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We have performed neutron scattering studies on two very different alloys which undergo transitions from ferromagnetic (FM) to spin-glass (SG) states as the temperature is reduced. The alloys are $Eu_xSr_{1-x}S$, a crystalline insulator, and $(Fe_xMn_{1-x})_{75}P_{16}B_6Al_3$, an amorphous metal, and their FM-SG multicritical points are at $x \cong 0.50$, $T = 4$ K and $x \cong 0.65$, $T = 42$ K respectively. In spite of the substantial differences between these materials, the neutron scattering data show that their spin correlations are remarkably similar. In particular, for the samples near the multicritical points, a single Lorentzian describes the magnetic scattering very well. Its width κ corresponds to a ferromagnetic correlation length ξ which, as T is reduced, first increases to a value indistinguishable from infinity, and then decreases to a finite value, as expected for a ferromagnet which evolves into a reentrant spin glass. As the Fe or Eu content is raised, the scattering function at low temperatures deviates increasingly from the Lorentzian form, and is better described by a power law $Q^{-\alpha}$ with $2 < \alpha < 3$. We find no evidence for coexistence of ferromagnetic order with freezing of the transverse spin components, as proposed by Gabay and Toulouse. We argue on general grounds that the zero-field Gabay-Toulouse state cannot exist in real magnets. However, our results can be explained in terms of the random field effects which arise when ferromagnetic and spin-glass order parameters are coupled together.

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I. INTRODUCTION

In the simplest spin glasses, ferromagnetic and antiferromagnetic interactions are equally large and equally probable. Under such circumstances, it is not difficult to imagine a transition where a disordered and rapidly relaxing paramagnetic (PM) phase evolves into a frozen, spin-glass (SG) state, which is still disordered. Sherrington and Kirkpatrick[1] predicted that an imbalance between the positive and negative interactions can lead to a transition which is more difficult to visualize. In this case, as the temperature is reduced, the PM phase first evolves into an ordered ferromagnetic (FM) phase, and then into a disordered SG state. Since 1975, when the Sherrington–Kirkpatrick (SK) paper was published, experimentalists have discovered many materials which seem to display this "reentrant" spin-glass (RSG) phenomenon.[2–9] While the existence of such materials is surprising enough, particularly in light of recent amendments to the SK theory,[10,11] the wide variety of materials with RSG behavior is even more striking. Indeed, RSG behavior has been observed in the polycrystalline metals $(Pd_{1-y}Fe_y)_{1-x}Mn_x$[2] and Fe_xCr_{1-x},[5] the amorphous metals $[Fe_x(TM)_{1-x}]_{75}P_{16}B_6Al_3$,[4,6,8] where TM = Ni, Mn, or Cr, the crystalline semiconductors $Eu_xSr_{1-x}S$,[3] and the dilute insulators $KMn_xZn_{1-x}F_4$.[7] The present paper deals with our neutron scattering studies of two very different dilution series, namely $(Fe_xMn_{1-x})_{75}P_{16}B_6Al_3$[8] and $Eu_xSr_{1-x}S$.[3,9] The corresponding magnetic phase diagrams,[3,4] established from bulk measurements and shown in Fig. 1, are very similar. Each of these systems has its advantages. $Eu_xSr_{1-x}S$ is crystalline, and it is therefore possible to study the zero-field magnetization by Bragg diffraction. Furthermore, the microscopic origin of the competing interactions is well-understood: the nonmagnetic Sr^{2+} ions are randomly substituted for magnetic Eu^{2+} ions which interact, as they do in pure EuS, via positive nearest- and negative next-nearest-neighbor couplings. Indeed, computer simulations reveal a breakdown of FM behavior at $x = x_c \cong 0.50$, in agreement with existing bulk measurements.[3]

The microscopic Hamiltonian for $(Fe_xMn_{1-x})_{75}P_{16}B_6Al_3$ is not known. The principal evidence for competing exchange interactions is the phase diagram itself (see Fig. 1). Note that the Fe concentration x_c at the FM-PM-SG multicritical point is ~ 0.65, which is far from a typical percolation threshold in three dimensions. In this sense $(Fe_xMn_{1-x})_{75}P_{16}B_6Al_3$ resembles $Eu_xSr_{1-x}S$ ($x_c = 0.50$, $x_p = 0.13$) more than another much-studied amorphous metal, $(Fe_xNi_{1-x})_{75}P_{16}B_6Al_3$, where $x_c \cong 0.17$.[4,6] Even though it is an amorphous alloy without a simple spin Hamiltonian, $(Fe_xMn_{1-x})_{75}P_{16}B_6Al_3$ remains an interesting system to study because its magnetic energy scale is very large. RSG and SG behavior occurs at convenient temperatures ($T \gtrsim 5$ K), and the spin dynamics can be productively studied using conventional triple-axis neutron spectroscopy.

