Magnetic Structure of the Single-Layer Graphene/Nickel Interface

We investigated the magnetic structure of the single-layer graphene/Ni(111) interface with depth-resolved X-ray magnetic circular dichroism spectroscopy. Ni *L*-edge analysis clarified that the easy magnetization direction changes from in-plane to out-of-plane in the Ni atomic layers near the interface, possibly associated with π-*d* hybridization. C *K*-edge analysis demonstrated that intensification of the spin–orbit interactions and spin polarization are induced in the π band region by the Ni atoms located near the interface. These results indicate the importance of understanding the interfacial magnetic structures for the development of graphene-based spin devices.

Spintronics is a new field of electronics that may lead to novel devices taking advantage of both the charge and spin of electrons. Graphene-based spintronics is a particularly active area of research because of the great potential of graphene for spin transport as represented by the long spin-diffusion length and extremely high carrier mobility. Efficient injection of spin-polarized currents into graphene is one of the most significant issues in developing graphene-based spintronics. Despite the theoretical prediction of the spin filtering effect of graphene/ferromagnetic metal (FM) interfaces [1], a satisfactory high efficiency of spin injection has not been reported yet [2]. Elucidation of the magnetic structures of graphene/FM interfaces may clarify the underlying reasons for the difficulty of efficient spin injection in graphene-based devices. In the present study, the electronic and magnetic structures of graphene/FM interfaces are being investigated for the bilayer structure of single-layer graphene (SLG) and Ni(111) thin film by applying X-ray magnetic circular dichroism (XMCD) with atomic-layer level depth-resolution [3].

Figures 1(a) and (b) show the Ni *L*-edge XMCD spectra measured at two different incident angles of the circularly-polarized X-ray beam. The X-ray incident angles of 30° and 60° are sensitive to the magnetic moments aligned in the in-plane and out-of-plane directions, respectively. The XMCD spectra were obtained by collecting the Auger electrons at different values of detection angle θ using a 2D electron detector. Since the effective escape depth of the emitted electrons changes depending on the detection angle due to the scattering attenuation in the sample, the relative fraction of the signals from the Ni atomic layer near the interface increases with decreasing detection angle θ. In Fig. 1(a), the XMCD intensity at $\theta = 30°$ (red line) is larger than that at $\theta = 3°$ (blue line), and this relationship is inverted in Fig. 1(b). This contrasting behavior indicates that the state of the magnetic moment in the Ni(111) thin film changes depending on the distance from the interface. To gain insight into the magnetic structure of the interface, the total magnetic moment (M_{tot}) and its angle γ from the in-plane direction were estimated for the Ni atoms near and far from the interface ($Ni_{interface}$ and Ni_{bulk}) by applying the magnetic sum rules [4, 5]. The obtained values were $M_{tot} = 0.55\mu_B$ and $\gamma = 87°$ for $Ni_{interface}$, and $M_{tot} = 0.66\mu_B$ and $\gamma = 3°$ for Ni_{bulk}. M_{tot} of Ni_{bulk} agrees well with that of the Ni(111) thin film [6]. This indicates that the Ni magnetic moment shows perpendicular magnetic anisotropy (PMA) at the interface, different from the in-plane orientation in the Ni(111) thin film.

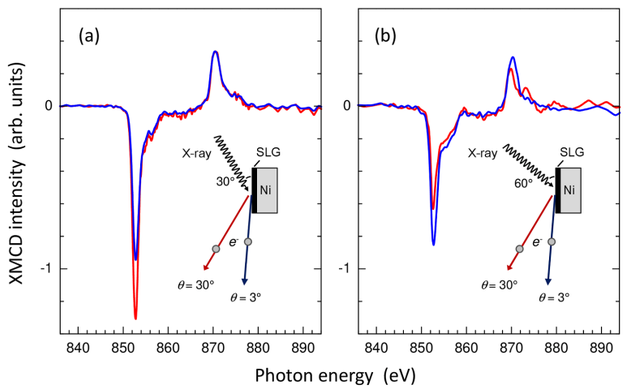

Figure 1: Depth-resolved Ni *L*-edge XMCD spectra at the X-ray incidence angles of 30° (left panel) and 60° (right panel). The blue and red lines in the spectra represent the data at the detection angle θ of 3° and 30°, respectively.

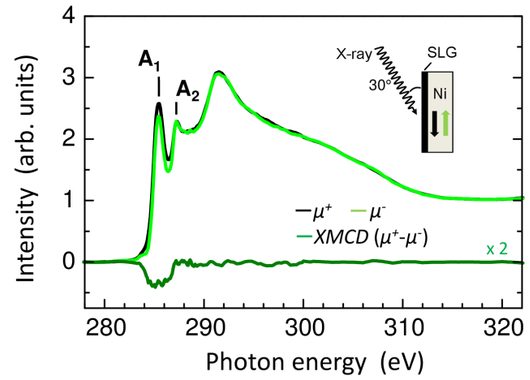

Figure 2: C *K*-edge XMCD spectra at the X-ray incidence angle of 30°. The black and light green spectra are the X-ray absorption spectra measured with the parallel (μ^+) and antiparallel (μ^-) Ni remanent magnetization to the propagation direction of the circularly-polarized X-ray beam.

Figure 2 shows the C *K*-edge XMCD spectrum measured at the X-ray incidence angle of 30° in remanence. Prominent XMCD signals can be seen at around the A_1 and A_2 peaks, which are assigned to the excitations from the 1s states to the hybridized states between the π^* (p_z) states around the *K*- and *M*-points of graphene and the Ni 3*d* states, respectively. Due to the absence of spin–orbit interactions (SOI) in the 1s initial states, the C *K*-edge XMCD is expected to reflect only the orbital magnetic moment of the 2*p* states. The intrinsic SOI strength of graphene is too small to generate sufficiently large orbital magnetic moment to be detectable as XMCD signals. It is therefore concluded that the spin polarization and also the enhancement of SOI are induced in SLG through π–d hybridization. This conclusion is in good agreement with theoretical studies [7].

In summary, the interface magnetic structure in SLG/Ni(111) thin film was investigated by depth-resolved XMCD spectroscopy. The results showed that on the Ni side of the interface the preferable spin orientation direction changes from in-plane to out-of-plane toward the interface, and on the SLG side not only the spin polarization but also the SOI enhancement are induced in the π^* states. The interfacial PMA could disturb the operation of graphene-based spin devices with conventional FM electrodes with in-plane magnetization. The present study highlights the importance of the design of interface magnetic structures at the atomic layer level for graphene-based spin devices.

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