By confining electrons in three dimensions inside semiconductors, quantum dots can recreate many of the phenomena observed in atoms and nuclei, making it possible to explore new physics in regimes that cannot otherwise be accessed in the laboratory

Quantum dots

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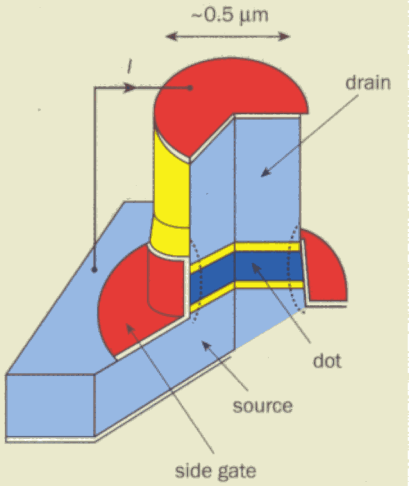
Quantum dots are man-made "droplets" of charge that can contain anything from a single electron to a collection of several thousand. Their typical dimensions range from nanometres to a few microns, and their size, shape and interactions can be precisely controlled through the use of advanced nanofabrication technology.

The physics of quantum dots shows many parallels with the behaviour of naturally occurring quantum systems in atomic and nuclear physics. Indeed, quantum dots exemplify an important trend in condensed-matter physics in which researchers study man-made objects rather than real atoms or nuclei. As in an atom, the energy levels in a quantum dot become quantized due to the confinement of electrons. With quantum dots, however, an experimentalist can scan through the entire periodic table by simply changing a voltage.

Many of these phenomena can be studied by allowing single electrons to tunnel into and out of the dot, since this reveals the quantized energy levels of the device. Experiments at our labs at the Delft University of Technology in the Netherlands and Stanford University in Calfornia have used this technique to probe various properties of quantum dots. What is most exciting is that many of the quantum phenomena observed in real atoms and nuclei - from shell structure in atoms to quantum chaos in nuclei - can be observed in quantum dots. And rather than having to study different elements or isotopes, these effects can be investigated in a quantum dot by simply changing its size or shape.

1 Vertical quantum dot structure



The quantum-dot structure studied at Delft and NTT in Japan is fabricated in the shape of a round pillar. The source and drain are doped semiconductor layers that conduct electricity, and are separated from the quantum dot by tunnel barriers 10 nm thick. When a negative voltage is applied to the metal side gate around the pillar, it reduces the diameter of the dot from about 500 nm to zero, causing electrons to leave the dot one at a time.

Symmetric quantum dots as artificial atoms

Many properties of quantum dots have been studied through electron tunnelling, a quantum effect that allows electrons to pass through a classically forbidden potential barrier. But electron tunnelling to and from a quantum dot is dominated by an essentially classical effect that arises from the discrete unit of charge on an electron. If the tunnelling to the dot is weak - which happens, for example, when relatively high potential barriers separate the dot from a source and drain of electrons - the number of electrons on the dot, N, is a well defined integer.

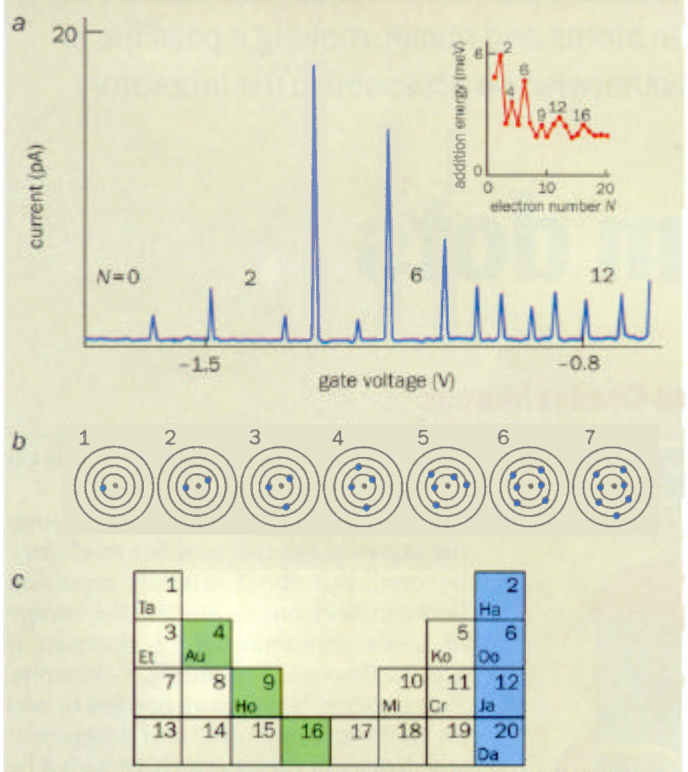
Any movement of electrons through the dot requires this number to change by one. But the Coulomb repulsion between electrons means that the energy of a dot containing N+1 electrons is greater than one containing N electrons. Extra energy is therefore needed to add an electron to the dot, and no current will flow until this energy is provided by increasing the voltage. This is known as the Coulomb blockade.

