

Metal-Organic Chemical Vapor Deposition of PtSe₂

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Abstract

Platinum diselenide (PtSe₂), a novel two-dimensional material from the class of noble-metal dichalcogenide (NMD), has recently received significant attention due to its outstanding properties. PtSe₂, which undergoes a semi metallic to semiconductor transition when thinned, offers a band-gap in the infrared range and good air stability. These properties make it a prime active material in optoelectronic and chemical sensing devices. However, a synthesis method that can produce large-scale and reliable high quality PtSe₂ is highly sought after. Here, we present PtSe₂ growth by metal organic chemical vapor deposition. Films were grown on a variety of centimeter scale substrates and were characterized by Raman, X-ray photoelectron and X-ray diffraction spectroscopy, as well as scanning tunneling microscopy and spectroscopy. Domains within the films are found to be up to several hundred nanometers in size, and atomic scale measurements show their highly ordered crystalline structure. The thickness of homogenous films can be controlled via the growth time. This work provides fundamental guidance for the synthesis and implementation of high quality, large-scale PtSe₂ layers, hence offering the key requirement for the implementation of PtSe₂ in future electronic devices.

1. Introduction

Two dimensional (2D) materials are the rising star of current materials research. Two element compound 2D materials with the general formula MX_2 (M = Metal, X = chalcogen) – the transition metal dichalcogenides (TMDs) – began to raise increasing interest as they offer unique electronic^{1,2}, photonic³, optoelectronic⁴ and sensing properties^{5,6}. One of the most recently research material class within the field of 2D materials are the noble-metal dichalcogenides. For those materials, the metal (M) is typically either palladium or platinum. NMDs like PdSe_2 or PtSe_2 offer, in addition to their unique properties⁷⁻¹¹, high-stability in air^{12,13} which is a key requirement for device integration.

One special material within the NMD family is PtSe_2 offering in total six possible crystallographic phases¹⁴ with the 1T phase being the most commonly experimentally observed. Notably, it undergoes a semimetal to semiconductor transition when thinned down^{15,16} along with a band gap in the infrared (IR) range^{7,17} and carrier mobilities¹² which is among the top performers when compared to other TMD materials. PtSe_2 has shown to be a prime material for chemical sensing devices^{6,18}, optoelectronic IR waveguide detectors¹⁹ but has also proven its viability when used as transistor¹⁵ or in thermoelectric devices due to its high Seebeck coefficient²⁰. Nevertheless, a high-quality, large-scale and reproducible synthesis approach of PtSe_2 is still highly sought after, potentially exploiting the materials unique properties in modern devices.

To date, a few synthesis techniques for thin PtSe_2 layers have been reported, which are mainly chemical vapor deposition²¹⁻²⁴ (CVD) and thermal assisted conversion (TAC)^{6,12,18,19}. In TAC pre-deposited Pt layers are converted with Se vapor to PtSe_2 which allows growth of continuous films with controlled thickness on the large scale. TAC synthesis can be performed at low temperatures allowing even synthesis on flexible substrates²⁵. Conformal and selective deposition has been reported with TAC in combination with atomic layer deposition of the Pt film. However, TAC grown films⁶ suffer from a low crystallinity with the individual PtSe_2 domains in the films having typically sizes of a few nanometers^{18,25,26} which sets a fundamental limit to the film quality, and, furthermore it is challenging to yield monolayer films. CVD growth of PtSe_2 yields monolayer individual flakes, in particular when using crystalline substrates such as sapphire substrates²¹⁻²⁴, however, it remains challenging to grow continuous films at larger scales. Hence, an improved growth technique is required, that combines the layer quality of CVD with the versatility of TAC synthesis.

In this work, we present for the first time the metal-organic (MO)CVD growth of PtSe_2 which is a highly reproducible synthesis route combining a MO-Pt-precursor source and H_2Se to scalable grow high-quality PtSe_2 layers on a variety of commonly used substrates such SiO_2 , c-plane sapphire and pyrolytic carbon (PyC). A custom-built synthesis setup fully encapsulated in a glovebox was used for the PtSe_2 growth, allowing versatility for deposition and the growth on areas up to 6" wafers. The quality of the grown films is evaluated using Raman spectroscopy, X-ray photoelectron spectroscopy (XPS), atomic force microscopy (AFM), X-Ray diffraction spectroscopy (XRD)

which together demonstrate high material quality. Furthermore, the atomic and electronic structure of the deposited layers is investigated by scanning tunneling microscopy (STM) and spectroscopy (STS), which reveal domain sizes within the layers of up to 300 nm. Within those domains, a defect free growth was visible in STM measurements. This study should give a growth and integration route of high-quality, large-scale PtSe₂ layers, laying the cornerstone for integration of PtSe₂ into modern devices.

