

Supplementary Information

Thermodynamically Guided Synthesis of 3R-TaSe₂ Nanocrystals and Their Superconducting Behavior

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Thermodynamic Simulations

Thermodynamic simulations were performed with the software “TRAGMIN 5.1” [1]. 9.4 mL volume, 5×10^{-9} mol H₂O, 10^{-9} mol Ar traces, 0.01 mmol TaSe₂, and 0.1 mmol iodine were used for the simulations.

The thermodynamic data used for the simulations were all adapted from the FactPS database, except for the case of TaSe₂ (s). We did not find data in the literature for TaSe₂ (s), therefore, the data was estimated as follows. $\Delta_f H^\circ$ was estimated to -230 kJ/mol based on the value of TaS₂ (s) of -353.966 kJ/mol [2] and assuming an increase in enthalpy from the S to the Se variant similar to what reported for other 2D TMD's such as Ti, Zr, Mo, Nb and W [2]. An error of about ± 30 kJ/mol for the estimated enthalpy of TaSe₂ (s) is to be expected by this method. However, even simulation with such deviating values did not change the qualitative results in a relevant way. S° and the C_p -function of TaSe₂ (s) were estimated to be equal to the values of WSe₂ [2] (90 J/(mol·K) and $C_p(T) = 71.086 \cdot T + 14.627 \cdot 10^{-3} + 0.253 \cdot 10^{-6} \cdot T^{-2} - 4718 \cdot T^{-1} - 125.2 \cdot T^{-0.5}$), as WSe₂ and TaSe₂ are isostructural and only have a difference in molar mass of 2.89 u, which is a good indicator for very similar values for S° and C_p according to similar comparisons of related compounds [2].

AFM images

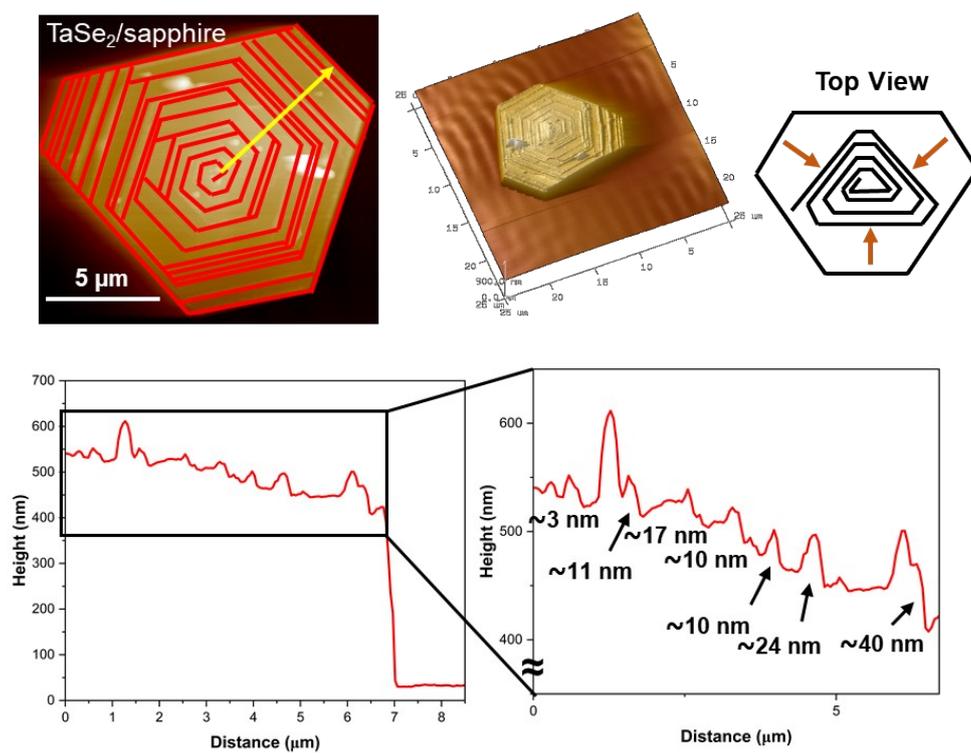


Figure S1. AFM image with height profile for continued growth of a TaSe₂ microcrystal on *c*-sapphire substrate. TaSe₂/sapphire typically exhibited a hexagonal spiral morphology, with varying thicknesses for each step. This progressive increase in height eventually developed into microcrystals.

