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Spin wave stiffness and the range of exchange interactions in amorphous $(\text{Fe}_{1-x}\text{Ni}_x)_{75}\text{B}_{25}$ and $(\text{Fe}_{1-y}\text{W}_y)_{83}\text{B}_{17}$ alloys

S N Kaul and T V S M Mohan Babu

School of Physics, University of Hyderabad, Central University PO, Hyderabad-500 134, India

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Abstract. The variation of saturation magnetisation, M_s , with temperature has been derived from FMR measurements performed on amorphous (a-) $(\text{Fe}_{1-x}\text{Ni}_x)_{75}\text{B}_{25}$ and $(\text{Fe}_{1-y}\text{W}_y)_{83}\text{B}_{17}$ alloys in the temperature range 77 to 300 K. These data have been analysed to yield reliable values of the coefficients B , C of the $T^{3/2}$, $T^{5/2}$ terms in the spin wave expression for the fractional decrease in magnetisation with temperature and the mean-square range of the exchange interaction, $\langle r^2 \rangle$. The spin wave theory for amorphous ferromagnets due to Krey adequately describes the observed dependence of the above parameters (B , C , $\langle r^2 \rangle$) on the Curie temperature, T_C (determined from low-field (≤ 10 Oe) bulk magnetisation data). The plot of spin wave stiffness coefficient, D , against T_C for a- $(\text{Fe}_{1-x}\text{Ni}_x)_{75}\text{B}_{25}$ alloys is a straight line, well represented by the empirical equation $D = D_0 + mT_C$, whose slope m exactly coincides with that predicted by the nearest-neighbour (nn) Heisenberg model and intercept, D_0 , on the ordinate gives the strength of the next-nearest-neighbour (nnn) exchange coupling constant which is at least one order of magnitude smaller than the nn exchange coupling constant. A plot of D against T_C for a- $(\text{Fe}_{1-y}\text{W}_y)_{83}\text{B}_{17}$ alloys is again a straight line with the same slope m but passing through the origin (i.e. $D_0 = 0$). This result is shown to imply that the competing interactions in W containing glasses confine the direct exchange to the nearest neighbours only whereas the exchange interactions in Ni containing alloys involve both the nearest as well as the next-nearest neighbours. Other important findings include (i) mixed exchange interactions cause more drastic reduction in the value of T_C , $M_s(0)$ and D than does simple magnetic dilution and (ii) for a given value of T_C , D possesses a considerably lower value for the W substituted alloys than for the Ni substituted ones.

1. Introduction

The nature of the low-lying magnetic excitations in amorphous metallic alloys has intrigued both theorists and experimentalists ever since the discovery of ferromagnetism in such materials. Ample theoretical (Herring and Kittel 1951, Kaneyoshi 1972, 1978, 1984, Continentino and Rivier 1977) and experimental (Kaul 1981a, 1983, 1984, Fernandez-Baca *et al* 1987) evidence has gathered over the years to demonstrate that the ferromagnets in the amorphous state, like their crystalline counterparts, exhibit well defined long-wavelength (wavevector $q \rightarrow 0$) spin-wave excitations which follow the dispersion relation (Keffer 1966)

$$\hbar\omega(q) = \Delta + Dq^2 + Eq^4 + \dots \quad (1)$$

where $\Delta \ll Dq^2$ is an effective energy gap which originates from the dipole-dipole

interactions (Keffer 1966, Axe *et al* 1977) and at low temperatures, both bulk magnetisation, M , and average hyperfine field, \bar{H}_{hf} , in these materials decrease with increasing temperature in accordance with the prediction of the Heisenberg model

$$[1 - (M(T)/M(0))] = [1 - (\bar{H}_{\text{hf}}(T)/\bar{H}_{\text{hf}}(0))] = BT^{3/2} + CT^{5/2} + \dots \quad (2)$$

where the coefficients B and C in equation (2) are related to the coefficients D (spin-wave stiffness constant) and E of the q^2 and q^4 terms in equation (1) through the expressions (Keffer 1966, Axe *et al* 1977, Kaneyoshi 1972, 1978)

$$B = 2.612(g\mu_B/M(0))(k_B/4\pi D)^{3/2} \quad (3)$$

and

$$C = 1.341(g\mu_B/M(0))(k_B/4\pi D)^{5/2}(3\pi\langle r^2 \rangle/4) \quad (4)$$

with the average mean-square range of exchange interaction, $\langle r^2 \rangle$, given by

$$\langle r^2 \rangle = \int r^2 J(r)g(r) dr / \int J(r)g(r) dr = -20(E/D). \quad (5)$$

$J(r)$ and $g(r)$ in equation (5) denote the range-dependent exchange interaction and the normalised pair correlation function, respectively. For a number of amorphous ferromagnetic alloys, spin-wave excitations completely account for the decrease in magnetisation (Birgeneau *et al* 1978, Kaul 1981a, Xianyu *et al* 1982, Kaul 1983, Fernandez-Baca *et al* 1987) and average hyperfine field (Chien and Hasegawa 1977, Dey *et al* 1980) with increasing temperature over a wide temperature range $0 \leq T \leq 0.5T_C$ (Curie temperature). The most significant effect of topological disorder is that the thermal demagnetisation occurs at a much *faster* rate (i.e. the value of the coefficient B in equation (2) is consistently several times *larger* and in view of equation (3) this implies that the value of the spin-wave stiffness constant, D , is considerably reduced) and the spin-wave behaviour persists to much higher temperatures ($T \leq 0.5T_C$) than in ordered (crystalline) ferromagnets ($T \leq 0.15T_C$). These observations support the theoretical viewpoint (Tahir-Kheli 1972, Kaneyoshi and Honmura 1972, Alben 1976) that the spin-wave density of states at low frequencies (low q) are greatly enhanced in the presence of disorder.

