

# Tm<sub>3</sub>Fe<sub>5</sub>O<sub>12</sub>/Pt Heterostructures with Perpendicular Magnetic Anisotropy for Spintronic Applications

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With recent developments in the field of spintronics, ferromagnetic insulator (FMI) thin films have emerged as an important component of spintronic devices. Ferrimagnetic yttrium iron garnet in particular is an excellent insulator with low Gilbert damping and a Curie temperature well above room temperature, and has been incorporated into heterostructures that exhibit a plethora of spintronic phenomena including spin pumping, spin Seebeck, and proximity effects. However, it has been a challenge to develop high quality sub-10 nm thickness FMI garnet films with perpendicular magnetic anisotropy (PMA) and PMA garnet/heavy metal heterostructures to facilitate advances in spin-current and anomalous Hall phenomena. Here, robust PMA in ultrathin thulium iron garnet (TmIG) films of high structural quality down to a thickness of 5.6 nm are demonstrated, which retain a saturation magnetization close to bulk. It is shown that TmIG/Pt bilayers exhibit a large spin Hall magnetoresistance (SMR) and SMR-driven anomalous Hall effect, which indicates efficient spin transmission across the TmIG/Pt interface. These measurements are used to quantify the interfacial spin mixing conductance in TmIG/Pt and the temperature-dependent PMA of the TmIG thin film.

include recently discovered phenomena such as the quantum anomalous Hall effect<sup>[2]</sup> in magnetic topological insulators (TIs),<sup>[3]</sup> spin transfer torque<sup>[4,5]</sup> effects in nonmagnetic metal/ferromagnetic metal/oxide heterostructures, and spin orbit torque (SOT) effects in heterostructures that include ferromagnetic metals/heavy metals,<sup>[6,7]</sup> magnetically-doped TIs,<sup>[8]</sup> and FMI/heavy metals where the FMI is a garnet<sup>[9]</sup> or Ba hexaferrite.<sup>[10]</sup> To realize novel circuit devices based on these effects a variety of magnetic materials and heterostructures has still to be developed in which the magnetic properties and interfacial spin transport can be controlled.

FIMs with PMA are of particular interest in spintronics. In FMI/heavy metal or FMI/TI heterostructures, current is limited to the metal or to the surface layer of the TI which reduces the conductivity (and potentially the power consumption) compared to all-metallic structures,

and avoids the possibility of direct spin transfer torque from current flow in the FMI layer. This facilitates the study of proximity effects, SOT and other exotic phenomena occurring at the interfaces, enabling for example an identification of the various contributions to spin orbit torques. Moreover, the presence of PMA in the FMI leads to stray-field-induced interface effects even at remanence. Domain walls in PMA films also have qualitatively different structures and dynamics compared to those of FIMs with in-plane magnetic anisotropy, which is relevant to racetrack memory or logic devices. Thus there is considerable interest in developing PMA FMI materials and heterostructures.

One of the most prominent classes of FMI is that of ferrimagnetic iron garnets, of which the best studied is Y<sub>3</sub>Fe<sub>5</sub>O<sub>12</sub> (YIG). The ultralow damping<sup>[11]</sup> and magneto-optical properties<sup>[12,13]</sup> of YIG are well known. The former makes YIG a suitable candidate for spin wave logic<sup>[14]</sup> and signal transmitters<sup>[15]</sup> due to the extremely large magnon propagation length of several tens of millimeters. YIG/heavy metal (e.g., Pt, W, Ta) and YIG/topological insulator heterostructures have demonstrated proximity effects, spin pumping, spin Seebeck, and other spintronic phenomena.<sup>[15–19]</sup> However, YIG films generally have an in-plane easy axis dominated by shape anisotropy due to their weak

## 1. Introduction

Harnessing the electron's second fundamental property, its spin, is the basis of spintronic phenomena and devices.<sup>[1]</sup> These

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DOI: 10.1002/aelm.201600376

magnetocrystalline anisotropy and magnetostriction. Epitaxial<sup>[20]</sup> and polycrystalline<sup>[21,22]</sup> YIG films have been reported to show PMA at low thickness as a result of magnetoelastic anisotropy arising from lattice mismatch and thermal mismatch respectively, but the fabrication of good quality YIG films with high out-of-plane remanence remains elusive. Fortunately, the magnetic properties of iron garnets can be dramatically altered by substitution of rare earths onto the Y sites. Kubota et al.<sup>[23,24]</sup> showed that 50 nm thick thulium iron garnet ( $\text{Tm}_3\text{Fe}_5\text{O}_{12}$ , TmIG) possesses PMA caused by magnetoelastic anisotropy<sup>[25]</sup> when grown epitaxially on (111)-oriented gallium gadolinium garnet ( $\text{Gd}_3\text{Ga}_5\text{O}_{12}$  or GGG), and we recently demonstrated<sup>[9]</sup> reversible magnetization switching via SOT in an 8 nm thick TmIG film grown on GGG, coated with Pt and patterned into a Hall cross.

In the present article, we provide a comprehensive description of the structural characteristics and magnetic properties of thin films of TmIG with thickness of 5.6–30 nm grown on GGG, as well as the electrical properties of TmIG/Pt heterostructures. The TmIG films show bulk magnetization, moderate coercivity ( $\approx 24 \text{ kA m}^{-1}$ ), high out-of-plane remanence, and a ferrimagnetic structure with compensation point below 1.5 K. We demonstrate that efficient spin transport can be achieved through the TmIG/Pt interface by measuring spin Hall magnetoresistance (SMR) in Pt, which also gives rise to an anomalous Hall effect (AHE) signal in the Pt heavy metal layer. Lastly, we use the strong SMR signal to measure the anisotropy field of the TmIG film electrically, which is inaccessible by conventional magnetometry measurements due to the dominant paramagnetic contribution of the GGG substrate. We identified the magnetoelastic and magnetocrystalline contributions to the total magnetic anisotropy, and we show that the spin mixing conductance is invariant in a temperature regime at and above room temperature. The TmIG/Pt heterostructure is thereby shown to be a robust system for further investigation of interfacial spintronic phenomena in a PMA FMI.

## 2. Structural Characterization of TmIG/GGG

Several TmIG films of thickness 5.6–30 nm were grown on (111)-oriented GGG by pulsed laser deposition as described in the Experimental Section. The results of structural characterization of the TmIG films are summarized in **Figure 1**. Thickness was determined by X-ray reflectometry (XRR) scans (not shown), and yielded a thickness of 5.6 nm ( $\pm 0.1 \text{ nm}$ ) for the thinnest film examined.

