

Supplementary Information

A universal approach for the synthesis of two-dimensional binary compounds

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Supplementary Methods

Sample characterisation. Optical microscopy images were taken in a Nikon Eclipse L200N optical microscope. Scanning electron microscopy (SEM) was conducted in a Zeiss Supra 40VP operated in in-lens detection mode at 5.00 keV. Atomic force microscopy (AFM) scans were done in a Bruker AFM Dimension Icon. X-ray photoelectron spectroscopy (XPS) measurements were conducted in a Thermo Fisher Scientific K-alpha XPS system using an AlK α X-ray source (1486.7 eV). Acquisition parameters were kept constant for all survey and high-resolution scans (300 ms x 10 scans). Raman spectroscopy was conducted in a Thermo Fisher DXR microscope equipped with a 455 nm laser (5 mW, 10 s x 5 exposure time, 50x objective). Photoluminescence spectra were obtained using a custom spectroscopy setup built from a Nikon Eclipse Ti-U inverted microscope and equipped with a 407 nm diode laser and a Shamrock 303i spectrometer with a 450 nm longpass filter (30 μ W, 1 s x 5 exposure time, 100x objective, 0.9 NA).

Electron backscatter diffraction (EBSD). EBSD measurements were used to determine the surface grain orientation of Au-*M* alloys after processing, which in the case of Supplementary Figure 1 was an Au-V alloy. Measurements were done in a FEI Nova NANO SEM 600 using the following parameters: 15 kV, 12 nA, 10.5 mm working distance, 50 μ m aperture, a 10 μ m x 10 μ m scan area and a step size of 280 nm. The obtained diffraction patterns were indexed against pure Au and Au-V (85%-15%) structures, and were found to match with the pure Au phase only. Various different regions were also scanned to ensure homogeneity throughout the sample. The SEM image in Supplementary Figure 1 was taken in the same instrument, using a secondary electron detector.

Transmission electron microscopy (TEM). TEM characterisation of W, Mo chalcogenides and CoS₂ was done in an FEI Tecnai T20 G2 operated at 200 kV. Bright field HRTEM images of transferred VN films were taken in an FEI Titan 80-300 environmental TEM with post-specimen spherical aberration correction, operated at 80 kV.

Terahertz-time domain spectroscopy (THz-TDS). THz-TDS mapping of the sheet conductivity of VN on quartz was acquired using a Picometrix T-ray 4000 fibre-coupled spectrometer. The sample was raster-scanned to form a spatial map with 200 μ m step size and a resolution of \approx 350 μ m at 1 THz. The frequency-dependent sheet conductivity was determined as described in detail in previous reports^{1,2}.

Ellipsometry. Ellipsometry measurements were performed in reflection mode with a rotating compensator spectroscopic ellipsometer (M-2000, J.A. Woollam Co.). The measurement of the ellipsometric quantities ψ and Δ in the spectral range 0.7-5.9 eV was repeated at seven angles of incidence (45-75°) at each measurement spot using a collimated beam with a spot size of approximately 200 x 300 μ m. Four spots were measured on the transferred VN film on quartz. The multi-angle ψ and Δ spectra for each spot were fitted simultaneously to an optical model, with thickness and optical functions of

VN being the unknown parameters. The optical functions of the quartz substrate were determined in a separate ellipsometry measurement. A multi-sample analysis routine was employed to accurately determine both the thickness and the optical functions of VN avoiding the risk of cross-correlation between fitted parameters. In one of the four spots, the thickness was fixed to the value of 5.5 nm as determined by AFM. The thicknesses of the three remaining spots were fitted independently. The optical functions of VN were parameterised as explained in the caption of Supplementary Figure 21, but were not allowed to vary from spot to spot.

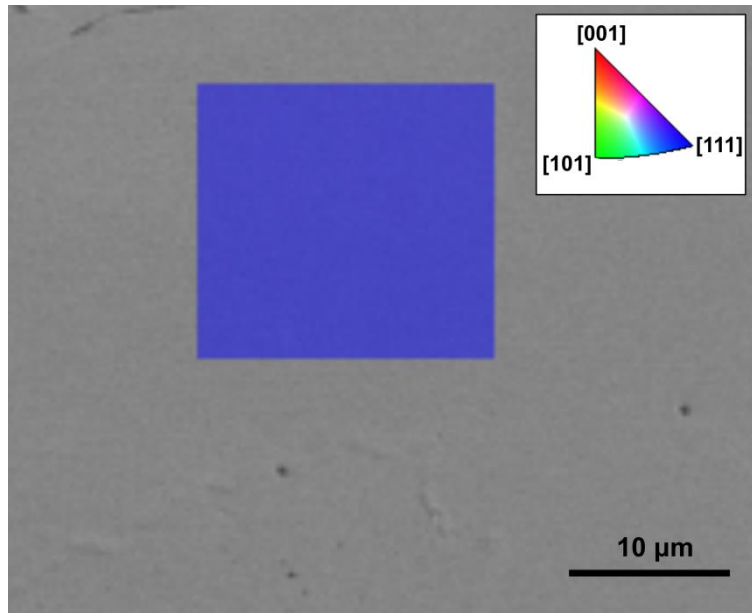
Density-functional theory (DFT) calculations. We use first-principles calculations based on density-functional theory to obtain the electronic properties of select chalcogenides. We employ a first-principles approach based on Kohn-Sham density functional theory (KS-DFT)³, as implemented in the Quantum ESPRESSO code⁴. The exchange correlation energy is described by the generalized gradient approximation (GGA) using the PBE⁵ functional. Interactions between valence and core electrons are described by Troullier-Martins pseudopotentials⁶.

Supplementary Table 1. Supplementary device performance characteristics of measured MoS₂ devices.

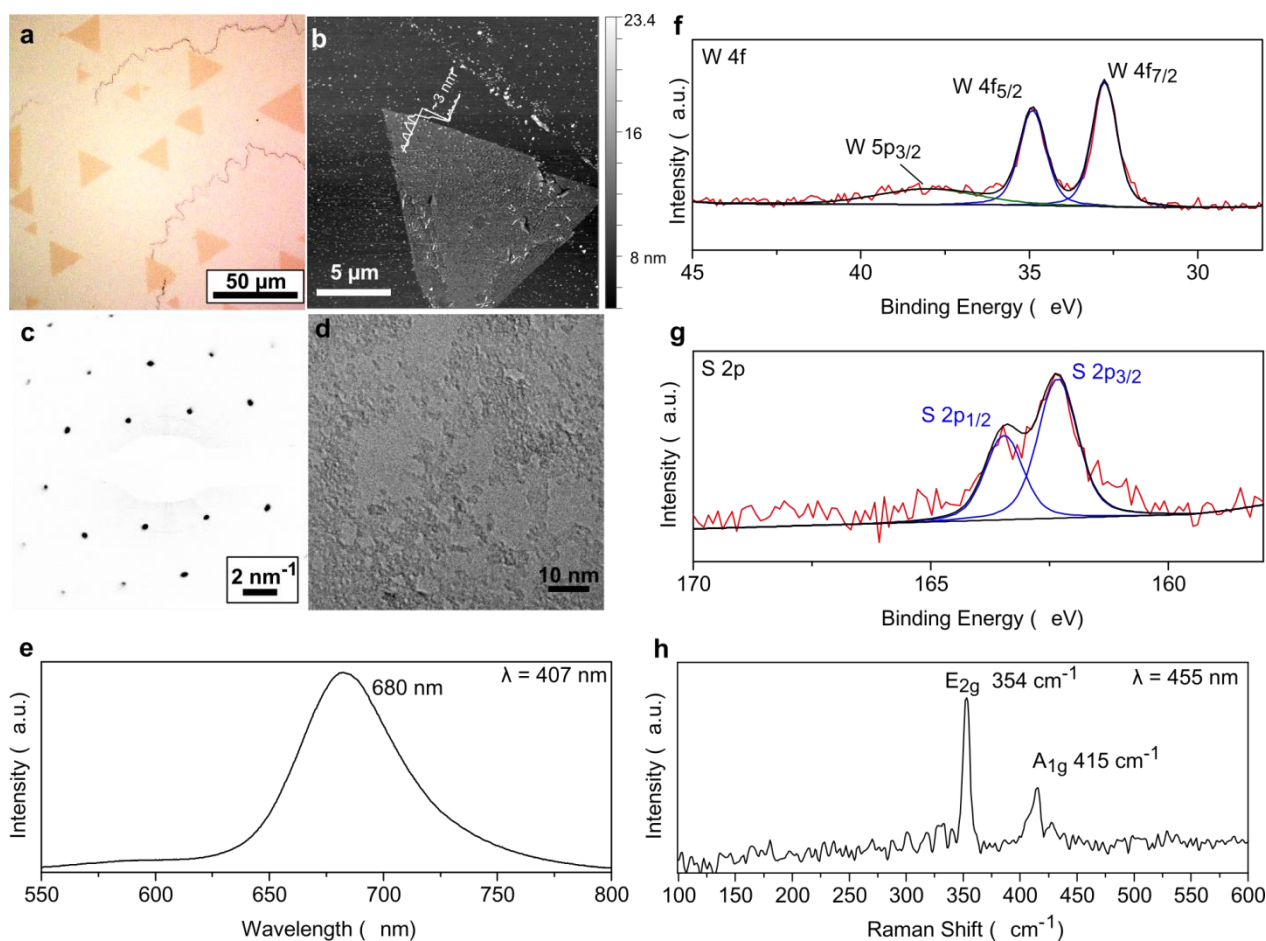
Device Number	μ (± 5 cm ² /Vs)*	On-off ratio**	Hysteresis (± 2 V)
1	4	10 ³	26
2	20	10 ⁴	9
3	25	10 ⁴	16
4	30	10 ⁴	11
5	30	10 ⁴	7
6	30	10 ²	7
7	5	10 ³	11
8	5	10 ³	12
9	15	10 ⁵	34
10	15	10 ⁴	14

*: Uncertainty in the estimation of μ for these devices is a result of the device geometry due to transfer of MoS₂ regions to pre-patterned electrodes (see main text).

