THE INFLUENCE OF HEAT TREATMENT ON MAGNETOSTRICTION OF Fe\textsubscript{74}Mo\textsubscript{8}Cu\textsubscript{1}B\textsubscript{17} METALLIC GLASS

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Abstract. Measurements of dilatation, magnetostriction, and electrical resistivity on rapidly quenched Fe\textsubscript{74}Mo\textsubscript{8}Cu\textsubscript{1}B\textsubscript{17} amorphous ribbons in as-quenched and annealed states was performed to determine changes of magnetic properties such as saturation magnetostriction and forced volume magnetostriction. The results have shown large changes of these quantities, which can be correlated with specific transformation processes.

1. INTRODUCTION

The structure of metallic glasses upon thermal treatment passes subsequently through different transformation processes such as amorphous phase relaxation, glass transition $T_g$, low temperature crystallization and transition through metastable crystalline structures in amorphous phase to stable crystalline structure. Ferromagnetic metallic glasses of the FeMoCuB type represent an important group of alloys where the presence of Mo and Cu atoms leads to drastic decrease of the Curie temperature $T_c$. Upon thermal treatment these alloys transform from the amorphous state into a state with very fine grained phase, nanocrystals, with grain sizes being about 15-20 nm. After this transformation, however, the material still contains a certain amount of the amorphous phase.

Magnetostriction is the deformation of a material related to its magnetization. From the phenomenological description of magnetoelastic coupling in isotropic magnetic medium we obtain [1,2]

$$\lambda_s(H) = \frac{\Delta l}{l} = \frac{1}{3} \omega(H) + \frac{2}{\lambda_v} \left(\frac{\cos^2 \Theta - 1}{3}\right) + \lambda_v^c,$$

where $\omega(H)$, $\lambda_v(H)$ and $\lambda_v^c$ are the volume magnetostriction, saturation magnetostriction and the form effects (bipolar magnetostriction), respectively. The quantity $\Theta$ is the angle between the direction of the measurements of the strain and the direction of magnetization. Magnetostriction is one of the physical properties which influences in an important way the use of ferromagnetic materials. Its value depends on external conditions ($T$, $H$, stress, ... ) and on the intrinsic properties of the magnetic materials, e.g. chemical and topological short-range order, CSRO and TSRO, homogeneity, density, internal stresses and shape. The value of $\lambda_v^c$ is nearly zero for a thin film (ribbon) magnetized in its plane.
and will be maximal for thin film magnetized perpendicularly to its plane.

The quantities $\lambda_S(H)$ and $\omega(H)$ are determined experimentally by measuring successively the strains observed in magnetic field applied in parallel and perpendicular directions to the measured direction of length change, $\lambda_{\text{par}}(H)$ and $\lambda_{\text{perp}}(H)$, respectively; then relations $\lambda_S = 2/3 (\lambda_{\text{par}} - \lambda_{\text{perp}})$ or $\lambda_S(H) = 2/3 [\lambda_{\text{par}}(H) - \lambda_{\text{perp}}(H)]$ and for the volume magnetostriction $\omega(H)$ holds $\omega(H) = \Delta V/V = \lambda_{\text{par}}(H) + 2\lambda_{\text{perp}}(H)$. The volume magnetostriction is composed of three parts of different origin $\omega = \omega_m(H) + \omega_x + \omega_s(H)$,

where $\omega_m$ and $\omega_x$ depend only on the amount of true magnetization and on the shape of the sample, respectively. The value $\omega_s$ describes the dependence of the crystal volume on the crystallographic directions and in isotropic materials on magnetic anisotropy of the spontaneous magnetization. Spontaneous volume magnetostriction is determined from dilatation measurements. Due to transformation processes in metallic glasses (i.e. nanocrystal formation) the quantity $\omega(H)$ is not always exactly defined, thus the quantity $\partial \omega/\partial H$, denoted as isotropic forced magnetostriction, is preferred in practice.

2. EXPERIMENTAL

Master alloy Fe$_{74}$Mo$_8$Cu$_1$B$_{17}$ has been prepared from elements with 99.8% purity in vacuum induction furnace. Amorphous ribbons 11 mm wide and 21 microns thick have been prepared by planar flow casting. The chemical composition of the ribbon has been determined by emission spectroscopy with inductively coupled plasma.

