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Quantitative XMCD mapping with high spatial resolution full-field magnetic transmission soft x-ray spectro-microscopy

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ABSTRACT

The spectroscopic analysis of X-ray magnetic circular dichroism (XMCD), which serves as strong and element-specific magnetic contrast in full-field magnetic transmission soft x-ray microscopy (MTXM) is shown to provide information on the local distribution of spin (S) and orbital (L) magnetic moments down to a spatial resolution of 25nm limited by the x-ray optics used in the x-ray microscope. The spatially resolved L/S ratio observed in a multilayered (Co 0.3nm/Pt 0.5nm)x30 thin film exhibiting a strong perpendicular magnetic anisotropy decreases significantly in the vicinity of domain walls, indicating a non-uniform spin configuration in the vertical profile of a domain wall across the thin film. Quantitative XMCD mapping with x-ray spectromicroscopy will become an important characterization tool for systems with topological or engineered magnetization inhomogeneities.

I. INTRODUCTION

The local distribution of inhomogeneities in magnetization plays a pivotal role in understanding magnetic nanoscale behavior [1] and harnessing magnetic properties for technological applications where the quest for increased information density, speed, and reliability poses significant challenges to control magnetization on the nanoscale. A non-uniform magnetization can be due to imperfections in fabrication, e.g. defects, grain size distributions, or non-uniform composition in multicomponent materials, but can also be artificially tailored, e.g. through nanopatterning [2] or local modifications of the magnetization, e.g. via ion beams to modify the local anisotropy [3],[4] .

The capability to characterize systems with topological or engineered magnetization inhomogeneities is thus a topic of paramount importance.

The spectroscopy of X-ray magnetic dichroism (XMCD) effects, which occur in the element-specific near edge X-ray absorption fine structure of ferromagnetic materials are an established and powerful method of locally probing magnetic structures[5]. Specifically, the applications of magneto-optical sum rules [6],[7] allows for the separation and determination of both spin and orbital magnetic moments of each constituent element in multicomponent systems. The utilization of XMCD as magnetic contrast mechanism in soft x-ray microscopies, such as X-ray photoemission electron microscopy (X-PEEM)[8] , Fresnel zone plate based full-field (TXM) and scanning transmission x-ray microscopies (STXM)[9], and lensless imaging techniques, e.g. x-ray holography [10] and coherent diffraction imaging [11] allows to image magnetic domains and spin textures at a spatial resolution, which ultimately will reach into the nanometer regime due to diffraction limit of the wavelength of soft x-rays. Combining the spectroscopic information with the spatial resolution in an x-ray spectromicoscopic approach can provide unique information on the local distribution of the local distribution of magnetization. This was demonstrated by X-PEEM on a study of the spin reorientation in a wedge-shaped fcc Co/Ni/Cu(001) epitaxially grown ultrathin film but with sub-micrometer spatial resolution only [12].

Here, we report a magnetic x-ray spectromicroscopic study of the local distribution of magnetization using magnetic full-field transmission X-ray microscopy (MTXM) with 25nm spatial resolution. We are able to map out locally the orbital/spin (L/S) ratio in the multidomain structure of a multilayered (Co 0.3nm/Pt 0.5nm)x30 thin film exhibiting a strong perpendicular magnetic anisotropy. We observe significant changes of the L/S ratio when approaching a magnetic domain wall.

II. EXPERIMENTAL DETAILS

The (Co 0.3nm/Pt 0.5nm)x30 thin film exhibiting a strong perpendicular magnetic anisotropy was fabricated by sputter deposition onto a 100nm thin Si_3Ni_4 membrane to allow for sufficient – x-ray transmission.

