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Charge Transfer Induced Persistent Current and Capacitance Oscillations

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The transfer of charge between different regions of a phase-coherent mesoscopic sample is investigated. Charge transfer from a side branch quantum dot into a ring changes the persistent current through a sequence of plateaus of diamagnetic and paramagnetic states. In contrast, a quantum dot embedded in a ring exhibits sharp resonances in the persistent current, whose sign is independent of the number of electrons in the dot if the total number of electrons in the system is even. It is shown that such a mesoscopic system can be polarized appreciably not only by the application of an external voltage, but also via an Aharonov-Bohm flux.

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The transfer of a single electronic charge from one region of a mesoscopic conductor into another region of the conductor can dramatically alter the mesoscopic properties of the conductor. In this work, we take the persistent current of a ring [1–3] as a phase sensitive probe of the equilibrium state of the conductor and investigate its properties under charge transfer. In Fig. 1, two samples are shown in which a ring-like structure is penetrated by an Aharonov-Bohm flux Φ and is connected to a quantum dot. If the sample is brought into an external capacitive circuit it can be polarized; charge transfer from one portion of the sample into the quantum dot can be induced. The charge transfer changes the potential landscape, and with it changes the phase sensitive properties of the mesoscopic sample. Both the electrochemical capacitance $C_\mu = ed\langle Q \rangle/d\mu$ and the flux-induced capacitance $C_\Phi = ed\langle Q \rangle/d\Phi$ are periodic functions of the AB-flux [4]. For the samples of Fig. 1, we find indeed very striking flux sensitive features in these capacitance coefficients. Measurement of such capacitance coefficients provides an important alternative to the difficult magnetization measurements [2] used to characterize the ground state of mesoscopic samples.

A purely one-dimensional ring exhibits a persistent current which is either diamagnetic or paramagnetic depending on the number of particles and their distribution over the flux sensitive states [5]. The persistent current is always an odd function of flux $I(\Phi) = -I(-\Phi)$. But the slope of the persistent current $dI(\Phi)/d\Phi$ for a small flux can be either negative (diamagnetic) or positive (paramagnetic). To be brief, we say that a diamagnetic ground state has a positive parity and a paramagnetic state has a negative parity. If we consider the contribution to the persistent current of each spin class separately, then the addition of a single electron changes the parity of its spin class [5]. For the sample in Fig. 1a, in which the dot acts as a fully coherent reservoir of carriers, charge transfer thus induces sharp transitions between plateaus of diamagnetic and paramagnetic states.

For the ring of Fig. 1b, the persistent current is suppressed by charging effects unless the conditions for res-

onant charge transfer are met, an effect analogous to the *Coulomb blockade* observed in the conductance through a quantum dot coupled to macroscopic leads [6–8]. The charge transfer discussed here should, however, be distinguished from the standard discussions of the Coulomb blockade [7], which treat charge transfer incoherently. Here we deal with coherent many-body states which are extended over *multiple* regions [9–11]. The surprising effect which we find for the ring of Fig. 1b is that the sign of the persistent current contributed by each spin class is *independent* of the number of electrons in the dot. The parity of each spin class is conserved under charge transfer, and is determined only by the total number of electrons in the sample, regardless of whether these electronic

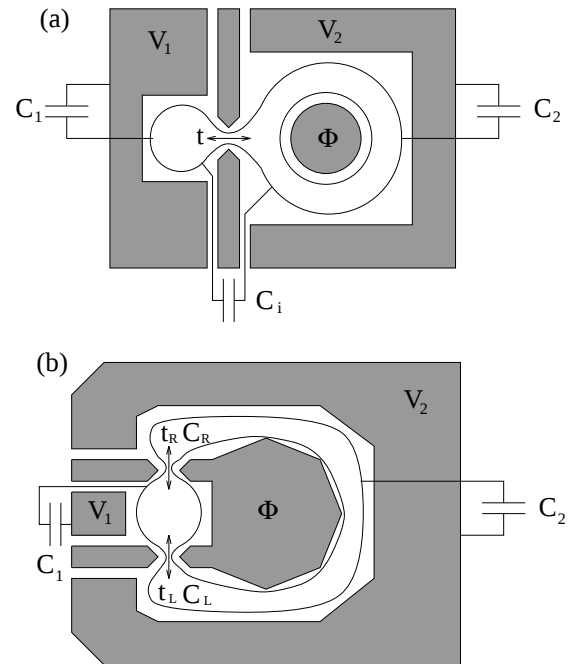


FIG. 1. (a) Ring with Aharonov-Bohm flux coupled to a side branch quantum dot. (b) Quantum dot with leads closed into a loop.

