Supplementary Information

Metallic *vs.* Semiconducting Properties of Quasi-One-Dimensional Tantalum Selenide van der Waals Nanoribbons

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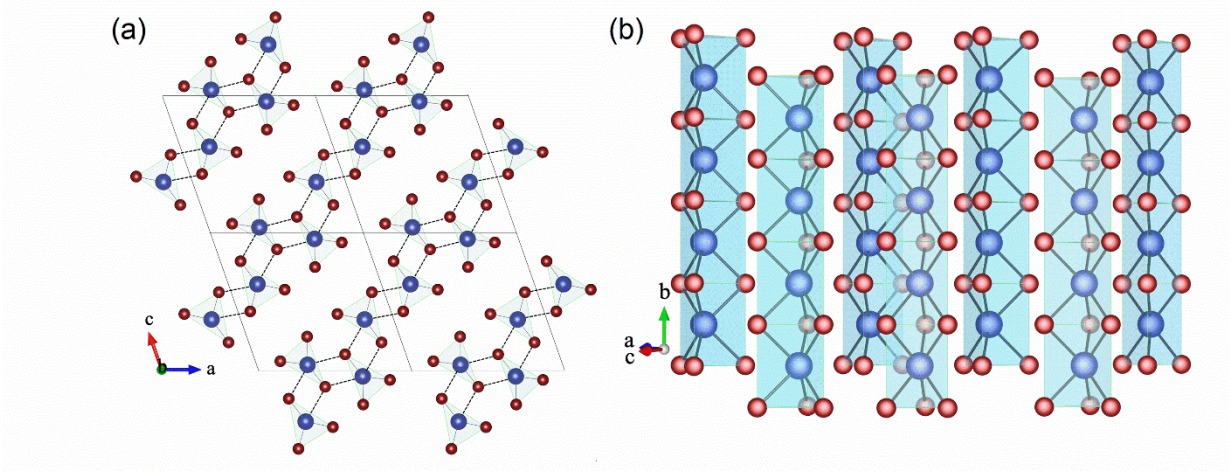

Figure S1: (a-b) Schematic of the crystal structure of quasi-1D $TaSe_3$ and the triangular prisms forming parallel van der Waals chains from different views. The red and blue circles represent Ta and Se atoms.]

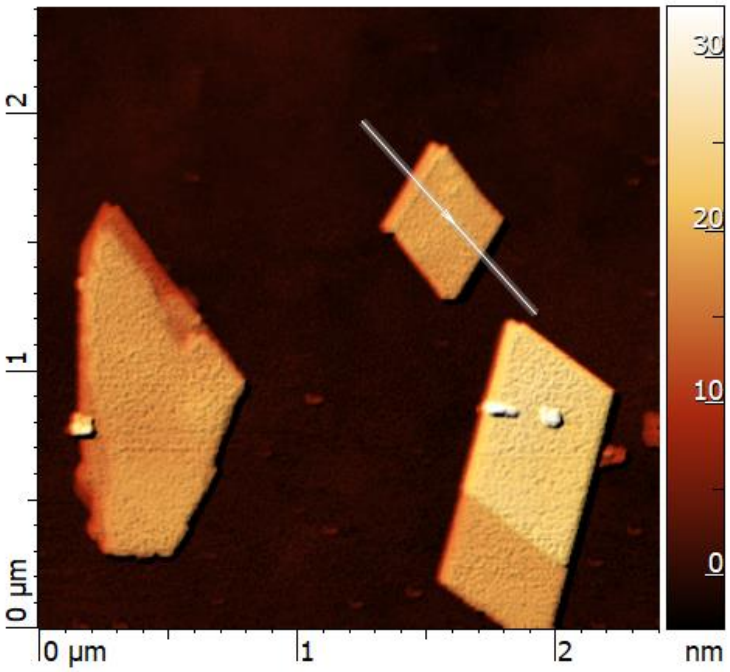

Figure S2: AFM image of the sample used in Figure 3a.