### 2.1. Temperature Dependent Strain of TmIG/GGG

Garnets crystallize in the cubic structure with lattice parameter around 1.2 nm and eight formula units in the unit cell. GGG has an excellent lattice match with YIG and other iron garnets, and as expected, TmIG grew epitaxially on GGG substrates. **Figure 1a** shows the symmetric high resolution X-ray diffraction (XRD) scan of a 19.7 nm thick TmIG film on GGG, revealing only (*hhh*)-type reflections, and yielding an out-of-plane lattice spacing of  $d_{\text{TmIG, XRD}}^{444} = 0.1769 \text{ nm}$ . Laue fringes around the TmIG peak indicate the high crystalline quality and thickness

uniformity of the layer. (Peaks from thinner films could not be resolved using XRD.)

A reciprocal space map of the 19.7 nm thick TmIG film around the TmIG and GGG (624)<sub>+</sub> asymmetric peaks (**Figure 1b**) confirms the coherent growth of the TmIG on the GGG substrate. The in-plane values for the (220) lattice spacing for both the TmIG film and the substrate are  $d_{220}^{\text{TmIG}} = d_{220}^{\text{GGG}} = q_x^{-1} = 0.4377 \text{ nm}$ , i.e., the cubic lattice parameter  $a = 1.238 \text{ nm}$  which is in very good agreement with literature values for GGG.<sup>[26]</sup> The out-of-plane lattice spacing for the TmIG film according to the reciprocal space map (RSM) is  $d_{444}^{\text{TmIG, RSM}} = q_z^{-1} = 0.1768 \text{ nm}$ , very close to that measured by symmetric XRD. The differences between the in-plane and out-of-plane lattice spacing of (111) TmIG show that the TmIG unit cell is compressively strained in the [111] out-of-plane direction due to its epitaxy with the GGG. The resulting distortion angle for the facets of the TmIG unit cell is  $\beta = 90.77^\circ$  (**Figure 1c,d**). We assume that, since the 19.7 nm thick film is fully strained to the substrate, thinner films are also fully strained.

Temperature-dependent measurements of the  $d_{444}$  lattice parameter of GGG, the pseudo-cubic  $d_{444}$  of TmIG (calculated as the cube root of the unit cell volume) and the distortion angle  $\beta$  are shown in **Figure 1c** for the temperature range of 50–200 °C. The data yield linear thermal expansion coefficients in the (111) direction of  $\alpha_{\text{L, GGG}} = 14.1 \times 10^{-6} \text{ K}^{-1}$  and  $\alpha_{\text{L, TmIG}} = 15.57 \times 10^{-6} \text{ K}^{-1}$  for the substrate and the film, respectively. Evidently, the lattice mismatch strain decreases slowly with increasing temperature.

### 2.2. Surface Morphology and Crystal Quality

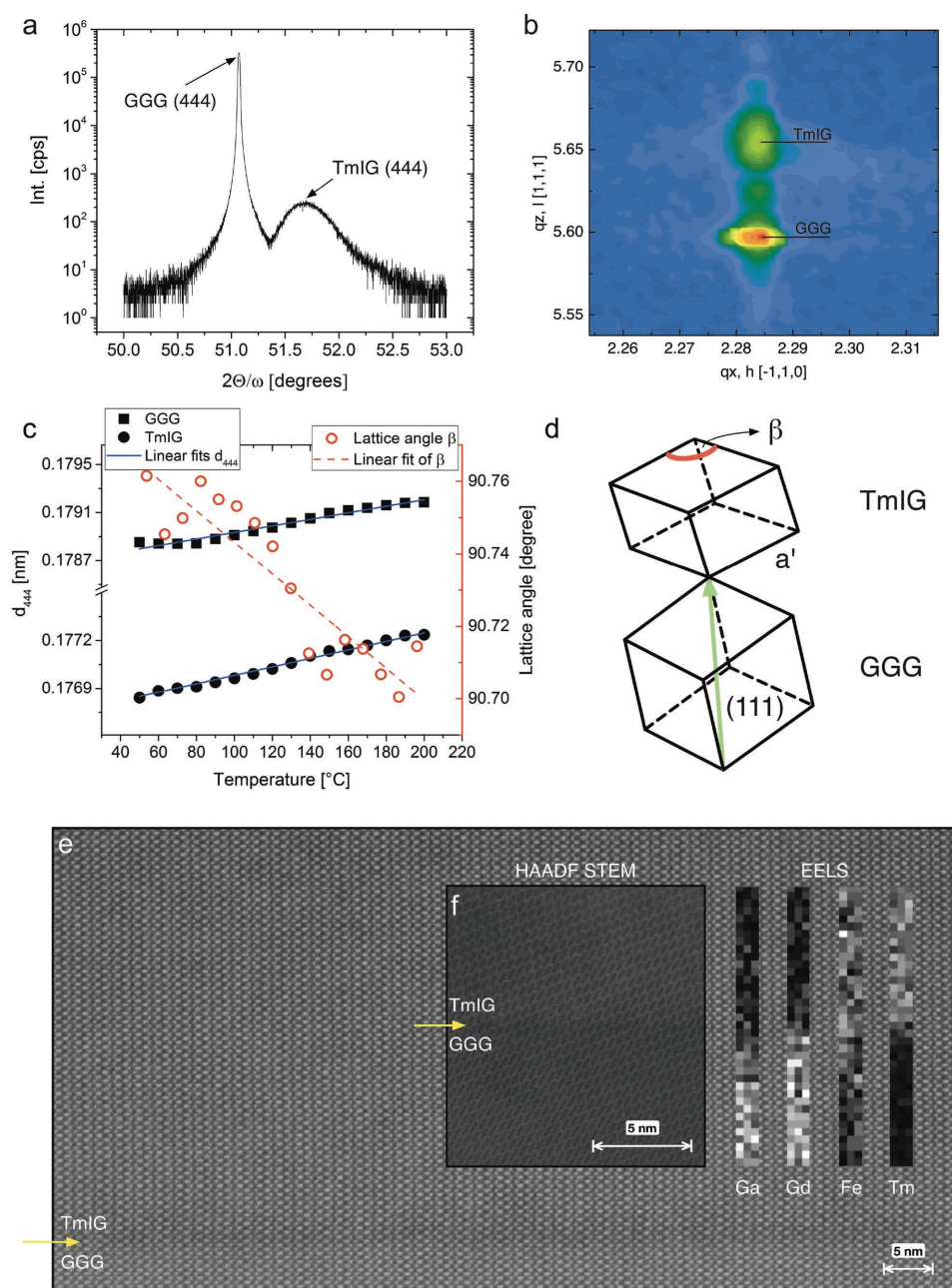
The surface morphology of the 19.7 nm thin TmIG film was measured via atomic force microscopy and exhibits an RMS roughness value of 1.5 nm calculated from the topographic image (**Figure 2a**). This film showed a few particles which led to the high roughness, but the roughness of the thinner films used for electrical studies was as low as 0.2 nm.

