Spin Reorientation Transitions in Perpendicularly Exchange-Coupled Thin Films Studied Using Element Specific Imaging

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Spatial variations in the spin-reorientation transition of an exchange-coupled Co-wedge/YIG ($Y_3Fe_5O_{12}$) bilayer, from perpendicular to in-plane domain structure, was studied using magnetic force microscopy (MFM) and photo-emission electron microscopy (PEEM). Even though MFM measurements of the YIG film showed perpendicular stripe domains, it was not possible to unambiguously resolve the domain structure of only the top ferromagnetic metal layer because of complications arising from the stray fields of the much thicker YIG underlayer. Hence, using element-specific, X-ray magnetic circular dichroism (XMCD) of the transition metal $L_{3,2}$ edges (Co and Fe, respectively) for magnetic contrast, PEEM measurements were carried out to resolve the domain structure of the individual Co and YIG layers. The two were identical up to a Co thickness of 4.5 nm, confirming that the Co layer was exchange coupled with the YIG underlayer; however, a transition of the Co domains, from perpendicular to in-plane, was observed at thickness of 4.5–6 nm. For thicker regions of the Co film (>6 nm), a sizeable portion of the Co layer showed in-plane domains; their spins were perpendicular to the domain walls of the stripe domains with Co XMCD values in the range of -5% to $+5\%$—a value much smaller than that of the perpendicular domains.

Index Terms—**Element specific imaging, exchange coupling, perpendicular domain, spin reorientation.**

I. Introduction

CONTROLLING the domain structure of thin ferromagnetic (FM) films, especially to orient their magnetization perpendicular to the film plane, is an ongoing challenge. This is because the magnetization of most FM thin films is dominated by their magnetostatic energy and result in their orientation parallel to the film surface. However, in ulltrathin films (less than a few AL thick, AL: atomic layer) [1] or in multilayer thin film structures [2], the break in symmetry either at the surface or interface, respectively, can give rise to an out of plane component that can overcome the shape anisotropy and exhibit a perpendicular magnetization. Alternatively, an out-of-plane anisotropy can also be achieved by the epitaxial growth of a thin film with strong uniaxial anisotropy such that its easy axis is oriented along the film normal and its magnetocrystalline energy becomes dominant [3]. Moreover, YIG thin films, usually fabricated by liquid phase epitaxy, also exhibit perpendicular magnetization arising from a growth induced anisotropy with strong contribution from the preferential site occupation of the rare earths in the garnet lattice [4], [5]. A defining characteristic of these garnet films is the observation of a stripe domain structure [6], with an out-of-plan magnetization component, arising from a balance between the magnetostatic and uniaxial anisotropy energies. Here, we demonstrate the possibility of stabilizing perpendicular magnetization in a thin metallic (Fe or Co) film, up to 4.5 nm in thickness, by growing it such that it is strongly exchange coupled to a garnet underlayer. This behavior is unlike that of a thin cobalt film [7], which is known to exhibit a stripe domain structure only for thicknesses greater than 40 nm. Moreover, the stripe period of the Co layer in the bilayer structure is not independent but mimics that of the garnet underlayer. This maybe an effective way to modulate the domain period of the metal layer by controlling the period of the garnet layer. We further demonstrate that magnetic imaging of such a bilayer (FM-metal/Garnet-underlayer) by magnetic force imaging is limited, and an unequivocal interpretation of the magnetic structure requires element specific imaging methods such as Photoemission electron microscopy (PEEM), with X-ray magnetic circular dichroism (XMCD) for magnetic contrast. Finally, such PEEM imaging reveals a spin reorientation transition in both layers at thicknesses greater that 4.5 nm.

