

The principle focus of research in our laboratory lies in the study of interacting electronic systems in low dimensional semiconductor structures. Systems in which electrons exist purely in two or one dimensions and even small boxes (quantum dots) containing as few as one electron can now be produced with relative ease. While simple quantum mechanical calculations determine the motion of a single electron in such a confining structures, it is far from simple to understand the behavior of many trapped electrons. Not only do the electrons repel one another, they are indistinguishable. This fact, along with the principle that only one electron can exist in any quantum mechanical orbit, produces unusual and sometimes counterintuitive correlations in the motions of electrons.

We use extremely sensitive methods to detect minuscule amounts of electrical charge inside materials. This capability has permitted us to perform some rather unique and fundamental measurements on low dimensional electronic systems.

Electrons Don't Repel? Probing Electron Localization in Quantum Dots

At low temperatures, a small metallic system which is poorly coupled to its surroundings must contain an integer number of electrons. Due to the repulsive Coulomb potential of electrons already in the system, the energy required to add an electron to the system increases by a fixed amount with each electron transfer to the system. An external gate electrode capacitively coupled to the system through a capacitance C_g can be used to cause the electron transfer. Additions of *single* electrons to the system occur periodically in gate voltage that draws electrons into the system, and the period is e/C_g (with e as the electron charge). This phenomenon is known as Coulomb blockade, and it is one of the most fundamental and robust concepts in the physics of small electronic systems.

Several years ago, we observed an extraordinary violation of Coulomb blockade. We found that it can cost no more energy to add a second electron to a system of electrons than it did for a first electron. Somehow the energy of repulsion between the two electrons appears to disappear completely. This occurs only for systems of electrons at low electron densities. Through a series of experiments we have found that the effect only occurs when electrons are, in some way, localized into separate regions within the system.

Figure 1a displays the typical arrangement in our experiments. A system of confined electrons (known as a "quantum dot") is positioned between two capacitor plates. It is close enough to one plate so that electrons tunnel between the dot and the plate and far enough from the other plate to prevent all tunneling. By adjusting the voltage on the top capacitor plate (or "gate"), we can draw electrons into the quantum dot (more positive top plate voltage) from the bottom plate or push them back into the bottom plate (more negative top plate voltage). When an electron tunnels into the dot, it induces a minuscule image charge on the top plate. We have learned to detect this charge, and we can thereby measure the gate voltages at which single electrons enter the dot.

Figure 1b shows the actual physical construction of the quantum dot. It exists inside of a wafer of in a semiconducting material and in a layer of gallium arsenide sandwiched between layers of insulating aluminum gallium arsenide. This layer is so thin that at low temperatures, only the lowest quantum level of this "quantum well" is occupied by electrons. With no further fabrication, such a sample will contain a two-dimensional (2D) electron gas in the gallium arsenide layer. With further processing involving etching a small pillar on the surface of the structure, we laterally confine electrons in the layer into a small disk (with radius ranging from 0.1 to 1.0 microns).

Figure 1c shows a typical charging spectrum of a dot with electrons confined to a ~0.2 micron diameter disk. The first peak on the left arises from the first electron the quantum dot. Notice that subsequent peaks appear nearly periodically in top plate voltage, although the period decreases as the electron number increases. As expected, the peaks avoid each other. It takes more gate voltage (energy) to add subsequent electrons because electrons already in the dot repel them. If the dot were of constant size and made from a perfect metal (with infinite density of states), one would expect electrons to be added with perfect periodicity. Our dots are confined laterally by a soft confinement potential. As electrons are added to them, they expand laterally. As the dot grows in size, charges of added electrons are spread over a larger area, and the amount by which they repel subsequent electrons therefore diminishes. This effect is responsible for the observed decrease of peak spacings.

