FERROMAGNETIC-SPIN GLASS TRANSITION IN AMORPHOUS AND CRYSTALLINE Fe-Cr[*]

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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.

Ferromagnetic (FM) – spin glass (SG) transitions -- which are predicted by various theoretical models[1-2] -- have now been found in several magnetic systems.[3-8] In recent[8] elastic neutron scattering studies of crystalline Fe_xCr_{1-x} alloys, spin wave excitations served to probe FM-SG transitions. We present here magnetic measurements on amorphous $(Fe_xCr_{1-x})_{75}P_{16}B_6Al_3$ alloys which also exhibit FM-SG transitions. The magnetic species in this system are Fe and Cr -- P, B and Al serve to stabilize the amorphous phase. Thus this system can be considered as the amorphous analog of Fe_xCr_{1-x}. The present work presents the phase diagram and an interpretation of the low temperature magnetization in this amorphous system in terms of a spin-wave model. This is the first direct comparison between the crystalline and amorphous magnetic analogs where both systems exhibit the FM-SG transition.

Amorphous Fe-Cr alloys were prepared by centrifugal spin quenching.[9] Small ribbons (5 mm x 1 mm x 25 μm) were used in a low field ac susceptibility bridge and in a vibrating sample magnetometer. In Fig. 1 we present the temperature dependence of the ac susceptibility (measured at ac fields of ≈3 Oe rms) for x=.70. This Figure exhibits the typical behaviour of a reentrant ferromagnet with a low temperature transition to the spin-glass phase.[3,7] The sharp rise at $T_c \simeq 85$ K marks the Curie temperature while the sharp drop at $T_{fg} \simeq 20$ K, the freezing temperature. In Fig. 2 we present the temperature dependence of the magnetization in applied field (50 G ~ 10 kG) for the x = .70 alloy. The mag-

