Fabrication of Fully Epitaxial Magnetic Tunnel Junctions with CoFe Electrodes and a MgO Barrier on Ge(001) Substrates via a MgO Interlayer

Gui-fang Li, Tomoyuki Taira, Hong-xi Liu, Ken-ichi Matsuda, Tetsuya Uemura, and Masafumi Yamamoto

Division of Electronics for Informatics, Hokkaido University, Sapporo 060-0814, Japan Phone: +81-11-706-6442, E-mail: li-guifang@nsed.ist.hokudai.ac.jp

1. Introduction

Spintronic devices that manipulate the spin degree of freedom of the electron are being extensively studied, which could lead to novel functionalities such as nonvolatility and reconfigurability.^{1,2} In particular, injection of spin-polarized electrons into semiconductor channels and manipulation of the injected spins have been intensively studied recently.³ The use of spin sources with the magnetization-reversal capability by a spin-polarized current is a promising method for providing reconfigurable logic functions in semiconductor-based spintronic devices. The use of a magnetic tunnel junction with this capability as a source or drain electrode for spin-based field-effect transistors (spin FETs) is highly required. Furthermore, the monolithic integration of MTJs and MOSFETs would lead to nonvolatile logic gates featuring ultralow-power logic circuits.⁴ On the other hand, a semiconductor Ge channel is highly favorable for future-generation metal-oxide-semiconductor (MOS) devices because of the high mobility of electrons and holes in Ge.⁵ Recently, ferromagnetic thin films of Fe,⁶ CoFe⁷ and Heusler alloy Co₂MnSi⁸ were prepared epitaxially on Ge(001) single-crystal substrates via a MgO interlayer. In these works, crystallographic relationship of MgO(001)[110] [Ge(001)[100] for a MgO thin layer grown on a Ge(001) substrate was confirmed. This relationship is as expected from the relatively small lattice mismatch (5.2%) between MgO(001) and Ge(001) on a 45° in-plane rotation and in contrast to the cube-on-cube epitaxial growth of MgO on GaAs(001)⁹ and MgO on Si(001)¹⁰ with a 4:3 coincident site lattice. Given these background, the purpose of the present study was to fabricate fully epitaxial MTJs on Ge(001) single-crystal substrates via a MgO interlayer. This is because the monolithic integration of MTJs with Ge FETs would lead to future-generation nonvolatile logic gates. Furthermore, a MgO interlayer with an appropriate tunnel resistance in a MTJ/MgO interlayer/Ge(001) heterostructure will be served as a tunnel barrier for spin injection¹¹ from an MTJ in a Ge channel. We will show a relatively high tunnel magnetoresistance (TMR) ratio of up to 212% at 293 K for prepared fully epitaxial $Co_{50}Fe_{50}$ (CoFe)/MgO/CoFe MTJs on a Ge(001) substrate via a 10-nm-thick MgO interlayer.

2. Experimental

The fabricated layer structure was as follows: (from the substrate side) MgO interlayer (5 or 10 nm)/ CoFe lower

electrode (50 nm)/MgO barrier (1.4–3.2 nm)/CoFe upper electrode (3 nm)/Ru (0.8 nm)/CoFe (2 nm)/IrMn (10nm)/Ru cap (5 nm) on a Ge(001) single crystal substrate. Each layer was successively deposited in an ultrahigh-vacuum (UHV) chamber (base pressure of $\sim 6 \times 10^{-8}$ Pa) through the combined used of magnetron sputtering for CoFe and electron beam evaporation for MgO. Before the Ge(001) substrates was installed in the UHV chamber, it was oxidized at 500 °C for 1 hour and then cleaned by HF solution. The Ge substrate was then annealed at 650 °C for 1 hour in the UHV chamber. The MgO interlayer was deposited at a substrate temperature of 125 °C.⁸ The layer structure was annealed *in situ* at 400 °C just after deposition of the CoFe lower electrode and also at 400 °C just after deposition of the CoFe upper electrode.

We fabricated MTJs with the layer structure described above using photolithography and Ar ion milling. The fabricated junction size was $10 \times 10 \ \mu m$. We investigated the structural properties of the heterostructures through reflection high-energy electron diffraction (RHEED) observations. We measured TMR characteristics of the fabricated MTJs using a dc four-probe method.

