Influence of varying metal-to-glass ratio on GMI effect in Co$_{70.3}$Fe$_{3.7}$B$_{10}$Si$_{13}$Cr$_3$ amorphous glass-coated microwires

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**ABSTRACT**

The influence of a varying metal-to-glass ratio on the GMI effect in amorphous glass-coated Co$_{70.3}$Fe$_{3.7}$B$_{10}$Si$_{13}$Cr$_3$ microwires has been investigated. In the range of frequencies investigated (1–10 MHz), the magnitude of the GMI effect increases as the metal-to-glass ratio (h) increases from 4.11 to 9.29. The GMI curves for the h = 4.11 microwire exhibit a single-peak feature for f ≤ 1 MHz and a double-peak feature for f > 1 MHz, whereas a consistent double-peak feature is observed for microwires with h = 8.07, 8.72, and 9.29. The largest GMI effect is achieved for microwires with h = 9.29. The anisotropy field ($H_{K1}$), determined from GMI curves, increases with h = 4.11 to h = 8.07 and decreases when h > 8.07. The calculated radial stress decreases as h increases from 4.11 to 9.29. These results provide further insights into the correlation between the GMI effect and microwire dimensions towards the GMI optimization of amorphous glass-coated magnetic microwires for sensor applications.

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1. Introduction

Research in magnetic materials with the giant magnetoimpedance (GMI) effect for high-performance sensor applications is of much interest [1–4]. Here, the GMI effect refers to the strong variation in the high frequency impedance of a magnetic conductor with a small change in applied dc magnetic field ($H_{dc}$). In the case of a cylindrical magnetic conductor, the impedance, Z, can be calculated by [5]

$$Z = R_{dc}kJ_0(ka)/2J_1(ka),$$

where $J_0$ and $J_1$ are the Bessel functions, a is the radius of the wire, $R_{dc}$ is the dc electrical resistance and $k = (1+j)/\delta$ with imaginary unit j. $\delta$ is the penetration depth in a magnetic medium and is calculated by

$$\delta = \frac{c}{\sqrt{4\pi^2\sigma\mu_0}},$$

where $c$ is the speed of light, $\sigma$ the electrical conductivity, $\mu_0$ the circumferential permeability and $f = \omega/2\pi$ is the frequency of the ac ($H_{ac}$) flowing along the wire. According to Eqs. (1) and (2), GMI can be understood as a consequence of the increase in $\delta$ until it reaches $a$, through the decrease of $\mu_0$ under $H_{dc}$. In order to achieve a large GMI, it is necessary to reduce $\delta$ by choosing magnetic materials that have a large $\mu_0$ and a small $\rho$ ($\rho = 1/\sigma$) [1–3].

Among the existing GMI materials, the amorphous glass-coated magnetic microwires are considered attractive candidates for making miniature GMI-based magnetic sensors [6–10]. These microwires consist of the metallic magnetic core and insulating glass coat of the order of several microns [9]. The presence of an insulating glass layer helps (i) protect the metallic core from oxidation and (ii) give a higher degree of freedom for controlling the domain structure and hence magnetic properties of the microwires [6,9]. The domain structure of the glass-coated microwires is often determined by magnetoelastic anisotropy arising from the coupling between magnetostriction and internal stress frozen-in during the fabrication process [11]. Since the internal stress is mainly caused by the difference in the thermal expansion coefficients between the glass layer and metallic core, any variation in the metallic core diameter and/or glass thickness of the microwire may lead to a significant change in magnetoelastic energy ($K_{me}$), thus exerting a pronounced influence on the magnitude and sensitivity of GMI [12–15], expressed as:

$$K_{me} = \frac{3}{2\lambda_t}\sigma,$$

where $\lambda_t$ is the magnetostriction constant and $\sigma$ the internal stress. While previous studies focused mainly on the effect of either the...
metallic core diameter or the glass thickness on GMI [16–24], it is vital to investigate in detail the influence of varying metal-to-glass ratio on the GMI effect in these glass-coated microwires. The overall aim of the present study is to address this important issue.

