the gravitational field $\psi(\boldsymbol{r})$, where the gravitational field is introduced as the mechanic counterpart of the temperature gradient, following Luttinger [18]. In the presence of these fields, the local energy density operator of the system is modified to [13]

$$\hat{h}_{\phi,\psi}(\boldsymbol{r}) = [1 + \psi(\boldsymbol{r})][\hat{h}(\boldsymbol{r}) + \phi(\boldsymbol{r})\hat{n}(\boldsymbol{r})], \tag{1}$$

where $\hat{n}(\boldsymbol{r})$ is the local density operator, and the Hamiltonian of the system is $\hat{H}_{\phi,\psi} = \int d\boldsymbol{r}\hat{h}_{\phi,\psi}(\boldsymbol{r})$.

The particle and energy current operators of the system are defined by the conservation equations [13]:

$$\frac{\partial \hat{n}(\boldsymbol{r})}{\partial t} \equiv \frac{1}{i\hbar}[\hat{n}(\boldsymbol{r}), \hat{H}_{\phi,\psi}] = -\boldsymbol{\nabla}\cdot\hat{\boldsymbol{J}}_N^{\phi,\psi}(\boldsymbol{r}), \tag{2}$$

$$\frac{\partial \hat{h}_{\phi,\psi}(\boldsymbol{r})}{\partial t} \equiv \frac{1}{i\hbar}[\hat{h}_{\phi,\psi}(\boldsymbol{r}), \hat{H}_{\phi,\psi}] = -\boldsymbol{\nabla}\cdot\hat{\boldsymbol{J}}_E^{\phi,\psi}(\boldsymbol{r}), \tag{3}$$

where $\hat{\boldsymbol{J}}_N^{\phi,\psi}$ and $\hat{\boldsymbol{J}}_E^{\phi,\psi}$ are particle and energy current operators, respectively.

We further require that the current operators in the presence of the external fields can be related to the zero-field current operators $\hat{\boldsymbol{J}}_N$ and $\hat{\boldsymbol{J}}_E$ by [13]

$$\hat{\boldsymbol{J}}_N^{\phi,\psi}(\boldsymbol{r}) = [1 + \psi(\boldsymbol{r})]\hat{\boldsymbol{J}}_N(\boldsymbol{r}), \tag{4}$$

$$\hat{\boldsymbol{J}}_E^{\phi,\psi}(\boldsymbol{r}) = [1 + \psi(\boldsymbol{r})]^2[\hat{\boldsymbol{J}}_E(\boldsymbol{r}) + \phi(\boldsymbol{r})\hat{\boldsymbol{J}}_N(\boldsymbol{r})]. \tag{5}$$

We note that the current operator is only defined up to a curl by Eqs. (2) and (3). As we will show later [see Eq. (22)], one may use this freedom to find appropriate forms of current operators that do satisfy these scaling relations [19].

When the system is in equilibrium and in the absence of the external fields, we have $\boldsymbol{\nabla}\cdot\boldsymbol{J}_N^{\mathrm{eq}} = \boldsymbol{\nabla}\cdot\boldsymbol{J}_E^{\mathrm{eq}} = 0$, where $\boldsymbol{J}_{N(E)}^{\mathrm{eq}}(\boldsymbol{r}) = \langle\hat{\boldsymbol{J}}_{N(E)}(\boldsymbol{r})\rangle_0$ is the expectation value of the particle (energy) current for the equilibrium density matrix $\hat{\rho}_0 = (1/Z_0)\exp[-\hat{K}/k_BT_0]$, where $\hat{K} = \int d\boldsymbol{r}\hat{K}(\boldsymbol{r})$ and $\hat{K}(\boldsymbol{r}) \equiv \hat{h}(\boldsymbol{r}) - \mu_0\hat{n}(\boldsymbol{r})$. As a result, we can introduce the zero-field particle magnetization density $\boldsymbol{M}_N(\boldsymbol{r})$ and the energy magnetization density $\boldsymbol{M}_E(\boldsymbol{r})$ so that

$$\boldsymbol{J}_{N(E)}^{\mathrm{eq}}(\boldsymbol{r}) = \boldsymbol{\nabla}\times\boldsymbol{M}_{N(E)}(\boldsymbol{r}). \tag{6}$$

The equation can also be considered as the (incomplete) definition of the magnetizations. To make the so-defined magnetizations physically meaningful, one needs to further require that the magnetizations are the properties of material; i.e., they should be well-behaved functions of $\boldsymbol{r}$ and vanish outside of the sample. We also introduce the zero-field heat magnetization: $\boldsymbol{M}_Q(\boldsymbol{r}) \equiv \boldsymbol{M}_E(\boldsymbol{r}) - \mu_0\boldsymbol{M}_N(\boldsymbol{r})$.

