Epitaxial wafer scale growth of tungsten dichalcogenides

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Transition metal dichalcogenides (TMDs) have been a focus of interest due to the direct band gap of the monolayers in the range of 1.6 – 2.0 eV and large exciton binding energies, which leads to interesting electronic and optical properties. One major challenge in harnessing the potential of these materials is the growth of high quality epitaxial mono- and few-layer films over large areas. The growth on C-plane sapphire is expected to lock the domain orientations at 0° and 60° due to the hexagonal symmetry, which can result in epitaxial TMD films with reduced high-angle grain boundaries.[1] Oriented growth for TMDs like MoS_2[1] and WSe_2[2] has been demonstrated by powder vapor transport on C-plane sapphire previously. However, we find that in a cold-wall metal organic chemical vapor deposition (MOCVD) process, even though this orientation relation is maintained for WSe_2, this locking does not extend universally to WS_2. The reason for this difference is crucial in not only obtaining oriented films but also understanding the basic interactions between the TMDs themselves and/or the precursors involved and the sapphire substrate.

In this work, WS_2 and WSe_2 mono- and few-layer films were deposited by MOCVD system on 2” C-plane sapphire wafers using tungsten hexacarbonyl ($W(CO)_6$), hydrogen selenide (H_2Se) and purified hydrogen sulfide (H_2S). The growth was carried out for 1 h at 800- 900°C for WSe_2 and 850-1000°C for WS_2 to achieve fully coalesced films with domains on the order of 1 μm in size. The results show that there is a distinct difference in the growth of WSe_2 and WS_2 films. Though both WSe_2 and WS_2 have an epitaxial relation with the underlying sapphire substrate, the WSe_2 domains are predominantly oriented at 0° and 60°, but the WS_2 films show presence of domains at other angles as well, as shown in Figure 1. In case of WSe_2, the orientation of 0° and 60° is maintained at all the growth temperatures investigated, but for WS_2, the domains are oriented at 0° and 30° at lower temperatures between 750-850°C. Additional orientations emerge at 900-1000°C. Further details about the epitaxial relation, the interface interaction and the differences in the growth of WS_2 and WSe_2 will be presented.

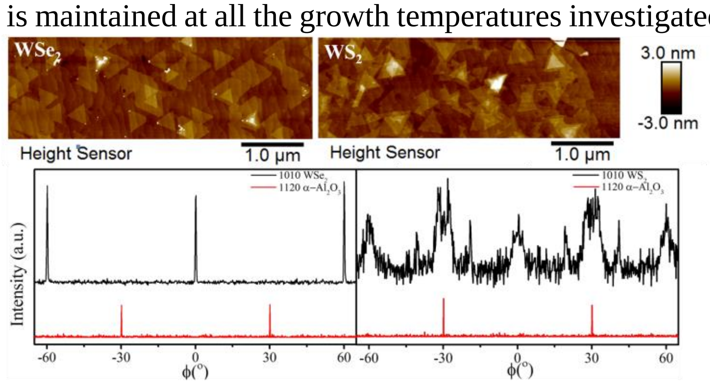

Figure 1: Atomic force microscopy images and the phi- scans highlighting the growth of WSe_2 oriented at 0° and 60° and the multiple orientations observed for WS_2.

[1] D. Dumcenco, .D. Ovchinnikov, K. Marinov, P. Lazi, M. Gibertini, N. Marzari, O. L Sanchez, Y. C. Kung, D. Krasnozhon, M. W. Chen, S. Bertolazzi, P. Gillet, A. F. Morral, A. Radenovic, A. Kis, *ACS Nano*, 9(4), **2015**, 4611–4620.
[2] L. Chen, B. Liu, M. Ge, Y. Ma, A. N. Abbas, C. Zhou*, *ACS Nano*, *9* (8),**2015**, 8368–8375.

