Low-temperature spin waves in amorphous $Fe_{90-x}Ni_xZr_{10}$

J. A. Fernandez-Baca
Solid State Division, Oak Ridge National Laboratory, Oak Ridge, Tennessee 37831 and Center of Materials Science, National Bureau of Standards, Gaithersburg, Maryland 20899

J. W. Lynn
Department of Physics, University of Maryland, College Park, Maryland 20742 and Center of Materials Science, National Bureau of Standards, Gaithersburg, Maryland 20899

J. J. Rhyne
Center of Materials Science, National Bureau of Standards, Gaithersburg, Maryland 20899

G. E. Fish
Allied Signal, Inc., Morristown, New Jersey 07960

Inelastic neutron scattering measurements have been performed on rapid-quenched ribbons of $Fe_{90-x}Ni_xZr_{10}$ ($x = 1,5,10$) to measure the small wave-vector spin-wave spectrum. At all three concentrations a quadratic dispersion relation typical of an isotropic ferromagnet is observed, but with anomalous properties at low temperatures. For $x = 5$ and 10 the excitations are found to broaden considerably for temperatures $\leqslant 0.3\,T_c$. For $x = 1$ the spin waves not only broaden for $T \leqslant 0.5T_c$, but the spin-wave energies decrease monotonically with decreasing temperature while a resolution-limited quasielastic component of the scattering develops. These results are consistent with a picture of competing ferromagnetic and antiferromagnetic interactions, with reentrant spin-glass behavior evident if the frustration is sufficiently strong.

The unusual compositional dependence of the magnetic properties of the amorphous Invar $Fe_{90-x}Ni_xZr_{10}$ system is of considerable interest. Bulk magnetization and susceptibility measurements show a paramagnetic to "ferromagnetic" transition at a temperature T_c that *decreases* with decreasing Ni content.[1] Furthermore, small-angle neutron scattering measurements have shown[2] that in the iron-rich limit the spin fluctuations are correlated over relatively short ranges of about 100 Å, but static domains also exist which are approximately five times larger. These results indicate that the phase below T_c is not a true ferromagnetic state. The appearance of an irreversible susceptibility below $T_F \approx 80$ K has been interpreted as a transition to a spin-glass-like phase[1] and the low-temperature magnetization fails to saturate in fields as large as 19 T.[3] These observations have suggested the description of amorphous $Fe_{90}Zr_{10}$ as a "wandering axis ferromagnet" with a low-temperature asperomagnetic phase.[3] This unusual magnetic ordering would be a consequence of strong competition between ferromagnetic and antiferromagnetic exchange interactions originating from a distribution of Fe-Fe nearest-neighbor distances.[1-3]

The partial substitution of Fe by Ni modifies this distribution, which can result in drastic changes of the magnetic properties, and favors long-range ferromagnetic order. To obtain a better understanding of how this transition to conventional ferromagnetism occurs we have undertaken a program to study the spin dynamics of $Fe_{90-x}Ni_xZr_{10}$ for various Ni concentrations x. For $x = 5$, 10, 20, we have reported[4] the existence of well-defined spin waves with a quadratic dispersion relation of the form

$$E_q = D(T)q^2 + \Delta. \tag{1}$$

In this equation D is the spin-wave stiffness parameter and Δ is a small effective gap in the spectrum originating from dipolar interactions. The stiffness parameter D renormalizes with temperature as

$$D(T) = D(0)[1 - A(T/T_c)^{5/2}], \tag{2}$$

as predicted by the magnon-magnon interaction theory of a Heisenberg ferromagnet. The value $D(0)$ increases with increasing Ni content x. In addition, $D(0)$ is consistently higher than the value estimated from bulk magnetization measurements, as found in other Invar alloys. The temperature dependence of the spin-wave intrinsic linewidths was found to be in disagreement with the T^2 form predicted from the two-magnon interaction theory.[5] Instead the linewidths were found to be independent of T in the range $0.55 \leqslant T/T_c \leqslant 0.90$, indicating that there are relevant spin-wave broadening mechanisms other than two-magnon interactions.

In this paper we report new results obtained at lower temperatures for $x = 5$, 10, as well as results for a new sample with $x = 1$.

