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Enhanced magnetoimpedance effect in Co-based amorphous ribbons coated with carbon nanotubes

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We report upon the enhancement of the giant magnetoimpedance (GMI) effect in Co-based amorphous ribbons coated with non-magnetic carbon nanotubes (CNTs). In our study, the CNTs were drop-casted onto the surface of a Metglas[®] 2714 A ribbon with three different concentrations (5, 10, and 15 μ L of CNTs). Relative to the plain ribbon, a 15% enhancement of the GMI effect was observed in the ribbon coated with 10 μ L of CNTs. The GMI effect first increased with the CNT concentration, to a maximum of 10 μ L of CNTs, and then decreased at higher concentrations. Noticeably, at a measured frequency of 10 MHz, the magnetic field-induced ac resistance change was about 35% larger for the ribbon coated with 10 μ L of CNTs than for the plain ribbon. These observations may reveal a new perspective for developing CNT-based gas sensors that operate using the principle of the GMI effect. © 2012 American Institute of Physics. [doi:10.1063/1.3676214]

The giant magnetoimpedance (GMI) effect, which refers to a large change in the ac impedance of a ferromagnetic conductor subject to a dc magnetic field, observed in a number of soft ferromagnetic ribbons,^{1,2} holds great promise in magnetic field sensing. The ac impedance, Z = R + jX (*R* and *X* are resistance and reactance, respectively), of a ferromagnetic ribbon can be calculated by,¹

$$Z = R_{dc} \cdot jka \coth(jka), \tag{1}$$

where *a* is half of the thickness of the ribbon, R_{dc} is the electrical resistance for a direct current, *j* is the imaginary unit, and $k = (1 + j)/\delta_m$. The impedance is related to the skin effect, characterized by the skin depth (δ_m) , which is given by,

$$\delta_m = \sqrt{\frac{\rho}{\pi \mu_T f}},\tag{2}$$

where ρ is the electrical resistivity, $\mu_{\rm T}$ is the transverse magnetic permeability, and *f* is the frequency of the alternating current. The application of a dc magnetic field ($H_{\rm dc}$) changes μ_T and δ_m , which alters *Z*, until δ_m reaches *a*. Since the GMI is often observed at high frequencies (f > 1 MHz), the skin effect is significant enough to confine the alternating current to a sheath close to the surface of the ribbon.¹ As a result, small variations in the magnetic signal near or on the surface of the ribbon can alter the GMI of that ribbon. Small variations could come from stray fields from surface irregularities^{3,4} or magnetic nanoparticles.⁵

In this study, we show that the presence of carbon nanotubes (CNTs) on the surface of a Co-based amorphous ribbon enhances the GMI effect of the ribbon. This observation is of potential interest in developing a new class of CNT-based gas sensors operating on the principle of the GMI effect. A change in the electrical resistance of the CNTs, when exposed to gases such as NO₂, NH₃, H₂O, CO, iodine, and ethanol, may alter the GMI of the ribbon.

Carbon nanotubes were grown in commercial porous alumina templates using a chemical vapor deposition (CVD) method without the use of metal catalysts.⁶ The alumina templates were etched away, leaving free standing CNTs. The resulting CNTs were between 250 and 300 nm in diameter, on average (see the inset of Fig. 1). The CNTs were then drop-casted onto a Metglas® 2714 A ribbon with the composition, Co₆₅Fe₄Ni₂Si₁₅B₁₄, before measuring the GMI. X-ray diffraction confirmed the amorphous nature of the ribbon. Three different concentrations (5, 10, and 15 μ L of CNTs) were used and compared to a plain ribbon with no CNTs. Magnetic measurements of CNTs were performed at room temperature using a vibrating sample magnetometer. Magnetoimpedance measurements in applied dc magnetic fields up to 120 Oe were carried out along the ribbon axis (1 cm long) over a frequency range of 0.1-10 MHz at a constant alternating current of 5 mA using an impedance analyzer (HP4192 A). The details of the measurement system have been reported elsewhere.⁷ The GMI ratio has been defined as,

$$\Delta Z/Z = 100\% \frac{Z(H) - Z(H_{\max})}{Z(H_{\max})},$$
(3)

where Z(H) and $Z(H_{max})$ represent the impedance in a magnetic field, H, and in the maximum field, H_{max} (120 Oe), respectively.

It has been reported that CNTs may behave ferromagnetically, resulting from the presence of magnetic impurities⁸ or due to sample synthesis conditions.⁹ To examine this, we measured the magnetization versus the magnetic field (the M-H loop) at 300 K for the presently fabricated CNTs, the result of which is shown in Fig. 1. As one can clearly see

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FIG. 1. Magnetic hysteresis loop (M-H) taken at 300 K for the CNTs. The inset shows a TEM image of the CNTs.

in this figure, the CNTs show a paramagnetic character at room temperature, which agrees with the fact that these CNTs were synthesized by the CVD method without the use of metal catalysts.⁶ Keeping this in mind, we investigated the influence of the CNTs on the GMI of the ribbon.

Figure 2 shows a 3 D plot for the magnetic field and frequency dependence of the GMI ratio ($\Delta Z/Z$) for the ribbon sample coated with 10 μ L of CNTs. It is observed that $\Delta Z/Z$ has a maximum at 2 MHz and the GMI profiles show a double-peak feature associated with the presence of transverse anisotropy in the ribbon. A similar behavior was also observed for the remaining samples.

