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Topological Hall effect in diffusive ferromagnetic thin films with spin-flip scattering

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We study the topological Hall (TH) effect in a diffusive ferromagnetic metal thin film by solving a Boltzmann transport equation in the presence of spin-flip scattering. A generalized spin diffusion equation is derived, which contains an additional source term associated with the gradient of the emergent magnetic field that arises from skyrmions. Because of the source term, spin accumulation may build up in the vicinity of the skyrmions. This gives rise to a spin-polarized diffusion current that in general suppresses the bulk TH current. Only when the spin diffusion length is much smaller than the skyrmion size does the TH resistivity approach the value derived by Bruno *et al.* [Phys. Rev. Lett. **93**, 096806 (2004)]. We derive a general expression of the TH resistivity that applies to thin-film geometries with spin-flip scattering, and show that the corrections to the TH resistivity become large when the size of room temperature skyrmions is further reduced to tens of nanometers.

I. INTRODUCTION

In the last few decades, the notion of topological order has led to several major breakthroughs in condensed matter physics. Along with the discovery of momentum space topology that unveils a new phase of matter – topological insulators^{1,2} – the spotlight has also been on magnetic skyrmions^{3,4}, which are spin textures possessing nontrivial real-space topology. Each magnetic skyrmion carries an integer topological charge protected by a finite energy barrier⁵. The topological attributes endow skyrmions with substantial robustness against boundaries and disorders during their current-induced motion, which makes them promising candidates for electronic applications^{4,6,7}.

After initially being identified in bulk chiral magnets^{8,9}, intensive efforts have been devoted to creating and manipulating nanoscale skyrmions at room temperature in magnetic thin films and multilayer structures^{10,11} that are more suitable for practical applications. A central issue in any application is a scheme to electrically detect magnetic skyrmions. In thin films and multilayers this is usually based on the topological Hall (TH) effect¹². This effect arises from the Berry phase acquired by conduction electrons when they traverse skyrmion textures and is therefore associated with topological charges carried by the skyrmions. The TH effect was first observed in bulk non-centrosymmetric magnetic materials such as MnSi^{13,14}, MnGe¹⁵ and *etc.*, following the discovery of the skyrmion lattice (SkX) phase in these materials. Later, TH measurements were also carried out in chiral magnet thin films with more stable SkX phase^{16,17} than bulk systems. Recently, a discretized TH effect induced by discontinuous motion, the creation and annihilation of individual skyrmions, was observed in nanostructured FeGe Hall-bar devices¹⁸.

So far, all electrical measurements of magnetic skyrmions have been based on the interpretation that the

TH resistivity is proportional to the number of skyrmions multiplied by the magnetic flux quantum $\psi_0 (= \frac{h}{e})$, *i.e.*, $\rho_{yx}^T \propto N_{sk}\psi_0$, a result originally derived by Bruno *et al.*¹² for bulk systems in the nondiffusive regime. We note that TH effect itself is *not* topologically protected in the presence of scattering of the conduction electrons. Indeed, Ndiaye and coworkers¹⁹ recently computed numerically the TH effect using the Landauer-Büttiker formulation and found it to be highly sensitive to spin-independent impurities. Furthermore, previous studies of the TH effect have been focused on the charge transport with conserved spin polarization, which only result in a spin-polarization dependent prefactor in the TH resistivity. However, this is not valid in the presence of spin accumulation and spin-flip scattering: The former locally alters the spin polarization and the latter mixes the two conduction channels for spin-up and spin-down electrons. These observations raise an important question of what the effect of spin-flip scattering is on the TH effect, and what the experimental consequences are of spin accumulation in thin film geometries.

Here, we investigate TH effect in a diffusive ferromagnetic metal (FM) thin film with spin-flip scattering by treating spin and charge transport on an equal footing. We show that skyrmions act as sources of spin accumulation, which may build up not only near the lateral boundaries but also in the vicinity of the skyrmions, as shown schematically in Fig. 1; consequently, the TH resistivity is in general reduced and is no longer proportional to the number of skyrmions.

FIG. 1: Schematics of the TH effect in a diffusive ferromagnetic thin film when an external electric field $\mathbf{E}$ is applied: (a) The trajectories of spin-up and spin-down electrons traversing a Néel-type skyrmion of radius r_{sk} due to the TH effect, and (b) spin accumulations built up at the lateral boundaries of the thin film as well as in the vicinity of the skyrmion within spin diffusion length λ_{sd} , owing to the nonuniform emergent magnetic field. Note that the emergent magnetic field acting on spin-up electrons, i.e., $\mathbf{B}_\uparrow$ (denoted by hollow green arrows), and that acting on spin-down electrons, i.e., $\mathbf{B}_\downarrow$ (denoted by hollow orange arrows), are in opposite directions.

