# Concentrated FeNiCr spin glasses in a magnetic field

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The effect of a static magnetic field on the freezing temperature  $T_f(H)$  of the spin glasses  $\operatorname{Fe}_x \operatorname{Ni}_{80-x} \operatorname{Cr}_{20}$  is investigated. In anisotropic iron-enriched alloys  $(56 \le x \le 61)$  the  $T_f(H)$  dependence is described by the de Almeida-Thouless equation. In isotropic alloys with low iron content  $(2 \le x \le 5)$ , the phase line in the *H*-*T* plane on which the paramagnet-spin glass transition takes place is approximated by the Gabay-Toulouse equation.

## **1. INTRODUCTION**

Edwards and Anderson<sup>1</sup> were the first to state that a paramagnet-spin glass (PM-SG) transition is a true phase transition at the freezing temperature  $T_f$ . It might seem that the PM-SG phase transition should vanish in the presence of a magnetic field, since the Edwards-Anderson order parameter  $q_{EA} = \langle \langle s_i \rangle_T^2 \rangle_J$  is finite in the paramagnetic (PM) phase. However, de Almeida and Thouless (AT) have demonstrated,<sup>2</sup> in the Ising model, that a PM-SG phase transition can take place also in a magnetic field, and the line that separates the PM and SG phases in the H-T plane is described by the equation

$$\tau = (H/H_{AT})^{\eta_{h}},\tag{1}$$

where  $\tau = 1 - T_f(H)/T_f(0)$ , and  $H_{AT}$  is a constant. Here  $T_f(0)$  and  $T_f(H)$  are the SG freezing temperatures in a zero magnetic field and in a field H.

In practice one deals frequently with Heisenberg systems. Such a case was theoretically considered in Refs. 3 and 4, in which it was shown that for isotropic SG in the presence of a magnetic field the transition to the SG state proceeds along the Gabay-Thouless (GT) line, the equation of which in the H-T plane can be written as

$$x = (H/H_{GT})^2.$$
 (2)

The spin components perpendicular to the direction of the external magnetic field freeze on the GT line,<sup>3</sup> and in contrast to the transverse ones, the longitudinal irreversible phenomena that occur in such a state are found to be "weak." <sup>4</sup> With further lowering of the temperature along the line whose equation is given by (1), the longitudinal components of the vector spins in a Heisenberg SG freeze, and this leads to the onset of "strong" longitudinal irreversible processes. In the opinion of several workers,<sup>5</sup> this line is not a phase boundary but is a "crossover" line between the regions of weak and strong longitudinal irreversibility.

Experimental investigations of the influence of a constant magnetic field on the freezing temperatures of various SG<sup>6</sup> have shown that in most cases  $T_f(H)$  is described by the AT equation [Eq. (1)]. In some experiments,<sup>7</sup> however, the GT transition [Eq. (2)] could be observed. Analysis of the experimental data shows that in the study of SG account must be taken of anisotropic interactions that are present in real systems. The presence of such interactions influences substantially both the temperature  $T_f(0)$  and the form of the PM-SG phase boundary in a magnetic field.<sup>8</sup>

We have investigated the influence of a constant magnetic field on the PM-SG phase boundary in a system of  $Fe_x Ni_{80-x} Cr_{20}$  alloys. It was shown earlier<sup>9</sup> that such a transition takes place in the composition ranges  $2 \le x \le 5$  and  $56 \le x \le 61$ . Alloys with higher iron concentrations are characterized by a noticeable anisotropy,<sup>10</sup> whereas iron-poor alloys are practically isotropic. It is this circumstance that permits assessment of the influence of anisotropy on the form of the PM-SG boundary in the H-T plane.

# 2. EXPERIMENT

The sample preparation procedure is described in Ref. 9. Immediately prior to the measurements, the cylindrical samples (15-20 mm long and 1-3 mm in diameter) were annealed in an inert atmosphere for two hours at 1400 K and then quenched in water.

The complex dynamic magnetic susceptibility in magnetization-reversing fields 1-3 Oe at frequencies 20-600 Hz were measured by the procedure described in Ref. 11. In all cases the phase shift between the reference channels for the measurement of the real ( $\chi'_0$ ) and imaginary ( $\chi''_0$ ) magnetization components was maintained at 90°  $\pm$  0.2°. The static susceptibility was measured with a vibrating-string magnetometer.

#### 3. EXPERIMENTAL RESULTS AND DISCUSSION

# 3.1. $Fe_x Ni_{a0-x} Cr_{20}$ alloys with high iron concentration (56<x < 61)

Dynamic magnetic susceptibility. At the phase transition temperature  $T_f$ , SG acquires besides the Edwards-Anderson order parameter  $q_{EA}$  also a parameter<sup>8</sup>  $\Delta(T, H)$  connected with non-ergodic character of the SG phase. One experimental manifestation of the nonergodic character of SG is the divergence of the imaginary component of the dynamic magnetic susceptibility (DMS) near  $T_f$  together with its anomalously high value below  $T_f$  (Refs. 8 and 12). Consequently, by placing the SG in a magnetic field and studying the temperature dependences of  $\chi_0^{"}(T)$  we can assess the field dependence of  $T_f(H)$  and thus verify the predictions of the theory [Eqs. (1) and (2)].