The main concern of this paper is to investigate the effect of magnetic dilution and mixed (ferromagnetic plus antiferromagnetic) exchange interactions on the magnitude of spin-wave stiffness constant and the range of exchange interactions in Fe-based amorphous ferromagnetic alloys. The alloy systems chosen for this type of study are amorphous (a-)Fe_{75-x}Ni_xB₂₅ and Fe_{83-y}W_yB₁₇ alloys. The rationale behind the choice of these systems is that Ni atoms in a-Fe_{75-x}Ni_xB₂₅ carry a negligibly small moment, which further decreases with increasing Ni concentration (Kaul 1981b), so that an increase in x results in increased magnetic dilution whereas in a-Fe_{83-y}W_yB₁₇ alloys, the exchange coupling constants between the spins on the Fe and W atomic sites are of different sign, i.e. $J_{\text{Fe-Fe}} > 0$, $J_{\text{Fe-W}} \geq 0$ (depending on the W concentration) and $J_{\text{W-W}} < 0$ (competing interactions), and W diminishes the moment on the neighbouring Fe site (Collins and Low 1965, Campbell 1966). The temperature and composition dependence of magnetisation, deduced from ferromagnetic resonance (FMR) measurements on the amorphous alloy series mentioned above, is reported and its physical implications are fully discussed.

2. Experimental details

Amorphous (a-)Fe_{75-x}Ni_xB₂₅ ($x = 35, 45, 55$) and Fe_{83-y}W_yB₁₇ ($y = 5, 9$) alloys were prepared by a single-roller melt quenching technique, under high purity argon atmosphere, in the form of long ribbons of width ~ 2 mm and thickness 30–40 μm . The amorphous nature of the ribbons so fabricated was verified by x-ray diffraction and electron microscopy. No attempt was made to find out the actual composition of the alloys and hence the nominal composition is used throughout this paper. Thermo-magnetic measurements on these alloys were carried out from room temperature to temperatures just above the Curie temperature, T_C , using a PAR vibrating sample magnetometer (model 155). The kink-point method (Kaul 1981c) was employed to determine the value of T_C from the low-field ($H \leq 10$ Oe) magnetisation data.

The first derivative with respect to the magnetic field (H) of the microwave power (P) absorbed during the ferromagnetic resonance (FMR) process, dP/dH , was measured on 4 mm long strips cut from the alloy ribbons and cleaned with ethyl alcohol before measurement, as a function of H at various fixed values of temperature in the temperature range 120–300 K in the X-band (operating frequency ≈ 9.3 GHz) on a JEOL FE-3X EPR spectrometer using sample geometries (i) horizontal-parallel (\parallel^h), in which the external magnetic field (H) is applied along the length in the ribbon plane and varied in the range zero to 5 kG and (ii) vertical-parallel (\parallel^v), in which H ($0 \leq H \leq 5$ kG) is directed along the breadth in the ribbon plane. Curves of dP/dH against H at 77 K were recorded for both the sample configurations mentioned above by immersing the sample, contained within the cavity, directly into liquid nitrogen. The sample temperature was monitored using a precalibrated copper-constantan thermocouple, placed just outside the cavity, and was held constant to within ± 0.1 K at every fixed temperature setting by means of a PID temperature controller.

Since the magnetic properties of the glassy alloys in question are known to be extremely sensitive to stress, the effects of which manifest themselves in one or more ways (Mohan Babu 1988), such as highly distorted lineshapes, large linewidths, multi-line FMR spectra, values of linewidth (ΔH) and resonance field (H_{res}) that cannot be reproduced when the measurements are repeated after remounting the sample, erratic variation of ΔH and H_{res} with temperature, a correct choice of method for sample mounting which completely avoids stress-induced effects and yields highly precise and reproducible results becomes imperative. Several methods of sample mounting were, therefore, tried out (Mohan Babu 1988) and the one that yielded the most accurate and reproducible results consisted of placing a folded piece of paper of thickness slightly greater than that of the sample on either side of the ribbon strip and sandwiching all of them between the half-cut lower portion of the quartz rod and the half-cut cylindrical quartz piece and then slowly inserting the whole assembly into the quartz tube without disturbing the sample position. The experimental results so obtained are presented and discussed in the following section.

3. Results and discussion

The experimentally observed functional dependence of the field derivative of FMR power absorption, dP/dH , on magnetic field, H , at a few selected values of temperature in the temperature range 77 to 300 K, using the horizontal-parallel sample configuration,

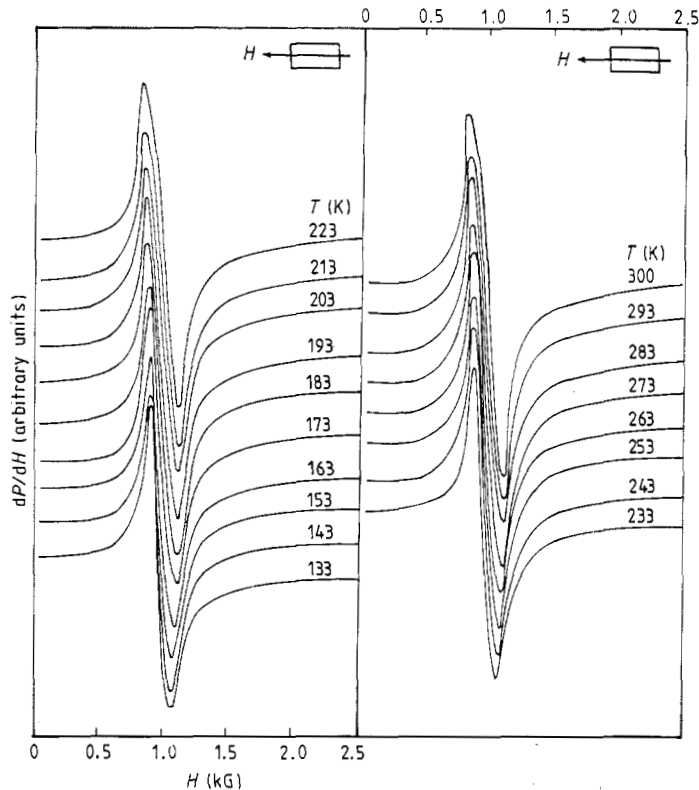


Figure 1. FMR power absorption derivative (PAD) curves for $\alpha\text{-Fe}_{30}\text{Ni}_{45}\text{B}_{25}$ taken at different temperatures in the horizontal-parallel configuration. To accommodate more curves, the saturated portions of the PAD curves for $H > 2500$ Oe are not shown in this figure.

