Fabrication of high quality, large-area twisted TMD bi-layers

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Since the discovery of the magic angle in twisted bilayer graphene, twisted bilayer systems have attracted significant research interest due to their potential applications in the devices of the future. These structures exhibit novel properties, including strongly correlated electronic effects, modifications of the electronic and phononic band structure and more. [1, 2] Among the most extensively studied materials in this context are transition metal dichalcogenides (TMDs), a class of Van der Waals materials whose properties depend on the number of layers and their relative twist angle. [3] However, the controlled growth and transfer of large, high-quality single-crystal monolayers remain challenging. This process is crucial, as the twisted bi-layers strongly depend on the interface quality. To address this, our research focuses the controlled growth and transfer of large-area, high-quality MoS₂ and WS₂ single-crystal monolayers using chemical vapor deposition (CVD), using metal precursor salts and optimizing the growth on a variety of substrates. Figure 1 a, schematically illustrates the conducted fabrication process and show microscopic images of the grown WS2 monolayers on **b** SiO2/Si and **c** sapphire. Monolayers are subsequently transferred using differently treated polydimethylsiloxane (PDMS) stamps for precise alignment and clean transfer. This process is conducted under low-pressure and lowtemperature conditions to minimize residue formation, as illustrated in Figure 1d. The resulting samples are characterized using photoluminescence spectroscopy, Raman spectroscopy, atomic force microscopy (AFM), and transmission electron microscopy (TEM) to determine the cleanest method for fabricating high-quality twisted bilayers (Fig. 1 e,f)



Figure 1. a) Capping method to grow big single crystal monolayers. b) WS2 monolayer in SiO2/Si c) WS2 monolayers CVD growth on sapphire. d) Transfer process for twisted bilayers. e) WS2-WS2 twisted homobilayers (green: bottom triangle, red: top triangle).

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- [2] Tang, B et. al. Small Structures, 2(5).
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