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Tunnel Spin Injection into Graphene Using Al_2O_3/PTCA Barrier Grown by Atomic Layer Deposition

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1. Introduction

Graphene is a particularly promising material for application to spintronics because it is expected to have a very long spin-diffusion length due to its weak spin-orbit and hyperfine interactions. In the spintronics, electrical spin injection from a ferromagnetic electrode (FM) to single-/multi-layer graphene is a key technology and it has recently been studied by several research groups. Toward more efficient spin injection, various type of interfaces between FM and graphene have been examined, including direct deposition of FM on graphene, reduction of contact area, insertion of a low-temperature-grown Al_2O_3 tunnel barrier, and a molecular beam epitaxy (MBE)-grown MgO tunnel barrier. Very recently, a large spin transport signal has been observed by non-local magnetoresistance (NLMR) measurements when the tunnel barrier is inserted between the FM and graphene. On the other hand, there is a strong demand for further improvement of spin injection efficiency, spin life time, and quality of the FM/graphene interface because the observed spin life time is still shorter than theoretical expectations and the spin injection yield is strongly influenced by the fabrication method. Thus, it is necessary to explore more reliable fabrication methods and alternative types of tunnel barrier material for fabricating uniform and pinhole-free tunnel barriers on the graphene surface.

Atomic layer deposition (ALD) is an emerging new technology for depositing smooth and high-quality oxide films. Given its self-limiting growth process, ALD has the potential to control the thickness of deposited layers on the order of the atomic level. Thus, ALD is a very promising method to fabricate thin tunnel barriers on the graphene surface. On the other hand, a clean graphene surface is inert for ALD precursors because there are no available dangling bonds. Recently, however, ALD of Al_2O_3 on graphene surfaces has been demonstrated using non-covalent surface functionalization layers. Among these, the 3,4,9,10-perylene tetracarboxylic acid (PTCA) self-assembled monolayer (SAM) [1] is advantageous for the fabrication of tunnel barriers on graphene.

2. Experimental

Single- and multi- layer graphene is fabricated by mechanically exfoliating Kish graphite and transferring it to SiO_2/n-Si substrate. Subsequently, ALD-Al_2O_3/PTCA is fabricated by the following sequence of steps. First, the sample is annealed at 600 °C in vacuum with 1 Torr argon atmosphere for surface cleaning. This produces a clean graphene surface with an RMS roughness of ~0.15 nm. Second, after the sample is dipped in the PTCA solution, some of the physically absorbed PTCA molecules are removed by rinsing with methanol and water. The graphene surface is covered with PTCA SAM at the end of this step. Third, Al_2O_3 is fabricated by ALD using trimethyl aluminum (TMA) and water vapor as precursors at sample temperature of 100 °C. The -OH groups of PTCA act as bonding sites for TMA precursor gas. This enables the deposition of TMA, which is subsequently oxidized by water vapor and converted to an Al-O atomic layer. The pulse/purge times used for TMA and water vapor are 0.1 s/15 s and 0.1 s/40 s, respectively. These conditions provide a growth rate of 0.12–0.15 nm/cycle for Al_2O_3. The RMS roughness of the surface after 11 cycles of Al_2O_3 deposition is ~0.17 nm; this is about the same as the RMS roughness before Al_2O_3 deposition. After the graphene and SiO_2 on the substrate surface is completely covered with Al_2O_3, ferromagnetic permalloy (Py = $Ni_{81}Fe_{19}$) and non-magnetic Au/Ti electrodes are fabricated with standard electron beam (EB) lithography and EB evaporation.

3. Results and discussion

The NLMR curve are measured under in-plane field $B_{//}$ using the 11-cycle ALD device at V_{BG} = 15 V as shown in Fig. 2. A clear NLMR signal is observed, and the change in non-local magnetoresistance R_{NL} is up to ~30 Ω at 45 K. Even though tunnel barrier fabrication process is not fully optimized yet, NLMR signal of exceeding ~10 Ω is obtained. If we take into account the fact that this is the first demonstration of ALD-Al_2O_3 as a tunnel spin injector, we think the observed amplitude of NLMR is very promising compared with the previously observed NLMR on a single-layer graphene.

To confirm whether we achieved tunnel spin injection into graphene, we examined the dependence of ΔR_{NL} on

V_{BG} for the device which has highest ΔR_{NL}. According to a simple spin drift-diffusion model, the shape of the function relating ΔR_{NL} and V_{BG} dramatically changes with the balance between the spin resistance of graphene R_G and the contact resistance R_J. When R_J increases, the function relating ΔR_{NL} and V_{BG} shows a clear transition from a valley shape (transparent) to a peak shape (tunneling). The experimental data show good agreement with the case of tunneling regime. This indicates that a high-quality tunnel barrier is formed with the ALD-grown Al_2O_3 barrier.

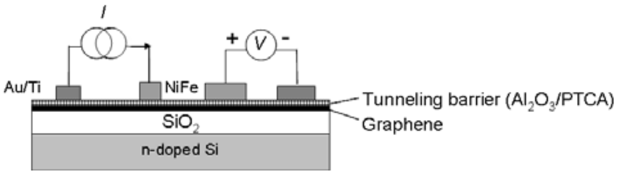

Fig. 1 A schematic diagram of the graphene spin-valve device with an Al_2O_3/PTCA tuunel barrier. Magnetoresistance is measured in non-local geometry.

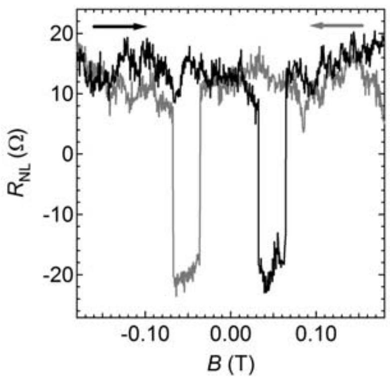

Fig. 2 Non-local magnetoresistance observed in the graphene spin-valve device (T = 45 K). Al_2O_3 layer was deposited with 11 ALD cycles.

4. Conclusions

We demonstrated electrical spin injection from a ferromagnet to graphene using an ALD-Al_2O_3/PTCA composite tunnel barrier. The results showed that tunnel spin injection is achieved and that a large non-local magnetoresistance signal is obtained. These performance characteristics of the Al_2O_3/PTCA composite tunnel barrier have potential applications for future graphene spintronics.

Acknowledgements

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