NEUTRON SCATTERING STUDIES OF AMORPHOUS INVAR ALLOYS

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ABSTRACT

This paper reviews recent inelastic neutron scattering experiments performed to study the spin dynamics of two amorphous Invar systems: $Fe_{100-x}B_x$ and $Fe_{90-x}Ni_xZr_{10}$. As in crystalline Invar $Fe_{65}Ni_{35}$ and Fe_3Pt , the excitation of conventional long-wavelength spin waves in these amorphous systems can not account for the relatively rapid change of their magnetization with temperature. These results are discussed in terms of additional low-lying excitations which apparently have a density of states similar to the spin waves.

Keywords: Invar effect, amorphous magnetism, neutron scattering.

1. INTRODUCTION

Neutron scattering studies on crystalline Invar $Fe_{65}Ni_{35}$ and Fe_3Pt have revealed that in these systems the excitation of conventional spin waves cannot account for the relatively rapid decrease of their magnetization with temperature. This finding led Ishikawa et al. [1] to propose the existence of "hidden" excitations which, in addition to conventional spin waves, contribute to the anomalous decrease of the magnetization with temperature. According to these authors these excitations should be spin-wave-like (*i.e.*, with a dispersion relation similar to that of the spin waves) but with linewidths different from those predicted by conventional spin-wave models. No clues were given, however, about the origin of these excitations. This anomalous decrease of the magnetization has, since then, been observed in a variety of other crystalline and iron-rich amorphous systems that also exhibit Invar anomalies (anomalous thermal expansion, large spontaneous and forced volume magnetostriction, large high-field susceptibility, and a negative pressure dependence of the Curie temperature T_c). Thus it is believed that this anomaly is an intrinsic dynamic property peculiar to the Invar state [2].

In order to obtain a better understanding of this problem, and in particular to probe the existence of these "hidden" excitations, we have performed a systematic neutron scattering study of the spin dynamics of two amorphous Invar systems: $Fe_{100-x}B_x$ ($x = 14, 18$) and $Fe_{90-x}Ni_xZr_{10}$ ($x = 5, 10, 20$). The results of these experiments are reviewed in this paper (detailed accounts of these experiments can be found in references [3-6]).

The advantage of studying these amorphous systems is that they can be prepared in a variety of concentrations without having the complications derived from the vicinity to the Martensitic transformation found in the crystalline Invar systems. These amorphous alloys have coordination numbers of about 12 (reminiscent of the *fcc* structure) and average nearest-neighbor distances which are in the vicinity of the *fcc*-Fe interatomic distance where the interaction between the moments changes from ferromagnetic (FM) to antiferromagnetic (AF) [7-8]. Thus in these systems a fair amount of spin frustration, originating from the distribution of FM and AF exchange, can be introduced by varying the

iron concentration. Significant amounts of frustration may result in non-collinear magnetic structures that can range from spin canting [9] to asperomagnetism [10]. From the two systems reviewed in this paper $Fe_{100-x}B_x$ exhibits the least degree of frustration and the highest ordering temperatures ($T_c = 617, 556\text{ K}$ for $x = 18, 14$). Recent studies by Cowley et al. [11] on $Fe_{83}B_{17}$ have suggested, however, that even this system shows signs of spin canting. The effects of frustration are more evident in $Fe_{90-x}Ni_xZr_{10}$ where T_c decreases from 455K for $x = 20$ to 250K for $x = 1$, with reentrant spin-glass (RSG) behavior evident for $x = 1$. [6] In order to distinguish between properties that might be relevant to the Invar effect and those which might be influenced by the presence of RSG phases, in this paper we review only the results obtained at the concentrations that do not exhibit signs of RSG behavior.

Like Ishikawa et al. [1] we concluded that the rapid decrease in the magnetization, characteristic of the Invar alloys, should be discussed in terms of low-lying magnetic excitations in addition to spin waves. These excitations apparently have a density of states similar to the spin waves since they contribute to the renormalization of spin-wave stiffness coefficient with temperature in the same manner as conventional spin waves. Unlike Ishikawa et al., however, we did not find anomalies in the wavevector dependence of the spin-wave linewidths.

