

Observation of Wannier-Rydberg and Charge-Transfer Hybrid Moiré Excitons under Pressure

Jing Song^{1,*}, Yifan Wang^{2,3,*}, Xuan Zhao^{1,4,*}, Yuxuan Song^{1,4,*}, Song Liu⁵, Chen Hu^{2,3,†} and Yang Xu^{1,4,‡}

¹Beijing National Laboratory for Condensed Matter Physics,

Institute of Physics, Chinese Academy of Sciences, Beijing, 100190, China

²Department of Physics, The Chinese University of Hong Kong, Hong Kong 999077, China

³State Key Laboratory of Quantum Information Technologies and Materials,

The Chinese University of Hong Kong, Hong Kong 999077, China

⁴School of Physical Sciences, University of Chinese Academy of Sciences, Beijing 100049, China

⁵Institute of Microelectronics, Chinese Academy of Sciences, Beijing 100029, China



(Received 26 March 2026; revised 20 July 2026; accepted 28 August 2026; published 6 October 2026)

Moiré superlattices in van der Waals heterostructures, arising from lattice mismatch or twist angle, provide a powerful platform for engineering correlated electronic and excitonic states. Moiré excitons form when electrons and holes are individually modulated by the moiré potential yet remain bound by Coulomb interaction, resulting in exciton wave functions with a highly nontrivial and rich real-space structure. However, achieving *in situ* and dynamic control over the interplay between excitonic Coulomb interaction and the moiré potential remains a central challenge. Here, we demonstrate that hydrostatic pressure provides a continuous and effective means of enhancing the moiré potential and tuning moiré excitons in WSe₂/WS₂ moiré heterobilayers. As pressure increases, intralayer moiré excitons exhibit systematic redshifts as the moiré potential deepens. Crucially, above ~3 GPa the system enters a strong-moiré regime, where a new higher-energy intralayer moiré exciton emerges that is not accessible otherwise. Combining dual-gate optical spectroscopy with Bethe-Salpeter equation calculations, we reveal that this emerging moiré exciton state arises from strong pressure-induced hybridization between two distinct exciton series: Wannier-Rydberg and charge-transfer excitons. Enhanced exciton-electron and exciton-exciton interactions observed in doping-dependent photoluminescence further evidence the strengthened moiré potential under pressure. Our work establishes a pioneering paradigm of moiré excitonic wave function engineering, enabling the on-demand creation of tailored excitonic species in moiré superlattices.

DOI: 10.1103/PhysRevX.16.041007

Subject Areas: Condensed Matter Physics,
Materials Science, Nanophysics

Moiré superlattices can be formed by stacking two-dimensional (2D) crystals with a small twist angle or lattice mismatch. Arising from the interference between the periodic lattices of the two layers, the moiré pattern introduces a long-wavelength potential whose characteristic length scale is much larger than the atomic lattice constant of the individual layers. This moiré potential can strongly reshape the electronic band structure and give rise to a variety of exotic quantum phases, including unconventional superconductivity, correlated insulating states, topological states, generalized Wigner crystals, ferromagnetism,

and Kondo lattice states [1–10]. Beyond ground-state phenomena, the interaction between 2D moiré superlattices and light generates a rich landscape of photoexcited states. Upon optical excitation, electrons and holes are created and bound via Coulomb attraction to form excitons that are correlated two-particle quasiparticles. Owing to reduced dielectric screening and quantum confinement, excitons in 2D semiconductors typically form a Wannier-Rydberg (WR) series, dominated by strong electron-hole Coulomb interactions [11]. Intriguingly, in transition-metal dichalcogenide (TMD) moiré superlattices, the presence of moiré potential introduces an additional degree of freedom that can strongly interplay with the excitonic Coulomb interaction, giving rise to moiré excitons.

Such moiré excitons arise when the intrinsic fermionic constituents, i.e., the electron and hole, are individually modulated by the moiré potential yet remain bound by the Coulomb interaction [Fig. 1(b)], resulting in exciton wave functions with highly nontrivial and rich real-space structures. In recent years, moiré excitons in TMD superlattices have attracted intense interest. Early studies demonstrated

*These authors contributed equally to this work.

