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Magnetic structures of $HoRh_{2-x}Ru_xSi_2$ compounds

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Abstract

The results of neutron diffraction measurements carried out for a number of compositions in the $HoRh_{2-x}Ru_xSi_2$ solid solution show that all samples exhibit the $ThCr_2Si_2$ type of structure. When the x increases, the magnetic ordering changes from the antiferro collinear AFI-type via complex structure to a sine-modulated structure.

Keywords: Rare-earth intermetallics; Antiferromagnetic; Magnetic structure

1. Introduction

$HoRh_2Si_2$ and $HoRu_2Si_2$ crystallize in the body-centered tetragonal $ThCr_2Si_2$-type of crystal structure [1]. Both compounds are antiferromagnets with a Néel temperature 27 K for $HoRh_2Si_2$ and 19 K for $HoRu_2Si_2$ and have different magnetic structures: $HoRh_2Si_2$ is a collinear antiferromagnet of the AFI-type whereas $HoRu_2Si_2$ has a modulated magnetic structure [2].

X-ray diffraction data indicate that the $Ho(Rh_{1-x}Ru_x)_2Si_2$ solid solution exists in the whole region of the concentration x. All compounds have the $ThCr_2Si_2$-type of crystal structure. The magnetic properties depend on the ruthenium concentration. With an increasing concentration of Ru up to $x \approx 0.4$, the Néel temperature (T_N) decreases and for higher concentrations T_N increases.

In this work, the results of neutron diffraction measurements for $HoRh_{2-x}Ru_xSi_2$ are presented and the crystal and magnetic structure parameters are determined.

2. Experimental details and results

The experiments were carried out on polycrystalline samples as reported in the previous paper [3].

Neutron diffractograms were obtained using the E6 diffractometer installed at the BERII reactor in the Berlin Neutron Scattering Centre, Hahn-Meitner Institute. The incident neutron wavelength was 2.426 Å. Data were collected at temperatures in the range from 1.5 to 40 K. The nuclear data processing was performed by the Rietveld type FULLPROF program [4] with neutron scattering lengths taken from Sears [5]. The parameters of the magnetic structure (value of the wave vector and type of the magnetic ordering) are firstly determined by the programme described by Tomkowicz [6] and next refined by the FULLPROF program with the Ho^{3+} form factor adopted from Freeman and Desclaux [7].

Neutron diffraction data obtained at the paramagnetic region confirm the body-centered tetragonal $ThCr_2Si_2$-type of crystal structure which is described

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by the space group I4/mmm with Ho, Rh or Ru and Si atoms located in the sites 2a, 4d and 4e, respectively.

For all samples, the neutron diffraction patterns taken at $T = 1.6$ K contain a number of magnetic peaks.

The analysis of the neutron diffraction data gives the following magnetic structures:

- For $x = 0.25$, below $T_N = 19$ K, the compounds have a collinear antiferromagnetic structure of the AFI-type with the magnetic moment parallel to the c-axis. Below 8 K an additional 001 peak appears which indicates that the Ho magnetic moment forms an angle $\theta = 25(2)°$ with the c-axis. At this temperature other peaks are present. They are described by the propagation vector $\mathbf{k} = (\frac{1}{2},\frac{1}{2},\frac{1}{2})$ and correspond to a collinear antiferromagnetic structure.
- For $x = 0.5$, below $T_N = 6.5$ K, the magnetic peaks correspond to a collinear antiferromagnetic structure with the propagation vector $\mathbf{k} = (\frac{1}{2},\frac{1}{2},\frac{1}{2})$. Below $T = 3.5$ K a sine modulated structure with the propagation vector $\mathbf{k} = (0.15, 0.15, 0.47)$ coexists with the collinear structure (see Fig. 1).
- For $x = 1.0$, below $T_N = 5.5$ K a sine modulated structure described by the propagation vector $\mathbf{k} = (0.135, 0.135, 0.285)$ is determined. Half-widths of the magnetic peaks are much larger than those for the crystallographic reflections, which suggests that the magnetic order is limited to clusters and does not extend over the whole crystal.
- For $x = 1.5$, below 8 K a collinear ferromagnetic ordering, with the magnetic moment parallel to the c-axis, is observed (see Fig. 2a). In the temperature region $7\ \mathrm{K} < T < 12$ K two sine modulated structures with the propagation vectors $\mathbf{k}_1 = (0.147, 0.147, 0)$ and $\mathbf{k}_2 = (0.28, 0, 0)$ coexist (see Fig. 2b).
- For $x = 1.75$, below $T_N = 15.5$ K, a sine modulated structure with the propagation vector $\mathbf{k} = (0.258, 0, 0)$ is observed.