states are localized or whether the states are extended and contribute to the persistent current. In contrast, the sample in Fig. 1a changes its parity with each electron that is transferred from the dot into the ring.

The geometry of Fig. 1a has been the subject of Refs. [4,12]. A recent experiment [13] and theory [14] investigated the AB effect of a quantum dot embedded in a loop and connected to two leads. Here we treat explicitly the *capacitively-coupled* closed structures and analyze the charge response.

The system of Fig. 1 is modeled in terms of a one-dimensional ring coupled capacitively and via tunneling to a quantum dot. The electron-electron interactions in the system are treated using a capacitive charging model, as indicated in Fig. 1: The system is coupled to two external metallic gates at voltages V_1 and V_2 with capacitance coefficients C_1 and C_2 . In addition, the quantum dot couples to the ring with capacitance C_i ($= C_R + C_L$ for the case shown in Fig. 1b). With the combined capacitances $C_e^{-1} = C_1^{-1} + C_2^{-1}$ and $C = C_e + C_i$, we can express the electrostatic Hamiltonian (which includes the work done by the voltage sources V_1 and V_2) in terms of the charge operator for the dot $Q = \sum_{n\sigma} d_{n\sigma}^\dagger d_{n\sigma}$, a polarization charge $Q_0 = C_e V$, and the externally applied voltage $V = V_2 - V_1$,

$$H_C = (1/2C)(Q - Q_0)^2 - (C_e/2)V^2. \quad (1)$$

The total Hamiltonian for the system is $H = H_0 + H_T + H_C$, where $H_0 = \sum_{k\sigma} \epsilon_{ak} c_{k\sigma}^\dagger c_{k\sigma} + \sum_{n\sigma} \epsilon_{dn} d_{n\sigma}^\dagger d_{n\sigma}$ describes the single-particle eigenstates in the ring and the dot, and the tunneling Hamiltonian is $H_T = \sum_{kn\sigma} (t_{kn} d_{n\sigma}^\dagger c_{k\sigma} + \text{H.c.})$. For the system of Fig. 1a, the AB-flux modulates the single-particle energy levels ϵ_{ak} in the ring, while for the system of Fig. 1b, the tunneling matrix elements t_{kn} connecting the dot to the ring are flux dependent. H_C favors integer charge states of the quantum dot [6–8], whereas H_T promotes hybridization of the localized states on the dot with the extended states of the ring. Our Hamiltonian is similar to the Anderson model [15] for a magnetic impurity (or quantum dot [16]) coupled to a Fermi sea of conduction electrons, but here the reservoir of conduction electrons is itself a mesoscopic system with a finite level spacing and bandwidth. In order to account for the tendency toward charge quantization in the system, H_C must be treated nonperturbatively. We therefore employ two complementary approaches: In the weak-tunneling limit, where hybridization occurs only between a single state in the ring and in the dot, H can be reduced to a 2×2 matrix (3×3 including spin), allowing for an explicit solution. This simple analytical solution correctly describes the interesting parity effects in the system. The ground state is also found exactly for arbitrary coupling using a numerical Lanczos technique.

