FERROMAGNETIC-SPIN GLASS TRANSITION IN AMORPHOUS AND CRYSTALLINE Fe-Cr

Y. Yeshurun, K. V. Rao and M. B. Salamon

Department of Physics, University of Illinois at Urbana-Champaign, Urbana, IL 61801, USA

and

H. S. Chen

Bell Laboratories
Murray Hill, NJ 07974

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Through a direct comparison of experimental results in amorphous and crystalline forms of Fe-Cr alloys we examine the effect of topological and compositional disorder on magnetic properties. Both types of disorder result in a decrease in the Curie temperature, the magnetic moment at $T = 0$ and the exchange stiffness constant. These results are discussed in the framework of several models.
Fig. 1. Low field $\chi$ susceptibility versus temperature for $(\text{Fe}_{0.70}\text{Cr}_{0.30})_{75}\text{P}_{16}\text{B}_{6}\text{Al}_{3}$.

Fig. 2. Magnetization versus temperature with different external fields for $(\text{Fe}_{0.70}\text{Cr}_{0.30})_{75}\text{P}_{16}\text{B}_{6}\text{Al}_{3}$. Arrows indicate the maximum magnetization.
Fig. 3. Phase diagram for amorphous (a) and crystalline (c) Fe-Cr alloys. Amorphous phases describe present results while crystalline phases are the result of neutron scattering experiments of Ref. 8. For the sake of simplicity we plot only the para-ferromagnetic and the FM-SG lines which are relevant to this paper. The squares denote the temperature of the drop in the ac signal (a alloys) and of the minimum in the neutron scattering intensity curve (c alloys).

To further investigate the differences between a and c systems we derive the stiffness constant from the temperature dependence of the magnetization. Bloch's law

\[ M(T) = M_0 \left[ 1 - b \frac{T}{T_c}^{3/2} \right] \]

is found to hold for the present amorphous samples with \( x \geq 0.70 \), indicating that long-wavelength spin-wave excitations determine the magnetization behaviour. The existence of spin waves in non crystalline ferromagnets has been demonstrated by many experiments\textsuperscript{12-15} and in theoretical discussions\textsuperscript{15-16}. We fit \( M(T) \), (measured at 10 kG) for \( x \geq 0.85 \) from 4.2 K to room temperature \( (T/T_c \geq 0.6) \) and for \( x = 0.80 \) and \( 0.70 \) from \( T_g \) up to and even above \( T_c \). Potted data for \( x = 0.70 \) is shown in Fig. 4. In Table I we summarize the values of \( M_0 \) and \( b \) for the present alloys as well as for pure a and c Fe\((x = 1)\), quoted from Ref. 12. The exceptionally good fit to a \( T^{3/2} \) law, (Fig. 4) although known to hold experimentally in many systems,\textsuperscript{13,14,18,19} is still a surprising result. Neutron scattering data\textsuperscript{8} on the crystalline analog exhibit a saturation value of \( D \) for \( x = 0.34 \) but a broad maximum for \( x = 0.26 \). A temperature independent value for \( b \) is expected in the regime where \( D \) saturates. The present high field data show that \( b \) is temperature independent even for alloys in which \( D \) has a maximum. One may attempt to find the origin for this disagreement in structural differences. However, recent neutron scattering experiments\textsuperscript{20} on amorphous \((\text{Fe}_{x} \text{Ni}_{1-x})\text{P}_{16}\text{B}_{6}\text{Al}_{3}\) yield a maximum in \( D(T) \) as in the crystalline case. We conclude that the changes in \( D \) are smaller in the present system although this may also indicate that external field causes qualitative changes in the spin-wave spectrum. Samples with compositions closer to the MCP point must be studied to clarify this point.

It is well known\textsuperscript{21} that

\[ D = \left( \frac{k_B}{4\pi} \right) \left( \frac{2.612 \, \text{g} \, \mu_B}{\text{b}} \right)^{2/3} (M_0 b)^{-2/3}. \]

Values of \( D \) as deduced from Eq. (2) are summarized in Table I and plotted in Fig. 5, together with \( D \)-values for the c system to show that \( D(x) \) can be described by a linear function which extrapolates to zero at \( x \approx X_{\text{MCP}} \). A zero value of \( D \) signals an instability of the FM phase\textsuperscript{22}. Note a similar linear decrease of \( D(x) \) in Ref. 11. Very similar results were reported recently\textsuperscript{14} for amorphous Co-Ni systems.
The experimental results described above (Table 1 and Figs. 3, 5) show clearly a reduction of $T_c$, $M_0$, and $D$ with the introduction of (i) compositional disorder (replacing Fe by Cr) and of (ii) structural disorder. In the following we discuss these results in terms of several models.

The effect of compositional disorder in both amorphous and crystalline alloys has been discussed in terms of electronic band filling, using a Slater-Pauling-type curve. In this model, the additional Cr causes a reduction in the average magnetic moment per atom and in the exchange interaction (and thus in $T_c$ and $D$ which are proportional to it). Calculations in this framework suggest antiferromagnetic (AFM) coupling between Cr atoms and the iron matrix. A different approach focuses on the competition between FM and AFM interactions to account for the changes in the magnetic properties. This later approach is also successful in predicting a magnetic phase diagram of the type exhibited in Fig. 3.

To explain the effect of structural disorder one may take the same two approaches. In the first one, as proposed by Mizoguchi et al., the changes in magnetic properties of the amor-
Table I.

