Magnetic properties of amorphous $(Co_{1-x}Ni_x)_{75}P_{16}B_6Al_3$ [a)]

Y. Yeshurun, K. V. Rao,[b)] and M. B. Salamon

University of Illinois at Urbana-Champaign, Urbana, Illinois 61801

H. S. Chen

Bell Laboratories, Murray Hill, New Jersey 07974

The temperature and field dependence of the magnetization in a series of amorphous Co-Ni alloys is reported. We discuss the critical behaviour and introduce a phase diagram which includes paramagnetic, ferromagnetic (FM) and spin glass (SG) phases. A $T^{3/2}$ dependence for the magnetization is established in the FM phase. Deviations from spin wave theory which are observed in alloys with a FM-SG transition are discussed in terms of single spin excitations.

PACS numbers: 75.30.Cr, 75.30.Ds, 75.30.Kz, 75.50.Kj

INTRODUCTION

In a recent paper (1) we described magnetic measurements of amorphous $(Fe_{1-x}Mn_x)_{75}P_{16}B_6Al_3$ which have proven to be very fruitful in demonstrating the critical behaviour at the ferromagnetic (FM) - spin glass (SG) transition temperature T_{fg} as well as at the Curie temperature T_c. In the present paper we introduce similar results for $(Co_{1-x}Ni_x)_{75}P_{16}B_6Al_3$ amorphous alloys. We discuss briefly the critical behaviour and derive a magnetic phase diagram with the qualitative characteristics of the theoretical phase diagram predicted by the Kirkpatrick-Sherrington (KS) model. (2) Finally, we focus our attention on the temperature dependence of the saturation magnetization below the Curie temperature. Spin-wave excitations result in a $T^{3/2}$ behaviour for FM materials at low temperature. (3) However, conventional spin-wave theories cannot be applied to a FM system which undergoes a SG transition at low temperature. In an attempt to gain more insight into the FM-SG transition, we analyze the magnetization data at low temperature for the various concentrations of Co and summarize the results in terms of the stiffness constant D.

EXPERIMENTAL

Ribbons of the amorphous materials were prepared by centrifugal spin quenching. (4) Small pieces of the material (~5mm x 1mm x 25 μm) were packed in the sample holder of a Vibrating Sample Magnetometer with the long axis parallel to the external field to minimize demagnetization effects. We used external fields from 10G to 15 kG starting at 4.2 K and in increasing order of temperature and field.

RESULTS AND DISCUSSION

Typical magnetization curves are shown in Fig. 1 for $(Co_{.40}Ni_{.60})_{75}P_{16}B_6Al_3$. For low fields (see insert of Fig. 1) the magnetization increases with temperature, reaches a maximum at T_m and then decreases. The maximum at T_m is shifted to lower temperatures as the external field is increased. For $H_{ext} > 50$ G the maximum totally disappears and the isofield curves exhibit the usual FM behaviour. Alloys with $.5 < x < .7$ exhibit the same behaviour - the lower x is the lower is the external field which washes out the maximum in M(T). This behaviour is typical of alloys with FM - SG transitions (compare ref. 1). For x = .8 the magnetization curves exhibit broader peaks and the temperature of maximum magnetization does not depend on external field. As we discuss later this is typical of SG behaviour.

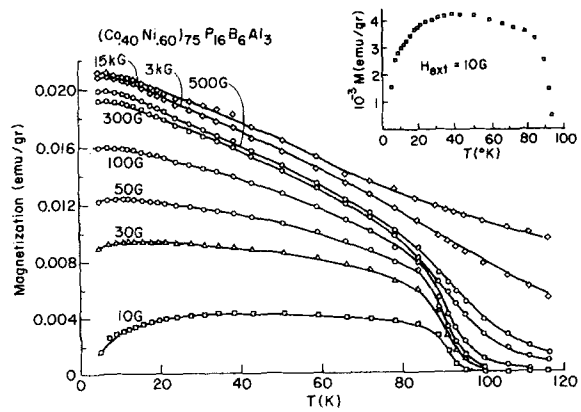

Fig. 1 Temperature dependence of the magnetization for $(Co_{.40}Ni_{.60})_{75}P_{16}B_6Al_3$ for different applied fields. Insert shows M(H,T) for H_{ext} = 10G.

