Universal superlattice potential for 2D materials from twisted interface inside h-BN substrate

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Lateral superlattices in 2D materials provide a powerful platform for exploring intriguing quantum phenomena, which can be realized through the proximity coupling in forming moiré pattern with another layer. This approach, however, is invasive, material-specific, and requires small lattice mismatch and suitable band alignment, largely limited to graphene and transition metal dichalcogenides (TMDs). Hexagonal boron nitride (h-BN) of antiparallel (AA') stacking has been an indispensable building block, as dielectric substrates and capping layers for realizing high-quality van der Waals devices. There is also emerging interest on parallelly aligned h-BN of Bernal (AB) stacking, where the broken inversion and mirror symmetries lead to out-of-plane electrical polarization. Here we show the that laterally patterned electrical polarization at a nearly parallel interface within the h-BN substrate can be exploited to create noninvasively a universal superlattice potential in general 2D materials. The feasibility is demonstrated by first principle calculations for monolayer MoSe2, black phosphorus, and antiferromagnetic MnPSe3 on such h-BN. The potential strength can reach 200 meV, customizable in this range through choice of distance of target material from the interface in h-BN. We also find sizable out-of-plane electric field at the h-BN surface, which can realize superlattice potential for interlayer excitons in TMD bilayers as well as dipolar molecules. The idea is further generalized to AB-stacked h-BN subject to torsion with adjacent layers all twisted with an angle, which allows the potential and field strength to be scaled up with film thickness, saturating to a quasi-periodic one with chiral structure.

INTRODUCTION

Lateral superlattices in graphene and transition metal dichalcogenides (TMDs) are drawing remarkable interest as fascinating arena to explore emergent quantum transport1–4, quantum optical5, and many-body phenomena6–8. Such superlattices have been realized through forming long-wave moiré pattern, where the registry-dependent proximity coupling from adjacent layers periodically modulate the electronic structures. This approach, however, requires the building blocks to have small lattice mismatch and suitable band alignment, and its implementation has so far been limited to graphene and TMDs. The resultant moiré superlattices are material-specific, and also dependent on the carrier species and internal degrees of freedom. Taking TMDs heterobilayers as example, the electrons at K and Q valleys at K and Γ points all experience distinct moiré superlattice potentials, and the energy landscape depend on choices of compounds9–23. The proximity coupling at the moiré interface also degrade or change other material properties of the target layer. For example, the TMD heterointerface for realizing superlattice potential of electrons18–23 also results in ultrafast electron-hole separation that completely alters the optical properties5–8.

The realization of the elegant moiré superlattices and high-quality van der Waals devices has relied on the use of hexagonal boron nitride (h-BN) as substrate and capping layer16,17, for its excellent thermal and chemical stability, and insulating band structures. Commonly used h-BN has the antiparallel alignment between adjacent layers, known as the AA' stacking, where boron atoms are vertically aligned with nitrogen atoms of neighboring layers. Another energetically favorable h-BN structure is to have the layers parallelly aligned with the AB (Bernal) stacking, and growth of such AB h-BN has been achieved using chemical vapor deposition28. Unlike the AA' one, the AB stacking allows the emergence of perpendicular electric polarization since the restriction from inversion symmetry and out-of-plane mirror symmetry are both lifted29. Observations of ferroelectricity in AB-stacked h-BN are reported very recently: under marginally twisting where structure relaxation leads to micron or sub-micron scale alternating AB and BA stacking domains, the opposite out-of-plane electrical polarizations are sensed by Kelvin probe force microscope30,31, and by the resistance of adjacent graphene32.

Here we propose a noninvasive approach to realize universal superlattice potential on a general 2D material, exploiting the electrical polarization at a nearly parallel interface inside the h-BN substrate or capping layer. Such interface can be readily created by stacking a n-layer AA' h-BN on another AA' film (Fig. 1), which hosts electrical polarization patterned with periodicity b = a/δ, a the lattice constant of h-BN and δ the twist angle from parallel. A triangular superlattice potential with period b is then electrostatically generated in the target 2D material placed on h-BN surface, regardless of its crystal structure and orientation. This is corroborated by our first principle calculations for monolayers of MoSe2, black phosphorus, and antiferromagnetic MnPSe3 on h-BN substrates. The potential strength is proportional to \(\exp(-nd\,\alpha\,n_d)\), \(nd\) the vertical distance of target material from the interface in h-BN, and can be customized in the range 0–200 meV. This rapid exponential dependence corresponds to sizable out-of-plane electric field \(-0.6V/b\), which can realize superlattice potential for dipolar objects at the h-BN surface, such as interlayer excitons in TMDs bilayers, or adsorbed dipolar molecules. Using AB h-BN film subject to torsion where adjacent

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layers are all twisted with a common angle $\delta$, the strengths of potential and electric field on the surface can be further scaled up with film thickness $N$, which saturates to a quasi-periodic form with a chiral structure when $N\delta$ becomes sufficiently large.

