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Nonlocal Anomalous Hall Effect in Heavy-metal/Ferromagnetic-insulator Bilayers

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The anomalous Hall effect is deemed to be a unique transport property of ferromagnetic metals, caused by the concerted action of spin polarization and spin-orbit coupling. Nevertheless, recent experiments have shown that the effect also occurs in a nonmagnetic metal (Pt) in contact with a magnetic insulator (yttrium iron garnet (YIG)), even when precautions are taken to ensure there is no induced magnetization in the metal. We propose a theory of this effect based on the combined action of spin-dependent scattering from the magnetic interface and the spin Hall effect in the bulk of the metal. At variance with previous theories, we predict the effect to be of first order in the spin-orbit coupling, just as the conventional anomalous Hall effect – the only difference being the spatial separation of the spin orbit interaction and the magnetization. For this reason we name this effect *nonlocal anomalous Hall effect* and predict that its sign will be determined by the sign of the spin Hall angle in the metal. The AH conductivity that we calculate from our theory is in order of magnitude agreement with the measured values in Pt/YIG structures.

Introduction.— The anomalous Hall (AH) effect is the generation of an electric current perpendicular to both the electric field and the magnetization in a ferromagnetic metal [1]. At variance with the ordinary Hall effect, which arises from the action of a magnetic field on the orbital motion of the electrons, the AH effect is ascribed to strong spin-orbit coupling in concert with spin-polarized itinerant electrons. The spin orbit coupling plays a central role in inducing a left-right asymmetry (with respect to the direction of the electric field) in the scattering of electrons of opposite spins. It is this asymmetry that generates a transverse charge current from a longitudinal spin polarized current. The same scattering process generates a pure transverse spin current for systems with spin unpolarized electrons, which is known as spin Hall effect [2–5]. Based on this picture, the conventional AH effect appears at *first order* in spin orbit coupling, no matter which kind of microscopic mechanisms predominates.

Recently, the spin and charge transport in a nonmagnetic heavy-metal (e.g., Pt) layer in direct contact with a ferromagnetic insulator (e.g., YIG) layer has been studied intensively [6–15]. While the focus of the attention has been on the anisotropic magnetoresistance, an AH effect has also been observed [6, 8, 15]. In view of the two aforementioned ingredients for the AH effect in ferromagnets, it is puzzling that an AH current would arise in Pt in the absence of spin polarized conduction electrons. In a first attempt to solve the puzzle, Huang et. al. [6] showed that the Pt layer in close proximity with YIG acquires ferromagnetic characteristics, which essentially subsumes the novel AH effect under the conventional AH effect for ferromagnetic metals. This explanation ran into difficulties when it was found that the AH effect persists in Pt/Cu/YIG trilayers [7] where a Cu layer is deliberately

inserted to eliminate the magnetic proximity effect.

An alternative explanation was then proposed [8, 14], based on the physical mechanism depicted in panel (a) of Fig. 1. In this mechanism the applied charge current j_x generates, via the spin Hall effect, a spin current Q_z^y propagating in the z -direction with spin along the y -direction. When those electrons carrying Q_z^y are reflected back from the magnetic interface, spin rotation occurs and gives rise to an additional spin current of Q_z^x , which in turn induces a transverse charge current j_y via the inverse spin Hall effect [16, 17]. Based on this picture, the transverse electric current is of *second order* in the spin orbit coupling or spin Hall angle; for this reason, we will hereafter refer to this mechanism as the *double spin-Hall mechanism*. It is worth mentioning that a fit to the experimental data based on this model [8, 14], requires a spin diffusion length on the order of 1 nm. Such a short spin diffusion length, an order of magnitude smaller than the room-temperature electron mean path of Pt [18], casts doubt on the internal consistency of the model.

