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Observation of magnetic moments at the interface region in magnetic tunnel junctions using depth-resolved x-ray magnetic circular dichroism

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The depth-resolved x-ray magnetic circular dichroism (XMCD) technique was applied to observe the interfacial and inner magnetic moments in CoFe/MgO and Co_2MnSi(CMS)/MgO. The magnetic moments of the real interface region (two monolayers from the interface) and the inner layers were separately analyzed using the XMCD sum rules. The observed interfacial moments of Co at the CMS/MgO interface show a remarkable reduction from that in the inner layers compared with the CoFe/MgO structure, suggesting small exchange stiffness in the CMS at the MgO interface. The weak exchange stiffness of Co is a possible reason for the large temperature dependence of tunnel magnetoresistance in CMS-based magnetic tunnel junctions.

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Achieving efficient spin-injection into nonmagnetic metals and semiconductors is important for new spintronic applications such as spin transistors. The magnetic properties in the real interface region [a few monolayers (MLs) from the interface] of a heterostructure with a ferromagnet (FM) and other materials largely determine the efficiency of spin injection from the FM. Therefore, before we can find ways to improve the spin-injection efficiency, we need to be able to directly observe the magnetic properties at the real interface.

The tunneling resistance in a magnetic tunnel junction (MTJ), a thin insulating layer (a tunnel barrier) sandwiched by two FM metallic layers (electrodes), varies depending on the relative magnetic alignment of the FM electrodes, which is called the tunnel magnetoresistance (TMR) effect.[1,2] It is well understood that the TMR effect is sensitive to the real interfacial region at the FM electrode/tunneling barrier. Extrinsic factors such as magnetic impurities or structural dislocations formed at the barrier interface are known to largely degrade the TMR properties.[3–5] Intrinsically, for example, large enhancement and reduction of the TMR ratio by inserting 4-ML bcc Co and 1-ML Cr at the barrier interface have been observed previously, suggesting the strong interface sensitivity of TMR.[6,7] The thermal fluctuation of interfacial magnetic moments is also considered to critically affect the temperature (T) dependence of TMR properties. However, up to now, no experimental study has investigated the relationship between the TMR properties and magnetic properties in the real interface region at the tunneling barrier.

There are several experimental techniques for characterizing surface/interface magnetic properties. Magnetization-induced second-harmonic generation (MSHG) is a powerful method for investigating surface magnetism because of its intrinsic surface/interface sensitivity.[8,9] However, it is difficult to compare the magnetic properties of the inner layers' region with those of the surface region, since no magnetic information about the inner layers can be obtained in MSHG. Alternatively, x-ray magnetic circular dichroism (XMCD) with the total electron yield (TEY) method is often used to measure surface/interface magnetic moments since XMCD signal with TEY detects the range of the escape depth of secondary and Auger electrons; typically 2–5 nm from the surface. Additionally, the element selectivity and the quantitativity of the magnetic moment of XMCD are also very attractive for studying magnetic properties in detail. However, when we want to access magnetic moments in a real surface/interface region, the TEY method is not appropriate because of insufficient sensitivity to the surface/interface. Although several groups have attempted to characterize magnetic properties a few MLs from the surface or interface by measuring a few-MLs-thick FM layer grown on a buffer layer by TEY,[10,11] the magnetic properties of such ultrathin films are not always identical to those at the surface/interface in the thick (3–30 nm) FM layers generally used in MTJs. Therefore the correlation between the magnetic moments of a FM electrode a few MLs from the barrier interface and the TMR properties is still unclear.

The depth-resolved XMCD method we have developed, in which electrons are collected using the detection-angle dependence of their effective escape depth, is a powerful tool for separately investigating magnetic properties in the real surface region and in the inner layers of the FM film.[12,13] Using this method for an MTJ structure without an upper FM layer (called a half MTJ structure here), we can evaluate magnetic moments of the lower FM layer in the region a few MLs from the interface of the tunneling barrier. In this study, we applied the depth-resolved XMCD method to half MTJ films with CoFe/MgO and Co_2MnSi(CMS)/MgO structures to study the relationship between the TMR properties and interfacial magnetic moments. In our recent studies on MTJs with CMS electrodes, we have found a large TMR ratio at low T, which reflects the half metallic nature of CMS.[14,15] However, in contrast to the small T dependence in MTJs with CoFe electrodes, the TMR ratio showed an unexpected drastic reduction with T even though Curie temperature of CMS and CoFe was almost the same.

