# Flat bands, strains, and charge distribution in twisted bilayer h-BN

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We study the effect of twisting on bilayer *h*-BN. The effect of lattice relaxation is included; we look at the electronic structure, piezoelectric charges, and spontaneous polarization. We show that the electronic structure without lattice relaxation shows a set of extremely flat in-gap states similar to Landau levels, where the spacing scales with twist angle. With lattice relaxation we still have flat bands, but now the spectrum becomes independent of twist angle for sufficiently small angles. We describe in detail the nature of the bands and we study appropriate continuum models, at the same time explaining the structure of the in-gap states. We find that even though the spectra for both parallel and antiparallel alignment are very similar, the spontaneous polarization effects only occur for parallel alignment. We argue that this suggests a large interlayer hopping between boron and nitrogen.

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## I. INTRODUCTION

The field of twisted bilayer materials has literally exploded in the past few years ever since the discovery of highly correlated phases in magic-angle twisted bilayer graphene (MATBG) [1]. The superconducting and insulating phases seen in such materials as a function of doping suggest that interactions play a crucial role.

Many other materials have been studied, both theoretically and experimentally, including transition metal dicalcogenides (TMDCs) [2,3], multilayer graphene systems such as twisted double bilayers [4–6], graphene stacks [6–10], and various forms of graphene twisted relative to hexagonal boron-nitride (*h*-BN) [11–14]; see also [15–18]. Twisted *h*-BN has also been proposed and studied [19–22], and it is the subject of this work.

There has been a great deal of recent interest in the electric properties of twisted-bilayer *h*-BN [23–26], where spontaneous charge polarization has been discovered for what is called "parallel" alignment, and none for the antiparallel one. Also, by mounting *h*-BN on a conducting substrate, we can look at the effect of an electric field. That leads to the question of the electronic structure of such materials: it is well known that flat bands occur in many such systems near the Fermi level, which drive most of the interacting physics since these are exquisitely sensitive to even weak residual forces. This clearly deserves investigation.

In MATBG, such continuum models are usually based on what is now called the Bistritzer-McDonald (BM) model [27,28], an inlayer continuum Dirac Hamiltonian with a very distinct threefold symmetry of the interlayer coupling. We have shown previously how we can derive a more detailed model from a tight-binding approach [29,30], still keeping much of the simplicity of such a model. Other approaches are discussed in the literature, for instance in Refs. [31–33].

It is an extremely interesting question whether we can find flat bands in twisted *h*-BN, and it is equally interesting to explore their nature and their continuum model description. With our toolbox we should be able to answer those questions, and we shall show that lattice relaxation has a surprising effect on the spectra, making an important part of it independent of twist angle.

Some large-twist-angle Density Functional based Tight Binding (DFTB) calculations for bilayer *h*-BN systems have been performed by Zhao *et al.* [20]. Another work along similar lines is that by Xian [19]. They do find flat bands, but the limitations of their computational techniques probably mean we can only use these results as indicative. The work by Javvaji *et al.* [22] takes an approach somewhat similar to ours, but it starts with a BM model with a gap (based on a tight-binding model like ours), thus it has some common elements but there are also clear differences. The main one is that their continuum model is not correct for small twist angles, as we shall discuss in detail.

The relaxation of the lattice gives rise to strain, and in piezoelectric materials this leads to charging in the areas of large strain. Since h-BN is a piezoelectric material, we could expect that charges are generated by the lattice relaxation. That leads to the question of whether this is the dominant mechanism for charge generation, and whether it is responsible for the charge domains observed in experiment.

The paper addresses these questions in order. We shall first look at lattice relaxation using an atomistic force model. We will then investigate the nature of the electronic states and

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especially the in-gap flat bands in twisted h-BN. We shall look at the nature of these states in both rigid and relaxed h-BN layers, and discuss continuum models that can be used to describe this. Finally, we turn our attention to the nature of the charge domains.

### **II. TWISTS AND RELAXATIONS**

There are two models predominantly used for relaxation: one is a simple harmonic potential model, often linked to DFT calculations, as done by, e.g., [34–38]. Such models work surprisingly well, but they lack some of the atomic detail for the smaller angles, which seems to lead to the occurrence of higher harmonics in the lattice deformation [29]. The only practical way this atomic nature can be reinstated is by using classical potential models, which is the approach taken here. Of course, such an approach has it own limitations, since it relies on a phenomenological potential model, normally fitted to both the results from microscopic models and experimental data.

#### A. Potential model

We use a standard approach, as we have discussed in great detail in, e.g., Ref. [29], where we use a classical atomistic model as implemented in the LAMMPS code [39]. We use this to relax the atomic positions to minimize a classical potential model for the energy. There are many suitable potential models for a single layer of a two-dimensional (2D) material; the difficulty is to find a good potential model for the interlayer interaction of these "van der Waals" layered materials. The most appropriate potential models for h-BN seem to be the "interlayer potential" (ILP) from Refs. [40,41]. This model was designed specifically for layered h-BN, and thus it should be a good choice for the problem at hand. The potential has been benchmarked in combination with the Tersoff inlayer potential [42–44], which is what we shall also adopt in this work. Of course, one should have a healthy skepticism as to the quantitative details of such models. Our experience [29] is that different potential models have a very close qualitative agreement, even though there are small quantitative differences (see also below).

