The phase transition and critical phenomena in concentrated spin glasses

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The linear and nonlinear dynamic magnetic susceptibility of the concentrated spin glasses (SG) $FeNiCr_{0.20}$, $FeNiMn_{0.15}$, and $NiMn_{0.27}$ have been studied. It is shown that the transition to the SG state in the $FeNiCr_{0.20}$ alloys possesses all the features of a true phase transition and is described by power-law dependences of the relaxation time, of the Anderson-Edwards order parameter and of the nonlinear magnetic susceptibility near the critical temperature T_f. The values of the critical indices obtained experimentally agree with the values predicted by mean field theory. It is shown that the appearance of SG in the alloys $NiMn_{0.27}$ and $FeNiMn_{0.15}$ is not related to a phase transition at T_f, but to processes in which the magnetic moments gradually freeze over a wide temperature range.

1. INTRODUCTION

There is at present active discussion of the question whether a spin glass (SG) is formed as the result of a true phase transition at a single, well defined temperature, or whether it is due to processes of slow relaxation of the magnetic moments of the system over a wide temperature range.[1] The first point of view is based on the ideas of Edwards and Anderson[2] who introduced a new order parameter for the SG characteristic

$$q_{EA}=\langle\langle s_i\rangle_T{}^2\rangle_J, \qquad (1)$$

where $\langle...\rangle_T$ indicates thermodynamic and $\langle...\rangle_J$ indicates configurational averaging. Then $q_{EA}=0$ if $T>T_f$ and $q_{EA}\leqslant 1$ for $T<T_f$, where T_f is the temperature of the phase transition. In the view of many authors, the appearance of a "paramagnetic–spin-glass (PM–SG) transition may be manifested in the frequently seen existence of maxima in the temperature dependences of the low-field magnetic susceptibility near T_f.[3]

According to one of the alternative points of view,[4] a SG can be regarded as an assembly of noninteracting superparamagnetic clusters, gradually aligning their magnetic moments in the fields of the anisotropy with decreasing temperature. Moreover, several computer calculations[5] indicate that the lowest critical dimensionality of a SG is $d=4$, and thus a PM–SG phase transition in a three-dimensional space is impossible. At the same time, numerical modeling of a SG carried out recently[6] nevertheless indicates the existence of a phase transition to the SG phase in a three-dimensional space.

It is clear from what has been said that on the theoretical level the situation is rather confused. It is also important to note that at present no reliable experimental criteria have been developed for determining the mechanism for a SG to arise in specific systems. More precisely, it is not quite clear which macroscopic SG properties should be measured and under what conditions measurements should be made, in order to provide an unequivocal answer to the question whether there is a process for establishing of the magnetic ground state of such systems or whether it is associated with the gradual freezing-in over a wide temperature range.

Although a PM–SG phase transition evidently does actually take place in the classical dilute spin-glass systems (CuMn, AuFe, PdFeMn, etc.), it remains an open question for concentrated SG.

In the present work we investigate the dynamic linear and nonlinear magnetic susceptibility of concentrated FeNiCr alloys and show that the PM–SG transition is a sharp phase transition at a temperature T_f, accompanied by a critical slowing-down of the relaxation time. At the same time, the appearance of a SG in some other systems (Ni_3Mn, $FeNiMn_{0.15}$) is the result of pure relaxation processes over a wide temperature range.

2. EXPERIMENTAL METHOD

For these investigations fcc alloys were chosen which go into the SG state at low temperatures: $Fe_x Ni_{0.80-x} Cr_{0.20}$ (Ref. 7), $Fe_x Ni_{0.85-x} Mn_{0.15}$ (Ref. 8) and $Ni_{0.73}Mn_{0.27}$ (Ref. 9). The method of preparing the specimens has been described earlier.[10]

Neutron diffraction analysis carried out on the diffractometer of the Institute of Metal Physics of the Ukrainian SSR showed that all the specimens were disordered and had a single-phase fcc structure. No noticeable excess intensity over the background was observed in the diffraction photographs obtained at a temperature of 4.3 K in the region of the (100) and (110) superstructure reflections. This is evidence of the absence in all the alloys studied of short-range and long-range antiferromagnetic order.