Half MTJs with a (001)-oriented epitaxial structure of MgO(100)-substrate/Cr-buffer (40)/CoFe(5) or CMS(30)/MgO(1)/Al-cap(1.5) (unit: nanometer) were fabricated using an ultrahigh-vacuum magnetron sputtering system without breaking vacuum. The CoFe and CMS layers were annealed at 300 and 450 °C, respectively. Then, the MgO barrier was

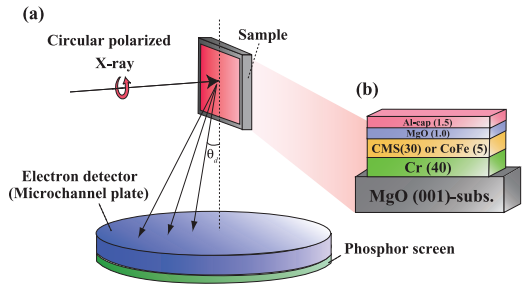

FIG. 1. (Color online) (a) Experimental setup for depth-resolved XMCD. (b) Stacking structures of the half MTJs using CoFe and CMS electrodes.

formed by electron-beam evaporation at ambient temperature. High degrees of B2 ordering and $L2_1$ ordering were confirmed in the CMS film from the superlattice diffraction peak in XRD measurement. The saturation magnetization of 1010 emu/cc was observed in the CMS film, which is almost the same as the bulk value. X-ray-absorption (XA) and XMCD spectra were measured in an ultrahigh-vacuum chamber at BL 7A and 16A at the Photon Factory of the High-energy Accelerator Research Organization, Japan. The sample was magnetized using pulsed current through a coil (about 500 G) oriented along the x-ray propagation direction. The coil was retracted during the measurement. Co L-edge XA spectra were measured with the field parallel and antiparallel to the fixed photon helicity, and XMCD spectra were obtained from the difference between the two spectra. Since both films exhibit in-plane magnetization, the magnetic moments were determined using XMCD spectra at grazing (30° from surface) x-ray incidence. Depth-resolved XMCD measurements were performed using an imaging-type microchannel plate detector with a phosphor screen and a charge-coupled device camera, operated in a partial electron yield mode, with an applied retarding voltage of 500 V for preferential collection of Co *LMM* Auger electrons. Spectra from different probing depths were collected simultaneously against various electron detection angles θ_d, as depicted in Fig. 1. The XA and XMCD spectra were measured at room temperature (RT) and liquid-nitrogen temperature (actual sample temperature was about 80 K). In this study, the spin and orbital magnetic moments in the interface region and in the inner layers were separately estimated using the XMCD sum rules,[16,17] where we analyzed the spectra by assuming that the magnetic moment in 2 MLs (meaning 0.28 nm for both CoFe and CMS) from the barrier interface is different from that in the inner layers. Note that, in the chemically ordered CMS, a Co layer and a Mn-Si layer are alternately stacked to the [001] direction. Thus, for perfectly chemically ordered CMS and an atomically flat interface, interfacial 2 MLs include one Co layer as a terminated or a second terminated layer with the MgO barrier.

