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I, Bryan J Hemingway, hereby submit this original work as part of the requirements for the degree of Doctor of Philosophy in Physics.

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Magnetoconductance and Dynamic Phenomena in Single-Electron Transistors

A dissertation submitted to the
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in partial fulfillment of the
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Doctor of Philosophy

in the Department of Physics
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by

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Acknowledgments

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The last two people I wish to thank have been the most influential people when it comes to my understanding of experimental physics. The first, Dr. Tai-Min Liu, made all my experiments possible, or at least expedited them. His fabrication skills of the single-electron transistors are absolutely remarkable. They are extremely stable and they seem to take a beating very well. During the years that Tai-Min and I were graduate researchers together, he played a very important role in discussing quantum dot models and always had a way of bringing me back to reality when some of my ideas became a bit too unreasonable. His work ethic and motivation are an example of what all scientists should strive to become.

Finally, this brings me to my advisor, Dr. Andrei Kogan. When Andrei offered to let me work for him as an undergraduate, the randomness of bookwork that courses can seem like began to disappear and the direction of my academic career arrived. For me, Andrei was the ideal advisor. He gives slack when creativity is necessary and reels in when the fundamentals are being forgotten. The constant discussions have always yielded deeper understanding of physics and his openness to questions and comments have always aided me (even if he tells me I’m wrong).
Abstract

The quantum mechanical nature of electrons at very low temperatures has posed interesting results in the resistivity of metals over the last century. When the temperature nears absolute zero, the resistivity due to electron-electron interactions and electron-phonon interactions become very small and the dominant source of resistance is the scattering of electrons due to small concentrations of impurity atoms in bulk metal. When the impurity atoms are not magnetic, such that there is no net electron spin, the zero temperature resistivity due to impurity scattering is proportional to the impurity concentration. When the impurity is magnetic, typically spin-1/2, the resistivity rises logarithmically as the temperature is lowered below a characteristic temperature. This is known as the Kondo effect and the characteristic temperature is called the Kondo temperature. This temperature is expected to be the only energy scale that is involved in a Kondo system. This report investigates the universality of the Kondo energy scale. Interactions between impurity atoms and itinerant electrons can be reproduced in a single-electron transistor (SET). By isolating a small region of bound electrons, called a quantum dot, and coupling these to reservoirs of electrons, the biased tunneling current is a direct measurement of the interactions between the bound electrons and the reservoir electrons. SETs provide a unique measurement system with great variability to measure the nature of the Kondo effect.

Five experiments have been performed in this report. The first two experiments, performed in a regime where the quantum dot is weakly coupled to the reservoirs, demonstrate measurements of the properties of a quantum dot, such as the tunneling rate and coupling symmetry. The co-tunneling regime introduces spin dependent transport through a quantum dot without strong spin correlations related to the Kondo effect. As the tunneling rate is increased, spin correlations form and the Kondo effect emerges as a peak in the conductance at zero bias. These delicate correlations are broken when external energies are added to the system and a characteristic energy scale arises, known as the Kondo temperature. Measurements of this energy scale are made by increasing the electron temperature and applying a magnetic field. Universality of both temperature and the magnetic field are demonstrated with respect to the Kondo temperature.

The main focus of this work is the relationship between the Kondo temperature and the time spin correlations take to form. I show a distinct regime transition when the Kondo temperature is held constant and the frequency of bias oscillations are increased past a characteristic frequency related to the Kondo energy. The transition also is seen when the Kondo temperature is increased. Lastly, a unique conductance signature is observed when
an “fast” oscillation is applied and the magnetic field is increased. At small magnetic fields, well below the Kondo temperature, the Kondo conductance peak is enhanced. I find that the magnetic field value at which the peak occurs is linearly proportional to the frequency applied and only occurs at frequencies above the Kondo temperature.
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Chapter 1

Introduction

For two centuries, understanding and harnessing the power of electricity has led to remarkable innovations, such as a simple incandescent light bulb or the transistor, and provided a large sandbox for the development of microscopic theories that encompass all modern physics research. Such a simple macroscopic law, \( V=IR \), has left the simple question, ”why does a constant electric force produce a limited electron velocity?” Theories such as the Drude model and Boltzmann’s transport equation have solved this question from single electron modeling by applying scattering mechanisms. The simple linear behavior of the current with an applied voltage was soon put to question with the observation of superconductivity and other mesoscopic phenomenon. A typical weak conductor has zero resistance below a critical temperature. Again, the question arises: ”What causes resistance?” Electrical and thermal conductivity of materials, as well as magnetic susceptibility, rely on electron transport which makes full understanding a necessity when improving nearly any daily product.

Development of new materials such as high temperature superconductors or quantum computing systems require careful examination of the microscopic phenomenon which often produce non-Fermi liquid behavior in the electron system. The quantum mechanical nature of electrons in metals have led to interesting properties in complex systems at very low temperature relating to the hybridization of electron orbitals. The Kondo effect, described
at length in chapter 4 of this report, demonstrates a logarithmic increase in the resistivity of dilute magnetic alloys as the temperature is reduced below a characteristic temperature. Heavy fermion systems, such as CeAl$_3$, have anomalously large linear specific heat capacities. Both require electron correlations between electron orbitals in the corresponding atoms, d-orbitals for the Kondo system and f-orbitals for the heavy fermion systems[1]. Likewise, high temperature superconductivity and Mott insulators require the investigation of nearest neighbor electron correlations[2]. The advancement of electronic systems requires quality understanding of electron correlations to better engineer the material properties desired.

This report focuses on the electron correlations which arise due to the interactions with conduction electrons in a host metal and a small percentage of impurity atoms, described by the Anderson impurity model. A historical example of such a system is iron atoms (3d$^6$4s$^2$ electron configuration) embedded into gold (5d$^{10}$6s$^1$ electron configuration). In this case, the 6s electrons from the gold act as the conduction electrons and the 3d orbitals of the iron are the scattering impurity sites. Three regimes arise in systems such as the iron/gold matrix when the conduction electrons interact with the impurity orbitals based on the materials used: Coulomb blockade (chapter 2), coherent tunneling (chapter 3), and the Kondo regime (chapters 4-6). In bulk metals, the regime is defined by the inherent properties of the system. In the experiment by de Haas et al. [3] in 1934, the resistivity of a gold wire showed a resistance minimum as a function of temperature at 3.47 Kelvin. The increase in resistance as the temperature is lowered does not follow the traditional resistivity that contribute to Matthiesen’s principle, namely electron-phonon scattering and electron-electron scattering. Both of which produce standard power law temperature dependence. This anomalous behavior was later described as electronic spin-spin correlations formed by spin flipping interactions between the conduction electrons and the impurity electrons, known as the Kondo effect[4].

The sheer volume of both experimental and theoretical research performed on the Kondo effect would make any complete compilation nearly impossible and simply compiling only
the review articles and books would be quite a formidable task. Modern electronic device fabrication, described in chapters 1 and 2 of this work, promotes much research which allows experiments to no longer be bound by the fixed parameter space that bulk material establish. Nanoscale electronic devices, called single-electron transistors (SETs)[5], isolate a small region of bound electrons by creating an electric potential barrier greater than the Fermi energy. These isolated electrons act as the bound electrons on a single impurity site. The height of the potential barrier described the remaining conduction electrons’ interaction with the impurity. This permits an SET to tune the coupling factor and hence the intrinsic energy scale associated with the Kondo effect, known as the Kondo temperature. Coupling the bound electrons to two different electron reservoirs permits a measurement of the enhanced density of states that the hybridization creates by measuring the differential conductance, dI/dV, across the impurity. The two reservoir system also allows the impurity scattering to be forced out of equilibrium. The ability to tune the Kondo temperature, directly measure the density of states, and measure the out of equilibrium Kondo effect makes the SET favorable to bulk systems.

The last benefit that SET gives to research on the Kondo effect is the ability to directly apply excitations to the impurity model system. The spin interaction required for the Kondo effect responds to external magnetic fields. The Zeeman energy shift that electrons have in the presence of a magnetic field causes spin decoherence in the electrons and breaks the electronic correlations. The Kondo state re-establishes itself when electrons can scatter from a higher energy, the high biased reservoir, to a lower energy, the low biased reservoir. This produces an out of equilibrium Kondo state. Lastly, the voltages which are used to construct the SET can be time dependent. This time dependence creates a photon bath with which the system can interact. This allows the measurement of the dynamic density of states of the Kondo system. The dynamic behavior is the primary focus of this research as well as interacting effects when both time dependence and magnetic fields are applied.

The remaining portion of the introduction describes the development of reduced dimen-
sions in the quantum electron gases. The second chapter covers the concepts of electronic transport through an Anderson impurity. The bound energy levels in the "zero-dimensional" (0D) system permit conductance through the device when they are matched to the Fermi energy of the source and the drain. The energy requirement is met by controlling the chemical potential of these states by a nearby metallic electrode known as the gate electrode. Introductions to the Coulomb Blockade regime and the Kondo regime are described and measurements which describe the parameters of the models are presented. The third chapter introduces the role of spin and the virtual singlet state in the Anderson model. This coherent tunneling (co-tunneling) state permits a small but finite conductance within an odd occupancy Coulomb blockaded regime caused by scattering through the virtual singlet. The magnetic field dependence is described. The fourth chapter describes the decoherence of the Kondo singlet as the magnetic field is increased and presents a quantitative relationship between the Kondo temperature and compares the results to previous measurements by Tai-Min Liu. The fifth chapter introduces the Kondo timescale and conductance measurements made as a function of both excitation frequency and amplitude. A relationship between the frequency and the Kondo time scale is investigated by demonstrating the change in conductance behavior in two difference Kondo temperature settings. The sixth chapter describes the anomalous conductance increase seem as a small magnetic field is applied along with the microwave irradiation.

To conclude this dissertation, three appendices have been added. The first appendix describes the circuit used to manipulate the electrodes and the measurement of the differential conductance. The second appendix contains the procedure used to control the state of the device. Both the Coulomb blockade and the Kondo regimes require careful manipulation of the controllable voltages to have functioning states. The third appendix explains the microwave circuit and application of microwave signal to the SET.
1.1 Quantizing into a 2 Dimensional Electron Gas

In 1884, Sir Edwin Abbott introduced those not of the scientific and mathematical persuasion to the remarkable concept of a world not of three dimensions in his satirical novel, *Flatland: A Romance of Many Dimensions*. Unlike Abbott who describes mathematically pure dimensions, the following sections of this introduction explains the realization of dimensional reduction through the quantum nature of the conduction electrons. The ability to take a completely three dimensional semiconductor and reduce it to a simple “particle in a box” system gives rise to a convenient model system for testing a single Anderson impurity. To do this a three dimensional electron system has to contain a small region of effectively zero dimensional electrons.

Any introductory course in statistical mechanics describes the behavior of electronic gases. Throughout the course, it is imperative to instruct the nature of one-dimensional (1D) and two-dimensional (2D) electron gases as well as a three dimensional (3D) electron gas. Though how often as an undergraduate or a beginning graduate student, does one question the physical reality of 1D and 2D electron gases when clearly the material they occupy is easily measured to be finite in all three dimensions? This introduces the interesting interplay between fermion statistics and the quantization conditions of a particle in a box. The simple solution of the wave-vectors for the particle in a box yields that the dimension contribution is proportional to $1/L$, where $L$ is the physical size of that dimension. This requires asking the basic question of what happens when one dimension becomes significantly smaller than the other two. Similar to the 4s orbital in an atom crossing over the 3d orbital, the small dimension requires much more energy to contribute another wavenumber than the larger dimensions. As one reduces the dimension more and more, the two others exhibit continuum behavior while the quantum nature of the third is maintain. This produces sub-bands of reduced dimensionality. What began as a 3D continuum in the bulk now holds the property of an infinite sum of 2D continuum sheets. The role of fermion statistics becomes increasingly interesting. It is easy to imagine the case in which the number of electrons is simply not
Figure 1.1: (a) A schematic of the GaAs/AlGaAs heterostructure containing the 2DEG 85 nm below the surface. (b) The band diagram for a typical modulation doped GaAs/AlGaAs. Typical band bending occurs as the GaAs/AlGaAs interface and provides a band minimum which extends below the Fermi energy and binds the 2DEG.[6]

enough to begin filling the higher sub-band. The originally perfect Fermi sphere no longer receives contribution in one dimension and becomes a 2D circle. It is in this fashion that a finite N dimensional system exhibits the behavior of an N-1 dimensional electron gas.

In the single-electron transistor used in these experiments, the physical dimension was not simply reduced as described above in the particle in a box scenario, which can be seen in systems such as graphene or carbon nanotubes. In this system, the technique of modulation doping in semiconductor heterostructure was employed. The following is a simplified description of the development of 2D electron gases (2DEG) which can be read in full by the various publications from our collaborator Dr. Michael Melloch at Purdue University[6].

To produce a two dimensional electron gas in a semiconductor heterostructure, the goal is to produce a small region in one dimension in which the conduction energy is lower than Fermi energy. This can be done by exploiting methods which alter the structure of the conductance band and the Fermi energy, as seen in figure 1.1 a. The natural difference in
band energy between GaAs and Al\textsubscript{x}Ga\textsubscript{1−x}As causes band bending at the interface between the two materials which produces a small region of low conductance band energy. The addition of electron donor silicon atoms raises the Fermi energy in the heterostructure. These electrons, residing at the Fermi energy of the heterostructure, now are energetic enough to begin filling the bent conduction band in the AlGaAs/GaAs at the boundary, as seen in figure 1.1 b)[7]. The addition of an undoped layer of AlGaAs provides a spatial separation between the positive dopant ions and the 2DEG. Being adequately separated, the electrons in the 2DEG have a very small scattering cross-section with the ions which aids in producing very high electron mobilities. The conduction band energy diagram describes the potential well for the electrons that make up the 2DEG conduction electrons. Through the process of Molecular Beam Epitaxy (MBE) used by Dr. Melloch, the layers can be carefully grown and doped to construct an appropriate potential well in which the quantization condition of a particle in a box is met in the z-direction and a 2DEG is formed.

The properties of our 2DEG was measured by the quantum Hall effect (QHE)[8]. A small rectangular region of 2DEG, known as a Hall mesa, is produced in the heterostructure by etching into the heterostructure, past the 2DEG, in the areas surrounding the desired rectangle. This region contains small areas at four points on the mesa to measure the longitudinal resistance and the perpendicular resistance. Applying a perpendicular magnetic field to the 2DEG, the QHE experiment produces resistance oscillations in the direction of the applied voltage as the magnetic field is increased, known as Shubnikov-deHaas (SdH) oscillations[9]. The resistance perpendicular to the applied voltage, produces resistance steps as the field is increased. This is the integer quantum Hall effect (IQHE). These measurements extract the important parameters for our applications: the sheet density \( n_{2d}=4.8 \times 10^{11} \text{ cm}^{-2} \), the electron mobility \( \mu \geq 5 \times 10^5 \text{ cm}^2/\text{V sec} \). Importantly, the mean free path, 5.76 \( \mu \text{m} \), is measured to be much longer than the lithographic designs of the SET.

The electron density can be calculated by observing the magnetic field necessary to
complete a full SdH oscillation,

\[ n_{2DEG} = \frac{2e}{\hbar} \frac{1}{1/B_m - 1/B_{m-1}} \]  

(1.1)

where \( B_m \) is the magnetic field where the longitudinal resistance drops to a local minimum. The electron mobility can be calculated by knowing the square resistance, \( R_{\text{square}} = R_{\text{total}} W/L \), for the mesa width, \( W \), and length, \( L \).

\[ \mu_e = \frac{1}{e n_{2DEG} R_{\text{square}}} \]  

(1.2)

Lastly, the mean free path is calculated by both the Fermi velocity, \( v_F \), and the electron mobility.

\[ l_{\text{mfp}} = \frac{v_F m^* \mu_e}{e} \]  

(1.3)

1.2 Lithographic Electrode Design to Create Electron Depletion

This section describes the fabrication design used in the experiments of this report which was performed by Dr. Tai-Min Liu and described in full within his dissertation[8]. Previously, I described the quantization into two dimensions via modulated doping techniques and the desire is to create a model system for a single atom. The first requirement for measuring the transport across the 2DEG is to supply a source of electrons and a drain for the transported electrons. To improve the measurement capabilities, the interface of the source and drain with the 2DEG should exhibit ohmic behavior. In these devices, a layered system of nickel, germanium, and gold are annealed into the heterostructure to contact the 2DEG. The top gold layer provides a quality material to contact external wires in order to supply the voltage. The source and drain contacts at equilibrium assist in defining the relative chemical potentials of the 2DEG system.
Given a large sub-band energy requirement in the 2DEG, the electron density can be varied with the application of electrostatic forces. To generate these forces, an electrode is added to the surface of the heterostructure and connected to a potential. A characteristic of this electrode is to maintain a strong Schottky barrier with the GaAs surface. This guarantees that the electrode only provides a source of electrostatic potential and not particle exchange for the 2DEG. For a simple wide electrode, the electron density follows a parallel plate capacitor model and is linear with the barrier voltage,

$$\delta n_{2\text{DEG}} = \frac{\epsilon}{d} \delta V_b,$$

(1.4)

where $\epsilon$ is the dielectric constant (12.9 for GaAs) and $d$ is the distance of the 2DEG from the surface (85 nm for our 2DEG). This provides a natural cut-off voltage in which 2DEG electrons are completely depleted under the surface of the electrode. In a SET system, the electrodes are often dimensionally equal to the depth of the 2DEG and so this model has a simple, qualitative modification. We wish to be in a tunneling regime, so the density does not need to be zero, but only maintain statistically less than one electron. In other words, the potential barrier is higher than the most energetic electrons. By varying the applied gate voltage, the tunneling rate can be manipulated.

A single electrode design obeys the basic principle of a traditional transistor. The gate voltage reaches a cut-off value and the electron flow stops. To create a single-electron transistor, two tunneling electrodes are required. In the case where both electrodes are tuned near the cut-off potential, resonant tunneling is permitted to transport electrons through the device. Creative design of these electrodes further confine the region between the electrodes into a small, finite two dimensional region. As this region becomes isolated from the source and drain regions of the 2DEG, it behaves as an independent “island.” The addition of another electrode which causes minimal potential well deformation enables control over the chemical potential of the small region via capacitive influences and this electrode has the
Figure 1.2: (a) A SEM micrograph of our single-electron transistor. The four leads used to control the device are labeled $V_t$, $V_b$, $V_s$, and $V_g$. (b) A simplified diagram of the manipulation technique for our device. The source and the drain electrodes have been thermally annealed to allow metal to contact the 2DEG via percolation and these "ohmics" are exaggerated by the gold spears. The electrodes in (a) typically have a potential of -1000 mV applied to be in the tunneling regime.

role of a traditional transistor’s gate voltage.

To review our SET design shown in figure 1.2, we utilize four electrodes. Three electrodes form the primarily potential well. These are denoted the top, bottom, and side electrodes. The potential created by the top and side electrode voltages form one tunneling barrier and the bottom and side form the second barrier. The gate voltage creates a small potential deformation at the joining of the top and bottom electrode potentials. The notably small lithographic region between the electrodes provide a substantial energy level spacing on the isolated region. This can be easily seen as the energy necessary to transition from N to N+1 electrons on the island. For a circular approximation this energy is proportional to $1/r^2$ and statistically $\delta E \approx 500\mu eV$ for our lithographic area.
Chapter 2

An Overview of Transport through a Single-Electron Transistor

The purpose of the chapter is ultimately to describe the effects photon absorption and emission has on the transport properties in a SET. A simple picture is described by using single particle transport through the device. It covers an explanation of why spin degeneracy is relieved on the dot, a thermally activated free energy description of the conductance through the device, a complete picture of the lineshape of the conductance starting from the Breit-Wigner form for resonant tunneling, and the addition of photo-assisted tunneling to the tunneling rates within the master equations for a single level. The data presented shows the characteristic behaviors associated with the Coulomb blockade regime and the complexity of multi-orbital effects when they are populated by photo-assisted tunneling.

2.1 The Single Particle Picture

Before describing many-body effects in a single-electron transistor, it is inherent to point out the calibration techniques related to the single particle behavior of the Anderson impurity model[10, 5]. The simplest picture of the lithographic system used is a one dimensional double potential barrier. The two electronic reservoirs act as a set of incident electrons upon the
electronic potential. As with any electronic model, the addition of electrochemical potential across the material shifts the Fermi sea in the direction of the gradient. This produces transport behavior in that direction due to the electrons in the vicinity of the Fermi energy. In this sense, models consider these electrons explicitly. The two potential barriers created by the top, bottom, and side electrodes contribute to the bound wavefunction energies and most importantly coupling energy of electrons in the reservoir with the electrons in the bound region. This behavior holds the inherent properties of the Anderson impurity model.

A coupling between itinerant electrons and bound electrons. The SET offers two variables to the model which bulk systems cannot. The addition of the gate electrode permits a variable bound state energy and the transport set-up permits out of equilibrium measurements.

