Insight into 3D structure formation during atomic layer deposition

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Nanoscale deposition techniques are becoming increasingly more important for various applications as the scalability of the current technology reaches the obtainable limit. Two-dimensional, layered materials, such as transition metal dichalcogenides (TMDs), have re-emerged as an intense area of study for future thin film applications [1]. Atomic layer deposition (ALD) is one synthetic technique that permits angstrom-level control of thin films that also exhibits a high degree of conformality over large, diverse surfaces [2]. However, development of ALD is still ongoing and mitigating a transition from in-plane to out-of-plane growth in TMD thin films is important. With increased film thickness, out-of-plane, or 3D, structures appear in addition to lateral film growth, a consequence of several factors (e.g. plane orientation variation, competition between grains, etc.). These 3D structures can significantly affect the overall film properties [3]. Several studies have address ed a formation mechanism for 3D structures, however, a detailed mechanism has not yet been well defined [3,4,5]. WS_2 ALD films of various thicknesses were studied with high resolution scanning transmission electron microscopy (STEM) to understand the 3D structure formation mechanism. Fast Fourier transforms (FFTs) were analyzed from areas surrounding 3D structures to study grain orientations. The angular difference of crystallographically equivalent diffraction spots in the FFTs reveals the misorientation between grains; misorientation angles were measured with respect to the horizontal. The 3D structures were found to predominantly form at grain boundaries with low misorientation angles, low angle grain boundaries, with an average of about 5.6 degrees. These misorientation angles were then compared with other grain boundaries with no 3D structure present. Misorientation angles at grain boundaries with no 3D structures was substantially higher with an average difference of about 16.5 degrees illustrating the preference for 3D structures to form at low-angle grain boundaries. $>$From these data and accounting for higher defect mobility at high angle grain boundaries [6], we propose three general formation mechanisms for 3D structures during ALD: (1) a discontinuous, (2) a preferential, and (3) a concerted mechanism. Understanding the mechanisms behind 3D structure formation during ALD is an important step toward further controlling and manipulating thin films at the nanoscale for future applications.

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