Vol. 34 (2003) *ACTA PHYSICA POLONICA B* No 2

WHAT IS THE REASON FOR DIFFERENT MAGNETIC PROPERTIES OF THE COMPOUNDS $\mathrm{TmRu_2Si_2}$ AND $\mathrm{YbRu_2Si_2}$ AMONG THE $\mathrm{RRu_2Si_2}$ COMPOUNDS?*

R. Michalski[a,b], A. Blaut[b], R.J. Radwanski[a,b]

[a]Institute of Physics, Pedagogical University
Podchorążych 2, 30-084 Kraków, Poland
[b]Center of Solid State Physics, św. Filipa 5, 31-150 Kraków, Poland

(Received July 10, 2002)

Experimental observations and theoretical analysis allow for understanding of the extremely anisotropic magnetic properties of compounds $\mathrm{ErRu_2Si_2}$ and $\mathrm{PrRu_2Si_2}$. The presented here calculations for $\mathrm{TmRu_2Si_2}$ and $\mathrm{YbRu_2Si_2}$ reveal that their properties are quite different from those of the Er and Pr compounds. We have found that a low-temperature ordering of $\mathrm{YbRu_2Si_2}$ and a weak ferromagnetism ($T_{\mathrm{C}} \approx 1$K) of $\mathrm{TmRu_2Si_2}$ can be obtained by using crystal-field parameters directly recalculated from $\mathrm{ErRu_2Si_2}$ and $\mathrm{PrRu_2Si_2}$. Moreover, an effect of the temperature-dependent rotation of the easy magnetic axis in $\mathrm{YbRu_2Si_2}$ is predicted.

PACS numbers: 75.10.Dg, 71.30.+h

1. Introduction

Magnetic properties of monocrystalline ternary compounds $\mathrm{RRu_2Si_2}$ (R= Pr, Nd, Tb, Dy, Ho, Er) are strongly anisotropic. Calculations of magnetocrystalline anisotropy for $\mathrm{ErRu_2Si_2}$ [1] and Pr $\mathrm{Ru_2Si_2}$ [2] were performed. A well-defined direction of the magnetic easy axis for these compounds was not changing in the whole temperature range and the magnetocrystalline anisotropy was significant. The direction and the value of the anisotropy field in case of $\mathrm{PrRu_2Si_2}$ and $\mathrm{ErRu_2Si_2}$ depends on the crystal field (CEF) that has similar values of the coefficients A_n^m for both compounds, but yields the different directions of easy axes [1–3]. In this situation the results of the similar calculations for $\mathrm{TmRu_2Si_2}$ and $\mathrm{YbRu_2Si_2}$ are surprising. We have treated $\mathrm{Tm^{3+}}$ and $\mathrm{Yb^{3+}}$ ions in the same manner as the $\mathrm{Er^{3+}}$ and $\mathrm{Pr^{3+}}$

---

[*] Presented at the International Conference on Strongly Correlated Electron Systems, (SCES 02), Cracow, Poland, July 10–13, 2002.

ions, but the obtained results are quite different from those for the Er and Pr compounds. Our calculations provide the answer to the question: why $YbRu_2Si_2$ and $TmRu_2Si_2$ have such low temperature of ordering (Ordering temperature $YbRu_2Si_2$ in not found and $TmRu_2Si_2$ exhibits a weak ferromagnetism ($T_C \approx 1$ K) [6]. We have established that these properties can be evoked by CEF parameters directly recalculated from $ErRu_2Si_2$ and $PrRu_2Si_2$. Moreover, an effect of the temperature-dependent rotation of the easy magnetic axis in $YbRu_2Si_2$ is predicted.

