Microscopic Theory for Conductance Oscillations of Electron Tunneling through a Quantum Dot

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(Received 2 February 1994)

A microscopic theory for conductance oscillations of electron tunneling through a quantum dot occupied by electrons with a strong Coulomb interaction is proposed. It is found that the conductance oscillations are approximately periodic only for a large number of electrons ($\gtrsim 30$). A "shell" structure in the oscillations for a small number of electrons is predicted. In addition, a pair structure reflected by even- and odd-occupation numbers is revealed in peak heights and linewidths as well as separations between adjacent peaks of the oscillations. Peak height and linewidth increase with increasing chemical potential. These predictions are in agreement with experimental measurements.

PACS numbers: 73.40.Gk, 73.20.Dx

Nanoscale quantum dots exhibit striking quantum phenomena [1–4]. The experimentally observed periodic conductance oscillations at low temperatures is one such phenomenon [5,6]. By virtue of an innovative device, Meirav, Kastner, and Wind were able to conclude that each cycle in the conductance oscillations corresponds to the addition of one electron to the dot, constituting a quantized charging process [7]. One interpretation of the conductance oscillations has been in terms of a classical Coulomb blockade theory (CBT) [8,9]. Alternatively, treating the dot as a single site, an interplay of the conductance oscillations and quantized energy spectrum can be qualitatively explained within the Wolff-Anderson model (WAM) for a single impurity as proposed by Meir, Wingreen, and Lee [10]. Based on the WAM, these authors discussed the temperature dependence of peak heights of the oscillations and the transport behavior in the Kondo regime [10,11]. More recently, an experimental investigation of channel competition for single electron tunneling through a dot has been reported [12] and an exact Breit-Wigner formula for 1D multibarrier resonance tunneling has been derived [13].

The Coulomb interaction in a quantum dot is very strong and thus plays a central role in the quantum transport process, as indicated in the WAM [10]. But that model can only qualitatively describe an energy spectrum for 2 electrons in the dot. Experimentally, the conductance oscillations were measured in a range of electron occupations 30–60 [14]. Similarly, the classical CBT itself cannot give any information related to the energy spectrum of the dot. Thus, it is difficult for either model to describe detailed structure of the oscillations. Again, only for a few electrons in the dot can an exact energy spectrum be numerically obtained by exact diagonalization [15,16]. This is far below the range in which the experiment was performed [14]. It is the purpose of this Letter to show for the first time how the strong Coulomb interaction (including both direct and exchange interactions), and hence the energy spectrum and the occupation number of electrons, microscopically affect the period and structure of the conductance oscillations.

We propose a microscopic resonant tunneling mechanism to interpret the conductance oscillations based on a Hartree-Fock (HF) calculation [17]. Our key findings are the following: (1) the conductance oscillations are aperiodic (approximating a "shell" structure) for a small number of electrons in the dot, and gradually become approximately periodic for large numbers ($\gtrsim 30$); (2) there is a pair structure (a reflection of even- and odd-occupation number) in peak heights and linewidths of the oscillations; and (3) the peak height and the linewidth tend to increase with increasing chemical potential, which is the energy carried by the tunneling electrons.

The quantum dot is considered as a two dimensional system in the x-y plane. We choose a parabolic confining potential, and take a typical value 2 meV for $\hbar\omega_0$ throughout this paper. We assume two perfect leads connecting to the dot on both sides. The total Hamiltonian of the system is given by

$$H = H_l + H_d + H_h, \tag{1}$$

where

$$H_l = \sum_{k,\sigma} \epsilon_k \hat{c}^\dagger_{k,\sigma} \hat{c}_{k,\sigma},$$

$$H_d = \sum_{\alpha,\beta,\sigma} \langle \alpha | T + U | \beta \rangle \hat{c}^\dagger_{\alpha,\sigma} \hat{c}_{\beta,\sigma} + \frac{1}{2} \times \sum_{\alpha,\beta,\gamma,\rho,\sigma,\sigma'} \langle \gamma\alpha | V^{\text{Coulomb}} | \beta\rho \rangle \hat{c}^\dagger_{\gamma,\sigma} \hat{c}^\dagger_{\alpha,\sigma'} \hat{c}_{\beta,\sigma'} \hat{c}_{\rho,\sigma},$$

$$H_h = \sum_{\alpha,k,\sigma} (V_{k,\alpha} \hat{c}^\dagger_{k,\sigma} \hat{c}_{\alpha,\sigma} + \text{H.c.}). \tag{2}$$

