

Journal of Magnetism and Magnetic Materials 198-199 (1999) 689-691



## Magnetic properties of thin Cr layers in multilayer systems studied through <sup>119</sup>Sn Mössbauer probes

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## Abstract

The magnetic properties of thin Cr layers in epitaxial  $[Cr(t_{Cr})/Sn(2 \text{ Å})]$  ( $t_{Cr} = 5$ , 10, 20, 30, and 40 Å) and [Fe(10 Å)/Cr(5 Å)/Sn(2 Å)/Cr(5 Å)] multilayers were studied through <sup>119</sup>Sn conversion electron Mössbauer spectroscopy. It was found that the magnetic ordering temperature of Cr in both Cr/Sn and Fe/Cr/Sn/Cr systems is much higher than the Néel temperature of bulk Cr (i.e. 311 K). It also turned out that the magnetic hyperfine fields at the Sn nuclear sites are quite different, i.e., 11–13 T for the Cr/Sn multilayers and 2 T for the Fe/Cr/Sn/Cr multilayer at 300 K, in spite of the similarity of the local crystallographic structure of the Sn probe layers. Possible magnetic structures of the Cr layers are inferred from the size and direction of the magnetic hyperfine field transferred at the Sn nuclear sites. © 1999 Elsevier Science B.V. All rights reserved.

Keywords: Epitaxial multilayers; Mössbauer spectroscopy; Magnetic hyperfine field

The magnetic properties of thin Cr layers are known to be difficult to clarify, because Cr is basically antiferromagnetic, where information from the magnetization measurements is limited, and there is no proper experimental method using a Cr nuclear probe. Since Sn is a nonmagnetic element, having limited miscibility with Cr in the equilibrium states, the <sup>119</sup>Sn Mössbauer nucleus can be an appropriate probe to study magnetic properties of Cr [1]. We have studied the magnetic properties of epitaxial Cr/Sn and Fe/Cr/Sn/Cr multilayers through <sup>119</sup>Sn Mössbauer spectroscopy. It was found that the magnetic hyperfine fields at the Sn nuclear sites are sensitively influenced by the magnetic properties of the Cr layers. The magnetic transition temperature and possible magnetic structures of the Cr layers are discussed on the basis of the size and direction of the hyperfine field transferred at the Sn nuclear sites.

The epitaxial Cr/Sn and Fe/Cr/Sn/Cr multilayers were prepared on MgO(001) substrates in an ultrahigh vacuum  $(10^{-9}$  Torr range). The substrate temperature was kept at 200°C during the deposition. A Cr(50 Å) layer was deposited as a buffer layer prior to the depositon of multilayers.  $[Cr(t_{Cr})/Sn(2 \text{ Å})]$   $(t_{Cr} = 5, 10, 20, 30, \text{ and})$ 40 Å) and [Fe(10 Å)/Cr(5 Å)/Sn(2 Å)/Cr(5 Å)] multilayers with <sup>119</sup>Sn enriched Sn layers, where the Sn layer thickness roughly corresponds to a monatomic layer, were prepared for <sup>119</sup>Sn Mössbauer measurements. From the reflection high energy electron diffraction patterns during the film growth and the X-ray diffraction patterns after the preparation, it was confirmed that the Cr layers and the substrate have the structural relations MgO(001)//Cr(001) in the growth direction and MgO[100]//Cr[110] in the film plane, and that the Sn probe layers grow epitaxially with the Cr layers [2]. The <sup>119</sup>Sn Mössbauer spectra were measured by means of conversion electron Mössbauer spectroscopy, using a gas-flow counter with  $He + 1\%(CH_3)_3CH$  gas at room temperature and a gas-filled counter [3] with  $He + 2\%CH_4$  gas at 100 K and  $H_2$  gas at 15 K [4].

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A Ca<sup>119m</sup>SnO<sub>3</sub> source was used to obtain  $\gamma$ -rays of 23.8 keV and the direction of the incident  $\gamma$ -rays was set parallel to the film normal.

The <sup>119</sup>Sn Mössbauer spectrum for [Cr(30 Å)/Sn(2 Å)]measured at 300 K is shown in Fig. 1(a). The spectrum indicates a magnetically split six-line pattern, suggesting that the Sn nuclei feel very large magnetic hyperfine fields. The spectrum was fitted with six-line components with a distribution of magnetic hyperfine fields [2]. The obtained distribution of magnetic hyperfine fields is shown in Fig. 1(b). The isomer shift (relative to that for  $CaSnO_3$ ) was fitted to be 1.56 mm/s; this is a reasonable value when the Sn atoms are sandwiched with Cr atoms. The hyperfine field at the peak in the distribution curve was estimated to be 11 T. The result that large hyperfine fields are observed even at room temperature shows that the magnetic ordering temperature of the Cr layers is much higher than the Néel temperature of bulk Cr (i.e., 311 K). Possible reasons of the increase of the magnetic transition temperature would be strain in the Cr layers inherent to the epitaxial Cr/Sn system and an interface effect or a finite size effect that is intrinsic to the multilayers. Note that a preliminary analysis of the X-ray patterns shows that the lattice spacing in the Cr layers appears to be somewhat larger than that in bulk Cr. It is not clear, at the present stage, whether the observed large hyperfine fields are due to a possible enhancement of Cr magnetic moments at interfaces [5] or not; the relationship between the size of the magnetic moment at the interface Cr sites and the size of the transferred hyperfine field at the Sn nuclear sites is not straightforward.

