

# Reentrant effects in Heisenberg antiferromagnetic alloys of fcc iron

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Reentrant transitions induced by temperature changes in the Heisenberg frustrated antiferromagnetic alloys  $\text{Fe}_x\text{Ni}_{80-x}\text{Cr}_{20}$  and  $\text{Fe}_x\text{Ni}_{80-x}\text{Mn}_{20}$  have been studied by magnetic methods, neutron diffraction, and  $\mu^+$  spin depolarization. As alloys with dopant concentrations close to the critical value for the appearance of a long-range antiferromagnetic order are cooled, the following sequence of magnetic phases and states occurs: paramagnet, collinear antiferromagnet, noncollinear antiferromagnet (antiasperomagnet), reentrant spin glass. In the reentrant spin glass phase there is no long-range antiferromagnetic order, but there are finite-radius antiasperomagnetic correlations.

## 1. INTRODUCTION

The problem of reentrant temperature-induced transitions from magnetically ordered (ferromagnetic and antiferromagnetic) states to a spin-glass (SG) state occupies a special place in the physics of SGs. The research on reentrant transitions in frustrated antiferromagnets (AFMs) is still in its infancy, in contrast with the research on ferromagnetic systems. A theory for these transitions has been derived for Ising<sup>1,2</sup> and Heisenberg<sup>3</sup> AFMs on the basis of the molecular-field approximation. It was concluded in those studies that in a zero magnetic field the primary magnetic state (at 0 K) is, for both Ising and Heisenberg AFMs with a competing exchange, the state of a so-called antiferromagnetic (AFM) SG, characterized by the coexistence of a long-range AFM order (LRAFMO) and a degenerate SG.

Most of the experimental work on reentrant AFM-SG transitions has been carried out on Ising systems. In particular, it has been shown by neutron diffraction that the transition to the state of a reentrant SG is not accompanied by a destruction of the LRAFMO in AFMs of this type [ $\text{Fe}_x\text{Mg}_{1-x}\text{Cl}_2$  (Ref. 4) and  $\text{Fe}_x\text{Mn}_{1-x}\text{TiO}_3$  (Ref. 5)], in agreement with the theory of Refs. 1 and 2.

There has been no systematic experimental work on Heisenberg frustrated AFMs, in contrast with Heisenberg systems. In particular, there are the unresolved questions of whether reentrant AFM-SG transitions occur in these systems, whether the LRAFMO is conserved as a result of such transitions, and whether the noncollinear AFM (antiasperomagnetic) state predicted in Ref. 6 arises.

In the present study we have undertaken an effort to find answers to these questions. Finding these answers will make it possible to draw general conclusions regarding the processes by which magnetic phases and states form in magnetically ordered frustrated Heisenberg systems.

## 2. EXPERIMENTAL PROCEDURE

For the experiments we selected the fcc alloys  $\text{Fe}_x\text{Ni}_{80-x}\text{Cr}_{20}$  ( $64 \leq X \leq 68$  at.%) and  $\text{Fe}_x\text{Ni}_{80-x}\text{Mn}_{20}$  ( $X = 52$  at.%), in which an AFM ordering<sup>7,8</sup> of the first

kind<sup>9</sup> occurs during cooling. According to Ref. 10, the exchange energy in these alloys is much larger than the anisotropy energy; i.e., these alloys are Heisenberg AFMs. The particular compositions of the alloys were chosen near corresponding critical concentrations for the appearance of an LRAFMO on the magnetic phase diagrams. Accordingly, the alloys have a substantial random exchange, satisfying a necessary condition for the occurrence of reentrant AFM-SG transitions in them.

The static susceptibility and the magnetization were measured on a vibrating-sample magnetometer. The real ( $\chi'_0$ ) and imaginary ( $\chi''_0$ ) components of the dynamic (ac) magnetic susceptibility were studied with the help of a mutual-induction bridge.

The local magnetic field distribution was studied by  $\mu^+$  spin depolarization on the apparatus described in Ref. 11. The temporal spectrum  $N(t)$  of positrons  $e^+$  from the decay  $\mu^+ \rightarrow e^+ + \nu + \bar{\nu}$  was measured:

$$N(t) = (N_0/N_\mu) \{ [1 + a_0 G(t)] \exp(-t/\tau_\mu) + N_\phi/N_0 \}. \quad (1)$$

Here the function  $G(t)$  characterizes the statics and dynamics of the local magnetic fields in the test sample,  $N_\mu$  is the number of muons stopped in the target,  $\tau_\mu$  is the muon lifetime, and  $N_0/N_\mu = 0.075$ ,  $a_0 = 0.3$ , and  $N_\phi/N_0 = 0.001$  are constants of the experimental apparatus and the muon beam.