2. Results and Discussion

2.1: Synthesis of large-scale PtSe₂ on various substrates

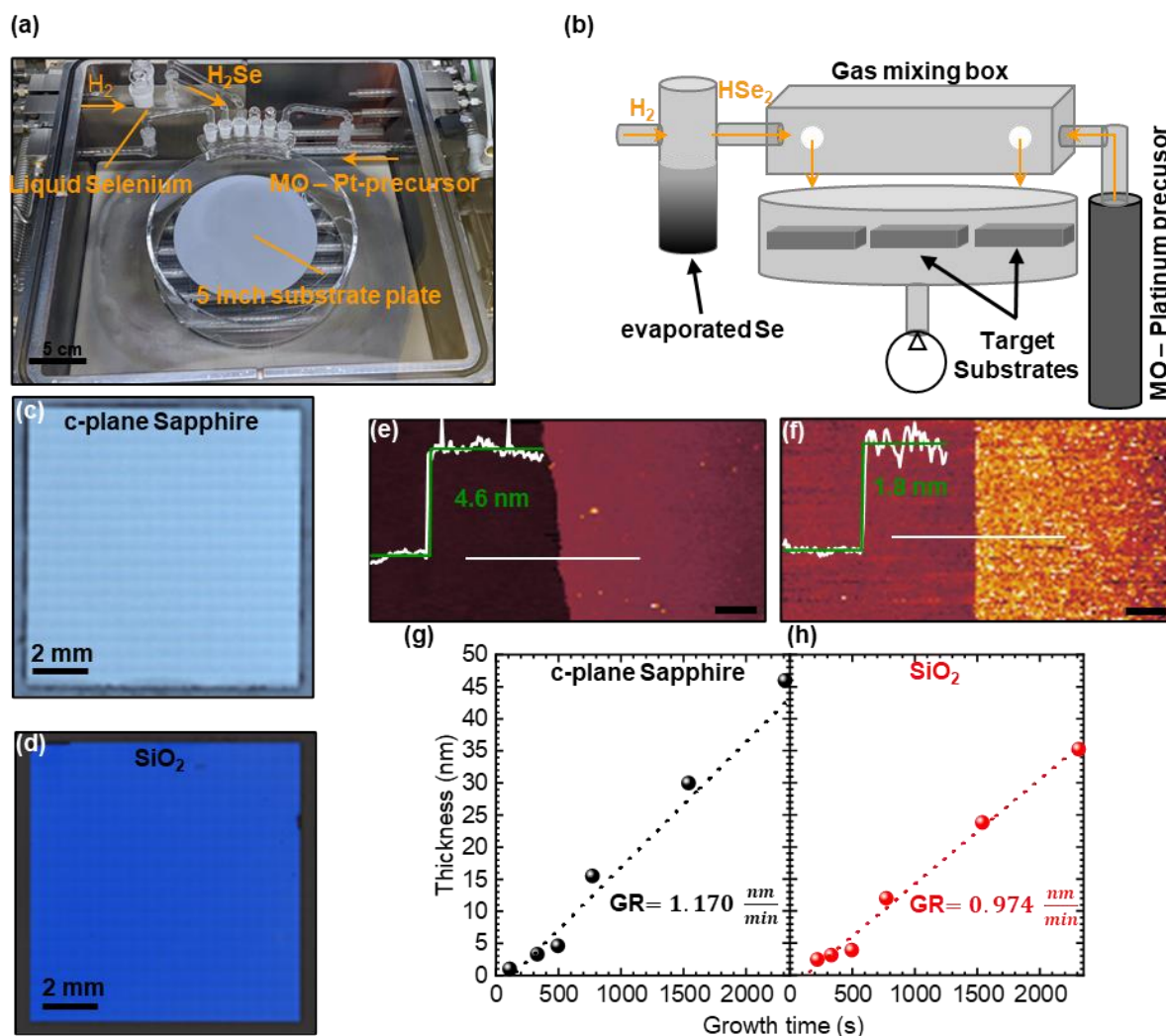


Figure 1: Synthesis of PtSe₂ by MOCVD. (a) Photograph of the 6" custom built MOCVD reactor. (b) Schematic of the reactor chamber (c) 1 nm and (d) 3.2 nm PtSe₂ layer homogeneously deposited on 1 * 1 cm² scale (c) c-plane sapphire and (d) SiO₂ substrates. (e) AFM image including height profile (white) and fit (green) for a 4.6 nm thick PtSe₂ grown on c-plane sapphire and (f) 1.8 nm PtSe₂ grown on SiO₂, profile positions are marked as white lines. The scale bars (black) are 1 μm. (g, h) PtSe₂ layer thickness versus growth time on c-plane sapphire and SiO₂ respectively. Growth rates have been calculated from linear fitting.

Large-scale PtSe₂ films have been synthesized by MOCVD in a custom-built cold-wall reactor shown in Figure 1 (a). A schematic of the growth chamber, showing gas flows during deposition, is shown in Figure 1 (b). The substrates are placed on a 6" substrate plate. H₂Se is produced in-situ from evaporated selenium powder heated to 225 °C in a compartment off the primary reactor and flushed with H₂. (Trimethyl)ethylcyclopentadienylplatinum(IV) ((CH₃)₃(C₂H₂C₅H₄)Pt) is the platinum precursor provided in a Swagelok® cylinder, which is externally heated to 40 °C and supplied through heated pipes. Prior to deposition the substrates were annealed in a H₂ atmosphere for 15 min at 600 °C. The growth of the PtSe₂ is typically carried out at 600 °C. Homogeneous deposition was achieved on 1 * 1 cm² c-plane sapphire (Figure 1 (c)) and SiO₂ (Figure 1 (d)) substrates. Additional SEM images of the deposited layers can be found in the Supplementary information.

