Large spin Hall magnetoresistance in easy-axis and easy-plane antiferromagnetic thin films analyzed using the complex Néel spin mixing conductance

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Abstract

Reading the magnetic state of antiferromagnetic (AFM) thin films is key for AFM spintronic devices. We investigate the underlying physics behind the spin Hall magnetoresistance (SMR) of bilayers of platinum and insulating AFM hematite (α-Fe₂O₃) and report a record SMR efficiency of up to ~1%. To understand the SMR field dependence, we analyse the role of the imaginary Néel spin-mixing conductance and find it key for the SMR response of uncompensated AFM interfaces. We demonstrate that the combined real and imaginary conductances can be used as a unique tool to unambiguously determine AFM textures.

Introduction

With spin dynamics in the terahertz regime and a robustness to external perturbations from magnetic fields, antiferromagnetic spintronics seeks to make use of this exciting ubiquitous class of materials for future spintronic devices [1,2]. A pure spin current can be generated in insulating antiferromagnets by several means, including the spin Hall effect [3,4] (SHE), the spin Seebeck effect [3,5] or spin pumping [6,7]. However, regardless of the excitation mechanism, the detection of such a pure spin current relies on the inverse spin-Hall effect in a heavy metal layer in contact with the insulating antiferromagnetic (AFMI) layer. A technique of choice is the spin-Hall magnetoresistance technique, for which a charge current $J_C$ flowing in the HM leads to a transverse spin current that flows towards the AFMI/HM interface where it is either absorbed or reflected by the AFMI. This then leads to a
modulation of the resistance of the HM as the angle between $J_C$ and the magnetic order is changed. One can thus theoretically determine the orientation of the antiferromagnetic order parameter, the Néel vector $n$, for both collinear and some non-collinear magnetic structures by means of electrical measurements [8–10] without the need for complex synchrotron-based measurements [11]. There have however been conflicting reports about SMR measurements in antiferromagnets, exhibiting so-called positive SMR and negative SMR on AFMs as well as the role of the antiferromagnetic symmetry and the interface quality.

Whilst the SMR signal is expected to depend on the compensated antiferromagnetic Néel vector direction, significant parasitic contributions can arise from both ordinary magnetoresistance (OMR) in the HM [8] and the net magnetic moment of canted antiferromagnets [12] and ferrimagnets [13] although no such influence was seen recently in a bulk canted antiferromagnet [14]. The SMR has also been investigated in AFMs with locally uncompensated interfaces, but no evidence of this is seen in the SMR signals, which are normally measured via angular dependent magnetoresistance [15]. On the theory side, antiferromagnets have been largely treated analogously to ferromagnets. However, the interaction between spin-polarized conduction electrons and the local magnetic order should be distinctly different in an AFM/HM structure with an interface with locally broken inversion symmetry around the magnetic atoms, which has previously been ignored. Furthermore, the quality and the nature of the interface between the antiferromagnet and the heavy metal is expected to impact the symmetry and the amplitude of the SMR signal, with different crystal orientations giving drastically different values for the SMR ratio in ferromagnets [16].

In this letter, we make use of the insulating antiferromagnet hematite, $\alpha$-Fe$_2$O$_3$, which exhibits a phase transition below $T_N$ from an easy-plane anisotropy to an easy-axis (EA), known as the Morin transition (MT) [17], to study the SMR for different antiferromagnetic symmetries. By varying the temperature at which the SMR is recorded, we control the antiferromagnetic spin structure and find distinctly different SMR behaviours with an extremely high SMR ratio of ~1% at low temperatures. This increase of the SMR ratio can be explained by the lowering temperature reducing fluctuations of the antiferromagnetic order that occur at the MT. It also demonstrates that spin-currents interact at least as
efficiently with compensated magnetic order (such as in antiferromagnets) as with uncompensated magnetic order (such as in ferromagnets). To explain the observed SMR below the Morin transition, we find that we cannot use conventional SMR theory. So we take into account the interface symmetry that proves critical in the spin transmission. In particular, for our growth direction, the uncompensated termination at the interface allows for an imaginary component of the Néel spin mixing conductance to appear. We observe an unconventional SMR signal through the rotation of the antiferromagnet symmetry at the Morin temperature. This is then naturally explained by taking into account the previously neglected imaginary component of the Néel spin mixing conductance.

