Energy levels of few-electron quantum dots imaged and characterized by atomic force microscopy

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Strong confinement of charges in few-electron systems such as in atoms, molecules, and quantum dots leads to a spectrum of discrete energy levels often shared by several degenerate states. Because the electronic structure is key to understanding their chemical properties, methods that probe these energy levels in situ are important. We show how electrostatic force detection using atomic force microscopy reveals the electronic structure of individual and coupled self-assembled quantum dots. An electron addition spectrum results from a change in cantilever resonance frequency and dissipation when an electron tunnels on/off a dot. The spectra show clear level degeneracies in isolated quantum dots, supported by the quantitative measurement of predicted temperature-dependent shifts of Coulomb blockade peaks. Scanning the surface shows that several quantum dots may reside on what topographically appears to be just one. Relative coupling strengths can be estimated from these images of grouped coupled dots.

nanoelectronics | single-electron charging | shell structure | electrostatic force microscopy

The ability to confine single charges at discrete energy levels makes semiconductor quantum dots (QDs) promising candidates as a platform for quantum computation (1, 2) and single-photon sources (3). Tremendous progress has been made not only in understanding the properties of single electrons in QDs but also in controlling their quantum states, which is an essential prerequisite for quantum computation (4). Single-electron transport measurements have been the main experimental technique for investigating electron tunneling into QDs (5). Charge sensing techniques using built-in charge sensors, such as quantum point contacts (6), complement transport measurements because lower electron tunneling rates can be monitored with even real-time detection being possible (7). It is instrumentally challenging to study self-assembled QDs via conventional transport and charge sensing methods because of the difficulty in attaching electrodes. Although progress is being made (8–12), these techniques have very small yield and therefore make it difficult to assess variation in QD electronic properties. Compared to typical QDs studied via transport measurements, in particular lithographically defined QDs, self-assembled QDs can be fabricated to have smaller sizes, stronger confinement potentials, and a more scalable fabrication process, all of which make them attractive for practical applications.

In this paper, we focus on an alternative technique for studying QDs that is better suited for self-assembled QDs: charge sensing by atomic force microscopy (AFM). Charge sensing by AFM is a convenient method to study the electronic structure of QDs because nanoelectrodes are not required and large numbers of QDs can be investigated in one experiment. Termed single-electron electrostatic force microscopy (e-EFM), this technique relies on the high force sensitivity of AFM to detect the electrostatic force resulting from single electrons tunneling into and out of the QD. It was first demonstrated on QDs formed in carbon nanotubes (13, 14) and later applied to self-assembled QDs (15, 16) and also to gold nanoparticles (17, 18). We focus on epitaxially grown self-assembled InAs/InP QDs in the few-electron regime. By using a dissipation model, we find compelling evidence for the existence of electronic degeneracies (i.e., shell structure) by measuring an effective temperature-dependent level repulsion of Coulomb blockade peaks in the AFM cantilever dissipation. This repulsion is a manifestation of the asymmetry between adding or removing electrons to or from a degenerate level on the dot; whereas similar effects were predicted for the conductance through a QD (19), we believe this to be a unique quantitative measurement. Further, we use the model to quantitatively extract various properties of both individual QDs, such as the tunneling rates and charging energy, and coupled QDs, such as the strength of coupling.

