Reduction of Magnetic Moments in Very Thin Cr Layers of Fe/Cr Multilayers: Evidence from ^{119}Sn Mössbauer Spectroscopy

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Fe/Cr multilayers with monatomic Sn layers embedded in the Cr layers were grown epitaxially on MgO(001) substrates, and the magnetic hyperfine field at the ^{119}Sn nuclear sites was examined using Mössbauer spectroscopy. It was found that nonzero hyperfine field is induced at the Sn sites at room temperature and that the value reduces drastically from 10 to 2 T when the Cr layer thickness decreases from 80 to 10 Å. The result indicates that the Cr layers are magnetically ordered even when the thickness is very small and that the magnetic moments of Cr become smaller as the Cr layer thickness decreases.

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Since the discovery of antiferromagnetic coupling between Fe layers through a Cr layer in Fe/Cr/Fe trilayers [1] and giant magnetoresistance in Fe/Cr multilayers [2], considerable attention has been paid to the magnetic properties of Cr in Fe/Cr multilayer systems. When the Cr layer thickness is around 10 Å, a strong antiferromagnetic coupling appears between Fe layers through the intervening Cr layer. However, it is still open to discussion whether the Cr layers are magnetically ordered or not in this thickness region. Since Cr metal is basically paramagnetic or antiferromagnetic [3], there are not many experimental methods to get effective information about the magnetic properties of Cr layers embedded between ferromagnetic Fe layers. Experimental works using perturbed angular correlation (PAC) [4], neutron diffraction [5,6], and x-ray magnetic circular dichroism (XMCD) measurement [7] have been performed to elucidate the magnetic structure of the Cr layers. The results from these experimental methods were, however, somewhat contradictory. For example, in the region where the Cr layer thickness is thinner than 42 Å, the PAC experiment indicated that the Cr is nonmagnetic, whereas the neutron diffraction measurements showed that the Cr has a commensurate antiferromagnetic structure. The XMCD result was explained with a model where only Cr atoms close to the Fe interface acquire a significant magnetic moment. In any case, it is difficult to elucidate magnetic properties of Cr layers sandwiched between ferromagnetic Fe layers, especially when the Cr layer thickness is very small. In the present work, ^{119}Sn Mössbauer spectroscopy was applied to obtain complementary information to the PAC and the neutron diffraction measurements. Since Sn is a nonmagnetic element, the ^{119}Sn Mössbauer nucleus can be an appropriate probe to study magnetic properties of Cr [8]. Monatomic Sn layers were embedded in the Cr layers of Fe/Cr multilayers, and the magnetic hyperfine field induced at the ^{119}Sn nuclear sites was measured using Mössbauer spectroscopy.

The Fe/Cr multilayers with a Sn layer embedded in each Cr layer (i.e., Fe/Cr/Sn/Cr multilayers) were prepared on MgO(001) substrates using an ultrahigh-vacuum deposition system with *e*-gun heating. A Cr(50 Å) buffer layer was deposited on the MgO(001) substrate prior to the deposition of a multilayer. Then, an [Fe(10 Å)/Cr(t_{Cr_1})/Sn(t_{Sn})/Cr(t_{Cr_2})] multilayer ($t_{\mathrm{Cr}} \equiv t_{\mathrm{Cr}_1} + t_{\mathrm{Cr}_2} \leq 80$ Å, $t_{\mathrm{Sn}} = 0.5$, 1, or 2 Å) with ^{119}Sn enriched (~85%) Sn layers was grown on the buffer layer. Here a thickness of about 1.7 Å for Sn corresponds to a monatomic layer. The period of Fe/Cr/Sn/Cr was repeated 29 or 39 times, then the deposition was ended with an additional Fe(10 Å) layer. The substrate temperature was kept at 200 °C during the deposition. The pressure during the film growth was in the 10^{-9} Torr range and the deposition rate was set around 0.3 Å/s, so that the contamination during the deposition is thought to be negligibly small. These growth conditions are the same as those for the previously reported epitaxial Cr/Sn multilayers [9]. The ^{119}Sn Mössbauer spectra were measured by means of conversion electron Mössbauer spectroscopy, using a gas-flow counter with He + 1%$(CH_3)_3CH$ at room temperature and a gas-filled counter with He + 2%CH_4 at 100 K and H_2 at 15 K [10]. A Ca$^{119m}SnO_3$ source was used to obtain γ rays of 23.8 keV and the direction of the incident γ rays was set parallel to the film normal. From reflection high energy electron diffraction and x-ray diffraction measurements, it was confirmed that the MgO, Cr, and Fe have the structural relation MgO(001)//Cr(001)//Fe(001) in the growth direction and MgO[100]//Cr[110]//Fe[110] in the film plane. The Sn atoms are thought to be located at the substitutional sites of bcc Cr(001) planes, and forming a somewhat strained bcc lattice together with the Cr atoms, as in the case of Cr/Sn multilayers [9]. The lattice spacing between the Cr(001) planes was estimated to be 1.44 ± 0.01 Å, and that between the Cr and Sn planes was 1.57 ± 0.04 Å. The in-plane (100) lattice parameter was somewhat larger than that for bulk Cr (i.e., 2.88 Å), ranging from 2.88 to 2.91 Å. The details of the structural study will be published elsewhere [11].

