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Electronic Supplementary Information for:

Flux Synthesis, Structure, Properties, and Theoretical Magnetic Study of Uranium (IV) Containing *A*2USi6O15 (*A* = K, Rb) with an Intriguing Green-to-Purple, Crystal-to-Crystal Structural Transition in the K Analogue

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Supplemental Table S1 Calculated ligand field potential (in cm^{-1}) obtained from the LFDFT calculation of the $(UO_6)^{-8}$ cluster in the high and the two U site of the low temperature structures of $K_2USi_6O_{15}$.

B_q^k	**High T structure**	**U(1) site Low T structure**	**U(2) site Low T structure**
B_0^2	413.41	1717.38	794.33
B_1^2	27.36+*i*286.87	-961.82+*i*1242.22	105.24+*i*967.93
B_2^2	-1087.83+*i*1118.52	2428.25+*i*92.31	1711.54+*i*93.38
B_0^4	8499.87	7481.32	9065.17
B_1^4	-7269.73+*i*7761.63	-8174.80+*i*7134.06	5904.72+*i*6728.88
B_2^4	857.03+*i*5676.61	79.58+*i*6994.80	-622.66+*i*4899.01
B_3^4	-1301.06+*i*3477.06	-230.80+*i*3148.32	-279.64+*i*3291.90
B_4^4	-8008.20+*i*4704.67	-7396.41+*i*1358.02	-6054.82+*i*550.24
B_0^6	-157.66	-521.03	-51.33
B_1^6	-355.80+*i*355.39	-524.93+*i*381.92	697.12+*i*534.21
B_2^6	-299.39+*i*75.83	-58.53+*i*340.74	-318.75+*i*38.98
B_3^6	430.72+*i*763.18	559.36+*i*1465.87	-464.51+*i*723.34
B_4^6	294.39+*i*398.36	657.59+*i*408.55	539.70+*i*450.98
B_5^6	-642.81+*i*291.19	-927.13+*i*49.44	443.76+*i*63.61
B_6^6	439.71+*i*34.58	-761.83+*i*878.80	-559.33+*i*356.21