The real and imaginary parts of the dynamic magnetic susceptibility and also the nonlinear dynamic susceptibility, measured at the third harmonic of the frequency of the exciting magnetic field were studied on an apparatus described elsewhere.[11] The static magnetization was measured on a vibrating specimen magnetometer.

3. EXPERIMENTAL RESULTS AND DISCUSSION

3.1 Critical SG dynamics

A dynamic theory of a SG, essentially based on the idea of a true PM–SG phase transition, has recently been developed

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through the efforts of a number of authors[12,13] using the molecular field approximation with finite interaction radius. According to this theory, the relaxation time Γ_0^{-1} of the magnetic moments of a spin-glass system undergoes a critical slowing down in a critical region near the temperature T_f, and the following relations are then valid

$$\Gamma_0^{-1} \propto \tau^{-\lambda}, \quad \lambda=1, \tag{2}$$

where $\tau = (T/T_f - 1)$ is the reduced temperature and λ is the dynamic critical index. On the assumption that the PM state is characterized by a single relaxation time, it is not difficult to find the relation between Γ_0^{-1} and the components of the dynamic magnetic susceptibility[14] for $T > T_f$:

$$\Gamma_0^{-1} = \chi_0''(\omega, T)/\omega\chi_0'(\omega, T), \tag{3}$$

where χ_0' and χ_0'' are the real and imaginary components of the dynamic susceptibility, and ω is the angular frequency of the exciting magnetic field.

It is shown in the dynamic SG theory[12,15] that

$$\chi_0''(\omega) \propto \omega^{\nu}, \tag{4}$$

where the dynamic critical index ν is temperature-dependent. In particular

$$\nu = \begin{cases} 1, & T>T_f, \\ {}^1/_2, & T=T_f, \\ 0{,}395, & T\to 0. \end{cases} \tag{5}$$

This dependence of ν is associated with the violation of the fluctuation-dissipation theorem for $T < T_f$.

Consequently, by studying the temperature dependences $\chi_0'(T)$ and $\chi_0''(T)$ for $T > T_f$ and the frequency dependence $\chi_0''(\omega)$ for specific SG, it is possible to verify the predictions of the theory [Eqs. (2), (4), and (5)]. In passing, we note that in spite of the appreciable amount of work on studying the dynamic magnetic susceptibility of SG,[16] as far as the present authors are aware this problem has not been considered previously.

We now go on to an analysis of the results obtained. We first consider the $Fe_xNi_{0.80-x}Cr_{0.20}$ alloys. Typical temperature dependences of the real χ_0' and imaginary χ_0'' components of the dynamic magnetic susceptibility of a SG (the alloy $Fe_{0.58}Ni_{0.22}Cr_{0.20}$ are shown in Fig. 1, measured in a 3-Oe alternating magnetic field. The component χ_0' has a sharp maximum near T_f (marked by an arrow), while χ_0'' grows strongly on lowering the temperature. This behavior of χ_0' and χ_0'' is perfectly characteristic of classical SG[16,17] and can serve as evidence of a true phase transition taking place. In fact, the temperature dependence of the relaxation time Γ_0^{-1} was calculated according to Eq. (3) by using the experimental results (the inset to Fig. 1). It can be seen that in the critical region near T_f, Γ_0^{-1} undergoes a sharp discontinuity characteristic of second-order phase transitions.[18]

