Spin-wave excitations and low-temperature magnetization in the amorphous metallic ferromagnetic $(Fe_xNi_{1-x})_{75}P_{16}B_6Al_3$

R. J. Birgeneau
Department of Physics and Center for Materials Science and Engineering, Massachusetts Institute of Technology, Cambridge, Massachusetts 02139

J. A. Tarvin and G. Shirane
Brookhaven National Laboratory, Upton, New York 11973

E. M. Gyorgy, R. C. Sherwood, and H. S. Chen
Bell Laboratories, Murray Hill, New Jersey 07974

C. L. Chien
Department of Physics, The Johns Hopkins University, Baltimore, Maryland 21218

(Received 10 May 1978)

We have carried out a systematic study of the spin-wave dispersion relations and the low-temperature magnetization in the amorphous metallic ferromagnets $(Fe_xNi_{1-x})_{75}P_{16}B_6Al_3$ for $x = 1.0$, 0.65, 0.5, and 0.30. In this series the Ni is, to a first approximation, nonmagnetic. The spin-wave measurements were carried out using inelastic neutron scattering techniques, the magnetization at finite fields (1.2 and 15 kOe) was measured via a vibrating sample magnetometer and the temperature dependence of the magnetization at zero field was deduced from the hyperfine field at the Fe sites as measured by Mössbauer techniques. The spin waves follow a normal ferromagnetic dispersion relation, $\hbar\omega(\vec{q}) = \Delta + Dq^2 + Eq^4 + \cdots$, in all materials. The magnetization follows the law $M(T) = M_0(1 - BT^{3/2} - \cdots)$. The $T^{3/2}$ coefficient B is nearly field independent for the samples with $x = 1.0$, 0.65, and 0.5. Further the Mössbauer and magnetometer measurements are in close quantitative agreement. We find that for these samples the observed B's are properly accounted for by the measured spin-wave dispersion relations in contrast to previous studies in other magnetic glasses. Anomalous effects are observed in the $x = 0.30$ sample.

In the past few years a number of studies have been reported of the magnetic excitations and of the low-temperature magnetic properties of amorphous metallic ferromagnets, especially of the metal-metalloid class.[1-8] In general, it has been found that at long wavelengths many such glasses exhibit well-defined spin-wave excitations[9] with a normal ferromagnetic dispersion relation

$$\hbar\omega(\vec{q}) = \Delta + Dq^2 + Eq^4 + \cdots \quad (Dq^2/\Delta \gg 1), \tag{1}$$

where Δ is an effective anisotropy gap due to the dipole-dipole interactions.[1] Correspondingly, at low temperatures, the magnetization is observed to exhibit the usual ferromagnetic behavior

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

In simple spin-wave theory,[9] the $T^{3/2}$ coefficient B and the spin-wave stiffness coefficient D are related by the equation

$$B = 2.612\,[g\mu_B/M(0)]\,(k_B/4\pi D)^{3/2}. \tag{3}$$

One of the most puzzling aspects of the results reported to data is that although Eqs. (1) and (2) have been found to hold rather well in the metal-metalloid ferromagnets, B calculated from Eq. (3) is consistently 30%–40% smaller than the measured B.[1-9] That is, the magnetization drops off much more rapidly with increasing temperature than one would anticipate from the observed spin-wave dispersion relation. This would seem to necessitate the existence of additional excitations, possibly nonpropagating modes which would be inseparable from the background in the neutron experiments. However, these modes would have to have the peculiar feature that they exhibit a $\omega^{1/2}$ density of states at low energies. Low-lying nonpropagating excitations are well known in the lattice dynamics of disordered systems; indeed they are believed to give rise to the ubiquitous linear term in the specific heat in glasses. However, in a concentrated magnetic system in which all the spins are tightly coupled, it is difficult to construct a plausible model which contains localized excitations with an $\omega^{1/2}$ density of states. Hence the above discrepancy has remained unexplained.

In an attempt to elucidate this problem we have carried out a series of measurements on the ferromagnetic glasses $(Fe_xNi_{1-x})_{75}P_{16}B_6Al_3$. The general properties of these glasses including the method of preparation are discussed in Ref. 8. The spin-wave dispersion relations were measured using inelastic neutron scattering techniques. The mag-

netization was measured using a vibrating sample magnetometer in applied fields of 1.2 and 15 kOe. The temperature dependence of the magnetization in zero field was obtained via the hyperfine field at the Fe site. In all cases, measurements were carried out using either the same samples or, at the minimum, samples obtained from the same batch.

