Neutron irradiation effect on permeability and magnetoimpedance of amorphous and nanocrystalline magnetic materials

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(Received 20 October 2004; revised manuscript received 14 January 2005; published 28 April 2005)

In this paper we provide physical insights into the effect of neutron irradiation on permeability spectra and magnetoimpedance of amorphous and nanocrystalline alloys. Experimental results indicate that neutron irradiation increases the permeability of the amorphous alloy but decreases the permeability of the nanocrystalline alloy in a high frequency region ($f \gg 1$ MHz), while the opposite is found in a low frequency region ($f < 1$ MHz). A careful examination of sample temperature during neutron irradiation process excludes thermal annealing as a possible origin of the observed irradiation effect. The magnetic relaxations in the low and high frequency regions are ascribed to the irreversible domain wall motion and reversible rotational magnetization, respectively. The enhancement in the permeability of the amorphous alloy upon neutron irradiation leads to a parallel improvement of magnetoimpedance response of the material, which is of practical use for sensing applications.

DOI: 10.1103/PhysRevB.71.134423 PACS number(s): 75.50.Kj, 75.75.+a

Suitable thermomagnetic processing of a metastable amorphous structure leads to parallel evolution of magnetic properties resulting in optimization of their properties. Most conventional procedures to achieve such transformations are performed by annealing at furnace, or under the presence of magnetic fields. In the present work, we investigate a novel processing technique involving neutron irradiation. Despite a number of previous studies, the understanding of the neutron irradiation effect on magnetic properties of amorphous and crystalline magnetic alloys remains controversial in part due to the complex nature of the problem. For instance, an increase in the permeability was revealed by relative permeability measurements of an as-quenched amorphous alloy irradiated in a low-field region with a 2.25 MeV proton fluence, was ascribed to the reduction of internal stresses. Brown et al. reported that, for both amorphous and crystalline magnetic alloys, the permeability decreased with increasing neutron fluence as a consequence of the increase of point defects produced by the irradiation and the domain walls pinning at defect clusters created by collision cascade. More recently, however, Kim et al. reported that neutron irradiation decreased the permeability of a nanocrystalline alloy but had a negligible effect on the as-quenched amorphous alloy. It is assumed that this discrepancy might be due to the irradiation with different neutron fluence. Since the magnetization processes are influenced by external and internal stresses originating at defects, the contribution of domain wall motion and rotational magnetization processes to the permeability of neutron irradiated amorphous magnetic materials is probably related to the neutron fluence.

To obtain deeper physical insights into the irradiation effect in both amorphous and crystalline materials, we investigated the effect of neutron irradiation on the magnetic properties of amorphous and nanocrystalline alloys by means of complex permeability spectra, magneto-impedance and the dc magnetization process. Amorphous alloy Fe$_{73.5}$Cu$_1$Nb$_{13}$Si$_{13.5}$B$_9$ ribbons with a thickness of ~20 μm and a width of 5 mm were prepared by a rapid quenching technique in vacuum. A subsequent thermal annealing treatment was carried out in vacuum at a temperature of 823 K for 1 h to achieve a stable and homogeneous nanocrystalline state. Both the amorphous and the nanocrystallized samples were then irradiated for 72 h using a HANARO research reactor at the Korea Atomic Energy Research Institute. The fluxes of thermal ($n_{th}$) and fast ($n_f$) neutrons were $3.09 \times 10^{13} n_{th}$ cm$^{-2}$ s$^{-1}$ and $1.87 \times 10^{11} n_f$ cm$^{-2}$ s$^{-1}$, respectively. In order to assess the possible thermal annealing effect due to irradiation, some as-quenched amorphous samples were also annealed at 473 K (the maximum sample temperature measured during the neutron irradiation) for 72 h. Magnetoimpedance (MI) and complex permeability were measured using a HP4129A impedance analyzer in the frequency range of $f = 0.1$–10 MHz. Magnetization measurements were performed using a vibrating sample magnetometer (VSM).

First, real and imaginary parts of the permeability of an as-quenched amorphous sample (denoted as No. 1) were measured. As shown in Figs. 1(a) and 1(b), at a small exter-
As the external field exceeds 23.4 mOe, the permeability spectra show an increase in the low-frequency region. This indicates that a different relaxation process from that observed in high-frequency region has developed in the low-frequency region. Therefore, the complex permeability $\mu^*(f)$ can be expressed by the addition of two decomposed relaxations as follows:

$$\mu^*(f) = \mu'_*(f) - j\mu''(f) = \mu'_0(f) + \mu'_h(f) - j(\mu''_0(f) + \mu''_h(f))$$

$$= 1 + \frac{\mu_{lo}(h_0)}{1 + (f/f_{lo})^2} + \frac{\mu_{hi}(h_0)(f/f_{hi})^2}{1 + (f/f_{hi})^2} + \frac{\mu_{hi}(h_0)(f/f_{hi})^2}{1 + (f/f_{hi})^2},$$

where $\mu_{lo}$ and $f_{lo}$ are the permeability and the relaxation frequency in the low-frequency region, $\mu_{hi}$ and $f_{hi}$ are the permeability and the relaxation frequency in the high-frequency region, and $h_0$ is the amplitude of the ac field. In general, the reversible magnetization process is much faster than the domain wall motion process. The magnetization by domain wall motion shows a threshold field to be activated due to the domain wall pinning at defects. Therefore, the relaxations in low and high frequency regions can be ascribed to the irreversible domain wall motion and the reversible magnetization rotation, respectively.