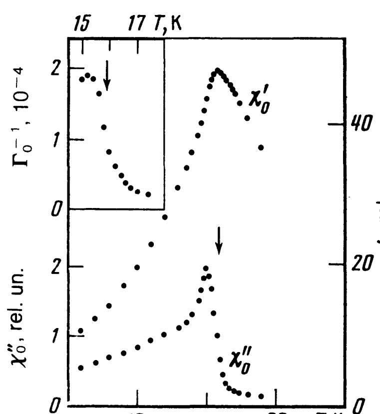

FIG. 1. The temperature dependence of the real (χ_0') and imaginary (χ_0'') components of the dynamic magnetic susceptibility and of the relaxation time Γ_0^{-1} (on the inset) for the spin glass $Fe_{0.58}Ni_{0.22}Cr_{0.20}$. The amplitude of the alternating magnetizing field was 3 Oe, the frequency 36 Hz.

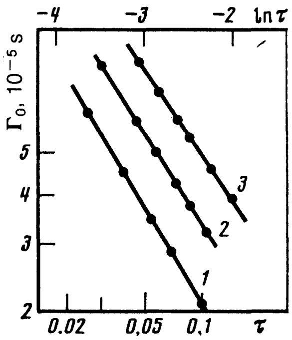

FIG. 2. The dependence of the relaxation time Γ_0^{-1} on the reduced temperature τ on a double logarithmic plot for the alloys $Fe_xNi_{0.80-x}Cr_{0.20}$ for the following values of x: 1) 0.20; (2) 0.22; (3) 0.04.

Analysis of the experimental data, the results of which are shown in Fig. 2, enabled, on the one hand, the true temperature T_f of the freezing-in of the SG to be determined, and on the other hand the dynamic critical index λ, characteristic of the critical lengthening of the relaxation time as the temperature approaches T_f from the paramagnetic region. It turned out that for the alloys studied (Fig. 2) in small (less than 3 Oe) exciting fields, Eq. (2) is satisfied with the dynamic critical index $\lambda = 1.00 \pm 0.05$ in complete agreement with the predictions of theory.[12]

It should be noted that an increase in the amplitude of the exciting magnetic field gives rise to a reduction in the critical index λ. For example, at a field amplitude of 10 Oe, the corresponding value of λ was found to be ≈ 0.3 for the alloy $Fe_{0.58}Ni_{0.22}Cr_{0.20}$. It is clear from what has been said that the critical behavior of Γ_0^{-1} given in Eq. (2) should disappear in sufficiently high magnetic fields.

In finishing the discussion of the critical dynamics of FeNiCr SG we note that the frequency dependences of the imaginary part χ_0'' of the dynamic magnetic susceptibility were also studied. It was found that Eq. (4) is satisfied over a wide temperature interval for the alloys $Fe_{0.04}Ni_{0.76}Cr_{0.20}$, $Fe_{0.60}Ni_{0.20}Cr_{0.20}$ and $Fe_{0.58}Ni_{0.22}Cr_{0.20}$, and in all cases the value of the critical index ν agreed with Eq. (5). As an example, the temperature dependence of ν for the alloy $Fe_{0.58}Ni_{0.22}Cr_{0.20}$ is shown in Fig. 3. It can be seen in fact that in the paramagnetic region $\nu = 1.00 \pm 0.05$. This indicates that the fluctuation-dissipation theorem is satisfied for

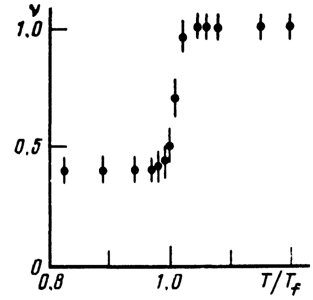

FIG. 3. The temperature dependence of the critical dynamical index for the alloy $Fe_{0.58}Ni_{0.22}Cr_{0.20}$.

$T > T_f$ and that the Debye approximation (Eq. 3) is justified in calculating the critical behavior of the relaxation time Γ_0^{-1}. In addition, in agreement with Eq. (5), in fact, $\nu = 0.5 \pm 0.1$ for $T = T_f$ and $\nu = 0.40 \pm 0.05$ in the temperature region $T < T$.

