CYCLOTRON RESONANCE IN GRAPHENE HETEROSTRUCTURES

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Cyclotron Resonance in Graphene Heterostructures

by

B. Jordan Russell

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The Graduate School
of Washington University in
partial fulfillment of the
requirements for the degree
of Doctor of Philosophy

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We present observations of cyclotron resonance in graphene Van der Waals heterostructure devices. Such devices provide dramatic improvements in sample quality and allow for advanced electronic control, opening windows on previously inaccessible physics. The design and construction of a dedicated system for the measurement of electronic transport and infrared magnetospectroscopy in microscopic samples of atomically thin materials at cryogenic temperatures is presented. In high-mobility encapsulated monolayer graphene, electron-electron interaction effects are unambiguously observed to impact the interband cyclotron resonance as the Landau level filling factor is varied in a quantizing magnetic field. Additionally, a splitting of transitions involving the zeroth Landau level is clearly resolved and the possible origins of the implied Dirac mass are discussed. Finally, we report the first measurements of cyclotron resonance in dual-gated bilayer graphene, making a novel optical determination of the electrostatically tunable band gap in that system at the lowest energies reported to date. At half-filling of the zero energy Landau level, a possible phase transition between ordered states is observed as the electric displacement field is varied.
Chapter 1

Introduction and Overview

Since it was first isolated in 2004 graphene has enticed scientists and the public alike with its unusual and superlative mechanical, electronic, and optical properties. Composed of a single layer of carbon atoms arranged on a hexagonal lattice, graphene is found abundantly in nature, locked away inside the countless layers of three-dimensional graphite. Though it represents the absolute extreme limit of thinness achievable in condensed matter, graphene is the strongest known material [1], it possesses the highest thermal conductivity observed to date [2–4], absorbs an enormous 2.3 % of incident light [5], and conducts electric currents at room temperature with an ease that no other naturally occurring material has yet to match. Deceptively simple in composition, graphene holds a secret: the behavior of its charge carriers mimics that of two-dimensional, massless, charged fermions, introducing the Dirac equation and elements of Quantum Electrodynamics (QED) to condensed matter for the first time [6–8]. And the first atomically-thin material proved not to be the last: the discovery of graphene single-handedly gave rise to an entire subfield of condensed matter physics concerned with the discovery and investigation of atomically-thin two-dimensional materials, with dozens found in the years since 2004 and new examples appearing regularly [9,10]. These atomically-thin crystals offer a remarkable opportunity toward realizing so-called “designer quantum materials,” as they can be re-stacked in any order, with any number of layers, of any composition,\(^1\) to produce an infinite variety of composite materials. The resulting

\(^1\) Not to mention the relative twist angle between layers, which has been profoundly exploited in recent years [11,12].
Van der Waals heterostructures (VDWHs), named for the weak Van der Waals force which holds them together, are often more than the sum of their parts [13–16].

1.1 Prior Investigation of Cyclotron Resonance in Graphene and Open Questions

In the presence of a magnetic field, the relativistic nature of electrons in graphene results in a Landau level (LL) sequence with energies exhibiting an unusual square-root dependence on both the magnetic field strength \( B \) and the LL index \( N \)

\[
E_N = \text{sign}(N) v_F \sqrt{2e\hbar|N|}, \quad N = 0, \pm 1, \pm 2, \ldots
\]  

(1.1)

where \( \hbar \) is the reduced Planck constant, \( e \) is the absolute value of the electron charge, and \( v_F \approx 1 \times 10^6 \) m/s is the Fermi velocity in graphene [17]. As we will discuss in detail in Chapters 2 and 3, the spectrum (1.1) differs significantly from that found in more conventional materials. Cyclotron resonance (CR) transitions between LLs \( N \rightarrow M \) can be optically-induced in graphene according to the dipole selection rule \( N = |M| \pm 1 \), which notably allows for both intra- and interband transitions, in contrast to conventional materials where only a single intraband transition is observed.

In 2007, cyclotron resonance measurements by Horst Störmer’s group at Columbia University [18] and others [19] provided a powerful confirmation of the Dirac-like nature of charge carriers in graphene, revealing the distinct \( \sqrt{NB} \) dependence of eq. (1.1) and the presence of multiple dipole-allowed interband transitions. However, these measurements also hinted at deviations from the simple single particle picture of CR in graphene, which were later more concretely observed by Henriksen, et al. [20].

Open questions regarding the nature of many-body interaction effects on the cyclotron resonance in graphene, combined with advances in the construction of high quality graphene-
based Van der Waals heterostructures (VDWHs), can be considered as the primary motivations for the work presented in this thesis.

1.2 General Overview of this Thesis

This thesis is structured as follows. In Chapter 2 we review the basic electronic properties of monolayer and bilayer graphene, starting from considerations of electron hopping between carbon atoms on the honeycomb lattice. We calculate the electronic band structure within the tight-binding framework, and explore the quasi-relativistic behavior that emerges at low energies. We then consider the effects of a quantizing magnetic field, arriving at the Landau level spectrum of eq. (1.1). We conclude Chapter 2 by repeating these calculations for the case of bilayer graphene.

Chapter 3 presents an experimental overview of two fundamental phenomena that occur in two-dimensional electron gasses (2DEGs) in the presence of strong magnetic fields: the quantum Hall effect and cyclotron resonance. An emphasis is placed on comparing and contrasting the manifestations of these phenomena in conventional 2DEGs and graphene systems. Where possible, an attempt is made to provide historical context for the various discoveries discussed in this chapter.

Observations and analysis of cyclotron resonance in a high-mobility monolayer graphene device are presented in Chapter 4. We find that electron-electron interactions significantly impact the cyclotron resonance as the carrier density is varied in a fixed magnetic field, manifesting as a non-monotonic variation of the effective Fermi velocity. Additionally, a splitting of transitions involving the zeroth LL are observed and the possible origins of the implied Dirac mass are discussed.

Finally, in Chapter 5 we present the first measurements of cyclotron resonance in a dual-gated, encapsulated bilayer graphene device. At complete filling of the zero energy LLs we make a novel optical determination of the electrostatically tunable band gap in that
system at the lowest energies reported to date. When the zero energy Landau levels are half filled the effects of small band parameters and possible interaction effects are observed as an applied perpendicular electric field is varied. In the absence of a perpendicular electric field significant electron-hole asymmetry is observed in the cyclotron resonance as the Landau level filling is varied.

Two appendices are included covering important aspects of the measurements presented in the main chapters. In Appendix A we discuss the design and construction of the system used to make the low temperature electronic transport and magneto-infrared spectroscopic measurements presented in Chapters 4 and 5. Appendix B provides a brief discussion of standard electronic transport measurements.

1.3 Future Work and Open Questions

While the work presented in this thesis constitutes a significant advancement in the study of cyclotron resonance in graphene-based systems, in many ways it also lays the foundation for future exploration of CR in graphene, other atomically-thin crystals, and new classes of quantum materials.

In graphene devices presently available for cyclotron resonance measurements, systematic studies of intraband transitions, which have yet to be performed, promise to offer better insight into the interaction effects observed in interband CR [21,22]. Fundamental physics is almost certainly also waiting to be found in the detailed behavior of intra- and interband resonance intensities, lineshapes, and linewidths [23,24].

The physics of quantum Hall ferromagnetism (QHFM) [25,26] and the fractional quantum Hall effect [27], two remarkable manifestations of electron-electron interactions never before seen in cyclotron resonance, will likely appear in cleaner graphene samples at lower temperatures. While each has been studied extensively by way of electronic transport, CR promises to offer complementary information that may lead to greater insights into these exotic many-
body phenomena. In samples where graphene and hexagonal boron nitride are brought to close rotational alignment and the fractal quantum Hall sequence of Hofstadter’s butterfly emerges [28–31], CR could provide direct measurements of the first fractal energy spectrum realized in Nature [32,33].

Coupling graphene to photonic cavities that resonate at one or more of graphene’s cyclotron frequencies has been proposed as a route to exploring the physics of cavity quantum electrodynamics (cQED) in the regime of strong coupling [34–37]. In addition to shedding light on the physics of electron-electron interactions in the presence of strong light-matter coupling, such work could lead to novel platforms for photodetection or new qubit architectures for quantum computation.

Twisted bilayer graphene, in which two graphene monolayers are stacked on top of one another with an arbitrary relative twist angle between their crystal axes, has recently emerged as an exciting new platform for the study of strong interactions and even superconductivity in graphene; its cyclotron resonance will surely carry interesting signatures of this physics [38].

Cyclotron resonance in high quality dual-gated Bernal bilayer graphene represents an open frontier, where a number of exotic predictions for CR remain unexplored [39–45] in a system known to host strongly interacting symmetry-broken states. And studies of trilayer and multilayer graphene could provide insight into the transition of electronic behavior from two- to three-dimensional [46–49].

Additionally, a number of two-dimensional crystals and entirely new classes of quantum materials have emerged since the discovery of graphene, very few of which have been explored by way of cyclotron resonance. In particular, a number of semimetals have been discovered in recent years which support entirely new types of fermions. Infrared optical and magneto-optical probes will surely play a role in uncovering the mysteries of type-I and type-II Weyl semimetals [50–54], 2D and 3D Dirac semimetals [55–57], and nodal semimetals [58–60].
Chapter 2

Dirac Electrons in Graphene

Monolayers and Bilayers

The remarkable electronic, optical, and mechanical properties of graphene are determined entirely by the deceptively simple arrangement of carbon atoms in a two-dimensional hexagonal lattice. At low energies, the resulting quasiparticle behavior that emerges is found to mimic that of relativistic, massless, charged, chiral fermions at a deep level, bringing the Dirac equation and elements of Quantum Electrodynamics (QED) to condensed matter. The relativistic nature of charge carriers in graphene leads to a number of unique phenomena, including a cyclotron resonance and quantum Hall effect qualitatively distinct from those observed in traditional two-dimensional electron gases (2DEGs). Furthermore, when two graphene monolayers are coupled, the resulting bilayer system hosts massive, chiral quasiparticles that exhibit behavior which interpolates between that of graphene and traditional 2DEGs and has no direct analogue in the Standard Model of particle physics. The ability to break inversion symmetry in graphene bilayers through the application of an electric field means it can be continuously driven from a semimetallic state to a semiconducting state with an electrostatically tunable band gap, giving rise to electronic behavior unseen in any other material system and making it enormously promising for applications.

Given the unrivaled properties found in monolayer and bilayer graphene, it is surprising that much of this behavior can be captured by a simple tight binding model. We begin this
chapter by calculating the band structure of monolayer graphene starting from the electronic orbitals of carbon. Special consideration is given to the low-energy behavior of electrons and holes in graphene where relativistic effects emerge. We then examine the effects of strong magnetic fields on the band structure of graphene, which give rise to a spectrum of quantized Landau levels qualitatively distinct from those found in conventional 2DEGs. This analysis is then repeated for the case of bilayer graphene, with an emphasis on the effects of an interlayer potential energy difference.

2.1 Carbon and the Honeycomb Lattice

Carbon demonstrates an unrivaled capacity to hybridize the orbitals of its valence electrons; this ability allows it to form an enormous array of compounds, earning carbon a role of out-sized importance in the study of chemical bonding and making it the only element known to form structures of every dimension between zero and three. From its ground state configuration, $1s^22s^22p^2$, carbon can promote one $2s$ electron into the empty $2p$ state, and from there form hybrid states in which one ($sp^1$), two ($sp^2$), or all three ($sp^3$) of its $2p$ orbitals mix with the remaining $2s$ orbital. The resulting hybrid states share characteristics of the constituent orbitals.

The five allotropes of carbon span zero (fullerene), one (nanotube), two (graphene), and three dimensions (graphite and diamond). While diamond is a result of $sp^3$ bonding between carbon atoms, the four remaining allotropes are all products of $sp^2$ hybridization [61]. Graphene is, in a sense, the progenitor of all $sp^2$ carbon allotropes: by stacking it layer by layer we can form graphite, by rolling it up we can produce nanotubes, while introducing defects and bending it from all sides yields the spherical fullerene. In the $sp^2$ state the three hybrid orbitals lay in a plane, which we arbitrarily take to be $x - y$, with the remaining unhybridized $p_z$ orbital oriented perpendicular to this plane.

The head-on overlap of these $sp^2$ orbitals gives rise to sigma bonding, the strongest form of
covalent bond, and naturally produces trigonal planar structures with bonds angles of 120°.

In graphene these sigma bonds not only define the characteristic honeycomb shape of the lattice, but also imbue it with enormous mechanical strength and stiffness. The stiffness and spatial symmetries of the lattice produce a unique phonon spectrum, leading to a reduction in electron-phonon scattering that, in part, explains the enormous electronic mobility in graphene. However, the sigma bonding and anti-bonding states contribute valence and conduction bands which, respectively, lay far below and above the Fermi energy and are, for all intents and purposes, electronically inert. As a result, the electronic behavior of graphene may be accurately described by considering only the single remaining $p_z$ orbital on each carbon atom. The weak, side-on overlap of the $p_z$ wavefunctions leads to the formation of π bonds much like those found in benzene, which are characterized by massive electronic delocalization, a phenomena known to chemists as aromaticity. The bands that result from the overlap of these $p_z$ orbitals cross the Fermi energy and dominate electronic transport and optical processes in graphene. In the following section we calculate the dispersion of these bands and show that at low energies they give rise to exotic electronic behavior.
Fig. 2.2: (a) The crystal lattice of monolayer graphene and (b) its first Brillouin zone.

2.1.1 The Hexagonal Lattice

The honeycomb lattice of monolayer graphene, illustrated in fig.2.2a, is in fact a triangular Bravais lattice with a two atom basis, which we arbitrarily label A and B. Equivalently, one can think of the honeycomb lattice as being composed of two inter-penetrating triangular sublattices related to one another by inversion symmetry. In either case, we find two atoms within the unit cell, that are related by inversion symmetry. This will be of fundamental importance to the band structure of graphene, discussed below. The unit cell is outlined and shaded in fig.2.2a.

The nearest neighbor carbon-carbon bond length $\delta$ is 1.42 Å, resulting in a very tight lattice. For comparison, in graphite the layer separation is considerably larger at 3.35 Å, due to the weakness of Van der Waals bonding relative to the strong in-plane sigma bonds. The lattice vectors, $\mathbf{a}_1$ and $\mathbf{a}_2$, also illustrated in figure 2.2, connect any point in the unit cell to the equivalent point in a neighboring unit cell. Their length, distinct from the carbon-carbon separation, is given by $a = |\mathbf{a}_1| = |\mathbf{a}_2| = \sqrt{3} \delta = 2.46$ Å. Explicitly, $\mathbf{a}_1$ and $\mathbf{a}_2$ can be written as

$$\mathbf{a}_1 = \left( \frac{a}{2}, \frac{\sqrt{3}a}{2} \right), \quad \mathbf{a}_2 = \left( \frac{a}{2}, -\frac{\sqrt{3}a}{2} \right),$$  \hspace{1cm} (2.1)$$

The honeycomb lattice is self-dual, in the sense that its corresponding reciprocal space lattice
is also hexagonal, as shown in fig. 2.2b. The primitive lattice vectors in momentum space are
\[ \mathbf{b}_1 = \left( \frac{2\pi}{a}, \frac{2\pi}{\sqrt{3}a} \right), \quad \mathbf{b}_2 = \left( \frac{2\pi}{a}, -\frac{2\pi}{\sqrt{3}a} \right). \] (2.2)

The six points of high symmetry at the corners of the Brillouin zone are of fundamental importance to the low energy electronic properties of graphene. Of these six points only two, which we label \( \mathbf{K}_\pm \), are inequivalent. This can be seen from the fact that any \( \mathbf{K}_+ (\mathbf{K}_-) \) point can be reached from another \( \mathbf{K}_+ (\mathbf{K}_-) \) point by a translation of \( \mathbf{b}_1 \) or \( \mathbf{b}_2 \). Explicitly, their position in momentum space is given by
\[ \mathbf{K}_+ = \left( +\frac{4\pi}{3a}, 0 \right), \quad \mathbf{K}_- = \left( -\frac{4\pi}{3a}, 0 \right). \] (2.3)

For reasons that will soon be clear, the points \( \mathbf{K}_\pm \) are commonly called valleys. We adopt the general notation \( \mathbf{K}_\lambda \), and call \( \lambda = \pm 1 \) the valley index. The existence of the \( \mathbf{K}_\pm \) valleys in momentum space is intimately related to the A-B sublattice symmetry of the real space graphene lattice. The threefold degeneracy of each valley within the first Brillouin zone is a consequence of the trigonal symmetry of graphene’s hexagonal lattice.

### 2.2 The Band Structure of Monolayer Graphene

The band structure of graphene was first theoretically explored by Philip Russell Wallace in 1947 [62]. The anisotropic electronic and thermal properties of graphite, the clearly laminar structure of graphite crystals, and the detailed crystal structure data of Bernal all suggested graphite was composed of many weakly bound layers, while the atoms within a layer were quite tightly bound to one another. As a starting point for uncovering the electronic, magnetic, thermal, and optical properties of graphite, Wallace first considered a single graphitic layer, adding the weak interlayer interactions to arrive at the band structure of graphite. He found that, in isolation, these single layers demonstrated unusual electronic
behavior, with an electronic energy dispersion that was linear in momentum at low energies and almost perfectly symmetric electron and hole bands that vanished at a single crossing point in momentum space. His approach was later refined by J. C. Slonczewski, P. R. Weiss, and J. W. McClure in 1957, resulting in the well-known SWM model of graphite which parameterizes the electronic hopping processes allowed purely by symmetries of the graphite crystal structure [63,64]. Because of its general nature the SWM model is still used in its originally published form; the value of many of its parameters are well known, though some still elude precise determination [65–68]. In this section we discuss the tight-binding method in general, before applying it to the case of MLG. Later in the chapter we will repeat this analysis for bilayer graphene.

2.2.1 The Tight-Binding Method

The tight binding method is a simple approach to calculating the single-particle band structure of a crystalline material. The organizing principle of the tight binding method is the assumption that the wavefunctions of crystal electrons may be expressed as a superposition of isolated atomic wavefunctions, which is reasonable provided the overlap of orbitals on neighboring atoms is weak. This condition of weak overlap is equivalent to requiring that the electrons remain tightly bound to the atoms that make up the crystal, hence the name of the method itself. The tight binding method provides an extremely accurate band structure for graphene, showing strong agreement with experimental results and more advanced theoretical methods. Here we present a brief review of the tight binding method in general, applying it to monolayer and bilayer graphene in the following sections. We largely adopt the notation of Saito, Dresselhaus, and Dresselhaus [61] and McCann and Koshino [69].

In the presence of a periodic potential, Bloch’s theorem guarantees the existence of an eigenstate basis of the form

\[ \phi(k, r) = e^{ikr}u(r), \]  

(2.4)
where $e^{i\mathbf{k} \cdot \mathbf{r}}$ are plane waves and the function $u(\mathbf{r})$ inherits the periodicity of the external potential. In a crystal with $m = 1, 2, ..., M$ atomic orbitals $\phi_m$ per unit cell, and $n = 1, 2, ..., N$ unit cells, this basis takes the explicit form

$$
\Phi_m(\mathbf{k}, \mathbf{r}) = \frac{1}{\sqrt{N}} \sum_{n=1}^{N} e^{i\mathbf{k} \cdot \mathbf{R}_{m,n}} \phi_m(\mathbf{r} - \mathbf{R}_{m,n}), \quad (2.5)
$$

where the vector $\mathbf{R}_{m,n}$ gives the position of the $m^{th}$ orbital in the $n^{th}$ unit cell of the crystal. The wavefunction $\Psi_j(\mathbf{k}, \mathbf{r})$ of an electron in the $j = 1, 2, ..., J$ band may be expanded in this basis of Bloch waves

$$
\Psi_j(\mathbf{k}, \mathbf{r}) = \sum_{m=1}^{M} \psi_{j,m}(\mathbf{k}) \Phi_m(\mathbf{k}, \mathbf{r}). \quad (2.6)
$$

For $M$ orbitals per unit cell, there will be $J = M$ bands in the band structure. The energy of the $j^{th}$ band can be found by solving the Schrödinger equation

$$
H\psi_j = E_j S\psi_j, \quad (2.7)
$$

where $\psi_j^T = (\psi_{j,1}, \psi_{j,2}, ..., \psi_{j,M})$, $H$ is the transfer or “hopping” matrix, and $S$ is the overlap matrix, which accounts for any non-orthogonality of the employed basis. The transfer and overlap matrices are each $M \times M$ with elements

$$
H_{m,m'} = \langle \Phi_m | \mathcal{H} | \Phi_{m'} \rangle, \quad S_{m,m'} = \langle \Phi_m | \Phi_{m'} \rangle. \quad (2.8)
$$

The elements $H_{m,m'}$ describe interatomic transfer or “hopping” processes; the hamiltonian $\mathcal{H} = \mathcal{H}_{\text{atom}} + \Delta U$ is the sum of the atomic hamiltonian plus the small corrections due to orbital overlap. The elements $S_{m,m'}$ quantify the spatial overlap of orbitals $\Phi_m$ and $\Phi_{m'}$. In practice, determining these matrix elements for a particular system is the primary challenge of the tight-binding method.
2.2.2 The Tight-Binding Model of Monolayer Graphene

The small number of atoms in the graphene unit cell, the need to include only one $2p_z$ orbital per atom, and the reasonably weak overlap of these orbitals all conspire to make application of the tight-binding method to monolayer graphene straightforward. We label these orbitals $m = A$ and $m = B$. If we neglect next nearest neighbor interactions, the diagonal elements of the hopping matrix give the on-site energies

$$H_{AA} \approx \frac{1}{N} \sum_{n=1}^{N} \langle \phi_A(r - R_{A,n})|H|\phi_A(r - R_{A,n}) \rangle,$$

which can be written as a simple constant, $H_{AA} = \epsilon_A$, and similarly for B site orbitals, $H_{BB} = \epsilon_B$. The off-diagonal hopping element

$$H_{AB} \approx \frac{1}{N} \sum_{n=1}^{N} \sum_{l=1}^{3} e^{ik \cdot \delta_l} \langle \phi_A(r - R_{A,n})|H|\phi_B(r - R_{B,n} - \delta_l^A) \rangle,$$

describes electronic transfer from A sites to B sites. The nearest neighbor vectors $\delta_l^A$ point to the location of the three B site atoms that surround a given A site and are given by

$$\delta_1^A = \left(0, \frac{a}{\sqrt{3}}\right), \quad \delta_2^A = \left(\frac{a}{2}, -\frac{a}{2\sqrt{3}}\right), \quad \delta_3^A = \left(-\frac{a}{2}, -\frac{a}{2\sqrt{3}}\right).$$

It is common practice to separate $H_{AB}$ into a constant

$$\gamma_0 = -\langle \phi_A(r - R_{A,n})|H|\phi_B(r - R_{B,n} - \delta_l^A) \rangle,$$

that sets the energy scale of the band\(^1\), and a function

$$f(k) = \sum_{l=1}^{3} e^{ik \cdot \delta_l^A} = e^{ik_ya/\sqrt{3}} + 2e^{-ik_ya/2\sqrt{3}} \cos(k_xa/2),$$

\(^1\) Commonly referred to as the bandwidth.
which is a sum of phase factors and characterizes the directional dependence of nearest neighbor hopping on the in-plane wavevector. We adopt the frequently used value $\gamma_0 = 3.0 \text{eV}$ [61, 70–72]. The two triangular sublattices of graphene are related by spatial inversion, so the nearest neighbor vectors from a given B site to the three surrounding A sites are $\delta_l^B = -\delta_l^A$. As a result, the hopping element describing transfer from B to A sites is related by complex conjugation to its time reversed process, $H_{BA} = H_{AB}^\ast$.

Combining these results and assuming an on-site energy difference $|\epsilon_A - \epsilon_B| = \Delta_{AB}$ between atoms A and B, we find

$$H_{MLG} = \begin{pmatrix} \Delta_{AB}/2 & -\gamma_0 f(k) \\ -\gamma_0 f^\ast(k) & \Delta_{AB}/2 \end{pmatrix}$$

(2.14)

The elements of the overlap matrix quantify the spatial overlap of the orbitals. Naturally, the result is simple for diagonal elements

$$S_{AA} = \langle \phi_A(r - R_{A,n})| \phi_A(r - R_{A,n}) \rangle = 1,$$

(2.15)

since the overlap of an orbital with itself is unity. By the same argument $S_{BB} = 1$. As with $H_{AB}$, the off-diagonal element

$$S_{AB} = \langle \phi_A(r - R_{A,n})| \phi_B(r - R_{B,n} - \delta_l^A) \rangle,$$

(2.16)

can be separated into a unitless measure of the overlap, $s_0$, and a function which captures the variation of the orbital overlap in space, which is again given by $f(k)$. The off-diagonal elements are once again related by complex conjugation and explicitly we find

$$S_{AB} = S_{BA}^\ast = s_0 f(k).$$

(2.17)
The full overlap matrix is
\[
S_{MLG} = \begin{pmatrix}
1 & s_0 f(k) \\
 s_0 f^*(k) & 1
\end{pmatrix},
\]
(2.18)

As with $\gamma_0$, the value of $s_0$ is found by fitting to experimental data, typically optical spectra. We adopt the commonly employed value $s_0 = 0.129$ [61, 69, 72].

Solving the Schrödinger equation (2.7) for our $2 \times 2$ Hamiltonian (2.14) yields eigenvalues for two bands
\[
E_{\pm} = \frac{\Delta_{\alpha\beta} \pm \gamma_0 |f(k)|}{1 \pm s_0 |f(k)|}.
\]
(2.19)

We plot the bands\(^2\) determined by equation (2.19) in fig. 2.3 for $\Delta_{\alpha\beta} = 0$, $s_0 = 0.129$, and $\gamma_0 = 3.0$ eV. In a departure from the behavior observed in virtually all other materials, we find the conduction and valence bands in graphene meet and vanish at six points in momentum space, at the corners of the hexagonal Brillouin zone, with a highly linear dispersion in the vicinity of this band crossing.

