High-temperature spin dynamics in an amorphous ferromagnet

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We have carried out an inelastic-neutron-scattering investigation of the high-temperature spin-wave excitations and the critical dynamics in the amorphous ferromagnet $(Fe_{65}Ni_{35})_{75}P_{16}B_6Al_3$ ($T_C = 572$ K). Well-defined spin-wave excitations are observed for wave vectors $0.06 \leq \vec{q} \leq 0.18$ Å^{-1} and for temperatures up to 555 K. The spin-wave dispersion relation over this q range is well described by the expression $\hbar\omega = \Delta + Dq^2$, where $\Delta(T=0) \simeq 0.05$ meV and $D = 115[1-0.45\,(T/T_C)^{5/2}]$ meV Å^2; the 5/2 power law appears to hold up to 450 K. Measurements at $T = 450$ K show that the spin-wave damping is consistent with the Heisenberg-model prediction $\Gamma(q) \sim q^4 \ln^2[k_B T/\hbar\omega(q)]$. In the critical region the spin-wave stiffness is found to follow the power law $D \sim (1-T/T_C)^{0.5 \pm 0.1}$ for $0.02 \leq 1-T/T_C \leq 0.2$, while at T_C the energy width is consistent with $\Gamma_C(q) \sim q^{2.7 \pm 0.2}$ for $0.05 \leq q \leq 0.18$ Å^{-1}. These results are in satisfactory agreement with dynamical scaling theory for the Heisenberg ferromagnet and further they are in good accord with similar, albeit more-detailed, measurements in the crystalline transition metals Fe, Co, and Ni.

I. INTRODUCTION

Recently there has been considerable interest in the static and dynamic properties of amorphous ferromagnets.[1] Detailed studies have now been reported on the low-temperature magnetic properties of a variety of amorphous metals, most notably the metal-metalloid systems such as $Fe_{75}P_{15}C_{10}$ (Ref. 2) and $Co_{80}P_{20}$.[3,4] Using inelastic-neutron-scattering techniques it has been clearly demonstrated that the amorphous lattice will support well-defined spin waves over a range of wave vectors up to at least 0.25 Å^{-1} and over a rather wide range of temperature.[1-5] Furthermore, at low temperatures the magnetization is consistent with the Heisenberg model prediction[6] $M = M_0(1 - BT^{3/2} - cT^{5/2})$ although a persistent discrepancy has been noted between the coefficient B obtained from the magnetization measurements and the B (spin wave) which one would infer from the measured spin-wave dispersion relation.[1,3] The source of this discrepancy is still not understood. Except for this discrepancy, however, conventional spin-wave theory gives a surprisingly good description of the low and intermediate temperature static and dynamic magnetic properties of the metal-metalloid amorphous ferromagnets.

Some studies have also been reported of the magnetic equation of state near the critical point.[7] In systems which exhibit a well-defined ferromagnetic phase transition, it is found that the critical exponents are similar to those observed in the crystalline transition metals Fe and Ni. The crystalline systems in turn are expected to exhibit Heisenberg model ($d=3$, $n=3$) critical behavior. In all cases, both amorphous and crystalline, there are some slight discrepancies, but the overall behavior seems to be at least reasonably consistent with theory. It should be emphasized that the sharp transition in the amorphous systems and the Heisenberg-like critical exponents rest crucially on the fact that these metals are magnetically isotropic on a microscopic scale. So far no measurements have been reported on the *dynamic* critical properties of an amorphous ferromagnet. The reasons for this are firstly that our overall understanding of amorphous systems has only recently reached the point where such studies are justified, and secondly that the measurements are feasible only in a restricted range of materials. In particular, the material must have a T_C well below the crystallization temperature; in addition, for neutron scattering reasons it must exhibit very weak incoherent and static-structure scattering so that the magnetic scattering may be unambiguously separated out. It is clear, however, that such a study is of considerable interest. In particular, it would be especially interesting to learn whether dynamical scaling theory[8] for the Heisenberg ferromagnet, which is so successful in the crystalline transition metals, will be equally accurate in describing the dynamical critical behavior of the metal-metalloid amorphous ferromagnets.