SEM - EDX

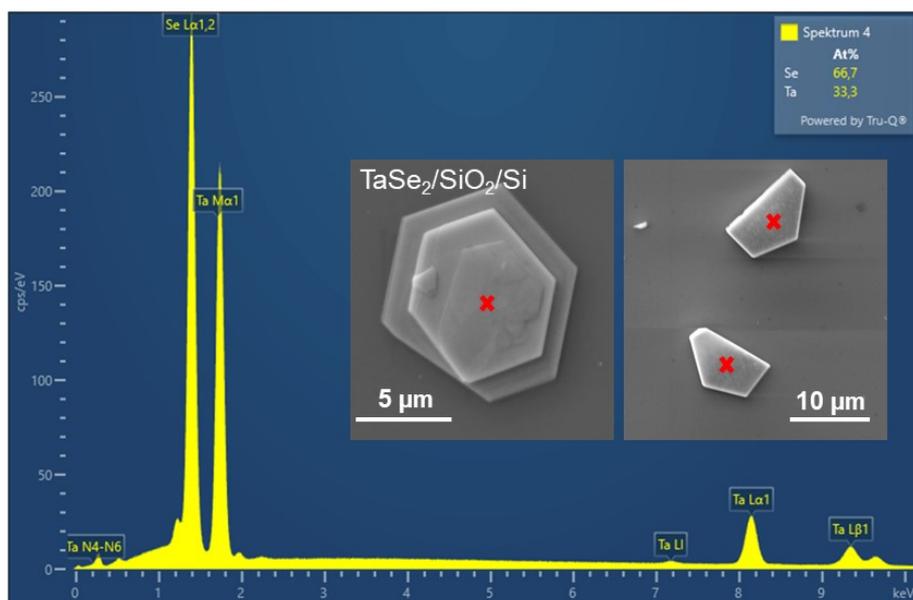


Figure S2. EDX image of 3R-TaSe₂ nanocrystals on SiO₂/Si substrate (Growth time = 6 h). Inset: SEM images of the analysed crystals; the red crosses indicate the positions of the EDX measurements.

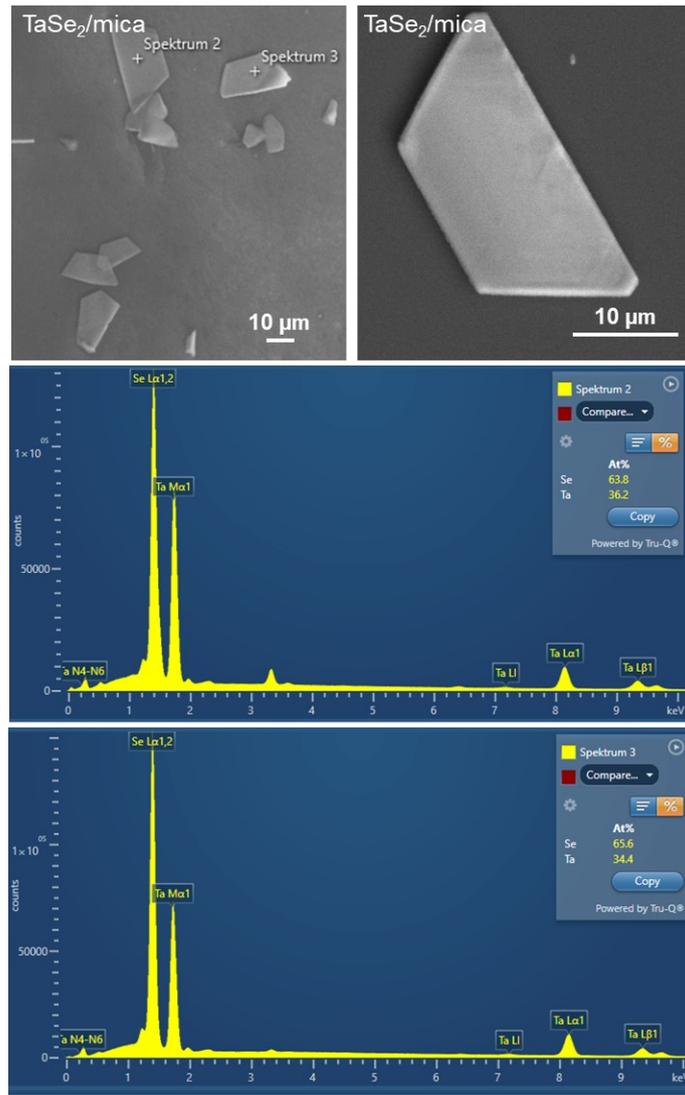


Figure S3. SEM-EDX images of TaSe₂ nanocrystals on mica substrate (Growth time = 30 minutes). The white crosses indicate the positions of the EDX measurements.