The growth method allows a high degree of control over the thickness by adjusting the growth duration. The growth rates (GRs) on different substrates were determined by AFM measurements (Figure 1 (e, f)) after deposition and result in $GR(\text{sapphire}) = 1.170 \frac{\text{nm}}{\text{min}}$ for c-plane sapphire (Figure 1 (g)) substrates and $GR(\text{SiO}_2) = 0.974 \frac{\text{nm}}{\text{min}}$ for SiO_2 substrates (Figure 1 (h)). Notably, the growth rate is significantly lower on SiO_2 compared to c-plane sapphire even though the intercepts with the x-axis for the linear fits, which define when successful seeding on the target substrate is achieved, are at 129 s and 120 s for c-plane sapphire and SiO_2 , respectively. However, no deposition could be observed on SiO_2 for a deposition time of 110 s by Raman and AFM measurements, a growth time which had already led to a 1 nm thick film on c-plane sapphire. From this, one can conclude that a closed layer is formed significantly faster on the c-plane sapphire (once seeding is achieved) than on the SiO_2 substrates.

2.2 Layer quality analysis by Raman spectroscopy, XPS and XRD measurements

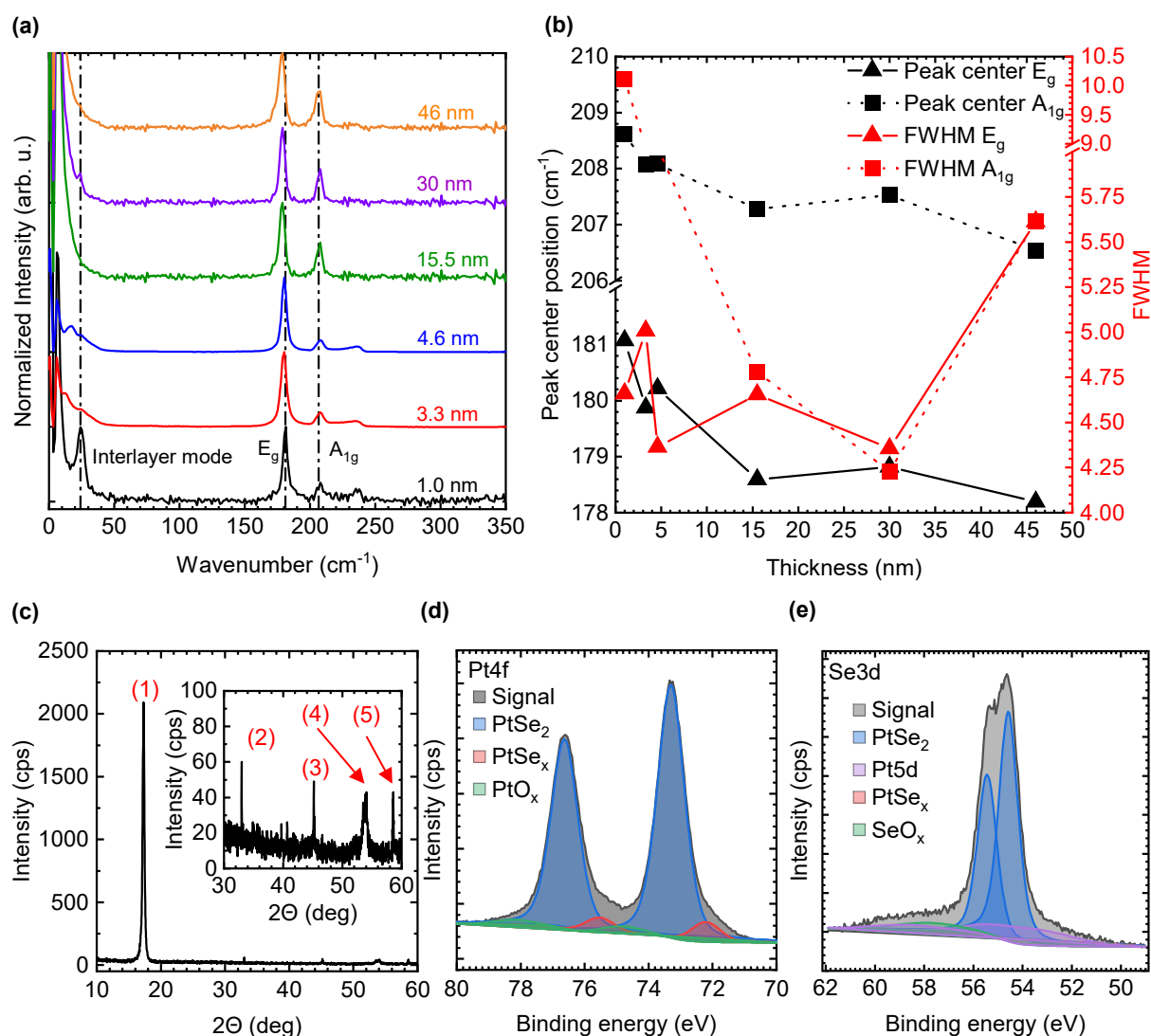


Figure 2: Raman spectra, XRD and XPS analysis of MOCVD grown PtSe_2 . (a) Normalized, averaged Raman spectra of differently thick PtSe_2 films grown on c-plane sapphire. (b) Peak center of the E_g (black triangle) and A_{1g} (black square) mode and respective FWHM (red) for different thickness PtSe_2 films. (c) XRD spectrum of a 35 nm thick PtSe_2 film. Inset showing a magnification of the same plot. XPS of the Pt4f core level region (d) and Se3d core level region (e). The spectra have been fitted to highlight the individual contributions to the raw signal (black).

To investigate the material quality, Raman spectra were acquired for PtSe₂ films of different thicknesses grown on c-plane sapphire substrates. The spectra were averaged over a total of 100 measured, distributed over the whole sample to increase statistical significance (Figure 2 (a)). A detailed Raman analysis for PtSe₂ grown on SiO₂ substrates can be found in the supplementary information. For all spectra, the fundamental PtSe₂ fingerprint E_g and A_{1g} modes can be identified, while also a prominent interlayer mode located at 24 cm⁻¹ is present in the spectrum of the 1 nm PtSe₂ (black). The absence of the interlayer mode for thicker layers is expected as it becomes less prominent for increasing film thickness²⁷. The evolution of the peak center position and FWHM for the E_g and A_{1g} mode are shown in (Figure 2 (b)). For the E_g (A_{1g}) mode a red shift from 181.07 cm⁻¹ (208.62 cm⁻¹) to 178.2 cm⁻¹ (206.54 cm⁻¹) is observed which is in good agreement with literature²⁷⁻²⁹. The FWHM of the E_g mode, which is directly correlated with layer quality^{29,30}, ranges from 4.3 cm⁻¹ to 5.6 cm⁻¹, giving an indication of high layer quality²⁹. The XRD spectrum (Figure 2 (c)) shows a strong feature at 17° which can be attributed to the (001) peak of PtSe₂^{30,31}, the higher order features (inset) at 