Phys. Status Solidi (b) 92, k57 (1979).
[5]See, for example, F. Bassani and G. P. Parravicini, *Electronic States and Optical Transitions in Solids* (Pergamon, Oxford, 1975).
[6]See, for example, M. Lax, *Symmetry Principles in Solid State Physics and Molecular Physics* (Wiley, New York, 1974), p. 431. Translations to other notations can be made with Table A11 in this reference.
[7]The number of holes is restricted to 4 in this representation. For BMEC with more than four holes, some of them would have to be placed in excited states.
[8]Details of the calculation will be presented in a future publication.
[9]A. Baldereschi and N. O. Lipari, Phys. Rev. B 6, 2697 (1973).
[10]M. L. W. Thewalt, Solid State Commun. 21, 937 (1977).
[11]E. C. Lightowlers, M. O. Henry, and M. A. Vouk, J. Phys. C 10, L713 (1977).
[12]Based on the local-density approximation, we expect that the correlation energy for the BE should be approximately the same for the ground and the one-electron low-lying excited states. These states both involve the same number of particles and have approximately the same densities.
[13]M. L. W. Thewalt, Can. J. Phys. 55, 1463 (1977).
[14]K. Elliott and T. C. McGill, Solid State Commun. 28, 491 (1978).

Spin-Wave Evolution Crossing from the Ferromagnetic to Spin-Glass Regime of Fe_xCr_{1-x}

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Inelastic-neutron-scattering studies on Fe_xCr_{1-x} for $x = 0.34$ and 0.26 reveal well-defined spin-wave excitations in the ferromagnetic regime and show a significant decrease of the spin-wave stiffness as the temperature is lowered towards the mictomagnetic or spin-glass regime. Within the spin-glass phase, no well-defined excitations are seen, but an intense quasielastic peak is observed.

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The statics and dynamics of spatially disordered systems with competing magnetic interactions have proven to be among the more interesting and difficult problems to study.[1,2] Even now, discussion continues on whether there is a transition to a true thermodynamically stable phase, as formulated by Edwards and Anderson, or a gradual freezing of spins with a widespread distribution of relaxation times.[3] The prototypical examples of spin-glass behavior are the alloys of $Cu_{1-x}Mn_x$ and $Au_{1-x}Fe_x$. Some theories[4] indicate the possibility of ferromagnetic (FM) ordering followed at lower temperatures by the onset of a spin-glass (SG) ordering. There have been several recent examples of systems which exhibit this sequence of phases. Examples are $(Pd\mathrm{Fe})_{1-x}Mn_x$,[5] Fe_{3-x}-Al_{1+x},[6] and the ionic solid $Eu_xSr_{1-x}S$.[7] Bulk measurements suggest that, for a range of x, Fe_xCr_{1-x} shows ferromagnetic features along with a more complicated behavior at lower temperatures. The low-temperature regime may be a spin-glass, mictomagnetic, or possibly a mixed-phase region[8]; however, since many of the characteristics of a spin-glass are observed, we will refer to Fe_xCr_{1-x} as a spin-glass in that part of the phase diagram. Our goal in these experiments is to probe the dynamics of the SG phase by studying the evolution of the spin waves from the FM phase, where the dynamics are reasonably well understood, into the SG phase. In the FM phase, well-defined spin waves are observed with a quadratic dispersion law, $E = Dq^2$. As the temperature is lowered, the stiffness D decreases until, within the SG phase, no well-defined ex-

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citations are observed.

Fe_xCr_{1-x} is a good system to study since the nearly equal lattice constants and identical bcc crystal structure of Fe and Cr improve the chances of having a random substitutional alloy. Also, the boundaries of the FM and SG regimes occur in convenient temperature ranges and the boundaries are well separated from each other. The studies were performed on polycrystalline samples of Fe_xCr_{1-x} with $x=0.34$, 0.28, 0.26, 0.24, and 0.22. We present the main features of the experiment by discussing the results for $x=0.34$ and $x=0.26$. The samples were prepared by arc melting, annealing at 1000°C, and quenching to room temperature. The room-temperature diffraction pattern revealed some diffuse scattering intensity indicative of some short-range nuclear order.[9]

The neutron scattering experiments were performed on a triple-axis spectrometer at the high-flux beam reactor of Brookhaven National Laboratory. Because the studies were performed on polycrystalline samples, the measurements were all near the forward direction. The incident energy was either 13.7 or 4.5 meV and the collimation was varied according to the intensity and resolution requirements.

