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Epitaxy and Magneto-Transport Properties in Fully Epitaxial Fe/GaO$_x$/Fe Magnetic Tunnel Junctions

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Abstract

We have grown fully epitaxial Fe(001)/GaO$_x$(001)/Fe(001) magnetic tunnel junctions (MTJs) by a solid-phase epitaxy with different growth conditions, where GaO$_x$ is amorphous in the as-grown state. We developed a novel fabrication process that can largely reduce the formation temperature of the fully epitaxial structures from 500°C to 250°C. At room temperature (RT), all the MTJs showed high tunneling magnetoresistance (MR) ratios of about 100% which was almost independent of the formation temperature. The results indicate that GaO$_x$ is an attractive tunnel barrier material for practical device applications.

1. Introduction

Semiconducting materials have recently attracted considerable attention to the tunnel barrier of MTJs because they provide unique properties and functions to the MTJ such as very low resistance-area product [1] and tunability of a tunneling current by electric fields [2].

Very recently, we have reported a high MR ratio up to 92% in fully epitaxial Fe(001)/GaO$_x$(001)/Fe(001) MTJs [3], where the GaO$_x$ is one of the emerging semiconductors for practical applications. Such a high MR evidently indicates the existence of a spin-polarized coherent tunneling as observed in MTJs with MgO [4,5] and $MgAl_2O_4$ [6] tunnel barriers. Although GaO$_x$ is amorphous in the as-grown state, a single-crystalline GaO$_x$ with a $MgAl_2O_4$–type spinel structure was successfully formed by an *in situ* annealing of the as-grown GaO$_x$ layer, the method of which is so called solid-state epitaxy technique. However, the formation temperature of the single-crystalline GaO$_x$ is too high (~500ºC) to apply to practical applications.

In this study, we developed a novel fabrication process that can largely reduce the formation temperature of the fully epitaxial MTJ from 500°C to 250°C.

2. Sample preparations

MTJ films were prepared by molecular beam epitaxy with the same growth system as our previous report [3]. The structure of the MTJ was Au (10 nm) cap / Co (5 nm) pinned layer / Fe (5 nm) upper electrode / GaO$_x$ (2 nm) tunnel barrier / MgO (1 nm) seed layer / Fe (30 nm) bottom electrode / MgO (10 nm) buffer layer on MgO(001) substrates. The Fe bottom electrode was annealed at 350°C for 10 min to improve the surface morphology. After the growth of MgO seed layer, the GaO$_x$ barrier layer was deposited at 80°C under an O_2 pressure of 1×10^{-6} Torr. Then, an *in situ* annealing at the temperature T_{GaO}, where T_{GaO} ranges from 250°C to 500°C, was carried out under an O_2 pressure of 1×10^{-7} Torr. The Fe upper electrode was grown and annealed at T_{Fe} = 250°C under the high vacuum below 1×10^{-9} Torr. The T_{GaO} and T_{Fe} of the present MTJs are listed in Table I. Finally, Co-pinned and Au-cap layers were respectively deposited at RT.

Table I. Sample name, *in situ* annealing temperatures of GaO$_x$ barrier (T_{GaO}) and Fe upper electrode (T_{Fe}) for the MTJ samples.

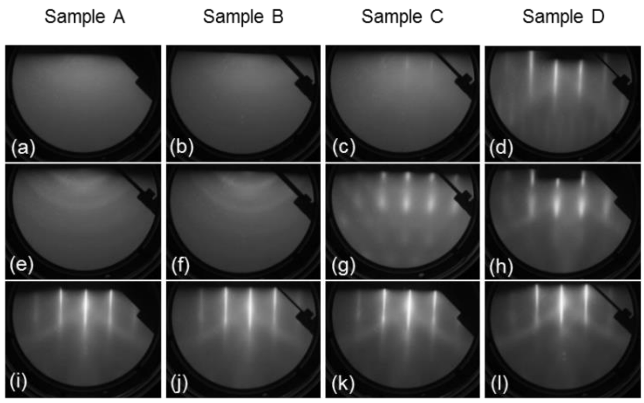
Sample name	T_{GaO} (°C)	T_{Fe} (°C)
A	w/o	250
B	250	250
C	350	250
D	500	250

