# A study of the magnetocrystalline anisotropy of $Sm_{1-x}Dy_{x}Fe_{10.5}Mo_{1.5}(x = 0-1.0)$

C. P. YANG\* Department of Material Science & Engineering, Tsinghua University, Beijing 100084, People's Republic of China E-mail: yangcp@263.net
Y. Z. WANG<sup>‡</sup> The State Key Laboratory for Magnetism, Institute of Physics, Chinese Academy of Sciences, Beijing 100080, People's Republic of China
B. P. HU
San Huan Research Laboratory, Chinese Academy of Sciences, P.O. Box 603, Beijing 100080, People's Republic of China
J. L. WANG
The State Key Laboratory for Magnetism, Institute of Physics, Chinese Academy of Sciences, Beijing 100080, People's Republic of China

Z. L. JIANG, X. Y. CHEN, Department of Material Science & Engineering, Tsinghua University, Beijing 100084, People's Republic of China

Z. X. WANG San Huan Research Laboratory, Chinese Academy of Sciences, P.O. Box 603, Beijing 100080, People's Republic of China

J. ZHU

Department of Material Science & Engineering, Tsinghua University, Beijing 100084, People's Republic of China

The magnetocrystalline anisotropy and the spin reorientation of  $Sm_{1-x}Dy_xFe_{10.5}Mo_{1.5}$  were investigated in detail. At room temperature, all  $Sm_{1-x}Dy_xFe_{10.5}Mo_{1.5}$  alloys possess easy *c*-axis anisotropy and the magnetocrystalline anisotropy field decreases with increasing Dy concentration. However, at low temperature, a spin reorientation transition of axis-to-cone type was observed in the  $Sm_{1-x}Dy_xFe_{10.5}Mo_{1.5}$  alloys with  $x \ge 0.8$ . The spin reorientation temperatures increase with increasing Dy concentration in the  $Sm_{1-x}Dy_xFe_{10.5}Mo_{1.5}$  alloys. © 1999 Kluwer Academic Publishers

## 1. Introduction

In the past ten years, a lot of studies were focused on the R(Fe, M)<sub>12</sub> compounds and their nitrides (R = rare earth and M = Ti, Cr, V, Mo, Si, W, Nb) for they are hopefully developed into excellent permanent-magnets [1–3]. Among the R(Fe, M)<sub>12</sub> compounds, R(Fe, Mo)<sub>12</sub> shows a remarkable difference from other R(Fe, M)<sub>12</sub> compounds such as M = Ti, Cr, V, Si, W, and Nb in the aspects of: (1) The Curie temperature great more quickly decreases with Mo concentration in R(Fe, Mo)<sub>12</sub> than with M in other R(Fe, M)<sub>12</sub> compounds [1, 2]; (2) Magnetohistory effects were observed in the R(Fe, Mo)<sub>12</sub> compounds with R = Y [4, 5], Lu [4], or Dy [6] on the contrast no magnetohistory efffects observed in other R(Fe, M)<sub>12</sub> compounds; (3) The R(Fe, Mo)<sub>12</sub> compounds and their nitrides can be easily formed and contain not much Mo concentration so that they were attracted more attention on the potential technological application [7]. Recently, many efforts were concentrated on R(Fe, Mo)<sub>12</sub>N<sub>y</sub> compounds including the anisotropic magnetic powders of NdFe<sub>10.5</sub>Mo<sub>1.5</sub>N<sub>y</sub>, which had a maximum energy product of (*BM*)<sub>max</sub> = 169.6 kJ/m<sup>3</sup> (21.2 MGOe), reported in 1997 [8]. The studies on spin reorientation transition (SRT) were considered as a significant method to understand the anisotropy and crystal-field interactions of the R-sublattice in R(Fe, M)<sub>12</sub>. Many authors have investigated on the magnetic anisotropy

<sup>\*</sup> Also with The State Key Laboratory for Magnetism, Institute of Physics, Chinese Academy of Sciences, Beijing 100080, People's Republic of China.

<sup>&</sup>lt;sup>‡</sup> Also with San Huan Research Laboratory, Chinese Academy of Sciences, P.O. Box 603, Beijing 100080, People's Republic of China.

in Sm(Fe, Mo)<sub>12</sub> [9–11] and SRT in Dy(Fe, Mo)<sub>12</sub> [12–16]. In this present work, We emphasise on investigating the magnetocrystalline anisotropy and spin reorientation in the Sm<sub>1-x</sub>Dy<sub>x</sub>Fe<sub>10.5</sub>Mo<sub>1.5</sub> series.