We study uncapped self-assembled InAs QDs grown on a 20-nm InP tunnel barrier, below which a two-dimensional electron gas (2DEG) is formed in an In0.53Ga0.47As quantum well. A dc-bias voltage, $V_B$, is applied to the 2DEG with respect to the grounded conductive AFM cantilever tip. Fig. 1A shows the sample structure and experimental setup. The AFM cantilever is driven at its mechanical resonance frequency, $\omega_0/2\pi \approx 166$ kHz, with constant oscillation amplitude (20). The voltage drop, $\alpha V_B$ (\(\alpha < 1\)), across the tunnel barrier between the QD and the 2DEG is only a fraction of $V_B$ with $\alpha = \alpha(xyz)$ being a function of the tip position. The tip–QD gap is tens of nanometers wide so that tip–QD tunneling is negligible. We thus have a single-electron box setup: The electrochemical potential of the 2DEG, $\mu_{\text{2DEG}}$, with respect to the QD, $\mu_{\text{QD}}$, is set by $\alpha V_B$ and a negative bias increases the number of electrons, $N$, on the QD in integer steps whenever the electrochemical potentials are aligned (called a charge degeneracy point). Tunneling between 2DEG and QD is suppressed by the electrostatic energy cost, $E_C$, of adding or removing an electron to the QD except near these charge degeneracy points (Coulomb blockade). The heart of the e-EFM technique lies in the fact that oscillations of the AFM cantilever modulate $\alpha$ and hence are equivalent to an effective oscillating gate voltage applied to the QD. Thus, motion of the cantilever induces a modulation of $N$ that will be slightly out of phase with the cantilever’s motion (a result of the finite response time of electrons on the dot). The electrostatic coupling between QD and cantilever tip implies an electrostatic force proportional to $N$ acting on the tip, the net result being both a frequency shift, $\Delta \omega$, and additional dissipation, $\gamma$, of the cantilever (21). These effects are maximal at charge degeneracy points because here $N$ can easily change in response to the effective oscillating gate voltage.


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Fig. 1B is an example of $\gamma(V_B)$ at 4.5 K with the tip positioned over the upper QD imaged in Fig. 1 D–E. The $\gamma(V_B)$ is equivalent to the energy addition spectra usually obtained from linear conductance or capacitance spectroscopy measurements (22). Similar to those measurements, Coulomb blockade peaks in $\gamma$ occur near charge degeneracy points of the QD. The peak positions are suggestive of the addition spectrum of a 2D circular QD with a parabolic confinement potential; each peak is separated by twice the capacitive charging energy, $2E_C$, with a further splitting between peaks 2 and 3 and between peaks 6 and 7 corresponding to the energy difference between shells, $\Delta E$. This type of shell structure has been previously observed in InAs QDs (8, 9, 23, 24). Fig. 1 C–E shows the topography, $\Delta \omega$, and $\gamma$ images of an elongated InAs island. The peaks in the $\gamma-V_B$ spectra radially surround the QD center so that the ring furthest from the center corresponds to the first electron entering the QD; the rings themselves are contour lines of constant $\alpha V_B$. Multiple sets of concentric rings appearing in the $\Delta \omega$ and $\gamma$ images indicate multiple QDs in the island. Such observations would not be as easily identified via other experimental means (25). The tip–2DEG capacitive force adds a large background in the signal that locally varies because of topography (15, 26), making it advantageous to focus on the $\gamma$ for image analysis.

Fig. 2 A shows $\gamma$ and $\Delta \omega$ as a function of $V_B$ with the tip positioned over the upper QD imaged in Fig. 1 D–E. The $\gamma$ as 30 K. Whereas the energy addition spectra shown in Figs. 1B and 2A are consistent with the expected shell structure for a 2D circular QD with a parabolic confinement potential, we obtain much stronger evidence of the shell structure from the temperature dependence of the peak positions. Theoretically, temperature-dependent shifts of Coulomb blockade peaks are expected whenever one has degenerate single particle levels, as predicted for the conductance peaks of a spin degenerate level (19). These shifts are a consequence of the asymmetry between adding or removing electrons; the direction of each peak shift versus $V_B$ depends on whether there are more ways to add or to remove an electron. These asymmetries for a twofold degenerate s shell and a fourfold degenerate p shell (expected for a circular QD) are illustrated in Fig. 3. The size of each shift is proportional to temperature, which enters through the Fermi distribution of electrons in the 2DEG, with a numerical prefactor that depends on how asymmetric the addition and removal processes are. The result is an effective temperature-dependent energy level repulsion: The peaks in each shell move apart as temperature is increased. Furthermore, our theoretical analysis (SI Text) suggests that this effect should be enhanced in the tunneling-induced cantilever dissipation compared to the conductance. Our measurements of these temperature-dependent peak shifts are in excellent agreement with theory; we are unaware of any experiments where these effects have been observed.

