

Magnetic properties and magnetostriction of a binary Dy₅₀Co₅₀ amorphous alloy

L. Xia^{1,2}, K. C. Chan^{1,*}, D. Ding², L. Zhao¹ and B. Z. Tang²

¹ Department of Industrial and Systems Engineering, The Hong Kong Polytechnic University, Hung Hom, Hong Kong

² Institute of Materials, Shanghai University, Shanghai 200072, China

Abstract

The magnetic properties and magnetostriction of a binary Dy₅₀Co₅₀ amorphous alloy were studied in the present work. The Dy₅₀Co₅₀ amorphous ribbon shows spin glass like characteristics with a spin freezing temperature of about 80 K. The magnetostriction of the amorphous ribbon is reversible at temperatures above 60 K, where the amorphous ribbon is soft magnetic; but is irreversible at temperatures well below 60 K, where the ribbon is hard magnetic. The effect of the magnetic structure on the magnetic and magneto-strictive behavior, and the mechanism for the large magnetostriction of the amorphous alloy, were investigated.

Keywords: amorphous materials, spin glass, magnetic properties, magnetostriction

* Corresponding authors. Email: kc.chan@polyu.edu.hk (K. C. Chan)

Magnetostriction, which was discovered in iron by J. P. Joule in 1842, is an intrinsic property of ferromagnetic materials in which their shape or dimensions change under a magnetic field¹⁻². Since the discovery of giant magnetostriction in the TbDyFe alloy (known as Terfenol-D)³, magnetostriction materials have received increasing attention due to their promising applications as actuators, sensors and energy harvesting devices. However, the shortcomings of Terfenol-D, such as brittleness, low corrosion resistance and high energy loss, have limited its industrial applications. Therefore, there have been huge efforts to address the limitations of this alloy over the last few decades. As a result, high performance magnetostriction has been achieved in several materials, such as the Gafenal alloys and magnetostrictive polymer based composites⁴⁻⁵. Nonetheless, to develop a novel magnetostriction material with a combination of giant magnetostriction under low magnetic fields, good corrosion resistance, good mechanical properties, and low energy loss is still a challenge in this field.

Amorphous alloys have been shown to demonstrate very attractive properties that can match the above requirements. Unfortunately, the magnetostriction of amorphous alloys is generally small due to the microstructural randomness, which suppresses the magnetoelastic interactions in the amorphous material^{2, 6-7}. However, recent results reported by Speliotis *et al.* have demonstrated the excellent magnetostriction of amorphous TbDyFe thin films⁸⁻⁹. Nevertheless, these amorphous alloys can only be prepared in the shape of thin films, indicating that their glass forming ability (GFA) is not sufficiently high.

The study of the GFA and magnetic properties of binary Tb(Dy)-TM amorphous alloys can provide fertile ground for developing multicomponent magnetostriction materials, with a combination of good GFA and giant magnetostriction¹⁰. On the other hand, systematic investigation of the relationship between microstructure, composition and magnetostriction, based on these binary amorphous alloys with simple composition, can reveal the mechanisms for the high magnetostriction of amorphous alloys. In the present work, we studied the GFA, magnetic properties and magnetostriction of a binary Dy₅₀Co₅₀ amorphous alloy. The amorphous alloy was prepared in the shape of a ribbon

by the melt-spinning method. The magnetic structure and its effect on magnetostriction of the amorphous alloy were investigated.

An ingot, with nominal Dy₅₀Co₅₀ composition was prepared by arc-melting a mixture of Dy and Co elements, with purity of above 99.9% (at%). The metals are purchased from the Trillion Metals Co., Ltd. Dy₅₀Co₅₀ as-spun ribbon was prepared by melt-spinning using a single copper wheel, with a surface speed of 30 m/s, under a high-purified argon atmosphere. The approximate width of the as-spun ribbon was about 1 mm and the average thickness about 40 μm. The structure of the ribbon was characterized by X-ray diffraction (XRD) on a Rigaku D\max-2550 diffractometer using Cu K_α radiation. A Perkin-Elmer DIAMOND differential scanning calorimetry (DSC) was used to measure the thermal properties of the amorphous ribbon under a purified argon atmosphere at a heating rate of 20 K/min, and the magnetic properties of the Dy₅₀Co₅₀ amorphous ribbon were measured by a Quantum Design Physical Properties Measurement System (PPMS 6000). In order to minimize the demagnetization factor, an applied field is parallel to the longitudinal direction along the length (~ 8 mm) of the glued ribbons. The hysteresis loops were measured at 10 K, 30 K, 60 K, 80 K, 100 K and 300 K under a field of 5 T. With the use of a foil strain gauge (KYOWA; model KFL-02-120-C1), the magnetostriction (λ) of the Dy₅₀Co₅₀ amorphous ribbon was measured by PPMS at 30 K, 60 K, 70 K, 80 K and 90 K under a magnetic field of 5 T. The strain gauge, which was calibrated by pure aluminum, was placed perpendicular to the longitudinal direction along the length of the sample, and KYOWA PC-600 strain gauge cement was used to fix the gauge on the Dy₅₀Co₅₀ amorphous sample.

