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Band Engineering of Large-Twist-Angle Graphene/*h*-BN Moiré Superlattices with Pressure

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Graphene interfacing hexagonal boron nitride (*h*-BN) forms lateral moiré superlattices that host a wide range of new physical effects such as the creation of secondary Dirac points and band gap opening. A delicate control of the twist angle between the two layers is required as the effects weaken or disappear at large twist angles. In this Letter, we show that these effects can be reinstated in large-angle ($\sim 1.8^\circ$) graphene/*h*-BN moiré superlattices under high pressures. A graphene/*h*-BN moiré superlattice microdevice is fabricated directly on the diamond culet of a diamond anvil cell, where pressure up to 8.3 GPa is applied. The band gap at the primary Dirac point is opened by 40–60 meV, and fingerprints of the second Dirac band gap are also observed in the valence band. Theoretical calculations confirm the band engineering with pressure in large-angle graphene/*h*-BN bilayers.

van der Waals (vdW) heterostructures by vertically stacking two-dimensional (2D) materials exhibit emergent properties distinct from those of the individual 2D layers [1–11]. For example, the periodic moiré potential imposed on monolayer graphene (MLG) by interfacing hexagonal Boron Nitride (*h*-BN) with delicately controlled rotational-angle alignment breaks the inversion symmetry and opens a band gap at the primary Dirac point (PDP) as well as a series of secondary Dirac band gaps in both the valence and conduction bands [12–15]; bilayer graphene with a “magic” twist angle of $\sim 1.1^\circ$ shows Mott-insulating behavior at half filling and superconductivity if doped by electric fields [16–18]. All these novel effects emerge only at very small, sometimes specific, twist angles between neighboring layers, which imposes an experimental challenge.

Interests are growing in the role that interlayer vdW forces play in regulating these emergent effects in vdW structures [19–22]. A recent study by Yankowitz *et al.* reports that hydrostatic pressure is powerful in tuning the interlayer coupling and hence band structures of graphene moiré superlattices [20]. The band gap at the PDP of 0.6° twisted moiré superlattices was raised from 30 meV at ambient to 50 meV at 2.3 GPa applied in a piston cell. Wider band gaps are intuitively expected in the graphene system under higher pressures, and also of importance for electronics applications. However, the maximum pressure

is typically limited to be below ~ 3 GPa in piston cells. In contrast, diamond anvil cells (DACs) can generate extremely high hydrostatic pressures (up to ~ 1 TPa), and they are widely used to tune the interlayer spacing of 2D materials. However, DACs are typically used to carry out nonelectrical experiments of ultrathin (few-layer) 2D materials such as Raman and photoluminescence [23–27] due to daunting technical difficulties in electrical wiring of atomically thin materials through the DAC [28,29]. A limited number of electrical measurements in DAC reveal unusual properties of ultrathin 2D materials at high pressures, such as much improved electron mobility in few-layer MoS₂ [28]. Therefore, it is intriguing to search for new electronic properties of graphene moiré superlattices at high pressures in a DAC. The electron-beam lithography (along with one-dimensional edge contact) commonly used for microdevice fabrication of vdW materials [30] is not compatible with the DAC, as the diamond is insulating and the epoxy (for fixing the diamond) is vulnerable to electron beams. Chen *et al.* [28] developed a new method to prepare and measure field effect transistors (FETs) of MoS₂ in a DAC by directly lithographically patterning microdevices onto the diamond culet. In this work, we advance this method to fabricate and measure three-terminal FETs of graphene moiré superlattices in the DAC.

