

ABSTRACT

Title of Dissertation: **CHIRAL LIGHT-MATTER INTERACTION
IN FERMIONIC QUANTUM HALL SYSTEMS**

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Achieving control over light-matter interactions is crucial for developing quantum technologies. This dissertation discusses two novel demonstrations where chiral light was used to control light-matter interaction in fermionic quantum Hall systems. In the first work, we demonstrated the transfer of orbital angular momentum from vortex light to itinerant electrons in quantum Hall graphene. In the latter, we demonstrated circular-polarization-dependent strong coupling in a 2D gas in the quantum Hall regime coupled to a microcavity. Our findings demonstrate the potential of chiral light to control light-matter interactions in quantum Hall systems.

In the first part of this dissertation, we review our experimental demonstration of light-matter interaction beyond the dipole-approximation between electronic quantum Hall states and vortex light where the orbital angular momentum of light was transferred to electrons. Specifically, we identified a robust contribution to the radial photocurrent, in an annular graphene sample within the quantum Hall regime, that depends on the vorticity of light. This phenomenon can be

interpreted as an optical pumping scheme, where the angular momentum of photons is transferred to electrons, generating a radial current, where the current direction is determined by the vorticity of the light. Our findings offer fundamental insights into the optical probing and manipulation of quantum coherence, with wide-ranging implications for advancing quantum coherent optoelectronics.

In the second part of this dissertation, we review our experimental demonstration of a selective strong light-matter interaction by harnessing a 2D gas in the quantum Hall regime coupled to a microcavity. Specifically, we demonstrated circular-polarization dependence of the vacuum Rabi splitting, as a function of magnetic field and hole density. We provide a quantitative understanding of the phenomenon by modeling the coupling of optical transitions between Landau levels to the microcavity. This method introduces a control tool over the spin degree of freedom in polaritonic semiconductor systems, paving the way for new experimental possibilities in light-matter hybrids.

Chiral light-matter interaction in 2D quantum Hall systems

by

Deric Session

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Foreword

The results of my graduate studies have contributed to the following publications / preprints, which have portions represented in this dissertation:

Deric Session, Mahmoud Jalali Mehrabad, Nikil Paithankar, Tobias Grass, Christian J. Eckhardt, Bin Cao, Daniel Gustavo Suárez-Forero, Kevin Li, Mohammad S. Alam, Kenji Watanabe, Takashi Taniguchi, Glenn S. Solomon, Nathan Schine, Jay Sau, Roman Sordan, Mohammad Hafezi, “Optical pumping of electronic quantum Hall states with vortex light”, *arXiv:2306.03417*.

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Dedication

To my parents for all their support throughout my many years of school.

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First, I would like to thank my advisor Mohammad Hafezi for welcoming me into his group and giving me this opportunity. Mohammad's broad knowledge of physics and ability to explain concepts in an understandable way have been a constant source of motivation. I am still amazed at how Mohammad has been able to successfully manage such a large group with broad interests including experimentalists and theorists.

I was fortunate to have multiple mentors during my time in graduate school and I would like to give special thanks to Bin Cao, Julia Sell, and Jon Vannucci. Their hard work was instrumental in building much of the lab we have today. In particular, my main experiment, which is the subject of Chapter 2 of this dissertation, would not have been possible without the years of dedicated work that Bin spent making the setup. In the last year before Bin left, he was gracious enough to spend time teaching me how to perform the measurements that I would later need for the project and testing the graphene samples that I fabricated. Testing my failed graphene samples with him was one of the most enjoyable times that I had in graduate school. I am also grateful to Julia for dedicating her time during my first year in the group to teaching me electron-beam lithography as well as other fabrication techniques. In working with Jon on the Bluefors setup, I was able to learn techniques such as data acquisition using Python, soldering electrical connectors, and how to use our dilution fridge. These are all things that I would need throughout my time in this group and will likely continue to need. In the last couple of years since these three left the group, I

have been the person other members of the group call on when there are problems in the lab. The reason why I can help in these situations is because of the time I spent working with Bin, Julia, and Jon.

I also would like to thank the postdocs in our group that I have had the opportunity to work with: Daniel Gustavo Suárez-Forero, Mahmoud Jalali Mehrabad, and Sunil Mittal. I was only able to work with Sunil for my first couple of years in the group before he started his faculty position at Northeastern University. However, I was inspired by his knowledge of physics and lab skills. Thanks to Daniel and Mahmoud, our group has been more productive than ever. The projects on which I have worked would not have been successful without their efforts and I would not have reached this point without their encouragement. I wish Daniel all the best in his new faculty position at the University of Maryland Baltimore County and Mahmoud in whatever his next position may be.

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The work discussed in this dissertation would not have been possible without the help of many collaborators not mentioned above. In particular, Nikil Paithanker and Roman Sordan were key in providing us with a graphene Corbino sample that we could use for our experiment, “Optical pumping of electronic quantum Hall states with vortex light.” For this experiment, I had

other great collaborators, including Tobias Grass, Christian Eckhardt, Glenn Solomon, Nathan Schine, and Jay Sau. For our experiment, “Spin-selective strong light-matter coupling in a 2D hole gas-microcavity system,” I would like to thank our coauthors, Patrick Knüppel, Stefan Fält, and Werner Wegscheider who were not mentioned above.

There are also many staff members at the University of Maryland who helped me with my research during my time here. The FabLab staff: Tom Loughran, Mark Lecates, Jon Hummel, and Nam Kim taught me a lot about fabrication and were always there to help when I needed it. I also want to thank Karen Gaskell, who was very helpful whenever I needed to use the AFM, and Alessandro Restelli for his help with electronics and 3D printing. In addition, I also thank Johnna Schmidt for her diligent administrative work in our group.

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List of Abbreviations

2D	two-dimensional
2DEG	Two-Dimensional Electron Gas
2DHG	Two-Dimensional Hole Gas
a_B	Bohr radius
BZ	Brillouin Zone
CB	Conduction band
DBR	Distributed Bragg reflector
E_F	Fermi energy
hBN	hexagonal boron nitride
IQHE	Integer Quantum Hall Effect
FQHE	Fractional Quantum Hall Effect
OAM	Orbital Angular Momentum
ℓ_B	Magnetic length
LG	Laguerre-Gauss
LL	Landau Level
NN	Nearest-neighbor
NNN	Next-nearest neighbor
PC	Photocurrent
QW	Quantum well
SAM	Spin angular momentum
SC	Strong Coupling
SLM	spatial light modulator
TIA	Trans-impedance amplifier
VB	Valence band
VTI	Variable temperature insert
Δ_{PC}	Photocurrent difference
ρ_{xx}	longitudinal conductivity
ρ_{xy}	Hall conductivity
σ_{xx}	longitudinal conductivity
σ_{xy}	Hall conductivity
σ	Pauli matrices
Φ_0	Magnetic flux quantum

Chapter 1: Introduction

1.1 Motivation

There has been a growing interest in investigating chiral and topological effects in photonic systems and also light-matter hybrids [1–3]. Such topological features can be either in the momentum domain and lead to Chern bands or simply in the spatial degrees of freedom, such as optical vortex beams. Specifically, in addition to spin, in the form of polarization, light can also carry orbital angular momentum (OAM) [4–6].

In Chapter 2, we experimentally demonstrate the transfer of OAM from light to electrons in quantum Hall graphene using optical vortex beams. This is primarily motivated by the theoretical work in Ref. [7], in which it is shown that for a quantum Hall system in which the light is resonant between two consecutive Landau levels (LLs), when OAM is transferred from the vortex beam to the electrons the radius of the electronic wavefunction expands, or contracts depending on the sign of the OAM. This would provide coherent manipulation of electronic wavefunctions, which is important for the advancement of future quantum technologies and optoelectronics [8, 9].

In Chapter 3, we demonstrate a method to induce a strong spin selectivity in the light-matter coupling between an electronic transition and a cavity mode. The system is a hole gas in a semiconductor quantum well embedded in a microcavity. It is subject to an external magnetic field such that it is in the quantum Hall regime. There has been increasing interest in combining

strong coupling with broken time-reversal symmetry in chiral systems, as it is expected to unlock new perspectives for the optical manipulation of correlated states of matter [8, 10, 11].

Before exploring Chapters 2 and 3, I will provide an overview of the quantum Hall effect in a two-dimensional electron/hole gas and graphene. Following that, I will explain the concept of chirality in light, focusing on vortex beams.

1.2 Quantum Hall physics

The Hall effect predicts that when charges are confined to two dimensions and exposed to a perpendicular magnetic field, the longitudinal conductivity, σ_{xx} , will go to zero and the Hall conductivity, σ_{xy} , increases proportionally with the magnetic field strength. In 1980, Von Klitzing discovered that the Hall conductivity was quantized,

$$\sigma_{xy} = \nu \frac{e^2}{h} \quad (1.1)$$

where e is the elementary charge, h is Planck's constant, and ν which is called the filling factor is an integer [12]. This is the integer quantum Hall effect (IQHE). It is independent of all microscopic details such as the type of semiconductor material and the purity of the sample. This has been shown to be related to the formation of Landau levels, and that quantization of the Hall conductivity is directly related to the topological properties of the system [12–14]. This will be discussed further in the subsequent sections.

In 1982 it was discovered that ν could also take rational fractional values by Tsui, Störmer, and Gossard [15]. This effect is called the fractional quantum Hall effect (FQHE). The physics of the FQHE differs from that of the IQHE, it involves strong Coulomb interactions and correlations

among the electrons [16]. This dissertation will focus on the integer quantum Hall.

1.2.1 Classical and semi-classical description

The classical equations of motion for a particle with charge q in a magnetic field $\mathbf{B} = B\hat{\mathbf{z}}$ are

$$m\mathbf{r} = q\mathbf{v} \times \mathbf{B}. \quad (1.2)$$

This can be written in matrix form as

$$\begin{pmatrix} \ddot{x} \\ \ddot{y} \end{pmatrix} = \begin{pmatrix} 0 & \omega_c \\ -\omega_c & 0 \end{pmatrix} \begin{pmatrix} \dot{x} \\ \dot{y} \end{pmatrix}, \quad (1.3)$$

where

$$\omega_c \equiv \frac{qB}{m} \quad (1.4)$$

is the cyclotron frequency. The solutions of these equations correspond to circular motion

$$\mathbf{r} = R \begin{pmatrix} \cos(\omega_c t + \varphi) \\ \sin(\omega_c t + \varphi) \end{pmatrix}, \quad (1.5)$$

where R is the radius of the circular orbit and φ is an arbitrary phase. Note that the direction of the circular orbits changes with the sign of the magnetic field.

By analyzing the Lagrangian of the system, it can be shown that the canonical momentum is

$$\mathbf{p} = \mathbf{p}_{\text{mech}} + q\mathbf{A}, \quad (1.6)$$

where $\mathbf{A}$ is the vector potential and $\mathbf{p}_{\text{mech}} = m\dot{\mathbf{r}}$ is the mechanical momentum. Using this in conjunction with the Bohr-Sommerfeld quantization condition ($\oint \mathbf{p} \cdot d\mathbf{q} = nh$), it can be shown that the kinetic energy of the charges is quantized. In the preceding condition, $\mathbf{q}$ is the canonical coordinate and $\mathbf{p}$ is the canonical momentum. To determine the vector potential, the symmetric gauge ($\mathbf{A} = -\frac{1}{2}\mathbf{r} \times \mathbf{B}$) can be used. In cylindrical coordinates for a magnetic field aligned to the z-axis, this is $\mathbf{A} = \frac{1}{2}rB\hat{\theta}$. The integral evaluates to $\oint \mathbf{p} \cdot d\mathbf{q} = q\pi R^2 B$. As the magnitude of the velocity for the particle in the orbit is $v = R\omega_c$, the kinetic energy is $n\hbar\omega_c$. As we will see in Section 1.2.2, this is very similar to the result we find using quantum mechanics.

Going back to the Bohr-Sommerfeld quantization condition:

$$\oint \mathbf{p} \cdot d\mathbf{q} = q \oint \mathbf{A} \cdot d\mathbf{q} = q\Phi_B,$$

where Φ_B is the magnetic flux. This implies that the magnetic flux is quantized,

$$\Phi_B = n \frac{h}{q}.$$

This allows us to define a length scale called the magnetic length:

$$\ell_B \equiv \sqrt{\frac{\hbar}{eB}}. \quad (1.7)$$

The interpretation of the magnetic length, is that for charge carriers with charge $|q| = e$, $2\pi\ell_B^2$ contains one quantum of magnetic flux, $\Phi_0 = h/e$.

Before moving on to the quantum description, we can use the Drude model to find the

classical expectation for the conductivity. The Drude equation of motion is

$$m \frac{d\mathbf{v}}{dt} = -m \frac{\mathbf{v}}{\tau} + \mathbf{F}. \quad (1.8)$$

Here τ is the average scattering time, and for this scenario $\mathbf{F}$ is the Coulomb and Lorentz force [17]. Consider the steady state, where $\frac{d\mathbf{v}}{dt} = 0$. Then $\mathbf{E} = \frac{m}{q\tau} \mathbf{v} - \mathbf{v} \times \mathbf{B}$. Making the substitution $\mathbf{v} = \frac{\mathbf{j}}{nq}$, where $\mathbf{j}$ is the current density and n is the charge carrier density,

$$\mathbf{E} = \frac{m}{nq^2\tau} \mathbf{j} - \frac{1}{nq} \mathbf{j} \times \mathbf{B}. \quad (1.9)$$

This can be rewritten in terms of Ohm's law, $\mathbf{E} = \boldsymbol{\rho} \mathbf{j}$, where $\boldsymbol{\rho}$ is the resistivity tensor. We find $\rho_{xx} = \rho_{yy} = \frac{m}{nq^2\tau}$ and $\rho_{xy} = -\rho_{yx} = -\frac{B}{nq}$. These values are referred to as the longitudinal and Hall resistivity, respectfully. The conductivity matrix can be found by taking the inverse of the resistivity matrix. These values can be measured in a sample, for example a Hall bar like in Fig. 1.1 where $\rho_{xx} = V_L/I$ and $\rho_{xy} = V_H/I$. Notice that the Hall resistivity increases proportional to B and inversely proportional to n . In Section 1.2.4 we will show that the Hall resistivity/conductivity is quantized.

1.2.2 Quantum Hall in a 2D gas

A gas of charges (electrons or holes) confined to two-dimensions (2D) in a material with a parabolic band structure has a Hamiltonian of the form

$$\hat{H}_0 = \frac{\hat{\mathbf{p}}^2}{2m}, \quad (1.10)$$

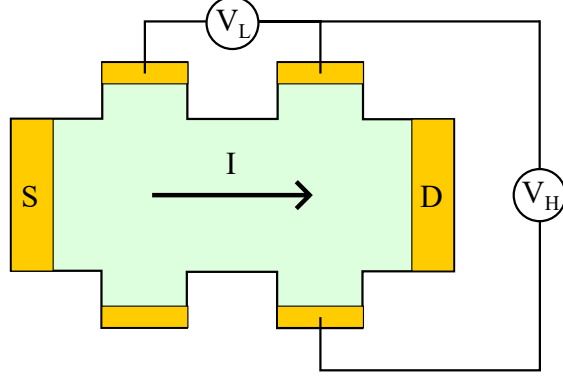


Figure 1.1: Illustration of the Hall bar geometry. The yellow areas represent metal contacts. A current, I , runs from the source (S) and drain (D) contact while the longitudinal voltage (V_L) and Hall voltage (V_H) are measured to obtain σ_L and σ_H .

where $\mathbf{p}$ is the in plane mechanical momentum and m is the effective mass of the charges. In the presence of a classical electromagnetic field, the Hamiltonian can be rewritten as

$$\begin{aligned}\hat{H} &= \frac{1}{2m} |\hat{\mathbf{p}} - q\hat{\mathbf{A}}|^2 \\ &= \frac{1}{2m} \left((\hat{p}_x - q\hat{A}_x)^2 + (\hat{p}_y - q\hat{A}_y)^2 \right).\end{aligned}\tag{1.11}$$

For a perpendicular magnetic field, it is common to use the Landau gauge $\mathbf{A} = (-yB, 0, 0)$. However, we will use the symmetric gauge, $\mathbf{A} = \frac{B}{2}(-y, x, 0)$, as we did in the last section as it is more applicable to the next chapter. We can define the mechanical momenta operators as

$$\hat{\Pi}_x \equiv \hat{p}_x - q\hat{A}_x \tag{1.12}$$

$$\hat{\Pi}_y \equiv \hat{p}_y - q\hat{A}_y. \tag{1.13}$$

The commutator for these two operators is $[\hat{\Pi}_x, \hat{\Pi}_y] = im\hbar\omega_c$. Notice the similarity to the position-momentum commutator. For a uniform magnetic field, these momenta operators can be re-scaled and can be considered as position and momentum operators. The Hamiltonian can then

be rewritten in the form of a harmonic oscillator:

$$\hat{H} = \frac{1}{2m}\hat{\Pi}_y^2 + \frac{1}{2}m\omega_c^2\hat{\Pi}_x'^2, \quad (1.14)$$

where $\hat{\Pi}_x' = \frac{1}{m\omega_c}\hat{\Pi}_x$. Since this is just the quantum harmonic oscillator, we can define ladder operators:

$$\hat{a} = \frac{1}{\sqrt{2m\hbar\omega_c}} \left(\hat{\Pi}_x - i\hat{\Pi}_y \right) \quad (1.15)$$

$$\hat{a}^\dagger = \frac{1}{\sqrt{2m\hbar\omega_c}} \left(\hat{\Pi}_x + i\hat{\Pi}_y \right), \quad (1.16)$$

and rewrite the Hamiltonian as $H = \hbar\omega_c(\hat{a}^\dagger\hat{a} + 1/2)$. This tells us that the energy spectrum is

$$\epsilon_n = \hbar\omega_c(n + 1/2), \quad (1.17)$$

but does not tell us the degeneracy.

To find the degeneracy, we can use another set of operators:

$$\tilde{\Pi}_x \equiv \hat{p}_x + q\hat{A}_x \quad (1.18)$$

$$\tilde{\Pi}_y \equiv \hat{p}_y + q\hat{A}_y. \quad (1.19)$$

Because we are in the symmetric gauge, these operators commute with the previously defined $\hat{\Pi}_x$ and $\hat{\Pi}_y$, and thus also commute with the Hamiltonian. These operators have a similar commutator relationship as the previous momenta operators, $[\tilde{\Pi}_x, \tilde{\Pi}_y] = -im\hbar\omega_c$ and so we can define

another set of harmonic oscillator ladder operators:

$$\begin{aligned}\hat{b} &= \frac{1}{\sqrt{2m\hbar\omega_c}} \left(\tilde{\Pi}_x + i\tilde{\Pi}_y \right) \\ \hat{b}^\dagger &= \frac{1}{\sqrt{2m\hbar\omega_c}} \left(\tilde{\Pi}_x - i\tilde{\Pi}_y \right).\end{aligned}\tag{1.20}$$

Therefore, the eigenstates are given by the Fock states corresponding to the two harmonic oscillators:

$$|n, m\rangle = \frac{\hat{a}^\dagger n \hat{b}^\dagger m}{\sqrt{n!m!}} |0, 0\rangle.\tag{1.21}$$

These states and their energy spectrum, $E_{n,m} = \hbar\omega_c(n + 1/2)$, are called Landau levels (LL).

Following Ref. [14] we can analyze the degeneracy of the ground state ($n = 0$) by looking at the lowest LL wavefunction :

$$\psi_{n=0,m} \sim \left(\frac{z}{l_B} \right)^m e^{-|z|^2/4l_B^2},\tag{1.22}$$

where $z = x - iy$. This state is an eigenstate of angular momentum with $L_z = m\hbar$. The profile of these wavefunctions form concentric rings with radii that increase monotonically with m . The expectation value of the radius is $r = \sqrt{2m}l_B$. Thus, for a disk-shaped region with area $A = \pi R^2$, the maximum number of states M is

$$M = \frac{R^2}{2l_B^2} = \frac{A}{2\pi l_B^2} = \frac{AB}{h/e} = \frac{\Phi_B}{\Phi_0},\tag{1.23}$$

which is just the number of flux quanta.

1.2.3 Quantum Hall in Graphene

Graphene is a 2D material consisting of a single layer of carbon atoms in a hexagonal lattice as shown in Fig. 1.2a. Before we discuss how graphene behaves in the quantum Hall regime, it is important to discuss the band structure of graphene, since it leads to a different Hamiltonian than we have for a 2D gas. The dispersion relation given by the tight-binding model with nearest-neighbor (NN) and next-nearest-neighbor (NNN) hopping is given by the following:

$$\epsilon_{\mathbf{k}}^{\lambda} = t'_{NNN} \left[3 + 2 \sum_{i=1}^3 \cos(\mathbf{k} \cdot \mathbf{a}_i) \right] + \lambda t \sqrt{3 + 2 \sum_{i=1}^3 \cos(\mathbf{k} \cdot \mathbf{a}_i)}, \quad (1.24)$$

where t is the NN hopping amplitude, t'_{NNN} is the effective NNN hopping amplitude, $\lambda = \pm 1$ is the index for the conduction and valence band, and $\mathbf{a}_i$ are the basis vectors of the graphene lattice [18]. In Fig. 1.2c, equation 1.24 is plotted over the Brillouin zone (BZ). The conduction band minima and the valence band maxima meet at the Dirac points which are situated at the K and K' points of the BZ. Near these points Eqn. 1.24 can be expanded such that it reads:

$$\epsilon_{\mathbf{k}, \xi=\pm}^{\lambda} = \lambda \hbar v_F |\mathbf{q}|, \quad (1.25)$$

where ξ is the valley pseudospin, v_F is the Fermi velocity, and $\mathbf{q}$ is the momentum relative to the Dirac point [18]. This is shown in Fig. 1.2d. The effective low-energy Hamiltonian can be represented as

$$H_{\mathbf{q}}^{\text{eff}, \xi} = \hbar v_F \tau^z \otimes \mathbf{q} \cdot \boldsymbol{\sigma}, \quad (1.26)$$

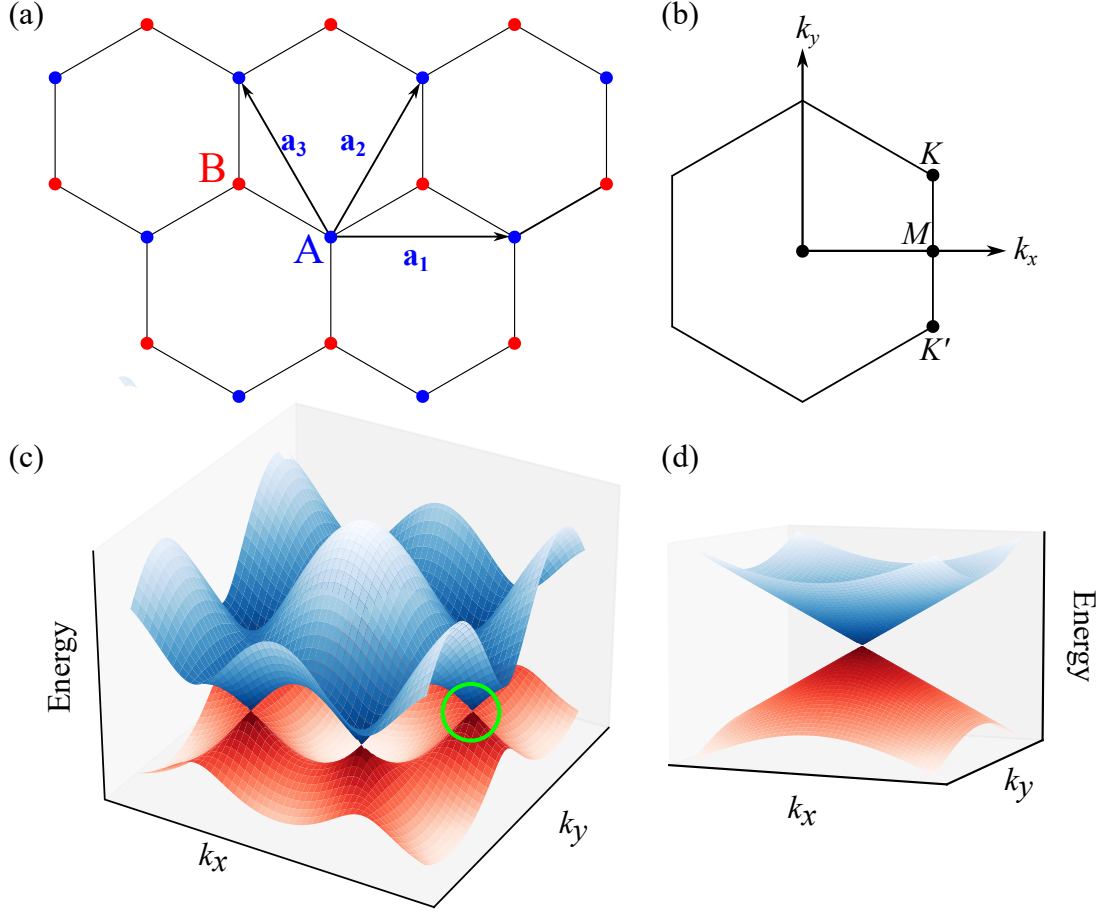


Figure 1.2: Graphene lattice and band structure. (a) The graphene lattice is made of two sublattices A (blue) and B (red). The vectors $\mathbf{a}_1$ and $\mathbf{a}_2$ are the basis vectors of the A lattice. The vector $\mathbf{a}_3 \equiv \mathbf{a}_2 - \mathbf{a}_1$ is used in the tight-binding model. (b) The first BZ of graphene. (c) The band structure of graphene calculated from Eqn. 1.24. The conduction and valence bands touch at K and K' points. (d) The low energy band structure around the green circled point in (c). This shows a linear dispersion without a bandgap.

where $\boldsymbol{\sigma}$ is the vector of Pauli spin operators and τ^z is a separate Pauli z-spin operator. This is two copies of the massless Dirac Hamiltonian [18]. Now that we know that the effective Hamiltonian for graphene is linear with respect to momentum, it is expected that quantum Hall in graphene will be different from quantum Hall in a 2D gas.

In the presence of an electromagnetic field, Eqn. 1.26 is modified using the same substitution as before, $\mathbf{p} \rightarrow \boldsymbol{\Pi} = \mathbf{p} - q\mathbf{A}$. Using a similar analysis as was used for the 2D gas in the

previous section [18, 19],

$$\epsilon_{\lambda,n} = \lambda \frac{\hbar v_F}{\ell_B} \sqrt{2n}. \quad (1.27)$$

There are two important differences from the result for the 2D gas. First, the LLs are not evenly spaced. Instead, the LL spacing decreases as the energy increases. Second, there is a zeroth LL which does not exist for the 2D gas. The zeroth LL is more complicated to understand, as it is occupied by both electrons and holes. The eigenstates are

$$\psi_{n=0,m}^{\xi} = \begin{pmatrix} 0 \\ |n=0, m\rangle \end{pmatrix} \quad (1.28)$$

and

$$\psi_{\lambda,n,m}^{\xi} = \frac{1}{\sqrt{2}} \begin{pmatrix} |n-1, m\rangle \\ \xi \lambda |n, m\rangle \end{pmatrix}, \quad (1.29)$$

where ξ is the valley index, Eqn. 1.28 is for the case where $n = 0$, and Eqn. 1.29 is for all other cases [18]. The states $|n, m\rangle$ are the same as for the 2D gas. The wavefunctions are analyzed in Section 2.9.2.

1.2.4 Integer quantum Hall effect

Before moving on from the introduction to quantum Hall, we should review the most famous manifestation of LLs, which is the quantized Hall conductance in the IQHE. As was first discovered by Thouless, Kohmoto, Nightingale, and den Nijs (TKNN), the quantized Hall conductance reveals the topological invariant of LLs below the Fermi level [13]. This result relies on

electrons in a periodic potential; thus we should start with the Bloch wavefunction:

$$\psi_{\mathbf{k}}^{\alpha}(\mathbf{x}) = e^{i\mathbf{k}\cdot\mathbf{x}} u_{\mathbf{k}}^{\alpha}(\mathbf{x}), \quad (1.30)$$

where $u_{\mathbf{k}}^{\alpha}(\mathbf{x})$ is the periodic part of the wavefunction on a unit cell within band α . The wavefunctions satisfy Schrödinger's equation, $H|\psi_{\mathbf{k}}^{\alpha}\rangle = E_{\mathbf{k}}|\psi_{\mathbf{k}}^{\alpha}\rangle$. By inserting Eqn. 1.30, we obtain

$$\tilde{H}|u_{\mathbf{k}}^{\alpha}\rangle = E_{\mathbf{k}}|u_{\mathbf{k}}^{\alpha}\rangle, \quad (1.31)$$

where $\tilde{H} = e^{-i\mathbf{k}\cdot\mathbf{x}} H e^{i\mathbf{k}\cdot\mathbf{x}}$. The Hall conductivity can now be calculated using the Kubo formula.

At a low frequency ($\omega \rightarrow 0$), the Kubo formula is given by

$$\sigma_{xy} = i\hbar \sum_{E_{\alpha} < E_F < E_{\beta}} \int_{\text{BZ}} \frac{d^2\mathbf{k}}{(2\pi)^2} \frac{\langle u_{\mathbf{k}}^{\alpha} | J_y | u_{\mathbf{k}}^{\beta} \rangle \langle u_{\mathbf{k}}^{\beta} | J_x | u_{\mathbf{k}}^{\alpha} \rangle - \langle u_{\mathbf{k}}^{\alpha} | J_x | u_{\mathbf{k}}^{\beta} \rangle \langle u_{\mathbf{k}}^{\beta} | J_y | u_{\mathbf{k}}^{\alpha} \rangle}{(E_{\beta}(\mathbf{k}) - E_{\alpha}(\mathbf{k}))^2}, \quad (1.32)$$

where the index α runs over the filled bands and β runs over the unfilled bands [14]. The current operator, $\mathbf{J}$ is defined with the group velocity,

$$\mathbf{J} = \frac{e}{\hbar} \frac{\partial \tilde{H}}{\partial \mathbf{k}}. \quad (1.33)$$

To use Eqn. 1.33 to simplify Eqn. 1.32 note that

$$\begin{aligned}
\langle u_{\mathbf{k}}^{\alpha} | \frac{\partial}{\partial k_i} \tilde{H} | u_{\mathbf{k}}^{\beta} \rangle &= \langle u_{\mathbf{k}}^{\alpha} | \frac{\partial}{\partial k_i} \left(\tilde{H} | u_{\mathbf{k}}^{\beta} \rangle \right) - \langle u_{\mathbf{k}}^{\alpha} | \tilde{H} | \frac{\partial}{\partial k_i} u_{\mathbf{k}}^{\beta} \rangle \\
&= (E_{\beta}(\mathbf{k}) - E_{\alpha}(\mathbf{k})) \langle u_{\mathbf{k}}^{\alpha} | \frac{\partial}{\partial k_i} u_{\mathbf{k}}^{\beta} \rangle \\
&= - (E_{\beta}(\mathbf{k}) - E_{\alpha}(\mathbf{k})) \langle \frac{\partial}{\partial k_i} u_{\mathbf{k}}^{\alpha} | u_{\mathbf{k}}^{\beta} \rangle.
\end{aligned}$$

Substituting this into Eqn. 1.32,

$$\begin{aligned}
\sigma_{xy} &= \frac{ie^2}{\hbar} \sum_{E_{\alpha} < E_F < E_{\beta}} \int_{\text{BZ}} \frac{d^2 \mathbf{k}}{(2\pi)^2} \langle \frac{\partial}{\partial k_y} u_{\mathbf{k}}^{\alpha} | u_{\mathbf{k}}^{\beta} \rangle \langle u_{\mathbf{k}}^{\beta} | \frac{\partial}{\partial k_x} u_{\mathbf{k}}^{\alpha} \rangle - \langle \frac{\partial}{\partial k_x} u_{\mathbf{k}}^{\alpha} | u_{\mathbf{k}}^{\beta} \rangle \langle u_{\mathbf{k}}^{\beta} | \frac{\partial}{\partial k_y} u_{\mathbf{k}}^{\alpha} \rangle \\
&= \frac{ie^2}{2\pi\hbar} \sum_{\alpha} \int_{\text{BZ}} d^2 \mathbf{k} \langle \frac{\partial}{\partial k_y} u_{\mathbf{k}}^{\alpha} | \frac{\partial}{\partial k_x} u_{\mathbf{k}}^{\alpha} \rangle - \langle \frac{\partial}{\partial k_x} u_{\mathbf{k}}^{\alpha} | \frac{\partial}{\partial k_y} u_{\mathbf{k}}^{\alpha} \rangle \\
&= -\frac{e^2}{2\pi\hbar} \sum_{\alpha} \int_{\text{BZ}} d^2 \mathbf{k} \nabla_{\mathbf{k}} \times \mathcal{A}_{\mathbf{k}}^{\alpha} \\
&= -\frac{e^2}{2\pi\hbar} \sum_{\alpha} \oint \mathcal{A}_{\mathbf{k}}^{\alpha} \\
&= \frac{e^2}{h} \sum_{\alpha} \mathcal{C}_{\alpha}.
\end{aligned} \tag{1.34}$$

Here, $\mathcal{A}_{\mathbf{k}}^{\alpha} = -i \langle u_{\mathbf{k}}^{\alpha} | \frac{\partial}{\partial \mathbf{k}} | u_{\mathbf{k}}^{\alpha} \rangle$ is the Berry connection and $\mathcal{C}_{\alpha}$ is the Chern number, which is an integer. From this we can see that the Hall conductivity is given by the total Chern number of the filled bands below the Fermi level. This gives us Eqn. 1.1 where ν is an integer.

