Transverse freezing in the amorphous spin-glass $Fe_{10}Ni_{70}P_{20}$

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The field-temperature phase diagram of the amorphous spin-glass $Fe_{10}Ni_{70}P_{20}$ has been explored using $\chi'_{\perp}$, the ac susceptibility perpendicular to an applied dc field. As the temperature is reduced in a finite magnetic field, the spin-glass first enters a regime characterized by weak freezing of the spin components perpendicular to the applied field. This weak transverse freezing region is entered along a line suggestive of the mean-field Gabay-Toulouse line. At lower temperatures strong freezing of the spin components both parallel and perpendicular to the applied field occurs along a de Almeida—Thouless—like line. Two new spin-glass features are observed along this line: (i) a shoulder in $\chi'_{\perp}$ in constant field, and (ii) a maximum in the isotherms of $\chi'_{\perp}$. The shoulder is of particular interest since it sharpens in high fields, in contrast to all previously observed spin-glass features. The suppression of strong transverse freezing in favor of a weak transverse freezing region is attributed to anisotropy effects.

I. INTRODUCTION

The phase diagram of a spin-glass in the presence of an applied field has attracted much attention recently. This interest was first aroused by de Almeida and Thouless, who discovered a field-dependent instability line in the mean-field model of an Ising spin-glass.[1] The expression for this line, referred to as the AT line, is given by

$$H = H_{AT}\tau^{3/2} , \tag{1}$$

where $\tau = 1 - T/T_g$ (T_g is the spin-glass freezing temperature). In units where k_B, μ_B, and T_g are unity $H_{AT} = 2/\sqrt{3}$. This line was later interpreted as a line of phase transitions by Parisi and Toulouse.[2]

More recently the mean-field theory of spin-glasses has been extended to include Heisenberg spins by Gabay and co-workers.[3,4] The most striking feature of this theory is the prediction of a second, field-dependent phase transition, referred to as the transverse freezing line. In this paper we focus on an experimental search for this line in the amorphous spin-glass $Fe_{10}Ni_{70}P_{20}$. Evidence is found for such a transition; however, the phase diagram appears to be more complicated than predicted by theory. Before presenting the data it will be useful to summarize the results of mean-field theory.

Assuming an isotropic Heisenberg Hamiltonian, Gabay and Toulouse employed replica analysis to investigate the free energy. The exchange interactions were assumed to be random with zero mean (i.e., no net ferromagnetic or antiferromagnetic tendency) and of infinite range. The phase diagram in the H-T plane obtained for this model is shown in Fig. 1. In finite fields the paramagnetic phase yields to a spin-glass phase characterized by the freezing of the spin components perpendicular to the applied field. This phase, dominated by transverse freezing, is referred to as the transverse spin-glass (TSG) phase. The TSG phase is entered along the line

$$H = H_{GT}\tau^{1/2} , \tag{2}$$

called the Gabay-Toulouse (GT), or transverse freezing line, where $H_{GT} = 10/\sqrt{23}$, for three-component spins. As the temperature is decreased further, the spin components parallel to the field also freeze. This phase, with both longitudinal and transverse spin components frozen, is called the mixed spin-glass (MSG) phase. The line separating the TSG and MSG phase is identical to that derived by de Almeida and Thouless, and is given by Eq. (1), with $H_{AT} = (\frac{2}{5})^{1/2}$.