The crystal quality and interface structure were examined using high resolution scanning transmission electron microscopy (STEM) of a cross-section of a TmIG/GGG sample. An image taken along the  $[11\bar{2}]$  direction is shown in **Figure 1e**. The yellow arrow indicates the interface between the TmIG and the GGG substrate. There is no evidence of dislocations in the crystal structure of TmIG or at the interface over the field of the images. **Figure 1f** shows a high angle annular dark field scanning transmission electron microscopy (HAADF-STEM) image in the [101] direction and corresponding electron energy loss spectroscopy (EELS) line scans (**Figure 1f**) perpendicular to the interface, which show Tm and Fe in the film and Ga and Gd in the substrate, indicating little or no interdiffusion of the cations between the garnet materials.

## 3. Magnetic Characterization

### 3.1. Hysteresis and Domain Structure

Polar magneto-optical Kerr effect (MOKE) and vibrating sample magnetometry (VSM) measurements on the 5.6 nm thin TmIG



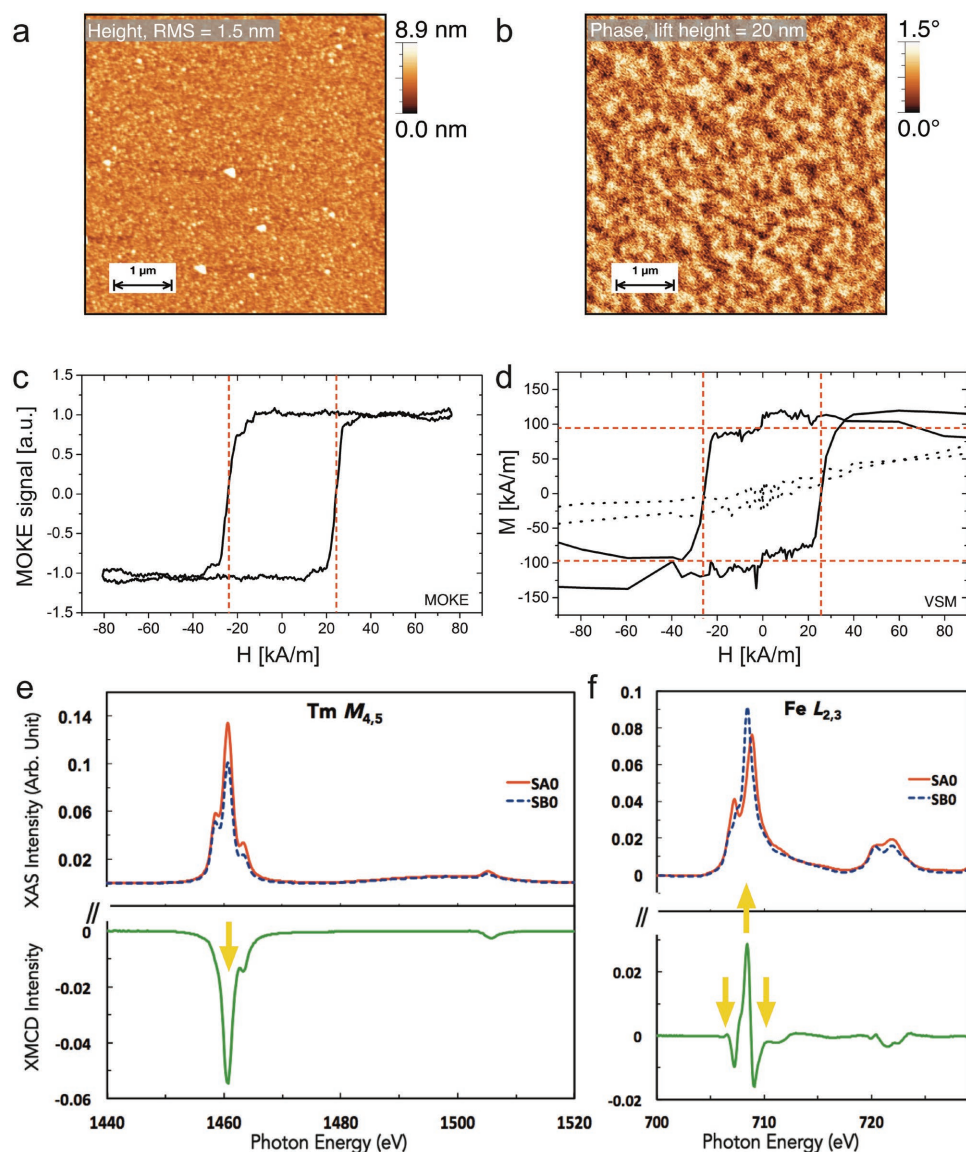
**Figure 1.** Structural characterization of TmIG. a) XRD symmetric scan of the TmIG (19.7 nm)/GGG sample around the (444) peak. b) Reciprocal space map of the asymmetric  $(624)_+$  diffraction peaks of the substrate GGG (lower peak) and the film (upper peak). c) Temperature evolution of the TmIG and GGG  $d_{444}$  lattice parameters and of the resulting lattice distortion angle  $\beta$ . d) Sketch of the two adjacent unit cells of TmIG and GGG at the interface. The rhombohedral deformation of the pseudocubic TmIG unit cell is exaggerated. e, f) HAADF STEM images of the TmIG/GGG structure at the interface in (e) the  $[112]$  direction and (f) the  $[10\bar{1}]$  direction. EELS line scans at the interface show the distribution of Ga, Gd, Fe, and Tm atoms as black–white contrast.

film are shown in Figure 2c,d. These both reveal the hysteresis loop shape and coercivity but only the VSM gives a quantitative measurement of magnetic moment; however, the paramagnetic background of the GGG substrate impedes measurements of hysteresis loops by VSM. The films exhibit a square hysteresis loop with coercivity of  $23.9 \text{ kA m}^{-1}$ . No hysteretic behavior was measurable for an in-plane loop, which indicates uniaxial anisotropy with easy axis along the surface normal. Based on

the thickness derived from an XRR measurement, the saturation magnetization  $M_S$  for the 5.6 nm thick TmIG sample and the Pt/TmIG(8 nm) sample was  $100 (\pm 1.5) \text{ kA m}^{-1}$ , and several other films of different thickness gave values of  $90\text{--}100 \text{ kA m}^{-1}$ . This is in good agreement with data measured for bulk TmIG.<sup>[25]</sup>