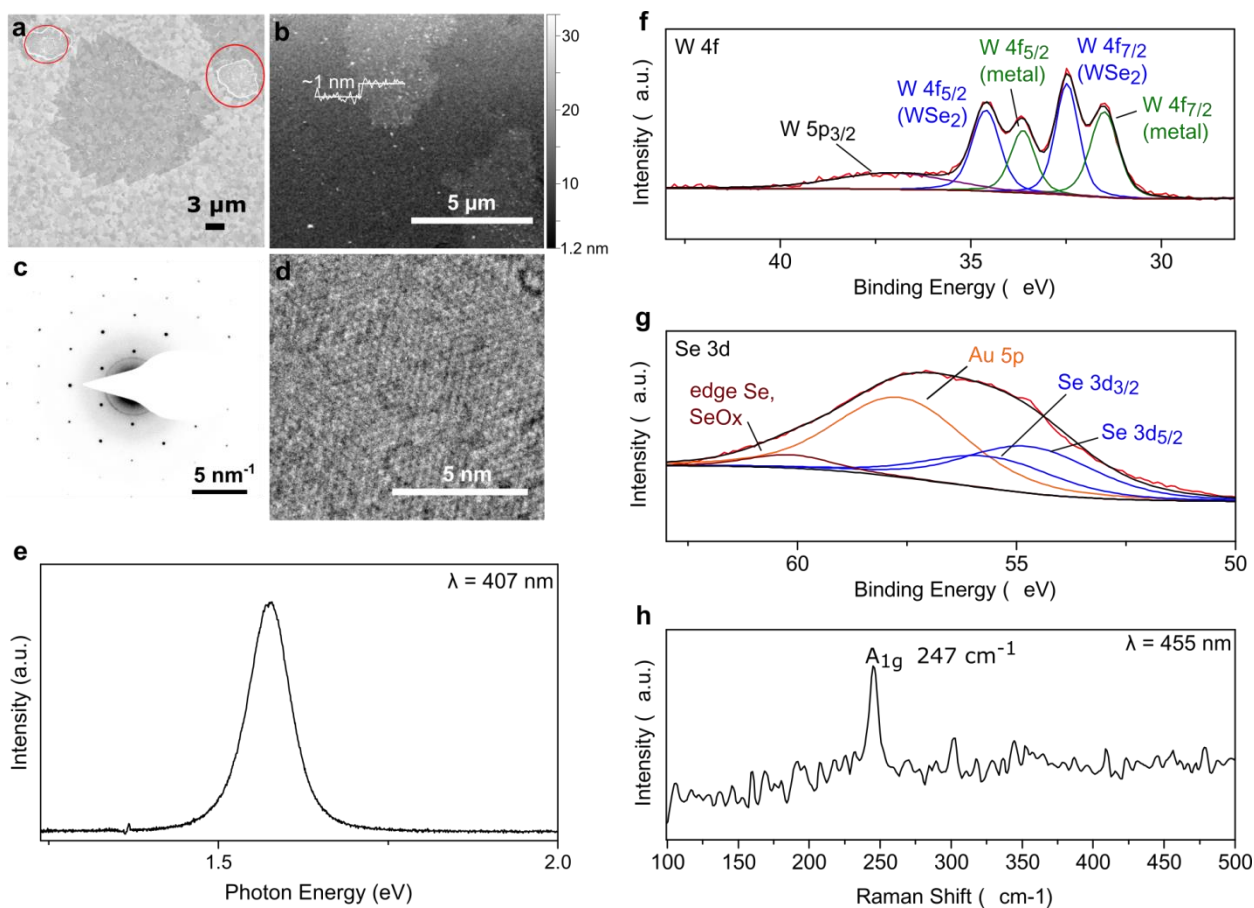
** : Gate leakage prevented driving the devices to saturation – the on-off ratio here is calculated from the largest achieved 'on' current in each case.



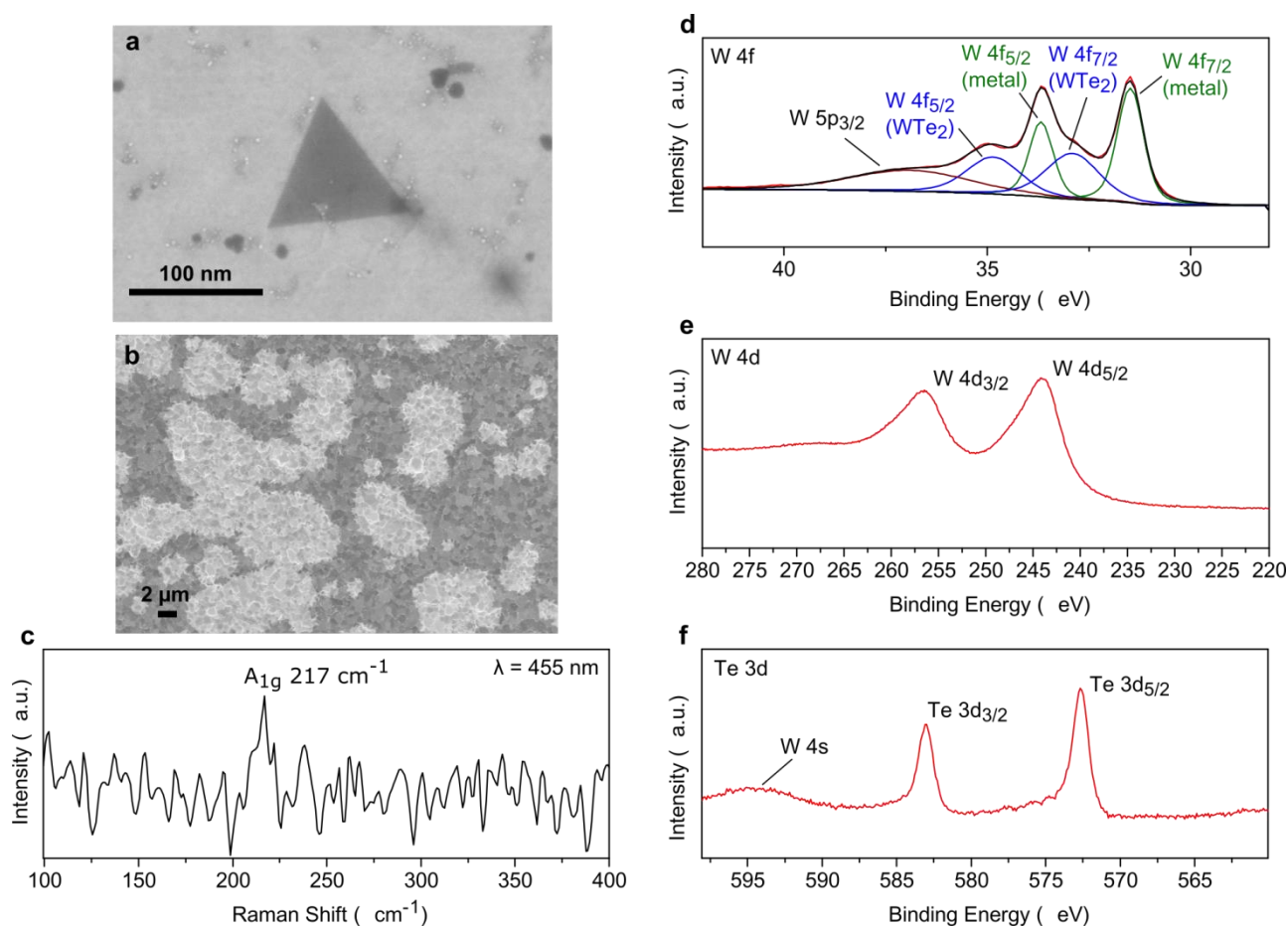
Supplementary Figure 1. EBSD of post-process Au alloys on c-plane sapphire. EBSD map of a $\sim 100 \mu\text{m}^2$ area overlaid onto an SEM image of the alloy surface. The EBSD clearly shows that the surface has the Au {111} orientation.



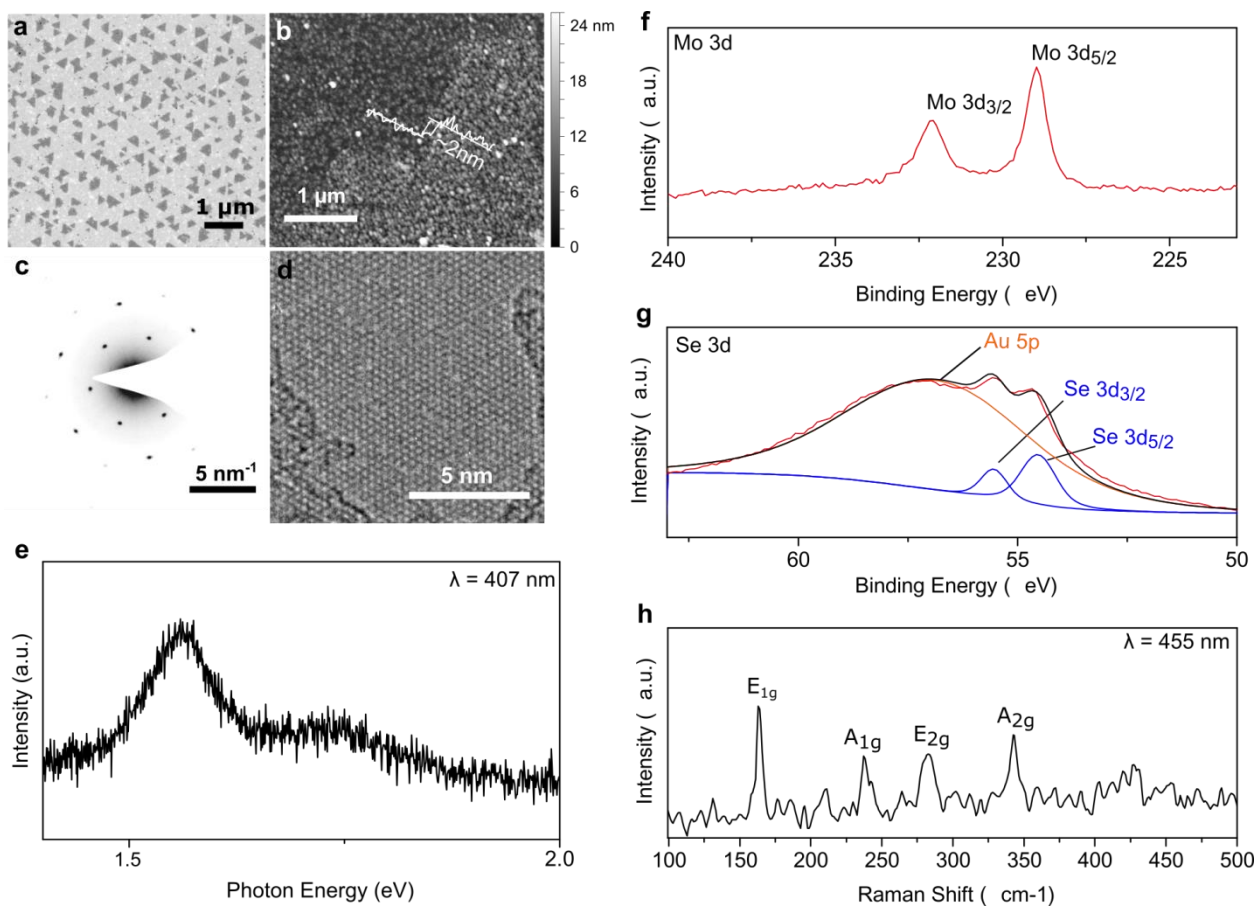
Supplementary Figure 2. Characterisation of tungsten disulphide (WS₂). (a) Optical image of WS₂ domains on gold, showing evidence of epitaxial alignment with the underlying substrate. (b) Atomic force microscopy (AFM) image of a WS₂ domain transferred onto 90 nm SiO₂ on Si substrate; the measured thickness of the domain is ~ 3 nm, which is within the thickness of a monolayer when taking into account polymer residues, surface oxidation, and tip and substrate interactions^{7,8}. (c) Selected area diffraction (SAED) pattern and (d) bright field TEM of a WS₂ domain transferred onto TEM grids. (e) Photoluminescence spectrum of WS₂ domains transferred onto 90 nm SiO₂/Si substrates, showing a strong peak at 680 nm. The peak is redshifted from reported values for monolayer WS₂⁹, which we suspect is due to screening and doping effects from transfer residues. (f-g) High resolution XPS spectra of (f) W 4f and (g) S 2p regions of as-grown WS₂ on the gold surface, where the peaks at 32.8 eV (W 4f_{7/2}), 34.9 eV (W 4f_{5/2}), 162.3 eV (S 2p_{3/2}) and 163.5 eV (S 2p_{1/2}) correspond to WS₂ (NIST XPS database). (h) Raman spectra of WS₂ domains on gold, indicating monolayer thickness¹⁰.



