LARGE REDUCTION OF FABRICATION TEMPERATURE FOR FULLY EPITAXIAL Fe/GaO$_x$/Fe MAGNETIC TUNNEL JUNCTIONS

H. SAITO[1], N. SAI KRISHNA[1], N. MATSUO[1,2], N. DOKO[1,2], and S. YUASA[1]

1) National Institute of Advanced Industrial Science and Technology, Spintronics Research Center
Umezono 1-1-1, Central 2, Tsukuba, Ibaraki 305-8568, Japan

2) On leave from Chiba Institute of Technology, 2-17-1 Tsudanuma, Narashino, Chiba 275-0016, Japan

I. 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. 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. 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.

II. SAMPLE PREPARATIONS

MTJ films were prepared by molecular beam epitaxy. 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 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.

III. 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.

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

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

HIDEKAZU SAITO
E-mail: h-saitoh@aist.go.jp
tel: +81-29-8615457

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. 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] As plotted 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.

IV. 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.

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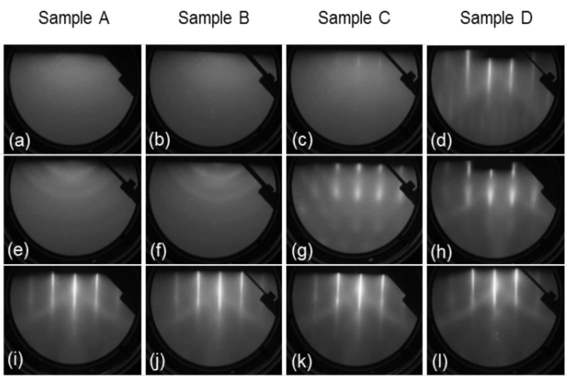
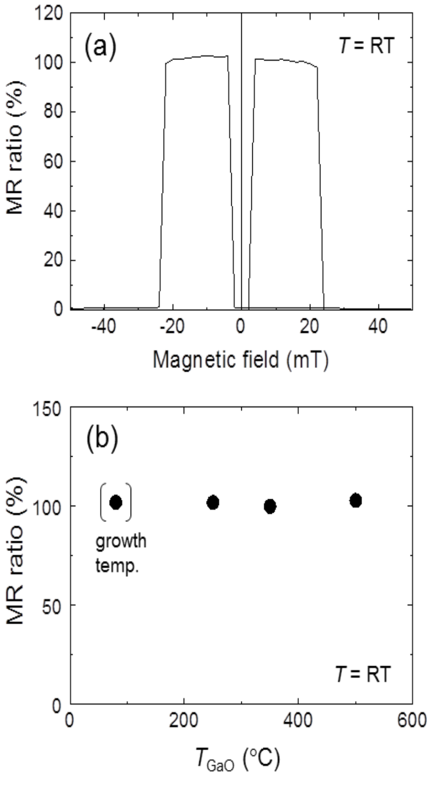


Figs. 1 RHEED images of the (a) GaO_x barrier layer in the as-grown state, (b)-(d) same layer after *in situ* annealing at T_{GaO}, (e)-(h) Fe upper electrode in the as-grown state, and (i)-(l) same layer after an *in situ* annealing at T_{Fe}, respectively.



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