

Journal of Magnetism and Magnetic Materials 203 (1999) 211-213



www.elsevier.com/locate/jmmm

## The effective anisotropy of nanocrystallized Co-based alloys P. García-Tello<sup>a</sup>, N. Murillo<sup>b</sup>, J. González<sup>b,\*</sup>, E. Amano<sup>c</sup>, R. Valenzuela<sup>c</sup>, J.M. González<sup>d</sup>

<sup>a</sup>Departmento de Física Aplicada I, EUITI, UPV/EHU, 20011 San Sebastián, Spain

<sup>b</sup>Departmento de Física de Materiales, Facultad de Químicas, UPV/EHU, P.O. Box 1072, 20009 San Sebastián, Spain

<sup>c</sup>Instituto de Investigaciones en Materiales, UNAM, P.O. Box 70-360, Mexico D.F., Mexico

<sup>d</sup>Instituto de Ciencia de Materiales, CSIC (Madrid), Cantoblanco, 28049 Madrid, Spain

## Abstract

The influence of the thermal treatments on the effective magnetic anisotropy of  $\text{Co}_{66}\text{Fe}_4\text{Mo}_2\text{Si}_{16}\text{B}_{12}$  metallic glass is examined through the results corresponding to the measurement of the transverse-biased initial susceptibility ( $\chi_t$ ). The annealing lead to a remarkable improvement of the magnetic softness, reaching the optimum properties at 648 K and 60 min. The analysis of the  $\chi_t$  data in terms of the micromagnetic magnetization ripple theory allowed us to evidence the occurrence in the annealed samples of coupled magnetization regions larger than the average grain size. © 1999 Elsevier Science B.V. All rights reserved.

Keywords: Anisotropy-magnetic; Annealing effect; Nanocrystalline materials

As it has been widely reported Fe(CuNb)SiB nanocrystalline alloys obtained by devitrifying the amorphous precursor exhibit a very attractive combination of soft magnetic properties. The origin of these excellent soft magnetic properties has been proposed by Herzer [1] to be a nanometer size effect, i.e., when the Fe(Si) BCC grain size is of a few nanometers ( $\approx 10$  nm), the effective nanocrystalline anisotropy is averaged out due to intergranular coupling over exchange length interaction larger than the grain size. The anisotropy reduction combines with the near-zero magnetostriction, associated to the two-phase nature of the sample, to produce excellent soft magnetic properties [1,2].

More recently, the hysteretic properties [3] and saturation magnetostriction [4] have been reported for this nanocrystalline and amorphous Co-rich alloy. Examination, by Transmission Electron Microscopy of these samples revealed the presence of nanocrystals with an average grain size of 20 nm [3]. In the present work, we deal with the influence of the thermal treatment on the effective magnetic anisotropy of the Vitrovac<sup>R</sup> 6025 alloy. Annealing conditions were chosen so as to result in the nanocrystallization of the samples.

The originally amorphous samples (nominal composition  $Co_{66}Fe_4Mo_2Si_{16}B_{12}$ ) were kindly supplied by Vacuumschmelze, Hanau, Germany in the as-cast state. Before the thermal treatments the original ribbon (4 mm wide and 25 µm thick) were cut by acid etching into disk (3 mm diameter) for the susceptibility measurement. The disk-shaped samples were annealed under argon atmosphere at temperatures ranging from 523 to 750 K and for different annealing times.

The anisotropy field of the samples was determined from the measurement of the transverse-biased initial susceptibility,  $\chi_t$ , [4]. For that purpose, the samples (cut into disks) were simultaneously submitted to a saturating DC field,  $H_1$ , and to an AC (73 Hz) field,  $H_2$ , perpendicular to  $H_1$  and small enough to deviate reversibly the direction of the mean magnetization from that of  $H_1$ . The component of the total magnetization parallel to  $H_2$  was measured by means of a pick-up coil and a lock-in amplifier tuned to the frequency of the field  $H_2$ .

<sup>\*</sup>Corresponding author. Tel.: + 34-43-21-66-00; fax: + 34-43-21-22-36.

E-mail address: wabmuhen@sq.ehu.es (J. González)



Fig. 1. Isothermal evolution of the anisotropy field,  $H_k$ , with  $t_{ann}$  for Co<sub>66</sub>Fe<sub>4</sub>Mo<sub>2</sub>Si<sub>16</sub>B<sub>12</sub> samples annealed at: ( $\bullet$ ) 648, ( $\bigcirc$ ) 673 and ( $\Box$ ) 773 K.

The anisotropy field,  $H_k$ , associated with the effective anisotropy,  $K_{eff}$ , and the dispersion field,  $H_d$ , developed by the inhomogeneities of the magnetization distribution of the as-quenched and treated samples were determined from the measurements of the transverse-biased initial susceptibility. More details of this method can be found in Ref. [5].

The variations of  $H_k$  with the annealing time in the heat-treated samples are presented in Fig. 1 for different treatment temperatures. As can be seen in this figure, the anisotropy field decreases with  $t_{ann}$  to a minimum value of the order of a few Oersted. The observed behaviour should be ascribed to the appearance (for  $t_{ann}$  range in which  $H_k$  reaches the minimum) of tiny crystallites with grain sizes of the order of a few nm which are embedded in an amorphous matrix [3]. These nanocrystals are strongly coupled which leads to a reduction (by average) of the effective anisotropy [1]. The increase of  $H_k$  at higher  $t_{ann}$  could be interpreted as due to grain growth or the appearance of the new phase [3].