33° and 45° are attributed to the (101) and (102) peaks, respectively, whose clear presence indicate the high crystallinity of the PtSe₂ film. For chemical binding analysis of the synthesized PtSe₂, spectra of the core level binding energy of the Pt4f (Figure 2 (d)) and Se3d (Figure 2 (e)) regions are acquired by XPS measurements. The core level region of the C1s peak (not shown here) was also recorded as reference to account for any residual surface charging not compensated by the tool built-in neutralization. The raw signal acquired from the Pt4f peak (grey) region consists of a total of three peaks. The main contribution (blue) has its maximum at 73.3 eV, indicating that 90 % of the Pt-atoms are present in the form of PtSe₂. About 6.8 % (red) of the contributing Pt-atoms are present in a sub-stoichiometric form PtSe_x. The remaining 3.2 % (green) are found in a slightly higher binding state which is attributed to surface oxidation. The latter two contributions are mainly present on the surface of the films, where a loss of selenium atoms (PtSe_x), and consequently oxidation (PtO_x) is expected³². The raw signal of the Se3d core level region (Figure 2 (e), grey) consists of three doublets and a singlet. The main contribution with 88.2 % (blue) originates from PtSe₂, while 1.7 % and 10.1 % are attributed to PtSe_x and SeO_x, respectively. The last contribution (purple) stems from the Pt5p orbital which was not taken into calculation for the PtSe₂ ratio. Furthermore, the survey spectrum (Supplementary Information) did not show any significant presence of other chemical components on our sample. In summary, detailed Raman analysis and XRD measurements confirm the high-quality growth of PtSe₂ by our newly developed MOCVD approach. In addition, XPS measurements reveal a stoichiometric ratio of 1:1.83, and that the signals of the Pt4f and Se3d peaks nearly exclusively originate from PtSe₂, supporting the conclusions drawn from Raman and XRD analysis.

2.3 Structural and electronic investigation by atomically resolved STM and STS measurements

STM (Figure 3 (a, b)) and STS (Figure 3 (c - i)) were used to investigate the quality of PtSe₂ films deposited on PyC. Individual PtSe₂ domains with sizes of 50 nm to 300 nm can be identified in large-scale STM images, as shown in Figure 3 (a). The high quality of the PtSe₂ within such domains is recognizable at smaller scales (Figure 3 (b)). For further evaluation of the atomic structure within one domain, STS measurements (Figure 3 (c - i)) were

acquired from the atomically clean region marked with a black square in Figure 3 (b). In Figure 3 (c) I/V (red) and dI/dV (black) spectra can be seen. From the dI/dV curve (black), the semi-metallic character of the 35 nm thick PtSe₂ film is confirmed (Figure 3 (c)). Furthermore, atomically resolved images acquired from bias voltages ranging from -0.4 V (Figure 3 (d)) to 0.4 V (Figure 3 (i)) show a perfectly ordered PtSe₂ lattice without any surface defects, once again underlining the high quality of PtSe₂ produced by our newly developed MOCVD approach.