2. SPIN DYNAMICS OF AMORPHOUS INVARI ALLOYS

The low-temperature long-wavelength elementary excitations of amorphous magnets are spin waves (or magnons) [12]. In the hydrodynamic limit (wavelengths much longer than the interatomic spacing), and in the regime of small dipolar effects, the energy of these excitations is given by:

$$E_q = \Delta + Dq^2 + \dots \quad (1)$$

In this expression D is the (exchange) stiffness coefficient, q is the magnitude of the wavevector and Δ is a small effective (pseudo-) gap due to dipolar interactions. The magnitude of the stiffness coefficient D in this expression depends on the microscopic details of the system but the form of this equation is model independent. Each spin wave reduces the magnetization of the system by one unit and $M(T)$, the magnetization at temperature T , can be calculated if the average number of spin waves excited at this temperature is known. In the limit of low temperatures and if spin waves are the only low temperature excitations $M(T)$ is given by:

$$M(T) = M(0)[1 - BT^{3/2}], \quad (2)$$

where $M(0)$ is the magnetization at $T = 0$ and

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

It has been shown that interactions between magnons result in the renormalization of the stiffness coefficient D , as well as in the reduction of the lifetime of the spin-wave excitations (*i.e.*, a increase in the spin-wave intrinsic linewidths). The two-magnon-interaction theory

of the Heisenberg ferromagnet predicts a low-temperature renormalization of the stiffness coefficient D given by the expression :

$$D(T) = D(0) [1 - A (T/T_c)^{5/2} + \dots], \quad (4)$$

where $D(0)$ is the spin-wave stiffness at $T = 0$. This theory also predicts a spin-wave intrinsic linewidth Γ_q of the form [13]

$$\Gamma_q(T) \propto q^4 [T \ln(k_B T/E_q)]^2. \quad (5)$$

In amorphous systems additional broadening of the spin waves, originating from the topological disorder, should be expected. This type of broadening, which has been predicted to have a q^5 or a q^7 dependence [14], has not been definitely observed yet.

The most direct method of studying the spin dynamics of magnetic systems is inelastic neutron scattering. This technique has been used to study the spin dynamics of many transition-metal-based amorphous ferromagnets [12]. In all of these materials spin waves were observed at small wavevectors q , which obey the dispersion relation of Eq. (1) with a spin-wave stiffness coefficient $D(T)$ that renormalizes with temperature consistently with the two-magnon-interaction prediction of Eq. (4). The low-temperature bulk magnetization measurements in these systems are usually in agreement with Eq. (2), with a $T^{3/2}$ behavior up to $0.2-0.4T_c$. It has also been observed that in the non-Invar ferromagnets the stiffness coefficient D_m derived from the coefficient B of the low-temperature dependence of the bulk magnetization [Eq. (2)] and Eq. (3) has the same value of the coefficient $D(0)$ determined from the neutron scattering measurements [Eq. (4)] [15]. This agreement indicates that the low-temperature decrease of the magnetization is due to the excitation of spin waves only, as it was assumed to obtain Eq. (2). This type of agreement, however, has not been found in Invar systems where the value $D(0)$ obtained from neutron scattering measurements is consistently higher than the value D_m obtained from the bulk magnetization measurements [2]. This discrepancy, which is common to both amorphous and crystalline Invar systems, suggests that in these systems there must be other excitations, in addition to the conventional spin waves, that contribute to the decrease of the magnetization with temperature. These should be low-lying excitations in order to contribute to the decrease of the magnetization even at the lowest temperatures.