We model the dissipation on the cantilever by using linear response and a master equation describing the charge state of the QD in the regime of weak coupling (19, 27). Details of the approach are provided in SI Text. Near a charge degeneracy point between $N$ and $N + 1$ electrons on the QD where the extra electron occupies a nondegenerate single particle level, the dissipation is (18, 28)

$$\gamma = \frac{\alpha_0^2 A T}{k_B T} \frac{1}{\omega^2 + \Gamma f(1 - f)},$$

where $\alpha_0$ and $k_B$ are the intrinsic cantilever resonance frequency and spring constant, respectively, $\omega = \alpha_0 + \Delta \omega$ is the measured resonance frequency because of forces on the cantilever, and $f = 1/(1 + \exp(E/k_B T))$ is the Fermi function evaluated at $E = \mu_{QD} - \mu_{2DEG} = eV_B (V_B > 0)$ ($V_B = -V_0$ is the point of charge degeneracy). The temperature, Boltzmann constant, and electron charge are described by $T$, $k_B$, and $e$, respectively. In the nondegenerate case, the rate to add an electron to the QD is $\Gamma = \Gamma_0$ and the rate to remove an electron is $\Gamma = \Gamma (1 - f)$, where $\Gamma$ is the 2DEG–QD tunneling rate. Last, $A = -2E_C \frac{e^2}{\pi} (1 - d) \frac{\alpha}{\pi}$ is the sensitivity of the potential on the QD to the cantilever motion and $C_{tip}$ is the tip–QD capacitance. We stress that Eq. 1 applies to each dissipation peak independently: $\Gamma$ and $A$ are obtained separately for each peak from the data with no assumption of constant $E_C$.

Whereas it describes the broadening of each peak very well, Eq. 1 only takes into account a single nondegenerate level. More generally, suppose the QD is occupied by $N + 1$ electrons, with $n_{shell} + 1$ in the valence shell. If this shell is $\nu$-fold degenerate, then near the charge degeneracy point between $N$ and $N + 1$ electrons on the QD, the dissipation is

$$\gamma(V_B) = \frac{\alpha^2 A T}{k_B T} \frac{1}{\omega^2 + (\phi f)\nu} \left[ (1 - f) \right],$$

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and \( \Gamma \) is the tunneling rate to one particular state on the QD, assumed to be equal for each degenerate state within the \( \nu \)-fold degenerate shell.\(^*\) Note that \( n_{\text{shell}} \) is the number of electrons occupying the given shell and not the total number of electrons on the dot, \( N \). Because of the factor \( \phi, \gamma(V_B) \) is no longer symmetric about its maximum, and the different coefficients of \( f \) and \( 1-f \) in \( \phi \) reflect that the rates to add and remove electrons are now asymmetric: \( \Gamma_+ = (\nu - n_{\text{shell}}) \Gamma f \) and \( \Gamma_- = (n_{\text{shell}} + 1) \Gamma (1-f) \). Note that a nondegenerate level corresponds to \( \nu - n_{\text{shell}} = n_{\text{shell}} + 1 = 1 \), for which Eq. 2 reduces to Eq. 1. The asymmetry in Eq. 2 causes each peak in \( \gamma(V_B) \) to be shifted by an amount proportional to temperature, because a dissipation peak maximum roughly corresponds to maximal tunneling on and off the dot, and this is achieved when the addition and removal rates are equal.\(^1\) The connection between the peak shifts and asymmetric addition and removal rates is depicted schematically in Fig. 3. By fitting \( \gamma(V_B) \) (e.g., Fig. 2A) to Eq. 2, we extract \( \alpha \), allowing us to convert the \( V_B \) axis into energy. This is done for all of the thermally limited peaks, yielding \( \alpha = 0.036 \pm 0.003 \). Fig. 2B shows \( \gamma(V_B) \) at different temperatures together with the fitted curves.