The x-ray microscopy data were obtained using the full-field transmission soft x-ray microscope XM-1 at beamline 6.1.2 of the Advanced Light Source (ALS) in Berkeley, CA. The experimental setup, specifically for magnetic TXM (MTXM) is described elsewhere, see e.g. [9] and references therein. Using circular polarized x-rays emitted either into the upper or lower half of the x-rya cone from a bending magnet magnetic domain images can be recorded utilizing XMCD as magnetic contrast mechanism. The transmitted intensity is a measure of the projection of the magnetization onto the photon propagation direction. Each image covering a field of view of about 8-10 micrometer diameter can be recorded within a few seconds of exposure times. For the spectromicroscopic study, MTXM images using the upper half of the x-ray cone from the bending magnet were recorded as a function of photon energy. The specific energy range for the PtCo multilayer was chosen from 725-805eV to cover both the pre-edge region as well as the L_3 and L_2 resonant absorption edges at 778.1eV and793.2eV resp. in steps of 0.5eV. This matches the spectral resolution of the instrument, which is largely determined by the illuminating optics, i.e. the combination of a condenser zone plate with 55nm outermost zone width and 10mm diameter and a pinhole of 15µm diameter located 250 µm upstream the sample. The transmitted x-rays are projected by a micro-zone plate used as objective lens with 25nm outermost zone width onto a 2048x2048-pixel CCD camera. Each pixel in the CCD camera corresponds to about 5.5nm.

Each image was corrected for nonlinear image drift and non-uniform background illumination using ImageJ [13] image analysis software. To derive local XMCD spectra, the intensity difference of areas with varying sizes but uniform and opposite magnetization were determined as a function of photon energy. From those spectra, the ratio of the local spin (S) and orbital (L) magnetic moments were derived following the standard formalism of magneto-optical sum rules.

$+¿+j^{-¿}$
$-¿$
$+¿-I^{¿}$
$I^{¿}$
$¿$
$j^{+¿}$
$-¿$
$+¿+I^{¿}$
$j^{-¿}$
$-¿$
$+¿-I^{¿}$
$I^{¿}$
$¿$
$¿$
$dE¿$
$I^{¿}-2\int_{¿}¿$
$dE¿$
$\int_{¿}¿$
$3¿$
$dE¿$
$j^{¿}¿$
$2\int_{¿}¿$
$\frac{L}{S}=¿$

with $\overset{+¿}{I^{¿}}$ and $\overset{-¿}{I^{¿}}$ being the intensities in the local areas with opposite magnetization and

$\overset{+¿}{j^{¿}}$ and $\overset{-¿}{j^{¿}}$ denoting the $p_{3/2}$ and $p_{1/2}$ electronic levels.

III. RESULTS AND DISCUSSION

Figure 1 a and b shows representative MTXM images recorded at 777.8eV and792.5eV, where the magnetic contrast is maximum. It shows the expected reversal of magnetic contrast due to the opposite spin orbit coupling of the inner-core $p_{3/2}$ and $p_{1/2}$ electrons involved in the x-ray transition process, resp. With regard to local inhomogeneities of the magnetization, our study focused on the domain wall region, i.e. the transition from an up to down magnetized area, which corresponds to areas with increased and decreased transmitted intensity.

The spatial resolution of the recorded images can be derived from analyzing the domain wall region, assuming that the domain wall width is below the spatial resolution, i.e. a sharp edge in the transition. The left panel in Figure 2 shows the derivative of Fig 1 a, where the domain wall regions show up as white lines. A typical intensity profile taken across the domain wall region (see Fig 1a) and orthogonal to the local domain wall direction is shown in Fig 2 b. The width of the transition derived from the 10% to 90% values of the intensity confirms that a spatial resolution of 25nm can be assumed in our MTXM images.

The spectroscopic capabilities of MTXM are demonstrated in Fig 3. Following the XMCD analysis procedure for the MTXM images, as described above, we determined XMCD spectra from the average intensity in areas of varying sizes ranging from more than 100nm x 100nm down to 25nm x 25nm, which is limited by the spatial resolution. Fig 3a shows, that although the signal to noise values increase with decreasing area sizes, yielding a 10% error for the smallest area recorded, all L/S values are compatible within each other with a L/S value around 0.37. This is in agreement with data from standard XMCD spectroscopy measurements, where a typical beam size in the 0.1mm range determines the area of the measured average XMCD signal. MTXM spectromicroscopy can therefore be used as a local probe of magnetization.