To see how this works in practice, Seigo Tarucha and colleagues at NTT in Japan and one of us (LK) and co-workers at Delft have studied what happens in a symmetric quantum-dot structure (figure 1). The structure contains a quantum dot a few hundred nanometres in diameter that is 10 nm thick and that can hold up to 100 electrons. The dot is sandwiched between two non-conducting barrier layers, which separate it from conducting material above and below. By applying a negative voltage to a metal gate around the dot, its diameter can gradually be squeezed, reducing the number of electrons on the dot - one by one - until there are none left.

This makes it possible to record the current flow as the number of electrons on the dot, and hence its energy, is varied. The Coulomb blockade leads to a series of sharp peaks in the measured current (figure 2a). At any given peak, the number of electrons on the dot alternates between N and N+1. Between the peaks, the current is zero and N remains constant. The distance between consecutive peaks is proportional to the so-called addition energy, E_{add}, which is the difference in energy between dots with N+1 and N electrons.

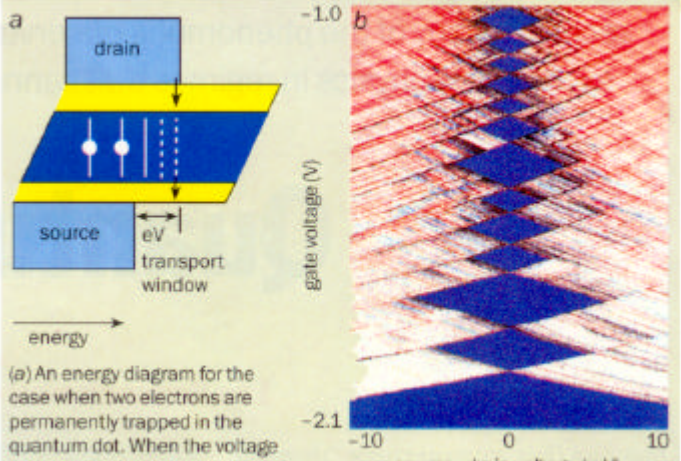
The simplest model to describe a quantum dot, the so-called constant-interaction model, assumes that the Coulomb interaction between the electrons is independent of N and is described by the capacitance, C, of the dot. In this model, the addition energy is given by $E_{add} = e^2/C + \Delta E$, where e is the charge on the electron, and ΔE is the energy difference between one quantum state and the next. Adding a single electron to the dot therefore requires a constant

2 Artificial atoms



The current flowing through the quantum-dot structure at a temperature of 0.1 K was measured as the gate voltage was varied. (*a*) The first peak corresponds to the voltage at which the first electron can enter the dot, and the number of electrons increases by one at each subsequent peak. The distance between peaks provide a measure of the addition energies (see inset). (*b*) The addition of single electrons to the quantum dot can be pictured in terms of circular orbits. The first shell can contain two electrons, the second can contain four and so on. This makes it possible to formulate a periodic table for these artificial two-dimensional atoms (*c*). Full shells correspond to the magic numbers $N = 2, 6, 12, 20$ and so on, while half-filled shells ($N = 4, 9, 16$, etc) correspond to maximum spin states. (The elements are named after team members from NTT and Delft.)