CRITICAL BEHAVIOUR

Scaling laws (5) imply a simple relationship $\gamma = \beta(\delta-1)$ between the critical exponents, and a simple form of the magnetic equation of state: $M/|\tau|^{\beta} = m^*(H/|\tau|^{\beta\delta}\cdot \mathrm{sgn}\tau)$, where $\tau \equiv (T-T_c)/T_c$. The scaling function m^* has two branches ($\tau>0$, $\tau<0$). By choosing the correct parameters T_c, β and δ all data points M(H,T) should collapse to these branches in an m^* vs $H/|\tau|^{\beta\delta}$ plot. (For more details, see ref. 1). This scaling procedure leads to non-Heisenberg critical magnetic exponents for the present system ($.40 < x < .66$): $\beta = .41 \pm .60$ and $\delta = 5.1 \pm .4$. Smaller δ ($4.3 \pm .5$) are obtained for x = .68, and x = .70. For higher concentrations of Ni the data do not exhibit scaling behaviour. Arrott plots for x>.70 show clearly the absence of spontaneous magnetization in those samples for the whole temperature regime.

As a summary of the above discussion, we show in Fig. 2 the phase diagram for the $(Co_{1-x}Ni_x)_{76}P_{16}B_6Al_3$ system. Qualitatively the same phase diagram was introduced in Ref. 1 for $(Fe_{1-x}Mn_x)_{75}P_{16}B_6Al_3$. However, the multicritical point is $x \simeq .72$ for the Co-Ni system

 0021-8979/81/031747-03$1.10

compared to x = .36 for the Fe-Mn system. Using the KS notations (2) we note that J/Jo is increasing with x for both systems. However, the multicritical value J/Jo = 1 is reached more rapidly in the Fe-Mn system. We suggest that this is the result of additional competing interaction between Mn spins in contrast to the paramagnetic nature of the $Ni_{75}P_{16}B_6Al_3$ matrix. (6) More evidence is necessary for support of this explanation.

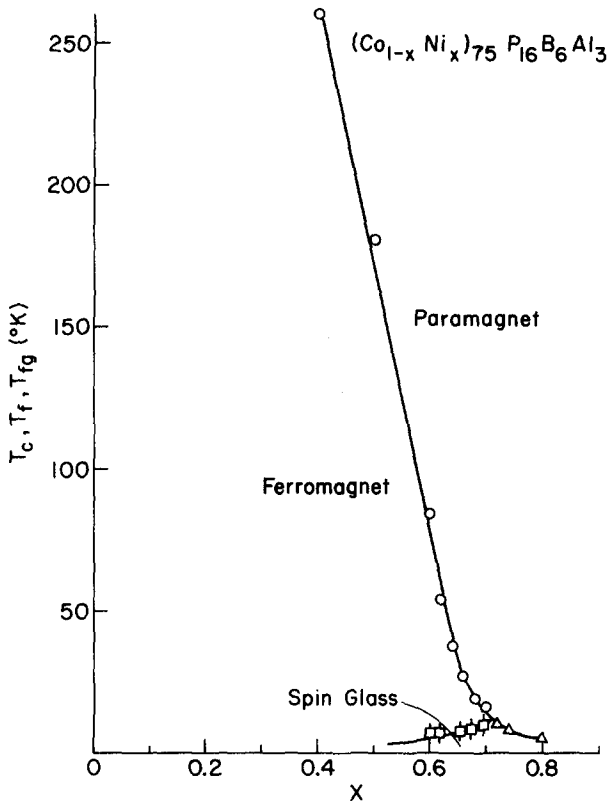

Fig. 2 Magnetic phase diagram for $(Co_{1-x}Ni_x)_{75}P_{16}B_6Al_3$.

LOW TEMPERATURE BEHAVIOUR

Spin wave excitations in ferromagnets have a dispersion relation of the form $\epsilon(q) = Dq^2+...$, where q is the wave vector and D, the stiffness constant, is proportional to the exchange integral J. Using this dispersion relation and neglecting magnon-magnon interactions (as well as single spin excitations) one gets for the saturation magnetization (3)

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

with b inversely porportional to $M_oD^{3/2}$, $M_o \equiv M_s(T=0)$.

