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EVOLUTION FROM FERROMAGNETISM TO SPIN-GLASS BEHAVIOR

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The magnetic properties of amorphous $(Fe_xNi_{100-x})_{75}P_{16}B_6Al_3$ have been investigated in the critical concentration region ($x_c \approx 17$) via neutron scattering. For $x>x_c$ a transition to ferromagnetism is first observed followed at lower temperatures by an evolution to spin-glass behavior. Associated with this low temperature state are two distinct time scales corresponding to a resolution-limited elastic peak and to excitations. The temperature dependence of the elastic component is directly related to the spin-glass order parameter and indicates that there is a region of temperature where ferromagnetism and spin-glass order coexist.

Randomized systems near the critical concentration where long range order develops have been the focus of considerable attention in recent years because of the rich variety of physical phenomena these systems exhibit. Examples of such systems include physisorbed and chemisorbed molecules on surfaces, spinodal decomposition in alloys, intercalated compounds, and substitutional magnetic alloys. Of these systems, random magnetic alloys are particularly well suited for study since the essential physical quantities of interest, such as the order parameter, static pair correlation function and dynamic response function, can be directly determined by neutron scattering without the application of a conjugate (magnetic) field. The magnetic behavior of such random substitutional magnetic systems can be generally catagorized as either dominated by percolation effects, or competing interactions (frustration). A percolative system has relatively simple (usually short range) magnetic interactions which lead to a behavior controlled by dilution, whereas frustration can lead to more complicated behavior as a function of temperature and concentration. In particular, theory indicates[1,2] that under suitable conditions a paramagnetic to ferromagnetic transition can occur near the threshold for long range order to develop, followed at lower temperatures by a transition to a spin-glass state. Whether this spin-glass state is a true thermodynamic phase as formulated by Edwards and Anderson[3] or represents a gradual freezing is still an open question, but there are a number of systems which appear experimentally to display this ferromagnetic to spin-glass "transition".[4-10]

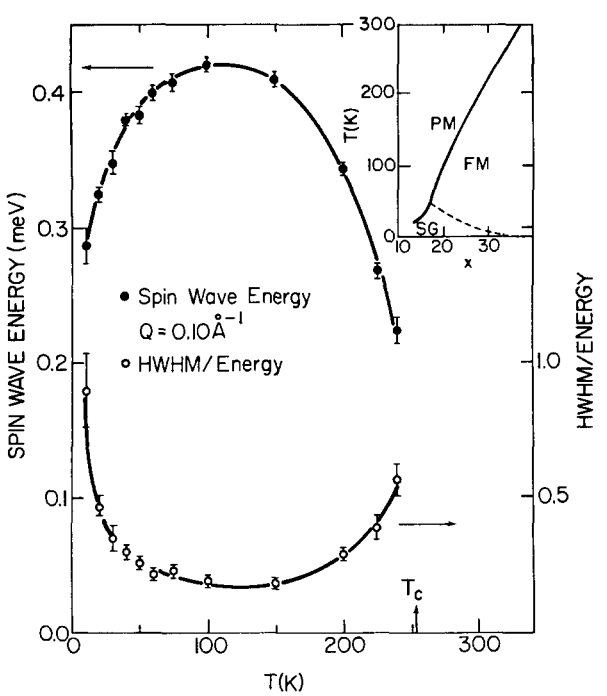
The new feature we find in our neutron scattering data is that at low temperatures there are two distinct time scales present in these systems. The long time scale (elastic) component is a direct measure of the spin-glass order parameter. The temperature dependence of this component yields a spin-freezing temperature which is well above the reentrant ferromagnetic transition T_R measured by low field susceptibility[9,11], indicating that there is a coexistent temperature regime.

To study these phenomena we have chosen to investigate the metallic glass $(Fe_xNi_{100-x})_{75}P_{16}B_6Al_3$. The magnetic phase diagram for this system, as determined by low-field susceptibility, magnetization and hysteresis loop measurements,[9,11] is shown in the inset to Fig. 1. Though this system is structurally amorphous, magnetically it is relatively simple. At high iron concentrations (x>40) it is a prototypical three-dimensional isotropic ferromagnet[12,13] with a characteristic quadratic spin-wave dispersion relation $E=Dq^2$ at small q. Here D is a temperature dependent constant of proportionality which contains the details of the system such as the average exchange interaction. With the substitution of nickel, which is nonmagnetic in this system, the transition temperature and D are reduced until at sufficiently low iron concentrations ($x < 17$) a ferromagnetic state can no longer be supported and a spin-freezing phenomenon, presumably due to frustration effects, is observed. In the intermediate concentration range ($17 < x < 40$) a paramagnetic-to-ferromagnetic transition first occurs with decreasing temperature, followed by a spin-freezing phenomenon at lower temperatures.

