Collective Coulomb Blockade in an Array of Quantum Dots: A Mott-Hubbard Approach

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We investigate the electron addition spectrum in a class of Hubbard-like models which describe arrays of coupled quantum dots. Interdot tunneling leads to a sequence of two phase transitions separating a region of collective Coulomb blockade from a region where the Coulomb blockade of individual dots is maintained and a region where the Coulomb blockade is destroyed altogether. Observable experimental consequences of our theory are discussed.

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Arrays of coupled quantum dots [1,2] provide a novel system in which one can study "solid-state" physics on a much lower energy scale. Because of the dominant role played by electron-electron interactions in quantum dots, as manifested in the phenomenon of the Coulomb blockade [3], it is expected that quantum dot arrays should mimic the physics of the Hubbard model, which has been studied extensively in the context of high-T_c superconductivity. To date, theoretical treatments of quantum dot arrays have focused largely on arrays of metallic dots [4], where the discreteness of the dot energy levels can be neglected, or arrays with nearly transparent barriers between dots [5], where the Coulomb blockade is absent. In this Letter, we investigate the electron addition spectrum in arrays of four coupled semiconductor quantum dots in the Coulomb blockade regime and its vicinity using a generalized Hubbard model to account for the effects of quantum confinement, intradot Coulomb interactions, and interdot tunneling. We find a remarkably rich phase diagram for tunneling-coupled quantum dot arrays which generically exhibits three phases: For weak interdot tunneling, the quantum dot states are split into minibands but the Coulomb blockade of individual dots is maintained; for intermediate tunneling strengths, the Coulomb blockade of individual dots is destroyed but there remains an energy gap of collective origin analogous to the energy gap in a Mott insulator (collective Coulomb blockade); and for strong tunneling, the Coulomb blockade is destroyed altogether. We argue that the experimental conductance spectra of Refs. [1,2] provide evidence for the existence of the latter transition—which is the analog of the Mott-Hubbard insulator-metal transition—in a one-dimensional (1D) array of quantum dots.

The system we wish to model consists of a linear or square array of identical quantum dots electrostatically defined [1,2] in a 2D electron gas, separated from a metallic backgate by a thin insulating layer. We consider the case where each quantum dot contains several electrons ($n > 10$) and the applied magnetic field is weak; then the electron-electron interactions within the parabolic confining potential of a single dot are rather well described by a self-consistent Hartree energy, and for small variations about some large, fixed n, can be parametrized by a capacitive charging energy $U = e^2/C(n)$, in accordance with the Coulomb blockade picture [3]. Longer range Coulomb interactions are screened by the backgate. We wish to model collective phenomena at the meV energy scale in GaAs quantum dots of area $\sim(100\ \mathrm{nm})^2$; given the density of states in GaAs of $(2.7\ \mathrm{meV}^{-1})/(100\ \mathrm{nm})^2$, it will be adequate to consider only the $M = 3$ or 4 single-particle energy levels in the confining potential of an isolated dot which are nearest the Fermi level, ε_α, $\alpha = 1, \ldots, M$ (α specifies the quantum state of both the orbital and spin degrees of freedom), which we take to be nondegenerate with level spacing Δ (the spin degeneracy can be lifted by a magnetic field or by spin-orbit coupling). The dominant effect of interdot coupling in such a system is to introduce a tunneling matrix element t_α between equivalent single-particle states in nearest-neighbor dots. We neglect the tunneling matrix elements between nonequivalent states; this is the usual tight-binding approximation, and is justified for nearly identical dots provided t_α is not too large. The Hamiltonian is

$$\hat{H} = -\sum_{\substack{\langle \mathbf{i},\mathbf{j}\rangle \\ \alpha}} (t_\alpha \hat{c}^\dagger_{\mathbf{i}\alpha}\hat{c}_{\mathbf{j}\alpha} + \mathrm{H.c.}) + \sum_{\mathbf{j},\alpha} \varepsilon_\alpha \hat{c}^\dagger_{\mathbf{j}\alpha}\hat{c}_{\mathbf{j}\alpha} + \frac{U}{2}\sum_{\mathbf{j}} \hat{n}_{\mathbf{j}}(\hat{n}_{\mathbf{j}} - 1), \qquad (1)$$