The neutron scattering was studied on polycrystalline samples with a high-resolution neutron diffractometer. A profile analysis of the (110) AFM superstructural reflections was carried out.

## 3. EXPERIMENTAL RESULTS AND DISCUSSION

### 3.1. Temperature dependence of the static susceptibility

To determine whether a reentrant AFM-SG transition can be induced by a temperature change in Heisenberg AFM alloys, we can make use of one of the basic properties of a SG: the appearance of irreversible effects below its freezing point  $T_f$ . In SGs one observes a thermomagnetic "past history;"<sup>12</sup>

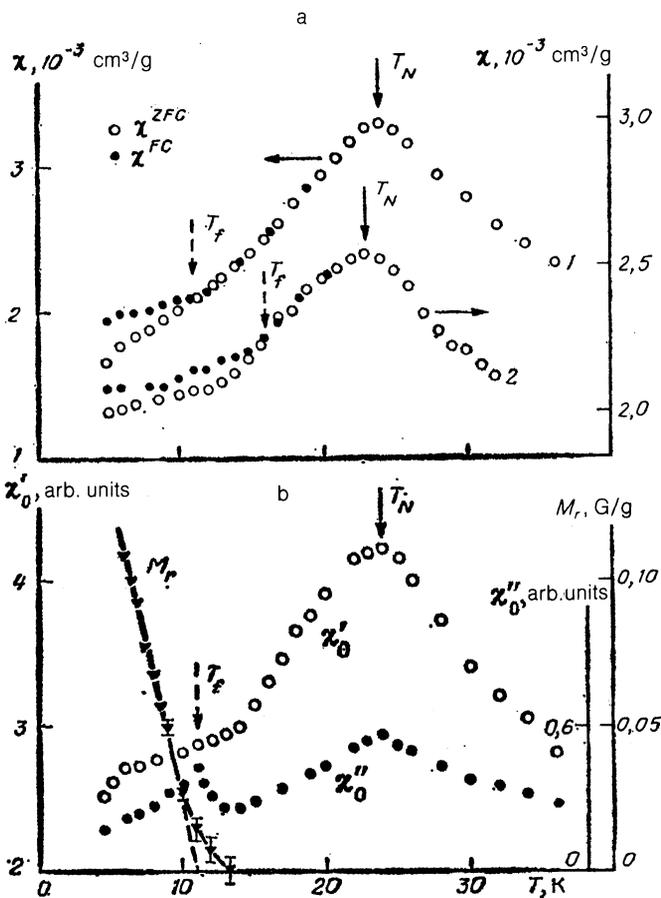


FIG. 1. Temperature dependence of certain properties of the alloy  $\text{Fe}_{64}\text{Ni}_{16}\text{Cr}_{20}$ . a: The susceptibilities  $\chi^{\text{ZFC}}$  and  $\chi^{\text{FC}}$ . 1—In a magnetic field of 0.1 kOe; 2—2.0 kOe. b: The thermoremanent magnetization  $M_r$  and the real  $\chi'_0$  and imaginary  $\chi''_0$  components of the ac susceptibility. The ac susceptibility was measured in a magnetization-reversal field of 10 Oe at a frequency of 270 Hz.

i.e., after cooling below  $T_f$  in a magnetic field, the static susceptibility  $\chi^{\text{FC}}$  of the SG is higher than the susceptibility  $\chi^{\text{ZFC}}$  after the sample is cooled in a zero magnetic field. The temperature at which  $\chi^{\text{FC}}$  becomes equal to  $\chi^{\text{ZFC}}$  is usually identified as the temperature of the transition to the SG phase.<sup>13</sup>

Figure 1a shows representative results on the temperature dependence of  $\chi^{\text{ZFC}}$  (open circles) and  $\chi^{\text{FC}}$  (filled circles) of the alloy  $\text{Fe}_{64}\text{Ni}_{16}\text{Cr}_{20}$ . These results were obtained in magnetic fields of (curves 1) 0.1 kOe and (curves 2) 2.0 kOe. It can be seen from these results that, for example, an irreversibility of the susceptibility arises in a field  $H = 0.1$  kOe below the temperature  $T = 11$  K (marked by the dashed arrow). In accordance with the discussion above, the temperature can be identified as the freezing point  $T_f$  of the SG in the given magnetic field  $H = 0.1$  kOe. It also follows from Fig. 1a that  $T_f$  increases with an increase in the magnetic field applied to the sample (curves 2). We need to stress that this dependence is qualitatively the same as the theoretical prediction of Refs. 1 and 2 and fundamentally different from the  $T_f(H)$  dependence for systems with paramagnet-SG (PM-SG) and ferromagnet-SG (FM-SG) transitions. The condition  $dT_f(H)/dH < 0$  always holds in the latter two cases.<sup>13,14</sup>

With increasing magnetic field, the Néel point  $T_N$  decreases anomalously rapidly, again in agreement with the calculations of Refs. 1 and 2.