2.1.1 Single Electron Tunneling and Coulomb Blockade

The easiest depiction of tunneling current across a potential barrier is seen as a comparison of the two reservoir density of states. For a single tunnel junction the density of states can be described as

\[
I = e \Gamma \int_{-\infty}^{\infty} D_L f_L(E + V_{ds})(1 - f_R(E))dE - e \Gamma \int_{-\infty}^{\infty} D_R f_R(E)(1 - f_L(E + V_{ds}))dE \quad (2.1)
\]

where \(e\) is the electron charge, \(\Gamma\) is the tunneling rate, \(D_{L,R}\) is the density of states of the left reservoir and the right reservoir, \(f_{L,R}\) is the Fermi-Dirac function, and \(V_{ds}\) is the potential difference. This equation consists of two parts. The first term describes the probability that an electron is in the left reservoir and a vacancy is in the right reservoir. For equivalent density of states on the left and right the current is proportional to difference in the adjusted energies created by applying a source-drain voltage.

Describing the singe-electron transistor, two tunneling events need to occur. An electron needs to tunnel through both potential barriers while an intermediate bound state on the quantum dot is available. This sequential tunneling current requires an electronic state
residing on the quantum dot at the Fermi level of the reservoirs in equilibrium. When a bias voltage is applied and the two reservoir Fermi energies are different, an electronic state must reside between the two energies. This is known as the transport window and the differential conduction described the enhanced tunneling density of states.

For the SET, we need to have a description for the single electron energy levels on the dot. These levels will be used as the intermediate tunneling states which will contribute to the enhanced tunneling density of states. Using a classical approximation for the isolated region between the two tunnel barriers, we can describe the energy necessary to maintain a charge, $Q$, in the area by a geometrically defined capacitance, $C$, between the isolated region and the rest of space as $E=Q^2/2C$. This is the energy necessary to manipulate the charged particles into the arrangement. The energy associated with the addition of another electron to the region is then $e^2/2C$. The uniqueness of a SET is that designing an additional electrode provides additional energy to the localized electrons. This energy is dependent upon the electrical potential on the electrode with respect to the energy of the bound electrons, $V_g$. The full electro-static energy of the bound charges then is

$$E(Q) = -QV_g + \frac{Q^2}{2C} \quad (2.2)$$

The first term varies the total energy of the bound charge relative to the Fermi energy of the reservoir electrons while the second term is the configuration energy. This energy function is shown as continuous black curves in the lower panels of fig. 2.1. Due to charge quantization, $Q$ must be an integer number of the electron charge, $Q=Ne$. This quantization is shown as the red dots in the lower panels of fig. 2.1. We can determine the relationship between the gate voltage and the number of electrons in the isolated region by considering the gate voltage that minimizes the energy. Taking the derivative of the energy function with respect to the charge, $Q$, we see that the minimum energy is

$$V_g = \frac{Q}{C} \quad (2.3)$$
which is consistent with basic electrostatic results. Considering the voltage difference between the charge quantization occupations, \( N \) and \( N+1 \) (\( Q = Ne \) and \( [N+1]e \)), we find an energy spacing to be

\[
\Delta V_g = \frac{e^2}{C} \tag{2.4}
\]

The previous equation describes the situation in which the number of electrons is well defined on the isolated region, such that \( N \) electrons minimizes the energy for a particular value of the gate voltage. Figure 2.1 (a) portrays the Coulomb blockade energy gap and the minimization for an integer number of electrons. Considering transport, we are primarily interested when electrons can flow to and from the isolated region to the reservoir. This is realized for the value of the gate voltage when the \( N \) occupation energy is equal to the \( N+1 \) occupation energy. In this case, there is no energy required when the isolated region transitions between \( N \) and \( N+1 \) electrons. Equating equation 2.2 for \( E(Ne) \) and \( E(Ne\pm e) \),
the gate voltage necessary is found to be

\[ V_g = (N \pm \frac{1}{2}) \frac{e}{C}. \] (2.5)

The result is that the minimum energy in equation 2.2 falls directly between the two occupations as seen in fig. 2.1. Observing the difference between equation 2.3 and 2.5, an energy of \( e^2/2C \) is necessary to transition from \( N \) to \( N+1 \) and an energy of \(-e^2/2C\) to transition from \( N \) to \( N-1 \). In agreement with equation 2.4, the voltage spacing between transition energies is \( e/C \). The gate voltage controls the number of electrons on the region and can tune the system into a conductive device when the energy levels overlap.

The previous description describes an arbitrary isolated region of charge with some arbitrary number of electrons. Now I describe the filling of a real region connected to two reservoirs of electrons. A typical transport device described in the previous chapter contains two electron reservoirs maintained at the same zero temperature Fermi energy, \( E_F \). We can define the gate voltage to be zero when the first electron enters the region. Because the reservoirs are the only source of electrons for the region, we know that the energy on the region, given by equation 2.2, needs to be equal to the Fermi energy. For \( Q=e \), equation 2.2 gives

\[ E_F = -eV_g + \frac{e^2}{2C}. \] (2.6)

For a typical single electron transistor, the Fermi energy is usually of the order of 10 meV and the charging energy, \( e^2/2C \) is 1 meV. This implies that a negative gate voltage is typically required to deplete the region. A new parameter, \( \Delta V_g \) is defined as the variation of the gate voltage with respect to a gate voltage that depletes the charge on the region and requires a change equal to the Coulomb energy. In respect to equations 2.3 and 2.5, \( \Delta V_g \) replaces \( V_g \). \( \Delta V_g = e/2C \) when the first electron enters the region. As the gate voltage is increased slightly, the Coulomb blockade energy gap arises and \( N=1 \) describes the number of electrons on the region until the gap is compensated for by the gate voltage. In the center of the \( N=0 \)
Figure 2.2: (a) The conductance enhancement due to the $N=(0,1)$ overlap and the $N=(1,2)$ overlap at half integer values of $e/C$. (b) The electron occupation number as $\Delta V_g$ is varied.
“valley,” the energy is minimized by the occupation in the continuous picture described by equation 2.2. When $\Delta V_g$ is increased by the Coulomb gap energy, the mixture of $N=1$ and $N=2$ electronic states permits transport, followed by another blockaded valley.

### 2.1.2 The Quantum Orbital Energy Contribution

To this point, the discussion of the Coulomb Blockade energy gap has only assumed the electrostatic configuration energy. The quantum mechanical nature of electrons require every odd bound electron added to begin filling a higher energy orbital. In similar fashion as before, the isolated region, now quantized into a quantum dot, can be described by the energy necessary to add another electron. Now this energy is the energy gap between orbital levels, $\delta \epsilon$. Ignoring the charging Coulomb energy, the total energy of the quantum dot can be expressed similar to equation 2.2.

$$E = -NeV_g + \sum_i n_i \epsilon_i \delta \sum n_i, N$$

(2.7)

The first term describes the energy contributed from the gate potential and the second term describes the energy contributions from each orbital energy level, $\epsilon_i$. The electron occupation number, $n_i$, can be 0 or 2 corresponding to the spin degeneracy for an orbital level. Without the Coulomb energy, there is not an energy difference between adding a single electron or adding both electrons into the orbital. The delta-function takes into account the total number of particles for a particular occupation arrangement, the set $\{ n_i \}$. In the zero temperature limit, the quantum dot will fill the lowest energy levels first and continue until the energy on the dot is greater than the Fermi energy of the reservoirs. Like the previous section, $N$ corresponds to the lowest integer value in which the energy in equation 2.7 does not exceed the Fermi energy. The energy gap now is the orbital energy spacing and for each blockaded region, two electrons can tunnel on and off the dot.
2.1.3 Coulomb Blockade with Orbital Energy

Equations 2.2 and 2.7 can be combined to give a full description of single-electron transport through the quantum dot.

\[ E = -NeV_g + \frac{(N\epsilon)^2}{2C} + \sum_i n_i \epsilon_i \delta \sum n_i, N \]  

(2.8)

Following the methodology of the classical picture, we can now describe two different gate voltage increases. The first describes an odd occupied dot gaining the second electron in the orbital and the second describes an even occupied dot gaining an electron that fills the next orbital. If we define the Coulomb blockade charging energy, U, to be the classical separation, \( U = e^2/C \), the periodicity of the conductance oscillations become

\[ (N = odd) e \Delta V_g = U \]  

(2.9)

and

\[ (N = even) e \Delta V_g = U + \delta \epsilon_i. \]  

(2.10)

This allows us to describe Coulomb blockade transport in terms of a discrete energy level diagram. Shown in figure 2.3 (a) is the adjusted Coulomb Blockade conductance peaks. The odd valley has a gate voltage separation only equal to the charging energy and the even valley has a separation consisting of both the charging energy and the orbital energy spacing.

Figure 2.3 (b) pictures the nature of the full Coulomb Blockade energy gap and the energy levels below the highest activated level. To describe the orbital energies and wave functions, an infinite potential well is used as an example. The wave functions are easily pictured as sine functions with increasing frequency modes. The energy scaled used to define the dot energy is the additional gate voltage, \( \delta V_g \), necessary to populate the largest N. As seen in (a), the gate voltage difference between N=1 and N=2 is smaller than the difference between N=2 and N=3. This is directly shown with dashed lines in (b). Transitioning from N=1 (left plot)
Figure 2.3: (a) The modified conductance enhancement as a function of the gate voltage due to the filling of electron orbitals on the quantum dot. The odd valley has a gap of $U$ and the even valley has a gap $U + \delta \epsilon$ (b) For $N=\{1,2,3\}$ occupancies, the dot energy is characterized into wave functions for an infinite potential well. The dot energy is described by the gate voltage needed to activate the largest $N$ available.
to N=2 (center plot), the same orbital is filled, but the gate voltage needs to compensate for the charging energy. The transition from N=2 to N=3 (right plot) fills the next orbital level in accordance with the Pauli exclusion principle. The energy necessary to complete this transition is both the charging energy and the orbital energy. A key point to note in this diagram is that all the orbital levels feel the additional charging energy. Even though the charging energy gap exists when transitioning into a new orbital level, the separation of the bound state orbitals is only the orbital energy spacing, $\delta \epsilon$. This is pictorially shown by the step increases in the energy of the lowest bound orbital as more electrons are added. The bound orbital energy separation will play an important role when excited quantum dot states are considered in Section 2.3.

2.1.4 Free Energy Description of Transport

Describing a two reservoir system connected by the quantum dot in an uncorrelated regime can be described by writing an equation in which the probability of the dot is non-zero and the leads contain an available energy level. We define transport conductance through the dot as[11]

$$G = \frac{e^2}{k_B T} \sum_{p=1}^{\infty} \sum_{N=1}^{\infty} \frac{\Gamma_p \Gamma_r}{\Gamma_p + \Gamma_r} P_{ensemble}(N, n_p = 1)[1 - f(E_p - U(N) - U(N - 1) - E_F)] \quad (2.11)$$

This form contains the total statistical probability of all orbital levels for any occupation number of electrons. The probability of the p-th level being occupied in an N-occupancy dot is given by

$$P_{ensemble}(N, n_p = 1) = \sum_{n_1} P_{ensemble}(\{n_i\}) \delta_N \sum_i n_i \delta_{n_p, n_p}.$$  

(2.12)

$P_{ensemble}(n_i)$ is the grand canonical ensemble for a given set of electron arrangement in the dot levels. The ratio of tunneling rates comes from the solution of the kinetic equation in linear response for a two reservoir contact to the dot. The ensemble probability for a set of
occupancy numbers $n_i$ is given by

$$P_{\text{ensemble}}(\{n_i\}) = \frac{1}{Z} e^{-\beta(\sum_{i=1}^{\infty} E_i n_i + U(N) - N E_F)}$$

(2.13)

in which $Z$ is the partition function for all arrangements. It is noticed that in conjunction with the previous section, the conductance is periodic with respect to the charging energy and the gate voltage. Here the important feature is that the change in energy due to the gate voltage can be seen as applied to the Fermi energy of the reservoirs, or the free energy of the dot energy. The condition that $\phi_g = U$ depends solely on the relative energy change between the dot energy and Fermi energy such that we describe the energy shift as

$$\delta \epsilon_{\text{dot}} / e = \alpha_g \delta V_g$$

(2.14)

for fixed barrier potential and a capacitance ratio $\alpha_g = C_g / C_{\text{total}}$. The significant low-temperature result is that conductance obeys a lineshape with respect to the gate voltage around the peak conductance.

$$G/G_{\text{max}} \approx \cosh^{-2}(\frac{\delta \epsilon_{\text{dot}}}{2.5 k_B T})$$

(2.15)

The peak conductance, $G_{\text{max}}$, satisfies the transport condition that $e \alpha_g V_g = 0, U$.

2.1.5 Energy Scales in Coulomb Blockade

The introduction of the resonant condition relying on a large charging energy introduces the first energy scale in SETs. The charging energy, $U$, must be greater than the temperature of the electrons. The inverse hyperbolic cosine squared behavior describes the conductance for a single peak and maintains a thermal width of the Coulomb blockade conductance peak. As the temperature is increased, the width gradually increases until two neighboring charging features cannot be individually identified. For the Coulomb blockade experiments in this chapter, temperatures need to be approximately 1K to observe a 1meV charging
energy clearly. In order to experimentally reach such low temperatures, a Leiden Cryogenics dilution refrigerator is used in all the experiments in the dissertation. The sample is thermally anchored to the coldest portion of the refrigerator, the mixing chamber, by a large copper cold finger. The wires used to operate the SET are anchored to the mixing chamber by wrapping many loops around a copper rod in equilibrium with the mixing chamber temperature. This method produces a minimum mixing chamber temperature of 10mK and a base electron temperature, as the measured by inelastic co-tunneling thermometry, of 100mK.

2.1.6 Zero Temperature Resonant Tunneling

The previous section describes a standard conduction result for thermally broadened transport windows in the Coulomb Blockade regime. Quite often, thermal broadening is not the dominant mechanism for the width of the conductance peak and is extremely important in photo-assisted tunneling measurements. It is very convenient to start with the Breit-Wigner distribution that describes the non-interaction resonant tunneling through a SET[11].

\[
G_{BW}(E) = \frac{e^2}{\hbar} \frac{\Gamma_r \Gamma_l}{\Gamma_r + \Gamma_l} \frac{\Gamma}{(\epsilon_{dot} - E)^2 + \Gamma^2}
\]  

(2.16)

Again the symmetry factor reduces the maximum conductance possible and there is no factor of two for spin degeneracy. The width of the Lorentzian function, \( \Gamma = (\Gamma_l + \Gamma_r)/2 \), is the average of tunneling to the left lead and the right lead. This equation represents the finite lifetime of an electron in the dot near the Fermi energy of the reservoir. This zero temperature functional form for the conductance can be thermally broadened by assuming the Fermi function for the reservoir occupation.

\[
G(T, \Gamma) = \frac{1}{4k_BT} \int_{-\infty}^{\infty} G_{BW}(E) \cosh^{-2}\left(\frac{E}{2k_BT}\right) dE
\]

(2.17)

This equation leads to the calculation of the thermally broadened conductance behavior as the dot energy is varied.
2.2 Coulomb Diamonds

To fully understand the parameter space measured in a SET, a calibration needs to be performed to extract the capacitance ratios and the charging energy of the dot. These are significant quantities for characterizing the Kondo effect and so the following describes the method for investigating these parameters. Generalizing eqn 2.14,

\[ \delta \epsilon_{\text{dot}} / e = \sum_{i=\{g,s,t,b,s\}} \alpha_i \delta V_i \]  

where again the values for ‘i’ are the set of electrodes. For fixed barrier electrodes s,t,b, the only two contributing components are \( V_{ds} \) and \( V_g \). Using the thermally broadened Lorentzian form for the conductance of the SET, eqn 2.17, and the above definition for the variable dot energy, we can construct the complete Coulomb Blockade features in the source-drain and gate voltage subspace.

To explain the diamond features, we consider the arrangement in which the gate voltage is tuned to a starting dot energy which is at the Fermi energy at bias equilibrium. By applying a gate voltage, we tune the orbital energy away from the Fermi level and the resonance condition. Resonance can be again achieved by adjusting the source or drain chemical potential. This can be imagined in one of two scenarios: (1) bringing the source chemical potential to the energy of the dot or (2) using the capacitive influence of the source potential to bring the dot energy to the chemical potential of the drain. These processes can be described by

\[ \alpha_g \delta V_g + \alpha_{ds} \delta V_{ds} = V_{ds} \]  

and

\[ \alpha_g V_g = \alpha_{ds} V_{ds} \]  

These two equations describe increased conductance not only as periodic as the gate voltage overcomes the charging energy but as continuous as both the gate and source/drain voltage.
Figure 2.4: (a) An instructional Coulomb diamond with four markers representing (b)-(e). The horizontal lines represent the increased conductance. (b) A SET at equilibrium in resonance with a dot energy level. (c) The energy of the dot was increased by making the gate voltage more negative. (d) and (e) Two possible ways to bring the energy level into resonance with one of the reservoirs. (d) The source voltage is increased until it reaches the dot level. (e) The source voltage is used as a level arm to bring the dot level to the drain potential.

is varied together. This continuous behavior describes two slopes in $V_g$ vs $V_{ds}$ space, $(1-\alpha_{ds})/\alpha_g$ and $\alpha_{ds}/\alpha_g$. The capacitance ratio for the gate voltage is used to determine the charging energy of the device.

2.3 Introduction of Photons

2.3.1 A Master Equation Approach to Transport

The ensemble statistics provide good insight into the charging behavior of the quantum dot as well as the lineshape around the charging conductance. In the interest of this dissertation, a simplified picture of the conductance given previously will be presented for a single charging peak around $N$ to $N+1$ electrons. The utility of rate equations, often called the master equations, of the flow of electrons can be used to describe how the tunneling rates change the population of the dot. The probabilities between $N$ and $N+1$ electrons are directly linked
due to the tunneling on and off of the dot. An N+1 electron system can lose an electron to create an N system, and vice versa, as it is shown in the following equations[12].

\[ \dot{P}_N = P_{N+1}(\Gamma^\text{out}_L + \Gamma^\text{out}_R) - P_N(\Gamma^\text{in}_L + \Gamma^\text{in}_R). \]  
(2.21)

and

\[ \dot{P}_{N+1} = P_N(\Gamma^\text{in}_L + \Gamma^\text{in}_R) - P_{N+1}(\Gamma^\text{out}_L + \Gamma^\text{out}_R) \]  
(2.22)

These equations can be solved to give the probabilities with the boundary condition that the quantum dot is always in either the N or N+1 state,

\[ P_N + P_{N+1} = 1. \]  
(2.23)

The tunneling rate in and out of the device is specifically noted to account for the electron occupancy of the reservoirs. The N+1 to N transition requires an empty electronic state in the reservoir while the N to N+1 transition requires an electron to be present in the reservoir. This is described by the product of the tunneling rate and the electron occupancy of the reservoirs.

\[ \Gamma^\text{in}_{l,r} = \Gamma_{l,r}f(\epsilon_\text{dot} - e\alpha_g V_g \pm 1/2\alpha_{ds} V_{ds}; T) \]  
(2.24)

\[ \Gamma^\text{out}_{l,r} = \Gamma_{l,r}(1 - f(\epsilon_\text{dot} - e\alpha_g V_g \mp 1/2\alpha_{ds} V_{ds}; T)) \]  
(2.25)

where \( f(E; T) \) is the Fermi distribution function for the reservoir leads at temperature, T.

It is noted that the energy of the dot has two corrections instead of one. The source and drain potentials contribute a shift in the orbital energy as well as the gate voltage yielding a combined energy shift, \( \alpha_{ds} V_{ds} + \alpha_g V_g \). This is the integral component of the Coulomb Blockade diamonds described later. Also, for these equations, the bias potential is divided between the source and drain in which the right tunneling carries a negative voltage and the left tunneling carries a positive voltage.

To obtain the current passing through the device, we maintain balance between the left
and right leads. The current is difference between the process of going from N to N+1 and N+1 to N.

\[ I = e(P_N \Gamma^\text{in}_I - P_{N+1} \Gamma^\text{out}_I) \] (2.26)

The same process is described for the right tunneling, although the two are mathematically identical if the directional tunneling rates are equal. A complete picture which accounts for the asymmetry of the left and right tunnel barriers can be derived similarly and results in the above asymmetry factor in eqn 2.11. The quantity of interest experimentally is the differential conductance

\[ G = \frac{d\langle I \rangle}{dV_{ds}} \] (2.27)

because the current in any tunneling process can be described as the convolution of a series of density of state functions. For a single tunnel junction the derivative of the current describes the difference in density of states of the initial and final states. For a SET, the dot can be viewed as an enhanced density of state. In fig. 2.5, the diamond pattern is observed in our SET. Using the equations for the positive and negative slopes in the diamond, I have calculated the capacitance ratios and the corresponding charging energy associated with this particular quantum dot setting. The figure also shows the gate dependent lineshape for the higher charging peak in the diamond. To obtain the tunneling rate, \( \Gamma \) the lineshape can be fitted to a thermally broadened Breit-Wigner form, eqn 2.17. \( U \) and \( \Gamma \) are important parameters to extract in order to define the Kondo effect as it will be explained in the following chapter.

### 2.3.2 Photon-Assisted Tunneling

The absorption and emission of photons in a tunneling process was first implemented by Tien and Gordon[13] to describe tunneling through a superconducting junction within the superconducting gap. This process can also be used to describe conductance enhancements at Coulomb blockaded gate voltages. When an oscillating bias voltage is applied to the system,
Figure 2.5: (a) The conductance measurement for the SET tuned to the Coulomb blockade regime. The source-drain voltage is varied as well as the gate voltage. The characteristic diamond pattern is observed. The capacitance ratios and the charging energy are calculated. (b) The gate dependent linseshape is shown for a single charging region. the width of the peak can be expresses as $\Gamma_{FWHM} = 0.74\Gamma + 3.5k_B T$.