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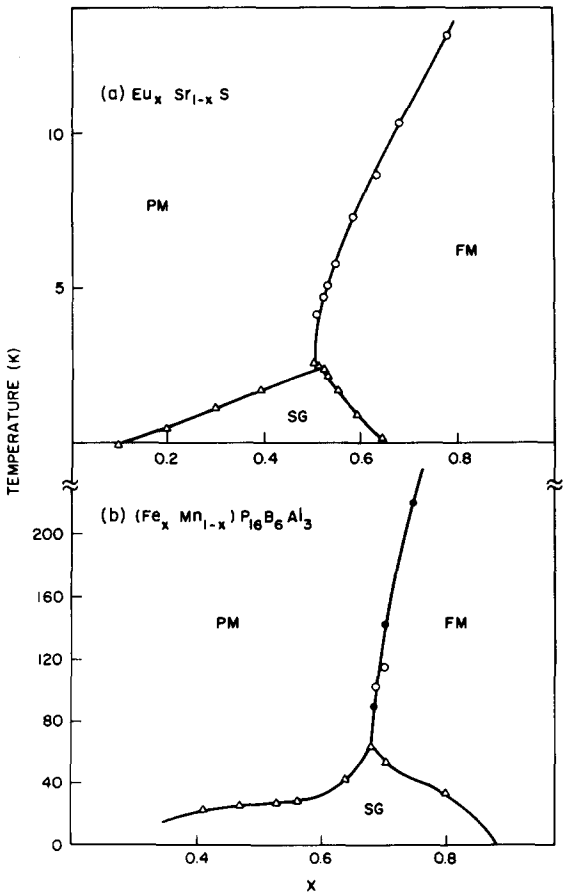

FIG. 1. Magnetic phase diagrams for $Eu_xSr_{1-x}S$ (from Ref. 3) and $(Fe_xMn_{1-x})_{75}P_{16}B_6Al_3$ triangles for $x > 0.65$ and open circles from Geohegan and Bhagat,[4] triangles for $x < 0.65$ from Yesharun *et al.*,[4] closed circles from SANS data.[8]

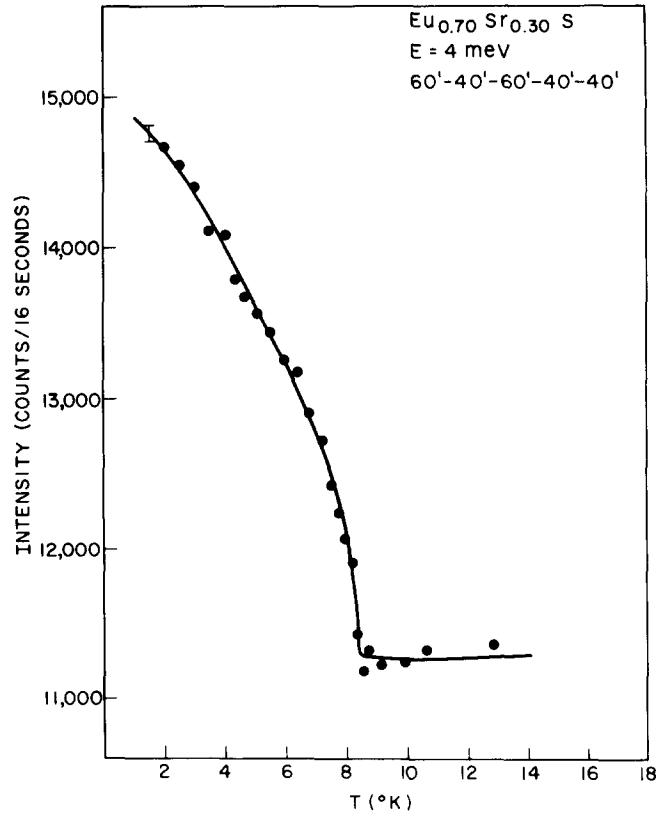

FIG. 2. Temperature dependence of (002) Bragg intensity for $Eu_{0.70}Sr_{0.30}S$.