### 3.2. $H$ - $T$ phase diagrams

Working from the results on the temperature dependence of  $\chi^{\text{ZFC}}/\chi^{\text{FC}}$ , one can construct  $H$ - $T$  phase diagrams of AFM alloys  $\text{Fe}_x\text{Ni}_{80-x}\text{Cr}_{20}$  of various compositions (Fig. 2). We first consider the alloys with  $X = 64$  and  $X = 66$  at.%. It can be seen from Fig. 2, a and b, that a double transition, PM-AFM-SG, does indeed occur in these alloys in a zero magnetic field. It also follows from this transition that the temperature region in which the SG exists (3) becomes broader, while that of the AFM phase (2) becomes narrower, with increasing magnetic field. Above a certain critical field  $H_c$  ( $H_c \approx 5$  kOe for the alloy with  $X = 64$ , and  $H_c \approx 11$  kOe for  $X = 66$ ), only the PM-SG (1-3) transitions occur. The derivative  $dT_f(H)/dH$  becomes negative, as in systems with PM-SG transitions.<sup>13,14</sup>

According to neutron diffraction data,<sup>7</sup> the critical concentration ( $X_0$ ) for the appearance of a LRAFMO in  $\text{Fe}_x\text{Ni}_{80-x}\text{Cr}_{20}$  alloys is in the interval 62–63 at.%. In the alloys which we just discussed, the iron concentration is just barely above  $X_0$ . Random exchange thus plays an important role in those alloys. There is accordingly interest in determining whether a PM-AFM-SG transition occurs in  $\text{Fe}_x\text{Ni}_{80-x}\text{Cr}_{20}$  alloys in which the contribution of random exchange to the total exchange energy is smaller. Figure 2c shows the  $H$ - $T$  phase diagram for the alloy with  $X = 68$ . We see that in a zero magnetic field we have  $H_0 \approx 5$  kOe  $< H < H_c \approx 17$  kOe, and a PM-AFM-SG transition occurs, as in the cases discussed above. Clearly, when the contribution of the AFM interaction is even larger (when  $X$  is even larger), and there is a simultaneous decrease in the relative importance of random exchange, the fields  $H_0$  and  $H_c$  will increase. In the limit of a "pure" AFM, the SG region (3) will disappear. We might add that the absolute value  $|dT_N(H)/dT|$  should decrease in this case, as can be seen clearly from a comparison of Figs. 2b and 2c.

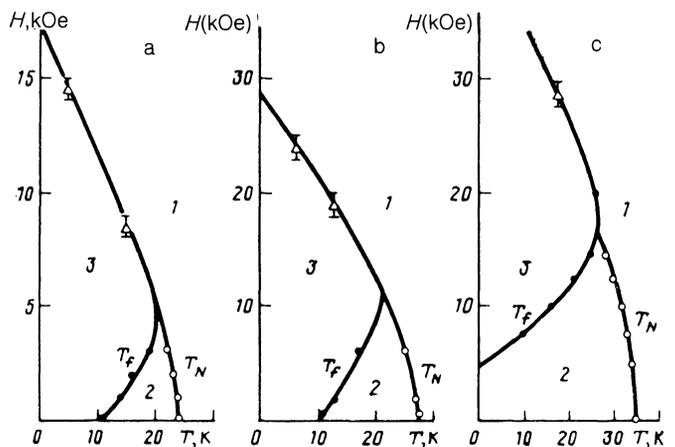


FIG. 2.  $H$ - $T$  phase diagrams of  $\text{Fe}_x\text{Ni}_{80-x}\text{Cr}_{20}$  alloys. a:  $X = 64$  at.%. b: 66 at.%. c: 68 at.%. 1—Paramagnet; 2—antiferromagnetic region; 3—spin-glass region.

