Large spin Hall magnetoresistance in antiferromagnetic $\alpha$-Fe$_2$O$_3$/Pt heterostructures

Johanna Fischer,$^{1,2}$ Matthias Althammer,$^{1,2}$ Nynke Vlietstra,$^{1,2}$ Hans Huebl,$^{1,2}$ Sebastian T.B. Goennenwein,$^{3,4}$ Rudolf Gross,$^{1,2,5}$ Stephan Geprägs,$^1$ and Matthias Opel$^{1}$

$^1$Walther-Meißner-Institut, Bayerische Akademie der Wissenschaften, 85748 Garching, Germany
$^2$Physik-Department, Technische Universität München, 85748 Garching, Germany
$^3$Institut für Festkörper- und Materialphysik, Technische Universität Dresden, 01062 Dresden, Germany
$^4$Center for Transport and Devices of Emergent Materials, Technische Universität Dresden, 01062 Dresden, Germany
$^5$Munich Center for Quantum Science and Technology (MCQST), 80799 Munich, Germany

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We investigate the spin Hall magnetoresistance (SMR) at room temperature in thin film heterostructures of antiferromagnetic, insulating, (0001)-oriented $\alpha$-Fe$_2$O$_3$ (hematite) and Pt. We measure their longitudinal and transverse resistivities while rotating an applied magnetic field of up to 17 T in three orthogonal planes. For out-of-plane magnetotransport measurements, we find indications for a multidomain antiferromagnetic configuration whenever the field is aligned along the film normal. For in-plane field rotations, we clearly observe a sinusoidal resistivity oscillation characteristic for the SMR due to a coherent rotation of the Néel vector. The maximum SMR amplitude of 0.25% is, surprisingly, twice as high as for prototypical ferrimagnetic Y$_3$Fe$_5$O$_{12}$/Pt heterostructures.

Despite lacking a net macroscopic magnetization, antiferromagnetic (AF) materials have moved into the focus of spintronics research [1–4]. Although L. Néel stated about 50 years ago that antiferromagnets “are extremely interesting from the theoretical viewpoint, but do not seem to have any applications” [5] this class of materials brings along two important advantages compared to ferromagnets: (i) they enable a better scalability and a higher robustness against magnetic field perturbations [13] and (ii) they offer orders of magnitude faster dynamics and thus switching times [6, 7]. From an application perspective, both switching the AF state as well as reading out the AF sublattice magnetization orientations are important challenges. It is evident that the vanishing net moment and the very small stray fields in AF materials call for new magnetization control and readout strategies.

Spin currents [8] were shown to interact with individual magnetic sublattices via spin transfer torques, also in antiferromagnets [9–12]. A particular manifestation of spin torque physics is the dependence of the resistivity of a metallic thin film with large spin-orbit coupling on the direction of the magnetization in an adjacent material with long range magnetic order, denoted as spin Hall magnetoresistance (SMR) effect [13–16]. Following earlier results in all-metallic systems [17], the SMR was first established in oxide spintronics [13] for insulating, collinear ferrimagnetic Y$_3$Fe$_5$O$_{12}$/Pt bilayers [13–15]. Upon rotating the magnetization in the magnet/metal interface plane, the SMR appears as a sinusoidal oscillation of the Pt resistivity, characterized by a specific amplitude and a phase. In compensated ferrimagnetic YGd$_2$Fe$_4$InO$_{12}$/Pt heterostructures, the pronounced temperature dependence of the SMR phase demonstrated the sensitivity of the effect to the individual canted Fe$^{3+}$ sublattice magnetizations [19]. Recently, the SMR effect was also identified in AF heterostructures. In spite of their zero net magnetization, the AF ordered magnetic sublattices contribute individually, resulting in a non-zero SMR. As the sublattice magnetizations are orthogonal to the applied magnetic field, a phase shift of 90° was reported for the SMR in NiO/Pt [20–22] and Cr$_2$O$_3$/Ta [23] as well as all-metallic PtMn/Pt [24] and PtMn/W [24] compared to that in the prototypical ferrimagnetic Y$_3$Fe$_5$O$_{12}$/Pt heterostructures. The SMR amplitude is still a matter of debate, since various extrinsic as well as intrinsic parameters play a crucial role [11, 25, 26].

In this Letter, we substantially complement the SMR data available for AF insulators by investigating $\alpha$-Fe$_2$O$_3$/Pt. We find a surprisingly large SMR amplitude of 0.25%, much higher than in AF NiO/Pt [21] and twice as large as in Y$_3$Fe$_5$O$_{12}$/Pt [15]. This finding supports the picture that each magnetically ordered sublattice contributes independently, regardless of the material’s net magnetization. The large SMR amplitude together with a moderate saturation field of ~3 T establishes $\alpha$-Fe$_2$O$_3$/Pt as a viable future SMR source and paves the way towards room temperature antiferromagnetic spintronic applications.

