Local magnetic study for cluster-layered Fe/Cr nanostructures

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Abstract. Local magnetic structures of cluster-layered Fe/Cr thin films are studied by X-ray magnetic circular dichroism (XMCD) and extended X-ray absorption fine structure (EXAFS). The magnetic moment of Fe atoms evaluated from XMCD for Al₂O₃/Cr(70 Å)/[Fe(1.2 Å)/Cr(10.5 Å)]₃₀/Cr(12 Å), which shows the minimum point in the resistivity, decreases as the temperature increases and disappears around the temperature of the minimum resistivity. EXAFS shows the local structure of this film around Fe atoms is rather disordered and it can be related the fluctuation of magnetic structure estimated from the theoretical analysis of the temperature dependence of magnetic moments.

1. Introduction

One of the great characteristic feature of Fe/Cr multilayered structure is giant magnetoresistance (GMR) discovered in 1988 [1,2]. On the other hand, the correlation between magnetic and transport properties is actively studied in such a multilayer structures and granular thin film with ferromagnetic nanoparticles [3-5]. Recently Kondo-like behavior, in which the minimum point is found in temperature dependence of the resistivity, has been observed in cluster-layered Fe/Cr nanostructures in 2006 [6]. In this ultrathin film, the small amount of Fe shows superparamagnetic feature but no GMR behavior.

In the MgO based thin-films; MgO/Cr(80 Å)/[Fe(tFe, Å)/Cr(10 Å)]₃₀; (tFe is thickness of Fe layer), Fe atoms are not continuous but exist as cluster in the range of tFe < 5 Å, the system shows superparamagnetic behavior for tFe < 2 Å, and the resistivity shows minimum point (Kondo-like behavior) for tFe = 0.3-1.2 Å [6]. To reveal the origin and mechanism of this interesting behavior in the system of such Fe multilayer films, the investigation from the view point of the local magnetic and atomic structure is important.

In this paper, we investigate X-ray magnetic circular dichroism (XMCD) and extended X-ray absorption fine structure (EXAFS) to study the local magnetic structures and the mechanism of the characteristic magnetic feature of the Al₂O₃ based cluster-layered Fe/Cr nanostructures.

2. Experimental

The samples discussed in this paper are Al₂O₃ based cluster-layered Fe/Cr films as; Al₂O₃/Cr(100 Å)/[Fe(8 Å)/Cr(10.5 Å)]₃₀/Cr(20 Å); (G1), Al₂O₃/Cr(70 Å)/[Fe(1.2 Å)/Cr(10.5 Å)]₃₀/Cr(12 Å); (K1) and Al₂O₃/Cr(63 Å)/Fe₀.17Cr₀.83 (24 Å) (A1) which are grown by molecular beam epitaxy (MBE) [6]. G1 sample shows GMR feature and K1 sample shows Kondo-like behavior in which the minimum point is appear in the resistivity at ~170 K.

XMCD spectra were measured at BL7A in KEK-PF, Japan. The applied magnetic field was 1.0 T and circular polarization of incident X-ray was 0.7. Fe L₁II and L₁III edge XMCD were measured by fluorescence mode using SDD (silicon drift detector). Angular dependent (θ = 30°, 45°, 55°, 70°, where
\( \theta \) is the angle between normal direction of the sample film and X-ray beam direction. XMCD spectra were measured at temperature range of 77 K–300 K. Fe K-edge EXAFS were also measured at BL9C in KEK-PF, Japan in Fluorescence mode using SSD (Ge-solid state detector) and Lytle detector at 25–300 K and \( \theta = 45^\circ \).

3. Results and discussion

The G1 sample which shows GMR behavior presents ferromagnetic feature through the present temperature ranges (77–300K) from Fe L-edge XMCD, in which XMCD intensity does not depend on temperature. A1 sample does not show any XMCD signal in this temperature range (77–300K). On the other hand, K1 sample shows interesting magnetic behavior in XMCD spectra. Figure 1 shows the typical X-ray Absorption Spectra (XAS) of Fe L_{II} and L_{III} edge for K1 sample measured at 158K with different circular polarizations. Figure 2 shows the Fe L-edge XMCD spectra for K1 sample \( \theta = 70^\circ \). Clear XMCD peak was found at 77 K but the intensity decreases as temperature increases (170K and 223K). For another angle of \( \theta = 30^\circ, 45^\circ, 55^\circ \), the similarly clear XMCD were observed.