†Contact author: chenhu@cuhk.edu.hk

‡Contact author: yang.xu@iphy.ac.cn

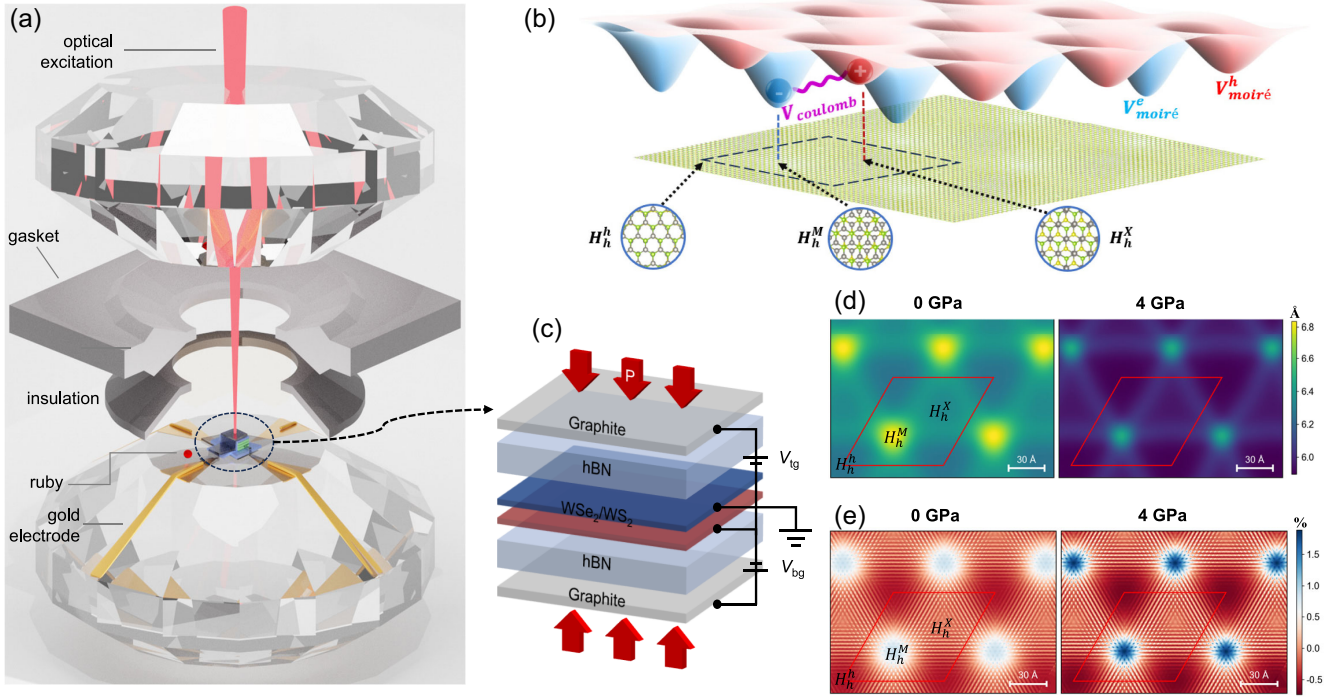


FIG. 1. H-stacked WSe₂/WS₂ moiré heterostructure under pressure. (a) Cartoon of high-pressure and dual-gate tunable spectroscopic measurements of moiré excitons in an H-stacked WSe₂/WS₂ heterostructure. An insulating layer is inserted between the metallic gasket and the diamond culet to prevent electrical shorting. (b) Schematic illustration of the intralayer moiré excitons arising from the interplay between the moiré potential ($V_{\text{moiré}}^e$ for electron and $V_{\text{moiré}}^h$ for hole) and the excitonic Coulomb interaction (V_{Coulomb}^e). The black dashed outline denotes the moiré supercell with a lattice constant of 8.3 nm, and the positions of three high-symmetry local stacking sites (H_h^h , H_h^M , and H_h^X) are labeled. H_h^h —W (Se) atoms of WSe₂ are on top of S(W) atoms of WS₂; H_h^M —W (Se) atoms of WSe₂ are on the hollow sites (top of S atoms) of WS₂; and H_h^X —Se (W) atoms of WSe₂ are on the hollow sites (top of W atoms) of WS₂. (c) Schematic of the WSe₂/WS₂ device with a dual gate configuration under pressure. (d),(e) Theoretical spatial distribution of (d) the interlayer distance and (e) the in-plane strain in the WSe₂ layer at 0 and 4 GPa.

moiré-trapped interlayer charge-transfer (CT) excitons [12,13], in which electrons and holes reside in different layers and are localized at the minima of the moiré potential. Such quantum-dot-like excitonic states were revealed through photoluminescence measurements [13]. In contrast, intralayer CT excitons, where electrons and holes occupy different regions within the same layer, are much rarer because their formation requires the moiré potential to counterbalance the intrinsic Coulomb attraction [Fig. 1(b)]. Recently, this new type of intralayer moiré CT exciton has been identified both experimentally and theoretically in WSe₂/WS₂ moiré superlattices, where its properties are highly sensitive to local strain and modulation of the moiré landscape [14]. Further theoretical work has predicted shift-current vortex crystals and associated all-optical control of magnetism induced by the intralayer moiré CT exciton [15]. Generally, CT excitons possess long lifetimes and large dipole moments, enabling a range of rich phenomena, including strong optical nonlinearities, Bose-Einstein condensation, and exciton superfluidity [16–19]. Despite these advances, achieving *in situ* and dynamic control over the interplay between excitonic Coulomb interaction and moiré

potential, thereby engineering the moiré excitonic wavefunction is highly desirable but remains a central challenge in moiré excitonic physics.

In this work, we demonstrate that hydrostatic pressure offers a powerful yet non-destructive control parameter for dynamically manipulating the interplay between the exciton's internal orbital character governed by the excitonic Coulomb interaction and its external spatial configuration governed by the moiré potential. By continuously tuning the interlayer coupling, structural reconstruction, and moiré potential strength, pressure drives a systematic evolution of moiré excitonic states. We apply hydrostatic pressure up to ~ 5 GPa to an H-stacked (60° -twisted) WSe₂/WS₂ moiré superlattice using a diamond anvil cell (DAC). Based on doping-dependent optical reflectance spectroscopy, we observe systematic pressure-induced energy shifts of the moiré excitonic states. Most strikingly, upon increasing pressure beyond ~ 3 GPa, the system is driven into a strong-moiré regime, where a new higher-energy moiré exciton state emerges that is not accessible otherwise. Our excitonic calculations based on the Bethe-Salpeter equation (BSE) reveal that this new moiré exciton originates from the pressure-enhanced moiré potential, which drives strong

hybridization between the WR exciton state and the CT exciton state, thereby inheriting the spatial characteristics of both. The marked strengthening of the moiré potential is further evidenced by enhanced electron-exciton and exciton-exciton Coulomb repulsions of the interlayer exciton observed in doping and electric field dependent photoluminescence spectroscopy, respectively. This direct observation of a pressure-induced exciton hybridization pathway underscores the unique potential of hydrostatic pressure for engineering the moiré potential and correlated excitonic phenomena in moiré superlattices.

Figure 1(a) presents a schematic of the DAC and the associated experimental setup [20]. Optical access is provided through the transparent top diamond window. Pressure (P) is applied by compressing the DAC chamber filled with liquid pressure-transmitting medium (PTM) and is calibrated using the energy shift of the ruby R_1 fluorescence line [21]. Dual-gated H-stacked WSe_2/WS_2 heterobilayer devices (see Supplemental Material [22], Fig. S1 for the device image) are fabricated and transferred onto the diamond culet, which is pre-patterned with Ti/Au electrodes for electrical access. The twisted angle ($\sim 60^\circ$) between the WSe_2 and WS_2 layers is achieved using polarization-dependent second-harmonic-generation (SHG) measurements [Supplemental Material [22], Fig. S1(b)] [44,45]. The WSe_2/WS_2 heterobilayers are fully encapsulated between few-layer hBN and graphite, which serve as dielectrics and gate electrodes on both sides, enabling independent control of the vertical electric field (E_z) and carrier doping density [as illustrated in Fig. 1(c)]. The encapsulation further preserves clean interfaces and maintains high optical quality under applied pressure. The exciton dynamics are primarily studied at ~ 8 K using optical reflectance and photoluminescence (PL) micro-spectroscopy with a probing spot of approximately $2\ \mu\text{m}$.

The H-stacked WSe_2/WS_2 heterostructure forms a moiré superlattice with a lattice constant of ~ 8.3 nm, arising from the $\sim 4\%$ lattice mismatch between WS_2 and WSe_2 [Fig. 1(b)]. Owing to the different energetics of local stacking configurations within the moiré pattern, the superlattice undergoes structural reconstruction to minimize the total energy by increasing the areas of the lower-energy H_h^h and H_h^x stacking regions while reducing the higher-energy H_h^M regions. Previous experimental and theoretical studies have shown that such structural reconstruction plays a key role in the electronic behavior of moiré superlattices [14,15,46,47]. Figures 1(d) and 1(e) present the spatial distribution of interlayer distance and intralayer strain in the WSe_2 layer across the reconstructed superlattice at 0 and 4 GPa, respectively, obtained from large-scale atomistic force-field simulations (see Methods in the Supplemental Material [22] for details). Earlier studies have shown that pressure can modulate the moiré structure without altering the intrinsic moiré periodicity [48–53]. The first observation is that pressure reduces the

interlayer distance across all stacking regions [Fig. 1(d)], as a direct consequence of the two layers being pushed closer together. More importantly, as shown in Fig. 1(e), pressure enhances the in-plane structural reconstruction, leading to a pronounced increase in the strain field, particularly within the H_h^M stacking regions. The calculated strain is strongly localized within the moiré unit cell and should not be interpreted as a uniform macroscopic strain. Optical micrographs show that the device remains intact during pressurization, without severe wrinkling or delamination (Supplemental Material [22], Fig. S9). This strengthened intralayer reconstruction, together with the reduced interlayer spacing, significantly enhances the interlayer coupling and moiré potential. As discussed below, these changes strongly modulate the electronic and excitonic properties of the moiré superlattice. Meanwhile, we note that low-frequency Raman spectroscopy of interlayer shear and breathing modes would provide another probe of pressure-enhanced interlayer coupling [54–59], which represents an interesting direction for future studies.

