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Effects of Gd substitution on the metamagnetic transition of $NdCo_{9.5}V_{2.5}$

X. Hai [a], W.F. Liu [a,*], P. Wu [a], Y.F. Gong [a], G.Y. Liu [b], G.H. Rao [c]

[a] Department of Applied Physics, Institute of Advanced Materials Physics, Tianjin Key Laboratory of Low Dimensional Materials Physics and Preparing Technology, Faculty of Science, Tianjin University, Tianjin 300072, China
[b] Beijing National Laboratory for Condensed Matter Physics, Institute of Physics, Chinese Academy of Sciences, Beijing 100190, China
[c] Department of Information Materials Science and Engineering, Guilin University of Electronic Technology, Guilin, Guangxi 541004, China

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ABSTRACT

Crystal structure and magnetic properties of the $Nd_{1-x}Gd_xCo_{9.5}V_{2.5}$ compounds (x=0–1) have been investigated. The compounds crystallize in the tetragonal $ThMn_{12}$ structure with space group $I4/mmm$. The lattice parameters a, c and the unit cell volume V decrease with the increase of Gd content. The Curie temperature of $Nd_{1-x}Gd_xCo_{9.5}V_{2.5}$ increases monotonically upon the substitution of Gd for Nd. The critical field H^L_{Crit} required to induce the magnetization jump decreases with increasing Gd content. The metamagnetic transition phenomenon becomes less obvious and even disappears with the substitution of Gd for Nd. The H^H_{Crit} required for the metamagnetic transition increased with increasing Gd content at low substitution content. The saturation moment M_S of the compounds decreases first then increases with the increased Gd content.

1. Introduction

Ternary rare-earth compounds with the tetragonal $ThMn_{12}$ structure belong to the family derived from the RT_5 ($CaCu_5$-type structure, space group $P6/mmm$) and form an important class of materials that find numerous industrial applications in permanent magnets and magneto-optical recording, as well as in the aerospace domain [1,2]. Among these intermetallic compounds, $R(Fe, M)_{12}$ compounds have attracted considerable attention owning to their relatively high Curie temperature and saturation magnetization as well as large uniaxial magnetocrystalline anisotropy at room temperature. In order to understand the magnetism of the $R(Fe, M)_{12}$ compounds and improve their magnetic properties, a series of experiments concerning the substitution of Fe by Co have been undertaken [3,4]. Jurczyk and Christyakov [5] showed that it is also possible to prepare Co rich compounds of the same structure. In general, the contribution of rare-earth metal to magnetic anisotropy energy has opposite signs in $RFe_{12-x}M_x$ and $RCo_{12-x}M_x$ [6]. Recently, we conducted a series of investigations on the ternary rare-earth compounds $R(Fe, Co and V)_{12}$ (R=Y and Nd) [7–10]. Some essential phenomena were observed: (i) a metamagnetization transition phenomenon occurred when a high field up to 140 kOe was applied, accompanied by an increase of the magnetization of about 3.1 μ_B per formula for $NdCo_{9.5}V_{2.5}$ [7]; (ii) Fe substitution for Co in $NdCo_{9.5}V_{2.5}$ could result in a shift of the critical field required for the metamagnetic transition, which may be understood by considering the combined effects of the exchange coupling parameter J_{RT} (between R and T atoms, where T represents the transition metal element) and the T-sublattice moment μ_T [9]; and (iii) nonmagnetic element Y substitution for Nd in $NdCo_{9.5}V_{2.5}$ decreases the critical field due to the weakening of the anisotropy field of R-sublattice. As a continuation of the systematic study on the metamagnetic transition of the compound $NdCo_{9.5}V_{2.5}$, in this paper we report the structure and magnetic properties of $Nd_{1-x}Gd_xCo_{9.5}V_{2.5}$. Since Gd is a magnetic element and the 4f shell is half filled so that its second Steven's factor α_J=0, the substitution of Gd for Nd provides an opportunity for obtaining a deeper understanding of the magnetic properties of the compounds.