### 3.3. Alternating-current susceptibility

The conclusion, reached above, that PM–AFM–SG transitions are induced by temperature changes in the alloys  $\text{Fe}_x\text{Ni}_{80-x}\text{Cr}_{20}$  was based on measurements of  $\chi^{\text{ZFC}}/\chi^{\text{FC}}$ . For SGs, measurements of the ac susceptibility are customary.

Figure 1b shows the temperature dependence of the real ( $\chi'_0$ ) and imaginary ( $\chi''_0$ ) components of the ac susceptibility of the alloy  $\text{Fe}_{64}\text{Ni}_{16}\text{Cr}_{20}$ . We see that  $\chi'_0(T)$  has a maximum at the Néel point  $T_N = 24$  K and an inflection-point anomaly at  $T_f = 11$  K. Qualitatively the same behavior has been predicted theoretically for systems with PM–AFM–SG transitions.<sup>1–3</sup>

To detect magnetic phase transitions in the magnetically ordered phase, it is far more convenient to study the imaginary component of the ac susceptibility. Being the Fourier transform of the two-spin correlation function, this component describes the dynamics of magnetic systems. In Fig. 1b we see a peak in  $\chi''_0(T)$  at  $T_f$  in the alloy  $\text{Fe}_{64}\text{Ni}_{16}\text{Cr}_{20}$ . This peak is evidence of the development of critical fluctuations in the course of an AFM–SG transition. It is important to note that for the alloys  $\text{Fe}_{64}\text{Ni}_{16}\text{Cr}_{20}$  and  $\text{Fe}_{66}\text{Ni}_{14}\text{Cr}_{20}$  the temperature  $T_f$  found from the  $\chi''_0(T)$  curves agrees well with the values found from the  $\chi^{\text{ZFC}}/\chi^{\text{FC}}$  curves in minimal magnetic fields. On the other hand, there are no anomalies on the temperature dependence  $\chi''_0(T)$  of the alloy  $\text{Fe}_{68}\text{Ni}_{12}\text{Cr}_{20}$ , according to Fig. 2c. This result confirms that a reentrant AFM–SG transition does not occur in this alloy in a weak magnetic field ( $\sim 10$  Oe).

It can also be seen from Fig. 1b that  $\chi''_0(T)$  has an anomaly near  $T_N$ . This effect is observed only in alloys which undergo PM–AFM–SG transitions in a zero magnetic field. For the alloy  $\text{Fe}_{68}\text{Ni}_{12}\text{Cr}_{20}$ , which has only a PM–AFM transition at  $H = 0$ , there is no anomaly on the  $\chi''_0(T)$  curve near  $T_N$ . The reason for this effect is not completely clear at this point.

### 3.4. Thermoremanent magnetization

One more piece of proof (in addition to the results presented above) of the occurrence of a reentrant SG in these alloys comes from data on their thermoremanent magnetization  $M_r$ . Figure 1b illustrates the situation with the temperature dependence  $M_r(T)$  of the alloy  $\text{Fe}_{64}\text{Ni}_{16}\text{Cr}_{20}$  found after this alloy was cooled from  $T > T_N$  to  $T = 4.5$  K  $< T_f$  in a magnetic field of 200 Oe. As expected,  $M_r(T)$  increases sharply below  $T_f = 11$  K. This increase is evidence of the appearance of a frozen SG state in the alloy at  $T < T_f$ . An extremely important result is that the remanent magnetization remains finite even in the AFM phase, vanishing only at a temperature  $T_f < T \approx 14$  K  $< T_N$ . This result is due to the appearance of a noncollinear AFM state (antiperomagnetic state) in this alloy. We postpone a discussion of this state to Subsection 3.6.

### 3.5. Isothermal magnetization

Figure 3 shows, as an example, the field dependence of the magnetization  $M$  of the alloy  $\text{Fe}_{64}\text{Ni}_{16}\text{Cr}_{20}$  at temperatures  $T < T_N$ . A hysteresis arises on the  $M(H)$  curves in magnetic fields  $H < H_p(T)$ . The corresponding values of

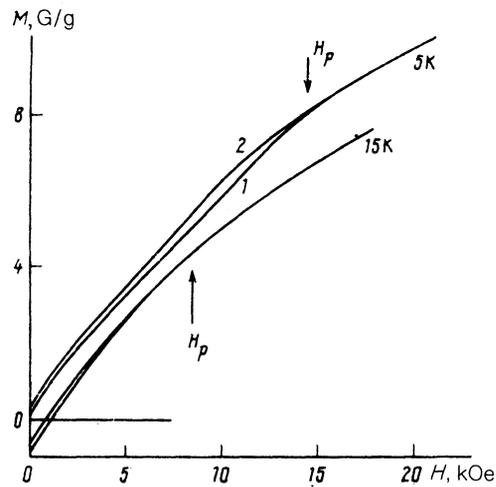


FIG. 3. Field dependence of the magnetization  $M$  of the alloy  $\text{Fe}_{64}\text{Ni}_{16}\text{Cr}_{20}$  at various temperatures.

