Interfacial disorder and magnetic frustration in epitaxial Fe/Cr/Fe trilayers studied by thermoremanent magnetization

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Abstract. In order to investigate the relaxation phenomena in Fe/Cr(001) system with various degrees of interfacial magnetic frustration, we have epitaxially grown Fe/Cr/Fe(001) trilayers on MgO(001) substrate under several thermal conditions by using molecular beam epitaxy (MBE) technique, and measured the time dependence of the thermoremanent magnetization (TRM) in the absence of magnetic field after field cooling process from 300 K to a measuring temperature 250 K under a magnetic field of 100 Oe after a waiting time \( t_w = 0 \sim 100 \text{ min} \). The reflection high energy electron diffraction (RHEED) was employed for confirming the epitaxial growth and for evaluating degree of the interfacial flatness in each sample. From the experimental results, we found that 1) all the present epitaxially grown Fe/Cr/Fe(001) trilayers are ferromagnetic at 300 K, 2) the TRM shows the logarithmic time dependence with larger magnetic viscosity coefficient for the samples which have more disordered interface relative to the other samples, and 3) the most disordered sample shows clearly \( t_w \) dependence as observed in spin glasses. These results indicate that the interfacial magnetic frustration in the epitaxial Fe/Cr(001) system increases with increasing the interfacial disorder.

1. Introduction

Fe/Cr multilayer is a well-known system to exhibit the giant magnetoresistance (GMR) [1] and the oscillatory interlayer exchange coupling through Cr spacer [2]. One of recent subjects which have attracted much attention is the interfacial magnetic frustration. It has been reported in the measurements of magneto-optical Kerr effect for epitaxial Fe/Cr [3] and Mössbauer spectroscopy for epitaxial Fe/Cr/Sn/Cr [4] that the interfacial frustration reduces the magnetic moments of Cr. The model proposed in ref. [4] is schematically shown in figure 1 for the spin arrangement near the epitaxial Fe/Cr(001) interface. Exchange interaction is ferromagnetic for Fe-Fe, and antiferromagnetic for Fe-Cr and Cr-Cr spin pairs. For the ideally flat interface, there is no frustration because all the spin pairs satisfy all the interactions (a). When there are Fe steps in Cr layer in the Fe/Cr interface, both directions of Fe spin in the step cannot satisfy all the interactions as shown in figures 1(b) and 1(c), causing the interfacial frustration. The interfacial frustration also occurs in the same way when there are Cr steps in Fe layer. The interfacial frustration in this model, therefore, increases with increasing the interfacial
Figure 1. Schematic model of the spin arrangement near the epitaxial Fe/Cr(001) interface. The arrows indicate the Fe and Cr spins. Exchange interaction between the nearest neighbor spins is ferromagnetic for Fe-Fe and antiferromagnetic for Fe-Cr and Cr-Cr, respectively. In the ideally flat interface, there is no frustration (a). The spin at the step site can not satisfy all the interactions for both spin directions (b) and (c).

disorder (atomic step). It is reasonable that such an interfacial frustration affects the magnetic relaxation mechanism and more or less causes magnetic viscosity as observed in spin glasses [5]. The direct observation of the slow dynamics is possible by measuring time dependence of the thermoremanent magnetization (TRM). As far as we know, time decay of the TRM has been measured only in a polycrystalline Fe/Cr system [6] or an epitaxial Fe/Cr with considerable interdiffusion [7] in which dominant origin of the frustration is not the interfacial disorder itself but that including substantial internal disorder. In fact, the magnetic transitions in such systems occur well below room temperature due to the bulk-like disorder [6, 7]. In this study, in order to investigate the relaxation phenomena of Fe/Cr(001) system in which the interfacial disorder is essential, we prepared the epitaxial Fe/Cr/Fe(001) trilayers with various degrees of the interfacial flatness by using the molecular beam epitaxy (MBE) technique and measured the time dependence of the TRM.

2. Experimental

Fe/Cr/Fe(001) trilayers were prepared by using MBE technique. The residual pressure in the MBE chamber was in the order of $10^{-9}$ Torr. MgO(001) single-crystal substrate was used to obtain the epitaxial Fe and Cr layers with (001) orientation [8]. The sample layer structure was MgO(001)/Cr(50 Å)/Fe(20 Å)/Cr(18 Å)/Fe(20 Å)/Au(10 Å). Cr(50 Å) was evaporated as a buffer layer prior to the Fe/Cr/Fe trilayer evaporation. Au(10 Å) was capped to prevent the oxidation. The thickness of the Cr spacer, 18 Å, was selected so that in our samples the first and second Fe layers are coupled ferromagnetically [2] to ensure sufficiently large magnetization for the relaxation measurement even in a small applied field. In order to obtain the samples with several degrees of the interfacial flatness, heat treatment condition was varied as below: 1) the growth temperatures of $T_s = 300$ °C and 400 °C were selected based on the fact that the optimal temperature to obtain the flat Fe(001) surface on the MgO(001) substrate is about 300 °C [9], 2) after evaporating each layer, the sample was annealed at 500 °C for 15 min, and 3) the samples without annealing process for each $T_s$ were also prepared. The reflection high
energy electron diffraction (RHEED) was employed for confirming the epitaxial growth and for evaluating of the interfacial flatness in each sample.