To systematically elucidate the pressure-driven evolution of moiré excitonic states, we combine reflectance-contrast spectroscopy measurements of the WSe_2/WS_2 superlattice with theoretical calculations of the exciton spectrum and optical absorbance across a pressure range of 0–5 GPa. Because the WSe_2 layer has a smaller band gap, the low-energy reflectance contrast spectrum is dominated by WSe_2 intralayer excitons ($< \sim 1.8$ eV, see Fig. 2), which are well separated from the higher-energy WS_2 intralayer excitons [45]. At the lowest pressure ($P = 0.2$ GPa), three distinct intralayer moiré exciton states (X_1 – X_3 , at energies of 1.683, 1.735, and 1.770 eV, respectively) at charge neutrality ($V_g = 0$ V) are observed experimentally [Fig. 2(a), left panel].

To reveal the microscopic nature of these moiré excitons, we perform theoretical calculations based on the BSE which is the state-of-the-art approach for accurately describing two-body excitonic states. Owing to the large size of the moiré supercells, full *ab initio* BSE calculations are computationally challenging [14,15]. We therefore employ a continuum-model-based BSE framework, in which the moiré potentials for electrons and holes are separately parametrized by using large-scale density functional theory (DFT) calculations of fully reconstructed moiré superlattices (see Methods in the Supplemental Material [22]). As shown in Fig. 2(c), the computed absorbance spectra capture the three moiré excitonic resonance peaks in good agreement with experiments [Fig. 2(a)]. The quantitative discrepancy between the calculated and experimentally measured moiré-exciton energies may arise from the complex dielectric screening associated with the spatially inhomogeneous moiré superlattice, substrate, and experimental device environment. Real-space analysis of the computed exciton wave functions further reveals their distinct spatial characters: the lowest-energy state X_1 corresponds to a typical modulated WR exciton with

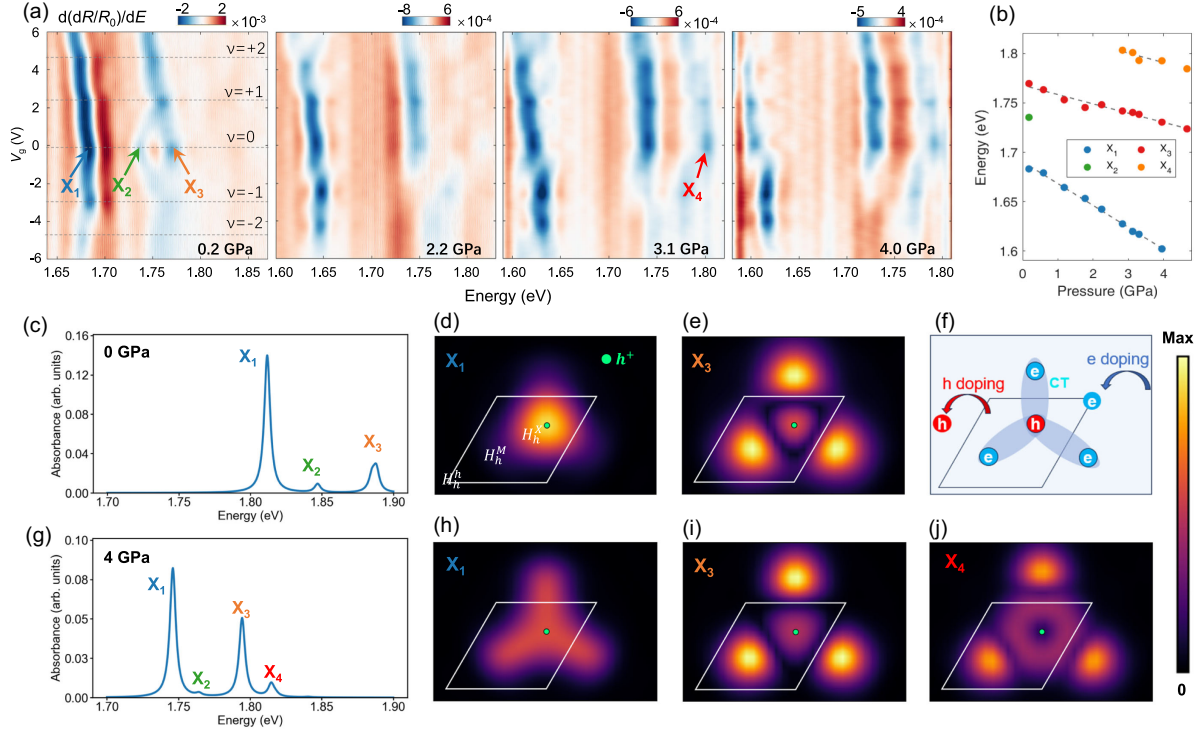


FIG. 2. Pressure-driven evolution of moiré excitons in an H-stacked WSe_2/WS_2 heterostructure. (a) Doping-dependent reflectance contrast spectra at 4 representative pressures. At low pressure (<0.2 GPa), three dominant peaks, X_1 , X_2 , and X_3 , are identified; notably, at high pressure (>3 GPa) a new high-energy exciton state (X_4) emerges. For data at additional pressures, see Fig. S7 in the Supplemental Material [22]. (b) A sum of excitonic energies as functions of pressure. (c),(g) Computed absorbance spectra of the moiré superlattices at 0 (c) and 4 GPa (g). (d),(e),(h),(i),(j) Real-space distributions of the electron density of the computed excitons, $|\Psi(r_e, r_h)|^2$, forming peaks X_1 and X_3 at 0 GPa [(d) and (e)], and peaks X_1 , X_3 , and X_4 at 4 GPa (h),(i), and (j). In the plots, the hole position r_h (green dot) is fixed at the H_h^X site, which yields the most dominant electron density distribution (see Supplemental Material [22], Fig. S5). At 0 GPa, X_1 and X_3 exhibit distinct features of a modulated Wannier-Rydberg exciton (electron and hole localized at the same stacking site) and an intralayer charge-transfer exciton (electron and hole separated across different stacking sites), respectively. At 4 GPa, stronger CT character develops: the electron density extends towards the H_h^M stacking site for X_1 (h) and increases at the H_h^M stacking site for X_3 (i). Most notably, the emerging peak X_4 (j) corresponds to a new hybrid exciton type, combining characteristics of the WR $3d$ exciton state (ringlike) and the CT exciton state. (f) Schematic illustration of the doping response of the CT exciton in the moiré superlattices. Upon electron (hole) doping, the doped electrons (holes) occupy the H_h^h (H_h^X) sites in the WS_2 (WSe_2) layer, respectively.

$1s$ character, in which the electron is tightly bound to the hole at the same H_h^X site [Fig. 2(d)]; by contrast, X_3 is an intralayer CT exciton in which the electron and hole densities are spatially separated by ~ 4.8 nm, residing at the H_h^M and H_h^X stacking regions, respectively [Fig. 2(e)]. The X_2 exciton exhibits a mixed character [14], and the details can be seen in the Supplemental Material [22] [Fig. S5(b)].