We consider firstly the neutron measurements. The experiments were carried out at the High Flux Beam Reactor at Brookhaven National Laboratory. Previously, results have been reported for two of the four compositions, $(Fe_{0.30}Ni_{0.70})_{75}P_{16}B_6Al_3$ (Ref. 5) and $(Fe_{0.65}Ni_{0.35})_{75}P_{16}B_6Al_3$ (Ref. 7). A complete description of the experimental techniques is given in Ref. 7. We have carried out similar measurements on the remaining two materials, $Fe_{75}P_{16}B_6Al_3$ and $(Fe_{0.50}Ni_{0.50})_{75}P_{16}B_6Al_3$, the former at $T=300$ K ($T_C=630$ K) and the latter at $T=80$ K ($T_C=482$ K). Due to neutron kinematical restrictions,[1,7] it was not possible to measure the pure iron sample at temperatures much below 300 K. However, as we shall show below, the extrapolation to 0 K represents only a minor correction. In both samples, sharp magnon excitations

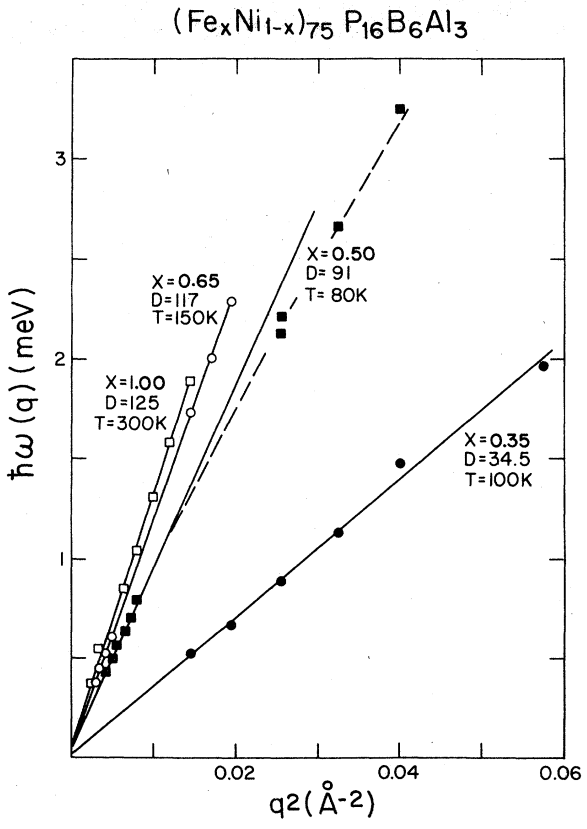

FIG. 1. Spin-wave dispersion relations in $(Fe_xNi_{1-x})_{75}P_{16}B_6Al_3$ for $x=1.0$, 0.65, 0.50, and 0.30.

are observed over the complete range of wave vectors accessible at long wavelengths. The spin-wave energies are plotted as a function of q^2 in Fig. 1. Here, we have also included data obtained in the previous studies.[5,7] In all cases, the data follow rather well the simple dispersion law Eq. (1). For the $x=0.50$ sample there is a suggestion of a measurable q^4 term.

In a more complete study of the $(Fe_{0.65}Ni_{0.35})_{75}P_{16}B_6Al_3$ sample, Tarvin *et al.*[7] showed that the spin-wave stiffness D renormalizes as

$$D(T)=D(0)[1-0.43(T/T_C)^{5/2}]. \quad (4)$$

The observed $T^{5/2}$ behavior is predicted by ordinary spin-wave theory. The prefactor, 0.43, should be only weakly sample dependent and, in the absence of individual measurements, we may take it as sample independent. The net corrections in extrapolating to 0 K from the data shown in Fig. 1 for $x=1.0$, 0.65, 0.50, and 0.30 are 6.7, 1.5, 0.5, and 4.0%, respectively. Thus, any error in the 0.43 prefactor (in $(Fe_{93}Mo_7)_{80}B_{10}P_{20}$ Axe *et al.*[1] find a prefactor of 0.61) will represent a negligible error in $D(0)$. The final results are given in Table I. We shall comment on these results after first presenting the Mössbauer and bulk measurements.

The magnetization as a function of temperature was measured using a recording vibrating sample magnetometer. A Chromel-Au (0.07-at.% Fe) thermocouple in contact with the sample was used to measure the temperature. For temperatures below 77 K, the reference junction was kept in liquid nitrogen; for temperatures above 77 K, the reference junction was in an ice bath. For all samples, with the exception of $(Fe_{0.30}Ni_{0.70})_{75}P_{16}B_6Al_3$, the measured magnetization is essentially field independent once the applied field is sufficiently large to overcome demagnetizing effects ($H\sim 700$ Oe). The magnetization of the $Fe_{0.30}Ni_{0.70}$ sample, however, is quite field dependent. A plot of $1-M/M_0$ as a function of $T^{3/2}$ for this sample taken with an applied field of 1.2 kOe is shown in Fig. 2. As seen from the figure, for temperatures less than 150 K, the behavior is well described by $1-M/M_0=BT^{3/2}$. For this sample $B=1.44\times10^{-4}$ ($K^{2/3}$) at 1.2 kOe and 1.25×10^{-4} ($K^{2/3}$) at 15 kOe. The Curie temperature was obtained by observing the disappearance of the ac susceptibility. All these magnetic results are summarized in Table I. The applied field used for the value of B in Table I was 15 kOe.

