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Neutron diffraction studies of the magnetic structures of the HoRhGe and ErRhGe compounds

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Abstract. HoRhGe and ErRhGe compounds crystallize in the orthorhombic TiNiSi-type of structure with the space group $Pnma$. Neutron diffraction measurements at $T = 1.6$ K indicate collinear magnetic structures with the propagation vector $\mathbf{k} = (1/2, 0, 1/2)$ for HoRhGe and $\mathbf{k} = (0, 1/2, 0)$ for ErRhGe. After increasing the temperature, the change to the incommensurate sine modulated structure is observed for ErRhGe at $T_i = 5$ K. The temperature dependence of the magnetic peak intensities gives the Néel temperature 4.6 K for HoRhGe and 9.5 K for ErRhGe.

1. Introduction

Ternary equiatomic RTX compounds, where R is a rare earth metal, T is a transition d element and X is a p-electron metal have been intensively investigated in recent years because of their interesting magnetic properties [1].

The RRhGe compounds crystallize in the orthorhombic TiNiSi type of structure [2]. CeRhGe and NdRhGe [3] as well as TbRhGe [4] have been found by neutron diffraction to order antiferromagnetically at low temperatures. The magnetic structure of the CeRhGe compound is collinear whereas that of NdRhGe is described by the wavevector $\mathbf{k} = (1/2, 0, 1/2)$ [3]. At $T = 1.7$ K the magnetic structure of TbRhGe is sine modulated with the wave vector $\mathbf{k} = (0, 0.45, 0.11)$ and at $T = 18$ K it transforms to the magnetic structure described by the wave vector $\mathbf{k} = (0, 0.44, 0)$.

To continue our studies of RTX compounds the neutron diffraction measurements of HoRhGe and ErRhGe compounds were performed and both crystal and magnetic structures were determined.

2. Experiment

HoRhGe and ErRhGe samples were synthesized by arc melting under argon atmosphere using high-purity component elements in the stoichiometric proportion 1:1:1. To obtain a homogeneous material, the samples were annealed at 800 °C for 1 week.

The homogeneity was tested by x-ray diffractometry (Co $K\alpha$ radiation). Both HoRhGe and ErRhGe were found to be single-phase samples. Lattice parameters were determined

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and refined by the least-squares method. They are in fair agreement with the data reported earlier [2]. Neutron diffraction patterns were measured in the temperature range 1.8–11 K using the E6 instrument installed at the BERII reactor at the Berlin Neutron Scattering Centre, Hahn-Meitner Institut in Berlin. The incident neutron wavelength was 2.378 Å. Data processing was performed by the Rietveld-type FULLPROF program [5] using nuclear scattering lengths taken from [6] and the magnetic form factors for Ho^{3+} and Er^{3+} ions from [7].

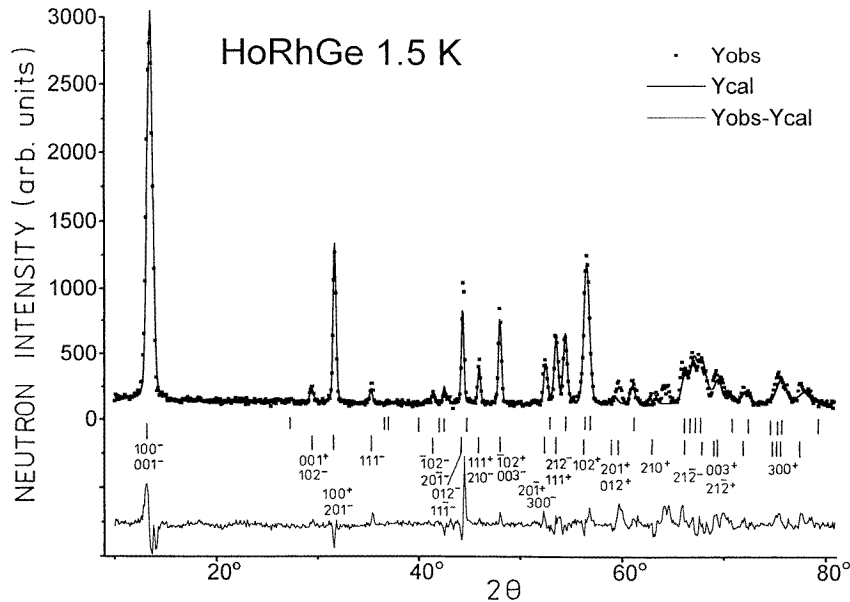


Figure 1. The observed and calculated neutron diffraction pattern of HoRhGe at 1.5 K. The filled squares represent the observed points, the solid lines the calculated profile and the difference between observed and calculated data (below). The vertical ticks indicate nuclear (top) and magnetic (bottom) Bragg peaks.