netization data exhibit a broad maximum at low fields. The maximum is pushed to lower temperatures with increasing external fields. This behaviour is typical of materials that undergo the FM-SG transition (compare the experimental results of Refs. 6 and 7). The magnetization data were processed by using a scaling technique. The resultant critical exponents are similiar to those described in Ref. 7. The Curie temperatures are in fair agreement with the ac susceptibility results. The same technique was also applied to the FM-SG transitions. In this case it seems that the true transition temperatures lies below the temperature of the drop in the ac signal, as in Ref. 6. However, the present results are much less reliable due to the disappearance of the maximum at quite low fields. Demagnetization and anisotropy fields can not be neglected in the scaling of data taken at such low fields.[6] In Fig. 3 we present a magnetic phase diagram for amorphous Fe-Cr, and, in the same Figure, the phase diagram for the crystalline materials.[8] (Amorphous phases are denoted by ã and crystalline phases by c̃). The most striking feature of Fig. 3 is the large separation between amorphous phase boundaries in the amorphous and crystalline states. Similiar results were described for amorphous $(Fe_xCr_{1-x})_{80}B_{10}P_{10}$[10] and amorphous $(Fe_xCr_{1-x})_{80}P_{13}C_7$.[11] Extrapolation of those results suggests a vanishing of the magnetization at x ≃ 0.6. The multicritical point (MCP) for the present amorphous alloys is at x ≃ .65 and T ≃ 25 K, while for the crystalline alloys it is at x ≃ .20 and T ≃ 60 K. Note, however that paramagnetic-ferromagnetic (PM-FM) lines have the same slopes over a wide range of temperature. Note also that the squares in Fig. 3 do not denote a sharp phase transition but rather a drop in the ac-susceptibility signal (ã curve) and a minimum in the neutron scattering intensity (c̃ curve).
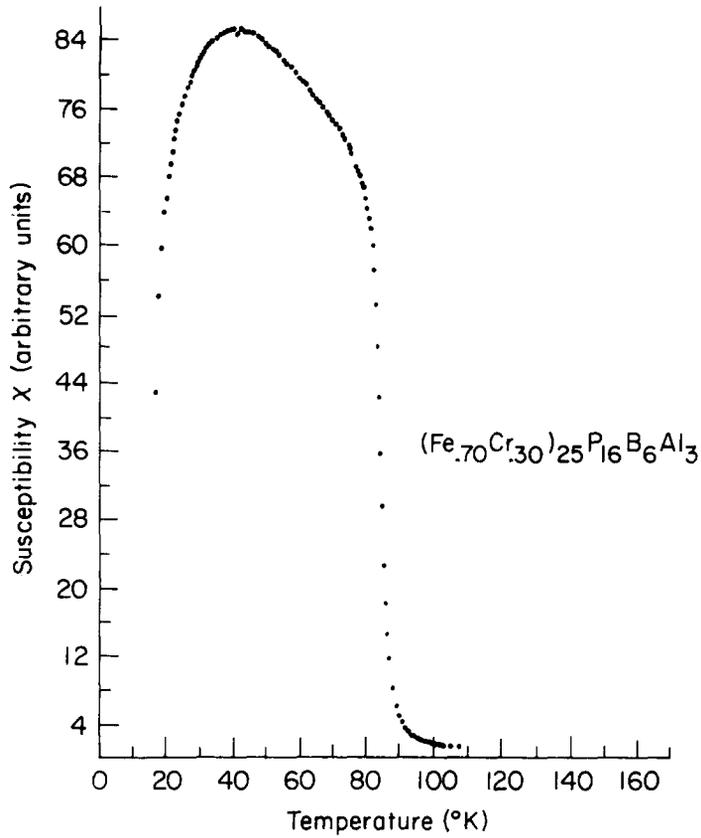
Fig. 1.   Low field ac susceptibility versus tem-
perature for $(Fe_{.70}Cr_{.30})_{75}P_{16}B_6Al_3$.
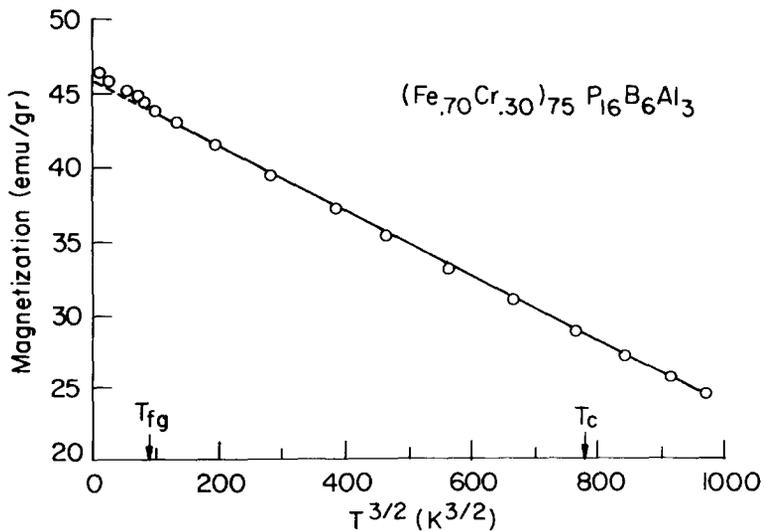

Fig. 2.   Magnetization versus temperature with
different external fields for
$(Fe_{.70}Cr_{.30})_{75}P_{16}B_6Al_3$.   Arrows indicate
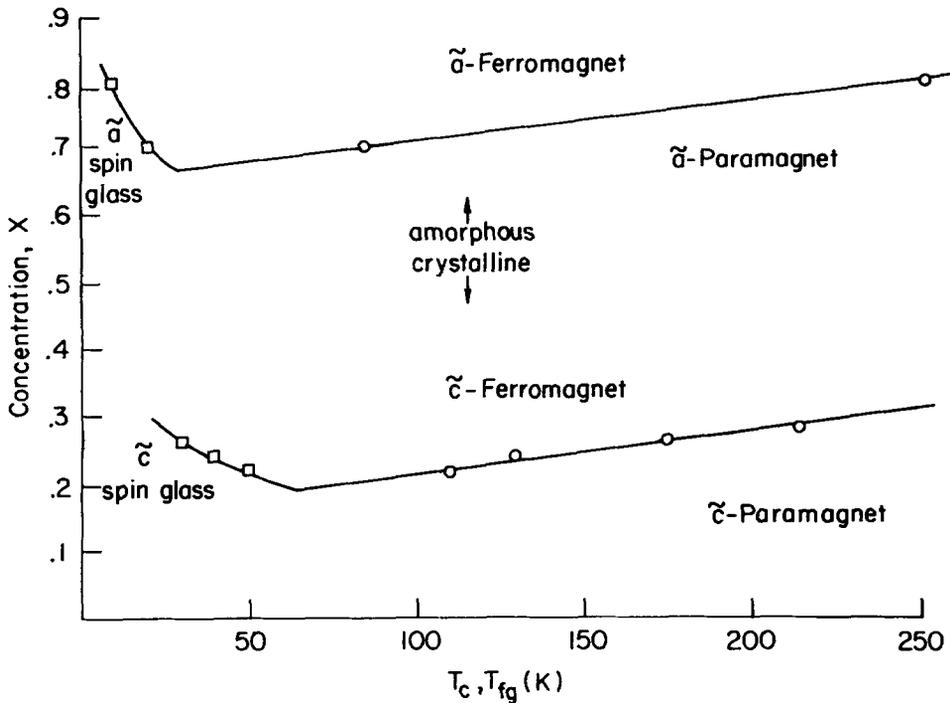the maximum magnetization.