The constant value of L/S across a magnetic domain changes dramatically in the proximity of a magnetic domain. Figure 4 shows the results from mapping the L/S ratio as a function of the distance to the domain wall and along the domain wall directions. As can be seen in the surface contour plot in Fig 4b, where the z-axis denotes the L/S value as a function of the distance from the domain wall and along the domain wall, rhere is a distinct decrease of the L/S value starting at about 80nm away from the domain wall reaching L/S values as low as about 0.2 close to the domain wall, i.e. a reduction to almost 60% of the original value. Interestingly, along the direction of the domain wall, this reduction is constant, indicating that the observed local reduction of the L/S value is inherent to the domain wall spin structure. To our knowledge, this is the first observation of a local change in L/S ratio at a domain wall, which can be seen as a representative for a local spin inhomogeneity.

As the measurements were done in perpendicular geometry, i.e. the photon beam direction was orthogonal to the sample’s surface normal, only magnetic moments with a projection onto the photon beam direction contribute to the XMCD signal. A reduction in the L/S value is therefore indicative of in-plane contributions occurring at the surface of the PtCo film.

In classical domain wall theory, there are two basic types of domain walls, i.e. Bloch and Neel walls. In PMA films such as the one presented here, the commonly assumed domain wall configuration is the Bloch wall, where the magnetization rotates uniformly from up to down direction in the plane of the wall. Our results are indicative, that this could be potentially an oversimplified representation. Neel caps, i.e. in-plane rotating spins have been predicted by micromagnetic modeling in magnetite to eliminate surface poles magnetized elements [14] and have been observed recently with X-PEEM in Fe (110) dots [15].

The existence of in-plane contributions to the domain wall spin structure in PtCo films emphasizes the 3dim character of a domain wall. The experimental confirmation requires the capability for 3d imaging of spin structures. Recent developments show that x-ray microscopies have the potential to expand into this capability [16].

IV. CONCLUSIONS

Combining the spatial resolution of MTXM with the magnetic information contained in XMCD spectroscopy we have studied the local distribution of L/S ratios in a Pt/Co multilayer with perpendicular magnetic anisotropy exhibiting a distinct domain structure. We have demonstrated that L/S ratios can be obtained with high statistical significance down to the spatial resolution fo MTXM limited by the x-ray optics. Whereas the obtained L/S values agree well inside a magnetic domain with XMCD spectroscopy data, in the proximity to a domain wall, the L/S ratio is significantly reduced keeping this a low value along the domain wall. The simplified representation of a Bloch wall, where the magnetization uniformly rotates at 180 degree needs to include in-plane contributions (Neel caps) at the surfaces.

The capability to map with nanoscale resolution and quantify local magnetic inhomogeneities will provide important information with regard to future developments of nanoscale magnetic devices and to obtain a fundamental understanding of nanoscale magnetic behavior.

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FIGURE CAPTIONS

Figure. 1: MTXM image of the domain structure recorded at the Co L_3 (a) and L_2 (b) x-ray absorption edge in a multilayered (Co 0.3nm/Pt 0.5nm)x30 thin film exhibiting a strong perpendicular magnetic anisotropy showing a reversal of magnetic contrast due to the reversal of spin-orbit coupling in the contributing 2p electronic levels.

Figure. 2: a) Image of derivative of Fig. 1a) emphasizing areas of magnetization inhomogeneities. The magnetic domain walls show up as pronounced white regions. b) Intensity profile across domain wall in Fig 1 and width of the 10% to 90% edge response from where the spatial resolution in the MTXM images can be derived.

Figure. 3: a) L/S ratios calculated from areas with varying size down to the spatial resolution of the MTXM instrument. These measurements were taken far away from the domain wall, where L/S ratios are expected to be constant. The size of 1px corresponds to about 5.5nm. (b) Typical XMCD difference spectrum of the multilayered (Co 0.3nm/Pt 0.5nm)x30 thin film derived from intensity differences in equally sized areas.

