Chapter 6. Single-Electron Spectroscopy

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6.1 Goals and Objectives

Several years ago, we developed a technique known as single-electron capacitance spectroscopy (SECS). We showed that a very sensitive electrometer could detect the motion of a single remote electron inside of a solid, and capacitance-voltage characteristics of very small structures could be performed. For small objects such as quantum dots, a spectrum of electron additions as a function of a gate bias can be clearly observed. We have produced quantum dots containing as few as one electron and up to 60 electrons. Additionally, electrons can be observed to enter different types of traps. Electron spectra for single impurity atoms have been determined this way.

SECS allows us to observe and manipulate single electrons to a previously unimagined degree. The main aim of our work is to advance and utilize these methods. All our work utilizes our specialized techniques for measuring very small amounts of electrical charge. The effort is divided into four projects which, while distinct, use considerably overlapping methods.

Our most significant advance in SECS will lie in our ability to perform the technique in a scanning fashion. By scanning a fantastically sensitive charge sensor, we are attempting to do something that is well beyond the capabilities of ordinary scanning tunneling microscopy: we will be able to "look" underneath surfaces. Further development of the method will allow scanning tunneling microscopy and spectroscopy of insulating surfaces.

A second key advance lies in our time domain SECS technique. We have developed a method which allows us to record, in the time domain, the motion of charge within a sample. This technique allows us to determine the nonlinear response of a sample to which we cannot make electrical contacts. So far, we have used the method to study the characteristics of electron tunneling between a two-dimensional electron gas and a three-dimensional electron gas. This measurement was not possible before. Our plan is to eventually apply this technique to the study of single traps containing single electrons.

Presently, SECS is a difficult technique to employ. We have used HEMT transistors to measure very small amounts of charge. Typically, electron motion in our samples results in a small fraction of one electron charge appearing at the gate of this transistor. There is just barely enough sensitivity to detect this charge, and large amounts of signal averaging are required. We plan to change this by using single-electron transistors (SETs). SETs will greatly improve our capacitance spectroscopy measurements. We expect signal to noise levels to increase by a factor of 100 compared to our measurements using field effect transistors. We now have functioning aluminum-aluminum oxide SETs.

Finally, we are extending the charge sensitivity that we have developed for the SECS technique to study other systems. Until our recent experiments, sensitive charge sensors had never been used to study transport in semiconductors. There is a wealth of experiments for which they may be employed to greatly enhance sensitivity. HEMT transistors and FETs will allow us to measure unprecedentedly small conductances in objects such as lateral quantum dots, point contacts, and tunnel junctions. They also allow study of the quantum Hall effect in a fundamentally new way. We have developed a capacitance experiment which we feel gives the clearest picture to date regarding the nature of edge states in the quantum Hall effect.

6.2 High-Resolution Scanning Capacitance Spectroscopy of Semiconductor Structures

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The first SECS measurements in a disordered 1 micron diameter quantum dot revealed that there exists a fascinating diversity of quantum states in this system. The magnetic field evolution of the spectra indicated states in which single electrons were trapped on (1) potential fluctuations, (2) single silicon impurities, and (3) strange sites which bound two electrons at precisely the same energy in spite of the Coulomb repulsion between electrons (Negative-U centers). The latter states have been of significant theoretical interest recently. Despite all of the information in the spectra, one gnawing question persists: What do the states "look" like? This kind of question can only be answered using a scanning probe technique. This probe will also help to answer critical questions related to localization in the quantum Hall effect, the structure of quantum Hall edge states, and the nature of dangling bonds in the Si-SiO$_2$ interface.