In real spin-glasses evidence for the AT line is plentiful. Using relaxation measurements, Yeshurun *et al.*[5] have shown that strong irreversibility in spin-glasses vanishes along a AT-like line. These results have been confirmed by a variety of experimental techniques performed on a wide spectrum of spin-glasses.[6–12] The AT line has also been observed in numerical simulations.[13,14] In both the experimental and numerical work, the observed AT line has the correct temperature dependence; however, the

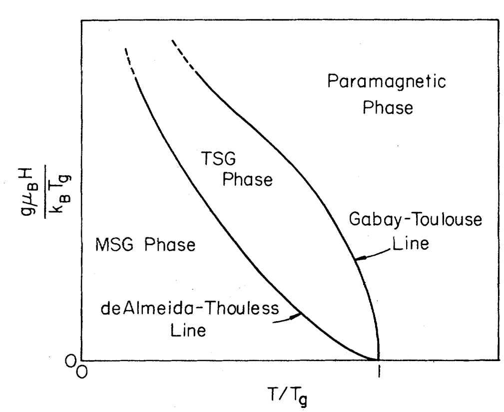

FIG. 1. Mean-field phase diagram for Heisenberg spin-glasses. The paramagnetic, transverse spin-glass (TSG) and the mixed spin-glass (MSG) phases are pictured.

characteristic fields are generally an order of magnitude smaller than those given by mean-field theory. The reason for this is not understood. Considerable effort has also been expended in search of the GT line in real spin-glasses. In some experiments, certain spin-glass characteristics (associated with magnetization, relaxation, heat capacity, etc.) are observed whose temperature and field dependence are qualitatively similar to the GT line.[10,11,15] Aside from this similarity, however, these features are not obviously related to transverse freezing. Furthermore some of these features are observed in Ising spin-glasses, which should not display transverse freezing.

The most direct method of searching for the GT line is through the susceptibility perpendicular to an applied field. In their original paper, Gabay and Toulouse derived expressions for the susceptibility of Heisenberg spins,[3] both parallel and perpendicular to an applied field. For three-component spins they are

$$\chi'_{\parallel}=\frac{C}{T}(1-q_{\parallel}+2x)\ , \tag{3a}$$

$$\chi'_{\perp}=\frac{C}{T}(1-x-q_{\perp})\ , \tag{3b}$$

where $x=([\langle S_{\parallel}^2\rangle]_{\rm av}-1)/2$ is the quadrupolar spin parameter (measuring the uniaxiality of the spin vector along the applied field), and $q_{\parallel}=[\langle S_{\parallel}\rangle^2]_{\rm av}$ and $q_{\perp}=[\langle S_{\perp}\rangle^2]_{\rm av}$ are Edwards-Anderson order parameters for spin components parallel and perpendicular to the applied field, respectively. The spins are normalized so that $\sum_{\mu}S_{i\mu}^2=3$, $\mu=x,y,z$. These equations are merely modified versions of the well-known Fischer relation for Ising spin-glasses. They arise from the absence of spatial correlations in spin-glasses and their validity is independent of mean-field theory.[16] The way in which $\chi'_{\parallel}(H,T)$ signals the onset of longitudinal freezing is not entirely clear since $q_{\parallel}$ is not a legitimate order parameter in finite field; it is always nonzero regardless of temperature. However, since $q_{\perp}$ is always a well-defined order parameter, transverse ordering should be accompanied by a decrease in $\chi'_{\perp}(H=\text{const}, T)$. These relations, unfortunately, cannot be applied quantitatively to real spin-glasses. The effects of ferromagnetic couplings and anisotropic interactions, which are unavoidably present in real systems, have not been included in the derivation of these equations. However, since most of the physical content of these relations can also be argued on intuitive grounds (regardless of the presence of ferromagnetic or anisotropic interactions), they will be useful in interpreting the transverse susceptibility measurements.

II. EXPERIMENTAL

The material used in this study was amorphous $Fe_{10}Ni_{70}P_{20}$. The amorphous nature of this system ensures that on a global scale the spins should be of Heisenberg type. However, random anisotropies are unavoidably present (e.g., dipole-dipole) and they may effect the local behavior of the spins.

