Irreversible thermodynamics of uniform ferromagnets with spin accumulation: Bulk and interface dynamics

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Using ideas from Landau's Fermi-liquid theory, we apply irreversible thermodynamics to conducting and insulating ferromagnets with magnetic variables \hat{M} for the quantization axis and for the spin accumulation \vec{m} of the nonequilibrium excitations; thus the total magnetization is taken to be $\vec{\mathcal{M}} = \vec{\mathcal{M}} + \vec{m}$. The resulting theory closely corresponds to the theory of Silsbee *et al.* [Silsbee, Janossy, and Monod, Phys. Rev. B **19**, 4382 (1979)]. For the bulk, in addition to confirming the usual Landau-Lifshitz equation for \hat{M} and a Bloch-like equation for \vec{m} (with a nonuniform precession term), there are two related cross-relaxation terms between the transverse parts of the nonequilibrium \vec{m} and \vec{M} . Unlike the *s*-*d* model, where in a field \vec{H} the equilibrium magnetization \vec{M} is nonzero. For the interface, the boundary condition for \hat{M} is given by micromagnetics, and that for \vec{m} is given by irreversible thermodynamics, where the current of transverse spins crossing the interface is proportional to the discontinuity in the transverse part of the vector spin chemical potential. \hat{M} , \vec{m} , and \vec{H} are coupled; in the decoupled approximation, we find the wave vectors for the modes of \hat{M} and the transverse \vec{m} . We discuss reciprocity between spin pumping ($\vec{\mathcal{M}}$ driven out of the ferromagnet) and spin transfer torque ($\vec{\mathcal{M}}$ driven into the ferromagnet).

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I. INTRODUCTION

In 1979, Silsbee, Janossy, and Monod [1] (SJM) employed a theory that was prescient relative to the modern field of spintronics [2,3]. In addition to a *d*-spin magnetization variable like the usual magnetization \vec{M} , it also included an *s*-spin magnetization variable like the spin accumulation \vec{m} of spintronics [4,5]. On the other hand, by considering how the (tensor) spin current \vec{j}_{mi} is driven through bulk and across interfaces, it could explain the phenomenon that the authors had observed, now known as *spin pumping* (SP) [6,7]. However, as we shortly show, it can also explain what was the yet-to-be-predicted phenomenon of *spin transfer torque* (STT) [8–13].

The relationship between these two reciprocal phenomena can be seen in the boundary condition on \vec{j}_{mi} for a conducting ferromagnet *F* to the left and an ordinary conductor *N* on the right [14]. With only transverse components \vec{m}_{\perp} and its effective field \vec{h}_{\perp}^* , SJM took \vec{j}_{mi} to be driven by the difference in \vec{h}_{\perp}^* across the interface. Since, as we show, the transverse part of the vector spin chemical potential $\vec{\mu}_{s\perp} = (\gamma \hbar/2)\mu_0 \vec{h}_{\perp}^*$, the SJM form for \vec{j}_{mi} is the spin analog of an ordinary electric current (or heat current) being driven across an interface by the difference across the interface in the ordinary chemical potential μ (or temperature *T*).

To discuss \dot{h}_{\perp}^* , consider a ferromagnet in a uniform equilibrium with static field \vec{H}_0 and equilibrium magnetization \vec{M}_0 along \hat{x} , and consider only the transverse components of various quantities.

Then, with dimensionless exchange constant λ and dimensionless magnetic susceptibility χ_f for the ferromagnet, and χ_n for the normal metal, the respective (spin-space) \vec{h}_{\perp}^* 's are

given by

$$\vec{h}_{f\perp}^* = \left(\vec{H}_f + \lambda \vec{M} - \frac{\vec{m}_f}{\chi_f}\right)_{\perp}, \quad \vec{h}_{n\perp}^* = \left(\vec{H}_n - \frac{\vec{m}_n}{\chi_n}\right)_{\perp}.$$
 (1)

(Below we show that these forms are consistent with local equilibrium.) Taking the \vec{H} 's to be the same for adjacent materials, with \mathcal{L} , a (surface) Onsager coefficient with dimension of velocity (Γ of Ref. [1]), Ref. [1] took the equivalent of

$$\vec{j}_{mx} = \mathcal{L}(\vec{h}_n^* - \vec{h}_n^*)_\perp \approx \mathcal{L}\left(\lambda \vec{M} - \frac{\vec{m}_f}{\chi_f} + \frac{\vec{m}_n}{\chi_n}\right)_\perp.$$
 (2)

For spin pumping, ferromagnetic resonance (FMR) drives a transverse $d\vec{M}$ that is a source term for the unknown spin flux \vec{j}_{mx} crossing the F/N interface, as well as the unknown spin accumulations \vec{m}_f and \vec{m}_n . These must all be determined by the boundary conditions. SJM found each of these quantities, with rightward \vec{j}_{mx} leaving F to enter N [1].

For spin transfer torque, a known leftward spin current \vec{j}_{mx} leaves N and enters F. Then, \vec{j}_{mx} in N serves as a source term for \vec{M} , \vec{m}_f , and \vec{m}_n ; these must all be determined by the boundary conditions. Using an equivalent bulk theory and special boundary conditions, in 2002, Zhang, Levy, and Fert (ZLF) studied spin transfer torque [5]. The sample average of the torque of the incoming \vec{m} yielded both the fieldlike and dissipationlike torque that were expected.

A. Outline

The present work applies irreversible thermodynamics to the bulk and surface dynamics of a conductor, using what we call the m-M model, which is distinct from the s-d model. In the m-M model, the total magnetization is

$$\dot{\mathcal{M}} = \dot{M} + \vec{m}. \tag{3}$$

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(Above we consider that $\overline{M} = M\widehat{M}$, where M often equals a fixed equilibrium value M_{eq} in a field \overline{H} . A change dM in $|\overline{M}|$ may be thought of as $\widehat{M} \cdot d\overline{m}$. Except when first-order terms in dM are important, in what follows we rarely distinguish between M and M_{eq} .) After discussing the thermodynamic variables and their thermodynamics, we write down their equations of motion with unknown flux and source terms. By imposing the condition that the fluxes and sources make the equations of motion consistent with thermodynamics at all times, and imposing non-negativity of entropy production and the appropriate symmetries, the form of the fluxes and sources is found. Appendix A considers the irreversible thermodynamics if \widetilde{M} and \widetilde{m} also apply to insulating ferromagnets. Appendix B considers issues of notation.

Irreversible thermodynamics largely yields agreement with the equations of motion assumed by SJM. There are a few differences: (1) There is a bulk precession term for \vec{m} when the system is nonuniform. (2) Rather than Bloch-like damping of \vec{M} , irreversible thermodynamics yields Landau-Lifshitz damping. (3) Both \vec{M} and \vec{m} have normally omitted cross-relaxation terms. (4) In equilibrium in a field, $\vec{m} = \vec{0}$, whereas $\vec{M} \neq \vec{0}$ in equilibrium in a field. (5) Irreversible thermodynamics yields boundary conditions for \vec{m} ; the boundary conditions on \vec{M} follow from micromagnetics.

The normal modes of \vec{H} , \vec{M} , and \vec{m} are coupled. Assuming perturbation theory, the normal modes and their wave vectors are found in zeroth order, and can be used to obtain the firstorder corrections to the normal modes. For the important case $\omega = 0$ (which is appropriate both to a dc spin current from *N* to *F*, and the second-order dc part of an ac spin pumped current from *F* to *N*), the \vec{H} mode decouples and the other coupled modes can be more readily obtained.

Besides ω , the equations of motion include two resonance frequencies, three independent decay times, a diffusion time, three dimensionless nondissipative parameters ($\lambda, \chi_f, \chi_\perp$), and the surface transport coefficient \mathcal{L} —a total of ten parameters. (This omits the new bulk nonuniform precession term and its surface analog.) Of these, at fixed temperature, only H and ω can be varied, but if T is permitted to vary (so M can vary), then three quantities are under experimental control.

Nevertheless, seven parameters are not under experimental control. For permalloy (Py), one can estimate all of these quantities, although the cross-relaxation times are relatively unknown. Although Ref. [14] obtained many of their parameters for a simpler theory involving electron spin resonance (ESR) of Li on Cu, it was achieved only through great effort in the data analysis.

In addition to its contributions to applied magnetics, by extending the idea of spin accumulation \vec{m} from nonmagnets to ferromagnets F, spintronics has brought attention to a more complex description of magnetic dynamics in the presence of collisions:

(1) The phenomenology for \vec{m} can be understood at the kinetic theory level as being associated with the out-of-equilibrium spinor distribution function for the electron states of the band theory [15]. Ultimately, \vec{m} is attributable to the spin excitations in Landau's Fermi-liquid theory when one includes collisions, which lead to the near-equilibrium phenomena of

diffusion and drift [16–19]. Although transverse excitations of the spin-density matrix, which lead to \vec{m}_{\perp} , are not eigenstates of a static Hamiltonian, they are nonzero when driven out of equilibrium, as in spin pumping and spin transfer torque.

(2) \hat{M} has a phenomenology [20] that can be understood at the energy band level by taking \hat{M} to be associated with the quantization axis, and M with the different occupations of the magnetic bands. Fermi-liquid theory has been used to determine the bending energy associated with nonuniform \hat{M} , from which the nonuniform exchange coefficient A can be determined (see below) [21].

