Double paramagnetism-ferromagnetism-spin glass temperature transition in alloys of ferromagnetic 3*d* metals and antiferromagnets

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Anomalously high values of the effective critical exponent γ_0^* of the susceptibility were observed for alloys of the $Fe_{65}Ni_{35-x}Cr_x$ system with $x \sim x_c^f$ at finite values of the reduced temperature $\varepsilon = (T - T_C)/T_C$ (T_C is the Curie temperature). The absence of this effect in $Ni_{100-x}Cr_x$ alloys is probably due to the fact that the latter alloys have no alternating-sign exchange interaction. It is shown that both stages of the temperature paramagnetism-ferromagnetism-spin glass transformation in alloys of the $Fe_{65}Ni_{35-x}Cr_x$ system are described by a scaling law.

I. INTRODUCTION

The possibility of a double paramagnetism-ferromagnetism-spin glass (PM-FM-SG) transition was first predicted by Sherrington and Kirkpatrick[1] for the Ising model with alternating-sign infinite-radius interaction. Within the framework of the phenomenological theory[2] the critical exponents for the PM-FM and FM-SG transition should be the same. The solution of Sherringon and Kirkpatrick, however, is unstable below the spin-glass "freezing" point. In a theory that takes into account the absence of replica symmetry (the Parisi scheme), the appearance of spin glass on cooling does not destroy the long-range ferromagnetic order (see, e.g., Ref. 3). In particular, for Heisenberg spins there are obtained within the framework of this theory an FM-asperomagnetism (ASM)-SG transition[4] (see also Ref. 5). It must be emphasized that the theory was developed only for a system with an infinite interaction radius, so that it is difficult to compare its conclusions with the properties of real alloys. The results of experimental studies of the PM-FM-SG(ASM) temperature transition are contradictory.[3,6–14]. This can be due both to difficulties entailed in the methods[3,6,12] and to the differences in the alloy compositions.

The present paper is devoted to the study of magnetization and the dynamic magnetic susceptibility of a number of quasibaric alloys, viz $Fe_{65}Ni_{35-x}Cr_x$ ($x = 5,10$), $Cr_{10}Fe_xNi_{90-x}$ ($x = 67$), $Mn_{20}Fe_xNi_{80-x}$ ($x = 40,43$). According to the form of the magnetic phase diagrams of the indicated alloys, [16–18] a double PM-FM-SG (ASM) temperature transition can occur in them. We have investigated alloys of the system $Ni_{100-x}Cr_x$ ($x = 5,8$). According to Ref. 19, a concentrational magnetic transition from ferromagnetism to Pauli paramagnetism takes place in the $Ni_{100-x}Cr_x$ system (at $x = x_c^f \approx 10$). There were grounds therefore to suggest that no spin glass is produced in alloys of the $Ni_{100-x}Cr_x$ system with $x \sim x_c^f$. All the investigated alloys had fcc crystal structure at temperature down to the boiling point of liquid helium (4.2 K). We investigated the behavior of the magnetization and of the dynamic susceptibility in the vicinity of the freezing point T_{fg} and of the Curie point T_C.

2. EXPERIMENTAL PROCEDURE

The magnetization was measured by an induction method. The change of the magnetic flux due to turning-on a current in a copper-wire solenoid was measured with an F190 microwebermeter and a PDP-4 automatic plotting potentiometer. The magnetization sensitivity of the apparatus was $1 \cdot 10^{-2}$ A/m. In the magnetization measurements we used cylindrical samples 100 mm long and 5 mm in diameter. The solenoid was capable of producing static magnetic fields up to $3.2 \cdot 10^5$ A/m.