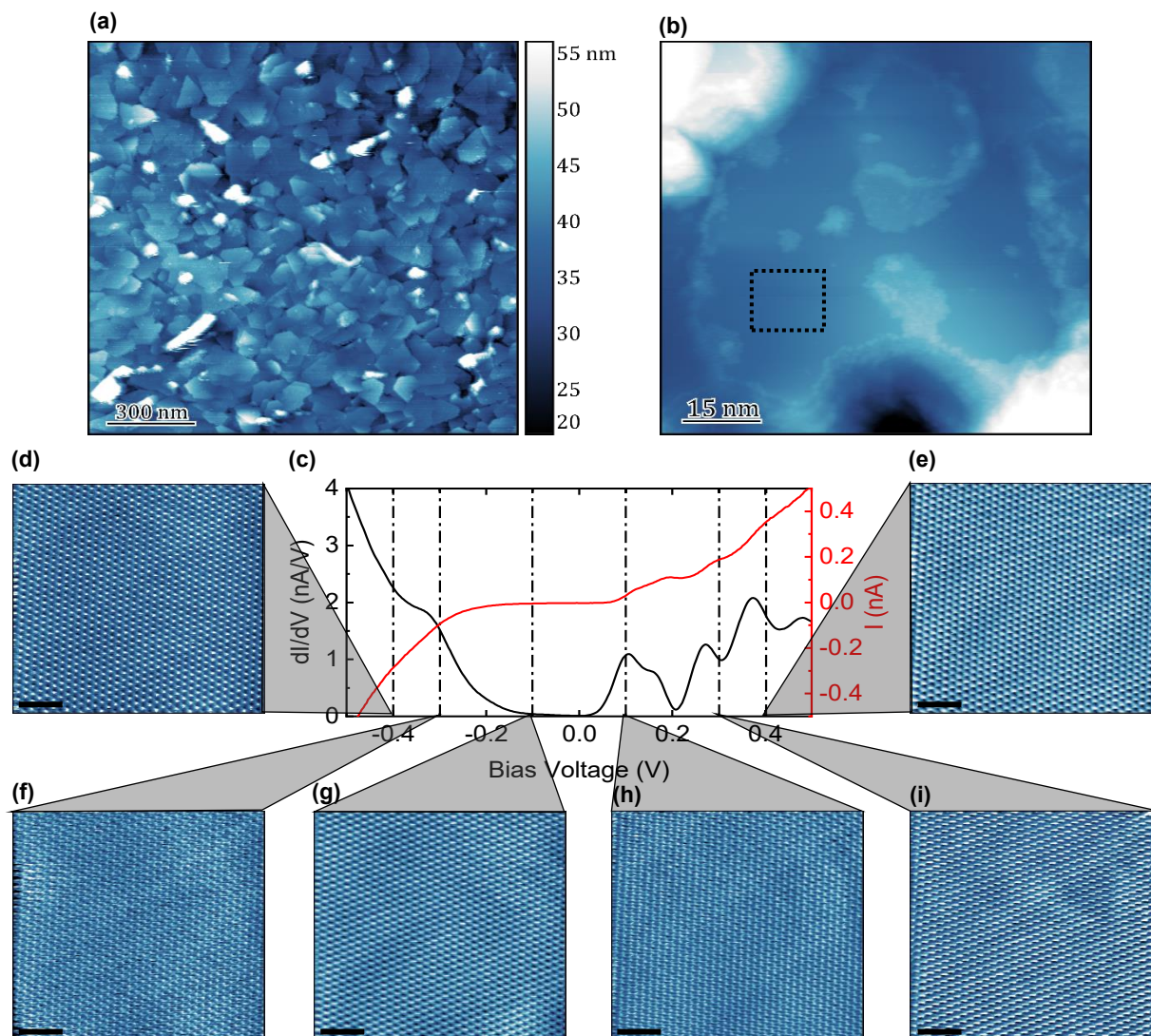


Figure 3: STM & STS of a 35 nm thick PtSe₂ film on PyC. (a, b) Large-scale STM image of PtSe₂ grown on PyC. Domain sizes in the range of 50 nm to 300 nm are visible. The black square in (b) indicated the area where high resolution STM and STS measurements have been conducted. (c) Derivation of current (black) and current (red) for different bias voltages used in STS. (d-i) Atomically resolved structure of PtSe₂ obtained for different bias voltages. The scale bars are 2 nm, the total size of all scans is 13 x 13 nm².

3. Conclusion

PtSe₂ was grown by combining a MO platinum precursor source and in-situ prepared H₂Se. Centimeter-scale synthesis was shown on SiO₂, c-plane sapphire and PyC substrates. The high quality of the synthesized material was verified by Raman, XPS and XRD measurements. Atomically resolved STM measurements revealed domains up to 300 nm wide and hexagonal PtSe₂ single crystals. Furthermore, atomically resolved STM images have confirmed locally defect free ordering within the single crystals, underlining the high material quality. In addition, the deposition was shown to be successful in a 6" wafer tool, paving the way towards industry scale synthesis of PtSe₂ and its future implementation into real world devices.