RESULTS

Electrostatic potential from a twisted interface in h-BN

We first examine the electrical polarization at a nearly parallel interface in h-BN, with twist angle $\delta < \pi$ but beyond the marginally twisting regime addressed in refs. 30–32 such that structural relaxation is not significant. This allows us to focus on a moiré pattern between rigid lattices at the nearly parallel interface. The local stacking registry can be characterized by the in-plane displacement $r$ between two near-neighbor B atoms across the interface (Fig. 2a), which is a linear function of the lateral position $R$. Having a few-layer h-BN to separate this interface from the surface may also help to stabilize the moiré from structural relaxation.

The nearly parallel interface is now described by a long-wavelength moiré pattern $r(R)$, with period $b > a$. There exists an intermediate length scale ($l$), large compared to lattice constant $a$ and small compared to $b$, such that the atomic registry in any region of size $l$ closely resembles a lattice-matched stacking, so the local electronic structures in moiré can be well approximated by band structures of the latter. First-principles calculations of various lattice-matched stacking can give the electrical polarization $P$ as function of registry $r$. Combined with the mapping between $r$ and location $R$ in the long-wavelength moiré, the laterally modulated electrical polarization can then be given as $P(R) \equiv P(r(R))$.

We start with first principle calculations of $n + n$ multilayers where the top $n$-layer AA’ h-BN is stacked on the bottom $n$-layer AA’ h-BN, with a lattice-matched parallel interface of various registry $r$. Figure 2b shows an example of the charge redistribution caused by the interlayer coupling at the parallel interface with $r = a/\sqrt{3}$ (i.e., an AB interface). The presented quantity is the differential charge $\Delta \rho \equiv \rho_{n+n} - \rho_{n} - \rho_{n+n}$, where $\rho_{n+n}$ is the DFT-calculated plane-averaged charge density of the $n + n$ multilayer, and $\rho_{n}$ is that of the pristine top (bottom) $n$-layer. Remarkably, for various thickness $n$ considered, $\Delta \rho$ is predominantly distributed with nearly the same profile on the two layers constituting the parallel interface. This shows that the charge redistribution is occurring at the parallel interface only, and the outer layers in antiparallel alignment only has very small effect on the resultant electrical polarization.

In the lattice-matched stacking, the spatially uniform electrical polarization corresponds to a voltage drop: $\Delta \phi = \frac{1}{C_0} \rho$, which can be directly extracted from the DFT calculations as the difference between the vacuum levels on the two side of the multilayer. The electrical polarization determined from this vacuum level difference, i.e., $\varepsilon_0 \Delta \rho$, is shown in Fig. 2c (triangle symbols). The magnitude of $P$ is $2\text{pC m}^{-1}$, consistent with other calculations for AB bilayers 29, while the modest dependence on the layer thickness $n$ can be attributed to the small screening effect by the outer layers. Through the induced carrier density change in an adjacent graphene layer, the experiment in ref. 32 has determined an electrical polarization of $P \sim 2.25 \text{pC m}^{-1}$ in AB-stacked bilayer h-BN, in good agreement with the calculations. Figure 2c also shows the electrical polarization evaluated from the differential charge, $P = \int z \Delta \rho(x) dx$ (star symbols), which is consistent with the $\varepsilon_0 \Delta \phi$ values.

Figure 2d plots the electrical polarization as function of $r$ in a $2 + 2$ configuration, $r = 2a/\sqrt{3}$ corresponds to an AB stacking interface, where $P$ is exactly the opposite of the AB stacking one. For the AA stacking ($r = 0$), the out-of-plane mirror symmetry forbids any polarization. In general, $r$ stacking registry is related to the $-r$ one by a spatial inversion, so $P$ is an odd function of $r$. Like many other quantities involving the interlayer processes 25,35, the registry dependence can be well fitted by the lowest Fourier harmonics observing the rotational and translational symmetry,

$$
P(r) = \frac{P_0}{2} \left( f_+(r) - f_-(r) \right),
$$

where $K$ is wavevector at the Brillouin zone corner. In Fig. 2d, the dots are the polarization calculated from $\Delta \rho$ under various $r$, while the curve is a plot of Eq. (1), showing excellent agreement.