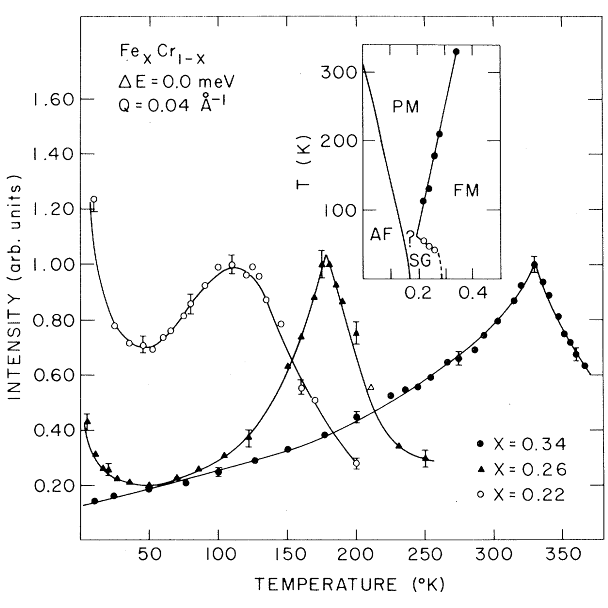

FIG. 1. Elastic scattering for small momentum transfer $Q = 0.04$ Å^{-1} as a function of temperature in Fe_xCr_{1-x}, $x = 0.34$, 0.26, and 0.22. Inset shows temperature-concentration phase diagram as deduced from the data. The open circles do not denote a sharp phase boundary. See text and Ref. 13.

The temperature variation of the intensities at $Q=0.04$ Å^{-1}, with the spectrometer set for zero energy transfer, is shown in Fig. 1. A peak corresponding to the Curie temperature is clearly observable for each alloy concentration. T_C decreases linearly with Fe concentration and extrapolates to 0 K for $x\simeq 0.16$ in agreement with other studies.[10] For $x=0.34$, the intensity decreases monotonically down to the lowest temperatures; however, data for $x=0.26$ and 0.22 show the scattered intensity increasing with lower temperatures below some temperature T_L, which depends on x. Similar temperature dependence of scattering has been previously observed in Fe_xCr_{1-x} (Ref. 11) as well as other SG systems.[7,12] Choosing T_L, which denotes the minimum in the intensity curves, as the boundary of the SG regime, one obtains the phase diagram shown in Fig. 1,[13] which is similar to other like systems, e.g., $Fe_{3-x}Al_{1+x}$ (Ref. 6) and $Eu_xSr_{1-x}S$ (Ref. 7).

The results for the inelastic scattering at $Q=0.075$ Å^{-1} are shown in Fig. 2. Below T_C, well-defined magnons are observed for $x=0.34$. As T is lowered, for the $x=0.34$ sample, the peak in the cross section shifts to larger energies because of the increase in the magnetic stiffness $\mathbf{D}$. This behavior is typical for a ferromagnet. On the other hand, for $x=0.26$, the development of the spectra at low T is surprisingly different. Just below T_C, the peaks shift to higher energies, but as T is lowered, their positions begin to move to lower energies and their linewidths Γ broaden. At the lowest temperatures, the peaks merge and finally at $T=10$ K a narrow quasielastic line remains with no evidence of propagating spin waves.

The cross section for inelastic scattering of neutrons from a FM is[14]

$$\frac{d^2\sigma}{d\Omega\, d\omega}=A\frac{k_f}{k_i}\frac{\hbar\omega/k_BT}{1-\exp(\hbar\omega/k_BT)}\frac{\chi(q)}{\chi_0}F(q,\omega). \qquad (1)$$

The constant A includes the form factor which is close to unity at these small $\mathbf{Q}$'s and some trivial constants. $\chi(q)$ is the transverse part of the static wave-vector–dependent susceptibility and χ_0 is the noninteracting atomic susceptibility. The spectral weight function $F(q,\omega)$ provides the description of the dynamic response of the system. Without theoretical arguments suggesting a particular form for $F(q,\omega)$, we have analyzed our data using several standard forms (Lorentzian, damped harmonic oscillator, and Halperin-Hohenberg).[15,16] Each of these results is a good description of the data whenever the magnon peaks are well defined. Therefore, one reaches the

