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Magnetic frustration of Cr at Fe(011)/Cr interfaces investigated by ¹¹⁹Sn Mössbauer spectroscopy

N Jiko^{1,5}, M Almokhtar², K Mibu^{3,6} and T Shinjo⁴

¹ Institute for Chemical Research, Kyoto University, Uji, Kyoto 611-0011, Japan

² Physics Department, Assiut University, Assiut, Egypt

³ Research Center for Low Temperature and Materials Sciences, Kyoto University, Uji, Kyoto 611-0011, Japan

⁴ International Institute for Advanced Studies, Kizu, Kyoto 619-0225, Japan

E-mail: jiko@ssc1.kuicr.kyoto-u.ac.jp

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Abstract

Magnetic properties of Cr layers in epitaxial Cr(011)/Sn and Fe(011)/Cr/Sn/Cr multilayers with monatomic Sn layers at the centre of the Cr layers were studied by means of ¹¹⁹Sn Mössbauer spectroscopy. It was found that Cr/Sn interfaces tend to enhance the magnetic moments of Cr layers and Fe/Cr interfaces tend to reduce the magnetic moments of Cr layers. These results were compared with those for (001)-oriented multilayers with the same composition. The large reduction of Cr magnetic moments has been found in Fe(011)/Cr/Sn/Cr multilayers and this is attributed to a magnetic frustration effect caused by the intrinsic magnetic structure of ferromagnetic Fe(011) and antiferromagnetic Cr(011) planes at the Fe/Cr interfaces.

1. Introduction

Fe/Cr multilayers are known as a system which shows the giant magnetoresistance (GMR) effect, and they are attracting scientific interest in view of both fundamental research and technical application. In order to study the fundamental mechanism of the GMR effect in this system, it is important to make clear the magnetic properties of the Cr layers, which mediate an interlayer exchange coupling between the Fe layers. When the Cr layers are so thin that the spin-density waves cannot be formed, the Cr layers are expected to be either antiferromagnetic with a simple commensurate structure or paramagnetic. However, it is difficult to obtain clear information on magnetism experimentally from antiferromagnetic thin layers.

⁵ Author to whom any correspondence should be addressed.

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⁶ Current address: Graduate School of Engineering, Nagoya Institute of Technology, Showa-ku, Nagoya 466-8555, Japan.



Figure 1. Relation between the magnetic moment of Cr in the Cr(001) layer which neighbours the inserted Sn layer (μ^{Cr}) and the magnetic hyperfine field (HF) at the Sn nuclear sites for Cr(001)/Sn multilayers (circles) and Fe(001)/Cr/Sn/Cr multilayers (squares) obtained from first principles (LDSA-FLAPW) calculations. The data shown by closed symbols are after Momida and Oguchi [1] and those with open symbols are after Arzhnikov *et al* [4]. The results for various multilayers with different Fe and Cr layer thicknesses and different lattice parameters are plotted in the same figure.

To study the magnetic properties of thin Cr layers, we have been using ¹¹⁹Sn Mössbauer spectroscopy with approximately monatomic Sn layers (0.2 nm) inserted in the Cr layers. Since Sn is a nonmagnetic element, we can interpret the hyperfine field (HF) induced at the Sn nucleus as reflecting the magnetic moments of Cr atoms adjacent to the Sn atom. The magnetism of Cr(001)/Sn multilayers with monatomic Sn layers was studied by Mibu *et al* [1]. It was suggested that the Cr layers are antiferromagnetically ordered at room temperature and that the Cr/Sn interfaces tend to enhance the magnetic moments of the Cr layers.

Fe(001)/Cr/Sn/Cr multilayers, where a monatomic Sn layer is inserted at the centre of the Cr layers, were also studied [2]. The result in contrast suggests that the Fe/Cr interfaces tend to reduce the magnetic moments of the Cr layers.

