Electrons in Boxes

Probing artificial atoms to stretch quantum physics

By IVARS PETERSON

Weird things happen when particles are confined to tiny boxes. An atom, for example, can be pictured as a spherical container for electrons. The attractive electrical force between the negatively charged electrons and the atom's positively charged nucleus serves as the container's walls.

According to quantum theory, electrons trapped in such minuscule packages follow only certain orbits, each representing a specific energy. Researchers can determine those energies by measuring the wavelengths of light absorbed or emitted by atoms as electrons jump abruptly from one energy level to another.

Nowadays, they can also probe the shenanigans of electrons inside microscopic semiconductor structures called quantum dots. In these artificial atoms, an electric field traps an electron in much the same way that a bowl confines a rolling marble.

"One can consider the artificial atom as a tiny laboratory in which quantum mechanics and the effects of electron-electron interactions can be studied," says Raymond C. Ashoori of the Massachusetts Institute of Technology.

Scientists can construct a wide variety of quantum dots to explore how electrons behave in boxes many times the size of a typical atom or in containers shaped like rods, pancakes, misshapen disks, or distorted spheres.

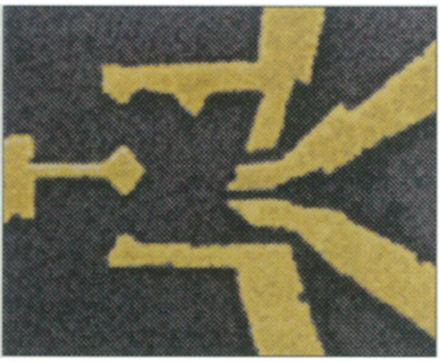
"It's fun to imagine and study how quantum mechanics plays itself out in all sorts of geometries—geometries that atoms can't have," says Paul L. McEuen of the University of California, Berkeley.

Surprises abound. No computer yet can handle the calculations necessary to determine the detailed behavior of a bunch of charged particles in a box of arbitrary shape. "So we don't know what's going to happen in our experiments," McEuen remarks.

There's also a practical aspect. The characteristics of solids typically reflect properties of their microscopic building blocks. "If we could engineer new types of artificial atoms, we could then assemble them into new kinds of solids—ones that could not be realized with real atoms," McEuen says.

Several research groups reported results of quantum dot experiments at an American Physical Society meeting held last month in Los Angeles.

Leo P. Kouwenhoven and his coworkers at the Delft University of Technology in the Netherlands studied the ground state—in which electrons have the lowest possible energy—as well as excited states of a pancake-shaped quantum dot, 0.1 micrometer µm across, containing 1 to 12 electrons.


Marcus et al.

Scanning electron micrograph showing the surface of a quatum dot device, about 1 micrometer wide, made up of layers of gallium arsenide and aluminum gallium arsenide.

By incorporating this quantum dot into a device resembling a transistor and measuring current-voltage relationships, the researchers determined the dot's ground state as they added electrons one by one.

In real atoms, the order in which electrons fill up different energy levels follows a set of rules devised many decades ago by German spectroscopist Friedrich Hund. The Dutch team discovered that electrons obey the same sort of rules in filling energy levels in what is essentially an oversized, two-dimensional atom. For example, the energy levels of pancake helium, with two electrons in the quantum dot, displayed the same sorts of complexities exhibited by real helium.

"Theorists had not predicted that one would see Hund's rules [applied] in quantum dots," Ashoori explains. The observations have since led to theoretical calculations that explain many features of a quantum dot's energy spectrum.

"These experiments beautifully illustrate that for a high-symmetry quantum dot of a few electrons, the ideas of atomic physics coupled with many-body quantum calculations can give a relatively complete qualitative and quantitative description of the observed behavior," McEuen commented in the Dec. 5, 1997 SCIENCE.

