changes during the transport cycle. In addition, the BtuCD structure clears the way to structural studies of intermediate states, which should further advance our understanding of the translocation mechanisms.

References and Notes
1. See http://ir2lcb.cnrs-mrs.fr/ABCdb/.
10. J. E. Walker et al., EMBO J. 1, 945 (1982).
27. Materials and methods are available as supporting material on Science Online at www.sciencemag.org/cgi/content/full/296/5770/1091/DC1.
57. Single-letter abbreviations for the amino acid residues are as follows: A, Ala; C, Cys; D, Asp; E, Glu; F, Phe; G, Gly; H, His; I, Ile; K, Lys; L, Leu; M, Met; N, Asn; P, Pro; Q, Gln; R, Arg; S, Ser; T, Thr; V, Val; W, Trp; and Y, Tyr.
58. See www.din3d.org.
60. We thank the staff at the Stanford Synchrotron Radiation Laboratory (SSRL), the Advanced Photon Source (APS), the Advanced Light Source (ALS), and the National Synchrotron Light Source (NSLS) for their support during crystal screening and data collection. We also thank M. Barclay, R. Bass, O. Einsle, I. Locher-Hinderling, and P. Strop for helpful discussions. We further thank M. Day for assistance in obtaining the cyclotetrahidrate and L. Ackerman and J. Bercaw for their help in the synthesis of inorganic clusters during the search for derivatives. The coordinates of the BtCD transporter with bound cyclotetrahidrate have been deposited in the Protein Data Bank (www.rcsb.org/pdb) with access code 1L7V.

REPORTS

**Scanned Probe Imaging of Single-Electron Charge States in Nanotube Quantum Dots**

Michael T. Woodside and Paul L. McEuen

An atomic force microscope was used to study single-electron motion in nanotube quantum dots. By applying a voltage to the microscope tip, the number of electrons occupying the quantum dot could be changed, causing Coulomb oscillations in the nanotube conductance. Spatial maps of these oscillations were used to locate individual dots and to study the electrostatic coupling between the dot and the tip. The electrostatic forces associated with single electrons hopping on and off the quantum dot were also measured. These forces changed the amplitude, frequency, and quality factor of the cantilever oscillation, demonstrating how single-electron motion can interact with a mechanical oscillator.

Single-electron charging phenomena are ubiquitous in atoms, molecules, and small electronic devices, and their effects are central to an understanding of the physics and technology of nanoscale systems. Single-electron effects arise because the number of electrons residing on a small, quasi-isolated, conducting island is quantized. Adding an additional charge to such a quantum dot costs an electrostatic energy on the order of $U = e^2/C$, where $C$ is the capacitance of the dot and $e$ is the electronic charge ($e$). This charging energy suppresses charge transport when $U \gg k_BT$, where $k_B$ is the thermal energy, leading to the Coulomb blockade of charge motion on and off the dot.

Although Coulomb blockade phenomena have been studied extensively with transport measurements (2), such measurements lack the spatial discrimination necessary to probe the interior of a dot or to probe complex multidot systems. An alternative approach is to detect single-charge motion using scanned probe techniques, such as scanned capacitance microscopy (3, 4), scanned single-electron transistors (5, 6), and atomic force microscopy (AFM). The first two of these have excellent charge sensitivity but are technically very difficult; moreover, they are not easily able to image the topography of the device under study. AFM-based techniques, on the other hand, can be used both to image the sample and to interact with it in a variety of ways. For example, electrostatic force microscopy (EFM), which measures the electrostatic force between a sample and a metallized AFM tip, has been used to detect the motion of single charges during contact electrification of insulating surfaces (7) and to image the potential profile in carbon nanotubes (8). In addition, scanned gate microscopy (SGM), in which the AFM tip is used to perturb the conducting properties of a sample, has been used to image electron trajectories and scattering centers in two-dimensional electron gases (9–11) and barriers in carbon nanotubes (8, 12, 13).

R E S E A R C H A R T I C L E S
We describe experiments that extend these AFM-based techniques into the single-electron regime, by using a low-temperature AFM (14) to perform single-electron scanned gate microscopy (e-SGM) and single-electron electrostatic force microscopy (e-EFM). The samples we study are individual single-walled carbon nanotubes grown by chemical vapor deposition (15) on a backgated Si substrate and attached to electrical contacts (16). Tunnel barriers arising from defects and/or imperfect electrical contacts to the nanotube define quantum dots in the tube (13, 16, 17). The charge state of a dot is controlled by changing either the position or the voltage of the AFM tip relative to the dot (Fig. 1, top). This charge state is then probed by monitoring the conductance of the dot (e-SGM) or the mechanical oscillation of the AFM cantilever (e-EFM). These measurements allow us to explore not only the local properties of nanotube quantum dots but also the influence of single-electron motion on a mechanical oscillator.