Magnetizations.—We rigorously prove that, with the appropriate current operators that follow the scaling laws Eqs. (4) and (5), the total magnetizations can be calculated from the following set of equations:

$$-\frac{\partial \boldsymbol{M}_N}{\partial\mu_0} = \frac{\beta_0}{2i}\boldsymbol{\nabla}_{\boldsymbol{q}}\times\langle\hat{n}_{-\boldsymbol{q}};\hat{\boldsymbol{J}}_{N,\boldsymbol{q}}\rangle_0|_{\boldsymbol{q}\to 0}, \tag{7}$$

$$\boldsymbol{M}_N - T_0\frac{\partial \boldsymbol{M}_N}{\partial T_0} = \frac{\beta_0}{2i}\boldsymbol{\nabla}_{\boldsymbol{q}}\times\langle\hat{K}_{-\boldsymbol{q}};\hat{\boldsymbol{J}}_{N,\boldsymbol{q}}\rangle_0|_{\boldsymbol{q}\to 0}, \tag{8}$$

$$-\frac{\partial \boldsymbol{M}_Q}{\partial\mu_0} = \frac{\beta_0}{2i}\boldsymbol{\nabla}_{\boldsymbol{q}}\times\langle\hat{n}_{-\boldsymbol{q}};\hat{\boldsymbol{J}}_{Q,\boldsymbol{q}}\rangle_0|_{\boldsymbol{q}\to 0}, \tag{9}$$

$$2\boldsymbol{M}_Q - T_0\frac{\partial \boldsymbol{M}_Q}{\partial T_0} = \frac{\beta_0}{2i}\boldsymbol{\nabla}_{\boldsymbol{q}}\times\langle\hat{K}_{-\boldsymbol{q}};\hat{\boldsymbol{J}}_{Q,\boldsymbol{q}}\rangle_0|_{\boldsymbol{q}\to 0}, \tag{10}$$

where $\langle\hat{a};\hat{b}\rangle_0 \equiv (1/\beta_0)\int_0^{\beta_0} d\lambda\,\mathrm{Tr}[\hat{\rho}_0\hat{a}(-i\hbar\lambda)\hat{b}]$ is the Kubo canonical correlation function ($\beta_0 \equiv 1/k_BT_0$) [20], $\boldsymbol{M}_{N(Q)} \equiv \int d\boldsymbol{r}\boldsymbol{M}_{N(Q)}(\boldsymbol{r})$, $\hat{\boldsymbol{J}}_Q(\boldsymbol{r}) \equiv \hat{\boldsymbol{J}}_E(\boldsymbol{r}) - \mu_0\hat{\boldsymbol{J}}_N(\boldsymbol{r})$, and $\hat{n}_{\boldsymbol{q}}$, $\hat{K}_{\boldsymbol{q}}$, $\hat{\boldsymbol{J}}_{N,\boldsymbol{q}}$, $\hat{\boldsymbol{J}}_{Q,\boldsymbol{q}}$ are the Fourier transform of $\hat{n}(\boldsymbol{r})$, $\hat{K}(\boldsymbol{r})$, $\hat{\boldsymbol{J}}_N(\boldsymbol{r})$, $\hat{\boldsymbol{J}}_Q(\boldsymbol{r})$ [$\hat{a}_{\boldsymbol{q}} \equiv \int d\boldsymbol{r}\hat{a}(\boldsymbol{r})e^{-i\boldsymbol{q}\cdot\boldsymbol{r}}$], respectively.

