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# First-principles study on half-metallicity of disordered $\text{Co}_2(\text{Cr}_{1-x}\text{Fe}_x)\text{Al}$

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We have investigated theoretically the effects of atomic disorder on the electronic and magnetic structures of the full-Heusler alloy  $\text{Co}_2(\text{Cr}_{1-x}\text{Fe}_x)\text{Al}$  using the first-principles density functional calculation with the Korringa–Kohn–Rostoker coherent-potential approximation. It was found that  $\text{Co}_2(\text{Cr}_{0.6}\text{Fe}_{0.4})\text{Al}$  preserves the high spin polarization even in the disordered B2 structure, where (Cr,Fe) and Al randomly occupy octahedral sites of the alloy. On the other hand, the disorder between Co and (Cr,Fe) considerably reduces the spin polarization. Furthermore, the total magnetic moment of  $\text{Co}_2(\text{Cr}_{0.6}\text{Fe}_{0.4})\text{Al}$  decreases with increasing disorder between Co and (Cr,Fe) due to the antiferromagnetic coupling of the antisite Cr with the ordinary site Cr. These results indicate that control of the disorder between Co and (Cr,Fe) is crucial in order to obtain highly spin polarized full-Heusler alloys  $\text{Co}_2(\text{Cr}_{1-x}\text{Fe}_x)\text{Al}$ . © 2004 American Institute of Physics.  
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Half-metallic ferromagnets (HMFs), where the majority-spin band is metallic, and the minority-spin band is semiconducting with an energy gap at the Fermi level, act as spin filters which provide current with a high degree of spin polarization. They are key materials in spintronics and crucial to spin-dependent phenomena, such as tunneling magnetoresistance (TMR)<sup>1</sup> and spin injection into semiconductors.<sup>2,3</sup> HMFs were first proposed by de Groot *et al.*<sup>4</sup> from band-structure calculations of the  $\text{C1}_b$ -type Heusler alloys (half-Heusler), NiMnSb and PtMnSb. So far, various kinds of HMFs have been studied, such as the rutile-type  $\text{CrO}_2$ ,<sup>5</sup> the transition-metal perovskites  $\text{La}_{0.7}\text{Sr}_{0.3}\text{MnO}_3$ ,<sup>6</sup>  $\text{L2}_1$ -type Heusler alloys (full-Heusler),<sup>7–12</sup> other half-Heusler alloys,<sup>13</sup> and zinc-blende type MnAs, CrAs, and CrSb.<sup>14–16</sup>

Though many materials have been predicted to be half-metallic, it is difficult to demonstrate their half-metallicity experimentally. The nonstoichiometry at the surface/interface is responsible for the reduction in spin polarization.<sup>17,18</sup> Recently Block *et al.*<sup>10,12</sup> have found that pressed powder compacts of the full Heusler alloy  $\text{Co}_2(\text{Cr}_{0.6}\text{Fe}_{0.4})\text{Al}$  have the disordered B2 structure and show a large magnetoresistive effect of 30% in a small magnetic field at room temperature (RT). Furthermore, Inomata *et al.*<sup>19</sup> have shown that a magnetic tunneling junction based on a thin film  $\text{Co}_2(\text{Cr}_{0.6}\text{Fe}_{0.4})\text{Al}$  with the disordered B2 structure has a relatively large TMR of 16% at RT. Their experimental results suggest that the spin polarization of  $\text{Co}_2(\text{Cr}_{0.6}\text{Fe}_{0.4})\text{Al}$  has highly spin polarized electronic structures even in atomically disordered states in contrast to NiMnSb<sup>20</sup> and is hopeful as a material in spintronic devices.

The electronic and magnetic properties of  $\text{Co}_2(\text{Cr}_{1-x}\text{Fe}_x)\text{Al}$  with the ordered  $\text{L2}_1$  structure have been investigated using the first-principles calculation.<sup>10–12,21,22</sup> It was found that the total magnetic moment of ordered  $\text{Co}_2(\text{Cr}_{1-x}\text{Fe}_x)\text{Al}$  follows the Slater–Pauling behavior  $M_t$

$=Z_t - 24$ , where  $M_t$  is the total magnetic moment in  $\mu_B$  per unit cell and  $Z_t$  is the total number of valence electrons, which scales linearly with the Fe concentration  $x$ .