$V(t) = V_{ds} + V_{AC} \sin(2\pi f)$, photons of energy, $hf$, can interact with the tunneling process. Figure 2.6 (b) demonstrates this process. When the gate voltage is tuned past the Coulomb blockade charging peak value the occupancy of the dot is well defined. In the presence of a photon bath, the electrons can have photon mediated tunneling processes. An quantum dot can be ionized due to the absorption of a photon by an electron which then tunnels out of the dot. To conserve energy, an electron can then emit a photon and tunnel onto the dot. This process is not excitations in the reservoir which then match resonant levels on the dot, but a combined process in which the electrons interact with the photons during the tunneling process. This leads to a frequency dependent adjustment to the tunneling rate used in the master equations approach.[12]

$$\Gamma_{in,out}^{l,r}(V_g, V_{ds}; T) \Rightarrow \Gamma_{in,out}^{l,r}(V_g, V_{ds}, f; T)$$ (2.28)

The new corrected forms to be used in the tunneling rates are

$$\Gamma_{in}^{l,r} = \sum_{n=-\infty}^{\infty} J_n^2 \left( \frac{eV_{AC}}{nhf} \right) \Gamma_{l,r} f(\epsilon_{dot} - e\alpha_g V_g \pm 1/2\alpha_{ds} V_{ds} + nhf; T)$$ (2.29)
and
\[ \Gamma_{l,r}^{\text{out}} = \sum_{n=-\infty}^{\infty} J_n^2 \left( \frac{\epsilon V_{AC}}{n h f} \right) \Gamma_{l,r}(1 - f(\epsilon_{\text{dot}} - e \alpha V_g \mp 1/2 \alpha ds V_{ds} + n h f; T)), \]

(2.30)

where \( J_n(x) \) is the Bessel function and \( n \) are integers. The Bessel function arises due to the adjustment to the tunneling wavefunction in the presence of an oscillating barrier. The sum can be interpreted as processes that involve \( n \) number of photons in which negative \( n \) represent emission processes and positive \( n \) represent absorption processes. It is seen that each Bessel function peaks at a larger argument than the previous. This requires a greater oscillation amplitude to account for the interaction with more photons. This can also be viewed in the Breit-Wigner form as multiple resonances with peak shifts of \( \Delta V_g = n h f \) and amplitudes reduced by the corresponding squared Bessel term. In order to observe these side peaks in the energy, the natural width of the peak \( \Gamma_{FWHM} = 0.74 \Gamma + 3.5 k_B T \), should be less than the photon energy, \( h f \). It should be noted here that the ability to tune the tunneling rates in order to observe the photo-assisted side peaks can be very difficult. The total tunneling rate, \( \Gamma = \Gamma_L + \Gamma_R \), and the symmetry, \( \Gamma_L \Gamma_R / (\Gamma_L + \Gamma_R) \) need to be carefully changed to produce the small tunneling rate necessary. In this process the sensitivity to the asymmetry increases. This potentially means that a particular dot arrangement may never achieve the small \( \Gamma \) and the tunneling parameters will need to be rearranged. The state in fig. 2.5 has been changed to account for the reduced tunneling rate, though in general, the charging energy changes very little for these types of modifications.

In this experiment, we applied microwave frequency excitations to the device by means of a capacitive coupling to the bias via the source voltage, which is fully explained in the appendix. Select frequencies demonstrate good transmission to the device and the conductance shows strong dependence on the input power; while the conductance does not respond well to other frequencies. This is the selection procedure for the applied frequencies in all experiments and is described in full in the appendix. For the “good” frequencies, the amplitude is varied by means of a controllable logarithmic power, \( N \text{ dBm} = 10 \times \log(P/1 \text{mW}) \), which the power is proportional to the square of the voltage. The input power to the system
Figure 2.6: (a) Normal single level, single particle resonant tunneling. (b) Photo-assisted tunneling through a single level tuned below the Fermi energies. (c) Multi-orbital photon interaction activates tunneling through the blockaded region when the photon can mediate the energy difference.
is controllable in a 0.1 dBm scale, though the actual input power, in dBm, will contain a frequency dependent coupling term.

\[ 10 \times \log\left( \frac{P}{1mW} \right) + A(f) \]  

(2.31)

Unfortunately the voltage amplitudes applied to the SET cannot be compared from frequency to frequency. The relative power for different frequencies can be compared because we believe the coupling term is power independent. The power is increased and we observe the change in the Coulomb blockade charging peak corresponding to the addition of photons. The predicted behavior is that as the oscillating voltage is increased, the zero photon process should give way to a one photon process and a side peak for a photon absorption and a peak for emission should emerge.

Comparing the applied microwave data to the no microwave data, we observe a strong reduction in the conductance and the emergence of side peaks in the gate voltage. However, when we label the integer photon energy in terms of the gate voltage, we find that several peaks do not fall in line with the photon energy. A pattern emerges in which the peaks alternate between two changes in gate voltage. Particularly, every other peak aligns roughly with the photon energy level, as seen in fig. 2.7. The second set of peaks demonstrate a constant shift in gate voltage from the first set of peaks.

\section*{2.3.3 Multi-Orbital Effect}

The poorly defined geometry of the quantum well in a SET can lead to orbital energy levels that have small energy separations, well below the 500µeV average separation for a circular region. Typically, orbitals above the highest occupied energy level are of no consequence to the conductance in a SET because the dot resides in the ground state for very low temperatures. The presence of photons produce a very interesting tunneling possibility. Shown in figure 2.6 (c) is a process that involves the quantum dot existing in an excited state
of energy $\delta \epsilon$ above the ground state. When the excited dot energy equals the Fermi energy, as shown, the photon absorption and emission tunneling process can excite the quantum dot into the higher energy state. This process permits tunneling through the excited state and produces photo-assisted excited tunneling conductance. Theoretically this can be handled by the master equation approach.

The original handling of the charging region assumed a single state of the quantum dot. However, for a dot occupancy of $N$, we can describe all of electron configurations for all the bound wavefunctions\cite{14, 15}. Under the pretense that $U \gg \delta \epsilon$, the number of possibilities could be very large. The single configuration wavefunction is often correct when the temperature is very low because the higher energy configurations cannot be populated. This assumption does not include the possibility of an electron absorbing a photon as it tunnels into the higher energy configuration. To account for this process, we re-investigate the master equations. Suppose a dot configuration, $\chi_N$, with $N$ electrons. The coupling to these occupations will be accounted for by the tunneling rates. The full master equations can now be rewritten as as sum of the tunneling to the desired occupation and configuration\cite{12},

$$
\dot{P}_{N, \chi'} = \sum_{\chi} P_{N+1, \chi} \left( \Gamma_{out}^{in} + \Gamma_{out}^{in} \right) - P_{N, \chi'} \left( \Gamma_{in}^{out} + \Gamma_{in}^{out} \right),
$$

(2.32)

and

$$
\dot{P}_{N+1, \chi'} = \sum_{\chi} P_{N, \chi} \left( \Gamma_{in}^{in} + \Gamma_{in}^{in} \right) - P_{N+1, \chi'} \left( \Gamma_{out}^{out} + \Gamma_{out}^{out} \right),
$$

(2.33)

where the complete photon tunneling rates are the same form as the original but account for the added energy, $\epsilon_1$ necessary to tunnel into the excited state. These equations now explain that when a photon energy is greater than the energy difference between the ground state and an excited state, tunneling can occur through the excited state. Again, as the amplitude is increased, both the ground state and the excited state favor tunneling through the multiple photon process. This explains the two sets of peaks which have a shift of $\delta \epsilon$ between the two.
2.4 Conclusions

An explanation of the additional energy, charging energy $U$, necessary to add an electron to the quantum dot has been given which focuses on a capacitive model of electrodes. The alignment of electronic bound state energies has been demonstrated experimentally to show the functionality of a SET and gives motivation for the variability necessary to pursue the Kondo effect. A simplified master equation approach was given to explain the change in tunneling rates to account for the presence of photons which yield multiple peaks in the conductance around the central charging peak as the gate voltage is varied. My photo-assisted tunneling data presented shows the complexity of the quantum dot energy spectrum. The inclusion of excited quantum dot states gives rise to the unique peak structure of the Coulomb blockade charging region.
Chapter 3

Co-Tunneling Regime

This chapter further explains the controllability of a single-electron transistor. The focus of this experiment is to describe the coherent tunneling (co-tunneling) through the Coulomb blockade region of the SET. From a purely mechanism point of view, co-tunneling is very similar to the enhanced conductance due to the Kondo effect and fits perfectly well as a bridge between Coulomb blockade and the Kondo effect. To investigate the co-tunneling regime, we vary the tunneling rates through the device and then observe the ratio of the elastic and inelastic co-tunneling processes. These measurements have been made over three different dot occupation arrangements.

3.1 The Co-Tunneling Virtual State

To this point, only the orbital energy and the charging energy has been considered relative to the Fermi energies of the electron reservoirs and any higher order interaction with the quantum dot have been neglected. Within the framework of Coulomb blockade, the device Hamiltonian can be represented as

\[ H = H_{left} + H_{right} + H_{dot} + H_{transition} \]  \hspace{1cm} (3.1)
with
\[ H_{\text{left,right}} = \sum_{n=1}^{N} \sum_{\sigma=\uparrow,\downarrow} \epsilon_n a_{n,\sigma}^\dagger a_{n,\sigma} \] (3.2)
\[ H_{\text{dot}} = U \sum_{m=0}^{M} \sum_{\sigma=\uparrow,\downarrow} d_{m,\sigma}^\dagger d_{m,\sigma} + \sum_{0}^{M} (\epsilon_m \pm g\mu_B) d_{m,\sigma}^\dagger d_{m,\sigma} \] (3.3)
\[ H_{\text{transition}} = \sum_{m=0,\sigma,\sigma'=\uparrow,\downarrow}^M T(a_{\sigma}^\dagger d_{\sigma'} + d_{\sigma'}^\dagger a_{\sigma}) + H.C. \] (3.4)

Here, \( a^\dagger(a) \) represents the creation (annihilation) operator for electrons in the conduction band reservoirs and \( d^\dagger(d) \) represents the creation (annihilation) operator for the electrons in the quantum dot. The summation over \( \sigma \) accounts for the spin orientations of the electrons. The electrons in the left and right reservoirs behave like normal Fermion filling. On the dot, both orbital energy and charging energy is taken into account. In order to correct for the magnetic field, the plus \( g\mu_B \) corresponds to anti-aligned spins states on the dot and the minus corresponds to the aligned spin states. The transition component represents the combination of the destruction of an electron on the dot and a creation in one of the reservoir and the reverse process in which an electron is created on the dot and destroyed in the reservoir.

This Hamiltonian, to first order interactions without magnetic field, describes the conductance behavior given in the previous chapter. The finite transition rate to and from the quantum dot defines a timescale in which virtual states can occupy the dot. Unlike the previous chapter as the dot can occupy higher energy states around the ground state conductance peak, this virtual state exists within the Coulomb blockaded regime. Figure 3.1 (a)-(d) demonstrates the tunneling and relaxation through the \( N+1 \) virtual state when the \( N \) (odd) electron dot energy is below the Fermi energy and the \( N+1 \) (even) electron dot energy is above.

For zero magnetic field, \( B = 0T \), the tunneling process that returns the dot to the original spin state, elastic co-tunneling, and the process that flips the dot spin, inelastic co-tunneling, are equivalent in nature\[16, 17, 18, 19\]. However, as the magnetic field is increased, the free energy of the dot is spin dependent and the ground state has a well defined magnetic moment.
Figure 3.1: (a) The SET is at equilibrium and is configured to an odd occupancy state well within the Coulomb blockade regime. A magnetic field causes the parallel and antiparallel spin states to be separated by the Zeeman energy and the ground state contains the aligned spin. (b) The virtual state occupied by a second electron in the orbital violates energy conservation. (c) The quantum dot relaxes into the original aligned spin state which is called the elastic co-tunneling scattering process. (d) A finite bias conserves energy when the dot relaxes into the anti-aligned spin state which is called the inelastic co-tunneling process. (e) The bias conductance data for the $B = 8.7$ Tesla co-tunneling regime.
In this arrangement, only the elastic co-tunneling event is permitted at equilibrium. In order to allow the inelastic process to occur, the source-drain bias needs to be equal to the difference in free energy of the quantum dot which is the Zeeman energy, $\delta E = g \mu B$. At this bias, the higher Fermi energy can supply the electron to the virtual state and the lower Fermi energy receives the tunneled electron. The difference in the Fermi energies constitute the gained energy of the quantum dot. This has been shown to fit to an empirical form very well[20, 17].

$$\frac{dI}{dV_{ds}} = A_e + A_i[F\left(\frac{eV_{ds} + \Delta Z}{k_B T}\right) + F\left(-\frac{eV_{ds} - \Delta Z}{k_B T}\right)] \quad (3.5)$$

where $A_e$ and $A_i$ are the elastic and inelastic conductances and $\Delta Z$ is the full Zeeman energy. $F(E)$ describes the electron tunneling between the reservoir and the quantum dot,

$$F(x) = \frac{1 + (x - 1)e^x}{(e^x - 1)^2} \quad (3.6)$$

The function $F(x)$ emerges from the resulting tunneling current, equation 2.1, through an insulator with an energy absorption mechanism connected to two electron reservoirs[20]. As seen in equation 3.5, there is a bias voltage threshold, +/- $\Delta Z$, to contribute substantially. This functional form has steps located at the Zeeman energy and have widths of $eV_{ds} = 5.4k_B T$. Importantly, the co-tunneling conductance is not a function of the total dot energy and persists through the Coulomb blockaded region whenever the dot occupancy is odd. The true power of this fitting function is the ability to extract electron temperatures and the effective g-factor which define the magnetic energy scales in a SET. This tunneling model sufficiently depicts the co-tunneling regime because it considers the transfer of energy to impurities as an electron tunnels from the source to the drain electrode. This is precisely the case for co-tunneling and, importantly, the spin flip process in a magnetic field. In order to leave the quantum dot into an excited state, the tunneling electron needs to “deliver” energy as it tunnels. This requires a finite bias voltage to make the transition to a lower energy.
Table 3.1: The electrode voltages used to sweep the tunneling rates for all three device configurations.

<table>
<thead>
<tr>
<th>COT1</th>
<th>$V_s$ (mV)</th>
<th>$V_t$ (mV)</th>
<th>$V_b$ (mV)</th>
<th>$V_g$ (mV)</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>-800 → -872</td>
<td>-816</td>
<td>-1151</td>
<td>-938 → -792</td>
</tr>
<tr>
<td>COT2</td>
<td>-960 → -1025</td>
<td>-750</td>
<td>-1090</td>
<td>-795 → -671</td>
</tr>
</tbody>
</table>

3.2 Tuning the Tunneling Rate

As described, the lifetime of the virtual state is dependent on the tunneling rate on and off of the quantum dot. The co-tunneling data presented in this chapter compares the response to the elastic and inelastic co-tunneling conductances as the tunneling rate is varied. Chapter 2 has already described the mechanisms to measuring the tunneling rate in the Coulomb blockade regime. We now invoke the thermally broadened Breit-Wigner form of the charging conductance line shape, equation 2.17, to measure the tunneling rates for each electrode configuration used.

Two configurations containing different numbers of electrons are investigated in the next section. The two configurations can be described by their electrode voltages as seen in Table 3.1: COT1 and COT2. As described full in detail in the appendix, the gate voltage is swept to maintain the quantum dot energy level relative to the Fermi energy of the conduction electrons. As the side voltage, $V_s$, becomes more negative, the tunnel barrier increases and the tunneling rate drops. This is the technique used to vary each configuration such that the tunneling rate varies and the center of the valley is maintained.

In the experiment which measures the tunneling rate change, the capacitance ratio and the charging energy needs to be examined as well. Such a strong change in the tunneling potential causes a physical shift in the quantum dot electronic wavefunction. This shift often changes the effect of the gate voltage to the dot energy level. As seen in Chapter 2, this factor is necessary in defining the energies of the SET system. For several $V_s$ in the COT1 configuration, we examine the changes to the diamond structure of the conductance pattern. Shown in Table 3.2, three values of the side gate demonstrate the effects on the
Table 3.2: The side gate, which is the primary control over the tunneling rates in our SET, is varied and the control parameters of the SET are measured for three representative values for $V_s$. As the side gate becomes more negative the tunneling rate is reduced and the gate dependence is enhanced.

<table>
<thead>
<tr>
<th>$V_s$ (mV)</th>
<th>$\alpha_g$</th>
<th>U (meV)</th>
<th>$\Gamma$ (meV)</th>
</tr>
</thead>
<tbody>
<tr>
<td>-800</td>
<td>0.027</td>
<td>2.77</td>
<td>0.19</td>
</tr>
<tr>
<td>-850</td>
<td>0.03</td>
<td>2.89</td>
<td>0.06</td>
</tr>
<tr>
<td>-900</td>
<td>0.036</td>
<td>3.11</td>
<td>0.03</td>
</tr>
</tbody>
</table>

diamond for $V_s = (-800\text{mV},-850\text{mV},-900\text{mV})$. The key features of the changes are that the gate dependence of the dot energy increases and the charging energy increases. These results are consistent with the changes in the electronic wavefunctions. As the side gate becomes more negative, the quantum well is pushed towards the gate electrode and away from the source and drain electrons. This reduces the capacitive influence of the reservoirs and more heavily relies on the gate voltage, thus enhancing $\alpha_g$. As seen in equation 2.2, the charging energy, $U$, is inversely proportional to the total capacitance and so removing the capacitance from the source and drain increases the energy needed to charge the device.

Measuring the behavior of $\alpha_g$ as $V_s$ varies allows the energy calibration necessary to give the conductance charging peak as a function of the gate voltage meaning as an energy scale. Fixing every parameter besides the gate voltage, we measure the conductance as we vary the gate voltage across both charging peaks enclosing the odd occupied co-tunneling valley. Both peaks are fitted to the thermally broadened Breit-Wigner formula to extract the tunneling rate at that gate voltage. Both peaks are used due to the small gate voltage dependence on the tunneling rate. The tunneling rate at the center of the valley is approximated as the average of both surrounding charging peaks, $\Gamma = (\Gamma_{N+1}+\Gamma_{N-1})/2$. For the three sample arrangement of COT1 shown in Table 3.2, we note the tunneling rate can be easily reduced by one order of magnitude.

Figure 3.2 demonstrates the relationship between the negative voltage on the side gate, $V_s$, and the tunneling rate. Agreeing with any quantum tunneling scheme, taller and wider tunnel barriers result in lower tunneling rates. In (a), several conductance peaks for COT 2
Figure 3.2: (a) Representative charging peaks are shown for different values of the side gate, \( V_s \). The width of the peak decreases as the voltage becomes more negative. (b) The fitted tunneling rate, \( \Gamma \), is shown to be linearly related to the side gate voltage.

are shown which have a reduction in the width if the tunneling rate as the side gate becomes more negative. These peaks are fitted to the thermally broadened Breit-Wigner form. These tunneling rates are plotted in (b) as a function of the side gate voltage. With this control, the co-tunneling conductance as a function of the tunneling rate can now be analyzed.