The dynamic magnetic susceptibility was investigated with two setups. In one case the contributions, linear and nonlinear in the magnetic field, to the dynamic susceptibility were investigated by a procedure similar to that described in Ref. 20. The measurements were performed at a frequency 62 Hz, the amplitude of the magnetization-reversing field did not exceed 8 A/m. The transverse component of the geomagnetic field was compensated for accurate to 1 A/m. The samples were rings with outside and inside diameters 5 and 3 mm and height 2 mm. In the second setup the reversible magnetic susceptibility was investigated by another procedure, similar to the one described in Ref. 21. The real and imaginary parts of the susceptibility were separated by a synchronous detector based on a circuit given in Ref. 22. The setup was calibrated by a method described in Ref. 23. It is known (see, e.g., Ref. 24) that in ac bridges a change in the signal amplitude is accompanied by a parasitic phase shift $\Delta\varphi$ in the measurement circuit. The phase shift $\Delta\varphi$ was taken into account by us by using a pure-imaginary-susceptibility standard (a coil in parallel with an active load[23]). Our investigations have shown that if the indicated correction is disregarded, the temperature dependence of the imaginary part χ'' of the susceptibility can have a parasitic maximum in the temperature interval in which the total dynamic susceptibility changes most.

The temperature was measured with a copper-constantan thermocouple and a KG semiconductor (or a TSU-2 carbon) resistance thermometer with relative error not larger than ± 0.01 K. The chemical composition and the heat treatment of the investigated samples are given in Refs. 16–18.

3. FM-PM MAGNETIC TRANSFORMATION AT THE CURIE POINT

The values of the Curie point T_C were found by Arrott-Noakes method and by analyzing the temperature dependences of the nonlinear contributions to the dynamic magnetic susceptibility:

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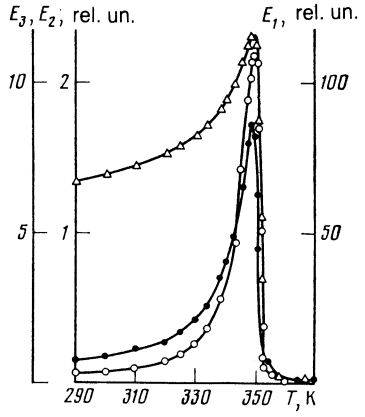

FIG. 1. Temperature dependences of the signals of the first (△) second (○), and third (●) harmonics for the $Ni_{95}Cr_5$ alloy. The amplitude of the magnetization-reversing field is 8 A/m.

$$\chi=\chi_0(\omega)+\chi_1(\omega)h_0+\chi_2(\omega)h_0^2+\chi_3(\omega)h_0^3+\ldots. \qquad (1)$$

where h_0 is the amplitude of the alternating magnetic field, and ω is the magnetization-reversal frequency.

Figure 1 shows by way of example the temperature dependence of signals $E_1=\chi_0 h_0$, $E_2\approx\chi_1 h_0^2$, and $E_3\approx\frac{3}{4}\chi_2 h_0^3$ measured respectively at the fundamental frequency ($\omega/2\pi=f=63$ Hz), at the second harmonic ($2f$), and at the third harmonic ($3f$) for the alloy $Ni_{95}Cr_5$.

Several methods exist for determining the Curie point T_C from the temperature dependence of the nonlinear susceptibility: from the position of the third-harmonic signal $E_3\propto\chi_2$ (Ref. 20), and by extrapolating to zero the steeply descending section of the second-harmonic signal E_2 which is proportional to the magnetization. In the case of homogeneous ferromagnets, these methods, and also the Arrott-Noakes method, give quite close values of T_C (the difference is usually less than 1%, see Fig. 1). The problem of finding the Curie point is more complicated for alloys with competing exchange interactions (e.g., of the Fe–Ni–Cr system), close in composition to the critical concentration x_c^f for the existence of the ferromagnetism. For example, the maximum of the third-harmonic signal of the $Fe_{65}Ni_{25}Cr_{10}$ alloy is observed at 107 K, while the second-harmonic signal E_2 is extrapolated to zero at 116 K, i.e., the values of the Curie point obtained by different methods differ by approximately 8%. The Arrott-Noakes method yields for the indicated alloy a value $T_C=116\pm1$ K. Analysis has shown that if the Curie point T_C is found from the position of the maximum of the third-harmonic signal, unphysical values ($\sim$5–10) are obtained for the critical exponent γ_0 of the linear susceptibility of Fe–Ni–Cr alloys with $x<x_c^f$. We therefore took the Curie point to be the temperature at which one can extrapolate to zero the steep section of the temperature dependence of the second-harmonic signal E_2 measured in the absence of a magnetizing field (in this case E_2 is proportional to the spontaneous magnetization).