Figs. 2a and b show numerical results for the persistent current $I = -cdE_0/d\Phi$ and the electrochemical capaci-

tance $C_\mu = -d^2E_0/dV^2$ of the systems of Figs. 1a and b, respectively, as a function of the polarization charge Q_0 . Here E_0 was evaluated computationally, with the single-particle energy levels ϵ_{ak} and ϵ_{dn} in the ring and dot and the tunneling matrix elements t_{kn} modeled using a one-dimensional tight-binding model in which the dot was represented by 2 sites, and the ring by 4 sites. The matrix element w of the kinetic energy operator between nearest-neighbor sites within the ring and the dot was taken to be unity, and the point contacts were modeled as weak links. For the case of 3 up-spin and 3 down-spin electrons, the persistent current of the quantum dot within the loop is diamagnetic, while the loop with a side branch quantum dot exhibits a sequence of plateaus of diamagnetic and paramagnetic states. The four peaks in C_μ in Figs. 2a,b, separated by $\Delta Q_0 \sim e$, correspond to the successive transfer of electrons from the ring to the dot (for decreasing Q_0), filling the four available single-particle states in the dot.

In order to understand the character of the charge transfer induced oscillations in I and C_μ , it is useful to consider the limit $t_{kn} \ll \Delta\epsilon$, e^2/C , where the dot and the ring are only weakly coupled. Then, in the vicinity of the charge transfer resonance $N \rightarrow N + 1$, where N

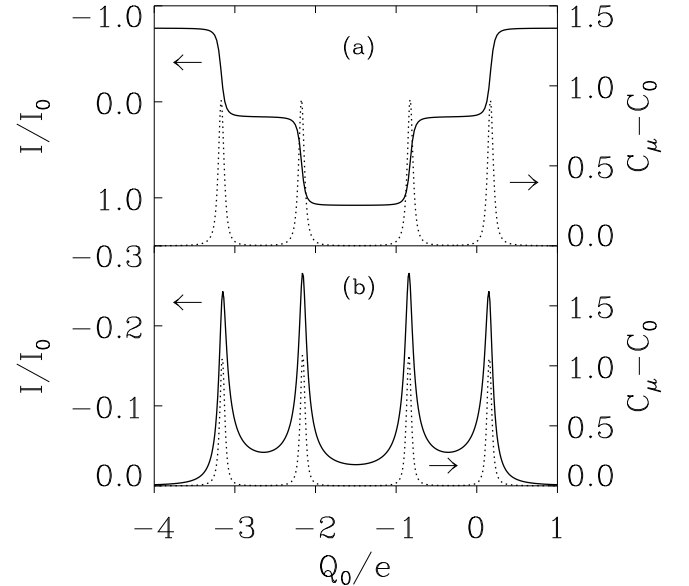


FIG. 2. Persistent current and differential capacitance as a function of the polarization charge $Q_0 = C_e V$ for (a) the sample of Fig. 1a with $t = 0.5$ and (b) the sample of Fig. 1b with $t_R = 0.2$ and $t_L = 0.3$. Each sample contains 6 electrons, with $\Phi/\Phi_0 = 1/4$, and $e^2/C = 10$. Energy is expressed in units of w , $4w$ being the bandwidth in the ring. The persistent current (solid curve) is expressed in units of $I_0 = \max e v_F / L$, where L is the circumference of the ring, and the capacitance (dotted curve) is expressed in units of $(C_e/C)^2 e^2 / w$. Each peak in C_μ corresponds to the transfer of one electron from the dot to the ring.

is the number of electrons in the dot, one need only consider the hybridization of the highest occupied level $|aM\rangle$ in the ring with the lowest unoccupied level $|d(N+1)\rangle$ in the dot. Neglecting spin (the effects of which will be considered further below), the Hamiltonian then reduces to a 2×2 matrix,

$$H_h = \begin{pmatrix} \epsilon_{aM} + \frac{(\epsilon_{aM} + Q_0)^2}{2C} & t \\ t^* & \epsilon_{d(N+1)} + \frac{[\epsilon_{d(N+1)} + Q_0]^2}{2C} \end{pmatrix} \quad (2)$$

plus an additive constant

$$E_1 = \sum_{k=1}^{M-1} \epsilon_{ak} + \sum_{n=1}^N \epsilon_{dn} - C_e V^2/2, \quad (3)$$

where $M+N$ is the total number of (spinless) electrons in the system. For the system of Fig. 1b, the matrix element t depends on the total number of nodes $M+N-1$ in the wave functions $|aM\rangle$ and $|d(N+1)\rangle$: its modulus squared is given by