Measurements of magnetic damping of the TmIG films were challenging due to the very low signal to noise ratio and the PMA, but gave values in the range of  $\alpha \sim 10^{-3}$  for a TmIG





**Figure 2.** Magnetic characterization of TmIG thin films. a) Topographic and b) phase images of a MFM scan on the 19.7 nm thick TmIG film. The RMS surface roughness is 1.5 nm and characteristic stripe domains possess a feature size below 1  $\mu\text{m}$ . c) Polar MOKE signal versus applied out-of-plane magnetic field for the 5.6 nm thick TmIG film. d) Vibrating sample magnetometer measurement on the same film with field applied normal to the sample plane (continuous line) and parallel to the sample plane (dashed line). e,f) The XAS (upper panel) and XMCD (lower panel) asymmetric intensity signals for e) the  $M_{4,5}$  edge of Tm and f) the  $L_{2,3}$  edge of Fe. The arrows in the lower panels correspond to the magnetization direction at the respective energy peak, pointing up (down) for positive (negative) XMCD signal.

film on (100) GGG which exhibited in-plane magnetization. Prior work on bulk TmIG showed an FMR line width of  $\Delta H = 5.17 \text{ kA m}^{-1}$ <sup>[27]</sup> which is one of the lower values reported for a range of rare earth garnets.<sup>[28]</sup>

Magnetic force microscopy (MFM) was used to image the domain structure of the thicker films. Figure 2b shows an MFM image of the 19.7 nm thin TmIG film after demagnetization, performed by placing the sample in a 10 Hz alternating magnetic field whose amplitude decreased approximately exponentially from 160 to 0  $\text{kA m}^{-1}$  over a time period of 2 min. The domain pattern is typical for PMA materials without lateral anisotropy<sup>[29]</sup> with a period of  $\approx 0.3 \mu\text{m}$ , and the area coverage

of up- and down-magnetized domains is, as expected, approximately equal.

### 3.2. Determination of the Magnetic Anisotropy

The magnetic anisotropy of the TmIG films includes contributions from magnetocrystalline, magnetoelastic, and shape anisotropies. Room temperature values for the first order cubic anisotropy constant  $K_1$  and the magnetostriction coefficients  $\lambda_{111}$  and  $\lambda_{100}$  of bulk TmIG have been reported to be  $-1.1 \text{ kJ m}^{-3}$   $< K_1 < -0.58 \text{ kJ m}^{-3}$ ,  $\lambda_{111} = -5.2 \times 10^{-6}$  and  $\lambda_{001} = 1.4 \times 10^{-6}$ .<sup>[25]</sup>

The uniaxial anisotropy  $K_u$  is the difference between the total magnetic energy when the magnetization is oriented in-plane versus out-of-plane, and can be written as<sup>[30]</sup>

$$K_u = E_{IP} - E_{OP} = -\kappa_{1/2} + \frac{1}{4} \lambda_{111} c_{44} \left( \frac{\pi}{2} - \frac{\beta}{2} \right) + \left( \frac{u_0}{2} \right) M_s^2 \quad (1)$$

In this expression the magnetocrystalline anisotropy is  $-\kappa_{1/2}$ , the magnetoelastic anisotropy is  $\frac{1}{4} \lambda_{111} c_{44} \left( \frac{\pi}{2} - \frac{\beta}{2} \right)$ , and the shape anisotropy is  $-(\frac{u_0}{2}) M_s^2$ . The magnetoelastic term is proportional to both the magnetostriction coefficient  $\lambda_{111}$  and to the shear strain which is related to the distortion angle  $\beta$ . The shear modulus is  $c_{44} = 76.4$  GPa<sup>[31]</sup> for YIG at room temperature, and 74–90 GPa for rare earth iron garnets. The magnetocrystalline anisotropy is the smallest contribution to  $K_u$ , and the magnetoelastic anisotropy competes with and overcomes the shape anisotropy.

### 3.3. Element-Specific Ferrimagnetic Configuration

X-ray magnetic circular dichroism (XMCD) measurements were carried out to determine the relative orientations of the magnetic moments of the Tm<sup>3+</sup> and Fe<sup>3+</sup> cations. XMCD is derived from X-ray absorption spectra (XAS) which clearly show a helicity-dependent X-ray absorption (Figure 2e,f, upper panels).

The XMCD signal is calculated as  $I_{asy} = \frac{I_{\sigma^-} - I_{\sigma^+}}{I_{\sigma^-} + I_{\sigma^+}}$ , where  $I$  is the total electron yield (TEY)-XAS intensities for the respective helicities of the emitted light (Figure 2e,f, lower panels).

The XMCD spectra around the Fe  $L_{2,3}$  edges are consistent with antiferromagnetic coupling between Fe in the a-sites (octahedral, two per formula unit) and d-sites (tetrahedral, three per formula unit) in the garnet unit cell, as expected for iron garnets. The spectra around the  $M_{4,5}$  edges of Tm confirm that the Tm<sup>3+</sup> ions are magnetized antiparallel to the a-site Fe<sup>3+</sup> ions.<sup>[32]</sup> This agrees with the fact that the d-site Fe<sup>3+</sup> peak is at lower photon energy than the a-site Fe<sup>3+</sup> peak, similar to the general case of iron spinels.<sup>[33,34]</sup> The spectral shape of the core spectra is characteristic of the Tm<sup>3+</sup> 4f<sup>12</sup> configuration.<sup>[35]</sup>

Our temperature-dependent XMCD measurements (not shown here) indicated that no compensation point was present for the TmIG thin films in the temperature range between 1.5 and 300 K, i.e., the net magnetization remains parallel to that of the tetrahedral Fe<sup>3+</sup>. The existence of a compensation temperature in bulk TmIG has not been confirmed.<sup>[36,37]</sup>

## 4. Spintronic Interface Properties of TmIG/Pt Heterostructures

### 4.1. Spin Hall Magnetoresistance

We use electrical measurements in GGG/TmIG(8 nm)/Pt(5 nm) heterostructures to access the spin mixing properties of the TmIG/Pt interface as well as measuring the PMA of TmIG. Pt is well suited to convert charge current into a pure spin current and vice versa by virtue of its large spin-orbit coupling combined with low resistivity. Recently, it has been discovered

that Pt in contact with an FMI can show magnetoresistance due to the interaction of the spin Hall effect-induced spin accumulation at the Pt/FMI interface with the magnetization ( $m$ ) of a FMI.<sup>[38]</sup> This so-called SMR increases (decreases) the longitudinal resistance when the spin current is absorbed (reflected) by the FMI.