The IQHE was first demonstrated in graphene in 2005 [20]. The quantized Hall conductance is slightly different. In graphene ν can only take values $2(2n+1)$, where n is an integer. This is due to a combination of spin and valley degeneracy, as well as particle-hole symmetry [18].

1.3 Chiral Light

The other piece of this dissertation is chiral light. The most common case of chiral light is circular polarization. From the perspective of a classical wave, a beam of circularly polarized light features an electric field that rotates as the beam propagates. Left- and right-handed circular polarizations rotate in the opposite direction and the two are related via time-reversal symmetry. Circularly polarized light has also been shown to carry an angular momentum $\pm\hbar$, which can be transferred to matter [21]. In the quantum theory of light, this angular momentum corresponds to the spin angular momentum (SAM) of the photons [22]. However, this is not the only form of chirality that light can take. By solving the wave equation within the paraxial approximation, it can be shown that, more generally, light can carry a non-zero phase twist which is associated with orbital angular momentum (OAM). Light that carries finite OAM is often referred to as vortex light or twisted light as the light twists around the axis of propagation, and the spatial profile of the beams contain vortexes.

1.3.1 Basic theory

To begin with, we start with Maxwell's equations:

$$\nabla \cdot \mathbf{E} = \frac{\rho}{\varepsilon_0} \quad (1.35)$$

$$\nabla \times \mathbf{E} = -\frac{\partial \mathbf{B}}{\partial t} \quad (1.36)$$

$$\nabla \cdot \mathbf{B} = 0 \quad (1.37)$$

$$\nabla \times \mathbf{B} = \mu_0 \mathbf{J} + \frac{1}{c^2} \frac{\partial \mathbf{E}}{\partial t}, \quad (1.38)$$

where $\mathbf{E}$ is the electric field, $\mathbf{B}$ is the magnetic field, ε_0 is the permittivity of free space, and μ_0 the permeability of free space. The electric and magnetic fields can also be associated with the electric scalar potential, ϕ , and the magnetic vector potential, $\mathbf{A}$,

$$\mathbf{B} = \nabla \times \mathbf{A} \quad (1.39)$$

$$\mathbf{E} = -\nabla\phi - \frac{\partial\mathbf{A}}{\partial t}. \quad (1.40)$$

With these potentials in combination with Maxwell's equations, equations for the potentials can be derived. In the Lorenz gauge, $\nabla \cdot \mathbf{A} = -\frac{1}{c^2} \frac{\partial\phi}{\partial t}$, the equations are:

$$\nabla^2 \mathbf{A} - \frac{1}{c^2} \frac{\partial}{\partial t^2} \mathbf{A} = -\frac{\rho}{\varepsilon_0} \quad (1.41)$$

$$\nabla^2 \phi - \frac{1}{c^2} \frac{\partial}{\partial t^2} \phi = -\mu_0 \mathbf{J}. \quad (1.42)$$

Since the rest of this discussion will be in vacuum, we can set $\rho = 0$ and $\mathbf{J} = 0$.

In geometric optics, light is analyzed using rays. If these rays have little dispersion from the propagation vector, then the paraxial approximation can be applied. A paraxial wave can be described with the vector potential $\mathbf{A} = \mathbf{A}(\mathbf{r})e^{i(\omega t - kz)}$. If we insert this into Eq. 1.41, then we obtain

$$\nabla^2 \mathbf{A}(\mathbf{r}) - 2ik \frac{\partial}{\partial z} \mathbf{A}(\mathbf{r}) = 0. \quad (1.43)$$

The assumption of the paraxial approximation is that the field changes along the propagation axis (z-axis) are much smaller than the wavelength of the light. Stated mathematically: $|k\mathbf{A}(\mathbf{r})| \gg$

$|\partial/\partial z \mathbf{A}(\mathbf{r})|$. Applying this to Eq. 1.43, we obtain the approximation

$$\nabla_{\perp}^2 \mathbf{A}(\mathbf{r}) - 2ik \frac{\partial}{\partial z} \mathbf{A}(\mathbf{r}) \approx 0, \quad (1.44)$$

where $\nabla_{\perp}^2$ is the transverse Laplacian operator. This is the paraxial wave equation. Typical solutions are Hermite-Gaussian beams which are best for Cartesian coordinates and Laguerre-Gaussian (LG) beams which are best for cylindrical coordinates as their intensity has azimuthal symmetry [22].

Solutions to Eqn. 1.44 can be written in the form

$$\mathbf{A}(\mathbf{r}, t) = \mathbf{A}_0 u(\mathbf{r}) e^{i(kz - \omega t)} + \text{c.c.}, \quad (1.45)$$

where $\mathbf{A}_0$ is a two-dimensional constant vector and $u(\mathbf{r})$ is a scalar mode function which describes the transverse spatial profile [22]. An important feature of this vector potential is that the polarization is described by $\mathbf{A}_0$, which is independent of $u(\mathbf{r})$. In polar coordinates, the LG modes can be written as

$$\begin{aligned} u(\mathbf{r}) = & \sqrt{\frac{2p!}{\pi(p+|\ell|)!}} \frac{1}{w(z)} \left(\frac{r\sqrt{2}}{w(z)} \right)^{|\ell|} e^{i\ell\theta} e^{i\Psi(z)} L_p^{|\ell|} \left(\frac{2r^2}{w^2(z)} \right) \\ & \times \exp \left(\frac{-r^2}{w^2(z)} \right) \exp \left(-ik \frac{r^2}{2R(z)} \right), \end{aligned} \quad (1.46)$$

where $w(z) = w_0 \sqrt{1 + (z/z_R)^2}$ is the beam radius, $R(z) = z(1 + (z_R/z)^2)$ is the radius of curvature of the wave front, $\Psi(z) = -(|\ell| + 2p + 1) \arctan(z/z_R)$ is the Guoy phase, $L_p^{|\ell|}$ is a generalized Laguerre polynomial, w_0 is the beam waist, and $z_R = kw_0^2/2$ is the Rayleigh range [22].

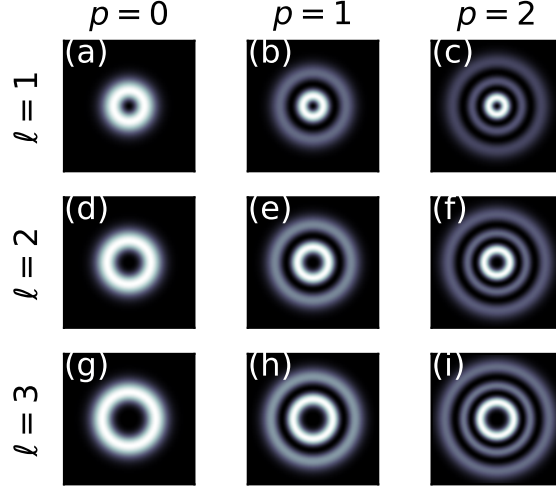


Figure 1.3: LG intensity profiles. (a)-(c) have $\ell = 1$, (d)-(f) have $\ell = 2$, and (g)-(i) have $\ell = 3$. Each column has different radial modes $p = 0$, $p = 1$, and $p = 2$.

Select intensity profiles of these LG beams for $z = 0$ are plotted in Fig. 1.3. Importantly, the value ℓ does not just change the intensity profile. It also changes the phase, and while the intensity profiles for beams with opposite values of ℓ are the same, their phase twists in the opposite direction with the number of times the phase winds around the axis being $|\ell|$, see Fig. 1.4. In analogy to circular polarization, where the electric field rotates in a specific direction, in LG modes the phase twists in a specific direction dictated by the sign of ℓ . LG modes have also been shown to have a well-defined OAM equal to $\ell\hbar$ per photon [4–6]. Hence, this is another source of chirality for light. In the next chapter, we discuss an experiment we conducted in which these vortex beams interact with quantum Hall graphene.

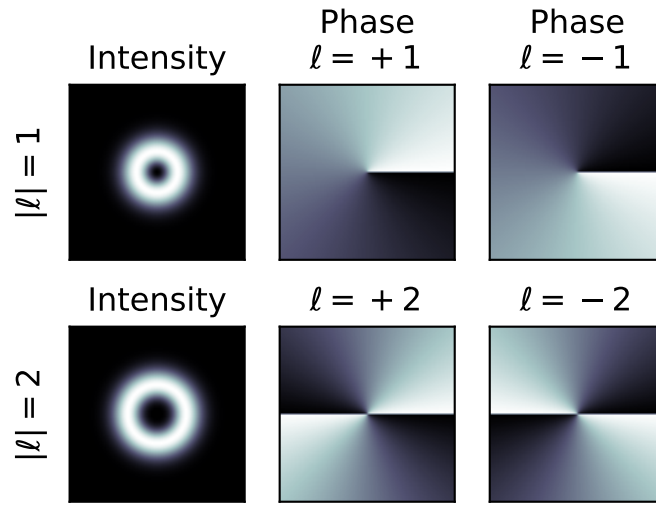


Figure 1.4: LG phase profiles. The first row shows the intensity profile for an LG mode with $|\ell| = 1$ and the phase profiles for $\ell = +1$ and $\ell = -1$. The second row shows the intensity profile for an LG mode with $|\ell| = 2$ and the phase profiles for $\ell = +2$ and $\ell = -2$. All plots are made with $z = 0$.

Chapter 2: Optical pumping of electronic quantum Hall states with vortex light

2.1 Introduction

Coherent manipulation of light-matter hybrids plays a crucial role in advancing future quantum technologies and optoelectronics [8, 9]. Particularly desirable is the control over the spatial degree of freedom in light-matter interactions. Typically, due to the presence of disorder or Coulomb binding, electronic wavefunctions are much more spatially confined than the wavelengths of associated optical transitions. Consequently, the light-matter interaction occurs locally, and neither the spatial profile of the optical field nor the spatial extent of the electron wavefunction has a significant influence on these interactions, a regime known as the dipole approximation. In other words, in this regime only direct optical transitions are accessible, and the transfer of linear and angular momentum, which enables optical control of the spatial degrees of electrons, is not possible.

To understand this, one can consider a simplified model of a hydrogen-like atom, where the typical Bohr radius (a_B) is much smaller than the corresponding optical transition wavelength (λ). Therefore, the next-order quadrupole transition is weaker by a factor of $(a_B/\lambda)^2$ than the dipole transition, yet it is still observable in experiments [23]. One approach to enhance such effects is to shrink the wavelength of the electromagnetic field, which can be achieved by using plasmonic effects [24, 25]. Alternatively, if the electronic wave function is coherently extended

over the associated optical transition wavelength, and electrons are more itinerant than bound, then a gross violation of the dipole approximation is expected. A striking example is the quantum Hall system, where the electrons in two dimensions are subject to a strong out-of-plane magnetic field. Consequently, the kinetic energy is suppressed and electrons exhibit cyclotron motions with chiral characteristics that make it a promising system to investigate the interplay of the chirality of electrons and photons and the transfer of angular momentum in between [7, 11, 26–28].

In particular, there has been a growing interest in investigating chiral and topological effects in photonic systems and also light-matter hybrids [1–3, 28]. Such topological features can be either in the momentum domain and lead to Chern bands, or simply in the spatial degrees of freedom, such as optical vortex beams. Specifically, in addition to spin, in the form of polarization, light can also carry OAM [5, 6]. Such an OAM is quantized and given by $\hbar\ell$, where $\hbar$ is the Dirac constant and ℓ is the mode number which determines the phase winding of a vortex beam. The interaction of such vortex beams with materials has led to a plethora of exciting phenomena [22], such as the orbital photogalvanic effect [29].

In this work, we experimentally demonstrate the transfer of OAM to electrons in a quantum Hall graphene device with annular geometry using optical vortex beams. In particular, harnessing non-conventional optical selection rules of LLs described in Fig. 2.1c, we show a vorticity-selective light-matter interaction between twisted light and the electronic wavefunctions manifesting as a radial photocurrent (PC). We show that this radial PC only depends on the vorticity of light as a direct indication of spatially coherent light-matter interaction. We provide further evidence of the robustness of this mechanism by comparison with circularly polarized light. Specifically, we find that the PC contribution from OAM is at least one order of magnitude larger than the contribution of spin angular momentum (polarization), allowing us to confirm the significant

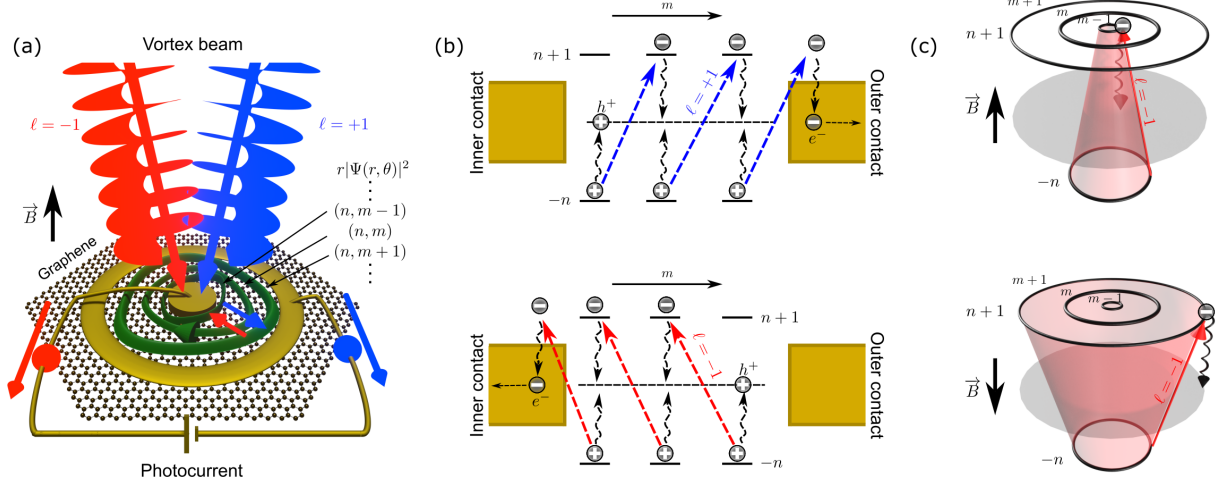


Figure 2.1: Concept of OAM pumping. (a) Schematic of LLs in the annular disc geometry subject to the vertically irradiated vortex beam. The green surfaces show representative states within a single LL. The electron distributions in the lowest LLs and directions of the induced radial PC are indicated by arrows. (b) Optical transitions and PC generation between inner and outer contacts using $\ell = +1$ (blue arrows) and $\ell = -1$ (red arrows) for σ^+ polarized light. For σ^- transitions see section 2.9.1. Light carrying OAM $\ell = +1$ ($\ell = -1$) increases (decreases) m and hence leads to expansion (shrinking) of the spatial extent of the electronic wavefunction. In this way, the shown scheme realizes an analog to optical pumping. (c) Schematic of the photoexcitation transitions between LLs with negative (red arrow) vortex beam in the presence of an upward (top) and downward (bottom) pointing magnetic field. In the top (bottom) panel, the magnetic field is anti-parallel (parallel) to the helicity of light, leading to the shrinking (expansion) of the wavefunction radius. In (c), only the electron relaxation is considered.

role of the beam's spatial topology, and its ability to control the spatial degree of electrons.

2.2 OAM pumping of electrons

To present the motivation and a basic understanding of our experiment, we discuss spatially-dependent light-matter interactions that can manipulate the spatial degrees of freedom of electrons within a quantum Hall system. In particular, using a LL picture, we observe how the transfer of OAM from photons to electrons results in a radial current, where the direction of the current is determined by the vorticity of the light. As shown in Fig. 2.1, we consider optical transitions between two LLs, in the presence of rotational symmetry perpendicular to the plane of the quantum

Hall sample. In this scenario, the sample is irradiated by an optical vortex where each photon is carrying an OAM $\hbar\ell$ [7, 26].

During the excitation process, the OAM of $\hbar\ell$ is transferred to electrons [7, 26]. As the radius of the electronic wavefunction increases monotonically with angular momentum, this optical transfer of angular momentum causes a radial change in the electronic wavefunction, which is solely determined by the vorticity of the light. The subsequent relaxation process conserves OAM on average and therefore maintains the OAM transfer from the original excitation. This concept is in direct analogy to optical pumping in atomic systems, wherein cyclical pumping among different hyperfine states of bound electrons within an atom transfers them to a specific quantum state [30].

Note that despite the absence of rotational symmetry in the presence of disorder, the optical pumping model continues to hold true [7]. Moreover, the optical pumping picture also provides a simple estimate of the resulting PC: assuming that OAM pumping was the only mechanism of charge transport, the OAM needed to carry one electron through the sample equals the number M of orbitals in a LL, which is given by $M = A/(\pi l_B^2)$, where A is the area of the sample and l_B is the magnetic length. The estimated transported charge, δQ , during the time interval δt by N_{ph} photons, is $\delta Q = eN_{\text{ph}}\ell/M$. Therefore, for photons with energy $\hbar\omega$, the PC obtained from the laser power $P = \hbar\omega \times N_{\text{ph}}/\delta t$ is $I = \delta Q/\delta t = eP\ell/(\hbar\omega M)$. This picture also implies that, upon inverting the direction of the magnetic field, the PC changes direction, as illustrated in Fig. 2.1c.

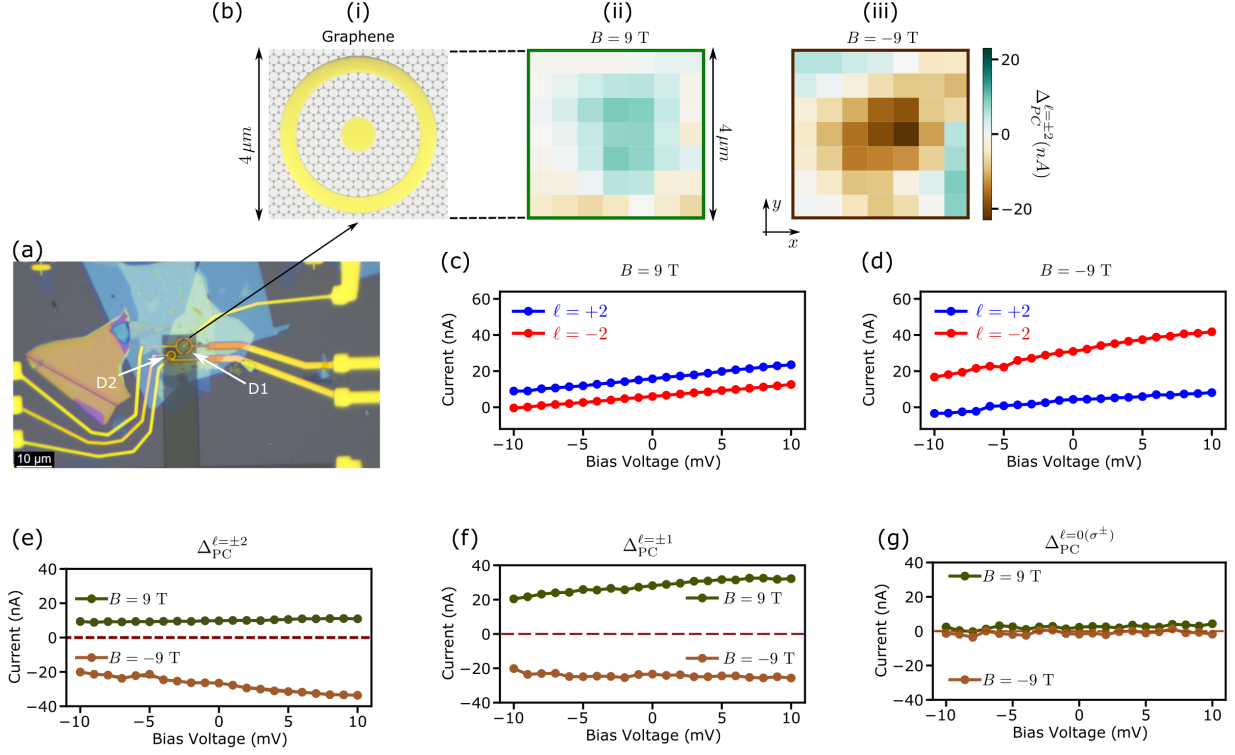


Figure 2.2: OAM-selective PC generation. (a) Optical-microscope image of the sample, showing two Corbino devices labeled D1 and D2, one with a smaller diameter ($\sim 2 \mu\text{m}$) and the other with a larger diameter ($\sim 4 \mu\text{m}$). D1 is the device used for the data in this chapter, for which the inner and outer contacts are marked. The gray rectangle shows the metallic back gate. (b) (i) Sample schematic, (ii-iii) Spatially-resolved PC difference Δ_{PC} for $\ell = \pm 2$, at $B = 9 \text{ T}$ and $B = -9 \text{ T}$, respectively. Bias voltage V_b for both cases was -4 mV . (c), (d) Measured PC as a function of bias voltage V_b generated using light carrying $\ell = +2$ (blue) and $\ell = -2$ (red) at (c) $B = 9 \text{ T}$ and (d) $B = -9 \text{ T}$. (e-g) Shows the Δ_{PC} for $B = 9 \text{ T}$ (green) and $B = -9 \text{ T}$ (brown) for $\ell = \pm 2, \pm 1, 0$, respectively. In (g), at each magnetic field, the Δ_{PC} is calculated by subtracting the PC generated using two Gaussian beams with opposite circular polarization ($\sigma^\pm$). In all panels, the gate voltage V_g and the average pump power were 1.78 V and $10 \mu\text{W}$, respectively.

2.3 Bias voltage dependence of PC

To experimentally demonstrate this mechanism, we use a device consisting of a hexagonal boron nitride (hBN) encapsulated monolayer graphene in an annular (Corbino) geometry as shown in Fig. 2.2a. The inner and outer contacts are used to apply an in-plane electric field and also measure the generated PC, while the back-gate voltage V_g controls the Fermi level. We apply an out-of-plane external magnetic field up to 9 T at 4.2 K to be in the quantum Hall regime. The optical vortex beams of different vortices ℓ are generated by a spatial light modulator (SLM) and are concentrically focused on the Corbino device (see sections 2.7.1 and 2.7.3 for the sample and optical setup). This enables us to excite the carriers in our device which undergo vorticity-selective optical transitions, shown in Fig. 2.1c. For all the measurements, we choose $V_g = 1.78$ V, which sets the Fermi energy near filling factor $\nu = 6$ (see Fig. 2.4 and associated text for discussion about this choice).

The generated PC for various optical vortices $\pm\ell$ are independently measured, while the beam is spatially scanned over the sample. Fig. 2.2b shows PC difference (subtracted), Δ_{PC} , where the vortex beam $\ell = \pm 2$ is spatially scanned over the sample (shown schematically in (i)), at (ii) $B = 9$ T and (iii) $B = -9$ T. It can be clearly observed that the Δ_{PC} flips sign when the magnetic field direction is reversed. This remarkable observation corroborates with the earlier optical pumping picture of Fig. 2.1a, where depending on the vortices of the optical beam the radial extent of the electrons either shrink or expand during the optical excitation. Note that the sign of the observed PC difference depends solely on the phase winding of the optical vortex beam, and not the intensity; therefore, one can not associate this (beyond the dipole-approximation) process with heating.

It is unlikely that any sample has pristine electrical conditions and it may harbor residual or intrinsic in-plane potential. Such inherent potential could potentially explain the presence of a radial PC in the Corbino sample. In order to rule out the origin of our observed effect to such an in-plane electric field, we apply a bias voltage V_b between the inner and outer contacts to create a controllable potential gradient in the radial direction. Figures 2.2c-d show the measured PC as a function of V_b for $B = 9$ T and $B = -9$ T, respectively. Remarkably, we observe multiple unambiguous signatures of the vorticity-selective light-matter interaction. First, as shown in Fig. 2.2c-d, for $\ell = +2$ (blue) and $\ell = -2$ (red) at each magnetic field, we observe a consistent and significant difference in the generated PC for a wide range of V_b ($-10 \leq V_b \leq 10$ mV). In other words, radially tilting the electric potential can change the total PC, however, the PC difference Δ_{PC} remains relatively constant. Second, for the opposite magnetic field, we observe a clear sign flip for the PC difference (Fig. 2.2e). Specifically, since the OAM is defined relative to the magnetic field, inverting the latter effectively inverts the OAM and should therefore lead to a sign-change of the observed radial PC, which is clearly observed in Fig. 2.2e. In an ideal case, the amplitude of this flipped current should be the same, however, due to slightly different spatial alignment for different magnetic fields, the magnitude of the PC is different (See Fig. 2.2b (ii) and (iii)). Third, to investigate the effect of the degree of vorticity on the generated PC, we illuminate the sample with $\ell = \pm 1$ beams. As shown in Fig. 2.2, robust PC difference $\Delta_{PC}^{\ell=\pm 1}$ is observed across a wide range voltage bias, and the sign reversal with the magnetic field is present. For a large sample subject to the optical vortex, one expects that Δ_{PC} to increase with the vorticity degree ℓ [7]. However, since our sample and optical vortex spatial profile is comparable, and in particular, the spatial overlap for $\ell = \pm 2$ is smaller than that of $\ell = \pm 1$, we observe a reduced Δ_{PC} for the larger value of $|\ell|$. Fourth, in order to rule out the origin of Δ_{PC} based on circular

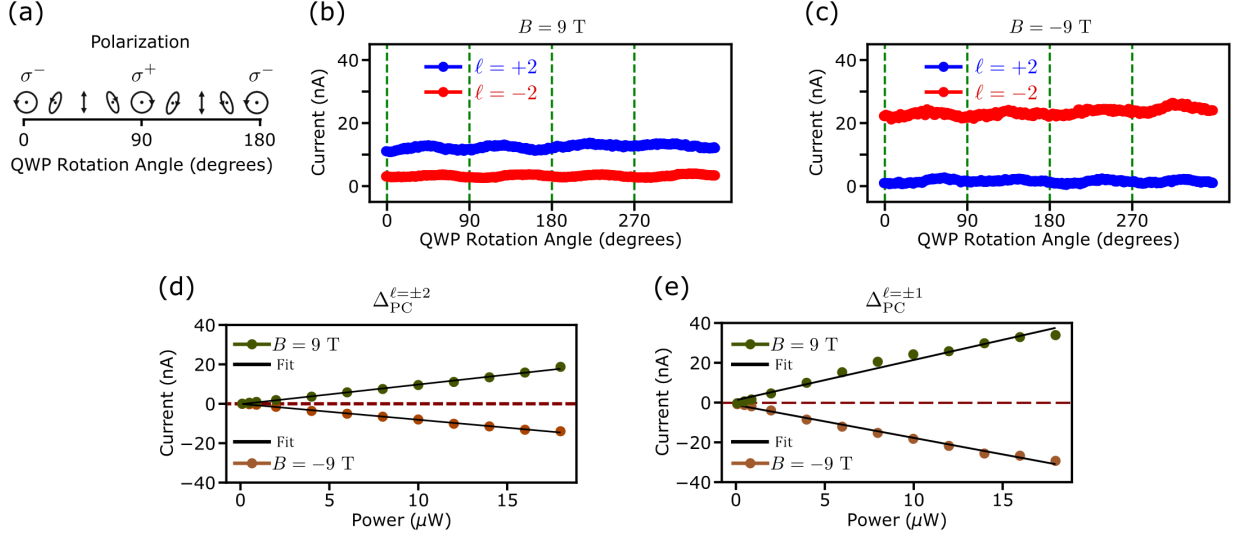


Figure 2.3: Polarization-resolved and power-dependent PC measurements. (a) polarization of the pump beam as a function of the QWP rotation, shown schematically at the top. σ^+ and σ^- denote positive and negative-circular polarization. Between each consecutive circular polarization, continuous rotation of the QWP changes the polarization periodically from circular to elliptical, to linear polarization. PC measured as a function of QWP rotation using light carrying $\ell = +2$ (blue) and $\ell = -2$ (red) at (b) $B = 9$ T and (c) $B = -9$ T, respectively. The green vertical dashed lines indicate the consecutive σ^+ and σ^- polarization. All measurements are done at a fixed $V_g = 1.78$ V, $V_b = -4$ mV, and average pump-power of 10μ W. (d), (e) Pump-power-dependence of PC: PC difference Δ_{PC} measured as a function of average pump-power using light carrying (d) $\ell = \pm 2$ and (e) $\ell = \pm 1$ OAM at $B = 9$ T (green circles) and $B = -9$ T (brown circles), respectively. The black lines show the least-squares linear fit. In all of the data presented in this chapter, the average pump power was set to 10μ W, at which a linear behavior is observed.

polarization, the sample is illuminated with $\ell = 0$ beams consisting of two Gaussian beams with opposite circular polarization (σ^+ and σ^-). As shown in Fig. 2.2g, the PC difference for different circular polarization and $\ell = 0$ is at least an order of magnitude smaller than the non-zero ℓ cases. Therefore, we associate the non-zero radial current with the OAM of light, rather than the spin angular momentum.

2.4 Polarization dependence of PC

To further decouple the role of the polarization of the optical field from OAM in our experiments, we perform polarization-resolved PC generation using OAM of light by using a variable quarter-wave plate (QWP) in the excitation path. We rotate the QWP such that the polarization continuously changes from linear to elliptical to circular while measuring the PC (Fig. 2.3a). The measured PC for $\ell = +2$ (blue) and $\ell = -2$ (red) at $B = 9$ T and $B = -9$ T is shown in Fig. 2.3b-c.