Fig. 3. Phase diagram for amorphous (ã) and
crystalline (c̃) 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 temper-
ature of the drop in the ac signal (ã
alloys) and of the minimum in the neu-
tron scattering intensity curve (c̃
alloys).

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

$$M(T) = M_o [1 - b \, T^{3/2}] \qquad (1)$$

is found to hold for the present amorphous
samples with $x \geq .70$, 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[12-15] and in
theoretical discussions.[15-16] We fit M(T),
(measured at 10 kG) for $x \geq .85$ from 4.2 K to
room temperature $(T/T_c \geq .6)$ and for $x = .80$ and
.70 from T_{fg}[17] up to and even above T_c. Fitted
data for $x = .70$ is shown in Fig. 4. In Table I
we summarize the values of M_o and b for the
present alloys as well as for pure ã and c̃
Fe(x = 1), quoted from Ref. 12. The exception-
ally good fit to a $T^{3/2}$ law, (Fig. 4) although
known to hold experimentally in many sys-
tems,[13,14,18,19] is still a surprising result.
Neutron scattering data[8] on the crystalline
analog exhibit a saturation value of D for
$x = .34$ but a broad maximum for $x = .26$. A tem-

perature 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[20]
on amorphous $(Fe_x Ni_{1-x})_{75} P_{16} B_6 Al_3$ yield a max-
imum in D(T) as in the crystalline[8] case. We
conclude that the changes in D are smaller in
the present system although this may also indi-
cate 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[21] that

$$D = (k_B/4\pi)(2.612 \, g\mu_B)^{2/3} (M_o b)^{-2/3}. \qquad (2)$$

Values of D as deduced from Eq. (2) are summar-
ized in Table 1 and plotted in Fig. 5, together
with D-values for the c̃ system, to show that
D(x) can be described by a linear function which
extrapolates to zero at $x \leq x_{mcp}$. A zero value
of D signals an instability of the FM phase.[22]
Note a similar linear decrease of D(x) in Ref.
11. Very similar results were reported recently[1 4]
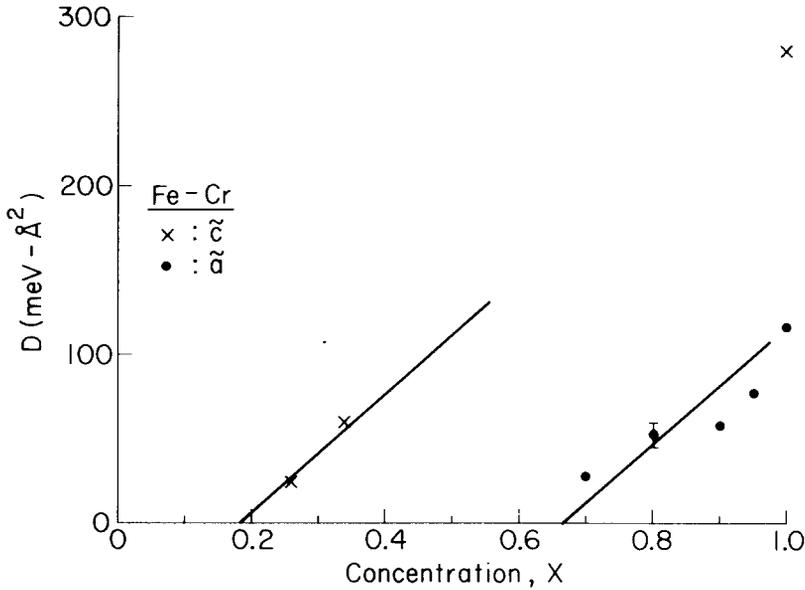for amorphous Co-Ni systems.

