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STRUCTURAL AND MAGNETIC PROPERTIES OF Co(AlCr) ALLOYS

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Abstract. — Magnetization, magnetic susceptibility, X-ray and neutron diffraction measurements have been made on CoAl\(_{1-x}\)Cr\(_x\) alloys with \(x \leq 0.6\). Alloys with \(x \geq 0.4\) are two-phase. For the remaining B2 alloys, the Curie temperature and magnetization increase with \(x\) beyond \(x_{\text{crit}} \approx 0.04\). The ratio \(q_c/q_s\) is \(> 1\) suggesting itinerancy.

Introduction

Although the equiatomic CoAl alloy having the ordered B2 (CsCl) structure is paramagnetic, deviations from perfect order occur giving rise to Co atoms on aluminium sites with which local magnetic moments may be associated. Furthermore, the partial substitution of Mn [1] or Ti [2] for Al results in the onset of ferromagnetic order beyond a critical concentration. In this respect the CoAl system resembles the isoelectronic CoGa system for which the effects on the magnetic properties of the partial substitution of a wide range of transition metals (Ti, V, Cr, Mn, Fe, Ni, Cu or Nb) for Ga have been investigated [3] and shown to lead to ferromagnetism at a high enough concentration. The corresponding CoAl systems have not yet been as extensively investigated and the present paper reports on the effects of Cr substitutions for Al in CoAl. The results have relevance to theoretical calculations of impurity moments in CoAl and CoGa [4] and the investigations form part of a general program of research on similar materials which has now been extended to high fields [5].

Experimental techniques

Alloys having the general formula CoAl\(_{1-x}\)Cr\(_x\) were fabricated having values of \(x\) equal to 0.05, 0.10, 0.20, 0.30, 0.40, 0.50 and 0.60 by arc melting of the constituents under a third of an atmosphere of argon. The powdered alloys were heat treated for 24 hours at 825 °C before being quenched into water. Within the limits of detection by X-rays the alloys with \(x \leq 0.30\) were single phase having the B2 ordered structure and the remainder two phase. Neutron diffraction using the LAD instrument at the Rutherford/Appleton Laboratory confirmed these results but information on any L2\(_1\)-type ordering is difficult to obtain in the present case because of the similarity of the Al and Cr scattering lengths. The intensities were consistent with Cr atoms entering Al sites. Magnetization and susceptibility measurements were obtained using a vibrating sample magnetometer (Princeton Applied Research) and a Sucksmith Ring balance in the range 4-1000 K.

Results

The structural studies indicated that the lattice parameters were practically independent of composition at 2.860 ± 0.004 Å for the single phase alloys. All the alloys were ferromagnetic and Curie temperatures \(T_c\) were determined by conventional Arrott plots. Figure 1 shows the variation of the intrinsic magnetizations \(\sigma_{0,0}\) at 4.2 K and the Curie temperatures as a function of \(x\). Both of these peak at \(x \approx 0.35\) corresponding to the observation of two-phase structures within the system for \(x \geq 0.40\). Extrapolation of the magnetization curve indicates a critical concentration for the onset of ferromagnetism of \(x_{\text{crit}} \approx 0.04\). Figure 2 shows the inverse susceptibilities \(\chi^{-1}\) as a function of temperature. These curves are seen to be linear over the major part of the temperature range enabling \(p_{\text{eff}} = g (S (S + 1))^{1/2}\) to be determined. Some preliminary low-field magnetization measurements on the alloy having \(x = 0.05\) have been carried out by Kepa

Fig. 1. — Magnetization \(\sigma_{0,0}\) and Curie temperature \(T_C\) as a function of \(x\) for the series CoAl\(_{1-x}\)Cr\(_x\). The vertical dotted line marks the transition from single-phase to two-phase alloys.

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Table I. — Magnetic parameters for the single phase CoAl$_{1-x}$Cr$_x$ alloys. The zero-field, zero-temperature magnetization $\sigma_{0,0}$, moment per formula unit $\mu_{\text{u.f.}}$, moment per Cr atom $\mu_{\text{Cr}}$, Curie temperature $T_C$, Weiss temperature $\theta_p$, and effective Bohr magneton number $p_{\text{eff}}$ are given. The quantity $q_c$ is defined by the equation $p_{\text{eff}}^2 = q_c (q_c+2)$ and $q_c$ is obtained from $q_c = N g_S \mu_B$ where $N$ is the number of formula units per gram.

