CONF-831187--10

October 1983

BNL--33827

DE84 002874

BNL--33827

DE84 002874

INVESTIGATION OF THE FERROMAGNETIC-SPIN GLASS TRANSITION IN a-$(Fe_{77}Cr_{23})_{75}P_{16}B_6Al_3$

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The submitted manuscript has been authored under contract DE-AC02-76CH00016 with the Division of Basic Energy Sciences, U.S. Department of Energy.

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Abstract

Elastic and inelastic neutron scattering studies were performed on the amorphous alloy $(Fe_{77}Cr_{23})_{75}P_{16}B_6Al_3$. These measurements were conducted over a temperature range from 1.8 K to room temperature. The elastic scattering results obtained at $Q = 0.025Å^{-1}$ indicated a Curie temperature of ~ 185 K in addition to a ferromagnetic-spin glass transition below 20 K. These values are consistent with the phase diagram construction resulting from the magnetization and ac susceptibility measurements of Yeshurun et al. Inelastic neutron scattering measurements resulted in well-defined spin waves being observed in the ferromagnetic phase. For the small Q range studied ($0.05Å^{-1}$ to $0.1Å^{-1}$), the spin wave energies followed a quadratic dispersion. As the temperature was lowered towards the spin glass regime, the spin wave energies decreased indicating a softening of the magnetic stiffness constant. Below 10 K, no well-defined spin wave excitations were present; however, a quasielastic peak, which increased slightly as the temperature was lowered, was observed.

Several magnetic systems have recently been studied which exhibit ferromagnetic ordering followed at lower temperatures by a spin glass-like state. These investigations were originally motivated by theories which predicted a sequence of phases based on calculations of an Ising system with infinite range random interaction.[1] This spin glass state is characterized by a random freezing of magnetic moments due to either competing magnetic interactions or percolation effects. There is still some debate, however, as to whether this state is a true thermodynamically stable phase as formulated by Edwards and Anderson[2] or a gradual freezing of spins.[3] Examples of materials which indicate a ferromagnetic to spin glass behavior include the crystalline alloys $(PdFe)_{1-x} Mn_x$,[4] $Fe_x Cr_{1-x}$,[5] amorphous alloys $(Fe_{1-x}Mn_x)_{75} P_{16}B_6Al_3$,[6] $(Fe_{1-x} Ni_x)_{75}P_{16}B_6Al_3$[7] and the ionic solid $Eu_{1-x}Sr_xS$.[8]

Neutron scattering is an ideal probe in studying the dynamics of the spin glass state. This is accomplished by following the evolution of spin waves from the ferromagnetic phase to the spin glass phase. Such investigations have been performed on polycrystalline $Fe_x Cr_{1-x}$[9] and amorphous $(Fe_{1-x} Mn_x)_{75} P_{16}B_6Al_3$[10] and $(Fe_{1-x}Ni_x)_{75} P_{16}B_6Al_3$.[11] The goal of the present work is to use this procedure in order to study the amorphous counterpart of Fe_xCr_{1-x}. Magnetization and ac susceptibility experiments have recently been conducted by Yeshurun et al.[12] on amorphous $(Fe_xCr_{1-x})_{75} P_{16}B_6Al_3$ resulting in a phase diagram for this amorphous alloy. This diagram shows a reentrant spin glass region for $x \geq 0.65$ with an extracted upper limit on x of about 0.9. It should be noted that earlier neutron scattering and magnetization experiments [13] have been performed on the amorphous $Fe_{70}Cr_{10} P_{13}C_7$ alloy where this iron concentration ($x > 0.9$) falls outside the spin glass region and thus only ferromagnetic behavior was observed.