The samples were prepared by centrifugal quenching. The ribbons were cut into pieces measuring $5\times1\times0.05$ mm^3. Previous work has shown that this material exhibits its typical spin-glass properties.[9] This particular sample has a Curie temperature of 18 ± 1 K and a spin-glass freezing temperature of 26.8 K. The dc magnetization data were taken in a Princeton Applied Research model 155 vibrating sample magnetometer. Temperature was measured using a type-E thermocouple mounted <1 mm from the sample. The ribbons were orientated so that the long axis was along the applied field in order to reduce demagnetizing effects.

The transverse susceptibility was measured using a conventional ac susceptibility apparatus. The sample was mounted in one of two counter wound coils; a solenoid was used to provide a small ($\sim$0.7 Oe) oscillating field along the axis of the coil while an electromagnet was used to establish a dc field perpendicular to this axis. All measurements were performed at 50 Hz. The temperature was measured with a type-E thermocouple mounted 3 mm from the sample. The sample was oriented so that the probe field was along the long axis and the applied dc field was in the plane of the ribbons in order to reduce demagnetizing effects. With this apparatus the real part of the transverse susceptibility ($\chi'_{\perp}$) was easily measured. However, the imaginary part ($\chi''_{\perp}$), due to its small magnitude, displayed considerable contamination by $\chi'_{\perp}$. As a result only the gross behavior of $\chi''_{\perp}$ could be determined.

III. RESULTS

The results shown in Fig. 2 are typical of spin-glasses with relatively large ferromagnetic coupling, namely, a maximum in $M_{\parallel}(H=\text{const}, T)$ and a large positive Curie temperature. The position of the maximum is field dependent, as with many spin-glasses, moving toward higher temperatures at low fields, and then toward lower temperatures at higher fields.

The main results reported here are for the field and temperature dependence of the real part of the transverse susceptibility $\chi'_{\perp}(H,T)$. The transverse susceptibility was measured in constant field under both zero-field-cooling (ZFC) and field-cooling (FC) conditions. As can be seen

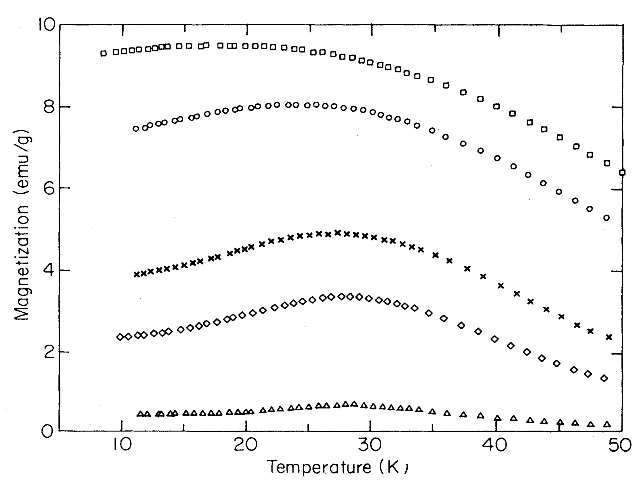

FIG. 2. Field-cooled longitudinal magnetization curves: △, 10 Oe; ◇, 100 Oe; ×, 200 Oe; ○, 500 Oe; □, 1000 Oe.

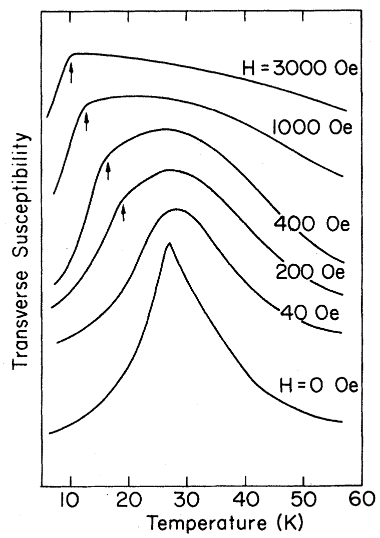

FIG. 3. Field-cooled transverse susceptibility $\chi'_\perp$. Arrows indicate the position of the shoulder. The zeros have been offset for clarity. Also the curves for fields greater than 40 Oe have been enlarged. The scale factors for the 200-, 400-, 100-, and 3000-Oe curves are, respectively, 2, 4, 8, and 40. Data were taken at 0.4-K intervals; scatter in the data is comparable to the thickness of the line drawn through the points.

