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The influence of quenching rate on magnetic properties of amorphous ribbons

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Five sets of amorphous ribbons of the composition $Fe_{73.5}Cu_1Nb_3Si_{13.5}B_9$ produced with different quenching rates (QR) were studied in order to verify the influence of the topological disorder on magnetic properties. For this purpose several magnetic methods were applied. Significant differences have been found between the results of magnetic disaccommodation $H_e \cdot \Delta \mu/\mu$, pinning field H_p and stress dependence of the magnetic permeability $\mu/\mu_0(\sigma)$ of as-cast materials produced at different QR. The values of $H_e \cdot \Delta \mu/\mu$ and H_p are directly proportional to the disorder degree of the samples. The $\mu/\mu_0(\sigma)$ curves display broad maxima, which are found to be related to the quenched-in stresses. In contrast with these results, no significant effects of the amorphous structure on the Curie temperature and Mössbauer spectra were found, because other parameters are dominating.

I. INTRODUCTION

It is clear that the degree of quenched atomic disorder is one of the most important parameters which determines the physical properties of amorphous ferromagnets. This structural disorder is directly related to the average quenching rate (QR) at which the ribbons are produced.^{1,2} However, due to intrinsic experimental difficulties, studies of the influence of the amorphous state on the magnetic properties are scarce and nonsystematic. The lack of results hinders a complete understanding of the magnetization processes that occur in the amorphous bulk. Furthermore, these studies can lead to the development of a simple magnetic method to characterize amorphous structures.

In the present work, the influence of the initial amorphous structure on several magnetic properties is analyzed using five sets of amorphous ribbons produced with different QR. The samples have the nominal composition $Fe_{73.5}Cu_1Nb_3Si_{13.5}B_9$, which is the typical composition of the new nanocrystalline material.³ Previous studies indicate that the as-cast amorphous state could also have an influence in the subsequent nanocrystalline structure.⁴

II. EXPERIMENT

Four sets of samples were produced in the Istituto Elettrotecnico "Galileo Ferraris," Torino, Italy, by meltspinning, changing systematically the wheel velocity. The samples are called A, B, C, and D, ordered by decreasing wheel velocity (thickness varying from 15 to 40 μ m). It was experimentally found that the ribbon thickness appears to be inversely proportional to the wheel speed.⁵ It is expected that a thinner ribbon would have been quenched at the higher average rate.¹ Therefore, from the ribbon thicknesses we can estimate the maximum relative variation of the QR to be about 25%. Another sample, obtained from Vacuumschmelze GmbH, Hanau, Germany, (called Vac) was analyzed to verify the applicability of the magnetic methods.

The pinning field (H_p) , initial permeability (μ/μ_0) and magnetic disaccommodation $(H_e \cdot \Delta \mu/\mu)$ were measured at room temperature in as-cast samples using a computer controlled system (driving field $H_e < 10$ A/m, f=90kHz). The H_p measurement is described in details in Ref. 6. The initial permeability relaxation was measured between 0.25 and 8 s after a domain wall rearrangement, using the impulsive method.⁷ The magnetic disaccommodation is considered taking the maximum in the B vs ΔB curve, where the results for different ribbons may be compared.⁷

The Curie temperature (T_c) was determined from the temperature dependence of the magnetic permeability in low ac fields (7.5 A/m). The heating rate was 6 K/min.

Mössbauer spectra were measured at room temperature in the transmission mode, using a conventional constant-acceleration setup with a $Rh(^{57}Co)$ source.