Equations (7)–(10) are the central results of this Letter. The total magnetizations can be obtained by integrating over either the chemical potential μ_0 [Eqs. (7) and (9)] or the temperature T_0 [Eqs. (8) and (10)]. The corresponding boundary conditions are that at $\mu_0 \to -\infty$, $\boldsymbol{M}_{N(Q)} \to 0$ and at $T_0 \to 0$, $\boldsymbol{M}_N$ ($2\boldsymbol{M}_Q$) coincides with the right-hand side (rhs) of Eq. (8) [(10)], respectively. For an electronic system, the two approaches are equivalent. On the other hand, for systems without the chemical potential, such as the phonon and magnon systems, Eq. (10) is the only option for calculating the heat (energy) magnetization.

In Ref. [21], a similar formula for the electromagnetic orbital magnetization $\boldsymbol{M} \equiv -e\boldsymbol{M}_N$ was derived from its thermodynamic definition $\boldsymbol{M} = -(\partial\Omega/\partial\boldsymbol{B})_{T_0,\mu_0}$, where Ω is the grand thermodynamic potential and $\boldsymbol{B}$ is the magnetic field. It is easy to identify that the rhs of Eq. (8) is just $-(\partial K/\partial\boldsymbol{B})_{\mu_0,T_0}$, where $K \equiv \Omega + T_0S$ and S is the entropy of the system. Similarly, Eq. (7) is just the Maxwell relation between $\partial\boldsymbol{M}/\partial\mu$ and $\partial N/\partial\boldsymbol{B}$, where N is the total particle number of the system.

One can develop a similar thermodynamic interpretation for the heat magnetization as well. For this purpose, it is necessary to introduce a fictitious "magnetic field" $\boldsymbol{B}_s$ which couples to $\boldsymbol{M}_s \equiv \boldsymbol{M}_Q/T_0$ so that $\boldsymbol{M}_s = -(\partial\Omega/\partial\boldsymbol{B}_s)_{\mu_0,T_0}$. $\boldsymbol{B}_s$ can be related to the physical gravitomagnetic field $\boldsymbol{B}_g$ [22] by $\boldsymbol{B}_s \equiv -(T_0/c^2)\boldsymbol{B}_g$. In analogy to the particle magnetization, the right-hand sides of Eqs. (9) and (10) are $-T_0(\partial N/\partial\boldsymbol{B}_s)_{T_0,\mu_0}$ and $-T_0(\partial K/\partial\boldsymbol{B}_s)_{T_0,\mu_0}$, respectively, and these equations are just the thermodynamic relations. It is important to note that the particular way to introduce the thermodynamic quantities (e.g., $\boldsymbol{M}_s$ instead of $\boldsymbol{M}_Q$) is necessary for accounting for the extra factor of 2 in front of $\boldsymbol{M}_Q$ in Eq. (10).

We sketch the proof of Eqs. (7)–(10) in the following [23]. We introduce the static response functions:

$$\boldsymbol{\chi}_{ij}(\boldsymbol{r},\boldsymbol{r}') = \beta_0\langle\Delta\hat{n}_j(\boldsymbol{r}');\Delta\hat{\boldsymbol{J}}_i(\boldsymbol{r})\rangle_0, \qquad i,j = 1,2, \tag{11}$$

where $\hat{n}_1(\boldsymbol{r}) \equiv \hat{n}(\boldsymbol{r})$, $\hat{n}_2(\boldsymbol{r}) \equiv \hat{K}(\boldsymbol{r})$, $\hat{\boldsymbol{J}}_1(\boldsymbol{r}) \equiv \hat{\boldsymbol{J}}_N(\boldsymbol{r})$, $\hat{\boldsymbol{J}}_2(\boldsymbol{r}) \equiv \hat{\boldsymbol{J}}_Q(\boldsymbol{r})$, and $\Delta\hat{a} \equiv \hat{a} - \langle\hat{a}\rangle_0$. Applying Eqs. (2) and (3), we obtain $\boldsymbol{\nabla}\cdot\boldsymbol{\chi}_{ij}(\boldsymbol{r},\boldsymbol{r}') = (1/i\hbar)\langle[\hat{n}_j(\boldsymbol{r}'),\hat{n}_i(\boldsymbol{r})]\rangle_0$, which implies

