Title: Application of the coupling model to magnetic after effects in the Fe72Co10Nb6B12 amorphous alloy

Author: Artur Chrobak, Grażyna Chełkowska, Grzegorz Haneczok, P. Kubik, Ł. Madej

Citation style: Chrobak Artur, Chełkowska Grażyna, Haneczok Grzegorz, Kubik P., Madej Ł. (2009). Application of the coupling model to magnetic after effects in the Fe72Co10Nb6B12 amorphous alloy. "Acta Physica Polonica A" (Vol. 115, nr 1 (2009), s. 396-398).

Proceedings of the European Conference "Physics of Magnetism" (PM'08), Poznań 2008

Application of the Coupling Model to Magnetic After Effects in the $Fe_{72}Co_{10}Nb_6B_{12}$ Amorphous Alloy

A. Chrobak, G. Chełkowska
Institute of Physics, University of Silesia
Uniwersytecka 4, 40-007 Katowice, Poland
G. Haneczok, P. Kubik and Ł. Madej
Institute of Materials Science, University of Silesia
Bankowa 12, 40-007 Katowice, Poland

It was shown that magnetic reluctivity measured versus time after demagnetization for pre-annealed samples of the $Fe_{72}Co_{10}Nb_6B_{12}$ amorphous alloy exhibits highly non-exponential behavior which can be described by the coupling model. The relaxation intensity and the coupling parameter describing correlation effects in free volume diffusion decrease with increasing 1 h annealing temperature indicating annealing out of free volume and formation of iron clusters in amorphous matrix.

PACS numbers: 75.50.−y, 75.75.+a

1. Introduction

It is known that soft magnetic properties of amorphous alloys based on iron can be improved by applying a suitable annealing at temperatures close to the crystallization temperature. After such annealing initial magnetic permeability can be of the order of 10^4 and coercive field even below 1 A/m [1–6]. This effect is usually explained by formation of α-Fe nanograins embedded into amorphous matrix. The microstructure with differently oriented nanograins averages out magnetic anisotropy and gives the enhancement of magnetic permeability [1, 4]. In some cases (e.g. [1, 5, 6]) the permeability enhancement effect may be observed without formation of a nanocrystalline phase. This was recognized in [5, 6] for different amorphous alloys and the observed effect was attributed to annealing out of free volume and internal stresses leading to formation of iron clusters with different magnetic order i.e. to the so-called relaxed amorphous phase [7–9].

The aim of the paper is to study free volume diffusion in $Fe_{72}Co_{10}Nb_6B_{12}$ amorphous alloy in the context of the permeability enhancement effect. Magnetic properties of this alloy were studied in [5] and it was found that permeability determined at room temperature for samples annealed for 1 h at temperatures T_a (ranging from 300 K to 900 K) shows a maximum at $T_a = 675$ K (see Fig. 1 in [5]). Moreover, it was also shown that the optimized microstructure is free of α-Fe nanograins, so the examined alloy is a suitable alloy for studying the influence of structural relaxation on magnetic properties. On the one hand, the paper concentrates on diffusion in correlated system and, on the other hand, we intend to clarify the mechanism of the permeability enhancement effect proposed in [5, 6].

2. Experimental

The $Fe_{72}Co_{10}Nb_6B_{12}$ amorphous alloy examined in this paper was fabricated by melt spinning in the form of strips with thickness and width of 25 μm and 1 cm, respectively. As quenched ribbons were annealed for 1 h at temperatures selected basing on the results of [5] i.e. 300 K (as quench state), 500 K, 625 K, 675 K and 700 K. These annealing as it was shown in [5] changes sample microstructure from the as quenched state (amorphous) to the fully relaxed amorphous state. For the pre-annealed samples (10 cm in length) magnetic susceptibility χ at field 0.5 A/m was measured versus time after demagnetization by making use of HP RLC 4284A meter working at frequency about 1 kHz. Figure 1 shows normalized magnetic reluctivity $r(t)$ ($r = 1/\chi$) versus time obtained for all examined samples. The normalization was carried out by subtracting $r(0)$ and dividing by the maximum value of reluctivity for the as quenched state. It can be seen that with increasing annealing temperature the curvature of the observed $r(t)$ curves is quite different indicating the influence of sample microstructure (progress in structural relaxation) on magnetic properties.

