High Magnetoresistance in Fully Epitaxial Magnetic Tunnel Junctions with a Semiconductor GaO_x Barrier

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Abstract

We have fabricated magnetic tunnel junctions with fully epitaxial $Fe(001)/GaO_x(001)/Fe(001)$ structure, where the GaO_x is a wide bandgap semiconductor with a cubic spinel-type crystal structure. Tunneling magnetoresistance (MR) ratios up to 92 % (125%) were observed at room temperature (20 K), which evidently indicates the existence of a spin-polarized coherent tunneling. Such a single-crystalline semiconductor tunnel barrier that shows a high MR ratio is an essential building block for a vertical-type spin field-effect transistor.

1. Introduction

Magnetoresistance (MR) effect is one of the most crucial operation principles in semiconductor (SC)-based emerging devices such as spin field-effect-transistors (spin-FET). Spin-dependent transport in SC is usually studied by using a planar-type device configuration, which has ferromagnetic (FM) source/drain electrodes on SC channel layer. However, the MR ratios so far achieved are still too low to meet the requirements for practical application of the spin-FET. Recently, Kanaki *et al.* [1] fabricated a vertical-type spin-FET based on (Ga,Mn)As/GaAs/(Ga,Mn)As magnetic tunnel junction (MTJ). They demonstrated the modulations of (Ga,Mn)As drain-source current and MR ratio by applying gate electric fields at low temperature. However, for room temperature (RT) operation, a new SC-based MTJ with *3d*-FM metal electrodes should be developed.

Gallium oxide (GaO_x) with a band-gap of ~ 5 eV has recently attracted much attention because of its great potential in various SC devices including FET [2]. This material is known to have several crystal structures, one of which is a cubic MgAl₂O₄-type spinel structure (γ -phase). It is noteworthy that the lattice constant of γ -Ga₂O₃ (0.824 nm) gives a relatively small lattice mismatch (~ 1.7 %) when the Fe unit cell is turned by 45° with respect to the γ -Ga₂O₃ unit cell in the same manner as the Fe(001) on MgAl₂O₄(001) [3]. In this study, we have developed a fully epitaxial MTJ with Fe(001)/ γ -GaO_x(001)/Fe(001) structure and demonstrated high MR ratio at RT.

2. Sample preparations

MTJ films shown in Fig.1 were prepared on MgO(001) substrates by molecular beam epitaxy using electron-beam evaporation. The Fe bottom electrode was grown on the substrate at 100 °C, followed by an *in situ* annealing at



Fig. 1 Schematic structure of the magnetic tunnel junction (MTJ) stack and *in situ* annealing conditions.

350°C for 10 minutes. A thin MgO seed layer with a thickness of 0.4 nm or 0.7 nm was then epitaxialy grown on the Fe electrode at 100° C. The single-crystalline GaO_x tunnel barrier was formed by solid phase epitaxy. The GaO_x layer was first deposited on the MgO insertion layer at 80 °C under an O_2 pressure of 1×10^{-6} Torr. No diffraction patterns were observed in reflective high-energy electron diffraction (RHEED) image, indicating an amorphous GaO_x. Then, an in situ annealing was carried out at temperature (T) up to 500°C under an O₂ pressure of 1×10^{-7} Torr. Clear streaky patterns appeared in the RHEED image after the annealing process (Fig. 2(b)), indicating the formation of a single-crystalline GaO_x with an atomically flat surface. Thanks to the single-crystalline barrier, we were able to epitaxialy grow the Fe upper electrode, which was grown at 100°C and then annealed for 10 minutes at 350°C. Finally, Co pinned and Au cap layers were deposited onto the Fe upper electrode at RT. As a reference, we also prepared the same MTJ stack without performing the *in situ* annealing for the GaO_x barrier and the upper Fe electrode.



Fig. 2 RHEED images of the GaO_x layer (a) as-grown and (b) after an *in situ* annealing at 500°C.

The samples were patterned into tunnel junctions (3 × 12 μ m²) using conventional micro-fabrication techniques (*e.g.*, photolithography, Ar ion milling, and SiO₂ sputtering) and magnetoresistance measurements were carried out at 20 K and RT.

3. Results

The cross-sectional scanning transmission electron microscopy (STEM) image (Fig. 3(a)) revealed a fully single-crystal MTJ stack with very few dislocations at the barrier/electrode interfaces. From the STEM image in the range of about 50 nm, the in-plane lattice mismatch $\Delta a/a$ between the barrier and Fe electrodes was estimated to be between 0.6% - 1.0%, where the lattice constant of Fe was smaller. The $\Delta a/a$ value is in reasonable agreement with that expected from the lattice constants of bulk Fe and γ -Ga₂O₃ (~ 1.7%). We confirmed the presence of two distinct layers, GaO_x and MgO, from the elemental mapping by energy dispersive X-ray spectroscopy (EDS). The Fig. 3(b) presents the electron nano-beam diffraction (NBD) pattern around the GaO_x layer. As confirmed by the simulated NBD pattern in Fig. 3(c)), it can be clearly assigned to the γ -Ga₂O₃(001). The epitaxial relations between the electrodes and the barrier layers were determined to be top $Fe(001)[110] \parallel \gamma$ -GaO_x(001)[100] \parallel MgO(001)[100] \parallel bottom Fe(001)[110].

Figures 4(a) and 4(b) show typical MR curves of the epitaxial and reference MTJs, respectively. The MR ratio was largely enhanced by the *in situ* annealing of the GaO_x barrier. The MR ratios of the epitaxial MTJ was 92% at RT (125% at 20 K), which is a few times those of the reference MTJ (34% at RT and 50% at 20 K) and those reported for GaO_x-based MTJs with polycrystalline FM electrodes [4]. The observed large MR ratio is considered to be due to the coherent spin-polarized tunneling which has been observed in MTJs with MgO [5,6] and MgAl₂O₄ [3] barriers.



Fig. 3 (a) Cross-sessional STEM image of the MTJ, (b) NBD pattern around the GaO_x layer ([100] azimuth of the MgO substrate), and (c) simulated NBD pattern of a spinel-type $Ga_2O_3(001)$ ([100] azimuth).



Fig. 4 Typical magnetoresistance (MR) curves of the (a) epitaxial and (b) reference MTJs at 20 K and RT. The total barrier thickness is 2.1 nm ($d_{GaO} = 1.4$ nm) and 2.7 nm ($d_{GaO} = 2.0$ nm) for the epitaxial and for the reference MTJs, respectively.

4. Conclusions

We have successfully fabricated fully epitaxial $Fe(001)/\gamma$ -GaO_x(001)/MgO(001)/Fe(001) MTJs and observed relatively high MR ratios (up to 92% at RT and 125% at 20 K). Such high MR ratios indicate the coherent spin-polarized tunneling in the epitaxial MTJs. This result demonstrates the high potential of GaO_x as a high-quality tunnel barrier material for SC-based MTJs.

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