2. Experimental details

Samples of polycrystalline $Nd_{1-x}Gd_xCo_{9.5}V_{2.5}$ (x=0, 0.1, 0.3, 0.6, 0.8, 0.9, and 1) were prepared by arc melting the appropriate amounts of the constituent elements Nd, Gd, Co, and V with purity better than 99.9% under a high purity argon atmosphere. Appropriate excess amounts of Nd and Gd were added to compensate for the weight loss during arc melting and subsequent heat treatment. After arc melting, the polycrystalline specimens (ingots) were wrapped in Ta foil, sealed into evacuated quartz tube and annealed at 1373 K for 1 week. To avoid possible phase transition during cooling, the samples were quenched into water. The crystal structure was determined by the

* Corresponding author. Tel.: +86 22 27406852.
E-mail address: wfliu@tju.edu.cn (W.F. Liu).

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X-ray powder diffraction (XRD) technique. The XRD experiments were performed on a Rigaku D/max 2500 diffractometer with CuK_α radiation in the 2θ range (20–120°). A step-scan mode was adopted with a sampling time of 1–2 s and a step width of $2\theta = 0.02°$. The data were further analyzed via the Rietveld refinement technique by using the program Fullprof [11]. The temperature dependence of the ac susceptibility of the samples was measured by a mutual inductance method with a fixed frequency of 240 Hz. Curves of magnetization versus temperature (M–T) for the samples were measured by a SQUID magnetometer in a field of 500 Oe. We have ground the bulk samples into fine powders using agate mortar under a high purity argon atmosphere and the field dependence of the magnetization (M–H) at 5 K of the fine powder was measured using a PPMS.

3. Results and discussion

3.1. Crystallography

The structure of $Nd_{1-x}Gd_xCo_{9.5}V_{2.5}$ ($x=0$, 0.1, 0.3, 0.6, 0.8, 0.9, and 1) was examined by X-ray diffraction and thermo-magnetic analysis. All the samples exhibit single-phase tetragonal $ThMn_{12}$ structure. The observed, calculated, and difference XRD patterns resulting from the Rietveld refinement [12,13] of $Nd_{0.4}Gd_{0.6}Co_{9.5}V_{2.5}$ is shown in Fig. 1. The unit-cell parameters a, c and unit-cell volume V of the compounds are shown in Fig. 2 and listed in Table 1. Gd substitution in the Nd site leads to a decrease in the unit-cell volume from 333.1 Å^3 for $NdCo_{9.5}V_{2.5}$ to 329.8 Å^3 for $GdCo_{9.5}V_{2.5}$. The decrease in unit-cell volume with increasing Gd content consists with the fact that the atomic radius of Gd is smaller than that of Nd. The values of a, c and V for $x=0$, 1 agree well with those reported in [10,14].

3.2. The Curie temperature and exchange interaction

Temperature dependence of the magnetization measured in an applied field of 0.5 kOe is shown in Fig. 3. The Curie temperature T_C was derived at the temperature where dM/dT shows minimum on the high temperature region. The dependence of Curie temperature on the Gd concentration is shown in inset of Fig. 3 and listed in Table 1. Fig. 4 shows the temperature dependence of the ac susceptibility χ_{ac} of $Nd_{1-x}Gd_xCo_{9.5}V_{2.5}$. χ_{ac} exhibits a distinct susceptibility peak near the Curie temperature. According to the Landau theory, the critical peak of χ_m can be derived on the basis of the expression for the susceptibility $\chi(T,H)=1/(C_1+C_3M^2)$, where C_1 and C_3 are the coefficients in the expression for the free energy [14]. The Curie temperature can be obtained from the peak temperature where $C_1(T_C)=0$ and listed in Table 1. T_C derived from M–T curves coincide well with that from the χ_{ac} peak. As listed in Table 1, the Curie temperature increases with increasing Gd content (shown in the inset of Fig. 3), reaching the value of 212 K for $x=1$, which is in good accordance with the value reported in Ref. [15].