II. EXPERIMENT

The neutron scattering experiments were performed at the Brookhaven National Laboratory High Flux Beam Reactor. The experimental procedures and samples were as described in Refs. 3 and 8. Inelastic measurements on $(Fe_xMn_{1-x})_{75}P_{16}B_6Al_3$ and quasielastic measurements on $Eu_xSr_{1-x}S$ were carried out using standard triple-axis spectrometers, while quasielastic data on $(Fe_xMn_{1-x})_{75}P_{16}B_6Al_3$ were collected with the small angle scattering spectrometer of the Brookhaven Biology Department.

The single crystals of $Eu_xSr_{1-x}S$ were grown with the ^{153}Eu isotope to limit neutron absorption. Morphologically, they are wafers with approximate dimensions $5\times5\times1$ mm^3; their large faces are (200) planes. We mounted these fcc crystals in a pumped ^3He cryostat so that the $(\bar{1}10)$ zone was in the scattering plane.

The $(Fe_xMn_{1-x})_{75}P_{16}B_6Al_3$ ribbons were prepared by centrifugal quenching from the melt. They differ in no respect from the samples studied using bulk techniques, except for the substitution of ^{11}B for ^{10}B to reduce neutron absorption. We packed approximately 5 g of the ribbon material in cylindrical sample holders, which in turn were mounted in Displex cryostats.

For both $Eu_xSr_{1-x}S$ and $(Fe_xMn_{1-x})_{75}P_{16}B_6Al_3$, there are uncertainties regarding the true concentrations x. We will label our data using concentrations determined from ratios of starting materials. To make comparisons, however, it is best to identify the samples by their Curie temperatures, which vary rapidly with x.

III. RESULTS

We begin with our most Eu-rich $Eu_xSr_{1-x}S$ sample, where $x = 0.70$. The principal characteristic of the data for this sample is magnetic Bragg scattering at the nuclear Bragg points. Figure 2 shows the temperature dependence of the (200) intensity. Note that there is a well-defined onset (at $T_c = 8.3$ K) for magnetic Bragg scattering which *rises* monotonically with decreasing temperature. Furthermore, the magnetic diffuse scattering, measured for nonzero reduced momentum transfers, reaches a maximum for $T = T_c$, and then *decreases* monotonically for $T < T_c$ (see Fig. 3). In other words, there seems to be a quite normal second order phase transition at T_c, characterized by the onset of long range FM order and a diverging susceptibility. At a more quantitative level, the FM phase does have some unusual features, the most obvious being that the magnetic diffuse intensity does not extrapolate to zero as $T\to0$ (see Fig. 3). Instead of dwelling on this issue, however, we turn to a less Eu^{2+}-rich sample, in which there are gross qualitative discrepancies from ordinary FM behavior.

Figure 4 shows the temperature dependence of Bragg and diffuse intensities for $Eu_{0.54}Sr_{0.46}S$. For higher temperatures ($T > 3.75$ K), there is a rise in diffuse intensity, which reaches a plateau at roughly the temperature (~4.3 K) where the nominal Bragg scattering begins to rise. Thus, the data are consistent with a smeared FM transition at 4.3 K. However, in contrast to what happens below the Curie point

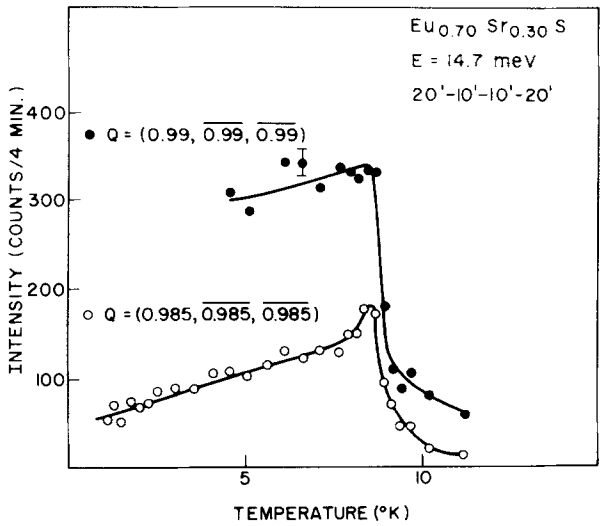

FIG. 3. Temperature dependence of diffuse scattering for slight offsets from (111) Bragg peak.

of an ordinary FM, the diffuse scattering rises again for $T < 3.5$ K, while the (200) intensity *falls*. This is indeed suggestive of the "inverted" FM or RSG transition indicated on the phase diagram [Fig. 1(a)].