These distinct spatial characters of the moiré WR exciton (X_1) and the CT exciton (X_3) can be further revealed by their markedly different doping dependences in the experimental optical spectra. Upon electron (hole) doping, the doped electrons (holes) predominantly occupy the H_h^h (H_h^X) sites in the WS_2 (WSe_2) layer, respectively [Fig. 2(f)] [60]. The doped carriers interact with the moiré exciton dipole through electrostatic Coulomb coupling, resulting in doping-induced energy shifts of the optical resonances. The moiré WR exciton X_1 , in which electrons and holes spatially overlap, possesses a negligible excitonic dipole

moment and therefore exhibits only a weak energy shift under carrier doping. In contrast, the moiré CT exciton X_3 carries a pronounced dipole moment arising from the large electron-hole separation, making its energy more sensitive to doping. Notably, the X_1 resonance recovers its oscillator strength when the system is doped to integer filling factors $\nu = \pm 1$, consistent with the formation of Mott insulating states, as previously reported [Fig. 2(a)] [45,61].

With increasing pressure, the intralayer moiré excitons exhibit systematic and distinct evolution. First, the mixed exciton state X_2 quickly loses its oscillator strength and becomes barely visible above ~ 0.5 GPa. Interestingly, the energies of X_1 and X_3 reduce linearly up to the highest applied pressure, with rates of ~ 22 and ~ 9 meV/GPa, respectively [Fig. 2(b)]. Such pressure-induced redshifts were also observed in previous reports based on fiber-tip-induced local deformation up to ~ 1.4 GPa [62]. This behavior originates mainly from the pressure-induced reduction of the moiré band gap in the WSe_2 layer

(Supplemental Material [22], Fig. S2). The slower redshift of X_3 than X_1 may reflect a stronger pressure dependence of the binding energy of the spatially extended CT-like exciton. Pressure-induced changes in screening (e.g., enhanced in the H_h^M region) and wave function extent can reduce the X_3 binding energy and thereby partially compensate the redshift of its single-body transition energy [63]. A quantitative analysis would require a spatially resolved calculation of the pressure-dependent dielectric screening of the moiré superlattice, which is beyond the scope of this work. In addition, the gate voltages (V_g) corresponding to integer fillings, extracted from the doping-dependent reflectance contrast (RC) spectra, exhibit an overall decreasing trend with increasing pressure [Supplemental Material [22], Fig. S3(a)]. This trend arises from the reduced interlayer spacing and the corresponding increase in the geometric capacitance of the hBN dielectrics under out-of-plane compression (Supplemental Material [22], Figs. S3(b)–S3(d)). In contrast, the 1s exciton peak energies in monolayer WSe₂ and WS₂ remain nearly pressure-independent in the RC and PL spectra (Supplemental Material [22], Fig. S4), consistent with a previous report of a negligible 1s-exciton shift in hBN-encapsulated monolayer WSe₂ under local compressive deformation [62]. This likely arises from the much weaker response of TMDs to out-of-plane strain than to in-plane strain [64]. Because the samples encapsulated by hBN adhere to the diamond surface, the applied stress deviates from isotropic hydrostatic pressure, leading to a near cancellation between the band gap increase from out-of-plane stress and the decrease from in-plane stress.

Most notably, when pressure exceeds ~ 3 GPa, a new high-energy exciton state X_4 emerges in our experimental reflectance spectra. Its energy redshifts with increasing pressure at a rate of ~ 9 meV/GPa, nearly identical to that of X_3 . This new moiré exciton state is also captured by our BSE calculations at 4.0 GPa [Fig. 2(g)]. Analysis of the computed spatial distribution of the exciton wave function reveals that X_4 possesses a complex hybridized character: its hole component is primarily localized at the H_h^X site, while its electron component is distributed between the H_h^X site and adjacent H_h^M sites. This spatial structure indicates a new type of WR-CT hybrid moiré exciton complex, combining the ringlike orbital character of a WR $3d$ exciton with the electron-hole separation characteristic of a CT exciton [Fig. 2(j)]. Experimental doping-dependent optical spectra measurements show that X_4 exhibits pronounced sensitivity to carrier doping [right panel of Fig. 2(a)], reflecting its large excitonic dipole arising from the highly delocalized and registry-separated charge distribution.

In the following, we focus our analysis on the underlying mechanism of how pressure drives the emergence of the previously unidentified WR-CT hybrid moiré exciton X_4 . As shown in Figs. 1(d) and 1(e), pressure-induced structural reconstruction introduces two major effects. First, the reduced interlayer spacing enhances interlayer coupling between the two layers. Second, the strengthened lattice reconstruction further redistributes local stacking domains

and generates enhanced local strain fields. Our large-scale DFT calculations show that the combination of these effects markedly amplifies the moiré potential under high pressure as illustrated in Figs. 3(a) and 3(b). On the other hand, we note that the widely used local stacking approximation (LSA), in which moiré potential is fitted from calculations of pristine bilayers (small unit cells) at different local regions [65], fails to capture the quantitative evolution of the moiré potential under pressure [Fig. 3(b)], highlighting the critical role of intralayer structural reconstruction in accurately describing pressure-induced moiré potential modulation.

A key feature of the system is that the moiré potentials for electrons and holes have different spatial distributions. The moiré potential minimum for holes is located at the H_h^X stacking site, while that for electrons lies at the H_h^M site [Fig. 3(a)]. This spatial separation between the energetically favorable locations of electrons and holes serves as the driving force for the formation of intralayer CT excitons [Fig. 1(b)]. The pressure-induced evolution of moiré excitons can therefore be understood through the dynamical interplay between the excitonic Coulomb potential V_{Coulomb} and the moiré potential $V_{\text{moiré}}$. For explicit illustration but without loss of generality, as shown in Figs. 3(c)–3(e), we fix the hole position and analyze the corresponding electron density distribution of the excitons. The Coulomb potential V_{Coulomb} , which is inversely proportional to the electron-hole separation $r_e - r_h$, favors Wannier-like excitons, where electrons and holes localize at the same positions to maximize their binding energy. In contrast, the moiré potential $V_{\text{moiré}}$, whose minima occur at different stacking sites for electrons and holes, tends to spatially separate them, thereby favoring CT character.

The pressure-driven dynamical interplay between V_{Coulomb} and $V_{\text{moiré}}$ organizes the moiré excitonic behaviors into three distinct regimes. In the no-moiré regime (absence of moiré potential), where the exciton is governed solely by Coulomb binding V_{Coulomb} [Fig. 3(c)], the exciton spectrum reduces to the standard WR series ($1s, 2p, 2s, 3d, \dots$) with different spatial extents of the wave functions [66]. States within the same principal shell but with higher orbital angular momentum lie at lower energies owing to strong spatially dependent dielectric screening [66]. In the moderate-moiré regime (upon forming moiré heterobilayers but without external pressure), the moiré modulation becomes relevant [Fig. 3(d)]. Electron and hole moiré potential minima occur at different stacking sites and give rise to moiré CT excitons. In this regime, the WR and CT exciton states coexist with little hybridization [corresponding to Figs. 2(d) and 2(e)]. The large electron-hole separation in CT excitons reduces their Coulombic attraction, yielding smaller binding energies and, consequently, higher exciton energies compared with the lowest WR exciton [Fig. 2(c)].