The Mössbauer measurements were carried out using the experimental techniques described in detail by Chien and Hasegawa.[6] The hyperfine field at the Fe site is plotted as a function of $(T/T_C)^{3/2}$ for all four samples in Fig. 3. As ex-

TABLE I. Spin-wave and magnetization parameters for $(Fe_xNi_{1-x})_{75}P_{16}B_6Al_3$ ferromagnetic glasses.

Compound	T_c(K)	M_0 (Gauss)	Moment per Fe (μ_B)	B (Mössbauer) (10^{-6} K$^{-3/2}$)	B (Magnetization) (10^{-6} K$^{-3/2}$)	$[D(0)]_{calc}$ (meVÅ^2)	$[D(0)]_{meas}$ (meVÅ^2)	$[D(0)]_{meas}/T_c$ (meVÅ^{2}K^{-1})
$(Fe)_{75}P_{16}B_6Al_3$	630 ± 12	1200	1.96	19.0 ± 2	18.6 ± 2	117 ± 10	134 ± 5	0.21
$(Fe_{0.65}Ni_{0.35})_{75}P_{16}B_6Al_3$	576 ± 6	832	2.05	28.0 ± 2	28.4 ± 2	114 ± 10	115 ± 3	0.20
$(Fe_{0.50}Ni_{0.50})_{75}P_{16}B_6Al_3$	482 ± 6	712	2.20	43.0 ± 3	46.3 ± 3	94 ± 10	91 ± 3	0.19
$(Fe_{0.30}Ni_{0.70})_{75}P_{16}B_6Al_3$	258 ± 3	475	2.39	117.0 ± 10	124 ± 10	61 ± 10	36 ± 3	0.13
Crystalline Fe	1042	1752	2.09		3.4	285	281	0.27

pected, the $T^{3/2}$ law holds rather well below 150 K. The final results for B as deduced from the hyperfine field measurements for all four samples are tabulated in Table I. Gratifyingly, the results are in close quantitative agreement with the magnetization measurements, especially for the $x = 1.0$, 0.65, and 0.50 samples.

As discussed in the beginning of this paper, ordinary spin-wave theory predicts that D and B should be directly related by Eq. (3). In implementing Eq. (3), we have taken $g = 2.09$, the value appropriate to crystalline Fe; we expect this to be accurate to at least 5%. The values for $D(0)$ calculated from the measured $M(0)$ and B are tabulated in Table I along with the values for $D(0)$ measured by neutron scattering. Perhaps surprisingly, for the $x = 1.0$, 0.65, and 0.50 samples the agreement is quite good. Thus, *ordinary spin-wave theory completely accounts for the low-temperature magnetic behavior of these ferromagnetic metallic glasses*. We find no evidence for subsidiary low-energy magnetic excitations. We can offer no unambiguous explanation for the discrepancies observed previously in other materials. As a note of caution, one should observe that because of kinematical restrictions the neutron experiments typically probe the spin-wave excitations only up to energies of ~ 30 K. On the other hand, B is typically determined by measurements of $M(T)$ up to 150 K in temperature; thus $M(T)$ effectively probes the magnetic excitations up to energies five times greater than those measured via neutrons. A breakdown of Eqs. (1)–(3) could occur at higher energies due to, for example, single-particle excitations or anomalously large q^6 etc., terms. This would undoubtedly account for the discrepancy observed in certain materials.

In our $(Fe_xNi_{1-x})_{75}P_{16}B_6Al_3$ series there is a significant disagreement for the $x = 0.30$ sample, but of the *opposite sign* to that observed previously. This system, however, differs in two essential

