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Upscaling two-dimensional $MoSe_2$ and hBN via controlled chemical vapor deposition

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ABSTRACT

Two-dimensional materials (2DM) are a topic of high interest in the materials science community to engineer innovative devices. Among them, hexagonal boron nitride (hBN) is an optimal candidate to serve as insulating/passivation layer in graphene-based devices, enabling high carrier mobilities [1]. Semiconducting 2D transition metal dichalcogenides (TMDC) have shown outstanding optoelectronic properties, such as thickness-dependent photoluminescence, combined with lightweight and flexibility [2,3]. Notwithstanding a considerable effort, a production method for 2DMs that would guarantee large scale, high throughput and low cost is still lacking. Chemical vapor deposition (CVD) is perhaps the most promising route for the batch production of 2DMs with high quality, but a few crucial challenges need to be addressed to attain cost-effective and reproducible processes over large areas [4].

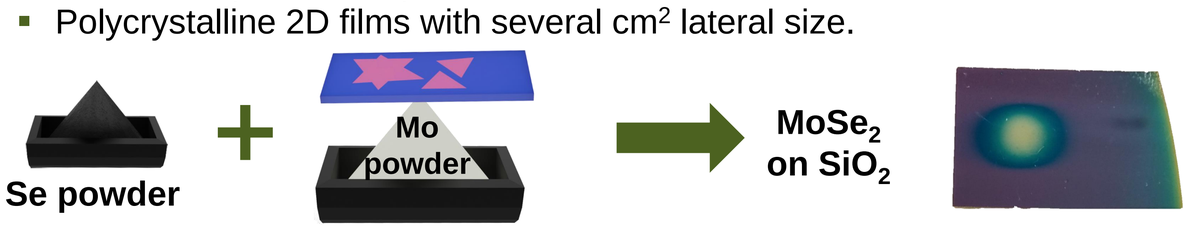
METHODOLOGY AND RESULTS

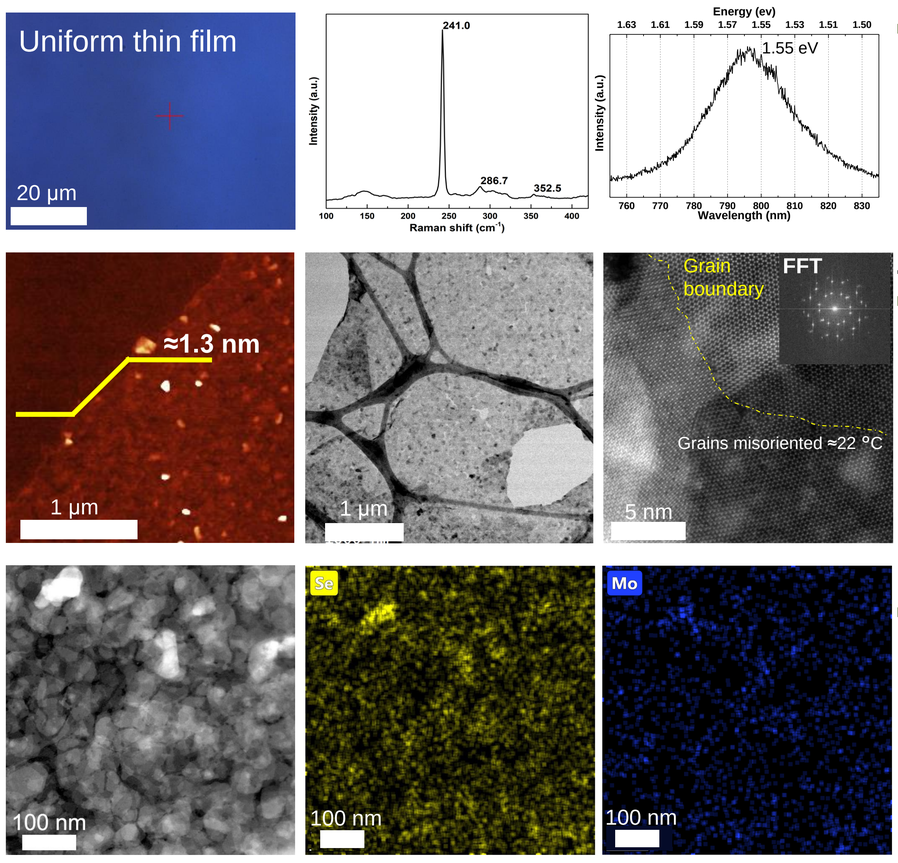
Two-dimensional $MoSe_2$ crystals grown by atmospheric pressure-CVD (AP-CVD)

The growth was carried out in a quartz-tube furnace at atmospheric pressure by two different approaches, using Ar + H_2 carrier gases. During the growth (10-30 min), the Se powder was heated to ~400 °C, while the Mo precursor and the substrate were held at 700-800 °C.

