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Magnetic and electrical properties of iron-nickel-chromium-metalloid glasses

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The magnetic and electrical properties of amorphous $(\text{Fe}_{1-x}\text{Ni}_x)_{78-y}\text{Cr}_y\text{Si}_{10}\text{B}_{12}$ alloys have been measured between 4.2 and 300 K. Alloys with $x \leq 0.9$ and $y = 5$ order ferromagnetically. High-field magnetization measurements show saturation in moderate fields suggesting a relatively high value of magnetic anisotropy which was estimated to be $K \simeq 10^6$ erg/cm³. The magnetization at low temperatures follows a $T^{3/2}$ law leading to a spin-wave stiffness coefficient $D \simeq 90$ meV Å². The $x = 1$, $y = 5$ alloy behaves like a "cluster glass" with a cluster ordering temperature around 20 K. Resistivity measurements show minima at low temperatures which can be explained with a modified Kondo mechanism. The resistivity maximum which appears around the cluster ordering temperature in $\text{Ni}_{73}\text{Cr}_5\text{Si}_{10}\text{B}_{12}$ is associated with a spin freeze-out of the Kondo effect due to local moments on Cr atoms.

I. INTRODUCTION

Metallic glasses are important from scientific¹ and technological^{2,3} viewpoints. Among them the transition-metal-metalloid alloys can be prepared over a wide range of composition and show outstanding mechanical, electrical, and magnetic properties.⁴⁻⁶

There have been several reports on the electrical and magnetic properties of metglasses. However, most of the papers so far published⁷⁻⁹ report different properties of the alloys but do not examine several electrical and magnetic properties of the same set of samples.

In this paper we report the electrical and magnetic properties of $(\text{Fe}_{1-x}\text{Ni}_x)_{78-y}\text{Cr}_y\text{Si}_{10}\text{B}_{12}$, amorphous alloys. Particularly we are interested in the kind of magnetic ordering which develops as the Ni concentration increases. High-field magnetization measurements were used over a wide range of temperatures to determine the spontaneous moment, the magnetic anisotropy, and the spin-wave stiffness at low temperatures. Additional points of interest include the resistivity minima which appear at low temperatures and the influence of Cr content on the electrical resistivity and coercive force.

II. EXPERIMENTAL

Samples of $(\text{Fe}_{1-x}\text{Ni}_x)_{78-y}\text{Cr}_y\text{Si}_{10}\text{B}_{12}$, with $y = 0.5$ and $0 \leq x \leq 1$, were prepared by the splat cooling technique. They were found to be amorphous by x-ray-diffraction analysis, as shown for a typical sample in Fig. 1. Magnetic susceptibility was measured between 4.2 and 300 K with the Faraday technique. Magnetization measurements were made with a vibrating sample magnetometer in magnetic fields up to 80 kOe and in the temperature range of 1.3 to 300 K. Resistivity measurements were performed between 4.2 and 300 K with the standard four-probe technique. Annealing was done at 300°C with the samples sealed under vacuum in a quartz tube.

III. RESULTS AND DISCUSSION

A. Magnetic ordering

Amorphous Fe-Ni-based alloys with more than 10 at. % Fe are ferromagnetic⁸ with ordering temperatures T_0 ranging up to 640 K (Table I). If Fe is completely replaced by Ni, T_0 drops to zero. The addition of Cr metal causes a rapid decrease in

FIG. 1. X-ray diffractogram of amorphous $\text{Fe}_{13}\text{Ni}_{60}\text{Cr}_5\text{Si}_{10}\text{B}_{12}$. $\text{MoK}\alpha$ radiation was used.