3. Results

The neutron diffraction patterns obtained at $T = 6$ K for HoRhGe and at 11 K for ErRhGe show that both compounds have the TiNiSi type of crystal structure ($Pnma$ space group). The 4 R, 4 Rh and 4 Ge atoms occupy the 4(c) sites: $x, 1/4, z$; $\bar{x}, 3/4, \bar{z}$; $1/2 - x, 3/4, 1/2 + z$; $1/2 + z, 1/4, 1/2 - z$ with different x and z coordinates. Their values are shown in table 1.

The neutron diffraction pattern of the HoRhGe compound recorded at 1.5 K shows additional peaks which are indexed with the wave vector $k = (1/2, 0, 1/2)$ (figure 1). The distribution of the peaks is similar to those observed in the isostructural NdRhGe [3] and HoRhSi [8,9]. The Ho magnetic moment occupies the following positions in the crystal unit cell:

$$\begin{array}{ll}
 S_1(x, 1/4, z) & S_2(\bar{x}, 3/4, \bar{z}) \\
 S_3(1/2 - x, 3/4, 1/2 + z) & S_4(1/2 + x, 1/4, 1/2 - z).
 \end{array}$$

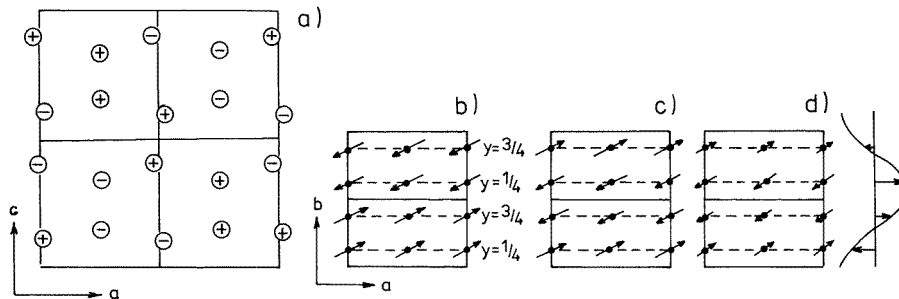
Table 1. Crystal structure and magnetic parameters for HoRhGe and ErRhGe compounds.

<i>T</i> (K)	HoRhGe		ErRhGe			
	1.5	7	1.5	4	6	11
<i>a</i> (Å)	6.819(3)	6.794(3)	6.805(3)	6.799(3)	6.810(3)	6.842(3)
<i>b</i> (Å)	4.252(2)	4.286(2)	4.263(2)	4.253(2)	4.248(2)	4.275(2)
<i>c</i> (Å)	7.468(3)	7.327(3)	7.495(4)	7.490(3)	7.502(3)	7.540(3)
<i>V</i> (Å ³)	216.50(26)	213.35(26)	217.43(28)	216.58(26)	217.02(26)	220.54(26)
<i>x</i>	0.007(5)	0.008(7)	0.028(4)	0.033(5)	0.016(7)	0.032(4)
R						
<i>z</i>	0.186(3)	0.202(3)	0.195(3)	0.194(3)	0.198(3)	0.192(4)
<i>x</i>	0.156(5)	0.147(4)	0.205(6)	0.155(8)	0.154(7)	0.159(5)
Rh						
<i>z</i>	0.555(5)	0.586(6)	0.621(8)	0.575(8)	0.561(6)	0.561(6)
<i>x</i>	0.782(4)	0.784(4)	0.811(4)	0.793(5)	0.790(5)	0.792(4)
Ge						
<i>z</i>	0.602(4)	0.561(5)	0.575(5)	0.591(7)	0.600(6)	0.613(6)
<i>R</i> _{Bragg} (%)	7.5	7.6	8.6	8.9	9.0	10.3
<i>R</i> _{prof} (%)	5.4	6.9	7.8	7.5	8.1	8.0
μ (μ_B)	6.96(12)		8.40(18)	8.33(18)	6.76(15)	
<i>R</i> _m (%)	9.7		5.7	4.3	7.8	