In this paper we report an inelastic-neutron-scattering study of the spin-wave excitations and critical dynamics in the amorphous ferromagnet $(Fe_{65}Ni_{35})_{75}P_{16}B_6Al_3$. We shall discuss the reasons for the choice of this particular material in Sec. II. The spin-wave experiments at intermediate temperatures represent an extension of those by Axe *et al.*[1] in $(Fe_{93}Mo_7)_{80}P_{20}B_{10}$. Our results reinforce their basic conclusion that classical spin-wave theory is quite successful in describing both the spin-wave-energy renormalization and the spin-wave lifetimes at intermediate temperatures. As noted above, the critical dynamics represents new terrain in the amorphous magnet problem. As we shall show, simple theory is quite successful.

The format of this paper is as follows. In Sec. II we give preliminary details including the materials preparation and characterization and the neutron scattering techniques used in these experiments. Section III reports the experimental results, the relevant theory, and the analysis for each of the spin-wave dispersion relations as a function of temperature, the renormalization of the spin-wave stiffness near T_C and the critical dynamics at T_C. Finally, the conclusions and suggestions for future experiments are given in Sec. IV.

II. PRELIMINARY DETAILS

As part of an overall Brookhaven–MIT–Bell Labs–Johns Hopkins collaborative study, a series of alloys of the general form $(Fe_xNi_{100-x})_{75}P_{16}B_6Al_3$ have been synthesized.[5,9] These alloys are produced in ribbons by spin quenching as described by Chen and Miller[10]; they are believed to have a random close-packed structure[11] which is independent of the Fe fraction. The isotope ^{11}B was used for our samples because natural boron contains about 19% of ^{10}B which has an extremely high absorption for thermal neutrons. To a first approximation Ni behaves nonmagnetically in this series.[12] Thus the effect of dilution is to decrease the moment M_0, the Curie temperature T_C, and the spin-wave stiffness D with increasing Ni concentration. In a separate paper[9] we shall report on the overall magnetic properties of this series, and especially on the temperature dependence of the magnetization as obtained from Mössbauer and magnetization-in-a-field measurements and as inferred from the measured dispersion relations. From this study it was found empirically that one of the alloys $(Fe_{65}Ni_{35})_{75}P_{16}B_6Al_3$, satisfied rather well the requirements noted in the Introduction for a study of the critical dynamics.

The $(Fe_{65}Ni_{35})_{75}P_{16}B_6Al_3$ sample was in the form of several long ribbons ~1 mm wide and 0.1 mm thick; the net amount of material was 5 g. The ribbons were wrapped around a thin aluminum cylinder yielding a sample with net dimensions ~2 cm in diameter by 3 cm in height. For measurements below room temperature the sample was placed in a Displex variable temperature cryostat; temperature could be controlled to 0.2 K or better. For the high-temperature measurements the sample was enclosed in a thin-walled quartz tube with a slight amount of argon gas to promote temperature uniformity. The quartz sample tube was then contained in a standard Brookhaven oven; near $T_C = 572$ K the temperature was held constant to better than 1 K during a typical run. Initial specific-heat measurements at Bell Laboratories indicated a T_C of 558 K; after subsequent annealing T_C increased to 569 K. In our quasielastic neutron-scattering measurements we found that the critical scattering showed a sharp peak at $T_C = 572 \pm 1$ K indicating a well-defined second-order phase transition. This final difference of 3 K may be simply a thermometer calibration error.

The experimental difficulties and peculiarities associated with inelastic-neutron-scattering studies of amorphous ferromagnets have been extensively discussed by Axe *et al.*[1] The essential difficulty is that all measurements must be perfomed around the forward direction rather than around a reciprocal lattice position as in single-crystal studies. We have followed rather closely the procedures outlined in Ref. 1. The experiments were performed with a triple-axis spectrometer at the Brookhaven high-flux beam reactor. The incoming neutron energy was fixed at either 13.7 meV ($13.7E_i$) or 47.3 meV ($47E_i$). For $13.7E_i$, prolytic graphite (002) was used for monochromator and analyzer, Be(002) was used for $47E_i$. The horizontal collimators were set at 20-10-10-20 full width at half-maximum (minutes), except for data near T_C with $13.7E_i$, for which the collimators were set at 10-10-10-10. Counting intervals ranged from 2 to 30 min. per point.

