Double paramagnetic–ferromagnetic–spin-glass temperature transition in disordered alloys with competing exchange interaction

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The temperature dependences of the dynamic (linear and nonlinear) magnetic susceptibility and also of the spontaneous and residual magnetization of a number of alloys that undergo a paramagnetic–ferromagnetic–spin-glass temperature transition are studied. It is shown that in classical spin-glass systems (FeNiCr, PdFeMn, AuFe) such a transition is accompanied by the appearance of the asperomagnetic state. In nonclassical systems (FeNiMn, NiMn) the transition from the ferromagnetic phase to the spin-glass phase occurs directly, without the formation of asperomagnetism.

INTRODUCTION

The temperature transition from the ferromagnetic (FM) phase to the spin-glass (SG) phase was first predicted theoretically for the Ising model with an infinite-range interaction.[1] Analysis of the analogous problem for Heisenberg spins[2,3] led to the prediction of a new, asperomagnetic (ASM) state, which is the result of the evolution of a disordered collinear ferromagnet as it cools. This state is characterized by the presence of spontaneous magnetization (ferromagnetic ordering) in a certain direction and by spin-glass ordering of the transverse components of the spins. Upon further lowering of the temperature the system goes over from the ASM phase to a new, mixed phase, frequently identified with the SG phase, for which strong degeneracy and related macroscopically irreversible phenomena are typical.[3]

Thus, the problem of the experimental study of such systems can be divided into two parts. One must ascertain first whether in reality, at the lowest temperatures, the SG state is realized and the long-range ferromagnetic order (LFMO) is destroyed in the process, and secondly whether in reality an ASM state arises at temperatures below the Curie temperature T_C.

A considerable number of papers have been devoted to the experimental study of the first of these questions. Studies of elastic and inelastic scattering of neutrons deserve special attention, since they give direct information on processes occurring in the magnets. However, the experimental data are highly contradictory. For example, investigations of the temperature dependence of the maximum intensity of magnetic scattering of neutrons by the magnetic semiconductors EuSrS indicate that in the FM–SG transition the LFMO is destroyed,[4] while Murani,[5] carrying out similar investigations but measuring the integral intensity in the classical spin-glass system AuFe, concluded that the LFMO is preserved.

The situation is no better with the results of investigations of inelastic neutron scattering. For example, for amorphous FeNi alloys[6] and crystalline FeCr (Ref. 7) it has been shown that for low ($T \ll T_C$) temperatures the spin waves disappear. Obviously, this should be evidence for the destruction of the LFMO. On the other hand, in crystalline disordered NiMn alloys with a double FM–SG transition spin waves coexist with the SG down to zero absolute temperature.[8]

As regards the experimental observation of the ASM state appearing in the FM phase, the authors know of one investigation,[9] in which this state was established in the alloy AuFe by means of γ-resonance methods. It should be emphasized, however, that in analogous work by other authors,[10] carried out on the same alloys, the ASM state was not detected.

In Refs. 11 and 12, for the example of FeNiCr alloys, it was concluded from the results of an investigation of the dynamic magnetic susceptibility (DMS) that the LFMO is destroyed in the FM–SG transition. It was also postulated that in the temperature range $T_f < T < T_C$ (T_f is the spin-glass freezing temperature) an ASM state is realized. It is necessary, however, to have direct experimental data that make it possible to trace the evolution of the LFMO through the FM–SG transition.

With this pupose, in the present work we have studied not only the DMS of various ferromagnetic alloys with competing exchange interaction but also the temperature dependences of the spontaneous and residual magnetization. This gives grounds for drawing more-definite conclusions about the processes occurring in the ferromagnetic phase.

2. EXPERIMENTAL METHOD

The FeNiCr, NiMn, FeNiMn, and AuFe alloys were melted out in an induction furnace in an atmosphere of purified argon. The ingots were homogenized at a temperature of 1200 K for 24 hours. The investigated samples, in the form of cylinders with a height-to-diameter ratio equal to ~ 10, were quenched in water from 1200 K immediately before the measurements.