The Bertaut theory [10] constructed on the basis of an irreducible representation for spin transformation of the 4(c) site in the space group *Pnma* gives three antiferromagnetic structures described by the vectors:

$$G = S_1 - S_2 + S_3 - S_4 \quad C = S_1 + S_2 - S_3 - S_4 \quad A = S_1 - S_2 - S_3 + S_4.$$

The best agreement with the experimental data is obtained for a collinear magnetic structure described by the *C* vector with the moment equal to 6.96(12) μ_B and parallel to the *b* axis.

**Figure 2.** Projection of the magnetic structure of (a) HoRhGe on (*a-c*) plane and ErRhGe on (*a-b*) plane for (b) F (ferromagnetic) mode, (c) G mode and (d) sine modulated structure.

The projection of this structure on the *a-c* plane is shown in figure 2(a). Figure 3 shows parts of neutron diffractograms recorded as a function of temperature for HoRhGe (a) and ErRhGe (b).

The temperature dependence of the magnetic intensity of 100⁻ and 001⁻ peaks gives the Néel temperature 4.6 K (see figure 4(a)).

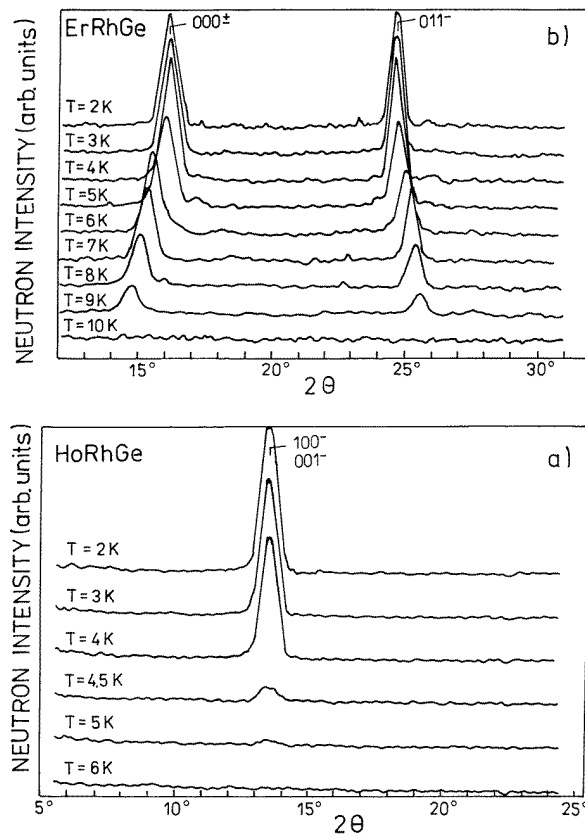


Figure 3. A part of the neutron diffraction pattern of (a) HoRhGe and (b) ErRhGe measured as a function of temperature.

The magnetic peaks observed on the neutron diffraction pattern of ErRhGe (figure 5) are indexed with the wave vector $\mathbf{k} = (0, 1/2, 0)$. The magnetic unit cell is doubled along the b -axis with respect to the crystal one. The minimum of the ‘agreement’ R factor corresponds to the collinear magnetic structure in which magnetic moments are coupled ferromagnetically (F mode) inside the chemical unit cell, but antiferromagnetically in the magnetic one (see figure 2(b)). A similar value of the R factor also corresponds to the model in which the Er^{3+} magnetic moments are coupled $+-+-$ (G mode) inside the chemical unit cell but antiferromagnetically in the magnetic one (see figure 2(c)). The magnetic moment equal to $8.40(18) \mu_B$ lies in the $(a-b)$ plane and forms an angle of $\Psi = 25^\circ$ with the a axis. Increasing the temperature up to 5 K does not change the diffraction patterns. The positions of the magnetic peaks on the diffraction pattern obtained at $T = 6$ K differ from those observed at 5 K. These peaks are indexed with the propagation vector $\mathbf{k} = (0, k_y, 0)$, where $k_y \neq 0.5$. The minimum of the reliability factor corresponds to the magnetic structure which forms a sine-modulated structure with the magnetic moment equal to $6.76(15) \mu_B$ in the $(a-b)$ plane ($\Psi = 25^\circ$) (see figure 2(d)).

