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Negative spin Hall magnetoresistance of Pt on the bulk easy-plane antiferromagnet NiO

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We report on spin Hall magnetoresistance (SMR) measurements of Pt Hall bars on antiferromagnetic NiO(111) single crystals. An SMR with a sign opposite to conventional SMR is observed over a wide range of temperatures as well as magnetic fields stronger than 0.25 T. The negative sign of the SMR can be explained by the alignment of magnetic moments being almost perpendicular to the external magnetic field within the easy plane (111) of the antiferromagnet. This correlation of magnetic moment alignment and the external magnetic field direction is realized just by the easy-plane nature of the material without the need of any exchange coupling to an additional ferromagnet. The SMR signal strength decreases with increasing temperature, primarily due to the decrease in Néel order by including fluctuations. An increasing magnetic field increases the SMR signal strength as there are fewer domains, and the magnetic moments are more strongly manipulated at high magnetic fields. The SMR is saturated at an applied magnetic field of 6 T, resulting in a spin-mixing conductance of ~10^18 Ω⁻¹ m⁻², which is comparable to that of Pt on insulating ferrimagnets such as yttrium iron garnet. An argon plasma treatment doubles the spin-mixing conductance. Published by AIP Publishing. [http://dx.doi.org/10.1063/1.4997588]

Antiferromagnets (AFMs) are mostly known for their exchange bias pinning effect on adjacent ferromagnetic (FM) layers. Owing to the robustness against magnetic perturbations of easy-plane AFMs, this coupling allows for giant superparamagnetic effects.1,2 and tunneling magnetoresistance devices. More recently, metallic AFMs have been manipulated and read out by applied spin polarized charge currents.4 Insulating AFMs can have spin waves carrying spin angular momentum,5–7 which switch ultrafast8 and act as efficient spin-current transmitters,6,9 thus playing an important role in spintronic applications.10

Injection and detection of spin angular momentum in insulating magnets can be done by the combination of the spin Hall effect (SHE)11 and the inverse spin Hall effect (ISHE)12 in normal metals (NMs). Rotating the magnetic moments in the adjacent magnet by an applied magnetic field can change the interaction of the Pt spins with the magnet. This leads to spin Hall magnetoresistance (SMR),13–15 which enables the study of various magnetic systems. In collinear ferrimagnets,13–15 the magnetic moments align collinear to the external magnetic field, resulting in positive SMR contributions. A reversed angular modulation or negative SMR signal has been observed when the average canting angle between the magnetic moments and the external magnetic field exceeds 45° in canted magnetic systems16 and spin spirals.17 The localized spins of bulk AFMs in an easy plane are nearly perpendicular to the applied magnetic field.18 The perpendicular alignment is expected to create a negative SMR due to the 90° angle shift, but this effect has not yet been studied in detail.

Spin-transfer measurements through insulating AFMs have been studied using stacked Pt/NiO/YIG (yttrium iron garnet) devices. Magnons are created in YIG by ferromagnetic resonance19,20 or the spin Seebeck effect,21,22 propagate through the NiO layer, and are detected in Pt by the ISHE. For NiO layers thicker than ~5 nm, the transmitted spin current decreased rapidly with thickness. The sign of the SMR signal in these Pt/NiO/YIG stacks is observed to be positive at room temperature and becomes negative at low temperatures.23,24 The authors explain this domination of the positive SMR at room temperature by spin currents injected at the Pt/NiO interface, transmitted through NiO, and partly reflected when entering the YIG. At low temperatures, the spin currents towards and from the YIG are suppressed due to the vanishing spin transmittance in NiO, and thus, the total signal is dominated by the negative SMR from NiO. For the Pt/NiO/YIG samples, the NiO magnetic moments are indirectly aligned perpendicular to the magnetic field via an exchange coupling with YIG, which is saturated at 0.06 mT.23,24

