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Domain Transformation and MI of Melt-extracted $Co_{68.15}Fe_{4.35}Si_{12.25}B_{13.25}Nb_1Cu_1$ Microwires by Cryogenic Joule Annealing

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Giant magneto-impedance (GMI) effect and domain transformation for melt-extracted $Co_{68.15}Fe_{4.35}Si_{12.25}B_{13.25}Nb_1Cu_1$ amorphous wires have been induced by a cryogenic Joule annealing (CJA) treatment with different DC current amplitude (0~350 mA) for 240s. Experimental results indicate that the maximum GMI ratio ($[\Delta Z/Z_0]_{max}$) achieves to 188.1% with exciting field increasing to 1.8 Oe monotonically for 300 mA annealing treated wires, which can attribute to the surface complex domain structure change formed by CJA treatment. The liquid nitrogen can protect the wire from crystallization when applied large DC currents. Moreover, the CJA treatment can improve the response sensitivities effectively. These remarkable characteristics make the melt-extracted microwires by CJA tailoring as the promising candidate material for small-sized magnetic sensors.

Keywords: *melt-extracted microwires, cryogenic Joule annealing, giant magneto-impedance, domain transformation*

1. Introduction

The ferromagnetic microwires have been recognized as promising materials for small-size magnetic sensors in recent years. The giant magneto-impedance (GMI) effect of these wires which used for weak-field detection and high-resolution sensors has been attracted much attentions[1-3]. Many previous works focused on annealing treatment for improving the GMI ratio and field response sensitivity of the microwires simultaneously by releasing the residual inner stress of an as-quenched microwire which enhances the circumferential permeability μ_φ, such as joule heat-treatment and stress annealing[4-8]. Based on the definition of GMI ratio, $\Delta Z/Z_0(\%)=((Z(H_{ex})-Z(H_0))/Z(H_0)\times100\%$, reducing Z_0 is an immediate method to enhance the GMI ratio through annealing treatment while increase the Z_{max} or keep it unchanged. The complex impedance Z can be expressed as $Z(H_0)=R(H_0)+iX(H_0)$, where $R(H_0)$ and $X(H_0)$ are the resistance and the reactance at zero external field, respectively. In this case, the reducing $R(H_0)$ and $X(H_0)$ simultaneously is favor of obtaining high GMI performance.

Another important factor for the wires used as practical GMI magnetic sensors is the field response range. In fact, the enhancement of GMI for the microwire is generally at the expense of enhancing its response field range thus reducing its field response sensitivity (ξ). The conflict limits the applications of GMI microwires on miniature sensors to some extent. Herein, for resolving this issue, i.e, obtaining high GMI ratio with broad response field concurrently, new tailoring techniques is necessary.

The origin of large GMI ratio is ascribed to the skin effect with small penetration depth δ, small R_{dc} and large circumferential permeability μ_φ, which is closely related to the microwires with fine surface and roundness[1]. It is well known that the melt-extraction technique with high cooling rate and process conveniently controlled (i.e. linear velocity of wheel, feed rate of the molten) can produce micorowires of uniform diameter and roundness which has excellent GMI microwires[9,10]. In our present work, compared with conventional Joule annealing (JA), we have induced a cryogenic Joule annealing (CJA) under larger amplitude current across the melt-extracted $Co_{68.15}Fe_{4.35}Si_{12.25}B_{13.25}Nb_1Cu_1$ amorphous microwire for effectively improving its anisotropy field and keep the regular outer-shell amorphous structure with characterizing the transformation of static domain structure (SDS) under large DC currents[11].

2. Experiments

We have fabricated soft magnetic amorphous microwires of $Co_{68.15}Fe_{4.35}Si_{12.25}B_{13.25}Nb_1Cu_1$ with diameter of 35 μm by a modified melt-extracted technique and employed by CJA technique[9]. Impedance measurements are carried out at the intermediate frequency range of 100 kHz~15 MHz using Agilent 4294A precision impedance analyzer. The samples with 18 mm length were subject to current annealing with

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current rang of 0~350 mA for 240 seconds and their MI properties were measured by 20 mA driving amplitude with an external magnetic field applied along the axial direction of the microwire. The GMI ratio of the wire is defined as: $\Delta Z/Z_0=((Z(H)-Z(H_0))/Z(H_0)\times 100\%$, and the field response sensitivity is defined as: $\xi=\Delta(\Delta Z/Z_0)/\Delta H_{ex}\times 100\%$. The observation of domain structures of the microwires was performed by a Nanoscope III multimode atomic force microscope from Digital Instruments. A Veeco micro-etched silicon probe tip is used to collect related information by applying a combination of tapping and lift mode. During lift mode, the magnetic data were collected and displayed. Selected lift height range is 100 nm. Liquid nitrogen was adopted to hinder the evolution of circumferential domain and protect the microwires against crystallization.