Following the same experimental procedure, we measured the real and imaginary parts of the permeability of samples: (No. 2) the amorphous ribbon annealed at 473 K, (No. 3) the neutron irradiated amorphous ribbon, (No. 4) the nanocrystalline ribbon, i.e., after annealing the precursor amorphous ribbon at 823 K, and (No. 5) the neutron irradiated nanocrystalline ribbon. For comparison, we summarized, in Table I, the experimental results including the relaxation frequency, $f_{lo}$ and $f_{hi}$, the initial permeability, $\mu_{lo}$ and $\mu_{hi}$, together with the coercivity, $H_C$, obtained from hysteresis loop measurements and the maximum value of MI measured at 5 MHz.

Comparing sample No. 2 with the amorphous sample No. 1, it can be observed that, after annealing at 473 K, both $f_{lo}$ and $f_{hi}$ decrease slightly whereas $\mu_{lo}$ and $\mu_{hi}$ increase slightly. That slight increase in the permeability can be attributed to the partial relief of internal stress by annealing at 473 K, but this thermal annealing effect (473 K) is negligible when compared with the effect of neutron irradiation effect.

TABLE I. The relaxation frequency $f_{lo}$ and $f_{hi}$, the initial permeability, $\mu_{lo}$ and $\mu_{hi}$, the coercivity, $H_C$, and the maximum value of MI ratio, $[\Delta Z/Z]_{max}(\%)$, measured at 5 MHz.

<table>
<thead>
<tr>
<th>Sample</th>
<th>$f_{lo}$ (kHz)</th>
<th>$f_{hi}$ (MHz)</th>
<th>$\mu_{lo}$</th>
<th>$\mu_{hi}$</th>
<th>$H_C$ (Oe)</th>
<th>$[\Delta Z/Z]_{max}(%)$</th>
</tr>
</thead>
<tbody>
<tr>
<td>No. 1</td>
<td>83.5</td>
<td>7.2</td>
<td>2.2</td>
<td>4.5</td>
<td>0.14</td>
<td>13.1</td>
</tr>
<tr>
<td>No. 2</td>
<td>73.1</td>
<td>6.84</td>
<td>2.4</td>
<td>5.8</td>
<td>0.138</td>
<td>17</td>
</tr>
<tr>
<td>No. 3</td>
<td>115</td>
<td>3.6</td>
<td>0.7</td>
<td>8</td>
<td>0.112</td>
<td>24</td>
</tr>
<tr>
<td>No. 4</td>
<td>13.9</td>
<td>1.7</td>
<td>2</td>
<td>20</td>
<td>0.015</td>
<td>57.1</td>
</tr>
<tr>
<td>No. 5</td>
<td>12.1</td>
<td>2</td>
<td>0.45</td>
<td>15</td>
<td>0.095</td>
<td>33.5</td>
</tr>
</tbody>
</table>

*(No. 1) as-quenched alloy, (No. 2) alloy annealed at 473 K, (No. 3) neutron-irradiated amorphous alloy, (No. 4) alloy annealed at 823 K only, and (No. 5) the 823 K-annealed and neutron irradiated alloy.*
NEUTRON IRRADIATION EFFECT ON ...
FIG. 4. The measured magnetoimpedance ratio (ΔZ/|Z|) as a function of frequency. (No. 1) the as-quenched alloy, (No. 2) the alloy annealed at 473 K, (No. 3) the neutron irradiated amorphous alloy, (No. 4) the alloy annealed at 823 K, and (No. 5) the 823 K-annealed and neutron irradiated alloy.

sample after neutron irradiation, but saturation magnetization was insensitive to the irradiation effect (see Fig. 3). No noticeable change of hysteresis loop was found in the sample annealed at 473 K for 72 h (sample No. 2), indicating that this annealing condition did not significantly affect the magnetic softness, and that the change in the M-H curve of sample No. 3 (Fig. 3) was caused solely by neutron irradiation. These results indicate that such an improvement in the magnetic softness of the as-quenched amorphous alloy subjected to neutron irradiation is likely due to the enhancement of rotational magnetization. In connection with the permeability data, we can attribute the increased permeability in the as-quenched amorphous alloy upon neutron irradiation (sample No. 3) to the enhancement of rotational magnetization.

