Spin glass-ferromagnetic phase transition in amorphous $(Fe_xNi_{1-x})_{0.75}P_{0.16}B_{0.06}Al_{0.03}$

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Inelastic neutron scattering studies have been carried out on the magnetically isotropic amorphous system $(Fe_xNi_{1-x})_{0.75}P_{0.16}B_{0.06}Al_{0.03}$ in the concentration range $(x>0.17)$ where both ferromagnetic and spin-glass behavior are observed. For $x=0.4$, well defined spin waves which obey the customary quadratic dispersion relation $E=DQ^2$ are observed below the Curie temperature ($T_c=365K$) and D is found to increase with decreasing temperature as usual. Below ~80K, however, D begins to *decrease* with decreasing temperature as the spin-glass state is approached. Accompanied by this decrease in D is an increase in the spin wave linewidths. Preliminary results for $x=0.3$ and $x=0.2$ show qualitatively similar behavior.

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INTRODUCTION

The structurally amorphous metal-metalloid system $(Fe_xNi_{1-x})_{.75}P_{.16}B_{.06}Al_{.03}$ has been found to be a prototype three dimensional isotropic ferromagnet at high iron concentrations. This behavior is reflected in the extremely low anisotropy observed in this system, in the bulk magnetization and in the spin dynamics (1-5). To a first approximation the nickel behaves simply as a magnetic diluent, so that there is only one magnetic species in the system. The initial effect of dilution is to decrease the specific magnetization, the Curie temperature T_c and the spin wave stiffness coefficient D. At sufficiently low concentrations, though, the system will be no longer capable of supporting a cooperative long range magnetic state. The demise of the ferromagnetism may be due to competing exchange interactions, which "frustrate" the system, or to percolation behavior. Recent spin resonance and susceptibility measurements (6,7) show that for iron concentrations below $x=0.17$ some kind of spin freezing phenomenon takes place, while for $x>0.17$ the system first orders ferromagnetically and then freezes at lower temperatures. Setting aside the delicate question of whether this spin freezing corresponds to a genuine thermodynamic transition, we will nevertheless refer to this region as the spin-glass phase. Our aim in the present investigation is to use inelastic neutron scattering to study the energetics of the system as we proceed from the paramagnetic to the ferromagnetic and finally to the spin glass state.

The characteristic feature of the long wavelength excitation spectrum of a uniform isotropic ferromagnet (8,9) is a collective excitation whose energy E varies as the square of the momentum Q:

$$E = DQ^2 \quad (1)$$

The isotropy of the system (10) demands that $E\to0$ as $Q\to0$ (Goldstone theorem (11)). Linear spin wave theory has in fact been found to give a good description of the magnetic properties of the ferromagnetic phase at high Fe concentrations (5).

The parameter D in Eq. 1 contains the details of the system such as the exchange interaction J, average number of neighbors, etc. One can qualitatively include the temperature dependence of the excitation spectrum by using mean field theory to renormalize the exchange interaction:

$$D \propto J \langle S_z\rangle \propto \langle M_z\rangle. \quad (2)$$

Thus D attains its maximum value at $T=0$ and monotonically falls to zero as T approaches T_c. This qualitative behavior is observed for all the conventional isotropic ferromagnets, both for structurally amorphous systems (3-5,12) as well as crystalline magnets such as iron (13), cobalt (14) and nickel (15).

The spin-glass (16) or mictomagnetic state, on the other hand, is characterized by $\langle M_z\rangle=0$ if the average is taken over a sufficiently large region. On the basis of Eq. 2 one can anticipate that the approach to the spin glass state will result in a decrease in D, and this indeed has been found to be the situation in the present case of interest. Similar results on the crystalline Fe_xCr_{1-x} system have also been reported very recently (17).

RESULTS

The neutron scattering measurements were carried out using the constant-Q mode on the triple-axis neutron spectrometers at the National Bureau of Standards research reactor. Pyrolytic graphite monochromator and analyzer crystals were used, along with Soller slit collimators of 10' or 20' FWHM as needed both before and after each crystal. The spin wave data have been corrected for the effects of finite horizontal and vertical instrumental resolution by convoluting Eq. 1 with the measured spectrometer resolution func-