$$\boldsymbol{\nabla}\cdot\boldsymbol{\chi}_{ij}^{\boldsymbol{q}}(\boldsymbol{r}) + i\boldsymbol{q}\cdot[\boldsymbol{\chi}_{ij}^{\boldsymbol{q}}(\boldsymbol{r}) - \boldsymbol{\nabla}\times\boldsymbol{M}_{ij}(\boldsymbol{r})] = 0, \tag{12}$$

where $\boldsymbol{\chi}_{ij}^{\boldsymbol{q}}(\boldsymbol{r}) \equiv \int d\boldsymbol{r}'\boldsymbol{\chi}_{ij}(\boldsymbol{r},\boldsymbol{r}')e^{-i\boldsymbol{q}\cdot(\boldsymbol{r}-\boldsymbol{r}')}$, $\boldsymbol{M}_{11}(\boldsymbol{r}) = 0$, $\boldsymbol{M}_{12}(\boldsymbol{r}) = \boldsymbol{M}_N(\boldsymbol{r})$, $\boldsymbol{M}_{21}(\boldsymbol{r}) = \boldsymbol{M}_N(\boldsymbol{r})$, $\boldsymbol{M}_{22}(\boldsymbol{r}) = 2\boldsymbol{M}_Q(\boldsymbol{r})$. In deriving Eq. (12), we have utilized Eqs. (2) and (3) and Eqs. (4) and (5), which imply the operator form of the commutators $[\hat{n}_j(\boldsymbol{r}'),\hat{n}_i(\boldsymbol{r})]$. Equation (6) is then used to

determine the equilibrium expectation values of the resulting commutators.

Therefore, $\boldsymbol{\chi}^{\boldsymbol{q}}_{ij}(\boldsymbol{r})$ must have the decomposition

$$\boldsymbol{\chi}^{\boldsymbol{q}}_{ij}(\boldsymbol{r}) = -i\boldsymbol{q}\times\boldsymbol{M}_{ij}(\boldsymbol{r}) + e^{-i\boldsymbol{q}\cdot\boldsymbol{r}}\boldsymbol{\nabla}\times\boldsymbol{\kappa}^{\boldsymbol{q}}_{ij}(\boldsymbol{r}). \tag{13}$$

Because both $\boldsymbol{M}_{ij}(\boldsymbol{r})$ and $\boldsymbol{\chi}^{\boldsymbol{q}}_{ij}(\boldsymbol{r})$ are properties of material, $\boldsymbol{\kappa}^{\boldsymbol{q}}_{ij}(\boldsymbol{r})$ must also be well behaved; i.e., it should be bounded and vanish outside of the sample. Moreover, the value of $\boldsymbol{\kappa}^{\boldsymbol{q}}_{ij}(\boldsymbol{r})$ at the long wave limit ($\boldsymbol{q}=0$) can be related to the macroscopic thermodynamic quantities $[\partial\boldsymbol{M}_{N(Q)}(\boldsymbol{r})/\partial\mu_0]_{T_0}$ and $[\partial\boldsymbol{M}_{N(Q)}(\boldsymbol{r})/\partial T_0]_{\mu_0}$ [23]. Applying $(i/2)\boldsymbol{\nabla}_{\boldsymbol{q}}\times$ to both sides of Eq. (13), taking limit $\boldsymbol{q}\to 0$, substituting $\boldsymbol{M}_{ij}$ and $\boldsymbol{\kappa}^{\boldsymbol{q}=0}_{ij}$, and integrating over $\boldsymbol{r}$, we obtain Eqs. (7)–(10).