The use of ^{119}Sn Mössbauer spectroscopy for the study on magnetic properties of Cr layers has already been demonstrated for Cr/Sn multilayers in Ref. [9]. In Fig. 1(a), the ^{119}Sn Mössbauer spectrum for [Cr(20 Å)/Sn(2 Å)] multilayer at 300 K is shown as a typical spectrum for [Cr(t_{Cr})/Sn(2 Å)] multilayers. A magnetic splitting with quite a large hyperfine field is observed in the spectrum. The appearance of a large hyperfine field at room temperature indicates that the magnetic ordering temperature of the Cr layers is much higher than the Néel temperature of bulk Cr (i.e., 311 K). The spectrum was fitted with six-line components with a distribution of magnetic hyperfine field. The hyperfine field has a Gaussian-like distribution with the maximum around 12 T and no component at zero field [Fig. 1(b)]. The isomer shift (relative to that for $CaSnO_3$) was fitted to be 1.56 mm/s, which is a reasonable value when the Sn atoms are sandwiched with Cr atoms. The distribution of hyperfine field in Fig. 1(b) gives information about the magnetic structure of Cr around the Sn layer. Suppose the multilayer has an ideal crystallographic structure, i.e., a monatomic Sn layer is embedded in a stack of bcc Cr(001) planes. When the Cr layer has an antiferromagnetic structure (including the incommensurate spin-density-wave (SDW) antiferromagnetic structure with the **Q** vector perpendicular to the film plane), the magnetic moment of Cr is aligned parallel in a (001) plane and antiparallel between adjacent (001) planes. If the magnetic moments of atomic Cr(001) planes in both sides of the Sn layer were oriented to the opposite direction with each other, the magnetic hyperfine field transferred at the Sn site would be canceled to zero. Therefore, the observation of a finite size hyperfine field indicates that the magnetic moments of atomic Cr planes that are sandwiching the Sn layer are oriented to the same direction and that this magnetic structure is established through the Sn layer. The Gaussian-like distribution is probably due to the deviation of the crystallographic structure from the ideal layered structure. The Gaussian-like feature also proves that all the Cr atoms that contact the Sn layer basically have the same size of magnetic moment. The existence of SDW with the **Q** vector in the film plane, for example, would result in an Overhauser-type distribution of hyperfine field, as observed for dilutely dissolved Sn in a Cr matrix [8]. The value of hyperfine field is larger than that reported for dilute Sn in Cr. The magnetic hyperfine field at the Sn site mainly reflects the spin polarization of 5*s* conduction electrons at the nuclear site, whereas the magnetic moment of Cr is mainly caused by the spin polarization of 3*d* electrons in the atomic site. In the case of dilutely dissolved Sn in a Cr or Cr-rich alloy matrix, the size of the hyperfine field at the Sn site is thought to be proportional to that of the magnetic moment of surrounding Cr atoms [12]. For layered structures, the neighboring effect might be different from that for a single impurity site. According to a band calculation on Cr/Sn multilayers by Oguchi and Momida [13], the magnetic moments of Cr adjacent to the Sn layer are somewhat enhanced in comparison with those inside the Cr layer, and the hyperfine field at the Sn site is also enhanced proportionally to the magnetic moment of the contacting Cr atoms. Thus the observed big hyperfine field at the Sn sites appears to be connected with the enhancement of Cr magnetic moments at the interface.