3. Results and Discussion

Figure 1 displays the CJA device schematically. This method is designed to cool the surface of microwire by immersing into liquid nitrogen medium and to prevent the microwire from oxidation or over-heating with applying the magnetic and stress anisotropy to alter intrinsic anisotropy field. In the CJA process, the microwire is surrounded by nitrogen gas. The excessively large Joule heat is conducted out by the nitrogen gas shell with low temperature role rendering the microwire crystallization process which obviously damages the GMI properties.

The fields dependence of GMI ratios ($\Delta Z/Z_0$) for CJA-ed microwires with selected annealing currents in range of 0~350 mA and frequencies of 100 KHz ~ 10 MHz are illustrated in Figure 2. For the as-cast state in Figure 2a, all the $\Delta Z/Z_0$ curves without any rising peak decrease when the applied fields increase at all frequencies, indicating small circumferential anisotropy. After 200 mA current annealing, the rising peaks of GMI curves appear and the positions of the peaks monotonically increase with the frequencies increase ranging from 10 KHz to 20 MHz. At 10 MHz, the linear response field H_p is 0.8 Oe with the $(\Delta Z/Z_0)_{max}$ of 76.9% and the field response sensitivity ξ is 104.5%/Oe. In the case of 300 mA annealing, the $(\Delta Z/Z_0)_{max}$ reaches to 188.1% with H_p of 1.8 Oe and ξ of 104.5%/Oe for f=10 MHz. This peak position presents the anisotropy effective field which corresponding to the maximum of circumferential permeability μ_φ. In this case, the nearly linear response field is enhanced to 1.8 Oe with the improving of the response sensitivity which

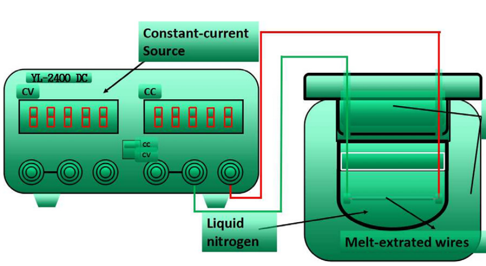

Figure 1. Schematic illustration of CJA process with insulation material and the microwire surrounding by nitrogen gap during annealing process.

is suitable for highly sensitive GMI sensors application. When annealing current was 350 mA, the $(\Delta Z/Z_0)_{max}$ reduces to 27.3% and the sensitivity ξ is only 13.7%/Oe in spite of the linear response field increasing to 2.0 Oe.

According to DC current through the microwire inducing the circumferential field instantaneously, the magnitude of magnetic field can be estimated by H_i=I/2πr with the strong skin effect. We obtained the induced magnetic field of the wires after 200 mA and 300 mA CJA which are $H_{i=200mA}\approx$22.9 Oe and $H_{i=300mA}\approx$34.3 Oe respectively. It can be clearly seen the induced circumferential field is larger than the corresponding field of the peak position for GMI profile H_p=0.8 Oe (200 mA CJA) and H_p=1.8 Oe (300 mA CJA). As well known, conventional Joule annealing can release the residual stress of microwires. It is also noted that the annealing current is selected to insure the temperature of the microwire between the T_c (Curie temperature) and the T_{cry} (Crystallization temperature) which avoiding the crystallization. It can be known from the induced M-H curves on the similar composition of Co-based microwire that the induced field H_i generated by large current can counteract the easy magnetization field and motivate the domain movement along the circular direction[12]. Meanwhile, the Joule heat generated by CJA is enough to meet the atoms redistribution or crystallization[13]. However, the cool role of the liquid nitrogen around the microwire protects the surface amorphous microstructure against crystallization or oxidation during CJA process[11]. Moreover, it restrains the formation of surface domain rotation. On another aspect, large current annealing also changes the microwire resistivity ρ and improves the circumferential permeability μ_φ. The skin depth is expressed as $\delta_s=(\rho/\pi f\mu_\varphi)^{1/2}$, δ_s increases with circumferential permeability μ_φ decrease and resistivity ρ increase.

