Spin-wave excitation studies in amorphous $Ni_{50}Co_{40}P_{10}$ films

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Spin-wave excitation studies have been made using ferromagnetic-resonance techniques on amorphous $Ni_{50}Co_{40}P_{10}$ films in the range 4.2 to 310 K. The coefficient D is higher than that usually found for amorphous alloys. D follows well a $T^{5/2}$ dependence, indicating the alloy is a Heisenberg ferromagnet which, to our knowledge, is the first reported of its kind. D, calculated from the spin-wave spectrum and from B, the $T^{3/2}$ coefficient in the low-temperature variation of magnetization, shows excellent agreement. The above results confirm that amorphous alloys exhibit spin-wave excitations similar to crystalline ones. However, using our values of B, C, D, and a leads to an over estimation of the mean-square range of exchange interaction $\langle r^2 \rangle$, which would indicate the inadequacy of the models which were developed and found valid for crystalline alloys.

I. INTRODUCTION

Recently there have been a number of studies on the magnetic excitations and low-temperature magnetic properties of glassy metallic ferromagnets of the metal-metalloid type.[1-3]

It is generally concluded that several aspects of the magnetic spin-wave excitations observed in crystalline alloys are equally found in the amorphous ones. Better preparation techniques and a rich variety of amorphous alloys available today are certainly contributing to the consolidation of the concepts of magnetism in the amorphous state. In the long-wavelength-region many of these glasses exhibit well-defined spin-wave excitations with a normal ferromagnetic dispersion relation.

$$\hbar\omega(\vec{q}) = \Delta + dq^2 + Eq^4 + \cdots \ (Dq^2 >> 1) \cdots \ , \quad (1)$$

where Δ is defined as an effective anisotropy gap due to dipole-dipole interactions.[1] The approach to saturation at lower temperatures of the magnetization obeys well the relation

$$M(T) = M(0)(1 - BT^{3/2} - CT^{5/2} \cdots) \ . \quad (2)$$

In the treatment of simple spin-wave theory,[4] the coefficient B in Eq. (2) and the spin-wave stiffness coefficient D are related by the equation

$$B = 0.0587 |g\mu_B / M_{(0)}| \left(\frac{k_B}{D}\right)^{3/2} \ . \quad (3)$$

There have been cases, in glassy ferromagnets, however, where D determined by neutron inelastic scattering differs by as much as 30% from that calculated from B using Eq. (2).[1,2] However it should be noted that D was measured at relatively high temperatures and $D(0)$ was obtained by extrapolation following a $T^{5/2}$ law. On the contrary when D is calculated from spin-wave spectra, the agreement with that calculated from B was shown to be quite good.[5,6] Finally, as regards the temperature dependence of D, one can expect the following behavior. For a localized Heisenberg model, Dyson[7] has shown that the low-temperature expansion of D is given by $D(T) = D(0)(1 - aT^{5/2} + \cdots)$.[4] For an itinerant-electron model a T^2 term is expected to be dominant.[8]

Inspection of the literature reveals the lack of proper experimental evidence for the temperature dependence of D; while in Ref. 5 measurements are at three fixed temperatures, in Ref. 6 Suran *et al.* found an anomalous decrease in D at low temperatures. Thus good experimental verification is needed, which has been satisfied by this present work. In our program on amorphous materials, we stumbled on an almost ideal candidate, which is the electroless deposit of amorphous NiCoP films which have been shown to exhibit excellent properties[9,10] such as (i) narrow ferromagnetic resonance (FMR) linewidths, (ii) standing spin-wave spectrum, and (iii) practically negligible anisotropy, which means that FMR could yield directly the magnetization $4\pi M_s$ of the film from the fields for resonance parallel and perpendicular to the film plane. This is indeed very useful because the temperature dependence of $4\pi M_s$ could then be studied with an accuracy of about 0.1%. Further, the NiCoP alloy we have studied is rather unique in the sense that it has a very low metalloid concentration, 10 at.% as against 20 to 25 at.% nor-

mally encountered in the literature. This amorphous alloy enabled us to study the different aspects of the spin-wave excitations and to establish the temperature dependence of D unequivocally as described below. All the parameters are obtained on the same samples and in the same experimental run, which certainly has many advantages.

II. EXPERIMENTAL

Amorphous NiCoP films were prepared by electroless deposition with a Brenner-type solution, as detailed in Ref. 10. The composition of the films was determined by chemical analysis, and their thickness by a Talystep instrument. FMR was observed at 17.38 GHz in the range 4.2 to 310 K at intervals of 10 K. $4\pi M_s$ was also checked at 290 K by the vibration sample magnetometer. Hysteresis loops were also taken as a routine check to estimate the uniaxial anisotropy which was of the order of 20 Oe, which is negligible as compared to $4\pi M_s$ of about 7000 G.