Fig. 4. $T^{3/2}$ fit to M(T) for
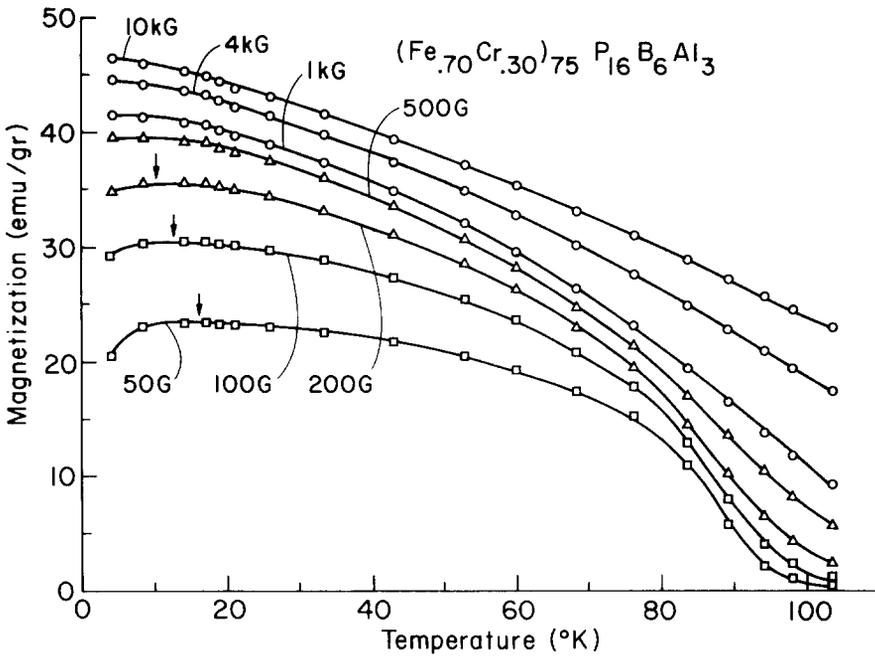$(Fe_{.70}Cr_{.30})_{75}P_{16}B_6Al_3$.

Fig. 5.   Stiffness constant D vs Fe concentration
x in amorphous and crystalline Fe-Cr.

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) com-
positional 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,[10]
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) cou-
pling between Cr atoms and the iron matrix.   A
different approach focusses on the competition
between FM and AFM interactions to account for
the changes in the magnetic properties.   This
later approach[1,2] is also successful in pre-
dicting a magnetic phase diagram of the type
exhibited in Fig. 3.

To explain the effect of structural dis-
order one may take the same two approaches.   In
the first one, as proposed by Mizoguchi et al,[10]
the changes in magnetic properties of the amor-

Table I.

Spin waves and magnetization parameters for amorphous (ã) and crystalline (c̃) Fe-Cr ferromagnets. D_{meas} is the stiffness constant as measured in neutron scattering. D_{calc} is the stiffness constant calculated from measured M_0 and b (Eq. 2). M_0 differs from the apparent magnetization at 0K due to deviation from Bloch's law. See text.

Fe Concentration x	T_c (K)	M_0 (Gauss)	b $10^{-6}K^{-3/2}$	D_{meas} (MeVA^2)	D_{calc} (meVA^2)
0.26 (c̃)[a]	178	–	–	25[b]	–
0.34 (c̃)[a]	330	–	–	60	–
1.00 (c̃)[c]	1042	1752	3.4	281	285
0.70 (ã)	85	365	490	–	29
0.80 (ã)	265	560	130	–	53
0.90 (ã)	440[d]	1070	60	–	58
0.95 (ã)	522[d]	1035	40	–	78
1.00 (ã)[c]	630	1200	18.6	134	117

a.   From Ref. 8.

