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# MAGNETIC PROPERTIES OF $\text{GdMn}_x\text{Fe}_{1-x}\text{Si}$ INTERMETALLIC COMPOUNDS

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Magnetic properties of  $\text{GdMn}_x\text{Fe}_{1-x}\text{Si}$  ( $x = 1, 0.9, 0.8, 0.7, 0.6, 0$ ) compounds were investigated. Both  $\text{GdMnSi}$  and  $\text{GdFeSi}$  crystallize in the  $\text{CeFeSi}$ -type tetragonal structure and form a complete solid solution without any change in the crystal structure. Magnetic measurements were made in static magnetic fields up to 1 MA/m in the temperature range from 77 K to 420 K. For  $\text{GdFeSi}$  these measurements were carried out on single crystal sample. It was found that the increase in Fe content leads to a sharp decrease in saturation magnetic moment. The Curie temperature increases in the concentration range  $0.7 \leq x \leq 1$  and monotonously decreases for  $x \leq 0.6$ . These effects could be explained by strong dependence of the exchange integrals on the interatomic distances.

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## 1. Introduction

The silicides  $\text{GdFeSi}$  and  $\text{GdMnSi}$  were first synthesized by Bodak et al. [1] and Yarovets et al. [2]. They crystallize in the tetragonal structure of  $\text{CeFeSi}$  type (space group,  $P4/nmm$ ), built of alternating (001) layers with sequence  $\text{R-Si-T}_2\text{-Si-R-R-Si-T}_2\text{-Si-R}$  (R — rare earth, T — transition metal, Si — silicon). This structure is closely related to the  $\text{ThCr}_2\text{Si}_2$ -type structure [3]. However, the magnetic ordering temperatures in  $\text{RTSi}$  compounds are much higher than in corresponding  $\text{RT}_2\text{Si}_2$  series, which makes these compounds more perspective of both fundamental and practical significance.

In our previous studies we investigated the magnetic properties of  $\text{GdMn}_x\text{Co}_{1-x}\text{Si}$  and  $\text{Gd}_x\text{La}_{1-x}\text{TSi}$  (T = Fe, Co) series [4, 5]. The  $\text{RFeSi}$  compounds were found to be ferromagnetic. The iron sublattice carries no magnetic moment in these compounds, in good accordance with neutronographic data for  $\text{RFeSi}$  (R = La–Nd, Tb–Dy) compounds [6]. It is noteworthy that T = Mn is a unique example of a 3d-transition metal with a magnetic moment in  $\text{RMn}_2\text{Si}_2$  [3] and  $\text{RMnSi}$  [7] compounds. The Mn sublattice orders at relatively high temperatures. From this point of view it was interesting to study the  $\text{GdMn}_x\text{Fe}_{1-x}\text{Si}$  series with substitution in 3d-sublattice in order to complete this study and to clarify the role of 3d-sublattice for magnetic properties of these compounds.

## 2. Experimental details and results

The  $\text{GdMn}_x\text{Fe}_{1-x}\text{Si}$  ( $x = 1, 0.9, 0.8, 0.7, 0.6, 0.4, 0$ ) alloys were prepared by induction melting of the components (of purity not worse than 99.9%) in an argon atmosphere. The purity of the samples was checked by X-ray diffraction. It was found that all samples were of single phase with the tetragonal  $\text{CeFeSi}$  structure. The lattice constants were determined using power diffraction patterns (DRON-3M diffractometer with  $\text{Cu } K_\alpha$  radiation).  $\text{GdFeSi}$  single crystals were detected by Laue reflection method. The magnetic measurements were made in the static magnetic fields up to 1 MA/m in the temperature range from 77 to 600 K (for example, see Figs. 1, 2). The experimental method was mainly described earlier in Ref. [8].

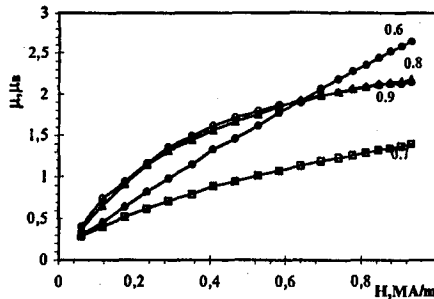


Fig. 1. Magnetization curves of  $\text{GdMn}_x\text{Fe}_{1-x}\text{Si}$  compounds at 78 K.

