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Spontaneous magnetostriction of $Dy_2AlFe_{13}Mn_3$ compound*

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The structural and magnetic properties of $Dy_2AlFe_{13}Mn_3$ compound have been investigated by means of x-ray diffraction and magnetization measurements. The $Dy_2AlFe_{13}Mn_3$ compound has a hexagonal Th_2Ni_{17}-type structure. Negative thermal expansion is found in the $Dy_2AlFe_{13}Mn_3$ compound at the temperatures ranging from 245 to 344K by x-ray dilatometry. The coefficient of the average thermal expansion is $\overline{\alpha} = -1.1 \times 10^{-4}K^{-1}$. The spontaneous magnetostrictive deformations in the 105–360K temperature range have been calculated by means of the differences between the experimental values of the lattice parameters and the corresponding values extrapolated from the paramagnetic range. The result shows that the spontaneous volume magnetostrictive deformation ω_s increases from 7.0×10^{-3} at 105K to 9.1×10^{-3} at 245K, and then decreases with the temperature increasing. The spontaneous linear magnetostrictive deformation λ_c along the c axis decreases as temperature increases. The spontaneous linear magnetostrictive deformation λ_a in basal-plane increases from 0.8×10^{-3} at 105K to 3.4×10^{-3} at 270K, and then decreases with the temperature increasing further.

Keywords: $Dy_2AlFe_{13}Mn_3$ compound, spontaneous magnetostriction, negative thermal expansion
PACC: 7550B, 7530C

1. Introduction

Materials with negative thermal expansion have many important applications as components in composites in order to reduce the composites' overall thermal expansion, but these materials are known in only several oxide systems[1,2] and a few Invar alloys.[3] When the negative thermal expansion occurs, the contraction is usually small and limited to a narrow temperature range beyond room temperature. Notable exceptions are ZrW_2O_8 and HfW_2O_8 with the cubic phase of the ZrW_2O_8-type, which show negative thermal expansion over a very broad range of temperatures from 0.3 to 1050K.[3,4] The discovery of the new materials with a negative coefficient of thermal expansion and further insight into the mechanism may, therefore, play an important role in theory and applications.

The relatively low values of Curie temperatures for the binary rare-earth(R)–iron intermetallic compounds with Th_2Zn_{17} or Th_2Ni_{17} structure and the fact that none of them exhibits easy-axis anisotropy at room temperature restrict the possible application of these materials as permanent magnets.[5] Numerous investigations have been made to improve their magnetic properties. The most effective way of increasing the Curie temperatures of R_2Fe_{17} compounds and modifying the anisotropy is to form interstitial or substitutional solid solutions. The discoveries that the introduction of nitrogen or carbon into R_2Fe_{17} leads to a dramatic increase in the Curie temperature and that nitrogenation of Sm_2Fe_{17} changes the magnetocrystalline anisotropy from basal to axial make these compounds to be potentially interesting materials in the application of permanent magnets.[6,7] Recent attention has focused on the investigation of the magnetic and structural properties of Mn-substituted R_2Fe_{17}.[7−12] It was found that the Curie temperatures of Mn-substituted R_2Fe_{17} compounds decreased fast. However they show a large posi-

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tive spontaneous volume magnetostriction in magnetic states, which leads to a large negative volume change anomaly near their Curie temperatures. For example, in $Y_2Al_3Fe_{11}Mn_3$ compound,[11] the average thermal expansion coefficient is $\bar{\alpha}=\frac{\Delta v}{v\Delta T}\approx -7.5\times10^{-5}$/K in the 185–200K temperature range. So, one is interested in the study of these materials interesting from the viewpoints of both basic research and applications. In this work, the thermal expansion behaviour of the unit-cell volume of $Dy_2AlFe_{13}Mn_3$ compound in a range of temperatures from 105 to 650K and its spontaneous magnetostrictions are investigated by means of x-ray dilatometry and magnetization measurements.

