Electronic spectroscopy of zero-dimensional systems

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We have measured the thermodynamic properties of dispersionless electronic systems. We observe structure in the capacitance of quantum dots reflecting the discrete energy levels of electrons confined in all three spatial dimensions. The spacings of the observed structure and the systematic variation with the degree of confinement are consistent with quantum-size effects.

Experimental studies of zero-dimensional (0D) systems have focused primarily on their optical properties.[1-3] This is due to the difficulty of fabricating 0D systems so that transport can be carried out. Nonetheless, some tunneling measurements on small structures have exhibited behavior characteristic of trapping and transport of very small numbers of charge carriers.[4,5] A powerful technique for probing small structures is the measurement of capacitance. Capacitance spectroscopy has been widely used to study electronic states at metal-semiconductor interfaces,[6] in semiconductors,[7] and in insulators.[8] In many instances the energy levels are atomic in nature and arise from isolated defects or impurities. In this paper we report the results of capacitance measurements on 0D electronic systems. These systems are also atomic in the sense that they are noninteracting and comprise very few electrons. However, unlike impurities in insulators, for example, these systems are tunable in that their size and energy-level spacings can be systematically varied. This allows one to tailor 0D systems for specific purposes. Our measurements reveal structure in the capacitance related to the presence of 0D quantum levels. The spacing of these levels and their shape vary systematically with the size of the confining potential.

The samples used in our experiments are modulation-doped GaAs-$Al_xGa_{1-x}As$ heterostructures with two unique features: a thick GaAs cap layer which is etched into dots, and a heavily doped GaAs substrate separated from the $Al_xGa_{1-x}As$ by 800 Å of GaAs with no intentional doping. The capacitors were fabricated with dots having 0.1-, 0.2-, 0.3-, and 0.4-μm diameters. Figure 1 shows scanning electron micrographs of two samples before and after reactive-ion etching (RIE). To fabricate the capacitors, dot patterns are written into polymethylmethacrylate (PMMA) using a high-resolution electron-beam pattern generator.[9] Next, thin metal is deposited over the PMMA and lifted off to define the dots. The GaAs cap layer is selectively etched in a CCl_2F_2 and He gas mixture[10] using the metal as an etch mask. To reduce the damage to the surface of the $Al_xGa_{1-x}As$ and the underlying structures, the etching is terminated immediately after the $Al_xGa_{1-x}As$ surface is exposed. Finally, a metal gate is deposited. Because the sample has a conducting substrate the mobility cannot be measured directly. However, an unpatterned sample grown to similar specifications has a mobility of 5×10^5 cm^2/V s and a carrier concentration of 3.4×10^{11} cm^{-2} at 4.2 K.

The potential profile produced by this confinement scheme is very anisotropic. The potential well confining the electrons at the GaAs-$Al_xGa_{1-x}As$ interface can be approximated by a triangular well with an effective width of about 150 Å at the Fermi energy. On the other hand, we have found (from two-dimensional calculations of the potential profiles for these structures) that the lateral confining potential is roughly circular and between 500 and 3000 Å in diameter. Although the patterns etched into the surface of the capacitors are rectangular with rounded corners, the confining potential near the heterojunction is rounder and smaller than the physical structures. The overall confinement is actually much closer to a thin disk than a box, and hence these structures have cylindrical symmetry rather than the spherical symmetry associated with atomic orbitals.

We performed our capacitance spectroscopy at cryogenic temperatures (4.2–0.4 K) under conditions such that the measured capacitance can be related to the thermodynamic density of states (DOS). The general features of the capacitance of our 0D capacitors are similar to those of the 1D capacitors we have studied.[11] This partly reflects the fact that the strongest confinement is that of the heterojunction. However, there are a number of important differences. The structure in the capacitance is weaker than in the 1D samples, the oscillations are more widely spaced, and noise and hysteresis effects are more pronounced. Each of these effects result from the nature of the confining potential and will be discussed in detail.

