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journal homepage: www.elsevier.com/locate/jmmmSingle crystal study of layered U_nRhIn_{3n+2} materials: Case of the novel U_2RhIn_8 compoundAttila Bartha^{a,*}, M. Kratochvílová^a, M. Dušek^b, M. Diviš^a, J. Custers^a, V. Sechovský^a^a Department of Condensed Matter Physics, Charles University, Ke Karlovu 5, 121 16 Praha 2, Czech Republic^b Department of Structure Analysis, Institute of Physics ASCR, Cukrovarnická 10, 162 00 Praha 6, Czech Republic

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ABSTRACT

We report on the single crystal properties of the novel U_2RhIn_8 compound studied in the context of parent $URhIn_5$ and UIn_3 systems. The compounds were prepared by In self-flux method. U_2RhIn_8 adopts the Ho_2CoGa_8 -type structure with lattice parameters $a=4.6056(6)$ Å and $c=11.9911(15)$ Å. The behavior of U_2RhIn_8 strongly resembles that of the related $URhIn_5$ and UIn_3 with respect to magnetization, specific heat and resistivity except for magnetocrystalline anisotropy developing with lowering dimensionality in the series UIn_3 vs. U_2RhIn_8 and $URhIn_5$. U_2RhIn_8 orders antiferromagnetically below $T_N=117$ K and exhibits a slightly enhanced Sommerfeld coefficient $\gamma=47$ mJ mol⁻¹ K⁻². Magnetic field leaves the value of Néel temperature for both $URhIn_5$ and U_2RhIn_8 unaffected up to 9 T. On the other hand, T_N is increasing when applying hydrostatic pressure up to 3.2 GPa. The character of uranium 5f electron states of U_2RhIn_8 was studied by first principles calculations based on the density functional theory. The overall phase diagram of U_2RhIn_8 is discussed in the context of magnetism in the related $URhX_5$ and UX_3 ($X=In, Ga$) compounds.

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1. Introduction

Magnetism of uranium compounds is characterized by the large spatial extent of the 5f wave functions which perceive their physical surroundings more intensively compared to the localized behavior of 4f electrons. Typical example of that is the 5f-ligand hybridization causing nonmagnetic behavior in several compounds characterized by the distance between the nearest U ions far larger than the Hill limit [1]. When considering the UX_3 ($X=p$ -metal) materials, the size of the p -atom is a very important parameter. In the case of smaller X -ions (Si, Ge) [2], the p -wave function decays slower at the U-site, resulting in strong 5f- p hybridization which leads to lack of magnetic ordering (UGe_3 , USi_3) [4,2,3] while larger X -ions (In, Pb) cause the hybridization to be weaker resulting in magnetic ground state (UIn_3 , UPb_3) [5,6].

The U_nTX_{3n+2} ($n=1, 2$; T =transition metal; $X=In, Ga$) [7-9,19,24] compounds adopt the layered Ho_nCoGa_{3n+2} -type structure which consists of n UX_3 layers alternating with a TX_2 layer sequentially along the [001] direction in the tetragonal lattice. They are isostructural with the thoroughly investigated Ce_nTX_{3n+2} [10] compounds known for their outstanding physical properties such as the coexistence of unconventional superconductivity and

magnetism or non-Fermi liquid behavior. These families of compounds provide unique opportunity to study the effect of dimensionality on physical properties due to their layered tetragonal structure. Adding a layer of TX_2 pushes the character of the structural dimensionality from 3D to more 2D.

Since the U_2RhIn_8 compound has not been reported yet, we focused in this paper on the structure study followed by investigation of magnetic, transport and thermodynamic properties with respect to applied magnetic fields and hydrostatic pressure. In order to study the evolution of ground state properties on the structural dimensionality, we also prepared and investigated single crystals of $URhIn_5$ and UIn_3 .

2. Experimental

Single crystals of UIn_3 , $URhIn_5$ and U_2RhIn_8 have been prepared using In self-flux method. High-quality elements U (purified by SSE [11]), Rh (3N5) and In (5N) were used. The starting composition of U:In=1:10, U:Rh:In=1:1:25 and U:Rh:In=2:1:25 was placed in alumina crucibles in order to obtain UIn_3 , $URhIn_5$ and U_2RhIn_8 , respectively. The crucibles were further sealed in evacuated quartz tubes. The ampoules were then heated up to 950 °C, kept at this temperature for 10 h to let the mixture homogenize properly and consequently cooled down to 600 °C in 120 h. After decanting, plate-like single crystals of U_2RhIn_8 ($URhIn_5$) with

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typical dimensions of $1 \times 0.5 \times 0.3 \text{ mm}^3$ ($1 \times 1 \times 0.5 \text{ mm}^3$) were obtained. In case of UIn_3 , however, our attempts led to growth of single crystals of typical masses $<0.1 \text{ mg}$. The single crystal of UIn_3 ($2 \times 2 \times 2 \text{ mm}^3$) suitable for the bulk measurements was obtained as a by-product of the URhIn_5 synthesis.