Thermal transport coefficients.—We can show that the magnetizations determined by Eqs. (7)–(10) will emerge naturally as corrections to the thermal transport coefficients. To see this, we calculate the full response of the currents to small deviation from the global equilibrium. In this case, the system can be approximately described by the density matrix

$$\hat{\rho}\approx\hat{\rho}_{\mathrm{leq}}+\hat{\rho}_1, \tag{14}$$

where $\hat{\rho}_{\mathrm{leq}}$ is the local equilibrium density matrix characterized by the local chemical potential $\mu(\boldsymbol{r})$ and local temperature $T(\boldsymbol{r})$:

$$\hat{\rho}_{\mathrm{leq}} = \frac{1}{Z}\exp\Big[-\int d\boldsymbol{r}\frac{\hat{h}(\boldsymbol{r})-\mu(\boldsymbol{r})\hat{n}(\boldsymbol{r})}{k_BT(\boldsymbol{r})}\Big]. \tag{15}$$

$\hat{\rho}_1$ is the linear response correction to the local equilibrium density matrix, determined by the Liouville equation $i\hbar\partial\hat{\rho}/\partial t+[\hat{\rho},\hat{H}_{\phi,\psi}]=0$ [24]. We define $\alpha(\boldsymbol{r})\equiv[1+\psi(\boldsymbol{r})][\phi(\boldsymbol{r})+\mu(\boldsymbol{r})]$, $\beta(\boldsymbol{r})\equiv 1/k_B[1+\psi(\boldsymbol{r})]T(\boldsymbol{r})$. It is easy to see that when $\alpha(\boldsymbol{r})$ and $\beta(\boldsymbol{r})$ are spatially uniform, $\hat{\rho}_{\mathrm{leq}}$ becomes the exact global equilibrium density matrix corresponding to the Hamiltonian $\hat{H}_{\phi,\psi}$, and $\hat{\rho}_1=0$. Therefore, the conditions of the global equilibrium are $\boldsymbol{\nabla}\alpha(\boldsymbol{r})=0$ and $\boldsymbol{\nabla}\beta(\boldsymbol{r})=0$ [18].

We define $\boldsymbol{J}^{\phi,\psi}_1=\boldsymbol{J}^{\phi,\psi}_N$ and $\boldsymbol{J}^{\phi,\psi}_2=\hat{\boldsymbol{J}}^{\phi,\psi}_Q\equiv\boldsymbol{J}^{\phi,\psi}_E-\alpha(\boldsymbol{r})\boldsymbol{J}^{\phi,\psi}_N$. The forces conjugate to these currents are $\boldsymbol{X}_1=-\beta(\boldsymbol{r})\boldsymbol{\nabla}\alpha(\boldsymbol{r})$ and $\boldsymbol{X}_2=\boldsymbol{\nabla}\beta(\boldsymbol{r})$, respectively, so that the entropy generation is $\partial s/\partial t+\boldsymbol{\nabla}\cdot(\beta\boldsymbol{J}^{\phi,\psi}_Q)=\sum_i\boldsymbol{J}^{\phi,\psi}_i\cdot\boldsymbol{X}_i$ [1]. The expectation values of the currents have two parts of contributions:

$$\boldsymbol{J}^{\phi,\psi}_i=\boldsymbol{J}^{\mathrm{leq}}_i+\boldsymbol{J}^{\mathrm{Kubo}}_i, \tag{16}$$

where $\boldsymbol{J}^{\mathrm{Kubo}}_i\equiv\mathrm{Tr}\hat{\rho}_1\hat{\boldsymbol{J}}^{\phi,\psi}_i$ is just the usual linear response contribution with the response coefficients determinable by the Kubo formula [24]. Besides this, there is an extra contribution $\boldsymbol{J}^{\mathrm{leq}}_i=\mathrm{Tr}\hat{\rho}_{\mathrm{leq}}\hat{\boldsymbol{J}}^{\phi,\psi}_i$, which is due to the inhomogeneous local chemical potential and temperature field. We assume that the deviation from the homogeneity is small so that $\mu(\boldsymbol{r})\approx\mu_0+\delta\mu(\boldsymbol{r})$, $1/T(\boldsymbol{r})\approx(1/T_0)+\delta[1/T(\boldsymbol{r})]$. By applying the static response theory [20], we obtain, to the linear order of $x_1(\boldsymbol{r})\equiv\delta\mu(\boldsymbol{r})$ and $x_2(\boldsymbol{r})\equiv-T_0\delta[1/T(\boldsymbol{r})]$,

$$\boldsymbol{J}^{\mathrm{leq}}_i(\boldsymbol{r})\approx\boldsymbol{J}^{\mathrm{eq}}_i(\boldsymbol{r})+\sum_{j=1}^{2}\int d\boldsymbol{r}'\boldsymbol{\chi}_{ij}(\boldsymbol{r},\boldsymbol{r}')x_j(\boldsymbol{r}'), \tag{17}$$