### 3.3 Elastic and Inelastic Co-tunneling Conductance

The co-tunneling mechanism that produces a finite conductance within the Coulomb blockade regime is strongly related to the tunneling rate which we have observed to be measurable by fitting the Coulomb blockade charging behavior. Importantly, the transmission coefficient, \( T \), in equation 3.4 is a function of the tunneling rates and the co-tunneling conductance amplitude is dependent on these parameters. To isolate the elastic conductance from the inelastic conductance, we apply a magnetic field much larger than the thermal excitations. In this experiment, we used \( B = 8.7 \) Tesla which is more than five times the natural step due to an electron temperature of 55 mK. The elastic conductance is given as \( G_0 = A_e \) and the inelastic conductance here is described as the conductance beyond the inelastic voltage threshold, \( G_+ = A_e + A_i \) in equation 3.5. We measure both the elastic and inelastic
co-tunneling conductances at all three occupations and their respective tunneling rates. For all occupations, both conductances are reduced below our measurement threshold as the tunneling rate becomes very small, \( \approx 20 \mu eV \). Figure 3.3 plots both the elastic conductance and the inelastic conductance as the tunneling rates are varied. For the first co-tunneling occupancy the conductance reaches the measurement threshold when the tunneling rate is 0.038 meV and the second occupancy reaches the threshold at 0.014 meV. We find that the elastic and inelastic behavior follow similar exponential behavior at higher tunneling rates for both occupancies. At lower tunneling rates the difficulty of measuring the conductance produces larger deviations from the high tunneling rate trends. The unique feature of the conductances exist in the ratio between the inelastic conductance and the elastic conductance, \( G_+ / G_0 \). For all measurements the ratio is greater than 2 which naively is expected if both processes behave as identical channels in a Landauer transport picture. The Landauer formula considers quantum transport through a tunneling process which can be described by a set of transmission probabilities and has the form

\[
G = \sum_i n_i |T_i|^2 \frac{e^2}{\hbar}
\]  

(3.7)

where \( I \) represents the individual transport mechanism, \( n_i \) is the degeneracy of the mechanism, and \( T_i \) is the transport coefficient. The last term is the quantum of electrical conductance. This behavior was seen in the Coulomb blockade theory where the degeneracy was one and the transport coefficient was determined by how close to resonance the system was in and how symmetric it is. If both the elastic and inelastic co-tunneling processes couple to the quantum dot equivalently, we would assume that each would contribute the same percentage of the conductance quantum and ratio of conductances would be 2. Having a ratio greater than 2 has been calculated[21] by our collaborators Drs. Ngo and Ulloa at Ohio University using the microscopic interaction Hamiltonian described above based on the measured parameters from our experiment. They calculate the ratio to be slightly less than
Figure 3.3: (a) and (b) $G_0$ and $G_+$ are plotted as a function of the fitted tunneling rates for both COT 1 and COT 2, respectively. (c) For both (a) and (b) data, the ratio of the inelastic co-tunneling conductance to the elastic co-tunneling conductance is given for the all tunneling rates.

the 2.4 measured experimentally. This calculation was performed in general by Lehmann and Loss in 2006[22]. They found that the ratio should reside near 2.4 for small tunneling rates and increase to approximately 3 for larger tunneling rates. These differences may be due to unexpected excited state interactions or that increasing bias brings the SET closer to the mixed-valence regime. The conductance ratio is also measured as a function of the dot energy level. By varying $V_g$, we measure all of the conductances as the level approaches, but never enters, the charging regime. We have guaranteed that $-\delta\epsilon_d/\Gamma$ and $U-\delta\epsilon_d/\Gamma \geq 4$. This prevents measuring the conductance enhancement due to resonant levels instead of the co-tunneling enhancement. Unlike the variable $\Gamma$ experiment, we find small, but notable, changes in the conductance ratio. Near the center of the valley, the ratio again is $\approx 2.4$ but
is parabolic around the center of the valley. This parabolic behavior may be caused again by the small influences of the mixed-valence regime that becomes more expressed away from the center of the valley. In all the dot occupancies the inelastic to elastic conductance ratios agree well with the microscopic model predictions.
Chapter 4

The Kondo Effect and the Magnetoconductance

Starting with de Haas’s measurements of gold wires in 1934[3] and continuing through the 1950s experimenters continuously investigated the anomalous logarithmic increase in the resistivity of a dilute magnetic alloys when the temperature was lowered below a certain temperature. Many experiments investigated the relationship between the impurity concentration and the low temperature resistance increase. At the time, it was speculated that this was a spin related phenomenon. In 1964, Jun Kondo proposed that intermediate states of the second order terms should be taken into account when considering the s-d interaction model for dilute magnetic alloy systems[4]. He demonstrates that this term would carry a resistivity term proportional to log(T). Shortly after, in 1966, Schrieffer and Wolff showed that the Kondo model Hamiltonian is consistent with the Anderson model Hamiltonian, by means of a unitary transformation[23]. Furthermore, the characteristic temperature of the Kondo effect was explicitly shown by Haldane in 1978 as a function of the model parameters.

We now enter the era of reduced dimensional model systems. In 1988, Glazman and Raikh showed that the conductance through an isolated region of bound electrons should exhibit the Kondo effect when the net spin in the region was non-zero, and of particular interest
A decade later, in 1998, Goldhaber-Gordon successfully observed this effect in a SET[26]. The simple question now is “why continue to study the Kondo effect?” First, the Kondo system is related to popular topics in condensed matter physics such heavy fermions and high-T_c superconductors. Secondly, the great successes in demonstrating agreement between theory and experiment permit a fantastic sandbox for innovative techniques, both theoretically and experimentally. The Kondo effect has been observed carbon nanotube devices[27, 28], nanowire devices[29], graphene devices[30], and quantum ring systems[31]. Exotic Kondo states are also under investigations, such as singlet-triplet transitions[32, 33], multi-dot Kondo effects[34, 35], and multi-orbital Kondo effects[36, 37, 38]. Third, much is still unknown about controlling the Kondo effect in SET, primarily correlations out of equilibrium and the dynamics of the Kondo correlations.

This chapter covers an introduction to the Kondo effect from the standpoint of a simplistic explanation of the second order spin-flip interaction that drives the Kondo effect. The use of characterization tools to express the idea of universality of of the Kondo energy scale is shown. The heart of this chapter demonstrates the relationship that the characteristic energy scale has in both temperature decoherence and magnetic field dot polarization observed in the equilibrium conductance.

### 4.1 The Spin-Flip Interaction

The ability to control the electron number described in chapter 1 permits tuning of the SET into an odd occupancy. As the dot fills with electrons, each orbital fills with both a spin up and spin down electron, odd occupancy leaves the dot with a net spin. The spin on the dot constitutes the equivalence to the magnetic impurities in the bulk alloys. This creates a single scattering site which the Kondo theory is based. A microscopic Hamiltonian can be constructed for the electrons in a single impurity and neglecting the impurity-impurity interactions. This is valid due to experimental evidence that the resistance ratio,
\( R(T=0)/R(T_{\text{min}}) \), is independent of impurity concentration. The simplified single particle Hamiltonian which describes the behavior in chapter 2 can be written as the sum of the conduction electrons and the spin-interaction with the impurity electrons.

The Coulomb blockade Hamiltonian representing the hybridization with local energy levels presented in the previous chapters describes the charging feature seen in chapter 1 and needs to be neglected for consideration of the Kondo model. This assumption, often called the “infinite-U” approximation of the Anderson model is very valid for the bulk representation of the Kondo effect. In bulk experiments where the Kondo effect is observed, the fixed energy levels in the d- and f-orbitals are often very separated such that ionization of the local impurity atoms is rare. The final description of the Kondo model extension only relies on spin-interaction perturbations from the Fermi liquid.

\[
H = H_{\text{left or right}} + H_{\text{Kondo}}
\]  

The Fermi liquid \( H_{\text{left or right}} \) is described as in the previous chapter to be the sum of the energy states in the reservoir. Using the form of Kondo’s original perturbation Hamiltonian for a single impurity[4]

\[
H_{\text{Kondo}} = -J[(a_+^\dagger a_+ - a_-^\dagger a_-)S_z + a_+^\dagger a_+ S_- + a_-^\dagger a_- S_+]
\]

The first two terms of the Kondo Hamiltonian represent no spin exchange with the dot and the last two terms represent the spin-flip interaction term. Kondo’s solution to a single reservoir Hamiltonian produces two results. The first is that \( J \) should be negative to agree with experimental evidence. The second is that the spin-flip interaction with the impurity demonstrates a logarithmic increase in resistivity with respect to temperature. Figure 4.1 is a graphical representation of the spin interaction terms of the Kondo perturbation. The first two terms of equation 4.2 can be seen in the lower process of the figure. Here the initial electron in the conduction band, represented by its momentum and spin state, \( k: \sigma \), interacts
with the local impurity spin and leaves in the final state, $k':\sigma$. The last terms of the Kondo perturbation is seen in the upper interaction diagram. The initial electron is scattered into the state, $k':\sigma$. The original calculations of the Kondo effect were performed for bulk material. The transition to the Kondo effect in a SET can be understood on a simple qualitative grounds. The above Hamiltonians are simplified for the purpose of basic understanding of the components of a SET model. The nature of the Kondo effect holds that electrons scatter from $k$ to $k'$ in the spin interaction. This is how bulk electrons produce resistivity. However, consider the scattering processes in a SET. There are four possibilities: left electron back scattering, right electron back scattering, left electron scattering into the right, and right electron scattering into the left. The current can be described by the strength of these processes. The conductance measurement requires comparison between two biases which will demonstrate the difference between two forward scattering probabilities. If the scattering mechanism is completely turned off, there is zero probability that an electron scatters through the dot. As the strength of the scattering interaction is increased these probabilities increase. In contrast to bulk material, which the scattering increases resistivity, the Kondo effect increases
Figure 4.2: (a) In bulk material, the spin-flip interaction which describes the Kondo effect scatters electrons from $k$ to $k'$. This scattering increases resistivity. (b) The Kondo scattering process in a SET can either back scatter or transport electrons. This increases conductance.

the conductance through the device. This can be seen schematically in 4.2. The left panel describes the scattering off several impurity sites. As an electric field is applied within the metal, the transport electrons develop a net momentum in the direction of the field and any scattering event reduces this net momentum, adding to resistance. The right panel shows the four paths which describe a two reservoir system. The process combinations which involve electrons having a net flow from the left to the right reservoir produces a tunneling current which is attributed to the Kondo effect.

4.2 Universality of the Kondo Effect

In the low temperature limit of Kondo model ($T \ll T_K$), the electron behavior of the Kondo state is often referred to being universal with respect to a single energy scale, the Kondo temperature ($T_K$). The result is that the major thermal and electrical properties [$C(T), \chi(T)$, and $\sigma(T)/\sigma(T=0)$][39] only relate to a single parameter in this low temperature regime. This universal behavior of the Kondo effect arises from the connection between the scattering cross-section given by the Friedel sum rule, $Q \sim \sin^2[\delta \sigma(\delta - \epsilon)]$, and the variation of the
Describing the relationship between the heat capacity, the magnetic susceptibility, and the conductivity, A.A. Abrikosov[39] applies the Fermi liquid approximation to the Friedel sum rule. The result is an approximation to the phase element $\delta_\sigma(\epsilon - \mu)$.

$$
\delta_\sigma(\epsilon - \mu) = \delta_0 + \alpha(\delta - \epsilon) - \frac{1}{2}\sigma \phi r
$$

(4.3)

$\alpha$ and $\phi$ are two interconnected constants related by fixing the phase when both the electron excitation energy $(\delta - \epsilon)$ and the chemical potential $(\mu)$ are both varied equally; $\alpha + \frac{1}{2}n_0 \phi = 0$. This relationship maintains that the Kondo effect is tied to the Fermi level of the conduction electrons. In the above equation, $r$ is the difference between the spin species density of states. Using renormalization techniques, Wilson showed that the characteristic parameter, $\alpha$ can be described, having units of inverse energy, as

$$
\alpha^{-1} = 3.08\epsilon_F(|J|n_0)^{1/2}e^{-\frac{n_0}{\pi}}
$$

(4.4)

and this universal parameter defined the Kondo temperature $\alpha^{-1} \equiv T_K$. Direct application of the scattering cross-section yields the universal low temperature conductance behavior with respect to the Kondo temperature,

$$
\sigma(T) = \sigma(0)(1 + \pi^2\alpha^2T^2) = \sigma(0)[1 + \pi^2\left(\frac{T}{T_K}\right)^2].
$$

(4.5)

Summarizing the results in the text referenced above by Abrikosov[39], the variation of the spin species density of states due to the spin-spin impurity interaction can be expressed as

$$
\frac{\partial n_\sigma}{\partial(\epsilon + \delta\epsilon_\sigma)} - \frac{\partial n_\sigma}{\partial\epsilon} = \frac{\alpha n_m}{\pi}
$$

(4.6)

where $n_m$ is the impurity per volume. The linear behavior of the heat capacity with respect
to the density of states yields a variation of the heat capacity,

\[ C_m = \frac{2}{3} \pi n_m \frac{T}{T_K}, \tag{4.7} \]

and the natural spin species difference of the phase \( \delta_\sigma \) yields a modified susceptibility,

\[ \chi_m = \frac{4}{2} \pi n_m \beta^2 \frac{T}{T_K}, \tag{4.8} \]

where \( \beta \) squared is defined by the usual paramagnetic susceptibility of electrons, \( \chi = \beta^2 n_0 \).

Though not rigorously proved in the dissertation, the results for the low temperature heat capacity, magnetic susceptibility, and conductance, all prove to contain a single universal energy parameter which will be later shown to be extracted from the conductance behavior as the temperature of the conduction electrons are increased.

### 4.3 The Many-Body Kondo Singlet and its Energy Scale

#### 4.3.1 From Co-Tunneling to Kondo

The spin-flip interaction describes the microscopic mechanism for the increased scattering off a magnetic impurity. This picture can lead to confusion about the difference between the Kondo state and the co-tunneling regime. In both cases, the spin-flip virtual state is investigated. The spin-flip interaction creates a virtual state in which energy is not conserved, though it relaxes fast into two possibilities, as seen by the Kondo Hamiltonian. The spin-flip component is the part in which the dot is no longer in the same spin configuration as it was before the interaction. For a dot far away from the Coulomb blockade charging regime, both virtual states are in continuous participation. Particularly, the co-tunneling regime demonstrates the spin-flip interaction distinctly as a coherent tunneling effect when a large magnetic field is applied. In order for the virtual state to relax into the anti-aligned, Zeeman split state, the device requires a finite bias to compensate for the energy. Experimentally,
the temperature can be lowered below a characteristic temperature in which the equilibrium
conductance is no longer reduced but is enhanced, the Kondo effect. The same effect is seen in
odd occupancies as the tunneling rate is increased from a low conductance, co-tunneling state,
to a high conductance, Kondo state. So what makes the weakly coupled (small tunneling
rate) co-tunneling regime so different from the strongly coupled (large tunneling rate) Kondo
regime? The answer lies in the changes in the local density of states due to spin-spin
correlations that only emerge in the strong coupling limit.

4.3.2 The Kondo Singlet

As a simple example of the exchange interaction (which produces correlations required for
the Kondo state), two hydrogen atoms that are very far apart do not feel any inclination to
change their inherent spin state. As the atoms become closer and closer, the two electronic
wavefunctions become intertwined and the ground state of the diatomic molecule forms a
singlet state. In a quantum dot weakly coupled to the reservoir electrons, the net magnetic
susceptibility on the dot is favorable for the ground state of the system. The co-tunneling
interactions do not maintain correlated electrons because the tunneling events are far too
infrequent. The electrons relax into the Fermi liquid in the reservoirs after participating in
the virtual state interaction. As the dot becomes more strongly coupled to the conduction
electrons, the system favors a many-body correlated ground state which contains a zero net
spin, called the Kondo singlet.

The Kondo singlet can be interpreted as the conduction electrons screening the net spin
of the dot. The conduction electrons begin to form spin-spin correlations with the bound
electronic states and, at zero temperature, all of the electrons in the vicinity of the Fermi
energy are contributing to the correlated state and the tunneling barrier becomes trans-
parent. The entirety of modern Kondo research investigates how this coherent many-body
state is affected when external forces begin to break down the weaker electron correlations
farther from the Fermi energy, beginning at the first observed challenge to the Kondo state:
4.3.3 Theoretically Predicting the Conductance Enhancement

The work by Haldane[40] extends the above theory into a characteristic energy scale for the Kondo system focused on the energy scales associated with the Anderson model described in chapter 2. Considering the tunneling rate, the charging energy and the orbital energy relative to the Fermi energy, the Kondo temperature can be represented as followed.

$$T_K = \frac{\sqrt{\Gamma U}}{2k_B} e^{\frac{\pi e_0(e_0 + U)}{\Gamma U}}$$

Note that the Kondo temperature is a function of the dot level and $T_K$ exponentially increases when the gate voltage is varied around the center of the odd occupied valley. This Kondo temperature is predicted to be the only necessary energy scale in the system to determine the conductance. Simply being able to understand the equilibrium conductance in terms of the Haldane formula (equation 4.9) does not make a SET unique. In fact, in order to measure the transport conductance a bias voltage is require to be supplied and the current measured. Being capable of supplying an arbitrary bias enables a measurement of the density of states.
enhancement due to the set of spin flip excitation composing the Kondo effect. The current has been given theoretically by using the Majorana Green’s function [41]

\[ G(\varepsilon, E_K, B) = \frac{\varepsilon + iE_K}{(\varepsilon + iE_K)^2 + (g^*\mu B)^2} \tag{4.10} \]

where the characteristic Kondo energy (E_K), the magnetic field (B), and the energy (\varepsilon) are used. The current is then given as the total number of states available in the transport window between the left chemical potential and the right chemical potential, V_{ds}, which is the source-drain bias voltage. The full expression for this current is given as

\[ I(V_{ds}) = \int_{-\infty}^{\infty} G(\varepsilon, E_K, B)(f_L(\varepsilon + eV_{ds}, T) - f_R(\varepsilon, T))d\varepsilon. \tag{4.11} \]

Here, f(\mu, T) is the Fermi function at chemical potential \mu and temperature, T.

the reduction in conductance is caused by the reduced coherence of the spin states that comprise the correlations. The empirical form for the conductance as a function of the temperature is described as being universal with respect to the Kondo temperature [26, 42, 43].

\[ G(T) = G(0)[1 + (2^{1/0.22} - 1)(T/T_K)^2]^{-0.22} \tag{4.12} \]

Here the Kondo temperature is defined as the temperature in which the conductance is half the zero temperature value. The functional exponent s=0.22 is predicted from the renormalization group calculations for the spin-1/2 Kondo effect. This universal behavior has been demonstrated in several experiments.

### 4.4 Experimental Observation of the Universality of the Kondo Effect

To this point, the Kondo effect has been described macroscopically as impurity spin screening and microscopically as a spin-flip interaction that permits scattering through a Coulomb
blockaded SET. The concept of universal behavior with respect to the Kondo temperature and how this is seen in a generalized conductance as a function of temperature is described in this section.

The conductance behavior in a SET due to the resonant tunneling behavior of Coulomb blockade has been shown in chapter 2. The Kondo conductance modifies this picture in the parameter space that defines the Kondo regime, namely at an odd occupancy dot at equilibrium and far from the charging peaks. Being far from the charging peak is necessary to maintain a "nearly infinite" U model. This means that the Kondo model is within predictability around the middle of the Coulomb valley even though there is distinct conductance enhancements across the entire valley.

The quintessential observation of the spin-1/2 Kondo effect is the even-odd valley occupancy behavior. Measuring the equilibrium conductance as a function of the gate voltage across many occupancies demonstrates conductance enhancements in alternating valleys. Even occupancies show blockaded behavior and odd occupancies do not. The Kondo effect appears throughout the entire odd occupancy because the density of state increase is at the Fermi level, not the level of the dot. As long as the level is occupied, the enhancement is observed at the same energy as the reservoirs and conductance is always increased.

4.4.1 Non-Equilibrium Conductance

In the charging regime, the SET demonstrates the density of states increase as the gate voltage is increased. The gate voltage behavior of the Kondo regime does not permit the same measurement because the enhancement persists across the valley. The typical modification to the Coulomb Blockade diamond in the Kondo regime is the thin band of increased conductance along the odd occupied valley. The Kondo conductance behavior does not persist away from equilibrium and the conductance is reduced. Typically, the lineshape of the Kondo enhancement in the source-drain bias is described as the density of states. At a finite bias, each reservoir builds a resonance with the spin on the dot and the important param-
Figure 4.4: (a) The Kondo enhanced conductance is clearly visible around equilibrium when the SET is tuned into a strong coupling regime. (b) A constant gate voltage cross-section of the conductance in the Kondo regime.

Para is how well the correlations couple the reservoirs. As the bias increases, this coupling is reduced and the resonance with each reservoir is reduced. This produces an all around reduction in the conductance.[41, 44]

4.4.2 Measuring the Universal Kondo Temperature

As seen in eqn. 4.12, the Kondo temperature can be extracted from the conductance behavior as a function of the temperature[26]. The particular behavior is that the zero temperature conductance is halved when the system is at the Kondo temperature. By applying a current across a resistor attached to the mixing chamber of the dilution refrigerator, the temperature of the electrons can be increased. Universality is described as the collapsing of all temperature dependent conductance traces into the single form of the conductance regardless of the Kondo temperature. The simplest method to demonstrate universality is to measure the temperature dependent conductance across the Kondo valley. According to the Haldane formula, eqn 4.9, the Kondo temperature is a function of the gate voltage and so this provides a useful variation of the Kondo energy. Fitting to eqn 4.12, I extract the Kondo temperatures
Figure 4.5: (a) The equilibrium conductance for several gate voltages as a function of the mixing chamber temperature. (b) The data from (a) when the conductance and temperature are scaled to the T=0 conductance value and T_K respectively. (c) The gate voltage dependence of the Kondo temperature and the conductance at high and low temperatures are shown.
as a function of the gate voltage and which shows parabolic behavior as expected. Away from
the center of the valley the Kondo model breaks down as the charging behavior increases.
This provides a range in which we measure the Kondo behavior. The functional exponent,
s, is measured to be $0.21 \pm 0.01$, which agrees with the theoretical value of 0.22.

The temperature dependent scaling is shown in figure 4.5. For several gate voltages,
the raw conductance is plotted versus the mixing chamber temperature. For all the gate
voltages, the conductance is clearly different. In figure 4.5 (b), the conductances have been
scaled to their low temperature values and the temperature has been scaled to their fitted
values as shown in equation 4.12. In order to perform this fit, the electron temperatures
need to be determined. Assuming the linear low temperature thermal conductivity,

$$\dot{Q} = \int_{T_{M/C}}^{T_{\text{electron}}} \lambda T dT,$$

a new temperature of the electrons have been added. Here, $\lambda$ incorporates the thermal
conductivity and the heat flow path dimensions. This relates to a squared temperature
function along a base temperature,

$$T^2_{\text{electron}} = T^2_{M/C} + T^2_{\text{base}}$$

where $T_{M/C}$ is the measured mixing chamber temperature and $T_{\text{base}}$ is a fitting parameter.$T_{\text{base}}$ is related the thermal equilibrium heat flow out of the system through the conductivity
of the attached wires and is interpretable as the lowest temperature the electrons can reach.
For this experiment, the fitted $T_{\text{base}} = 100$ mK. This measurement is made when the ther-
rometer is attached to the mixing chamber which introduces noise into the system. This
adds heat flow to the electrons and is notably larger than the 70 mK electron temperature
measure via cotunneling without the thermometer cables attached.

