High-quality WS2 monolayers growth via two-step approach based on pulsed laser deposition of tungsten oxide

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Two-dimensional (2D) layered atomic crystals have received great scientific attention during the past decade since the discovery of graphene [1]. Among 2D materials crystal family transition metal dichalcogenides (TMDs) are the most promising candidates for potential applications in optoelectronic devices, and great efforts have been devoted to study their electronic and optical properties [2]. It was shown that single layers of MoS2, WS2, MoSe2 and WSe2 exhibit direct bandgaps ranging from visible to near-infrared (between 1-2 eV), which gives rise to a strong light-matter coupling in these materials. Moreover, beyond the devices consisting of single layer TMDs, atomically-thin van der Waals heterostructures based on 2D TMDs with a varied bandgaps can be synthesized, without a stringent requirement of lattice matching between two constituent materials [3]. Tungsten disulfide (WS2) is a promising representative of the semiconducting TMDs. Unlike its bulk counterpart, WS2 monolayers exhibit a direct bandgap of ~2eV at the corners (K and K' points) of the Brillouin Zone, one of the lowest electron effective mass, and exceptionally bright photoluminescence (PL). A strong light absorption and emission at visible wavelengths opens great opportunities for its integration in ultrathin optoelectronic devices, such as light-emitting diodes, photovoltaic cells, and microcavity lasers. However, one of the top challenges has being faced is designing a method to grow uniform and highly oriented WS2 monolayers with a precise control over the layers number on a large scale. In this study we report on high-quality WS2 films synthesis using two-step approach based on pulsed laser deposition (PLD) of WOx epitaxial oxide. PLD of oxides using background gas is applied to grow high-quality epitaxial oxides with sharp heterointerfaces, and offers great opportunities towards crystalline quality of the material and its composition control. The controllability of the oxide growth conditions and its crystallinity is favorable for a second step, which is oxide high-temperature sulfurization. This is an advantage of this method unlike other two-step processes. We found that conversion from oxide to sulfide appears to be efficient if the crystallinity of the oxide precursor is increased by oxide high-temperature growth. Also, we explored that oxygen vacancies presence is favorable for oxide conversion efficiency to WS2 films. As a result, high quality monolayers of WS2 with a low amount of defects were synthesized over a larger area. Our WS2 films show strong light-matter coupling, as indicated by PL signal. Tungsten oxide composition and crystallinity and WS2 films' properties were analyzed by Raman spectroscopy, PL spectroscopy, atomic for microscopy (AFM), scanning tunneling microscopy (STEM), and X-ray photoelectron spectroscopy.

References

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Figures

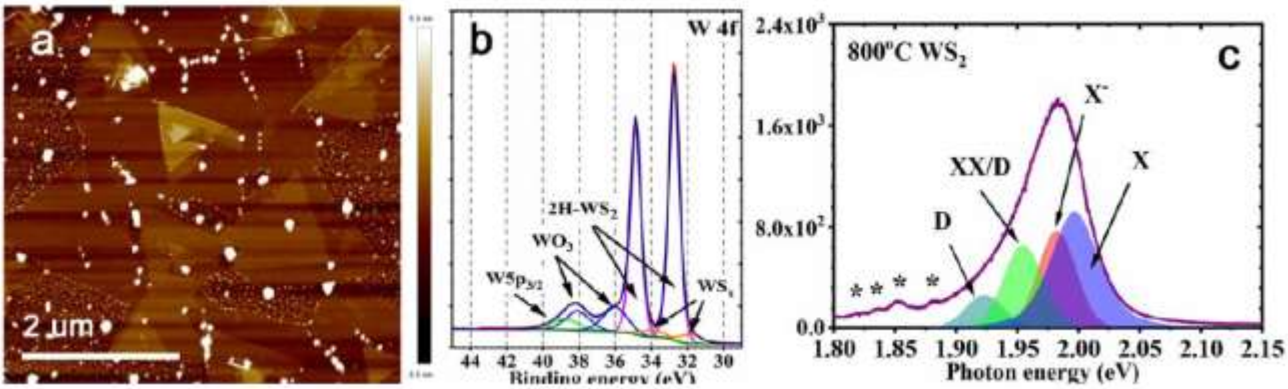

Figure 1: a) AFM, b) W4f XPS spectrum, and c) PL spectrum of a single layer WS2 grown by high temperature sulfurization of PLD-deposited WOx film