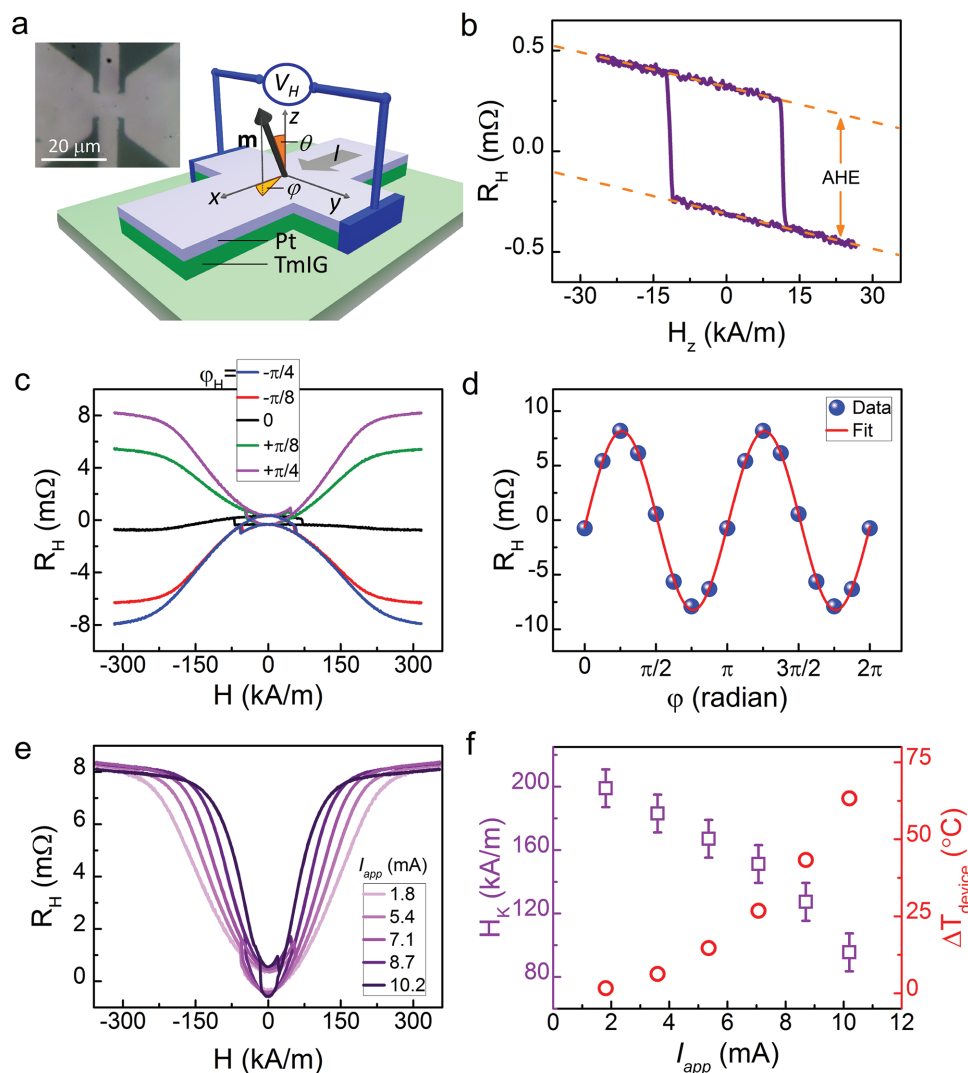
The spin current transmission across interfaces is dictated by the spin mixing conductance concept, which parameterizes the interface transparency to the spin current, and is crucial for SMR,<sup>[16,39]</sup> spin Seebeck effect,<sup>[40]</sup> and spin-orbit torque<sup>[41]</sup> studies. The additional SMR contribution to the longitudinal resistance scales with  $\Delta R \propto m_y^2$ , assuming charge current flow along  $x$ , with  $z$  being the direction normal to the layer plane. The SMR also has a transverse (Hall resistance) component with symmetry  $R_{SMR} \propto m_x m_y$  analogous to the planar Hall effect in ferromagnets. Additionally, it has been theorized<sup>[42]</sup> and measured<sup>[17]</sup> that the imaginary part of the spin mixing conductance gives rise to a transverse anomalous Hall-like signal with symmetry  $R_{AHE} \propto m_z$  that has much lower amplitude compared to  $R_{SMR}$ . By combining these and the ordinary Hall effect of Pt we can write the expected angular symmetry of the transverse Hall effect resistance ( $R_H$ ) as follows

$$R_H = R_{SMR} \sin^2 \theta \sin 2\varphi + R_{AHE} \cos \theta + R_{OHE} H_z \quad (2)$$

where  $\theta$  and  $\varphi$  are the polar and azimuthal angles of  $m$ , respectively, as depicted in Figure 3a, and  $R_{OHE}$  is the ordinary Hall resistance due to an out-of-plane field  $H_z$ .

To characterize the SMR, anomalous Hall effect and ordinary Hall effect (OHE) in 5 nm Pt/8 nm TmIG bilayers we injected an ac current of root mean square amplitude  $I_{ac} = 1.8$  mA with frequency  $\omega/2\pi = 3678$  Hz, and measured the first harmonic Hall voltage as shown schematically in Figure 3a. This is equivalent to standard dc measurements but with higher signal-to-noise ratio. The inset shows the optical microscope image of the device utilized for electrical measurements. Figure 3b depicts the Hall resistance for an out-of-plane field sweep. We recognize the AHE contribution following  $m_z$  which reverses between up and down states at the coercive field of  $\approx 11.9$  kA m<sup>-1</sup>. The linear slope independent of the direction of  $m$  (orange dashed lines) gives  $R_{OHE}$ . This measurement shows that  $m$  has 100% remanence, and sharp transitions between up and down states suggest low pinning and efficient propagation of domain walls, assuming that the reversal is mediated by domain nucleation and propagation.

To determine  $R_{SMR}$  and the spin mixing conductance at the TmIG/Pt interface, we measure  $R_H$  for different in-plane angle  $\varphi$  when the film is saturated in plane. Figure 3d shows  $R_H$  obtained at  $H = \pm 318.3$  kA m<sup>-1</sup> versus  $\varphi$ . We fit the data by using Equation (2) and obtain  $R_{SMR} = 8.2$  m $\Omega$ . By transforming  $R_{SMR}$  and  $R_{AHE}$  into resistivity, taking  $\rho_{Pt}^{Pt} = 40$   $\mu\Omega$  cm as measured on the same device, assuming the spin Hall angle  $\theta_{SH} = 0.08$  for Pt and taking the spin diffusion length to be  $\lambda_{Pt} = 1.4$  nm, we calculate the real and imaginary part of the spin mixing conductance as  $G_r^{\uparrow\downarrow} = 1.3 \times 10^{14}$   $\Omega^{-1}$  m<sup>-2</sup> and  $G_i^{\uparrow\downarrow} = 4.8 \times 10^{12}$   $\Omega^{-1}$  m<sup>-2</sup>, respectively. These values are close to the ones reported for Pt/YIG<sup>[16,17]</sup> and our measurement on another Pt/TmIG device<sup>[9]</sup> and suggest highly efficient transmission of spin current across the Pt/TmIG interface.



