Chapter 2. Single-Electron Capacitance Spectroscopy

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2.1 Introduction

The principal focus of research in our laboratory is the study of interacting electronic systems in low dimensional semiconductor structures. Systems in which electrons exist purely in one or two 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 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.

2.2 Single-Electron Capacitance Spectroscopy of Quantum Dots

Sponsors

Joint Services Electronics Program
Grant DAAG55-98-1-0080
National Science Foundation
National Science Foundation/Materials Research Science and Engineering Centers
David and Lucille Packard Foundation
U.S. Navy - Office of Naval Research

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We developed a method known as single electron capacitance spectroscopy (SECS) which permits measurement of the electronic energy levels of a single quantum dot or “artificial atom.” This method allows us to vary controllably the number of electrons

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in artificial atoms (starting from the first electron) and to measure precisely the energy required to add successive electrons. Our artificial atoms are much larger than real atoms, and this has the consequence of greatly accentuating the effects of electron-electron interactions and resulting in quite different physics.

While some features appear in the spectra that could be predicted from a simple noninteracting model of the artificial atom, the spectra clearly display features attributable to electron-electron interactions. For instance, we observe effects of ferromagnetism of the electron gas: for particular values of an applied magnetic field, all electronic spins flip and line up in the same direction. At higher or lower fields, the spins depolarize. At high magnetic fields, the electron density in the artificial atom even undergoes a bifurcation into discrete inner and outer shells with low-electron density between the shells.

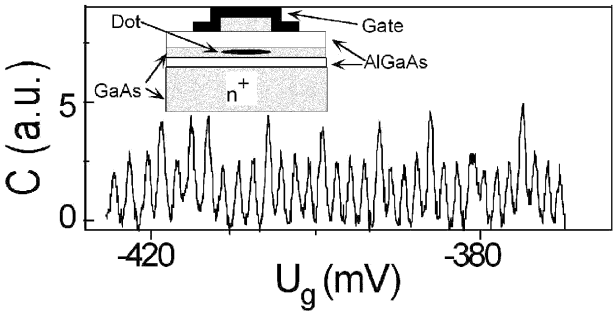

Figure 1. Inset: Schematic of the dot embedded within a capacitor. Electrons can tunnel from the bottom n+ electrode to the dot. Main Figure: Quantum dot capacitance as a function of gate voltage. Each peak denotes the appearance of an electron in the dot. Double height peaks indicate the addition of two electrons. The temperature is 0.3 K.

While many of the spectroscopic features that we have observed can now be understood theoretically, we have uncovered a profound puzzle. In larger artificial atoms that contain few electrons, a physical process exists which appears to exactly cancel the interaction between electrons! In SECS spectra of small artificial atoms, we always observe that there is an increased energy cost for adding successive electrons to the system. This is simple to understand: electrons already in the artificial atom repel additional electrons from being added to the system. However, in larger artificial atoms, sometimes two or even more electrons can be added to the system with no additional energy cost for successive electrons. Even more striking, for intermediate size artificial atoms, every fourth and fifth electron added to the system appears as a pair. Figure 1 displays an SECS spectrum from a dot displaying this behavior. The periodicity of the bunching suggests that it is associated with electron additions into spatially distinct regions within the artificial atoms. Recently, we have performed experiments on artificial atoms with adjustable shape, allowing us to separately adjust energies of electrons in different positions within the artificial atoms. After the shape is varied beyond a threshold, the pairs suddenly split, and, with more shape variation, new pairs form. This behavior is consistent with the model that paired electrons enter distinct positions within the atom. However, the long-range mechanism that binds the two distant electrons into pairs is still entirely a mystery.

2.3 Imaging Electrons Inside Materials

Sponsors

Joint Services Electronics Program
Grant DAAG55-98-1-0080
National Science Foundation
National Science Foundation/Materials Research Science and Engineering Centers
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Our work comprises study of other low-dimensional electronic systems. One of the most intriguing among these is the two-dimensional electronic system (2DES). In an applied magnetic field, this system gives rise to the quantum Hall effect. As quantum dots often consist simply of confined regions of 2DES, study of quantum dots is often closely related to study of the physics of the quantum Hall effect. Moreover, the potential landscape within a two-dimensional system inevitably contains local potential minima and maxima. For this reason, it may be

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sometimes appropriate to think of the system as consisting of many quantum dots (potential valleys) and antidots (potential maxima).