Before discussing $Eu_xSr_{1-x}S$ in greater detail, we present constant-q data for $(Fe_xMn_{1-x})_{75}P_{16}B_6Al_3$. For the amorphous alloy, the reduced momentum transfer q is of necessity nonzero and measured with respect to the forward direction. Figure 5 shows our results for two samples which, according to the bulk phase diagram [Fig. 1(b)], should display RSG behavior at low temperatures. For the more iron-rich of the two ($x = 0.70$), there is a well-defined and q-independent maximum in the diffuse scattering at $T_c = 143$ K.

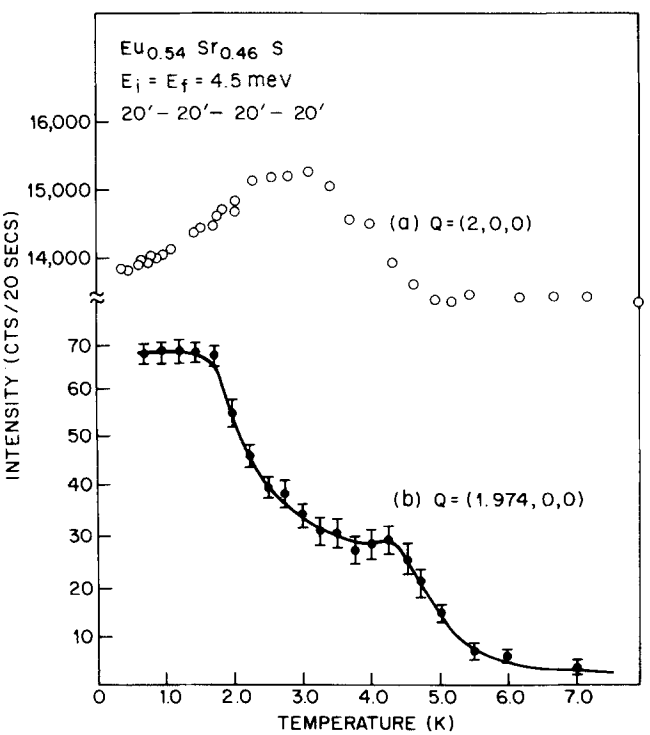

FIG. 4. Temperature dependence of Bragg and diffuse intensities for $Eu_{0.54}Sr_{0.46}S$.

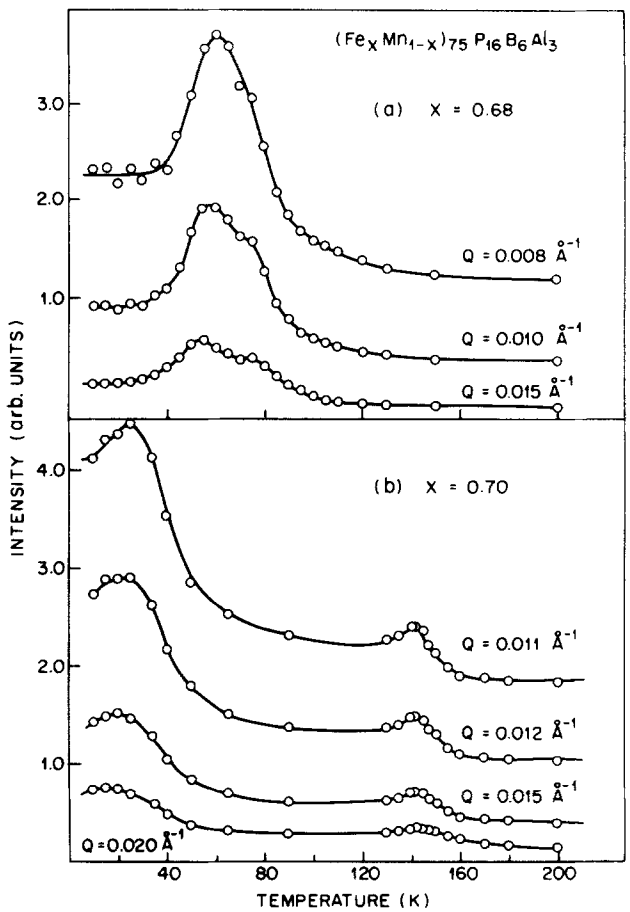

FIG. 5. Temperature dependence of SANS intensity measured at various momentum transfers q for $(Fe_xMn_{1-x})_{75}P_{16}B_6Al_3$ with $x = 0.68$ and 0.70.