(3) Even treating the total magnetization \mathcal{M} as the relatively complex quantity $\mathcal{M} = \mathcal{M} + \mathcal{m}$ is a simplification, as we now argue. For each momentum \vec{p} , let a uniform nonequilibrium magnetization density $\mathcal{M}(\vec{p})$ be given, whose sum over \vec{p} gives the full \mathcal{M} . The time development of $\mathcal{M}(\vec{p})$ is determined by the collision operator and precession in the exchange field, with a spectrum of times that quickly eliminates all but the parts of $\mathcal{M}(\vec{p})$ that contribute to \vec{m} and \vec{M} . The subsequent time development determines the amounts of \vec{m} and \vec{M} , with \vec{m} decaying much more rapidly than \vec{M} [22]. From \vec{m} and \vec{M} , one then obtains (3).

(4) Inclusion of a driving field at the relatively low frequency of ferromagnetic resonance will not change this general picture, and the resulting theory will be based on the response of \hat{M} and \vec{m} to the driving field, as described by irreversible thermodynamics.

II. VARIABLES, THERMODYNAMICS, AND CONSTITUTIVE RELATIONS

We now consider the irreversible thermodynamics for a conducting system with, in equilibrium, a uniform magnetization \vec{M} associated with a quantization axis \hat{M} , a longitudinal spin accumulation $dM\hat{M}$, and a transverse spin accumulation \vec{m}_{\perp} . $dM = \hat{M} \cdot d\vec{m}$ and \vec{m}_{\perp} are, by definition, zero in equilibrium. We will employ

$$\vec{m} = \vec{m}_{\perp} + \hat{M}dM. \tag{4}$$

Consider a two-band conducting magnet with electrons of charge -e and gyromagnetic ratio $-\gamma$, where $\gamma = |g|\mu_B/\hbar > 0$ and $\mu_B = e\hbar/2m$ (for free electrons, we take g = -2). It is characterized by temperature *T*, up and down number densities $n_{\uparrow,\downarrow}$ (with $n_{\uparrow} < n_{\downarrow}$), equilibrium magnetic moment direction \hat{M} , and magnetization

$$M = -(\gamma \hbar/2)(n_{\uparrow} - n_{\downarrow}) \equiv -(\gamma \hbar/2)n_z.$$
 (5)

Note that the spin density $\vec{S} = -\vec{M}/\gamma$.

Clearly the total number density *n* is given by

$$n = n_{\uparrow} + n_{\downarrow}. \tag{6}$$

When we later consider the equations of motion, we will employ n_{\uparrow} and n_{\downarrow} , but for other purposes we will employ *M* and *n*. The longitudinal spin accumulation is

$$dM = -(\gamma \hbar/2)(dn_{\uparrow} - dn_{\downarrow}); \tag{7}$$

and

$$dn = dn_{\uparrow} + dn_{\downarrow}. \tag{8}$$

Because for equilibrium in a field $\vec{H}, \vec{m} = \vec{0}$, the *m*-*M* model differs from the *s*-*d* model. There the analogous quantity \vec{M}_s is nonzero in equilibrium for a field \vec{H} .

Thermodynamics. Irreversible thermodynamics requires that if a system initially satisfies thermodynamics, then it satisfies thermodynamics at all future times.

We thus consider the differential of the thermodynamic energy density ε (including the interaction with the static field \vec{H}_0), which must hold at all times. With V the voltage and -ethe charge on the electron, the electrochemical potentials $\tilde{\mu}_{\uparrow,\downarrow}$ are given in terms of the chemical potentials $\mu_{\uparrow\downarrow}$ as

$$\tilde{\mu}_{\uparrow\downarrow} = \mu_{\uparrow\downarrow} - eV. \tag{9a}$$

The differential $d\varepsilon$ includes terms involving the entropy density *s* and the temperature *T*, and the energy of interaction of the magnetizations with appropriate fields. Let the field \vec{H}^* couple to \vec{M} [23], and the field \vec{h}^*_{\perp} couple to \vec{m}_{\perp} . In terms of these and previously defined quantities, and the definitions

$$\mu_{\uparrow,\downarrow}^* = \tilde{\mu}_{\uparrow,\downarrow} \pm \mu_0(\gamma\hbar/2)\vec{H}^*\cdot\hat{M},\tag{9b}$$

we take (in SI units, where \vec{H} and \vec{M} are in A/m)

$$d\varepsilon = T ds + \tilde{\mu}_{\uparrow} dn_{\uparrow} + \tilde{\mu}_{\downarrow} dn_{\downarrow} - \mu_0 \vec{H}^* \cdot d\vec{M} - \mu_0 \vec{h}_{\perp}^* \cdot d\vec{m}_{\perp}$$

$$= T ds + \mu_{\uparrow}^* dn_{\uparrow} + \mu_{\downarrow}^* dn_{\downarrow}$$

$$- \mu_0 M \vec{H}^* \cdot d\hat{M} - \mu_0 \vec{H}^* \cdot \hat{M} dM - \mu_0 \vec{h}_{\perp}^* \cdot d\vec{m}_{\perp}.$$

(10)

This use of \vec{m} as a thermodynamic variable—even though it is zero in equilibrium—is implicit in Ref. [1].

Constitutive relations for \vec{H}^* and \vec{h}^* . Both \vec{H}^* and \vec{h}^* include the true magnetic field \vec{H} , which is the sum of the applied field \vec{H}_a and the dipole (or demagnetization) field \vec{H}_D . In addition, they include uniform exchange fields coupling one another, with interaction energy density

$$\varepsilon_{Mm} = -\mu_0 \lambda \vec{M} \cdot \vec{m}, \qquad (11)$$

and susceptibility fields proportional to the respective magnetization deviations. Here the "molecular field coefficient" λ is a dimensionless measure of the *uniform* exchange between \vec{M} and \vec{m} . This exchange term has the same structure as in Refs. [21,24].

Form for H^* . With χ_{\parallel} dimensionless, we take

$$H_{\parallel}^* = \vec{H}^* \cdot \hat{M} = \vec{H} \cdot \hat{M} - \frac{M - M_0}{\chi_{\parallel}} \equiv -\frac{dM}{\chi_{\parallel}}, \qquad (12)$$

so that in equilibrium both $H_{\parallel}^* = 0$ and the longitudinal spin accumulation dM = 0.

 \vec{H}^* is taken to include a nonuniform exchange field $(2A/\mu_0 M)\nabla^2 \hat{M}$, where A has units of J/m. Permitting \vec{M} to tip slightly from the equilibrium direction (which can be taken to be along \hat{z}), we take

$$\vec{H}_{\perp}^{*} = \left(\vec{H} + \lambda \vec{m} + \frac{2A}{\mu_{0}M} \nabla^{2} \hat{M}\right)_{\perp} - \frac{H}{M} \vec{M}_{\perp} \equiv -\frac{H}{M} \delta \vec{M}_{\perp},$$
(13)

so that in equilibrium $\vec{H}_{\perp}^* = \vec{0}$. In uniform equilibrium, \vec{M} rotates if the field rotates, giving a transverse susceptibility

$$\chi_{\perp} \equiv \frac{M}{H}.$$
 (14)

It is convenient to rewrite (13) as

$$\delta \vec{M}_{\perp} = -\chi_{\perp} \vec{H}_{\perp}^* = \vec{M}_{\perp} - \chi_{\perp} \left(\vec{H} + \lambda \vec{m} + \frac{2A}{\mu_0 M} \nabla^2 \hat{M} \right)_{\perp}.$$
(15)

We interpret δM_{\perp} as the deviation of M_{\perp} from instantaneous local equilibrium. When the system has wave vector k, so $\nabla^2 \rightarrow -k^2$, it is convenient to employ the nonuniform exchange wave vector k_A ,

$$k_A = \sqrt{\frac{\mu_0 M H}{2A}}.$$
 (16)

Then, (15) may be rewritten as

$$\delta \vec{M}_{\perp} = \vec{M}_{\perp} \left(1 + \frac{k^2}{k_A^2} \right) - \chi_{\perp} (\vec{H} + \lambda \vec{m})_{\perp}.$$
(17)

Form for \vec{h}^* . Since we have already considered equilibrium of the longitudinal spin accumulation in Eq. (12), we do not have to consider all three components from the viewpoint of equilibrium. However, we now endow \vec{h}^* with a nonzero longitudinal component $h_{\parallel}^* = \vec{h}^* \cdot \hat{M}$ about which \vec{m} precesses:

$$\vec{h}_{\parallel}^{*} = \vec{h}^{*} \cdot \hat{M} = \vec{H} \cdot \hat{M} + \lambda M.$$
(18)

Moreover, the transverse component of \vec{h}^* is given by

$$\vec{h}_{\perp}^{*} = \left(\vec{H} + \lambda \vec{M} - \frac{\vec{m}}{\chi_{f}}\right)_{\perp} = -\frac{\delta \vec{m}_{\perp}}{\chi_{f}}, \qquad (19)$$

so that in equilibrium $\vec{h}_{\perp}^* = 0$ and $\delta \vec{m}_{\perp} = \vec{0}$. We later show that the $\lambda \vec{M}_{\perp}$ term in \vec{h}_{\perp}^* is responsible for spin transfer torque and spin pumping. It is convenient to rewrite (19) as

$$\delta \vec{m}_{\perp} = \vec{m}_{\perp} - \chi_f (\vec{H} + \lambda \vec{M})_{\perp}. \tag{20}$$

We interpret $\delta \vec{m}_{\perp}$ as the deviation of \vec{m}_{\perp} from instantaneous local equilibrium [1,25–29].

