Determination of the spin Hall angle in single-crystalline Pt films from spin pumping experiments

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Abstract
We report on the determination of the spin Hall angle in ultra-clean, defect-reduced epitaxial Pt films. By applying vector network analyzer ferromagnetic resonance spectroscopy to a series of single crystalline Fe (12 nm)/Pt (tPt) bilayers we determine the real part of the spin mixing conductance \( (4.4 \pm 0.2) \times 10^{-19} \text{ m}^{-2} \) and reveal a very small spin diffusion length in the epitaxial Pt (1.1 \( \pm \) 0.1) nm film. We investigate the spin pumping and ISHE in a stripe microstructure excited by a microwave coplanar waveguide antenna. By using their different angular dependencies, we distinguish between spin rectification effects and the inverse spin Hall effect. The relatively large value of the spin Hall angle (5.7 \( \pm \) 1.4)% shows that ultra-clean e-beam evaporated non-magnetic materials can also have a comparable spin-to-charge current conversion efficiency as sputtered high resistivity layers.

1. Introduction
In spintronics, bilayers composed of a ferromagnetic (FM) and a nonmagnetic (NM) layer with large spin-orbit-interaction are used to investigate spin-to-charge current conversion. At ferromagnetic resonance (FMR) the spin pumping (SP) effect results in the injection of a pure spin current into the NM layer [1] and the inverse spin Hall effect (ISHE) generates a charge current by spin-dependent deflection [2]. For magnon spintronics the spin-to-charge current conversion may play an important role for its success since it can define the interface to common CMOS technology. Many aspects of the SP effect using FM metals [3] and insulators [4] with Pt capping or Py with varying non-magnetic layers [5, 6] as well as metallic [7] and dielectric interlayer [8] materials have been investigated.

The parameter describing the efficiency of the spin-to-charge current conversion is the spin Hall angle \( \Theta_{\text{SH}} \). It is defined as the ratio of the measured charge current deflected by ISHE in the NM material and the spin current injected into the NM material. Another parameter, strongly correlated to the spin Hall angle, is the spin diffusion length \( \lambda_{\text{SD}} \). \( \lambda_{\text{SD}} \) describes the typical length scale in which the spin current is attenuated after its injection from the FM into the NM material. This attenuation in the NM is originates from spin-flip processes and from the defection of the spin current caused by the ISHE. For the commonly used NM material Pt, \( \Theta_{\text{SH}} \) and \( \lambda_{\text{SD}} \) have a wide span in the literature, namely from 1% up to 11% and from under 1 nm to roughly 10 nm, respectively [4–6, 9–16]. If the depolarization of the spin current by spin memory loss can be neglected, both parameters, \( \Theta_{\text{SH}} \) and \( \lambda_{\text{SD}} \), can show a reciprocal behavior [10]: a small \( \lambda_{\text{SD}} \) is then connected to a large \( \Theta_{\text{SH}} \) and vice versa. Often, this correlation can be seen in the literature. The experimental finding of a small \( \lambda_{\text{SD}} \) is crucial for determining \( \Theta_{\text{SH}} \) and will be discussed in this paper.

When a metallic ferromagnet is used, spin rectification (SR) effects, in particular anisotropic magnetoresistance and anomalous Hall effect, generate a DC voltage in the FM material at FMR. This DC voltage is added to the ISHE voltage making it difficult to disentangle the signals. There are various experimental
methods to separate ISHE from SR effects, e.g. by applying a microwave cavity to minimize the microwave currents or making use of the external magnetic field dependence of the effects [17, 18]. In a recent publication we demonstrate the complexity of the results of an angle-resolved SP setup with macroscopic bilayer samples [19].

Most of the studies on metallic systems are devoted to sputtered polycrystalline samples, thus the influence of the crystal structure of those materials on the SP experiments is neglected. Recently, studies on epitaxial bilayer systems are arising in literature [3, 19, 20]. In a prior publication [21], we optimized the growth of epitaxial Fe/Pt bilayers for SP experiments: we have shown that the real part of the spin mixing conductance $g^{\text{real}}$ (parameter which quantifies the transport of angular momentum through the FM/NM interface) and the inverse spin Hall efficiency (inverse spin Hall current divided by the absorbed microwave power) could be increased by increasing annealing temperature from room temperature to 300 °C. With in situ scanning tunneling microscope imaging it was shown, that with increasing annealing temperature the Fe/Pt interface roughness was also increased. With the present work, we aim to determine the spin Hall angle of epitaxial Fe/Pt at optimal annealing temperature (300 °C).