Both elastic and inelastic neutron scattering measurements were performed using the H-9 cold source spectrometer at the Brookhaven National Laboratory's High Flux Beam Reactor. This triple axis spectrometer is equipped with a double crystal monochromator allowing incident energies < 5 meV to be used. A liquid nitrogen cooled Beryllium filter was needed to eliminate higher order contamination. Pyrolytic graphite (002) monochromators and analyzers were used with the analyzer fixed at E_F = 4.5 meV. The collimation of the system was 30'-40'-30'-20'-20'. The resulting energy resolution for zero energy transfer was 0.09 meV (FWHM). Since the material was amorphous and thus lacked translational order, all measurements were conducted near the forward direction, i.e. the (000) "Bragg Peak". The amorphous ribbon samples used in our experiments were prepared by the centrifugal spinning technique.[14] Approximately 5 grams of the sample were packed into a He-filled aluminum sample holder which was mounted on the cold finger of a flow cryostat or a closed cycle displex refrigerator. The temperature range covered 1.8 K to 300 K with regulation better than 0.2 K.

The temperature variation of the intensity measured at Q = $0.025Å^{-1}$, with the triple axis spectrometer set for zero energy transfer, is given in Fig. 1. The rounded peak corresponds to the Curie temperature of the sample which is approximately 184 ± 2 K. The scattering intensity, below T_C, decreased linearly with decreasing temperature which is expected for ferromagnetic materials. However, around 50 K, the intensity decreased less rapidly until it reached a minimum between 10 K and 20 K after which it increased slightly at lower temperature. The temperature at which this minimum occurs suggests the ferromagnet - spin glass phase boundary. This temperature, in addition to the value of the Curie temperature given above, is consistent with the phase diagram constructed by Yeshurun et al.[12] for this concentration of Fe (x = 0.77).

Inelastic constant Q scans were performed for Q values ranging from 0.05Å^{-1} to 0.1Å^{-1}. Fig. 2 shows the observed spectra measured at Q = 0.07Å^{-1} with E_F = 4.5 meV. As the temperature decreased below T_C, spin waves appeared shifting to higher energies which is normal for conventional ferromagnets. However, below 50 K, the spin wave peaks started to decrease in energy with a corresponding increase in their linewidths. For temperatures less than 10 K, no propagating spin waves were observed. Instead, a quasielastic peak was observed which increased slightly while the temperature was lowered. This is clearly seen in Fig. 2. The inelastic scattering at T = 10 K is larger while its peak at zero energy transfer is smaller when compared to the data obtained at T = 1.82 K (broken curve). A constant background which is energy independent has been subtracted from spectra obtained at temperatures of 30 K, 10 K, 1 82 K.

The data were analyzed using the scattering cross section for spin waves in a ferromagnet [15] plus an elastic scattering cross section contribution:

$$\frac{d^2\sigma}{d\Omega d\omega} = A_S \frac{k_f}{k_i} \left[\frac{\hbar\omega/kT}{1-\exp(-\hbar\omega/kT)}\right] \frac{F_S(Q,\omega)}{Q^2} + \frac{A_G}{Q^2} \delta(\omega) \qquad (1)$$

where A_G and A_S are the integrated intensities of the elastic part and the spin wave part, respectively. $F_S(Q,\omega)$ is the spectral weight function which we chose to be of the form of a double Lorentzian:

$$F_S(Q,\omega) = \frac{1}{2} \left\{ \frac{\Gamma}{(\omega-\omega_Q)^2 + \Gamma^2} + \frac{\Gamma}{(\omega + \omega_Q)^2 + \Gamma^2} \right\} \qquad (2)$$

where Γ is the half-width at half maximum (HWHM) and $\hbar\omega_Q$ is the energy of the spin wave excitation. For small Q, the dispersion law for spin waves in a ferromagnet has a quadratic form:

$$\hbar\omega_Q = \Delta + DQ^2 \qquad (3)$$

where D is the spin wave magnetic stiffness constant and Δ is the anisotropy gap. The solid lines in Fig. 2 represent the best fit of the cross section expression (Eqs. 1-3) convoluted with the instrumental resolution, to the observed spectra. The parameters D, Γ, A_G, A_S were varied during the non-linear least squares fitting procedure. A value for Δ of ~ 0.02 meV was obtained from the analysis, which increased slightly for T < 50 K.