For the graphene/*h*-BN moiré superlattices, experimental and theoretical studies have demonstrated the significance and sensitivity of the rotational alignment between the two layers [31–37]. For example, the band gap near the PDP is around 30 meV if the twist angle is $<2^\circ$ and progressively decreases if the twist angle increases. The band gap at the secondary Dirac points (SDPs) rapidly decreases when the twist angle increases and vanishes when the twist angle is $>1^\circ$ [31]. Most recent studies of graphene/*h*-BN moiré superlattices are focused on systems with small twist angles ($<\sim 1^\circ$), since most of the intriguing physical phenomena are believed to appear only in these well-aligned systems. However, making a nearly perfectly aligned graphene/*h*-BN stack is extremely challenging even for very experienced hands and the possible exotic properties of systems with large twist angles remain nearly unexplored. In this work, electrical transport measurements are carried out on graphene/*h*-BN moiré superlattices with a rather large twist angle of $\sim 1.8^\circ$. We show that the PDP band gap increases by 40–60 meV at a pressure of 8.3 GPa. Theoretical calculations confirm these findings and further predict that an indirect band gap opens at the SDP in the valence band when pressure exceeds 7.3 GPa, and reaches 4.7 meV at 8.4 GPa. Experimental fingerprints are observed to be consistent with the prediction about the secondary band gap.

Figures 1(a) and 1(b) illustrate our experimental setup. Cubic-BN (*c*-BN) mixed with epoxy gel was coated onto a stainless-steel gasket to avoid shorting between electrodes. Ruby particles were used for pressure

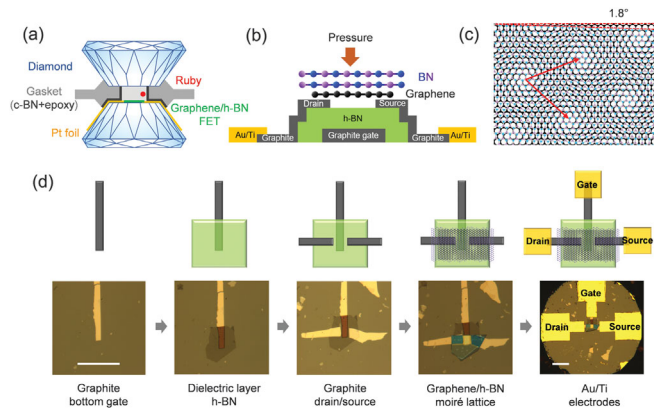


FIG. 1. Experiment schematic and fabrication process of the graphene/*h*-BN field effect transistor in DAC. (a) A schematic of the experimental setup. The graphene/*h*-BN field effect transistor is directly fabricated on the diamond culet and sealed inside the gasket chamber. Pt foils are manually mounted as feedthrough wires. The pressure is calibrated by ruby particles. (b) Schematic side view of the device. Graphite flakes are used as part of the electrodes. (c) A cartoon showing the graphene/*h*-BN moiré superlattices with twist angle of $\sim 1.8^\circ$. The red lines indicate the moiré wavelength. (d) Fabrication process of the device. All the 2D flakes are dry transferred. The diamond culet size is $300\ \mu\text{m}$ and the scale bars are $50\ \mu\text{m}$.