Figure 2 shows the T dependence of the TMR ratio in CoFe/MgO/CoFe and CMS/MgO/CoFe MTJs with the same bottom structures as the half MTJs and deposited under the same conditions. The CoFe/MgO/CoFe MTJ showed small T dependence; the observed TMR ratios were 170% at 300 K and 240–260% at around 2–30 K. In contrast, the CMS/MgO/CoFe MTJ has extremely large T dependence; the giant TMR ratio of 753% at 2 K drastically decreased with T toward 208% at

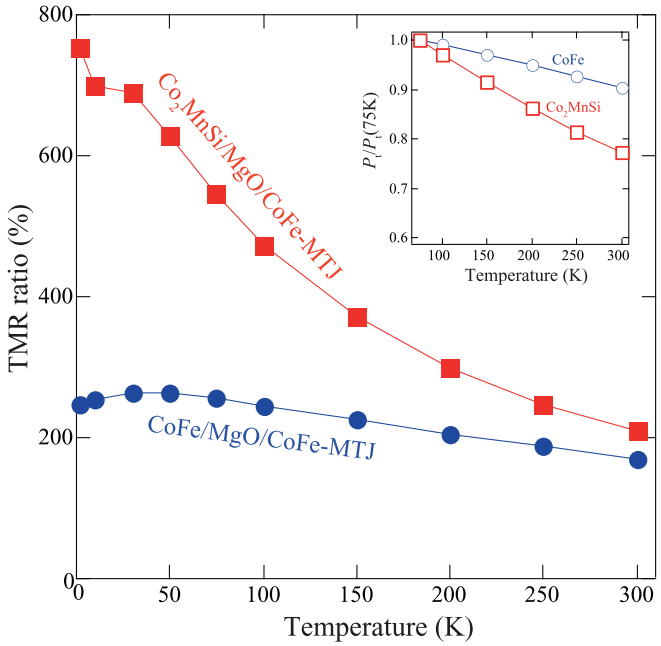

FIG. 2. (Color online) Temperature dependence of TMR ratios in CoFe/MgO/CoFe and CMS/MgO/CoFe MTJs. The inset shows the temperature dependence of effective spin polarization $P_{\rm eff}$ normalized by that at 75 K, estimated from Julliere's formula.

300 K. The inset shows the T dependence of the tunneling spin polarization P_t (i.e., spin polarization including the enhancement by the spin-filtering effect of the MgO barrier) of CoFe and CMS. The data are normalized by P_t at 75 K. The estimated P_t(300 K)/ P_t (75 K) for CoFe and CMS were 0.90 and 0.77, respectively, indicating a rapid reduction of tunneling spin polarization in the CMS electrode compared to that in the CoFe electrode. Note that the spin bands crossing the Fermi level are similar in CoFe and CMS, i.e., only the spin-up channel has a Δ_1 electron that dominantly tunnels through the MgO barrier.[18] Therefore the observed large difference in the temperature dependence in TMR ratio is caused by the temperature dependence of interfacial spin polarization at the electrode/MgO not by tunneling probability.

Figures 3(a)–3(d) show the XA and XMCD spectra around Co $L_{2,3}$-absorption edges in the CoFe/MgO and CMS/MgO half MTJs measured at RT. The spin and orbital magnetic moments ($m_s^{\rm eff}$ and m_l) were estimated by XMCD sum rules using the 3d hole number of inner-layer Co $n_h = 2.50$ for CoFe and 2.24 for CMS.[10] The effective spin moment $m_s^{\rm eff}$ is defined as $m_s + 7m_T$, where m_s and m_T denote the spin magnetic moment and magnetic dipole, respectively, the details of which are described in the literature.[17] In any case, the contribution of m_l is negligibly small against $m_s^{\rm eff}$, which is consistent with previous reports.[10,19] In the CoFe/MgO, $m_s^{\rm eff}$ is smaller in the interface region but preserved to be large (1.90μ_B, almost 80% of that in the inner layers). In contrast, in the CMS/MgO, $m_s^{\rm eff}$ at the interface is just 0.60μ_B, which is 57% of that observed in the inner layers of the CMS. The observed $m_s^{\rm eff}$ in the inner layers of CMS agrees with the previous study using TEY.[10] The XA spectra of Co and Mn (not shown here) at the interface exhibit no feature suggestive of oxidization at the interface, indicating a high interfacial quality. Therefore it is considered

