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## MAGNETORESISTANCE OF EPITAXIALLY GROWN METALLIC MULTILAYERS

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### ABSTRACT

The epitaxial TM/bcc-Cr(001) (TM=Fe, Co, Ni) multilayers have been prepared using MBE. The crystal structure, interlayer exchange coupling and magnetoresistance of those multilayers have been discussed. The structure of Fe, Co and Ni grown on bcc-Cr(001) exhibited bcc(001), distorted hcp(11 $\bar{2}$ 0) and fcc(110), respectively. In Fe/Cr multilayers, an oscillatory exchange coupling has been observed, but not observed in Ni/Cr system, which may come from the large mixing at interfaces. Large MR ratio (116%, 4.2K) has been obtained in Fe/Cr system, but only 2% in Co/Cr system. This difference can be understood from the view point of the relative potential height for down spin electrons between TM and Cr.

### INTRODUCTION

The oscillatory exchange coupling<sup>[1]</sup> and associated giant magnetoresistance (GMR) effect<sup>[2]</sup> in magnetic multilayers are currently of great interest. Since the discovery, much work has been carried out on these phenomena in various kinds of multilayers. Recently, a maximum magnetoresistance (MR) ratio of 220% (1.5K) has been found in high-quality epitaxially grown Fe/Cr(001) multilayers<sup>[3]</sup>. Since both the antiferromagnetic (AF) coupling and the magnetoresistance are very sensitive to the structural quality of multilayers, high quality samples and detailed structural investigation are necessary to understand these phenomena. The crystal structure of Fe and Cr are the same body-centered-cubic (bcc), and lattice mismatch is

very small, which is an advantage to prepare good samples. On the other hand, those of Co and Ni in the bulk state are hexagonal-closed packed(hcp) and face centered cubic(fcc) structure, therefore their lattice matchings to Cr are not good and well controlled epitaxial sample preparation is very difficult. Although it is interesting to know the exchange coupling and the MR effect of the epitaxial [Co/Cr] and [Ni/Cr] multilayers, little studies have been reported so far, for these reasons. The MR of the [Co/Cr] multilayer was first measured by Parkin et al.<sup>[1]</sup>, however the film structure was not reported in detail. The preparation of the epitaxial [Co/Cr] multilayers were reported by Th. Zeidler et al.<sup>[4]</sup>, but the MR measurement was not performed because of thick buffer layers. In the [Ni/Cr] case no study of epitaxial multilayers has

been reported so far.

In this paper we present the magnetization and MR effect measurements of TM/bcc-Cr(001) (TM=Fe, Co, Ni) multilayers prepared carefully by molecular beam epitaxy (MBE) technique, and compare those properties to each other.

## EXPERIMENTAL PROCEDURE

Samples were grown on MgO(001) substrates in an ultrahigh vacuum chamber (VG Semicon V80M system) with a base pressure of  $1 \times 10^{-11}$  Torr. The Fe, Co and Ni layers were deposited by eb-guns and Cr is using Knudsen cell with a rate of 3-5 Å/min. The 50-100 Å Cr buffer layers were deposited onto the substrate, and followed by [TM/Cr]<sub>20</sub> multilayers. The substrate temperature was held at 500°C during the buffer deposition, and down to 90°C. the surface structure of the films was examined by reflection high-energy-electron diffraction (RHEED) observations during the growth. The structure of multilayers perpendicular to the film plane was determined by X-ray diffraction using CuK $\alpha$  radiation. Magnetization measurements were carried out by vibrating sample magnetometer (VSM) and superconducting quantum interference device (SQUID) magnetometry. In the case of the [Co/Cr], which shows the perpendicular anisotropy, magnetic field was applied both in the plane of the sample and perpendicular to it. The MR effect was measured by the conventional four point method. A superconducting magnet (10T) was used for the [Fe/Cr] because of the large saturation magnetic field. In this study, the MR ratio is defined as  $(\rho_0 - \rho_s) / \rho_s$ , where  $\rho_0$  is the

resistivity at  $H=0$  kOe and  $\rho_s$  is at the saturation field  $H_s$ .