FIG. 1. Temperature dependences of the real  $(\chi'_0)$  and imaginary  $(\chi''_0)$  components of the dynamic magnetic susceptibility of the Fe<sub>58</sub>Ni<sub>22</sub>Cr<sub>20</sub> alloy in static magnetic fields: 1–0, 2–4, 3–8, 4–16 Oe. Magnetization-reversing field strength is 1 Oe, frequency is 36 Hz.

Figure 1 shows by way of example typical temperature dependences of the real  $(\chi'_0)$  and imaginary  $(\chi''_0)$  components of the DMS of the alloy  $Fe_{58}Ni_{22}Cr_{20}$ , measured in static magnetic fields of various strengths. It can be seen that  $\chi_0''(T)$  has the maximum typical of SG, near which is observed also an abrupt increase of  $\gamma_0''(T)$ , due to the critical slowing down of the spin fluctuations as  $T_f$  is approached from the PM region. The experimental  $\chi'_0(T)$  and  $\chi''_0(T)$ dependences, both in a zero magnetic field and in fields of various strengths, together with calculations of the reciprocal relaxation rate  $\Gamma_0^{-1}$  of the magnetic moments of the system and extrapolation  $\Gamma_0^{-1} \rightarrow \infty$  at  $T \ge T_f$  (Ref. 12), were used to calculate the true freezing temperatures  $T_f(H)$  of this alloy and to plot the H-T diagram shown in Fig. 2 (dark circles). The shape of the obtained diagram is similar to the theoretically predicted<sup>2</sup> one, and can be described analytically by Eq. (1) with  $H_{AT} = 150 \pm 10$  Oe and with the exponent of H equal to  $0.66 \pm 0.3$ .

It must be noted that the measured exponent agrees very well with the value predicted theoretically for the Ising model. At the same time, the constant  $H_{AT}$  is in this case anomalously small. The small  $H_{AT}$  of the studied alloy can be understood from the following considerations. In accordance with the conclusions of Ref. 2,

$$H_{AT} = [2kT_{f}(0)/\sqrt{5}\mu_{eff}](1 - \mathcal{J}_{0}/\mathcal{J}).$$
(3)

Here  $\mu_{\text{eff}} = 2s\mu_B$ ,  $\tilde{J}_0 = \langle J_{jj} \rangle$ , and  $\tilde{J} = (\langle J_{ij}^2 \rangle - \langle J_{ij} \rangle^2)^{1/2}$ . Assuming for the given alloy  $\mu_{\text{eff}} = 2\mu_B$  and  $1 - \tilde{J}_0/\tilde{J} = 1 - \theta/T_f(0)$ , where  $\theta = 4$  K is the paramagnetic Curie temperature, we obtain  $H_{\text{AT}} \sim 80$  kOe, much higher than the measured value. It must be recognized, however, that this alloy is close to the critical concentration ( $x_c = 55$ ) at which long-range ferromagnetic order is produced.<sup>9</sup> Consequently, ferromagnetic correlations with large values of the magnetic moments exist in this alloy. The value of  $\mu_{\text{eff}}$  in Eq. (3) is therefore not  $2\mu_B$  but is much higher. Furthermore  $J_0/J \rightarrow 1$  as  $x \rightarrow x_c$ , so that  $H_{\text{AT}} \rightarrow 0$ .

Statistical magnetization. Besides the anomaly of  $\chi_0^{"}$ near  $T_f$ , one more manifestation of the non-ergodic behavior of spin glasses is their sensitivity to the thermomagnetic pri-



FIG. 2. *H-T* phase diagram of the spin glass  $Fe_{58}Ni_{22}Cr_{20}$ ,  $\tau = 1 - T_f(H)/T_f(0)$  is the relative temperature.

or history.<sup>13</sup> In other words, below  $T_f(H)$  the magnetization measured after cooling the sample in a zero magnetic field  $(I^{ZFC})$  differs from the magnetization measured after cooling it in a field of finite strength  $(I^{FC})$ . The temperature at which  $I^{ZFC}$  and  $I^{FC}$  begin to differ is in fact the SG freezing temperature  $T_f(H)$  in the given magnetic field H.

Figure 3 shows the temperature dependences of  $I^{ZFC}$  (light circles) and  $I^{FC}$  (dark circles) of the Fe<sub>58</sub>Ni<sub>22</sub>Cr<sub>20</sub> alloy we are studying. Just as in dilute SG,<sup>14</sup> in this alloy the temperature  $T_f(H)$  at which the  $I^{ZFC}$  and  $I^{FC}$  are no longer equal (marked by arrows) decreases as the measuring-field strength increases. The  $T_f(H)$  values obtained from measurements of the static magnetization are marked on the H-T diagram (Fig. 2, light circles) and agree well with the data obtained from DMS measurements. This is due to the relatively large ( $\sim 10^2$  s) characteristic time scale of static-magnetization measurements, and confirms the conclusion that the true SG freezing temperature must be determined in an approximation with an infinite observation time.