The role of \( \phi \) is further elucidated by the relation

\[
\phi \Gamma = -2 \alpha \omega_0 \frac{\Delta \omega_{\text{dip}}}{\gamma},
\]

where \( \Delta \omega_{\text{dip}} \) is the size of the frequency shift dip because of the single-electron tunneling. The ratio in Eq. 4 defines an inverse time scale set by the relative in-phase and out-of-phase parts of the electrostatic force; this is simply \( \Gamma \) for a nondegenerate QD (18, 28) but modified by degeneracy through the factor \( \phi \). By using Eqs. 2 and 4 and the measured values of \( \gamma \) and \( \Delta \omega_{\text{dip}} \), we calculate the tunneling rates at the maxima of dissipation peaks 1–6, obtaining \( \Gamma/2\pi = 70, 90, 160, 180, 230, \) and 330 kHz. As expected, \( \Gamma \) increases with increasing \( V_B \) as the height of the potential barrier between the 2DEG and the QD is reduced.

After extracting the tunneling rates, we fit each dissipation peak by using Eq. 2 and measure the spacing between peaks as functions of temperature from 4.5 to 30 K and from 78 to 95 K. We focus on the relative shifts between peaks, because these are less sensitive than the absolute peak positions to slight offsets because of small changes in the tip–QD distance. The size and direction of each peak shift is different (see Fig. 2A), in a manner that is completely captured by our model: The two peaks in the s shell shift apart, as do the four peaks in the p shell. The measured relative peak shifts of repelling pairs are shown in Fig. 2C–D and compared to the theoretical shifts from Eq. 2, where the sole fit parameter is the \( \gamma \) intercept corresponding to \( 2E_C \). In addition, we expect that multiple shells, not just the valence shell, \( s \) and \( p \) shells toward each other that we believe is a consequence of strong repulsion of the \( p \) shell by the \( d \) shell, predicted to be sixfold degenerate.

The agreement between the data and theory shown in Fig. 2C–D is strong evidence that the peak shifts are because of level degeneracy in the QD. We performed several additional checks to support this conclusion. First, our explanation of the observed peak shifts requires only approximate level degeneracies, and the predicted shifts are unchanged provided that the degeneracy splitting is smaller in energy than \( k_B T \). This is important because

\[\phi = (\nu - n_{\text{shell}}) f + (n_{\text{shell}} + 1)(1-f),\]

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we expect level degeneracies to be lifted in the real, imperfect QDs that we measure. We note that, according to our theory, the observation of peak shifts down to 4.5 K implies that the level splitting is smaller than this temperature corresponding to roughly 0.4 meV. Furthermore, the same degeneracy theory leads to small, but measurable, shifts between the dissipation peak and the frequency peak corresponding to the same charge degeneracy point. This is visible for the third peak (from the right) in Fig. 2 in which the $\gamma$ and $\Delta \omega$ peaks do not exactly line up, with the measured shifts compared to theory in Fig. S1 (see SI Text). These dissipation-frequency shifts further support our model, ruling out the alternative of a temperature-dependent renormalization of $E_C$ or $AE$.

Finally, before we discuss results for coupled QDs, it is worth noting that our theoretical treatment of degeneracies also predicts that the Coulomb blockade cantilever dissipation peaks should have a slightly asymmetric line shape; cf. Eq. 2. This effect was unfortunately too small to be resolved in the current experiment, which focuses on cantilever–dot couplings that are weak enough that the motion of the cantilever only weakly perturbs the dot. We recently studied both theoretically and experimentally the more complex regime where this coupling strength becomes strong, because of a much larger cantilever oscillation amplitude. We find that in this strong coupling regime, degeneracy-induced line shape asymmetries become much more pronounced and clearly resolvable (29).