Approach #1: Atomic-thick $MoSe_2$ film on Si/SiO_2

- Se and Mo powder as precursors.
- Polycrystalline 2D films with several cm^2 lateral size.

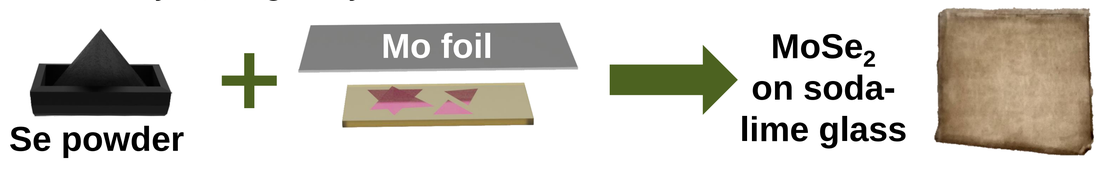


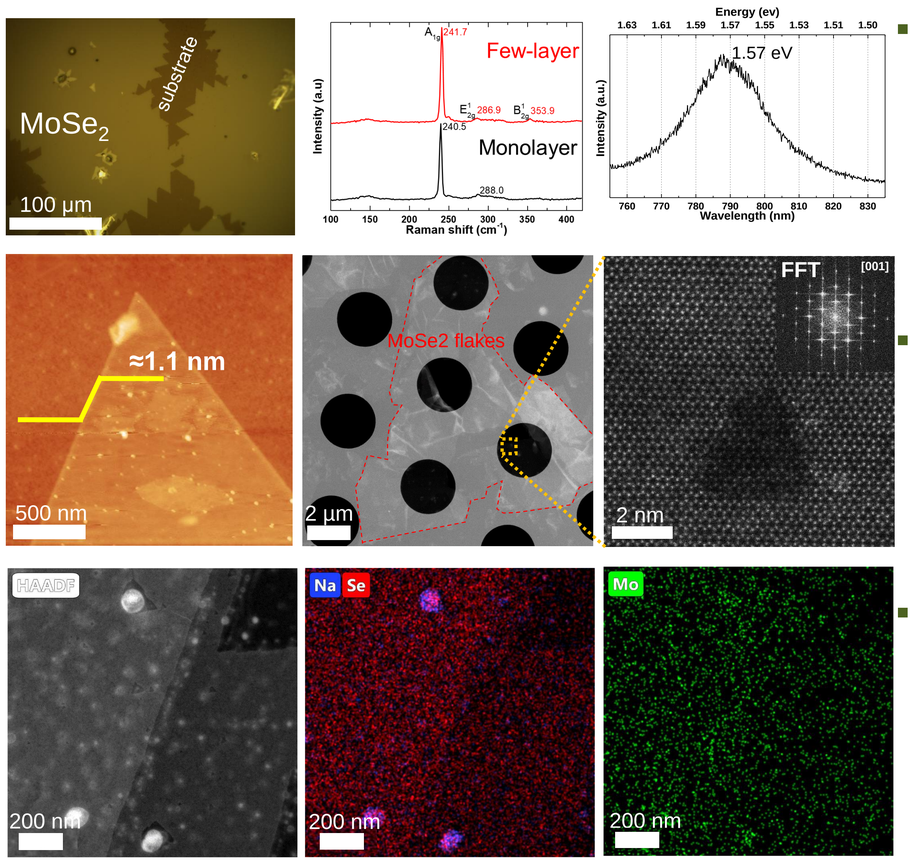


- 2 cm^2 uniform region. Raman peak at 241 cm^{-1} and PL emission at 1.55 eV: bilayer $MoSe_2$ film.
- AFM height profile of 1.3 nm. STEM shows the polycrystalline structure of the film, with grain size 60-100 nm.
- Mo and Se are the main elements detected by EDS, no contamination or impurities found.