#### **B.** Alignment

Since we have five potential alignments with complex energetics (see, e.g., Refs. [45,46] and Fig. 1), we will have to extend the analysis of our previous work. The main difference is that if we invert one of the layers, we change the main alignment, since we swap boron with nitride atoms: this corresponds to what is called "antiparallel alignment" in the literature.

#### C. Results

In Fig. 2 we show the alignments, defined using an extension of the method in our previous work [29]; see Appendix B. We use color saturation to show the quality of the dominant alignment, and a specific color for each type of this dominant alignment. The twist angle is 0.33°.



FIG. 1. The five different alignments, labeled as in Ref. [45], plus the top-bottom inversion BA. Vertical green lines denote aligned atoms. All alignments in the left column (AA, AN, and BA) are parallel; the ones on the right (AA', AB', and BA') are antiparallel.

As we can see in the figure, we find a substantial reorganization of alignment in both cases, with a very different pattern for parallel or antiparallel h-BN. Such patterns will induce an inhomogeneous strain in the h-BN, and since the material is piezoelectric, will also induce charge (or in other words,



FIG. 2. The dominant alignment structures (a), (c) parallel and (b), (d) antiparallel for a twist angle  $\theta = 0.33^\circ$ , L = 43.15 nm. Each image shows four primitive cells. Images (a) and (b) are rigid, undeformed structures; (c) and (d) are the associated relaxed structures. Each color shows a specific dominant alignment. Left column: blue, *AA* alignment (*B* above *B*, *N* above *N*); purple, *AB* alignment; red *BA* alignment. All three correspond to parallel alignment. Right column: green, *AA'* (*B* in one layer above *N* in another); orange/brown, *AB'* with aligned *N*; yellow, *AB'* with aligned *B*. All three antiparallel. The darker the color, the stronger the alignment.



FIG. 3. The induced piezoelectric charge in a single layer after relaxation of a *h*-BN bilayer. (a)–(c) Parallel at angles (a) 0.33°, (b) 0.67°, and (c) 1.05°. (d), (e) Antiparallel at angles (d) 0.33°, (e) 0.67°, and (f) 1.05°. The electron density *n* (scale on the right) is given in units of  $10^{12}$  cm<sup>-2</sup>. All images are drawn to the same scale.

the change in the inlayer hopping parameters, although small, will induce a charge). We have evaluated this by adopting expressions from continuum theory for these systems. The required strain tensor is the Lagrangian finite-difference one,  $E = \frac{1}{2}(F_S^T F_S - I)$ , where  $F_S$  is evaluated using the method for discrete hexagonal lattices from Ref. [47]; we then turn this into a piezoelectric charge using the method in Ref. [48]. The derivatives that are required in that last method are replaced with a finite difference on the *h*-BN lattice sites.

In Fig. 3 we notice that the charge density concentrates around the channels where the alignment changes, but also that it seems to decrease as we decrease the angle; this is slightly surprising, since the strain concentrates around the channels, but it seems that the rate of change of strain saturates. This cannot be the whole electronic charge of this system, since the concentration around these bands is not what is seen in experiment, where homogeneous charging is observed across each triangular domain [23–26].

#### **III. ELECTRONIC STRUCTURE**

We shall first concentrate our attention on the electronic hopping in the bilayer system. As discussed before, there are a few papers that discuss the spectrum of h-BN. The core motivation for the work here is the paper by Ribeiro and Peres [45], who were the first to derive a tight-binding model for a bilayer—the ingredient crucial to our work.

They start from a rather simple DFT calculation of two infinite bilayers aligned in the five positions shown in Fig. 1. The weaknesses of their DFT inputs are a substantial mismatch between the calculated and actual layer distance (the calculated one in their work is substantially larger than the accepted value of 3.33 Å), and what seems to be a gross underestimate of the gap—they find values around 4 eV, and *GW* calculations may be closer to 8 eV [49–52]. For some reason that is unclear to us, rather than fitting a single tight-binding model to all alignments, they fit a different model to each alignment.



FIG. 4. Left column: spectra for the naive tight-binding model for three different alignments as labeled. We use  $\Delta = 4.5$  eV, t = 2.33 eV, and find  $t_{BB} = 0.5$  eV,  $t_{NN} = 0.1$  eV, and  $t_{NB} = 0.2$  eV fit the spectra in Ref. [45]. The decay is parametrized by x = 0.25. We also show in the right column the results of the tight-binding model including the complete longer-ranged interlayer hopping.

Since the idea to fit a tight-binding model seems reasonable, we use a single more complete version of such a model and see what physics we can describe: We shall make use of an exponential parametrization of the interlayer hopping parameters, and we will ignore the nearest-neighbor inlayer one. Thus our interlayer hopping parameters will be assumed to take the simple, and potentially still too naive, form

$$t_{XY}(r) = t_{XY} \exp[-\alpha(r-d)].$$
(1)

We shall use d = 3.33 Å and  $\alpha = 4.4$  Å<sup>-1</sup> [53].