## RESULTS AND DISCUSSION

Fig. 1 shows the typical high angle X-ray diffraction patterns of [TM/Cr] on bcc-Cr(001) multilayers. Several satellite peaks in the pattern indicate the evidence for the good artificial periodic structure. The large intensity of satellite peaks in [Co/Cr] and [Ni/Cr] suggest that there is a difference between the interlayer spacing of TM(=Co, Ni) and Cr. According to the analysis of them<sup>[5]</sup> using step model as shown in Fig. 2, the lattice spacing of Fe and Cr perpendicular to the film plane are almost the same as bulk values of  $d_{(002)}$ . On the contrary, those of Co and Ni are small compared to Cr, and reduced asymptotically to  $d_{(1120)}$  of bulk hcp-Co and  $d_{(220)}$  of fcc-Ni as the Co or Ni thickness is thicker. These results indicate that the crystal

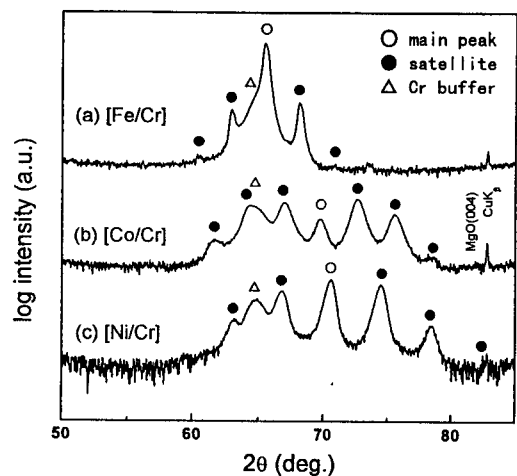


Fig. 1 High angle X-ray diffraction patterns of [TM/bcc-Cr(001)] (TM=Fe, Co, Ni) multilayers. (a) [Fe20 Å/Cr20 Å]<sub>20</sub>. (b) [Co20 Å/Cr18 Å]<sub>20</sub>. (c) [Ni16 Å/Cr12 Å]<sub>20</sub>.

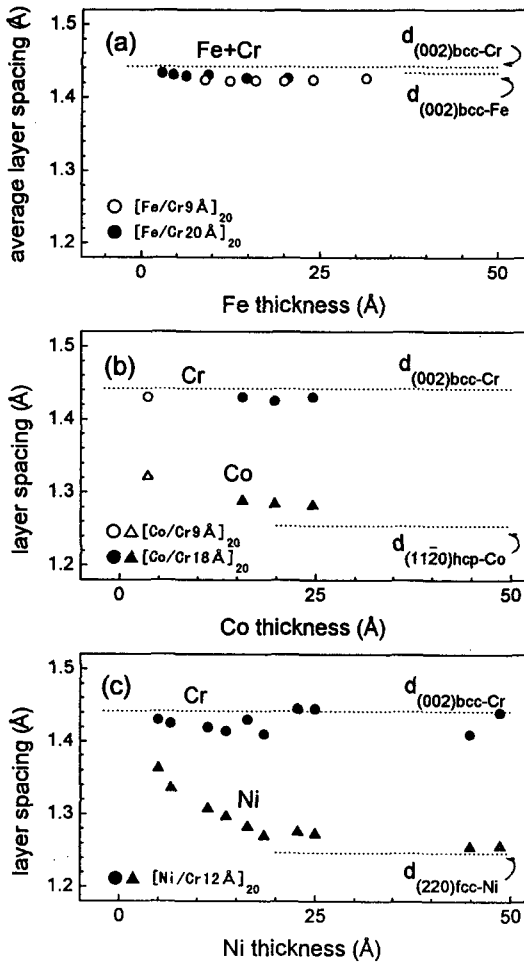


Fig. 2 Variations of layer spacing of TM and Cr in  $[\text{TM}/\text{bcc-Cr}(001)]$  (TM=Fe,Co,Ni) multilayers as a function of TM layer thickness. (a)  $[\text{Fe}/\text{Cr}]$ . In  $[\text{Fe}/\text{Cr}]$  (b)  $[\text{Co}/\text{Cr}]$ . (c)  $[\text{Ni}/\text{Cr}]$ , the average layer spacing of Fe and Cr is plotted since it is difficult to separate similar layer spacings.

structure of Fe and Cr in  $[\text{Fe}/\text{Cr}]$  multilayers are bcc, however Co and Ni are distorted hcp  $(11\bar{2}0)$  and fcc(110) respectively. Details are described elsewhere together with the analysis of RHEED observations<sup>[5]</sup>.