We begin by discussing e-SGM, in which the charge state of a quantum dot is sensed by transport measurements. An e-SGM measurement of the conductance of a metallic nanotube device as a function of the tip voltage \( V_{\text{tip}} \) is shown in Fig. 1A, made at \( T = 0.6 \) K when the position of the AFM tip is fixed 120 nm above the tube (18). The sharp peaks observed in the conductance are the Coulomb oscillations that occur at low temperature (\( k_B T \ll U \)) each time a single electron is added to a dot in the nanotube (1). These Coulomb oscillations arise because the tip voltage would like to induce a continuous charge \( q \) (the “control charge”) on the dot, but the charge \( e \) that can actually be transferred to the dot is quantized. This produces periodic steps in the occupancy of the dot as a function of the gate voltage (Fig. 1A), resulting in peaks in the conductance each time an electron is added to the dot.

The control charge \( q \) induced on the dot by the AFM tip is given by:

\[
q(r, \Delta V) = C_{\text{sd}}(r) \cdot \Delta V
\]

where \( C_{\text{sd}}(r) \) is the tip-dot capacitance as a function of the tip position \( r \), and \( \Delta V = V_{\text{tip}} - V_{\text{dot}} - \Phi_r \) is the electrostatic potential difference between the tip and the dot (\( \Phi_r \approx 0.2 \) to 0.3 V is the work function difference between the tip and the dot). The conductance of the dot can thus be changed not only by varying \( V_{\text{tip}} \) but also by moving the tip over the sample. Imaging the conductance of the device as a function of the tip position in \( (r, x) \) for fixed \( V_{\text{tip}} = -200 \) mV (Fig. 2A), we observe two sets of concentric rings of conductance peaks, centered at different locations on the nanotube. Each ring in the image corresponds to a single Coulomb oscillation on the dot enclosed by the ring: it is the locus of tip positions that correspond to the particular control charge producing the conductance peak. As the tip approaches the dot, \( C_{\text{sd}} \) grows, causing the negative tip bias to push electrons off the dot one by one, producing successive concentric rings.

The two sets of rings in Fig. 2A indicate that there are two quantum dots in series along the tube, located at the center of each set of rings. A defect or local potential perturbation in this nanotube apparently creates a tunnel barrier that breaks the tube into two dots (13, 19). The size of the elliptically shaped Coulomb oscillations for the dot on the right side of Fig. 2A indicates a dot of length \( L \approx 1.5 \) \( \mu \)m, which corresponds well to the length of 1 to 2 \( \mu \)m that is inferred from the data in Fig. 1A (16, 17). The rings around the dot on the left side are further apart and nearly perfectly circular, indicating that this dot is significantly smaller (\( L \approx 0.7 \) \( \mu \)m). These e-SGM images thus give valuable spatial information about the location and size of the quantum dots formed in nanotubes.

We next consider e-EFM measurements, in which we measure the dynamical response of the AFM cantilever to the force from single-electron motion. The cantilever responds to an external driving force \( F_{\text{ext}} \), as a damped simple harmonic oscillator (20).
force driving the cantilever in e-EMF is the electrostatic force exerted on the tip by a single electron moving on/off the quantum dot. This charge motion produces a change $U/e$ in the electrostatic potential of the dot, which in turn exerts a force $F_e$ on the tip (21)

$$F_e = C_{ud}(\Delta V \cdot U/e)$$

where $C_{ud} = dC_{ud}/dz$ is the derivative of the tip-dot capacitance. For typical parameters ($\Delta V \sim 0.5$ V, $U/e = 2$ mV, and $C_{ud} = 3 \times 10^{-11}$ F/m (22)), we expect $F_e \sim 30$ nN. This force is two orders of magnitude larger than the sensitivity of the AFM on resonance ($F_{\text{res}} \sim 0.3$ fN/√Hz (14, 21)), hence single-electron forces should be readily detectable (23).