T_0 since the Cr moments couple antiferromagnetically with the dominant Fe moments.¹⁰

Evidence of magnetic ordering has been observed in $\text{Fe}_{13}\text{Ni}_{60}\text{Cr}_5\text{Si}_{10}\text{B}_{12}$ where magnetic susceptibility measurements show a broad peak around 22 K (Fig. 2). No peak in χ was observed in $\text{Ni}_{73}\text{Cr}_5\text{Si}_{10}\text{B}_{12}$ above 4.2 K (Fig. 3). The high-temperature data were fitted to a Curie-Weiss law,

$$\chi - \chi_0 = \frac{Np_{\text{eff}}^2\mu_B^2}{3k_B(T - \Theta)}, \quad (1)$$

where χ_0 is a temperature-independent contribution to susceptibility including the signal of the sample holder and N is the number of spins per unit mass. In $\text{Fe}_{13}\text{Ni}_{60}\text{Cr}_5\text{Si}_{10}\text{B}_{12}$, the effective moment per average atom, p_{eff} , was found to be 4.2 and the Curie temperature $\Theta = 102$ K indicating an overall ferromagnetic behavior. However, in $\text{Ni}_{73}\text{Cr}_5\text{Si}_{10}\text{B}_{12}$ $p_{\text{eff}} = 0.28$ and $\Theta = -15$ K suggesting the existence of antiferromagnetic interactions.

B. High-field magnetization measurements

Magnetization measurements show saturation in a relatively high field (Fig. 4). Values of the spontaneous moment and the high-field susceptibility were found from the law of approach to saturation (see Sec. III D). The spontaneous magnetization is significantly reduced upon substitution of Fe with Ni and it drops close to zero in $\text{Ni}_{73}\text{Cr}_5\text{Si}_{10}\text{B}_{12}$ (Fig. 4). Values of moment per average atom, μ , were found from the spontaneous magnetization values and are listed on Table I. In $\text{Fe}_{78}\text{Si}_{10}\text{B}_{12}$ the average value of the iron moment was found to be $\mu_{\text{Fe}} = 2\mu_B$. This value was used for the rest of the alloys in order to find μ_{Cr} and μ_{Ni} . In $\text{Fe}_{73}\text{Cr}_5\text{Si}_{10}\text{B}_{12}$, μ_{Cr} was found to be $4.8\mu_B$ and antiferromagnetically coupled to the Fe moment. For $\text{Fe}_{37}\text{Ni}_{36}\text{Cr}_5\text{Si}_{10}\text{B}_{12}$ and $\text{Fe}_{13}\text{Ni}_{60}\text{Cr}_5\text{Si}_{10}\text{B}_{12}$, using the previously found values of μ_{Fe} and μ_{Cr} , μ_{Ni} was found to be $0.6\mu_B$ and $0.3\mu_B$, respectively. This seems to be consistent with other observations¹¹ which suggest that the Ni atoms can be progressively polarized when the Fe content increases.

TABLE I. Summary of the magnetic and electrical properties of amorphous $(\text{Fe}_{1-x}\text{Ni}_x)_{78-y}\text{Cr}_y\text{Si}_{10}\text{B}_{12}$.

Composition	T_0 (K)	$\mu_0(\mu_B)^a$	p_{eff}^b	B ($10^{-6} \text{ K}^{-3/2}$)	D ($\text{meV} \text{ \AA}^2$)	T_{max} or T_{min} (K) ^c	ρ ($\mu\Omega \text{ cm}$)
$\text{Fe}_{78}\text{Si}_{10}\text{B}_{12}$	640	1.560				16	280
$\text{Fe}_{73}\text{Cr}_5\text{Si}_{10}\text{B}_{12}$	620	1.220		31.4	94	22	390
$\text{Fe}_{37}\text{Ni}_{36}\text{Cr}_5\text{Si}_{10}\text{B}_{12}$	600	0.718		67.14	78	65	270
$\text{Fe}_{13}\text{Ni}_{60}\text{Cr}_5\text{Si}_{10}\text{B}_{12}$	120	0.197	4.21			32	180
$\text{Ni}_{73}\text{Cr}_5\text{Si}_{10}\text{B}_{12}$	20	0.002	0.28 ^d			15	110
$\text{Ni}_{78}\text{Si}_{10}\text{B}_{12}$							100

^a μ_0 is the moment per average atom of alloy (determined from spontaneous magnetization).

^b p_{eff} is the paramagnetic moment per average atom of alloy [see Eq. (1)].

^cTemperatures where a resistivity maximum or minimum occurs.