calibration. A bar-shaped, $\sim 50\ \text{nm}$ thick graphite flake was first transferred onto the diamond culet ($\sim 300\ \mu\text{m}$ in diameter) as the bottom gate. The dielectric *h*-BN and drain-source graphite flakes were then sequentially transferred onto the bottom graphite. The graphite flakes serve as part of the drain and source electrodes to extend the contacts to graphene, so that metal-electrode deposition could be done in the final step once for all to minimize the damage and enhance the yield. A monolayer graphene was aligned with and picked up by a top *h*-BN flake ($\sim 40\text{--}80\ \text{nm}$ thick) to obtain the moiré superlattices. Mechanically exfoliated graphene and *h*-BN tend to break along the zigzag or armchair axis. Therefore, the key to obtain a precisely controlled alignment is to optically identify MLG and *h*-BN flakes with straight edges and match the straight edges in the pickup process. A rotational stage was incorporated to provide the degree of freedom of controlling the relative alignment. The alignment angle was then estimated from the width of the Raman 2D peak as it is quantitatively established that a smaller-angle alignment leads to a wider Raman 2D peak [38]. More details about the twist-angle determination and the discussions of the possible experimental artifacts are in the Supplemental Material [39]. Then the graphene/*h*-BN stack was carefully transferred onto the dielectric *h*-BN. It is noted that the MLG was intentionally misaligned with the bottom dielectric *h*-BN layer, such that the moiré superlattices are defined only by the alignment angle of the MLG with the top *h*-BN. The MLG was then well protected by the two encapsulating *h*-BN layers [59]. Importantly, transferring of the aligned graphene/*h*-BN bilayer requires extremely careful positioning, such that the MLG is in contact with both the drain and source graphite flakes. All the 2D flakes were transferred using the conventional polymer-assisted dry transfer method to ensure clean interfaces [33,60]. The device was carefully sonicated in acetone after each transfer to remove the polymer residues and the surface cleanliness was confirmed with atomic force microscopy. Afterward, three blocks of Au/Ti ($70\ \text{nm}/8\ \text{nm}$) were patterned using photolithography and deposited as gate-drain-source electrodes onto the corresponding graphite flakes. Finally, narrow strips of Pt foil ($\sim 5\ \mu\text{m}$ thick) were manually mounted onto each Au/Ti electrode as feedthrough wires to external electronics. The actual fabrication process is summarized in Fig. 1(d). More details about the fabrication process are in the Supplemental Material [39]. This is a general and feasible device fabrication strategy that can be applied to preparation of other vdW heterostructure micro-devices in the DAC. Figure 2(a) shows current-voltage (*I*-*V*) characteristics of the $\sim 1.8^\circ$ twisted graphene/*h*-BN FET measured in the DAC at room temperature. The drain-source graphite flakes provide excellent Ohmic contact, as shown in Fig. 2(a). The *I*-*V* curves at different pressures all display good linearity, proving robustness of the FET device under high pressure.

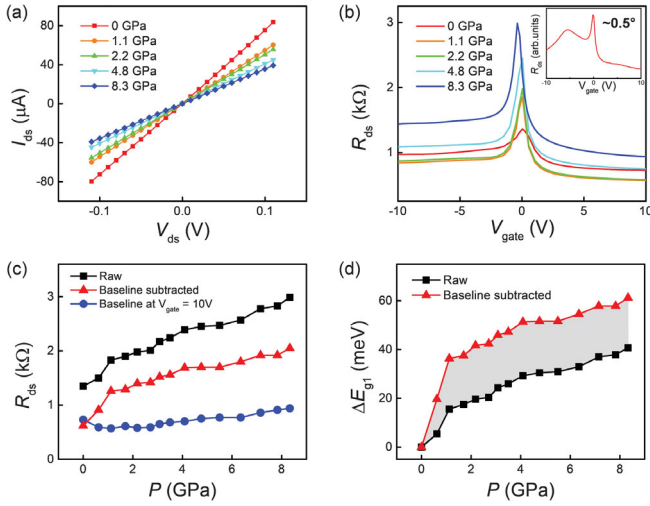


FIG. 2. Transport measurements of the $\sim 1.8^\circ$ twisted graphene/ h -BN FET in DAC. (a) Two-probe I - V curves of the FET in DAC. (b) Transfer curves of the same device in the DAC. Inset: transfer curve of a smaller-angle (0.5°) twisted graphene/ h -BN device recorded at ambient pressure. Clear electron-hole asymmetry is shown in both small and large twist-angle systems. (c) Drain-source resistance at the primary Dirac point, baseline resistance (at $V_{\text{gate}} = 10$ V) and the baseline-subtracted resistance at the primary Dirac point, under different pressures. (d) Pressure induced PDP band gap opening ΔE_{g1} with respect to the band gap at ambient pressure. The shadow area indicates the possible PDP band gap widening range considering the contact resistance.

Shown in Fig. 2(b) is the channel resistance (R_{ds}) of the FET as a function of gate voltage (V_{gate}) under different pressures. The peak near zero gate voltage is associated with the PDP, which increases from 1.3 k Ω at ambient pressure to 3 k Ω at 8.3 GPa, suggesting opening of a band gap (E_{g1}) at the PDP by the pressure. Another critical effect in Fig. 2(b) is the asymmetry of the baseline resistance under positive vs negative gate voltages. We have tested four other FETs with different twist angles and all FETs show such asymmetry. When the twist angle is small, the SDP shows up as a broad peak at both positive and negative V_{gate} , corresponding to the band gap of the SDP in the conduction and valence band, respectively [31]. This is also shown in the inset for a $\sim 0.5^\circ$ aligned graphene/ h -BN FET measured at ambient pressure and room temperature. It is clear that the SDP peak in the valence band is much stronger than in the conduction band. This asymmetry has been observed in previous experiments [12–14,31], and theoretically attributed to the difference between the wave functions in the conduction-valence bands combined with the inversion asymmetry introduced by the h -BN layer [34,36,61–63]. The inversion asymmetry creates two correlated terms in the perturbed Hamiltonian: the honeycomb sublattice site energy term and the intersublattice hopping term. These two sublattice-related terms will cancel (in the