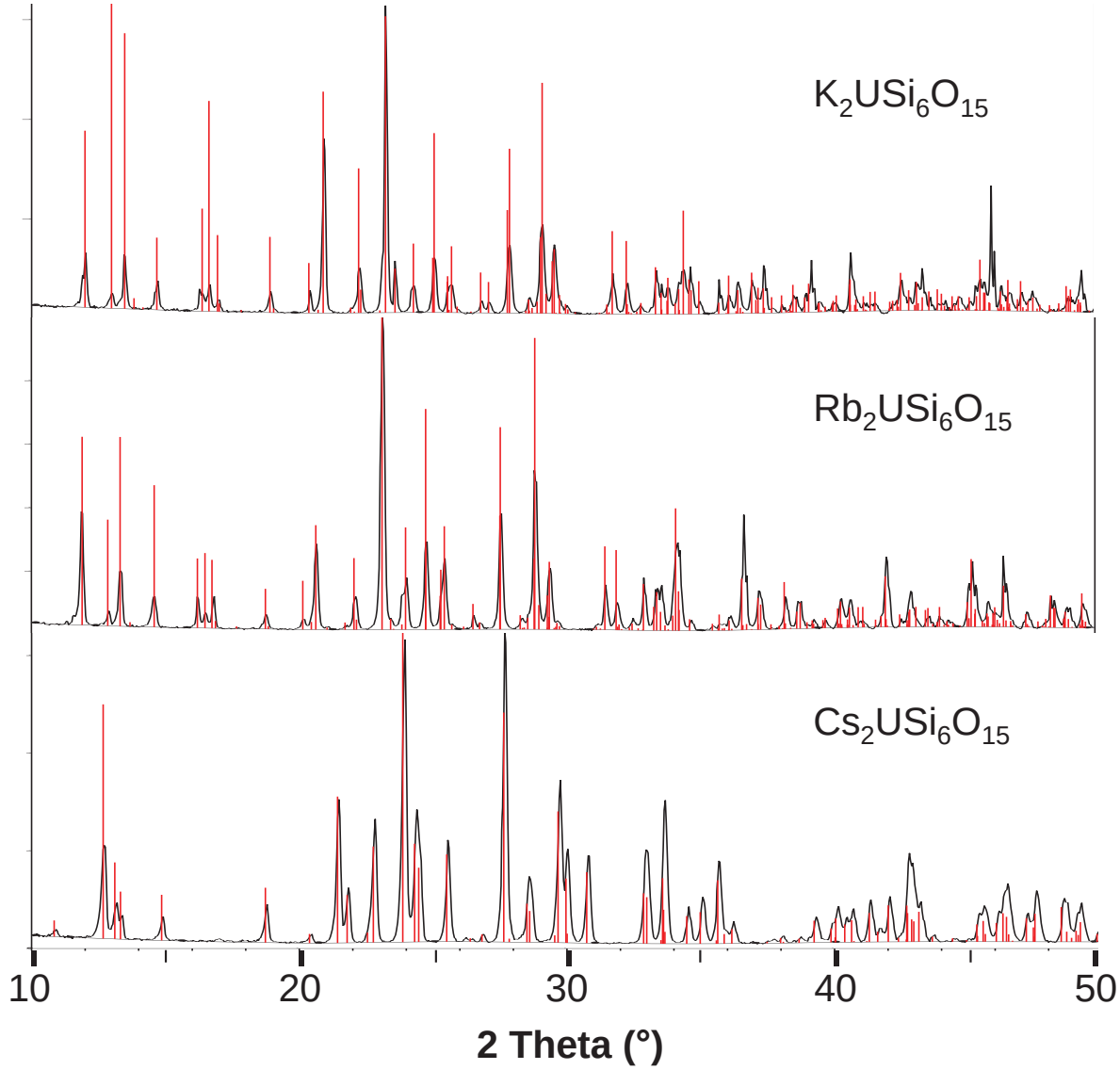

Figure S1 Powder diffraction patterns of $A_2USi_6O_{15}$ (*A* = K, Rb, Cs) with the calculated diffraction peaks shown in red.

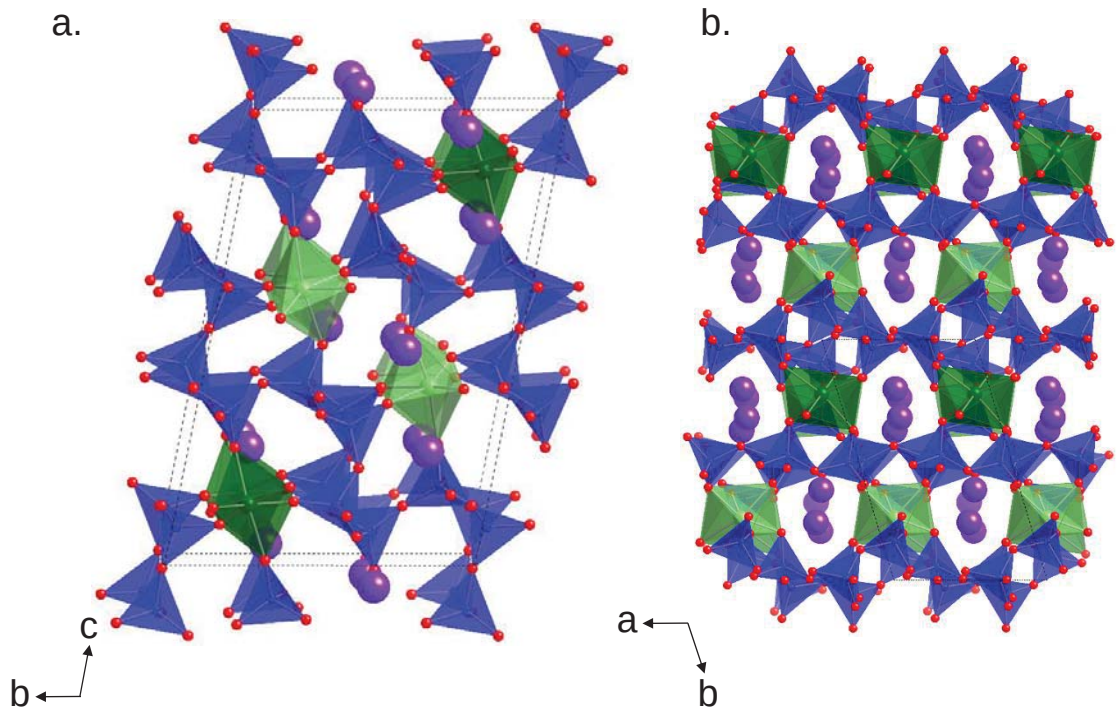

Figure S2 Low temperature structure of $K_2USi_6O_{15}$ showing a) the three dimensional connectivity of the uranium and silicon polyhedra and b) the *c*-direction highlighting the channels in the framework. Uranium atoms are shown in green, silicon in blue, potassium in purple, and oxygen in red.