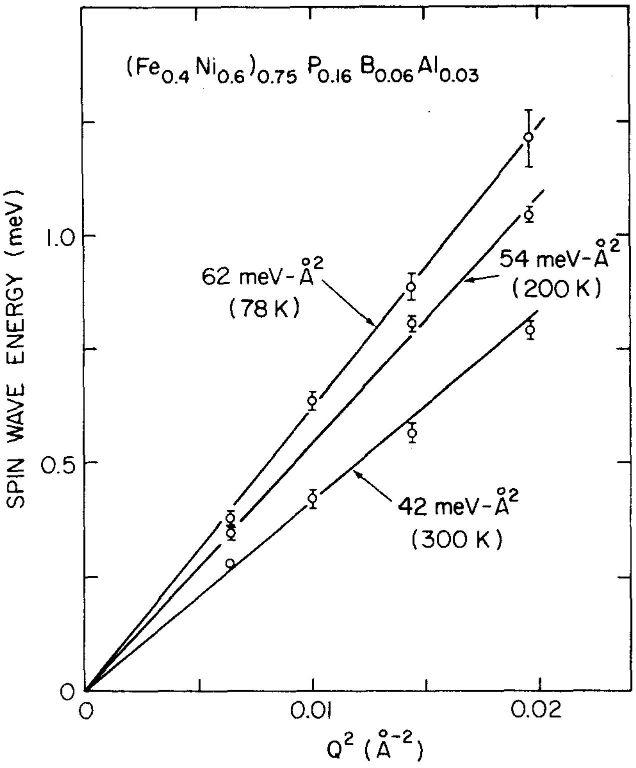

Fig. 1. Spin wave energy as a function of the square of the wavevector, showing that the long wavelength excitation spectrum obeys the quadratic dispersion relation $E=DQ^2$.

tion and adjusting D to give the best least-squares fit to experimental spectra at each Q. Further experimental details can be found in Ref. 12.

The samples were made by the centrifugal spinning technique (18) and were in the form of long ribbons. The nominal concentrations studied were x=0.2, 0.3, and 0.4, with each sample having a total weight of ~4 gm. The B^{11} isotope was used in the preparation of the samples to avoid the large neutron absorption cross section of B^{10}. The transition temperatures measured by neutron scattering agreed well with the transition temperatures determined by susceptibility on the identical samples, but are somewhat higher than the transitions reported in Refs. 6 and 7 at the same nominal concentrations.

The results for the spin wave dispersion relation are shown in Fig. 1, where the spin wave energy is plotted as a function of Q^2 for the x=0.4 alloy. The data are seen to obey Eq. 1 as expected, with D increasing with decreasing temperature below the Curie temperature of 365K. At 78K we obtain a value of D=(62±2)meV-Å^2, which is the maximum value attained. Figure 2 shows that below ≈78K the spin wave energy decreases with further decrease of temperature as some of the spins freeze. Preliminary measurements for x=0.3 show an even more dramatic decrease at low T.

Figure 3 shows the scattering observed at a wavevector of 0.10Å^{-1} for x=0.4. The peak at zero energy transfer originates primarily from nuclear incoherent scattering as well as scattering from the cryostat and sample holder. There may also be a small magnetic component at E=0 which is unresolved; this will be the subject of further studies. Here we are interested in the spin waves, which appear symmetrically around E=0 for neutron energy loss (E>0; spin wave creation) and for neutron energy gain (E<0; spin wave destruction). The three sets of data have been successively shifted by 25 counts along the ordinate to separate the spectra for clarity. At T=280K the spin waves have a substantial intrinsic linewidth. By cooling to 78K the spin waves are seen to renormalize to higher energy and sharpen. The reduction in intensity is due to the decrease in the Bose thermal population factor. Further decrease of the temperature (to 30K) is seen to result in a shift of the peak to smaller energy, and also to an increase in the linewidth of the spin wave scattering. The solid curves in Fig. 3 are least-squares fits to the spectra using Eq. 1 convoluted with the instrumental resolution as discussed above. The intrinsic spin wave lineshape was assumed to be Lorentzian. The peak at E = 0 was taken to be a gaussian whose width was fixed to the measured (gaussian) energy resolution of 0.35 meV FWHM.

These results show that D decreases at low temperatures as the spin-glass state is approached. This decrease is presumably due to a decrease in the average exchange interaction as some of the spins freeze out. An anomaly is also observed in the low temperature magnetization (6) for the x=0.4 alloy. The dominant contribution to the increased spin wave linewidths at low T is probably due to an increased scattering rate as the magnetic disorder increases. There could also be a contribution from a distribution of magnetic states at low T, which would tend to smear the excitation spectrum when averaged over the whole crystal.