The electrical insulator $\alpha$-Fe$_2$O$_3$ (hematite) crystallizes in a rhombohedral structure and can be described in the hexagonal system with the lattice constants $a = 0.5032$ nm and $c = 1.3748$ nm [27]. In bulk, it exhibits a Néel temperature of $T_N = 953$ K and undergoes a spin reorientation (“Morin” transition) at $T_M \approx 263$ K [28]. For $T_M < T < T_N$ and in the absence of an external magnetic field, the $S = 5/2$ spins of the Fe$^{3+}$ ions are ordered ferromagnetically in the (0001) planes. Along the crystallographic [001] direction, these easy planes form a “++−−−+” sequence, resulting in a net AF order [29].
reflections. The reciprocal lattice direction, the longitudinal (rlu) direction, and the magnetic field magnitude of only 0°.

A finite anisotropic spin-spin ("Dzyaloshinskii-Moriya") interaction leads to a small canting of the two AF sublattice magnetizations \( M_1 \) and \( M_2 \) with a canting angle of 0.13° ± 0.01°. This results in a small net magnetization \( M = M_1 + M_2 \) in the (0001) plane. Similar to the situation in NiO, \( \alpha \)-Fe\(_2\)O\(_3\) displays three AF domains rotated by 120° with respect to each other and a domain population dependent on the direction and magnitude of the external magnetic field. The monodomainization field \( \mu_0 H_{\text{MD}} \) is reported to be above 600 mT.

Since thin films are key for applications, we here study \( \alpha \)-Fe\(_2\)O\(_3\)/Pt bilayer heterostructures, fabricated on single crystalline, (0001)-oriented \( \text{Al}_2\text{O}_3 \) substrates. First, we deposit epitaxial \( \alpha \)-Fe\(_2\)O\(_3\) thin films at a substrate temperature of 320°C in an oxygen atmosphere of 25 μbar via pulsed laser deposition. Without breaking the vacuum, the films are covered in-situ by thin layers of Pt via electron beam evaporation. High-resolution X-ray diffractometry (HR-XRD) measurements reveal a high structural quality of the \( \alpha \)-Fe\(_2\)O\(_3\)/Pt heterostructures. The \( 2θ-ω \) scan (Fig. 1(a)) shows only reflections from the epitaxial \( \alpha \)-Fe\(_2\)O\(_3\) thin film, the Pt layer, and the \( \text{Al}_2\text{O}_3 \) substrate. No secondary crystalline phases are detected. A broad feature below the \( \alpha \)-Fe\(_2\)O\(_3\) (0006) reflection (grey shaded area) can be assigned to Pt(111) expected at 39.8° and points to a textured nature of the Pt top electrode. On an enlarged scale (upper inset in Fig. 1(a)), satellites due to Laue oscillations are detected around the \( \alpha \)-Fe\(_2\)O\(_3\) (0006) reflection, evidencing a coherent growth with low interface roughness of the \( \alpha \)-Fe\(_2\)O\(_3\) thin film. The asymmetry on both sides of the \( \alpha \)-Fe\(_2\)O\(_3\) (0006) reflection is caused by interference with the broad Pt(111) reflection. Furthermore, \( \alpha \)-Fe\(_2\)O\(_3\) shows a low mosaic spread as demonstrated by the full width at half maximum of the rocking curve around the \( \alpha \)-Fe\(_2\)O\(_3\) (0006) reflection of only 0.027° (lower inset in Fig. 1(a)). They reveal the epitaxial relations \( [0001]_{\alpha \text{-Fe}_2 \text{O}_3} || [0001]_{\text{Al}_2 \text{O}_3} \) and \( [10\bar{1}0]_{\alpha \text{-Fe}_2 \text{O}_3} || [10\bar{1}0]_{\text{Al}_2 \text{O}_3} \). We derive lattice constants of \( a = 0.505 \text{ nm} \) and \( c = 1.372 \text{ nm} \) very close to the respective bulk values, indicating a nearly fully relaxed strain state for our \( \alpha \)-Fe\(_2\)O\(_3\) films. Furthermore, a low surface roughness below 1.1 nm (rms value) is confirmed by X-ray reflectometry as well as atomic force microscopy (not shown here). In summary, our \( \alpha \)-Fe\(_2\)O\(_3\)/Pt bilayer is of the same high structural quality as the prototypical ferrimagnetic \( \text{Y}_3\text{Fe}_2\text{O}_{12}/\text{Pt} \) heterostructures reported earlier.