We identify this maximum with the FM transition found in the bulk studies. At lower temperatures, we observe deviations from ordinary FM behavior: the diffuse scattering increases again and reaches a second maximum at a temperature $T_l(q)$ which rises with decreasing q. The limiting value T_l ($q = 0$) should be the zero-field RSG transition temperature, if the notion of an "inverted" FM transition is correct. For the more Mn-rich sample (x = 0.68), the upper FM and lower RSG anomalies move towards each other; note that at the smallest q (0.008 Å^{-1}), T_c is very difficult to identify. For $q = 0.008$ Å^{-1}, the maximum is at 60 K, which is indistinguishable from the spin glass transition temperature of 63 K found in the low field magnetization measurements of Geohegan and Bhagat[4] on a sample with the same nominal x.

At a qualitative level, the results that we have shown for $Eu_xSr_{1-x}S$ and $(Fe_xMn_{1-x})_{75}P_{16}B_6Al_3$ are very similar, and also consistent with the phase diagrams reproduced in Fig. 1. Indeed, our data clearly indicate that FM order is reduced, if not eliminated, at low temperatures in the RSG samples. On the other hand, we have no evidence that true FM order, as it exists in, for example, pure EuS, is ever established *at any temperature* in the RSG samples. The remarkable resemblance of the $q = 0.008$ Å^{-1} data for $(Fe_{0.68}Mn_{0.32})_{75}P_{16}B_6Al_3$ and the nominal Bragg scattering for $Eu_{0.54}Sr_{0.46}S$ is particularly germane in this context. Of course, neutron scattering experiments give no direct information concerning length scales larger than those corre-

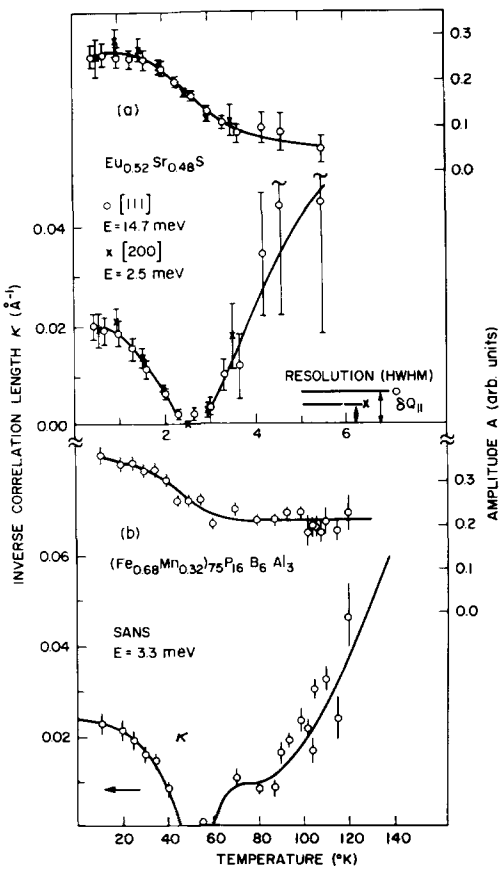

FIG. 6. Temperature dependence of inverse correlation length κ (lefthand scale and lower curves in each frame) and Lorentzian amplitude (righthand scale and upper curves in each frame) for $Eu_{0.54}Sr_{0.46}S$[3] and $(Fe_{0.68}Mn_{0.32})_{75}P_{16}B_6Al_3$.[8]