Fig. 3 shows the magnetization curves of  $[\text{TM}/\text{Cr}]$  multilayers. The measured saturation

magnetization ( $M_s$ ) of Fe in  $[\text{Fe}/\text{Cr}]$  is almost the same to the bulk value, however that of Co and Ni in multilayers are much reduced from bulk values ( $M_s$  of the bulk bcc-Fe, hcp-Co and fcc-Ni are 221.9, 162.5 and 57.5 emu/g respectively at 0K). It suggests the possibility of the formation of CoCr and NiCr alloys later at interfaces of multilayer, which is discussed in later.

The hysteresis loop of  $[\text{Co}8\text{Å}/\text{Cr}20\text{Å}]_{20}$  indicates that the 8Å Co layer on Cr has the perpendicular anisotropy and that the Co layers are ferromagnetically coupled the large remanent magnetization is observed. By varying Cr thickness, loop shapes are drastically changed. The hysteresis loop of  $[\text{Co}8\text{Å}/\text{Cr}9\text{Å}]_{20}$  has on remanent magnetization and the spin flop transition is observed at approximately 9kOe. As calculated the similar type of the spin flop in the bilayer film<sup>[6]</sup>, the magnetization of Co layers in  $[\text{Co}8\text{Å}/\text{Cr}9\text{Å}]_{20}$  are antiferromagnetically coupled through the Cr layer and oriented perpendicular to the film plane, at zero field.

As described above, the interlayer exchange coupling could be evaluated by the remanent magnetization ( $M_r$ ). In Fig. 4, the magnitude of  $M_r/M_s$  was plotted for  $[\text{TM}/\text{Cr}]$  multilayers as a function of Cr layer thickness. The data for the  $[\text{Fe}/\text{Cr}]$  multilayers are quoted from those obtained by E.E. Fullerton et al<sup>[7]</sup> as plotted in Fig. 4b.

As shown in Fig. 4a, an oscillatory Cr thickness dependence of  $M_r/M_s$  was observed for the  $[\text{Co}/\text{Cr}]$  multilayers, which is almost the same as the  $[\text{Fe}/\text{Cr}]$  multilayers (Fig. 4b). On the other hand, the magnitude of  $M_r/M_s$  of the  $[\text{Ni}/\text{Cr}]$  multilayers is relatively high, and has no Cr thickness dependence,