Using the typical sum rule for XMCD [7], the contribution from spin and orbital magnetic moments were derived from the intensity of XMCD spectra individually. In this paper we discuss the total magnetic moment (spin + orbital moment). Figure 3 shows temperature dependence of the total magnetic moment of K1 sample measured at \( \theta = 30^\circ \) and \( 70^\circ \). The magnetic moment is decreases as temperature increases and the XMCD peaks disappear at the temperature more than ~200 K.

We analyze the temperature dependence of the magnetic moments obtained from XMCD by mean field theory. The function for the fitting is

\[
m_{\chi}(T; T_c, \Delta) = m_{\chi} \frac{\mu}{T_c} \int_{T_c}^{T} L(T' - T_c, \Delta) \left[ T_c^2 - T_c^2 \right] dT_c
\]

(1)

where the convolution function \( L(T, \Delta) \) is expressed as

\[
L(T, \Delta) = \frac{\Delta / (2\pi)}{T^2 + (\Delta / 2)^2}
\]

(2)

![Figure 1](image_url). Fe L_{II} and L_{III} edge XAS spectra for K1 sample at 158K. \( \theta=70^\circ \).
Figure 2 Fe $L_{\text{II}}$- and $L_{\text{III}}$-edge XMCD spectra for K1 sample at (a) 77K, (b) 170K, (c) 223K. $\theta=70^\circ$.

Figure 3 Temperature dependence of total magnetic moment derived from XMCD for (a) $\theta=30^\circ$ and (b) $\theta=70^\circ$.

We obtained a diffuse phase transition from the ferromagnetic to the paramagnetic state with the Curie temperature distributed in the interval $\Delta=56$ K around the average value $T_c=129$ K for several angles. Strong fluctuations occur for the system in this area and they may lead to an increase in electrical resistance.
It is important that the Curie temperature is reduced by several times with the thick films. This phenomenon was also observed in the two-dimensional magnetic trilayers with spin fluctuations [8].

The local structure around Fe atom for G1 and K1 sample were also studied in comparison with the alloy film of A1 by Fe K-edge EXAFS. Figure 4 shows (a) Fe K-edge XAFS $k^2\chi(k)$ and (b) Fourier transform for K1 sample at various temperature as an example. Same quality of EXAFS data was obtained for G1 and A1 samples. We analyzed the data at the range of $k=2$ to $12 \text{Å}^{-1}$ by use of Athena & Artemis [9].

Figure 5 shows the temperature dependence of (a) interatomic distance and (b) Debye-Waller factor (or mean square relative displacement) of 1st nearest neighbor (1NN) Fe-Fe. The interatomic distance of 1NN Fe-Fe for K1 sample is close to that for A1 sample, and longer than that for G1 at lower temperature. This indicates that the local structure around Fe of K1 sample is similar to that of A1 sample. Debye-Waller factor generally indicates the disorder or fluctuation of the atomic position from the X-ray absorbing atom.

The Debye-Waller factor of K1 sample is also similar to that of A1 sample and larger than that of G1 sample; this EXAFS result indicates that the local structure around Fe atoms for K1 sample is disordered like as alloy sample (A1).

Electron spin density fluctuations associated with the transition cause temperature anomalies of electrical resistivity and other transport properties due to carrier scattering on fluctuations.

4. Conclusion
Local magnetic and atomic structures of cluster-layered Fe/Cr thin films are studied by XMCD and EXAFS. The magnetic moment of K1 sample is around $0.8 \mu_B$ at 77K for K1 sample and decreases as temperature increases then disappear near the minimum point of electric resistivity. Analysis of the temperature dependence of the magnetic moment suggests that the strong spin fluctuations occur for the system and they may lead to an increase in electrical resistance. The EXAFS result shows that the local structure disorder of cluster-layered film is close to the FeCr alloy film. The connection between the magnetic fluctuation and the local structure disorder can be expected.

![Figure 4](image-url) **Figure 4** Fe K-edge EXAFS $k^2\chi(k)$ and Fourier transform for K1 sample at various temperature with Fe foil as standard sample.
Figure 5 Temperature dependence of (a) interatomic distance and (b) Debye-Waller factor of 1NN Fe-Fe. K1; blue square, G1; red circle, A1; green triangle.

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