Spin waves in amorphous $Fe_{1-x}B_x$ alloys

J. J. Rhyne

National Measurement Laboratory, National Bureau of Standards, Washington, D.C. 20234

G. E. Fish

Corporate Research and Development, Allied Corporation, Morristown, New Jersey 07960

J. W. Lynn

Department of Physics and Institute for Physical Science and Technology, University of Maryland,[a] College Park, Maryland 20742

The temperature dependence of spin excitations has been studied in amorphous $Fe_{1-x}B_x$ (x = 0.18 and 0.14) by inelastic neutron scattering. The spin-wave stiffness constant D was determined directly from the magnon dispersion curves ($E = Dq^2$) over the temperature range from below room temperature ($T/T_C = 0.48$ and 0.36, respectively, for x = 0.18 and 0.14 alloys) up to 548 K ($T/T_C = 0.75$ and 0.99), which is just below the crystallization temperature. For both alloys the temperature dependence of D was found to be proportional to $(T/T_C)^{5/2}$ up to (T/T_C) greater than 0.8. The extrapolated values of D at T = 0 were D = 167 and 138 meV A^2, respectively, and were nearly twice as large as those determined from the $T^{3/2}$ coefficient found from magnetization studies. These anomalies may be related to the Invar characteristics of the thermal expansion in these alloys. Limited data were also obtained for an alloy $Fe_{76}B_{24}$ which exhibits a higher D (> 174 meV A^2).

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INTRODUCTION

Amorphous alloys of composition $Fe_{1-x}B_x$ can be prepared by melt-spinning techniques throughout the composition range (1,2) from x = 0.12 up to about x = 0.28. In the vicinity of x $\gtrsim$ 0.17, these alloys have been shown to exhibit Invar characteristics (3,4) in which their normal thermal contraction is nearly completely compensated by a large positive magnetostriction. As a function of increasing Boron composition, the Curie temperature increases from 509 K (x = 0.12) to 760 K (x = 0.28), and a corresponding decrease occurs in the Fe moment from 2.14 μ_B to 1.94 μ_B (1). The alloys show progressive crystallization at elevated temperatures (2). Initial crystallization temperatures range from 570 K for x = 0.12 and increase with boron concentration up to about 685 K for x = 0.26. For x $\gtrsim$ 0.20, the alloys crystallize below their Curie temperature. Hasegawa and Ray (2) have suggested a subtle change in structure from a bcc-like atomic stacking near x $\gtrsim$ 0.12 to a dense random packed arrangement for x $\gtrsim$ 0.20.

Magnetization measurements (1) have shown a proportionality between the magnetization and $T^{3/2}$, characteristic of linear spin-wave behavior, over an unusually wide range up to $T/T_C \gtrsim 0.33$. From the observed magnetization and the relationship

$$M(T) = M(0)\ (1 - B\ T^{3/2}) \tag{1}$$

values of the coefficient B have been determined (1). The spin-wave stiffness D predicted by the magnetization can then be calculated from

$$B = 2.6212\ \frac{g\mu_B}{M(0)} \left(\frac{k_B}{4\pi D}\right)^{3/2} . \tag{2}$$

The derived values of D can then be compared to those determined directly from the spin-wave dispersion as measured by inelastic neutron scattering,

$$E = E_g + D(T)\ q^2 + \ldots, \tag{3}$$

where E_g is the spin-wave energy gap at zero wavevector transfer q = 0. For most amorphous alloys, E_g is immeasurably small. Higher order terms in eq. (3) have also been found to be small.

EXPERIMENTAL

This paper reports results of inelastic neutron scattering on the amorphous alloys $Fe_{86}B_{14}$, $Fe_{82}B_{18}$, and $Fe_{76}B_{24}$, prepared in ribbon form by the inside-roll-chill-block melt-spinning technique in vacuum. Boron enriched to 98.5 percent ^{11}B was used to reduce the neutron absorption, and the ribbons were wound between two aluminum posts to produce a flat plate-like sample of optimum absorption thickness. Neutron data were taken in a vacuum furnace on a conventional triple-axis spectrometer at the National Bureau of Standards Reactor taking precautions to minimize air and sample-container scattering at the small q required for these studies.

Amorphous alloys require inelastic scattering experiments to be done near the forward beam position ([000] in reciprocal space), and thus neutron energy and momentum conservation conditions place severe limitations on the range of E and q space accessible for measurements. This is particularly critical for measurements on alloys with the larger spin stiffnesses D. The results reported here were taken with an incident neutron energy of 28 meV (also 14.8 meV for $Fe_{86}B_{14}$) obtained from a graphite monochromator with graphite filtering to reduce higher order wavelength contaminations. Söller slit collimations of angle 10', 12', 11', 16' were used before and after the monochromator and analyzer respectively, resulting in an overall intrinsic resolution width of 0.8 meV for elastic scattering. Depending on the alloy and temperature range, useful data were obtained from $q = 0.08\ \text{Å}^{-1}$ up to $q \geq 0.20\ \text{Å}^{-1}$.