where $\mathbf{i}, \mathbf{j}$ are vectors of integers labeling the positions of the dots, $\hat{c}^\dagger_{\mathbf{j}\alpha}$ is the creation operator for an electron in state α of the $\mathbf{j}$th dot, $\hat{n}_{\mathbf{j}} \equiv \sum_\alpha \hat{c}^\dagger_{\mathbf{j}\alpha}\hat{c}_{\mathbf{j}\alpha}$, and the sum over $\langle \mathbf{i},\mathbf{j}\rangle$ is over nearest neighbors only. Since the phase diagram which we propose for tunneling-coupled quantum dot arrays is not expected to depend sensitively on the details of the Hamiltonian, the simplifications involved in Eq. (1) should be irrelevant.

In this Letter, we calculate the equilibrium electron addition spectrum of (1), $\partial\langle\hat{N}\rangle/\partial\mu = kT\,\partial^2 \ln \mathcal{Z}/\partial\mu^2$, by numerically diagonalizing $\hat{H}$ and evaluating the grand partition function $\mathcal{Z} = \mathrm{Tr}\{\exp[-(\hat{H} - \mu\hat{N})/kT]\}$. For the system of 4 quantum dots with $M = 4$ levels/dot, $\mathcal{Z}$

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involved a sum over 65 536 states. The differential self-capacitance of the experimental quantum dot system [6] is given by $\partial Q/\partial V = e^2\partial\langle\hat{N}\rangle/\partial\mu$, provided the tunneling rate from the backgate is much less than $kT/\hbar$.

We first consider an array of four quantum dots arranged in a square with three single-particle energy levels per dot, and study the evolution of the addition spectrum as the interdot tunneling is increased (see Fig. 1). Here we have set the tunneling matrix elements for all three quantum levels equal to t and have taken $\Delta = 0.3U$ and $kT = 0.04U$. At $t = 0$, the behavior characteristic of isolated dots [3] is evident in Fig. 1, namely, three peaks separated by the Coulomb blockade energy $U + \Delta$. Each peak at $t = 0$ represents the addition of four electrons to the array (one to each quantum dot). As t is increased, the quantum dot states are split into minibands and each capacitance peak is split into three peaks, the central peak representing the addition of two electrons at the same value of the chemical potential. When $t \sim \Delta$, this degeneracy is lifted and one can see three Hubbard minibands, each composed of four states. Finally, when $t \sim U/2$ the energy gap between the minibands is no longer discernible.

Figure 1 shows evidence of three distinct phases in the quantum dot array, which we will now analyze in detail. In the weak-tunneling phase, characterized by the degeneracies in the addition spectrum, $\partial\langle\hat{N}\rangle/\partial\mu$ has peaks at zero temperature at $\mu = -2t$, 0 ($\times 2$), and $2t$, with this pattern repeated, centered at $U + \Delta$ and at $2(U + \Delta)$. The energies of the lowest miniband are those of non-interacting electrons in a tight-binding band with four lattice sites, $E = -2t\cos k$, $k = 0, \pm\pi/2, \pi$. This reflects the fact that at sufficiently small t there is no admixture of the higher single-particle states ε_2 and ε_3 in the many-body ground state, so that the Pauli principle prevents "double occupancy" at $T = 0$, thus negating the interaction term in Eq. (1). The minibands centered at $U + \Delta$ and $2(U + \Delta)$ are identical, but occur when the states ε_1 and ε_2 are completely filled. The Coulomb blockade of the individual quantum dots is thus maintained in this weak-tunneling phase, which is analogous to the ferromagnetic phase of the Hubbard model in a strong magnetic field.

As t is increased, it becomes energetically favorable to admix the higher single-particle states ε_2 and ε_3 in the many-body ground state—thereby allowing charge fluctuations $\Delta n_j > 1$—in order to lower the kinetic energy of the system. The degeneracies present in the weak-tunneling phase are then lifted by the interaction term in Eq. (1). This transition represents a breakdown of the Coulomb blockade of individual quantum dots, and occurs in Fig. 1 when $t \sim \Delta$. For the case of an infinite 1D array of quantum dots with $M = 2$ levels per dot, this phase transition is equivalent to the ferromagnetic-antiferromagnetic phase transition of the 1D Hubbard model in a magnetic field [7], and in the limit $U \gg \Delta$ the critical value of the tunneling matrix element is [7]

