Frequency and field dependence of the "freezing" temperature of FeNiMn spin glasses

A. V. Deryabin and A. V. T'kov

Scientific-Research Institute of Applied Physics at the A. A. Zhdanov State University, Irkutsk
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A study is made of the inverse magnetic susceptibility of alloys of the quasibinary tie line $Mn_{20}Fe_x Ni_{80-x}$. A wide existence region for the spin glass (asperomagnetism) is found, and it is discovered that the position of the peak on the temperature curve of the inverse susceptibility of FeNiMn spin glasses depends strongly on the magnetization-reversal frequency. It is shown that the field and temperature dependence of the inverse susceptibility in the vicinity of the "freezing" temperature T_f is described by a power law, but the critical exponents have anomalous values. The features of the magnetic transformation at T_f are attributed to the nonergodicity of the spin glass.

1. INTRODUCTION

There is particular interest right now in the dynamical properties of spin glasses; in particular, in the temperature, frequency, and field dependence of the inverse magnetic susceptibility.[11] The main goal of such studies is to find out whether or not the formation of the spin glass is a true phase transition. Properties characteristic of a spin glass have in recent years been detected in alloys of ferromagnetic 3*d*-metals with antiferromagnets (e.g., FeNiMn,[2–5] FeCr,[6,7] and FeNiCr[8–10]). In these alloys the direct exchange interaction varies in sign; this corresponds to the spin-glass model[11,12] considered in the majority of the theoretical papers. The most convenient objects of study are the FeNiMn spin glasses, since the "freezing" temperature T_f in these alloys reaches 100–110 K.[2,3,5]

We have studied the differential inverse magnetic susceptibility χ in alloys of the quasibinary tie line $Mn_{20}Fe_x Ni_{80-x}$. It is known that a concentrational ferromagnetic-antiferromagnetic transition occurs on this tie line (the critical concentration for the existence of ferromagnetism is $x_c \approx 43$).[2,3] However, the frequency and field dependence of the inverse susceptibility of these alloys had not been studied.

In addition to the linear magnetic susceptibility, we also studied the nonlinear susceptibility. The measurement techniques were described in Ref. 10. The samples were fused from pure components, forged, drawn down in diameter from 8 to 5, and, after annealing at 800 °C, quenched in oil. According to x-ray diffraction data, all the samples had a stable fcc structure at temperature down to 4.2 K.

2. MAGNETIC PHASE DIAGRAM

Figure 1 show the temperature dependence of the linear differential inverse magnetic susceptibility χ of a number of alloys of the system $Mn_{20}Fe_x Ni_{80-x}$. It is seen that these curves are transformed by the application of a static magnetic field H parallel to the alternating magnetization-reversing field. In alloys with $x = 45$–49 the $\chi(T, H)$ curves exhibit a single peak at a temperature T_f. With increasing bias field H this peak decreases in height and shifts to lower temperatures (see Fig. 1c) in a manner similar to that in RKKY[1] and

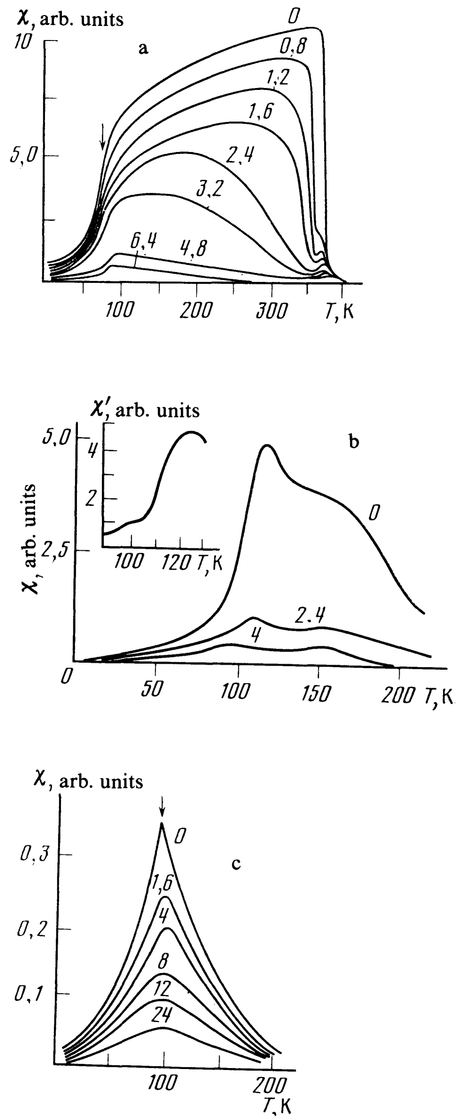

FIG. 1. Temperature dependence of the linear inverse susceptibility of alloys of the system $Mn_{20}Fe_x Ni_{80-x}$ with $x = 30$ (a), 43 (b), and 45 (c). The numbers on the curves give the values of the static bias field (in kA/m). The inset in (b) shows the temperature dependence of the nonlinear susceptibility χ' of the alloy $Fe_{43}Ni_{37}Mn_{20}$ as measured in the absence of a bias field.