Ishikawa et al. [1] proposed that these “hidden” excitations have a dispersion relation similar to that of spin waves, but with linewidths different from those predicted by conventional spin-wave models. From the results of their inelastic neutron scattering experiments on crystalline $Fe_{65}Ni_{35}$, Fe_3Pt , and amorphous $Fe_{86}B_{14}$ [16], they suggested that the linewidths of their magnetic excitations do not obey the two-magnon-interaction form of Eq. (5), but rather the empirical relation:

$$\Gamma_q \propto \Gamma_0 (1 + c T^\alpha) q^2, \quad (6)$$

where $\alpha \approx 1$. Wohlfarth [17], on the other hand, proposed that the additional excitations should be single-particle (Stoner) excitations. Ishikawa’s neutron inelastic scattering measurements on crystalline $Fe_{65}Ni_{35}$, however, showed no evidence of Stoner excitations in this system. Continentino and Rivier [18] proposed that the additional excitations are

diffusive modes that originate from longitudinal spin-wave fluctuations and give rise to an additional $T^{3/2}$ term in the low-temperature magnetization of amorphous ferromagnets. They argued that spin-canted structures may exhibit longitudinal fluctuations that can be as strong as the transverse ones and therefore their elementary excitations would contribute to an “anomalous” decrease of the magnetization. A recent report [11] that amorphous $Fe_{83}B_{17}$ exhibits a noncollinear arrangement of moments below T_c suggests that many systems that were thought to be collinear ferromagnets might be “canted” ferromagnets and that the argument of Continentino and Rivier may be valid. It is not clear, however, if this argument could be applied to Invar systems that are crystalline. This topic will be further discussed in section 5.

3. EXPERIMENTAL

The samples used in the experiments were amorphous ribbons of $Fe_{100-x}B_x$ ($x = 14, 18$) and $Fe_{90-x}Ni_xZr_{10}$ ($x = 5, 10, 20$). At these concentrations these systems exhibit Invar behavior [19-20]. The details of the sample environment and inelastic neutron scattering measurements have been described in references [3-6]. The amorphous nature of the samples under study required that the measurements of the small q excitations be made in the small-angle scattering region, in the vicinity of the incident beam [(000) direction]. The energy that can be transferred in these experiments is severely restricted to a narrow range around $E = 0$. Thus the experiments that can be performed by this technique are limited to systems of moderate or low stiffness coefficient D such that E_q at small wavevectors q falls in the “accessible” energy transfer range.

Figure 1 shows an example of inelastic scattering spectra taken at $T = 495K$ on one sample of $Fe_{86}B_{14}$ for $q = 0.08, 0.10$, and $0.12 \text{ \AA}^{-1}$. The peaks on the neutron energy gain ($E < 0$) and energy loss ($E > 0$) sides of these spectra correspond to the annihilation and creation of spin waves. Because of the restricted energy range accessible to this type of experiment it was not always possible to scan the full lineshapes. The central peaks of the inelastic scattering spectra are the result of small-angle and incoherent scattering from the sample and environment, and are elastic but q dependent.

In analyzing the spin-wave data the inelastic scattering spectra were least-squares fitted to the convolution of a parametrized form of the cross section and the triple-axis spectrometer resolution function. The solid curves shown in figure 1 are the results of these fits. The cross section was assumed to have a double Lorentzian spectral weight function (the Lorentzians corresponding to the creation and annihilation of spin waves of energy E_q and linewidth Γ_q) plus a δ function centered at $E = 0$. The central component of this cross section accounts for the small angle and incoherent scattering from the sample and environment. The parameters of the cross section that were obtained from the least-squares procedure were: (i) the spin-wave excitation energy E_q , (ii) the spin-wave linewidth Γ_q , (iii) an amplitude parameter for the inelastic peaks, proportional to the transverse susceptibility, and (iv) an overall amplitude for the central ($E = 0$) component of the cross section.