We have constructed a scanning tunneling microscope (STM), presently functioning at room temperature, which we are preparing to cool down to 300 mK. The room temperature STM works in a mode which is entirely different from a typical scanning tunneling microscope. Rather than detecting a current from electron tunneling between a scanning tip and the sample, our STM is sensitive to the induced charge in the vicinity of the tip. This means that the STM can be used to study *insulating* surfaces and to do spectroscopy on *buried* traps and surfaces. We are now taking our first images using the new method. It is our hope that this method will vastly expand the utility of STM. Shown in figure 1 is a scanning tunneling image (a) and a scanning capacitance image (b) taken with our microscope. Remarkably, the scanning capacitance image was taken with the tip at a distance of 700 Å from the sample surface.
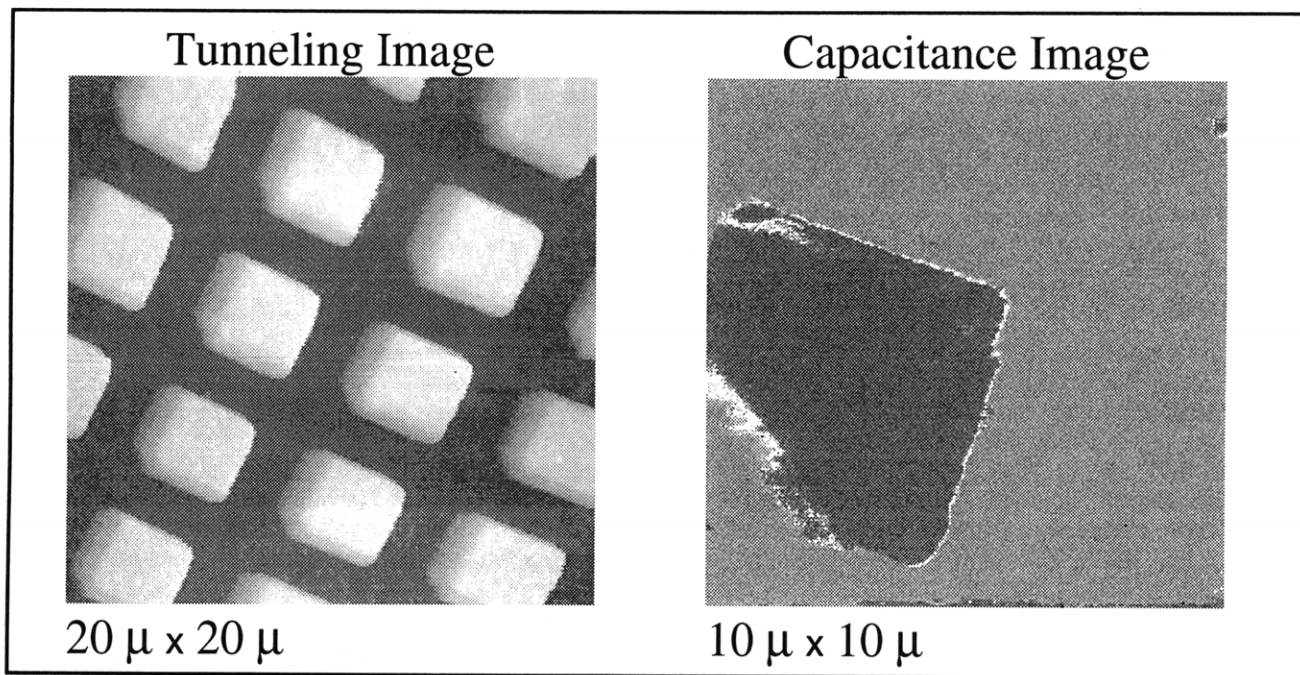

Figure 1. Scanned images of a microfabricated Au sample. The tunneling image of the surface topography shows an array squares of height 480 Å. It was acquired with the use of feedback, so that the tip maintained a 5 Å distance from the surface. Conversely, the capacitance image was acquired without feedback using a tip-sample distance of roughly 500 Å. The contrast reflects both the geometrical capacitance between the tip and surface and the impedance of an individual square with respect to the substrate.

6.3 Single Electron Transistors for Studying Quantum Dots and other Nanoscale Objects

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Project Staff

David Berman, Professor Raymond C. Ashoori, Professor Henry I. Smith

The major research thrust of our group is the study of the motion of small amounts of charge. In our experiments in capacitance spectroscopy, charges from a quantum dot sample are coupled to an amplifier. It is extremely important that the amplifier is located as close to the sample as possible to reduce the capacitance of the gate electrode, since the signal at the amplifier is inversely proportional to the gate electrode capacitance. This capacitance includes the coupling capacitance to the quantum dot, the gate capacitance of the transistor (the amplifier is usually a HEMT transistor), and all stray capacitance to the rest of the world. We are using the single-electron transistor (SET) as an ultra-sensitive electrometer in this application because it has a much smaller gate capacitance than the HEMT transistor. Also the SET can be fabricated in very close proximity to the quantum dot, thus reducing stray capacitance.

To further increase the sensitivity of our measurement, we will couple our signals into the central island of a SET with a gate. We will bias this gate separately with a specially designed low capacitance resistive element. In previous experiments with SET electrometers, the central island of the SET was directly coupled to the device being measured. By capacitively coupling a gate to the quantum dot, we hope to gain a threefold advantage. First, the size of the Coulomb blockade region is independent of the length of the gate wire and the size of the quantum dot, since it is capacitively coupled. Second, the potential on the coupling electrode is not affected by the SET itself, since the tunnel junctions are decoupled from this electrode. Third, we have greater control over the biasing of the amplifier circuit. We can bias the gate electrode with a resistor, as we have done in our circuits employing HEMT transistors. This would give us the ability to bias the gate of the SET at the optimum position. In addition, since we have a back contact to the quantum dot, we would also be able to set the voltage across the quantum dot exactly.