As previously discussed,[11] we determined the positions of the peaks in the DOS by measuring the first derivative of the capacitance (see Fig. 2). For all but the smallest dots there is a large peak reflecting the initial turn-on of the capacitors. We were not able to observe any clear structure in the 100-nm dots and none of the 100-nm-dot samples exhibited a clear turn-on. Rather, the capacitance and its derivative both increased monotonically. The fact that we did not observe any clear structure may be related to the large spacings of the energy states in these capacitors. The initial peak in the derivative of the capacitance of the 200-, 300-, and 400-nm-dot samples is fairly broad. Above turn-on, we observe several oscillations in the signal amplitude. These oscillations directly reflect the change in the density of states as the Fermi energy passes through 0D quantum levels. The spacings be-

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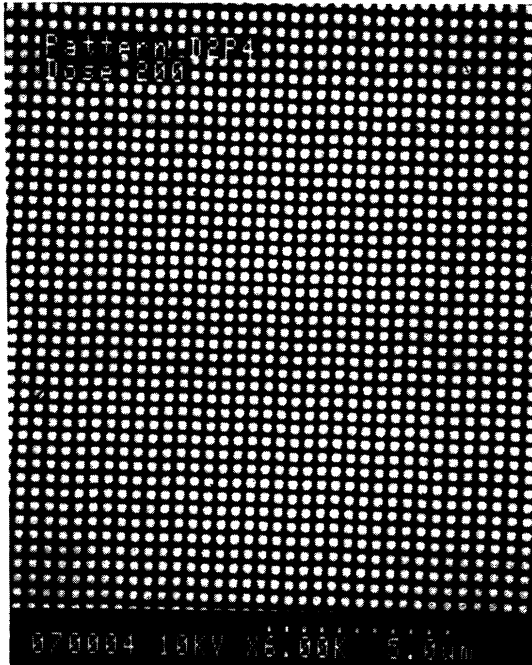
tween the peaks in the signal are about 105 mV for the 200-nm dots, 70 mV for the 300-nm dots, and 50 mV for the 400-nm dots. This reduction in spacing with increasing dot size is consistent with a simple particle-in-a-box picture.



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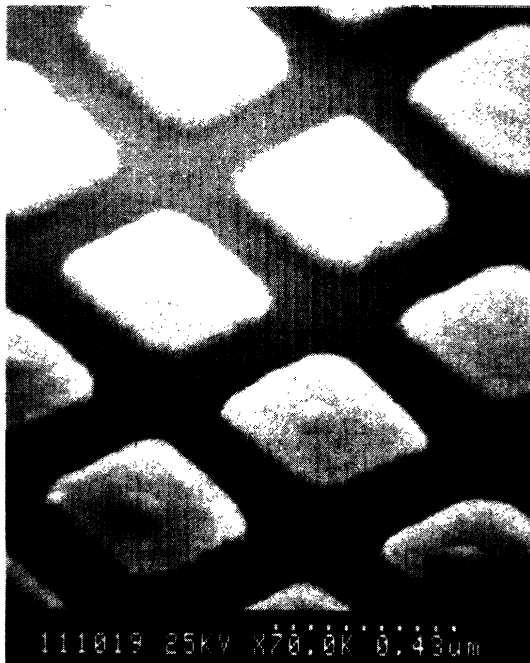

(b)

FIG. 1. (a) Scanning electron micrographs of many nominally 200-nm metal dots used as a RIE mask as they appear before etching. (b) A high-resolution micrograph of 300-nm dots. No surface roughness is resolved on the etched $Al_xGa_{1-x}As$.

Interestingly, the peaks in the signal are clearest in the 300-nm-dot samples. This probably occurs because of two competing effects: the 0D energy eigenstates become more widely spaced as the size of the dots is reduced and this tends to sharpen the structure, while the broadening of the levels due to boundary effects tends to become larger as the dots become smaller and this tends to smear out the structure in the capacitance. Another interesting observation is the systematic shift in the turn-on voltage of the capacitors. Although there are small differences among samples with the same dot sizes, in all cases the smallest dots require the highest positive bias to induce electrons at the heterojunction and the largest dots require the lowest bias. This is consistent with the electrostatic confinement scheme we employ. If we assume that the depletion width is the same for all samples at the same bias then the smallest dots can be completely depleted at the same bias that the largest dots are turned on. This provides further evidence that the electrostatic potentials in our samples truly reflect the lithographically defined structures.