The temperature variation of the magnetic stiffness constant D and the parameters A_G and A_S are given in Fig. 3. D increases below the Curie temperature and then exhibits a decrease for T < 50 K. It is not possible to state whether D actually goes to zero. In the simple molecular field approximation, this drop indicates a decrease in the magnetization. This behavior has been observed in several ferromagnetic materials, both crystalline and amorphous, upon approaching a spin glass-like state. The maximum value of D obtained at T = 50 K is ~ 26 meV-Å^2 which is 2/3 of the value extrapolated from Ref. 12. It should be noted that D deduced from magnetization measurements depends on the applied magnetic field.

The integrated spin wave intensity, A_S, decreases with temperature, but does not extrapolate to zero at T = 0 which simple spin wave theory suggests. The amplitude of the central peak, A_G, shows only a slight increase at low temperatures as seen in the data of Fig. (3b). Note that A_G never exceeds A_S. In contrast, the behavior of A_G in the other amorphous systems studied $(Fe_{1-x} T_x)_{75} P_{16} B_6 Al_3$, with T = Mn and Ni, shows a significant increase. Our results are more like those of polycrystalline Fe_xCr_{1-x} with x = 0.26 where T_c = 175 K. In that case D showed significant softening while a small increase in the critical scattering at lower temperatures was observed. For our amorphous sample of x = 0.77 with T_c = 185 K, the small change in A_G suggests that at low temperatures, the spin glass state is not fully developed. It will be useful to measure this amorphous alloy at smaller Q's and lower temperatures to see if there is additional enhancement of the elastic scattering.

ACKNOWLEDGMENTS

The authors would like to thank G. Aeppli, K. Motoya and B. H. Grier for helpful discussions and assistance. Work at Brookhaven supported by the Division of Materials Sciences U. S. Department of Energy under contract De-AC02-76CH00016.