The content of the chromiumatom in the $Fe_{65}Ni_{25}Cr_{10}$ alloy is quite close to the critical value for the existence of ferromagnetism in the quasibinary tie line of $Fe_{65}Ni_{35-x}Cr_x$, ($x_c^f\approx13$ Ref. 16). Therefore, in accordance with the results of the theoretical papers,[25,26] one can expect the second-order phase transition of this alloy to be "smeared out" at the Curie point. This, however, is obviously not the case, since the behavior of the magnetization of this alloy in the vicinity of the Curie point is described by the scaling law

$$M(H,T)=|\varepsilon|^{\beta}f_{\pm}(H/\varepsilon^{\beta\delta}) \qquad (2)$$

($\varepsilon=(T-T_C)/T_C$, H is the magnetic field, and β and δ are the critical exponents), which is valid for second-order phase transitions.[27] The scaling law for the magnetization of the $Fe_{65}Ni_{25}Cr_{10}$ alloy is obeyed at the following values of the critical exponents:

$$\beta=0.60\pm0.05,\quad \gamma=2.4\pm0.1,\quad \delta=5.0\pm0.4.$$

The values of the exponents β and γ of the $Fe_{65}Ni_{25}Cr_{10}$ alloy differ substantially from those obtained for a homogeneous Heisenberg (Ising) ferromagnet.[27]

Figure 2 shows the dependence of the linear susceptibility χ_0 on the reduced temperature (in log-log scale) for a number of alloys of the systems $Fe_{65}Ni_{35-x}Cr_x$ and $Ni_{100-x}Cr_x$. It can be seen that the power law

$$\chi_n=\chi_n^{+}\varepsilon^{-\gamma_n} \qquad (3)$$

(χ_n^{+} is the critical amplitude at $\varepsilon>0$) describes satisfactorily the behavior of the linear susceptibility of the indicated alloys in a certain interval $\varepsilon_0-\varepsilon_1$ of reduced temperatures. On the whole, the critical exponent γ_0 of the linear susceptibility depends on the reduced temperature, and it is more convenient to consider the effective Kouvel-Fisher critical exponent $\gamma_0^{*}=-d\lg\chi_0/d\lg\varepsilon$. Figure 3 shows a plot of $\gamma_0^{*}(\varepsilon)$ for a number of alloys of the $Ni_{100-x}Cr_x$ and $Fe_{65}Ni_{35-x}Cr_x$ systems located on the composition axis in the vicinity of the critical concentration at which the ferromagnetism vanishes, $x_c^f=10$ (Ref. 19) and $x_c^f=13$ (Ref. 16). Also shown are the values of γ_0^{*} calculated from the data of Ref. 28 for the $Eu_{0.6}Sr_{0.4}S$ alloy ($x_c^f\approx0.5$ in this system), and the $\gamma_0^{*}(\varepsilon)$ dependence for nickel and the amorphous alloy $Fe_{20}Ni_{56}B_{24}$.[29,30] The set of curves shown in Fig. 3 can be divided into three groups. For Ni and alloys of the system $Ni_{100-x}Cr_x$ with $x\lesssim x_c^f\approx10$ the critical exponent γ_0^{*} does not exceed the value $\gamma_0^{H}=1.38$ calculated for the three-dimensional Heisenberg model.[27] For alloys of the systems $Fe_{65}Ni_{35-x}Cr_x$ and $Eu_xSr_{1-x}S$ with $x\lesssim x_c^f$ the values of γ_0^{*}

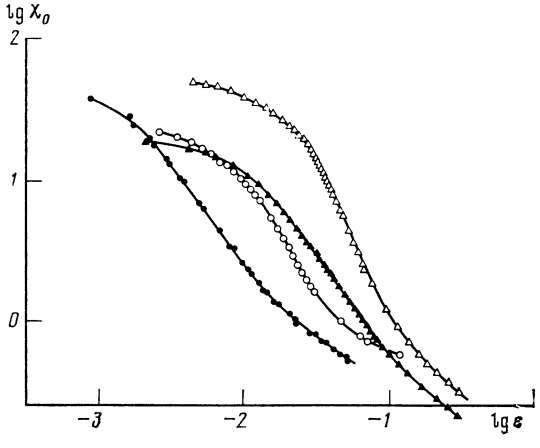

FIG. 2. Linear dynamic susceptibility vs reduced temperature for the alloys $Ni_{95}Cr_5$ (●), $Ni_{92}Cr_8$ (▲), $Fe_{65}Ni_{30}Cr_5$ (○) and $Fe_{65}Ni_{25}Cr_5$ (△).