**Figure 3.** SMR measurements on a TmIG/Pt bilayer device. a) Schematics of the electrical measurements and the definition of coordinate axes and angles. Upper left: optical image of the measured device. b) Hall resistance as a function of the out-of-plane field showing the anomalous Hall (two levels) and ordinary Hall (linear slope) contributions. c) Hall resistance as a function of an in-plane field sweep applied at different angles  $\varphi$ . d) In-plane angular variation of the Hall resistance recorded at  $H = \pm 318 \text{ kA m}^{-1}$ . Fit according to Equation (2) shows that the signal is purely driven by the SMR. e) Variation of Hall resistance as a function of applied current. Different shapes suggest that the effective anisotropy field depends on  $I_{\text{app}}$  and therefore on the Joule heating. f) Calculated PMA field (left axis) and the variation of the device resistance (right axis) as a function of  $I_{\text{app}}$ . We see that the increase of the resistance due to Joule heating correlates with the decrease in  $H_K$ .

#### 4.2. Measurement of the Total Magnetic Anisotropy

Figure 3c exhibits  $R_H$  for in-plane field sweeps at various  $\varphi$ . In this geometry  $R_{\text{OHE}}$  does not contribute to the signal, and thus we can characterize  $R_{\text{SMR}}$  by analyzing the angular dependence of the signal at high fields where  $m$  tilts in-plane thus  $R_{\text{AHE}}$  becomes negligibly small. The specific U-shape of the signal is due to coherent rotation of the magnetization toward the film plane at higher  $H$ . We find that  $H \geq 199 \text{ kA m}^{-1}$  is necessary to saturate  $m$  fully in-plane, which gives an indication of the effective perpendicular magnetic anisotropy field ( $H_K$ ) of TmIG. Macrospin simulations<sup>[9]</sup> suggest that for this measurement  $H_K \approx 198.9 \pm 15.9 \text{ kA m}^{-1}$ .

The features around  $H = 0 \text{ kA m}^{-1}$  are due to  $R_{\text{AHE}}$  since  $m$  switches between up and down states due to a small unintentional out-of-plane component of the field. With the values for  $H_K$  and  $M_s$ , the total anisotropy energy at room temperature was calculated to be  $K_u = 11.88 \text{ kJ m}^{-3}$ . The contribution from shape anisotropy is well known, but the measurement cannot separately identify the contributions of magnetocrystalline and magnetoelastic anisotropies. However, based on the estimate of  $K_1$  from the literature, magnetoelastic anisotropy is found to be the dominant contribution to producing PMA in the TmIG film. From the negative sign of  $\lambda_{111}$  and the out-of-plane compressive strain state in the film, the magnetoelastic anisotropy favors an out-of-plane easy axis so  $K_{\text{ME}}$  is positive, with magnitude  $\approx 17.5 \text{ kJ m}^{-3}$ .

### 4.3. The Effect of Joule Heating on Magnetic Anisotropy

We now discuss the effect of temperature rise due to Joule heating on  $H_K$ . Figure 3e shows  $R_H$  measured at  $\phi_H = +\pi/4$  with applied current varying between 1.8 and 10.2 mA, corresponding to  $j = 4.8 \times 10^{10}$  and  $2.72 \times 10^{11} \text{ A m}^{-2}$ , respectively. We observe that, although the SMR amplitude remains nearly the same, the field required to saturate  $m$  fully in-plane decreases systematically, as well as the coercivity. We attribute this behavior to  $H_K$  decreasing as a function of increasing temperature due to Joule heating rather than to changes in the  $M_S$ . In Figure 3f we plot  $H_K$  as a function of  $I_{\text{app}}$  (purple squares). When  $I_{\text{app}}$  is increased from 1.8 to 10.2 mA,  $H_K$  decreases more than 50%.

To further support the hypothesis that heating lowers  $H_K$ , we measure the device resistance as a function of  $I_{\text{app}}$  and independently measure the device resistance for low  $I_{\text{app}}$  (i.e., with minimum Joule heating) at different temperatures by placing the device in a heated environment. By correlating these two measurements we can determine the actual changes in the device temperature ( $\Delta T_{\text{device}}$ ) as a function of  $I_{\text{app}}$  (Figure 3f right axis). At 10 mA the temperature is 85 °C (i.e., the temperature increase was 65 °C above ambient). From the XRD data, this corresponds to a lattice distortion angle of 90.75°, compared to 90.77° at room temperature (Figure 1c). The small change in strain between RT and 85 °C, and the dominant magnetoelastic contribution to  $K_u$ , suggest that the change in  $H_K$  is not caused by a change in the magnetocrystalline anisotropy. Instead, we believe the significant fall in  $H_K$  due to heating is primarily governed by a strong temperature dependence of the magnetoelastic coefficient.

The net PMA was confirmed by magnetometry measurements on partly strain-relaxed TmIG films grown on (111)-oriented GGG substrates. These films were grown then immediately annealed for 3 h at the deposition temperature and pressure. XRD scans (not shown) indicated closer to bulk values of the out-of-plane lattice parameters in the films. Despite this, magnetometry measurements showed that the films still exhibited PMA. This shows that PMA could be present even in partly strain-relaxed films with a smaller magnetoelastic anisotropy.

## 5. Conclusion

In summary, we characterized thin film ferrimagnetic garnet TmIG grown epitaxially on GGG that possesses high structural quality and robust perpendicular magnetic anisotropy down to a thickness of 5.6 nm. We showed strong spin mixing conductance at the interface between Pt and TmIG, which led to a large SMR signal in the Pt layer and enabled electrical measurement of the magnetic state of the TmIG. By exploiting the SMR of the TmIG/Pt bilayer, we directly measured the anisotropy field of the TmIG and its temperature dependence due to Joule heating. The PMA is mainly a result of magnetoelastic anisotropy with a contribution from magnetocrystalline anisotropy (see Table 1).