RESULTS AND DISCUSSION

Figure 1 shows typical constant-q neutron scans for the $Fe_{86}B_{14}$ alloy as a function of both q and T. Spin waves for both neutron energy gain ($E < 0$) and energy loss ($E > 0$) are easily observable and well resolved up to temperatures where they merge with the central peak (elastic) scattering.

For the $Fe_{86}B_{14}$ alloy, which has a Curie temperature of 556 K, meaningful spin-wave data were obtained over the range from 200 K ($T/T_c = 0.36$) up to 548 K ($T/T_c = 0.99$). For $Fe_{82}B_{18}$ ($T_c = 617$ K), data were taken from room temperature ($T/T_c \approx 0.48$) up to 548 K ($T/T_c = 0.89$). The spin-wave linewidths observed for all temperatures below 0.9 T_c were that of the instrumental resolution. Above 500 K the $Fe_{86}B_{14}$ alloy, which has the lower Curie temperature, showed distinct linewidth broadening which increased rapidly as T_c was approached. At 513 K the observed width was 1.0 meV, almost double the instrumental width of 0.55 meV. (The intrinsic spin-wave width is less than the elastic resolution of 0.8 meV due to focussing effects.) After resolution corrections were applied, the dispersion data for these alloys were quadratic in q ($E = Dq^2$). The third alloy, $Fe_{76}B_{24}$, ($T_c = 723$ K) exhibits a higher spin stiffness which is almost beyond the range of these neutron experiments. Meaningful results could be obtained only at q = 0.08 Å and for temperatures above approximately 450 K ($T/T_c = 0.62$).

Figure 2 is a plot of the temperature dependence of the values of D for two alloys. As shown, the spin-

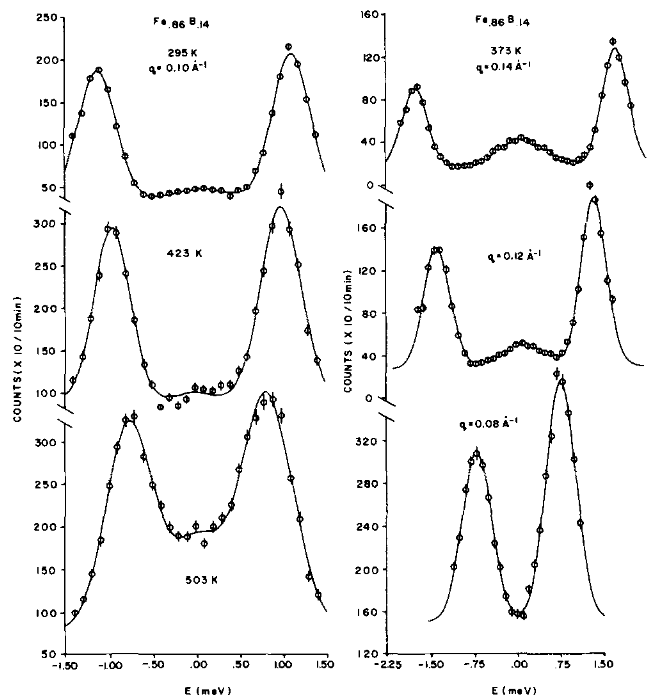

Fig. 1. (Left) Temperature dependence of magnon excitations in amorphous $Fe_{86}B_{14}$ at $q = 0.10\ \text{Å}^{-1}$. Solid lines are Gaussian least squares fits to the data. (Right) q-dependence of the spin waves for amorphous $Fe_{86}B_{14}$ at 373 K. Spin-wave peaks are shown for both neutron energy gain ($E < 0$) and energy loss ($E > 0$). Incident energy was 28 meV, and Söller slit collimations were 10-12-11-16 minutes (FWHM).

