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Antiferromagnetic properties in RFe_2Ge_2 ($R = Tb, Dy, Ho$)

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Abstract. X-ray and neutron diffraction measurements confirm that RFe_2Ge_2 ($R = Tb, Dy$ and Ho) compounds show the tetragonal $ThCr_2Si_2$ type of crystal structure. Magnetization and magnetic susceptibility data indicate antiferromagnetic behaviour below the Néel temperatures at 8.5 K in $TbFe_2Ge_2$ and 3.35 K in $DyFe_2Ge_2$. A metamagnetic transition at $H_c = 11$ kOe and $T = 4.2$ K has been detected in $TbFe_2Ge_2$. The other two compounds give at 4.2 K magnetization curves characteristic for paramagnetic materials up to $H = 50$ kOe. The magnitudes of magnetic moments at $H = 50$ kOe are smaller than the free ion values for all three compounds. $TbFe_2Ge_2$ and $DyFe_2Ge_2$ exhibit at 1.5 K modulated antiferromagnetic order described by two component wavevectors $\mathbf{k} = |k_x, 0, k_z|$. The magnetic moments are localized on the lanthanide ions and amount at 1.5 K to 7.68(11) and 7.3(3) μ_B in $TbFe_2Ge_2$ and $DyFe_2Ge_2$ respectively. Diffuse magnetic peaks observed at 1.5 K in the neutron diffraction pattern of $HoFe_2Ge_2$ indicate that a long-range antiferromagnetic order starts to set up at this temperature.

1. Introduction

Ternary lanthanide–iron–germanium (or silicon) phases with 1:2:2 stoichiometry crystallize in the tetragonal $ThCr_2Si_2$ -type structure [1] and show complex magnetic ordering schemes at low temperatures [2–7]. Magnetization measurements carried out for $TbFe_2Ge_2$ and $DyFe_2Ge_2$ have revealed that both are antiferromagnetic at low temperatures [8, 9]. No magnetization data are available for $HoFe_2Ge_2$. Neutron diffraction results reported for $TbFe_2Ge_2$ [10] confirmed the existence of magnetic ordering at low temperatures; however, the magnetic structure has not been solved.

Continuing our studies on the magnetic properties of RFe_2X_2 ($R =$ lanthanide element, $X = Si, Ge$) phases, we present in this paper the results of neutron diffraction experiments performed on polycrystalline samples of $TbFe_2Ge_2$, $DyFe_2Ge_2$ and $HoFe_2Ge_2$ supplemented by new data obtained in the course of magnetization measurements.

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2. Experimental details

The samples were obtained by arc melting of the constituent elements taken in the stoichiometric ratio 1:2:2. The purity was 3N (lanthanide and iron) and 5N (germanium). The melting was performed in purified argon atmosphere. The samples were afterwards annealed at 800 °C for 100 h. X-ray patterns (Fe K α radiation) showed that all were single phases with the tetragonal ThCr₂Si₂-type structure.

Magnetization and magnetic susceptibility measurements were performed in the temperature range 2 to 300 K using a vibrating sample magnetometer and SQUID magnetometer. In the latter experiment the maximum external magnetic field strength was 50 kOe.

Neutron diffraction measurements were carried out in the temperature range from 1.5 to 15.2 K on the E6 instrument at the BER II reactor (Hahn-Meitner Institute, Berlin) with the incident neutron wavelength of 2.422 Å. The data were processed by the Rietveld method using the FULLPROFF program [11] with neutron scattering lengths taken from [12] and the R³⁺ form factors adopted following [13].

The ⁵⁷Fe Mössbauer effect measurements were performed in transmission geometry using a constant-acceleration-type spectrometer with ⁵⁷Co source in a Cr matrix. The low-temperature measurements at 80 and 4.2 K were made using a ‘top-loading’-type cryostat.