Here, there are three clear observations confirming the robustness of OAM-induced PC generation. First, the OAM-induced PC difference is almost an order of magnitude larger than the amplitude of the current oscillations induced by QWP rotation. Second, the OAM-induced PC difference never changes sign as a function of QWP rotation, further confirming the domination of the observed vorticity-selective PC. Finally, this measurement sheds light on the role of focusing the optical beam on its polarization properties. This is crucial since in our measurements, we focus the beam onto the sample using an aspheric lens with a numerical aperture (NA) of 0.68. This large NA may distort the polarization beyond the paraxial approximation. However, Fig. 2.3b-c shows that this distortion of polarization is relatively insignificant and it does not affect the validity of our results.

2.5 Power dependence of PC

In order to verify the linear power dependence of the OAM transfer we also investigate the power dependence of the OAM-induced PC in our measurements as a function of optical

pump power by varying the average power of our excitation beam within the range 0.1 to 18 μW . Figures 2.3d-e show that the generated PC increases linearly with the pump intensity. From the above optical pumping estimate, we have $l_B \approx 26 \text{ nm}/\sqrt{B/(1 \text{ tesla})} \approx 9 \text{ nm}$, and $M = A/(\pi l_B^2) \approx 3 \times 10^4$. The PC is estimated to be $I \approx (0.1 \text{ nA}/\mu\text{W})P$. Interestingly, the slope of the experimental power dependence ($\sim 1 \text{ nA}/\mu\text{W}$) significantly exceeds this estimate. As we discuss in more detail below, the PC signal also exhibits a strong gate voltage dependence due to relaxation effects, and the experimental values are taken at local PC difference maxima, where significant carrier multiplication can be expected [31]. In addition, the donut-shaped intensity profile of the light, not taken into account in the estimate, enhances the current flow into the outer contact. Additionally, the OAM-induced PC differences increase linearly with pump power which indicates that our experiment was performed in the linear regime away from the Pauli-blockade in Ref. [7].

2.6 Gate voltage dependence of PC

Next, we investigate the role of the Fermi energy (E_F) in the generated vorticity-selective PC. By changing the gate voltage V_g , we tune E_F between LLs, that is, we tune the LL filling factor ν . As shown in Fig. 2.4b, the subtracted PC (for $\ell = \pm 2$) changes sign as a function of V_g , with the direction of PC changing twice between each consecutive conductance peak. In Fig. 2.4b, green (brown) denotes the subtracted PC at $B = 9 \text{ T}$ ($B = -9 \text{ T}$), respectively. Recently, an effect known as the “bottleneck effect”, was developed to describe the observation of a similar PC direction change as a function of V_g in a rectangular geometry [31]. This picture describes this V_g -dependent sign based on the relative position of the electrons and holes com-

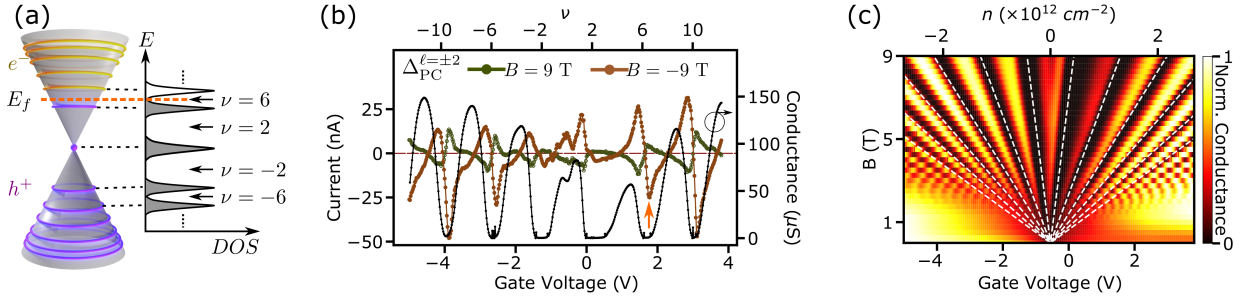


Figure 2.4: Gate voltage dependence of PC. (a) Schematic of graphene's band structure near the Dirac point with LLs shown in yellow and purple for the electron and holes, respectively. The inset shows the density of states (DOS) as a function of energy with the Fermi energy. (b) Measured PC difference as a function of gate voltage using light carrying $\ell = \pm 2$ at $B = 9$ T (green) and $B = -9$ T (brown), respectively. The top x-axis shows the calculated integer filling factors (see section 2.7.2). The two-terminal longitudinal conductance measured at $B = 9$ T (black circles) is also shown. This measurement was done at $V_b = -4$ mV, and an average pump-power of $10 \mu\text{W}$. (c) Landau fan measured via magneto-transport as a function of the magnetic field, gate voltage (carrier density marked on the top x-axis). The conductance is normalized for each magnetic field value. The white dashed lines denote integer filling factors calculated from the conversion of gate voltage to carrier density (see section 2.7.2).

pared to the Fermi energy and their propagation through the edge states. However, in contrast to graphene in a rectangular geometry, in our Corbino device, edge states do not contribute to the measured PC. Therefore, our measurements are only sensitive to bulk physics.

2.7 Methods

2.7.1 Device fabrication

Graphene was exfoliated from natural graphite crystals (HQ Graphene) and hBN was exfoliated from lab-grown [32] or commercial (HQ Graphene) synthetic crystals. Monolayer graphene and hBN were identified based on their color contrast with an optical microscope, while the thickness of the hBN layers used as a gate insulator was measured by atomic force microscopy. A hot pickup technique was used to stack the hBN/graphene/hBN heterostructure,

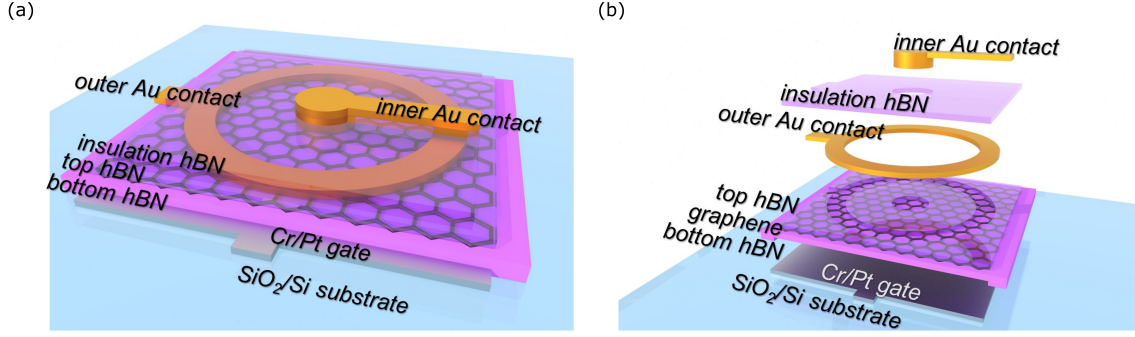


Figure 2.5: Sample fabrication. (a) Schematic of the graphene Corbino sample. (b) Exploded view of (a).

which was then transferred onto a pre-patterned local metallic back-gate made of 3 nm of Cr and 2 nm of Pt [33]. The area for the outer contact was etched using a selective reactive ion etching which was followed by the evaporation of 50 nm of Au to form the outer contact [34]. Next, a third layer of hBN was dropped on top of the heterostructure to act as an insulating layer between the overlapping parts of the contacts. The same selective etch was used to expose the area for the inner contact and 100 nm of Au was evaporated to make the contact. This process is illustrated in Fig. 2.5. The fabricated devices were wire-bonded to chip carriers for the electrical and PC measurements.

2.7.2 Transport measurements

Transport measurements are carried out with low-frequency lock-in techniques either by biasing with a current of 20 nA or with a voltage at a frequency of 13 Hz. Gate voltage sweeping is performed using a DC source measure unit (Keithley 2450). The two-terminal longitudinal conductance for various magnetic fields was measured to obtain the Landau fan; see Fig. 2.4c. These measurements are performed using the circuits depicted in Fig. 2.7a-b.

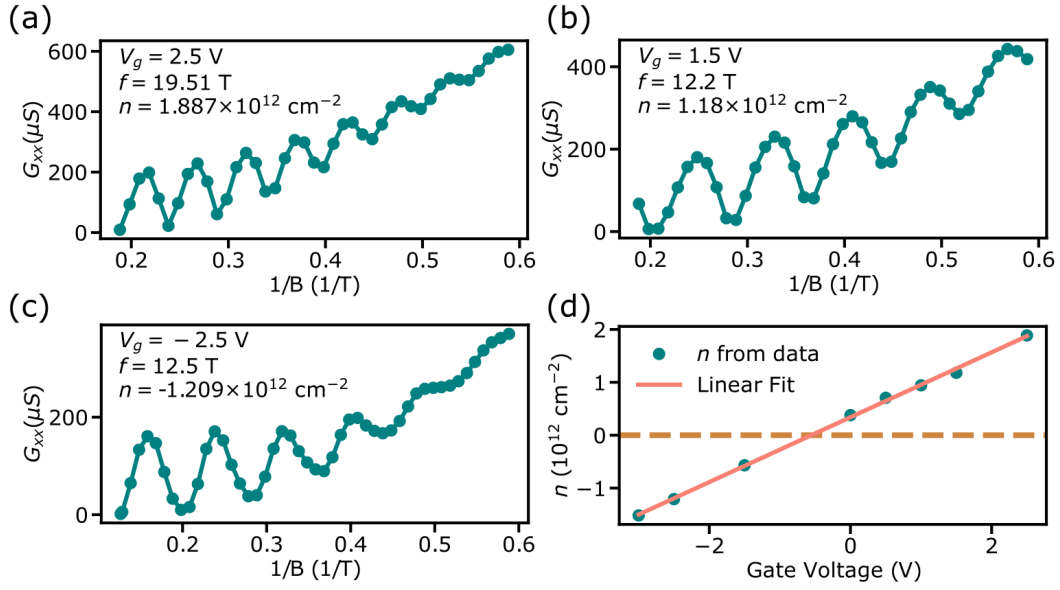


Figure 2.6: Shubnikov-de Haas oscillation measurement. Longitudinal conductance measured as a function of $1/B$ at gate voltages: (a) 2.5 V, (b), 1.5 V, and (c) -2.5 V. Using a Fourier transform to extract the frequency, the carrier densities were calculated to be (a) $1.887 \times 10^{12} \text{ cm}^{-2}$, (b) $1.18 \times 10^{12} \text{ cm}^{-2}$, and (c) $-1.209 \times 10^{12} \text{ cm}^{-2}$. (d) A linear fit to the extracted carrier densities as a function of gate voltage. The slope of the least-squares linear fit is $(0.614 \pm 0.009) \times 10^{12} \text{ cm}^{-2} \text{ V}^{-1}$ and the y-intercept is $(0.34 \pm 0.02) \times 10^{12} \text{ cm}^{-2}$.

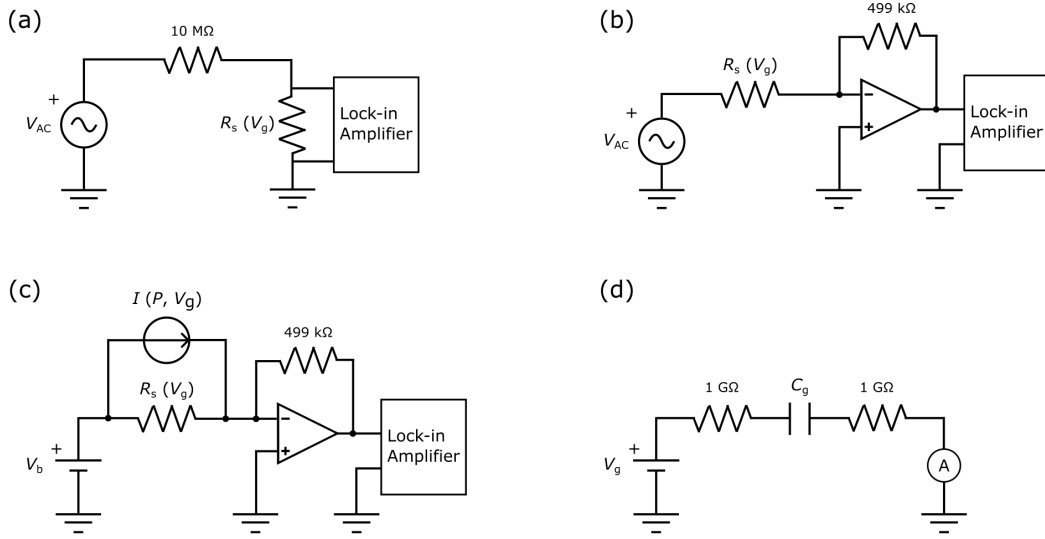


Figure 2.7: Electronic measurement circuits. (a) The current bias circuit used for transport measurements. A sinusoidal voltage is provided by the lock-in amplifier. The constant resistor ($10\text{ M}\Omega$) is in series with the sample R_S . This large resistor is used to current bias the sample. The voltage across the sample is measured through the inputs of the lock-in amplifier. (b) The voltage bias circuit is alternatively used for transport measurements. We use the same source as in (a), however, there is no large resistor to convert the voltage signal to current. In this case, the resulting current from the sample is passed through a trans-impedance amplifier (TIA), and the resulting voltage is measured with the lock-in amplifier. (c) The effective circuit for measuring PC. In this circuit, the sample is modeled as a current source, $I(P, V_g)$, in parallel with a resistor (sample resistance). The sample is in series with a DC voltage source. The TIA is in series with the sample and is connected to both terminals of the lock-in amplifier. (d) The effective circuit used to gate the sample: in this circuit, the sample is treated as a capacitor with capacitance C_g . It is in series with two $1\text{ G}\Omega$ resistors to decrease the current in the circuit.

To calibrate the conversion between gate voltage and carrier density, we measured Shubnikov-de Haas oscillations by setting the gate voltage and sweeping the magnetic field. The carrier density is extracted from the frequency of the oscillation (f) by $n = 4ef/h$ [35]. We extracted the carrier density for gate voltages between -3 V and 2.5 V and used a linear fit as a conversion between gate voltage and carrier density, see Fig. 2.6.

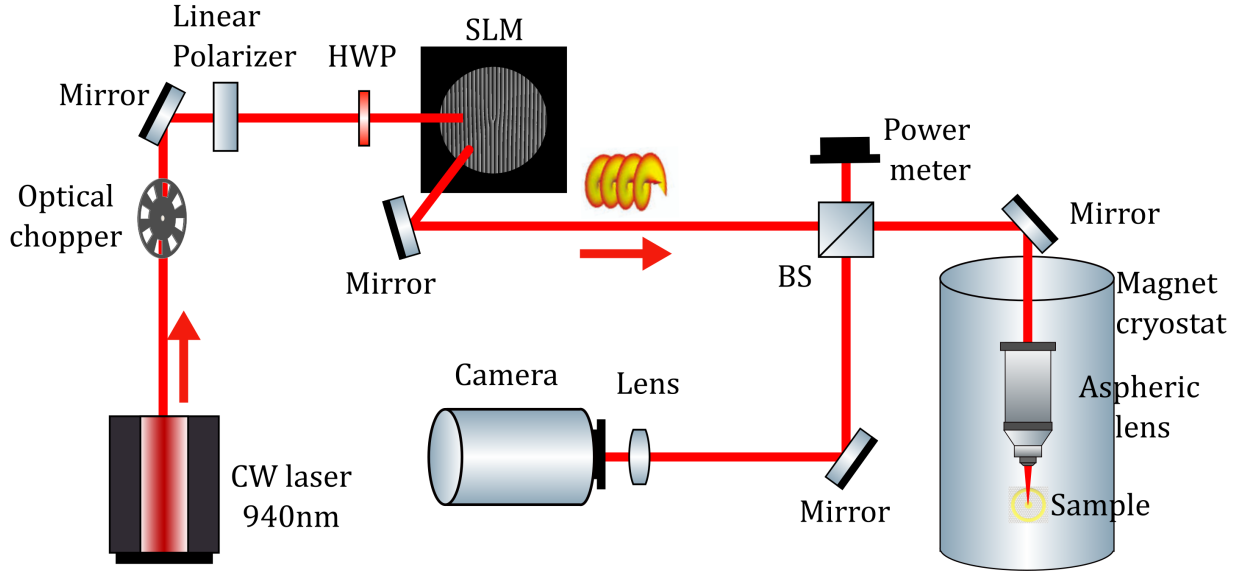


Figure 2.8: The optical setup used for the PC generation.

2.7.3 PC measurements

The sample, inside a variable temperature insert (VTI), is mounted on top of a piezo-electric stack (scanners (ANSxy100) and positioners (ANPx101, ANPz201)). It is cooled down to 4.2 K using liquid helium and can reach magnetic fields up to 9 T. The VTI has an optical window on top and a confocal microscope is built above to optically resolve the sample. The pump laser is illuminated through the same window and the laser spot's alignment to the sample can be monitored with the microscope. During measurements, the laser power is constantly monitored with a power meter right before the beam enters the optical window, and a feedback is given to a proportional-integral-derivative loop controlling a laser power control module. The power control module consists of a DC voltage applied to a Thorlabs electronic variable optical attenuator.

The two sample contacts are connected to a custom-built trans-impedance amplifier (TIA) outside of the cryostat [31]. The outputs of the TIA are connected to a lock-in amplifier (SRS

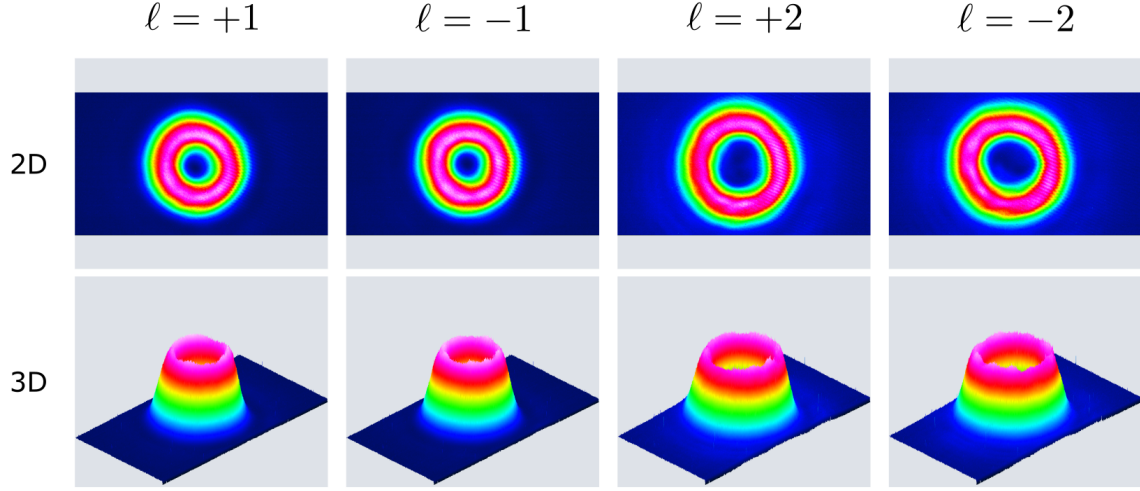


Figure 2.9: Beam profiles are imaged with a beam profiler for $\ell = +1$, $\ell = -1$, $\ell = +2$, and $\ell = -2$. Images are taken of the collimated beam diverted from the sample path.

SR860). The pump laser is chopped at a frequency of 308 Hz and the lock-in is frequency-locked to the chopper.

We sweep the gate in the same way as in the transport measurement. However, in the PC measurements, we apply a DC voltage bias across the sample using an SRS SIM928. The PC measurements are performed using the circuit depicted in Fig. 2.7c and the optical setup depicted in Fig. 2.8.

2.7.4 Generation and optimization of orbital angular momentum beams

To generate beams with orbital angular momentum (OAM) a Gaussian beam is diffracted off of a phase-only spatial light modulator (SLM). Ideally, the pattern displayed to achieve this would be

$$h(r, \theta) = \frac{1}{2\pi} \text{mod} \left(\ell\theta - \frac{2\pi}{D} r \cos(\theta), 2\pi \right). \quad (2.1)$$

This blazed hologram maximizes the diffraction efficiency [36, 37]. The obtained diffraction modes are a superposition of Laguerre-Gaussian (LG) modes with the same ℓ but various p which is the transverse index of the LG mode [38]. A Meadowlark optics 1920x1152 XY Phase series SLM is used. The performance of the pixels in the SLM is not homogeneous. The phase hologram can be modified to compensate for this. The phase hologram used in the experiment is

$$h(r, \theta) = \mathcal{M}(r, \theta) \bmod \left(\mathcal{F}(r, \theta) + \frac{2\pi}{D} r \cos(\theta), 2\pi \right), \quad (2.2)$$

where $\mathcal{M} = 1 + \frac{1}{\pi} \text{sinc}^{-1}(A(r, \theta))$ and $\mathcal{F} = \Phi - \pi\mathcal{M}$. The beam can be optimized by setting $\Phi = \ell\theta$ and manually adjusting the function $A(r, \theta)$ until a satisfactory beam is reached [39, 40]. The OAM beams after optimization are shown in Fig. 2.9.

To verify that the beams used in the experiment have the correct OAM, the beam's reflection off of the sample was interfered with a Gaussian reference beam. The results can be seen in Fig. 2.10. The interference patterns show the same pitchfork but in opposite directions, verifying that one has $\ell = +1$ and the other has $\ell = -1$.

2.7.5 Profile Intensity Comparison

To minimize the beam profile intensity mismatch effects in the PC measurements, beam optimization was performed to make the beams with opposite OAMs as similar as possible. However, naturally, the beams will not be perfectly identical, as can be seen in Fig. 2.9. This is important since differing beam profiles can cause a difference in generated PC. To quantify and demonstrate the insignificance of this effect in our experiments, the overlap of the beams is

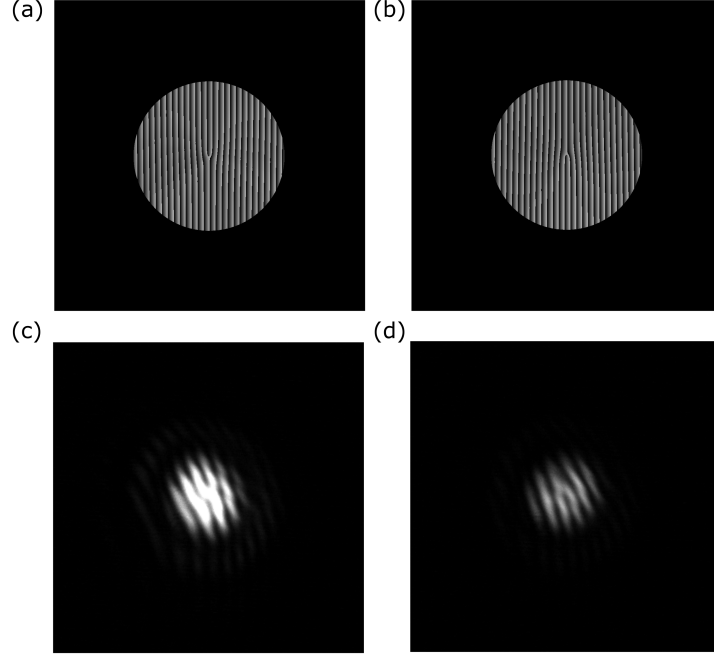


Figure 2.10: (a) Grating displayed on SLM to generate $\ell = +1$ as the first diffraction order. (b) The same as (a), but for $\ell = -1$. (c) Interference pattern when the $\ell = +1$ beam interferes with a Gaussian beam on the sample substrate. (d) The same as (c), but with $\ell = -1$.

calculated using the following function:

$$O[I(x, y), J(x, y)] = \frac{|\int \int I(x, y) J(x, y) dx dy|^2}{\int \int |I(x, y)|^2 dx dy \int \int |J(x, y)|^2 dx dy}, \quad (2.3)$$

where $I(x, y)$ and $J(x, y)$ are the two beam profiles with opposite OAMs. The maximum possible overlap is one. The calculated overlap for the $\ell = \pm 1$ and $\ell = \pm 2$ beams is 0.96 and 0.92 respectively. In other words, the $\ell = \pm 1$ beams have a 4% difference and the $\ell = \pm 2$ beams have an 8% intensity profile difference. The minimum percent difference between the average total current and the subtracted current for $\ell = \pm 2$ at $B = 9$ T is 60%. For $B = -9$ T it is 134%. For $\ell = \pm 1$ the minimum percent differences are 78% and 75% respectively. This confirms that the beam profiles' intensity mismatch cannot be affecting our observations.

Moreover, the beam profiles do not change with flipping the magnetic field, further confirming that the intensity profile mismatch cannot cause the sign of the PC difference to flip for the opposite magnetic field.

2.7.6 Reflection Imaging of the Sample

In the PC measurement setup depicted in Fig. 2.8, it is difficult to obtain a high enough quality image of the sample to properly align the beam to it. Another method is to use a Gaussian beam, the same used for PC measurements with $\ell = 0$, and move the sample using scanners (AN-Sxy100) while collecting the reflection with a photodiode. The measured voltage from the photodiode can be used to make an image of the sample; one of these images is shown in Fig. 2.11b. The gold contacts on the sample are highly reflective in comparison to the hBN/graphene/hBN heterostructure, therefore, in the image, the gold contacts will appear bright and areas of the sample without gold will appear dark (with the chosen color map). Since the outer boundary of the Corbino sample is covered with a gold contact and the center of the Corbino also has a gold contact, the image clearly defines the boundaries of the sample which can then be used for beam alignment.

2.8 Additional data

2.8.1 Additional Bias and Polarization data for $B = \pm 9$ T

Figures 2.2 and 2.3 show the bias voltage and polarization-dependent measurements for $\ell = \pm 2$. These measurements were also performed for $\ell = \pm 1$ as shown in Fig. 2.12. The results are consistent with what is seen in Fig. 2.2 and Fig. 2.3; the sign of the PC difference does not

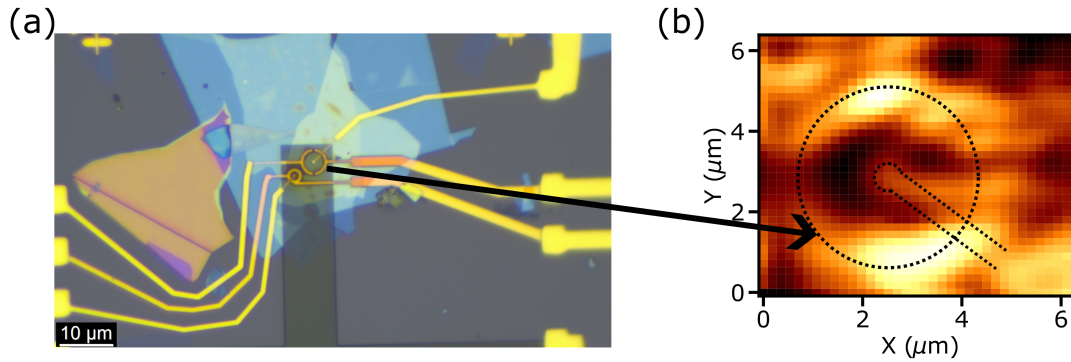


Figure 2.11: (a) Image of the sample taken with an optical microscope. (b) Image of the sample taken by measuring the optical reflection of the pump beam with a photodiode while moving the sample with scanners. The dashed curves show the outline of the Corbino sample.

change and the magnitude changes very little with bias voltage and polarization.

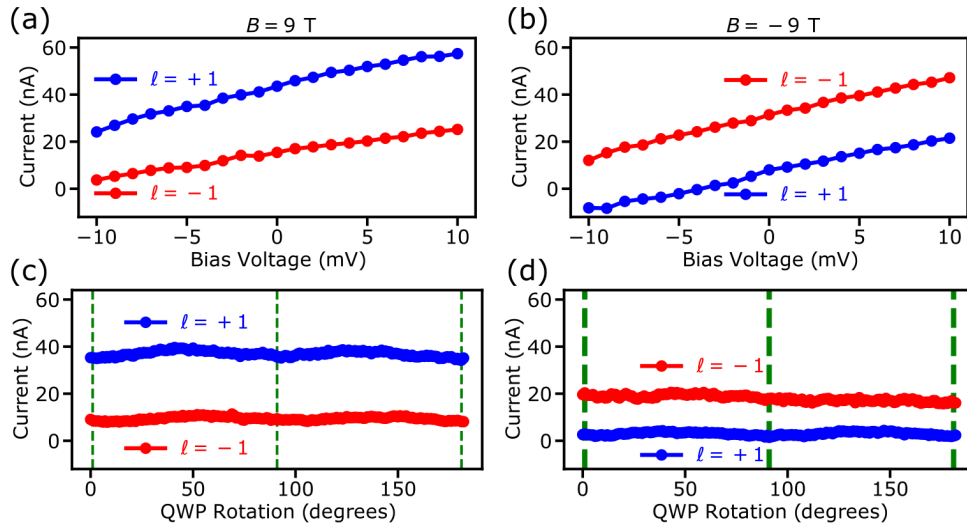


Figure 2.12: Measured PC as a function of bias voltage and polarization using light carrying $\ell = +1$ (blue) and $\ell = -1$ (red) at (a, c) $B = 9$ T and (b, d) $B = -9$ T.

2.8.2 Gate voltage sweep measurements for different bias

The gate voltage dependence was also measured for different bias voltages. This is shown in Fig. 2.13. It is observed that the gate voltage dependence changes very little with bias voltage and the flip with magnetic field stays the same.

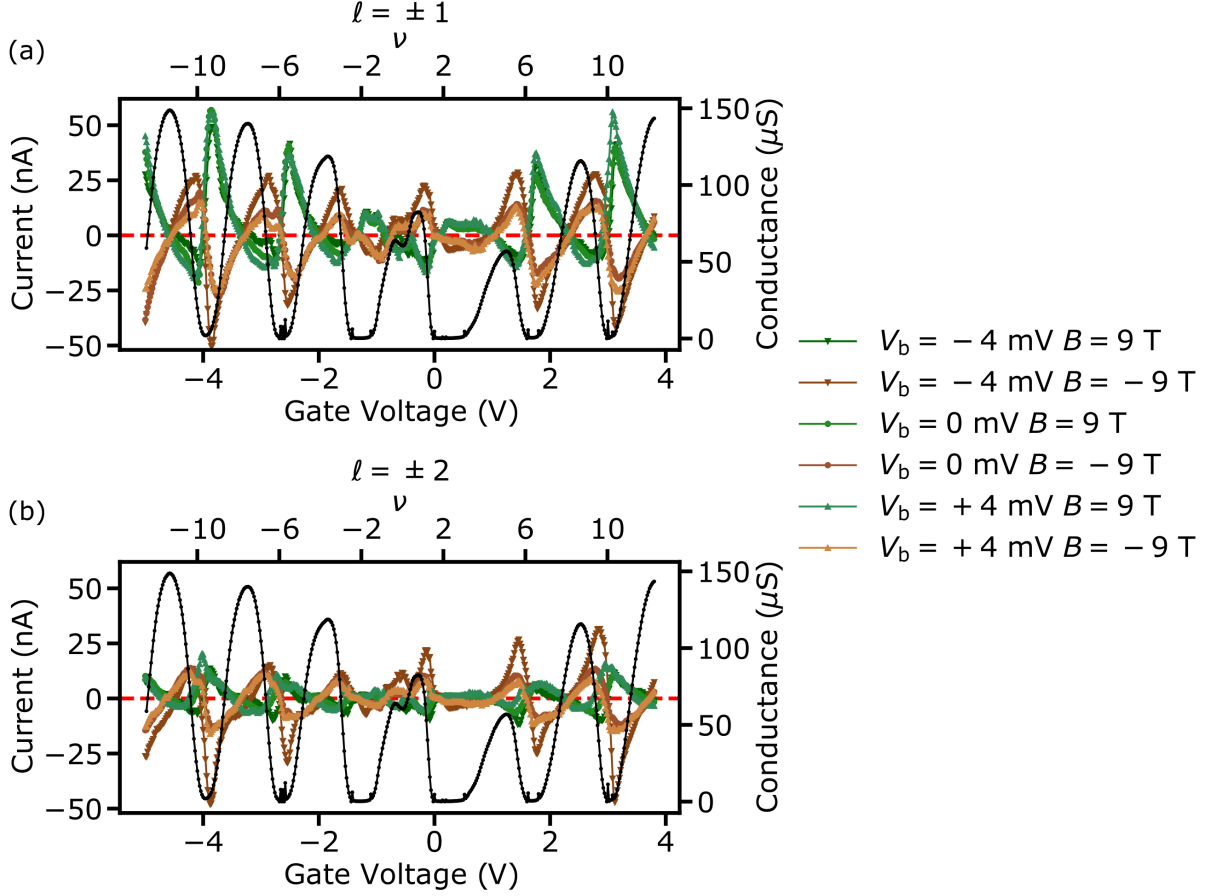


Figure 2.13: Measured PC difference as a function of gate voltage for different V_b at $B = 9$ T: (a) Subtraction of PC for $\ell = \pm 1$. (b) Subtraction of PC for $\ell = \pm 2$.