As the external pressure increases, the system enters a strong-moiré regime in which the enhanced moiré potential $V_{\text{moiré}}$ strengthens the CT character of the

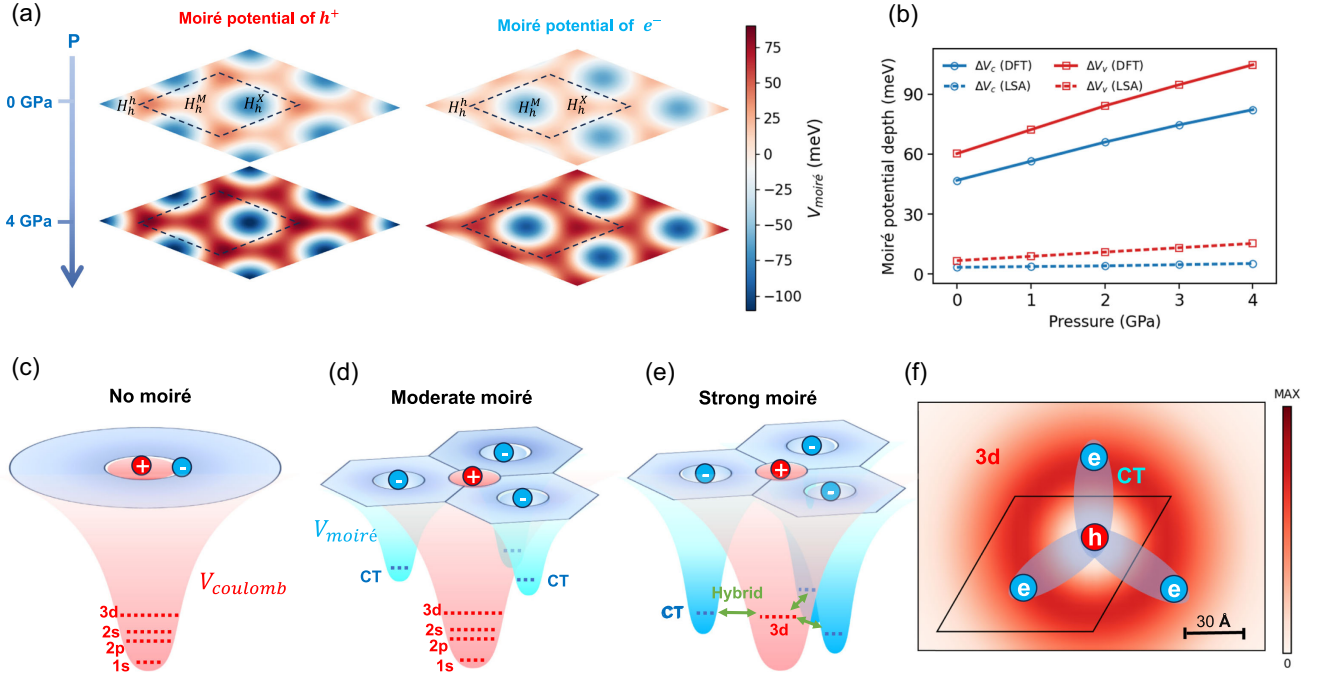


FIG. 3. Calculated moiré potential distribution and moiré exciton evolution under pressure. (a) Theoretical spatial distributions of the moiré potential $V_{moiré}(r)$ for holes (h^+ , left panel) and electrons (e^- , right panel) at 0 and 4 GPa, obtained from large-scale DFT calculations. (b) Pressure dependence of the moiré potential depth for electrons (V_c) and holes (V_v), obtained from explicit large-scale DFT calculations (solid lines) and local-stacking approximations [LSA]; dashed lines]. (c) Schematic illustration of the WR series ($1s, 2p, 2s, 3d, \dots$) [66], governed solely by the electron-hole Coulomb potential (red cone), in pristine monolayer WSe₂ without a moiré potential. (d), (e) Schematic illustration of the pressure-induced evolution of the excitonic states in the WSe₂/WS₂ moiré superlattice. For clarity, we fix the hole at its moiré-potential minimum and illustrate only the effective potential experienced by the electron. (d) At zero applied pressure, the system lies in a moderate-moiré regime. The moiré potential (blue cone) favors the localization of the electron at the moiré potential minimum, thereby leading formation of CT excitons in which the electron and hole remain bound by the Coulomb potential. (e) At high pressure, the system enters a strong-moiré regime. The deepened moiré potential brings the CT exciton into energetic proximity with the WR 3d state, resulting in strong hybridization and the emergence of a CT-3d hybrid excitonic state. (f) Real-space distribution of the electron density of the pristine 3d exciton state (without moiré) with the hole fixed at the H_h^X stacking site. The location of the CT exciton is illustrated. The radius of the 3d exciton is close to the electron-hole separation of the CT exciton, leading to strong spatial overlap and thus pronounced coupling between them.

excitons [Figs. 2(h)–2(j)]. The CT exciton states become progressively stabilized, with their energies decreasing toward those of the WR excitons, leading to coupling between them. Remarkably, strong hybridization occurs between the CT exciton state and the WR 3d state due to their resonant energies and significant electron wave function overlaps [Fig. 3(f)]. This gives rise to the emergence of a new exciton X_4 , which simultaneously exhibits the ringlike orbital character of a WR 3d exciton and the electron-hole separation characteristic of a CT exciton [as shown in Fig. 2(j)]. This behavior is further revealed by the calculated optical absorbance spectra under fine pressure tuning (Supplemental Material [22], Fig. S8). The schematic in Figs. 3(c)–3(e) summarizes the crossover driven by the interplay between $V_{Coulomb}$ and $V_{moiré}$, highlighting how pressure continuously tunes the system from a moderate moiré regime to a strong moiré regime where the WR-CT hybrid moiré exciton X_4 emerges. A similar pressure-induced hybrid moiré exciton also emerges in

the WS₂ layer (Supplemental Material [22], Fig. S16), although its high-energy optical resonance is difficult to resolve experimentally.

The enhancement of the moiré potential under pressure is further corroborated by the gate-dependent spectral evolution of interlayer excitons (Fig. 4). Doping-dependent PL spectra [Figs. 4(a)–4(c)] reveal a pressure-induced strengthening of the electron-exciton interaction U_{e-ex} , defined as the energy cost of adding an interlayer exciton to a moiré site already occupied by one electron. As summarized in Fig. 4(c), U_{e-ex} increases from ~ 25 to ~ 70 meV with increasing pressure. In addition, electric-field-dependent PL spectra [Figs. 4(c) and 4(d)] demonstrate an enhanced exciton-exciton interaction U_{ex-ex} under pressure. This enhancement is manifested by the increasing energy separation between the singly occupied state (IX_1) and the doubly occupied state (IX_2) at the charge neutrality, as illustrated in Fig. 4(e). The higher-energy branch IX_2 corresponds to a repulsively interacting

