# Magnetic properties of Fe–Ni–Cr(Mn) spin glasses

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Measurements of the magnetic-field and temperature derivatives of the magnetization are used to determine the $H$-$T$ diagram and to elucidate the nature of the change in the nonlinear magnetic susceptibility in iron–nickel–chromium and iron–nickel–manganese spin glasses. The results of the nonlinear-susceptibility study point to the existence of a phase transition to a "spin glass" state. The magnetic properties of Fe–Ni–Cr(Mn) spin glasses are compared with the theoretical predictions.

## 1. INTRODUCTION

Interest in the spin-glass state continues unabated. The primary reason for this is that the ergodic hypothesis, which is fundamental to statistical mechanics, may not apply to the description of this state.[1,2] The absence of ergodicity has been demonstrated for spin-glass models with an infinite-range exchange interaction.[3] The presence of a large number of equivalent energy minima (valleys) separated by infinite barriers (hills) in the Sherrington–Kirkpatrick spin glass[3] comes about because the minimum number of frustrated bonds in a system with an interaction of alternating sign can be reached in many different ways.[4] Computer modeling of a spin glass with a short-range exchange interaction[5] also points to the existence of an energy relief in the form of hills and valleys[6] (but the barriers between the valleys, although they can be very large, are finite). If the energy barriers between the valleys are infinite, then an averaging over all possible thermodynamic states (valleys) is not equivalent to an averaging over time (having entered a valley, the system remains in it for an infinite time), i.e., the ergodic principle is violated. When the energy barriers between the valleys are very high, but finite, a quasinonergodicity arises—the system can sample all the valleys only over enormous observation times.

The theory incorporating the presence of random degenerate states is very complex and has been adequately developed only for a model with an infinite-range interaction[3] (for which the mean field approximation is exact). Both static[7–9] and dynamic[10,11] versions of the theory of a spin glass with an infinite-range interaction have been proposed, generally yielding the same results. Analytical expressions are obtained[7–11] only near the upper existence boundary of the spin-glass phase on the $H$-$T$ diagram (the de Almeida–Thouless line[12]). In recent years attempts have been made to go beyond the mean field approximation and develop a theory for a spin glass with a finite interaction range (see, e.g., Refs. 13–15).

The subject of the present paper is a comparison of the magnetic properties of the disordered fcc alloys $Fe_{65}Ni_{20}Cr_{15}$, $Fe_{70}Ni_{20}Cr_{10}$, and $Fe_{45}Ni_{35}Mn_{20}$ with the predictions of the theory of spin glasses. These alloys have no long-range order[16,17] and exhibit all the characteristic properties of spin glasses as low temperatures:

a) a sharp maximum (cusp) on the linear magnetic susceptibility at a certain temperature $T_f$; this feature is suppressed by a relatively weak (of the order of $10^3$–$10^4$ A/m) magnetic field[17–20];

b) irreversible magnetic behavior below the freezing point $T_f$: a large magnetic aftereffect and an increase in the coercive force and remanent magnetization on cooling below $T_f$ (Refs. 16, 18, 21, 22);

c) a magnetic contribution to the specific heat that is linear in the temperature for $T \lesssim T_f$ (Refs. 20, 23);

d) a negative magnetoresistance at low temperatures.[24]

Other fcc alloys of the Fe–Ni–Cr and Fe–Ni–Mn systems have also been found to have physical properties similar to those of "classical" (RKKY) spin glasses (see, e.g., Refs. 25, 26). However, the similarity may be purely superficial. A more detailed comparison with the conclusion of the present-day theory of spin glasses (Refs. 7–15, 27–30) is needed. In particular, no one has considered the question of whether there is a paramagnetic-to-spin-glass transition at $T_f$ or the form of the $H$-$T$ diagram in concentrated spin glasses, i.e., alloys between ferromagnetic and antiferromagnetic $3d$ metals.

## 2. EXPERIMENTAL TECHNIQUES AND SAMPLES

The magnetization was measured by a ballistic method. The use of an external digital instrument in conjunction with an F190-1 microfluxmeter made it possible to decrease the error to 0.2–0.5%. Magnetic fields of up to $1.6 \cdot 10^5$ A/m were produced with a copper-wound solenoid. The vertical component of the geometric field was compensated to within $\pm$ 1 A/m. To decrease the error in the determination of the total susceptibility $M/H$ and differential susceptibility $\partial M/\partial H$ for $T \geqslant T_f$ under quasisteady conditions, we measured the field dependence of $\partial M/\partial H$ with the aid of an R3003 comparator and a PDP-4 plotter. The total susceptibility was found by numerical integration on a computer. We also measured the temperature derivative $\partial M/\partial T$ of the magnetization with the aid of an electronic circuit capable of maintaining a constant heating rate to $\pm$ 1%. The emf, which is

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proportional to the derivative $\partial M/\partial T$, was recorded continuously with the aid of an R3003 comparator and PDP-4 plotter. The temperature markers were registered by a digital printer. The temperature was measured by a carbon (TSU-2) or semiconductor (KG) thermometer. The temperature of the sample was held constant to $\pm 0.01$ K over the course of the measurements by means of a VRT-2 regulator. All the samples had a face-centered cubic crystal structure.

The alloys $Fe_{65}Ni_{20}Cr_{15}$ and $Fe_{70}Ni_{20}Cr_{10}$ were smelted at the Central Scientific Research Institute of Ferrous Metallury. Neutron-diffraction studies of samples of these compositions, smelted in the same furnace and subjected to the same heat treatment, showed that the alloys contained no structural irregularities larger than two or three interatomic distances.[31] A study on a Cameca microanalyzer showed that there were no macroscopic inhomogeneities of the composition. The $Fe_{45}Ni_{35}Mn_{20}$ alloy was smelted at the Institute of Metal Physics in Sverdlovsk. No structural irregularities were observed by neutron diffraction.[17] However, a magnetic thermal analysis revealed the presence of microscopic amounts (of the order of 0.1–0.01%) of a second magnetic phase in this alloy. This phase is ferromagnetic, and the value of the Curie temperature suggested the presence of regions with incomplete atomic ordering of the $Ni_3Mn$ type. Quenching in oil from a temperature of 1100–1200 °C minimized the influence of the second magnetic phase on the properties of $Fe_{45}Ni_{35}Mn_{20}$ (in many cases the contribution of the second phase was at the level of the experimental error). The samples were in the form of cylinders with a length-to-diameter ratio $l/d \geqslant 20$.