conduction band) or add up (in the valence band) for the SDP gap, due to the opposite chiral convolutions of the bands. The SDPs will be broadened by temperature, disorder, and inhomogeneities, lifting up the baseline resistance between the PDP and the SDPs. As a consequence, the baseline of R_{ds} for negative V_{gate} (valence band) is usually higher than for the positive V_{gate} (conduction band). For this reason, we believe the asymmetric baseline in Fig. 2(b) is the residue of the SDPs in the case of large twist angles, and implies that moiré superlattices have indeed formed in the graphene. The SDP band gap is further discussed in conjunction with theoretical calculations.

It is noted that R_{ds} of our FETs was measured in a two-terminal configuration, so contact resistance is expected. However, one could estimate the lower limit of the real PDP peak resistance by subtracting the raw data with the baseline resistance, which can be taken at $V_{\text{gate}} = 10$ V where R_{ds} no longer changes with V_{gate} . The raw PDP peak resistance, baseline resistance at $V_{\text{gate}} = 10$ V, and the baseline-subtracted PDP peak resistance are plotted as a function of pressure in Fig. 2(c). Next, we use the PDP peak resistance to extract the band gap opening by using the Arrhenius dependence [20]:

$$\sigma = A e^{-(E_{g1}/2k_B T)}, \quad (1)$$

where σ is the channel conductivity calculated from R_{ds} , and A is the coefficient. Although all our measurements were done at room temperature, previous reports [20] have shown that the plot of $\ln \sigma$ vs $1/T$ of graphene/ h -BN moiré superlattices is linear under different pressures. More importantly, all the $\ln \sigma$ vs $1/T$ lines under different pressures tend to converge onto a single point on the $\ln \sigma$ axis, indicating that $\ln A$ is nearly pressure independent. Therefore, we estimate the change in E_{g1} with respect to E_{g1} in ambient pressure,

$$\Delta E_{g1} = -2k_B T * \ln \left(\frac{\sigma}{\sigma_0} \right), \quad (2)$$

where σ_0 is the PDP peak resistance at ambient pressure.

The extracted ΔE_{g1} values are plotted as a function of pressure in Fig. 2(d), where the values obtained from both raw R_{ds} and baseline-subtracted R_{ds} are shown. The actual ΔE_{g1} should lie between these bounds, as shown by the shaded area. The band gap at the PDP is expanded by 40–60 meV at 8.3 GPa. Using the previously reported experimental value of $E_{g1} \sim 30$ meV [31] for similar twist angles at ambient pressure, the final E_{g1} at 8.3 GPa is between 70 and 90 meV, enhanced by 2–3 times over the ambient value. Data above 8.3 GPa are not included here because h -BN undergoes a structural phase transition to c -BN at about 9 GPa, resulting in destruction of the moiré superlattices. The complete set of transport data is in Fig. S3.

To better probe the physical origin of the band gap opening in large-twist-angle graphene moiré superlattices, we performed theoretical calculations using models with inputs from *ab initio* density functional theory (DFT) computation. Fully relaxing the graphene/*h*-BN structure with a twist angle of $\sim 1.8^\circ$ and a fixed average interlayer space (h_0), the equilibrium pressure (P) at given h_0 is obtained and illustrated in Fig. S4. Subjecting to high pressures up to 10 GPa, the equilibrium h_0 is significantly reduced in the graphene/*h*-BN bilayer. The band structure of the relaxed graphene/*h*-BN under pressure was calculated by employing the effective lattice model of the moiré superlattices in graphene with the Hamiltonian,