4. Experimental Section:

AFM analysis: A Jupiter XR AFM in tapping mode was used for acquisition of AFM data within this study. OMCL-AC160TS tips were used in tapping mode, the tapping frequency was set in between 200 to 400 kHz. Processing of the acquired raw data and extraction of topographical information was done using Gwyddion (64bit) 2.60.

Characterization by Raman and X-ray photoelectron spectroscopy: All Raman data was acquired using a WITec Alpha 300 Confocal Raman system. For all measurements, a green laser (532 °C) with a power of 2 mW (on sample surface) was used, with a 100x objective (NA = 0.9). A grating with 1800 lines per mm was used. All Raman data shown are averaged data from 100 points gather from a 1 x 1 cm² PtSe₂ film. Data was analyzed and processed using software provided with the tool. For XPS data a Versa Probe XPS tool (Physical Electronics GmbH) using the monochromated Al K α line (1486.7 eV). Single spectra were acquired using 61.8 W beam power paired with a 100 μ m spot size. Spectra have been fitted with Gaussian-Lorentzian fit functions using Multi-Pak.

X-ray diffraction spectroscopy: High resolution XRD analysis was obtained using Bruker D8 Discover equipment with a monochromated Cu K-alpha source.

Scanning tunneling microscopy and spectroscopy: STM and STS measurements were conducted on a low temperature microscopy (Createc) at liquid nitrogen temperature (77 K). The base pressure in the main STM chamber was $5 \cdot 10^{-11}$ mbar. Data were obtained in constant current mode using a single-crystalline W(001) tip.

Supporting Information:

The data that supports the findings of this study are available from the corresponding author upon reasonable request.

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Author contributions

M.P., C.C. and G.S.D. conceived and defined the study. G.S.D and I.S. supervised the study. M.P. and M.B. fabricated and conducted Raman and AFM measurements. A.Z. conducted XRD measurements and analyzed the data. O.H. conducted XPS measurement. T.S.L. and O.H. analyzed the spectra by multiple peak fitting. K.Z. and S.B. acquired STM and STS images, C.C., K.Z. and I.S. analyzed the respective data. K.L. acquired and analyzed electrical data. All authors extensively discussed and reviewed the manuscript. M.P., C.C. and G.S.D. wrote the manuscript with inputs from all authors.

Competing interests:

The authors declare no competing interests.

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Supporting Information

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1. SEM images of PtSe₂ grown on SiO₂

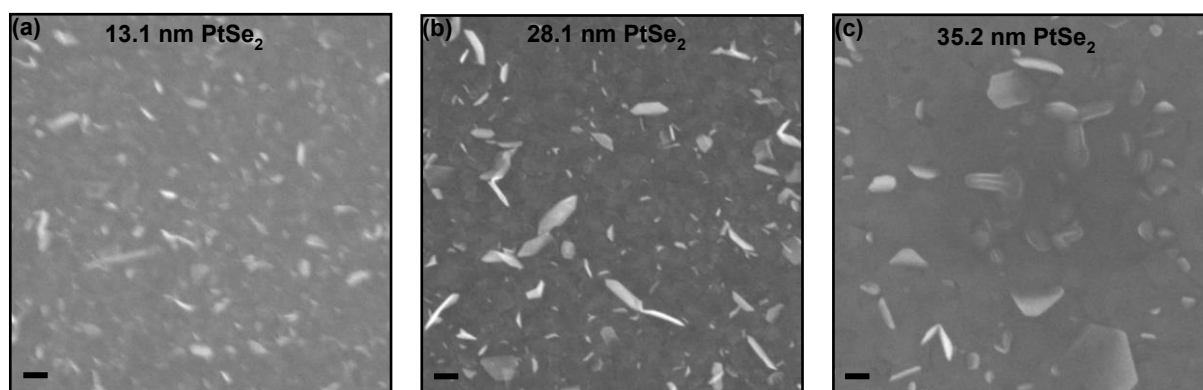


Figure S1: SEM imaging of PtSe₂ grown on different substrates. SEM images of MOCVD grown PtSe₂ on (a – c) SiO₂, (d – f) c-plane sapphire and (g – i) pyrolytic carbon. All scale bars are 100 nm.

Figure S1 shows SEM images for 13.1 nm thick (a), 28.1 nm thick (b) and 35.2 nm thick (c) PtSe₂ layers synthesized on SiO₂ substrates. The hexagonal shape of the deposited flakes are clearly visible.

