Electronic and optical excitations in two-dimensional systems are distinctly sensitive to the presence of a moiré superlattice. We used cryogenic transmission electron microscopy and spectroscopy to simultaneously image the structural reconstruction and associated localization of the lowest-energy intralayer exciton in a rotationally aligned WS$_2$-WSe$_2$ moiré superlattice. In conjunction with optical spectroscopy and ab initio calculations, we determined that the exciton center-of-mass wave function is confined to a radius of approximately 2 nanometers around the highest-energy stacking site in the moiré unit cell. Our results provide direct evidence that atomic reconstructions lead to the strongly confining moiré potentials and that engineering strain at the nanoscale will enable new types of excitonic lattices.

**M**oiré superlattices formed by stacking monolayers of van der Waals crystals are a burgeoning platform for discovery of fundamental physical phenomena. For example, the moiré superlattice of semiconducting transition-metal dichalcogenides (TMDCs) has been predicted to form a topographically protected lattice of bound electron-hole pairs or excitons that can act as a model system for quantum simulations and technologies. Recent optical spectroscopy studies have found signatures of interlayer and intralayer moiré excitons in TMDC heterostructures, investigated exciton diffusion in a superlattice potential, and found evidence for the cooperative nature of moiré excitons.

An important parameter less well studied is the nature of the excitons at the nanoscale level. Nanoscale modulation or confinement of the center of mass of interlayer excitons has been recently probed in momentum space by using ultrafast angle-resolved photoemission electron spectroscopy (ARPES). Similarly, the coupling and decoupling of interlayer excitons and phonons was probed by means of ultralow-frequency tip-enhanced Raman spectroscopy. A clear understanding of the degree of confinement of intralayer excitons as well as a measurement of the stacking at which the localization occurs has remained out of reach because of the diffraction limit of optical probes or the loss of phase information in ARPES. Such real-space visualization of the exciton localization is necessary to address the fundamental question of whether a moiré superlattice can support a periodic array of well-localized quantum excitations.

Electron microscopy can be used to measure structure and electronic transitions at high resolution. Using TMDC heterostructures as model systems, previous studies have measured either their atomic-scale structural reconstruction with annular dark-field scanning transmission electron microscopy (ADF-STEM) or the excitonic signatures by using spatially averaged STEM low-loss electron energy-loss spectroscopy (STEM-EELS). The advent of high-sensitivity direct electron detectors incorporated into EEL spectrometers along with cryogenic holders and monochromation provides an opportunity to simultaneously probe the weak exciton states and the structural reconstruction at the sub-nanometer scale.

We used simultaneous ADF-STEM and low-loss STEM-EELS mapping with hyperspectral analysis to directly image the in-plane structural reconstruction of a rotationally aligned (R-stacked) WS$_2$-WSe$_2$ moiré superlattice and the corresponding oscillator strength of the moiré exciton within the moiré unit cell. We prepared an R-stacked heterostructure of WS$_2$ and WSe$_2$ monolayers encapsulated within ~16 nm and 10-100 nm-thick hexagonal boron nitride (hBN) on opposing sides. The sample was suspended over a 20-nm hole in a custom fabricated silicon TEM grid.

hBN creates a uniform dielectric environment that narrows exciton linewidths and protects the sample from oxidation and beam damage. We verified the twist angle between the layers to be near-zero or R-stacked using second harmonic generation (SHG) and position-averaged convergent beam electron diffraction (fig. S2), along with the emergence of strong emission from a lower-lying, interlayer exciton state in photoluminescence measurements (fig. S3). We then collected hyperspectral images from multiple regions under cryogenic conditions using simultaneous ADF-STEM-EELS with ~100-meV monochromated electron beam focused to less than a nanometer (Fig. 1A). We used simultaneous ADF-STEM-EELS with a ~100-meV monochromated electron beam focused to less than a nanometer (Fig. 1A). We implemented a custom data analysis routine based on unit cell averaging to improve the signal-to-noise ratio in the structural image and final EEL spectra and map (supplementary text and figs. S7 to S17).