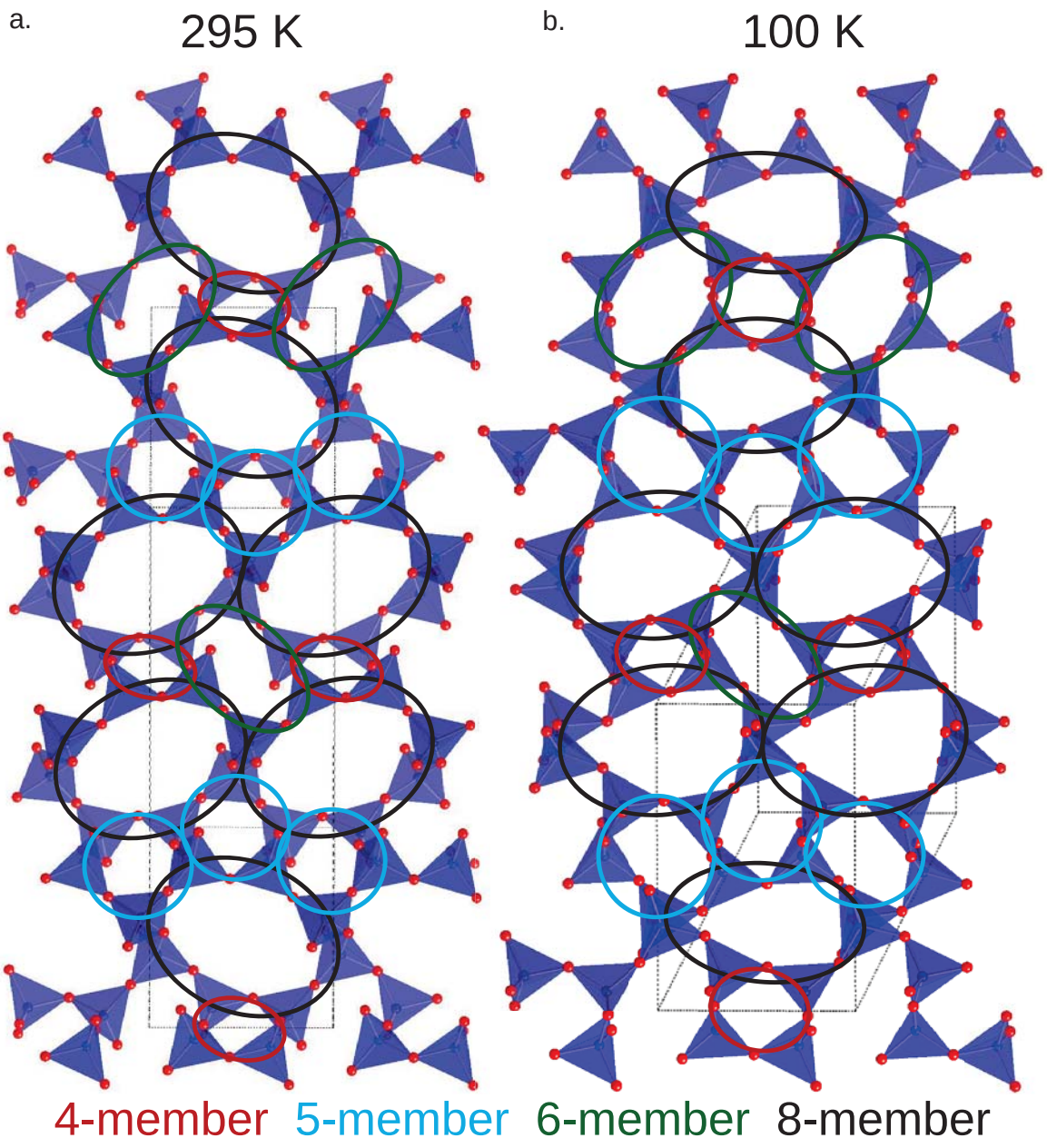

Figure S3 The silicate slabs in the a) room temperature and b) low temperature structures of $K_2USi_6O_{15}$. The silicate rings are color coded to highlight the relationship between the two slabs, see legend.