Charles M. Marcus and his colleagues at Stanford University have taken a somewhat different tack, focusing on irregularly shaped quantum dots containing about 200 electrons chilled to millikelvin temperatures. In this case, no simple pattern, or shell structure, is evident in the energy spectrum.

"Many basic questions about the level structure remain a mystery," McEuen notes. For example, no one knows whether electrons in the ground state tend to pair up so that they have opposite spins, as they do in atoms. Such pairing would influence the quantum dot's magnetic characteristics.

The large number of electrons also makes such a quantum dot the rough equivalent of an atom that has far more electrons than occur in natural elements. Thus, researchers have a vehicle for probing the quantum physics of electrons in elements that can't otherwise be synthesized and studied.

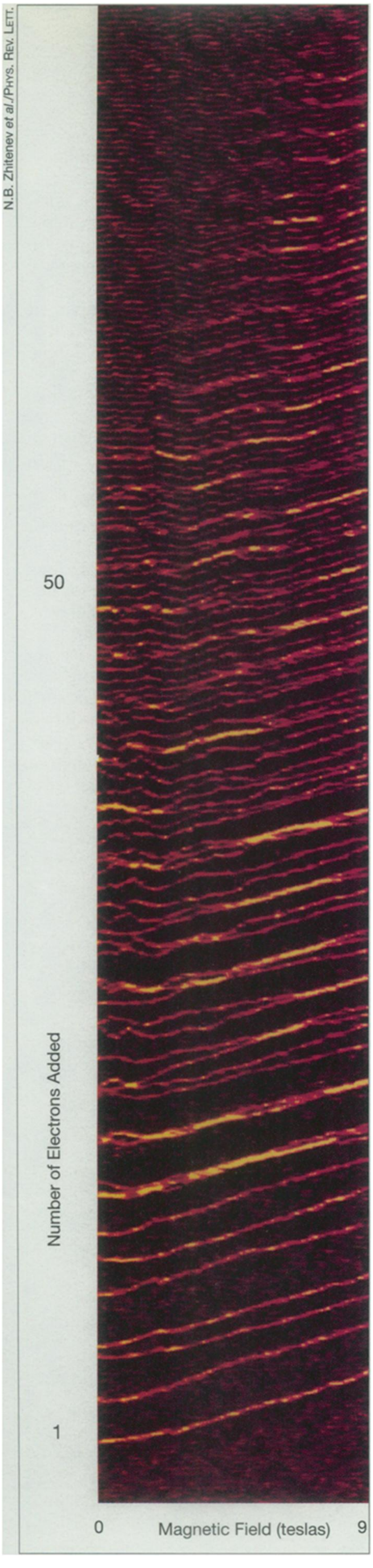
In beginning to address these issues, Marcus and his coworkers showed that, although the energy level structure of such a quantum dot is complex, there are discernible patterns. For instance, adding an extra electron to the quantum dot generally produces a ground state structure of energy levels resembling that observed when the quantum dot, was in its first excited state, before the addition.

Theorists had incorrectly expected that electrical interactions between so many electrons would thoroughly scramble the quantum states, washing out the possibility of strong correlations between ground and excited states of quantum dots with different numbers of electrons.

The new results provide valuable insights into the relationship between magnetism and the spins of electrons in quantum dots, Marcus says.

Instead of measuring current-voltage relationships, Ashoori and his colleagues use a technique known as single-electron capacitance spectroscopy to deduce the energy required to add successive electrons to an initially vacant quantum dot (SN: 4/4/92, p. 222).

In effect, the researchers observe how the arrival of each newcomer must overcome (with the help of an increasing applied voltage) the mutual repulsion of



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In this spectrum, each wavering trace shows the amount of energy needed to add an electron to a quantum dot (0.5 micrometer in diameter), starting at the bottom with the first electron injected into an empty dot. As the magnetic field increases from 0 to 9 teslas, that energy changes to reflect various types of quantum interactions.
Note that the first seven electrons enter the quantum dot at widely spaced energies. Three electrons then enter the dot at nearly the same energy. The next two electrons join the assemblage as a pair. After about 40 electrons are added to the dot, the bunching develops a pattern, with one bunch appearing for each four to six electrons added to the dot.

those electrons already in place. The sequence of peaks observed at particular voltages reflects the dot's electronic spectrum.