where $\boldsymbol{\chi}_{ij}(\boldsymbol{r},\boldsymbol{r}')$ is the static response function defined in Eq. (11) and $\boldsymbol{J}^{\mathrm{eq}}_i(\boldsymbol{r})\equiv\langle\hat{\boldsymbol{J}}^{\phi,\psi}_i(\boldsymbol{r})\rangle_0$, which can be determined by Eqs. (6), (4), and (5). Substituting Eq. (13) into Eq. (17), and after some algebra, we obtain, to the linear order of ϕ, ψ, $\delta\mu$, and $\delta(1/T)$ [23],

$$\boldsymbol{J}^{\mathrm{leq}}_1(\boldsymbol{r})\approx\boldsymbol{\nabla}\times\boldsymbol{M}^{\phi,\psi}_N(\boldsymbol{r})-\frac{1}{\beta}\boldsymbol{M}_N(\boldsymbol{r})\times\boldsymbol{X}_2, \tag{18}$$

$$\boldsymbol{J}^{\mathrm{leq}}_2(\boldsymbol{r})\approx\boldsymbol{\nabla}\times\boldsymbol{M}^{\phi,\psi}_E(\boldsymbol{r})-\alpha(\boldsymbol{r})\boldsymbol{\nabla}\times\boldsymbol{M}^{\phi,\psi}_N(\boldsymbol{r}) -\frac{1}{\beta}\boldsymbol{M}_N(\boldsymbol{r})\times\boldsymbol{X}_1-\frac{2}{\beta}\boldsymbol{M}_Q(\boldsymbol{r})\times\boldsymbol{X}_2, \tag{19}$$

where $\boldsymbol{M}^{\phi,\psi}_N(\boldsymbol{r})\equiv[1+\psi(\boldsymbol{r})]\boldsymbol{M}_N(\boldsymbol{r})+\delta\boldsymbol{M}_N(\boldsymbol{r})$, $\boldsymbol{M}^{\phi,\psi}_E(\boldsymbol{r})\equiv[1+\psi(\boldsymbol{r})]^2[\boldsymbol{M}_E(\boldsymbol{r})+\phi(\boldsymbol{r})\boldsymbol{M}_N(\boldsymbol{r})]+\delta\boldsymbol{M}_E(\boldsymbol{r})$, and $\delta\boldsymbol{M}_{N(E)}(\boldsymbol{r})$ is the correction to the particle (energy) magnetization due to the spatial gradients of the chemical potential and temperature, determinable by $\boldsymbol{\kappa}^{\boldsymbol{q}}_{ij}(\boldsymbol{r})$.

Applying Eqs. (16), (18), and (19), we can obtain the total currents responding to the nonequilibrium forces. However, due to the presence of $\boldsymbol{J}^{\mathrm{leq}}_i$, such responses break the fundamental nonequilibrium thermodynamic relations [1]: (1) Onsager reciprocal relations, (2) Einstein relations; i.e., the currents should only be proportional to $\boldsymbol{\nabla}\alpha$ and $\boldsymbol{\nabla}\beta$ and vanish when the system is in the global equilibrium. The problem can be remedied by defining the transport currents as $\boldsymbol{J}^{\phi,\psi,\mathrm{tr}}_{N(E)}=\boldsymbol{J}^{\phi,\psi}_{N(E)}-\boldsymbol{\nabla}\times\boldsymbol{M}^{\phi,\psi}_{N(E)}$, and the corresponding transport responses then become

$$\begin{bmatrix}\boldsymbol{J}^{\mathrm{tr}}_1\\ \boldsymbol{J}^{\mathrm{tr}}_2\end{bmatrix}=\begin{bmatrix}\overleftrightarrow{L}^{(11)} & \overleftrightarrow{L}^{(12)}-\frac{\boldsymbol{M}_N}{\beta_0V}\times\\ \overleftrightarrow{L}^{(21)}-\frac{\boldsymbol{M}_N}{\beta_0V}\times & \overleftrightarrow{L}^{(22)}-\frac{2\boldsymbol{M}_Q}{\beta_0V}\times\end{bmatrix}\begin{bmatrix}\boldsymbol{X}_1\\ \boldsymbol{X}_2\end{bmatrix}, \tag{20}$$