II. THE SEMICLASSICAL TRANSPORT THEORY

Our starting point is the steady-state Boltzmann transport equation

$$\mathbf{v} \cdot \frac{\partial f_s}{\partial \mathbf{r}} - e(\mathbf{E} + \mathbf{v} \times \mathbf{B}_s) \cdot \frac{\partial f_s}{\hbar \partial \mathbf{k}} = -\frac{f_s - \langle f_s \rangle}{\tau_s} - \frac{\langle f_s \rangle - \langle f_{-s} \rangle}{\tau_{sf}}, \quad (1)$$

where $f_s(\mathbf{r}, \mathbf{k})$ is the distribution function for electrons with $s = 1$ (-1) or $\uparrow$ ($\downarrow$) denoting spin-up (spin-down) with respect to the local magnetization direction, $\langle f_s \rangle \equiv \int d^2 \Omega_{\mathbf{k}} f_s(\mathbf{r}, \mathbf{k}) / \int d^2 \Omega_{\mathbf{k}}$ with $\Omega_{\mathbf{k}}$ the solid angle in $\mathbf{k}$ -space, and τ_s and τ_{sf} are the momentum and spin-flip relaxation times respectively; $\mathbf{E}$ is the external electric field applied in the longitudinal direction of the FM thin film, i.e., $\mathbf{E} = E_x \hat{\mathbf{x}}$, and $\mathbf{B}_s$ is the emergent magnetic field^{20,21} given by

$$B_{s,i} = -s \frac{\hbar}{2e} \epsilon_{ijk} \mathbf{m} \cdot (\partial_j \mathbf{m} \times \partial_k \mathbf{m}) \quad (2)$$

with $\mathbf{m}$ the unit vector denoting the direction of the magnetization, ∂_j the short-hand denotation for the spatial derivative of $\frac{\partial}{\partial r_j}$ and ϵ_{ijk} the antisymmetric Levi-Civita tensor. We assume the film is sufficiently thin so that the magnetization is uniform along the direction perpendicular to the thin film (i.e., the z -direction), and focus on the transport in the x - y plane. Note that, for this effectively two-dimensional system, only the z -component of the emergent magnetic field is nonzero, i.e., $\mathbf{B}_s = sB\hat{z}$.

Note that by including the emergent magnetic field in Eq. (1) as a driving force acting on the conduction electrons, we have assumed a *strong* exchange coupling J_{ex} between the local magnetization and conduction electron

spins, i.e., $\tau_s J_{ex} / \hbar \gg 1$. In this case, the transverse spin component decays on a time scale of $\tau_{ex} (\sim \hbar / J_{ex})$ much shorter than the electron momentum relaxation time τ_s . Therefore, the spin of the conduction electrons can adiabatically follow the surrounding local magnetization. Recently, the THE in the *weak* exchange coupling regime has also been studied theoretically^{22,23}. Here we shall focus on the strong exchange coupling regime where the emergent magnetic field (or the real-space Berry phase) picture is still valid¹². Also note that the emergent magnetic field was derived^{20,21,24–26} by projecting the scalar and vector gauge potentials that emerge from a canonical transformation that rotates quantization axis to align with the local magnetization onto the spin-up and spin-down bands with respect to the local magnetization orientation. In other words, intrinsic interband spin-flip transitions are neglected²⁷. The spin-flip scattering considered in our transport equation (1) may originate from extrinsic spin-dependent scatterings such as electron-magnon scattering and weak spin-orbit interactions with magnetic impurities, and the relaxation times, i.e., τ_s and τ_{sf} , are associated with the transition probabilities in the standard way.

Next, we separate the distribution function into an equilibrium component $f_{0,s}(\mathbf{k})$ and small nonequilibrium perturbations,

$$f_s(\mathbf{r}, \mathbf{k}) = f_{0,s}(\mathbf{k}) - \frac{\partial f_{0,s}}{\partial \epsilon_{ks}} [-e\mu_s(\mathbf{r}) + g_s(\mathbf{r}, \mathbf{k})], \quad (3)$$

where $-e\mu_s(\mathbf{r})$ and $g_s(\mathbf{r}, \mathbf{k})$ are the zeroth and first velocity moments respectively (the latter satisfies $\int d^2 \mathbf{k} g_s(\mathbf{r}, \mathbf{k}) = 0$), and $\epsilon_{ks} = \frac{\hbar^2 k^2}{2m} - sJ_{ex}$ denotes the energy of spin- s electrons with J_{ex} the exchange splitting of the conduction band. Placing Eq. (3) in the Eq. (1) and separating the odd and even velocity moments of the

distribution function, we find, up to $\mathcal{O}(B)$,

$$g_s \simeq -e\tau_s \mathbf{v} \cdot \left(\mathbf{E} - \nabla_{\mathbf{r}} \mu_s - \frac{\tau_s e}{m} \mathbf{E} \times \mathbf{B}_s \right), \quad (4)$$

and a generalized diffusion equation for the spin accumulation, defined as $\delta\mu \equiv \frac{1}{2}(\mu_{\uparrow} - \mu_{\downarrow})$:

$$\nabla_{\mathbf{r}}^2 \delta\mu - \frac{\delta\mu}{\lambda_{sd}^2} = \frac{\tau e}{m} (\hat{\mathbf{z}} \times \mathbf{E}) \cdot \nabla_{\mathbf{r}} B. \quad (5)$$

