Spin-reorientation in the heterostructure Co/SmFeO$_3$

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Abstract
Element-specific imaging of magnetic domains in a ferromagnet/antiferromagnet
heterostructure consisting of Co/SmFeO₃ has been performed in the vicinity of the
spin-reorientation phase transition in SmFeO₃ using a photoemission electron microscope.
Evidence is shown that a 90° in-plane spin-reorientation in the antiferromagnetic SmFeO₃
triggers a similar reorientation of spins of the ferromagnetic Co layer on top of it. The
possibility of triggering the spin-reorientation with the help of laser-excitation is demonstrated.

Heterostructures consisting of ferromagnetic and antiferromagnetic thin films are indispensable building blocks in magnetic data storage applications [1]. In these systems, the antiferromagnet is used to pin the magnetization direction of the ferromagnet via the exchange bias effect [2]. For changing the magnetization direction, the antiferromagnet is usually treated as a static system and only the ferromagnet is considered as the dynamic part. Recent work, for example on NiO [3–5], has shown that the antiferromagnet is not static but can change its magnetic configuration due to the deposition of a ferromagnetic layer. An intriguing possibility is now the reverse, to control the spins of the ferromagnet using the dynamics of the antiferromagnet. The spin-reorientation of an antiferromagnet could be a possible solution and, for example, the rare-earth orthoferrites RFeO₃ (with R a rare-earth element) are known to have a strong temperature dependent anisotropy [6, 7]. However, whether the spins of a ferromagnetic layer placed on top of RFeO₃ are coupled to the antiferromagnet and can indeed be manipulated using the spin-reorientation in the orthoferrite remains a question since it is not clear whether the coupling will be maintained during the transition.

In this paper we show evidence that the spin-reorientation transition in SmFeO₃ can be used to change the magnetization direction of a ferromagnetic Co layer on top of it. Using a photoemission electron microscope (PEEM) we investigated the orientation of the antiferromagnetic axis of the SmFeO₃ employing x-ray magnetic linear dichroism (XMLD) and of

the ferromagnetic axis of the Co employing x-ray magnetic circular dichroism (XMCD) as a function of the temperature. Besides using resistive heating we demonstrate that the reorientation of the Co layer can be induced by laser heating which paves the way for ultra-fast switching of this system, since it has been shown that the spins of an antiferromagnet can be manipulated on a timescale of a few picoseconds in contrast to the hundreds of picoseconds in a ferromagnet [8].

SmFeO₃ is a canted antiferromagnet. The spins of four sublattices S_1, S_2, S_3, and S_4 are antiferromagnetically coupled with the antiferromagnetic vector $\mathbf{L} = \mathbf{S}_1 - \mathbf{S}_2 + \mathbf{S}_3 - \mathbf{S}_4$, see inset in figure 1. Due to the Dzialoshinsky–Moria antisymmetric exchange the spins are slightly canted and a weak ferromagnetic moment arises $\mathbf{M} = \mathbf{S}_1 + \mathbf{S}_2 + \mathbf{S}_3 + \mathbf{S}_4$ in a direction perpendicular to $\mathbf{L}$ [9, 10]. In bulk SmFeO₃ below $T = 460$ K the vectors $\mathbf{L}$ and $\mathbf{M}$ are aligned along the a-axis [100] and the c-axis [001], respectively. In the range between 460 and 470 K the spins continuously rotate over 90° in the (010) plane so that above 470 K $\mathbf{L}$ and $\mathbf{M}$ are aligned along the c-axis and the a-axis, respectively. For our experiments a bulk single crystal of SmFeO₃ with a b-surface ([010]) orientation was first polished, then sputtered and plasma cleaned before a 2 nm Co layer and a 1 nm Pt capping layer were deposited with magnetron sputtering. A reference sample for optical measurements was polished to a thin slice and the birefringence measurement showed an in-plane spin-reorientation at around 465 K, as shown in figure 1. Magnetic imaging was performed with an Elmitec PEEM at the Surface/Interface: Microscopy (SIM) beamline at the Swiss Light Source. The ⩾98% linear or ⩾98% circular polarized x-rays impinged on the sample at a

