Antiferromagnetism and reentrant spin glass in disordered Ni–Mn alloys

A. D. Gezalyan and s. V. Shul'pekova
I. P. Bardin Central Scientific-Research Institute of Ferrous Metallurgy, 107005, Moscow

(Submitted 29 April 1991; resubmitted 3 June 1991)
Pis'ma Zh. Eksp. Teor. Fiz. **54**, No. 1, 48–51 (10 July 1991)

The formation of a reentrant spin glass from an antiferromagnetic phase has been discovered in Ni–Mn alloys by neutron diffraction analysis and measurements of the magnetic susceptibility. A cluster mechanism has been identified as responsible for the formation of the spin glass from adjacent magnetic states.

In the Ni–Mn system we would expect to find a ferromagnetic–antiferromagnetic transition induced by a change in concentration and accompanied by the formation of a spin glass. The spin glass in the course of a temperature-induced paramagnet–ferromagnet–(spin glass) transition was studied in Refs. 1 and 2 from the standpoint of mixed exchange interactions. In contrast, there has been essentially no experimental study of the reentrant spin glass in the course of paramagnet–antiferromagnet–(spin glass) transitions. The magnetic properties of alloys of the Ni–Mn system are very sensitive to the atomic order.[3]

Studies by neutron diffraction show that quenched alloys acquire regions in which a short-range atomic and magnetic order is retained. The neutron diffraction patterns in Fig. 1 reflect the growth of a short-range order, which is evidence that the anitferromagnetic intermetallic compound NiMn is contributing to the intensity. These ordered regions may be either ferromagnetic or antiferromagnetic clusters, de-

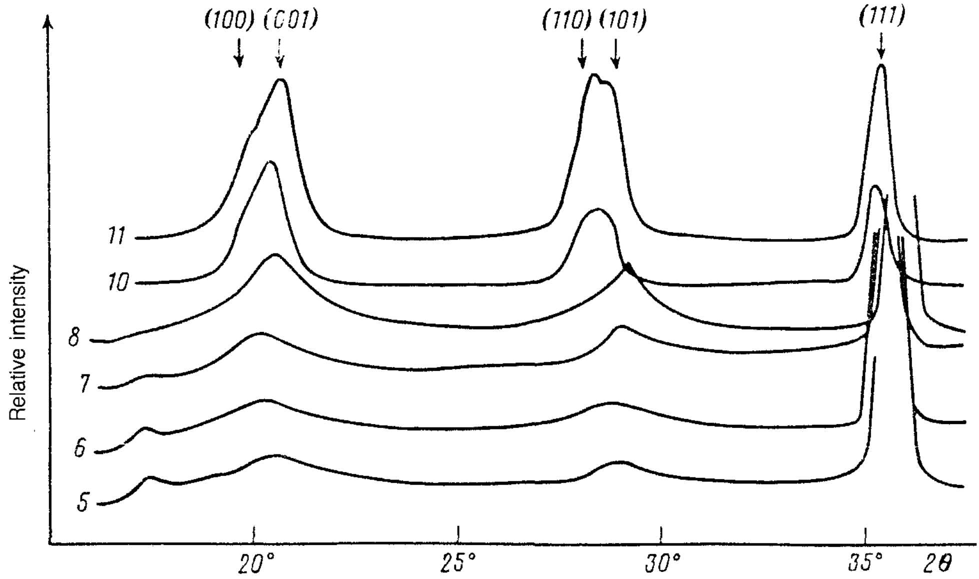

FIG. 1. Neutron diffraction patterns of quenched alloys. *5*—29.0% Mn; *6*—30.9% Mn; *7*—33.3% Mn; *8*—35.4% Mn; *10*—41.7% Mn; *11*—45.4% Mn.

pending on the Mn content. With increasing concentration, $c_{\mathrm{Mn}} > 25\%$ (here and everywhere the percentages are atomic), antiferromagnetic clusters are created along with the ferromagnetic clusters, because of a prior precipitation of the intermetallic compound NiMn. The latter compound is an antiferromagnet with an anomalously high Néel temperature, $T_N = 1073 \pm 40$ K. We will thus treat the spin glass as a cluster spin glass here.