III. EXPERIMENTAL RESULTS, THEORY AND ANALYSIS

A. Spin waves at low and intermediate temperatures

Typical experimental results at $T = 300$ K, $q = 0.07$ Å^{-1} and $q = 0.14$ Å^{-1} are shown in Fig. 1. The former were taken with $E_i = 13.7$ meV and the latter with $E_i = 47.3$ meV. The temperature-dependent spin-wave response at $q = 0.06$ Å^{-1}, with the background and the central peak subtracted, is shown in Fig. 2. The solid lines in both figures are theoretical fits as we shall discuss below. It is evident from these results that the $(Fe_{65}Ni_{35})_{75}P_{16}B_6Al_3$ amorphous alloy does indeed sustain well-defined spin-wave excitations over a wide range of

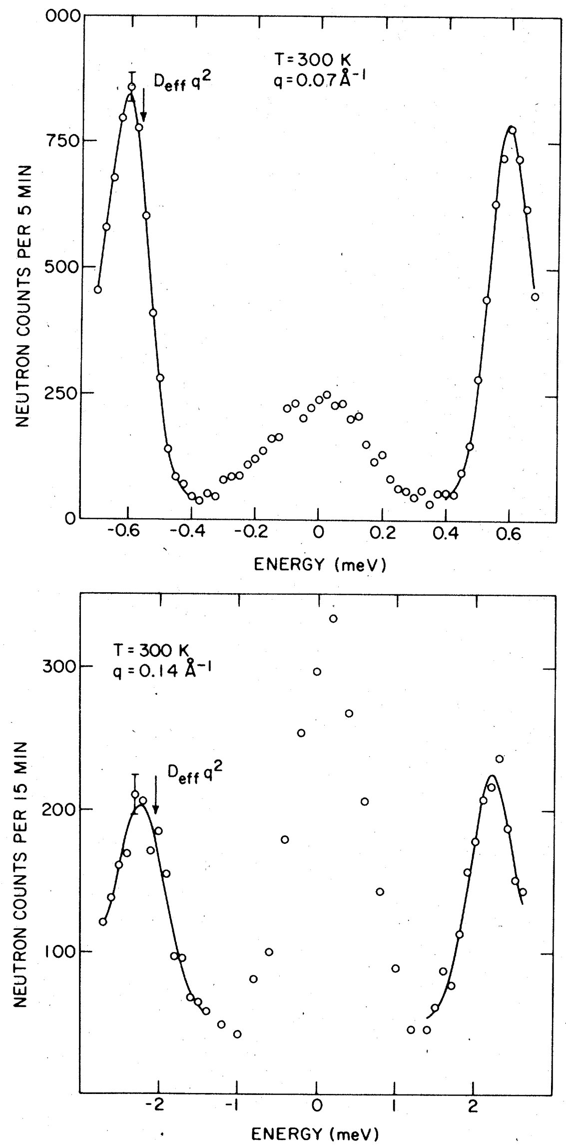

FIG. 1. Typical spin-wave scans at $T = 300$ K. The upper data were taken with $E_i = 13.7$ meV, the lower with $E_i = 47.3$ meV. The solid lines are least-squares fits to the data as described in the text. No background has been subtracted.

wave vectors and temperatures. The incoherent energy resolution of the instrument is given by the widths of the central peaks in Fig. 1. An unusual feature of the small-angle scattering is that both the energy-gain and the energy-loss spin-wave peaks are focused so that the observed spin-wave peaks are both narrower than the incoherent and static structure scattering centered at $E = 0$. We should note also that because of kinematical restrictions, that is for momentum transfer q only $\sim\hbar^2 k_i q/m$ energy can be transferred, it was not possible in many cases to scan the full spin-wave peak.

As discussed extensively by Axe *et al.*,[1] in order to extract the spin-wave energy and damping from the measured profiles it is necessary to convolute a theoretical cross section with the instrumental resolution. At small angles the vertical resolution, which is q independent, is typically comparable with the horizontal momentum transfer; thus the mean momentum transfer, averaged over the resolution ellipse, is measurably higher than the nominal setting. Fortunately, for a given spectrometer arrangement the resolution function may be accurately calculated using the method of Cooper and Nathans.[13] We have assumed a Lorentzian cross section of the form

$$\mathcal{S}(\vec{q},\omega) = C(\vec{q})\left(\frac{\hbar\Gamma(q)[n(\omega)+1]}{[\hbar\Gamma(q)]^2+[\hbar\omega-\hbar\omega(q)]^2} + \frac{\hbar\Gamma(q)n(\omega)}{[\hbar\Gamma(q)]^2+[\hbar\omega+\hbar\omega(q)]^2}\right), \qquad (1)$$

where $n(\omega) = [\exp(\hbar\omega/kT) - 1]^{-1}$.