typical of $\alpha\text{-Fe}_{75-x}\text{Ni}_x\text{B}_{25}$ and $\alpha\text{-Fe}_{83-y}\text{W}_y\text{B}_{17}$ alloys is shown in figures 1 and 2. Temperature dependence of the resonance field, H_{res} (defined as the field where dP/dH possesses half the peak-to-peak value) and 'peak-to-peak' linewidth, ΔH_{pp} , deduced from the curves of dP/dH versus H , is depicted in figures 3 and 4. Now that identical results, within the error limits, are obtained for the horizontal- and vertical-parallel sample geometries, neither raw (i.e. dP/dH against H curves at different temperatures) nor derived (i.e. $H_{\text{res}}(T)$ and $\Delta H_{\text{pp}}(T)$) data for the latter geometry are presented here. In view of the expressions (Conger and Essig 1956)

$$H_{\text{res}}^{\text{h}} = H_{\text{res}}^{\parallel} - H_{\text{k}} \quad (6a)$$

and

$$H_{\text{res}}^{\text{v}} = H_{\text{res}}^{\parallel} + H_{\text{k}} \quad (6b)$$

where $H_{\text{res}}^{\text{h}}$ and $H_{\text{res}}^{\text{v}}$ are resonance fields observed in the horizontal- and vertical-parallel configurations, respectively, and $H_{\text{res}}^{\parallel}$ is the resonance field in the absence of the 'in-plane' uniaxial anisotropy field, H_{k} , the above finding that $H_{\text{res}}^{\text{h}}(T) \approx H_{\text{res}}^{\text{v}}(T)$ indicates that the value of H_{k} at any temperature within the investigated temperature range does not exceed 50 G. Figures 3 and 4 reveal that H_{res} increases with increasing temperature and the variation with T becomes steeper as T is progressively increased towards room

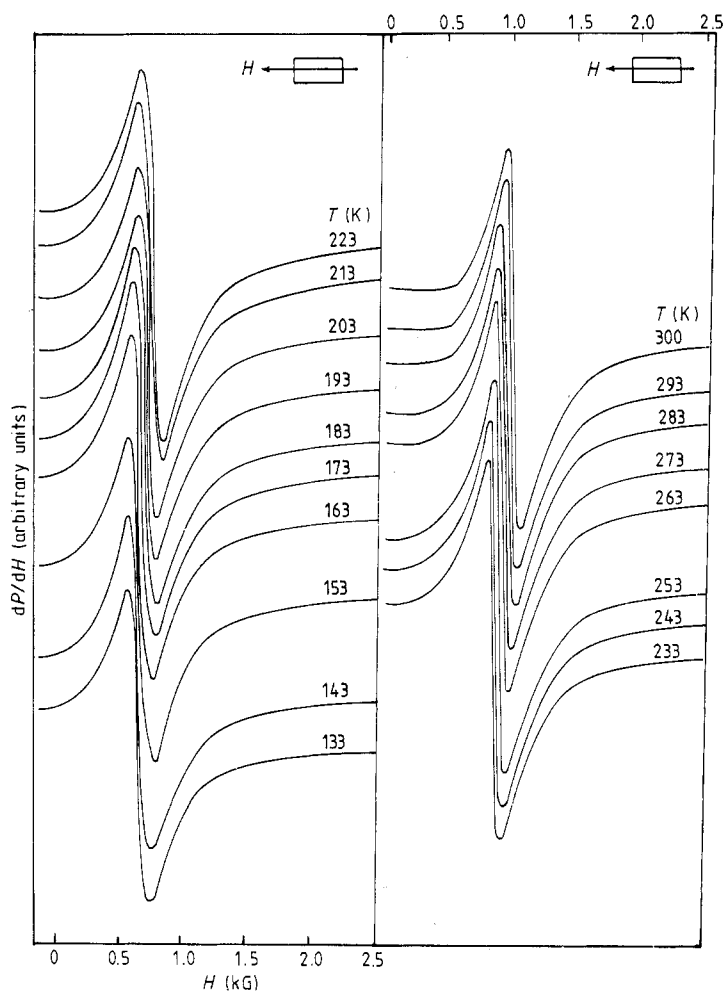


Figure 2. FMR power absorption derivative curves for a-Fe₇₈W₅B₁₇ taken at different temperatures in the horizontal-parallel configuration for $H \leq 2500$ Oe.

temperature (≈ 300 K) while ΔH_{pp} remains practically constant (within the error limits) in the entire temperature range covered in the present experiments.

With a view to ascertain whether the finite FMR linewidth has any influence on the observed values of H_{res} , a detailed lineshape analysis (Kaul and Siruguri 1987) of the FMR spectra taken at different temperatures for all the investigated amorphous alloys has been carried out with the result that the splitting factor, g , possesses a temperature-independent value of 2.09 ± 0.02 , the saturation magnetisation, M_s , has a temperature dependence as shown in figures 5 and 6, and the observed linewidths do not cause any noticeable shift in the resonance field (primarily because ΔH_{pp} is small (≤ 200 Oe) while $4\pi M_s$ is larger (≥ 3 kG) (Webb and Bhagat 1984, Kaul and Siruguri 1987)). In figures 5 and 6, the straight lines through the data points represent the best least-squares theoretical fits based on equation (2) with the choice (that yields the least value for the sum of the squares of the deviations, χ^2) of the coefficients B and C (coefficient B) and $M_s(0)$ given in table 1 when both $T^{3/2}$ and $T^{5/2}$ terms (the $T^{3/2}$ term only) in equation (2) are

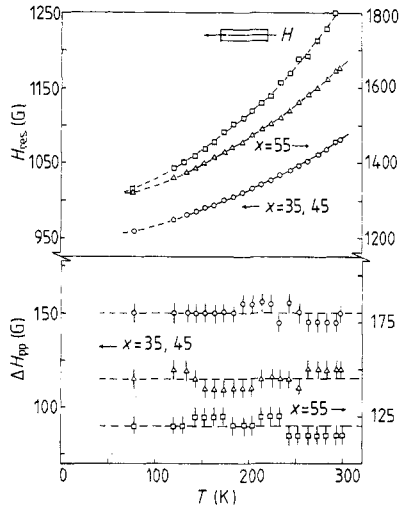


Figure 3. Resonance field, H_{res} , and peak-to-peak linewidth, ΔH_{pp} , as functions of temperature for amorphous $\text{Fe}_{75-x}\text{Ni}_x\text{B}_{25}$ alloys in the horizontal-parallel configuration. \circ , $x = 35$; \triangle , $x = 45$; \square , $x = 55$.