2. Experimental details

Soft magnetic amorphous glass-coated microwires of Co70.3 Fe2.3 B2.7 Si13 Cr3 with different geometrical parameters have been fabricated by a modified Taylor–Ullovski method [25,26]. The metallic core diameter and the glass thickness of the microwires were measured by scanning electron microscopy (SEM). The geometrical parameters of the microwires (the metallic core diameter \(d\), the glass thickness \(t\), and the metal-to-glass ratio \((h = d/t)\)) are summarized in Table 1. These microwires possess good soft magnetic properties owing to the nearly zero and negative magnetostriction \((\lambda \sim -10^{-3})\). The magnetoimpedance was measured by a HP8753E network analyzer in the frequency range of 1–10 MHz. The microwire samples were set in the measuring cell subjected to dc magnetic fields up to 30 Oe along the microwire axis and vertical to the geomagnetic field. S11 port of the two-port network was applied to pick up the signal of output power and the S-parameter was then converted to the impedance by program calculation. The magnetoimpedance ratio \(\Delta Z/Z\) is defined as

\[
\frac{\Delta Z}{Z} (%) = \frac{Z(H) - Z(H_{\text{max}})}{Z(H_{\text{max}})} \times 100% ,
\]

where \(Z(H)\) and \(Z(H_{\text{max}})\) are the impedance values of the microwire in the measured external magnetic field and maximum magnetic field to saturate the impedance, respectively.

3. Results and discussion

Fig. 1 shows the magnetic field dependence of GMI (\(\Delta Z/Z\)) at representative frequencies of \(f = 1\) MHz, 4 MHz, and 10 MHz. It can be observed that the GMI curves show a single-peak feature for \(f \leq 1\) MHz and a double-peak feature for \(f > 1\) MHz for Sample No. 1 \((h = 4.11)\), whereas a double-peak feature is observed for Sample No. 2 \((h = 8.07)\), Sample No. 3 \((h = 8.72)\), and Sample No. 4 \((h = 9.29)\) over the frequency range of 1–10 MHz. The height and shape of GMI curves vary strongly with \(h\).

In Fig. 2 we plot the \(h\) dependence of maximum GMI ratio \((\Delta Z/Z)_{\text{max}}\) at \(f = 1\) MHz, 4 MHz, and 10 MHz. The corresponding \(d\) dependence of \((\Delta Z/Z)_{\text{max}}\) is also included in the inset of Fig. 2. It can be seen that the \((\Delta Z/Z)_{\text{max}}\) increases as the metal-to-glass ratio \((h)\) increases from \(h = 4.11\) to \(h = 9.29\). Remarkably, the \((\Delta Z/Z)_{\text{max}}\) starts to increase strongly for \(h \geq 8.07\) relative to the case of \(h = 4.11\). A similar trend is also found for the \(d\) dependence of \((\Delta Z/Z)_{\text{max}}\) (see inset of Fig. 2). Another noticeable feature is that the magnetic field corresponding to the double-peak position in GMI curves (defined as the anisotropy field, \(H_{\text{a}}\)) increases from \(h = 4.11\) to \(h = 8.07\) and then decreases for \(h > 8.07\), as shown in Fig. 3 and its inset. In addition, the magnitude of \(H_{\text{a}}\) increases with the measurement frequency.

Internal stress plays a crucial role in determining the domain structure and hence GMI behavior. We have calculated the internal stress in the radial direction using the following equation [27]:

\[
\sigma_r = \varepsilon E_g (1 - p^2) / [(k/4)(1 - p^2) + 4p^2/3] = (\alpha_m - \alpha_g)(T' - T),
\]

where \(\sigma_r\) is the residual radial stress, \(k\) is the metal-to-glass ratio of Young’s modulus, \(\alpha_m\) and \(\alpha_g\) are the metal and glass thermal expansion coefficients, respectively, \(T\) is the room temperature, \(T'\) is the glass solidification temperature. Using the values of \(T' = \)

\(823\) K, \(E_g = 64\) GPa, \(E_m = 110\) GPa, \(\alpha_m = 1.2 \times 10^{-5}\) K\(^{-1}\), and \(\alpha_g = 3.2 \times 10^{-6}\) K\(^{-1}\) in Ref. [28], we have calculated the radial stresses for Co70.3 Fe2.3 B2.7 Si13 Cr3 microwires with different \(h\) values. The calculated results are summarized in Table 1. As one