In the first section, we focus on the growth of the Fe/Pt bilayers, via x-ray diffraction (XRD), high resolution transmission electron microscopy (HRTEM) imaging and selected area electron diffraction (SAED) analysis, and explain the microstructuring process and design. In the second part, we determine the spin diffusion length of the Pt and the real part of the spin mixing conductance of the Fe/Pt interface by vector network analyzer ferromagnetic resonance spectroscopy (VNA-FMR) measurements of unstructured Fe/Pt films. For this, we investigated the Gilbert damping parameter of the films with varying Pt thickness. In the third section, we show the results of the angle-resolved SP measurements of the microstructured Fe(12 nm)/Pt(6 nm) sample. There, we discuss, how the ISHE and AMR voltages change, when the magnetization rotates in-plane, and, how the strong anisotropy of Fe influences the resonance condition and the ellipticity of the precession. In the last part, we calculate and discuss the spin Hall angle and compare the efficiency of the spin-to-charge current conversion in the epitaxial Pt to the results provided by recent publications.

2. Structural quality and microstructuring process of the Fe/Pt samples

Fe/Pt bilayers with a constant Fe thickness of 12 nm and a varying Pt thickness have been grown by molecular beam epitaxy on MgO (100) substrates with a growth temperature of 300 °C in an ultra-high vacuum system with a base pressure of $5 \times 10^{-10}$ mBar. To prevent oxidization we additionally capped the samples with Pt thicknesses below 1 nm with a 10 nm MgO layer. We have previously shown that this MgO capping layer has no influence on the Gilbert damping parameter $\alpha$ in FMR spectroscopy measurements [3]. To confirm the epitaxial growth of the samples we performed XRD and HRTEM measurements. The XRD pattern of the Fe(12 nm)/Pt (6 nm) sample is indicatively shown in figure 1: Pt(200) and (400) as well as Fe (200) diffraction peaks beside the MgO(200) and (400) peaks of the substrate are present, showing the formation of single crystalline grains of both materials with distinct orientation. In figure 2(a), a HRTEM image of the Fe(12 nm)/Pt(6 nm) sample is depicted, illustrating the epitaxial structural quality of the individual layers that exhibit a single crystalline nature. The Fe/Pt interface is coherent and continuous across the bilayer presenting an rms roughness of $(0.7 \pm 0.1)$ nm (2–3 Fe monolayers), while the Pt layer compensates this roughness, exhibiting a minimal rms surface roughness of $(0.2 \pm 0.02)$ nm. Later we discuss, how the atomic roughness of the Fe/Pt interface influences the ISHE in epitaxial Pt. The HRTEM imaging along with SAED analysis (figure 2(b)) showed the perfect epitaxial growth relation of the bilayer on the MgO substrate, considering that the Fe lattice is 45° in-plane rotated relative to the MgO and Pt lattices. Moreover, HRTEM measurements demonstrated that the resolved interplanar $d$-spacing values of the involved lattices are close to their bulk values and, therefore, a relaxed structural configuration is anticipated. This implies that the mismatch between consecutive lattices is accommodated by misfit dislocations.

For angular dependent SP measurements the Fe(12 nm)/Pt(6 nm) sample has been microstructured into small stripes of $200 \mu\text{m} \times 10 \mu\text{m}$ along the Fe (010) axis by means of electron beam lithography and ion beam etching. The microstructuring process did not change the SP properties, since we performed VNA-FMR spectroscopy measurements with the sample before and after the process. They showed that the lineshapes and the Gilbert damping did not change. Afterwards, Cr/Au/Cr contacts for the Fe/Pt stripes, a dielectric layer (SiO$_2$) and a coplanar waveguide (CPW)-like Ti/Cu/Ti antenna structure have been fabricated by electron-beam lithography, electron-beam evaporation and lacquering technique. The dielectric layer prevents crosstalk between the Cr/Au/Cr contacts and the microwave antenna. A scheme of those microstructures can be seen in figure 3(a).
3. VNA-FMR measurements

Here, we address the VNA-FMR measurements of the unstructured Fe/Pt sample series with constant Fe thickness of 12 nm and varying Pt thicknesses. The scope of these measurements is to measure the actual spin diffusion length of Pt and the real part of the spin mixing conductance $g_{22}$, which are critical parameters for calculating the spin Hall angle for the Fe/Pt microstructures. The Gilbert damping parameter $\alpha$ is extracted by the dependence of the linewidth on the resonance frequency:

$$\mu_0 \Delta H = \mu_0 \Delta H_0 + \frac{2\alpha f_{\text{res}}}{\gamma}$$

with the inhomogeneous broadening $\Delta H_0$ being related to the film quality. The Pt thickness dependence of $\alpha$ in figure 4 can be described by the following expression [20, 23]:

$$\alpha(d_{\text{Pt}}) = \alpha_0 + \frac{g_{22} \mu_B g_{11}}{4\pi M_s f_c} \left(1 - e^{-\frac{2\alpha}{\Delta H_0}}\right) = \alpha_0 + \Delta \alpha \left(1 - e^{-\frac{2\alpha}{\Delta H_0}}\right)$$