$H_p(H)$  for the various  $\text{Fe}_x\text{Ni}_{80-x}\text{Cr}_{20}$  alloys are shown on the  $H$ – $T$  phase diagrams (the triangles in Fig. 2). It can be seen from the  $H$ – $T$  phase diagrams that the experimental points found in this manner lie on the  $T_f(H)$  phase line constructed from the  $\chi^{\text{ZFC}}/\chi^{\text{FC}}$  measurements, and these experimental points correspond to a PM–SG transition. There is nothing surprising here, since the SG phase should be characterized by not only a thermal hysteresis but also a field hysteresis of the static magnetization.<sup>15</sup>

Let us examine the features in the magnetization reversal of the alloy  $\text{Fe}_{64}\text{Ni}_{16}\text{Cr}_{20}$  at a temperature  $T = 5$  K  $< T_f$  ( $H = 0$ ) (Fig. 3). The experimental results show that, after the magnetizing field is reduced from  $H > H_p$  to zero, a hold of the alloy at 5 K for a certain time ( $\sim 600$  s) has the consequence that a repeated magnetization corresponds to an  $M(H)$  curve (not shown in Fig. 3) which lies between curves 1 and 2. This fact is unambiguous evidence that the magnetization curves for this alloy at  $T = 5$  K in magnetic fields  $H < H_p$  correspond to a metastable SG state which is undergoing a slow relaxation to equilibrium.

### 3.6. $\mu^+$ spin relaxation

It was shown in the preceding sections of this paper, on the basis of results obtained by magnetic measurements, that a double PM–AFM–SG transition can be induced by temperature changes in Heisenberg AFMs. On the other hand, it has been shown in several papers that, as frustrated ferromagnets are cooled, the appearance of a reentrant SG phase is preceded by the formation of a noncollinear FM state (an asperomagnetic state), which may be thought of as a superposition of an FM ordering of spins along a selected direction and a SG order of their transverse components.<sup>16–18</sup>

We would like to know whether a noncollinear AFM (antiperomagnetic) state can form in a Heisenberg frustrated AFM.

Unfortunately, the results of the magnetic measurements do not support unambiguous conclusions regarding the magnetic structure of the phases and states which arise in these systems in the course of reentrant PM–AFM–SG tran-

sitions. Information of this sort is provided by the results on the  $\mu^+$  spin depolarization, discussed below. These results make it possible to draw conclusions regarding the distribution of local magnetic fields in a test sample.

We studied the alloy  $\text{Fe}_{64}\text{Ni}_{16}\text{Cr}_{20}$ . To analyze the experimental data, we chose the function  $G(t)$  in (1) in the form

$$G(t) = G_d(t)G_s(t), \quad (2)$$

where  $G_d(t)$  and  $G_s(t)$  characterize the dynamics and statics, respectively, of the local magnetic fields. The choice of the functions  $G_d(t)$  and  $G_s(t)$  in the form<sup>19</sup>

$$G_d(t) = \exp(-\lambda t) \quad (3)$$

and

$$G_s(t) = \sum_i \frac{a_i}{a_0} \left\{ \frac{1}{3} + \frac{2}{3} \left[ \cos \gamma_\mu B_i t - \frac{(\gamma_\mu \Delta_i t)^\alpha}{\gamma_\mu B_i t} \sin(\gamma_\mu B_i t) \right] \right. \\ \left. \times \exp \left[ -\frac{(\gamma_\mu \Delta_i t)^\alpha}{\alpha} \right] \right\} \quad (4)$$

resulted in the best description of the experimental data on the  $\text{Fe}_{64}\text{Ni}_{16}\text{Cr}_{20}$  sample at various temperatures. In (3) and (4),  $\lambda$  is the rate of the muon spin depolarization,  $\gamma_\mu$  is the gyromagnetic ratio of the muon,  $B_i$  is the mean value of the local magnetic field,  $\Delta_i$  is a measure of the dispersion of the local magnetic field, and  $a_i$  is the volume fraction of magnetic phase  $i$  which arises in the sample as it is cooled.

