Using a multiscale computational approach, we probe the origin and evolution of ultraflatbands in moiré superlattices of twisted bilayer MoS$_2$ (TBLM), a prototypical transition metal dichalcogenide. Unlike twisted bilayer graphene, we find no special magic angles in TBLM. Ultraflatbands, which form at the valence band edge for twist angles ($\theta$) close to 0$^\circ$ and at both the valence and conduction band edges for $\theta$ close to 60$^\circ$, have distinct origins. For $\theta$ close to 0$^\circ$, inhomogeneous hybridization in the relaxed moiré superlattice is sufficient to explain the formation of flatbands. On the other hand, for $\theta$ close to 60$^\circ$, local strains cause the formation of modulating triangular potential wells such that electrons and holes are spatially separated. This leads to multiple energy-separated ultraflatbands closely resembling eigenfunctions of a quantum particle in an equilateral triangle well.

Correlated insulating behaviour and unconventional superconductivity was recently observed in twisted bilayer graphene (TBLG) at a 'magic' angle of 1.1$^\circ$ [1–3]. While the nature of superconductivity is still contested, formation of ultraflatbands near the Fermi level at this angle is essential to understanding these phenomena [4–11]. Since this discovery, ultraflatbands have been predicted in other twisted 2D materials [12–18] including small angle twisted bilayer MoS$_2$ (TBLM), a prototypical transition metal dichalcogenide (TMD) [19, 20]. For TBLG, the bands flatten in a narrow range of 0.1$^\circ$ about 1.1$^\circ$ [21, 22], making their experimental realisation challenging. The existence or absence of similar unique 'magic' angles in TBLM has not been explored. Ultraflatbands and localization also has significant implications on excitonic properties of the material [23, 24].

Properties of TBLM are remarkably different from TBLG. Due to sublattice symmetry breaking, TBLM forms distinct moiré patterns for twist angles close to 0$^\circ$ and 60$^\circ$ [19]. The sliding potential energy landscape is more corrugated in TBLM compared to TBLG leading to larger deformation of the moiré superlattice (MSL) in TBLM [25–27]. These deformations, distinct for twist angles close to 0$^\circ$ and 60$^\circ$, involve a change in the distribution of stackings and interlayer spacings from the rigidly twisted structure [19, 27, 28]. This influences the local electronic hybridization between the layers and controls the localization of the flatbands [19]. In TBLG, on the other hand, localization at the magic twist angle is largely independent of lattice relaxations [6]. The change in band structure and band width due to relaxation is minor compared to TBLM [25, 20, 52]. The origin of the flatbands in TBLM is thus expected to be different from TBLG.

Quantum dots using 2D materials have several potential applications including quantum emission, design of solar cells and photocatalysis [33–38]. The current route to obtain quantum dots is through preparation of a colloidal suspension of 2D material flakes [34, 36, 69]. This leads to poor control over the size and shape of quantum dots [37]. Obtaining quantum dot arrays in a dry and systematic manner has been a challenge [59]. While the possibility of obtaining quantum dots in moiré patterns of twisted bilayers has been proposed [35, 19, 41], explicit predictive calculations on the MSL including crucial atomic relaxation effects are lacking. Moreover, signa-
tures of trapped excitons in moiré superlattices have been recently observed in photoluminescence experiments [42–44], indicating the presence of localised states.

In this letter, we use an efficient multiscale approach to study the evolution and origin of ultraflatbands in TBLM. We establish that unlike TBLG, there are no regions in the MSL. Wavefunctions and degeneracies of the ultraflatbands at the valence band edge are in excellent agreement with states of a particle in an infinite equilateral triangle potential well (IETPW). The wavefunctions at the conduction band edge also resemble IETPW states, but the degeneracies are modified by valley degeneracies in the unit-cell Brillouin zone. The ultraflatbands form due to two factors: 1) inhomogeneity in the interlayer hybridization in the MSL due to the distribution of stackings and interlayer spacing (ILS), and 2) local strains due to soliton formation. We would also like to point out that neglecting lattice relaxations in the study of TMD MSL leads to spurious flatbands and localizations (see supplementary materials (SM) [45]).

The MSL are constructed using the Twister code [46]. Structural relaxations are performed using classical force-field calculations as implemented in the LAMMPS package [47, 48]. The intralayer forces are described using Stilinger-Weber [19, 50] forcefield. The interlayer interactions are taken into account using the recently parametrized Kolmogorov-Crespi [51] potential which is shown to accurately capture relaxations in TMD MSL [52]. The electronic structure calculations are performed on the relaxed MSL using density functional theory (DFT) calculations with the SIESTA [53] package. The DFT wavefunctions are expanded in a double-$\zeta$ plus polarization basis. Norm-conserving pseudopotentials [54] and the local density approximation to the exchange-correlation functional are employed (see SM for further details [45]).