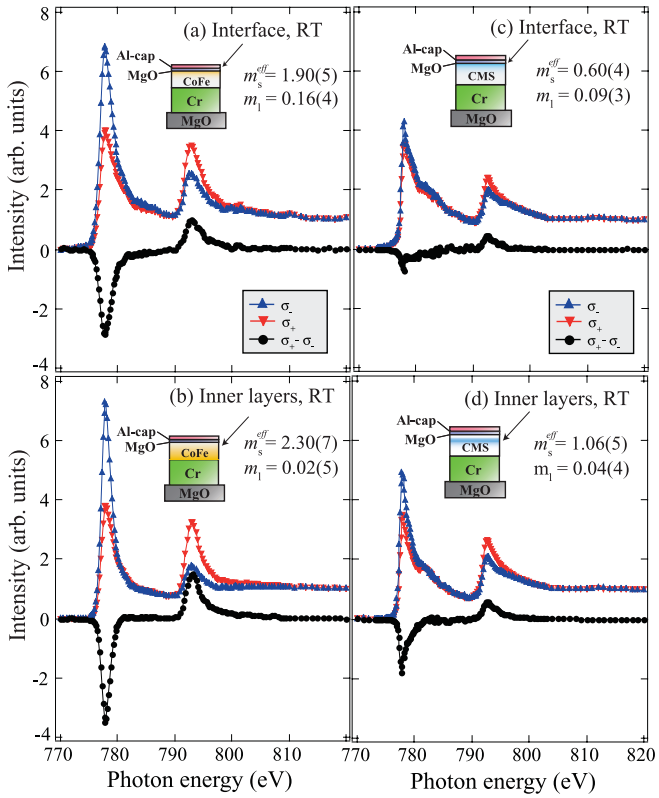

FIG. 3. (Color online) XA (blue and red curves) and XMCD (black curve) spectra at Co $L_{2,3}$ edges in the CoFe/MgO and CMS/MgO half MTJs measured at RT. Spectra of the interface region in the CoFe/MgO (a) and the CMS/MgO (c) and in the inner layers for the CoFe (b) and the CMS (d). The spin and orbital magnetic moments ($m_s^{\rm eff}$ and m_l) were estimated by XMCD sum rules. The values in parentheses are the estimation errors in the XMCD sum-rule analysis.

that there is an intrinsic reason for the large reduction of $m_s^{\rm eff}$ at the CMS/MgO interface. A recent first-principles calculation of the magnetic moment in the (001)-CMS/MgO structure predicted a small magnetic moment of $0.40\mu_{\rm B}$ in a Co layer terminated with a MgO barrier.[20] In contrast, for Mn-Si layer termination, a large magnetic moment of $1.28\mu_{\rm B}$ is calculated for the second terminated Co layer. Therefore, although a previous study suggested a Mn-Si termination with MgO in the CMS/MgO structure,[10] our present results imply the existence of partial Co termination due to interface roughness or chemical disordering at the interface region. The possibility of chemical disordering at the terminated layer is also suggested from the scanning transmission electron microscopy observation of the CMS/MgO MTJ.[21]

Our present experiment using depth-resolved XMCD found a large reduction of magnetic moments of Co at the CMS/MgO interface from that in the inner layers. It is well known that the stiffness of exchange coupling A is proportional to JM^2, where J and M represent the exchange energy and magnetic moment, respectively. Thus small M at the interface inevitably results in large thermal fluctuation of the magnetic moment. From a simple estimation of A in the Co interfacial magnetic moment using the observed $m_s^{\rm eff}$, A becomes just one-third of that in the inner layers in the CMS/MgO MTJ even when the reduction of J at the interface is not taken into account. In the first-principles calculation of tunneling conductance with noncollinear spin configuration at the interface taken into consideration, the spin-flip conductance increases with increasing tilting angle of interfacial magnetic moment in antiparallel magnetic configuration due to the creation of Δ_1 conductive channel at the Γ point in the $k_{//}$ plane.[22] Thus a large reduction of TMR until a few orders of magnitude was predicted to occur even by a small tilting of the interfacial magnetic moments. Therefore it is concluded that the large difference in the interfacial magnetic moment in the CoFe/MgO and CMS/MgO structures can qualitatively explain the remarkable difference in the T dependence of the TMR ratio between them. The weak exchange stiffness indicated from the small $m_s^{\rm eff}$ of Co at the CMS/MgO interface is the most likely cause of the extremely large temperature dependence of the TMR ratio in the MTJs with CMS electrodes.[14,15,23,24]