III. RESULTS AND DISCUSSION

The amorphous state of the sample was confirmed by x-ray diffraction and also by transmission electron microscopy.[11] The composition of the films studied here is $Ni_{50}Co_{40}P_{10}$. Several small samples were cut from three different specimens of the same batch, and their FMR properties agreed to within 3%, indicating their high homogeneity. For low-temperature measurements two samples were chosen. Figure 1 shows the standing spin-wave spectrum for the field applied perpendicular to the film plane where one could observe as many as nine modes. The resonance fields of the various modes follow a quadratic dispersion law, whence $Hu - Hn = D/\gamma h[(\pi/t)^2 n^2]$, where u denotes the uniform mode, t is the thickness of the film, n is the number of the mode (in our case, $n = 0, 1, 2, 3$, etc.) and the other symbols have their usual meaning. Except for $n = 0$ to 2, the other

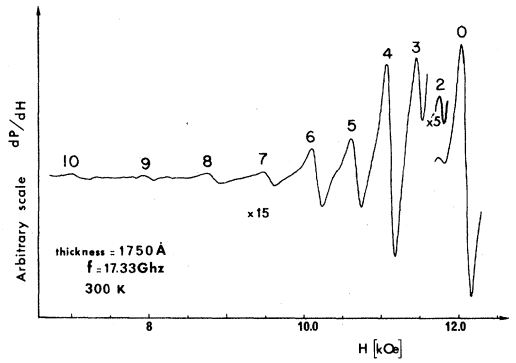

FIG. 1. Standing spin-wave spectrum of the amorphous $Ni_{50}Co_{40}P$ film at 300 K. One can note ten modes.

modes follow very well a quadratic behavior with deviations less than 1%, which enabled us to calculate D. As the spin-wave (SW) spectrum was observed down to 4.2 K, D could be calculated in the whole temperature range. The relevant results are tabulated in Table I. Data on other alloys are also given for comparison.

Let us first discuss D. When calculating this parameter from the spin-wave spectrum, some caution has to be exercised for the following reasons. Surface conditions perturb the spin-wave modes of only lower-order numbers, $0 < n < 3$, as in our case; the higher-order modes obey the quadratic dispersion law, and D is determined from this quadratic part of the spectrum. Also the exact pinning conditions cannot be easily determined which means it is difficult to choose for k_n the eigenvalues between $(\pi/t)^2 n^2$ and $(\pi/t)^2(n+1)^2$ (all symbols have the usual meanings). Under these conditions the measured value of D can be considered to be accurate to only about 10%, which of course is quite satisfactory for our purpose.[12] On the contrary, in neutron inelastic experiments, which are expensive, such problems as discussed above do not arise. At 290 K our experimental value of D is 210 meV Å^2, which is found to be

TABLE I. Spin-wave parameters and magnetization.

Compound	$M(0)$ gauss	B 10^{-6} K$^{-3/2}$	C 10^{-8} K$^{-5/2}$	$D(0)_{SW}$ meV Å^2	$D(0)$ cal meV Å^2	a 10^{-8} $T^{-5/2}$	Ref.
$Ni_{50}Co_{40}P_{10}$	560	15.2	1.0 ± 0.5	230 ± 2	231 ± 5	5.4	This work
$Fe_{75}P_{16}B_6Al_3$	1200	18.6		134 ± 5	117 ± 10		1
$(Fe_{0.65}Ni_{0.35})_{75}P_{16}B_6Al_3$	832	28.4		115 ± 3	114 ± 10		1
Crystalline Fe	1042	3.4	0.1	281	285		1
Crystalline Ni		7.5	1.5				13

somewhat higher than those reported for other amorphous alloys and rather closer to crystalline Fe.[1] This, in our opinion, could be due to the high concentration of transition metals in our case, as has been mentioned earlier. Figure 2 shows that the temperature dependence of D, and it is seen that D follows very well a $T^{5/2}$ dependence. It was found that $T^{3/2}$ dependence gave a poor fit. Therefore this amorphous alloy could be treated as a localized Heisenberg ferromagnet. The slope a, which is 5.4×10^{-8}, agrees well with that obtained in Ref. 2 (it is unfortunate that there is some confusion between a and a' in this paper). So it turns out that $T^{5/2}$ law holds good for a very wide range of temperature.