in Fig. 3, in zero field $\chi'_\perp(0,T)$ $[=\chi'_{\parallel}(0,T)]$ displays the sharp cusp characteristic of the spin-glass transition. When a finite field is established $\chi'_\perp$ decreases, as does the longitudinal ac susceptibility. However, at fields greater than 40 Oe it becomes apparent that a shoulder is present on the low-temperature side of the maximum. Such a prominent feature is not observed in $\chi'_{\parallel}(H=\text{const},T)$. The temperature at which this shoulder appears depends on the magnitude of the longitudinal field. For relatively small fields (<900 Oe) this dependence is given by

$$H_c = H_0 \tau^{1.8\pm0.1} \, . \tag{4}$$

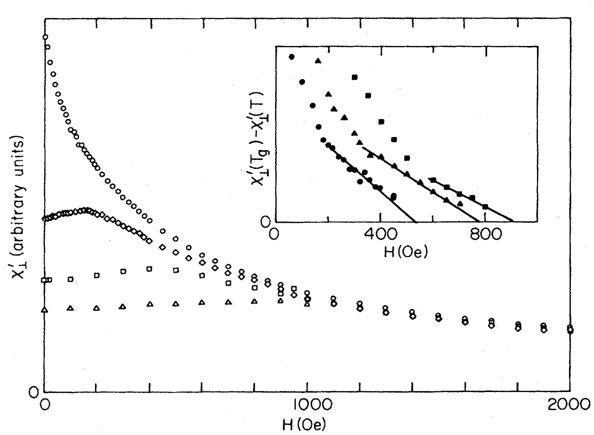

FIG. 4. Zero-field-cooled isotherms for the transverse susceptibility $\chi'_\perp$: ○, 26.8 K; ◇, 21.2 K; □, 17.2 K; △, 13.5 K. The inset shows the difference between the freezing isotherm and several isotherms slightly below the freezing temperature: ●, 26.5 K; ▲, 25.0 K; ■, 21.2 K.

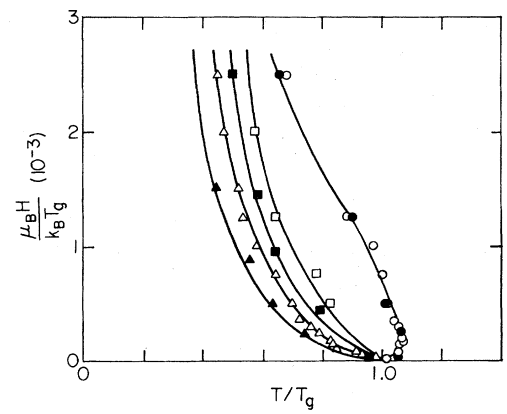

FIG. 5. Plot of the various field-temperature-dependent features seen in the transverse susceptibility $\chi'_\perp$ and longitudinal magnetization $M_{\parallel}$: ○, maximum in $\chi'_\perp(H=\text{const},T)$; ●, maximum in $M_{\parallel}(H=\text{const},T)$; □, shoulder in ZFC $\chi'_\perp(H=\text{const},T)$; ■, maximum in ZFC $\chi'_\perp(H,T=\text{const})$; △, shoulder in FC $\chi'_\perp(H=\text{const},T)$; ▲, maximum in FC $\chi'_\perp(H,T=\text{const})$.

At larger fields H_c increases more strongly with τ. The characteristic field, H_0, which appears in Eq. (4) depends on the cooling condition. For FC, we find that $H_0=2000$ Oe, while for ZFC, $H_0=3600$ Oe. The shoulder sharpens dramatically with increasing longitudinal field; see Fig. 3. This is markedly different from all other spin-glass features, which tend to flatten and become less distinct in large fields. This shoulder also coincides with a maximum in $\chi''_\perp(H=\text{const},T)$, indicating that it is associated with viscous behavior of the transverse spin components. In addition, $\chi'_\perp(H=\text{const},T)$ displays a field-dependent maximum, similar in behavior to that of the longitudinal magnetization.

