Observation of an intrinsic bandgap and Landau level renormalization in graphene/boron-nitride heterostructures

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Van der Waals heterostructures formed by assembling different two-dimensional atomic crystals into stacks can lead to many new phenomena and device functionalities. In particular, graphene/boron-nitride heterostructures have emerged as a very promising system for band engineering of graphene. However, the intrinsic value and origin of the bandgap in such heterostructures remain unresolved. Here we report the observation of an intrinsic bandgap in epitaxial graphene/boron-nitride heterostructures with zero crystallographic alignment angle. Magneto-optical spectroscopy provides a direct probe of the Landau level transitions in this system and reveals a bandgap of $\sim 38$ meV (440 K). Moreover, the Landau level transitions are characterized by effective Fermi velocities with a critical dependence on specific transitions and magnetic field. These findings highlight the important role of many-body interactions in determining the fundamental properties of graphene heterostructures.
Heterestructures consisting of two-dimensional (2D) layers of graphene, hexagonal boron-nitride (h-BN), MoS$_2$ and so on coupled by van der Waals interactions exhibit many intriguing physical properties and new device functionalities that are not achievable by individual constituting materials. In particular, graphene/h-BN heterostructures have shown great potentials for band structure engineering of graphene including inducing a bandgap (Fig. 1a), which is of great fundamental and technological interest. The coupling between graphene and h-BN results in a periodic moiré superlattice potential due to a 1.8% lattice mismatch, which gives rise to superlattice minibands and new Dirac points near the edges of the superlattice Brillouin zone. Furthermore, the local sublattice symmetry of graphene is broken owing to different local potentials produced by boron and nitrogen atoms (Fig. 1b), inducing a local bandgap. Although this effect varies spatially and is predicted to nearly disappear after spatial averaging, transport studies showed signatures of a global bandgap in these heterostructures. It is observed bandgap, but the issue remains unresolved.

Here we report the observation of a finite bandgap in epitaxially grown graphene/h-BN heterostructure with zero crystallographic rotation angle (Fig. 1c; see Methods section) and so on coupled by van der Waals interactions. The coupling between graphene and h-BN results in a periodic moiré superlattice potential due to a 1.8% lattice mismatch, which gives rise to superlattice minibands and new Dirac points near the edges of the superlattice Brillouin zone. Furthermore, the local sublattice symmetry of graphene is broken owing to different local potentials produced by boron and nitrogen atoms (Fig. 1b), inducing a local bandgap. Although this effect varies spatially and is predicted to nearly disappear after spatial averaging, transport studies showed signatures of a global bandgap in these heterostructures. It is suggested that many-body interactions may be responsible for the observed bandgap, but the issue remains unresolved experimentally.

At zero magnetic field, the energy dispersion of graphene with a bandgap $\Delta$ is 

$$E_n = \text{sgn}(n) \sqrt{2e \hbar v_F |B| n} + (\Delta/2)^2, \quad (2)$$

where $e$ is the elementary charge, $\hbar$ is Planck’s constant divided by $2\pi$, the integer $n$ is LL index, and $\text{sgn}(n)$ is the sign function. The LLs for gapped graphene have the form:

$$E_n = \pm (\Delta/2) \delta_{n,0} + \text{sgn}(n) \sqrt{2e \hbar v_F |B| n} + (\Delta/2)^2, \quad (2)$$

which features two zeroth LLs labelled as $n = 0$ and $n = -0$ with energies of $E_{\pm 0} = \pm \Delta/2$. Here $\delta$ is the Kronecker delta function. Therefore, the bandgap of graphene can be explored by probing its LL energy spectrum.

Our study provides a direct spectroscopic determination of the bandgap in epitaxial graphene/h-BN heterostructures from optical measurements of LL transitions. We observe an intrinsic bandgap of ~38 meV (440 K) in this system, which is comparable to the gap value found in transport studies. Moreover, we find different values of effective Fermi velocity for different LL transitions, indicating LL renormalization by interaction effects. These findings have broad implications for the fundamental understanding of graphene heterostructures and their potential applications.