In a model proposed by Kouvel[4] for interacting ferromagnetic and antiferromagnetic clusters, a frozen state arises as a result of a blocking of the magnetic moments of superparamagnetic clusters in exchange-anisotropy fields. In Ni–Mn alloys, composition fluctuations may lead to the formation of regions of various sizes which are enriched in Mn (antiferromagnetic regions) and Ni (ferromagnetic regions). The antiferromagnetic regions have a more pronounced anisotropy.[5] At high temperatures, these regions behave as superparamagnets. During cooling, large antiferromagnetic regions arise first. Because of the strong exchange interaction at the interface between ferromagnetic and antiferromagnetic clusters, there is a blocking of the magnetic moments of the ferromagnetic clusters which accompany the appearance of magnetic irreversibilities. A further lowering of the temperatures leads to the realization of smaller antiferromagnetic regions, so new ferromagnetic clusters are blocked in the exchange-anisotropy fields.

It has been demonstrated for the first time here that a temperature-induced paramagnet–antiferromagnet–(spin glass) transition can occur in Ni–Mn alloys near the critical concentration for the onset of a long-range antiferromagnetic order. Figure 2a

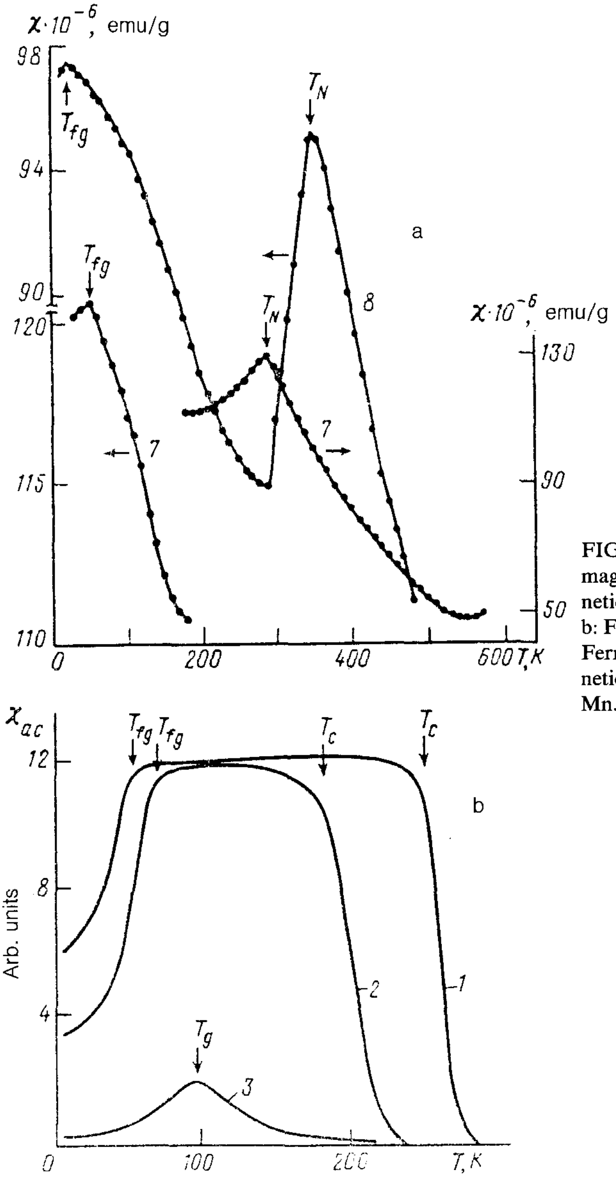

FIG. 2. Temperature dependence of the magnetic susceptibility. a: Antiferromagnetic alloys. *7*—33.3% Mn; *8*—35.4% Mn. b: Ferromagnetic alloys and spin glass. *1*—Ferromagnetic, 21.2% Mn; *2*—ferromagnetic, 23.1% Mn; *3*—spin glass, 25.3% Mn.

shows the temperature dependence of the magnetic susceptibility measured by the Faraday method. These results demonstrate a coexistence of a long-range antiferromagnetic order and a spin glass. The paramagnet–ferromagnet–(spin glass) transition can be detected by measuring the differential magnetic susceptibility χ_{ac} as a function of the temperature in a weak magnetic field ($H_0 = 0.5$ Oe; Fig. 2b), because of the fairly strong interactions of the ferromagnetic clusters. With increasing c_{Mn}, however, these ferromagnetic clusters break up into small clusters, which coexist with growing antiferromagnetic clusters. The result is to hinder a detection of the paramagnet–antiferromagnet–(spin glass) transition in a weak field. It was only through measurements of the magnetic susceptibility χ by the Faraday method in a strong magnetic field [1.8 kOe and 2.4 kOe, for samples 7 and 8 (Fig. 2a)] that it was found possible to detect the paramagnet–antiferromagnet–(spin glass) transition. The value of χ at the peak at the temperature T_{fg} in sample 8 is lower than the corresponding value for sample 7, confirming that the frozen ferromagnetic clusters are small. Neutron diffraction has revealed[6] an antiferromagnetism in polycrystalline samples at $c_{\mathrm{Mn}} \geqslant 5\%$, but the Néel temperature T_N has not been determined because of the small Bragg peak and the obvious temperature dependence of the diffuse scattering. In the present study, along with detecting the antiferromagnetism we have also determined the value of T_N and the freezing point of the spin glass, T_{fg} (for 7 and 8). We have simultaneously refined the value of $c_{\mathrm{Mn}}^{\mathrm{cr}}$, at which the antiferromagnetism appears.