Isotherms of $\chi'_\perp(H,T)$ were measured under both ZFC and FC conditions. The most striking feature of the isotherms is a temperature-dependent maximum; see Fig. 4. This feature is retraceable: If the field is increased above the maximum then returned to its original value there is no observed hysteresis. The occurrence of a maximum is surprising and represents a new feature of the spin-glass phase. It is in contrast to the monotonic field dependence of $\chi'_{\parallel}(H,T=\text{const})$ observed in a paramagnet. The loci of points described by this maximum follow the functional form given in Eq. (4) for low fields. The characteristic field H_0 in Eq. (4) is also dependent on cooling condition, with $H_0=1480$ Oe for FC, and 2750 Oe for ZFC. Another interesting feature of the isotherms is that for sufficiently high fields they appear to join with the freezing temperature isotherm. This is most dramatic for temperatures near T_g (see inset, Fig. 4). The positions of the

TABLE I. Coefficients of $\tau^{1.8}$ for various features in the susceptibility of $Fe_{10}Ni_{70}P_{20}$.

Property	FC	ZFC
Shoulder in isochamps	2.00 kOe	3.60 kOe
Maximum in isotherms	1.48	2.75
Parallel irreversibility		2.30

various features in $\chi'_\perp(H,T)$ under differing field cycling conditions are given in Fig. 5, and the values of the various characteristic fields are summarized in Table I.

IV. DISCUSSION

As may be seen in Fig. 5, the loci of points determined from the shoulder in $\chi'_\perp(H, T=\text{const})$ are suggestive of the AT longitudinal spin-glass freezing lines. Indeed, previous examination of longitudinal relaxation showed that longitudinal freezing does occur in the vicinity of the observed anomalies.[17] In those experiments the sample was cooled in zero field to a fixed temperature below the freezing temperature, then a field was established in a stepwise fashion. Strong irreversibility in the longitudinal magnetization was found to set in below a line given by Eq. (4), with a characteristic field of $H_0=2300$ Oe. It seems therefore that the anomalies in $\chi'_\perp(H,T)$ are correlated with the freezing of the longitudinal spin components.

The only distinctive feature of the transverse susceptibility that bears a qualitative resemblance to the GT line is the maximum in $\chi'_\perp(H=\text{const}, T)$. As can be seen from Fig. 5, it coincides with the maximum in $M_{\parallel}(H=\text{const}, T)$. Similar field-dependent behavior of the maximum of $M_{\parallel}(H=\text{const}, T)$ has been observed in Y—4 at. % Er,[18] which is known to be an Ising spin-glass, and thus displays no transverse freezing, and nor, it seems reasonable to conclude, is the maximum in $M_{\parallel}(H=\text{const}, T)$ seen in $Fe_{10}Ni_{70}P_{20}$ associated with transverse freezing. Because the field dependences of the maximum in $M_{\parallel}(H=\text{const}, T)$ and $\chi'_\perp(H=\text{const}, T)$ are identical, it is likely that they have a common origin, and that neither is associated with transverse freezing.