Figure 1. X-ray diffraction (XRD) pattern of the unstructured Fe/Pt bilayer. It displays Pt (200) and (400) as well as Fe (200) diffraction peaks beside the MgO (200) and (400) peaks of the substrate showing the formation of single crystalline grains of both materials with distinct orientation.

Figure 2. (a) Cross-sectional HRTEM image of the Fe (12 nm)/Pt (6 nm) bilayer on MgO (100), along the [001] projection direction. Perfect alignment of the in-plane crystal planes in all lattices is evident, while their experimental d-spacing values are close to their stress-free bulk values. (b) Corresponding common SAED pattern showing the good alignment of the (200) MgO and (200) Fe (gray indices) crystal planes along the growth direction. (c) In the magnified part, the perfect epitaxy of the (200) MgO, (011) Fe and (020) Pt planes (in-plane direction) is also shown. Occasionally, the Pt lattice slightly deviates towards the [110] direction.

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with $g_e$ being the Landé-factor of the free electron, $\mu_B$ the Bohr magneton, $M_S$ the saturation magnetization, $t_{Fe}$ the Fe thickness and $t_{Pt}$ the Pt thickness. $\alpha_0$ denotes the intrinsic Gilbert damping of the Fe (12 nm) reference sample, where no SP takes place. $\Delta \alpha$ is the saturation value of the Gilbert damping parameter increase due to SP.

As part of equation (2), the real part of the spin mixing conductance $g^{\parallel}$ is the defining parameter of the transport of angular momentum through the interface [22].

The obtained value of the spin mixing conductance is then $g^{\parallel} = (4.4 \pm 0.2) \times 10^{19}$ m$^{-2}$ and stands in the same order of magnitude as for other metallic systems, like Py/Pt and CoFeB/Pt [10, 24, 25]. Zhang et al. have shown with their first-principles calculations, that the real part of $g^{\parallel}$ can be increased up to 25% in highly ordered Py/Pt bilayers by introducing interface roughness [26]. Experimentally, this was observed in a prior publication [21], where both the interface roughness as well as $g^{\parallel}$ of epitaxial Fe/Pt bilayers could be enlarged by increasing the annealing temperature up to 300 °C. This optimized annealing temperature of 300 °C was also applied for the growth of the samples in this publication.

Therefore, we can use equation (2) to fit the Pt thickness dependence of $\alpha$ in figure 4 and extract the spin diffusion length of the epitaxial Pt $\lambda_{SD} = (1.1 \pm 0.1)$ nm, which is a rather small value for Pt in comparison to
other authors. This value of \( \lambda_{SD} \) implies that all spin currents are transmitted through the interface and are contributing to the ISHE. It has been shown that angular momentum can be lost at the interface due to spin memory loss [10] or other interface effects may potentially also influence the Gilbert damping parameter (e.g. proximity effect [7]). Therefore, \( g^{-1} \) has to be seen as effective spin mixing conductance including all possible interface effects [3] and displays an upper limit value. Furthermore, we show through the Pt thickness dependent FMR measurements that all spin currents in Pt relevant to ISHE are located close to the Fe/Pt interface: above a Pt thickness of about \( 2\lambda_{SD} \), all spins are flipped and additional Pt cannot contribute to the ISHE voltage. In the next section, we can also show that spin Hall angle is relatively high as well, correlating with \( \lambda_{SD} \).

4. Angle-resolved SP measurements on microstructured Fe/Pt stripes

Now, we are addressing the ISHE through angle-resolved SP measurements: a system, consisting of two perpendicularly aligned magnetic coil pairs, is providing an in-plane rotating external magnetic field magnitude for the SP measurements. For each individual external field angle, the field is swept and the DC-voltage is measured by lock-in amplification technique. The measured loops are then fitted by equation (3) involving symmetric and antisymmetric Lorentzians [25]:

\[
V_{\text{DC}}(H) = V_{\text{sym}}^z (\Delta H)^2 \left( H - H_0 \right)^2 + V_{\text{sym}}^z (\Delta H)^2 \left( H - H_0 \right)^2 + \frac{\Delta H (H - H_0)}{(H - H_0)^2 + (\Delta H)^2},
\]

where \( \Delta H \) is the linewidth, \( H_0 \) the magnetic resonance field, \( V_{\text{sym}} \) the symmetric and \( V_{\text{sym}}^a \) the antisymmetric voltage amplitude for the case in which contacts are in z-direction (as schematically shown in figure 3). Each fit parameter can then be displayed in dependence of the external magnetic field angle.