The determined parameters of the magnetic structures are presented in Table 1. The Ho magnetic moment is parallel to the c-axis in all $HoRh_{2-x}Ru_xSi_2$ compounds with $x \geq 0.5$. The values of T_N and T_t are determined from the temperature dependence of the intensity of the magnetic reflections. The determined magnetic phase diagram is shown in Fig. 3.

3. Discussion

In these compounds the Ho^{3+}–Ho^{3+} interionic distances are long enough (~4 Å in the basal plane and ~5 Å between planes) so the magnetic interactions in these compounds are probably of the RKKY type [2].

The results presented in this work show that the

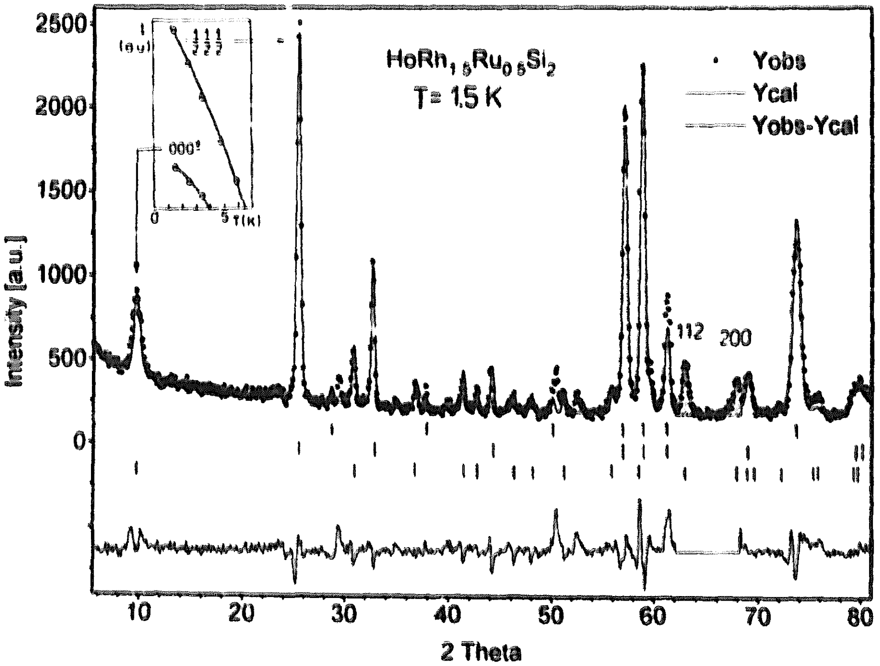

Fig. 1. Neutron diffraction pattern of $HoRh_{1.5}Ru_{0.5}Si_2$ at 1.6 K. The open squares represent the observed points, the solid lines the calculated profile and the difference between observed and calculated data (below). The vertical ticks indicate, respectively (from top to bottom) nuclear and magnetic corresponding to the wave vector (1/2, 1/2, 1/2) and (0.15, 0.15, 0.47) Bragg peaks. Insert shows the temperature dependences of the intensities of the (1/2, 1/2, 1/2) and $(0, 0, 0)^{\pm}$ reflections.