If, as discussed above, none of the features in the transverse susceptibility correspond to the predicted GT freezing line, what are the causes of the observed anomalies? Using the Fischer relations [Eqs. (3a) and (3b)], we can ascribe the shoulder in the transverse susceptibility observed in Fig. 3 to two sources: (a) the rapid freezing of the transverse spin components (i.e., a large increase in $q_\perp$), or (b) an increased tendency for the spins to lie along the longitudinal field axis (increase in x). In case (a) the decrease in $\chi'_\perp(H=\text{const}, T)$ should be accompanied by an increase in the relaxation times associated with the transverse spin components. Indeed, a maximum in $\chi''_\perp(H=\text{const}, T)$ is observed at the same temperature as the shoulder in $\chi'_\perp(H=\text{const}, T)$, indicating that this anomaly is associated with transverse freezing. In case (b), a strong increase in $\chi'_{\parallel}(H=\text{const}, T)$ should be observed at the same temperature, but is not. Although, a rapid decrease in $q_\perp$ with increasing temperature is required to produce the observed anomaly, this does not exclude a subsequent gradual decrease in $q_\perp$ toward 0 at higher temperatures. The data, in fact, suggest that such a tail in $q_\perp$ may exist. The quadrupolar spin parameter x, because it is not associated directly with the freezing phenomenon, ought not to vary rapidly with temperature near T_g. Thus, the Fischer relation [Eq. (3b)] suggests that

$$q_\perp \alpha \chi'_\perp(T_g,H)-\chi'_\perp(T,H), \quad T\le T_g .$$

This means that the point at which isotherms of the transverse susceptibility in Fig. 4 join the freezing isotherm marks the vanishing of $q_\perp$ (to the extent that x is temperature independent). The fields at which this occurs have been estimated for several isotherms (see inset, Fig. 4) with the results displayed in Fig. 6. As can be seen in Fig. 6 the apparent vanishing of $q_\perp$ occurs at field-temperature points suggestive of the GT transverse freezing line. Although this is not conclusive evidence for GT-like freezing, it does suggest that a weak form of transverse freezing occurs along such a line. Careful dynamical measurements are required to determine the true nature of this weak transverse freezing regime.

With this in mind some conclusions may be drawn using the Fischer relation for $\chi'_\perp(H,T)$ [Eq. (3b)]. If the shoulder is indeed the mark of strong transverse freezing (i.e., a rapid increase in $q_\perp$) then the maximum that occurs at higher temperatures must be mainly attributed to changes in x. As the spin-glass regime is approached from the paramagnetic regime in finite field there must be a rise in the uniaxial nature of the system causing an increase in x and, consequently, the observed decrease (below the maximum) in $\chi'_\perp(H=\text{const}, T)$. The more dramatic decrease observed at lower temperatures (i.e., the shoulder) is then attributed to an increase in $q_\perp$. In large fields, the spins lie predominantly along the applied longitudinal field, causing x to approach its limiting value of unity, regardless of temperature, and thus making the effect of $q_\perp$ relatively more important. This accounts for the sharpening of the shoulder in large fields.

The ideas developed in the preceding paragraph can explain the maximum in $\chi'_\perp(H, T=\text{const})$. In zero field, at temperatures below the spin-glass freezing temperature, and due to the absence of a preferred axis, the system must be in the MSG phase, characterized by freezing along all directions [i.e., $q_\perp(=q_{\parallel})\neq 0$, and $x=0$]. As the field is increased the transverse order parameter is suppressed, thus tending to increase $\chi'_\perp(H, T=\text{const})$

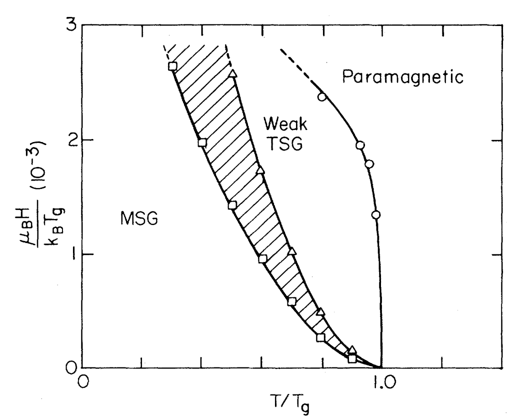

FIG. 6. The experimentally observed field-temperature spin-glass phase diagram. The paramagnetic, weak transverse spin-glass, and mixed spin-glass phases are shown. The lines separating these regions are ○, the vanishing of $q_\perp$ as determined by the joining of the isotherm; △, the ZFC shoulder in $\chi'_\perp(H=\text{const}, T)$; □, the FC shoulder in $\chi'_\perp(H=\text{const}, T)$. Both strong transverse and strong longitudinal freezing occur inside the shaded region.