2.8.3 $B = \pm 4$ T Measurements

All previous measurements were taken at $B = \pm 9$ T. This is the maximum accessible magnetic field. To verify that these results are consistent with B , measurements were also taken at $B = \pm 4$ T. The results of the bias voltage-dependent measurements for $\ell = \pm 1$ and $\ell = \pm 2$ are shown in Fig. 2.14, the results for the polarization-dependent measurements for $\ell = \pm 2$ are shown in Fig. 2.15, and the gate voltage dependence of the PC difference ($\Delta_{PC}^{\ell=\pm 2}$ and $\Delta_{PC}^{\ell=\pm 1}$) are shown in Fig. 2.16. It is observed that these results are consistent with the $B = \pm 9$ T results. For

the $B = \pm 4$ T bias voltage and polarization measurements, the gate voltage was set such that the Fermi energy was close to $\nu = 6$ just like the $B = 9$ T data.

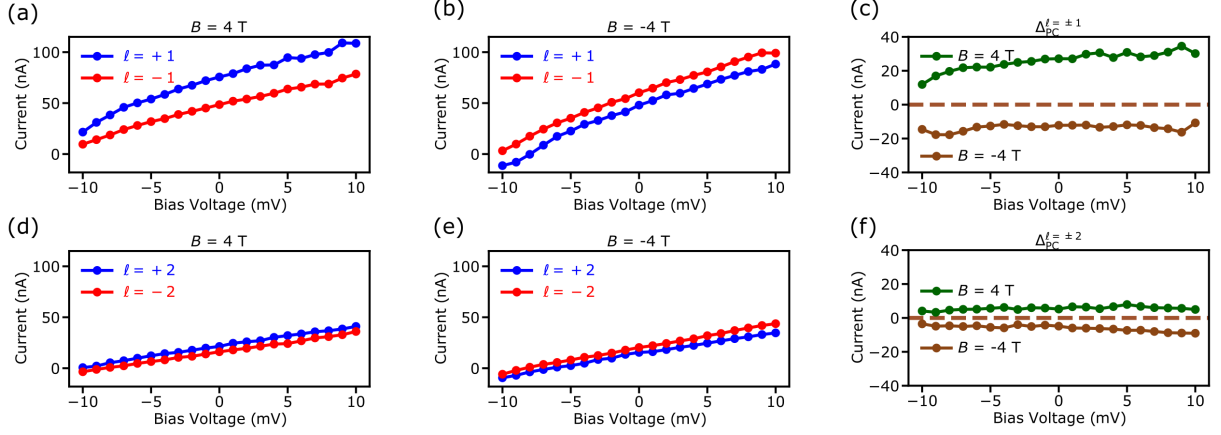


Figure 2.14: (a), (b) Measured PC as a function of bias voltage V_b generated using light carrying $\ell = +1$ (blue) and $\ell = -1$ (red) at (a) $B = 4$ T and (b) $B = -4$ T. (c) Shows $\Delta_{PC}^{\ell=\pm 1}$ for $B = 4$ T (green) and $B = -4$ T (brown), respectively. (d), (e) Measured PC as a function of bias voltage V_{rmb} generated using light carrying $\ell = +2$ (blue) and $\ell = -2$ (red) at (d) $B = 4$ T and (e) $B = -4$ T. (f) Shows $\Delta_{PC}^{\ell=\pm 2}$ for $B = 4$ T (green) and $B = -4$ T (brown), respectively. In all the panels, the gate voltage V_g and average pump power were 0.5 V and 10 μ W, respectively.

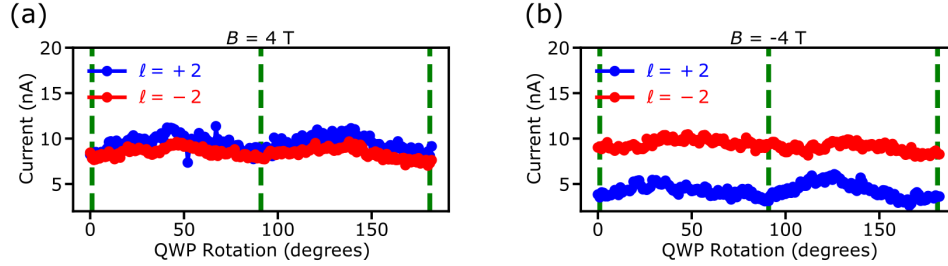


Figure 2.15: Polarization dependence of PC for $\ell = \pm 2$ at (a) $B = 4$ T (b) $B = -4$ T.

2.8.4 PC Laser Wavelength Dependence

To make sure the experimental results do not depend on the wavelength of the pump laser, the PC is measured as a function of wavelength. This measurement was performed using a Ti-sapphire laser (M2 SolsTiS) and placing a mirror in front of the SLM. The wavelength of the

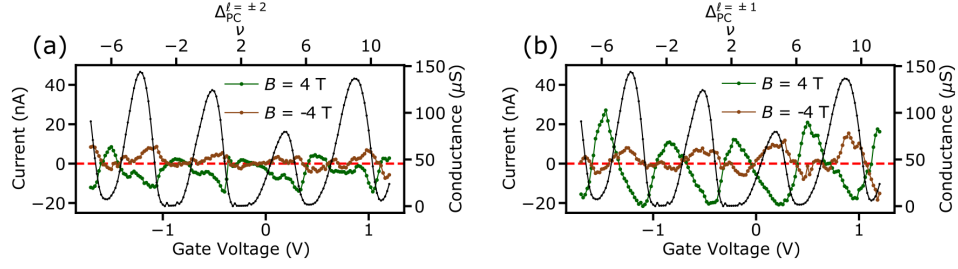


Figure 2.16: Gate voltage dependence of the PC difference for (a) $\ell = \pm 2$ and (b) $\ell = \pm 1$ for $B = \pm 4$ T.

laser was swept between 905 and 942 nm. The magnetic field was set to 9 T and the gate voltage was set either to filling factor $\nu = +2$ or to the zeroth LL. As seen in Fig. 2.17, the PC changes very little with the wavelength of the laser.

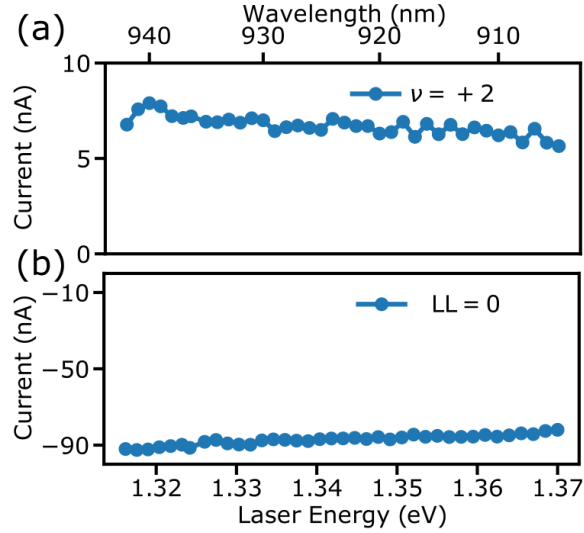


Figure 2.17: PC measured as a function of the wavelength of the pump laser. In (a) the gate voltage is set such that $\nu = +2$ and in (b) the gate voltage is set at charge neutrality (the zeroth LL).

2.8.5 Temperature Dependence of PC

To test the dependence of the vorticity-selective PC on the spatial coherence of electrons, the PC for $\ell = \pm 1$ at $B = 9$ T is measured as a function of temperature. As the temperature of

the system is increased, the spatial coherence of the electrons will diminish. Thus, it should be expected that the PC difference between $\ell = \pm 1$ should go to zero with increasing temperature. This is observed in Fig. 2.18, where the PC difference goes to zero, within error, by a temperature of 10 K.

This measurement was performed by finding the spatial point at which the PC difference flips with magnetic field, just as in previous measurements. The difference between the measured PC values in this data and other sets of data in the paper is due to the sample degrading with time and being removed from the cryostat in between measurements. The bias voltage was fixed at -4 mV and the gate voltage was swept between the filling factors $\nu = -2$ and $\nu = +10$. The average current shown in Fig. 2.18 is the average of the absolute value of the PC over the gate voltage range. The error bars were calculated by changing the beam back and forth between $\ell = \pm 1$ and calculating the standard deviation of the mean of the measured PC for each gate voltage in a smaller range. Error propagation was used to find the error for the average value, and this was assumed to be the same for each temperature.

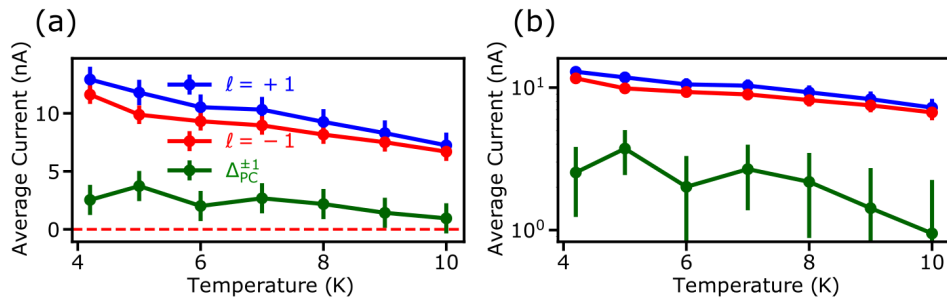


Figure 2.18: (a) Linear and (b) semi-logarithmic plot of the temperature dependence of the vorticity-selective PC. The PC measured for $\ell = +1$ ($\ell = -1$) is shown in blue (red) and the difference between PC measured with $\ell = \pm 1$ ($\Delta_{PC}^{\pm 1}$) is shown in green.

2.9 Theory

2.9.1 Selection Rules

We discuss the selection rules of exciting electrons with light from the lower to the upper band in graphene in the Quantum Hall regime, considering the possibility that the light might carry non-zero OAM. We consider the electromagnetic vector potential $\vec{A}$ of incident light coupled via Peierls substitution $\vec{k} \rightarrow \vec{k} - e\vec{A}$ and compute transition matrix elements as

$$M(n_f, m_f, n_i, m_i) = -ev_F \langle n_f, m_f | \vec{A}(\vec{r}) \cdot \vec{\sigma} | n_i, m_i \rangle \quad (2.4)$$

where $|n_x, m_x\rangle$ denotes initial ($x = i$) and final ($x = f$) states. n_x denotes the LL and m_x the quantum number related to angular momentum. The expected angular momentum in z -direction can be computed as $\langle L_z \rangle = \hbar(m - n)$. We compute the matrix elements in the spinor representation of the wave-function limiting to transitions from the lower band of graphene (negative n) to the upper band (positive n) and assuming $n \neq 0$. For σ^+ polarized light, $\vec{A}(\vec{r}) \cdot \vec{\sigma} = A(\vec{r})\sigma^+$, we get

$$\begin{aligned} M^+(n_f, m_f, n_i, m_i) &= -\frac{ev_F}{2} \begin{pmatrix} \langle n_f - 1, m_f | & \langle n_f, m_f | \end{pmatrix} A(\vec{r})\sigma^+ \begin{pmatrix} |n_i - 1, m_i\rangle \\ -|n_i, m_i\rangle \end{pmatrix} \\ &= \frac{ev_F}{2} \langle n_f - 1, m_f | A(\vec{r}) | n_i, m_i \rangle \end{aligned} \quad (2.5)$$

while for σ^- polarized light we get

$$M^-(n_f, m_f, n_i, m_i) = -\frac{ev_F}{2} \langle n_f, m_f | A(\vec{r}) | n_i - 1, m_i \rangle. \quad (2.6)$$

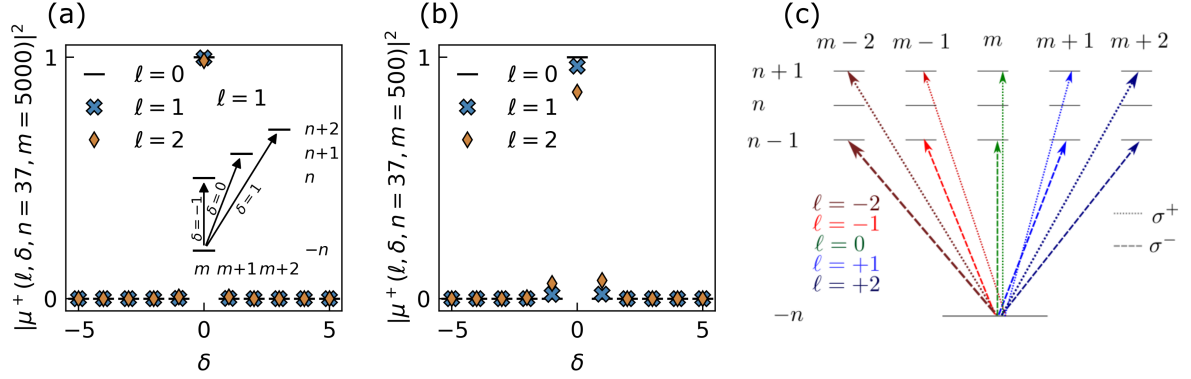


Figure 2.19: (a) Matrix element $\mu^+(\ell, \delta, n, m)$ as defined in Eq. (2.10) as a function of δ for $\ell = 0, 1, 2$ as in the experiment (limiting to positive OAM). $n = 37$ is fixed as in the experiment and $m = 5000$ which corresponds to a state with expected radial position $\langle r \rangle_{n=37, m=5000} \approx 1 \mu\text{m}$ which is somewhere in the middle of the sample, with radius $R = 2 \mu\text{m}$. The inset illustrates the effect of δ in the case of $\ell = 1$: $\delta = 0$ corresponds to the situation where the OAM of the light only changes the quantum-number m while $\delta \neq 0$ considers further transitions where n and m are simultaneously changed. Only the $\delta = 0$ transition has substantial weight. (b) Same as in (a.) just with $m = 500$ which corresponds to a radial position of the electron of $\langle r \rangle_{n=37, m=500} \approx 300 \text{ nm}$ hence close to the inner contact. The $\delta = 0$ transitions also dominate here but for smaller m , $\delta \neq 0$ transitions have an increased weight. (c.) Illustration of selection rules considering only $\delta = 0$ that have the dominant weight (see (a) and (b)).

For light carrying OAM, we write $A(\vec{r}) = Ae^{i\varphi\ell}$, $\ell = 0, \pm 1, \pm 2, \dots$ assuming no radial dependence of the light field. We compute the dimensionless and normalized matrix elements for σ^+ polarized light

$$\mu^+ = \langle n_f - 1, m_f | e^{i\varphi\ell} | n_i, m_i \rangle \quad (2.7)$$

and σ^- polarized light

$$\mu^- = \langle n_f, m_f | e^{i\varphi\ell} | n_i - 1, m_i \rangle \quad (2.8)$$

using the wave-functions

$$\langle r, \varphi | n, m \rangle = \frac{i^{(m-n)}}{\sqrt{2\pi\ell_B}} \sqrt{\frac{n!}{m!}} e^{-\frac{r^2}{4\ell_B^2}} e^{i\varphi(m-n)} r^{(m-n)} L_n^{(m-n)} \left(\frac{r^2}{2\ell_B^2} \right). \quad (2.9)$$

Here $L_a^b(x)$ are the associated Laguerre polynomials. We assume $n < m - 1$, since this is the relevant case for the experiment. In what follows, we will focus on σ^+ polarized light computing μ^+ . However, analogous steps can be performed to compute the matrix elements μ^- . Inserting the wave function [18, 41] Eq. (2.9) into Eq. (2.7) we get

$$\mu^+ = i^{(m_i - m_f - n_i + n_f - 1)} \sqrt{\frac{n_i!(n_f - 1)!}{m_i!m_f!}} \frac{1}{2\pi} \int_0^{2\pi} d\varphi e^{i\varphi(m_i - m_f - n_i + n_f - 1 + \ell)} \int_0^\infty dx 2x e^{-x^2} x^{m_i + m_f - n_i - n_f + 1} L_{n_i}^{m_i - n_i}(x^2) L_{n_f - 1}^{m_f - n_f + 1}(x^2). \quad (2.10)$$

The angular integral enforces angular momentum conservation. We introduce $n := n_i$ and $m := m_i$ and define

$$n_f =: n + \delta + 1 \quad (2.11)$$

$$m_f =: m + \ell + \delta$$

such that angular momentum conservation is fulfilled for any choice of n , m , ℓ , and δ . Note that Refs. [26, 31] only considered $\delta = 0$ case. With this

$$\mu^+(\ell, \delta, n, m) = i^\ell \sqrt{\frac{n!(n + \delta)!}{m!(m + \delta + \ell)!}} \int_0^\infty dx 2x e^{-x^2} x^{2(m-n)+\ell} L_n^{m-n}(x^2) L_{n+\delta}^{m-n+\ell}(x^2). \quad (2.12)$$

We plot μ^+ as a function of δ for $\ell = 0, +1, +2$ in Fig. 2.19 for $n = 37$ and $m = 5000$ (a) and $m = 500$ (b). For $m = 5000$ the expected radial position of the electron is $\langle r \rangle_{n=37, m=5000} \approx 1 \mu\text{m}$ hence somewhat in the radial middle of the sample that has an overall radius of $R \approx 2 \mu\text{m}$. For $m = 500$ we have $\langle r \rangle_{n=37, m=500} \approx 300 \text{ nm}$, which is close to the radius of the inner contact. For $\ell = 0$ the orthogonality of the Laguerre polynomials enforces $\delta = 0$ such that we get the well

known selection rules [41]

$$\mu^+(\ell = 0, \delta, n, m) = \delta_{\delta,0} \Rightarrow \mu^+ = \delta_{n_f-1, n_i} \delta_{m_f, m_i} \quad (2.13)$$

and for μ^-

$$\mu^-(\ell = 0, \delta, n, m) = \delta_{\delta,0} \Rightarrow \mu^- = \delta_{n_f, n_i-1} \delta_{m_f, m_i}. \quad (2.14)$$

For $\ell \neq 0$ we don't get strict Kronecker-Delta-like selection rules but instead, multiple transitions are in principle allowed. However, as can be seen in Fig. 2.19, transitions with $\delta \neq 0$ are suppressed compared to the $\delta = 0$ transitions. We illustrate the selection rules, only considering $\delta = 0$, in Fig. 2.19 (c).

In the main experimental data, as a control experiment, we have inverted the B -field $\vec{B} \rightarrow -\vec{B}$. It is therefore interesting to consider how the selection rules change in this case. For deriving the selection rules as above, we have used the B -field to define the z -axis of the system. Therefore the sign of the OAM of light as well as the handedness of the polarization is determined by whether they are aligned or anti-aligned with the magnetic field. Inverting the B -field is therefore equivalent to inverting the OAM and the polarization. Hence we can relate the transition matrix elements at magnetic field $\vec{B}$ and $-\vec{B}$ as

$$\mu^+(\ell, \delta, n, m) \Big|_{\vec{B}} = \mu^-(-\ell, \delta, n, m) \Big|_{-\vec{B}}. \quad (2.15)$$

Based on this observation, when measuring the subtracted photo-current at magnetic field $\vec{B}$

$$\Delta I \Big|_{\vec{B}} = I^{+\ell} \Big|_{\vec{B}} - I^{-\ell} \Big|_{\vec{B}} \quad (2.16)$$

one expects this to change sign when inverting the magnetic field hence

$$\Delta I \Big|_{\vec{B}} = -\Delta I \Big|_{-\vec{B}}. \quad (2.17)$$

This is what we have used as a control experiment and indeed we observe the same sign reversal in the experiment.

2.9.2 Relation between expected radius and quantum numbers n and m

In this part, we compute the expected radius of the electronic wavefunctions in graphene in the quantum Hall regime. In spinor representation, they can be computed as

$$\langle r \rangle = \begin{cases} \langle n = 0, m | r | n = 0, m \rangle ; & n = 0 \\ \frac{1}{2} (\langle n, m | r | n, m \rangle + \langle n - 1, m | r | n - 1, m \rangle) ; & n \neq 0 \end{cases}. \quad (2.18)$$

The wave functions $\phi_{n,m}(r) = \langle r, \phi | n, m \rangle$ read [18, 41]

$$\phi_{n,m}(r, \varphi) = \frac{i^{|n-m|}}{\sqrt{2\pi\ell_B}} \sqrt{\frac{\min(n, m)!}{\max(n, m)!}} e^{-\frac{r^2}{4\ell_B^2}} e^{i\varphi|n-m|} r^{|n-m|} L_{\min(n, m)}^{|n-m|} \left(\frac{r^2}{2\ell_B} \right) \quad (2.19)$$

and are normalized according to

$$\int_{\mathbb{R}^2} d\vec{r} \phi_{n,m}(\vec{r}) \phi_{n',m'}^*(\vec{r}) = \delta_{n,n'} \delta_{m,m'}. \quad (2.20)$$

We show the expected radii as a function of the quantum number m for different values of n in Fig. 2.20. For $n = 0$ the expected position approximately coincides with the position of the

guiding center $r_{\text{Guide}} = \ell_B \sqrt{2m + 1}$ while for larger n one gets larger radii as expected.

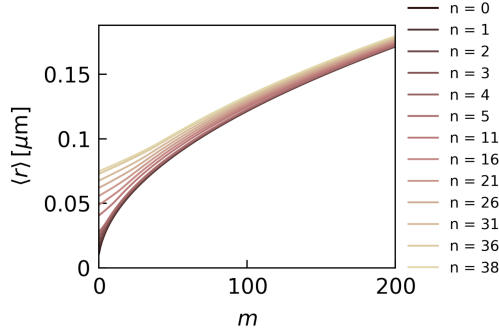


Figure 2.20: Expected radial position $\langle r \rangle$ according to Eq. 2.18 as a function of quantum number m in different LLs n .

2.9.3 Ideal interacting quantum Hall OAM response

The above argument can be generalized to understand the OAM PC in the presence of Coulomb interactions. Let us consider photons incident on disorder-free Graphene in a perfect Corbino geometry with a magnetic field and Coulomb interactions. The Coulomb interaction is key to helping the carriers to thermalize and our discussion includes relaxation processes such as Auger. Let us further assume that the bulk is gapped and there are two edges, i.e., an inner and outer edge, which for now we will assume to be isolated from contacts.

Given the circular symmetry, the angular momentum is the key quantum number to think about. In fact, in this ideal situation, energy E and angular momentum L are the only two conserved quantities. For a gapped bulk, the system thermalizes to a state with a low density of bulk quasiparticles. This means that the edge charge Q is also a conserved quantity. As a consequence, the final state (after the absorption of some number of photons) is characterized by edges with quantum numbers L , E , and Q .

The edge theory is more conventionally described in terms of momentum k rather than

angular momentum. Note that in a magnetic field, momentum is shifted by the vector potential at radius R of the edge is $k_F = A = BR/2$. Transferring charge Q to the edge changes momentum by Qk_F . Adding energy through particle-hole excitations changes momentum by E/v where v is the edge velocity. The change in angular momentum is $L = R(Qk_F + E/v) = R(QBR/2 + E/v)$. This is consistent with the wave-functions of LLs at angular momentum m seen in the last section being at a radius $\sim \sqrt{2m/B}$.

Photons supply both OAM angular momentum l and energy E_{ph} . Considering angular momentum and ignoring the contribution of the photon energy, the total charge transferred is $\delta Q_L = 2N_{\text{ph}}l/BR^2 = 2N_{\text{ph}}l(l_B/R)^2$. The photon energy contribution turns out to be independent of OAM and therefore even in magnetic field. The charge transfer expression matches that from the LL wave-function picture in the previous section and shows that interactions (at least in the absence of phonons and disorder) do not affect the OAM PC, which is a result of angular momentum pumping.

2.9.4 PC sign oscillation: LLs and injection current picture

LLs picture: As described in a simplified picture earlier (Fig. 2.1), in the quantum Hall regime, light with non-zero OAM can induce optical transitions between the LLs such that the spatial extent of these states depends on the associated angular quantum number m , where the radius of LLs increases with m . In our Corbino device, depending on the helicity of light, this spatial expansion (shrinking) of the carriers' wavefunction manifests itself as an outward or inward radial current.

This intuitive picture for the PC generation due to the radial modulation of the electron

wavefunction caused by the optical vortex does not immediately suggest PC oscillations as a function of gate voltage (V_g), which are seen in Fig. 2.4. To understand this effect, one must note that the PC is actually the result of optical excitation and subsequent carrier relaxation. As known from earlier studies [31,42] the position of E_F within a LL, controlled by V_g , gives rise to a relaxation bottleneck for either electrons or holes. Due to the opposite charges of the different carriers (electrons and holes), this leads to a change of PC polarity. That is, PC oscillations upon sweeping through a LL. While in the rectangular geometry experiment of Ref. [31] this bottleneck argument relied on equal chirality of electrons and holes at the edge of the sample, in the present scenario, the direction of transport is determined by the relative helicity of twisted light and magnetic field. Naively, one might expect that the OAM would lead to a relative shift of electron and hole position, i.e., to opposite shifts for the two types of carriers. However, Coulomb attraction ensures that the shift is experienced by the center of mass of the electron-hole-pair (see also Ref. [43] for the case of excitons). Hence, electrons and holes are moved in the same direction which explains the observed current oscillations.

Unfortunately, formalizing this picture, i.e., via optical Bloch equations as has been done in Ref. [31], is extremely complicated. Not only would it be necessary to explicitly account for LL and orbital degrees of freedom, but the picture also suggests that properly accounting for the Coulomb attraction between electrons and holes will be crucial to capture the oscillations. However, an alternative description is able to provide a formalism which is suited to explain the observed behavior.

Injection current picture: In our system, since the photon frequency is in the optical regime (pump wavelength 940 nm = 1.32 eV and $E_{n=37} - E_{n=-36} = 1.315$ eV), one expects the LL spacing ($E_{n=37} - E_{n=36} = 9$ meV) to be much smaller than the lifetime broadening of

excited electrons and holes. Therefore, we can consider optically excited electrons and holes as approximately free Dirac particles. Beyond the dipole approximation, the OAM of the pump photons locally imparts momentum to the electrons and holes in the tangential direction (in the polar coordinates of the Corbino disk). Moreover, the Lorentz force from the external out-of-plane magnetic field imparts radial components to the velocity of the electron and holes. As detailed in the following section, the radial current of both the electrons and holes in this scenario is therefore given by the following

$$I_{\text{radial}} = -\frac{Bq}{k^4} \cos(2\theta)(\tau_e + \tau_h) \quad (2.21)$$

where B is the external out-of-plane magnetic field, q is the local OAM-induced momentum, θ is the angle between the electron and hole's momentum k , and τ_e and τ_h are the scattering times of the electrons and holes, respectively. From the Dirac equation $k = \omega/(2v_D)$. The total induced current is

$$I_{\text{meas}} = \int d\theta W(\theta) I_{\text{radial}}(\theta) \quad (2.22)$$

where $W(\theta)$ is the weight of the current contribution from the electrons and holes moving at the angle θ . In an ideal case, W should be θ -independent, leading to a vanishing of the measured current in our system. However, as explained later in the following section, W can be a function of both θ and the longitudinal conductivity, which is a function of the carrier density (and therefore gate voltage V_g) and B . This can potentially explain the observed V_g -dependent polarity change of the measured PC.

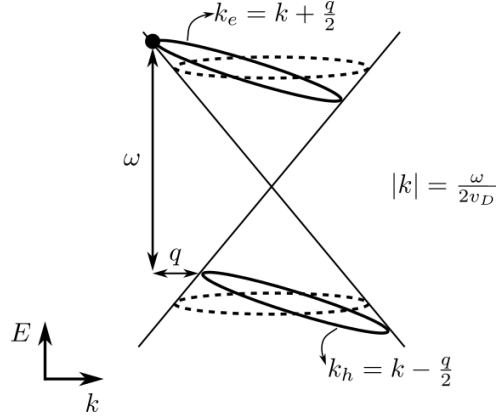


Figure 2.21: Optical excitation by finite wave-vector light in a band structure of Dirac electrons in graphene.

2.9.5 Theory of OAM photoresponse in the strong scattering regime

The OAM field from light with frequency ω can be written as $A(\vec{r}) \propto e^{il\theta}$, where (r, θ) are the radial and angular coordinates. This can then be expanded in terms of a tangential coordinate $x_t = r\theta$ as $A(\vec{r}) \propto e^{ilx_t/r} = e^{iqx_t}$ where $q = l/r$. Locally, in the Corbino geometry, we can approximate the OAM as a plane wave with wave-vector q .

Considering the absorption of the photon by Dirac electrons, at rate R per unit area, shown in Fig. 2.21, leads to an electron-hole pair with momenta $k_e = k + q/2$ and $-k_h = -(k - q/2)$, respectively, where $|k| = \omega/2v_D$ is independent of q . To check that this is correct note that the energy of such a pair is $|k + q/2| + |k - q/2| = 2|k| + o(q^2)$.

Next, we consider (seen in Fig. 2.22) the generated electron-hole pair in momentum space along with the velocity of each particle as well as the Lorentz-force induced momentum change. For a quasiparticle at momentum k (note that we designate holes to be at the momentum of the missing electron), the velocity is $u_{e,h}(k) = \pm v_D \vec{k}/k$. The Lorentz-force induced momentum change is $B \times u_{e,h}(k) \delta\tau_{e,h}$ where τ_e (τ_h) is the scattering time of electrons (holes). The average

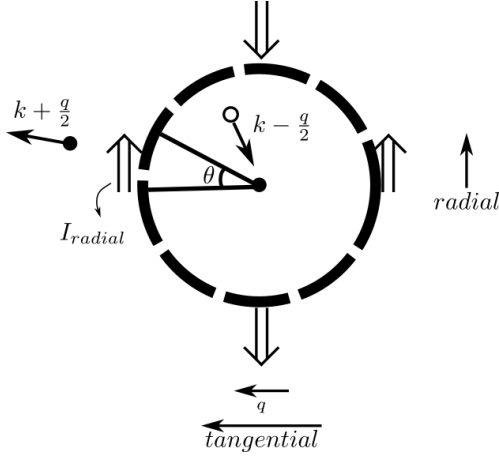


Figure 2.22: Wave-vector and velocity structure of electron-hole pairs in a magnetic field excited by light at wave-vector q , which is tangential in direction relative to the Corbino geometry. The dotted line shows the wave-vector $|k| = \omega/2v_D$ where the optical excitations originally occur.