For the purposes of the deconvolutions we have assumed that within the resolution ellipse $\hbar\omega(q) = D_{\text{eff}} q^2$ and $\hbar\Gamma(\vec{q}) = \text{const}$. The cross section [Eq. (1)] convoluted with the instrumental resolution function is then fitted in a least-squares sense to the instrumental data to obtain $C(q)$, $\Gamma(q)$, and $\omega(q)$. The solid lines in Figs. 1 and 2 are the calculated profiles so obtained. It is evident that the Lorentzian cross section describes the experimental data very well. This is found to hold true at all wave vectors at all temperatures measured. It is also clear from Fig. 1 that $\hbar\omega(q) = D_{\text{eff}} q^2$ occurs

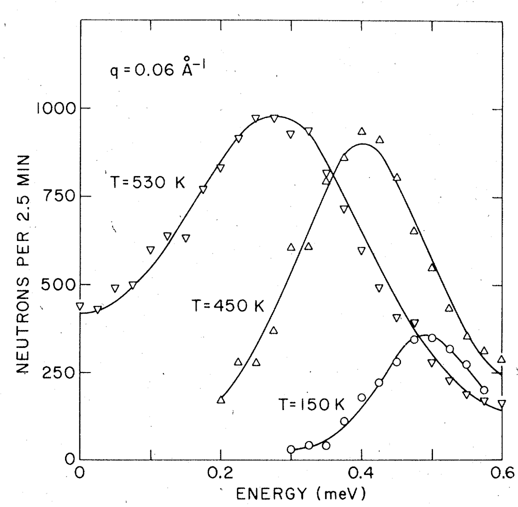

FIG. 2. Neutron groups for three temperatures at $q = 0.06$ Å^{-1}. The solid lines are best fits of Lorentzians to the data. A central peak and a constant background have been subtracted.

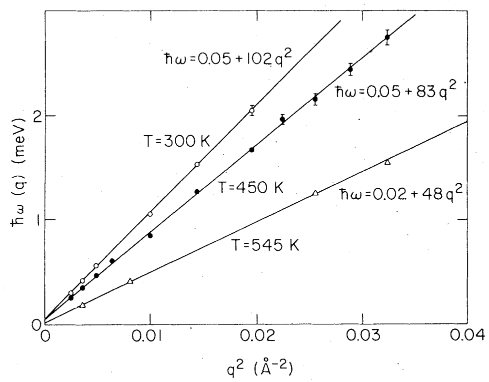

FIG. 3. Spin-wave energies vs q^2 at three temperatures. The lines are fits to the expressions $\hbar\omega = \Delta + Dq^2$. Terms of order q^4 are below our resolution limit.

~10% below the peak energy. This 10% difference is the correction for vertical resolution. The resultant spin-wave dispersion relations at several temperatures are shown in Fig. 3. In Fig. 4 we show the spin-wave damping, $\hbar\Gamma(q)$ versus q at $T = 450$ K. We should note that to within the errors $C(q)$ appears to be q independent as expected for ferromagnetic spin waves.

We now consider the theory for the magnons in an amorphous ferromagnet. Of course, spin-wave theory is a mature, well-tested subject in insulating systems, especially those with short range predominantly isotropic interactions.[14] The extension to amorphous systems at long wavelengths has been discussed in detail by Axe *et al.*[1] We refer the reader to that paper for a full description. Quite generally one has[6]

