MAGNETIC PHASE TRANSITIONS AND MAGNETIC PROPERTIES OF $Nd_3(Fe, Mo)_{29}$ COMPOUND AND ITS NITRIDE①

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ABSTRACT Iron-rich ternary intermetallic compound $Nd_3(Fe, Mo)_{29}$ with the $Nd_3(Fe, Ti)_{29}$-type monoclinic structure and its nitride were prepared. The intrinsic magnetic properties, magnetic phase transitions, and the magnetocrystalline anisotropy, have been investigated. The results show that the Curie temperature of $Nd_3(Fe, Mo)_{29}$ nitride is 675 K, which is 70.9% higher than that of parent compound (395 K), the saturation magnetization σ_S and anisotropy fields H_A are 151.0 Am^2/kg and 14.0 T at 4.2 K, 116.0 Am^2/kg and 1.6 T at 300 K for $Nd_3(Fe, Mo)_{29}$ compound, 161.0 Am^2/kg and 18.0 T at 4.2 K, 143.5 Am^2/kg and 4.2 T at 300 K for $Nd_3(Fe, Mo)_{29}N_X$ nitride, respectively; the magnetocrystalline anisotropy of the $Nd_3(Fe, Mo)_{29}$ compound is planar above about 230 K and easy cone from 4.2 K to 230 K, after nitrogenation, the spin reorientation phenomenon was no longer observed.

Key words magnetic properties magnetic phase transitions $Nd_3(Fe, Mo)_{29}$ $Nd_3(Fe, Mo)_{29}$

1 INTRODUCTION

Since the discovery of the novel ternary rare earth iron-rich intermetallic compound with the nominal composition $Nd_3(Fe_{0.95}Ti_{0.05})_{29}$[1], a lot of studies dealing with the structure and magnetic properties of this phase have been reported[2-22]. Firstly, Li *et al*[6] and Hu *et al*[11] reported that the structure of the $Nd_3(Fe, Ti)_{29}$ compound is monoclinic with the space group $P2_{1/c}$. Recently, Kalogirou *et al*[13] pointed out that this kind of compound belongs to the space group $A_{2/m}$. The complexity of the structural arrangement of the 3∶29 phase has led to contradictory reports concerning the kind of magnetocrystalline anisotropy of the $Nd_3(Fe, Ti)_{29}$ compound, based on X-ray diffraction (XRD) data of an aligned sample. Collocott *et al*[1] initially proposed that this compound exhibits planar anisotropy. However, they had indexed the XRD patterns of $Nd_3(Fe, Ti)_{29}$ compound based on an hexagonal structure. Hu *et al*[11] predicted, from neutron diffraction data using a monoclinic description, that at room temperature the moments lie along the *a* axis but the possibility of a small component along the *b* axis is not excluded. In another paper, the same authors have reported that the easy direction of this compound is found to change from the *a* axis at room temperature to the *a*—*b* plane at 12.5 K. A magnetic phase transition at 230 K, first reported by Morellon *et al*[8] by means of a. c. susceptibility measurement, has been explained in terms of a spin reorientation (SRT) from planar to easy

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cone anisotropy. Recently, Kalogirou *et al*[13] reported the presence of an easy cone anisotropy at room temperature in the $Nd_3(Fe, Ti)_{29}$ compound. The purpose of this work is to investigate the magnetocrystalline anisotropy phenomena in the $Nd_3(Fe, Mo)_{29}$ compound and its nitride.

2 EXPERIMENTAL

Ingots of $Nd_3(Fe_{0.956}Mo_{0.044})_{29}$ compound were prepared by argon arc melting using starting elements of purity at least 99.9% with an excess amount of Nd element, to compensate for the loss during melting[18]. Ingots were melted in the water copper hearth and remelted at least five times for homogenization. The ingots were annealed at 1 453 K for 48 h under argon atmosphere, then quenched in water. In order to prepare the nitride, the ingots were pulverized into fine powders with an average particle size of 10 ~ 15 μm, which were then nitrogenated by heating in nitrogen atmosphere of 100 kPa at a temperature of 818 K for about 2.5 h. The nitrogen content of the powder was determined from the mass difference before and after nitrogenization.