On the other hand, XMCD spectra at 80 K (not shown here) exhibit no remarkable variation of $m_s^{\rm eff}$ compared with those at RT in either sample even in the interface region ($1.87\mu_{\rm B}$ and $0.58\mu_{\rm B}$ for the interface with CoFe and CMS, respectively). This result agrees with a previous XMCD study on temperature dependence using TEY,[25] but seems to contradict the above-discussed T dependence of TMR. However, the tiny T dependence of $m_s^{\rm eff}$ in contrast to the large dependence of a TMR is reasonably explained as follows: In this study, we measured a remanent magnetization state in XMCD spectra; thus the magnetic moment is aligned to an in-plane easy magnetization direction. At finite temperature, however, a magnetic moment thermally fluctuates from the exact easy-axis direction with tilting angle $\theta(T)$. The $\theta(T)$ can be estimated from the exchange stiffness constant A by calculating the Boltzmann average of $\theta(T)$ against thermal fluctuation energy $k_{\rm B}T$. Miura *et al.* reported that exchange stiffness A of Co in the Co-terminated layer at the CMS/MgO interface is 145 meV per unit-cell area, which is almost one-third of that calculated in bulk (414 meV), as we deduced from our depth-resolved XMCD measurements.[22] The $\theta(T)$ of the interfacial Co magnetic moments estimated from this exchange stiffness A are about 12° at 80 K and 22° at 300 K. The net reductions of the interfacial Co magnetic moment due to thermal fluctuation from 0 K, which can be calculated in $m_{\rm s}[1\text{-}\cos\theta(T)]$, are 0.02 $m_{\rm s}$ at 80 K and 0.07 $m_{\rm s}$ at 300 K, indicating a tiny reduction of the magnetic moment from 80 to 300 K even when we consider a drastic decrease of A at the MgO interface. Therefore it is understandable that there is no remarkable temperature dependence of $m_s^{\rm eff}$ from 80 to 300 K in our XMCD measurements not only in CoFe/MgO but also in CMS/MgO half MTJs. In stark contrast, the TMR ratio is predicted to be reduced strongly even by a tiny $\theta(T)$ in the CMS/MgO MTJ because of the creation of spin-flipping conductance at the interfacial region.[22]

In conclusion, in this study, we performed depth-resolved XMCD measurements on half MTJs with CoFe/MgO and CMS/MgO structures to investigate the relationship between the T dependence of the TMR ratio and interfacial magnetic moment. The observed interfacial spin moments of Co within 2 MLs from the interface with the MgO barrier are $1.90\mu_{\rm B}$ for CoFe and $0.60\mu_{\rm B}$ for CMS, suggesting a large difference

of interfacial exchange stiffness between CoFe and CMS, because the exchange stiffness is proportional to the square of the magnetic moment. The large difference in the T dependence of TMR is attributed to the difference in exchange stiffness between the CoFe/MgO and CMS/MgO interfaces. It should be noted that the reduction of the magnetic moment of Co at the CMS/MgO interface observed in this study is opposite to the result observed in ultrathin CMS films deposited on an Fe buffer layer, in which the Co spin moment is enhanced with decreasing CMS thickness.[10] This indicates that the magnetic properties in an ultrathin FM film are different from those at the surface/interface of a thick FM film. Therefore this result also demonstrates the usefulness of the depth-resolved XMCD technique as a tool to investigate the magnetism in the real interface region of a heterostructure, which critically affects spin-dependent transport properties in various kinds of spintronic devices.

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[1] T. Miyazaki and N. Tezuka, J. Magn. Magn. Mater. **139**, L231 (1995).

[2] J. S. Moodera, L. R. Kinder, T. M. Wong, and R. Meservey, Phys. Rev. Lett. **74**, 3273 (1995).

[3] S. Yuasa, T. Nagahama, A. Fukushima, Y. Suzuki, and K. Ando, Nat. Mater. **3**, 868 (2004).

