Spin dynamics of amorphous $Fe_{90-x}Ni_xZr_{10}$

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Neutron inelastic scattering experiments have been performed in order to study the long wavelength spin dynamics of the amorphous Invar system $Fe_{90-x}Ni_xZr_{10}$ (for $x = 5,10$). Spin waves were observed over the entire range of wave vectors (0.05–0.15 $Å^{-1}$) and temperatures (0.3–0.9 T_C) under study. The spin-wave energies are well described by the quadratic dispersion relation $E_q = Dq^2 + \Delta$, where Δ is a small gap due primarily to dipolar interactions. The stiffness parameter D renormalizes with temperature as $D(T) = D(0) \times [1 - A(T/T_C)^{5/2}]$ throughout the temperature range under study. The values of $D(0)$ corresponding to $x = 5$, 10, are 42.1 and 78.0 meV $Å^2$, respectively, with $A = 0.83$. In the temperature range 0.55–0.90 T_C the intrinsic spin-wave linewidths Γ_q can be fitted equally well to the q^4 dependence predicted by hydrodynamic spin-wave theory, or to the q^5 dependence predicted from the topological disorder in amorphous systems, but they did not show the T^2 behavior expected from magnon interactions. These findings suggest that there might be relevant spin-wave broadening mechanisms, perhaps due to the magnetic disorder in the system, in addition to magnon-magnon interactions.

INTRODUCTION

The magnetic properties of the iron-rich $Fe_{90-x}Ni_xZr_{10}$ amorphous system are very peculiar. When $x \to 0$ there is no uniform long-range ferromagnetic order: Small-angle scattering experiments on $Fe_{90}Zr_{10}$ have shown that the average spin correlation length at low temperatures remains small ($\approx$25 Å) and nearly temperature independent,[1] reflecting the effect of competing ferromagnetic and antiferromagnetic exchange interactions. The addition of Ni in the system, on the other hand, induces conventional long-range ferromagnetic order. In $Fe_{70}Ni_{20}Zr_{10}$,[2] for example, spin waves have been observed at temperatures between 0.55 and 0.89 T_C. It has also been shown that for $x = 10$, 20, the system exhibits Invar characteristics,[3] where the thermal expansion and a large positive spontaneous volume magnetostriction nearly cancel below the Curie temperature.

In order to obtain a better understanding of the magnetic properties of the iron-rich Fe-Ni-Zr alloys, we have studied the spin dynamics of this system for various concentrations of Ni. Our results for $x = 20$ have been reported already,[2] and in this paper we report results for $x = 5$, 10.

EXPERIMENTAL DETAILS

The samples used in our measurements were amorphous ribbons of $Fe_{90-x}Ni_xZr_{10}$ with concentrations $x = 5$ ($T_C = 306$ K) and $x = 10$ ($T_C = 359$ K). Ribbons (16 g for $x = 5$ and 9 g for $x = 10$) approximately 25 μm thick and 0.5 cm wide, were prepared by the inside-roll chill-block melt-spinning technique in vacuum, and were loosely wound between two aluminum posts to produce flat platelike samples. The neutron scattering experiments were performed on a triple axis spectrometer at the National Bureau of Standards Reactor, with pyrolytic graphite (PG 002) crystals used as monochromator and analyzer. The incident energy was fixed at 13.5 meV and a PG filter was placed after the monochromator to suppress higher-order wavelength contaminations. Soller slit horizontal collimators of 15′-12′-11′-25′ produced an energy resolution ΔE of 0.35 meV full-width-at-half-maximum (FWHM)] at the elastic position. The amorphous nature of the system under study required that measurements be taken near the forward (000) beam position. The wave vector transfers examined were in the range $0.07\ Å^{-1} \leqslant q \leqslant 0.15\ Å^{-1}$, and the temperature ranges of the experiments were 0.33–0.90 T_C for $Fe_{85}Ni_5Zr_{10}$, and 0.28–0.83 T_C for $Fe_{80}Ni_{10}Zr_{10}$.