4. EXPERIMENTAL RESULTS

4.1 $Fe_{100-x}B_x$

Two samples of $Fe_{86}B_{14}$ and one of $Fe_{82}B_{18}$ were studied [3]. Due to the restricted energy transfer that can be studied in this type of experiment our measurements were performed at temperatures $T \geq 0.5T_c$ and at wavevectors $q \leq 0.13 \text{ \AA}^{-1}$. Well defined spin waves were observed at all temperatures and wavevectors studied. In all cases, the spin-wave energy E_q , obtained from the fits, could be well described by the quadratic dispersion relation of Eq. (1) with a small gap $\Delta \approx 0.04 \text{ meV}$. To illustrate this, figure 2 shows the plot of E_q vs. q^2 for one of the samples of $Fe_{86}B_{14}$. Similar plots were obtained for the other samples. The stiffness coefficients $D(T)$, obtained from the slopes of E_q vs. q^2 curves, followed a temperature dependence consistent with the two-magnon-interaction form of Eq. (4). Figure 3 shows the values of $D(T)$ for all the Fe-B samples vs. $(T/T_c)^{5/2}$. The solid lines are results of least-squares fits to the form of Eq. (4) and show that agreement to the $(T/T_c)^{5/2}$ could be obtained up to $T = 0.9T_c$. The values of $D(0)$ and the coefficient "A", obtained from these fits, are listed in table I. These values are considerably larger than the stiffness coefficient D_m , obtained from magnetization measurements and Eq. (3), which are also listed in the same table. The differences of the values of $D(0)$ (i.e., 131 and 122 $\text{meV \AA}^2$) obtained from the two samples of $Fe_{86}B_{14}$ indicate that there might be some small differences in the composition of these samples. These values should also be compared to 118 and 138 $\text{meV \AA}^2$ obtained by Ishikawa et al. [16] and Rhyne et al. [21] from previous experiments on samples of the same nominal concentration. In table I we have included also, for comparison purposes, the results on $(Fe_{65}Ni_{35})P_{16}B_6Al_3$ (non-Invar) [15].

A comparison of the temperature dependence of the spin-wave stiffness $D(T)$ of several amorphous systems (see figure 6 of reference [3]) suggested that the renormalization of $D(T)$ is more rapid in the Invar than in the non-Invar systems [i.e., the coefficient "A" of Eq. (4) is larger in Invar $Fe_{86}B_{14}$ than in non-Invar $(Fe_{65}Ni_{35})_{75}P_{16}B_6Al_3$ (table I)]. The fact that the $T^{5/2}$ dependence of Eq. (4) is found in a rather broad range of temperatures (up to $0.9T_c$!) in the Invar systems is just a consequence of the fact that in these systems the coefficient "A" of Eq. (4) is relatively large. An interpretation of the relatively large value of the coefficient "A" in these systems is that there might be additional low-lying excitations that contribute to the rapid renormalization of D with temperature. The $T^{5/2}$ dependence of D suggests that these excitations have the same form of the (energy) density of states as the spin waves.

We have also performed a detailed analysis of the wavevector and temperature dependence of the spin-wave intrinsic linewidths of Fe-B. The values of Γ_q obtained from high-resolution measurements performed on one of the specimens of $Fe_{86}B_{14}$ showed a wavevector and temperature dependence consistent with the prediction of the two-magnon-interaction theory of a Heisenberg ferromagnet [Eq.(5)], with no resemblance to the empirical form [Eq. (6)] of Ishikawa et al.[16]. In order to illustrate this we have plotted Γ_q vs. $q^4 [T \ln(k_B T/E_q)]^2$ in figure 4. In this figure the linewidth data for the other sample of $Fe_{86}B_{14}$ (asterisks), obtained from lower instrumental resolution measurements, are also shown. As it can be seen in this figure the linewidth data are in excellent agreement

with the predictions of the two-magnon-interaction theory of the Heisenberg ferromagnet of Eq. (5).

