Reentrant antiferromagnet-(spin glass) transition in disordered alloys of fcc iron

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A double temperature transition, from a paramagnet to an antiferromagnet to a spin glass, has been found in disordered fcc FeNiCr alloys.

"Reentrant" phenomena in systems with competing exchange have been the subject of a fair number of studies. In the overwhelming majority of cases, however, both the theoretical studies and the experimental studies have focused on double temperature transitions from a paramagnet to a ferromagnet to a spin glass (PFS).[1,2] Some calculations carried out in the Heisenberg model and the Ising model[4] show that a spin-glass phase may also arise as a result of evolution of the magnetic structure of a disordered antiferromagnet as it cools. However, the experiments in which attempts have been made to study paramagnet-antiferromagnet-(spin glass) transitions[5–7] have been only sporadic and do not clarify the question.

In the present study, of disordered antiferromagnetic alloys of the system $Fe_C Ni_{80-C} Cr_{20}$, we have shown (for the first time) that a double temperature transition from a paramagnet to an antiferromagnet to a spin glass (PAS) does indeed occur in a certain composition region in a zero magnetic field. In alloys in which the random exchange plays a lesser role, a transition of this sort would be possible only in a sufficiently strong magnetic field.

To determine the possibility of a PAS temperature transition, we made use of one of the basic properties of a spin glass: the appearance of irreversible phenomena below its freezing point T_f. For very single spin glass, at measurement times $\sim 10^2$ s, one observes a thermomagnetic "past-history" effect[2]: After cooling below T_f in a magnetic field (FC), the magnetization of the spin glass is above its magnetization after cooling in a zero field (ZFC). The temperature at which the FC and ZFC magnetizations are equal is the freezing point in the given magnetic field.[8] $T_f(H)$.

Figure 1 illustrates the results with the temperature dependence of the ZFC magnetization (open circles) and that of the FC magnetization (field circles) found for the alloy $Fe_{66}Ni_{14}Cr_{20}$ in magnetic fields of various strengths. We see that in a field $H = 0.5$ kOe, for example, below $T = 11$ K, the magnetization becomes irreversible. In accordance with the discussion above, this temperature can be identified as the freezing point T_f of the spin glass at the given H. As the magnetic field applied to the sample is increased, T_f increases monotonically (as shown by the broken arrows). We emphasize that this behavior is fundamentally different from the behavior $T_f(H)$ for systems with paramagnet-(spin glass) and ferromagnet-(spin glass) transitions. In the latter case, T_f always decreases with increasing H (Refs. 2 and 8).

We might note that, as the magnetic field is increased, there is an anomalously rapid decrease in the temperature (T_N) of the maximum magnetization I (marked by

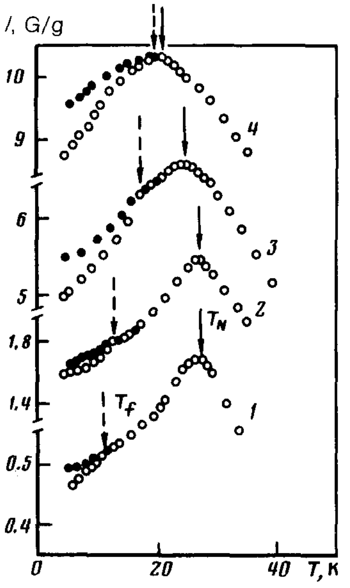

FIG. 1. Temperature dependence of the zero-field-cooling magnetization (open circles) and of the field-cooling magnetization (field circles), I, of the alloy $Fe_{66}Ni_{14}Cr_{20}$ in magnetic fields of various strengths: 1—0.5; 2—1.8; 3—6; 4—10 kOe (T_N is the Néel temperature, and T_f is the freezing point of the spin glass).

the solid arrows), due to the onset of an antiferromagnetic order in the alloy, according to neutron-diffraction data.[9] It is clear from Fig. 1 that the difference $T_N - T_f$ should decrease with increasing field. Finally, at $H > 10$ kOe the nature of the temperature dependence $I(T)$ is the same as that for ordinary spin glasses. Consequently, for this alloy at $H > 10$ kOe the lines of $T_f(H)$ and $T_N(H)$ in the H, T plane should intersect. The situation can be illustrated by the corresponding phase diagram in Fig. 2a, from which we see that in a zero magnetic field a double PAS temperature transition does in fact occur. With increasing field, the temperature region in which the spin glass exists (3) expands, while that of the antiferromagnetic phase (2) contracts. Above the critical magnetic field $H_c \simeq 11$ kOe, there is only a transition from the paramagnetic phase (1) to the spin glass.

