Hydrogen-Induced Epitaxial Growth of Monolayer WS_2 and Orientation-Dependent Grain Boundaries

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Transition metal dichalcogenides (TMDCs) have been attracting a great interest, because they have a finite band gap, high mobility, and mechanical flexibility which promise applications in flexible electronics and optoelectronics[1]. For such applications, chemical vapor deposition (CVD) is a powerful method, because it enables the large-scale growth with relatively low cost. However, in the standard CVD system using Ar carrier gas, TMDC grains are randomly oriented and a multilayer grain exists at the center of each grain, resulting in degraded device performance[2]. Therefore, it is very important to develop a new method to grow multilayer-free monolayer TMDCs with controlled grain orientation.

In this work, we demonstrate that high concentration of hydrogen gas in the CVD process enhances the interaction between WS_2 and sapphire surface, resulting in the highly aligned WS_2 grains. Figure 1 compares the WS_2 grains obtained by the standard CVD with Ar gas and our method using high concentration of H_2. It is clearly seen that WS_2 grains are oriented, registered by the underlying the sapphire surface. In addition, the AFM measurements revealed that the WS_2 grains grown with H_2 have no multilayer grains at the center of monolayer grains. Photoluminescence (PL) quenching was observed, reflecting strong coupling between the WS_2 and the substrate. Figure 2(a,b) shows scanning transmission electron microscopy (STEM) images of interfaces between two merged grains. The misaligned WS_2 showed various kinds of GB structures depending on misalign angles[3]. On the other hand, the aligned WS_2 grains possessed unique dislocations with less point defects and impurities. Finally, field-effect transistors (FETs) were fabricated to understand the influence of GBs on the carrier transport property (Fig. 2c,d). The conductance degradation across GBs observed in the misoriented WS_2 grains was not seen for the oriented WS_2, suggesting the importance of the controlling the grain orientation for obtaining high device performance. Our work offers a new approach to control the WS_2 orientation, which will accelerate the potential applications of 2D materials.

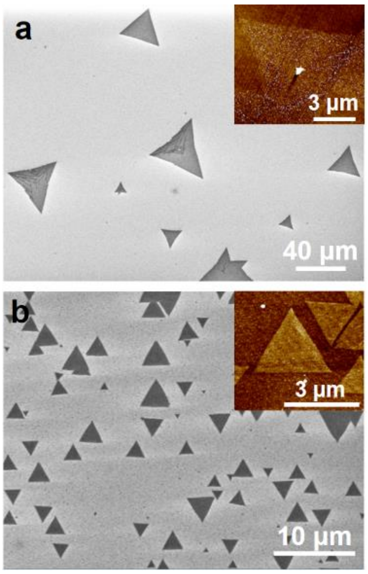

Fig. 1. SEM images of WS_2 grains grown on sapphire. (a) Ar flow, (b) Ar-H_2 flow.

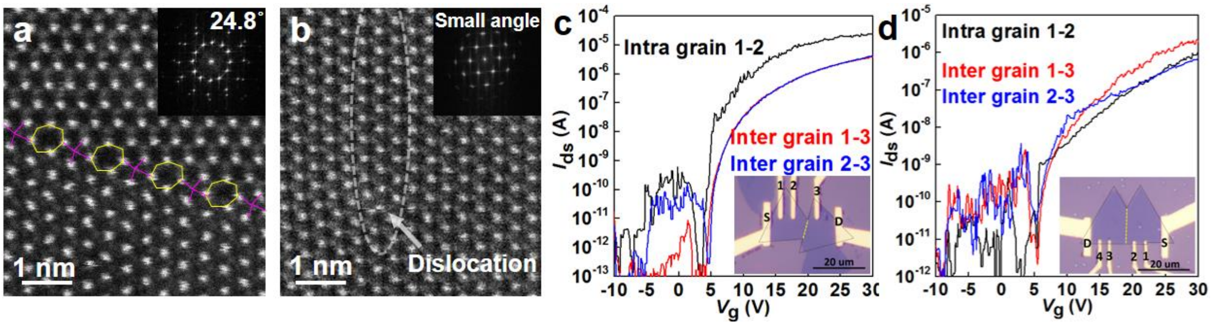

Fig. 2 Annular dark-field (ADF) images of merging grain boundaries (a,b) and corresponding transfer curves (c,d). (a,c) a large misalign angle, (b,d) a very small misalign angle.

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