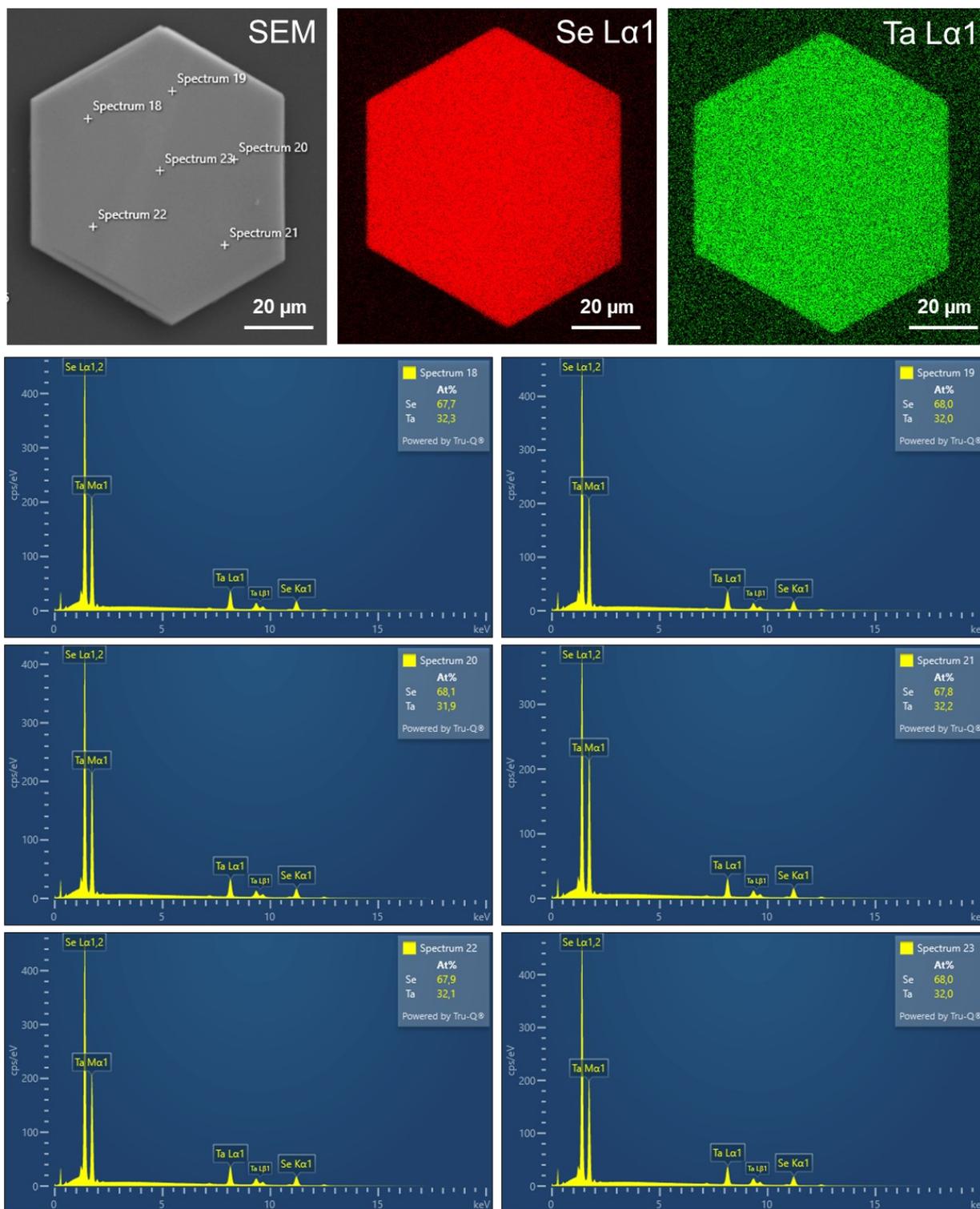


Figure S4. Structural and compositional homogeneity of CVT-grown 3R-TaSe₂ nanocrystals. SEM image of a representative hexagonal 3R-TaSe₂ crystal with marked EDX acquisition points, together with corresponding elemental maps of Se (Se La₁) and Ta (Ta La₁). The maps reveal a uniform spatial distribution of both elements across the entire crystal area, confirming chemical homogeneity.

