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Magnetothermal Properties of $NdCo_{5-x}Ga_x$ Compounds

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Abstract—The magnetization and magnetocaloric effect (MCE) of $NdCo_{5-x}Ga_x$ compounds are measured. It is found that the magnetic ordering temperatures of $NdCo_{5-x}Ga_x$ compounds with Ga substitution fall in proportion to the square of the magnetic moment of the Co sublattice, due to the major contribution from the Co sublattice in the magnetic ordering of these compounds. Reducing the Ga concentration from 1.7 to 1.3 sharply raises the values of both the magnetic ordering temperature and the MCE. With a further decrease in Ga concentration, the magnetic anisotropy of the Co sublattice begins to play an important role, leading to the formation of noncollinear magnetic structures, the emergence of spin-reorientation magnetic phase transitions, and a reduction in the MCE.

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INTRODUCTION

Compounds based on RCo_5 are a wide class of magnetically ordered substances with high values of magnetization and magnetic anisotropy constants. Many such compounds are considered materials for permanent magnets [1, 2]. Attenuating the Co sublattice with *p*-elements allows us to vary the temperatures of the magnetic ordering of these compounds widely while maintaining high values of magnetization. This is of interest in studying magnetic effects observed in the range of critical temperatures (e.g., the magnetocaloric effect (MCE)) [3, 4]. In this work, we present the results from investigating the magnetization and magnetocaloric effect of $NdCo_{5-x}Ga_x$ compounds. The concentration of Ga was chosen such that the temperatures of magnetic ordering of the investigated compounds varied from 130 to 400 K, and the combination of Nd and Co allowed us to obtain compounds with high values of magnetization due to the ferromagnetic interaction between the rare-earth and 3d-sublattices.

EXPERIMENTAL

Our $NdCo_{5-x}Ga_x$ compounds (x = 1.1, 1.3, 1.5, and 1.7) were synthesized by means of electric-arc melting. The samples were melted three times with subsequent annealing over 200 hours at 1070 K. According to X-ray structural analysis data, all compounds were single-phase and had a hexagonal crystal structure of the $CaCu_5$ type (spatial group $P6/mmm$). The homogeneity of the compounds was confirmed via metallography.

RESULTS AND DISCUSSION

We measured (i) magnetization in fields of up to 12 kOe in the 78 to 450 K range of temperatures, and in fields of up to 60 kOe at 4.2 K; and (ii) the magnetocaloric effect directly in fields of up to 13 kOe in the 78 to 400 K range of temperatures.

The main magnetic data for the studied compounds are given in Table 1.

Table 1. Main crystallographic and magnetic data for $NdCo_{5-x}Ga_x$ compounds (x is the concentration of Ga, a and c are the parameters of the crystal lattice, T_C is the Curie temperature, T_{SR} is the temperature of spin-reorientation, μ_s is the spontaneous magnetic moment, μ_{ext} is the magnetic moment of saturation, μ_{Co} is the magnetic moment per one Co atom, and ΔT is the maximum value of the magnetocaloric effect in a field of 13 kOe

x	a, nm	c, nm	T_C, K	T_{SR}, K	μ_s, μ_B	μ_{ext}, μ_B	μ_{Co}, μ_B	ΔT, K at ΔH = 13 kOe
1.1	0.50820(2)	0.40238(2)	376	303	5.9	6.43	0.81	0.11
1.3	0.51015(3)	0.40232(2)	285	–	4.4	5.14	0.51	0.4
1.5	0.51218(2)	0.40298(3)	194	–	4.6	5.11	0.53	0.19
1.7	0.51238(3)	0.40462(2)	131	–	3.6	4.33	0.32	0.18

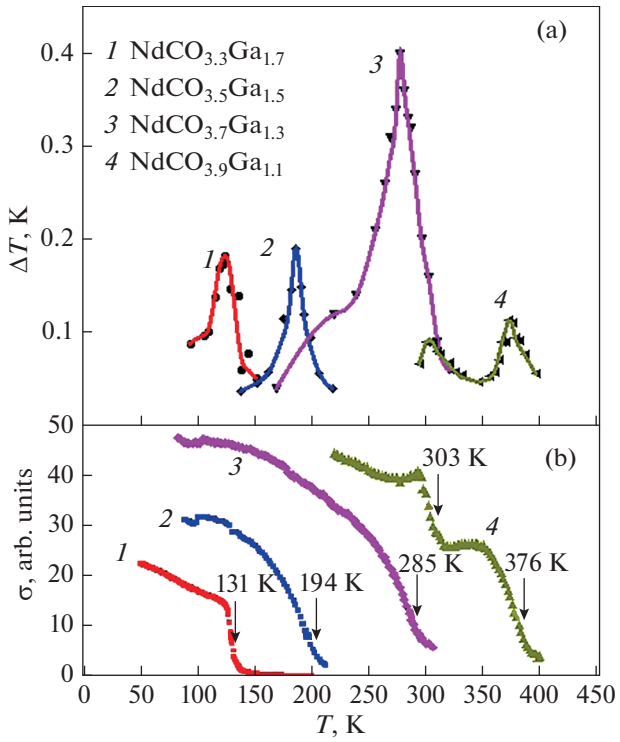

Fig. 1. Temperature dependences: (a) magnetocaloric effect of $NdCo_{5-x}Ga_x$ compounds in a field of 13 kOe; (b) magnetization of $NdCo_{5-x}Ga_x$ compounds in weak magnetic fields.