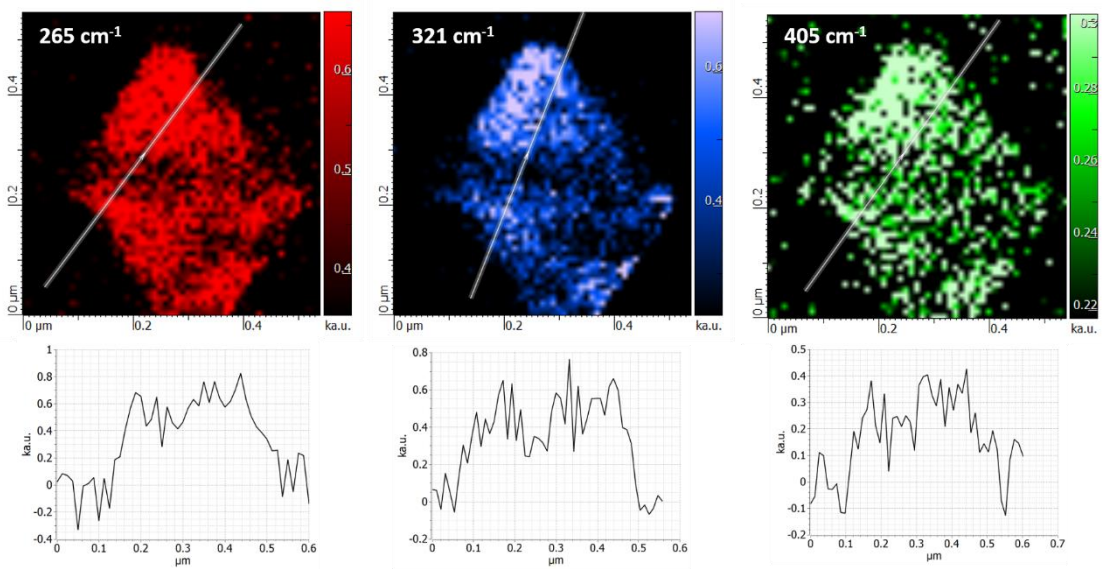

Figure S3: Individual TERS maps of the Raman peaks at 265 cm^{-1}, 321 cm^{-1}, and 405 cm^{-1} bands.

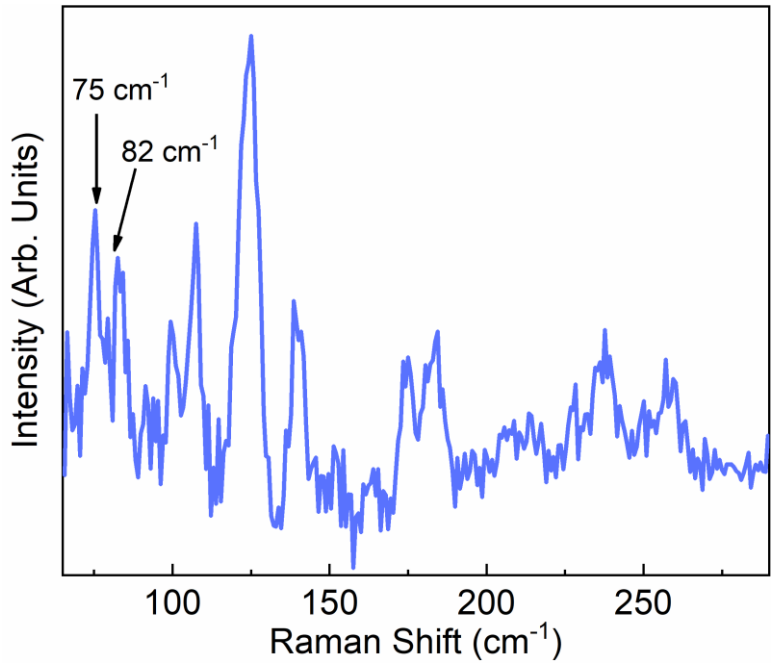

Figure S4: (a-b): Low-wave-number Raman experiments showing the low-frequency Raman modes at 75 cm^{-1} and 82 cm^{-1}.