3. Results

3.1. Crystal structure

Neutron diffractograms of all title compounds recorded above their Néel points contain reflections obeying the rule $h + k + l$ even which is characteristic for the ThCr₂Si₂-type structure (space group $I4/mmm$) [14]. The atoms are located in the following positions:

- (i) R atoms in 2(a) 0, 0, 0;
- (ii) Fe atoms in 4(d) 0, $\frac{1}{2}$, $\frac{1}{4}$; $\frac{1}{2}$, 0, $\frac{1}{4}$;
- (iii) Ge atoms in 4(e) 0, 0, z ; 0, 0, \bar{z} ;
- (iv) body centring translation.

The free z parameter for the Ge atom was refined and is listed in table 1, which contains also the lattice parameters determined at temperatures a few degrees above the respective Néel points.

Table 1. Crystal structure parameters.

	TbFe ₂ Ge ₂ $T = 15.2$ K	DyFe ₂ Ge ₂ $T = 8$ K	HoFe ₂ Ge ₂ $T = 6$ K
a (Å)	3.9705(14)	3.929(6)	3.9278(14)
c (Å)	10.3733(35)	10.230(24)	10.2755(53)
c/a	0.3828(2)	0.3826(26)	0.3807(4)
V (Å ³)	163.53(17)	158.57(82)	159.18(29)
z	0.3826(9)	0.3766(30)	0.3840(10)
R_{Bragg} (%)	5.4	6.7	5.5
$R_{prof.}$ (%)	5.6	5.6	5.8

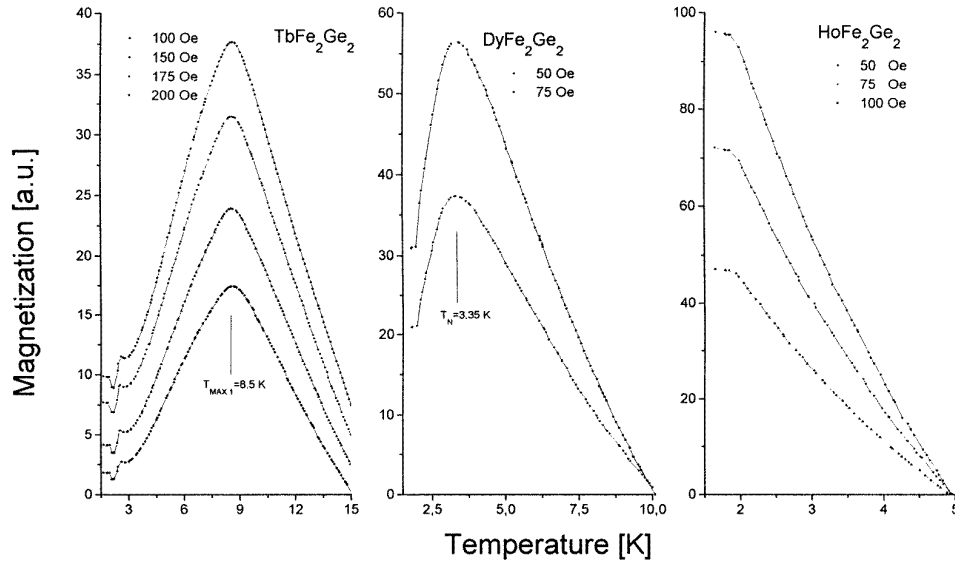


Figure 1. Magnetization curves for $TbFe_2Ge_2$, $DyFe_2Ge_2$ and $HoFe_2Ge_2$ obtained at low temperatures.

3.2. Magnetic data

Magnetization curves recorded at low temperatures in the presence of magnetic fields from 50 to 200 Oe (see figure 1) indicate the transition to the paramagnetic state at 8.5 K in $TbFe_2Ge_2$ and 3.35 K in $DyFe_2Ge_2$, while $HoFe_2Ge_2$ remains paramagnetic at 2 K. Figure 2 illustrates how the magnetization curves recorded at 4.2 K behave as the magnetic field changes from 0 to 50 kOe. The curve obtained for $TbFe_2Ge_2$ indicates the presence of a critical field $H_c = 11$ kOe connected most probably with a metamagnetic transition. The two remaining curves are characteristic of paramagnetic materials. At $H = 50$ kOe and $T = 4.2$ K the magnetic moments localized on the lanthanide ions amount to $5.3 \mu_B$ for $TbFe_2Ge_2$ and $6.3 \mu_B$ for $DyFe_2Ge_2$ and $HoFe_2Ge_2$ i.e. they are much smaller than the respective free ion values.