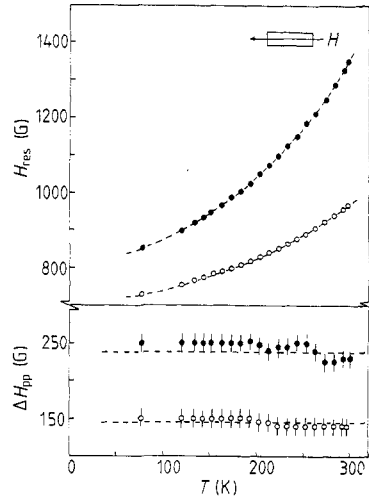


Figure 4. Resonance field, H_{res} , and peak-to-peak linewidth, ΔH_{pp} , as functions of temperature for amorphous $\text{Fe}_{83-y}\text{W}_y\text{B}_{17}$ alloys in the horizontal-parallel configuration. \circ , $y = 5$; \bullet , $y = 9$.

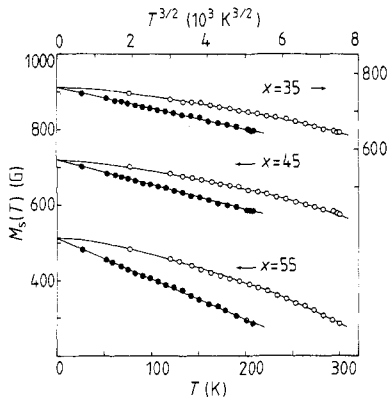


Figure 5. Temperature dependence of magnetisation and plots of M_s against $T^{3/2}$ for amorphous $\text{Fe}_{75-x}\text{Ni}_x\text{B}_{25}$ alloys. Full curves through the data points on the M_s against T plots are the least-squares fits to the data based on equation (2) of the text with the choice of the coefficients B and C given in table 1. \circ , M_s against T ; \bullet , M_s against $T^{3/2}$.

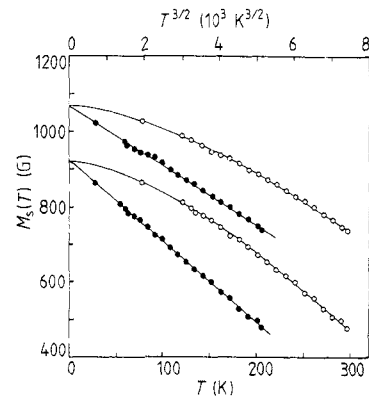


Figure 6. Experimentally observed (open circles) and theoretically predicted (full curves through the data points) variation of magnetisation with temperature in amorphous $\text{Fe}_{83-y}\text{W}_y\text{B}_{17}$ alloys based on equation (2) (in the text). This figure also shows M_s plotted against $T^{3/2}$ (\bullet) for different compositions in this alloy series.

(is) considered. Inclusion of the $T^{5/2}$ term leads to a significant improvement in the quality of the theoretical fits as inferred from the appreciable reduction in the value of χ^2 . The values of the parameters B , C and $M_s(0)$ so determined and $g = 2.09 \pm 0.02$ when used in equations (3) and (4) give the corresponding values of the spin wave stiffness constant, D , and the average mean-square range of the exchange interaction,

Table 1. Magnetic and spin wave parameters, mean-square range of the exchange interaction and Gilbert damping parameter for amorphous $Fe_{78-x}Ni_{10}B_{12}$ and $Fe_{85-y}W_{10}B_5$ alloys. Numbers in parentheses denote the estimated uncertainty in the least significant figure.

Sample/ alloy	Ref.†	Method‡	$M_s(0)$ (G)	T_c (K)	B		D (meV Å ³)	D/T (meV Å ² K ⁻¹)	$\langle r^2 \rangle$ (Å ²)	λ (10 ⁸ s ⁻¹)	
					(10 ⁶ K ^{3/2}) [$B_{1/2}$]	(10 ⁸ K ^{3/2}) [$C_{3/2}$]				77 K	300 K
x = 35	PW	FMR, KPM	765(10)	637.0(10)	28.8(22)		120(5)	0.19(1)		1.36(9)	1.17(8)
	a	BM	889	677.0(10)	[0.46(4)] 19.1 [0.34]		142	0.21			
x = 45	PW	FMR, KPM	770(10)	637.0(10)	28.4(21)		120(5)	0.19(1)	2.3(17)		
	PW	FMR, KPM	720(5)	539.0(10)	[0.46(3)] 36.2(23) [0.45(3)]	0.45(25) [0.05(2)]	107(5)	0.20(1)		0.97(8)	0.80(7)
x = 55	a	BM	708	568.0	34.5(1) [0.47]		111(1)	0.20			
	PW	FMR, KPM	720(5)	539.0(10)	36.5(20)	0.70(20)	106(5)	0.20(1)	2.5(14)		
x = 55	PW	FMR, KPM	515(5)	373.0(10)	[0.46(2)] 83.4(26) [0.60(2)]	[0.05(2)]	78(4)	0.21(1)		0.70(6)	0.42(3)
	a	BM	528	395.0	72.5 [0.57]		83	0.21			
y = 5	PW	FMR, KPM	515(5)	373.0(10)	78.2(18)	2.10(50)	80(3)	0.21(1)	2.6(8)		
	b	BM, AA	372.9(2)		[0.56(2)]						
y = 5	PW	FMR, KPM	1066(4)	450.0(10)	59.0(40)		59(3)	0.13(1)		1.79(25)	1.28(20)
	PW	FMR, KPM	1070(4)		[0.56(4)] 54.8(42) [0.52(4)]	1.50(50) [0.06(3)]	62(3)	0.14(1)	2.0(11)		
y = 9	b	BM, AA	449.2(2)		93.3(27)		48(2)	0.14(1)		2.48(26)	1.36(14)
	PW	FMR, KPM	922(4)	354.0(10)	[0.62(2)]		49(2)	0.14(1)	2.0(4)		
Fe	PW	FMR, KPM	923(4)		91.7(23) [0.61(2)]	3.00(50) [0.07(1)]	286(11)	0.27(1)	10.1(123)		
	b	BM, AA	353.6(2)		3.4(2)	0.10(10)					
	c	BM	1752	1043	[0.115(7)]	[0.04(4)]				0.70	0.70
	d	FMR									

† PW, present work; †† Chattopadhyay (1982); †† Kellner *et al.* (1986); †† Kaul (1983); †† Bhagat (1973).

