

Observation of Atomic Ordering in a Monolayer Semiconductor Alloy

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Alloying of semiconductors enables fabrication of materials with continuously tunable properties such as lattice constant, bandgap, and carrier mobility, which is the key in development of next generation devices with new design and function (1–4). However, it has long been an open question whether the atoms in the lattice of a multispecies alloy are distributed randomly or they are ordered in some way (5). The answer is fundamentally important as the fundamental properties of the alloy could be significantly affected by the ordering (3, 6–9). Ordering in semiconductor alloys has been experimentally studied, but most of the reports have been limited to reciprocal space (10, 11). Atomically-thin two-dimensional (2D) materials (12–15) offer an ultimate playground to explore atomic-scale structural features in the lattice through direct imaging of their atomic registry in real space.

In this work we synthesize bulk single crystals of $\text{Re}_{0.5}\text{Nb}_{0.5}\text{S}_2$ with a layered structure using chemical vapor transport. We exfoliate the crystals with scotch tape to prepare monolayer crystals for atomic-scale imaging. Figure 1A shows an aberration-corrected annular dark-field scanning transmission electron microscope (ADF-STEM) image of a $\text{Re}_{0.5}\text{Nb}_{0.5}\text{S}_2$ monolayer. Due to the Z-contrast mechanism, Re (heavier) and Nb (lighter) atoms show brighter and dimmer contrasts, respectively. The most noticeable structural feature is the formation of meandering Re (or Nb) stripes, quite different from the atomic registry of other reported monolayer alloys (2, 16, 17). Using statistical analysis on the ADF-STEM images, we verify the presence of short-range ordering in monolayer $\text{Re}_{0.5}\text{Nb}_{0.5}\text{S}_2$. This can be attributed to the interplay between Coulomb interaction between Re and Nb atoms and the geometrical constraints of the lattice, leading to correlated nearest-neighbor pairs.

In addition, the bandgap of a semiconductor is perhaps the most important parameter for optoelectronic applications. Since there are limited bandgap ranges offered by elemental or binary individual semiconductors; alloying has been the most common approach to expand the bandgap range by controlling the alloy composition (18). In particular, 2D materials provide an ideal platform for bandgap engineering by alloy composition (2) and layer number (4). Here, we demonstrate how the induced atomic ordering can be used as another degree of freedom to tune the alloy band gap.

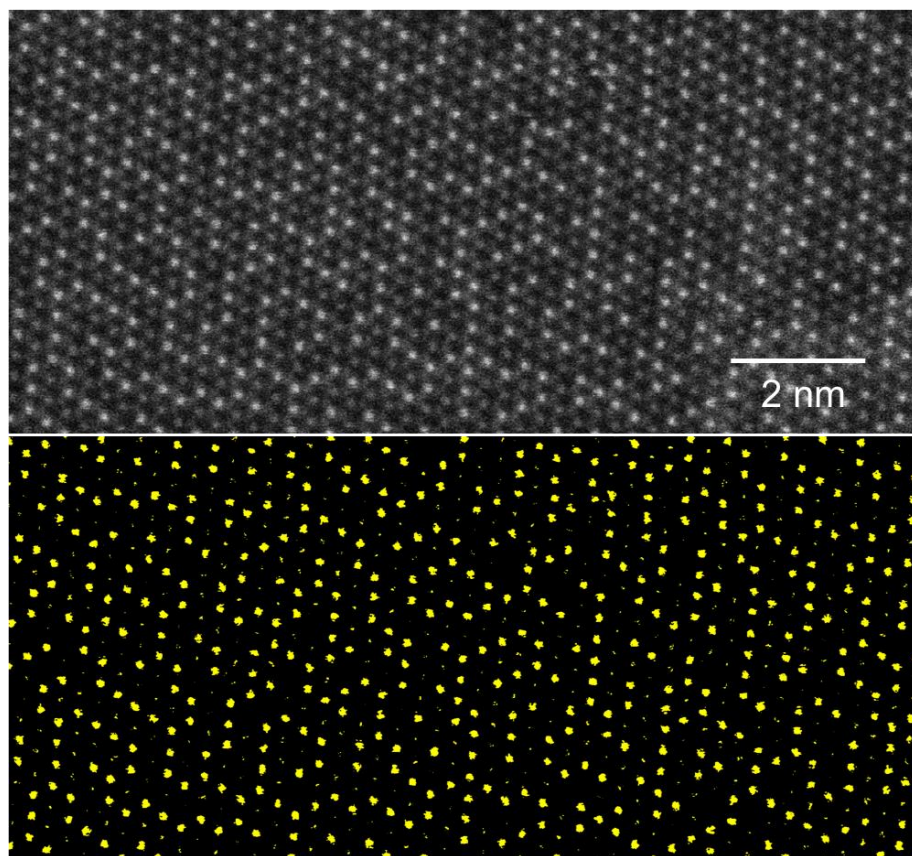


Figure 1. (A) An atomic-resolution ADF-STEM image of the $\text{Re}_{0.5}\text{Nb}_{0.5}\text{S}_2$ monolayer (top) and its filtered version (bottom) showing the meandering stripes formed by Re atoms.

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