The ^{119}Sn Mössbauer spectra for [Fe(10 Å)/Cr(t_{Cr_1})/Sn(2 Å)/Cr(t_{Cr_2})] multilayers ($t_{Cr_1} = t_{Cr_2} = 40$, 20, 10, and 5 Å), where Sn(2 Å) is embedded at the center of Cr(t_{Cr}) layer ($t_{Cr} = 80$, 40, 20, and 10 Å), measured at 300 K are shown in Fig. 2(a). A big change in the spectra is observed when t_{Cr} is varied between 80 and 10 Å. Each spectrum was fitted with six-line components with a distribution of magnetic hyperfine field. The obtained distribution is shown in Fig. 2(b). The hyperfine field at the peak in the distribution curve was estimated to be 10 T for [Fe(10 Å)/Cr(40 Å)/Sn(2 Å)/Cr(40 Å)] and 2 T for [Fe(10 Å)/Cr(5 Å)/Sn(2 Å)/Cr(5 Å)]. Thus, the hyperfine field drastically reduces as the Cr layer

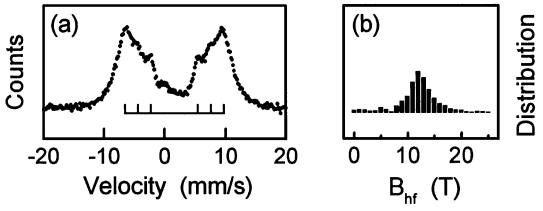

FIG. 1. (a) ^{119}Sn Mössbauer spectrum for [Cr(20 Å)/Sn(2 Å)] multilayer at 300 K. The peak positions for the magnetically split sextet are a guide to the eye. (b) The distribution of hyperfine field obtained from the fitting of the spectrum.

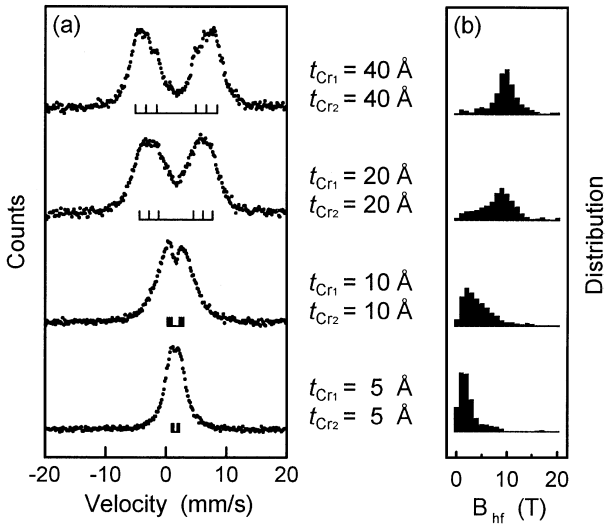

FIG. 2. (a) ^{119}Sn Mössbauer spectra for [Fe(10 Å)/Cr(t_{Cr_1})/Sn(2 Å)/Cr(t_{Cr_2})] multilayers ($t_{Cr_1} = t_{Cr_2} = 40$, 20, 10, and 5 Å) at 300 K. The peak positions for the magnetically split sextet are a guide to the eye. (b) The distribution of hyperfine field obtained from the fitting of the spectra.

