SYSTEMATIC STUDY OF THE TEMPERATURE DEPENDENCE OF THE SATURATION MAGNETIZATION IN Fe, Fe-Ni AND Co-BASED AMORPHOUS ALLOYS

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Abstract. We have measured magnetization versus temperature in the temperature interval 2-200 K, for amorphous alloys of the three different compositions: $Fe_{81.5}B_{14.5}Si_4$, $Fe_{40}Ni_{38}Mo_4B_{18}$ and $Co_{70}Fe_5Ni_2Mo_3B_5Si_{15}$. The measurements were performed by means of a SQUID magnetometer. A powerful data analysis technique based on successive minimization procedures, has allowed us to conclude that Stoner excitations of the strong ferromagnetic type play a significant role in the Fe-Ni alloy studied. The Fe rich and Co rich alloys do not show a measurable contribution from single particle excitations.

Introduction

The knowledge of the fundamental properties of amorphous ferromagnets is of great importance from the standpoint of applications, i.e. in an Fe(BC) alloy, the replacement of B by C increases the room temperature m considerably through the increase of both the spin wave stiffness constant and the density {1}.

In particular, the saturation magnetization change with temperature m(T), has been the object of a great deal of experimental work. There are many reports about the different mechanisms which contribute to the decrease of the saturation magnetization with temperature in amorphous alloys, but a complete picture has not yet been achieved and there is still controversy over the role of the different terms of the spin wave expansion and the weak and strong itinerant models.

The presence of long wavelength spin waves in amorphous alloys, indeniably proved by inelastic neutron scattering measurements, is also assessed by the thermal demagnetization behaviour, which has been repeatedly reported to follow Bloch's law:

$$m(T) = m(0)\left(1 - BF\left(\frac{3}{2},\frac{T}{T_g}\right)T^{3/2} - CF\left(\frac{5}{2},\frac{T}{T_g}\right)T^{5/2}\right) \qquad (1)$$

where m(0) is the saturation magnetization at T=0K, and the functions $F(\frac{3}{2},\frac{T}{T_g})$ and $F(\frac{5}{2},\frac{T}{T_g})$, given in ref {2} contain the magnetic field dependence of the spin wave spectrum, the gap temperature T_g being related to the gap introduced in this spectrum by the effective magnetic field (H_{eff}) being $T_g=(g\mu_B H_{eff})/k$, where g is the gyromagnetic ratio, μ_B Bohr's magneton and k Boltzmann's constant.

The coefficients B and C relate to the stiffness constant D, and the average mean square of the exchange interaction $\langle r^2 \rangle$ through the appropiate ferromagnetic model, having been shown that the Heisenberg model leads to satisfactory D values for most alloy compositions.

However, in some cases there is a profound disagreement between the D values obtained from inelastic neutron scattering and those resulting from magnetization measurements. In the Fe rich Invar-type alloys, for example, $D_{neut} \sim 1.5\ D_{mag}$.

In order to account for this disagreement, some authors have invoked the sensitivity of the saturation magnetization m(T) to mechanisms of thermal decay other that the collective (spin wave) excitations, such as the single particle, or Stoner excitations {3}.

The demagnetization of a system exhibiting both spin waves and single particle excitations, would be described by

$$m(T) = m(0)(1 - F(3/2, T/T_g)\ BT^{3/2} - D'T^{3/2}\ e^{-\Delta/kT}) \qquad (2)$$

for the strong ferromagnetic case, and

$$m(T) = m(0)(1 - BF(3/2, T/T_g)T^{3/2} - ET^2) \qquad (3)$$

for a weak ferromagnet, where the $T^{5/2}$ term of the spin wave expansion has not been considered. The aim of this work is to study the different contributions to the thermal demagnetization in amorphous alloys based in Fe, Fe-Ni, and Co.

Experimental and data analysis

The samples are amorphous ribbons of compositions $Fe_{81.5}B_{14.5}Si_4$, $Fe_{40}Ni_{38}Mo_4B_{18}$ and $Co_{70}Fe_5Ni_2Mo_3B_5Si_{15}$. In the following we call them samples A, B and C respectively. The former is from General Electric and the other two from Allied Corp.

Measurements of magnetic moment versus temperature at constant applied field, were performed with a SHE-SQUID magnetometer.