3. Coupling model, data analysis and discussion

In ferromagnetic materials any diffusing atom possessing a magnetic moment interacts with magnetization

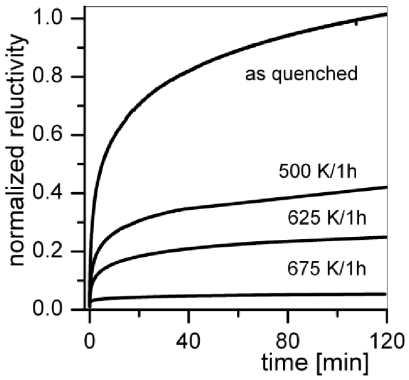

Fig. 1. Normalized magnetic reluctivity versus time after demagnetization determined at room temperature for the $Fe_{72}Co_{10}Nb_6B_{12}$ amorphous alloy preliminary annealed at different temperatures.

which modifies its diffusion walk. Application of an external weak magnetic field changes energetic levels of different sites and according to the Néel approach leads to magnetic after effects [2, 10–13]. Macroscopically it is observed as a time increase in magnetic reluctivity measured after demagnetization. The most spectacular case is the well known Snoek relaxation observed in alpha iron containing carbon atoms in solid solution form [2].

In ferromagnetic amorphous materials diffusing object consists of atomic pairs of alloying additions in the vicinity of free volume as it was considered by Kronmüller (see Figs. 2–5 in [11]). An individual atomic jump requires a correlated atomic displacement of the nearest neighbors so some correlation effects would play an important role [14, 15]. Let us notice that for a random walk the mean displacement $\langle x(t)\rangle$ of a diffusing atom is zero but the $\langle x(t)^2\rangle$ is proportional to the diffusivity D and time t, so we have $\langle x(t)^2\rangle \propto Dt$. In correlated systems one can define the so-called diffusion exponent d by the relation

$$\sqrt{\langle x^2(t)\rangle}^d \propto Dt\,, \tag{1}$$

which allows introducing the time dependent transition rate $W_{\alpha\alpha'}$ (the frequency of jump from one site — let us say α — to another one α') by taking into account that

$$\frac{\sqrt{\langle x^2(t)\rangle}^d}{t} \propto D \quad \text{and} \quad D \propto W_{\alpha\alpha'}\,. \tag{2}$$

According to Eqs. (1) and (2) $W_{\alpha\alpha'}$ can be expressed as

$$W_{\alpha\alpha'} = W_0 t^{-n}, \tag{3}$$

where W_0 is the unperturbed frequency of jump and n is the coupling parameter defined by the relation $n = 1 - \frac{2}{d}$ and restricted to $0 \le n \le 1$ since $d \ge 2$. Let us notice that for the random walk $d = 2$ and $n = 0$ which means that the transition rate is constant and the corresponding response function is proportional to $\exp(-t/\tau)$, where τ is the relaxation time depending on temperature via the Arrhenius relation. In correlated systems the transition rate is time dependent which leads to the response function known as the universal one (i.e. $\propto \exp(-(t/\tau)^{1-n})$) [2, 14, 15], as it can be applied to many correlated systems like polymers, dielectrics, metals etc. Finally, for magnetic reluctivity measured in this paper with the initial condition $r(0) = r_0$ the model function can be written as

$$r(t) = I[1 - \exp(-(t/\tau)^{1-n})] + at + r_0\,, \tag{4}$$

where I is the intensity of the observed process and parameters a and r_0 correspond to the irreversible component of the measured magnetic after effects and can be estimated from a long time tail of the measured curves.

Numerical analysis was carried out by applying Eq. (4) as the model function in the fitting procedure (for detail see [13]). Parameters to be fitted are: I, τ, n plus a and r_0. In all cases we have got a very good agreement between theory and experiment with n ranging from 0.75 to 0.50. This means that the observed phenomenon is highly non-exponential. An example of the fit quality is presented in Fig. 2. It should be underlined that the relative difference between theory and experiment does not exceed 0.05%. The results of the numerical analysis are presented in Fig. 3 where the fitted parameters are plotted versus the 1 h annealing temperature T_a, so versus a degree of the structural relaxation progress.

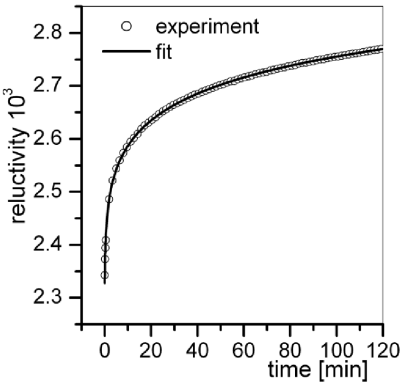

Fig. 2. Experimental data from Fig. 1 for the as quenched sample and the theoretical curve calculated according to Eq. (4) (only every 10th point is plotted).