Our single-electron transistors are composed of aluminum. Tunnel junctions, which establish most of the operational characteristics of the SET are formed by a small overlap of two layers of metal with a thin dielectric layer in between. This dielectric layer is formed by oxidizing the surface of the bottom layer of metal and then introducing the top layer. The thickness of this oxide layer is only a few atomic spacings, so it is very important to ensure that it is very uniform and pure.

The tunnel junctions have a capacitance mostly determined by the area of the metal overlap. In order for SET to operate properly, this overlap area must be less than 0.01 μm^2. Electron beam lithography is used to pattern our devices. It is performed on a JEOL 6400 scanning electron microscope which we modified to be able to do lithography. The Ashoori group installed a pattern generation system on the SEM to control and scan the beam of the SEM. Using this system, we have been successful in patterning 60 nm features in electron-beam resist.

We form the metal layers in an e-beam metal evaporator. The high degree of directionality of the evaporated metal gives us more control over the patterning of the metal. Both layers of metal of the transistor are evaporated in one vacuum cycle, to prevent the bottom layer, which must be oxidized to form the tunnel barrier, from being exposed to oxygen-rich air, which would prevent us from controlling the thickness of the barrier. Double-angle evaporation, or shadow evaporation, is the method which we use to fabricate both layers so that the small overlap between them forms the tunnel junctions. Figure 2 shows a schematic of the shadow evaporation technique.

The electron-beam resist which we use in the shadow evaporation method plays a crucial role in the success of the process. In addition to the usual planar pattern, we must also engineer the profile of the patterned resist. As depicted in figure 1, the resist consists of two layers. The lower layer, MAA copolymer, is more sensitive to electron-beam bombardment than the top layer, PMMA. Thus, during development, MAA is dissolved more rapidly than PMMA. The result is that we get a large degree of undercut in the pattern profile. The result of this undercut is that we can get a suspended bridge in the PMMA by placing two lines in close proximity. In fact, we use a two-step development process to increase the extent of undercut. First, we develop the sample in a solution of methyl isobutyl ketone (MIBK) and isopropyl alcohol, which develops exposed regions of PMMA and MAA.

Then, we develop in a solution of poly glycol methyl ether acetate (PGMEA) and ethanol, which develops only the MAA copolymer layer. This technique allows us to increase the degree of undercut without affecting the linewidth. This process is shown in figure 3.

After its development, the sample is placed in a metal evaporator on a tilting stage. In order to form tunnel junctions, we perform two separate evaporations at two different angles, as shown in figure 4. The oxide barrier is formed by introducing a controlled amount of oxygen gas into the chamber after the first metal evaporation.

We have recently produced our first SETs for measuring single quantum dots on a GaAs sample. Our first measurements using these devices are presently underway.
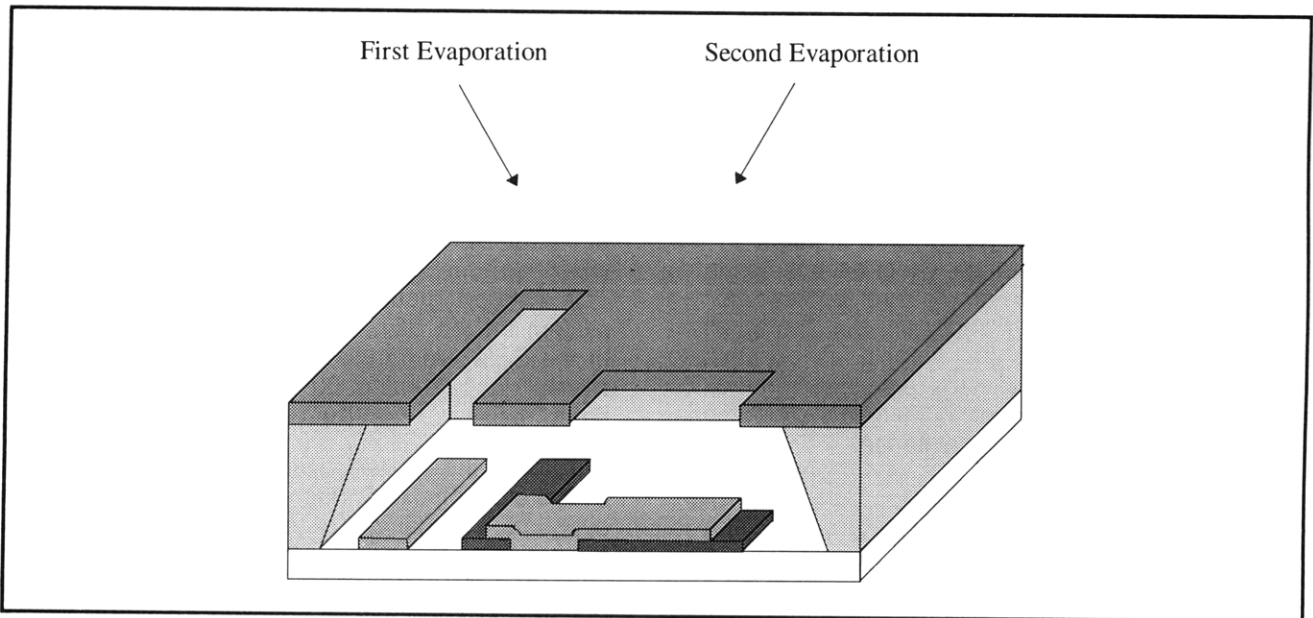