## 3. PREDICTIONS OF THE SPIN-GLASS THEORY

Upon transition to a nonergrodic state, the theory of a spin glass with an infinite-range interaction[7,8,10,11] predicts that the equilibrium susceptibility will be independent of temperature:

$$\chi_e = M_{FC}/H = \text{const}, \quad T < T_f, \tag{1}$$

where $M_{FC}$ is the magnetization measured after cooling in a magnetic field $H$ below the freezing point $T_f$. The equation describing the line of this transition on the $H$-$T$ plane was obtained by de Almeida and Thouless.[12] In the limit of small magnetic fields

$$T_f(0) - T_f(H) = (H/H_0)^n, \tag{2}$$

where $n = 2/3$ for the Ising model. According to Ref. 28, in a Heisenberg spin glass the transverse components of the spin freeze first (for this process $n = 2$) and then the longitudinal components ($n = 2/3$).

Below the de Almeida–Thouless line $T_f(H)$ (in the nonergodic phase) the field dependence of the equilibrium susceptibility of the spin glass is nonanalytic (see, e.g., Refs. 8, 29):

$$\chi_e = \chi_0 - \chi_1 H^P + O(H^2), \tag{3}$$

where $\chi_1$ is the generalized nonlinear susceptibility, $P = 4/3$ for $T < T_f$ (we note that $P = 1$ for $T = T_f$ and $P = 2$ for $T > T_f$),[29] and $O(H^2)$ denotes terms of higher order.

In the nonergodic phase, according to Refs. 8 and 10, we have for $\varepsilon = (T_f - T)/T_f \ll 1$

$$\Delta\chi = \chi_e - \chi_{ne} = A(H)\varepsilon + B(H)\varepsilon^2, \tag{4}$$

where $\chi_{ne} = M_{ZFC}/H$ is the nonequilibrium susceptibility, $M_{ZFC}$ is the magnetization measured after cooling in the absence of magnetic field, and $A(H)$ and $B(H)$ are coefficients, with $A(H) \to 0$ for $H \to 0$.

The transition to a "spin-glass" state from the high-temperature region, $T \to T_f^+$, was first considered by Suzuki.[27] It turned out that when the Edwards–Anderson parameter $q_{EA}$ is taken into account together with the reduced magnetization $m$ in the Ginzburg–Landau funtional, the anomalous quantities at $T_f$ are not the thermodynamic variables themselves (the magnetization, specific heat, etc.) but rather their derivatives with respect to the magnetic field, i.e., in the expansion

$$m = \chi_0 H + \chi_2 H^3 + \chi_4 H^5 + \chi_6 H^7 + \ldots, \tag{5a}$$

where

$$\chi_0 \sim \varepsilon^{-\gamma}, \quad \chi_2 \sim \varepsilon^{-\gamma_2}. \tag{5b}$$

the exponent $\gamma$ equals $-1$ but $\gamma_2$ equals 1 (in the mean field approximation).

The nonlinear susceptibilities of higher order (e.g., $\chi_4$) also diverge at $T_f$. Comparison of expressions (3) and (5a) show that the generalized nonlinear susceptibility $\chi_1$ should diverge at the phase transition point $T_f$. According to Ref. 27, at a paramagnetic-to-spin-glass transition the behavior of the nonlinear susceptibility should be described by the scaling law

$$(m/H) - \chi_0 = \chi_s = \varepsilon^{-\beta} g(H/\varepsilon^{(\gamma_2+\beta)/2}), \tag{6}$$

where $\beta$ and $\gamma_s \approx \gamma_2$ are critical exponents.

Going beyond mean field theory will give a correction to the values of the critical exponents and the exponents $n$ and $P$ in expressions (2) and (3).[13–15] The theory of a spin glass with a large but finite interaction radius[15] yields a step-like phase transition to a "spin-glass" state. Consequently, according to Ref. 15, the nonlinear susceptibility $\chi_2$ should increase on approach to the transition point in accordance with power law (5b), oscillating between two envelopes ($\chi_2^+$ and $\chi_2^-$), and the scaling-law behavior should not occur.

## 4. EXPERIMENTAL RESULTS AND DISCUSSION

### 4.1. The *H-T* diagram

Figure 1a shows as an example the temperature dependence of the total susceptibility of the alloy $Fe_{45}Ni_{35}Mn_{20}$ in various magnetic fields. The measurements were made on heating from 4.2 K after cooling from $T > T_f$ in a magnetic field $H$ ($\chi_e = M_{FC}/H$) and in the absence of field ($\chi_{ne} = M_{ZFC}/H$). It is seen that at every value of the magnetic field the peaks of $\chi_e$ and $\chi_{ne}$ occur at the same temperature $T_f(H)$, and the difference $\Delta\chi = \chi_e - \chi_{ne}$ becomes nonzero at another value of the temperature:

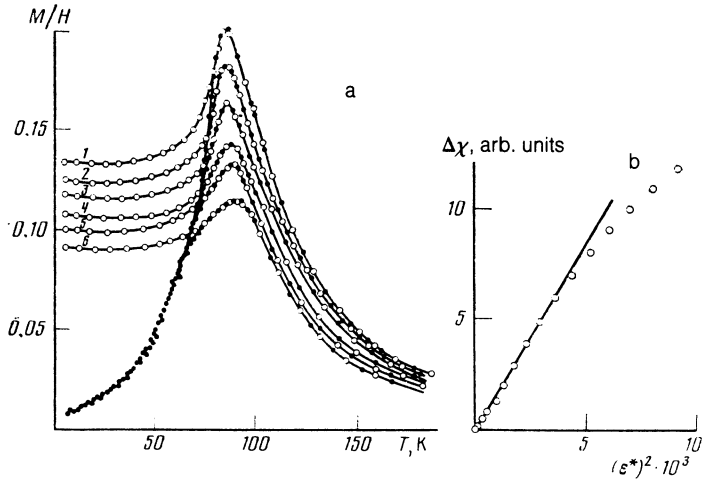


FIG. 1. a) Temperature dependence of the equilibrium susceptibility $\chi_e = M_{FC}/H$(○) and nonequilibrium susceptibility $\chi_{ne} = M_{ZFC}/H$(●) of the alloy $Fe_{45}Ni_{35}Mn_{20}$ in magnetic fields $H$ [in kA/m]: 1) 0.4, 2) 0.8, 3) 1.6, 4) 4, 5) 8, and 6) 16. The equilibrium susceptibility was measured after cooling to 4.2 K in the same magnetic field $H$; b) the difference between the equilibrium and nonequilibrium susceptibilities versus the square of the reduced temperature $\varepsilon^* = (T - T_R(H))/T_R(H)$ for $Fe_{70}Ni_{20}Cr_{10}$ in a magnetic field of 0.8 kA/m.

