Low Temperature Magnetic Transition of Y(Fe,Mo)₁₂ Alloys

Yi-Zhong Wang^a, Ji-Fan Hu^b and Bo-Ping Hu^a

^a San Huan Research Laboratory, Chinese Academy of Sciences, P.O. Box 603, Beijing 100080, P.R. China

^b Department of Physics, Shandong University, Jinan 250100, P.R. China

(received April 30, 1998; accepted August 26, 1998)

The magnetic properties of YFe_{12-x}Mo_x alloys (x=1-3) have been studied by ac susceptibility χ_{ac} , low field thermomagnetic analysis (TMA) and an extracting sample magnetometer in a temperature range of 1.5-300K. Data from χ_{ac} and TMA indicate that there is a magnetic phase transition occurring below 200K for the alloys with high molybdenum concentrations. The magnetic transition temperature T_f increases as the molybdenum concentration increases. At room temperature, the YFe_{12-x}Mo_x alloys have a hard ferromagnetic phase with uniaxial magnetocrystalline anisotropy. But below T_f , high molybdenum alloys exhibit mectomagnetic behaviour and show obvious magnetohistory effects and a large coercivity, e.g., about 1.7 Tesla for YFe_{9.5}Mo_{2.5} at 1.5 K.

Key words: magnetic transition, ThMn₁₂ structure, Y(Fe,Mo)₁₂ compounds, magnetocrystalline anisotropy, mectomagnetism, narrow-domain-wall pinning.

1. Introduction

In recent years R(Fe,Mo)₁₂ compounds with the ThMn₁₂ structure (1:12) and their nitrides were investigated extensively, since the Mo-containing 1:12 compounds have some unusual magnetic properties, e.g., their Curie temperatures and saturation magnetizations decrease more quickly than that of other third element 1:12 compounds, and they exhibit multiform magnetic transitions at low temperature¹⁾⁻¹¹⁾. In order to understand the influence of Mo on the magnetic properties of the Fe sublattice, numerous studies on crystallographic and magnetic properties of YFe_{12-x}Mo_x have been carried out⁶⁾⁻¹²⁾. Concerning a anomaly appearing below 150 K in the temperature dependence of magnetization for the YFe₁₀Mo₂ alloy, Kou et al. claimed that it results from a temperature-induced competition of the anisotropy among different Fe sites⁷⁾, and then Christides et al. suggested existence of a spin-glass-like magnetic phase due to randomness of exchange interactions from variation of Fe-Fe distances¹⁰⁾, while Wang et al. suggested that it can be attributed to the domain-wall pinning model¹¹⁾.

In this paper we report some new results on magnetic properties in YFe_{12-x}Mo_x alloys (x = 1 - 3) to improve understanding of the magnetic transition at a low temperature.

2. Experiment

Alloys of YFe_{12-x}Mo_x with x = 1, 1.25, 1.5, 1.75, 2, 2.5 and 3 were prepared by arc melting from 99.9% starting materials under an argon gas atmosphere and vacuum

annealed at 1100 °C for 24 h. The annealed alloys have been checked by X-ray diffraction. They are of single phase with the 1:12 type structure. The temperature dependence of the preliminary ac susceptibility χ_{ac} (T) at an ac-field of 80 A/m and a frequency of 220 Hz were measured in the temperature range from 1.5 to 300 K in order to detect a possible temperature-induced magnetic transition. Samples of cylindrical shape with a diameter of 3 mm and a length of 6 mm were made at room temperature by aligning the alloy powders along the cylinder axis in a magnetic field of 1 T and fixing their direction with epoxy resin. The magnetization curves and thermomagnetic data were obtained using an extracting sample magnetometer with applied fields up to 7 T at 1.5 - 300 K and a vibrating sample magnetometer at 77 - 1000 K, respectively.

3. Results and Discussion

In order to obtain information about magnetic transitions in YFe_{12-x}Mo_x alloys, the low field thermomagnetic scan and the ac susceptibility χ_{ac} measurements were used. Figure 1 shows several typical thermomagnetic curves of YFe_{12-x}Mo_x alloys at an applied field of 45 mT after zero-field cooling (ZFC) process. Results of the thermomagnetic measurements were shown that the samples with x = 1 and 1.25 exhibit a normal ferromagnetic behaviour, while the magnetization of samples with x ≥ 1.5 show an anomalous decrease in the thermomagnetic curves below 150 K, and the magnetization of these samples with higher molybdenum

Fig. 1 several typical thermomagnetic curves of YFe_{12-x}Mo_x alloys after zero-field cooling (ZFC) process

concentrations even drops close to zero at 0 K. This implies that local magnetization for thermally demagnetized samples, which on random distribution at room temperature were frozen into different orientations after zero-field cooling (ZFC) process. However after a magnetic field cooling (FC) process, samples with high Mo concentrations have a large magnetization at 0 K and exhibit normal ferromagnetic behaviour. Such irreversible behaviour or magnetohistory effects in $\text{YFe}_{12-x}\text{Mo}_x$ alloys have been investigated in our previous work (see ref.11), where the irreversible behaviour was explained by the domain-wall pinning mechanism. Similar magnetohistory effects were also observed in spin glass¹³⁾, mictomagnetic¹⁴⁾, amorphous¹⁵⁾ and narrow domain wall materials¹⁶⁾.