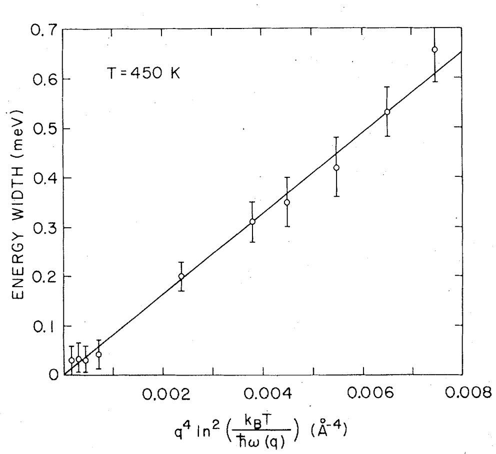

FIG. 4. Spin-wave Lorentzian half-widths at $T = 450$ K. The straight line indicates that Γ follows rather well the predicted $q^4 \ln^2[k_B T/\hbar\omega(q)]$ q dependence.

$$\hbar\omega(\vec{q}) = \Delta + Dq^2 + Eq^4 + \cdots . \tag{2}$$

This expression is valid for $Dq^2 \gg \Delta$; in this case Δ represents an effective gap due to the dipole-dipole interactions.[15] D measures the second moment of the exchange interaction, E the fourth, etc. The ratio E/D thence measures the range of the exchange interaction. Explicitly, in an amorphous material

$$E = -\frac{D}{20} \frac{\int r^4 J(r) g(r)\, dr}{\int r^2 J(r) g(r)\, dr}, \tag{3}$$

where $J(r)$ is the range-dependent exchange interaction and $g(r)$ is the pair distribution function.[1] From simple spin-wave theory for the Heisenberg ferromagnet[6] one predicts in addition

$$D(T) = D(0)(1 - aT^{5/2} - bT^{7/2} \cdots), \tag{4}$$

and correspondingly for the low-temperature magnetization

$$M(T) = M_0(1 - BT^{3/2} - CT^{5/2} \cdots). \tag{5}$$

It can be shown that[1]

$$a \sim \frac{\langle r^2 \rangle k_B}{4} \frac{B}{D}, \tag{6}$$

where $\langle r^2 \rangle$ is the mean range of interaction, k_B is Boltzmann's constant, and a, B, D are defined by Eqs. (4) and (5). Each of Eqs. (2)–(5) have been found to hold rather well in the crystalline transition metals.

The spin-wave energies are plotted versus q^2 in Fig. 3. It is evident that the dispersion relation Eq. (2) holds very well at all q's and at all temperatures. Further, to within the errors, we find that E, the quartic coefficient, is below our resolution limit. This in turn implies that the exchange interactions are quite short range in this system. For $T \leq 450$ K the dipole-dipole effective gap $\Delta = 0.05 \pm 0.01$ meV. We note that in $(Fe_{93}Mo_7)_{80}B_{10}P_{20}$, with a flat-plate geometry Axe *et al.*[1] deduce $\Delta = 0.035$ meV. From the measured dispersion relations one may obtain D as a function of temperature. We show in Fig. 5, $D(T)$ plotted versus $T^{5/2}$ as anticipated from Eq. (4). It is evident that the $T^{5/2}$ diminution law holds very well up to at least $0.8T_C$. A similar result, albeit over a smaller temperature range, has been found previously by Axe *et al.*[1] The spin-wave stiffness D is well described by the law

$$D(T) = (115 \pm 5)[1 - 0.45(T/T_C)^{5/2}] \text{ meV Å}^2 . \tag{7}$$

In $(Fe_{93}Mo_7)_{80}B_{10}P_{20}$ Axe *et al.*[1] find a value of 0.61 for the coefficient of the $T^{5/2}$ term. If we assume that the range of the interaction $\langle r^2 \rangle$ is the same in the two materials then according to Eq. (6) the $T^{5/2}$ coefficients in the two systems ought to be

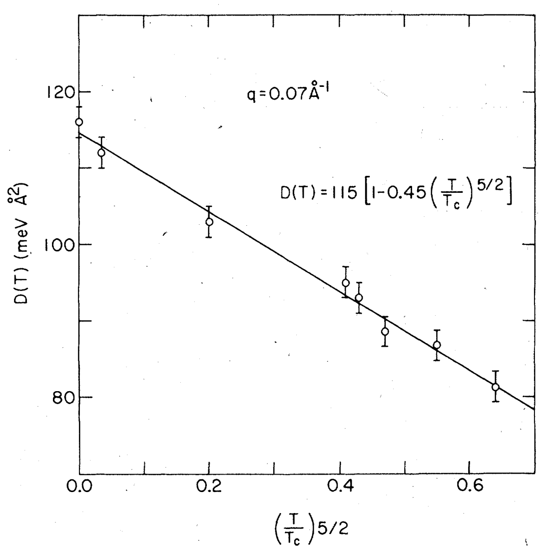

FIG. 5. Renormalization of the spin-wave stiffness $D(T)$. The change in D is proportional to $T^{5/2}$ up to at least $T=0.8\,T_C$.