$$H = \sum_i (H_{A,i} c_{A,i}^\dagger c_{A,i} + H_{B,i} c_{B,i}^\dagger c_{B,i}) - \sum_i \sum_{n=1}^3 t_{n,i} (c_{A,i}^\dagger c_{B,i,n} + c_{B,i,n}^\dagger c_{A,i}), \quad (3)$$

where $c_{\alpha,i}^\dagger$ ($\alpha = A, B$) is the creation and $c_{\alpha,i}$ is the annihilation operator of a p_z -like state at the site in the sublattice α and unit cell i of the MLG hexagonal lattice, $c_{B,i,n}^\dagger$ and $c_{B,i,n}$ are operators at the nearest sites of the sublattice-A atom in cell i , and the on-site energies and hopping terms are represented by $H_{\alpha,i}$ and $-t_{n,i}$, respectively. The hopping and on-site terms at a site with a local shift vector \mathbf{d} are expanded in Fourier series with the expansion parameters decaying exponentially with the local interlayer spacings as described in Supplemental Material [39]. For the considered system twisted by $\sim 1.8^\circ$, only a small gap of $E_{g1} = 8$ meV is opened at the PDP at ambient pressure, and bands around the SDPs in both the valence and conduction bands are significantly overlapped, as seen in Fig. 3(a) and Fig. S5(a) in the Supplemental Material [39]. Since this twisted system has a much smaller moiré supercell (moiré wavelength ~ 4 nm) compared to the perfectly aligned system (moiré wavelength ~ 14 nm), the sublattice asymmetry and the in-plane structural deformation are much less pronounced, leading to the smaller PDP gap [36,37]. With increasing pressure, the interlayer spacing decreases and the energy differences between domains with different stackings grow, resulting in stronger in-plane structural relaxation and larger domains with energetically favored stacking. Consequently, the sublattice asymmetry in the Hamiltonian becomes stronger, and the gap at the PDP increases. The calculated PDP gap (E_{g1}) vs pressure is plotted in Fig. 3(a), along with the experimentally evaluated band gap range. The calculated E_{g1} rises from 8 meV at ambient to 50 meV at 8.4 GPa, as shown in Fig. 3(a). The gap opening of $\Delta E_{g1} = 42$ meV is in good agreement with the experimental value. The band structures near the PDP at 0, 4.9, and 8.4 GPa are also plotted in Fig. 3(b). The gap is continuously opened by pressure whereas away from the gap the linear dispersion stays. Therefore, both experiments and calculations

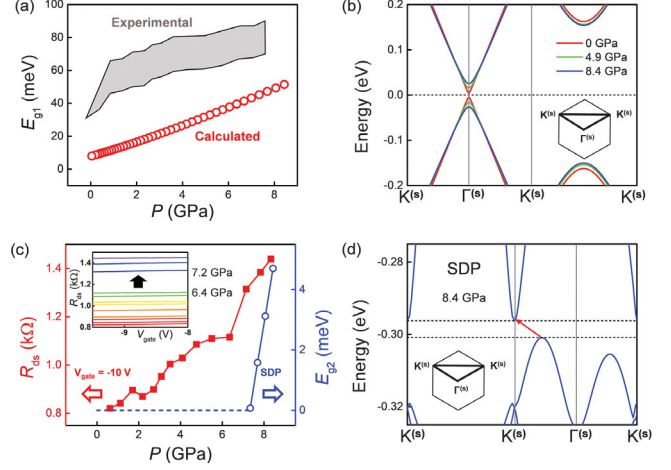


FIG. 3. Calculations of the band structure of the $\sim 1.8^\circ$ twisted system. (a) the calculated and experimental band gap at the PDP vs pressure. The red circles depict the band gap evolution obtained from theoretical calculations and the gray area is the estimated band gap range from Fig. 2(d) using the gap value of 30 meV at 0 GPa from Ref. [31]. (b) Calculated band structure near the PDP at 0 GPa, 4.9 GPa, and 8.4 GPa, respectively. Inset: the k path in the moiré Brillouin zone. (c) Calculated gap near the SDP in valence band and the drain-source resistance at $V_{gate} = -10$ V as a function of pressure. Blue dashed line indicates overlapped bands (i.e., zero gap). Inset: raw resistance data at $V_{gate} = -8$ V to -10 V. A none-zero SDP gap emerges at 7.3 GPa and an abrupt resistance increase on the hole side is also observed near 7 GPa. (d) Band structure near the SDPs in the valence band at 8.4 GPa. An indirect gap of 4.7 meV is opened.