The conclusion that the smaller dots are more depleted than larger ones comes from analysis of solutions to the Poisson and Schrödinger equations for 1D structures. For the 0D structures, however, there are a number of

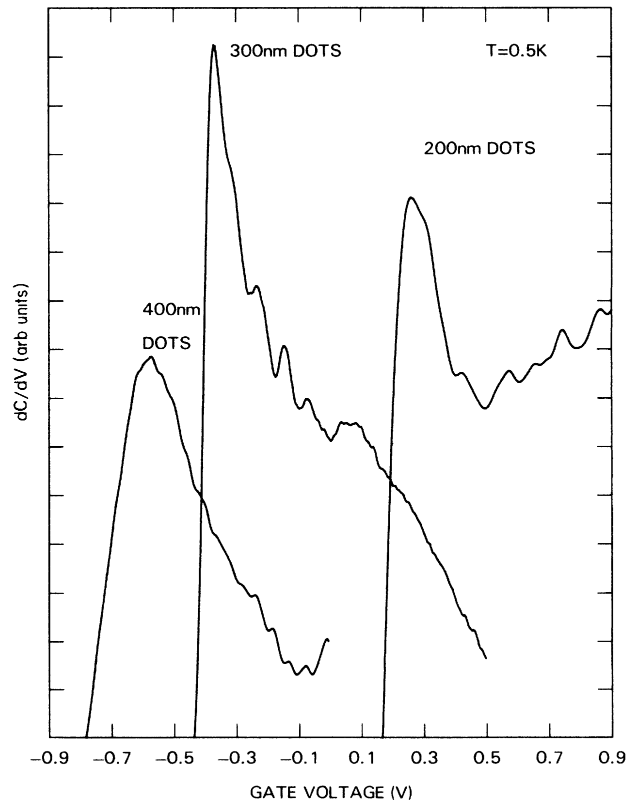

FIG. 2. The derivative of the capacitance vs gate voltage for three quantum dot sizes. The main peak in the derivative is due to turn-on of the capacitors.

differences which render further extrapolation meaningless. First the symmetry of the electrostatic problem is different. In the 1D case we are studying a line charge with its essentially $\ln(r)$ potential, while the 0D case involves a disk of charge which is closer to the point charge potential of $1/r$. Since the charge distribution and confining potential are intimately coupled one cannot simply take the cross section of a 1D line as representative of a 0D disk. In addition, the dependence of the Fermi energy on applied bias cannot be inferred from the 1D results. An essential difference between these two systems is that the states of the 1D lines have dispersion in phase space while the 0D states do not. If one ignores this and attempts a simple extrapolation from 1D, the fundamental physics of the problem is lost. For accurate solutions, the best candidates appear to be either a fully-self-consistent three-dimensional numerical solution to the Poisson and Schrödinger equations or perhaps an analytical solution to the Schrödinger equation coupled self-consistently with a numerical solution to the three-dimensional Poisson equation. Both are large undertakings.

Figure 3 shows the signals from 300-nm dots and lines. The turn-on of the samples with lines are consistently sharper than the turn-on of the dots (as measured by the width of the main peak in the derivative of the capacitance). In addition, the oscillations in the capacitance are much clearer for the samples with lines. This is due to several effects. First, the active area of line samples is about twice that of dot samples and this tends to reduce the signal-to-noise ratio. Second, the boundary effects are much more pronounced in the dot samples. This reflects the fact that the electron wave functions overlap the boundaries of the confining potential in all directions in the dots but only in two directions in the lines. Third, the variations in dot size over the sample may be more deleterious than the variations in the 1D samples. And finally, there seems to be some noise generated in the dot samples which is not detectable in the line samples. This noise is not reproducible but shows a strong dependence on gate bias. It may be that this noise is related to the small number of electrons which can occupy each of the energy states. In the line samples the eigenstates have very large degeneracies whereas the energy levels in the quantum boxes are often only twofold degenerate (spin "up" and spin "down"). Although we sample many dots at the same time (in parallel) the change in total charge may still be discrete if only a few dots are involved at a