The analysis of the experimental results given above and their comparison with theoretical calculations thus enable one to confirm that in the disordered $FeNiCr_{0.20}$ alloys, the SG state arises in the result of a true phase transition, accompanied by a critical increase in the relaxation time within a narrow temperature region near T_f. In the two following sections of the present work this conclusion will be confirmed. The alloy system just discussed and those similar to it can be called typical, or Edwards-Anderson, SG. The dilute AuFe, CuMn, and PdFeMn alloys and some amorphous alloys based on iron, nickel and manganese should evidently be included among them.

In contrast to the case described above, a completely different picture is observed in the alloys $Ni_{0.73}Mn_{0.27}$ and $Fe_{0.58}Ni_{0.27}Mn_{0.15}$, where in the view of Menshikov *et al.*[8]

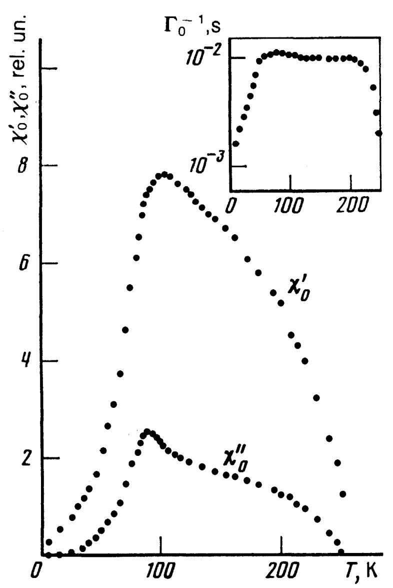

FIG. 4. The temperature dependence of the real and imaginary components of the dynamic magnetic susceptibility and of the relaxation time Γ_0^{-1} (on the inset) of the disordered alloy $Ni_{0.73}Mn_{0.27}$. The amplitude of the alternating magnetizing field was 1 Oe, the frequency 36 Hz.

and Goldfarb and Patton[9] SG also arise. As an example, the temperature dependences of χ_0', χ_0'' and Γ_0^{-1} for the alloy $Ni_{0.73}Mn_{0.27}$ are shown in Fig. 4. It can be seen that unlike χ_0' and χ_0'', Γ_0^{-1} has no sharp discontinuity and remains practically constant over the whole temperature interval, including the vicinity of the temperature of the maxima of χ_0' and χ_0''. Consequently, no critical increase in Γ_0^{-1} in the form of Eq. (2) and characteristic of FeNiCr alloys is observed in the present case. It can be concluded from this that a sharp phase transition to the SG state is absent in the alloy $Ni_{0.73}Mn_{0.27}$. For such systems the formation of the magnetic ground state is most likely the result of pure kinetic freezing-in processes, which take place over a wide temperature interval.

As confirmation of what has been said, we give the results of a study of the static magnetization of the alloy considered (Fig. 5). The magnetization of the alloy when it is cooled in a constant field of 10 Oe from a temperature of 210 to 4.2 K (curve 2) is higher that the magnetization obtained after cooling it to 4.2 K in zero magnetic field (curve 1). It should, however, be taken into account that in this case curves 1 and 2 coincide close to or below the temperature T_M of the maximum magnetization.[19,20] In the present case such a coincidence occurs at the temperatures T_{cool}, from which the alloy was cooled, and as can be seen from Fig. 5, $T_{cool} \gg T_M$. This indicates unequivocally that the freezing-in processes in the present case do not take place at one strictly determined temperature but over a wide temperature range.

In conclusion we note that a qualitatively similar picture of the temperature variation of χ_0', χ_0'', Γ_0^{-1} and the magnetization is also observed in the alloy $Fe_{0.58}Ni_{0.27}Mn_{0.15}$.