Homogeneity and chemical composition of the single crystals were confirmed by scanning electron microscope (Tescan MIRA I LMH SEM) equipped with energy dispersive X-ray analyzer (Bruker AXS). The crystal structures were determined by single crystal X-ray diffraction using X-ray diffractometer Gemini, equipped with an Mo lamp, graphite monochromator and an Mo-enhance collimator producing Mo K_α radiation, and a CCD detector Atlas. Absorption correction of the strongly absorbing samples ($\mu \sim 50 \text{ mm}^{-1}$) was done by the combination of the numerical absorption correction based on the crystal shapes and empirical absorption correction based on spherical harmonic functions, using the software of the diffractometer CrysAlis PRO. The crystal structures were solved by SUPERFLIP [13] and refined by software Jana2006 [14].

The electrical resistivity measurements were done utilizing the standard four-point method down to 2 K in a Physical Property Measurement System (PPMS). The specific heat measurements down to 400 mK were carried out using the He3 option. Magnetization measurements were performed in a superconducting quantum interference device (MPMS) from 2 to 300 K/400 K and magnetic fields up to 7 T.

To investigate the effect of hydrostatic pressure on the transition temperature T_N , we measured the temperature dependence of electrical resistivity using a double-layered (CuBe/NiCrAl) piston-cylinder type pressure cell with Daphne 7373 oil as the pressure-transmitting medium [15,16]. Pressures up to 3.2 GPa were reached.

In order to acquire information about formation of magnetic moments in U_2RhIn_8 , we applied the theoretical methods based on the density functional theory. The electronic structure and magnetic moments were calculated using the latest version of APW+lo WIEN2k code [17]. The $5f$ electrons form the Bloch states with non-integer occupation number. The spin-orbit coupling was included using second-order variational step [18]. Since we found the smaller value of the total magnetic moment than expected, we applied the LSDA+U method [17] and tuned the effective U to obtain the required total magnetic moment. The electronic structure calculations were performed at experimental equilibrium. The calculations were ferromagnetic for the sake of simplicity, since we have no information about the character of the antiferromagnetic ground state.

3. Results and discussion

The obtained diffraction patterns revealed the Ho_2CoGa_8 -(HoCoGa_5)-type structure (P4/mmm) for U_2RhIn_8 (URhIn_5). Table 1 summarizes the lattice parameters, atomic coordinates and the equivalent isotropic displacement parameters U_{eq} . The refinement parameters of the obtained data for U_2RhIn_8 equal $R_{\text{int}} = 0.076$, $R[F^2 > 3\sigma F^2] = 0.035$, the largest peak/hole in difference Fourier map $\Delta\rho_{\text{max}} = 5.84 \text{ e}\text{\AA}^{-3}/\Delta\rho_{\text{min}} = -4.04 \text{ e}\text{\AA}^{-3}$. For URhIn_5 : $R_{\text{int}} = 0.041$,

$R[F^2 > 3\sigma F^2] = 0.022$, $\Delta\rho_{\text{max}} = 2.55 \text{ e}\text{\AA}^{-3}/\Delta\rho_{\text{min}} = -2.01 \text{ e}\text{\AA}^{-3}$.

The temperature dependence of the specific heat $C(T)$ divided by temperature for U_2RhIn_8 and URhIn_5 is presented in Fig. 1; a clear λ -shaped anomaly at $T_N = 117 \text{ K}$ and $T_N = 98 \text{ K}$, respectively, indicates a second-order phase transition in both materials. Closer observation of the $C(T)$ vs. T curve of U_2RhIn_8 reveals a small anomaly at $T \sim 100 \text{ K}$, which arises from a tiny amount of URhIn_5 .

Table 1

Lattice parameters, fractional atomic coordinates and isotropic or equivalent isotropic displacement parameters for U_2RhIn_8 and URhIn_5 .

