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Long magnetic relaxation time of Fe–Bi spin-glass system

Jen-Hwa Hsu*, J.T. Lee, Ching-Ray Chang, M.T. Lin

Department of Physics, National Taiwan University, Taipei, Taiwan 106, ROC

Abstract

Bi and Fe are mutually insoluble either in the solid or liquid states. However, metastable Fe–Bi alloys can be made using the rapidly quenching method. We have successfully prepared metastable $\text{Fe}_x\text{Bi}_{1-x}$ alloys over a wide composition range by DC magnetron co-sputtering. The X-ray diffraction results indicate that the magnetic Fe atoms are gradually replacing the Bi atoms in a rhombohedral structure for $x < 0.6$. The magnetic measurements show that a spin-glass results with the freezing temperature increasing with x . In addition, a long relaxation characteristic time τ is observed which is about 20 h at room temperature. The slow relaxation behavior of the Fe–Bi system can be qualitatively explained from the picture that Bi has a large spin–orbit splitting. © 2001 Elsevier Science B.V. All rights reserved.

Keywords: Magnetic relaxation; Metastable; Spin-glass; Spin–orbit splitting

The magnetic relaxation has been extensively studied in the past both in theory as well as in experiment. The efforts of research are particularly devoted to the spin-glass [1,2] and fine particle systems [3]. The magnetic relaxation was first discussed in the framework of the Debye model. Then it was extended to a “stretched” exponential form [4]. More recently a so-called $T \ln(t/\tau_0)$ scaling was used in describing the magnetic relaxation in spin-glass systems with a distribution of energy barriers due to the frustration of the competing magnetic interactions among the individual spins [5]. However, the above theoretical explanations are just phenomenological approaches and none is able to fit experimental data over a large interval of time with a single characteristic time. On the other hand, the observable relaxation events in most spin-glass systems in general are very short and the phenomenon is notable only at low temperatures. Recently, we have discovered a very long relaxation time on the scale of more than 10,000 s at room temperature in Bi-based alloy systems. Bi is a semi-metal and possesses several unique physical properties. For example, the Fermi wavelength of the

semi-metallic Bi is as large as 400 Å, as compared to a few Å for most metals [6]. The carrier’s mean free path in Bi can be as much as a few millimeters at 4.2 K, several orders of magnitude larger than in most metals. In this work, we have investigated the magnetic relaxation phenomenon in the Fe–Bi alloy system and obtained an unusually large relaxation time.

The metastable $\text{Fe}_x\text{Bi}_{1-x}$ alloy films were prepared by DC magnetron sputtering with composite targets made from Fe and Bi powders. In this work, x denotes the relative volume concentration of Fe everywhere. Since the sputtering rate of Bi is about 20 times that of Fe and the melting point of Bi is low, a small DC current of 20 mA was adopted. The substrates of kapton foil were cooled by liquid LN_2 during sputtering. The thickness of the films is 1–2 μm .

The film structure and actual film composition were determined from X-ray powder diffraction method and by electron probe X-ray micro-analysis (EPMA), respectively. The differential thermal calorimeter (DSC) was used to investigate the thermal stability of as-made metastable alloys. The magnetic measurements were performed on a superconducting quantum interference device (SQUID) between 5 and 400 K.

From Fig. 1, it can be seen that for the films with $x < 0.6$, a single rhombohedral structure is present and the peak positions of diffraction lines are slightly different

* Corresponding author. Tel.: + 886-2-23661867; fax: 886-2-23639984.

E-mail address: jhhsu@phys.ntu.edu.tw (J.-H. Hsu).

Fig. 1. Representative X-ray diffraction patterns of as-made $\text{Fe}_x\text{Bi}_{1-x}$ alloys.

from those of pure Bi, shifting to larger angles as more Fe atoms substitute Bi. This is because the atomic radii are 1.2 and 1.7 Å for Fe and Bi, respectively. As x exceeds 0.6, all diffraction lines from rhombohedral Bi disappears, while the phase corresponding to BCC Fe appears. No amorphous phase has been identified as in Ref. [7]. The thermal analysis for metastable alloy films indicates that no other phase transformation has been observed except at the melting point of Bi which is 271°C. We can conclude that the phase segregation of Fe and Bi atoms should occur at temperatures higher than the melting temperature of Bi.

In Fig. 2(a), the zero-field-cooled (ZFC) and field-cooled (FC) magnetization curves of $\text{Fe}_{0.06}\text{Bi}_{0.94}$ are shown. In these experiments, a small field of 100 Oe was applied. The as-made $\text{Fe}_{0.1}\text{Bi}_{0.9}$ is undoubtedly identified as a spin glass with the freezing temperature (T_f) of ~30 K. The freezing temperature rises rapidly with increasing x and it is higher than 300 K for $\text{Fe}_{0.4}\text{Bi}_{0.6}$ as shown in Fig. 2(b). A drastic increase of magnetization at temperatures higher than 300 K is also observed, which is due to the slow magnetic relaxation. Fig. 3 depicts the magnetization process of $\text{Fe}_{0.4}\text{Bi}_{0.6}$ under 100 Oe at 300 K. The experimental results can be fitted with a “stretched” exponential form

$$m(t) = m_s - B \exp[-(t/\tau)^\beta] \quad (1)$$

with $\tau = 4.8$ h and $\beta = 0.43$. The value of τ increases to 5.4 h as the measuring temperature is reduced to 100 K. The value of β is essentially the same at different temperatures and close to 0.5 as predicted in Ref. [4]. The spin-glass system with such a large characteristic time is unusual at room temperature. We have observed a similar behavior in the Bi-Co system. We believe that this unusually large relaxation time is the result of the large spin-orbit coupling in the high-Z element Bi. This large

Fig. 2. The zero-field-cooled (ZFC) and field-cooled (FC) magnetization curves for (a) as-made $\text{Fe}_{0.1}\text{Bi}_{0.9}$, and (b) as-made $\text{Fe}_{0.4}\text{Bi}_{0.6}$. The applied external field is 100 Oe.

Fig. 3. The normalized magnetization as a function of time for as-made $\text{Fe}_{0.4}\text{Bi}_{0.6}$ at room temperature. The solid line is the fitting of Eq. (1) with $\tau = 4.8$ h and $\beta = 0.43$.

spin-orbit splitting of Bi affects the Fe electronic structure due to the hybridization effect in the Fe-Bi alloy. Therefore, the energy splittings in the Fe-Bi system are large and the localized Fe-spins, thus, cannot relax their energy to the system continuously. In other words, the discrete and large energy gap makes the relaxation unusually slow. For comparison, we have also studied Fe-Cu and Fe-Ag systems and did not see similar effects. These results support our picture described above. A further experimental investigation, which probes spin-orbit splitting directly using the photoemission technique, is in progress.

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