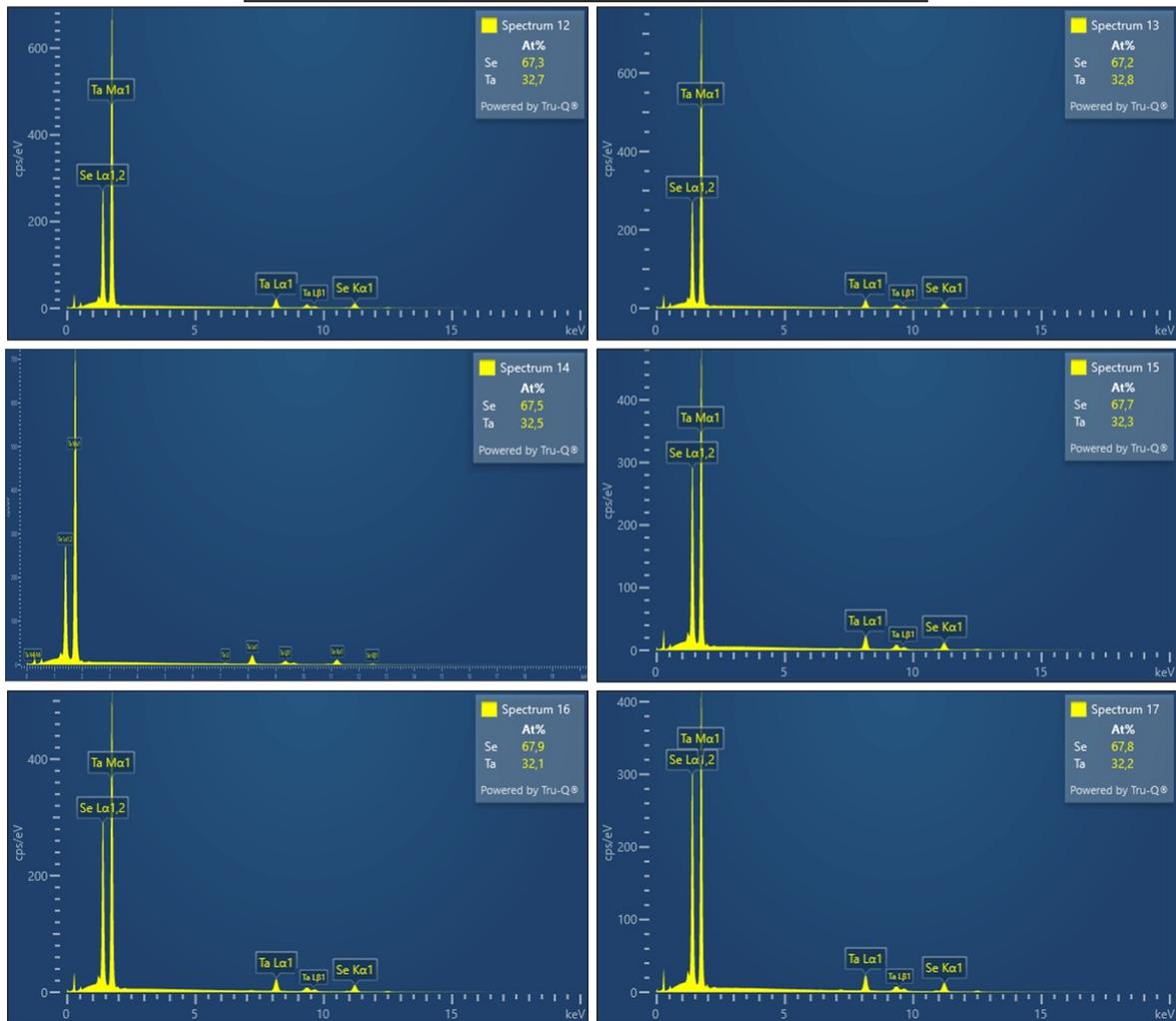
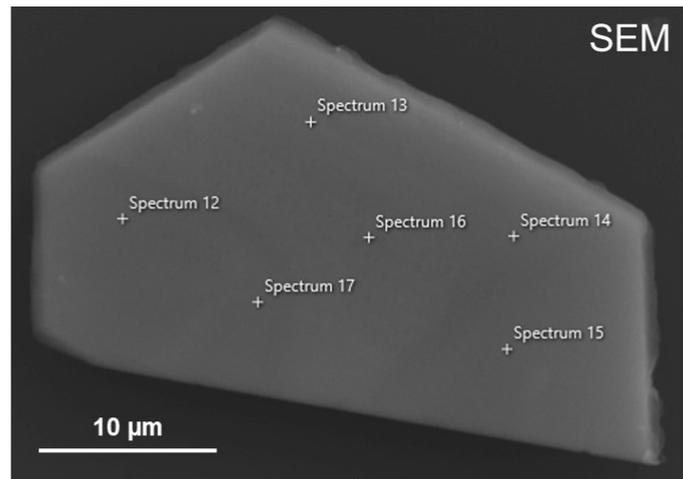