Here, the spin averaged diffusion length λ_{sd} is given by $\lambda_{sd}^{-2} \equiv (l_{\uparrow}^{-2} + l_{\downarrow}^{-2})/2$ with $l_s = \sqrt{\frac{1}{2}v_{F,s}^2 \tau_s \tau_{sf}}$, $\tau = \frac{\tau_{\uparrow} + \tau_{\downarrow}}{2}$ is the spin averaged momentum relaxation time, and we have used $\mathbf{B}_{\pm} = -\mathbf{B}_{\mp} = B\hat{\mathbf{z}}$; detailed derivation of the generalized spin diffusion equation is given in Appendix A. We note that the nonuniform emergent magnetic field introduces a source term in the spin diffusion equation, Eq. (5); this may be understood as follows: In analogy to the force acting on magnetic moments induced by magnetic field gradient, the gradient of the emergent magnetic field may serve as a spin-dependent driving force in addition to the Lorentz force; in this case, it acts like a local spin source and hence enters the spin diffusion equation as a source term. In the absence of this source term, spin accumulation can only occur through spin injection at boundaries or interfaces.⁴² The source term gives rise to spin accumulation in the vicinity of the skyrmions where the gradient of the emergent magnetic field is non-zero. This also imparts nonlocal features in the spin and charge transport, as we will show below.

The current density of the spin- s conduction channel can be calculated by $\mathbf{j}_s = \frac{-e}{(2\pi)^2} \int d^2\mathbf{k} f_s(\mathbf{r}, \mathbf{k}) \mathbf{v}$; using Eqs. (3) and (4), it can be expressed as

$$\mathbf{j}_s = \sigma_s (\mathbf{E} - \nabla_{\mathbf{r}} \mu_s) - \sigma_s \left(\frac{\tau_s e}{m} \right) \mathbf{E} \times \mathbf{B}_s, \quad (6)$$

where $\sigma_s = \frac{e^2 \tau_s n_{e,s}}{m}$ is the longitudinal conductivity with $n_{e,s}$ the density of spin- s conduction electrons. The charge and spin current densities by definition are given by the sum and difference of the current densities in the two spin channels, i.e., $\mathbf{j}_{ch} = \mathbf{j}_{\uparrow} + \mathbf{j}_{\downarrow}$ and $\mathbf{j}_{sp} = \mathbf{j}_{\uparrow} - \mathbf{j}_{\downarrow}$.

The TH effect is associated with the transverse current densities that arise from spatial variations of the chemical potentials along the y -direction. Within linear response, the leading-order correction to the longitudinal current due to gradients in the chemical potentials is of the second order in the emergent magnetic field $\mathbf{B}_s$. Therefore up to $\mathcal{O}(B)$, we may focus only on the y -components of the charge and spin current densities by integrating out the skyrmions along x -direction, i.e.,

$$\bar{j}_{ch,y} = -\sigma \left(\frac{d\bar{\mu}}{dy} + p_{\sigma} \frac{d\bar{\delta\mu}}{dy} \right) + \sigma (p_{\tau} + p_{\sigma}) E_x \left(\frac{\tau e \bar{B}}{m} \right) \quad (7)$$

and

$$\bar{j}_{sp,y} = -\sigma \left(p_{\sigma} \frac{d\bar{\mu}}{dy} + \frac{d\bar{\delta\mu}}{dy} \right) + \sigma (1 + p_{\tau} p_{\sigma}) E_x \left(\frac{\tau e \bar{B}}{m} \right), \quad (8)$$

where $\sigma = \sigma_{\uparrow} + \sigma_{\downarrow}$ and $\mu = \frac{\mu_{\uparrow} + \mu_{\downarrow}}{2}$ are the total longitudinal conductivity and the spin averaged chemical potential, $p_{\sigma} \equiv \frac{\sigma_{\uparrow} - \sigma_{\downarrow}}{\sigma_{\uparrow} + \sigma_{\downarrow}}$ and $p_{\tau} \equiv \frac{\tau_{\uparrow} - \tau_{\downarrow}}{\tau_{\uparrow} + \tau_{\downarrow}}$ are the spin asymmetries of the conductivity and relaxation time respectively, and we have defined $\bar{F}(y) \equiv (2l)^{-1} \int_{-l}^l dx F(\mathbf{r})$ with $F(\mathbf{r})$ denoting an arbitrary function and $2l$ the length of the film. Similarly, we can cast the spin diffusion equation along the y direction as follows

$$\frac{d^2 \bar{\delta\mu}}{dy^2} - \frac{\bar{\delta\mu}}{\lambda_{sd}^2} = E_x \left(\frac{\tau e}{m} \frac{d\bar{B}}{dy} \right), \quad (9)$$