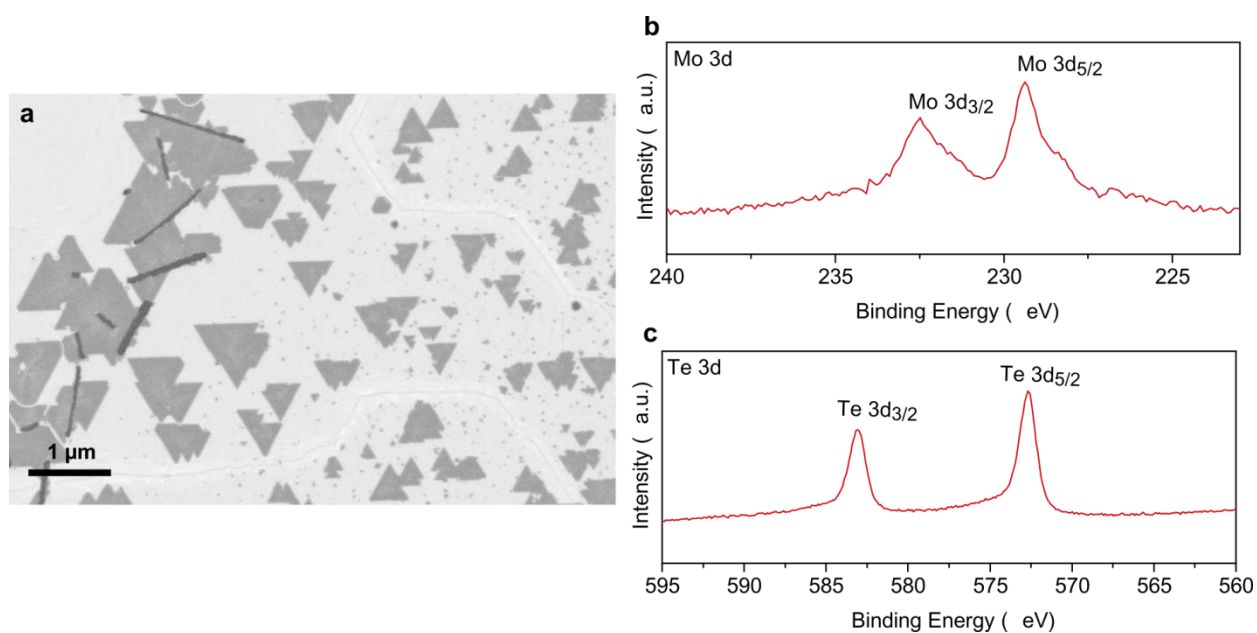
Supplementary Figure 3. Characterisation of tungsten diselenide (WSe₂). (a) SEM image of WSe₂ domains on gold. Red circles in (a) indicate areas where the gold film has dewetted the underlying tungsten layer due to its thickness (initial thickness of the gold film here was 300 nm). (b) AFM image of a WSe₂ domain transferred onto 90 nm SiO₂/Si substrate; the measured thickness of the domain is ~ 1 nm, not accounting for transfer residues and chemical contrast between tip and substrate. (c) SAED pattern and (d) bright field TEM of WSe₂ transferred onto TEM grids. (e) Photoluminescence spectrum of WSe₂ domains transferred onto 90 nm SiO₂/Si substrates, which shows a strong peak at 1.57 eV. (f-g) High resolution XPS spectra of (f) W 4f and (g) Se 3d regions of as-grown WSe₂ on the gold surface. Peaks at 32.5 eV and 34.6 eV in (f) correspond to WSe₂, while the peaks at 31.5 eV and 33.6 eV correspond to the exposed W metal in the de-wetted areas in (a) (NIST XPS Database). Peaks in (g) at 54.7 eV and 55.6 eV correspond to Se 3d peaks of WSe₂, while we attribute the peak at 60.2 eV to oxidised/edge selenides^{11,12}. (h) Raman spectra of WSe₂ domains on gold, which indicates that they are monolayers¹³.



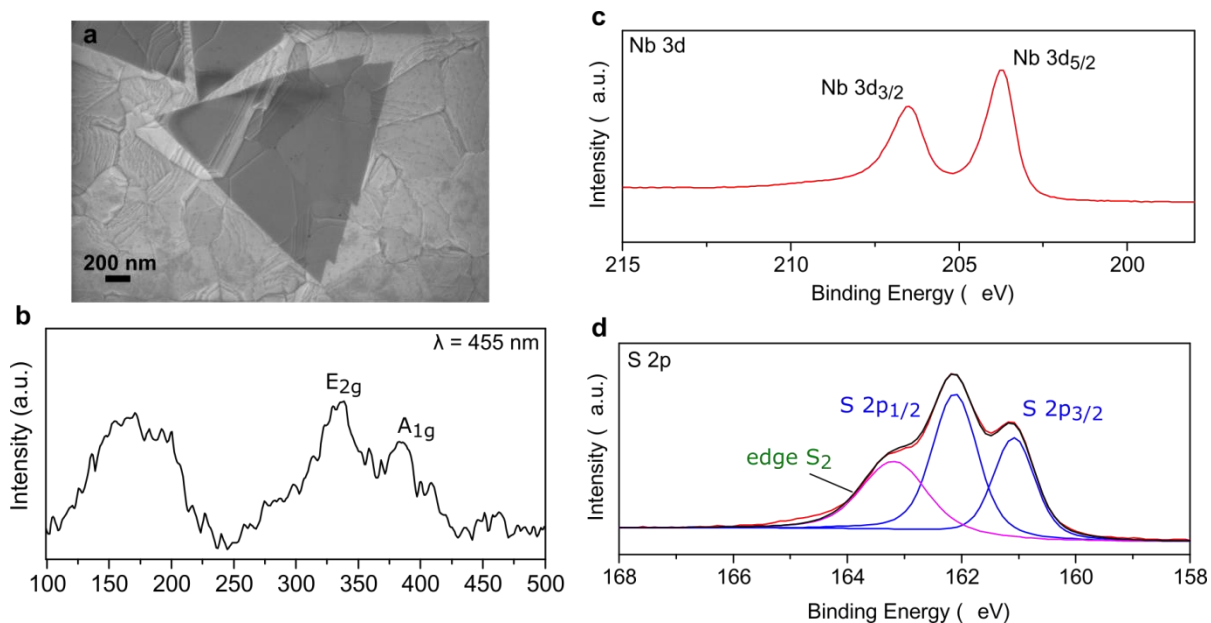
Supplementary Figure 4. Characterisation of tungsten ditelluride (WTe_2). (a) High magnification of isolated WTe_2 domains and (b) low magnification SEM images of merged WTe_2 domains on gold. Dark coloured regions in (b) are partially grown WTe_2 films. (c) Raman spectra of WTe_2 on gold, which indicates that the film is a monolayer¹⁴, (d-f) High resolution XPS spectra of (d) W 4f, (e) W 4d and (f) Te 3d regions of as-grown WTe_2 on gold. W 4f peaks in (d) at 32.9 eV and 34.85 eV correspond to WTe_2 , whereas the peaks at 31.5 eV and 33.7 eV correspond to W metal from dewetted areas, similar to the case of WSe_2 (the initial gold film thickness was 300 nm in this case as well). W 4d peaks in (e) at 244.2 eV and 256.5 eV correspond to reported values for WTe_2 ¹⁵. Te 3d peaks in (f) are at 572.7 eV and 583 eV, corresponding to WTe_2 ¹⁵. WTe_2 films did not appear to survive transfer onto 90 nm SiO_2 on Si substrates.



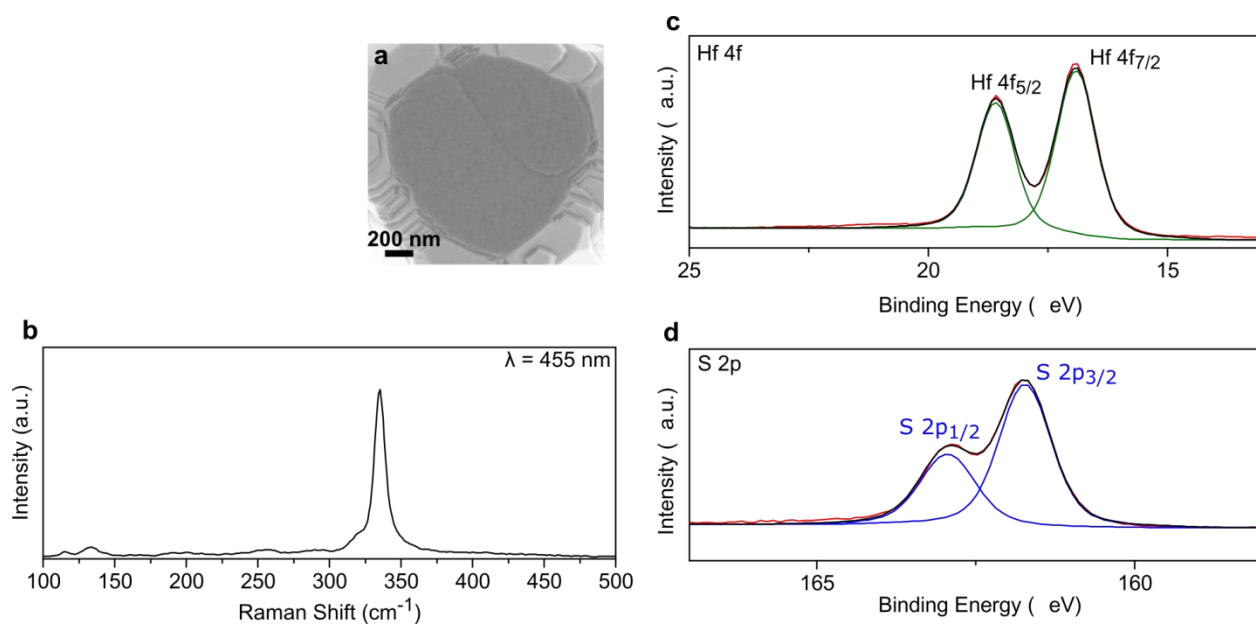
Supplementary Figure 5. Characterisation of molybdenum diselenide (MoSe₂). (a) SEM image of MoSe₂ domains on gold, showing evidence of epitaxial alignment with the underlying gold. (b) AFM on MoSe₂ domains transferred onto 90 nm SiO₂/Si substrate; the measured thickness of the domain is ~ 2 nm, not accounting for transfer residues and chemical contrast between tip and substrate. (c) SAED pattern and (d) high resolution TEM image of MoSe₂ domains transferred onto TEM grids. Visible defects are caused by knock-on damage during imaging at 200 kV. (e) Photoluminescence spectrum of MoSe₂ domains transferred onto 90 nm SiO₂/Si substrates, showing A and B exciton peaks at 1.57 eV and 1.75 eV, respectively¹⁶. (f-g) High resolution XPS spectra of the (f) Mo 3d and (g) S 2p regions of MoSe₂ on gold. Mo 3d peaks in (f) at 228.9 eV and 232.1 eV and Se 3d peaks in (g) at 54.6 eV and 55.5 eV correspond to MoSe₂¹⁷. (h) Raman spectra of MoSe₂ transferred onto 90 nm SiO₂/Si substrate; the pronounced E_{1g}, E_{2g}, and A_{2g} peaks are attributed to the excitation wavelength used in this work¹⁸.



