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MAGNETIC PROPERTIES OF CO-HEUSLER AND RELATED MIXED ALLOYS

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Abstract. Magnetic and crystallographic measurements were made for Co_{2-x}Fe_xMnM (M = Si, Ge, Sn and Ga) and Co_{2-x}Mn_xSi. In Co_{2}MnM, the magnetic moment (\mu_P) in the paramagnetic temperature region is generally much smaller than the ferromagnetic moment (\mu_F) at T = 0 K, which implies that the Co-moment disappears above T_c. The other results are also discussed.

Ferromagnetic Co-Heusler alloys of the type Co_{2}MnM (M = Si, Ge etc.) have been extensively studied by many authors [1-5]. One of the present authors [2] pointed out by the susceptibility measurements of Co_{2}MnM (M = Si, Ge) up to 1300 K that the paramagnetic moment per formula unit, \mu_P, takes about one half value of the ferromagnetic moment per formula unit, \mu_F, where \mu_F = 2S_{Co} + S_{Mn} and \mu_P = 2S\mu_B respectively. In these equations, S_{Co} and S_{Mn} are spin values of Co- and Mn-atoms respectively, and S is an averaged spin expressed by equation S (S + 1) = 2S_{Co} (S_{Co} + 1) + S_{Mn} (S_{Mn} + 1), whose right hand appears in the Curie-Weiss constant of material which has two kinds of magnetic atoms. To confirm and extend the above results, we have studied Co_{2}MnGa, Co_{2-x}Fe_xMnM (M = Si, Ge and Sn) and Co_{2-x}Mn_{1+x}Si in addition to Co_{2}MnM (M = Si, Ge and Sn). The Heusler type (L2_1) alloys in this work were prepared by the sintering method [2], and confirmed to have the good atomic ordering by X-ray diffraction. Saturation magnetization (\sigma_s) versus temperature curves in this work agree well with the data of Webster [1]. The \sigma_s - T curves drop relatively rapidly in the temperature region just below the Curie temperatures (T_c) for M = Si and Ge. The paramagnetic susceptibilities for Co_{2}MnM (M = Si, Ge, Sn) obey well the Curie-Weiss law in the wide temperature regions except for the small parts of the \chi^{-1} - T curves as seen in figure 1. Similar results have been obtained also for Co_{2-x}Fe_xMnM (M = Si, Ge, Sn). As seen in figure 2, the data for x = 2 is somewhat different from the others. The magnetic parameters obtained are summarized in table I and figure 3. Data similar to figure 3 are also obtained for M = Ge and Sn. The values of \mu_F and \mu_P are especially noted. Since the averaged spin S in \mu_P (= 2S) was defined in the equation S (S + 1) = 2S_{Co} (S_{Co} + 1) + S_{Mn} (S_{Mn} + 1), if S_{Co} is not zero, the Mn-moment (= 2S_{Mn}) in the paramagnetic region becomes smaller than the experimental \mu_F in table I and figure 3. As Mn atoms in the Heusler alloys generally have the moment, 3 \sim 4\mu_B, it seems very likely that the Co atoms in Co_{2}MnM lose their magnetic moment in the para-

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Table I. Magnetic and crystallographic data for Co_{2}MnM.

<table>
<thead>
<tr>
<th>Alloy</th>
<th>a (Å)</th>
<th>T_c (K)</th>
<th>\theta_p (K)</th>
<th>\mu_F (µB/f.u.)</th>
<th>\mu_P (µB/f.u.)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Co_{2}MnSi</td>
<td>5.65</td>
<td>1034</td>
<td>1044</td>
<td>5.10</td>
<td>2.03</td>
</tr>
<tr>
<td>Co_{2}MnGe</td>
<td>5.74</td>
<td>905</td>
<td>890</td>
<td>4.66</td>
<td>2.61</td>
</tr>
<tr>
<td>Co_{2}MnSn</td>
<td>6.00</td>
<td>826</td>
<td>870</td>
<td>5.37</td>
<td>3.35</td>
</tr>
<tr>
<td>Co_{2}MnGa</td>
<td>5.77</td>
<td>695</td>
<td>770</td>
<td>4.09</td>
<td>3.28</td>
</tr>
</tbody>
</table>
magnetic temperature region. According to Webster [1], the Co atoms in Co$_2$MnM (M = Si, Ge etc.) have a magnetic moment of 0.75 $\mu_B$ at $T = 0$ K. Therefore the Co-moment would be induced by a molecular field of the Mn-sublattice moment below $T_C$. In figure 3, the $\mu_t$ value coincides with the $\mu_p$ value at $x \approx 1$, which would mean that the condition for the A-site atoms to be magnetically polarized disappears by the change of averaged number of d electrons.

The lattice parameter of Co$_2$MnGa has been measured by X-ray diffraction technique as shown in figure 4. No anomaly was observed at $T_C$, which seems to contradict the polarization of the Co atoms below $T_C$. To investigate the effect of the atomic disorder such as an exchange of atoms between the A site (Co) and the B site (Mn), we prepared Co$_{2-x}$Mn$_{1+x}$Si alloys. When the composition $x$ is positive, the excess Mn atoms will occupy the A site and the situation will be reversed for the negative $x$. Magnetic data for Co$_{2-x}$Mn$_{1+x}$Si are shown in figure 5. Kinks appear at $x = 0$ on the curves of $\mu_t(x)$, $T_C(x)$ and the paramagnetic Curie point $\theta_p(x)$, which would mean that the atomic ordering of Co$_2$MnSi is good and stable. We also found that several Co$_2$MnSi alloys which experienced various heat treatments, have the same magnetization within the limit of error at $T = 77$ K. Therefore the atomic ordering is also stable for temperature variation in Co$_2$MnM. The $\mu_t$ vs. $x$ curve in figure 5 indicates that the excess Co atoms occupy the B-site ferromagnetically to the matrix magnetization and the excess Mn atoms occupy the A-site antiferromagnetically to the matrix magnetization.