II. Experimental Method

Bilayer films of FM metal and YIG were deposited on GGG (Gadolinium–Gallium–Garnet) substrate, which has a lattice constant and thermal expansion coefficient similar to those of YIG. First, a YIG film was deposited (6 h at 50-W RF power) on a GGG single crystal substrate at room temperature using magnetron sputtering. The thickness of the YIG film was optimized at $\sim$890 nm to maximize the perpendicular anisotropy, post-annealed at $\sim$1050 °C to crystallize the film and magnetized by applying a magnetic field loop of $\pm$3000 G. The domain structure was initially measured by magnetic force microscopy (MFM), a routinely used method to observe the domain structure of thin films, with contrast proportional to the stray field gradient. Next, a ferromagnetic metal thin film (Fe or Co) with thickness ranging from 0–20 nm was deposited at room temperature on the YIG underlayer using ion beam sputtering. After growth of the bilayer the domain structures of these films, measured by MFM, show a strong influence of the stray fields emanating from the under-layer, making it difficult to resolve the domain structure of the individual layers. So, element specific XPEEM (XMCD-PEEM) measurement was used to study the FM(Co-wedge)/YIG bilayer system. For element specific XPEEM measurement, the post-annealed YIG film was transported to the PEEM chamber and then a wedge shaped Co thin film Fig. 2(e), with thickness varying from 0 to 15 nm over a distance of 3 mm, was deposited *in situ* using a controlled moving shutter. The domain structures of both Co and YIG in the Co-wedge/YIG bilayer were then measured as a function of position each corresponding to a different thickness of the Co (top) layer.

III. Results and Discussion

The observation of stripe domains in a single YIG film by MFM [8] measurements, as shown in Fig. 1(c), confirmed the

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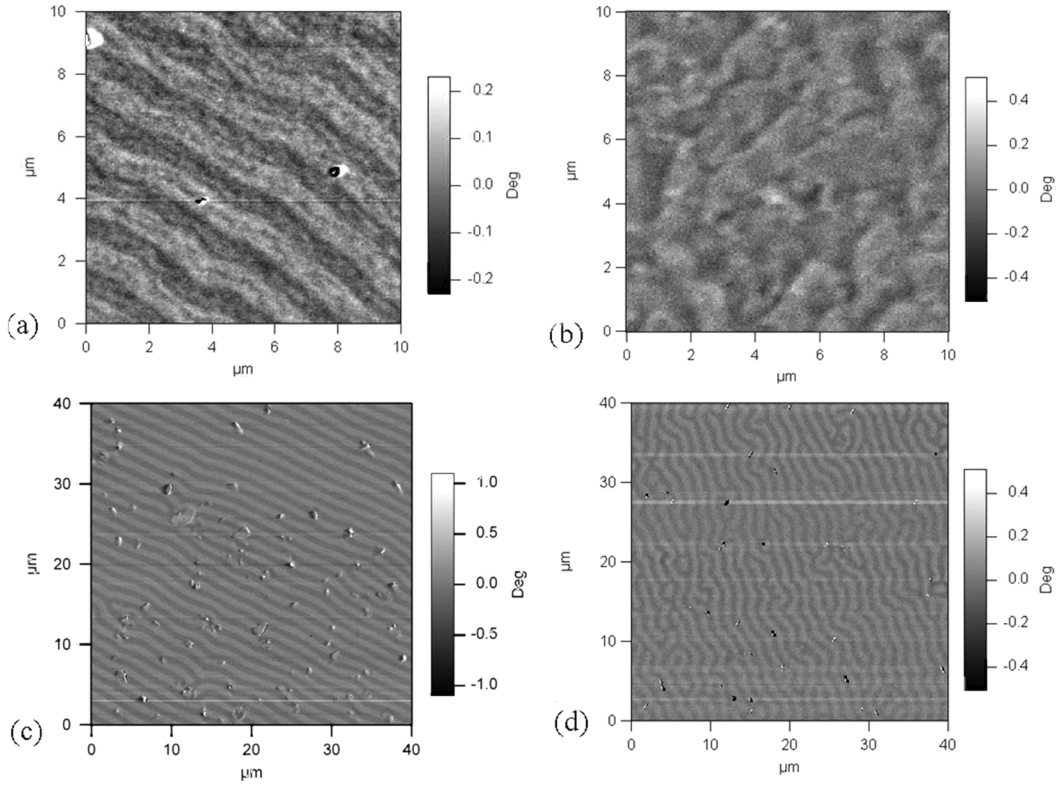

Fig. 1. Domain structure measured by MFM for the samples of (a) Fe(20 nm)/YIG: 10×10 μm scan area; (b) Fe(20 nm)/Al(2 nm)/YIG: 10×10 μm scan area; (c) YIG only: 40×40 μm scan area; (d) Cu (50 nm)/YIG: 40×40 μm scan area.

