

Review: Spatial inhomogeneities, moiré potential and moiré excitons.

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In this short review, we provide an overview of recent progress in deploying advanced characterization techniques to understand the effects of local inhomogeneities in moiré heterostructures over multiple length scales. Particular emphasis is placed on correlating the impact of twist angle misalignment, nano-scale disorder, and atomic relaxation on the moiré potential and its collective excitations, particularly moiré excitons. Finally, we discuss future technological applications leveraging based on moiré excitons.

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I. INTRODUCTION

In condensed matter, moiré patterns emerge when two or more atomically thin layers are artificially stacked with a slight angular misalignment or, in the case of heterolayers, are stacked featuring a small lattice constant mismatch (Du *et al.*, 2023b). Remarkably, the moiré interference patterns create a superstructure, the moiré lattice, with a new periodicity that is usually much larger than the underlying atomic lattices. This superlattice can dramatically alter the electronic band structure of the material, leading to the formation of miniband structures and flat bands that directly affect the transport behaviour. Some key discoveries in this field of moiré engineering include the formation of flat bands (Bistritzer and MacDonald, 2011), promoting the relative relevance of correlations, but also the engineering of topological properties (Choi *et al.*, 2021), controlling polaritons (Zhang *et al.*, 2021) or networks of solitons (Fu *et al.*, 2020) and excitons, the key quasi-particle we will focus upon in our review. We refer the reader to recent reviews of moiré physics for an in-depth discussion of moiré physics: (Alexeev *et al.*, 2019; Behura *et al.*, 2021; Ciarrocchi *et al.*, 2022; Huang *et al.*, 2022; Mueller and Malic, 2018; Zhang *et al.*, 2023a)

Moiré excitons refer to photo-excited electron-hole bound states that form within (intra-) or between (inter-) layers with the potential defined by the moiré superpattern (Tran *et al.*, 2019). Moiré excitons exhibit distinct energy levels and behaviors compared to their semiconductor-monolayer or three-dimensional counterparts. For a review of exciton physics in transition metal dichalcogenides and their applications, we refer the reader to Ref. (Mueller and Malic, 2018). Moiré excitons can host a rich tapestry of phenomena with valuable insights into novel quantum phenomena and many-body effects in low-dimensional systems, including exciton condensation, superfluidity, and topologically protected states (Choi *et al.*, 2021; Shimazaki *et al.*, 2020). Moiré excitons also offer opportunities for tuning the electronic band structure of materials, enabling tailored properties for semiconductors, insulators, and topological materials, paving the way for technological innovation (Ciarrocchi *et al.*, 2022). Additionally, they contribute a new dimension to valleytronics, allowing for control over valley polarization and dynamics in van der Waals heterostructures (Jin *et al.*, 2019; Unuchek *et al.*, 2019). Furthermore, moiré excitons play a pivotal role in realizing topological phases and the quantum anomalous Hall effect, leading to the development of innovative electronic and spintronic devices (Li *et al.*, 2021b). Interactions between moiré excitons and antiferromagnetic ordering enable the manipulation of spin-dependent properties and spin information in optoelectronic devices (Ramos *et al.*, 2022). Moreover, moiré excitons present opportunities for controlling carrier dynamics and charge transport properties

in heterostructures, resulting in high-performance optoelectronic devices with improved efficiency and reduced energy consumption. Excellent, complementary reviews of moiré excitons are (Huang *et al.*, 2022) focusing on early optical experiments and selection rules and (Ciarrocchi *et al.*, 2022) which highlights device architectures and engineering approaches to tailor excitons toward future applications.

Controlling and understanding moiré superlattices, arising from the periodicity of moiré patterns, is fundamental to engineer moiré excitons and their dynamics phenomena (Brem *et al.*, 2020; Meneghini *et al.*, 2024; Merkl *et al.*, 2020). However, the excitonic properties of moiré heterostructures are extremely sensitive to local deviations from the ideal Lego-like description. In this short overview, we embark on a journey through multiple length scales and how new theoretical descriptions and advanced characterization techniques (Figure 1) have revealed the effects of twist angle misalignment, local inhomogeneities, and atomic relaxation in the physics of moiré excitons.

II. EFFECT OF SPATIAL INHOMOGENEITIES ON MOIRÉ HETEROSTRUCTURES

Excitonic states in moiré van der Waals superlattices offer exciting prospects for novel optoelectronics and quantum technologies (Ciarrocchi *et al.*, 2022). The strong exciton physics in TMD-based materials has been extensively investigated using different optical spectroscopy techniques, such as absorption spectroscopy, photoluminescence and photocurrent measurements, and optical pump-probe measurements (Huang *et al.*, 2022). While these techniques provide access to spectral and temporal degrees of freedom, they lack information about the real space properties of excitonic TMDs. However, local spatial modulations of the moiré superstructure due to strain fields, atomic defects (Guo *et al.*, 2021), and twist angle change extremely influence their optoelectronic properties. Correlating the averaged optical and transport properties with these local variations is needed to unambiguously resolve the role of the moiré energy and length scale. In the following, we review recent applications of advanced characterization techniques to disentangling emergent excitonic phenomena from moiré potential disorder in order of spatial resolution.

A. Determining the twist angle of moiré heterobilayers via non-linear optical techniques

In the fabrication of heterobilayer devices, the twist angle, the effective misalignment between the main crystallographic directions of the two stack monolayers has emerged as a novel degree of freedom that controls the moiré potential and enables tunability of the optical