	Atom	x	y	z	$U_{\text{iso}}/U_{\text{eq}}$
U_2RhIn_8					
$a = 4.6056(6) \text{ \AA}$	U	0.5	0.5	0.30883(7)	0.0059(3)
$c = 11.9911(15) \text{ \AA}$	Rh	0.5	-0.5	0	0.0078(6)
	In(1)	0.5	0	0.5	0.0080(5)
	In(2)	0.5	0	0.12263(11)	0.0091(4)
	In(3)	0	0	0.30916(14)	0.0079(4)
URhIn_5					
$a = 4.6210(5) \text{ \AA}$	U	0	0	0	0.00474(19)
$c = 7.4231(7) \text{ \AA}$	Rh	1	0	0.5	0.0059(4)
	In(1)	0.5	0	0.30179(11)	0.0078(2)
	In(2)	0.5	0.5	0	0.0076(3)

The magnitude of the phonon contribution to the specific heat for both ternary compounds was determined from a $C/T = \gamma + \beta T^2$ fit to the data (fit interval $1 \text{ K} < T < 10 \text{ K}$). For the U_2RhIn_8 compound, the value of Sommerfeld coefficient yields $\gamma = 47 \text{ mJ mol}^{-1} \text{ U K}^{-2}$ and the β coefficient equals $3.4 \text{ mJ mol}^{-1} \text{ K}^{-4}$ which corresponds to a Debye temperature $T_D = 150 \text{ K}$. The Sommerfeld coefficient of URhIn_5 equals $60.7 \text{ mJ mol}^{-1} \text{ U K}^{-2}$ while the β coefficient yields the value of $3.3 \text{ mJ mol}^{-1} \text{ K}^{-4}$ corresponding to the Debye temperature of 165 K . The values for URhIn_5 are close to those presented recently [24].

Fig. 1(b) represents data in an applied magnetic field of 9 T for U_2RhIn_8 along the [001] axis. The direct comparison with the zero field measurement reveals that T_N is almost unaffected within experimental uncertainty. Equivalent behavior is observed, Fig. 1(c), in the case of URhIn_5 . Similar response to magnetic field is observed in the structurally related Ce-based compound, CeRhIn_5 , where the transition temperature T_N tends to be rather insensitive to the application of magnetic field along the same direction [25].

Fig. 2 shows the temperature dependence of the $\chi(T)$ and $1/\chi(T)$ of U_2RhIn_8 and URhIn_5 in magnetic field oriented along [100] and [001] directions and [110] and [001] in the case of UIn_3 . The analysis of the $\chi(T)$ data leads to the determination of Néel temperatures as proposed by Fisher [20]; the maximum of the $\partial(\chi(T))/\partial T$ curve. The behavior of the susceptibility curves resembles the one shown for URhIn_5 and UIn_3 [19,5], therefore we conclude that the phase transition drives the compound into an antiferromagnetic state. The magnetic susceptibility increases in all compounds with decreasing temperature and this increase is much pronounced for the [001] direction in the ternary compounds. The maximum value of susceptibility is reached at $T_{\chi\text{max}} = 130 \text{ K}$ for UIn_3 (consistently with the literature [5]), at $T_{\chi\text{max}} = 160 \text{ K}$ for URhIn_5 [24] and at $T_{\chi\text{max}} = 150 \text{ K}$ for U_2RhIn_8 . Such behavior was previously observed in several different uranium compounds [21] and it is generally supposed that this character of the susceptibility curves is associated with antiferromagnetic correlations when approaching T_N . The value of $T_{\chi\text{max}}$ for both studied ternary compounds is the highest among uranium compounds up to our knowledge (UPd_2Al_3 : $T_{\chi\text{max}} = 30 \text{ K}$; URu_2Si_2 : $T_{\chi\text{max}} = 60 \text{ K}$ [22]).

In the vicinity of $T_N = 117 \text{ K}$ (98 K, 88 K) for U_2RhIn_8 (URhIn_5 , UIn_3), a sharp drop of the magnetic susceptibility is observed in agreement with the literature [5,24,19]. This rapid decrease is again more pronounced in the [001] direction in the studied ternary compounds, pointing to the fact that the magnetic moments probably lie in this direction in the ordered state.

At low temperatures ($T \sim 40 \text{ K}$) the susceptibility reaches its minimum value and rises up again. This increase is negligible in the case of URhIn_5 and U_2RhIn_8 ; however, it plays a dominant role

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