In this paper, we propose a different mechanism for the AH effect observed in hybrid heavy-metal/ferromagnetic-insulator structures. The essential new ingredient is the scattering of electrons from the (rough) metal-insulator interface. Because the insulator is magnetic, the scattering rate is spin-dependent [19]. This means that a charge current flowing parallel to the interface is partially converted to a spin current, while a spin current flowing parallel to the interface is partially converted to a charge current. The surface-induced conversion of charge to spin current and vice versa conspires with the spin Hall effect in the bulk of the metal to produce the observed AH current. As shown in Fig. 1(b), this may happen in two ways: in the first process, (b1), the charge current j_x generates, via spin-dependent interfacial scattering a spin current

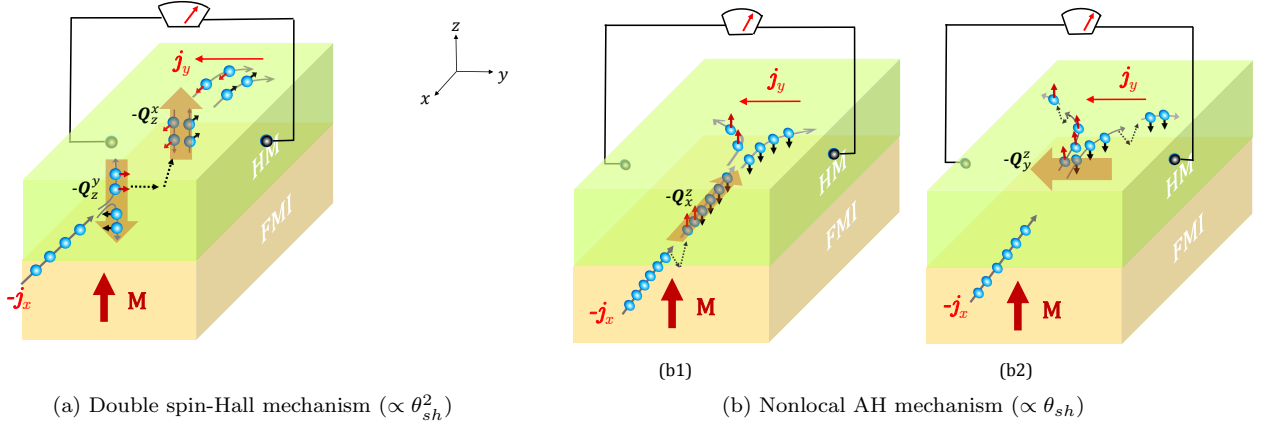


FIG. 1: Schematics of two different mechanisms of the AH effect in heavy-metal (HM)/ferromagnetic-insulator (FMI) bilayers: (a) the double spin-Hall mechanism and (b) nonlocal AH mechanism with two coexisting physical processes depicted separately in panels b1 and b2. The curved arrows represent the trajectories of electrons upon spin orbital scattering and the dotted arrows stand for spin dependent scattering at the magnetic interface.

Q_x^z , which subsequently gives rise to the transverse spin polarized current j_y via the inverse spin Hall effect; in the second process, (b2), j_x first generates, via spin Hall effect, a transverse spin current Q_y^z , which is then turned into a spin polarized current j_y due to spin dependent interfacial scattering. Both physical processes involve spin orbit scattering only once (through the spin Hall effect) and hence the resulting AH current is of *first order* in the spin orbit coupling or spin Hall angle. As a matter of fact, this AH effect has the same physical nature as its conventional counterpart in bulk ferromagnets, and differs from the latter only in the spatial separation of the spin orbit interaction and the magnetization: it is for this reason that we name it *nonlocal AH effect*.

Compared to the double spin-Hall mechanism [8, 14], our mechanism requires a single action of the spin Hall effect to generate a spin current flowing *in the plane of the layers*, which cooperates with the spin-dependent interfacial scattering to produce the AH current. The new mechanism has distinctive features that can be tested experimentally, the most striking one being the sign of the effect, which we predict to track the sign of the bulk spin Hall angle. In addition, our mechanism, at variance with the double spin-Hall mechanism, is *not* associated with spin diffusion, and hence also applies in the ballistic regime when the thickness of the metal layer is smaller than the electron mean free path.