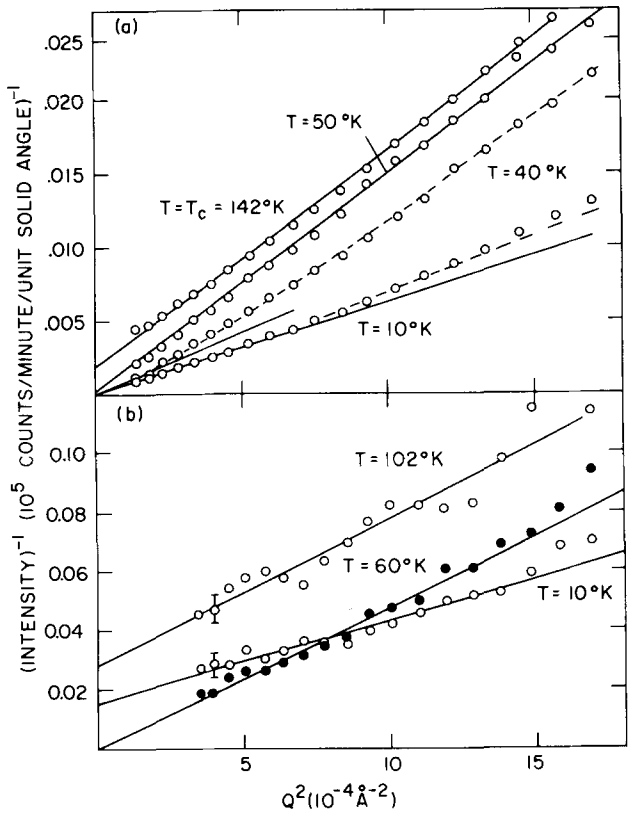

FIG. 7. Inverse SANS intensity vs Q^2 for $(Fe_xMn_{1-x})_{75}P_{16}B_6Al_3$ with (a) $x=0.70$ and (b) $x=0.68$.[8] Background spectra were taken at $T=330$ and 250 K for $x=0.30$ and $x=0.32$, respectively. The sample for which the data in (b) were taken is different from that used to obtain the results shown in Fig. 5(a).

sponding to their q-resolution or low q cutoffs. However, it is possible to establish consistency of the data with models which entail specific large-distance behavior. We have carried out and discussed elsewhere[3] such a detailed study of $Eu_{0.52}Sr_{0.48}S$. The principal finding was that for each temperature, the magnetic scattering at all q (including $q=0$) is very well described by a single Lorentzian,

$$S(q)=\frac{A}{(q^2+\kappa^2)}. \tag{1}$$

Figure 6 shows the temperature dependence of the corresponding amplitudes A and inverse correlation lengths κ. For comparison, Fig. 6(b) gives the results of Lorentzian fits to the SANS data ($q>0.02$ Å^{-1}) for one of our $(Fe_{0.68}Mn_{0.32})_{75}P_{16}B_6Al_3$ samples. Again, the results for the two very different materials are very similar. As the temperature is reduced, κ decreases to a value indistinguishable from zero. At the lowest temperatures, κ increases again; this result together with the lack of discernible magnetic Bragg scattering in $Eu_{0.52}Sr_{0.48}S$ implies that the magnetic correlations in the RSG state are purely short range.

As we move away from the FM-RSG-SG multicritical point, matters become more complicated because the simple Lorenztian form, Eq. (1), no longer describes the data. Figure 7 shows $I^{-1}(Q)$ plotted against Q^2 for $(Fe_xMn_{1-x})_{75}P_{16}B_6Al_3$ with $x=0.68$ and 0.70. On such plots, Lorentzian profiles reduce to straight lines, with slopes A^{-1} and intercepts $-\kappa^2$. As noted above, Eq. (1) describes the $x=0.68$ data very well. However, for $x=0.70$, there are clear deviations from Lorentzian behavior. We find that power law singularities $Q^{-\alpha}$ with $\alpha>2$, as indicated by the dashed lines in Fig. 7(a), give a better account of the data. The exponent α is largest not for the lowest temperatures, but for $T\cong 30$ K. Essentially the same results obtain for $Eu_{0.54}Sr_{0.46}S$.[9] Here we fitted the magnetic scattering to the form

$$S(Q)=\frac{A}{[q^2+\kappa^2]^{\alpha/2}}+B\delta(q). \tag{2}$$

The exponent α reaches its maximum value 2.6 at $T\cong 2.3$ K. While we could not exclude a nonzero magnetic Bragg contribution ($B>0$), we were able to fit all of the data equally well with B fixed at zero as with B allowed to vary. More detailed experiments to address the issue of whether B is ever nonzero for this sample are underway.

We have shown that our RSG samples produce considerable diffuse scattering which does not exist for ordinary ferromagnets. Given that these are Heisenberg systems, it is natural to ask whether the unusual scattering in the RSG regime is simply due to unusual spin wave behavior. Our inelastic neutron scattering study of $(Fe_xMn_{1-x})_{75}P_{16}B_6Al_3$ suggests that this is not the case. Figure 8 shows energy scans taken at $q=0.06$ Å^{-1} for the most Fe-rich ($x=0.80$, $T_C=342\pm 5$ K) sample considered. For all temperatures except 5 K, there are well-resolved spin-wave peaks at finite energy transfers. Below T_C, these peaks move first to higher energies, as they do in ordinary ferromagnets when the mag-