RESULTS AND DISCUSSION

The samples used in our experiments were amorphous ribbons of $Fe_{90-x}Ni_xZr_{10}$ ($x = 1$, 5, 10) prepared by the flow casting technique in vacuum. The ribbons, approximately 25 μm thick and 0.5 cm wide, were loosely wound between two aluminum posts to produce flat platelike samples. The samples were then placed in a Displex closed-cycle refrigerator. The neutron scattering experiments were performed on the BT-4 triple axis spectrometer at the National Bureau of Standards Reactor, with pyrolytic graphite (PG 002) crystals used as monochromator and analyzer. The neutron incident energy was fixed at 3.7, 4.9, or 13.5 meV for $x = 1$, 5, and 10, respectively. In order to suppress higher-order wavelength contaminations a filter (cooled beryllium for $E_i = 3.7$ and 4.9 meV, and PG for $E_i = 13.5$ meV) was

 0021-8979/88/083749-03$02.40

placed in the incident beam. Suitable Söller slit horizontal collimators were chosen to produce energy resolutions ΔE (FWHM) of 0.065 meV ($E_i = 3.7$ meV), 0.089 meV ($E_i = 4.9$ meV), or 0.40 meV ($E_i = 13.5$ meV).

In analyzing the spin-wave data, excitation energies and linewidth information were obtained by least-squares fitting the inelastic scattering spectra to a parametrized theoretical cross section convoluted with the instrumental resolution function. Two forms of the spectral weight function were used in this analysis: Double-lorentzian and damped harmonic oscillator. Both forms of the spectral weight function yielded qualitatively similar results and in this paper we make reference only to the results corresponding to the double lorentzian-type cross section.

(a) $Fe_{89}Ni_1Zr_{10}$ ($T_c = 248$ K)

Constant-q scans for wave vectors 0.04 $Å^{-1} \leqslant q \leqslant 0.10$ $Å^{-1}$ at temperatures 0.04 $T_c \leqslant T \leqslant 0.91$ T_c were taken for $Fe_{89}Ni_1Zr_{10}$. Figure 1 shows the constant-q spectra obtained for $q = 0.08$ $Å^{-1}$ at $T = 10, 30, 50, 75$, and 125 K. The peaks for energy gain ($E<0$) and energy loss ($E>0$) correspond to annihilation and creation of spin-wave excitations, and the (resolution-limited) peak at $E = 0$ corresponds to elastic and quasielastic scattering from the sample and environment. Due to resolution effects and the fact that our measurements were performed with fixed incident energy, the peaks on the energy-gain side of the spectra appear larger than the peaks on the energy-loss side. The solid lines shown in the figure are the result of the least-squares fits.

In the analysis of the inelastic scattering data for this concentration, we have identified three different temperature regions, corresponding to $T = 125$–225 K ($0.5T_c$–$0.91T_c$), 50–125 K ($0.2T_c$–$0.5T_c$), and 10–50 K ($0.04T_c$–$0.2T_c$). In the $T = 125$–225 K region, well-defined excitations that are consistent with the dispersion relation of Eq. (1) are observed. In addition, the temperature dependence of the stiffness parameter D is consistent with Eq. (2), with $D(0) = (24.57 \pm 0.72)$ meV $Å^2$. However, in this region the spin-wave intrinsic linewidths appear to be temperature independent.

In the $T = 50$–125 K region well-defined excitations which are consistent with Eq. (1) are also observed. However, the stiffness parameter D decreases with decreasing temperature with a concomitant increase in the damping of the excitations, while the intensity of the resolution-limited peak at $E = 0$ increases considerably. This increase of the quasielastic scattering intensity is associated with the development of a spin-glass-order parameter.[6] Hence we find in this region a coexistence of spin-wave excitations and spin freezing phenomena, similar to that found in other "reentrant spin-glass" systems.[7]

Below $T = 50$ K no well-defined excitations can be identified. In this region the inelastic scattering spectra can be well described either by overdamped spin waves with energies that continue to decrease with the decreasing temperature and collapse at $T = 10$ K, or by a spin diffusion cross section with no propagating features. In both cases the inelastic scattering coexists with the intense resolution-limited peak at $E = 0$, which continues to increase with the decreasing temperature.

(b) $Fe_{85}Ni_5Zr_{10}$ ($T_c = 306$ K)

Constant-q scans for wave vectors 0.04 $Å^{-1} \leqslant q \leqslant 0.10$ $Å^{-1}$ in the temperature range $0.07\,T_c \leqslant T \leqslant 0.33 T_c$ were taken for this sample. Figure 2 shows the spectra obtained at $q = 0.08$ $Å^{-1}$ for $T = 20, 40, 60, 100$ K. At all temperatures under study well-defined spin waves were identified that are consistent with Eq. (1). Unlike the case for $x = 1$ no indication of softening of the spin-wave modes at low temperatures