simply related. Using the measured values for B and D in the two materials we calculate $a=0.43$ for $(Fe_{65}Ni_{35})_{75}P_{16}B_6Al_3$ in good agreement with the measured value of 0.45. Further, a calculated from Eq. (6) is at least of the right order of magnitude. We should emphasize, however, that in our system E seems to be below our resolution limit so that this apparent agreement between experiment and theory fcr a may be somewhat accidental.

Finally, we consider the spin-wave damping. At long wavelengths and intermediate temperatures the spin-wave damping in a Heisenberg ferromagnet is predicted to have the form[16,17]

$$\Gamma(q)\sim T^2q^4\ln^2[k_BT/\hbar\omega(q)]\,. \tag{8}$$

Axe *et al.*[1] have shown that their data in $(Fe_{93}Mo_7)_{80}B_{10}P_{20}$ have the temperature dependence predicted by Eq. (7). The spin-wave linewidths in our sample are plotted versus $q^4\ln^2[k_BT/\hbar\omega(q)]$ in Fig. 4. It is evident that the data obey this law rather well. The reader should be cautioned, however, that the error bars are such that we really have only demonstrated consistency and certainly we have not shown the uniqueness of Eq. (8). Indeed Harris[17] has shown that there are rather large correction terms which must be taken into account in any more refined test of the theory. It is amusing to note, nevertheless, that Eq. (8), which was calculated on the basis of a Heisenberg model has now been verified in two amorphous ferromagnets. No such tests, except very near T_C in EuO,[14] are available for crystalline systems.

B. Critical dynamics

The modern era of critical dynamics studies in magnets began with the dynamic scaling hypothesis and the hydrodynamic spin-wave calculations by Halperin and Hohenberg.[8] The theory of dynamical critical phenomena as it pertains to Heisenberg ferromagnets has been extensively reviewed by Dietrich *et al.*[14] and by Glinka *et al.*[18] in the context of their experiments in EuO and Co, respectively. Accordingly, we shall mention only the aspects immediately relevant to our experiments and the reader is referred to the above papers for a full discussion. According to the hydrodynamic theory[8] the spin-wave stiffness D in an isotropic ferromagnet should renormalize near T_C like

$$D(T)\sim(1-T/T_C)^{\nu'-\beta}\sim\kappa^{1/2-\eta}\,, \tag{9}$$

where κ is the inverse correlation length and β, ν', η are the usual critical exponents.[19] Hence,

$$\hbar\omega(\vec{q})\sim\kappa^{1/2-\eta}q^2=q^{5/2-\eta}(\kappa/q)^{1/2-\eta}\,. \tag{10}$$

Note that since the damping $\Gamma(q)\sim q^4\ln^2q$, then there should always be underdamped spin waves at long enough wavelengths below T_C. According to the dynamic scaling hypothesis[8] one may write quite generally

$$\hbar\omega(q)=q^z\Omega(\kappa/q)\,. \tag{11}$$

Thus for $T<T_C$ and $q\ll\kappa$, $Z=\frac{5}{2}-\eta$ and $\Omega(\kappa/q)\sim(\kappa/q)^{1/2-\eta}$. However, for $T=T_C$ so that $\kappa=0$, one then has $\Omega(\kappa/q)=\Omega(0)=\text{const}$ and therefore

$$\hbar\omega_{T=T_C}(q)\sim q^{5/2-\eta}\,. \tag{12}$$

These results have been approximately confirmed in Fe, [15]Ni,[20] Co,[18] and EuO.[14]

