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1992 J. Phys.: Condens. Matter 4 1373

(http://iopscience.iop.org/0953-8984/4/5/017)

J. Phys.: Condens. Matter 4 (1992) 1373–1378. Printed in the UK

Ferromagnetic resonance in amorphous $Fe_{20}Ni_{60}B_{20}$

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Received 11 June 1991, in final form 27 September 1991

Abstract. Ferromagnetic resonance has been used to study the isothermal annealing behaviour of metallic glass $Fe_{20}Ni_{60}B_{20}$ at temperatures between 300 K and 473 K. The temperature dependence of the resonance field (H_{res}) and the linewidth (ΔH) for both parallel vertical (PV) and parallel horizontal (PH) configurations suggest the formation of less random clusters leading to a second amorphous (paramagnetic) phase. Landau–Lifshitz–Gilbert theory has been used to obtain estimates of the effective magnetization (M_{eff}) and the Gilbert damping or relaxation parameter (λ). Well below and near the Curie temperature (T_c), λ varies linearly with saturation magnetization (M_s) whereas at intermediate temperatures the behaviour is non-linear.

1. Introduction

Ferromagnetic resonance (FMR) has been found to be a sensitive probe in observing effects of thermal aging, surface effects and structural and long-range magnetic ordering in metallic glasses [1–5]. Most previous FMR studies on metallic glasses have been done on iron-rich compositions exhibiting strong ferromagnetic behaviour at room temperature. In this paper we report the results of FMR studies on the nickel-rich $Fe_{20}Ni_{60}B_{20}$ alloy at temperatures between 300 K and 473 K to investigate changes in its magnetic properties induced by thermal annealing. The FMR data have been analysed to obtain the effective magnetization, M_{eff}, the Gilbert damping or relaxation parameter (λ) as a function of temperature, and the structural changes due to the thermal treatment.

2. Experimental details

FMR measurements are carried out on amorphous $Fe_{20}Ni_{60}B_{20}$ alloy at 9.3 GHz on a JEOL FE-3X spectrometer employing 100 kHz modulation with an incident microwave power level of about 1 mW.

As-received ribbons (about 1 mm wide and 20–25 μm thick) of amorphous $Fe_{20}Ni_{60}B_{20}$ alloy are carefully cut with scissors and electro-polished into rectangular specimens about 3 mm long in the manner described earlier elsewhere [6]. They are mounted on a teflon sample holder [7] to investigate its isothermal annealing behaviour by holding the sample in the microwave cavity at selected temperatures between 300 K

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0953-8984/92/051373 + 06 $04.50

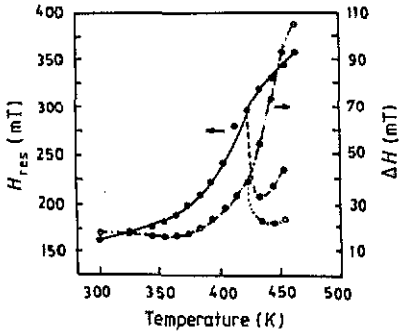

Figure 1. Variation of H_{res} and ΔH with temperature for a-$Fe_{20}Ni_{60}B_{20}$ in the PV configuration.

and 473 K for 20 min in PV (the plane of the ribbon parallel to the magnetic field and the long axis of the ribbon perpendicular to the magnetic field) and PH (both the plane of the ribbon and the long axis of the ribbon parallel to the magnetic field) configurations. Clean FMR signals (single line) were obtained in each case, indicating that the samples were magnetically homogeneous. The ribbons were annealed separately, in air and under vacuum, at 473 K, for 6 h and subsequently examined by x-ray diffractometry. The Curie temperature, T_c, of amorphous $Fe_{20}Ni_{60}B_{20}$ alloy is found to be 420 ± 5 K by Mössbauer spectroscopy.

3. *Results and discussion*

The variation of the resonance field, H_{res}, and the linewidth, ΔH, for amorphous $Fe_{20}Ni_{60}B_{20}$ alloy with temperature in the range 300–473 K in the PV configuration is shown in figure 1. H_{res} increases monotonically with temperature, showing a rapid rise after 383 K, while ΔH remains constant until 383 K and increases thereafter. The temperature-independent linewidths are of the same order of magnitude as observed by Spano *et al* [8]. Most interestingly, an additional resonance signal with considerably low H_{res} begins to appear at about 383 K. This signal increases very slowly in the beginning as the temperature of the sample is increased. Only at about 433 K does the signal become big enough to calculate H_{res} and ΔH without any ambiguity. At 473 K it becomes fully resolved. Figure 1 also shows H_{res} and ΔH for this additional peak. It is seen that ΔH for this line is smaller than ΔH of the original line. This additional signal is also observed when measurements are done in PH configuration and on a different piece of the sample taken from the same batch of the ribbon as shown in figure 2. The variation of H_{res} and ΔH with temperature in PH configuration is shown in figure 3, which is similar to figure 1. A comparison of linewidths (ΔH) shows that $[\Delta H]_{PV}$ is always larger than $[\Delta H]_{PH}$. This is in good agreement with the observation reported for amorphous $Fe_{40}Ni_{40}B_{20}$ alloy [9] and also for METGLAS 2826 [4].