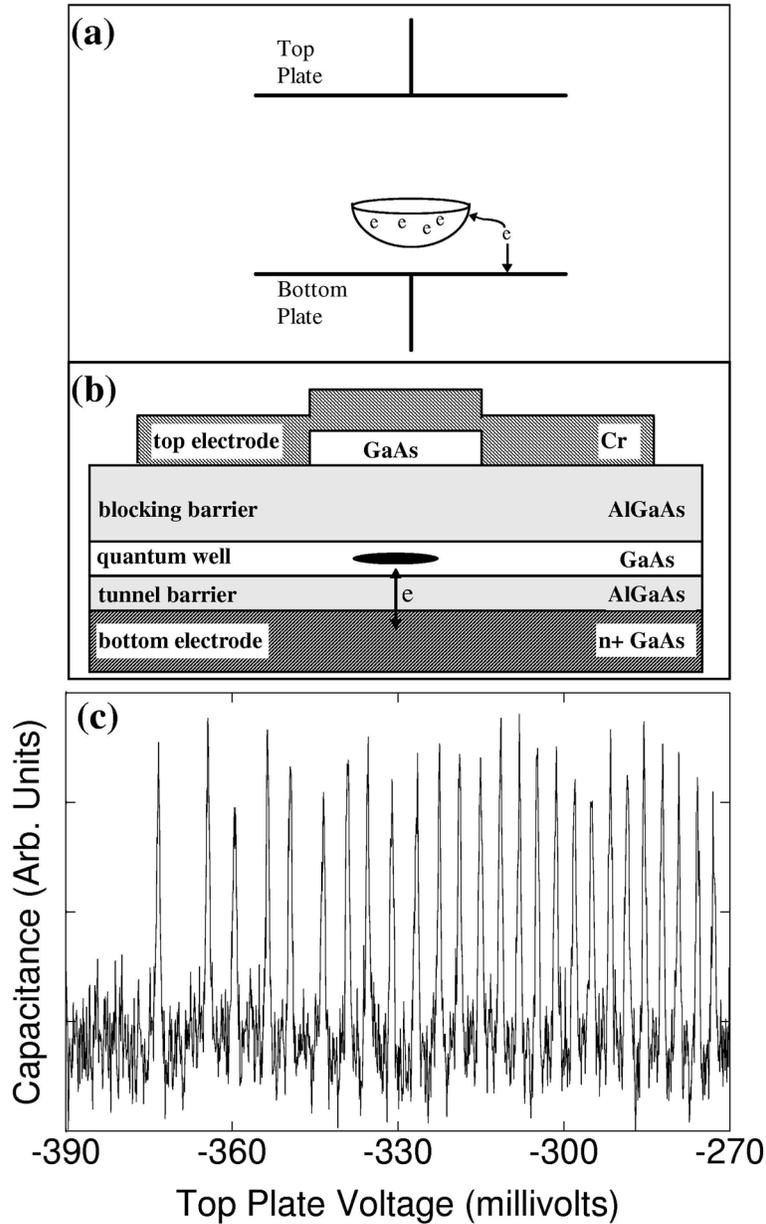


Fig. 1 a) Schematic diagram of a quantum dot located between two capacitor plates. The quantum dot is actually two-dimensional; the bowl-like shape represents the force tending to move electrons to the center of the quantum dot. b) Diagram of the sample used in single-electron capacitance experiments in a crystal grown using molecular-beam-epitaxy. The quantum dot is the black disk in the quantum well. c) Capacitance (charging) of the sample containing the artificial atom as a function of the top plate (gate) voltage. The first peak on the left represents the first electron entering the quantum dot.

Upon building dots of larger diameter, we found a profound surprise. Electrons sometimes enter such dots in pairs and even in bunches of three or more. Addition of subsequent electrons cost no more energy than the addition of prior ones.

The largest dots (1.0 micron pillar diameter) showed many randomly appearing pairs and bunches, while smaller dots showed no pairs. We then performed a systematic study of different dots of different sizes. For dots made from pillars of diameters between 0.4-0.7 microns, we found another surprise. The spectra did indeed display pairs, but the pairing effect itself occurred periodically. Figure 2 shows data from a dot created under a 0.5 micron pillar. The first peak on the left corresponds to about

the 30th electron entering the dot. Notice that some peaks appear in the spectrum which are higher than others. These arise from 2 electrons entering the dot at the same gate voltage. Moving to higher electron numbers, the spectrum shows similar periodic electron additions as in Fig. 1. However, every 4th and every 5th electron enter the dot as a pair. This behavior continues over about ten (five electron) periods over a total range of about 50 additions. For yet higher electron numbers, the pairing effect disappears and the spectrum appears as ordinary Coulomb blockade with nearly equal spacings between all electron additions. This transition from a pairing regime to Coulomb blockade occurs at an electron density of 1.1×10^{11} electrons/cm². All dots with diameters larger than 0.4 microns display the same transition (whether from periodically or randomly occurring pairs) to Coulomb blockade.