2. Raman Analysis of MOCVD grown PtSe₂ on SiO₂

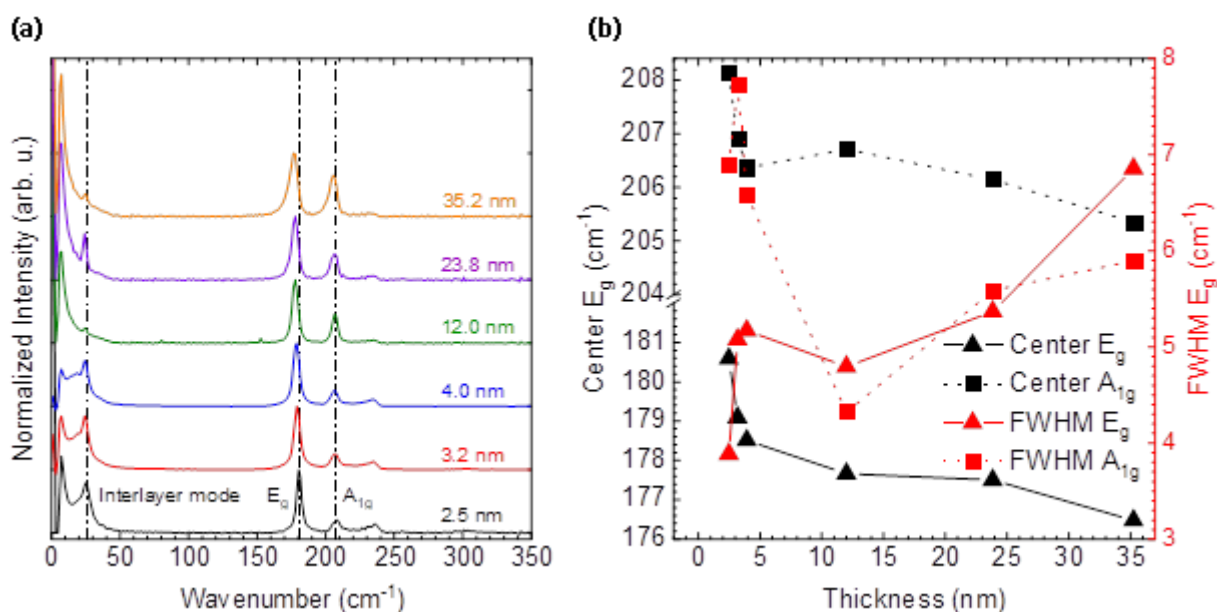


Figure S2: Raman analysis of MOCVD grown PtSe₂ on SiO₂ substrates. (a) Normalized, averaged Raman spectra of different thickness PtSe₂ films grown on c-plane sapphire. (b) Peak center of the E_g (black triangle) and A_{1g} (black square) mode and respective FWHM (red) for different thickness PtSe₂ films.

Figure S2 (a) shows six Raman spectra acquired from different thicknesses of PtSe₂ deposited on SiO₂ using the MOCVD growth approach presented in the main text. The spectra have been averaged (100 spots equally distributed on a 1 x 1 cm² sample) and normalized to the E_g mode for better comparison. For all thicknesses, the Raman fingerprint mode of PtSe₂ (E_g and A_{1g}) can be identified. With the peak position of the E_g (A_{1g}) mode located at 180.6 cm⁻¹ (208.12 cm⁻¹) for the 2.5 nm thick PtSe₂ (black), a blue shift of 3.9 cm⁻¹ (3.7 cm⁻¹) can be identified with increasing film thickness up to 35.2 nm (Figure S2 (b)). This is in good agreement with literature¹⁻³. Furthermore, a prominent interlayer mode evolving around 24 cm⁻¹ can be identified for the thinnest layer, becoming less prominent with increasing film thicknesses⁴. A FWHM of 3.9 cm⁻¹ is extracted from the E_g mode of the thinnest layer, increasing to 6.86 cm⁻¹ for the thickest PtSe₂ layer. This indicated high layer quality, as the FWHM of the E_g mode has been reported to be a quality indicator².