Recently, Momida and Oguchi [3] and Arzhnikov *et al* [4] studied the magnetism of Cr(001)/Sn and Fe(001)/Cr/Sn/Cr multilayers by first principles (LDSA-FLAPW) calculations, and it was found that the magnitude of the HF at the Sn nuclei is proportional to that of the magnetic moments of Cr neighbouring the Sn nuclei, as shown in figure 1. The results obtained from these calculations also explain the rough trend observed in Fe(001)/Cr/Sn/Cr multilayers that the HF reduces with a decrease of Cr layer thickness. The theoretical calculations, however, predict HFs larger than those observed experimentally. One reason for this discrepancy between the experimental and theoretical results is that the LDSA-FLAPW calculations tend to overestimate the size of the HF, so the experimentally obtained value of the HF is somewhat smaller than the calculated value for each multilayer with certain layer thicknesses. Another possible reason is that the real Fe/Cr interface has atomic steps and interdiffusion, whereas in the calculations all the interfaces in the multilayers are assumed to be perfectly flat. Atomic steps cause a magnetic frustration at interfaces [5], and this frustration is expected to be relieved by the reduction of magnetic moments of Cr [6, 7]. The experimental results support this theoretical suggestion.

We mentioned above multilayers with bcc (001) orientation. In the present paper, we discuss the magnetic properties of Cr layers in Cr/Sn and Fe/Cr/Sn/Cr multilayers with bcc

(011) orientation. When the Cr layer has a commensurate antiferromagnetic structure, the magnetic moments at the cubic corners and body-centres are antiparallel, so that the (011) plane has a compensated antiferromagnetic structure. Therefore, a strong frustration effect is expected to occur at the Fe/Cr interfaces even if the interfaces are atomically flat. We applied ¹¹⁹Sn Mössbauer spectroscopy to this system with inserted monatomic Sn layers in the Cr layers and compared the results obtained with those of the (001)-oriented multilayers.

2. Experiments

We have grown Cr/Sn and Fe/Cr/Sn/Cr multilayers with (011) orientation on MgO(111) substrates using an ultrahigh-vacuum deposition system. The epitaxial growth of Cr(011) on MgO(111) was reported by Mattson *et al* [8]. We performed x-ray diffraction measurements with the scattering vectors normal to the surface using Cu K α radiation. ¹¹⁹Sn Mössbauer spectroscopic measurements were carried out at room temperature by means of conversion electron Mössbauer spectroscopy using a gas-flow counter with He + 1%(CH₃)₃CH. The isomer shift is expressed relatively to that of CaSnO₃.

The configurations of the prepared samples are as follows. Cr/Sn multilayers: MgO(111)/Cr(5.0 nm)/[¹¹⁹Sn(0.2 nm)/Cr(t_{Cr})] × 30 or 40 (t_{Cr} = 8.0, 4.0, 2.0 and 1.0 nm) and Fe/Cr/Sn/Cr multilayers: MgO(111)/Cr(5.0 nm)/[Fe(1.0 nm)/Cr(t_{Cr})] × 29/Fe(1.0 nm) (t_{Cr} = 8.0, 4.0, 2.0 and 1.0 nm). The MgO(111) substrates were annealed at 400 °C before film growth in the deposition chamber. The Cr buffer layers (5.0 nm) were deposited at the substrate temperature of 200 °C for all samples. To determine the optimum substrate temperature for the Cr/Sn multilayers, we deposited the sample with t_{Cr} = 4.0 nm at different substrate temperatures from 50 to 600 °C. After considering the results of RHEED patterns, x-ray patterns and ¹¹⁹Sn Mössbauer spectra, we selected 400 °C as the substrate temperature. The Fe/Cr/Sn/Cr multilayers were prepared at 200 °C to minimize interdiffusion at the Fe/Cr interfaces.

3. Results

Figure 2 shows the x-ray diffraction patterns for Cr/Sn and Fe/Cr/Sn/Cr multilayers with $t_{Cr} = 4.0$ nm. Bragg peaks corresponding to the artificial period appear in the patterns at low angles in (a) and (b). In the patterns around Cr(011) peaks in (c) and (d), satellite peaks are clearly visible. These facts indicate that the artificial periodicity is well established in these samples.

