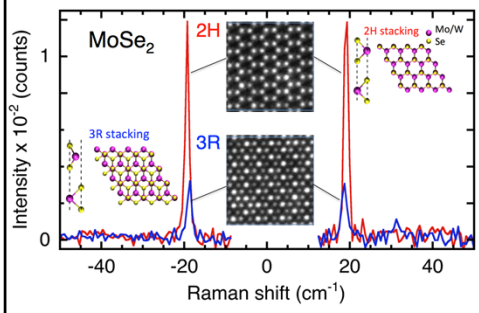
A simple, optical method to determine how two-dimensional layers are stacked in a crystal



Characteristic optical fingerprints of two different stacks of bilayer $MoSe_2$ crystals. Raman spectra of two-layer $MoSe_2$ measured on a TEM grid for 2H and 3R stacking patterns verified by atomic resolution Scanning Transmission Electron Microscopy (STEM.) Stokes (right) and anti-Stokes (left) terahertz Raman spectra of $MoSe_2$ bilayers suspended on a TEM grid. The insets show atomic resolution STEM images of the 2H (top) and 3R (bottom) stacking patterns and their schematics taken at the locations used for the Raman measurements.

Work was performed at the Center for Nanophase Materials Sciences and at Rensselaer Polytechnic Institute

Scientific Achievement

An optical method (Terahertz Raman spectroscopy) was developed for fast remote screening of layer stacking in new two-dimensional (2D) materials, which is orders of magnitude faster than conventional electron microscopy.

Significance and Impact

Fast screening and understanding of the stacking of 2D layers is crucial for accelerating the development of new artificially structured materials for electronic and optoelectronic applications.

Research Details

- High-resolution Raman spectral 'fingerprints' of $MoSe_2$ and WSe_2 bilayers and trilayers were measured at the CNMS in the sub-50 cm^{-1} regime.
- Z-contrast atomic-resolution scanning transmission electron microscopy and first-principle calculations determined the stacking orientations.

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Narrative Highlight Text:
In this work it was discovered that layers of two-dimensional (2D) materials with different atomic registries have characteristic Raman spectra fingerprints in the low frequency spectral range that can be used to characterize stacking patterns of these materials. Stacked monolayers of 2D materials present a new class of hybrid materials with tunable optoelectronic properties determined by their stacking orientation, order, and atomic registry. Fast optical determination of the exact atomic registration between different layers, in few-layer 2D stacks is a key factor for rapid development of these materials and their applications. Using two- and three-layer $MoSe_2$ and WSe_2 crystals synthesized by chemical vapor deposition we show that the generally unexplored low frequency Raman modes (< 50 cm^{-1}) that originate from interlayer vibrations can serve as fingerprints to characterize not only the number of layers, but also their stacking configurations. *Ab initio* calculations and group theory analysis corroborate the experimental assignments and show that the calculated low frequency mode fingerprints are related to the 2D crystal symmetries.

Reference

"Low-Frequency Raman Fingerprints of Two-Dimensional Metal Dichalcogenide Layer Stacking Configuration"

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