The Néel temperature can be obtained from the temperature dependence of $(000)^+$ is equal to 9.5 K. The propagation vector below 4 K is constant and equal to $k_y = 0.5$. Between 5 and 6 K a jump of the vector is observed and above 6 K it decreases with increasing temperature (see figure 4(b)).

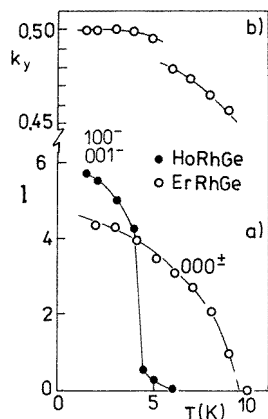


Figure 4. Temperature dependence of the (a) magnetic intensity of the reflection for HoRhGe and reflection for ErRhGe and (b) k_y component of the wave vector for ErRhGe.

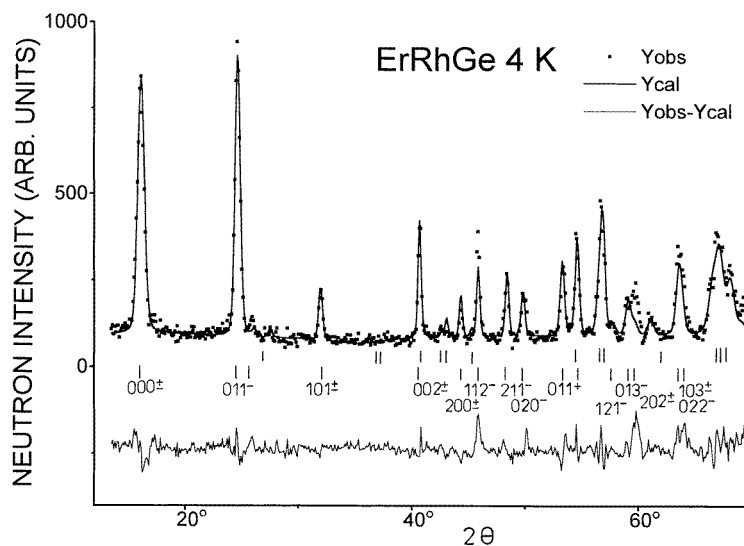


Figure 5. The observed and calculated neutron diffraction pattern of ErRhGe at 4 K. The squares represent the observed points and the solid lines the calculated profile and the difference between observed and calculated data (below). The vertical ticks indicate nuclear (top) and magnetic (bottom) peaks.

4. Discussion

The investigated compounds order antiferromagnetically at low temperatures. The Néel temperatures are equal to 4.6 K for HoRhGe and 9.5 K for ErRhGe.

The interatomic distances between rare earth atoms suggest that direct magnetic interactions are highly improbable. The stability of the observed magnetic ordering scheme may thus be considered as due to the interactions via conduction electrons (RKKY model). The magnetocrystalline anisotropy is caused by the influence of the crystalline electric field (CEF) on 4f electrons.

The magnetic structure of HoRhGe is similar to those observed for isostructural NdRhGe [3], HoRhSi [8, 9], HoNiGe [11] and HoNiSi [12]. HoRhGe has a collinear magnetic structure with the magnetic unit cell doubled along a and c axis with respect to the crystal ones. In all holmium compounds magnetic moment is parallel to the b axis.

For the compounds with Rh the magnetic structure is stable with temperature up to the Néel point. A different situation is observed in the Ni compounds, in which near the Néel temperature a change to the sine modulated structure is observed [11, 12].

At low temperatures, the ErRhGe compound has a collinear antiferromagnetic structure with the magnetic unit cell doubled along the b -axis in respect to the crystal one. With an increasing temperature a change of the magnetic structure to the incommensurate sine modulated one described by the propagation vector is observed. In the isostructural ErRhGe a helicoidal magnetic order with the propagation vector $\mathbf{k} = (0, 0.4, 0)$ is observed [9].