Equation (4) is valid for a Heisenberg magnetic material without a magnetic texture.<sup>19</sup> The parameter value  $\alpha = 1$  corresponds to a Lorentz distribution, and  $\alpha = 2$  to a Gaussian distribution, of the local magnetic fields. In (4), the following relations are assumed for the cases of a collinear magnetic material, a noncollinear magnetic material (an antiasperomagnet), and a spin glass, respectively:

$$\left. \begin{array}{l} \text{collinear magnet} \\ \text{noncollinear magnet (asperomagnet)} \\ \text{spin glass} \end{array} \right\} \begin{array}{l} B_i \gg \Delta_i \approx 0, \\ B_i \approx \Delta_i, \\ \Delta_i \gg B_i \approx 0. \end{array} \quad (5)$$

The experimental data were analyzed by the method of least squares using Eqs. (1)–(4) with conditions (5). The results confirm the presence of a reentrant PM–AFM–SG transition in the alloy  $\text{Fe}_{54}\text{Ni}_{16}\text{Cr}_{20}$ . The results also reveal features of the magnetic states which are realized in this alloy at the various temperatures.

Figure 4 shows the temperature dependence  $\lambda(T)$ . We see that near  $T_N$  and  $T_f$  (the phase-transition temperatures determined above by magnetic methods) there are some clearly defined peaks in the relaxation rate of the muon spin. According to Ref. 11, these peaks are evidence of the development of critical fluctuations near the corresponding temperatures.

Analysis of the distributions of local magnetic fields shows that different types of magnetic states are realized in different temperature intervals in the alloy  $\text{Fe}_{64}\text{Ni}_{16}\text{Cr}_{20}$  (Fig. 5). In particular, at temperatures  $T_A \approx 17\text{K} < T < T_N = 24\text{K}$  this alloy is a collinear AFM with a Lorentz distribution of local magnetic fields. At temperatures  $T_f = 11\text{K} < T < T_A = 17\text{K}$  the magnetic struc-

ture of this alloy may be thought of as a superposition of a collinear AFM and a noncollinear AFM (antiasperomagnet), with the same distribution of local magnetic fields. Finally, at  $T < T_f = 11\text{K}$  the alloy goes into a reentrant SG phase, in which SG and antiasperomagnetic states coexist with a Gaussian distribution of local magnetic fields.

It thus follows from the results presented in this section of the paper that the appearance of a reentrant-SG phase is preceded by the formation of a nonlinear AFM (antiasperomagnetic) state in the course of reentrant temperature-induced transitions in Heisenberg frustrated AFMs. Confirmation of this conclusion follows from an analysis of results on the unidirectional anisotropy, to which we now turn.

### 3.7. Unidirectional anisotropy

The Dzyaloshinskii–Moriya anisotropy plays an important role in metal systems. This anisotropy stems from the spin-orbit interaction of a pair of spins  $\mathbf{S}_1$  and  $\mathbf{S}_2$  with coordinates  $\mathbf{R}_1$  and  $\mathbf{R}_2$  through a third atom, at the origin of coordinates.<sup>20</sup> The energy of this interaction can be written in the form

$$H_{DM} = -\mathbf{D}(\mathbf{R}_1, \mathbf{R}_2) [\mathbf{S}_1 \mathbf{S}_2], \quad (6)$$

where  $\mathbf{D}(\mathbf{R}_1, \mathbf{R}_2)$  is a constant.

Interaction (6) gives rise to a macroscopic anisotropy, which is unidirectional and which is manifested by a displacement of the magnetization-reversal loop with respect to the origin of coordinates (toward negative magnetic fields) after the system is cooled in a magnetic field. It can be seen from (6) that the Dzyaloshinskii–Moriya anisotropy does not arise in a collinear magnetic material. It should be seen only in magnetic structures in which the spins are not parallel to each other, i.e., in an antiasperomagnetic state and in a spin-glass state.