‡ AA, asymptotic analysis; BM, bulk magnetisation; FMR, ferromagnetic resonance; KPM, kink-point method (for details see Kaul 1981c).

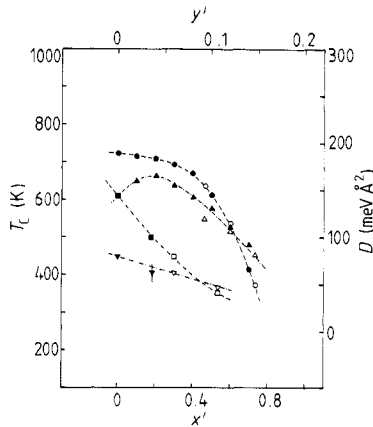


Figure 7. Composition dependence of the spin wave stiffness coefficient, D , and Curie temperature, T_C , in amorphous $(\text{Fe}_{1-x}\text{Ni}_x)_{75}\text{B}_{25}$ and $(\text{Fe}_{1-y}\text{W}_y)_{83}\text{B}_{17}$ alloys. For $(\text{Fe}_{1-x}\text{Ni}_x)_{75}\text{B}_{25}$: \circ , \bullet , T_C ; \triangle , \blacktriangle , D . For $(\text{Fe}_{1-y}\text{W}_y)_{83}\text{B}_{17}$: \square , \blacksquare , T_C ; ∇ , \blacktriangledown , D . Open symbols represent results from present work; closed symbols: \bullet , Vincze *et al* (1980); \blacktriangle , Chattopadhyay (1982); \blacksquare , \blacktriangledown Potocky *et al* (1984).

$\langle r^2 \rangle$. The values of D and $\langle r^2 \rangle$ and those of T_C determined by the kink-point method (Kaul 1981c) along with reported values of the parameters $M_s(0)$, B , D , T_C and D/T_C for the same compositions (Chattopadhyay 1982, Kellner *et al* 1986) are also listed in this table.

Composition dependence of the spin wave stiffness constant, D , and Curie temperature, T_C , in amorphous $(\text{Fe}_{1-x}\text{Ni}_x)_{75}\text{B}_{25}$ and $(\text{Fe}_{1-y}\text{W}_y)_{83}\text{B}_{17}$ alloys is depicted in figure 7. Similar data derived from bulk magnetisation measurements on other compositions in the same alloy series as above (Vincze *et al* 1980, Chattopadhyay 1982, Potocky *et al* 1984) are also included in this figure to clearly illustrate the functional dependence of these quantities on composition over a wider composition range. The following observations can be made based on data presented in figures 5–7 and table 1: (i) the values determined here for the parameters B , CD , T_C and D/T_C are in reasonable agreement with the corresponding values reported earlier; (ii) a much steeper drop in the values of both T_C and $M_s(0)$ is caused by replacing Fe with W than with Ni, i.e. $(\Delta T_C/\Delta y') \approx 30(\Delta T_C/\Delta x')$ and $(\Delta M_s(0)/\Delta y') \approx 10(\Delta M_s(0)/\Delta x')$ in the concentration range $0 \leq x', y' \leq 0.1$; (iii) within the composition range $0.5 \leq x' \leq 0.733$ $(\Delta T_C/\Delta x') \approx 12.5 \text{ K/at. \%Ni}$ and $(\Delta D/\Delta x') \approx 2.5 \text{ meV \AA}^2/\text{at. \%Ni}$ so that $(\Delta D/\Delta T_C) \approx 0.20 \text{ meV \AA}^2 \text{ K}^{-1}$ whereas in the concentration range $0.036 \leq y' \leq 0.108$ ($0 \leq y' \leq 0.108$), $(\Delta T_C/\Delta y') \approx 20.8(23.6) \text{ K/at. \%W}$ and $(\Delta D/\Delta y') \approx 2.9(2.8) \text{ meV \AA}^2/\text{at. \%W}$ such that $(\Delta D/\Delta T_C) \approx 0.14(0.12) \text{ meV \AA}^2 \text{ K}^{-1}$; (iv) with decreasing T_C , (a) the temperature range over which Bloch's $T^{3/2}$ law dominates becomes wider and wider, (b) the average mean-square range of the exchange interaction, $\langle r^2 \rangle$ and D/T_C (which is also a measure of the range of the exchange interaction) both exhibit a slight increase (more so for the alloys containing Ni) and (c) the normalised coefficient $C_{5/2} = CT_C^{5/2}$ apparently retains its crystalline Fe or Ni value in contrast with the reduced coefficient $B_{3/2} = BT_C^{3/2}$ whose magnitude is greatly enhanced and which for the present alloys possesses values that are roughly 4 to 5 times larger than those found in crystalline ferromagnets.

Observation (ii) above lends itself to a straightforward explanation as follows. In the molecular-field approximation, T_C and the exchange coupling constant between the nearest neighbours, J , are directly related as

$$T_C = [2S(S+1)/3k_B]zJ \quad (7)$$

where z is the number of nearest neighbours. The average value of J in the glassy alloys of present interest is given by

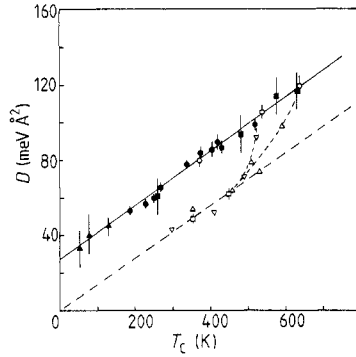


Figure 8. Spin wave stiffness coefficient, D , as a function of Curie temperature, T_C , for amorphous $(\text{Fe}_{1-x}\text{Ni}_x)_{75}\text{B}_{25}$ and $(\text{Fe}_{1-x}\text{W}_x)_{83}\text{B}_{17}$ alloys. $D(T_C)$ data available in the literature on other similar glassy alloy systems are also included in this figure (see the text). \circ , $(\text{Fe}_{1-x}\text{Ni}_x)_{75}\text{B}_{25}$ (present work); \bullet , $(\text{Fe}_{1-x}\text{Ni}_x)_{80}(\text{P}, \text{B}, \text{Si})_{20}$; \blacktriangle , $(\text{Fe}_{1-x}\text{Ni}_x)_{77}\text{B}_{13}\text{Si}_{10}$; \blacksquare , $(\text{Fe}_{1-x}\text{Ni}_x)_{75}\text{P}_{16}\text{B}_6\text{Al}_3$; \square , $(\text{Fe}_{1-x}\text{W}_x)_{83}\text{B}_{17}$ (present work); ∇ , $(\text{Fe}_{1-x}\text{Cr}_x)_{85-y}\text{B}_{15+y}$; \triangle , $(\text{Fe}_{1-x}\text{Mn}_x)_{78}\text{B}_{22}$.