perpendicular domain structure in our high-quality garnet thin films [9]. The Fe/YIG bilayer samples with different Fe thicknesses, prepared by ion beam sputtering were then evaluated. MFM images of these Fe/YIG samples showed the same stripe domain structure for Fe thickness up to 20 nm Fig. 1(a). This may suggest that the domain structure of the Fe film, grown on top of the YIG, is magnetically very well exchange-coupled to the YIG underlayer. However, MFM measurement of a Cu(50 nm)/YIG bilayer control sample still showed the same stripe domain image Fig. 1(d). Since Cu is a non-magnetic thin film, the stripe domain images must be the result of the interaction between the MFM tip and the stray field gradients from the YIG underlayer. Hence we concluded that in the magnetic bilayer films, the MFM image of the top layer may not necessarily represent the true domain structure of the metal layer. However, the existence of exchange coupling between the FM metal (Fe) and the YIG layer at the interface was indirectly confirmed by comparing the domain images of a Fe(20 nm)/YIG bilayer with that of a Fe(20 nm)/Al(2 nm)/YIG trilayer sample [Fig. 1(a) and (b), respectively]. In the latter case, the Al (2 nm) buffer layer was used to break the exchange interaction at the Fe-YIG interface. The domain image of Fe(20 nm)/Al(2 nm)/YIG trilayer did not show the stripe domain images and only showed a uniform contrast, which is the indication of in-plane domains. From these MFM measurements of the bilayer and trilayer samples, it is clear that both the exchange interactions at the Fe-YIG interface and the stray fields from the underlying YIG layer contribute to the observed stripe domains. However, it was not possible to unambiguously determine, separately and independently, the domain structure of the top ferromagnetic metal layer and YIG underlayer. Hence, to resolve the domain structures of the individual components of such a bilayer, additional element specific domain imaging method, such as XPEEM, was performed on the *in-situ* grown Co-wedge/YIG bilayer. In XPEEM at the ALS [10], element-specific contrast for the two bilayers is achieved by tuning the incident X-ray wavelength through the $L_{3,2}$ absorption edges of the transition metals and using XMCD, which permits the determination of spin and orbital moments using sum rules [11], to obtain the magnetic contrast.

XPEEM measurements [12], [13] were carried out on a Co-wedge/YIG bilayer using element-specific XMCD of the principal transition metal $L_{3,2}$ edges of Co and Fe (in YIG) for magnetic contrast. The spatially-resolved domain structure of the individual Co and YIG layers were measured by XPEEM as shown in Fig. 2. The domain structure of a pure YIG sample showed the same stripe domain images, with the same domain width, in both XPEEM and MFM measurements. Also, the stripe domain structures of the Co layer, up to a thickness $t_{Co} = 4.5$ nm, in the Co-wedge/YIG bilayer were identical with that of YIG layer, with a stripe domain width of 2 μm [Fig. 2(a) and (b)]. Since, in earlier measurements [7], stripe domains images were only observed in much thicker (>40 nm) films of epitaxially grown Co and with a stripe width of $85 \sim 250$ nm, these results clearly show that the perpendicular anisotropy in the Co layer arises from the exchange coupling at the Co/YIG interface. Moreover, the line scan data (XMCD intensity) of the domain images of Co and YIG from exactly the same area shows a very good match in all details (peak and crossing point values) of the magnetic structure. However, as the Co thickness increased above $t_{Co} > 4.5$ nm the domain pattern exhibited by the Co layer is increasingly determined by the Co bulk anisotropy that forces the magnetization of the Co layer to be in-plane as indicated by the gray contrast in the XPEEM images Fig. 2(c). This is clear from the XPEEM images of the Co layer for $t_{Co} > 6$ nm. In addition, the Fe image, representative of the YIG layer, shows not only black