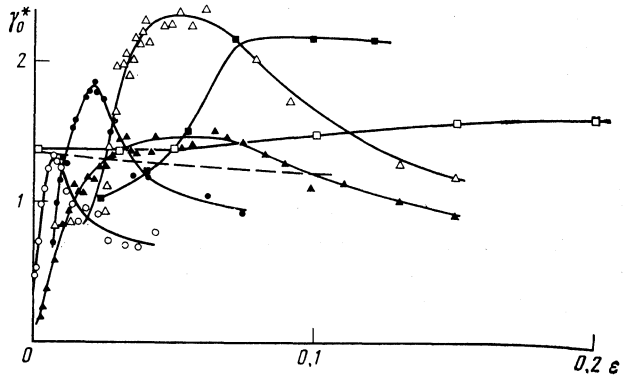

FIG. 3. Effective critical exponent γ_0^* of the Kouvel-Fisher linear susceptibility vs reduced temperature for the alloys $Ni_{95}Cr_5$ (○), $Ni_{92}Cr_8$ (▲), $Fe_{65}Ni_{30}Cr_5$ (●), $Fe_{65}Ni_{25}Cr_{10}$ (△), $Eu_{0.6}Sr_{0.4}S$ (■), (Ref. 28) and $Fe_{20}Ni_{56}B_{24}$ (□) (Ref. 29), and for Ni (dashed line).

in a certain reduced-temperature interval are substantially higher than $\gamma_0^H(\gamma_0^* \gtrsim 2)$. For amorphous alloys, the effective exponent has intermediate values ($\gamma_0^* \lesssim 1.6$–1.8). The experimental γ_0^* of all the considered ferromagnets varies along a curve with a maximum at a certain value $\varepsilon = \varepsilon_m$ ($\varepsilon_m \sim 10^{-3}$–$10^{-4}$ for Ni, and the maximum of γ_0^* is probably very broad for the $Eu_{0.6}Sr_{0.4}S$ alloy). At small ε, the effective exponent γ_0^* decreases as a result of the inhomogeneity and the finite dimensions of the sample, and also because the condition that the magnetic field be weak,[27]

$$H \ll H' = (k_B T_C / g\mu_B)\varepsilon^{\gamma_0+\beta}, \tag{4}$$

is not met; here g is the Landé factor, μ_B the Bohr magneton, and k_B the Boltzmann constant.

The amplitude of the magnetic field in our experiments was low enough ($H \sim 1$ A/m), so that the suppression of the critical fluctuations by the magnetic field was not the principal mechanism responsible for the substantial decrease of the exponent γ_0^* at $\varepsilon < \varepsilon_m$.

The scaling law (2) describes the behavior of the magnetization of Fe–Ni–Cr alloys with $x \lesssim x_c^f$ near the Curie point. As a result, the high values of the exponents γ_0^* of these alloys can obviously not be explained within the framework of a theory[29,31] in which the principal role is played by noncritical correlations and quasi-one-dimensionality effects. If the theory of Refs. 29 and 31 is used, it is difficult to understand why anomalously large values of γ_0^* are observed for Fe–Ni–Cr alloys with $x \lesssim x_c^f$ but not for Ni–Cr alloys (see Fig. 3). It is assumed in Ref. 31 that the increase of the effective exponent γ_0^* is due to "magnetic dilution" (decrease of the number of "magnetic" atoms in the "site problem"). The alternation of the signs of the exchange interaction (which is obviously a feature of Fe–Ni–Cr but not of Ni–Cr alloys) does not play a significant role in the theory of Ref. 31.