The dynamic magnetic susceptibility in the temperature range 1.5–300 K was measured on the apparatus described in Ref. 13. The static magnetization was measured by means of a vibromagnetometer in magnetic fields of up to 300 Oe, produced by a copper solenoid. In all cases the vertical component of the Earth's magnetic field was compensat-

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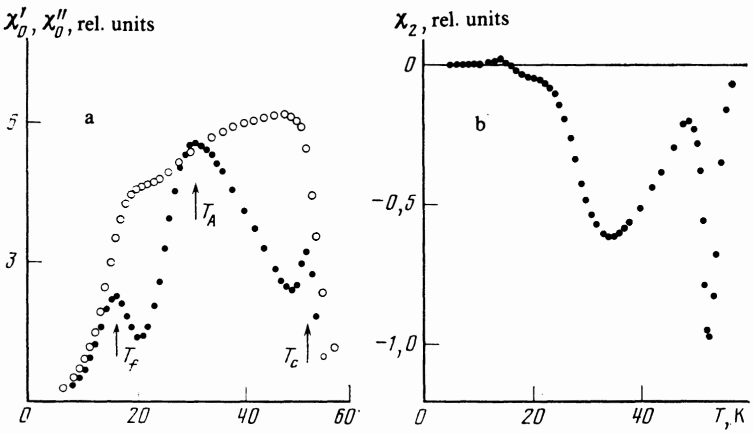

FIG. 1. Temperature dependences of a) the real part χ_0' (the light circles) and imaginary part χ_0'' (the dark circles) of the dynamic magnetic susceptibility, and b) the nonlinear susceptibility χ_2 of the alloy $Fe_{54}Ni_{26}Cr_{20}$. The measurement field has intensity 1 Oe and frequency 60 Hz.

ed with an accuracy of $\sim 10\%$.

3. EXPERIMENTAL RESULTS AND DISCUSSION

The alloy $Fe_{54}Ni_{26}Cr_{20}$. The dynamic magnetic susceptibility

In an alternating magnetic field $h = h_0\cos\omega t$ the dynamic magnetic susceptibility χ_0 of a magnet can be represented in the form

$$\chi_0(\omega)=\chi_0'(\omega)-i\chi_0''(\omega),$$

where the imaginary part χ_0'' of the susceptibility, being the Fourier transform of the two-spin correlation function, characterizes the dynamics of the magnetic system, and, consequently, should display anomalous behavior near points of magnetic phase transitions.[14] In particular, it may be expected that in the vicinity of the Curie temperature T_C of ferromagnetic alloys χ_0'' will have anomalies. Moreover, if the magnetic structure of the ferromagnetic phase itself undergoes changes, this should also be manifested in the temperature dependences of χ_0''. Another quantity that is extremely sensitive to processes occurring in magnets is the nonlinear magnetic susceptibility χ_2 (Ref. 15). Indeed, in Fig. 1, as an example, we give the temperature dependences of χ_0', χ_0'', and χ_2 of the alloy $Fe_{54}Ni_{26}CR_{20}$, which is near the critical concentration $C_0 = 25$at% for the appearance of LFMO (Ref. 16). The susceptibility χ_0' has two sharp anomalies, typical of systems undergoing a FM–SG transition. The high-temperature rise, as will be shown below, is connected with the onset of spontaneous magnetization in the alloy, while the sharp low-temperature fall is connected with the transition to the spin-glass state. The behavior of χ_0'' and χ_2 is more complicated. Near the Curie temperature $T_C = 52$ K one observes sharp extrema of χ_0'' and χ_2, reflecting the onset of LFMO in the alloy. At low temperatures ($T_f = 16$ K) the anomalies of χ_0'' and χ_2 are associated with the transition of the sample to the SG phase. In the intermediate region of temperatures, in the vicinity of $T_A = 31$ K, a further anomaly of χ_0'' and χ_2 is observed, whose causes we shall try to elucidate.