In this letter, SMR signals are obtained from Pt/NiO heterostructures by direct manipulation of the AFM spins in the magnetic easy plane with an applied magnetic field and without the need of any exchange coupling to an additional ferromagnetic ferrimagnet. The surface of the studied NiO bulk single crystal has a (111) cut so that the Pt/NiO interface has an easy plane of the NiO magnet. A strong magnetic field will align the moments perpendicular to the magnetic field direction due to Zeeman energy reduction, aside from contributions due to magnetic anisotropy or domain formation by magnetostriiction. Therefore, by rotating the magnetic field, the magnetic moments follow the rotation with almost a 90° angle shift within this magnetic easy plane.18
Figure 1(a) shows the atomic face centered cubic unit cell of NiO. The superexchange interaction between Ni$^{2+}$ ions mediated by O$^{2-}$ ions aligns the Ni$^{2+}$ magnetic moments antiparallel. Below the Néel temperature of 523 K, the total interaction causes the spins to have their preferential orientation in one of the (111) planes. Magnetostriction creates rhombohedral distortions in the diagonal orientation in one of the {111} planes. Magnetostriction creates the total interaction causes the spins to have their preferential orientation in one of the {111} planes. However, the spin angular momentum can be transferred to imaginary components of the spin-mixing conductance.

In a NM with large spin-orbit coupling, the electrons deflect in a direction depending on their spin orientation, resulting in a spin current perpendicular to the charge current—known as SHE. Since NiO is an insulator, a vertical spin polarized charge current in the NM results in a spin accumulation at the interface, which is shown in Fig. 1(b). However, the spin angular momentum can be transferred to NiO when the magnetic moments of NiO are perpendicularly aligned to the accumulated spins. Figure 1(c) shows the spin transfer of perpendicular ($\mu_{\perp}$) and the reflection of collinear ($\mu_{\parallel}$) components of the spin accumulation.

The ISHE converts the reflected collinear component into charge current as shown in Fig. 1(d). The spin transfer depends on the microscopic interaction of the spin accumulation with NiO and can reduce the back-flow of the spin current depending on the direction of the magnetic moments of NiO at the interface. The changes in reflected spin current and, thus, in the resistivity of the NM can be measured both longitudinally and transversally.

The spin transfer through the interface is given by:

$$J_s = \frac{G_s}{4\pi} \mathbf{n} \times \mathbf{\mu} \times \mathbf{n} + \frac{G_i}{4\pi} \mathbf{\mu} \times \mathbf{n},$$

where $\mathbf{n}$ is the Néel vector and $G_s$ and $G_i$ are the real and imaginary components of the spin-mixing conductance $G_{\parallel}$, respectively. The first part of Eq. (1) containing $G_s$ is governed by Umklapp reflections, and the second part containing $G_i$ is associated with specular reflections similar to the ferromagnetic case.6,25

AFMs can be described by the Néel vector $\mathbf{n} = (\mathbf{m}_A - \mathbf{m}_B)/2$ instead of the total magnetic moment $\mathbf{m} = (\mathbf{m}_A + \mathbf{m}_B)/2$. Using the exchange approximation $\mathbf{n} \gg \mathbf{m}$, we obtain $n^2 = n^2 + \mathbf{m}^2 = \mathbf{m}_A^2 + \mathbf{m}_B^2$ for the SMR description. Still, a small canting of the moments lowers the Zeeman energy and aligns the NiO magnetic moments almost perpendicular to the applied in-plane magnetic field. Therefore, the magnetic field couples to the resulting but quite small total magnetic moment. The magnetic moment directions of $\mathbf{m}_A$ and $\mathbf{m}_B$ and, thus, the Néel vector $\mathbf{n}$ follow the magnetic field with an angle shift of about 90°.