3.3. Magnetic structures

Figure 3 shows the neutron diffraction pattern in $TbFe_2Ge_2$ recorded at the temperature of 1.5 K and the corresponding diagram showing the difference between observed intensities and those calculated for this assumed structure. The intensities of peaks of magnetic origin were calculated for this assumed structure. The modulated magnetic order is described by the propagation vector $\mathbf{k} = |0.1536(4), 0, 0.4358(20)|$. The magnetic structure of $TbFe_2Ge_2$ is thus represented by a static linear spin wave displayed schematically in figure 4 with magnetic moment of $7.68(11) \mu_B$ at 1.5 K localized on the Tb^{3+} ion and parallel to the c -axis. No localized magnetic moment on the Fe ion could be detected within the accuracy of a powder neutron diffraction experiment, in agreement with the Mössbauer spectroscopy results shown below. Table 2 lists the observed and calculated magnetic intensities.

The variation of satellite peak 000^\pm intensity with temperature displayed in figure 5 shows that $TbFe_2Ge_2$ does not undergo any magnetic phase transition between 1.5 and

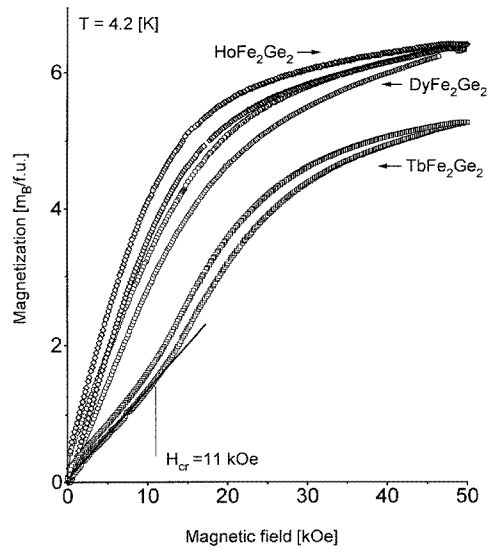


Figure 2. Magnetization curves at 4.2 K as a function of the applied external magnetic field strength.

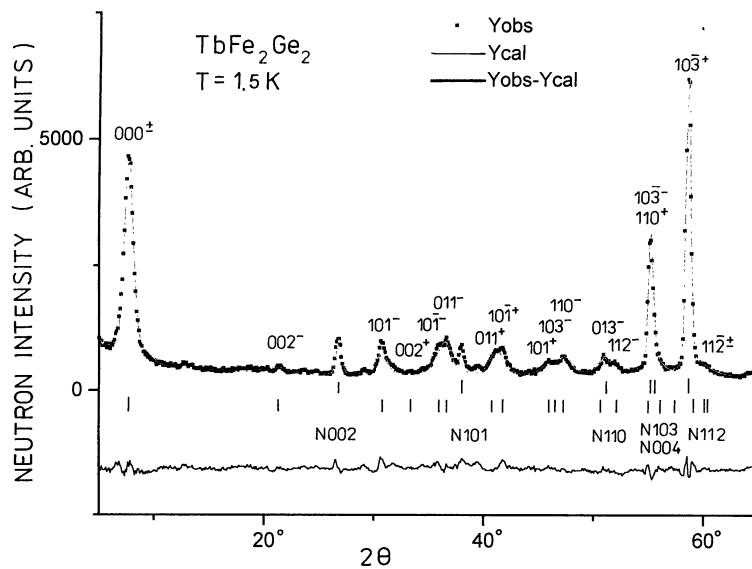


Figure 3. The neutron diffraction pattern of TbFe_2Ge_2 at 1.5 K and the difference pattern.