We obtained subnanoscale structural information by analyzing ADF-STEM images, in which intensity is proportional to the locally summed atomic number (Z) at each atomic column along the electron beam direction (materials and methods and supplementary text). Quantitative ADF-STEM has been used in the past to study in-plane reconstructions in large-period, lattice-matched moiré systems such as twisted bilayer MoS$_2$, WS$_2$, and MoS$_2$-WS$_2$ (14). To help interpret our experiments, we simulated ADF-STEM images of the theoretically predicted unrelaxed and relaxed moiré superlattice using multislice simulations (supplementary text). The simulation of the unrelaxed moiré heterostructure in which circular regions of higher intensity correspond to the AA, BSe/W, and B/W stacking arrangements is shown in Fig. 1D. The AA region (Z$_{\text{eff}}$ = 142) appears brighter than the BSe/W (Z$_{\text{eff}}$ = 88) and B/W (Z$_{\text{eff}}$ = 54) regions.

Without accounting for a structural reconstruction, all the stacking configurations have roughly the same area (fig. S18). However, the ADF-STEM simulation of the relaxed moiré heterostructure (Fig. 1E) shows clear differences among the sizes of the bright and dark regions. Driven by the stacking energy landscape (Fig. 1C), the effective

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**QUANTUM MATERIALS**

**Hyperspectral imaging of exciton confinement within a moiré unit cell with a subnanometer electron probe**

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Electronic and optical excitations in two-dimensional systems are distinctly sensitive to the presence of a moiré superlattice. We used cryogenic transmission electron microscopy and spectroscopy to simultaneously image the structural reconstruction and associated localization of the lowest-energy intralayer exciton in a rotationally aligned WS$_2$-WSe$_2$ moiré superlattice. In conjunction with optical spectroscopy and ab initio calculations, we determined that the exciton center-of-mass wave function is confined to a radius of approximately 2 nanometers around the highest-energy stacking site in the moiré unit cell. Our results provide direct evidence that atomic reconstructions lead to the strongly confining moiré potentials and that engineering strain at the nanoscale will enable new types of excitonic lattices.

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A fine balance between the strain energy cost of ~1% in the individual layers because of a whole moiré unit cell (with a maximum strain (Fig. 1C) led to a strain redistribution over the present work is distinct from those in et al. Susarla et al. to the relaxed structure, and the BSe/W and BW/S regions configuration is reduced compared with the unrelaxed structure, and the BSe/W and BW/S regions form larger, triangular domains. Experimentally, we imaged an area of the sample that contained many moiré unit cells with a 5-Å probe step size. Although direct atomic resolution imaging was challenging because of the overlap of atoms with very small projected distances (fig. S19), our simulations allowed us to differentiate AA sites from B sites. We averaged the ADF-STEM intensities over 162 moiré unit cells (materials and methods and fig. S7) to obtain the final image shown in Fig. 1F. The experimental ADF-STEM unit cell is consistent across scans (fig. S8), not affected by hBN owing to incommensurate moire with respect to the TMDCs, and in agreement with the multislice simulation of the relaxed heterostructure (Fig. 1E, fig. S18, and table S1), providing the first direct evidence of in-plane structural reconstructions in a WS2-WSe2 moiré superlattice.

The structural transformation we observed (Fig. 1C) led to a strain redistribution over the whole moiré unit cell (with a maximum strain of ~1%) in the individual layers because of a fine balance between the strain energy cost and stacking energy gain (fig. S20) (24). The observed in-plane structural reconstruction in the present work is distinct from those in previous first-principle calculations (22, 25) and ADF-STEM reports (14) in twisted lattice-matched systems, in which the strain was localized to the boundary between the lowest-energy stacking configurations. Usually, when a lattice-mismatched TMDC heterostructure is formed by use of thermodynamics-based growth techniques such as chemical vapor deposition, the heterostructure does not reconstruct (10). The unexpectedly large in-plane structural reconstruction we observed in the R-stacked van der Waals interfaces was recently predicted by theory, and the associated inhomogeneous strain in the individual layers is responsible for the formation of flat electronic bands (24) and spatially modulated excitonic states (9).