The observed double-resonance behaviour in amorphous $Fe_{20}Ni_{60}B_{20}$ due to heat treatment at temperatures well below the crystallization temperature (~685 K) could be due to any of the following reasons: (i) structural relaxation, (ii) pre-crystallization, and (iii) amorphous phase separation, i.e. thermal evolution of two amorphous phases. The structural relaxation caused by heat treatment does not produce any drastic changes (for PV or PH configuration) in ΔH until 383 K, above which it rises steeply. However, H_{res}

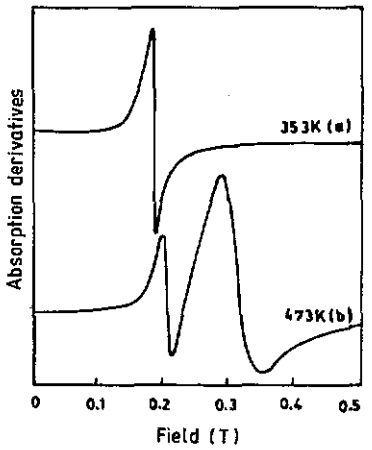

Figure 2. Magnetic-field dependence of the derivative of microwave power absorption for amorphous $Fe_{20}Ni_{60}B_{20}$ (PH configuration) at (*a*) 353 K and (*b*) 473 K.

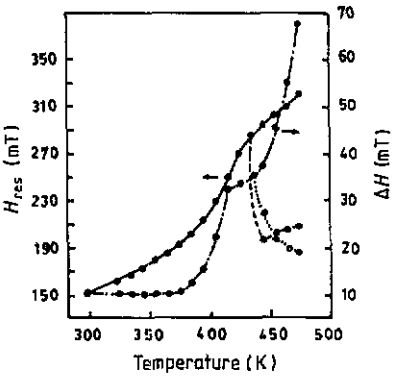

Figure 3. Variation of H_{res} and ΔH with temperature for a-$Fe_{20}Ni_{60}B_{20}$ in the PH configuration.

does increase slightly. A similar conclusion was also reached by Bork, who investigated $Fe_{40}Ni_{40}P_{14}B_6$ by FMR [10].

One is tempted to explain the appearance of the second FMR line as being due to crystallization and/or surface oxidation of the sample after it has been annealed. X-ray diffractograms of the annealed samples were found to be identical to those of the as-quenched samples—this rules out crystallization as the possible cause. A careful investigation by scanning electron microscope (SEM) of the surfaces of these samples also ruled out detectable crystallization and surface oxidation. Therefore, the additional FMR signal cannot be attributed to two distinct crystallized products. Moreover, if it were due to crystallization, the second peak would be expected to appear suddenly at the onset of the crystallization. The observations that this second peak develops slowly and that its intensity increases continuously as annealing is performed at higher and higher temperatures, indicate that there is thermal evolution of two amorphous paramagnetic phases that have different atomic environments to that of the original homogeneous glassy phase of $Fe_{20}Ni_{60}B_{20}$. The assertion, that these two resonances are due to two different paramagnetic amorphous phases and not due to thermally developed ferromagnetic inhomogeneities, arises from the observations that: (i) the annealing temperature (433 K) is higher than T_c (=420 ± 5 K), and (ii) Mössbauer measurements showed only broad quadrupole-split lines at 420 ± 5 K [11].

Annealing at temperatures greater than 383 K will cause diffusion of Fe and Ni atoms and changes in the neighbourhood of the Fe and Ni. This is expected to result in a rearrangement of atoms in the sample, and therefore, in view of the large Ni concentration, it is likely that two distinct distributions of amorphous clusters may evolve on annealing, one richer in Ni than the other. This may be looked upon as amorphous phase separation via cluster formation leading to two distinct FMR peaks. The phase that is rich in Ni will give rise to FMR at lower fields—this is what we observe. The exact nature (size and distribution) of clusters could only be determined by a technique like small-angle scattering, which is not available in our laboratory. However, these clusters are still

defined as amorphous since x-rays/SEM do not show any detectable crystallization. It may be noted that a similar observation of amorphous phase separation has been reported in the Mössbauer spectra of Co–Fe amorphous alloys [12].