$T_m(H) \leqslant T_f(H)$. In a certain interval of temperatures below $T_m(H)$ the equilibrium susceptibility $\chi_e$ depends relatively weakly on temperature, in qualitative agreement with the prediction of the analytical theory of the spin glass.[7-12] Below the line $T_m(H)$ the magnetic behavior becomes irreversible: there is a rapid increase in the remanent magnetization, coercive force, and magnetic aftereffect (see Ref. 22). The appearance of magnetic irreversibility is customarily attributed to a transition to a quasinonergodic state.[1,2] The problem is to determine precisely the position of the $T_m(H)$ line on the $H$-$T$ diagram. The relative error in finding the difference $\Delta\chi = \chi_e - \chi_{ne}$ at small $\Delta\chi$ becomes extremely large, and the values of the temperature $T_m(H)$ can be determined only to very low accuracy from an analysis of the temperature dependence of $\chi_e$ and $\chi_{ne}$. Different methods of finding the $T_m(H)$ line are described in the literature. The most accurate way of finding this line is to analyze the temperature dependence of the derivative $\partial M/\partial T$ (Ref. 32) and the imaginary susceptibility $\chi''$ (see, e.g., Ref. 34). Figure 2 shows the temperature dependence of the derivative $\partial M_{FC}/\partial T$ and $\partial M_{ZFC}/\partial T$ for $Fe_{70}Ni_{20}Cr_{10}$ (in the first case the temperature derivative of the magnetization was measured after cooling in a magnetic field; in the second case, after cooling in the absence of field). Direct measurement of the derivatives $\partial M_{FC}/\partial T$ and $\partial M_{ZFC}/\partial T$ permitted very accurate (better than $\pm 0.5\%$) determination of the values of certain characteristic temperatures. The shape of the temperature dependence of $\partial M_{FC}/\partial T$ and $\partial M_{ZFC}/\partial T$ in Fe–Ni–Cr spin glasses is approximately the same as in the spin glasses Ag–Mn (Ref. 32). There is a characteristic temperature $T_R$ at which the temperature dependence of $\partial M_{FC}/\partial T$ intersects the line $0A$, which is the continuation of the rapidly changing part of the curve of the derivative $\partial M_{ZFC}/\partial T$ (see Fig. 2).

In a certain interval of reduced temperatures $\varepsilon^* = (T - T_R)/T_R$ the difference $\Delta\chi_e = \chi_e - \chi_{ne}$ is described well by the expression (4) from the analytical theory (Refs. 8, 10), which implies that $\Delta\chi \sim \varepsilon^2$ in the limit of small $H$ (see Fig. 1b). An analogous result is obtained[33] for the amorphous spin glasses $(Fe_{0.64}Mn_{0.36})_{75}P_{16}B_6Al_3$. The temperature curves of $\partial M_{FC}/\partial T$ and $\partial M_{ZFC}/\partial T$ ultimately converge at a temperature $T_p$, which corresponds to an inflection point. The temperature $T_f$ of the susceptibility peak corresponds to $\partial M/\partial T = 0$. Finally, for $T > T_f$ there is a

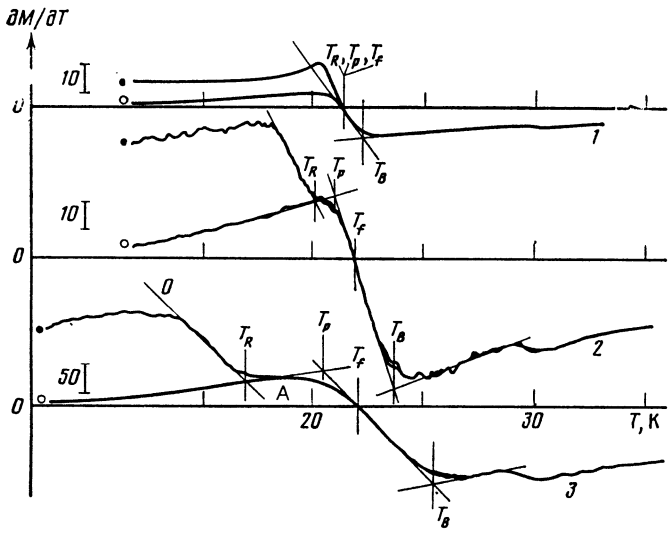


FIG. 2. Temperature dependence of the derivative of the magnetization with respect to temperature for the alloy $Fe_{70}Ni_{20}Cr_{10}$. The measurements were made on heating from 4.2 K at a constant rate in magnetic fields [in kA/m]: 1) 0.8, 2) 4, 3) 15.2. The open points correspond to a preliminary cooling in a magnetic field of the same strength ($\partial M_{FC}/\partial T$), the filled-in points to a preliminary cooling in the absence of magnetic field ($\partial M_{ZFC}/\partial T$).

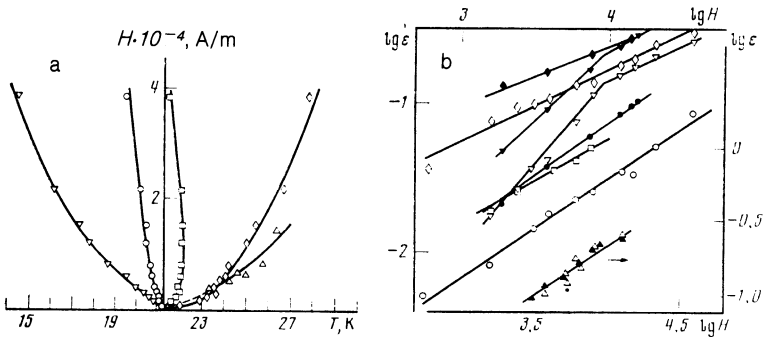


FIG. 3. a) The $H$-$T$ diagram of the alloy $Fe_{70}Ni_{20}Cr_{10}$, showing the values of the characteristic temperatures $T_R$ (▽), $T_p$ (○), $T_f$ (□), and $T_B$ (◇). The $T_k$ ($H$) line (△) indicates the values of the magnetic field $H_{cr}$ above which (at a fixed value of the reduced temperature) the first four terms of the expansion (5a) are no longer sufficient for describing the field dependence of the total susceptibility; b) the field dependence of the temperatures $T_R$ (▽,▼), $T_p$ (○,●), $T_f$ (□), $T_B$ (◇,◆), and $T_k$ (△,▲) (in logarithmic scale). The filled-in points correspond to the alloy $Fe_{65}Ni_{20}Cr_{15}$, the open points to $Fe_{70}Ni_{20}Cr_{10}$.

characteristic temperature $T_B$, which corresponds to an inflection point of the curve $\partial M(T)/\partial T$ in the region of negative values (see Fig. 2).