### Results

**Transmission spectra in magnetic field.** Infrared (IR) transmission spectra $T(B)$ were measured in magnetic field applied perpendicular to the samples as shown in Fig. 1d (see Methods section). Figure 2 depicts the $T(B)/T(B_0)$ spectra for a...
representative sample, where \(B_0 = 0\) T. Data for more samples are shown in Supplementary Fig. 1. Three dip features denoted as \(T_1\), \(T_2\) and \(T_3\) are observed, all of which systematically shift to higher energies with increasing magnetic field. The zero-field transmission spectrum \(T(B_0)\) of either pristine or gapped graphene shows a step-like feature without any sharp resonances in the energy range explored here, so the observed dip features in the \(T(B)/T(B_0)\) spectra are corresponding to transmission minima in \(T(B)\) and thus absorption peaks in magnetic fields (Supplementary Fig. 2 and Supplementary Note 1).

The effective bulk mobility of our samples estimated from the widths of the resonances in the optical spectra is higher than 50,000 cm\(^2\) V\(^{-1}\) s\(^{-1}\) (Supplementary Fig. 3 and Supplementary Note 2). Our optical data also indicate that the Fermi energy for our samples is in the range of \(E_F < 19\) meV (Supplementary Note 3).

**Observed LL transitions.** The energies \((E)\) of all observed features in graphene/h-BN exhibit an approximate linear dependence on \(\sqrt{B}\) (or equivalently, \(E^2\) has an approximate linear dependence on \(B\)) in our spectral range as shown in Fig. 3. However, they all show non-zero energy intercepts at zero magnetic field under linear extrapolations, in stark contrast to the LL transitions of pristine graphene described by equation (1), which converge to zero energy at zero field\(^2\)\(^,\)\(^3\)\(^,\)\(^9\)\(^,\)\(^30\).

Similar behaviours were observed in all five samples we have measured (Supplementary Fig. 1). Within the non-interacting single-particle picture, the finite zero-field extrapolation values of all observed absorption energies suggest that the LLs of our graphene/h-BN samples are described by equation (2). From the selection rule\(^3^1\) for allowed optical transitions from \(LL_n\) to \(LL_{n'}\), \(\Delta n = |n| - |n'| = \pm 1\), and a quantitative comparison with equation (2), we assign feature \(T_1\) to transitions of \(LL_1 \rightarrow LL_0\) and \(LL_0 \rightarrow LL_{-1}\) (Fig. 2b). With an energy given by:

\[
E_{T_1} = \sqrt{2\hbar v_F^2 B + (\Delta/2)^2} \mp \Delta/2
\]

(Fitting the \(T_1\) feature based on equation (3) from a least squares fit yields a bandgap \(\Delta \approx 38 \pm 4\) meV and an effective Fermi velocity \(v_F^1 \approx (0.96 \pm 0.02) \times 10^6\) m s\(^{-1}\) (Supplementary Table 1 and Supplementary Note 4). We emphasize that the bandgap explored here is at the main Dirac point of graphene instead of the secondary Dirac points at the edges of graphene/BN superlattice Brillouin zone\(^8\)\(^–\)\(^12\). The \(T_1\) transition energies in Fig. 2 are well below the energy (\(~200\) meV) of the secondary Dirac points\(^9\)\(^–\)\(^12\), so the LLs in this energy region are not significantly affected by the strong band structure modifications at the boundary of the superlattice Brillouin zone, which is supported by the observed linear \(\sqrt{B}\) dependence of the LL transition energy.

The observed \(T_2\) and \(T_3\) features have higher energies compared with \(T_1\) features and can be assigned as (Fig. 2b):
Figure 3 | LL transition energies of graphene/h-BN. (a) All observed transitions shown in a $E^2$-B plot. Symbols: data for sample 1 and 2. Solid lines: best fits to the data for sample 1 using equations (3)-(5) and parameters discussed in the text. $\Delta = 38\,\text{meV}$ and $v^f_{\text{ch}} \approx 1.20 \times 10^6\,\text{m/s}$ are used for the fit to $T_3$ transition shown here. (b) The low energy part of (a) to highlight the extraction of the gap. Dashed line: a guide for eye showing linear extrapolation of the data. The error bars in both panels, $\delta(E^2)$, are calculated as $\delta(E^2) = 2\delta(E)/\delta(E)$, where $\delta(E)$ is the uncertainty in determining the energy of each LL transition from the $T(B)/T(B_0)$ spectra.