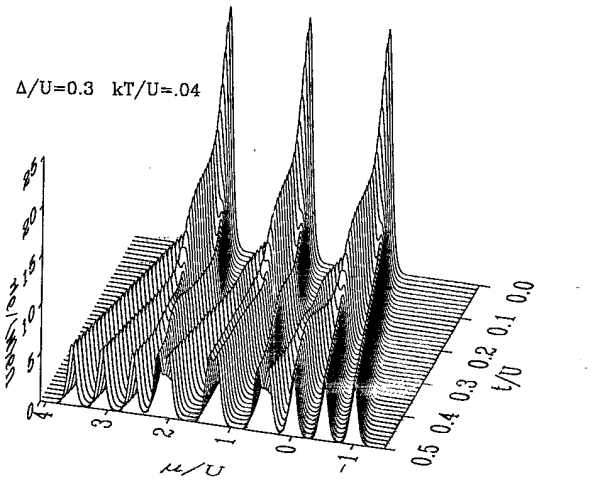

FIG. 1. A plot showing the evolution of the electron addition spectrum $\partial\langle\hat{N}\rangle/\partial\mu$ as a function of the interdot tunneling matrix element t for an array of four quantum dots with three single-particle energy levels per dot, arranged in a square.

$t_{c1}^2 = (\pi\Delta U/4)/(2\pi n - \sin 2\pi n)$, where $n < 1$ is the filling factor of the lowest miniband. We find that this transition is qualitatively similar for $M = 3$ and 4, and in finite arrays (although it is of course not a true phase transition in a finite array). Because t is many orders of magnitude smaller than the band width in conventional solid-state systems, it should be possible to observe this phase transition at Zeeman splittings Δ easily obtainable in the laboratory, whereas an astronomically large magnetic field would be required to spin polarize a conventional metal-oxide system.

Despite the destruction of the Coulomb blockade in the individual quantum dots when $t > t_{c1}$, there is still an energy gap between the minibands above this transition. This energy gap is a collective effect which we refer to as "collective Coulomb blockade" (CCB), and is analogous to the energy gap in a Mott insulator [8]. The CCB regime is characterized by strong interdot correlations which are analogous to the antiferromagnetic correlations in Mott insulators [9], i.e., occupancy of the state ε_α in one quantum dot is anticorrelated with the occupancy of that state in its nearest neighbors.

When t is increased still further, the energy gap between minibands in Fig. 1 collapses. The breakdown of CCB [10] in the strong-tunneling regime is directly analogous to the Mott-Hubbard insulator-metal transition [8,11]. For $M = 2$ single-particle energy levels per dot, this transition occurs at $t/U = \infty$ for a 1D array [11], but for $M > 2$, the transition is expected to occur at a finite value of t due to the absence of Fermi-surface nesting [12]; Fig. 1 is consistent with the critical value obtained in Ref. [12] for an infinite array with $M = 3$, $t_{c2}/U = 0.39$ [although the metal-insulator transition (MIT) studied in Ref. [12] occurs in an unspecified model which is merely analogous to Eq. (1)].

To this point, we have assumed equal tunneling matrix

elements for each of the quantum dot states, but in fact the more weakly bound states should be connected by stronger tunneling matrix elements. To simulate this effect, in Fig. 2(a) we show the electron addition spectrum for a linear array of four quantum dots with four single-particle energy levels per dot with tunneling matrix elements $t_\alpha = 0.1U(1.5)^{\alpha-1}$, $\alpha = 1,\ldots,4$. The level spacing is $\Delta = 0.3U$. Note that for a linear array with open boundary conditions, the degeneracies discussed above do not occur. When disorder was introduced into the tunneling matrix elements [Fig. 2(b)], the gross features of the spectrum were unchanged, but the peak spacings within the minibands were different. The energy gap between the minibands decreases with increasing chemical potential; when the value of U was decreased 40% relative to Δ and t_α, the energy gap between the third and fourth minibands was no longer discernible [Fig. 2(c)], indicating an effective $t_{c2}/U \sim 0.5$. As discussed above, this breakdown of CCB [10] is analogous to the Mott-Hubbard insulator-metal transition. Quantum dot arrays should thus provide a system where this transition can be studied by *tuning a gate voltage*, which would be inconceivable in the metal-oxide systems which display conventional Mott-Hubbard MITs. We emphasize our finding that this quantum phase transition—which strictly exists only for the infinite system at $T = 0$—shows up strikingly in a small array of dots at finite temperature, as our numerical diagonalization clearly demonstrates.

