Magnetic properties of systems near a concentrational ferro-antiferromagnetic transition

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The parameters of the spin-density fluctuations that accompany a concentrational ferro-antiferromagnetic transition in systems based on iron are obtained using magnetic small-angle neutron scattering, and are used to determine the concentration and temperature dependences of the saturation magnetization. The magnetic-properties anomalies that arise in the transitions are well described with the framework of a first-order phase transition.

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INTRODUCTION

Concentrational magnetic phase transitions, which are of first order, are accompanied by formation of spin-density fluctuations whose parameters determine the diversity of many magnetic properties of alloys. Investigations of these properties by traditional magnetic method yield averaged density and temperature dependences of the magnetic characteristics, thereby masking the true magnetic state of this system and obscuring the physical causes of the appearance of various anomalies. This is precisely why numerous models were proposed for systems based on iron and nickel, where concentrational ferro-antiferromagnetic transitions occur.[1,2] These models were called upon to explain the anomalies of the magnetic properties without taking into account the parameters of the spin-density fluctuations.

In this paper we use a cluster model of the transition to describe the magnetic properties of an iron + nickel system and of $Fe_{65}(Ni_{1-x}Mn_x)_{35}$ near the critical concentration c_0 of the transition.

It is assumed that the seeds of the antiferromagnetic state in the ferromagnetic matrix are atom clusters that do not contain nickel atoms in the first coordination sphere around the iron and manganese atoms. At the temperature 0 K these clusters are in a state of short-range or local antiferromagnetic order and perturb the surrounding matrix, causing a decrease of the magnetic moments of its atoms that spans 10–12 coordination spheres[3] and producing thereby static fluctuations of the spin density.

The fluctuation is characterized by an average total decrease of the magnetic moment $Q(0)$, and by a spin correlation parameter $\varkappa$; these are determined from the cross sections $(d\sigma/d\Omega)_0$ for small-angle magnetic scattering of the thermal neutrons.[3,4]

With decreasing nickel content c_{Ni}, the concentration C of the fluctuations and their parameters $|Q(0)|$ and $1/\varkappa$ increase, and at c_0 the long-range ferromagnetic order is destroyed. Below c_0, the fluctuations give rise to a long-range antiferromagnetic order and to production of an antiferromagnetic matrix.

CONCENTRATION DEPENDENCES OF AVERAGE MAGNETIC MOMENTS $\bar{\mu}$

The mechanism acting in the proposed model is nucleation and growth of a new antiferromagnetic state in a ferromagnetic matrix. The quantity $\bar{\mu}$ can be expressed in terms of the linear Slater-Pauling relation μ_0, which describes the unperturbed part of the matrix, and a "fluctuation" term:

$$\bar{\mu}=\mu_0-C|Q(0)|. \tag{1}$$

Here $\mu_0=c_{Ni}\mu_{Ni}+c_{Fe}\mu_{Fe}$ with $\mu_{Ni}=0.6\mu_B$ and $\mu_{Fe}=2.8\mu_B$.

According to the foregoing model, the values of C are calculated using the formula for the binomial distribution of the atoms, with allowance for the short-range atomic-order parameters α_1:

$$C=c_{Fe}(c_{Fe}+c_{Ni}\alpha_1)^{12}. \tag{2}$$

The concentration dependences of $|Q(0)|$ for Fe–Ni and $Fe_{35}(Ni_{1-x}Mn_x)_{35}$ systems are shown in Fig. 1. The increase of $Q(0)$ with decreasing c_{Ni} reflects the connection between the parameters $|Q(0)|$ and $\varkappa$, indicated in

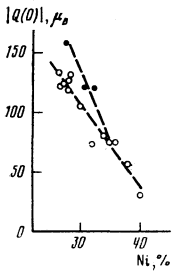

FIG. 1. Concentration dependence of the average total decrease of the magnetic moment in the fluctuation of $|Q(0)|$ at 4.2 K; ○—Fe - Ni system, ●—$Fe_{65}(Ni_{1-x}Mn_x)_{35}$.

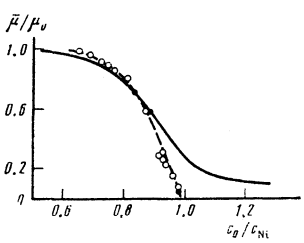

FIG. 2. Concentration dependence of the average magnetic moment $\bar{\mu}/\mu_0$; μ_0 is the linear Slater-Pauling dependence; the symbols are the same as in Fig. 1.