TBLM is composed of distinct high-symmetry stackings for twist angles close to $0^\circ$ (AA, $B^{Mo/5}$ and $B^{S/Mo}$ stackings) and $60^\circ$ (AB, $B^{S/S}$ and $B^{Mo/Mo}$) [19]. Relaxations in TBLM arise from competition between in-plane strain cost and stacking energy gain [19, 26–28]. The low-energy stacking regions in the MSL are separated by domain walls (solitons) [55] that hold the shear strain built up in the system [19, 28] (Fig. 1). Furthermore, atomic displacements in the out-of-plane direction lead to an undulating ILS in the MSL. [19, 27]. The stacking and interlayer spacing distribution are shown in the SM [45]. For twist angles close to $0^\circ$, the $B^{Mo/S}$ and $B^{S/Mo}$ regions are equal in area since they are the same energy and lead to a six-fold symmetry around the AA stacking region (similar to TBLG). For angles close to $60^\circ$, the AB stacking is lower in energy than $B^{Mo/Mo}$ [27, 28]. Hence the area of AB stacking in the MSL is larger and leads to a reduced three-fold symmetry around $B^{S/S}$ [27].

Construction of a MSL by introducing a twist is accompanied by shrinking of the unit-cell Brillouin zone (UBZ). The bands from the UBZ of the two layers fold into the moiré BZ (MBZ), Fig. 1 (a) shows the band structure of 2.65° TBLM. The bands of TBLM are flatter than those of AB stacking of same supercell size. The valence bands, v1 and v2 in inset of Fig. 1 (a) are degenerate at the K point of the MBZ corresponding to the symmetry of the underlying lattice. This degeneracy at the K point is present in all twist angles close to $0^\circ$ in our study. Application of an external uniaxial strain can break this symmetry and degeneracy (see SM [45]). The wavefunctions of the v1 and v2 states are localised at the $B^{Mo/S}$ and $B^{S/Mo}$ regions (Fig. 1 (b)). This localization occurs due to inhomogeneous hybridization between the two layers in the MSL. As discussed previously [19], this can be qualitatively understood in terms of ordering of the VBM with respect to the vacuum level amongst different high-symmetry stackings (see SM [45]). The conduction band edge lines up among the stackings [19] and provides no hint at a preferred localisation site. The inhomogeneous hybridization in the MSL can be captured by a map of the local barrier potential in the MSL, $V_{barr}(x,y)$ [45, 56]. Macroscopic averaging is performed in a unit-cell around each Mo atom of the bottom layer to remove atomic-scale oscillations. Smaller the potential barrier, larger the interlayer hybridization. This shows (Fig. 1 (d)) larger hybridization at the $B^{Mo/S}$ and $B^{S/Mo}$ regions.

In $58^\circ$ TBLM, several ultraflatbands form at the valence and conduction band edges (Fig. 2 (a)) and are well separated in energy. From hybridization considerations, as discussed above, we expect the valence band edge states to localise at the AB and $B^{Mo/Mo}$ sites (see Fig. 2 (d)), and no localisation at the conduction band edge. However, the valence band edge states localise at the AB stacking alone and conduction band edge states at $B^{Mo/Mo}$ (Fig. 2 (b) and (c)). We find that an additional modulating potential is introduced in the MSL (Fig. 2 (c)) for twist angles close to $60^\circ$. To calculate the modulating potential, we first average the DFT potential in a slab of length 17 Å in the out-of-plane direction containing the bilayer, to obtain $V_{MSL}(x,y)$. We then macroscopic average $V_{MSL}(x,y)$, as discussed above, to obtain $V_{barr}(x,y)$, which confines the electrons (Fig. 2 (c)), and a maximum at $B^{S/S}$ and AB. The hybridization
FIG. 2. (a) Band structure of 58° TBLM showing the multiple energy-separated ultraflatbands. The inset shows the enlarged version of the valence and conduction band edges. (b) Distribution, $|\psi(r)|^2$, the states labeled in (a) averaged in the out-of-plane ($z$) direction. The corresponding equilateral triangle quantum well wavefunctions of the ground state and first five excited states are also shown. (c) $\Delta V(x_{Mo}, y_{Mo})$, for 58° TBLM. (d) Local potential barrier distribution (in eV) between the layers, $V_{bar}(x_{Mo}, y_{Mo})$, in 58° TBLM. (e) $|\psi(r)|^2$ of the conduction states labeled in (a), c1-c4, averaged along the $z$-direction. The degeneracies of the wavefunctions in (b) and (e) are shown in brackets.

FIG. 3. (a) and (b) Modulating potential, $\Delta V(x_{Mo}, y_{Mo})$ (in eV), for 57.35° and 58.5° TBLM, respectively. (c) Evolution of the ultraflatbands in the energy range -4.5 and -3.35 eV, with respect to the vacuum level for twist angles 57.35°, 58.0 and 58.5°. The band gap is marked by green arrow and not in scale. (d) and (e) $V_{bar}(x_{Mo}, y_{Mo})$ (in eV) for 57.35° and 58.5° TBLM, respectively. (f) Evolution of the splittings between the first two states at the valence band edge, $\Delta E_{v1-v2}$, first ($\Delta E_{c1-c2}$) and second two states ($\Delta E_{c2-c3}$) at the conduction band edge. (g) and (h) Evolution of the density of states as a function of twist angles close to 0° and 60°, respectively.

rules out localisation at $B^{S/S}$, hence inducing the holes to localise at the AB stacking. The confining potential has the shape of an equilateral triangle (Fig. 2(c)). This is absent in twist angles close to 0° (Fig. 1(c)).