For the ErNiX compounds the magnetic ordering is described by the propagation vector $\mathbf{k} = (0, 1/2, 1/4)$ for X = Si [12] and $\mathbf{k} = (0, 1/2, 0)$ for X = Ge [11] at low temperatures. An increasing temperature changes the magnetic structure into incommensurate sine modulated with the wave vector $\mathbf{k} = (0.197, 0.515, 0.154)$ for X = Si and $\mathbf{k} = (0, 0.5, 0.084)$ for X = Ge.

The presented experimental data lead to the following conclusions.

(1) The Néel temperatures of the germanides are smaller than those observed in isostructural silicides. It indicates that the magnetic interactions depend on interatomic distances because the lattice constants of germanides are larger than those of the silicides (see table 2).

Table 2. Comparison of magnetic data for HoTX and ErTX compounds (T = Rh, Ni, X = Si and Ge).

Compound	V (\AA^3)	T_N (K)	T_i (K)	μ (μ_B)	Ref.
HoRhSi	209.00	8.4; 11	—	9.07, 8.7(2)	[8, 9]
HoRhGe	213.35	4.6	—	6.96(12)	This work
HoNiSi	202.09	4.1	3.2	7.08(7)	[12]
HoNiGe	209.12	2.75	2.2	9.98(5)	[11]
ErRhSi	209.20	12	—	6.6(2)	[9]
ErRhGe	220.54	9.5	5	8.40(18)	This work
ErNiSi	200.76	3.3	2.25	7.68(9)	[12]
ErNiGe	206.30	2.9	2.36	9.05(5)	[11]

(2) The change of the magnetic structure from commensurate to incommensurate near the Néel temperature and sine modulated ordering observed in these last phases imply that the RKKY model of magnetic interactions can be applied here. However the Néel dependence of the de Gennes factor is not obeyed (see figure 6). These results suggest that the other factors influence on the magnetic ordering.

(3) In the erbium compounds the direction of the magnetic moment differs from that observed in the holmium compounds. It results from the influence of the crystalline electric field (CEF). The CEF Hamiltonian for the lanthanide atoms which occupy the sites of the C_s symmetry in the TiNiSi type of crystal structure is the following:

$$H_{CEF} = B_2^0 O_2^0 + B_2^2 O_2^2 + B_4^0 O_4^0 + B_4^2 O_4^2 + B_4^4 O_4^4$$

where B_n^m are the crystal field parameters and O_n^m represent polynomials of the angular momentum operators [13]. In this Hamiltonian the second-order CEF parameter B_2^0 is

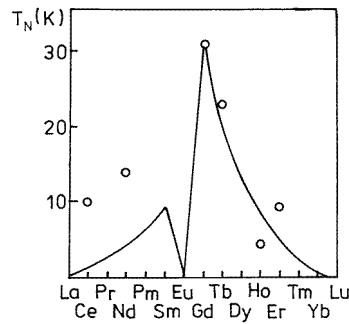


Figure 6. Ordering temperatures of RRhGe compounds plotted against the number of 4f electrons localized on the R^{3+} ions. The solid line represents the de Gennes function normalized with respect to GdRhGe.

dominant and the magnitude and the sign of this parameter determine the direction of the magnetic moment. For example, in TbNiSn $B_2^0 = -1.11$ K, $B_2^2 = 0.08$ K, $B_4^2 = 0.0007$ K, $B_4^4 = -0.0055$ K, $B_4^6 = 0.0059$ K [14] and the magnetic moment is parallel to the b axis [15]. The experimental data and the results of the point charge calculation of the crystal field parameters for the isostructural RSi compounds [16] indicate that for the negative sign of the B_2^0 parameter the magnetic moment is parallel to the c axis, whereas for the positive sign the moment lies in the (a, c) plane. The observed change of the direction of the magnetic moment from parallel to the b axis for HoRhGe to nearly parallel to the a axis for ErRhGe causes the change of the sign of the B_2^0 parameter.

The CEF effect manifests itself by an attenuation of the observed magnitude of the magnetic moment localized on the lanthanide ion (see table 2).

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