thickness decreases. Since the Sn layer is sandwiched between Cr layers, it appears reasonable to conclude that (i) the decrease of the magnetic hyperfine field at the Sn sites reflects the reduction of the magnetic moments of Cr around the Sn layer, and that (ii) this change of magnetic moments occurs not only around the Sn layer but throughout the Cr layer. In the Cr/Sn multilayers, the hyperfine field at the Sn sites increases a little as the Cr layer thickness becomes smaller, i.e., around 10 T for [Cr(80 Å)/Sn(2 Å)] and 13 T for [Cr(10 Å)/Sn(2 Å)] [9]. The in-plane (100) lattice parameter is estimated to be 2.90 Å for [Fe(10 Å)/Cr(40 Å)/Sn(2 Å)/Cr(40 Å)] and 2.91 Å for [Fe(10 Å)/Cr(5 Å)/Sn(2 Å)/Cr(5 Å)], whereas that is 2.90 Å for [Cr(80 Å)/Sn(2 Å)] and 2.94 Å for [Cr(10 Å)/Sn(2 Å)]. The reduction of the magnetic hyperfine field is observed only for the Fe/Cr/Sn/Cr multilayers, although the strain is prominent for the Cr/Sn multilayers. Therefore, the strain would not be the main reason for the reduction of magnetic moments in the present case. In Fe/Au/Sn/Au multilayers, where Sn layers are inserted in nonmagnetic Au layers, the hyperfine field increases as the Au layer thickness decreases [14]. The conduction electrons in the Au layer are spin polarized by the ferromagnetic Fe layer, and the spin polarization gets smaller as the distance from the Fe interface becomes larger. In the Fe/Cr/Sn/Cr multilayers, the hyperfine field reduces as the Cr layer thickness decreases, so that such a polarization effect from the ferromagnetic layer is not the main origin of the observed change of hyperfine field.

The Mössbauer spectra for [Fe(10 Å)/Cr(5 Å)/Sn(2 Å)/Cr(5 Å)] multilayer were measured also at low temperatures. The hyperfine field at the peak in the distribution, which was obtained from the spectra, is plotted as a function of temperature in Fig. 3. The hyperfine field increases a little as the temperature decreases, but the value is merely around 3 T at 15 K. Therefore, the difference of the spectra at 300 K shown in Fig. 2(a) is not simply due to the change of the magnetic transition temperature of Cr, but due to the change of Cr magnetism in the temperature range from 0 K. From the extrapolation of the hyperfine field vs temperature curve to higher temperatures, it appears that the magnetic transition temperature of the Cr layers in [Fe(10 Å)/Cr(5 Å)/Sn(2 Å)/Cr(5 Å)] is over 400 K, which is much higher than the Néel temperature of bulk Cr. Since the hyperfine fields at room temperature is even larger for other samples, the magnetic ordering temperature of Cr is probably much higher than that of bulk Cr as well. Moreover, the change in the magnetic hyperfine field in Fig. 3 is larger than the temperature dependence of Fe layer magnetization. This fact also shows that the effect due to the conduction electron spin polarization induced by the Fe layer, as observed in Fe/Au/Sn/Au multilayers [14], is not dominant in Fe/Cr/Sn/Cr multilayers.

Figure 4 shows the change of the Mössbauer spectra as a function of the position (i.e., the depth from the Fe interface) of the Sn probe layer for [Fe(10 Å)/Cr(44 Å)] multilayers with Sn(1 Å) at 300 K. The size of the hyperfine field is not much dependent on the position of the Sn layer so long as the Cr layer thickness t_{Cr} is fixed. This result implies that the size of the magnetic moment of Cr is almost the same throughout the layer. This is reasonable because neutron diffraction measurements indicate the existence of a commensurate antiferromagnetic structure in the present thickness region at room temperature [5,6]. Note that the insertion of Sn would also act to stabilize the commensurate antiferromagnetic phase. The fact that the hyperfine field is not much dependent on the depth from the Fe interface again shows that the effect due to the conduction electron spin polarization induced by the Fe layer [14] is not dominant in the Fe/Cr/Sn/Cr multilayers. The direction of the magnetic hyperfine field (hence that of the magnetic moments of Cr) was also estimated from the intensity ratio of magnetically split six-lines in the spectra. It appears that the magnetic hyperfine field is almost in the film plane when the Sn layer is situated near the Fe/Cr