High and composition-dependent values of the exponents β and γ_0 were obtained for dilute ferromagnets by Sobotta[32] by the renormalization-group method. This result does not agree with the existing notions concerning the isomorphism of the critical behavior outside the immediate vicinity of the polycritical point.[27,33] Nonetheless, the experimentally obtained critical exponents of dilute ferromagnets agree better with the conclusion of the theory of Ref. 32 (see also Ref. 34). Anomalously high values of the exponents β and γ_0 are possessed by Fe–Ni–Cr (present paper), Ni–Cu (Ref. 34), and Fe–Cr (Ref. 36) alloys, and by the dielectric compounds EuSrS (Refs. 18 and 34) and EuSrSSe (Ref. 37). As the critical concentration x_c^f for the vanishing of the ferromagnetism is approached along the composition axis, no substantial change of the critical exponents of the magnetic transition at the Curie point is observed in Ni–Cr (present paper) and in Ni–Rh (Ref. 36) alloys.

It can be assumed that the critical exponents are influenced by the character of the exchange interaction. An alternating-sign exchange interactions (alloy systems EuSrS, EuSrSSe, Fe–Cr, and Fe–Ni–Cr) corresponds to the highest values of the critical exponents β and γ_0. Intermediate values of the critical exponents correspond to amorphous alloys. The lowest (close to those typical of a homogeneous ferromagnet) critical exponents obviously correspond to collectivized magnets such as Ni–Cr. The latter is probably not subject to large fluctuations of the exchange interaction. As for the collectivized magnets Ni–Cu and Ni–Rh, cluster effects are quite strong in them (alloys with "giant moments"[35,36]), making their classification difficult. Notice should be taken of the correlation between the values of the critical exponents that describe a phase transition at the Curie point, on the one hand, and the existence of spin glass at low temperatures, on the other. The SG state is a feature of alloys with anomalously large critical exponents β and γ_0 (Fe-Ni-Cr, EuSrS, Fe-Cr)[16,17,34,38] but not of alloys with "normal" values of the critical exponents (e.g., Ni-Cr, Ref. 39). Note that a substantial increase of the exponent γ_0 in $Fe_{65}Ni_{35-x}Cr_x$ sets in in that composition region ($x \approx 5$) in which SG appears on the magnetic phase diagram.[16]. As to the experimentally observed broadening of the critical region near T_C as x_c^f is approached on the composition axis (see Fig. 3 and Ref. 40), it is a feature of the vicinity of a multicritical point.[41] The broadening of the critical region as $x \to x_c^f$ suggests that the corresponding change of the observed critical exponents β and γ_0 is not due to crossover from a critical to a polycritical behavior (and the observed critical exponents are true ones rather than effective, se also Refs. 34 and 41).

Analysis of the critical behavior of quasibinary alloys $Mn_{20}Fe_xNi_{80-x}$ in the vicinity of the Curie point T_C is difficult. The plot of the magnetization vs temperature of these alloys has a long "tail" at $T \gtrsim T_C$. The presence of a magnetization tail all the way to $\varepsilon \approx 1$ may be due to the presence of chemical clusters of statistical origin or of regions of the ordered ferromagnetic phase $Ni_3(FeMn)$ (Ref. 42).

4. THE FM-SG TEMPERATURE TRANSITION

Figure 4 shows by way of example the temperature dependences of the real χ' and imaginary χ'' parts of the dynamic susceptibility of the alloys $Fe_{65}Ni_{25}Cr_{10}$ and $Fe_{40}Ni_{40}Mn_{20}$. The χ'' (T) plots of the investigated alloys has two maxima, one in the vicinity of the Curie point T_C and the other at low temperatures in the region where the real part χ' of the susceptibility falls off (see Fig. 4). The low-temperature maximum of χ'' is customarily attributed to SG formation.[3,13,14] Below the maximum point χ'' ($T < T_{fg}$) the alloys considered begin to exhibit other symptoms of irreversibility of the magnetic behavior, viz., an aftereffect with anomalously long characteristic times, an appreciable remanent