Figure 3a-c depict the zero-field frequency dependence of the resistance $R(H_0)$, inductive reactance $X(H_0)$ and impedance $Z(H_0)$ with different current amplitude of 0~350 mA. Obvious monotonic increasing trends in the resistance $R(H_0)$ (Figure 3a), inductive reactance $X(H_0)$ (Figure 3b), and impedance $Z(H_0)$ (Figure 3c) are observed with the frequency range of 100 kHz ~ 15 MHz. The values of R and Z show the trends of firstly increasing less than 100 mA CJA compared with that of as-cast wires, then decreasing with the currents increase to minimum values at 300 mA CJA. Compared with $R(H_0)$ and $Z(H_0)$, the values of $X(H_0)$ with current amplitude larger than 200 mA are much smaller than those with corresponding values in current amplitude larger than 100 mA at high frequency (f>1 MHz). It illustrates that the DS has changed a lot when the annealing current amplitude larger than 200 mA. Figure 3d-f display the fields dependence of the magneto-resistance $\Delta R/R$ (%) (d), magneto-inductive reactance $\Delta X/X$ (%) (e) and magneto-impedance $\Delta Z/Z$ (%) (f) ratios at f=10 MHz for different annealing currents. The insets give the magnified plots of curves at the peak positions. After 200 mA CJA-ed microwire, the magneto-resistance ratio $\Delta R/R$ (%) and magneto-inductive reactance $\Delta X/X$ (%) reach to maximums of 76.4% and 88.9% at H_{ex}=1.0 Oe, respectively. The magneto-impedance $\Delta Z/Z_0$ (%) is up to 76.9% at H_{ex}=0.8 Oe. After 300 mA CJA, the ratio of $(\Delta R/R)_{max}$ (%) is 232.7% at H_{ex}=1.4 Oe, the $(\Delta X/X)_{max}$ (%)