Table 1 The Curie temperature T_c , the freeze temperature T_f obtained from TMA and $\chi_{ac}(T)$ curves in $\text{YFe}_{12-x}\text{Mo}_x$.

X	$T_c(\text{K})$	$T_f^{\text{TMA}}(\text{K})$	$T_c^{\chi_{ac}}(\text{K})$
1.00	481	0	0
1.25	464	0	0
1.50	440	0	0
1.75	413	30	30
2.00	360	126	137
2.50	300	136	124
3.00	175	150	175

Fig. 2 Several typical ac susceptibility $\chi_{ac}(T)$ curves for the $\text{YFe}_{12-x}\text{Mo}_x$ alloys

Several typical ac susceptibility $\chi_{ac}(T)$ curves for the $\text{YFe}_{12-x}\text{Mo}_x$ alloys are shown in Fig. 2. The samples with $x = 1$ and 1.25 do not show any anomaly in the ac susceptibility $\chi_{ac}(T)$ curves, however samples with $x \geq 1.5$ obviously show one or two cusps on the ac susceptibility $\chi_{ac}(T)$ curves, which revealed the existence of magnetic transitions at low temperature in the samples with $x \geq 1.5$ for $\text{YFe}_{12-x}\text{Mo}_x$ alloys. We can find from Fig.1 and Fig.2 that the dropping temperature of thermomagnetic curves just corresponds to the cusp temperatures on the ac susceptibility $\chi_{ac}(T)$ curves. The higher cusp temperature associates with the Curie temperature, while the lower

cusp temperature, which is called as a freezing temperature T_f , may be associated with another magnetic transition (see below discussion) in $\text{YFe}_{12-x}\text{Mo}_x$ alloys with $x \geq 1.5$.

Fig. 3 The magnetic phase diagram of $\text{YFe}_{12-x}\text{Mo}_x$ alloys.

The magnetic transition temperatures T_f and the Curie temperatures T_c of $\text{YFe}_{12-x}\text{Mo}_x$ alloys are listed together in Table 1 and plotted in Figure 3 gives the magnetic phase diagram of $\text{YFe}_{12-x}\text{Mo}_x$ alloys. It is well known that the $\text{YFe}_{12-x}\text{Mo}_x$ alloys possess a uniaxial magnetocrystalline anisotropy at room temperature^{3),7),11)}. Therefore, the magnetic phase above T_f in $\text{YFe}_{12-x}\text{Mo}_x$ alloys are clear: a paramagnetic phase above T_c and a hard ferromagnetic phase between T_c and T_f . As for the magnetic phase below T_f , we believe that it can be classified into mictomagnetism¹⁴⁾.

The mictomagnetism is characterised by the presence of antiferromagnetic interactions, which may be ascribed to short interatomic Fe-Fe distances of 4f sites, and the presence of concentration fluctuations at 3d sites, where Fe-rich regions behave as ferromagnetic clusters, in $\text{YFe}_{12-x}\text{Mo}_x$ alloys with high Mo concentration. The local magnetic coupling to ferromagnetic clusters leads to a quasi-random orientation of the net cluster moment parallel and antiparallel with the c-axis and form a mictomagnet. Thus, the ferromagnetic configuration is frozen in only for FC process, and a large magnetization appears on the thermomagnetic curves with increasing the temperature. But for ZFC process the magnetizations of ferromagnetic clusters are frozen into different directions and the magnetization become to be very small.

A huge coercivity of 1.7 Tesla at 1.5 K was observed in the $\text{YFe}_{9.5}\text{Mo}_{2.5}$ alloy. Figure 4 shows hysteresis loops of the $\text{YFe}_{9.5}\text{Mo}_{2.5}$ alloy at 1.5 K after FC and ZFC process. The loops display obviously an asymmetry since the applied field of 6.5 Tesla is not enough to saturate the magnetization of the $\text{YFe}_{9.5}\text{Mo}_{2.5}$ sample at 1.5 K. But the coercivity $\mu_0 H_{ci}$ is still very high, with values of 1.7 Tesla for FC process and 1.0 Tesla for ZFC process, respectively. They are far larger than 0.15 tesla obtained for a $\text{YFe}_{10}\text{Mo}_2$ alloy¹⁰⁾.

Fig. 4 The hysteresis loops of the YFe_{9.5}Mo_{2.5} alloy at 1.5 K after FC and ZFC process.

Fig. 5 The magnetization curves obtained parallel and perpendicular to the c-axis on the aligned YFe_{9.5}Mo_{2.5} sample at 1.5 K after FC process.

