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X-ray magnetic circular dichroism spectroscopy analysis of graphene/YIG heterostrcuture with Dirac cone spin polarization

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1 Introduction

Graphene and other two-dimensional materials are receiving increasing attention as potential materials for nanospintronics devices due to their outstanding spin transport properties and low-dimensionality useful for spin manipulation. The use of the proximity effect in graphene/magnetic dielectrics heterostructures has been proposed as an alternative to make up for the weakness of the spin-orbit interaction and gap-less nature of graphene in developing graphene spin transistors [1,2]. Recently, it has been revealed that a large degree of spin polarization is generated in the Dirac cone electrons of single layer graphene (SLG) proximity contacted with yttrium iron garnet (YIG, $Y_3Fe_5O_{12}$) [3]. In this study, we investigated the electronic and magnetic states of the interface region in the SLG/YIG heterostructure with the proximity-induced Dirac cone spin polarization by using X-ray absorption (XAS) and magnetic dichroism (XMCD) spectroscopy.

2 Experiment and Results

The SLG/YIG heterostructure was synthesized by transferring a large SLG sheet, which was grown on a Cu foil by chemical vapor deposition, on a YIG thin film epitaxially deposited on a GGG(111) substrate. In accordance with the manifestation of the magnetic proximity effect [3], the SLG/YIG sample was annealed at 1000 K in ultra-high vacuum prior to the measurements, in order to reduce residues and contamination at the SLG/YIG interface. The XAS and XMCD measurements were performed at BL-7A of Photon Factory (KEK, Japan). The XAS and XMCD spectra were taken at the C *K*-edge, O *K*-edge and Fe $L_{2,3}$-edge in the total electron yield (TEY) and partial electron yield (PEY) modes with larger and smaller probing depths (~5 nm in the TEY mode and < 2 nm in the PEY mode), respectively.

Figure 1 shows the C *K*-edge XAS spectra taken in the PEY mode with linearly polarized x-ray beams at grazing incidence (GI) (θ = 30°) and normal incidence (NI) (θ = 90°), respectively. A sharp peak A_1 (~285 eV) and a broad structure A_2 (~292 eV) in the spectra are in agreement with those of pristine graphite and attributed to the transition from the 1s to unoccupied π and σ states of graphene, respectively. The remarkable change of the π peak intensity between GL and NI confirms the adhesion of SLG on the surface of the YIG thin film. Since the π peak shape is affected by the electronic structure around the K and M points of graphene, the narrow shape of the π peak indicates that there is no significant modification of the π band of graphene at the SLG/YIG interface different from the strong degradation of the graphitic electronic structure caused by the chemical interfacial interactions [4,5].

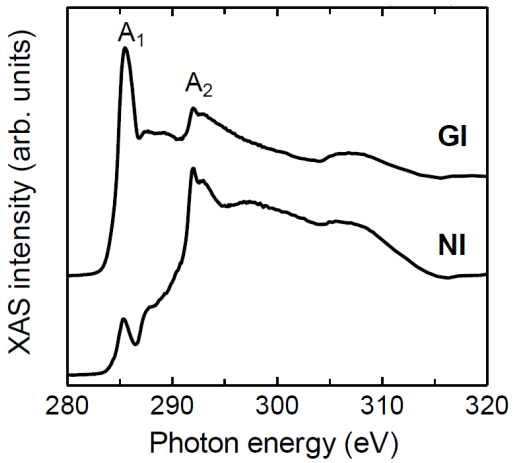

Fig. 1: C *K*-edge XAS spectra of the SLG/YIG heterostructure taken at GI (θ = 30°) and NI (θ = 90°).

Figures 2a and 2b show (a) the XAS and XMCD spectra at the Fe $L_{2,3}$-edge taken at GI in the TEY and PEY modes, and (b) the magnetic hysteresis loops obtained from the asymmetry of the XAS intensity at the L_3 and L_2 edges; $I(L_3)-I(L_2)$ / $I(L_3)+I(L_2)$, as a measure of the magnetization. In Fig. 2a, the spectra were measured at 100 K in remanence after magnetic saturation by applying a pulsed magnetic field (300 Oe) along the incidence direction of circularly-polarized x-ray beams (the circular polarization factor P_C = 0.8). In both the TEY and PEY modes, the XAS spectra show a double-peak structure at both the L_3 and L_2 edges consistent with the previous report for YIG [6]. The shapes of the XAS and XMCD spectra are almost similar regardless of the different probing depth in the TEY and PEY modes. This indicates that the proximity contact with SLG has a minor impact on the electronic structure and magnetism of YIG even in the vicinity of the SLG/YIG interface, possibly related to the physical nature of the interfacial interactions between graphene and YIG which can be deduced from the graphitic feature in the C *K*-edge XAS spectra (Fig. 1). The weak XMCD intensity in remanence (Fig. 2a) is ascribed to the small magnetization of YIG in remanence

as shown in Fig. 2b. The negligible remanent magnetization of YIG at 300 K might be responsible for the unsuccessful detection of spin polarization of graphene in the SLG/YIG heterostructure at 300 K in contrast to the remarkable spin polarization at 100 K [3].