2. Experiment

The compound of $Dy_2AlFe_{13}Mn_3$ was prepared by arc melting in high-purity argon atmosphere. The raw materials of Dy, Fe, Mn and Al used in the experiment had at least 99.95% purity. The ingot was remelted at least three times to ensure its homogeneity and sealed in a silicon vacuum tube, then annealed at 950°C for 7 days, and after that quenched in water. The ingot was ground into powder. To reduce the stress, the powder was sealed in a silicon vacuum tube, annealed at 300°C for 5h and slowly cooled down to room temperature. The powder x-ray diffraction with Cu Kα radiation was used to examine the phase structure of the sample.

The Curie temperature T_C was derived from the temperature dependence of the magnetization curve measured by a vibrating sample magnetometer in a low field of 40 kA/m. The thermal expansion was measured by x-ray dilatometry on a diffractometer. For the determination of the lattice parameters a and c of the $Dy_2AlFe_{13}Mn_3$ compound in the temperatures ranging from 105 to 650K, the sample was placed into an evacuated chamber, and the step-scanning (at 0.01° intervals) x-ray diffraction patterns of the (112) and (222) and reflections were recorded by the x-ray diffractometer with Cu K_α radiation monochromatized by a single-crystal graphite monochromator. The experimental error in determination of a and c was 10^{-4} nm. The magnetostrictive deformations ω_s, λ_a, λ_c were determined by means of the differences between the experimental values v_m, a_m, c_m of the lattice parameters at a given temperature and the corresponding values v_p, a_p, c_p extrapolated from the paramagnetic range according to Debye theory and Grüneisen relation. The value of the Debye temperature T_D, which is necessary for the extrapolation, was estimated from acoustical measurements to be 450K for Y_2Fe_{17} and 400K for other R_2Fe_{17} compounds.[13] We used the same value 400K for the extrapolation of the temperature dependence of the lattice parameters of the sample.

3. Results and discussion

Figure 1 gives the x-ray pattern of $Dy_2AlFe_{13}Mn_3$ compound at room temperature (300K). The indices of crystallographic plane (h k l) of reflections are shown in Fig.1. They indicate that $Dy_2AlFe_{13}Mn_3$ compound is in a single phase with the Th_2Ni_{17}-type structure.

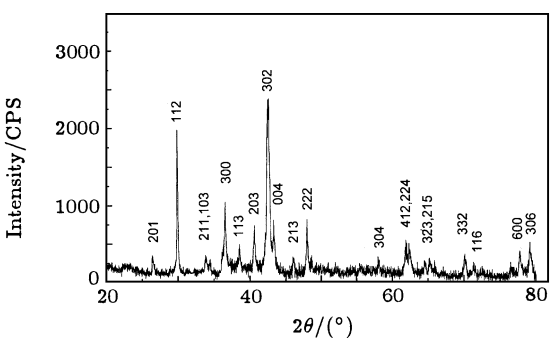


Fig.1. X-ray pattern of the $Dy_2AlFe_{13}Mn_3$ compound at room temperature (300K).

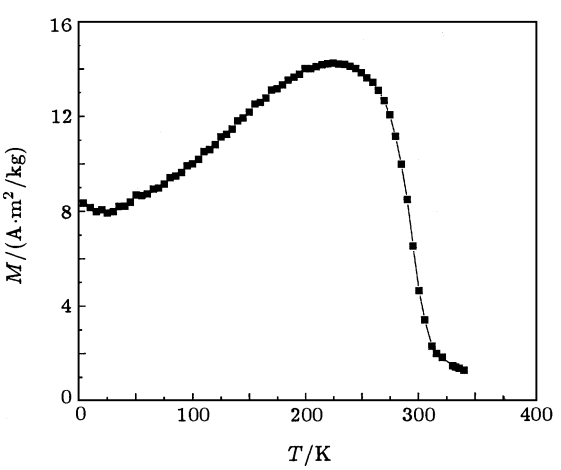


Fig.2. Temperature dependence of the magnetization of $Dy_2AlFe_{13}Mn_3$ compound in a low field of 40kA/m.

Figure 2 shows the temperature dependence of the magnetization of $Dy_2AlFe_{13}Mn_3$ compound in a low field of 40kA/m from which we can derive the Curie temperature of the sample, and it is about 320K.