## 2. Experimental details

Alloys with composition of  $\text{Sm}_{1-x}\text{Dy}_x\text{Fe}_{10.5}\text{Mo}_{1.5}$ (x = 0, 0.2, 0.4, 0.6, 0.8 and 1.0) were prepared by arc melting the starting materials with a purity better than 99.5% under an atmosphere of high pure argon, and then annealing in vacuum at 1050 °C for 5–10 h. The quality of the samples was checked by powder X-ray diffraction (XRD) and thermomagnetic analysis (TMA) method. The spin reorientation transition was detected both by TMA using an extracting-sample magnetometer in an applied field of 40 mT from 1.5 K to room temperature and by ac susceptibility ( $\chi_{ac}$ ) measurement in an ac field of 0.1 mT with a frequency of 220 Hz from 4.2 K to room temperature.

The magnetic structure of  $Sm_{1-x}Dy_xFe_{10.5}Mo_{1.5}$  at room temperature and 1.5 K were determined by X-ray diffraction and the magnetization curves on the magnetically aligned samples respectively. The anisotropy field  $\mu_0H_a$  at room temperature was measured by the singular point detection (SPD) [17] technique on the aligned  $Sm_{1-x}Dy_xFe_{10.5}Mo_{1.5}$  samples.

### 3. Results and discussion

Studies of X-ray diffraction patterns and thermomagnetic analysis indicate that all the  $Sm_{1-x}Dy_{x}Fe_{10.5}$ Mo1.5 samples consist of the main phase of  $R(Fe, Mo)_{12}$ , which has a tetragonal ThMn<sub>12</sub>-type structure, and a few  $\alpha$ -Fe as the impurity phase. The values of Curie temperatures  $T_c$  obtained by TMA are listed in Table I. Fig. 1 shows several thermomagnetic curves of  $Sm_{1-x}Dy_{x}Fe_{10.5}Mo_{1.5}$  measured in a low field of 40 mT from 1.5 K to room temperature. It can be seen that the thermomagnetic curves go smoothly when x < 0.8 as seen in the Sm<sub>0.6</sub>Dy<sub>0.4</sub>Fe<sub>10.5</sub>Mo<sub>1.5</sub> and Sm<sub>0.6</sub>Dy<sub>0.4</sub>Fe<sub>10.5</sub>Mo<sub>1.5</sub>. However, an evident cusp, which indicating a spin reorientation transition, was observed on the samples with x = 0.8-1.0. It means that the magnetic structure was changed at low temperature for the  $Sm_{1-x}Dy_{x}Fe_{10.5}Mo_{1.5}$  samples with a high Dy Concentration.

In order to determine the spin reorientation transition temperature ( $T_{sr}$ ) precisely, susceptibility measurements were performed on the Sm<sub>1-x</sub>Dy<sub>x</sub>Fe<sub>10.5</sub>Mo<sub>1.5</sub> samples. Fig. 2 gives several typical results of the  $\chi_{ac}$ 

TABLE I The data of the Curie temperatures  $T_c$ , the saturation magnetization  $M_s$ , the spin reorientation transition temperatures  $T_{sr}$ , the easy magnetization direction EMD and the magnetocrystalline anisotropy fields  $\mu_0 H_a$  for Sm<sub>1-x</sub>Dy<sub>x</sub>Fe<sub>10.5</sub>Mo<sub>1.5</sub> alloys

x	<i>Т</i> <sub>с</sub> (К)	$\frac{M_s \text{ (A m^2/kg)}}{1.5 \text{ K}}$	$\frac{T_{\rm sr}~({\rm K})}{{\rm by}~\chi_{\rm ac}}$	EMD		$\mu_0 H_a$ (T)	
				1.5 K	293 K	1.5 K	293 K
0	485	113.90	0	c-axis	c-axis	14.0	7.90
0.2	481	112.82	0	c-axis	c-axis	10.5	5.70
0.4	472	93.67	0	c-axis	c-axis	6.0	3.70
0.6	466	75.65	0	c-axis	c-axis	2.0	2.49
0.8	454	66.16	98	cone	c-axis	_	2.02
1.0	440	62.21	156	cone	c-axis	_	1.03



*Figure 1* Typical thermomagnetic curves at 1.5–293 K in a field of 40 mT for  $Sm_{1-x}Dy_xFe_{10.5}Mo_{1.5}$ .