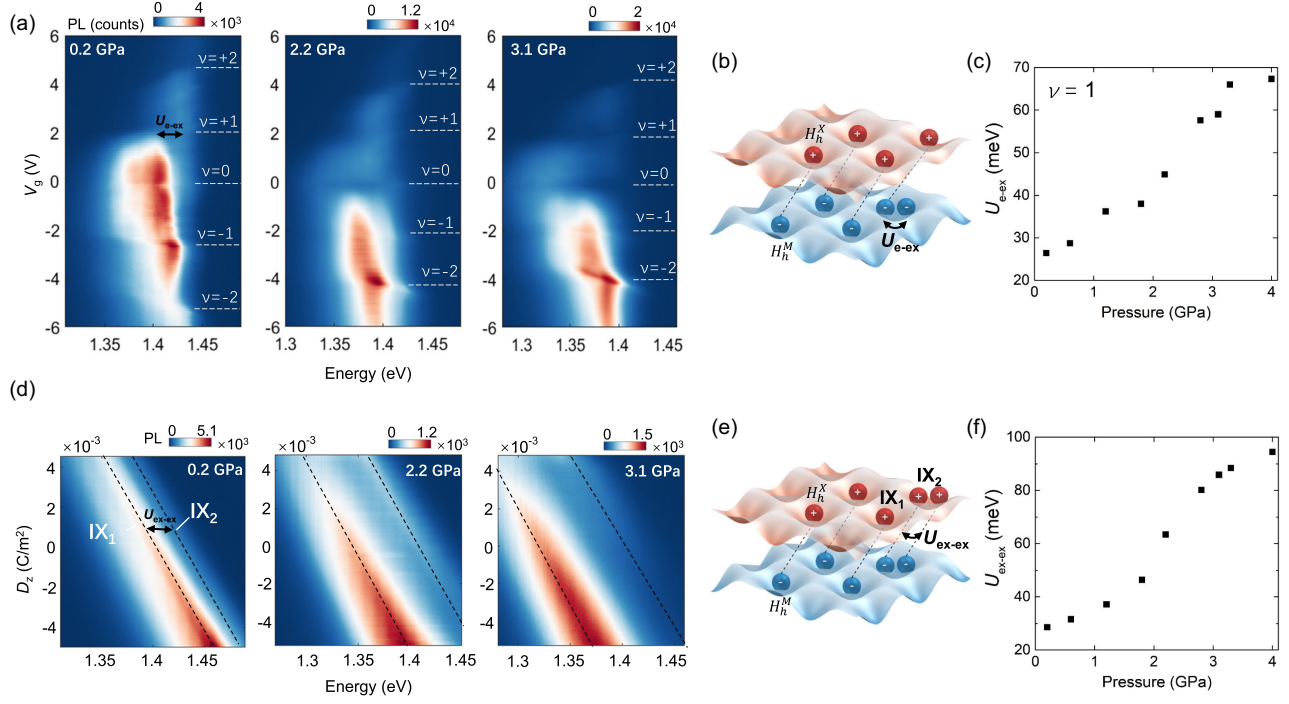


FIG. 4. Pressure tuning of interlayer exciton-electron and exciton-exciton interactions. (a) Doping-dependent (with $V_g = V_{tg} = V_{bg}$) PL spectra at three representative pressures. (b) Schematic illustration of exciton–electron interactions. (c) Energy discontinuity at filling factor $\nu = 1$, corresponding to the interaction energy (U_{e-ex}) between an electron and an exciton within the same moiré supercell. (d) PL spectra versus vertical displacement field (D_z) at three representative pressures, where gate voltages are applied antisymmetrically ($V_{tg} = -V_{bg}$) to maintain charge neutrality of the sample. (e) Schematic illustration of exciton-exciton interactions (U_{ex-ex}). (f) Extracted energy separation between IX₁ and IX₂, reflecting exciton-exciton repulsion within the same moiré supercell, under different pressures. For data at additional pressures, see Fig. S12–S14 in the Supplemental Material [22].

two-interlayer-exciton configuration in a localized moiré site [67–69]. This is analogous to an on-site Hubbard-type interaction energy, in the sense that double occupancy of a localized site is shifted upward by a repulsive interaction. The role of the moiré potential is to localize the excitons and make this repulsively interacting two-particle configuration spectroscopically accessible. In this case, after one interlayer exciton already occupies a moiré potential minimum, adding a second interlayer exciton to the same moiré site costs a positive interaction energy U_{ex-ex} . The extracted U_{ex-ex} , summarized in Fig. 4(f), increases from ~ 30 to ~ 95 meV. Stronger confinement generally reduces the in-plane extent of the localized exciton wave function and thereby enhances the on-site electron-exciton and exciton-exciton interactions. The observed increase of U_{e-ex} and U_{ex-ex} with pressure is therefore consistent with pressure-enhanced moiré confinement and stronger interaction effects within a moiré cell.

In summary, we have markedly enhanced the moiré potential in an H-stacked WSe₂/WS₂ moiré superlattice by applying hydrostatic pressure up to ~ 5 GPa using a diamond anvil cell. Doping-dependent reflectance spectroscopy reveals the concurrent emergence of a new, higher-energy moiré exciton peak. BSE calculations confirm that this new moiré exciton resonance originates from

the strong hybridization between WR 3d and CT excitons under a pressure-enhanced moiré potential. The strengthened moiré potential is further manifested in the enhanced electron-exciton and exciton-exciton Coulomb interactions of the interlayer exciton, as evidenced by gate-dependent PL spectroscopy. This establishes pressure as a powerful tuning knob governing the interplay between intrinsic excitonic Coulomb attraction and moiré confinement, and provides a microscopic framework for engineering exciton character and pressure-tunable optical selection in moiré superlattices.

This work is supported by the National Key R&D Program of China (Grants No. 2021YFA1401300 and No. 2023YFA1406000), the National Natural Science Foundation of China (Grants No. 92476108, No. 12504222, and No. 12674034), and the Innovation Program for Quantum Science and Technology (Grants No. 2021ZD0302400 and No. 2021ZD0302300). C.H. acknowledges support from the start-up fund and the Direct Grant (Grant No. 4053726) of the Chinese University of Hong Kong, and the State Key Laboratory of Quantum Information Technologies and Materials. J.S. acknowledges support from Beijing Natural Science Foundation (Grant No. 1262040).

DATA AVAILABILITY

The data that support the findings of this article are available from the authors upon reasonable request.