The experimental procedures for the studies near T_C are essentially idential to those discussed in the previous sections. The critical dynamics measurements, however, are complicated by the elastic central-peak scattering evident in Fig. 1. In order to account for this extra scattering we have fitted it with a Gaussian at each q vector at room temperature, where it is well resolved, and we have then subtracted this fitted central peak uniformly from the data near T_C. This introduces an extra uncertainty of typically 10% in the final parameters. Well-defined spin waves could be observed up to at least 555 K, that is, to $0.975T_C$. Uncertainties associated with the central-peak subtraction prevented us from obtaining reliable results close to T_C. The final values for the spin-wave stiffness $D(T)$ are shown in Fig. 6. Within the decade $0.02\leq 1-T/T_C\leq 0.2$ the data are consistent with a power law of 0.5 ± 0.1. From the most recent estimates[21] $\nu'=0.705\pm0.001$, $\beta=0.365\pm0.001$ one has a theoretical exponent of 0.341 ± 0.002. In

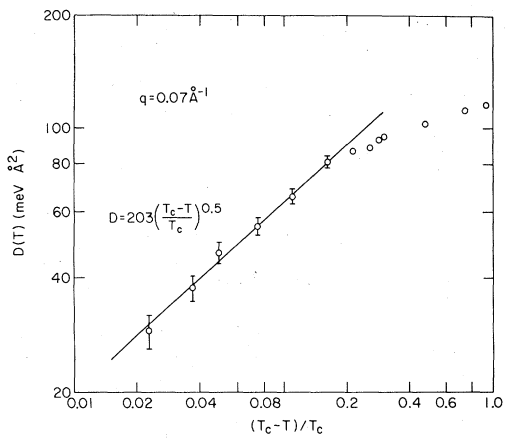

FIG. 6. Renormalization the spin-wave stiffness $D(T)$ near T_C.

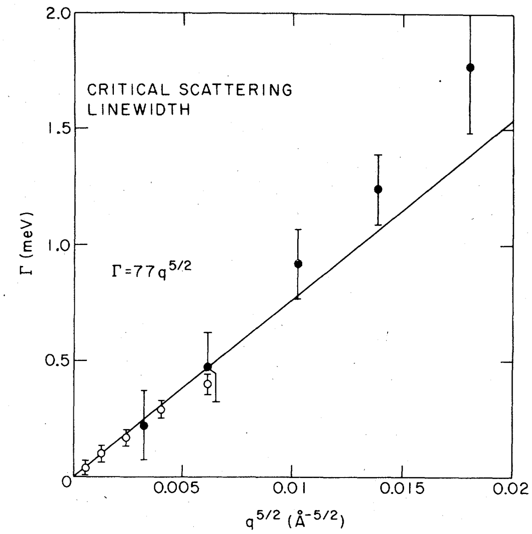

FIG. 8. Lorentzian half-width of the critical scattering at T_C (572 K). The open and filled circles were taken with 13.7 and 47.3 meV, respectively. The line is the best fit to a $\frac{5}{2}$ power law.

the transition metals Fe, Co, and Ni[15,18,20] one finds as a mean value an exponent of 0.39 ± 0.06. Our measured value, 0.5 ± 0.1, for $(Fe_{65}Ni_{35})_{75}P_{16}B_6Al_3$ agrees with this value to within the errors but the theoretical prediction 0.341 ± 0.002 is outside of the error limits. However, we do not regard this discrepancy as significant given the uncertainties in our central-eak substraction procedure and the fact that our data do not extend very close to T_C

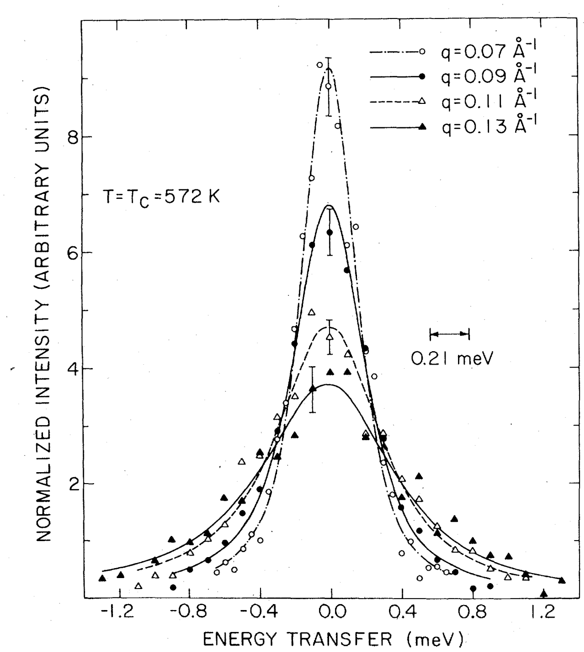

FIG. 7. Dynamical critical scattering at T_C. The background has been subtracted and the overall intensity for each q has been normalized. The lines are the results of fits to a Lorentzian profile convoluted with the instrumental resolution function.

