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## **Report:**

An enhancement of the spin-orbit effects arising on an interface between a ferromagnet (*FM*) and a heavy metal (*HM*) is possible through the strong breaking of the structural inversion symmetry in the layered films. We have found that the introduction of an ultrathin W interlayer between Co and Ru in Ru/Co/Ru films enables to preserve perpendicular magnetic anisotropy (PMA) and simultaneously induce a large interfacial Dzyaloshinskii-Moriya interaction (iDMI). We find that the Ru/Co/W/Ru films have PMA up to 0.35 nm of the nominal thickness of W ( $t_W$ ). The study of the spin-wave propagation in the Damon-Eshbach geometry by Brillouin light scattering (BLS) spectroscopy reveals the drastic increase of the iDMI value with the rising  $t_W$ . The maximum iDMI of -3.1 erg/cm<sup>2</sup> is observed for  $t_W=0.24$  nm, which is 10 times larger than the latter for the quasi-symmetrical Ru/Co/Ru films. The ability to simultaneous control the strength of PMA and iDMI in symmetrical *HM/FM/HM* trilayer systems through the interface engineered inversion asymmetry at the nanoscale excites new fundamental and practical interest to the chiral ferromagnets, which are a potential host for stable magnetic skyrmions.

Our polycrystalline Ru/Co/W/Ru films were prepared by magnetron sputtering on the SiO<sub>2</sub> substrates at room temperature. The base pressure in the chamber was  $10^{-8}$  Torr. The working pressure of  $Ar^+$  was  $10^{-4}$  Torr. In order to precise control the thickness of layers, we used low sputtering rates: V(Ru)=0.011 nm/s, V(Co)=0.018 nm/s, V(W)=0.02 nm/s. The Co thickness ( $t_{Co}$ ) was varied from 0.7 to 1.5 nm. The thickness of the buffer and capping Ru layers ( $t_{Ru}$ ) was 10 and 2 nm, correspondingly. The W thickness ( $t_W$ ) was taken in the range from 0 to 0.4 nm. The structural and magnetic properties of the quasi-symmetrical Ru/Co/Ru were systematically studied.

Magnetic properties of films were investigated with vibrating sample magnetometer (7410 VSM, LakeShore). Kerr microscopy (Evico Magnetics) was used to study domain structure and its field driven dynamics. Kerr microscope images were captured using an immersion oil in order to increase optical resolution for clear observation of skyrmions with the size as small as 700 nm. Brillouin light scattering (BLS) spectroscopy measurements were conducted to probe the spin-wave frequency non-reciprocity in the Damon-Eshbach geometry in order to define the iDMI sign and value. The crystal structure and interface quality was studied by X-ray diffraction (XRD) and X-ray reflectivity (XRR) measurement techniques (SmartLab, RIGAKU) at CuK $\alpha$  radiation wavelength (1.54 Å). The fitting of XRR spectra were performed with GenX software. Micromagnetic simulations of the domain structure were executed using MuMax<sup>3</sup> software package.

Polarization dependent X-ray spectroscopy measurements at the Co K- (7709 eV), W L<sub>3</sub>- (10207 eV) and W L<sub>2</sub>- (11544 eV) edges on the Ru(10nm)/Co(1nm)/W(x)/Ru(2nm) structures were performed at the

ESRF ID12 beamline. Four different samples have been measured with x=0, 0.21, 0.25 and 0.29, where x is the thickness of the layer in nm. The APPLE-II undulator and a Si(111) double crystal monochromator were used to collect the spectra. Circular polarization rate of monochromatic x-rays was in excess of 95% at the W  $L_{3,2}$ -edges. Magnetic field of 0.27 T generated by an electromagnet was applied in the film plane and nearly parallel to the X-ray beam direction. This filed was sufficient to reach magnetic saturation of the samples at room temperature. In order to detect x-ray absorption spectra from our samples, we had to use an energy resolved detector - Si drift diode with an active area of 10 mm<sup>2</sup>. To avoid saturation of the detector with fluorescence of Ru and of Si as well as Compton scattering from SiO<sub>2</sub> substrate a 50 µm Al filter was inserted between sample and the diode. The XMCD spectra were recordered by flipping the direction of applied magnetic field two times at every energy point of the x-ray absorption spectra. To make sure that XMCD signal is free of experimental artefacts, we checked that the sign of XMCD was reverted when it was recorded with opposite x-ray helicity. It should be underlined that the attenuation length for tungsten  $L_{3,2}$ edges is about 4 orders of magnitude larger than the total thickness of W in the samples. That's why very long acquisition time was needed to observe XMCD signals well above the noise level.



**FIG 1.** The comparison of normalized Co *K*- XANES spectra for the Ru(10nm)/Co(1nm)/Ru(2nm) reference sample with W containing ones: Co(1nm)/W(0.21nm), Co(1nm)/W(0.25nm), Co(1nm)/W(0.29nm). Inset: K-edge XANES spectra of Co with *hcp* and *fcc* structures taken from [Physics of the Solid State 53 (1) 1–5 (2011); Appl. Sci. 2 396-442 (2012)].

In Fig. 1 we show normalized x-ray absorption spectra at the *K*-edges of Co in Ru/Co/W/Ru trilayers in comparison with a sample without W (9.27(1) sample). The sensitivity of the XANES technique to a valence state of absorbing atoms and to their local atomic structure allows one to use this technique for "fingerprinting" the crystallographic structure of a material under study [Physics of the Solid State 53 (1) 1–5 (2011); Appl. Sci. 2 396-442 (2012)]. Comparison of our normalized XANES spectra recorded at the *K*-edge of cobalt with published results [Physics of the Solid State 53 (1) 1–5 (2011); Appl. Sci. 2 396-442 (2012)] clearly shows that in all studied samples Co has *hcp* structure (see inset of fig. 1) and practically not affected by the deposition of tungsten.