3. XPS survey spectra of MOCVD synthesized PtSe₂

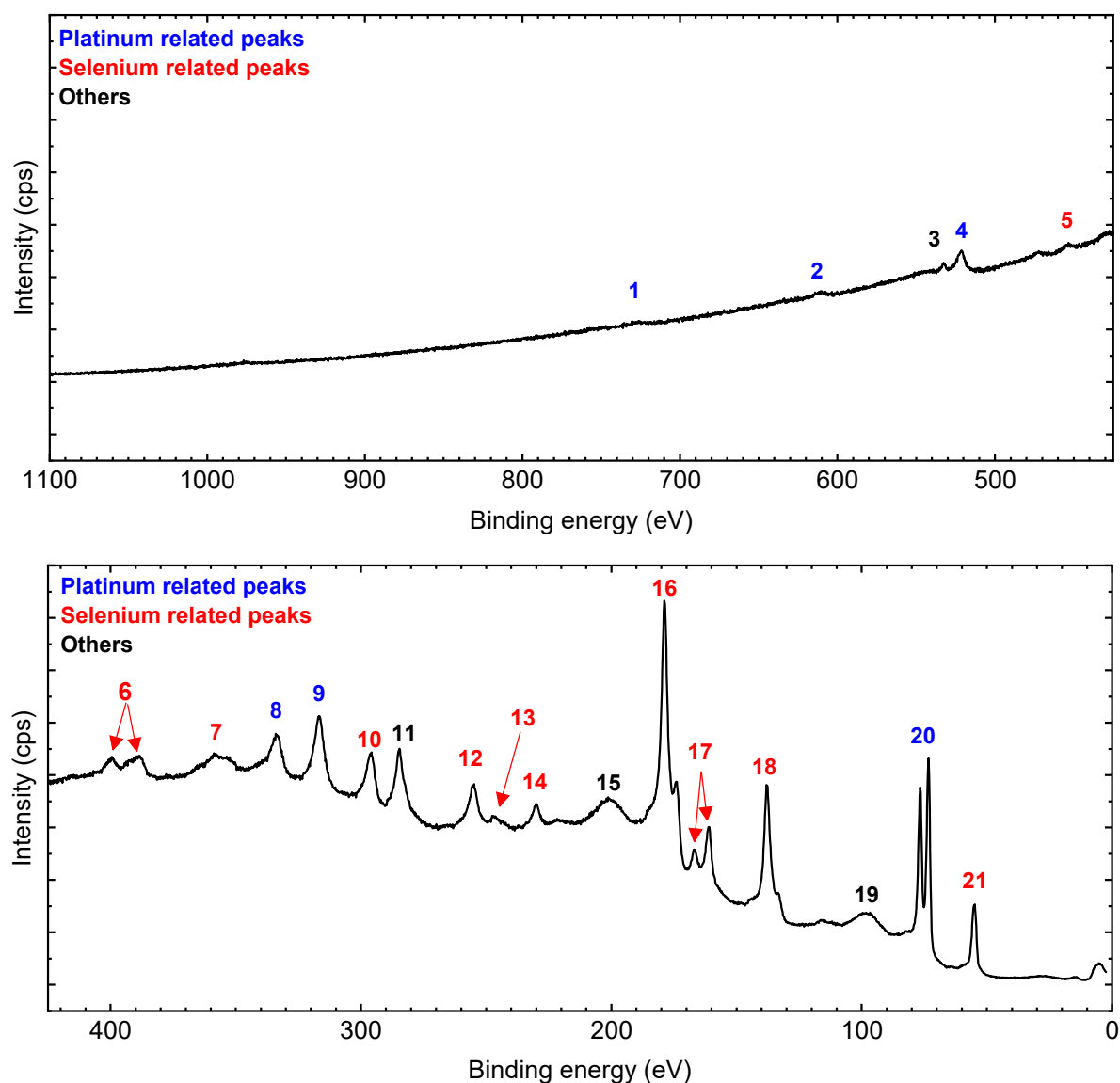


Figure S3: XPS survey spectrum. XPS survey spectrum of MOCVD 13 nm thick PtSe₂ deposited on SiO₂ for binding energies (a) 1100 eV to 425 eV and (b) 425 eV to 0 eV. The primary platinum related peaks have been marked in blue, selenium related peaks have been marked in red and the remaining peaks have been left as black.

For investigation of possible contaminants in our MOCVD grown PtSe₂ we acquired a high-resolution survey spectrum (Figure S3) of a 13 nm thick PtSe₂ sample deposited on SiO₂. The spectrum has been shifted using the C1s peak (284.6 eV⁵) as a reference. All peaks in the spectra have been labelled while platinum and selenium marked features are marked in blue and red, respectively⁵. Other features, originating from plasmon losses, carbon and oxygen have been marked in black. A conclusive summary of all features is given in Table S1. Notably, a strong nonlinear background was observed during acquisition. The background originates from plasmon losses which are caused by the sample surface morphology⁶. This is well supported by strong plasmon peak features (label 15 and 19). In addition to selenium and platinum related features, we can identify a small amount of oxygen on the sample (label 3) which we attribute to surface oxidation occurring on the PtSe₂ layer. Nevertheless, with the exception of

oxygen, the overall signal originates from selenium and platinum related features, showing a high purity of the layers without any surface contaminations.

Table S1: Summary table of all peaks which are visible in Figure S3.

Label in figure	Peak origin	Peak position(measured) [eV]	Literature value ⁵ [eV]
1	Pt4s	725	725
2	Pt4p _{1/2}	609	609
3	O1s	532	531
4	Pt4p _{3/2}	521	520
5	Se	453	-
6	Se	399 & 389	-
7	Se	359	-
8	Pt4d _{3/2}	334	332
9	Pt4d _{5/2}	316	315
10	Se Auger line L ₃ M ₂₃ M ₄₅ (¹ P)	297	299
11	Carbon 1s	284.6 (shift reference)	298.4
12	Se Auger line L ₂ M ₂₃ M ₄₅ (¹ P)	255	257
13	Se	248	-
14	Se3s	231	232
15	Energy loss peak of 16		
16	Se Auger line L ₃ M ₄₅ M ₄₅ (¹ P)	179	181
17	Se3p _{1/2} & Se3p _{3/2}	167 & 161	169 & 163
18	Se Auger line L ₂ M ₄₅ M ₄₅	138	140
19	Energy-loss peak of 20	99	-
20	Pt4f _{5/2} & Pt4f _{7/2}	77 & 73	74 & 71
21	Se3d _{3/2} & Se3d _{5/2} & Pt5p	55	74 & 71
21	Se3d _{3/2} & Se3d _{5/2} & Pt5p	55	57 & 56 & 52

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