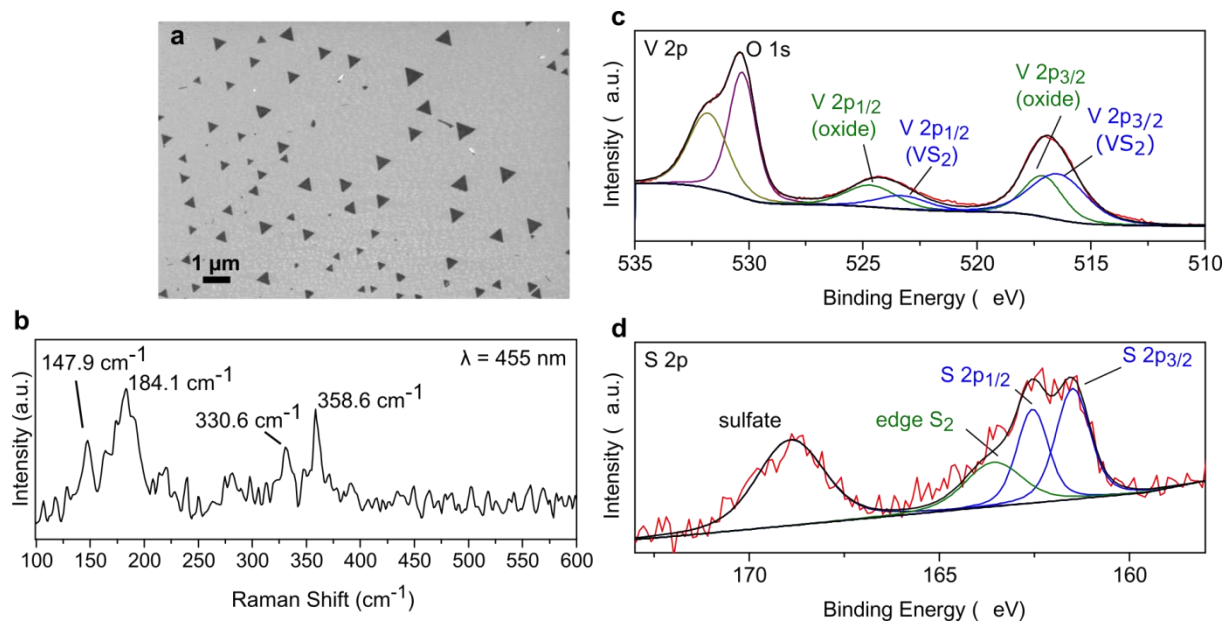
Supplementary Figure 6. Characterisation of molybdenum ditelluride (MoTe₂). (a) SEM image of a MoTe₂ domain on gold. (b-c) High resolution XPS spectra of the (b) Mo 3d and (c) Te 3d regions of MoTe₂ on gold. Mo 3d peaks in (b) at 229.2 eV and 232.3 eV and Te 3d peaks in (c) at 572.6 eV and 582.9 eV correspond to MoTe₂¹⁹. We were unable to detect a sufficiently strong Raman signal from the domains on gold with our experimental setup, and the domains did not appear to survive transfer onto SiO₂ substrates.



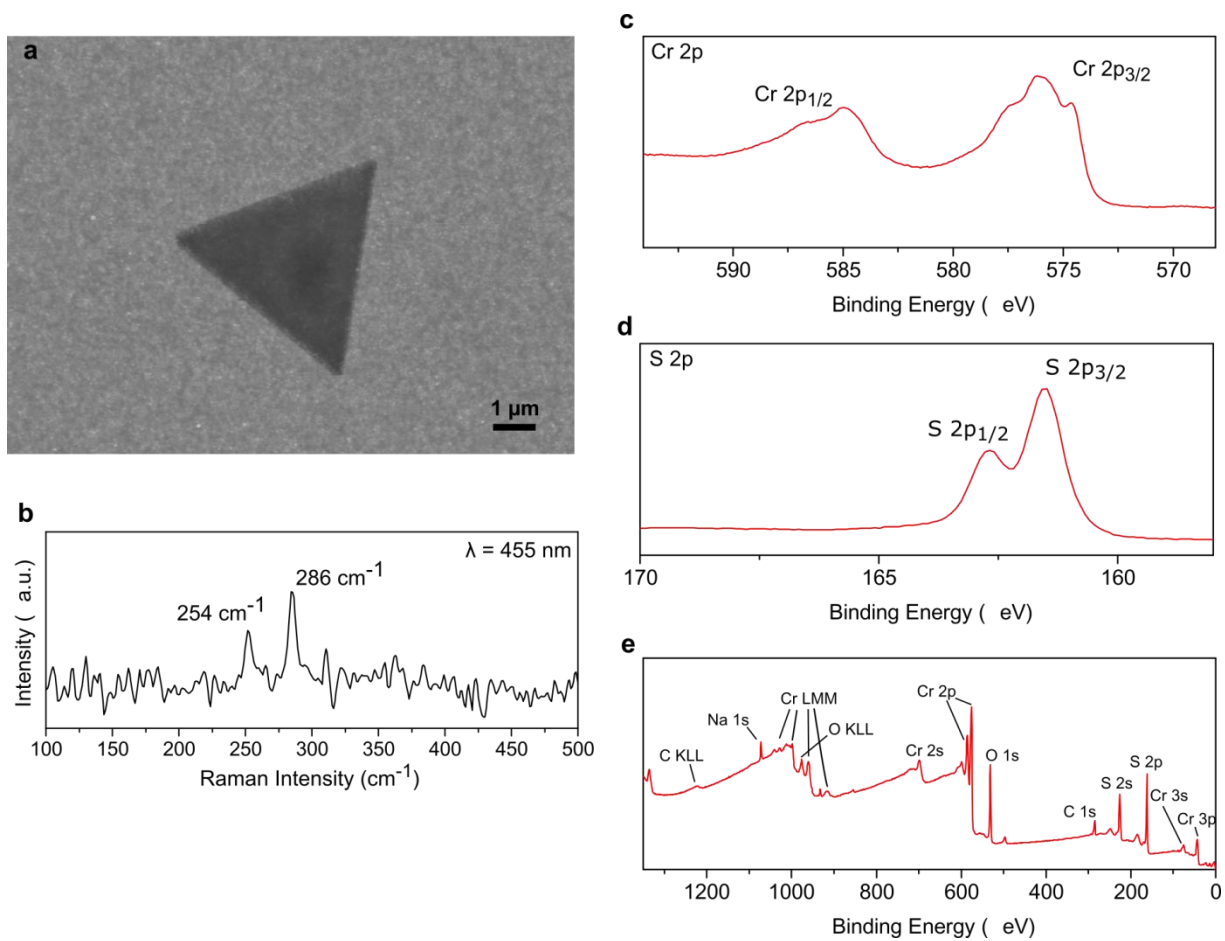
Supplementary Figure 7. Characterisation of niobium disulphide (NbS₂). (a) SEM image of an NbS₂ domain on gold. (b) Raman spectrum of an NbS₂ film on gold, where the E_{2g} and A_{1g} correspond to reported values for NbS₂²⁰. High resolution XPS spectra of the (c) Nb 3d and (d) S 2p regions of NbS₂ on gold. Nb 3d peaks in (c) are at 203.7 eV and 206.5 eV and S 2p peaks in (d) are at 161.1 eV, 162.2 eV and 163.2 eV.



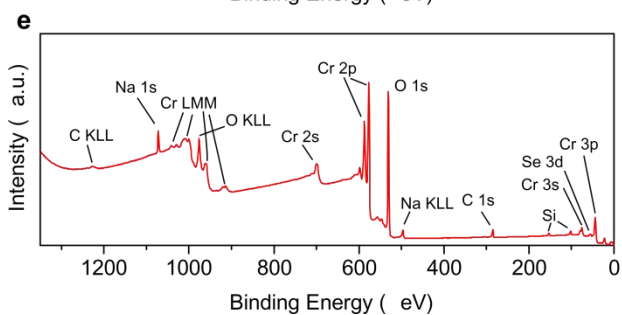
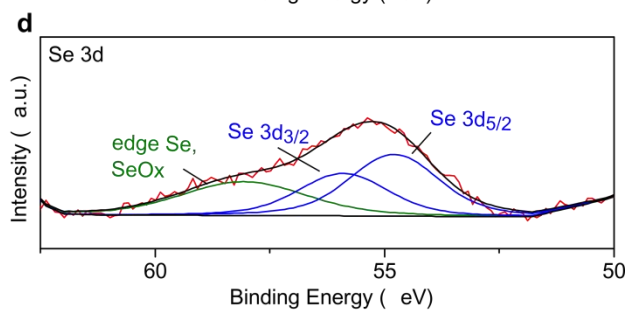
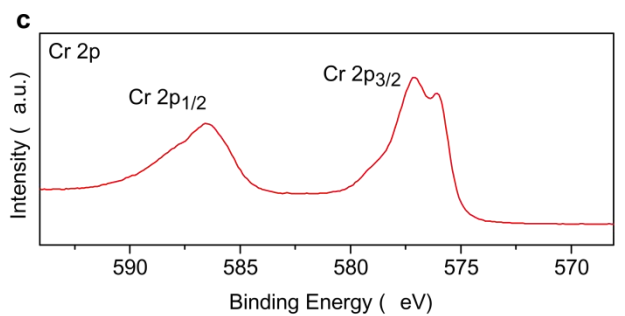
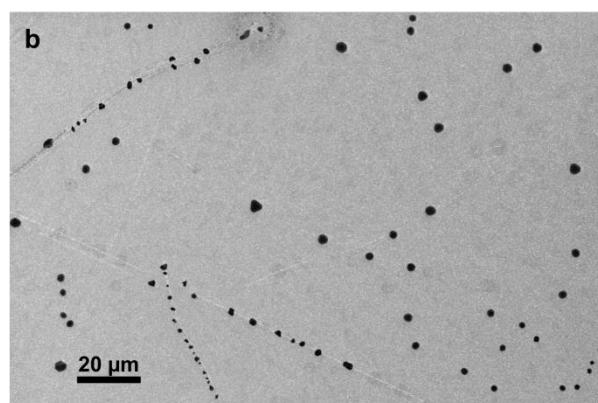
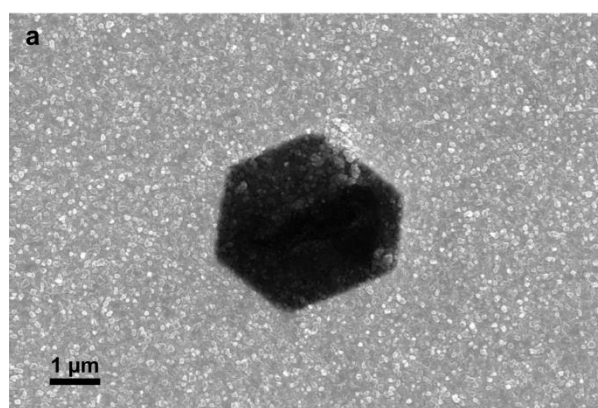
Supplementary Figure 8. Characterisation of hafnium disulphide (HfS₂). (a) High magnification SEM image of an isolated HfS₂ domain on gold. (b) Raman spectrum of an HfS₂ film on gold, which shows a pronounced peak at ~335 cm⁻¹ corresponding to reported values for HfS₂²¹. (c-d) High resolution XPS spectra of the (c) Hf 4f and (d) S 2p regions of HfS₂ on gold; Hf 4f peaks are at 16.9 eV and 18.6 eV and S 2p peaks are at 161.7 eV and 162.9 eV. The Hf:S ratio here is 1:2.



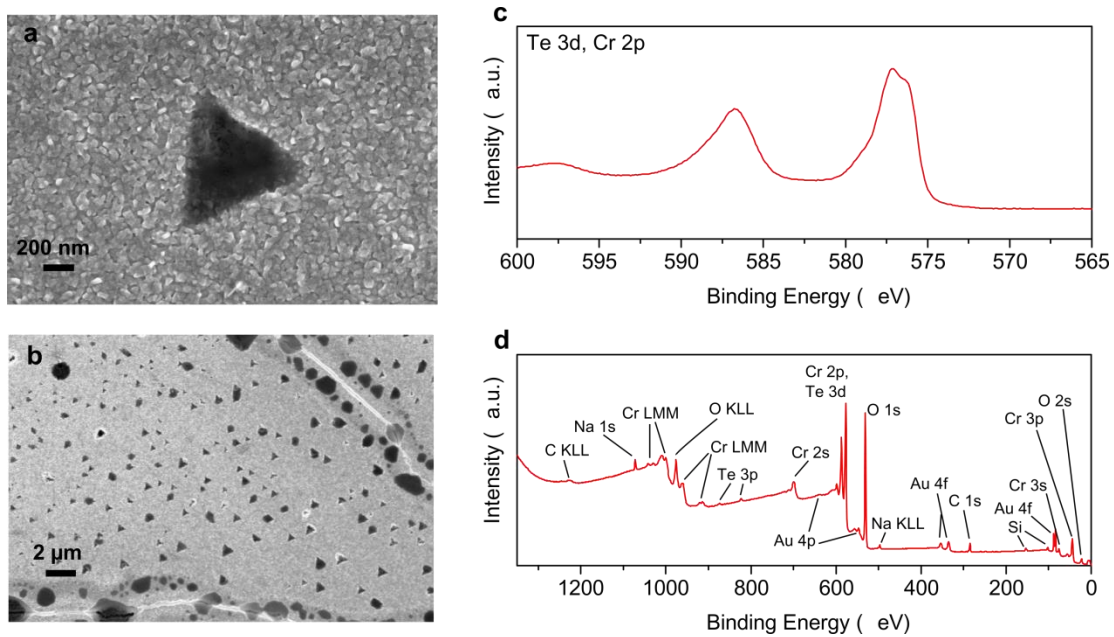
Supplementary Figure 9. Characterisation of vanadium disulphide (VS₂). (a) SEM images of VS₂ domains on gold, showing evidence of epitaxial alignment with the underlying Au (111) substrate. (b) Raman spectrum of VS₂ film on gold, where the peaks correspond to reported values for VS₂ nanosheets²². (c-d) XPS spectra of the gold surface post-growth: (c) V 2p and (d) S 2p scan. The V 2p spectrum shows peaks at 516.4 eV and 523.3 eV, which correspond to the V⁴⁺ oxidation state in VS₂²³. The peaks at 517.1 eV and 524.7 eV can be attributed to vanadium oxides¹¹. The S 2p peaks at 161.5 eV and 162.6 eV are attributed to VS₂. VS₂ is air-sensitive and degrades upon exposure to ambient, as evidenced by the prominent sulphate peak in (d).