In order to understand the origin of so large coercivity, the magnetization curves parallel $M_{\parallel}$ and perpendicular $M_{\perp}$ to the c-axis were measured at 1.5 K on the aligned YFe_{9.5}Mo_{2.5} sample after FC process. Figure 5 shows the magnetization curves obtained parallel and perpendicular to the c-axis on the aligned YFe_{9.5}Mo_{2.5} sample at 1.5 K after FC process. From the extrapolated intersection of both magnetization curves, we can obtain the magnetic anisotropy field B_a . This value is about 12 Tesla and was listed in Table 2. From the magnetization curve parallel to the c-axis, the saturation magnetization M_s can be deduced by the low of approach to saturation (see Table 2).

Since the magnetization curves under FC show a uniaxial feature, the value of B_a at 1.5 K should reflect the uniaxial anisotropy of ferromagnetic clusters. The first order anisotropy constant K_1 can be deduced from the equation $K_1 = \mu_0 M_s B_a / 2$. In addition, the exchange integral

Table 2 Several magnetic parameters of the YFe_{9.5}Mo_{2.5} alloy at 1.5 K.

M_s (Am ² /kg)	H_a (kA/m)	K_1 (J/m ³)	δ_B (nm)
480	9554	2.9×10^6	2.0

J can be obtained by the mean-field approximation. J expressed as $J = 3k_B T_c / (2ZS_{Fe}(S_{Fe} + 1))$, where Z being the average number of Fe neighbours (about 8) and S_{Fe} the average iron spin magnetic moment ($0.421\mu_B$). Then the exchange constant A can be deduced from the equation $A = JS_{Fe}^2 / d_{Fe}$ where d_{Fe} being the Fe-Fe average interatomic distance (about 0.26 nm). The value of A is 11.7×10^{-3} J/m in the YFe_{9.5}Mo_{2.5} alloy. Thus, using the classical formula $\delta_B = \pi \times 10^{-2} (A/K_1)^{1/2}$ we can estimate the domain wall width of the YFe_{9.5}Mo_{2.5} alloy at 1.5 K. The domain wall width is only 2.0 nm. Therefore so large coercivity seen above may be attributed to the presence of the narrow-domain-walls.

Reference

- 1). M. Anagnostou, C. Christides and D. Niarchos: *Solid State Commun.* **78**, 681 (1991).
- 2). Y. Z. Wang, G. C. Hadjipanayis, A. Kim, J. D. Sellmyer and W.B. Yelon: *J. Magn. Magn. Mater.* **104-107**, 1132 (1992).
- 3). Y. Z. Wang, B. P. Hu, X. L. Rao, G. C. Liu, L. Yin, W. Gong and G. C. Hadjipanayis: *J. Appl. Phys.* **73**, 6251 (1993).
- 4). Y. C. Yang, Q. Pan, X. D. Zhang, J. Yang, M. H. Zhang and S. L. Ge: *Appl. Phys. Lett.* **61**, 2723 (1992).
- 5). Y. Z. Wang, B. P. Hu, K. Y. Wang, G. C. Liu and W. Y. Lai: *J. Magn. Magn. Mater.* **140-144**, 1019 (1995).
- 6). H. Sun, M. Akayama, K. Tatami and H. Fujii: *Physica B* **183**, 33 (1992).
- 7). X. C. Kou, C. Christides, R. Grossinger, H. R. Kirchmayr and A. Kostikas: *J. Magn. Magn. Mater.* **104-107**, 1341 (1992).
- 8). X. C. Kou, R. Grossinger, G. Wiesinger, J. P. Liu, F. R. de Boer, I. Kleinschroth, H. Kronmüller: *Phys. Rev. B* **51**, 8254 (1995).
- 9). Y. Z. Wang, B. P. Hu, L. Song, K. Y. Wang, G. C. Liu: *J. Phys.: Condens. Mater.* **6**, 7085 (1994).
- 10). C. Christides, A. Kostikas, G. Zouganelis, V. Psyharis, X. C. Kou, R. Grossinger: *Phys. Rev. B* **47**, 11220 (1993).
- 11). Y. Z. Wang, B. P. Hu, G. C. Liu, J. F. Hu, L. Song, K. Y. Wang and W. Y. Lai: *J. Appl. Phys.* **76**, 6383 (1994).
- 12). H. Sun, Y. Moril, H. Fujii, M. Akayama and S. Funahashi: *Phys. Rev. B* **48**, 13333 (1993).
- 13). K. moorjani and J. M. D. Coey: *Magnetic Glasses* (Elsevier, New York, 1984).
- 14). R. D. Shull, H. Okamoto and P. A. Beck: *Solid State Commu.* **20**, 863 (1976).
- 15). J. M. D. Coey, D. Givord, A. Lienard, and J. P. Rebouillat: *J. Phys. F* **11**, 2707 (1981).
- 16). B. Barbara, C. Becle, R. Lemaire, and D. Paccard: *J. Phys. (Paris)*, **32**, C1-299 (1971).