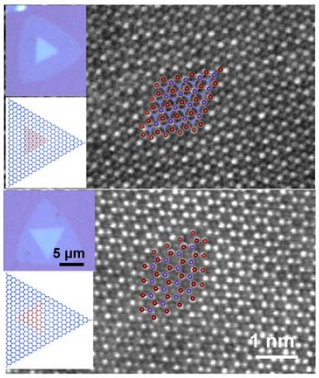
Untangling How Crystals Stack



Atomic images of two layers of GaSe crystals show how they stack for two typical orientations (insets show optical images, and theoretical models).

Scientific Achievement

Untangled how the individual atoms align between two layers of a semiconductor crystal (GaSe) deposited from the vapor phase.

Significance and Impact

Understanding the driving forces that cause crystals to orientate in preferred directions is crucial for guiding synthesis toward achieving new/improved properties.

Research Details

- Bilayer crystals with different twist angles were synthesized through vapor phase deposition directly on microscope grids to enable atomic-scale characterization of the stacking configurations and edge structure by atom-resolved microscopy.
- Calculations revealed that the observed preferred orientations among the bilayer GaSe crystals are minima on the total energy landscape.

Work was performed at the Center for Nanophase Materials Sciences, ORNL

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Narrative Highlight Text:
A coordinated synthesis, characterization, modeling, and theory development enable understanding the numerous metastable configurations orientations that are possible due to interlayer orientations and stacking of two-dimensional (2D) bilayer crystals. Characterizing and controlling the interlayer orientations and stacking order of two-dimensional (2D) bilayer crystals and van der Waals (vdW) heterostructures is crucial to optimize their electrical and optoelectronic properties. Here, through a controllable vapor-phase deposition method we selectively grew bilayer GaSe crystals directly on windowed TEM grids to permit high angle annular dark field electron microscopy in order to determine the atomic configurations corresponding to the preferred 0° or 60° interlayer rotations found experimentally. These rotations were found to correspond with two stackings (AA' and AB-stacking) of the four polymorphs of bulk GaSe. Total energy calculations incorporating hybrid orbital overlap interactions revealed that these two preferred orientations correspond to interlayer stackings with the minimum energy configurations.

Reference
"Revealing the Preferred Interlayer Orientations and Stackings of Two-Dimensional Bilayer Gallium Selenide Crystals"
Xufan Li[1], Leonardo Basile[1,2], Mina Yoon[1], Cheng Ma[1], Alexander A. Puretzky[1], Jaekwang Lee[1], Juan C. Idrobo[1], Miaofang Chi[1], Christopher M. Rouleau[1], David B. Geohegan[1], Kai Xiao[1],*
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