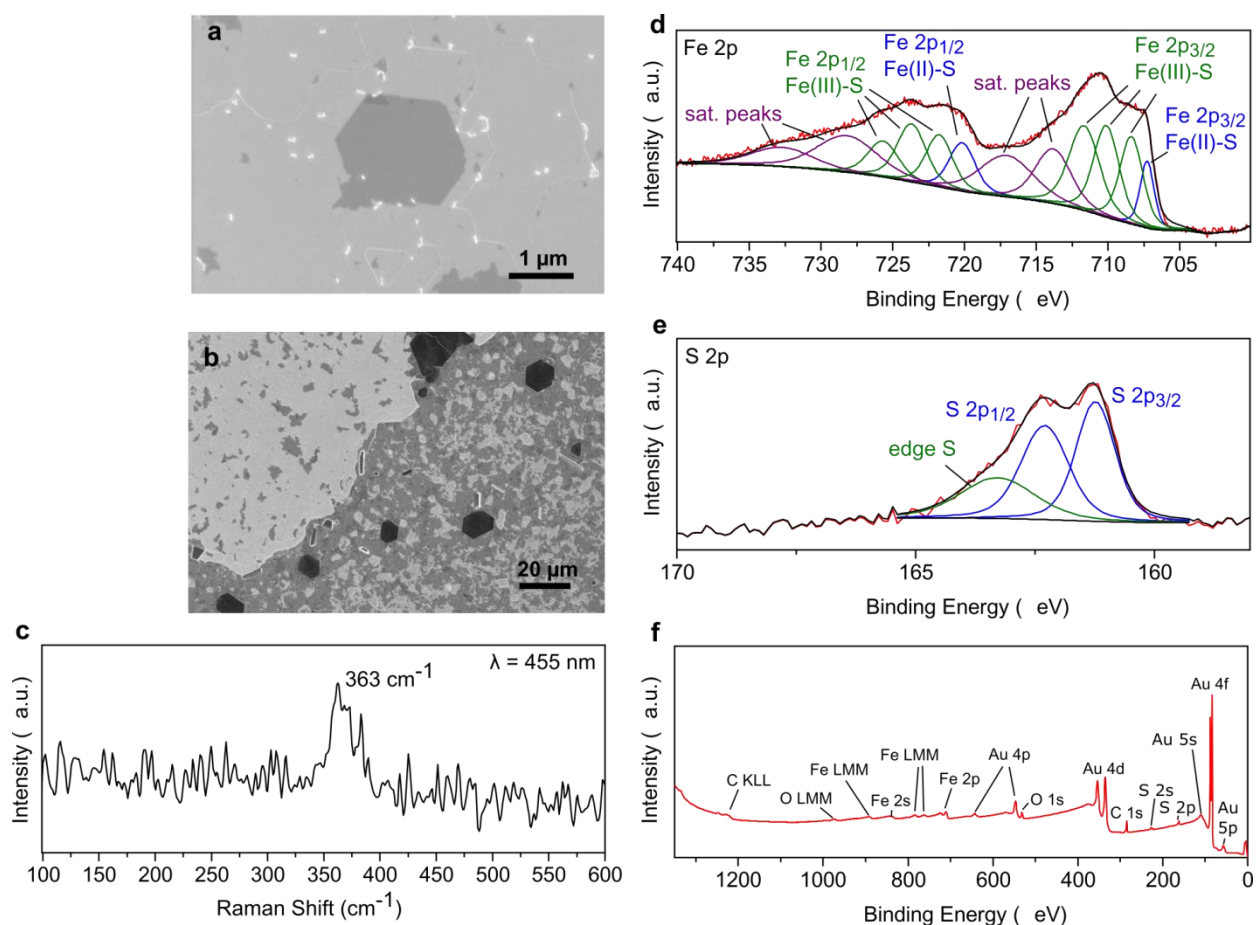
Supplementary Figure 10. Characterisation of chromium disulphide (CrS₂). (a) SEM image of a CrS₂ domain on gold. (b) Raman spectrum of a CrS₂ domain on gold. (c-e) XPS spectra of the gold surface post-growth: (c) Cr 2p, (d) S 2p, and (e) survey scan.



Supplementary Figure 11. Characterisation of chromium diselenide (CrSe_2). (a) High magnification and (b) low magnification SEM images of CrSe_2 on gold. (c-e) XPS spectra of the gold surface post-growth: (c) Cr 2p, (d) Se 3d, and (e) survey scan.

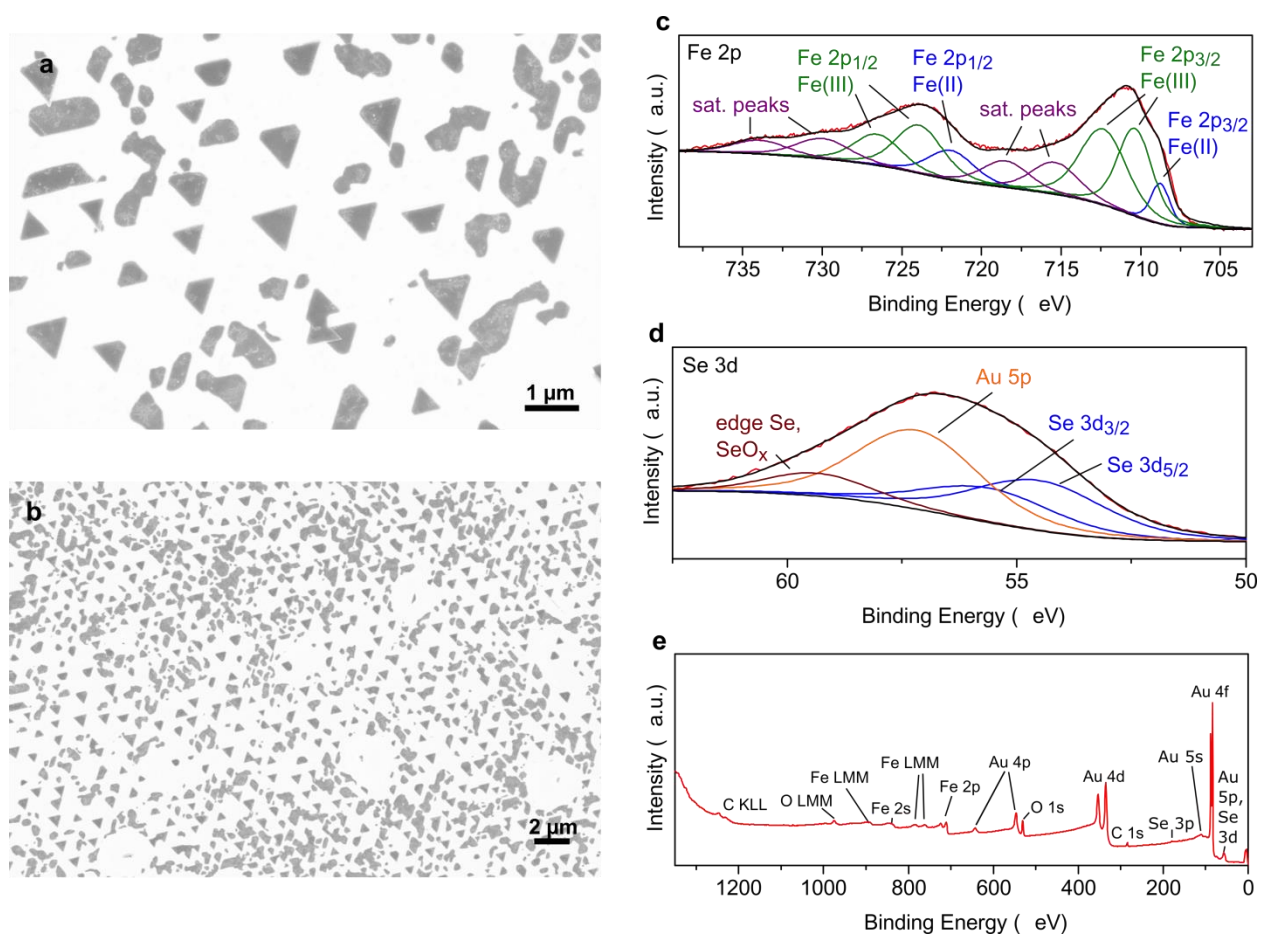


Supplementary Figure 12. Characterisation of chromium ditelluride (CrTe_2). (a) High magnification and (b) low magnification SEM images of CrTe_2 on gold. (c-d) XPS spectra of the gold surface post-growth: (c) Te 3d, Cr 2p and (d) survey scan.



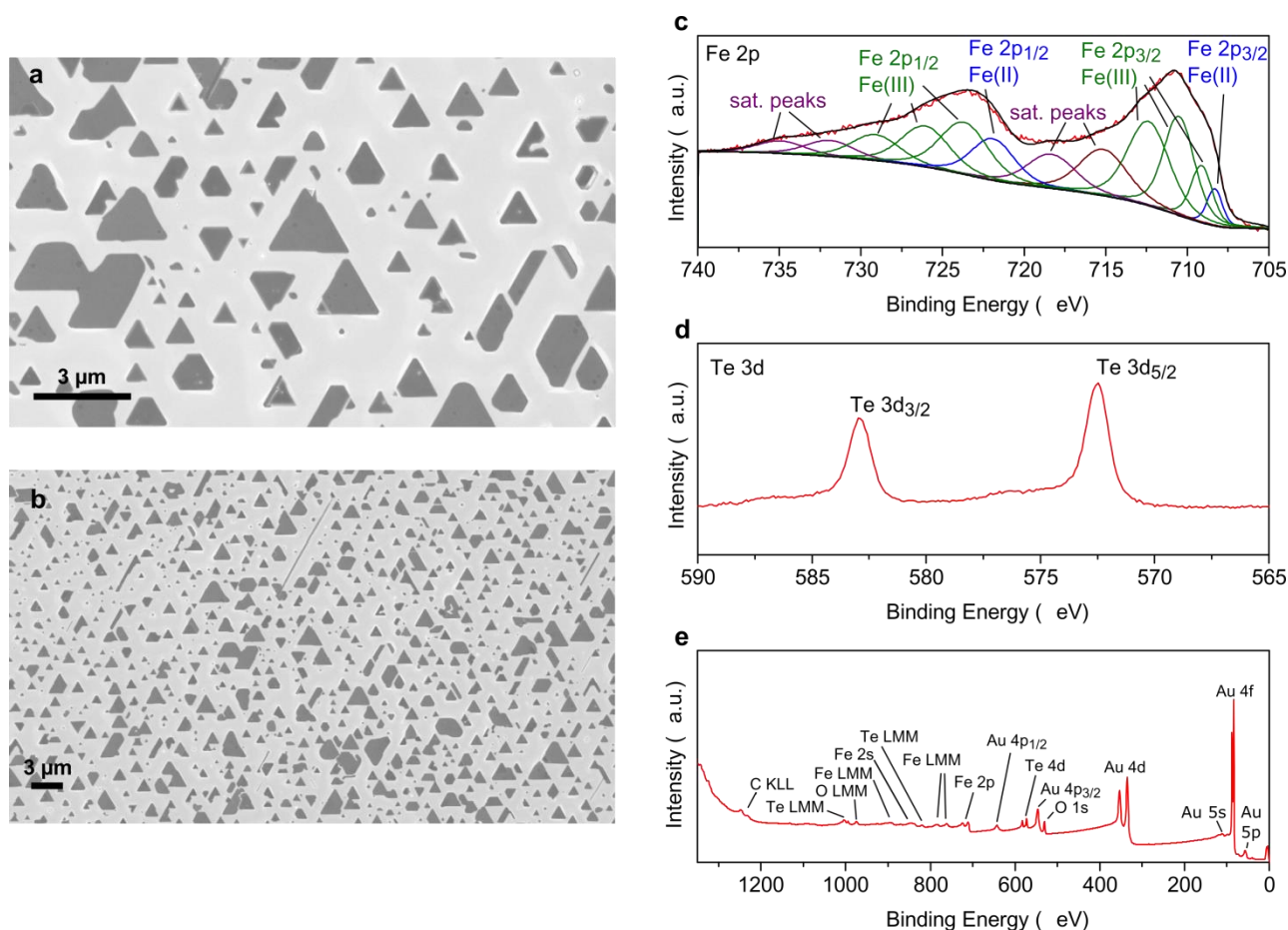
Supplementary Figure 13. Characterisation of iron sulphide (FeS). (a) High magnification and (b) low magnification SEM images of FeS on gold. (c) Raman spectrum of FeS domains on gold. (d-f) XPS spectra of the gold surface post-growth: (d) Fe 2p, (e) S 2p, and (f) survey scan.