The concentration (C_0) at which the antiferromagnetic order arises in the alloys of the system $Fe_CNi_{80-C}Cr_{20}$ is in the range[9] 62–63 at. %. At $C < C_0$, these alloys

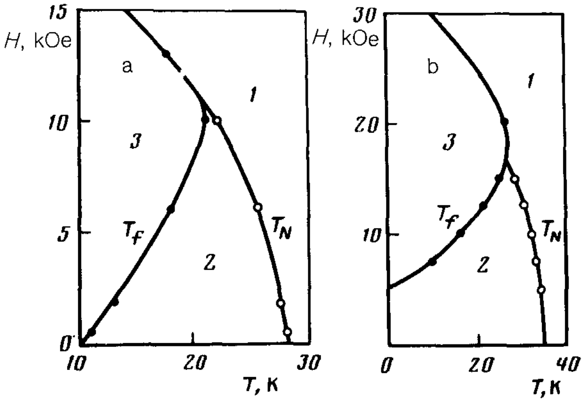

FIG. 2. H, T phase diagrams of the alloys (a) $Fe_{66}Ni_{14}Cr_{20}$ and (b) $Fe_{68}Ni_{12}Cr_{20}$. 1, 2—Paramagnetic and antiferromagnetic regions; 3—spin glass.

typically exhibit paramagnet–(spin glass) temperature transitions.[10] In the alloys studied above, on the other hand, the iron concentration is only slightly above C_0, so that a competing exchange plays an important role. It is interesting to determine whether a PAS temperature transition occurs in $Fe_C Ni_{80-C} Cr_{20}$ alloys, in which the contribution of random exchange to the total exchange energy is smaller. Figure 2b shows an H, T phase diagram of the alloy $Fe_{68}Ni_{12}Cr_{20}$, from which we see that only the paramagnet-antiferromagnet transition occurs at $H = 0$. In the magnetic field interval $H_0 \simeq 5 < H < H_c \simeq 17$ kOe, however, as in the case discussed above, there is a PAS transition. It is quite understandable that as the contribution of the antiferromagnetic interaction becomes even greater, with a simultaneous decrease in the relative importance of random exchange, the fields H_0 and H_c will increase, and the spin-glass region (3) will disappear in the limiting case of a "pure" antiferromagnetic.

It should be noted that the conclusion which we reached above regarding the existence of the PAS transition in these alloys is based on measurements of the static ZFC and FC magnetizations. For a spin glass, it has been customary to study the dynamic susceptibility. Figure 3 shows the temperature dependence of the real part χ_0' and the imaginary part χ_0'' of the dynamic susceptibility of the alloy $Fe_{66}Ni_{14}Cr_{20}$. As expected, $\chi_0'(T)$ has a maximum at the Néel temperature $T_N = 29$ K and a slight anomaly in the form of an inflection point near $T_f = 9$ K. In order to detect phase transitions in magnetically ordered phases, it is far more convenient to study the