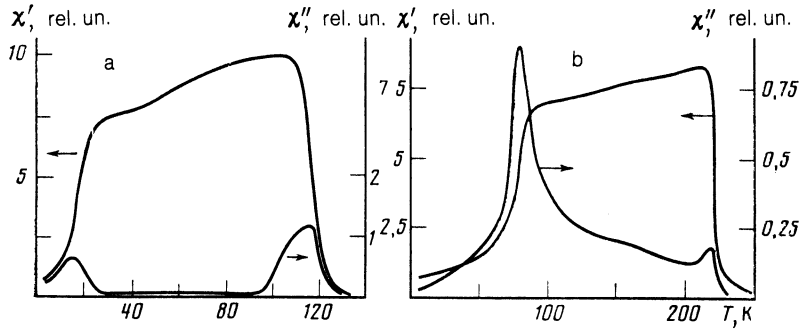

FIG. 4. Temperature dependences of the real (χ') and imaginary (χ'') parts of the linear dynamic magnetic susceptibility of the alloys $Fe_{65}Ni_{25}Cr_{10}$ (a) and $Fe_{40}Ni_{40}Mn_{40}$ (b).

magnetization, and others.[16,18] Note that in contrast to Refs. 9 and 10, we observed no additional maximum on the temperature dependence of the imaginary part χ'' of the susceptibility in the temperature interval $T_{fg} - T_C$ for Fe–Ni–Cr alloys with $x \lesssim x_c^f$.

In Fe–Ni–Cr alloys with composition close to the critical concentration for the vanishing of the FM, the behavior of the magnetization near the temperature T_{fg} can be described by a scaling law such as (2) (with ε replaced by $\varepsilon^* = (T - T_{fg})/T_{fg}$, β by $\bar{\beta}$, and δ by $\bar{\delta}$). As seen from Fig. 5, the set of points forms two branches, one corresponding to $T < T_{fg}$ and the other to $T > T_{fg}$. A similar result is obtained in investigations of a double temperature transition in amorphous alloys.[13,43,44] According to the phenomenological theory of Ref. 2 this points to the existence of two phases, FM and SG, separated by a critical line in the HT plane (and hence to violation of the long-range FM order below T_{fg}).

It must be recognized, however, that the theory of Ref. 2 does not take into account the replica symmetry breaking upon formation of SG below the temperature T_{fg}. It is therefore not clear how well the approach of Ref. 2 describes the behavior of the magnetization when SG is formed from the FM state. Note also that investigations of the $Fe_{0.55}Mg_{0.45}Cl_2$ alloy offer quite convincing evidence of the conservation of the long-range antiferromagnetic order on cooling below the SG freezing point.[45] The foregoing indicates that the question of conservation of the FM long range order when SG is produced below the temperature T_{fg} in the investigated Fe–Ni–Cr is still moot.

As shown by our investigations, the use of the Arrott-Noakes method and the kink method to study the FM-SG (ASM) transition encounters substantial difficulties. In particular, the Arrott-Noakes relation at $T \sim T_{fg}$ is linear only in strong magnetic fields $\sim 10^5$ A/m. Extrapolation to $H = 0$ from the interval $H \gtrsim 10^5$ A/m overestimates the spontaneous magnetization M_s at all temperatures below T_{fg}. Owing to the large deviation of the Arrott-Noakes relation from linearity, it is impossible to obtain M_s by extrapolation from the region of weak magnetic fields.

In the case of alloys with very low magnetic anisotropy (e.g., amorphous ones), the spontaneous magnetization M_s at $T \lesssim T_{fg}$ can be determined by using one modification, described in Ref. 46, of the kink method. The value of M_s is obtained from the intersection of the magnetization isotherm $M(H)$ with the "Rayleigh tie line" $H = NM$ (N is the demagnetizing factor). If, however, the investigated alloy has appreciable magnetic anisotropy, this can underestimate the spontaneous magnetization. Figure 6 shows the temperature dependence of the spontaneous magnetization. M_s obtained for the $Fe_{65}Ni_{25}Cr_{10}$ alloy by the kink method and by Arrott-Noakes method (with extrapolation from the magnetic-field region where the Arrott-Noakes relations are linear).

It can be concluded that the spontaneous magnetization M_s decreases below the freezing temperature T_{fg}. It must be borne in mind, however, that the abrupt decrease of M_s near T_{fg}, which was obtained by using the kink method (see also Ref. 10), can be due to an error introduced into this method by the presence of magnetic anisotropy (see above).