In Fig. 2 we reproduce normalized XANES and XMCD spectra recorded at the  $L_{3,2}$ -edges of W in Co/W/Ru trilayers in comparison with those measured on bulk Co<sub>3</sub>W alloy [PRB 86 064428 (2012)]. For the XANES spectra the ratio of the tungsten  $L_3/L_2$  was taken equal to 2.19/1 according to [At. Data Nucl. Data Tables 54 181-342 (1993)]. The normalized intensities of XMCD signals is about 1% and evidence that 5*d* states of W carry a magnetic moment for all measured samples. This induced magnetic moment is aniparallel to applied field and therefore to the 3*d* magnetic moment of Co.

In order to minimize an influence of the noise in the XMCD spectra on final results, we decided to exploit a differential formulation of the X-ray magneto-optical sum-rules [The Physics of Metals and Metallography 116(13) 1285-1336 (2015); J. Phys.: Condens. Matter 8 L747-L752 (1996); PRB 60 7156 (1999)]. The XANES spectra were first shifted in energy one with respect to the other in such a way that the EXAFS wiggles at both  $L_2$  and  $L_3$  edges coincides when both spectra are normalized to unity. The XMCD spectra are then shifted in the same way before applying the differential sum rules. We have applied the same procedure to the spectra recorded in [PRB 86 064428 (2012)] on bulk Co<sub>3</sub>W alloy. The results of application of the differential magneto-optical sum-rules, i.e.  $\langle I_Z \rangle$  and  $\langle 2s_Z \rangle$  5d DOS of tungsten for all three measured

samples in comparison with Co<sub>3</sub>W alloy are presented in Fig. 3(a,b,c). It should be noted that the form of the  $\langle 2s_Z \rangle$  spin contribution function for all measured samples is consistent for the same function measured on bulk Co<sub>3</sub>W alloy. In turn the  $\langle l_Z \rangle$  orbital contribution is close to zero. It becomes obvious that the XMCD signals, as in the case of Co<sub>3</sub>W, originate predominantly from the spin polarization of the 5*d*-DOS, because  $\langle 2s_Z \rangle$  largely exceeds the contribution of  $\langle l_Z \rangle$ . Due to a rather high level of noise it is very difficult to discuss the orbital magnetic moment and its sign. We can only give its estimate, it is about 10 times weaker than the spin magnetic moment. Integrating the  $\langle l_Z \rangle$  and  $\langle 2s_Z \rangle$  spectra and taking x-ray absoprtion cross-section per hole from [PRB 86 064428 (2012)], one obtains  $m_L < \pm 0.002$  and  $m_S \sim -0.023 \pm 0.004$  for all three studied samples. Similar values were also obtained in [PRB 86 064428 (2012)]:  $m_L < \pm 0.004(1) \mu_B$  and  $m_S \sim -0.023(2) \mu_B$  for W on bulk Co<sub>3</sub>W alloy.



**FIG 2.** XAS and XMCD spectra at the W  $L_{3,2}$  for three samples (Co(1nm)/W(0.21nm), Co(1nm)/W(0.29nm)) measured at H = 0.27 T at RT in comparison with Co<sub>3</sub>W alloy [PRB 86 064428 (2012)] measured at H = 5 T and T = 10 K.

What has attracted our attention is some visible difference in the absorption white-line intensities at W  $L_{3,2}$  edges for three samples (Fig. 3) that can results from variation of the 5*d* DOS above the Fermi level. Specifically,  $L_3$  absorption edge reflects both  $5d_{3/2}$  and  $5d_{5/2}$  DOS whereas only the  $5d_{3/2}$  band is probed at the  $L_2$ -edge as given by the dipolar selection rules for corresponding optical transitions:  $2p_{3/2} \rightarrow 5d_{3/2}$  and  $5d_{5/2}$  at the  $L_3$ -edge and  $2p_{1/2} \rightarrow 5d_{3/2}$  at the  $L_2$ -edge. Surprisingly, Co(1nm)/W(0.21nm) and Co(1nm)/W(0.29nm) show nearly the same structures whereas an increase of the number of W  $5d_{3/2}$  empty states for Co(1nm)/W(0.25nm) sample is clearly observed. This is a clear difference, eventhough quite minor, in population of spin-orbit split 5*d* states between these three samples.



**FIG 3.** X-ray absorption spectra for all samples (Co(1nm)/W(0.21nm) (a), Co(1nm)/W(0.25nm) (b), Co(1nm)/W(0.29nm) (c)) at the tungsten  $L_{3,2}$ -edges plotted as a function of the photoelectron energy  $\Delta E = E - E_F$ , as well as magnetically polarized  $\langle l_Z \rangle$  and  $\langle 2s_Z \rangle$  5*d* DOS of tungsten as a result of analysis based on the differential sum rules [J. Phys.: Condens. Matter 8 L747-L752 (1996); PRB 60 7156 (1999)], in comparison with magnetically polarized  $\langle l_Z \rangle$  and  $\langle 2s_Z \rangle$  5*d* DOS of tungsten for bulk Co<sub>3</sub>W alloy [spectra are taken from PRB 86 064428 (2012)].

In summary, we show that there is an induced spin magnetic moments of about -0.02  $\mu_B$  per atom for W for all studied samples with a much weaker orbital contribution. The magnetic moment of W is antiferromagnetically coupled with the Cobalt moment in agreement with other studies on 3*d*/W systems [PRL 87 207202 (2001); PRB 86 064428 (2012)]. Small changes in the population 5*d* spin-orbit split bands for different thicknesses of tungsten were observed.