3 Excited states in quantum dots



(*a*) An energy diagram for the case when two electrons are permanently trapped in the quantum dot. When the voltage is small, electrons can only tunnel through the three-electron ground state (solid line). But at larger voltages the third electron can also tunnel through one of the excited states (dashed lines) in the transport window. (*b*) The current flow was measured as both the gate voltage and the voltage between the source and drain were varied. The vertical axis shows a change in electron number from 0 to 12, while the change in source–drain voltage opens the transport window symmetrically around zero. (A voltage of zero provides the linear response, as shown in figure 2*a*). The blue diamonds correspond to zero current, while the lines running parallel to the borders of the diamonds reflect the excitation energies of the dot.

charging energy, e^2/C, plus the difference in energy between the quantum states.

Despite its simplicity, this model is remarkably accurate and allows us to describe the measurements in more detail. The first peak on the graph marks the energy at which the first electron enters the dot, the second records the entry of the second electron and so on. But the spacings between the peaks are not constant, and significantly more energy is needed to add the second, sixth and twelfth electrons.

We can picture this in terms of two-dimensional electron orbits, since the shape of the quantum dot restricts electron motion to this plane (figure 2b). The orbit with the smallest radius corresponds to the lowest energy state. This state has an angular momentum of zero and - as with atoms - can only contain two electrons with opposite spin.

This means that the charging energy, e^2/C, is enough to increase the number of electrons on the dot from one to two. But extra energy, ΔE, is needed to add a third electron, since the innermost orbit will be full and the electron must go into a higher energy state. Electrons in this orbit have an angular momentum of ± 1 and two spin states, which means that this shell can contain four electrons. This shell will be full once the dot contains six electrons, and so extra energy is needed to add the seventh electron.

The third shell presents a special case if the confining potential is parabolic in the radial direction, because this introduces a radial quantum number. States in this shell can have an angular momentum of 0 and a radial quantum number of 1, or an angular momentum of ± 2 and a radial quantum number of 0. Together with the spin states, this means that the third shell can contain six electrons and will be full when $N = 12$.

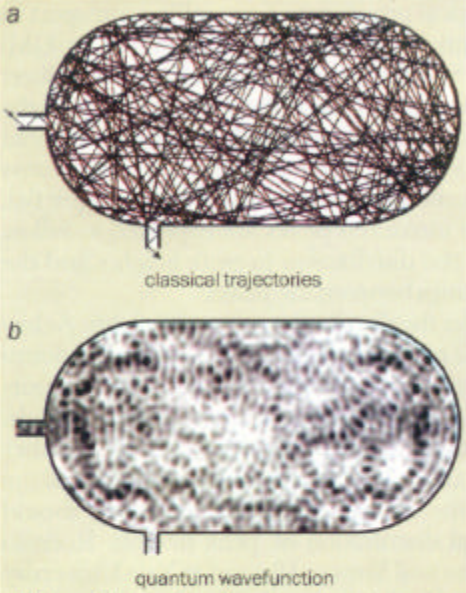
This sequence, $N = 2, 6, 12, 20$ and so on, provides the "magic numbers" of electrons in a circularly symmetric harmonic potential confined to two dimensions. The energy states for such a system were calculated in the 1920s by Charles G Darwin and independently by Vladimir Fock.

The addition energy (inset to figure 2a) also shows smaller peaks at $N = 4, 9$ and 16. This substructure reflects how the interactions between electrons can influence the filling of energy states. In atomic physics these effects are formulated as Hund's rules, which state that electrons enter a shell with parallel spins until the shell is half full, and then enter with opposite spin. The sequence N= 4, 9 and 16 corresponds to a half filling of the second, third and fourth shells, where the total spin of the electrons reaches a maximum value.

This picture is summarized in a new "periodic table" of two-dimensional elements (figure 2c). The rows are shorter than those of the familiar periodic table because the dot is defined in two dimensions rather than three.

