Capacitive probes of broken symmetries in bilayer graphene

Competing valley, spin, and orbital symmetry breaking in bilayer graphene
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Recommended with a Commentary by S. A. Parameswaran, UC Irvine

The quenching of kinetic energy of two-dimensional electron gases in high magnetic fields leads to the formation of extensively degenerate Landau levels (LLs), providing a canonical setting for interaction-driven phenomena. When a LL is partially filled, the ground state is determined solely by interactions, as in the fractional quantum Hall effect. Interactions can also play a central role at integer[1] filling in resolving degeneracies that stem from internal quantum numbers such as those corresponding to spin, valley, or layer degrees of freedom. The resulting 'quantum Hall ferromagnets' exhibit a variety of interesting phenomena[2,3], such as charged skyrmions, Josephson-like effects, and symmetry-breaking phase transitions, that can be linked to competition between different possible ordered phases.

A particularly pretty example of the complex phase structure that can result due to the interplay of LL physics and electronic correlations is furnished by bilayer graphene (BLG). A single graphene layer has a two-site unit cell; in the 'Bernal stacking' common to both BLG and 3D graphite, A sites in one layer are stacked above B sites in the adjacent layer, with the B (A) sites in the first (second) layer positioned above interstices in the second (first) layer. Using a simple tight-binding model in a magnetic field to determine the LL structure[5], one finds an octet of states pinned to zero energy, the 'zeroth LL'. A fourfold degeneracy due to the combination of spin and valley is shared with graphene; an additional twofold 'orbital' degeneracy is unique to the quadratic dispersion of the bilayer. This latter degeneracy is between states that correspond to the lowest and first LLs in a conventional, parabolically dispersing 2DEG. If we define the LL filling factor ν (where $\nu = 0$ is the neutrality point), successively filling this octet of LLs corresponds to integer quantum Hall states at $\nu = -3, -2, -1, 0, 1, 2, 3, 4$. The puzzle of exactly how the combination of interactions and weak single-particle symmetry-breaking lift this degeneracy has led to much theoretical and experimental effort over the past decade, summarized in the article. (Symmetry breaking in BLG at *zero* field[4], is a separate issue that neither the authors nor I discuss.)

In the present work, the authors use a clever combination of capacitive measurements on BLG samples encapsulated in hexagonal boron nitride (a 'van der Waals (vDW) heterostructure') to tease out microscopic details and mechanisms of the broken symmetries. Specifically, the authors measure an 'antisymmetric capacitance' that probes the layer polarization, i.e. the charge imbalance between the two layers. To do so, they independently control top and bottom gate voltages; in the limit of an infinitesimally spaced, perfectly metallic bilayer, the symmetric (antisymmetric) combination of these voltages, suitably weighted by geometric capacitances, correspond to idealized electron density (n_0) and layer polarization (p_0), respectively. These are modified to the actual values (denoted n, p) due to the bilayer's intrinsic capacitance, as well as due to the formation of insulating electronic states in the bilayer. These in turn can be related to symmetric (antisymmetric) combinations $C_{S,A}$ of measured capacitances at each gate[6].

Why is layer polarization a sensitive probe of symmetry breaking? For this, we must delve a little more deeply into the structure of the ZLL. Denoting the eight degenerate single-particle states $|\xi N \sigma\rangle$ where $\xi = +, -$ denotes the valley, $N = 0, 1$ the orbital index and $\sigma =\uparrow, \downarrow$ the spin, and ignoring the spin structure for the moment, the simplest tight-binding model reveals that the $+(-)$ valley resides entirely in the top (bottom) layer. At this level of analysis, layer polarization is already a proxy for the valley index. However, the situation is richer: additional corrections show that already at the single-particle level, the $N = 1$ orbital is only partially layer-polarized, in contrast to the perfect polarization of $N = 0$, with the dominant piece still set by the valley index. Therefore, layer polarization can distinguish all four distinct (valley, orbital) combinations.