$$|t_{\pm}|^2 = t_R^2 + t_L^2 \pm 2t_R t_L \cos(2\pi\Phi/\Phi_0), \quad (4)$$

where the $+$ sign holds for $M+N-1$ even, and the $-$ sign holds for $M+N-1$ odd. Here $\Phi_0 = hc/e$ is the single-charge flux quantum and $t_{R/L}$ are energies proportional to the transmission amplitudes through the two point contacts. The hybridization of the localized state of the dot with the extended state of the ring is a maximum when the polarization charge takes the value

$$Q_* = -e(N+1/2) + (C/e)[\epsilon_{aM} - \epsilon_{d(N+1)}]. \quad (5)$$

Note that this is precisely the polarization charge which would be needed to transfer an electron in the classical approach to the Coulomb blockade. For this polarization charge, in the classical case, the energy has the form of a cusp. In the quantum mechanical case, the ground state energy is a smooth function of the polarization charge,

$$E_0 = E_1 + \frac{\epsilon_{aM} + \epsilon_{d(N+1)}}{2} + \frac{e^2}{8C} + \frac{[e(N+1/2) + Q_0]^2}{2C} - \frac{1}{2} \left(\left[\frac{e}{C}(Q_0 - Q_*) \right]^2 + 4|t_{\pm}|^2 \right)^{1/2}. \quad (6)$$

Due to quantum mechanical tunneling, the energy barrier is lower. Note that after transfer of an electron to the dot the next hybridization will take place between the state $|a(M-1)\rangle$ of the ring and the state $|d(N+2)\rangle$ of the dot. The total number of nodes $(M-1) + (N+2) - 2 = M+N-1$, which determines the parity of the system, is left invariant. Let us next explore a few consequences of this simple result.

Differentiating Eq. (6), one obtains the persistent current for the sample of Fig. 1b,

$$I(\Phi) = \mp \frac{e}{\hbar} \frac{4\pi t_R t_L \sin(2\pi\Phi/\Phi_0)}{[e(Q_0 - Q_*)/C]^2 + 4|t_{\pm}|^2}^{1/2}. \quad (7)$$

The persistent current is a sharply peaked function of the polarization charge, obtaining a maximum value of $I_{\max} = e \partial|t_{\pm}|/\partial\Phi$ at $Q_0 = Q_*$, and being of order $(e/\hbar)[t_R t_L/(e^2/C)]$ far from resonance. The parity of $I(\Phi)$ is determined by the matrix element $t_{\pm}$, and is independent of the polarization charge Q_0 . This result is a consequence of the Friedel sum rule [14], which links the phase acquired by an electron traversing the system to the *total* charge in the system, which is invariant under polarization. Consequently, the parity effects on the persistent current described here for the case of a strictly one-dimensional ring are expected to be quite general. Eq. (7) indicates that the peaks in the persistent current exhibit long non-Lorentzian tails away from resonance due to charge fluctuations on the quantum dot, as is evident in Fig. 2b.

The charge on metallic gate 1 is determined by $Q_e = -dE_0/dV$. The electrochemical capacitance between gates 1 and 2 is thus $C_\mu = -d^2 E_0/dV^2$. From Eq. (6), we find

$$C_\mu - C_0 = \frac{2e^2(C_e^2/C^2)|t_{\pm}|^2}{([e(Q_0 - Q_*)/C]^2 + 4|t_{\pm}|^2)^{3/2}}, \quad (8)$$