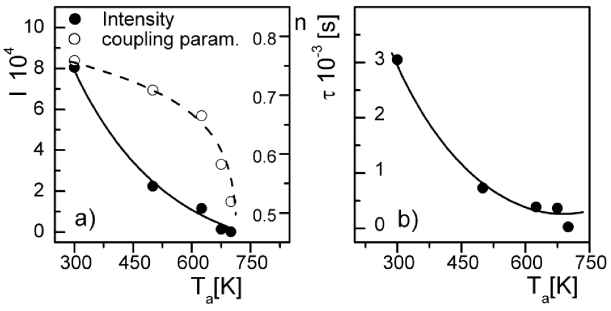

Fig. 3. Results of numerical analysis of the data presented in Fig. 1 carried out by making use of Eq. (4): (a) intensity I and coupling parameter n versus 1 h annealing temperature T_a, (b) relaxation time τ versus T_a.

According to Fig. 3a the intensity I of the observed phenomenon decreases with increasing T_a indicating annealing out of free volume (and internal stresses) and proving that after the optimization annealing thermal/time instabilities characteristic of the as quenched state practically disappear.

Figure 3a shows also that the coupling parameter n decreases with T_a but the curvature of this dependence is quite different. Initially in the range 300 K $\leq T_a \leq$ 625 K n decreases relatively slow (i.e. about 0.1 per 300 K) and its main drop is observed for $T_a >$ 625 K (0.15 per 75 K) so just before the optimization annealing. This fact suggests that the decrease in the coupling parameter is correlated with the increase in magnetic permeability observed in [5] and is caused by a certain kind of short range ordering cutting out the correlations. Such an order causes that internal stresses and what follows magnetoelastic energy are significantly reduced. Finally, one can conclude that the obtained results are consistent with the idea of formation of iron clusters in amorphous surroundings being the final stage of structural relaxation just as it was proposed in [5, 6].

Figure 3b shows the relaxation time τ versus T_a. It can be seen that τ decreases with increasing T_a which means that the progressing annealing out of free volume causes a cut-out of the long time components of the full spectrum of relaxation times.

4. Conclusions

The paper shows that magnetic after effect technique and the coupling model describing diffusion processes in correlated systems are applicable to the examination of structural relaxation in amorphous ferromagnetic alloys. The main conclusions of the paper can be summarized as follows: (i) magnetic reluctivity measured as a function of time for samples of the $Fe_{72}Co_{10}Nb_6B_{12}$ amorphous alloy pre-annealed at temperatures 300 K $\leq T_a \leq$ 700 K (with different progress in structural relaxation) shows highly non-exponential behavior which can be described by the coupling model, (ii) the relaxation intensity and the coupling parameter describing correlation effects in free volume diffusion decrease with increasing T_a indicating annealing out of free volume and a short range ordering which is consistent with the idea of formation of iron clusters in amorphous matrix, and (iii) the progress in structural relaxation causes a cut-out of the long time components of the spectrum of relaxation times.

References

[1] G. Haneczok, J. Rasek, *Def. Diff. Forum* **224-225**, 13 (2004).

[2] G. Haneczok, J. Rasek, *Def. Diff. Forum* **188-190**, 3 (2001).

[3] T. Kulik, *J. Non-Cryst. Solids* **287**, 145 (2001).

[4] G. Herzer, L.K. Varga, *J. Magn. Magn. Mater.* **215-216**, 506 (2000).

[5] A. Chrobak, G. Haneczok, P. Kwapuliński, D. Chrobak, Z. Stokłosa, J. Rasek, *J. Alloys Comp.* **423**, 77 (2006).

[6] G. Haneczok, J.E. Frackowiak, A. Chrobak, P. Kwapuliński, J. Rasek, *Phys. Status Solidi A* **202**, 2574 (2005).

[7] R. Lorenz, J. Hafner, *J. Magn. Magn. Mater.* **139**, 209 (1995).

[8] T. Sitkowski, L. Dominguez, K. Kułakowski, A.Z. Maksymowicz, *J. Magn. Magn. Mater.* **254-255**, 413 (2003).

[9] J. Marcin, A. Wiedenmann, I. Skorvanek, *Physica B* **276- 278**, 870 (2000).

[10] L. Néel, *J. Phys. Radium* **13**, 249 (1952).

[11] H. Kronmüller, *Philos. Mag. B* **48**, 127 (1983).

[12] J. Rivas, M.A. Lopez-Quintela, D. Martinez, F. Waltz, H. Kronmüller, *J. Non-Cryst. Solids* **131-133**, 1235 (1991).

[13] W. Ciurzyńska, G. Haneczok, J. Zbroszczyk, *J. Magn. Magn. Mater.* **189**, 384 (1998).

[14] K. Funke, *Berichte Bunsenges Phys. Chem.* **95**, 955 (1991).

[15] K.L. Ngai, A.K. Jonsher, G.T. White, *Nature* **277**, 185 (1979).