with the general solution given by

$$\begin{aligned} \bar{\delta\mu}(y) = & C_- \exp\left(-\frac{y}{\lambda_{sd}}\right) + C_+ \exp\left(\frac{y}{\lambda_{sd}}\right) - \lambda_{sd} E_x \\ & \times \int_{-w}^{+w} d\tilde{y} \left(\frac{\tau e}{2m} \frac{d\bar{B}}{d\tilde{y}} \right) \exp\left(-\frac{|y - \tilde{y}|}{\lambda_{sd}}\right), \end{aligned} \quad (10)$$

where $C_{\pm}$ are two constants of integration to be determined by the boundary conditions, and $2w$ is the film width. Note that the spatially varying emergent magnetic field induces a *nonlocal* term in the spin accumulation, with the range of nonlocality set by the spin diffusion length. A similar nonlocal feature has also been found in the weak exchange coupling regime²³.

With open boundary conditions in the transverse direction, the transverse current density in each spin-channel vanishes at the boundaries, i.e., $\bar{j}_{s,y}(\pm w) = 0$, and it follows that $\bar{j}_{sp}(\pm w) = 0$. The transverse component of the charge current density, up to $\mathcal{O}(B)$, must be zero everywhere, i.e., $\bar{j}_{ch,y}(y) = 0$, due to the absence of bulk charge accumulation in ferromagnetic metals with screening lengths of a few Ångströms. We can thus express the transverse electric field E_y in terms of the spin accumulation and the emergent magnetic field as

$$E_y = -\frac{d}{dy} \bar{\mu} = p_{\sigma} \frac{d}{dy} \bar{\delta\mu} - (p_{\tau} + p_{\sigma}) E_x \frac{e\tau \bar{B}}{m}. \quad (11)$$

Placing Eqs. (7), (8) and (10) in the boundary conditions, we can determine the spin accumulation $\bar{\delta\mu}(y)$ and hence E_y .

III. RESULTS AND DISCUSSIONS

The magnitude of the TH effect can be characterized by the TH resistivity, given by $\rho_{yx}^T = \frac{\bar{E}_y}{j_x}$ where $\bar{E}_y = \frac{1}{2w} \int_{-w}^{+w} dy E_y$. By placing Eq. (10) in Eq. (11) and using the property that the total flux of the emergent magnetic field associated with a skyrmion is equal to the magnetic flux quantum ψ_0 ³, we obtain

$$\rho_{yx}^T = \rho_{yx}^{T,(0)} \left[1 - \frac{p_{\sigma}}{p_{\tau} + p_{\sigma}} \int d^2\mathbf{r} \frac{B(\mathbf{r}) \cosh\left(\frac{y}{\lambda_{sd}}\right)}{N_{sk} \psi_0 \cosh\left(\frac{w}{\lambda_{sd}}\right)} \right] \quad (12)$$

FIG. 2: Spatial profiles of (a) spin accumulation $\bar{\delta\mu}$ and (b) transverse spin current density $\bar{j}_{sp,y}$ in the y -direction for several different spin diffusion lengths, where $\delta\mu_0 \equiv E_x r_{sk} (\frac{e\tau B_0}{m})$ and $j_{sp,y}^{(0)} \equiv (1 - p_\sigma^2) \sigma E_x (\frac{e\tau B_0}{m})$, and $w = 3r_{sk}$.

where $\rho_{yx}^{T,(0)} = (p_\tau + p_\sigma) n_{sk} R_H B_0$, $B_0 = \frac{\psi_0}{\pi r_{sk}^2}$ is the averaged emergent magnetic field per skyrmion with radius r_{sk} , $n_{sk} = \frac{N_{sk} \pi r_{sk}^2}{4w l}$ is a dimensionless skyrmion density ($n_{sk} \rightarrow 1$ for a close packed SkX) with N_{sk} the total number of skyrmions contained in the thin film, $R_H = \frac{1}{en_e(1+p_\tau p_n)}$ is the Hall coefficient with $n_e = n_{e,\uparrow} + n_{e,\downarrow}$ and $p_n = \frac{n_{e,\uparrow} - n_{e,\downarrow}}{n_{e,\uparrow} + n_{e,\downarrow}}$ the total conduction electron density and its spin polarization respectively. We note that the TH resistivity is proportional to the Hall coefficient and hence inversely proportional to the carrier density; in other words, the TH effect may become more sizable in magnetic materials with lower carrier density. Also note that in deriving Eq. (12), we have assumed that the effects of the emergent magnetic fields from different skyrmions are additive, i.e., $B(\mathbf{r}) = \sum_i B(\mathbf{r} - \mathbf{r}_i)$ with $\mathbf{r}_i$ denoting the central position of the i -th skyrmion⁴³.