(over a scattering time) change in velocity induced by the Lorentz force turns out to be $\delta u_{e,h}(k) = v_D^2 \tau_{e,h} \frac{B \times \vec{k}}{2k^2}$, which is the same for electrons and holes. The sign is independent of electrons or holes because both the velocity as well as the momentum change changes sign for both electrons or holes.

The Lorentz-force-induced current for electrons and holes at wave-vector k would cancel (since the velocities are the same) except for the relative wave-vector q induced by the OAM. This leads to an OAM-B-field induced velocity $\delta_2 u(k) = (q \cdot \nabla_k) \delta u(k) = \frac{qv_D^2 \tau B}{k^2} (\sin 2\theta, -\cos 2\theta)$ where θ is the angle of $\vec{k}$ relative to the tangential direction $\vec{q}$. This velocity is opposite for electrons and holes so that the electron and hole components of the current now add up. The relevant current in the Corbino geometry is the radial current $I_{\text{rad}} = \frac{q_e^2 R v_D^2 (\tau_e^2 + \tau_h^2) q B}{k^2} \cos 2\theta = q_e (R l_B^2) (2\pi l) \{(\tau_e^2 + \tau_h^2) \omega_c^2\} \cos 2\theta$. The prefactor $2q_e \pi l (R l_B^2)$ is the ideal expectation based on angular momentum.

As shown in the figure, the radial component of the current flips sign with θ , i.e., $I_{\text{rad}}(\theta \sim 0, \pi) > 0$ while $I_{\text{rad}}(\theta \sim \pm\pi/2) < 0$. In fact, the straightforward average over $\cos 2\theta$ would

vanish. However, for reasons we describe below, the total measured radial current $I_{\text{meas}} = \int d\theta W(\theta) I_{\text{rad}}(\theta)$ is a weighted average of the radial current where the presence of a $\cos 2\theta$ Fourier component of $W(\theta)$ would lead to a non-vanishing measured current.

To understand the origin of a non-trivial weight function $W(\theta)$, we note that θ represents the angle of the wave-vector k (and hence velocity) relative to the tangential direction in the Corbino geometry at which the electron-hole pair is excited. Electron-hole pairs at angles $\theta \sim \pm\pi/2$ are dominantly moving in the radial direction either towards or away from the Corbino edge. In contrast, pairs at $\theta \sim 0, \pi$ are moving tangentially. As clear from Fig. 2.23, the formal set of electron/hole pairs and the associated current can hit the edge on a lower time scale. This reduces the scattering time $\tau_{e,h}$ for such quasiparticles. Thus, the weight function in the radial direction $W(\theta \sim \pm\pi/2) \sim (\tau_e^{(r)} + \tau_h^{(r)})/(\tau_e + \tau_h)$, where $\tau_{e,h}^{(r)}$ is the quasiparticle scattering time for particles moving in the radial direction and $\tau_{e,h}$ is the scattering time of the quasi-particles averaged over the Fermi surface. In contrast, the tangentially moving quasiparticles have an averaged scattering time, but have a contribution to the edge conductance that is suppressed by the longitudinal conductivity $\sigma_{xx,W}$ of warm carriers. Note that the relevant conductivity is that of electron-hole pairs that are relaxing from the high optical energy scale to the ground state. The relevant weight function in the tangential direction, which arises from a current divider effect between the bulk and contact resistance, is $W(\theta \sim 0, \pi) \propto R_{\text{contact}} \sigma_{xx,W}$, where R_{contact} is the contact resistance and $\sigma_{xx,W}$ is the two-dimensional conductivity (which has the same units as R_{contact}^{-1}). This is a rough qualitative estimate that may be refined by more detailed modeling of the conductivity. However, we expect the key feature of dependence on bulk conductivity to be robust.

The form of the weight function discussed above can predict a switching of sign with

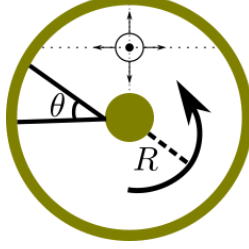


Figure 2.23: Scattering in Corbino geometry. Quasiparticles moving radially towards the edge, i.e., $\theta \sim \pm\pi/2$, have a shorter scattering time resulting in smaller B-field current though can more efficiently reach the edge because of ballistic transport relative to those moving tangentially, i.e., $\theta \sim 0, \pi$.

changing gate voltage of the measured B-dependent part of the OAM PC. As the gate voltage changes, the longitudinal conductance $\sigma_{xx,W}$ is expected to change as one passes through LLs. This leads to modulation of $W(\theta \sim \pm\pi/2)$, while one can expect $W(\theta \sim 0) \lesssim 1$ to have minor variations. At the same time $I_{\text{rad}}(\theta \sim 0, \pi) \sim -I_{\text{rad}}(\theta \sim \pi/2)$. Therefore, $I_{\text{meas}} \sim I_{\text{rad}}(\theta \sim 0, \pi)[W(\theta \sim 0, \pi) - W(\theta \sim \pm\pi/2)]$ changes sign when the bulk conductivity changes from low to high or vice-versa as the gate voltage sweeps through LLs.

2.10 Summary

In summary, we demonstrated a novel mechanism for transferring OAM from photons to electrons. Our OAM optical pumping scheme is analogous to cold-atom and ion optical pumping, and can be used to manipulate itinerant electrons. Additionally, our results suggest that the control of spatial degrees of freedom in light-matter interactions can become a new and versatile toolbox in solid-state systems. This approach heralds a new ability to image the spatial coherence of electrons, a fundamentally new probe of quantum materials, inaccessible through existing measurements such as multi-port transport and scanning tunneling microscopy [44]. For example, our work highlights the potential of using the OAM degree of freedom as a powerful tool to

further the field of quantum Hall physics. One immediate direction is to employ THz fields, as opposed to the optical fields used in our case, to excite the two nearest LLs [45]. The advantage of this approach is the absence of cascade relaxation. Also, the influence of gradient fields on the quantum Hall system has been recently observed in the THz domain, suggesting that this platform is a promising candidate [46]. A more ambitious direction involves the strongly interacting limit, where one could exploit the transfer of OAM to probe fractional quantum Hall states and excite and manipulate anyons [47–51].

Moreover, our experiment provides a unique testbed for investigating the interplay between topology and chirality in the interactions between electrons and photons. While our experiment was performed in the low excitation limit, there are several intriguing proposals to use a strong drive field and exploit the spatially coherent light-matter interaction to induce a wider class of topological insulators in electronic systems by using various structured light [6, 52–55]. Furthermore, while our graphene system lacks a photoluminescence response, our scheme can be applied to materials where emission from electronic LLs is possible [56], potentially enabling the observation of chiral photon emission. Another promising avenue is the prospect of coherent wavefunction spectroscopy, where interferometric techniques can be integrated into our experimental scheme to measure and modulate the spatial distribution of wavefunction amplitudes and phases [57].

Chapter 3: Spin-selective strong light-matter coupling in a 2D hole gas-microcavity system

3.1 Introduction

A gas of charged particles confined in two dimensions has been a cornerstone system to investigate numerous single-particle and many-body quantum mechanical effects. In these systems, the reduction in the dimensionality has enabled the demonstration of integer [12, 58] and fractional [59] quantum Hall effects, Mott-insulating phases [60, 61], superconductivity [62], formation of skyrmions [63, 64] and spin-ordered states [65] among other effects [55]. Recently, the possibility of coupling such 2D gases to an optical mode, forming cavity exciton-polaritons, has opened up exciting new directions. For example, this capability has been exploited to optically study integer and fractional quantum Hall states [66, 67], formation of skyrmions and manipulation of the magnetic Landé g factor [68], and the enhancement of optical non-linearities, when an electron gas, in the fractional quantum Hall regime, is dressed with the cavity mode [48]. Recent experiments have also demonstrated giant Zeeman splitting and valley-selective light-matter coupling in a monolayer TMD [69]. The demonstration of the mentioned effects requires that the light-matter interaction strength in the microcavity exciton-polaritons exceeds cavity and matter dissipation — a regime known as strong coupling (SC) [70].

Combining SC with broken time-reversal symmetry in chiral systems is expected to unlock new perspectives for the optical manipulation of correlated states of matter [8, 10, 11]. The chirality can arise from either electronic or photonic degrees of freedom, and in exciton-polariton systems these two are intertwined. This work illustrates a method to induce strong spin selectivity in the light-matter coupling between an electronic transition and a cavity mode. To achieve this, we used a hole (h^+) gas in a semiconductor Quantum Well (QW), embedded in a microcavity. The system was subjected to an external magnetic field (B) to be in the quantum Hall regime. The modification of the density of states induced by B resulted in the selectivity of the light-matter coupling for each circularly polarized state of light. We explain this selectivity, in a single-particle picture, by combining an exciton-polariton model and the LL filling picture. We note that the robustness of this mechanism is demonstrated at a temperature of 3.5 K, where the Coulomb-induced matter correlations are expected to be irrelevant.

3.2 Results

3.2.0.1 System description

Our physical system consists of a doped GaAs QW whose electronic states can be manipulated either through an applied gate voltage or a magnetic field. The sample is a p-doped grown QW, which constitutes a 2-dimensional h^+ gas (2DHG). To strongly enhance the interaction with an electromagnetic mode, the QW is embedded in a Distributed Bragg Reflector (DBR) semiconductor microcavity. Such a hybrid system provides a powerful tool to analyze the electron spin properties through optical experiments by exploiting the selection rules, which determine a spin selective transition depending on the circular polarization of light. The origin of this selectivity is

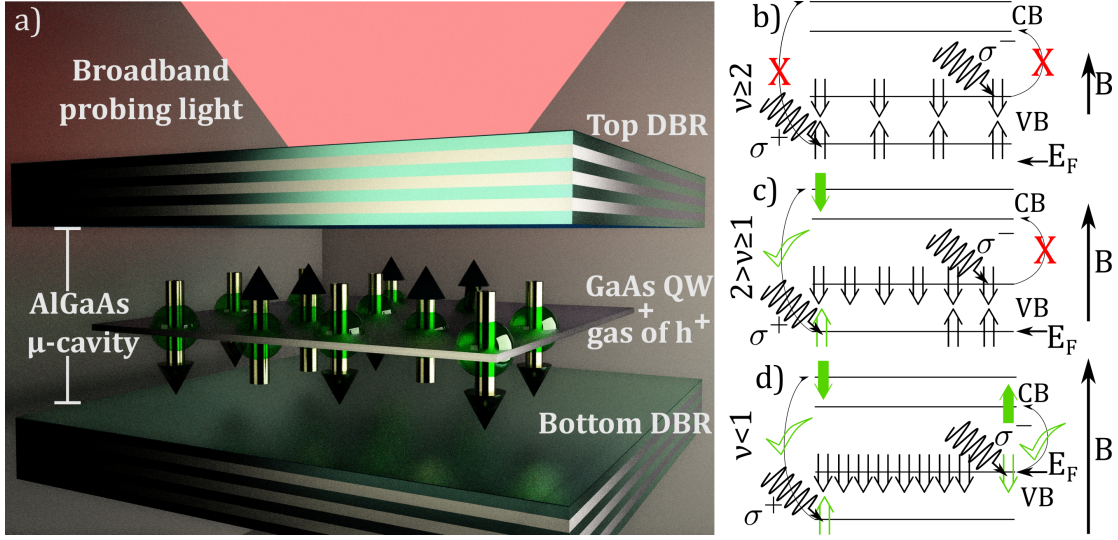


Figure 3.1: a) Diagram of the sample, including an illustration of the 2DHG embedded in an optical microcavity. b-d) mechanism to induce spin-selectivity for light-matter coupling benchmarked by three ranges of the filling factor ν . VB and CB stand for Valence Band and Conduction Band, respectively. b) For $\nu \geq 2$, none of the spin VBs have available states and therefore no electronic transition can take place. As a consequence, the SC regime will not be reached. c) As the magnetic field increases and enters the $1 \leq \nu < 2$ regime, the density of states is tuned such that the σ^+ sub-band becomes available. Unlike the former case, the electronic transition is not blocked anymore and this spin population reaches the SC regime. The σ^- states are still Pauli-blocked, so no electronic transition is allowed for them. The result is the spin-selective light-matter coupling. d) As B is further increased such that the system is tuned into the $\nu < 1$ regime, the σ^- band starts to have available states and can reach SC. The asymmetry in the Rabi frequency persists because the σ^- band is still populated, in contrast to the σ^+ band that is fully available. The Fermi energy (E_F) is indicated for each regime in panels b to d.

in the crystalline structure and spin-orbit coupling properties of GaAs QWs, whose upper valence band is a heavy hole-band (with total angular momentum $J_z^h = \pm 3/2$). A schematic representation of the 2D gas of semi-integer spin-charged particles confined to the QW and interacting with the electromagnetic field is shown in Fig. 3.1a. All data in this chapter, unless otherwise labeled, were obtained at 3.5 K, where we do not expect any correlated electron physics to be present. A more detailed discussion of the selection rules and comparison with data taken at 40 mK can be found in Sections 3.5 and 3.7, respectively. The details of the sample composition and fabrication can be found in Section 3.4.

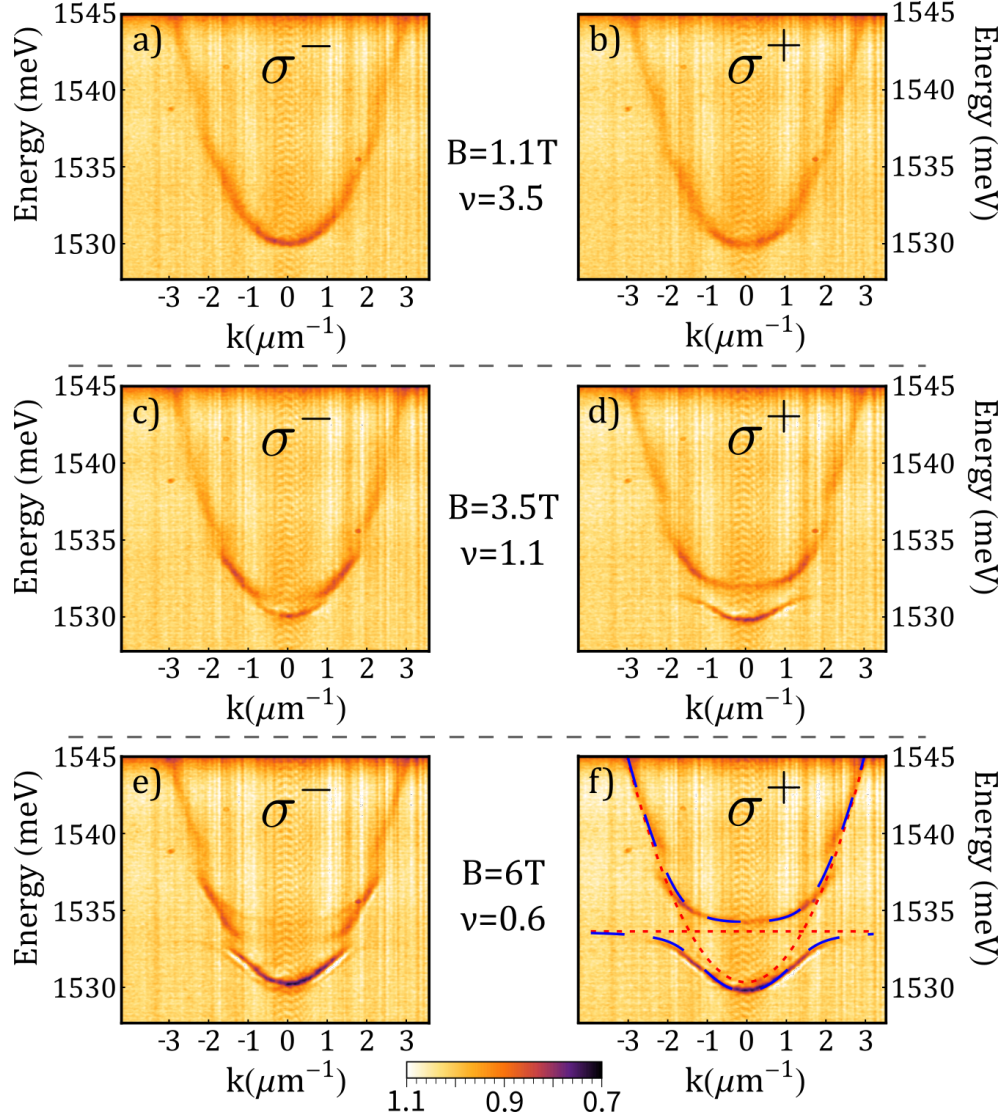


Figure 3.2: Energy dispersion of the system probed at values of B corresponding to the three ranges of ν shown in Fig. 3.1b-d. Panels a, c, and e correspond to σ^- polarization, and panels b, d, and f to σ^+ . a) and b): when $\nu \geq 2$ both spin sub-bands are occupied and no electronic transition can take place in the system. Only the bare cavity mode is visible. c) and d): in the regime $2 > \nu \geq 1$ only the σ^+ spin sub-band has available states, and hence the spin selectivity manifests. Only the dispersion probed with σ^+ polarization shows polaritonic bands, while σ^- remains decoupled. e) and f): upon further increasing B , also the σ^- population can reach the SC, but the remaining h^+ population in this sub-band makes the light-matter coupling asymmetric, making the Rabi splitting larger for σ^+ . Panel f includes the fitting obtained from a theoretical model (Eq. 3.1). Cavity and exciton modes are depicted in red dotted lines, while upper and lower polariton branches are depicted in dashed blue lines.

3.2.0.2 Mechanism for spin-selective SC

An external magnetic field acting on the 2D gas induces LLs that are non-degenerate for the spin eigenstates. This spin energy splitting depends on the magnitude of B , and in particular, for a h^+ doped system, it has a strong dependence on the particle density due to the spin-orbit interaction [71, 72]. At the same time, the h^+ density and the magnetic field determine the Fermi energy and hence the existence of an optically active transition of the electrons in the valence band [73, 74]. We exploit this particular interplay to obtain a spin-selective light-matter interaction. Specifically, for a fixed density of particles, we tune B to consider three relevant situations with fundamentally different light-matter coupling properties. The quantity that determines the three cases is the quantum Hall filling factor $\nu = \rho/n_B$, defined as the ratio between the h^+ density ρ and the density of magnetic states $n_B = eB/h$. The three cases are depicted in Fig. 3.1(b-d). The first scenario (b) takes place when $\nu > 2$. In this condition, there are no available states in any of the two spin sub-bands, inhibiting the possibility of an electronic transition from the lowest LL and therefore keeping the system in the so-called weak coupling regime. The next scenario is where $1 \leq \nu < 2$ (c). In this case, one of the sub-bands (σ^-) is still full, but the opposite spin state (σ^+) can instead host optically generated electron-hole pairs, leading to a polarization-selective SC. Finally, when $\nu < 1$, as shown in Fig. 3.1(d), both spin transitions are open, but due to the imbalance in the h^+ population, the system still has a strong asymmetry in the light-matter coupling.

3.2.0.3 Optical measurements

To investigate the proposed mechanism, we performed angular and spectrally-resolved reflectivity measurements to reconstruct the energy dispersion in a confocal setup. Such a setup is capable of mapping the far-field for any polarization state, using a broadband light source for excitation. A complete description of the optical setup can be found in Section 3.4. ρ is manipulated through a gate voltage and it is set to $\sim 9.4 \cdot 10^{10} \text{ cm}^{-2}$; a density high enough to guarantee that at low B , both spin valence sub-bands (σ^+ and σ^-) are occupied, but low enough to allow the desired effect to take place at a few Teslas, i.e., the higher the density, the higher the magnetic field at which the conditions $\nu = 2$ and $\nu = 1$ take place. We sweep B to tune the system over a range of filling factors that covers the three regimes described in Fig. 3.1(b-d). Figure 3.2 shows the measured dispersion of the exciton-polariton system for both circular polarizations and values of B in the three ranges of ν . For $\nu \geq 2$, as shown in panels a and b, and in agreement with the above description, only the parabolic dispersion from a bare cavity mode can be observed. This indicates that none of the spin sub-bands have available states, and hence no optical electronic transition can take place. Next, by increasing B , we reach the $2 > \nu \geq 1$ regime, and then the asymmetry in SC is manifested. Although the dispersion is still parabolic for σ^- (panel c), the σ^+ channel has clearly reached the SC regime, as observed from the formation of lower and upper polariton branches in the energy dispersion (panel d). By further increasing B , we reach the regime $\nu < 1$ (panels e and f), where both spin sub-bands can now reach SC, but the asymmetry in the h^+ distribution causes the population of σ^- to couple less efficiently to the cavity mode than the population in the σ^+ sub-band. As a consequence, the vacuum Rabi splitting is different for each state of circular polarization.

A coupled oscillator model is employed for the quantitative analysis of our observation, characterized by the polaritonic Hamiltonian:

$$\hat{H} = \sum_k \left(\omega_c + \frac{k^2}{2m} \right) \hat{a}_k^\dagger \hat{a}_k + \omega_X \sum_k \hat{b}_k^\dagger \hat{b}_k + \Omega \sum_k \hat{b}_k^\dagger \hat{a}_k + \hat{b}_k \hat{a}_k^\dagger, \quad (3.1)$$

for each polarization, where ω_c is the cavity mode's energy, k its momentum, m its effective mass, and $\omega_X(B)$ and $\Omega(B)$ are the energy of the electronic transition and the Rabi splitting, respectively (the latter two quantities depend on B). The operators $\hat{a}_k$ correspond to the annihilation operators of the cavity mode k , and $\hat{b}_k$ to the electronic transition with corresponding momentum k [75]. An example of the obtained polaritonic eigenfunction after fitting the data is included in Fig. 3.2f. The cavity and exciton modes are depicted (dotted red lines) together with the upper and lower polariton branches (dashed blue lines) that match the experimental dispersion.

The trend of spin-selective light-matter coupling as a function of the filling factor is illustrated in Fig. 3.3. To obtain it, we extract a vertical section of the energy dispersion corresponding to the momentum at which the bare cavity and the electronic transition cross (deduced from the theoretical model). From Fig. 3.3 one can observe how the filling factor $\nu = 2$ benchmarks the establishment of SC for σ^+ , which is observed as the emergence of two resonances instead of one, while σ^- remains decoupled. Only for filling factors above $\nu = 1$ the two polariton resonances can be observed for this spin state, although, as previously mentioned, there is a persistent difference in the value of Ω . Here it is worth noting the feeble trace of the opposite spin reflection contrast that can be detected on each panel of Fig. 3.3. We associate it with three effects: imperfections in the polarization filters, linear birefringence of the cavity, and TE-TM splitting. Birefringence has the ability to change the system's polarization, and TE-TM splitting has shown

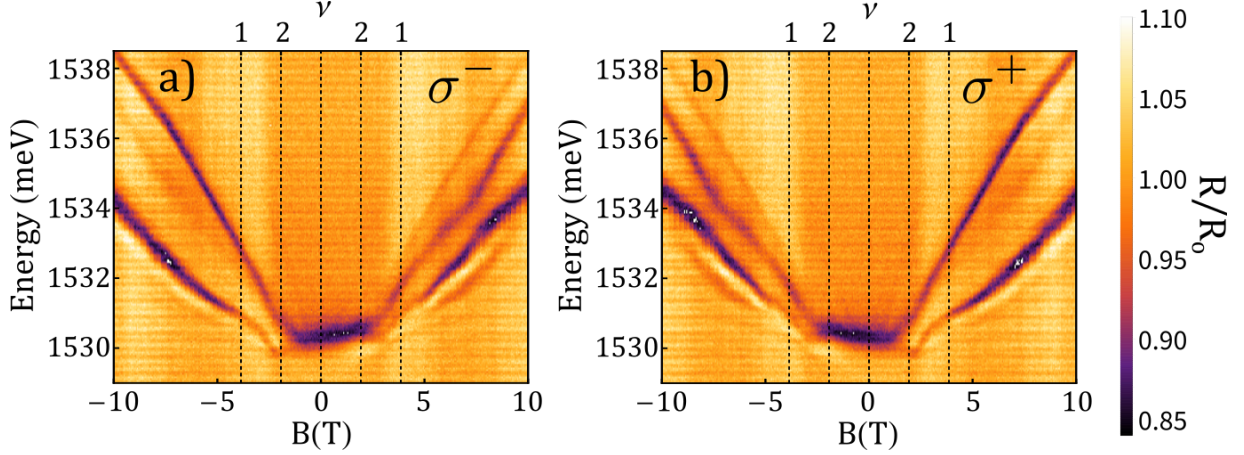


Figure 3.3: Magnetic field dependence of the system resonance at momentum at which the bare cavity and exciton energies cross. Due to both Zeeman splitting and diamagnetic shift, this crossing occurs at different momentum for each B . The values of B for filling factors 1 and 2 that delimit the three regimes of the system are indicated by black dashed lines for positive and negative values of B . The data is displayed for both spin states (a) σ^- , (b) σ^+ .

the capability of introducing intrinsic chiral behavior in the system [76].

To obtain a quantitative description of the spin-selective SC effect, we extract the values of ω_X and Ω from the described theoretical model for each magnitude of B and for each spin state. The results are displayed in Fig. 3.4. From the Rabi splitting value (upper panels), one can observe how the integer filling factors (marked on the top axis) coincide with the magnetic fields at which the populations σ^+ and σ^- reach the SC: $\nu = 2$ and $\nu = 1$, respectively. As previously mentioned, for $\nu < 1$ the asymmetry in Ω persists due to the h^+ population in the σ^- sub-band; we expect that for even higher magnetic fields, the difference will be asymptotically eliminated as $\nu \rightarrow 0$. Notice that for $\nu < 1$, Ω further increases for the σ^+ polariton resonances. This is due to the electron and hole confinement induced by the magnetic field that increases the oscillator strength and hence Ω [77]. Finally, for a fully depleted h^+ gas (panel c) the selectivity is completely removed, and both spin sub-bands are in SC with the cavity mode, in agreement with the expected behavior of a polaritonic system.

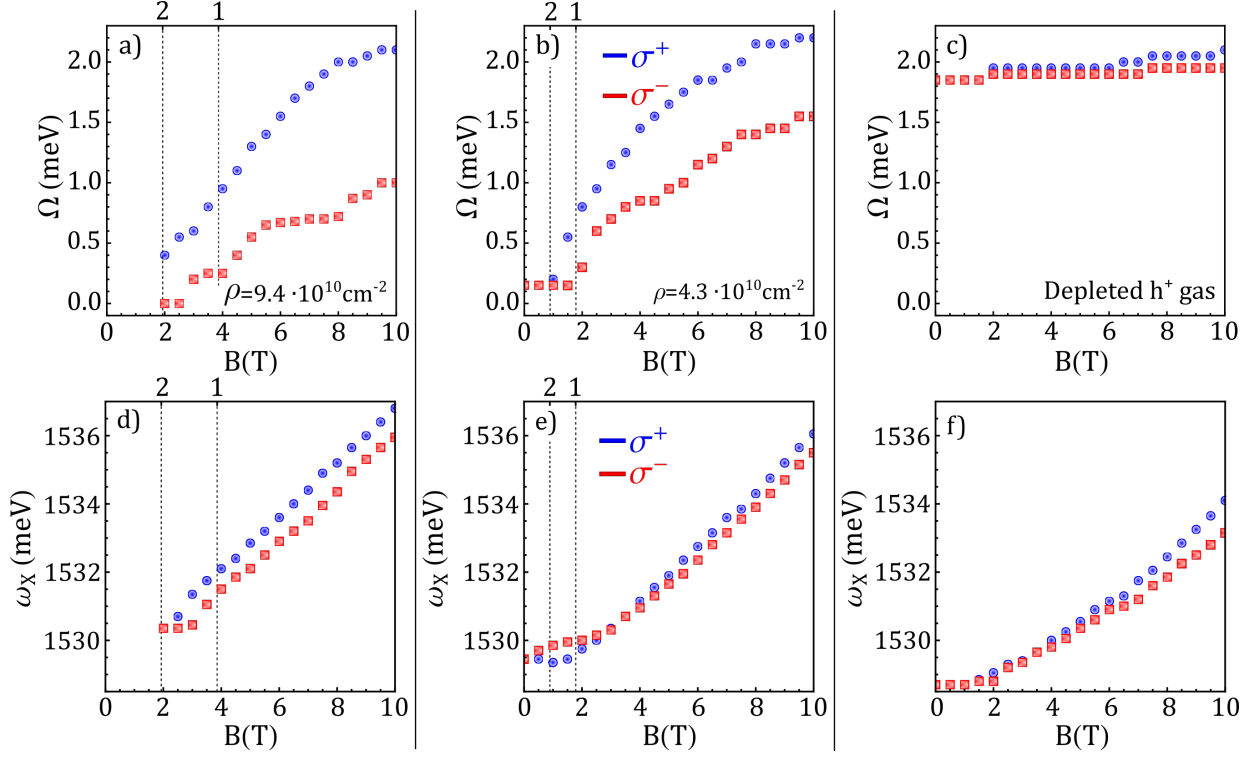


Figure 3.4: a-c) Rabi splitting obtained from the fitting to a polariton model (equation 3.1) for selected magnetic fields and for three different h^+ densities. d-f) Energy of the electronic transition obtained from the theoretical model for the same selected magnetic fields and densities. a) and d) $\rho = 9.4 \cdot 10^{10} \text{ cm}^{-2}$. b) and e) $\rho = 4.3 \cdot 10^{10} \text{ cm}^{-2}$. c) and f) fully depleted h^+ gas. As expected, the light-matter coupling selectivity is not observed in this last situation.

3.3 Discussion

The theoretical model (equation 3.1), in close agreement with the experimental observation, provides the value of the electronic transition energy (Fig. 3.4d-e). However, it is important to mention that a complete comprehension of the nature of this optically induced transition requires further experimental and theoretical investigation. Although the existence of this transition depends on the filling factor, its microscopic properties are determined by the interplay between: 1. electron-hole Coulomb interaction, 2. interaction with the 2DHG, and 3. interaction with the external magnetic field. Since none of these interactions dominates over the other two, a complete description of this polaritonic quasi-particle is not straightforward. Indeed, recent theoretical efforts are directed to investigate the nature of the light-matter coupling under these circumstances [78]. It is also important to highlight that although the nature of this quasi-particle is still an object of active research, the demonstration of the selective coupling to the cavity mode does not depend on it. Hence, the results presented in this work remain valid regardless of the nature of the matter excitation.

An asymmetry in the light-matter coupling has been reported for similar systems at lower charge densities when the 2DHG is in a spin-ordered state in the frame of integer or fractional quantum Hall effects [67, 68, 79]. In those cases, the asymmetry takes place for the specific values of B at which correlated states of matter dominate. This is not the case for the system in this work. In our case, any trace of correlated matter states at integer filling factors is ruled out by increasing the temperature (T) of the system to 3.5 K (in contrast to Refs. [67, 68] where T is kept at ~ 100 mK). The fact that the presented results are collected at a relatively high T (3.5 K) indicates that our observation is not related to many-body electronic correlations, highlighting the

robustness of this mechanism. Section 3.7 includes a comparison between the system's response at 40 mK and 3.5 K and section 3.6 shows the method used for calibrating the h^+ density that requires these ultra-low T .