EXPERIMENTAL RESULTS AND DISCUSSION

Well-defined spin waves were observed at all temperatures and wave vectors studied. In analyzing the spin-wave data, excitation energies and linewidth information were obtained by convoluting a theoretical cross section with the instrumental resolution and least-squares fitting to the observed spectra, after subtraction of the background scattering. Two forms of the spectral weight function in the spin wave cross section were used in this analysis: double Lorentzian and damped harmonic oscillator. Both forms yielded qualitatively similar results for the spin-wave energies and linewidths, and therefore in this paper we report only the results for the double Lorentzian-type cross section.

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The spin-wave energies were found to obey the quadratic dispersion relation expected for an isotropic ferromagnet:

$$E_q = \Delta + B(T)q^2 + \cdots , \qquad (1)$$

where $D(T)$ is the stiffness parameter, and Δ is an effective anisotropy gap which originates mainly from dipolar interactions. For both concentrations $\Delta \approx 0.06$ meV and showed no significant temperature dependence. The quadratic dispersion is illustrated in Fig. 1 for $Fe_{85}Ni_5Zr_{10}$ at $T = 100$, 175, 225, 250, and 275 K. A similar plot was obtained for $Fe_{80}Ni_{10}Zr_{10}$.

The spin-wave stiffness parameter $D(T)$ can be obtained from the slopes of the dispersion curves of the type of Fig. 1. The two-magnon interaction theory of a Heisenberg ferromagnet predicts a low-temperature dependence of $D(T)$ of the form[4]

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

In Fig. 2 we have plotted $D(T)$ vs $(T/T_C)^{5/2}$ for $Fe_{90-x}Ni_xZr_{10}$ ($x = 5$, 10, 20). The values corresponding to $x = 20$ have been taken from Ref. 2. In all cases the values of $D(T)$ were found to be in agreement with the $(T/T_C)^{5/2}$ dependence of Eq. (2). The values of the stiffness parameter extrapolated to $T = 0$ were found to be $D(0) = 42.05 \pm 0.37$ and 78.03 ± 0.49 meV Å^2 for the samples with $x = 5$ and 10, respectively, and for both samples $A = 0.83$. This value of A is of the same order of magnitude as the ones found in the Fe-B amorphous Invar system,[5] and its relatively large magnitude (i.e., $A \approx 1$) means that the low-temperature expansion of $D(T)$ [Eq. (2)] should represent the stiffness parameter data over a wide range of temperatures, as found in Fig. 2.

The observed spin-wave peaks will be broadened by any intrinsic linewidth. The magnitude of these linewidths can be extracted from the spin-wave data by convoluting the cross section with the instrumental resolution and least-squares fitting the energy, linewidth, and scattering strength to the data. Typically, reliable linewidth data can be obtained in this manner when the linewidths Γ_q are greater than about 30% of the instrumental energy resolution. Figure 3 shows the plot of the linewidths Γ_q (FWHM of the Lorentzian spectral weight function), for $x = 5$, vs q for $T = 175$, 225, 250, and 275 K. The error bars shown are statistical only (i.e., no systematic errors have been included).

If the dominant spin-wave broadening mechanism in the system is due to magnon-magnon interactions, then the

