Onsager relations between spin currents and charge currents

Wayne M. Saslow *
Department of Physics, Texas A&M University, College Station, Texas 77843, USA

Chen Sun †
School of Physics and Electronics, Hunan University, Changsha 410082, China

(Received 3 November 2023; revised 9 August 2024; accepted 9 August 2024; published 23 August 2024)

We consider the macroscopic dynamics of systems with charge and spin currents, using the methods of Onsager's irreversible thermodynamics. Applied to systems with spin-orbit interaction (SOI), we derive Onsager relations showing that, if electrical disequilibrium leads to spin currents, magnetic disequilibrium leads to charge currents. We consider three examples of such SOI. Two of these predicted charge currents have not previously appeared. By measuring these charge currents one can infer the corresponding spin currents.

DOI: 10.1103/PhysRevB.110.054438

I. INTRODUCTION

Although spin flux (or, effectively, spin current) is essential to the field of spintronics, except for optical techniques that usually do not apply to metals, spin flux cannot be observed directly. Typically spin flux is inferred from a measurement like the inverse spin Hall effect, whereby for a surface (e.g., x-y) along which the spin polarization points (e.g., x), a spin flux leaving a material (here, along z) produces a spin-orbit interaction (SOI)-induced charge flux (and thus voltage difference) in the other surface plane direction (e.g., y). Generally, measurement of charge flux is much easier than that of spin flux.

The main result of this paper is that, for two theoretical models of systems with SOIs that predict spin fluxes driven by electrical disequilibrium, Onsager reciprocity predicts corresponding charge fluxes driven by magnetic disequilibrium. It would be of interest to measure such charge fluxes.

In 1971 the effects of SOI were studied in two pioneering works by Dyakonov and Perel, DP1 [1] and DP2 [2], in which (dimensionless) charge flux and (dimensionless) spin flux were considered for semiconductors. Later the charge flux measurement was called the spin Hall effect and the spin flux measurement was called the inverse spin Hall effect, and their theory implied the anomalous Hall effect [3]. DP2 [2], which was submitted a week after DP1 [1], gave additional spin flux terms due to previously neglected spin-orbit effects. These new terms were later expanded on and named "spin-swapping" by Lifshits and Dyakonov [4]. We refer to these works [1,2,4] and one by Dyakonov alone [5] collectively as the *DP model.*

DP considered only a nonequilibrium, flux-carrying spin density that they called the *accumulation of spin*. Later, in a context without SOI, Valet and Fert [6] introduced the term *spin accumulation* for this nonequilibrium spin density. Often spin density $\vec{S}$ and the proportional magnetization density $\vec{M}$ are used interchangeably.

For a ferromagnet with local equilibrium magnetization $\vec{M}$ along $\hat{z}$, versions of the charge flux and spin flux including spin-orbit effects were given by Taniguchi *et al.* (TGS) [7]. This included the anisotropic magnetoresistances (relative to the magnetization direction $\hat{M}$) associated with gradients of the electrochemical potential $\tilde{\mu}$ and gradients of only the longitudinal part $\hat{M}\cdot\vec{\mu}$ of the spin electrochemical potential $\vec{\mu}$ [8]. Here μ and $\hat{M}\cdot\vec{\mu}$ are the symmetric and antisymmetric combinations of the up-spin and down-spin electrochemical potentials. Reference [7] includes the anomalous Hall flux due to both of these gradients, but ignores the transverse part of the spin electrochemical potential $\vec{\mu}$. (In equilibrium $\vec{\mu}=\vec{0}$; see Sec. II.)

More recently, Amin *et al.* (ALSH) gave additional SOI-induced spin flux terms [9]; they are new and are not analogous to the spin-swapping terms of Dyakonov [2,4].

Because of the role of the SOI in the powerful spintronics probes of the spin Hall effect and the inverse spin Hall effect, SOI-related spin currents in ferromagnets have been the subject of ongoing theoretical and experimental interest. The purpose of the present work is to apply Onsager reciprocity to the three theoretical models with SOI mentioned above, namely, the DP2 [2], TGS [7], and ALSH [9] models. This reciprocity holds for any model where the spin current arises primarily from a single energy band, in which case the spin chemical potential is associated with that band.

We consider weakly spin-polarized systems (paramagnets) in Sec. IV and strongly spin-polarized systems (ferromagnets) in Sec. V. We find that TGS [7] are consistent with that reciprocity, but that for consistency the other two theories [2,9] must be given new charge fluxes that correspond to the spin flux already in the models. For the new charge current associated with DP2 [2], see Eq. (29); for the new charge current associated with ALSH [9], see Eq. (33). In Sec. VI we estimate the size of these effects and propose experiments to

*Contact author: wsaslow@tamu.edu
†Contact author: chensun@hnu.edu.cn

measure them. The Appendix compares notation in the current work and in previous work.

These two new charge currents, together with the proposal to measure them to verify the original spin currents, are the main points of this paper.