The resulting changes in the longitudinal and transverse SMR ($\rho_L$ and $\rho_T$, respectively) are adapted from the regular ferromagnetic SMR equations14,15 to

$$\rho_L = \rho + \Delta \rho_0 + \Delta \rho_1 (1 - n^2),$$

$$\rho_T = \Delta \rho_1 (\mathbf{n} \cdot \mathbf{n}) + \Delta \rho_{Hall} B_z,$$

where $\rho$ being the electrical resistivity of Pt and $\Delta \rho_{Hall} B_z$ the change in resistivity caused by the ordinary Hall effect with an out-of-plane component of the magnetic field $B_z$, $\mathbf{n}$ and $\mathbf{n}$ are the x- and y-components of the Néel vector. Due to the quadratic dependence on $\mathbf{n}$, resistivity changes are equal for the two sublattices with the rotation of the magnetic field. $\Delta \rho_0$ and $\Delta \rho_1$ are resistivity changes defined as14

$$\frac{\Delta \rho_0}{\rho} = -\theta_{SH}^2 \frac{2\lambda}{2\sigma} \left( \frac{\lambda}{\lambda} \right),$$

$$\frac{\Delta \rho_1}{\rho} = \theta_{SH}^2 \frac{\lambda}{2\sigma} Re \left( \frac{2\lambda G_{\parallel}}{2\sigma} \frac{d\lambda}{d\lambda} \right),$$

where $\lambda$, $d\lambda$, $\sigma$, and $\theta_{SH}$ are the spin relaxation length, thickness, bulk conductivity, and the spin Hall angle in the NM, respectively. $G_{\parallel}$ is the spin-mixing conductance of the NM/(A)FM interface.

To investigate the effect of Ar-sputtering on the spin-mixing conductance between Pt and NiO, we etched one of the two used bulk single NiO crystals. The two crystals investigated are black colored due to vacancies with the dimensions of $5 \times 5 \times 1$ mm$^3$. For the device fabrication, the crystals are polished along the (111) surface in the line of the technique described by Aqeel et al.26 The crystals have been ground (SiC P4000) and polished (diamond 6 µm and 3 µm, silica 0.04 µm, and AlOx 0.02 µm). To remove residuals of polishing, the samples are rinsed (hot water, ethanol), blow dried, rinsed again (propanol), and annealed at 200°C. The peak-to-peak surface roughness is 0.24 nm as determined by atomic force microscopy shown in Fig. 2(a).

A Pt Hall bar with a $100 \times 1000$ µm$^2$ main bar and four longitudinally 753 µm separated, $100 \times 20$ µm$^2$ Pt side contacts was patterned by e-beam lithography as can be seen in Fig. 2(b). 5 nm thick Pt is sputtered at a base pressure of 2.5 × 10$^{-7}$ mbar and a sputter pressure of 4.9 × 10$^{-7}$ mbar.
The etched sample was given an additional 15 s argon plasma exposure at 200 W. The effect on the roughness could not be checked as the etching occurred in situ before sputtering the Pt Hall bar.

The crystallographic (111) surfaces are confirmed by x-ray diffraction with a crystallographic data based fit as can be seen in Fig. 2(c) for the unetched sample. The magnetic moment is nearly a linear function of the magnetic field as is shown by the SQUID measurements in Fig. 2(d). The effect of etching on the magnetic moment is negligible; the SQUID curves of the samples overlap as the etching was done on a relatively small area and affects only the top atomic layers.

The electrical connections for the longitudinal and transverse measurements are shown in Fig. 2(b). An AC source of 1 mA was used with a frequency of 17.777 Hz. The SMR signals were separated from any heating effects by measuring the first harmonic signal using the lock-in technique.

Figure 3(a) shows the relative change in resistivity and the corresponding change in resistance (0.25 × 10⁻³ and 124 mΩ for 8 T, respectively) of the longitudinal SMR at room temperature. The resistivity is minimal when the accumulated spins and the magnetic moments are collinear since the interface electrons are deflected by the ISHE into the direction of the current at the corresponding field angles, 0° and 180°. At other field angles, there is a spin current into NiO, which decreases the spin accumulation and increases the resistivity as sin α.