We propose that the high spin mixing conductance and perpendicular anisotropy found in TmIG/Pt heterostructures are promising attributes for future studies of a range of spintronic heterostructures such as FMI/topological insulators and phenomena such as quantum anomalous Hall and proximity

**Table 1.** Summary of some important parameters of the Pt/TmIG heterostructure.

Parameters of Pt/TmIG [8 nm]/GGG with PMA	Value
Measured total anisotropy	$K_u = 11.88 \text{ kJ m}^{-3}$
Shape anisotropy	$K_{\text{shape}} = -5.67 \text{ kJ m}^{-3}$
Magnetoelastic anisotropy	$K_{\text{ME}} = 17.5 \text{ kJ m}^{-3}$
Spin mixing conductance: real part	$G_i^{\uparrow\downarrow} = 1.3 \times 10^{14} \Omega^{-1} \text{ m}^{-2}$
Spin mixing conductance: imaginary part	$G_i^{\uparrow\downarrow} = 4.8 \times 10^{12} \Omega^{-1} \text{ m}^{-2}$

effects. This emphasizes the value of TmIG for spintronic experiments that require FMI layers with PMA.

## 6. Experimental Section

**Fabrication of the Thin Films:** TmIG thin films were deposited using pulsed laser deposition (PLD) on single-crystal GGG substrates with (111) orientation. The stoichiometric target used during the deposition process was made in house by mixing powders of Tm and Fe oxides in the appropriate weight ratios, and then calcining the mix at 1150 °C for 5 h, cold-pressing it into a target, and sintering the target at 1350 °C for 10 h. The stoichiometry of the target was confirmed by wavelength dispersion spectroscopy conducted at various locations on the target surface. To ensure epitaxial growth conditions during the PLD process, a substrate temperature of 650 °C, a laser fluence of  $2 \text{ J cm}^{-2}$ , a laser repetition rate of 10 Hz, and an  $\text{O}_2$  pressure of 200 mTorr were applied. After the deposition the substrate was cooled down to room temperature in 200 mTorr  $\text{O}_2$  at a rate of  $2 \text{ K min}^{-1}$ . It was found that an additional annealing step was not necessary, but the slow cooling step after deposition was crucial in order to obtain the best quality films.

**Transmission Electron Microscopy:** Cross-sectional TEM imaging of the structure was carried out using a FEI TITAN (S)TEM with a CEOS probe aberration corrector operated at 200 kV, which achieves a spatial resolution of less than 0.8 Å. The use of a HAADF detector in STEM mode allowed the authors to record elastic, thermal diffuse scattering events that are proportional to  $Z^2$  and thus determine the position of atom columns or individual atoms as brightness contrast proportional to their atomic number. The TEM lamellas were prepared using a focused ion beam (Zeiss Auriga cross beam system).

**Synchrotron Study:** A detailed investigation of the element-specific magnetic structure of TmIG was conducted on a 19.7 nm thin sample using XMCD at the Diamond Light Source, UK. XAS were collected at the Tm  $M_{4,5}$  and Fe  $L_{2,3}$  absorption edges by TEY measurements, assisted by an ultrathin (3 nm) Au capping layer on top of the TmIG. Oppositely circular polarized X-rays with 100% degree of polarization were used successively to resolve XMCD signals from the respective elements. The light-helicity was switched in a saturating magnetic field of 6 T, which was applied normal to the film plane and in parallel with the incident beam.

**Hall Cross Fabrication:** To study the properties of TmIG as a spininterface material, the authors sputtered 5 nm Pt on top of an 8 nm thin TmIG film. The authors then defined  $8 \times 6 \mu\text{m}^2$  sized Hall crosses in resist by a photolithography process and used ion milling to create Pt/TmIG mesa structures. Using etching instead of a liftoff process improved the quality of the Pt/TmIG interface by avoiding residual resist layers between the TmIG and the Pt.

## Acknowledgements

The authors acknowledge support from C-SPIN, one of the six SRC STARnet Centers sponsored by MARCO and DARPA. A.Q.



acknowledges funding from the Max Planck Society and the Deutsche Forschungsgemeinschaft (DFG, German Research Foundation). J.W.A.R. and W.L. acknowledge support from the EPSRC Programme Grant EP/N017242/1.