II. THERMODYNAMICS AND EQUATIONS OF MOTION

We approach the problem using Onsager's irreversible thermodynamics. We consider charge carriers with negative charge $-e$ and gyromagnetic ratio $-\gamma$, as normally appropriate for electrons. They are in an electrical potential V and have chemical potential μ. The electrochemical potential $\tilde{\mu} = \mu - eV$. With $E_i = -\partial_i V$, we employ the effective electric field

$$E_i^* = \frac{1}{e}\partial_i\tilde{\mu} = E_i + \frac{1}{e}\partial_i\mu. \tag{1}$$

A. Thermodynamics

We take the energy density to be a function of the entropy density s (with thermodynamic conjugate the temperature T), the carrier density n (with thermodynamic conjugate the electrochemical potential $\tilde{\mu} = \mu - eV$), and the spin density $\vec{S}$ (proportional to the magnetization $\vec{M}$ and the vector polarization density $\vec{n}$), where

$$\vec{M} = -\gamma\vec{S} = -\frac{\gamma\hbar}{2}\vec{n}. \tag{2}$$

We use units where B is in tesla (T) and both M and H are in A/m: $\vec{B} = \mu_0(\vec{H} + \vec{M})$.

Then for the differential of the energy density we take

$$d\varepsilon = T\,ds + \tilde{\mu}\,dn + \vec{\mu}\cdot d\vec{n}. \tag{3}$$

Here $\vec{\mu}$ is the spin-space vector chemical potential, as defined in Eq. (16) of Ref. [8], which calls $\vec{\mu}$ the spin accumulation, a term that properly refers to either $\vec{S}$ or $\vec{M}$ [1,6].

$\vec{\mu}$ is related to the effective magnetic field $\vec{B}^* \equiv -\partial\varepsilon/\partial\vec{M}$, which includes both the applied field $\vec{B}$ and the internal field. In local equilibrium $\vec{B}^*_{le} = \vec{0}$. This condition is satisfied by writing the (effective) magnetic field energy as

$$-\vec{B}^*\cdot d\vec{M} = \vec{\mu}\cdot d\vec{n}, \tag{4}$$

from which we deduce that

$$\vec{\mu} = \frac{\gamma\hbar}{2}\vec{B}^*. \tag{5}$$

Thus, in equilibrium $\vec{\mu} = \vec{0}$.

B. Equations of motion

With unknown fluxes (we use J rather than j, in order to free j to be used as an index) and unknown sources R, the equations of motion are taken to be

$$\partial_t\varepsilon + \partial_i J_i^\varepsilon = 0, \tag{6}$$

$$\partial_t s + \partial_i J_i^s = R^s \geqslant 0, \tag{7}$$

$$\partial_t n + \partial_i J_i^n = 0, \tag{8}$$

$$\partial_t\vec{n} + \partial_i\vec{J}_i^{\vec{n}} = \gamma\vec{n}\times\vec{B} + \vec{R}^{\vec{n}}, \tag{9}$$

where J_i^ε, J_i^s, J_i^n, and $\vec{J}_i^{\vec{n}}$ are the energy density flux, the entropy density flux, the charge current (charge flux), and the spin current (spin polarization flux), respectively, R^s is the entropy density production rate, and $\vec{R}^{\vec{n}}$ is the source of the vector number density. $\vec{R}^{\vec{n}}$ can include decay of the magnetization $\vec{M} = -(\gamma\hbar/2)\vec{n}$.

We now rewrite TR^s in terms of a divergence and of products of thermodynamic fluxes or sources with their respective thermodynamic forces. To do so we use the energy differential (3) and the equations of motion. We find that

$$\begin{aligned} 0 \leqslant TR^s = &-\partial_i\big(J_i^\epsilon - TJ_i^s - \tilde{\mu}j_i^n - \vec{\mu}\cdot\vec{J}_i^{\vec{n}}\big) \\ &- J_i^s\partial_i T - J_i^n\partial_i\tilde{\mu} - \vec{J}_i^{\vec{n}}\cdot\partial_i\vec{\mu} - \vec{\mu}\cdot\vec{R}^n. \end{aligned} \tag{10}$$

The divergence term in Eq. (10) must vanish since otherwise it can be of either sign, and $TR^s \geqslant 0$ cannot be satisfied.

We can think of the thermodynamic forces as an abstract vector $F = (\partial_i T, \partial_i\mu, \partial_i\vec{\mu}, \vec{\mu})$ and the thermodynamic fluxes and sources as another abstract vector $C = (J_i^s, J_i^n, \vec{J}_i^{\vec{n}}, \vec{R}_i^n)$. They are connected by

$$C_a = -L_{ab}F_b, \tag{11}$$

where L_{ab} is the Onsager matrix of transport coefficients and we sum on the index b. The entropy production equation (10) then becomes (on summing over repeated indices)

$$0 \leqslant TR^s = -C_aF_a = L_{ab}F_bF_a. \tag{12}$$

If a pair of forces has the same time-reversal signature, then Onsager showed that $L_{ab} = L_{ba}$, leading to dissipation. If a pair of forces have the opposite time-reversal signature, then their product is odd under time reversal. Thus if their product is nonzero, it can change sign under time reversal. However, this is forbidden by the non-negative rate of entropy production. Therefore, if their product is nonzero, $L_{ab} = -L_{ba}$, with no dissipation.