[4] S. Zhang, P. M. Levy, A. C. Marley, and S. S. P. Parkin, Phys. Rev. Lett. **79**, 3744 (1997).

[5] N. Tezuka and T. Miyazaki, Jpn. J. Appl. Phys. **37**, L218 (1998).

[6] S. Yuasa, A. Fukushima, H. Kubota, Y. Suzuki, and K. Ando, Appl. Phys. Lett. **89**, 042505 (2006).

[7] T. Nagahama, S. Yuasa, E. Tamura, and Y. Suzuki, Phys. Rev. Lett. **95**, 086602 (2005).

[8] M. Straub, R. Vollmer, and J. Kirschner, Phys. Rev. Lett. **77**, 743 (1996).

[9] J. Reif, J. C. Zink, C. M. Schneider, and J. Kirschner, Phys. Rev. Lett. **67**, 2878 (1991).

[10] T. Saito, T. Katayama, T. Ishikawa, M. Yamamoto, D. Asakura, T. Koide, Y. Miura, and M. Shirai, Phys. Rev. B **81**, 144417 (2010).

[11] K. Miyokawa, S. Saito, T. Katayama, T. Saito, T. Kamino, K. Hanashima, Y. Suzuki, K. Mamiya, T. Koide, and S. Yuasa, Jpn. J. Appl. Phys. **44**, L9 (2005).

[12] K. Amemiya, S. Kitagawa, D. Matsumura, H. Abe, T. Ohta, and T. Yokoyama, Appl. Phys. Lett. **84**, 936 (2004).

[13] K. Amemiya, E. Sakai, D. Matsumura, H. Abe, T. Ohta, and T. Yokoyama, Phys. Rev. B **71**, 214420 (2005).

[14] Y. Sakuraba, M. Hattori, M. Oogane, Y. Ando, H. Kato, A. Sakuma, T. Miyazaki, and H. Kubota, Appl. Phys. Lett. **88**, 192508 (2006).

[15] S. Tsunegi, Y. Sakuraba, M. Oogane, K. Takanashi, and Y. Ando, Appl. Phys. Lett. **93**, 112506 (2008).

[16] C. T. Chen, Y. U. Idzerda, H.-J. Lin, N. V. Smith, G. Meigs, E. Chaban, G. H. Ho, E. Pellegrin, and F. Sette, Phys. Rev. Lett. **75**, 152 (1995).

[17] P. Carra, B. T. Thole, M. Altarelli, and X. Wang, Phys. Rev. Lett. **70**, 694 (1993).

[18] Y. Miura, H. Uchida, Y. Oba, K. Abe, and M. Shirai, Phys. Rev. B **78**, 064416 (2008).

[19] I. Galanakis, Phys. Rev. B **71**, 012413 (2005).

[20] A. Sakuma, Y. Toga, and H. Tsuchiura, J. Appl. Phys. **105**, 07C910 (2009).

[21] T. Miyajima, M. Oogane, Y. Kotaka, T. Yamazaki, M. Tsukada, Y. Kataoka, H. Naganuma, and Y. Ando, Appl. Phys. Express **2**, 093001 (2009).

[22] Y. Miura, K. Abe, and M. Shirai, Phys. Rev. B **83**, 214411 (2011).

[23] T. Ishikawa, H. Liu, T. Taira, K. Matsuda, T. Uemura, and M. Yamamoto, Appl. Phys. Lett. **95**, 232512 (2009).

[24] T. Taira, H.-x. Liu, S. Hirata, K.-i. Matsuda, T. Uemura, and M. Yamamoto: *55th Annual Conference on Magnetism & Magnetic Materials, Abstracts* (CD-ROM), BH-10, Atlanta, Georgia, USA, November 14-18, 2010, pp. 118–119.

[25] N. D. Telling, P. S. Keatley, L. R. Shelford, E. Arenholz, G. van der Laan, R. J. Hicken, Y. Sakuraba, S. Tsunegi, M. Oogane, Y. Ando, K. Takanashi, and T. Miyazaki, Appl. Phys. Lett. **92**, 192503 (2008).