The $H$-$T$ diagram in Fig 3a shows the values of $T_R$, $T_p$, $T_f$, and $T_B$ for $Fe_{70}Ni_{20}Cr_{10}$. We have to decide which of the characteristic temperatures ($T_p$, where a difference $\Delta\chi$ arises between the equilibrium and nonequilibrium susceptibilities, or $T_R$, where $\Delta\chi$ begins to increase sharply; see Fig. 2) is most suitable for comparison with the theory. Chamberlin *et al.*[32] chose the temperature $T_p$, because (unlike $T_R$) its values for dilute Ag–Mn alloys scale in the coordinates $\mu_B gH/k_B T_f(0) - T/T_f(0)$. This principle cannot be applied to concentrate Fe–Ni–Cr spin glasses, for which (unlike dilute alloys) the freezing temperature $T_f(0)$ divided by the magnetic field $H_0$ from expression (2) is not proportional to the fraction of magnetic atoms (see Tables I and II). The field dependence of the temperature $T_p$ in each of the two Fe–Ni–Cr spin glasses studied is described well by expression (2) with the same values of the field $H_0^p$ and exponent $n_p$ over the entire range of magnetic fields studied (0–40 kA/m) (see Fig. 3b). The value of the exponent $n_p$ (see Table I) is very close to the value predicted by the mean field theory of the spin glass (for the Ising model, $n = 2/3$).[8,12] At the same time, the values of the characteristic temperature $T_R$ in the indicated magnetic-field interval cannot be described by a power law of the form (2) with a single value of the exponent $n_R$. In particular, for $Fe_{70}Ni_{20}Cr_{10}$ in weak magnetic fields the values of $n_R$ for the $T_R$ ($H$) line are close to unity for $H \sim 10^4$ A/m: $n_R \approx 0.6$ (see Fig. 3b). Thus the field dependence of the temperature $T_p$ has the better agreement with mean field theory (2). However, expression (4) from the theory of Refs. 8–12 does hold in the neighborhood of the other characteristic temperature $T_R$ ($T \lesssim T_R$, see Fig. 1b).

The values of the normalizing magnetic field $H_0^p$ in Fe–Ni–Cr spin glasses is several times smaller than the estimate found from the theoretical expression[28]:

$$H_0 = \left[\frac{(m+1)(m+2)}{8}\right]^{-1/2} \frac{k_B T_f(0)}{g\mu_B S}, \qquad (7)$$

where $S$ is the magnitude of the spin, $g$ is the Landé factor, $\mu_B$ is the Bohr magneton, $k_B$ is Boltzmann's constant, and $m$ is the number of spin components.

A similar disagreement between the theoretical estimate and experiment arises in RKKY spin glasses.[2,7,34] It is obviously due to cluster effects, which make it necessary to replace the atomic spin $S$ in expression (7) by the cluster spin $S_{eff} > S$. We see from Table I that the values of the normalizing field $H_0^p$ differ strongly in the two Fe–Ni–Cr spin glasses, which have very similar freezing temperatures. This is probably because the concentrated spin glasses have an effective cluster size (and hence $S_{eff}$ and $H_0$) that depends strongly on the proximity to the critical concentration $x_c^f$ from the onset of ferromagnetism. In particular, for $Fe_{58}Ni_{24}Cr_{18}$ a value $H_0^p = 3.2 \cdot 10^3$ A/m has been found[35] (corresponding to $S_{eff} \sim 10^3$–$10^4$).

We did not observe any indication of the presence of a line associated with the freezing of the transverse spin components on the $H$-$T$ diagram of Fe–Ni–Cr spin glasses (the Gabay–Toulouse line[28]). According to Ref. 28, this line should correspond to an exponent $n = 2$ in expression (2).

It is possible that the transverse and longitudinal components of the spins are weakly coupled to each other and that the freezing of the transverse components is not reflected in the measurement of the longitudinal susceptibility. More likely, however, is the situation described in Ref. 30. According to Ref. 30, the Gabay–Toulouse line arises for weak anisotropy:

$$d < h^{2/3}, \qquad (8)$$

where $d = D/k_B T$, $h = \mu H/k_B T$, $\mu = S\mu_B$, and $D$ is the anisotropy constant. Contrarily, for strong anisotropy ($d \gg h^{2/3}$) the $H$-$T$ diagram should exhibit only the de Almeida–Thouless line.[30] The case of strong anisotropy can obtain in Fe–Ni–Cr spin glasses during measurements in weak

TABLE I. Values of the exponent and normalizing magnetic field in power law (2) for various lines on the $H$-$T$ diagram of several spin glasses.

| Composition [Ref.] | $n_p$ | $H_0^p$, $10^6$ A/m | $n_f$ | $H_0^f$, $10^6$ A/m | $n_B$ | $H_0^B$, $10^6$ A/m | $n_k$ | $H_0^k$, $10^6$ A/m |
|---|---|---|---|---|---|---|---|---|
| $Fe_{70}Ni_{20}Cr_{10}$ | 0.67±0.05 | 1.78±0.15 | 0.60±0.05 | 1.12 | 0.35±0.05 | 0.34±0.04 | 0.66±0.05 | 0.10±0.01 |
| $Fe_{65}Ni_{20}Cr_{15}$ | 0.67±0.03 | 1.05±0.05 | – | – | 0.35±0.05 | 0.25±0.03 | 0.66±0.05 | 0.10±0.01 |
| Ag – 2.6% Mn [32] | 2/3 | – | – | – | 0.51±0.10 | 2.48 | – | – |
| Ag – 4% Mn [32] | 2/3 | – | – | – | 0.56±0.10 | 2.02 | – | – |
| $Gd_{37}Al_{63}$ [14] | 0.57 | 0.36 | – | – | – | – | 0.60 | 0.99 |

TABLE II. Values of the temperature $T_f$ of the magnetic transition, the critical exponents, and certain other parameters for various spin glasses.