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Ref. 5: $|Q(0)| = 4\pi A I_s \varkappa^{-2}$ (here A is a constant and I_s is the saturation magnetization); this relation is well satisfied within the limits of the measurement errors of $(d\sigma/d\Omega)_0$ and $\varkappa$ (Refs. 3 and 4), and indicates that the fluctuations increase in the systems when c_0 is approached.

The concentration dependence of $\bar{\mu}$, obtained using Eqs. (1) and (2) as well as the data of Fig. 1, is shown in Fig. 2. The values of c_0 were determined experimentally with $\bar{\mu}/\mu_0$ extrapolated to the concentration axis. They are equal to 26% Ni and 27% Ni for the Fe–Ni and $Fe_{65}(Ni_{1-x}Mn_x)_{35}$ systems, respectively, and agree with other measurements.[4] The solid line shows the results of Refs. 2 and 6, obtained by the usual magnetic methods. Figures 1 and 2 show that the deviation of $\bar{\mu}$ from the linear dependence of μ_0 with decreasing c_{Ni} is due to the fluctuation term of Eq. (1).

Figure 2 demonstrates the decrease of the degree of long range ferromagnetic order in the concentrational transformation, and the values of $\bar{\mu}$ measured by both methods are in good agreement for compositions with $c_{Ni} < 28\%$. The density in the values of $\bar{\mu}$ as c_0 is approached is due to the influence exerted on the parameter $\varkappa$ by the external magnetic field used in the magnetic measurements in Ref. 2, whereas the values of $(d\sigma/d\Omega)_0$ were obtained in Refs. 3 and 4 without application of magnetic fields.

Since a long-range antiferromagnetic order is produced from the fluctuations in the transition, the fluctuation itself can be represented as a cluster with a short-range antiferromagnetic order, surrounded by spin glass without a sharp boundary between the fluctuation and the ferromagnetic matrix.[7] The spin glass is produced in this case as a result of the reversal of the sign of the exchange interaction, from negative in the cluster to positive in the matrix.

The external magnetic field decreases the fluctuations,[7] thus limiting the possibility of "leakage" of the antiferromagnetic short-range order to larger volumes, i.e., its transformation into long-range order. It consequently shifts c_0, so that a ferromagnetic-order "tail," similar to the tails of the spontaneous magnetization above the Curie temperatures,[8] is observed for the magnetic measurements of Fig. 2.

TEMPERATURE DEPENDENCES OF THE SATURATION MAGNETIZATION

With increasing temperature, the magnetization of the unperturbed matrix decreases in the usual manner, but the change of the fluctuation parameters is complicated, since the clusters first go over into the paramagnetic state ("unfreezing") and then the fluctuation behaves like a paramagnetic impurity, increasing in size.[3] The behavior of the latter at small values of C was considered in Ref. 5, and two competing effects were found to affect the temperature dependence of $|Q(0)|$. The decrease of I_s with increasing temperature leads to a decrease of $|Q(0)|$, but the increase of the size of the fluctuations, like $\varkappa^{-2}$, increases $|Q(0)|$. The second effect exceeds the first considerably, causing a deviation of the temperature dependence of the magnetization from the Brillouin curve in such magnetically inhomogeneous systems.

The average magnetization atom as a function of temperature takes according to Eq. (1), for a given composition, the form

$$M(T) = M_0(T) - C|Q(0)|, \tag{3}$$

where $|Q(0)|$ is also a function of temperature. The magnetization of the unperturbed ferromagnetic matrix is given at low temperatures by

$$M_0(T) = \mu_0[1 - aT^{3/2} - bT^2], \tag{4}$$

where the terms a and b are the coefficients of the temperature dependences due respectively to the spin waves and the Stoner excitations.

For a matrix with 40% Ni and negligibly small number of fluctuations ($C = 0.08\%$) the values of a and b were obtained in Ref. 9 and will be used in the calculations that follow for all the investigated compositions.

The low-temperature parts of the temperature dependences of $|Q(0)|$ are plotted for three compositions in Fig. 3 on the basis of measurements of $(d\sigma/d\Omega)_0$.[3,4] For compositions with $c_{Ni} > 32\%$ (curve 1), $|Q(0)|$ increases monotonically with temperature, as predicted theoretically for a ferromagnetic system with a small concentration of nonmagnetic impurities.[5] For samples with $c_{Ni} \leq 32\%$ (curves 2 and 4) one can see anomalous increases of $|Q(0)|$ with decreasing temperature. This, together with the decrease of the parameter $\varkappa$, offers evidence of the spin depolarization of the ferromagnetic matrix on the boundaries with the fluctuations as they "freeze".[4]

The solid lines in Fig. 4 show the temperature dependences of $M(T)$ [Eq. (3)], obtained using the results of Fig. 3 and of Ref. 4. For compositions with $c_{Ni} \geq 32\%$, good agreement is observed between the $M(T)$ dependences and the data of Refs. 6 and 9 (circles) measured by the usual methods.