To be able to interpret the angular dependence of the fit parameters, especially the voltage amplitudes, one needs to consider the microwave dynamic fields (in- and out-of-plane) and the induced microwave currents in the Fe as well as the the contact geometry. Since the Fe/Pt stripe is located between one ground line and the signal line (see figure 3) the out-of-plane dynamic magnetic microwave field \( h_x \) is about one magnitude stronger than the in-plane field \( h_y \). The other field component \( h_z \) is negligible (see appendix). With the DC contacts along the z-direction, the angular dependence of the voltage amplitudes are then given by [17–19]:

\[
V_{\text{sym}} = V_{\text{ISHE,AMR}}^h \sin(2\Theta_M) \cos(\Theta_M) + V_{\text{AMR}}^h \sin(2\Theta_M) + V_{\text{AMR}}^h \sin(2\Theta_M) + V_{\text{AMR}}^h \sin(2\Theta_M),
\]

\[
V_{\text{sym}} = V_{\text{ISHE,AMR}}^h \sin(2\Theta_M) \cos(\Theta_M) + V_{\text{AMR}}^h \sin(2\Theta_M) + V_{\text{AMR}}^h \sin(2\Theta_M) + V_{\text{AMR}}^h \sin(2\Theta_M),
\]

where \( h_x \) and \( h_y \) in the superscript of the voltages depict effects excited by in-plane and out-of-plane dynamic magnetic fields. In-plane excited AMR and ISHE have the same dependence (\( \sin(2\Theta_M) \)) and can therefore not be separated (\( V_{\text{ISHE,AMR}}^h \) or \( V_{\text{ISHE,AMR}}^h \) only in \( V_{\text{sym}} \)). In-plane (\( V_{\text{AMR}}^h \)) and out-of-plane (\( V_{\text{AMR}}^h \)) excited AHE have unique shapes, but are strongly suppressed with the DC-contacts in z-direction, since microwave currents flowing in x-direction are negligible. What is expected to appear mainly, are out-of-plane excited AMR (\( V_{\text{AMR}}^h \)) and out-of-plane excited ISHE (\( V_{\text{ISHE}}^h \)) in the symmetric and antisymmetric voltage and out-of-plane excited ISHE (\( V_{\text{ISHE}}^h \)) in the symmetric voltage amplitude.

Since the SR effects and ISHE depend on the alignment between the magnetization and the z-axis (\( \Theta_M \)), which differs from the angle between the external magnetic field and the z-axis (\( \Theta_H \), due to the anisotropy field, as schemed in figure 3(b). The measured data sets of fit parameters in dependence of \( \Theta_H \) were readjusted to \( \Theta_M \). Therefore, the cubic anisotropy constant \( K_1 \) (arising from the bcc Fe lattice) and uniaxial anisotropy constant \( K_u \) (arising from shape anisotropy of the Fe/Pt stripe) are extracted from the change in resonance field \( H \) (shown in figure 5) according to:

\[
f = \mu_0 \gamma \sqrt{(H_z \cos(\Theta_M) - \Theta_M) + (N_x + N_y) M_0 (H_z \cos(\Theta_M) - \Theta_M) + (N_y + N_y) M_0},
\]

with the demagnetization factor for a thin magnetic film \( N_x = 0, N_y = 1 \) and \( N_z = 0 \), using effective demagnetization factors \( N_x^e \) and \( N_y^e \) to take the cubic and uniaxial anisotropy into account for the resonance condition (described in equation (6)) [27, 28], and with \( \gamma = 28 \, \text{GHz} \, \text{T}^{-1}, M_0 = 1712 \, \text{kA} \, \text{m}^{-1} \) (measured with VNA-FMR), \( K_1 = 44290 \, \text{J} \, \text{m}^{-3} \) and \( K_u = 3600 \, \text{J} \, \text{m}^{-3} \).
When the angle of magnetization rotates $N_y$ varies between 0.026 and 0.012 and $N_x$ varies between 0.026 and $-0.024$ for the given parameters. Equation (5) is then used to calculate $\Theta_M$ for each individual measurement point of the external field angle. The fit function parameters from equation (3) are then redistributed to $\Theta_M$ and the conversion from $\Theta_H$ to $\Theta_M$ is performed.