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"frustrated-insulator"[13] spin glasses. In alloys with $x = 50$ and 52 the inverse susceptibility is very small, so that one can consider their ground state to be antiferromagnetic (see also Ref. 3).

The temperature curves of the inverse susceptibility $\chi(T, H)$ have a different shape in alloys of the $Mn_{20}Fe_xNi_{80-x}$ system with an iron content of from 0 to 40 at. %. Below a certain temperature T_f there is a drop-off on the $\chi(T, 0)$ curve (see Fig. 1a). In the presence of a bias field H the $\chi(T, H)$ curves have two local maxima: one near T_f, and the other in the vicinity of the Curie point T_c. With increasing H the low-temperature peak is shifted downward along the temperature axis, while the high-temperature peak is shifted upward. Such a shape of the $\chi(T, H)$ curves is characteristic for a ferromagnet–spin-glass mixed state (the asperomagnetic state).[1,9,10,14]

Of particular interest is the shape of the $\chi(T, H)$ curve in the alloy $Mn_{20}Fe_{43}Ni_{37}$ (Fig. 1b). The temperature dependence of the inverse susceptibility measured in the absence of a bias field H exhibits a single peak with an asymmetric shape. Applying a bias field, however, will "split" this peak into two (i.e., the $\chi(T, H)$ curve will resemble that in alloys with $x = 0$–40). The temperature dependence of the nonlinear susceptibility x shows splitting even at $H = 0$ (see the inset in Fig. 1b). The behavior of the inverse susceptibility of the alloy $Mn_{20}Fe_{43}Ni_{37}$ suggests that its composition corresponds to a tricritical point on the magnetic phase diagram of the $Mn_{20}Fe_xNi_{80-x}$ system. We note that at the present time much significance is attached to the study of multicritical points on the magnetic phase diagram.[15]

The magnetic phase diagram of the $Mn_{20}Fe_xNi_{80-x}$ system, as constructed from our results on the differential inverse susceptibility (and also from neutron-diffraction data[3]), is shown in Fig. 2. The values of the freezing temperature T_f in alloys with $x = 0$–40 were determined from the bend on the $\chi(T, 0)$ curve or from the position of the low-temperature peak on the $\chi(T, H)$ curve for $H = 4$–8 kA/m. The values obtained in the two cases were rather similar. The values of the Curie temperature T_c were found from the position of the peak on the $\chi(T, 0)$ curve (see Fig. 1a) and also from analysis of the field dependence of the position of the high-temperature peak of χ. It is seen that the asperomagnetism region in the $Mn_{20}Fe_xNi_{80-x}$ system is considerably wider ($0 < x < 43$) than was assumed in Ref. 3.

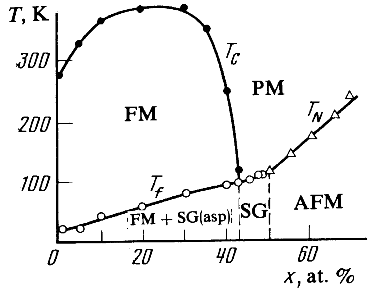

FIG. 2. Magnetic phase diagram of the quasibinary tie line $Mn_{20}Fe_xNi_{80-x}$. The diagram was constructed using our results on the inverse susceptibility (●, ○) and the data of Ref. 3 (△); T_n is the Néel temperature.

3. FREQUENCY DEPENDENCE OF THE FREEZING TEMPERATURE T_f

The manner in which the freezing temperature T_f of the spin glass depends on the measurement time τ (magnetization-reversal frequency ν) is a test of whether the transformation at T_f is a true phase transition.[16] In RKKY and frustrated-insulator spin glasses there is an observable shift in T_f when ν changes, but this effect, as a rule, is small (of the order of 0.1 K.[17]

We have studied $T_f(\nu)$ in alloys of the $Mn_{20}Fe_xNi_{80-x}$ system with $x = 43$ and 45. It was found that a change in the magnetization-reversal frequency by less than two orders of magnitude (from 17 to 1020 Hz) caused the peak of the linear inverse susceptibility to be shifted upwards by $\Delta T_f = 2.2$ K. This is twice as high as the largest value of ΔT_f previously observed in spin glasses.[17]

An even larger value was found for the shift of the peak of the linear susceptibility χ as a function of ν in the alloy $Fe_{43}Ni_{37}Mn_{20}$. However, measurments of the nonlinear susceptibility χ' have shown that as the magnetization-reversal frequency increases, the χ' peak, which corresponds to the Curie point T_c (see the inset in Fig. 1b), is shifted to higher temperature. The shift of the high–temperature peak of χ' is anomalously large (it reaches 6 K for a change in the magnetization-reversal frequency ν by a factor of less than twenty). So far as we know, this effect has not been seen before.