$$\langle J \rangle = (1-x)^2 J_{\text{Fe-Fe}} + 2x(1-x) J_{\text{Fe-imp}} + x^2 J_{\text{imp-imp}} \quad (8)$$

where x and the subscript 'imp' stand for x' or y' and Ni or W, respectively. In $a\text{-(Fe}_{1-x}\text{Ni}_x)_{75}\text{B}_{25}$ alloys, $J_{\text{Fe-Ni}}$ and $J_{\text{Ni-Ni}}$ are both positive but small and their magnitude decreases further when the Ni concentration is increased (Kaul 1981b) whereas a progressive replacement of Fe with Ni reduces the contribution due to the first term in equation (8). Thus, $\langle J \rangle$ (and hence T_C) decreases as x' increases. An interpretation of the $T_C(y')$ data in $a\text{-(Fe}_{1-y'}\text{W}_{y'})_{83}\text{B}_{17}$ alloys along similar lines requires a complete knowledge of the relative magnitudes and signs of various exchange interactions in these alloys as a function of y' which is lacking at present. However, in view of the general observation that the short-range order in amorphous alloys is roughly the same, though not so well defined as that in their crystalline counterparts, a tentative explanation could nevertheless be attempted by making use of the relevant results available (Campbell 1966) on crystalline $\text{Fe}_{100-x}\text{W}_x$ alloys and by assuming that the topological disorder affects the strength of $J_{\text{Fe-Fe}}$, $J_{\text{Fe-W}}$ and $J_{\text{W-W}}$ to some extent but not their sign. Therefore, in $a\text{-(Fe}_{1-y'}\text{W}_{y'})_{83}\text{B}_{17}$ alloys with $y' = 0.036$ and 0.108 , both $J_{\text{Fe-W}}$ and $J_{\text{W-W}}$ are negative (antiferromagnetic coupling) and comparable in magnitude to $J_{\text{Fe-Fe}}$ (Campbell 1966) while the ferromagnetic coupling between Fe atoms, $J_{\text{Fe-Fe}}$, is reduced in strength as the W concentration increases because the average distance between such Fe atoms is considerably increased on account of the greater atomic diameter of W (by about 10%) compared with that of Fe. Consequently, a sharper drop in $\langle J \rangle$ (and hence in T_C , cf equation (7)) is expected with y' rather than with x' . The effect of adding Ni at the expense of Fe in $a\text{-(Fe}_{1-x'}\text{Ni}_{x'})_{75}\text{B}_{25}$ alloys is to dilute the system magnetically, because Ni atoms in these alloys carry a negligibly small moment (Kaul 1981b), and to make the charge transfer from B atoms to the holes in the 3D-subbands of Fe more effective. In contrast, besides the simple magnetic dilution and charge-transfer effects, the presence of W in the first or second coordination shell of Fe diminishes the moment at the Fe site (Campbell 1966) so that $M_s(0)$ should decrease more rapidly in the case of alloys containing W than those containing Ni. Observation (iii) points to the existence of a linear relation between D and T_C . Figure 8 testifies to the validity of this prediction by demonstrating that when D is plotted against T_C , the D values for the amorphous alloys with $x' = 0.47, 0.60$ and 0.733 in the alloy series $(\text{Fe}_{1-x}\text{Ni}_x)_{75}\text{B}_{25}$, like those in other amorphous (Fe, Ni)-M alloys (where M = P, B, Si, Al), fall on a straight line represented by the equation

$$D = D_0 + mT_C \quad (9)$$

with the intercept $D_0 = 27 \pm 2 \text{ meV } \text{Å}^2$ and slope $m = 0.144 \pm 0.020 \text{ meV } \text{Å}^2 \text{K}^{-1}$ (Kaul

1983) whereas the values of D for the compositions $y' = 0.060$ and 0.108 in the glassy alloy series $(\text{Fe}_{1-y'}\text{W}_{y'})_{83}\text{B}_{17}$ fall on another straight line parallel (i.e. $m = 0.140 \text{ meV } \text{\AA}^2 \text{ K}^{-1}$) to the earlier straight line and passes through the origin (i.e. $D_0 = 0$). Previously reported values of D for $a\text{-(Fe}_{1-x}\text{Cr}_x)_{85-y}\text{B}_{15+y}$ ($y = 0, 5, 10$) and $a\text{-(Fe}_{1-x}\text{Mn}_x)_{78}\text{B}_{22}$ alloys (Dey *et al* 1980, Soumura *et al* 1986) (except for the alloys with $x < x_C \approx 0.065$), when plotted in the form of D against T_C , are noticed to fall on the latter straight line (figure 8). The theoretical basis for a linear variation of D with T_C of the form given by equation (9) when D_0 is either finite or zero has already been provided by Kaul (1983) in terms of the Heisenberg model which considers the nearest-neighbour (nn) and/or the next-nearest-neighbour (nnn) spin pairs to be coupled via an exchange interaction of the Heisenberg type with the corresponding exchange coupling constants denoted by J_1 and J_2 , respectively, and yields the following expressions (Kaul 1983) for D_0 and m in the two cases

$$D_0 = z_2 J_2 (a_2^2 - a_1^2) S / 3 \quad m = k_B a_1^2 / 2 (S + 1) \quad (10a)$$

$$D_0 = 0 \text{ (since } J_2 = 0) \quad m = D / T_C = k_B a_1^2 / 2 (S + 1) \quad (10b)$$