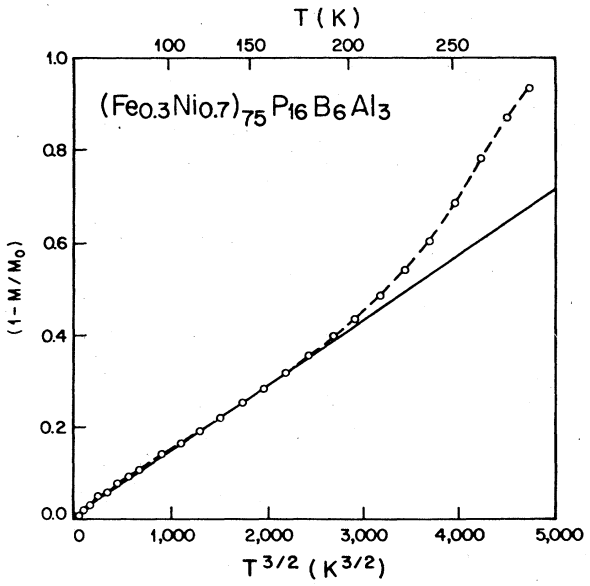

FIG. 2. Plot of $1 - M(T)/M(0)$ vs $T^{3/2}$ in $(Fe_{0.3}Ni_{0.7})_{75}P_{16}B_6Al_3$ as measured via a recording vibrating sample magnetometer. The applied field was 1.2 kOe.

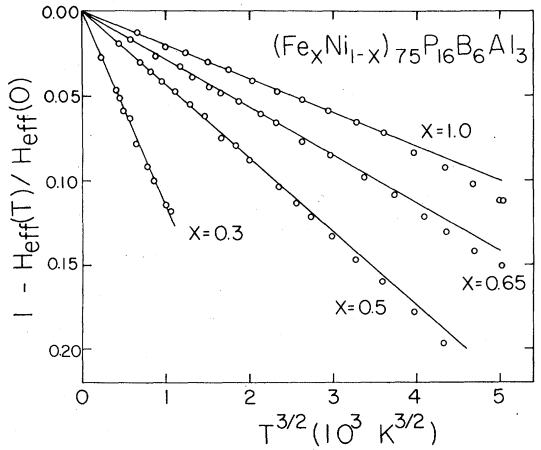

FIG. 3. Plot of $1 - H_{eff}(T)/H_{eff}(0)$ vs $T^{3/2}$ in $(Fe_xNi_{1-x})_{75}P_{16}B_6Al_3$.

ways from the other samples. Firstly, from Fig. 1 of Ref. 5, it is evident that the spin waves are never sharp excitations so that a spin-wave description is probably not valid. Secondly, the magnetization is markedly field dependent in a fashion which cannot be explained within the spin-wave picture. We believe, therefore, that in this sample where the Fe constitute only ~ 22% of the atoms, dilute alloy effects dominate completely. One might note that in the close-packed fcc lattice the nearest-neighbor percolation threshold occurs at 19.5% concentration. We are quite close to this threshold and one might therefore anticipate unusual effects as indeed are observed. A theory for amorphous magnets in this regime would be most welcome.

Finally, we note from Table I that D/T_C, which measures the range of the exchange interaction, is approximately constant for the $x = 1.0$, 0.65, and 0.50 materials. The observed ratio $D/T_C = 0.20 \pm 0.01$ (meV Å^{2}K^{-1}) is somewhat less than that in pure iron, $D/T_C = 0.27$, implying shorter-range interactions in the amorphous metals. This is consistent with the conclusion of Tarvin *et al.*[7] based on their spin-wave measurements.

ACKNOWLEDGMENTS

The work at the Massachusetts Institute of Technology and the Johns Hopkins University was supported by the NSF under contract Nos. DMR78-02397 and DMR77-09598, respectively. The work at the BNL was supported by the Division of Basic Energy Sciences, Department of Energy, Contract No. EY-76-C-02-0016.

[1] J. D. Axe, G. Shirane, T. Mizoguchi, and K. Yamauchi, Phys. Rev. B 15, 2763 (1977).
[2] J. D. Axe, L. Passell, and C. C. Tsuei, AIP Conf. Proc. 24, 119 (1974).
[3] R. W. Cochrane and G. S. Cargill III, Phys. Rev. Lett. 32, 476 (1974).
[4] H. A. Mook, N. Wakabayashi, and D. Pan, Phys. Rev. Lett. 34, 1029 (1975).
[5] J. W. Lynn, G. Shirane, R. J. Birgeneau, and H. S. Chen, AIP Conf. Proc. 34, 313 (1976).
[6] C. L. Chien and R. Hasegawa, Phys. Rev. B 16, 2115 (1977).
[7] J. A. Tarvin, G. Shirane, R. J. Birgeneau, and H. S. Chen, Phys. Rev. B 17, 241 (1978).
[8] E. M. Gyorgy, in *Metallic Glasses*, (Metals, Metals Park, Ohio, 1978), Chap. 11.
[9] For a review of elementary spin-wave theory see, F. Keffer in *Handbuch der Physik*, edited by S. Flügge (Springer-Verlag, Berlin, 1966), Vol. 18, Part 2, p. 1.