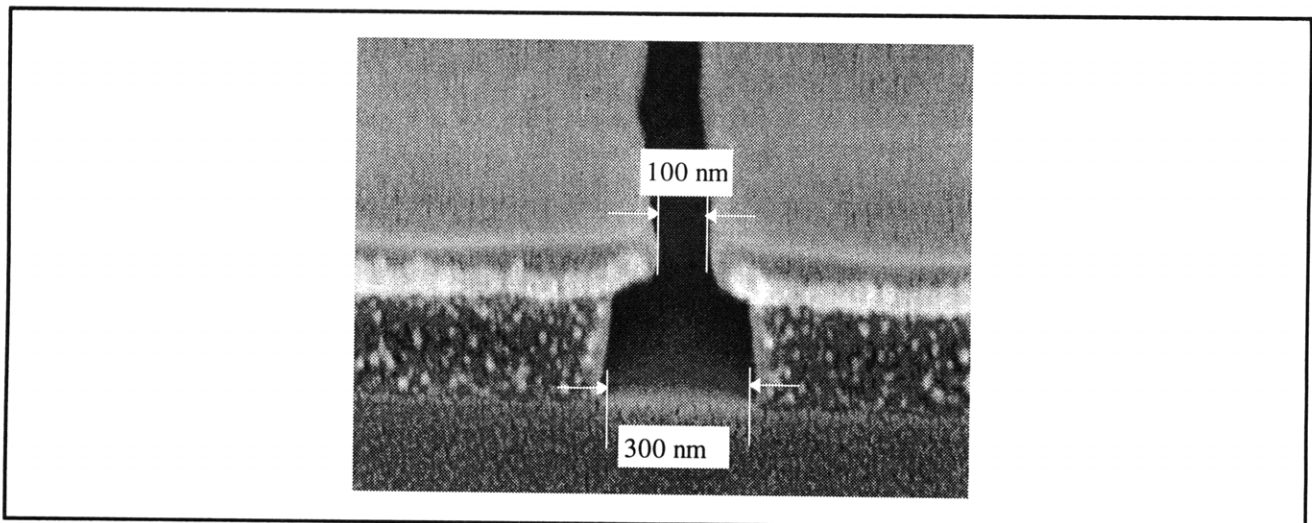**Figure 2.** Double-angle evaporation schematic.