4.2 $F\epsilon_{90-x}Ni_xZr_{10}$

Specimens of $F\epsilon_{90-x}Ni_xZr_{10}$ with $x = 20, 10, 5$, were studied [4-6]. As in the Fe-B system spin-wave excitations were observed at all concentrations x and at all temperatures under study. In all cases the spin-wave energies E_q could be well described by the dispersion relation of Eq. (1). Figure 5, for example, shows the plot of E_q vs. q^2 for $F\epsilon_{85}Ni_5Zr_{10}$ [5]. The temperature dependence of the stiffness coefficients $D(T)$ could be fitted to the $T^{5/2}$ dependence of Eq. (4) up to $T = 0.9 T_c$. The values of $D(0)$ and the coefficient "A" obtained from these fits are listed in table I. As in the case of Fe-B these values of $D(0)$ are larger than the values D_m , obtained from the magnetization measurements, which are also shown in table I. The relatively low values of $D(0)$ for the samples with $x = 5, 10$, allowed the spin-wave measurements to be performed at temperatures as low as $T = 0.07 T_c$.

A detailed analysis of the linewidth data for the samples with $x = 5, 10$ revealed that the values of Γ_q could be adequately fitted to the q^4 dependence of Eq. (5) or to a q^5 predicted for topological disorder [14], but not to the q^2 dependence of Eq. (6) [See figure (6)]. The temperature dependence of these linewidths, on the other hand, was different from that expected for a conventional ferromagnet. At intermediate temperatures the linewidths were virtually temperature independent [see figure (6)], and at lower temperatures the linewidths increased considerably. In all cases the linewidths for $x = 5, 10$ were significantly broader than those of the Fe-B system [5]. It was also observed that, at the same temperatures where the spin waves broaden, an apparent enhancement of the transverse susceptibility similar to that found in RSG systems [25], occurs. There was no evidence, however, of any enhancement of the central component of the scattering (that could be associated with the development of a spin-glass order parameter) or of low-temperature spin-wave energy softening in any of the concentrations under review.

5. CONCLUSIONS AND DISCUSSION

Inelastic neutron scattering measurements have been performed on amorphous specimens of two Invar systems: Fe-B and Fe-Ni-Zr. It is expected that all the specimens under study exhibit some degree of spin-canting due to frustration [11], but none of them exhibits signs of RSG behavior. In all cases well defined spin-waves were observed, with a excitation spectra well described by a quadratic dispersion relation with a small gap. The temperature dependence of the spin-wave stiffness coefficients and the wavevector dependence of the spin-wave linewidths were consistent with the predictions of a model that is based on magnon-magnon interactions. As in other Invar systems the $T = 0$ stiffness coefficient is consistently larger than the value D_m derived from low-temperature magnetization experiments (see table I). It has been observed [3] that the renormalization of the stiffness coefficient $D(T)$ with temperature in these systems seems to be more rapid than in the non-Invar systems. This is reflected in the relatively large values of the coefficient "A" of Eq. (4), in comparison with those for the non-Invar systems. An interpretation of this observation is that there might be additional excitations that contribute to the rapid decrease of D with increasing temperature. These excitations would have the same form

of the (energy) density of states as that for spin waves since, apparently, they contribute to the renormalization of the spin-wave stiffness with temperature in the same manner as conventional spin waves. An observation that would support this interpretation is the anomalous temperature dependence of the spin-wave linewidths accompanied by an apparent enhancement of the transverse susceptibility found in $Fe_{90-x}Ni_xZr_{10}$ ($x = 5, 10$) at low temperatures.

The origin of these excitations might be a combination of the effects suggested by Continentino and Rivier [18] and Vaks et al. [26]. According to Continentino and Rivier the spin canting in a (moderately) frustated system will originate longitudinal fluctuations with amplitudes as large as the transverse ones, which would result in an additional $T^{3/2}$ temperature term in the low-temperature magnetization. Vaks et al. suggested that the coupling of the transverse and longitudinal fluctuations should result in an observed enhancement of the spin-wave cross section (*i.e.*, the longitudinal spectral weight function should consist of a pair of broad peaks at the spin-wave energies, in addition to a much smaller central peak corresponding to spin diffusion). It should be noted, however, that there is no conclusive evidence, as yet, of these propagating modes proposed by Vaks et al.