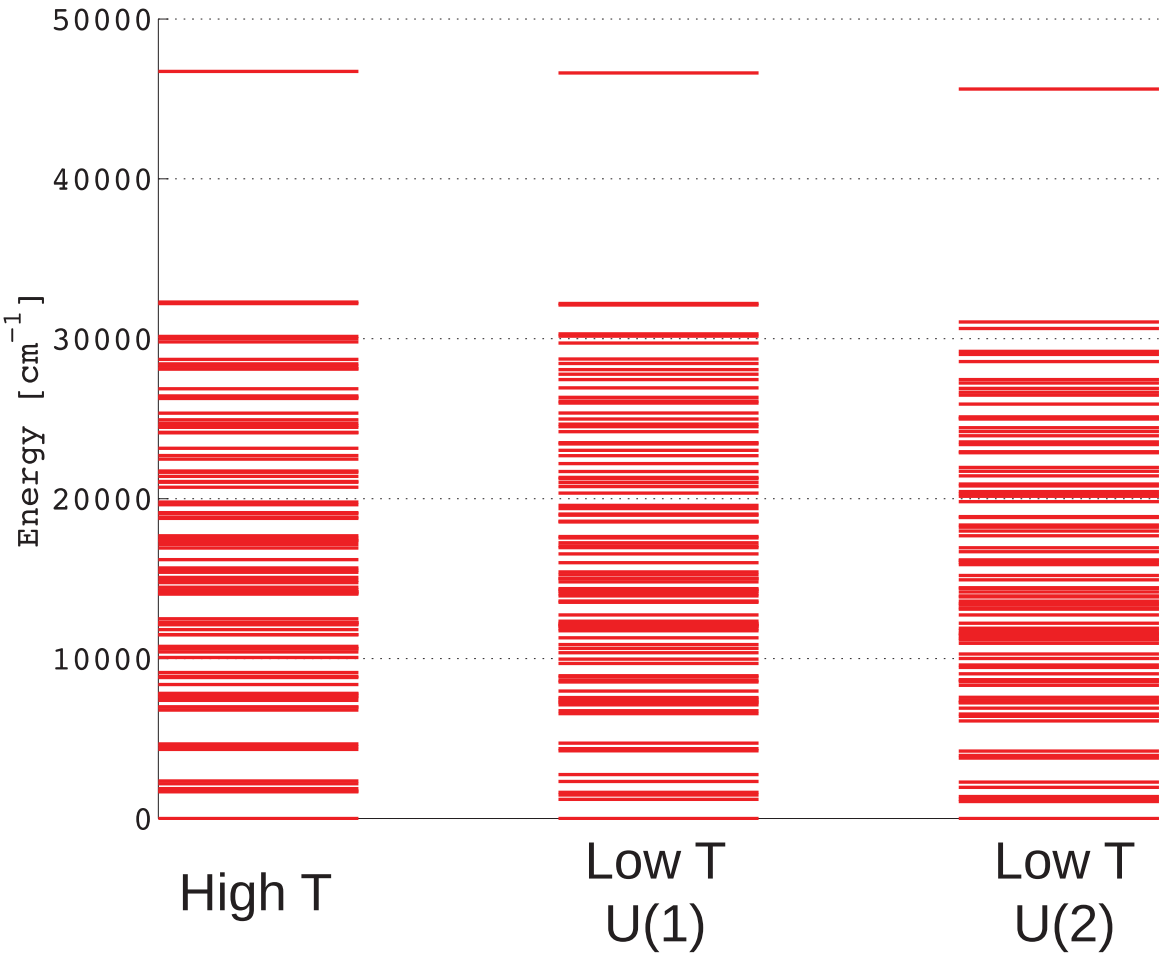

Figure S4 Calculated LFDFT multiplet energy levels obtained for $(UO_6)^{-8}$ from the structure belonging to the high temperature U site (left hand side) and the low temperature U(1) site (middle) and U(2) site (right hand side) of $K_2USi_6O_{15}$.