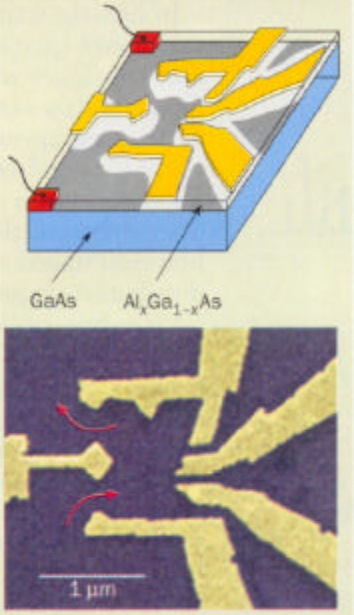
But these dots are more useful than simply providing analogues of real atoms, since their larger and more controllable size allows experiments to be carried out in regimes that cannot readily be accessed for real atoms. For example, the electron orbits in both atoms and quantum dots are altered by a magnetic field, but the effect of a I T magnetic field on a quantum dot is comparable with the effect of a one million tesla field on a real atom. Such high magnetic fields cannot be produced in the lab.

4 Classical and quantum chaos



(a) If a particle is confined in an irregular shape – i.e. not a circle or polygon – the classical bouncing-ball motion will generally lead to chaos. (b) In the same geometry for a quantum dot, a theoretical density plot of the square amplitude of a single-particle wavefunction reveals an irregular standing-wave pattern. The statistical properties of such a chaotic or disordered quantum system, including its wavefunctions and energy levels, share a number of common, or universal, features independent of the details of the particular structure. This theoretical example would roughly correspond in size to electrons on a micron-sized dot formed in a gallium arsenide heterostructure. The classical trajectory was calculated by S McDonald and A Kaufman; and the wavefunction was calculated by Richard Akis and David Ferry at Arizona State University.

5 Dots without symmetry



A lateral quantum dot is made by trapping electrons at the two-dimensional interface between gallium arsenide and aluminium gallium arsenide ($Al_xGa_{1-x}As$, with $x \sim 7\%$). Negative voltages are applied to the surface gates to confine the electrons. The gates are patterned using electron-beam lithography, then chromium and gold layers are evaporated onto the surface to form the patterns. Electrons can pass in and out of the dot through two leads (red arrows on micrograph), but a depletion region of ~ 100 nm around the gates prevents them from passing through the other gaps.

Indeed, studies of the electronic orbits of quantum dots in a magnetic field have provided further support for the Darwin- Fock energy spectrum. And at high magnetic fields it is even possible to study the quantum Hall effect in quantum dots. For example, Ray Ashoori of the Massachusetts Institute of Technology in the US has measured how the electron states evolve as the magnetic field is increased from zero into the quantum HO regime. And in 1991 Paul McEuen - then working with Marc Kastner at MIT and now at the University of California at Berkeley - showed that the simple model of constant Coulomb interactions no longer applies at high magnetic fields. In this regime, the interactions and confinement should be treated on an equal footing, and a more sophisticated calculation is needed to get the physics right.

We have so far focused on electron transport in the linear regime, where the voltage across the dot is small compared with the charging energy and the separation between quantum states. But this only allows us to study the ground-state energies of the artificial atoms. By increasing the voltage further, a "transport window" can be opened across the quantum dot, allowing excited states to contribute to electron transport (figure 3a).

This makes it possible to measure the energy spectrum of excited states (figure 3b). Such excitation spectra, together with the effect of a magnetic field, can be calculated numerically for up to six electrons. At magnetic fields of around 1 T, for example, experimental and numerical results indicate that interesting transitions occur between the energy states on the dot. These are related to changes in the exchange energy, which accounts for the interactions between electrons with parallel spins.

