Spin polarization at the graphene-ferromagnetic metal interface

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Graphene attracts much attention as the most promising material for realizing molecular spintronic devices, due to the possible extremely long spin relaxation length which reflects the weak spin-orbit interaction, hyperfine interaction and the specific π band structure. In the graphene spintronic devices, the control of the spin injection/detection processes through the contact between graphene and ferromagnetic metal (FM) electrodes are crucial for the device operation. Therefore, the elucidations of the electronic structures of the graphene/FM interface including the spin polarization in graphene are essentially important for designing molecular spintronic devices based on graphene.

In order to investigate the interface electronic structures, we need to address the following two issues. The first issue is the formation of a well-defined interface between graphene and FM. So far, several fabrication methods have been developed for the deposition of graphene on various substrates, such as micromechanical exfoliation (ME) from bulk graphite [1] and ultrahigh vacuum chemical vapor deposition (UHV-CVD) [2]. ME results in small flakes of graphene with significant inhomogeneity in the atomic-layer thickness and electronic state. Moreover, contamination and oxidization of the graphene/substrate interface are unavoidable in practice during the fabrication processes. On the other hand, UHV-CVD can be an alternative fabrication method to overcome the above problems. The chemical growth of graphene on the clean catalytic metal surface in UHV enables the interface to be kept free from contamination. In the present study, UHV-CVD was adopted for the deposition of graphene on the FM substrates. The second issue is to employ an appropriate spectroscopic technique which allows highly-sensitive detection of atomically thin graphene. The conventional techniques such as spin-resolved ultraviolet photoelectron spectroscopy are not satisfactory due to the superposition of large background signals from the substrate and small signals from graphene. We have directed our attention toward spin-polarized metastable-atom deexcitation spectroscopy with the extremely high sensitivity to the topmost atomic layer on the sample surface, which makes it possible to selectively detect the spin-resolved electronic structure of graphene contacted with FM substrates.

The graphene with coverage ranging from 0% to 100% was deposited on the ferromagnetic nickel, Ni(111), surface by controlling the exposure amount of benzene, which is a precursor for the graphene growth, in UHV-CVD [3]. The spin-resolved electronic structure of graphene was investigated by SPMDS as follows: The secondary electron emission was detected during irradiation of the spin-polarized triplet metastable helium atom, He*(2^3S), beam with a thermal velocity onto the sample surface. The ejected electron intensity (SPMDS spectrum) depends on degree of the spin polarizatin of the surface electrons since only a surface electron with the same sign of spin as that of the 1s hole of He*(2^3S) can be transferred to and fill the 1s hole following the Pauli's exclusion principle [4].

Figure 1 shows the spin asymmetry $(I_p – I_a)/(I_p + I_a)$ obtained from the SPMDS measurements, where I_p and I_a represent the secondary electron intensities obtained under irradiation of the spin-polarized He*(2^3S) beams with electron spins parallel and antiparallel to the majority spin of the Ni substrate, respectively. The sign and magnitude of the spin asymmetry change with the coverage of graphene, and, at the coverage of 100%, the asymmetry becomes approximately -2% at the Fermi energy of graphene. This negative spin asymmetry of graphene is different from the positive asymmetry of approximately +7% at the Fermi energy of Ni measured for the Ni(111) clean surface at the coverage of 0%. The above results lead to the conclusion that the conduction electrons in graphene at the graphene/Ni(111) interface have the opposite sign of the spin polarization with respect to those in Ni(111) [5]. It is suggested that the opposite spin polarization as clarified in the present study possibly results in the degradation of the spin polarization of the charge current passing through the interface between graphene and FM in spin valve devices.

The present study has, for the first time, provided direct information on how the conduction electrons in graphene are affected by the contact with FMs. Our research achievement is expected to greatly contribute to the designing of spin-related properties of molecular spintronic devices by taking advantage of graphene and other two-dimensional materials.

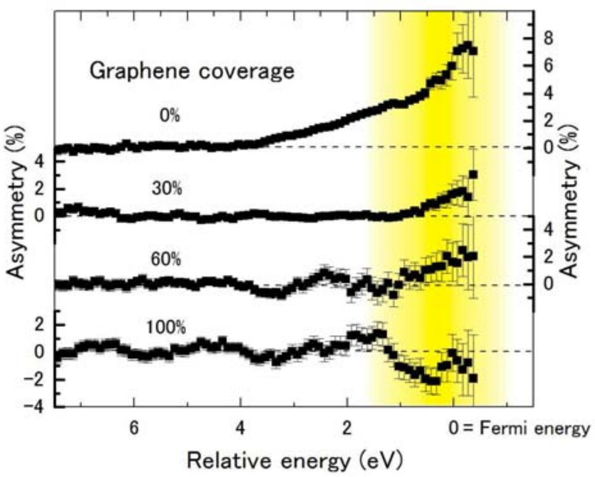

Fig.1 Spin asymmetries of graphene/Ni(111) obtained from the SPMDS measurements with the He*(2^3S) beams aligning the spin parallel and anti-parallel to the majority spin of Ni(111).

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