The three terms in Eq. (1) are associated with the leads, the dot, and the hopping process between the leads and dot, respectively. An electron state in the leads (dot) is labeled by k (α) with a spin index σ. $T + U = \vec{p}^2/2m^* + m^*\omega_0^2\rho^2/2$ ($m^* = 0.067 m_e, m_e$ is an electron bare mass) is a one-body Hamiltonian

0031-9007/94/73(4)/585(4)$06.00

which contains the confining potential. The corresponding single electron eigenfunctions are $\phi_{n,m} = \rho^{|m|}\exp(-im\theta)L_n^{|m|}(\rho^2/2a^2)\exp(-\rho^2/4a^2)$ [18], where $a = (\hbar/2m^*\omega_0)^{1/2}$, and m and n are an azimuthal angular momentum and a principle quantum number, respectively. The eigenvalues are $\epsilon^{(0)}_{n,m} = (2n + |m| + 1)\hbar\omega_0$ [18] with $n = 0, 1, 2, \ldots$ and $m = 0, \pm 1, \pm 2, \ldots$. V is the barrier between the leads and dot and is denoted by a sum of U and the Hartree potential V^H of the dot. We calculate the quasiparticle (QP) energy spectrum by solving the zero temperature HF equation [19]

$$\left[-\frac{\hbar^2\nabla^2}{2m^*} + \frac{1}{2}m^*\omega_0^2\rho^2\right]\psi_\alpha(\vec{x}_1) + \int d^2x_2\,\hbar\Sigma(\vec{x}_1,\vec{x}_2)\,\psi_\alpha(\vec{x}_2) = \epsilon_\alpha^{\mathrm{HF}}\psi_\alpha(\vec{x}_1), \quad (3)$$

where

$$\begin{aligned}\hbar\Sigma(\vec{x}_1,\vec{x}_2) = {} & \delta^2(\vec{x}_1 - \vec{x}_2)\int d^2x_3\, V^{\mathrm{Coulomb}}(\vec{x}_1 - \vec{x}_3) \\ & \times \sum_\beta g_\beta\,|\psi_\beta(\vec{x}_3)|^2\,\theta(\epsilon_F^{(N)} - \epsilon_\beta^{\mathrm{HF}}) \\ & - V^{\mathrm{Coulomb}}(\vec{x}_1 - \vec{x}_2) \\ & \times \sum_\beta \psi_\beta(\vec{x}_1)\,\psi_\beta^*(\vec{x}_2)\,\theta(\epsilon_F^{(N)} - \epsilon_\beta^{\mathrm{HF}}).\end{aligned} \quad (4)$$

Here, α is a label identifying the QP state (m, ν) (ν is the principle quantum number, but is different from n). $V^{\mathrm{Coulomb}} = (e^2/\kappa)|\vec{x}_i - \vec{x}_j|$ with a dielectric constant $\kappa = 13$ for the host semiconductor. $g_\beta = 2$ (1) corresponds to 2 (1) electrons in state β. $\epsilon_F^{(N)}$ is the Fermi energy for N electrons in the dot. Equation (3) is diagonal in m. The Coulomb interaction makes the QP wave function completely different from that of the single electron. One way to obtain a QP wave function is to rewrite the Fock term as an equivalent local potential with a QP wave function in its denominator, and then to solve the resulting differential equation. Another approach is to map the HF equation into a matrix form in terms of a complete basis, the QP energy levels and wave functions can be found by diagonalization of the matrix in a truncated space. We adopt the second one, which is well established in atomic and nuclear physics [20].