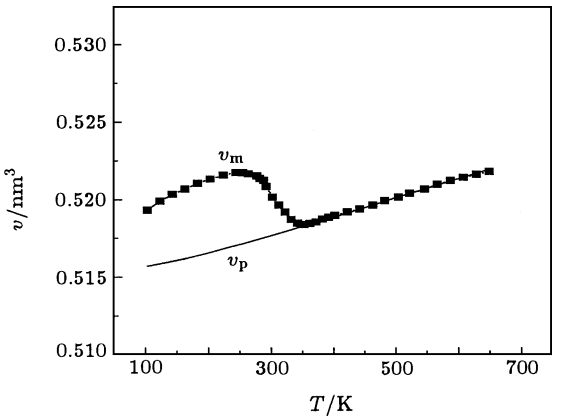


Fig.3. Temperature dependence of the unit cell volume v of $Dy_2AlFe_{13}Mn_3$ compound.

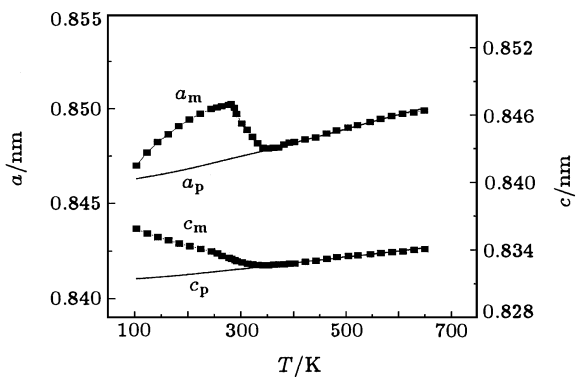


Fig.4. Temperature dependences of lattice parameters a and c of $Dy_2AlFe_{13}Mn_3$ compound.

Figure 3 shows the temperature dependence of the unit cell volume v of $Dy_2AlFe_{13}Mn_3$ compound. If the variation of the rate of v (v_m) is considered in a range of temperatures from 105 to 650K, we will have the average thermal expansion coefficients $\overline{\alpha} = \Delta v/v\Delta T = 3.2 \times 10^{-5}$/K in the 105–245K temperature range, $\overline{\alpha} = -1.1 \times 10^{-4}$/K in 245–344K, and $\overline{\alpha} = 2.3 \times 10^{-5}$/K in 344–650K. We suppose that the negative thermal expansion behaviour in the temperatures ranging from 245 to 344K results from the decrease not only in the magnetic interactions but also in the sharp of magnetic moment. This can be seen in Fig.2. It shows that the magnetic order rapidly drops from about 240K. This transition would be accompanied by a large negative volume change anomaly. Just as $Y_2Al_3Fe_{11}Mn_3$ compound in Ref.[11], we suppose that its origin is intimately related with the loss of vibrating energy which is due to an increase in the kinetic energy of the d electrons through the lattice contraction. It is obvious that the average thermal expansion coefficient in the 105–245K temperature range is larger than that in the 344–650K temperature range. It is correlated with the increase in the magnetic interaction and magnetic moment. It maybe implies that the moment of Mn turns partly to the direction of the moment of Fe with temperature increasing from 105 to 245K.

Figure 4 shows the temperature dependences of lattice parameters a and c respectively, indicating that the negative thermal expansion of the sample in the temperatures ranging from 245 to 344K is anisotropic.

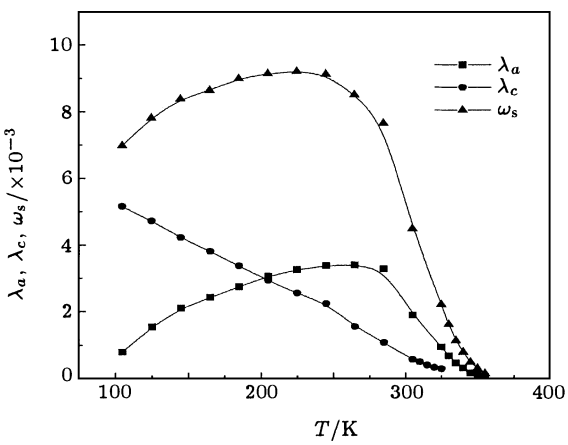


Fig.5. Temperature dependences of the spontaneous magnetostrictive deformation respectively for ω_s, λ_c, andλ_a of $Dy_2AlFe_{13}Mn_3$ compound.