The two $p_z$ orbitals within the unit cell can each accommodate a total of four electrons after accounting for spin degeneracy. In charge neutral graphene these orbitals are precisely half-filled, a fact that we arrive at by simply counting the electrons contributed by each carbon atom. It follows that the Fermi energy in pristine graphene lies precisely at the six unusual points in momentum space where the conduction and valence bands meet, which is defined as the natural zero of energy. The valence band is completely full of electrons, while the conduction band hosts an equivalent number of holes.

It is interesting to consider the topology of the band structure illustrated in fig. 2.3. If the Fermi energy lies immediately above or below the point of band crossing, one finds the Fermi “surface” to be a one-dimensional ring, but as $E_F$ passes through the point of contact this Fermi surface collapses to a zero-dimensional point. As discussed below, when

\(^2\) For intrinsic graphene the on-site energies of orbitals A and B are exactly equal and given by the energy of the $2p_z$ orbital, $\epsilon_A = \epsilon_B = \epsilon_{2p_z}$, and it is customary to let $\epsilon_{2p_z} = 0$. A non-zero difference in on-site energies has the effect of opening a band gap equal to $\Delta_{\alpha\beta}$ in graphene’s otherwise gapless band structure. There are important situations in which a non-zero $\Delta_{\alpha\beta}$ can be realized in practical graphene devices; we return to this point at a later time.
Dirac Electrons in Graphene Monolayers and Bilayers

Fig. 2.3: The band structure of monolayer graphene as determined by a nearest neighbor tight-binding model using parameter values $\Delta_{AB} = 0$, $s_0 = 0.129$, and $\gamma_0 = 3.0 \text{ eV}$. The band energies are given by eq. (2.19). At low energies the band structure is almost perfectly linear, forming Dirac cones which meet at a single point in momentum space.

electrons and holes execute orbits\(^3\) that enclose these singularities in the band structure, their wavefunctions develop a non-trivial geometric phase with significant consequences [73, 74].

2.2.3 Low Energy Effective Theory of Graphene Charge Carriers

From figure 2.3 it is clear that at low energies the electronic properties of graphene are dominated by the points $K_{\pm} = \pm(4\pi/3a, 0)$ in momentum space. To gain a clearer understanding of the electronic behavior in the vicinity of these points we can shift the origin to one of the two valleys

$$\kappa = k - K_\lambda \quad (2.20)$$

\(^3\) The most obvious case being the cyclotron orbits that charge carriers execute in the presence of a magnetic field.
and expand the sum of phase factors \( f(\mathbf{k}) \) in powers of \( \mathbf{k} \). Keeping only terms linear in wavevector we find

\[
f(\mathbf{k}) \approx -\frac{\sqrt{3}a}{2}(\lambda \kappa_x - i \kappa_y),
\]

which is valid for \( \kappa a << 1 \), near \( \mathbf{K}_\pm \).

If we identify \( v_F = \sqrt{3}a\gamma_0/2\hbar \) as the Fermi velocity\(^4\) of charge carriers in graphene, which defines the slope of the band structure at low energies, and recall \( \mathbf{p} = \hbar \mathbf{\kappa} \), we can rewrite the low-energy Hamiltonian in the vicinity of the valley \( \mathbf{K}_\lambda \) as

\[
H_\lambda = \lambda v_F \begin{pmatrix}
0 & (p_x - ip_y) \\
(p_x + ip_y) & 0
\end{pmatrix}.
\]

The corresponding solutions

\[
|\Psi_\lambda\rangle = \begin{pmatrix}
\psi_{A,\lambda} \\
\psi_{B,\lambda}
\end{pmatrix},
\]

give the amplitude of the electronic wavefunctions on sublattice A and B in valley \( \mathbf{K}_\lambda \).

Considering the valley \( \mathbf{K}_+ \) explicitly, we may rewrite Hamiltonian (2.22) in the form

\[
\mathcal{H}_+ = v_F \mathbf{\sigma} \cdot \mathbf{p},
\]

where \( \mathbf{\sigma} = (\sigma_x, \sigma_y) \) is the two-dimensional Pauli vector operating on the basis of sublattices A and B [7]. The solutions to eq. (2.24) are two-component spinors

\[
\Psi_+(\mathbf{\kappa}) = \frac{1}{\sqrt{2}} \begin{pmatrix}
e^{-i\theta_{\kappa}/2} \\
s e^{i\theta_{\kappa}/2}
\end{pmatrix},
\]

where the \( s = +1(-1) \) indexes electron (hole) states and the angle \( \theta_{\kappa} = \text{Arctan}(\kappa_x/\kappa_y) \) is set by the direction of the in-plane momentum. The spin-like degree of freedom implicit in the wavefunction (2.25) is commonly referred to as the sublattice pseudospin, which arises

---

\(^4\) Using \( \gamma_0 = 3.0 \) eV we find \( v_F = 9.7 \times 10^5 \) m/s \( \approx c/300 \). We will round this up to \( 1 \times 10^6 \) m/s.
from the bipartite nature of the hexagonal lattice and, notably, under a $2\pi$ rotation of $\kappa$ the wavefunction accumulates a phase shift of $\pi$ \[75\]. In a clear departure from the parabolic dispersion found at low energies in virtually all other materials, the eigenvalues of eq. (2.24)

$$E = s v_F \hbar \kappa,$$

(2.26)

mimic the linear-in-momentum behavior of relativistic particles.

For completeness, in the $K$ valley we find\(^5\)

$$\mathcal{H}_- = v_F \sigma^* \cdot \mathbf{p},$$

(2.27)

which has solutions

$$\Psi_-(\kappa) = \frac{1}{\sqrt{2}} \begin{pmatrix} e^{i\theta \kappa/2} \\ s e^{-i\theta \kappa/2} \end{pmatrix}.$$  

(2.28)

Thus we find the wavefunctions in the two valleys are related by time reversal symmetry (TRS).\(^6\)

Finally, one may define a chirality operator $\hat{\eta} = (\kappa \cdot \sigma) / |\kappa|$ which has eigenvalues $\pm 1$ and quantifies the fact that quasiparticle motion on the hexagonal lattice is locked to the underlying pseudospin degree of freedom \[78\]. The chirality $\eta$, valley quantum number $\lambda$, and band index $s$ are then related to one another by

$$s = \lambda \eta,$$

(2.29)

which reveals the fact that the handedness of the pseudospin is opposite in the two valleys \[78\].

---

\(^5\) It is common to invert the role of sublattice in the $K$ valley, which allows one to write a single Hamiltonian for the two valleys (see eq. 2.22). In this case the wavefunctions may be written as four-component bispinors, completing the analogy to Dirac’s theory \[76\].

\(^6\) The combination of this TRS with the inversion symmetry of the graphene lattice yields charge conjugation (electron-hole) symmetry \[77\].
To conclude, as first shown by Semenoff in 1984 [79] and expounded upon by others [80, 81], the behavior of low-energy charge carriers in graphene is governed by a massless two-dimensional Dirac equation, with the analogy running much deeper than the linear dispersion of eq. (2.26).

2.2.4 Landau Levels in Monolayer Graphene

The effects of a magnetic field on the unusual Dirac-like charge carriers in graphene can be accounted for by rewriting Hamiltonian (2.24) in terms of the kinetic momentum \( \pi = p + eA \)

\[
\mathcal{H} = v_F \sigma \cdot \pi, \quad \text{(2.30)}
\]

where the magnetic field and vector potential are related by \( B = \nabla \times A \). Acting on the wavefunction \( \Psi = (\psi_A, \psi_B)^T \) with Hamiltonian (2.30) yields

\[
v_F \left[ (p_x + eA_x) - i(p_y + eA_y) \right] \psi_B = E\psi_A \quad \text{(2.31a)}
\]
\[
v_F \left[ (p_x + eA_x) + i(p_y + eA_y) \right] \psi_A = E\psi_B. \quad \text{(2.31b)}
\]

Substituting eq. (2.31a) into eq. (2.31b), choosing the Landau gauge \( A = Bx\hat{y} \) to generate a magnetic field \( B = B\hat{z} \) applied perpendicular to the graphene plane, and expanding we find

\[
v_F^2 \left( -\hbar^2 \frac{\partial^2}{\partial x^2} - \hbar^2 \frac{\partial^2}{\partial y^2} - 2i\hbar eBx \frac{\partial}{\partial y} + e^2B^2x^2 - eB\hbar \right) \psi_B = E^2\psi_B. \quad \text{(2.32)}
\]

Looking forward to chapter 3, where we derive the Landau level (LL) spectrum for the case of a parabolic dispersion, we recognize eq. (2.32) to be equivalent to the quantum harmonic oscillator given by eq. (3.5) with three noteworthy differences: here the energy on the right-hand side is squared, the factor of \( 1/2m^* \) has been replaced by \( v_F^2 \), and there is an additional

---

7 This so-called minimal coupling procedure is discussed at greater length in Ch. 3.
8 The final term \( ieB(xp_x - px) = -e\hbar B \) follows from \([x, p_x] = i\hbar\).
term, $-eB \hbar$, not present in the parabolic case. Thus we take the spectrum determined by
the lefthand side of eq. (2.32) to be that of eq. (3.12), making the substitution $1/m^* \rightarrow 2v_F^2$
in $\omega_c$ and keeping the additional term. The result

$$E^2 = 2\hbar eBv_F^2 \left( N + \frac{1}{2} \right) - \hbar eBv_F^2,$$

(2.33)
simplifies to

$$E^2 = \hbar eBv_F^2 (2N + 1 - 1),$$

(2.34)
which leads us to the famous Landau level energy spectrum for massless Dirac charge carriers

$$E = \pm \sqrt{2\hbar eBv_F^2 N} \quad N = 0, 1, 2, \ldots$$

(2.36)
which is independent of the valley index $\lambda$.

The spectrum (2.36) is distinguished from the LL spectrum of a parabolic system in four
ways: (i) It is electron-hole symmetric, as evidenced by the positive and negative solutions. (ii) In contrast to the usual ladder of uniformly spaced LLs, the $\sqrt{N}$ dependence
of eq. (2.36) imparts each LL with a unique energetic spacing from its nearest neighbors. (iii) The cyclotron energy in graphene has a $\sqrt{B}$ dependence on the applied magnetic field, while conventional two-dimensional electron gases (2DEGs) are linearly dependent on $B$ (see eq. (3.7) and eq. (3.12)). (iv) Finally, the presence of a Berry’s phase of $\pi$ due to the unusual
topology of the Dirac cones is directly responsible for the additional term in eq. (2.32), which
cancels the factor of $+1/2$ in the harmonic oscillator spectrum (3.12). The result is a true
zero energy zeroth Landau level with deep connections to the axial anomaly in quantum
electrodynamics [79, 82–85]. While all other LLs in graphene have either electron-like or
hole-like character, the zeroth LL is uniquely equal parts both.

---

9 We will more frequently write the spectrum (2.36) in the form

$$E_N = \text{sign}(N) v_F \sqrt{2e\hbar B|N|} \quad N = 0, \pm 1, \pm 2, \ldots$$

(2.35)
to remove the ambiguity when referencing specific electron or hole LLs.
Fig. 2.4: Illustrations of the Landau level (a) density of states, (b) ladder diagram, and (c) dispersion as a function of magnetic field. Optically-induced inter- and intraband LL transitions are illustrated in (b). All interband transitions are allowed at \( \nu = 0 \) while a maximum of two intraband transitions are allowed simultaneously depending on the position of the Fermi energy. Notably, transitions involving the zeroth LL have both intra- and interband characteristics. For more information regarding the allowed CR transitions in graphene, see section 2.2.5 below.

In the Landau gauge, the wavefunctions of charge carriers in the \( N^{th} \) LL of the \( \mathbf{K}_+ \) valley are [86]

\[
|N, \mathbf{K}_+\rangle = \frac{C_N}{\sqrt{2}} \begin{pmatrix}
 i^{N}|\phi_{|N|}(x - \ell_B^2 k_y) \\
 \text{sign}(N)i^{N-1}\Phi_{|N|-1}(x + \ell_B^2 k_y) \end{pmatrix} e^{i k_y y},
\]

(2.37)

where the normalization factor \( C_N \) is given by

\[
C_N = \begin{cases} 
\sqrt{2} & N = 0 \\
1 & |N| > 0
\end{cases}
\]

(2.38)
and $\phi_{|N|}$ is defined as

$$
\phi_{|N|}(x + \ell_B^2 k_y) = \frac{1}{\sqrt{2^{|N|} |N| ! \pi \ell_B L_y}} \exp \left[ \frac{-(x + \ell_B^2 k_y)^2}{2 \ell_B^2} \right] H_{|N|} \left( \frac{x + \ell_B^2 k_y}{\ell_B} \right). \tag{2.39}
$$

In the above expressions $\ell_B = \sqrt{\hbar/eB}$ is the magnetic length, $L_y$ is the extent of the graphene sheet in the $\hat{y}$-direction, $k_y$ is the $y$-component of the in-plane wavevector, and $H_N(x)$ is the $N^{th}$ Hermite polynomial.\(^{10}\)

Notably, for $N = 0$ we find the second component of wavefunction (2.37) is zero, corresponding an amplitude entirely on the A sublattice. Recalling that the role of sublattice is inverted in the two valleys, we can deduce that in a similar fashion the amplitude of $|0, K_-\rangle$ lies entirely on sublattice B. Thus we find that for $N = 0$ electrons and holes in opposite valleys are polarized on opposite sublattices, further distinguishing this LL from all the others \([76,87]\).\(^{11}\)

### 2.2.5 Optical Transitions Between Landau Levels in Graphene

Semiclassically, the Hamiltonian governing the interaction of Landau quantized charge carriers with an incident electromagnetic field takes the form

$$
\mathcal{H}_{el-ph} = i \hbar \frac{e}{m_0} \sum_{i,f} M_{i,f} \cdot \mathbf{A}' \hat{a}_f^\dagger \hat{a}_i, \tag{2.40}
$$

where $\mathbf{A}'$ is the vector potential of the incoming radiation.\(^{12}\) The operators $\hat{a}_f^\dagger$ and $\hat{a}_i$, which create and annihilate states in the indexed Landau level, are discussed in section 3.3.3. The optical matrix elements $M_{i,f} = \langle \Psi_f | \nabla | \Psi_i \rangle$ are computed using the wavefunctions (2.37) and determine the absorption intensity of a given transition, as well as the dipole selection rules governing which transitions are allowed or forbidden. As shown in Refs. \([23,88–90]\), the

---

\(^{10}\) See the discussion surrounding eq. (3.6) and eq. (3.13) for further explanation of these quantities.

\(^{11}\) This means that for $N = 0$, and only for $N = 0$, we may identify valley with sublattice pseudospin.

\(^{12}\) In a fully quantum treatment of this interaction, the vector potential $\mathbf{A}'$ would be written in terms of photon creation and annihilation operators.
optical matrix elements for Landau quantized graphene are

\[
    M_{if} = \frac{i\alpha_N \alpha_{N_f} m_0 v_F}{2\sqrt{2}\hbar} \delta_{\lambda_i,\lambda_f} \delta_{\sigma_i,\sigma_f} \delta_{k_i,k_f} \left[ s_{N_i} e^{-\delta_{N_f,|N_i|-1}} + s_{N_f} e^{+\delta_{N_f,|N_i|+1}} \right],
\]  

where \( \alpha_{N=0} = \sqrt{2} \) and \( \alpha_{N\neq0} = 1 \), \( m_0 \) is the free electron mass, \( v_F \) is the Fermi velocity in graphene, and \( s_N = \text{sign}(N) \). The Jones vectors \( e^\pm = (\hat{e}_x \mp \hat{e}_y)/\sqrt{2} \) describe left-handed (+) and right-handed (−) circularly polarized light. The initial and final states \( |i\rangle = |N_i,\lambda_i,k_i,\sigma_i\rangle \) and \( |f\rangle = |N_f,\lambda_f,k_f,\sigma_f\rangle \), are characterized by four quantum numbers corresponding to the Landau level index \( N \), the valley \( \lambda \), the intrinsic spin of the electron \( \sigma \), and the in-plane wavevector \( k \) which is related to the cyclotron guiding center.\(^{13} \) The first three Kronecker delta functions in eq.\((2.41)\) impose the constraint that optically-induced, dipole-allowed transitions between Landau level states conserve valley, spin, and the cyclotron guiding center, as required by momentum conservation. The last two delta functions in eq.\((2.41)\) give rise to the selection rules \( |N_f| = |N_i| - 1 \) for right-handed and \( |N_f| = |N_i| + 1 \) for left-handed polarizations, which may be written as

\[
    \Delta N = \pm 1, 
\]  

in contrast to the CR selection rule \( \Delta N = +1 \) found in other systems.

The unusual selection rule \((2.42)\), which follows from the fact that both the conduction and valence bands in graphene form from the same overlapping \( \pi \) orbitals, gives rise to three types of cyclotron resonance in graphene. Interband transitions \((N \geq 1)\)

\[
    -N \rightarrow N + 1
\]

\[
    -N - 1 \rightarrow N,
\]

which have energies

\[
    \Delta E = v_F \sqrt{2e\hbar B} \left( \sqrt{N+1} + \sqrt{N} \right),
\]  

\(^{13}\) See eq.\((3.9)\) and the surrounding discussion of the cyclotron guiding center.
Dirac Electrons in Graphene Monolayers and Bilayers

occur in degenerate pairs due to the electron-hole symmetry found in graphene. The intra-band transitions \((N \geq 1)\)

\[
N \rightarrow N + 1 \\
-N - 1 \rightarrow -N,
\]

have relatively lower energies given by

\[
\Delta E = v_F \sqrt{2e\hbar B} \left( \sqrt{N+1} - \sqrt{N} \right).
\]

Finally, resonances involving the zero energy zeroth Landau level

\[
N_{-1} \rightarrow N_0 \\
N_0 \rightarrow N_1,
\]

occur at energy

\[
\Delta E = v_F \sqrt{2e\hbar B},
\]

and may be equivalently classified as the highest energy intraband or lowest energy interband resonance.

Several intra- and interband transitions are depicted schematically in fig. 2.4b. It must be stressed that CR transitions in graphene are subject to Pauli blocking,\(^{14}\) and are therefore allowed or forbidden depending on the location of the Fermi energy: Optical transitions are only allowed out of filled states and into empty states. If a transition’s initial state is completely unoccupied, or its final state is fully occupied, the corresponding resonance will not occur. Near charge neutrality, corresponding to a Landau level filling factor \(\nu \approx 0\), all interband transitions, including the mixed-character \(0 \Rightarrow \pm 1\) resonances, may be observed simultaneously. In contrast, a maximum of two intraband transitions may be observed simultaneously at low temperatures.

\(^{14}\) For further details on Pauli blocking of optical transitions in graphene, the reader is referred to Ref. [91] and references therein.
Pauli blocking also provides a clean method for spectroscopic background removal in CR measurements of density-tunable graphene devices: Spectra are first acquired at a Landau level filling factor of interest. The Fermi energy is then moved, typically to the highest or lowest carrier density available when interband transitions are being studied,\textsuperscript{15} and spectra are again collected. Division of the spectra collected at these different filling factors allows for the removal of all sample-independent features. This backgrounding technique is discussed in more detail in Ch. 4.

\section*{2.3 The Density of States and Electric Field Effect in Graphene}

\subsection*{2.3.1 The Density of States in Graphene}

The density of states (DOS) plays a fundamental role in determining the optical and dielectric properties of a material and the scattering rates of its charge carriers. In two-dimensions the DOS per unit area can be calculated from \cite{92}

$$\text{DOS}(E) = \frac{g}{(2\pi)^2} 2\pi k \frac{dk}{dE},$$  \hspace{1cm} (2.49)

where \(g\) counts the degeneracy of internal degrees of freedom such as spin and valley. By integrating the DOS up to the Fermi energy we can obtain the two-dimensional charge carrier density

$$n = \int_{0}^{E_F} \text{DOS}(E) \, dE,$$  \hspace{1cm} (2.50)

where \(n = \mathcal{N}/S\) is the total number of carriers in the sample, \(\mathcal{N}\), divided by the area of the sample, \(S\).

\textsuperscript{15} At very high or low carrier densities all but the very lowest energy intraband transitions are blocked, which allows for clean backgrounding of interband transitions up to a certain energy. If intraband transitions are being studied, it is often preferable to background at charge neutrality.
In a typical 2DEG with an isotropic parabolic dispersion, \( E(k) = E_0 + \hbar^2 k^2 / 2m^* \), where \( E_0 \) gives the energy at the band edge and \( m^* \) is the carrier effective mass, we find \( (dE/dk)^{-1} = m^*/\hbar k \) and therefore

\[
\text{DOS}(E) = \begin{cases} 
0 & E < E_0 \\
g m^* / 2\pi \hbar^2 & E > E_0.
\end{cases}
\]  

(2.51)

Integrating, we find

\[
n = \begin{cases} 
0 & E < E_0 \\
g m^* (E_F - E_0) / 2\pi \hbar^2 & E > E_0.
\end{cases}
\]  

(2.52)

Thus the carrier density is zero below the band edge and increases linearly with \( E_F \) above the band edge. The Fermi surface in such a system is a circle with a radius equal to the Fermi wavevector

\[
k_F = \sqrt{4\pi n / g}.
\]  

(2.53)

In graphene one finds that the linear dispersion relation, \( E = v_F \hbar k \), leads to a qualitatively distinct DOS [93, 94]. Substituting \( (dE/dk)^{-1} = 1/v_F \hbar \) and \( k = E/v_F \hbar \) into eq. (2.50), and explicitly including the spin and valley degeneracy \( g = g_s g_v = 2 \times 2 = 4 \), we find a density of states

\[
\text{DOS}_{\text{MLG}}(E) = \frac{2E}{\pi v_F^2 \hbar^2},
\]  

(2.54)

which varies linearly from zero with energy, in contrast to the step function behavior (2.51) found in 2D parabolic systems. The resulting carrier density

\[
n_{\text{MLG}} = \frac{E_F^2}{\pi \hbar^2 v_F^2} = \frac{k_F^2}{\pi},
\]  

(2.55)

is quadratic in the Fermi energy, but the dependence of the Fermi wavevector on the carrier density

\[
k_F = \sqrt{\pi n},
\]  

(2.56)

is the same as a typical 2DEG (2.56) with a degeneracy of four.
In bilayer graphene, which we discuss in detail in the following section, the density of states is

$$\text{DOS}_{BLG}(E) \approx \frac{2E + \gamma_1}{\pi v_F^2 \hbar^2}, \quad (2.57)$$

when the band gap is zero,\(^{16}\) which interpolates between the behavior of monolayer graphene (2.54) and a traditional parabolically dispersing 2DEG (2.51), where \(E_0 = 0\) and the effective mass is set by the interlayer coupling, \(m^* = \gamma_1/2v_F^2\).

The gapless, zero-overlap band structure of MLG earns it the unusual classification of zero bandgap semiconductor. In intrinsic bilayer graphene, a finite density of states at zero energy makes it a semimetal, like graphite [96-98]. Uniquely, bilayer graphene becomes a semiconductor with an electrostatically tunable band gap in the presence of interlayer asymmetry [99].

### 2.3.2 The Electric Field Effect in Graphene

One of the first breakthroughs in graphene research came when it was discovered that its charge carrier density could be efficiently tuned between electron and hole gases by applying a voltage between the sample and its silicon substrate [96]. Graphene’s atomic thinness renders it incapable of screening the electric field that results from the application of this gate voltage, \(V_g\), allowing a surface charge density, \(n_s\), to be induced. For graphene separated from a metallic substrate by an insulating material of thickness \(t\) and relative permittivity \(\epsilon_r\), the induced carrier density is

$$n = \frac{\epsilon_r \epsilon_0}{te} V_g. \quad (2.58)$$

The quantity \(\alpha = \epsilon_r \epsilon_0 / te\), commonly referred to as the gate efficiency, is simply the capacitance per unit area per unit charge of the graphene-gate parallel plate capacitor. For

\(^{16}\) It is worth emphasizing that this expression is valid only in the presence of interlayer symmetry. As discussed below, interlayer asymmetry in bilayer graphene dramatically reshapes the band structure by inducing a so-called sombrero shape in addition to opening a bandgap. This sombrero geometry produces a divergent DOS [95]. Interlayer asymmetry can be induced by an electric field applied perpendicular to the bilayer plane.
an insulating layer of silicon dioxide ($\epsilon_r = 3.8$) with a thickness of 300 nm one finds $\alpha \approx 7 \times 10^{10} \text{ cm}^{-2} \text{ V}^{-1}$. In Combining eq.(2.58) with eq.(2.55), we see that the Fermi energy in graphene can be varied electrostatically.

![Diagram](image)

**Fig. 2.5:** (a) An illustration of a typical encapsulated monolayer graphene device. Electrical contacts to the device are omitted and layer thicknesses are not to scale. (b) The measured resistivity and conductivity of a high-quality monolayer graphene device at $B = 0$ T. This sample (bngbn062216) is discussed in detail in Chapter 5. The FWHM of the Dirac peak is $\delta n = 1.12 \times 10^{11} \text{ cm}^{-2}$ and $V_{DP} = -10.7 \text{ V}$. A field effect mobility of 190 000 (290 000) cm$^2$ V$^{-1}$ s$^{-1}$ was calculated for electrons (holes).