The spatial distribution of the wavefunctions can be understood in terms of IETPW. A particle in an IETPW (of side $a$) can be described using two quantum numbers, $p$ and $q$. The energies are given by $E_{p,q} = (p^2 + q^2 + pq)E_0$ [57], $q$ takes the values $0, 1/3, 2/3, 1, ..., p = q + 1, q + 2, q + 3, ...$. The ground state, $E_{1,0} = E_0 = \hbar^2 / 3ma^2$. 


The eigenfunctions can be further labelled by $A_1$, $A_2$ and $E$. Where the $A_1$ and $A_2$ states are singly degenerate, while the $E$ states are doubly degenerate [57]. In $58^\circ$ TBLM, the wavefunctions and degeneracies of the ultraflatbands of the first six states close to the valence band edge are in excellent agreement with those of the ground state and first five excited states of the IETPW (Fig. 2 (b)). Similarly the conduction band edge states also agree well with the IETPW, however the degeneracies are intriguing. $c_1$ and $c_2$ both are doubly degenerate states, while the $c_3$ state is four-fold degenerate. The wavefunctions and degeneracies of the IETPW are hence unaffected and follow those of the IETPW. We should note that in contrast to the ideal infinite IETPW, the potential is periodic and of finite depth. We thus expect only a few states close to the valence and conduction band edge to be confined in a triangular region.

The evolution of the density of states (Fig. 3 (f) and (g)) shows the stark contrast in electronic structure between twist angles close to $60^\circ$ and close to $0^\circ$. As the twist angle approaches $60^\circ$, the AB stacking area grows larger than the other stackings (Fig. 3 (d) and (e)) [27]. As the area of confinement of holes increases, the spacing between the flatbands close to valence band edge decreases as shown in Fig. 3 (c) and (f). While the area of $B_{Mo/Mo}$ region does not grow appreciably as the twist angle approaches $60^\circ$ [27], the confining potential depth increases (Fig. 3 (a) and (b)). The spacings between levels at the conduction band edge are thus relatively unaffected (Fig. 3 (c) and (f)). The flatbands and the band gap is thus tunable with twist angle. This also indicates the absence of special "magic" angles for flatband formation in TBLM [55].

To understand the origin of the confining triangular potential in twist angles close to $60^\circ$ we probe the role of local strains [45] in the MSL. The strain is localised at the shear soliton regions in each layer (Fig. 4 (a) and (b)). We can construct a strain-free MSL by allowing atomic relaxations only in the out-of-plane direction (from the rigidly twisted MSL). The ILS is allowed to vary in this procedure (Fig. 4 (c) and (d)), hence the ordering of the VBM among the stackings is unaffected. We find that in this structure the multiple energy-separated ultraflatbands in $57.35^\circ$ vanish (Fig. 4 (f)). The conduction bands are delocalised due to the absence of a modulating potential well and the valence bands are localised at $B_{Mo/Mo}$ and AB sites (Fig. 4 (h)), as expected from the hybridization arguments. $V_{bare}(x_{Mo}, y_{Mo})$ and $\Delta V(x_{Mo}, y_{Mo})$ for this structure are shown in the SM [45]. This clearly establishes local strains as the driving mechanism for formation of the modulating potential well, which in turn determines localisation. The localisation in $2.65^\circ$ MSL, on the other hand, is not affected by removal of the strains (Fig. 4 (e) and (g)).

In conclusion, we have demonstrated the formation of an array of triangular quantum dots in MSL of TBLM with twist angles close to $60^\circ$. The holes and electrons are spatially separated which could lead to long-lifetime confined excitons in this system. Unlike TBLG, no special 'magic' angles exist in TBLM for the formation of

![Fig. 4](image-url)
ultraflatbands. This makes it easier to experimentally probe ultraflatbands in these systems. The contrasting electronic structure of twist angles close to 0° and 60° is due to an additional modulating confining potential in angles close to 60°. Local strains in the MSL due to soliton formation are responsible for the formation of this confining potential. External strains can thus be used to engineer the confining potential and flatbands in these systems. The effect of finite temperature on the soliton networks and their effect on the quantum dots, if any, is a worthwhile future direction.

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See Supplementary Materials for details regarding the commensurate superlattices and computational methods employed. Electronic structure of rigidly (unrelaxed) TBLM. Distribution of ILS and order-parameter in the relaxed MSL as a function of twist angle. VBM ordering and barrier potential in the isolated stackings. \( V_{\text{barr}}(x_{\text{Mo}}, y_{\text{Mo}}) \) and \( \Delta V(x_{\text{Mo}}, y_{\text{Mo}}) \) for contrain relaxed 57.35° TBLM.

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