The dynamical-critical-scattering measurements at T_C are shown in Fig. 7. The arrow indicates the experimental energy resolution of 0.21 meV. For these data the background has been removed and the neutron groups have been normalized so that they have the same integrated intensity. The lines are the results of least-squares fits to a Lorentzian line shape [Eq. (1) with $\omega(q) = 0$] convoluted with the instrumental resolution function. It is evident that the agreement is in general very good. The net half-widths so deduced are shown in Fig. 8 plotted versus $q^{5/2}$. The open and filled circles correspond to scans taken with $E_i = 13.7$ and 47.3 meV, respectively. The predicted $q^{5/2}$ law holds reasonably well although there is a measurable deviation at the highest q's. A least-squares fit to a general power law gives

$$\hbar\Gamma_{T=T_C}(q) = 114 q^{2.7\pm0.2}\ \text{meV Å}^{2.7}\,. \tag{13}$$

These results are in good agreement with theory. In crystalline iron[15] one also finds an exponent $Z = 2.7 \pm 0.3$. It is also interesting to note that from the dynamic scaling hypothesis $Z = 2.7$ implies $D \sim (1 - T/T_C)^{0.5}$ as we in fact observe.

IV. CONCLUSIONS

This work together with that of Axe *et al.*[1] gives a rather complete characterization of the long-wavelength spin dynamics in the metal-metalloid amorphous ferromagnets both at general tempera-

TABLE I. Comparison of the properties of spin dynamics in Fe and $(Fe_{65}Ni_{35})_{75}P_{16}B_6Al_3$.

Law	Quantity	Fe	$(Fe_{65}Ni_{35})_{75}P_{16}B_6Al_3$
	Moment per Fe	$2.09\ \mu_B$	$2.0\ \mu_B$
	T_C	1042 K	572 K
$\hbar\omega = \Delta + Dq^2 + Eq^4$	Δ	0.1 meV	0.05 meV
	$D(0)$	281 meV Å^2	115 meV Å^2
	E	-270 meV Å^4 at room temperature	$<\vert 30\vert$ meV Å^4 in magnitude
$D = D(0)\left[1 - a\left(\frac{T}{T_C}\right)^{5/2}\right]$	a		0.45
$D = d_0\left(1 - \frac{T}{T_C}\right)^x \quad (T < T_C)$	d_0	320 meV Å^2	203 meV Å^2
	x	0.37 ± 0.03	0.5 ± 0.1
$\hbar\Gamma = \hbar\Gamma_0 q^Z \quad (T < T_C)$	Γ_0	225 meV Å$^{2.7}$	114 meV Å$^{2.7}$
	Z	2.7 ± 0.3	2.7 ± 0.2

tures and near T_C. It is evident that many detailed predictions of the theory for the Heisenberg ferromagnet are confirmed in these amorphous systems. Although some similarities might be expected based on dimensionality and symmetry grounds we find the detailed agreement surprising. We summarize in Table I various measured properties in crystalline iron and in amorphous $(F_{65}Ni_{35})_{75}P_{16}B_6Al_3$. Clearly, the two systems are closely similar in all respects. As discussed in Ref. 9, the relationship between D and B in the material is also satisfactory. Thus we seem to have a rather complete description of the magnetism based on the Heisenberg picture.

A more detailed study of the critical dynamics both above and below T_C in an amorphous ferromagnet would now be of considerable value. One would like especially to map out the dynamic scaling function $\Omega(\kappa/q)$. The material studied here is ideal except for the appreciable incoherent scattering at $E=0$. Hopefully, an amorphous ferromagnet with much smaller incoherent scattering may be discovered so that a complex study will be possible.

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