Narrow FMR lines and rather small anisotropy fields (of the order of 30 Oe) gave us confidence in our sample homogeneity and quality and, therefore, the FMR data of a-$Fe_{20}Ni_{60}B_{20}$ have been analysed up to 423 K (before the phase separation takes place) in the framework of the Landau–Lifshitz–Gilbert theory to obtain estimates for the Gilbert relaxation parameter, λ, the effective magnetization, M_{eff}, and their temperature dependences. In this analysis we have assumed that (i) $g = 2.1$ (a value taken from literature for iron-based alloys [1, 13]), and (ii) $4\Pi M_{\text{eff}} = 4\Pi M_s$ (where M_s is the static saturation magnetization). The second assumption is based on the closeness of M_{eff} obtained by FMR and M_s obtained using a vibrating-sample magnetometer [13].

The equations relating the resonance field, H_{res}, to the effective magnetization, M_{eff}, and the linewidth, ΔH, to the Landau–Lifshitz damping parameter, λ, are

$$(\omega/\gamma)^2 = H_{\text{eff}}(H_{\text{eff}} + 4\Pi M_{\text{eff}}) \tag{1}$$

$$\lambda = (\Delta H)M_s\gamma^2/2\omega \tag{2}$$

where

$$H_{\text{eff}} = H_{\text{res}} - H_{\text{dem}} + H_{\text{an}}. \tag{3}$$

H_{dem} and H_{an} are the demagnetizing and anisotropy fields, respectively, $\omega = 2\Pi\nu$ is the frequency of the microwave field and $\gamma = -g|e|/2mc$ is the gyromagnetic ratio. H_{dem} is calculated using the expression [8]

$$H_{\text{dem}} = 8M_s \cot^{-1}[(a^2 + b^2 + c^2)^{1/2}\, c/ab] \tag{4}$$

where a, b, c are the measured values of width, thickness and length of the amorphous alloy ribbon. Equation (4) is valid only for the sample configuration in which the applied field, H_a, is parallel to the c axis (PH). So, an equation with suitably changed parameters has to be used for the configuration of the ribbon where H_a is parallel to the a axis (i.e. PV). H_{dem} values thus obtained are $\approx$8 and 25 Oe, for PH and PV configurations, respectively. Thus, H_{dem} effectively cancels out H_{an} ($\approx$30 Oe) for the present alloy in PV configuration to produce a resonance, as observed in the experiment. However, H_{eff} for the PH configuration has to be corrected for H_{dem} and H_{an}. In the following analysis, we have used corrected values of H_{eff} for the PH configuration.

Using the experimental values of microwave frequency, ΔH and also $g = 2.1$, the values of $4\Pi M_{\text{eff}}$ and λ are obtained for the amorphous $Fe_{20}Ni_{60}B_{20}$ sample in the temperature range 300 K $< T \leqslant T_c$. Figure 4(a) shows the plot of $4\Pi M_{\text{eff}}$ against T and figure 4(b) shows the variation of λ with $4\Pi M_s$ for amorphous $Fe_{20}Ni_{60}B_{20}$ alloy in the PV configuration. Similarly, in the PH configuration, the plot of $4\Pi M_{\text{eff}}$ against T is as shown in figure 5(a) and the variation of λ with $4\Pi M_s$ is as shown in figure 5(b). Data of $4\Pi M_{\text{eff}}$ against T are used to obtain T_c for both PV and PH configurations, which is found to be $T_c = 430 \pm 5$ K. This is about 10 K different from that obtained by Mössbauer measurements [11]. This could be due to slight differences in the nominal composition of the samples used for Mössbauer and FMR measurements. It is also well known that different experimental techniques give slightly different Curie temperatures for iron-based amorphous alloys having the same nominal composition.