Spin chemical potential $\vec{\mu}_s$ from \vec{h}_s^* . Associated with the spin accumulation, we define

$$\vec{h}_{s}^{*} \equiv \vec{h}_{\perp}^{*} + (\vec{H}^{*} \cdot \hat{M})\hat{M}, \quad d\vec{m} = d\vec{m}_{\perp} + \hat{M}dM.$$
(21)

Thus, $\vec{h}_{s\perp}^* = \vec{h}_{\perp}^*$ and $\vec{h}_{s\parallel}^* = H_{\parallel}^*$, which uses (12) [not $\vec{h}_{s\parallel}^* = h_{\parallel}^*$, which would use (18)].

We now rewrite the last two terms of (10) as $-\mu_0 \vec{h}_s^* \cdot d\vec{m}$, so that (10) becomes

$$d\varepsilon = T ds + \tilde{\mu}_{\uparrow} dn_{\uparrow} + \tilde{\mu}_{\downarrow} dn_{\downarrow} - \mu_0 M \vec{H}^* \cdot d\hat{M} - \mu_0 \vec{h}_s^* \cdot d\vec{m}.$$
(22)

In equilibrium, both \vec{m} and \vec{h}_s^* are zero.

Further, by defining a vector spin chemical potential

$$\vec{\mu}_s \equiv \frac{\gamma \hbar}{2} \mu_0 \vec{h}_s^*, \quad \vec{m} \equiv -(\gamma \hbar/2) \vec{n}, \tag{23}$$

so
$$\vec{\mu}_s \cdot d\vec{n} = -\mu_0 \vec{h}_s^* \cdot d\vec{m}$$
, we can rewrite (22) as
 $d\varepsilon = T ds + \tilde{\mu}_{\uparrow} dn_{\uparrow} + \tilde{\mu}_{\downarrow} dn_{\downarrow} - \mu_0 M \vec{H}^* \cdot d\hat{M} + \vec{\mu}_s \cdot d\vec{n}.$
(24)

 $\vec{\mu}_s$ also has been called the spin accumulation. However, $\vec{\mu}_s$ does not have the same dimensionality as the true spin accumulation \vec{m} . Moreover, because of the $\lambda \vec{M}$ term in \vec{h}^* , $\vec{\mu}_s$ and \vec{m} are not even proportional to one another. We present (22) and (24) for completeness; we do not employ them further [30].

III. EQUATIONS OF MOTION, "FLUXES"

For each of the thermodynamic densities, we assume an equation of motion that is first order in time, with associated fluxes and source terms to be determined. (We sometimes lump fluxes and sources together as "fluxes.") For the magnetic variables, we include appropriate precession terms. We linearize about equilibrium, so that when M appears with terms in dM, $d\hat{M}$, or \vec{m}_{\perp} , it is shorthand for its equilibrium value. Moreover, \vec{m} will be used for \vec{m}_{\perp} .

We take, for the energy and entropy densities, both of which have fluxes *j*,

$$\partial_t \varepsilon + \partial_i \, j_{\varepsilon i} = 0, \tag{25}$$

$$\partial_t s + \partial_i j_{si} = R_s \ge 0. \tag{26}$$

The fluxes $j_{\varepsilon i}$, j_{si} and the entropy source term $R_s \ge 0$ are to be determined. ε includes the Zeeman energy. For a static field, as here, we may neglect any energy changes from the field, and include the field-interaction energy with the internal energy of the magnetic system.

For the number densities, which have both fluxes and sources, we take

$$\partial_t n_{\uparrow} + \partial_i j_{\uparrow i} = R_{\uparrow}, \qquad (27)$$

$$\partial_t n_{\downarrow} + \partial_i j_{\downarrow i} = R_{\downarrow} = -R_{\uparrow}. \tag{28}$$

We have $R_{\downarrow} = -R_{\uparrow}$ because up and down spin together are conserved. The fluxes $j_{\uparrow i}$, $j_{\downarrow i}$ and the source term R_{\uparrow} are to be determined.

Equations for $\partial_t \hat{M}$ and $\partial_t \vec{m}_{\perp}$. As a unit vector, the quantization axis \hat{M} is subject only to rotation, and therefore has no flux term. On the other hand, \vec{m} is due to excitations and has both a flux term and a source term. Since for electrons the spin angular momentum and magnetization are oppositely directed, we take

$$\partial_t \hat{M} = -\gamma \hat{M} \times \mu_0 \vec{H}^* - \hat{M} \times \vec{\Omega}_M, \qquad (29)$$

$$\partial_t \vec{m} + \partial_i \vec{j}_{mi} = -\gamma \vec{m} \times \mu_0 \vec{h}^* + \vec{R}_m.$$
(30)

For \hat{M} , the dissipative "rotation rate" $\vec{\Omega}_M$ must be determined. For \vec{m} , the flux term \vec{j}_{mi} (which leads to spin diffusion) and the source term \vec{R}_m (which leads to spin decay) must be determined. To make the equations of motion for \hat{M} and \vec{m} more symmetrical, we use the fact that \vec{m} is normal to \hat{M} , so we may rewrite \vec{R}_m as

$$\vec{R}_m \equiv -\vec{M} \times \vec{\Omega}_m, \quad M\vec{\Omega}_m \equiv \hat{M} \times \vec{R}_m,$$
 (31)

thus introducing the "rotation rate" $\vec{\Omega}_m$.

Derived equations for $\partial_t n$ and $\partial_t M \equiv \partial_t m_{\parallel}$. The equations for $\partial_t \hat{M}$ and $\partial_t \vec{m}$ follow from (27) and (28). For *n*, we have

$$\partial_t n = \partial_t n_{\uparrow} + \partial_t n_{\downarrow} = -\partial_i j_{\uparrow i} - \partial_i j_{\downarrow i}, \qquad (32)$$

from which we deduce that

$$\partial_t n + \partial_i j_i = 0, \quad j_i \equiv j_{\uparrow i} + j_{\downarrow i}.$$
 (33)

Further, in $\vec{H}^* \cdot d\vec{M}$, the $\vec{H}^* \cdot dM$ term requires that

$$\partial_t M = -(\gamma \hbar/2)\partial_t (n_{\uparrow} - n_{\downarrow}) = (\gamma \hbar/2)\partial_i (j_{\uparrow i} - j_{\downarrow i} - 2R_{\uparrow}).$$
(34)

Introducing the longitudinal magnetization current j_{Mi} (or spin current) and the longitudinal magnetic source R_M via

$$j_{Mi} \equiv -(\gamma \hbar/2)(j_{\uparrow i} - j_{\downarrow i}), \qquad (35)$$

$$R_M \equiv -\gamma \hbar R_{\uparrow}, \qquad (36)$$

we rewrite (34) as

$$\partial_t M + \partial_i j_{Mi} = R_M. \tag{37}$$

Neither *n* nor *M* is relevant to the magnetic response transverse to \hat{M} .

IV. ENTROPY PRODUCTION, "FLUXES"

We now rewrite TR_s following a well-established procedure from irreversible thermodynamics [31–33]. Equations (25) and (27)–(30) placed in the time derivative of (26), on using (10), yield

$$0 \leqslant TR_{s} = T(\partial_{t}s + \partial_{i}j_{si})$$

$$= -\partial_{i}[j_{\varepsilon i} - Tj_{si} - \mu_{\uparrow}^{*}j_{\uparrow i} - \mu_{\downarrow}^{*}j_{\downarrow i} + \mu_{0}\vec{h}^{*} \cdot \vec{j}_{mi}]$$

$$+ [-j_{si}\partial_{i}T - j_{\uparrow i}\partial_{i}\mu_{\uparrow}^{*} - j_{\downarrow i}\partial_{i}\mu_{\downarrow}^{*} + \mu_{0}\vec{j}_{mi} \cdot \partial_{i}\vec{h}^{*}]$$

$$+ [-R_{\uparrow}(\mu_{\uparrow}^{*} - \mu_{\downarrow}^{*}) + \mu_{0}\vec{\Omega}_{M} \cdot (\vec{M} \times \vec{H}^{*}) + \mu_{0}\vec{R}_{m} \cdot \vec{h}^{*}].$$
(38)

The bracket associated with the divergence terms contains products of various intensive (i.e., volume-independent) thermodynamic quantities and their respective fluxes. The second bracket contains products of the four to-be-determined thermodynamic fluxes $(j_{si}, j_{\uparrow i}, j_{\downarrow i}, \vec{j}_{mi})$ with the corresponding gradients of their respective intensive thermodynamic quantities $(\partial_i T, \partial_i \mu^*_{\uparrow}, \partial_i \mu^*_{\downarrow}, \partial_i \vec{h}^*)$. The third bracket contains products of the three to-be-determined thermodynamic sources $(R_{\uparrow}, \vec{\Omega}_M, \vec{R}_m)$ with their respective intensive thermodynamic forces $[(\mu^*_{\uparrow} - \mu^*_{\downarrow}), \vec{M} \times \vec{H}^*, \vec{h}^*_{\perp}]$. Because the divergence term can have either sign, we set it to zero, thus yielding $j_{\varepsilon i}$ in terms of the other fluxes, once they are determined.

Each of the seven nondivergence terms in Eq. (38) has a clear physical interpretation as a means to produce entropy: j_{si} to thermal conduction, $j_{\uparrow i}$ and $j_{\downarrow i}$ to (spin-dependent) electrical conduction, \vec{j}_{mi} to diffusion of transverse spin accumulation (excitations), R_{\uparrow} to longitudinal spin flip (from one energy band to another), $\vec{\Omega}_M$ to transverse magnetic damping (of \hat{M}), and \vec{R}_m to decay of transverse spin accumulation \vec{m} .