where a_1 and a_2 are the nn and nnn distances and z_2 is the number of the next-nearest neighbours. Using $S = 1$, $D_0 = 27 \text{ meV } \text{\AA}^2$ (the observed value) and the typical values $a_1 = 2.55 \text{ \AA}$, $a_2 = 4.35 \text{ \AA}$ and $z_2 \approx 6$ in equations (10a) and (10b), the values of J_2 and m come out to be $J_2 \approx 1 \text{ meV}$ and $m = 0.140 \text{ meV } \text{\AA}^2 \text{ K}^{-1}$ (Kaul 1983). While the value of m , so computed, is the same (within error limits) as the observed gradients of both the straight lines, J_2 is at least one order of magnitude smaller than J_1 (Kaul 1983), which normally ranges between 10 and 50 meV for amorphous ferromagnetic alloys of the type considered in this work (Kaul 1981b). The above finding that the $D(T_C)$ data for $a\text{-(Fe, Ni)-M}$ and $a\text{-(Fe, M')-B}$ alloys ($M' = \text{Mn, Cr, W}$) fall on two (roughly) parallel 'universal' straight lines, which result when the values of D_0 and m given by equations (10a) and (10b) are used in equation (9), implies that the direct exchange interactions in the latter alloy series are confined to the nearest neighbours only whereas in the former set of alloys they involve not only the nearest neighbours but also the next-nearest neighbours. This inference finds further support from the fact that the D/T_C values for $a\text{-(Fe}_{1-x}\text{Ni}_x)_{75}\text{B}_{25}$ alloys are consistently higher than those calculated from equation (10b) while those for $a\text{-(Fe}_{1-y'}\text{W}_{y'})_{83}\text{B}_{17}$ alloys exactly match the calculated value of $D/T_C = 0.14 \text{ meV } \text{\AA}^2 \text{ K}^{-1}$ (see table 1). For impurity (M') concentrations $x < x_C$ (x_C is the solute concentration below which D values start deviating from the straight line corresponding to $D_0 = 0$ and $D/T_C = m$, equation (10b)), the range of exchange interactions extends beyond the nearest-neighbour distance (i.e. a 'crossover' from short-ranged to long-ranged exchange takes place). The above arguments permit us to conclude that the competing interactions tend to limit the range of direct Heisenberg ferromagnetic exchange interactions and confine them to the nearest neighbours only. Another important point that deserves attention is that for a given value of T_C D has a considerably lower value in the case of Mn, Cr and W substituted (type II) alloys than in the Ni substituted (type I) ones (figure 8). Although this result could be interpreted as higher spin-wave density of states at low frequencies (Tahir-Kheli 1972, Kaneyoshi and Honmura 1972, Alben 1976) in type II alloys, a more lucid explanation, given below, can be provided in terms of the expression derived for the spin wave stiffness constant in the quasicrystalline approximation (Kaul 1981a), i.e.

$$D = D(0)[1 - 2z(\Delta r/r_0)(\Delta J/J(r_0))] \quad (11a)$$

with

$$D(0) = zSJ(r_0)r_0^2/3 \quad (11b)$$

where r_0 is the mean nn distance, $(\Delta r)^2$ is the mean-square deviation of the nn pair distances and ΔJ is the contribution to the exchange integral resulting from the distribution in the nn distance due to structural fluctuations in amorphous materials. In the case of Ni-substituted alloys, no appreciable change in $(\Delta r/r_0)$ from its value for the parent alloy, a-Fe₇₅B₂₅, is expected to occur since Ni and Fe atoms have roughly the same atomic diameter, whereas ΔJ should undergo a marked change because of alteration in the environment (both the number and type of the nearest neighbours) of a given Fe atom. Thus, the quantity $\Delta J/J(r_0)$ is mainly responsible for the reduction in the value of D as x' increases in a-(Fe_{1-x'}Ni_{x'})₇₅B₂₅ alloys. By comparison, the atomic diameters of Fe and W are widely different so that both Δr and r_0 should possess larger values when W replaces Fe than when only Fe is present, i.e. for a-Fe₈₃B₁₇. Moreover, exchange fluctuations, ΔJ , shall have greater amplitude in a-(Fe_{1-y'}W_{y'})₈₃B₁₇ alloys as a result of the random mixture of ferromagnetic and antiferromagnetic interactions than in a-(Fe_{1-x'}Ni_{x'})₇₅B₂₅ alloys in which only the ferromagnetic interactions exist. From the foregoing remarks, it is evident that for any specified value of T_C (which, according to equation (7), constrains $J(r_0)$ to have the same value for both the glassy alloy systems), D possesses a considerably smaller value for the type II alloys than for the type I alloys because in the former case the quantities $(\Delta r/r_0)$ and $(\Delta J/J(r_0))$ both change substantially from their values for a-Fe₈₃B₁₇ in contrast with the latter case wherein only $\Delta J/J(r_0)$ changes and that, too, to a lesser extent. Observations (iv), (a)–(c), are not only in consonance with similar observations made earlier on other amorphous ferromagnetic alloys (Kaul 1981a, 1983) but also conform very well with the predictions of the Heisenberg model (Krey 1978), which assumes the functional dependence of the exchange integral, J , on distance, r , to be of the form $J(r) = J_1 \exp\{\alpha[(r/a_1) - 1]\}$ with $J(r) = 0$ for $r \geq 1.25 a_1$ and considers α to be negative. Negative sign of α is also consistent with our finding that $J_2 \ll J_1$. Furthermore, the presently determined values of the normalised coefficient $B_{3/2}$ for the amorphous alloys in question (table 1) are noticed to agree well with the theoretical values $B_{3/2} = 0.512$ for $S = 1$ and $B_{3/2} = 0.587$ for $S = \frac{1}{2}$ (Krey 1978).

