On the crystal structure and magnetic properties of new quaternary compounds in the system U-Cr-Si-C

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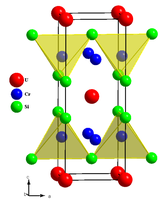
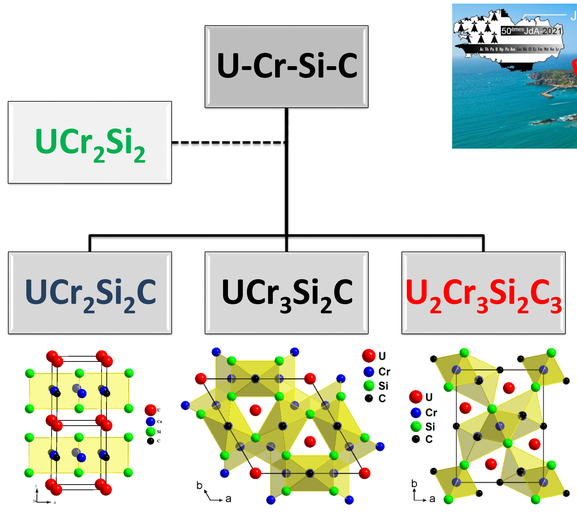
UCr_2Si_2

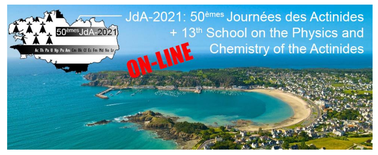
Structural transition at low temperature (T_t = 210 K) from $ThCr_2Si_2$-type ($I4/mmm$) to its own-type ($C\,2/m$) [1]

Antiferromagnetic ordering of the U-sublattice at low temperature (T_N = 27K) [1-3]

Singular structural and magnetic behaviors in comparison to the other RCr_2Si_2 compounds







$U_2Cr_3Si_2C_3$

New compound crystallizing in its own-type [1], a carbon-filled variant of the $Sc_2Pt_3Si_2$-type [7]

Pbam
a = 7.052 Å, b = 9.106 Å, c = 3.957 Å
V = 254.09 Å³

No magnetic ordering detected by specific heat, magnetic measurement or NPD

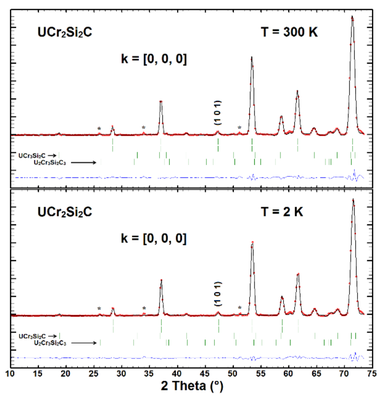
→ $d_{U\text{-}U}$ relatively short (3.275 Å), well bellow the Hill limit (3.5 Å)

→ $d_{Cr\text{-}Cr}$ (2.812 Å) equivalent to those of UCr_2Si_2C (2.816 Å) but very short $d_{Cr\text{-}C}$ bonds (1.908 Å) suggesting strong chemical bonds

UCr_2Si_2C

P4/mmm
a = 3.983 Å, c = 5.160 Å
V = 81.84 Å³

New member of the RCr_2Si_2C series crystallizing in the $CeCr_2Si_2C$-type [4], a carbon-filled variant of the $CeMg_2Si_2$-type



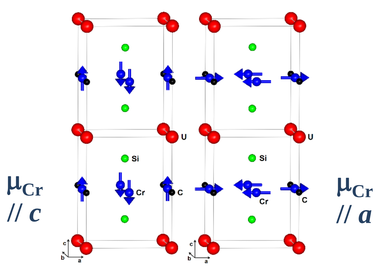
Specific heat and magnetic measurements suggest a Pauli paramagnetic behavior

Neutron powder diffraction reveals a magnetic contribution to the nuclear Bragg peak (1 0 1) from 300 K

→ AF ordering of the Cr-sublattice (AFl-type) at high temperature

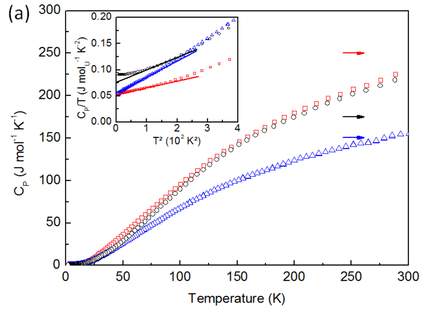
→ No magnetic ordering detected on the U-sublattice