We expand ψ_α by $\{\phi_{n,m},\ n = 0, 1, 2, \ldots\}$ with a fixed value of m as

$$\psi_\alpha = \sum_n c_n^\alpha \phi_{n,m}. \quad (5)$$

Here, each value of m corresponds to a matrix equation

$$\sum_{n'}\left[(\epsilon_\alpha^{\mathrm{HF}} - \epsilon^{(0)}_{n,m})\delta_{n,n'} - \int d^2x_1 d^2x_2\,\phi^*_{n,m}(\vec{x}_1)\,\hbar\Sigma(\vec{x}_1,\vec{x}_2)\phi_{n',m}(\vec{x}_2)\right]c^\alpha_{n'} = 0, \quad (6)$$

where $\epsilon^{(0)}_{n,m}$ is a diagonal matrix element of the one-body Hamiltonian. Equation (6) is solved self-consistently. We fill electrons starting from the lowest energy empty states (ν, m)'s. According to our convergence test, five terms in the expansion of Eq. (5) give satisfactory results for up to 40 electrons in the dot.

We have compared our HF ground state energies with those calculated by using an exact diagonalization method for a few electrons [15]. For the case of four electrons in the dot, our total binding energy is about 5% higher. However, the HF result is expected to rapidly improve with increasing occupation number. It is worth pointing out that the Fock term contributes to the energy spectrum as much as 20%, so it is very important.

Following the treatment in Ref. [10], we use a generalized Landauer-type formula [21] to calculate the conductance:

$$\sigma = \frac{e^2}{h}\sum_\alpha \int_{-\infty}^{\infty} d\epsilon\, f'_{\mathrm{FD}}(\epsilon)\,\frac{\Gamma_\alpha(\epsilon)}{2}\,\mathrm{Im}[G_\alpha(\epsilon)], \quad (7)$$

where f_{FD} is the Fermi-Dirac distribution function, Γ is an elastic coupling between the leads (the two leads are assumed to be symmetric) and dot, and in the Born approximation $\Gamma_\alpha(\omega) = 2\pi\sum_k |V_{k,\alpha}|^2\,\delta(\omega - \epsilon_k)$. $\mathrm{Im}[G_\alpha]$ is given by

$$\mathrm{Im}[G_\alpha(\epsilon)] = \frac{\mathrm{Im}\Sigma_\alpha}{(\epsilon - \epsilon_\alpha^{\mathrm{HF}})^2 + (\mathrm{Im}\Sigma_\alpha)^2}, \quad (8)$$

where $\mathrm{Im}\Sigma_\alpha = \Gamma/2 + \mathrm{Im}\Sigma_\alpha^{(c)}$. ($\mathrm{Im}\Sigma_\alpha^{(c)}$ is also attributed to both the Coulomb interaction and Γ.) At the zero temperature, $f'_{\mathrm{FD}}(\epsilon) = -\delta(\epsilon - \mu)$ (μ is the chemical potential of the lead). Suppose that a dot is occupied by N electrons and an incoming electron tunnels through the barrier and enters the dot. There are many states for this electron to mediate. Since $\mathrm{Im}[G_\alpha(\mu)]$ has a Lorentzian form with a narrow width, it decays rapidly with increasing deviation of the mediating energy $\epsilon_\alpha^{\mathrm{HF}}$ from μ. Our calculation of the self-energy of electron-electron correlations shows that $\mathrm{Im}\Sigma_\alpha$ is at least 2 orders smaller than the energy gaps of quasiparticle energy spectra. Thus there exists a dominant resonant state which is the highest occupied state of the $N + 1$ electron system and has a contribution to the conductance at least 4 orders larger than any other energy level. This result immediately leads to a well approximated Breit-Wigner type conductance. As shown below, the degeneracy of this state will be "physically" invisible because the addition of an electron to the dot changes the entire energy spectrum completely due to the strong Coulomb interaction. This is contrary to the single electron case in which a peak can be contributed to by multielectrons.

According to the experimental conditions [7], the chemical potential on both sides of the dot can be thought of as being controlled by varying a gate voltage applied to the substrate. The bias between the left and right leads is very small and is neglected here. Suppose that there are N electrons in the dot. As the chemical potential is raised above an experimental threshold, it is initially lower than

the resonant energy level $\epsilon_F^{(N+1)}$, and the Breit-Wigner type conductance will have a peak only when μ coincides with $\epsilon_F^{(N+1)}$. Upon raising the chemical potential μ further, such that $\mu \sim (\epsilon_F^{(N+1)} + \epsilon_F^{(N+2)})/2$, one electron will be trapped in the dot to form a $N+1$ electron system. The next electron tunneling into the dot will interact with the $N+1$ electrons self-consistently and mediate through a new resonant energy level $\epsilon_F^{(N+2)}$. Continuing this sequence, the conductance shows oscillations with Lorentzian line shapes, and each peak in the oscillations corresponds to the addition of one electron to the dot, as confirmed by experiments [7,14].