^dThe effective moment per Cr atom (p_{Cr}) is 1.25 assuming only Cr atoms have a moment in this sample.

FIG. 2. Susceptibility and Curie-Weiss fit for amorphous $\text{Fe}_{13}\text{Ni}_{60}\text{Cr}_5\text{Si}_{10}\text{B}_{12}$.

However, a more complex behavior has been observed in $\text{Ni}_{73}\text{Cr}_5\text{Si}_{10}\text{B}_{12}$. The high-field magnetization measurements (Fig. 5) suggest the existence of a predominantly ferromagnetic cluster in a paramagnetic matrix. The high-field susceptibility and the extrapolated values of the nearly linear high-field portion of the magnetization curve (Fig. 5) are plotted as a function of temperature in Fig. 6. This gives a rough estimate of the cluster contribution to the magnetization. The magnetic moment of the cluster is found to be $\mu_{cl} = 0.002\mu_B$ per atom. Assuming $\mu_{Ni} = 0$, which is found by others¹¹ for highly concentrated Ni amorphous alloys, the cluster magnetic moment due to Cr spins is

FIG. 3. Susceptibility and Curie-Weiss fit for amorphous $\text{Ni}_{73}\text{Cr}_5\text{Si}_{10}\text{B}_{12}$.

FIG. 4. Magnetization curves for amorphous $(\text{Fe}_{1-x}\text{Ni}_x)_{78-y}\text{Cr}_y\text{Si}_{10}\text{B}_{12}$ at 4.2 K.

found to be $0.034\mu_B$ per average Cr atom. This value is substantially less than the typical Cr moment and is consistent with susceptibility measurements which show a relatively small value of $p_{Cr} = 1.25$ (Table I). The small value of the magnetic moment suggests either not all Cr atoms have moment or the moment of the Cr atom is significantly reduced. The high-field susceptibility values

FIG. 5. Magnetization curves for amorphous $\text{Ni}_{73}\text{Cr}_5\text{Si}_{10}\text{B}_{12}$ at several temperatures.

FIG. 6. Temperature dependence of the extrapolated value of the high-field magnetization curve to $H=0$ and the high-field susceptibility dM/dH in $\text{Ni}_{73}\text{Cr}_5\text{Si}_{10}\text{B}_{12}$.

(Fig. 6) were fitted with the Curie-Weiss law and the Curie temperature was found to be $\Theta = -17$ K which is very close to the value obtained from the Faraday susceptibility data (Fig. 3). The cluster ordering temperature is estimated to be around 17 K.

C. Temperature dependence of magnetization

The magnetization curves were studied at different temperatures in order to determine the temperature dependence of the spontaneous moment (Fig. 7). At low temperatures the decrease in moment with increasing temperature can be well represented by a $T^{3/2}$ law (Fig. 8),

$$M = M_0(1 - BT^{3/2}), \quad (2)$$

where M_0 is the spontaneous magnetization per unit volume. If the decrease in moment is due to the excitation of long-wavelength spin waves the coefficient B is given by¹²

$$B = 2.612(g\mu_B M_0^{-1}) \left(\frac{k_B}{4\pi D} \right)^{3/2}, \quad (3)$$

where D is the spin-wave stiffness coefficient used in the ferromagnetic dispersion relation

$$\hbar\omega_q = Dq^2 + Eq^4. \quad (4)$$

Assuming density values of 7.0 and 7.5 g/cm³ for $\text{Fe}_{73}\text{Cr}_5\text{Si}_{10}\text{B}_{12}$ and $\text{Fe}_{37}\text{Ni}_{36}\text{Cr}_5\text{Si}_{10}\text{B}_{12}$, respective-

FIG. 7. Magnetization curves in amorphous $\text{Fe}_{37}\text{Ni}_{36}\text{Cr}_5\text{Si}_{10}\text{B}_{12}$ at temperatures 4.2, 74, 105, 130, 151, and 230 K (top to bottom).

ly,¹³ leads to the values of B and D listed in Table I. These values are comparable with those reported¹⁴ for $(\text{Fe}_{1-x}\text{Ni}_x)_{75}\text{P}_{16}\text{B}_6\text{Al}_3$. However, they appear to be slightly lower than the values measured by inelastic neutron scattering.¹⁵

D. Magnetic anisotropy

The magnetization curves of Fig. 4 suggest the presence of a relatively high anisotropy.¹⁶ The law

FIG. 8. Spontaneous magnetization versus $T^{3/2}$ for amorphous $\text{Fe}_{37}\text{Ni}_{36}\text{Cr}_5\text{Si}_{10}\text{B}_{12}$.