With these additional hopping parameters, if we truncate the interlayer coupling to atoms placed directly above each other and their nearest neighbors, we find Hamiltonian matrices that are a slight generalization of those in Ref. [45]; see (A1) for detailed expressions. These depend on the gap  $\Delta$  and the inlayer hopping *t*. Unlike in Ref. [45], we assume that these parameters are the same for all alignments. We then fit the tight-binding model, in a truncated form that can be evaluated analytically, to the spectra in Ref. [45]. The results are shown on the left in Fig. 4; in that figure, we also show on the right that the full numerical tight-binding calculation does not differ substantially from the analytical model.

With the parameters as shown in Fig. 4, we get a reasonable representation of the spectra and gap as compared to DFT. Since we are describing a system with smaller *d* and a larger gap than in Ref. [45], we find that we get similar band structures with this larger gap if we use the parameters  $\Delta = 8 \text{ eV}$ , t = 2.33 eV, and the hopping parameters increase by almost 50%,  $t_{BB} = 0.7 \text{ eV}$ ,  $t_{NN} = 0.15 \text{ eV}$ , and  $t_{NB} = 0.3 \text{ eV}$ —these are obtained by a rough fit to the dispersion of DFT calculations for aligned *h*-BN (we shall argue below that the experimental data suggest that  $t_{NB}$  may well



FIG. 5. Relaxation for  $\theta = 1.05^{\circ}$ : (a) parallel and (b) antiparallel alignment. See the caption of Fig. 2 for details of the color coding. Each image shows four primitive cells.

be larger). We ignore next-nearest-neighbor inlayer couplings; as shown in Ref. [54], their effect is small, and inclusion of next-nearest-neighbor hoppings would require us to determine two additional parameters for the calculations.

We can now use these parameters to try and find the spectra of twisted *h*-BN layers. For computational efficiency, we shall initially study an angle of  $1.05^{\circ}$ —full tight-binding models are rather expensive for smaller angles, but we shall investigate an alternative approach in a later section.

#### **Detailed comparison**

We start with a detailed tight-binding calculation for a twist angle of  $1.05^{\circ}$ , and a cell spacing (also called the moiré wavelength  $L_s$ ) of 13.6 nm. The alignment for the two relaxed structures considered here is shown in Fig. 5. We have calculated the spectra for both relaxed and unrelaxed structures of this nature; see Fig. 6. We note that the spectra for the unrelaxed structures are identical, but the Hamiltonians are completely different, and thus we need to identify a mechanism that gives rise to this. The spectra for these two cases seem similar to Landau levels, which is also reflected in the increasing degeneracy: 2 for the states deepest inside the gap, then 4, 6, 8, ... as the energy increases. As we shall show below, their origin is very different from the related argument made for twisted bilayer graphene in Ref. [55].

For the relaxed positions, we see rather different spectra. In all cases, the spectra are extremely flat: the bandwidth of each state is only a fraction of an meV until we reach the quasicontinuum at the gap energy.



FIG. 6. Tight-binding spectra for  $\theta = 1.05^{\circ}$ . (a) Unrelaxed and (c) relaxed lattice corresponding to (a) in Fig. 5. (b) Unrelaxed and (d) relaxed lattice corresponding to (b) in Fig. 5.



FIG. 7. Tight-binding spectra for (a), (d)  $\theta = 1.05^{\circ}$ , (b), (e)  $\theta = 0.67^{\circ}$ , and (c), (f)  $\theta = 0.33^{\circ}$ . This is for the relaxed lattice (c) in Fig. 2. (a), (b), (c) no relaxation; (d), (e), (f) relaxed lattice. The sharp cutoff at the top and bottom of the spectrum for the smallest angles is an artifact of our numerical approach due to the calculation of a finite number of eigenvalues.

There are some very intriguing features in the spectra as we change twist angle: As can be seen in Fig. 7, when there is no relaxation we see an equally spaced set of levels that also show a typical two-dimensional harmonic-oscillator degeneracy (which is doubled due the valley degeneracy), where the spacing decreases with an increasing moiré wavelength (supercell size). On relaxation a few flat bands remain, with the same 2-4-6-... degeneracy, but the most surprising result is that the in-gap states now appear to be independent of twist angle: their energies are so similar that we had to check twice that we had actually used the right images.

We conclude that we will have to find an explanation for two different phenomena: the occurrence for in-gap flat bands (flat to within a fraction of an meV), which show a harmonic oscillator type spacing for lattices that do not relax at the interface, with the spacing decreasing as the twist angle decreases, and the occurrence of twist-angle independent flat bands if we relax the lattice at sufficiently small angles. Both of these should be described by a type of continuum model. The first case should be described by a generalized BM model discussed in the next section, and the second by an alternative approach, probably in real space.

## **IV. CONTINUUM PROJECTION**

To understand the flatness of the bands, we first plot some wave functions from a tight-binding calculation, Fig. 8, and we see that these indeed look like 2D harmonic-oscillator states shifted by a value proportional to the momentum. To get an analytical handle on this, we turn to a continuum projection, using a "generalized Bistritzer-MacDonald model" [27,28]. We follow the approach set out in [29]. This established technology is known to generate continuum models that completely reproduce the full tight-binding calculations near the Fermi energy for graphene; we just need to check the equivalent result for h-BN.