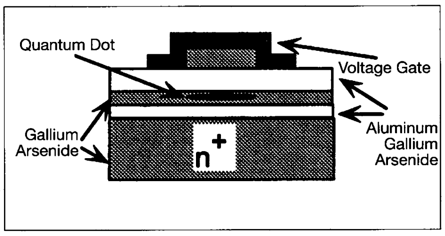
Such spectra "depend drastically upon the size of dots and on the electron number," Ashoori says.

For small quantum dots, just 0.2 μm across, a graph of the charge versus the voltage looks like a staircase—one step for each additional electron. In contrast, the spectra of larger dots (1 μm wide) reveal an astonishing feature. The staircase becomes irregular, indicating that electron additions occur in bunches. For example, under certain conditions, each step up in voltage allows not just one, but two electrons to join the assemblage.

"You would expect that after one electron, it would take more energy to add a second electron because the electrons repel each other," Ashoori says. "What you often see is that you don't have to have additional energy. In fact, you find that for the same amount of energy, you can sometimes add as many as six electrons."

For intermediate-sized quantum dots, the physics gets even weirder. As the number of electrons increases, bunching changes from an occasional occurrence to once for every fifth electron added to the group.

Such regularities and other experimental evidence suggest that electrons settle into two distinct regions—one at the center and another near the edge of a quantum dot. "As you add electrons, four electrons go to the center, then the fifth electron goes to the edge," Ashoori says. "Once that electron is added to the edge, you can add another electron at no additional energy cost."



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Schematic view of the quantum dot used to investigate electron bunching.

Why electrons pair up or bunch in large and medium-sized quantum dots isn't understood. "It's all a huge mystery," Ashoori insists. "Something very strange is going on."

"One does not expect such results in a semiconductor or a normal metal sample," Ashoori and his colleagues note in the Sept. 22, 1997 Physical Review Letters.

"The systematic exploration of artificial atoms is well under way," McEuen says. Such investigations are beginning to reveal how electrons behave in a wide variety of geometries, not just ordinary atoms.

"In a sense, atoms represent a special case from the point of view of quantum systems," Marcus says. "To me, the ultimate aim is to understand the quantum mechanics of coherent, interacting charged systems without requiring the perfect symmetry of the atomic world."

McEuen and his collaborators have been developing techniques to fabricate and study quantum dots only a fraction of the size of those now conventionally used. "Our big push has been toward new geometries on a smaller scale," McEuen says. "We want to move this kind of science into chemically derived nanostructures, such as nanocrystals and nanotubes."

Another important step is to start assembling artificial atoms into molecules and, eventually, solids. In the Dec. 5, 1997 Science, Werner Wegscheider and his coworkers at the Munich Technical University in Garching, Germany, described the fabrication of an artificial molecule consisting of two adjacent quantum dots. The team studied bond strength as the separation of the dots was varied.

"This model system opens new insights into the physics of coupled quantum objects," the researchers note.

Scientists are also striving to develop techniques to image where electrons are located and what they are doing inside quantum dots. "We need to keep refining our experiments," Ashoori contends. "We're very good at adding electrons and measuring how much energy it takes. We need to be able to go in and look for the electrons."

"Finding new ways of interrogating these systems is important," McEuen agrees. At the same time, quantum dot devices can themselves serve as sensitive probes of all sorts of physical, chemical, and biological phenomena on a microscopic level.

More surprises are undoubtedly in store for those researchers working with artificial atoms. So far, "we have not been very good at predicting the next neat thing to happen," McEuen says. "One finds a richness of phenomena that one would not have naively expected to be there." □