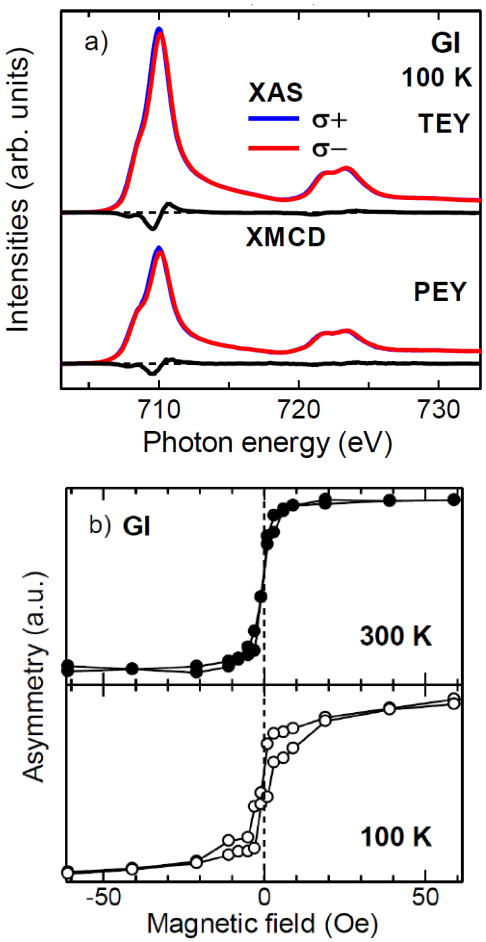

Fig. 2: a) Fe $L_{2,3}$-edge XAS and XMCD spectra of the SLG/YIG heterostructure taken at GI and b) magnetic hysteresis loops taken at GI with sweeping the magnetic field along the x-ray incident direction. The measurement temperatures are 100 K in Fig. 2a and 300 K (solid circles) and 100 K (open circles) in Fig. 2b.

Figure 3 shows the TEY and PEY O *K*-edge XAS spectra taken at GI with linearly polarized x-ray beams. A sharp peak B_1 (~529 eV) and a broad structure B_2 (~535 eV) are attributed to the transitions to the unoccupied oxygen 2p states (B_1) hybridized with Fe 3d band and (B_2) weakly hybridized with Fe 4s and 4p states, respectively. [7]. It is notable that the intensity of peak A with respect to peak B is small in the PEY spectrum compared to that in the TEY spectrum. The decrease of peak A can be attributed to the decrease of the hole population in the O 2p-Fe 3d hybridized states. The redistribution of the charge density between the less electronegative carbon to the more electronegative oxygen at the SLG/YIG interface might lead to the p-type doping of SLG as observed in SPMDS [3] and also the decrease of the hole population in the O 2p-Fe 3d hybridized states as suggested from the PEY spectrum with higher sensitivity to the interface region located near the sample surface.

Fig. 3: O *K*-edge XAS spectra of the SLG/YIG heterostructure taken at GI in the TEY and PEY modes.

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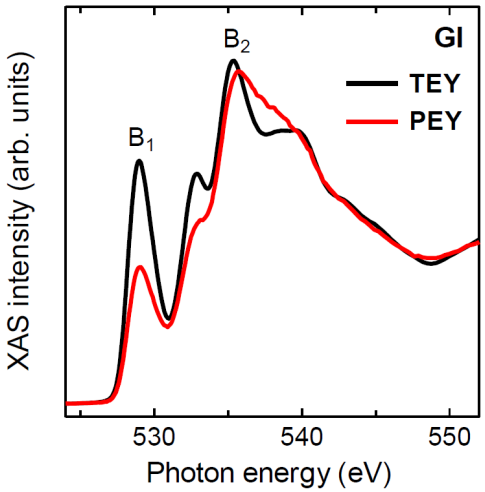

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