b.   A maximum value.

c.   From Ref. 10.

d. T_c results from Ref. 27.

phous alloys are due to electron transfer from metalloid atoms to the holes of 3d metal atoms. The band filling causes, as above, a reduction in the average magnetic moment. This reduction is a function of the metalloid and its concentration in the alloy. For constant metalloid conditions (25 at% of P-B-Al in the present case) one expects a parallel shift of the crystalline Slater-Pauling curve as a function of the iron concentration. This shift can explain a reduction in the magnetic moment in the amorphous alloys as well as a disappearance of spontaneous magnetization at higher concentration of Fe as compared to crystalline Fe-Cr. Indeed the magnetic moment curve of amorphous Fe-Cr does lie below the crystalline curve (Table 1 and Ref. 10) but decreases much more rapidly with Cr addition than in crystalline alloy. A simple transfer model would yield a parallel shift only. To account for the additional reduction it is essential to consider the random nature of the magnetic interactions in amorphous solids. This kind of approach was first introduced by Gubanov[23] to explain ferromagnetism in amorphous materials. In his model random variation in the Heisenberg coupling constant J induces a reduction of T_c and of M_0. A simple expression for the reduced magnetization as a function of $\delta^2 = <\Delta J^2>/<J>^2$ was derived by Handrich:[24]

$$\sigma(T) = 0.5 \{B_s[(1+\delta)y] + B_s[(1-\delta)y]\} \quad (3)$$

where B_s is the Brillouin function and y = $[3\sigma(T)/(S+1)]T_c/T$. Numerical calculations of Eq. 3 show clearly that $\sigma(T)$ decreases with increasing structural randomness (see Fig. 1 of Ref. 24). According to this model a decrease of

b in Eq. 1 means a decrease in δ. On the Fe-rich side δ is smaller and increases towards the MCP. Also, δ is smaller in the c̃ alloys due to lack of structural disorder. Although this simple model suffers from intrinsic defects, (e.g. the Curie temperature does not depend on structure fluctuations[24]) it serves quite well to demonstrate the importance of structural disorder (see e.g. Ref. 25).

The effect of randomness in the exchange interaction is discussed also by Kirkpatrick and Sherrington (KS)[1] in their model for spin glasses. In this model the exchange interaction has a Gaussian distribution with average at $\tilde{J}_0$ and width of $\tilde{J}$. This model predicts a decrease of the magnetization at T = 0.

$$m_0 \sim 1 - (2/\pi)^{1/2} \tilde{J}/\tilde{J}_0 \exp (-\tilde{J}_0^2/2\tilde{J}^2) \quad (4)$$

for $\tilde{J}_0 \gg \tilde{J}$, and vanishing of m_0 for $J/J_0 \gtrsim (2/\pi)^{1/2}$ as

$$m_0 \sim (\tilde{J}/\tilde{J}_0)^2 [(2/\pi)^{1/2} - \tilde{J}/\tilde{J}_0]^{1/2} \quad (5)$$

The main conclusion drawn from Eqs. 4, 5 is that m_0 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 $T_c = \tilde{J}_0$, M_0 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^a_{mcp} = .65 compared with x^c_{mcp} = .20. However, it is difficult to see why the MCP temperature is higher in the $\tilde{c}$ alloys than in the $\tilde{a}$ system. In the KS model $\tilde{J}_o = \tilde{J}$ at the MCP and this means that the width $\tilde{J}^{\tilde{c}}$ (=60 K) > $\tilde{J}^{\tilde{a}}$ (=20 K). We suggest that in the amorphous system where the width is a result of composition and structural disorder, to a first approximation

$$\tilde{J} = \tilde{J}^{\tilde{c}} + \tilde{J}^{\tilde{a}} \qquad (6)$$

where $\tilde{J}^{\tilde{c}}$ is a rapidly decreasing function of x (with $\tilde{J}^{\tilde{c}}$ = 60 K at x = .20) and $\tilde{J}^{\tilde{a}}$ is approximately constant in view of the x independence of the amorphous structure.[26] Since for the amorphous system the width of the distribution equals 20 K at x = .65 we can set an upper limit of 20 K on $\tilde{J}^{\tilde{a}}$. 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 quantitatively for experimental observations.

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