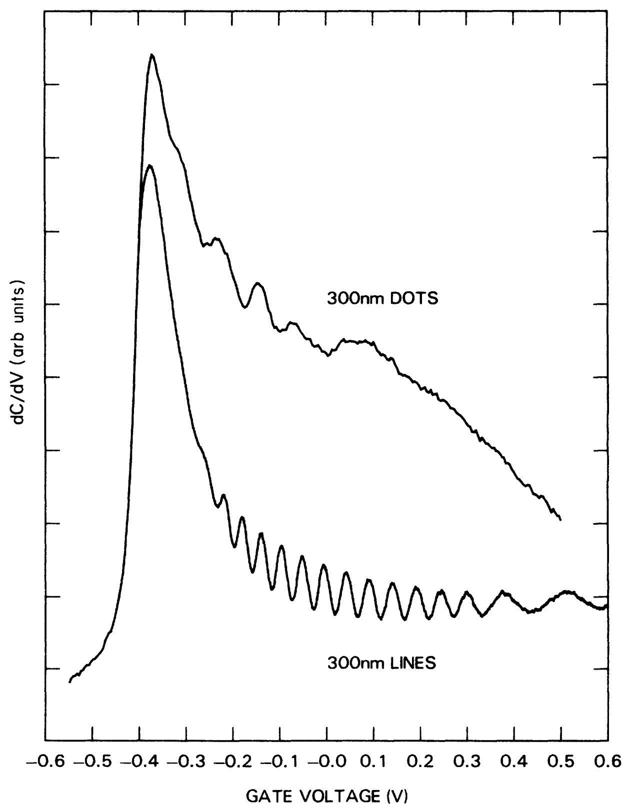

FIG. 3. A comparison of the signal from 300-nm lines and 300-nm dots.

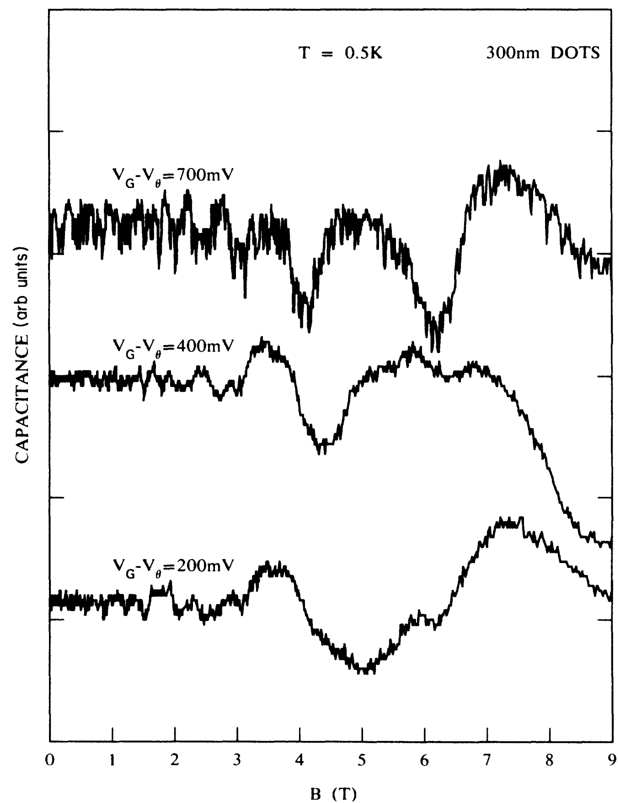

FIG. 4. The measured capacitance of a sample with 300-nm dots vs magnetic field. The three traces were taken under identical measurement conditions except for the dc bias. The bottom curve is for a bias 200 mV above threshold, the middle curve is for 400 mV above threshold, and the top curve is for 700 mV above threshold. The noise is attributed to the leakage of small numbers of electrons through the capacitor.

particular bias. Another interesting result is that the oscillations in the derivative of the capacitance of the dot samples are spaced more widely in gate voltage than the oscillations in the line samples. This probably reflects the fact that the effective width of the confining potential of the 0D dots is smaller than that of the lines of the same lithographic feature size under similar bias conditions. As mentioned above, resolution of this question requires more complete calculations.