In this work, we investigate and discuss the effects of atomic disorder on the electronic and magnetic properties of  $\text{Co}_2(\text{Cr}_{0.6}\text{Fe}_{0.4})\text{Al}$ , using the first-principles calculation. We consider two types of disorder of the alloy, one is a disorder between Co and  $(\text{Cr}_{0.6}\text{Fe}_{0.4})$  (Co–CrFe type disorder), and the other is a disorder between  $(\text{Cr}_{0.6}\text{Fe}_{0.4})$  and Al (CrFe–Al type disorder).

The calculations are performed using the *ab initio* calculation code based on the Korringa–Kohn–Rostoker method<sup>23</sup> developed by Akai *et al.*<sup>24</sup> and the atomic disorder calculations are implemented within the coherent potential approximation. The space is divided into nonoverlapping muffin-tin spheres. Our results are obtained by the so-called scalar-relativistic calculation, in which the spin-orbit interaction is neglected. We adopt the generalized gradient approximation<sup>25</sup> for the exchange and correlation term. The unit cell of  $\text{Co}_2(\text{Cr}_{0.6}\text{Fe}_{0.4})\text{Al}$  is a fcc lattice with four atoms. In the ordered  $\text{L2}_1$  structure, each atom of  $\text{Co}_2(\text{Cr}_{0.6}\text{Fe}_{0.4})\text{Al}$  is located at Co=(0,0,0) and  $(\frac{1}{2}, \frac{1}{2}, \frac{1}{2})$ ,  $(\text{Cr}_{0.6}\text{Fe}_{0.4})=(\frac{1}{4}, \frac{1}{4}, \frac{1}{4})$ , Al= $(\frac{3}{4}, \frac{3}{4}, \frac{3}{4})$  in Wyckoff coordinates. The disordered  $\text{Co}_2(\text{Cr}_{0.6}\text{Fe}_{0.4})\text{Al}$  is characterized by the disorder level  $y$ , i.e., the Co–CrFe type disorder corresponds to  $(\text{Co}_{2-y}\text{Cr}_{0.6y}\text{Fe}_{0.4y})[\text{Cr}_{0.6(1-y)}\text{Fe}_{0.4(1-y)}\text{Co}_y]\text{Al}$  and the CrFe–Al type disorder corresponds to  $\text{Co}_2[\text{Cr}_{0.6(1-y)}\text{Fe}_{0.4(1-y)}\text{Al}_y](\text{Al}_{1-y}\text{Cr}_{0.6y}\text{Fe}_{0.4y})$ . For the Brillouin zone (BZ) integration, we have included 256 **k** points in the full BZ.

In Fig. 1, we show the spin polarization as a function of the disorder level  $y$  for the Co–CrFe type disorder and the CrFe–Al type disorder. The spin polarization  $P$  is defined by  $P=(D_\uparrow - D_\downarrow)/(D_\uparrow + D_\downarrow)$ , where  $D_\sigma$  denotes the majority( $\uparrow$ )- and minority( $\downarrow$ )-spin components in the density of states at the Fermi level. It is found that the spin polarization of  $\text{Co}_2(\text{Cr}_{0.6}\text{Fe}_{0.4})\text{Al}$  is 0.90 in the ordered  $\text{L2}_1$

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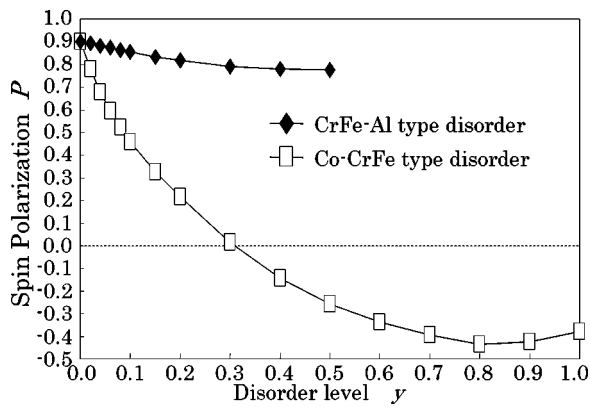