X-ray diffraction with CuK_α radiation was used to identify the phases of the compounds and to determine the lattice parameters. The thermomagnetic analysis (TMA) was performed in a low field of about 0.04 T in the temperature range from 300 K to above the Curie temperature. The Curie temperatures T_C were determined from $\sigma^2 - T$ plots by extrapolating σ^2 to zero. The magnetization curves were measured by extracting sample magnetometer (ESM) with a superconducting magnet with maximum magnetic field up to 7T. Saturation magnetization σ_S were derived from $\sigma - 1/B$ plots based on the magnetization curves. Anisotropy field H_A were determined from the intersection point of two magnetization curves measured in the magnetic field applied parallel and perpendicular, respectively, to the alignment direction of the cylinder samples. The a.c. susceptibility measurements in the temperature range 50~ 300 K on polycrystalline powder samples were carried out. ^{57}Fe-Mössbauer spectra on magnetically aligned sample of the $Nd_3(Fe, Mo)_{29}$ compound were obtained at 50, 100, 150, 200, 250 and 300 K on a conventional constant acceleration spectrometer with ^{57}Co(Rh) source moving at room temperature while the absorber was at the desired temperature. The direction of X-rays was parallel to the alignment direction of the $Nd_3(Fe, Mo)_{29}$ compound.

3 RESULTS AND DISCUSSION

The unit cell parameters a, b, c, β and unit cell volume V is 10.638 Å, 8.583 Å, 9.748 Å, 96.86° and 884.0 Å for $Nd_3(Fe, Mo)_{29}$ and 10.856 Å, 8.753 Å, 9.924 Å, 96.72° and 936.4 Å^3 for $Nd_3(Fe, Mo)_{29}N_X$ nitride, respectively. The unit cell volume V of investigated nitride is 5.9% greater than that of parent compound. The nitrogen content X is about 3.9, which is similar to that of the $Nd_3(Fe, Ti)_{29}$ nitride.

Fig. 1 shows the thermomagnetic curves of $Nd_3(Fe, Mo)_{29}N_X$ nitride (a) and parent compound (b). The Curie temperature of $Nd_3(Fe, Mo)_{29}$ nitride is 675 K according to Fig. 1, which is 70.9% higher than that of parent compound (395 K). It is higher than that of $NdFe_{10.7}Mo_{1.5}N_X$ nitride[24] but lower than that of $Nd_2Fe_{17}N_X$ nitride[25]. The strong increase in Curie temperature T_c upon nitrogenization may partly be explained in terms of lattice expansion of the nitride which leads to an increase in the average nearest-neighbor Fe-Fe exchange interaction. A theoretical analysis shows that the increase in Curie temperature T_c may also be ascribed to the increase in magnetization upon nitrogenization and the decrease in the spin up density of states at the Fermi level E_F associated with narrowing of the 3d band[26].

Table 1 shows the saturation magnetization σ_s and anisotropy field H_A of $Nd_3(Fe, Mo)_{29}$ and its nitride measured at 4.2, 50, 100, 150, 200, 250 and 300 K. The saturation magnetization σ_S are 151.0 Am^2/kg at 4.2 K and 116.0 Am^2/kg at 300 K for $Nd_3(Fe, Mo)_{29}$ compound, and 161.0 Am^2/kg at 4.2 K and 143.5 Am^2/kg at

300 K for $Nd_3(Fe, Mo)_{29}N_X$ nitride, respectively. The saturation magnetizations of nitride are about 6.6% (at 4.2 K) and 23.7% (at 300 K) higher than those of parent compound. The increase of saturation magnetization σ_S is attributed to an increase of average Fe moments after nitrogenization[26]. The saturation magnetization σ_S of investigated nitride is also higher than that of corresponding 1∶12 nitride[24] but similar to that of corresponding 2∶17 nitride[25]. It can be seen, from Table 1, that the saturation magnetization σ_S of both $Nd_3(Fe, Mo)_{29}$ compound and its nitride decrease with increase of temperatures, but the σ_S of $Nd_3(Fe, Mo)_{29}$ compound decreases faster than that of nitride.