| Composition [Ref.] | $T_f$, K | $\gamma_1$ | $\gamma_2$ | $\gamma_4$ | $\gamma_6$ | $\beta$ | $P(T_f)$ | $\varepsilon=(T_f-T)/T_f$ |
|---|---|---|---|---|---|---|---|---|
| $Fe_{70}Ni_{20}Cr_{10}$ | 20.6±0.1 | 2.75±0.07 | 2.6±0.1 | 5.3±0.2 | 8.3±0.5 | 0.55±0.05 | 0.50±0.05 | 0.05–0.3 |
| $Fe_{70}Ni_{20}Cr_{10}$ | 21.2±0.1 | 2.65±0.10 | 2.4±0.1 | 5.1±0.2 | 7.8±0.5 | 0.60±0.05 | 0.50±0.05 | 0.05–0.4 |
| $Fe_{55}Ni_{25}Mn_{20}$ | 89.0±0.5 | 2.2±0.2 | – | – | – | – | 0.60±0.05 | – |
| Mn aluminosilicate [37] | 2.95 | – | 3.1±0.1 | 7.6±0.3 | 12.1±0.5 | 1.4±0.1 | 0.35 | 0–0.4 |
| Cu – 4.6% Mn [36] | 26.2 | 3.1±0.4 | – | – | – | 1 | 0.7±0.1 | 0.1–2 |
| $Gd_{37}Al_{63}$ [36] | 15.8 | 3.8±0.5 | – | – | – | – | 0.5±0.1 | 0.1–2 |
| Au – 1.5% Fe [40] | 8.9±0.1 | – | 1.1±0.2 | – | – | – | – | 0.01–0.5 |
| Ag – 10.6% Mn [41] | 37.4 | – | 1.5±0.5 | – | – | – | – | 0–0.5 |

magnetic fields because of the extremely large unidirectional anisotropy of these materials.[18]

It should be noted that in Fe–Ni–Cr spin glasses the line $T_f(H)$ of susceptibility maxima and the line $T_m(H)$ along which magnetic irreversibility arises [here, as in Ref. 32, $T_p(H)$ is clearly the better choice for the latter temperature] do not coincide. This behavior is typical in other spin glasses as well.[1,32] In the theory of the Ising spin glass[8–12] the lines $T_f(H)$ and $T_m(H)$ are identical, in disagreement with experiment.

Mean field theory[8–12] is also unable to explain the field dependence of the position of the susceptibility peaks in Fe–Ni–Cr spin glasses. On increasing magnetic field $H$, the susceptibility peak is shifted first to higher and then to lower temperatures (see Fig. 3a). The same kind of field dependence $T_f(H)$ has been observed in the spin glasses $Gd_{37}Al_{63}$ and Cu–Mn (Ref. 14) and in the alloy $Fe_{45}Ni_{35}Mn_{20}$ (Ref. 19). In Ag–Mn spin glasses, however, $T_f$ decreases monotonically on increasing magnetic field.[32] It was shown in Ref. 14 that the nonmonotonicity of $T_f(H)$ can be explained in the framework of the mean field approximation by assuming a definite form of the scaling function $g(x)$.

The $T_B(H)$ line in Fe–Ni–Cr spin glasses is described well by a power law of the form (2) with values of the exponent $n_B$ that are close to those found for Ag–Mn alloys[32] (see Fig. 3b and Table I). In Ref. 32 it is assumed that $T_B$ is not a well-defined temperature but corresponds to the middle of the transition region between the region of critical behavior for $T=T_f(0)$ and the temperature region in which the Curie–Weiss law holds. Our findings are consistent with this view. The $H$-$T$ diagram (Fig. 3a) exhibits another crossover line $T_k(H)$ which was found from analysis of the nonlinear susceptibility (see Subsec. 4.2 below). As will be shown in Subsec. 4.2, the line $T_k(H)$ has a clearer physical meaning. The susceptibility in Fe–Ni–Cr spin glasses begins to obey the Curie–Weiss law at temperatures considerably higher than $T_B$ and $T_k$.

The values of the exponents $n_p$, $n_f$, and $n_B$ ($n_k$) are connected by certain critical-exponent relations.[1,13,14,32] This topic will also be discussed in the next subsection.

### 4.2. Nonlinear susceptibility

According to theory,[27] in the vicinity of the phase transition temperature one should analyze the susceptibility corresponding to the static limit. The separation of the nonlinear contributions to the static susceptibility with the aid of both (3) and (5a) has been described in the literature.[36,37] Our study and the results of Ref. 37 indicate that the use of expansion (5a) yields more-reliable results (but only if at least four terms are taken into account).

The field dependence of the susceptibility at different temperatures was analyzed on a computer by the least-squares method. According to Ref. 37, for each isotherm there is a critical magnetic field $H_{cr}$ above which there is a rapid increase in the standard deviation when the experimental $M(H)/H$ curves are described by the first four terms of expansion (5a). The values of $H_{cr}$ for different values of the reduced temperature are given in Fig. 3a [the line $T_k(H)$]. The temperature dependence of the coefficients $\chi_2$, $\chi_4$, and $\chi_6$ for $Fe_{70}Ni_{20}Cr_{10}$ is shown in Fig. 4a. We see from Fig. 4b that this dependence conforms well to a power law

$$\chi_n = a_n \varepsilon^{-\gamma_n}, \tag{9}$$

[$\varepsilon=(T-T_f)/T_f$, $a_n$ is a coefficient] with the values of the critical exponents $\gamma_n$ given in Table II.

Scaling law (6) gives a rather good description of the behavior of the nonlinear susceptibility of Fe–Ni–Cr spin glasses at values of $\varepsilon$ in the interval from 0.01 to 0.3–0.4 (see Fig. 5). The smallest scatter in the points is obtained at the values of the critical exponents $\beta$ and $\gamma_2$ given in Table II. The form of the scaling function $g(x)$ ($x=H^2/\varepsilon^{\gamma_2+\beta}$) is approximately the same for Fe–Ni–Cr spin glasses and manganese aluminosilicate.[37]

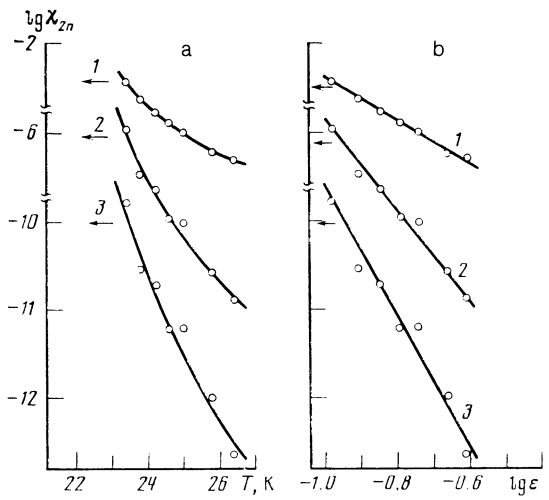


FIG. 4. a) Semilogarithmic plot of the temperature dependence of the coefficients of the nonlinear susceptibility of the alloy $Fe_{70}Ni_{20}Cr_{10}$: 1) $\chi_2$, 2) $\chi_4$, 3) $\chi_6$; b) log–log plot of the coefficients $\chi_2$ (1), $\chi_4$ (2), and $\chi_6$ (3) versus the reduced temperature $\varepsilon=(T-T_f)/T_f$ for the same alloy.