As we notice in Fig. 9, for undeformed lattices the projections of parallel and antiparallel layers are identical. We have seen that the Hamiltonians are rather different, but the results here are no surprise due to the similarity of the tight-binding spectra calculated earlier. Upon relaxation, we see that higher harmonics, corresponding to a larger superlattice momentum transfer, start playing a role.



FIG. 8. The absolute value of the four components of the realspace wave function multiplied with the phase  $\exp[i(\mathbf{k} - \mathbf{K}_i) \cdot \mathbf{r}]$  for (a)  $\mathbf{k} = \mathbf{K}_1$ , (b)  $\mathbf{k} = \mathbf{K}_2$ , and (c)  $\mathbf{k} = \mathbf{K}_1/5 + \mathbf{K}_2/3$ . The four columns are layer 1 N (A) sites, layer 1 B sites, and the same for layer 2. The hue of the coloring shows the phase of the wave function. Thus the first column is real and positive, while the third column is real and negative. The B site wave functions show an (almost) uniform phase change of  $2\pi$  as a function of the polar angle around the origin. Space axis units in Å.

We have investigated the source of the flat bands using this model—we shall concentrate here on the larger spacing at the positive side of the gap, but a similar analysis applies at the other end. For the case of the flat bands, we have the classic Bistritzer-MacDonald model with an extra gap added to the inlayer Hamiltonian,

$$H = \begin{pmatrix} h(\mathbf{K}_1, \theta/2) & U(\mathbf{r}) \\ U^{\dagger}(\mathbf{r}) & h(\mathbf{K}_2, -\theta/2) \end{pmatrix},$$
(2)

$$h(\boldsymbol{k},\theta) = \boldsymbol{h}_{1}(-i\boldsymbol{\nabla}-\boldsymbol{k},\theta)\cdot\boldsymbol{\sigma} + \frac{\Delta}{2}\sigma_{3}.$$
 (3)

Unlike for the case of graphene, we find little benefit using the full inlayer tight-binding dispersion for  $h_1$  rather than the simpler linear expansion  $h_1(k) = \hbar v_F(-i\nabla - k)$ .

In its simplest form, the matrix U takes the form

$$\begin{pmatrix} u_{AA}(\boldsymbol{r}) & u_{AB}(\boldsymbol{r}) \\ u_{AB}^{\dagger}(\boldsymbol{r}) & u_{BB}(\boldsymbol{r}) \end{pmatrix},$$
(4)

where

$$u_{AA}(\boldsymbol{r}) = u_{0BB}g(\boldsymbol{r}), \tag{5}$$

$$u_{BB}(\boldsymbol{r}) = u_{0NN}g(\boldsymbol{r}), \tag{6}$$

$$u_{AB}(\boldsymbol{r}) = u_{0BN}g'(\boldsymbol{r}), \tag{7}$$

$$g(\mathbf{r}) = (1 + e^{-i\mathbf{G}_{1}\cdot\mathbf{r}} + e^{i\mathbf{G}_{2}\cdot\mathbf{r}}),$$
(8)

$$g'(\mathbf{r}) = (1 + e^{-i2\pi/3}e^{-iG_1\cdot\mathbf{r}} + e^{i2\pi/3}e^{iG_2\cdot\mathbf{r}}), \qquad (9)$$

where  $G_1$  and  $G_2$  are the reciprocal unit vectors of the superlattice.



FIG. 9. Projection of the interaction onto a continuum model as described in [29]. Each hexagon is one interaction matrix element for a given momentum transfer. Zero momentum transfer is denoted by the pink dot, and the three dominant matrix elements for the unrelaxed lattice are exactly those for the BM model. In each panel, the left top figure shows the AA projection, and the right top one the AB case. The lower row shows the BA (left) and BB case (right), respectively. Unlike in graphene, the AA and BB cases are not identical. The (a)–(d) labels are as in Fig. 6. All couplings are given in units of eV. Each panel represents the Fourier decomposition of the matrix in Eq. (4) at k = M.

### A. In-gap states

We first look numerically at which parameters are most relevant; we find that the energies are largely insensitive to the value of  $u_{0BN}$  and  $u_{0NN}$ , and the wave functions are dominantly located on the boron sites (there is a small component on the *N* sites); they look very much like two (discrete) Gaussians centered on the momentum  $\mathbf{k} - \mathbf{K}_i$ ; the signs of the Gaussians are opposite for the two layers. Unlike for the case of TBG [54], we find that in this case the effect of replacing  $\mathbf{h}_1$  by its linear expansion is small, and we will thus work with the latter.

The result we see brings to mind the analysis of Ref. [55], even though that work is for a different problem, and seems to ignore the mismatch between the two Dirac points, which is crucial for a cancellation of the gauge fields; see below. We look at the Hamiltonian in coordinate space, where the inlayer potential is expanded about the two K-points, following the standard BM continuum model [28], with the addition of a gap; see also [22]. To simplify the analysis, we define the wave function with a momentum translation to the relevant K point by writing

$$\psi_{k}(\mathbf{r}) = (e^{iK_{1}\cdot\mathbf{r}}\psi_{B1k}(\mathbf{r}), e^{iK_{1}\cdot\mathbf{r}}\psi_{N1k}(\mathbf{r}),$$
$$\times e^{iK_{2}\cdot\mathbf{r}}\psi_{B2k}(\mathbf{r}), e^{iK_{2}\cdot\mathbf{r}}\psi_{N2k}(\mathbf{r})).$$
(10)