Because L_{ab} can have parts that are both dissipative (irreversible) and nondissipative (reversible, or reactive), we can summarize this by writing $L_{ab} = L_{ab}^D + L_{ab}^R$, where

$$L_{ab}^D = L_{ba}^D \text{ (dissipation)}, \quad L_{ab}^R = -L_{ba}^R \text{ (no dissipation)}. \tag{13}$$

Another way to look at dissipation (irreversibility) versus nondissipation (reversibility) is to consider a flux term driven by a thermodynamic force. If that driven flux term has the opposite (as opposed to the same) time-reversal signature to the intrinsic time-reversal signature of the flux—see below—then that driven flux term is dissipative (as opposed to nondissipative).

The intrinsic time-reversal signature of a flux is determined by the product of the time-reversal signatures of its corresponding density and velocity. Thus, the entropy density flux J_i^s has entropy density s (even) times velocity (odd), so its intrinsic time-reversal signature is odd, but the spin polarization flux $J_i^{\vec{n}}$ has spin polarization density $\vec{n}$ (odd) times velocity (odd), so its intrinsic time-reversal signature is even.

C. Fluxes and sources

For the diagonal transport coefficients, where the fluxes are proportional to the associated thermodynamic forces, we have

$$J_i^s = -\frac{\kappa}{T}\partial_i T, \tag{14}$$

$$J_i^n = -\frac{\sigma}{e^2}\partial_i \tilde{\mu}, \tag{15}$$

$$\vec{J}_i^{\vec{n}} = -\frac{\sigma'}{e^2}\partial_i \vec{\mu}, \tag{16}$$

$$\vec{R}^n = -\Gamma\vec{\mu}. \tag{17}$$

For completeness, note that the intrinsic time-reversal signatures of the "fluxes" (J_i^s, J_i^n, $\vec{J}_i^{\vec{n}}$, $\vec{R}^n$) are (odd, odd, even, even), and the time-reversal signatures of the "forces" ($\partial_i T$, $\partial_i\tilde{\mu}$, $\partial_i\vec{\mu}$, $\vec{\mu}$) are (even, even, odd, odd). Therefore, all of the above terms are dissipative. Here κ, σ, and σ' all represent diagonal dissipative processes (of s, n, and $\vec{n}$), and Γ represents a decay process. To have $TR^s \geqslant 0$ these all are non-negative.

We do not include anisotropy due to the magnetization $\vec{M}$, although each of the dissipative coefficients (κ, σ, σ', Γ) can be replaced by a tensor of the form $A_\parallel \hat{M}_\alpha \hat{M}_\beta + A_\perp(\delta_{\alpha\beta} - \hat{M}_\alpha\hat{M}_\beta)$. These represent diffusion and decay effects that depend on whether they involve processes along or normal to $\hat{M}$.

D. Off-diagonal Onsager relations

We now consider the off-diagonal Onsager coefficients. In the remainder of this section only, we give them the cumbersome subscript *od*. In the present case, the SOI causes mixing of transport from $\tilde{\mu}$ and $\vec{\mu}$ [10]. We neglect temperature gradients.

Therefore, when the off-diagonal transport coefficients correspond to reactive (nondissipative) processes, the cross terms cancel. Thus

$$J_{i,od}^{n,R}\partial_i\tilde{\mu} = -\vec{J}_{i,od}^{\vec{n},R}\cdot\partial_i\vec{\mu} \quad \text{(nondissipative)}. \tag{18}$$

When the off-diagonal transport coefficients correspond to dissipative processes (either diffusion or decay), the cross terms add, thus doubling the rate of entropy production. Thus,

$$J_{i,od}^{n,D}\partial_i\tilde{\mu} = \vec{J}_{i,od}^{\vec{n},D}\cdot\partial_i\vec{\mu} \quad \text{(dissipative)}. \tag{19}$$

Relation (19) is analogous to what is used to derive the reciprocal relations between heat current driven by electrochemical potential gradients and charge current driven by temperature gradients.

We apply Eqs. (18) and (19) to the spin currents of the theories of DP [2], TGS [7], and ALSH [9].

III. DISSIPATIVE (IRREVERSIBLE) FLUXES VS NONDISSIPATIVE (REVERSIBLE) FLUXES

Fluxes can be driven by the intrinsic signature of fluxes, such as of charge and heat, and are determined independently of their cause. The charge and heat fluxes are proportional to the velocity of the excitations, and thus their intrinsic time-reversal signature is always odd, independent of what drives these fluxes. The time-reversal signatures of thermodynamic forces are determined from those forces alone.

Because of Joule heating, the charge flux J_i^n, driven by a gradient in the electrochemical potential $\partial_j\tilde{\mu}$, has an even signature under time reversal. This is opposite the intrinsic time-reversal signature of the charge flux and thus is dissipative, or irreversible.

On the other hand, the intrinsic flux has an odd signature under time reversal. When the flux driven by a thermodynamic force has the same (opposite) time-reversal signature as that of the intrinsic flux, the driven flux is reversible (irreversible).

