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Spin correlations near the ferromagnetic–spin-glass crossover point in amorphous Fe-Mn alloys

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We have performed a neutron scattering study of the spin correlations in the amorphous alloys $(Fe_{1-x}Mn_x)_{75}P_{16}B_6Al_3$ with $x=0.35$ and 0.25; the former exhibits short-range spin correlations at all temperatures. The latter displays a paramagnetic-ferromagnetic transition followed by a gradual evolution into a state characterized by $Q^{-2.6}$ power-law decay at low temperatures. We present a simple model for this behavior based on random field effects.

Recently, a number of concentrated alloy systems have been discovered which exhibit spin-glass (SG), ferromagnetic (FM) and apparently reentrant spin-glass (RSG) phases. These include the diluted insulating systems $Eu_xSr_{1-x}S$,[1] the metallic alloys PdFeMn[2] and Fe_xCr_{1-x}[3] and the amorphous alloys $(Fe_{1-x}Mn_x)_{75}P_{16}B_6Al_3$.[4] In general, these have been discussed in terms of a multicritical phase diagram [see, for example, Fig. 1(a)] of the sort first predicted by Sherrington and Kirkpatrick.[5] In this description, the so-called RSG phase is entered via a second-order transition from the FM phase with decreasing temperature. This model has been applied most extensively to the amorphous alloy $(Fe_{1-x}Mn_x)_{75}P_{16}B_6Al_3$ by Yeshurun *et al.*[4] These authors have used a scaling equation of state analysis to extract the critical exponents characterizing the FM-RSG transition. We emphasize, however, that in the published data there is no sharp feature acting as a signature of this phase transition. Instead, the transition temperature is included as a parameter in the scaling analysis.

Stimulated by the above, we have carried out a neutron scattering study of the spin fluctuations in $(Fe_{1-x}Mn_x)_{75}P_{16}B_6Al_3$ with $x=0.35$ and $x=0.25$. The $x=0.35$ material has x very near the critical concentration for FM order and exhibits a PM-SG transition near 40 K [see Fig. 1(a)].[4] The $x=0.25$ alloy exhibits a PM-FM transition at $T=221$ K followed by an apparently gradual transition into an SG-like state.

The amorphous $(Fe_{1-x}Mn_x)P_{16}B_6Al_3$ ribbons used for our neutron scattering experiments at Brookhaven were prepared in the same fashion as those which have been extensively characterized by ac susceptibility and dc magnetization measurements.[4] The quasielastic experiments were carried out using the Small Angle Neutron Scattering (SANS) facility of the Brookhaven Biology Department. The incident neutron energy was 14.6 meV, and the net resolution was 0.006 $Å^{-1}$ half-width at half maximum (HWHM). The inelastic experiments were performed on a triple-axis spectrometer with a fixed incoming neutron energy of 4.5 meV. We note that our inelastic measurements justify the quasielastic approximation in the SANS studies for both samples.

We discuss first the quasielastic scattering in the $x=0.35$ alloy. Figure 1(b) shows the temperature dependence of the scattered intensity at two representative momentum transfers. The principal feature of these data is the broad maximum centered near 40 K, which is approximately where the phase diagram of Yeshurun *et al.*[4] [Fig. 1(a)] would place the SG transition. To estimate the Q dependence of the magnetic scattering we subtract the high-temperature data at 200 K as background. Figure 2(a) shows the resulting inverse intensities $I^{-1}(Q)$ plotted as a function of Q^2 at various temperatures. The diffuse scattering follows the Ornstein-Zernike form $I(Q)=A/(\kappa^2+Q^2)$ where $\kappa=\xi^{-1}$ is the FM inverse correlation length. A maximum length of $\xi=25$ Å is achieved near the SG transition temperature. Similar behavior has been found previously in other systems.[1,3]

Much richer behavior is obtained in the $x=0.25$ alloy. As illustrated in Fig. 1(c), for all wave vectors between 0.020 and 0.11 $Å^{-1}$, the diffuse scattering shows a sharp peak at 221 K signaling the transition into the ferromagnetic phase; the scattering is then only weakly temperature dependent between 200 and