FIG. 1. Spin polarization  $P$  as a function of the disorder level  $y$ . The line with diamond points corresponds to the CrFe–Al type disorder and the line with square points corresponds to the Co–CrFe type disorder of  $\text{Co}_2(\text{Cr}_{0.6}\text{Fe}_{0.4})\text{Al}$ .

structure ( $y=0.0$ ), and 0.77 in the disordered B2 structure ( $y=0.5$ ). The CrFe–Al type disorder slightly reduces the spin polarization of  $\text{Co}_2(\text{Cr}_{0.6}\text{Fe}_{0.4})\text{Al}$ , however,  $\text{Co}_2(\text{Cr}_{0.6}\text{Fe}_{0.4})\text{Al}$  still retains the high spin polarization even in the disordered B2 structure. The reason why the CrFe–Al type disorder does not significantly reduce the spin polarization of  $\text{Co}_2(\text{Cr}_{0.6}\text{Fe}_{0.4})\text{Al}$  can be attributed to the orbital character of the minority-spin states near the Fermi level. It is considered that the minority states close to the Fermi level mainly consist of Co  $3d$  states. In addition, those states arise from Co–Co hybridization<sup>11</sup> and do not couple to Cr  $3d$  and Fe  $3d$  orbitals. Thus, the disorder between  $(\text{Cr}_{0.6}\text{Fe}_{0.4})$  and Al hardly affects this Co–Co interaction, and the minority-spin states near the Fermi level of  $\text{Co}_2(\text{Cr}_{0.6}\text{Fe}_{0.4})\text{Al}$  still have an energy gap even in the disordered B2 structure. The slight decrease in the spin polarization in the disordered B2 structure is mainly due to the life time effects caused by the disorder scattering. This effect broadens the Fe minority conduction bands of  $\text{Co}_2(\text{Cr}_{0.6}\text{Fe}_{0.4})\text{Al}$ , and generates additional minority-spin states at the Fermi level.<sup>26</sup> This leads to a slight decrease in the spin polarization of  $\text{Co}_2(\text{Cr}_{0.6}\text{Fe}_{0.4})\text{Al}$  with the disordered B2 structure.

On the other hand, the spin polarization rapidly decreases with increasing disorder level  $y$  in the Co–CrFe type disorder. In this case, even the disorder level  $y=0.1$  produces a considerable reduction (less than 0.5) in the spin polarization. We consider that the additional minority-spin states at the Fermi level caused by the antisite Co  $3d$  are the main contributor in reducing the spin polarization, as is the disorder between Co and Cr in  $\text{Co}_2\text{CrAl}$ .<sup>26</sup> The energy level of the antisite Co is located in the energy gap of the minority-spin states, and it does not hybridize with the orbital of the first nearest-neighbor ordinary-site Co. Thus, the states are nonbonding and a sharp peak appears in the minority-spin states at the Fermi level, which significantly reduces the spin polarization of the alloy.

As shown in Fig. 2, the total magnetic moment of  $\text{Co}_2(\text{Cr}_{0.6}\text{Fe}_{0.4})\text{Al}$  decreases with increasing  $y$  in the Co–CrFe type disorder ( $\approx 2.7\mu_B$  for  $y=0.8$ ), while it shows a slight decrease in the CrFe–Al type disorder ( $3.8\mu_B \rightarrow 3.7\mu_B$ ) which is consistent with the Slater–Pauling

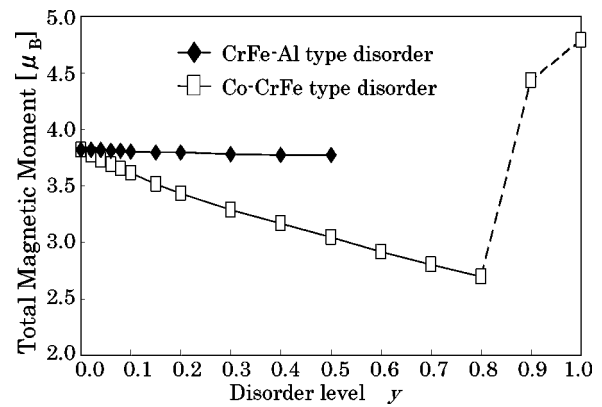


FIG. 2. Total magnetic moment per unit cell in  $\mu_B$  as a function of the disorder level  $y$ . The line with diamond points corresponds to the CrFe–Al type disorder and the line with square points corresponds to the Co–CrFe type disorder of  $\text{Co}_2(\text{Cr}_{0.6}\text{Fe}_{0.4})\text{Al}$ .