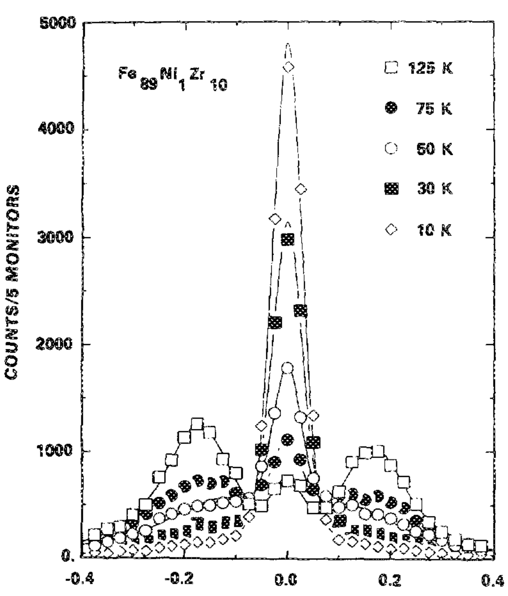

FIG. 1. Constant-q scans at $q = 0.08$ $Å^{-1}$ for several temperatures for amorphous $Fe_{89}Ni_1Zr_{10}$. The intensities have been normalized to 5 monitor counts (≈ 5 min). The solid lines are the result of the least-squares fit to a double lorentzian-type cross section convoluted with the instrumental resolution, as explained in the text.

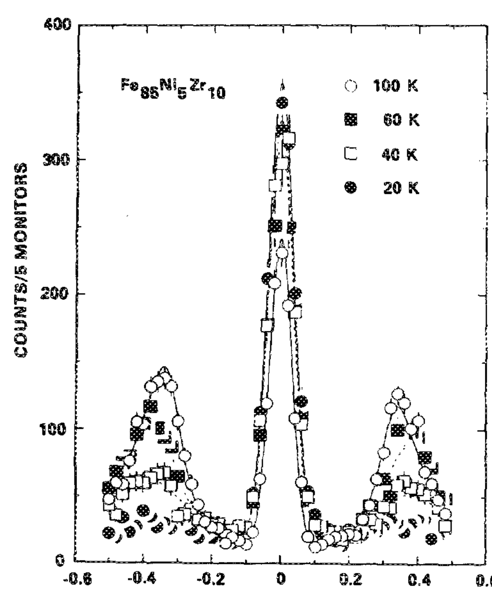

FIG. 2. Constant-q scans at $q = 0.08$ $Å^{-1}$ for amorphous $Fe_{85}Ni_5Zr_{10}$. The solid lines are from the fits.

was observed, and the temperature dependence of the stiffness parameter D was consistent with Eq. (2) with $D(0) = (46.90 \pm 0.72)$ meV Å^2. In addition, the resolution-limited peak at $E = 0$ showed no significant temperature dependence, unlike the findings for $x = 1$. The lower-temperature excitations, however, are considerably broadened and this broadening is strongly q dependent. This low-temperature broadening causes the apparent increase of the elastic peak intensity with decreasing temperature evident in Fig. 2.

(c) $Fe_{80}Ni_{10}Zr_{10}$ ($T_c = 359$ K)

Constant-q scans for wave vectors 0.06 Å$^{-1} \leqslant q \leqslant 0.09$ Å^{-1} and temperatures $0.06T_c \leqslant T \leqslant 0.28T_c$ were taken for this sample. At the lowest temperature studied ($T = 20$ K) no excitations could be identified due to the coarse resolution of these measurements ($\Delta E = 0.40$ meV) and the small thermal population factor in the spin-wave cross section. For all the other temperatures the results are qualitatively similar to those for the $x = 5$ sample. Above 40 K well-defined spin waves were identified, with no indication of softening at low temperatures. The spin-wave stiffness parameter D renormalized with temperature according to Eq. (2), with $D(0) = (74.2 \pm 1.3)$ meV Å^2. No indications of a low-temperature elastic (or resolution-limited quasielastic) component of the magnetic scattering were found, but the spin-wave excitations broadened considerably at lower temperatures.