*Figure 2* Temperature dependence of ac susceptibility  $\chi_{ac}(T)$  at 4.2–293 K for several  $Sm_{1-x}Dy_{x}Fe_{10.5}Mo_{1.5}$  alloys.

measurements. The position of the peak in  $\chi_{ac}$  curves was defined as spin reorientation transition temperature  $T_{\rm sr}$  which marked by an arrow in Fig. 2. The values of  $T_{\rm sr}$  measured from  $\chi_{\rm ac}$  curves are also listed in Table I. It follows that  $T_{\rm sr}$  increases with Dy concentration in the  $Sm_{1-x}Dy_{x}Fe_{10.5}Mo_{1.5}$  alloys. Theoretically, the magnetic structure of  $R(Fe, M)_{12}$  is a result of the anisotropy competition between the R and Fe-sublattices [18]. In the  $Sm_{1-x}Dy_{x}Fe_{10}$  5Mo<sub>1</sub> 5 system,  $SmFe_{10}$  5Mo<sub>1</sub> 5 has an easy *c*-axis type due to the uniaxial anisotropy of Sm, because of a positive second-order Stevens coefficient  $\alpha_2$ . However, with increasing the Dy concentration, a spin reorientation transition could be expected to occur in the  $Sm_{1-x}Dy_{x}Fe_{10.5}Mo_{1.5}$  alloys with a high Dy concentration when the planar anisotropy of Dy-sublattice overcomes the total uniaxial anisotropy of Sm and Fesublattice. A similar change of  $T_{\rm sr}$  was also observed in



Figure 3 Typical X-ray diffraction patterns on magnetically aligned samples of (a) Sm<sub>0.8</sub>Dy<sub>0.2</sub>Fe<sub>10.5</sub>Mo<sub>1.5</sub> and (b) Sm<sub>0.2</sub>Dy<sub>0.8</sub>Fe<sub>10.5</sub>Mo<sub>1.5</sub>.



Figure 4 SPD curve of DyFe<sub>10.5</sub>Mo<sub>1.5</sub> at room temperature.

 $Dy_{1-x}Y_xFe_{11}Mo$  [19],  $Dy_{1-x}Y_xFe_{11.35}Nb_{0.65}$  [20] and  $Dy_{1-x}Y_xFe_{11}Ti$  compounds [21].

Fig. 3 illustrates typical X-ray diffraction patterns on the magnetically aligned samples at room temperature. It indicates all  $\text{Sm}_{1-x}\text{Dy}_x\text{Fe}_{10.5}\text{Mo}_{1.5}$  alloys exhibit uniaxial anisotropy at room temperature for only line (002) left in the diffraction patterns. It is reasonable as considering of even  $\text{DyFe}_{10.5}\text{Mo}_{1.5}$ , which possessing the largest planar anisotropy in the  $\text{Sm}_{1-x}\text{Dy}_x\text{Fe}_{10.5}\text{Mo}_{1.5}$  system, still shows a uniaxial anisotropy of 1.03 T at room temperature [6]. The anisotropy field  $\mu_0 H_a$  at room temperature was measured by singular point detection (SPD) technique on the magnetically aligned sample in a pulsed-field up to 25.0 T. Fig. 4 gives the SPD curve of  $\text{DyFe}_{10.5}\text{Mo}_{1.5}$ 



*Figure 5* Dy-concentration dependence of the magnetocrystalline anisotropy fields measured by SPD at room temperature for  $Sm_{1-x}Dy_x$  Fe<sub>10.5</sub>Mo<sub>1.5</sub>.



Figure 6 Magnetization curves of magnetization parallel component  $M_{\parallel}$  and perpendicular component  $M_{\perp}$  at 1.5 K as a function of applied field  $\mu_0 H_a$ .

as a typical example. The position of the cusp in the curve of the second-order time derivative  $dM^2(B_0)/dt^2$  presents the magnetocrystalline anisotropy field. The  $\mu_0 H_a$  values are listed in Table I and plotted in Fig. 5. The magnetocrystalline anisotropy field remarkably decreases monotonically with increasing Dy concentration till a lowest value of  $\mu_0 H_a = 1.03$  T arriving at DyFe<sub>10.5</sub>Mo<sub>1.5</sub>.

Fig. 6 gives the applied field dependence of magnetization parallel component  $M_{\perp}$  and perpendicular component  $M_{\perp}$  for several Sm<sub>1-x</sub>Dy<sub>x</sub>Fe<sub>10.5</sub>Mo<sub>1.5</sub> samples at 1.5 K. Saturation magnetizations  $M_s$ , obtained from  $M_{\parallel}$  curves by applying the law of approach to saturation, are also listed in Table I. From Fig. 6, we can see that the feature of the magnetization curve of Sm<sub>0.8</sub>Dy<sub>0.2</sub>Fe<sub>10.5</sub>Mo<sub>1.5</sub> shows a great difference from Sm<sub>0.2</sub>Dy<sub>0.8</sub>Fe<sub>10.5</sub>Mo<sub>1.5</sub> for the two samples have the different types of magnetic structure and anisotropy at 1.5 K. It is clear that Sm<sub>0.8</sub>Dy<sub>0.2</sub>Fe<sub>10.5</sub>Mo<sub>1.5</sub> shows a canted anisotropy for the magnetization  $M_{\perp}(0)$  of the hard direction has a considerable value of about 12% of the saturation magnetization  $M_s$  near to the zero field. A similar spin reorientation transition of easy *c*-axis to cone was also observed in the Dy(Fe, Mo)<sub>12</sub> compounds [6, 15].