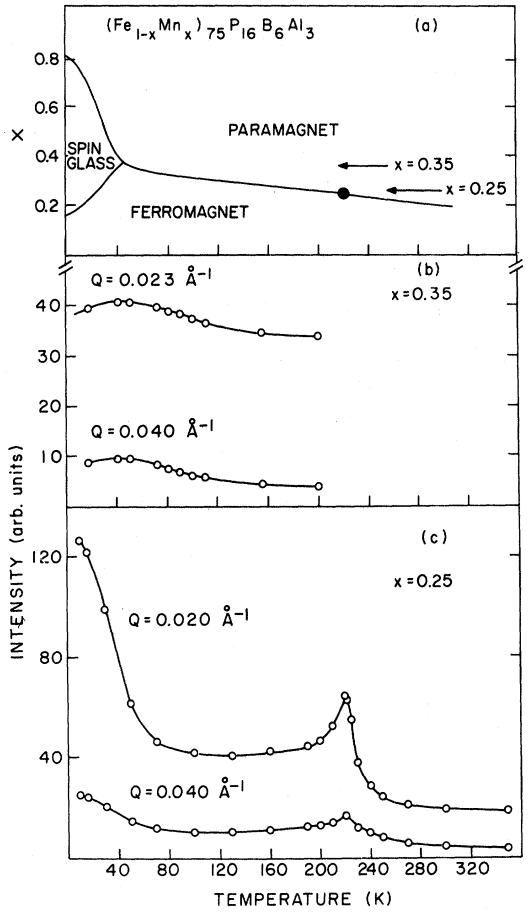

FIG. 1. (a) Magnetic phase diagram of $(Fe_{1-x}Mn_x)_{75}P_{16}B_6Al_3$ as deduced by Yeshurun *et al.* (Ref. 4). Solid point represents our measured T_c. (b), (c) Temperature dependence of small angle neutron scattering spectra, uncompensated for nonmagnetic background. Intensities are in the same units for both samples.

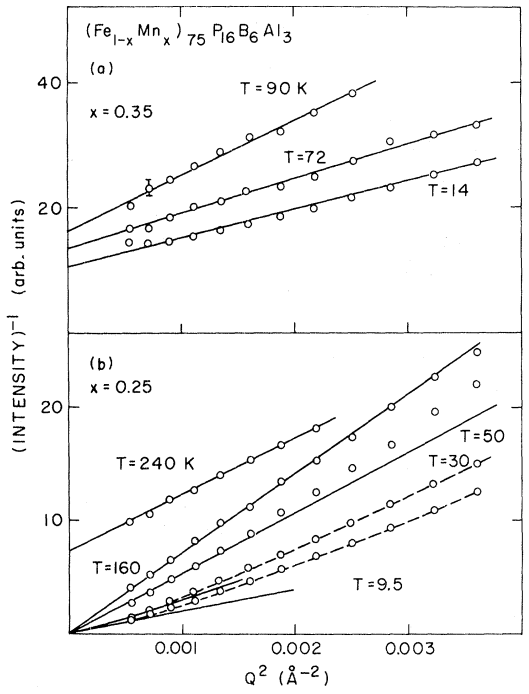

FIG. 2. Inverse intensity I^{-1} vs Q^2 at various temperatures for (a) $x=0.35$ and (b) $x=0.25$ samples. The $T=200$ and 350 K spectra were taken as background for the $x=0.35$ and 0.25 data, respectively. The dashed line passing through the $T=9.5$ K data ($x=0.25$) represents a $Q^{-2.6}$ power law.

80 K. Below 80 K, a dramatic rise occurs in the scattering intensity. The corresponding Q dependence of the scattering, with the 350-K data subtracted as background, is shown in Fig. 2(b) for selected temperatures. Above 221 K, the intensity follows the Ornstein-Zernike form for $Q^2 < 0.003$ Å^{-2} and $\kappa \to 0$ as $T \to T_C$, which is what occurs at a normal PM-FM transition. At intermediate temperatures, we observe that $I \sim Q^{-2}$, as expected for spin-wave scattering in a ferromagnet. However, in the low-temperature region, where the anomalous increase in intensity occurs, significant departures from the Q^{-2} law are observed. At the lowest temperatures, the data are well described by a $Q^{-2.6}$ singularity. We stress that because the data only extend to $Q=0.02$ Å^{-1}, we cannot exclude correlation lengths $\xi > 100$ Å and the related possibility of a second-order FM-RSG transition into a true SG phase with $\infty > \xi > 100$ Å.

The dynamics in this system are similar to those observed[3] in Fe_xCr_{1-x}. For both the $x=0.35$ and $x=0.25$ samples, the low-temperature spectra are dominated by a resolution-limited peak centered at $E=0$. The primary effect of increasing temperature on the $x=0.35$ spectra is to reduce the intensity of this peak. In contrast, for the $x=0.25$ sample, resolvable spin-wave peaks are found for $80 < T < 200$ K; the positions of these peaks follow the usual ferromagnetic dispersion relation $E=DQ^2$. As T is lowered below the Curie temperature $T_C=221$ K, D initially increases, as it does in a normal ferromagnet. However, below about 110 K, D begins to decrease dramatically, as in $Fe_{0.24}Cr_{0.76}$.[3] Unfortunately, our instrumental resolution was too coarse to decide whether underdamped modes exist at the lowest temperatures in the $x=0.25$ sample.