FIG. 9. Magnetization versus $1/H$ in amorphous $\text{Fe}_{37}\text{Ni}_{36}\text{Cr}_5\text{Si}_{10}\text{B}_{12}$ at 4.2 K.

of approach to saturation¹⁷ and the magnetization-area method were used to determine the anisotropy constant K .

The approach to saturation is usually expressed in the following empirical form,

$$M = M_0(1 - AH^{-1} - BH^{-2}) + \chi_H H, \quad (5)$$

where χ_H is the high-field susceptibility, A/H is the term due to pinning of domain walls by the microstructure inhomogeneities and B/H^2 is due to rotations of the magnetization vector against the magnetic anisotropy with $B = 4K^2/15M_0^2$. A plot of M vs $1/H$ (Fig. 9) in $\text{Fe}_{37}\text{Ni}_{36}\text{Cr}_5\text{Si}_{10}\text{B}_{12}$ shows that the A/H term is predominant for low fields up to 10 kOe above which deviations occur.

Therefore a least-squares fit of the magnetization

data above 10 kOe was attempted using Eq. (5) with $A = 0$. The values of M_0 , K , and χ_H obtained are listed in Table II. Similar fits were performed at different temperatures and on samples annealed for 6 h at 300°C. The results are shown in Fig. 10.

In the magnetization-area method the anisotropy K is found by the following expression¹⁸:

$$\int_{M_R}^{M_0} H dM = \frac{2}{3} K, \quad (6)$$

where M_R is the remanent magnetization and the integral corresponds to the area to the left of the magnetization curve as the field is decreased from the saturation value to zero. This expression is valid assuming a random distribution of easy axes in the different domains and a spherical average of

TABLE II. Values of the spontaneous moment, magnetic anisotropy and high-field susceptibility at 4.2 K in $(\text{Fe}_{1-x}\text{Ni}_x)_{78-y}\text{Cr}_y\text{Si}_{10}\text{B}_{12}$ amorphous alloys.

Composition	M_0 (emu/g)	K (10^6 erg/cm ³)	χ_H (10^{-5} emu/g)
$\text{Fe}_{78}\text{Si}_{10}\text{B}_{12}$	186.9	7.9	1.0
$\text{Fe}_{73}\text{Cr}_5\text{Si}_{10}\text{B}_{12}$	144.5	5.5	1.7
$\text{Fe}_{37}\text{Ni}_{36}\text{Cr}_5\text{Si}_{10}\text{B}_{12}$	82.7	1.9 (1.6) ^a	1.0
$\text{Fe}_{37}\text{Ni}_{36}\text{Cr}_5\text{Si}_{10}\text{B}_{12}$ (annealed)	81.9	1.6	1.2
$\text{Fe}_{13}\text{Ni}_{60}\text{Cr}_5\text{Si}_{10}\text{B}_{12}$	28.7	2.2	4.0

^aMagnetization-area method.

FIG. 10. Temperature dependence of magnetic anisotropy in amorphous $\text{Fe}_{73}\text{Ni}_{36}\text{Cr}_5\text{Si}_{10}\text{B}_{12}$ and $\text{Fe}_{73}\text{Cr}_5\text{Si}_{10}\text{B}_{12}$ alloys.

the anisotropy over the easy axes.