It is not possible to determine the frequency dependence of T_f from the inverse-susceptibility results for the alloy $Fe_{43}Ni_{37}Mn_{20}$ because the peak in the linear susceptibility χ is too diffuse.

We therefore carried out a more detailed study of the frequency dependence $T_f(\nu)$ in the alloy $Fe_{45}Ni_{35}Mn_{20}$. This dependence was found to be described well by the Vogel-Fulcher law[17]:

$$\frac{1}{\nu} = \tau = \tau_0 \exp \frac{E_a}{k_B(T_f - T_0)}, \qquad (1)$$

where E_a is the activation energy, τ_0 and T_0 are certain characteristic values of the time and temperature, and k_B is the Boltzmann constant.

The time constant $\tau_0 \approx 10^{-9}$ sec was determined in our earlier study[5] by analysis of the time dependence of the remanent magnetization of the alloy $Fe_{45}Ni_{35}Mn_{20}$. Figure 3 shows a plot of T_f versus $(\ln \nu_0 - \ln \nu)^{-1}$, from which we have determined that parameters T_0 and E_a/k_B. These two parameters were found to be significantly larger in $Fe_{45}Ni_{35}Mn_{20}$ ($T_0 = 80$ K, $E_a/k_B = 146$ K) than in "classical" spin glasses (in which $T_0 < 40$ K, $E_a/k_B \leqslant 80$ K).[17]

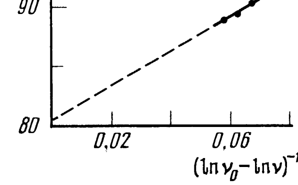

FIG. 3. Freezing temperature of the spin glass $Fe_{45}Ni_{35}Mn_{20}$ versus the quantity $(\ln \nu_0 - \ln \nu)^1$.

In accordance with the conclusions of theoretical studies,[18,19] this fact can be explained as follows. In the framework of the "cluster" model of the spin glass[18] the high value of T_0 in FeNiMn is indicative of a strong intercluster interaction in this spin glass in comparison with other spin glasses. Estimates made in Ref. 19 suggest that ferromagnetic correlations are dominant in the spin glass $Fe_{45}Ni_{35}Mn_{20}$. The large value of E_a/k_B means that at $x \sim x_c$ the clusters in FeNiMn alloys are larger than in impurity magnets of the AuFe type.[18]

4. FIELD DEPENDENCE OF T_f

To ascertain whether there is a phase transition at T_f it is of considerable interest to study the field dependence of the inverse susceptibility χ in the neighborhood of T_f. In a number of studies (see, e.g., Refs. 14 and 20) it is assumed that the magnetic-field (H) dependence of the height and position of the $\chi(T_f)$ peak can be described in the static scaling model with the aid of the power laws

$$[T_f(H)-T_f(0)]/T_f(0) \propto H^{(\gamma+\beta)^{-1}}, \quad (2)$$

$$\chi \propto H^{1/\delta-1}. \quad (3)$$

A dependence of the form (2) with $\gamma+\beta=3/2$ was obtained by de Almeida and Thouless.[21] It describes a "line instability" below which on the H–T diagram the solutions of the Sherrington-Kirkpatrick model are unstable.[12]

At the same time, the idea that spin glasses can be described by the N'eel theory of superparamagnetism (and that there is no phase transition at T_f) leads to[22]

$$T_f(H)/T_f(0) \propto (1-H/H_k)^2, \quad (4)$$

where H_k is the anisotropy field.

We made an experimental check of expressions (2)–(4) in alloys of the $Mn_{20}Fe_xNi_{80-x}$ system with $x=45$ and 48. It turns out that expressions (2) and (3) give a good description of the results of our experiments, whereas the dependence of $[T_f(H)/T_f(0)]^{1/2}$ on H is nonlinear (i.e., expression (4) is incorrect, see Fig. 4). For the alloy $Fe_{48}Ni_{32}Mn_{20}$ we obtain $(v+\beta)^{-1} \approx 0.3$, a value about half as large as the value obtained by de Almeida and Thouless.[21] This most likely means that the de Almeida-Thouless instability line should not be determined from the position of the peak on the temperature dependence of the inverse susceptibility. In the present theory of spin glasses there is a temperature T_i (in general different from T_f) below which macroscopic irreversible effects arise.[23] It is assumed that the $T_i(H)$ curve coincides with the de Almeida-Thouless line. The problem of making an unambiguous experimental determination of the line $T_i(H)$ has yet to be solved.