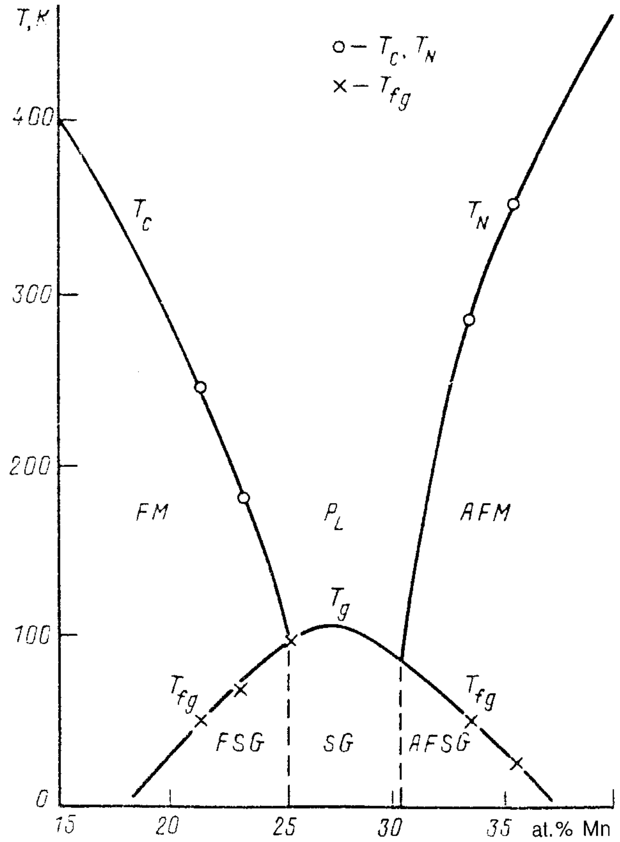

FIG. 3. Magnetic phase diagram of disordered Ni–Mn alloys. FM—Ferromagnetic phase; AFM—antiferromagnetic phase; P_L—Langevin paramagnetic phase; FSG—ferromagnet and spin glass; SG—normal spin glass; AFSG—antiferromagnet and spin glass.

Calculations of the Curie temperature T_C of the ferromagnetic phase from the formula in Ref. 7 yield results in good agreement with the experimental data ($T_C = 246$ K and 182 K for samples 1 and 2; Fig. 2b).

Figure 3 shows the complete magnetic phase diagram. The ferromagnetic part of this diagram agrees with the data of Ref. 1. We see that as the Langevin paramagnet is cooled down, it ultimately goes into a state of either a normal (or ordinary) spin glass or a reentrant spin glass, depending on c_{Mn}.

In summary, this study verifies the concept of a cluster spin glass in the formation of a long-range atomic order. It has yielded the first observation of a reentrant spin glass in Ni–Mn alloys at $c_{\mathrm{Mn}} > 30\%$

We wish to thank V. I. Goman'kov, B. N. Tret'yakov, and V. V. Sumin for a discussion of these results and for assistance in the experiments.

[1]W. Abdul-Razzag and J. Kouvel, Phys. Rev. B. **35**, 1764 (1987).
[2]Yu. P. Grebenyuk, M. V. Gavrilenko *et al.*, Preprint, Institute of Metal Physics, Kiev, 1989.
[3]V. I. Goman'kov, A. D. Gezalyan, B. N. Tret'yakov *et al.*, Fiz. Met. Metalloved., No. 12, 49 (1990).
[4]J. J. Kouvel, Phys. Chem. Solids **24**, 795 (1963).
[5]L. Néel, Adv. Phys. **4**, 191 (1955).
[6]O. Moze, T. Hicks, and P. J. Blanckenhagen, Magn. Magn. Mat. **42**, 103 (1984).
[7]H. Tange, T. Tokunaga, and M. Goto, J. Phys. Soc. Jpn. **45**, 105 (1978).

Translated by D. Parsons