A reexamination of the work of Kouwenhoven *et al.* [1] leads us to conclude that the analog of the Mott-Hubbard MIT may have already been observed in a 1D array of quantum dots. In Ref. [1], the conductance spectrum of a linear array of fifteen GaAs quantum dots ~ 100 nm in diameter was measured. While our capacitance spectra cannot be compared directly to those conductance spectra, the peak positions should be the same in linear response, though the peak heights and possibly the line shapes would be different. At low values of the gate voltage V_{g2} (corresponding to low values of the chemical potential in the array), conductance peaks separated by 4 mV are evident in Fig. 1 of Ref. [1]. Given the electrostatic "lever arm" of the gate with respect to the quantum dots of ~ 4, this implies a 1 meV spacing in energy between the peaks, which is roughly the Coulomb blockade energy $e^2/\epsilon d$ for a GaAs quantum dot of diameter $d = 100$ nm. We therefore interpret these peaks as Hubbard minibands in which the individual discrete states are unresolved due to a finite electron temperature (cf., the lowest miniband in Fig. 2). As the gate voltage V_{g2} was increased, the conductance peaks broadened [1], and some structure in the peaks became observable, as would be expected due to the exponential increase of the tunneling matrix elements with increasing chemical potential. The center to center peak spacing, however, appears to be constant to $\pm 10\%$, indicating that it is a reasonable approximation to neglect the dependence of U and Δ on

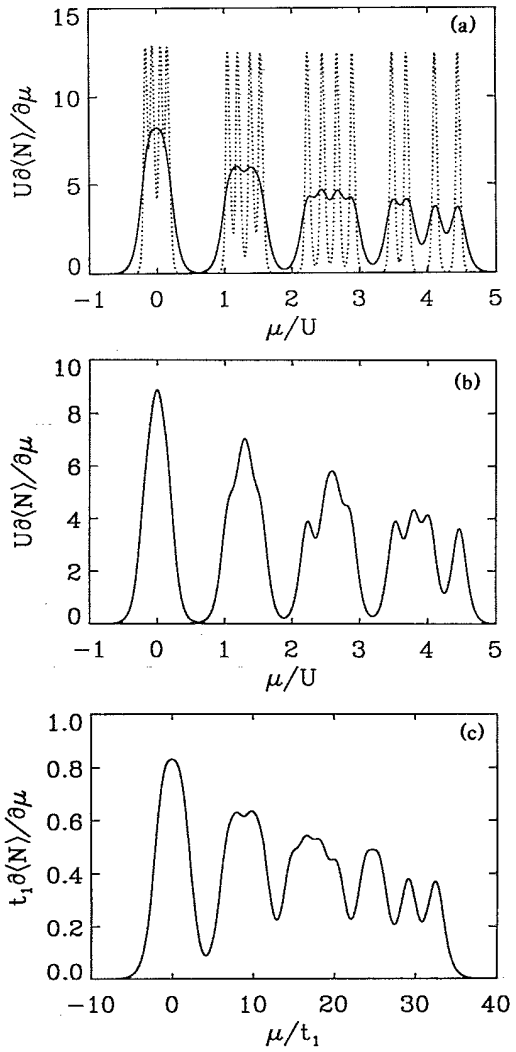

FIG. 2. (a) A plot of the electron addition spectrum versus chemical potential for linear array of four quantum dots with four single-particle energy levels per dot. $\Delta = 0.3U$ and $t_\alpha = 0.1U(1.5)^{\alpha-1}$, $\alpha = 1,\ldots,4$. Solid curve: $kT = 0.7t_1$. Dotted curve: $kT = 0.2t_1$. (b) Same as in (a), but with disorder $\delta t_\alpha/\bar{t}_\alpha = 0.3$. (c) Same as in (a), but with $U = 2\Delta = 6t_1$.

the chemical potential in this regime, as we have done in our calculation. Finally the gap structure with a period of 4 mV was destroyed at $V_{g2} \approx -0.76$ V, which we interpret as the breakdown of CCB. (This MIT is particularly evident in the spectra at $V_{g1} = -0.44$ V and -0.46 V.) Qualitatively similar behavior was observed in an array of four quantum dots by Haug *et al.* [2].

