Band Gap Opening in Bilayer Graphene-CrCl$_3$/CrBr$_3$/CrI$_3$ van der Waals Interfaces

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We report experimental investigations of transport through bilayer graphene (BLG)/chromium trihalide (CrX$_3$; X=Cl, Br, I) van der Waals interfaces. In all cases, a large charge transfer from BLG to CrX$_3$ takes place (reaching densities in excess of $10^{13}$ cm$^{-2}$), and generates an electric field perpendicular to the interface that opens a band gap in BLG. We determine the gap from the activation energy of the conductivity and find excellent agreement with the latest theory accounting for the contribution of the $\sigma$ bands to the BLG dielectric susceptibility. We further show that for BLG/CrCl$_3$ and BLG/CrBr$_3$ the band gap can be extracted from the gate voltage dependence of the low-temperature conductivity, and use this finding to refine the gap dependence on the magnetic field. Our results allow a quantitative comparison of the electronic properties of BLG with theoretical predictions and indicate that electrons occupying the CrX$_3$ conduction band are correlated.

Van der Waals (vdW) interfaces provide a vast playground for creating new systems with engineered electronic properties, by stacking suitably chosen atomically thin crystals (or 2D materials) on top of each other. Examples include hexagonal Boron Nitride (hBN) encapsulation of graphene [1–3], proximity induced spin-orbit coupling in graphene on semiconducting transition metal dichalcogenide substrates [4–10], or the creation of so-called $\Gamma$–$\Gamma$ interfaces [11, 12]. Recently, the discovery of 2D magnets and their use in vdW heterostructures has further broadened the scope of phenomena that can be explored [13–18]. In these systems, the wave functions of electrons in the non-magnetic material extend into the magnetic one and experience some of the magnetic interaction, enabling magnetism to be proximity induced in graphene or other 2D materials [13, 19–28]. However, deterministically controlling magnetism by proximity and predicting what aspect of magnetism can be induced into non-magnetic materials are challenges that remain to be solved because many different phenomena – such as strain, hybridization, charge transfer, and more – occur simultaneously at van der Waals interfaces [15, 29–31], influencing the interfacial electronic properties. In particular, electrostatic effects often dominate the behavior of heterostructures formed by low-charge-density systems, such as 2D semiconductors and semimetals. As a result, significant charge transfer can occur and lead to new phenomena mediated by changes in electron concentration or orbital occupation [32]. Indeed, recent work reported spin-dependent interlayer charge transfer in magnetic vdW heterostructures [33–35] and concluded that its detailed analysis is of key importance for improving the control of interfacial properties.

Here, we report the systematic behavior of vdW interfaces formed by bilayer graphene (BLG) [36, 37] and chromium trihalide crystals (CrX$_3$; X=Cl, Br, I) [38–45]. All systems exhibit a large transfer of electrons from graphene to the magnetic material, reaching values in excess of $10^{13}$ cm$^{-2}$, producing a large electric field perpendicular to the interface and a gap in BLG. When gating the BLG at the charge neutrality point (CNP), the gap induces a low-temperature suppression of the conductance of four orders of magnitude or more, exhibiting a sharp onset as a function of gate voltage. We determine the size of the band gap by analyzing the temperature dependence of the transfer curves (i.e., conductance-vs-gate voltage), and find excellent agreement with the results of the latest ab initio calculations of the electrostatically induced gap in BLG, which include dielectric screening due to the polarizability of the $\sigma$ bonds in the graphene lattice [46]. We also find that the gap in BLG can be determined by looking exclusively at the low-temperature gate voltage dependence of the conductance, a result that provides information about the nature of the electronic states in the CrX$_3$, and that enables the quantitative determination of the dependence of the BLG band gap on the applied magnetic field.