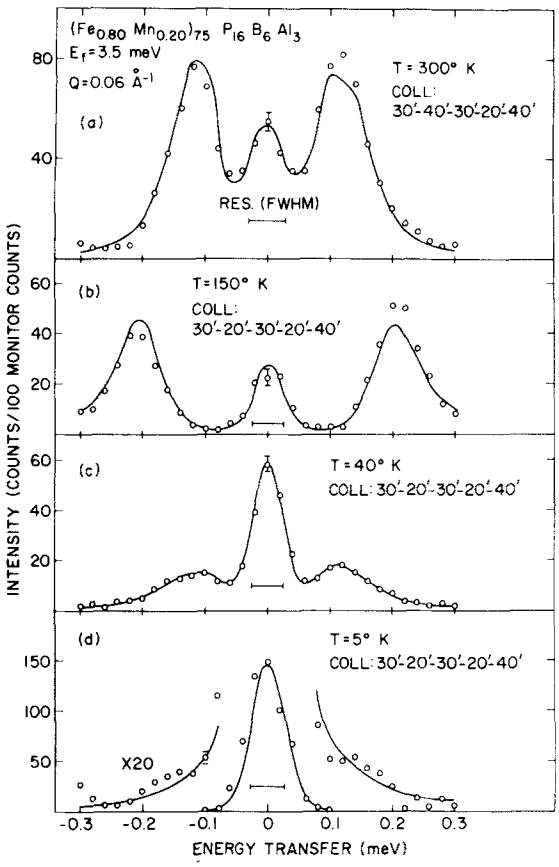

FIG. 8. Evolution with temperature of inelastic spectra for $Q = 0.06$ Å^{-1}.[8] 100 monitor counts correspond to counting times of roughly 30 s for (a), (c), and (d) and 1 min for (b). Solid lines represent results of fits to the sum of a double Lorentzian and an elastic peak.

netization increases. Very peculiar behavior occurs for $T < 150$ K: here the spin wave energy *decreases* with temperature. Furthermore, for $T \leqslant 50$ K, there is increased, resolution-limited quasielastic scattering which coexists with the spin-wave sidebands. We have analyzed our data using a scattering function consisting of an elastic term, with integrated intensity A_G, and an inelastic spin-wave contribution, with integrated intensity A_S. Figure 9 shows the temperature dependence of A_S, A_G and their sum $A_S + A_G = A_Q$,

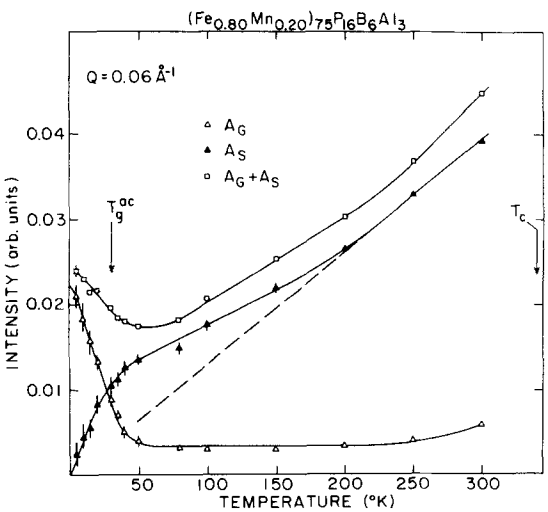

FIG. 9. Temperature dependence of integrated spin-wave intensity A_S, elastic peak amplitude A_G, and net scattering amplitude $A_G + A_S$.[8]

which is what would be measured in a quasielastic scattering experiment. These results indicate that for $T > 50$ K, the deviation from the standard proportionality $A_Q \sim T$ is due to the spin-wave softening, while below 30 K, it is due to the appearance of substantial scattering which is elastic on the scale of the 60 μeV (FWHM) experimental resolution.

IV. DISCUSSION: FRUSTRATION, RANDOM FIELD EFFECTS, AND THE GABAY-TOULOUSE STATE

The experimental results for $Eu_xSr_{1-x}S$ and $(Fe_xMn_{1-x})_{75}P_{16}B_6Al_3$ are very similar to those for many other RSG alloys.[2,4–7] Notable features are the finite FM correlation lengths near the multicritical point, the deviations of the q-dependent scattering profiles from the Lorentzian form, the decline of the magnetic stiffness with temperature, and the appearance of a "central peak" in the low temperature inelastic spectra. Whatever model describes this behavior should be quite general, in view of the wide variety of materials for which RSG behavior has been observed.[2–9]