2. Outline of theory

Our approach bases on the crystal fields model, that points out that f atoms partially preserve their atomic properties even becoming the full part of a solid [8]. We have attributed the magnetic properties of $TmRu_2Si_2$ and $YbRu_2Si_2$ to be predominantly due to the $4f^{12}$ and $4f^{13}$ electronic system of the Tm^{3+} and Yb^{3+} ions respectively [4]. The used method is based on according to the local symmetry of the surroundings of the R ions, which leads to the tetragonal CEF+Zeeman Hamiltonian:

$$\begin{aligned} H_{\text{tetra}} &= B_2^0\hat{O}_2^0(J,J_z) + B_4^0\hat{O}_4^0(J,J_z) + B_4^4\hat{O}_4^4(J,J_z) \\ &\quad + B_6^0\hat{O}_6^0(J,J_z) + B_6^4\hat{O}_6^4(J,J_z) + g_J\mu_B \boldsymbol{J}\cdot\boldsymbol{B}_{\text{ext}} , \end{aligned}$$

where B_n^m are CEF parameters, $\hat{O}_n^m$ are Stevens operators, g_J is Landé factor, $\boldsymbol{B}_{\text{ext}}$ is the external magnetic field. The values of total momentum quantum numbers $\boldsymbol{J}$ of the R ions are 6 and $\frac{7}{2}$ for Tm^{3+} and Yb^{3+} ions, respectively. The details of the calculations of the magnetic properties are explained in Ref. [9]. The main idea of the theoretical estimation of the properties of the considered compounds is possibility of the selection of the CEF parameters for the $YbRu_2Si_2$ and $TmRu_2Si_2$ according to parameters established for $ErRu_2Si_2$ and $PrRu_2Si_2$ [1–3]. Direct recalculation from it base at the subtraction of the local-potentials coefficients A_n^m according to the formula:

$$B_n^m = A_n^m \left\langle r_{4f}^n \right\rangle \theta_n ,$$

where the $\left\langle r_{4f}^n \right\rangle$ is average of n-power of radius of the $4f$ shell and θ_n are the scaling parameter, Stevens coefficients, dependent on the number of $4f$ electrons. The values of θ_n can be found in [7]. Coefficients A_n^m carry information of the potential of the local environment of rare ions in the crystal structure. We assume that proper values of A_n^m should be similar for the whole family of compounds RRu_2Si_2.

3. Results for $TmRu_2Si_2$

The main properties of 3 lowest lying states are collected in Table I. Independently of the used set of parameters (recalculated from $ErRu_2Si_2$ or $PrRu_2Si_2$), practically the same energy structure has been found: the

TABLE I

Sets of CEF parameters for $TmRu_2Si_2$ and $YbRu_2Si_2$ together with the magnetic characteristics of 3 lowest states. g.s. denotes the ground state, 1^{st}, 2^{nd} e.s. denote the first and second excited states, $\langle J_x\rangle$ and $\langle J_z\rangle$ are the expectation values of x and z components of the total angular momentum J; **K-doublet** and **nk-doublet** denote Kramers and non-Kramers doublet, δ_1 and δ_2 are energies of the excited states with respect to the ground state, $m(B)$ is the magnetic moment at 1 K in the external field B.