Figure. 4: a) Grid of the areas and MTXM pixel contents used to generate the L/S ratio mapping. (b): 3D contour map, where the z-values corresponds to the XMCD values and x and y are locations on the sample used in the quantitative 2D mapping of L/S ratios near the domain wall.

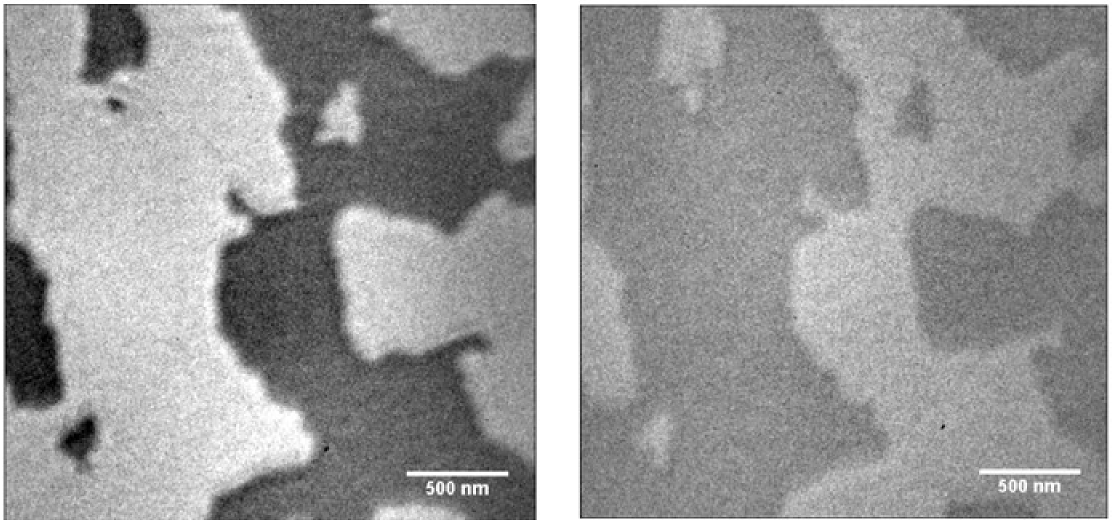

FIGURE 1

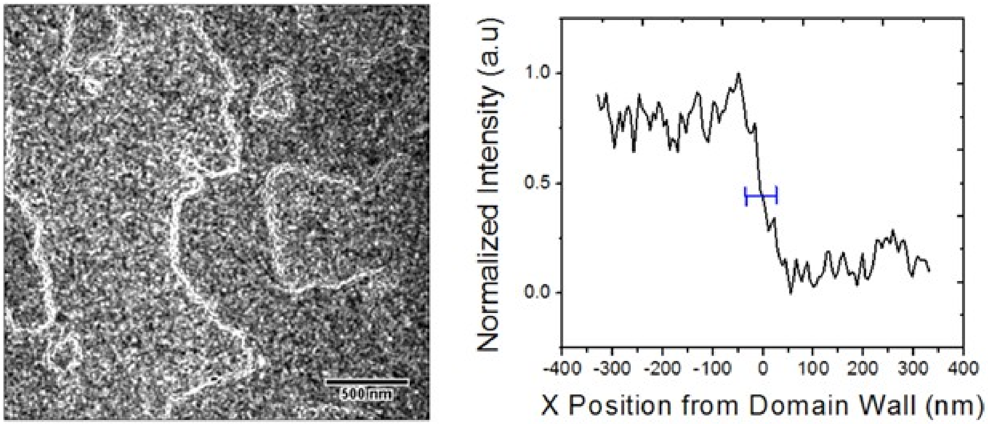
500 nm
Normalized Intensity (a.u)
1.0
0.5
0.0
-400
-300
-200
-100
0
100
200
300
400
X Position from Domain Wall (nm)