Analytical results can be obtained in the following two limiting cases. When $\lambda_{sd} \ll r_{sk} < w$, the TH resistiv-

ity coincides with the bulk value, i.e., $\rho_{yx}^T \rightarrow \rho_{yx}^{T,(0)12}$ (provided $B(\mathbf{r})$ has mirror reflection symmetry about the $y = 0$ plane), whereas in the opposite limit of $\lambda_{sd} \gg w > r_{sk}$, we find the TH resistivity is reduced to $\rho_{yx}^T \rightarrow (\frac{p_\tau}{p_\tau + p_\sigma}) \rho_{yx}^{T,(0)}$. At a first glance, this observation may seem a little counterintuitive as one would expect ρ_{yx}^T to approach its ideal bulk value in the weak diffusive limit (i.e., $\lambda_{sd} \rightarrow \infty$). However, this is *not* the case in the presence of spin accumulation in thin FM films. To illustrate this point, it is instructive to examine the spatial distributions of the spin current and spin accumulation induced by a single skyrmion.

Let us consider a single skyrmion residing in the center of the thin film. The magnetization unit vector defining a skyrmion may be expressed as $\mathbf{m}[\Theta(r), \Phi(\phi)] = (\sin \Theta \cos \Phi, \sin \Theta \sin \Phi, \cos \Theta)$, where Θ and Φ are the polar and azimuthal angles of the magnetization at a given position $\mathbf{r} = r(\cos \phi, \sin \phi)$ relative to the center of the skyrmion. In FM thin films with perpendicular anisotropy in contact with a spin-orbit scatterer such as Ta or Pt, interfacial Dzyaloshinskii-Moriya interactions favor Néel-type skyrmions^{11,28–30}, for which the azimuthal angle can be written as $\Phi(\phi) = \nu\phi + \gamma_c$ with vorticity $\nu = 1$ and chirality $\gamma_c = \pm\pi$ and the polar angle of the magnetization – varying linearly with its distance from the center of the skyrmion – may be described as $\Theta(r) = \pi \left(1 - \frac{r}{r_{sk}}\right) H(r_{sk} - r)$ with $H(x)$ is the unit step function.

In Fig. 2, we show the spatial distributions of the spin accumulation $\bar{\delta\mu}$ and transverse spin current density $\bar{j}_{sp,y}$ along the y -direction of the thin film (averaged over the x -coordinate across the skyrmion). When $\lambda_{sd} \ll r_{sk}$, because of the short spin diffusion length both $\bar{\delta\mu}$ and $\bar{j}_{sp,y}$ are spatially localized to within the skyrmion texture, and hence the boundaries have no effect on the spin transport. However, when λ_{sd} becomes comparable to or larger than r_{sk} , both $\bar{\delta\mu}$ and $\bar{j}_{sp,y}$ spread out over the spin diffusion length. It follows that the diffusive spin current generated by the gradient of the spin accumulation will be partially converted to a charge current due to the spin asymmetry of conductivities for spin-up and spin-down electrons (i.e., $p_\sigma \neq 0$), which consequently suppresses the bulk contribution of the TH current, as indicated by Eq. (11). Similar nonlocal spin and charge transport driven by the emergent electric field was studied earlier^{31,32} in the context of the spin electromotive force^{26,33} and enhanced damping induced by magnetization dynamics^{24,27}.

Using Eq. (12), we can also numerically calculate the TH resistivity induced by a Néel skyrmion for any arbitrary magnitude of the spin diffusion length, as shown in Fig. 3. We find that the TH resistivity decreases with increasing spin diffusion length, reaching its maximum and minimum values in the two limits of $\lambda_{sd}/r_{sk} \rightarrow 0$ and $\lambda_{sd}/r_{sk} \rightarrow \infty$ respectively; this is because longer spin diffusion length leads to larger gradient of spin accumulation and hence greater backflow spin (polarized) diffusion

FIG. 3: TH resistivity ρ_{yx}^T generated by a single Néel skyrmion as a function of spin diffusion length λ_{sd} in a thin film of half width $w = 3r_{sk}$ for several different p_τ , where $\rho_{yx}^{T,(0)}$ is the ideal bulk TH resistivity independent of λ_{sd} ; the insets show the corresponding spatial profile of the emergent magnetic field as a function of the distance from the skyrmion center. Other parameters used: $J_{ex}/\epsilon_F = 0.2^{34}$.

current that effectively suppresses the source current generated by the skyrmion, as indicated by Fig. 2. For given ratios of λ_{sd}/r_{sk} and J_{ex}/ϵ_F , ρ_{yx}^T decreases with the spin asymmetry of the electron momentum relaxation p_τ . We also note that, with fixed topological charge ν and the materials parameters λ_{sd}/r_{sk} and p_τ , the TH resistivity is *independent* of the detailed spin configuration of the skyrmion; the analyses of the TH effect for a Bloch-type skyrmion and a chiral skyrmion bubble with the same topological charge can be found in Appendix B.