To assess the influence of the CNTs on the GMI of the ribbon, we display, in Fig. 3(a) the magnetic field dependence of the GMI ratio at a frequency of 2 MHz for the ribbon samples with and without CNTs. Figure 3(b) shows an enlarged portion of the GMI profile at low magnetic field ranges. It is very interesting to note that relative to the plain ribbon, the presence of the CNTs significantly increased the GMI ratio for the ribbon samples coated with CNTs. At f = 2 MHz, the $\Delta Z/Z$ increased from 34% for the plain ribbon to 40% for the ribbon coated with 10 μ L of CNTs. With increasing CNT concentration, the $\Delta Z/Z$ first increased, reached a maximum for the sample with 10 μ L of CNTs (15 μ L of CNTs).

This feature is seen more clearly in Fig. 4(a), which shows the frequency dependence of the maximum GMI ratio



FIG. 2. (Color online) Magnetic field and frequency dependences of the GMI ratio ($\Delta Z/Z$) for the ribbon coated with 10 μ L of CNTs.



FIG. 3. (Color online) Magnetic field dependence of the GMI ratio ($\Delta Z/Z$) measured at f=2 MHz for the plain ribbon and the ribbons coated with 5, 10, and 15 μ L of CNTs. (b) is a close-up.

 $([\Delta Z/Z]_{max})$ for all of these samples. As one can see in this figure, for all samples investigated, the $[\Delta Z/Z]_{max}$ first increased, reached a maximum at 2 MHz, and then decreased for higher frequencies. This frequency dependence of $[\Delta Z/Z]_{max}$ can be understood by considering the relative contributions to the impedance from transverse permeability resulting from the domain wall motion and spin rotation processes.² It is also noted that the increase of $[\Delta Z/Z]_{max}$ in the ribbon samples coated with CNTs was achieved only at f > 1 MHz, where the skin effect was strong and the surface effects were more important.

To understand this further, we considered the relative contributions to the impedance (*Z*) from the ac resistance (*R*) and reactance (*X*) components (note: Z = R + jX). The changes in *R* and *X* with H_{dc} were evaluated for all samples. We show the frequency dependence of the maximum field-induced resistance and reactance change ratios ($[\Delta R/R]_{max}$ and $[\Delta X/X]_{max}$, respectively) for the ribbon samples with and without CNTs in Figs. 4(b) and 4(c). Except for the case of the ribbon sample coated with 5 μ L of CNTs, the $[\Delta R/R]_{max}$ was found to be greater for the ribbon samples coated with the CNTs (10 and 15 μ L of CNTs) than for the plain ribbon.



FIG. 4. (Color online) Frequency dependence of the maximum field-induced impedance change $[\Delta Z/Z]_{max}$, maximum field-induced resistance change $[\Delta R/R]_{max}$, and maximum field-induced reactance change $[\Delta X/X]_{max}$ for the plain ribbon and the ribbons coated with 5, 10, and 15 μ L of CNTs.

The largest value of $[\Delta R/R]_{\text{max}}$ was achieved for the ribbon sample coated with 10 μ L of CNTs. At the highest measured frequency of 10 MHz, the $[\Delta R/R]_{\text{max}}$ of the ribbon coated with 10 μ L of CNTs was about 35% larger than that of the plain ribbon. We note that the difference in $[\Delta R/R]_{\text{max}}$ occurred at f > 2 MHz. However, the situation is different for the frequency dependence of $[\Delta X/X]_{\text{max}}$ (Fig. 4(c)). In the frequency range 0.1–10 MHz, the $[\Delta X/X]_{\text{max}}$ was larger in all ribbon samples coated with CNTs relative to the plain ribbon. Among these samples, the largest $[\Delta X/X]_{\text{max}}$ was achieved for the ribbon coated with 10 μ L of CNTs. This explains why the largest $[\Delta Z/Z]_{\text{max}}$ was achieved for this sample (Fig. 4(a)). Note that we performed similar experiments on different ribbon samples to confirm the reproducibility of our results. Our results seem to suggest that the presence of CNTs on the surface of the ribbon could reduce stray fields due to surface irregularities and close up the magnetic flux path, resulting in the enhancement of the GMI effect. Nevertheless, further study is needed to better understand the physical origin of the observed effect.

From a gas sensor application perspective, it has been shown that CNTs are ideal for gas molecule adsorption and storage, owing to their extremely high surface-to-volume ratio.¹⁰ The CNT-based gas sensors with high sensitivity and selectivity (where sensing is achieved by the dc resistance change upon adsorption of analytic molecules) are important for leakage detections of explosive gases such as hydrogen, and for real-time detections of toxic or pathogenic gases in various industries.¹¹ However, these resistive sensors possess limited sensitivities $(\Delta \rho / \rho \sim 2-10\%)$.^{10,11} Therefore, there is a need for developing alternative techniques, that allow detecting gases with a higher degree of sensitivity. While the origin of the enhanced GMI effect in the magnetic ribbons coated with CNTs remains to be investigated, our observations may provide a way for developing CNT-based gas sensors that operate based on the principle of the GMI effect. These sensors would detect the magnetoimpedance change of the sensing element (e.g., the magnetic ribbon) through variations in the electrical resistance of CNTs when exposed to gases such as NO2, NH3, H2O, CO, iodine, and ethanol. Future research should be aimed at testing the sensor's abilities for detecting and distinguishing different kinds of gases.

In summary, the influence of CNTs on the GMI effect of the Co-based amorphous ribbons was systematically investigated. It was shown that the presence of CNTs on the surface of the ribbon increased the GMI effect of that ribbon; the GMI effect varied with the CNT concentration. Since change in the electrical resistance of the CNTs subject to absorbed gases can alter the impedance of the ribbon, a new class of CNT-based gas sensors that operate based on the principle of the GMI effect might be developed.

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