For a typical maximum gate voltage of $\sim 100 \text{ V}$, the maximum induced carrier density is $\sim 7 \times 10^{12} \text{ cm}^{-2}$. The minimum carrier density achievable in a real graphene device is determined by the level of disorder in the sample. Disorder produces so-called electron-hole puddles, spatial inhomogeneities of the carrier density amounting to spatial variation of the Fermi energy at which charge neutrality occurs. The Dirac peak observed in zero field resistance measurements of graphene (see fig. 2.5) is essentially the average of many Dirac peaks, and thus the width of this peak, $\delta n$, is an indicator of sample homogeneity and, therefore, sample quality. In graphene on silicon dioxide, which is known to have a surface that hosts dangling bonds and is rough on an atomic scale, one typically finds $\delta n \gtrsim 5 \times 10^{11} \text{ cm}^{-2}$. When graphene is encapsulated in hexagonal boron nitride (hBN), a wide band gap insulating van der Waals material that is atomically-flat and free of dangling bonds, one typically finds $\delta n \lesssim 1 \times 10^{11} \text{ cm}^{-2}$ [100]. More recently it has been demonstrated that
hBN encapsulated graphene on a graphite back gate offers another substantial improvement in device quality, with $\delta n \sim 1 \times 10^{10} \text{ cm}^{-2}$ [101].

In addition to broadening the Dirac peak, the presence of charged disorder can induce an overall shift of the carrier density that must be overcome to reach charge neutrality. As a result, eq. (2.58) is usually given in the more practical form

$$n = \alpha (V_g - V_{DP})$$

(2.59)

where $V_{DP}$ is the voltage at which minimum conductivity is observed and $n_0 = \alpha V_{DP}$ is the induced carrier density in the absence of an applied gate voltage.

### 2.4 The Band Structure of Bilayer Graphene

Bilayer graphene consists of two monolayers stacked on top of one another. These two layers may be arranged in three distinct ways: twisted bilayers, AA stacking, and AB stacking. Twisted bilayer graphene, in which one monolayer is stacked on top of another with an arbitrary relative twist angle between their common crystallographic axes, has generated a great deal of interest recently following the 2018 discovery that this system displays flat bands, Mott insulator physics, and, at certain “magic angles”, tunable superconductivity [11,12]. The AA stacking arrangement of bilayer graphene, in which atoms in the two layers lay directly on top of each other, is naturally occurring, but extremely rare due to a lack of thermodynamic stability. Theory predicts exotic electronic behavior in AA bilayers due to the presence of extremely flat bands, but little is known of it experimentally [102].

The AB, or “Bernal”, stacking arrangement, which we consider in the following and refer to simply as bilayer graphene (BLG), is by far the most commonly encountered in nature.\(^{17}\)

As shown in figure 2.6, in an AB bilayer every other carbon atom within a layer has a

\(^{17}\) In 1924 John Desmond Bernal demonstrated conclusively that the atomic layers in graphite predominantly take on an AB stacking arrangement.
neighboring atom in the opposing layer, while the remaining atoms are positioned over or under the center of the hexagons in the neighboring layer. We will refer to those pairs with neighbors in the opposing layer as “dimer” sites, and those that lay above or below hexagon centers as “non-dimer” sites. The separation between layers is 3.35 Å [103], which is considerable compared to the intralayer nearest neighbor separation of 1.42 Å, and it is reasonable to expect that the band structure should closely resemble that of two decoupled graphene monolayers. However, as we will see, the interlayer coupling between dimer sites breaks the sublattice symmetry within each layer, giving rise to massive chiral quasiparticles which have no direct analogue in the Standard Model of particle physics.

Fig. 2.6: The lattice of AB stacked bilayer graphene from (a) top and (b) side views. The site indices, lattice vectors, unit cell, and hopping parameters are labeled. Adapted from Ref. [69].

The tight-binding calculation for BLG proceeds in largely the same way as the monolayer calculation, with the exception that there are now four atoms within the unit cell. We label the two atoms in the bottom layer \( m = A1, B1 \) and the two in the top layer \( m = A2, B2 \), defined such that \( B1-A2 \) form dimer sites. Each atom contributes one \( p_z \) orbital, and as such we anticipate four bands. However, matters are complicated by the presence of four hopping terms and three on-site energy parameters, compared to the single hopping term and single asymmetry parameter in the monolayer case. Many of these additional terms are relatively small and can often be neglected, however there are situations in which each can play an important role.

\(^{18}\) This value is virtually unaltered from the graphite layer separation.
2.4.1 Bilayer Hopping and Asymmetry Parameters

The bilayer lattice vectors $a_1$ and $a_2$, reciprocal lattice vectors $b_1$ and $b_2$, and Brillouin zone are all identical to those of monolayer graphene. The intralayer hopping parameter $\gamma_0$ is also unchanged from the monolayer case. However, as illustrated in fig. 2.6b, there are now three additional hopping parameters between atoms in opposing layers that must be accounted for [71]. Interlayer hopping between non-dimer sites is parameterized by $\gamma_3$, while interlayer hopping between dimer and non-dimer sites is determined by $\gamma_4$. But it is hopping between dimer sites, given by $\gamma_1$, which has the most profound impact on the low energy behavior of BLG.

To summarize, the hopping parameters in BLG are [69, 71]

$$\gamma_0 = -\langle \phi_{A1} | \mathcal{H} | \phi_{B1} \rangle = -\langle \phi_{A2} | \mathcal{H} | \phi_{B2} \rangle \approx 3000 \text{ meV} \quad (2.60a)$$

$$\gamma_1 = \langle \phi_{A2} | \mathcal{H} | \phi_{B1} \rangle \approx 400 \text{ meV} \quad (2.60b)$$

$$\gamma_3 = -\langle \phi_{A1} | \mathcal{H} | \phi_{B2} \rangle \approx 300 \text{ meV} \quad (2.60c)$$

$$\gamma_4 = \langle \phi_{A1} | \mathcal{H} | \phi_{A2} \rangle = \langle \phi_{B1} | \mathcal{H} | \phi_{B2} \rangle \approx 150 \text{ meV}. \quad (2.60d)$$

The parameters $\gamma_2$ and $\gamma_5$ describing next nearest layer hopping in the SWM model are, of course, absent in bilayer graphene.

In addition to hopping processes we must also account for the difference in on-site energies between the four atoms within the unit cell.$^{19}$ In the most frequently employed parameterization, $\delta_{dim}$ accounts for differences between a dimer and non-dimer site ($A1$ vs $A2$ or $B1$ vs $B2$)$^{20}$, $\delta_{AB}$ gives the difference between intralayer sites ($A1$ vs $B1$ or $A2$ vs $B2$), and $\Delta$ describes a possible interlayer energy difference ($A1$ and $B1$ vs $A2$ and $B2$). Of the three, only $\delta_{dim}$ is an intrinsic asymmetry present even in intrinsic bilayers. Experimentally, $\delta_{AB}$ and $\Delta$ are enormously important, with the former invariably generated by the presence of

$^{19}$ It is important to note that while three numbers are enough to completely define the difference in four on-site energies, there are multiple acceptable parameterizations.

$^{20}$ Note that in bilayers $\delta_{dim} = \delta_{dim}^*/2$ where $\delta_{dim}^*$ is the standard graphite SWM parameter.
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a boron nitride substrate and the latter produced by application of a perpendicular electric field.

Summarizing, one finds the following on-site energy differences in intrinsic bilayer graphene [71,104]:

\[
\delta_{dim} = \left( (\epsilon_{B1} + \epsilon_{A2}) - (\epsilon_{A1} + \epsilon_{B2}) \right)/2 \approx 18 \text{ meV} \quad (2.61a)
\]
\[
\delta_{AB} = \left( (\epsilon_{A1} + \epsilon_{A2}) - (\epsilon_{B1} + \epsilon_{B2}) \right)/2 = 0 \quad (2.61b)
\]
\[
\Delta = \left( (\epsilon_{A1} + \epsilon_{B1}) - (\epsilon_{A2} + \epsilon_{B2}) \right)/2 = 0. \quad (2.61c)
\]

Parameters \(\gamma_0\) through \(\gamma_3\) have been well determined by experiments for some time, while \(\gamma_4, \gamma_5\), and \(\delta_{dim}\) are known with less precision [71].

### 2.4.2 Four-Band Tight-Binding Model of Bilayer Graphene

Combining the various hopping elements we find the full Hamiltonian for BLG

\[
H_{\text{BLG}} = \begin{pmatrix}
\epsilon_{A1} & -\gamma_0 f(k) & \gamma_4 f(k) & -\gamma_3 f^*(k) \\
-\gamma_0 f^*(k) & \epsilon_{B1} & \gamma_1 & \gamma_4 f(k) \\
\gamma_4 f^*(k) & \gamma_1 & \epsilon_{A2} & -\gamma_0 f(k) \\
-\gamma_3 f(k) & \gamma_4 f^*(k) & -\gamma_0 f^*(k) & \epsilon_{B2}
\end{pmatrix}. \quad (2.62)
\]

Defining the wavefunctions and overlap matrix elements in the same way as the monolayer case and solving for the solutions of eq.(2.7) we find four bands, as anticipated. These bands closely resemble those of monolayer graphene, with linear dispersion \(E \approx v_F p\) for momenta \(p \gamma_1/2v_F\), but now we find that the sublattice symmetry breaking effect of the dimer site interaction has imparted a curvature to all four bands near the band minima.

This curvature produces a massive dispersion \(E \approx p^2/2m^*\) for momenta \(p \lesssim \gamma_1/2v_F\), where the effective mass\(^{21}\) is \(m^* = \gamma_1/2v_F^2\). Two low-energy bands, associated with hopping on

\(^{21}\) We note that this expression for band mass is only valid in the absence of gap-opening asymmetries.
sublattices $A_1$ and $B_2$, still meet at a single point\textsuperscript{22} in momentum space, which coincides with the position of the Fermi energy in pristine bilayers.\textsuperscript{23} The other two bands, associated with hopping on dimer sites $B_1$ and $A_2$, are pushed away from the Fermi energy by an amount $\gamma_1 \approx 400$ meV.

In figure 2.7 we plot numerical solutions for all four bands for several values of interlayer asymmetry $\Delta$. Notably, the opening of a bandgap due to an interlayer asymmetry does not simply shift the low-energy bands away from zero, but gives rise to a so-called ‘sombrero’ shape \cite{95,109} which generates Van Hove singularities in the zero field density of states \cite{95,110} and exotic LL crossings in the presence of a magnetic field \cite{111}; the high-energy dimer bands are largely unaffected by $\Delta$. The ability to tune this gap from zero to $\sim 300$ meV has been demonstrated using several methods \cite{99,112,113}, with the most conclusive evidence\textsuperscript{24} coming from infrared optical determinations \cite{71,115–119}.

Finally, we note that an analytical expression for all four bands in the absence of a magnetic field has been derived by several authors \cite{69,120} by taking $\gamma_4 = \delta_{dim} = \delta_{AB} = 0$. Adapting to our own notation, we find the sometimes useful result

$$E(p) = s \sqrt{\frac{\gamma_1^2}{2} + \frac{\Delta^2}{4} + p^2 \left( v_F^2 + \frac{v_3^2}{2} \right) + (-1)^\alpha \sqrt{\Gamma}},$$

(2.63)

where

$$\Gamma = \frac{1}{4} \left( \gamma_4^2 - p^2 v_3^2 \right)^2 + v_F^2 p^2 \left( \gamma_1^2 + \Delta^2 + p^2 v_3^2 \right) + 2\lambda \gamma_4 p^3 v_3 \cos(3\phi).$$

(2.64)

and $s = +1 (-1)$ indexes the conduction (valence) band, while $\alpha = 1 (2)$ addresses the non-dimer low-energy (dimer high-energy) bands. Here we generally define the effective velocity $v_i = \sqrt{3a\gamma_i/2\hbar}$, though we will continue to refer to $v_0$ as $v_F$.

\textsuperscript{22} In BLG, this singularity in the band structure gives rise to a Berry phase of $2\pi$, in contrast to the previously discussed MLG Berry phase of $\pi$ \cite{74,105}.

\textsuperscript{23} Some theoretical work predicts that interaction effects could open a gap in the zero field bandstructure of BLG \cite{106}. Some experimental evidence for this has been found in suspended bilayers \cite{107,108}.

\textsuperscript{24} Transport and optical determinations of the bandgap in BLG are in strong disagreement, with optical bandgaps consistently appearing up to two order of magnitude larger than those measured in transport \cite{113}. Various theories have been put forth invoking mid-gap impurity states in the presence of disorder \cite{114} or exotic topological edge states.
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Fig. 2.7: The low-energy band structure of bilayer graphene for four values of the interlayer potential difference $\Delta$. Notably the high-energy dimer bands are largely unaffected by the interlayer asymmetry $\Delta$.

2.4.3 Low-Energy Effective Theory of Bilayer Graphene

As with monolayer graphene, a simplified effective theory emerges if we expand the function $f(k)$ in Hamiltonian (2.62) in terms of the momentum measured from the Dirac points. Upon doing so we find

$$H_{BLG} = \begin{pmatrix} 
\epsilon_{A1} & v_F p^\dagger & -v_4 p^\dagger & v_3 p \\
v_F p & \epsilon_{B1} & \gamma_1 & -v_4 p^\dagger \\
-v_4 p & \gamma_1 & \epsilon_{A2} & v_F p^\dagger \\
v_3 p^\dagger & -v_4 p & v_F p & \epsilon_{B2} 
\end{pmatrix}, \quad (2.65)$$

where $p = \hbar k$ as before.

To acquire an effective description at energies very near the Dirac point there exists a prescription for decoupling the dimer and non-dimer sites $[69, 78, 120, 121]$, in which case the four-band effective Hamiltonian (2.65) can be further reduced to a two-band effective Hamil-
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\[ H_{BLG} = H_0 + h_{\gamma_3} + h_{\gamma_4} + h_{dim} + h_{\Delta} + h_{AB} \]  \hspace{1cm} (2.66)

where all the matrices in eq. (2.66) are 2 × 2 and operate in the basis of non-dimer sites A1 and B2

\[ |\Psi\rangle = \begin{pmatrix} \psi_{A1} \\ \psi_{B2} \end{pmatrix}. \]  \hspace{1cm} (2.67)

The behavior at low energies is dominated by

\[ H_0 = -\frac{1}{2m^*} \begin{pmatrix} 0 & p^\dagger p^\dagger \\ p^\dagger p & 0 \end{pmatrix}, \]  \hspace{1cm} (2.68)

which describes massive, chiral quasiparticles with a Berry phase of 2π

\[ \Psi_+ (\kappa) = \frac{1}{\sqrt{2}} \begin{pmatrix} e^{-i\kappa} \\ e^{i\kappa} \end{pmatrix}, \]  \hspace{1cm} (2.69)

and has an eigenvalue spectrum

\[ \pm \frac{p^2}{2m^*}. \]  \hspace{1cm} (2.70)

We can treat all other terms in (2.66) as perturbations of \( H_0 \). Each is associated with a particular band parameter and each has a unique effect on the band structure. Referring the interested reader to the detailed analysis in Refs. [41, 69, 71, 104], we note only that \( h_{\gamma_3} \) is responsible for the so-called trigonal warping of the band structure, while \( h_{\gamma_4} \) and \( h_{dim} \) are responsible for electron-hole symmetry breaking effects. These three terms, plus \( H_0 \), are intrinsic to BLG and must respect the inversion symmetry of the lattice. However, the extrinsic terms

\[ h_{\Delta} = -\frac{\Delta}{2} \begin{pmatrix} 1 & 0 \\ 0 & -1 \end{pmatrix} - \frac{2v_F^2}{\gamma_1} \begin{pmatrix} p^\dagger p & 0 \\ 0 & -p^\dagger p \end{pmatrix}, \]  \hspace{1cm} (2.71)
and

\[ h_{AB} = \frac{\delta_{AB}}{2} \begin{pmatrix} 1 & 0 \\ 0 & -1 \end{pmatrix}, \tag{2.72} \]

respectively break inter- and intralayer symmetry and are the only terms in eq. (2.66) capable of opening a bandgap in bilayer graphene [99,122].

### 2.4.4 Landau Levels in Bilayer Graphene

As in the case of monolayer graphene, we may solve for the spectrum of bilayer graphene in the presence of a magnetic field by making the substitution \( p \rightarrow \pi = p + eA \) in the Hamiltonian of interest. Doing so for \( H_0 \), we find eigenvalues

\[ E_N = \pm \hbar \omega_c \sqrt{N(N-1)} \quad N \geq 0 \tag{2.74} \]

where \( \omega_c = eB/m^* \) is the cyclotron frequency and the effective mass in BLG is a very light \( m^* \sim 0.035 m_0 \), where \( m_0 \) is the free electron mass [123]. The spectrum (2.74) is notably independent of the valley quantum number. The corresponding eigenvectors in valley \( K_+ \) are

\[ |N, K_+\rangle = \begin{pmatrix} \phi_N \\ 0 \end{pmatrix} \quad N = 0, 1 \tag{2.75} \]

\[ |N, K_+\rangle = \frac{1}{\sqrt{2}} \begin{pmatrix} \phi_N \\ s_N \phi_{|N|-2} \end{pmatrix} \quad N = \pm 2, \pm 3, ... \tag{2.76} \]

25 Similar to the monolayer case, we will usually write this as

\[ E_N = \text{sign}(N) \hbar \omega_c \sqrt{|N|||N|-1|} \quad N = 0, 1, \pm 2, \pm 3, ... \tag{2.73} \]

to make referencing specific electron or hole LLs more straightforward.

26 In Ref. [123] Shubnikov-de Haas oscillations in single-gated BLG devices indicated effective masses for electrons (holes) to be \( m_e^* \approx 0.033 m_0 \) \( m_h^* \approx 0.043 m_0 \) at a carrier density of \( n = 2 \times 10^{12} \text{ cm}^{-2} \). The ratio \( m_h^*/m_e^* = 1.2 - 1.3 \), found over a range of densities, supports the expectation of significant electron-hole asymmetry in BLG, with holes some 20% heavier than electrons. However, it should be noted that measurements of cyclotron resonance in BLG, including the work presented in Ch. 5 and that in Ref. [124], indicate a somewhat lighter effective masses \( m^* \sim 0.023 m_0 \).
Fig. 2.8: Illustrations of the bilayer Landau level (a) density of states and ladder diagram at (b) zero and (c) finite $\Delta$.

but in valley $K_-$ we find

$$ |N, K_-\rangle = \begin{pmatrix} 0 \\ \phi_N \end{pmatrix} \quad N = 0, 1 $$

(2.77)

$$ |N, K_+\rangle = \frac{1}{\sqrt{2}} \begin{pmatrix} \phi_{|N|-2} \\ s_N \phi_N \end{pmatrix} \quad N = \pm 2, \pm 3, \ldots $$

(2.78)

where $s_N = \text{sign}(N)$ and the $\phi_N$ are the same as those defined for MLG (2.39).
Studying eqs. (2.74) - (2.78) we can identify several remarkable features in the LL spectrum of bilayer graphene. First, in contrast to the single zero energy LL in MLG, the spectrum (2.74) of BLG features two fourfold degenerate Landau levels at zero energy, which is a consequence of the $2\pi$ Berry phase in bilayer graphene and is responsible for the unusual quantum Hall sequence observed in BLG \[74\]. The resulting massive degeneracy due to spin, valley, and this additional orbital degree of freedom gives rise to strong Coulomb and exchange interactions between an enormous number of electrons and holes \[120\], making the quantum Hall octet in BLG a fertile environment to explore the effects of symmetry breaking in the quantum Hall regime. Theory \[41,106,111,125–133\] and experiment \[107,108,134–141\] both suggest the presence of competing ordered states.

Further, examining eq. (2.75) through (2.78) also reveals that the wavefunctions for $N = 0$ and 1 are layer polarized, with the zero energy LLs in valley $K_+$ completely localized on sublattice $A1$ and those in valley $K_-$ completely localized on sublattice $B2$. This means that within the quantum Hall octet we can identify the valley and layer degrees of freedom with one another, which is made all the more remarkable by the fact that we can break the symmetry of the two layers, and therefore the two valleys, by the application of an electric field perpendicular to the bilayer \[109,120,142\]. For higher LLs, we find that the wavefunctions (2.76) and (2.78) carry weight on both sublattices and the relationship between valley and layer is not as simple, however, as we discuss momentarily, the orbital structure still allows for the degeneracy of the two valleys to be broken, even for $N \geq 2$.

The spectrum (2.74), derived from the effective Hamiltonian (2.68), neglects the effects of parameters $\gamma_3$, $\gamma_4$, $\Delta$, $\delta_{\text{dim}}$, and $\delta_{\text{AB}}$, which can profoundly impact the band structure. In Ref. \[111\], Zhang, et al. derive analytical expressions for the LL energies neglecting only the hopping parameter $\gamma_3$ and the intralayer potential difference $\delta_{\text{AB}}$. Defining the quantities

$$\Theta = \left( \frac{\hbar eB}{\gamma_1 m^*} \right)^2 = \frac{2\hbar eB v_F^2}{\gamma_1^2},$$  \hspace{1cm} (2.79)
which is the cyclotron energy normalized by the interlayer coupling strength $\gamma_1$, and

$$\tilde{\delta} = \delta_{\text{dim}} + \frac{2 \gamma_1 \gamma_4}{\gamma_0}, \quad (2.80)$$

which sets the energy scale of electron-hole symmetry breaking effects,\textsuperscript{28} they show that the LL energies may be expressed as

$$E_{N=0}^{\lambda=\pm} = \lambda \frac{\Delta}{2} \quad (2.81a)$$

$$E_{N=1}^{\lambda=\pm} = \Theta \tilde{\delta} + \lambda(1 + 2\Theta) \frac{\Delta}{2} \quad (2.81b)$$

$$E_{|N|\geq 2}^{\lambda=\pm} = \left(|N| + \frac{1}{2}\right) \Theta \tilde{\delta} - \lambda \Theta \frac{\Delta}{2} + s_N \sqrt{\left[(2|N| + 1)\Theta \frac{\Delta}{2} - \frac{\lambda}{2} \Theta \tilde{\delta} - \frac{\Delta}{2}\right]^2 + |N|(|N| + 1)\Theta^2 \gamma_1^2} \quad (2.81c)$$

where electron and hole LLs are indexed by $s_N = \text{sign}(N)$. As shown in fig. 2.9, the LL spectrum given by eq. (5.5) displays a wealth of interesting behaviors not captured by the commonly encountered approximation given in eq. (2.74). First, the degeneracy of $N = 0$ and 1 is now broken by an amount $\Theta \tilde{\delta}$, even in the absence of an interlayer potential difference. At $B = 10$ T we find $\Theta \tilde{\delta} \approx 4.4$ meV, which is still reasonably small relative to the cyclotron energy $\hbar \omega_c \approx 39$ meV at this field. Furthermore, we find that eq. (2.81) captures the valley splitting behavior in the presence of an interlayer potential difference, as anticipated from our examination of the wavefunctions (2.75) - (2.78), with the valley splitting of $N = 0$ and 1 displaying a simple linear dependence on $\Delta$ and a more complicated dependence for higher LLs. Interestingly, LL crossings can also be seen at certain values of $\Delta$, which are discussed in detail in Ref. [111]. Cyclotron resonance between valley-split LLs in bilayer graphene is the subject of Chapter 5.

\textsuperscript{28} Using $\gamma_0 = 3000$ meV, $\gamma_1 = 400$ meV, $\gamma_4 = 150$ meV, and $\delta_{\text{dim}} = 18$ meV, we find $\tilde{\delta} = 58$ meV.
Fig. 2.9: The dependence of bilayer Landau level energies on (a) interlayer potential difference and (b) magnetic field.
Chapter 3

Cyclotron Resonance and The Quantum Hall Effect

Cyclotron resonance and the quantum Hall effect are part of a family of remarkable phenomena in condensed matter with a common origin in the quantization of energy that charge carriers undergo in the presence of a magnetic field.\(^1\) Both emerge under the extreme conditions of very low temperatures and intense magnetic fields, where Landau levels dramatically reshape the band structure of the host system. In this chapter we give a brief accounting of these phenomena, with an emphasis on their experimental manifestations.

We begin with a discussion of charge carriers confined to two dimensions, a scenario realized in only a few physical systems that has, nevertheless, played an enormously important role in condensed matter physics for the last forty years. We then consider the effect of a magnetic field on the energy-momentum relationship of electrons in two dimensions, arriving at Landau’s titular discovery: under such conditions the energies of charge carriers take on quantized values. We then explore the effect of large magnetic fields on electronic transport in two dimensions, arriving at one of the most exotic phenomena known to occur in condensed matter: the quantum Hall effect. We conclude with a discussion of cyclotron resonance, the resonant absorption of light by charged particles in a magnetic field.

\(^1\) The resulting discrete spectrum was first calculated by Lev Landau in 1930 [144].
3.1 The Two-Dimensional Electron Gas and Its Landau Level Spectrum

The Hamiltonian of a free electron may be written as

\[ H = \frac{\hbar^2 k^2}{2m^*}, \]  

where \( \hbar \) is the reduced Planck constant, \( k \) is the three-dimensional electron wavevector \( k = (k_x, k_y, k_z) \), and \( m \) is the electron mass.\(^2\) The solutions to (3.1) are well known to be plane waves of wavevector \( k \). If electronic motion is restricted in one direction, let it be \( \hat{z} \), by some infinite potential well of width \( L_z \), one finds the values of \( k_z \) take on a discrete spectrum \( k_{z,n} = n\pi/L_z \), where \( n \) is a positive integer indexing the allowed subbands, while a continuum of allowed wavevectors persists in the \( x-y \) plane. If the width of this potential is sufficiently narrow, and the temperature and density of electrons in the well is sufficiently low, one finds that only the lowest value of \( k_{z,n} \) may be occupied and electronic motion in the \( \hat{z} \)-direction is effectively frozen. Under these conditions, the wavevector is effectively reduced to \( k = (k_x, k_y) \) and the electron gas is, for all intents and purposes, two-dimensional. Later in this chapter we discuss some physical realizations of two-dimensional electron gases (2DEGs) in condensed matter systems.