demonstrate that high pressure is an effective tool to open the band gap of graphene/*h*-BN moiré superlattices even at rather large twist angle.

Moreover, in contrast to small-twist-angle systems that show a sizable direct gap at the SDP in the valence band, the bands around the SDPs now overlap due to the much weaker moiré potential, as shown in Fig. S5. The hydrostatic pressure reduces the band overlapping around the SDPs in the valence band and eventually opens an indirect gap (E_{g2}). The band gap opening at the SDPs is due to the enhanced moiré potential caused by the pressure, as the Hamiltonian parameters increase exponentially with decreasing interlayer spacing (details in Supplemental Material [39]). As shown in Fig. 3(c), under pressures higher than a critical value of 7.3 GPa, an indirect gap emerges in the valence band around the SDPs, and keeps increasing to a small but non-negligible value of 4.7 meV at 8.4 GPa. Even considering the usual underestimate of the band gap given by first-principle calculations, the indirect secondary gap of the $\sim 1.8^\circ$ system is likely to be quite narrow, which can explain why the SDPs in the large-twist-angle system have not been experimentally reported in previous studies. The narrow and indirect secondary gap is likely to generate a rather weak and broad secondary Dirac peak on the hole side, leading to a moderately flat baseline

[instead of an upward shape like the inset in Fig. 2(b)] even at high pressure. Interestingly, the resistance R_{ds} at $V_{gate} = -10$ V (probing the valence band) shows an abrupt rise at the pressure of about 7 GPa, as plotted also in the inset of Fig. 3(c). As illustrated both in the literature [31] as well as in Fig. 2(b), the hole side R_{ds} is a qualitative measure of the SDP peak resistance. Therefore, the abrupt rise of the hole side R_{ds} near 7 GPa is likely an experimental fingerprint to trace the SDP gap opening. The band structure near the SDPs in the valence band at 8.4 GPa is plotted in Fig. 3(d). Our calculations of the system show that the gap at the SDPs remains indirect even when the pressure reaches 16.4 GPa (Fig. S6). The indirect feature of the SDP gap could be attributed to the rather small moiré supercell in the considered graphene/*h*-BN structure, which makes the dispersion of the band right below the SDPs insensitive to pressure. It is also noted that pressure cannot open the SDP gap in the conduction band even at 16.4 GPa, as shown in Fig. S6, further testifying the particle-hole asymmetry in the system.

In conclusion, we demonstrate that large-twist-angle graphene/*h*-BN moiré superlattices can also host emergent electronic physics under mildly high pressures. For $\sim 1.8^\circ$ twisted superlattices, the band gap at the primary Dirac point is opened to ~ 70 – 90 meV at a pressure of 8.3 GPa. Therefore, we show that poorly aligned graphene/*h*-BN moiré superlattices can also serve as a platform for exotic physical effects, adding more freedom in preparation and investigation of graphene moiré superlattices. We also for the first time theoretically predict and experimentally trace the fingerprint of the secondary Dirac gap opened by pressure in large-twisted graphene superlattices, providing opportunities for a wide range tuning of the moiré electronics with pressure. The work shows that pressure can effectively unlock and control moiré physics in graphene superlattices, and similar tool could be used to probe emerging physics in heterostructures of transition metal chalcogenides.

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J. W. conceived the work. Y. G. conducted the experiments and data acquisition. X. L. and J. N. developed the theory and performed the *ab initio* calculations. P. C., T. S., and R. J. contributed to device and DAC preparations. K. W. and T. T. prepared the *h*-BN crystal. J. W., Y. G., X. L., and R. J. participated in writing the paper.

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