Main Text

In order to investigate the spin Hall magnetoresistance, epitaxial (0001) oriented 100 nm thin films of hematite were deposited on like-wise orientated sapphire (α-Al2O3) via pulsed laser deposition from a stoichiometric Fe2O3 target at 800°C [11,18]. Hall bars were defined by electron beam lithography and the subsequent deposition of 7 nm platinum by DC sputtering in an argon atmosphere without additional interfacial treatments, a schematic for which is shown in Fig. 1(a). The samples were then mounted onto a piezorotating element and installed in a variable temperature insert of a superconducting magnet. A charge current $J_C$ is passed through the Pt Hall bar along the $x$-axis indicated in Fig. 1(a) whilst a longitudinal voltage is detected.
Fig. 1. (a) Schematic of the platinum Hall bar atop a thin film of (0001) oriented hematite. The crystal structure of half the unit cell of hematite is shown. The direction of the magnetic moments corresponds to the easy-plane (orange plane) antiferromagnetic phase above the Morin temperature. The red and blue arrows represent spins on the Fe atoms (light blue). A charge current is applied and a longitudinal voltage is recorded. (b) Normalized longitudinal resistance ($\Delta R_{xx} = R_{xx} - R_0$, where $R_0$ is the zero-field resistance) of the Hall bar as a function of the magnetic field applied along the three geometric axes. (c) Normalized longitudinal resistance for an in-plane rotation of the magnetic field. ($\Delta R_{xx} = R_{xx} - R_{\alpha=0}$) (d) Normalized longitudinal resistance for a magnetic field rotated in the yz plane. ($\Delta R_{xx} = R_{xx} - R_{\beta=0}$.) Error bars where visible represent the standard deviation of the measurement points.

Alongside the dominating exchange interaction giving rise to antiferromagnetic ordering, hematite also has an additional antisymmetric exchange interaction known as Dzyaloshinskii-Moriya Interaction
DMI oriented along the (0001) direction, i.e. perpendicular to the easy-plane [19]. At room temperature, this DMI has an impact on the equilibrium position of \( n \), serving to cant the magnetic moments towards one another resulting in a net magnetic moment \( m \perp n \) [17,20]. The SMR has been previously reported to be positive in some canted antiferromagnets [12], and follow the orientation of \( m \), or negative, and follow the orientation of \( n \), in others [14], showing the complex roles of the canted moment and of the interface.

We first investigate the magnetoresistance at room temperature in the easy-plane phase of the films, the antiferromagnetic structure of which is shown in Fig. 1(a). We find for the SMR (Fig. 1(b)) that when \( H//x \), there is initially a decrease of the signal before a slight and sustained increase with increasing magnetic field. This initial decrease represents the magnetic field \( H_{\text{MD}} \) required to create a monodomain, when the degeneracy of the zero-field domain structure is broken and \( n \) aligns perpendicular to \( H \) as previously reported for antiferromagnetic thin films of the easy-plane antiferromagnet nickel oxide [8,21]. If \( H \) is now directed along \( y \), there is a steep increase of the SMR ratio that plateaus at the same field value \( H_{\text{MD}} \). However, there is no further increase of the signal above \( H_{\text{MD}} \) unlike for \( H//x \).

When the magnetic field is applied out of the plane, along \( z \), \( n \) is already perpendicular to \( H \) and thus no change in the resistance is expected. However, we again observe above \( H_{\text{MD}} \) an increase of the resistance with field. To understand this first surprising field dependence, we consider possible mechanisms: Previously, OMR has been shown to have an impact on the transport signal, however, it does not account for the positive increases observed here for both \( H//x \) and \( H//z \), the absence of increase for \( H//y \) and was reported to be a full order of magnitude smaller than the SMR value reported here [8].