3.1.1 Quantization in a Magnetic Field

In the presence of a magnetic field \( \mathbf{B} \) the gauge invariant kinetic momentum is

\[ \pi = p + eA, \]  

\(^2\) For electrons propagating in a crystal lattice \( m \to m^* \), where \( m^* \) is the effective mass. In general the effective mass is anisotropic and may be greater than or less than the bare electron mass, though the latter is more common.

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and the Hamiltonian 3.1 becomes

\[ H = \frac{1}{2m} (\mathbf{p} + e\mathbf{A})^2, \tag{3.3} \]

where we have made use of the relationship \( \mathbf{p} = \hbar \mathbf{k} \) between the wavevector and momentum operators and \( \mathbf{A} \) is the vector potential associated with the magnetic field by

\[ \mathbf{B} = \nabla \times \mathbf{A}. \tag{3.4} \]

We consider the case of a magnetic field oriented perpendicular to the plane of the 2DEG, \( \mathbf{B} = B\mathbf{\hat{z}} \), and choose to work in the Landau gauge \( \mathbf{A} = Bx\mathbf{\hat{y}} \). For simplicity we neglect the Zeeman term related to the electron spin.\(^4\)

By choosing the Landau gauge we find that eq. (3.3) can be explicitly written as

\[ H = \frac{1}{2m} \left( -\hbar^2 \frac{\partial^2}{\partial x^2} - \hbar^2 \frac{\partial^2}{\partial y^2} - 2i\hbar eB_x \frac{\partial}{\partial y} + e^2 B^2 x^2 \right), \tag{3.5} \]

where we have made use of the fact that \( \mathbf{p} = -i\hbar \nabla \). Noting the absence of terms that break translational invariance in the \( \mathbf{\hat{y}} \)-direction, we may assume solutions to eq. (3.5) are wavefunctions of the form

\[ \psi(x, y) = \phi(x) e^{i\mathbf{k} \cdot \mathbf{y}}, \tag{3.6} \]

where the exact form of \( \phi(x) \) is undetermined and \( e^{i\mathbf{k} \cdot \mathbf{y}} \) are plane waves of wavevector \( k_y \).

We now define three important quantities. The cyclotron frequency of a non-relativistic

\(^3\) For a free electron, \( \mathbf{p} \rightarrow \mathbf{\pi} = \mathbf{p} + e\mathbf{A} \), is known as minimal coupling. The same procedure is widely used to incorporate the effects of a uniform magnetic field on electrons in a crystal, which is valid as long as the vector potential varies slowly on the scale of the crystal lattice constant [28]. In the latter case this is known as Peierls substitution.

\(^4\) This is generally a good approximation given the smallness of the Bohr magneton, \( \mu_B = e\hbar / 2m_e \approx 0.058 \) meV/T. At \( B = 10 \) T, this amounts to a Zeeman splitting of \( \sim 1.16 \) meV, easily an order of magnitude less than the cyclotron energy scale in conventional 2DEGs.
particle with effective mass $m^*$ and charge $e$ is

$$\omega_c = \frac{e B}{m^*}. \quad (3.7)$$

The cyclotron energy, $\hbar\omega_c$, sets the fundamental energy scale of charge carriers in the presence of a magnetic field. The magnetic length

$$\ell_B = \sqrt{\frac{\hbar}{e B}}, \quad (3.8)$$

defines the characteristic length scale of the wavefunctions of charged particles undergoing cyclotron motion in their ground state;\(^5\) intuitively, it is the quantum analog to the cyclotron radius, $r_c = mv/eB$. Finally, we define the cyclotron guiding center

$$X_k = -\frac{\hbar}{e B} k = -\ell_B^2 k. \quad (3.9)$$

Acting on wavefunction (3.6) with Hamiltonian (3.5), making the substitution\(^6\) $p_y = -i\hbar \frac{\partial}{\partial y} \rightarrow \hbar k_y$, and rewriting the results in terms of eqs. (3.7) - (3.9), we find

$$\mathcal{H}\psi(x,y) = \left[ -\frac{\hbar^2}{2m^*} \frac{\partial^2}{\partial x^2} + \frac{1}{2} m^* \omega_c^2 (x - X_k)^2 \right] \phi(x) = \mathcal{E}_{k,N} \phi(x). \quad (3.10)$$

Equation (3.10) is immediately identifiable as the Schrödinger equation for a quantum har-

\(^{5}\) In a magnetic field of $B = 10$ T, one finds $\ell_B = 8.1$ nm.

\(^{6}\) This substitution is allowed since $[p_y, \mathcal{H}] = 0$. 

monic oscillator centered at \( x = X_k \), which has the well-known discrete\(^7\) spectrum

\[
E_{k,N} = \hbar \omega_c \left( N + \frac{1}{2} \right) \quad N = 0, 1, 2, \ldots,
\]  

(3.12)

where the integer \( N \) is known as the Landau level index; all states with quantum number \( N \) compose the \( N^{th} \) Landau level (LL). Ignoring degeneracies due to spin and other internal degrees of freedom, eq. (3.10) describes a single harmonic oscillator located on each guiding center \( x = X_k \). The normalized solutions for \( \phi(x) \) are

\[
\phi(x) = \left( \frac{1}{\sqrt{2^N N! \sqrt{\pi}}} \right) \left( \frac{m^* \omega_c}{\hbar} \right)^{1/4} \exp \left[ -\frac{(x - X_k)^2}{2\ell_B^2} \right] H_N \left( \frac{x - X_k}{\ell_B} \right),
\]  

(3.13)

where \( H_N(x) \) is the \( N^{th} \) order Hermite polynomial.

If each oscillator occupies an area \( 2\pi \ell_B^2 \), then the number of allowed states per LL sub-band\(^8\) in a 2DEG of area \( S \) is

\[
N_\Phi = \frac{S}{2\pi \ell_B^2} = \frac{eBS}{\hbar}.
\]  

(3.14)

This is generally a very large number and, therefore, LLs are massively degenerate in energy.\(^9\)

Alternatively, consider the total magnetic flux penetrating a 2DEG of area \( S \), which is

\[
\Phi = \mathbf{B} \cdot \mathbf{S}.
\]  

(3.15)

\(^7\) Had we considered a three-dimensional system we would still find discrete Landau level energies associated with motion perpendicular to the magnetic field, but with a continuum of allowed energies due to motion in the \( \hat{z} \)-direction that would smear out the quantization. If we include Zeeman spin splitting, the spectrum is more generally given by

\[
E_{k,N} = \hbar \omega_c \left( N + \frac{1}{2} \right) + \frac{\hbar^2 k_z^2}{2m^*} \pm \frac{1}{2} g^* \mu_B B
\]  

(3.11)

where here \( g^* \) is the effective electron \( g \)-factor.

\(^8\) There is occasionally some ambiguity in how the term “Landau level” is used. We refer to all states with quantum number \( N \) as a “Landau level.” The true degeneracy of a LL is given \( gN_\Phi \), where \( g \) counts the internal degrees of freedom of the system. Each LL, \( N \), contains \( g \) LL sub-bands.

\(^9\) In a magnetic field of 10 T a 100\(\mu m^2\) 2DEG can accommodate \( 2.4 \times 10^5 \) carriers per Landau level per degree of freedom.
The quantum of magnetic flux is given by

$$\Phi_0 = \frac{h}{e}, \quad (3.16)$$

and the ratio of the two gives the total number of flux quanta penetrating the 2DEG

$$\frac{\Phi}{\Phi_0} = \frac{eBS}{h} = N_\Phi, \quad (3.17)$$

which is exactly the number of allowed states per LL sub-band given by (3.14). Thus, the number of charge carriers that may occupy a given Landau level, $N$, is equal to the number of magnetic flux quanta penetrating the 2DEG, $N_\Phi$, multiplied by the degeneracy, $g$, due to internal degrees of freedom.

It is useful to define a quantity that tracks the filling of Landau levels. If the total number of charge carriers in the 2DEG is $N$, the carrier density is $n = N/S$. The ratio of the total number of charge carriers in a 2DEG to the number required to fill a LL sub-band

$$\nu = \frac{N}{N_\Phi} = \frac{h n}{e B}, \quad (3.18)$$

is known as the Landau level filling factor and plays an important role in the physics of the quantum Hall effect.

The spectrum (3.12) also leads to a dramatic rearrangement of the density of states (DOS). In the complete absence of disorder, the DOS of a Landau quantized 2DEG becomes an equally spaced ladder of Dirac delta functions\textsuperscript{10} of magnitude $g N_\Phi$ at energies $E_N = h \omega_c (N + 1/2)$.

It is instructive to consider the behavior of the system as we add charge carriers to it. Starting from zero carrier density, an energy $h \omega_c (1/2)$ is required to add each carrier, which subsequently occupy the $N = 0$ LL. Once $g N_\Phi$ carriers have been added, the addition of the next now requires an energy $h \omega_c (1 + 1/2)$, and it occupies $N = 1$. This pattern repeats

\textsuperscript{10} Disorder serves to broaden these delta functions into bands of finite width.
for all $N$, with the chemical potential, $\mu$, pinning to the lowest unfilled LL and jumping discontinuously to the next once it is filled. The behavior of the chemical potential as carriers are added or removed can be related to the thermodynamic compressibility of the system, which measures the energetic cost of shifting $\mu$ an infinitesimal amount. When $\mu$ lies inside a LL, shifting it infinitesimally requires an infinitesimal cost in energy, and the system is said to be compressible. Upon complete filling of a LL, shifting $\mu$ infinitesimally requires a finite energy, and the system is rendered incompressible. A complete explanation of the quantum Hall effect, which is not attempted here,\textsuperscript{11} follows from the fact that this incompressibility leads to quantized electronic conduction [146]. It is worth noting that this incompressible behavior may arise in two distinct ways [145]: (1) The quantization of kinetic energies into units of $\hbar \omega_c$ which, as shown above, may be accounted for by considering only single-particle physics, results in the integer quantum Hall effect (IQHE). (2) Interactions between electrons can produce gapped behavior at energies other than $\hbar \omega_c$, resulting in the fractional quantum Hall effect (FQHE). A brief discussion of the IQHE and FQHE are given in the following section.

\textbf{3.2 The Quantum Hall Effect}

It was recognized in the first half of the twentieth century that a two-dimensional electron gas (2DEG) could be trapped at the interface between two dissimilar materials. Practical devices, in the form of metal-oxide-semiconductor field-effect transistors (MOSFETs), were first realized in 1959 at Bell Laboratories and saw intense development through the 60’s and 70’s, largely driven by the incentives of an emerging semiconductor industry to realize transistors with ever greater electron mobility. In these devices a metal layer is placed on silicon dioxide that was grown on silicon. Applying a voltage to the metal “gate” electrode, charge carriers could be induced at the silicon-silicon dioxide interface with a density of $10^{10}$

\textsuperscript{11}For a full account of the quantum Hall effect, we refer the interested reader to the excellent review by A. H. MacDonald (p.195-217) in Quantum Coherence in Mesoscopic Systems [145].
to $10^{13}$ cm$^{-2}$. The so-called inversion layer of charge carriers in such systems are typically less than 10 nm thick in the direction perpendicular to the interface [147].

![Fig. 3.1: The integer quantum Hall effect at constant carrier density as a function of magnetic field. Adapted from figure 14 of reference [147].](image)

3.2.1 The Integer Quantum Hall Effect

In 1980,\textsuperscript{12} while exploring the Hall effect in a silicon MOSFET 2DEG, physicist Klaus von Klitzing made a remarkable discovery: at certain values of electronic density and magnetic field, when the Landau level filling factor $\nu$ is an integer,\textsuperscript{13} the conductance transverse to an applied current, also known as the Hall conductance, took on quantized values given by

$$\sigma_{xy} = \nu \frac{e^2}{h},$$

(3.19)

\textsuperscript{12} Investigations of the high field magnetotransport in 2DEGs had suggested anomalous behavior as early as 1978 [148]

\textsuperscript{13} This integer has been shown to have topological origins rooted in Chern-Simmons theory [75]
while simultaneously the resistance parallel to the current, also known as the magnetoresistance, $R_{xx}$, became vanishingly small\textsuperscript{14} [147]. This incredible behavior arises when the Fermi energy lies between Landau levels: while the Fermi energy in the bulk of the 2DEG lies inside the cyclotron gap, there exist gapless current carrying edge states at the boundary of the sample. These exotic edge states, one for each filled LL sub-band, have a topological origin and each carry a dissipationless current\textsuperscript{15} with conductance $e^2/h$. When the Fermi energy lies inside a Landau level the bulk once again supports extended states which are subject to scattering from various sources of disorder, the Hall conductance resumes its classical value of $\sigma_{xy} = ne/B$, and the magnetoresistance is once again finite [151].

Quantization is so fundamental to quantum mechanics that it literally lends its name. What is so remarkable about the quantization of the Hall conductance discovered by von Klitzing is that it is observed in macroscopic, disordered samples and yet its value has been demonstrated to deviate from eq. (3.19) at the level of only a few parts per million. Klaus von Klitzing was awarded the 1985 Nobel Prize in Physics “for the discovery of the quantized Hall effect.”

### 3.2.2 The Fractional Quantum Hall Effect

In the history of condensed matter there is a pattern that has repeated itself over and over again: cleaner samples inevitably lead to new physics. The discovery of the integer quantum Hall effect (IQHE) in silicon MOSFETs naturally set off intense interest in the study of 2DEG magnetotransport. Attention was quickly turned toward samples in which the 2DEG could be confined to potential wells of considerably higher uniformity, which was realized in the recently developed modulation doped GaAs/AlGaAs heterostructure quantum wells grown by molecular beam epitaxy.

\textsuperscript{14} See Appendix B for a summary of transport measurements.

\textsuperscript{15} The quantity $e^2/h$, also known as the quantum of conductance, is the conductance of a single one-dimensional quantum channel in the limit that the probability of transmitting an electron through that channel goes to 1, a situation achieved when the electron’s motion is ballistic [149,150]. The inverse of the conductance quantum, $R_K = h/e^2 = 25812.8165 \, \Omega$, is known as the von Klitzing constant.
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The disorder in these epitaxial devices proved to be so low that the previously obscured energy scale on which electron-electron interactions occur was revealed, with the correlated behavior of many electrons manifesting new incompressible ground states and, therefore, quantized Hall conductance at *fractional* values of $\nu$. Many aspects of the fractional quantum Hall effect (FQHE) are now well-understood; others remain deep mysteries and the subject of intense research [152,153]. Horst Störmer, Daniel Tsui, and Robert Laughlin were awarded the 1998 Nobel prize in Physics “for their discovery of a new form of quantum fluid with fractionally charged excitations.”

![Graph of the fractional quantum Hall effect](image)

**Fig. 3.2:** The fractional quantum Hall effect observed at constant carrier density as a function of magnetic field. Reproduced from figure 1 of reference [151].

### 3.2.3 The Relativistic Quantum Hall Effect

The most recent leap in the development of quantum Hall physics arguably came in 2004, with the discovery of the relativistic quantum Hall effect\(^\text{16}\) in graphene [73,154,155]. Graphene’s band structure is defined by four unique properties: a linear energy-momentum relationship, an additional spin-like internal degree of freedom associated with the bipartite honeycomb

\(^{16}\) More commonly known as the “half-integer quantum Hall effect” and less frequently as the “chiral quantum Hall effect.”
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lattice, a high level of electron hole symmetry, and a topological singularity at zero energy that imparts a non-trivial geometric phase to the electron and hole wave functions when they undergo cyclotron motion. All four are deeply related to the quasi-relativistic nature of the charge carriers in graphene, and all four are manifest in the way it forms LLs. The result is a Hall conductance given by

\[ \sigma_{xy} = \pm 4 \left( N + \frac{1}{2} \right) \frac{e^2}{h} \]

where the factor of 4 is due to the combination of spin and valley degeneracies, and the \( \pm \) accounts for both electron and hole LLs. The shift of the filling factor sequence \( \nu = N \rightarrow \nu = N + 1/2 \) relative to that in eq. (3.19) is a direct consequence of the Berry phase of \( \pi \) accumulated by charge carriers in a magnetic field, which gives rise to a true zero energy zeroth Landau level and, at charge neutrality, is equally occupied by electrons and holes. Andre Geim and Konstantin Novoselov were awarded the 2010 Nobel prize “for groundbreaking experiments regarding the two-dimensional material graphene.”

![Fig. 3.3:](image)

Fig. 3.3: The relativistic quantum Hall effect observed in graphene at a constant magnetic field as the carrier density is swept. Reproduced from figure 1 of reference [155].
3.3 Cyclotron Resonance

3.3.1 The Evolution of Cyclotron Resonance

Cyclotron resonance, the resonant absorption of light by charged particles undergoing cyclotron motion in the presence of a magnetic field, has been employed to study charge carriers in solid state materials for over half a century. In that time it has undergone several significant developments in the way it is applied.

Predictions by Dorfman and Dingle [156] in 1951, and Shockley in 1953 [157], had indicated that cyclotron resonance was a promising method for experimentally probing the shapes of Fermi surfaces in materials, a problem for which few solutions were known. Shortly thereafter, the first observation of cyclotron resonance (CR) in condensed matter was made by Dresselhaus, Kip, and Kittel studying germanium crystals in 1953 [158]. Follow up observations of CR in Si in 1954 by the same authors [159] set off an avalanche of interest in applying the technique to semiconductors and, shortly thereafter, to metals and semimetals [157,160].

Classically, the Lorentz force acting on a charged particle induces cyclotron motion with frequency $\omega_c = eB/m$. In materials, charge carriers near the band edge are well-described in the effective mass approximation in which $m \rightarrow m^*$, where the effective mass $m^*$ describes the inertial response of a charged particle propagating in a periodic potential.

Observation of resonant absorption of electromagnetic radiation at a frequency $\omega = \omega_c = eB/m^*$, in

\[ m^* = \frac{m}{m_e} \]

In germanium crystals held at 4 K they observed resonant absorption of 9.05 GHz radiation in a magnetic field of 37 mT, corresponding to an effective mass of $m^* = 0.11 m_e$.

In the more general case where motion is unconfined in the z-direction these orbits are helical.

An external force acting on a crystal electron produces a change in its wavevector $\mathbf{k}$ and, therefore, its energy, $\mathcal{E}(\mathbf{k})$, through $\mathbf{F} = \hbar \partial \mathbf{k} / \partial t$. The expectation value of the velocity of a crystal electron is $\langle v \rangle = (1/\hbar) \partial \mathcal{E} / \partial \mathbf{k}$. From these relations one finds

\[ \frac{\partial \langle v \rangle}{\partial t} = \frac{1}{\hbar^2} \frac{\partial \mathcal{E}^2}{\partial \mathbf{k}^2} = \frac{1}{m^*} \mathbf{F}, \]

from which the definition of $m^*$ in eq. (3.22) follows [161] [162]. While it must be noted that the cyclotron mass $m_c$ measured in CR is not formally equivalent to the effective band mass $m^*$, they are very nearly identical in most cases.
a known magnetic field $B$, thus allows for an accurate determination of $m^*$. In general, $m^* = m^*_{ij}$ is a tensor quantity that varies with respect to the crystal axes of the investigated material. Measurement of $\omega_c$ for various orientations allows for determinations of

$$\frac{1}{m^*_{ij}} = \frac{1}{\hbar^2} \frac{\partial \mathcal{E}^2}{\partial k_i \partial k_j}.$$ (3.22)

As elucidated by Azbel and Kaner, observations of the number of resonances for a given crystal orientation and their anisotropies\(^{20}\) allows for a determination of the geometry and topology of a material’s Fermi surface.

Resonance intensities and widths were found to be sample and temperature dependent, and it was quickly recognized that CR contained a wealth of information beyond the effective mass. While the integrated intensity of a resonance is proportional to the number of excited

---

\(^{20}\) Here anisotropy includes not just the variation of a resonance as the angle between crystal axes and \(B\) is changed, but also the dependence of a resonance on the polarization state of the incident radiation.
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charge carriers, the resonance width\(^{21}\) \(\Gamma\), which defines the cyclotron lifetime

\[
\tau_{CR} = \frac{\hbar}{\Gamma};
\]  

was found to be a sensitive probe of various scattering mechanisms \cite{164}. Thus CR evolved into a powerful probe of carrier dynamics\(^{22}\). Thus it fed the material characterization needs of a nascent semiconductor industry and simultaneously drove new theoretical developments in the understanding of carrier dynamics in condensed matter.

The next evolution in the application of CR came when it was applied (reword) to the two-dimensional electron gas. Observation of CR in a silicon MOSFET 2DEG was reported as early as 1973 \cite{166,167}, but the discovery of the integer and fractional quantum Hall effects in the early 1980s naturally set off an explosion of interest in turning the now mature technique of CR on these exotic new material systems, particularly the cleaner GaAs/AlGaAs heterostructures. CR was found to offer an experimental probe complementary to that of electronic transport; when the Fermi energy lies inside a cyclotron gap, transport probes only the edge states of a quantum Hall system, while CR always probes LLs in the bulk of the material.

Given the strong interactions between electrons confined to two dimensions, and the explicitly many-body nature of the FQHE, one might assume that cyclotron resonance applied to such systems would also be subject to interaction effects. A 1961 theorem\(^{23}\) by Walter Kohn, which we consider in detail in the final section of this chapter, would seem to eliminate this possibility: on simple theoretical grounds he argued that in a Landau quantized, translationally-invariant system possessing a parabolic dispersion relation, radiation couples

\(^{21}\) Note that \(\Gamma = \text{HWHM}\) is half the width at half the maximum resonance intensity, which we refer to simply as the width. The half-width is employed due to the definition of the Lorentzian lineshape typically observed in CR \cite{163}.

\(^{22}\) As noted in Ref. \cite{165}, the CR lifetime \(\tau_{CR}\) is closely related to the transport lifetime \(\tau_\mu\) only in the low-field limit. At high fields, when the majority of carriers occupy the lowest LLs, \(\tau_{CR}\) is more closely related to the quantum scattering time \(\tau_\sigma\).

\(^{23}\) Kohn’s theorem should not be confused with the more widely known Kohn-Sham theorem that underpins much of Density Functional Theory.
only to the center-of-mass motion, forbidding the interactions between charge carriers from impacting the cyclotron resonance energy. Fortunately, in real material systems Kohn’s theorem is broken to varying degrees by the presence of disorder and, in some cases, non-parabolic energy-momentum relations. A number of unusual observations were subsequently reported in the CR of 2DEGs including anomalous splitting and shifting of resonances [168–170], filling factor dependent oscillations of resonance linewidths and amplitudes [171–174], and signatures of Wigner crystallization [175] that have been explained on the grounds of many-particle interactions [176]. Many of these observations still lack widely accepted explanations.

Fig. 3.5: The first observations of cyclotron resonance in exfoliated monolayer graphene. Reproduced from figure 2 of reference [18].

The most recent evolution in the use of CR came with its application to the atomically thin, two-dimensional material graphene. After the discovery by Novosolov and Geim in 2004 [6,155,177] that graphene monolayers hosted an electron gas displaying the long-expected
behavior of Dirac electrons, CR measurements by Jiang, et al. [18] and others [19] in 2007 confirmed the unusual $\sqrt{NB}$ scaling of LL energies expected for relativistic Dirac fermions. These measurements also suggested deviations from the simple single particle picture of CR in graphene, which were later more concretely observed by Henriksen, et al. [20]. Open questions regarding the nature of many-body interaction effects on the cyclotron resonance in graphene, combined with advances in the construction of high quality graphene-based Van der Waals heterostructures (VDWHSs), can be considered as the primary motivations for the work presented in this thesis. Cyclotron resonance will likely continue to play a role in investigating graphene VDWHSs as they grow in quality and complexity. Just as exciting is the idea of using CR to study the cornucopia of atomically-thin materials that have been discovered over the last decade and half, as well as new classes of materials which have emerged over the same period such as topological insulators, and Dirac, Weyl, and nodal semimetals.

### 3.3.2 Conditions for Observing Cyclotron Resonance

Cyclotron resonance is observed when three basic criteria are satisfied. In materials, scattering due to impurities, defects, and phonons introduces a natural timescale, $\tau$, on which cyclotron motion remains coherent. The first condition quantifies the requirement that a well-defined resonance is observed when the scattering rate is comparable to the timescale of cyclotron motion $^{24}$

$$\omega_c \tau = \mu B >> 1,$$

where $\mu$ is the carrier mobility$^{25}$ [158]. Intuitively, this means the system must be clean enough, or the magnetic field strong enough, that charge carriers execute at least one complete cyclotron orbit between scattering events. Electron-phonon scattering also generally

$^{24}$ The equality in eq. (3.24) can be found by making use of the relationship $\omega_c \tau = (eB/m^*)\tau = \mu B$.

$^{25}$ As noted earlier for $\tau$, at low fields the mobility $\mu$ is approximately the same as the mobility measured in transport.
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decreases with temperature, thus cryogenic environments are usually required\(^{26}\). For the second condition, recall from earlier in the chapter that the density of states in a Landau quantized system is composed of a series of narrow peaks separated by the cyclotron energy \(\hbar \omega_c\). Charge carriers occupy these Landau levels according to the Fermi-Dirac distribution, \(f_{FD}(E) = \left[ e^{(E-E_F)/k_B T} + 1 \right]^{-1}\), and at finite temperature \(T\) there will be a region of width \(\sim k_B T\) about the Fermi energy over which LLs will be partially occupied. Thus, observation of a well-defined resonance between neighboring LLs requires the energy scale associated with thermal broadening to be small relative to the cyclotron energy

\[
k_B T \ll \hbar \omega_c, \tag{3.25}
\]

a condition that can always be met with a large enough magnetic field.\(^{27}\)

Finally, the third condition for observing cyclotron resonance

\[
\omega = \omega_c, \tag{3.26}
\]

quantifies the fact that only incident electromagnetic radiation with a frequency equal to the cyclotron frequency\(^{28}\) will be resonantly absorbed.