It follows from what has been said that the temperatures of the maxima of χ_0', and of the low-field magnetization which are often identified with the temperatures of the SG freezing-in, are not of this kind in the alloys $Ni_{0.73}Mn_{0.27}$

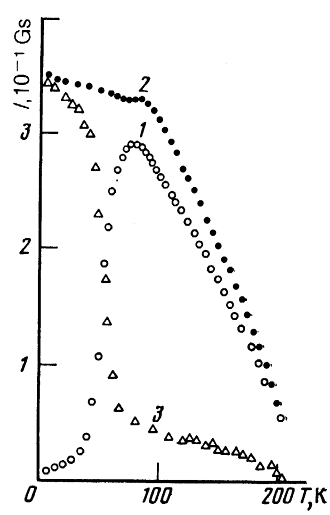

FIG. 5. The temperature dependence of the static magnetization of a specimen of the alloy $Ni_{0.73}Mn_{0.27}$ for different thermomagnetic treatments: 1) after cooling to 4.2 K in zero magnetic field, 2) after cooling in the measuring field of 10 Oe. Curve 3 was obtained by subtracting curve 1 from curve 2.

and $Fe_{0.58}Ni_{0.27}Mn_{0.15}$. Therefore, the lines on the corresponding phase diagrams[8] defining the SG region are not critical lines and are devoid of physical meaning.

3.2 Critical behavior of the Edwards-Anderson order parameter

At present two types of magnetic susceptibility of SG are distinguished: the susceptibility in thermodynamic equilibrium χ_e and the nonequilibrium form χ_{ne}.[1] The former is pratically independent of time and temperature in the region $T< T_f$ and can be obtained after cooling a SG in a sufficiently weak magnetic field. It is important to emphasize that a SG possesses magnetic viscosity in the temperature region $T<T_f$. Because of this χ_{ne} grows with time, tending towards its limiting value χ_e.[1] On the other hand the real part χ_0' of the magnetic susceptibility of a SG, being completely reversible,[16] can be identified with the true nonequilibrium susceptibility χ_e. It is not difficult to determine the temperature dependence of the order parameter q_{EA} of the defining equation (1) after obtaining by experiment the variation of $\chi_0'(T)=\chi_{ne}(T)$. In fact, the relation between q_{EA} and χ_{ne} has been obtained on the basis of the Edwards-Anderson model in the molecular field approximation:

$$\chi_{ne}=(C/k_BT)(1-q_{EA}), \tag{6}$$

where C is the Curie constant, while the other symbols have the usual meaning.

In the dynamic SG theory[12,21] a new order parameter Δ is introduced for the characteristics of the equilibrium susceptibility χ_e, so that

$$\chi_e(T)=\chi_{ne}(T)+\Delta(T)C/k_BT.$$

The parameter Δ is a measure of the nonergodicity of a SG. Leaving aside a consideration of the $\Delta(T)$ dependence, first obtained by Yeshurun,[22] we shall be interested in the temperature dependence of q_{EA}. Then, taking into account that for real alloys the paramagnetic Curie temperature satisfies $\theta\neq 0$, Eq. (6) can be transformed into the form

$$q_{EA}=1-T\chi_{ne}(T)/[C+\Theta\chi_{ne}(T)]. \tag{7}$$

The temperature dependence of q_{EA} has been calculated[12,23] and it was shown that $Q_{EA}=\tau+\tau^2$ where $\tau=1-T/T_f$. The theory thus predicts a power law for the temperature variation of q_{EA} near T_f:

$$q_{EA}\propto\tau^{\beta},\quad \beta=1. \tag{8}$$

As an example we shall consider the alloy $Fe_{0.58}Ni_{0.22}Cr_{0.20}$. The temperature dependence of q_{EA} was calculated (Fig. 6a) according to Eq. (7), using the experimental results (Fig. 1). It is easy to convince oneself that the behavior of q_{EA} resembles qualitatively the variation of the spontaneous magnetization (the order parameter) of a uniform ferromagnet. This fact indirectly points to a PM–SG phase transition taking place at $T=T_f$. Additional arguments for such a conclusion can be obtained from the analysis of the critical behavior of $q_{EA}(T)$. The temperature dependence of q_{EA} for this alloy is shown in Fig. 6b on a double logarithmic plot. The results are well approximated by a