-
- [1] Y. Cao, V. Fatemi, S. Fang, K. Watanabe, T. Taniguchi, E. Kaxiras, and P. Jarillo-Herrero, *Unconventional superconductivity in magic-angle graphene superlattices*, *Nature (London)* **556**, 43 (2018).
- [2] Y. Cao *et al.*, *Correlated insulator behaviour at half-filling in magic-angle graphene superlattices*, *Nature (London)* **556**, 80 (2018).
- [3] Y. Xia, Z. Han, K. Watanabe, T. Taniguchi, J. Shan, and K. F. Mak, *Superconductivity in twisted bilayer WSe₂*, *Nature (London)* **637**, 833 (2025).
- [4] Y. Guo *et al.*, *Superconductivity in 5.0° twisted bilayer WSe₂*, *Nature (London)* **637**, 839 (2025).
- [5] E. C. Regan *et al.*, *Mott and generalized Wigner crystal states in WSe₂/WS₂ moiré superlattices*, *Nature (London)* **579**, 359 (2020).
- [6] Y. Tang *et al.*, *Simulation of Hubbard model physics in WSe₂/WS₂ moiré superlattices*, *Nature (London)* **579**, 353 (2020).
- [7] Y. Xu, S. Liu, D. A. Rhodes, K. Watanabe, T. Taniguchi, J. Hone, V. Elser, K. F. Mak, and J. Shan, *Correlated insulating states at fractional fillings of moiré superlattices*, *Nature (London)* **587**, 214 (2020).
- [8] L. Wang *et al.*, *Correlated electronic phases in twisted bilayer transition metal dichalcogenides*, *Nat. Mater.* **19**, 861 (2020).
- [9] C. Hu, V. Michaud-Rioux, W. Yao, and H. Guo, *Moiré valleytronics: realizing dense arrays of topological helical channels*, *Phys. Rev. Lett.* **121**, 186403 (2018).
- [10] A. L. Sharpe, E. J. Fox, A. W. Barnard, J. Finney, K. Watanabe, T. Taniguchi, M. A. Kastner, and D. Goldhaber-Gordon, *Emergent ferromagnetism near three-quarters filling in twisted bilayer graphene*, *Science* **365**, 605 (2019).
- [11] G. Wang, A. Chernikov, M. M. Glazov, T. F. Heinz, X. Marie, T. Amand, and B. Urbaszek, *Colloquium: Excitons in atomically thin transition metal dichalcogenides*, *Rev. Mod. Phys.* **90**, 021001 (2018).
- [12] H. Yu, G.-B. Liu, J. Tang, X. Xu, and W. Yao, *Moiré excitons: From programmable quantum emitter arrays to spin-orbit-coupled artificial lattices*, *Sci. Adv.* **3**, e1701696 (2017).
- [13] K. Tran *et al.*, *Evidence for moiré excitons in van der Waals heterostructures*, *Nature (London)* **567**, 71 (2019).
- [14] M. H. Naik *et al.*, *Intralayer charge-transfer moiré excitons in van der Waals superlattices*, *Nature (London)* **609**, 52 (2022).
- [15] C. Hu, M. H. Naik, Y.-H. Chan, J. Ruan, and S. G. Louie, *Light-induced shift current vortex crystals in moiré heterobilayers*, *Proc. Natl. Acad. Sci. U.S.A.* **120**, e2314775120 (2023).
- [16] A. Kogar *et al.*, *Signatures of exciton condensation in a transition metal dichalcogenide*, *Science* **358**, 1314 (2017).
- [17] Z. Wang, D. A. Rhodes, K. Watanabe, T. Taniguchi, J. C. Hone, J. Shan, and K. F. Mak, *Evidence of high-temperature exciton condensation in two-dimensional atomic double layers*, *Nature (London)* **574**, 76 (2019).
- [18] M. M. Fogler, L. V. Butov, and K. S. Novoselov, *High-temperature superfluidity with indirect excitons in van der Waals heterostructures*, *Nat. Commun.* **5**, 4555 (2014).
- [19] L. Gu *et al.*, *Giant optical nonlinearity of Fermi polarons in atomically thin semiconductors*, *Nat. Photonics* **18**, 816 (2024).
- [20] X. Zhao *et al.*, *Pressure tuning of layer-hybridized excitons in trilayer WSe₂*, *Nano Lett.* **26**, 1035 (2026).
- [21] G. Shen *et al.*, *Toward an international practical pressure scale: A proposal for an IPPS ruby gauge (IPPS-Ruby2020)*, *High Press. Res.* **40**, 299 (2020).
- [22] See Supplemental Material at <http://link.aps.org/supplemental/10.1103/lgl7-ww2r> for experimental and computational details, additional discussions, and supporting figures, which includes Refs. [23–43].
- [23] Y. Ye, X. Dou, K. Ding, D. Jiang, F. Yang, and B. Sun, *Pressure-induced K–Λ crossing in monolayer WSe₂*, *Nanoscale* **8**, 10843 (2016).
- [24] X. Ma *et al.*, *Robust interlayer exciton in WS₂/MoSe₂ van der Waals heterostructure under high pressure*, *Nano Lett.* **21**, 8035 (2021).
- [25] L. Fu *et al.*, *K–Λ crossover transition in the conduction band of monolayer MoS₂ under hydrostatic pressure*, *Sci. Adv.* **3**, e1700162 (2017).
- [26] S. Plimpton, *Computational limits of classical molecular dynamics simulations*, *Comput. Mater. Sci.* **4**, 361 (1995).
- [27] F. H. Stillinger and T. A. Weber, *Computer simulation of local order in condensed phases of silicon*, *Phys. Rev. B* **31**, 5262 (1985).
- [28] J.-W. Jiang and Y.-P. Zhou, *Handbook of Stillinger-Weber Potential Parameters for Two-Dimensional Atomic Crystals* (IntechOpen, London, 2017), 10.5772/intechopen.71767.
- [29] M. H. Naik, I. Maity, P. K. Maiti, and M. Jain, *Kolmogorov–Crespi potential for multilayer transition-metal dichalcogenides: Capturing structural transformations in moiré superlattices*, *J. Phys. Chem. C* **123**, 9770 (2019).
- [30] A. N. Kolmogorov and V. H. Crespi, *Registry-dependent interlayer potential for graphitic systems*, *Phys. Rev. B* **71**, 235415 (2005).
- [31] D. Xiao, G.-B. Liu, W. Feng, X. Xu, and W. Yao, *Coupled spin and valley physics in monolayers of MoS₂ and other group-VI dichalcogenides*, *Phys. Rev. Lett.* **108**, 196802 (2012).
- [32] R. Wang, K. Chang, W. Duan, Y. Xu, and P. Tang, *Twist-angle-dependent valley polarization of intralayer moiré excitons in van der Waals superlattices*, *Phys. Rev. Lett.* **134**, 026904 (2025).
- [33] C. Wang, X.-W. Zhang, X. Liu, Y. He, X. Xu, Y. Ran, T. Cao, and D. Xiao, *Fractional Chern insulator in twisted bilayer MoTe₂*, *Phys. Rev. Lett.* **132**, 036501 (2024).
- [34] J. M. Soler, E. Artacho, J. D. Gale, A. García, J. Junquera, P. Ordejón, and D. Sánchez-Portal, *The SIESTA method for ab initio order-N materials simulation*, *J. Phys. Condens. Matter* **14**, 2745 (2002).
- [35] D. R. Hamann, *Optimized norm-conserving Vanderbilt pseudopotentials*, *Phys. Rev. B* **88**, 085117 (2013).