Figure S5. SEM image of a representative hexagonal crystal with marked EDX acquisition points.

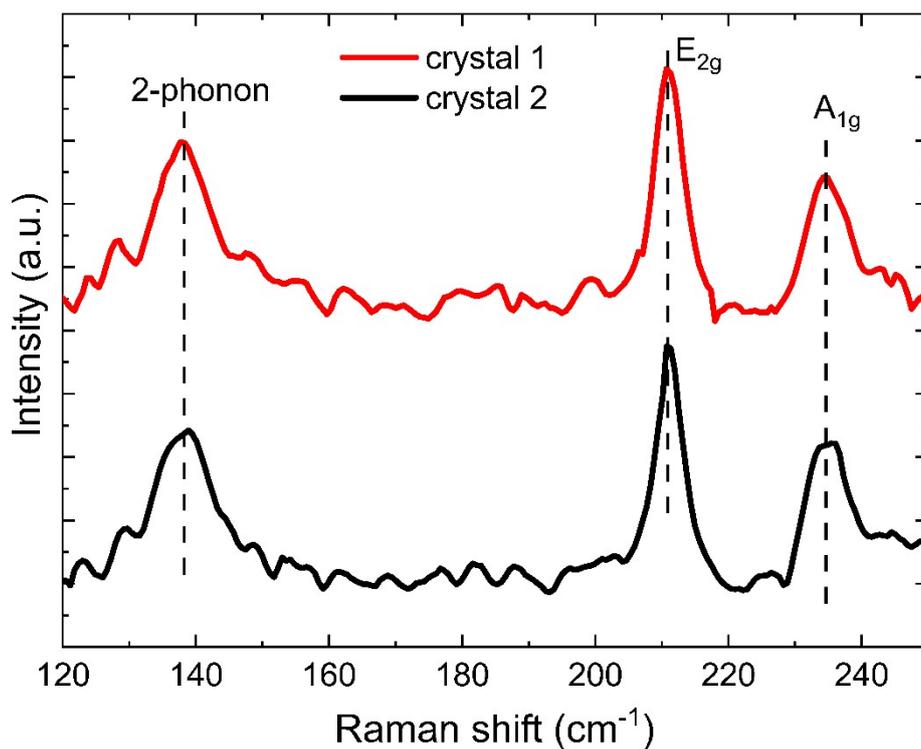


Figure S6. Raman spectra of two different TaSe₂ hexagonal crystals deposited on SiO₂/Si substrate (grown under the same conditions). The peaks at $\sim 234.5 \text{ cm}^{-1}$, $\sim 211 \text{ cm}^{-1}$, and 138.5 cm^{-1} correspond to out-of-plane vibrational mode, in-plane vibrational mode, and two-phonon process, respectively, matching well with the literature on 3R-TaSe₂ [3].

References

1. Krabbes, G., et al., *Programmpaket TRAGMIN, Version 5.1*. 2014, IFW Dresden, TU Dresden, HTW Dresden.
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3. Deng, Y., et al., *Controlled growth of 3R phase tantalum diselenide and its enhanced superconductivity*. Journal of the American Chemical Society, 2020. **142**(6): p. 2948–2955.