Figure 1 shows the XRD pattern of the Dy₅₀Co₅₀ as-spun ribbon, exhibits typical amorphous characteristics of broadened diffraction maxima. The amorphous feature of the Dy₅₀Co₅₀ as-spun ribbon can also be illustrated by the typical glass transition and crystallization behavior on the continuous DSC trace, as shown in the inset of Fig. 1. The onset temperature for the glass transition (T_g) and the crystallization (T_x) for the Dy₅₀Co₅₀ amorphous ribbon is about 521 K and 577 K, respectively. The liquidus

temperatures (T_l) of the Dy₅₀Co₅₀ binary alloy can be obtained from the binary Dy-Co equilibrium phase diagram¹¹. Therefore, the reduced glass transition temperature ($T_{rg} = T_g/T_l$)¹² and the parameter $\gamma (=T_x/(T_g+T_l))$ ¹³ were calculated to be 0.42 and 0.3253, respectively. Hence, the critical section thickness ($Z_c = 2.8 \times 10^{-7} \exp(41.7\gamma)$)¹³ for the Dy₅₀Co₅₀ amorphous alloy is about 0.218 mm. Although it is not as high as for bulk metallic glasses (BMGs), it is sufficient to be fabricated in the shape of ribbons or wires.

The temperature dependence of the zero field cooled (ZFC) and field cooled (FC) magnetization ($M-T$) curves were measured in the heating process under a field of 0.03 T from 2 K to 300 K, as shown in Fig. 2 (a). The sample was preliminarily cooled from 300 K to 2 K under a zero field for ZFC measurement, and cooled from 300 K to 2 K under a field of 0.03 T for FC measurement. The ZFC $M-T$ curve and the FC $M-T$ curve are almost the same within the temperature range of 80 - 300 K. The Curie temperature (T_c) of the Dy₅₀Co₅₀ amorphous ribbon obtained from the derivative of the $M-T$ curves is about 106 K. Divergence between the ZFC and FC $M-T$ curves appears at temperatures lower than 80 K, as shown in Fig. 2 (a). The difference between ZFC and FC $M-T$ curves of the Dy₅₀Co₅₀ amorphous ribbon is similar to those of canonical spin glass systems^{9, 14-15}. The spin freezing temperature (T_f) is about 80 K.

Figure 2 (b) shows the isothermal magnetization ($M-H$) curves of the ribbon measured at the 10 K, 30 K, 50 K, 60 K, 70 K, 80 K, 90 K, 100 K, 150 K and 300 K under a field of 5 T. The ribbon was cooled from room temperature under a zero magnetic field so as to avoid the effect of magnetization history on the low temperature measurement. The magnetization under a low magnetic field decreases from 60K to 10 K, which is in accordance with the ZFC $M-T$ curve, indicating the spin glass like behavior of the Dy₅₀Co₅₀ amorphous ribbon¹⁴⁻¹⁵.

Figure 3 (a) shows the Arrott plots of the Dy₅₀Co₅₀ amorphous ribbon. The plots at 10 K and 30 K exhibit an s-shape feature, which is a typical characteristic of a spin glass system. It is known that Dy(Tb)-based amorphous systems involve huge random magnetic anisotropy (RMA) due to the local random electrostatic field^{9, 14-16}. Competition between the RMA and the exchange interaction dominates the magnetic

behavior of the Dy(Tb)-based amorphous alloys. The magnetization behavior of the Dy₅₀Co₅₀ amorphous ribbon, including the M-T curves, magnetization curves and the Arrott plots, are similar to those of other Tb(Dy)-based metallic glasses, indicating that the magnetic behavior of the Dy₅₀Co₅₀ amorphous alloy is dominated by the RMA at temperatures well below T_f but is controlled by the exchange interaction at temperatures above T_f ^{9, 14-15}.