while the quadrupolar spin parameter x is enhanced tending to decrease $\chi'_\perp(H, T=\text{const})$. The competition between $q_\perp$ and x is the most likely source of the maximum in $\chi'_\perp(H, T=\text{const})$. This explanation predicts that the shoulder occurs at a higher field than the maximum in $\chi'_\perp(H, T=\text{const})$—above the shoulder $q_\perp$ is small and $\chi'_\perp(H,T)$ is dominated by x, whose monotonic dependence on H would not lead to a maximum. This is indeed the observed sequence of events, as seen in Fig. 5.

V. CONCLUSIONS

The measurements and analysis presented here suggest that spin freezing in real spin-glasses occurs in the following fashion: The paramagnetic phase yields to a phase characterized by weak transverse freezing along a line reminiscent of the mean-field GT line; at lower temperatures a region is entered where strong transverse and longitudinal freezing is exhibited. Due to the ambiguity in experimentally defining the longitudinal freezing line it cannot be determined if both strong longitudinal and strong transverse freezing occurs precisely at the same temperature. If a difference exists it is relatively small. The experimentally observed phase diagram is portrayed in Fig. 6. Such behavior is not predicted by mean-field theory. The key to this difference may lie in the numerical work of Soukoulis, Grest, and Levin.[14] In their work on the free energy surface of Heisenberg spin-glasses, they included the effects of anisotropy [uniaxial and Dzyaloshinsky-Moriya (DM)] and net ferromagnetic interactions, both of which are neglected in most mean-field treatments. They found that both types of interactions significantly affect the nature of the transverse freezing line. Uniaxial and ferromagnetic interactions suppress, while the DM interaction enhances, the transverse freezing temperature. Unfortunately, their results cannot be compared directly to this study, since the amorphous nature of $Fe_{10}Ni_{70}P_{20}$ precludes uniaxial anisotropy resulting solely from crystal-field effects (although a random or induced uniaxial anisotropy may exist), and the absence of a displayed hysteresis loop suggests that the DM interaction does not play an important role.[19] Further, dipole-dipole forces are present, which have not been treated in the numerical work. However, the results of Soukoulis *et al.* do indicate that anisotropy cannot be neglected when examining transverse freezing. The fact that anisotropy may play a role in the observed suppression of strong transverse freezing is supported by the observed difference between the FC and ZFC transverse susceptibility results. In the FC case the field applied during the cooling process may induce a frozen-in anisotropy. Such an anisotropy must be uniaxial-like, since no evidence for unidirectional anisotropy is seen in this system. According to the numerical work, transverse freezing would then be suppressed. In the ZFC case any frozen-in anisotropy must be random in nature. Thus the observed transverse freezing line should more clearly resemble that for the Heisenberg case. Thus strong transverse freezing should occur at a higher temperature in the ZFC case than in the FC case. This is consistent with the experimental results. Although such field-induced anisotropy is difficult to explain, microscopically it is undoubtedly related to "movable" anisotropy observed in other systems.[20–23]

These results are probably applicable to spin-glass systems similar to $Fe_{10}Ni_{70}P_{20}$. However, they may not be relevant to systems in which the DM interaction plays an important role, as appears to be the case in *Cu*Mn and *Au*Fe. Numerical work has shown that transverse freezing is not easily suppressed in such systems.

While the interpretation of the present results has been guided by recent numerical work, it is evident that the models used in such studies are not yet sufficiently close to real spin-glass systems. The effects of random anisotropy (dipole-dipole, random uniaxial, etc.) and net ferromagnetic interactions require further study.

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