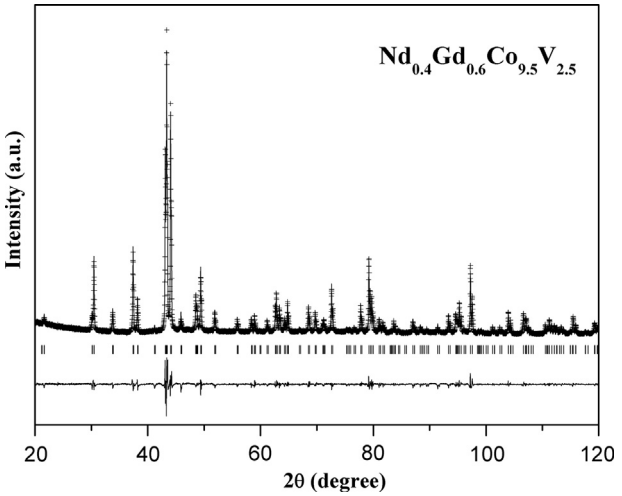

Fig. 1. Observed (crosses) and calculated (solid line) XRD intensities of $Nd_{0.4}Gd_{0.6}Co_{9.5}V_{2.5}$ at room temperature. The vertical lines indicate the Bragg peak positions. The differences between the observed and calculated intensities are shown at the bottom of the figure.

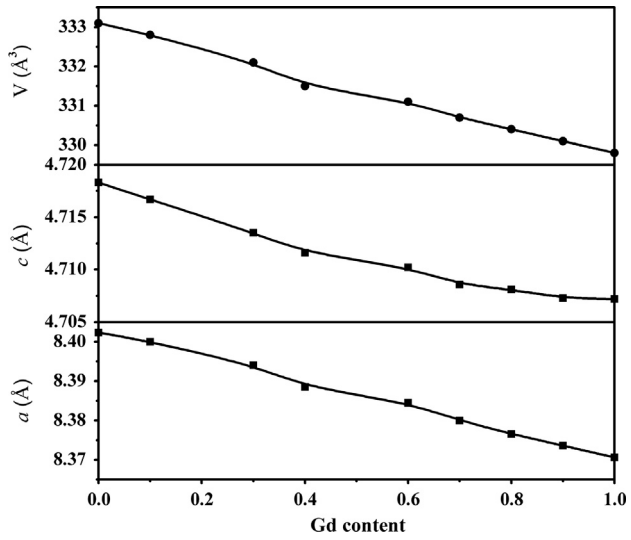

Fig. 2. The Gd content dependence of the lattice parameters a, c and the unit-cell volume V.

Table 1
Structural and magnetic properties of $Nd_{1-x}Gd_xCo_{9.5}V_{2.5}$ compounds, T_C' and T_C were derived from ac susceptibility and M–T curve, respectively.

x	a(Å)	c(Å)	V(Å^3)	T_C(K)	T_C'(K)	M_s(μ_B/fu)	H^L_{Crit}(Oe)
0	8.4024	4.7183	333.1	177	180	7.80	6411
0.1	8.4000	4.7167	332.8	180	182	5.57	6002
0.3	8.3941	4.7135	332.1	190	191	3.05	4036
0.6	8.3845	4.7102	331.1	197	200	1.09	4033
0.8	8.3766	4.7081	330.4	200	205	0.76	1528
0.9	8.3736	4.7073	330.1	204	207	1.0	521
1	8.3706	4.7072	329.8	212	215	2.32	219

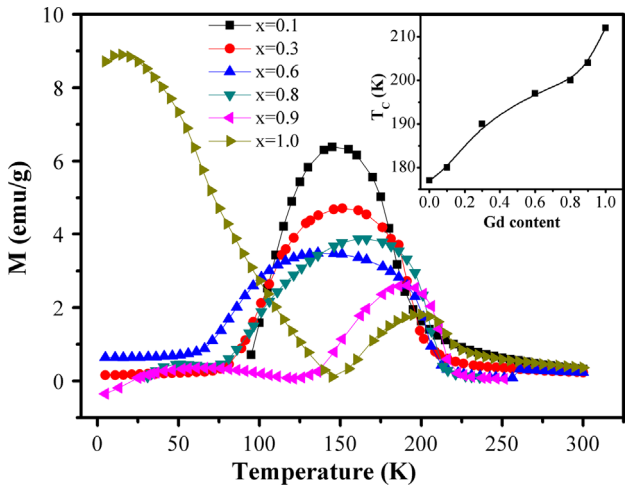

Fig. 3. The temperature dependence of the magnetization for $Nd_{1-x}Gd_xCo_{9.5}V_{2.5}$ compounds. The inset shows concentration dependence of Curie temperature.