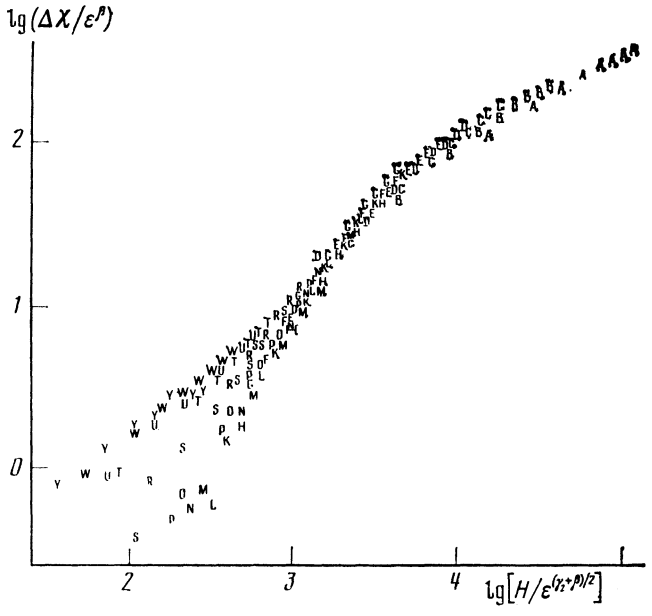


FIG. 5. Plot illustrating the applicability of scaling law (6) for describing the nonlinear susceptibility $\chi_5$ of the alloy $Fe_{65}Ni_{20}Cr_{15}$. The measurement temperatures [in K] are: A) 21.0; B) 21.4; C) 21.8; D) 22.2; E) 22.6; F) 23.0; G) 23.4; H) 23.6; K) 24.0; L) 24.4; M) 24.8; N) 25.2; O) 25.6; P) 26.0; R) 27.0; S) 28.0; T) 29.0; U) 30.0; W) 32.0; Y) 35.0.

For $Fe_{45}Ni_{35}Mn_{20}$, owing to the presence of microscopic quantities of a second (ferromagnetic) phase, there was a large error in the analysis of the field dependence of the susceptibility above $T_f$ with the aid of expansion (5a). We shall therefore omit the results of this analysis. However, an analysis could be done using expression (3), since $\lg(\chi_e - \chi_0)$ as a function of $\log H$ is practically a straight line.

Figure 6 shows the temperature dependence of the generalized nonlinear susceptibility $\chi_1$ and the exponent $P$ for the alloy $Fe_{45}Ni_{35}Mn_{20}$ (similar curves are found for $Fe_{65}Ni_{20}Cr_{15}$ and $Fe_{70}Ni_0Cr_{10}$). The values of the exponent $P$ in Fe–Ni–Cr(Mn) spin glasses and the spin glasses Cu–Mn, Gd–Al, and manganese aluminosilicate[36,37] are quite similar (see Table II). The predictions of the analytical theory of the spin glass[7-11] are confimed in the sense that the field dependence of the equilibrium susceptibility of the spin glass is nonanalytic. The temperature dependence of the generalized nonlinear susceptibility $\chi_1$ above $T_f$ can be described by power law (9) with the values of the critical exponent $\gamma_1$ given in Table II.

On the whole, the results on the nonlinear susceptibility of Fe–Ni–Cr and (somewhat less reliably) Fe–Ni–Mn spin glasses indicate the existence of a phase transition to a "spin-glass" state. The model of Ref. 38 which describes the blocking of the magnetic moments of the clusters in the anisotropy field cannot explain the presence of a well-defined transition point (see Fig. 6, for example) and the existence of a scaling law for the nonlinear susceptibility (see Fig. 5). However, the following questions arise.

a) Why do the critical exponents have different values in different spin glasses (see Table II)?

b) Why does scaling law (6) hold over such an extremely wide temperature interval?

As to the wide interval of $\varepsilon$ over which the scaling law holds, a similar situation also arises in the neighborhood of the Curie point $T_c$ in inhomogeneous ferromagnets (for example, in the alloy $Eu_{0.7}Sr_{0.3}S$ in the interval $\varepsilon \lesssim 0.5$–$0.6$).[39] However, the large temperature region in which the scaling law holds does not preclude true critical behavior in the phase transition point itself.[39]

As to the difference in the values of the critical exponents, one can suggest the following possibilities:

a) The difference might be due to the use of different methods of measurement and data processing. In particular, it is possible that the reason for the low value of the exponent $\gamma_2$ in the spin glass Au–1.5% Fe (Ref. 40) is that it was obtained from analysis of dynamic susceptibility measurements (moreover, in this case it is harder to separate contributions to the nonlinear susceptibility);

b) It is possible that there are several universality classes for spin glasses depending on the type of anisotropy and its value, the characteristic interaction radius, etc. In this case the Fe–Ni–Cr and Fe–Ni–Mn spin glasses would clearly belong to the same class as Cu–Mn alloys (with $\gamma_2 \sim 3, \beta < 1$). Then the other class would include the spin glasses Au–Fe (Ref. 40) [and possibly Ag–Mn (Ref. 41)], with $\gamma_2 \sim 1$.

To distinguish these two possibilities it is necessary to analyze the nonlinear susceptibility of different spin glasses by the same method [best done using expression (5a) with allowance for at least four terms]. Such an analysis has so far been carried out only for manganese aluminosilicate[37] and Fe–Ni–Cr spin glasses (present study);

c) The difference in the values of the critical exponent $\gamma_2$ could be due to the variance of the exchange interaction and its influence on the static (noncritical) correlations of the spin orientations at finite values of the reduced temperature $\varepsilon$ (Ref. 42). In this case the effective critical exponent $\gamma_2^* = (d \ln \chi_2^{-1})/(d \ln \varepsilon)$ could take on a continuous set of values. However, in the theory of Ref. 42 the exponent $\gamma_2^*$ depends on $\varepsilon$, whereas no such dependence is seen in experiment (see Fig. 4b).