We also examined the properties of our 0D capacitors in the presence of a magnetic field applied perpendicular to the plane of the heterojunction. Figure 4 shows the capacitance versus magnetic field for a sample with 300-nm dots. The samples exhibit Shubnikov–de Haas–type oscillations at high magnetic fields where the magnetic length ($\sqrt{\hbar/eB}$) is much shorter than the effective size of the dots. In this regime the electrons can execute several cyclotron orbits before being scattered. However, there are small changes in the capacitance superposed on the major oscillations. They are more prominent at low biases but are seen in all the spectra. They may be related to the confinement. Oscillations in the capacitance due to the presence of Landau levels are not seen below about 2 T. This corresponds to a magnetic length of 180 Å. If we assume that the Landau-level spacing is roughly equal to the level broadening when we first observe these oscillations this corresponds to an energy of about 3.4 meV. This would explain why the oscillations in the capacitance are very weak since this is comparable to the 0D energy-level spacings. Magnetic field measurements also allow us to determine an effective carrier density from the periodicity of these oscillations. The data shown are for biases of 200, 400, and 700 mV above threshold, V_θ. At the highest bias the carrier concentration is 6×10^{11} cm^{-2} and this is above the range where we observe 0D levels. At a bias of 300 mV above threshold we observe the 0D levels in the 300-nm samples. At this bias the carrier concentration deduced from the magnetocapacitance oscillations is 3.4×10^{11} cm^{-2}. If we assume that the effective diameter of our dots is about 1000 Å at this bias, then this corresponds to a total electron density of about 27 per dot. This would mean that we have filled less than ten 0D levels at this bias.

In all our measurements (both electric field and magnetic field sweeps) we observe noise which is larger than the ambient noise level. This noise becomes more pronounced at large positive biases. Because of its strong bias dependence (see Fig. 4) we believe it is related to leakage currents in the capacitors. Although, the total leakage current is less than 30 pA at the highest biases studied this could be sufficient to produce the observed noise. This noise could be related to the tunneling of individual charges through the isolated capacitors. It may also be related to charging of the capacitors, but the changes in the capacitance are too large to represent the change in occupation by a single electron. This noise does not appear to have the temporal properties of the "telegraphic noise" first reported by Ralls *et al.*[12] and it is not reproducible. Noise of this nature was not observed in the 1D capacitors and this supports the hypothesis that it is related to the lack of dispersion in the density of states of the 0D capacitors.

Finally, we should point out that in all our 0D samples we observe hysteresis in the capacitance-voltage characteristics. The problems associated with this hysteresis can be avoided by sweeping the voltage very slowly so that is not important technically. However, it may be important in understanding the basic properties of the structures.

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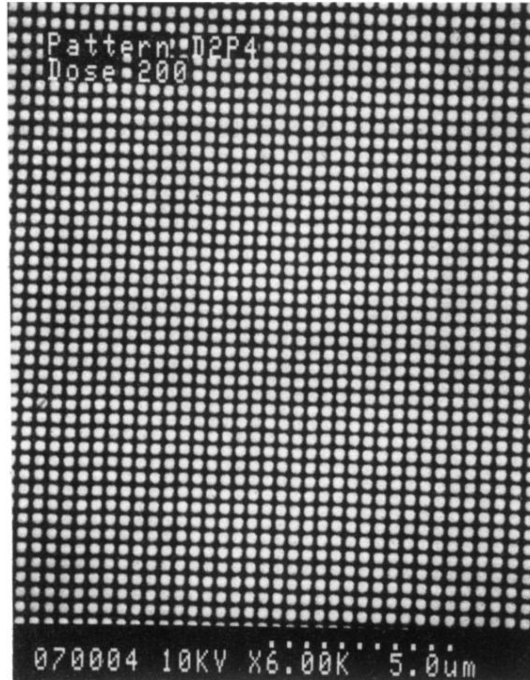

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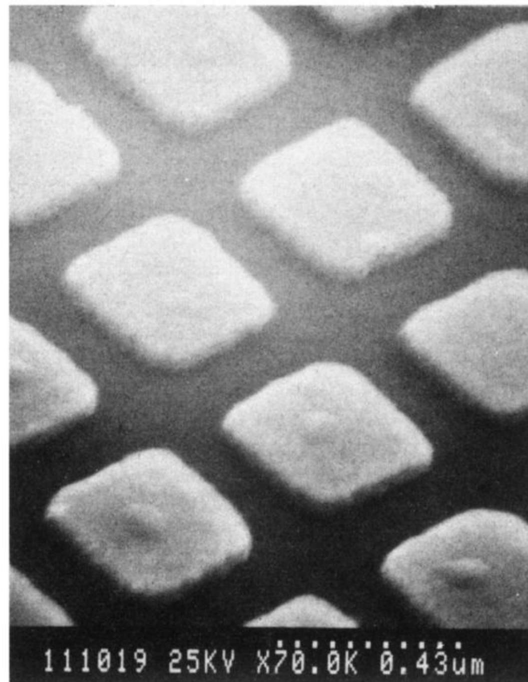

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