Linear response theory—Let us consider a heavy-metal/ferromagnetic-insulator bilayer as shown in Fig. 1 with an external electric field applied in the x -direction (i.e., $\mathbf{E}_{ext} = E_{ext}\hat{\mathbf{x}}$) and with the magnetization of the insulator layer pointing in the z direction, i.e., $\mathbf{m} = \hat{\mathbf{z}}$. We also assume that both surfaces of the metal are rough, but on the average translational invariance is recovered so that the transport properties are independent of x and

y coordinates. The linear response of current densities to spin dependent electric fields can be written as follows

$$\begin{aligned} \mathbf{j}(z) &= C_0\mathbf{E}(z) + C_s\boldsymbol{\mathcal{E}}_{\parallel}(z) \\ \mathbf{Q}_{\parallel}(z) &= C_0\boldsymbol{\mathcal{E}}_{\parallel}(z) + C_s\mathbf{E}(z) \\ \mathbf{Q}_{\perp}(z) &= C'_{\perp}\boldsymbol{\mathcal{E}}_{\perp}(z) + C''_{\perp}\hat{\mathbf{z}} \times \boldsymbol{\mathcal{E}}_{\perp}(z) \end{aligned} \quad (1)$$

where $\mathbf{j}(z) = (j_x, j_y)$ is the in-plane current density (note that $j_z = 0$ everywhere in the metal layer due to the open boundary conditions), $\mathbf{Q}_{\parallel} = (Q_x^z, Q_y^z)$ is the in-plane spin-current density (with spin in the z direction), and $\mathbf{Q}_{\perp} = (Q_x^y, Q_y^x)$ is the perpendicular-to-plane spin current density carrying the x and y components of the spin. The corresponding electric fields are $\mathbf{E} = (E_x, E_y)$, $\boldsymbol{\mathcal{E}}_{\parallel} = (\mathcal{E}_x^z, \mathcal{E}_y^z)$ and $\boldsymbol{\mathcal{E}}_{\perp} = (\mathcal{E}_x^y, \mathcal{E}_y^x)$. Notice that C_k is defined as the integral operator with kernel $c_k(z, z')$, i.e., $C_k f(z) \equiv \int dz' c_k(z, z') f(z')$. While C_0 is an ordinary in-plane conductivity, C_s describes the generation of an in-plane spin current from an electric field in the presence of spin-dependent surface scattering. As we show below, C_s is the essential ingredient of our theory, producing an AH current of first order in the spin Hall angle. C'_{\perp} and C''_{\perp} are the perpendicular-to-plane spin conductivities related to the spin-mixing conductance [20, 21]. In particular, C''_{\perp} is the essential ingredient of the double spin-Hall mechanism [14], but *plays no role* in the first order AH effect studied here.

In the presence of the bulk spin-orbit scattering, the driving electric fields \mathbf{E} , $\boldsymbol{\mathcal{E}}_{\parallel}$ and $\boldsymbol{\mathcal{E}}_{\perp}$ are self-consistently determined by the internal current densities as follows

$$\begin{aligned} \mathbf{E} &= \mathbf{E}_{ext} + \rho_{sh}\hat{\mathbf{z}} \times (\mathbf{Q}_{\parallel} - \mathbf{Q}_{\perp}) \\ \boldsymbol{\mathcal{E}}_{\parallel} &= \rho_{sh}\hat{\mathbf{z}} \times \mathbf{j} \\ \boldsymbol{\mathcal{E}}_{\perp} &= -\rho_{sh}\hat{\mathbf{z}} \times \mathbf{j} \end{aligned} \quad (2)$$

where $\rho_{sh} \equiv \rho_0\theta_{sh}$ with ρ_0 being the Drude resistivity and θ_{sh} the spin Hall angle of the metal layer. Solving

the system of linear equations (1) and (2), we obtain a general expression for the AH current density up to $O(\theta_{sh}^2)$

$$j_y(z) = [\rho_{sh} \{C_0, C_s\} - \rho_{sh}^2 C_0 C_\perp'' C_0] E_{ext} \quad (3)$$

where $\{, \}$ represents the anti-commutator of the two integral operators. Note that with finite C_s the AH effect appears already at the first order of θ_{sh} . The two orderings of C_0 and C_s in the anti-commutator of Eq. (3) correspond to the processes b1 and b2 of Fig. 1. The sec-

ond term on the right hand side of Eq. (3) corresponds to the double spin-Hall mechanism which is of second order in θ_{sh} and is proportional to conductivity kernel C_\perp'' . In what follows, we employ the Boltzmann transport theory to explicitly construct the integral kernels C_0 and C_s in the presence of a rough magnetic interface.