Finally, to complete the analysis, we measured the magnetoimpedance ratio (ΔZ/|Z|=[Z(H)−Z(Hmax)]/|Z(Hmax)|) as a function of the applied dc magnetic field (H_{max}=35 Oe) at different frequencies up to f=10 MHz for all the samples investigated. Figure 4 shows the frequency dependence of the maximum value of MI ratio (i.e., [ΔZ/|Z|]_{max}) for the samples. It can be seen that [ΔZ/|Z|]_{max} starts to increase with increasing frequency up to 5 MHz and then decreases at higher frequencies.

This feature can be explained by considering the model of skin effects for thin ribbons.12 Within the framework of this model, at sufficiently high frequencies (the order of MHz), the cross section through which an ac current I=I_{0} exp(−jωt) flows is reduced due to the generation of eddy current and, consequently, the current flows through a thin sheath near the surface of the ribbon because of the skin effect. By solving the classical Maxwell equations of electrodynamics, the ac impedance Z=R+jωL (R and L are resistance and inductance, respectively) of the ribbon can be expressed in the form

\[ Z = R_{dc} \cdot j k a \coth(j k a), \]  

where 2a is the thickness of the ribbon, R_{dc} is the electrical resistance for a dc current, k=(1+j)/δ_{m} with imaginary unit j, δ_{m} is the penetration depth in a magnetic medium with the transverse permeability of μ_{T} and conductivity of σ and is expressed as

\[ \delta_{m} = \frac{c}{2 \pi \omega \mu_{T}}, \]  

where c is the speed of light and f=2πω is the frequency of the ac current.

At a given frequency, the application of a dc magnetic field (H_{dc}) changes the transverse permeability μ_{T} and hence the magnetic penetration depth δ_{m} that in turn alters the magnetoimpedance until the value of δ_{m} reaches the half thickness of the sample (a). At high frequency (δ_{m} ≪ a), Eq. (3) is reduced to the expression of Z ∝ (fμ_{T})^{1/2}. This means that, in this frequency region, the total impedance is proportional to the square root of the transverse permeability.12,13 Based on these analyses, it is pointed out that the GMI effect can be achieved when μ_{T} is large and δ_{m} and R_{dc} are small.

In this context, the GMI results can be interpreted by considering the change in δ_{m} in relation to the change of μ_{T} caused by the application of an external magnetic field according to Eq. (3). At frequencies below 1 MHz (a ≪ δ_{m}), the maximum value of GMI, [ΔZ/|Z|]_{max}, was relatively low due to the contribution of the induced magnetinductive voltage to magnetoimpedance. When 1 MHz ≲ f ≲ 5 MHz (a ≈ δ_{m}), the skin effect is dominant, a higher [ΔZ/|Z|]_{max} was found. Beyond f=5 MHz, [ΔZ/|Z|]_{max} decreases with increasing frequency (see Fig. 4). It is believed that, in this frequency region (f ≳ 5 MHz), the domain wall displacements were strongly damped owing to eddy currents, thus contributing less to the transverse permeability (μ_{T}), i.e., a small [ΔZ/|Z|]_{max}. Furthermore, the highest value of [ΔZ/|Z|]_{max} corresponds to the largest value of μ_{T} according to Eq. (3). Meanwhile, the transverse permeability (μ_{T}) is directly proportional to the longitudinal permeability due to rotational magnetization (μ_{hi}).14 Therefore, the largest value of [ΔZ/|Z|]_{max} corresponds to the largest value of μ_{hi}. As one can see from Table I, the nanocrystalline sample (No. 4) among the samples investigated has the largest value of μ_{hi} thus resulting in the largest value of [ΔZ/|Z|]_{max}. As compared with the as-quenched sample (No. 1), [ΔZ/|Z|]_{max} is considerably larger in the neutron-irradiated sample (No. 3) and is likely due to the higher value of μ_{hi} (see Table I). The decrease of μ_{hi} and the increase of H_{c} in the 823 K-annealed alloy after neutron irradiation (No. 5) lead to a considerable reduction in [ΔZ/|Z|]_{max}, as compared to the 823 K-annealed alloy (No. 4).

In the present work, it should be emphasized that amorphous alloys increase their permeability upon neutron irradiation, while nanocrystalline alloys decrease their permeability upon neutron irradiation. This fact has important consequences in the application of these materials as sensing
elements in a nuclear environment when the magnetoimpedance effect is used,\textsuperscript{13,15} because, when compared with the annealed amorphous alloy, the amorphous materials are less brittle and easier to handle which provides the necessary manufacturing flexibility and, more importantly, their magnetoimpedance properties can be enhanced by subsequent neutron irradiation.

The authors are grateful to Dr. B. G. Kim (HANARO center, Korea Atomic Energy Research Institute, Taejon 305-600, South Korea) for the neutron irradiation experiments. This work was partially supported by the Korea Science and Engineering Foundation through the Research Center for Advanced Magnetic Materials at Chungnam National University.

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