For the twisted interface, $\Delta \rho$ and $P$ as functions of $r$ can then be mapped to the dependences of these quantities on location $R$, by...
The approximation in Eq. (2) holds well under the condition \( \Delta \phi \equiv \Delta \phi_r \equiv \rho_{bn} \), where \( \rho_{bn} \) is the plane-averaged charge density of the \( n + n \) multilayer calculated using DFT, and \( \rho_{bn} \) is that of the pristine top/bottom \( n \)-layer. Dashed vertical lines indicate atomic planes. For various thickness \( n \) considered, \( \Delta \phi \) is predominantly distributed with nearly the same profile on the two layers forming the interface. Electrical polarization in the \( n + n \) multilayers, evaluated using \( \int z \Delta \phi_r (z) I dz \) (stars), and using \( \epsilon_0 \phi \Delta \phi \) (triangles), \( \Delta \phi \) being the vacuum level difference on the two sides of multilayer directly extracted from DFT. \( \mathbf{P} \) as a function of \( r \) in the \( 2 + 2 \) configuration. The dots denote the values from the DFT-calculated \( \Delta \phi \), and the curve is the fitting using Eq. (1). Lateral modulated polarization \( \mathbf{P}(\mathbf{R}) \) when the interface has a rigid twist from parallel by small angle \( \delta \), and the resultant electrostatic superlattice potential \( \Delta \phi(\mathbf{R}, z) \) on constant \( z \) plane (from Eq. (2), c.f. text). \( \Delta \phi(\mathbf{R}, z) \) is shown at three vertical distance from the interface which, for the example of \( \delta = 1.5^\circ \) (period \( b = 9.6 \) nm), corresponding to thickness of \( n = 2, 4, \) and 6 layers, respectively.

The thin limit \( G |z| \approx 1 \), the potential strength measured by \( V_{\text{max}} - V_{\text{min}} \) reaches 203 meV.

When a general 2D material is placed on the h-BN surface, the electrons/holes experience the electrostatic potential through their charge, which shall produce a universal superlattice energy landscape determined by the h-BN interface, regardless of the crystal type and orientation of the target materials. This is corroborated by our first principle calculations of the band-edge energies in three distinct examples, i.e., MoSe\(_2\), black phosphorus (BP), and the antiferromagnetic MnPSe\(_3\) monolayers.

Figure 3a, b shows the DFT-calculated band edges, in a monolayer MoSe\(_2\) placed on an h-BN bilayer, as functions of stacking registries. In these calculations, the MoSe\(_2\) has the crystal type and orientation of the target materials. This is corroborated by our first principle calculations of the band-edge energies in three distinct examples, i.e., MoSe\(_2\), black phosphorus (BP), and the antiferromagnetic MnPSe\(_3\).
the superlattice energy landscape for electrons in MoSe₂, which has the same profile for K, Q, and Γ valleys.

It is generally expected that the heterointerface between h-BN and target material such as MoSe₂ does not change the energy landscape of the latter, which underlies the extensive use of h-BN as substrate and capping layer. This is confirmed in our calculations, as shown in Fig. 3b, where the band-edge energies show no visible dependences on the stacking registry \( r_{bn} \) between MoSe₂ and h-BN. It is worth noting that there is a charge transfer between MoSe₂ and h-BN (c.f. Supplementary Note 3), but its magnitude of the modulation, i.e., \( E_{\text{max}} - E_{\text{min}} \), is 193 meV for K and Q valleys, and 202 meV for Γ point. The dependence on \( r_{bn} \) is also visible in Fig. 3a, where the band-edge energies of Q and Γ valleys show the same behaviors as the CBM and VBM @ K valleys. The energies of Q and Γ valleys are similar to those of MoSe₂ on a bilayer h-BN substrate. We have also performed calculations for a different angular alignment between MoSe₂ and h-BN that forms \( \sqrt{7} \times \sqrt{7} \) periodic cell, which points to the same conclusion (Supplementary Fig. 3).