For the ordinary Hall effect, the Hall charge flux, driven by the gradient $\partial_j\tilde{\mu}$, is normal both to that gradient and to the applied magnetic field $\vec{B}$. As above, the intrinsic time-reversal signature of the charge flux is odd. Also, the Hall charge flux has an odd time-reversal signature. Therefore, the Hall charge flux is reversible.

In the Nernst effect, a temperature gradient in a normal magnetic field drives a charge flux that is normal to both of them. The charge flux driven by the temperature gradient has an odd time-signature because it is proportional to the (odd) magnetic field. This is the same as the time-reversal signature of the intrinsic charge flux and therefore is reversible.

In the thermoelectric effect, a charge flux is induced by a temperature gradient, and in the electrothermal effect, a heat flux is driven by an electrochemical potential gradient. The rate of heating from the thermoelectric effect involves the product of the temperature gradient and the electrochemical potential gradient

IV. WEAKLY SPIN-POLARIZED SYSTEM WITH SOI: REWRITING THE DP MODEL

Before considering ferromagnets, we discuss paramagnets, as considered in the DP model [1,2], which we consider to be weakly spin-polarized systems. Dyakonov and Perel considered the spin flux first and then the charge flux [1,2], but we consider them in the opposite order.

For a paramagnet in equilibrium, we define χ via $\vec{M} = (\chi/\mu_0)\vec{B}$. Then, out of equilibrium, we take

$$\vec{B}^* = \vec{B} - \frac{\mu_0}{\chi}\vec{M} = \vec{B} + \frac{\gamma\hbar}{2}\frac{\mu_0}{\chi}\vec{n}, \tag{20}$$

so

$$\vec{\mu} = \frac{\gamma\hbar}{2}\vec{B} + \lambda\vec{n}, \quad \lambda \equiv \frac{\mu_0}{\chi}\left(\frac{\gamma\hbar}{2}\right)^2. \tag{21}$$

Also, for a spin-1/2 paramagnet,

$$\chi = \left(\frac{\gamma\hbar}{2}\right)^2\frac{n_c}{k_B T}, \tag{22}$$

where n_c is the average number density of charge carriers.

A. Charge flux J_i^n

Following (8.7) of Ref. [11], and eliminating gradients of the density by using E_i^* for E_i, with μ_c being the charge mobility (μ has already been used for the chemical potential), we have

$$J_i^n = -\mu_c n_c E_i^* - \beta(\vec{E}^*\times\vec{n})_i - \delta(\vec{\nabla}\times\vec{n})_i, \tag{23}$$

where β and δ (which can have either sign) are parameters due to the SOI. For a nondegenerate semiconductor, $\beta = e\delta/k_BT$ [1,2]. For a paramagnet, the conductivity $\sigma = en_c\mu_c$. For a non-negative rate of entropy production, we have $\sigma, \mu_c \geqslant 0$. In Eq. (23) the first term is dissipative, and the second and third terms are reactive.

We now use Eqs. (1) and (21) at fixed $\vec{B}$. Then we may rewrite Eq. (23) as

$$J_i^n = -\frac{n_c\mu_c}{e}\partial_i\tilde{\mu} - \frac{\beta}{e}(\vec{\nabla}\tilde{\mu}\times\vec{n})_i - \frac{\delta}{\lambda}(\vec{\nabla}\times\vec{\mu})_i. \quad (24)$$

The first term is the ordinary conductivity, the second term leads to the anomalous Hall effect, and the third term leads to the inverse spin Hall effect. Consistent with Onsager's irreversible thermodynamics, the driving terms are gradients of $\tilde{\mu}$ and $\vec{\mu}$; these gradients are zero in equilibrium. In Ref. (24), the first term is dissipative, and the second and third terms are reactive.

B. Spin polarization flux $\vec{J}_i^{\vec{n}}$

It is not uncommon for "spin flux" to refer to spin polarization flux (a density times a velocity), spin flux (an extra factor of $\hbar/2$), or magnetization flux (for electrons, an extra factor of $-\gamma\hbar/2$).

We follow Eq. (37) of Ref. [12], which is a rewritten version of Eq. (8.8) of Ref. [11]:

$$\left(\vec{J}_i^{\vec{n}}\right)_j \equiv J_{ij} = -\frac{\mu_c}{e}(\partial_i\tilde{\mu})n_j - \frac{D}{\lambda}\partial_i\mu_j + \epsilon_{ijk}\frac{\beta n_c}{e}\partial_k\tilde{\mu}, \quad (25)$$

where for simplicity we denote $(\vec{J}_i^{\vec{n}})_j \equiv J_{ij}$. For a semiconductor, an Einstein relation gives the diffusion constant

$$D = \frac{\sigma}{e^2}\frac{\partial\mu}{\partial n_c} \approx \frac{\sigma}{e^2}\frac{k_BT}{n_c} = \frac{n\mu_c}{e}k_BT. \quad (26)$$

In Eq. (25), the first and second terms are dissipative, and the third term is reactive.

