Graphene/Half-Metallic Heusler Alloy: A Novel Heterostructure towards High-Performance Graphene Spintronic Devices

A novel heterostructure consisting of single-layer graphene (SLG) and half-metallic $Co_2Fe(Ge_{0.5}Ga_{0.5})$ (CFGG) Heusler alloy ferromagnet were synthesized, and the electronic properties of the interface were examined by depth-resolved X-ray magnetic circular dichroism spectroscopy. We found that in the SLG/CFGG heterostructure the SLG has a quasi-free-standing nature and the CFGG preserves the half-metallic ferromagnetism in the vicinity of the interface, which suggested that the SLG/CFGG heterostructure possesses distinctive advantages over other reported graphene/ferromagnet heterostructures for realizing high-performance spintronics devices.

Graphene-based vertical spin-valve (SV) is expected to offer a large magnetoresistance effect without impairing the high electrical conductivity, which could pave the way for the next generation of spintronic memory devices offering higher speed and lower power consumption. However, since the ferromagnets adopted to date in graphene-based vertical SV studies are limited to materials with low spin polarization, the graphene-based vertical SV has failed to perform as expected due to the lack of a graphene/ferromagnet heterostructure that can provide highly efficient spin transport. We recently succeeded in the fabrication of a novel heterostructure [1] consisting of CVD-grown high-quality single-layer graphene (SLG) on a magnetron sputtered half-metallic full Heusler alloy $Co_2Fe(Ge_{0.5}Ga_{0.5})$ (CFGG, hereafter) [2] thin film with high spin polarization. We investigated the interfacial properties of the SLG/CFGG heterostructure by means of depth-resolved X-ray magnetic circular dichroism spectroscopy in order to evaluate the feasibility for spintronic device applications.

The quality of the SLG grown on CFGG was characterized by Raman spectroscopy as shown in Fig. 1(a). The Raman peaks of graphene (the D, G, and 2D bands) are visible in the spectrum. The small intensity of the defect-induced D band compared to the G band suggests a high crystal quality of SLG formed on CFGG. The electronic state of graphene in the SLG/CFGG heterostructure was investigated by X-ray absorption spectroscopy (XAS). Figure 1(b) shows the XAS spectra at the C *K*-edge obtained in the partial-electron-yield (PEY) mode at the X-ray incident angle α of 30° (grazing incidence) and 90° (normal incidence) with respect to the sample surface. We observed a sharp peak (285 eV) due to the π^* resonance (C 1s-π^* transition) of graphene, as well as the peak (290–300 eV) due to the σ^* resonance (C 1s-σ^* transition), where the significant increment in the π^* peak intensity at α = 30° compared to α = 90° shows that SLG is flat-lying on the CFGG surface. The sharpness of the π^* peak in the SLG/CFGG heterostructure is markedly different from that in the heterostructures of SLG and other ferromagnets such as Co and Ni, [3, 4] where a significant splitting and broadening of the π^* peak was reported in association with the orbital hybridization between SLG and metals. This suggests that the SLG on CFGG has a quasi-free-standing nature and CFGG does not cause severe degradation of the electronic properties of graphene in contrast to other graphene/ferromagnet heterostructures.

The magnetic properties at the interface region of the CFGG thin film in the SLG/CFGG heterostructure were explored by depth-resolved (DR) X-ray magnetic circular dichroism (XMCD) spectroscopy. Figures 2(a) and (b) compare the XAS and XMCD spectra at the Co and Fe $L_{2,3}$-edges obtained in the bulk-sensitive total-electron-yield (TEY) mode with the mean probing depth λ_p ~ 5 nm and the DR-PEY mode under the surface-sensitive condition of λ_p ~ 4 Å, which corresponds to the outermost surface located at the SLG/CFGG interface. The spectroscopic features of the TEY and DR-PEY XAS spectrum are compatible with the features in other Co-based Heusler alloys, and no sign of carbide formation is observed in the spectra. Indeed, the $L2_1$ structure of the CFGG in the vicinity of the interface was directly confirmed by reflection high-energy electron diffraction. The total magnetic moments (m_{total}) per atom and their spin and orbital components (m_{spin} and m_{orb}, respectively) evaluated from the TEY XMCD spectrum and the series of the DR-PEY XMCD spectra obtained at different emission angles are summarized in Figs. 2(c) and (d), which demonstrate how the element-specific magnetic moments change with depth in the region near the interface. It is found that, except for the outermost surface (λ_p ~ 4 Å) adjacent to SLG, m_{total} of both Co and Fe gradually decreases by ~10% from the values in the bulk region as λ_p decreases. In addition, enhancements of the magnetic moments in the outermost region (λ_p ~ 4 Å) are also commonly observed for Co and Fe, which can be responsible for the orbital rearrangement in the atomic layer terminating the CFGG surface [1]. The preservation of the large magnetic moment throughout the interface region is in sharp contrast to the significant reduction as reported for the SLG/Fe [5] and SLG/Ni [6] heterostructures, where chemical bonding is formed between graphene and ferromagnets. In summary, a newly developed SLG/CFGG heterostructure is found to show several remarkable features such as the quasi-free-standing nature of graphene with little modification of the π band, and the robust magnetism of CFGG even at the interface. This unusual preservation of the inherent electronic properties at the SLG/CFGG interface could lead to a dramatic improvement in the efficiency of spin transport in graphene-based vertical SV, and paves the way for the future development of high-performance spintronic devices based on the graphene/half-metallic Heusler alloy heterostructures.