		$PrRu_2Si_2$		**$ErRu_2Si_2$**	
n	***m***	B_n^m	A_n^m	B_n^m	A_n^m
2	**0**	**- 22.0 K**	**+867 K/a_0^{-2}**	**+ 1.6 K**	**+ 870 K/a_0^{-2}**
4	**0**	**+ 220 mK**	**-88.2 K/a_0^{-4}**	**- 5.0 mK**	**- 88 K/a_0^{-4}**
6	**0**	**- 12 mK**	**-11 K/a_0^{-6}**	**- 5 µK**	**-0.5 K/a_0^{-6}**
4	**4**	**+ 200 mK**	**- 80.2 K/a_0^{-4}**	**- 4.5 mK**	**- 80 K/a_0^{-4}**
6	**4**	**-45 mK**	**- 39 K/a_0^{-6}**	**- 0.52 mK**	**- 50 K/a_0^{-6}**
		Recalculated parameters	**Properties**	**Recalculated parameters**	**Properties**
$TmRu_2Si_2$		B_2^0**=+6.0 K**	easy axis: [1 0 0] $m_z(6T)=0.00\mu_B$*/ion* $m_x(6T)=2.8\mu_B$*/ion* **g.s.- Singlet** $\langle J_z\rangle=\langle J_x\rangle=0.00$. **$1^{st}$ e.s.- n-K doublet** δ_1 = 74 K *(INS visible)* $\langle J_z\rangle=\pm0.85$, $\langle J_x\rangle=0.00$. **2^{nd} e.s.- singlet** δ_2 = 222K *(INS visible)* $\langle J_z\rangle=\langle J_x\rangle=0.00$.	B_2^0**=+6.0 K**	easy axis: [1 0 0] $m_z(6T)=0.01\mu_B$*/ion* $m_x(6T)=4.5\mu_B$*/ion* **g.s. -Singlet** $\langle J_z\rangle=\langle J_x\rangle=0.00$. **$1^{st}$ e.s.- n-K dublet** δ_1 = 33.8 K *(INS visible)* $\langle J_z\rangle=\pm0.68$, $\langle J_x\rangle=0.00$. **2^{nd} e.s.- singlet** δ_2 = 98.7K *(INS visible)* $\langle J_z\rangle=\langle J_x\rangle=0.00$.
		B_4^0**=-16.9 mK**		B_4^0**=-16.9 mK**	
		B_6^0**=+256 µK**		B_6^0**=+12 µK**	
		B_4^4**=-15.4 mK**		B_4^4**=-15.4 mK**	
		B_6^4**=+0.96 mK**		B_6^4**=+1.27 mK**	
$YbRu_2Si_2$		B_2^0**=+18.1 K**	easy axis: [? ? ?] $m_z(6T)=1.64\mu_B$*/ion* $m_x(6T)=0.94\mu_B$*/ion* $m_z(30T)=1.67\mu_B$*/ion* $m_x(30T)=1.78\mu_B$*/ion* **g.s.- K-doublet** $\langle J_z\rangle=\pm1.63$, $\langle J_x\rangle=\pm0.55$. **$1^{st}$ e.s.- K-doublet** δ_1 = 123K *(INS visible)* $\langle J_z\rangle=\pm0.57$, $\langle J_x\rangle=\pm2.28$. **$2^{nd}$ e.s.- K-doublet** δ_2 = 235K *(INS visible)* $\langle J_z\rangle=\pm2.77$, $\langle J_x\rangle=\pm2.29$.	B_2^0**=+18.4 K**	easy axis: [? ? ?] $m_z(6T)=1.45\ \mu_B$*/ion* $m_x(6T)=1.86\ \mu_B$*/ion* **g.s.- K-doublet** $\langle J_z\rangle=\pm1.39$, $\langle J_x\rangle=\pm1.05$. **$1^{st}$ e.s.- K-doublet** δ_1 = 24.9 K *(INS visible)* $\langle J_z\rangle=\pm0.57$, $\langle J_x\rangle=\pm2.26$. **$2^{nd}$ e.s.- K-doublet** δ_2 = 146.4 K *(INS visible)* $\langle J_z\rangle=\pm2.53$, $\langle J_x\rangle=\pm1.02$.
		B_4^0**=+166 mK**		B_4^0**=+167 mK**	
		B_6^0**=-6.12 mK**		B_6^0**=-0.292 mK**	
		B_4^4**=+151 mK**		B_4^4**=+150 mK**	
		B_6^4**=-22.9 mK**		B_6^4**=-30.4 mK**	

singlet ground state and a non-Kramers doublet as the 1st excited state. Such configuration is hard to polarize magnetically by the mechanism found in $PrRu_2Si_2$ [1]. In the case of $TmRu_2Si_2$ the external field of 6T parallel to the local x-axis (easy axis) induces only 0.4 of the Tm^{3+} full moment of $7\mu_B$. Thus, according to us, this inactive singlet ground state is the reason for the low-temperature ordering of $TmRu_2Si_2$.