Infinite-range models[1,10,11] are interesting because they can be analytically tractable, but they do not describe real concentrated spin systems such as $(Fe_xMn_{1-x})_{75}P_{16}B_6Al_3$ and $Eu_xSr_{1-x}S$, where the near-neighbor exchange interactions are random in sign and strong relative to the longer range interactions (e.g., dipolar) present. More useful in this sense are computer simulations by Binder and coworkers,[12] and some calculations by Barahona *et al.*[13] on spin systems with competing short-range interactions. The important result is that in such systems, FM behavior disappears at a critical ratio of positive to negative bonds because there is no longer a single infinite, connected FM network. This is analogous to what happens in ordinary percolating magnets. Above the percolation threshold, a single infinite network of coupled spins coexists with finite clusters of neighboring spins decoupled from the infinite network by virtue of physical separation. In the mixed exchange problem, the finite clusters are "decoupled" because of frustration. Should these clusters freeze, as they do on the SG side of the phase diagram for the alloys in question, they will impose a random field on the FM network. Random fields arise similarly in percolation-type RSG alloys, such as Fe_xCr_{1-x}[5] and $(Fe_xNi_{1-x})_{75}P_{16}B_6Al_3$.[6] Here, the FM network is simply the percolating network of magnetically active ions. The relatively weak, but longer range RKKY and dipolar interactions then account for the SG behavior among the spins in the finite clusters, and the associated random field acting on the FM network.

The principal consequences of the coupling between FM and SG order parameters are as follows. First, in short-range coupled, three-dimensional Heisenberg systems, the imposition of an infinitesimal random field destroys long-range magnetic order.[14] Hence, as the "decoupled" spins freeze—much as they do in ordinary spin glass—at some characteristic temperature T_g below the Curie point of the RSG alloy, the net FM moment will vanish. Concomitantly, the spin-wave lifetimes and stiffness will be reduced as T is decreased towards T_g. For $T < T_g$, the inverse FM correlation length will grow, as we have observed in our experi-

ments (see Fig. 6).

It has been shown[14] that random fields transform magnetic Bragg peaks into diffuse profiles of the form

$$S(q) = \frac{a}{q^2 + \kappa^2} + \frac{b}{(q^2 + \kappa^2)^2}. \quad (3)$$

For static random fields, the Lorentzian squared portion of $S(q)$ represents strictly elastic scattering. Over relatively wide ranges of q, (3) is difficult to distinguish from the power law form $S(q) \sim q^{-\alpha}$ with $2 < \alpha < 4$. Our data for $S(q)$, as well as that collected by many other groups, do indeed deviate in this manner from Lorentzian behavior. Thus, the random fields produced by the freezing "decoupled" clusters account for the enhanced quasielastic neutron scattering characteristic of RSG alloys.

Very recently, Gabay and Toulouse[11] (GT) have proposed a phase diagram for the infinite range Heisenberg model with random exchange interactions. They found that with decreasing temperature, the FM phase could evolve into a state where a net moment **M** coexists with SG order among the spin components transverse to **M**. However, it could not transform into an ordinary SG state with vanishing **M**, as suggested by Sherrington and Kirkpatrick.[1] In a neutron scattering experiment, such a state would be characterized by (1) resolution limited magnetic Bragg scattering, with intensity proportional to $|\mathbf{M}|^2$, and (2) elastic diffuse scattering, corresponding to the frozen transverse spin degrees of freedom, whose intensity would rise with decreasing T for *all* momentum transfers q. Our results clearly cannot be interpreted within this picture because (1) we find *no magnetic Bragg scattering* in the RSG state, and (2) there is very substantial elastic diffuse scattering whose intensity at small q actually decreases after the RSG phase is entered. However, it is worth noting that in the GT state, off-diagonal (in spin space) interactions will couple the randomly frozen transverse and ferromagnetically aligned parallel spin components. Thus, there will be an effective random field acting to reduce **M**. The consequences for real three-dimensional systems would be very similar to those of the model described above. In particular, long range FM order would be destroyed, which means that $\mathbf{M} = 0$. Dipolar interactions, present in all real magnets, lead to off-diagonal coupling, and consequently the *zero-field* GT state cannot exist in practice. Indeed, to the best of our knowledge, there is no experimental evidence for such a state in any compound. Contrary to assertions in the literature,[15] the observation of well-defined spin waves at finite wave-vectors does not demonstrate the existence of long-range FM order. Of course, systems with relatively long-range (e.g., RKKY) interactions between spins, when subjected to uniform fields with magnitudes exceeding typical internal random field amplitudes, could very well behave in a manner consistent with the GT solution.

In summary, we have shown that two very different random alloys exhibit very similar magnetic behavior. Notably, both are characterized by substantial diffuse scattering with an unusual power law dependence on momentum transfer. In the crystalline system $Eu_xSr_{1-x}S$ we find no evidence for magnetic Bragg scattering in the reentrant spin glass state. This means that in this compound, the *zero-field* Gabay-Toulouse state does not exist. We argue on general grounds that this state cannot exist in real materials. A model[8] based on frustration decoupling and random field effects accounts for all of our data and similar observations on other RSG systems.

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