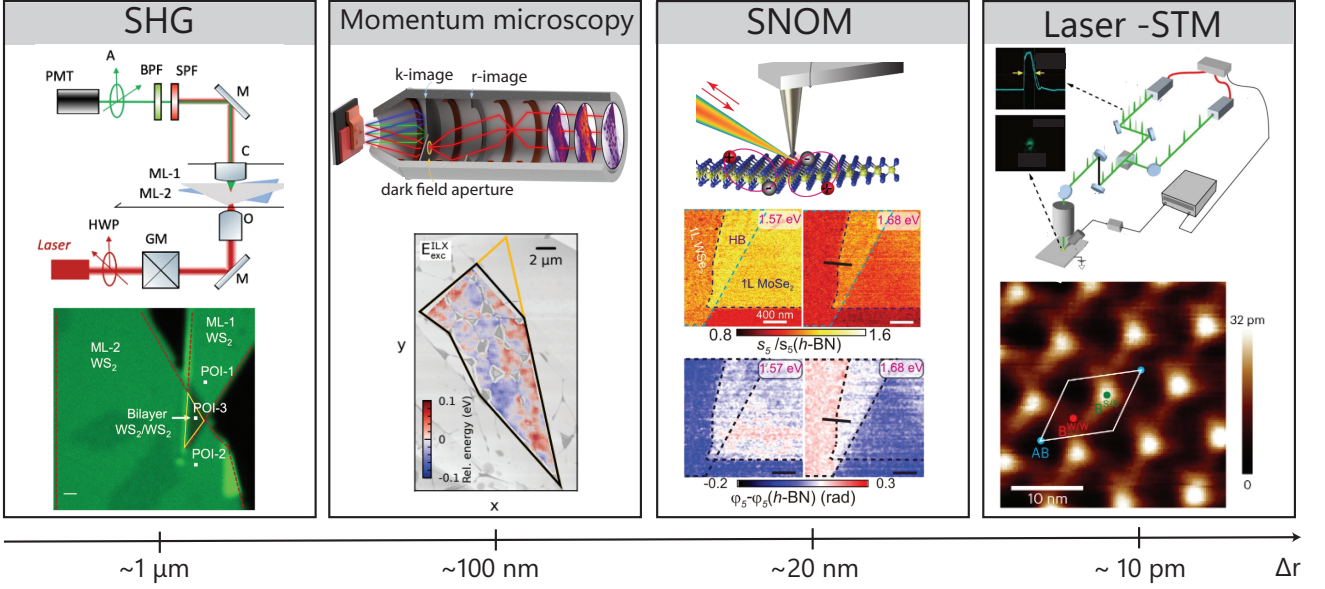


FIG. 1 A subset of advanced characterization techniques deployed to study the spatial variation of moiré heterostructures. From left to right, in order of increasing resolution, Second Harmonic Generation (SHG) ($\sim 1\mu\text{m}$), photoelectron momentum microscopy ($\sim 100\text{ nm}$), scanning near-infrared optical microscopy (SNOM, $\sim 20\text{ nm}$), and laser scanning tunneling microscopy and spectroscopy (laser-STM/S, $\sim 1\text{ pm}$). Adapted with permission from (Li *et al.*, 2024; Mogi *et al.*, 2019; Psilodimitrakopoulos *et al.*, 2021; Zhang *et al.*, 2022)

emission and absorption spectrum of TMD heterobilayers (Huang *et al.*, 2022) (Fig. 2-(a)). For example, in closely aligned $\text{WS}_2/\text{MoSe}_2$ heterobilayers, corresponding to $\theta = 0^\circ$ (AA stacking) and $\theta = 60^\circ$ (AB stacking), the almost perfect overlap between the two Brillouin zones leads to direct band gaps between inter- and intra-layer transitions. However, even small deviations from the AA or AB stacking lead to large changes in the band overlap in momentum space. Figure 2-(b) shows the evolution of the lower ($\approx 1.6\text{ eV}$) and upper hybrid excitons ($\approx 1.7\text{ eV}$) with twist angle as measured from reflection spectroscopy (Zhang *et al.*, 2020a) revealing changes to the excitonic spectrum of more than 100 meV as a function of the twist angle. Thus, it is fundamental to precisely determine the stacking angle between the heterobilayers for future devices based on moiré excitons.

In this context, the observation of strong non-linear responses dependent on the twist angle has emerged as a unique and versatile tool to characterize the formation of the moiré excitons (Jin *et al.*, 2019; Tran *et al.*, 2019). Similarly, changes of symmetry and nonlinear responses associated by the interlayer coupling and charge transfer associated with the moiré potential have also been detected via non-linear responses (Deng *et al.*, 2022; Shree *et al.*, 2021b; Yang *et al.*, 2020). For strong electric fields, the interaction of light can be modeled by a resulting po-

larization given by:

$$P_i(t) = \varepsilon_0(\chi_i^{(1)} E_i(t) + \chi_{ij}^{(2)} E_i(t)E_j(t) + \chi_{ijk}^{(3)} E_i(t)E_j(t)E_k(t) + \dots) \quad (1)$$

with $\chi^{(i)}$ as the linear and non-linear optical susceptibility tensors and ε_0 as the permittivity of vacuum. These high-order processes are governed by the crystallographic symmetry and the electronic band structure, carrying fundamental information about the wavefunctions involved in the two-photon absorption process (Ma *et al.*, 2021). Terms with an even number of powers on the electric field E are only allowed in inversion symmetry-breaking compounds. The second-order term carries special importance as it is responsible of second-harmonic generation (SHG), in which the constructive interference of the two oscillating electric fields with frequency ω can give rise to a polarization density at twice the frequency $P(2\omega)$ (Zhao *et al.*, 2018). In a normal-incidence SHG experiment, a single wavelength or broadband light pulse with controlled polarization is focused onto the sample by a microscopic objective. Emission from the sample is first collimated by the objective and a short-pass filtered to eliminate contributions from the fundamental before being focused onto, a generally fixed, photodiode or CCD camera. The angular dependence of the intensity of the 2ω response with respect to the main crystallographic symmetries and the polarization of the incoming and outgoing beam enables the resolution of the underlying structural point group.