In $\text{Fe}_{37}\text{Ni}_{36}\text{Cr}_5\text{Si}_{10}\text{B}_{12}$ the values of K found by this way are very close to the values obtained from the law of approach to saturation (Table II). However, the observed anisotropies are much higher than those reported by other workers^{19,20} for different Metglasses. The small temperature dependence of the estimated anisotropy and its persistence after annealing implies that it is not due to the larger internal stresses produced by the rapid quenching of the samples.⁸ Possibilities for the origin of the observed anisotropy include exchange anisotropy (see next subsection) and shear stresses arising from anisotropic metal-metalloid atomic arrangements.¹⁹

E. Coercivity

Figure 11 shows the coercive force H_c as a function of iron content in the glass containing chromium. Because of trapped flux in our superconducting solenoid it was not possible for us to measure H_c in the $\text{Fe}_{78}\text{Si}_{10}\text{B}_{12}$ sample. The remanent field in our solenoid is about 10 Oe and it is well known that Fe-metalloid glasses of the form $\text{Fe}_{80}\text{M}_{20}$ have H_c values in the range of 10^{-1} – 10^{-2} Oe.²¹ The samples containing Cr have coercivities of several hundred oersteds, with the Ni-rich samples being somewhat harder than the Fe-rich ones. It appears that the most likely origin for the rather large values of H_c (and K —see previous subsection) is

FIG. 11. The coercive field H_c at 4.2 K vs Fe concentration in $(\text{Ni}_{1-x}\text{Fe}_x)_{73}\text{Cr}_5\text{Si}_{10}\text{B}_{12}$.

exchange anisotropy associated with the coupling of the Cr local moments with each other and with their surroundings. An analogous, but more extreme, example of this behavior was reported by Sherwood *et al.* in a more concentrated cluster glass: $(\text{Fe}_{0.5}\text{Mn}_{0.5})_{0.75}\text{P}_{0.16}\text{B}_{0.06}\text{Al}_{0.03}$.²² In this case typical spin-glass or mictomagnetic phenomena were observed with H_c values ranging up to about 7 kOe. The results were interpreted in terms of local exchange anisotropy in a glass which naturally can have no long-range magnetocrystalline anisotropy.

In general the microscopic origins of anisotropy and of the coercive force are not well understood in Fe-Ni alloys in which the glass formers are Si and B. For example Masumoto *et al.*²³ have found that in $(\text{Fe-Ni})_{78}\text{Si}_y\text{B}_{22-y}$, the anisotropy is a relatively strong function of y . Moreover, annealing for 1 h at 380°C decreases K by two orders of magnitude for $y \approx 0.07$, but by only a small factor for $y = 0.04$ and 0.11. They speculate that at the latter composition there may be microcrystalline precipitates or microscopic compositional fluctuations which lead to the relative hardness and insensitivity to annealing.

In one sample (see Table II) K was found to be insensitive to annealing. Again this suggests a major part of the origin of the relative magnetic hardness of our glasses is due to the exchange anisotropy rather than stress. On the other hand, as demonstrated by the above-mentioned work of

Masumoto *et al.*, it could well be the case that subtle structural effects, involving the glass forming atoms as well as Cr, could be playing an important role in our alloys. These structural effects are far beyond the sensitivity of standard x-ray diffraction measurements.

F. Electrical resistivity

Approximate values of resistivity for all samples are listed in Table I. A gradual decrease of resistivity is observed upon substitution of Fe with Ni.

The temperature dependence of the electrical resistivity is shown in Figs. 12 and 13. For all the samples the change in resistivity between 4.2 and 300 K is only a few percent. The temperature coefficient of resistivity β [$\beta = (1/\rho) \partial \rho / \partial T$] at room temperature is positive in all alloys except $\text{Ni}_{73}\text{Cr}_5\text{Si}_{10}\text{B}_{12}$. The positive β is indicative of either $2k_F \neq k_p$ (where k_F is the radius of the Fermi sphere and k_p is the position of the first peak of the structure factor), or the Ziman theory²⁴ and its extensions²⁵ do not hold for these alloys. However, assuming the number of conduction electrons n_e for the transition metals and glass formers to be 1 and 4, respectively, it is found that $2k_F \neq k_p$. It is clearly seen in Fig. 13 that 5 at. % Cr causes β to change its sign from positive in $\text{Ni}_{78}\text{Si}_{10}\text{B}_{12}$ to

FIG. 12. Temperature dependence of resistivity in amorphous $\text{Fe}_{78}\text{Si}_{10}\text{B}_{12}$, $\text{Fe}_{78}\text{Cr}_5\text{Si}_{10}\text{B}_{12}$, $\text{Fe}_{37}\text{Ni}_{36}\text{Cr}_5\text{Si}_{10}\text{B}_{12}$, and $\text{Fe}_{13}\text{Ni}_{60}\text{Cr}_5\text{Si}_{10}\text{B}_{12}$ alloys.