The magnetic field was always applied along the sample surface, in order to minimize demagnetizing effects. The values of the magnetic field corresponding to the fittings in table I, were H = 25 kOe for samples A and C and H = 50 kOe for sample B. These high values, as well as sample geometry considerations (our sample dimensions were around 10x3x0.025 mm), allow us to neglect the anisotropy and demagnetizing field contributions to the effective magnetic field H_{eff}.

The temperature was changed between 1.8 and 200 K (200 K is around 1/3 of the Curie temperatures for the three samples). Temperature control down to 0.01 K was possible, and we were able to measure magnetic moment variations of one part in 10^4.

The relevant physical parameters have been estimated by means of non-linear regressions. The strategy for locating the minima of the sum -of- squares function was the following: an initial search within the space of the N parameters was carried out using a SIMPLEX algorithm, similar to the method of Puzniak et al {4}. For such a purpose, all the free parameters were bounded to intervals obtained from physical considerations.

Once a stable minimum was located, a gradient search was started from the final point provided by the previous algorithm, using an implementation of the Gill-Murray method {5}. Finally, in cases were convergence within the feasible region was attained, a Montecarlo search, starting from the final point, was performed, in order to locate any other surrounding minima. The programme architecture was basically the one corresponding to the CERN computer library code MINUIT {6} where some improvements were

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introduced. The reported χ^2 statistic given in table I represents the sum of the residuals weighted by their normalized weights. This powerful fitting method allows us to get very low χ^2 values (in the range of 10^{-7}-10^{-8}) even for not very high experimental data density.

Results and discussion

In order to establish a point of departure to which we may refer in our later analysis, we have represented in Fig. 1, the curves $(\Delta m/m(0))(F_{3/2})^{-1}$ versus $T^{3/2}$ for the three samples studied.

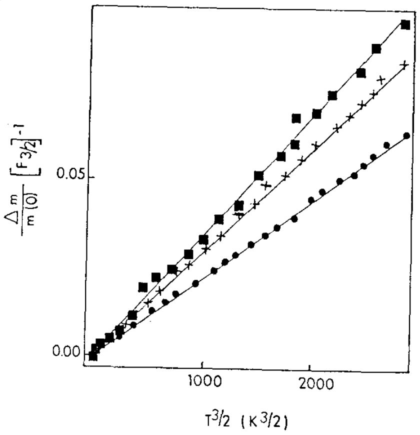

Fig. 1. $(\Delta m/m(0))(F_{3/2})^{-1}$ vs $T^{3/2}$ for + ($Fe_{81.5}B_{14.5}Si_4$), ■ ($Fe_{40}Ni_{38}Mo_4B_{18}$), ● ($Co_{70}Fe_5Ni_2Mo_3B_5Si_{15}$)

The linear dependence, apparent at first sight, only confirms the well known result of the predominant role of long wave-length spin waves in the thermal demagnetization of amorphous alloys.

The slopes of these curves give the values of B, the coefficient of the $T^{3/2}$ term in eq. (1) corresponding to a fitting where the term $T^{5/2}$ would have been dropped.

These values are shown in the first column of Table I, together with the values of T_g used (the g values have been 2.09, 2.05 and 2.18 for the samples A, B and C respectively). The other three columns contain the results obtained for the coefficients of the fittings to eqs. (1), (2) and (3). The first column will serve then as a reference throughout the subsequent discussion.

Sample A: $Fe_{81.5}B_{14.5}Si_4$

The results of the fittings for this sample are represented in the first row of Table I

As one can see, all three equations represent a slightly better fit to the m(T) behaviour than the $T^{3/2}$ term only. But the B coefficient for this latter case, $B=2.8\times10^{-5}$, hardly changes upon introduction of the single-particle excitations terms.

The largest correction to the $T^{3/2}$ term is thus due to the $T^{5/2}$ term.

The coefficients B and C are related to the spin wave stiffness constant D, and the average mean square of the exchange interaction $<r^2>$ {7} through:

$$B = \frac{\zeta(3/2)\, g\mu_B}{m(0)} \left(\frac{k}{4\pi D}\right)^{3/2} \quad (4)$$

$$C = \zeta\left(\frac{5}{2}\right) (g\mu_B/m(0)) \left(\frac{k}{4\pi D}\right)^{5/2} \left(\frac{3}{4}\pi\right) <r^2> \quad (5)$$

where $\zeta(\frac{3}{2})$ and $\zeta(\frac{5}{2})$ are the Rieman functions.

In table II, the values of D, $<r^2>$ and m(0) for our three samples, are presented.

Table II. Saturation magnetization, densities and spin wave parameters.