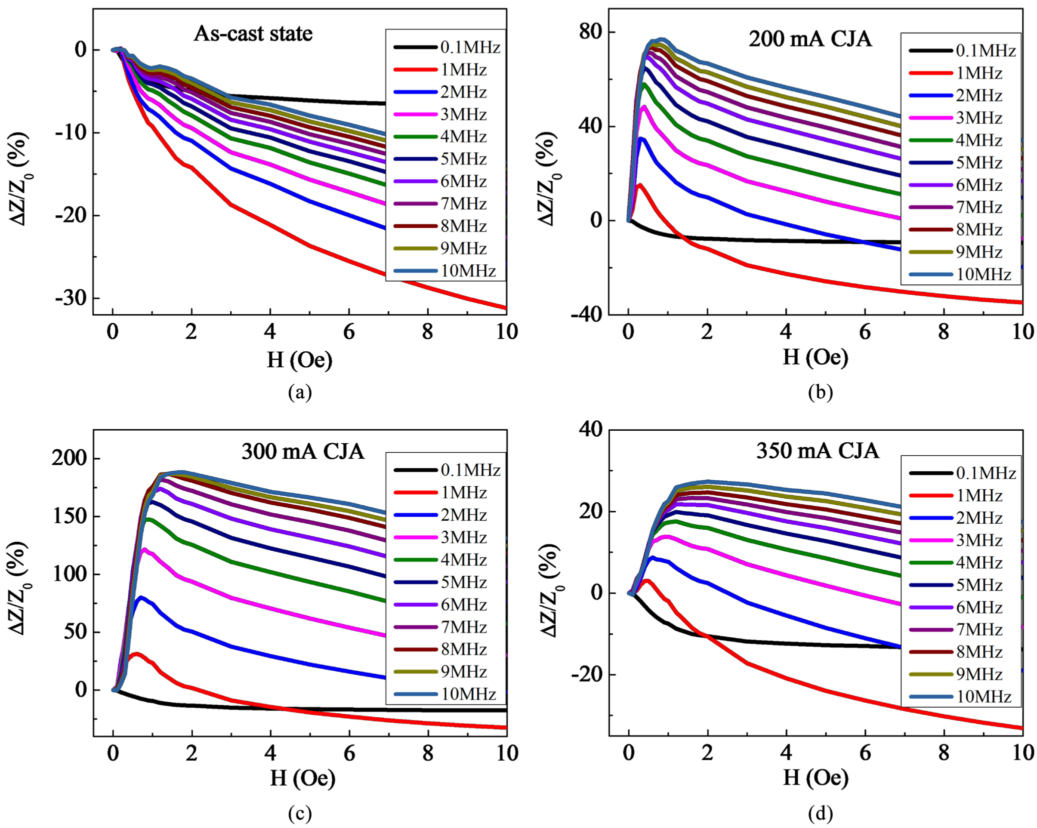

Figure 2. Field dependence of GMI curves measured with selected CJA-ed sample for t=240 s. (a) As-cast; (b) 200 mA; (c) 300 mA; (d) 350 mA.

is 86% at H_{ex}=3 Oe and the $(\Delta Z/Z_0)_{max}$ (%) is 188.1% at the H_{ex}=1.8 Oe. After 350 mA CJA, the $(\Delta R/R)_{max}$ (%) decreases to 24.9% at H_{ex}=1.4 Oe, the $(\Delta X/X)_{max}$ (%) decreases to 40.9% at H_{ex}=5 Oe and the $(\Delta Z/Z)_{max}$ (%) decreases to 27.3% at H_{ex}=2 Oe due to the over Joule heated with large current.

The maximum value of $\Delta X/X$ (%) obtained after 200 mA CJA. The reason for this behavior can be inferred for the anisotropy field induced by the couple effect between the large radial stress for the large temperature gradient with the circumferential field and the inner axial field. These two factors play the main role for the physical properties of the microwires surface. The result of the coupling effect leads to the unique surface domain structure of the microwire. The phenomenon that the response of the $\Delta X/X$ (%) obviously hysteretic to the $\Delta Z/Z$ (%) nearly 1.2 Oe is resulting from the crystallization of inner part due to the over Joule heat under large current while the outer-shell part keeping amorphous microstructure owing to the protection of liquid nitrogen[11]. In this case, the induced circumferential fields observed for the $\Delta X/X$ (%) curves after 300 mA CJA appear a large platform without sharp peak which corresponding to the response field H_{ex}.

In fact, the impedance test is a dynamic magnetization process, during which the domain walls movement, magnetization rotation and the magnetic permeability μ changing alter the MI ratios. For the microwires with different preparation methods, the difference of domain structure causes the disparity of response sensitivity to the external field. In order to improve the circumferential permeability and change the magnetization process thus improving the MI effect, it is necessary to carry out annealing modulation for the microwire[13]. Figure 4 exhibits the microwires DSs treated by different currents in CJA. For as-cast state, the weak circumferential domain with uneven width and indistinct wall due to the residual stress generated during preparation process is displayed in Figure 4a. The average width of weak circumferential domains is about 1.1 μm. The DS for 100 mA CJA-ed microwire is still an unclear circumferential domain with width of 0.9 μm. Most areas of the surface microwire are filled by unclear maze domains marked by white square zones. This DS almost exists in every CJA-ed microwire surface. After 200 mA CJA in Figure 4c, the width of local domain is about 1.71 μm with only a small part on the microwire surface. The majority of the surface domains are the maze domains. Notably, the circumferential domain is improved obviously for a large proportion of the microwire surface with clear domain wall and width of 0.94 μm for 300 mA CJA wire as seen in Figure 4d. This state corresponds to the large GMI ratio with relatively higher circumferential permeability μ_φ. For larger current of 350 mA CJA in Figure 4e, only a small proportion shows circumferential domain width is about 0.94 μm and most areas are the maze domains. In this case, the maze domain distribution may be resulted from the competitive