Figure 3. A 100 nm line drawn in PMMA and copolymer resists using the PGMEA process.
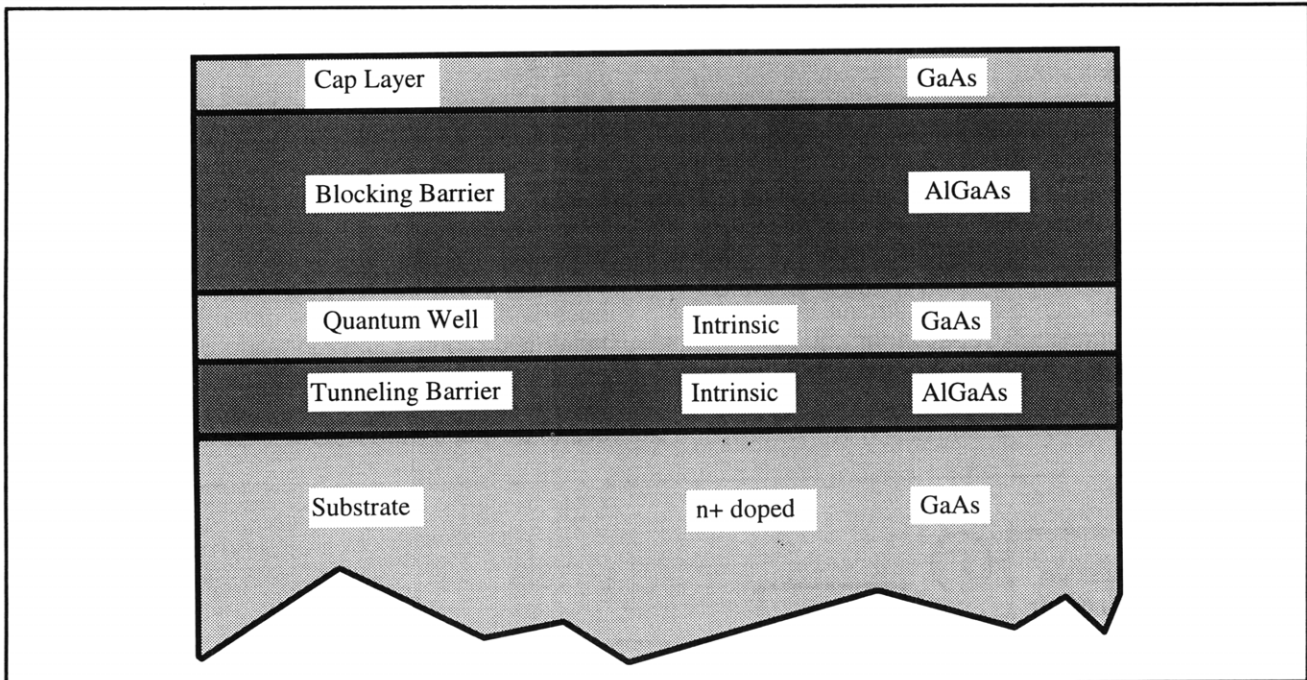
Figure 4. Structure for magnetocapacitance measurements of the 2DEG.

6.4 Time Domain Capacitance Spectroscopy

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Ho Bun Chan, Professor Raymond C. Ashoori

We have developed a method which allows us to record, in the time domain, the motion of charge within a sample. This technique permits the determination of the nonlinear response of a sample to which we cannot make electrical contacts. So far, we have used the method to study the characteristics of electron tunneling between a two-dimensional and a three-dimensional electron gas. This measurement was not possible previously. We are now working to apply this technique to the study of single traps containing single electrons.

6.4.1 Motivation

One of the chief limitations of SECS measurements have been that they only probe the ground electronic states of quantum dots, impurities, and other charge traps. We have devised a method for using SECS to probe excited states. We expect that this will be a very powerful probe, capable of determining excitation spectra for single traps of many sorts. We decided to first try the technique on samples which would create much larger signals than would quantum dots. Our first experiments using this new technique have been carried out on a two-dimensional electron gas sample. We will describe these measurements first and then discuss how the measurement can be extended to be used on quantum dot samples.

We have been using magnetocapacitance measurements to study zero bias tunneling into a two-dimensional electron gas (2DEG). A magnetic field induced energy gap is observed in the tunneling spectrum of the 2DEG at low temperatures. This suppression of tunneling is believed to be a many body phenomena and is interpreted as a manifestation of a Coulomb gap.

Our experiment is performed on a GaAs tunneling capacitor (figure 4) grown by molecular beam epitaxy. The bottom electrode consists of an n⁺ doped GaAs region. Above this an AlGaAs tunnel barrier is grown followed by a GaAs quantum well defining the 2DEG. After the quantum well, there is a thick AlGaAs blocking barrier which prevents

electrical conduction between the quantum well and the top electrode. A DC bias is applied at the top electrode to control the number density of electrons in the quantum well. Sinusoidal excitations are used to measure the equilibrium tunneling conductivity. The Fermi energies on both sides of the tunnel barrier are kept within kT of each other. Time domain experiments yield a much clearer picture of the tunneling than our previous frequency domain measurements. The chief reason is that, using the time domain, we can easily determine the I-V characteristic for tunneling. Capacitance measurements are only useful for the tunneling resistance for V=0.