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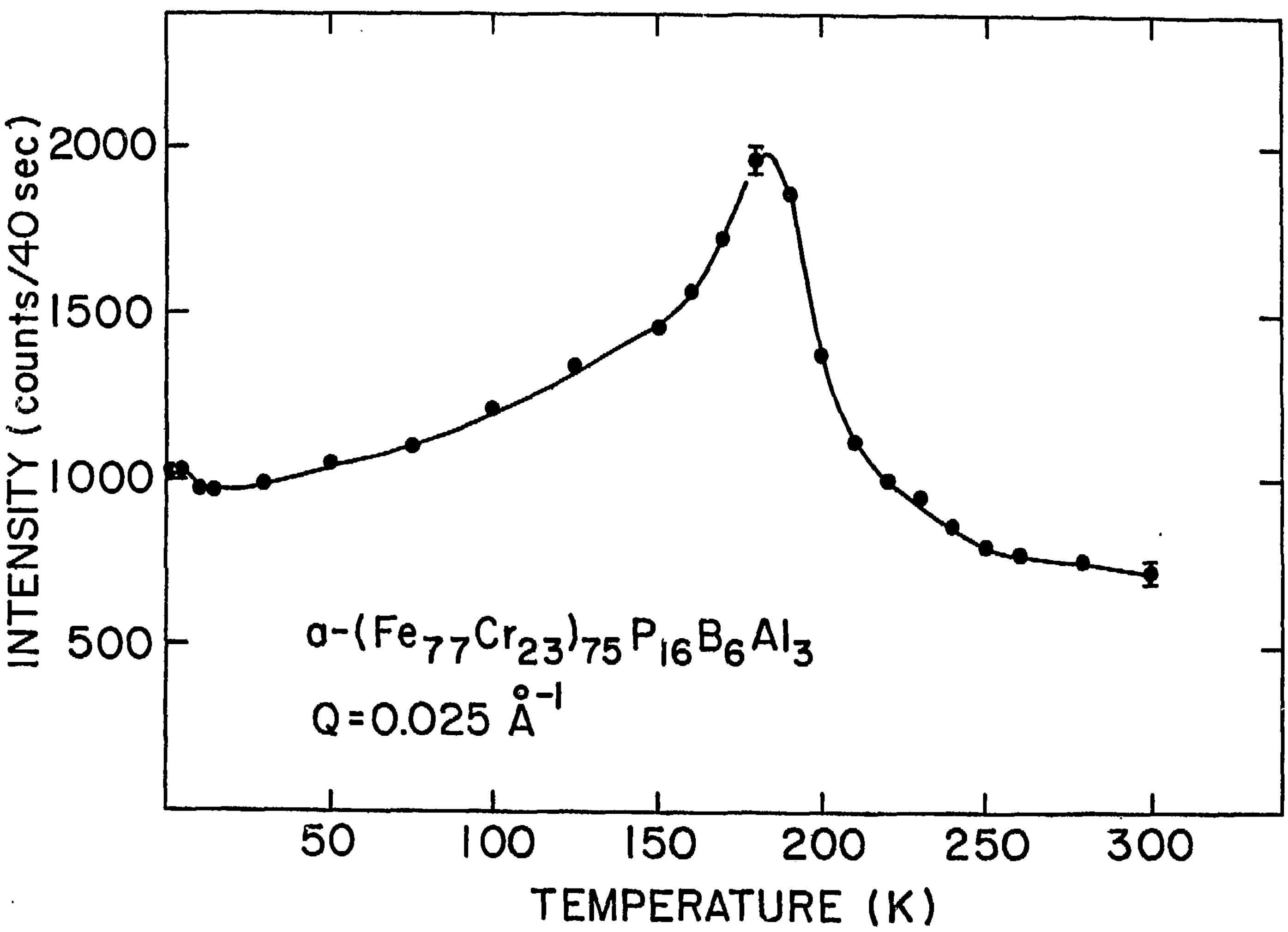
Figure Captions