In conclusion, the Curie temperatures, saturation magnetizations, and especially, the anisotropy and spin reorientation transition were investigated on the  $Sm_{1-x}Dy_xFe_{10.5}Mo_{1.5}$  system. The Curie temperatures and saturation magnetizations at 1.5 K monotonically decrease with increasing Dy concentration. At room temperature, all  $Sm_{1-x}Dy_xFe_{10.5}Mo_{1.5}$  alloys exhibit uniaxial anisotropy when rapidly decrease from 8.2 T for  $SmFe_{10.5}Mo_{1.5}$  to 1.03 T for  $DyFe_{10.5}Mo_{1.5}$ . However, a spin reorientation transition of axis-to-cone type take place in the  $Sm_{1-x}Dy_xFe_{10.5}Mo_{1.5}$  samples with a high Dy concentration of x = 0.8-1.0. The spin reorientation temperatures  $T_{sr}$  are 98 K and 156 K for  $Sm_{0.2}Dy_{0.8}Fe_{10.5}Mo_{1.5}$  and  $DyFe_{10.5}Mo_{1.5}$ 

#### References

- K. H. J. BUSCHOW and D. B. DE MOOIJ, "Concerted European Action on Magnets (CEAM)" (Elsevier, London, 1989) p. 63.
- HONG-SHOU LI and J. M. D. COEY, "Handbook of Magnetic Materials," Vol. 6 (Elsevier Science Publishers B. V., 1991).

- B. P. HU, K. Y. WANG, Y. Z. WANG and Z. X. WANG, *Phys. Rev. B* 51 (1995) 2905.
- 4. C. CHRISTIDES, A. KOSTIKAS, G. ZOUGANELIS and V. PSYCHARIS, *ibid. B* 47 (1993) 1220.
- Y. Z. WANG, B. P. HU, G. C. LIU, J. H. HU, L. SON, K. Y. WANG and W. Y. LAI, *J. Appl. Phys.* 76 (1994) 6383.
- 6. C. P. YANG, Y. Z. WANG, B. P. HU and Z. X. WANG, J. Phys.: Condens. Matter 10 (1998) 4177.
- 7. M. S. ANAGNOSTON, C. CHRISTIDES, M. PISSAS and D. NIARCHOS, J. Appl. Phys. 70 (1991) 6013.
- 8. J. B. YANG, W. H. MAO, B. P. CHENG, Y. C. YANG,
  H. XU, B. S. HAN, S. L. GE and W. J. KU, *Appl. Phys. Lett.* **71** (1997) 3290.
- 9. A. MOLER, J. Appl. Phys. 64 (1988) 249.
- 10. C. KOUSTLER, L. SCHULTZ and G. THOMAS, *ibid.* 67 (1990) 2532.
- M. ANAGNOSTON, C. CHRISTIDES and D. NIARCHOS, Solid State Commun. 78 (1991) 681.
- X. C. KOU, C. CHRISTIDES, R. GROSSINGER, H. R. KIRCHMAYR and A. KOSTIKAS, J. Magn. Magn. Mater. 104–107 (1992) 1341.
- 13. ROY TUCKER, XIE XU and S. A. SHAHEEN, *J. Appl. Phys.* **75** (1994) 6229.
- 14. C. CHRISTIDES, A. KOSTIKAS, X. C. KOU, R. GROSSINGER and D. NIARCHOS, J. Phys.: Condens. Matter 5 (1993) 8611.
- 15. X. C. KOU, R. GROSSINGER and WIESINGER, *Phys. Rev. B* 51 (1995) 8254.
- K. YU. GUSLIENKO, E. H. C. P. SINNECKER and R. GROSSINGER, *ibid.* 55 (1997) 380.
- 17. G. ASTI and S. RINALDI, Phys. Rev. Lett. 28 (1972) 1584.
- HU BO-PING, LI HONG-SHOU, GAVIGAN J P and COEY J M D, J. Phys.: Condens. Matter 1 (1989) 775.
- C. P. YANG, Y. Z. WANG, B. P. HU and Z. X. WANG, J. Alloys and Compounds 4 (1998) 258.
- 20. K. Y. WANG, C. Y. PAN, Y. Z. WANG, B. P. HU, J. F. HU, W. Y. LAI and Z. X. WANG, 1995 Intermag' 95, San Autonio, Texas USA, FE-07.
- 21. P. H. QUANG, N. H. LUONG, N. P. THUY, T. D. HIEN and J. J. M. FRANSE, J. Magn. Magn. Mater. 128 (1993) 67.

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