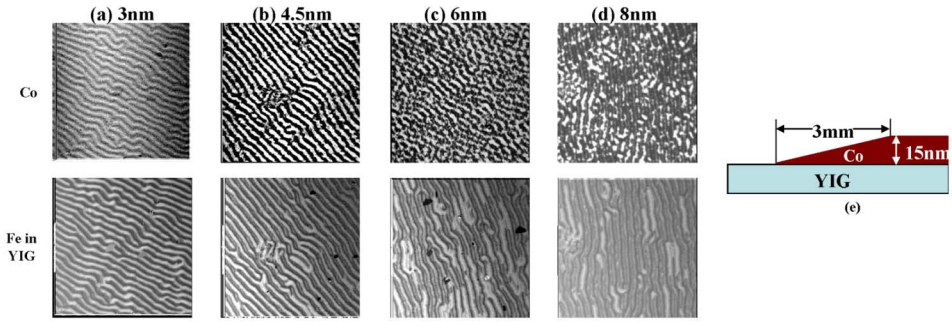

Fig. 2. Domain images of the individual Co and YIG layers measured by XPEEM as the thickness of Co is varied: (a) 3, (b) 4.5, (c) 6, and (d) 8 nm. All scan sizes are $40 \times 40\ \mu$m. The stripe domain structure was identical (shape and period) in both Co and YIG layers for Co thickness <4.5 nm. However, as the Co thickness increased, an in-plane domain component (gray contrast) was observed in the domain images of both Co and YIG layers. The top row shows the domain images of the Co layer, and the bottom row shows the domain images of the YIG layer. (e) Illustration of Co-wedge/YIG sample; the thickness of Co was varied from 0 to 15 nm over the 3-mm distance.

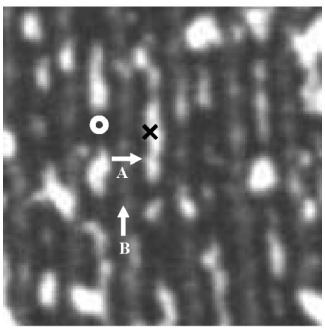

Fig. 3. Domain configuration of the Co film at $t_{Co} = 8$ nm: o (black region) and x (white region) are the perpendicular domain structure. The in-plane domain can be magnetized either perpendicular (Case **A**: Co XMCD $\sim$5%), or parallel (Case **B**: Co XMCD= $\pm$10%) to the stripe.

and white contrast but also some intermediate grey contrast indicating that the YIG layer starts orienting in-plane as well. At this point the Co layer shows a complicated domain patterns and future micromagnetic simulation (in progress) will show precisely what the exact reasons are for this behavior.

The in-plane domain elements (gray contrast) of Co at $t_{Co} = 8$ nm may have their magnetization direction either perpendicular to the stripe domain wall (case A) or parallel to the stripe (case B) as illustrated in Fig. 3. A quantitative analysis of the XMCD/PEEM intensity spectrum of the gray regions shows a variation of $\sim \pm 5\%$ and is much smaller than the black and white regions corresponding to the up and down contrast in the stripe domains. For the experimental geometry used, in order to have a parallel orientation of the in-plane elements (case B), the observed XMCD intensities should be higher $(\sim \pm 10\%)$ than that of the perpendicularly magnetized regions because the incident X-ray beam would make a smaller angle with the parallel in-plane magnetization direction (Case B) than that with the up and down spin directions. Hence, we conclude that the magnetization direction in the in-plane regions (gray contrast) is rather perpendicular to the domain walls (Case A) than parallel to the domain wall. Also, comparing the domain configuration of both the parallel and perpendicular cases, the perpendicular in-plane domain configuration shows an overall facile flux closure configuration; this would correspond to a lower magnetostatic energy state.

IV. Conclusion

In this study, the direct exchange coupling of a FM metal layer and a magnetic oxide garnet (YIG) film was studied. To overcome the stray filed affect from underlayer and the inability of MFM measurements to individually resolve the magnetization of each component of a bilayer thin film sample, an element specific domain imaging technique, XPEEM was employed. It clearly confirmed the perpendicular exchange coupling at the Co-YIG bilayer interface, and the domain structure of FM (Co) was controlled by the YIG under layer. Further, it showed a spin reorientation transition of FM metal (Co), at a thickness $t_{Co} \sim 4.5$ nm, in the perpendicularly exchanged coupled bilayer samples.

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