The temperature dependence of $4\pi M_s$ shows (Fig. 3) that $T^{3/2}$ law holds for a broad range of temperature. For $T > 180$ K, the coefficient C is found necessary. B and C values are given in Table I. The value of B in our case is smaller than those observed in other cases, probably again due to high transition-metal concentration as explained earlier. The value of C has to be treated with caution where the error is large, and it is estimated to be of the (Table I) right order. Using our value of $B = 15 \times 10^{-6}$ K$^{-3/2}$, one obtains through Eq. (3) $D = 231 \pm 5$ meV Å^2 which agrees indeed very well with $D(0) = 230$ meV Å^2 obtained from SW spectra. The different properties shown in Table I reveal that, in amorphous $Ni_{50}Co_{40}P_{10}$ film, the spin-wave excitations are similar to those in crystalline alloys.

From the results thus obtained, we can go a little further as follows. It can be shown that[1]

$$a \sim \frac{\langle r^2 \rangle k_B}{4} \frac{B}{D} , \tag{4}$$

where $\langle r^2 \rangle$ is the mean-square range of exchange interaction; other symbols have been described before. Substituting in the above our values of B, D, and a, one obtains $\langle r^2 \rangle = 38$ Å^2. This is certainly higher than the 22 Å^2 obtained from neutron scattering data of Ref. 1. This result is difficult to reconcile, since we have a reliable and fairly accurate determination of B, D, and a. One can also estimate $\langle r^2 \rangle$ by another method as follows. In the spin-wave theory with Heisenberg exchange,[1] C/B is governed by the mean-square range of exchange interaction $\langle r^2 \rangle$ by the relation

$$\langle r^2 \rangle = \frac{16}{3k_B} \frac{\zeta\frac{3}{2}}{\zeta\frac{5}{2}} \frac{CD}{B} . \tag{5}$$

Hence knowing B, C, and D one can estimate $\langle r^2 \rangle$, which in our case turns out to be 18 ± 9 Å^2. This is indeed closer to plausible values. The uncertainty here comes from C. Some disagreement for C/B, estimated from neutron-diffraction data for $\langle r^2 \rangle$ and from magnetization measurements, was reported in Ref. 1; however, a calculation made by us using their data showed that the agreement was satisfactory. In all events it has to be mentioned that relations (4) and (5) have been found valid for crystalline alloys and are probably not adequate for amorphous systems which call for more appropriate treatment for the amorphous state.

In conclusion, FMR studies on electroless deposited amorphous $Ni_{50}Co_{40}P_{10}$ films have enabled us to show that the temperature dependence of the spin-wave stiffness coefficient D can be well described by $T^{5/2}$ law and that of the magnetization by $T^{3/2}$ law. Also, D calculated from both spin-wave spectra and from B (magnetization measurements) show very good agreement. However, applying the classical theories of spin waves developed for the crystalline materials to estimate the mean-square range of exchange of interactions gives poor results in our case. This would indicate the inadequacy of the existing theories for the amorphous state.

At this stage of the present work, in our opinion, any agreement with a theory based on a certain model cannot be taken as an infallible proof of its validity, and by the same token, a disagreement does

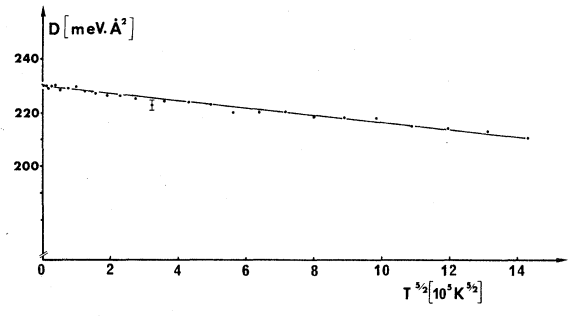

FIG. 2. $T^{5/2}$ dependence of the spin-wave-stiffness coefficient D.

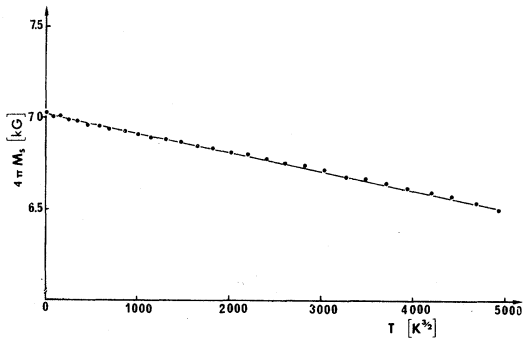

FIG. 3. $T^{3/2}$ dependence of the magnetization in amorphous $Ni_{50}Co_{40}P_{10}$ film.

not prove the contrary. All one could say is that recent papers on this subject do indicate and strengthen the viewpoint that the amorphous state itself seems to have no specific influence on the general behavior of spin waves, which are understandable. All the same, some detailed examination of the problem makes it clear that more experimental work on a wider variety of amorphous alloys is called for, which we hope could trigger more detailed theoretical work pertaining to the amorphous state, leading to a more sound basis for understanding amorphous materials.

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