Figure 5(b) shows that for $0 \leqslant 4\Pi M_s \leqslant 0.15$ T (near T_c) and $4\Pi M_s \geqslant 0.4$ T (well below T_c), $\lambda \propto M_s$. However, λ goes through a shallow minimum as $4\Pi M_s$ decreases

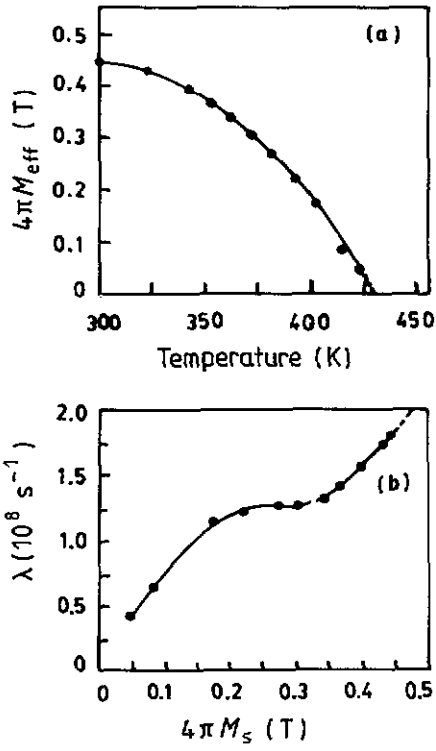

Figure 4. (*a*) The temperature dependence of $4\Pi M_{eff}$. (*b*) The relaxation parameter (λ) as a function of saturation magnetization for a-$Fe_{20}Ni_{60}B_{20}$ in the PV configuration.

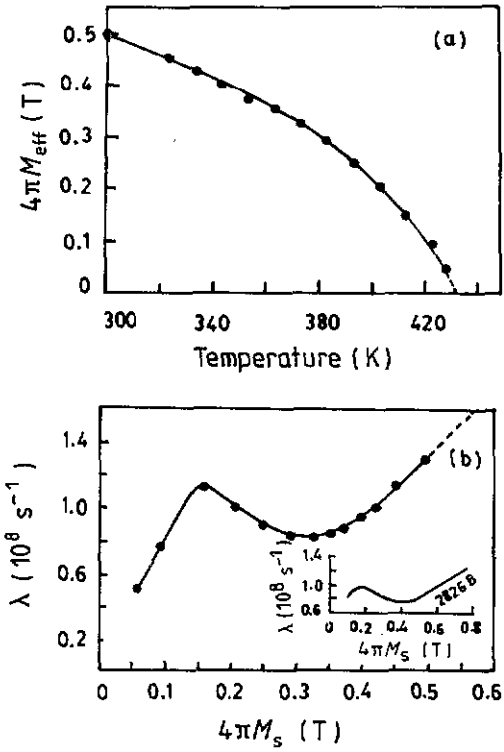

Figure 5. (*a*) The temperature dependence of $4\Pi M_{eff}$. (*b*) The relaxation parameter (λ) as a function of saturation magnetization for a-$Fe_{20}Ni_{60}B_{20}$ in the PH configuration.

Table 1.

Work/Sample	$4\Pi M_s$ (Oe)	λ ($10^8 s^{-1}$)	Frequency (GHz)
Present work on $Fe_{20}Ni_{60}B_{20}$ alloy	5008	1.3	9.3
Bhagat *et al* [1] for 2826B	4800	0.8	35

from 0.4 to 0.15 T. This behaviour of λ with $4\Pi M_s$ is similar to that of Metglas 2826B ($Fe_{29}Ni_{49}P_{14}B_6Si_2$) as reported by Bhagat *et al* [1] which is shown as an inset in figure 5(*b*). Table 1 also shows a comparison of values of $4\Pi M_s$ and λ for a-$Fe_{20}Ni_{60}B_{20}$ in PH configuration at room temperature and Metglas 2826B calculated using $g = 2.1$ [1]. λ in both cases is of the same order, affirming that the two assumptions made (i.e. $g = 2.1$ and $4\Pi M_{eff} = 4\Pi M_s$) in our analysis of FMR data are reasonable. Our results on amorphous $Fe_{20}Ni_{60}B_{20}$ along with that of Bhagat *et al* on Metglas 2826B and 2826 ($Fe_{40}Ni_{40}P_{14}B_6$) [1] indicate that a similar behaviour between λ and $4\Pi M_s$ may exist in other iron-based amorphous metallic systems.

In conclusion our FMR studies on amorphous $Fe_{20}Ni_{60}B_{20}$ alloy have shown the following.

(i) A short-duration annealing at temperatures of 433 K and above, but below the crystallization temperature, produces two well resolved magnetic-resonance peaks. It is suggested that these two resonances arise from annealing-induced phase separation in the original homogeneous amorphous phase into two distinct amorphous paramagnetic phases.

(ii) The variation of λ with $4\Pi M_s$ shows that, at temperatures well below T_c and near T_c, λ varies linearly with $4\Pi M_s$ and that the behaviour is non-linear at intermediate temperatures.

Acknowledgments

The work was supported by the Department of Atomic Energy (India) through grant no. 34/2/90-G. One of us (GVR) thanks the Council of Scientific and Industrial Research, New Delhi (India) for the award of Research Associateship.

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