In this new experiment, we start with the 2DEG in equilibrium with the n⁺ substrate. A sudden voltage step is then applied to create an offset in the Fermi energies between the two sides of the tunnel barrier (figure 5). As the electrons tunnel, this offset in Fermi energies equilibrate to zero. The amount of charge induced on the top gate is recorded in real time as electrons tunnel into the 2DEG. If the tunneling conductance is independent of the excitation amplitude, a familiar RC decay will be observed. The situation is different when the magnetic field reduces the tunneling density of states at the Fermi energy of the 2DEG. The tunneling conductance becomes voltage dependent and the signal deviates from a pure exponential decay.
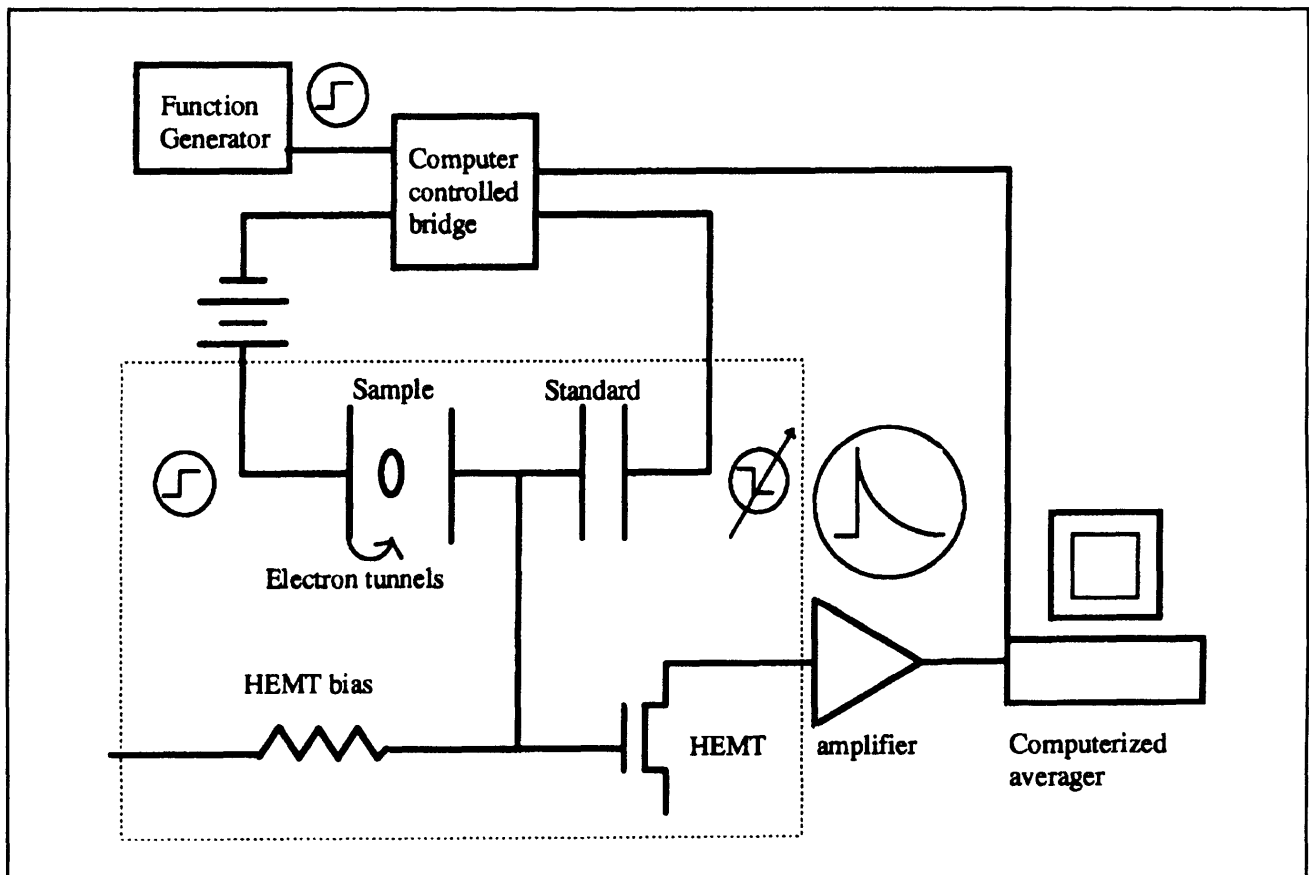

Figure 5. Schematic of the time domain experiment. Presently, the sample is a "tunnel capacitor" containing a 2DEG. In the future, it will be a quantum dot. Components enclosed in box sit inside cryostat.