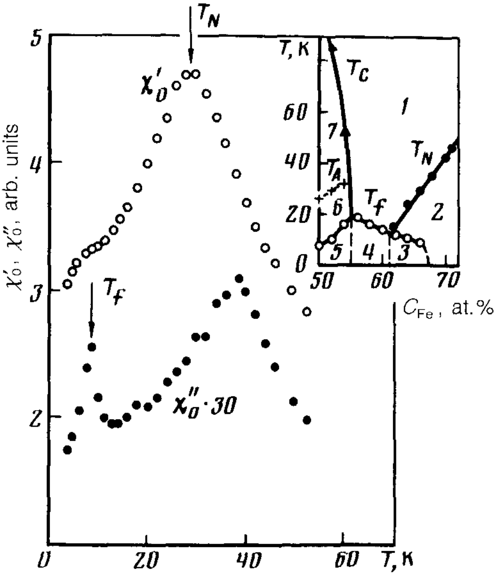

FIG. 3. Temperature dependence of the real and imaginary parts, x_0' and x_0'', respectively, of the dynamic magnetic susceptibility of the alloy $Fe_{66}Ni_4Cr_{20}$ in a magnetization-reversing field of 10 Oe at the frequency 240 Hz. The inset shows the complete magnetic phase diagram of the alloys $Fe_C Ni_{80-C} Cr_{20}$; T_c, T_N, and T_f are the Curie temperature, the Néel temperature, and the freezing point of the spin glass; T_A is the temperature at which the asperomagnetic state arises. 1—paramagnet; 2—antiferromagnet; 3, 4, 5—regions in which a spin glass exists; 6 and 7—asperomagnetic and ferromagnetic regions.

imaginary susceptibility, $\chi_0''(T)$, which describes the dynamics of the magnetic moments of a system.[11] We see from Fig. 3 that there is a sharp anomaly on the $\chi_0''(T)$ curve near T_f, which is evidence of a transition of the alloy to a spin-glass state. An important point is that the temperature T_f found from the measurements of $\chi_0''(T)$ agrees well with the value found by extrapolating the $T_f(H)$ curve to a zero field (Fig. 2a).

We turn now to the complete magnetic phase diagram of the $Fe_CNi_{80-C}Cr_{20}$ alloys (the inset in Fig. 3), constructed from the data of the present study and earlier studies.[10,11] In this diagram, the composition regions in which the double temperature transitions PFS (1–7–5) and PAS (1–2–3) typically occur have been identified experimentally for the first time. We will not go into a discussion here of the nature of the spin-glass phase which results from the latter transition or possible manifestations of random-field effects. These questions will be discussed in detail in a future paper.

We note in conclusion that the basic results of this study—the shape of the complete magnetic phase diagram and of the H, T diagrams for antiferromagnetic $Fe_CNe_{80-C}Cr_{80}$ alloys—agree qualitatively with the theoretical predictions of Ref. 4.

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[1] K. H. Fischer, Phys. Status Solidi **b116**, 357 (1983); **b130**, 13 (1985).
[2] C. Y. Huang, J. Magn. Magn. Mater. **51**, 1 (1985).
[3] M. V. Medvedev and S. M. Goryanova, Phys. Status. Solidi **b98**, 143 (1980).
[4] I. Ya. Korenblit and E. F. Shender, Zh. Eksp. Teor. Fiz. **89**, 1785 (1985) [Sov. Phys. JETP **62**, 1030 (1985)].
[5] T. Datta, D. Thornberry, E. Jones, and H. M. Ledbetter, Solid State Commun. **52**, 515 (1984).
[6] G. V. Lecomte, H. Lohneysen, W. Bauhofer, and G. Guntherodt, Solid State Commun. **52**, 535 (1984).
[7] J. Gelard, F. Bensamka, D. Bertand, A. R. Fert, J. P. Redoules, and S. Legrand, J. Phys. C **16**, L939 (1983).
[8] G. A. Takzei, A. M. Kostyshin, and Yu. P. Grebenyuk, Fiz. Tverd. Tela (Leningrad) **26**, 2722 (1984) [Sov. Phys. Solid State **26**, 1648 (1984)].
[9] A. K. Majumdar and P. J. Blanchenhagen, J. Magn. Mater. **40**, 227 (1983).
[10] A. Z. Men'shikov, G. A. Takzei, and A. E. Teplykh, Fiz. Met. Metalloved. **54**, 465 (1982).
[11] G. A. Takzei, and A. M. Kostyshin, Pis'ma Zh. Eksp. Teor. Fiz. **40**, 308 (1984) [JETP Lett. **40**, 1099 (1984)]; G. A. Takzei, A. M. Kostyshin, Yu. P. Grebenyuk, and I. I. Syts, Zh. Eksp. Teor. Fiz. **89**, 2181 (1985) [Sov. Phys. JETP **62**, 1259 (1985)].

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