The temperature dependences of the magnetization demonstrate the ferromagnetic behavior typical of the studied compounds (see Fig. 1). An increase in Ga concentration shifts the Curie temperature toward low temperatures. Two features are observed on the magnetization curve of the $NdCo_{3.9}Ga_{1.1}$ sample. We assume that spin-reorientation occurs at one of the temperatures, due to the competition between the positive exchange energy and the energy of magnetic anisotropy, while the other temperature is the Curie temperature. Similar behavior with a reorientation transition was observed earlier for the closely related $NdCo_4Ga_1$ compound [5].

From the magnetization curves around the Curie temperature, we calculated Belov–Arrott dependences $H/\sigma(\sigma^2)$, whose linear form indicates there were phase transitions of the second kind.

The high anisotropy of the investigated compounds does prevented total saturation of the magnetization curves at 4.2 K in fields up to 6 T. The obtained values of spontaneous magnetization μ_s at 4.2 K appear to be far below the expected values of magnetization μ_{ext} derived from extrapolating the $\mu(1/H)$ curves to zero in the domain of strong fields.

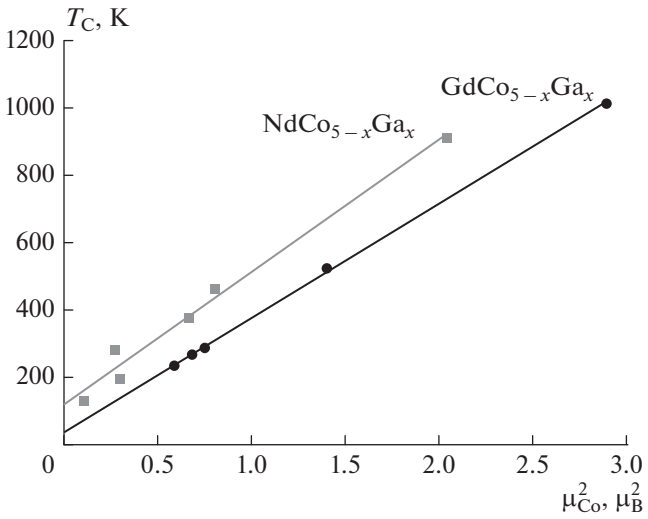

Fig. 2. Dependences of the Curie temperatures of $NdCo_{5-x}Ga_x$ and $GdCo_{5-x}Ga_x$ compounds on the square of the magnetic moment per one Co atom.

Since the exchange energy between the Co and Nd moments in the above compounds is considerably higher than the magnitude of splitting by the crystal field [6], the Nd magnetic moment at low temperatures may be considered approximately equal to the value for a free Nd ion: $\mu_{Nd} = gJ = 3.27\ \mu_B$. Because of the parallel orientation of Nd and Co sublattices, the magnetic moment per one Co atom was calculated as the difference between μ_e and μ_{Nd}, divided by the Co concentration. It is seen that the introduction of Ga not only attenuates the Co sublattice, but also sharply reduces the magnetic moment in Co, as was observed earlier for different substitutions by *p*-elements [3, 4].

Calculations of the molecular field done earlier for $GdCo_{5-x}Ga_x$ compounds showed that coefficient n_{RR} determining the exchange interaction in the rare-earth sublattice is much lower than coefficients n_{RT} and n_{TT} responsible for exchange among the sublattices and inside the 3d-sublattice [4]. In addition, the introduction of Ga has virtually no effect on the value of n_{RT}, but it substantially reduces n_{TT}. For the above compounds, the Curie temperature is proportional to the square of magnetic moment on cobalt: μ^2_{Co}. Dependence $T_C(\mu^2_{Co})$ obtained from our data and [3–5, 7, 8] was also a straight line (Fig. 2).

Linear dependence $T_C(\mu^2_{Co})$ testifies to the major contribution from the Co sublattice to exchange interaction Co–Co. Exchange interaction R–Co makes a small contribution to the Curie temperature, as is apparent from curve $T_C(\mu^2_{Co})$ not passing through zero. The shift of this linear dependence along the *y* axis is due to the contribution from exchange interaction R–Co.

The results from direct MCE measurements are presented in Fig. 1a. Reducing the Ga concentration initially increases the MCE substantially. The magnitude of the MCE is apparently very sensitive to the magnetic moment on Co, which determines the exchange interaction in these compounds. With a further reduction in concentration, however, the role of magnetic anisotropy grows, resulting in the spin-reorientation transition. We therefore observe two maxima on curve ΔT that correspond to the reorientation transition and the Curie temperature, respectively.

CONCLUSIONS

Our investigation of the concentration dependences of magnetization and the MCE showed that the Co sublattice determines the character and temperature of the ordering of these compounds. Attenuation of the Co sublattice by gallium (i) lowers the temperature of magnetic ordering in proportion to the square of the magnetic moment on a cobalt atom and (ii) alters the magnitude of the MCE. The maximum MCE value was observed for a Ga concentration of 1.3.

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