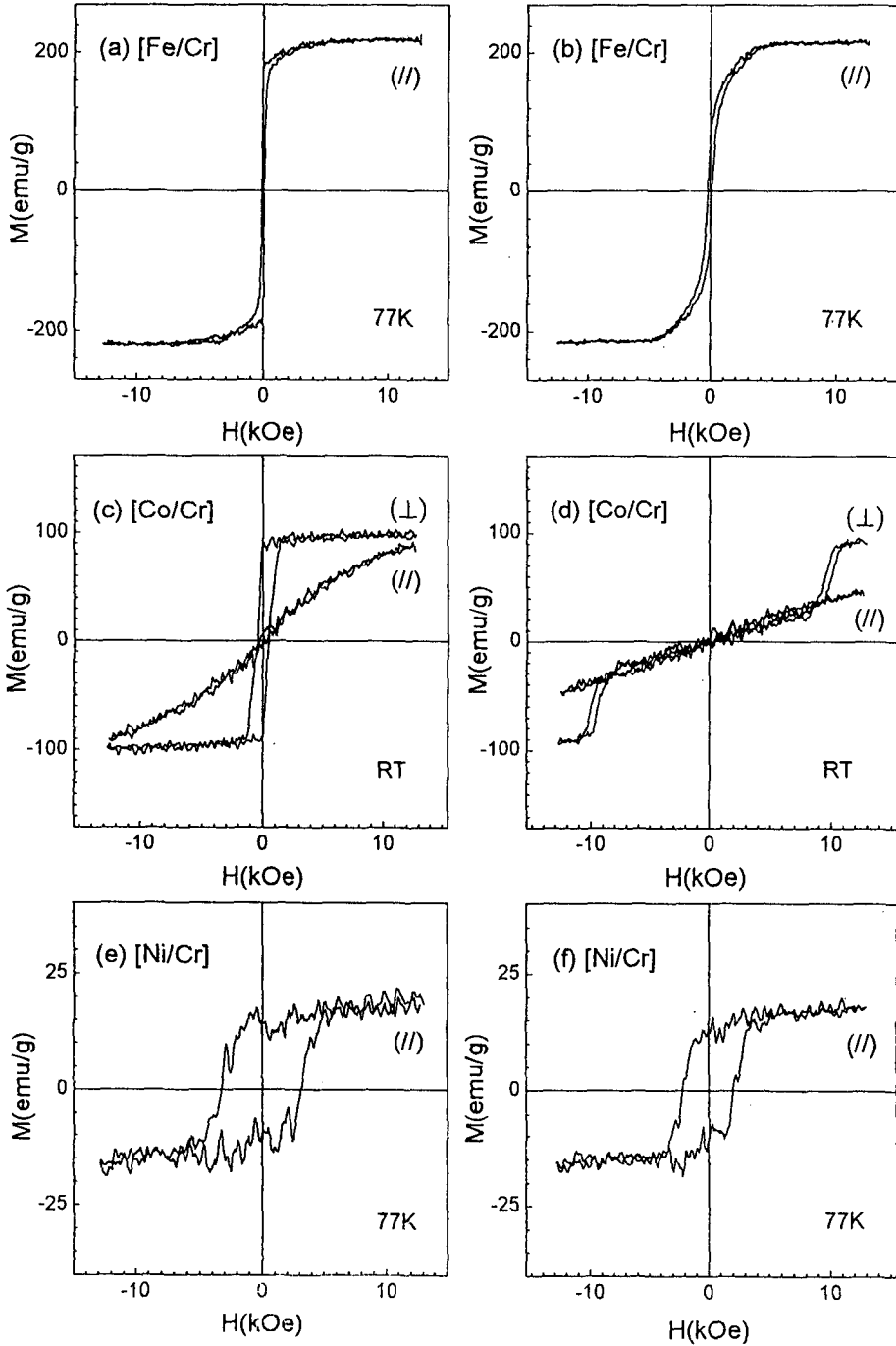


Fig. 3 Magnetization curves of  $[TM/bcc-Cr(001)]$  ( $TM=Fe, Co, Ni$ ) multilayers. (a)  $[Fe\text{\AA}/Cr20\text{\AA}]_2$ . (b)  $[Fe20\text{\AA}/Cr13\text{\AA}]_{20}$ . (c)  $[Co8\text{\AA}/Cr18\text{\AA}]_{20}$ . (d)  $[Co8\text{\AA}/Cr9\text{\AA}]_{20}$ . (e)  $[Ni18\text{\AA}/Cr22\text{\AA}]_{20}$ . (f)  $[Ni18\text{\AA}/Cr12\text{\AA}]_{20}$ . ( $//$ ): a magnetic field was applied in the film plane. ( $\perp$ ): perpendicular to the film plane.

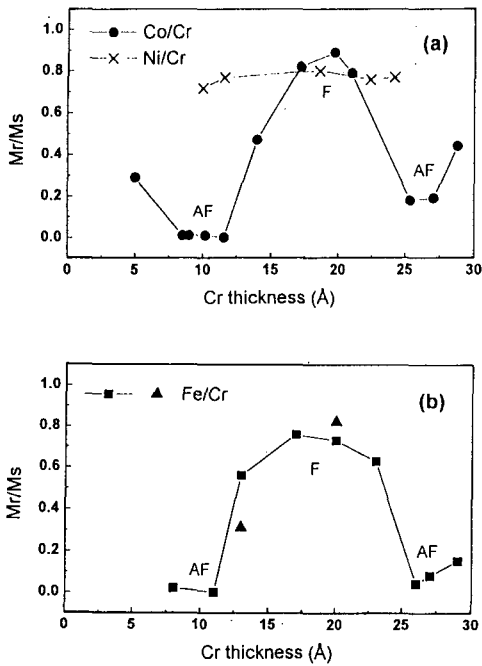


Fig. 4 Variations of  $M_r/M_s$  of  $[tM/bcc-Cr(001)]$  (TM=Fe, Co, Ni) multilayers as a function of Cr thickness. (a)  $[Co8\text{\AA}/Cr]_{20}$  at RT and  $[Ni18\text{\AA}/Cr]_{20}$  at 77K. (b) circle: rif.7, triangle: present data of  $[Fe20\text{\AA}/Cr]_{20}$  at 77K.

which indicate the ferromagnetic or magnetically uncoupled state between Ni layers. In order to explain the disappearance of AF coupling in the  $[Ni/Cr]$ , differences of spacer layer are considered from the view point of mixing of Ni and Cr at interfaces, as follows.