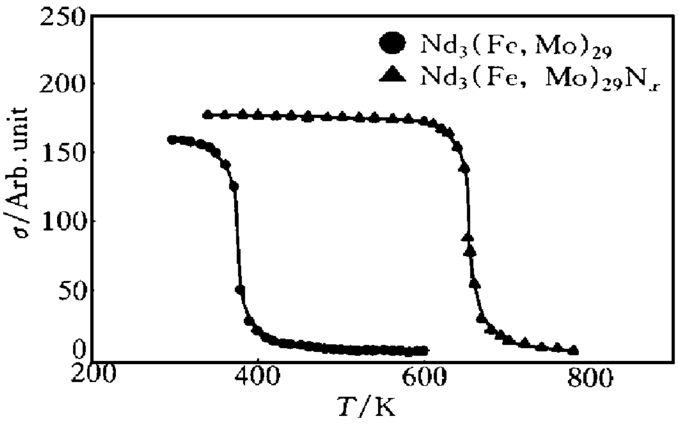

Fig. 1 Magnetization as a function of temperature for $Nd_3(Fe, Mo)_{29}N_X$ nitride compared with that of parent compound at magnetic field of 0.04 T

Table 1 Saturation magnetization σ_s and anisotropy field H_A of $Nd_3(Fe, Mo)_{29}$ and its nitride measured at 4.2, 50, 100, 150, 200, 250 and 300 K

Temperature / K	$Nd_3(Fe, Mo)_{29}$		$Nd_3(Fe, Mo)_{29}N_X$	
	σ_S/ (Am^2kg^{-1})	H_A/T	σ_S/ (Am^2kg^{-1})	H_A/T
4.2	153.0	14.0	161.0	18.0
50	152.5	12.51	60.0	17.0
100	148.0	11.5	157.1	15.5
150	142.0	8.4	154.3	12.4
200	133.0	6.6	148.5	10.3
250	125.0	4.5	146.0	8.0
300	115.5	1.6	143.0	4.2

Fig. 2 shows the X-ray diffraction patterns of the magnetically aligned samples for $Nd_3(Fe, Mo)_{29}$ (a) at room temperature. For comparison, the X-ray diffraction patterns of magnetically aligned samples of the $Y_3(Fe, Mo)_{29}$ at 300 K are given in Fig. 2(b). Both of the X-ray diffraction patterns are very similar, which indicates that the magnetocrystalline anisotropy of the $Nd_3(Fe, Mo)_{29}$ compound is planar. The X-ray pattern of magnetically aligned sample of the nitride likes that of parent.

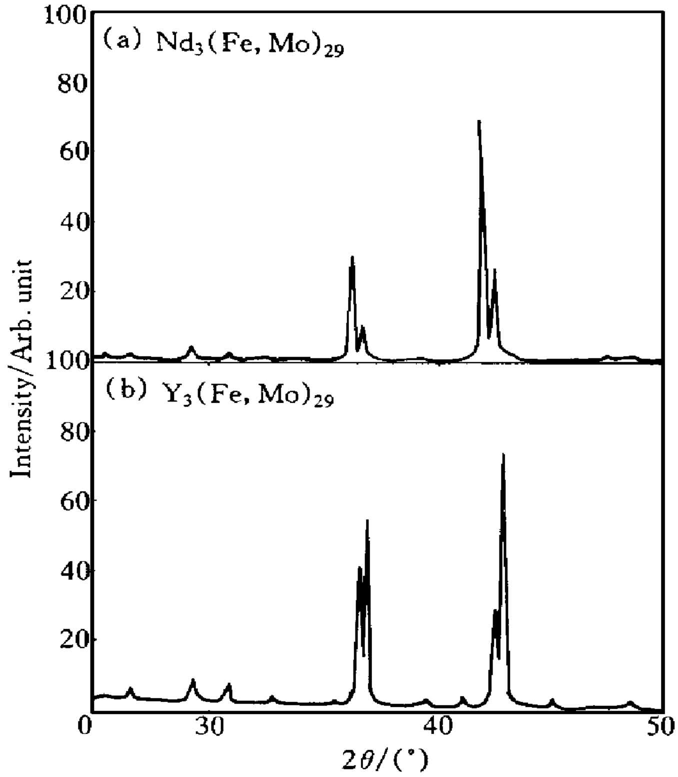

Fig. 2 X-ray diffraction patterns of magnetically aligned samples of $Nd_3(Fe, Mo)_{29}$(a) and $Y_3(Fe, Mo)_{29}$(b)