Figure 4 | Many-body effects on LL transitions for pristine and gapped graphene. (a) Energy ratios of different LL transitions for graphene on SiO$_2$ (blue colour) and graphene/h-BN sample 1 (red colour) shown in a common vertical scale. Open symbols: $E_r/(\sqrt{2} + 1)E_r$. Solid symbols: $E_r/(\sqrt{3} + \sqrt{2})E_r$. Red solid line: theoretical result of $E_r/(\sqrt{2} + 1)E_r$ based on equations (3) and (4) for gapped graphene. The ratios for graphene on SiO$_2$ are $>1$, which is a signature of interaction effects in pristine graphene (Supplementary Note 5). On the other hand, the ratios for graphene/h-BN exhibit an entirely different behaviour, which is consistent with gapped graphene. The error bars of energy ratios are calculated using standard formulas for propagation of uncertainty for division based on the uncertainty in determining the energy of each LL transition from the $T(B)/T(B_0)$ spectra. (b) Fermi velocity ratios of different LL transitions for graphene/h-BN with a constant $v^f_{\text{ch}} \approx (0.96 \pm 0.02) \times 10^6\,\text{m/s}$. For $T_2$ transition, a constant Fermi velocity $v^f_{\text{ch}} \approx (1.20 \pm 0.01) \times 10^6\,\text{m/s}$ is extracted from the data. These ratios are distinct from the value expected from the single-particle picture, which is indicative of interaction effects in gapped graphene. The error bars indicate the range of Fermi velocity values that could fit the data in Fig. 3 (Supplementary Note 4).

$T_2$, $LL_{-2} \rightarrow LL_{+1}$ and $LL_{-1} \rightarrow LL_{+2}$; $T_3$, $LL_{-3} \rightarrow LL_{+2}$ and $LL_{-2} \rightarrow LL_{+3}$. Their energies are described by:

$$E_{T_2} = \sqrt{2E_h v^f_B B + (\Delta/2)^2} + \sqrt{(2E_h v^f_B B) \times 2 + (\Delta/2)^2}$$ (4)

$$E_{T_3} = \sqrt{(2E_h v^f_B B) \times 2 + (\Delta/2)^2} + \sqrt{(2E_h v^f_B B) \times 3 + (\Delta/2)^2}$$ (5)

The energies of $T_2$ transition show a deviation from linear $\sqrt{B}$ dependence above 4T (or 220 meV in energy) as shown in Fig. 3, with $E^2$ deviating from a linear $B$-dependence, which is perhaps due to the moiré superlattice or many-body interactions. Therefore, we focus on the low field ($< 4\,\text{T}$) region where the $T_2$ transition exhibits an overall linear $\sqrt{B}$ dependence, which most likely arises from the intrinsic behaviours of gapped graphene alone. We observed a splitting of the $T_2$ transition near 169 meV owing to the coupling to the IR active phonon of h-BN, which nonetheless does not affect the main conclusions of our analysis because this effect only occurs in a very narrow field and energy range. Based on equation (4), we find that the $T_2$ transition in low field is consistent with a bandgap similar to that extracted from the $T_1$ transition, $\Delta \approx 38 \pm 4\,\text{meV}$, and an effective Fermi velocity $v^f_{\text{ch}} \approx (1.20 \pm 0.01) \times 10^6\,\text{m/s}$ (Supplementary Fig. 4 and Supplementary Note 4). The $T_3$ transition is discussed in details below and in Supplementary Fig. 5 and Supplementary Note 4.

Comparison with pristine and gapped graphene. We stress that our data on graphene/h-BN cannot be explained by many-body effects of pristine graphene (Supplementary Fig. 6 and...
Supplementary Note 5). One prominent feature of interaction effects in pristine graphene is that the effective Fermi velocity varies for different LL transitions, so that the energy ratios \( E_{T_1}/(\sqrt{2}+1)E_{T_1} \) and \( E_{T_2}/(\sqrt{3}+\sqrt{2})E_{T_2} \) are higher than one, as demonstrated by previous IR studies \(^{29}\) and our data on graphene on SiO\(_2\). However, the data for graphene/h-BN exhibit an entirely different behaviour (Fig. 4a) compared with interaction effects in pristine graphene. Instead, the energy ratios of different LL transitions for graphene/h-BN are consistent with the behaviours of gapped graphene. A theoretical result of \( E_{T_1}/(\sqrt{2}+1)E_{T_1} \) based on equations (3) and (4) is shown in Fig. 4a, with \( \Delta \approx 38 \text{ meV} \), \( v_F = 0.96 \times 10^6 \text{ m s}^{-1} \) and \( v_F^1 = 1.20 \times 10^6 \text{ m s}^{-1} \), which agrees very well with the experimental results.

