Spin stiffness anomaly in the reentrant spin-glass $(Fe_{0.25}Ni_{0.75})_{0.75}P_{0.16}B_{0.06}Al_{0.03}$

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Low field magnetization measurements have shown that the amorphous alloy $(Fe_{0.25}Ni_{0.75})_{0.75}P_{0.16}B_{0.06}Al_{0.03}$ becomes a demagnetization-limited ferromagnet below a Curie temperature T_c of 200 K, but that the spontaneous magnetization vanishes again below a reentrant temperature T_R of 4 K. Inelastic neutron scattering measurements have been performed to study the spin-wave dynamics as the spin-glass state is entered from the ferromagnetic state. The observed spin-wave peak positions decrease in energy and the linewidths increase as the temperature is lowered towards T_R. A detailed analysis based on both the double Lorentzian and damped harmonic oscillator forms of the spectral weight function shows that the spin-wave energies strongly decrease as the temperature is lowered and the excitations become heavily damped over the range of wave vectors ($0.04\ \text{Å}^{-1} < q < 0.18\ \text{Å}^{-1}$) studied. An elastic component of the magnetic scattering, attributed to a spin-glass order parameter, is also observed.

In the metallic glass system $(Fe_xNi_{1-x})_{0.75}P_{0.16}B_{0.06}Al_{0.03}$, the Curie temperature T_c for the ferromagnetic phase transition in these alloys decreases with decreasing x until for $x \leqslant 0.16$ the system exhibits only a spin-glass transition.[1] For intermediate concentrations $0.16 < x < 0.40$, the ferromagnetic phase is accompanied at lower temperatures by spin-glass behavior,[2] which includes the disappearance of ferromagnetism below a reentrant transition temperature T_R.[1] The reentrant temperature increases with decreasing x until the multicritical point at $x = 0.16$ is reached. We have employed inelastic neutron scattering techniques to study the spin dynamics in this system[2] as the spin-glass state is approached from the ferromagnetic state for the alloy with $x = 0.25$. This value of x is near the lower limit in concentration for the feasibility of studying spin waves in this system with presently available instrumental resolution and intensity.

Examples of the observed neutron scattering spectra are shown in Fig. 1 at a fixed wave vector transfer $q = 0.08$ Å^{-1} measured from the forward beam direction (000). The temperature-independent (nonmagnetic) scattering consists of two components, an energy independent intrinsic background which has been subtracted from these data, and an elastic peak at $E = 0$ due to nuclear incoherent and small-angle nuclear scattering from the sample and experimental apparatus. This elastic component has not been subtracted from the data shown in Fig. 1. The temperature-dependent (magnetic) scattering consists of an elastic component ($E = 0$) related to the magnetic correlations which are static on the time scale of these measurements, and inelastic magnetic scattering. The centroids of the scattering for the creation ($E > 0$) and destruction ($E < 0$) of magnetic excitations clearly move to smaller energies as the temperature is lowered towards T_R, and the width of the scattering distribution increases as well.

In order to analyze these data we have least-squares fit them to a spectral weight function plus a central elastic component convoluted with the instrumental resolution func-

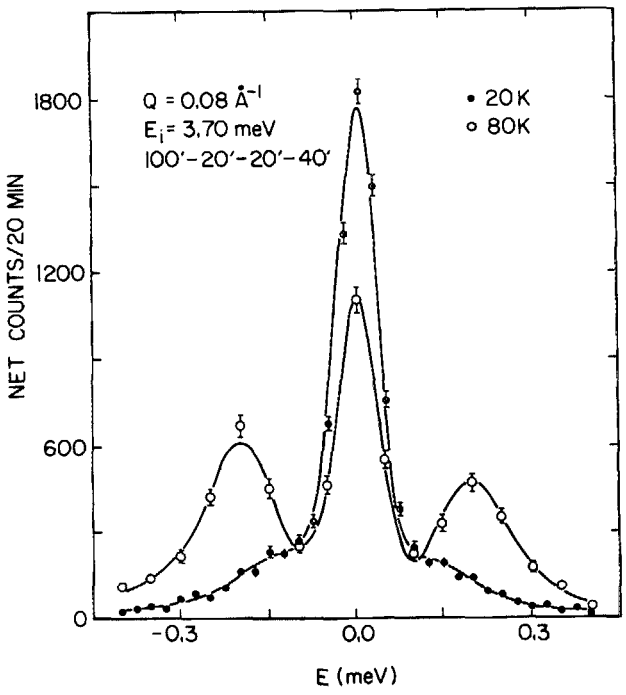

FIG. 1. Constant-q spectra at $q = 0.08$ Å^{-1} showing two components of the temperature-dependent magnetic scattering. Below 80 K, the resolution limited elastic component ($E = 0$) increases above the nonmagnetic background while the inelastic spin-wave scattering on either side of $E = 0$ shifts to lower energies and broadens.