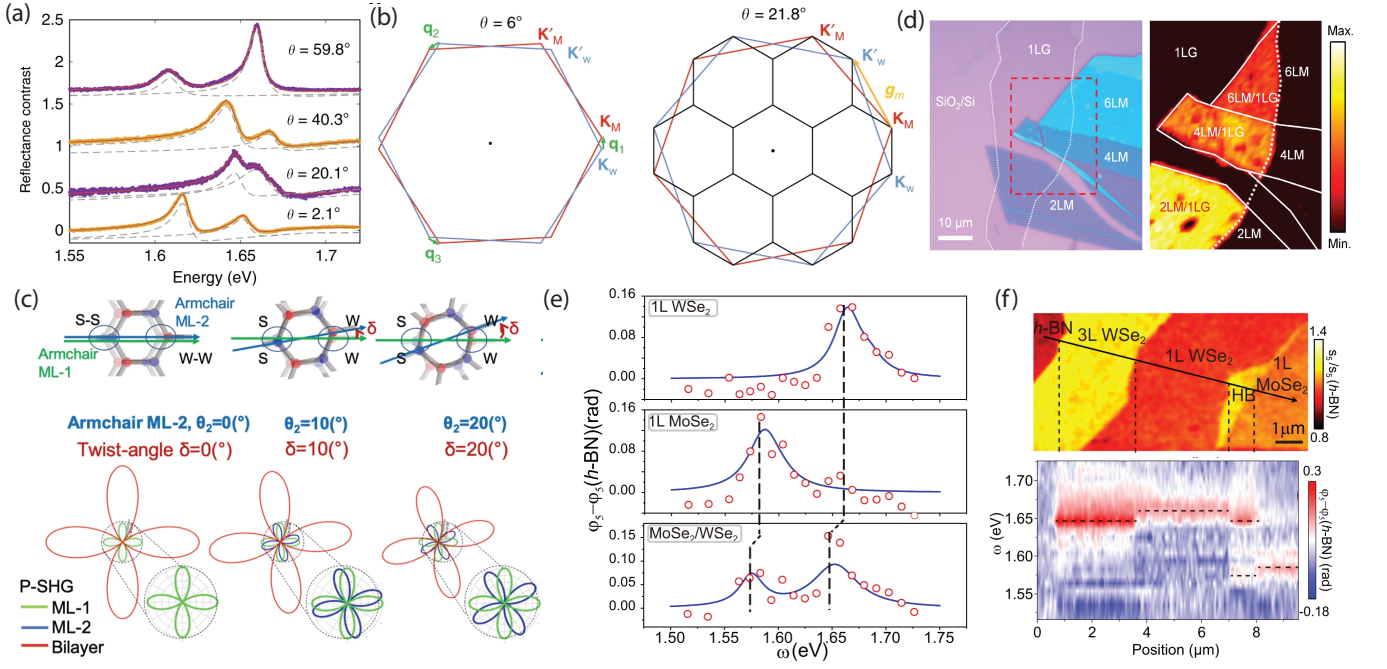


FIG. 2 a) Normalized reflections spectroscopy of $\text{WS}_2/\text{MoSe}_2$ bilayers as a function of twist angle. (b) Schematic of the Brillouin zones for each of the two ML (red, blue) and the moiré heterostructures (black) for a small twist angle (6°) and a large twist angle (21.8°). (c) Schematic of the alignment of the main crystallographic directions function of twist angle, θ , keeping the armchair direction of the bottom ML fixed and rotating the armchair direction of the top ML. The simulated interference SHG pattern reflects the twist angle by the rotation of the lobes and the modulation of the overall intensity. (d) Micrograph of MoSe_2 (LM) and Graphene (LG) for different stacking configurations and the associated second-harmonic generation (SHG) spatial imaging map in the highlighted dashed red box. (e) Normalized phase spectra at different positions in the sample indicating the excitonic responses for the WSe_2 , MoSe_2 monolayers, and the $\text{MoSe}_2/\text{WSe}_2$ heterobilayer. (f) Near field image away from resonance and phase resolve map as a function of energy revealing the spatial dependence of the excitonic energies. Panels adapted with permission from (Psilodimitrakopoulos *et al.*, 2021; Zhang *et al.*, 2020a, 2023b, 2022)

For 2D transition metal dichalcogenide (TMD) monolayers, a strong SHG response with maxima along the armchair or zig-zag direction can be measured due to their inversion symmetry-breaking nature (point group D_{3h}). When a heterobilayer is formed, the SHG signal from each layers coherently interferes resulting in a pattern that is extremely sensitive to small twist angle misalignment from perfect AA and AB stacking, as shown in Fig.2 (Hsu *et al.*, 2014; Psilodimitrakopoulos *et al.*, 2021). Note that a twist angle of $\theta = 60^\circ$ results in a completely destructive interference, fully suppressing the SHG response. Moreover, strong electric dipole SHG from centrosymmetric bilayer MoS_2 with 2 - H stacking has been observed (Shree *et al.*, 2021a; Zhang *et al.*, 2023b) due to a resonance enhancement associated with inter- and intra-layer excitons, a phenomenon also expected to occur in heterobilayers hosting moiré excitons. Indeed, strong twist angle enhancement has been observed in $\text{MoS}_2/\text{WSe}_2$ devices at the intralayer hybridized exciton energies (Paradisanos *et al.*, 2022). Comprehensive reviews of the enhancement of non-linear processes in twisted and heterostructures of TDMs can be found in Ref. (Wen *et al.*, 2019; Zhou *et al.*, 2022).

Optical contrast of the moiré heterobilayers beyond subwavelength resolution (~ 20 nm) can be achieved by scanning near-field optical microscopy (SNOM) (Vincent, 2021). In a scattering SNOM (s-SNOM), the metallic AFM tip locally confines the electric light field. Thus, the excitonic spectrum of TMD heterostructures could be resolved by measuring the scattered light amplitude and phase as a function of position and energy (McLeod *et al.*, 2014). s-SNOM has been recently deployed to monolayers of MoSe_2 and WSe_2 and $\text{MoSe}_2/\text{WSe}_2$ heterobilayers to extract the excitonic resonance energy and interlayer hybridization with nanometer resolution (Zhang *et al.*, 2022). These early measurements have open the way for local measurements and control of moiré excitons. For example, the AFM tip from a SNOM has been used to introduce a vertical deformation in heterostructures of WSe_2/WS_2 at cryogenic temperatures and concomitantly measure a shift on the moiré intralayer exciton as a function of uniaxial pressure (Zhao *et al.*, 2021), as shown in Fig. 2.

Optical characterization techniques with real space imaging capabilities are thus fundamental to studying the properties of moiré heterostructure and excitons. More-

over, their large versatility enables a multi-messenger approach in which the combination of different wavelengths and scanning techniques characterize the excitation spectrum over a broad range of energies (Sternbach *et al.*, 2023).

B. Nano-imaging of moiré exciton using photoemission microscopes

Angle-resolved photoemission spectroscopy (ARPES) is a technique that explicitly accesses the material-specific electronic band structure (Sobota *et al.*, 2021). Its extension to the time domain (tr-ARPES) is sensitive to the transient dynamics of carriers with momentum resolution. In a tr-ARPES measurement, a pump pulse, typically in the visible or near-infrared range, excites electrons at a specific energy in the conduction band. Then, a second extreme ultraviolet probe pulse is applied to eject electrons from the material (Fig. 3 (a)) – according to Einstein’s photoelectric effect. Measuring the kinetic energy and the emission angle of the ejected electrons allows us to extract the energy and momentum of the initially excited electrons in the material (Karni *et al.*, 2023). Moreover, the successful development of high-repetition-rate extreme ultraviolet light sources paved the way for tr-ARPES measurements on TMD materials. This powerful technique has been demonstrated to visualize momentum-dark excitons in TMD monolayers directly (Madéo *et al.*, 2020) and the momentum-resolved charge transfer dynamics in twisted TMD heterostructures (Bange *et al.*, 2024; Schmitt *et al.*, 2022). While the first ARPES studies were performed on exciton dynamics in bulk TMD materials (Bertoni *et al.*, 2016; Dong *et al.*, 2021; Grubišić Čabo *et al.*, 2015; Wallauer *et al.*, 2020), the more recent results, enabled by the development of time-of-flight momentum microscopes, have focused on the exciton physics of TMD monolayers (Kunin *et al.*, 2023; Madéo *et al.*, 2020; Wallauer *et al.*, 2021). Typically, tr-ARPES experiments do not spatially resolve the carrier dynamics, but they average over large sample areas of typically $10\mu\text{m}$ or more. Thus, they cannot access spatial inhomogeneities, dielectric disorder, strain gradients, or local lattice reconstructions. Recently, ultrafast dark field momentum microscopy has been introduced as a new technique for simultaneous nano-imaging and nano-spectroscopy of materials (Schmitt *et al.*, 2023). The technique has a temporal resolution of 55 fs and a spatial resolution of ≈ 100 nm and has been applied to study the spatial heterogeneity in a WSe_2 - MoS_2 heterostructure as well as its impact on the energy and the formation time of interlayer excitons. For a more in-depth discussion of the techniques’ details we refer the reader to other recent reviews (Karni *et al.*, 2023; Sobota *et al.*, 2021).

For excitonic materials, such as TMDs, tr-ARPES

measurements provide access to exciton binding energies and their momentum in the reciprocal space. Here, the excitation pulse first creates excitons in the material. In contrast, the energy of the second pulse is used to break up the Coulomb-bound electron-hole pairs, photo-ejecting the electrons while leaving a hole density behind (Fig. 3 (a)). The ARPES signal of the photoemitted electrons reflects the excitonic band structure since the Coulomb-bound electrons and holes present a strongly correlated system (Meneghini *et al.*, 2023). The excitonic ARPES signal appears at the momentum corresponding to the ejected electron’s reciprocal-space valley. It is spectrally located one exciton binding energy below the conduction band. The momentum-resolved shape of the signal is characterized by the negative curvature of the valence band of the remaining hole. This negative dispersion from the photoemitted electron is the hallmark of the excitonic origin of the ARPES signal (Christiansen *et al.*, 2019; Man *et al.*, 2021; Meneghini *et al.*, 2023).