Table S1: Raman active modes of TaSe3 in the range of 65 cm^{-1} to 270 cm^{-1}		
Experiment	Theory	Symmetry
75	74.5	Bg
82	82.7	Ag
-	107.5	Ag
114	116.7	Ag
-	123.4	Bg
127	127.6	Bg
-	133.4	Bg
-	138.1	Bg
142	140.1	Bg
-	146.4	Bg
152	155.4	Ag
166	168.0	Ag
174	173.2	Bg
187	185.3	Ag
-	191.7	Bg
	201.8	Ag
213	216.8	Ag
-	224.9	Ag
229 (very weak)	228.3	Ag
-	239.9	Ag
264	269.4	Ag

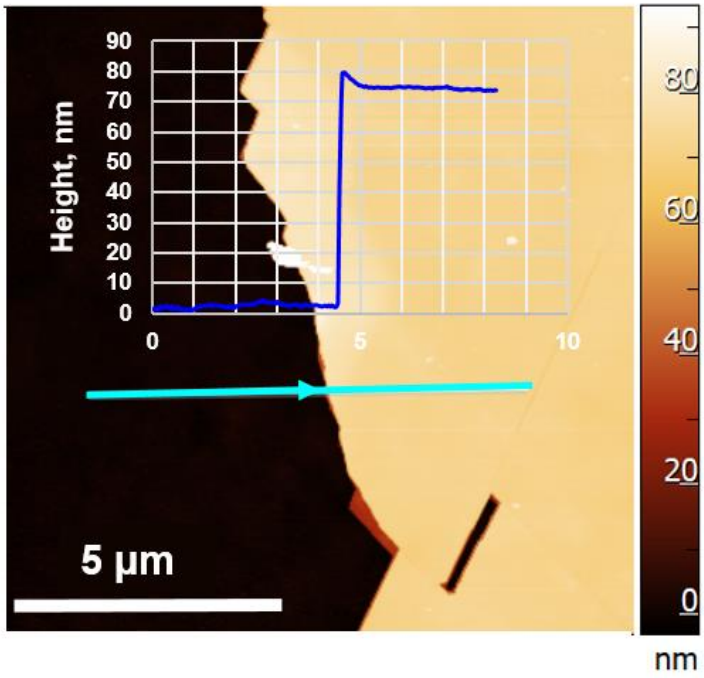

Figure S5: (a) AFM topography image (12 µm × 12 µm) of a large $TaSe_{2.75}$ crystal on a template stripped gold substrate. In the insert-section graph along the cyan line showing that the height of the crystal was about 70 nm.