3.4 Methods

3.4.1 Sample fabrication and preparation

Our sample consists of a $\text{Al}_{0.2}\text{Ga}_{0.8}\text{As}$ λ cavity embedded between two $\text{AlAs}/\text{Al}_{0.2}\text{Ga}_{0.8}\text{As}$ DBRs (24 bottom pairs and 19 top pairs) with a p-doping (Carbon) layer and a 16 nm GaAs QW, grown by molecular beam epitaxy. To control the h^+ density, a 60 nm Si-doped n-type GaAs layer is grown between the GaAs substrate and the bottom DBR. This acts as a gate that can control the density of the 2DHG. Contacts were fabricated using In for the n-type GaAs gate and $\text{In}_{0.96}\text{Zn}_{0.04}$ for the 2DHG. The device's h^+ density can then be controlled by applying a gate voltage.

This sample was fabricated at ETHZ and shipped to us at the University of Maryland. It had to be transferred to a sample holder that could be used in our systems and the wiring for the contacts had to be re-made, see Fig. 3.5a-b. To verify that it survived this process and that the backgate was still working, we tested the resistance between QW contacts and measured the current between the back gate and QW as a function of voltage. Since the structure forms a P-I-N junction, we expect to see no current when the structure is reverse-biased and an increasing current when the system is forward-biased. This is what we observe in Fig. 3.5c.

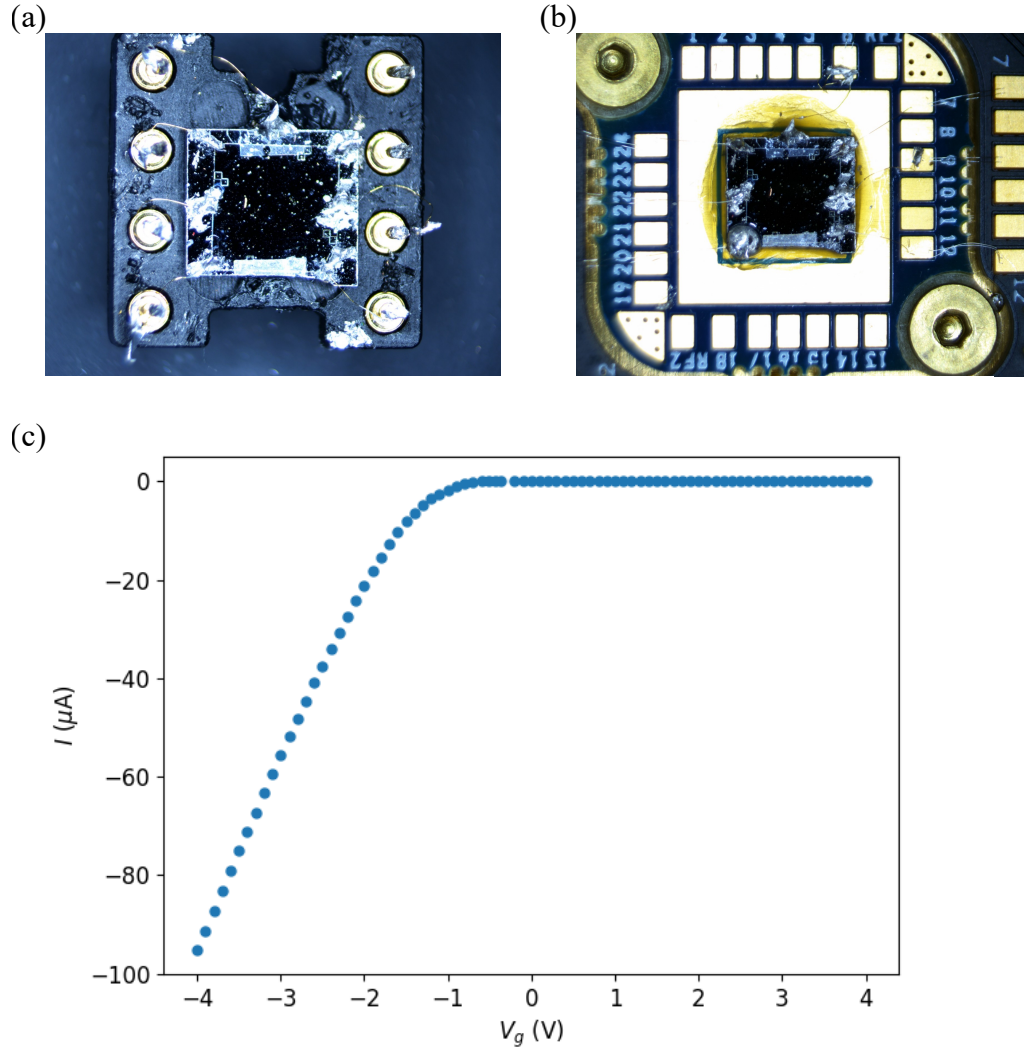


Figure 3.5: GaAs structure optical images and I-V curve. (a) Microscope image of the sample after receiving it from ETHZ. (b) Microscope image of the sample after transferring it to one of our sample carriers and re-wiring it. The sample is adhering to the carrier with thermal grease. (c) Measured I-V curve between a QW contact and a back gate contact. The low current at positive voltages and increasing current at negative voltages verifies that the back gate is functional since the structure is a P-I-N junction.

3.4.2 Setup for optical measurements

The sample is placed in a dilution refrigerator with the capability of reaching temperatures down to 40 mK. Although all the measurements presented in the manuscript are taken at a temperature of 3.5 K, we perform measurements at a base temperature of 40 mK to calibrate the h^+ density, using the optical signature of correlated states at integer filling factors. The comparison of the data taken at base temperature and 3.5 K as well as the details of the density calibration procedure can be found in sections 3.7 and 3.6, respectively.

For the optical measurements, we use a confocal setup in reflectivity configuration with numerical aperture $NA = 0.5$. A tungsten lamp is employed as a broadband light source, but consistent results were obtained using a supercontinuum white light source. Polarization-resolved spectra were collected by placing a linear polarizer followed by a quarter wave plate in the excitation path.

3.5 Spin selection rules for optical transitions

A fundamental condition, necessary for the spin-selective strong coupling reported in this work, is the existence of spin-selection rules that determine the optical transitions that can take place in the system. In this section, we will discuss the nature and origin of these selection rules, mostly based on Ref. [75]. They originate from the dipolar nature of the light-matter interaction, which in turn arises from the spin conservation in photoabsorption events. Due to the strong spin-orbit coupling, the valence band (VB) energy is non-degenerate in quantum wells with a Zincblende crystalline structure. The VBs are composed of a band with a total angular momentum $J_z^h = \pm 3/2$ usually referred to as the “heavy-hole” (HH) band and one with $J_z^h =$

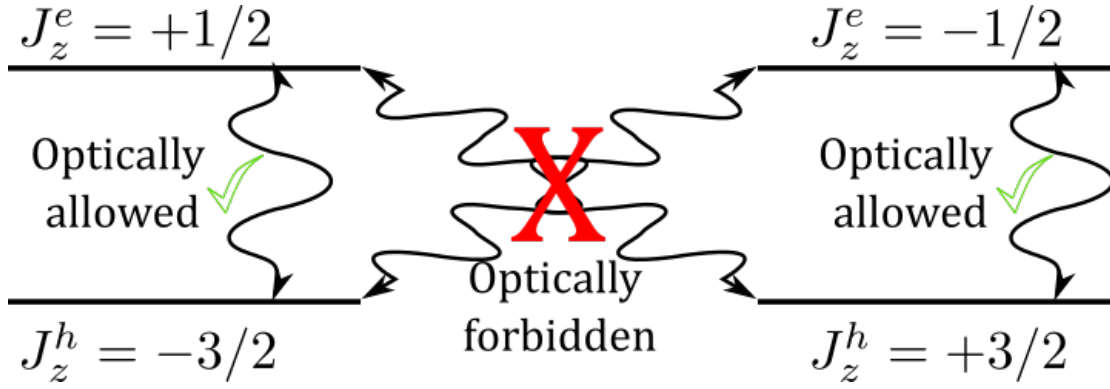


Figure 3.6: Optical selection rules for a GaAs quantum well for transitions between the HH band and the CB.

$\pm 1/2$, referred to as “light-hole” (LH) band. It is the relative orientation of the spin and the orbital angular momentum which determines the nature of the hole band. In typical GaAs structures, the HH band is closer to the conduction band (CB) than the LH band, hence, in this analysis, we focus on the lowest energy optical transitions, i.e., those between the HH band and the CB.

The optical transition has a total angular momentum projection corresponding to the projection of the addition of angular momentum of electrons ($J_z^e = \pm 1/2$) and HH ($J_z^h = \pm 3/2$). This transition can then have projections of the total angular momentum $J_z^T = \pm 1, \pm 2$. The angular momentum conservation implies that only those transitions matching the photon’s angular momentum ± 1 can take place; hence, only the configurations in which the projections of electron and hole are anti-parallel are optically allowed. Typically, for a neutral QW system, the exciton corresponding to this projection is called “bright” exciton, in contrast to the $J_z^T = \pm 2$ case, referred to as “dark” exciton.

These selection rules determine the existence of spin selectivity in the strong coupling. Figure 3.6 shows a diagram of the optical selection rules. It shows how the transition from the spin up (down) VB to the spin up (down) CB is optically forbidden, which justifies why these transitions are not indicated in Fig. 3.1b-d.

3.6 Hole density calibration

At a temperature of 40 mK, the hole gas is capable of sustaining correlated states of matter at integer filling factors. This result has previously been reported [68]. This gives a very accurate method for calibrating the hole density: since the magnetic field strength and the gate voltage are known quantities, one can infer the hole density from the optical signatures of the integer filling factor states $\nu = 1$ or $\nu = 2$. According to quantum Hall physics, these correlated states are established when $\rho h/eB \in \mathbb{Z}$. Depending on ρ , one can observe both correlated states, one of them, or none. This limitation is imposed by the finite hole mobility in the sample. Figure 3.7 shows the reflectivity spectrum of the system at low numerical apertures, i.e., close to zero in-plane momentum, as a function of the magnetic field. After systematically collecting data for different gate voltages, and recording the magnetic fields at which the Rabi splitting is modified, one can obtain an accurate linear model that gives the hole density of the system as a function of the gate voltage.

The density is not perfectly uniform across the sample; we performed this calibration in different spots and obtained different results. For this reason, it is important to perform a calibration procedure every time the position of the excitation spot on the sample changes. Figure 3.8 shows the deduced hole density for each given gate voltage at a given spot of the sample. The procedure consists of registering the magnetic field at which the Rabi splitting is modulated for a given gate voltage. With this information, we infer the hole density according to the formula $\rho = eB\nu/h$, with ν taking the values 1 or 2. Notice from the figure that for some gate voltages both integer filling fractions are displayed, because in those cases the optical signatures of both states are observable. This corresponds, for example, to the case displayed in Fig. 3.7a.

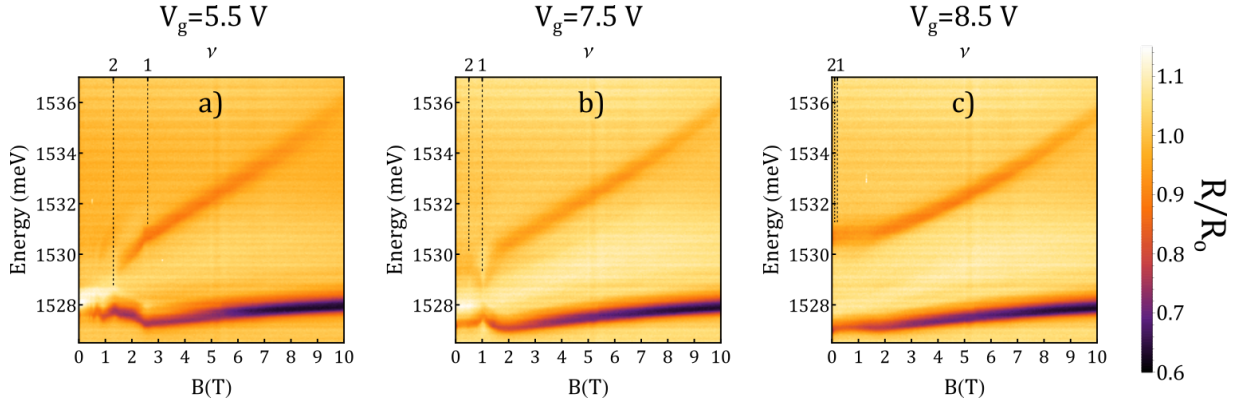


Figure 3.7: Differential reflectivity of the system at 40 mK. The displayed data corresponds to low numerical apertures ($k \sim 0$). The gate voltage sets the hole density and a sweep over the magnetic field allows us to identify the correlated states of matter associated with integer filling fractions. Due to the finite hole mobility of the sample, the effect is not visible for every density. In the figure, one can observe that for high densities, the modification of the absorption spectrum corresponding to $\nu = 1$ and $\nu = 2$ can be observed (panel a). For medium densities, only $\nu = 1$ shows the feature (panel b) and for low densities, none of the correlated states is observable (panel c). The top axis indicates the magnetic fields corresponding to integer filling fractions obtained from an interpolation of the results.

For other gate voltages, only one of the correlated states can be detected as shown, for example, in Fig 3.7b. However, for very high or very low densities, none of the correlated states can be observed (Fig. 3.7c). After recording the calculated density for each gate voltage, we use the best-fitting linear function as a reference for the system's density once the temperature is increased from 40 mK to 3.5 K. Finally, it is worth mentioning that we did not detect any optical signal related to fractional quantum Hall states in the full range of charge density. The formation of these states requires very high charge mobility (μ). For example, the reported fractional states in polaritonic microcavities [67] were achieved in a system with $\mu = 1.6 \cdot 10^6 \text{ cm}^2 \text{V}^{-1} \text{s}^{-1}$, which according to our characterization, is at least 5 times higher than the mobility of the sample employed in the present work.

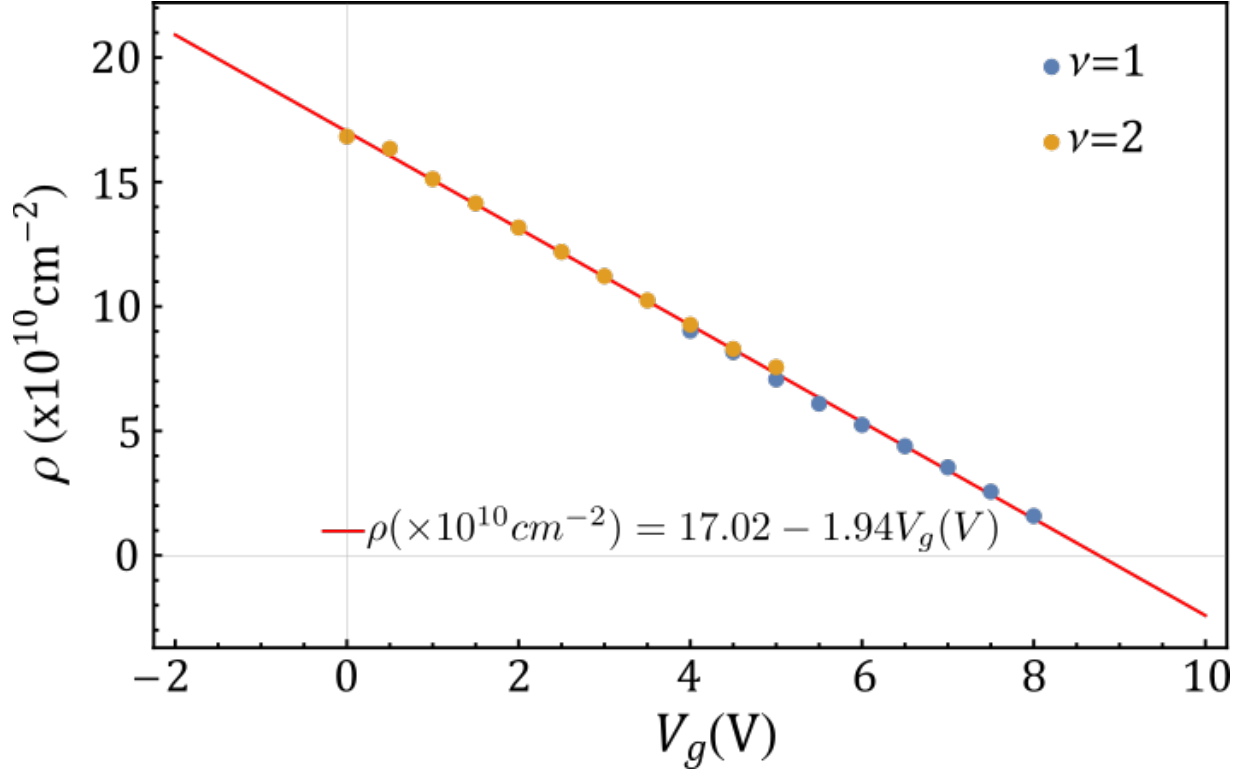


Figure 3.8: Voltage to hole density calibration. From the low momentum reflectivity spectrum, we extract the values of the magnetic fields for which the correlated states of matter take place for each voltage. In some cases both ($\nu = 1$ and $\nu = 2$) states are observable, in some cases only one of them is, and for very high and very low densities, none of the correlated states are observable. The red line shows the fitting line for the data set, which we use as a reference to obtain the hole density at 3.5 K. The range of the horizontal axis corresponds to the range at which we collected data.

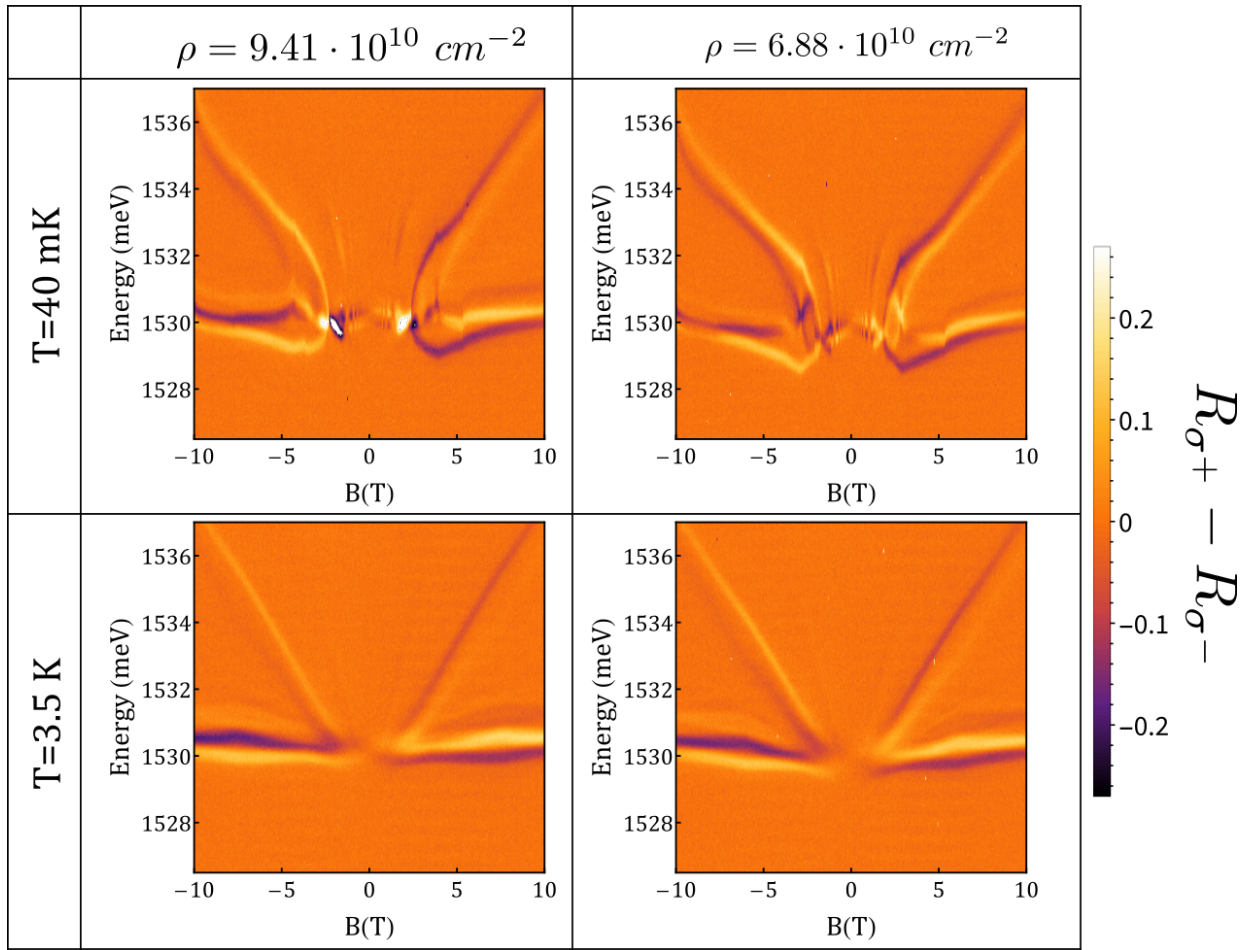


Figure 3.9: Difference of the reflectivity spectra for the two circular states of polarization. The data is presented for two densities ($9.41 \cdot 10^{10} \text{ cm}^{-2}$ and $6.88 \cdot 10^{10} \text{ cm}^{-2}$) and for two temperatures (40 mK and 3.5 K). The modulation of the resonance energy peak associated with correlated states of matter is visible only for the data collected at the dilution unit base temperature (40 mK).

3.7 Comparison of the system response at $T = 40$ mK vs $T = 3.5$ K

In this section we discuss the fundamentally different response of the physical system when probed at 40 mK or 3.5 K. This is important because the previously reported modulation of the Rabi splitting [66–68] relies on correlated states of matter that can only take place at these ultra-cold temperatures, while the effect we exploit to induce the spin-selective strong coupling does not rely on this physics. The optical features from the correlated states at integer filling fractions are completely washed out when the temperature is 3.5 K. A magnetic field dependence was collected in identical conditions at 40 mK and 3.5 K for two densities: ($9.4 \cdot 10^{10} \text{ cm}^{-2}$ and $6.88 \cdot 10^{10} \text{ cm}^{-2}$). The results, displayed in Fig. 3.9 respond to the difference of the reflectivity spectra for σ^+ and σ^- polarization collected with low numerical aperture. The subtraction of the reflectivity spectra allows us to show both of the spin (polarization) states in a single frame. In contrast with the data presented in Fig. 3.3, the in-plane momentum is constant over this set of data ($k \sim 0$).

Figure 3.9 features a much richer behavior at a temperature of 40 mK with respect to the 3.5 K case. For example, at specific values of the magnetic field, the Rabi splitting is strongly modified. While the overall splitting of different polarization branches can be understood using our single-particle picture, the finer features require more investigation. Generally, one can associate such features with Coulomb interaction in creating correlations at such low temperatures. For example, at $\nu = 1$, certain skyrmion physics can be observed similar to the earlier work by ETHZ [68]. At mK temperatures, the thermal energy is low enough to allow the establishment of these Coulomb-induced correlations that go beyond the Pauli exclusion. On the other hand, at higher temperatures (3.5 K, shown in the lower panels of Fig. 3.9), the thermal excitations

have enough energy to bring the system into excited states, which consist of delocalization of the charges and destruction of the spin order. As pointed out previously, the spin selectivity reported in this work does not rely on the Coulomb-induced spin order, but its robustness depends on the high Zeeman splitting between LLs with opposite spin.

There are two energies that, when compared with T , determine the ground state of the system: the Coulomb interaction and the Zeeman splitting between LLs. In the previous paragraph, we established that at 3.5 K, the thermal fluctuations destroy the localization and magnetization induced by charge repulsion, for which we will focus now on the second quantity: Zeeman splitting vs. T . For a system following the Fermi-Dirac distribution, one would expect T to challenge the spin selectivity by inducing thermal transitions in the hole gas. In this scenario, the hole population at the σ^- level could be excited to the σ^+ level; or in other words, T would induce electronic transitions from σ^+ to σ^- . The consequence would be the loss of spin selectivity, because now both bands would have electrons available to perform an optical transition. Although we cannot measure the band structure of the system, the robustness of the effect indicates that the energy splitting between the σ^+ and σ^- levels is higher than the thermal energy ($T=0.3$ meV at 3.5 K). This allows us to neglect the effect of T in our description. It is important to mention that the energy of the LLs of the valence band has a non-trivial dependence on both the magnetic field and the charge density, as it can be observed in reference [72]. Therefore, the energy splitting between the spin sub-bands cannot be easily estimated.

3.8 Details on the theoretical formalism for data fitting

The coupled oscillators formalism employed for the theoretical modeling of the polaritonic system is described by equation 3.1. Once collected, we fit the data by using ω_X and Ω as fitting parameters for each magnetic field. The data is then fitted to the eigenfunctions of the Hamiltonian which are given by:

$$E_{U/L} = \frac{1}{2} \left(\omega_X + \omega_c + \frac{k^2}{2m} \right) \pm \frac{1}{2} \sqrt{\Delta_k^2 + 4\Omega^2} \quad (3.2)$$

with $\Delta_k = \omega_c + \frac{k^2}{2m} - \omega_X$. The cavity's energy and effective mass is accurately extracted from the case $\Omega = 0$ since in this condition, only the bare cavity mode is visible. For the mass, we obtain the value $m = (2.4 \cdot 10^{-5})m_e$, with m_e the free electron mass. This value is kept constant over the fitting procedure of the full data set and it is consistent with typical values for these devices. The cavity energy accounts only for minor changes due to the response of the optical properties of the bare electromagnetic mode to the high magnetic and electric fields. In the absence of any field, the cavity mode at 0 in-plane momentum has energy $\omega_C = 1530$ meV, and for the full set of data (0 V to 10 V and -10 T to 10 T), it never changes by more than 0.5 meV.

3.9 Summary

In summary, we introduced an experimental method for spin-selective light-matter coupling in a microcavity-quantum well system through the modification of the hole density and the strength of the applied magnetic field. This result opens perspectives on the control of non-linear effects in light-matter hybrid systems. For example, in exciton-polariton systems, the $\chi^{(3)}$ optical

nonlinearity could be easily controlled using the polarization of the light. As shown in Fig. 3.9, the spin selectivity persists even at lower temperatures (~ 40 mK), where correlated states of electrons are expected, such as fractional quantum Hall states. However, samples with higher mobilities are required to further investigate the interplay between spin selection and correlated states of electrons.

Importantly, the application of this method is not limited to III-V semiconductor devices. One potential alternative platform is TMD materials where spin and valley degrees of freedom are locked. This method, combined with the possibility of embedding a monoatomic semiconductor in a 2D microcavity [80] opens interesting perspectives for the valley selective light-matter SC previously reported in the literature [69]. Such versatility is also expected to unlock the experimental realization of recent theoretical proposals for the optical study and manipulation of light-matter interaction in different 2D correlated matter systems. Although this perspective is very promising, a deep theoretical understanding of these phenomena is still under development [7, 47, 81]. Overcoming both the technical and conceptual challenges promises to pave the way for the investigation of this exotic physics. Our system might also enable technological implementations such as variable circular polarization filters with ultra-narrow bandwidth, and from a more fundamental point of view, it is a suitable platform to optically study the spin physics of 2D gases of charged fermions, thanks to the degree of control on the light-matter interaction that can be achieved by modifying the h^+ density.

Appendix A: Graphene device fabrication

A.1 Heterostructure fabrication

Before receiving the Corbino device used for the experiment in Chapter 2, I spent a significant amount of time fabricating graphene devices. These techniques can also be applied to the fabrication of other 2D materials samples. This appendix will review this fabrication process.

The process begins with the mechanical exfoliation of graphene and hBN flakes onto Si/SiO₂ (285 nm) substrate. The substrate is precleaned first in acetone and then in isopropanol. When trying to exfoliate monolayers, I often also cleaned the substrate with a piranha etch or oxygen plasma to remove residuals of organic compounds prior to exfoliation. The stacking process is depicted in Fig. [A.1](#). Stacking these flakes together requires making a stamp from polydimethylsiloxane (PDMS) and a poly(bisphenol A carbonate) film. A PDMS hemisphere is formed by first cutting out a small square of a flat PDMS film, either made in the lab or from commercially available PDMS. Then using Sylgard 184 form a small droplet on top of the PDMS square and let it cure overnight. The surface tension of the droplet should cause it to cure in a hemisphere shape. It is not necessary to use a hemisphere for stacking, flat PDMS will also work, however, this provides the benefit of having a smaller more precise contact area with the substrate while stacking. An example of the result of this stacking can be seen in Fig. [A.2a-e](#) where four flakes are stacked together to make an hBN encapsulated monolayer graphene stack

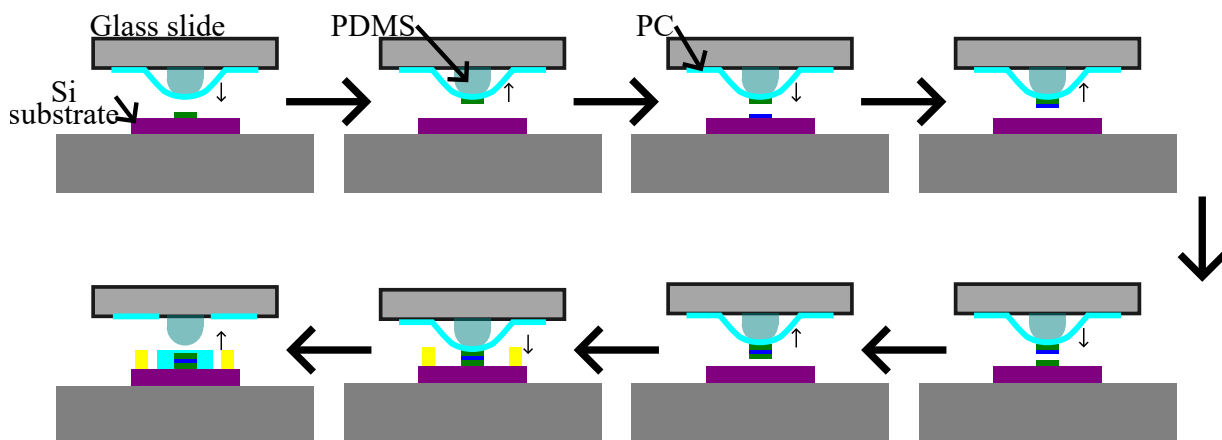


Figure A.1: Schematic showing the stacking process. A glass slide with a stamp made from a PDMS hemisphere and a PC film is used to sequentially pick up each layer of the structure. All of these steps except for the last two are performed by making contact with the flake at a temperature between 50°C and 70°C. The subsequent pickup step can either be performed by manually retracting the glass slide or by letting the substrate and poly(bisphenol A carbonate) film cool, which causes the film to contract. In the last two steps the poly(bisphenol A carbonate) film with the stack is melted onto a substrate with alignment markers at 180°C. The poly(bisphenol A carbonate) is then removed in a chloroform bath. This schematic is only for a three layer stack. More steps will need to be added for additional layers.

with a graphite gate.

The stack is deposited on a substrate with alignment markers. The alignment markers allow us to use electron-beam lithography (EBL) to further process the sample. At the University of Maryland campus we have access to two EBL tools: the Elionix GS-100 and the Raith e-LiNE. We primarily use the Elionix, which has a fixed acceleration voltage of 100 kV. However, when investigating if I could improve the quality of the graphene sample contacts, I tried the Raith, which has a variable acceleration voltage between 10 kV and 30 kV. The hypothesis was that the higher-energy electrons from the Elionix were damaging the graphene in the contact regions, however, this did not show any improvement. Unfortunately, I was only able to test this once, so it may be worth testing again if more of these samples need to be fabricated.