In order to illustrate the enhancement of ferromagnetic ordering by the partial substitution of Fe by Ni in $Fe_{90-x}Ni_xZr_{10}$, we have plotted in Fig. 3 the ratio D/kT_c vs T/T_c for $x = 1, 5, 10, 20$. The higher-temperature values of D/kT_c for $x = 5, 10, 20$ have been taken from our previous measurements. The solid lines are the result of least-squares fits to the two-magnon interaction form of renormalization of the stiffness parameter [Eq. (2)]. The ratio D/kT_c is directly related to the mean-square range of the exchange interaction, and clearly decreases with decreasing x. The substitution of Fe by Ni increases the average Fe-Fe distance and thereby decreases the competition between ferromagnetic and antiferromagnetic interactions. A similar enhancement of ferromagnetism has also been obtained by partial substitution of Fe by Co and by hydrogenation.[1,3]

The onset of the low-temperature softening of the spin-wave energies for $x = 1$ occurs at a temperature ($T \approx 125$ K) considerably higher than the reported temperature where $Fe_{90}Zr_{10}$ (no Ni) enters a asperomagnetic phase ($T_F \approx 40$ K).[3] The lack of low-temperature spin-wave softening for $x = 5$ and 10 would indicate that, at these Ni concentrations, the short-range asperomagnetic phase is gradually replaced by a more aligned structure. The low-temperature broadening of the spin waves at these concentrations, however, would indicate that there remains some noncollinearity of the spins in the ground state. The origin of this broadening might be the superimposition of spin waves and a diffusive component, such as the elementary excitations of the noncollinear configuration of spins suggested by Continentino and Rivier.[8] These additional modes would cause an additional $T^{3/2}$ decrease of the magnetization and would explain the discrepancy between the stiffness parameter $D(0)$ obtained from neutron scattering and magnetization measurements.

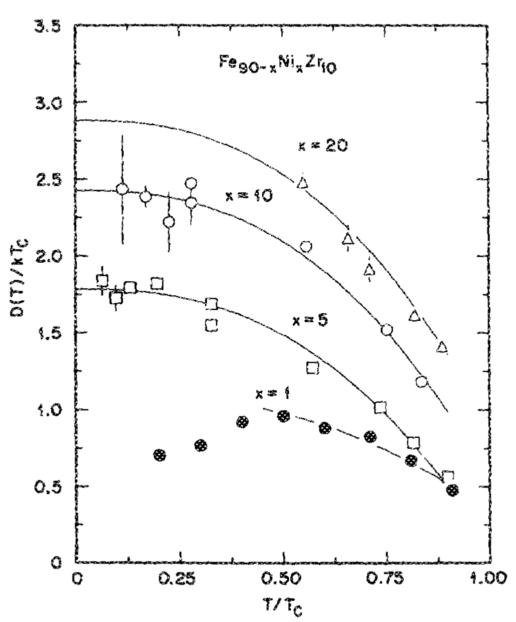

FIG. 3. Plot of D/kT_c vs T/T_c for $Fe_{90-x}Ni_xZr_{10}$ for $x = 1, 5, 10, 20$. The decrease of D/kT_c with decreasing Ni content is indicative of strong competition between ferromagnetic and antiferromagnetic interactions.

ACKNOWLEDGMENTS

The work at ORNL was supported by the U. S. Department of Energy under contract No. DE-AC05-840R21400 with Martin Marietta Energy Systems, Inc. The work at the University of Maryland was supported by the National Science Foundation, grant No. DMR 86-20269.

[1] K. Shirakawa, S. Ohnuma, M. Nose, and T. Masumoto, IEEE Trans. Magn. MAG-16, 910 (1980); *ibid.* 1129 (1980).

[2] J. J. Rhyne, R. W. Erwin, J. A. Fernandez-Baca, and G. E. Fish (these proceedings).

[3] D. H. Ryan, J. M. D. Coey, E. Batalla, Z. Altounian, and J. O. Strom-Olsen, Phys. Rev. B **35**, 8630 (1987), and references therein.

[4] J. A. Fernandez-Baca, J. W. Lynn, J. J. Rhyne, and G. E. Fish, J. Appl. Phys. **61**, 3406 (1987); J. A. Fernandez-Baca, J. J. Rhyne, and G. E. Fish, J. Magn. Magn. Mater. **54-57**, 289 (1986).

[5] A. B. Harris, Phys. Rev. **175**, 674 (1968); **184**, 606 (1969).

[6] J. W. Lynn, R. W. Erwin, H. S. Chen, and J. J. Rhyne, Solid State Commun. **46**, 317 (1983).

[7] For a review see J. W. Lynn and J. J. Rhyne, in *Spin Waves and Magnetic Excitations*, edited by A. S. Borovik-Romanov and S. K. Sinha (North-Holland, Amsterdam, 1988), Part II, Chap. 4; S. M. Shapiro, *ibid.*, Part II, Chap. 5.

[8] M. A. Continentino and N. Rivier, J. Phys. F **9**, L145 (1979); J. Magn. Magn. Mater. **15–18**, 1419 (1980).