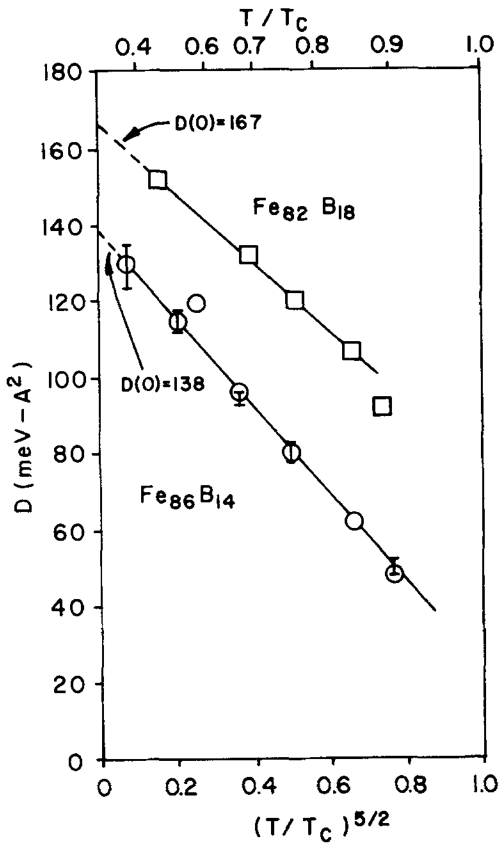

Fig. 2. Temperature dependence of spin-wave stiffness for $Fe_{86}B_{14}$ and $Fe_{82}B_{18}$. The data points are the average values for D obtained after resolution correction of the spin-wave energy for $0.06 < q < 0.10\ \text{Å}^{-1}$ ($Fe_{86}B_{14}$) and $q = 0.08\ \text{Å}^{-1}$ ($Fe_{82}B_{18}$). The error bars represent the variation in D obtained from the range of q's measured.

wave stiffness exhibits the following $T^{5/2}$ renormalization over virtually the entire temperature range studied:

$$D(T) = D(T = 0)\left(1 - a\left(\frac{T}{T_c}\right)^{5/2}\right) \qquad (4)$$

The validity of the $T^{5/2}$ relationship over this wide of a temperature region is anomalous and would be expected to result in significant departures of the magnetization from the $T^{3/2}$ low-temperature behavior (eq. 1). As mentioned earlier (1,5), these departures are not observed for $T < 0.33\ T_c$. The $(T/T_c)^{5/2}$ dependence of the spin-wave stiffness is consistent with that derived theoretically for a two-magnon interaction for a Heisenberg spin system and is the same behavior seen in crystalline Invar-type alloys $Fe_{65}Ni_{35}$ and Fe_3Pt (6) and in an independent study of the amorphous alloy $Fe_{86}B_{14}$ (7). The $Fe_{76}B_{24}$ alloy data do not appear to follow the $T^{5/2}$ dependence; however, this is not considered significant due to their limited range and lower reliability. The 0 K extrapolated values of D for each alloy and the slope D(0)a of eq. (4) are listed in Table I. For $Fe_{86}B_{14}$, values of $D(T = 0)$ are somewhat higher than that found by Ishikawa (7) et al. with a more rapid decrease of D with temperature than reported in ref. 6. They also find a non-zero value for the gap parameter E_g [eq. (3)] which we do not observe after accurately correcting the raw data for the calculated instrumental resolution. This correction was also verified experimentally by introducing additional vertical collimation. The origin of these discrepancies between our results and those of ref. 7 is being investigated. Also included in the table are

Table I. Neutron and Magnetization Determined Values of Spin Stiffness, D, for T = 0 K. The units of D are meV-A^2.

	$Fe_{86}B_{14}$	$Fe_{82}B_{18}$	$Fe_{76}B_{24}$	$Fe_{83}B_{16.5}Si_{0.5}$
D(T = 0) (neutron)	138 118 (ref. 6)	167	>175*	125 (ref. 7)
D(T = 0) (magnet.)	65	71	96	--
D(0)a	115 94 (ref. 6)	90	--	

*Approximate value, limited by energy-momentum restrictions of experiment.

values of D derived by Hasegawa and Ray (1) from their low temperature magnetization studies which values are in substantial agreement with those from Mössbauer results by Chien et al. (5). Comparison of these values illustrates the second anomaly of $Fe_{1-x}B_x$ spin-wave results. The neutron-determined D is almost twice as large as that calculated from magnetization. At the same time $D_{neutron}/T_c$ has approximately the same ratio (0.25 for $Fe_{82}B_{18}$ and 0.21 for $Fe_{86}B_{14}$) at room temperature as those found for other amorphous alloys (8) which do not exhibit similar discrepancies in the value of neutron- and magnetization-determined values of D. Similar anomalies have been reported for the $Fe_{65}Ni_{35}$ and Fe_3Pt crystalline alloys (5). These results suggest that both the wide-range of the $(T/T_c)^{5/2}$ dependence of the spin stiffness D and the anomalously small values of D may be intrinsic, but not yet understood, dynamic properties of the magnetic and magneto-elastic interactions peculiar to the Invar state. Preliminary examination of the temperature and q-dependence of the quasi-elastic peak intensity (see Fig. 1) shows that additional magnetic scattering is present and possibly represents a spin diffusion phenomena in these alloys. This is being investigated further as an explanation for the inability of the neutron-determined spin-wave stiffness to account for a significant portion of the reduction in bulk magnetization with temperature.

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