**Discussion**

Previous transport measurements on graphene/h-BN heterostructures indicated a gap of \(~300 \text{ K at 0.4}\) \(^\circ\) crystallographic rotational angle (\( \theta \)) \(^{11}\). A recent study \(^{13}\) reported the existence of large domains of graphene with the same lattice constant as h-BN separated by domain walls with concentrated strain for small \( \theta \), and a gap of 360 K was found for \( \theta = 0^\circ \). Our optical study provides a direct spectroscopic determination of the bandgap with similar magnitude (\(~440 \text{ K}) in epitaxial graphene/h-BN heterostructures. This bandgap value is larger than those found in theories within the single-particle picture \(^{18-20}\), which suggests the relevance of many-body interactions in generating the gap \(^{15,16}\). It was argued that the gap at the Dirac point is greatly enhanced by interaction effects due to coupling to a constant sublattice-asymmetric superlattice potential \(^{15}\), which is not affected by the spatial variations shown in Fig. 1b. The intrinsic gap value for graphene/h-BN obtained in our study provides a critical input for the basic understanding of the gap in this system.

Our study further reveals the crucial role of many-body interactions in renormalizing LL transitions \(^{32}\) in graphene/h-BN heterostructures. Specifically, the effective Fermi velocity associated with the LL transitions varies with particular transitions as well as the magnetic field. We find that the T\(_3\) transition cannot be consistently fitted by equation (5) using a constant \( v_F \), so we use an effective field-dependent parameter \( v_F^1 (B) \) to describe this transition (Supplementary Note 4). Figure 4b depicts the Fermi velocity ratios \( v_F^1/v_F^0 \) and \( v_F^1 (B)/v_F^1 \), both of which are higher than one and therefore very different from the constant \( v_F \) for all transitions expected from single-particle pictures. Intriguingly, \( v_F^1 (B)/v_F^1 \) shows a systematic increase in low magnetic fields. For T\(_2\) transitions (consider LL\(_{ -1 } \rightarrow \text{LL}\(_{ +2 }\)), for example, it shows \( v_F^1 \sim 1.20 \times 10^6 \text{ m s}^{-1} \) even at 1 T field with \( E_{T_1} = 115 \text{ meV} \) (Supplementary Fig. 1c), which corresponds to \( E_{\text{LL}_{+2}} \approx 66 \text{ meV} \) and \( E_{\text{LL}_{-1}} \approx 49 \text{ meV} \). According to theoretical studies \(^{33}\), the band structure of graphene/h-BN at such low energy scales are quite linear and not strongly modified by the superlattice Dirac points (\(~200 \text{ meV}) refs 9–12), so the value \( v_F^1 \sim 1.20 \times 10^6 \text{ m s}^{-1} \) extracted from data at low magnetic field (thus low energy) is little affected by the superlattice Dirac points. Similar argument can be made for T\(_1\) and T\(_2\) transitions at low magnetic fields and low energy. Note that the LL transitions at high field and high energy (for instance, T\(_2\) transition above 4 T field) may be affected by the band structure modification due to the superlattice Dirac points \(^{8,33}\), but our discussions here are only focused on the low field regime shown in Fig. 4b. Our results in Fig. 4b indicate LL renormalization due to many-body interactions in magnetic field. Theoretical studies \(^{34-36}\) showed that interaction effects of electron-hole excitations between LLs, such as direct Coulomb interactions between the excited electrons and holes and the exchange self-energy of electrons and holes between LLs, can significantly renormalize the inter-LL transition energy. The observation shown in Fig. 4b in gapped graphene is qualitatively similar to the results from many-body theories \(^{34-37}\) as well as experimental studies \(^{29}\) on pristine graphene, so our results strongly suggest contributions of many-body effects to the inter-LL transitions \(^{34-37}\). Further theoretical investigations are required to quantitatively understand these interactions in gapped graphene, with many open questions yet to be addressed such as the role of superlattice potential \(^{15}\) and bond distortion \(^{20}\) in graphene/h-BN heterostructures.