Finally, we return to the discussion of the $\Delta H_{pp}(T)$ data displayed in figures 3 and 4. ΔH_{pp} is found to be independent of temperature within the investigated temperature range (77–300 K). Insensitivity of ΔH_{pp} to changes in temperature over a wide temperature range which lies well below T_C is known to be a characteristic property (Bhagat *et al* 1977, Bhagat *et al* 1980, Bhagat *et al* 1985, Spano and Bhagat 1981, Webb and Bhagat 1984) of amorphous ferromagnetic alloys with composition well above the percolation threshold (i.e. the composition at which long-range ferromagnetic order first sets in). Two main contributions to ΔH_{pp} are ΔH_1 , which does not depend on the microwave field frequency, $\nu = \omega/2\pi$, and is caused by two-magnon scattering from spatially localised magnetic inhomogeneities (Heinrich *et al* 1984), and ΔH_{LLG} ($= 1.45\lambda\omega/\gamma^2 M_s$, where λ is the Gilbert damping parameter and $\gamma = -g|e|/2mc$ is the gyromagnetic ratio), which has a linear dependence on ν and results from the Landau–Lifshitz–Gilbert (LLG) relaxation mechanism. Additional contribution to ΔH_{pp} , besides ΔH_{LLG} and ΔH_1 , arises from the skin-depth effect (which makes the magnetisation induced by the microwave field non-uniform in the volume of the surface penetration layer) but this contribution for the investigated alloys turns out to be as small as ~ 10 Oe (Kaul and Srinivasa Kasyapa 1989). Note that this value lies well within the observed error limits and hence can be dropped out of the discussion. Unambiguous

determination of the FMR linewidth contributions ΔH_I and ΔH_{LLG} calls for investigations which involve measurement of ΔH_{pp} as a function of ν at various fixed values of temperature since a plot of ΔH_{pp} against ν is a straight line whose intercept on the ordinate yields ΔH_I and the slope gives ΔH_{LLG} . Such frequency dependent FMR data (Spano and Bhagat 1981, Cochran *et al* 1982, Heinrich *et al* 1984, Webb and Bhagat 1984 and Bhagat *et al* 1985) reported on a number of widely different amorphous ferromagnetic alloy systems and commercial metallic glasses in the temperature range ($T < T_C$) where ΔH_{pp} is found to remain constant reveal that $\lambda \propto M_s$ and $\Delta H_{pp} (\nu = 9.3 \text{ GHz}) = \Delta H_I + \Delta H_{LLG} (\nu = 9.3 \text{ GHz}) \approx 0.7 \Delta H_{pp} (\nu = 9.3 \text{ GHz}) + 0.3 \Delta H_{pp} (\nu = 9.3 \text{ GHz})$. An immediate consequence of the result $\lambda \propto M_s$ is that ΔH_{LLG} does not vary with temperature so that in view of a constant value of ΔH_{pp} even ΔH_I should not depend on temperature. This deduction is consistent with our present finding that thermal cycling between 77 and 300 K has no noticeable influence on the value of ΔH_{pp} and with our previous observation (Kaul and Srinivasa Kasyapa 1989) on similar glassy alloys as those investigated in this work that the annealing treatment at moderately high temperatures ($T \approx 500 \text{ K}$) does not have any detectable effect on the bulk values of λ and M_s . Values of the damping parameter, $\lambda = (\gamma^2 M_s / 1.45\omega) \Delta H_{LLG}$, at the extreme temperatures of the temperature range investigated (i.e. 77 and 300 K) deduced using the above result, i.e. $\Delta H_{LLG} (\nu = 9.3 \text{ GHz}) \approx 0.3 \Delta H_{pp} (\nu = 9.3 \text{ GHz})$, and the presently determined values of g and $M_s(T)$ are listed in table 1. The values of λ so calculated are found to be in excellent agreement with those reported previously (Bhagat *et al* 1977, Bhagat *et al* 1980, Spano and Bhagat 1981, Cochran *et al* 1982, Heinrich *et al* 1984) for amorphous alloys with similar 3D transition metal content.

4. Summary and conclusions

The first set of detailed FMR measurements presently performed on amorphous $(\text{Fe}_{1-x}\text{Ni}_x)_{75}\text{B}_{25}$ and $(\text{Fe}_{1-y}\text{W}_y)_{83}\text{B}_{17}$ alloys go beyond the previously reported bulk magnetisation measurements on similar compositions in the glassy alloy series in question on two main counts. First, all the parameters $M_s(0)$, T_C , $\langle r^2 \rangle$ and D have been determined from FMR and low-field magnetisation data taken on the same sample for a given alloy composition rather than from measurements on alloy samples from completely different batches (figure 7 and Potocky *et al* 1984). Second, the effect of magnetic dilution and mixed exchange interactions on magnetic and spin wave parameters is clearly illustrated, and D and T_C values determined in this work, along with those available in the literature on other amorphous ferromagnetic alloys when plotted in the form of D against T_C (figure 8) are shown to fall on two different 'universal' straight lines for glassy magnetic systems with or without competing exchange interactions. Moreover, based on the present FMR results, the following conclusions can be drawn.

(i) Mixed exchange interactions are more effective in reducing the value of Curie temperature, T_C , saturation magnetisation at 0 K, $M_s(0)$ and spin wave stiffness constant, D , than exchange interactions resulting in simple magnetic dilution.

(ii) Direct exchange interactions in $a\text{-(Fe}_{1-y}\text{W}_y)_{83}\text{B}_{17}$ alloys with $y' = 0.060$ and 0.108 are confined to the nearest neighbours only whereas in $a\text{-(Fe}_{1-x}\text{Ni}_x)_{75}\text{B}_{25}$ alloys they not only couple spins on the nearest-neighbour (nn) sites but also those on the next-nearest-neighbour (nnn) sites. For impurity ($M' = \text{Mn, Cr, W}$) concentrations $x < x_C$ ($x_C \approx 0.06$) a 'crossover' from short-ranged ($(\langle r^2 \rangle)^{1/2} \approx a_1$, nn distance) to long-ranged ($a_1 < (\langle r^2 \rangle)^{1/2} \approx a_2$, nnn distance) exchange occurs.

(iii) For a given value of T_C , the spin wave stiffness constant has a substantially lower value for the W substituted alloys than for the Ni substituted ones primarily because both structural and exchange fluctuations have much greater amplitude in the former alloy series than in the latter.

(iv) In conformity with the predictions of the Heisenberg model, which assumes the exchange integral to have an exponential dependence on distance, r , for $r \leq 1.25a_1$ and a sharp cut-off beyond this distance, it is observed that as T_C decreases, (a) the spin-wave behaviour persists to higher temperatures, (b) the average mean-square range of the exchange interaction exhibits a slight increase and (c) the normalised coefficient $C_{5/2}$ retains its crystalline value while the reduced coefficient $B_{3/2}$ possesses considerably higher values for ferromagnets in the amorphous state than those in the crystalline state and increases in magnitude.

(v) The Gilbert damping parameter, λ , for the amorphous alloys investigated has values that are of the same order of magnitude as those found for crystalline Permalloys.

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