8.5 K. The latter temperature is the Néel point of TbFe_2Ge_2 (see the inset of figure 5).

The reflections due to magnetic order observed in the neutron diffraction pattern of DyFe_2Ge_2 recorded at 1.5 K (see figure 6) were indexed also for a modulated structure described by the wavevector $\mathbf{k} = |0.209(1), 0, 0.421(4)|$ with a magnetic moment of $7.3(3) \mu_B$ at 1.5 K localized on the Dy^{3+} ion aligned with the c -axis. The comparison of observed and calculated magnetic intensities is given in table 3.

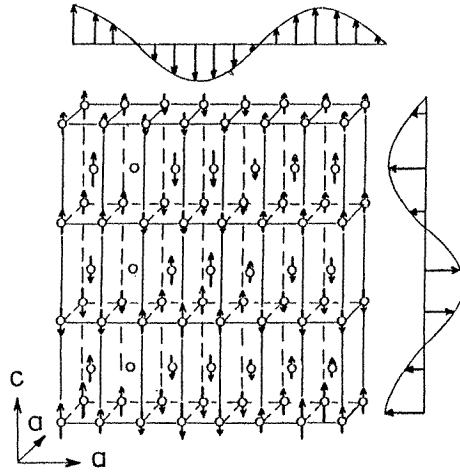


Figure 4. A schematic representation of the magnetic structure of $TbFe_2Ge_2$.

Table 2. Bragg angles and integrated magnetic intensities for $TbFe_2Ge_2$.

hkl	2θ	I_{calc}	I_{obs}
000^\pm	8.0	4086	3994
002^-	21.7	73	49
101^-	31.0	534	698
002^+	33.6	12	24
101^-	36.0	290	375
011^-	36.8	754	868
011^+	41.2	476	544
101^+	42.0	287	369
101^+	46.0	198	203
103^-	46.6	101	101
110^-	47.6	442	366
004	49.5	3	1
013^-	51.0	200	128
112^-	52.2	304	168
103	55.0	95	77
110^+	56.0	310	294
103^+	57.2	44	47
112^-	58.9	186	182
112^+	60.0	232	216
013^+	61.0	96	70
103^+	64.6	40	51

Neutron diffraction patterns of $HoFe_2Ge_2$ recorded at 1.5 K and at a number of temperatures above do not show coherent magnetic reflections (see figure 7). Instead, diffuse scattering is observed near the angles at which magnetic reflections are present in $TbFe_2Ge_2$ and $DyFe_2Ge_2$. The shape of this diffuse peak suggests that long-range antiferromagnetic order in $HoFe_2Ge_2$ starts to set up at 1.5 K.

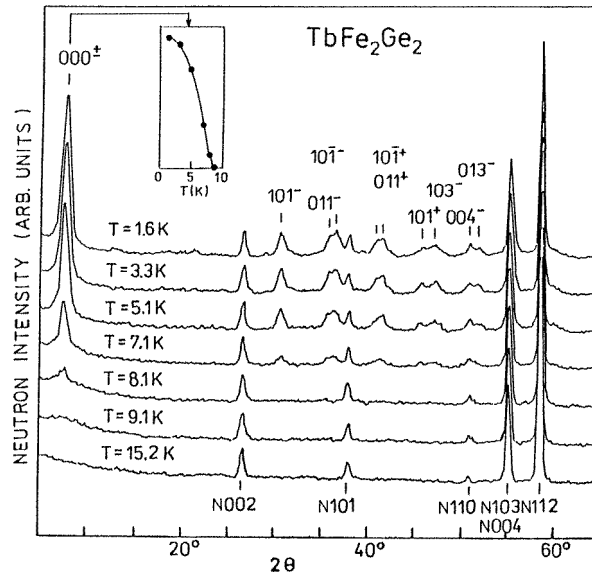


Figure 5. The neutron diffraction patterns of TbFe_2Ge_2 taken at a number of temperatures between 1.6 and 15.2 K. The inset shows how the integrated intensity of the satellite peak 000^\pm varies with temperature, yielding the Néel point at 8.5 K.