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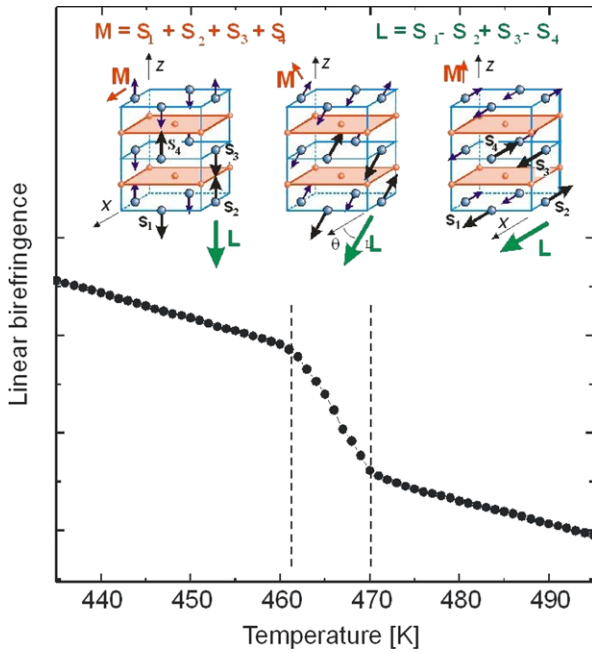
Figure 1. Linear optical birefringence in SmFeO$_3$ as a function of temperature. Insets show the corresponding arrangement of spins. The change of birefringence in the range of 460–470 K is due to the reorientation of the vector **L** in the *x*–*z* plane.

(This figure is in colour only in the electronic version)

is obtained by subtracting a spectrum taken with the linear polarization vector **E** perpendicular to the sample plane from a spectrum recorded with **E** parallel to the sample plane. The sample is heated either by a filament mounted in the sample holder or by a picosecond pulse laser (wavelength 532 nm, repetition rate 62.5 MHz). The temperature is measured with a thermocouple mounted on the sample holder.

In order to determine the orientation of the antiferromagnetic axis at room temperature the XMLD spectra of the Fe L$_{3,2}$ edge was measured for different azimuthal orientations of the sample. Figure 2(a) shows the x-ray absorption and XMLD spectrum for x-rays parallel to the *a*-axis, and figure 2(b) with x-rays parallel to the *c*-axis. While a strong dichroism was observed for x-rays parallel to the *a*-axis no significant dichroism was detected for x-rays parallel to the *c*-axis, showing that at room temperature the antiferromagnetic axis is parallel to the *c*-axis as expected [9].

The temperature dependence of the Co magnetization direction is determined by taking XMCD images for one sample angle with increasing temperature. After cooling down to room temperature the measurement is repeated with a 90° rotated sample. Note that the crystallographic axis of the SmFeO$_3$ is used to describe the direction of the polycrystalline Co film. The Co XMCD images at room temperature and above the spin-reorientation transition at 485 K are shown in figure 3(a). The black (white) areas are domains pointing parallel (antiparallel) to the x-ray directions whereas gray corresponds to a perpendicular alignment. While at room temperature x-rays parallel to the *a*-axis yield a black/white contrast, above the spin-reorientation transition x-rays parallel to the *c*-axis give rise to a black/white contrast. Hence, the orientation of the Co film changed by 90° with temperature from parallel to the *a*-axis to parallel to the *c*-axis and is collinear with the direction of the weak ferromagnetic moment of SmFeO$_3$.

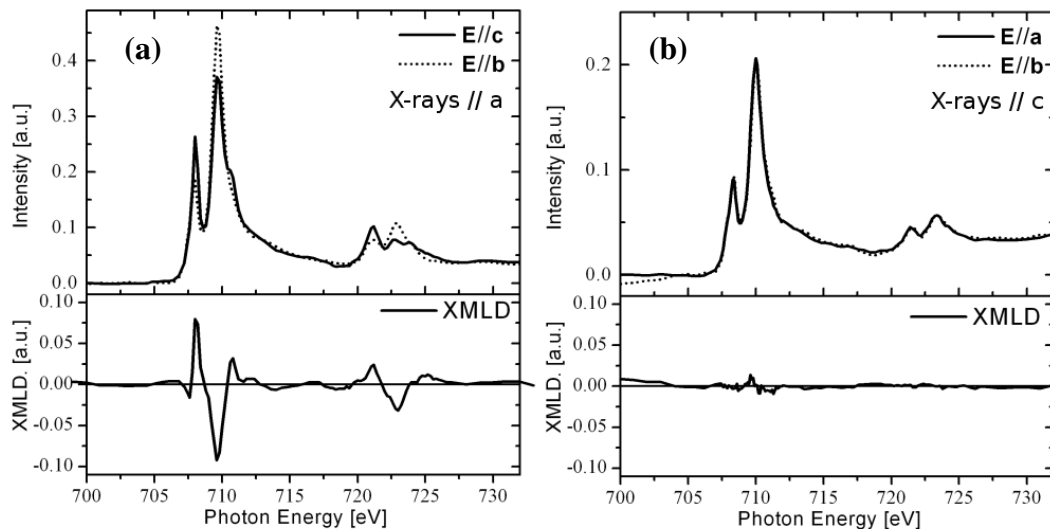In order to obtain a more detailed picture of the reorientation process we analyzed the contrast change of