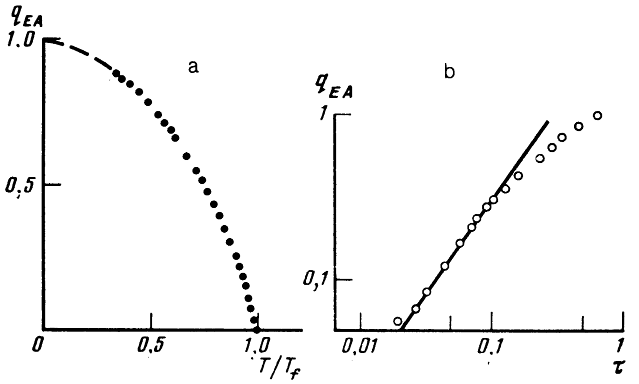

FIG. 6 a) The temperature dependence of the Edwards-Anderson order parameter q_{EA} for the spin glass $Fe_{0.58}Ni_{0.22}Cr_{0.20}$ b) the dependence of q_{EA} on the reduced temperature τ.

straight line in the region of temperatures near T_f. The power law of Eq. (8) is thus valid, with the experimental value $\beta=1.00\pm0.05$ in complete accord with theory. It is worth remembering that values of β close to this were obtained in static experiments[19] on the classic SG $Cu_{0.98}Mn_{0.02}$.

It can thus be concluded from an analysis of the temperature dependence of the Edwards-Anderson order parameter q_{EA} that the PM–SG transition in the alloy $Fe_{0.58}Ni_{0.22}Cr_{0.20}$ is really an Edwards-Anderson phase transition.

3.3 The nonlinear dynamic magnetic susceptibility

If a small magnetic field h is applied to a SG, then its total susceptibility $\chi(T)$ can be represented in the form

$$\chi(T)=\chi_0(T)+\chi_2(T)h^2+\chi_4(T)h^4+\dots \tag{9}$$

While the linear susceptibility $\chi_0(T)$ does not show critical behavior at the SG freezing-in point, the nonlinear susceptibility $\chi_2(T)$, being negative, has a singularity at T_f (Ref. 24). The temperature dependence $\chi_2(T)$ of the following form was obtained[25] within the framework of the dynamical SG theory,[12] based as indicated on the concept of a PM–SG phase transition:

$$|\chi_2|\propto\tau^{-\gamma_s},\quad \gamma_s=\begin{cases}1, & T>T_f\\ 2, & T<T_f\end{cases}, \tag{10}$$

where $\tau=|1-T/T_f|$.

We shall explain the applicability of Eq. (10) to the real SG $Fe_{0.58}Ni_{0.22}Cr_{0.20}$, studied in the preceding sections. The temperature dependence of the dynamic nonlinear susceptibility $\chi_2(T)$ obtained in an alternating magnetic field of 1 Oe is shown in Fig. 7a, together with χ_0'. As might be expected, the anomaly in χ_2 is much greater than in χ_0 near T_f. It follows from this that for locating the PM–SG characteristic transition temperatures, it is more convenient to make use of the results of measurements of $\chi_2(T)$.