We also permute rows and columns, so that the positive gap appears in the upper left-hand block,

$$H = \begin{pmatrix} \Delta/2 & u_{0BB}g(r)e^{i\delta K \cdot r} & v_F p_- & u_{0BN}g'(r)e^{i\delta K \cdot r} \\ u_{0BB}e^{-i\delta K \cdot r}g(r)^* & \Delta/2 & u_{0BN}g'(r)e^{-i\delta K \cdot r} & v_F p_- \\ v_F p_+ & u_{0BN}e^{i\delta K \cdot r}g'(r)^* & -\Delta/2 & u_{bb}g(r)e^{i\delta K \cdot r} \\ u_{0BN}e^{-i\delta K \cdot r}g'(r)^* & v_F p_+ & u_{bb}e^{-i\delta K \cdot r}g(r)^* & -\Delta/2 \end{pmatrix},$$
(11)

where  $\delta K$  is the difference in momenta between the two K points,  $\delta K = K_1 - K_2$ . We use the notation p to denote the momentum operator, with  $p_{\pm} = p_1 \pm i p_2$ . We assume we are looking at an in-gap eigenvalue just below the top of the gap,  $E = \Delta/2 - \epsilon$ , with  $\epsilon \ll \Delta$ . We solve for the lower two components to eliminate the B wave functions, and find to first order in  $1/\Delta$  [strictly speaking, we expand in terms of all five of the small scales  $v_F \langle p \rangle / \Delta$ ,  $u_i / \Delta$ , and  $\epsilon / \Delta$ ]

$$H_0 = \begin{pmatrix} \Delta/2 & 0\\ 0 & \Delta/2 \end{pmatrix} \tag{12}$$

and

$$H_{1} = \frac{1}{\Delta} \begin{pmatrix} -u_{0BN}^{2} |g'(r)|^{2} + v_{F}^{2} p^{2} & e^{i\delta K \cdot r} [\Delta u_{0BB} g(r) - 2u_{0BN} v_{F} \operatorname{Re}(p_{+}g'(r))] \\ e^{-i\delta K \cdot r} [\Delta u_{0BB} g(r)^{*} - 2u_{0BN} v_{F} \operatorname{Re}(p_{+}g'(r))] & -u_{0BN}^{2} |g'(r)|^{2} + v_{F}^{2} p^{2} \end{pmatrix}.$$
 (13)

Intriguingly enough, we see that the kinetic energy actually only appears at first order. So where does the harmonicoscillator potential we hope to see appear? To see that, we must reexpress our results in the basis where the Hamiltonian  $H_0$  is diagonal. We suffer from the problem that the basis now depends on r, and thus the kinetic energy acts nontrivially on this.

Let us ignore the terms proportional to  $u_{0BN}$ —numerically we see that these are unimportant relative to the lowest-order potential due the fact that  $u_{0BN} \ll \Delta$  and  $u_{0BN} < \Delta$ . We diagonalize the Hamiltonian to first nonvanishing order, which gives two contributions: the diagonalized matrix

$$H_1' = \begin{pmatrix} v_F^2 / \Delta p^2 - u_{0BB} | g(r) | & 0\\ 0 & v_F^2 / \Delta p^2 + u_{0BB} | g(r) | \end{pmatrix},$$
(14)

and a second term due to the fact that the momentum operator in  $H_1$  acts nontrivially on the transformation matrix T that brings the Hamiltonian to diagonal form,

$$T = \frac{1}{\sqrt{2}} \begin{pmatrix} e^{i[\delta \boldsymbol{K} \cdot \boldsymbol{r} + \phi_{g}(r)]} & 1\\ -e^{i[\delta \boldsymbol{K} \cdot \boldsymbol{r} + \phi_{g}(r)]} & -1 \end{pmatrix},$$
(15)

with

$$e^{\phi_g(r)} = \sqrt{g(r)} / \sqrt{g(r)^*}.$$
 (16)

The derivative in the Hamiltonian acts on the phase factor in T as

$$\nabla e^{i[\delta \mathbf{K} \cdot \mathbf{r} + \phi_g(r)]} = (\delta \mathbf{K} - \mathbf{G}_1 e^{-i\mathbf{G}_1 \cdot \mathbf{r}} + \mathbf{G}_2 e^{i\mathbf{G}_2 \cdot \mathbf{r}}) e^{i[\delta \mathbf{K} \cdot \mathbf{r} + \phi_g(r)]}.$$

Since  $\delta K = G_1 - G_2$ , this quantity vanishes for small r, and thus we can safely ignore the resulting vector potential near the origin, where all the wave functions are located.