Loss of symmetry

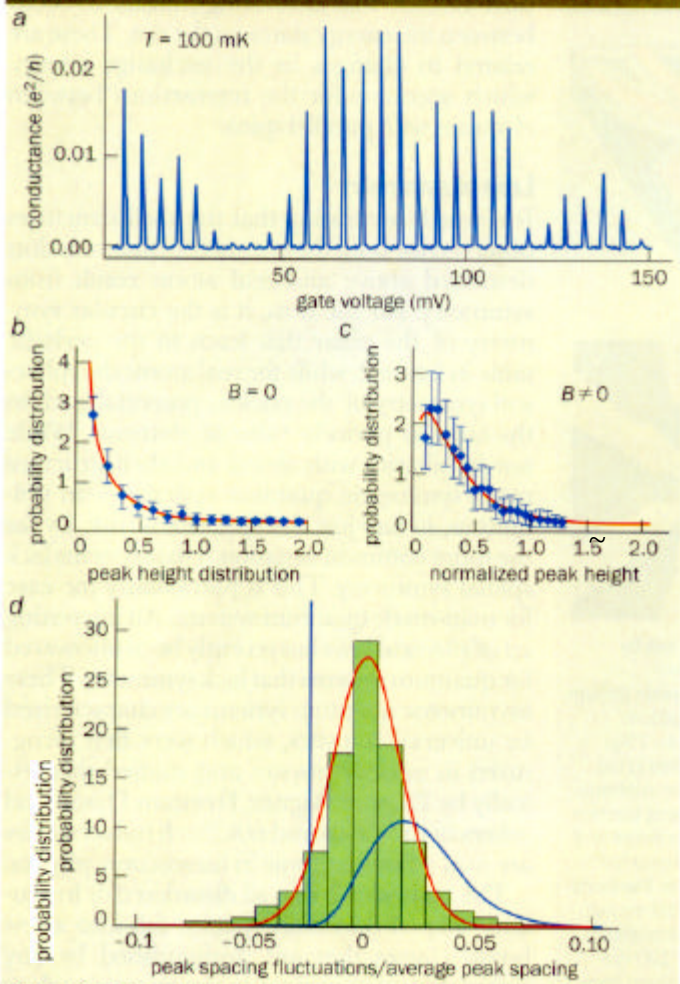
It is worth reiterating that the shell structures observed for both the symmetric quantum dots described above and real atoms result from symmetry. For the dots, it is the circular symmetry of the pillar that leads to the periodic table in figure 2, while for real atoms the spherical symmetry of the nuclear potential leads to the familiar periodic table of elements. While our familiarity with atoms and shell structure make symmetric quantum systems seem ubiquitous, in fact just the opposite is true. By far the more common situation is that systems lack spatial symmetry. This is particularly the case for man-made quantum systems. An interesting set of physical laws has recently been uncovered for quantum systems that lack symmetry. These asymmetric quantum systems are characterized by universal statistics, which were first recognized in nuclear physics and studied theoretically by Eugene Wigner, Freeman Dyson and others in the 1950s and 60s. Such universalities are now a familiar topic in mesoscopic physics.

The basic idea is that all disordered or irregularly shaped quantum systems fall into a few broad classes that are distinguished by any symmetries that remain in the system, such as symmetry under time-reversal. The quantum systems within each class share various statistical properties that describe, for example, their energy-level structure or scattering behaviour.

A surprising aspect of the universal statistics relating to quantum systems is that they seem to be connected with chaotic dynamics observed in the equivalent classical system. For example, the famous "stadium billiard" (a two-dimensional region bounded by hard walls) is a simple shape, consisting of semicircles connected by straight edges. Nonetheless, the classical trajectory of a ball bouncing in a stadium produces random motion for almost any initial condition, as proved by Leonid Bunimovich (figure 4a). The corresponding quantum system (figure 4b) has wavefunctions and energy levels that share universal features with other classically chaotic systems. This example has since become a mainstay of research into the quantum aspects of classical chaos, a subject now known as "quantum chaos".

It is presumably the same universal behaviour of quantum chaos that leads to the random, but reproducible, fluctuations that are observed in the conductance of micron-scale metals at low temperatures. Such "mesoscopic" systems are small enough to exhibit quantum interference, but large enough to contain a random distribution of scatterers such as impurities and dislocations. These scatterers act as a source of chaos, causing electrons to move along random paths that depend on the initial conditions, just like a ball bouncing in a pinball machine.