Having understood the TH effect in the diffusive regime for a single skyrmion, we now proceed to estimate the TH resistivity induced by a hexagonal SkX. Figure 4 shows the TH resistivity ρ_{yx}^T as a function of the number of skyrmions N_{sk} in FM thin films with different spin diffusion lengths. We find that ρ_{yx}^T increases linearly with the number of skyrmions contained in the thin film, as the emergent magnetic field of different skyrmions are additive. However, only in the limit of short spin diffusion length (*i.e.*, $\lambda_{sd}/r_{sk} \rightarrow 0$), is the TH resistivity proportional to the number of skyrmions, in agreement with the ideal bulk TH effect¹². For a close-packed skyrmion lattice (*i.e.*, $n_{sk} \rightarrow 1$) with $r_{sk} = 100$ nm and a spin diffusion length of $\lambda_{sd} = 10$ nm^{35,36}, the estimated TH resistivity is about $\rho_{yx}^T \sim 0.016 \mu\Omega \cdot \text{cm}$, where we have used $R_H = 0.05 \mu\Omega \cdot \text{cm}$ and $p_\sigma = 2p_\tau = 0.4$. This is in order-of-magnitude agreement with recent measurements of the TH resistivity in transition metal thin films and multilayers^{16,36}. When the skyrmion size is further reduced, the magnitude of the TH resistivity depends on the trade-off between the increased emergent magnetic field B_0 and the reduction due to spin accumulation.

For example, with the same material parameters but a smaller skyrmion radius $r_{sk} = 0.5\lambda_{sd} = 5$ nm, ρ_{yx}^T will be reduced by 40%; in the extreme case of $p_\tau/p_\sigma \rightarrow 0$ and $r_{sk}/\lambda_{sd} \rightarrow 0$, ρ_{yx}^T might even be completely suppressed.

FIG. 4: The ratios of ρ_{yx}^T / ρ_{xx} and $\rho_{yx}^T / \rho_{yx}^{T,(0)}|_{N_{sk}=1}$ as a function of skyrmion number for several spin diffusion lengths λ_{sd} , where ρ_{xx} is the longitudinal resistivity and $\rho_{yx}^{T,(0)}|_{N_{sk}=1}$ is the ideal TH resistivity for a single skyrmion. Parameters used: $p_\tau = 0.1$, $J_{ex}/\epsilon_F = 0.2$, $\tau = 10^{-13}$ s, $m = 10^{-30}$ kg, $w = 6r_{sk}$, and inter-skyrmion distance $d_{sk-sk} = 4 r_{sk}$.

As a final point, we discuss material considerations in observations of the TH effect in FM thin films and multilayers. The spin diffusion length of typical transition metal ferromagnets, typically of the order of 10 nm³⁵, is much smaller than the room temperature skyrmions (with size in the range of $0.1 \sim 1 \mu\text{m}$) observed in magnetic thin films^{10,11,37,38}. In this case, the spin and charge transport remain spatially localized to within the skyrmion spin texture, and the measured TH resistivity will therefore still agree with the ideal bulk TH effect¹², as shown by Figs.(2) and (3). However, increasing efforts have been directed at searching for nanoscale skyrmions at room temperature, which is desirable for device applications¹⁰. According to our theory, when the skyrmion size approaches the spin diffusion length of the FM layer, a reduction of the TH resistivity should be expected, as indicated by Eq. (12). In order to minimize such reduction, FMs with small p_σ but sizable p_τ would be advantageous. For transition metals, p_σ is in fact dominated by scattering of the *s*-electrons from the 3*d*-shell, which can be tuned in their alloys. For example, Fert and Campbell³⁹ have demonstrated that in Ni-based binary alloys, p_σ may be varied significantly and even tuned to change signs.

IV. SUMMARY AND CONCLUSION

In this work, we have used a semi-classical spin-dependent Boltzmann equation to study the effect of

spin-flip scattering and spin accumulation on the TH effect in ferromagnetic thin films. We found that the nonuniform emergent magnetic field serves as a source term for the spin diffusion and imparts nonlocal features to the spin and charge transport. A generalized spin diffusion equation was derived, the solution of which shows that spin accumulation may build up not only at the lateral boundaries of a current-carrying thin film but also in the vicinity of the magnetic skyrmions; such spin accumulation gives rise to a spin-polarized diffusion current that flows against the bulk TH current and hence attenuates the TH signal. A general expression for the TH resistivity was obtained which applies to diffusive ferromagnetic thin films especially when the size of the magnetic skyrmions is comparable or smaller than the spin diffusion length.