Multi-valley (band extrema in momentum space) Dirac systems such as gapped graphene, silicene and 2D transition metal dichalcogenides are described by the same Dirac Hamiltonian and share several essential properties such as valley-dependent orbital magnetic moment and Berry curvature \(^{23,38}\), which are intimately related to their unconventional valley-dependent LL structures \(^{24-46}\). In this context, the strong LL renormalization observed here has broad implications for fundamental studies of many novel phenomena related to LLs in these Dirac materials, such as magnetic control of valley degree of freedom \(^{38}\) and valley-spin-polarized magneto-optical response \(^{39,40}\).

In summary, we have observed a bandgap of \(~38 \text{ meV} \) (440 K) in graphene/h-BN heterostructures with zero crystallographic rotation angle using magneto-optical spectroscopy. The intrinsic gap value reported here is important for fundamental understanding of the bandgap and many-body interaction effects in this system. Our demonstration of a finite bandgap in epitaxial graphene/h-BN heterostructures can also lead to novel applications in electronics and optoelectronics.

**Methods**

**Sample preparation and characterization.** h-BN was mechanically exfoliated onto double-side-polished SiO\(_2\)/Si substrates with 300 nm SiO\(_2\). Graphene was epitaxially grown on h-BN by remote plasma enhanced chemical vapour deposition \(^{12}\). Some multilayer graphene can be found on monolayer graphene in as-grown samples, so hydrogen plasma etching technique \(^{22}\) was applied to reduce these additional graphene. The resulting sample is continuous monolayer graphene with minor etched hexagonal pitches in plane, as shown in Fig. 1c. The moiré pattern (Fig. 1c) due to lattice mismatch shows a periodicity of 15 ± 1 nm as measured by atomic force microscopy, indicating zero crystallographic alignment angle between graphene and h-BN \(^{12}\). This moiré pattern is observed over the entire areas of all samples, establishing these epitaxial samples as single-crystalline and single-domain graphene heterostructures. The samples studied in this work have typical lateral sizes of \(~100 \mu\)m. The observed LL transitions indicate that the optical absorption of our samples is little affected by defects or grain boundaries. The effective mobility of our samples estimated from the widths of the resonances in the optical spectra is higher than 50,000 cm\(^2\) V\(^-1\) s\(^{-1}\) (Supplementary Fig. 3 and Supplementary Note 2). The absence of the LL\(_{ -1 } \rightarrow \text{LL}\(_{ +2 }\) and LL\(_{ -1 } \rightarrow \text{LL}\(_{ +1 }\) transitions in our optical data indicates that the Fermi energy is within the gap for our samples, namely \( E_F < 19 \text{ meV} \) (Supplementary Note 3).

**Magneto-transmission measurements.** The measurements were performed at \(~4.5 \text{ K}\) in a superconducting or resistive magnet in the Faraday geometry (magnetic field perpendicular to the sample surface). IR light from a Fourier transform spectrometer is delivered to the sample using a copper light pipe, and the light transmitted through the sample is detected by a composite Si bolometer. The focus of the IR light on the sample is \(~0.5-1 \mu\)m. To reduce the stray light around our small samples, an aluminium aperture of \(~200 \mu\)m diameter was placed around the sample. We report data at energies above 60 meV corresponding to wavelengths shorter than 20 \mu m, which ensures that the wavelength is significantly smaller than the sizes of the samples, and therefore a macroscopic description of the data using optical constants is applicable.

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Author contributions
Z.-G.C. and Z.S. initiated the optical studies; Z.-G.C. carried out the optical experiments; Z.S. and Y.L. participated in part of the measurements; W.Y. and X.L. grew and characterized the graphene/bn BN sample; H.Y. provided the graphene on SiO2 samples; F.W., G.Z. and Z.L. supervised the project; Z.-G.C. and Z.L. analyzed the data and wrote the manuscript. All authors discussed the results and commented on the paper.

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