Equation (1) is valid for amorphous ferromagnets as well, since the long wave length spin waves can be clearly defined. (7) Indeed, a $T^{3/2}$ dependence is found experimentally in many amorphous alloys and over a wide range of temperature. (8-10) This is also the case for the present system. The saturation magnetization for alloys with concentration $0 \le x \le .50$ exhibit $T^{3/2}$ behaviour up to $T/T_c \simeq 0.6$. However, for $.60 \le x \le .70$ -- namely for alloys with a FM-SG transition -- we find deviations from $T^{3/2}$ law at low temperatures. Similar deviations were found in spin-resonance measurements with $(Fe_{1-x}Ni_x)_{75}P_{16}B_6Al_3$ amorphous alloys. (10) These deviations, in both systems, can be correlated with recent neutron scattering measurements. (11) In these experiments it was found that D is almost temperature independent over a wide range of temperature unless the sample undergoes a FM-SG transition. In the latter case D has a broad maximum within the FM phase and practically vanishes below the transition temperature T_{fg}. A temperature independent D suggests that $M_S(T)$ can be described with a temperature-independent parameter b as in Eq. (1). But, if the sample undergoes a FM-SG phase transition, one may expect single spin instead of spin-wave excitations to dominant the low temperature behaviour. In order to clarify the role of single spin excitations, we recall the mean field approximation (MFA) result at low temperature:

$$M_S(T) = M_0 - aT \qquad (2)$$

where a is proportional to P(H=0), the internal field distribution at H=0. (2,12) In a FM phase a is small (compared to the value at SG phase) and the linear contribution from single spin excitations is negligible compared with the collective, spin wave excitations. However, as the temperature decreases, P(H=0) increases and the aT term becomes the dominant contribution below T_{fg}.

In the following we present results of fitting $M_S(T)$ with Eq. (1) for $0 \le x \le .7$. For alloys with FM-SG transition we ignore the lower temperature data and start the fit, quite arbitrary, at $2T_{fg}$. Also, it is worthwhile to mention here that for $x \le .5$, saturation is achieved with fields on the order of 10 kG while for larger x we have to use the $(1/H + 1/H^2)$ law to evaluate $M_S(T)$.

The first remarkable result of the procedure is the behaviour of b, which increases rapidly with x from a value of $.25 \times 10^{-4}\ K^{-3/2}$ at x = 0 to a value of $16.9 \times 10^{-4}\ K^{-3/2}$ at x = .7. To compare these results with other materials it is common to describe the dimensionless quantity $B = b\,T_c^{3/2}$ instead. The B values range from .22 to .49 which is common for amorphous ferromagnets. (8,9) However, a plot of B vs x (Fig. 3) exhibits a very unusual characteristic of this system, namely a maximum of B at $x \simeq .6$ (where FM-SG transitions appear). Linear extrapolation of B above x = .7 (see Fig. 3) gives zero T_c for $x \simeq .74$ which is the multicritical concentration.

The stiffness constants determined from the coefficients b(x) are plotted in Fig. 4 as a function of the concentration. Over a wide range of concentrations ($x \gtrsim .4$) D decreases linearly toward a zero value at $x_c \simeq .74$. Zero (and negative) values of D signal an instability of the FM phase (13) of the system.

In Fig. 5 we replot D as function of T_c. This Figure suggests an empirical relation $D = D_o + k\,T_c$ with positive D_o and slope k. Similar experimental expressions, though with different D_o and k, are

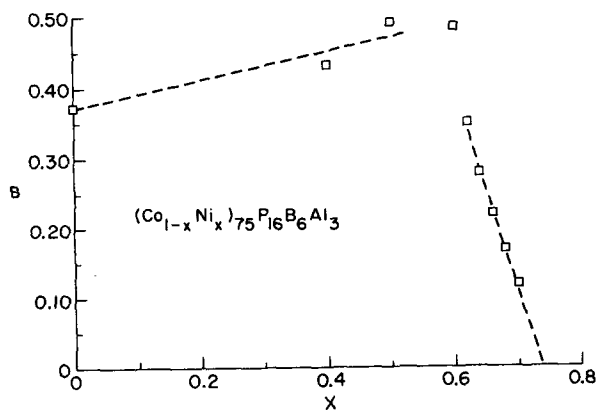

Fig. 3 Slope of spin wave equation B as a function of Ni concentration x.