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tion. Two forms of the spectral weight function have been employed, the double Lorentzian,

$$F_{\mathrm{LOR}}(q,E)=\frac{1}{\pi}\left(\frac{\Gamma_q}{(E-E_q)^2+\Gamma_q^2}+\frac{\Gamma_q}{(E+E_q)^2+\Gamma_q^2}\right) \tag{1}$$

and the damped harmonic oscillator,

$$F_{\mathrm{DHO}}(q,E)=\frac{1}{\pi}\frac{2\Gamma_q E_q^2}{(E^2-E_q^2)^2+(2\Gamma_q E)^2}. \tag{2}$$

These expressions have the same limiting form as $\Gamma_q/E_q\to 0$, where Γ_q is the half width at half maximum, but they differ substantially in shape when Γ_q and E_q are comparable since the harmonic oscillator form has a larger weight at small E than the double Lorentzian. We remark that in the regime where Γ_q/E_q is comparable to or larger than unity care must be exercised in interpreting the parameters for the DHO function. Note, in particular, that in the limit $\Gamma_q/E_q\to\infty$ the harmonic oscillator function approaches the unphysical form of a delta function $\delta(E)$ while the double Lorentzian becomes a single Lorentzian of half width Γ_q centered at $E=0$. Thus, it is physically impossible for the bare spin-wave energy E_q in Eq. (2) to approach zero, while from general theoretical considerations we expect the spin-wave energies to renormalize to zero at a second-order phase transition. We suggest that this unphysical behavior of the DHO function arises from an incorrect interpretation of the parameter E_q, and that the relevant excitation energy should rather be $\tilde{E}_q=(E_q^2-\Gamma_q^2)^{1/2}$. The excitation energy, as represented by the real part of the poles of the spectral weight function (and reflected in the temporal Fourier transform) is explicitly displayed when Eq. (2) is rewritten in terms of $\tilde{E}_q$:

$$F_{\mathrm{DHO}}(q,E)=\frac{1}{\pi}\frac{2\Gamma_q(\tilde{E}_q^2+\Gamma_q^2)}{[(E-\tilde{E}_q)^2+\Gamma_q^2][(E+\tilde{E}_q)^2+\Gamma_q^2]}. \tag{3}$$

Now the limit $\Gamma_q/\tilde{E}_q\to\infty$ is well defined, yielding a squared Lorentzian of half-width $(\sqrt{2}-1)^{1/2}\Gamma_q$ centered at $E=0$. Thus, the renormalization of $\tilde{E}_q$ to zero will result in spin diffusion, with the time dependence of the two-spin correlation function given by $(1+\gamma_q t)\exp(-\gamma_q t)$ with $\Gamma_q=\hbar\gamma_q$.

In the temperature range between T_c and T_R there is a substantial wave vector range over which elementary magnetic excitations are observed. The fits for the spin-wave energies in this case are reasonably well described by the usual quadratic dispersion relation of the form

$$E_q=Dq^2+\Delta, \tag{4}$$

with D the spin-wave stiffness parameter and Δ a small (dipole) energy gap. The value determined for Δ at 80 K is 0.015 ± 0.006 meV although possible systematic errors preclude an accurate determination of this very small energy gap. The results for the temperature dependence of E_q, $\tilde{E}_q$, and Γ_q are shown in Fig. 2. The behavior of the spin-wave energies (E_q for the Lorentzian, and $\tilde{E}_q$ for the DHO) are qualitatively quite similar. After increasing in energy with decreasing temperature below T_c in the usual manner, these excitation energies begin to decrease again below about 80 K. The intrinsic linewidth also increases again below ~ 80 K. As $T\to T_R$ we find that $\Gamma_q/E_q\to 1$ which for the DHO function would imply that $\tilde{E}_q\to 0$. The data are not of sufficient quality, though, to determine whether or not these excitation energies renormalize completely to zero. We remark that the decrease of spin-wave energies with the magnetization as either phase boundary of the ferromagnetic state is approached is expected from both mean-field and hydrodynamic theories[3–6] since these two quantities are directly related in the FM phase. Of course strictly speaking only the $q\to 0$ spin-wave modes are required to renormalize completely to zero energy at a second-order phase transition while finite energies may still be observed at finite q.