Figure 3c, d shows the DFT-calculated CBM and VBM energies in monolayer MnPSe₃ with the Neel type antiferromagnetic order, placed on the bilayer h-BN substrate. Figure 3e, f is for BP monolayer on the bilayer h-BN substrate. We find that the band energies in these materials are also modulated with the same profile as functions of the stacking registry \( r_{bn} \), between the two parallel h-BN layers. These results are in full support of the expectation that the heterointerface between h-BN and target material such as MoSe₂ on a bilayer h-BN substrate. From DFT-calculated band structures, the energies of conduction band minima (CBM) and valence band maxima (VBM) are examined as functions of \( r_{bn} \) (registry between the two parallel BN layers, c.f. inset of a), and \( r_{t} \) (stacking registry between MoSe₂ and h-BN, c.f. inset of b). The expected modulation by the \( r_{bn} \)-dependent electrical polarization in h-BN is seen in a. The magnitude of the modulation, i.e., \( E_{\text{max}} - E_{\text{min}} \), is 193 meV for K and Q valleys, and 202 meV for Γ point. The dependence on \( r_{bn} \) is not visible in b. The energies of Q and Γ valleys show the same behaviors as the CBM and VBM @ K valleys. c, d and e, f are similar plots for MnPSe₃ and BP monolayer, respectively, on the bilayer h-BN substrate. In c, \( E_{\text{max}} - E_{\text{min}} \) is 233 meV for CBM and 223 meV for VBM. In e, \( E_{\text{max}} - E_{\text{min}} \) is 195 meV for CBM and 207 meV for VBM. See Supplementary Fig. 2 for more details.

**Fig. 3** Universal modulation of band-edge energies in target materials by the electrical polarization in h-BN substrate. a, b Monolayer MoSe₂ on a bilayer h-BN substrate. From DFT-calculated band structures, the energies of conduction band minima (CBM) and valence band maxima (VBM) in MoSe₂ are examined as functions of \( r_{bn} \) (registry between the two parallel BN layers, c.f. inset of a), and \( r_{t} \) (stacking registry between MoSe₂ and h-BN, c.f. inset of b). The expected modulation by the \( r_{bn} \)-dependent electrical polarization in h-BN is seen in a. The magnitude of the modulation, i.e., \( E_{\text{max}} - E_{\text{min}} \), is 193 meV for K and Q valleys, and 202 meV for Γ point. The dependence on \( r_{bn} \) is not visible in b. The energies of Q and Γ valleys show the same behaviors as the CBM and VBM @ K valleys. c, d and e, f are similar plots for MnPSe₃ and BP monolayer, respectively, on the bilayer h-BN substrate. In c, \( E_{\text{max}} - E_{\text{min}} \) is 233 meV for CBM and 223 meV for VBM. In e, \( E_{\text{max}} - E_{\text{min}} \) is 195 meV for CBM and 207 meV for VBM. See Supplementary Fig. 2 for more details.

**Multiple twisted parallel interfaces in h-BN**

We first consider an AB stacking trilayer, which has two parallel interfaces both with stacking registry \( r = a/\sqrt{3} \). The bottom panel of Fig. 4a presents the charge redistribution due to the interlayer coupling at both interfaces, \( \Delta \rho \equiv \rho_{2L} - \rho_{2M} - \rho_{2M} \), where \( \rho_{2M} \) is trilayer charge density from DFT, and \( \rho_{2M/o} \) is that of the pristine top/middle/bottom monolayer. Through the comparison with the charge redistribution due to a single interface (c.f. top and middle panels of Fig. 4a), it is remarkable to note that \( \Delta \rho \) in the trilayer is simply the sum of that due to each individual interface, showing that charge redistribution at adjacent interfaces can be treated as independent of each other. Figure 4b plots the electrical polarization \( P \) in trilayers where the two parallel interfaces have identical registry \( r \). Under the various \( r \), the trilayer \( P \) is all twice of that in the corresponding bilayer.

The above findings point to the possibility to design superlattices from the interference of electrostatic potentials from the electrical polarizations at multiple twisted parallel interfaces. For example, creating two independently configured twisted interfaces in h-BN can allow the generation of bichromatic superlattice potentials. Here we consider the scenario of staring from an N-layer film of AB stacking and apply torsion such that adjacent layers are all twisted from each other by a common angle \( \delta \). This creates \( N - 1 \) nearly parallel interfaces with the same periodicity, but with a small rotation by \( \delta \) from each other. With the charge redistribution at different interfaces being independent of each other, the electrostatic potentials contributed by adjacent interfaces are related by: \( V_{ij,l} (\mathbf{R}, z) = V_j (\mathbf{R}, z + d) \), where \( \mathbf{R} \) denotes...
the in-plane rotation by δ, with \( V(R, z) \) described by Eq. (2). The overall electrostatic potential is then,

\[
V_{\text{sum}}(R) = \sum_i V_0(R, z + ld),
\]

In the limit \( N \delta \ll 1 \), \( V_{\text{sum}}(R) \approx \sum_i V_0(R, z + ld) \), one obtains a superlattice potential with the strength scaled up with the thickness \( N \). In Fig. 4d, we show a plot of Eq. (3) at the surface of the h-BN film for \( N = 10 \) and \( \delta = 0.5^\circ \). The superlattice potential strength reaches \( \sim 1.3 \text{ eV} \). Figure 4c characterizes the potential strength with the increase of \( N \). For larger \( \delta \), the potential strength saturates at smaller \( N \). It is interesting to note that \( V_{\text{sum}}(R) \) always saturates with a common spatial profile with a chiral structure, as shown in Fig. 4e for the example of \( N = 20 \) and \( \delta = 1^\circ \).