It might be supposed that the appearance of the intermediate anomaly is associated with features of the domain structure of the ferromagnetic alloy. But then this should inevitably be reflected in the behavior of χ_0'. However, as the experiment shows (Fig. 1), χ_0' has no noticeable features at the relevant temperature. In addition, the domain structure of the ferromagnet is very sensitive to magnetic annealing in the transition through T_C (Ref. 17). Therefore, two types of experiment were carried out. First, the alloy was cooled from a temperature $T > T_C$ to $T = 4.2$ K in an alternating magnetic field with $h_0 = 10$ Oe. Then the field amplitude was reduced to the appropriate measurement value ($h_0 \cong 1$ Oe), after which, as the sample warmed up, measurements of χ_0', χ_0'', and χ_2 were carried out. In the second experiment the alloy was cooled from $T > T_C$ to $T = 4.2$ K in zero field. After this an alternating magnetic field with $h_0 = 10$ Oe was applied and then reduced to the measurement value, and the temperature dependences of χ_0', χ_0'', and χ_2 were again recorded. Within the limits of the experimental error the two experiments gave the same results, given in Fig. 1. From this it can be concluded that the intermediate anomaly of χ_0'' and χ_2 is in no way connected with the domain structure of the sample. Moreover, it is unlikely that the dynamics of the domain walls played an important role at such low magnetization-reversal frequencies (10–100 Hz).

Unfortunately, the DMS data do not yield direct information about the evolution of the LFMO in the case under consideration. Considerably more information can be obtained from analysis of the experimental data on the temperature dependence of the spontaneous and residual magnetizations.

Temperature dependence of the spontaneous and residual magnetizations

Usually, the spontaneous magnetization I_s of ferromagnets is determined by the method of thermodynamic coefficients.[18] However, this method, which is justified for homogeneous ferromagnets near the Curie temperature, is unsuitable for the study of systems with competing exchange interaction. This is due to at least two circumstances. First, magnetization measurements are usually performed in saturating magnetic fields, which themselves destroy the SG state.[19] Secondly, in the SG phase, strong irreversible phenomena leading to a dependence of the magnetization on the measurement time (magnetic viscosity) manifest themselves. For these reasons, the authors of Ref. 20 proposed another method of measuring I_s—a method that is free from the above defects and consists in measuring the magnetiza-

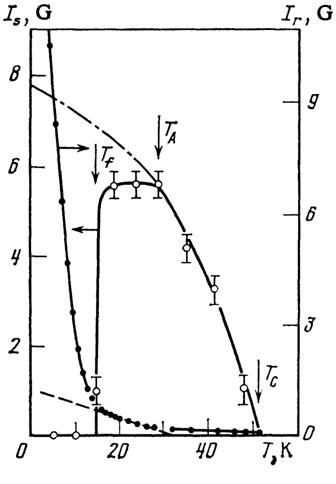

FIG. 2. Temperature dependences of the spontaneous magnetization I_s and residual magnetization i_r of the alloy $Fe_{54}Ni_{26}Cr_{20}$.

tion during cooling of the ferromagnet from temperatures $T > T_C$ to $T = 4.2$ K in magnetic fields that do not destroy the spin glass.

Using this technique we have measured the spontaneous magnetization I_s of the alloy $Fe_{54}Ni_{26}Cr_{20}$ (Fig. 2). In the same Figure we give data on the temperature dependence of the residual magnetization I_r of the same alloy, which were obtained after cooling of the alloy from temperatures $T > T_C$ to $T = 4.2$ K in a constant magnetic field 100 OE. It can be seen that spontaneous magnetization appears at the temperature $T_C = 52$ K. As the temperature is lowered, I_s increases, but in the vicinity of $T_A = 30$ K it deviates from the quasi-Brillouin dependence (the dashed-dotted curve) in the direction of lower values. Upon further cooling, near $T_f = 16$ K, the spontaneous magnetization disappears. This indicates unambiguously the complete destruction of the LFMO in the sample. It should be noted that the characteristic temperatures of the changes of I_s (Fig. 2) are in very good agreement with the temperatures at which the anomalies of χ_0'' and χ_2 are observed (Fig. 1).