Figure 1a shows an optical microscope image of a...
FIG. 1. Band gap opening in BLG/CrX₃ interfaces. (a) Optical micrograph and (b) schematics of a representative device (the scale bar in (a) is 20 µm), based on a BLG/CrX₃ heterostructure encapsulated in hBN. A metallic gate electrode is deposited onto the top hBN layer, and is coupled to two distinct regions: a central part formed by the BLG/CrX₃ interface (region 1) and two adjacent parts where BLG is in contact only with hBN (region 2). Transport is measured using metallic source (S) and drain contacts (D) and probes the two regions connected in series. (c) Square conductance $G_\square$ as a function of gate voltage $V_G$ measured in a heterostructure of BLG-on-CrCl₃ at 250 mK (green curve, top). Two characteristic features are visible in the transfer curve: a small conductance dip close to $V_G = 0$ V corresponding to the CNP of graphene in region 2 ($V^{(2)}_{\text{CNP}}$) and a pronounced suppression (four orders of magnitude) at large $V_G$ ($V^{(1)}_{\text{CNP}}$) that originates from gating BLG-on-CrCl₃ (i.e., region 1) to charge neutrality. The black dashed lines represent linear extrapolations to extract the threshold voltages for holes $V_{h_{\text{th}}}$ and electrons $V_{e_{\text{th}}}$.

The observed charge transfer generates a strong electric field perpendicular to the interface that causes the opening of a band gap in BLG (10⁻¹³ cm⁻²). Analogous behavior and a large hole doping in BLG (see Section S1 of the Supporting Information for transfer curves in a broader range of gate voltages) are also observed in heterostructures formed by BLG and CrBr₃ (light-blue curve) and in BLG-on-CrI₃ (red curve; in agreement with the earlier observations [32, 48]).
BLG/CrX₃ interfaces, indicating that charge transfer from CrX₃ to BLG is rather homogeneous. To compare quantitatively the experimental observations made in heterostructures based on the different CrX₃, we convert the applied gate voltage to the corresponding accumulated charge density $n$, as $n = \frac{\varepsilon}{t \varepsilon_\text{rel}} \frac{V_{G \text{CNP}} - V_{\text{CNP}}^{(1)}}{V_{\text{CNP}}^{(2)} - V_{\text{CNP}}^{(1)}}$ ($\varepsilon$ and $t$ are the relative dielectric constant and thickness of the hBN layer, and $V_{\text{CNP}}^{(1)}$ is the gate voltage corresponding to the CNP in region 2, see Figure 1c). The concentration of electrons transferred from BLG in CrX₃ is then given by $\Delta n = \frac{\varepsilon}{t \varepsilon_\text{rel}} \frac{V_{G \text{CNP}} - V_{\text{CNP}}^{(1)}}{V_{\text{CNP}}^{(2)} - V_{\text{CNP}}^{(1)}}$ (where $V_{\text{CNP}}^{(1)}$ is the gate voltage corresponding to the CNP of BLG-on-CrX₃, i.e. in region 1; see Figure 1c again), and is directly proportional to the displacement field present at the BLG/CrX₃ interfaces, $D = e \Delta n$ (we use this relation to calculate the values of the displacement field in Figure 3).

We determine the size of the band gap from the temperature ($T$) evolution of the charge transfer curves, plotted in Figure 2a-c for CrCl₃, CrBr₃, and CrI₃, respectively. Data are shown for selected values of $T$ between 250 mK (blue curve) and 250 K (red). The activation energies for the three heterostructures are extracted by looking at the minimum square conductance $G_{\text{min}}$ (corresponding to $G_{\square}$ at the CNP of BLG/CrX₃), by fitting the linear part of the Arrhenius plot in the high-temperature range, where the charge carriers are dominated by a thermally activated behavior (see Figure 2d). The larger activation energy $E_a$ is observed in BLG-on-CrCl₃ (green hexagons) where the gap ($E_a = 2E_a$) is estimated to be 162 meV; band gaps of 124 meV and 108 meV are found for BLG-on-CrI₃ (red squares) and on CrBr₃ (light-blue circles), respectively.

The same measurements show that the position of the CNP, i.e., the density of charge transferred from BLG to CrX₃, is temperature-dependent (as summarized in Figure 2e), implying that the perpendicular electric field responsible for the opening of the band is not constant as $T$ is varied. For BLG-on-CrCl₃ (green) and on CrBr₃ (light-blue) the position of CNP changes by less than 10% throughout the full range investigated, and by significantly less over the range used to determine the size of the band gap, so that the effect can be disregarded. For CrI₃ (red) the change is larger, corresponding to a more sizable indetermination for the electric field value responsible for the opening of the gap in BLG. The precise microscopic origin of the $T$ dependence of CNP in BLG-on-CrI₃ is currently not understood, and is likely determined by the electronic properties of CrX₃, which are materials with very narrow bands, whose behavior deviates from that of conventional semiconductors (see also the below discussion on the determination of the gap from the gate voltage dependence of the transfer curves).