<table>
<thead>
<tr>
<th>$x$</th>
<th>$\sigma_{0,0}$ (emu g$^{-1}$)</th>
<th>$\mu_{\text{u.f.}}$ ($\mu_B$)</th>
<th>$\mu_{\text{Cr}}$ ($\mu_B$)</th>
<th>$T_C$ (K)</th>
<th>$\theta_p$ (K)</th>
<th>$p_{\text{eff}}$ ($\mu_B$)</th>
<th>$q_c$ ($\mu_B$)</th>
<th>$q_c/q_s$</th>
</tr>
</thead>
<tbody>
<tr>
<td>0.05</td>
<td>-</td>
<td>-</td>
<td>-</td>
<td>&lt; 10</td>
<td>50</td>
<td>1.06</td>
<td>0.43</td>
<td>-</td>
</tr>
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<td>0.10</td>
<td>11.1</td>
<td>0.17</td>
<td>1.7</td>
<td>144</td>
<td>220</td>
<td>1.06</td>
<td>0.43</td>
<td>2.5</td>
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<tr>
<td>0.20</td>
<td>26.5</td>
<td>0.43</td>
<td>2.2</td>
<td>225</td>
<td>280</td>
<td>1.38</td>
<td>0.54</td>
<td>1.3</td>
</tr>
<tr>
<td>0.30</td>
<td>39.1</td>
<td>0.65</td>
<td>2.2</td>
<td>290</td>
<td>315</td>
<td>1.58</td>
<td>0.61</td>
<td>1.0</td>
</tr>
</tbody>
</table>

Fig. 2. — Inverse susceptibility $\chi^{-1}$ against temperature for the series of CoAl$_{1-x}$Cr$_x$ alloys. Parameters deduced from these curves are given in table I.

[6] using a VSM in fields of 5 Oe. The zero-field cooled magnetization indicated a transition temperature at $\sim 15$ K. The thermoremanent magnetization suggested spin glass behaviour up to $\sim 100$ K.

Discussion and conclusions

The alloys from this system have the B2 CsCl structure for $x \leq 0.3$ and are two-phase for larger $x$. The independence of lattice parameter on composition contrasts with the effects of Ti substitutions for which a sharp increase is observed as the larger Ti atom is substituted [2]. Magnetization data indicate the onset of ferromagnetism in the Cr system to occur at $x \sim 0.04$. The results for the corresponding Ti and Mn systems indicate critical concentrations of $x = 0.4$ and $\sim 0.03$ respectively. The critical concentrations for Mn and Cr are very similar to those observed for the isostructural CoGa systems for which the critical values are $x \sim 0.05$ (Cr), $\sim 0.04$ (Mn) and 0.25 (Ti). Until the site occupation is known little can be put forward by way of explanation for the disparity between the two Ti concentrations but it is possible that differing numbers of antistructure atoms are partly responsible. Results for the vanadium substituted alloys are awaited with interest. Table I lists $\sigma_{0,0}$, the magnetic moments per formula unit $p$ deduced from $\sigma_{0,0}$ and the effective Bohr Magneton numbers $p_{\text{eff}}$ for the CoAl$_{1-x}$Cr$_x$ alloys. The slope of the inverse susceptibility curve is approximately identical for $x = 0.05$ and 0.1 but decreases at higher values of $x$ leading to a larger $p_{\text{eff}}$. A comparison of the ferromagnetic and paramagnetic moments for each composition shows that the two quantities are not identical except for $x = 0.3$. This inequality is usually expressed in terms of the Rhodes-Wohlfarth ratio $q_c/q_s$ [7] where $q_c$ is defined by $p_{\text{eff}}^2 = q_c (q_c+2)$ and $q_s$ is equal to 25, where $S$ is obtained from $\sigma_{0,0} = N g_S \mu_B$, $N$ being the number of formula units per gram. The disparity between the para- and ferromagnetic moments is also suggestive of an itinerant magnetic structure [8]. Ab initio self-consistent calculations of the electronic structure of 3d impurities substituted into CoAl and CoGa in the framework of the density functional theory using local spin density approximation have suggested a moment of 2.47 $\mu_B$ for Cr atoms on Al sites [4]. This is reasonably consistent with the moments given in table I if it is assumed that the Co atoms have a negligible moment. Finally, the preliminary low-field magnetization results on CoAl$_{0.95}$Cr$_{0.05}$ suggest there may be some clustering effects at low concentrations in this system.