Our preliminary neutron results show that a larger reduction in D occurs for the x=0.3 concentration, which would be expected on the basis of the spin resonance and susceptibility results. Indeed in the ear-

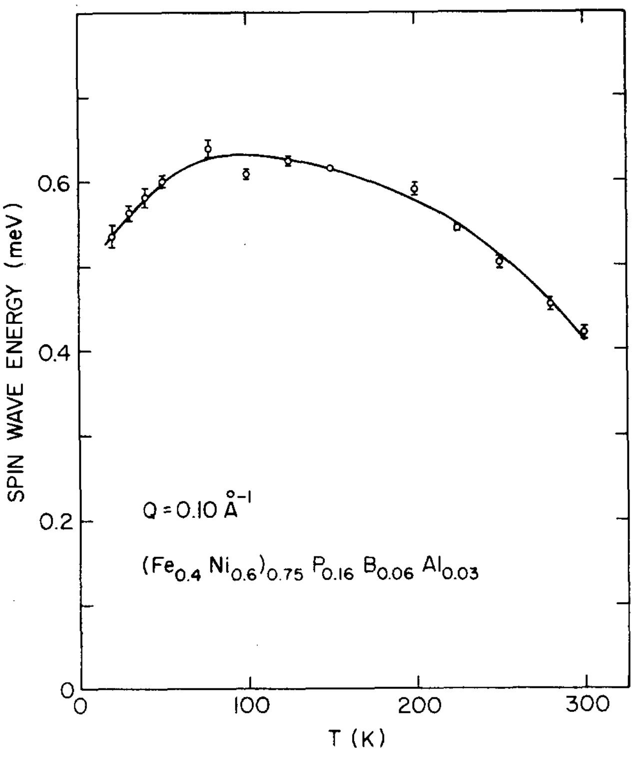

Fig. 2. Temperature dependence of the spin wave energy at Q=0.10 Å^{-1}. Below T_c (365K) the energy increases with decreasing temperature as usual. Below ~80K, however, the energy begins to decrease with decreasing temperature as the spin-glass state is approached.

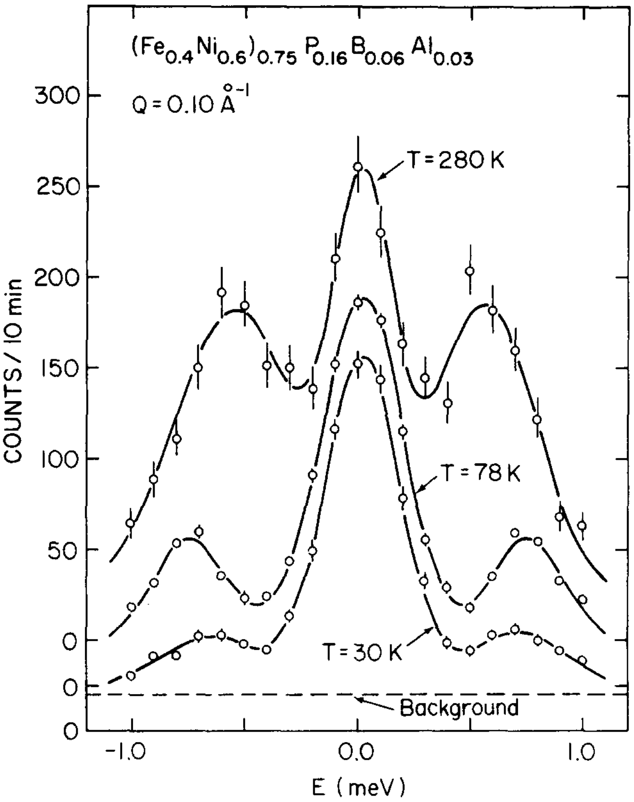

Fig. 3. Observed scattering at a wavevector of $0.10\ Å^{-1}$ at three different temperatures. The spin wave peaks are located symmetrically about E=0. With decreasing temperature the spin waves initially increase in energy while the linewidths decrease. As the spin-glass state is approached at low temperature the opposite behavior ensues; the energy decreases and the linewidth increases.

lier neutron results (at higher temperatures) it was already noted (3-5) that significant deviations from spin wave theory occurred for $x=0.3$. Our preliminary results for $x=0.2$ ($T_c=118K$) show only very broad spin wave excitations. Further work is in progress.

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