The <sup>119</sup>Sn Mössbauer spectra for  $[Cr(t_{Cr})/Sn(2 \text{ Å})]$  $(t_{\rm Cr} = 40, 20, 10, \text{ and } 5 \text{ Å})$  measured at 300 K are shown in Fig. 2. All the spectra show the same extent of magnetic splitting indicating that they have similar distribution of magnetic hyperfine field. The hyperfine fields at the peak in distribution were estimated to be about 11 T for [Cr(40 Å)/Sn(2 Å)], 12 T for [Cr(20 Å)/Sn(2 Å)], and 13 T for [Cr(10 Å)/Sn(2 Å)]. The thickness of 40 Å corresponds to one half of the wavelength  $\Lambda$  of the spin density wave (SDW) in bulk Cr at room temperature [5]. The fact that the size of the hyperfine field is not much dependent on the Cr layer thickness between 5 and 40 Å implies that the Cr layer has a commensurate antiferromagnetic structure rather than an incommensurate SDW structure in this thickness range. The intensity ratio of the six lines of the spectra for the samples with  $t_{\rm Cr} = 20, 30, \text{ and } 40 \text{ Å}$  was fitted to be 3:1:1:1:1:3, which means  $\langle \cos^2 \theta \rangle = 0.6$ , where  $\theta$  is the direction of the hyperfine field relative to the film normal. In an antiferromagnetic Cr film with (001) orientation, either perpendicular magnetic moments or in-plane magnetic moments are expected to be stabilized if the magnetostrictive effect is sufficiently large. Neither of them is, however, the case in the present system.



Fig. 1. (a) <sup>119</sup>Sn Mössbauer spectrum for [Cr(30 Å)/Sn(2 Å)] at room temperature. The fitting line is also shown by the solid line. (b) The distribution of magnetic hyperfine fields obtained from the fitting of the spectrum for [Cr(30 Å)/Sn(2 Å)].



Fig. 2. <sup>119</sup>Sn Mössbauer spectra for  $[Cr(t_{cr})/Sn(2 \text{ Å})]$  ( $t_{cr} = 40$ , 20, 10, and 5 Å) at 300 K.



Fig. 3.  $^{119}Sn$  Mössbauer spectra for [Fe(10 Å)/Cr(5 Å)/Sn(2 Å)/Cr(5 Å)] at 300, 100, and 15 K.

For [Fe(10 Å)/Cr(5 Å)/Sn(2 Å)/Cr(5 Å)], where the crystallographic circumstances of the Sn layers are thought to be the same as those for [Cr(5 Å)/Sn(2 Å)], the magnetic splitting in the <sup>119</sup>Sn Mössbauer spectrum at 300 K (in Fig. 3) was found to be smaller than that for [Cr(5 Å)/Sn(2 Å)]. The hyperfine field was estimated to be 1.7 T at the maximum in distribution, and the distribution width was almost the same as that for [Cr(5 Å)/Sn(2 Å)]. The spectra at 100 and 15 K are also



Fig. 4. Temperature dependence of the hyperfine field (at the maximum in distribution) obtained from the <sup>119</sup>Sn Mössbauer spectra for [Fe(10 Å)/Cr(5 Å)/Sn(2 Å)/Cr(5 Å)] in Fig. 3.

shown in Fig. 3. The temperature dependence of the hyperfine field at the peak in the distribution estimated from the spectra in Fig. 3 is shown in Fig. 4. It appears that the magnetic transition temperature of the Cr layers in the Fe/Cr/Sn/Cr multilayer is also higher than the Néel temperature of bulk Cr. On the magnetism of thin Cr layers in Fe/Cr multilayers, contradictory results have been obtained from different experimental techniques. Perturbed angular correlation experiments have indicated that the Cr layers are nonmagnetic when the Cr layer thickness is smaller than 60 Å [6], and neutron diffraction measurements have shown that incommensurate SDW order is suppressed when the Cr layers are thinner than 42 Å but that commensurate antiferromagnetic order exists instead [7,8]. The present result that the Sn nuclei in [Fe(10 Å)/Cr(5 Å)/Sn(2 Å)/Cr(5 Å)] feel finite hyperfine fields at 300 K is consistent with the neutron diffraction experiments, although there might be an effect originating from the inserted Sn layers in the Fe/Cr/Sn/Cr system. The difference in the hyperfine fields between the Cr/Sn and the Fe/Cr/Sn/Cr multilayer at 300 K is not due to a difference in the magnetic transition temperature, but due to a difference in the magnetic properties of the Cr layers at 0 K. At the present stage, it is not clear whether this difference originates from an intrinsic difference in the electronic structures or from a difference in the strains of the Cr layers. The magnetic frustration effect accompanied by steps at the Fe/Cr interface also would reduce the size of Cr magnetic moments [9] and hence the hyperfine fields at the Sn nuclear sites. Further study is required to solve this problem.

The authors would like to thank Drs. T. Ono, and N. Hosoito for fruitful discussions during this work. This work was partially supported by a Grant-in-Aid for Creative Basic Research from Monbusho.

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