The gate dependence of the conductances in figure 4.5 (a) are no longer presence in (b)
which demonstrates the scaling nature of the Kondo effect with respect to temperature.
Table 4.1: The electrode voltages used to throughout this dissertation to establish the Kondo state. The Kondo temperature for each setting is given as well as the section numbers in which they are used.

<table>
<thead>
<tr>
<th>Setting</th>
<th>SET</th>
<th>$V_s$</th>
<th>$V_t$</th>
<th>$V_b$</th>
<th>$V_g$</th>
<th>$T_K$</th>
<th>Sections</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>2</td>
<td>-850</td>
<td>-1024</td>
<td>-760</td>
<td>-1160</td>
<td>250</td>
<td>4.4</td>
</tr>
<tr>
<td>2</td>
<td>2</td>
<td>-900</td>
<td>-916</td>
<td>-822</td>
<td>-673</td>
<td>400</td>
<td>5.4</td>
</tr>
<tr>
<td>3</td>
<td>1</td>
<td>-1050</td>
<td>-876</td>
<td>-964</td>
<td>-600</td>
<td>800</td>
<td>5.4.4; 6.2; 6.3</td>
</tr>
<tr>
<td>4</td>
<td>2</td>
<td>-565</td>
<td>-800</td>
<td>-990</td>
<td>-535</td>
<td>1500</td>
<td>6.3</td>
</tr>
</tbody>
</table>

Figure 4.5 (c) shows the gate dependence of the Kondo temperature. As predicted by the Haldane formula (eqn. 4.9), the Kondo temperature shows the characteristic rise away from the center of the odd valley. The high and low temperature conductance traces are presented to demonstrate the difference in conductance drop with respect to the Kondo temperature. High $T_K$ regions observe smaller temperature ratios as the temperature is increased. This reduces the observed change in conductance across the available temperature range.

4.5 Analysis of the Kondo States

Throughout this report, many Kondo states have been used over a very long span of time. Two different SETs have been measured and for each several thermal cycling of the 2DEG have been performed. For each set of data, the Kondo temperature needed to be calibrated in order to investigate the universality of the measurement. As described above, the electrons have been heated from a mixing chamber temperature of 10 mK to 700 mK. Four Kondo states have been measured and Table 4.1 denoted the electrode voltages used and the center of the valley gate voltage. The respective Kondo temperature and the sections the state in which the is used are shown in the table also.

The four settings measured have a wide range in the Kondo temperature, from 250 mK, in the magnetoconductance experiment, to 1500mK used in the dynamic magnetoconductance experiment. Figure 4.6 describes the universal behavior of the Kondo conductance with respect to temperature increases for all of the settings used. In figure 4.6 (a), the equilibrium
conductance at the center of the valley is plotted as a function of the electron temperature for each setting. The electron temperatures have fits to $\approx 100\text{mK}$ for all settings, as seen by the similar range of temperatures; the scaling exponents fit between 0.2 and 0.25. Figure 4.6 (b) plots the electron temperatures scaled to the fitted Kondo temperature. In the fourth setting, $T_K = 1500\text{mK}$, the fitting routine was altered to only consider the low temperature conductance values, which yielded the best fit to the scaling exponent, $s = 0.25$. The higher temperature values can be seen to approach a minimum well above the $G(0)/2$ conductance value which may demonstrate a limiting feature to a pure Kondo model. The remaining three settings fit very well to the empirical Kondo formula. The empirical Kondo formula, black trace in figure 4.6 (b), is also presented to show this agreement.

The bias conductance lineshape also demonstrates pseudo-scaling features because the zero temperature conductance is believed to be half the peak value when the bias voltage is equal to the Kondo temperature[41]. Figure 4.6 (c) shows this behavior as the Kondo temperature is reduced. The larger Kondo temperatures are broader in lineshape than the lower $T_K$'s. A quantitative analysis of this feature is difficult because the lower $T_K$ traces are clearly suppressed by the finite electron temperature; their peak conductances are less than the fitted zero temperature conductance. The reduction in the peak due to the decoherence of the weakest spin correlations produces an illusionary effect which can result in misinterpreting the Kondo temperature if only the width of the bias conductance peak is considered. Comparing the peak to minimum conductance for all the states, there is a difference between all four lineshapes. The two medium Kondo temperatures, 750 and 350 mK, show strong differences between the peak conductance and the minimum conductance at the bias where the Coulomb blockade conductance meets the Kondo conductance. The 350 mK data both demonstrates a reduced conductance to that of the 750 mK data and a smaller peak width. The smaller peak width signifies the notably smaller Kondo temperature because thermally limited Kondo states appear to be broadened, as seen in the $T_K = 250$ mK data, when considering the half maximum conductance. These conductance trends
Figure 4.6: (a) The temperature dependent equilibrium conductance, scaled to its fitted $T = 0\text{mK}$ value, is given for each setting. The temperature has been scaled to the fitted electron temperature as given in equation 4.14. (b) The universal conductance behavior of equation 4.12 is shown by scaling the electron temperature to the fitted Kondo temperature associating $G(T_K)/G(0) = 0$. (c) The biased conductance lineshapes are given to demonstrate the difference in behavior away from equilibrium as the Kondo temperature is increased. The conductances are scaled to their respective $G_fitted(T=0)$ values.
require careful fitting in order to correctly obtain the appropriate Kondo temperature and simply measuring the width of the peak will overestimate the Kondo temperature if \( T_K \) is approaching the electron temperature.

4.6 Measurement of the Magnetoconductance

The universality of the Kondo state with respect to temperature is a well-known result with much empirical evidence. This section presents the universality of the Kondo state as a function of the magnetic field which is functionally identical to the empirical Kondo function. By aligning the device so that the 2DEG is parallel, within 1° uncertainty, with the magnetic field, I provide a situation in which only the spin interaction with the magnetic field is considered and neglect spin-orbit interactions. This section provides extensive equilibrium measurements to demonstrate the relationship between the decoherence affects of temperature and magnetic field. I also present the well-known non-equilibrium behavior in which the bias voltage supplies the necessary energy to establish the non-equilibrium Kondo effect.

4.6.1 Source-Drain Dependent Magnetoconductance

The spin nature of the Kondo effect makes the magnetoconductance an inherent property. As the magnetic field becomes large, the Zeeman splitting of the spin species is inevitable and a spin-flip process is no longer elastic\cite{45, 46}. As the elasticity breaks, the Kondo singlet cannot form and the quantum dot is polarized. Like temperature, the increasing magnetic field slowly removes all but the strongest correlated electrons and the conductance is reduced. At small fields, \( g \mu_B B > 0.5T_K \), the Kondo singlet screens the net spin on the dot and the conductance is reduced slowly. As the field is increased, the singlet state weakens and the spin species are expose to the magnetic field. In this limit, the Zeeman energy determines the energy shift of the resonance for both spin up and down. The equilibrium
Figure 4.7: The bias conductance measurements for several sample magnetic fields. At zero magnetic field we observe the characteristic equilibrium conductance enhancement. As the magnetic field is increased past the critical magnetic field value, $B = 3.3$ Tesla, we observe the formation of two distinct peaks at finite bias. At large magnetic fields, the split peaks are dependent upon the Zeeman energy.

The result of the spin splitting can be seen if a source-drain bias is applied to the SET[26, 47, 48]. As the Zeeman energy is applied to the spin state on the dot, a finite energy is required to flip the spin through. The bias voltage is this source of energy and the conductance is enhanced again at the bias, $V_{ds} = g\mu B$, which is the total energy difference between the two spins in a magnetic field. Experimentally, this arises as two conductance peaks in the source-drain bias and, along with temperature dependence, is regarded as a defining property of the Kondo effect in a SET. This behavior is shown in figure 4.7 for a set of magnetic fields. The total separation between the peaks is $2g\mu B$. For the screening limit, the Kondo singlet is still favorable energetically and the spin splitting effect of the magnetic field is not observed. The screening arises due to the singlet nature of the Kondo state. When the many-body correlated state forms, the overall electronic wavefunction has singlet spin behavior. The causes the state to not be effected by the magnetic field and the local spin component is screened. As the coherence breaks with increasing magnetic field, the local spin moment emerges and the peaks split. This splitting value for the magnetic field is $B = T_K/2$. 
4.6.2 Universality of the Equilibrium Magnetoconductance

As with the temperature dependent conductance, the effects of magnetic field on the Kondo singlet has been calculated with the renormalization group technique. Predictions[46] show that the magnetoconductance should behave functionally as the temperature dependence,

\[ G(B) = G(0)[1 + (2^{1/0.47} - 1)(B/B_K)^2]^{-0.47}, \]

(4.15)

with a different universality exponent, s=0.47[46]. The difference in scaling exponent describes the “rate” at which decoherence occurs. In both cases the conductance reduces to \( G(0)/2 \) when \( T_K \), or \( B_K \) is achieved but the lineshapes around these values are different. The behavior of the conductance at large \( T \) and \( B \) also change with this parameter. The conductance reduces to its co-tunneling value much slower as temperature is increased than as the magnetic field is increased. Again, experimentally, I vary the Kondo energy scale by observing the conductance at various gate voltages. Fitting to eqn 4.15, the values for \( B_K \) and \( s \) were extracted. The result in the valid Kondo regime fitted for \( s = 0.42 \pm 0.03 \). The value of \( B_K = 2.4 \) Tesla in the center of the valley and the expected upward trend as the gate voltage moves away from the center. Figure 4.8 (a) shows the equilibrium conductance decrease as the magnetic field is increased. The conductance traces for a set of representa-
tive gate voltages are given. Figure 4.8 (b) shows the same data sets, only scaled to their respective $B_K$ and $G_0$.

Figure 4.8 (c) plots the magnetic field scaling parameter across the Kondo valley. It is compared to the zero magnetic field conductance for the respective valley. There exists a notable difference between the center of the valley as observed by the conductance and the minimum in the scaling energy. This difference can be described by the small effect the gate voltage has on the tunneling rate. Whereas the model typically assumes that the gate voltage only acts an energy variable, it too adds to the confining potential. The effect is weak but shows as a linear term in the Kondo temperature as the orbital energy is changed. The tunneling rate is decreased as the gate voltage becomes more negative and shifts the Haldane equation by this small correction. The Kondo temperature is changed and the symmetry of the SET is changed also. This can be seen by the general slope of the conductance enhancement. The gate voltage affects the two tunneling rates differently creating a change in the conductance maximum as the gate voltage is swept.

4.6.3 The Universal Kondo Energy Scale

In the Kondo regime, it is predicted that there exists a single Kondo energy scale. This has been referred to in this report as the Kondo temperature due to historical reasons. However, the true universality is the energy associated with this temperature, $E_K = k_B T_K$. In association with the many-body correlations and the respective energy, this universal energy is a representation of the correlations themselves. In such a view, the source of the correlation breaking should be universal regardless, such that $g^*\mu B_K = k_B T_K$ [46]. This chapter has demonstrated the measurement of the conductance as both energies are increased past their respective $G/G_0$ values. To end the discussion of the universality of the Kondo energy scale, I present a comparison between the Kondo temperature across the valley and the Kondo magnetic field in figure 4.9. The data presented is a comparison between two different SETs at two different Kondo temperatures. The first SET is set to a Kondo temperature
equal to 1.2K[8] and the second is 0.45K. All the energies are plotted on the same scale for comparison. This demonstrates the small curvature on the second SET due to the scale. The uncertainty in the Kondo temperatures for the first SET is much larger than the second because a true $G/G_0=1/2$ cannot be reached when the device is heated to 700mK and so the fitting routine is not as sensitive to the temperature. The ratio, $T_K/B_K$, is found to be 1.65 for the first SET and 1.3 for the second SET. They are both consistent across the valid Kondo regime which suggests a real ratio between the two energies. It is wholly possible that the first SET ratio is larger than the second due to the uncertainty in the Kondo temperature, though I propose that this ratio should be further investigated whenever Kondo universality is considered in order to reach a better understanding.

### 4.6.4 Conclusions and Observations

The role of a universal Kondo energy scale has been investigated in this chapter. The strength of the Kondo correlations have been understood by the conductance through the SET. As the
correlations are weakened, the conductance is reduced. In order to weaken the correlations, both magnetic field and increased temperature have been applied to the system. As expected the conductance is reduced in both cases. In accordance with renormalization group calculations, the conductance behavior in both cases are expressed in the same functional form with different scaling exponents which reasonably agree with predictions for the spin-1/2 Kondo effect. The characteristic energy scales, $T_K$ and $B_K$, do not fully agree with the prediction that both should halve the conductance at the same energy. However, the difference between the two ratios suggest further measurements are needed, measurements which could easily become standard characterization in all future measurements of the Kondo state.

I would like to extend considerations in the magnetic field scaling for future measurements based on observations I have made. In an ideal depiction of the Kondo enhancement within the Coulomb blockade framework, only a simple conductance enhancement around equilibrium would be observed. However, the Kondo regime puts a strain on the non-equilibrium Coulomb blockade regions and warps the shape of the diamond. This produces noon-equilibrium conductances that rarely reach the cotunneling conductance limit. In most experimental observations of the Kondo effect, researchers often find a ratio of the equilibrium peak conductance to the lowest bias conductance to be a value of 2. This ratio however is not a fundamental measurement result of the system and quite often is less. As a large magnetic field is applied, the equilibrium conductance can display several behaviors as a function of the large magnetic field. Though an extensive investigation was never performed, I have personally observed that this high field tail is affected by this $G_{max}/G_{min}$ ratio. This greatly affects the fitted universality across the valley and the largest ratio possible is desirable for the best measurement of the Kondo effect.
Chapter 5

Dynamics of the Kondo Effect

A simple reality is that the universe is not static. Time plays a very integral part in physics and daily life. Often it is simply a parameter which describes the state of an object; at time $t$, a ball is located at position $x$. Time describes how a particle moves; momentum involves how an object traverses space with respect to time. Newton’s Laws require time to describe how an object interacts with outside forces; an object changes its momentum based on the time it interacts with its surroundings. Important to this work, electrical current can simply be described as the passage of charge with respect to time. In many real situations, asking how an object moves amongst its environment is not enough. What if the environment itself changes with respect to time. A popular question for introductory physics students is what is the position of a swinging ball attached to a sting before and after the breaking of the string. Clearly the changing of the environment has altered the steady state of the swinging ball. The inevitability of energy loss in physical systems introduces a new function of time; how long does a system survive in its initial state? The amplitude of an oscillating pendulum slowly reduces in the presence of a dampening force and the charge on a capacitor dissipates when connected to a resistor. For a resistor and capacitor connected to a battery, there is a well known time scale in which the capacitor collects charge, $\tau = \frac{1}{RC}$. The simple RC circuit example can be extended to having a time dependent voltage charging the capacitor. Given a
square wave pulse that alternates between on and off much slower than $\tau$, the system reaches both its “on” and “off” steady state behavior in between pulses. The natural question to follow is what happens when the pulses become faster than the intrinsic time scale in the RC circuit. The system never reaches its slow varying steady states and induces a frequency dependent behavior. In electronics, this behavior is utilized to develop frequency filters. For low frequencies, well below $1/\tau$, all of the voltage drop in a series RC circuit is across the capacitor and high frequencies the voltage drop is across the resistor. Having an output voltage defined by the resistor only permits high frequencies to pass through and defined by the capacitor permits low frequencies.

Quite often time scales in solid state physics are not as simple as the basic high-pass and low-pass filter example. Defining the intrinsic timescale may not come easily or the system may have multiple time scales. The frequency dependence of the second coefficient of viscosity as related to the dispersion of sound in a fluid has two distinct frequency behaviors[49]. For low frequencies, the viscosity has a frequency dependence; but for large frequencies, the dependent vanishes. This is attributed to the effectiveness of energy transfer between internal degrees of freedom. Another example of distinct frequency dependence is the dielectric function[50]. Across the frequency spectrum, several time scales govern the problem. These regimes are determined by the internal process that dominates that frequency range.

The Anderson and Kondo models also have internal processes that determine the time dependence of the system. The tunneling rate of electrons on and off the quantum dot has been shown in Chapter 2 to describe an intrinsic time scale. When the frequency became larger than the tunneling rate independent frequency modes emerged from the Coulomb Blockade peak and side peak structures developed. As the tunneling rate increases and the SET transitions from the co-tunneling regime to the Kondo regime, the intrinsic time scale is predicted to also transition from being proportional to the tunneling rate to the Kondo time scale which is proportional to the Kondo temperature, $hf = k_B T_K$. This chapter investigates the realization of the Kondo time scale for several frequency ranges and amplitudes. The
first section demonstrates the ultra-low frequency (adiabatic) behavior which obeys a time average current model. The following sections detail the transition into frequencies that approach and surpass the predicted Kondo time scale. The theoretical models of Schiller and Herschfield[51] and Kaminski, et al.[52] are employed to describe the dynamic conductance measurements of the Kondo effect.

5.1 Previous Work on the Kondo Timescale

The previous chapter describes how the correlations produced due to the spin-flip interaction of conduction electrons with a magnetic impurity have a specific energy scale. The strong spin flip interactions form correlations that require a finite time to be established. This work investigates the intrinsic timescale associated with the formation of these correlations by varying the bias voltage both slower and faster than this timescale. The goal is to demonstrate that the timescale is the Kondo energy scale, $k_B T_K=hf$, as predicted in theory[53]. Ultimately, the change from a system that permits correlations to form at each increment in time to a system that incorporates a new density of states needs to be investigated to determine the dynamics of the Kondo state.

Dynamics of the Kondo effect is the least known of its properties, experimentally. Many theoretical works have been proposed on a myriad of experimental systems[53, 54, 52, 55, 56, 57, 58]. In 1995, Hettler and Schoeller proposed that the frequency dependence of the Kondo density of states can be split into two regimes separated by the Kondo energy[53]. Their dynamic predictions showed the first examples of the Kondo effect present away from the chemical potential of the leads, $E_F+nhf$, for an excitation frequency, $f$. Schiller and Herschfield shortly after proposed an analytical form for the current with a time dependent bias voltage, $V(t)=V_{ds}+V_{AC}\sin(\omega t)$. This theory will be described and used to compare with experiment in the following sections. Between 1998 and 2001, new theories investigating the formation time of Kondo correlations and one experiment was performed. Nordlander et al.
investigated the increasing Kondo density of states when the bound energy level was stepped from a non-Kondo regime into the Kondo regime while Philal et al. and Schiller and Herschfield started at an energy level in the Kondo regime and stepped to a finite bias and calculated the time dependent current. All three groups determined that the correlations formed at a rate relative to the Kondo energy. Around this time, Goldhaber-Gordon et al. had measured the Kondo effect in a SET and Elzerman et al. supplied a capacitively coupled oscillation to the gate voltage. Using frequencies larger than the Kondo temperature, Elzerman et al. measured oscillation amplitude dependence of the equilibrium conductance for several frequencies. Their results showed that all frequencies behaved similarly when the oscillation amplitudes were scaled with frequency. Interestingly, their bias conductance did not demonstrate the characteristic "Kondo side bands" that were predicted by theory. Shortly after the Elzerman experiments and described in a following subsection, Kaminski et al.[52] predicted scaling behavior of the equilibrium conductance in terms of both oscillations of the gate voltage and oscillations of the bias voltage. They found agreement with the Elzerman experiment, both that an oscillating gate voltage would suppress the equilibrium conductance as the amplitude scaled to the frequency and that side bands may not form for the time dependence used. Specific to this work, Kaminski demonstrated the differences between gate and bias oscillations and predicted three regimes for bias oscillations. In 2004, Kogan et al.[59] demonstrated the conductance maximum when the bias equals the photon energy. In this experiment, the bias voltage was oscillated and the frequency used was $2T_K$. Recently in 2011, two experiments have taken new approaches on measuring the dynamics of the Kondo effect. Delbecq et al.[60] coupled a carbon nanotube SET to a microwave resonator. The complex impedance of the SET arises as a power loss in the resonator and broadens the resonance peak. Latta et al.[61] measured the frequency dependence of the quenching of Kondo correlations due to exciton formations in an optical quantum well. Relying on the overlap of the correlated state and the un-correlated state Hamiltonians, the reduced optical absorption was measured as the frequency was increased. Their primary
result of interest to this work is that the absorption curves demonstrated that the frequency dependent absorption scales with respect to $T_K$.