**Electric field on h-BN and superlattice for dipolar objects**

As shown in Eq. (2), the potential \( V(R, z) \) decays exponentially in the out-of-plane direction in a length scale \( G^{-1} = \frac{\pi \delta}{2b} \). This is associated with an out-of-plane electric field on the h-BN surface with magnitude inversely proportional to superlattice period \( b \). Figure 5a shows the electric lines of forces, which have a common profile when plotted in the dimensionless coordinates \((R/b, z/b)\). For the example of a single parallel interface with \( \delta = 2^\circ \), which corresponds to \( b = 7.2 \text{ nm} \), the spatial profile of \( E_z \) and total field strength \( |E| \) are shown in Fig. 5b, along the long diagonal of a supercell, at a vertical distance of \( z = 2d \) from the interface. The field strength can reach \( \pm 6 \text{ mV}/\text{Å} \). This sizable electric field further enables the possibility to create electrostatic superlattice potential for dipolar objects such as interlayer excitons in bilayer TMDs, and dipolar molecules. Taking interlayer exciton as an example, it couples to \( E_z \) through its out-of-plane electric dipole, and the superlattice potential strength in the field of Fig. 5b can reach \( \sim 80 \text{ meV} \), assuming the interlayer distance of 7 Å in TMDs. Dipolar molecules adsorbed could have their orientations adjusted to the total field \( E \), then the potential landscape is determined by \( |E| \). The electric field strength can also be scaled up by having multiple twisted parallel interfaces in h-BN, for creating a quasi-periodic disordered landscape for the dipolar object, as shown in Fig. 5c.

**DISCUSSION**

We have shown the possibility to introduce a superlattice energy landscape on a general 2D material by exploiting twisted parallel interfaces inside the widely used h-BN substrate. Unlike the existing moiré superlattices, this noninvasive approach can be applied to the h-BN encapsulated high-quality monolayers, bilayers, etc., not limited by the lattice type and periodicity of the target materials. It is also worth noting that the orientation of the superlattice potential is determined by the h-BN interface only, and can be placed at an arbitrary angle to the crystalline axis of the target material, which is also a control parameter to tailor the minibands through various zone folding schemes. The approach can be further combined with the existing moiré superlattices in twisted graphene, twisted TMDs, and TMDs heterobilayers. In addition to the substrate, the twisted parallel interfaces can also be created in the h-BN capping layer where similar effect is expected, or even in the h-BN spacing layers exploited as tunneling barriers. The latter could point to interesting superlattice effects on the tunnelling processes.

In reality, structural relaxation can be relevant at the interfaces when the twisting angle is small30–32. The structural relaxation can increase the area of certain energetically favorable stacking while shrink the unfavorable ones, at the cost of inhomogeneous strain. This changes the mapping function \( r(R) \) in a supercell, and can quantitatively change the spatial profile of \( P(R) \) from the rigid twist one shown in Fig. 2e. Correspondingly, the spatial profile of the electrostatic potential can be modulated by such structural relaxation. As long as the local approximation is still applicable,
parallel interface with periodicity \( \delta \) on the h-BN surface can thus experience a superlattice with the laterally modulated polarization \( P \) from the lateral modulation of out-of-plane field \( E_z \), b Spatial profile of \( E_z \) and total field strength \( |E| \) along the long diagonal of a supercell, at a vertical distance of \( Z = 2d \) from a single interface with \( \delta = Z^* \). c \( E_z \) and \( |E| \) on the surface of a 20-layer film under torsion with \( \delta = 1^\circ \), along the dashed line in Fig. 4e.

the electrical polarization profile can still be quantified using \( P(R) \) from the relaxed texture \( r(R) \) of the interface. On the other hand, for the multilayers subject to torsion (Fig. 4 inset), the structural relaxation can be complicated with the large number of twisted interfaces. Compared to those shown in Fig. 4c–e for the rigid interfaces, the potential profile can get much more complex and the scaling of the potential strength with \( N \) can be significantly changed by such relaxation in the small \( \delta \) limit.

DATA AVAILABILITY

All data needed to evaluate the conclusions in the paper are included in the paper and/or the Supplementary Materials. Additional data related to this paper may be requested from the authors.

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W.Y. conceived and designed the research. P.Z. and C.X. performed the calculations. All authors analyzed the results and wrote the manuscript.

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