It is clear that the difference between neighboring resonant energies $\epsilon_F^{(N)}$ and $\epsilon_F^{(N+1)}$ determines the separation between two adjacent peaks (corresponding to N and $N+1$ electrons in the dot, respectively) in the oscillations. The QP Fermi energies for 1–10 and 31–40 electrons are shown in columns (a) and (b) of Fig. 1, respectively. There are two striking features in Fig. 1: a "shell" structure in the spectrum for the range 1–10, and a spectrum of approximately equally separated energy levels in the occupation range 31–40. The shell structure is attributed to the following residue effect from the degeneracy of the single electron energy level: Suppose that the first resonant energy level $\epsilon_F^{(1)}$ is empty. An (first) electron enters the dot and occupies this level. The next (second) incoming electron also occupies this level, but the level has been pushed up to $\epsilon_F^{(2)}$ by the Coulomb repulsion. The third electron has to occupy the next higher level $\epsilon_F^{(3)}$. Since the first and second electrons occupy the same level, the Coulomb repulsion is stronger than that between the third electron and the rest. Thus the gap between $\epsilon_F^{(3)}$ and $\epsilon_F^{(2)}$ is smaller than that between $\epsilon_F^{(2)}$ and $\epsilon_F^{(1)}$. In column (a) of Fig. 1, the first two levels belong to the lowest shell ($\nu = m = 0$), the next four to ($\nu = 0, m = \pm 1$), the 7th and 8th levels to ($\nu = 1, m = 0$), and the last two levels to ($\nu = 0, m = 2$). Although the HF result for small numbers of electrons is not as reliable as that for large numbers, the variation among the gaps in Fig. 1(a) is so large that the shell structure will qualitatively remain in the exact spectra. As the occupation number of electrons increases, the Coulomb interaction is increasingly dominant, and QP energy levels are more and more in balance self-consistently so that the gap in the resonant energy spectrum decreases and the degeneracy of the single electron energy level is smeared out. The latter effect leads to the observation that the oscillations in the conductance gradually become periodic when a sufficient number of electrons ($\gtrsim 30$ in our calculation) are accumulated in the dot. It should be pointed out that the oscillations are not perfectly periodic. There is about a 10% difference in the periods for the occupation range 31–40, which is the reflection of a 10% difference among the energy gaps in column (b) of Fig. 1, and in coincidence with experiment [7].

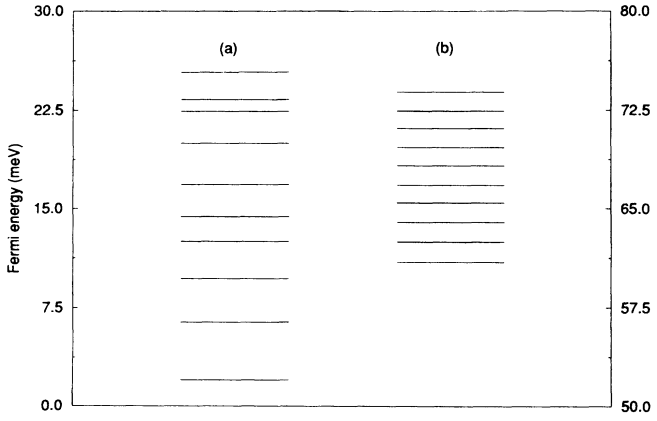

FIG. 1. The Fermi energies calculated in the HF approximation corresponding to the ranges of occupations (a) 1–10 and (b) 31–40 electrons, respectively.