5.1.1 Current Calculations by Schiller and Herschfield

Expanding on their static perturbation calculations for the tunneling current, Schiller and Herschfield considered a spin dependent Hamiltonian for a SET in which the voltage difference between the two reservoirs maintains a time dependent oscillation. This system maintains a chemical potential between the leads as $\mu_L - \mu_R = V_{ds} + V_{AC}\sin(\omega t)$. They determined an analytical form for the current that generalizes the static case.

$$I(t) = \frac{e^i}{\hbar} \Gamma \text{Im} \{e^{i(eV_{AC}/\hbar)\sin(2\pi ft)} \sum_{n=-\infty}^{\infty} e^{-i2\pi nf} J_n(eV_{AC}/\hbar)g(eV + nhf)\} \quad (5.1)$$

Investigating this equation, we start from the modal terms at the end of the equation. Here, $g(x)$ is again the tunneling density of states within the window of transport given in the previous chapter. However, with the absorption and emission of photons, the energy window is adjusted by the energy of the photon(s), $nhf$. The Bessel function arises because the transition is described as a tunneling function as seen in photo-assisted tunneling. Because these are modal functions, we sum over all the modes along with their oscillatory behavior. The complex coefficient depends on the statistical dependence of the sinusoidal drive. This term is also seen when the system is instantaneously moved into a finite bias to produce current. This “ringing” describes the oscillatory behavior due to a change in the system. In a system that steps the bias, the ringing term decays as a function of the Kondo energy. In the sinusoidally driven system, this ringing needs to be compared with the natural oscillatory behavior of each mode. When one only considers the time averaged current that is measured in these experiments, the ringing term and the modal oscillations combine to produce another Bessel function term dependent upon the ratio of the oscillation amplitude and the photon energy. The result is a direct comparison between the AC time averaged current and the
DC current.

\[
\langle I_{AC} \rangle = \sum_{n=-\infty}^{\infty} J_n^2 \left( \frac{eV_{AC}}{\hbar f} \right) I_{DC}(V_{ds} + nhf/e)
\]  

(5.2)

This result, similar to the photo-assisted tunneling result, can predict the AC conductance behavior as a function of the DC characteristics. The difference between photo-assisted tunneling and dynamic Kondo effect is that the effects in the dynamic Kondo regime are dependent upon the relationship between the photon energy and the Kondo energy, not the tunneling rate as seen in photo-assisted tunneling.

### 5.1.2 Three Regimes for Dynamic Kondo Behavior

One more theoretical description needs to be pointed out in order to correctly interpret my measured AC Kondo conductance. The comprehensive theoretical work by Kaminski et al.[52] describes the zero-bias conductance behavior for a set of oscillations on the source-drain voltage. They predict five regimes that are governed by the parameters $eV_{AC}$, $hf$, and $k_B T_K$. They first define one specific relationship that divides two regimes. In this division, the photon energy must be greater than both the Kondo energy and the oscillation amplitude. In this regime, they find that the scaling parameter is the Kondo energy. This arises from the fact that the governing parameter is the decoherence time of the spin-spin correlations. The decoherence in terms of the Kondo energy is given by Kaminski et al. [52]

\[
\bar{h}/\tau k_B T_K = \frac{1}{\pi} \frac{G_U (eV_{AC})^2 T_K}{hf ln(hf/k_BT_K)^2}
\]

(5.3)

The authors also present two limits for this “fast” AC bias regime. For large decoherence rates, $\bar{h}/\tau k_B T_K > 1$ the zero bias conductance is inversely proportional to the natural log squared of the decoherence rate.

\[
G_{V_{ds}=0} = \frac{3\pi^2}{16} \frac{1}{ln(h/\tau k_B T_K)^2} G_0
\]

(5.4)
where $G_0$ is given by the tunneling symmetry of the device, $4\Gamma_L \Gamma_R / (\Gamma_L + \Gamma_R)^2$, in the Kondo regime. For a constant frequency, this parameter space covers amplitudes less than the photon energy but greater than $T_K$. The small amplitude zero bias conductance is proportional to the decoherence rate.

$$G_{V_{ds}=0} = (1 - \hbar/\tau k_B T_K) G_0 \label{eq:5.5}$$

Now we see an amplitude squared dependency on the zero bias conductance.

“Slow” AC bias can be considered under one of two conditions. The first is that the frequency is less than the Kondo energy and the second is when the frequency is less than the oscillation amplitude. This regime is considered the “adiabatic” regime. The physical interpretation is that the spin-spin correlations are capable of rearranging to form the DC current at all times. The low frequencies, in which excitations are dependent upon the Kondo energy, never develop discernible differences from making a voltage step and waiting for the Kondo time to build up to the DC current before making another step. An interesting result is that the adiabatic regime also resides at frequencies greater than the Kondo energy at large amplitudes. In comparison to the theory by Schiller and Herschfield, this can be described by a modal decomposition argument. When the amplitude becomes large enough to contain several Bessel terms, there are enough higher order modes of the driving frequency present to “fill out” the short time details of the DC behavior. The conductance is then given by the time averaged DC current in presence of an oscillating bias.

$$G_{V_{ds}=0} = \langle G(V_{AC} \cos(2\pi ft)) \rangle \label{eq:5.6}$$

To better illustrate the different regimes, I have recreated the parameter space diagram by Kaminski’s figure 3[52]. In fig. 5.1, the two parameters, $V_{AC}$ and $hf$, are scaled to the Kondo energy to produce the y and x axes respectively. The solid red lines define the fast and slow regimes. To the left of the red line, the oscillations obey the adiabatic prediction of equation
Figure 5.1: This schematic diagrams the different regimes of the time dependent Kondo problem. The solid red line is the divider between “fast” oscillations to the right and “slow” oscillations to the left. The dashed black lines divide the small (below) and large (above) amplitudes.
5.6; and to the right, the oscillations obey equations 5.4 and 5.5. The dashed black lines denote the transition from small amplitudes to larger amplitudes. It should be noted that the crossover from one regime to another is only continuous in the adiabatic limit, crossing over the horizontal dashed line. All other crossovers have are not analytically calculable as the frequency or amplitude is increased.

The unique result is that when the frequency is only slightly larger than the Kondo energy, \( hf \geq k_B T_K \), the large amplitude dynamic regime is very small, as seen by the small area between the diagonal red line and the logarithmic black dashed line. In fact, many of the frequencies used in these experiments are not even an order of magnitude greater than the Kondo energy. In this case, the large amplitudes for all frequencies may be explained by the adiabatic regime, not the dynamic regime.

5.2 Adiabatic Limit of the Kondo Effect

In order to understand the dynamic behavior of the Kondo state, I must first examine the adiabatic behavior and how to interpret results from slow oscillations. When the state is coupled to a source of photons via oscillating electric fields in the source-drain bias, the energy of the photon must be capable of enhancing the conductance of the SET by forming new correlations at energies away from the Fermi energy. The adiabatic limit does not meet this requirement and behaves in the classical regime. The simplest interpretation is that at each moment in time, the change in the voltage is not enough to greatly affect the system and the static correlations described in the previous chapter are present as they would be at that static bias voltage. From this, it is clear that the measured current through the SET can be described as a time average current that the static picture describes.
5.2.1 Predicting and Measuring the Adiabatic Regime

The adiabatic regime depends on how a sinusoidal bias oscillation changes the time dependent current. When the change is slow, both theories predict that the current behaves as a sinusoidal path through the DC current. This makes an easy expression for the time averaged current being an integral over time when only a sine wave voltage is applied on the DC current.

\[ \langle I \rangle = \frac{1}{T} \int_0^T I_{\text{static}}(V_{ds} + V_{AC}\sin(2\pi ft))dt. \]  

(5.7)

The steady state current oscillates around the static bias voltage, \( V_{ds} \), with an oscillating amplitude, \( V_{AC} \). This is demonstrated in figure 5.2 (b). After the new time averaged current is calculated, a numerical derivative is taken to generate the prediction for the adiabatic conductance, figure 5.2 (c). This calculation shows the distinct peak splitting in the bias voltage which is characteristic of the adiabatic regime. This splitting represents the amount of time that a sine wave spends at each value. When the amplitude becomes larger than the Kondo energy, the peak begins to split into two peaks at finite bias. These peaks arise because a sine wave lingers at its extremes and quickly passes through its zero value. When the DC bias voltage is comparable to the amplitude of the oscillations, the
Conductance ($e^2/h$)

-100 0 100

V$_{ds}$ ($\mu$V)

V$_{AC}$ = 70$\mu$V

Conductance ($e^2/h$)

200 150 100 50 0

V$_{AC}$ ($\mu$V)

V$_{ds}$ ($\mu$V)

Figure 5.3: (a) The equilibrium conductance measured at increasing amplitudes. (b) A sample of the lineshape agreement between the adiabatic prediction and the measured values.

biased conductance begins to collect from the equilibrium peak conductance. In reverse, the equilibrium conductance begins to drop as the majority of the current is collected from the slowly changing regions at finite bias.

In order to confirm that the time averaging prediction of the adiabatic behavior is correct, a low frequency (1 kHz) signal is introduced to the source-drain bias voltage. To do so, I have removed the typical DC source from the source-drain and introduced a waveform generator so that I can control both the AC and DC components. Such low frequency is transmitted through the DC circuitry with the same change in amplitude as the DC coupling. This provides a known amplitude applied to the device which can be used directly to compare theory with measurement. The introduction of this new instrument creates more noise than the original measurement set-up so I have ran long equilibrium measurement scans at individual V$_{ds}$ and V$_{AC}$ values. The measurements have been averaged to produce the adiabatic conductance value at that source-drain voltage. These measurements are shown in figure 5.3. Both the equilibrium conductance as a function of the amplitude and the non-equilibrium behavior for a given amplitude demonstrate good agreement with the adiabatic prediction. The agreement between measurement and prediction only using direct input of measurement parameters gives good reason to use deviations from the adiabatic prediction as evidence of a change of behavior.
5.3 Dynamic Measurements

With the good agreement between the adiabatic prediction and the 1 kHz measurements, I transition to observing the conductance behavior as the frequency nears the Kondo energy and surpasses it. The goal is to rectify the Kaminski prediction that large amplitudes behave adiabatically in order to directly compare the Schiller and Herschfield closed-form perturbation calculation. This ultimately can show the change from no frequency dependence (adiabatic) to the regime where the frequency scales to the Kondo energy (dynamic) as the amplitude is increased. A main result of this section is that the measured zero bias conductance does not behave as the Schiller and Herschfield perturbation theory predicts but does agree with small and large amplitude predictions of the Kaminski prediction.

As stated earlier, the adiabatic prediction can be used to contrast results that are distinguished as “non-adiabatic.” The first goal is to tune the device into the Kondo state and measure the Kondo temperature. For these experiments, two SETs have been measured, one at 800 mK and one at 1200 mK. The characteristic frequencies are then 16 GHz and 24 GHz respectively. As seen in the experiment by Kogan et al., the high frequency regime will be viewed in the regime of $T_K$ and above. For both SETs, these frequencies fall within the frequency bandwidth of the AC circuit.

5.3.1 Comparison to Adiabatic at $T_K=800$ mK

Figure 5.4 shows the enhanced conductance around equilibrium due to the Kondo effect. As the previous chapter describes, this enhancement exists across the entire odd Kondo valley. All measurements are considered at the center of the valley marked by a dashed line. This measurement also constitutes the static conductance which will be used for predictions. Figure 5.4 (b) demonstrates the difference between the adiabatic agreement with prediction and the excitation of a 20 GHz oscillation. The top scale is the oscillating voltage of an applied 1 kHz signal. As before, direct calculation with experimental parameters of the
Figure 5.4: (a) The Kondo enhanced conductance around zero bias is observed for a SET tuned to $T_K = 800 \text{mK}$. The dashed line represents the gate voltage in which the conductance is measured. (b) The adiabatic behavior of the equilibrium conductance of the Kondo state, scaled to the static conductance, is compared to the 20.3 GHz excitation on the device. The root power expression comes from the generators output in dB into a 50 Ohm microwave circuit.

adiabatic prediction agrees with the measured values. The lower scale is the square root of the power that the input of the generator would deliver if the circuit was replaced with the attenuation of the line and a 50 Ohm load. We know that for a resistor the power is proportional to voltage squared so I can justify that there is at least a coefficient which connects the top scale with the bottom scale. There is an obvious shift in the root power due to unforeseen differences in the room temperature behavior and the low temperature behavior. There are distinct difference between the adiabatic and dynamic regimes even if the power was adjusted. In the log plot, there is a difference in high amplitude slopes between the two which describes the difference in functional behavior.

This deviation from the adiabatic prediction does not fully describe the high frequency data as being dynamic. It could easily be surmised that nonlinearities in the conductance could affect the transmission of the microwave signals in an unpredictable manner and hence produce a monotonic transmission amplitude as a function of $V_{AC}$. I check this by observing the the biased conductance behavior for matching zero bias amplitudes of the adiabatic prediction and the 20.3 GHz data. Figure 5.5 demonstrates the lineshape behavior of the 20.3 GHz oscillations as the input power is increased from -58 dBm to -52 dBm. The difference of
Figure 5.5: (a) The static source-drain behavior of the Kondo enhancement. (b) and (c) The application of two amplitudes of 20.3 GHz oscillations and comparison to the adiabatic prediction of same equilibrium conductance.

6 dBm represents a factor of two change in voltage. When the equilibrium conductances are matched between the adiabatic prediction and the 20.3 GHz data, the high frequency data is below the adiabatic prediction. This is demonstration of divergence from the adiabatic behavior because it represents coherence breaking. In the adiabatic regime, the conductance enhancement due to the Kondo effect is “conserved” because coherence time is much shorter than the oscillations and the steady state current is an oscillation of the static I-V curve. The coherence is not broken in this regime. If the 20.3 GHz were dynamic, the reduced conductance would not be observed. As the amplitude is increased, the difference becomes much more noticeable. An interesting behavior of the lineshape is that all the data trends to the same conductance far from equilibrium. This can be explained by the fact that all excitations return to the non-Kondo limit far from equilibrium as the timescale of the non-Kondo processes are very fast in comparison.

### 5.3.2 Characterizing $V_{AC}$

Several times so far it has been noted that the true amplitude of the voltage oscillations delivered by the AC circuit is not well defined. Figure 5.4 cannot be a true understanding of the amplitude characterization even though any coefficient would simply translate the data. To describe any universal behavior with respect to the Kondo temperature and the frequency requires a characterization of the amplitude. Previous scaling by Elzerman et
al., uses the electron temperature to describe the amplitude in which the conductance is no longer the static conductance. This argument does not agree with the theory by Kaminski et al., in which the any experimentally observable change in equilibrium conductance will be a function of the frequency for the high frequency limit. The following attempts to explain a new characterization of $V_{AC}$ in the framework of the large amplitude behavior of the equilibrium conductance.

Calibrating the oscillation amplitudes can be directly understood by high amplitude predictions by both Schiller and Herschfield and Kaminski, et al. Both predictions deduce that large amplitudes should trend towards the adiabatic prediction. The perturbation prediction suggests that as more modes, $n_{hf}$, are present, the driving frequency becomes less dominant on the behavior of the current as a function of time. Kaminski’s renormalization group calculations directly relate the coherence rate to the amplitude of the oscillations. In particular, the coherence rate is not the dominant feature when the amplitudes are large. This is the basis for the characterization of the amplitude, $V_{AC}$.

To justify this calibration technique, I investigate two behaviors of the measured conductance. The first is the far from equilibrium behavior as the amplitude is increased. In this regime, the high frequency data trends towards the adiabatic prediction, as seen in figure 5.5. This behavior suggests that the dynamics of the Kondo state far from equilibrium is minimal. While it cannot be concluded whether this is the sheer reduction of the Kondo correlations due to the bias voltage or some integrated effect between dynamics and the bias, we do notice that the trend is consistent. These small amplitude out of equilibrium conductance behaviors are relevant because the time dependent current will be a result of the time spent at these high bias when a large amplitude is applied with zero bias.

The second behavior is comparing the high amplitude zero bias conductance for several frequencies. According to Kaminski’s prediction for the large amplitude and Schiller and Herschfield’s prediction for very large amplitudes, the conductance should behave similar to the adiabatic prediction. I have measured several frequencies at both low and high powers
for a single Kondo state. The intent is to demonstrate that all equilibrium conductance trends to the same behavior at very high amplitudes. To define “very high” amplitude, I use the common behavior that the equilibrium conductance begins to increase. This represents the amplitude in which the far from equilibrium Coulomb blockade behavior is noticeable. In figure 5.6 a), the conductance is plotted versus the unscaled root power for the frequencies used. Several external attenuation settings were required to obtain the wide spread amplitude range. As it is seen, the frequencies exhibit a trend that lower attenuation, and so lower amplitudes, suppress the Kondo conductance as the frequency is increased. This is clearly not in agreement with the predictions of Kaminski et al. because in such a scaling prediction, the larger frequencies require larger amplitudes to promote electron-photon tunneling processes which suppress the conductance. This experimental observation also holds no meaning in the “randomness” of power delivered to the SET. To account for this, I align the large amplitude behavior to scale the power of each frequency to the corresponding amplitude. When the amplitude is corrected, shown in figure 5.6 b), the large amplitude tails for each frequency agrees well with the adiabatic prediction. This is the amplitude calibration technique that will be used to compare the frequencies shown.
5.3.3 Frequency Dependent Conductance Suppression

As seen in figure 5.6, the amplitude calibration is very much needed to compare frequencies. More so, in figure 5.6 b), the frequency dependence of the suppression becomes visible. To understand the frequency dependent nature of the Kondo effect, I begin by comparing the low frequencies (1 GHz and 140 Mhz) which I define as the system dependent adiabatic response to a frequency faster than the Kondo time scale, 20 GHz. Plotting 20 GHz and 1 GHz, we observe that the 20 GHz peak conductance is suppressed much greater than the 1 GHz conductance for small amplitudes. This behavior directly opposes the perturbation predictions of Schiller and Herschfield which propose the relationship between amplitude and frequency. Under this assumption, higher frequencies should always require a greater amplitude to cause as much suppression as a lower frequency. This behavior can be explained by the renormalization group predictions of Kaminski which denotes a transition from the adiabatic behavior to the dynamic behavior. In particular, the scaled decoherence time given in eqn. 5.3 defines this the behavior for “fast” frequencies. Investigating this function, we see that when hf approaches k_B T_K the decoherence time is only definable when the amplitude is zero and will immediately reduce the conductance to zero for any finite amplitude. For frequencies faster than the Kondo time scale, the amplitude with which the conductance begins to be suppressed will increase monotonically with frequency. The behavior of the 20 GHz data is such that the ratio of f to T_K, hf/k_B T_K = 1.2, causes a roll-off at an amplitude below the adiabatic roll-off amplitude. It still holds that a non-perturbative method is required to describe the lower roll-off value.

I have measured two different frequencies, 20 GHz and 29 GHz, for the full amplitude range to better describe the nature of the dynamic regime. Figure 5.7 (a) shows the amplitude dependence of the zero bias conductance for both frequencies. We observe an increase in the low amplitude roll-off as the frequency is increased by 50%. This behavior is in qualitative agreement with predictions. To determine the quantitative validity of the calibration technique and the theoretical predictions, I need to compare the low amplitude behaviors
Figure 5.7: (a) The zero bias conductance dependence on the oscillation amplitude for two frequencies greater than $T_K$. (b) The amplitudes scaled in terms of the square root of the decoherence rate. Both data sets overlap in the region where the decoherence rate is less than 1.
of both frequencies. To do this I directly calculate eqn. 5.5 using the parameters \((T_K, f)\) into the scaled decoherence rate. Without any further adjustment to the amplitude beyond the calibration technique and using the suggested coefficient of \(a=1\), I find very good agreement with the roll-off amplitudes for both 20 GHz and 29 GHz. These calculations are plotted in figure 5.7 (a) as solid lines. The excellent agreement leads to a scaling argument for the dynamic Kondo behavior based on the decoherence rate. According to eqn. 5.3 the decoherence rate is universal with regards to the Kondo temperature. Whereas the Kondo temperature was not changed in this experiment, the decoherence rate can be used to compare the two different frequencies for the same Kondo temperature. This is demonstrated by rewriting the oscillation amplitudes in terms of the square root of the decoherence rate. Using this method, the axis is still proportional to the amplitude as seen by the square dependence on the amplitude in the decoherence rate. When both the 20 GHz data and the 29 GHz data are scaled in this fashion we notice that the low amplitude dependence are equivalent for both data sets. Figure 5.7 (b) shows the conductances versus the scaled amplitudes and the agreement is easily seen. In the regime where the decoherence rate is larger than 1, the scaling relationship breaks down and the frequency independent regime emerges.