C. DP1 and Onsager reciprocity

Equation (10) contains the terms $-J_i^n\partial_i\tilde{\mu} - \vec{J}_i^{\vec{n}}\cdot\partial_i\vec{\mu}$, which must be non-negative. Using Eq. (24) for J_i^n, the first term of $-J_i^n\partial_i\tilde{\mu}$ and, using Eq. (25) for $\vec{J}_i^{\vec{n}}$, the second term of $-\vec{J}_i^{\vec{n}}\cdot\partial_i\vec{\mu}$, are indeed non-negative. Being quadratic in the respective thermodynamic forces $\partial_i\tilde{\mu}$ and $\partial_i\vec{\mu}$, Onsager reciprocity does not apply to these terms.

Onsager reciprocity applies to the off-diagonal terms associated with the third terms of Eqs. (24) and (25). These give products of $\partial_i\tilde{\mu}$ and $\partial_i\vec{\mu}$ with respective coefficients δ/λ and $\beta n_c/e$, which can be shown to satisfy Onsager reciprocity. These cross terms can be of either sign and thus must not be too large or TR^s can become negative.

The second term of Eq. (24), multiplied by $\partial_i\tilde{\mu}$, is identically zero. The first term of Eq. (25), multiplied by $\partial_i\vec{\mu}$, is nonzero, but proportional to the small quantity $\vec{n}$, so we neglect it. Thus, in the end, the original theory of DP1 satisfies Onsager reciprocity [1].

D. DP2 spin-swapping spin flux

A week after giving the first version of the spin flux, DP2 added to the spin flux some additional terms due to the SOI [2]. Decades later, Lifshits and Dyakonov called these "spin-swapping" terms [4].

The spin-swapping terms, which are corrections to Eq. (25), are written implicitly in Eq. (3) of Ref. [4], which introduces the new spin-swapping parameter κ. Reference [12], in Eq. (38), gives them as

$$\Delta J_{ij} = -\frac{\kappa\mu_c}{e}(n_i\partial_j\tilde{\mu} - \delta_{ij}\vec{n}\cdot\vec{\nabla}\tilde{\mu}) - \frac{\kappa D}{\lambda}(\partial_j\mu_i - \delta_{ij}\vec{\nabla}\cdot\vec{\mu}). \quad (27)$$

Reference [12] uses κ_s for κ; these are dimensionless. In Eq. (27), all of the terms are dissipative.

The κD terms in the spin-swapping part of the spin flux are diagonal in the matrix of fluxes vs thermodynamic forces. However, the $\kappa\mu_c$ terms are off-diagonal, so by Onsager's reciprocity principle they must have corresponding terms in the charge current. Let us write these $\kappa\mu_c$ terms in Eq. (27), involving gradients of $\tilde{\mu}$ explicitly:

$$\Delta J_{ij}^{\tilde{\mu}} = -\frac{\kappa\mu_c}{e}(n_i\partial_j\tilde{\mu} - \delta_{ij}\vec{n}\cdot\vec{\nabla}\tilde{\mu}). \quad (28)$$

DP2 gives no corresponding charge flux terms, which by Onsager should appear in the theory.

E. Onsager gives DP2 a spin-swapping charge flux

The intrinsic (and reversible) time signature of ΔJ_{ij} is that of spin times velocity, and thus is even. On the other hand, irreversible thermodynamics gives Eq. (27) for ΔJ_{ij}, for which the time signature of each term on the right-hand-side is odd; this is because $\tilde{\mu}$ is even, but $\vec{n}$ and $\vec{\mu}$ are odd. Thus, the intrinsic and irreversible thermodynamics time signatures are opposite, indicating that ΔJ_{ij} is irreversible. This is consistent with the condition that the dissipation rate $\Delta J_{ij}\partial_i\mu_j$ be positive and invariant under time reversal.

By Onsager's reciprocity principle for dissipative terms, Eq. (19) then gives

$$\Delta J_i^{\vec{\mu}} = -\frac{\kappa\mu_c}{e}(n_j\partial_j\mu_i - n_i\partial_j\mu_j). \quad (29)$$

This result is implicit, but not commented on, in Ref. [12].

V. STRONGLY SPIN-POLARIZED SYSTEM: SPIN AND CHARGE FLUXES FROM TGS AND ALSH

We now present the spin and charge flux from both TGS and ALSH, which are specifically for ferromagnets and, more generally, for strongly spin-polarized systems.

A. TGS has spin flux and charge flux

TGS [7] use the spin flux $Q_{ji} = (\hbar/2)J_{ij}$, with indices opposite of the DP convention that we employ. Their Eqs. (6) and (7) use conductivities σ, $\sigma_{\rm AH}$, and $\sigma_{\rm AMR}$ and introduce the dimensionless coefficients β (which we write as β^T because it differs from β of DP), ζ, and η. We write their charge flux as J_i^{T} and their spin flux as J_{ij}^{T}:

$$J_i^{\mathrm{T}} = -\frac{\sigma}{e^2}\partial_i\{\tilde{\mu} + \beta^T(\hat{M}\cdot\vec{\mu})\} - \frac{\sigma_{\mathrm{AH}}}{e^2}(\hat{M}\times\vec{\nabla})_i\{\tilde{\mu} + \zeta(\hat{M}\cdot\vec{\mu})\} - \frac{\sigma_{\mathrm{AMR}}}{e^2}\hat{M}_i(\hat{M}\cdot\vec{\nabla})\{\tilde{\mu} + \eta(\hat{M}\cdot\vec{\mu})\}, \tag{30}$$