Significant efforts are ongoing towards understanding and controlling the properties of coupled QDs, in particular double QDs or “artificial molecules” (30). The $\gamma$ images that we obtain for double QDs are equivalent to stability diagrams that depict the charge state of the double QD system. This is because of the position dependence of the lever arm $\alpha(x,y,z)$ for each QD that results in two electrochemical potentials, $\mu_{QD1}$ and $\mu_{QD2}$. Scanning the AFM tip at constant height and $V_B$ corresponds to sweeping the $\mu_{QD1}$–$\mu_{QD2}$ space through changing $\alpha_1$ and $\alpha_2$ even though only a single electrode (the tip) is being used. In a conventional stability diagram, lines of constant electrochemical potential for each QD are plotted as a function of two gate voltages. When the two QDs are coupled, intersection points are split into two points (triple points), showing avoided crossings (30). In the $\gamma$ images the avoided crossings are observed when the ring radii suddenly change at intersection points. Fig. 4 A and B show the same three QDs as in the lower part of Fig. 1E, now imaged at −9 and −7.6 V, respectively. Such avoided crossings are highlighted in the circle and box in Fig. 4A, representing an example of weak and strong coupling, respectively.

We characterize the coupling strength by comparing the ratio of the change in ring radius to the separation between the first two rings ($2E_C$) for QD2 (30). This method is valid only when both rings are far enough from the QD center that the voltage drop between them is approximately linear. By following this procedure, the coupling of QD2 to QD1 (circle) and QD3 (box) can be compared. Whereas the change in radius of QD2 is approximately $0.10 \pm 0.01$ of $2E_C$ because of QD1, it is $0.46 \pm 0.03$ because of QD3, indicating a much stronger coupling between QD2 and QD3. We consider the former to be an example of weak coupling because the triple points are nearly joined. This is consistent with a small capacitive coupling between the two dots. The charging of one dot effectively gates the second dot, causing a sudden change in ring radius.

Conversely, the boxed region in Fig. 4A is an example of strong coupling because there is a large gap at ring intersections as in the triple points of a stability diagram. In Fig. 4B, the same three QDs as in Fig. 4A are imaged at smaller $V_B$. This image allows for a more intuitive explanation of the coupling. Consider the diagonal line from the center of QD3 outward; initially, the AFM tip is over QD3. As it moves closer to QD3, the charging of one dot effectively gates the second dot, causing a sudden change in ring radius.

Fig. 4 C–E shows another example of coupled QDs at 4.5 K. The InAs structure (Fig. 4C) contains coupled QDs as shown in the $\gamma$ image (Fig. 4D). Fig. 4E zooms up on the region in Fig. 4D showing many avoided crossings. Within this distance range from the QD centers, each $\alpha$ is approximately linearly dependent on the tip position so that scanning the tip more closely resembles sweeping two gate voltages, resulting in the image resembling a conventional stability diagram. Fig. 4 also highlights how advantageous it is to have images in addition to the $\gamma$-$V_B$ spectra be.
cause the spectra alone will contain the peaks from nearby QDs that can be identified by using the images. Yet another advantage of this technique is the ability to spatially resolve the effects of changes in the local electrostatic environment; for self-assembled QDs, no other technique is capable of doing this. The AFM images show how the QD confinement potential is being influenced by charge reconfiguration. Fig. 5 shows two such cases. The structure in Fig. 5A shows a fluctuation in electron population because of nearby fluctuations in the electrostatic background at both 78 (Fig. 5B) and 4.5 K (Fig. 5C). The missing sections of the first ring in Fig. 5B indicate that the number of electrons in the QD is fluctuating by one in this region. Interestingly, depending on the scan direction (left to right or right to left) over the QD, the missing section may appear. A similar reconfiguration was observed in the -V_f B spectra. Whereas all the peaks appear in the reverse V_f sweep (positive to negative), the first peak disappears in the forward sweep. Fig. 5D–G shows a more dramatic change. During the scan, a sudden switch in the confinement potential occurs, leading to the transition from the single QD (Fig. 5D) to a coupled double QD (Fig. 5E). This double QD state could be switched back to the single state (Fig. 5G) by scanning over the same area with a positive V_f (Fig. 5F). Although both of these changes are readily identified in the images, having a spectrum alone may cause confusion as is the case in conventional transport measurements. These observations indicate that the QD confinement potentials are very sensitive to the electrostatic background and can be modified, or switched, controllably.

