Uncovering Stacking Disorder in the Weyl Semimetal $TaRhTe_4$

F. Eder[1], B. Stöger[2], E. Giannini[1], F. O. von Rohr[1]

[1] University of Geneva, Department of Quantum Matter Physics, Quai Ernest-Ansermet 24, 1211 Geneva, Switzerland,
[2]Technical University of Vienna, X-ray Centre, Getreidemarkt 9, 1060 Vienna, Austria

felix.eder@unige.ch

Topological van-der-Waals materials like $TaIrTe_4$ are of growing interest for their exotic electronic properties and exfoliable structures. The related $TaRhTe_4$ has been identified as isotypic to the Ir variant [1], but no structural determination on single crystals has been published so far. Both phases were synthesized in a Te self-flux synthesis [2], in which the educts are heated in an excess of Te to 1000 °C, slowly cooled to the quenching temperature T (600–750 °C), and then quenched by hot-centrifugation to remove excess Te-flux.

Single crystal X-ray diffraction (SXRD) patterns of $TaRhTe_4$ crystals, exhibit a different unit-cell compared to previous reports, accompanied by diffuse streaks along the **c*** direction (Fig. 1 (a)). These can be rationalized by stacking disorder and application of OD-theory [3]. The crystal structure is formed by one non-polar layer type A, which has $P12_1/m(1)$ symmetry (Fig. 1(b)). Neighboring layers are mirrored by $m_{[100]}$ and shifted by either $0.214\mathbf{a} + 0.5\mathbf{b}$ (A^+) or $-0.214\mathbf{a} + 0.5\mathbf{b}$ (A^-) within the layer plane, and the resulting layer pairs are symmetrically equivalent. From this, two maximum degree of order (MDO) polytypes [4] can be derived. We observed for $TaRhTe_4$ the MDO_1 stacking (Fig. 1(c)), despite $TaIrTe_4$ and $TaRhTe_4$ having been reported to follow the MDO_2 stacking (Fig. 1(d)).

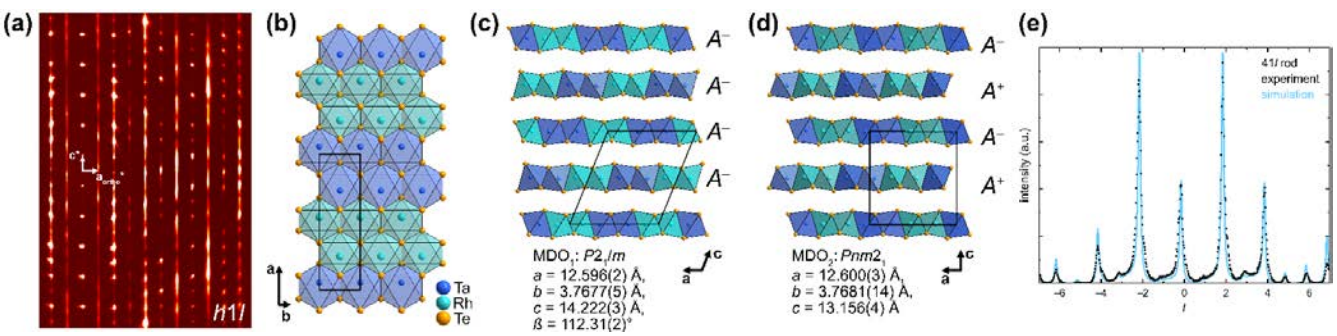

Figure 1. (a) reconstructed $h1l$ plane of $TaRhTe_4$ in orthorhombic setting; (b) monolayer of $TaRhTe_4$ (c, d) the two MDO stackings and their lattice parameters determined at 300 K; (e) measured intensities and intensities calculated with DIFFaX [5] for the $41l$ rod;

When simulating the intensities of the observed diffuse scattering using DIFFaX [5], it was found that two sets of probability parameters, p/q and r/s, were needed. p/q describe the preference for either MDO_1 (p) or MDO_2 ($q = 1–p$) layer triples, while r/s ($s = 1–r$) introduce a potential general A^+ vs. A^- preference. Surprisingly, the best agreement with experimental data was achieved with $p = 0.3$ and $r = 0.85$ (Fig. 1(e)). Concerning the two possible layer triples, there exists a preference for MDO_2 relative to MDO_1, but a stronger external influence parameter, possibly local temperature or concentration gradients, shifts the layer stacking in the crystal to the MDO_1 polytype.

Stacking disorder was observed consequently over crystals from numerous batches. Hereby, we identified the quenching temperature T as a possible influence parameter, a lower T leads to a preference for the orthorhombic MDO_2 stacking in $TaRhTe_4$. For $TaIrTe_4$, we likewise detected diffuse scattering following the MDO_2 stacking; hereby, the degree of stacking disorder is decreasing when reducing T.

In this talk, I will establish a disorder model guided by OD-theory, present the influence of p/q and r/s on the simulated patterns and highlight, how the different stacking orders can be controlled by the synthetic conditions. Our results demonstrate how stacking order in quantum materials can be tuned through synthetic control, offering a new handle on structural engineering at the atomic scale.

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