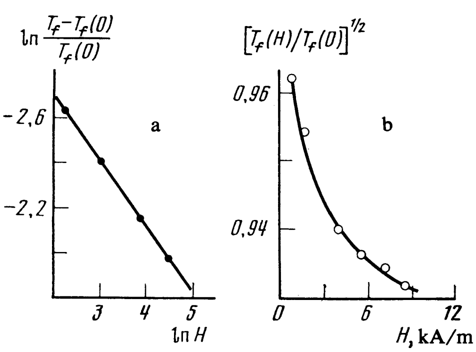

FIG. 4. a) Shift of the freezing temperature of the spin glass $Fe_{48}Mi_{32}Mn_{20}$ versus the bias field (in logarithmic coordinates); b) field dependence of the quantity $[T_f(H)/T_f(0)]_{1/2}$ for the alloy with $x=48$.

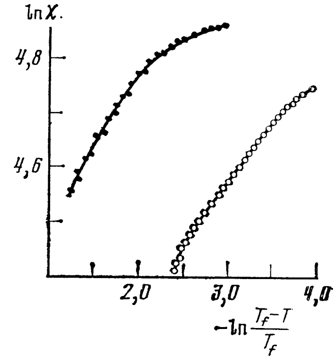

FIG. 5. Linear inverse susceptibility of alloys of the system $Mn_{20}Fe_xNi_{80-x}$ with $x=45$ (○) and $x=48$ (●) versus the reduced temperature $(T_f-T)/T_f$ (in logarithmic coordinates).

We4 have also made an attempt to verify the applicability of the power law

$$\chi \propto [(T_f-T)/T_f]^{-\gamma'} \quad (5)$$

for description of the behavior of the linear inverse susceptibility near the freezing temperature T_f. Figure 5 shows a plot of ln χ versus ln $[(T_f - T)/T_f]$ (for $T \lesssim T_f$) for alloys of the $Mn_{20}Fe_xNi_{80-x}$ system with $x=45$ and 48. It is seen that on each of the curves there is a rather large rectilinear segment. However this segment corresponds to an extremely small value of the critical exponent γ' (approximately 0.22 and 0.26 for the alloys with $x=45$ and $x=48$, respectively). On the whole, one can say that the critical exponents corresponding to the magnetic transformation at T_f differ strongly from the values corresponding to the phase transition at the Curie point in typical ferromagnets (e.g., in $Fe_{48}Ni_{32}Mn_{20}$ one has $\delta \approx 2.4$, $\beta \approx 3.0$, $\gamma' \approx 0.26$). We note that anomalously low values of the exponent γ' (of the order of 0.1–0.2) have also been obtained in a study of the behavior of the susceptibility of the spin glasses CuMn and GdAl.[24] Very high values of the exponent β (around 2) have been obtained[25] for the transformation at T_f in CuMn spin glasses.

For the investigated alloys of the $Mn_{20}Fe_xNi_{80-x}$ system the scaling relations between critical exponents[26] are not satisfied in the form of equalities. However the relations

$$\alpha'+2\beta+\gamma' \geqslant 2, \quad (6)$$

$$\alpha'+\beta(\delta+1) \geqslant 2 \quad (7)$$

do hold as inequalities. This indicates that the magnetic transformation at T_f is of a nonequilibrium character.

5. CONCLUSION

Our studies of the inverse susceptibility have refined the magnetic phase diagram of the system $Mn_{20}Fe_xNi_{80-x}$. We found that the shift of the freezing temperature T_f as a function of the magnetization-reversal frequency in alloys of this system can reach several degrees. We have shown that this shift is described well by the Vogel-Fulcher law, and we have determined the parameters T_0 and E_a/k_B characterizing the size and interaction of clusters in the spin glass.

We discovered that expression (4), which was obtained in the Néel superparamagnetism theory,[22] is unsuitable for explaining the field dependence of T_f. At the same time, the change in the position and height of the inverse-susceptibility peak at T_f and also the functional dependence $\chi(T)$ near T_f are described satisfactorily by power laws of the form in (2), (3). and (5). However, the values thus obtained for the critical exponents β, γ', and δ differ markedly from the values predicted by mean field theory or obtained in the fluctuation theory of phase transitions. In addition, the critical exponents do not obey the thermodynamic equalities which fellow from the scaling hypothesis. The anomalously large shifts of the freezing temperature of FeNiMn spin glasses are also difficult to explain if the transformation at T_f is treated as an equilibrium process.[5,16]

The features of the transition to the spin-glass state are obviously due to the nonergodicity of this state. In this case the usual methods of evaluating the thermodynamic quantities become unsuitable and only a time-dependent description of the system is possible.[27] The nature of the nonergodicity of the spin glass, as we know, is due to the presence of a great number of degenerate states separated by large energy barriers.

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