The increasing magnetic field can also lead to two additional effects detectable in transport measurements: a canting of the magnetic moments in the direction of the applied field, or a spin-dephasing through a Hanle effect in the platinum stripes. Given the large exchange field of hematite \((H_{\text{ex}} \sim 1000 \text{ T})\), the canting angle increases by less than 10 mrad from 0 to 10 T. As the additional increase of resistance is also present only for \( H//x \) and \( H//z \) (and not for \( H//y \)), we associate it to the Hanle effect (HMR) which has been reported to have a contribution of up to 50% in ferrimagnetic insulator (FMI)/Pt bilayers [22].
Although the behaviour of the SMR for a field along a single direction indicates that the Néel vector dominates the transport response, it does not exclude the possibility of the canted moment playing a role as it has been reported to contribute strongly to the SMR in thin films of a canted antiferromagnet [12]. To identify the governing factor for the SMR, we check the expected signals for the SMR being dominated by $n$ versus $m$. These should result in different symmetries for a rotating magnetic field, which we therefore next rotate $H$ in the $xy$-plane (Fig 1(c)). In this plane, the SMR signal shows a behaviour that increases up to $H_{MD}$ and can be modelled by a $\sin^2\alpha$ relationship between the direction of the field and the charge current and is thus again dominated by the orientation of $n$, even in the presence of a spontaneous, net moment. If instead, $H$ is rotated in the $yz$-plane (Fig. 1(d)), the signal is smooth as the angle is rotated through the in-plane direction and drops when the field is perpendicular to the easy-plane given the field is insufficient to rotate $n$ fully out of the easy-plane.

Next we check the effect of the antiferromagnetic symmetries, for which the surface sensitive nature of the SMR theoretically down to the monolayer limit is well suited [13,14], rather than other methods relying on large scale facilities [11] or are not applicable to thin antiferromagnetic films due to small signals and contributions from substrates. From SQUID magnetometry [23], the Morin transition of the films here can be measured down to films of 100 nm thickness (Fig 2(a)). If the films are thinner than this, the Morin transition is heavily suppressed or cannot be resolved [24]. Around the Morin transition there are significant thermal fluctuations as $n$ rotates along the (0001) direction and the antiferromagnetic structure transitions to that of an easy-axis anisotropy. Below $T_M$, for a magnetic field parallel to the easy-axis, there will be a classical spin flop at a critical field $H_c^\parallel$ [25]. There is also a unique spin transition in hematite, due to the DMI field, for a field perpendicular to the easy-axis at another critical field denoted $H_c^\perp$ [14,26,27]. It is well known that the magnetic anisotropies of bulk hematite have strong temperature dependencies and determine the critical fields $H_c^\parallel$ and $H_c^\perp$ as well as at the Morin transition where the critical fields drop to zero [27]. For a magnetic field both parallel ($H//z$) and perpendicular ($H//x$) to the (0001) direction, we can extract from the SMR measurements the values of these critical fields as a function of temperature. Below the Morin temperature, the values of the critical fields begin to increase (Fig 2(b)) and can be fitted by a well-known Bloch 3/2 law, $M = \ldots$
\[ M_0(1 - \frac{T}{T_M}^{3/2}) \] which is indicated by the dashed lines, resulting in a value for \( T_M \) of 210-240 K, in line with the measured Morin transitions via SQUID and other methods on similar films [11,28]. Alongside the critical fields, the SMR ratio can also be extracted at \( H_{MD} \) above \( T_M \) and at both \( H_{c\parallel} \) and \( H_{c\perp} \) below \( T_M \), shown in Fig 2(c) in line with two very recent studies, where very thin films without an accessible Morin transition were used [29,30]. We find that the behaviour at room temperature above the Morin transition is thus similar to ferromagnets. However by going below the Morin transition, a much larger SMR ratio, up to nearly 1% for \( H_{\parallel x} \) is found, far higher than previous reports on any FMI/Pt bilayers or hematite films. This increase cannot be explained through the properties of the Pt alone. However, we can explain this increase by the reduction of thermal fluctuations of the antiferromagnetic order far from \( T_M \) [9,31], showing that intrinsically hematite/Pt shows a record high SMR at low temperatures.