Sample	m(0) emu/g	ρ g/cm^3	D_0 meVÅ^2	$<r^2_0>$ (Å)^2
$Fe_{81.5}B_{14.5}Si_4$	182.0	7.4	94	15.9
$Fe_{40}Ni_{38}Mo_4B_{18}$	100.5	8.02	125	-
$Co_{70}Fe_5Ni_2Mo_3B_5Si_{15}$	81.2	8.1	178	112

The stiffness constant value obtained for this iron rich sample, D=94 meVÅ^2, falls between the values for the samples $Fe_{82}B_{18}$ (D=71 meVÅ^2) and $Fe_{81.5}B_{14.5}Si_4$(D=110 meVÅ^2), given in ref {8}.

This result is consistent with the observation {9},{10} that the replacement of B with any of the group IV elements C, Si and Ge, leads to a "hardening" of the stiffness constant.

Also, this D value is well below the one measured by inelastic neutron scattering D_{neut}=192Å, for a similar composition sample $Fe_{81}Si_9B_{10}$ {8}. This behaviour, not as yet understood, is displayed by most of the "Invar" type alloys {4}. Some authors {3} have claimed that this disagreement between D values obtained from direct measurement of the spin wave dispersion relation $\varepsilon = Dq^2$ (neutron inelastic scattering) and from thermal demagnetization m(T), is due to the contribution of single particle excitations of the strong ferromagnetic type to the m thermal decay.

As we have already explained, we haven't been able to detect any significant contribution of this effect.

The average mean square the exchange interaction, as obtained from eqs. (4) and (5) is $<r^2>$ =16 Å^2. This value, comparable to the one obtained in ref. {9} from the D_{neut} temperature dependence, corresponds to an exchange interaction extending slightly further than the nearest-neighbour distance.

Sample B: $Fe_{40}Ni_{38}Mo_4B_{18}$

The value of the B coefficient obtained from the fit to the $T^{3/2}$ term only expression, is $B=3.4\times10^{-5}$ (see table I). Notice that the introduction of the long-range exchange term $T^{5/2}$, as well as those of the single particle excitations, either conserves or improves the accuracy of the fit to the $T^{3/2}$ expression. The value of B corresponding to this later case, is scarcely changed in introducing the terms $T^{5/2}$ or T^2, whereas there is a significant decrease for the Stoner gap case. Taking into account the fact that the best value of m(0) is the same for all cases, one concludes that the most relevant correction to the long wave-lengh spin wave contribution must come from the single particle excitations of the strong type.

The small value of the Stoner gap Δ would reflect the fact that this alloy composition belongs to the transition region from strong to weak ferromagnetism, as has been proposed by O'Handley et al. {12}.

From the value of B for the fit to the strong ferromagnetic expression, one obtains the stiffness constant D=125 meV Å^2, which agrees well with the values in the literature {13}. This value of D is in the range of values obtained from direct measurement of the spin wave dispersion relation {11}. One argument in favor of the predominance of the Stoner gap term over the $T^{5/2}$ term is the small value reported in the literature for the second moment of exchange interaction $<r^2>$ =4.5 Å^2 {2} which would imply an exchange interaction of the localised Heisenberg ferromagnet type.

Table I: The fitted functions and the best fitted values of the parameters for samples (S) A ($Fe_{81.5}B_{14.5}Si_4$), B($Fe_{40}Ni_{38}Mo_4B_{18}$)and C($Co_{70}Fe_5Ni_2Mo_3B_5Si_{15}$).