Supplementary Page

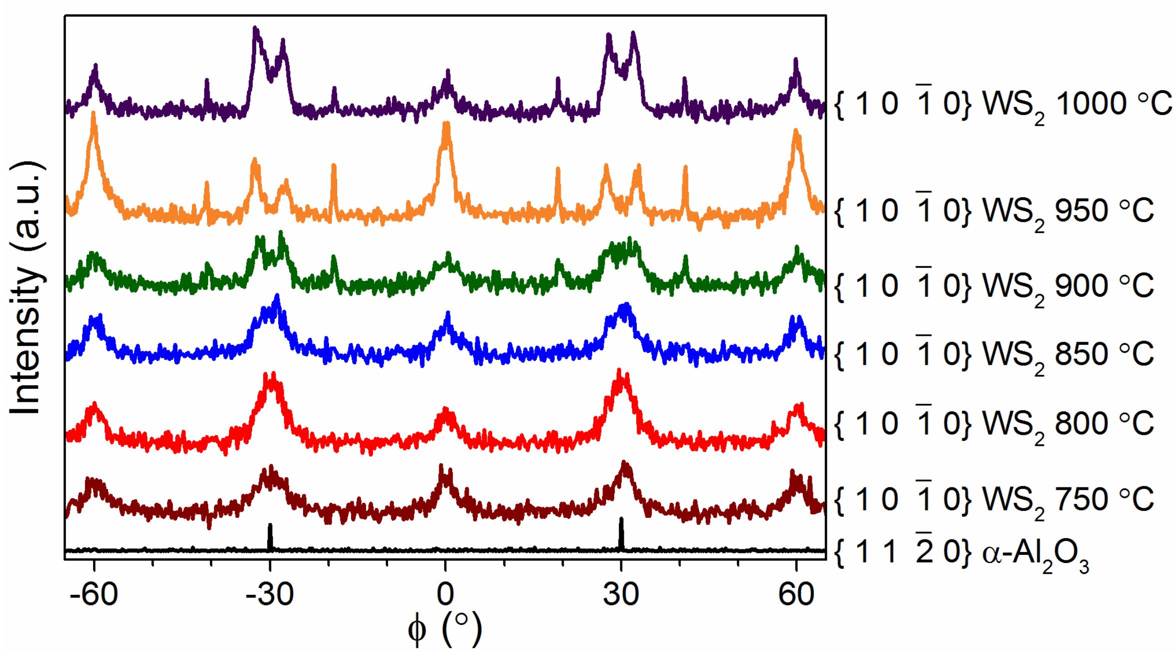

Figure 2 shows the evolution of the in-plane orientations of WS_2 as a function of the deposition temperature.

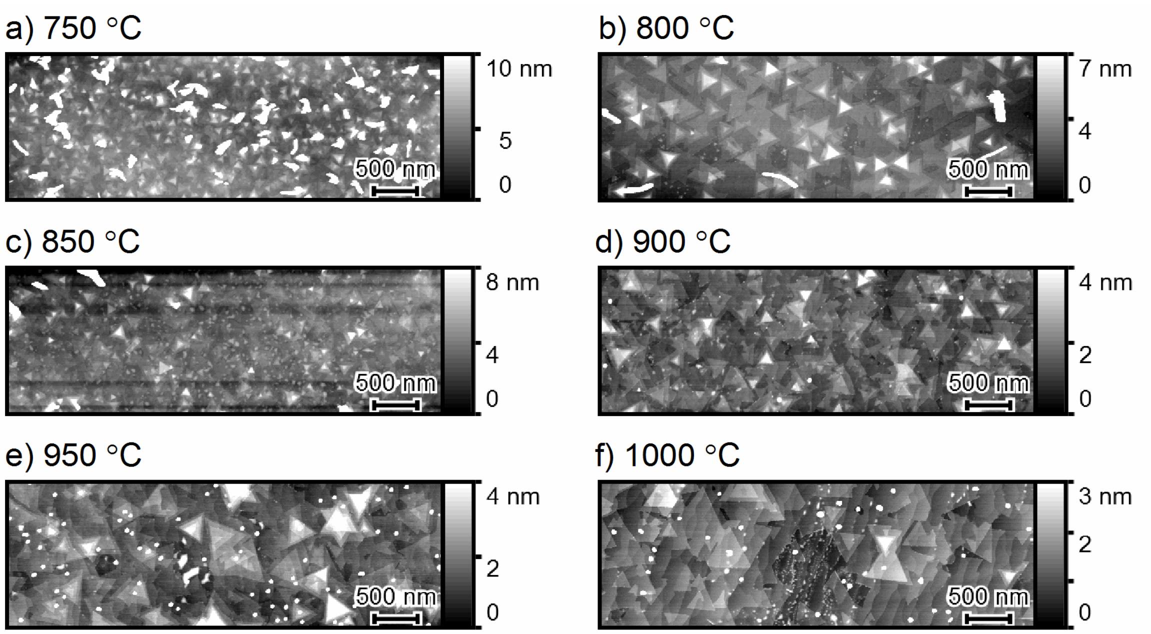

Figure 3 (a-f) shows the atomic force morphology of WS_2 as a function of the deposition temperature.

At lower temperatures, the films are oriented at 0 and 30° but at temperatures greater than 850°C, other orientations become prominent. The growth temperature plays a significant role in determining the orientation of growth.