Furthermore, the spin transfer alters the spin direction of the accumulated electrons. This affects the direction of the ISHE mediated electron deflection, creating transverse deflection and more scattering of electrons in the longitudinal geometry. Since the change in the spin direction is a function of its relative direction with the NiO magnetic moments, this also leads to an increase in resistance as sin α. The combination of the two angular dependencies causes the observed modulation of the resistance $R_{\alpha,xx} \propto \sin^2 \alpha$ in the longitudinal geometry.

In FMs, however, the longitudinal resistivity is maximal when the magnetic field and the spin accumulation are perpendicular since the magnetic moments coherently follow the applied magnetic field. Therefore, a phase shift of 90° arises in the angular dependence of the SMR of an AFM as compared to a FM. The $\cos^2 \alpha$ angular dependent SMR of a FM changes into $\cos^2(\alpha - 90°) = \sin^2 \alpha = 1 - \cos^2 \alpha$ for an AFM. The modulation has changed from a positive to a negative $\cos^2 \alpha$, giving reason to call it a negative SMR.

**FIG. 2.** Characterization of the samples. (a) Atomic force microscopy data giving the height profile and roughness of the sample. (b) Optical microscopy image of the Pt Hall bar with Ti/Au contacts on top of the NiO(111) crystal. Black lines indicate the connections for the voltage measurements in the longitudinal ($V_{xx}$) and transverse ($V_{xy}$) configuration. (c) X-ray diffraction measurement with data (black dots) and fit (red line) confirming that the reflection peak is at an angle corresponding to (111) crystallographic surfaces. (d) SQUID measurement showing an almost linear dependence of magnetic moment on an applied magnetic field and overlapping graphs for the two samples.

**FIG. 3.** SMR signals in the (a) longitudinal and (b) transversal geometry, obtained at 300 K on the unetched sample. The right axes show the change in resistance $R_\alpha - R_0$, with $R_\alpha$ being the angular dependence and $R_0$ the constant resistance; $R_{ax,xx} = 460$ Ω and $R_{ax,xy} = 0.685$ Ω for the longitudinal and transverse geometry, respectively. The left axes show the relative change in resistivity, where $\Delta \rho / \rho = (\rho_\alpha - \rho_0) / \rho_0 = (R_{ax,xx} - R_0) / R_{ax,xx}$ (or $R_{ax,xy} / R_{ax,xx}$) in the longitudinal (transverse) direction and 7.53 is the geometric conversion factor.
Since a transverse deflection creates a voltage difference in the transverse geometry as \(-\cos x\), the angular dependence in the transverse geometry is \(R_{\text{xy}} \propto -\cos x \times \sin x\) with \(0.25 \times 10^{-3}\) and 15 mΩ for 8 T, respectively) are measured as shown in Fig. 3(b). These transverse results agree with the longitudinal relative change in resistivity taking the geometric conversion factor of 7.53 into account, which is the ratio between the length and the width of the main Hall bar segment. The peak to peak changes of the angular dependent resistivity parts in Eqs. (2) and (3) match up to an average factor of 0.95 ± 0.06. This means that for both the transverse and the longitudinal case, \(\Delta \rho_{\text{LY}}\) is equal and there is no difference in spread as a result of domain formation, anisotropy, or spin-mixing conductance. For the etched sample, however, this ratio is 0.85 ± 0.02. This is most likely due to a local variation in spin-mixing conductance, creating a difference in the signal size between the longitudinal and transverse SMR measurements.

Besides the SMR signal, there is also a 360° period Hall contribution in the transverse geometry, which originates from a slight misalignment of the sample, resulting in a small out-of-plane component of the magnetic field. A fit shows that the signal is one order of magnitude lower than the SMR signal, as can be seen in Figs. 4(a)–4(c). The Hall component of both samples increases linearly with magnetic field strength as expected. However, the Hall contribution is of both samples increases linearly with magnetic field strength on the (a) unetched and (b) etched sample at 5 K. (c) The relative SMR signal as a function of temperature for the etched sample obtained at 4 T

The background resistivity increases linearly with increasing temperature as expected for a metal. The relative SMR signal strength [Fig. 4(c)], however, decreases with temperature for the etched sample and is fitted with \(\propto (T_N - T)^{0.7}\). This expression for the square of the order parameter, depending on \(\propto (T_N - T)^{0.35}\), is obtained by a mean-field-like approach, including small fluctuations around the average magnetization.\(^{30}\) The obtained parameters of Néel temperatures are 551 ± 16 K and 531 ± 25 K for the longitudinal and transverse SMR signals, respectively, and compare relatively well with the established value of 523 K.