Received: September 13, 2016  
Published online: December 12, 2016

- [1] C. Chappert, A. Fert, F. N. Van Dau, *Nat. Mater.* **2007**, 6, 813.
- [2] C.-Z. Chang, J. Zhang, X. Feng, J. Shen, Z. Zhang, M. Guo, K. Li, Y. Ou, P. Wei, L.-L. Wang, Z.-Q. Ji, Y. Feng, S. Ji, X. Chen, J. Jia, X. Dai, Z. Fang, S.-C. Zhang, K. He, Y. Wang, L. Lu, X.-C. Ma, Q.-K. Xue, *Science* **2013**, 340, 167.
- [3] Y. Ando, *J. Phys. Soc. Jpn.* **2013**, 82, DOI: 10.7566/JPSJ.82.102001.
- [4] J. C. C. Slonczewski, *J. Magn. Magn. Mater.* **1996**, 159, DOI: 10.1016/0304-8853(96)00062-5.
- [5] L. Berger, *Phys. Rev. B* **1996**, 54, 9353.
- [6] I. M. Miron, K. Garello, G. Gaudin, P. J. Zermatten, M. V. Costache, S. Auffret, S. Bandiera, B. Rodmacq, A. Schuhl, P. Gambardella, *Nature* **2011**, 476, 189.
- [7] L. Q. Liu, C.-F. Pai, Y. Li, H. W. Tseng, D. C. Ralph, R. A. Buhrman, *Science* **2012**, 336, 555.
- [8] Y. Fan, P. Upadhyaya, X. Kou, M. Lang, S. Takei, Z. Wang, J. Tang, L. He, L. Chang, M. Montazeri, G. Yu, W. Jiang, T. Nie, R. N. Schwartz, Y. Tserkovnyak, K. L. Wang, *Nat. Mater.* **2014**, 13, 699.
- [9] C. O. Avci, A. Quindeau, C.-F. Pai, M. Mann, L. Caretta, A. S. Tang, M. C. Onbasli, C. A. Ross, G. S. D. Beach, *Nat. Mater.* **2016**, DOI: 10.1038/nmat4812.
- [10] P. Li, T. Liu, H. Chang, A. Kalitsov, W. Zhang, G. Csaba, W. Li, D. Richardson, A. DeMann, G. Rimal, H. Dey, J. S. Jiang, W. Porod, S. B. Field, J. Tang, M. C. Marconi, A. Hoffmann, O. Mryasov, M. Wu, *Nat. Commun.* **2016**, 7, 12688.
- [11] C. Hauser, T. Richter, N. Homonnay, C. Eisenschmidt, H. Deniz, D. Hesse, S. Ebbinghaus, G. Schmidt, N. Weinberg, *Sci. Rep.* **2016**, 6, DOI: 10.1038/srep20827.
- [12] Y. Shoji, T. Mizumoto, H. Yokoi, I. W. Hsieh, R. M. Osgood, *Appl. Phys. Lett.* **2008**, 92, 071117.
- [13] B. J. H. Stadler, T. Mizumoto, *IEEE Photonics J.* **2014**, 6, 0600215.
- [14] T. Schneider, A. A. Serga, B. Leven, B. Hillebrands, R. L. Stamps, M. P. Kostylev, *Appl. Phys. Lett.* **2008**, 92, 022505.
- [15] Y. Kajiwara, K. Harii, S. Takahashi, J. Ohe, K. Uchida, M. Mizuguchi, H. Umezawa, H. Kawai, K. Ando, K. Takanashi, S. Maekawa, E. Saitoh, *Nature* **2010**, 464, 262.
- [16] N. Vlietstra, J. Shan, V. Castel, B. J. van Wees, J. Ben Youssef, *Phys. Rev. B* **2013**, 87, 184421.
- [17] S. Meyer, R. Schlitz, S. Geprags, M. Opel, H. Huebl, R. Gross, S. T. B. Goennenwein, *Appl. Phys. Lett.* **2015**, 106, 132402.
- [18] M. Schreier, N. Roschewsky, E. Dobler, S. Meyer, H. Huebl, R. Gross, S. T. B. Goennenwein, *Appl. Phys. Lett.* **2013**, 103, 242404.
- [19] A. Hamadeh, O. D. Kelly, C. Hahn, H. Meley, R. Bernard, A. H. Molpeceres, V. V. Naletov, M. Viret, A. Anane, V. Cros, S. O. Demokritov, J. L. Prieto, M. Munoz, G. de Loubens, O. Klein, *Phys. Rev. Lett.* **2014**, 113, 197203.
- [20] H. Wang, C. Du, P. C. Hammel, F. Yang, *Phys. Rev. B* **2014**, 89, 134404.
- [21] E. Popova, N. Keller, F. Gendron, M. Guyot, M. C. Brianso, Y. Dumond, M. Tessier, *J. Appl. Phys.* **2001**, 90, 1422.
- [22] E. Popova, N. Keller, F. Gendron, L. Thomas, M.-C. C. Brianso, M. Guyot, M. Tessier, S. S. P. Parkin, *J. Vac. Sci. Technol., A* **2001**, 19, 2567.
- [23] M. Kubota, A. Tsukazaki, F. Kagawa, K. Shibuya, Y. Tokunaga, M. Kawasaki, Y. Tokura, K. Shibuya, Y. Tokunaga, M. Kawasaki, Y. Tokura, *Appl. Phys. Express* **2012**, 5, 103002.
- [24] M. Kubota, K. Shibuya, Y. Tokunaga, F. Kagawa, A. Tsukazaki, Y. Tokura, M. Kawasaki, *J. Magn. Magn. Mater.* **2013**, 339, 63.
- [25] A. Paoletti, *Physics Of Magnetic Garnets*, North-Holland Publishing Company, Amsterdam **1978**.
- [26] H. Sawada, *J. Solid State Chem.* **1997**, 132, 300.
- [27] V. V. Randoshkin, V. I. Kozlov, Y. A. Koksharov, A. M. Saletski, N. N. Sysoev, N. N. Usmanov, *Tech. Phys.* **2007**, 52, 495.
- [28] V. J. Fratello, I. Mnushkina, S. J. Licht, R. R. Abbott, *Mater. Res. Soc. Symp. Proc.* **2005**, 834, J6.2.1.
- [29] A. Hubert, R. Schäfer, in *Magnetic Domains*, Springer, New York **1998**, pp. 500–507.
- [30] D. H. Kim, N. M. Aimon, L. Bi, J. M. Florez, G. F. Dionne, C. A. Ross, *J. Phys. Condens. Matter* **2013**, 25, 26002.
- [31] Y. A. Burenkov, S. P. Nikanorov, *Magn. Ferroelectr.* **2002**, 44, 318.
- [32] W. Meyer, W. Sturhahn, E. Gerdau, *Hyperfine Interact.* **1994**, 94, 2059.
- [33] Y. Liu, L. He, Y. Zhou, K. Murata, M. C. Onbasli, C. A. Ross, Y. Jiang, Y. Wang, Y. Xu, R. Zhang, K. L. Wang, *AIP Adv.* **2016**, 6, DOI: 10.1063/1.4943157.
- [34] W. Liu, L. He, Y. Xu, K. Murata, M. C. Onbasli, M. Lang, N. J. Maltby, S. Li, X. Wang, C. A. Ross, P. Bencok, G. van der Laan, R. Zhang, K. L. Wang, *Nano Lett.* **2015**, 15, 764.
- [35] B. T. Thole, G. van der Laan, J. C. Fuggle, G. A. Sawatzky, R. C. Karnatak, J.-M. Esteve, *Phys. Rev. B* **1985**, 32, 5107.
- [36] S. Geller, J. P. Remeika, R. C. Sherwood, H. J. Williams, G. P. Espinosa, *Phys. Rev.* **1965**, 137, 1034.
- [37] R. L. Cohen, *Phys. Lett.* **1963**, 5, 177.
- [38] H. Nakayama, M. Althammer, Y. T. Chen, K. Uchida, Y. Kajiwara, D. Kikuchi, T. Ohtani, S. Geprags, M. Opel, S. Takahashi, R. Gross, G. E. W. Bauer, S. T. B. Goennenwein, E. Saitoh, *Phys. Rev. Lett.* **2013**, 110, 206601.
- [39] M. Weiler, M. Althammer, M. Schreier, J. Lotze, M. Pernpeintner, S. Meyer, H. Huebl, R. Gross, A. Kamra, J. Xiao, Y. T. Chen, H. J. Jiao, G. E. W. Bauer, S. T. B. Goennenwein, *Phys. Rev. Lett.* **2013**, 111, 176601.
- [40] K. I. Uchida, H. Adachi, T. Ota, H. Nakayama, S. Maekawa, E. Saitoh, *Appl. Phys. Lett.* **2010**, 97, 18.
- [41] C. F. Pai, Y. X. Ou, L. H. Vilela-Leao, D. C. Ralph, R. A. Buhrman, *Phys. Rev. B* **2015**, 92, 64426.
- [42] Y. T. Chen, S. Takahashi, H. Nakayama, M. Althammer, S. T. B. Goennenwein, E. Saitoh, G. E. W. Bauer, *Phys. Rev. B* **2013**, 87, 144411.