grazing incidence of 16°. Dividing two images taken at the Co L$_3$-edge with left and right circular polarized light leads to an XMCD image where the intensity *I* is a measure of the angle α between the x-ray propagation direction and the magnetic spins in the ferromagnetic domains $I \propto \cos(\alpha)$ [11]. The antiferromagnetic SmFeO$_3$ is studied using linearly polarized x-rays. Dividing images taken at two peaks of the multiplet structure at the Fe L$_3$ absorption edge, i.e. at 708.9 and 709.6 eV, an XMLD image is obtained. An XMLD spectrum

Figure 2. X-ray absorption and XMLD spectrum at room temperature at the Fe L$_{3,2}$ edge with linearly polarized light with (a) x-ray along the *a*-axis, i.e. **E** vector along the *c*- and *b*-axes, (b) x-ray along the *c*-axis, i.e. **E** vector along the *a*- and *b*-axes.
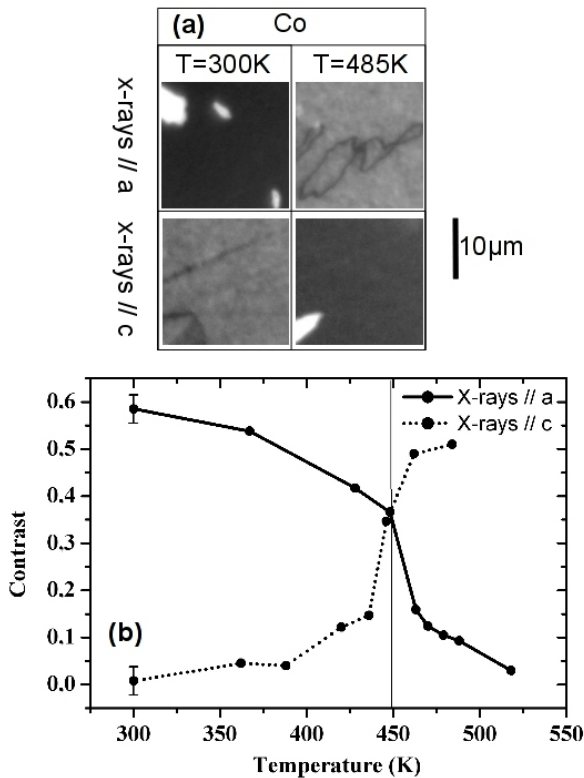
Figure 3. (a) XMCD images at the Co L_3 edge at room temperature and at 485 K. (b) Contrast of the XMCD images as a function of temperature for the two different sample orientations. The lines are guides to the eyes.

Figure 4. (a) XMLD images at the Fe L_3 edge at room temperature and at 485 K. (b) Contrast of the XMLD images as a function of temperature for the two different sample orientations. The lines are guides to the eyes.

the XMCD images by plotting the difference between the black and white areas as a function of temperature. For x-rays along the a-axis we observed a strong contrast at room temperature, decreasing with increasing temperature and with an abrupt drop around 450 K. Correspondingly, when the x-rays are along the c-axis (90° rotated sample), we observed a small contrast at room temperature, which increases with temperature and saturates above 455 K. From figure 3(b) we can deduce that the Co magnetization rotated from the a-axis at room temperature to the c-axis for higher temperature and that the spin-reorientation transition takes place around 450 K. This 90° rotation is reversible with temperature but the Co domain configuration is not identical. However, the Co domain orientation was always uniaxial which is not expected for polycrystalline Co film and can only be due to the coupling to the antiferromagnet as has been observed in exchange bias systems, see for example [12, 13].