Magnetic properties were measured by using a SQUID magnetometer (Quantum Design MPMS). The procedure of the relaxation measurement of the TRM was as follows: first, a sample was rapidly cooled (-10 K/min) in the field $H_{FC}$ (= 100 Oe) from room temperature to a measuring temperature $T_m$ (= 250 K), and after a waiting time $t_w$ (= 0, 5, 15, and 100 min), the measurement was started in the absence of field and continued for about 10,000 sec.

3. Results and discussion

3.1. Interfacial structure observed by the RHEED

From in situ RHEED observations, we confirmed that all the layers in Cr/Fe/Cr/Fe/Au grew epitaxially with the (001) basal plane. Figure 2 shows the RHEED patterns at the surface of the first Fe layer in the sample of $T_s = 300 \, ^\circ C$ before annealing (a) and after annealing (b), respectively. The so-called $c(2 \times 2)$ reconstructed structure was observed as shown in figure 2 (a). It has been reported, in the study of the flat surface structure for Fe(001) epitaxially grown on MgO(001) substrates, that the $c(2 \times 2)$ reconstruction indicates two fold periodicity in the Fe[110] direction of the arrangement of the topmost Fe atoms and the surface with atomically flat and wide terraces separated by monoatomic steps [10, 11]. On the other hand, in the sample of $T_s = 400 \, ^\circ C$, no reconstructed structure was observed before annealing as shown in the figure 3 (a). By annealing at 500 \, ^\circ C for 15 min, $c(3\sqrt{2} \times \sqrt{2})$ reconstructed structure with three fold periodicity in the Fe[110] direction emerged in both samples as shown in figure 2 (b) and figure 3 (b). Such a $c(3\sqrt{2} \times \sqrt{2})$ reconstructed structure was also observed for Cr layer surface after annealing as shown in figure 2 (c). It has been reported that $c(3\sqrt{2} \times \sqrt{2})$ reconstruction is formed by C atoms segregated from the MgO(001) substrate [12]. Although it remains controversial what kind of element forms the reconstructed structure, an atomically flat surface at least equivalent to the $c(2 \times 2)$ case is needed to induce the $c(3\sqrt{2} \times \sqrt{2})$ reconstructed structure.

**Figure 2.** (Color online) RHEED patterns for the sample of $T_s = 300 \, ^\circ C$ at the surface of the first Fe (a), (b) and Cr (c) layers obtained in the Fe[110] and Cr[110] directions. Arrows in pattern (a) indicate the $c(2 \times 2)$ reconstructed structure, and arrows in pattern (b) and (c) indicate the $c(3\sqrt{2} \times \sqrt{2})$ reconstructed one. By annealing at 500 \, ^\circ C for 15 min, the RHEED pattern changes from (a) into (b). The lower panels show the Miller’s indices of the RHEED patterns. The dotted lines indicate the Miller’s indices for $c(2 \times 2)$ and $c(3\sqrt{2} \times \sqrt{2})$ reconstructed surfaces.
Figure 3. (Color online) RHEED patterns for the sample of $T_s = 400 \, ^\circ\text{C}$ at the surface of the first Fe layer obtained in the Fe[110] direction. Pattern (a) shows the $(1 \times 1)$ unreconstructed surface and the arrows in (b) indicate the $c(3\sqrt{2} \times \sqrt{2})$ reconstructed surface. By annealing at $500 \, ^\circ\text{C}$ for 15 min, the RHEED pattern changes from (a) into (b). The Miller’s indices of streak lines in (a) is the same as those in figure 2(a) excluding the reconstructed pattern (dotted line), and those in (b) is the same as in figure 2(b).

Table 1. Reconstructed structure observed at the surface of the first Fe layer in each sample and relative order of the interfacial flatness (larger number indicates relatively more disordered interface).

| Sample          | Reconstructed structure (before annealing) | Reconstructed structure (after annealing) | Relative order of the interfacial flatness (sample number) |
|-----------------|-------------------------------------------|------------------------------------------|----------------------------------------------------------|
| $T_s = 300 \, ^\circ\text{C}$ (with annealing) | $c(2 \times 2)$                         | $c(3\sqrt{2} \times \sqrt{2})$           | 1                                                        |
| $T_s = 300 \, ^\circ\text{C}$ (without annealing) | $c(2 \times 2)$                         | -                                        | 1’                                                       |
| $T_s = 400 \, ^\circ\text{C}$ (with annealing) | unreconstructed                         | $c(3\sqrt{2} \times \sqrt{2})$           | 2                                                        |
| $T_s = 400 \, ^\circ\text{C}$ (without annealing) | unreconstructed                         | -                                        | 3                                                        |

The reconstructed structures observed at the first Fe layer in each sample are summarized in the table 1. For estimating the interfacial flatness in each sample, we assumed that the reconstructed surface is more flat than the unreconstructed surface. Based on the reconstruction, relative order of the interfacial flatness is determined as shown in the rightmost column of the table 1. Here, the larger number indicates relatively more disordered interface. We note that the number 1 and 1’ means nearly the same order of the interfacial flatness as described above. We have also performed the RHEED observations at the other surfaces, but the order of the interfacial flatness is not changed. Hence, each sample is labeled as the number.