6 Universal statistics in a quantum dot



(a) In an irregular quantum dot, the peaks due to Coulomb blockade show random fluctuations in height. These are caused by variations in the coupling between the wavefunction in the dot and the single-channel leads. (*b*) The experimentally measured probability distribution of normalized peak heights (blue diamonds) shows excellent agreement with the theoretical distribution (red line). (c) The distributions in (*b*) are sensitive to the breaking of time-reversal symmetry, which occurs in the presence of a magnetic field. (*d*) A simple (but, it turns out, incorrect) theoretical model for the peak spacing (blue line) can be obtained by adding a smoothly varying charging energy to the well known Wigner–Dyson distribution for the spacing of quantum energy levels. This leads to a bimodal distribution, where the single spike is due to the filling of even-numbered electrons and the broad, continuous part is due to the filling of odd-numbered electrons. However, the experimental data (green bars/red line) show no bimodal structure.

In the case of quantum dots, however, chaos is not generated by scatterers, since the mean free path for elastic scattering is several times larger than the dot. Although the interactions between electrons - just like those between nucleons in the nucleus - can lead to quantum chaos and universal statistics, experiments to study these effects do not only rely on these interactions to generate chaos. Instead, the dots are designed so that their shape lacks all spatial symmetry.

In practice, these dots can readily be fabricated by laterally confining a two-dimensional sheet of electrons created at the interface between layers of gallium arsenide and aluminium gallium arsenide (figure 5). Several metal gates are used to confine the electrons in an irregular shape, and individual electrons can enter or leave the dot through two tunnelling barriers or "leads". Both the shape of the dot and the movement of electrons through the leads can be controlled by altering the gate voltages. Dots of this type can contain any- thing from a few to several thousand electrons.

In 1996 the statistical properties of tunnelling transport in this type of quantum dot were measured by one of us (CNI) and colleagues at Stanford, and independently by Albert Chang and co-workers at Bell Labs, New Jersey, US. As in the earlier experiments, the movement of electrons is dominated by the Coulomb blockade. The conductance therefore shows a series of peaks as the gate voltage is increased (figure 6a). Two aspects of the measured peaks are expected to follow universal statistics: the distribution in peak heights and the distribution of spacings between the peaks.

Let us first consider the distribution of peak heights. At low temperatures (- 100 millikelvin), the heights of the Coulomb peaks are inversely proportional to temperature and proportional, on average, to the series conductance of the two leads. However, changes in the shape of the dot - or in the number of electrons it contains - lead to random fluctuations in the coupling between the dot and the leads, and these should result in a universal distribution of peak heights. Rodolfo Jalabert, Doug Stone and Yoram Alhassid at Yale University in the US calculated this distribution in 1992, using ideas that lead to the well known Porter-Thomas distribution of scattering widths observed in compound nuclear scattering.

Measurements of the distribution in peak heights can be made by altering the shape of the quantum dot to collect data on large numbers of different - but similar - quantum systems. Distributions measured in this way agree well with theory (figures 6b). As long as the dot generates chaotic dynamics, the distribution is independent of the size, shape and transmission of the single-channel leads. It only changes in the presence of a magnetic field, since this breaks the symmetry under time-reversal, and here again the theory matches the experimental results (figure 6c).

What about the spacing of the Coulomb peaks? A simple universal prediction can be made for any chaotic quantum dot by simply applying the approach taken earlier for symmetric dots. In this case the charging energy needed to add electrons to the dot is simply added to the universal distribution for spacings between quantum levels, which is well known from other quantum systems. Indeed, these so-called Wigner-Dyson distributions are the most famous example of the universality of quantum chaos, and seem to describe everything from the spacing between resonances in com- pound nuclear scattering to single-particle energy level spacings of classically chaotic dynamical systems.

According to the charging-energy model, the distribution of peak spacings should follow a Wigner-Dyson distribution with a width that depends on the properties of a particular dot. This theoretical distribution shows two main features: a single spike due to even-numbered electrons that only require a charging energy to enter the dot, and a broader peak due to odd-numbered electrons that require extra energy to enter the next quantum level (figure 6d).

However, this simple scheme requires a series of question- able assumptions. It supposes that the charging effects can be described by a single, slowly varying charging energy. It also assumes that the electron spins dictate how the energy levels are filled, and that fluctuations in the capacitance of the dot are small. This last assumption has been called into question by Uri Sivan and colleagues at the Technion in Israel, who have investigated the peak spacings in quantum dots and wires.