In the following, we first review the exciton dynamics in TMD homo- and heterobilayers, which have attracted huge interest in current research. The latter exhibits a type-II band alignment favoring the tunneling of an electron or hole into the opposite layer and resulting in the formation of spatially separated interlayer excitons (Yong *et al.*, 2019), where the Coulomb-bound electrons and holes are located in different layers (Fig. 3 (b)-(c)). Finally, we discuss how the deployment of ultrafast dark field momentum microscopy has revealed local inhomogeneities of the dark excitons.

ARPES signatures of the electron and hole bound in an interlayer exciton in a twisted WSe_2 - MoS_2 heterostructure have been observed [Fig. 3(d)-(i)] thanks to the high signal-to-noise ratio and high-quality samples with narrow initial valence band line widths (Karni *et al.*, 2022). The unexcited band structure of the heterostructure is characterized by two spin-split valence band maxima of the WSe_2 layer at the K valley and a single broadband at lower energies corresponding to the valence band of the MoS_2 layer (Fig. 3 (d)). After optically exciting the material resonant at the intralayer exciton in WSe_2 , clear signatures of interlayer excitons about 1 eV above the valence band maximum are found (Fig. 4 (e)). Interestingly, the signal around the WSe_2 valence band maximum becomes slightly depleted at the K and K’ points (Fig. 4 (f)). This was assigned to the holes created in WSe_2 during the photoexcitation. Finally, the interlayer exciton signature at 1 eV exhibits a negative dispersion resembling the negative curvature of the WSe_2 valence band (Fig. 3(i)). This anomalous dispersion has been theoretically predicted (Christiansen *et al.*, 2019) and is a hallmark of the excitonic origin of the observed ARPES signal.

Recently, it was shown that tr-ARPES is an ideal technique to visualize hybrid exciton states directly (Meneghini *et al.*, 2023). Tunneling between two TMD lay-

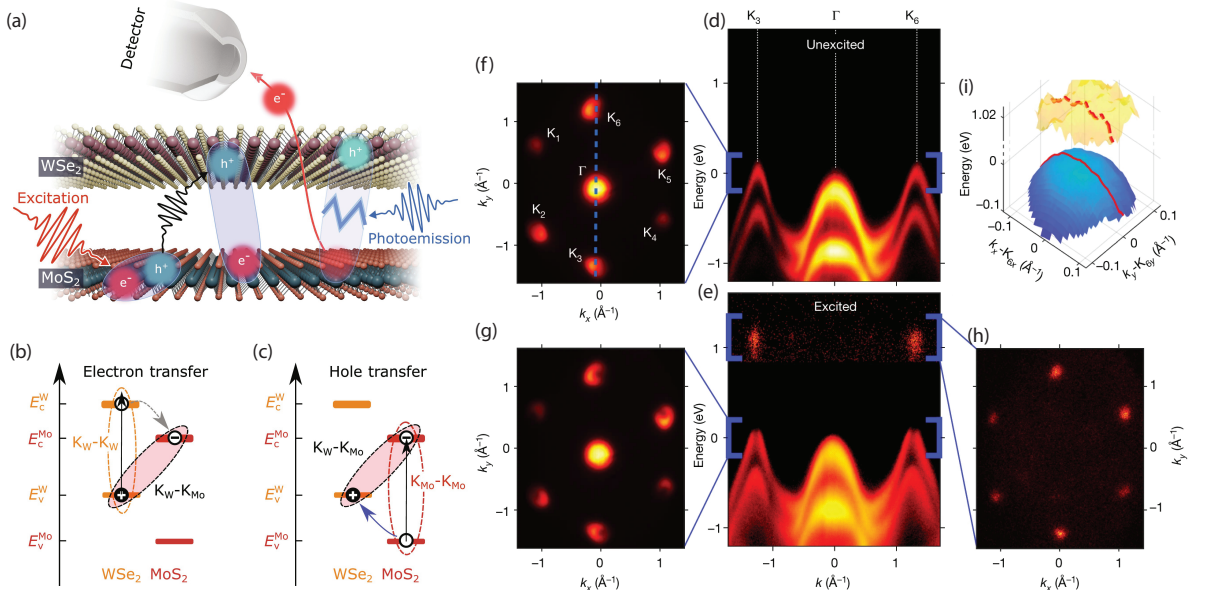


FIG. 3 (a) Schematic on the ARPES process in the exemplary WSe₂-MoS₂ heterostructure after optical excitation of excitons. A second extreme ultraviolet laser pulse breaks the exciton, and the ejected electrons are detected while the holes remain in the material. (b)-(c) Electronic band alignment in the heterostructure with the formation of interlayer excitons via electron or hole transfer, depending on the excitation condition. Energy-momentum cuts of the ARPES signal along the K- Γ direction in the Brillouin zone (d) before and (e) after optical excitation of excitons in the WSe₂-MoS₂ heterostructure. The insets show the zoomed-in momentum-space images around the valence band minimum (f,g) and around the interlayer exciton energy (h). Note that the interlayer exciton signal has been enhanced by a factor of 50. (i) The ARPES signal from the ejected electrons exhibits the negative dispersion of the remaining holes. Figures adapted with permission from Ref. (Bange *et al.*, 2024; Karni *et al.*, 2022).

ers leads to a mixture of intra- and interlayer exciton states, resulting in hybrid excitons that inherit properties of both exciton species. Here, electrons and/or holes are in a superposition of both layers. The formation of such hybrid states is facilitated in naturally stacked TMD homobilayers, whereas the dominating excitons in TMD heterostructures are mostly of interlayer character. Thus, the performed study focuses on MoS₂ homobilayers, where hybridized momentum-dark $\Gamma_{\text{hyb}}\text{K}$ excitons are the energetically lowest states. Here, the hole at the Γ valley is strongly delocalized between the layers. During the ARPES process, the hybrid exciton breaks up into an ejected free electron from the K valley and a superposition of hybrid holes. This gives rise to a double-peak signal reflecting the superposition of the remaining holes between the two hybrid valence bands at the Γ point.

Figure 4 (a) illustrate how the momentum- and energy-resolved ARPES signal evolves in time. During the optical excitation, the spectrum is characterized by a pronounced single peak stemming from the optically excited intralayer exciton (Fig. 4 (a)). On a sub-100 fs timescale, these excitons scatter via emission of phonons to the energetically lower hybridized $\Gamma_{\text{hyb}}\text{K}$ excitons and the ARPES signal shows signatures of both KK intralayer and hybridized $\Gamma_{\text{hyb}}\text{K}$ excitons. After approx. 600 fs, an equilibrium distribution is reached with $\Gamma_{\text{hyb}}\text{K}$ exci-

tons carrying nearly the entire population. Interestingly, their ARPES signature is characterized by two peaks, one slightly above the KK exciton and one red-shifted by more than 600 meV.