The ¹¹⁹Sn Mössbauer spectra for the Cr(011)/Sn multilayers with $t_{Cr} = 4.0$ and 2.0 nm are shown in figures 3(a) and (b). The ¹¹⁹Sn Mössbauer spectra were least-square fitted. The results of the fitting are drawn with lines in the same figures. There are two kinds of components in each spectrum. The minor component has a zero isomer shift and a zero HF. This component would come from the Sn oxide at the surface of the evaporated source metal of Sn. The major component has a variety of finite HFs but a common isomer shift between 1.5 and 1.6 mm s⁻¹. It was assumed for the fitting that the angle between the HF and the normal direction to the film plane is 54.7°, which corresponds to the random orientation of HFs. It was confirmed by the magic-angle method [9] that the HFs are oriented at random in the Cr(011)/Sn multilayer with $t_{Cr} = 4.0$ nm. The HF distributions for the major component are shown in figures 3(c) and (d). The ¹¹⁹Sn Mössbauer spectra for the Fe(011)/Cr/Sn/Cr multilayers with $t_{Cr} = 4.0$ and 2.0 nm are shown in figures 4(a) and (b), and the HF distributions are shown in figures 4(c) and (d). The angle of the HFs was assumed to be random in for the Cr(011)/Sn multilayers.



Figure 2. X-ray diffraction patterns at low angles, (a) and (b), and patterns around the Cr(011) peak, (c) and (d). Figures (a) and (c) are for the [Cr(4.0 nm)/Sn(0.2 nm)] multilayer and (b) and (d) are for the [Fe(1.0 nm)/Cr(4.0 nm)/Sn(0.2 nm)] multilayer.



Figure 3. ¹¹⁹Sn Mössbauer spectra at room temperature for Cr(011)/Sn multilayers with (a) t_{Cr} = 4.0 and (b) t_{Cr} = 2.0 nm. The results of fitting are drawn with lines. The small peaks at zero isomer shift come from Sn oxide. Hyperfine field distributions at the Sn sites, (c) and (d), obtained by fitting the ¹¹⁹Sn Mössbauer spectra in (a) and (b), respectively.

The averaged HFs for the Cr/Sn and Fe/Cr/Sn/Cr multilayers with both (011) and (001) orientations are plotted versus t_{Cr} in figure 5(a). As shown in figures 3 and 4, there are a few sites in the spectra. The origin of the sites except for the one with maximum distribution is unclear, but it may be the disordered Sn sites. We think it is informative to show also the HF with the maximum distribution in order to see the intrinsic properties. In figure 5(b), the HFs at the maximum in distribution for the Cr/Sn and Fe/Cr/Sn/Cr multilayers are plotted



Figure 4. ¹¹⁹Sn Mössbauer spectra at room temperature for the Fe(011)/Cr/Sn/Cr multilayers with (a) $t_{Cr} = 4.0$ and (b) $t_{Cr} = 2.0$ nm. The results of fitting are drawn with lines. Hyperfine field distributions at Sn sites, (c) and (d), obtained by fitting the ¹¹⁹Sn Mössbauer spectra (a) and (b), respectively.



Figure 5. (a) Averaged hyperfine field at Sn sites and (b) hyperfine field with the maximum distribution. The symbols denote the data as follows: O, Cr(001)/Sn; \bullet , Fe(001)/Cr/Sn/Cr; \Box , Cr(011)/Sn; \blacksquare , Fe(011)/Cr/Sn/Cr.