The temperature dependences of the extrapolated values v_p, a_p, and c_p are given in Fig.3 and Fig.4, respectively. From Fig.3 and Fig.4, we can derive the temperature dependence of the spontaneous volume magnetostrictive deformation ω_s by the relation $\omega_s = (v_m - v_p)/v_p$, and the temperature dependence of the spontaneous linear magnetostrictive deformations λ_c along the c axis and λ_a in the basal-plane by the relations $\lambda_c = (c_m - c_p)/c_p$ and $\lambda_a = (a_m - a_p)/a_p$, respectively. Here the subscript letters m and p denote the unit cell parameters in the magnetic state and paramagnetic state, respectively. The temperature dependence of ω_s is shown in Fig.5. It is obvious that the ω_s increases from 7.0×10^{-3} at 105K to 9.1×10^{-3} at 245K and then decreases with temperature increasing. We supposed that the increase in ω_s in the 105–245K temperature range was correlated with the increase in the interaction and the magnetic moment of $Dy_2AlFe_{13}Mn_3$ compound. As mentioned above, this implies that the moment of Mn turns partly to the direction of the moment of Fe with temperature increasing. Above 245K, the ω_s decreases with temperature increasing, which may be due to the decrease in magnetic interaction and magnetic moment of $Dy_2AlFe_{13}Mn_3$ compound. The ω_s

disappears at about 40K which is above T_C. This apparently reflects short-range ordering. The temperature dependences of λ_c and λ_a are both shown in Fig.5. It is noticed that the spontaneous linear deformation λ_c is larger than λ_a at the same temperature below 200K, and the λ_c decreases with temperature increasing. The change of λ_a with temperature increasing is complicated. At low temperatures (T <270K),the λ_a increases with temperature increasing. It maybe implies that the interaction and the magnetic moment in the basal-plane both increase. On the other hand, at high temperatures (T >270K), the λ_a decreases with temperature increasing. This is due to the decrease in the magnetic interaction and magnetic moment in the basal-plane.

4. Conclusions

We can conclude that there exist an anisotropic strong spontaneous magnetostriction and a negative thermal expansion coefficient in $Dy_2AlFe_{13}Mn_3$ compound at the temperatures ranging from 245 to 344K.

References

[1] Evans J S O, Hu Z, Jorgensen J D, Argyriou D N, Short S and Sleight A W 1997 *Science* **75** 61

[2] Mary T A, Evans J S O, Vogt T and Sleight A W 1996 *Science* **272** 90

[3] Lagarec K and Rancourt D G 2000 *Phys. Rev.* B **62** 978

[4] Evans J S O, Mary T A and Sleight A W 1998 *J. Solid State Chem.* **138** 148

[5] Buschow K H J 1977 *Rep. Phys.* **40** 1179

[6] Coey J M D and Sun H 1990 *J. Magn. Magn. Mater.* **87** L251

[7] Shen B G, Wang F W, Kong L S and Cao L 1993 *J. Phys.: Condens. Mater.* **5** L685

[8] Chen Z H, Shen B G, Yan Q W, Guo H Q, Chen D F, Gou C, Sun K, de Boer F R and Buschow K H J 1998 *Phys. Rev.* B **57** 14299

[9] Shen B G, Cheng Z H, Wang F W, Yan Q W, Tang H, Liang B, Zhang S Y, de Boer F R, Buschow K H J and Ridwan S 1998 *J. Appl. Phys.* **83** 5945

[10] Hao Y M, Gao Y, Qu J P and Yong Z 2002 *J. Appl. Phys.* **91** 8219

[11] Hao Y M, Gao Y, Wang B W, Qu J P, Li Y X, Hu J F and Deng J Ch 2001 *Appl. Phys Lett.* **78** 3277

[12] Hao Y M, Zhao W and Gao Y 2003 *Acta Phys. Sin.* **52** 2612 (in Chinese)

[13] Andreev A V, Deryagin A V, Zadvorkin S M, Kudrevatykh N V, Levitin R Z, Moskalev V N, Popov Yu F and Yumaguzhin R Yu in: 1985 *Fizika Magnitnykh Materialov* (Physics of Magnetic Materials) ed Mishin D D (Kalinin, USSR: Kalinin University) p21 (in Russian)