In order to evaluate the thickness of the interlayer mixing, the following approximations were adopted.

- 1) The compositional profile is assumed to be trapexoidal.
- 2) The magnetization of the pure Co (or Ni) lauer is taken to be the bulk hcp-Co value  $M_{s,hcp-Co}$  (or fcc-Ni,  $M_{s,fcc-Ni}$ ).
- 3) The magnetization of the mixing region is taken to be the same compositional depen-

dence as bulk magnetization, where the bulk CoCr and NiCr allous become nonmagnetic at 25% at.Cr and 12% at.Cr respectively.

According to these assumptions, a schematic representation of the TM (Co, Ni) concentration and magnetization profiles along the growth direction is given in Fig. 5, where  $t_{Co(or\ Ni)}$  and  $t_{mix}$  denote the thickness of Co (or Ni) effective lauers and that of mixing region, respectively. Using thes model, the measured magnetizations satisfy the follow- ing expressions.

$$M_{s\ t_{Co}} = M_{s\ hcp-Co} (t_{Co} - 0.75t_{mix}) : \text{for} [Co/Cr] \quad (1)$$

$$M_{s\ t_{Ni}} = M_{s\ fcc-Ni} (t_{Ni} - 0.88t_{mix}) : \text{for} [Ni/Cr] \quad (2)$$

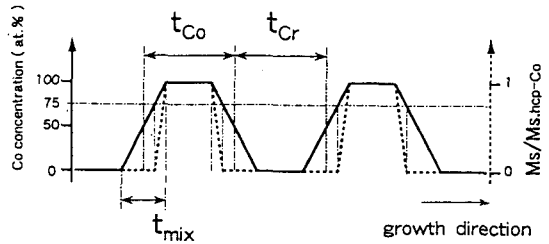


Fig. 5 schematic representation of the TM (Co, Ni) concentration and magnetization profiles along the growth diection.

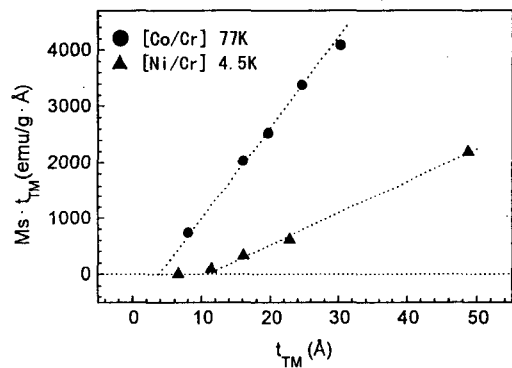


Fig. 6  $M_s \cdot t_{TM}$  as a function of the effective layer thickness,  $t_{TM}$  (TM=Co, Ni).

The experimental results of  $M_s \cdot t_{TM}$  as a function of the effective layer thickness,  $t_{TM}$  ( $TM=Co, Ni$ ) are shown in Fig. 6. As a result of the analysis,  $t_{mix}$  of about  $5\text{\AA}$  for  $[Co/Cr]$  and about  $12\text{\AA}$  for  $[Ni/Cr]$  were obtained.

In the  $[Ni/Cr]$  multilayer, the large interlayer mixing might reduce the AF coupling between Ni layers. As another reason, we propose that the existence of the different two kinds of materials (Cr and NiCr) in a spacer layer causes the discontinuity of the reciprocal lattice vectors which contribute to the interlayer exchange coupling.