In figure 6 the symmetric and antisymmetric voltage amplitudes in dependence of the magnetization angle with fit functions according to equation (4) are shown. The antisymmetric voltage (green dots) has almost no deviation from the expected $\sin(2\Theta_M)$-shape for out-of-plane excited AMR indicating low AHE (due to longitudinal contacts) and low in-plane excitation fields. The shape of the symmetric voltage (black squares) can be described by the overlapping of out-of-plane AMR and out-of-plane excited ISHE $\propto \sin(\Theta_H)$. The ISHE component excited by the out-of-plane dynamic magnetic field (1.92 $\mu V$, component of $V_{sym}$ according to equation (4)) is plotted as a blue dashed line. As one can see, the ISHE voltage is of similar magnitude as the AMR voltage. To compare all signal components, the voltages of the contributing effects are listed in table 1.
see that effects generated by in-plane excitation fields are a magnitude smaller than effects generated by out-of-plane excitation fields and AHE contributions, in general, are negligible, since AHE with contacts in z-direction scales with $j_x$. As we show in appendix A, the excitation by the CPW generates mainly microwave currents in z-direction and $j_x$ is negligible. Small deviations of the data points from the fit functions can be seen for the symmetric and antisymmetric voltage amplitudes at the angles of $\pm 135^\circ$ and $\pm 45^\circ$. Around these angles double resonances appear in the measurements due to the strong magneto-crystalline anisotropy. Since our theory only account single resonances, the experimental data deviates from the fit function.

5. Calculation of the spin Hall angle

To calculate the spin Hall angle, the cone-angle of the magnetization precession $\theta$ is needed to be known. With COMSOL Multiphysics, we simulate the microwave currents inside the Fe layer and the magnetic microwave fields around it. Then, with the Polder susceptibility tensor (with relevant components $A_{xy}$ and $A_{yy}$) which transforms the magnetic excitation field $h_{ij}$ into the dynamic components of the magnetization $m_x$, $m_y$ (with the rotating coordinates $x^\prime$, $y^\prime$, $z^\prime$, where the $z^\prime$ and $x^\prime$-axis is rotating with the magnetization), we are able to calculate the cone-angle of the FMR precession $\theta$ with the following expression:

$$P \sin^2(\theta) = A_{xy}A_{yy} h_{ij}^2 / M_S^2,$$

where $P$ is the correction factor for the precession ellipticity and $M_S$ is the static component of the magnetization. Further information about the COMSOL simulation, the method of calculating $h_{ij}$ and about the Polder tensor can be found in appendices A and B.

Equation (8) describes the strength of the ISHE voltage generated by the FMR precession and the SP through the Fe/Pt interface. As the magnetization precesses elliptically with the cone-angle $\theta$ at a frequency $\pi / 2\pi$, a spin current proportional to $P \sin^2(\theta)$ is flowing through the Fe/Pt interface and angular momentum is transferred into the Pt, which is described by the real part of the spin mixing conductance $g^r$. This spin current is then deflected by ISHE inside the Pt over the characteristic length scale $\lambda_{SD}$ with the spin-to-charge current efficiency $\Theta_{SH}$. With this, a DC voltage arises between the contact points over the stripe length $l$, which is attenuated by the electrical shunting of the metallic bilayer itself [4, 17]. The measured ISHE voltage is then expressed by:

$$V_{ISHE}^z = - \frac{l}{M_S^2} \sin(\Theta_M) A_{xy} A_{yy} h_{ij}^2 \Theta_{SH} \lambda_{SD} g^r_{film} \left( \frac{\omega}{2\pi} \right) \tanh \left( \frac{\eta_{pl}}{2\lambda_{SD}} \right),$$

where $l = 200 \mu m$ is the length of the Fe/Pt stripe, $A_{xy} (\Theta_M = 90^\circ) = 173.7$ and $A_{yy} (\Theta_M = 90^\circ) = 35.6$ are the Polder tensor components, $\sigma_{film} = 5.4 \times 10^6 (\Omega m)^{-1}$ is the measured film conductivity, $t_{film} = 18 nm$ is the Fe/Pt film thickness, $\omega = 2\pi \times 13 GHz$ is the angular frequency of the microwave and $\eta_{pl} = 6 nm$ is the Pt layer thickness.