EBL can then be used to define the sample geometry by writing an etch mask into resist.

The resist used in this step for graphene samples is typically two layers of polymethyl methacrylate (PMMA) 950A4. Each layer is spin-coated onto the sample at 4000 rpm for one minute. The first layer is baked at 180°C for five minutes and the second layer is baked at the same temperature but for ten minutes. The resist is exposed in the EBL system and then developed in a solution composed of water and isopropanol, mixed in a ratio of one to three by volume. After the etch mask is made, the sample can be etched using a dry etch process. I used two types of dry etching processes for these samples: selective and non-selective etching. We will review the difference in Section [A.2](#).

The result of the etching can be seen in Fig. [A.2f](#), where three Corbino devices of different sizes have been etched into the sample. To finish the fabrication process, metal contacts must be deposited. These will form one-dimensional edge contacts to the graphene [\[82\]](#). This process starts in the same way as the etching process in that a pattern must be written into the resist using EBL. For the deposition step, the same resist can be used; however, an alternative is to substitute the first layer with PMMA 495A8. Doing this will cause an undercut to form in the resist after it has been exposed and developed. The undercut can make liftoff of the metal easier. After the pattern for the contacts has been formed in the resist, the sample is loaded into a thermal or electron-beam evaporator. This consists of a vacuum chamber loaded with the materials needed for deposition. In the thermal evaporation process, a current is run through a boat or rod that holds the material, and the heat causes the material to evaporate and then deposit onto the sample. In the electron-beam evaporation process, the material is held in a crucible, and a focused electron beam causes the material to heat up and evaporate. For graphene samples, we typically use 3 nm of chromium, which acts as an adhesion layer, and 70 nm of gold. However, in Section [2.7.1](#), only gold is used. The deposition is uniform over the entire substrate, which is why lithography was

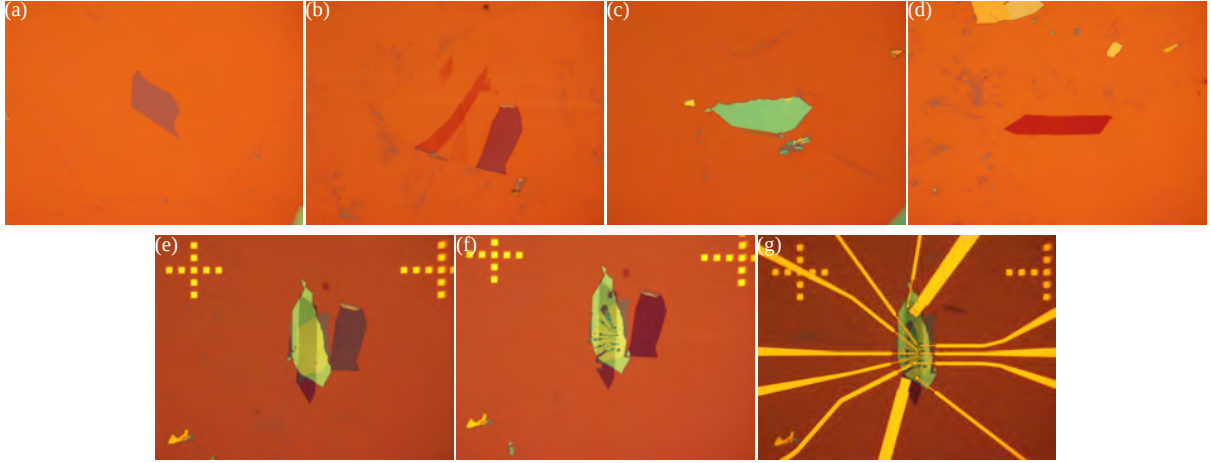


Figure A.2: Steps in the graphene fabrication process. (a)-(d) Are the flakes that are used for the stack where (a) is the top hBN, (b) is the monolayer graphene, (c) is the bottom hBN, and (d) is the graphite used for the back gate. (e) The stack after being deposited onto a substrate with alignment markers. For reference each alignment marker square has a width of $5\ \mu\text{m}$. (f) The sample after the sample geometry is etched into the stack. (g) The sample after contacts composed of 3 nm of chromium and 70 nm of gold are deposited.

needed. The sample is then placed in acetone to remove the PMMA and excess metal, leaving the deposited metal only in the regions that had previously been exposed in the EBL step. This process is called liftoff. In Fig. A.2g, the metal contacts have been formed for the three Corbino devices, including the graphite back gate.

A.2 Etching techniques

The first graphene devices that I fabricated used a non-selective etch recipe based on CHF_3 and O_2 . This recipe was based on the one developed in [82]. This was performed using the Trion RIE and the Oxford ICP etcher (fluorine) in the Maryland Nanocenter. For devices that incorporated a graphite or metal back gate rather than a Si back gate, this technique became problematic because, if not timed properly, it would etch through the entire heterostructure, making it impossible to make contacts to the sample that did not short-circuit to the back gate. An example of

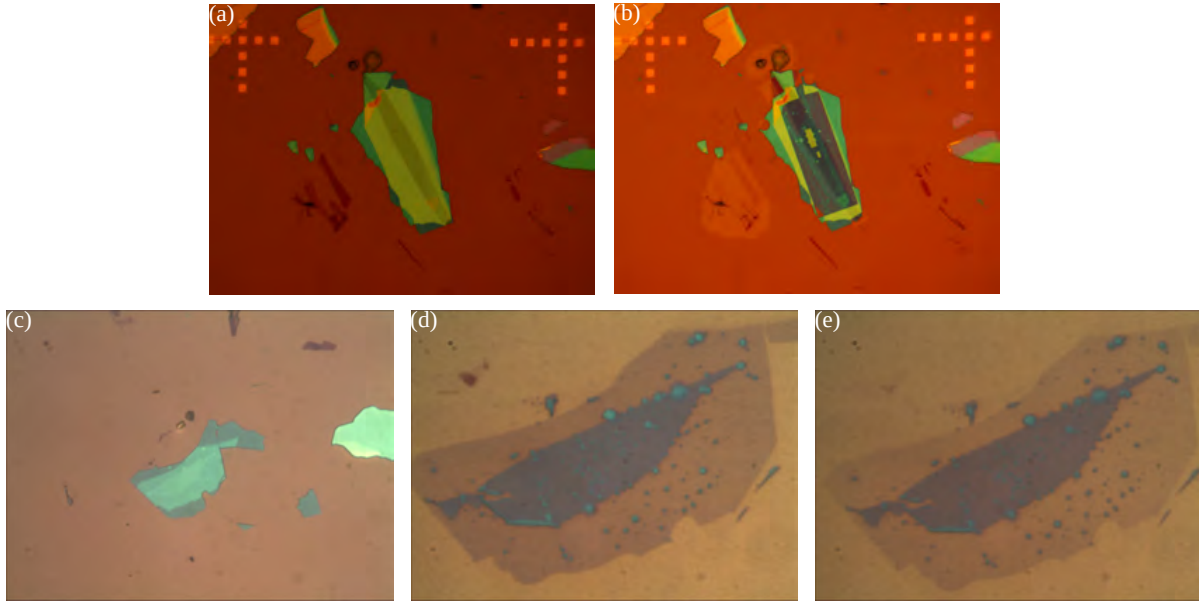


Figure A.3: Graphene etching examples. Graphene heterostructure etched with a non-selective etch recipe based on CHF_3 and O_2 (a)-(b). Graphene heterostructure first etched with an etch recipe based on SF_6 which is selective to hBN over graphene and then etched with a recipe based on O_2 and Ar which is selective to graphene over hBN (c)-(e).

this can be seen in Figs. A.3a-b. In this device the etching was timed properly so that the contacts could be formed without short-circuiting to the back gate; however, after several attempts, I determined that a reliable etch rate could not be found; it varied too much from sample to sample.

An alternative to the non-selective etching process is to use a selective etching process. A selective dry etch process can be made by using a different fluorine based compound than CHF_3 , by increasing the pressure in the etch chamber, and using a low power for etching. The effect of increasing the pressure of the etch chamber and using low power is to decrease physical etching, which is a non-selective process. This leaves chemical etching, which, depending on the compounds used for the etching can be selective [83]. Recipes based on SF_6 and CF_4 have been developed [34, 83]. Both of these recipes work based on the idea that the SF_6 or CF_4 etch will etch through hBN, but will form fluorinated graphene, which resists fluorine-based etching. The fluorinated graphene can then be etched away with an O_2 plasma etch, and edge contacts can

be made. However, for the sample described in Section 2.7.1, the gold contacts were deposited directly on the fluorinated graphene. I was able to make etch recipes based on both Refs. [34] and [83]. In Figs. A.3c-e, the etching was performed by first using a selective SF₆ etch on the hBN-encapsulated graphene and then a selective O₂ and Ar etch of the graphene. For both of these recipes to work, the etcher needed to run a cleaning cycle before etching.

Appendix B: Photocurrent setup modifications

When performing the experiment discussed in Chapter 2, we found that alignment of the beam with the sample was crucial to collecting reliable data. As can be seen in Figs. B.1a-b, the image quality in the measurement system was not very good. There were always reflections of the sample image present. To go around this we needed to perform time-consuming spatially resolved photocurrent measurements. For future work, we implemented changes to the optical setup.

A schematic of the probe before making any modifications is shown in Fig. B.1c and a schematic of the probe after making modifications is shown in Fig. B.1d. In the original setup light is focused onto the sample using an aspheric lens, however, the aspheric lens is about 1.4 m from the top of the probe. To improve the imaging quality as well as decrease the spot size of the focused beam, we wanted to install a high NA Attocube cryogenic objective as well as two plano-convex lenses in the probe to reconstruct and collimate the image from the objective.

Making these modifications required designing custom parts to hold the objective, which has a larger diameter than the tube which holds the aspheric lens. The designs of these parts are shown in Figs. B.2a-b, and a picture of the assembled parts with the objective is shown in Fig. B.2c. This assembly is composed of a circular holder piece that connects with four pins with 8-32 threads. These pins then connect to the probe rods. The objective is screwed into

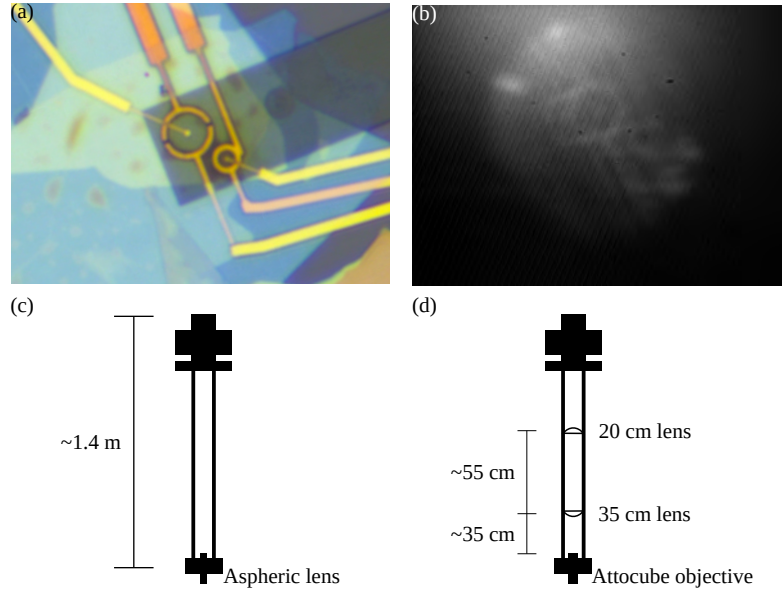


Figure B.1: Photocurrent setup imaging before modifications. (a) Optical microscope image of a Corbino sample. (b) Image of the same sample taken in the photocurrent measurement setup. (c) Schematic of the probe before modifications. There is approximately 1.4 m between the window at the top and the aspheric lens at the bottom. (d) Schematic of the probe with the modifications. The aspheric lens is replaced with an objective and two lenses are mounted in the probe.

a cylindrical adapter component using RMS threads and is connected to the holder with a set screw. To install the plano-convex lenses, we needed to design a part that could be installed without changing the wiring of the probe. The designed part as well as the mounted lenses are shown in Figs. B.2d-e. The lenses could only be mounted in a small number of places on the probe, so it was determined that the best lenses would be a 35 cm and a 20 cm lens, as depicted in Fig. B.1d. The lens mounts are attached to the probe using titanium m3 screws. The parts were designed using Autodesk Inventor and machined in grade 5 titanium.

The results of these modifications are shown in Fig. B.3. As we can see, the image quality is greatly increased, there is no more doubling of the image, and the reflection of the vortex beams also looks good. The only downside is that since the working distance of the objective is so small (0.7 mm) it is more difficult than before to find the correct sample position which corresponds to

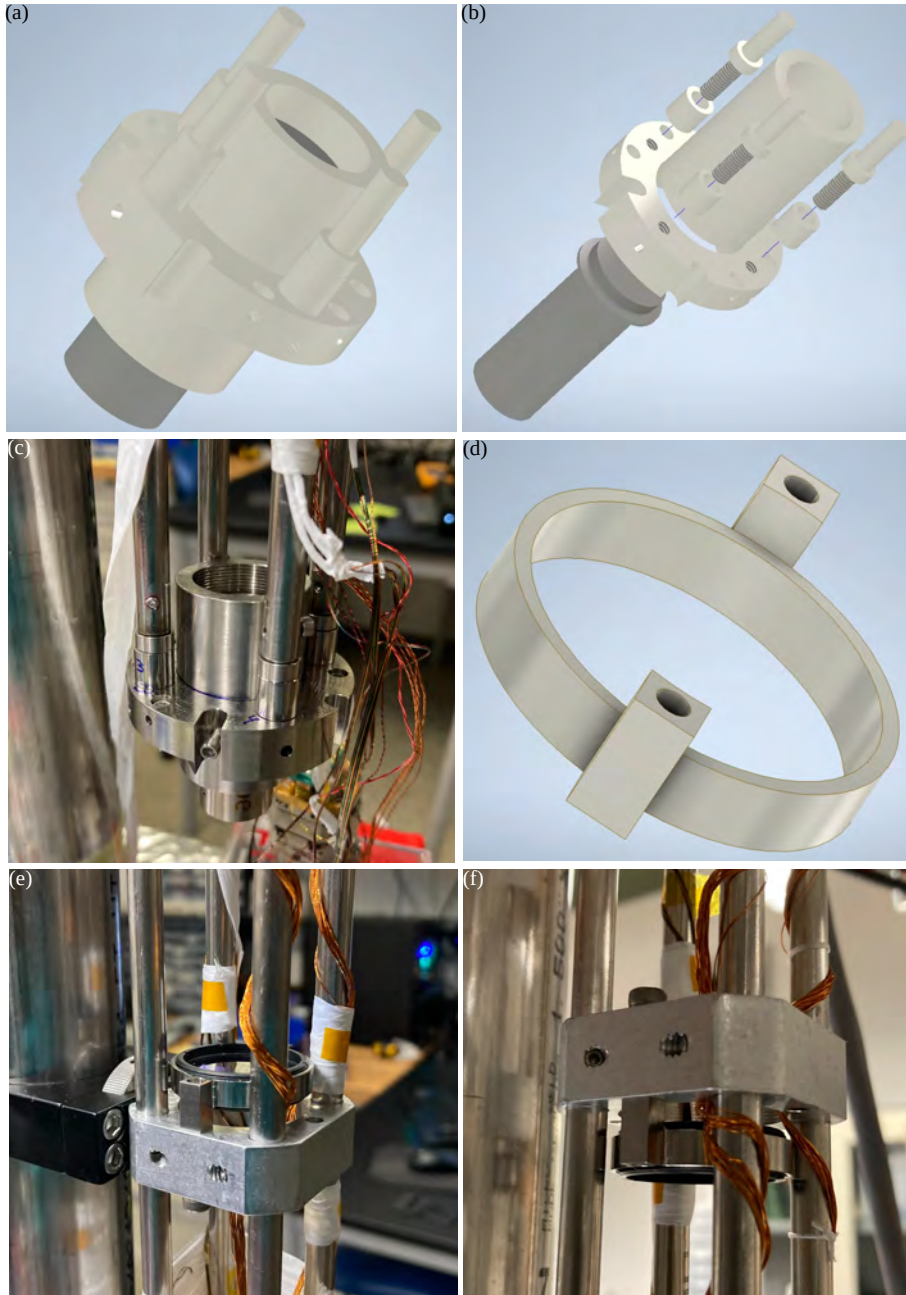


Figure B.2: Custom parts made to modify the photocurrent measurement setup. (a) Assembly of parts designed to attach an Attocube cryogenic compatible objective to the system probe. This consists of four pins connecting the rods of the probe to the objective holder, four washers for the pins, and an adapter to hold the objective in the designed holder. (b) An exploded view of (a). (c) Picture of objective and designed objective holder attached to the probe. (d) Custom designed lens holder. The lens holder also features SM1 internal threads which are not shown in the figure. (e) 35 cm plano-convex lens mounted using the custom lens holder. (f) 20 cm plano-convex lens mounted using the custom lens holder.

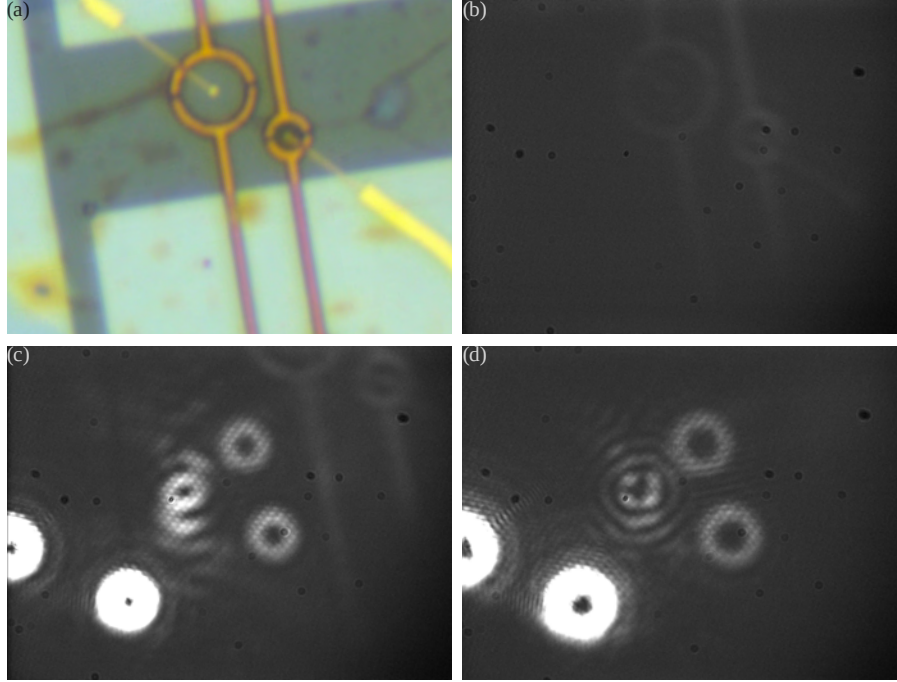


Figure B.3: Imaging with the modified photocurrent setup. (a) Optical microscope image of a Corbino sample. (b) Image of the same Corbino sample taken using the modified photocurrent setup. (c) Image of a beam with an OAM of +1 next to the sample. There are also four reflections from other optical components in the setup. (d) The same as (c), but with a beam that has an OAM of +2.

the working distance than it was when using the aspheric lens which has a focal length of 3.1 mm.

Still, this should improve the data acquisition process for future experiments.

Appendix C: Bluefors 2023

C.1 Wiring

The Bluefors 2023 probe is equipped with a total of 48 DC lines, which are separated into two sets of 24. One set is designated for the DC lines which are used for the sample contacts, and the other is designated for the Attocube stack. Inside of the fridge the 24 lines designated for the sample's DC lines are composed of copper wire from the top of the fridge to the 4 K plate and are NbTi/CuNi from the 4 K plate to the probe. Inside of the probe, we used 32 AWG single-strand wire from LakeShore. The other 24 lines that are for the Attocube stack use Attocube wiring from the top of the fridge through the probe.

The connectors on the top of the probe and bottom of the mixing chamber flange are micro-D25 connectors, while the connectors on the room temperature plate are 24-pin Fischer connectors. The Fischer connectors and micro-D25 connectors use different number conventions, thus it is important to know how each DC line and Attocube line corresponds to the pins on the Fischer connectors and the pins on the micro-D25 connectors. The pin out for the DC lines and the Attocube lines are given in Tables [C.1](#), [C.2](#), [C.3](#), [C.4](#), [C.5](#), and [C.6](#), respectively.

For the external wiring two Fischer cables are used to connect the fridge wiring to the breakout box. The breakout box connects to BNC connectors which can be used to connect electronic equipment to the sample and also to the Attocube controller (ANC 300). The BNC

connectors for the DC lines are also equipped with toggle switches which allow each line to be grounded when not in use. Pictures of the breakout box are shown in Fig. C.1.

C.2 Internal optics

When the Bluefors was installed it was equipped with ports in which we could install windows and the top of the probe was designed with a hole in the center so that light could pass through it. However, we needed to install a 45 degree mirror to direct light to the probe and install components in the probe such as a lens, an objective, and the Attocube stack. This required the design and installation of custom parts. These parts are similar to the ones we used for Bluefors 2017, however, they needed to be adjusted due to the difference in probe size. The parts used in Bluefors 2017 are discussed in Ref. [84]. The Bluefors 2017 probe was designed to hold 1” optics while the Bluefors 2023 probe was designed to hold 2” optics. These components were machined out of copper and gold plated. All of them except for the 45 degree mirror mount are installed in the probe. The 45 degree mirror mount, as shown in Figs. C.2a-b is installed on the cold plate, which is located between the mixing chamber flange and the still plate. The probe with all installed components is shown in Figs. C.2c-d.

C.3 Differences between Bluefors 2023 and Bluefors 2017

Apart from the difference in the size of the optics and probe, which was discussed in the previous section, there are other differences in the loading/unloading procedure as well as the Attocube stack. In Bluefors 2017, as discussed in Ref. [84], the bottom loading fast sample exchange system (FSE) has three feet that are used to lift the loading mechanism up to the fridge



Figure C.1: Bluefors 2023 breakout box. Inside of the breakout box showing the wiring from the Fischer connectors to the BNC connectors for the Attocube lines (a), and the wiring for the BNC connectors for the DC lines (b). (c) The breakout box mounted on an electronics rack outside of the fridge.

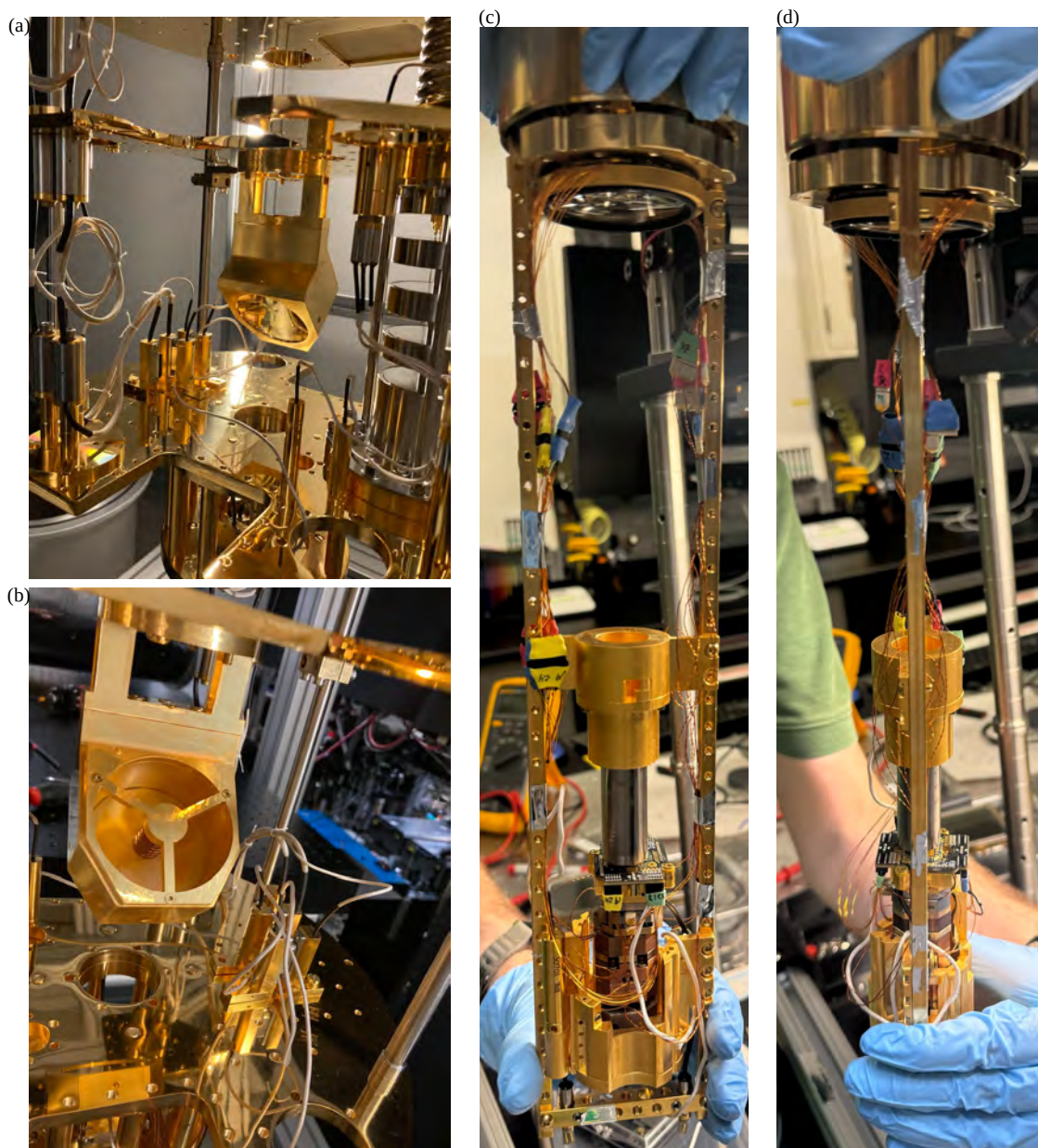


Figure C.2: Bluefors 2023 internal optics. 45 degree mirror mount attached to the cold plate (a) front and (b) back. (c) Front view of the probe with the lens, objective, Attocube stack, sample, and wiring installed. (d) Side view of the probe.

or lower it back to the ground. The Bluefors 2023 uses a new way to do this. The FSE is fitted with two latch clamps that can be used to raise or lower the probe loading system. Another difference is that the cables that connect to the temperature sensor and heater are shorter. Due to these two differences, the loading/unloading procedure is modified.

In Fig. C.3 we review the unloading procedure. First, the M8 screws that secure the FSE to the gate valve are removed. Then the FSE is vented by opening the pump valve, V19, and V20 and then injecting nitrogen using the needle valve shown in Fig. C.3c. The FSE is then removed from underneath the fridge and moved up using the home in button until the screws securing the sample space tube are accessible. These screws are then removed, and the sample space tube is lifted while a second person disconnects the temperature sensor and heater cables. The sample holder stage can then be removed from the sample space tube by twisting the top and carefully lifting it up. The sample space tube can be placed somewhere safe and the sample holder stage can be placed in the probe loading station, which was made for the Bluefors 2017 setup. Unlike the probe for the Bluefors 2017, the probe for the Bluefors 2023 does not fit properly in the probe loading station due to the design for 2" optics. However, it can still be placed here to exchange samples, but it cannot be screwed in place. In the future, it may be good to make a separate probe loading station.

The probe loading procedure is mostly the reverse of the unloading procedure. However, just as in the Bluefors 2017 procedure, we must make sure the probe is aligned properly by using the probe alignment tool supplied by Bluefors. We must also clean each flange with isopropanol and coat each flange with graphite as a lubricant. If too much graphite is applied, then a second lighter cleaning with isopropanol should be performed. The graphite should not be very visible. Also, just as before, the probe loading pin must be pushed into place. In the Bluefors

2017, to load the probe into the fridge, the FSE must first be pumped out before executing the probe-loading program. In the Bluefors 2023 the probe loading program performs the pumping of the FSE, simplifying the loading procedure. We have found that it is best to use the cooldown without condensing program to load the probe into the fridge; this is done with the script “BLFSE_SO01541.10_4K_cooldown_NO_condensing_2_3_pumping_VC_temperatures_variable.txt.” This script has been modified after the probe was repaired to fix the home in position at 5.3 mm. To start condensing either use “2_condensing_v2_51.txt” or “2_PPC+condensing_v2_51.txt.” Both will start the condensing process and bring the fridge to base temperature, but the latter will first circulate the mixture without condensing.

We had some difficulty reaching lower temperatures in this fridge. In our first attempt to condense and reach base temperature after installing the windows and internal optics, the probe only reached 450 mK. We then tried removing all of the custom probe components so that it was just the rails. With this, the probe reached 81 mK. We were able to reach 79.7 mK by increasing the still heater power to 25 mW. Bluefors support recommended that we let the system thermalize at 4 K longer after the probe is loaded, hence why it is best to load the probe using the no condensing script. We tried loading the probe with all custom components loaded back into the probe and allowed it to sit at 3.7 K for approximately six hours. After running the condensing procedure, the probe reached 100 mK. By setting the still heater to 25 mW we were able to reach 97.8 mK. This is warmer than the Bluefors 2017 base temperature, but so far we have not found a way to reach colder temperatures.

Another difference is the Attocube stack. This is shown in Fig. C.4. The larger space between the rails allowed us to place the heater and temperature sensor closer to the sample. We also decided to place Attocube x and y scanners in the stack. These should only be used when

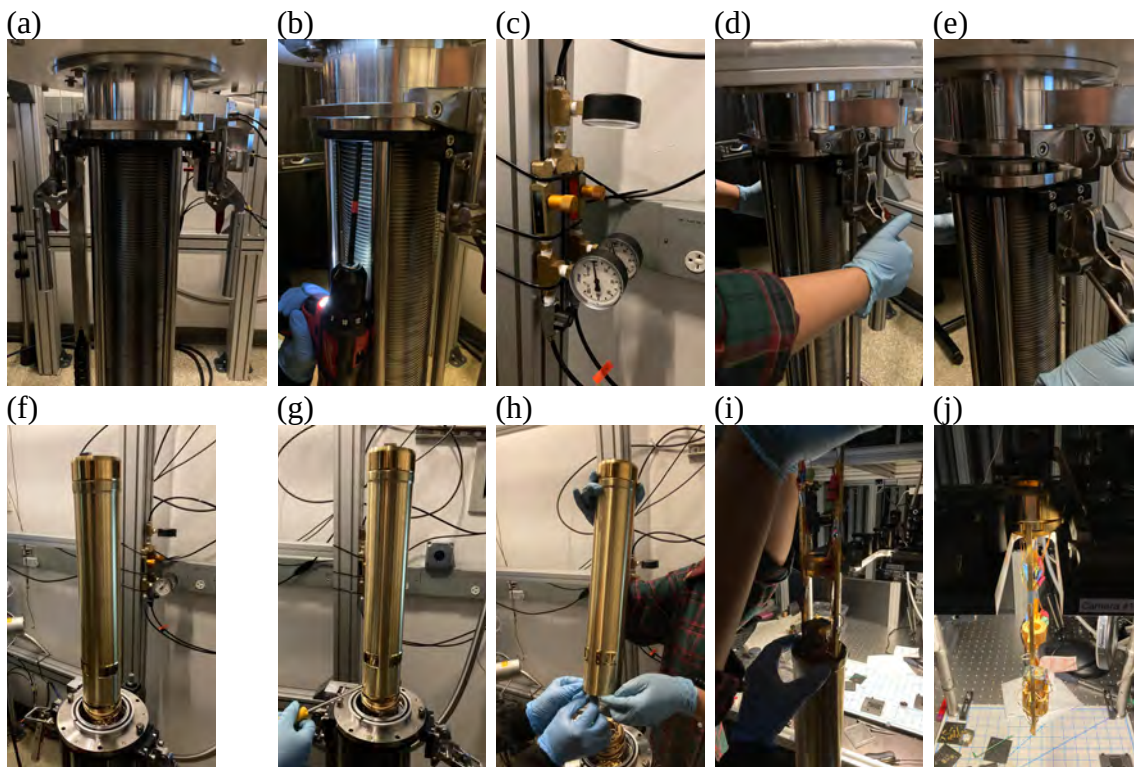


Figure C.3: Bluefors 2023 probe unloading. (a) FSE attached to the gate valve. (b) Removing screws that attach the FSE to the gate valve. (c) Needle valves used to inject nitrogen or air into the system. (d) Disengaging the latch clamps. (e) Lowering the FSE to the floor. (f) Moving the probe to the home in position. (g) Remove screws connecting which connect the sample space tube to the loading mechanism. (h) Lift the sample space tube and disconnect the temperature sensor and heater cables. (i) Remove the sample holder stage from the tube. (j) Place the sample holder stage on the probe loading station.