3. Results

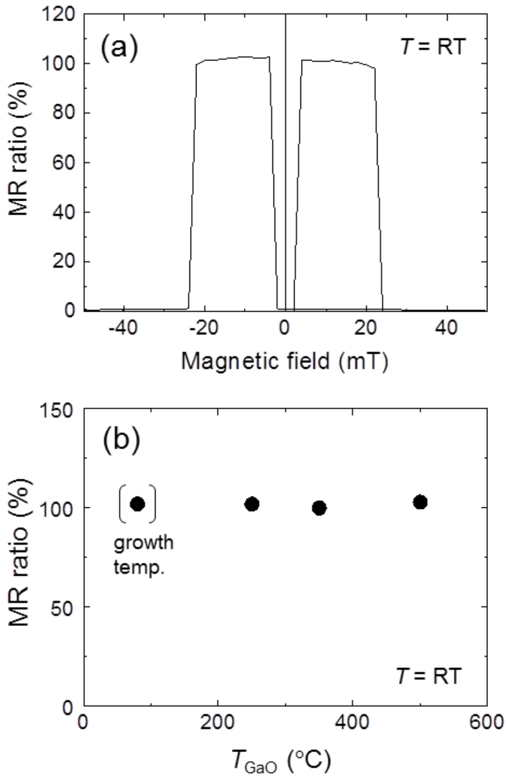
Figures 1 (a)-(l) show reflection high-energy electron diffraction (RHEED) images of the GaO$_x$ barrier layers (upper panels), the Fe upper electrode in the as-grown state (middle panels) and after an *in situ* annealing at T_{Fe} = 250°C (bottom panels) of the MTJs, respectively. For the GaO$_x$ layers, no clear diffraction patterns were observed in the RHEED images for the as-grown state (Fig. 1a) and after the annealing at T_{GaO} = 250°C (Fig. 1b). With increasing T_{GaO}, streaky patterns started to appear at around T_{GaO} = 350°C (Fig. 1c), and finally sharp streaky patterns could be observed at T_{GaO} = 500°C (Fig. 1d). These indicate that the GaO$_x$ barrier layers are amorphous for the samples A and B, mixture of amorphous and crystalline for the sample C and single-crystalline for the sample D, respectively.

The Fe upper electrodes of the samples A and B exhibited broad ring RHEED patterns in the as-grown state (Figs. 1e and 1f), suggesting polycrystalline Fe. In contrast, RHEED images of the samples C and D showed spotty patterns (Figs. 1g and 1h, respectively), implying single-crystalline Fe electrodes. It should be remarked that the broad ring patterns observed in the samples A and B changed to streak ones after an *in situ* annealing at T_{Fe} = 250°C as displayed in Figs. 1(i) and 1(j), respectively. Consequently, the Fe upper electrodes for all the samples revealed similar sharp streak patterns after the *in situ* annealing at T_{Fe} = 250°C. This strongly suggests that a single-crystalline Fe upper electrode can be formed even on the as-grown GaO_x barrier layer without a high temperature annealing up to 500°C.

From the RHEED observations, we can expect the existence of coherent spin-polarized tunneling, and thereby a high MR ratio beyond the Julliere's model even for the samples A and B. Here, MR ratio is defined as $(R_{AP} - R_P)/R_P$ where R_P and R_{AP} are the resistances between the two Fe electrodes with parallel and antiparallel magnetization alignments, respectively. Figure 2(a) shows a typical MR curve of sample A. The MR ratio up to 102% was observed at RT, which is close to the reported value in the fully epitaxial MTJ (92%) [3], strongly suggesting the existence of coherent spin-polarized tunneling. The MR ratios of the present MTJs are summarized in Fig. 2(b). The MR ratio hardly depends on the T_{GaO}, suggesting that there is no remarkable difference in the magneto-transport properties among the MTJ samples.



Figs. 1 RHEED images of the (a) GaO_x barrier layer in the as-grown state, (b)-(d) same layer after *in situ* annealing at T_{GaO}, (e)-(h) Fe upper electrode in the as-grown state, and (i)-(l) same layer after an *in situ* annealing at T_{Fe}, respectively.



Figs. 2 (a) Typical MR curve of sample A and (b) MR ratio as a function of T_{GaO} at RT, respectively.

4. Conclusions

We investigated structural and magneto-transport properties of Fe/GaO_x(MgO)/Fe MTJs grown by different *in situ* annealing conditions for amorphous GaO_x tunnel barrier. Fabrication of fully epitaxial MTJ was possible even without the *in situ* annealing of the GaO_x barrier, resulting in a large reduction on the formation temperature of the fully epitaxial structure from 500°C to 250°C. At RT, all the MTJs showed high MR ratios of about 100% which was almost independent of T_{GaO}. These findings will open a new pathway for developing GaO_x -based practical applications.

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