The experiments were carried out utilizing the triple-axis neutron spectrometers at the National Bureau of Standards Research Reactor. Typically the incident energy was held fixed at

3.7, 4.9 or 14.8 meV, and the collimations before and after the pyrolytic graphite monochromator and analyzer were either 10', 20' or 40' (FWHM) depending on intensity and resolution considerations. The samples were prepared[14] by the melt-spun quench-cooled technique, and were ~ 9 gm in mass. Iron concentrations of x = 15,20,30 and 40 were investigated, which span the critical concentration of x_c = 17. For $x < x_c$ only spin-glass behavior is observed, while for x = 40 a small anomalous decrease in the spin-wave stiffness parameter D is found at low temperatures.[13,15] We find that the x = 20 and x = 30 alloys both show the same qualitative behavior, that is a transition (T_C) from paramagnetism to ferromagnetism followed at lower temperatures by an evolution to behavior characteristic of a spin glass.

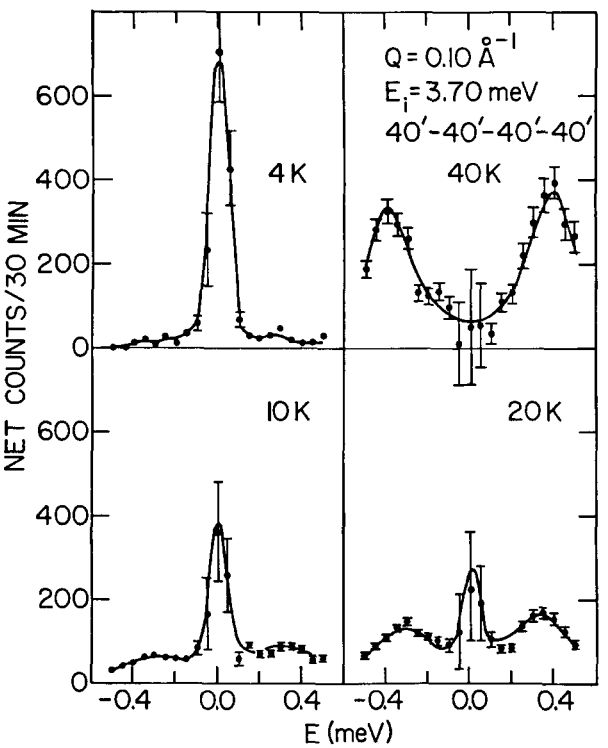
Since the x = 30 concentration is experimentally more favorable for us, we will focus our attention on this alloy. In the ferromagnetic regime (T_C = 255 K) we observe spin-wave excitations, with a D that first increases with decreasing temperature but then decreases as the spin-glass state is approached. The decrease in the spin-wave energy E in the ferromagnetic state is accompanied by an increase in the intrinsic linewidth Γ such that Γ/E exceeds one just below 10 K as shown in Fig. 1.



1. Spin wave energy and (intrinsic half-width)/energy as a function of temperature for the x=30 alloy. With decreasing temperature the energy and lifetime increase, but then decrease at lower temperatures as the spin-glass state is approached. Note that the spin waves have an intrinsic linewidth at all temperatures. The solid curves are a guide to the eye. The inset shows the phase diagram for the system as determined by low-field susceptibility and magnetization measurements.[9]

The low-field susceptibility data on the identical sample establish that the reentrant ferromagnetic transition temperature T_R is 8 K. Thus our inelastic scattering data are consistent with the measured reentrant transition.

The temperature evolution of the magnetic scattering is shown in Fig. 2 as a function of energy for a wave vector of 0.10 $Å^{-1}$. At relatively low temperatures, but still in the ferromagnetic regime, we observe spin waves in neutron energy gain (E<0) and energy loss (E>0). With decreasing temperature these excitations continue to shift to lower energy (in contrast to the conventional behavior) and broaden. The data at 10 K show that the spin waves are very broad; this is the lowest temperature that we could observe spin waves at this Q. Similar behavior has been observed recently[16] in the crystalline system $Cr_{100-x}Fe_x$. These results for the spin dynamics (of both systems) suggest that the spin-wave spectrum collapses as the ferromagnetism is lost, indicating that a phase transition has indeed taken place. However, establishing from the dynamics that a transition has occurred is not straightforward because of the ambiguities introduced by the choice of the spectral weight function used to analyze the data.[17] We used a double-Lorentzian form for the cross section, which we found to give the best