Approach #2: Monolayer $MoSe_2$ flakes on glass

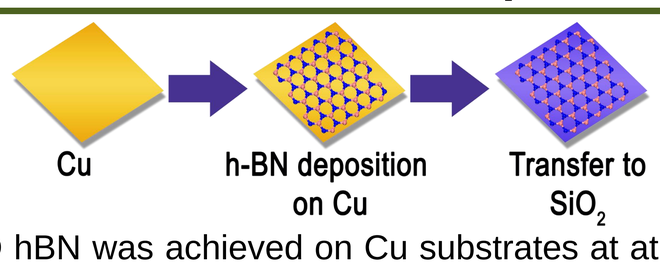
- Se powder and Mo foil structure covering the glass substrate as precursors.
- Monolayer, single crystals with sub-mm lateral size.



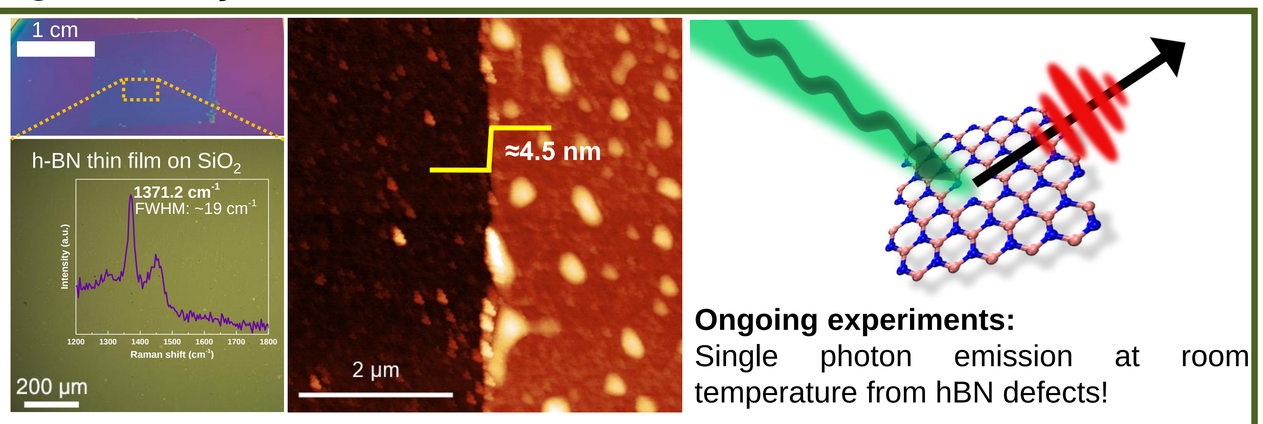


- Single crystals with lateral size > 100 µm. Raman peak at 240.5 cm^{-1} and PL emission at 1.57 eV: monolayer $MoSe_2$.
- AFM height profile of 1.1 nm. STEM shows highly crystalline structure of the layer, with few defects.
- Other than Mo and Se, EDS shows the presence of Na particles, acting as growth promoter during the process

Two-dimensional hBN films up to several cm^2 size grown by AP-CVD



The growth of 2D hBN was achieved on Cu substrates at atmospheric pressure in a two-zone quartz-tube furnace. A mixture of Ar/H_2 was used as carrier gas and ammonia borane (AB) powder as solid precursor. After the substrate annealing (40 min at 1020 ºC) and a pre-treatment of the AB powder (150 min at 100 ºC), the growth step begun (30 min at 1020 ºC). The transfer process to a SiO_2 substrate was done via a wet-etching method, using a PMMA as the sacrificial polymer layer and $FeCl_3$ (0.5M) as Cu etchant.



Ongoing experiments:
Single photon emission at room temperature from hBN defects!

- Raman spectroscopy: E_{2g} phonon mode at 1371.2 cm representative of few-layer hBN, with no contaminations detected.
- AFM analysis confirms a film thickness below 5 nm.

CONCLUSIONS

- **Growth of atomic-thick $MoSe_2$ by AP-CVD using two approaches:**
  - **1) Few-cm^2 polycrystalline films on Si/SiO_2**
  - **2) Sub-mm^2 single crystals on soda-lime glass, exploiting the effect of Na atoms released by the substrate during CVD to increase the lateral size of the 2D crystals.**
- **Growth of continuous few-layer hBN films by AP-CVD**
  - **High reproducibility towards wafer-size areas.**
  - **Samples with controlled features to be applied and studied as single photon emitters at room temperature.**

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