We investigated the antiferromagnetic axis orientation as a function of the temperature by using Fe XMLD images for the two orientations of the sample which are represented in figure 4(a). The experimental procedure is the same as for the Co images above. The XMLD images of the SmFeO$_3$ have a homogeneous contrast, indicating that the SmFeO$_3$ is in a single domain state. Only some lines with a low contrast are visible, which probably stem from the polishing. Similar lines have been observed on NiO surfaces [14] and were linked to a pinning of the antiferromagnetic regions to the surface structure.
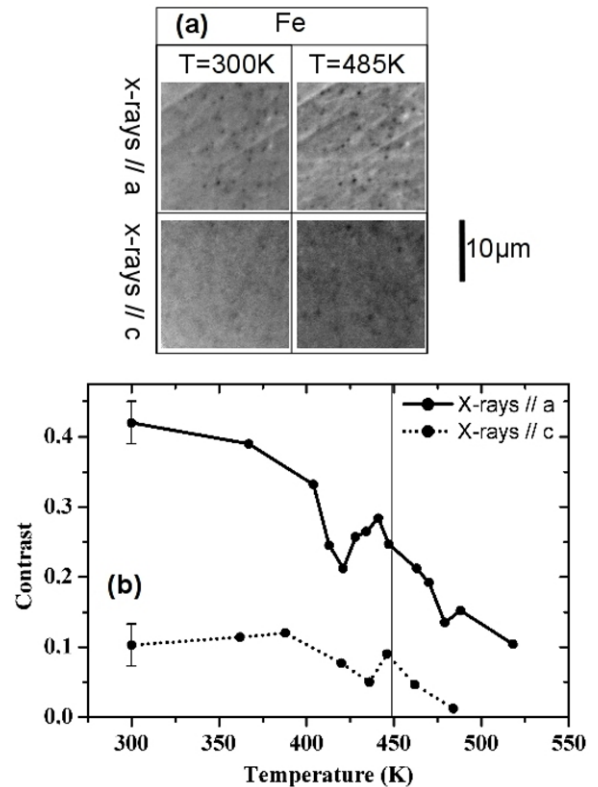
The detailed temperature dependence of the Fe XMLD signal is plotted in figure 4(b) for the two orientations of the sample, where the signal is the difference of the intensity of the Fe XMLD spectrum at the L_3 multiplet edges. At room temperature and for x-rays along the a-axis we observed an XMLD signal globally decreasing with increasing temperature but with an increase around the spin-reorientation transition. For the 90° rotated sample, the XMLD signal is weak and more or less constant up to 425 K and then decreases with temperature.

The behavior shows two surprising characteristics. First, while the behavior for x-rays parallel to the a-axis is consistent with a rotation of the antiferromagnetic axis from the c-axis to the a-axis, no increase of the XMLD signal for x-rays parallel to the c-axis is observed. Second, around the spin-reorientation temperature a non-monotonic behavior is observed. In particular, the first point is surprising since we clearly see a reorientation of the Co film which can only be induced by a change of the antiferromagnet. Most likely this is due to a reduction of the Néel temperature of the SmFeO$_3$ surface with respect to the bulk behavior [15, 16]. The observed XMLD signal is a superposition of the reorientation of the antiferromagnetic axis with temperature and the reduction of the magnetization when approaching the Néel temperature. As a consequence, the antiferromagnetic axis of SmFeO$_3$ can be unambiguously determined at room temperature but above the spin-reorientation the overall XLMD signal is too small for a precise determination. Taking a closer look at the lines we
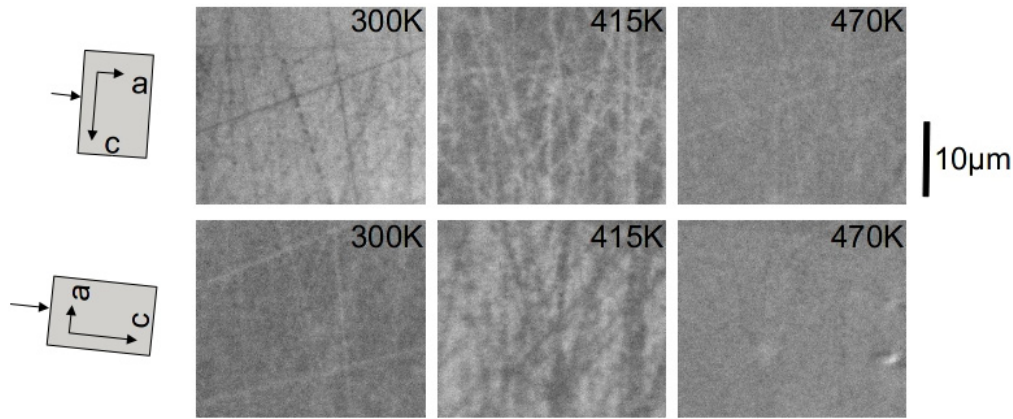
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Figure 5. XMLD images at the Fe L_3 edge for both directions of the sample and for temperatures below, above, and well above the spin-reorientation.