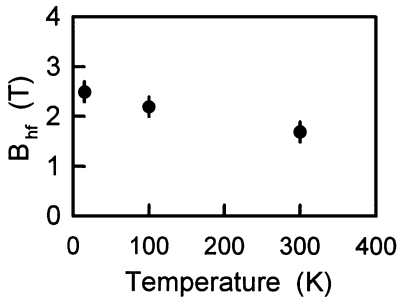

FIG. 3. Temperature dependence of the hyperfine field (at the maximum in distribution) for [Fe(10 Å)/Cr(5 Å)/Sn(2 Å)/Cr(5 Å)] multilayer obtained from the ^{119}Sn Mössbauer spectra.

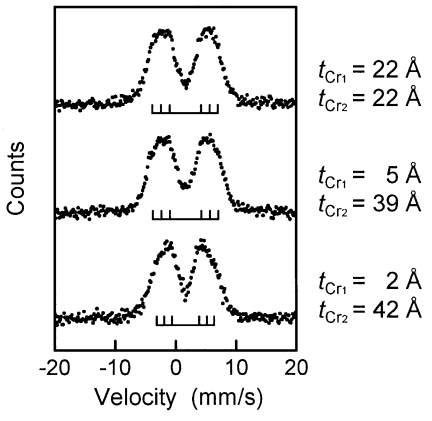

FIG. 4. ^{119}Sn Mössbauer spectra for [Fe(10 Å)/Cr(t_{Cr_1})/Sn(1 Å)/Cr(t_{Cr_2})] multilayers ($t_{Cr} = t_{Cr_1} + t_{Cr_2} = 44$ Å, $t_{Cr_1} = 22$, 5, and 2 Å) with the Sn layer in the different depth from the Fe/Cr interface at 300 K. The peak positions for the magnetically split sextet are a guide to the eye.

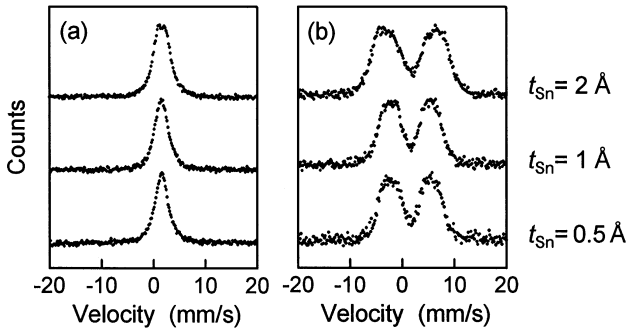

FIG. 5. ^{119}Sn Mössbauer spectra for (a) [Fe(10 Å)/Cr(5 Å)/Sn(t_{Sn})/Cr(5 Å)] multilayers (t_{Sn} = 2, 1, and 0.5 Å) and (b) [Fe(10 Å)/Cr(22 Å)/Sn(t_{Sn})/Cr(22 Å)] multilayers (t_{Sn} = 2, 1, and 0.5 Å) measured at 300 K.

interface, and it gradually turns toward the perpendicular direction as the position becomes closer to the center of the Cr layer. This tendency is surprisingly consistent with the result from the PAC experiment on Fe/Cr multilayers with thicker Cr layers [4] and the neutron diffraction measurements on Cr films with Fe cap layers [15], although the insertion of Sn would more or less influence the magnetism of the Cr layers.