We now discuss the results. In the transition from the paramagnetic (PM) phase at the temperature $T_C = 52$ K spontaneous magnetization (and, hence, long-range ferromagnetic order) arises in the sample. This also finds its reflection in the anomalies of χ_0', χ_0'', and χ_2 (Fig. 1). Upon further cooling, in the vicinity of $T_A = 30$ K, I_s decreases, and this can be related to the appearance of an ASM state in the alloy. Indeed, let the direction of the spontaneous-magnetization vector in the ferromagnetic state coincide with the z axis. As indicated above, in the transition to the ASM phase randomly frozen x and y components of the spontaneous magnetization appear. This is equivalent to an effective decrease of the z component of I_s. Precisely this situation is observed in reality, since below $T_A = 30$ K the spontaneous magnetization I_s does not increase but, within the limits of the experimental error, remains constant. Upon further decrease of the temperature, in the vicinity of $T = T_f = 16$ K, the spontaneous magnetization I_x becomes equal to zero and the alloy goes over into the spin-glass state. Strong degeneracy and the associated strong irreversibilities are characteristic for this state.[3] Data on the temperature dependence of the residual magnetization I_r (Fig. 2) can serve as confirmation of this. It can be seen that below $T_f = 16$ K the residual magnetization I_r depends very sharply on the temperature, and this is highly characteristic of systems in the spin-glass state.[20] We note in passing that, according to the theory of Ref. 21, weak irreversibilities arise at the temperature T_A of the onset of the ASM state, and this is also noticeable in the temperature dependence of I_r (the dashed curve).

PdFeMn and AuFe alloys

In Ref. 13 it was shown that in the system $Fe_C Ni_{80-C} Cr_{20}$, to which the alloy investigated above be-

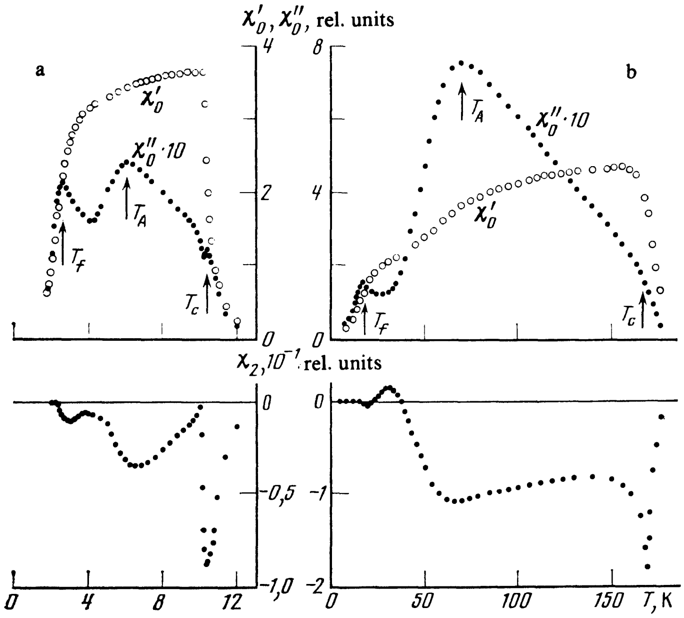

FIG. 3. Temperature dependences of the real part χ_0' and imaginary part χ_0'' and imaginary part χ_0'' of the dynamic magnetic susceptibility, and nonlinear susceptibility χ_2, of the alloys $(Pd_{99.65}Fe_{0.35})_{95.0}Mn_{5.0}$ (a) and $Au_{81}F_{19}$ (b). The measurement field has frequency 60 Hz and intensity 1 Oe (a) and 3 Oe (b).