3.2. Relaxation of the TRM
We measured magnetization curves for all samples from -50 kOe to +50 kOe, and observed magnetic hysteresis with coercive forces (about 1 Oe for the samples of $T_s = 300 \, ^\circ\text{C}$ and $30 \sim 100 \, \text{Oe for } T_s = 400 \, ^\circ\text{C}$) at 300 K for all samples, which indicates that our epitaxially grown Fe/Cr/Fe trilayers are ferromagnetic at least at 300 K. The result suggests that the internal disorder due to the interdiffusion can be almost disregarded in our samples because the transition temperature becomes lower far below room temperature when such a bulk-like
frustration occurs [6, 7].

Figure 4 shows time dependence of the TRM in each sample in the linear scale (a) and the logarithmic scale (b). Measurement conditions were $H_{\text{FC}} = 100$ Oe, $T_m = 250$ K, and $t_w = 0$ min. We found that time decay of the TRM is dramatically changed depending on the interfacial flatness as shown in the figure 4 (a). In the samples of number 2 and 3, which have relatively disordered interfaces, time decay of the TRM is well fitted to the following logarithmic equation (1) as shown in the figure 4 (b),

$$M(t) = M(0) - S \ln t,$$

where $M(0)$ is the first measuring point after removing the $H_{\text{FC}}$, $t$ is the elapsed time after starting the measurement, and coefficient $S$ is the magnetic viscosity which indicates the slope of the logarithmic decay. For the samples of number 1 and 1’, which have relatively flat interfaces, it is considered that the relaxation is very rapid, then, it has been already finished before measuring the first point. Logarithmic decay observed in the samples 2 and 3 can be attributed to the effect of the interfacial frustration. In addition, it is found that $S/M_s$ increases with increasing the interfacial disorder, where $S$ is divided by $M_s$ (saturated magnetization obtained from the M-H curve at 300 K) to compare the relative value in each sample. The values of $S/M_s$ for the samples 2 and 3 are $1.09 \times 10^{-3}$ and $3.39 \times 10^{-3}$, respectively. These results support consistently the schematic models demonstrated in the figure 1.

Figures 5 (a) and (b) show $t_w$ dependences of the $S/M_s$ in the samples 2 and 3, respectively. In the samples 2 and 3, particularly in the sample 3, $S/M_s$ decreases substantially with increasing $t_w$ as shown in the figures 5 (a) and (b). This $t_w$ dependence has been generally observed in spin glasses [13]. These results suggest that dynamical behavior of the epitaxial Fe/Cr/Fe(001) trilayers with the interfacial magnetic frustration is close to that of the spin glasses. But, we emphasize that Curie temperature in the present Fe/Cr/Fe(001) system is above 300 K, that is, the epitaxially grown system is ferromagnetic reflecting dominantly to the interfacial disorder, therefore, to the interfacial magnetic frustration. Further experiments are needed to discuss the detailed relaxation mechanism.
4. Conclusion
We prepared epitaxial Fe/Cr/Fe(001) trilayers with different degrees of the interfacial flatness and measured the relaxation of the TRM in each sample. The relative order of the interfacial flatness was determined from the reconstructed structure observed in the RHEED. We found that in the samples with disordered interfaces TRM shows logarithmic time dependence and magnetic viscosity coefficient $S$ increases with increasing the interfacial disorder. In addition, the most disordered sample shows clear $t_w$ dependence as observed in spin glasses. These results definitely indicate that the interfacial magnetic frustration increases with increasing the interfacial disorder in the epitaxial Fe/Cr(001) system. The effect of ferromagnetic interlayer exchange coupling (IEC) of Fe layers on the interfacial frustration has not yet been clarified. However, our preliminary investigation for a simple Fe/Cr bilayer system without IEC shows that the logarithmic TRM decay is also observed in the bilayer system as in the present Fe/Cr/Fe one, which indicates that the ferromagnetic IEC is not essential at least in the present sample structure. Recently, we have also found the logarithmic time dependence of the TRM in the epitaxial Fe/Cr/Fe(011) trilayers. But, in contrast to Fe/Cr/Fe(001) case, $S$ decreases with increasing the interfacial disorder suggesting the decrease of the interfacial magnetic frustration. We will report elsewhere about the Fe/Cr/Fe(011) trilayers as second article.

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