The dependence, noted by a number of workers, <sup>15</sup> of the position of the PM-SG phase boundary in a magnetic field on



FIG. 3. Temperature dependences of the static magnetization of the  $Fe_{58}Ni_{22}Cr_{20}$  alloy in fields of various strengths: 1-2, 2-8, 3-16, 4-32 Oe.



FIG. 4. Temperature dependence of the ergodicity parameter  $\Delta$  of the spin glass  $Fe_{58}N_{22}Cr_{20}.$ 

the characteristic time scale of the experiment is apparently due to the fact that, unlike our study, they failed to consider scaling and the  $T_f(H)$  were taken to be the temperatures of the characteristic anomalies of the SG magnetic properties (e.g., the temperature of the inflection on the  $\chi_0''(T)$  plots), the positions of which depend strongly on the measurement time.

It should be noted that, in the opinion of the authors of Ref. 16, the temperature  $T_f(H)$  can be determined also from the temperature dependence of the nonergodicity parameter  $\Delta(H,T)$ . We have calculated in the present paper, by the procedure proposed in Ref. 16, the temperature dependence of  $\Delta(T)$  for the alloy described above in a 25 Oe magnetic field.

Figure 4 shows the behavior of  $\Delta(T)$  in the critical region near  $T_f$  (H = 25 Oe). It can be seen that on a log-log plot the experimental points lie well on a straight line. This means that the  $\Delta(T)$  temperature dependence can be described by the power law typical of phase transitions

$$\Delta(\tau) \propto \tau^{\beta'}, \beta' = 1.6^{+0.4}_{-0.1}, \tag{4}$$

where  $\beta'$  is the critical exponent.

The dynamic SG theory<sup>8</sup> predicts for H = 0 a critical behavior of  $\Delta(T)$  in the form (4), with  $\beta' = 2$ . The somewhat smaller value of  $\beta'$  obtained in our case is apparently due to the fact that  $\Delta T$  was measured in a nonzero magnetic field. The temperature  $T_f$  determined in this experiment (H = 25 Oe) is marked on the H-T diagram by a black triangle, and can be seen from Fig. 2 to be likewise located on the PM-SG transition line determined above.

To conclude this section, we note that similar patterns in the variation of  $T_f(H)$  were observed also in other alloys of the Fe<sub>x</sub> Ni<sub>80-x</sub> Cr<sub>20</sub> system with high iron content (x = 60 and 56 in Refs. 11 and 17, respectively).

It should be emphasized that while these alloys are Heisenberg magnets in the ferromagnetic region (x < 56),<sup>18</sup> as shown above, in the presence of a magnetic field they behave in the SG region as Ising systems. It was shown in Ref. 10 that such alloys, with large iron content, are characterized at temperatures lower than  $T_f$  by a shift of the hysteresis loop after they are cooled in a magnetic field. This phenomenon is usually attributed to the presence of unidirectional (ex-



FIG. 5. H-T phase diagram of the alloy Fe<sub>4</sub>Ni<sub>76</sub>Cr<sub>20</sub>.

change) anisotropy<sup>19</sup> or to Dzyaloshinskiĭ-Moriya anisotropy.<sup>20</sup> Without dwelling in detail on the anistropy types present in this SG, we note that in both cases their presence should lead to an Ising behavior of  $FeNiCr_{20}$  spin glass, as is indeed observed.

We consider now SG of the  $Fe_x Ni_{80-x} Cr_{20}$  system, for which the anisotropic interactions can be neglected.

## 3.2. Fe<sub>x</sub> Ni<sub>80-x</sub> Cr<sub>20</sub> alloys with low iron content ( $2 \le x \le 5$ )

Just as for the alloys with high iron content, we investigated for these alloys the temperature dependences of  $\chi'_0$ ,  $\chi''_0$ ,  $I^{\rm ZFC}$  and  $I^{\rm FC}$  in magnetic field of various strengths. The experimental data obtained for alloys with x = 2, 3, and 4 are qualitatively similar to the results shown in Figs. 1 and 3. The equation for the phase-transition line in the H-T plane takes for these alloys (Fig. 5) the GT form (2), with an exponent  $1.8 \pm 0.2$  for H and with  $H_{\rm GT} = (57 \pm 2)$  Oe. It can be deduced from the foregoing that FeNiCr<sub>20</sub> alloys with low iron content are Heisenberg SG. It should be noted that in this case, too, the low value of the constant  $H_{\rm GT}$  has the same causes as in alloys with high iron content.

#### 4. CONCLUSION

We have investigated the dynamic magnetic susceptibility and the static magnetization of  $Fe_x Ni_{80-x} Cr_{20}$  spin glasses in magnetic fields. In SG with large (55 < x < 61)iron content the phase line on which the PM-SG phase transition takes place in a magnetic field can be described by the de Almeida-Thouless equation (1) for Ising SG. For Heisenberg SG with low iron content (2 < x < 5) the equation of the corresponding line takes the form (2) predicted by Gabay and Toulouse. This reason for this difference is that the alloys of the first type have a noticeable anisotropy whereas those of the other are practically isotropic.

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