The temperature dependence of planar anisotropy field H_A for $Nd_3(Fe, Mo)_{29}$ compound and its nitride are also listed in Table 1. At the same temperature, the anisotropy field H_A of $Nd_3(Fe, Mo)_{29}N_X$ nitride is higher than that of parent compound.

Fig. 3 shows the temperature dependence of the a.c. magnetic susceptibility of the $Nd_3(Fe, Mo)_{29}$ compound in the temperature range from 4.2 K to 300 K. It exhibits an anomalous peak at about 230 K, which is attributed to spin reorientation phenomenon.

The Mössbauer spectra of magnetically

aligned powder samples of $Nd_3(Fe, Mo)_{29}$ were measured at 50, 100, 150, 200, 250 and 300 K, and are presented in Fig. 4. The fit was performed with four components with an area ratio of 1∶2∶1.5∶1.2 with respect to the population of these Fe site group and taking into account the Mo occupancy according to Ref. [23]. A fifth component was introduced for α-Fe. The fit yields an average hyperfine field of 26.4, 25.5, 24.7, 23.3, 21.9, and 19.2 T at 50, 100, 150, 200, 250, and 300 K, respectively. The average hyperfine field decreases with increasing temperature. The marked decrease of the average hyperfine field between 250 K and 300 K confirms that a spin reorientation takes place in this temperature region. This result only shows a small difference when compared with that observed for the a. c. magnetic susceptibility measurements.

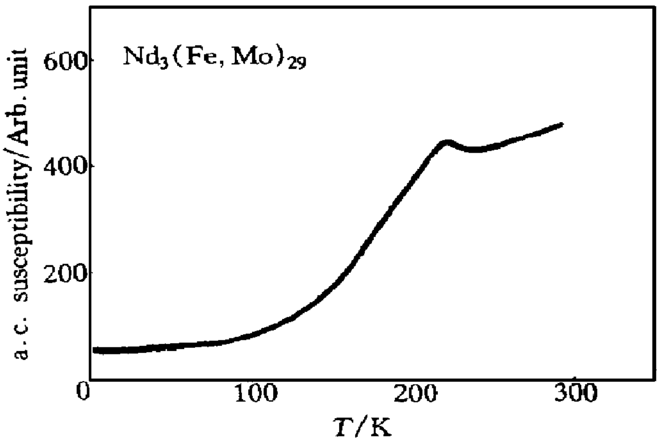

Fig. 3 Temperature dependence of a. c. magnetic susceptibility of $Nd_3(Fe, Mo)_{29}$ in temperature range from 4.2 K to 300 K

Fig. 5 shows the thermomagnetic curve of the $Nd_3(Fe, Mo)_{29}$ compound. It can be seen that this compound has an easy cone anisotropy in the temperature range from 4.2 K to 230 K. Therefore, the spin reorientation that occurs at 230 K is from an easy cone to a planar anisotropy.

Fig. 6 shows the temperature dependence of the a. c. magnetic susceptibility of the $Nd_3(Fe, Mo)_{29}$ nitride in the temperature range from 4.2K to 300K. It can be seen that the spin reorientation phenomenon is no longer observed after nitrogenization. The X-ray diffraction pattern of the magnetically aligned sample of the $Nd_3(Fe, Mo)_{29}N_X$ compound at room temperature, is very similar to that of its parent compound, which indicates that the magnetocrystalline anisotropy of the $Nd_3(Fe, Mo)_{29}$ nitride is also planar.