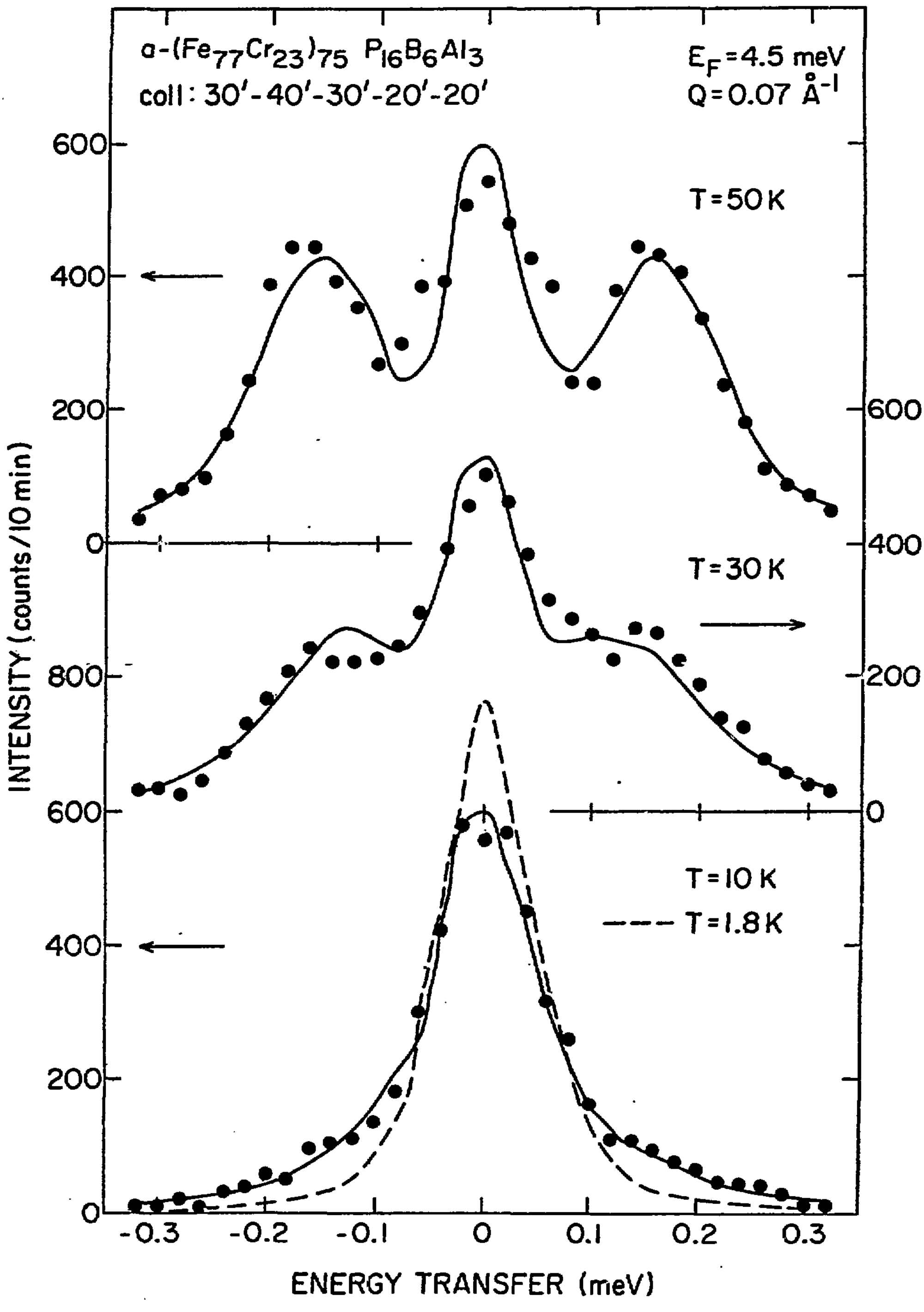
Fig. 1 Elastic scattering measured at $Q = 0.025Å^{-1}$ for $\Delta E = 0$ for $a-(Fe_{77}Cr_{23})_{75} P_{16}B_6Al_3$.

Fig. 2 Temperature dependent spectra of $a-(Fe_{77}Cr_{23})_{75} P_{166}Al_3$. An energy independent background has been subtracted from each data point for temperatures less than 50 K. Solid lines are fits to the data as described in the text. Broken line represents data obtained at 1.82 K.

Fig. 3 Temperature dependence of parameters resulting from least squares fits to the data (Fig. 2). (a) Spin wave magnetic stiffness constant D and (b) elastic A_G and inelastic A_S integrated intensity parameters for $Q = 0.07Å^{-1}$.



INTENSITY (counts/40 sec)
2000
1500
1000
500
50
100
150
200
250
300
TEMPERATURE (K)
a-(Fe77Cr23)75P16B6Al3
Q=0.025 Å-1



a-(Fe77Cr23)75 P16B6Al3
coll: 30′-40′-30′-20′-20′
EF=4.5 meV
Q=0.07 Å-1
T=50 K
T=30 K
T=10 K
T=1.8 K
INTENSITY (counts/10 min)
ENERGY TRANSFER (meV)

FIGURE 2

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PACS Numbers: 75.25.+z, 75.30.Ds, 75.30.Kz, 75.50.Kj