Fig. 7 shows the MR curves of  $[TM/Cr]$  multilayers. The maximum MR ratio of the  $[Fe/Cr]$  multilayers is 116% at 4.2K, however that of the  $[Co/Cr]$  is 2% at most. This difference can be understood by the calculated results obtained by Inoue et al.<sup>[8]</sup>. According to their model, the conduction electrons are scattered by the spin dependent random exchange potential, which is defined as the impurity potential caused by the random substitution of atoms at the interface. As a result of their calculation, a larger MR ratio is obtained for the  $[Fe/Cr]$  multilayer since the difference between the potential of down spin conduction electron in Fe and Cr,  $\Delta\epsilon(Fe/Cr)$ , is nearly equal, and the resistivity of down spin electrons is much reduced at the ferromagnetically coupled configuration of Fe layers. In the case of the  $[Co/Cr]$  multilayer  $\Delta\epsilon(Co/Cr)$  is larger than  $\Delta\epsilon(Fe/Cr)$  and the MR ratio becomes small. It is difficult for the quantitative comparison of our experimental results with the model. However, a large difference of the MR ratio between  $[Fe/Cr]$  and  $[Co/Cr]$  found in our

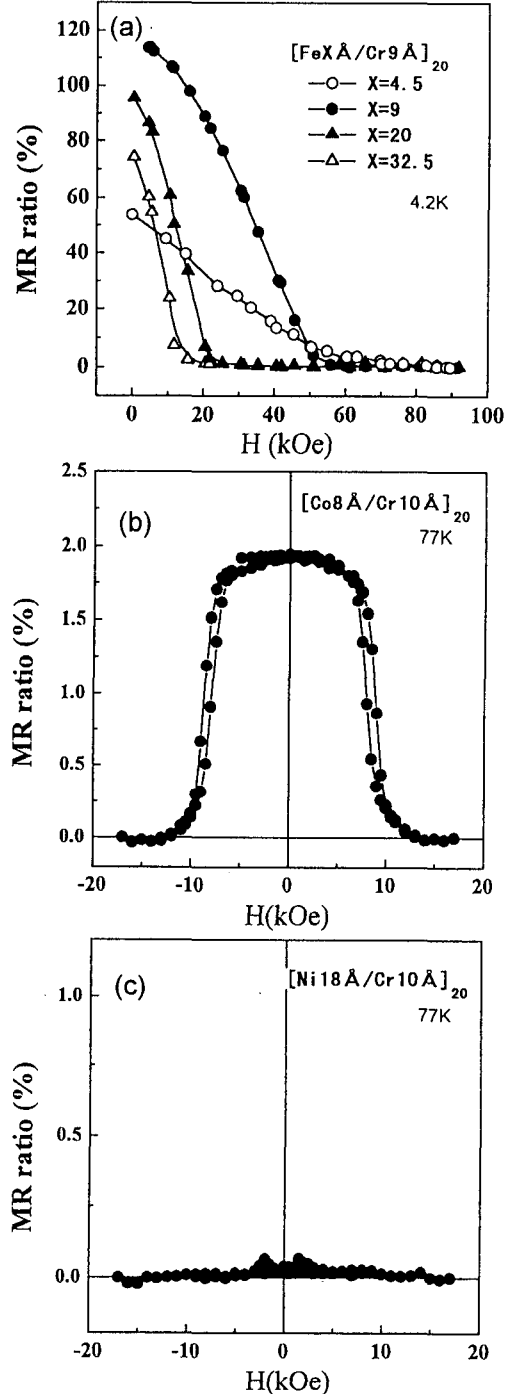


Fig. 7 MR curves of  $[TM/bcc-Cr(001)]$  ( $TM=Fe, Co, Ni$ ) multilayers.

works is qualitatively consistent with the calculated results. The MR ratio of the [Ni/Cr] multilayers is much small (about 0.1%), which is explained by nin AF coupling between Ni layers.

### CONCLUSIONS

The epitaxial [TM/bcc-Cr(001)]<sub>20</sub> (TM= Fe,Co,Ni) multilayers were prepared using MBE methods. The crystal structure of Fe, Co and Ni grown on the bcc-Cr(001) were bcc(001), distorted hcp(1120) and distorted fcc(110) respectibely. The oscillatory behavior of the interlayer exchange coupling was found in [Fe/Cr] and [Co/Cr], but no oscillatory behavior in [Ni/Cr]. By means of an analysis of the saturation mgnetizationm a thickness of the mixing layer was evaluated. The MR ratio of 116% for [Fe/Cr] and 2% for [Co/Cr] were obtained, which is explained by Inoue's calculation. The MR ratio of [Ni/Cr] was 0.1% at most because of a large interlayer mixing of [Ni/Cr].

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