This new technique of time-resolved capacitance measurement has a number of advantages compared to the frequency domain technique. In the frequency domain measurement, electrons tunnel into and out of the 2DEG with a frequency of the sinusoidal excitation. It is not possible to distinguish between tunneling of electrons into the 2DEG from the n⁺ substrate or from the 2DEG into the n⁺ substrate. In the time domain experiment, tunneling in both directions can be measured separately by

using steps of opposite polarity. Preliminary data indicate that an asymmetry of the tunnel gap does exist between electrons tunneling into and out of the 2DEG. The degree of asymmetry seems to depend strongly on the filling factor in the 2DEG. Even in the absence of a magnetic field, we have been able to detect the effect of momentum conservation on electrons tunneling into and out of the 2DEG. Consider the case where we increase the density of electrons in the 2DEG so that the two-

dimensional Fermi disk has a larger diameter than the three-dimensional Fermi sphere in the n⁺ substrate. The tunneling rate for electrons entering the 2DEG will be suppressed because all the states which satisfy momentum conversation are already filled. On the other hand, such suppression does not exist for electrons tunneling out of the 2DEG since the states in the 3D substrate are empty. Our preliminary data demonstrate clearly the effect of this 2D-3D momentum conservation.

This voltage stepping technique has other advantages over the sinusoidal technique. In the frequency domain experiment, small excitations are used so that the system is very close to equilibrium. Results from larger excitations are difficult to interpret because of the oscillating energy configuration in the sample. In the time domain experiment, the excitation is in the form of a voltage step. The energy configuration of the sample immediately after the voltage step can easily be determined. We can easily and precisely map out the excitation dependence of electron tunneling, a feat that is impossible using sinusoidal excitations. We will directly extend this technique for studying excited states in quantum dots using the method depicted in figure 6.
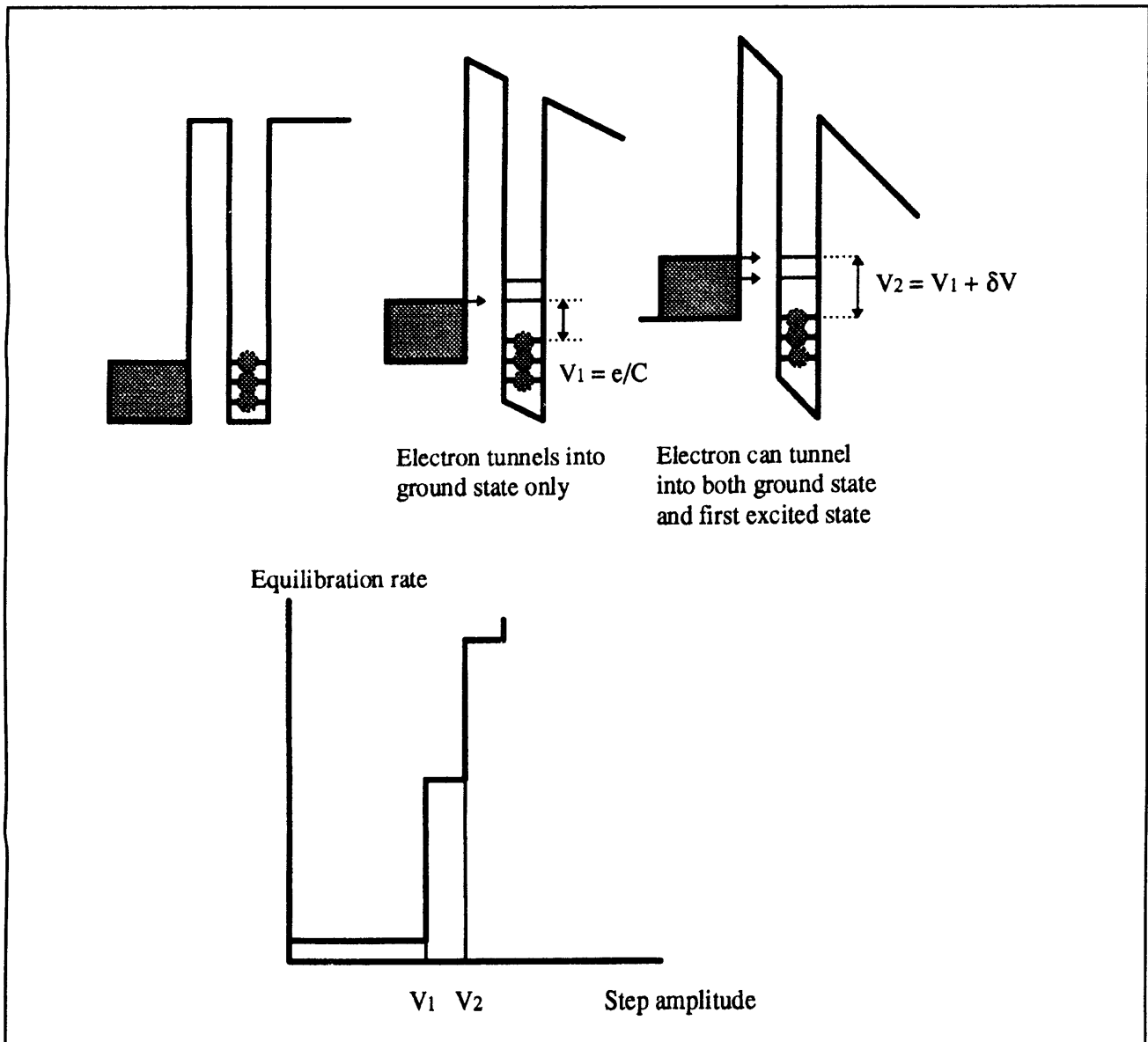