A. Diagonal terms in the flux-force matrix

We consider first the contributions to the rate of entropy production that are diagonal (d) in what we will call the thermodynamic "flux-force" matrix. (Thus the entropy flux j_{si} is driven only by the temperature gradient.) Recall that \vec{R}_m and \vec{j}_{mi} are all taken to have only the two components transverse to \hat{M} . Then, defining the projector in spin space that is transverse to \hat{M} as $\mathbb{1}_{\perp} \equiv \mathbb{1} - \hat{M}\hat{M}$, we have

$$j_{si}^{(d)} = -\frac{\kappa}{T} \partial_i T, \qquad (39)$$

$$j_{\uparrow i}^{(d)} = -\frac{\sigma_{\uparrow}}{e^2} \partial_i \mu_{\uparrow}^*, \tag{40}$$

$$j_{\downarrow i}^{(d)} = -\frac{\sigma_{\downarrow}}{e^2} \partial_i \mu_{\downarrow}^*, \tag{41}$$

$$\vec{j}_{mi}^{(d)} = L\mathbb{1}_{\perp} \cdot \partial_i \vec{h}^* + L_R \hat{M} \times \partial_i \vec{h}^*, \qquad (42)$$

$$R^{(d)}_{\uparrow} = -G(\mu^*_{\uparrow} - \mu^*_{\downarrow}), \qquad (43)$$

$$\vec{\Omega}_M^{(d)} = \alpha \gamma \mu_0 \hat{M} \times \vec{H}^*, \tag{44}$$

$$\vec{R}_m^{(d)} = K_m \mathbb{1}_\perp \cdot \vec{h}^*, \text{ or } \vec{\Omega}_m^{(d)} = \alpha_m \gamma \mu_0 \hat{M} \times \vec{h}^*.$$
(45)

Many of these Onsager coefficients are already familiar.

- (i) κ is the thermal conductivity.
- (ii) σ_{\uparrow} and σ_{\downarrow} are electrical conductivities.
- (iii) L is a susceptibility times a diffusion coefficient, or

$$L = \chi_f D_f. \tag{46}$$

(iv) L_R , also a susceptibility times a diffusion coefficient, introduces a term that, from its time-reversal signature, is nondissipative.

(v) *G* has units of number density per energy (e.g., $\partial n/\partial \mu$) times the inverse of a longitudinal spin-flip time τ_{sf} , so we write

$$G \equiv \left(\frac{\partial n_{\uparrow}}{\partial \mu_{\uparrow}} + \frac{\partial n_{\downarrow}}{\partial \mu_{\downarrow}}\right) \frac{1}{\tau_{sf}}.$$
(47)

(vi) α is the dimensionless Gilbert constant. Although irreversible thermodynamics inevitably leads to Landau-Lifshitz damping rather than Gilbert damping, we employ α [34] because it is useful as the rotational analog of the inverse Q factor of a linear oscillator.

(vii) α_m also is a dimensionless constant, relating to damping of \vec{m} . It is related to K_m of (45) by

$$K_m \equiv \alpha_m \mu_0 \gamma M. \tag{48}$$

Because K_m is associated with relaxation of \vec{m} , we introduce the relaxation rate τ_f^{-1} via

$$K_m \equiv \frac{\chi_f}{\tau_f}.$$
 (49)

B. Off-diagonal terms in the flux-force matrix

In addition, there are terms that are off-diagonal (*od*) in the thermodynamic forces. They give rise, among other things, to the spin Seebeck effect and the spin Peltier effect. For a non-

spin-orbit-active material and a uniform equilibrium, we have

$$j_{si}^{(od)} = -L_{s\uparrow}\partial_i\mu_{\uparrow}^* - L_{s\downarrow}\partial_i\mu_{\downarrow}^*, \qquad (50)$$

$$j_{\uparrow i}^{(od)} = -L_{\uparrow s} \partial_i T - L_{\uparrow \downarrow} \partial_i \mu_{\downarrow}^*, \tag{51}$$

$$j_{\downarrow i}^{(od)} = -L_{\downarrow s} \partial_i T - L_{\downarrow \uparrow} \partial_i \mu_{\uparrow}^*, \qquad (52)$$

$$\vec{j}_{mi}^{(od)} = 0,$$
 (53)

$$R^{(od)}_{\uparrow} = 0, \tag{54}$$

$$\vec{\Omega}_{M}^{(od)} = \alpha_{Mh} \gamma \mu_0 \hat{M} \times \vec{h}^*, \tag{55}$$

$$\vec{R}_m^{(od)} = K_{mH} \mathbb{1}_\perp \cdot \vec{H}^*, \quad \vec{\Omega}_m^{(od)} = \alpha_{mH} \gamma \mu_0 \hat{M} \times \vec{H}^*.$$
(56)

Many of these Onsager coefficients are already familiar.

(a) $L_{\uparrow s}$ and $L_{\downarrow s}$ imply separate up- and down-spin Seebeck effects, and $L_{s\uparrow}$ and $L_{s\downarrow}$ imply separate up- and down-spin Peltier effects. They all have dimensionality of s⁻¹ m⁻¹ K⁻¹ [diffusion constant/(number density-K)]. The spin Seebeck effect was observed and explained by Uchida *et al.* [35]. The spin Seebeck and spin Peltier effect terms in Eqs. (50)–(52) also appear in Eqs. (A14)–(A16) of Ref. [23].

(b) The cross-conductivity terms $L_{\uparrow\downarrow}$ and $L_{\downarrow\uparrow}$ have dimensionality of conductivity/charge².

(c) α_{Mh} is a dimensionless cross-damping term. A term like (55) has appeared in the spintronics literature [36]; as we show below, Onsager reciprocity on (55) then leads to the cross-damping term in Eq. (56).

(d) α_{mH} is related to α_{Mh} by an Onsager relation (see below).

All of these off-diagonal Onsager terms are dissipative because their time-reversal signatures are opposite those of their primary quantities: entropy flux has odd time-reversal signature, but $j_{si}^{(od)}$ is even under time reversal; $\partial_t \hat{M}$ has even timereversal signature, but $\hat{M} \times \vec{\Omega}_M^{(od)}$ is even under time reversal; etc. The cross-damping coefficients α_{Mh}, α_{mH} introduce two distinct but related relaxation rates, as we show below.

Onsager reciprocity ensures that each pair of even-in-time off-diagonal terms in Eq. (38) has the same coefficient, and thus leads to conditions on the spin Seebeck and spin Peltier coefficients that

$$L_{s\uparrow} = L_{\uparrow s}, \quad L_{s\downarrow} = L_{\downarrow s}, \tag{57}$$

and that

$$L_{\uparrow\downarrow} = L_{\downarrow\uparrow}, \quad K_{mH} \equiv \gamma \mu_0 M \alpha_{mH}, \quad \alpha_{mH} = \alpha_{Mh}.$$
 (58)

Thus there is only one truly distinct damping coefficient, $\alpha_{mH} = \alpha_{Mh}$. The requirement for positivity of entropy production, which leads to non-negativity of $\kappa, \sigma_{\uparrow}, \sigma_{\downarrow}, L, \alpha, \alpha_m, G$, and $L_{\uparrow\downarrow}^2 - L_{\uparrow\uparrow}L_{\downarrow\downarrow}$, includes the important constraint that

$$\alpha_{Mh}^2 \leqslant \alpha \alpha_m. \tag{59}$$

We will see that requiring positive cross-relaxation times further gives $\alpha_{Mh} \leq 0$.

V. FULL EQUATIONS OF MAGNETIC DYNAMICS

It is convenient to define two frequencies:

$$\omega_H \equiv \gamma \mu_0 H, \tag{60}$$

$$\omega_X \equiv \gamma \mu_0 h_{\parallel}^* = \gamma \mu_0 (H + \lambda M) \approx \gamma \mu_0 \lambda M.$$
 (61)

Using Gaussian units, Ament and Rado define five dimensionless quantities that have been employed in interpreting ferromagnetic resonance experiments [37]. Reference [38] converts to SI units in the supplemental material. See our Appendix B.

A. $\partial_t \hat{M}$

 $\partial_t \hat{M}$ follows from (29), with $\vec{\Omega}_M^{(d)}$ from (44) and cross-term term $\vec{\Omega}_M^{(od)}$ from (55),

$$\partial_t \hat{M} = -\gamma \mu_0 \hat{M} \times \vec{H}^* - \alpha \gamma \mu_0 \hat{M} \times (\hat{M} \times \vec{H}^*) - \alpha_{Mh} \gamma \mu_0 \hat{M} \times (\hat{M} \times \vec{h}^*).$$
(62)

Except for the cross-relaxation term involving α_{Mh} , this is basically the Landau-Lifshitz equation.