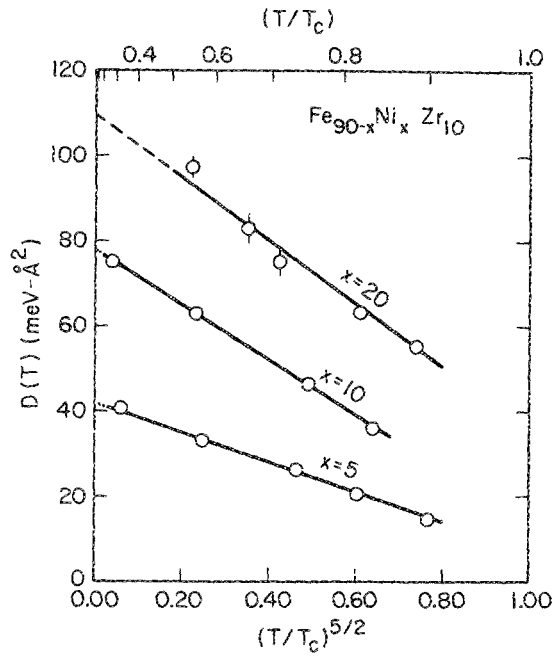

FIG. 2. Stiffness constant $D(T)$ vs $(T/T_C)^{5/2}$ for $Fe_{90-x}Ni_xZr_{10}$, with $x = 5$, 10, and 20. The values for $x = 20$ are from Ref. 2. In all cases $D(T)$ renormalizes with temperature according to the prediction of the two-magnon interaction theory of a Heisenberg ferromagnet [Eq. (2)].

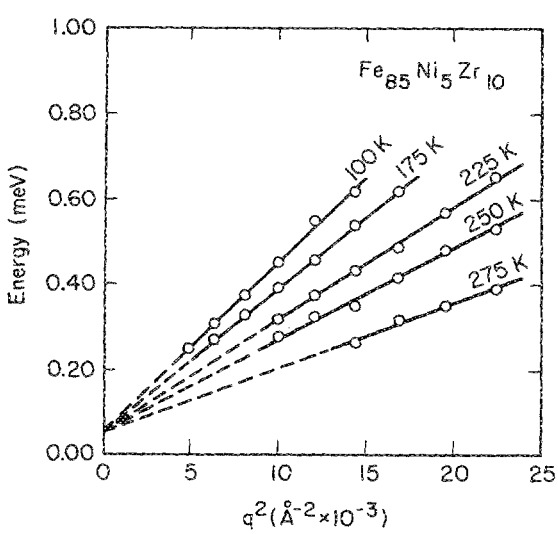

FIG. 1. E_q vs q^2 for $Fe_{85}Ni_5Zr_{10}$ ($T_C = 306$ K) at $T = 100$, 175, 225, 250, and 275 K. The spin-wave energies obey the quadratic dispersion relation of Eq. (1) in the range of temperatures and wave vectors under study.

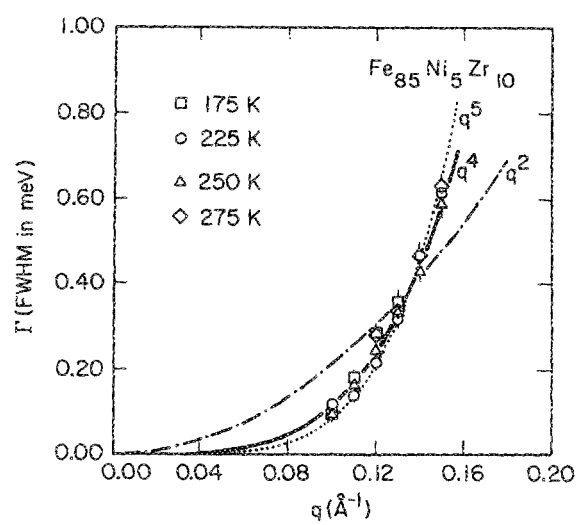

FIG. 3. Intrinsic spin-wave linewidth Γ_q (FWHM of the Lorentzian spectral weight function) vs q for $Fe_{85}Ni_5Zr_{10}$ ($T_C = 306$ K) at $T = 175$, 225, 250, and 275 K. The linewidths can be fitted to a q^4 or q^5 dependence, with limited temperature dependence. The dot-dash line is the result of the fit to the q^2 form suggested by Ishikawa *et al.* (Ref. 7). The instrumental energy resolution (FWHM at the elastic position) was $\Delta E = 0.35$ meV.