In the following, we discuss an \( \alpha \)-Fe\(_2\)O\(_3\)/Pt bilayer sample with thicknesses of \( t_{\text{Fe}_2\text{O}_3} = 90 \text{ nm} \) and \( t_{\text{Pt}} = 3 \text{ nm} \). For transport measurements, a Hall bar-shaped mesa structure with a width of \( w = 80 \mu\text{m} \) and a longitudinal contact separation (length) of \( l = 600 \mu\text{m} \) was patterned into the bilayer via photolithography and Ar ion milling (Fig. 2). For a dc current of ±100 μA applied in the [10\bar{1}0] direction, the longitudinal \( (ρ_{\text{long}}) \) and the transverse \( (ρ_{\text{trans}}) \) resistivities are measured in a standard four-probe configuration. A current-reversal method is applied to eliminate thermal effects. We restrict our investigation to room temperature, where the (0001) plane is a magnetic easy plane. We perform angle-dependent magnetoresistance (ADMR) measurements by rotating an external magnetic field of constant magnitude \( H \) in three different orthogonal planes of the (0001)-oriented \( \alpha \)-Fe\(_2\)O\(_3\) using the same notation as in Ref. 15 and Fig. 2(a-c): (0001) = "ip" (in-plane, angle \( α \), black);
The data are well described by

$$\rho_{\text{long}} = \rho_0 + \frac{\rho_1}{2} (1 - \cos 2\alpha)$$

$$\rho_{\text{trans}} = -\frac{\rho_3}{2} \sin 2\alpha$$

(black line in Fig. 2(d)) as demonstrated earlier for AF NiO/Pt [21]. However, the SMR amplitude of 0.25% for $\alpha$-Fe$_2$O$_3$/Pt is more than a factor of 3 higher than for NiO/Pt and, remarkably, even twice as large as for the prototypical ferrimagnetic Y$_3$Fe$_2$O$_{12}$/Pt heterostructures with similar Pt thickness [15]. In fact, it is larger than for any other reported bilayer compound so far.

The ADMR of the out-of-plane rotations is qualitatively different. For oopj rotations of $\mathbf{H}$, we observe $\rho_{\text{long}}$ in the maximum resistive state for a wide range of angles $\beta$ around $\mathbf{H} \parallel \pm \mathbf{t}$ (blue squares in Fig. 2(d)), indicating a monodomain state with $\ell \parallel \pm \mathbf{j}$. For the oopt geometry, on the other hand, $\rho_{\text{long}}$ stays in the minimum resistive state for a wide range of angles $\gamma$ around $\mathbf{H} \parallel \pm \mathbf{n}$ (green triangles in Fig. 2(d)), indicating again a monodomain state with $\ell \parallel \pm \mathbf{t}$. Both observations show that $\ell$ does not follow $\mathbf{H}$ for out-of-plane rotations. $\rho_{\text{long}}$ changes significantly only close to $\mathbf{H} \parallel \pm \mathbf{n}$: both oopj and oopt curves meet for magnetic fields perpendicular to the sample surface at the midpoint of the two states with extremal resistances. According to the SMR model for a multidomain antiferromagnet [21], we interpret this observation with the “decay” of a monodomain into a three-domain state when $\mathbf{H}$ points orthogonal to the magnetic easy (0001) plane of $\alpha$-Fe$_2$O$_3$, in agreement with a recent preprint [37].

To obtain further insight into the AF domain configurations, we perform ip ADMR measurements of both $\rho_{\text{long}}$ and $\rho_{\text{trans}}$ at different magnitudes of the magnetic field from 10 mT to 17 T (Fig. 3). The expected $(-\cos 2\alpha)$ and $(-\sin 2\alpha)$ dependencies of $\rho_{\text{long}}$ and $\rho_{\text{trans}}$, respectively, are clearly observed for $\mu_0 H \geq 300$ mT (Fig. 3(b-e)). This angular dependence is fully consistent with the model introduced earlier for NiO/Pt [21] and Eq. (1) and clearly shows that our $\alpha$-Fe$_2$O$_3$ is AF with the resistivity of Pt being sensitive to $\ell$, which rotates coherently in the easy (0001) plane perpendicular to $\mathbf{H}$. The data is further fully consistent with recent experiments in Pt on canted ferrimagnets, where the same angular dependence is observed close to the compensation temperature [19], as well as experiments in Y$_3$Fe$_2$O$_{12}$/NiO/Pt [38, 41] and NiO/Pt [20, 22]. For $\mu_0 H \lesssim 100$ mT, the applied field is smaller than $\mu_0 H_{\text{MD}}$, resulting in hardly detectable SMR oscillations (Fig. 3(a)).