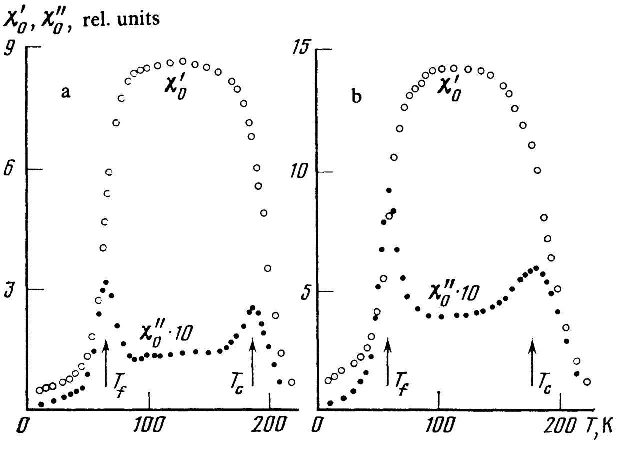

Fig. 4. Temperature dependences of the real part χ_0' and imaginary part χ_0'' of the dynamic magnetic susceptibility of the alloys $Fe_{52.5}Ni_{32.5}Mn_{15.0}$ (a) and $Ni_{77.9}Mn_{22.1}$ (b). The measurement field has intensity 1 Oe and frequency 36 Hz.

longs, in the region of compositions $C = 56$–60 at% the alloys experience a true (Edwards-Anderson) PM–SG phase transition. Such systems may be called classical spin glasses. PdFeMn and AuFe alloys are such systems. It is therefore of interest to investigate their dynamic magnetic susceptibilities, since in this case too a PM–SG temperature transition occurs. From Fig. 3, in which the temperature dependences of χ_0', χ_0'', and χ_2 for the alloys $(Pd_{99.65}Fe_{0.35})_{95.0}Mn_{5.0}$ and $Au_{81}Fe_{19}$ are given, it can be seen that all the characteristic features of the DMS are preserved. Therefore, one can conclude that for the given alloys the process of establishment of the ground magnetic state, as in the FeNiCr alloy considered above, follows the scheme PM–FM–ASM–SG.

FeNiMn and NiMn alloys

A completely different picture is observed, e.g., in FeNiMn and NiMn alloys (Fig. 4). On the temperature dependences of χ_0'' there are only two anomalies, near T_C and T_f. This indicates that in these systems the *SG* state arises directly from the FM phase. This conclusion is confirmed also by the results of an investigation of inelastic neutron scattering in NiMn alloys.[8]

It is important to note that we have carried out special investigations of the *DMS* of alloys of other compositions ($Fe_{58.5}Ni_{26.5}Mn_{15.0}$ and $Ni_{75}Mn_{25}$) that undergo a PM–SG transition. Processing of the results by the method described in Ref. 13 makes it possible to conclude that in the latter case the process of establishment of the ground magnetic state (the spin glass) is not the result of a phase transition but rather is associated with processes of gradual freezing of the magnetic moments in a wide range of temperatures. Such alloys may be called nonclassical spin glasses.

From this we can conclude that in systems in which the PM–SG transition is not a true phase transition the process of formation of the ground magnetic state in the ferromagnetic region follows the scheme PM–FM–SG. In other words, in the FM phase an ASM state does not arise. In conclusion, we note that the reasons for the difference in the evolution of the LFMO in the PM–FM–SG transition in classical and nonclassical systems are not clear at present.

4. CONCLUSION

On the basis of the experimental results presented in the paper we can conclude that in classical spin-glass systems (FeNiCr, PdFeMn, AuFe) the following hierarchy of magnetic states arises in the PM–FM–SG temperature transition: At the curie point T_C long-range ferromagnetic order appears, and down to the temperature T_A the magnetic structure of such alloys is that of a disordered collinear ferromagnet. In the temperature range $T_f < T < T_A$ an ASM state, characterized by the appearance of randomly frozen magnetic moments perpendicular to the spontaneous-magnetization vector, is characteristic for the alloys. Upon further cooling the ASM is transformed into the degenerate-SG phase.

In nonclassical spin-glass systems (FeNiMn, NiMn) an ASM phase is not observed, and the degenerate-SG phase arises directly from the FM phase.

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