To calculate the conductance at zero temperature, we approximate the elastic coupling using the WKB method. $\mathrm{Im}\Sigma_\alpha^{(c)}$ is calculated by considering the diagonal components of the next order Feynman diagrams beyond those of Hartree-Fock in which the imaginary self-energy due to the elastic coupling, $-i\Gamma/2$, is added in the QP Green's function denominator. A cutoff 0.084 eV is chosen for the total confining potential $0.8V^H + U$, where a factor 0.8 is introduced to account for the reduction from the nonlocal exchange interaction that contributes about 20% on the average. We choose to plot in Fig. 2 ten oscillations of the conductance in the occupation number range 31–40, since this is a region in which the experiments were conducted [7,14]. Several features should be noticed: (1) A pair structure appears in the peak heights. This behavior reflects the even- and odd-occupation numbers and the double degeneracy of two spin freedoms. Consider, for example, the second and third peaks in Fig. 2, which correspond to the occupation numbers 32 and 33, respectively. There is only one conduction channel for the 32nd electron, but two channels for the 33rd electron because of the spin degree of freedom [22]. Besides a factor of 2, the Fermi energy $\epsilon_F^{(33)}$ is closer to the cutoff edge of the total confining potential and the elastic coupling Γ becomes larger so that the third peak is pushed up even higher. This explanation of the alternation in peak heights can be generalized to nonzero magnetic field cases, as can be seen by noticing that there is also a double degeneracy of states ($m + \frac{1}{2}$ and $m + 1 - \frac{1}{2}$) in the presence of a magnetic field. Since a strong field produces an edge state near the boundary of the dot which prevents electrons from approaching, we can argue that the magnetic field "quenches" the disorder near the boundary and makes the alternation structure more regular and explicit in experiments. (2) A pair structure occurs in the linewidths. This can be clearly seen in the last five lines of Fig. 2 corresponding to the occupation numbers 36–40. An even-occupation number gives a configuration of total

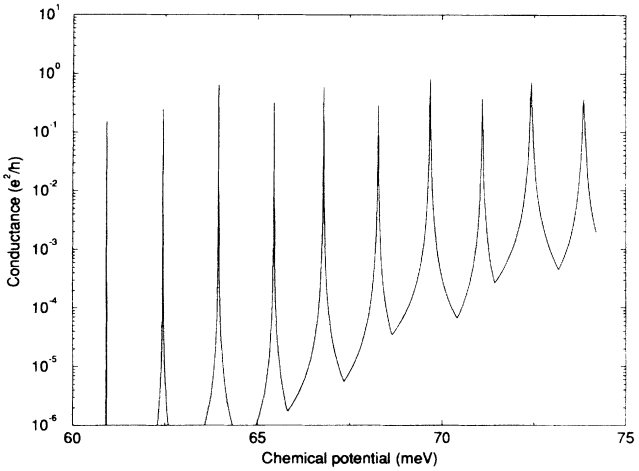

FIG. 2. Ten conductance oscillations as a function of chemical potential corresponding to the electron occupation range 31–40.

spin $\vec{S} = 0$ such that every state has two electrons and their wave functions completely overlap. The Coulomb interaction makes the energy level separation close to the Fermi surface bigger than that for an odd-occupation number. This is similar to a closed shell atom or nucleus. Thus, the resonant state for the occupation numbers 36 [38] has a narrower linewidth, a longer lifetime, and more stability compared with that for 37 [39]. (3) The peak height and the linewidth increase with the chemical potential μ. This result can be understood by noting that the transmission probability continuously increases as the chemical potential approaches the cutoff edge of the total confining potential.

In conclusion, we have proposed a microscopic resonant tunneling theory based on the HF QP energy spectra to quantitatively interpret the observed striking conductance oscillations. The strong Coulomb interaction between electrons in a quantum dot is very important and essential for explaining the quantized electron charging process. The exchange interaction (Fock term) contributes as much as 20% to the QP energies, and cannot be neglected. We predict that the oscillations are not periodic for a small number of electrons in the quantum dot, but will be approximately periodic for a large occupation number. A pair structure (even- and odd-occupation numbers) is found in the peak heights and the linewidths as well as separations between adjacent conductance peaks. A tendency that the peak height and the linewidth increase with increasing chemical potential is revealed. The experimental observations [7,14] are in excellent agreement with our theoretical predictions.

We would like to acknowledge useful discussions with S. Trugman, K. Likharev, and K. B. Efetov, and to thank S. Feng for his fruitful suggestions. Work at Los Alamos is performed under the auspices of the U.S. DOE.

Note added.—More recently, we heard that our three findings concerning the effect of even- and odd-occupation number have been observed in experiments in the absence of a magnetic field by R. Westervelt and colleagues (private communication).

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