For a detailed analysis of the field dependence of $\rho_{\text{long}}$ and $\rho_{\text{trans}}$, we fit our data analogous to Eq. (1) using cos $2\alpha$ and sin $2\alpha$ functions, respectively, (solid lines in Fig. 3 and plot the SMR amplitudes $\rho_{\text{long}}$ and $\rho_{\text{trans}}$ (double arrows in Fig. 3(e)) as a function of the magnetic field magnitude in Fig. 4). Remarkably,
SMR\text{trans} exceeds SMR\text{long} for fields above 100 mT. We attribute this unexpected observation to the presence of 180° domain walls in α-Fe\textsubscript{2}O\textsubscript{3}. These domain walls may either be separated by a distance larger than the transverse contact width of the Hall bar or be located at pinning sites created by the patterning process along the length of the Hall bar, effectively reducing the SMR in the longitudinal voltage, but not in the transverse one. Furthermore, the field evolution of the SMR amplitude is qualitatively different from the one in AF NiO/Pt [21]. In α-Fe\textsubscript{2}O\textsubscript{3}/Pt, we find that both SMR\text{long} and SMR\text{trans} saturate already around 3 T and then gradually decrease again from 5 T to 17 T. This gradual decrease can be traced back to an increasing canting of the AF sublattices thus reducing the value of ℓ and an emerging non-zero net M [42]. The fast saturation, on the other hand, points to a lower destressing energy compared to NiO. Applying the same mean field simulation [21], we approximate the low-field data with a quadratic function in \( H \)

\[
\text{SMR}_{\text{long,trans}} = \frac{\rho_{1.3}}{\rho_0} \frac{H^2}{H_{\text{MD}}} \quad (2)
\]

(the green line in the inset of Fig. 4 shows the simulation based on SMR\text{long}) and obtain a monodomainization field of \( \mu_0 H_{\text{MD}} = 240 \) mT. With an exchange field of \( \mu_0 H_{\text{ex}} = 900 \) T [32] [13], we derive a destressing field of \( \mu_0 H_{\text{dest}} = \mu_0 H_{\text{MD}}^2/(4H_{\text{ex}}) \approx 0.016 \) mT, drastically smaller than the 46 mT in epitaxial NiO thin films on Al\textsubscript{2}O\textsubscript{3} [21]. We note that \( H_{\text{MD}} \) highly depends on the strainability of the specific sample and is expected to be larger for thin films than for bulk [21]. We further note that \( \mu_0 H_{\text{MD}} = 240 \) mT, being smaller than reported for single crystals [44], results as a parameter from a mean field simulation of our low-field data. Our α-Fe\textsubscript{2}O\textsubscript{3} thin film does not become single domain at 240 mT, but at a higher field of \( \approx 3 \) T where the SMR amplitude starts to saturate (Fig. 4). With \( \mu_0 H_{\text{MD}} = 3 \) T, we derive \( \mu_0 H_{\text{dest}} \approx 2.5 \) mT. This value is reasonable, since the magnetostriction \( \lambda = 4 \times 10^{-6} \) in the basal plane of α-Fe\textsubscript{2}O\textsubscript{3} at 293 K [44] is by a factor of \(~20\) smaller than \( \lambda = (9 \pm 3) \times 10^{-5} \) in NiO [45].

In summary, we present a detailed investigation of the SMR in antiferromagnetic α-Fe\textsubscript{2}O\textsubscript{3}/Pt heterostructures at room temperature studying three orthogonal rotation planes of the magnetic field. We consistently describe the angular dependence of the data in a three-domain model, considering a field-dependent canting of the AF sublattices. Our data supports the picture that each magnetic sublattice contributes separately to the SMR. Surprisingly, we find a large SMR amplitude of 0.25%. This value well exceeds the established values for Y\textsubscript{3}Fe\textsubscript{5}O\textsubscript{12}/Pt or any other Pt-based thin film heterostructures reported in literature so far. AF materials are therefore expected to play an important role in SMR-related research and applications. Due to the small destressing field, the SMR amplitude reaches 0.20% (corre-
sponding to \( \frac{2}{5} \) of its maximum value) already at 300 mT, i.e. at much smaller magnetic fields as in comparable AF NiO/Pt heterostructures [21]. This combination of high sensitivity at low magnetic fields and room temperature operation makes \( \alpha \)-FeO\(_3\)/Pt a promising material system both for a viable SMR source and future spin transfer torque based devices and applications in the emerging field of antiferromagnetic spintronics.

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