\(^{26}\) The weak electron-phonon coupling and high phonon energies in graphene allow CR to be observed as high as room temperature [178].

\(^{27}\) The cyclotron energy scale in graphene is naturally very high. At \(B = 10\ T\) the characteristic cyclotron energy is \(v_F \sqrt{2eB} = 114.7\ \text{meV} = 1331\ \text{K}\) and condition 2 is easily met. Combined with the weak temperature dependence of scattering in graphene, which enters through condition 1, one finds the temperature dependence of cyclotron resonance in graphene is weak.

\(^{28}\) In GaAs at 10 T one finds \(\omega_c = eB/m^* \approx 23\ \text{meV} = 5.6\ \text{THz}\), deep in the far-infrared. For graphene in the same magnetic field one finds \(\omega_c = v_F \sqrt{2eB/\hbar} = 114.7 \text{meV} \approx 28\ \text{THz}\), placing cyclotron resonance in the mid-infrared.
The Quantum Theory of Cyclotron Resonance and Kohn’s Theorem

To give a quantum accounting of cyclotron resonance, we now revisit the Landau level derivation from section 3.1. We begin by rewriting Hamiltonian (3.3) in terms of the kinetic momentum

\[ H_0 = \frac{1}{2m} \left( p + eA \right)^2 = \frac{1}{2m} \left( \pi_x^2 + \pi_y^2 \right). \]  

(3.27)

In addition to the usual commutation relations

\[ [x, p_x] = [y, p_y] = i\hbar \]  

(3.28a)

\[ [x, y] = 0 \]  

(3.28b)

\[ [p_x, p_y] = 0 \]  

(3.28c)

we find

\[ [x, \pi_x] = [y, \pi_y] = i\hbar \]  

(3.29a)

\[ [x, \pi_y] = [y, \pi_x] = 0 \]  

(3.29b)

\[ [\pi_x, \pi_y] = -i\hbar^2/\ell_B^2. \]  

(3.29c)

Thus, in contrast to the components of the canonical momentum, the components of the kinetic momentum in the presence of a magnetic field do not commute with one another. As with any quantum harmonic oscillator, we can define raising and lowering operators

\[ \hat{a}_\pm = \frac{\ell_B^2}{\sqrt{2}\hbar} \left( \pi_x \pm i\pi_y \right), \]  

(3.30)

which are related to one another by

\[ [\hat{a}_+, \hat{a}_-] = 1, \]  

(3.31)
and to the Hamiltonian (3.27) by

\[ [H_0, \hat{a}_\pm] = \pm i\hbar \omega_c. \]  

(3.32)

The action of \( \hat{a}_\pm \) on an eigenstate \( |N\rangle \) of Hamiltonian (3.27) is

\[ \hat{a}_+ |N\rangle = \sqrt{N+1} |N+1\rangle \]  

(3.33a)

\[ \hat{a}_- |N\rangle = \sqrt{N} |N-1\rangle, \]  

(3.33b)

and the combined action of \( \hat{a}_+ \) and \( \hat{a}_- \) defines the number operator

\[ \hat{N} = \hat{a}_+ \hat{a}_-, \]  

(3.34)

which we may then use to rewrite Hamiltonian (3.27) as

\[ H_0 = \hbar \omega_c \left( \hat{a}_+ \hat{a}_- + \frac{1}{2} \right). \]  

(3.35)

So far we have only quantized the harmonic oscillator spectrum discussed at the beginning of the chapter. Following Ref. [163], and making use of the electric dipole approximation,\(^{29}\) we introduce a perturbing vector potential

\[ \mathbf{A}_\pm' = - \frac{\mathcal{E}_0}{\sqrt{2}i\omega} (\hat{e}_x \pm \hat{e}_y) e^{-i\omega t}, \]  

(3.36)

which represents the time-varying electric field of left-handed (\( \mathbf{A}_+ \)) or right-handed (\( \mathbf{A}_- \)) circularly polarized light of frequency \( \omega \). Adding the perturbation (3.36) to the Hamiltonian

\(^{29}\) The electric dipole approximation, in which \( \exp^{ik\mathbf{r} \cdot \mathbf{r}} \rightarrow 1 \), is valid provided that the dimensions of the relevant charge distribution are small compared to the wavelength of the light being emitted or absorbed. Put another way, it is valid when the electric field of the electromagnetic wave is spatially uniform on the scale of the emitter or absorber.
(3.27) we find

\[ \mathcal{H} = \frac{1}{2m^*} \left( \mathbf{p} + e \mathbf{A} + e A'_{\pm} \right)^2 \approx \frac{1}{2m^*} \pi^2 + \frac{e}{m^*} \mathbf{\pi} \cdot \mathbf{A}'_{\pm} = \mathcal{H}_0 + \mathcal{H}'. \]  

(3.37)

Noting that the perturbing Hamiltonian \( \mathcal{H}' \) can be rewritten in terms of the raising and lowering operators (3.30)

\[ \mathcal{H}' = \frac{e}{m^*} \left( \pi_x A'_{\pm,x} + \pi_y A'_{\pm,y} \right) = \frac{-e \mathcal{E}_0}{\sqrt{2i\omega m^*}} (\pi_x \pm i\pi_y) e^{-i\omega t} = \frac{-e \hbar \mathcal{E}_0}{i\omega m^* \ell_B} \hat{a}_{\pm} e^{-i\omega t}, \]  

(3.38)

we find that radiation of frequency \( \omega = \omega_c \) can be absorbed by charges undergoing cyclotron motion, inducing transitions \( N \rightarrow N + 1 \) between Landau levels.

In 1961 it was shown by Walter Kohn [179] that in a translationally-invariant system of Landau quantized charge carriers interacting via the short range Coulomb force, \( U \), electromagnetic radiation couples only to the center of mass motion of the system, corresponding to excitations between LLs \( N \rightarrow N + 1 \) at the cyclotron energy \( \hbar \omega_c \). Adding the interaction \( U \) to the Hamiltonian (3.27), one finds that the commutation relation (3.32) remains unchanged and, therefore, the cyclotron resonance frequency is insensitive to electron-electron interactions.

Kohn’s derivation, valid for materials with parabolic band structures and therefore harmonic oscillator Landau level spectra, clearly breaks down when applied to graphene’s unusual \( \sqrt{N} \) LL sequence. While in practice, disorder or non-parabolicity of the band structure can render cyclotron resonance in traditional, parabolic 2DEGs weakly sensitive to interactions [168, 170–173, 180, 181], graphene’s linear dispersion should enable CR that is intrinsically sensitive to interactions [18, 20–22, 42–44, 182–192].
Chapter 4

Cyclotron Resonance in Encapsulated Monolayer Graphene

In the presence of a magnetic field, the linear dispersion of electrons in graphene is quantized into a set of discrete Landau levels (LLs) with energies exhibiting an unusual square-root dependence on both the magnetic field strength \( B \) and the LL index \( N \)

\[
E_N = \text{sign}(N) \left| v_F \sqrt{2e\hbar B} \right| N, \quad N = 0, \pm 1, \pm 2, \ldots
\]  

(4.1)

where \( v_F \approx 1 \times 10^6 \) m/s is the Fermi velocity [17]. This unusual spectrum is a direct consequence of the relativistic nature of charge carriers in graphene.\(^1\) At magnetic fields accessible in the laboratory, cyclotron resonance (CR) transitions between LLs in graphene occur at energies corresponding to light at far- and mid-infrared frequencies. Cyclotron resonance measurements were crucial in confirming the unusual index and field dependence of eq. (4.1), providing strong evidence for the Dirac-like nature of charge carriers in graphene; subtle deviations from the expected behavior also hinted at the presence of interaction effects in these early graphene on silicon dioxide devices [18,19,88,193].

The parabolic dispersion found in more conventional two-dimensional electron gas (2DEG) systems, such as GaAs heterostructures, leads to an evenly spaced harmonic oscillator LL

\(^1\) For a discussion of how this spectrum compares to a conventional 2DEG see Ch. 2. For details concerning its relativistic origin see Ch. 3.
spectrum, resulting in a single CR absorption peak. In contrast, the square-root index dependence and dipole selection rules in graphene enable many transitions to occur simultaneously, yielding richer optical spectra. Further, in parabolic systems long-wavelength light couples only to the center-of-mass motion, so that in principle electron-electron interactions do not impact the CR, in accordance with Kohn’s theorem\(^2\) [179]; in practice, disorder or non-parabolicity of the band structure can render the CR weakly sensitive to interactions in parabolic systems [168, 170–173, 180, 181]. Intriguingly, the linear dispersion found in graphene should enable CR that is intrinsically sensitive to interactions [18, 20, 43, 182–184], leading to widespread anticipation that optical probes of graphene will provide a novel window on the many-body problem [21, 22, 42, 44, 185–192].

Performing CR measurements for the first time on gate-tunable, high-mobility encapsulated monolayer graphene, we observe unambiguous evidence of many-body interaction contributions to the single-particle CR transition energies, manifesting as a non-monotonic variation of the charge carrier effective Fermi velocity in a magnetic field. The success of these measurements followed two key developments within the Henriksen lab: The first was the design and construction of a unique experimental system capable of performing broadband infrared spectroscopy and simultaneous electronic transport on microscopic, atomically thin materials held at cryogenic temperatures and subjected to intense magnetic fields up to 14 tesla.\(^4\) The second key was the in-house development of a capability for producing the high-quality, hexagonal boron nitride encapsulated graphene devices that were first demonstrated by Dean, et al. in 2010 [100], with the added challenge that, in order to obtain a useable optical signal, these devices must be relatively large (lateral dimensions of \(l \sim 10 \mu m\) or more) compared to typical devices used in electronic transport studies (\(l \sim 2 \mu m\)). The results that follow stand as proof that the wedding of high performance magneto-optical probes

\(^2\) For further details, see the discussion of Kohn’s theorem in Ch. 3.

\(^3\) Note that the theoretical work in references [22] and [21] were written in response to the results presented in this chapter and published in Physical Review Letters in 2018 [194]

\(^4\) For detailed discussion of the design, construction, and capabilities of this infrared system the reader is referred to Appendix A.
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with high quality samples has much to offer in the pursuit of electron-electron interaction physics in condensed matter.

4.1 Electron-Electron Interaction Effects on the Cyclotron Resonance of Monolayer Graphene

As seen from eq. (4.1) and discussed in detail in chapter 2, the single-particle theory of graphene predicts an unusual spectrum of LL energies. Uniquely, both intraband and interband Landau level transitions are allowed, with the interband transition energies given by

$$\Delta E_{MN} = v_F \sqrt{2e\hbar B} \left( \sqrt{|M|} + \sqrt{|N|} \right),$$  

(4.2)

where $M$ and $N$ index the initial and final LLs, and $v_F \approx 10^6$ m/s is the carrier Fermi velocity which is nominally constant. However, electron-electron interactions can contribute an additional energy $\delta E_{ee} = C_{MN}(B, \mu) e^2/\ell_B$, where the transition matrix elements $C_{MN}(B, \mu)$ may depend on the initial and final LL states involved in the transition, the magnetic field strength, and the chemical potential, $\mu$. The quantity $e^2/\ell_B$ sets the Coulomb energy scale in the presence of a magnetic field, where the so-called magnetic length, $\ell_B = \sqrt{\hbar/eB}$, is the quantum mechanical analogue of the classical cyclotron radius. Combining the single-particle transition energy with the many-body contributions yields

$$\Delta E_{total}^{MN} = \Delta E_{MN} + \delta E_{ee} = (v_F^{eff}/v_F) \Delta E_{MN},$$  

(4.3)

where the “effective” Fermi velocity $v_F^{eff} \geq v_F$ accounts for the observed interaction contribution (cite). Because $\Delta E_{MN}$, $C_{MN}$, and $e^2/\ell_B$ all vary with $B$, it is difficult to clearly distinguish many-body contributions to the transition energy by changing the magnetic field alone, however, any variation of $v_F^{eff}$ at fixed field will reflect changes arising from electron-electron interactions.
Therefore, in this work we elected to study the CR transitions in our high-mobility graphene monolayer at constant $B$, while tuning the carrier density $n$ through its dependence\(^5\) on the back gate voltage $V_g$ applied to the silicon substrate. Changing the carrier density $n$ in the presence of a magnetic field $B$ amounts to controlling the Landau level filling factor, $\nu = nh/eB$, where $h$ is Planck’s constant. Thus, while the LL spacings and Coulomb energy are held fixed, the physics of correlated electrons in partially filled LLs may yet impact the CR. Indeed, as we will discuss in detail, we observe a clear non-monotonic dependence of $v_{\text{eff}}^\nu$ on the filling factor (FF), with the detailed behavior changing for each of the six interband LL transitions measured.

### 4.2 Monolayer Device, Electronic Transport, and Magnetospectroscopy

The sample consists of a graphene monolayer encapsulated between two crystals of hexagonal boron nitride (hBN), stacked on top of 300 nm of thermal silicon dioxide which was grown on a lightly doped, 620 $\mu$m thick silicon wafer. The thickness of the hBN crystals is estimated at 30 and 15 nm for the top and bottom layers, respectively; the total area of the encapsulated stack is 250 $\mu$m$^2$. Although the device has two interfaces with hBN crystals, there is no evidence of the satellite Dirac peaks that are indicative of a moiré superlattice or the accompanying Hofstadter butterfly physics [29].

Electrical contact to the device was made using the widely employed one-dimensional edge contact method [195] (Science.614). First, the hBN-graphene-hBN stack was etched to expose the single-atom thick edge of the graphene. Next, a contact-defining etch mask was then written using electron beam lithography and 60 nm of Ti-Al was deposited so as to

\[^5\] $n = \alpha (V_g - V_D)$ where $\alpha$ is the gate efficiency and $V_D$ is the Dirac point voltage. $V_D$ is subtracted off to account for the effects of unintentional charge doping due to the presence of charged impurities near the graphene. Here we find $\alpha \approx 7 \times 10^{10}$ V$^{-1}$ cm$^{-2}$ and $V_D = -10.7$ V, both typical for devices of this architecture.
Fig. 4.1: (Left) An illustration of the device architecture employed in this work. A large monolayer of graphene is encapsulated between two crystals and deposited on an oxidized silicon wafer. Application of a gate voltage between the graphene and the silicon substrate allow the carrier density to be tuned over a wide range, as discussed in Ch. 2. (Right) The top image shows the monolayer device on a sample mount which provides a simple interface with measurement probes. The next image in the sequence shows the $\sim 90 \mu m$ foil aperture placed over the device to reduce the amount of unwanted light reaching the infrared detector. The final image is an optical micrograph of the finished device, which has an area of $\sim 250 \mu m^2$

overlap the sides of the stack. Finally, the etch mask was removed. The resulting contact resistances were found to be $\sim 300 \Omega$. Electronic transport measurements were made using standard low-frequency lock-in techniques. From these measurements it is apparent that the device exhibits the hallmarks of high-quality graphene. As shown in figure (a), the zero field resistivity exhibits a sharp Dirac peak with a full width at half maximum of only $\delta n = 1.12 \times 10^{11} \text{cm}^{-2}$ centered around zero carrier density. The electron (hole) field effect mobility of $190 000 \ (290 000) \ \text{cm}^2 \ \text{V}^{-1} \ \text{s}^{-1}$ was measured.

In figure (4.2b) the magnetoresistance, $R_{xx}$, is shown in what is commonly referred to as a Landau fan, where it is plotted versus both the magnetic field $B$ and the gate voltage $V_g$. Additional measurements of $R_{xx}$ are shown at constant field fig. (4.2c) and constant $V_g$ fig. (4.2d). Minima in the magnetoresistance $R_{xx}$ and plateaus in the Hall resistance
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Fig. 4.2: Electronic transport at T = 6 K. (a) Zero-field conductivity and resistivity vs carrier density, $n$; the inset shows an optical micrograph of the 250 $\mu$m$^2$ encapsulated graphene device used in this work. (b) Magnetoresistance vs gate voltage, $V_g$. Note that up to $V_g = \pm 80$ V, there is no evidence of additional Dirac peaks sometimes observed in transport of hBN-encapsulated graphene [31, 32, 196, 197]. (c) Magnetoresistance traces at constant field $B = 11$ T, and (d) Shubnikov-de Haas oscillations.

$R_{xy}$ were observed at carrier densities corresponding to LL filling factors $\nu = \pm 4(N + 1/2)$, characteristic of monolayer graphene in the absence of symmetry breaking effects. The deeper minima in $R_{xx}$ at negative $V_g$ correlate with the higher hole field effect mobility. The transport was negligibly impacted by exposure to IR light.

Spectroscopy was performed using a custom built system which combines a Fourier transform infrared spectrometer with a closed cycle dilution refrigerator by way of geometric optics. Broadband infrared light is sourced from a silicon carbide globar inside the spectrometer, transmitted through vacuum to the base of the cryostat, and admitted into the fridge through a KBr window. Once inside the fridge the light is focused onto the sample and subsequently re-collimated using custom paraboloidal reflecting optics, before finally reaching a compound parabolic condensing cone. At the end of the condenser, light is detected by a high performance composite silicon bolometer, which is maintained at $\sim 4$ K.

---

6 The absence of symmetry breaking of the four-fold degenerate Landau levels in a monolayer device of this quality is likely attributable to the elevated temperature of $\sim 6$ K [87].
4.3 Cyclotron Resonances and Effective Fermi Velocities

Mid-infrared spectra were acquired at a sample stage temperature of $T = 6$ K in magnetic fields of $B = 5$, 8, and 11 T as a function of the Landau level filling factor $\nu$. Spectral coverage was from roughly 60 to 600 meV ($500$ to $5000$ cm$^{-1}$) with a resolution of 1 meV ($8$ cm$^{-1}$). In order to remove sample independent features in the raw IR spectra, traces taken at filling factors of interest were divided by spectra obtained at high filling factors, where all observable interband transitions are Pauli blocked. Normalizing spectra in this way, by electronically modulating the allowed transitions in the sample, ensures that all substrate absorption features are removed. To study the interband transitions for $|\nu| \leq 10$, the corresponding spectra were normalized to the transmission at $\nu = 22$ and plotted as $S(\nu)/S(\nu = 22)$, where $S$ is the raw transmission signal. Thus at a given $\nu$ value, resonant absorption of IR light leads to reductions in $S$ which appear as dips in the normalized spectra; in fig. (4.3a) and fig. (4.4) we invert the normalized spectra so these resonances appear as peaks. An example of a typical normalized spectra taken at $\nu = 0$ and $B = 8$ T is shown in fig. (4.3a). Several resonances labeled $T_1$ through $T_6$ are clearly visible, spanning the measured range of frequencies. A series of such traces taken over a range of $\nu$ values at $B = 8$ T is combined into a colormap in fig. (4.3b), from which it is clear that the observed transitions hew closely to the square root dependence on LL index described by eq. (1).

(4.3c) illustrates the allowed interband Landau level transitions at a filling factor $\nu = +2$ according to the selection rule $N = |M| \pm 1$ [23]. Figure (4) shows the detailed $\nu$-dependence of the four lowest transitions, $T_1$ ($0 \rightarrow 1$ and $-1 \rightarrow 0$), $T_2$ ($-1 \rightarrow +2$ and $-2 \rightarrow +1$), $T_3$ ($-2 \rightarrow +3$ and $-3 \rightarrow +2$), and $T_4$ ($-3 \rightarrow +4$ and $-4 \rightarrow +3$). Notably, $T_2$, $T_3$, and $T_4$ show similar behavior: for $|\nu| < 2$, a single peak remains largely unchanged as $\nu$ is varied, but for $|\nu| > 2$ the peak begins to broaden and show shifts of the central energy; this effect is most pronounced for $T_3$ and $T_4$. Note that,
Fig. 4.3: Interband cyclotron resonance in monolayer graphene at $B = 8$ T. (a) Typical normalized transmission, at $\nu = 0$, showing six resonances. The labels identify the same transitions in all plots. (b) Colormap of resonances vs $\nu$. The color scale is expanded above 290 meV to better show lower intensity peaks. (c) Schematic of Landau levels, and allowed interband transitions (arrows in color) at $\nu = +2$ [84].
while not shown, T\textsubscript{5} and T\textsubscript{6} show the same overall behavior as T\textsubscript{2} through T\textsubscript{4}. Furthermore, while the higher energy resonances become difficult to see on the same scale as lower energy peaks, fits to the data remain robust. Meanwhile T\textsubscript{1} exhibits a more elaborate behavior, with a maximum in the resonance energy occurring at \( \nu = 0 \), and the appearance of a splitting centered near \( \nu = +2.5 \) and \( \nu = -2 \). Except for the splitting, the behavior of T\textsubscript{1} is consistent with the findings of Ref. [20] which were interpreted as an interaction-induced gap at half-filling of the \( N = 0 \) Landau level [87].

Figure (4.5) shows the detailed behavior of the effective Fermi velocity for \( B = 5, 8, \) and 11 T, directly extracted from the CR energies by\textsuperscript{7}

\[
v_{\text{F}}^{\text{eff.}} = \frac{\Delta E_{MN}^{\text{meas}}}{\sqrt{2e\hbar B}} \left( \sqrt{|M|} + \sqrt{|N|} \right), \quad (4.4)
\]

as a function of \( \nu \). Focusing on the 8 T data, several intriguing features appear: (1) \( v_{\text{F}}^{\text{eff.}} \) shows a non-monotonic dependence on filling factor; (2) the behavior of \( v_{\text{F}}^{\text{eff.}} \) falls in two groups: T\textsubscript{3} through T\textsubscript{6} show a generally similar response as \( \nu \) changes, while T\textsubscript{1} and T\textsubscript{2} each display a unique dependence on \( \nu \); (3) the peak values of \( v_{\text{F}}^{\text{eff.}} \) also vary non-monotonically with the transition number; and (4) \( v_{\text{F}}^{\text{eff.}} \) is also found to decrease monotonically with increasing magnetic field.

We address each of these in turn: (1) At constant \( B \), the single-particle cyclotron energies \( v_{\text{c}} \sqrt{2e\hbar B N} \) and bare Coulomb interaction \( e^2/\ell_B \) are fixed, therefore the variations in \( v_{\text{F}}^{\text{eff.}} \) must arise from changes in the many-particle interactions experienced by the excited electron and the hole it leaves behind. The density of states at the Fermi level and therefore the effects of many-particle screening will change with filling factor, so the exciton’s energy will change as well. (2) The variation of \( v_{\text{F}}^{\text{eff.}} \) with filling factor is different for each transition, but the data in Fig. (4.5) can be divided into two groups: transitions T\textsubscript{3} through T\textsubscript{6} show similar

\textsuperscript{7} This approach differs slightly from other works, where the Fermi velocity is studied in ratio to that of the \( 0 \rightarrow 1 \) transition: \( v_{\text{c}}(T_j)/v_{\text{c}}(T_1) \) [43,198]. Since T\textsubscript{1} varies in a distinct fashion from the other transitions, indicating different physics is at work, the meaning of that ratio is not obvious and so we plot \( v_{\text{F}}^{\text{eff.}} \) for each transition independently.
Fig. 4.4: Filling factor dependence of the first four interband transitions. For $|\nu|>2$—when the Fermi level lies outside the lowest Landau level—all transitions are seen to broaden and undergo shifts in the central energy. Nonetheless good fits are readily made [?]. Note the larger vertical scale for $T_1$.
ν-dependence suggesting a common origin, with (i) a peak or plateau for |ν| < 2 that is centered on ν = 0; and (ii) a sharp 4-5% decrease for |ν| > 2. In contrast, transitions T1 and T2 both show a maximum at ν = 0 but otherwise show distinct behavior. The existence of these groupings is likely due to the fact that, for almost all filling factors studied, the Fermi level lies inside the N = 0 or ±1 LLs which participate in the T1 and T2 transitions. If any of these three lowest levels shift or split when partially filled, the CR transition energies should reflect this. For instance, the sharp minima in T2 at |ν| = 4 (half-filling of N = ±1) are reminiscent of exchange-enhanced gaps seen at half-filling in high-mobility GaAs 2D systems [199]. Similarly, a sharp maximum in T1 is seen for ν = 0 (half-filling of N = 0) [20]. On the other hand, the T3–T6 transitions involve electrons excited over the Fermi sea from completely full to completely empty LLs. For these, the enhanced values of $v^\text{eff.}_F$ arise purely from many-body interactions with states at the Fermi energy in a distant, partially filled LL [43,182].

These many-particle contributions to T3–T6 are fairly constant until |ν| > 2, after which $v^\text{eff.}_F$ abruptly decreases. Similar behavior at 11 T emphasizes this decrease is associated with changes in the LL filling rather than a particular density [?]. At ν = ±2, the Fermi level shifts from N = 0 to N = ±1, so the decrease may be due to enhanced screening from the presence of a partially-filled level lying close to levels involved in the T3–T6 transitions. On the other hand, the unique valley-polarized nature of the N = 0 level may play a role [8].

We note the values of $v^\text{eff.}_F$ at ν = ±10, where the N = +2(−2) levels are filled (empty), largely agree with the ν = ±6 values, implying the decrease is in fact linked to filling of the N = 0 level.

The dashed lines in Fig. 4.5 are calculated using a many-particle theory of graphene magnetoexcitons [182]. As shown, these predictions roughly capture the decrease of $v^\text{eff.}_F$ with increasing |ν|, but are almost identical for T2 and T3 and do not reflect the observed plateaus about ν = 0 or the |ν| = 4 minima in T2.

(3) The several transitions recorded probe the graphene dispersion at energies up to ±300
Fig. 4.5: Effective Fermi velocity at $B = 5, 8, \text{ and } 11 \text{ T}$ for all six transitions as a function of filling factor, $\nu = nh/eB$. The black dashed lines in $T_2$ and $T_3$ are calculated from the theory of Ref. [182].