Figure 4-(b) shows the momentum-integrated energy distribution of the tr-ARPES signal in WSe₂-MoS₂ heterostructures (Bange *et al.*, 2024). The highest contribution at early delay times occurs at around 1.7 eV (orange dashed line) corresponding to the energy of the optically excited $\text{K}_\text{W}\text{K}_\text{W}$ intralayer excitons in the WSe₂ layer. Interestingly, one observes a signal transition to a second long-lived peak at the lower energy of about 1.1 eV (red dashed line) occurring on a femtosecond timescale. This peak has been identified as the photoemitted electronic contribution of the $\text{K}_\text{W}\text{K}_\text{Mo}$ interlayer exciton. Here, the first/second capital letter denotes the position of the hole/electron in the reciprocal space (K, Λ valley), while the subindex stands for the layer (W or Mo). To support this assignment, the same measurement was performed on a WSe₂ monolayer. No spectral weight is observed in the momentum and energy space region associated with the interlayer excitons.

The observed charge transfer dynamics, Fig. 4(c), involves a phonon-driven two-step scattering process (Meneghini *et al.*, 2022). The optically excited intralayer $\text{K}_\text{W}\text{K}_\text{W}$ excitons in the WSe₂ layer cannot efficiently scat-

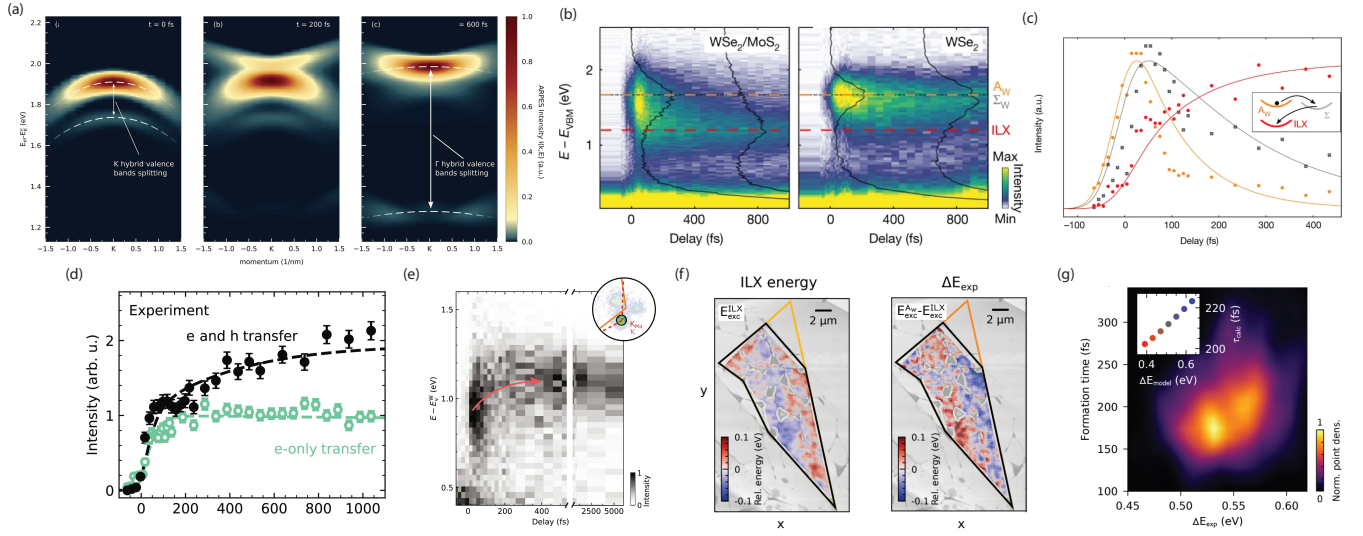


FIG. 4 (a) Theoretical prediction for the hybrid exciton dynamics in the MoS₂ homobilayer showing the excitation of the nearly purely intralayer KK excitons (0 fs), the formation of strongly hybridized $\Gamma_{\text{hyb}}\text{K}$ excitons (200 fs), and the thermalized hybrid exciton distribution (600 fs), respectively. The dashed lines denote the shifted split valence bands of the hybrid hole at the K and the Γ point. (b) Momentum-integrated energy-distribution for the heterostructure and in comparison also for the WSe₂ monolayer, respectively. (c) Charge-transfer dynamics from optical excitation of excitons in the WSe₂ monolayer ($\text{K}_\text{W}\text{K}_\text{W}$, orange line) via phonon-driven scattering into the hybrid dark excitons ($\text{K}_\text{W}\Lambda_{\text{hyb}}$, grey line) to the energetically lowest interlayer exciton states ($\text{K}_\text{W}\text{K}_{\text{M}_0}$, red line). The points represent experimental data and solid lines the theoretical prediction. (d) Direct comparison of the charge transfer dynamics when the heterostructure is excited resonant to the excitons either in the WSe₂ layer (green circles, 1.7 eV) or in the MoS₂ layer (black circles, 1.9 eV). While the first excitation scenario leads to an electron transfer only, the latter gives rise to a combined electron- and hole-transfer. (e) Time evolution of the energy distribution filtered at the momentum region of the K valley in MoS₂ layer for the excitation at 1.9 eV. As the interlayer exciton is formed, the photoelectron energy shows an unintuitive blue-shift. (f) Spatial heatmaps illustrating the variation of the interlayer exciton (ILX) energy and the energy difference ΔE_{exp} to the optically excited intralayer exciton, respectively. (g) The interlayer exciton formation time plotted as a function of the energy difference ΔE_{exp} . The inset shows a theoretical prediction confirming qualitatively the measured correlation between the formation time and the energy difference. Figures adapted with permission from Ref. (Bange *et al.*, 2024; Meneghini *et al.*, 2023; Schmitt *et al.*, 2022, 2023)

ter directly to the energetically lowest $\text{K}_\text{W}\text{K}_{\text{M}_0}$ interlayer exciton states (hole at the K valley of the WSe₂ layer and electron in the K valley of the MoSe₂) [Fig. 3-(b)]. The $\text{K}_\text{W}\text{K}_\text{W}$ excitons rather scatter first with phonons to the momentum-dark $\text{K}\Lambda$ excitons, where the electrons at the Λ valley are delocalized over both layers. In a second step, these hybridized excitons scatter into the $\text{K}_\text{W}\text{K}_{\text{M}_0}$ interlayer excitons, where the electron has been transferred to the MoSe₂ layer. The direct scattering process is prevented by the weak wave function overlaps at the K valley. In contrast, the electronic wave function at the Λ valley has large contributions at the outer selenium atoms, resulting in much more efficient hybridization of $\text{K}\Lambda$ states (Merkl *et al.*, 2020).

