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Quantification of Metal Atom Ordering in Engineered $W_{1-x}Mo_xS_2$ Monolayers

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Engineering the physical and device properties of two-dimensional (2D) materials requires precise understanding of the position and bonding environment of individual atoms [1,2]. Numerous characterization techniques, such as X-ray diffraction (XRD) [3], spatially resolved photoluminescence (PL) [4], X-ray photoelectron spectroscopy (XPS) [5], and Raman spectroscopy [6], are frequently applied to the study of 2D materials and provide a wealth of information. However, most characterization techniques do not directly probe individual atoms and are unable to map atomic positions locally and across length scales.

Aberration-corrected annular dark-field scanning transmission electron microscopy (ADF-STEM) enables the direct visualization of alloy structures and the atomic number-dependent scattering that makes it possible to identify the distributions of dopants or alloying atoms. Several examples have demonstrated this for the alloy $W_{1-x}Mo_xS_2$ [6-9] and linked the atomic distributions with physical properties such as vibrational anisotropy [8] and composition-dependent spin–orbit splitting [9]. For instance, density functional theory predicts that striping of metal atoms in the alloy $W_{1-x}Mo_xS_2$ can lead to a material that is electronically isotropic but possesses anisotropic thermal conductivity [8], and angle-resolved photoemission spectroscopy (ARPES) shows that increasing the W concentration leads to increased spin–orbit splitting between the upper valence bands [9]. The understanding of such phenomena is critical to engineering physical properties, and it is especially important for the controlled design of lateral heterojunctions, which are an emerging class of 2D material monolayers. In monolayer 2D materials such as transition metal dichalcogenides (TMDs), lateral heterojunctions hold great promise for new designs of transistors and other electronic devices that take advantage of the unique geometry, properties, and precise covalent, in-plane bonding in 2D materials [10].

To date, efforts to analyze and quantify metal atoms in 2D material alloys have yielded information about nearest neighbors and coordination shells, and they have shown that overall, many of the materials synthesized display a nearly random distribution of metals [6-9]. A recent analysis of $Mo_xRe_{1-x}S_2$ investigated the thermodynamics of metal distributions and proposed a model from which it is possible to make predictions about the statistics of atomic ordering and the thermodynamic history of the sample. In our work, we build on previous efforts to characterize and classify metal distributions in 2D monolayer alloys. We present results from $W_{1-x}Mo_xS_2$ films grown by metal–organic chemical vapor deposition (MOCVD) that have been intentionally grown with various degrees of anisotropy. Because MOCVD allows the controllable introduction of precursors, it is possible to target the growth of

predominantly random alloys, as well as alloys with ordering of the metal atoms. This, we present quantification of the atomic positions and make connections between the atomic structure and the growth conditions. We have previously demonstrated the importance of considering growth parameters when analyzing atomic-scale features in 2D material monolayers [11-13], and the work presented here makes it possible to uncover trends and engineering principles for the more complicated case of TMD alloys [14].

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