If we now look at the top entry of the Hamiltonian, which describes the in-gap modes, we find an effective Hamiltonian,

$$H_{\rm eff} = \frac{v_F^2}{\Delta} p^2 - u_{0BB} |g(r)|.$$
(17)

5)

If we now expand |g(r)|, we find that we can express this in terms of the moiré wavelength  $L_s$  as

$$|g(r)| = 3 - \frac{4[\pi^2(x^2 + y^2)]}{3L_s^2}.$$
 (18)

Thus all together, we have a harmonic oscillator

$$H_{\rm eff} = -3u_{0BB} + \frac{v_F^2}{\Delta}p^2 + \frac{4\pi^2 u_{0BB}}{3L_s^2}r^2.$$
 (19)

Thus the spacing of the levels is

$$\hbar\omega = \sqrt{4\frac{\hbar^2 v_F^2}{\Delta} \frac{4\pi^2 u_{0BB}}{3L_s^2}} = \frac{4\pi \hbar v_F}{L_s} \sqrt{\frac{u_{0BB}}{3\Delta}}, \qquad (20)$$

with the full energy of the *n*th state (with degeneracy 2*n*)

$$E_n = -3u_{0BB} + (n+1)\hbar\omega.$$
 (21)

This agrees well with the data in Fig. 10; a small renormalization (a 3% increase) of  $u_{0BB}$ , probably due to neglected higher-order effects, gives results indistinguishable from the linear fit.



FIG. 10. The in-gap spectrum for  $L_s = 13.6$  (blue points), 21.5 (orange points), and 43.1 nm (green points) from Figs. 7(a)-7(c) vs harmonic-oscillator quantum number n. Solid lines show a linear fit; dashed lines show the result from Eq. (21).



FIG. 11. Comparison of continuum projections for (a), (d)  $\theta = 1.05^{\circ}$ ; (b), (e)  $\theta = 0.67^{\circ}$ , and (c), (f)  $\theta = 0.33^{\circ}$ . (a), (b), (c) hexagonal lattice; (d), (e), (f) relaxed lattice. Whereas the reciprocal space interlayer coupling does not depend on twist angle for the rigid lattice, relaxation gives a very long range in the *AA* and *BB* couplings. In each panel, the left top figure shows the *AA* projection, and the right top one shows the *AB* case. The lower row show the *BA* (left) and *BB* projection (right), respectively. All couplings are given in units of eV.

## **B.** Relaxation

We have already seen in Figs. 7(d)-7(f) that for the relaxed lattices the in-gap spectra are roughly independent of twist angle. Since the Wannier functions corresponding to these bands are still like Gaussians, the real-space continuum model describing these states must be essentially the same.

Let us take a more detailed look at the continuum model projection as a function of twist angle. As we can see in Fig. 11, the results from the projection are indeed very different in the AA and BB channel after relaxation, and they are strongly dependent on twist angle. The reason is that while the physical size of the AA aligned regions is constant, independent of twist angle, and thus we would expect the real-space potential to be independent of angle, the reciprocal-lattice spacing reduces substantially as we change the angle, and thus many more Fourier components of smaller magnitude are needed to describe this potential. The AB regions grow, leading to almost constant coupling terms.

The model that seems to describe this behavior is a slightly extended version of the real-space model derived in the previous section: a confined potential well, where the well is centered on the region of AA alignment, with a sharp cutoff at the edges. This is very difficult to describe in momentum space, but the real-space wave functions all look very similar, independent of twist angle. We have not pursued such a model here, since we know we can do very accurate tight-binding and continuum model calculations at an an angle of, say,  $1.05^{\circ}$ . We can either turn that into a real-space Hamiltonian as above, which will then have spatially localized solutions, or we can solve the problem in *k* space for such a large twist angle. The (real-space) solutions for the in-gap flat bands are no longer dependent on twist angle, and we have thus solved the problem for these states for all smaller twist angles. It is thus absolutely incorrect to apply the lowest-harmonic Bistritzer-MacDonald model to such situations: if there is relaxation of the atomic lattice, this will fail drastically at small angles. The behavior is more like states localized at the *AA* impurity.

#### V. CHARGE DENSITY

Since experimental data [23–26] suggest twisted *h*-BN is electrically charged, it would be interesting to try to understand the charge density in detail. It appears that the only way to get reliable results is to sum over *all* occupied states, since converged results are only found when summing over all eigenvectors.

As can be seen in Fig. 12, for the triangular relaxation the charge density indeed has the triangular pattern observed in experiment. This charge density is largely carried by the B atoms, due to the difference in hopping parameters. There is no charge density for the antiparallel twisted case, and there is a three orders of magnitude smaller charge density when we relax the antiparallel crystal.



FIG. 12. Twist-induced charge density at neutrality in the top layer for  $\theta = 1.05^{\circ}$ . (a) Unstrained layer as in (a) in Fig. 5. (b) Relaxed layer as in (c) in Fig. 5. We clearly note the enhanced triangular symmetry. The units used are the same as in Fig. 3.

The uncertainty and sensitivity to parameters of the charge density need quantification, and we have performed a more detailed analysis; see Fig. 13. There is some sensitivity to the range parameter  $\alpha$  in (1): The charge density falls with an increase of the range parameter. We conclude that the experimental results suggest a relatively short-range hopping, maybe driven by many-body screening as in Ref. [29]. However, the dominant parameter is the hopping  $t_{NB}$ : increasing that from 0.3 to 0.5 eV increases the maximum in the electron density to  $2 \times 10^{12}$  cm<sup>-2</sup>. This value of the hopping is still quite reasonable, and may well help us to put constraints on microscopic calculations of such parameters.