Figure 6. Simple pictures of the magnetic configuration at (a) Fe(001)/Cr and (b) Fe(011)/Cr interfaces. Atomic steps cause the magnetic frustration at the Fe(001)/Cr interface. The Cr(011) plane is magnetically compensated so that the ferromagnetic Fe(011) on the Cr(011) plane causes the magnetic frustration.

versus t_{Cr} . The results of multilayers with both (011) and (001) orientations are shown in order to make clear the orientation dependence. The HF in the Cr/Sn multilayers increases slightly with the decrease of t_{Cr} . This tendency is common for both (011) and (001) orientations. We think that such a tendency comes from two factors: i.e., the Cr/Sn interfaces tend to enhance the Cr magnetic moments, whereas Cr in the middle of the Cr layers tends to keep the magnitude of the magnetic moments in its bulk state. The former effect becomes dominant when t_{Cr} decreases. Note that Dubiel studied the HFs at Sn atoms dissolved in bulk Cr [10]. In Cr_ySn_{1-y} (y = 0.07-1.18 at.%) alloys, the HF distribution curves at room temperature have many peaks, with positions ranging from 0 to 8 T, which are smaller than the peaks with the maximum distribution of both Cr(001)/Sn and Cr(011)/Sn multilayers.

The HF in the Fe/Cr/Sn/Cr multilayers, on the other hand, is reduced with a decrease of t_{Cr} . This behaviour is common for both orientations and can be attributed to an effect at the Fe/Cr interfaces, which reduces the magnetic moments of Cr. However, there is a big difference in the magnitude of the HF between the Fe/Cr/Sn/Cr multilayers with different orientations, although the magnitude of the HF in the Cr/Sn multilayers is not so different between the two orientations. The HF in the Fe(011)/Cr/Sn/Cr multilayers is clearly smaller than that in the Fe(001)/Cr/Sn/Cr multilayers with the same t_{Cr} . This fact suggests that there is a larger effect that reduces the magnetic moments of Cr at the Fe(011)/Cr interfaces.

4. Discussion

We have found that there is an effect at the Cr(011)/Sn interfaces that enhances the magnetic moments of Cr. Momida and Oguchi found by means of a first-principles method that, in Cr(001)/Sn multilayers, interface Cr-d states with non-bonding character cause an enhancement of magnetic moments of Cr [3]. We speculate that such interface states of Cr are also produced in Cr(011)/Sn multilayers.

In Fe(001)/Cr/Sn/Cr multilayers, the reduction of magnetic moments of Cr observed experimentally is roughly reproduced by first-principles calculations and is interpreted as a band effect at the Fe/Cr interfaces [3, 4, 11]. The difference of the magnitude of the reduction in the HF between the calculations and experiments may come from the magnetic frustration at the Fe/Cr interfaces due to atomic steps and interdiffusion. Although all the exchange interactions of Fe–Cr, Fe–Fe and Cr–Cr spin pairs are energetically satisfied at a perfect interface, a magnetic frustration occurs at a real interface with atomic steps. The frustration reduces the magnetic moments of Cr. A simple picture is illustrated in figure 6(a).

In Fe(011)/Cr/Sn/Cr multilayers, on the other hand, the extent of the reduction of Cr magnetic moments is rather large compared to multilayers with the (001) orientation. We think that there is an intrinsic origin of magnetic frustration in the Fe(011)/Cr/Sn/Cr multilayers. In these multilayers, a Cr(011) plane is magnetically compensated, i.e., there are the same number of Cr moments aligned in one direction and the opposite direction if Cr has the simple antiferromagnetic structure with the antiparallel magnetic moments at the cubic corners and the body-centres. When a ferromagnetic Fe(011) plane is deposited on it, one half of the Fe–Cr spin pairs can couple so that the Heisenberg energy is minimized, whereas the other half cannot (figure 6(b)). Such a situation leads to a large frustration and causes a large degree of reduction of the Cr magnetic moments.

5. Conclusion

We have shown that there is an effect at the Cr(011)/Sn interfaces that enhances the magnetic moments of Cr and an effect at the Fe(011)/Cr interfaces that reduces the magnetic moments of Cr. The former effect is similar to that at the Cr(001)/Sn interfaces in magnitude and the latter effect is more pronounced than that of the Fe(001)/Cr/Sn/Cr interfaces. A large dependence of the interface effects on the growth orientation is thus elucidated.

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