Iron sulphides show various growth modes on gold as seen in (b), with some regions on the surface appearing to favour the growth of monolayers, while others favouring the growth of multilayers and 3D crystals. The Fe(II)-S peaks at 707.3 eV and 720.8 eV in the Fe 2p XPS spectrum (d) correspond to reported values for mackinawite^{24,25}, the layered phase of iron sulphide. We attribute the additional peaks at 708.4 eV, 710.1 eV, 711.7 eV, 722.4 eV, 724.1 eV, and 727.2 eV to spin multiplets of Fe³⁺ oxidation states at the edges and defects in the sulphide domains²⁵. The peaks at 713.9 eV, 730.5 eV and 716.9 eV, 733.9 eV are attributed to satellite peaks of the Fe²⁺ and Fe³⁺ oxidation states, respectively. The S 2p region shows 3 peaks at 161.6 eV, 162.9 eV and 164.2 eV, which are attributed to the S 2p_{3/2}, S 2p_{1/2} and edge states of the FeS domains^{24,26}, respectively. A small amount of oxygen can be seen in the survey spectrum; as such, it is plausible that oxide peaks may be convoluted with the satellite peaks in (d). The Raman spectrum (c) shows a peak at 363 cm⁻¹ which is associated with the greigite phase of iron sulphide²⁷ – it is known that the mackinawite phase oxidizes to the greigite phase upon exposure to ambient air^{24,28}.



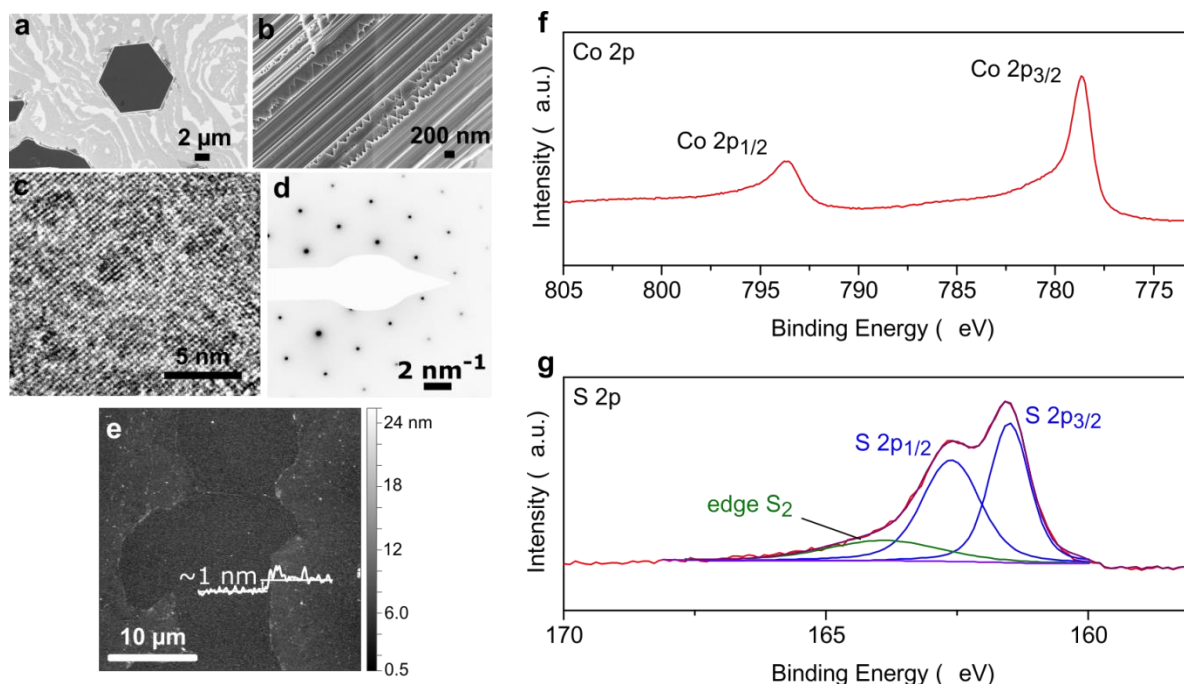
Supplementary Figure 14. Characterisation of iron selenide (FeSe). (a) High magnification and (b) low magnification SEM images of FeSe on gold. (c-e) XPS spectra of the gold surface post-growth: (c) Fe 2p, (d) Se 3d, and (e) survey scan.

Deconvolution of the Fe 2p XPS spectra for FeSe was done using the FeS spectrum as a guide. Since a considerable amount of oxygen is present from the survey spectrum, we have only identified the peaks from the Fe oxidation states, as the iron oxide peaks are convoluted with the selenide peaks. The Fe(II) peaks at 708.8 eV and 722.1 eV correspond to the Fe²⁺ oxidation state. Additional peaks at 710.4 eV, 712.4 eV, 723.9 eV and 726.5 eV are attributed to Fe³⁺ oxidation states. Satellite peaks at 715.4 eV, 729.9 eV and 718.5 eV, 733.8 eV are attributed to the Fe²⁺ and Fe³⁺ states, respectively. Further oxide/multiplet peaks may be convoluted with the satellite and Fe³⁺ multiplet peaks. The peaks at 54.5 eV and 55.6 eV in the Se 3d region correspond to FeSe^{29,30}.



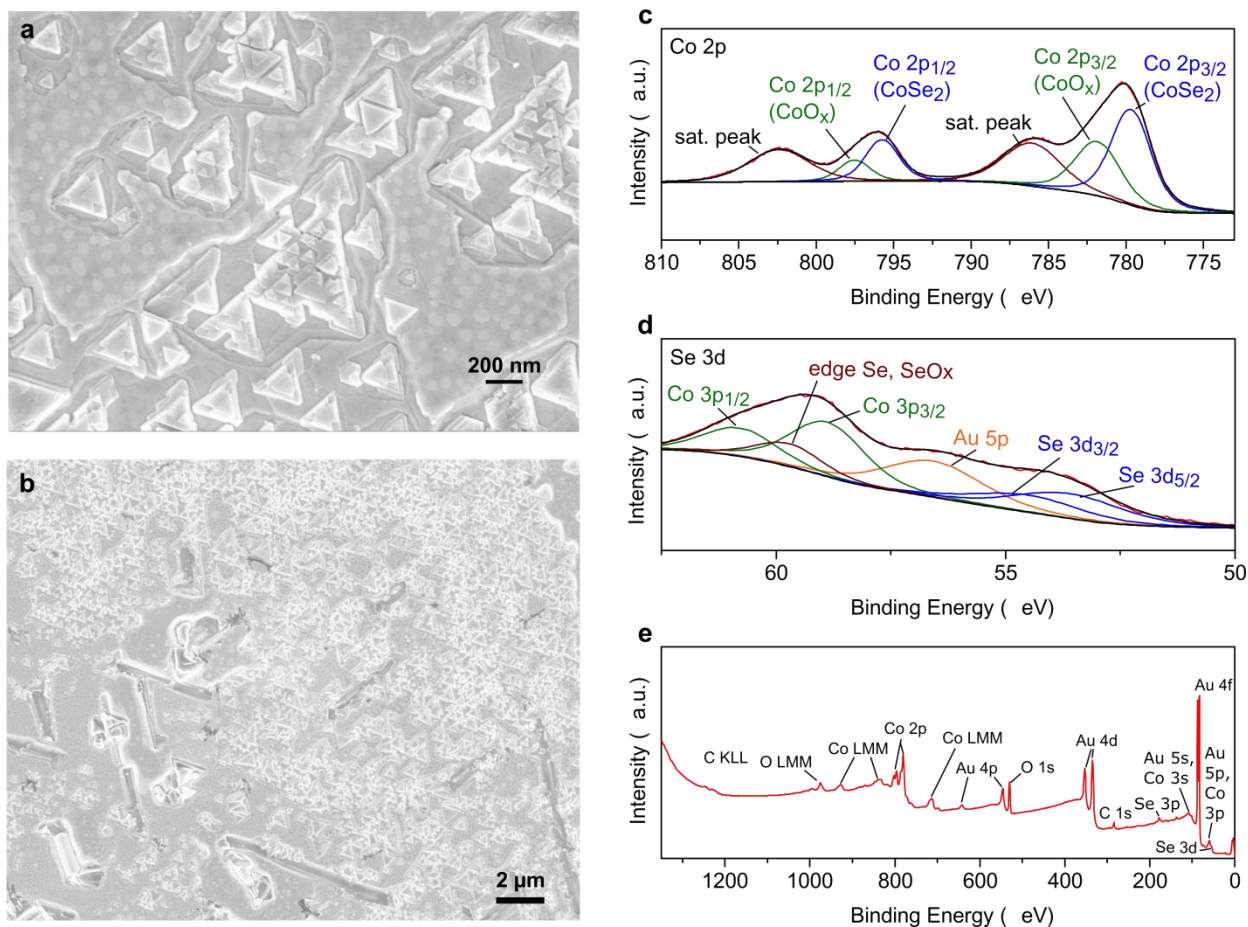
Supplementary Figure 15. Characterisation of iron telluride (FeTe). (a) High magnification and (b) low magnification SEM images of FeTe on gold. (c-e) XPS spectra of the gold surface post-growth: (c) Fe 2p, (d) Te 3d, and (e) survey scan.

Deconvolution of the Fe 2p region for FeTe was done using FeS and FeSe XPS spectra as guides. As with the case of FeSe, oxygen is present on the surface, and as such only the oxidation states are identified in the Fe 2p region. The peaks at 708.3 eV and 721.4 eV are assigned to the Fe²⁺ oxidation state. Further peaks in (c) at 709.1 eV, 710.5 eV, 712.4 eV, 723.4 eV, 726.1 eV and 729.1 eV are attributed to multiplets of the Fe³⁺ oxidation state. The peaks at 715.2 eV, 731.9 eV and 718.3 eV, 734.9 eV are assigned to the Fe²⁺ and Fe³⁺ satellite peaks, respectively. Te 3d peaks at 572.5 eV and 582.9 eV in (d) correspond to FeTe^{11,30}. FeTe also shows evidence of epitaxy with the underlying gold substrate, as seen in the SEM images (a-b).