**Fig 2.** (a) Temperature dependence of the magnetic susceptibility of 100 nm thick (0001) orientated hematite for a field parallel to the c-axis. The Morin temperature is indicated. (b) Temperature dependence of the critical fields for \( H_{\parallel x} (H_{\parallel y}) \) (black) and \( H_{\parallel z} \) (blue). (c) Temperature dependence of the SMR ratio for \( H_{\parallel x} \) (black) and \( H_{\parallel y} \) (grey).

To understand in more detail the role of the antiferromagnetic symmetries and the differences to ferromagnets, we need to investigate the SMR below the Morin transition in-depth. At 175 K, \( n \) rotates along an easy-axis pointing out of the plane along the (0001) direction (Fig. 3(a)). For a magnetic field parallel to the direction of the uniaxial anisotropy (\( H_{\parallel z} \), i.e. \( z/EA \), Fig. 3(b)), the resistance decreases with increasing magnetic field as due to a small field misalignment the antiferromagnetic order \( n \) begin to rotate before reaching the spin-flop field, \( \mu_0 H_{c\parallel} = 6.5 \) T. As the magnetic field is lowered, a hysteretic
field dependence appears, where the critical field is shifted slightly due to the additional magnetoelastic energy present above $H^\parallel_c$ stabilising the spin-flopped state [32].

If $H$ is perpendicular to the EA ($H\parallel x$ and $H\parallel y$) $n$ will undergo a second-order transition and smoothly rotate perpendicular to both the EA and $H$. At some critical field $H^\perp_c$ [33], a transition is induced by the DMI parallel to the EA. Focusing first on the magnetic field parallel to $x$, the resistance begins to decrease until it reaches $\mu_0 H^\perp_c = 2.5 \, T$ and saturates at 0.32%, a much higher amplitude than in the easy-plane phase above $T_M$. Above $H^\perp_c$, $n\parallel/\mu_s$ at the $\alpha$-Fe$_2$O$_3$/Pt interface leads to a lower resistance state. However, we note that there is still an additional increase of the signal even though there is no further rotation of $n$ just as in the easy-plane phase, which is distinctly different from what is expected from conventional theory [34,35].

Surprisingly, an increase of the resistance is also observed for $H\parallel y$, where $n$ remains perpendicular to $\mu_s$ both below and above $H^\perp_c$ and no change in the resistance is conventionally expected. Above $H^\perp_c$, the resistance saturates unlike for $H\parallel x$ or $H\parallel z$, confirming the previous conclusion that this increase comes from HMR rather than OMR [8,22]. Whilst the change of resistance for the previous field direction ($H\parallel x$) indicates that the hematite film is antiferromagnetic even at the interface with the Pt, this latter field ($H\parallel y$) direction demonstrates that there is an additional contribution to the SMR. At $H^\perp_c$, $n$ rotates such that it remains perpendicular to $\mu_s$ at all times, failing to explain the increase of the resistance that we observe whilst the non-monotonic behaviour and the saturation show that $n$ is responsible for this signal. This is consistent with the temperature dependence of the SMR for $H\parallel y$, where we observe a reduced, as we no longer have an appropriate rotation of $n$, but persisting SMR ratio below the Morin transition.

To investigate the origin of this surprising signal, we note that, whilst hematite is a compensated antiferromagnet in general in the bulk, the (0001) growth orientation allows for an uncompensated monolayer at the interface not present in other growth orientations conventionally used [36–38]. The existence of such an uncompensated interface has previously not been considered in AFM-SMR. The
general equation for a spin current, polarised along $y$, and crossing an interface along $z$, is however then given by,

$$j_s = \frac{g_{n,r}^{(1)}}{4\pi} n \times (\mu \times n) + \frac{g_{m,r}^{(1)}}{4\pi} m \times (\mu \times m) + \frac{g_{m,i}^{(1)}}{4\pi} \mu \times m,$$