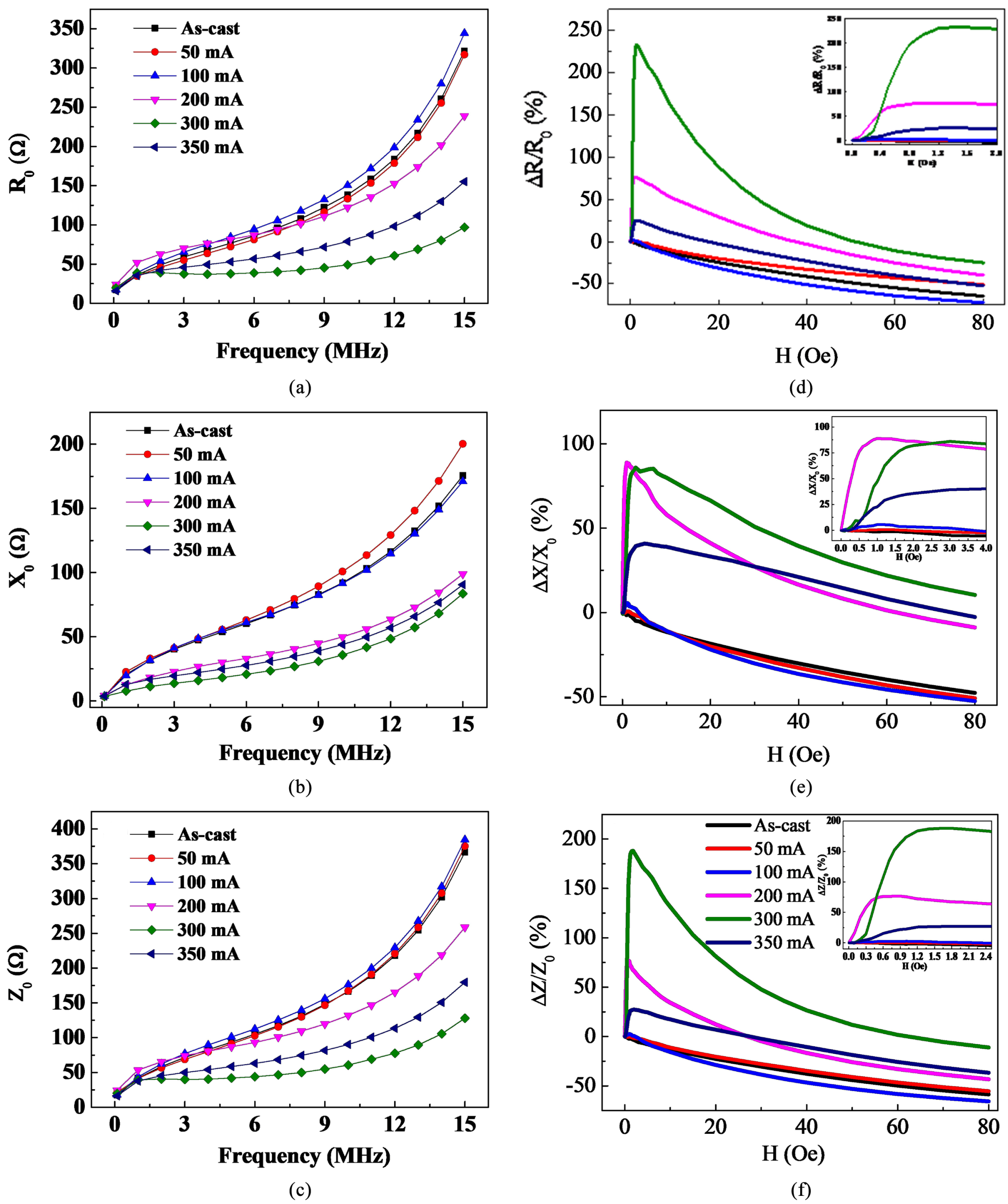

Figure 3. Frequency dependence of (a) resistance $R(H_0)$, (b) inductive reactance $X(H_0)$ and (c) impedance $Z(H_0)$ and field dependence of (d) magneto-resistance ($\Delta R/R_0$), (e) magneto-reactance ($\Delta X/X_0$), and (f) magneto-impedance ($\Delta Z/Z_0$) ratios with annealing current in the range of 0~350 mA. Insets of the magnified peak positions of $\Delta R/R_0$, $\Delta X/X_0$ and $\Delta Z/Z_0$ in (d), (e) and (f) respectively.

effect between the atoms severe thermal motion aroused by the large Joule heat and the forming of domain ordering distribution induced by the low temperature cooling around the microwire. The expansion of maze domain distribution represents the increasing of the leakage magnetic energy.