Tr-ARPES has also revealed the hole transfer process mechanism across the WSe₂-MoS₂ interface (Meneghini *et al.*, 2023). The $\text{K}_{\text{M}_0}\text{-K}_{\text{M}_0}$ intralayer excitons in the MoS₂ layer can be resonantly excited at 1.9 eV (Fig. 4-(d)). However, this still leads to a partial electron transfer due to the off-resonant excitation of the energetically lower $\text{K}_\text{W}\text{-K}_\text{W}$ excitons (1.7 eV) in the WSe₂ layer. To distinguish the impact of the electron and hole transfer,

the temporally resolved formation of $\text{K}_\text{W}\text{-K}_{\text{M}_0}$ interlayer excitons have been studied for both scenarios, i.e. excitation at 1.7 eV (only electron transfer expected) and excitation at 1.9 eV (electron and hole transfer expected). For the electron-only transfer process, the interlayer excitons are formed quickly (~ 35 fs), and the ARPES signal saturates on the sub-200 fs timescale (Fig. 4-(d)). In contrast, the joint electron- and hole-transfer is found to be much slower, with a formation time of approximately 3 ps. This can be traced back to the different relaxation pathways of optically excited $\text{K}_{\text{M}_0}\text{-K}_{\text{M}_0}$ and $\text{K}_{\text{M}_0}\text{-K}_{\text{M}_0}$ excitons. The degree of the hybridization of the intermediate hybrid states and the energetic offset to the initial and final states determine the efficiency of the charge transfer process. While the first scatter via hybridized $\Gamma_{\text{hyb}}\text{-K}_{\text{M}_0}$ excitons, the latter scatter via hybridized $\text{K}_\text{W}\text{-}\Lambda_{\text{hyb}}$ states towards to energetically lowest $\text{K}_\text{W}\text{-K}_{\text{M}_0}$ interlayer excitons.

Assuming that ARPES only provides information on the ejected electrons, one would not expect any difference whether the measured photoelectrons stem from the break-up of the intralayer $\text{K}_{\text{M}_0}\text{-K}_{\text{M}_0}$ or interlayer $\text{K}_\text{W}\text{-}$

K_{Mo} excitons as the electron remains in the MoSe_2 layer, while the hole transfers across the interface (Fig. 4-(d)). Interestingly, one finds a non-intuitive blue shift of about 170 meV of the ARPES signal for increasing delay times (Fig. 3-(e)). During this time, the optically excited intralayer excitons scatter into the energetically lower interlayer states, so one would rather naively expect a red-shift in the ARPES signal. Auger processes and energy renormalization could be excluded as possible explanations for the unexpected blue-shift. The unintuitive shift towards higher energies is a hallmark of the correlated nature of the electron-hole pairs.

Typically, tr-ARPES experiments do not spatially resolve the carrier dynamics, but they rather average over large sample areas of typically $10\mu\text{m}$ or more. Thus, they cannot access spatial inhomogeneities, dielectric disorder, strain gradients, or local lattice reconstructions. Recently, ultrafast dark field momentum microscopy has been introduced as a new technique for simultaneous nano-imaging and nano-spectroscopy of materials (Schmitt *et al.*, 2023). The technique has a temporal resolution of 55 fs and a spatial resolution of 500 nm and has been applied to study the spatial heterogeneity in a WSe_2 - MoS_2 heterostructure as well as its impact on the energy and the formation time of interlayer excitons. Interestingly, distinct spatial heterogeneity is found even in seemingly flat areas of the sample exhibiting high-quality photoemission spectra. Positioning the dark-field aperture on the momenta of the interlayer excitons provides access to the spatially and spectrally resolved evolution of the interlayer exciton energy. The nanoscale variation of the latter as well as its difference ΔE_{exp} to the energy of the optically excited intralayer exciton is shown in spatial heat maps in Fig. 4-(f). Interestingly, the experiment reveals a correlation between the interlayer exciton formation time τ and the energy difference ΔE_{exp} , i.e. τ gets larger with the increasing ΔE_{exp} (Fig. 4-(g)). This observation is qualitatively confirmed by the many-particle theory (inset), which could trace this correlation back to nanoscale variations of the strength of interlayer hybridization in the WSe_2 - MoS_2 bilayers. This results in a changed efficiency for the phonon-driven charge transfer to the intermediate hybridized states and thus has a direct impact on the formation time of interlayer excitons.

C. Atomic relaxation and its influence on moiré excitons

The emergence of two-dimensional van der Waals materials has raised the intriguing idea of mixing and matching layers of different materials to realize novel properties in a given stack very early on. With the rush to build new Lego-like heterostructures in which to combine the vastly different electronic and magnetic ground states (Geim and Grigorieva, 2013), two fundamental real

space properties of van der Waals materials have been predominantly ignored: (i) their lattice constants are extremely dependent on local changes of composition and (ii) the atomistic positions in van der Waals materials are not rigid, but rather follow a membrane-like behavior. The second point becomes most prominent in large moiré unit cells formed by small twist angles between adjacent layers or by small lattice constant mismatches in heterostructures. In this scenario, the relaxation of atomistic positions in and out of the plane can minimize the energy of the system, and the reconstructed atomistic registry might be very different from that expected for unrelaxed, rigid heterobilayers with severe consequences for electronic, excitonic and mechanical properties (Carr *et al.*, 2018a; Halbertal *et al.*, 2022; Susarla *et al.*, 2022). Indeed, today, it is known that atomic corrugation plays a crucial role in the celebrated flat bands and, in particular, the energy gaps of twisted bilayer graphene at the magic angle (Lucignano *et al.*, 2019; Uchida *et al.*, 2014). Recently, even the role of moiré phonons, as vibrational stacking re-arrangements, have been highlighted as an unconventional way to boost exciton diffusion (Rossi *et al.*, 2023). Thus, it is fundamental to characterize experimentally the local stacking arrangements. Piezoresponse force microscopy (PPM) is an extension of atomic force microscopy that has emerged as a universal and easily implemented technique to map the real space variation of the moiré potential (McGilly *et al.*, 2020). PPM measures the amplitude and phase of the electromechanical response of a material to local deformations induced by an AC bias between its surface and the AFM tip. PPM maps the moiré superlattice by recording the changes in the flexoelectric response of heterobilayers associated with the inherent strain gradients of moiré heterostructures. This manifestation of the moiré potential as polar domain walls in a non-polar background is irrespective of the inversion symmetry of the constituent mono-layers (McGilly *et al.*, 2020).

While scanning tunneling microscopy and spectroscopy experiments (STM/S) lack the momentum resolution to resolve the reciprocal space information of the electronic band structure of moiré heterobilayers, STM/S has emerged as the only techniques with the required atomic and energy resolution to observe changes to the excitonic landscape due to atomic relaxation. For the STM/STS technique and its application to quantum materials, we refer the reader to other previous reviews (Bian *et al.*, 2021; Yin *et al.*, 2021). Early STM results on moiré heterostructures including Ref. (Li *et al.*, 2021a; Zhang *et al.*, 2017, 2020b) were reviewed in Ref. (Huang *et al.*, 2022). We now discuss examples of the effects of local strain in the electronic structure of moiré heterobilayers. In small-angle heterostructures of $\text{WSe}_2/\text{MoSe}_2$ STM experiments revealed spatially varied moiré period up ranging from 5 to 17nm wavelengths over a $500 \times 500 \text{nm}^2$ (Shabani *et al.*, 2021). The resulting strain associated