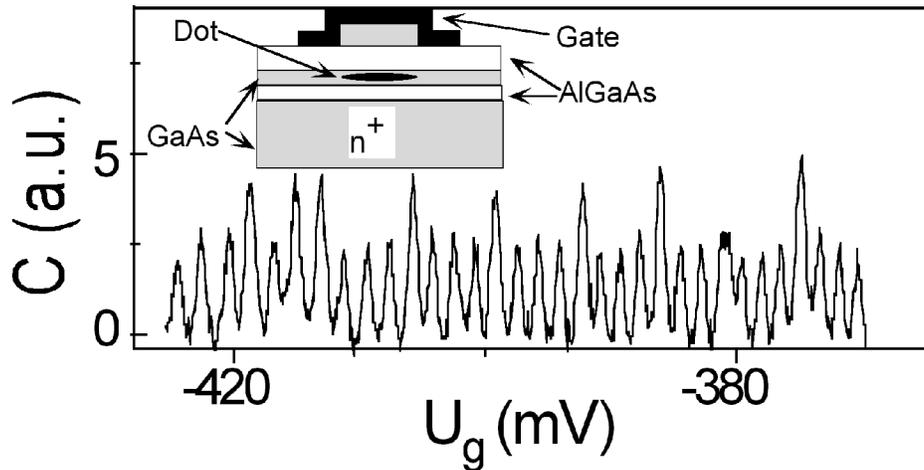


Fig. 2. Quantum dot capacitance as a function of gate voltage. Each peak denotes the appearance of an electron in the dot. Double height peaks indicate the addition of two electrons. $T = 0.3$ K. Inset: Schematic of the dot.

What produces the pairs, and why do they occur periodically in some cases? While we still do not have a clear answer to the first question, we can answer the second one. Prof. Leonid Levitov performed calculations for classical 2D electrons in a cylindrical confining potential. He found that classical electrons arrange themselves into a hexagonal lattice. The simulations show that electrons at the edge (the outermost row) of the dot are isolated from interior by dislocations in the crystal. In these calculations, the quantum dot breaks up into two separate electron "puddles": a core of electrons and a ring surrounding the core. Prof. Levitov then calculated the sequence for adding electrons to these two puddles. His calculations showed that 4 electrons enter the core puddle in succession and then a 5th electron enters the surrounding ring. As in the experiment, this 5-electron periodicity continues for many periods.

There exists a substantial distinction between the classical calculations of Levitov and the quantum mechanical system of electrons in our quantum dots. Nonetheless, we should expect a correspondence when the electron density is extremely low. The important dimensionless parameter determining whether or not classical calculations apply is r_s . r_s is the ratio of the inter-electron spacing to the Bohr radius (in GaAs this is about 100 Å), and r_s therefore decreases as the density of electrons increases. It turns out that r_s is also the ratio of the Coulomb energy of electrons to their kinetic energy. When r_s is large, electrons arrange themselves in a crystal because the kinetic energy cost of for localizing electrons is small compared to the Coulomb energy gained. Detailed calculations predict that electrons crystallize in two-dimensions when r_s is larger than 37. However, our experiments show the periodic pairing effect at densities so high that $r_s=1$. Do the classical calculations still contain some glimmer of validity even though the measurements lie well outside the range where they are ordinarily applied?

We tested Levitov's hypothesis by performing an experiment to test whether every 5th electron was indeed added to the periphery of the dot while the other 4 were added to the dot's core. In our laboratory, Nikolai Zhitenev and Misha Brodsky fabricated a sample that contained an additional that surrounded the quantum dot. A schematic of this sample is shown in Fig. 3a. The ring affects most strongly electrons that reside near the periphery of the dot.