The experiments carried out here show that after the alloy  $\text{Fe}_{64}\text{Ni}_{16}\text{Cr}_{20}$  is cooled from  $T = 30\text{K}$  to  $T = 4.6\text{K}$  in a magnetic field of 200 Oe the magnetization-reversal loop is

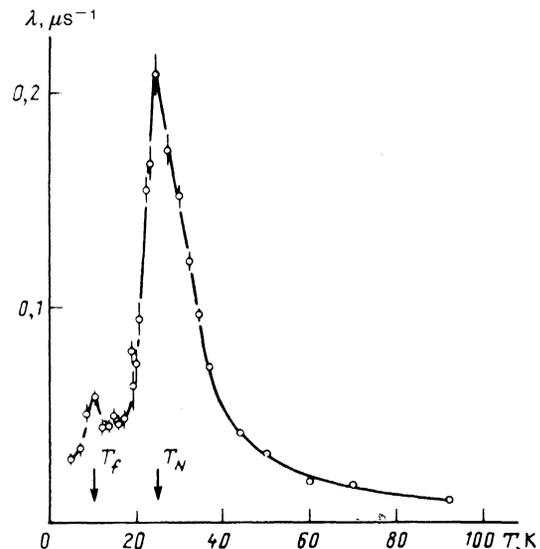


FIG. 4. Temperature dependence of the depolarization rate  $\lambda$  of the  $\mu^+$  spin for the alloy  $\text{Fe}_{64}\text{Ni}_{16}\text{Cr}_{20}$ .

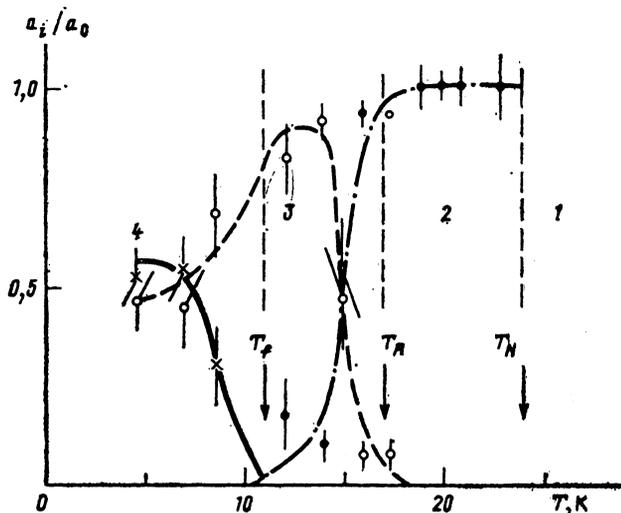


FIG. 5. Volume fraction ( $a_i/a_0$ ) of the magnetic phases which arise in the alloy  $\text{Fe}_{64}\text{Ni}_{16}\text{Cr}_{20}$  at various temperatures. 1—Paramagnet; 2—collinear antiferromagnet; 3—noncollinear antiferromagnet (antiasperomagnet); 4—spin glass. Here  $T_N$ ,  $T_A$ , and  $T_f$  are respectively the Néel point, the temperature at which the antiasperomagnetic state appears, and the state at which the reentrant spin glass appears.

shifted an amount  $\delta H = 12$  Oe in the negative-field direction. In other words, there is a unidirectional anisotropy at  $T < T_f$  for this alloy. According to the discussion above, this result means that a noncollinear magnetic state—in the case at hand, an SG state—is realized in this alloy at this temperature.

An extremely important point is that in the temperature interval  $T_f < T \leq 17$  K we again observe a displacement of the magnetization-reversal loop. The shift  $\delta H$  decreases with increasing experimental temperature. Finally, at temperatures  $17 \text{ K} < T < T_N$  the shift vanishes:  $\delta H = 0$ . These results confirm that a collinear AFM state  $\{[S_1 S_2] = 0$  in Eq. (6) $\}$  forms in the alloy in the temperature interval  $17 \text{ K} < T < T_N$ , while an antiasperomagnetic state ( $[S_1 S_2] \neq 0$ ) forms at temperatures  $T_f < T < 17$  K.

### 3.8. Neutron scattering

As we mentioned in the Introduction, it has been concluded in most of the previous theoretical and experimental studies that a long-range antiferromagnetic order (LRAFMO) coexists with a spin glass (SG) in the magnetic ground state in frustrated antiferromagnets in a zero magnetic field. However, one can cite arguments to support the suggestion that an LRAFMO should be disrupted in Heisenberg AFMs in the course of reentrant AFM-SG transitions. Specifically, according to Refs. 21 and 22 a “freezing” of the frustrated spins in a magnetically ordered matrix gives rise to random magnetic fields at the sites of the surrounding ordered matrix. As a result, the lower critical dimensionality of the magnetic material doubles. Consequently, in that interpretation the LRAFMO should not be disrupted in three-dimensional Ising AFMs upon the transition to a reentrant-SG state. This conclusion agrees with what is actually observed.<sup>4,5</sup> In Heisenberg systems in the reentrant-SG phase, in contrast, there should be no LRAFMO.