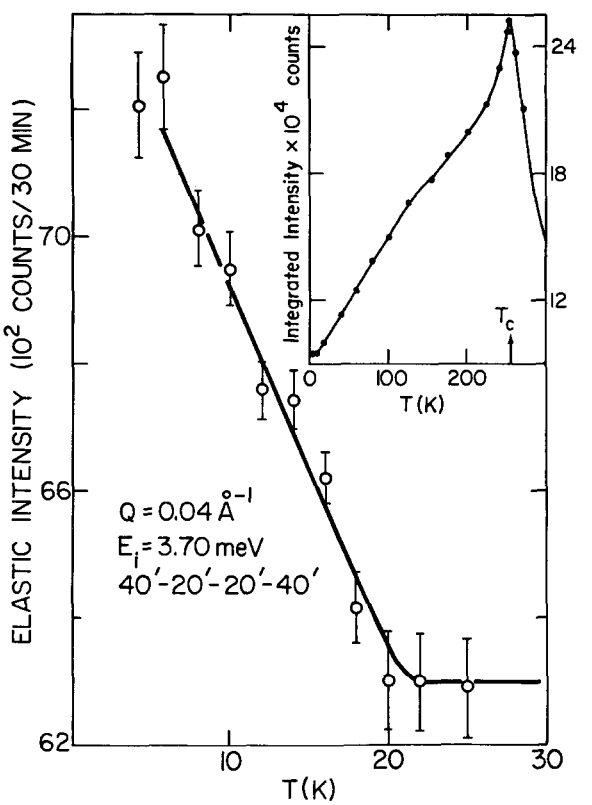
2. Energy dependence of the magnetic scattering as a function of temperature. The spin wave excitations observed in energy gain (E<0) and energy loss (E>0) in the ferromagnetic phase decrease in energy and broaden with decreasing temperature. At low temperatures the scattering intensity is dominated by an elastic (resolution limited) component, plus a broad inelastic component.

fit. The parameter Γ used above is the half-width of the Lorentzian obtained from the data, after correction for the instrumental resolution.

At low temperatures the scattering is characterized by two time scales, evidenced by a broad inelastic component, and a (resolution limited) elastic component. The upper limit to any intrinsic width to the central component is established at 20 μeV HWHM. The temperature dependence of the elastic component to the scattering is shown in Fig. 3 for a wave vector of 0.04 $Å^{-1}$. These data were obtained by setting the spectrometer for the elastic scattering configuration, so that experimentally we measure the truly elastic scattering, plus any inelastic component to the scattering which falls within the resolution "window" of the spectrometer (which is a Gaussian centered at E=0, with a FWHM of 64 μeV). The increase in the scattering at low temperatures is due primarily to the elastic component as the spin-glass state develops. We remark that if the system were an ideal Edwards-Anderson spin glass, then the intensity of this elastic scattering would be independent of the wave vector and thus would be a direct measure of the order parameter in the spin-glass phase. In these relatively concentrated systems, however, we find that there are short-range (ferromagnetic) correlations between the spins, which introduce a wave vector dependence to the elastic scattering. If these (frozen) spatial correlations are temperature dependent (e.g. the size of an average magnetic cluster is a function of temperature) then the intensity of the scattering at any particular value of the wave vector Q would be a product of the order parameter and the (static) correlation function, and we are not able to seperate these two contributions at present. Nevertheless the relatively sharp onset of the elastic scattering is suggestive of the occurrence of a phase transition. The freezing temperature T_f we obtain from such data is not dependent on Q, and is well above the reentrant ferromagnetic transition of 8 K determined by susceptibility measurements. We interpret these results as indicating that the spin-glass state develops before the ferromagnetism is completely destroyed, so that the two phenomena coexist over a region of temperature. This interpretation is consistent with our inelastic scattering measurements (Fig. 1 and 2) which show that the ferromagnetic spin-wave excitations exist well below 20 K, and become overdamped around $T_R \sim 8$ K rather than $T_f \sim 20$ K. The nature of the inelastic scattering below 8 K is not completely clear at present and is under further study.

The increase in the elastic magnetic scattering contrasts with the total (energy integrated) scattering, which was found to monotonically decrease with decreasing temperature below T_C as shown in the inset to Fig. 3. These data taken together indicate that the scattering is shifting from ferromagnetic to spin-glass behavior. In particular the scattering we observe cannot be explained as due to separate groups of clusters freezing, with the ferromagnetic "backbone" persisting unperturbed.

Finally we remark that the presence of an elastic component to the scattering is an essential characteristic of an order parameter. Truly elastic scattering [δ(E)] signifies that the associated parameter has an infinite lifetime, i.e. that an ordered state exists. For a conventional magnetic state such as a crystalline ferromagnet or antiferromagnet we have spin-coherence in space as well as time, and this spatial coherence gives rise to a delta function in wave vector [δ(Q −τ)] as well as energy, that is to Bragg peaks located at the reciprocal lattice vectors τ. For a spin-glass system there is no long range spatial coherence, so we only have a delta function in energy. An important question with regard to spin-glass systems is whether this central component to the scattering really is elastic, or only quasielastic with a width which is smaller than the instrumental resolution. Experimentally we can of course only put an upper limit to any intrinsic linewidth, but from a pragmatic viewpoint there are clearly two time scales in the problem. This behavior is typical of the spin-glass state.[18]



3. Temperature dependence of the elastic component of the scattering, which is related to the spin-glass order parameter. The solid curve is a guide to the eye. The freezing temperature obtained from these data (T_f = 20 K) is well above the reentrant ferromagnetic transition (T_R = 8 K), indicating that the ferromagnetic and spin-glass states coexist in the intervening temperature interval. The inset shows the temperature dependence of the total (energy integrated) scattering.

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