The theoretical conclusions of Ref. 15 do not agree well enough with the results on the nonlinear susceptibility of Fe–Ni–Cr spin glasses. No noticeable oscillations are seen on the

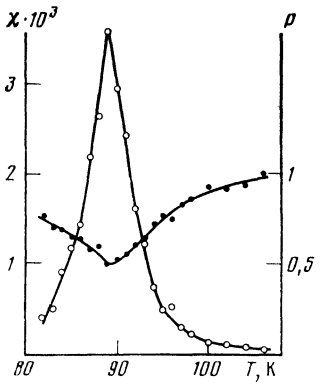


FIG. 6. Temperature dependence of the generalized nonlinear susceptibility $\chi_1$ (○) and of the exponent $P$ (●) from expression (3) for the alloy $Fe_{45}Ni_{35}Mn_{20}$.

temperature dependence of the nonlinear susceptibility $\chi_2$ (see Fig. 4), and, in disagreement with Ref. 15, a scaling law holds (see Fig. 5). It is possible that the effective range of the exchange interaction in Fe–Ni–Cr alloys is not large enough that the model of Ref. 15 can be used to describe its properties.

Finally, we turn to a comparison of the exponents $n_p$, $n_f$, $n_B$, and $n_k$ (corresponding to different lines on the $H$-$T$ diagram; see Fig. 3a and Table I) and critical exponents (see Table II). According to Refs. 13 and 14,

$$n_p=2/\varphi_-, \tag{10}$$

$$n_f=2/(\gamma_s+1) \tag{11}$$

(in the magnetic-field region in which the freezing temperature $T_f$ increases with increasing magnetic field),

$$n_k=2/(\gamma_s+\beta), \tag{12}$$

with $\gamma_s+\beta=\varphi_+$; $\varphi_+$ and $\varphi_-$ are the values of the crossover exponents above and below $T_f$, respectively, and $\gamma_s$ is the critical exponent of the nonlinear susceptibility.

Generally speaking, it is not clear[13,14] which nonlinear-susceptibility exponent should be used in expressions (11) and (12): $\gamma_1$ or $\gamma_2$. In Fe–Ni–Cr spin glasses relations (11) and (12) are better satisfied if we take $\gamma_s=\gamma_2$. The crossover line $T_k(H)$, which separates the region of "low" magnetic fields from the region of "high" magnetic fields (fields which suppress the critical fluctuations), has approximately the same value of the critical exponent ($\varphi_+$) as does the line $T_p(H)$ on which the irreversibility appears ($\varphi_-$). This means that the lines $T_k(H)$ and $T_p(H)$ and also $T_f(H)$ can be described consistently in a non-mean-field theory of the spin glass.[14] Mean field theory[7–12,27] gives $\varphi_-=3$ but $\varphi_+=2$, whereas experimentally $\varphi_+\geqslant 3$ (see Table II). We note that for the line $T_B(H)$ a relation of form (12) (with values of the exponent $\gamma_s$ equal to $\gamma_1$ or $\gamma_2$) is satisfied considerably less well than for the line $T_k(H)$.

## 5. CONCLUSION

Mean field theory[7–12] gives a qualitatively correct description of the magnetic properties of the investigated alloys of the systems Fe–Ni–Cr and Fe–Ni–Mn, which lack long-range magnetic order. Below the point $T_f$ of the transition to the "spin-glass" state, the equilibrium susceptibility $\chi_e$ is weakly temperature-dependent. The line along which magnetic irreversibility arises is described by (2) with an exponent $n$ that is close to the theoretical values for the Ising model (2/3). The difference between the equilibrium and nonequilibrium susceptibilities $\Delta\chi=\chi_e-\chi_{ne}$ is described by expression (4) in a certain temperature interval, while the field dependence of $\chi_e$ is nonanalytic.

One wonders why a theory developed for a magnetic insultor[7–12] gives a satisfactory description of certain properties of metallic spin glasses. This question pertains not only to the 3$d$-metal alloys studied in the present paper (such as Fe–Ni–Cr and Fe–Ni–Mn) but also to dilute metallic alloys with the RKKY interaction. In alloys of the Fe–Ni–Cr(Mn) type the exchange interaction is long-ranged and has a rather complex spatial structure. However, this interaction (like the RKKY interaction in dilute alloys) is characterized quite well by a model[5] with a nearest-neighbor interaction of alternating sign (see Ref. 43 for details).

The experiment also revealed a number of discrepancies with the predictions of mean field theory[7–12]:

a) The theory[7–12] cannot give a consistent explanation of the form of the $H$-$T$ diagram of Fe–Ni–Cr spin glasses. In diagreement with the predictions of the theory,[7,12] the line on which the magnetic irreversibility arises in the experiment does not coincide with the line $T_f(H)$ of susceptibility maxima. In Fe–Ni–Cr spin glasses the field dependence of $T_f$ is more complex than predicted by the theory[7–12] (according to which $T_f$ always decreases with increasing magnetic field).

b) The values of the exponent $P$ in expression (3) and of the critical exponents, $\beta$, $\gamma_2$, and $\varphi_+$ in Fe–Ni–Cr and Fe–Ni–Mn spin glasses are substantially different from the mean field values (see Table II).

By going beyond the mean field approximation[13–15] (i.e., taking fluctuations into account), one can explain the form of the $H$-$T$ diagram of Fe–Ni–Cr spin glasses and also (in principle) the difference of the critical exponents from the mean field values. However, the non-mean-field theory of the spin glass requires further development. For example, the results of Refs. 13 and 14 depend substantially on the choice of scaling function, which was found by the principle of optimizing the agreement with experiment.

The results on the nonlinear susceptibility that the Fe–Ni–Cr and Fe–Ni–Mn alloys have a phase transition to a "spin-glass" state. The question of universality of the critical behavior of spin glasses will require studying the nonlinear susceptibility of different spin glasses by the same method.

Spin glasses of the systems Fe–Ni–Cr and Fe–Ni–Mn are concentrated spin glasses and cannot be expected to exhibit appreciable "cluster" effects. Our studying (and studies of other concentrated spin glasses, e.g., Refs. 36 and 37) show, however, that correlations in the orientation of neighboring spins will necessitate only the replacement of the atomic magnetic moment $S\mu_B$ in the formulas [e.g., in (7)] by a certain effective magnetic moment $S_{\text{eff}}\mu_B$. Such a correction is also necessary in RKKY spin glasses at magnetic atom concentrations $\sim 1\%$ (Refs. 32, 33). The nature (at least) of the magnetic transition at $T_f$ will clearly not be affected much by "cluster" effects of this kind. The size of the static spin clusters is probably substantially smaller than the correlation length of the critical fluctuations for the phase transition at $T_f$ (see Ref. 42 for details).