The alloys  $\text{Fe}_x\text{Ni}_{80-x}\text{Cr}_{20}$  are ill-suited for an experimental resolution of this question by neutron diffraction, because the AFM reflections on the neutron diffraction patterns are very faint. More convenient are the alloys  $\text{Fe}_x\text{Ni}_{80-x}\text{Mn}_{20}$ , which have a higher Néel point and a more intense (110) AFM reflection.<sup>8</sup>

For the present study we selected the alloy  $\text{Fe}_{52}\text{Ni}_{28}\text{Mn}_{20}$ , which has a Néel point  $T_N = 115$  K and which goes into a reentrant-SG state at  $T_f = 55$  K (Ref. 23).

A (110) superstructural reflection is observed on the neutron diffraction pattern of this alloy below  $\sim 120$  K. The temperature dependence of the integral intensity of this reflection is shown in Fig. 6a. It can be seen from these results that below  $T_N = 120$  K an AFM order is established in the alloy, and the Néel point agrees with the values found from magnetic measurements.<sup>23</sup>

The profile of the (110) AFM reflection at  $T < T_f = 60$  K is Lorentzian (we are allowing for the instrumental resolution). An attempt was made to describe the profile of this reflection as a sum of two components: a Gaussian line with the instrumental width and a Lorentzian line convolved with the first component. However, it was not found possible to perform this separation, because of the inadequate statistical accuracy.

Figure 6b shows the temperature dependence of the half-width (the width at half-maximum) of the (110) AFM reflection. We see that the reflection is broadened at temperatures  $T < T_f$ , and the broadening increases as the alloy is cooled. The horizontal line in Fig. 6b corresponds to the instrumental half-width (an error  $\pm 0.001^\circ$ ). The latter was found from measurements of the (220) nuclear reflection, which arises in place of the magnetic reflection in the absence of the filter which prunes the contribution to the scattering from half-wavelength neutrons.

The broadening of the magnetic reflection below  $T_f$  is

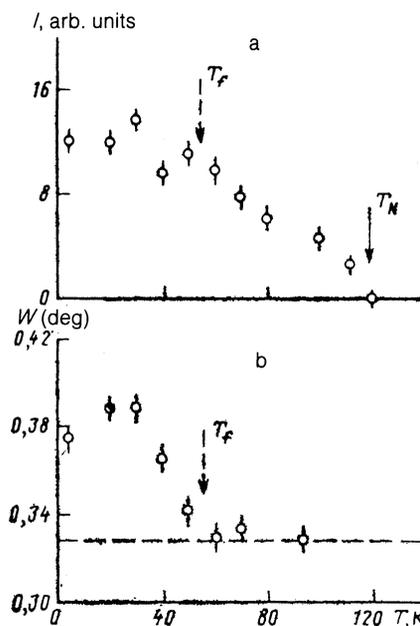


FIG. 6. Temperature dependence of (a) the integral intensity  $I$  of the (110) antiferromagnetic reflection and (b) the half-width  $W$  of this reflection for the alloy  $\text{Fe}_{52}\text{Ni}_{28}\text{Mn}_{20}$ .

unambiguous evidence that the LRAFMO is disrupted in the reentrant SG phase. An estimate of the AFM correlation radius at 4.2 K yields  $\sim 700 \text{ \AA}$ .

#### 4. CONCLUSION

With  $\gamma$ -iron alloys as an example, it has been shown that reentrant PM–AFM–SG transitions are induced by temperature changes in strongly frustrated Heisenberg AFMs in a zero magnetic field. In alloys in which the random-exchange component of the total exchange energy is smaller, such transitions are possible only in sufficiently strong magnetic fields.

The onset of a reentrant SG phase in Heisenberg systems is preceded by the formation of a noncollinear AFM (antiasperomagnetic) state. This state may be thought of as a superposition of an AFM ordering of spins and a spin-glass ordering of their components in the plane perpendicular to the antiferromagnetism vector. The formation of an antiasperomagnetic state is not accompanied by the development of dynamic processes (within the range of the time scales of these experiments). The realization of such a state apparently involves random static distortions of the collinear AFM structure of the alloys; these distortions become larger as the temperature is lowered.

A comparison of the results found by neutron diffraction with those found by  $\mu^+$  spin depolarization indicates that there is no LRAFMO in the reentrant SG phase, but there are antiasperomagnetic correlations of finite radius.

This picture of the hierarchy of magnetic phases and states is also applicable at a qualitative level to the temperature-induced reentrant transitions which occur in frustrated Heisenberg ferromagnets.<sup>17,18,23,24</sup>

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