Figure 6. (top) Schematic of the use of the stepping technique to study quantum dots. (bottom) The expected change in the equilibration rate as tunneling is permitted for higher lying states of the dot.

6.5 Use of High Sensitivity Electrometry to Study the Quantum Hall Effect

Sponsor

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We have developed a new technique for direct investigation of the resistance of incompressible strips adjoining edge channels under quantum Hall effect conditions. We measure the frequency dependence of the capacitance of a small gate covering the edge and part of a 2DEG from 100 Hz to 20 MHz, employing an ac cryogenic bridge. Geometrical capacitances are 0.01-1 pF, depending on the size of the gate. Varying the gate bias changes the character of the electron gas underneath the gate. While the portion of the electron gas nearest to the edge remains compressible (edge channel), the remainder evolves from compressible, to incompressible, and then to compressible encircled by incompressible as the electron concentration is enhanced by the gate bias.

The latter situation of a compressible puddle surrounded by an incompressible strip (IS) is realized when $\nu_g > \nu_s \geq \nu_b$. Here ν_g and ν_b are filling factors under the gate and in the bulk; ν_s is the integer filling factor of the IS. To charge the puddle, electrons must tunnel from the edge channel through the IS. Typical capacitance versus gate voltage traces, measured on a sample with gate dimensions 32 μm (along the edge) × 5 μm (perpendicular) at 100 kHz, are shown in the figure (curves for different fields are shifted vertically). Peaks seen in the capacitance correspond to conductance resonances for electrons tunneling through the edge IS.

There is a striking similarity of the resonance spectrum observed for samples with vastly different gate
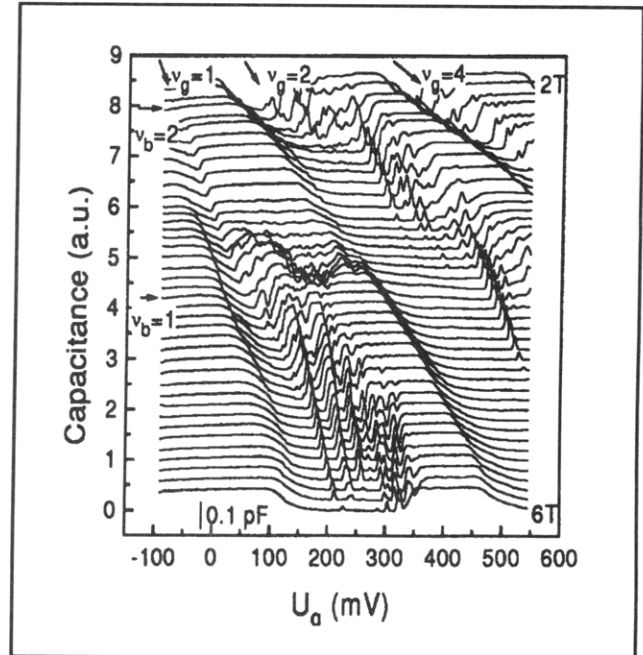

Figure 7. Capacitance versus gate bias scans for a small gate over the edge of a two-dimensional electron gas. The different scans are for different magnetic field valves varying from 2-6 Tesla. Note the resonances.

lengths along the edge (1-170 μm). The resonances are no broader and appear no more frequently in samples with longer gates. This fact, as well as the character of the temperature dependence of the peaks, excludes impurity-assisted tunneling as the origin of the resonances.

We speculate that the resonances arise from abrupt transitions in structure in or near edge incompressible strips. The sharpness of the resonances in the case of long gates indicates an unusual ubiquity of the transitions, despite the expected inhomogeneity of the sample edge. There appears to be an underlying phenomenon which forces large regions of the edge to behave as a single structure.

6.6 Publications

Ashoori, R.C. "A New Class of Resonances at the Edge of the Two Dimensional Electron Gas." Submitted to *Phys. Rev. Lett.*

Ashoori, R.C. "Electrons in Artificial Atoms." *Nature* 379: 413-419 (1996).