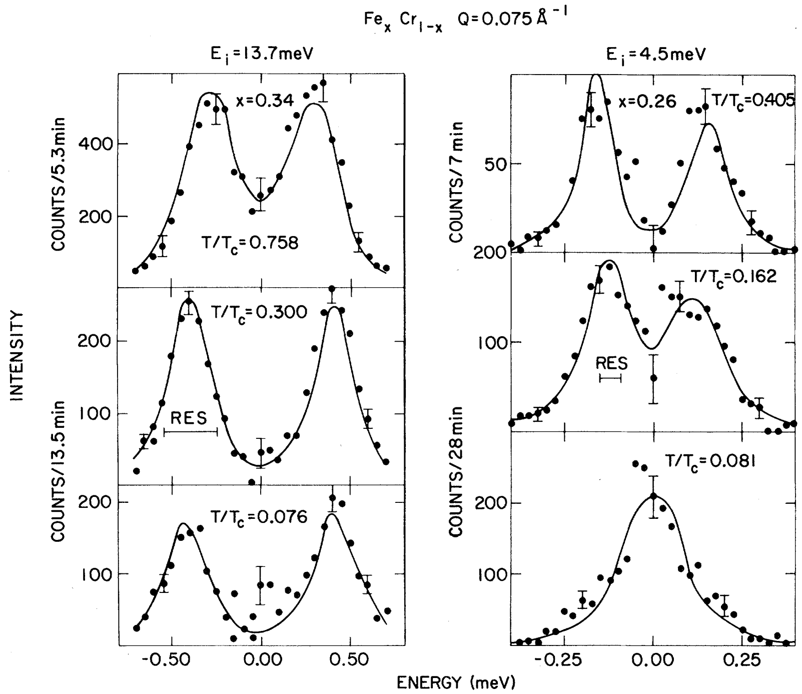

FIG. 2. Temperature evolution of spin-wave scattering in Fe_xCr_{1-x}, $x=0.34$ and 0.26. Solid line represents the convolution of the spectrometer resolution and a Lorentzian parametrization of the scattering. Experimental energy resolution is denoted.

same conclusions independent of the form of $F(q, \omega)$ used in the analysis. However, it should be mentioned that all of the above forms for $F(q,\omega)$ have inadequacies when the spin waves are overdamped. This problem has been discussed for ferromagnets near the Curie temperature.[15] For these reasons, we view the parameters obtained by fitting the data as not necessarily intrinsic to the system but certainly as the clearest and most efficient means of summarizing the data.

The solid line in Fig. 2 is the result of the convolution of Eq. (1) with the spectrometer resolution function. A Lorentzian form for $F(q,\omega)$ was taken and the parameters adjusted to give the best least-squares agreement with the data. A quadratic form of the spin-wave dispersion, $E=Dq^2$ was used since this was obeyed whenever well-defined spin-wave excitations were present. A constant background has been subtracted in addition to a resolution-limited peak due to elastic incoherent scattering and scattering from the sample container and cryostat. This elastic background was determined from the central part of the spectra at temperatures where spin waves are well defined. Because of this subtraction procedure, the errors at $E=0$ are considerably larger than at finite energy transfer.

Figure 3(a) shows the temperature dependence of D for $x=0.26$ and $x=0.34$. Both show an increase just below T_C and in the $Fe_{0.34}Cr_{0.66}$ sample, the stiffness eventually saturates at $D=60$ meV Å^2. In $Fe_{0.26}Cr_{0.74}$ one observes radically different behavior with D decreasing at temperatures below T_L. When the spectra coalesce into a peak about $E=0$, we can no longer assign a nonzero value for the stiffness and the spectra resemble those observed in the more conventional SG systems.[3]

Figure 3(b) shows the temperature dependence of the quantity $\chi(q)/\chi_0 T$ obtained from the computer fits. For conventional spin-wave theory, this quantity should be temperature independent as is the case for $x=0.34$. For $x=0.26$, however, there is an increase at low T, in contradiction to the predictions of spin-wave theory. The low-T spectrum for $x=0.26$ shows a quasielastic peak (Fig. 2). Part of this peak is due to the fall in D, but there may be an additional elastic scattering not accounted for in the cross section of Eq. (1). This may be due to the growth of short-ranged magnetic correlations or clusters, which have been observed in most spin-glasses. One also expects an increase in the truly elastic scattering from the freezing of spins into a glass-like state.