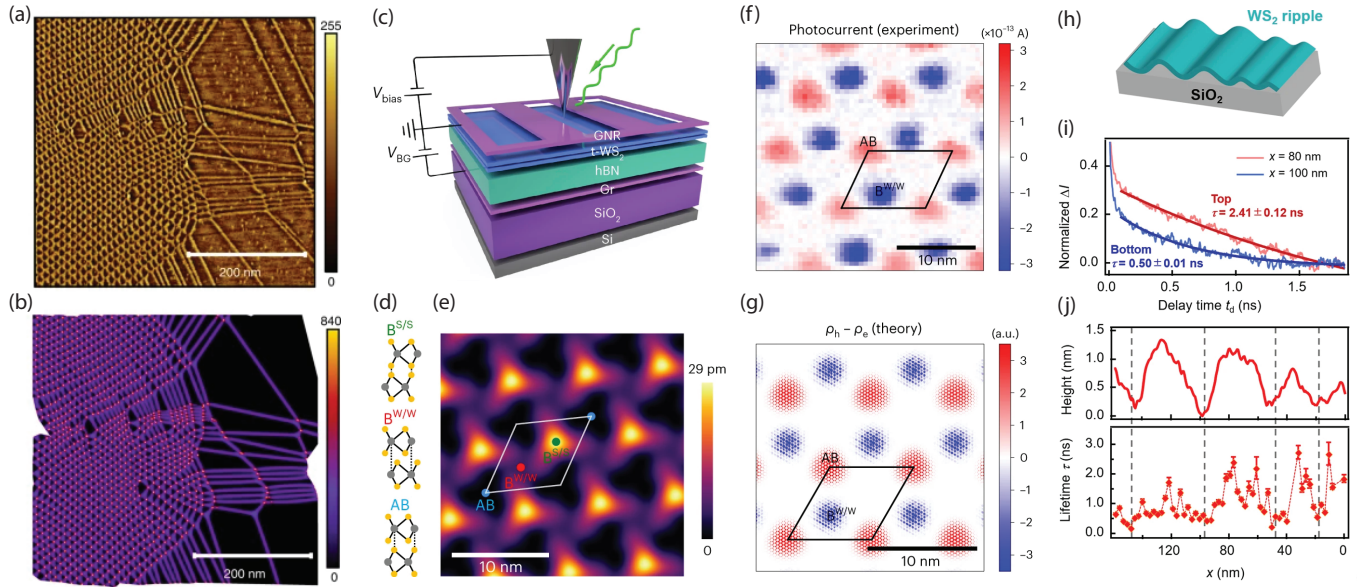


FIG. 5 (a) Large STM topography in a 1.7° $\text{WSe}_2/\text{MoSe}_2$ heterostructure showing a spatially dependent moiré periodicity. (b) Calculation of the stacking energies for the relaxed atomic energy reflecting the relation between the moiré pattern and strain. (c) Schematic of the laser-STM to measure a near- 58° twisted WS_2 bilayer sample. A bias between the STM tip and the sample in the presence of a 520 nm continuous wave laser enables measurements of the photocurrent with near-atomic resolution. (d) Illustration of the possible local stacking structures in twisted WS_2 . (e) Calculated topographic variation twisted WS_2 bilayers at $B^{W/W}$ and AB stacking sites. (f) Comparison of the experimentally observed spatial dependence of the charge transfer excitonic photocurrent sign and (g) GW-Bethe-Salpeter equation calculations. (h)-(j) Intralayer exciton lifetime dependence at the top and bottom of a strain induced ripple in WS_2 . Adapted with permission from (Li *et al.*, 2024; Mogi *et al.*, 2022; Shabani *et al.*, 2021)

with the moiré structure induces systematic large variations on the moiré potential depths and local changes to the near E_F states, see Fig. 5. In 3° twisted bilayers of WS_2 STS revealed the existence of flat bands associated with localized states that cannot be accounted for with first-principle calculations, but that can be recovered when the atomic displacements due to strain are included (Molino *et al.*, 2023). STM has also been deployed to observe changes of more than 10% in the moiré period of heterostructures of MBE grown multilayer PdTe_2 on Bi_2Se_3 (Figure 6) (Halbortal *et al.*, 2023).

In studying intra- and interlayer moiré excitons, it is also vital to understand and characterize the precise atomistic registry as it will modify the specific interactions and relaxation pathways. Laser-STM experiments in which STM/STS measurements are performed while a continuous-wave laser shines on the sample, enabling resolving the electronic structure of moiré excitons with sub-nm resolution. Laser-STM experiments in twisted bilayer WS_2 revealed nanoscale modulation in the moiré excitons (Li *et al.*, 2024) (Figure 5). The STM topography maps in these heterostructures are characterized by a moiré pattern with a 9 nm period. STS measurements show clear differences in the electronic band structure at $B^{W/W}$ and AB stacking sites reflecting the deep moiré potential in twisted WS_2 . Upon turning the laser on, the emerging excitonic photocurrent displays different

behavior for both sites. When the bias voltage between tip and sample is set in the region $-2V < V_{bias} < 1V$, the photocurrent is concomitantly positive in the $B^{W/W}$ sites and a negative in the AB stacking sites (Figure 5), indicating a dominant electron and hole contribution respectively. This phenomenology was explained by GW-Bethe-Salpeter equation calculations, including the moiré potential and electron-hole Coulomb interactions, which indicate the formation of in-plane charge transfer (ICT) moiré excitons. Thus, the STM tip-position photocurrent can be explained as a result of the lateral separation of electrons and holes in an ICT excitons (Li *et al.*, 2024).

Optical pump-probe STM setups with picosecond resolution could enable studying the atomic dependence of exciton dynamics (Liang *et al.*, 2023). The challenge in tr-STM measurements is to remove thermal fluctuations associated with the optical pump-induced expansion of the atomic tip and sample, which can result in large artifacts in the tunneling currents. The development of shaken-pulse-pair-excited STM, in which the pump beam is electronically modulated by an external trigger acting as the reference signal for lock-in amplifier detection (Mogi *et al.*, 2019; Takeuchi *et al.*, 2002), has enabled the required high-signal-to-noise ratios and time resolution to study non-equilibrium dynamics with atomic resolution (Iwaya *et al.*, 2023). Recently, tr-STM has been

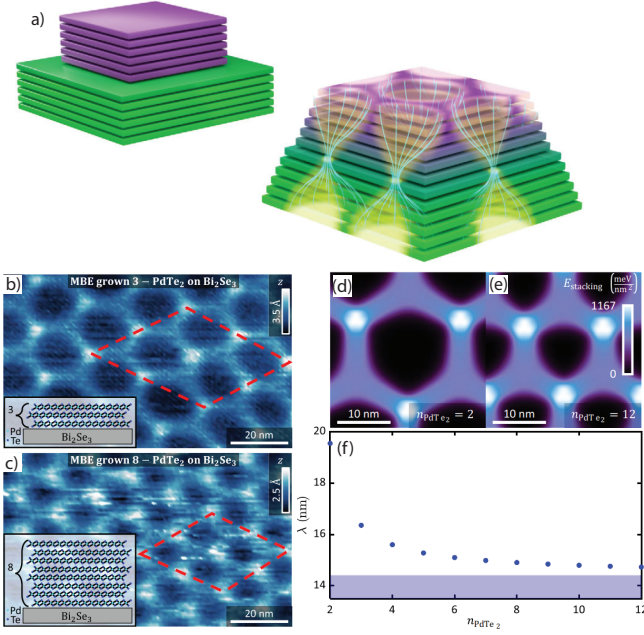


FIG. 6 (a) Schematic comparison of the lego-like and continuous layer-dependent interpretation of 2D heterostructures. (b)-(c) STM topography of 3 and 8 ML, respectively, of PdTe₂ on Bi₂Se₃ heterostructures. (d)-(e) Corresponding calculated moiré pattern after. (f) Calculated moiré potential wavelength dependence on the number of PdTe₂. Adapted with permission from (Halbertal *et al.*, 2023)