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Appendix A: Derivation of the generalized spin diffusion equation

Let us begin with the following Boltzmann equation

$$\mathbf{v} \cdot \frac{\partial f_s}{\partial \mathbf{r}} - e(\mathbf{E} + \mathbf{v} \times \mathbf{B}_s) \cdot \frac{\partial f_s}{\hbar \partial \mathbf{k}} = -\frac{f_s - \langle f_s \rangle}{\tau_s} - \frac{\langle f_s \rangle - \langle f_{-s} \rangle}{\tau_{sf}}, \quad (\text{A1})$$

where $f_s(\mathbf{r}, \mathbf{k})$ is the distribution function for electrons with spin character s , $\langle f_s \rangle \equiv \int d^2 \Omega_{\mathbf{k}} f_s(\mathbf{r}, \mathbf{k}) / \int d^2 \Omega_{\mathbf{k}}$ with $\Omega_{\mathbf{k}}$ the solid angle in $\mathbf{k}$ -space, and τ_s and τ_{sf} are the momentum and the spin-flip relaxation times respectively; $\mathbf{E}$ is the external electric field is applied along the longitudinal direction of the FM thin film, *i.e.*, $\mathbf{E} = E_x \hat{\mathbf{x}}$, and $\mathbf{B}_s$ is the emergent magnetic field. Separating the distribution function into an equilibrium component $f_{0,s}(\mathbf{k})$ and small nonequilibrium perturbations as follows

$$f_s(\mathbf{r}, \mathbf{k}) = f_{0,s}(\mathbf{k}) - \frac{\partial f_{0,s}}{\partial \varepsilon_{ks}} [-e\mu_s(\mathbf{r}) + g_s(\mathbf{r}, \mathbf{k})], \quad (\text{A2})$$

where $e\mu_s(\mathbf{r})$ and $g_s(\mathbf{r}, \mathbf{k})$ are the zeroth and first velocity moments respectively (the latter satisfies $\int d^2 \mathbf{k} g_s(\mathbf{r}, \mathbf{k}) = 0$), and $\varepsilon_{ks} = \frac{\hbar^2 k^2}{2m} - sJ_{ex}$ denotes the energy of spin- s electrons with J_{ex} the exchange splitting of the conduction band. Placing Eq. (A2) in the Eq. (A1) and separating the odd and even velocity moments of the distribution function, we find, up to $O(\mathbf{B}_s)$,

$$-e(\mathbf{E} - \nabla_{\mathbf{r}} \mu_s) \cdot \mathbf{v} + e(\mathbf{v} \times \mathbf{B}_s) \cdot \frac{\partial g_s}{\partial \mathbf{k}} = \frac{g_s}{\tau_s} \quad (\text{A3})$$

and

$$\mathbf{v} \cdot \frac{\partial}{\partial \mathbf{r}} g_s(\mathbf{r}, \mathbf{k}) = \frac{e(\mu_s - \mu_{-s})}{\tau_{sf}}, \quad (\text{A4})$$

where we have assumed a spherical Fermi surface and have neglected higher order terms of the order of $O\left(\frac{J_{ex}\tau_s}{\varepsilon_F\tau_{sf}}\right)$ [noting that $\frac{J_{ex}}{\varepsilon_F} \sim 0.1$ for typical transition metal ferromagnets and $\frac{\tau_s}{\tau_{sf}} \ll 1$ for the diffusive regime]. By placing the ansatz $g_s = \mathbf{A}_s \cdot \mathbf{v}$ (where $\mathbf{A}_s$ is an arbitrary vector independent of $\mathbf{v}$) in Eq. (A3) and solving the resulting vector equation for $\mathbf{A}_s$, up to $O(\mathbf{B}_s)$, we find

$$g_s \simeq -e\tau_s \mathbf{v} \cdot \left(\mathbf{E} - \nabla_{\mathbf{r}} \mu_s - \frac{\tau_s e}{m} \mathbf{E} \times \mathbf{B}_s \right), \quad (\text{A5})$$

Note that since spatial variation in chemical potential is induced by $\mathbf{B}_s$, so we have discarded the higher order term $\nabla_{\mathbf{r}} \mu_s \times \mathbf{B}_s$ as well. Plugging Eq. (A5) back in Eq. (A4) and carrying out the angular integration in the momentum space on both sides of the resulting equation, we arrive at

$$\nabla_{\mathbf{r}}^2 \mu_s - \frac{\mu_s - \mu_{-s}}{l_s^2} = -\frac{\tau_s e}{m} \mathbf{E} \cdot (\nabla_{\mathbf{r}} \times \mathbf{B}_s), \quad (\text{A6})$$

where $l_s = \sqrt{\frac{1}{2} v_{F,s}^2 \tau_s \tau_{sf}}$. Subtracting Eq. (A6) for $s = \uparrow$ from that for $s = \downarrow$, we arrive at the generalized spin diffusion equation

$$\nabla_{\mathbf{r}}^2 \delta\mu - \frac{\delta\mu}{\lambda_{sd}^2} = \frac{\tau e}{m} (\hat{\mathbf{z}} \times \mathbf{E}) \cdot \nabla_{\mathbf{r}} B, \quad (\text{A7})$$

where the spin averaged diffusion length λ_{sd} is given by $\lambda_{sd}^{-2} \equiv (l_{\uparrow}^{-2} + l_{\downarrow}^{-2})/2$, $\tau = \frac{\tau_{\uparrow} + \tau_{\downarrow}}{2}$ is the spin averaged momentum relaxation time and we have used the relation $\mathbf{B}_+ = -\mathbf{B}_- = B\hat{\mathbf{z}}$. Note that the generalized spin diffusion equation (A7) involves an additional source term associated with the spatial gradient of the emergent magnetic field.