On the other hand, according to the random anisotropy model, the RMA existing in the amorphous alloys will break the rotational symmetry of the Hamiltonian and increase the hysteresis^{9, 17}. Figure 3 (b) shows the hysteresis loops of the Dy₅₀Co₅₀ amorphous ribbon measured at 10 K, 30 K, 60 K, 80 K, 100 K, 150 K and 300 K under a field of 5 T. The amorphous ribbon is paramagnetic at 300 K and soft magnetic with negligible coercivity at 60 K – 100 K, but is hard magnetic at 10 K and 30 K. The coercivity is about 0.175 T at 30 K and about 0.633 T at 10 K. The large hysteresis at low temperature is due to the strong RMA, and the decreased coercivity with increasing temperature is due to the unfreezing of the magnetic moment with thermal fluctuation^{9, 14}.

It is reported that the spin freezing behavior makes the magnetic entropy change irreversible at temperatures well below the spin freezing temperature, and reduce the magneto-caloric properties of the Dy-based bulk metallic glasses¹⁴. In the present work, we attempt to study the effect of the magnetic structure on the magnetostriction of the Dy₅₀Co₅₀ amorphous ribbon. Figure 4 shows the field dependence of the magnetostriction (λ - H) curves of the Dy₅₀Co₅₀ amorphous ribbon measured at 30 K, 60 K, 70 K, 80 K and 90 K from 0 to 5 T, and then from 5 to 0 T. The λ - H curves are reversible at 60 K, 70 K, 80 K and 90 K, but irreversible at 30 K because the magnetostriction of the ribbon is still about 37 ppm when the magnetic field returns to zero, which is closely related to the RMA and hysteresis of the amorphous ribbon. The reversible magnetostriction of the Dy₅₀Co₅₀ amorphous ribbon increases with decreasing temperature and reaches to a high value of up to 320 ppm under 2 T and nearly 600 ppm under 5 T, both of which are rather high for Tb(Dy)-TM amorphous

alloys⁹⁻¹⁰. The large magnetostriction of the Dy₅₀Co₅₀ amorphous ribbon is anomalous because amorphous alloys are considered to be zero-magnetostriction alloys according to the random anisotropy model for magnetostriction developed by O'Handley and Grant¹⁸. However, we consider that the randomly oriented clusters surrounded by the disordered matrix in amorphous alloys rotate upon magnetization so as to minimize the local anisotropic energy. Such rotation will induce the shear stresses on the surrounding matrix, leading to the macro-deformation of the whole specimen, and will result in large magnetostriction of amorphous alloys.

In summary, we studied the glass forming ability, magnetic properties and magnetostriction of a binary Dy₅₀Co₅₀ amorphous alloy. The binary Dy₅₀Co₅₀ alloy shows a GFA good enough for it to be fabricated in the shape of ribbons or wires. The magnetic behavior of the Dy₅₀Co₅₀ amorphous ribbon is similar to that of canonical spin glass systems, with a Curie temperature of about 106 K and a spin freezing temperature of about 80 K. Hysteresis loops indicate that the amorphous ribbon is paramagnetic at room temperatures, soft magnetic at 80 K to 100 K, and hard magnetic at temperatures well below 80 K. The magnetostriction of the Dy₅₀Co₅₀ amorphous alloy was measured from 30 K to 90 K, with a high reversible magnetostriction of 320 ppm under a field of 2 T and about 600 ppm under a field of 5 T at 60 K. The magnetostriction at 30 K, however, is irreversible and is supposed to deteriorate due to the existence of spin freezing behavior of the amorphous alloys. Considering its simple composition and excellent magnetostriction, the Dy₅₀Co₅₀ binary amorphous alloy is an ideal model alloy for investigating the mechanism of large magnetostriction in Tb(Dy)-TM amorphous alloys. Furthermore, the binary alloy also shows attractive potential for industrial applications by playing a basic alloy role for developing multicomponent amorphous alloys with enhanced GFA and magnetostriction.

The work described in this paper was supported by the Hong Kong Polytechnic University (grant number B-Q53Z).