4. Results for $YbRu_2Si_2$

For the Yb^{3+} ion the recalculated CEF parameters yield the energy scheme with intriguing properties. For both sets of CEF parameters the 1^{st} excited Kramers doublet has the opposite direction of the magnetic moment than the Kramers-doublet ground state. Moreover, the moment of the excited state is much larger than that in the ground state (Fig. 1). Such a situation causes rotation of the easy magnetic axis with the increasing temperature from the direction [1 0 0] (or [0 1 0]) to [0 0 1] when the excited state becomes populated. The computer simulation of the influence of the external magnetic field reveals the crossing of the magnetization curves for different directions. It means that the easy magnetic axis changes with the field. The rotation field B_R depends on the chosen set of the parameters (recalculated from $ErRu_2Si_2$ or $PrRu_2Si_2$). Such rotation of the easy magnetic axis of the Yb^{3+} ion prevents, the formation of the long-range magnetic order in $YbRu_2Si_2$.

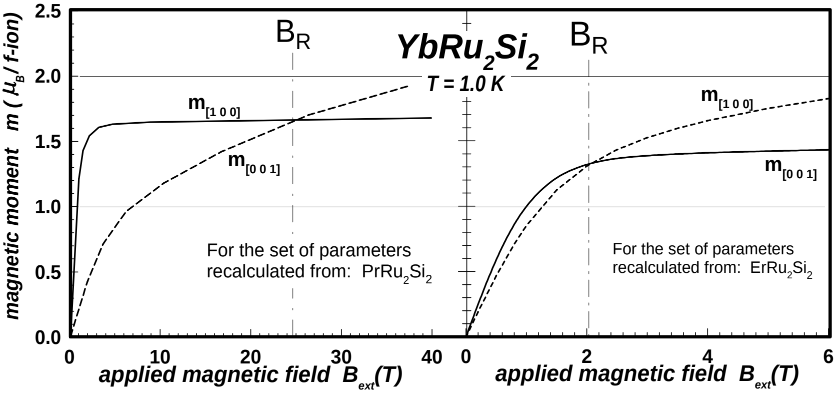

Fig. 1. Magnetization curves of $YbRu_2Si_2$ in applied fields along 2 main directions. B_R is the rotation field.

5. Conclusions

Magnetic properties of $TmRu_2Si_2$ and $YbRu_2Si_2$ have been discussed. Our results reveal the reason for the extremely low ordering temperature of these compounds. The isolated singlet for $TmRu_2Si_2$ and the rotating easy axis in $YbRu_2$ Si_2 could be studied further, both theoretically and experimentally. This work is only the starting point towards the understanding these compounds. The final evaluation of the fine electronic structure of these compounds will be possible once the inelastic neutron scattering for $TmRu_2Si_2$ and $YbRu_2Si_2$ are carried out.

Computations were performed with the help of the computer package `BIREC 1.5 for Windows` developed by Center for Solid State Physics Ltd [6]. This work was supported by KBN, project no. 2 PO3B 134 22.

REFERENCES

[1] R. Michalski, Z. Ropka, R.J. Radwanski, *J. Phys.: Condens. Matter* **12**, 7609 (2000).

[2] R. Michalski, Z. Ropka, R.J. Radwanski, *Physica B* **312–313**, 858 (2002).

[3] R. Michalski, R.J. Radwanski, Z. Ropka, *Acta Phys. Pol. B* **31**, 3079 (2000).

[4] K. Hiebl, C. Horvath, P. Rogl, M.J. Sienko, *J. Magn. Magn. Mater.* **37**, 287 (1983).

[5] K. Muramoto, T. Takeuchi, T. Taniguchi, Y. Miyako, *Physica B* **206–207**, 519 (1995).

[6] `http://www.css-physics.edu.pl/download.html`

[7] M.T. Hutchings, *Solid State Phys.* **16**, 227 (1964).

[8] R.J. Radwanski, Z. Ropka, `cond-mat/0010081`.

[9] R.J. Radwanski, N.H. Kim-Ngan, F.E. Kayzel, J.J.M. Franse, D. Gignoux, D. Schmitt, F.Y. Zhang, *J. Phys.: Condens. Matter* **4**, 8853 (1992).