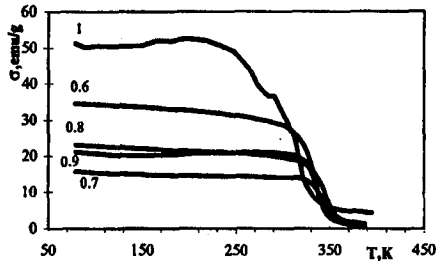


Fig. 2. Temperature dependences of the magnetization of  $\text{GdMn}_x\text{Fe}_{1-x}\text{Si}$  compounds at 0.16 MA/m.

The magnetic and crystal data for  $\text{GdMn}_x\text{Fe}_{1-x}\text{Si}$  compounds are summarized in Table. It was found that  $\text{GdMnSi}$  is a ferrimagnetic with opposite orientation of magnetic moments of Gd and Mn sublattices. The magnetic curve is characterized by the Curie temperature at 316 K and an inflection point at  $\approx 270$  K, which is in good agreement with [7]. The manganese magnetic moment at 4.2 K is near  $1.63\mu_B$  [8].

In  $\text{GdMn}_x\text{Fe}_{1-x}\text{Si}$  compounds the lattice parameters decrease with substitution of Mn. The increase in Fe content leads to a sharp decrease in saturation magnetic moment (for  $x > 0.6$ ) (see Table). The Curie temperature increases in the concentration range  $0.7 \leq x \leq 1$  and monotonously decreases for  $x \leq 0.6$ .

TABLE

Unit-cell parameters, magnetic ordering temperature  $T_C$ , paramagnetic Curie temperature  $\theta_p$  and saturated (at  $T = 77$  K and  $H = 1$  MA/m) magnetic moments of  $GdMn_xFe_{1-x}Si$  compounds.

$x$	$a$ [Å]	$c$ [Å]	$V$ [Å <sup>3</sup> ]	$T_C$ [K]	$\mu_0$ [K]	$\theta_p$ [ $\mu_B$ ] 77 K
1	4.024	7.180	116.26	317	316	4.2
0.9	4.020	7.170	115.87	337	328	1.3
0.8	4.011	7.168	115.32	345	317	1.3
0.7	4.001	7.170	114.78	348	305	0.5
0.6	3.983	7.140	113.27	335		0.1
0.4	3.983	7.070	112.16	309		0.8
0	3.996	6.817	108.85	124	125	5.5

GdFeSi was found to be classical ferromagnetic with  $T_C = 125$  K. The easy magnetization direction is along  $c$  axis. The anisotropy field  $H_a$  is near 0.3 MA/m. The X-ray data [5] obtained for GdFeSi show that the magnetic ordering in the temperature region  $T < T_C$  for this compound is accompanied by a sharp increase in the lattice parameter  $c$  and small changes of the parameter  $a$ . The value of the magnetic moment of GdFeSi at  $T = 4.2$  K and  $H = 20$  MA/m is close to that expected for the  $Gd^{3+}$  free ion [5]. This shows the lack of the magnetic moment on iron in this compound.

### 3. Discussion

To understand the magnetic behavior of RTX compounds it is necessary to consider several types of exchange interactions: T-T, R-R and R-T. The magnetic measurements performed on the isotypic  $GdMn_xFe_{1-x}Si$  and  $GdMn_xCo_{1-x}Si$  [4] series show very similar behavior. The absence of saturation in the magnetization curves for these compounds with substitution of Mn by Fe or Co may be connected to the shift of the  $3d$ -subbands of the "up" spins and the "down" spins, under an action of a magnetic field which according to the model [9] can lead to monotonous increase in magnetization.

The increase in Fe content of  $GdMn_xFe_{1-x}Si$  compounds results in a decrease in lattice parameters  $a$ ,  $c$  and volume  $V$ . Therefore an increase in the overlap of  $3d$  electron wave functions can be expected, which results in increase in the R-T and T-T exchange integrals and hence in increase in the Curie temperature for  $x \geq 0.7$ . On the contrary, the abrupt decrease in  $T_C$  for  $x < 0.7$  ( $T_C = 335$  K for  $x = 0.6$ ,  $T_C = 124$  K for  $x = 0$ ) is probably caused by a decrease in the resulting magnetic moment in these compounds since Fe has no magnetic moment in these compounds. The high positive values of paramagnetic Curie temperatures in these compounds suggest that the ferromagnetic intralayer interactions are dominant. The present study suggests the strong dependence of R-T and T-T exchange integrals on the interatomic distances.

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