$$J_{ij}^{\mathrm{T}} = -\frac{\sigma}{e^2}\hat{M}_j\partial_i\{(\hat{M}\cdot\vec{\mu}) + \beta^T\tilde{\mu}\} - \frac{\sigma_{\mathrm{AH}}}{e^2}\hat{M}_j(\hat{M}\times\vec{\nabla})_i\{(\hat{M}\cdot\vec{\mu}) + \zeta\tilde{\mu}\}_i - \frac{\sigma_{\mathrm{AMR}}}{e^2}\hat{M}_i\hat{M}_j(\hat{M}\cdot\vec{\nabla})\{(\hat{M}\cdot\vec{\mu}) + \eta\tilde{\mu}\}. \tag{31}$$

The off-diagonal terms in ΔJ_i^{T} and $\Delta J_{ij}^{\mathrm{T}}$ for which Onsager relations apply are those proportional to β^T, ζ, and η. [We neglected these terms in Eqs. (15) and (16).] Indeed they satisfy the appropriate Onsager relations.

Because TGS omit transverse spin diffusion, this J_{ij} includes no terms normal to $\hat{M}_j$. Note that the third set of terms in Eqs. (30) and (31) can be thought of as due to "tensorization" of the gradients of its first set of terms. In Eqs. (15) and (16), the only reversible terms are those proportional to σ_{AH}; the others are dissipative.

B. ALSH has only spin flux

ALSH find a new spin flux term, which in its general form is given in their footnote [16] in Ref. [9]. Their symbol σ, which is associated with spin-scattering, is *not* the conventional conductivity; to avoid confusion in sections involving ALSH, only in these sections do we employ $\tilde{\sigma}$ for the conductivity.

On rewriting their spin flux in tensor form, we find

$$J_{ij}^{\mathrm{A}} = \frac{\tilde{\sigma}_\parallel - \tilde{\sigma}_\perp}{e^2}\hat{M}_j\hat{M}_k\varepsilon_{ikl}\partial_l\tilde{\mu} + \frac{\tilde{\sigma}_\perp}{e}\varepsilon_{ijl}\partial_l\tilde{\mu}. \tag{32}$$

This J_{ij}^{A} is driven by $\partial_i\Delta\tilde{\mu}$, which means that the Onsager coefficients are off-diagonal. Therefore, there must be corresponding J_i^{A} terms driven by $\partial_i\vec{\mu}$. The first term above has the symmetry of the ζ term in Eq. (31). The second term corresponds to the last (spin Hall effect) term in Eq. (25). In Eq. (32) all of the terms are reversible.

C. Onsager reciprocity and ALSH spin flux implies ALSH charge flux

The time signature of J_{ij}^{A} is even, and each of the new terms in Eq. (32) is even under time reversal. Therefore the new terms are nondissipative and do not contribute to the rate of entropy production. Thus, Eq. (18) applies.

From Eq. (18) we deduce that

$$J_i^{\mathrm{A}} = -\frac{1}{e^2}[(\tilde{\sigma}_\parallel - \tilde{\sigma}_\perp)\hat{M}_l\hat{M}_k\varepsilon_{ijk} + \tilde{\sigma}_\perp\varepsilon_{ijl}]\partial_j\mu_l. \tag{33}$$

For $\hat{M} = \hat{z}$, so $\hat{M}_k = \delta_{kz}$, this gives

$$J_i^{\mathrm{A}} = -\frac{1}{e^2}[(\tilde{\sigma}_\parallel - \tilde{\sigma}_\perp)\varepsilon_{ijz}\partial_j\mu_z + \tilde{\sigma}_\perp\varepsilon_{ijl}\partial_j\mu_l]. \tag{34}$$

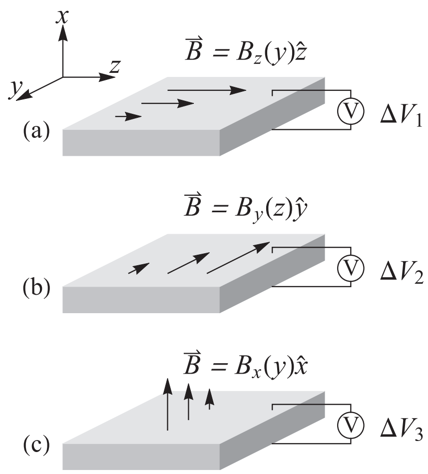

FIG. 1. Experimental geometries for measuring voltages corresponding to the new charge currents that will verify the existence of the spin currents predicted by (a), (b) the ALSH model and (c) the DP model.

The first term and the second term with $l = z$ together give a contribution of $J_i = -(1/e^2)\tilde{\sigma}_\parallel\varepsilon_{ijz}\partial_j\mu_z$, and the second term with $l = (x, y)$ gives a contribution of $J_i = -(1/e^2)\tilde{\sigma}_\perp\varepsilon_{ijl}\partial_j\mu_l$. The sum is thus

$$J_i^{\mathrm{A}} = -\frac{\tilde{\sigma}_\parallel}{e^2}\varepsilon_{ijz}\partial_j\mu_z - \frac{\tilde{\sigma}_\perp}{e^2}[\varepsilon_{ijx}\partial_j\mu_x + \varepsilon_{ijy}\partial_j\mu_y]. \tag{35}$$

In Eq. (35), all of the terms are reversible.