Curves 3–5 of Fig. 4 show the anomalous decrease of the magnetization with decreasing temperature, which is typical of the "mictomagnetic" behavior of the system.[10] It is due to the growth of the fluctuation term in Eq. (3) with decreasing temperature (Fig. 3). With increasing c_{Ni}, the anomaly on the $M(T)$ curves begins to be masked, both on account of the decrease

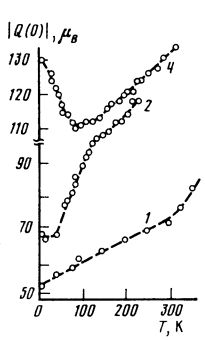

FIG. 3. Temperature dependences of $|Q(0)|$ of the system Fe – Ni: 1—38% Ni; 2—32% Ni; 4—28% Ni.

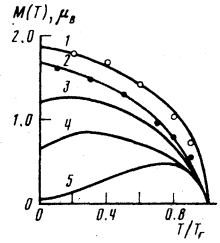

FIG. 4. Temperature dependences of the saturation magnetization: 1—38% Ni; 2—32% Ni; 3—30% Ni; 4—28% Ni, 5—6.8% Mn.

of C and on account of the enhancement of the magnon scattering of the neutrons which lower the sensitivity of the neutron-diffraction method in the measurements of $|Q(0)|$. For these reasons, the decrease of $M(T)$ for the composition $c_{Ni}=32\%$ at $T<30$ K (curve 2) lies within the limits of the errors of the measurement of $M_0(T)$.

The temperature dependence of $M(T)$ of the Fe–Ni system at $c_{Ni}\leq 32\%$ cannot be measured by magnetic methods because of the martensitic transformation. Curves 3 and 4 are therefore experimental plots of $M(T)$ of the γ phase, first obtained in a zero magnetic field in the presence of an α phase in the samples.[4]

Similar $M(T)$ plots were obtained for ferromagnetic samples of the $Fe_{65}(Ni_{1-x}Mn_x)_{35}$ system[4] which did not undergo a martensitic transformation. Curve 5 (Fig. 4) pertains to the temperature dependence of $M(T)$ of an alloy with 6.8% Mn, whose composition is close to c_0. It is compared with the temperature dependences of the magnetization B of Fig. 5, measured by the Faraday method at a fixed value of the external magnetic field. The decrease of the magnetization in a field of 300 Oe with decreasing temperature from 100 K, shown in Fig. 5, agrees with the corresponding decrease of $M(T)$ in Fig. 4.

For a boundary composition with $c_{Ni}=35\%$, the anisotropy field $H_a=\frac{4}{3}K_1/I_s$ ($K_1=6.7\times10^4$ erg/cm^3 is the anisotropic constant[11] and $I_s=1400$ G) is equal to 64 Oe. For the composition with 6.8% Mn we have $I_s=1400$ G, is equal to 64 Oe. For the composition with 6.8% Mn

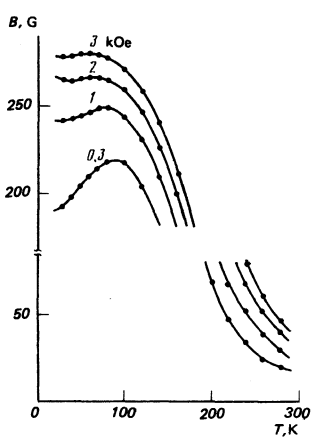

FIG. 5. Temperature dependence of the saturation magnetization of the alloy with 6.8% Mn.

TABLE I.

H, kOe	T_{max}, K	H, kOe	T_{max}, K
0.3	90	2	70
1.0	80	3	60

we have $I_s=350$ G, and at the same value of K_1 we have $H_a=250$ Oe. One can expect, however, the doping with manganese to make K_1 much smaller,[12] therefore H_a is patently over-estimated. Hence, the decrease of B with decreasing temperature, observed in fields up to 3 kOe (Fig. 5), is not due to the anisotropy field.