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[1]K. H. Fischer, Phys. Status Solidi B **116**, 357 (1983).
[2]I. Ya. Korenblit and E. F. Shender, Izv. Vyssh. Uchebn. Zavod. Fiz., No. 10, 23 (1984).
[3]D. Sherrington and S. Kirkpatrick, Phys. Rev. Lett. **35**, 1792 (1975).
[4]A. J. Bray and M. A. Moore, J. Phys. C **13**, L469 (1980).
[5]S. F. Edwards and P. W. Anderson, J. Phys. F **5**, 965 (1975).
[6]A. J. Bray and M. A. Moore, J. Phys. C **14**, 1313 (1981).
[7]G. Parisi, Phys. Rev. Lett. **43**, 1754 (1979).
[8]G. Parisi, J. Phys. A **13**, 1887 (1980).
[9]G. Parisi, Phys. Rev. Lett. **50**, 1946 (1983).
[10]H. Sompolinsky, Phys. Rev. Lett. **47**, 935 (1981).

[11]H. Sompolinsky and A. Zippelius, Phys. Rev. B **25**, 6860 (1982).
[12]J. R. L. de Almeida and D. J. Thouless, J. Phys. A **11**, 983 (1978).
[13]H. Bouchiat, J. Phys. C **16**, L145 (1983); **30**, 3963 (1984).
[14]A. P. Malozemoff, S. E. Barnes, and B. Barbara, Phys. Rev. Lett. **51**, 1704 (1983); B. Barbara, A. P. Malozemoff, and S. E. Barnes, J. Appl. Phys. **55**, 1655 (1984).
[15]L. V. Ioffe and M. V. Feĭgel'man, Zh. Eksp. Teor. Fiz. **89**, 654 (1985) [Sov. Phys. JETP **62**, 376 (1985)].
[16]A. Z. Men'shikov and A. E. Teplykh, Fiz. Met. Metalloved. **44**, 1215 (1977).
[17]A. Z. Menshikov, P. Burlet, A. Chemberod, and J. L. Tholence, Solid State Commun. **39**, 1093 (1971).
[18]A. V. Deryabin, Yu. A. Chirkov, and A. V. T'kov, Zh. Eksp. Teor. Fiz. **86**, 609 (1984) [Sov. Phys. JETP **59**, 355 (1984)].
[19]A. V. Deryabin and A. V. T'kov, Zh. Eksp. Teor. Fiz. **88**, 237 (1985) [Sov. Phys. JETP **61**, 138 (1985)].
[20]A. V. Deryabin, A. V. T'kov, B. N. Shvetsov, A. P. Larionov, and I. V. Zakharov, Fiz. Tverd. Tela (Leningrad) **27**, 834 (1985) [Sov. Phys. Solid State **27**, 511 (1985)].
[21]A. V. Deryabin and I. G. Pislar', Fiz. Tverd. Tela (Leningrad) **20**, 3456 (1978) [Sov. Phys. Solid State **20**, 1996 (1978)].
[22]A. V. Deryabin, A. Z. Men'shikov, and Yu. A. Chirkov, Fiz. Met. Metalloved. **57**, 897 (1984).
[23]A. V. Deryabin, V. I. Rimlyand, and A. P. Larionov, Zh. Eksp. Teor. Fiz. **84**, 2228 (1983) [Sov. Phys. JETP **57**, 1298 (1983)].
[24]A. V. Deryabin and V. E. Rode, Fiz. Nizk. Temp. **2**, 1450 (1976) [Sov. J. Low Temp. Phys. **2**, 710 (1976)].
[25]A. Z. Menshikov, N. N. Kuzmin, Yu. A. Dorofeev, V. E. Kazantsev, and S. K. Sidorov, J. Magn. Magn. Mater. **20**, 134 (1980).
[26]G. A. Takzeĭ, I. I. Sych, A. Z. Men'shikov, and A. E. Teplykh, Fiz. Met. Metalloved. **52**, 960 (1981).
[27]M. Suzuki, Prog. Theor. Phys. **58**, 1151 (1977).
[28]M. Gabay and G. Toulouse, Phys. Rev. Lett. **47**, 201 (1981).
[29]G. Toulouse and M. Gabay, J. Physique Lett. **42**, L103 (1981).
[30]G. Kotliar and H. Sompolinsky, Phys. Rev. Lett. **53**, 1751 (1984).
[31]V. I. Goman'kov, Tonkaya Kristallicheskaya i Atomnaya Magnitnaya Struktura Splavaov na Osnove Sisteme Zhelezo–Nikel' [Crystalline and Atomic Magnetic Fine Structure of Iron–Nickel Alloys], Author's Abstract of Doctoral Dissertation, Moscow (1980).
[32]R. V. Chamberlin, M. Hardiman, L. A. Turkevich, and R. Orbach, Phys. Rev. B **25**, 6720 (1982).
[33]Y. Yeshurun, L. J. P. Ketelsen, and M. V. Salamon, Phys. Rev. **26**, 1491 (1982).
[34]M. B. Salamon and J. L. Tholence, J. Magn. Magn. Mater. **31–34**, 1375 (1983).
[35]G. A. Takzeĭ, A. M. Kostyshin, and Yu. P. Grebenyuk, Fiz. Tverd. Tela (Leningrad) **26**, 2722 (1984) [Sov. Phys. Solid State **26**, 1648 (1984)].
[36]B. Barbara, A. P. Malozemoff, and Y. Imry, Phys. Rev. Lett. **47**, 1852 (1981).
[37]P. Beauvillain, C. Chappert, and J. P. Renard, J. Physique Lett. **45**, L665 (1984); P. Beauvillain, C. Dupas, J. P. Renard, and P. Veillet, Phys. Rev. **29**, 4086 (1984).
[38]E. P. Wohlfarth, Physica B **86–88**, 852 (1977).
[39]K. Siratory, K. Kohn, H. Suwa, E. Kita, S. Tamura, F. Sakai, and T. Tasaki, J. Phys. Soc. Jpn. **51**, 2746 (1982).
[40]S. Chikazawa, S. Taniguchi, H. Matsuyama, and Y. Miyako, J. Magn. Magn. Mater. **31–34**, 1355 (1983).
[41]P. Monod and H. Bouchiat, J. Physique Lett. **43**, L45 (1982).
[42]M. Fähnle, G. Herzer, H. Kronmüller, R. Meyer, M. Saile, and T. Egami, J. Magn. Magn. Mater. **38**, 240 (1983).
[43]M. V. Medvedev, Izv. Vyssh. Uchebn. Zavod. Fiz., No. 10, 3 (1984).

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