With equation (8) and $h_{ij} = 273.7 A m^{-1}$ from simulation, the spin Hall angle is then calculated as $\Theta_{SH} = (5.7 \pm 1.4)^\circ$. Therefore, the value of epitaxial Pt lies within the range of 1% up to 10% reported for mostly polycrystalline RF-sputtered Pt [9] and is comparable to epitaxial Pt reported by other authors [20]. The origin of ISHE is the spin–orbit coupling of Pt, whereas theoretically there is a distinction between the intrinsic and the extrinsic mechanism behind ISHE. With the intrinsic mechanism, the spin–orbit coupling effects directly the electronic band structures. Therefore, it is independent of the electron scattering with defects, grain boundaries or phonons, contrary to the extrinsic mechanism. Intrinsic ISHE in Pt has a strong temperature dependence due to the change of resistivity. Sagasta et al have shown [29] how to tune the spin Hall angle of Pt in spin valve devices by tuning the resistivity through defect density from the moderately dirty to the ultra-clean region for their Pt. They differentiate the intrinsic from the extrinsic (skew scattering) SHE contribution by the means of changing the temperature of their devices. Their ultra-clean, electron beam evaporated Pt showed a SHA of about 2%–3% due to their low resistivity of their Pt. This low resistance is also connected to the large Pt thickness of their devices (20 nm) compared to ours. There, the intrinsic bulk contribution to the SHA of Pt is reduced and has the same weight as the extrinsic contribution. Although our Pt is e-beam evaporated, its low conductivity and relatively large SHA are similar to the ones of the sputtered devices. This is due to the small
layer thickness of our Pt (6 nm) and furthermore due to the fact that in the very first nanometers of Pt a larger disorder in form of dislocations is expected near the Fe/Pt interface, where the lattice mismatch between Fe and Pt must be compensated (see section 2). Despite the similarity of Sagastas Pt resistivity dependence of the spin Hall angle with our results, it is necessary to point out the differences: the measurements of Sagasta et al are performed on spin valve devices with Pt, where spin currents are flowing in lateral dimensions (spin currents in SP experiments are in the transversal dimension). Additionally, there the Pt has no contact with the ferromagnet. For both, the sputtered and the e-beam evaporated devices, the Pt should be polycrystalline. Therefore, the spin Hall angle of spin valve devices is not perfectly comparable to the one of SP experiments. In short, the reduced conductivity of our e-beam evaporated Pt allows for the intrinsic ISHE to strongly contribute to the spin Hall angle. Additionally to the intrinsic contribution, extrinsic mechanisms to the ISHE in our Pt could be present. The extrinsic ISHE contribution from the bulk-Pt should be largely reduced in our experiment (single-crystalline, highly ordered Pt layers) compared to polycrystalline Pt layers [29]. On the other side, the small spin diffusion length points to the lead that the atomically rough interface (2–3 monolayers) could have an extrinsic contribution (surface-assisted skew scattering) to the relatively large spin Hall angle, which was measured here at room temperature. Other authors reported similar theoretical and experimental findings: Gu et al [30] strongly enhanced the room temperature spin Hall angle of Au films by experimentally realizing a surface-assisted spin Hall effect with Pt impurities on the Au (111) surface. In a following publication, Gu et al [31] have shown, that this surface-assisted skew scattering mechanism strongly depends on the surface morphology of Au, on which Pt is located. Doping bulk-Au with Pt resulted in a much smaller enhancement of the spin Hall angle. Additionally, Hou et al investigated SP and ISHE in Bi/Pt bilayers and discovered, that the spin Hall angle of Bi was significantly enlarged in the vicinity of the interface and was small in the bulk-Bi [32]. And recently, Alves-Santos et al have greatly enlarged the spin Hall angle of Pt by inserting Ag nano-particles inside of the Pt layer, where the local spin orbit-coupling is enhanced by the Rashba effect [33]. Considering the above reported findings and the prior publication [21], we propose the ISHE in ultra-clean epitaxial Pt is very sensitive to the conductivity of the Pt and, therefore, to the Pt layer thickness. Potentially, the interface morphology with its distinct atomic interface roughness might support ISHE by surface-assisted skew-scattering, when the conductivity of Pt is in the transition region between extrinsic and intrinsic dominated ISHE.