We studied the influence of the in-plane structural reconstruction on the moiré excitonic states through ensemble optical spectroscopy and localized low-loss STEM-EELS on the same sample, obtaining higher spectral resolution with the former technique and higher spatial resolution with the latter. The optical image of the R-stacked WS2-WSe2 heterostructure embedded within hBN is shown in Fig. 2A. A representative band-pass filtered ADF-STEM image of the heterostructure is shown in Fig. 2B, displaying a moiré pattern over a 100- by 100-nm region with 0.5-nm resolution. Shown in Fig. 2C are the average EEL spectra acquired from regions of monolayer WSe2 (Fig. 2C, orange), monolayer WS2 (Fig. 2C, blue), and WS2-WSe2 (Fig. 2C, green) compared with optical reflectivity spectra (Fig. 2C, gray). We performed EELS background subtraction using Gauss-Lorentz fitting in the pre- and postedge onset of the exciton peak (26) and smoothed with a Savitzky-Golay algorithm (figs. S9 to S13). STEM-EELS measures the energy lost by incident electrons to electrons in the target atoms excited into empty states, and the EEL signal is proportional to the imaginary part of the dielectric constant of the material (27). Thus, we measured the local probability of creating an exciton, rather than the excitons themselves. The split A and B exciton peak positions arising from spin-orbit coupling of both WS2 (A, 2.10 eV; B, 2.40 eV) and WSe2 (A, 1.75 eV; B, 2.10 eV) monolayers in low-loss EELS match well with optical data, suggesting that the two spectroscopic techniques display a similar oscillator strength for the main exciton peaks. The zoom-in moiré exciton peaks are shown in Fig. 2D. The lowest energy peak (1.65 eV) is slightly red-shifted compared with the WSe2 monolayer A exciton peak (1.75 eV). The optical reflectivity measurement shows the three lowest-energy intralayer moiré exciton peaks (labeled I, II, and III). Although we could only resolve this fine structure of the moiré exciton peaks optically, subsequent broadening of the optical spectra by 100 meV yielded good agreement with the EEL spectra (Fig. 2D and table S2). The largest spectral contribution (58%) in the 1.6 to 1.7 eV range comes from the moiré exciton peak I (fig. S21), which is consistent with the largest oscillator strength predicted with ab initio GW plus Bethe-Salpeter equation (GW-BSE) calculations. The intralayer excitons can decay rapidly into lower-energy interlayer excitons; however, interlayer excitons were not observed in optical reflectance or EELS for this heterostructure system because of their negligible (~1000x smaller) oscillator strength as compared with that of the intralayer excitations (28).
The splitting of the WS\textsubscript{2} A exciton peak into three peaks in the WS\textsubscript{2}-WSe\textsubscript{2} moiré superlattice is predicted (9) to be a result of the strong in-plane structural reconstruction observed in ADF-STEM measurements (Fig. 1), which is unlike other moiré systems in which layer hybridization can also contribute to the moiré-exciton fine structure (7). The in-plane structural reconstruction of the WSe\textsubscript{2} layer leads to flat electronic bands at the valence and conduction band edge. The wave functions corresponding to these flat band states are spatially modulated in the moiré superlattice (fig. S23). In the absence of the reconstruction, the flat bands were not observed (fig. S22), and the electronic states were delocalized in the moiré superlattice. Our first-principles GW-BSE calculations, which fully account for the spatial modulation of the valence and conduction band states owing to the structural reconstruction, reproduced the three peaks observed in the optical reflection contrast spectrum (fig. S24). The GW-BSE calculation for the large-area moiré superlattice was made possible by a recently developed pristine unit cell matrix projection (PUMP) method (9). The wave function that describes a moiré exciton state |S⟩ is a linear combination of valence-to-conduction transitions in the Brillouin zone: $\psi(r_e, r_h) = \sum_{\mathbf{k}=\text{val}} A_{\mathbf{k}}^{\text{val}} \phi_{\mathbf{k}}^{\text{val}}(r_e) \phi_{\mathbf{k}}^{\text{con}}(r_h)$, where $r_e$ and $r_h$ are the electron and hole coordinates, respectively; $A_{\mathbf{k}}^{\text{val}}$ is the exciton envelope function; $\phi_{\mathbf{k}}^{\text{val}}$ and $\phi_{\mathbf{k}}^{\text{con}}$ are the single-particle Bloch states of the valence and conduction bands, respectively; $\mathbf{k}$ is the electron wave vector; and $S$ is an exciton band index. Furthermore, our calculations show that the exciton associated with peak I forms a spatially modulated Wannier exciton with maximum charge density at the AA stacking in the moiré superlattice (Fig. 3A). In the absence of structural reconstructions, we expected only a single excitonic peak in this energy range, with no spatial modulation. We compared the oscillator strength of the moiré exciton, primarily peak I, with the experimental data by applying a 1.3-nm hard mask around the AA, B\textsubscript{WSe}\textsubscript{2}, and B\textsubscript{WS}\textsubscript{2} positions determined from ADF-STEM structural images (supplementary text and figs. S14, S16, and S17). Although the electron beam was spatially confined to less than a nanometer in diameter, low-loss EELS delocalization limited the achievable spatial resolution to about 1 nm (27, 29). We observed a clear difference in the peak intensities derived from the three stacking regions, indicating that the excitonic peak we observed in EELS (principally peak I) has an inhomogeneous oscillator strength in the moiré unit cell, with the maximum intensity at AA stacking regions (Fig. 3B and figs. S16 and S17). This is in very good agreement with the results from our first-principles GW-BSE calculations that constrained the exciton-photon coupling matrix element $\langle S|d\psi^\dagger(r)A(r)\cdot\mathbf{v}\psi(r)|0\rangle$ to a specific spatially resolved region by taking the vector potential $\mathbf{A}(r)$ to have a Gaussian profile, and where $\mathbf{v}$ is the velocity operator, $\psi(r)$ is a field operator, and $|0\rangle$ is the ground-state wave function (Fig. 3C and supplementary text). The oscillator strength of peak I has the largest contribution from around the AA stacking because the photoexcited electron and hole charge density are maximized in this region (Fig. 3A). For a Wannier-type exciton, the oscillator strength distribution is proportional to $\psi^2(r_e=x, r_h=0)$, which corresponds to the wave function of the exciton center-of-mass coordinate, $R = r_e + r_h = x$, and relative coordinate $r = r_e – r_h = 0$—that is, $\psi^2(R = x, r = 0)$.