For frequencies notably slower than the Kondo time scale, 140 MHz and 1 GHz, frequency independent behavior is observed as the amplitude is increased and frequency dependence emerges when frequencies are notably faster than the Kondo time scale, 20 GHz and 29 GHz. Figure 5.8 (a) plots these frequencies to compare the roll-off amplitudes which is denoted by the black horizontal line. However, the 10 GHz signal is slower than the Kondo time scale and is anticipated to behave adiabatically. The central question here is why does the 10 GHz signal suppress the Kondo peak much greater than the adiabatic frequencies. Figure 5.8 (b) plots the 90% roll-off amplitude for the frequencies used in this experiment. This figure makes it quite clear that the perturbation prediction (dashed trace) does not allow for a transition regime in which the roll-off value has a minimum. The renormalization group calculations
Figure 5.8: (a) Zero bias conductance traces as a function of amplitude for two frequencies below the Kondo frequency (1 GHz and 10 GHz) and two frequencies above (20 and 29 GHz). The horizontal black line is the 90% roll-off value. (b) The oscillation amplitudes corresponding to where the peak conductance is 90% of the static conductance. The 10 GHz data shows the roll-off drop below the Kondo frequency. The black trace represent the renormalization group predicted roll-off point in the dynamic regime. The dashed line is the perturbation predicted roll-off for all frequencies.

have a distinct change in behavior which the data demonstrates. This investigation requires understanding of how the adiabatic/dynamic transition occurs. The theory of Kaminski only supplies the appropriate results for the slow and fast regimes and does not consider the transition. Though this work does not investigate the transition regime, the 10 GHz data fully motivated the necessity for future work. Making a rather rash assumption that the decoherence time of the Kondo state still is a valid parameter, the frequency dependence will rapidly increase the roll-off amplitude past the adiabatic regime. It would be logical that the transition regime should approach the adiabatic regime as the frequency is lowered. The fact that the 10 GHz data shows suppression of the conductance at a significantly lower amplitude than adiabatic data may lead to one of two frequency behaviors. The first is that there is an abrupt change in dynamical behavior at a frequency slower than the time scale. In this possibility, the lower conductances for 10 GHz than the 20 GHz data demonstrates a compression of the roll-off amplitude below the Kondo frequency. The second, and more reasonable possibility, is that the transition is not abrupt as the frequency is increased past the Kondo frequency. The existence of a minimum roll-off amplitude between 10 GHz and
20 GHz is clearly seen in figure 5.8 (b). Future research is necessary to better explain the nature of the transition between adiabatic and dynamic behaviors.

5.3.4 Dynamics in the Bias Conductance

The primary feature of in all theoretical work is the presence of photon side bands in the out of equilibrium conductance. This prediction, as described quite often, is very volatile and is often thought to not be measurable due to the enhanced decoherence at finite bias. In this system, the dynamic frequencies seen in the previous section cannot be used in the bias analysis because the conductance behavior when $eV_{AC} = hf$ proceeds into the non-Kondo regime. The anticipated side peaks lose their meaning when other processes dominate the conductance. However, the conductance lineshape does not necessarily lose importance. Using the previous Kondo state, I compare the bias conductance lineshape as both the frequency and the oscillation amplitude is varied. The goal of this section is to better explain the transition regime, $f = 10$ GHz, by comparing the conductance peaks for both the adiabatic frequencies and the 10 GHz data.

Figure 5.9 plots the conductance value at several bias voltages as a function of the calibrated oscillation amplitude. The bias voltages used are color coded in figure 5.9 a) with the black data being the peak heights. As the bias voltage is increased all the frequencies demonstrate a reduction in conductance as expected given the natural lineshape of the Kondo enhanced conductance. These plots show several interesting features when inspected. For bias voltages near the equilibrium peak, the conductance does not decrease for small oscillation amplitudes. This is because the small bias voltage has already broken the weakest correlated electrons from the singlet state. As the bias voltage is increased, the conductance roll off amplitude increases because larger bias voltages have suppressed more energetic correlations. Ultimately, far from equilibrium, the conductance is observed to increase as the oscillation amplitude becomes very large. Interestingly, the oscillation amplitude at which a peak occurs is not constant between the transition frequency and low frequencies.
Figure 5.9: (a) The conductance vs $V_{ds}$ trace for the Kondo peak at $V_g=-673$. The vertical lines cut the bias voltages displayed in (b)-(d). (b)-(d) The conductance vs oscillation amplitude for representative bias voltages for 140 MHz, 1 GHz, and 10 GHz respectively.

At low frequencies (140 MHz and 1 GHz), the peak aligns at roughly $V_{AC,RMS}$ which is consistent with the adiabatic description. At 10 GHz, the peak is observed at a factor of five lower in amplitude. This difference represents a major shift from low frequencies to the transition frequencies which may be a demonstration of the beginnings of a shift to the dynamic Kondo regime.

Again in figure 5.9, the similarities between the two low frequencies arise while the transition frequency shows signs of disagreement. Increasing the oscillation frequency by a factor of ten does not notably change the location of the biased conductance peak with respect
to the amplitude for the low frequencies. As the frequency is increased by another factor of ten to 10 GHz, but still below the Kondo frequency, the biased conductance behavior develops peaks earlier than the lower frequencies. Even though 10 GHz is less than the Kondo frequency, we can still investigate two questions with this observation in respect to dynamic Kondo predictions. This will serve as a check as to whether or not dynamic predictions explain frequency slightly below the Kondo frequency. First, the photon side bands theoretically appear at $V_{AC}=hf$. For a 10 GHz excitation, the peak would correspond to $V_{AC} \approx 40 \mu V$ (the mustard trace) and possibly emerge at the second mode when the amplitude is equal to 80 $\mu V$. This trace does not demonstrate an increase in conductance as the oscillation amplitude reaches the photon energy. Second, the conductance peak as a function of the amplitude shifts for different bias voltages. The side bands demonstrate a logarithmic divergence as the bias voltage approaches the photon energy that is scaled as the oscillation amplitude surpasses the photon energy. This introduces whether or not the increased biased conductance due to the increasing oscillation amplitude can overcome the natural decoherence produced from the bias voltage. If so, this may produce the observed result that the conductance maximum in amplitude increases with respect to the bias voltage.

The frequency dependence of the Kondo effect are universally determined by the Kondo temperature. To check the validity of this, I have measured the time dependent conductance behavior of a second SET at a much higher Kondo temperature in order to compare to the previous results. Using the same frequency ranges as the first SET, the 10 GHz data has transitioned from being near the Kondo energy in the transition regime to being half the Kondo energy. In figure 5.10 a), the equilibrium conductance for several frequencies has been measured and, at this $T_K$, the 10 GHz conductance suppression is clearly distinct from the 16 GHz and 23 GHz conductance suppression. This can easily be seen in comparison to the data shown in figure 5.6 b). The suppression as the oscillation voltage is increased for the 10 GHz excitation is notably more similar to the adiabatic prediction as compared to the low frequencies in the low $T_K$ SET. To compare the lineshape of the 10 GHz conductance,
the conductance versus bias voltage is given in 5.10 b) and the biased conductance is given is 5.10 c). The lineshape for the biased conductance in 5.10 b) demonstrates a small positive curvature around equilibrium which is not seen in the low T_K setting. This curvature represents the relocation of the Kondo conductance to a finite bias which agrees with the adiabatic prediction. The adiabatic prediction is given to show the expected behavior for frequencies less than T_K. Clearly, similar featured are present; though the conductance is further suppressed at all bias voltages for an uncertain reason. In figure 5.10 c), the biased conductance traces show the formation of a large amplitude increase near the oscillation voltage that corresponds with V_{ds}. This is another demonstration that the 10 GHz excitation conductance has transitioned from being very dissimilar to the adiabatic prediction to agreeing with the prediction.

5.4 Conclusions and Discussion

The equilibrium measurements shown in this chapter are the first transport conductance measurements through a quantum dot to demonstrate a transition from a frequency de-
pendent regime to a frequency independent regime. Two different transitions have been observed. The first transition is from low frequency to high frequency separated roughly by the Kondo temperature. Frequencies sufficiently below the Kondo temperature show agreement through the entire amplitude range investigated. As frequencies approach the Kondo temperature, frequency dependence arises and different frequencies exhibit different amplitude dependence. In this high frequency regime, all frequencies reduce to the adiabatic limit for large amplitudes. However, these “large” amplitudes are much less than perturbation predictions, but agree with the renormalization predictions. The return to adiabatic behavior at large amplitudes permits a direct technique to calibrate the frequency dependent potential amplitudes delivered by the microwave circuit. By only adjusting the amplitude according to the large amplitude limit, two frequencies greater than the Kondo temperature suggests that small amplitude suppression of the Kondo conductance are universal with respect to the decoherence rate. These measurements are the first to demonstrate this scaling as a function of frequency as the amplitude is increased. In order to explore the transition regime for frequencies lower than, but close to, the Kondo energy, biased conductances were compared to the adiabatic frequencies and the transition frequency demonstrated a change in conductance increases far from equilibrium. Adiabatic conductances show a peak in the amplitude dependent conductances that shift roughly as $V_{ds}=V_{AC}$, whereas the transition frequency shows peaks at a much lower amplitude. Further experimental data is necessary to better understand the transition period.

Beyond being an example of novel scaling with respect to the Kondo temperature, the dynamic response of the Kondo effect demonstrates several invaluable concepts that brings the entire field of quantum dot based Kondo related research into perspective. As with the results by Latta et al, the direct scaling with respect to theoretical predictions reconfirms the Kondo temperature itself. Experimentally, the Kondo temperature has always been confirmed by thermally breaking the spin correlations. Fitting the peak conductance as a function of the temperature as seen in the previous chapter has always been assumed
accurate. The dynamic response of the Kondo state not only has effect in understanding the Kondo time scale, but reconfirms the importance of the Kondo temperature. By simply inserting the experimental parameters alongside the fitted Kondo temperature into the small amplitude prediction of Kaminski et al, a direct overlay is observed for both the 20 and 29 GHz data sets. This result is quite remarkable and enhances the experimental validity of the thermal and magnetic field measurements. Furthermore, the inclusion of the new fitting parameter for the base electron temperature is now justified and should become the experimental standard for future measurements. Without this adjustment to the temperature, the Kondo temperature would be notably less which would no longer demonstrate such excellent agreement with the frequency dependent dynamic suppression.

Lastly, the data presented in this chapter gives renewed interest in the complete modeling of a single-electron transistor. The equilibrium conductance behavior as the amplitude is increased for all frequencies demonstrate the exit from the Kondo regime. In this report, this transition only has concern for the calibration technique which is immediately justified by the low amplitude roll-off in the dynamic regime. In full regard, the transition between low frequency and high frequency agreement with theory diverges from the predicted boundaries of the regimes. It is clear that this divergence is caused by the non-Kondo mechanisms that exist alongside the Kondo density of states and emerge when the equilibrium Kondo conductance is sufficiently suppressed by the microwave signals. There is plenty of room for new physics to emerge in the dynamic regime as interplay between strongly suppressed Kondo correlations and single particle physics that exist out of equilibrium. The following chapter will divulge into anomalous magnetoconductance behavior in the presence of microwave signals.

5.4.1 Unexplained Conductance Suppression

Throughout this chapter, the conductance has shown an consistent trend of being less than expected as the amplitude of oscillations are increased. This extra suppression of the con-
ductance is by no means anomalous and is not intended to imply new physics. The probable explanation is a systematic uncertainty which further decoheres the Kondo state, of which may include electron heating and the noise spectrum. These effects, and many more, consistently plaque the entire field of quantum coherence and often are difficult to quantify.

Considering temperature, the conventional technique used to measure the electron temperature is the width of the inelastic cotunneling step in the presence of a large magnetic field. As the bias voltage is greater than the Zeeman orbital splitting, the spin-flip cotunneling process is activated. The voltage at which this is activated is determined by the thermal population of the electron gas. Time dependent excitations created in the electron gas are also present in the weakly coupled cotunneling process just as they are in the Kondo state. This prevents measurements from decoupling thermal effects from the inherent time dependent conductance. With direct measurement impossible, a consideration of the electronic heating effects in the 2DEG can be made. Given the dimensionality of the 2DEG region, roughly 100µm, and the GaAs/AlGaAs heterostructure in general, roughly 1mm, the wavelength of even the largest frequencies, 4mm, used are larger than the features in consideration. This implies that Joule heating should dominate. Even if all the power supplied by the microwave generator to the SET, -70 dBm (10^{-10}W), is much less than the heat flow from the SET to the mixing chamber (approximately 20µW).
Chapter 6

Dynamic Magnetoconductance

In the previous two chapters, the conductance has been measured as the magnetic field is increased and as the amplitude of voltage oscillations is increased. This chapter introduces a new measurement that has never been considered: dynamic magnetoconductance. This chapter cannot consist of theory for none has been proposed, though the measurements and unique behavior of the magnetoconductance should pose excellent reason for future theoretical works. The first section describes qualitative predictions for the conductance under the pretense that a simple combination of dynamic behavior and magnetoconductance is permitted in the equilibrium conductance. The second section describes the experiment performed and the anomalous equilibrium conductance behavior observed.

6.1 The Dynamic Magnetoconductance Hypothesis

The absence of theoretical work on dynamic magnetoconductance makes any true predictions very difficult for little is fully known about the dynamic Kondo effect in general. This section attempts to utilize the understanding of the two previous chapters to describe a simple combined pictorial model. The true intent is to provide insight into how remarkable the data truly is.

The simplest hypothesis of dynamic magnetoconductance easily combines both features
Figure 6.1: The predicted evolution of the Kondo density of states as both magnetic field and high frequency oscillations are present. (a) The static, B=0 Kondo density of states. (b) Magnetic splitting of the density of states at B_μB = h/2. (c) Dynamic sideband formation around each split peak. (d) The magnetic field is reduced to gμB = hf and an enhancement is formed at equilibrium.

previously discussed. When a magnetic field is applied to the SET, the spin species split as it has been discussed earlier. The split conductance peaks in the bias voltage relate to the individual correlations affiliated with each spin and should have real photon absorption and emission processes for each peak. Within the dynamic regime, the formation of dynamic Kondo sidebands should form around each conductance split peak in accordance with dynamic theory. The intent of this experiment is that when the magnetic splitting, gμB, is equal to the photon energy, the necessity of a bias voltage to flip the spin of the quantum dot is relieved by the energy of the photon. This should reestablish the Kondo effect at equilibrium. The importance of this experiment is to demonstrate that two out of equilibrium Kondo effects can combine to produce an equilibrium effect. Shown in figure 6.1 is the simplified evolution of the possible reemergence of the equilibrium conductance as the photon sidebands converge. In (a), the static, B=0 T Kondo density of states is shown. (b)-(d) demonstrate the shift from a large magnetic field splitting the spin states with photon sidebands to the overlap as a photon is emitted from one state and absorbed by the other. If this mechanism is permitted, applying a constant oscillation in the dynamic regime should produce a peak in conductance as the magnetic field is swept through the photon energy.
6.2 Measuring the Magnetoconductance in a Time Dependent System

Measuring the time dependence of the magnetoconductance simply combines the two previously discussed techniques in chapters 3 and 4. This experiment was performed on two different SETs during two different runs of the dilution refrigerator. Each SET was tuned into the large $T_K$ regime ($T_K \approx 1K$) which corresponds to a frequency of 21 GHz and a magnetic field of 8 Tesla. The large $T_K$ state was chosen because of the poor stability of a low $T_K$ state in the SET and the small curvature of $T_K$ as the center of the valley shifts in the magnetic field. The change in $T_K$ would add an extra dimension to the complexity of the problem.

6.2.1 Effects of Microwaves at High Magnetic Fields

First, I demonstrate the effects of microwave radiation on both $B = 0T$ and $B = 8.7T$. It has previously been shown that the increased amplitude for a constant frequency, 27.611 GHz in the case of figure 6.2 (a), monotonically decreases the equilibrium conductance while slowly
increasing the biased conductance. When the device is exposed to a large magnetic field, $B = 8.7T$, the conductance behavior reverses. The split peaks around $V_{ds} = \pm g\mu B$ lower in conductance when the amplitude is increases. This demonstrates the breaking of correlations when the microwaves become significant in amplitude regardless if they are in equilibrium or far from equilibrium. The equilibrium conductance increases as the oscillation amplitude is increased. This may be explained by an adiabatic depiction in which, for large amplitude $V_{AC} = g\mu B$, the equilibrium conductance begins to feel the weight of the Kondo correlations away from equilibrium. This does not explain the relatively constant conductance behavior of the bias voltages greater than the peak voltage location. In figure 6.2 (b), the conductance suggests that at bias voltages greater than $g\mu B$ the conductance shows little response, let alone the peak splitting that the adiabatic prediction contains.

As with many results in this chapter, distinct analysis is difficult to perform with the microwave response at high magnetic fields. The data does not demonstrate the conductance spread in both lower and higher bias voltages as the adiabatic prediction suggests. Previous demonstration of the conductance lineshape in the predicted dynamic regime can agree with the high magnetic field behavior in the sense that conductance increases away from the conductance peak cannot be visually observed, but can play role when the amplitude dependence is considered. The conductance increase around equilibrium and not at large bias voltages may suggest that applying a bias voltage inherently affects the intrinsic time scale of the conductance mechanism. Just as the static conductance peaks at finite bias at a large magnetic field do not maintain the total conductance of the original Kondo peak, the coherence of the state is affected significantly as the SET is driven out of equilibrium. The implies that the time dependent effects far from equilibrium are significantly weaker than the effects near equilibrium. And so, the same excitation increases the conductance around equilibrium much greater than it does far from equilibrium. This observation further motivated the proposed dynamic magnetocondcutance measurement in order to observe behavior near equilibrium.
6.2.2 Equilibrium Magnetoconductance

To test the equilibrium conductance as the time dependent behavior is driven towards equilibrium, the magnetic field is varied at constant microwave frequency and amplitude. The first measurement while maintaining a constant oscillation of 27.611 GHz and an input power of -10 dBm demonstrated a very unexpected result in the equilibrium magnetoconductance. The conductance increases at a very small magnetic field, \( B \approx 2 \) Tesla, well below the photon energy and the Kondo temperature. This measurement was made at \( V_g = -600 \) mV while scanning the bias voltage back and forth from \(-350 \mu V\) to \(350 \mu V\). The magnetic field current was constantly decreasing from 90 Amp to 0 Amp while the bias voltage sweep was performed.

Figure 6.3 demonstrates the first observation of the anomalous increase in conductance at a small magnetic field, shown as the bright white spot near equilibrium and \( B \approx 1.6T \) in (a). Figure 6.3 (b) shows the cross-sections of (a) for the lowest and highest magnetic fields while microwaves are present. The \( B = 1.6T \) trace is clearly larger than the zero magnetic field trace. To compare the increased conductance, the zero excitation conductance trace is shown. The peak in the magnetoconductance rises to \( 0.9 \frac{e^2}{h} \) which is a 25% reduction from the full Kondo conductance and a 20% reduction from the magnetically scaled conductance for \( B \).
= 1.6T and no microwaves. This peak compares to the 45% reduction that the equilibrium conductance shows due to the application of microwave. The location of the conductance peak in the magnetic field is \( \approx \frac{1}{4} T_K \) and \( \frac{1}{6} h f \).

### 6.2.3 Circuitry Influences

Next, I present data which demonstrates that the observed increase in conductance is not caused by systematic changes to the measurement due to the presence of the magnetic field. For such a large magnetic field, even slow time dependent changes in the magnetic field strength may produce large induced currents in the system which can affect the measured conductance. Primarily, this would be bound to the microwave circuitry because no conductance increase was observed in any data presented in chapter 3 and these measurements were made using the same procedure. To observe any possible effects on the conductance due to the changing magnetic field, I have fixed the magnetic field to a particular value and measured the conductance. This was continued for several magnetic field values. The data clearly presents that for static magnetic fields, the increase in conductance persists, as seen in figure 6.4.

Another possible effect of the magnetic field is the magnetization of the components of the microwave circuit which would lead to the possibility of both frequency and magnetic field dependence of the transmission amplitude. The glaring downside to this possibility is that the effect seemingly turns off as the magnetic field is increased. This is not consistent with normal magnetization effects which have a threshold field value and different behaviors above and below. However, to check the effects of the magnetic field, I have measured the reflection of the microwave signal at the source and shown that a monotonic decrease of 5% in the reflection amplitude and \( \pm 1\% \) change in the phase of the reflection as the magnetic field was swept from 0 to 8.7T. More importantly, no signs of resonant behavior was observed in the reflection. Such a resonance would be the only explanation for the reduction in reflection without the assumption that the amplitude was increased onto the device. Increasing the
amplitude is the opposite of the behavior observed.

The last possible circuit behavior is a possible rectification induced by the magnetic field and the microwaves. Rectification can occur in a time dependent system when a directional component, such as a diode, is introduced to the system and the result is that a net DC bias is observed. This possibility would potentially explain the reduced conductance in the previous chapter’s measurements as well as the effect seen above. For the $B = 0$T behavior, the effect of rectification would produce a reduced conductance due to the presence of another bias voltage. In the case of the magnetically split peaks, the rectification would drive the conductance up the split peak. To test the possible rectification, I measured the signal across the 2DEG while varying the magnetic field and the input microwave power. With no compensation bias, the system naturally maintains a 165mV bias due to the inclusion of the current amplifier and any rectification would be observed as a change to this systematic bias. As the power and magnetic field were increased only monotonic behavior persists and a maximum of $-5\mu$V shift was ever observed. This is not nearly large enough to produce any real effect on the system.

The previous results all demonstrate monotonic behaviors and very small effects. The anomalous increase in conductance demonstrated requires a non-monotonic response with the magnetic field.

### 6.3 Amplitude Dependence

With the immediate systematic explanations removed, I continue to pursue an explanation that is maintained within the Kondo model or an increased complexity version of the Kondo model. Many variables can be changed and so the first is the amplitude of the oscillations. Previously shown, increasing the amplitude changes the conductance profile in two distinct manners when there is now magnetic field and when there is high magnetic field. The observed anomalous conductance in the dynamic Kondo magnetoconductance requires very
Figure 6.4: (a) The conductance measured for several powers of a 27.611 GHz signal is plotted as the magnetic field is changed but maintained constant during the measurement. (b) The conductance has been scaled to the $B = 0$ T conductance for each microwave power in (a).

little magnetic field to be activated. In the first experiment to characterize the anomaly, I vary the amplitude of the microwave signals. The intent of the measurements are to observe if the strength of the electron interaction with the photons, as controlled by the amplitude, affect the behavior of the anomaly.