S	$\Delta m/m(0)=BF_{3/2}T^{3/2}$	$\Delta m/m(0)=BF_{3/2}T^{3/2}+CF_{5/2}T^{5/2}$	$\Delta m/m(0)=BF_{3/2}T^{3/2}e-D'T^{3/2}e^{-(\Delta/k)T}$	$\Delta m/m(0)=BF_{3/2}T^{3/2}-ET^2$
	m(0)=180.6 emu/g	m(0)=182.0 emu/g	m(0)=182.1 emu/g	m(0)=182.1 emu/g
	$B=2.85x10^{-5}\ K^{-3/2}$	$B=2.34x10^{-5}\ K^{-3/2}$	$B=2.93x10^{-5}\ K^{3/2}$	$B=2.81x10^{-5}\ K^{-3/2}$
A	$T_g=3.5$ K	$C=0.33x10^{-7}\ K^{-5/2}$	$D'=7.1x10^{-7}\ K^{-3/2}$	$E=0.31x10^{-7}\ K^2$
			$(\Delta/k)=21$ K	
	$\chi^2=6.4x10^{-8}$	$\chi^2=3x10^{-8}$	$\chi^2=0.7x10^{-8}$	$\chi^2=0.6x10^{-8}$
	m(0)=100.5 emu/g	m(0)=100.6 emu/g	m(0)=100.6 emu/g	m(0)=100.5 emu/g
	$B=3.4x10^{-5}\ K^{-3/2}$	$B=3.5x10^{-5}\ K^{-3/2}$	$B=2.5x10^{-5}\ K^{-3/2}$	$B=3.2x10^{-5}\ K^{-3/2}$
B	$T_g=6.9$ K	$C=-0.09x10^{-7}\ K^{-5/2}$	$D'=66x10^{-7}\ K^{-3/2}$	$E=1.0x10^{-7}\ K^2$
			$(\Delta/k)=0.15$ K	
	$\chi^2=2.5x10^{-7}$	$\chi^2=2.5x10^{-7}$	$\chi^2=2.4x10^{-8}$	$\chi^2=3.1x10^{-8}$
	m(0)=81.2 emu/g	m(0)=81.2 emu/g	m(0)=81.2 emu/g	m(0)=81.2 emu/g
	$B=2.1x10^{-5}\ K^{-3/2}$	$B=1.9x10^{-5}\ K^{-3/2}$	$B=2.06x10^{-5}\ K^{-3/2}$	$B=2.05x10^{-5}\ K^{-3/2}$
C	$T_g=3.7$ K	$C=1.0x10^{-7}\ K^{-5/2}$	$D'=17x10^{-7}\ K^{-3/2}$	$E=0.91x10^{-7}\ K^2$
			$(\Delta/k)=86$ K	
	$\chi^2=3.7x10^{-8}$	$\chi^2=3.7x10^{-8}$	$\chi^2=0.15x10^{-8}$	$\chi^2=0.12x10^{-8}$

Sample C: $Co_{70}Fe_5Ni_2Mo_3B_5Si_{15}$

Close examination of the third row of table I, allows us to conclude that the fit to the expression containing the $T^{3/2}$-term only, is hardly affected by any of the correcting terms. m(0) stays within 10^{-4} emu/g of the $T^{3/2}$ fit value, whilst B undergoes slight variations, the most marked one corresponding to the addition of the spin wave term $T^{5/2}$

The B value obtained for this $T^{3/2}+T^{5/2}$ fit, leads to a value of D=178 meV Å^2. This is in satisfactory agreement with the reported values for samples of similar composition, which range between 153 meV Å^2 {13} and 198 meV Å^2 {14}. On the other hand, this D value falls within the range scanned by the inelastic neutron scattering results in Co-rich samples: 185 meV Å^2 for Co_4P {15} and 140 meV Å^2 for $Co_{70}P_{10}B_{20}$ {10}. This comes in support of our conclusion that single particle excitations are not an appreciable contribution to the thermal demagnetization of this sample.

The average mean square of exchange interaction $<r^2>$ resulting from the B, C values of this fit is obtained from eqs. (4) and (5): $r^2=112$ Å^2. This apparently overestimated value for an amorphous alloy, has already been reported and discussed for a $Co_{74}B_{26}$ sample {14}. There is previous evidence, then, that Co-B glassy alloys, unless Fe-B materials, behave very much like their crystalline counterparts.

Conclusions

We have measured the low temperature region of the magnetization versus temperature curves for glassy alloy samples of three different compositions: one iron rich, one Fe-Ni and one Co rich. Our aim was to extract distinct information about the different mechanisms contributing to the thermal demagnetization. It results from our study that only the Fe-Ni alloy exhibits clearly the presence of Stoner-like excitations, of the strong ferromagnetic type.

For the Fe-rich and Co-rich alloys, the short wavelength spin wave term $T^{5/2}$ has a significant contribution. In the Fe-rich sample single-particle excitations do not seem to explain the disagreement between the values of the stiffness constant as obtained from inelastic neutron scattering and magnetization measurements.

The second moment of exchange interaction has very different values in the three alloys studied: whereas the Fe-rich and Fe-Ni samples seem to behave as localised ferromagnets, according to their low $<r^2>$ values (which are of the order of the nearest neighbor distance), the Co-rich alloy exhibits long range interactions, not far from the crystalline behaviour.

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