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Characterization of growth and crystallization processes in CoFeB/MgO/CoFeB magnetic tunnel junction structure by reflective high-energy electron diffraction

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We performed reflective high-energy electron diffraction observations to investigate the growth and crystallization processes of $\text{Co}_{60}\text{Fe}_{20}\text{B}_{20}/\text{MgO}/\text{Co}_{60}\text{Fe}_{20}\text{B}_{20}$ magnetic tunnel junction structures. A MgO layer grown on an amorphous CoFeB layer has an amorphous structure up to the MgO thickness (t_{MgO}) of 4 monoatomic layers (ML) and begins to crystallize with (001) preferred orientation when $t_{\text{MgO}} \geq 5$ ML. By annealing, an amorphous CoFeB layer grown on MgO(001) crystallizes in a body-centered-cubic structure with (001) orientation because MgO(001) acts as a template to crystallize CoFeB. The results give important information for understanding the mechanism of giant tunneling magnetoresistance effect in CoFeB/MgO/CoFeB MTJs. © 2005 American Institute of Physics. [DOI: 10.1063/1.2140612]

Magnetic tunnel junctions (MTJs), consisting of two ferromagnetic electrode layers separated by an ultrathin insulating layer (a tunneling barrier), exhibit magnetoresistance (MR) at room temperature (RT).^{1,2} This phenomenon, known as the tunneling MR (TMR) effect, has been extensively studied in an attempt to develop magnetic sensors and magnetoresistive random-access memory. Although an amorphous Al–O tunneling barrier has been mainly used in device applications in the last decade, a crystalline MgO(001) barrier is now considered a key for next-generation device applications because of its giant TMR effect at RT.^{3–8} Based on first-principle theoretical calculations, a huge MR ratio is expected only in epitaxial MTJs, such as $\text{Fe}_{1-x}\text{Co}_x(001)/\text{MgO}(001)/\text{Fe}_{1-x}\text{Co}_x(001)$ ($x=0-1$), as a result of the coherent tunneling of highly spin-polarized Δ_1 Bloch states in the $\mathbf{k}_{\parallel}=0$ direction.^{9–11} Theoretically, a four-fold symmetry at the barrier/electrode interfaces is essential to produce the giant TMR effect. On the other hand, a giant MR ratio greater than 200% has been observed not only in fully epitaxial or textured MTJs (Refs. 6 and 7), but also in MTJs with a MgO(001) barrier sandwiched between amorphous CoFeB electrode layers.⁸ Reports have indicated that CoFeB electrodes might be crystallized locally near the interfaces because the MgO(001) layer could act as a template to crystallize CoFeB layers.^{8,12} The MR ratio has also been observed to be sensitive to the cap layer deposited on the CoFeB top electrode layer.¹³ The cap layer also seems to have an influence on the crystallization process of CoFeB. However, the precise crystallization process of CoFeB has not yet been experimentally studied.

Another puzzling issue is the growth process of the highly oriented polycrystalline MgO(001) barrier layer on the amorphous CoFeB bottom electrode layer. The growth

process is especially important in low-resistance CoFeB/MgO(001)/CoFeB MTJs for magnetic sensor application. The MgO layer thickness (t_{MgO}) should be less than 1 nm to achieve a resistance-area (RA) product less than a few $\Omega (\mu\text{m})^2$, which is required for the read heads of hard disk drives. However, the MR ratio is observed to drop sharply when the t_{MgO} is reduced to less than 1 nm.¹⁴ Although the drop in MR ratio is attributed to a deterioration in the crystalline orientation of the MgO barrier, the precise mechanism is not clear at the present stage.

As described above, the growth and crystallization process of CoFeB/MgO/CoFeB MTJs is of great importance for applications and for basic physics of the giant TMR effect. X-ray diffraction and cross-section transmission electron microscopy (TEM) analyses are usually used in the structural analysis of MTJs. These techniques, however, are not sensitive enough to determine the structure of an ultrathin MgO barrier (typically 0.8–2 nm) and CoFeB electrode layers (typically 3 nm). In addition, cross-section TEM observations may damage the sample during the preparation process. We used *in situ* reflective high-energy electron diffraction (RHEED) observations in this study to investigate the growth process of the MgO layer and the crystallization process of the CoFeB layer. We found that RHEED is a powerful tool to analyze the structure of ultrathin layers. The observed growth and crystallization process clearly explains the TMR effect in the CoFeB/MgO/CoFeB MTJs.