6. Conclusions

Fe/Pt bilayers have been grown stress-free with a distinct roughness of 2–3 monolayers at the Fe/Pt interface. With unstructured, large area Fe/Pt samples with Pt thicknesses ranging from 0 to 18 nm we determined the spin mixing conductance \((4.4 \pm 0.2) \times 10^{19} \text{ m}^{-2}\) by VNA-FMR. By fitting the Gilbert damping dependence on the Pt thickness for the samples with very thin Pt we extracted the spin diffusion length \((1.1 \pm 0.1) \text{ nm}\), which is atypically small for Pt. With angle-resolved SP measurements on a microstructured Fe/Pt stripe and with a COMSOL simulation of the microwave excitation, we obtained the spin Hall angle. Its value of \((5.7 \pm 1.4)\) is unexpectedly large for highly pure, epitaxial Pt. We explain this behavior due to the reduced conductivity of the Pt and the emerging contribution of the intrinsic mechanism.

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Appendix A. COMSOL simulation of the microstructured CPW

To quantify the efficiency of the ISHE in epitaxial Fe/Pt, it is necessary to determine the cone-angle and the ellipticity of the precession in Fe at the excitation power used in our experiment (18 dBm). Since the absorbed microwave power is very small and, therefore, hard to quantify experimentally in microstructured samples, the precession dynamic is not easily accessible. We simulated the microwave excitation fields with COMSOL Multiphysics and extracted the dynamic magnetic field \(h_z\) around the Fe layer and the microwave induced electrical current \(j_z\) inside the Fe from the simulation. Later on, we compare the simulated values of \(h_z\) and \(j_z\) with the AMR signal from figure 6, which is strongly dependent on these two. Additionally, the microwave current is
estimated by the theoretical impedance of the structures. With this, we ensure, that the microwave power level of the simulation is comparable to the experiments. This way, the precession dynamics can be calculated.

The radio frequency (RF) module of COMSOL was used to simulate the excitation of our small Fe/Pt stripe by the CPW antenna (scheme shown in figure 3). We used lumped ports, the known impedance of 50 Ω and microwave power of 18 dBm to excite the CPW. To account for the small thicknesses of Fe and Pt the meshing was chosen finer around the stripe. Literature values for electrical conductance, permeability and susceptibility of the relevant materials was used (except for Fe and Pt, there we estimated $\sigma_{Fe} = 3.3 \times 10^6 (\Omega m)^{-1}$ and $\sigma_{Pt} = 2.1 \times 10^6 (\Omega m)^{-1}$, since they have reduced conductivities due to very small thicknesses). Since COMSOL can only describe static magnetization behavior, but not magnetization precession, the microwave dynamic magnetic field of the sample-free space at the same position was used and the dynamics were calculated theoretically. For the AMR, we assume, that the interaction of the magnetization and the microwave electric field, which induces microwave currents inside the Fe, can be neglected. Therefore, we use the value of the microwave currents given by the COMSOL simulation.

In figure A1, a 2D color mapping of the normalized current densities inside of the CPW (a) and the Fe layer (b), taken from COMSOL, are displayed. The current densities in the CPW and the Fe are peaking in the order of $10^9$ Am$^{-2}$ and are concentrating around the inner edges (skin effect). The same behavior is seen in figure A2(b), where one can see the non-normalized current density components. Additionally, circular currents at the inner edges of the CPW in the $x$–$z$ plane are visible, but mainly the $j_z^{CPW}$ component will induce the currents inside the
Fe. The microwave currents in the CPW have opposite signs between ground and signal line, as expected from the design. To compare the simulation results with the theoretical current density (according to impedance and excitation power), $j_{\text{CPW}}$ all over the $x$–$y$–plane was integrated. With this, the absolute current inside the CPW is $I_{\text{CPW}} = 55.5$ mA. Compared to the current expected by a simplified analytical model including the microwave power and CPW impedance $\sqrt{0.0631 \, \text{W}/50 \, \Omega} \cdot \sqrt{2} = 50.2$ mA (taken the factor of $\sqrt{2}$ for transforming the effective current into the physical amplitude), there is only a small 10% derivation. The current inside the Fe layer was also integrated: $I_{\text{Fe}} = 0.39$ mA. To this point no microwave reflection (by cables or by the picoprobes itself) was taken into account. Therefore, the simulated current inside Fe can be considered as upper limit value.

In figure A3 the excitation microwave (a) electric and (b) magnetic field components in dependence on the Fe cutline $x$ position (see cutline scheme figure A2(a)) can be seen. One can see, that there are significant electric in-plane fields at the edges of the CPW and strong electric out-of-plane fields directly beneath the CPW. At the position of the Fe sample, one the other hand, the electric fields are strongly reduced. There, the electric field from signal and ground line are compensating each other. The microwave magnetic in- and out-of-plane fields are peaking strongly at positions near the CPW edge due to the skin effect at high microwave frequencies. At the Fe position the in-plane magnetic fields are mostly compensated and the preferred out-of-plane component $h_y$ is relatively homogeneous. Between the left ground and the signal line, $h_y$ is dropping because of the large (static) susceptibility of Fe in the simulation. For calculating the magnetization dynamics, the magnetic microwave field surrounding Fe is needed and we extract the value of $h_y = 310$ A m$^{-1}$ from the position between the signal and the right ground line for further evaluations (figure A3(b) indicated by the arrow). This value as well, later, needs to be adjusted because of microwave reflection losses of the cables, picoprobes and so on.