Fig. 4.6: (Left) $v_F^{\text{eff}}$ values for $\nu = 0$, plotted vs. transition number for all three magnetic field values. The symbol colors match those used in figures 4.3 and 4.5. (Right) Illustrations of the zero magnetic field Dirac cone band structures implied by the observed dependence of $v_F^{\text{eff}}$ on transition number. Plots of $v_F^{\text{eff}}$ for $|\nu| > 0$ are shown in fig. 4.7.
Fig. 4.7: Plots of \( v_{\text{eff}} \) vs. transition number for several key integer values \( |\nu| > 0 \). See fig. 4.6 for more details. Here \( B = 5, 8, \) and 11 T data are shown in red, green, and blue, respectively.

meV away from the Fermi level. In detail the dependence of \( v_{\text{eff}} \) on transition number evolves with \( \nu \), and we have re-plotted these data to make this plain in fig. 4.6a and fig. 4.7. Indeed, at \( \nu = 0 \) the peak value of \( v_{\text{eff}} \) occurs for \( T_3 \), but as \( \nu \) increases the peak shifts to lower transition numbers. We represent this schematically in fig. 4.6b, sketching a “zero-field” Dirac cone with slope \( v_{\text{eff}} \). This procedure results in an intriguing departure from the familiar linear dispersion. In contrast, the renormalized dispersion found by scanning tunneling spectroscopy remains linear over a comparable energy range about the Fermi level, with higher \( v_{\text{eff}} \) for lower densities [200]. These divergent results can perhaps be reconciled by noting that in Ref. [200], electrons tunneled into a single LL of an interacting system, while here we are sensitive to the physics of an electron-hole pair with each particle in a separate LL having different index (and energy), noting that many-particle interactions differ for an exciton vs. single electrons [176].
(4) The $v_{\text{eff}}/F$ found at $B = 5$ and 11 T are qualitatively similar to those of fig. 4.5, except the overall magnitude decreases with increasing $B$. This running of the velocity enhancement was predicted [43] and also recently observed in graphene magneto-Raman experiments [184].

4.4 Dirac Mass

Returning to fig. 4.4, $T_1$ exhibits a $\sim 2.5$ meV splitting near $\nu = \pm 2$. We speculate the $N = 0$ level has become gapped due to coupling of the graphene and hBN lattices, which breaks the sublattice symmetry and generates a so-called Dirac mass [84, 197, 201] (Phys-RevLett.98.157402*). The resulting splittings around $\nu = \pm 2$ are due to transitions from (into) the $N = 0$ LL sub-bands on either side of the gap, which we illustrate schematically in fig. 4.8.

Separately, the blueshift of $T_1$ at $\nu = 0$ indicates the presence of an interaction-induced gap at half-filling of the $N = 0$ level, which has been observed even in the absence of a boron nitride substrate [20]. Analyzed along the lines of Ref. [20], we infer this interaction-induced gap to be $\sim 6$ meV.

A gap at zero energy will shift the LL energies according to [198]

$$E_N = \pm \frac{\Delta}{2} \delta_{N,0} + \text{sign}(N) \sqrt{2\epsilon_N^2 B |N| + (\Delta/2)^2},$$

(4.5)

where $\Delta$ is the gap energy. In the case of finite $\Delta$ the two valley-split $N = 0$ LLs are pushed to energies $\pm \Delta/2$, with the effects on LLs $|N| > 0$ being substantially smaller. Even for the larger 6 meV interaction-induced gap, this implies the energy of the $N = 1$ LL is shifted by less than 0.05%. Since the variations we observe due to many body interactions (see fig. 4.5) are of order a few %, far larger than the gap-induced shifts of the higher LL energies, we do not account for shifts due to $\Delta$ in our analysis.

Lattice couplings between graphene and hBN also generate additional Dirac peaks on both sides of the main peak, which are not observed in transport measurements of this device.
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Fig. 4.8: A Landau level diagram illustrating the way a fixed valley splitting of the $N=0$ LL might appear at certain filling factors. Because the valley degree of freedom is preserved in cyclotron resonance, only the energetically degenerate transitions $|−1, \uparrow\rangle \rightarrow |0, \uparrow\rangle$ and $|0, \downarrow\rangle \rightarrow |+1, \downarrow\rangle$ are allowed at $\nu = 0$. At $\nu = 2$ the transitions $|0, \uparrow\rangle \rightarrow |+1, \uparrow\rangle$ and $|0, \downarrow\rangle \rightarrow |+1, \downarrow\rangle$ are observed, with an energy difference dominated by the valley splitting of the $N=0$ LL. Similarly, at $\nu = −2$ the transitions $|−1, \uparrow\rangle \rightarrow |0, \uparrow\rangle$ and $|−1, \downarrow\rangle \rightarrow |0, \downarrow\rangle$ show the same splitting observed at $\nu = 2$.

(see fig.4.2); however, as previously noted, these peaks shift to higher density for increasing graphene-hBN rotational misalignment, and thus may lie outside the $V_g$ range of this device. More work on samples with a range of relative graphene-hBN rotations is needed to verify this, but if correct, our method can be used to accurately determine the gap size and its dependence on rotation or other symmetry-breaking mechanisms.

4.5 Cyclotron Linewidths and Lineshapes

In addition to the peak energy, cyclotron resonances contain a wealth of additional information. The integrated intensity of a resonance is directly proportional to the number of charge carriers participating in the associated transition, while the linewidth is a sensitive probe of the various scattering processes that carriers may undergo in the host system [163].
The half width at half maximum (HWHM) intensity for all the observed transitions as a function of LL filling factor at $B = 5$, 8, and 11 T. The widths observed in our hBN encapsulated device are an order of magnitude narrower than those reported in previous studies of unencapsulated graphene samples on SiO$_2$. Some transitions appear to display oscillatory behavior as the filling factor is varied, similar to observations made in CR of GaAs samples [171–174]. At all magnetic fields, the $T_1$ transition is significantly narrower than the higher transitions, indicating that different scattering mechanisms may effect excitations involving the zeroth LL. The splitting of $T_1$ in the vicinity of $\nu = \pm 2$ leads to the observation of significant linewidth narrowing at these values of $\nu$.

The linewidth can be related to the cyclotron lifetime $\tau_{CR}$ by

$$\Gamma = \frac{\hbar}{\tau_{CR}},$$

where $\Gamma = \text{HWHM}$ is the half width of the resonance at half its maximum intensity.$^8$

It is interesting to compare the observed cyclotron lifetimes to the mobility and quantum scattering times found in this device. From the field effect mobility we find $\tau_\mu \approx 3000$ fs, while measurements of Shubnikov-de Haas oscillations at carrier densities of -1.89, -1.22, 1.08, and $1.94 \times 10^{12}$ cm$^{-2}$ yielded corresponding quantum scattering times of $\tau_q = 139$, 76, 66, and 81 fs. From the observed linewidths we find cyclotron lifetimes of $\sim 50$ fs for transitions $T_2$ through $T_6$ at $B = 5$, 8, and 11 T. The lifetime of the $T_1$ transition is considerably longer at $\sim 150$ fs, indicating it is subject to broadening mechanisms in a way that is distinct from the higher transitions.

As noted in Ref. [23], several mechanisms contribute to the broadening including Coulomb...

$^8$ The use of the half-width, rather than the full-width commonly used to define lifetimes in high energy and nuclear physics, comes from the definition of the Lorentzian lineshape found in CR. [163]
interactions, scattering from optical and acoustic phonons, impurity scattering, and radiative
decay. Radiative broadening, which is due to spontaneous emission and exists in the absence
of all scattering mechanisms, is particularly interesting as it sets the fundamental minimum
limit on the cyclotron linewidth in any system. In graphene the linewidth due purely to
radiative decay is found to be less than $\sim 10^{-1}\mu\text{eV}$, corresponding to a theoretical maximum
cyclotron lifetime of order 10 ns [23]. As more samples are studied in time, a comparison
of the observed linewidths may allow for detailed conclusions to be made about the relative
importance of the different broadening mechanisms.

4.5.1 Optical Modeling of Transmission Through Layered Thin
Film Media

The varying thicknesses and dielectric functions of our Van der Waals heterostructure device
and its host substrate constitute a complicated thin-film optical system. Transmission of
light through such a structure results in frequency dependent distortions of the Lorentzian
lineshapes of the CR absorptions in the graphene layer, preventing simple extraction of the
central energies, linewidths, and oscillator strengths, particularly in the vicinity of substrate
absorptions.

The composition of our five layer heterostructure device is as follows: a 620 $\mu$m thick, lightly
doped silicon wafer; a 300 nm thermal oxide on the wafer surface; a thin crystal of hexagonal
boron nitride (hBN) estimated to be 15 nm thick; a monolayer of graphene; and a top
layer of hBN estimated to be 30 nm thick. Due to multiple reflections at the interfaces
of different layers and the presence of multiple absorbing materials, transmission through
such a stratified system is an involved function of the layer thicknesses and complex indices
of refraction. The observed resonances appear asymmetric, with the spectral weight and
central peak shifted from their actual values. To account for these distortions we performed
a nonlinear fitting procedure using a model for transmission through a multilayer thin-film
system [20].
Fig. 4.10: Normalized infrared magneto-spectroscopy data plotted as $S(\nu)/S(\nu = 22)$, showing representative fits to several cyclotron resonance transitions in traces having (a) high, (b) moderate, and (c) low signal-to-noise ratios. For the fits in (c), error bars for the energies of the three highest energy resonances are taken to be the width of the transitions.

Constructing the multilayer reflection model requires accurate determination of the complex index of refraction $\tilde{n} = n + ik$ for each layer. For the Si/SiO$_2$ substrate this was accomplished via careful measurements of a piece of the same silicon wafer used in fabricating the device, both before and after removing the oxide layer. Standard fitting procedures employing the Reffit software package were then used to construct a model of the Si and SiO$_2$ complex indices of refraction [?](Review of Scientific Instruments 76, 083108 (2005)*). For hexagonal boron nitride, values of $n = 1.8$ and $k = 0$ were assumed for all frequencies.

The dielectric function of the graphene monolayer was modeled as a set of Drude-Lorentz oscillators corresponding to distinct Landau level transitions. Each Lorentzian is character-
ized by three parameters: a center frequency, a plasma frequency, and a linewidth. These parameters were allowed to vary simultaneously until good agreement was achieved between the model and data, as determined by a quasi-Newton algorithm and checked by eye (see fig. 4.10). In most cases, error bars for center frequencies are taken as the standard errors computed by the fitting procedure. For certain data traces where low signal-to-noise ratios prevented unambiguous determination of the center frequency (see fig. 4.10c), error bars are taken as the width of the resonance.

4.6 Conclusions and Future Outlook

In conclusion, we have performed infrared magneto-spectroscopy on a high quality encapsulated monolayer graphene device. Several interband transitions provide direct evidence of contributions from many-particle physics via a renormalized Fermi velocity that depends on the Landau level filling factor and the magnetic field strength. Furthermore, the good quality of this sample leads to very narrow resonances with halfwidths approaching 1 meV for the sharpest transitions, leading to high-precision measurements that are likely to enable future spectroscopy of broken symmetry states [26], the fractal Landau levels underlying Hofstadter’s butterfly [29–31], or even the fractional quantum Hall effect at lower temperatures [27]. This work demonstrates that infrared magneto-spectroscopy can provide a novel tool for the study of correlated electron physics in graphene.
Chapter 5

Cyclotron Resonance in Dual-Gated Bilayer Graphene

In this chapter we present the first measurements of cyclotron resonance (CR) in dual-gated bilayer graphene (BLG). As with our work on monolayer graphene, we improve on prior results by encapsulating our sample in hexagonal boron nitride, while the inclusion top and bottom gates allows for independent electrostatic control of both the bandgap and charge carrier density.

At the heart of this work is the exotic zero energy Landau level (LL) in BLG: due to a combination of spin, valley, and an additional orbital degeneracy the zero energy LL in BLG is eight-fold degenerate, giving rise to a so-called quantum Hall octet [120]. This massive degeneracy, and the couplings of external fields to the involved degrees of freedom, make the quantum Hall octet in BLG a fertile environment to explore the effects of symmetry breaking in the quantum Hall regime. Furthermore, Kohn's theorem [179], which prevents the observation of many-body interactions in translationally invariant 2DEGs with parabolic dispersions, and fails in monolayer graphene [18,20–22,43,44,182,184,185,187–192,194,198,202], also fails in bilayer graphene due to the chiral nature of its charge carriers [41,45]. As a result, many-body interactions are expected to impact the cyclotron resonance in BLG [39,42–45,127].
As of 2019 only one investigation of cyclotron resonance in exfoliated\(^1\) BLG, from 2008, has been reported in the literature to the authors’ knowledge \([124]\). In this work a back-gated, low-mobility BLG on silicon dioxide device was utilized and the four lowest energy intraband transitions were studied at five magnetic fields from 10 to 18 T. As expected, a transition from linear-in-B to \(\sqrt{B}\) behavior was observed with increasing LL index, reflecting the unique hyperbolic dispersion found in BLG, which is parabolic at low energies and linear at higher energies. Additionally, the authors were able to analyze the data in such a way that allowed for a determination of the density of states (DOS), which showed satisfactory agreement with the expected bilayer DOS, \(g(E) = (2E + \gamma_1)/(\pi\hbar^2v_F^2)\). However, several anomalies were observed: a large spread in \(v_F\) and \(\gamma_1\) values were required to achieve good fits to the data, and the observed transition from linear-in-B to \(\sqrt{B}\) behavior occurred more suddenly and at much lower energies than predicted from the simplest single-particle, nearest-neighbor tight-binding model. Possible explanations for these deviations could include neglected band parameters in the tight-binding model, a finite electric displacement field that varied with filling factor and was surely present, or many-body interaction effects. These anomalies, the unexplored effects of electric displacement field controlled independently of carrier density, and the many exciting discoveries made in quantum Hall studies of BLG in the last decade all leave considerable room for further experimental exploration of cyclotron resonance in BLG.

In this work we perform far-infrared magneto-spectroscopy at \(B = 13\) T for electric displacement fields \(D < |700|\) mV/nm and Landau level filling factors \(|\nu| \leq 6\), using electronic transport to guide these measurements.\(^2\) The observed transport behavior matches that reported for a number of high quality dual-gated bilayer graphene devices at charge neutrality: in a quantizing magnetic field insulating behavior is observed at small and large electric dis-

\[^1\] We note that CR measurements of epitaxial BLG on SiC have also been reported \([203]\).

\[^2\] As noted in previous chapters, the Landau level filling factor is

\[
\nu = \frac{\hbar n}{eB}
\]

where \(n\) is the two-dimensional electronic carrier density and \(B\) is the applied magnetic field.
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placement fields, separated by a region of metallic conductance [107, 108, 137, 139, 141, 204]. The insulating state near $D \approx 0$ has been attributed to a canted antiferromagnetic (CAF) quantum Hall insulator, while the high-$D$ insulating behavior has been ascribed to the trivial layer polarized (LAP) state [108, 135, 137, 139, 204–206]. Cyclotron resonance is measured at $\nu = 0$ and we observe three distinct regimes as a function of $D$, which notably do not coincide with the three distinct regions observed in transport. At $\nu = 4$, where interaction effects are minimized, we present a novel spectroscopic measurement of the optical band gap in bilayer graphene as a function of the applied electric displacement field. These results are in excellent agreement with previous optical determinations of the band gap made at significantly higher energies. Finally, CR at $D = 0$ is measured as a function of $\nu$, revealing distinct electron-hole asymmetry.

5.1 Landau Levels and Cyclotron Resonance in Bilayer Graphene

Bilayer graphene hosts an unusual hyperbolic band structure which gives rise to a unique Landau level spectrum in the presence of a magnetic field [95, 120, 127]. As discussed in Ch. 2, the commonly encountered approximation for the LL energies in bilayer graphene

$$E_N = \text{sign}(N)\hbar \omega_c \sqrt{|N|(|N| - 1)}, \quad N = 0, 1, \pm 2, \pm 3, ...$$

(5.2)

is insufficient for describing the effects of an interlayer potential difference $\Delta$, such as arises in the presence of an electric field applied perpendicular to the BLG plane. We follow Ref. [111] in defining

$$\Theta = \left( \frac{\hbar e B}{\gamma_1 m^*} \right)^2 = \frac{2\hbar e B v_F^2}{\gamma_1^2},$$

(5.3)
which is the cyclotron energy normalized by the interlayer coupling strength $\gamma_1$, and

$$\tilde{\delta} = \delta_{\text{dim}} + \frac{2 \gamma_1 \gamma_4}{\gamma_0}, \quad (5.4)$$

which sets the energy scale of electron-hole symmetry breaking effects.\footnote{Using $\gamma_0 = 3000$ meV, $\gamma_1 = 400$ meV, $\gamma_4 = 150$ meV, and $\delta_{\text{dim}} = 18$ meV, we find $\tilde{\delta} = 58$ meV.} Then, neglecting only the hopping parameter $\gamma_3$ and the intralayer asymmetry $\delta_{AB}$, the LL energies in BLG are

$$E_{N=0}^{\lambda=\pm} = \lambda \frac{\Delta}{2} \quad (5.5a)$$

$$E_{N=1}^{\lambda=\pm} = \Theta \tilde{\delta} + \lambda (1 + 2 \Theta) \frac{\Delta}{2} \quad (5.5b)$$

$$E_{|N|\geq2}^{\lambda=\pm} = \left( \frac{1}{2} |N| + \frac{1}{2} \right) \Theta \tilde{\delta} - \lambda \Theta \frac{\Delta}{2} + s_N \sqrt{\left[ \frac{1}{2} (2 |N| + 1) \Theta \frac{\Delta}{2} - \lambda \Theta \frac{\Delta}{2} \right]^2 + |N|(|N| + 1) \Theta^2 \gamma_1^2} \quad (5.5c)$$

where $\Delta$ is the interlayer potential difference, $\lambda = +1(-1)$ indexes the $K^+(K^-)$ valley, and electron and hole LLs are indexed by $s_N = \text{sign}(N)$.

As shown in figure 5.1, the LL spectrum given by eq. (5.5) displays a wealth of interesting behaviors not captured by eq. (5.2). We consider three such effects relevant to our observations: (i) In the presence of an interlayer potential difference $\Delta$, the valleys within each LL are no longer degenerate in energy. This can be seen as a consequence of the identification of valley and layer in BLG [109, 120, 142]. The simple linear dependence of this splitting on $\Delta$ in the case of $N = 0$ and 1 comes from the fact that the wavefunctions in opposite valleys for those LLs are polarized on opposing layers. We refer the reader to the discussion surrounding eq. (2.75 - 2.78) for more detail. (ii) At certain values of $\Delta$, states of opposite valley in adjacent LLs are observed to cross. As shown in Ref. [111], this occurs when the phase acquired by charge carriers undergoing cyclotron motion, which in BLG is the sum
Fig. 5.1: BLG LL energies vs D. See eq. (5.5) and the surrounding discussion for details.

of the Berry phase and an additional phase related to the pseudospin Zeeman coupling, is equal to $\pi$. This is related to the development of a sombrero shape in the zero field band structure under application of a perpendicular electric field.\(^4\) (iii) Notably, the $N = 0$ and $N = 1$ LLs are no longer degenerate at zero energy for all $B$; they are weakly split even at $\Delta = 0$ by an amount $\tilde{\delta}$, which is of order $\sim 5.7$ meV at $B = 13$ T.

It is worth emphasizing that the LL spectrum given by eq. (5.5) accounts for strictly single particle effects. Departures from this behavior are predicted to occur in the presence of Coulomb and exchange interactions and many competing ground states have been predicted

\(^4\) LL crossing are also directly related to quantum Hall ferromagnetic ordering [111, 207–209].
[40, 106, 125, 128–130, 132, 133, 210, 211]. Notably, observations of Hall quantization at all integer filling factors $\nu < 4$ lend strong experimental support to the presence of exchange-driven quantum Hall ferromagnetism in BLG [108, 134–136, 140, 212, 213].

5.2 Dual-Gated, Encapsulated Bilayer Graphene Device

5.2.1 Device Architecture

The sample investigated in the present work has a top gated area of approximately 550 $\mu$m$^2$. An image of the device under 100x magnification is shown in figure 5.2d. Our exfoliated graphene bilayer was encapsulated in hexagonal boron nitride (hBN) using standard dry transfer techniques [100, 195]. Electrical contact was made using the widely employed one-dimensional edge contact method [195]. In the final step of device fabrication, a mask was defined by way of electron beam lithography and 20 nm of chromium was sputtered to form the top gate. The resulting metal thin film is thick enough to allow for application of a uniform electric field, yet thin enough to enable transmission of infrared light though the multilayer device. In total our device consists of seven layers, which are illustrated in figure 5.2b. From top to bottom they are 20 nm of Cr, 40 nm of hBN, bilayer graphene, 30 nm of hBN, 293 nm of thermal silicon dioxide ($\text{SiO}_2$), 380 $\mu$m of lightly doped silicon, and another 293 nm layer of $\text{SiO}_2$. Roughly half of the BLG crystal that composes our device is covered by the top gate (see discussion in fig. 5.2). The effects of this un-gated region on transport measurements are discussed in the relevant sections below.

The finished device was aligned over a $\sim 1$ mm copper foil aperture and wire bonded. Immediately prior to loading the sample into the cryostat, a $\sim 90 \mu$m aluminum foil aperture was placed over the device. The results are shown in fig. 5.2c.
Fig. 5.2: (a) An illustration of the Van der Waals heterostructure device employed in this work. The SiO$_2$ layer on the bottom of the Si wafer is not pictured and the layer thicknesses are not to scale. Voltages applied to the top and bottom gates allow for independent control of the electric displacement field and carrier density in the sample. In the series of images one can see (b) an example of the silicon wafer with pre-fabricated contacts on which our dual-gated bilayer device was constructed, (c) the 90 $\mu$m aluminum foil aperture that was placed over the device, and (d) an optical micrograph of the device taken at 100x magnification. The transparent 20 nm Cr top gate, with an area of $\sim$550 $\mu$m, is visible in the latter. While not visible, the bilayer graphene crystal extends below the lower boundary of the top gate. The area of this un-top-gated bilayer is approximately equal to that of the top-gated region.

5.2.2 Dual-Gating of a Graphene Bilayer

The carrier density and electric displacement field in the sample each depend on the voltages applied to both the top and bottom gate. The carrier density is given by

$$n = \alpha_b(\Delta V_b + \beta \Delta V_t),$$

(5.6)
while the electric displacement field is

\[
D = \frac{(D_t - D_b)}{2} = \left(\frac{\epsilon_t}{d_t} \Delta V_t - \frac{\epsilon_b}{d_b} \Delta V_b\right)/2, \tag{5.7}
\]

where \(D\) is defined such that a positive displacement field points up, in the direction of the top gate. The relative gate voltage on the \(i^{th}\) gate is

\[
\Delta V_i = V_i - V_i^0, \tag{5.8}
\]

where \(V_i\) is the applied voltage and \(V_i^0\) and \(V_b^0\) are the offset voltages required to bring the sample to \(n = D = 0\). As with single-gated samples, \(\alpha_b\) is the so-called “gate efficiency” of the bottom gate, which relates the applied gate voltage to the charge carrier density induced in the sample; the top gate efficiency \(\alpha_t\) is defined in the same way. These gate efficiencies, which are the capacitances per unit area per unit charge of the graphene-gate capacitor, are set by the thicknesses and zero-frequency relative permittivities of the dielectric materials above and below the graphene and have units of \(V^{-1} \text{ cm}^{-2}\). The value of the offset voltages were found to be very near to zero volts, \((V_b^0, V_t^0) = (-0.104 \text{ V}, -0.082 \text{ V})\). This means the level of charged impurities trapped between the BLG and either gate is low, which is generally a good indicator of high electronic quality in such a device.

The ratio of the top and bottom gate efficiencies,

\[
\beta = \frac{d_b \epsilon_t}{d_t \epsilon_b} = \frac{\alpha_t}{\alpha_b} = -1/m, \tag{5.9}
\]

is equal to the negative inverse of the slope \(m\) of the high resistance \(n = 0\) ridge in \(V_b-V_t\) space, which can be seen in figures (5.3a) and (5.4). The zero-frequency relative permittivities of hBN and SiO\(_2\) are \(\epsilon_{\text{hBN}} = 3.76\) [214] and \(\epsilon_{\text{SiO}_2} = 3.7 - 3.9\). We take \(\epsilon_{\text{hBN}} \approx \epsilon_{\text{SiO}_2} \approx 3.8\), allowing us to make the simplification of treating the materials between the bilayer and silicon bottom gate as a single, uniform dielectric. Thus, we find \(\epsilon_t = \epsilon_b = 3.8 \epsilon_0\), where \(\epsilon_0\) is
the permittivity of free space. The thickness of the top and bottom hBN layers are estimated to be $d_{hBN}^{top} \approx 40$ nm and $d_{hBN}^{bottom} \approx 30$ nm. Using ellipsometry, the thickness of the oxide on our silicon wafers was measured to be $d_{SiO_2} = 293$ nm. In total we have $d_t = d_{hBN}^{top} \approx 40$ nm and $d_b = d_{hBN}^{bottom} + d_{SiO_2} \approx 323$ nm. The ratio $d_b \epsilon_t / d_t \epsilon_b \approx d_b / d_t = 8.08$ agrees well with the value $-1/m = 7.63$ taken from transport measurements. Using high field quantum Hall plateaus for calibration, a bottom gate efficiency of $\alpha_b = 5.9 \times 10^{10}$ cm$^{-2}$ V$^{-1}$ was found, which in turn implies $\alpha_t = 4.6 \times 10^{11}$ cm$^{-2}$ V$^{-1}$.