The CoFeB/MgO/CoFeB MTJs are usually prepared by sputtering deposition at RT, followed by annealing. The annealing temperature, at around 360 °C, is an optimum condition to obtain a giant MR ratio.⁸ In this study, however, we used the sputtering method for the growth of CoFeB and the molecular-beam epitaxy (MBE) method for the growth of MgO. This is because *in situ* RHEED observations cannot be performed in a magnetron-sputtering chamber. It should also be noted that we performed *in situ* annealing under an ultra-high vacuum (UHV) to observe the crystallization of CoFeB

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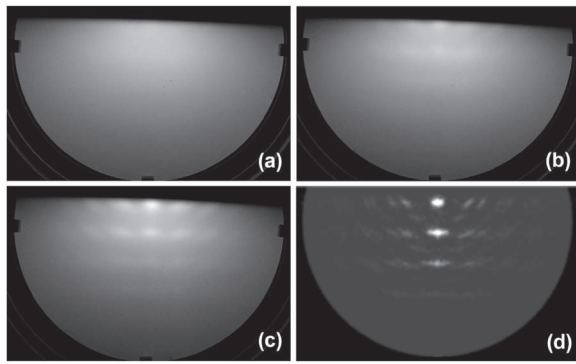


FIG. 1. (a)–(c) RHEED images of MgO layer grown on amorphous $\text{Co}_{60}\text{Fe}_{20}\text{B}_{20}$ layer. MgO layer thickness (t_{MgO}) are (a) 3 ML, (b) 5 ML, (c) 10 ML, and (d) simulated RHEED image for highly oriented polycrystalline MgO(001) layer with NaCl-type structure. Simulated coherent length and c -axis orientation distribution were assumed to be 15 Å and 10° .

using RHEED. MTJs are usually annealed *ex situ* after the deposition of an entire MTJ structure.⁸ Therefore, the sample preparation conditions in this study were not the same as those for conventional MTJs. Despite these differences due to experimental restrictions, the present study yielded valuable information for the fabrication of MTJs because their basic metallurgical nature is independent of the preparation methods. The MTJs prepared by sputtering and MBE methods showed quite similar MR ratios and RA product when the preparation conditions were optimized. The $\text{Co}_{60}\text{Fe}_{20}\text{B}_{20}$ electrode layers were deposited using radio-frequency magnetron sputtering with an Ar gas pressure of 2×10^{-3} Pa. The MgO barrier layers were deposited using an electron-beam evaporation of stoichiometric MgO source material under a UHV.⁶ The thickness of the MgO layer was measured using a quartz monitor. The samples were transferred between the sputtering and MBE chambers without breaking the UHV conditions (below 1×10^{-7} Pa). The RHEED observations and *in situ* annealing were performed in the MBE chamber with a base pressure of 2×10^{-8} Pa. The RHEED was observed with a 20 kV electron beam at an incident angle of about 2° .

The RHEED observations revealed that the CoFeB layer deposited using sputtering at RT always produced an amorphous structure regardless of its thickness and underlying layer. The RHEED images of the MgO layer grown on the amorphous CoFeB layer at RT are shown in Fig. 1. When $t_{\text{MgO}} \leq 3$ monoatomic layers (ML; 1 ML=0.21 nm), the MgO layer is amorphous, as shown in Fig. 1(a). The image has no diffraction spots, streaks, and ring patterns. At $t_{\text{MgO}} = 4$ ML, a very faint diffraction pattern appeared. The RHEED image for $t_{\text{MgO}} = 4$ ML is not shown because the diffraction pattern was too faint to be identified in a printed image. When $t_{\text{MgO}} \geq 5$ ML, a clear diffraction pattern appeared as shown in Figs. 1(b) and 1(c). From these results, we concluded that the MgO layer begins to crystallize when $t_{\text{MgO}} \geq 5$ ML. The faint diffraction pattern at $t_{\text{MgO}} = 4$ ML probably originated from a spatial distribution of t_{MgO} in the sample. Note that the diffraction patterns were independent of the in-plane angle of incidence, indicating that the MgO layer had no in-plane crystalline orientation. Figure 1(d) is a simulated diffraction pattern for MgO(001) textured layer with a NaCl-type structure. The RHEED pattern was simulated using a simple kinematics theory, in which the scattering factors were assumed to be proportional to the square of