To measure the single-electron force, we push an electron on/off the dot at the resonant frequency $\omega_0$ of the cantilever by oscillating at $\omega_0$ either the voltage between the tip and the dot or the height of the tip above the dot. A control charge modulation of $\delta Q(\omega_0)$ changes the occupancy of the dot by $dQ(\omega_0) = (dQ/dq)(\delta Q(\omega_0))$, where $q$ is the probability that the dot is occupied (24), producing a force

$$F(\omega_0) = (d/dq)(\delta Q)(\omega_0)F_e$$

For a nanotube quantum dot of length $1 - 3$ μm, we estimate $\Gamma \sim (10^{-3} - 10^{-4}) S$, where $S$ is the transmission coefficient for tunneling off the dot. The ratio $\alpha_0/\Gamma \sim 10^{-7}/3$ is very small, except for very opaque barriers.

The real part of the force in Eq. 4 corresponds to an additional effective spring constant for the cantilever $\delta k$, producing a resonant frequency decrease $\delta \omega_0 = \delta k/2k$. Physically, as the tip approaches the sample during each cycle of the oscillation, the additional charge induced on the dot pulls the tip toward the dot, slowing down the oscillation and reducing $\omega_0$. The magnitude expected for typical parameters is $(\delta \omega_0/\omega_0)_{\text{max}} \sim 5 \times 10^{-6}$, in good agreement with the observed values.

The imaginary part of the force in Eq. 4 corresponds to an effective damping term, producing a decrease in $Q$ by $\delta Q/Q \sim (1/Q)/\Gamma$. Physically, this additional energy dissipation is due to the resistive losses from single electrons moving on and off the dot during the cantilever oscillations. It is a manifestation of the well-known fluctuation-dissipation theorem, which requires that the energy delivered to the cantilever by the fluctuating single-electron forces be balanced by additional dissipation. The single-electron dissipation depends inversely on $S$, becoming maximal when the time for hopping on/off the dot is comparable to the oscillation frequency of the cantilever ($\Gamma \sim 10^{-7}$). This hopping rate can vary by orders of magnitude from one dot to the next (or even from one Coulomb oscillation to the next in the same dot). There are therefore large variations in the magnitude of the $Q$ degradation signal for different dots or even for different Coulomb oscillations in the same dot (Fig. 1D) (26).

Just as with e-SGM, e-EMF can also be used to make images of the charge state of a quantum dot as a function of the tip position. In Fig. 2B, the single-electron force is measured on a different nanotube by applying an ac voltage $V(\omega_0)$ to the sample electrodes. Concentric rings of peaks in the force on the AFM tip are observed, centered on the nanotube. As with the rings in the conductance in Fig. 2A, each ring corresponds to a change of a single electron in the charge state of the quantum dot at the center of the rings. Figure 2C shows a measurement of the $Q$ degradation as a function of tip position for yet another nanotube. Two sets of well-
Creation and Manipulation of Three-Dimensional Optically Trapped Structures

M. P. MacDonald,1 L. Paterson,1 K. Volke-Sepulveda,2 J. Arlt,3 W. Sibbett,1 K. Dholakia1**

An interferometric pattern between two annular laser beams is used to construct three-dimensional (3D) trapped structures within an optical tweezers setup. In addition to being fully translatable in three dimensions, the trapped structure can be rotated controllably and continuously by introducing a frequency difference between the two laser beams. These interference patterns could play an important role in the creation of extended 3D crystalline structures.

At a microscopic level, transparent objects can be trapped and manipulated using the forces exerted by a tightly focused laser beam. This technique, known as “optical tweezers” (1, 2), has enabled major advances in numerous areas of science, including force detection measurements on biological samples, such as the determination of the elastic response of DNA (3). Recent work has also demonstrated the use of optical tweezers for developing optical microscopy and micro-components (4–8). In parallel with this, the extension of optical tweezers to multiple beam sites to create two-dimensional particle arrays (9, 10) has been investigated. We take this technique a step further by creating vertical arrays of particles (stacking) in multiple trapping sites, forming the basis for creating 3D trapped structures.

Stacking of a small number of particles in standing-wave geometries (11) and Bessel light beams (12) has been observed experimentally, whereas Gauthier and Ashman theoretically predicted stacking in a Gaussian beam (13). Experimentally, we have observed controlled stacking of large numbers of particles in optical tweezers using a single Gaussian beam. By extending this to multiple trapping sites, formed in the interference pattern generated between two annular (Laguerre-Gaussian) light beams, we have created 3D trapped structures. Furthermore, we use the angular Doppler effect to achieve continuous and controlled rotation of the 3D structure.

The mechanism for creating particle stacks...