The dependence of the Mössbauer spectra on the Sn layer thickness is shown for [Fe(10 Å)/Cr(5 Å)/Sn(t_{Sn})/Cr(5 Å)] and [Fe(10 Å)/Cr(22 Å)/Sn(t_{Sn})/Cr(22 Å)] multilayers in Figs. 5(a) and 5(b). The hyperfine field tends to decrease a little as t_{Sn} is reduced from 2 to 1 Å and stay constant from 1 to 0.5 Å. A big difference in the size of hyperfine field between the samples with different Cr layer thickness remains even when t_{Sn} is reduced to 0.5 Å. The spectra for [Fe(10 Å)/Cr(5 Å)/Sn(1 Å)/Cr(5 Å)] and [Fe(10 Å)/Cr(5 Å)/Sn(0.5 Å)/Cr(5 Å)] look like a single-line pattern, but the linewidth (2.9 mm/s) is larger than that for a paramagnetic peak ($\sim$1 mm/s). Therefore, the Cr layers in these samples are also regarded as magnetic at room temperature. For [Fe(10 Å)/Cr(5 Å)/Sn(t_{Sn})/Cr(5 Å)] multilayers, where a strong antiferromagnetic coupling between Fe layers and a large magnetoresistance effect are expected to appear when the Sn layer thickness is zero, the magnetoresistance ratio decreases as t_{Sn} increases from 0 to 2 Å; this is because the antiferromagnetic coupling is weakened and spin-independent electron scattering is increased by the existence of Sn layers. The basic features of Mössbauer spectra, on the other hand, do not change much when t_{Sn} decreases from 2 to 0.5 Å. (Note, in comparison, the big dependence on the Cr layer thickness.) Therefore, the change in the size of the magnetic moments of Cr as a function of the Cr layer thickness is thought to be a common and essential feature for both Fe/Cr/Sn/Cr and Fe/Cr multilayer systems.

The reason why the Cr magnetic moments reduce as a function of Cr layer thickness is thought to be either an intrinsic (thin film) band effect or a frustration effect. A band calculation of epitaxial Fe/Cr/Sn/Cr multilayers with an ideal layered structure is now in progress [13]. The magnetic frustration effect accompanied by steps at the Fe/Cr interface would reduce the size of Cr magnetic moments [16] and hence the hyperfine field at the Sn nuclear sites. This frustration effect is thought to become dominant as the Cr layer thickness becomes smaller (i.e., a frustration and size effect). Further study is required to conclude which effect is more dominant for the reduction of the magnetic moments. The reduction of magnetic moment would make the neutron diffraction signal weaker and the PAC signal more or less nonmagnetic. Thus, the Mössbauer results link the contradictory results from the PAC and the neutron diffraction measurements on Fe/Cr multilayers. Although the Mössbauer spectroscopy requires a large amount of probe atoms, it gives effective information about the local magnetism of Cr especially for the samples with thin Cr layer thickness, where both PAC and neutron diffraction measurements are getting experimentally difficult.

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[1] P. Grünberg *et al.*, Phys. Rev. Lett. **57**, 2442 (1986).
[2] M. N. Baibich *et al.*, Phys. Rev. Lett. **61**, 2472 (1988).
[3] For magnetic properties of Cr in general, see E. Fawcett, Rev. Mod. Phys. **60**, 209 (1998), and references therein.
[4] J. Meersschaut *et al.*, Phys. Rev. Lett. **75**, 1638 (1995).
[5] E. E. Fullerton, S. D. Bader, and J. L. Robertson, Phys. Rev. Lett. **77**, 1382 (1996).
[6] A. Schreyer *et al.*, Phys. Rev. Lett. **79**, 4914 (1997).
[7] M. A. Tomaz *et al.*, Phys. Rev. B **55**, 3716 (1997).
[8] S. M. Dubiel, J. Magn. Magn. Mater. **124**, 31 (1993).
[9] K. Mibu, S. Tanaka, and T. Shinjo, J. Phys. Soc. Jpn. **67**, 2633 (1998).
[10] K. Fukumura, A. Nakanishi, and T. Kobayashi, Nucl. Instrum. Methods Phys. Res., Sect. B **86**, 387 (1994).
[11] K. Mibu *et al.* (unpublished).
[12] S. M. Dubiel, J. Cieślak, and F. E. Wagner, Phys. Rev. B **53**, 263 (1996); (private communication).
[13] T. Oguchi and H. Momida (unpublished).
[14] T. Emoto, N. Hosoito, and T. Shinjo, J. Phys. Soc. Jpn. **66**, 803 (1997).
[15] P. Bödeker *et al.*, Phys. Rev. Lett. **81**, 914 (1998).
[16] D. Stoeffler and F. Gautier, J. Magn. Magn. Mater. **147**, 260 (1995).