The band gap dependence on the electric field for the three different BLG/CrX₃ interfaces is compared to the calculated gap in Figure 3. The electric field dependence of the gap predicted for $\varepsilon_z = 2.6$ –corresponding to the theoretically expected dielectric susceptibility when accounting for the polarizability of the $\sigma$ bands– is represented by the blue line. For comparison we also show calculations with $\varepsilon_z = 1$ (grey line) as in [37]. The
on the analysis of the low-temperature G dependence of the square conductance, used in earlier experiments [54]. From analyzing our data, we find that for BLG on both the temperature dependence of the charge transferred from BLG to CrX termination on the electric field due to the temperature experimental data, and the error bar denotes the inde-

dependence of the conductance on ε proposed in [46], and deviate very significantly from with theory that considers ε as just discussed above. These data agree perfectly with the agreement with the empty symbols of different colors (CrCl3, green hexagon; CrI3, red square; CrBr3, light-blue circle) represent our experimental data, and the error bar denotes the inde-
terface. The continuous lines represent the band gap as a function of displacement field D predicted by ab initio calculations, considering or ignoring the contribution to the dielectric susceptibility ε due to the electrons that occupy the σ band of BLG [46]. The empty symbols represent the experimental data obtained from the temperature dependence of the conductance measured in our devices. It is apparent that the experimental data are in excellent agreement with theoretical prediction for ε = 2.6. The error bars for the displacement field (D = eΔn, see main text) correspond to the variation of charge transferred from BLG to CrX3 (and consequently of D) as temperature is varied. Filled symbols indicate the experimental values of E extracted from the threshold voltages of low-temperature transfer curves using Equation (1). For CrCl3 and CrBr3, the agreement with the gap values obtained from the temperature-dependent measurements is excellent.

Having determined the band gap from the analysis of the temperature dependence of the square conductance, we now present an alternative way that relies exclusively on the analysis of the low-temperature G-vs-VG curves. From analyzing our data, we find that for BLG on both CrCl3 and CrBr3, the gap is quantitatively given by

\[ E_g = \frac{C (V_{th}^e - V_{th}^h)}{e \rho_{BLG}} \]  (1)

Here, V_{th}^e and V_{th}^h are the threshold voltages for electron and hole conductance (obtained by extrapolating to zero the conductance measured as a function of gate voltage, as illustrated by the dashed lines in Figure 1c) and \( \rho_{BLG} = 2m^*/\pi \hbar^2 \) is the density of states in gapless BLG contacts (i.e., region 2 in Figure 1b), next to the gapped region where BLG is on the CrX3 layer (region 1). Equation (1) gives the gap values represented with filled symbols in Figure 3, in perfect agreement with the values obtained from the T dependence of the minimum conductance for both BLG-on-CrCl3 and BLG-on-CrBr3; for BLG-on-CrI3, instead, Eq. (1) gives a value that deviates by nearly a factor of 2 from the correct one. We discuss below the origin of Eq. (1), the condition for its validity, and why it fails to give the correct value of the gap for CrI3.