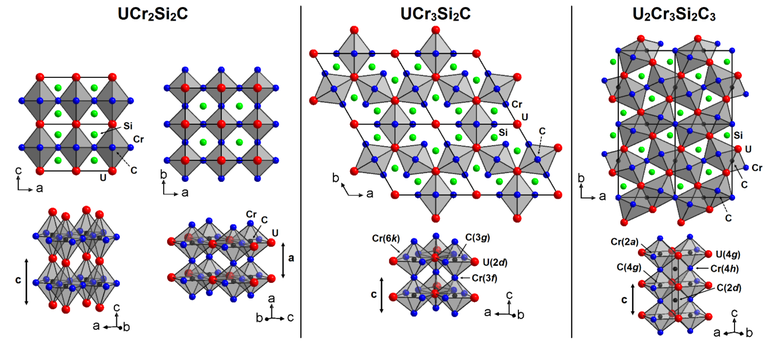
- Magnetic SG $P\bar{1}$
- k = [0, 0, 0]
- $\overrightarrow{m_{Cr}}$ coupled AF in the (a,b) plan and F along the c axis



R_{magn} values in favor of a planar orientation of the $\overrightarrow{m_{Cr}}$

	300 K // c	300 K // a	2 K // c	2 K // a
μ_{Cr} (μ_B)	0.56(5)	0.66(5)	0.58(5)	0.69(5)
χ^2	21.3	20.9	60.3	58.5
R_{magn}	14.1	**3.2**	19.8	**7.9**





UCr_3Si_2C

Member of the RCr_3Si_2C series crystallizing in the YCr_3Si_2C-type [5], a carbon-filled variant of the YCo_3Ga_2-type [6]

P6/mmm
a = 8.968 Å, c = 4.003 Å
V = 278.78 Å³

No magnetic ordering detected by specific heat, magnetic measurement or NPD

→ $d_{U\text{-}U}$ relatively long (4.003 Å) but structural disorder on the U-sublattice

→ $d_{Cr\text{-}Cr}$ relatively short (2.533 Å) suggesting strong chemical bonds

- Crystal structure and stability of UCr_2Si_2C, UCr_3Si_2C and $U_2Cr_3Si_2C_3$ based on the full occupancy of interstitial sites by carbon atoms (i.e. C-filled variant of ternary U-Cr-Si compounds and frameworks related to CU_2Cr_4, CU_3Cr_3 and CU_4Cr_2 octahedron)

- No magnetic transition detected by specific heat and magnetic measurements

- Magnetic behavior of UCr_2Si_2C inverse in comparison to the others RCr_2Si_2C compounds

- Absence of magnetic ordering in UCr_3Si_2C and $U_2Cr_3Si_2C_3$ explains by crystal chemistry investigations

[1] P. Lemoine, A. Vernière, M. Pasturel, G. Venturini, B. Malaman, *Inorg. Chem.* **2018**, 57, 2546 ; [2] T.D. Matsuda, N. Metoki, Y. Haga, S. Ikeda, T. Okubo, K. Sugiyama, N. Nakamura, K. Kindo, K. Kaneko, A. Nakamura, E. Yamamoto, Y. Onuki, *J. Phys. Soc. Jpn.* **2003**, 72, 122 ; [3] K. Hiebl, P. Rogl, C. Horvath, K. Remschnig, H. Noël, *J. Appl. Phys.* **1990**, 67, 943 ; [4] C. Tang, S. Fan, M. Zhu, *J. Alloys Compd.* **2000**, 299, 1 ; [5] P. Lemoine, J. Tobola, A. Vernière, B. Malaman, *J. Solid State Chem.* **2013**, 201, 293 ; [6] M.A. Fremy, D. Gignoux, J.M. Moreau, D. Paccard, L. Paccard, *J. Less-Common Met.* **1985**, 106, 251 ; [7] A. Gribanov, A. Grytsiv, P. Rogl, Y. Seropegin, G. Giester, *J. Solid State Chem.* **2010**, 183, 1278