Boltzmann theory—To quantitatively describe the non-local AH effect in a heavy metal thin layer with an external electric field applied in the x -direction (see Fig. 1(b)), we make use of the spinor Boltzmann equation in the relaxation time approximation [3, 22–25]

$$v_z \frac{\partial \hat{f}(\mathbf{k}, z)}{\partial z} - e E_{ext} v_x \left(\frac{\partial \hat{f}_0}{\partial \varepsilon_k} \right) + \frac{\boldsymbol{\sigma} \cdot [\mathbf{e}_k \times \hat{\mathbf{l}}(k, z)]}{\tau_{so}} = - \frac{\hat{f}(\mathbf{k}, z) - \hat{f}(k, z)}{\tau} - \frac{2\hat{f}(k, z) - \hat{I} T r_\sigma \hat{f}(k, z)}{\tau_{sf}} \quad (4)$$

where \hat{f}_0 and $\hat{f}(\mathbf{k}, z)$ are 2×2 matrices represent respectively the equilibrium and nonequilibrium spinor distribution functions, $\mathbf{v} = d\varepsilon_k / \hbar d\mathbf{k}$ is conduction electron velocity, $\hat{f}(k, z) \equiv (1/4\pi) \int d\Omega_{\mathbf{k}} \hat{f}(\mathbf{k}, z)$ is the angular average of the distribution and $\hat{\mathbf{l}}(k, z) \equiv (1/4\pi) \int d\Omega_{\mathbf{k}} \mathbf{e}_k \hat{f}(\mathbf{k}, z)$ is its dipolar moment, with \mathbf{e}_k the unit vector of \mathbf{k} . Non-spin-flip and spin-slip processes are included, with τ and τ_{sf} being the momentum and spin relaxation times respectively. The additional source term $\tau_{so}^{-1} \boldsymbol{\sigma} \cdot [\mathbf{e}_k \times \hat{\mathbf{l}}(k, z)]$, where τ_{so}^{-1} is the spin-orbit scattering rate, is responsible for the spin Hall effect [26–28]. It is this term that generates the current-dependent fields in Eq. (2).

The crucial step in our theory is the description of spin-dependent interfacial scattering via boundary conditions for the distribution function. For the interface (at $z = 0$) between the heavy metal and the ferromagnetic insulator, we impose the following generalized Fuchs-Sondheimer boundary condition [29],

$$\hat{f}^+(\mathbf{k}, 0) = \frac{1}{2} \hat{s} \hat{R}^\dagger \hat{f}^-(\mathbf{k}, 0) \hat{R} + \frac{1}{2} (\hat{I} - \hat{s}) \langle \hat{f}^-(\mathbf{k}, 0) \rangle + h.c. \quad (5)$$

where the superscripts $+$ and $-$ label the distribution functions for $v_z > 0$ and $v_z < 0$ respectively, the hermitian conjugate ensures \hat{f}^+ to be an hermitian, \hat{I} is the 2×2 identity matrix, $\langle \hat{f} \rangle = (2\pi)^{-1} \int d\phi_{\mathbf{k}} \hat{f}$ with $\phi_{\mathbf{k}}$ the \mathbf{k} -space azimuthal angle, and both \hat{s} and \hat{R} are 2×2 matrices in spin space which are responsible for spin dependent specular reflection and spin rotation of incident electrons.

The matrix \hat{R} , satisfying $\hat{R}^\dagger \hat{R} = \hat{I}$, is the reflection amplitude matrix which captures the spin rotation of electrons that are *specularly reflected* from the magnetic

interface (Note that we assume such a coherent spin rotation does not occur for the diffusively scattered electrons). The explicit form of \hat{R} can be determined by electron wave function matching subject to the following *spin-dependent* potential barrier

$$\hat{V}(z) = \left(V_b \hat{I} - J_{ex} \hat{\sigma}_z \right) \Theta(-z) \quad (6)$$