3. Results and discussion

First, we describe the structural properties of CoFe/MgO/CoFe MTJs grown on Ge(001) substrates via a 10-nm-thick MgO interlayer. RHEED patterns, along two different azimuths [100]_{Ge} and [110]_{Ge}, were obtained in situ for each successive layer in the heterostructure, having 10-nm-thick MgO interlayer in Fig. 1. Here both the CoFe lower and upper electrodes were deposited at RT and successively annealed at 400 °C just after deposition of the respective layers. Streak patterns dependent on the electron injection direction, parallel to [100]_{Ge} and [110]_{Ge}, were obtained for all the layers in the heterostrucure consisting of MgO interlayer/CoFe lower electrode/MgO barrier/CoFe upper electrode. Furthermore, the spacing of the observed streak patterns of all these layers agreed well with that of the Ge(001) substrate for both electron beam injection directions, parallel to [100]_{Ge} and [110]_{Ge}, indicating that the all these layers were grown epitaxially on a Ge(001) substrate with а crystallographic relationship of CoFe(001)[100]||MgO(001)[110]||Ge(001)[100], including the MgO interlayer, the MgO barrier, the lower CoFe and upper CoFe electrodes.

The crystallographic relationship of MgO interlayer(001)[110]||Ge(001)[100] is consistent with previous works on MgO/Ge(001) heterostructures.⁶⁻⁸ And the crystallographic relationship of CoFe lower electrode(001)[100]||MgO interlayer(001)[110] on Ge(001)[100] is consistent with previous work and is as expected from the relatively small lattice mismatch (4.3%) between CoFe(001) and MgO(001) on a 45° in-plane rotation.

Figure 2(a) shows typical a magnetoresistance curve at 293 K for a fabricated fully epitaxial CoFe/MgO/CoFe MTJ (CoFe MTJ) with a MgO interlayer thickness (tinterlayer) of 10 nm. The bias voltage was 5 mV. The MTJ showed clear exchange-biased TMR characteristics with a relatively high TMR ratio of 212% at 293 K. This value is comparable and slightly lower than the TMR ratio of 260% at 293 K obtained for fully epitaxial and identically fabricated CoFe MTJs on MgO-buffered MgO(001) substrates. Figure 2 (b) shows TMR ratios at 293 K for CoFe MTJs on MgO interlayer/Ge(001) substrates as a function of t_{interlayer}. TMR ratios of up to 180% at 293 K were obtained for CoFe MTJs with $t_{\text{interlayer}} = 5$ nm. The slight increase in the TMR ratios with increasing the MgO interlayer thickness may be due to a possible improvement of the structural quality of CoFe/MgO/CoFe MTJ trilayers grown on the MgO interlaver. On the other hand, the observed weak dependence of the TMR ratio on $t_{interlayer}$ suggests that there is room for further decreasing the MgO interlayer thickness. These results suggest the promise of the monolithic integration of epitaxial CoFe MTJs and Ge MOSFETs for future-generation nonvolatile logic circuits. It also suggests the promise of the heterostructure consisting of CoFe MTJ/MgO interlayer/Ge(001) substrate as a key device structure for efficient spin injection into a Ge channel from a MTJ.

4. Conclusion

Fully epitaxial magnetic tunnel junctions (MTJs) with Co₅₀Fe₅₀ (CoFe) electrodes and a MgO barrier were prepared on Ge(001) single crystal substrates via a MgO interlayer. Reflection high-energy electron diffraction patterns observed *in situ* for each layer during preparation clearly indicated that all layers grew epitaxially. The microfabricated epitaxial CoFe/MgO/CoFe MTJs (CoFe MTJs) with a 10-nm-thick (5-nm-thick) MgO interlayer demonstrated a relatively high tunnel magnetoresistance ratio of 212% (180%) at 293 K. These results suggest the promise of the monolithic integration of epitaxial CoFe MTJs and Ge MOSFETs for future-generation nonvolatile logic circuits and of CoFe MTJ/MgO interlayer/Ge(001) heterostructures as a key device structure for efficient spin injection into a Ge channel from a MTJ.

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FIG. 1. RHEED patterns along two different azimuths $[100]_{Ge}$ and $[110]_{Ge}$, corresponding to $[100]_{CoFe}$ and $[110]_{CoFe}$, respectively, obtained *in situ* for each successive layer in the heterostructure consisting of (from the lower side) Ge(001) substrate/MgO interlayer (10 nm)/CoFe lower electrode/MgO barrier/CoFe upper electrode, where both the CoFe lower and upper electrodes were deposited at RT and successively annealed at 400 °C just after deposition of the respective layers.



FIG 2. Typical magnetoresistance curves at 293 K for a fabricated fully epitaxial CoFe/MgO/CoFe MTJ with a MgO interlayer thickness of 10 nm. (b) TMR ratios at 293 K for CoFe MTJs on MgO interlayer/Ge(001) substrates as a function of MgO interlayer thickness.

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