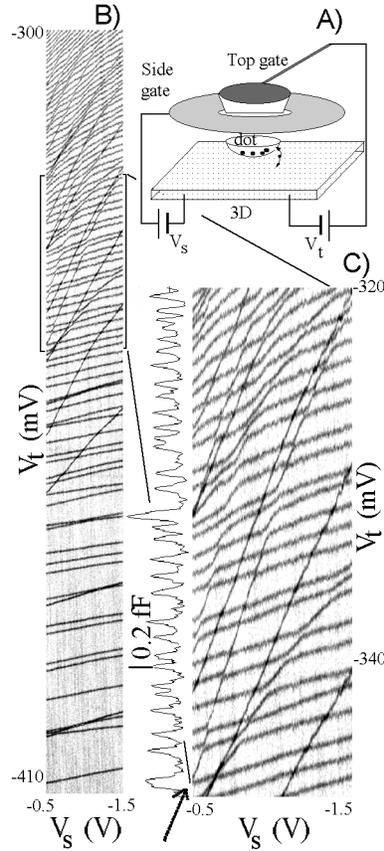


Fig.3. a) Schematic of quantum dot structure with side gate and scheme for application of bias voltages. b) Greyscale panel of measured capacitance with dark lines denoting capacitance peaks. Black color denotes the capacitance peaks. Each peak corresponds to an electron addition to the dot. Top-gate voltage scale can be converted to the addition energy scale by multiplying by 0.5 (lever-arm) Temperature 50 mK. c) Zoom-in to a part of the panel b). The arrow points to a characteristic edge localized state. The line trace on the left shows a single experimentally measured capacitance trace as a function of top gate voltage. Peaks arise from single electrons moving back and forth between the dot and the substrate in response to a small ac excitation. The panel is built from 250 separate capacitance traces.

If the electrostatic potential on the ring gate is made more negative, it becomes more difficult to add electrons to the periphery of the dot. One should observe peaks of periphery electrons moving higher in energy (to more positive top gate potential) with more negative ring gate potential and only a small effect on core electrons. The measurements shown below are on a 0.8 micron diameter dot displaying the pairing effect. However, the dot is large, and the pairs do not appear periodically with electron number as in Fig. 2.

Figures 3b and 3c (zoom-in) display results from this experiment. The figures plots the sample capacitance in gray scale as a function of both ring gate (abscissa) and top gate voltage (ordinate). The dark traces arise from capacitance peaks resulting from the addition of single electrons to the dot. The first trace at the bottom of Fig. 3b arises from the very first electron in the dot. Notice that all of the traces display a positive slope throughout the image. The slope measures the sensitivity of the energies of the electronic states to the ring gate and thereby their positions within the dot.

Clearly most all traces in Figs. 3b and 3c can be grouped into two distinct categories, those with steep slopes (periphery electrons) and shallow slopes (core). The distinctness of these two groups establishes that the dot does indeed break-up into isolated core and periphery regions. Notice also that at high electron numbers (near the top of Fig. 3b) the two groups blend into one and the distinction between core and periphery electrons breaks down. The electrons effectively delocalize over the full area of the dot.

Closer examination in Fig. 3c reveals another feature. As one still expects Coulomb repulsion between electrons in the two regions, the existence of levels that cross directly (rather than anticross) is a surprise. This behavior corresponds to the pairing behavior observed in Fig. 2. At the direct crossings, two electrons enter the dot at the same top gate voltage. The absence of Coulomb repulsion leading to the pairing effect is still a mystery, but we now understand that the paired electrons enter isolated positions within the dot.

The problem of electron localization has remained a prime focus of experimental and theoretical research over past 40 years. Misha Brodsky has now built several structures that contain several potential nearby wells, and he has observed sudden transitions from localized electrons in distinct wells to delocalized electrons over several adjacent wells. We hope to achieve fundamental understanding of the phenomenon of localization by building structures that probe the location of a single electron.

Single Electron Contours in the Quantum Hall Liquid

What creates the amazing properties of electrons moving in two-dimensions inside of a semiconductor? Upon changing the strength of a perpendicularly applied magnetic field, one finds that over ranges of fields the system behaves as a perfect conductor along its edges but as a perfect insulator in the interior. For other fields, the interior conducts electricity as a normal metal. Discoveries in this system have rewarded researchers with the 1985 and the 1998 Nobel Prizes in Physics.

The fact that the two-dimensional systems that experimenters study lies 1000 Angstroms beneath the surface has made determination of the microscopic character of the quantum Hall state quite difficult.

Our group has built a scanning microscope that creates images of the quantum Hall state by sensing charges in the 2D system. A sharp metal tip, scanned just above the semiconductor surface connected to a charge sensor, detects charges in the 2D through its capacitive coupling to the 2D layer (see figure 4). The apparatus, operating at 0.3 Kelvin, is so sensitive that it can detect single electrons in the 2D layer.