where $g_{n,r}^{(1)}$ represents the Néel spin mixing conductance in the case of a compensated interface and $g_{m,i}^{(1)}$ is the imaginary (real) part of the magnetic spin mixing conductance [34,35]. In order to explain the increasing resistance for a magnetic field applied along the $y$ direction, we introduce to the above equation a term of the form $\frac{g_{n,i}^{(1)}}{4\pi} \mu \times n$ where $g_{n,i}^{(1)}$ represents the imaginary part of the Néel spin mixing conductance. This term quantifies the precession of spins crossing the interface which is opposite for the two sublattices of an AFM and has previously been ignored on the assumption of no sublattice symmetry breaking at the interface, which is correct for many orientations but not for the one used here [8,9,34]. This term allows for a modification of the spin current transmissivity through changing the $x$ component of $n$, which occurs for $H//y$ in the (0001) hematite films (see model details in Ref. [23]). The expected SMR ratio, expressed as the change in spin current across the interface in the presence of a magnetic field $\Delta \eta = \eta - \eta_0$ (Equation (S10) of Ref [23]), of excluding or including this additional $g_{n,i}$ are shown in Figure 3(c) and 3(d) respectively demonstrating that only by taking this previously neglected symmetry breaking into account can the experimental result be reproduced.

At the pressure our films were grown (10 mTorr [11]), we would expect a coexistence of oxygen terminated and iron terminated domains with lateral sizes of a few tens of nanometers as previously shown [36]. These iron-terminated domains would then give rise to an uncompensated layer of iron at the interface. Through atomic force microscopy (AFM), we can gain a qualitative insight into the crystal terminations of our films through the size of grains indicating regions of differing terminations as previously identified in literature [23,36,39]. Thus, our qualitative assessment from AFM [23] in line with literature supports the assumption that the (0001) films terminate with multiple domains that can be both Fe or O terminated, where the Fe termination results in an uncompensated interface.
Fig. 3 (a) Schematic for a Hall bar on (0001) oriented hematite in the easy-axis phase. (b) Longitudinal resistance for a magnetic field applied along three orthogonal directions. $\Delta R_{xx} = R_{xx} - R_0$ where $R_0$ is the zero-field resistance. (c) Expected effect on the SMR ratio for three orthogonal directions providing $g_{ni}$ is zero. (d) Expected effect on the SMR ratio for three orthogonal directions when $g_{ni}$ is included with a value of $0.7 \times 10^{18}$. $H_0 = \sqrt{2JK_z}$ in both c) and d) where $J$ is the strength of the exchange interaction and $K_z$ is the uniaxial anisotropy.

In conclusion, the spin Hall magnetoresistance technique is shown to be an excellent tool for investigating antiferromagnets to understand the effects of anisotropies, symmetries and surface symmetry breaking. The surface sensitivity nature of the technique allows for the extraction of the Morin transition even for thin films of hematite where conventionally used bulk-sensitive techniques fail. We find distinctly different behaviours above and below the Morin transition, where we observe record high values for the spin Hall magnetoresistance. By probing the SMR for different directions, the effects of the uncompensated surface of the (0001) films becomes visible through a field dependence.
of the SMR that cannot be described by conventional SMR theory that does not take into account symmetry breaking. We show that this leads to the emergence of an imaginary Néel spin mixing conductance that together with its more conventional real conductance proves to be a versatile tool to ascertain antiferromagnetic spin textures. So without the need for more involved techniques accessible only at large scale facilities, we have developed a tool that allows for the necessary detection of antiferromagnetic orientations as a key step for all-electrical operation of AFMs for logic and storage devices.

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Author contributions

R.L. and M.K. proposed and supervised the project. A.R. performed the transport experiments. A.R. patterned with R.L. the samples. D. G, A. K and Av. R grew and optimized the films. A.R
and R.L. analyzed the data with input from M.K. C.U. developed the theoretical model with input from A.R. A. R wrote the paper with R.L. and M.K. All authors commented on the manuscript.

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