The origin of $n$-$D$ space, the point $(n, D) = (0, 0)$, coincides with the point $(V_b, V_t) = (V_b^0, V_t^0)$ in $V_b$-$V_t$ space. The axes $n$-$D$ are rotated relative to the $V_b$-$V_t$ axes by an angle determined by the relative efficiencies of the top and bottom gates. While we operate in $V_b$-$V_t$ space as a matter of practicality, clearly $n$-$D$ space is the more physically relevant of the two. In the presence of a quantizing magnetic field we will be more interested in the Landau level filling factor, $\nu = nh/eB$.

Throughout we divide the applied displacement field by the permittivity of free space $\epsilon_0$ so that we may work with the more familiar units of electric field. This field is related to an important energy scale in BLG

$$\Delta \approx \frac{eDd}{\epsilon_0},$$

which is the interlayer energy difference, where $e$ is the electron charge and $d = 0.335$ nm is the interlayer separation. It is important to note that self-screening within the bilayer acts to reduce $\Delta$ from the value give by eq. (5.10) [115,122], however the exact magnitude of this screening depends sensitively on interactions in the quantum Hall regime, which are not known a priori [135]. As such, we report all quantities in terms of the applied electric field and use eq. (5.10) sparingly.
5.2.3 Electronic Transport

The two-terminal resistance, $R_{2pt}$, and Hall resistance, $R_{xy}$, were measured at $B = 0$ and 13 T as a function of top and bottom gate voltages. The zero field transport seen in figure 5.3a shows the response expected in a dual-gated BLG device: at zero carrier density the opening of a band gap for $|D| > 0$ produces insulating behavior, which can be seen as a diagonal high-resistance ridge in fig. 5.3a. The zero field insulating behavior reported at $n = D = 0$ in some BLG devices was not seen, though the very slow sweep rates and measurement optimization reportedly required were not reproduced in our measurements [108,215]. From the line cut at $D = 0$ in fig.5.3b, we extract a field-effect mobility $\mu_{FE} = (1/e)(d\sigma/dn)$ of 20 000 cm$^2$/V s.

The presence of the un-top-gated bilayer can be seen in fig.5.3a as a faint vertical ridge at $V_{BG} \approx 0$ V, in agreement with observations of transport in split-gate BLG devices [113,216]. As can be see in fig.5.3c, at large displacement fields and zero carrier density we observe a non-monotonic variation of the two-point resistance. We attribute this apparently non-ideal response to the un-top-gated bilayer region at the edge of our sample: as the back gate voltage is increased this region becomes heavily doped, shorting the highly resistive dual-gated region.

The two-point magnetoresistance and the Hall resistance at $B = 13$ T as a function of $V_b$ and $V_t$ are shown in figure 5.4. Our device exhibits the same behavior now widely observed in dual-gated bilayer graphene at high field: at charge neutrality two distinct insulating phases are observed in the magnetoresistance, separated by a region of metallic conductance $\sim e^2/h$. This metallic phase boundary has been reported to disperse linearly in $B$ at 7 to 18 mV/nm T for $5 \lesssim B \lesssim 12 - 15$ T [107,108,137–139,141]. At 13 T we find these metallic regions centered at 241 mV/nm (-216 mV/nm) corresponding to a dispersion of 18.5 mV/nm T (16.6 mV/nm T) for $D > 0$ ($D < 0$). The two-point conductance in these regions is $G \sim 0.7h/e^2$. Incredibly, slow gate sweeps in the vicinity of $n = D = 0$ reveal a sharp peak in the resistance in excess of 500 kΩ, dramatically higher than the $\sim 6k\Omega$. 

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Fig. 5.3: (a) Dual-gated two-point resistance measurement of bilayer graphene at $B = 0$ T and a sample temperature of 4 K. The axes have been shifted in the direction of $n$ so as not to obscure the high-resistance ridge. The faint vertical feature seen at $V_b = 0$ is due to the presence of an adjacent, un-gated region of BLG. Line cuts at (b) $D = 0$ and (c) $n = 0$.

observed at zero field.

The Hall resistance, shown in fig.5.4b, displays four rough plateaus on either side of the $\nu = 0$ ridge, suggesting complete symmetry breaking of the $N = 0, 1$ LLs [134]. Line cuts reveal $\sigma_{xy}$ approaching the expected values of $\nu e^2/h$ for $\nu = 0, \pm 1, \pm 2, \pm 3,$ and $\pm 4$ in these regions, but the quantization is incomplete. We attribute this to a combination of elevated sample temperature ($T \approx 4$ K) and mixing with edge states from the un-top-gated region of our device. We associate the strong vertical features seen in fig.5.4a with Hall quantization in the un-top-gated region. Notably, in performing spectroscopy, the background spectra required for normalization were obtained by heavily doping the sample using only the top gate; back gate voltages were held constant for the collection of each spectra-background pair. Doing so removes all signatures of the un-top-gated bilayer from our spectroscopic data.
5.3 Cyclotron Resonance at Constant Filling Factors

With the ability to dual-gate our bilayer sample we can explore the evolution of cyclotron resonance at a single filling factor while tuning the layer symmetry-breaking electric displacement field. A number of potential ground states have been predicted at $\nu = 0$ in bilayer graphene [125, 130, 132, 133], with several CR-specific predictions for optical probes of the ground state order in BLG having been put forward to date [40, 128, 129].

5.3.1 Cyclotron Resonance at $\nu = 0$

We begin with our observations of cyclotron resonance studied as a function of $D$ at $\nu = 0$, shown in fig. 5.5. The observed resonance behavior can be roughly divided into three distinct regions, which we label the (i) low-, (ii) intermediate-, and (iii) high-$D$ regimes. We focus
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on $D > 0$, as shown in fig. 5.5, and discuss each regime in turn.$^5$

(i) In the low-$D$ region, observed between 0 and 12 mV/nm, we observe two peaks separated by a small splitting. We label the higher energy peak, C, and the lower peak, B (see fig. 5.6). The peak splitting varies from 2.3 meV at 0 mV/nm, to less than 1.7 meV at 12 mV/nm, while the average energy of the two resonances remains centered around 67.5 meV. This region is observed exclusively inside the high resistance region seen in transport near $n = D = 0$.

(ii) In the intermediate-$D$ regime observed between 27 and 110 mV/nm, a third, lower energy peak gradually emerges, which we label A. Peak C is observed to blueshift monotonically, while peaks A and B redshift monotonically. The splitting between peaks A and B is relatively constant at $\sim 3.4$ meV, while the splitting between peaks B and C grows from 2.2 to 7.2 with increasing $D$. Above 99 mV/nm all three peaks appear to undergo strong broadening. Within region (ii) peak B redshifts from $\sim 67.2$ meV to 64.6 meV.

At 122 mV/nm peaks A and C seemingly disappear and a single peak with an unusually long low-energy tail is observed. The energy of the single remaining peak coincides with the energy of peak B at 110 mV/nm, but the spectral weight and lineshape character differs.

(iii) A high-$D$ region is observed starting from 134 mV/nm and persisting until the resonances are blueshifted outside of the observable spectral window at 529 mV/nm. We label these peaks A’ and B’, as their exact relation to the resonances observed in regions (i) and (ii) is unclear. The splitting between these peaks reaches a maximum value of 3.2 meV at 134 mV/nm, gradually decreases until dropping below the measurement resolution of 1 meV at 350 mV/nm, and reappears at less than 2 meV for higher values of $D$. The average resonance energy undergoes a strong linear blueshift from $\sim 64$ meV to $\sim 84$ meV in this region.

We find that much of the detailed behavior observed at $\nu = 0$ can be explained simply by accounting for some of the often neglected small band parameters present in BLG. Using eqs.(5.5) we plot the dispersion of both valleys in the $N = -2, 1, \text{ and } +2$ LLs for small

$^5$ We note that, while only five spectra were collected for $D < 0$, reaching a maximum of -70 mV/nm, their peak energies, resonance count, linewidths, and lineshape character agree strongly with their $D > 0$ counterparts, suggesting some degree of symmetry.

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Fig. 5.5: (a) Evolution of the observed cyclotron transitions at \( \nu = 0 \) as a function of \( D \). Note that spectra are shown with equal spacing in the vertical direction. A sense of the true spacing can be found by comparing to data in figures 5.6 and 5.7. A splitting of \( \sim 2.3 \) meV is observed at \( D = 0 \). (b) A Landau level ladder diagram illustrating the LL dispersion vs. \( \Delta \) for small displacement fields, as calculated from eqs. (5.5). The \( N = 0 \) LL has been removed here for clarity, but can be seen in fig. 5.1. Vertical arrows indicate optically-active transitions, which preserve spin and valley, for different values of \( \Delta \).
values of $\Delta$ in fig. 5.5b, excluding $N = 0$ for visual clarity and indicating the dipole-allowed transitions for different values of $\Delta$ with vertical arrows. As in monolayer graphene, the dipole-allowed cyclotron transitions are governed by the selection rule $N = |M| \pm 1$ and preserve the spin and valley degrees of freedom.

At $D = 0$ the Fermi energy lies between the $N = 0$ and 1 LLs and two resonances, $|N, \lambda\rangle = |-2, +\rangle \rightarrow |1, +\rangle$ and $|-2, -\rangle \rightarrow |1, -\rangle$, are allowed. As $D$ is increased, the $K^-$ valley of the $N = 1$ LL disperses downward in energy, eventually intersecting the Fermi energy. Over a small range of displacement field, determined by the LL broadening, the $|1, -\rangle$ state is partially occupied and the transition $|1, -\rangle \rightarrow |2, -\rangle$ is allowed in addition to the two that were active at smaller displacement fields, yielding three observable transitions. Eventually, $|1, -\rangle$ fully submerges below the Fermi energy and the transition

---

The optical selection rule for CR in BLG is identical to that of MLG, $N = |M| \pm 1$, permitting cyclotron transitions from $N = 0$ to $N = 1$. Because of the small energy difference between the two, this resonance falls at THz or very far-infrared frequencies at $B = 13$ T, even for modestly large electric displacement fields, placing it below the observable spectral range of these measurements. The nature of this cyclotron transition has been discussed theoretically in Refs. [40] and [41].
$|−2, −⟩ \to |1, −⟩$ is completely Pauli blocked. For displacement fields beyond this point only transitions $|−2, +⟩ \to |1, +⟩$ and $|1, −⟩ \to |2, −⟩$ are allowed, once again producing two observable resonances.

This sequence gives a good accounting of the 2-3-2 peak pattern visible in fig.5.5a. Using typical values for BLG band parameters (see footnote 3), the three peak region should be centered at $\Delta \approx 5$ meV, corresponding to an unscreened displacement field of $D \approx 15$ mV/nm. We observe the three peak regime to extend from approximately $D = 25$ to 100 mV/nm, centered at roughly 60 mV/nm. The range of displacement fields over which the three peaks persist indicates a width of $\sim 25$ meV for the $N = 1$ LL. The discrepancy in the expected (15 mV/nm) and observed (60 mV/nm) central $D$ values can likely be explained by a combination of screening effects (see discussion of eq.5.10) and differences in one or more of the true band parameters $\delta_{\text{dim}}, \gamma_0, \gamma_1,$ and $\gamma_4$ from those used in calculating fig.5.5b. Additionally, the parameters $\gamma_3$ and $\delta_{\text{AB}}$ may contribute to the dispersion of $|1, −⟩$. At the time of writing numerical work is being conducted to determine whether a full accounting of all BLG band parameters can explain this discrepancy.

In the high-$D$ region the small splitting between the two observed resonances appears to gradually diminish with increasing $D$, disappearing near 350 mv/nm and then reemerging at still higher displacement fields. This behavior, indicative of a crossing of the energies of transitions $|−2, +⟩ \to |1, +⟩$ and $|1, −⟩ \to |2, −⟩$, is also observed in numerical calculations performed by Zhang, et al. at 5 T (see figure 6 of Ref. [111]). Further work, in conjunction with Zhang, et al., is being conducted at the time of writing to confirm this effect at 13 T.

We note that even though available theory seems to explain much of the behavior at $\nu = 0$, several mysteries persist. While two transitions, $|−2, +⟩ \to |1, +⟩$ and $|−2, −⟩ \to |1, −⟩$, are expected at $D = 0$, they should also be energetically degenerate, leaving the observed splitting of $\sim 2$ meV unexplained. It is possible that such a splitting could arise from the sublattice symmetry-breaking effects of the boron nitride substrate; splittings of similar size have been seen in the CR of monolayer graphene on hBN (see Ch.4 and Ref. [194]).
Accounting for $\delta_{AB}$ in future numerical work should determine whether this is the case. Alternatively, splittings could arise in the presence of the canted antiferromagnetic ground state observed near $\nu = D = 0$ in BLG.

Additionally, in the intermediate-$D$ regime, two resonances are observed to redshift while the third blueshifts. This is at odds with the behavior predicted by eqs.(5.5) and the numerical work of Ref. [111], both of which would suggest that two resonances should blueshift while the third redshifts. And finally, the apparent observation of a single resonance near $D = 120$ mV/nm, at the transition between the intermediate- and high-$D$ regions, is unexpected, as a continuous crossover from three to two peaks should occur.

### 5.3.2 Cyclotron Resonance at $\nu = 4$

At a filling factor of $\nu = 4$ the Fermi energy lies between the $N = 1$ and $N = 2$ LLs in BLG and interaction effects are minimized. Here, cyclotron transitions may occur between states $|N, \lambda\rangle = |1, +\rangle \rightarrow |2, +\rangle$ and $|1, -\rangle \rightarrow |2, -\rangle$, with transitions into LL $|N\rangle = |1\rangle$ forbidden by Pauli blocking. One therefore expects to observe a single resonance at $D = 0$, where the
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Fig. 5.8: (a) Landau level ladder diagram showing the observed transitions at $\nu = 4$. The involved states are labeled by $|N, \lambda \rangle$, where $N$ is the Landau level index and $\lambda = \pm$ labels the valleys within each LL. (b) Individual spectra characteristic of observations at $\nu = 4$ for zero, intermediate (73 mV/nm), and large (318 mV/nm) displacement fields. (c) The energy difference between the observed resonances as a function of the applied displacement field. The dashed lines are extrapolations to zero energy of mid-infrared optical bandgap measurements made in Refs. [115,116].

Valleys in all LLs are degenerate, which splits in two for $|D| > 0$. At small displacement fields, where the valley splitting in the $|N \rangle = |1 \rangle$ LL is very nearly equal to $\Delta$ while the valley splitting within the $|N \rangle = |2 \rangle$ LL is negligible, one anticipates that the difference in energy between the two transitions should hew closely to that of the single particle band gap at $B = 0$.

The location of the $\nu = 4$ line at $B = 13$ T was determined from dual-gated transport measurements and spectra were collected at a number of displacement fields $D < |660|$ mV/nm. The resulting spectra were combined into a color map, shown in figure 5.9, from which the splitting of the resonances can clearly be seen. The center energies of these resonances were determined from Lorentzian fits, and the gap was calculated as the difference of the high and low energy peaks out to $D = 320$ mV/nm, beyond which the high energy resonance moves outside of the observable spectral window. The results are plotted in fig.5.8c. For comparison we include determinations of the optical gap in BLG at $B = 0$.
made by Zhang, et al. [115] and Mak, et al. [116] using mid-infrared spectroscopy. Both groups directly observed band gaps between 150 and 250 meV at large displacement fields $|D| > 1$ V/nm; the lines in figure 5.8c are extrapolations of this data to zero energy. We find excellent agreement between these extrapolations and the observed cyclotron splittings. Notably, while our cyclotron absorption lines are measured in the far-infrared, at energies between 40 and 90 meV, the splittings observed range from approximately 30 meV at $D = 320$ mV/nm to less than 2 meV at $D \sim 10$ mV/nm.

### 5.4 Cyclotron Resonance at Zero Displacement Field

Cyclotron resonance was also studied as a function of filling factor at $D = 0$. Spectra were collected from $\nu = 0$ to +6 at all integer and half-integer filling factor values, and from $\nu = 0$ to -6 at integer values. The resulting spectra are shown in figure 5.10, while peak energies, extracted from Lorentzian fits to the data, are plotted in figure 5.11a. Comparison of peak energies at $\nu = \pm 4$ are made to observations by Henriksen, et al. [124] at $\nu = \pm 4$ in
Cyclotron Resonance in Dual-Gated Bilayer Graphene

Fig. 5.10: Cyclotron resonance measured as a function of the Landau level filling factor $\nu$ at $D = 0$. Significant electron-hole asymmetry is observed, as are several unidentified resonances which can be seen to disperse with filling factor. Where possible, arrows marking distinct resonances have been included which are color coded to the data plotted in fig.5.11a.

In addition to significant electron-hole asymmetry, several unexpected peaks appear and disperse with changing filling factor. The splitting at $\nu = 0$ is seen to disappear by $\nu = \pm 1$, while additional peaks (green and yellow) of lower intensity appear at lower energies near the same filling factors. These new peaks seem to carry less electron-hole asymmetry relative to the primary peak. While only a single peak is seen for $\nu > 0$, the corresponding peak for
$\nu < 0$ appears to be split by $\sim 2$ meV.

**Fig. 5.11:** (a) Peak energies extracted from the data shown in fig.5.10. (b) Comparisons of resonance energies at $D = 0$ and $\nu = \pm 4$ with observations of Henriksen, et al. [124]. Discrepancies are likely due to the presence of an uncontrolled displacement field in the single-gated samples used in Ref. [124].

Returning to the primary resonance, we note that the observed variation of the resonance energy versus filling factor is reminiscent of effects seen in the CR of monolayer graphene, which have been attributed to many-body interaction effects (see Refs. [124,194] and Ch. 4). Using the relation $\omega_c = eB/m^*$, we find effective masses\(^7\) ranging from $m^*/m_0 = 0.0218$ up to 0.0247, where $m_0$ is the free electron mass, with the lightest mass found at $\nu = 0$. The ratio of the hole and electron effective masses, $m_h^*/m_e^*$, is found to go from 0.988 at $|\nu| = 2$, to 1.052 at $|\nu| = 4$, to 1.065 at $|\nu| = 6$. Masses calculated from the observations in Ref. [124] are found to closely match those reported here. In comparison, studies of Shubnikov-de Haas oscillations in BLG by Zou, et al. [123] previously found $m_h^*/m_e^* = 1.2 - 1.3$, with effective

\(^7\) Masses calculated from the lower energy resonances of uncertain origin correspond to $m^*/m_0 \approx 0.027$. 

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masses of $m_e^* \approx 0.033 m_0$ and $m_h^* \approx 0.043 m_0$ at carrier densities of $|n| = 2 \times 10^{12} \text{ cm}^{-2}$. These discrepancies may well be indicative of electron-electron interactions, which have been predicted to impact the cyclotron resonance in BLG [39, 41–45, 45, 127]. Additionally, large variations in the linewidth, which may also be indicative of many-body effects, are observed in the primary resonance, most notably for $\nu < 0$.

5.4.1 Conclusions

We have presented the first observations of cyclotron resonance in a dual-gated, encapsulated bilayer graphene device. The inclusion of top and bottom gates allows for independent control of both the Landau level filling factor and a layer symmetry-breaking perpendicular electric field, granting access to previously unexplored physics. Our measurements at $\nu = 4$ confirm the presence of an electrostatically controlled optical bandgap at the lowest energies to date. At $\nu = 0$, our observations suggest the importance of the often neglected small band parameters of bilayer graphene in precision measurements. At $D = 0$ our observations of cyclotron resonance suggest less electron-hole asymmetry and lighter effective masses relative to measurements of these quantities made by electronic transport, possibly indicating that electron-electron interactions play a significant role in the CR of bilayer graphene. While this work represents a significant advancement in the study of cyclotron resonance in bilayer graphene, it also indicates a strong need for additional work, both experimental and theoretical.
Appendix A

Infrared Magnetospectroscopy

As discussed in Chapter 3, cyclotron resonance experiments involve spectroscopic measurements of clean samples held at cryogenic temperatures and subjected to intense magnetic fields. The unique sample environment required for such measurements precludes the use of typical spectroscopy instrumentation, necessitating the construction of a dedicated system. In this chapter we present the design and construction of such a system for performing infrared magnetospectroscopy on microscopic samples of atomically-thin materials.

We begin with a general discussion of the technique known as Fourier transform infrared (FTIR) spectroscopy and the FTIR spectrometer we employ in measuring cyclotron resonance, an understanding of which will greatly inform everything that follows. From there we will consider the objectives and constraints that shaped the design of the system. We then discuss the optics which connect the spectrometer to the dilution refrigerator that holds our samples and 14 tesla superconducting magnet at cryogenic temperatures, as well as the optics internal to the fridge, which are responsible for focusing light onto microscopic samples and carefully guiding infrared radiation through a cryogenic environment. Finally, we will consider the means by which infrared light is detected inside the cryostat and the way the resulting signal is processed.
A.1 Fourier Transform Infrared Spectroscopy

Fourier Transform Infrared Spectroscopy (FTIR) is a powerful technique and a staple of modern spectroscopy at frequencies spanning the ultraviolet, visible, near- (NIR), mid- (MIR), and far-infrared (FIR), and offers significant advantages over spectrometers based on dispersive optics.\(^1\) Generally, an FTIR spectrometer works in the following way: Broadband light, covering frequencies of interest, is generated by a source. This light is then conditioned with apertures and filters and passed through an interferometer. The interferometer modulates the light in a frequency dependent way; this modulation is key to the operation of an FTIR spectrometer. Optics then direct the modulated light exiting the interferometer through a sample. Upon passing through the sample, the modulated light now carries information about sample absorption features in the form of missing or reduced spectral content. This light is then collected and directed onto an appropriate photodetector, which transduces it into an electronic signal known as an interferogram. This signal is amplified and digitized by the detector’s associated electronics before being passed to a computer, along with information about the position of the moving mirror inside the interferometer. The computer further conditions and then Fourier transforms the interferogram from the time domain to the frequency domain, reproducing the original broadband spectrum of the light source, now superimposed with absorption features from the sample. Comparison of this spectrum with one collected when no sample is present allows for complete isolation of the sample absorption features. Next we consider in detail some of the major components involved in this process, with an emphasis on the specific components found in our Bruker Vertex 80V.

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\(^1\) For readers interested in learning about FTIR spectrometers and spectroscopy, the author strongly recommends *Fourier Transform Infrared Spectrometry* by Griffiths and de Haseth [217]. For details on the general principles and operating conditions of infrared detectors the reader is referred to *Fundamentals of Infrared Detector Operation and Testing* by John David Vincent [218].
**Fig. A.1:** An FTIR spectrometer. Under typical operation light leaves an internal source (MIR or NIR), passes through an aperture (APT) and an optional optical filter (OF), and is directed into a Michelson interferometer which houses a beamsplitter (BMS) at its center. Under typical operation, optics direct the modulated light exiting the interferometer through an internal sample stage. This light is then collected and directed onto an infrared detector ($D_1$ or $D_2$). The modulated signal transduced by the detector, known as an interferogram, is electronically amplified and digitized before being sent to a computer (not pictured). The computer performs additional signal conditioning and Fourier transforms the digitized interferogram, producing the original spectrum of the internal source, which is now superimposed with sample absorption features.

**Source** - A variety of light sources can be used in an FTIR spectrometer depending on the spectral range of interest. The most commonly employed source for mid- and far-IR spectroscopy is the globar, a silicon carbide filament typically heated to temperatures between 1200 and 1900 K. The standard MIR globar source inside the Bruker Vertex 80v has a temperature of ~1370 K under vacuum and produces 25 mW of power at the focus of the internal sample stage when an 10 mm diameter source aperture is used. An image of an actual globar can be seen in fig. A.2; plots of its mid-IR and far-IR blackbody spectra are shown in fig. A.5 and fig. A.6, respectively.

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2 This information was provided by Sergey Shilov of Bruker Optics in a private communication.
**Interferometer** - The interferometer most commonly found in FTIR spectrometers is a Michelson interferometer. To achieve modulation of the incoming light a mirror at the end of one of the two arms of the interferometer is moved forward and backward along the direction of the arm. The movement of this mirror establishes a time dependent modulation of all wavenumbers, $\tilde{\nu} = 1/\lambda$, of light in the interferometer.\(^3\) For a given mirror velocity $V_m$, each wavenumber in the interferometer interferes constructively and destructively at a frequency $f_{\tilde{\nu}}$ unique to that wavenumber as determined by

$$f_{\tilde{\nu}} = V_m \tilde{\nu}.$$  \hspace{1cm} (A.1)

The precision of an FTIR spectrometer ultimately depends on the precision with which $V_m$ is known. In almost all modern FTIR spectrometers the position and velocity of the moving mirror are determined through measurements of the interference pattern of a helium neon

\(^3\) The electromagnetic wavenumber, $\tilde{\nu} = 1/\lambda$, often measured in units of inverse centimeters (cm\(^{-1}\)), is the conventional unit of FTIR spectroscopy. These units are nice because they are directly proportional to energy, like temporal frequency, $\nu$, while still retaining some sense of the length scales involved.
(HeNe) laser\(^4\) of wavelength \(\lambda_{HN}\), which is also directed through the interferometer. Measuring the frequency \(f_m\) at which the laser interference fringes go through a complete cycle of constructive and destructive interference allows for direct measurement of the mirror velocity, \(V_m = \lambda_{HN} f_m\), with the accuracy of the mirror position measurement being comparable to \(\lambda_{HN}\). In a Michelson-type FTIR spectrometer the measured modulation frequency of a given wavenumber is

\[
f_\nu = 2\lambda_{HN} f_m \tilde{\nu},
\]

where \(2\lambda_{HN} f_m\) is the effective mirror velocity [217]. For a HeNe modulation frequency of \(f_m = 5\) kHz, corresponding to a mirror velocity of 0.3164 cm/s, wavenumbers between \(\tilde{\nu} = 50\) cm\(^{-1}\) and 5000 cm\(^{-1}\) will be modulated at frequencies between \(f_\nu = 32\) Hz and 3.2 kHz.

\[\text{Fig. A.3:}\] An interferogram collected using a KBr mid-IR beamsplitter. The observed waveform is characteristic of a broadband light source. The large signal seen at zero mirror displacement, or zero path difference (ZPD), occurs when all wavelengths of light inside the interferometer interfere constructively.