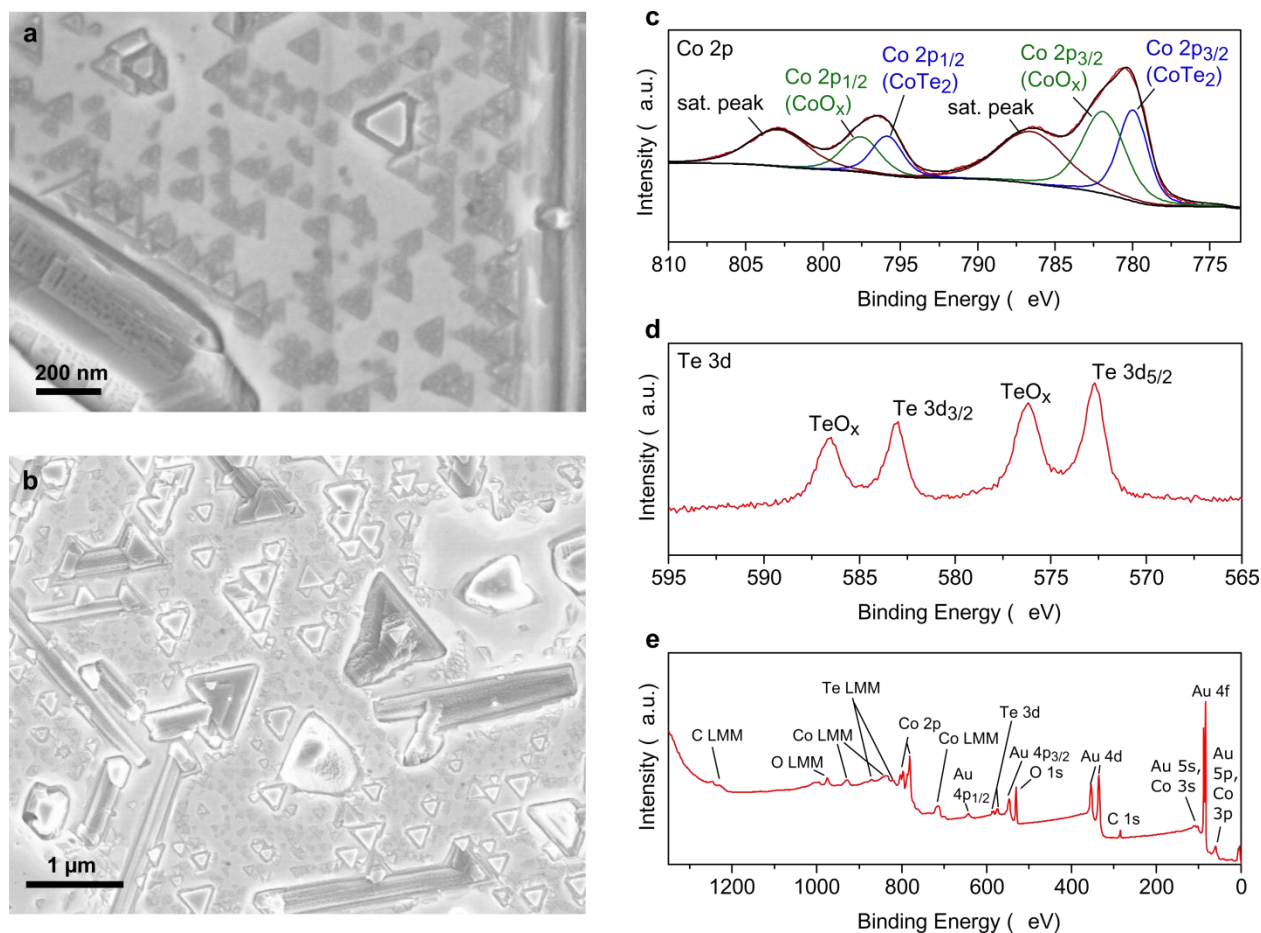
Supplementary Figure 16. Characterisation of cobalt disulphide (CoS₂). (a) Low magnification SEM image of CoS₂ on gold, and (b) a high magnification SEM image of one of the edges of the 3D crystals in (a), showing a resemblance to layered crystals. (c) high resolution TEM image and (d) SAED pattern of one of the edges of the 3D crystals in (a). Monolayered regions in (a) were too unstable under e-beam irradiation for TEM imaging at 200 kV. (e) AFM of transferred domains in (a) onto 90 nm SiO₂/Si substrate, where the measured thickness of the monolayered regions is ~1 nm, not accounting for chemical contrast between tip and surface. (f-g) XPS spectra of the gold surface post-growth: (f) Co 2p and (g) S 2p regions.

The growth behaviour of sulphides of the ferrous metals Co and Fe bear similarities. We observe monolayer films of CoS₂ (light gray films in (a)) as well as 3D crystals (black hexagon in (a)) similar to those seen for FeS but not seen for any of the other metals tested in this work. The 3D crystals here appear to resemble layered crystals from the SEM (b), but appear to have a cubic crystal structure as seen from the BF-TEM and SAED patterns in (c-d). The XPS Co 2p peaks at 778.7 eV and 793.7 eV, and the S 2p peaks at 161.5 eV and 162.6 eV correspond to literature values for CoS₂^{31,32}, whereas as the Co:S ratio is 1:2. We attribute the peak at 163.9 eV in the S 2p spectrum to sulphide edges²⁶. No Raman signal was measureable on as-grown or transferred films with our setup.



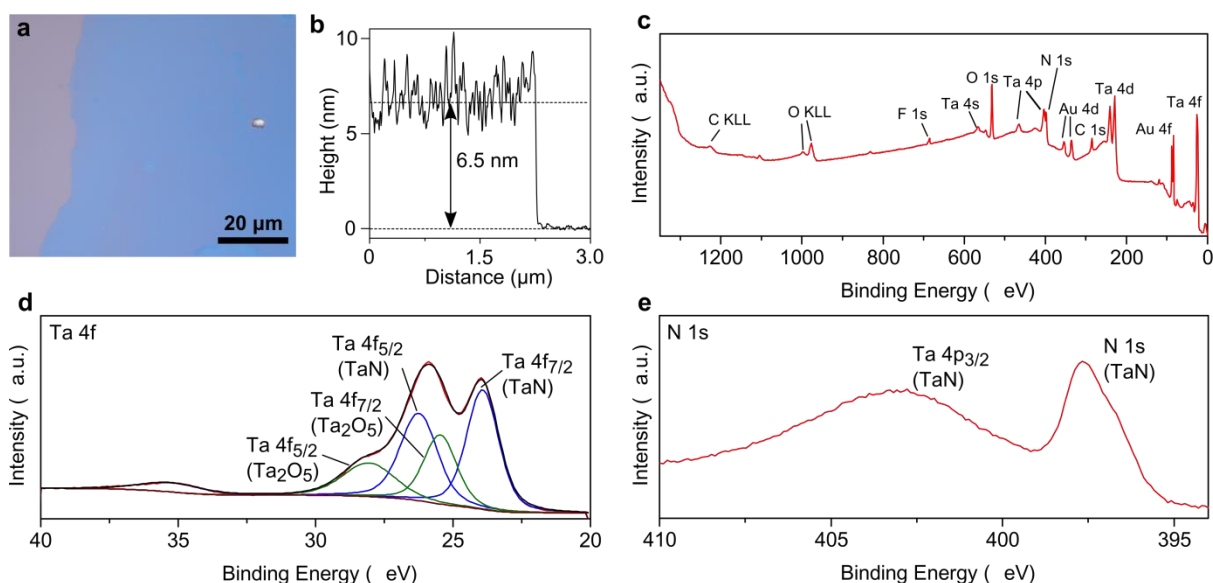
Supplementary Figure 17. Characterisation of cobalt diselenide (CoSe₂). (a) High magnification and (b) low magnification SEM images of CoSe₂ on gold. (c-e) XPS spectra of the gold surface post-growth: (c) Co 2p, (d) Se 3d, and (e) survey scan.

A number of structures are observed in the SEM images (a-b), which we believe are a combination of CoSe₂ and cobalt oxides. XPS scan of the Co 2p region confirms the presence of CoSe₂: Co 2p peaks at 779.7 eV and 795.8 eV correspond to reported values for CoSe₂^{33,34}. Co 2p peaks at 781.8 eV and 797.5 eV are attributed to cobalt oxides, while the peaks 786.1 eV and 802.4 eV are satellite peaks of the Co²⁺ oxidation state^{33,34}. Se 3d peaks at 53.6 eV and 54.5 eV correspond to CoSe₂³³⁻³⁵.



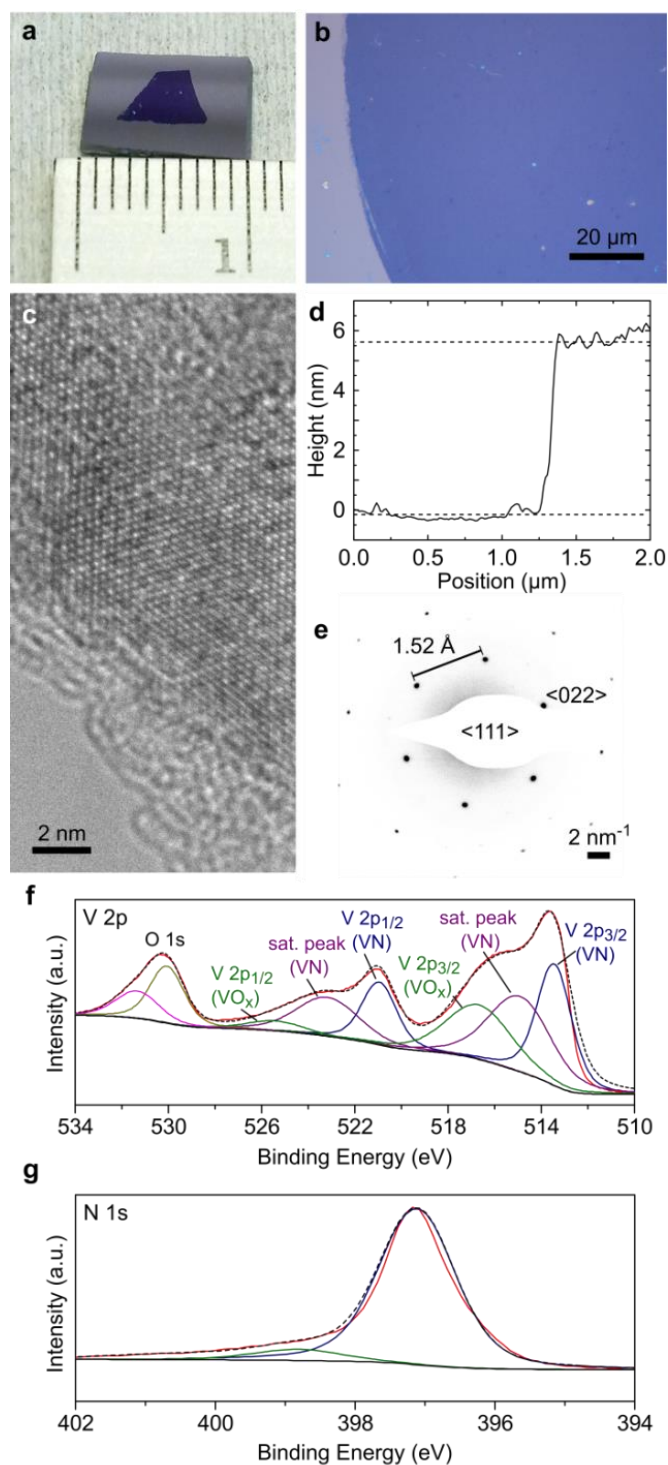
Supplementary Figure 18. Characterisation of cobalt ditelluride (CoTe₂). (a) High magnification and (b) low magnification SEM images of CoTe₂ on gold. (c-e) XPS spectra of the gold surface post-growth: (c) Co 2p, (d) Te 3d, and (e) survey scan.