References

1. E. du Trémolet de Lacheisserie, *Magnetostriction, Theory and Application of Magnetoelasticity* (CRC Press, Boca Raton, Florida, USA, 1993).
2. J. M. Barandiarán, J. Gutiérrez and A. Garcia-Arribas, *Phys. Status Solidi A*, 208, 2258 (2011).
3. R. Abbundi and A. E. Clark, *Tran. Magn.* 13, 1519 (1977).
4. J. Atulasimha and A. B. Flatau, *Smart Matter. Struct.* 20, 043001 (2011).
5. L. Sandlund, M. Fahlander, T. Cedell, A. E. Clark, J. B. Restorff, M. Wun-Fogle, *J. Appl. Phys.* 75, 5656 (1994).
6. D. Atkinson, P. T. Squire, M. R. J. Gibbs and S. N. Hogsdon, *J. Phys. D: Appl. Phys.* 27, 1354 (1994).
7. M. Han, D. F. Liang and L. J. Deng, *J. Mater. Sci.* 40, 5574 (2005).
8. A. Speliotis and D. Niarchos, *Sens. Actuators A*, 106, 298 (2003).
9. Th. Speliotis and D. Niarchos, *Microelectro. Eng.*, 112, 183 (2013).
10. B. Z. Tang, D. Q. Guo, L. Xia, D. Ding and K.C. Chan, *J. Alloys Compd.* 728, 747 (2017).
11. T. B. Massalski, H. Okamoto, P. R. Subramanian, L. Kacprzak, *Binary Alloy Phase Diagrams*, 2nd ed. ASM international, 1990.
12. D. Turnbull, *Contemp. Phys.* 10, 473 (1969).
13. Z. P. Lu, C. T. Liu, *Phys. Rev. Lett.* 91, 115505 (2003).
14. Q. Luo, B. Schwarz, N. Mattern, J. Eckert, *Phys. Rev. B*, 82, 024204 (2010).
15. Q. Luo, P. N. Dinh, X. Kou and J. Shen, *J. Alloys Compd.* 725, 835 (2017).
16. C. Jayaprakash, S. Kirkpatrick, *Phys. Rev. B*, 21, 4072 (1980).
17. R. Harris, M. Plischke and M. J. Zuckermann, *Phys. Rev. Lett.* 31, 160 (1974).
18. R. C. O'Handley and R. J. Grant, in: *Proc. Conf. on Rapidly Quenched Metals*, edited by S. Steeb and H. Warlimont, (Elsevier, Amsterdam, 1985), p. 1125.

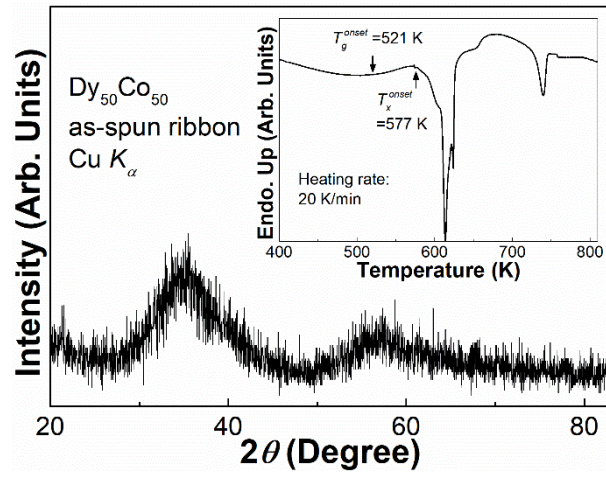
Figure captions

Figure 1 XRD pattern of the Dy₅₀Co₅₀ as-spun ribbon, the inset is the DSC traces of the ribbon at a heating rate of 20 K/min.

Figure 2 (a) ZFC and FC $M-T$ curves of the Dy₅₀Co₅₀ amorphous ribbon under a field of 0.03 T; (b) $M-H$ curves of the ribbon measured at various temperature under a field of 5 T.