<table>
<thead>
<tr>
<th>Fe Concentration x</th>
<th>T&lt;sub&gt;c&lt;/sub&gt; (K)</th>
<th>M&lt;sub&gt;s&lt;/sub&gt; (Gauss)</th>
<th>b·10&lt;sup&gt;-6&lt;/sup&gt;·K&lt;sup&gt;-3/2&lt;/sup&gt;</th>
<th>D&lt;sub&gt;meas&lt;/sub&gt; (MeV·Å&lt;sup&gt;2&lt;/sup&gt;)</th>
<th>D&lt;sub&gt;calc&lt;/sub&gt; (meV·Å&lt;sup&gt;2&lt;/sup&gt;)</th>
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</thead>
<tbody>
<tr>
<td>0.26 (α)&lt;sup&gt;a&lt;/sup&gt;</td>
<td>178</td>
<td>-</td>
<td>-</td>
<td>25&lt;sup&gt;b&lt;/sup&gt;</td>
<td>-</td>
</tr>
<tr>
<td>0.34 (α)&lt;sup&gt;a&lt;/sup&gt;</td>
<td>330</td>
<td>-</td>
<td>-</td>
<td>60</td>
<td>-</td>
</tr>
<tr>
<td>1.00 (α)&lt;sup&gt;c&lt;/sup&gt;</td>
<td>1042</td>
<td>1752</td>
<td>3.4</td>
<td>281</td>
<td>285</td>
</tr>
<tr>
<td>0.70 (α)</td>
<td>85</td>
<td>365</td>
<td>490</td>
<td>-</td>
<td>29</td>
</tr>
<tr>
<td>0.80 (α)</td>
<td>265</td>
<td>560</td>
<td>130</td>
<td>-</td>
<td>53</td>
</tr>
<tr>
<td>0.90 (α)</td>
<td>440&lt;sup&gt;d&lt;/sup&gt;</td>
<td>1070</td>
<td>60</td>
<td>-</td>
<td>58</td>
</tr>
<tr>
<td>0.95 (α)</td>
<td>522&lt;sup&gt;d&lt;/sup&gt;</td>
<td>1035</td>
<td>40</td>
<td>-</td>
<td>78</td>
</tr>
<tr>
<td>1.00 (α)&lt;sup&gt;c&lt;/sup&gt;</td>
<td>630</td>
<td>1200</td>
<td>18.6</td>
<td>134</td>
<td>117</td>
</tr>
</tbody>
</table>

<sup>a</sup> From Ref. 8.
<sup>b</sup> A maximum value.
<sup>c</sup> From Ref. 10.
<sup>d</sup> T<sub>c</sub> results from Ref. 27.

Spin waves and magnetization parameters for amorphous (α) and crystalline (α) Fe-Cr ferromagnets. D<sub>meas</sub> is the stiffness constant as measured in neutron scattering. D<sub>calc</sub> is the stiffness constant calculated from measured M<sub>s</sub> and b (Eq. 2). M<sub>s</sub> differs from the apparent magnetization at zero field due to deviation from Bloch's law. See text.

The effect of randomness in the exchange interaction is discussed also by Kirkpatrick and Sherrington (KS) in their model for spin glasses. In this model the exchange interaction has a Gaussian distribution with average at J<sub>0</sub> and width of J. This model predicts a decrease of the magnetization as T = 0.

\[
m_0 \sim 1 - \left(\frac{2}{\pi}\right)^{1/2} \frac{J}{J_0} \exp\left(-\frac{J}{j_j_0}^2\right) (4)
\]

for J<sub>0</sub> << J, and vanishing of m<sub>0</sub> for J/J<sub>0</sub> \(\approx (2/\pi)^{1/2}\) as

\[
m_0 \sim \frac{(j_j_0)^2}{(2/\pi)^{1/2}} - \frac{J}{j_j_0}^{1/2} (5)
\]

The main conclusion drawn from Eqs. 4, 5 is that m<sub>0</sub> decreases with increasing randomness. Comparing this result with the experimental results (Table I) we reach the same conclusion as in the previous paragraph, namely that randomness of the exchange interactions increases as a result of structural as well as compositional disorder. Comparing the relative changes of T<sub>c</sub> = J<sub>0</sub>, M<sub>s</sub> and b we conclude that structural disorder makes a significant contribution to the changes of the magnetic properties. In view of this conclusion it is possible to under-
stand the relative high value $x^{\text{MC}} = 0.65$ compared with $x^{\text{MC}} = 0.20$. However, it is difficult to see why the MCP temperature is higher in the \( \alpha \) alloys than in the \( \alpha \) system. In the KS model $J_0 = J$ at the MCP and this means that the width $J^\text{C} (=60 K) > J^\text{D} (=20 K)$. We suggest that in the amorphous system where the width is a result of composition and structural disorder, to a first approximation

$$J = J^\text{C} + J^\text{D} \tag{6}$$

where $J^\text{C}$ is a rapidly decreasing function of $x$ (with $J^\text{C} = 60 K$ at $x = 0.20$) and $J^\text{D}$ is approximately constant in view of the $x$ independence of the amorphous structure. Since for the amorphous system the width of the distribution equals 20 K at $x = 0.65$ we can set an upper limit of 20 K on $J^\text{D}$. In view of the number of parameters involved in determining $J(x)$ it is clear that more, independent (e.g. Mössbauer) experimental data are necessary to support this simple model.

In conclusion, we have shown that the Curie temperature, the magnetic moment and the stiffness constant are reduced as a result of induced compositional and structural disorder. The effect of both types of disorder on the magnetic properties may be interpreted in terms of electron transfer and band fillings as well as in terms of distribution of the exchange interaction. The first approach accounts for the main magnetic changes, however not for the more subtle ones. The second approach ignores any possible chemical effects but still accounts qualitatively for experimental observations.

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References

17. Slight deviations from spin wave theory are observed below $T_f$. Several explanations have been given to similar phenomena. See e.g. Ref. 14.