The possibility to use Eq. (1) to extract the band gap of BLG enables the detailed dependence of the gap on the magnetic field, not yet addressed in previous studies, to be probed in an experimentally straight-forward way. To this end, it suffices to measure the conductance as a function of gate voltage for different values of the magnetic field, as shown in Figure 4a for a BLG-on-CrCl3 device. The left and right panels zoom in on the onset of threshold for both electron and hole conduction, which shift upon increasing the applied magnetic field, resulting in a decrease in \( (V_{th}^e - V_{th}^h) \), and therefore a decrease in the BLG band gap. The full dependence of the gap on B for BLG-on-CrCl3 and for BLG-on-CrBr3 (represented by the green hexagons and light-blue circles, respectively) is compared to the theoretically calculated dependence (orange line) in Figure 4b. Theory predicts that the gap decreases as a result of the formation of Landau levels [1], which causes the top of the valence band to increase in energy and the bottom of the conduction band to decrease (see Figure 4c). This is a counter-intuitive behavior that contrasts what would be expected for electrons in a conventional two-dimensional electron gas (i.e., electrons described by a scalar wave function, for which the formation of Landau levels would lead to an increase in the gap at finite B). At a quantitative level, a change in the gap between 10% and 15% is expected as B increases up to 13 T, which results in perfect agreement with experiments without the need to introduce any free fitting parameters. To confirm the soundness of this result and exclude significant contributions of other mechanisms to the observed magnetic field dependence of the band gap, we also considered whether the charge transferred from BLG to CrX3 depends on the magnetic field. This is important because a change in charge transfer would lead to a corresponding change in the perpendicular electric field and thus in the size of the band.
Such excellent quantitative agreement shows the usefulness of Eq. (1) and confirms its validity. To understand heuristically the origin of Eq. (1) we look at how the electrostatic and electrochemical potentials vary in the different regions of our devices (see Figure 1b), i.e., in region 1 where BLG is in contact with the CrX$_3$ layer and in region 2 where BLG is on hBN. A change $\Delta V_G$ in applied gate voltage causes a variation in the electrochemical and electrostatic potentials $\Delta \mu$ and $\Delta \phi$ in both regions, with the two quantities related by $\Delta \mu = e\Delta \phi + \Delta E_F$ ($\Delta E_F$ is the change in Fermi energy induced by the variation in the density of accumulated electrons in BLG, i.e., $\Delta E_F = C\Delta V_G/e\rho_{BLG}$). Since the entire structure is at equilibrium for all gate voltages, the change in electrochemical potential is uniform, such that $\Delta \mu^{(1)} = \Delta \mu^{(2)}$, or $e\Delta \phi^{(1)} = e\Delta \phi^{(2)} + \Delta E_F^{(2)}$. Whenever the electrochemical potential in region 1 is inside the gap of BLG—and at sufficiently low temperature $\Delta E_F^{(1)} = 0$, because no states are available to add charge. Under these conditions, therefore, a variation in gate voltage only changes the electrostatic potential in region 1, so that we have $e\Delta \phi^{(2)} = \Delta E_F^{(2)}$. As $\Delta E_F^{(2)} = C\Delta V_G/e\rho_{BLG}$, we obtain $e\Delta \phi^{(1)} = e\Delta \phi^{(2)} = C\Delta V_G/e\rho_{BLG}$, a relation that determines the relative band alignment between region 1 and 2. While we sweep the gate voltage from $V_G = V_{th}^h$ to $V_G = V_{th}^e$, this relation always holds, because throughout this $V_G$ interval the electrochemical potential in region 1 is inside the gap. Since at $V_G = V_{th}^h$ the electrochemical potential in region 2 is aligned with the valence band edge in region 1 and at $V_G = V_{th}^e$ the electrochemical potential in region 2 is aligned with the conduction band edge in region 1, we obtain $E_g = e\Delta \phi^{(1)} - e\Delta \phi^{(2)}$, and Eq. (1) then follows directly using that $e\Delta \phi^{(1)} - e\Delta \phi^{(2)} = C\Delta V_G/e\rho_{BLG}$. We interpret this result by saying that a change in $V_G$ lowers the bands of BLG in region 1 and in region 2 (which effectively forms the source and drain contacts to the transistor channel), but changes $E_F$ only in region 2 (because only region 2 is gapless), and it is this change in Fermi energy that shifts the electrochemical potential from the valence to the conduction band edge.