\(^4\) The HeNe wavelength employed is \(\lambda_{HN} = 632.8\text{nm} = 6.328 \times 10^{-5}\text{cm}\).
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The sum of the modulated light leaving the interferometer as a function of mirror position is known as an interferogram, an example of which can be seen in fig.A.3. When the mirror position is such that both arms of the interferometer are of equal length, all wavelengths of light in the interferometer constructively interfere and a large signal is observed. This occurs under the condition of zero path difference, or ZPD. As the mirror moves away from this point all wavelengths interfere constructively and destructively at unique frequencies $f_\nu$, with the sum of these various interference patterns producing the interferogram waveform shown in fig.A.3. Measurement and subsequent Fourier transformation of this interferogram allows for precise reconstruction of the light source’s broadband spectrum.

The spectral resolution of a measurement is determined by the distance the moving mirror travels during the collection of a spectrum; as the mirror moves further from ZPD, higher frequency content is added to the interferogram. The resolution of a spectrum, $\Delta\tilde{\nu}$, is given by $\Delta\tilde{\nu} = 1/d$, where $d$ is the distance traveled away from ZPD by the mirror. The tradeoff in collecting higher resolution spectra is an increase in the time required to achieve an equivalent signal-to-noise (SNR) ratio.

A beamsplitter, which can be found at the center of the interferometer, must be appropriately selected depending on the spectral range of interest. While a wide variety of materials are available, two types of beamsplitter were employed in the measurements presented in this thesis: a potassium bromide (KBr) beamsplitter, best suited for mid-IR wavenumbers, and a multilayer mylar beamsplitter appropriate for work in the far-IR. Images of both beamsplitters are shown in fig.A.4, while plots of spectra collected with the KBr and mylar beamsplitters can be seen in fig.A.5 and fig.A.6, respectively.

5 If the interferometer is the heart of an FTIR spectrometer, the interferogram is its heartbeat.
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**Fig. A.4:** (Left) A KBr beamsplitter, commonly employed for work at mid-IR wavenumbers between 400 and 7000 cm\(^{-1}\). A spectrum collected using this beamsplitter can be seen in fig. A.5. (Right) A multilayer mylar beamsplitter, best suited for work at far-IR wavenumbers between 50 and 400 cm\(^{-1}\). A spectrum collected using this beamsplitter can be seen in fig. A.6.

**Translating Optics** - Upon leaving the interferometer, the collimated beam of modulated light must be directed to its intended destination, which is typically through a sample and onto a detector. This may involve not only redirecting the light, but also changing its divergence for the purpose of focusing it onto a sample or detector. While changing the direction and divergence can be accomplished with lenses, mirrors are virtually always employed in modern FTIR spectrometers since they do not suffer from chromatic aberration. In general, to minimize the loss of light that inevitably occurs upon reflection, the number of mirrors is minimized and a material with the highest possible reflectivity over the spectral range of interest is used. Aluminum or gold mirrors are most often found in FTIR spectrometers due to their high infrared reflectivity. Simple redirection of light can be achieved with flat
mirrors, while focusing and defocusing of light is accomplished using spherical, ellipsoidal, or off-axis paraboloidal mirrors, with the latter being most common. In fig. A.1 multiple flat and off-axis paraboloidal mirrors can be seen, as well as a single spheroidal mirror behind the MIR source. Note that the direction of propagation and the divergence always changes when light leaves an off-axis paraboloidal mirror, while only the direction is changed by a flat mirror.

**Detector** - Once the modulated light has interacted with the sample it must be transduced into an electronic signal. A wide variety of photoresponses exist in nature and almost all of them are utilized to make photodetectors. Broadly, IR detectors are classified as either thermal or quantum, with the major photoresponses employed including bolometric, photovoltaic, photoconductive, thermoelectric, and pyroelectric [218]. Two pyroelectric detectors, one suited for work at mid-IR frequencies and one optimized for detection in the far-IR, are included in the Vertex 80v and provide convenient room temperature operation. The ideal photodetector would produce a large response and little noise, respond to changes in incident light in a negligible amount of time, be sensitive to arbitrarily low levels of illumination, display the same response to every frequency of light over a broad range of the electromagnetic spectrum, and operate at room temperature. In practice, real detectors can only achieve a few of these criteria simultaneously. To meet the significantly higher sensitivity requirements of our magneto-optical system a 4 K composite silicon bolometer is used, which we discuss in greater detail below.

**Electronics** - The electronic signal generated by a photodetector is often too small to be useful, so it is typically amplified. For the resulting signal to be manipulated by a computer it must first be digitized, which is achieved by passing the amplified signal to an analog-to-digital convertor (ADC). Often overlooked, the computer is essential for processing the fast Fourier transform algorithms on which FTIR spectroscopy is so crucially reliant. In 2019, virtually any consumer computer is more than capable of performing such calculations, but the lack of computing resources long prevented early work on FTIR from proceeding beyond
Fig. A.5: A mid-infrared (MIR) spectra collected using a KBr beamsplitter, KBr optical window, and composite silicon bolometer. Sharp peaks visible near 400 and 550 cm\(^{-1}\) correspond to harmonics of 60 Hz noise. The y-axis is proportional to detected intensity, the units are arbitrary. Comparison to the y-axis of fig. A.6 should be avoided.
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Fig. A.6: An FIR spectra collected using a mylar beamsplitter, polyethylene optical window, and composite silicon bolometer. The y-axis is proportional to detected intensity, the units are arbitrary. Comparison to the y-axis of fig. A.5 should be avoided.

Fig. A.7: A useful plot to consider when deciding which optical setup to employ. Shown is the intensity of light detected using the KBr beamsplitter (blue), the mylar beamsplitter (red), and the mylar beamsplitter with a 3.2 mm thick polyethylene window in the beam path (green). Using the mylar-polyethylene configuration is preferable only when working below 400 cm$^{-1}$. 

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A.2 System for Low Temperature Infrared Magnetospectroscopy

The unique sample conditions required for cyclotron resonance measurements, namely cryogenic sample temperatures and the application of intense magnetic fields to the sample,\textsuperscript{6} preclude the possibility of using the sample stage inside an FTIR spectrometer. Fortunately, these instruments typically provide multiple options for directing light modulated by the interferometer out of the spectrometer. In some sense, the challenge of designing and constructing a system for performing FTIR spectroscopy inside a remote cryostat then becomes a matter of unfolding the spectrometer: the roles of the translating optics, detector, and electronics discussed in the last section must be reproduced outside of the spectrometer, though they may be subject to additional constraints. Our system consists of a precision FTIR spectrometer (Bruker Vertex 80v) coupled to a closed-cycle dilution refrigerator (BlueFors LD 400) which houses a 14 tesla superconducting magnet and under normal operating conditions has achieved a base temperature below 10 mK. An illustration (fig. A.8) and image (fig. A.9) of the system are provided.

\textsuperscript{6} See Chapter 3 for a detailed discussion these requirements.
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Fig. A.8: An image of the final design of our system for infrared magnetospectroscopy made using computer aided design software (SolidWorks). Ray tracing software (APEX) was used to verify the optical performance of the system during the design phase.

A.2.1 Objectives and Constraints

The design goals for our magneto-optical system are simple to state. We need to direct the light out of an FTIR spectrometer and into a cryostat. Upon entering the cryostat, that light must be focused onto a sample with typical linear dimensions of $\sim 10\mu m$, which must be held at a particular point inside the bore of a superconducting magnet.\(^7\) After passing through the sample, this light must be re-collimated to allow for efficient propagation over a distance of approximately 0.5 m, after which it must be collected and condensed onto an infrared detector. The infrared detector must operate at low temperatures ($\sim 4$ K) over a broad

\(^7\) A uniform magnetic field is obtained within a 1 cm\(^3\) volume at the magnet’s center.
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range of frequencies spanning the mid- and far-IR and be sensitive enough for observations of very small changes ($\lesssim 1\%$) in the spectral radiance of a small amount of incident light without the need for excessively long measurement integration times. The electronics that amplify and digitize the resulting signal should be quiet so as not to degrade the detector’s signal-to-noise ratio. The radiation entering the cryostat should be handled so as to minimize heating: unwanted background radiation should be rejected to the greatest extent possible while the optics internal to the cryostat should be efficient enough that only low intensity light from the spectrometer is required to produce a useable signal. It is crucial that the heat load on the superconducting magnet be minimized to the greatest extent possible since even low level heating will reduce the maximum magnetic field that can be obtained. Obtaining the lowest sample stage temperatures possible is also desirable, since much of the physics we eventually hope to explore only emerges at temperatures below 1 K.

The intense magnetic fields generated by the magnet lead to the first major design constraint: to protect the electronics inside the spectrometer it must be located at a considerable distance (2 m) from the cryostat. While the light exiting the interferometer is reasonably well-collimated, its small divergence would nevertheless lead to an unacceptable loss of light as it propagates to the fridge. This necessitates the use of custom beam-shaping optics to condition the light’s divergence as it moves from the spectrometer to the cryostat.

The small linear dimensions ($\sim 10 \mu\text{m}$) of our samples requires that the beam of light entering the cryostat, which has an initial diameter of $\sim 40 \text{ mm}$, be focused to a point comparable in size to the sample. In practice, a focal point with a diameter between 1 and 2 mm is achieved and the excess light is rejected by apertures placed immediately above and below the sample, as shown in fig.A.15. Additionally, the need to perform electronic transport measurements on our samples requires appropriate wiring of the sample stage, as well as a system for making breakable electronic connections to contacts on the sample mount.

To avoid absorption of infrared light due to ambient water vapor, the Vertex 80v operates under a vacuum of $\sim 1 \text{ mbar}$, while our closed cycle dilution refrigerator operates under a
considerably harder vacuum of $1 \times 10^{-6}$ mbar. The need to operate these two machines at significantly different pressures necessitates a design in which the two vacuum chambers are separated by an infrared-transparent optical window. The optics joining the two machines are housed inside a vacuum-tight enclosure which shares the spectrometer’s vacuum. Additionally, a pneumatically-operated gate valve is mounted at the base of the fridge, allowing the cryostat to be completely isolated whenever the need arises.

Fig. A.9: An image of the assembled system in operation.
A.2.2 External Optics

The design of the optics responsible for directing light from the spectrometer to the base of the cryostat is essentially that of a telescope with unity magnification: To avoid propagation losses the nearly-collimated light exiting the spectrometer is brought to an intermediate focus roughly halfway between the spectrometer and cryostat (see fig. [?]). This is achieved using a custom gold coated off-axis paraboloidal (OAP) mirror with a 1 m focal length, which was made by ThorLabs. A custom optical enclosure was machined to house this mirror and associated off-the-shelf optical components (fig. A.10).

To re-collimate the light before it enters the fridge an identical OAP is mounted directly under the cryostat, which also serves to redirect the light from a horizontal to vertical direction (fig. A.11). The mirror is set in a three-way adjustable kinematic mount that allows for adjustment of the mirror’s pitch and yaw and also for small amounts of translation in the $\hat{z}$-direction. Vacuum feedthroughs allow these adjustments to be made while the system is under vacuum. This adjustability is key to maximizing alignment with the optical axis of the sample stage, which has a strong impact on the detected signal.
**Fig. A.10:** A raytracing simulation and image of the beam shaping optics responsible for focusing the light exiting the spectrometer. The direction of light travel is from top to bottom. The first mirror is off-axis paraboloidal and has a focal length of 1 m. The last three mirrors are flat. All four are gold coated for maximum infrared reflectivity and are 50.8 mm in diameter. The second and third mirrors are mounted on sliding tracks, allowing them to translate toward or away from the other two. The first and fourth mirrors are set in three-way adjustable kinematic mounts, allowing for considerable control of the optical alignment.
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Fig. A.11: (Left) The vacuum enclosure housing the second 1 m focal length OAP and hardware for mounting the optical windows that hermetically separate the fridge and spectrometer. The pneumatic gate valve used to isolate the cryostat can be seen immediately below the fridge. The valve visible at the top left of the enclosure is used to evacuate the space between the window and the gate valve prior to opening the gate. Visible below the enclosure are the three knobs used to adjust the mirror. A hatch on the backside allows for easy access to the optical window. (Right) The second OAP.

A.2.3 Internal Optics

The design of the internal optics was specialized enough to preclude the use of any off-the-shelf components and, as such, each piece was machined in-house. The requirement of focusing a $\sim 40$ mm diameter beam to a point $\sim 1$ mm across was achieved using a paraboloidal cone. Parabolas have the defining geometric property that any paraxial\(^8\) ray entering the parabola will be reflected such that it intersects the parabola’s focal point; conversely, any ray emanating from a parabola’s focus will be redirected such that it becomes paraxial upon intersecting the parabola. Light entering the parabola at an angle relative

\(^8\) In optics, a paraxial ray is one that propagates parallel to the optical axis of an optical system or component.
to the symmetry axis will not pass though the focus and is instead rejected, being reflected back out of the opening of the parabola. The sample stage design chosen is that of two back-to-back paraboloidal cones, machined such that their foci precisely overlap with the sample mounted at exactly this point. Raytracing was used to confirm this design (fig. A.12), after which the complex optical surfaces were machined using a computer-controlled three-axis mill. Machining of the outer surfaces and polishing of the interior was done by hand and, finally, each piece was chemically gold plated (fig. A.13). A hollow oxygen-free copper tube mounted to the mixing chamber plate of the dilution refrigerator suspends the top sample stage cone such that its focal point lies precisely at the center of the magnet’s bore (fig. A.14). This top cone also integrates the wiring required for electronic transport measurements.

The design of the sample stage is such that one first aligns the sample over a hole in the center of a specially designed sample mount (fig. A.15). This mount then rigidly attaches to the top face of the top cone in such a way that the center of the mount is automatically aligned with the optical axes of the cones, with the sample positioned precisely at the location of the overlapping foci. The bottom cone is then attached to the top cone using three screws. Attaching the bottom cone to the top cone also brings the copper or gold contacts of the sample mount into contact with “fuzz buttons”, which are mounted in a ceramic ring on the bottom face of the top cone, making direct, but breakable, electrical contact to phosphor bronze measurement wires (fig. A.14).
Fig. A.12: (Left) Raytracing simulation of the sample stage paraboloids and an exploded view showing both cones and the ceramic sample mount and fuzz button ring. (Right) The compound paraboloidal concentrator to which the detector is mounted accepts incident light over a range of angles and concentrates it to a region of finite width.
Fig. A.13: Top and bottom views of (left to right) the compound paraboloidal concentrator, the bottom sample stage paraboloid, and the top sample stage paraboloid.
Fig. A.14: Three views of the mounted internal optics. (Left) The top sample stage paraboloid suspended from the mixing chamber plate. The integrated wiring and fuzz button contacts are also visible. (Center) The sample stage with the lower paraboloid in place. (Right) After leaving the sample stage, collimated light passes through holes in several of the fridge plates before being collected by the directed onto a bolometer by the compound paraboloidal concentrator, which can be seen here hanging from the 4 K plate of the cryostat.

As previously noted, the focal point of the sample stage is considerably larger than a typical sample. To reduce the excess light that would otherwise pass on all sides of our sample and drown out the signal of interest we place metal film apertures directly above and below the sample.\(^9\) When mounting the sample to the sample mount, a copper foil aperture with a

\(^9\) If you’re planning to attempt this, allow me to share a few tips and tricks. (1) Find yourself a sharp needle and hide it. (2) Use the stereoscope and find some bright lights that you can easily move around. Figuring out the right lighting is crucial. (3) When aligning the device over the bottom aperture, use the edge of a razor blade to check the alignment with the crosshairs. (4) In making the top aperture I would use the very tip of the sharpest pin I could find; half the time the hole was still too big. After poking an appropriately sized hole, flip it over and use the pin to flatten out the material around the hole. (5) Bend the corners or edges of the aperture up to form handles for your tweezers to hold on to. This allows you to drop the aperture onto the device and can prevent sliding of the aperture when you pull away. (6) To apply the vacuum grease that holds the foil in place, take the wooden stick from a Q-tip, cut it at an angle, and get an extremely small amount of vacuum grease on the end. Dab tiny amounts directly on the wafer, close enough that the outer edges of the foil will rest on it, but as far from the sample as possible. (7) While it’s typically a no-no to bring metal tweezers anywhere near a device, I found that I couldn’t reliably get carbon fiber tweezers to release the aperture when the time came. Caution is obviously a must if metal tweezers are used. (8) Place you elbows on the counter to steady your hands. Holding your breath also helps for the
hole slightly less than 1 mm across is first applied to the mount. Crosshairs scored into the foil are used to align the device, which typically lives on an opaque silicon substrate, with the hole in the foil. After electrical contact is made between the sample bond pads and the sample mount contacts, either by wire bonding or soldering, an aluminum foil aperture with a typical hole diameter of 100 \( \mu \text{m} \) or less is placed over the device. Using foil with photoresist spun on the underside prevents electrical short between sample wires, while a very small amount of vacuum grease is used to hold the foil in place. Pictures illustrating this process are included in figure A.15.

really delicate parts. (8) Find some dead devices and practice. A lot. (9) Alternatively, you could always fabricate an aperture into or onto the device, though this comes with its own set of issues.
**Fig. A.15:** (Top left) A copper foil bottom aperture on a sample mount. The hole is slightly less than 1 mm in diameter. (Top right) A device after being aligned over a bottom aperture. Magnification is 2.5x. (Bottom right) A $\sim 90 \mu m$ top aperture in place over a device. Vacuum grease is visible as dark blobs around the perimeter of the aperture. (Bottom left) The same device and aperture shown at right, here at 20x magnification. The device area is $\sim 700 \mu m^2$

Unwanted light entering the fridge, either in the form of background radiation or non-paraxial light from the spectrometer, poses a significant problem. Left unaddressed, much of this light is deposited on the inner surface of the magnet bore or on the magnet’s underside. This heat also overwhelms the cooling power of the dilution refrigerator, preventing operation at
sample stage temperatures below several kelvin. The addition of a simple radiation shield (fig. A.16) prevented much of this stray light from directly reaching the magnet, resulting in noticeably lower magnet temperatures and higher attainable magnetic fields, though it did nothing to reduce the base temperature of the sample stage (∼6 K). More recently this simple shield was replaced by a second set of back-to-back paraboloidal cones (fig. A.16), which act to reject a great deal of the light that would otherwise be deposited on the magnet or rejected by the lower cone of the sample stage. Stray light is further rejected by placing a 1 mm aluminum foil aperture at the shared focus of the two cones. With this addition, the magnet temperature has dropped such that the magnet’s maximum field can be achieved, while the sample stage temperature when operating only the cryocooler has dropped to ∼4 K. Most significantly, the heat load has been reduced to the point that the dilution refrigerator can now be operated in the infrared configuration. Recently, broadband infrared spectroscopy has been performed at a sample stage temperature of ∼200 mK, granting this system a truly unique capability. With further optimization lower temperatures will likely be obtainable.
Fig. A.16: (Left) First-generation radiation shield for diverting heat away from the magnet. (Right) Second-generation radiation shield made from back-to-back paraboloidal cones. This improved shielding dramatically reduces the total heat load on the fridge, allowing for operation of the dilution refrigerator while spectroscopy is being performed. A $\sim 1$ mm foil aperture is located at the foci of the two cones.

A.2.4 Infrared Detector

Infrared detection is accomplished using a composite silicon bolometer which is mounted to the 4 K plate of the fridge. To collect the light that has been nominally re-collimated by the top cone of the sample stage a compound paraboloidal concentrator, also known as a Winston cone, is mounted below the bolometer (figs. A.13 and A.14). Such concentrators have a finite angle of acceptance, making them excellent for collecting light that is not perfectly collimated, and concentrate light to a region of finite extent rather than a point (fig.A.12).

The bolometer, which acts essentially like a temperature dependent resistor, was removed
from a dewar system sold by Infrared Laboratories, Inc., and mounted inside the fridge along with its accompanying electronics (fig. A.17). Two twisted pairs of phosphor bronze wires were non-inductively hand-wound and sunk to the 50 K and 4 K stages of the fridge using oxygen-free copper bobbins. These four wires supply a bias voltage to the bolometer, a grounding wire, and a source and drain for biasing a J230 JFET transistor (fig. A.18). This JFET, which is mounted to the top of a thermally isolating fiberglass post and heated to a temperature of approximately 70 K by a biased resistor, serves as an electronic buffer between the bolometer and a room temperature voltage preamplifier. The JFET and 4 K heat sinking bobbin are mounted to an oxygen-free copper accessory ring, which simplifies the mounting and un-mounting process. The bolometer is wired to ground in series with a matched 10 MΩ load resistor and biased by a 9 V battery. The pair act essentially as a voltage divider, with the bolometer playing the role of a variable resistor. As the light incident on the bolometer is modulated, its resistance varies in time, producing a varying voltage signal on the gate of the JFET. The resulting signal at the JFET source, which is the interferogram, is then amplified by a preamplifier, digitized by a Bruker ADC, fed to the spectrometer, and finally conditioned and Fourier transformed by a computer, producing an infrared spectrum.
Fig. A.17: (Left) A composite silicon bolometer shown bonded to a diamond absorber and suspended between two wires. (Right) The backside of the bolometer enclosure and the accompanying accessory ring. The 10 MΩ load resistor can be seen mounted to the back of the enclosure. The JFET on its fiberglass post and the 4 K wire heat sinking bobbin can both be seen mounted to the accessory ring.

![Diagram of the bolometer circuit mounted inside the fridge.](image)

**Fig. A.18:** A diagram of the bolometer circuit mounted inside the fridge. Reproduced from a document made and distributed by Infrared Laboratories, Inc.
Appendix B

Electronic Transport Measurements

Here we give a brief overview of some commonly employed electronic transport measurements. It should be stressed that these techniques are by no means unique to graphene and are applied to an enormous variety of materials in condensed matter experiments and industrial semiconductor characterization. These simple measurements provide a great deal of insight into the electronic properties of materials, especially when used in conjunction with an applied magnetic field. In the following discussion we consider only two-dimensional systems, largely following Datta [149] and Ando, et al. [92], however, we note that the more general three-dimensional relations may be found in a number of excellent and widely-available sources.
The simplest transport measurement one can perform consists of driving a current, \( I \), through a material and measuring the corresponding voltage drop, \( V \), along the direction of current flow.\(^1\) The resistance may then be calculated from

\[
V = IR,
\]

which is the well-known Ohm’s relation. The resistance, \( R \), is an extrinsic quantity, but normalizing by the relevant sample dimensions (see fig. B.1) yields the resistivity

\[
\rho = R \frac{W}{L},
\]

which is an intrinsic property of a material. An important related quantity, the electronic

\(^1\) We take the direction of current flow to be in the \( \hat{x} \)-direction.
Electronic Transport Measurements

conductivity, $\sigma$, is the inverse of the resistivity

$$\sigma = \rho^{-1}. \quad (B.3)$$

The electric field established in the sample, $E = V/L$, and the resulting current density, $J = I/W$, are related by the resistivity

$$E = \rho J. \quad (B.4)$$

In the presence of a magnetic field applied perpendicular to the conducting channel, moving charges are deflected by the Lorentz force such that a non-zero current appears in the $\hat{y}$-direction, and the tensor nature of the resistivity is revealed:

$$\begin{pmatrix} E_x \\ E_y \end{pmatrix} = \begin{bmatrix} \rho_{xx} & \rho_{xy} \\ \rho_{yx} & \rho_{yy} \end{bmatrix} \begin{pmatrix} J_x \\ J_y \end{pmatrix}. \quad (B.5)$$

The magnetoresistivity, or longitudinal resistivity, $\rho_{xx}$, is

$$\rho_{xx} = \frac{V_{xx}W}{I\cdot L}, \quad (B.6)$$

which is identical to the definition of the zero field resistivity in eq. (B.6). The Hall resistance, also known as the transverse resistivity, $\rho_{xy}$, is defined as

$$\rho_{xy} = \frac{V_{xy}}{I}. \quad (B.7)$$

The magnetoresistivity retains its zero field relationship to the conductivity, $\rho_{xx} = \sigma^{-1}$, while the off-diagonal components of the resistivity tensor are related by

$$\rho_{xy} = -\rho_{yx} = \frac{\mu B}{\sigma} = \frac{B}{en}. \quad (B.8)$$
where $\mu$ is the electronic Hall mobility$^2$, $e$ is the absolute value of the electron charge, and $n$ is the two-dimensional charge carrier density.

Crucially, electronic transport measurements at low magnetic fields$^3$ allow for precise determination of the electronic carrier density through the relationship

$$n = \frac{1}{e} \left[ \frac{d\rho_{xy}}{dB} \right]^{-1}. \quad \text{(B.10)}$$

With the carrier density in hand, the Hall mobility, which is a metric of the ease with which charge carriers move through a material, may be determined from

$$\mu = \frac{1}{e} \left[ n \rho_{xx} \right]^{-1}. \quad \text{(B.11)}$$

$^2$ Not to be confused with the commonly encountered field effect mobility,

$$\mu_{FE} = \frac{1}{e} \frac{d\sigma}{dn}. \quad \text{(B.9)}$$

$^3$ We take 'low' magnetic fields to mean those fields below the onset of Shubnikov de Haas oscillations and well below the onset of Hall quantization. At these low fields the classical, linear relationship between $\rho_{xy}$ and $B$, given in eq. (B.8), is maintained.
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