Dilatometric measurements have been performed on as-quenched samples 11 mm wide and 30 mm long using a dilatometer especially designed for measuring the length changes on this type of materials [3]. The results from these measurements have been used to determine the spontaneous volume magnetostriction, the invar effect and the values of $T_c$ (Fig. 1).

Thermal treatment of the materials has been selected in such a way as to optimize the study of the transformation from metallic glass to nanocrystalline material. Firstly, the 11 mm wide ribbon has been cut into samples 5 mm wide and 60 mm long; the samples have then been annealed for 1 h at temperatures from 603 to 923K. Samples treated in this way, cut from the same position on the ribbon, have then been used for measurements of different physical properties.

The phase formation in the amorphous structure after annealing has been checked by trans-
mission electron microscopy and by electron diffraction and X-ray diffraction. The magnetostrictions \( \lambda_{\text{par}}(H) \) and \( \lambda_{\text{perp}}(H) \) have been measured directly using the three terminal capacitance method [4]. This method allows determining the change of dimensions of the order of 0.02 microns.

The as-quenched and annealed ribbons have been chemically etched to prepare circular samples with 6 mm diameter, the shape being selected in order to obtain the same demagnetization factor when determining \( \lambda_{\text{par}}(H) \) and \( \lambda_{\text{perp}}(H) \). During measurements the sample orientation versus applied magnetic field has been such that the sample axis (ribbon axis) coincided with the direction of magnetic field when measuring \( \lambda_{\text{par}} \) and has been perpendicular to it, yet in the sample plane, when measuring \( \lambda_{\text{perp}} \). For ferromagnetic alloys with \( \lambda_S > 0 \) the dependencies \( \lambda_{\text{par}} \) and \( \lambda_{\text{perp}} \) are similar to those in Fig. 2, measured on thermally treated samples in order to evaluate also other physical quantities.

For the calculation of \( \lambda_S \) we have used the values of \( \lambda_{\text{par}} \) and \( \lambda_{\text{perp}} \) obtained as the intersection of the tangent of the corresponding measured curve with the ordinate at \( H = 0 \). The complete curves \( \lambda(H) \) have been used to determine \( \omega(H) \) or \( \partial\omega/\partial H \) in such a way as to obtain the dependence of \( \omega(H) \) on \( H \); the value \( \partial\omega/\partial H \) has been calculated as the average value over the applied field \( H \) (Fig. 3).

### 3. RESULTS AND DISCUSSION

The value of \( T_c \) for the investigated alloys lies at 350K; the alloy exhibits the invar effect and the value of the spontaneous volume magnetostriction is rather low, \( \sim 1.6 \cdot 10^{-3} \) at 300K, see curves 2 and 3 in Fig. 1. Temperature dependencies of dilatation and electrical resistivity exhibit also a region of relaxation up to 700K. Above this temperature the dependencies reflect the formation of metastable crystalline phases, namely \( \alpha \)-Fe(Mo) and subsequently also boride phases, as seen on curve 1 in Fig. 1. Curve 2 corresponds to dilatation of a sample upon heating after being heated up as shown by curve 1, showing dilatation of a system consisting of \( \alpha \)-Fe(Mo) nanograins in amorphous remains with partial formation of borides. The position of \( T_c \) of the remaining amorphous matrix (after heating - curve 1) is indicated on curve 2 by the tangents to the dilatation curve 3 at \( \sim 420K \).

The field dependencies of magnetostrictions \( \lambda_{\text{par}}(H) \) (full symbols) and \( \lambda_{\text{perp}}(H) \) (open symbols) on samples annealed at the temperatures indicated for 1 hour.