Based on experimental data and numerical work with Richard Berkovits at Bar-Ilan University in Israel, Sivan and colleagues find that changes in capacitance can significantly contribute to, and in some cases dominate, the peak-spacing statistics.

Indeed, the measured distribution of the peak spacings - obtained by the Stanford group for about ten thousand peaks - disagrees with the simple theoretical model (figure 6d). In particular, the distribution shows no hint of the even-odd features seen in the predicted distribution, challenging the assumption that each level fills first with one spin then the other before the next level is occupied. Further work is needed to understand how the spin, Coulomb interactions and chaotic wavefunctions combine to produce this measured distribution.

Several theorists, in particular Michael Stopa, while at Riken in Japan, Berkovits, and Ned Wingreen and Kenji Hiroshi at the NEC Research Center in Princeton, US, have recently approached the problem numerically, allowing the possibility of more complicated arrangements of occupations, and that the total spin may minimize the energy of the dot. But it remains to be seen whether there are some universal statistics for these interacting systems. Recent experiments by Dan Ralph and Michael Tinkham at Harvard University in the US also emphasize the importance of spin and inter- action in the properties of quantum dots. In their case, the dots are made of aluminium, which leads to superconductivity effects at low temperatures and magnetic fields.

Future horizons

Much of the physics observed in quantum dots has also been revealed in other quantum systems. For example, the move- ment of single electrons has recently been measured in carbon nanotubes and other molecular systems, and shows similar features to those observed for quantum dots. These similarities suggest that the physics of quantum dots applies to many other systems containing confined electrons.

Meanwhile, the continual development of nanotechnology will allow a greater range of artificial quantum structures to be studied. More refined theories of quantum transport will be needed to consider not only the quantization of energy and charge, but also the interactions between confined and non-confined electrons.

Some work has already started in this direction. For ex- ample, a collaboration between MIT and the Weizmann Institute in Israel and also the group in Delft with one of us (LK) has recently used quantum dots as artificial magnetic impurities embedded between metallic leads. If the dot contains an odd number of electrons, the total spin will be non- zero and electron tunnelling between the dot and the leads changes the spin on the dot. The coupling between the dot and leads therefore has the effect of a spin-exchange coup- ling. Experiments in this regime have shown new transport features that resemble the Kondo effect in metals containing magnetic impurities.

While the Kondo effect owes its existence to interactions between many electrons, interactions can actually destroy other quantum transport phenomena. For instance, in the case of more complex circuits, the question arises whether electrons can retain their quantum mechanical properties, or whether interactions with the outside environment will lead to phase decoherence. In the last few years Moty Heiblum's group at the Weizmann Institute has carried out an interesting series of experiments aimed at measuring these coherence properties, and has observed, for instance, how placing a micro-detector near one arm of the interferometer causes decoherence. More work along these lines needs to be done before we can know if it is possible to use quantum dots as the building blocks for quantum circuits.

This is a question with practical importance, since it has been suggested that such quantum circuits could form the basis of a quantum computer. However, it will take many more fundamentally interesting experiments before we get to such practical applications.

Further reading

R CAshoori 1996 Electrons in artificial atoms Nature 379413

CWJ Beenakker 1997 Random-matrix theory of quantum transport Rev. Mod. Phys. **69** 731; cond-mat/9612179

TGuhr, A Mueller-Groeling and HA Weidenmueller 1998 Random matrix theories in quantum physics: common concepts *Phys. Rep.;* cond-mat/9707301 at press

L P Kouwenhoven et al. 1997 Electron transport in quantum dots Mesoscopic *Electron Transport* led) L L Sohn, L P Kouwenhoven and G Schoen (NATO Series, Kluwer, Dordrecht) See also: http://vortex.tn.tudelft.nV-Ieok/papers/ and http-.//www-leland.stanford.edu/group/MarcusLab/grouppubs.html

P L McEtien 1997 Artificial atoms: new boxes for electrons science **273** 1682

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