where $C_0^{-1} = C_e^{-1} + C_i^{-1}$ is the classical series capacitance. The total change of the charge on gate 1 integrated over such a charge transfer resonance (excluding the contribution from C_0) is $|\Delta Q_e| = e(C_e/C)$, corresponding to the transfer of one electron between ring and dot. The quantum corrections to C_μ reach a maximum of $(eC_e/2C)^2/|t_{\pm}|$ at $Q_0 = Q_*$, and are of order $C[|t_{\pm}|/(e^2/C_e)]^2$ far from resonance, decreasing faster than a Lorentzian. The coherent backscattering in such a phase-coherent system thus leads to a suppression of charge transfer away from resonance vis à vis a system with incoherent charge transfer, such as that studied by Ashoori *et al.* [17] or Lafarge *et al.* [18], which would be expected to exhibit Lorentzian peaks at zero temperature. That is to say, coherence suppresses charge fluctuations of the type $\delta Q = \langle Q - Ne \rangle$, which contribute to the capacitive response of the system, while enhancing charge fluctuations of the type $\delta Q = (\langle Q^2 \rangle - \langle Q \rangle^2)^{1/2}$, which govern the persistent current. The parity of $t_{\pm}$ determines the phase of the AB-effect on C_μ , which exhibits a phase-shift of π on resonance.

Let us next briefly describe how the above results change for the loop with the side branch quantum dot. If the loop and the side dot are disconnected, the ring supports flux dependent states with energies $\epsilon_{ak}(\Phi)$ whereas the dot supports flux independent states with energies ϵ_{dn} . Thus, for this system, we have a persistent current $I(\Phi)$ even in the absence of coupling to the dot. To take the Coulomb interaction into account in the presence of a weak coupling to the dot, we again need only consider the hybridization of the topmost electron in the ring with the lowest empty state in the dot. For the energy of the

topmost electron, this leads to an eigenvalue problem of the same form as Eq. (2), but now with coupling matrix elements t which are independent of flux. The total energy is of the same form as Eq. (6), except that the flux dependence is now determined by the states of the uncoupled ring.

The sensitivity of the persistent current to changes in the gate voltage can be characterized by the flux-induced capacitance [4] C_Φ . This capacitance is measured in response to an oscillating AB-flux $d\Phi(\omega) \exp(-i\omega t)$ superimposed on the static AB-flux, and is given by $C_\Phi = -d^2 E/d\Phi dV = -(1/c)dI(\Phi)/dV$. The flux-induced capacitance is, like the persistent current, an odd function of flux. It has a particularly interesting behavior for the system of Fig. 1a, for which case it takes the form

$$C_\Phi = \frac{4t^2 e(C_e/C) d\epsilon_{aM}(\Phi)/d\Phi}{(\{e[Q_0 - Q_*(\Phi)]/C\}^2 + 4t^2)^{3/2}} \quad (9)$$

near resonance. Because Q_* is now a function of the AB-flux Φ , one can pass through the charge transfer resonance by varying Φ . Integrating Eq. (9) with respect to Φ , one finds $|\Delta Q_e| = e(C_e/C)$ for the case where the bandwidth in the ring is large compared to t , corresponding to the transfer of one electron between ring and dot.

So far we have neglected spin. For the sample of Fig. 1b, the discussion given above still applies in the vicinity of a single resonance for the case when there are an unequal number of up-spin and down-spin electrons in the system. However, the parity of the persistent current on resonance is then determined by the spin of the electron being transferred. If the up-spin and down-spin systems have different parity, this leads to resonances of *alternating* sign in the persistent current. For equal numbers of up-spin and down-spin electrons, the ground state forms a Kondo singlet. In the weak-coupling limit, the Hamiltonian reduces to a tridiagonal 3×3 matrix similar to Eq. (2), where the diagonal terms give the energies of the three possible charge states in the absence of tunneling, and the terms nearest the diagonal are $\sqrt{2}t_\pm$ and $\sqrt{2}t_\pm^*$. This leads to an enhancement of the persistent current on resonance by a factor of $\sqrt{2}$ compared to Eq. (7), and an enhancement by a factor of 2 midway between the two resonances. In such a system, the parity of the persistent current is again invariant under charge transfer, as illustrated in Fig. 2b.

The transfer of a single electronic charge from one region of a mesoscopic conductor into another region of the conductor can dramatically alter the mesoscopic properties of the conductor. In this work we have taken the persistent current as an example. We have emphasized that the measurement of capacitance coefficients C_μ and C_Φ provides an interesting possibility to characterize the ground state of such closed systems. The charge transfer in quantum-coherent mesoscopic conductors or large molecules thus provides a very interesting future avenue of research.

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