To determine the spatial extent of the exciton modulation within a moiré unit cell, we generated a hyperspectral unit cell by shifting a 1.3-nm mask to different unit cell locations across a 100- by 100-nm scan, effectively creating a rolling average (supplementary text and figs. S14 and S15). Shown in Fig. 3D is the unit cell averaged and periodically repeated EELS intensity map generated by summing spectral intensity from 1.61 to 1.71 eV (figs. S15, S25, and S26). We observed that the intarlayer exciton was constrained to the AA sites, which formed a triangular arrangement in the moiré pattern. Furthermore, the exciton intensity rapidly reduced beyond ~2 nm from the AA site, which is in good agreement with the corresponding GW-BSE-calculated oscillator strength map of the moiré exciton peak I (Fig. 3E).

The effect of structural reconstruction in moiré lattices has been predicted to generate strong electronic and excitonic confinement (9, 24). By taking advantage of the spatial and spectral resolution in cryogenic correlated ADF-STEM and STEM-EELS techniques, we directly captured the subnanometer scale in-plane structural reconstruction and the corresponding
exciton localization without the limitations of diffractive optics. The structural reconstruction was not observed in thermodynamically grown heterostructures (20). We found that the structural reconstruction was robust over a submicrometer area of the sample, despite potential sources of disorder introduced by the multilayer mechanical stacking process. The in-plane structural reconstruction led to spatially modulated electronic bands (24–30) in this system, which was corroborated by the recent observations of strongly correlated Wigner crystal and Mott insulating phases (31, 32).

The exciton wave function as a function of the center-of-mass coordinate (with small electron-hole relative coordinate) was localized to the AA site within a radius of ~2 nm. This suggests that the moiré potential was preserved over a large area of the sample and could support the formation of a triangular lattice of excitons. The diminished exciton intensity at the Bernal stacking regions (Fig. 3, D and E) is in agreement with the unexpected absence of trion formation upon hole doping of the same heterostructure system (supplementary text) (5, 9). Unit cell averaging, which sampled across hundreds of square nanometers, greatly improved the signal-to-noise ratio of the EELS intensity map.

We discovered that the strong atomic relaxations within each moiré cell leads to confinement of excitons at a particular stacking site. This opens up the possibility of nanoscopic engineering of bosonic lattices in moiré heterostructures through external strain, twist-angle, and number of layers.

FIG. 3. Subnanoscale spatial mapping of moiré excitons: (A) Calculated electron charge density contributing to the peak I exciton, \( \nu_0(r_x, r_y) \), for a fixed position of the hole, \( r_h \), in the moiré superlattice. The evolution of the electron charge density was plotted by moving the hole position across the longer diagonal of the superlattice, showing the spatially modulated character of the peak I exciton. (B and C) Comparison of spatially resolved experimental EELS and GW-BSE theory for different stacking configurations AA, B̅Se/W, and B̅W/S. (D) Experimental unit cell averaged and tiled EELS map, obtained by a rolling average with a 1.3-nm radius at different unit cell locations. Shown is the modulation of the EELS intensity, which is primarily composed of peak I at 1.65 eV. (E) Theoretically calculated oscillator strength distribution of the peak I exciton, with similar radial blurring as the experiment.

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**SUPPLEMENTARY MATERIALS**

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Materials and Methods
Supplementary Text
Figs. S1 to S26
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