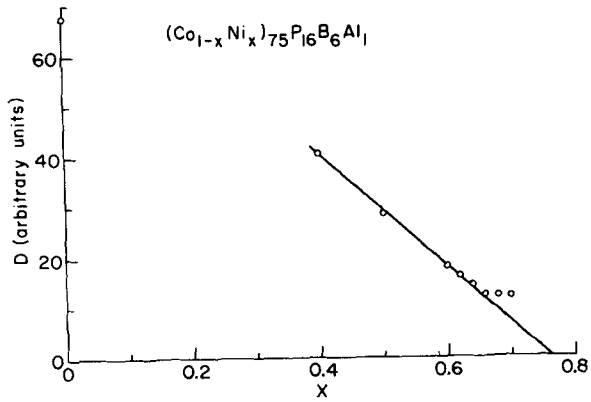

Fig. 4. Stiffness constant D as a function of Ni concentration x.

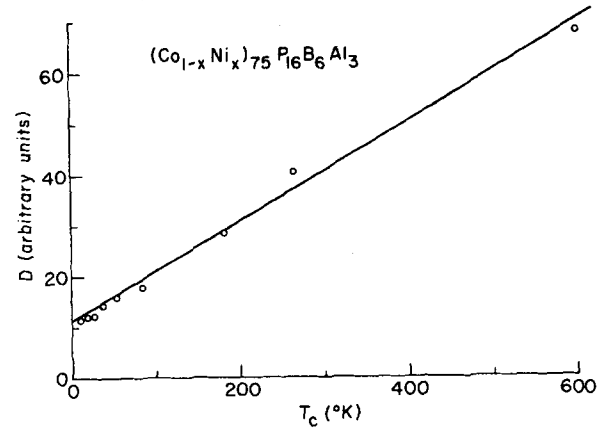

Fig. 5. Stiffness constand D as a function of Curie temperature T_c.

known in the literature. (8) Theoretical linear correlation has been discussed for the itinerant electron model. (13) However, other features of this simple model are not met by experimental results. (e.g. $D_o = 0$)

In conclusion, through the temperature dependence of the magnetization of amorphous $(Co_{1-x}Ni_x)_{75}P_{16}B_6Al_3$ alloys we have located the ferromagnetic and spin-glass phase boundaries. In the critical regime we find the usual non-Heisenberg exponents. At low temperatures we find a $T^{3/2}$ dependence but with deviations from spin-wave theory below the FM-SG transition temperature. The results of $T^{3/2}$ fit within the FM regime are discussed in terms of the stiffness constant and compared to previous experiments.

REFERENCES

a) Supported in part by the National Science Foundation, MRL grant DMR 77-23999.

b) Present address: Department of Solid State Physics, Royal Institute of Technology, Stockholm, Sweden.

1. Y. Yeshurun, M. B. Salamon, K. V. Rao and H. S. Chen (to be published).
2. S. Kirkpatrick and D. Sherrington, Phys. Rev. B17, 4384 (1978).
3. C. Kittel, "Quantum Theory of Solids" (John Wiley, N.Y., 1963).
4. H. S. Chen and C. E. Miller, Mater. Res. Bull. 11, 49 (1976).
5. H. G. Stanley, "Introduction to Phase Transition Critical Phenomena" (Oxford University, N.Y. 1971).
6. H. S. Chen, Rep. Prog. Phys. 43, 353 (1980).
7. T. Kaneyoshi, J. Phys. Soc. Japan 45, 1835 (1978).
8. F. G. Luborsky, J. L. Walter, H. H. Liebermann and E. P. Wohlfarth, J. Magn. Magn. Mat 15, 1351 (1980).
9. S. Hatta and T. Egami, J. Appl. Phys. 50, 1589 (1979)
10. S. M. Bhagat, M. L. Spano, H. S. Chen and K. V. Rao, Solid State Comm. 33, 303 (1980).
11. C. R. Fincher, S. M. Shapiro, A. H. Palumbo and R. D. Parks, Phys. Rev. Lett. 45, 474 (1980). J. W. Lynn, R. W. Erwin, J. J. Rhyne and H. S. Chen, Proceedings of this Conference.
12. M. W. Klein, L. J. Schowalter and P. Shukla, Phys. Rev. B19, 1492 (1979).
13. A. Katsuky and E. P. Wohlfarth, Proc. Roy. Soc. A295, 182 (1966).