where V_b is the averaged potential barrier of the insulator, J_{ex} measures the spin splitting of the energy barrier, $\hat{\sigma}_z$ is the z -component of the Pauli spin matrices, and $\Theta(z)$ is the unit step function. Explicitly, \hat{R} takes the following form [30]

$$\hat{R} = \left(\frac{R^\uparrow + R^\downarrow}{2} \right) \hat{I} + \left(\frac{R^\uparrow - R^\downarrow}{2} \right) \hat{\sigma}_z \quad (7)$$

where $R^\alpha = -(\kappa^\alpha + ik_z) / (\kappa^\alpha - ik_z)$ with k_z the z -component of the electron wave vector, $\kappa^\alpha \equiv \sqrt{2m_e^* (V_b - \alpha J_{ex}) - k_z^2}$ ($\alpha = \pm$ or $\uparrow\downarrow$ and we have let $\hbar = 1$ for notation convenience) and m_e^* being the electron effective mass.

The matrix \hat{s} , on the other hand, is introduced to describe the averaged effects of spin dependent scattering at the magnetic interface due to roughness, impurities, and etc. In general, we write [31, 32]

$$\hat{s} = s_0 \left(\hat{I} + p_s \hat{\sigma}_z \right) \quad (8)$$

where $s_0 \equiv (s^\uparrow + s^\downarrow) / 2$ is the average of the specular reflection coefficients s^\uparrow and s^\downarrow for spin-up and spin-down electrons with “up” and “down” defined with respect to \mathbf{m} ($= \hat{\mathbf{z}}$), and $p_s \equiv (s^\uparrow - s^\downarrow) / (s^\uparrow + s^\downarrow)$ is their asymmetry. A simple model calculation for the rough interface

yields [19], to the lowest order in J_{ex}/V_b , the specular reflection asymmetry $p_s \simeq -\frac{2J_{ex}}{V_b}(1-s_0)$ for $s_0 \lesssim 1$. Note that p_s is *negative*, meaning that more spin-down electrons are specularly scattered than spin-up electrons, for the former encounter a higher energy barrier. Also, we notice that a rough magnetic interface is essential for the spin asymmetry of the specular reflection coefficients: for an ideally flat interface, both s^\uparrow and s^\downarrow are exactly equal to one, and no charge/spin conversion can occur.

For the outer surface at $z = d$, we assume, for simplicity, that the scattering is diffusive, i.e.,

$$\hat{f}^-(\mathbf{k}, d) = \langle \hat{f}^+(\mathbf{k}, d) \rangle \quad (9)$$

Note that the boundary conditions given by Eqs. (5) and (9) demand that both charge and spin currents flowing along the z -direction vanish at the outer (non magnetic) surface, whereas only the charge current and the z -component of the spin current flowing along the z -direction vanish at the magnetic surface.

By solving the Boltzmann equation (4) with the boundary conditions given by Eqs. (5) and (9), we have calculated the current densities in the heavy-metal layer. Up to first order in $\theta_{sh}(\equiv \tau/\tau_{so})$, the Hall current density can be expressed as follows

$$j_y^{ah}(z) = \rho_{sh} E_{ext} \int_0^d \frac{dz'}{l_e} [c_s(z, z') \bar{c}_0(z') + c_0(z, z') \bar{c}_s(z')] \quad (10)$$

where l_e is the electron mean free path and the nonlocal integral kernels $c_s(z, z')$ and $c_0(z, z')$ are given by

$$c_0(z, z') = \frac{3}{4} \sigma_0 \int_0^1 d\xi (\xi^{-1} - \xi) \left(s_0 e^{-\frac{z+z'}{l_e \xi}} + e^{-\frac{|z-z'|}{l_e \xi}} \right) \quad (11)$$

and

$$c_s(z, z') = \frac{3}{4} p_s \sigma_0 \int_0^1 d\xi (\xi^{-1} - \xi) s_0 e^{-\frac{z+z'}{l_e \xi}} \quad (12)$$