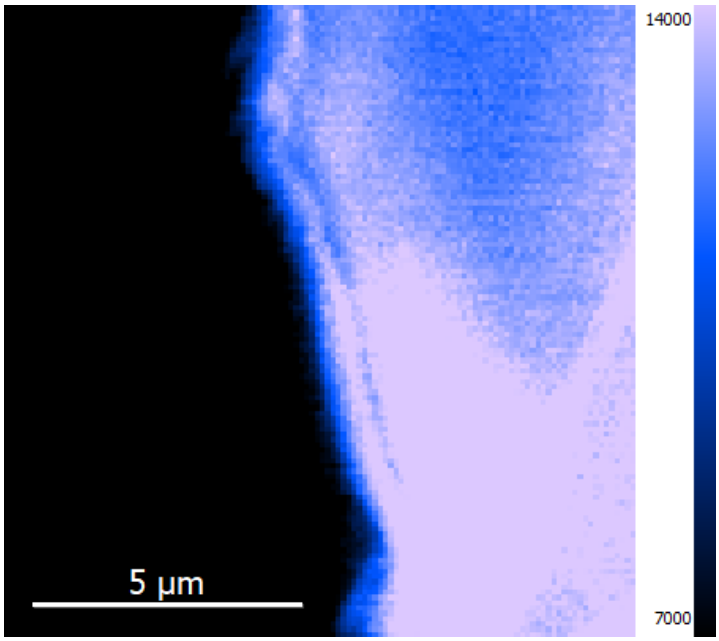

Figure S6: Confocal PL map of the crystal used in PL measurement.]

TERS measurement procedure in DualTwoPass^{TM} mode

As the TERS signal gets strongly enhanced when the TERS probe is in direct contact with the sample, TERS measurements presented in current study were performed using proprietary DualTwoPass imaging mode. The measurement procedure was the following: in the beginning of each line of the TERS map the AFM was operating in AC (tapping) mode. Then the feedback was switched to the contact mode with normal force exerted on the samples of about 100 nN, and this contact mode feedback was maintained for the duration of the line. A full Raman spectrum was collected in each pixel along the line with integration time ranging from a few hundred milliseconds to a few seconds per pixel. At the end of each line the feedback was switched to AC (tapping) operation, the sample was moved to the beginning of the next line, switched to contact mode and so forth. Since in every pixel of the map a full spectrum was collected, TERS maps could be rendered as the intensity or the peak position maps of up to three different peaks.

When regular confocal Raman signal was to be collected along with the TERS signal, after the contact mode pass, the same line was scanned again in AC (tapping) mode when the Raman signal was of purely far field nature. Thus, we collected simultaneously two maps- a far field map and the map containing both the TERS contribution and (usually much weaker) the far field signal. Subtracting the far field map from the combined map we could obtain the pure TERS (near field) map.

DOS calculation:

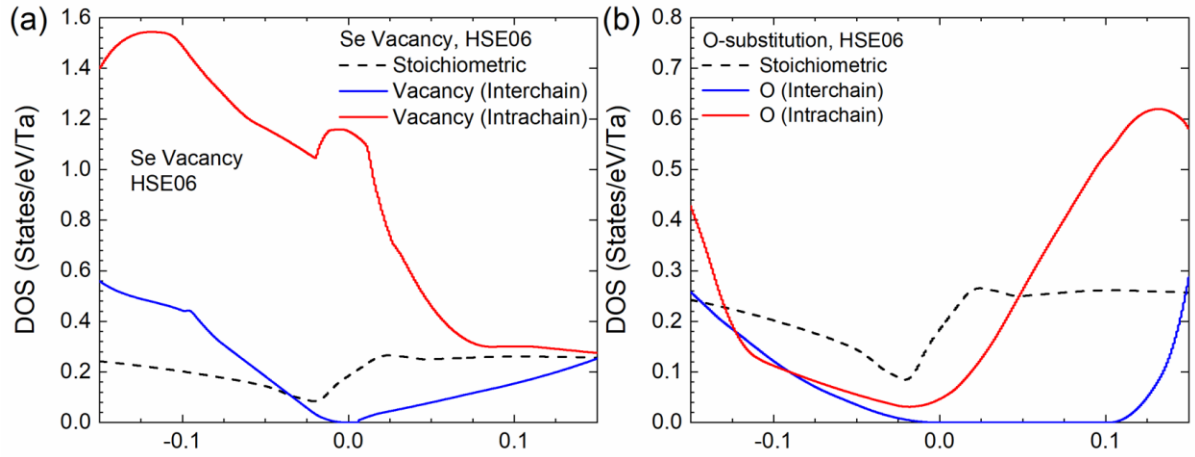

Figure S7: Electronic Density of states for (a) Se deficient structure $TaSe_{2.75}$, (b) oxygen substituted structure $TaSe_{2.75}O$ with HSE only (no SOC).