where $\boldsymbol{J}^{\mathrm{tr}}_i\equiv(1/V)\int d\boldsymbol{r}\boldsymbol{J}^{\phi,\psi,\mathrm{tr}}_i(\boldsymbol{r})$ and V is the total volume of the system. $\overleftrightarrow{L}^{(ij)}$ is a tensor of rank two with the component $L^{(ij)}_{\alpha\gamma}=\int_0^\infty dte^{-st}\langle\hat{J}_{j,\gamma};\hat{J}_{i,\alpha}(t)\rangle_0$ ($\alpha,\gamma=x,y,z$), which is the usual response coefficient determined by the Kubo formula [24]. It is easy to verify that both the Onsager relations and the Einstein relations are recovered. The magnetizations determined in Eqs. (7)–(10) naturally emerge as the corrections to the thermal transport coefficients.

Application.—We can apply these general results to study the thermal Hall coefficient of a noninteracting anomalous Hall system [16,25] and show how the unphysical divergence is eliminated and the Wiedemann-Franz law emerges. The energy density of such a system, in the

presence of the external fields $\phi(\boldsymbol{r})$ and $\psi(\boldsymbol{r})$, can in general be written as

$$\hat{h}_{\phi,\psi}(\boldsymbol{r}) = [1+\psi(\boldsymbol{r})]\Big\{\frac{m}{2}[\hat{\boldsymbol{v}}\,\hat{\varphi}(\boldsymbol{r})]^\dagger\cdot[\hat{\boldsymbol{v}}\,\hat{\varphi}(\boldsymbol{r})] + \hat{\varphi}^\dagger(\boldsymbol{r})[V(\boldsymbol{r})+\phi(\boldsymbol{r})]\hat{\varphi}(\boldsymbol{r})\Big\}, \tag{21}$$

where $\hat{\varphi}(\boldsymbol{r})$ $[\hat{\varphi}^\dagger(\boldsymbol{r})]$ is the electron annihilation (creation) field operator with the two spin components, $\hat{\boldsymbol{v}} \equiv (1/m)[-i\hbar\boldsymbol{\nabla} + \boldsymbol{A}_{\rm so}(\boldsymbol{r})]$ is the velocity operator with $\boldsymbol{A}_{\rm so}(\boldsymbol{r})$ being the non-Abelian gauge potential characterizing the spin-orbit coupling, and $V(\boldsymbol{r})$ is the periodic potential. The field operator $\hat{\varphi}(\boldsymbol{r})$ satisfies the Schrödinger equation: $i\hbar\partial\hat{\varphi}/\partial t = \hat{\mathcal{H}}_{\phi,\psi}\hat{\varphi}$ with $\hat{\mathcal{H}}_{\phi,\psi} = (m/2)\hat{\boldsymbol{v}}\cdot[1+\psi(\boldsymbol{r})]\hat{\boldsymbol{v}} + [1+\psi(\boldsymbol{r})][V(\boldsymbol{r})+\phi(\boldsymbol{r})]$. An appropriate energy current operator that does satisfy both Eq. (3) and the scaling law Eq. (5) is

$$\hat{\boldsymbol{J}}_E^{\phi,\psi}(\boldsymbol{r}) = \frac{1+\psi}{2}[(\hat{\boldsymbol{v}}\,\hat{\varphi})^\dagger(\hat{\mathcal{H}}_{\phi,\psi}\hat{\varphi}) + (\hat{\mathcal{H}}_{\phi,\psi}\hat{\varphi})^\dagger(\hat{\boldsymbol{v}}\,\hat{\varphi})] + \frac{i\hbar}{8}\boldsymbol{\nabla}\times[(1+\psi)^2(\hat{\boldsymbol{v}}\,\hat{\varphi})^\dagger\times(\hat{\boldsymbol{v}}\,\hat{\varphi})]. \tag{22}$$

The presence of the last term is essential for satisfying the scaling law Eq. (5).