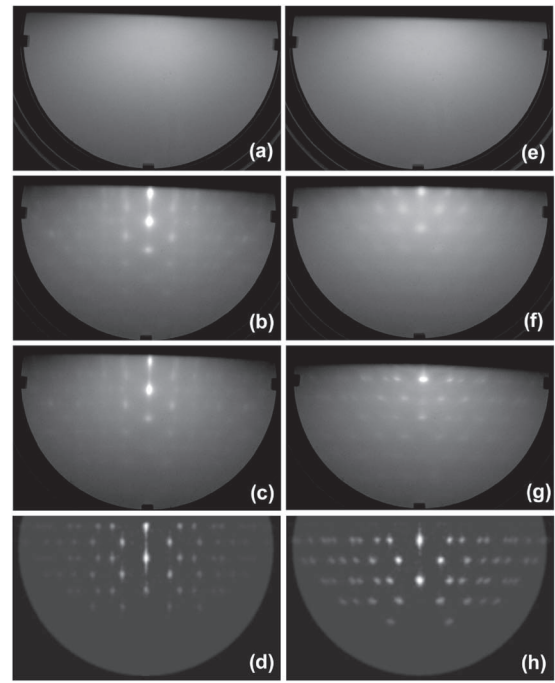


FIG. 2. (a)–(c) RHEED images of $\text{Co}_{60}\text{Fe}_{20}\text{B}_{20}$ layer (3 nm) grown on amorphous SiO_2 ; (a) as-grown state, (b) annealed at 360°C for 30 min, (c) annealed at 430°C for 30 min, and (d) simulated RHEED image for fcc CoFeB(001) textured layer. Simulated coherent length and c -axis orientation distribution were assumed to be 40 Å and 0° . (e)–(g) RHEED images of $\text{Co}_{60}\text{Fe}_{20}\text{B}_{20}$ layer (3 nm) grown on MgO(001) textured layer (10 ML); (e) as-grown state, (f) annealed at 360°C for 30 min, (g) annealed at 430°C for 30 min, and (h) simulated RHEED image for bcc CoFeB(001) textured layer. Simulated coherent length and c -axis orientation distribution were assumed to be 30 Å and 2° .

the atomic numbers. Coherent length and crystal orientation distribution were the fitting parameters. Note that positions of the diffraction spots are independent of these adjustable parameters. The comparison between the observed and simulated images clearly indicates that the MgO layer grown on an amorphous CoFeB had a highly oriented polycrystalline MgO(001) structure when $t_{\text{MgO}} \geq 5$ ML. Therefore, the MR ratio is expected to decrease significantly when $t_{\text{MgO}} < 5$ ML (1.05 nm) because the crystalline MgO(001) barrier is essential to produce the giant TMR effect. Thus, the observed sharp drop in the MR ratio for $t_{\text{MgO}} < 1$ nm (Ref. 14) can be attributed to the formation of an amorphous MgO barrier layer rather than the deterioration of the crystalline orientation.

The following is a discussion on the crystallization of the CoFeB layer. A RHEED image of CoFeB (3 nm) deposited on an amorphous SiO_2 at RT is shown in Fig. 2(a). The CoFeB layer was amorphous in the as-grown state. When the CoFeB was annealed, clear diffraction patterns appeared, as shown in Figs. 2(b) and 2(c), indicating crystallization of CoFeB. The observed diffraction pattern shows good agreement with the simulated pattern for the polycrystalline face-centered-cubic (fcc) (001) textured layer [Fig. 2(d)]. The origin of the (001) orientation is not clear at this stage. The fact that CoFeB crystallizes in a fcc structure without any template layer means that fcc is a stable crystal structure. Note that this is not a rigorous metallurgical discussion. The stable structure simply means that fcc is more stable than other structures that were obtained in this temperature range.

Figure 2(e) is the RHEED image of an amorphous CoFeB (3 nm) grown on a highly oriented polycrystalline

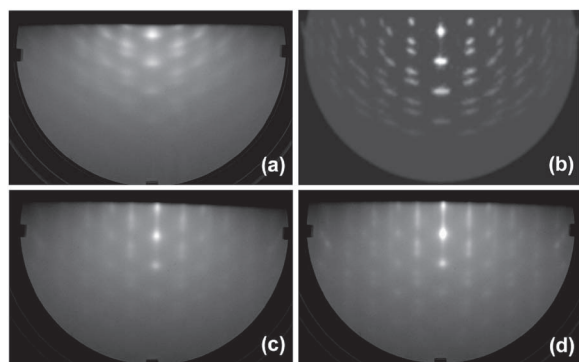


FIG. 3. (a) RHEED image of $\text{Ni}_{0.8}\text{Fe}_{0.2}$ layer (20 nm) grown on an amorphous $\text{Co}_{60}\text{Fe}_{20}\text{B}_{20}$ layer. (b) Simulated RHEED image for fcc NiFe(111) textured layer. Simulated coherent length and (111)-axis orientation distribution were assumed to be 40 Å and 5°. (c) and (d) RHEED images of $\text{Co}_{60}\text{Fe}_{20}\text{B}_{20}$ layer (3 nm) grown on fcc NiFe (111) textured layer; (c) annealed at 360 °C for 30 min, (d) annealed at 430 °C for 30 min.