Charge sensing with AFM can be used to investigate the electronic structure of single and coupled self-assembled QDs. The technique enables the quantitative extraction of the tunneling rate, charging energy, and the QD interaction energies; further, we have used it to perform a unique measurement of temperature-dependent Coulomb blockade peak shifts confirming the shell degeneracy of the QD. The dissipation images proved especially useful in analyzing multiple QDs and changes in QD confinement potential resulting from nearby charge fluctuations. The images also revealed that what looked like a single QD structure topographically can actually contain multiple QDs, which might be a result of the local modulation of the confinement potential caused by oxidation or structural defects. Additionally, the imaging capability of AFM provides insight into the link between QD electronic structure and topography, which is of great importance in developing electronic devices on the basis of QDs.

Materials and Methods

Sample. The sample, grown by chemical beam epitaxy (31), consists of the following layers: 460 nm undoped InP grown on top of an InP substrate, followed by a 10 nm Si-doped InP layer, 10 nm undoped layer, 10 nm In₀.₅Ga₀.₅As layer, 20 nm undoped layer, and a 1.82 ML InAs layer that results in the formation of InAs QDs by Stränki–Kratanov growth. The QDs cover the surface with a density of ~2.5 QDs per μm² having diameters in the range of 30–95 nm and heights of 0.5–6 nm. The 2DEG layer formed in the InAs layer serves as a back electrode, and an Ohmic contact to the 2DEG is made by indium diffusion.

Experiment and Data Processing. Our home-built cryogenic AFM (32) includes an rf-modulated fiber optic interferometer (33) with 1,550-nm wavelength for cantilever position detection. We coat Si AFM cantilevers (Nanosensors PPP-NCLR) with 10 nm titanium (adhesion layer) and 20 nm platinum. The cantilevers typically have a spring constant of k = 48 N/m, with a 160 kHz resonance frequency and a quality factor, Q, between 100,000 and 1,000,000 at 4.5 K. All of the images were taken in frequency modulation mode [(20)]. In this mode, the cantilever is self-oscillated at its resonance frequency with a constant amplitude. The frequency shift and dissipation were measured with a commercially available phase-locked loop frequency detector (Nanosurf, easyPLL plus). The topography images were taken in constant frequency shift mode where a constant frequency shift is maintained by regulating the cantilever tip-sample distance by using a feedback controller. The frequency shift and dissipation images were taken in constant-height mode with a typical tip height of 20 nm. Dissipation images are shown in Fig. S2 as a function of V_f. More negative V_f results in additional atoms to the QD. Areas of increased dissipation mark 2DEG–QD tunneling events. Each time a ring is crossed when traveling toward the quantum dot center marks the addition of an electron to the dot. More details of the AFM images are listed in Table S1. The amplitude of the cantilever excitation signal, A_exc, is provided as a measure of the dissipation signal from the Nanosurf oscillator controller. It is converted to units of 1/V via 


Phys 64:701–736.


10. Igarashi Y, et al. (2007) Spin-half Kondo effect in a single self-assembled InAs quantum


double-barrier tunnelling structure observed by noncontact atomic-force spectroscopy.


teraction forces with a dynamic force microscope using the frequency modulation


to atomic force microscopy cantilevers with controlled angle, length, and radius for

27. Clerk AA, Bennett S (2005) Quantum nonelectromechanics with electrons, quasi-
particles and cooper pairs: effective bath descriptions and strong feedback effects.
New J Phys 7:238.

28. Brink M (2007) Imaging single-electron charging in nanostructures by low-tempera-

coupling of an atomic force microscope cantilever to a quantum dot. Phys Rev Lett
104:017203.


19:1467–1470.

71:3782–3787.

force microscopy.