### Table 1. The values of magnetostriction and forced volume magnetostriction after annealing.

<table>
<thead>
<tr>
<th>( T ) [K]</th>
<th>( \lambda_S \cdot 10^6 )</th>
<th>( \partial\omega/\partial H \cdot 10^{10} )</th>
</tr>
</thead>
<tbody>
<tr>
<td>603</td>
<td>1.93</td>
<td>0.920</td>
</tr>
<tr>
<td>683</td>
<td>1.66</td>
<td>1.47</td>
</tr>
<tr>
<td>723</td>
<td>1.50</td>
<td>1.35</td>
</tr>
<tr>
<td>783</td>
<td>2.60</td>
<td>2.17</td>
</tr>
<tr>
<td>843</td>
<td>7.14</td>
<td>0.953</td>
</tr>
<tr>
<td>873</td>
<td>9.66</td>
<td>-1.854</td>
</tr>
<tr>
<td>923</td>
<td>5.55</td>
<td>0.983</td>
</tr>
</tbody>
</table>
temperature $T_{\text{meas}}$ (room temperature). Different cases may occur:

1. $T_c \gg T_{\text{meas}}$. In this case practically all atoms form dipoles and are arranged in domains, thus contributing to the values of $\lambda_s$ and $\lambda_s(H)$ and $\lambda_{\text{par}}$ may be $\lambda_s > 0$, $\lambda_s < 0$ or $\lambda_s \sim 0$, depending on the type of atoms in the alloy.

2. $T_c << T_{\text{meas}}$. In this case the material is in paramagnetic state and no dipoles or domains are formed. Deformation due to $H$ is caused by individual atoms which are oriented according to their magnetic (para or dia) states, $\lambda_s = 0$ and the dependencies $\lambda_{\text{par}}(H)$ and $\lambda_{\text{perp}}(H)$ are practically linear all the way up to saturation.

3. $T_c \sim T_{\text{meas}}$. The magnetostriction dependencies are in this case a superposition of the first two cases.

The shape of magnetostriction dependencies is also influenced by the structural state of the material, being either amorphous isotropic, polycrystalline or crystalline. The dependencies $\lambda_{\text{par}}(H)$ and $\lambda_{\text{perp}}(H)$ are practically identical for samples in as-quenched state and annealed at 603, 683, and 723K; only the dependencies for 683K are shown in Fig. 2 as representative for this state. This is confirmed also by the values of saturation magnetostriction $\lambda_s$ shown in Fig. 3 and in Table 1. The curves correspond to the case 3 from above, i.e. being a superposition of cases 1 and 2.

Annealing at 783K already leads to increase of $\lambda_s$ as well as of $\partial \omega(H)/\partial H$ due to formation of nuclei of crystalline phases which leads to deformation and to changes in both CSRO and TSRO and to changes in the slope of the dilatation curves in Fig. 1. These changes point to the directional effect of the bonding of individual paramagnetic atoms.

Annealing at 843K leads to changes on the $\lambda_{\text{par}}(H)$ and $\lambda_{\text{perp}}(H)$ dependencies and to changes in $\lambda_s$ and $\partial \omega(H)/\partial H$, being influenced by the magnetostriction dependence of $\alpha$-Fe(Mo). At 873K the crystallization of $\alpha$-Fe(Mo) is practically completed, $\lambda_s$ and $\partial \omega(H)/\partial H$ attain their maximum and minimum value, respectively. Annealing at 923K leads to formation of additional crystalline phase (borides). The magnetostriction dependencies are again as in case 3, the value of $\lambda_s$ decreases and $\partial \omega(H)/\partial H$ increases, again indicating a change of internal stresses and TSRO and CSRO.

4. CONCLUSIONS

Changes of field dependencies of magnetostric-

\begin{figure}[h]
\centering
\includegraphics[width=0.8\textwidth]{fig3}
\caption{Dependence of saturation magnetostriction $\lambda_s$ and forced volume magnetostriction $\partial \omega(H)/\partial H$ on annealing at given temperatures for 1 hour in argon atmosphere.}
\end{figure}
The influence of heat treatment on magnetostriction of Fe$_{74}$Mo$_8$Cu$_1$B$_{17}$ metallic glass

The function $\partial \omega(H)/\partial H$ can be used to identify the processes taking part in the deformation of samples in external magnetic field. Magnetostrictions reflect the effects of annealing temperature leading to transformations from as-quenched amorphous state and allow to design heat treatment forming a material with magnetic properties suitable for technical applications.

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