behavior<sup>11,21</sup> throughout the entire range of  $y$ . The local magnetic moments per unit cell of  $\text{Co}_2(\text{Cr}_{0.6}\text{Fe}_{0.4})\text{Al}$  in the Co–CrFe type disorder with  $y=0.8$  are  $1.1\mu_B$  (ordinary-site Co),  $1.2\mu_B$  (ordinary-site Cr),  $2.4\mu_B$  (ordinary-site Fe),  $1.5\mu_B$  (antisite Co),  $-1.3\mu_B$  (antisite Cr),  $2.0\mu_B$  (antisite Fe), and  $-0.048\mu_B$  (Al). It was found that the antisite Cr moment antiferromagnetically couples with the ordinary-site Cr moment, which reduces the total magnetic moment of the alloy. The abrupt increase in the total magnetic moment at  $y=0.9$  and  $1.0$  is due to the antisite Cr moment which ferromagnetically couples with the ordinary-site and antisite Co and Fe moments.

The total magnetic moment from the superconducting quantum interference device measurement for  $\text{Co}_2(\text{Cr}_{0.6}\text{Fe}_{0.4})\text{Al}$  is  $3.4\mu_B$  in Ref. 12,  $3.2\mu_B$  in Ref. 22 and  $2.0\mu_B$  in Ref. 19, which are smaller than the theoretical estimates  $\approx 3.8\mu_B$ . Our results in Fig. 2 suggest that the experimentally observed lower total magnetic moment compared with the theoretical prediction might be due to the Co–CrFe type disorder of  $\text{Co}_2(\text{Cr}_{0.6}\text{Fe}_{0.4})\text{Al}$ . If the origin of the decrease of the total magnetic moment in the experimental results is due wholly to the effects of the Co–CrFe type disorder, the observed total magnetic moment  $3.2\mu_B$  in Ref. 22 corresponds to a disorder level  $y=0.3$ – $0.4$  (see Fig. 2). In this case, our results show that the spin polarization  $|P| \approx 0.02$ – $0.2$  (see Fig. 1), and that the  $\text{Co}_2(\text{Cr}_{0.6}\text{Fe}_{0.4})\text{Al}$  is therefore no longer half-metallic. Consequently, it is considered to be important to control the disorder between Co and  $(\text{Cr}_{0.6}\text{Fe}_{0.4})$  in order to obtain highly spin-polarized  $\text{Co}_2(\text{Cr}_{1-x}\text{Fe}_x)\text{Al}$ . In our results, the Co–CrFe type disorder could not explain the total magnetic moment  $2.0\mu_B$  in Ref. 19. Thus, it is believed that surface and interface effects are also important factors in explaining the decrease in the total magnetic moment of  $\text{Co}_2(\text{Cr}_{1-x}\text{Fe}_x)\text{Al}$ .

In conclusion, we have found that the disorder between  $(\text{Cr}_{0.6}\text{Fe}_{0.4})$  and Al does not significantly degrade the spin polarization, while the disorder between Co and  $(\text{Cr}_{0.6}\text{Fe}_{0.4})$  causes a considerable reduction in the spin polarization of  $\text{Co}_2(\text{Cr}_{0.6}\text{Fe}_{0.4})\text{Al}$ . Furthermore, the total magnetic moment of  $\text{Co}_2(\text{Cr}_{0.6}\text{Fe}_{0.4})\text{Al}$  decreases with increasing disorder between Co and  $(\text{Cr}_{0.6}\text{Fe}_{0.4})$  due to the antiferromagnetic cou-

pling of antisite Cr with ordinary-site Cr. We conclude that control of the disorder between Co and ( $\text{Cr}_{0.6}\text{Fe}_{0.4}$ ) is crucial in order to obtain half-metallic full-Heusler alloys  $\text{Co}_2(\text{Cr}_{1-x}\text{Fe}_x)\text{Al}$  that demonstrate good performance as spintronic devices.

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