To adjust the absolute microwave power level of the simulation to the experiments, we utilize the out-of-plane excited AMR signal shown in figure 6 and listed in table 1. It will be used to estimate the microwave transmission of the setup with the following equation [18]:

$$V_{\text{AMR}} = \frac{\Delta R}{2M_s} A_{xy} h_y I_{\text{Fe}} \sin(2\Theta_M),$$  \hspace{1cm} (A.1)

with the Polder tensor component $A_{xy}(\Theta_M = 45^\circ) = 161.7$ and $\Delta R = 0.67 \, \Omega$ being the change of film resistance with the angle of magnetization. $\Delta R$ was separately measured by resistance measurements with rotating external magnetic field without microwave excitation and calculated according to $R_{\text{film}} = R_0 - \Delta R \sin^2(\Theta_M)$. The left side of equation (A.1) is already measured and shown at figure 6 and table 1, but the average from the symmetric and antisymmetric voltage has to be taken, since AMR contributes to both amplitudes in general:

$$V_{\text{AMR}} = \sqrt{(0.73 \, \mu V)^2 + (2.95 \, \mu V)^2} = 3.04 \, \mu V.$$  \hspace{1cm} (A.2)

Using equations (A.1) and (A.2) the product $h_y I_{\text{Fe}}$ for the measurement can be calculated: $(h_y I_{\text{Fe}})_{\text{meas}} = 0.096$ A$^2$ m$^{-1}$. Since the microwave induced current in Fe is proportional to the microwave electric field, this factor is proportional to the microwave power transmitted to the CPW. The comparison with the factor extracted from the simulation $(h_y I_{\text{Fe}})_{\text{COMSOL}} = 0.120$ A$^2$ m$^{-1}$ shows that 78% of the microwave power is transmitted to the CPW. With this, the microwave magnetic field from the simulation needs to be adjusted to the experiment:

$$h_y = \sqrt{0.096} \cdot 310 \, \text{A} \cdot \text{m}^{-1} = 273.7 \, \text{A} \cdot \text{m}^{-1}.$$  

This value is then used to calculate the spin Hall angle using equation (8) in section 5.
Appendix B. Polder susceptibility tensor

In order to calculate the magnetization precession dynamics from the microwave magnetic fields, the Polder susceptibility tensor needs to be taken into account [18, 28]:

\[
\chi = \begin{pmatrix}
\chi_{xx} & i\chi_{xy} & 0 \\
-i\chi_{xy} & \chi_{yy} & 0 \\
0 & 0 & 0
\end{pmatrix},
\]

(B.1)

with:

\[
(\chi_{xx}, \chi_{xy}, \chi_{yy}) = (D + iL)(A_{xx}, A_{xy}, A_{yy})
\]

(B.2)

and

\[
L = \frac{\Delta H^2}{(H-H_s)^2 + \Delta H_s^2}, \quad D = \frac{\Delta H (H-H_s)}{(H-H_s)^2 + \Delta H_s^2},
\]

(B.3)

\[
A_{xx} = \frac{\gamma \mu_0 M_S [\mu_0 M_s (N_y + N_y) + (\mu_0 H - N_x \mu_0 M_s)]}{\alpha [2(\mu_0 H - N_x \mu_0 M_s) + \mu_0 M_s (N_z + N_y + N_y)]},
\]

(B.4)

\[
A_{xy} = -\frac{\mu_0 M_S}{\alpha [2(\mu_0 H - N_x \mu_0 M_s) + \mu_0 M_s (N_z + N_y + N_y)]},
\]

(B.5)

\[
A_{yy} = \frac{\gamma \mu_0 M_S [\mu_0 M_s (N_x + N_y) + (\mu_0 H - N_x \mu_0 M_s)]}{\alpha [2(\mu_0 H - N_x \mu_0 M_s) + \mu_0 M_s (N_z + N_y + N_y)]},
\]

(B.6)

Here, the anisotropy is taken into account with the effective demagnetization factors from equation (6). For the AMR and ISHE $A_{xy}$ and $A_{yy}$ needs to be calculated for each magnetization angle, individually.

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