FIGURE 2

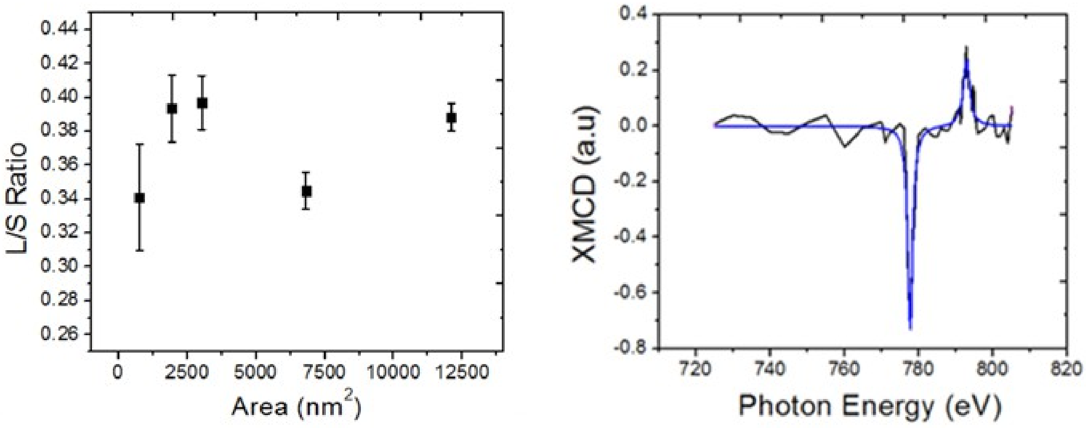
L/S Ratio
0.44
0.42
0.40
0.38
0.36
0.34
0.32
0.30
0.28
0.26
0
2500
5000
7500
10000
12500
Area (nm^2)
XMCD (a.u)
0.4
0.2
0.0
-0.2
-0.4
-0.6
-0.8
720
740
760
780
800
820
Photon Energy (eV)

FIGURE 3

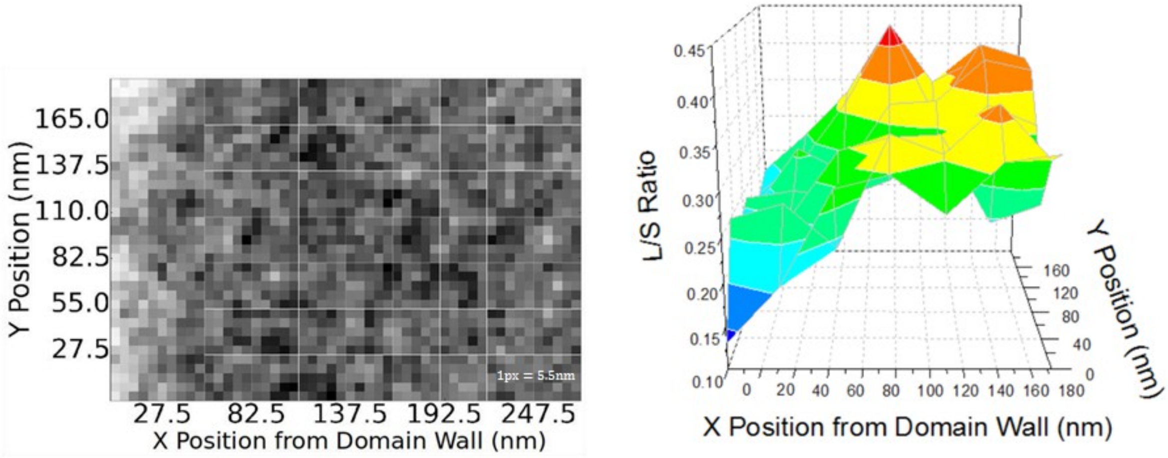
Y Position (nm)
165.0
137.5
110.0
82.5
55.0
27.5
27.5
82.5
137.5
192.5
247.5
X Position from Domain Wall (nm)
1px = 5.5nm
L/S Ratio
0.45
0.40
0.35
0.30
0.25
0.20
0.15
0.10
0
20
40
60
80
100
120
140
160
180
X Position from Domain Wall (nm)
160
120
80
40
0
Y Position (nm)

FIGURE 4