For the case with the highest electron density, the charge density is about  $\sigma = \pm 2 \times 10^{12} \times 10^4 \times 1.6 \times 10^{-19} = \pm 3 \times 10^{-3}$  C/m<sup>2</sup>. If we take the vacuum value  $\epsilon_r = 1$  between the *h*-BN layers [56,57], we find, assuming the triangular domains are large enough to apply an infinite-parallel-plate approximation,

$$V = \frac{\sigma}{\epsilon_r \epsilon_0} d = \frac{3 \times 10^{-3} \times 3.33 \times 10^{-10}}{8.85 \times 10^{-12}} = 110 \text{ mV}.$$
 (22)

### VI. EFFECT OF CHARGE ON ELECTRONIC SPECTRUM

Of course such a charge density can impact the electronic spectrum of the in-gap states, especially since the Wannier states are localized at the point where the positive charge density meets the negative one. Clearly, in this case we need to look at both the ionic and the piezoelectric charges, since they are both of similar magnitude near the AA/AA' points. The total charge-carrier density does not exceed the value of  $n_0 = 2 \times 10^{16}$  cm<sup>2</sup>. Define a dimensionless carrier density  $\bar{n}(\mathbf{x}) = n(\mathbf{x})/n_0$ . Expressing all distances in angstroms, we find that the Coulomb force due to the charge in the two layers is

$$V(\mathbf{x}) = \frac{\alpha}{\epsilon_r} n_0 \hbar c \int \bar{n}(\mathbf{y}) \left( \frac{1}{|\mathbf{x} - \mathbf{y}|} - \frac{1}{|\mathbf{x} - \mathbf{y} + d\mathbf{e}_z|} \right) d^2 \mathbf{y}$$
  
= (3 meV/Å)  $\int \left( \frac{1}{|\mathbf{x} - \mathbf{y}|} - \frac{1}{|\mathbf{x} - \mathbf{y} + d\mathbf{e}_z|} \right) d^2 \mathbf{y}.$  (23)

Using a model for large charge domains, where just six domains meet at a point, we find that the integral above is at most 2 Å. Since the potential has positive and negative contributions, we estimate the maximum effect of the perturbation of the charge as a function of momentum to be much less than 1 meV. This clearly does not modify the isolated bands by a significant amount.

#### VII. CONCLUSIONS

We conclude that twisted h-BN has in-gap flat bands, which are extremely flat. If the crystal were not to relax, these would be a set of equally spaced levels, similar to Landau levels, and we have clearly identified their origin and location. As we relax the lattice, we lose some of these levels, even though they remain extremely flat (to numerical accuracy, about  $10^{-2}$  meV). For large twist angles, these can be described as a continuum model with a gap, in the Bistritzer-MacDonald mold [22]. For smaller twist angles this is not the correct description, since the in-gap spectrum becomes independent of twist angle, showing these are a set of states all localized near the AA' aligned point, which is a region that becomes independent of twist angle as that decreases. A continuum description of these states can then be trivially extracted for the larger twist angles, where a BM model captures these states. In the end, the width of these states will be determined by the induced charge density, which may lead to a bandwidth of about 1 meV.



FIG. 13. Twist-induced charge density at neutrality in the top layer for  $\theta = 1.05^{\circ}$  for a relaxed layer for change to the basic parameter choice  $t_{BB} = 0.7$ ,  $t_{NB} = 0.3$ ,  $t_{NN} = 0.15$ ,  $\Delta = 8 \text{ eV}$ , and  $\alpha = 4.4 \text{ Å}^{-1}$ . (a) Basic parameters, (b)  $t_{BB} = 0.5 \text{ eV}$ , (c)  $\alpha = 2.2 \text{ Å}^{-1}$ , (d)  $\alpha = 6.6 \text{ Å}^{-1}$ , (e)  $\Delta = 6 \text{ eV}$ , (f)  $t_{NB} = 0.5 \text{ eV}$ , (g)  $t_{NB} = 0.15 \text{ eV}$ , and (h)  $t_{BB} = 1.0 \text{ eV}$ . The electron density N (scale on the right) is given in units of  $10^{12} \text{ cm}^{-2}$ .

Without lattice relaxation, parallel and antiparallel alignments have identical in-gap spectra. Since, as discussed in the body of the paper, these lattices relax in a very different way, the spectra are also very different, but both have flat bands, caused by the same mechanism of a fixed size island of AA or AA' alignment where these states live.

One of the surprising features of this work is that the parallel case has a permanent dipole polarization, which dominates the piezoelectric charge that is caused by the strain to relaxation. This agrees with the observations in Refs. [23-26], and it has been described with different theoretical methods in those works.