For purposes of interpretation, following the form employed previously in a related context [39], it is useful to rewrite the above equation. We multiply (62) by M and use $\vec{H}_{\perp}^* = -\chi_{\perp}^{-1}\delta \vec{M}_{\perp}, \vec{h}_{\perp}^* = -\chi_f^{-1}\delta \vec{m}_{\perp}$ and (14) to obtain

$$M\partial_t \hat{M} = \gamma \mu_0 H \hat{M} \times \delta M_\perp - \alpha \gamma \mu_0 H \delta M_\perp - \frac{\alpha_{Mh} \gamma \mu_0 M}{\chi_f} \delta \vec{m}_\perp.$$
(63)

Because an excess $\delta \vec{m}_{\perp}$ should in part decay to \vec{M}_{\perp} , the coefficient of $\delta \vec{m}_{\perp}$ should be positive, meaning that

$$\alpha_{Mh} = \alpha_{mH} < 0. \tag{64}$$

We now define

$$\frac{1}{\tau_{ML}} + \frac{1}{\tau_{Mm}} \equiv \alpha \omega_H, \quad \frac{1}{\tau_{mM}} \equiv -\alpha_{Mh} \gamma \mu_0 \frac{M}{\chi_f}, \qquad (65)$$

where τ_{ML}^{-1} is the decay rate from \vec{M} to the lattice, and τ_{Mm}^{-1} is the decay rate from \vec{M} to \vec{m} ; only the sum is specified at the moment. τ_{mM}^{-1} is the decay rate from \vec{m} to \vec{M} ; (64) implies that $\tau_{mM} > 0$. Then (63) becomes

$$\partial_t \vec{M}_{\perp} = \omega_H \hat{M} \times \delta \vec{M}_{\perp} - \left(\frac{1}{\tau_{ML}} + \frac{1}{\tau_{Mm}}\right) \delta \vec{M}_{\perp} + \frac{1}{\tau_{mM}} \delta \vec{m}_{\perp}.$$
(66)

Physically speaking, \vec{M}_{\perp} decreases by decay both to the lattice (L) and to \vec{m}_{\perp} in proportion to the disequilibrium $\delta \vec{M}_{\perp}$, and it increases by decay from \vec{m}_{\perp} in proportion to the disequilibrium $\delta \vec{m}_{\perp}$. Not only is there a new "source" due to decay from $\delta \vec{m}_{\perp}$, there is also a new "sink" due to decay to $\delta \vec{m}_{\perp}$; the latter requires a reinterpretation of the Gilbert parameter α as including decay to the lattice and to \vec{m}_{\perp} [40,41]. In the context of magnetic alloys, such cross relaxation has been known for many years [24].

B. $\partial_t \vec{m}_{\perp}$

 $\partial_t \vec{m}_{\perp}$ follows from (30), with $\vec{R}_m^{(d)}$ from (45) and cross term $\vec{R}_m^{(od)}$ from (56). With \vec{h}_{\perp}^* from (19), we find

$$\partial_t \vec{m}_\perp = -\gamma \vec{m}_\perp \times \mu_0 \hat{M} h_\parallel^* - \chi_f D_f \nabla^2 \vec{h}_\perp^* - L_R \hat{M} \times \nabla^2 \vec{h}_\perp^* + K_m \vec{h}_\perp^* + \alpha_{mH} \gamma \mu_0 M \vec{H}_\perp^*.$$
(67)

We now use
$$\vec{H}_{\perp}^* = -\chi_{\perp}^{-1}\delta\vec{M}_{\perp}, \vec{h}_{\perp}^* = -\chi_f^{-1}\delta\vec{m}_{\perp}$$
. Then,
 $\partial_t \vec{m}_{\perp} = -\gamma \mu_0 h_{\parallel}^* \vec{m}_{\perp} \times \hat{M} + D_f \nabla^2 \delta\vec{m}_{\perp} + L_R \hat{M} \times \nabla^2 \vec{h}_{\perp}^*$
 $- (K_m/\chi_f)\delta\vec{m}_{\perp} - \alpha_{mH}\gamma \mu_0 H\delta\vec{M}_{\perp}^*$. (68)

To put this in more familiar form we define

$$\frac{1}{\tau_{mL}} + \frac{1}{\tau_{mM}} \equiv \frac{1}{\tau_f} \equiv \frac{K_m}{\chi_f},$$
$$\frac{1}{\tau_{Mm}} \equiv -\alpha_{mH}\gamma\mu_0 H = -\alpha_{mH}\omega_H, \qquad (69)$$

where τ_{mL} is the decay rate from \vec{m} to the lattice, τ_{mM} of (65) is the decay rate from \vec{M} to \vec{m} , and τ_{Mm} of (69) is the decay rate from \vec{m} to \vec{M} ; (64) implies that $\tau_{Mm} > 0$.

By (61), (65), and (69), and the Onsager relation $\alpha_{mH} = \alpha_{Mh}$, we find that τ_{Mm} and τ_{mM} are related by

$$\frac{\chi_{\perp}}{\tau_{Mm}} = \frac{\chi_f}{\tau_{mM}}.$$
(70)

Then (68) becomes, on using (61) for ω_X ,

 $\partial_t \vec{m}$

Except for the distinct cross-relaxation term, this equation for $\partial_t \vec{m}$ is basically the Bloch equation with diffusion [25,42,43]. In this form, the physical interpretation is that \vec{m}_{\perp} decreases by decay both to the lattice (*L*) and to \vec{M} in proportion to the disequilibrium $\delta \vec{m}_{\perp}$, and it increases by decay from \vec{M} in proportion to the disequilibrium $\delta \vec{M}$.

Equations (66) and (71) may be compared to Eq. (1) of Ref. [39] when the two types of spin have equal γ 's. For comparison, we rewrite τ_{ML}^{-1} of (65) and τ_{mL}^{-1} of (69) and τ_{mM} . Using (64) and (48), we find that

$$\frac{1}{\tau_{ML}} = \omega_H(\alpha - |\alpha_{Mh}|), \quad \frac{1}{\tau_{mL}} = \gamma \mu_0 \frac{M}{\chi_f} (\alpha_m - |\alpha_{Mh}|).$$
(72)

Clearly $|\alpha_{Mh}| \leq \alpha, \alpha_m$, and this implies (59).

C. $\partial_t M$

The longitudinal spin accumulation satisfies (37) with j_{Mi} given by (35), (40), and (41), and R_M given by (36), (43), and (47). We find that

$$\partial_{t}M + (\gamma\hbar/2)(\sigma_{\uparrow}\nabla^{2}\mu_{\uparrow}^{*} + \sigma_{\downarrow}\nabla^{2}\mu_{\downarrow}^{*}) = \gamma\hbar\left(\frac{\partial n_{\uparrow}}{\partial\mu_{\uparrow}} + \frac{\partial n_{\downarrow}}{\partial\mu_{\downarrow}}\right)\frac{1}{\tau_{sf}}(\mu_{\uparrow}^{*} - \mu_{\downarrow}^{*}), \qquad (73)$$

so M decays both by spin flip and by spin diffusion.

VI. TRANSVERSE RESPONSE

The transverse normal modes couple \vec{M}_{\perp} , \vec{m}_{\perp} , and \vec{H}_{\perp} . We assume a dependence $e^{i(\omega t \pm kz)}$, corresponding to leftward (L) and rightward (R) waves. For simplicity, we set $L_R = 0$.

A. $\partial_t \vec{M}_\perp$

In
$$\partial_t M_{\perp}$$
, on substituting for δM_{\perp} and $\delta \vec{m}_{\perp}$ in terms of M_{\perp} , \vec{m}_{\perp} , and H_{\perp} , and using (70), (66) becomes, with k_A from (16)

$$i\omega\vec{M}_{\perp} = \left\{\omega_{H}\hat{M}\times\vec{M}_{\perp}\left(1+\frac{k^{2}}{k_{A}^{2}}\right) - \vec{M}_{\perp}\left[\left(1+\frac{k^{2}}{k_{A}^{2}}\right)\left(\frac{1}{\tau_{ML}}+\frac{1}{\tau_{Mm}}\right) + \frac{\lambda\chi_{f}}{\tau_{mM}}\right]\right\} + \left\{-\omega_{H}\chi_{\perp}\hat{M}\times\vec{H}_{\perp} + \frac{1}{\tau_{ML}}\chi_{\perp}\vec{H}_{\perp}\right\} + \left\{-\omega_{H}\chi_{\perp}\hat{M}\times\lambda\vec{m}_{\perp} + \vec{m}_{\perp}\left[\lambda\chi_{\perp}\left(\frac{1}{\tau_{ML}}+\frac{1}{\tau_{Mm}}\right) + \frac{1}{\tau_{mM}}\right]\right\}.$$

$$(74)$$

B. $\partial_t \vec{m}_{\perp}$

In $\partial_t \vec{m}_{\perp}$, on substituting for $\delta \vec{M}_{\perp}$ and $\delta \vec{m}_{\perp}$ in terms of \vec{M}_{\perp} , \vec{m}_{\perp} , and \vec{H}_{\perp} and using (70) and (71) becomes

$$\begin{aligned}
\omega \vec{m}_{\perp} &= \left\{ \omega_X \hat{M} \times \vec{m}_{\perp} - \vec{m}_{\perp} \left[Dk^2 + \left(\frac{1}{\tau_{mL}} + \frac{1}{\tau_{mM}} \right) + \frac{\lambda \chi_{\perp}}{\tau_{Mm}} \right] \right\} + \left\{ \vec{H}_{\perp} \chi_f \left(Dk^2 + \frac{1}{\tau_{mL}} \right) \right\} \\
&+ \left\{ \vec{M}_{\perp} \left[\lambda \chi_f \left(Dk^2 + \frac{1}{\tau_{mL}} + \frac{1}{\tau_{mM}} \right) + \frac{1}{\tau_{Mm}} \left(1 + \frac{k^2}{k_A^2} \right) \right] \right\}.
\end{aligned}$$
(75)

Recall that (75) omits the L_R term from (71).