III. RESULTS AND DISCUSSION

The amorphous state of each sample was initially characterized by the room temperature value of the magnetic disaccommodation. The amplitude of the magnetic disaccommodation taken as the maximum of the *B* vs ΔB curve, $H_e \cdot \Delta \mu/\mu$, is very sensitive to structural modifications in the amorphous bulk.⁸ Although controversial, both theories proposed to explain this phenomenon arrive at a direct relationship between the disaccommodation value and the number of structural defects, or amount of free volume, of the ribbons.^{7,9,10} It is well known that samples produced with higher QR (thus having larger free volume and higher quenched-in stresses) display higher disaccommodation amplitudes.⁸ Figure 1 shows the *B* vs



FIG. 1. The magnetic induction decay from $t_1=0.25$ s to $t_2=8$ s after a domain wall rearrangement, as a function of the magnetic induction $B(t_2)$. The disaccommodation values for all samples are calculated in the maxima of these curves.⁵

 ΔB curves from the samples A to D. The results of $H_e \cdot \Delta \mu/\mu$ for all the samples are given in Table I. Even when there is no narrow maximum in the $\Delta B(B)$ curve, the accuracy of the disaccommodation values is better than 5%, once the error due to the determination of the ribbons' cross section is avoided. The amplitude $H_e \cdot \Delta \mu/\mu$ decreases from sample A to D, as expected. In fact, the disaccommodation amplitude of sample D is four times lower than the amplitude of sample A, which, in turn, displays 3/4 of the amplitude of Vac sample. This fact indicates that the Vac sample has a greater degree of structural disorder, when compared to all the other ribbons.

The pinning field (H_p) is another particular characteristic of amorphous ferromagnets. It has been observed that a sharp variation in the magnetization curve occurs for very low critical fields (called H_p). Using a very simple model, it is found that the pinning field is directly proportional to the internal stresses present in the ribbon.¹¹ For $\sigma_{\text{ext}} = 0$, there is no detectable H_p in any sample. However, applying tensile stress, a clearly defined pinning field appears gradually. The H_p values found for $\sigma_{ext} \approx 100$ MPa are shown in Table I. The results agree qualitatively with the expected behavior. H_p decreases from sample A to D, indicating clearly the level of stress present in each sample. The Vac sample displays the highest H_p value, showing, as before, that this sample has the most disordered amorphous structure. However, the results do not agree quantitatively with disaccommodation data, e.g., the H_p observed for sample D is just a factor 3/5 lower than the H_p

TABLE I. Disaccommodation amplitudes $(H_e \cdot \Delta \mu/\mu)$, position of the maxima in the *B* vs σ curve (σ_{max}), pinning field (H_p) for $\sigma_{ext} \cong 100$ MPa, and Curie temperature (T_c).

Sample	$\frac{H_e \cdot \Delta \mu / \mu}{(10^{-2} \text{ A/m})}$	σ _{max} (MPa)	$H_p (A/m) \\ [\approx 100 \text{ MPa}]$	<i>T_C</i> (°C)
A	28	100	2.3	317
В	15	60	2.0	309
С	10	50	1.4	313
D	7	20	1.4	315
Vac	37	60	3.4	306

6604 J. Appl. Phys., Vol. 73, No. 10, 15 May 1993



FIG. 2. Stress dependence of the initial permeability (μ/μ_0) for samples A to D.

of sample A. This fact may indicate that the stress dependence of the pinning field cannot be described with such a simple model, and some extra contributions must be added to describe $H_p(\sigma)$ correctly.⁶

Figure 2 shows the stress dependence of the initial permeability (μ/μ_0) for samples A to D in the as-cast state. Comparing the curves it is worth noticing that the permeability of $\sigma_{\text{ext}} = 0$ is very similar for all the samples, whereas $\mu/\mu_0(\sigma)$ differs very drastically, indicating a strong variation in the quenched-in stresses (σ_i) . The approximate stress value where the maxima occur (σ_{max}) is written in Table I for all samples. The error is very large, because besides the error due to cross section determination, the curves are very broad, being difficult to determine the maximum exactly. However, the ratio between σ_{max} values for the different samples agrees surprisingly well with disaccommodation results (with the exception of Vac sample, which does not have a well defined maximum). There is no theory which describes correctly the stress dependence of the initial permeability, but, comparing with the above results, it is clear that the maximum must be directly related with the strength of the internal stresses. Additionally, the maximum in the μ vs σ curve must have a connection with the minimum observed in the stress dependence of the coercivity $H_{c}^{4,6}$

The Curie temperature is very sensitive to changes in the density ρ of the amorphous materials,¹² and therefore could be suitable to study changes in the amorphous state of the ribbons. However, as can also be seen in Table I, measurements of T_C do not directly reflect the initial amorphous state. This is obvious because during the T_C determination the samples suffer annealing, which, in turn, produces irreversible changes in the amorphous bulk (stress relief, free volume reduction, etc...). The kinetics of structural relaxation is very complex, depending on several factors, including the as-cast structure of the samples.¹³ Hence, it is not possible to extract any information about differences in the initial amorphous state of the material from T_C data.