Figure 6.4 demonstrates the conductance measurements as the magnetic field and frequency are held constant and the amplitude is varied. This measurement technique, as stated earlier, eliminates the possibility of induced currents in the system. The conductance increase is clearly observed using this technique for all the powers used. The most important results here is that the peak location in the magnetic field does not vary as the amplitude of
the oscillations are increased for a constant frequency. This feature is extremely important to understanding the nature of the dynamics of the Kondo state. For a constant frequency, any dynamic feature should appear at the same energy set by the photon. To further investigate the combined features of magnetic field and AC oscillations, the rest of the behavior may contain important information. Again, at very large magnetic fields, we observe an increasing conductance with increasing amplitude. This again brings to question the nature of the Kondo effect far from equilibrium. This is continuous support that the equilibrium dynamic nature of the Kondo effect changes when the correlated state is pushed out of equilibrium by means of a magnetic field. Figure 6.4 (b) shows that the nature of the conductance anomaly is affected slower by increasing the amplitude than the rest of the magnetic field regimes. By scaling the magnetoconductance with the $B = 0$T conductance, the peak feature is more prominent as the amplitude is increased. This may suggest that complexities in the dynamic model of the Kondo effect create a stronger correlated state at this small magnetic field which would be less affected by the decoherence mechanisms limiting the conductance.

In the scaling framework of chapter 3, the Kondo temperature increases as the correlations become stronger, and the thermal effects are reduced. For two frequencies, 10 GHz and 27.611 GHz, figure 6.5 shows that the conductance anomaly is constant with amplitude. For 10 GHz extra features are observed.

### 6.4 Frequency Dependence

The conductance anomaly is shown to be frequency dependent in figure 6.5. The lower frequency, 10 GHz, has an increased conductance at a lower magnetic field than the 27.611 GHz data. To investigate the frequency dependence of the anomalous conductance peak, the magnetoconductance peak has been measured for several frequencies on two different SETs and two different thermal cycling of the refrigerator. Measuring this effect in this manner further eliminates two systematic concerns. If the magnetic field affects the transmission
Figure 6.5: A comparison between the 10 GHz features (a) observed in the magnetoconductance and 27.611 GHz features (b) as the power in increased.
Figure 6.6: For both SET 1 (a) and SET 2 (b), the anomalous conductance peak is demonstrated for several frequencies. The anomaly peak location clearly is frequency dependent for both SETs.

of the microwaves on an electrical length change, two thermal cycling of the microwave circuit would not produce the same mechanical fluctuations of the transmission properties. Thermally cycling the 2DEG and using a second SET provides insurance that the effect is not produced by a resonant tunneling through lattice impurities near the 2DEG. The two SETs were tuned to similar Kondo temperatures, $T_K \approx 1K$, in order to vary as few parameters as possible.

The frequency dependence of the anomalous conductance peak in magnetic field is demonstrated for both SETs in figure 6.6. The primary feature can be seen in all the frequencies and is clearly frequency dependent in both cases. The frequency dependence of the peak location is linear and demonstrates a slope of $1/7$ when the magnetic field energy is the Zeeman energy. This scale of energies is very different from the one to one scale that a simple energy exchange would explain. The relationship between the magnetic field and the photon energy can be seen in figure 6.7. For both SETs, the anomalous conductance peak location in magnetic field is plotted as a function of the frequency. For frequencies that show multiple peaks, the peak at the highest magnetic field is recorded. The linear behavior extracts to a slope approximately $1/7$ and intercepts at $B = 0.28T$. The remarkable result is that both
Figure 6.7: For both SETs, the location of the anomalous peak is linearly related to the frequency of the applied signal. Both SETS demonstrate identical linear behavior.

SETs have produced the same relationship between the frequency and the magnetic fields.

6.5 Discussion

No mechanism currently explains the anomalous conductance peak shown in the data. The complexity of the set-up would make any theory very daunting. The possibility of the tunneling rates, $\Gamma$, being functionally dependent on the magnetic field could produce a region of symmetry as $\Gamma_L(B) = \Gamma_R(B)$, though it would not explain why this is not observed in the static behavior. The first chapter describes possible excited orbital states. Theoretically, the presence of excited orbital states can produce a second Kondo enhancement. These measurements may be a new spectroscopy into the orbital energy structure of a quantum dot. However, the magnetic field and the frequency should still have a one to one relationship and the statistical probability of two devices have the same excited orbital energy level is very low. These anomalies readily suggest more experimental and theoretical works to unravel the complexities of the single-electron transistor system.
Chapter 7

Final Remarks

This dissertation has demonstrated the full range of the functionality of the a single-electron transistor. Beginning with the Coulomb blockade regime and traversing the entirety of the spin-1/2 Kondo effect, the data presented covers all the major results and adds much needed investigation to the dynamic regime of the Kondo effect. Universality of the conductance in the Kondo regime shows the true coherent nature of correlated electron systems by focusing on the breaking of correlations as the coherence of the conduction electrons is reduced. Furthermore, the time scales of non-critical coherent phenomena are unique in their own right and are much needed for the next generation of electronics. The Kondo state, as shown throughout this work, is extremely volatile as the parameter space is varied and coherence is very sensitive to these changes. This provides great framework to investigate even the most sensitive systems and further provide the need to develop better and better test equipment and techniques. The sections of this final chapter intends to collate the limitations of the experiments described and the need for future research into the dynamic Kondo effect.

7.1 Providing Better Dynamic Coherence

As the introduction to this dissertation described, coherence is the foundation of many novel systems and the future of electronics. The primary experimental necessity is improving the
quantum coherence of nanoscale devices. The Kondo effect is clearly no different. The thermal and magnetic effects on the Kondo state are very well known and this work has demonstrated great agreement with both experimental and theoretical works before it. Both the Kondo time scale and far from equilibrium effects add unique elements to the concept of the coherence of the Kondo state which have been shown to require better understanding of the single-electron system as a test device for the Kondo effect.

Experimentally, there is one distinct difference between the experiment that observed the Kondo side bands and those which do not. The use of a resonator to filter the oscillations at very low temperature may indeed be required to observe such sensitive effects because there is limited understanding to the effects of an entire spectrum of excitation frequencies. In terms of this experiment, the capacitive coupling technique used, and the bandwidth it demonstrates, does not filter the thermal noise that the DC circuitry does. The decoherence caused by this noise may produced the unexpected results that have been observed in these experiments.

### 7.2 The Future of Dynamic Kondo Research

Since the start of the experiments presented here, many advances have been made in single-electron transistor measurements and coherence systems in general. The use of planar microwave resonators have proven to show agreement between the lock-in amplification measurements shown here and device induced changes to the resonant behavior of the AC circuit. Removing the use of a resonator was the primary focus of this dynamic experiment due to the stability of the SET in several Kondo temperature regimes, but current research in single molecule magnets have employed planar microwave resonators which perform very well at two modes. These modes have been shown to be in the frequency range of interest to most Kondo systems. Designing such a resonator such that the Kondo temperature lies between these two modes may provide the test structure necessary to have adiabatic to dynamic
transition measurements. Any stability at multiple Kondo temperatures would simply be a bonus for fully describing the state. There are thermal advantages to this arrangement as well. In such a structure, the microwave circuit is thermally anchored to a large temperature bath that also anchors the SET. This would reduce the extra heating that may occur due to loss in the coaxial cable and improve the quality of the measurement.
Appendix A

Measurement Setup

In order to perform the low current measurements necessary, this first appendix is dedicated to discussing the DC circuitry set-up used. This will be split into two parts, one for the control over the electrodes and one for the measurement of the conductance. In figure A.1, the entirety of the circuit is shown. Several components, mostly filters, are used in both circuits.

A.1 The Electrode Circuit

To deliver the voltages to the nanoscale electrodes described in the introduction, the voltage is initially supplied by a computer controlled National Instruments voltage card. This card supplied 16 bit precision and these voltages are labeled in figure A.1 as $V_{AVE}$, $V_1$, $V_2$, $V_3$, and $V_4$. These voltages pass through low-pass filters which send the signal from outside of the shielded room to inside of the shielded room. The purpose of these filters are to eliminate the stray AC signals that exist naturally due to the multitude of electrons in the laboratory. The frequency dependence for this filter can be seen in figure A.2. The roll off occurs around 10 kHz. The voltages are combined inside the shielded room, but external to the refrigerator, and the input to output voltage matrix in figure A.3. The average voltage, $V_{AVE}$, contributes the majority of the voltage for each output voltage. This permits a single voltage to control
Figure A.1: As schematic of the measurement set-up used to control the single-electron transistor and to measurement the differential conductance.

the entire closing of the device and the other voltages fine tune the device. From this combining circuit, as we call it the M Box, the voltage are sent into the dilution refrigerator. At the mixing chamber of the refrigerator, the voltages are again filter by absorbing powder which has been measured to filter signals above 1GHz. This filters the thermal noise that is induced by the connection to room temperature. At this point, the signal is connect to the electrodes atop the GaAs/AlGaAs heterostructure. Dot manipulation is performed by inverting the M Box matrix and constantly changing the voltages from the voltage card to produce the desired voltages at the device.

A.2 Source-Drain Circuit

The most important measurement in the experiments presented in this work is the differential conductance. A better measurement of the conductance means a better demonstration of
Figure A.2: The frequency dependence of the first filter sending the voltages into the shielded room.

![Transmission vs Frequency](image)

Figure A.3: The voltage contribution matrix in the M-Box. The $V_{AVE}$ voltage has a major contribution to each of the output voltages. Small cross contributions exist that are controlled by inverting this matrix.

<table>
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<th>V2</th>
<th>V3</th>
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</tbody>
</table>
the Kondo enhanced density of states. The energy scales of the SET require low voltage excitations, which in turn require a very sensitive current measurement. To measure the current on the scale of picoamps, a lock-in amplifier is used. The lock-in amplifier excites the device with a low frequency, low amplitude signal. In these experiments, a $3.16 \mu V_{RMS}$ at 17 Hz signal is used. This signal travels through exterior filter box and is combined through a transformer with the source-drain DC bias voltage. The overall output of the combined signal is a factor of 2043 smaller than the input signals. This signal is a bias voltage with a small oscillation, $V(t) = V_{ds} + V_{exc}(t)$ that is sent to the internal filter and then to the high side of the SET and then brought to ground. The signal-to-noise ratio is enhanced by a current amplifier in series with the source-drain voltage. Exiting the screened room through the filter box, the amplified current signal is inputted into the lock-in amplifier. The lock-in amplifier uses the source signal as a reference frequency for the noisy input current signal. To separate the signal of interest (the 17 Hz response of the SET) from the noisy spectrum, the input current signal and the reference signal are convoluted and pass through a low pass filter. This convolution only has a DC component for the portion of the input current signal that is near the reference frequency ($f_{in} - f_{ref}$). The lock-in amplifier outputs this average of the signal which is in turn used to calculate the differential conductance.
Appendix B

Device Manipulation

Throughout this report many changes have been made to the device due to the long time span necessary to acquire the data and the several different regimes that have been presented. Quite often the tunneling rates and the charging energy are difficult to measure in the Kondo regime and hence measurements in the Coulomb blockade regime need to be made in order to understand the Kondo state. Measurements of the photo-assisted tunneling require vast manipulation to obtain the conditions needed. This appendix covers the characterization used to understand the parameter flow as the voltages are varied.

B.1 Tuning \( \Gamma \)

The tunneling rate, as it has been seen is one of the most vital parameters in both the Coulomb blockade and Kondo regimes. In the Coulomb blockade regime, this is typically directly measured by fitting the thermally broadened Breit-Wigner form for the gate dependent equilibrium conductance. As the confining voltages, \( V_L, V_b, \) and \( V_s, \) are varied, both the symmetry factor and the total tunneling are varied.

\[
Symmetry = \frac{\Gamma_L \Gamma_R}{2\Gamma}; \quad \Gamma = (\Gamma_L + \Gamma_R)/2
\]  

(B.1)
Each gamma is controlled by the two potentials that create it. The principle control variable for the total tunneling rate, $\Gamma$, is the side electrode and it does not influence the individual rates evenly. This requires an intelligent user to constantly interact and interpret the scan by scan results to bring the SET into a near symmetric state with an appropriate tunneling rate.

From the above equation, we notice that the symmetry term is equal to $\Gamma_{L,R}$ when the two tunneling rates are equal. This represents its maximum value and behaves as a single, resonant tunneling junction. Any change from symmetry rapidly reduces the maximum conductance that can be observed. Quite often, this quickly brings the SET to a conductance value that cannot be interpretable from the natural noise of the system. The following is the procedure that I have used to bring the SET as close to symmetric as possible.

- Fit the current state’s conductance profile to check the tunneling rate. Here I use a coefficient to account for the symmetry term.
- Slowly step the side electrode voltage, $V_s$, to vary the tunneling rate. More negative to decrease the rate and more positive to increase the rate.
- Visually observe the change in width of the conductance profile to guarantee correct behavior.
- Choose one of the independent tunneling voltages, $V_t$ or $V_b$, to vary to check the symmetry nature of the SET.
- Increase the tunneling rate to this reservoir by slowly applying less negative voltage on this electrode. If the peak conductance increases (decreases), this tunneling rate is the lower (higher) of the two.
- If the desired total tunneling rate is to be lower, choose the electrode of the higher tunneling rate; else, choose the smaller tunneling rate electrode.
• Either increase or decrease the negative voltage of the chosen electrode to decrease or increase the tunneling rate. Observe the peak conductance as this voltage is changed. As the electrode brings the tunneling rates into symmetry, the conductance will hit a maximum and then begin to decrease.

• Repeat procedure until the desired tunneling rate is achieved.

This process is typically very efficient for the Coulomb blockade regime. Even more, identifying the charging peaks are very easy. The Kondo regime is not as simple because the total tunneling rate affects the Kondo temperature which can lead to misinterpretation of the conductance behavior during the symmetrizing process. When selecting the independent electrode to vary, four possibilities arise.

• Increase the independent tunneling rate; the equilibrium Kondo conductance decreases. The only explanation to this is that the SET is becoming less symmetric.

• Increase the independent tunneling rate; the equilibrium Kondo conductance increase. This outcome is difficult to explain because the Kondo temperature may be increasing as well as the symmetry factor.

• Decrease the independent tunneling rate; the equilibrium Kondo conductance decreases. The Kondo temperature may be decreasing as well as the symmetry factor

• Decrease the independent tunneling rate; the equilibrium Kondo conductance increase. The symmetry factor is increasing.

The end result is that only two of the scenarios can be distinctly interpretable as a change in the symmetry factor. The obvious downside to this process is that the two “good” scenarios still change the Kondo temperature if the voltage is varied by a large amount. In short, any utilization of this process should be followed by a Kondo temperature measurement to fully characterize the device.
B.2 Maintaining Orbital Level

Throughout the measurements, it is desirable to measure the Kondo conductance in a smooth flow with respect to the tunneling rates. These typically give a qualitative, monotonic change with respect to the tunneling rates and hence the Kondo temperature. The main challenge to these scans is maintain a constant orbital energy state so that measurements are made in the center of the Kondo valley. As noted in the body of the text, the change in the orbital energy can be described as the sum of the capacitive effects of each electrode.

\[
\delta \epsilon_{d\alpha} / e = \sum_{i=ds,s,t,b,g} \alpha_i V_i \quad \text{(B.2)}
\]

The process described in the previous section of this appendix does not maintain constant confining potentials and so the changes in the orbital energy need to be taken into account. This is often performed by describing the energy change due to the confining potentials in terms of the gate voltage for zero bias measurements.

\[
V_g = \frac{\alpha_{s,b,t}}{\alpha_g} V_{s,b,t} \quad \text{(B.3)}
\]

The gate voltage can be varied to account for the orbital energy shift when one of the confining potentials is changed. When tuning the symmetry in the Kondo regime, a constant energy level can be achieved by this method and correct interpretation of the conductance change can be made. The ratio of the capacitances can be measured by tuning the device to the Coulomb blockade regime and observing the charging peak shift in gate voltage as each electrode is varied. This shift can be seen in figure B.1. Once these rations are obtained, the orbital energy level can be tuned very well for a full manipulation of the SET.
Figure B.1: The linear behavior of the Coulomb blockade charging peak location in the gate voltage as each electrode is varied. The negative slope is due to the necessity of lowering the dot energy via the gate voltage to account for the increase in dot energy due to the electrode.
Appendix C

Experimental Application of Microwaves

The purpose of this experiment is to apply a wide range of frequencies to the source-drain voltage in order to test the variability of the Kondo dynamics. The ability to fix the Kondo temperature and sweep the frequency through the characteristic timescale described above is of particular interest. This will permit observation of the change from adiabatic behavior to dynamic behavior.

In order to test the dynamics of the Kondo state in a SET frequencies in the microwave range, 10 GHz, are necessary. These frequencies are much too large to be used in the DC circuitry and so a secondary circuit is required to deliver these signals. This circuit consisted of several lengths of UT-85 coaxial cable. The majority of the cable used is a copper/silver/aluminum alloy delivering signal to the 1K level of the dilution refrigerator. High attenuating stainless steel coaxial cable connects the 1K level to the mixing chamber where temperatures are the lowest, 10 mK. This high attenuation created a very poor thermal connection between these two area of the refrigerator which is necessary to maintain the cooling power of the mixing chamber. From the mixing chamber to the sample, pure copper coaxial cable is used to prevent magnetization effects in the cable when high magnetic fields
are applied. The coaxial cable is connected to the sample by capacitively coupling the signal conductor to the DC pin on the chip socket corresponding to the source-drain bias. This capacitive coupling prevents DC current to leak through the AC circuit and the filters in the DC circuit prevent the AC signal from traveling backwards up the DC path. This acts as a natural bias-T delivering a combined AC and DC signal to the device. Based on reflection theory of microwaves, reflection = \( \frac{Z_{\text{in}} - Z_{\text{load}}}{Z_{\text{in}} + Z_{\text{load}}} \), there exists a low frequency cutoff of the coupling to the device. Also, the functionality of the coaxial cable and the connectors, there is a high frequency cutoff. These yield a broadband range of 100MHz-35GHz of potential functioning frequencies.

The last circuit component of consideration is the thermal disconnect between the outer ground conductor and the inner signal conductor due to the poor thermal conductivity of the Teflon dielectric. To assist cooling the inner conductor, several thermal heat sinks were constructed to place at various functioning levels of the dilution refrigerator, the 1 Kelvin condenser, the still (300 mK), and the mixing chamber (10 mK). These heat sinks are microstrip transmission lines that consist of a surface signal line and a ground line below the dielectric. The signal line is a thin layer of gold to reduce the resistive loss and the dielectric is sapphire, chosen for its good thermal conductivity and poor electrical conductivity at low temperatures. This permits the signal line to be cooled by the ground line connected to the refrigerator. The coaxial cables are connected via copper clamps and conductive epoxy securing contact between the center conductor of the coaxial cable to the microstrip signal line. Using a vector network analyzer (VNA), these connectors transmitted an average of 0.7V_{\text{input}}, or 3 dB of attenuation. Imperfect boundary conditions at the coax/microstrip interface leads to small oscillations of the transmission of the order of 0.1V_{\text{input}}. The unfortunate effect of these heat sinks is that the low temperature behavior is unknown leading to the uncertainty in the transmitted signal as a function of frequency. At room temperature, the total loss of the system is measured to be 0.4% V_{\text{input}}, or 47dB of attenuation. If we take the absorption of a photon with one electron in the energy
range necessary, 40-100\(\mu\text{eV}\), this corresponds to a necessary attenuation of -74dB. The room temperature measurements suggest that the signal will need to be reduced in order to maintain a low signal. However, the true attenuation necessary is determined on a frequency by frequency basis described next.

C.1 Determining Usable Frequencies

As described above, not all frequencies within the frequency range are usable frequencies. The main determination of the usability of a frequency is whether or not the device responds to the applied oscillations. In all regimes the device should be amplitude dependent and so any large variation in the delivered power should produce an observable change in the conductance. In principle, no frequency has a zero transmission amplitude and a large enough power will demonstrate behavior. This, however, is often impractical due to unfavorable heating in the refrigerator.

For any thermal cycle of the sample, it is very possible that the various contractions and torques on the microwave transmission lines will produce a change in the transmission properties of the microwave circuit. It is very important that a characterization technique is used to understand which frequencies can be used for measurements and which cannot. This following demonstrates the technique I have used to determine the frequencies that I present.

- Tune the SET to a desirable setting: Gate voltage sweep for Coulomb blockade and source-drain sweep for Kondo regime.

- Set the sweep to measure the conductance as the frequency is varied at a fixed power.

- Frequencies that show strong response get recorded.

- For each frequency of interest, fix the frequency and increase in large increments of power to observe a full reduction of the static phenomenon.
Typically, the several frequencies within 1GHz span are found throughout the 1-30 GHz range. Frequencies below 1 GHz are more difficult to obtain and, in my measurement, the 140 MHz was found over a very long search for low frequencies. Also, by the 35 GHz range, the number of frequencies are few.
Bibliography