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### APPENDIX A: TIGHT-BINDING MODEL EXPRESSIONS

With these additional hoppings, if we truncate the interlayer coupling to atoms placed directly above each other and their nearest neighbors, we find Hamiltonians that are a slight generalization of those in Ref. [45] [see (A1)],

$$h_{AA} = \begin{pmatrix} -\frac{\Delta}{2} & tg(\mathbf{k}) & t_{NN} & t_{NB}xg(\mathbf{k}) \\ tg(\mathbf{k})^* & \frac{\Delta}{2} & t_{NB}xg(\mathbf{k})^* & t_{BB} \\ t_{NN} & t_{NB}xg(\mathbf{k}) & -\frac{\Delta}{2} & tg(\mathbf{k}) \\ t_{NB}xg(\mathbf{k})^* & t_{BB} & tg(\mathbf{k})^* & \frac{\Delta}{2} \end{pmatrix},$$

$$h_{AA'} = \begin{pmatrix} -\frac{\Delta}{2} & tg(\mathbf{k}) & t_{NB} & t_{NN}xg(\mathbf{k}) \\ tg(\mathbf{k})^* & \frac{\Delta}{2} & t_{BB}xg(\mathbf{k})^* & t_{NB} \\ t_{NB} & t_{BB}xg(\mathbf{k}) & \frac{\Delta}{2} & tg(\mathbf{k}) \\ t_{NN}xg(\mathbf{k})^* & t_{NB} & tg(\mathbf{k})^* & -\frac{\Delta}{2} \end{pmatrix},$$

$$h_{AB'} = \begin{pmatrix} -\frac{\Delta}{2} & tg(\mathbf{k}) & t_{NN} & t_{NB}xg(\mathbf{k}) \\ tg(\mathbf{k})^* & \frac{\Delta}{2} & t_{NB}xg(\mathbf{k})^* & 0 \\ t_{NN} & t_{NB}xg(\mathbf{k}) & -\frac{\Delta}{2} & tg(\mathbf{k}) \\ t_{NB}xg(\mathbf{k})^* & 0 & tg(\mathbf{k})^* & \frac{\Delta}{2} \end{pmatrix},$$

$$h_{AB'} = \begin{pmatrix} \frac{\Delta}{2} & tg(\mathbf{k}) & t_{BB} & t_{NB}xg(\mathbf{k}) \\ tg(\mathbf{k})^* & -\frac{\Delta}{2} & tg(\mathbf{k}) \\ t_{NB}xg(\mathbf{k})^* & 0 & tg(\mathbf{k})^* & 0 \\ t_{BB} & t_{NB}xg(\mathbf{k}) & \frac{\Delta}{2} & tg(\mathbf{k}) \\ t_{NB}xg(\mathbf{k})^* & 0 & tg(\mathbf{k})^* & -\frac{\Delta}{2} \end{pmatrix},$$

$$(A1)$$

Here  $\Delta$  is the gap, and *t* is the inlayer hopping;  $g(\mathbf{k}) = e^{i(\frac{\sqrt{3}k_y}{2} - \frac{k_x}{2})} + e^{i(\frac{k_x}{2} + \frac{\sqrt{3}k_y}{2})} + 1$  is the standard sum of three phase factors usually found in these calculations. *x* is the suppression factor for hopping to a next-to-nearest neighbor. Unlike in



FIG. 14. Graphical representation of the terms used in Eqs. (B1) and (B2) for the atoms labeled "c." The first term is for AA-type alignment, the last two define two situations in AB alignment. The dotted circles are the inverted positions of the upper-layer nearest-neighbor atoms relative to the central one.

Ref. [45], we assume that these parameters are the same for all alignments.

## **APPENDIX B: DEFINITION OF ALIGNMENT MEASURE**

To compare the size of the *AA* and *AB* aligned domains, we construct a measure of alignment, made of a measure for *AA* and *AB* alignment. We first define a measure of *AA* alignment by the function (*l* labels the layer,  $\bar{l}$  denotes the opposite layer,  $\langle r_{li}r_{\bar{l}j}\rangle$  denotes the atom *j* closest to atom *i* but in the opposite layer,  $\delta_{li}^{(k)}$  denotes the three vectors connecting atom *i* to its nearest neighbors in the same layer, and  $\mathbf{r}_{lik\sigma} = \mathbf{r}_{li} + \sigma \delta_{li}^{(k)}$ )

$$w_{AA}(\mathbf{r}_{li}) = \frac{1}{a^2} \delta_{\langle r_{li} r_{\bar{l}j} \rangle} \left[ 3(\mathbf{r}_{l,i} - \mathbf{r}_{\bar{l},j})^2 + \sum_k (\mathbf{r}_{l,i,k,+} - \mathbf{r}_{\bar{l},j,k,+})^2 \right].$$
(B1)

In a similar way, we define the quality of any *AB* alignment as the following function:

$$w_{AB}(\mathbf{r}_{li}) = \frac{1}{a^2} \min\left(\delta_{\langle l_i \bar{l}_j \rangle} 3(\mathbf{r}_{li} - \mathbf{r}_{\bar{l}j})^2 + \sum_k (\mathbf{r}_{lik+} - \mathbf{r}_{\bar{l}jk-})^2, \sum_{k\sigma} (\mathbf{r}_{lik\sigma} - \mathbf{r}_{\bar{l}j} \delta_{\langle \mathbf{r}_{lik\sigma}, \mathbf{r}_{\bar{l}j} \rangle})^2\right).$$
(B2)

The factors of 3 in front of the central terms ensure that we use six atoms in every expression; they also weigh the central atom more heavily, which has proven to be an effective approach. The value of a we shall use is the *h*-BN nearest-neighbor spacing.

We then use

$$w = \max(w_{AA}, w_{AB}, w_{BA}, w_{AA'}, w_{AB'}, w_{A'B})$$
(B3)

as a measure of alignment, and we choose the color according to the dominant choice. Note that w is extremal for perfect alignment. See Fig. 14 for a graphical explanation of each of the terms.

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