We now discuss a possible model for the results presented here. We emphasize that similar empirical behavior has been found in both metallic[2,3] and insulating alloys.[1] Therefore, the underlying physics cannot depend on the detailed nature of the system but must rest primarily on the competing near-neighbor interactions. Our model, which uses concepts from

magnetic percolation theory,[6] was motivated by the computer simulations of Binder *et al.*[7] together with recent theoretical[8] and experimental[9] work on random field effects. For ferromagnets near the FM-SG critical concentration x_c, only a fraction of the spins participate in the ferromagnetism. Thus, by analogy with a diluted magnet near, but above, the percolation threshold, we can divide the system into two parts—the infinite network and finite clusters. In the percolation problem, the finite geometrical clusters are isolated by a closed boundary of vacancies. In this problem, a "magnetic cluster"[7] corresponds to a set of strongly coupled spins with fixed relative orientations at low temperatures but with a very small or vanishing interaction with the ferromagnetic network due to frustration of the exchange bonds around the boundary. The effect of decreasing the concentration of spins having ferromagnetic bonds is to diminish the number of spins in the infinite ferromagnetic network until at the critical concentration, x_c, only finite magnetic clusters remain. We now discuss the consequences of such a model for the $x=0.25$ alloy.

Above $T_C=221$ K the diffuse scattering will originate from a combination of Lorentzian critical scattering from the ferromagnetic network together with broad Lorentzian scattering from the finite clusters. At intermediate temperatures below 221 K, the scattering should be dominated by a sharp elastic peak centered at $Q=0$ and a Q^{-2} diffuse spin-wave contribution. As the temperature is further decreased the finite clusters will freeze—or, equivalently, participate in an SG transition—with cluster spin orientations which will be random with respect to the FM moment. We now make the important assumption that the relaxation times associated with this freezing process are much longer than the domain formation times for the infinite network. In other words, the activation barriers, due to intracluster exchange and intercluster dipolar interactions, that must be overcome to reorient the finite clusters are much larger than those hindering the formation of Bloch walls within the FM network. This assumption is supported by the many observations of hysteresis effects and long relaxation times in the SG and RSG, but not the FM states of random magnetic alloys. The net coupling between the FM network and the frozen clusters then enters the effective Hamiltonian for the infinite network like a *random field.*[8]

This random field, in turn, will destroy long-range order in the ferromagnetic network, causing the decrease in the magnetization with decreasing T observed in the bulk measurements.[4] Furthermore, according to recent theory[8] and experiments,[9] the random field converts the delta-function order parameter scattering at $Q=0$ into diffuse scattering which is predominantly of the form $B/(\kappa^2+Q^2)$.[2] Thus, there should be a dramatic increase in the scattering intensity at finite wave vectors with decreasing temperature, which is what we observe in $(Fe_{0.75}Mn_{0.25})_{75}P_{16}B_6Al_3$ [Fig. 1(c)] and has been seen previously in other systems.[1-3] At low temperatures, the diffuse scattering would then be a combination of Q^{-2} spin-wave scattering and, for random field correlation lengths >100 Å, Q^{-4} "order parameter" scattering. Therefore, we expect $Q^{-\alpha}$ diffuse scattering at low temperatures with the exponent α intermediate between 2 and 4. This is observed in all of the spin-glass systems referred to above[1-3] and in the random antiferromagnets in dc fields, for reduced wave vectors >0.01 Å^{-1}.[9] Presumably, the decrease in the spin-wave stiffness is also caused by the gradual breakup of the infinite network due to the finite cluster random fields.

Similar effects should occur in the pure SG regime. Specifically, we expect that the finite cluster FM correlation length ξ will exhibit a maximum at the "SG transition temperature" with the decrease in ξ with decreasing temperatures precipitated by random field effects. Concomitantly, the finite wave-vector susceptibility will show a peak at about the same temperature. These effects have been identified by SANS measurements in $Eu_{1-x}Sr_xS$[1] and Fe_xCr_{1-x},[3] and are indicated by our own measurements in $(Fe_{0.65}Mn_{0.55})_{75}P_{16}B_6Al_3$.

In summary, for $(Fe_{1-x}Mn_x)_{75}P_{16}B_6Al_3$ near but on the FM side of the FM-SG critical concentration, we observe a gradual evolution with decreasing temperature into an SG-like state. We argue that this behavior can be understood in terms of random field effects. Clearly, further experiments and a proper theoretical formulation of our ideas are required.

After completing this manuscript, we received a report prior to publication[10] which describes a SANS study of amorphous $(Fe_{1-y}Mn_y)_{80}P_{16}C_4$. The results for $y=0.24$ and 0.34 are similar to those described above. Gratifyingly, for samples very near the multicritical concentration on the ferromagnetic side, a secondary maximum was observed at low temperatures for $Q=0.012$ Å^{-1}; we have now obtained similar results in our system with $x=0.30$ and 0.28. This, of course, is anticipated in our model when the infinite network inverse correlation length exceeds ~ 0.01 Å^{-1} due to the random fields.

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