find an indication of the spin-reorientation. In figure 5 XMLD images of the reference sample for optical measurements are shown. The sample was prepared in the same way as described above but resulted in a smoother surface. The Co domain structure showed the same in-plane rotation (not shown) and the XMLD images revealed again a homogeneous contrast but now with much better visible lines, which might be due to the improved spatial resolution. On azimuthal rotation of the sample the contrast is inversed which indicates an in-plane variation of the antiferromagnetic axis. Upon heating the sample above the spin-reorientation temperature a contrast reversal of these lines is also observed, e.g. for x-rays parallel to the *c*-axis from bright lines on a dark background to dark lines on a bright background. Still, the global XMLD signal for temperatures above the spin-reorientation is zero[5].

The non-monotonic behavior of the Fe XMLD signal around the spin-reorientation transition could be associated with the recently discovered anisotropy of the XMLD [17–19]. The XMLD depends on the relative orientation of the polarization vector, the magnetization direction and the crystallographic axis. During the spin-reorientation transition the antiferromagnetic axis is rotating with respect to the crystallographic axis, resulting in a change of the XMLD signature. A more detailed study would be necessary to reveal the details of this spin-reorientation transition which is outside the scope of this paper.

For applications a faster stimuli than filament heating will be needed. Therefore we heated the sample using a picosecond pulsed laser incoming onto the sample with a grazing incidence of 16° with a minimum size of the laser spot of about 10 μm $\times$ 70 μm and a tunable power between 0 and 1 W. In order to minimize the needed heating power we placed the temperature of the sample at 425 K, which is just below the spin-reorientation transition.

As can be seen from figure 6, for x-rays parallel to the *a*-axis black and white areas are observed with the laser switched off while with the laser on only gray areas are visible. Measurements of the 90° rotated sample confirmed that this

Figure 6. Co XMCD images at the L_3 edge for a temperature of 425 K with the laser switched off (left column) and on (right column) for the x-rays along the *a*-axis (a) and along the *c*-axis (b).

contrast change is not due to a demagnetization of the sample, which can be seen in figure 6(b). It should be noted that now the contrast with the laser off is not completely gray since we are close to the spin-reorientation temperature. As with the resistive heating this means that the Co magnetization rotated from the *a*-axis to the *c*-axis. Also, as for the resistive heating, only a reduction of the $SmFeO_3$ XMLD signal is observed. Since it is very unlikely that the laser heating of the Co film leads to this reorientation it must be related to the coupling to the antiferromagnet.

In conclusion we studied the spin-reorientation in a heterostructure and showed strong evidence that a 90° in-plane rotation of the Co magnetization can be induced by the spin-reorientation of antiferromagnetic $SmFeO_3$. The coupling of the Co film to the $SmFeO_3$ is maintained during this spin-reorientation which can be controlled by heating with a filament or using laser irradiation. The triggering of the Co

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[5] The variation of the observed temperature of the spin-reorientation might be due to the mounting of the thermocouple on the sample holder and the different thickness of the $SmFeO_3$ samples.
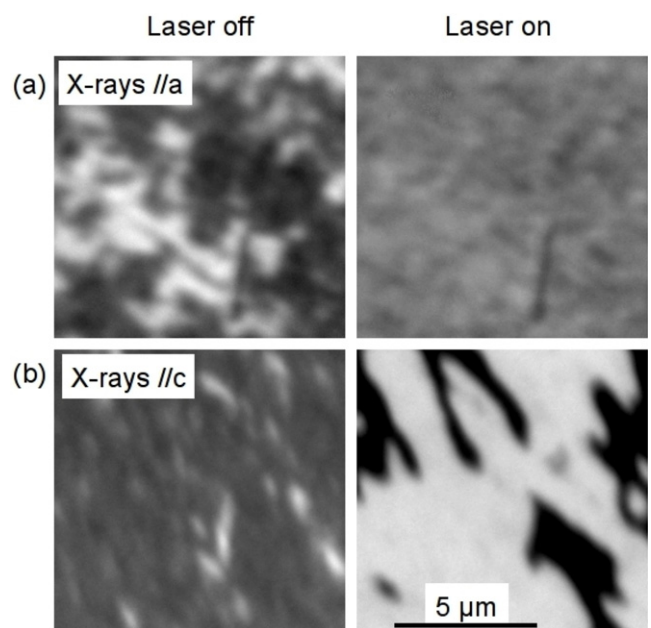
magnetization rotation by a laser is the first step towards ultra-fast switching employing the spin-reorientation transition in an antiferromagnet.

Acknowledgments

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