VI. EXPERIMENTAL CONSEQUENCES

A. ALSH

This charge current can be measured. Consider a thin sample with dimensions l_x, l_y, and l_z, where $l_x \ll l_y, l_z$ (as shown in Fig. 1). Recall that, by Eqs. (5) and (20) $d\vec{\mu} = (\gamma\hbar/2)d\vec{B}^* \approx (\gamma\hbar/2)d\vec{B}$, where we assume that $\vec{M}$ is nonzero but effectively pinned in place, so that for purposes of making an estimate the deviation $d\vec{M}$ can be neglected. According to Eq. (35), if we apply a field $\vec{B} = B_z(y)\hat{z}$, then a charge current along x, of $J_{x,1}^{\mathrm{A}} = -(\tilde{\sigma}_\parallel/e^2)\partial_y\mu_z$, will be generated.

Let us assume isotropic charge conductivity σ. If there are no leads along $\hat{x}$, then the equilibrium ΔV_1 along x is determined by the condition that the charge current $J_{x,1}^{\mathrm{A}}$ is canceled by the conventional dissipative charge current $\sigma_x\Delta V_1/l_x$. Thus,

$$\Delta V_1 = -\frac{l_x\tilde{\sigma}_\parallel}{e\sigma}\partial_y\mu_z = -\frac{\tilde{\sigma}_\parallel}{\sigma}\frac{\gamma\hbar}{2e}l_x\partial_y B_z, \tag{36}$$

where the negative sign means that the electric potential at the $x = l_x$ surface is higher than that at the $x = 0$ surface [see Fig. 1(a)].

Similarly, if a magnetic field of the form $\vec{B} = B_y(z)\hat{y}$ is applied, then a charge current along x, of $J_{x,2}^{\mathrm{A}} = (\tilde{\sigma}_\perp/e^2)\partial_z\mu_y$, will be generated, and there should be a voltage ΔV_2 along x:

$$\Delta V_2 = \frac{l_x\tilde{\sigma}_\perp}{e\sigma}\partial_z\mu_y = \frac{\tilde{\sigma}_\perp}{\sigma}\frac{\gamma\hbar}{2e}l_x\partial_z B_y. \tag{37}$$

Therefore, measurements of finite ΔV_1 and ΔV_2 will test the present theory and determine the values of $\tilde{\sigma}_\parallel$ and $\tilde{\sigma}_\perp$ [see Fig. 1(b)].

We now estimate the magnitude of the magnetic field gradient needed to produce a measurable voltage ΔV. From Table I of Ref. [9], values of $\tilde{\sigma}_\parallel$ and $\tilde{\sigma}_\perp$ for Fe, Ni, and Co range from 10^2 to $2\times 10^3\ \Omega^{-1}\,\mathrm{cm}^{-1}$. We take $\sigma = 1.0\times 10^5\ \Omega^{-1}\,\mathrm{cm}^{-1}$, which is not far from the values for Fe, Ni, and Co, and $l_x = 1$ mm. Then to produce a voltage of $|\Delta V| = 100\ \mu$V, we estimate the field gradient $\partial_y B_z$ or $\partial_z B_y$ to be from 0.086 to 1.7 T/μm.

B. DP2

We now discuss possible experimental observation of the spin-swapping charge currents (29) from DP. Again consider a thin sample with dimensions l_x, l_y, and l_z, where $l_x \ll l_y, l_z$. From Eq. (29), we have

$$\Delta J_x = -\frac{\kappa\mu_c}{e}(n_y\partial_y\mu_x + n_z\partial_z\mu_x - n_x\partial_y\mu_y - n_x\partial_z\mu_z). \quad (38)$$

If $\vec{n} = P\hat{y}$ and we apply a field $\vec{B} = B_x(y)\hat{x}$, then a charge current $\Delta J_x = -(\kappa\mu_c/e)P\partial_y\mu_x$ will be generated, and analysis similar to that for ALSH predicts the following voltage:

$$\Delta V_3 = -\frac{l_x\kappa\mu_c n_c}{\sigma}\partial_y\mu_x = -\frac{\kappa\mu_c P}{\sigma}\frac{\gamma\hbar}{2}l_x\partial_y B_x \quad (39)$$

[see Fig. 1(c)].

We take $P = 0.01n_c$, where n_c is the charge carrier density and note that $\mu/\sigma = 1/(n_c e)$. Then $\Delta V_3 = -0.01\kappa\gamma\hbar/(2e)l_x\partial_y B_x$. Lifshits and Dyakonov [4] estimate κ to be 0.3 for InSb (large SOI) and 0.003 for GaAs (small SOI). Thus, to produce a voltage of $|\Delta V_3| = 100\ \mu$V for a $l_x = 1$ mm sample, we estimate $\partial_y B_x = 0.57$ T/μm for $\kappa = 0.3$ and $\partial_y B_x = 57$ T/μm for $\kappa = 0.003$.