The argument above relies on the assumption that charge transferred from BLG to the underlying CrX$_3$ layer is fixed: at low temperature, a change in $V_G$ does not change the charge accumulated in the CrX$_3$ layer. This is not what would happen if electrons in CrX$_3$ behaved as independent, non-interacting particles, i.e., if CrX$_3$ could be described as a conventional semiconductor. We attribute this behavior to the very narrow bands of CrX$_3$—electrons added to CrX$_3$ are virtually localized on the Cr orbitals—which make electrons hosted in these materials strongly correlated, because the strength of their Coulomb interaction is larger than the bandwidth. As a result, at low temperature, the electrons transferred from BLG to the surface of CrX$_3$ create an energetically

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**FIG. 4. Magnetic field dependence of the BLG band gap.** (a) Square conductance $G_\square$ as a function of gate voltage $V_G$ shifted with respect to the value of CNP $V_{CNP}^{(1)}$, measured in a BLG/CrCl$_3$ heterostructure at 0 T and with an applied magnetic field of 13 T. The left and the right panels zoom in on the onset of conductance for holes and electrons: the corresponding threshold voltages $V_{th}^h$ and $V_{th}^e$ shift upon increasing the magnetic field, resulting in a decrease in ($V_{th}^h - V_{th}^e$) and therefore in a decrease in the band gap extracted using Eq. (1). (b) Magnetic field dependence of the energy gap for BLG-on-CrCl$_3$ (green empty hexagons) and for BLG-on-CrBr$_3$ (light-blue empty circles); the size of the symbols corresponds to the experimental uncertainty associated with the error in the determination of the threshold voltages. The continuous orange lines represent the calculated band gap considering appropriate screened interlayer asymmetry potentials and including a screening potential for non-zero magnetic fields, as predicted by theory to calculate the Landau level spectrum. The experimental data are in excellent agreement with the theoretical predictions. (c) Landau levels calculated for a screened interlayer asymmetry potential of $|\Delta| = 215$ meV resulting in an experimentally observed gap of $E_g = 170$ meV at zero applied magnetic field. The dependence of the gap on the magnetic field is determined by the difference in the energies of the lowest Landau level in the conduction and valence bands.
stable correlated state (we imagine a spatially ordered distribution of electrons localized on Cr atoms that minimizes energy), which has an energy gap for adding or removing electrons. This assumption appears to be fully consistent with the behavior observed in BLG interfaces with CrCl$_3$ and CrBr$_3$, for which Eq. (1) works perfectly, but not for BLG-on-CrI$_3$, for which Eq. (1) gives a factor of 2 deviation as compared to the actual gap. The reason for this difference between the different CrX$_3$ compounds likely originates from the fact that the charge transferred from BLG to CrI$_3$ does vary as $V_G$ is varied (possibly because the width of the conduction band of CrI$_3$ is somewhat larger than that of CrCl$_3$ and CrBr$_3$), an observation that seems consistent with the pronounced temperature dependence of charge transfer from BLG to CrI$_3$ (see Figure 2e). This conclusion underscores the unconventional nature of the semiconducting properties of chromium trihalides, which calls for more detailed future investigations, and the fact that the study of transport through BLG/CrX$_3$ interfaces allows differences in the electronic properties of the different members of this family to be evidenced.

In summary, we have performed a systematic analysis of different phenomena determining the transport properties of vdW interfaces based on BLG and chromium trihalide crystals (CrCl$_3$, CrBr$_3$ and CrI$_3$). In all cases, a very large charge transfer from graphene to CrX$_3$ is found to occur, which causes the opening of a band gap in BLG. A detailed comparison shows that the values of the gap determined experimentally are in excellent agreement with the latest ab initio calculations, which include the effect of the polarizability of the $\sigma$ bands in the graphene honeycomb lattice. We furthermore show that it is possible to determine the band gap quantitatively by looking exclusively at the low-temperature gate voltage dependence of the conductivity, a finding that we exploit to determine how the band gap depends on the applied magnetic field. Besides providing indications as to the correlated nature of electrons transferred onto the very narrow conduction band of CrX$_3$, our work establishes a remarkable quantitative agreement between different electronic properties of BLG and corresponding theoretical predictions.

**SUPPORTING INFORMATION**

The Supporting Information is available free of charge on the ACS publications website at https://pubs.acs.org/doi/10.1021/acs.nanolett.2c02369.

- Transfer curves for BLG-on-CrBr$_3$ and BLG-on-CrI$_3$ devices; Magnetic field dependence of charge neutrality point.

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**NOTES**

G.T. and D.S.D. contributed equally to this work. The authors declare no competing financial interest.

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