with their spatial integrations defined as $\bar{c}_0(z) \equiv \int_0^d \frac{dz'}{l_e} c_0(z, z')$ and $\bar{c}_s(z) \equiv \int_0^d \frac{dz'}{l_e} c_s(z, z')$ and $\sigma_0 = \rho_0^{-1}$ being the Drude conductivity. Physically, $\bar{c}_0(z)$ and $\bar{c}_s(z)$ are the linear response functions of the charge and spin current densities to the uniform external electric field up to $O(\theta_{sh}^0)$, i.e., $\bar{c}_0(z) = \sigma_{xx}(z) \equiv j_x(z)/E_{ext}$ [33] and $\bar{c}_s(z) = Q_x^z(z)/E_{ext}$. The nonlocality of the AH effect, i.e., the spatial separation of the spin-orbit scattering and the magnetization, is clearly reflected in the structure of these integral kernels which depend on the relative distance between the current and field points as well as the distance of their center of mass coordinates from the interface. Equations (10)-(12) are the main results of this paper.

One of the most remarkable features of the nonlocal AH effect is that it appears at the first order of the spin

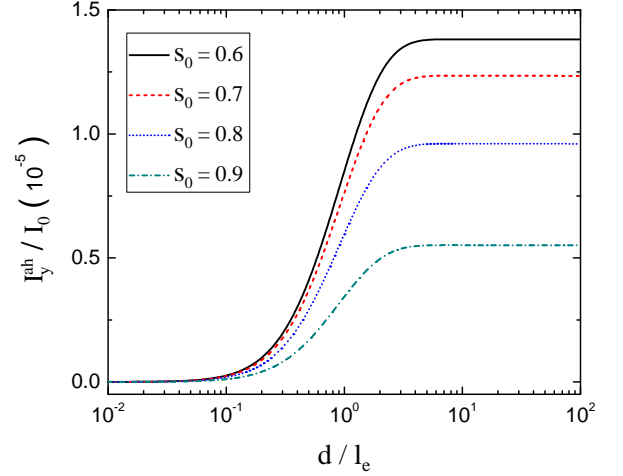


FIG. 2: The ratio of total AH current I_y^{ah} to $I_0 (= \sigma_0 E_{ext} w d)$ as a function of the thickness of the heavy metal layer for several specular reflection parameters. Other Parameters: $\theta_{sh} = 0.05$, $J_{ex} = 0.01$ eV and $V_b = 12$ eV.

Hall angle, which is distinctly different from the double spin-Hall mechanism that occurs at the second order. Since p_s is negative, the directions of the AH currents due to the nonlocal AH mechanism and the double spin-Hall mechanism would be the *same* for positive θ_{sh} but the *opposite* for negative θ_{sh} , as can be seen from Eq. (3). Furthermore, the nonlocal AH is independent of spin diffusion and thus is present in both ballistic and diffusive regimes, whereas the AH effect due to the double spin-Hall mechanism vanishes as the thickness of the metal layer becomes much smaller than the spin diffusion length [14].

The total AH current can be calculated from Eq. (10) by integrating the AH current density over the thickness of the layer, i.e., $I_y^{ah}(d) \equiv w \int_0^d dz j_y^{ah}(z)$ with w being the width of the metal bar. By doing so, we find $I_y^{ah}(d) = 2\rho_{sh} E_{ext} w \int_0^d \frac{dz'}{l_e} \bar{c}_s(z') \bar{c}_0(z')$ where the factor of 2 shows that the two physical processes that we described in Fig. 1b contribute *equally* to the total AH current. In Fig. 2, we show the thickness dependence of the total AH current for several values of the specular reflection coefficient. We find that I_y^{ah} begins to saturate when the thickness reaches the electron mean free path. Also, we note that the saturation current is smaller for a smoother surface (larger s_0), as expected from the above discussions.