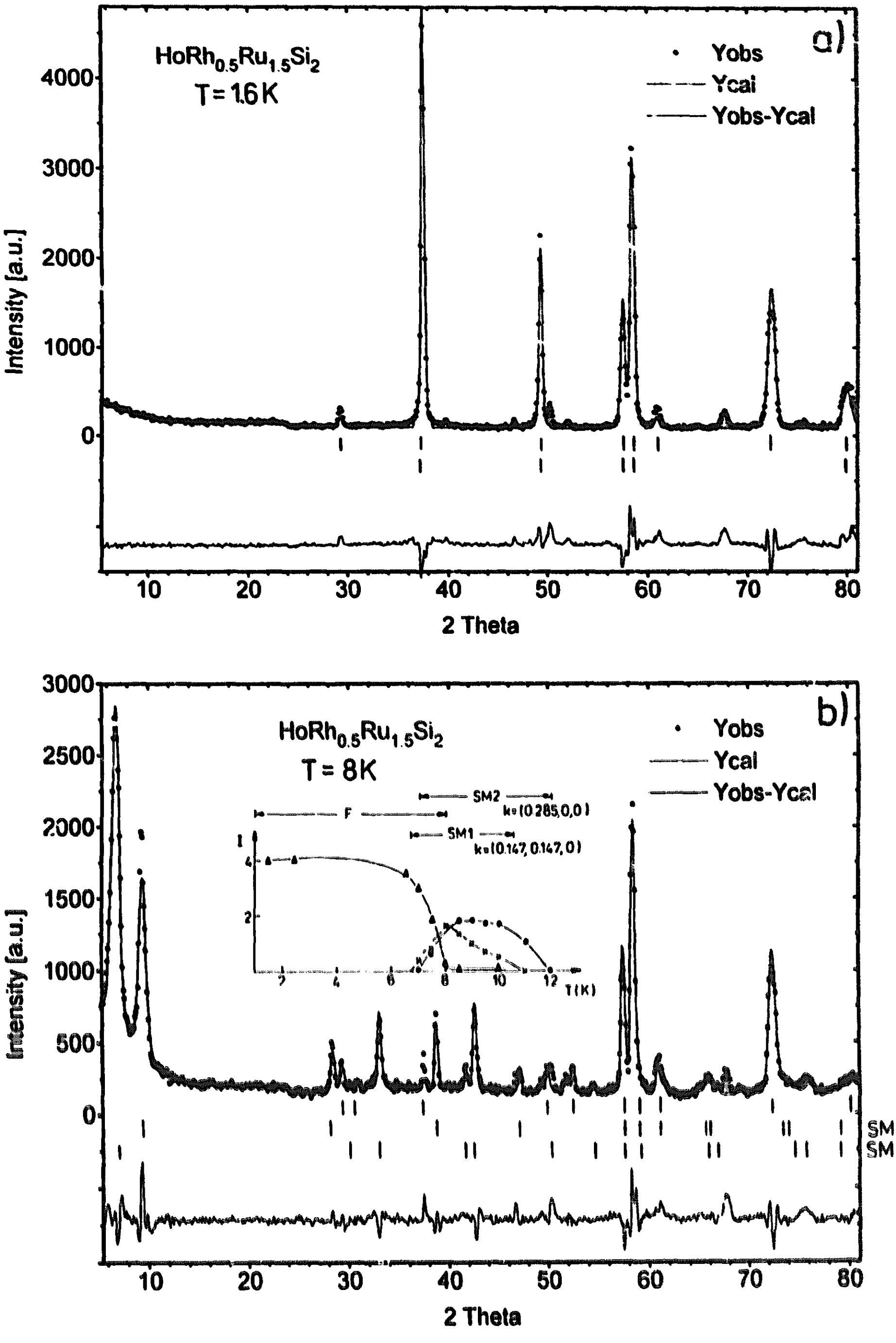

Fig. 2. Neutron diffraction patterns of $HoRh_{0.5}Ru_{1.5}Si_2$ at (a) 1.6 K and (b) 10 K. The open squares represent the observed points, the solid lines the calculated profile and the difference between observed and calculated data (below). The vertical ticks indicate, respectively (from top to bottom) nuclear and magnetic peaks. Insert shows the temperature dependences of the intensities dependence of (101) and $(000)^{\pm}$ magnetic peaks.

substitution of rhodium by ruthenium changes the magnetic structure from a collinear antiferro-AFI-type to magnetic structures described by the different wave vectors (1/2, 1/2, 1/2), (k_x, k_y, k_z), (0, 0, 0) and $(k_x, 0, 0)$. This is very surprising in the tetragonal system where a strong uniaxial behaviour should be expected. We have to consider that the Fourier transform of the exchange coupling $J(k)$ seems to present flat dependence in the k-space, allowing the existence of extrema in several positions out of the main symmetry directions (see Ball et al. [8]). This gives the magnetic ordering with the different values of the wave vector. The change of the magnetic structure is connected with a decrease of the Néel temperature when concentration increases up to $x = 0.4$ and next it increases for high concentrations of Ru. This effect is also observed for other $RRh_{2-x}Ru_xSi_2$ systems, where R = Ce [9], Nd [10], Tb [11] and U [12]. It suggests

Table 1
Magnetic structure parameters for $\mathrm{HoRh}_{2-x}\mathrm{Ru}_x\mathrm{Si}_2$ compounds determined from neutron diffraction data

Low temperature phase								High-temperature phase				
x	T_N (K)	T_t (K)	T (K)	k-vector	μ (μ_B)	θ (°)	R_m (%)	T (K)	k-vector	μ (μ_B)	θ (°)	R_m (%)
0[a]	27	11	4.2	[0, 0, 1]	8.8(2)	28(3)						
0.25	19	8	1.6	[0, 0, 1]	5.94(10)	26(2)	11.7					
				[1/2, 1/2, 1/2]	5.64(9)	0	9.7	8.0	[0, 0, 1]	4.8(1)	0	6.3
0.5	6.5	3.5	1.6	[1/2, 1/2, 1/2]	5.10(8)	0	10.5	3.5	[1/2, 1/2, 1/2]	4.1(2)	0	8.1
				[0.15, 0.15, 0.468]	3.83(13)[c]		14.7					
1.0	5.5		1.6	[0.132, 0.132, 0.297]	9.05(8)[c]	0	7.4					
1.5	12	7	1.6	[0, 0, 0]	8.32(20)	0	10.2	8	[0.147, 0.147, 0]	4.7(1)[c]	0	11.4
									[0.285, 0, 0]	4.4(1)[c]	0	9.3
								10	[0.287, 0, 0]	5.63(9)[c]	0	8.8
1.75	15.5		1.6	[0.252, 0, 0]	9.03(5)[c]	0	4.6					
2.0[b]	19		4.2	[0.22, 0, 0]	9.25(5)[c]	0						

[a] Data from Ślaski et al. [16].
[b] Data from Ślaski et al. [17].
[c] The effective value of the magnetic moment.