FIG. 13. Resistivity curves in amorphous $\text{Ni}_{78}\text{Si}_{10}\text{B}_{12}$ and $\text{Ni}_{73}\text{Cr}_5\text{Si}_{10}\text{B}_{12}$ alloys.

negative in $\text{Ni}_{73}\text{Cr}_5\text{Si}_{10}\text{B}_{12}$. This is consistent with other observations²⁶ which show that the addition of several percent Cr to $\text{Pd}_{80}\text{Si}_{20}$ leads to crossover from positive to negative β . The negative β together with the weak maximum in resistivity observed around 15 K can be explained with the ordinary spin freeze-out of the Kondo²⁷ effect due to Cr spins. It is worthwhile pointing out that the temperature where the maximum in $\rho(T)$ occurs is very close to the estimated cluster ordering temperature for this alloy (Table I).

The resistivity minima observed at low temperatures in the strongly ferromagnetic Fe-Ni amorphous alloys can be explained by a modified Kondo mechanism as proposed by Grest and Nagel.²⁸ It has been shown, in a concentrated ferromagnetic alloy of the transition-metal-metalloid type, that there is an internal-field distribution function, $p(H)$, which is large for large values of H , but which has a long tail at the low field end and is small but nonzero at $H=0$. The spins which exist in zero field will give rise to a Kondo minimum in resistivity. The temperature of the resistivity minimum initially increases by increasing the Ni concentration and then decreases. The increase is consistent with the previous model since by diluting the alloy, the tail in $p(H)$ becomes more pronounced and extends to more negative fields. Therefore, the Kondo effect will be more pronounced and the Kondo minimum will shift to higher temperatures. However, the subsequent de-

crease in resistivity minimum temperature is not clear at present. Similar behavior has been observed in $(\text{Fe}_{1-x}\text{Mn}_x)_{75}(\text{AlBP})_{25}$ amorphous alloys.²⁹

IV. SUMMARY AND CONCLUSIONS

This study has focussed on the effects of Cr on the magnetic and electron-transport properties of glasses of the form $(\text{Fe-Ni})_{78-y}\text{Cr}_y(\text{Si-B})_{22}$. For large Fe concentrations the magnetism is ferromagnetic in character with local moments on Cr atoms which couple antiferromagnetically with the Fe moments. For small or zero Fe concentrations, the Cr local moments couple to each other via the conduction electrons and partially perhaps via direct *d-d* interactions. This coupling, which is on average antiferromagnetic in character, leads to susceptibility and resistivity anomalies at low temperature which are similar to those seen in conventional spin-glass or mictomagnets. However, the magnetic properties of $\text{Ni}_{73}\text{Cr}_5(\text{Si-B})_{22}$, for example, indicate that the Cr moments in part are due to ferromagnetic clusters which then coupled with each

other and perhaps with "isolated" Cr-atom moments. Thus these alloys are probably best described as magnetic cluster glasses. Other similar magnetically dilute glasses such as $\text{Zr}_{40}\text{Cu}_{60-x}\text{Fe}_x$ and $\text{Nb}_{50}\text{Ni}_{50-x}\text{Fe}_x$ were discussed by Gruzalski and Sellmyer.³⁰

In the Fe-rich strongly-ferromagnetic alloys there are low-temperature resistance minima that are consistent with the modified Kondo mechanism as suggested by Grest and Nagel. The addition of Cr to the ferromagnetic alloys makes them much more magnetically hard than typical transition-metal-metalloid glasses. It was argued that the most likely origin of the high coercivities in these alloys is exchange anisotropy associated with the coupling of local moments on Cr atoms.

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