deployed to study the spatial dependence exciton dynamics in 7.6° heterobilayers of WS₂/WSe₂ (Mogi *et al.*, 2022) near grain boundaries and strain-induced ripples (see Figure 5). The authors revealed a $\times 10$ larger exciton lifetime between the top and bottom of the ripple, suggesting that interaction with the substrate phonon modes can lead to shorter excitonic lifetimes. Similarly, substrate-induced strain near grain boundaries also induced faster excitonic relaxation. The effect diminishes rapidly in space, with measurements 8 nm away from the grain boundary, where long exciton lifetimes are recovered.

Theoretical modeling of the atomic relaxation requires taking the multi-layered nature of vdW heterostructures into account properly, as it has been shown that the strain fields emerging from moiré defects can penetrate deep into a vdW material stack (Cook *et al.*, 2022; Halbertal *et al.*, 2023). However, a theoretical description of vdW heterostructures with large moiré periods and/or many layers on the atomic scale is computationally out of reach, and as a consequence, sophisticated coarse-grained models were developed that address atomic relaxation at large length scales. If the moiré lattice spans many unit cells of a single layer, the local atomic stacking registry and in- and out-of-plane strain fields, can be described by generalized forces acting on a stack of membranes (i.e., one models each single layer as one of these

membranes) (Carr *et al.*, 2020, 2018b; Cazeaux *et al.*, 2020; Halbertal *et al.*, 2021, 2023; Zhu *et al.*, 2020). In such membrane models, each layer of the vdW stack is allowed to relax individually. The total energy functional is then a sum of the generalized stacking fault energy, which accounts for lattice mismatch at any of the stack’s interfaces, and the elastic energy, which accounts for the in- and out-of-plane strain of each of the membranes. The parameters for both the elastic and the stacking fault energy are readily obtained from *ab-initio* simulations of the individual constituent layers and interfaces (Halbertal *et al.*, 2023). The multi-layered relaxation process in vdW heterostructures can heavily affect electronic and, thus, excitonic properties. Therefore, the theoretical modeling of relaxation, e.g., by the aforementioned membrane models, is an essential ingredient also in describing these properties in moiré structures even far away from the interface (Halbertal *et al.*, 2023; Mullan *et al.*, 2023).

These experimental and theoretical studies exemplify that it is essential to understand the relaxation mechanisms of moiré heterostructures toward fully understanding their emergent excitonic properties. In a second step one might consider using long-ranged relaxation even for engineering-by-stacking purposes.

III. OUTLOOK

New applications are emerging as our understanding of moiré physics progresses. In the following, we highlight a few recent applications:

Optics, photonics, and optoelectronic devices: moiré excitons have been shown to play a pivotal role in developing tunable optoelectronic devices across various heterostructures and materials (Behura *et al.*, 2021; Brem *et al.*, 2020; Guo *et al.*, 2021; Liu *et al.*, 2022). Moiré excitons serve as a platform for optoelectronic modulation, offering efficient control over light absorption, emission, and transmission essential for devices like optical switches, photodetectors, and modulators, ensuring high-speed operation and low energy consumption (Ramos *et al.*, 2022; Yu *et al.*, 2017). For example, moiré excitons enhance device efficiency in WS₂/graphene heterostructures by promoting efficient carrier extraction and utilization with high sensitivity and signal-to-noise ratios (Trovatello *et al.*, 2022). Integration of moiré excitons into integrated photonic circuits enables the development of compact, scalable systems for telecommunications, sensing, and quantum information processing. Specifically, MoS₂/FePS₃ heterostructures have potential for new integrated photonics (Duan *et al.*, 2022), offering multifunctional devices with advanced performance and functionality (Onga *et al.*, 2020). Novel excitonic devices exploiting strong light-matter interactions and long-range effects will result in novel excitonic transis-

tors, photodetectors, LEDs, and solar cells with enhanced efficiency and tunability.

Quantum Information Processing: The long coherence times and strong interactions of moiré excitons position moiré devices as promising candidates for quantum information processing (Huang *et al.*, 2022). Moreover, moiré excitons serve as valuable sources of quantum light, capable of generating non-classical light states like single photons and entangled photon pairs (Baek *et al.*, 2020; Yu *et al.*, 2017). Researchers aim to leverage these characteristics for quantum computing, communication, and cryptography, harnessing coherence and entanglement properties to encode and process quantum information effectively. Their utilization in semiconductor moiré superlattices, particularly in Kekulé/moiré superlattices, offers new prospects for practical quantum devices (Ye *et al.*, 2023). Additionally, moiré excitons show potential in magnetic field sensing applications, especially in antiferromagnet-semiconductor heterostructures (Onga *et al.*, 2020). Their sensitivity to external magnetic fields enables high-resolution and sensitive detection, making them suitable for various magnetic imaging, navigation, and magnetometry applications.

Other devices and applications: moiré excitons offer significant potential for enhancing the efficiency and functionality of various energy and detection devices (Ciarrocchi *et al.*, 2022; Du *et al.*, 2023a). Their strong light-matter interaction in energy harvesting and photovoltaics allows for efficient photon absorption, leading to higher photoconversion efficiencies (Duan *et al.*, 2022). Additionally, their tunability enables optimization across different wavelengths, making them suitable for diverse photonic applications. In broadband photodetection, MoSe₂/FePS₃ heterostructures demonstrate a broad spectral response, enhancing versatility for applications like imaging and spectroscopy (Onga *et al.*, 2020). These photodetectors operate self-driven due to efficient carrier generation and transport, resulting in low power consumption and enhanced portability. Moreover, moiré exciton-based photodetectors exhibit fast response times and low noise performance, crucial for high-speed data acquisition and sensitive detection of weak optical signals in noisy environments. Enhanced light-matter interaction in moiré excitons, particularly in Kekulé/moiré superlattices and 2D TMD materials, improves optical absorption and emission properties, benefiting applications like optical sensing and imaging (Ye *et al.*, 2023). Furthermore, moiré excitons facilitate efficient hot carrier transfer in WS₂/graphene heterostructures, enabling rapid carrier dynamics and high-speed device operation essential for photodetectors and communication systems.

These applications and recent understanding of the spatial variation of moiré require a new paradigm for understanding the effects of spatial inhomogeneities and atomic relaxation on the electronic properties of moiré devices. A tandem effort of new advanced characteri-

zation techniques and new machinery to calculate the structural variation of the moiré potential and collective excitations is needed towards new excitonic devices. For an extensive review of the applications of moiré optoelectronics we refer the reader to Ref. (Du *et al.*, 2023b) and Ref. (Ciarrocchi *et al.*, 2022).

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