Mössbauer spectra for samples A–D, measured at room temperature, show strongly overlapping magnetically split lines characteristic of amorphous ferromagnets. Good fits were obtained with asymmetric Gaussian hyper-

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TABLE II. Fitted parameters from Mössbauer spectra: most probable hyperfine field (B_{ij}) , half-maximum width of $P(B_{hf})$ (ΔB), average B_{hf} ($\langle B_{hf} \rangle$), and 2nd to 3rd line intensity ratio $(I_{2,5}/I_{3,4})$.

Sample	$B_0(T)$	$\Delta B(T)$	$\langle B_{nf} \rangle(T)$	I _{2,5} /I _{3,4}
A	23.0	10.2	20.6	2.7
в	23.4	09.8	21.2	2.6
С	23.7	10.5	21.1	3.1
D	23.3	10.5	20.6	2.7

fine field distributions including a linear correlation between B_{hf} and isomer shift.¹⁴ Sextet line intensity ratios were left as free parameters. While $I_{1,6}/I_{3,4}$ is always close to 3, $I_{2,5}/I_{3,4}$ is substantially larger than 2, indicating a preferential orientation of the local magnetization parallel to the ribbon plane. Relevant fitting parameters are summarized in Table II.

It can be seen in Table II that the parameters related to $P(B_{hf})$ change very little from sample to sample and do not appear to exhibit any definite correlation to sample quenching rate. The insensitivity of Mössbauer spectra to the QR of amorphous magnets has already been noticed.¹⁵ According to Allia *et al.*, the B_{hf} distribution is primarily determined by the degree of local magnetic disorder, whereas macroscopic magnetic properties such as those discussed in this work are most sensitive to local stresses. These, in turn, are more dependent on the QR than on the degree of magnetic disorder. Our results are consistent with those conclusions.¹⁵

IV. CONCLUSIONS

We have used several magnetic methods to study the influence of the quenching rate on the magnetic properties of amorphous materials. It was found that the magnetic properties which are particular of amorphous ferromagnets, like the magnetic disaccommodation and pinning field, appear to be greatly influenced by the alloy's QR. The stress dependence behavior of the magnetic permeability is found also to be very sensitive to structural changes. The position of the maxima in the $\mu/\mu_0(\sigma)$ curves agrees surprisingly well with the above mentioned results, indicating that the existence of a maximum must be related to the fraction of atomic-level stress fluctuations. On the other hand, the Curie temperature varies from 306 to 317 °C, but not following a direct relationship with the QR. This may be explained by considering that the structural relaxation kinetics must also depend on the as-cast amorphous structure. Mössbauer spectra were found to be relatively free from the effect of variations of topological disorder. This result indicates that the variation of the pressure-density fluctuations in differently quenched systems does not modify the value of the hyperfine field, which is rather dominated by the effect of the local magnetic disorder, and not substantially influenced by small variations of the QR.

In conclusion, when materials with the same composition are produced with different QR, the magnetic properties can vary drastically. This fact prevents a reliable comparison between results obtained from different samples produced by different groups. The initial amorphous state should always be taken into account. For this purpose, some very sensitive methods could be applied, like magnetic disaccommodation, stress dependence of permeability and pinning field. Although experimentally simple, these methods can only determine roughly (and comparatively) the amorphous structure of the samples. Further research effort is needed to describe correctly these phenomena, in order to obtain a precise description of the effect of the topological disorder on the physical properties of amorphous solids.

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