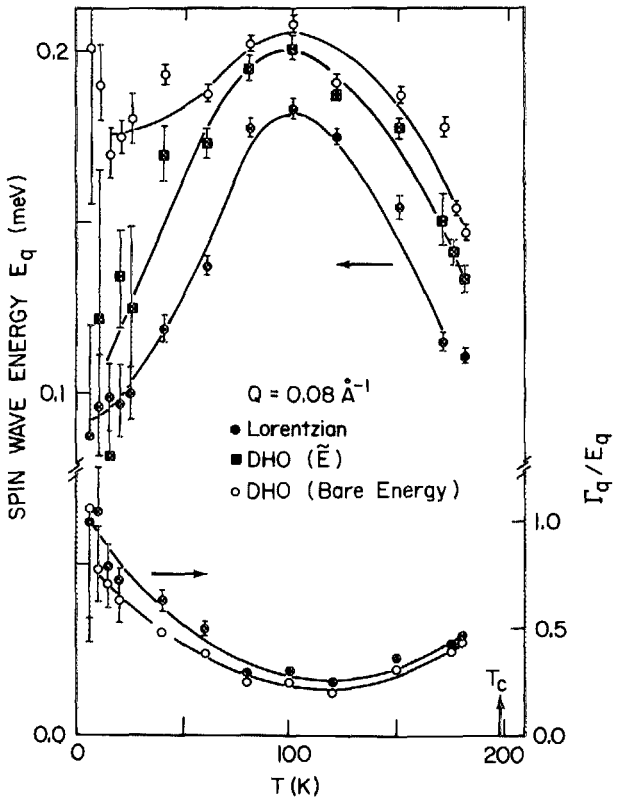

FIG. 2. Temperature dependence of the spin-wave energy (E_q for the double Lorentzian, $\tilde{E}_q$ for DHO) and the damping ratio Γ_q/E_q, determined from the convolution fits. The excitation energies decrease as the temperature is lowered towards the reentrant spin-glass temperature ($T_R=10$ K) and Γ_q approaches critical damping ($\Gamma_q/E_q=1$). Also shown is the parameter E_q for the DHO, which becomes unphysical when Γ_q/E_q becomes comparable to unity.

Results for the temperature dependence of the resolution-limited elastic component of the scattering as determined from the fits are shown in Fig. 3. The magnetic part of this intensity, if it is truly elastic, should be directly related to the spin-glass order parameter, which we can write as $q_{\mathrm{EA}}-m^2$ where $q_{\mathrm{EA}}=N^{-1}\Sigma\langle \mathbf{S}_i\rangle^2$ is the generalized Edwards–Anderson[7] parameter for N spin variables, $\mathbf{S}_i$, and $m^2=(N^{-1}\Sigma\langle S_i^z\rangle)^2$ is the square of the site-averaged magnetization. Note that $q_{\mathrm{EA}}-m^2$ is a nonnegative quantity. The data show an increase of the elastic scattering above the nuclear background for temperatures below about 80 K, in good agreement with our inelastic measurements and the low field magnetization measurements which demonstrate that m begins to decrease at this same temperature.[1] Thus,

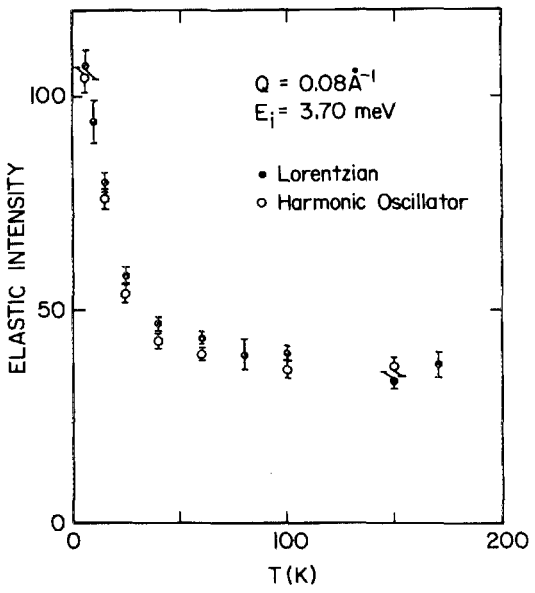

FIG. 3. Temperature dependence of the elastic component of the neutron scattering determined from the convolution fits. The magnetic part of this intensity represented by the increase below about 80 K can be attributed to a spin-glass order parameter.

our results are consistent with the assumption that spin-glass order coexists with ferromagnetism over a limited temperature range, but is detrimental to the ferromagnetic state and eventually destroys it.

The decrease of the observed spin-wave peak positions and concomitant increase of their linewidths at low temperatures has also been observed in the related systems $(Fe_{1-x}Mn_x)_{0.75}P_{0.16}B_{0.06}Al_{0.03}$,[8] Fe_xCr_{1-x},[9] Fe_xAl_{1-x},[10] $Au_{1-x}Fe_x$,[11] and NiMn,[12] with varied interpretations of the results. In Refs. 11 and 12, the authors argue against the breakup of ferromagnetism on the basis of the temperature dependence of the parameter E_q in the DHO function which shows little or no softening at the reentrant temperature. However, we have shown that the correct spin-wave energies $\tilde{E}_q$ have a temperature dependence which is very similar to that given by the double Lorentzian form for the spectral weight function. Thus, both forms of $F(q,E)$ yield the same physical behavior when properly interpreted. We conclude that the present measurements of the dynamics in a reentrant spin glass are consistent with a picture in which long-range ferromagnetic order is destroyed at a reentrant phase transition, and in which ferromagnetism may coexist with spin-glass order in a limited temperature range.

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