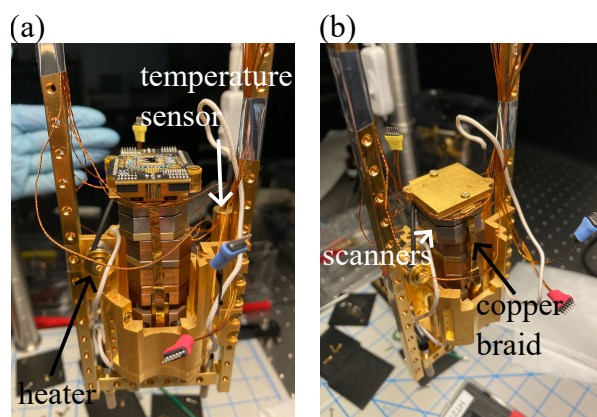


Figure C.4: Attocube stack. (a) The Attocube stack with a sample attached. The heater and temperature sensor are identified. (b) The Attocube stack without a sample. The Attocube thermal coupling device is identified with its copper braid. The scanners are also labeled.

the fridge is operating at 4 K. Because they operate using a DC voltage, they generate heat which would warm the sample if the fridge were condensing. The scanners are titanium, which is a poor thermal conductor, and thus we added an Attocube thermal coupling device. This is a thermal contact between the bottom of the stack and the top. In Fig. C.4 the copper braid belongs to the thermal coupling device. In order to have the sample the correct working distance from the objective, we needed to add a spacer. For the spacer, we used the gold plated copper thermal contact used for our sample carriages. With the added spacer, we needed to use longer brass M2 screws than in our other setups. The only commercially available M2 brass screws that were long enough are slotted flat head screws. Thus, when loading a sample, a flat headed screw driver must be used instead of a hex key.

DC lines	Fischer connector pin	Micro-D25 connector pin
1	1	1
2	2	14
3	3	2
4	4	15
5	5	3
6	6	16
7	7	4
8	8	17
9	9	5
10	10	18
11	11	6
12	12	19
13	13	7
14	14	20
15	15	8
16	16	21
17	17	9
18	18	22
19	19	10
20	20	23
21	21	11
22	22	24
23	23	12
24	24	25

Table C.1: Pin out for the DC lines.

X-positioner	Fischer connector pin	Micro-D25 connector pin
V+	1	1
V-	2	14
S+	3	2
S0	4	15
S-	5	3

Table C.2: Pin out for the X-positioner.

Y-positioner	Fischer connector pin	Micro-D25 connector pin
V+	6	16
V-	7	4
S+	8	17
S0	9	5
S-	10	18

Table C.3: Pin out for the Y-positioner. Note that the connection to S+ is not working. One of the solder joints must have broken. Since we are not measuring position we have chosen to keep it this way for now rather than making a new set of connectors.

Z-positioner	Fischer connector pin	Micro-D25 connector pin
V+	11	6
V-	12	19
S+	13	7
S0	14	20
S-	15	8

Table C.4: Pin out for the Z-positioner.

X-scanner	Fischer connector pin	Micro-D25 connector pin
V+	17	9
V-	18	22

Table C.5: Pin out for the X-scanner.

Y-scanner	Fischer connector pin	Micro-D25 connector pin
V+	21	11
V-	22	24

Table C.6: Pin out for the X-scanner.

Appendix D: Utilizing shot noise measurements to detect electron pairing in 2D semiconductors

In a recent theoretical work by Dehghani *et al.*, a mechanism for light-induced superconductivity using Floquet engineering is proposed in two-valley semiconductors with a massive Dirac-type band structure. This mechanism occurs by exciting photocarriers in one of the two valleys, leading to an asymmetric occupation distribution. This non-equilibrium occupation creates an effective pairing population inversion around one of the valleys, which leads to an inter-band pairing of electrons. The electron pairs would only have a lifetime of several picoseconds, making it difficult to detect superconductivity through traditional measurements of resistivity. The shot noise from an electronic system can be used to detect the charge of carriers. Hence, we propose a way to experimentally detect the formation of electron pairs due to the attractive potential between electrons in the $\mathbf{K}$ and $-\mathbf{K}$ valleys in a transition metal dichalcogenide monolayer under a Floquet drive via shot noise measurements in a mesoscopic tunnel junction.

Light-induced superconductivity in semiconductors has been theoretically studied since the 1970s [85–90]. It has received renewed attention due to recent theoretical and experimental advances in Floquet engineering of quantum matter, such as the light-induced anomalous Hall effect in graphene and light-enhanced superconductivity in organic superconductors by using a periodic drive [91–93]. However, light-induced superconductivity has not been shown in a

semiconductor.

In Ref. [89, 90] it is theoretically shown that by applying a periodic drive to a two-valley semiconductor with a massive Dirac-type band structure, such as a monolayer transition metal dichalcogenide (TMD), electron pairing can be induced. This is a non-equilibrium process where the electron pairs would have a short lifetime on the order of picoseconds. Instead of looking for the resistance to drop to zero, it would be better to be able to tell if electron pairs are being formed. Measurement of shot noise through a tunnel junction allows for measurement of the charge of carriers.

Shot noise in electronic systems is a current noise which results from the discreteness of charge. Because of this shot noise is proportional to the charge of the quasiparticles and the average current. Shot noise has been used to observe fractional charges in fractional quantum Hall systems and doubled charges in superconducting systems [94–98]. These measurements require the isolation, amplification, and detection of small currents. In the case of fractional charges, quantum point contacts have been used, which are constrictions in the plane of a two-dimensional electron gas (2DEG) that partly reflect the current [94, 95]. To detect electron pairs (Cooper pairs) in superconducting systems, tunnel junctions have been implemented either in the form of mesoscopic tunnel junctions [97] or tunnel junctions formed by a scanning tunneling microscope (STM) [96, 98]. Both of these methods require the use of low temperature amplifiers that function at high frequencies (~ 1 MHz).

Due to the equipment limitations of not having a scanning tunneling microscope and needing optical access to the sample, it is best that we start working with mesoscopic tunnel junctions which can be measured in cryostats equipped with RF electrical lines. This has worked before in detecting electron pairs copper oxide mesoscopic tunnel junctions [97]. However, work by Niu

et al., suggests that detecting electron pairing with this measurement can be difficult due to the transparency of the junctions and the tunneling of quasiparticles with $1e$ charge [99]. Using the lessons learned in this paper we can try and fabricate a sample with a smaller junction area and lower transparency which should give better results.

One limitation to the shot noise measurement is that it is averaged over time and the electron pairs that we are trying to detect have a short lifetime. This measurement could still work since in TMDs the free electrons would quickly bind to holes and form charge-neutral excitons. Since the shot noise is proportional to the charge of the quasiparticle, the charge neutral excitons should not contribute to the measured shot noise.

D.1 Light-induced electron pairing via Floquet interaction engineering

The mechanism for light-induced electron pairing presented in Ref. [90] is based on Floquet engineering, which is the control of quantum systems using a periodic drive. The periodic drive can change the band structure of the material, leading to new properties. The electron pairing process is shown in Fig. D.1. The two-valleys with a massive Dirac-type band structure are shown in Fig. D.1a. Circularly polarized light is used to selectively excite carriers in one of the two valleys. By changing to the rotating frame (Fig. D.1b) we can see that anticrossings form in the valley that is being driven by the circularly polarized light. The population probabilities at the crossing points in the $-\mathbf{K}$ valley and the anticrossing points in the $\mathbf{K}$ valley are asymmetric. This is a nonequilibrium process. To see how this results in interband electron pairing, we can

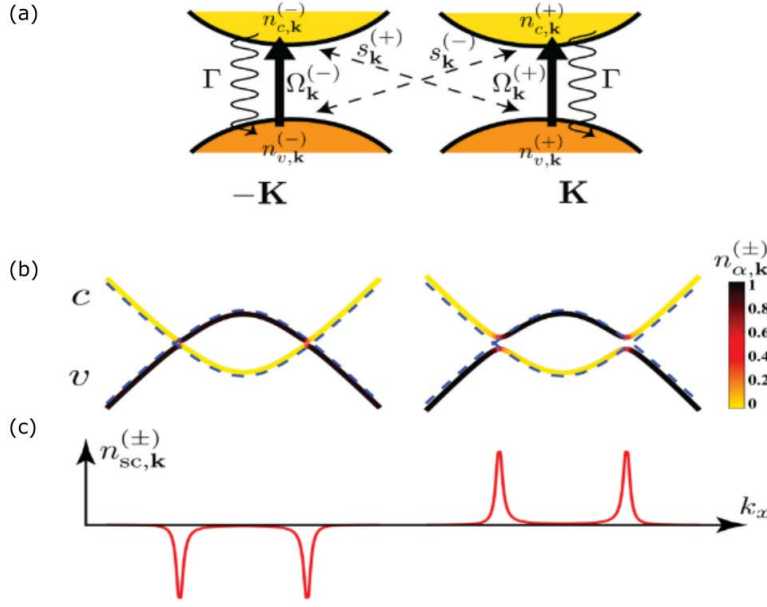


Figure D.1: Adapted from Ref. [90]. (a) The relevant processes in a 2D semiconductor around the two $\mathbf{K}$ points. The Rabi frequency and system-reservoir couplings are labeled Ω_k^+ (Ω_k^-) and Γ , respectively. $n_{v,k}^\pm$ ($n_{c,k}^\pm$) and $s_k^{(+)}$ ($s_k^{(-)}$) represent the occupation probabilities of the valence (conduction) band and the anomalous interband pairing between the conduction band around $\mathbf{K}$ ($-\mathbf{K}$) and the valence band around valley $-\mathbf{K}$ ($\mathbf{K}$), respectively. (b) The driven (static) energy spectrum in the rotating frame is plotted with a solid (dashed) line colored according to their population probabilities. (c) The interband pairing population, $n_{sc,k}^\pm = 1 - n_{v,k}^\pm - n_{c,-k}^\mp$, in terms of momentum.

look at the equation for the pairing order parameter:

$$\Delta_{\mathbf{k}}^\eta = - \sum_{\mathbf{k}'} \bar{U}_{\mathbf{k}\mathbf{k}'} \frac{\epsilon_{t,\mathbf{k}}}{\epsilon_{t,\mathbf{k}}^2 + \Gamma_t^2} n_{sc,\mathbf{k}'}^\eta \Delta_{\mathbf{k}'}^\eta, \quad (\text{D.1})$$

where $\bar{U}_{\mathbf{k}\mathbf{k}'}$ is the density-density interaction, $\epsilon_{t,\mathbf{k}}$ is the total kinetic energy at momentum $\mathbf{k}$, Γ_t is the total decay rate, and the interband pairing population is $n_{sc,\mathbf{k}}^\eta = 1 - n_{v,\mathbf{k}}^\eta - n_{c,-\mathbf{k}}^{-\eta}$. From Fig. D.1c, this value is negative around the $-\mathbf{K}$ valley and positive around the $\mathbf{K}$ valley. Equation (D.1) implies that states with negative $n_{sc,\mathbf{k}}$ can form pairing as the electron-electron interaction ($\bar{U}_{\mathbf{k}\mathbf{k}'}$) effectively changes sign.

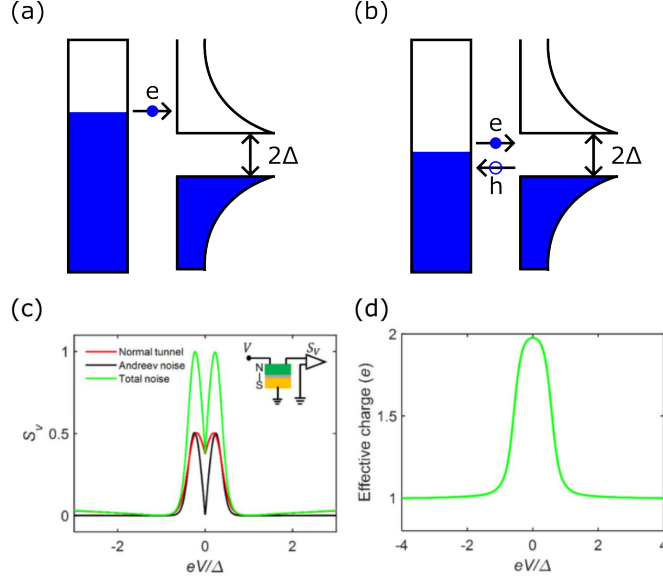


Figure D.2: (a)-(b) The density of states of the superconductor and normal metal in a NIS junction with filled states colored blue. (a) Single electron tunneling at biases larger than the gap. (b) Andreev reflection process when the bias voltage is smaller than the gap leading to $2e$ shot noise. (c) Voltage noise power calculated at different bias voltages according to BTK formulas of NIS junction with superconducting gap $\Delta = 3.8$ meV, $T = 4$ K, and transparency $\tau = 0.01$. The red line is the noise curve if there is only normal tunneling, the black line is the shot noise from Andreev reflection processes, and the green line is the total noise which is what is measured in experiments. The inset is the schematic of the electrical circuit needed to measure the noise of an NIS junction. (e) The effective charge calculated from the total noise. Adapted from Ref. [99].

On this basis, to maximize the electron pairing effect a material platform should be selected which has high valley selectivity and high Coulomb interaction. A good choice for this is monolayer TMDs, which have two-valleys with a massive Dirac-type band structure. The two valleys are related by time-reversal symmetry and thus have opposite Berry curvature. This leads to circular dichroism, meaning that the valleys are selectively excited by circularly polarized light with opposite handedness, i.e., the $\mathbf{K}$ valley is excited by σ^+ light and the $-\mathbf{K}$ valley is selected by σ^- light [100].

D.2 Shot Noise Theory

Before collecting any noise data we need to know what to expect. In this section we present some of the theory of shot noise in a normal metal-insulator-superconductor (NIS) junction. Tunneling in a NIS junction is depicted in Fig. D.2. If the Fermi energy of the normal metal is above the superconducting gap (2Δ) then there will be normal tunneling of $1e$ charges. If the Fermi energy of the normal metal is within the gap, then there will be an Andreev reflection leading to a $2e$ shot noise.

The $I - V$ curve and the spectral density of the noise power in an NIS junction can be calculated using the Blonder-Tinkham-Klapwijk (BTK) formulas [99, 101]:

$$I_{NS} = 2N(0)e v_F \mathcal{A} \int_{-\infty}^{\infty} [f_0(E - eV) - f_0(E)] [1 + A(E) - B(E)] dE, \quad (\text{D.2})$$

where $N(0)$ is the one-spin density of states at the Fermi energy E_F , E is the energy minus the Fermi energy, e is the electron charge, v_F is the Fermi velocity, $\mathcal{A}$ is the effective neck cross-sectional area, $f_0(E)$ is the Fermi-Dirac distribution function, $A(E)$ is the coefficient of Andreev reflection, and $B(E)$ is the probability of ordinary reflection. $A(E)$ and $B(E)$ are given by,

$$A(E) = \begin{cases} \frac{\Delta^2}{E^2 + (\Delta^2 - E^2)(1 + 2Z^2)^2} & \text{if } E < \Delta \\ \frac{u_0^2 v_0^2}{\gamma^2} & \text{if } E > \Delta \end{cases} \quad (\text{D.3})$$

and

$$B(E) = \begin{cases} 1 - A & \text{if } E < \Delta \\ \frac{(u_0^2 - v_0^2)Z^2(1+Z^2)}{\gamma^2} & \text{if } E > \Delta \end{cases}. \quad (\text{D.4})$$

In the above equations for $A(E)$ and $B(E)$, $\gamma^2 = [u_0^2 + Z^2(u_0^2 - v_0^2)]^2$, $u_0^2 = 1 - v_0^2 = \frac{1}{2}(1 + [(E^2 - \Delta^2)/E^2]^{1/2})$, and $Z = \sqrt{1/\tau - 1}$ is the barrier strength where τ is the transparency of the tunnel junction. The shot noise power spectral density function (PSD) can be calculated by [102, 103]:

$$S_I = 4N(0)e^2v_F\mathcal{A} \int_{-\infty}^{\infty} [f_0(E - eV) - f_0(E)] [A(E)[1+A(E)] + B(E)[1-B(E)] + 2A(E)B(E)] dE. \quad (\text{D.5})$$

If there is only normal tunneling ($1e$ shot noise), than $A(E) = 0$ and the expression for S_I is simplified to

$$S_{I,1e} = \frac{4e^2}{h} \int_{-\infty}^{\infty} [f_0(E - eV) - f_0(E)] [B(E)[1 - B(E)]] dE. \quad (\text{D.6})$$

If there is Andreev reflection, then there is an added contribution to the shot noise:

$$S_{I,Andreev} = \frac{4e^2}{h} \int_{-\infty}^{\infty} [f_0(E - eV) - f_0(E)] [B(E)[1 - B(E)] + 2A(E)B(E)] dE. \quad (\text{D.7})$$

More practically, at finite temperature it can be shown that the PSD of noise in a tunnel barrier can be written as

$$S_I = 2q|I| \coth\left(\frac{q|V|}{2k_B T}\right). \quad (\text{D.8})$$

This can be converted to a voltage noise if we know the differential resistance of the tunnel

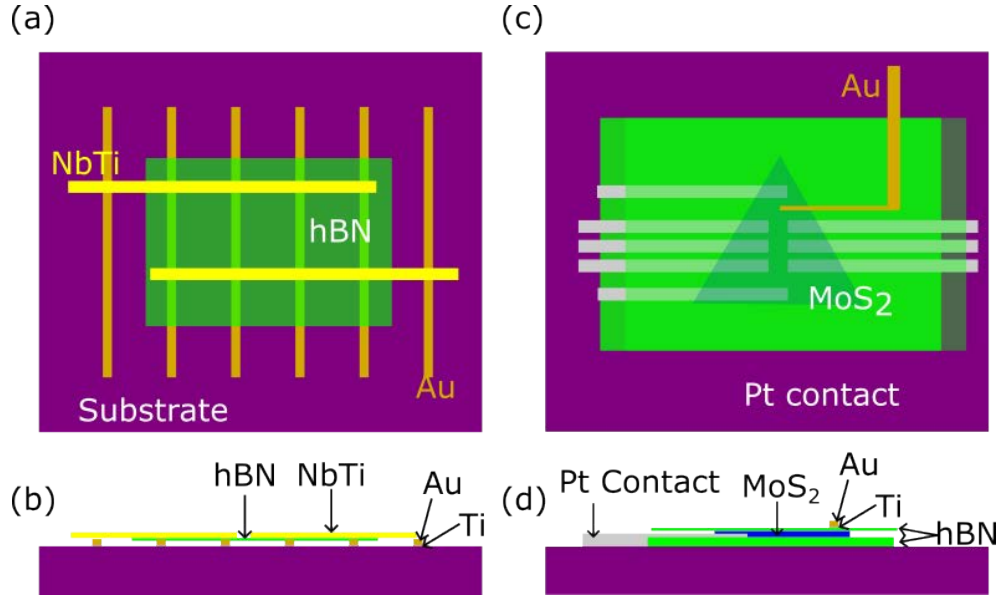


Figure D.3: (a) Top-down view and (b) side view of the designed test NIS junction. The normal metal is gold with a titanium adhesion layer, the insulating layer is a thin layer of hexagonal boron nitride, and the superconductor is niobium titanium. (c) Top-down view and (d) side view of the designed sample for light-induced pairing. This junction consists of molybdenum disulfide and gold separated by a thin layer of hexagonal boron nitride.

junction:

$$S_V = S_I R_{\text{diff}}^2, \quad (\text{D.9})$$

where R_{diff} is the differential resistance. This equation in combination with thermal noise is used to extract the carrier charge from the noise spectrum as a function of the voltage at the two ends of the junction (V).

D.3 Methods

D.3.1 Samples

Ultimately, the goal of this project is to detect light-induced electron pairing in a TMD. To do this, we need to design and fabricate a sample for this measurement. We propose the use

of monolayer MoS₂ because it has the best valley selectivity and Coulomb interaction strength among semiconducting TMDs [104]. This is important because the pairing mechanism for the electrons involves selectively exciting electrons in only one of the valleys and changing the sign of the Coulomb interaction. The sample design is depicted in Figs. D.3c-d. The MoS₂ will be encapsulated by hexagonal boron nitride (hBN), where the top layer will be only a few layers thin since it will be the tunnel barrier [105]. We will need at least two contacts to the TMD so that we can measure the resistance of the tunnel junction; for this, we can use pre-patterned platinum contacts on the bottom layer of hBN. A thin strip of gold with a titanium adhesion layer can be used as the normal metal part of the tunnel junction. If made thin enough, the gold will be optically transparent and the excitation beam will not be blocked. It is important to make sure the junction is small enough so that it is completely covered by the excitation beam. Since the spot size of the beam will be about 1 μm , this should not be difficult given the resolution of electron-beam lithography. We should be able to fabricate the gold (normal metal) part of the junction to have a width smaller than 500 nm.

Before we try to measure light-induced electron pairing, it would be better to test our system using a material that we know superconducts and thus should have $2e$ shot noise. To do this, we will fabricate a NIS tunnel junction consisting of gold as the normal metal, few-layer hBN as the insulator, and NbTi as the superconductor. NbTi has a critical temperature (T_c) of 9.2 K, so it should work well in our 4 K cryostat [106]. We will measure the noise spectrum of these NIS tunnel junctions as a function of DC bias voltage and temperature to see if we can detect enhanced shot noise within the superconducting gap.

D.3.2 Shot Noise Measurement

The sample will be mounted in a 4 K optical cryostat equipped with high-frequency coaxial cables and DC lines. Using the DC lines, the tunnel junction can be biased using a Yokogawa low-noise DC voltage source. The DC lines can also be used to measure the differential resistance of the tunnel junction for the full range of bias voltages. At each bias voltage, the current through the sample goes to a low-temperature amplifier discussed below where it is converted to a voltage and amplified. The signal is further amplified outside the cryostat at room temperature using a commercial voltage amplifier (FEMTO HAS-X-1-40). After the room temperature amplifier, a spectrum analyzer (Zurich MFLI) is used to measure the voltage signal. The spectrum analyzer can display the data as a PSD in the frequency domain. The Zurich MFLI can measure signals up to 5 MHz. We should look at part of the signal that is clean and below the RC roll-off, which can be seen where the spectral density decreases near 10 MHz in Fig. D.4d. In Ref. [99] the value of the noise used is the average over the frequency range of 1.1 to 1.5 MHz.

The voltage noise data can be taken as a function of bias voltage and temperature. Using the equation for shot noise at finite temperature (equation D.8), thermal noise, and the conversion to voltage noise (equation D.9) we can fit the data to

$$S_V = 2q|I| \coth\left(\frac{q|V|}{2k_B T}\right) R_{diff}^2 + 4k_B T R_{diff}, \quad (\text{D.10})$$

which gives the effective charge of the carriers.

According to ref. [90] the TMD should be driven by a beam whose frequency is slightly larger than the band gap. Depending on the dielectric environment, the band gap for MoS₂

has been measured to be in the range of 1.39 to 2.16 eV [107–110]. In units of wavelength, this is between 574 and 892 nm. This lower energy (longer wavelength) value of 1.39 eV was measured for a CVD-grown sample on a substrate of Au (111) [108]. Most of the other values were measured to be in the visible range. Apart from the wavelength of the light, we also need the electric field of the light to be high enough to see this effect. According to ref. [90] this should be greater than 5×10^6 V/m. This electric field strength can be accomplished using pulsed lasers. For example, the optical parametric oscillator (OPO) X has a wavelength range of 500 to 740 nm, a maximum average power of 520 mW, a pulse width of 80 fs or 3 ps, and a repetition rate of 80 MHz. The peak power for a pulsed laser can be calculated as the average power/(repetition rate times pulse duration). This gives a peak power of 81.25 kW. The intensity of the beam is given by the time average of the Poynting vector, $I = \frac{1}{2}c\epsilon_0 E^2$. For a spot size with a radius of 1 μm , a peak power of 81.25 kW would correspond to an electric field strength of 4.4×10^9 V/m or 7.2×10^8 V/m depending on the pulse width. Therefore, electric fields of magnitude 5×10^6 V/m should be within reach. To perform this measurement, we would just need to make sure that the average power is low enough to not burn the sample. This can be achieved through chopping the beam.

D.3.3 Amplifier

There are many types of noise that can influence tunneling measurements, including mechanical noise, 1/f noise, and thermal noise. Mechanical noise for a pulse tube cryostat can occur at frequencies up to 15 kHz, and the PSD function of 1/f noise decreases inversely with frequency (as the name implies) [111]. Thermal noise and shot noise are white noise, which means

that their PSD functions are independent of frequency. However, the PSD function of thermal noise is $S_V = 4k_B T R$, where k_B is the Boltzmann constant, T is the temperature, and R is the resistance of the sample. Therefore, as depicted in Fig. D.4a, if the measured signal is amplified at higher frequencies and the measurement is performed at lower temperatures, the shot noise can be isolated from other forms of noise.

This is accomplished by using an amplifier designed to work at cryogenic temperatures and measuring signals in the MHz regime [99, 112]. The circuit diagram and the amplifier image are given in Figs. D.4b-c. The amplifier is designed around a low-temperature high-electric-mobility transistor (HEMT), which is biased with a DC voltage. The tunneling current is fed into the gate of the HEMT and the signal is converted into a voltage noise across the HEMT and amplified.

D.4 Conclusion

In summary, we have proposed a method to detect light-induced electron pairing in a TMD monolayer by measuring shot noise while driving with a high-powered pulsed laser. Light-enhanced superconductivity has already been shown in organic superconductors by using a periodic drive [92, 93]. However, if this experiment is successful, it would be the first demonstration of light-induced electron pairing in a semiconductor. After verifying electron-pairing it would be interesting to test for signs of superconductivity using resistivity measurements and magnetic measurements to test for the Meissner effect.

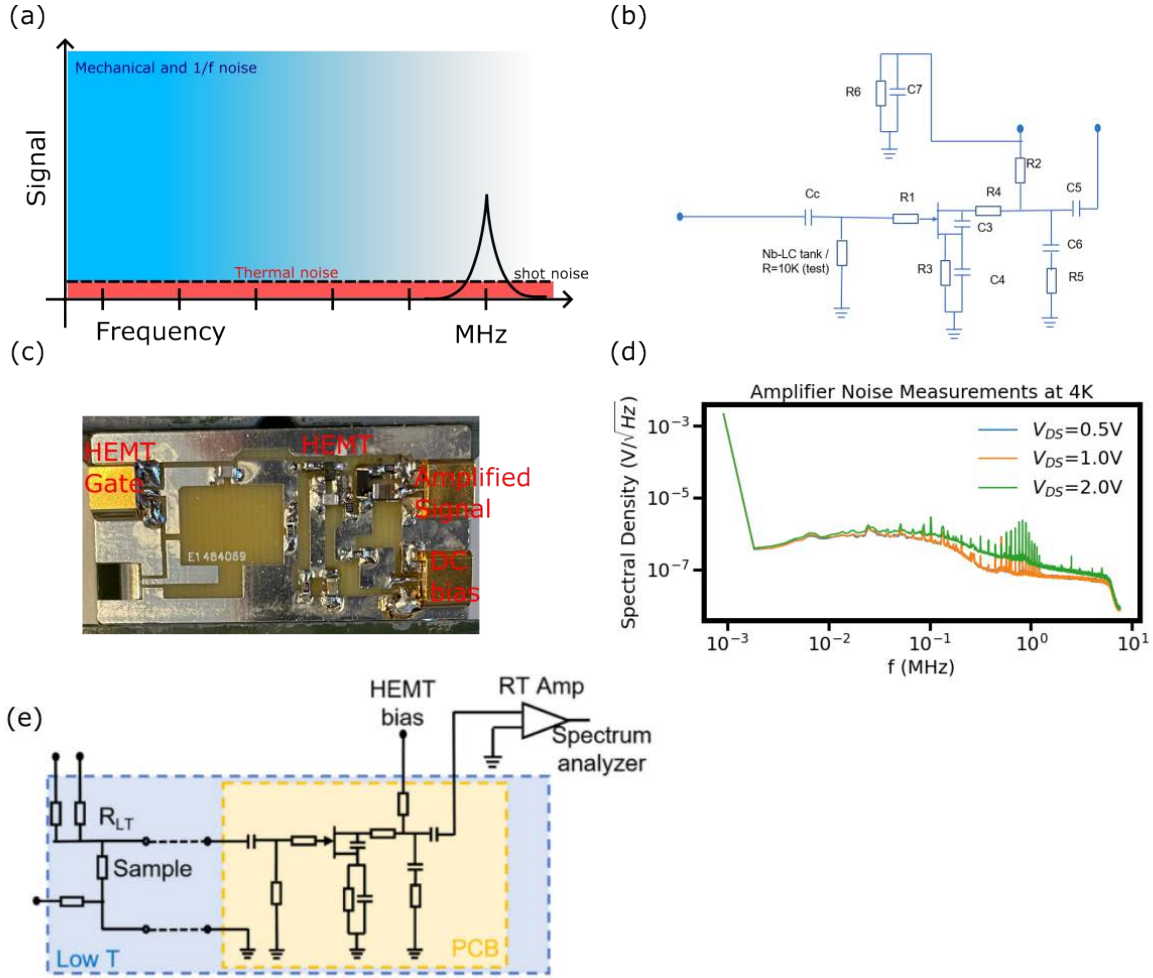


Figure D.4: (a) The different sources of noise in the tunneling measurement including mechanical, 1/f, thermal, and shot noise. This figure is adapted from Ref. [112]. (b) The circuit diagram for high frequency amplifier used for shot noise measurements. (c) Image of the amplifier made in Milan Allan's lab. (d) Noise spectrum of the amplifier measured at liquid helium temperature (4 K) in Milan Allan's lab. (e) Shot noise measurement schematic showing parts inside and outside the cryostat including the amplifier circuit (PCB) taken from Ref. [99].

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