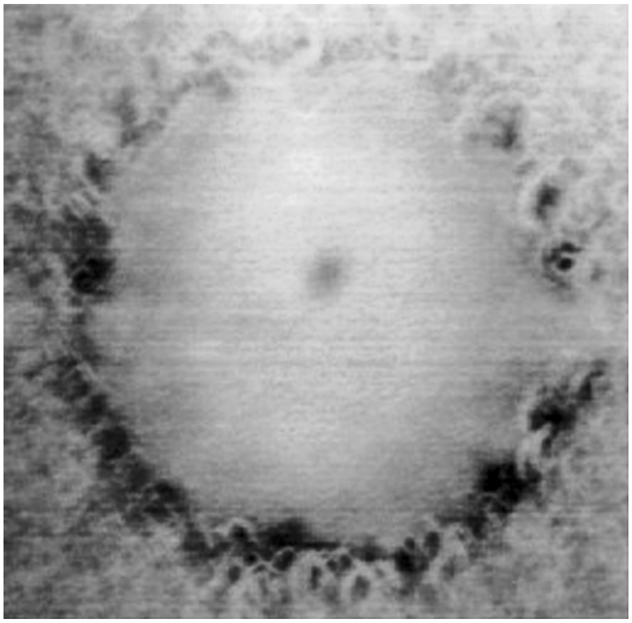

Figure 2. A subsurface charge accumulation (SCA) image of a two-dimensional electron gas (2DEG) in the vicinity of a prepared local density maximum of the 2DEG exists at the center of the image. The scale of the image is 10×10 µm. The electron density is approximately 10% higher at the center of the image than at the edges. A 3.07 tesla magnetic field is applied perpendicular to the plane of the 2DEG. This field causes electrons to be quantized into Landau levels, and their subsequent filling depends on the local electron density. Regions of completely filled levels cannot accommodate more electrons and therefore cannot accumulate charge. The SCA images measure the ability of the system to absorb charge. The black ring is a region of precisely filled Landau level. Outside the ring is a region of partially filled Landau level, and inside the ring is an area in the next (higher) Landau level, also partially filled.

Of course, actual images of the distribution of electronic charges inside confined structures may be even more valuable than inferences drawn from spectroscopic measurements. Such imaging is useful not only for quantum dot systems, but also for a variety of low-dimensional systems. A major challenge arises in producing such images because the electronic systems exist deep beneath the surfaces of semiconductors. We have overcome this difficulty in developing a cryogenic scanned probe technique called subsurface charge accumulation (SCA) imaging. It permits very high resolution examination of electronic systems *inside* materials. We have used our SCA microscope to image directly the nanoscale structures that exist in the 2DES. Amazingly, we can now actually *see* some of the processes that give rise to features in SECS spectra. We have learned how to produce samples with tailored local density minima or maxima. "Compressible" (high thermodynamic density of states) and "incompressible" (very low density of states) regions associated with regions of different Landau level filling factor can be clearly identified. Finally, images of regions of local shallow potential minima directly depict the bifurcation of the electron density into inner and outer shells as discussed above for structures similar to artificial atoms. Figure 2 displays a typical SCA image demonstrating this behavior.

Our work on SCA imaging is still in its infancy, but we can already make some key remarks about the behavior of a 2DES that does not contain intentionally produced density minima or maxima. First, the 2DES appears uniformly compressible for fields away from the integer Landau level filling factors (i.e., away from quantum Hall plateaus). Second, spatial compressibility structure appears only near integer Landau level filling factors, and this structure evolves with enormous sensitivity to magnetic field. Imaging the compressibility at fields differing by only 0.5% yields images which appear to have no correlation of spatial features. Both of these results are fundamentally surprising; several theoretical notions exist, and some predicted structure between the plateaus, but none seem to have predicted the rapid evolution of the observed structure.

2.4 Tunneling Into Two-Dimensional Electron Systems

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Joint Services Electronics Program
Grant DAAG55-98-1-0080
National Science Foundation
National Science Foundation/Materials Research
Science and Engineering Centers
David and Lucille Packard Foundation
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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 described two basic questions that the experiments can answer: (1) how much energy does it take to add an electron to a quantum dot, and (2) where the charge flows 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 contacts 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 six samples (including high mobility samples) that we measured.

2.5 Chemically Derived Quantum Dots

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Kazumasa Nomoto, Dmitri Pouchine, Professor Raymond C. Ashoori, in collaboration with the group of Professor Moungi G. Bawendi

Professor Moungi G. Bawendi's group in MIT's chemistry department has learned to make chemically derived small clusters (size scale around 30-100 Å) with remarkable uniformity. They can also produce highly regular arrays of particles in two or three dimensions. We have started working with Professor Bawendi's group to place such arrays inside capacitors so that we may perform SECS measurements. Because the coupling between dots can be adjusted by changing the chemical constituents on the dot surfaces, many different types of 2-D 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. We do not need to transfer electrons 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.

2.6 Publications

Ashoori, R.C., N.B. Zhitenev, L.N. Pfeiffer, and K.W. West. "Paired Additions of Electrons into Quantum Dots: How and Why does Localization Destroy Coulomb Blockade to Produce Pairing?" *Physica E* 3: 15-22 (1998).

Berman, D., N.B. Zhitenev, R.C. Ashoori, and M. Shayegan. "Observation of Quantum Fluctuations of Charge on a Quantum Dot." *Phys. Rev. Lett.* 82: 161-64 (1999).

Koltonyuk, M., D. Berman, N.B. Zhitenev, R.C. Ashoori, L.N. Pfeiffer and K.W. West. "Single Electron Capacitance Spectroscopy of Vertical Quantum Dots Using a Single Electron Transistor." *Appl. Phys. Lett.* 74**:** 555-57 (1999).

Tessmer, S.H., P.I. Glicofridis, R.C. Ashoori, and M.R. Melloch. "Subsurface Charge Accumulation Imaging of a Quantum Hall Liquid." *Nature* 392: 51-54 (1998).