The dependence of the dispersion field,  $H_d$ , on the  $t_{\rm ann}$  is shown in Fig. 2 (for different annealing temperatures). It can be seen that  $H_d$  behaves in a similar way to  $H_k$ . Regarding the results corresponding to the dispersion field,  $H_d$ , it is possible to remark that this parameter had, in most of the samples, a value clearly larger than the effective anisotropy field,  $H_k$ . This should reflect the disordered nature of the samples (characterized either by the distribution of the easy-axis direction, in the axis of nanocrystalline samples, or by the lack of long-range structural order, in the case of the amorphous phase). This correlation between  $H_k$  and  $H_d$  can be understood as a consequence of the very good homogeneity which exhibit all the samples treated around 648 K during 1 h with an average grain size of about 15 nm [3]. The lower  $H_{\rm d}$  and  $H_{\rm k}$  values measured at these annealing conditions could result from the compromise between the intensity of the intergranular exchange coupling (increasing as the volume of the nanocrystalline phase) and the degree of homogeneity of the grain-size distribution. The grain-size



Fig. 2. Dependence of the dispersion field,  $H_d$ , with  $t_{ann}$  for  $Co_{66}Fe_4Mo_2Si_{16}B_{12}$  samples treated isothermally at: (•) 648, ( $\bigcirc$ ) 673 and ( $\square$ ) 773 K.

homogeneity could correspond to a predominance of the nucleation process during crystallization.

According to Hoffmann [5], the interaction of the local magnetic moments with the diverse microstructural features present in the sample results in small deviations from the direction of the average magnetization (magnetization ripple), the local moments are coupled in ellipsoidal regions where the minor axis is directed parallel to the average magnetization direction. This particular magnetization distribution causes a dependence of  $H_d$  on the applied field proportional to  $(H_1 \pm H_k)^{(-1/4)}$  which is followed by our samples. In this way, the micromagnetic theories of magnetization in ripple [6] allow to evaluate the minor,  $R_{qe}$ , and major,  $R_{me}$ , semiaxes of the coupled regions according to [4]

$$R_{\rm qc} = \{AH_{\rm k}/K_{\rm eff} (H_1 \pm H_{\rm k})\}^{1/2} \tag{1}$$

and

$$R_{\rm mc} = 2^{3/2} d^{1/2} A^{1/4} K_{\rm eff}^{3/4} M_{\rm s} [H_{\rm k}/(H_1 \pm H_{\rm k})]^{3/4}, \qquad (2)$$

where A is the exchange constant ( $A = 10^{-10}$  J/m) and d is the thickness of the sample (around 25 µm).

In Fig. 3a and Fig. 3b we have plotted the variation of these parameters with  $t_{ann}$ . It is worth noting that the major semiaxes of the coupled regions (of the order of  $10^{-3}$  cm) are much larger than the average grain size measured by TEM in the crystallized samples [3]. The dimensions of the minor semiaxes ( $\sim 10^{-7}$  cm) are of the order of the average grain diameter. We should remark that the large transverse dimensions of the coupled units evidence that the magnetostatic interactions are relevant in the formation of these regions.

Concluding, from the analysis of the transverse susceptibility results in an amorphous and nanocrystalline Corich alloy (very low magnetostriction), it was possible to obtain the typical dimension of the regions of coupled magnetic moments. Considering that these dimensions, especially when measured perpendicularly to the mean magnetization direction, were larger than the mean grain



Fig. 3. Annealing time dependence of the minor (a),  $R_{qc}$ , and major (b),  $R_{mc}$ , semiaxis of the coupled regions in  $Co_{66}Fe_4Mo_2Si_{16}B_{12}$  samples annealed at: (•) 648, ( $\bigcirc$ ) 673 and ( $\square$ ) 773 K.

diameter, and taking into account the magnitude of the reduction of the magnetic anisotropy, we propose that the enhancement of the soft magnetic properties in this alloy is linked to the decrease in the magnitude of the interactions between the microstructure and the magnetization due to the occurrence of intergranular coupling.

This work was supported in part by the Excma Diputación Foral de Guipúzcoa, Spain, and in part by DGAPA (IN101093)-UNAM, México.

## References

- [1] G. Herzer, IEEE Trans. Magn. Mag- 26 (1990) 1397.
- [2] Y. Yoshizawa, S. Oguma, K. Yamamuchi, J. Appl. Phys. 64 (1988) 6044.
- [3] P. Quintana, E. Amano, R. Valenzuela, J.T.S. Irvine, J. Appl. Phys. 75 (1994) 6490.
- [4] J.M. González, N. Murillo, J. González, J.M. Blanco, J. Echeberria, J. Mater. Res. 11 (1996) 512.
- [5] H. Hoffmann, IEEE Trans. Magn. Mag-15 (1979) 1215.
- [6] H. Hoffmann, IEEE Trans. Magn. Mag-4 (1968) 32.