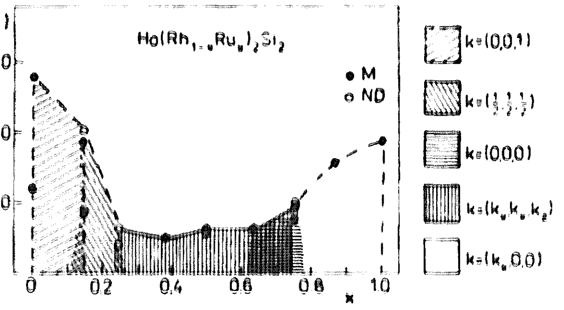

Fig. 3. The (T, x) magnetic phase diagram of $\mathrm{Ho(Rh}_{1-x}\mathrm{Ru}_x)_2\mathrm{Si}_2$.

that the d-electrons form the conduction bands and change the density of the states at the Fermi surface which modify magnetic interactions leading to the observed changes of the magnetic structure. A similar dependence of the Néel temperature in the function of the number of d-electrons was observed in $\mathrm{GdT_2Si_2}$ [13], $\mathrm{ErT_2Si_2}$ [14] and in $\mathrm{EuT_2Ge_2}$ and $\mathrm{GdT_2Ge_2}$ compounds [15]. The results of the Mössbauer measurements for isostructural $\mathrm{GdT_2Si_2}$ compounds also indicate an influence of the transition d-band metal on many properties of these compounds [13].

In all $\mathrm{HoRh}_{2-x}\mathrm{Ru}_x\mathrm{Si}_2$ compounds with $x \geq 0.5$ the Ho-magnetic moment is parallel to the c-axis. It indicates a strong Ising-type anisotropy and a small influence of the substitution of rhodium by ruthenium on the crystal electric field parameters.

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References

[1] W. Rieger, E. Parthé, Mh. Chem. 100 (1969) 444.
[2] A. Szytuła, J. Leciejewicz, in: K.A. Gschneidner, L. Eyring (Eds.), Handbook on the Physics and Chemistry of Rare Earths, vol. 12, North-Holland, Amsterdam, 1989, Ch. 83, p. 139.
[3] V. Ivanov, T. Jaworska, L. Vinokurova, T. Mydlarz, A. Szytuła, J. Alloys Comp. 234 (1996) 235.
[4] J. Rodriguez-Carvajal, Physica B 192 (1993) 55.
[5] V.F. Sears, Neutron News 3 (1992) 26.
[6] Z. Tomkowicz, Universitas Jagellonica, Acta Sientiarum Literarumque 1979, Schadae Physicae, Fasciellus 16.
[7] A.J. Freeman, J.P. Desclaux, J. Magn. Magn. Mater. 12 (1979) 11.
[8] A.R. Ball, D. Gignoux, D. Schmitt, F.Y. Zhang, J. Magn. Magn. Mater. 140/144 (1995) 1121.
[9] Calemczuk, E. Bonjour, J. Rossat-Mignod, B. Chevalier, J. Magn. Magn. Mater. 9091 (1990) 477.
[10] T. Jaworska, A. Szytuła, H. Ptasiewicz-Bak, J. Leciejewicz, Solid State Commun. 65 (1988) 261.
[11] T. Jaworska, A. Szytuła, J. Leciejewicz, H. Ptasiewicz-Bak, J. Magn. Magn. Mater. 82 (1989) 313.
[12] P. Burlet, F. Bourdarot, S. Quezel, et al., J. Magn. Magn. Mater. 108 (1992) 202.
[13] K. Łatka, Magnetism and Hyperfine Interactions in $\mathrm{GdT_2Si_2}$ Systems, Report No 1443/PS, Institute of Nuclear Physics, Cracow, 1989.
[14] J. Leciejewicz, S. Siek, A. Szytuła, J. Magn. Magn. Mater. 40 (1984) 265.
[15] I. Felner, I. Nowik, J. Phys. Chem. Solids 39 (1978) 763.
[16] M. Ślaski, J. Leciejewicz, A. Szytuła, J. Magn. Magn. Mater. 39 (1983) 268.
[17] M. Ślaski, A. Szytuła, J. Leciejewicz, A. Zygmunt, J. Magn. Magn. Mater. 46 (1984) 114.