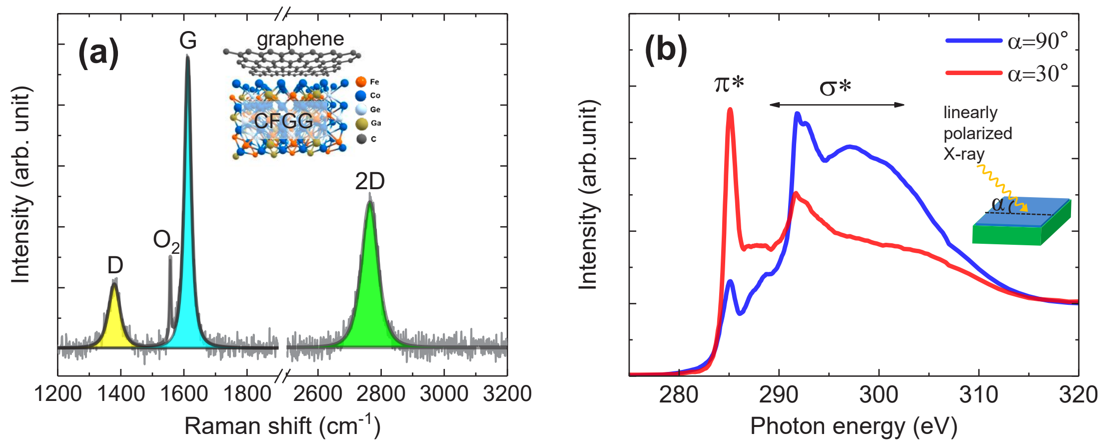

Figure 1: (a) Raman spectrum of SLG/CFGG heterostructure. (b) C *K*-edge XAS spectra measured at the incident angles of α = 30° (red solid line) and α = 90° (blue solid line). The sharp peak at 1557 cm^{-1} of (a) is attributed to O_2 in ambient air. The inset of (a) is a sketch of the SLG/CFGG heterostructure.

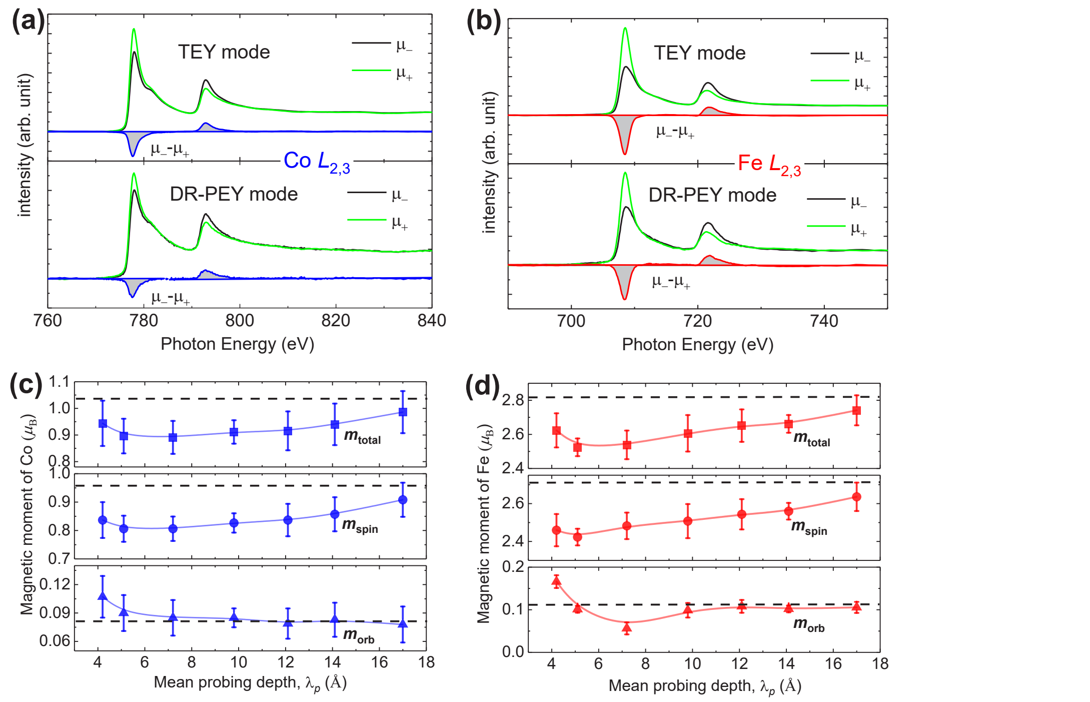

Figure 2: (a, b) Comparison of XAS and XMCD spectra of SLG/CFGG heterostructure at Co and Fe $L_{2,3}$-edges obtained in TEY and DR-PEY modes. (c, d) Plots for orbital, spin and total magnetic moment of Co, and Fe *vs.* mean probing depth (λ_p). The DR-PEY spectra in (a) and (b) are at λ_p ~ 4 Å. The dashed lines in (c) and (d) indicate the magnetic moments evaluated from the TEY XMCD spectra.

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