The spin-mixing conductance is \(\sim 10^{18} \Omega^{-1} \text{m}^{-2}\) as calculated from Eq. (5) for the unetched sample with \(\lambda, d_N\), and \(\theta\) assumed being constants of 1.1 nm, 5 nm, and 0.08, respectively.\(^{15}\) All fabrication steps of both samples are the same, except for the argon plasma treatment. Therefore, the magnitude of the SMR signal must originate from an enhancement of the spin-mixing conductance by a factor of two. So, the etching significantly increases the interface transparency. These spin-mixing conductance results are comparable to the SMR results of heterostructures with ferrimagnets such as Pt/YIG.\(^{31}\) Recently, an increase in the spin-mixing conductance after an etch step is observed in Pt/YIG systems as well, where it is further improved by an annealing step.\(^{32}\)

Based on the bulk exchange coupling, the spin-mixing conductance is expected to be smaller than that of ferromagnets. A possible explanation for the high spin-mixing conductance in NiO is a lowered surface exchange coupling compared to the bulk.\(^{7}\) The results agree with the theoretical prediction of increased spin-mixing conductance due to the interface disorder.\(^{6}\) The change in spin-mixing conductance due to the argon plasma treatment supports this theory as it affects the surface.

An attempt to detect SMR in Pt/AFM bilayers has already been reported by Han et al., where the SMR of Pt on bulk AFM SrMnO\(_3\) had a positive sign.\(^{33}\) Taking our results into account, their study is evidence that the SMR signals observed there are not due to an antiferromagnetic alignment but rather due to spin canting with respect to the magnetic field direction.

In summary, SMR for Pt/NiO(bulk) heterostructures with a (111) oriented surface has been studied. Negative SMR is observed at fields higher than 0.25 T without the need of exchange coupling to an additional ferromagnet. The sign of the SMR indicates that the applied magnetic field couples

\[\Delta \rho_{\text{xy}} = a \cos \theta \sin \theta,\]

where the SMR of Pt on NiO has been reported by Han et al., where the SMR of Pt on bulk AFM SrMnO\(_3\) had a positive sign.\(^{33}\) Taking our results into account, their study is evidence that the SMR signals observed there are not due to an antiferromagnetic alignment but rather due to spin canting with respect to the magnetic field direction.

In summary, SMR for Pt/NiO(bulk) heterostructures with a (111) oriented surface has been studied. Negative SMR is observed at fields higher than 0.25 T without the need of exchange coupling to an additional ferromagnet. The sign of the SMR indicates that the applied magnetic field couples
perpendicular to the magnetic moments. The SMR signal increases by decreasing the temperature, which is attributed to the lowering of the Néel order and its spatial fluctuations. The relative change of resistivity in the transverse geometry agrees with the longitudinal geometry in the sample without argon plasma treatment and follows $R_{xy} \propto -\frac{1}{2} \sin 2\alpha$ and $R_{xx,xy} \propto \sin^2 \alpha$ angular dependences, respectively.

A maximum SMR signal is observed at around 6 T with a corresponding spin-mixing conductance of $\sim 10^{18} \, \Omega^{-1} \, m^{-2}$, which is comparable to that of YIG. SMR proves to be an effective technique to investigate and manipulate the magnetic properties of AFMs. The simultaneous electrical injection and detection of spin currents, while having control over the magnetic moment directions, open up the possibility of ultrafast and lossless AFM transport devices.

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