Based on the dynamic domain model[14] $\mu_{dc}=\mu_0^2M_s^2/dq$, the permeability $\mu_{dc}\propto d^{-1}$ (d is the width of dynamic domain and q is the pinning force coefficient), the growth of dynamic domain width and the domain wall pinning both are bad to obtain high magnetic permeability and GMI effect. Therefore, it is very important to select a suitable parameter for obtaining an ideal domain structure. For as-cast state in Figure 4a, due to the residual stress during the microwire fabrication process, the circumferential domain is non-uniform and the maze domains cover the whole microwire surface. Furthermore, the circumferential anisotropy is weak with low permeability. With the annealing current amplitude increasing in Figure 4b, the inner stress is released at a certain extent. However, the radial stress regenerate is adverse to the domain wall movement and moment rotating for the low temperature cooling of liquid nitrogen surrounding. The circumferential field is improved by large current which

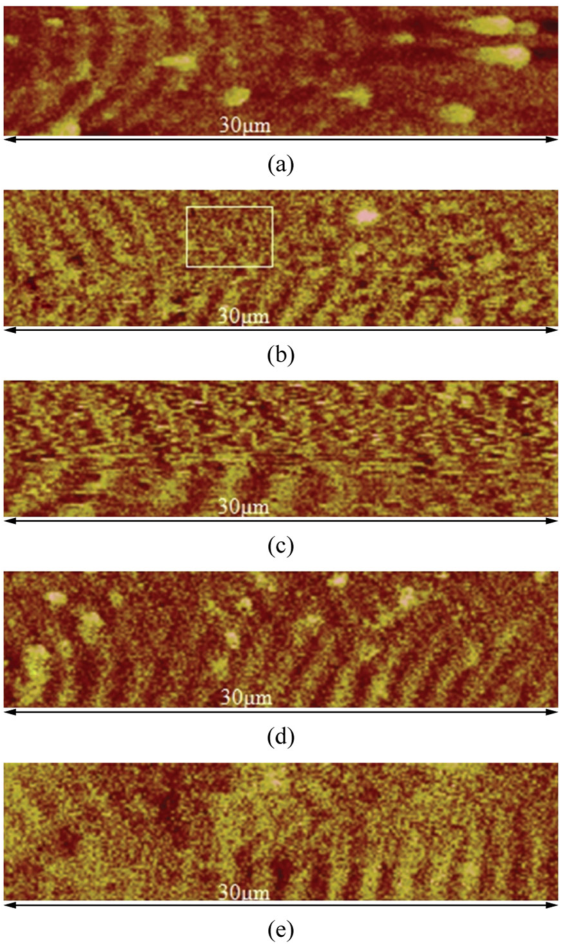

Figure 4. Magnetic domain structures of melt-extracted Co-based amorphous wires: (a) as-cast state; (b) 100 mA; (c) 200 mA; (d) 300 mA; (e) 350 mA; Ruler point is axial direction of microwire.

resulting in higher GMI ratio and response sensitivity (seen in Figure 4d). Combined with the large response field range of 1.8 Oe, the microwire during the CJA can be considered as promising miniaturized and high sensitive GMI sensors.

4. Conclusion

In conclusion, we have conducted a systematic investigation of the effect of CJA on melt-extrated $Co_{68.15}Fe_{4.35}Si_{12.25}B_{13.25}Nb_1Cu_1$ microwires. We obtained the direct evidence that the enhanced GMI effect under large current annealing result from the improving the circumferential domains through the surrounding nitrogen gap for cooling and coupling effect between the radial stress field and induced circumferential field, respectively. After 300 mA CJA, the maximum $\Delta Z/Z$ (%) achieves to 188.1% with field sensitivity ξ of 104.5 (%/Oe). Moreover, the linear response range expands to 1.8 Oe. Based on the cryogenic effect, large annealing current amplitude to 350 mA is achieved for the GMI profile. Therefore, an effective method to modulate the GMI effect towards a wider response field range for sensor applications is obtained.

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