Experimentally, a most relevant quantity is the ratio of the spatially averaged AH resistivity to the longitudinal resistivity, i.e., $\theta_{ah} \equiv \bar{\rho}_{xy}^{ah}(d)/\bar{\rho}_{xx}(d)$. The AH resistivity can be obtained by inverting the conductivity tensor. Since $p_s s_0 \theta_{sh} \lesssim 10^{-1}$, to a good approximation, we can take $\bar{\rho}_{xy}^{ah} \simeq \bar{\sigma}_{xy}^{ah}/\bar{\sigma}_{xx}^2$ where $\bar{\sigma}_{xy}^{ah} \equiv d^{-1} \int_0^d dz j_y^{ah}(z)/E_{ext}$

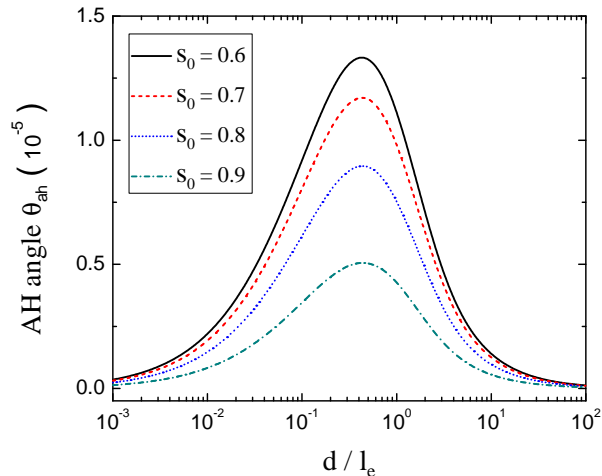


FIG. 3: The AH angle θ_{ah} ($\equiv \bar{\rho}_{xy}^{ah}(d)/\bar{\rho}_{xx}(d)$) as a function of thickness of the heavy metal layer for several values of the specular reflection coefficient. Other Parameters: $\theta_{sh} = 0.05$, $J_{ex} = 0.01$ eV and $V_b = 12$ eV.

with $j_y^{ah}(z)$ given by Eq. (10). In Fig. 3, we show the thickness dependence of θ_{ah} for several values of the specular reflection coefficient s_0 . For $d \ll l_e$, θ_{ah} tends to zero, because $\bar{\rho}_{xx}(d)$ increases with decreasing layer thickness. In the opposite limit of $d \gg l_e$, θ_{ah} also diminishes since the nonlocal AH effect is essentially an interface effect, which saturates for thicknesses larger than the electron mean path. A hint of a peak in the thickness dependence of the Hall resistivity can be found in a systematic experimental study on YIG/Pt bilayers (see Table I in Ref. [8]). By choosing the following parameters for a Pt (7 nm)/YIG bilayer at room temperature: $\theta_{sh} = 0.05$ [34], $s_0 = 0.6$, $J_{ex} = 0.01$ eV [35], $V_b = 12$ eV and $l_e = 20$ nm [18], we estimate the AH angle arising from our mechanism to be about 1.3×10^{-5} , which is in good agreement in the order of magnitude with the experimental observations [6, 8]. For the same system parameters, the spin diffusion length λ_s , which must be larger than the electron mean free path [23], is at least an order of magnitude larger than the film thickness d . In this limiting case, the AH effect due to the double spin-Hall mechanism vanishes due to the spin current continuity and the open boundary condition [14]. In the opposite limit, in which $\lambda_s \ll d$, the AH angle due to the double spin-Hall mechanism has the simple expression of $\theta_{ah}^{dsH} = \rho_0 \theta_{sh}^2 \bar{C}_{\perp}''$ with $\bar{C}_{\perp}'' \simeq G_i \left(\frac{\lambda_s^2}{d} \right)$ [21]. Taking $G_i \simeq 10^{13} \Omega^{-1} m^{-2}$ [36], $\rho_0 = 40 \mu\Omega \cdot cm$ and $\lambda_s = 1$ nm from Ref. [8], we find $\theta_{ah}^{dsH} \simeq 1 \times 10^{-6}$ which is an order of magnitude smaller than the value we calculated for the nonlocal AH effect.

As a final point, we suggest a crucial verification of our mechanism by contrasting the directions of the Hall current (or the signs of Hall voltages) of two trilayer struc-

tures Pt/Cu/YIG and β -Ta/Cu/YIG. Since the spin Hall angles of Pt and β -Ta are of opposite signs [9, 37, 38] we predict that the Hall current directions in these two trilayers will be opposite. A Cu layer, thinner than the electron mean free path, may be inserted between the heavy-metal and the magnetic insulator in order to eliminate the magnetic proximity effect, while the nonlocal AH effect will still be operative.

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