The breakdown of CCB in the quantum dot array of Ref. [1] appears to occur at the foot of the first conductance plateau when $G \simeq e^2/h$, i.e., when there is one channel of nearly perfect transmission through the array. Dimensional arguments imply that the hopping matrix element between two quantum dots of size d at the threshold for unit transmission is $t \sim \hbar^2/m^* d^2$; a numer-

ical simulation of single-particle tunneling through 1D barriers of various heights and widths at the threshold for unit transmission gave $t \simeq 4\hbar^2/m^*d^2$, with the numerical prefactor ranging from 2 to 8. We therefore estimate t_c for the MIT of the system of Ref. [1] to be ~ 0.5 meV, yielding the ratio $t_c/U \sim 0.5$. This is consistent with the theoretical prediction [12], $0.29 \leq t_c/U \leq 0.39$ for $\infty \geq M \geq 3$, thus substantiating our interpretation of the data as evidence for a Mott-Hubbard MIT.

An alternative hypothesis which could account for the data is that the conductance of the array near pinchoff is dominated by a single ~ 100 nm Coulomb island, with the electrons traveling ballistically through the rest of the array. While the breakdown of the Coulomb blockade in a single dot generically occurs when $G \sim e^2/h$, the breakdown of CCB is expected to occur at a characteristic value $t_c/U \sim 0.4$ for a 1D array, which implies by the arguments above that $G_c \sim (0.1d/a_B)^2 e^2/h$ for quantum dots of size d, where $a_B = 10.5$ nm. Comparing the spectra of Ref. [1] at $V_{g1} = -0.44$ V and -0.46 V (where the effective size of the dots is smaller), the MIT at -0.46 V does indeed appear to occur at a smaller value of G. A naive extrapolation of these arguments for dots larger than 100 nm would imply $G_c > e^2/h$ (more than one channel of perfect transmission through the array), but in that case the Hubbard model description of the system would break down and the system would behave like a quantum wire, with no Mott-Hubbard gap. Thus e^2/h is an upper bound on G_c. Here we have implicitly assumed that the barriers between the leads and the array are comparable to the interdot barriers, as in the system of Ref. [1]. Since the conductance of the array is proportional to the coupling to the leads, one way to decrease G_c is simply to decrease the coupling to the leads, while keeping the interdot coupling fixed.

The effects of disorder, magnetic fields, and interdot Coulomb interactions on the above results should be mentioned. In order for the degeneracies in the electron addition spectrum which occur in the weak-tunneling regime to be discernible experimentally, the disorder in any of the parameters in Eq. (1), as well as the magnitude of the interdot Coulomb interactions, would have to be less than the level spacing within a miniband, which may be impossible to achieve in practice. The modification of the tunneling matrix elements by the Peierls phase factor in a magnetic field would also lift these degeneracies. Even in the presence of these perturbations, however, there remains a weak-tunneling phase in which the Coulomb blockade of individual quantum dots is maintained, and this phase is expected to have clear signatures in both optical absorption and conductance experiments. Interdot Coulomb interactions, which can give rise to interesting effects in arrays of electrically isolated dots [13], are expected to be less important in arrays coupled by tunneling, and should be irrelevant to the phase diagram in the presence of disorder [14]. Our model is not adequate to describe the intricate magnetic field dependence of the electron addition spectrum in the strong-field regime [6] because the Hubbard-type interaction term in Eq. (1) does not account for the strong *intradot* electron correlations present there. Nonetheless, Eq. (1) may be adequate as a phenomenological Hamiltonian to describe *collective* effects in quantum dot arrays at fixed field even in the strong-field regime.

In conclusion, we have argued on general grounds that the interplay of quantum confinement, interdot tunneling, and strong intradot Coulomb interactions leads to three distinct zero temperature phases in quantum dot arrays, and have identified experimental evidence [1,2] for the analog of the Mott-Hubbard metal-insulator transition in a 1D array of quantum dots. Our results suggest that quantum dot arrays should provide ideal experimental systems in which to study a variety of interaction-driven quantum phase transitions predicted to occur in Hubbard-like models [7,8,11,12], albeit at very different energy scales (~ 1 meV) compared with what these models were originally intended for (~ 1 eV).

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