An investigation of the rotational hysteresis[13] shows that an alloy with this composition has the maximum unidirectional anisotropy. Our measurements of the magnetization at 4.2 K after cooling the samples from 300 to 4.2 K in a field 50 kOe shows a shift of the hysteresis loop near 300 Oe, which is also evidence of considerable unidirectional anisotropy.

Temperature dependences similar to the curves of Fig. 5 were obtained in Ref. 14 for the system Fe–Ni–Mn without any connection with the fluctuation parameters.

Thus, the observed decrease of the magnetization at low temperatures, shown in Figs. 4 and 5, is due to freezing of the fluctuations. The freezing temperature T_{max} depends on the external magnetic field, as reflected in Table I.

The influence of the magnetic field on the sizes of the fluctuations takes place primarily on the periphery of the fluctuations, where formation of spin glass is assumed. When cooled in a magnetic field, the spins of the periphery of the fluctuations "align themselves" with the orientation of the spins of the ferromagnetic matrix, and the fluctuations decrease in size. In sufficiently strong magnetic fields the anomalous low-temperature decrease of the magnetization vanishes (Fig. 5), since the concentration of the antiferromagnetic clusters is on the whole small ($C<2\%$). The antiferromagnetic clusters themselves are found to be sharply demarcated from the matrix, and this leads to the shift of the hysteresis loop.

Since the shift of the hysteresis loop is observed in sufficiently strong magnetic fields,[11,14] it must be assumed that the antiferromagnetic clusters have a $3d$-electron structure (apparently of the γ-Fe type), which is entirely different from the electronic structure of the matrix.

CONCLUSION

A study of the concentration and temperature dependences of the parameters of the fluctuations of the spin density in a ferromagnetic matrix has shown that the number and dimensions of the seeds of the short-range antiferromagnetic order influence substantially the concentration and temperature dependences of the saturation magnetization. Thus, the concentration dependences of the fluctuation parameters at 4.2 K describe

quantitatively the anomalous deviation of the values of $\bar{\mu}$ from the Slater-Pauling linear relation. In turn, the temperature dependences of the fluctuation parameters reveal the physical causes of the anomalously low temperature-dependent decrease of the magnetization and shift of the hysteresis loop, which are due to effects of freezing of the fluctuations.

In an antiferromagnetic matrix, below c_0, one observes also fluctuations of the spin density,[4] in the centers of which is produced a short-range ferromagnetic order. Their parameters and concentration can be used to describe the magnetic properties of antiferromagnetic alloys.

The foregoing analysis of the concentrational ferro-antiferromagnetic transition within the framework of a first-order phase transition only implies the presence of a mixed exchange interaction on the boundary between the fluctuation and the matrix.

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[1]Y. Nakamura, IEEE Trans. Magn. Mat. **12**, 278 (1976).
[2]S. Kochi and H. Asano, J. Phys. Soc. Jpn. **37**, 536 (1969).
[3]V. I. Goman'kov, E. V. Kozis, and B. N. Mokhov, Zh. Eksp. Teor. Fiz. **70**, 327 (1976) [Sov. Phys. JETP **43**, 170 (1976)].
[4]V. I. Goman'kov, and B. N. Mokhov, and N. I. Nogin, *ibid.* **77**, 630 (1979) [**50**, 317 (1979)].
[5]S. W. Lovesey and W. Marshall, Proc. Phys. Soc. **86**, 613 (1966).
[6]J. Crangle and G. C. Hallam, Proc. Roy. Soc. **272**, 119 (1963).
[7]M. Shimizu, J. Magn. and Magn. Mat. **10**, 231 (1979).
[8]K. P. Belov, Magnetic Transitions, Consultants Bureau, 1961.
[9]R. W. Cochrane and G. H. Graham, Canad. J. Phys. **48**, 264 (1970).
[10]P. A. Beek, Progr. Mat. Scien. **23**, 1 (1978).
[11]I. M. Puzei, Fiz. Met. Metallov. **11**, 525 (1961).
[12]U. Hofmann, A. Handstein, and K. Hausmann, Phys. Stat. Sol. **40**, k81 (1970).
[13]Y. Nakamura and N. Miyata, J. Phys. Soc. Jpn. **23**, 223 (1967).
[14]A. Z. Men'shikov, V. A. Kazantsev, and N. N. Kuz'min, Zh. Eksp. Teor. Fiz. **71**, 648 (1976) [Sov. Phys. JETP **44**, 341 (1976)].

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