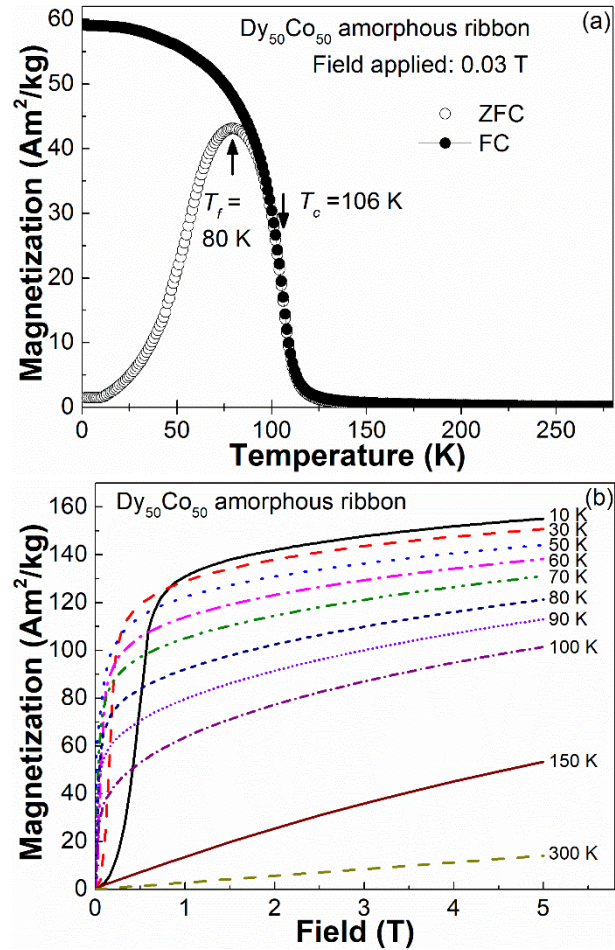
Figure 3 (a) Arrott plots of the Dy₅₀Co₅₀ amorphous ribbon; and (b) the hysteresis loops of ribbon under a field of 5 T.

Figure 4 $\lambda-H$ curves of the Dy₅₀Co₅₀ amorphous ribbon measured at 30 K, 60 K, 70 K, 80 K and 90 K from 0 to 5 T, and then from 5 to 0 T.



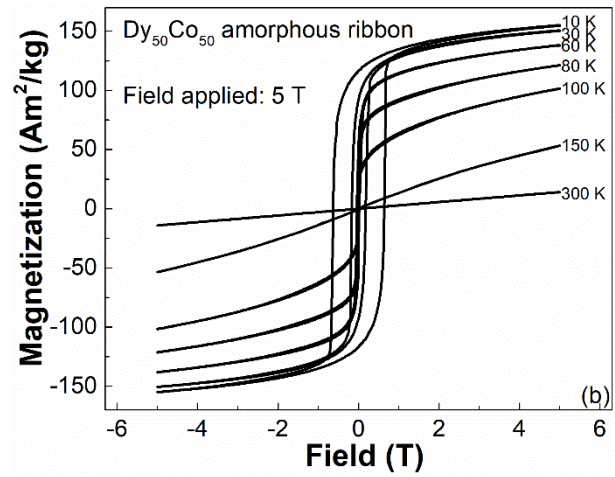
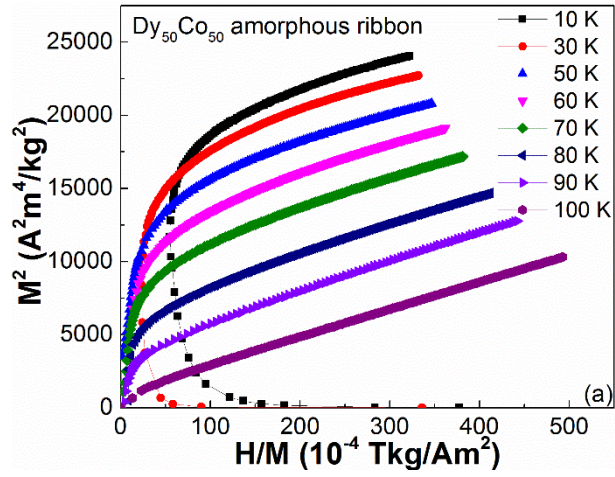
L. Xia *et al.*

Figure 1



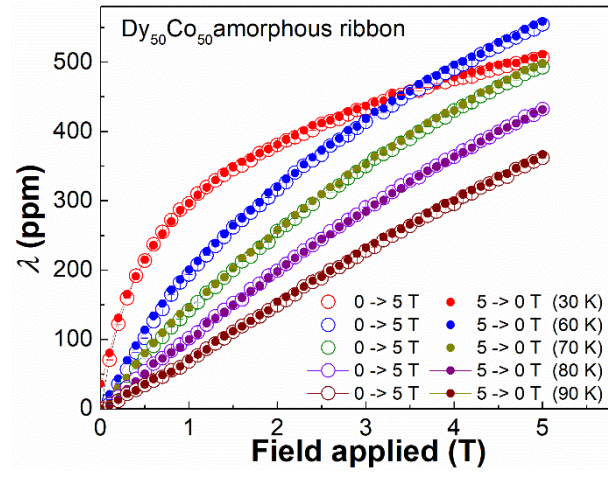
L. Xia *et al.*

Figure 2



L. Xia *et al.*

Figure 3



L. Xia *et al.*

Figure 4