It can be seen from Fig. 7b, where the temperature dependence of χ_2 is shown on a double logarithmic plot, that the power law of Eq. (10) is satisfied on both sides of T_f, with the experimental values of the critical index for the alloy studied coming out as $\gamma_s=0.8\pm0.1$ for $T>T_f$ and $\gamma_s=1.4\pm0.1$ for $T<T_f$. The result obtained is in satisfac-

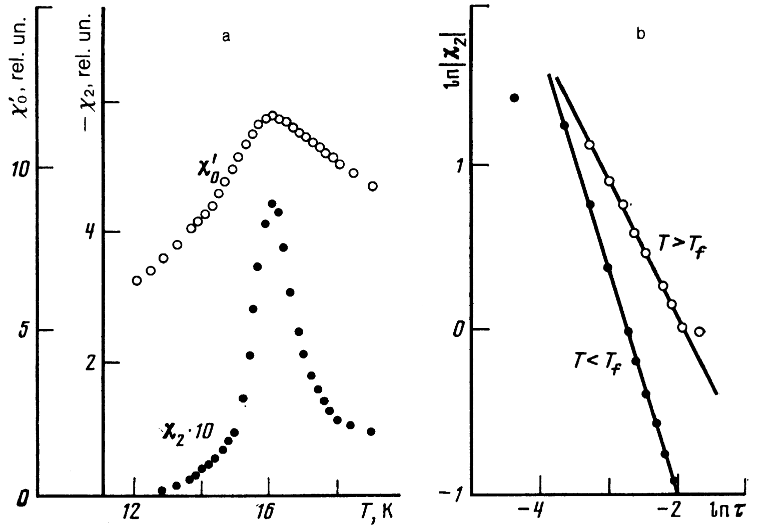

FIG. 7. a) The temperature dependence of the dynamic linear χ'_0 and nonlinear χ_2 magnetic susceptibility of the alloy $Fe_{0.58}Ni_{0.22}Cr_{0.20}$; b) the dependence χ_2 on the reduced temperature τ. The amplitude of the alternating magnetizing field was 1 Oe, the frequency 60 Hz.

tory agreement with the predictions of the mean field theory.[25]

It should be noted that on increasing the amplitude of the exciting magnetic field up to 10 Oe, the power-law divergence of χ_2 changes to a logarithmic form $\chi_2 \propto \ln T$. A similar result was obtained for a number of spin-glass systems[26] and is associated with the growth in the contribution to the measured nonlinear susceptibility of the terms $\chi_4(T)$, $\chi_6(T)$, etc., [Eq. (9)].

The experimental results on the critical behavior of the nonlinear susceptibility $\chi_2(T)$ presented in this section thus also indicate the onset of a PM–SG phase transition in the alloy studied.

4. CONCLUSIONS

It has been shown experimentally as a result of an investigation of the linear and nonlinear magnetic susceptibility, that in the disordered alloys $Fe_x Ni_{0.80-x} Cr_{0.20}$ the PM–SG transition possesses the features of a true phase transition and is well described by the present SG theories,[12,15,25] based on the mean field approximation with infinite interaction radius. Such a transition is characterized by a critical slowing down of the relaxation time Γ_0^{-1}, by a critical behavior of the Edwards-Anderson order parameter q_{EA} and by a power law divergence of the nonlinear magnetic susceptibility $\chi_2(T)$ near the temperature T_f of the freezing-in of the SG [Eqs. (2), (8) and (10)]. Moreover, the study of the behavior of the freezing-in temperature of the $FeNiCr_{0.20}$ SG in magnetic fields[11,20] indicates that the equation of the $T_f(H)$ line in the H–T plane is also described by the mean field theory.

The concentrated $Fe_x Ni_{0.80-x} Cr_{0.20}$ alloys together with the classical spin-glass systems (AuFe, CuMn, etc.) can, in view of what has been said, be called typical or Edwards-Anderson SG. In addition, cases are possible when the SG state arises as a result of slow relaxation processes or of the gradual freezing-in of the magnetic moments over a wide temperature region (the alloys $Ni_{0.73}Mn_{0.27}$ and $Fe_{0.58}Ni_{0.27}Mn_{0.15}$). Such systems should be regarded as atypical SG.

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