MgO(001) layer (10 ML). When the CoFeB layer was annealed at 360 °C, which is an optimum annealing temperature for the CoFeB/MgO/CoFeB MTJs,⁸ broad diffraction spots appeared as indicated in the RHEED image [Fig. 2(f)]. This indicates a nanocrystallization or a crystallization with distortions. When the CoFeB layer was annealed at 430 °C, sharper diffraction spots appeared as indicated in the RHEED image [Fig. 2(g)]. The diffraction patterns in Figs. 2(f) and 2(g) are basically the same except for the broadness of diffraction spots, indicating the same crystal structure and orientation. The observed diffraction pattern shows good agreement with the simulated pattern for a body-centered-cubic (bcc) structure with (001) orientation [Fig. 2(h)]. Although a fcc structure is more stable than a bcc structure, CoFeB crystallizes in a bcc(001)-textured structure when grown on MgO(001). This clearly indicates that the MgO(001) layer acted as a template to crystallize the amorphous CoFeB layer because of a good lattice match between the MgO(001) and bcc CoFeB(001) layers. These results suggest that the CoFeB/MgO/CoFeB MTJs annealed at 360 °C should have a bcc CoFeB(001)/NaCl-type MgO(001)/bcc CoFeB(001) structure on a microscopic scale. Therefore, the observed giant TMR effect in CoFeB/MgO/CoFeB MTJs can be explained within the framework of the first-principle theories for epitaxial MTJs, in which the coherent spin-polarized tunneling of Δ_1 electrons plays an important role.^{9–11}

We also investigated the crystallization process of CoFeB when it faces a permalloy ($\text{Ni}_{0.8}\text{Fe}_{0.2}$) layer. A NiFe cap layer on a CoFeB top electrode layer (a free layer) is important for applications because this bilayered structure is effective in achieving soft magnetic properties, such as a high permeability and a low coercive field. The bilayered NiFe/CoFeB electrode, however, resulted in a considerable reduction of the MR ratio of less than 50%.¹³ We first deposited a NiFe layer (20 nm) on an amorphous CoFeB layer to characterize the structure of a NiFe layer. A RHEED image of the NiFe layer is shown in Fig. 3(a). The observed image shows good agreement with the simulated diffraction pattern for a fcc NiFe(111) textured layer [Fig. 3(b)]. When CoFeB (3 nm) was deposited onto this fcc NiFe(111) layer at RT, the CoFeB had an amorphous structure. When the CoFeB layer was annealed, the layer crystallized as shown in Figs. 3(c) and 3(d). The CoFeB layer crystallized in the fcc structure

with (111) orientation. This suggests that the CoFeB electrode would have a fcc(111)-textured structure if the crystallization began from the interface with the NiFe layer. The RHEED observations actually showed that the crystallization of CoFeB on NiFe(111) began at around 200 °C, while the crystallization of CoFeB on MgO(001) began at around 250 °C. Therefore, the NiFe cap layer is considered to have crystallized the CoFeB electrode in the fcc(111) structure, resulting in a reduction in the MR ratio.

In conclusion, we performed *in situ* RHEED observations to investigate the growth and crystallization processes of CoFeB/MgO/CoFeB MTJ structures. The MgO layer grown on an amorphous CoFeB layer had an amorphous structure for $t_{\text{MgO}} < 5$ ML and crystallized with a (001) preferred orientation for $t_{\text{MgO}} \geq 5$ ML. This growth process is consistent with the observed t_{MgO} dependence of the MR ratio. When a CoFeB layer is grown at RT, it has an amorphous structure regardless of its underlying layer. By *in situ* annealing, we found that the CoFeB layer grown on MgO(001) crystallized in a bcc structure with (001) preferred orientation, although a fcc structure is more stable than a bcc structure. This indicates that the MgO(001) layer acted as a template to crystallize the CoFeB layer because of a good lattice match between the MgO(001) and bcc CoFeB(001). The results suggest that the giant TMR effect observed in the CoFeB/MgO/CoFeB MTJs can be explained within the framework of the first-principle theories for epitaxial MTJs. When a CoFeB layer faces an fcc NiFe(111) layer, the CoFeB crystallizes in a fcc(111) structure. This explains the reduced MR ratio as result of the NiFe cap layer on the CoFeB electrode.

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