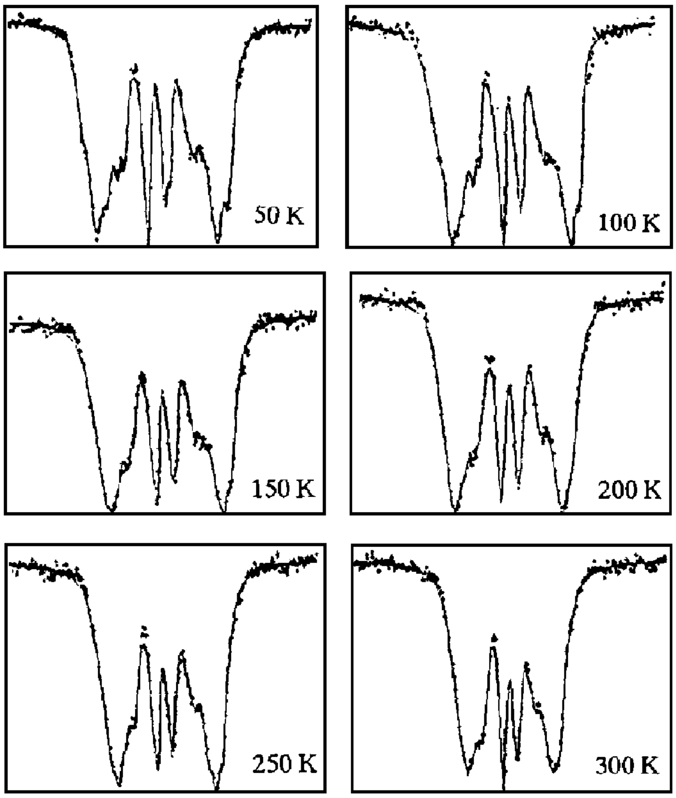

Fig. 4 Fitted Mössbauer spectra at 50, 100, 150, 200, 250, and 300 K of magnetically aligned powder samples of $Nd_3(Fe, Mo)_{29}$

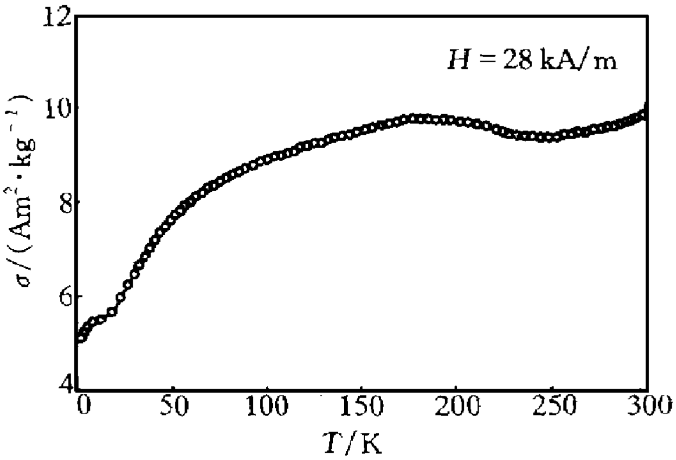

Fig. 5 Magnetization as a function of temperature for $Nd_3(Fe, Mo)_{29}$ compound

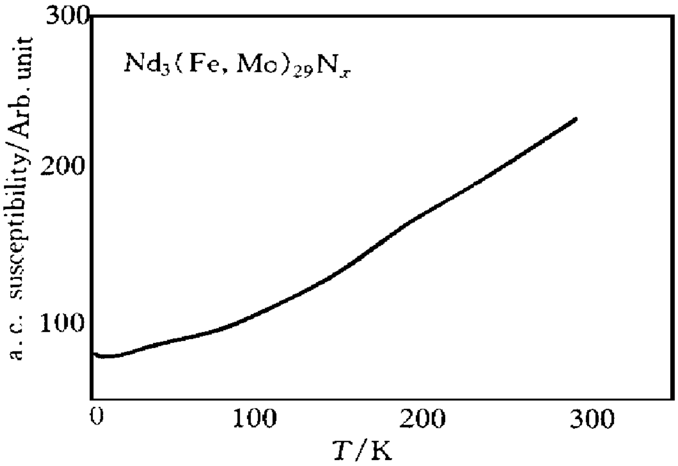

Fig. 6 Temperature dependence of a. c. magnetic susceptibility of $Nd_3(Fe, Mo)_{29}N_X$ in temperature range from 4. 2 K to 300 K

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