A variety of structures are observed in the SEM images (a-b), which bear resemblance to those seen in the case of CoSe₂. Deconvolution of the Co 2p spectrum (c) was done using the CoSe₂ case as a guide: Co 2p peaks at 779.9 eV and 795.9 eV correspond to CoTe₂, while the peaks at 781.9 eV and 797.6 eV are attributed to cobalt oxides; Peaks at 786.6 eV and 802.9 eV are Co²⁺ satellite peaks. Telluride peaks in the Te 3d region are seen at 572.7 eV and 583.0 eV, while the peaks at 576.2 eV and 586.6 eV correspond to oxidised tellurides¹¹. CoTe₂ appears to be more air-sensitive than CoSe₂, as seen from the oxidised telluride peaks and the prominent cobalt oxide peaks, and from higher oxygen content in the survey spectrum. The triangular structures in (a) correspondingly show visible signs of oxidation/degradation.



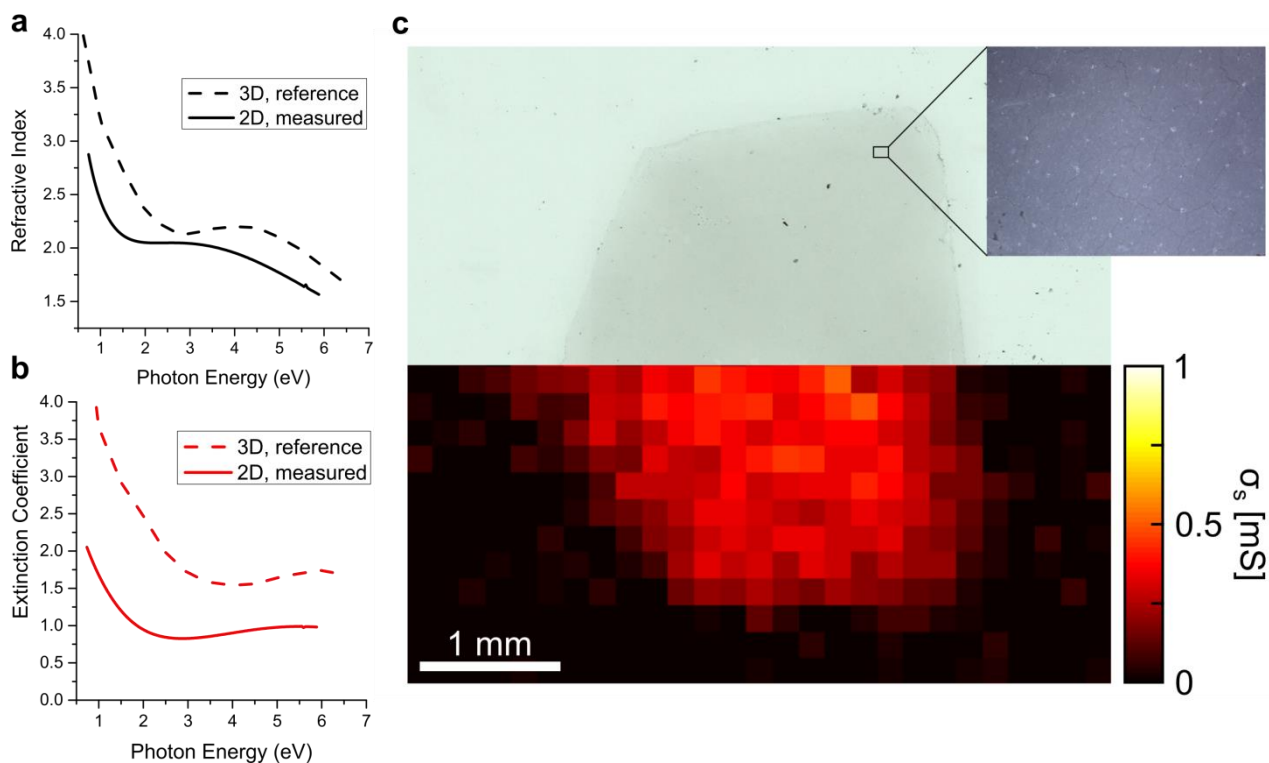
Supplementary Figure 19. Characterisation of ultrathin tantalum nitride films (TaN).

(a) Optical image of transferred TaN on 90 nm SiO₂/Si substrate. **(b)** AFM line profile of transferred TaN films; the film thickness is ~6.5 nm. **(c-e)** XPS spectra of the gold surface post-growth: **(c)** survey spectrum, **(d)** Ta 4f, and **(e)** N 1s scan. Ta 4f_{7/2} and Ta 4f_{5/2} peaks at ~24.0 eV and ~26.0 eV in (d) and the Ta 4p_{3/2} and N 1s peaks in (e) correspond to reported values for hexagonal TaN³⁶; the Ta:N ratio here is ~1.1:1. The films show a noticeable amount of oxidation, as seen by the Ta₂O₅ peaks in (d) and the large oxygen peak in the survey spectrum. The fluorine and carbon signals in the survey spectrum are from backstreamed carbon contaminants³⁷.



Supplementary Figure 20. Characterisation of ultrathin vanadium nitride films (VN).

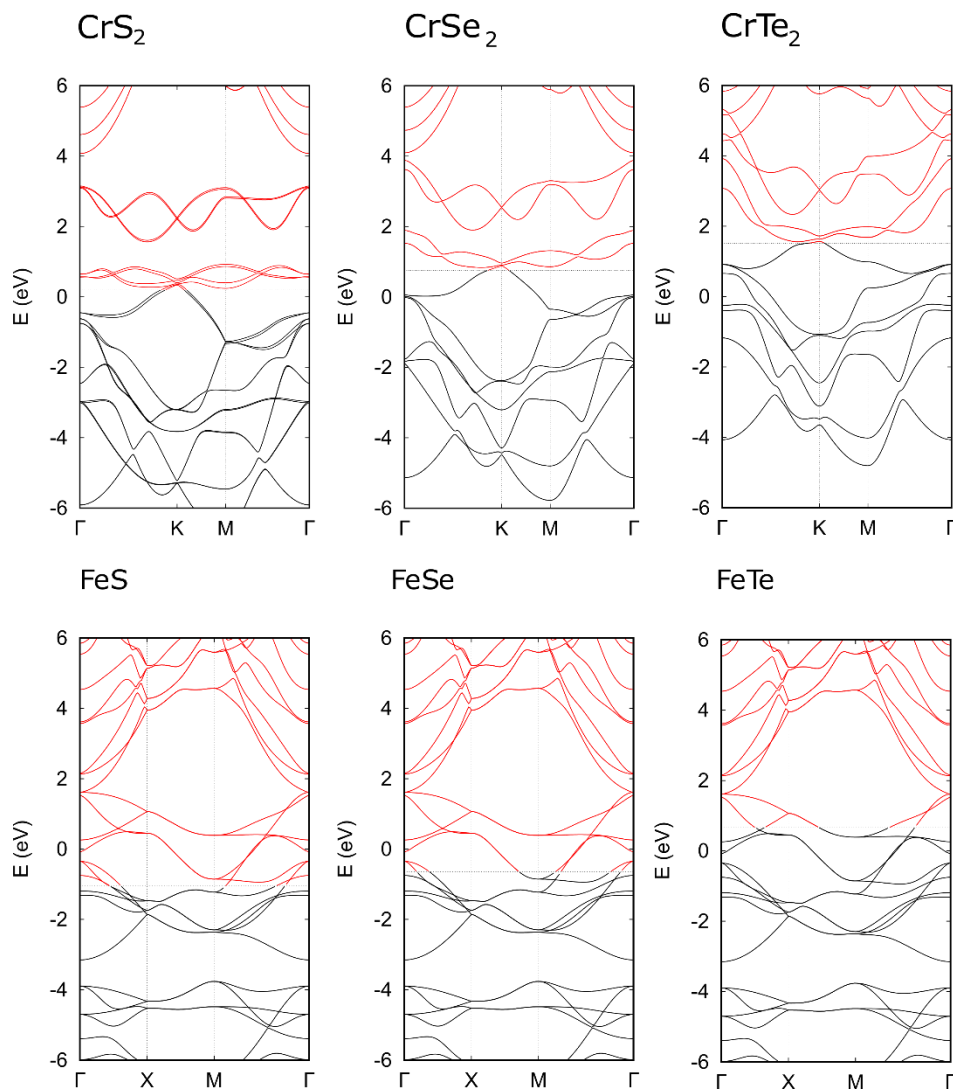
(a) Photograph of ultrathin vanadium nitride film transferred to 90 nm SiO₂ on Si substrate (scale in cm). **(b)** Optical microscope image of film in (a). **(c)** High resolution bright field TEM micrograph of suspended vanadium nitride film. **(d)** Atomic force microscopy line scan of edge of transferred vanadium nitride film; the measured AFM thickness is ~ 5.5 nm. **(e)** SAED pattern of suspended vanadium nitride film. **(f-g)** XPS spectra of as-grown vanadium nitride on gold. Partial oxidation is visible in the spectrum in (f).



Supplementary Figure 21. Ellipsometry, optical and THz-TDS characterisation of ultrathin vanadium nitride films on quartz. (a-b) Optical functions of 5.5 nm VN (labelled 2D) extracted by least squares fitting of ellipsometry spectra using a parameterised optical function model. The model consists of one Drude oscillator (three fitting parameters) to model free carrier absorption in the infrared, and one Lorentz oscillator (three fitting parameters) to model the response in the visible and near ultraviolet. Optical functions obtained from literature³⁸ for the bulk VN films are also shown (labelled 3D). (c) Optical/THz-TDS sheet conductivity map (mirrored) of transferred VN films on quartz. The sheet conductivity map shows the average sheet conductivity in the range from 0.8-0.9 THz.

Supplementary Figure 20a-b show a centimetre-scale continuous layer of ultrathin vanadium nitride (VN) transferred onto a 90 nm SiO₂ on Si substrate. The size of the film here was limited only by the dimensions of the growth substrate. The thickness of the film is ~5.6 nm, as measured by AFM (Supplementary Figure 20d), and is uniform across the sample, as confirmed by optical microscopy and ellipsometry (Supplementary Figure 20b, Supplementary Figure 21a-b). HRTEM and SAED of the VN films (Supplementary Figure 20c, e) establishes an FCC lattice structure with the {111} facet perpendicular to the growth substrate. Notably, the lattice spacing here, 1.52 Å, is ~4% larger than that of bulk cubic VN, 1.46 Å (ICDD database). XPS characterisation of as-grown VN films on gold (Supplementary Figure 20f, g) shows the expected nitride bonding between vanadium and nitrogen³⁹, with a V:N ratio of ~1:1. Some degree of oxidation was always present in the

films even when grown under highly reducing conditions, likely due to exposure of the films to ambient conditions. The sheet resistance of transferred VN layers as determined by van der Pauw measurements is $\approx 2 \text{ k}\Omega\text{sq}^{-1}$ immediately after transfer, increasing to $\approx 50 \text{ k}\Omega\text{sq}^{-1}$ after 100 days of exposure to ambient conditions, which supports the idea that VN layers are air- or humidity-sensitive. Terahertz time domain spectroscopy results performed 15 days after transfer show an average sheet resistivity of $2 \text{ k}\Omega\text{sq}^{-1}$ across the transferred VN film (Supplementary Figure 21c).



Supplementary Figure 22. Band structure calculations for select materials synthesized. Simulated structures for the CrS₂, CrSe₂, and CrTe₂ are of the CdI₂ type monolayers, while for FeS, FeSe, FeTe the simulated structure is of the PbO monolayer type.

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