\begin{table}
\centering
\begin{tabular}{|c|c|c|c|}
\hline
Sample & \(h\) & \(d\) & \(t\) \\
\hline
No.1 & 4.11 & 14.8 & 0.3 \\
No.2 & 8.07 & 24.2 & 0.3 \\
No.3 & 8.72 & 26.6 & 0.3 \\
No.4 & 9.29 & 30.2 & 0.3 \\
\hline
\end{tabular}
\end{table}
Fig. 4a shows that for glass-coated microwires with negative magnetostriction, the single-peak behavior in the GMI profile is caused by the axial anisotropy of the central part of the metallic core, whereas the double peak behavior is due to the circumferential anisotropy of the outer shell [21,22]. This leads to a general expectation that these microwires should show a single-peak feature at a low frequency but a double-peak feature at a high frequency. This consistently explains why for the case of Sample No. 1 (h = 4.11), the GMI curve exhibits a single-peak feature at f = 1 MHz and a double-peak feature at f = 4 MHz. However, for Samples No. 2–No. 4 the strong increase of h is the result of the increased metallic core diameter (d) of the microwires (note that t is not much different in these microwires). In these cases, the increase of the metallic core diameter leads to the increase of the outer shell circular domain volume creating a circumferential anisotropy, thus resulting in a significant increase of the GMI effect with an observed double-peak structure [22]. The larger values of the GMI effect at high frequencies over 1 MHz (Fig. 2) arise from the fact that the skin effect becomes dominant and the magnetic penetration depth limits the flow of the ac driving current to the surface region of the microwire as the frequency is increased [3].

As reported previously by Chiriac et al. [21] for Co$_{68.15}$Fe$_{3.35}$Si$_{12.5}$B$_{15}$ glass-coated microwires having the same glass thickness (d = 5 µm, 10 µm, and 20 µm), the circumferential anisotropy constant first increased with an increasing d from 5 µm to 20 µm and then decreased for d > 20 µm. Also, the largest GMI effect was obtained for microwires with d = 20 µm (the...
largest circumferential anisotropy). In the present case, however, for Co$_{70.3}$Fe$_{23.7}$B$_{6.0}$Si$_{13}$Cr$_{3}$ microwires with $t \sim 3$ µm, the magnitude of the GMI effect increases continuously as the metallic core diameter ($d$) increases from 14.8 µm to 30.2 µm. The largest value of GMI is achieved for microwires with $d = 30.2$ µm. This difference suggests that the distribution of internal stresses in microwires with thinner glass thicknesses ($t \sim 3$ µm) could be rather different from that in microwires with thicker glass thicknesses ($t \geq 5$ µm) [12]. This points to an important fact that for $t < 5$ µm, glass thickness has a significant influence on the magnitude of the GMI effect of amorphous glass-coated microwires. This finding is of practical importance with respect to optimizing the GMI effect in amorphous glass-coated microwires for high-performance sensor applications.

For Co$_{70.3}$Fe$_{23.7}$B$_{6.0}$Si$_{13}$Cr$_{3}$ microwires with $t \sim 3$ µm, the increase of the metallic core diameter results in a significant reduction in the radial stress. This can be attributed to the fact that the same glass coating layer has a larger effect on a thinner metallic core and the sample with a smaller total diameter undergoes rapid solidification at a higher cooling rate [22]. This can also be related to the decrease of $H_k$ and the increase of the GMI effect, when the metallic core diameter increases from $d = 24.2$ µm to 30.2 µm, as shown in Fig. 3. This result is consistent with that reported in the previous study by Zhukova and coworkers [16–18,24]. The increase of $H_k$ with frequency is in agreement with the theoretical calculation that the radial distribution of magnetoelastic anisotropy increases in the outer shell surface region [21,22]. The increase of the GMI effect is also consistent with the ratio $\delta / \alpha$ decreasing for $d \geq 24.2$ µm. Although the variation of the radial internal stress in the present microwires arises mainly from the difference in the thermal expansion coefficient between the metallic nucleus and the insulating glass cover, one should also consider the effect of inhomogeneity of internal stress that could modify the $\lambda$ and hence $\mu_0$ of the microwires, according to $\lambda = \lambda_0 + \sigma \lambda_1$, where $\lambda_0$ is the saturation magnetization at zero tensile stress. $\sigma$ is a coefficient susceptible to thermal treatments. This effect may also account for the complex dependence of $H_k(h)$. A detailed study of the effect of inhomogeneity of internal stress is beyond the scope of this paper, but it would be an interesting subject for the future work.

4. Conclusions

The influence of varying metal-to-glass ratio on the GMI effect in amorphous glass-coated Co$_{70.3}$Fe$_{23.7}$B$_{6.0}$Si$_{13}$Cr$_{3}$ microwires in the frequency range of 1–10 MHz has been investigated. The magnitude and shape of GMI curves vary strongly with the metal-to-glass ratio. For microwires with the glass thickness varying between 3 µm and 4 µm, the magnitude of the GMI effect increases continuously with increasing the metallic core diameter from 14.8 µm to 30.2 µm. The calculated radial stress decreases as the metallic core diameter increases from 14.8 µm to 30.2 µm.

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