With the appropriate energy current operator at hand, we calculate the thermal Hall coefficient. The usual Kubo formula yields

$$\kappa_{xy}^{\rm Kubo} \equiv \frac{L_{xy}^{(22)}}{k_B T_0^2} = \frac{1}{2T_0\hbar V}\sum_{n\boldsymbol{k}}\Pi_{n\boldsymbol{k}}^z f_{n\boldsymbol{k}}, \tag{23}$$

where $\Pi_{n\boldsymbol{k}}^z = {\rm Im}\langle\frac{\partial u_{n\boldsymbol{k}}}{\partial k_x}|(\hat{\mathcal{H}}_{\boldsymbol{k}} + \epsilon_{n\boldsymbol{k}} - 2\mu_0)^2|\frac{\partial u_{n\boldsymbol{k}}}{\partial k_y}\rangle$, $u_{n\boldsymbol{k}}$ is the periodic part of the Bloch wave function for band n and quasimomentum $\boldsymbol{k}$, $f_{n\boldsymbol{k}} \equiv f(\epsilon_{n\boldsymbol{k}})$ is the Fermi distribution function, $\hat{\mathcal{H}}_{\boldsymbol{k}} = (1/2m)[-i\hbar\boldsymbol{\nabla} + \boldsymbol{A}_{\rm so}(\boldsymbol{r}) + \hbar\boldsymbol{k}]^2 + V(\boldsymbol{r})$, and $\epsilon_{n\boldsymbol{k}}$ is the electron dispersion [8]. It is easy to see that the coefficient diverges at zero temperature.

We calculate $\tilde{M}_Q^z \equiv (\beta_0/2i)\boldsymbol{\nabla}_{\boldsymbol{q}}\times\langle\hat{K}_{-\boldsymbol{q}};\hat{\boldsymbol{J}}_{Q,\boldsymbol{q}}\rangle_0|_{z,\boldsymbol{q}\to 0}$ and obtain

$$\tilde{M}_Q^z = -\frac{1}{4\hbar}\sum_{n\boldsymbol{k}}\{\Pi_{n\boldsymbol{k}}^z[2f_{n\boldsymbol{k}} + (\epsilon_{n\boldsymbol{k}}-\mu_0)f'_{n\boldsymbol{k}}] + 2\Omega_{n\boldsymbol{k}}^z(\epsilon_{n\boldsymbol{k}}-\mu_0)^3 f'_{n\boldsymbol{k}}\}, \tag{24}$$

where $\Omega_{n\boldsymbol{k}}^z \equiv -2{\rm Im}\langle\partial u_{n\boldsymbol{k}}/\partial k_x|\partial u_{n\boldsymbol{k}}/\partial k_y\rangle$. M_Q^z is obtained by integrating Eq. (10). After some algebra, we obtain $\kappa_{xy}^{\rm tr} \equiv \kappa_{xy}^{\rm Kubo} + (2M_Q^z/T_0V)$:

$$\kappa_{xy}^{\rm tr} = -\frac{1}{e^2T_0}\int d\epsilon(\epsilon-\mu_0)^2\sigma_{xy}(\epsilon)f'(\epsilon), \tag{25}$$

where $\sigma_{xy}(\epsilon) = -(e^2/\hbar)\sum_{\epsilon_{n\boldsymbol{k}}\le\epsilon}\Omega_{n\boldsymbol{k}}^z$ is the zero temperature anomalous Hall coefficient for a system with the chemical potential ϵ [16,25]. It recovers the Wiedemann-Franz law at low temperature $k_BT_0 \ll \mu_0$ [12,26], and the unphysical divergence is eliminated.

In summary, we have developed a systematic approach for calculating the particle and heat (energy) magnetizations. We also explicitly show that these magnetizations naturally emerge as the corrections to the thermal transport coefficients, recovering the Onsager and Einstein relations, and eliminating the unphysical divergences. Our approach makes no assumption on the nature of the system, so it is equally applicable to fermionic (e.g., electron) or bosonic (e.g., phonon, magnon) systems, either noninteracting or interacting. The approach does not involve the ill-defined spatially extended operators, so it is usable in practical calculations.

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