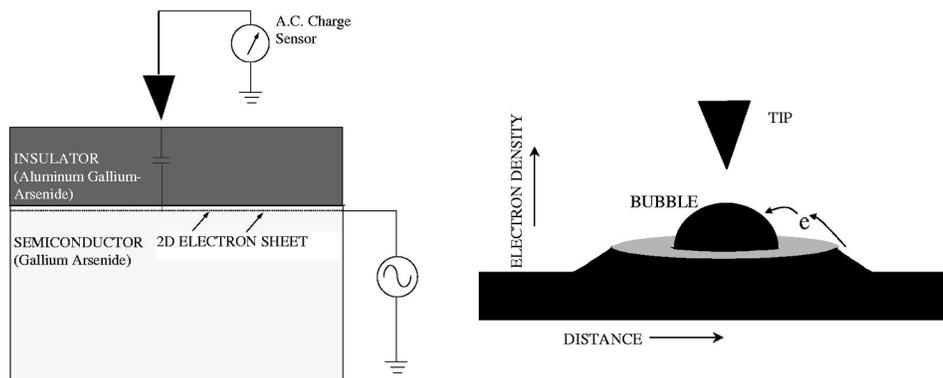


Fig. 4. (left image) Schematic of the subsurface charge accumulation imaging scheme. An ac excitation is applied to a buried two-dimensional electron gas. Due to the self capacitance of the 2D system, charge enters and leaves the 2D. Regions of the 2D system vary in their compressibility (propensity to absorb charge), and a scanning tip attached to a sensitive transistor detects these variations. Aside from sensing compressibility variations, we can now measure directly the electrostatic potential inside the 2D system. We do this by monitoring the motions of single electrons into and out of an "accumulation bubble", as shown in the right image. A positive voltage applied to the scanning tip accumulates electrons underneath the tip. If more electrons accumulate than can be contained in one Landau level, they form a bubble of electrons in the next level. This bubble must hold an integer number of electrons. As we scan the bubble around with the tip, electrons enter or leave it in response to changes in the local electrostatic potential. The tip registers these single electron changes to produce a contour map of the electrostatic potential in the buried system.

Postdoc Gleb Finkelstein and graduate Paul Glicofridis used the tip locally increase the density of electrons within the 2D layer by applying a positive voltage to it with respect to the 2D layer. Thus, in scanning the tip around, the researchers also scan around a local enhancement of the electron density. In magnetic field, electrons in the 2D system must occupy electronic states within discrete Landau energy

levels. As each Landau level can hold only a certain density of electrons, it is possible for the researchers to create a "bubble" of electrons in a higher Landau level than the bubble's surroundings. This bubble is surrounded by a region where the lower Landau level is exactly filled. This region acts similarly to a filled band in an insulator, and electrons must tunnel across this region to travel between the bubble and the surroundings.

As the bubble is scanned around within the 2D layer, electrons tunnel into or out of the bubble. They do so in response to changes in the electrostatic potential at the position of the bubble. The apparatus registers a peak in signal each time the charge inside the bubble changes by a single electron. In sensing the transfer of single electrons in and out of the bubble, the microscope produces a contour map of the electrostatic potential inside the quantum Hall system (see figure 5). By comparing this contour map under different quantum Hall effect conditions, our group has now been able to ascertain basic features of the microscopic character of the quantum Hall liquid in their samples.

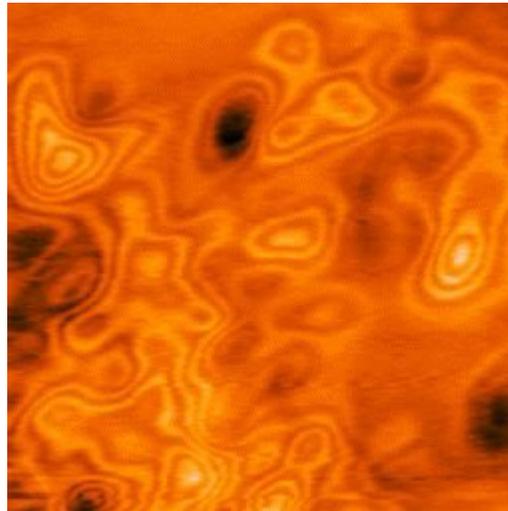


Fig. 5. A 2 μm by 2 μm image produced by scanning an accumulation bubble. Each bright contour results from a single electron entering or leaving the scanned bubble, and the contour map represents the electrostatic potential inside the buried 2D system. Each successive contour represents a potential energy change of about 0.3 meV. The magnetic field is set to 5.9 Tesla, and the Landau level filling fraction is slightly less than one.