As to $Mn_{20}Fe_xNi_{80-x}$ alloys with $x \lesssim x_c^f$, analysis of the behavior of their magnetization in the vicinity of the freezing point T_{fg} is made difficult by the presence of a small amount of "parasitic" FM phase at all temperatures from 4.2 K to $(1.5–2)T_C$ (most likely, of regions with partial ordering of

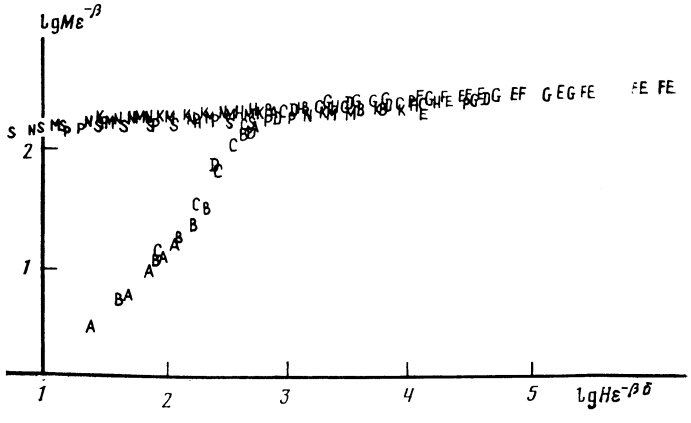

FIG. 5. Applicability of the scaling law to the description of an aggregate of magnetization isoterms of the alloys $Fe_{65}Ni_{25}Cr_{10}$ in the vicinity of the magnetic-transition temperature T_{fg}: A—4.2 K; B—7.4 K; C—10.8 K; D—14.5 K; E—18.2 K; F—20.9 K; G—22.3 K, H—27.6 K, K—30.0 K; M—35.3 K, N—38.0 K; P—44.2 K, S—53.5 K; $\beta = 0.28$; $\delta = 8$.

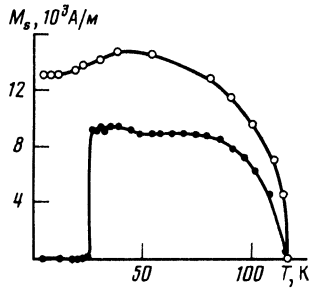

FIG. 6. Temperature dependence of the spontaneous magnetization M_s of the alloy $Fe_{65}Ni_{25}Cr_{10}$, determined by the Arrott-Noakes method (○)— and by the kink method (●).

the Ni_3(FeMn) type[42]). It can be assumed that spin waves with a "ferromagnetic" dispersion law, observed at $T < T_{fg}$ in $Ni_{100-x}Mn_x$ alloys with $x \lesssim x_c^f$ (Ref. 7) are also due to the presence of a parasitic FM phase.

5. CONCLUSION

In quasibinary alloys $Fe_{65}Ni_{35-x}Cr_x$ with $x \lesssim x_c^f$, in a definite interval of the reduced temperatures near the Curie point, the effective critical exponent of the linear susceptibility reaches anomalously high values ($\gamma_0^* \geqslant 2$), whereas the exponent γ_0^* of the corresponding alloys of the $Ni_{100-x}Cr_x$ system does not exceed the values calculated for the Heisenberg model ($\gamma_0^* \approx \gamma_0^H \approx 1.38$). The reason may be that the Ni–Cr alloys (in contrast to fcc Fe–Ni–Cr alloys) are not characterized by an alternating-sign exchange interaction. The behavior of the magnetization of alloys of the Fe–Ni–Cr system with $x \lesssim x_c^f$ in the vicinity of the SG formation temperature T_{fg} is described by a scaling law of the type (2). Within the framework of a phenomenological theory (see, e.g., Ref. 2) this means destruction of the long-range FM order on cooling below T_{fg}. Results of analysis using the kink method and the Arrott-Noakes method point to a decrease of the spontaneous magnetization M_s when SG is produced. Recognizing, however, that the applicability of the aforementioned methods of determining the temperature dependence of M_s to analysis of the FM–SG transition is not obvious, one must refrain from drawing final conclusions.

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