- [36] M. Schlipf and F. Gygi, *Optimization algorithm for the generation of ONCV pseudopotentials*, *Comput. Phys. Commun.* **196**, 36 (2015).
- [37] M. Rohlfing and S. G. Louie, *Excitonic effects and the optical absorption spectrum of hydrogenated Si clusters*, *Phys. Rev. Lett.* **80**, 3320 (1998).
- [38] M. Rohlfing and S. G. Louie, *Electron-hole excitations and optical spectra from first principles*, *Phys. Rev. B* **62**, 4927 (2000).
- [39] L. V. Keldysh, *Coulomb interaction in thin semiconductor and semimetal films*, *JETP Lett.* **29**, 658 (1979), http://jetpletters.ru/ps/1455/article_22207.pdf.
- [40] A. Chernikov, T. C. Berkelbach, H. M. Hill, A. Rigosi, Y. Li, B. Aslan, D. R. Reichman, M. S. Hybertsen, and T. F. Heinz, *Exciton binding energy and nonhydrogenic Rydberg series in monolayer WS₂*, *Phys. Rev. Lett.* **113**, 076802 (2014).
- [41] E. Mostaani, M. Szyniszewski, C. H. Price, R. Maezono, M. Danovich, R. J. Hunt, N. D. Drummond, and V. I. Fal'ko, *Diffusion quantum Monte Carlo study of excitonic complexes in two-dimensional transition-metal dichalcogenides*, *Phys. Rev. B* **96**, 075431 (2017).
- [42] H. C. Kamban and T. G. Pedersen, *Interlayer excitons in van der Waals heterostructures: Binding energy, Stark shift, and field-induced dissociation*, *Sci. Rep.* **10**, 5537 (2020).
- [43] G. Onida, L. Reining, and A. Rubio, *Electronic excitations: Density-functional versus many-body Green's-function approaches*, *Rev. Mod. Phys.* **74**, 601 (2002).
- [44] Y. Li, Y. Rao, K. F. Mak, Y. You, S. Wang, C. R. Dean, and T. F. Heinz, *Probing symmetry properties of few-layer MoS₂ and h-BN by optical second-harmonic generation*, *Nano Lett.* **13**, 3329 (2013).
- [45] C. Jin *et al.*, *Observation of moiré excitons in WSe₂/WS₂ heterostructure superlattices*, *Nature (London)* **567**, 76 (2019).
- [46] A. Weston *et al.*, *Atomic reconstruction in twisted bilayers of transition metal dichalcogenides*, *Nat. Nanotechnol.* **15**, 592 (2020).
- [47] H. Li *et al.*, *Imaging moiré flat bands in three-dimensional reconstructed WSe₂/WS₂ superlattices*, *Nat. Mater.* **20**, 945 (2021).
- [48] Y. Wang *et al.*, *Pressure-driven moiré potential enhancement and tertiary gap opening in graphene/h-BN heterostructure*, *Phys. Rev. Lett.* **135**, 046303 (2025).
- [49] M. Yankowitz, J. Jung, E. Laksono, N. Leconte, B. L. Chittari, K. Watanabe, T. Taniguchi, S. Adam, D. Graf, and C. R. Dean, *Dynamic band-structure tuning of graphene moiré superlattices with pressure*, *Nature (London)* **557**, 404 (2018).
- [50] Y. Gao, X. Lin, T. Smart, P. Ci, K. Watanabe, T. Taniguchi, R. Jeanloz, J. Ni, and J. Wu, *Band engineering of large-twist-angle graphene-hBN moiré superlattices with pressure*, *Phys. Rev. Lett.* **125**, 226403 (2020).
- [51] B. Szentpéteri *et al.*, *Tailoring the band structure of twisted double bilayer graphene with pressure*, *Nano Lett.* **21**, 8777 (2021).
- [52] M. Yankowitz, S. Chen, H. Polshyn, Y. Zhang, K. Watanabe, T. Taniguchi, D. Graf, A. F. Young, and C. R. Dean, *Tuning superconductivity in twisted bilayer graphene*, *Science* **363**, 1059 (2019).
- [53] P. Jiao *et al.*, *Hydrostatic pressure-enhanced correlated magnetism and Chern insulator in moiré WSe₂*, *Phys. Rev. X* **16**, 011068 (2026).
- [54] K. Wang *et al.*, *Interlayer coupling in twisted WSe₂/WS₂ bilayer heterostructures revealed by optical spectroscopy*, *ACS Nano* **10**, 6612 (2016).
- [55] I. Stenger, L. Schué, M. Boukchicha, B. Berini, B. Plaçais, A. Loiseau, and J. Barjon, *Low frequency Raman spectroscopy of few-atomic-layer thick hBN crystals*, *2D Mater.* **4**, 031003 (2017).
- [56] C. H. Lui *et al.*, *Observation of interlayer phonon modes in van der Waals heterostructures*, *Phys. Rev. B* **91**, 165403 (2015).
- [57] J. Yan, J. Xia, X. Wang, L. Liu, J.-L. Kuo, B. K. Tay, S. Chen, W. Zhou, Z. Liu, and Z. X. Shen, *Stacking-dependent interlayer coupling in trilayer MoS₂ with broken inversion symmetry*, *Nano Lett.* **15**, 8155 (2015).
- [58] J. Xia, J. Yan, Z. Wang, Y. He, Y. Gong, W. Chen, T. C. Sum, Z. Liu, P. M. Ajayan, and Z. Shen, *Strong coupling and pressure engineering in WSe₂/MoSe₂ heterobilayers*, *Nat. Phys.* **17**, 92 (2021).
- [59] L. Liang, J. Zhang, B. G. Sumpter, Q.-H. Tan, P.-H. Tan, and V. Meunier, *Low-frequency shear and layer-breathing modes in Raman scattering of two-dimensional materials*, *ACS Nano* **11**, 11777 (2017).
- [60] X. Wang *et al.*, *Intercell moiré exciton complexes in electron lattices*, *Nat. Mater.* **22**, 599 (2023).
- [61] E. Liu, T. Taniguchi, K. Watanabe, N. M. Gabor, Y.-T. Cui, and C. H. Lui, *Excitonic and valley-polarization signatures of fractional correlated electronic phases in a WSe₂/WS₂ moiré superlattice*, *Phys. Rev. Lett.* **127**, 037402 (2021).
- [62] W. Zhao *et al.*, *Dynamic tuning of moiré excitons in a WSe₂/WS₂ heterostructure via mechanical deformation*, *Nano Lett.* **21**, 8910 (2021).
- [63] B. Aslan, M. Deng, and T. F. Heinz, *Strain tuning of excitons in monolayer WSe₂*, *Phys. Rev. B* **98**, 115308 (2018).
- [64] P. Steeger, J. H. Graalman, R. Schmidt, I. Kuppenko, C. Sanchez-Valle, P. Maruhn, T. Deilmann, S. M. de Vasconcellos, M. Rohlfing, and R. Bratschitsch, *Pressure dependence of intra- and interlayer excitons in 2H-MoS₂ bilayers*, *Nano Lett.* **23**, 8947 (2023).
- [65] F. Wu, T. Lovorn, E. Tutuc, I. Martin, and A. H. MacDonald, *Topological insulators in twisted transition metal dichalcogenide homobilayers*, *Phys. Rev. Lett.* **122**, 086402 (2019).
- [66] Z. Ye, T. Cao, K. O'Brien, H. Zhu, X. Yin, Y. Wang, S. G. Louie, and X. Zhang, *Probing excitonic dark states in single-layer tungsten disulphide*, *Nature (London)* **513**, 214 (2014).
- [67] H. Park *et al.*, *Dipole ladders with large Hubbard interaction in a moiré exciton lattice*, *Nat. Phys.* **19**, 1286 (2023).
- [68] R. Xiong, J. H. Nie, S. L. Brantly, P. Hays, R. Sailus, K. Watanabe, T. Taniguchi, S. Tongay, and C. Jin, *Correlated insulator of excitons in WSe₂/WS₂ moiré superlattices*, *Science* **380**, 860 (2023).
- [69] Z. Lian *et al.*, *Valley-polarized excitonic Mott insulator in WS₂/WSe₂ moiré superlattice*, *Nat. Phys.* **20**, 34 (2024).