C. \vec{E} and \vec{H}

 \vec{M} and \vec{m} couple to \vec{H} , and \vec{H} couples to \vec{E} . Therefore, a complete discussion should include the appropriate equations for \vec{E} and \vec{H} .

Vacuum H response. Maxwell's equations in a conductor are

$$\vec{\nabla} \times \vec{H} = \epsilon_0 \frac{\partial \vec{E}}{\partial t}, \quad \vec{\nabla} \times \vec{E} = -\mu_0 \frac{\partial \vec{H}}{\partial t}.$$
 (76)

From these, we can show that in vacuum

$$\vec{E} = \mp \mu_0 c\hat{z} \times \vec{H}.$$
(77)

Conductor H response. Maxwell's equations in a conductor with conductivity σ are

$$\vec{\nabla} \times \vec{H} = \vec{j}_{\text{free}} + \frac{\partial \vec{D}}{\partial t} \approx \sigma \vec{E},$$

$$\vec{\nabla} \times \vec{E} = -\mu_0 \frac{\partial}{\partial t} (\vec{H} + \vec{M} + \vec{m}).$$
(78)

In writing this, we take the ordinary electric current due to free carrier $\vec{j}_{\text{free}} = \sigma \vec{E}$, which thus omits complications associated with up- and down-spin conductivities. Using \pm for R and L waves, by $\vec{\nabla} \times \vec{H} \approx \sigma \vec{E}$ we have, for all three modes,

$$\vec{E} = \pm \frac{ik}{\sigma} \hat{z} \times \vec{H}.$$
(79)

 \vec{E} can reflect the presence of the *M* and *m* variables in their contributions to \vec{H} . Taking the curl of (78), for $\vec{\nabla} \cdot \vec{H} = 0$, leads to

$$-\nabla^2 \vec{H} = -\mu_0 \sigma \frac{\partial}{\partial t} (\vec{H} + \vec{M} + \vec{m}).$$
(80)

With an $e^{i\omega t \pm kz}$ dependence, this leads to

$$(k^2 + i\mu_0\sigma\omega)\vec{H} = -i\mu_0\sigma\omega(\vec{M} + \vec{m}).$$
(81)

D. Decoupled transverse modes

Consider a film geometry where the applied field and the equilibrium point normal to the plane, with $\hat{M} = \hat{z}$. Then,

with $M_+ \equiv M_x + iM_y$, it follows that $(\hat{M} \times \hat{M})_+ = iM_+$, and similarly for the other vectors in the problem. Because the demagnetization field for this geometry is isotropic in the plane, these variables are useful for solving the above equations.

(i) When \vec{H} and \vec{m} are neglected, (74) leads to a normal mode of \vec{M} with wave vector k_M satisfying

$$i\omega = i\omega_H \left(1 + \frac{k^2}{k_A^2} \right) - \left[\left(1 + \frac{k^2}{k_A^2} \right) \left(\frac{1}{\tau_{ML}} + \frac{1}{\tau_{Mm}} \right) + \frac{\lambda \chi_f}{\tau_{mM}} \right].$$
(82)

Solving for k_M^2 then yields the zeroth-order result,

$$\frac{k_M^2}{k_A^2} = \frac{\omega - \omega_H - i\left(\frac{1}{\tau_{ML}} + \frac{1}{\tau_{Mm}} + \frac{\lambda\chi_f}{\tau_{mM}}\right)}{\omega_H + i\left(\frac{1}{\tau_{ML}} + \frac{1}{\tau_{Mm}}\right)}.$$
(83)

(ii) When \vec{H} and \vec{M} are neglected, (75) leads to a normal mode of \vec{m} with wave vector k_m satisfying

$$i\omega = i\omega_X - \left[D_f k^2 + \frac{1}{\tau_{mL}} + \frac{1}{\tau_{mM}} + \frac{\lambda\chi_\perp}{\tau_{Mm}}\right].$$
 (84)

With

$$\tau_D^{-1} \equiv D_f k_A^2, \tag{85}$$

solving for k_m^2 then yields the zeroth-order result,

$$\frac{k_m^2}{k_A^2} = \tau_D \bigg[-i(\omega - \omega_X) - \bigg(\frac{1}{\tau_{mL}} + \frac{1}{\tau_{mM}} + \frac{\lambda \chi_\perp}{\tau_{Mm}} \bigg) \bigg]. \quad (86)$$

When L_R is included, (85) for τ_D^{-1} should have $D_f \to D_f + iL_R/\chi_f$.

(iii) When \vec{M} and \vec{m} are neglected, (81) leads to a normal mode of \vec{H} , with

$$k_H^2 = -i\mu_0\sigma\omega$$
 (conductor) (87)

corresponding to the usual skin depth. For an insulating magnet, such as yttrium iron garnet (YIG), with index of

refraction *n* (*c* is the speed of light),

$$k_H^2 = \frac{n\omega^2}{c^2} \quad \text{(insulator).} \tag{88}$$

E. On resonances

Near the usual resonance $\omega \approx \omega_H$, the damping terms dominate \vec{M}_+ . On neglecting m_+ , the leading term in \vec{H}_{\perp} in Eq. (74) gives $M_+/H_+ \approx -i\chi_\perp/\alpha$, which is of the order of 10^3 .

Near the high-frequency resonance, $\omega \approx \omega_X$, where the damping terms dominate \vec{m}_+ . On neglecting M_+ , the leading term in \vec{H}_{\perp} in Eq. (75) gives $m_{\perp}/H_{\perp} \approx \chi_f$, which is of the order of 10^{-5} . Moreover, for this mode, $M_+ + m_+ \approx 0$. Therefore, although in principle there is a resonance of m_+ at ω_X , even if it were possible to drive m_+ without driving $M_+ \approx$ $-m_+$, this mode seems unlikely to be detectable without great effort.

Similarly, the s-d model of Hasegawa [24] for Mn in Cu, where $\gamma_{Mn} \approx \gamma_{Cu}$, has two resonances, and the high-frequency exchange-driven resonance has $(\dot{M}_s + \dot{M}_d)_{\perp} = 0$. On the other hand, in a study of Er (d) in Ag (s), where $\gamma_{\rm Er} \approx 6.8$ and $\gamma_{Ag} \approx 2.0$ –2.6, the theory gives two resonances, both with $\vec{M}_s + \vec{M}_d \neq 0$ and both observed [39].

Note the large number of materials- and experiment-related parameters that appear in the bulk theory:

(a) the frequencies ω_H and ω_X ; (b) the rates τ_{ML}^{-1} , τ_{mL}^{-1} , τ_{Mm}^{-1} , and τ_D^{-1} [τ_{mM}^{-1} and τ_{Mm}^{-1} are related by (70)];

(c) the dimensionless parameters λ , χ_f , and χ_{\perp} ; only *H* and ω can be controlled by the experimentalist;

(d) L_R .

VII. SURFACE EFFECTS AND BOUNDARY CONDITIONS

This section applies irreversible thermodynamics to obtain the boundary conditions on \vec{m} in the absence of spin-orbit scattering. It also discusses the boundary conditions on \hat{M} , which follow from micromagnetics in the continuum limit. The boundary conditions on \vec{E} and \vec{H} are, as usual, that their components transverse to the normal are continuous.

A. Boundary conditions from irreversible thermodynamics

For simplicity, we have not included spin-orbit effects in bulk, so that the anomalous Hall effect (AHE) and anisotropic magnetoresistance (AMR) are not included [44]. Correspondingly, in the present section we neglect spin-orbit scattering at surfaces and interfaces [45]. For simplicity, as boundary conditions we take the continuity of various fluxes across an interface,

$$j_{si}, j_{\uparrow i}, j_{\downarrow i}, \bar{j}_{mi}$$
 (continuity across interface). (89)

Other than the truly conserved number flux $j_i = j_{\uparrow i} + j_{\downarrow i}$, the fluxes j_{si} , j_{mi} , and $j_{Mi} = -(\gamma \hbar/2)(j_{\uparrow i} - j_{\downarrow i})$ are due to the excitations of the adjacent systems.

Beyond continuity, in irreversible thermodynamics a full statement of the boundary conditions relates the fluxes across the interfaces to differences in the various intensive thermodynamic quantities across the interface. Typically, the fluxes are proportional to the difference across the interface (such as ΔT) of various intensive quantities. We now derive these boundary conditions using irreversible thermodynamics.

Consider Eq. (38) for TR_s , which holds for each side of an interface. Integrate it over an atomic distance across the interface between the two materials. Because $j_{\varepsilon i}$ is chosen to make the term in parentheses zero, this term will not contribute. If we assume that the terms R_{\uparrow} , Ω_M , and R_m in the last (third) bracket remain finite at the interface, then for these terms integration over an atomic distance gives a negligible contribution. However, the terms from the middle bracket, involving products of (continuous fluxes) and gradients of intensive thermodynamic quantities, do contribute.