Appendix B: Topological Hall effect induced by skyrmions with different spin configurations

1. Bloch-type skyrmion

A standard Bloch-type skyrmion has the same polar angle profile as a standard Néel-type skyrmion but with a different chirality of $\gamma_c = \pm \frac{\pi}{2}$. However, the emergent magnetic field does not rely on γ_c , as can be explicitly

seen from the expression of the emergent magnetic field in polar coordinate system³, *i.e.*,

$$B(r) = -\frac{\hbar}{2e} \sin \Theta \frac{1}{r} \frac{d\Theta}{dr} \frac{d\Phi}{d\phi} \quad (\text{B1})$$

It follows that the topological Hall resistivity of Bloch-type skyrmions (or skyrmion lattices) should be the same as that of Néel-type skyrmions (or skyrmion lattices) which was examined in the main text.

2. Chiral skyrmion bubble

Skyrmion bubbles (or chiral bubbles) were also found in magnetic thins and multilayers in recent experiments^{11,40}. At variance with a prototypical Néel skyrmion, the domain of reversed magnetization in a skyrmion bubble is more extended and surrounded by a narrow Néel domain wall with fixed chirality. The magnetization polar angle profile may be expressed as $\Theta(r) = \frac{\pi}{2} \{1 - \tanh[f(r)]\}$ ⁴¹ with $f(r) = \ln\left(\frac{2r}{r_{sk}}\right) + \frac{2r-r_{sk}}{2a_{DW}}$ and a_{DW} the Néel wall width. The emergent magnetic field for a skyrmion bubble can be written as

$$B_z = \frac{\pi\hbar}{4e} \frac{\sin \Theta f'(r)}{(\cosh[f(r)])^2 r} \quad (\text{B2})$$

In Fig. B.1, we show the spatial distributions of the spin accumulation $\bar{\delta\mu}$ and transverse spin current density $\bar{j}_{sp,y}$ along the y -direction of the thin film (averaged over the x coordinate across the skyrmion). When the spin diffusion length λ_{sd} is much smaller than radius of the skyrmion bubble r_{sk} (see the solid blue line for $\lambda_{sd} = a_{DW} = 0.2 r_{sk}$), the magnitudes of both $\bar{\delta\mu}$ and $\bar{j}_{sp,y}$ reach their maxima around the narrow Néel wall, at variance with the case for a single Néel skyrmion as shown in Fig. 2. When λ_{sd} is comparable or larger than r_{sk} , the spatial profiles of $\bar{\delta\mu}$ and $\bar{j}_{sp,y}$ for a skyrmion bubble coincide with those for a standard Néel skyrmion, since the characteristic range of nonlocality, set by the spin diffusion length, spans the entire skyrmion which makes the transport properties insensitive to the local spin structure of the skyrmion.

FIG. B.1: Spatial profiles of (a) spin accumulation $\bar{\delta\mu}$ and (b) transverse spin current density $\bar{j}_{sp,y}$ in the y -direction induced by a single skyrmion bubble for several different spin diffusion lengths, where $\delta\mu_0 \equiv E_x r_{sk} (\frac{e\tau B_0}{m})$ and $j_{sp,y}^{(0)} \equiv (1 - p_\sigma^2) \sigma E_x (\frac{e\tau B_0}{m})$, and the skyrmion radius is taken to be $r_{sk} = \frac{w}{3}$. we have also used $a_{DW} = 0.2 r_{sk}$ and $J_{ex} = 0.2 \epsilon_F$.

In Fig. B.2, we show the topological Hall resistivity as a function of the ratio of λ_{sd}/r_{sk} . We find that the topological Hall resistivity induced by a single skyrmion bubble turns out to be exactly the same as that for a standard Néel skyrmion. This is understandable since the topological Hall resistivity is calculated by integrated over the entire skyrmion and hence only relies on the topology (or topological charge) of the skyrmion rather than its detailed spin structure.

FIG. B.2: Topological Hall resistivity ρ_{yx}^T generated by a single skyrmion bubble as a function of spin diffusion length λ_{sd} in a thin film of width $w = 6r_{sk}$ for several different p_τ , where $\rho_{yx}^{T,(0)}$ is the ideal bulk TH resistivity independent of λ_{sd} ; the insets show the corresponding spatial profile of the emergent magnetic field. We have defined $\rho_{TH}^{(0)} = (p_\tau + p_\sigma) R_H \psi_0 / S$ and used $a_{DW} = 0.2r_{sk}$ and $J_{ex} = 0.2\epsilon_F$.

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- ⁴³ This is true within linear response so long as the skyrmions are well separated so that $2r_{sk}$ is smaller than the skyrmion-skyrmion separation.