Tunneling Into Two-Dimensional Electron Systems

Our ability to sense very small amounts of electrical charge has permitted us to make some basic queries about interacting electronic systems. So far I have describe two basic questions that the experiments can answer: how much energy does it take to add an electron to a quantum dot and where does charge flow in an electron system as a result of a change in the chemical potential of the system? The simplicity of the charge sensing measurement can often result in fundamental information about low dimensional electronic systems not available with other measurement techniques.

There are many other basic questions that can be answered with these techniques. Among them is the following: how likely is it that an electron with a given energy will be able to tunnel into a two-dimensional or any other electronic system? Such measurements have often been unrealizable because it may be practically impossible to produce separate electrical to an isolated low-dimensional electronic system and a neighboring metallic electron "injector". We have overcome this difficulty by developing a contactless capacitance method for making such measurements. We call this method "time domain capacitance spectroscopy" (TDCS). We have used TDCS to understand in detail the characteristics of tunneling of electrons into a 2DES in magnetic field. We discovered a universal shape of the tunneling density of states (growing linearly with excitation) that has arisen in each of the 6 samples (including high mobility samples) that we measured.

Our recent measurements have allowed us to measure directly spin relaxations inside of a ferromagnetic 2D electronic system. We find that initially electrons can tunnel into this system quickly, but

then the tunneling rate drops markedly. According to a theory developed to explain our experiments, electrons entering the system must eventually relax into a ground state structures known as Skrymions, before other electrons can enter. We have thus found a new way for probing spin dynamics and discovered a "spin blockade" that prevents electrons from tunneling into a system if sufficient time is not for electrons to relax into their spin groundstate.

Chemically Derived Quantum Dots

The Bawendi group in the chemistry department at MIT has learned to make chemically derived small clusters (size scale around 30-100 Angstroms) with remarkable uniformity. They can also produce highly regular arrays of particles in two or three dimensions. We have started working with the Bawendi group to place such arrays inside capacitors so that we may perform SECS measurements on them. As the coupling between dots can be adjusted by changing the chemical constituents on the dot surfaces, many different types of 2D materials, from tight binding lattices to nearly free electron metals, can be approximated by this type of lateral "superlattice". The SECS measurements can then be used as a sensitive probe of the type of "material" produced this way. Moreover, large arrays are not required for these measurements. In SECS, electrons are transferred vertically inside a capacitor and into the quantum dots. Electrons do not need to be transferred through the dots for the measurement to sense them. We should be able to measure isolated clusters this way. The SECS measurements can therefore be used to study spectra of single particles, dimers, small clusters, and large arrays.

We have made significant progress in building capacitor structures incorporating the Bawendi dots. How does one embed small particles inside of an insulator but with the particles close enough to one metal electrode to allow tunneling? We have two approaches; one uses self assembled monolayers as the insulating medium, and the other uses a novel technique to deposit ZnS at low temperatures (100 C). As this fabrication is entirely new, we have encountered several obstacles, but we expect to have functioning structures this year. We hope to eventually learn to measure directly the number of electrons in a single dot while simultaneously observing its luminescence properties.

Publications

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5. G. Finkelstein, P.I. Glicofridis, S.H. Tessmer, R.C. Ashoori, and M. Shayegan, "Imaging of Low Compressibility Strips in the Quantum Hall Liquid, *submitted to Physical Review Letters* and appearing as cond- mat/ 9910061.
6. H. B. Chan, R. C. Ashoori, L. N. Pfeiffer, and K. W. West, "Observation of a Spin Bottleneck for Tunneling into the $\nu=1$ Quantum Hall State", *to appear in Physica E*

7. M. Brodsky, N.B. Zhitenev, R.C. Ashoori, L.N. Pfeiffer, and K.W. West, "Localization-Delocalization Transition in Double Quantum Dots", *to appear in Physica E*
8. G. Finkelstein, P.I. Glicofridis, S.H. Tessmer, R.C. Ashoori, and M. R. Melloch, "Imaging the Low Compressibility Strips Formed by the Quantum Hall Liquid in a Smooth Potential Gradient", *to appear in Physica E*
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