From the discussion above, integration of Eq. (38) across the interface, along surface normal \hat{N} , then gives, for the surface rate of entropy production,

$$0 \leqslant T\mathcal{R}_s = \hat{N}_i [-j_{si}\Delta T - j_{\uparrow i}\Delta\mu^*_{\uparrow} - j_{\downarrow i}\Delta\mu^*_{\downarrow} + \vec{j}_{mi}\cdot\Delta\vec{h}^*].$$
(90)

The (assumed) continuous fluxes across the interface satisfy, with surface transport coefficients defined by analogy to their bulk symbols (and not retaining all the possible off-diagonal terms),

$$\hat{N}_i j_{si} = -\frac{\mathcal{K}}{T} \Delta T, \qquad (91)$$

$$\hat{N}_i j_{\uparrow i} = -\frac{\Sigma_{\uparrow\uparrow}}{e^2} \Delta \mu_{\uparrow}^* - \frac{\Sigma_{\uparrow\downarrow}}{e^2} \Delta \mu_{\downarrow}^*, \qquad (92)$$

$$\hat{N}_i j_{\downarrow i} = -\frac{\Sigma_{\downarrow\downarrow}}{e^2} \Delta \mu_{\downarrow}^* - \frac{\Sigma_{\downarrow\uparrow}}{e^2} \Delta \mu_{\uparrow}^*, \qquad (93)$$

$$\hat{N}_i \vec{j}_{mi} = \mathcal{L} \mathbb{1}_\perp \cdot \Delta \vec{h}^* + \mathcal{L}_R \hat{M} \times \Delta \vec{h}^*.$$
(94)

For the above equations,

(i) \mathcal{K} has units of thermal conductivity.

(ii) The Σ 's have units of electrical conductivity per unit length. Here we keep the off-diagonal conductivities in order to represent the effect of scattering at the interface.

(iii) \mathcal{L} and \mathcal{L}_R have units of susceptibility times velocity.

We note two works giving related longitudinal spin accumulation results [46.47].

With the above equations for the fluxes crossing the surface, (90) becomes

$$0 \leqslant T\mathcal{R}_{s} = \frac{\mathcal{K}}{T} (\Delta T)^{2} + \frac{\Sigma_{\uparrow\uparrow}}{e^{2}} (\Delta \mu_{\uparrow}^{*})^{2} + \frac{\Sigma_{\downarrow\downarrow}}{e^{2}} (\Delta \mu_{\downarrow}^{*})^{2} + \frac{\Sigma_{\uparrow\downarrow} + \Sigma_{\downarrow\uparrow}}{e^{2}} (\Delta \mu_{\uparrow}^{*}) (\Delta \mu_{\downarrow}^{*}) + \mathcal{L} (\Delta \vec{h}^{*})^{2}.$$
(95)

By the Onsager condition that cross terms give the same rate of entropy production, $\Sigma_{\uparrow\downarrow} = \Sigma_{\downarrow\uparrow}$. Requiring $\mathcal{R}_s \ge 0$ for arbitrary values of $\Delta \mu^*_{\uparrow}$ and $\Delta \mu^*_{\downarrow}$ yields $\Sigma_{\uparrow\uparrow} \Sigma_{\downarrow\downarrow} \ge (\Sigma_{\downarrow\uparrow})^2$.

The boundary conditions on the number current $j_i =$ $j_{\uparrow i} + j_{\downarrow i}$ and the longitudinal magnetization current $j_{Mi} =$ $-(\gamma \hbar/2)(j_{\uparrow i} - j_{\downarrow i})$ may be obtained by combining (92) and (93). At a vacuum interface, in the absence of surface spin-flip scattering, $j_{mi} \sim \partial_i \vec{m} = 0$. Reference [14] employed $M\delta \hat{M}$ for \vec{m} , and thus took $\partial_{\vec{n}} \hat{M} = 0$ at a vacuum interface [48]. The next section shows that \hat{M} satisfies a boundary condition determined by micromagnetics.

The term $\mathcal{L}_R \hat{M} \times \Delta \vec{h}^*$ of (94) in $\hat{N}_i \vec{j}_{mi}$ is even under time reversal, and thus is not dissipative. Mathematically, it gives zero contribution to R_s because it is normal to $\Delta \vec{h}^*$.

B. Boundary condition on \hat{M}

The boundary conditions on \hat{M} are obtained from the equation of motion for \hat{M} evaluated at each surface. We first consider the case where, if anisotropy is present, it is the same in bulk and at the surface. (This ensures that in the absence of any degree of freedom other than \vec{M} , Landau-Lifshitz dynamics holds.) Neglecting damping at the surface, and in the continuum limit, if across the interface is a neighboring magnetization $\vec{M'}$, then

$$\partial_t \hat{M} = -\gamma \,\mu_0 \hat{M} [\vec{H} + \lambda \vec{m} + J_S \hat{M}' + A a^{-1} (\hat{N} \cdot \vec{\nabla} \hat{M})].$$
(96)

Here, J_S is the interface coupling (units of A/m), *a* is an atomic length, and *A* is the nonuniform exchange constant (units of A m).

Linearization of (96) gives the form one would expect in the long-wavelength limit for a magnetics calculation,

$$\partial_t \hat{M} = -\gamma \mu_0 \delta \hat{M} \times (\vec{H}_0 + J_S \hat{M}') -\gamma \mu_0 \hat{M} \times (\vec{H}_{rf} + \lambda \vec{m} + J_S \delta \hat{M}') -\gamma \mu_0 \hat{M} \times [Aa^{-1}(\hat{N} \cdot \vec{\nabla} \delta \hat{M})].$$
(97)

We can make this look like the bulk equation for \hat{M} if we impose the boundary condition

$$-A\hat{M} \times \nabla^2 \delta \hat{M} = -Aa^{-1}\hat{M} \times \hat{N} \cdot \bar{\nabla} \delta \hat{M} -J_S(\delta \hat{M} \times \hat{M}' + \hat{M} \times \delta \hat{M}').$$
(98)

At a vacuum interface, $J_S = 0$; in that case, (98) becomes

$$-A\hat{M} \times \nabla^2 \delta \hat{M} = -Aa^{-1}\hat{M} \times \hat{N} \cdot \vec{\nabla} \delta \hat{M}, \qquad (99)$$

which relates, at the interface, the first and second spatial derivatives of \hat{M} .

However, as noted by Kittel, surface anisotropy can have a significant effect on the boundary conditions, causing pinning [49–51]. Consider uniaxial surface anisotropy K_s (units of J/m²) that favors pinning of \vec{M} along or against the surface normal. Neglecting the second derivative term employed in the free boundary condition (99), one has

$$A\hat{N}\cdot\vec{\nabla}\hat{M}-K_s\delta\hat{M}=\vec{0}.$$
 (100)

The results of the present section, for the M mode and the m mode, when appended by the H mode, enable one to solve very general boundary conditions problems, including spin transfer torque and spin pumping at any frequency for which irreversible thermodynamics is valid.

VIII. HEAT PRODUCTION AND ENERGY ABSORPTION

We now derive two important relations associated with heat flow and heat production. The first shows the relatively well-known result that temperature gradients alone do not contribute to the heat production, although they *are* associated with entropy production. Although we do not show it, the off-diagonal terms in the heat flux can lead to heating or cooling due to either electric currents (Seebeck effect) or spin currents (spin Seebeck effect). The second relation shows that, on average, rf energy absorbed in a volume goes entirely into heating that volume and/or to heat flow in or out of that volume.

A. Heat production

Defining

$$dq \equiv Tds, \quad j_i^q \equiv Tj_{si}, \tag{101}$$

we deduce from (26) that

$$\partial_t q = T \partial_t s = -T \partial_i j_{si} + T R_s. \tag{102}$$

Thus,

$$\partial_t q + \partial_i j_i^q = j_{si} \partial_i T + T R_s. \tag{103}$$

If there is only a thermal gradient, so that $TR_s = -j_{si}\partial_i T$ by (38), then the right-hand side of the above equation is zero, and the "heat" is conserved—as expected, heat simply flows from hot to cold, without being produced or absorbed.

B. Time-averaged energy absorption

With dots referring to rate of changes of the nonentropic part of the internal energy density, the energy conservation equation (25) can be rewritten using (102) as

$$\partial_t \varepsilon = -\partial_i j_i^\varepsilon$$

= $(T \partial_t s + \cdots) - \vec{H} \cdot (\partial_t \vec{m} + \partial_t \vec{M})$
= $\partial_t q + (\cdots) - \vec{H} \cdot (\partial_t \vec{m} + \partial_t \vec{M}).$ (104)

For an oscillating rf field, the time average of the term in dots (the nonentropic part of the internal energy density) is zero. Then,

$$\overline{\partial_t q} = -\overline{\partial_i j_i^\varepsilon} + \overline{\vec{H} \cdot (\partial_t \vec{m} + \partial_t \vec{M})}.$$
 (105)

This immediately leads to a physical interpretation. $\overline{\partial_t q}$ is the average volume rate of heating, and $\overline{\vec{H}} \cdot (\partial_t \vec{m} + \partial_t \vec{M})$ is the rate of absorption of energy from the oscillating field (rf heating). Without energy flow, these two are equal. With energy flow, the rate of heating equals the rf heating less any net energy flow from the volume in question, as can occur when a charge current or spin current crosses the interface. For a related discussion, see Ref. [52].

IX. ON RECIPROCITY OF SPIN TRANSFER TORQUE AND SPIN PUMPING

Section I discussed the issue of reciprocity of spin transfer torque and spin pumping, but some of it bears repeating with some context. For two paramagnets, spin pumping—without the current spintronics terminology [6,7]—was observed in 1976 and explained using irreversible thermodynamics [14]. This important but neglected work studied Li on Cu, where there are slightly different but distinguishable g factors; at fixed Cu thickness, a thin layer of Li gave a single common resonance line, whereas a thicker layer of Li gave two resonance lines. Attributing only a single magnetization to each material, Eq. (1) of Ref. [14] had the spin flux across the interface driven by the difference in \vec{M}/χ across the interface. For thin Li on Cu, the electron spin resonance in Cu "pumped" a spin current into Li, and thus there was a spin transfer torque from Cu to Li.