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linewidths Γ_q are expected to follow a wave vector and temperature dependence of the form[6]

$$\Gamma_q(T) \propto q^4[T\ln(kT/E_q)]^2, \tag{3}$$

provided that $E_q \ll kT$. On the other hand, Ishikawa *et al.*[7] have suggested that the spin-wave intrinsic linewidths in amorphous and crystalline Invar systems do not obey Eq. (3), but rather the empirical relation $\Gamma_q \propto q^2(1+bT^\alpha)$, where $\alpha \leqslant 1$. In addition to the thermal broadening of the spin-wave excitations, broadening due to the topological disorder is also expected in amorphous systems. Singh and Roth,[8] Mano,[9] and Ishkakov[10] have predicted that this additional broadening is of the form $\Gamma \propto q^5$, with no temperature dependence, similar to that expected for dilute crystalline ferromagnets.

Our analysis revealed that the linewidths Γ_q for both samples did not show significant temperature dependence in the temperature range 0.55–0.90 T_C. These linewidth data were fitted to (a) the q^4 dependence of Eq. (3), (b) the q^5 dependence predicted from the topological disorder in amorphous systems, and (c) the empirical q^2 form suggested by Ishikawa *et al.*[7] The results of the least-squares fits are shown in Fig. 3. It can be seen that the q^2 form is clearly inappropiate, while the q^4 and q^5 forms both adequately represent the linewidth data. In order to distinguish between the q^4 and q^5 forms more data will need to be taken.

In order to compare the relative magnitudes of the spin-wave damping in the Fe-Ni-Zr system we have plotted (see Fig. 4) $1/2\ \Gamma_q/E_q$ [i.e., the ratio of the half width at half maximum (HWHM) linewidth to the spin-wave energies] for $q = 0.12\ \text{Å}^{-1}$ vs T/T_C. Although Γ_q in our system was almost temperature independent at the temperatures shown in Fig. 4, the ratio $1/2\ \Gamma_q/E_q$ shows a strong temperature dependence due to the renormalization of the spin-wave energies with temperature. In this figure we have also included linewidth data from the amorphous Invar system $Fe_{86}B_{14}$,[5] which have been found to be in agreement with the magnon-magnon interaction prediction (including the temperature dependence) of Eq. (3). It can be seen that the relative damping in Fe-Ni-Zr is significantly larger than in the Fe-B system. These findings suggest that there might be spin-wave broadening mechanisms, presumably due to the magnetic disorder, in addition to the magnon-magnon interaction in the Fe-Ni-Zr system.

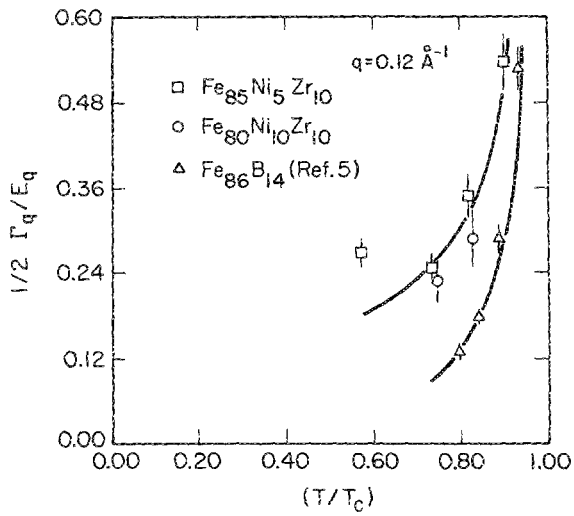

FIG. 4. Ratio of the half width at half maximum (HWHM) of the Lorentzian linewidths vs T/T_C for Fe-Ni-Zr, and $Fe_{86}B_{14}$ (after Ref. 5). The relative damping of the spin-wave excitations in the Fe-Ni-Zr system is significantly larger, indicating that there may be additional broadening mechanisms in this system besides magnon-magnon interactions. The solid lines are guides to the eye.

ACKNOWLEDGMENT

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