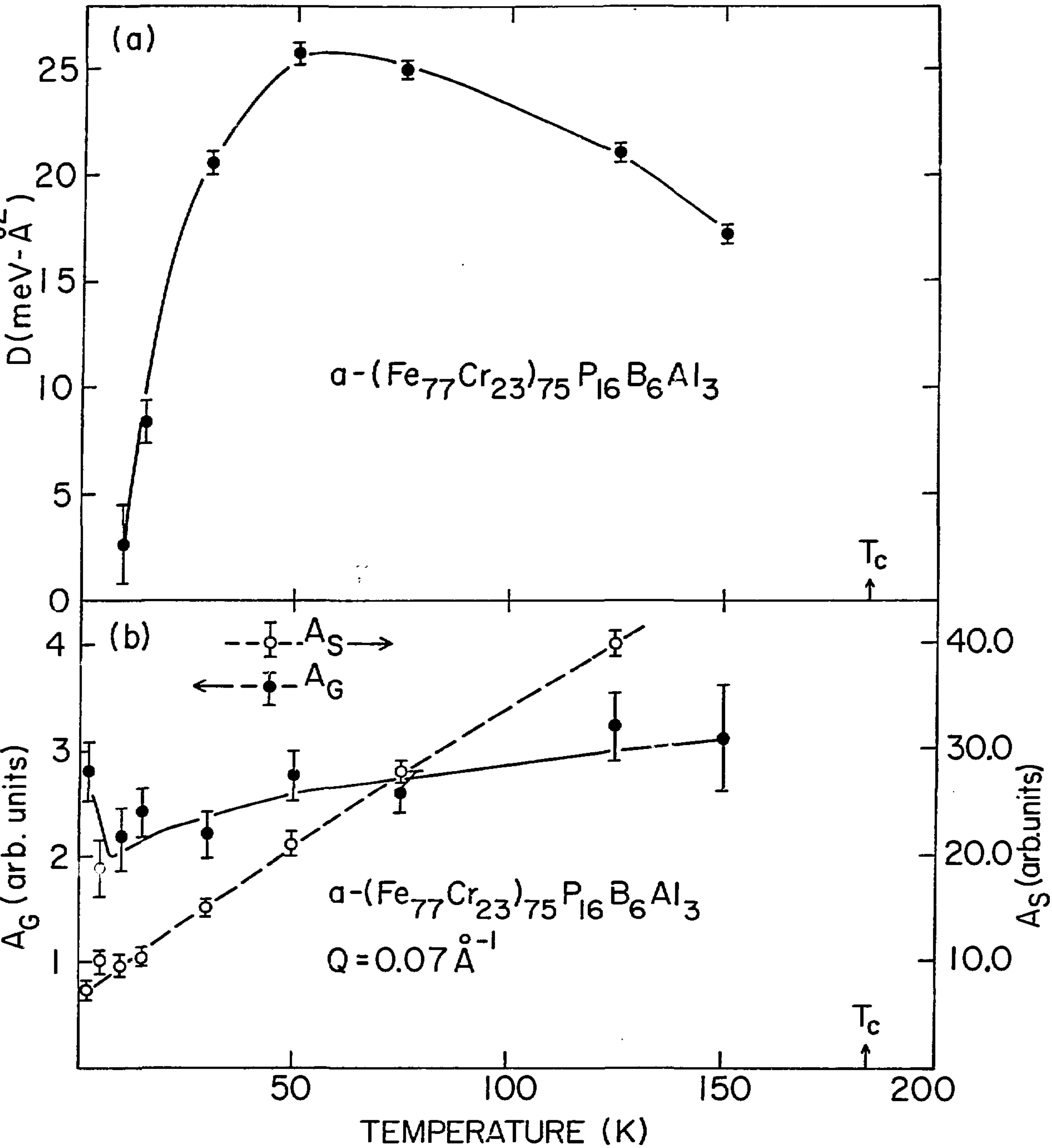
(a)
D(meV-Å^2)
0
5
10
15
20
25
a-$(Fe_{77}Cr_{23})_{75}P_{16}B_6Al_3$
T_c
(b)
A_S
A_G
A_G (arb. units)
A_S (arb.units)
1
2
3
4
10.0
20.0
30.0
40.0
a-$(Fe_{77}Cr_{23})_{75}P_{16}B_6Al_3$
Q = 0.07 Å^{-1}
T_c
50
100
150
200
TEMPERATURE (K)

FIGURE 3