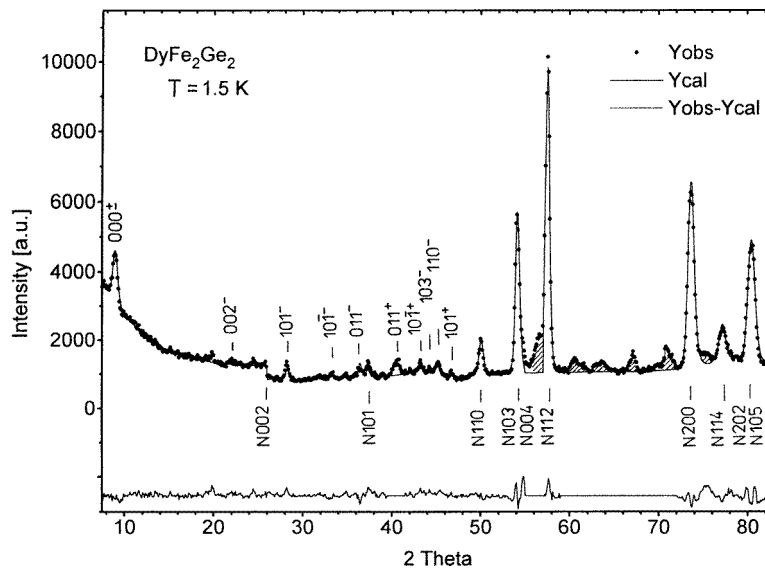


Figure 6. The neutron diffraction pattern of DyFe_2Ge_2 at 1.5 K and the difference pattern between experiment and calculations.

3.4. Results deduced from ^{57}Fe Mössbauer spectra

The ^{57}Fe Mössbauer absorption spectra of TbFe_2Ge_2 recorded at room, liquid nitrogen and liquid helium temperatures have the form of non-resolved quadrupole doublets (see

Table 3. Bragg angles and integrated magnetic intensities for $DyFe_2Ge_2$.

hkl	2θ	I_{calc}	I_{obs}
000^\pm	9.1	1094	1016
002^-	22.1	34	128
101^-	28.5	179	293
002^+	33.1	7	4
101^-	33.4	98	84
011^-	36.6	236	246
011^+	40.5	220	215
101^+	43.4	90	215
103^-	44.5	35	99
110^-	45.3	172	294
101^+	47.0	69	77
004^-	49.1	0	5
112^-	50.1	122	132
013^-	50.5	76	66
103^-	54.5	17	18

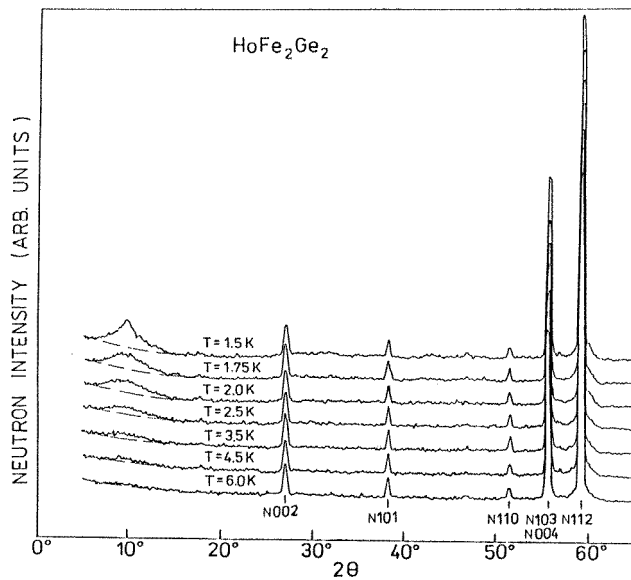
**Figure 7.** Neutron diffraction patterns of $HoFe_2Ge_2$ recorded at temperatures between 1.5 and 6 K.

figure 8). The recorded spectra show that the iron atom does not carry any magnetic moment. The values of the isomer shift and the quadrupole splitting change only slightly with the temperature, in accordance with the results published previously [15]. The liquid helium temperature spectrum does not indicate the presence of any magnetic hyperfine field caused by the magnetic moment localized on the Tb^{3+} ion or by electron spin polarization in the material.