As noted in Sec. I, for a ferromagnet *F* against a paramagnet *N*, in 1979 spin pumping was observed and explained using a more phenomenological form of irreversible thermodynamics than in the present work [1]. The authors attributed to *F* the equivalent of \hat{M} and \vec{m} , with the ferromagnetic spin resonance driving \hat{M} in bulk *F*; then they used the irreversible thermodynamics boundary conditions, where the tipped \hat{M} causes a nonzero \vec{h}_{\perp}^* (or, equivalently, $\vec{\mu}_{s\perp}$) to drive a spin flux across the interface to *N*, possessing only an \vec{m} . (This mechanism is independent of frequency, as opposed to the mechanism of Ref. [6], where the driving term $d\hat{M}/dt$ is proportional to the resonance frequency.)

One might expect spin pumping (SP) and spin transfer torque (STT) to be reciprocal phenomena in the Onsager sense, just as the Seebeck effect and the Peltier effect have related transport coefficients. Indeed, for a single magnetic conductor with a nonuniform texture, there are such Onsager relationships between SP and STT, and they are local [23]. (Consideration of time-reversal properties further shows that the so-called adiabatic STT and SP are dissipative and the so-called nonadiabatic STT and SP are nondissipative [23].) However, for an $F_1/N/F_2$ system with only F_1 driven into resonance, the two effects occur at different places, with spin pumping at F_1/N and spin transfer torque at N/F_2 . Therefore, *spin transfer torque and spin pumping satisfy a nonlocal relationship*, as shown explicitly in Sec. I.

X. SUMMARY AND CONCLUSIONS

The present work has given the foundations for an irreversible thermodynamics approach to spin pumping and spin transfer torque in the framework of what we have called the *m*-*M* model. Although it is closely related to the *s*-*d* models of Refs. [1,5], the *m*-*M* model is distinguishable from these *s*-*d* models in part because $\vec{m} = \vec{0}$ in equilibrium. This work also finds that in bulk, there are previously neglected relaxation times associated with

cross relaxation between M and m, that there is a new precession term for \vec{m} in nonuniform systems, and that at surfaces there are distinct boundary conditions for M and m. A qualitative analysis has been given of spin pumping and spin transfer torque for an $F_1/N/F_2$ system: the spin pumped M in the F_1 drives a spin current across the F_1/N interface, and a spin current in the N drives an M on the F_2 side of the N/F_2 interface.

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APPENDIX A: IRREVERSIBLE THERMODYNAMICS OF MAGNETIC INSULATORS

The irreversible thermodynamics of magnetic insulators is similar to but distinct from that for magnetic conductors. For magnetic insulators, we consider that spin waves, as in the Heisenberg model, can be out of equilibrium and therefore produce a spin accumulation \vec{m} . We therefore treat this electric current-free case separately.

The thermodynamic differential is taken to be

$$d\varepsilon = Tds - \mu_0 \vec{H}^* \cdot (Md\hat{M}) - \mu_0 \vec{H}^* \cdot \hat{M}dM - \mu_0 \vec{h}^* \cdot d\vec{m}.$$
(A1)

For \vec{H}^* and \vec{h}^* , we continue to employ (13) and (19). Likewise, for the equations of motion, for ε and s we continue to employ (25) and (26). For \hat{M} and M, we continue to employ (29) and (37), although in the latter we dispense with the band-related definitions of j_{Mi} and R_M . We use (30) for \vec{m} , but we combine (30) and (31) to yield

$$\partial_t \vec{m} + \partial_i \vec{j}_{mi} = -\vec{m} \times (\mu_0 \gamma \vec{h}^* + \vec{\Omega}_m).$$
(A2)

The approach of the text then yields

$$0 \leqslant TR_{s} = T(\partial_{t}s + \partial_{i}j_{si})$$

$$= -\partial_{i}[j_{\varepsilon i} - Tj_{si} + \mu_{0}\vec{H}^{*} \cdot \hat{M}j_{Mi} + \mu_{0}\vec{h}^{*} \cdot \vec{j}_{mi}] + [-j_{si}\partial_{i}T + \mu_{0}j_{Mi} \cdot \partial_{i}(\vec{H}^{*} \cdot \hat{M}) + \mu_{0}\vec{j}_{mi} \cdot \partial_{i}\vec{h}^{*}]$$

$$+ [\mu_{0}\vec{\Omega}_{M} \cdot (\vec{M} \times \vec{H}^{*}) + \mu_{0}\vec{\Omega}_{m} \cdot (\vec{M} \times \vec{h}^{*}) + \mu_{0}R_{M}\vec{H}^{*} \cdot \hat{M}].$$
(A3)

The diagonal terms in the thermodynamic "flux-force" matrix are

 $\vec{j}_{mi}^{(d)} = L\mathbb{1}_{\perp} \cdot \partial_i \vec{h}^* + L_R \hat{M} \times \partial_i \vec{h}^*,$

 $j_{si}^{(d)} = -\frac{\kappa}{T}\partial_i T,$

 $j_{Mi}^{(d)} = L_M \partial_i (\vec{H}^* \cdot \hat{M}),$

 $R_M^{(d)} = \alpha_M \gamma \mu_0 \vec{H}^* \cdot \hat{M},$

 $\vec{\Omega}_{M}^{(d)} = \alpha \gamma \mu_{0}(\vec{M} \times \vec{H}^{*}),$

 $\vec{\Omega}_m^{(d)} = \alpha_m \gamma \,\mu_0(\vec{M} \times \vec{h}^*).$

All of these are dissipative. The *L*'s have units of susceptibility times diffusion constant. The α 's are all dimensionless.

The off-diagonal terms (again, all dissipative) are

$$\dot{j}_{si}^{(0d)} = L_{sM} \partial_i (H^* \cdot \hat{M}), \tag{A10}$$

$$j_{Mi}^{(od)} = L_{Ms}\partial_i T, \tag{A11}$$

$$\vec{j}_{mi}^{(od)} = 0, \tag{A12}$$

$$R_M^{(od)} = 0, \tag{A13}$$

$$\vec{\Omega}_M^{(od)} = \alpha_{Mh} \gamma \mu_0 (\vec{M} \times \vec{h}^*), \qquad (A14)$$

$$\vec{\Omega}_m^{(od)} = \alpha_{mH} \gamma \mu_0 (\vec{M} \times \vec{H}^*).$$
(A15)

(A4)

(A5)

(A6)

(A7)

(A8)

(A9)

There are no separate up- and down-spin Seebeck/spin Peltier effects (described by the coefficients L_{sM} and L_{Ms}). The off-diagonal Onsager coefficients satisfy

$$L_{sM} = L_{Ms}, \quad \alpha_{Mh} = \alpha_{mH}. \tag{A16}$$

APPENDIX B: ON NOTATION

In an early work, Ament and Rado (AM) established a dimensionless notation in Gaussian units that is often followed [37]. More recently, Li and Bailey (LB) have converted this to SI units [38]. Below we compare their definitions and ours (S). Reference [37] employs λ for the dimensional Landau-Lifshitz damping constant, whereas we employ λ_{LL} , with λ reserved for the dimensionless mean-field exchange constant (which determines how effectively the tipped \hat{M} at the F/N interface drives a spin current). Normally, δ denotes the skin depth, but LB employ δ_0 .

Notation of Ament-Rado (Gaussian).

$$\eta = \frac{H_z}{4\pi M_s} \rightarrow \eta = \frac{H}{M} = \frac{1}{\chi_{\perp}} \cdot (AM \rightarrow S), \quad (B1)$$

$$\Omega = \frac{\omega}{4\pi M_s \gamma} \to \Omega = \frac{\omega}{\omega_M}, \quad \omega_M \equiv \gamma \mu_0 M_s \cdot (AM \to LB),$$

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$$L = \frac{\lambda_{LL}}{M_s \gamma} \to \alpha = \frac{\lambda_{LL}}{\gamma M_s} \cdot (AM \to S), \qquad (B3)$$

$$\epsilon \delta = \sqrt{\frac{A}{2\pi M_s^2}} \to \epsilon \delta = \sqrt{\frac{2A}{\mu_0 M_s^2}} = \delta_{ex} \cdot (AM \to LB), (B4)$$

$$K = k\epsilon\delta \to \kappa = k\delta_{ex} \cdot \text{ (AM} \to \text{LB)}. \tag{B5}$$

Notation of Li-Bailey.

$$\delta_{ex} = \sqrt{\frac{2A}{\mu_0 M_s^2}}, \quad \delta_0 = \sqrt{\frac{2}{\mu_0 \sigma \omega}} (= \delta).$$
 (B6)

$$\kappa = k\delta_{ex}, \quad \epsilon = \frac{\delta_{ex}}{\delta_0}, \quad \Omega = \frac{\omega}{\omega_M}, \quad \omega_M \equiv \gamma \mu_0 M_s.$$
 (B7)

Notation of present work.

(B2)

$$k_H^2 = -i\mu_0\sigma\omega = -\frac{2i}{\delta^2}, \quad \chi_\perp = \frac{M}{H}.$$
 (B8)

$$k_A^2 = \frac{\mu_0 M H}{2A} = \frac{H}{M} \frac{\mu_0 M^2}{2A} = \frac{1}{\chi_\perp \delta_{ex}^2}.$$
 (B9)

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