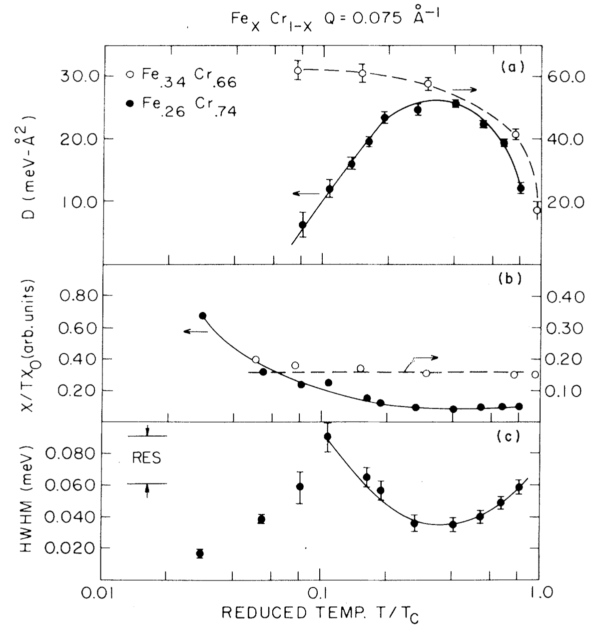

FIG. 3. Temperature dependence of parameters of the fits shown in Fig. 2. (a) Spin-wave dispersion constant; (b) interacting susceptibility divided by the noninteracting susceptibility and temperature; (c) Lorentzian half-width at half maximum for $x = 0.26$.

Figure 3(c) shows the temperature dependence of the parameter Γ for the $x = 0.26$ sample. The results for the $x = 0.34$ sample are not shown because the linewidths are resolution limited except very near T_C. For $x = 0.26$, Γ increases with decreasing temperature until approximately $T = 20$ K, reflecting the increasing linewidth of the spin waves. Below $T = 20$ K ($T/T_C = 0.112$), Γ represents the narrowing of the quasielastic scattering which remains after the collapse of the spin waves.

These results provide a framework for understanding the differences between the dynamics of a ferromagnet and a disordered SG-like system. It is clear that the dynamics of systems which exhibit a breakdown of the ferromagnetic state with a decrease in temperature cannot be explained by conventional spin-wave theory. Insight into the low-temperature behavior of D may be obtained by comparing it with the known behavior of a ferromagnet upon approaching T_C. Because of thermal fluctuations, the system breaks up into correlated regions and the magnetization decreases from its saturation value. The resulting decrease in the molecular field reduces the restoring forces for spin waves and critical slowing down occurs. As the SG regime is approached at lower temperatures, clusters of correlated spins freeze out such that the long-range order is decreased. The molecular field must also be reduced which causes a decrease in the stiffness. In the SG phase, the spins are frozen and the disorder appears as elastic scattering rather than quasielastic as in a paramagnet where the zero moment arises due to the thermal average of the spin fluctuations.

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[1]S. F. Edwards and P. W. Anderson, J. Phys. F 5, 965 (1975).

[2]J. A. Mydosh, J. Magn. Magn. Mater. 7, 237 (1978).

[3]A. P. Murani and A. Heidman, Phys. Rev. Lett. 41, 1402 (1978); A. P. Murani, J. Appl. Phys. 49, 1604 (1978).

[4]S. Kirkpatrick and D. Sherrington, Phys. Rev. B 17, 4384 (1978).

[5]B. H. Verbeek, G. J. Nieuwenhuys, H. Stocker, and J. A. Mydosh, Phys. Rev. Lett. 40, 586 (1978).

[6]R. D. Shull, H. Okamoto, and P. A. Beck, Solid State Commun. 20, 863 (1976).

[7]H. Maletta and P. Convert, Phys. Rev. Lett. 42, 108 (1979).

[8]S. Fishman and A. Aharony, Phys. Rev. B 21, 280 (1980).

[9]A. J. Aldred, B. D. Rainford, J. S. Kouvel, and T. J. Hicks, Phys. Rev. B 14, 228 (1976).

[10]F. Adcock, J. Iron Steel Inst. London 124, 129 (1931).

[11]S. K. Burke, R. Cywinski, and B. D. Rainford, J. Appl. Cryst. 11, 644 (1978).

[12]S. M. Shapiro, G. Shirane, B. H. Verbeek, G. J. Nieuwenhuys, and J. A. Mydosh, to be published.

[13]The boundary of the antiferromagnetic region is taken from other work: Y. Ishikawa, S. Hoshino, and Y. Endoh, J. Phys. Soc. Jpn. 22, 1221 (1967).

[14]W. Marshall and S. W. Lovesey, *Theory of Thermal Neutron Scattering* (Oxford Univ. Press, Oxford, 1971).

[15]L. Passell, J. Als-Nielsen, and O. W. Dietrich, in *Proceedings of the Fifth IAEA Symposium on Neutron Inelastic Scattering, Grenoble, France, 1972* (International Atomic Energy Agency, Vienna, 1972), p. 619.

[16]C. J. Glinka, V. J. Minkiewicz, and L. Passell, Phys. Rev. B 16, 4084 (1977).