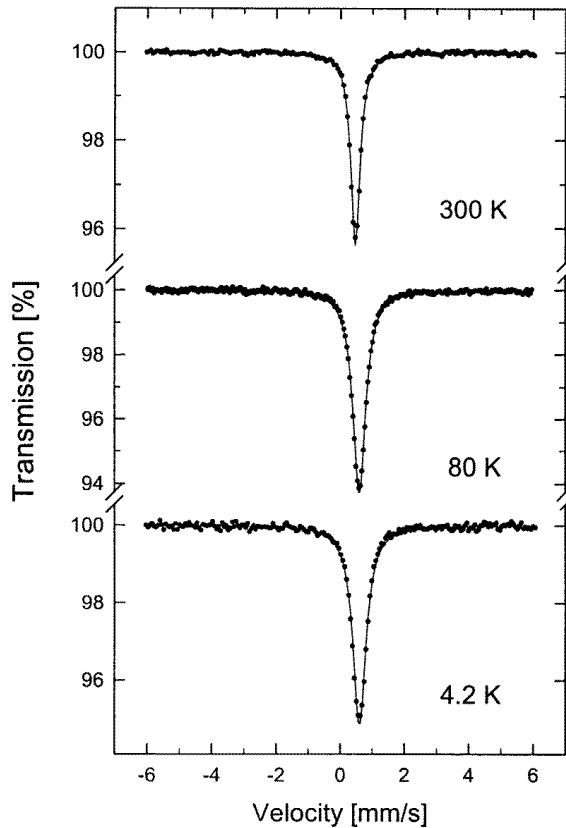


Figure 8. ^{57}Fe Mössbauer spectra of TbFe_2Ge_2 recorded at $T = 4.2, 80$ and 300 K.

4. Comment

The modulated antiferromagnetic structures observed in TbFe_2Ge_2 and DyFe_2Ge_2 imply that the dominant magnetic interactions responsible for their stability may be discussed in terms of the RKKY model.

Although the RFe_2X_2 silicides and germanides crystallize in the same tetragonal ThCr_2Si_2 structure type, the germanides show larger magnitudes of a and c lattice parameters and consequently longer R–R interatomic distances in the (001) plane and along the tetragonal axis. If one connects these experimental observations with the Néel temperatures which are smaller in the germanides (see table 4) the conclusion may be drawn that the exchange interactions operating in the germanides are weaker than those in the silicides.

Since the components of the propagation vectors determined for the RFe_2Ge_2 and RFe_2Si_2 compounds are different (see table 4), one may conclude that this effect is also brought about by the sizes of Ge and Si atoms, which affect the interatomic distances in the crystals.

It is known that the orientation of the magnetic moment with respect to the tetragonal axis is connected with the sign of the B_2^0 coefficient in the crystal electric field Hamiltonian [16]. This coefficient was found to be negative for RFe_2Si_2 ($\text{R} = \text{Tb, Ho}$) compounds [4–6] and was correlated with the observation that the magnetic moments in these compounds are

Table 4. A comparison of the Néel temperatures T_N , the components of the propagation vector (k_x , k_y , k_z) and values of ordered magnetic moments μ observed in RFe_2X_2 silicides (at 4.2 K) and germanides (at 1.5 K).

R	Silicides					Germanides						
	T_N (K)	k_x	k_y	k_z	μ (μ_B)	Ref.	T_N (K)	k_x	k_y	k_z	μ (μ_B)	Ref.
Tb	10.5	0.3226	0	0.1708	7.9(1)	[5]	8.5	0.1536	0	0.4358	7.68(11)	This work
Dy	3.8	0.335	0	0.136	7.52(5)	[4]	3.35	0.209	0	0.421	7.3(3)	This work
Ho	2.3	0.328	0	0.152	7.4(3)	[6]	<1.5					This work

aligned along the tetragonal axis. Since the orientation of magnetic moment in $TbFe_2Ge_2$ and $DyFe_2Ge_2$ observed in this experiment is the same as that in the RFe_2Si_2 compounds one may predict that the B_2^0 coefficient is most probably also negative.

Acknowledgment

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