The antisymmetric capacitance (C_A) depends on the charging energy across the two atomic layers, and is hence directly related to the layer polarization. As the layers are separated on a scale $d \approx 3.35$ Å, measuring C_A is far more challenging than measuring C_S, which is sensitive only to the total density of states, and is the primary technical advance of the present paper. As the total electron density (n_0) and the applied layer imbalance (p_0) are tuned, the authors track the change in both C_S and C_A. They use the C_S measurements (their Fig. 2A) to detect the formation of insulating (QH) states, identifying a total of 16 distinct phase transitions: as insulating states are incompressible they appear as 'gaps' with low capacitance, with a transition appearing as a compressible spike in C_S. Each such transition corresponds to a change in the nature of the filled LLs, due to the combination of single-particle and correlation effects. The authors then correlate the four valley-orbital combinations $(|+0\sigma\rangle, |+1\sigma\rangle, |-1\sigma\rangle, |-0\sigma\rangle)$ with distinct layer polarizations to four discrete 'contrast' levels in the C_A data (red, orange, cyan and blue, respectively), allowing them to determine the valley-orbital state of the sample (their Fig. 2B). Spin is not captured directly by either C_S or C_A but is accounted for by the usual Zeeman contributions. Taken together, these data allow the authors to map out the structure of filled LLs at different densities and applied layer imbalances (e.g. their Fig. 2D). By incorporating interaction effects due to exchange energy contributions, and comparing with the observed sequence of LL filling, the authors can also roughly pin down the strength of Coulomb interactions $V \sim e^2/\epsilon_\parallel \ell_B^2$ parametrized by the dielectric constant $\epsilon_\parallel \sim 10-11.4$, and the charging energy $U_c = e^2 d/\pi\epsilon_\perp \ell_B^2$ of the bilayer capacitor, with $\epsilon_\perp \sim 7-8$ (ℓ_B^2 is the magnetic length).

However, the simple model of single-particle splitting augmented by exchange corrections still leaves some features of the data unexplained. First, there is a particle-hole asymmetry that the authors suggest emerge from ν-dependent contributions to ϵ due to 'intrinsic screening' by electrons in the bilayer itself, rather than the Boron Nitride layers. Second, the domains of stability of phases observed near $\nu = 0$ departs significantly from the theoretical model, which the authors relate to known subtleties of the $\nu = 0$ point, and flag for future work. Third, they observe that several of the transitions exhibit a pronounced slope in the $n_0 - p_0$ plane, indicating that the energy difference *per particle* for the relevant phases itself is density-dependent, strongly suggesting significant correlation effects. Furthermore, they note that these effects are most pronounced when the straddling phases have $N = 0, 1$, which differ strongly in exchange energies rather than layer polarization. Therefore, they argue that this is likely more than a simple charging effect, and may point to a role for partial LL fillings in any explanation.

In my view, the measurements described in the present paper represent both an end and a beginning. They add to and unify the wealth of existing experimental data on bilayer graphene, while providing an elegant validation of a (to my knowledge) novel probe of symmetry breaking. In the near future such measurements will potentially settle various questions on competing orders within the ZLL. Personally, I am more excited about the potential future applications the authors suggest for their technique. These include studying the competition between different fractional quantum Hall states in half-filled LLs, examining charged valley or orbital skyrmions[8,9]. Farther afield, I would be interested also in what their tools have to say about other vDW heterostructures or transition-metal dichalcogenides, where again determining interlayer charge order can play a central role in elucidating effects of electronic correlations and broken symmetries.

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1 As is standard, I reserve 'fractional filling' for cases where the electron density is not an integer multiple of the number of magnetic flux quanta threading the sample. In this convention, interactions may be necessary at some integer fillings: for instance $\nu = 1$ in a spinful LL is integer filling, but interactions are required to lift the spin degeneracy.

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7 A drawback with the authors' capacitive method is that the gapped states that form at exactly commensurate filling away from such transitions are inaccessible to their technique, since the system is too insulating to fully charge over the measurement timescale; hence, evidence is presented – hence the masks on their data for integer ν.

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