Further measurements are necessary to resolve the nature of these low-temperature features. In particular further experiments like the one by Cowley et al. [11] are necessary to ascertain the noncollinear magnetic structures of these systems and give further support to the prediction of Continentino and Rivier. If the rapid decrease of the magnetization in amorphous Invar alloys is due to longitudinal fluctuations originated by noncollinear magnetic arrangements, as proposed by Continentino and Rivier, one might ask if this mechanism could be extended to the crystalline Invar systems. This would require noncollinear magnetic structures in these crystalline Invar systems, like the one proposed by Dubinin et al. [27] for the Fe-Ni Invar. This topic also requires further study.

ACKNOWLEDGMENTS

It is a pleasure to acknowledge my collaborators J. W. Lynn, J. J. Rhyne and G. E. Fish. The work at ORNL was supported by US DOE under contract DE-AC05-84OR21400, with Martin Marietta Energy Systems, Inc.

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TABLE I
EXPERIMENTAL RESULTS

	T_c (K)	$D(0)$ meV Å ²	D_m meV Å ²	A
$Fe_{82}B_{18}$	617	165 ¹	71 ⁶	0.64
$Fe_{86}B_{14}$	556	131 ¹	65 ⁶	0.82
		122 ¹		0.79
		138 ²		
		118 ³		
$Fe_{70}Ni_{20}Ni_{10}$	455	113 ⁴	71 ⁷	0.69
			88 ⁸	
$Fe_{80}Ni_{10}Ni_{10}$	359	74 ⁵	52 ⁹	0.83
$Fe_{85}Ni_5Ni_{10}$	306	47 ⁵		0.83
$(Fe_{65}Ni_{35})_{75}P_{16}B_6Al_3$	576	114 ¹⁰	115 ¹⁰	0.45

¹ Reference [3].

² Reference [16].

³ Reference [21].

⁴ Reference [4].

⁵ Reference [6].

⁶ Reference [22].

⁷ Reference [23].

⁸ value for $Fe_{72}Ni_{18}Zr_{10}$, reference [24].

⁹ value for $Fe_{81}Ni_9Zr_{10}$, reference [24].

¹⁰ After Tarvin et al. and Birgeneau et al. [15].

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FIGURE CAPTIONS

FIGURE 1. Constant- q scans for one of the samples of $Fe_{86}B_{14}$. The solid curves are the result of the least-squares fits explained in the text. The spectral weight function used in the fits was a double Lorentzian plus a δ function centered at $E = 0$. After reference [3].

FIGURE 2. E_q vs. q^2 for one of the samples of $Fe_{86}B_{14}$. After reference [3].

FIGURE 3. $D(T)$ vs. $(T/T_c)^{5/2}$ for the two samples of $Fe_{86}B_{14}$ and $Fe_{82}B_{18}$. The solid and dotted lines are the result of least-squares fits to Eq. (4). After reference [3].

FIGURE 4. Intrinsic spin-wave linewidth Γ_q (FWHM of the Lorentzian spectral weight function) vs. $q^4 [T \ln(k_B T/E_q)]^2$ for $Fe_{86}B_{14}$. The solid curve is the result of a fit to the prediction of the two-magnon-interaction theory of a Heisenberg ferromagnet [Eq. (5)]. After reference [3].

FIGURE 5. E_q vs. q^2 for $Fe_{85}Ni_5Zr_{10}$. After reference [5].

FIGURE 6. Intrinsic spin-wave linewidth Γ_q (FWHM of the Lorentzian spectral weight function) vs. q for $Fe_{85}Ni_5Zr_{10}$. The linewidths can be fitted to a q^4 (solid curve) or a q^5 (dotted curve) dependence, with a limited temperature dependence at the temperatures shown. The dot-dash line is the result of the fit to the q^2 form of Eq. (6). After reference [5].

FIGURE 1.

FIGURE 2.

FIGURE 3.

FIGURE 4.

FIGURE 5.

FIGURE 6.