One may be concerned about the feasibility of the proposed experiments because the estimated magnetic field gradients are large. However, these estimates are done for a sample thickness of $l_x = 1$ mm and a measured voltage of $|\Delta V| = 100\ \mu$V, whereas in an experiment one could take a larger l_x and/or a smaller $|\Delta V|$ to reduce the required magnetic field gradient. Moreover, one could also employ other materials with material parameters requiring a smaller field gradient, e.g., one with a smaller charge conductivity σ. The expressions for ΔV_1, ΔV_2, and ΔV_3 show clearly how to choose experimental parameters to achieve a smaller field gradient.

VII. SUMMARY AND CONCLUSIONS

We have developed the irreversible thermodynamics of magnets supporting spin currents and charge currents. We specifically showed that Onsager relations imply that, when there are spin currents driven by voltage gradients, there are also charge currents driven by field gradients, which may enable measurement of effects predicted by Dyakonov and Perel [2] and by Amin *et al.* [9] Observation of these charge currents would corroborate the spin currents predicted by Refs. [2,9].

ACKNOWLEDGMENTS

C.S. was supported by the National Natural Science Foundation of China under Grant No. 12105094 and by the Fundamental Research Funds for the Central Universities from China.

APPENDIX: COMPARISON OF VARIOUS NOTATIONS

This Appendix compares the notation of the current work and several previous works. Recall that we use $\vec{n}$, J_i^n, and $J_i^{\vec{n}}$ for the vector polarization density, the charge current, and the spin current, respectively.

DP and Ref. [12] use q_i and $\vec{q}_i$ to denote the dimensionless charge and spin currents. Their relations to our notation are

$$q_i \equiv J_i^n, \quad \vec{q}_i \equiv \vec{J}^{\vec{n}}. \quad (A1)$$

Their dimensionless spin polarization $\vec{P}$ is equivalent to the vector number density $\vec{n}$ used in this work:

$$\vec{P} \equiv \vec{n}. \quad (A2)$$

The corresponding charge and magnetization currents are related by

$$J_i^q \equiv -J_i^n = -q_i, \quad \vec{J}_i^{\vec{M}} \equiv -\frac{\gamma\hbar}{2}\vec{J}_i^{\vec{n}} = -\frac{\gamma\hbar}{2}\vec{q}_i. \quad (A3)$$

As discussed in the main text, TGS use the spin flux $Q_{ji} = (\hbar/2)J_{ij}$, with indices opposite those of our notation. Also note that in Eqs. (30) and (31) we have changed TGS's coefficient β to β^T, since β has been used in Eq. (23) of DP.

[1] M. I. Dyakonov and V. I. Perel, Possibility of orienting electron spins with current, Sov. Phys. JETP Lett. **13**, 467 (1971).

[2] M. I. Dyakonov and V. I. Perel, Current-induced spin orientation of electrons in semiconductors, Phys. Lett. A **35**, 459 (1971).

[3] With no knowledge of Refs. [1,2], and using only qualitative reasoning, the spin Hall and inverse spin Hall effects were predicted and named by J. E. Hirsch, Spin Hall effect, Phys. Rev. Lett. **83**, 1834 (1999); see also, Overlooked contribution to the Hall effect in ferromagnetic metals, Phys. Rev. B **60**, 14787 (1999).

[4] M. B. Lifshits and M. I. Dyakonov, Swapping spin currents: interchanging spin and flow directions, Phys. Rev. Lett. **103**, 186601 (2009).

[5] M. I. Dyakonov, Magnetoresistance due to edge spin accumulation, Phys. Rev. Lett. **99**, 126601 (2007).

[6] T. Valet and A. Fert, Theory of the perpendicular magnetoresistance in magnetic multilayers, Phys. Rev. B **48**, 7099 (1993).

[7] T. Taniguchi, J. Grollier, and M. D. Stiles, Spin-transfer torques generated by the anomalous Hall effect and anisotropic magnetoresistance, Phys. Rev. Appl. **3**, 044001 (2015).

[8] Y. Tserkovnyak, A. Brataas, G. E. W. Bauer, and B. I. Halperin, Nonlocal magnetization dynamics in ferromagnetic heterostructures, Rev. Mod. Phys. **77**, 1375 (2005).

[9] V. P. Amin, J. Li, M. D. Stiles, and P. M. Haney, Intrinsic spin currents in ferromagnets, Phys. Rev. B **99**, 220405(R) (2019).

[10] Off-diagonal transport coefficients are well-known in the thermoelectric effect, where a temperature difference drives a charge current, and its inverse, where a voltage difference drives a heat current.

[11] M. I. Dyakonov and A. V. Khaetskii, Spin Hall effect, in *Spin Physics in Semiconductors*, Springer Series in Solid-State Sciences, edited by M. Dyakonov (Springer, Berlin, 2008), pp. 211–243.

[12] W. M. Saslow, Spin Hall effect and irreversible thermodynamics: Center-to-edge transverse current-induced voltage, Phys. Rev. B **91**, 014401 (2015).