Quantum inductance and negative electrochemical capacitance at finite frequency in a two-plate quantum capacitor

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We report on theoretical investigations of frequency-dependent quantum capacitance. It is found that at finite frequency, a quantum capacitor can be characterized by a classical *RLC* circuit with three parameters: a static electrochemical capacitance, a charge relaxation resistance, and a quantum inductance. The quantum inductance is proportional to the characteristic time scale of electron dynamics, and due to its existence, the time-dependent current can accumulate a phase delay and lags behind the applied ac voltage, leading to a negative effective capacitance.

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Understanding dynamic conductance of quantum coherent conductors is a very important problem in nanoelectronics theory. When two quantum coherent conductors form a double plate "quantum capacitor," its dynamic conductance $G(\omega)$ is given by the frequency-dependent electrochemical capacitance[1–3] $C_\mu(\omega)$, $G(\omega)=-i\omega C_\mu(\omega)$, where ω is the frequency. At low frequency, $C_\mu(\omega)$ can be expanded in frequency, and at the linear order, it is described[2] by an equivalent classical circuit consisting of a static capacitor C_μ in series with a "charge relaxation resistor" R_q. For a conductor having a single spin-resolved transmission channel, R_q was predicted[2] to be half the resistance quantum, $R_q=1/2G_o$, where $G_o\equiv h/e^2$. The factor 1/2 in R_q is of quantum origin[2,4] and has recently been confirmed experimentally.[5] In the experiment of Gabelli *et al.*,[5] a submicron two-dimensional electron-gas quantum dot (QD) is capacitively coupled to a gold plate forming a double plate capacitor, where the QD connects to the outside reservoir by a single-channel quantum point contact (QPC). The dynamic conductance $G(\omega)$ is then measured at 1.2 GHz, and the data are well fit to the equivalent circuit characterized by two parameters (C_μ,R_q).

The experiment of Gabelli *et al.*[5] opened the door for elucidating important and interesting physics of high-frequency quantum transport in meso- and nanoscale devices. An important question is what happens to electrochemical capacitance at higher frequencies beyond the linear ω regime, and, in particular, whether the two-parameter (C_μ,R_q) equivalent circuit is adequate at higher frequencies to describe a quantum capacitor. It is the purpose of this paper to address these issues.

In the following, we report a microscopic theory for high-frequency quantum transport in a two-plate quantum capacitor. Our results show that to characterize its high-frequency dynamic response, one needs—in addition to C_μ and R_q—another quantity L_q having the dimension of inductance. L_q is found to have purely quantum origin and will be named "quantum inductance." Therefore, the frequency dependent electrochemical capacitance of a quantum capacitor $C_\mu(\omega)$ is equivalent to a classical *RLC* circuit characterized by three parameters (C_μ,R_q,L_q) at high frequency. Due to L_q, electrons dwell in the neighborhood of the capacitor plates causing a phase delay. At low frequencies, the dynamic response is capacitivelike and voltage lags current. At larger frequencies when $\omega>1/\sqrt{C_\mu L_q}$, inductive behavior dominates and voltage leads current: in this case, the quantum capacitor gives a negative capacitance value. It is, indeed, surprising that a quantum capacitor can give an inductive dynamic response. For the experimental setup of Ref. 5, we estimate that when $\omega\sim 3$ GHz, the predicted high-frequency effects should be observable.

Let us first work out a simple expression for the frequency-dependent electrochemical capacitance $C_\mu(\omega)$ following the work of Büttiker.[2] We consider a two-plate capacitor similar to the experiment of Gabelli *et al.*:[5] a QD, labeled I, and a large metallic electrode, labeled II. Each plate is connected to the outside world through its lead, and a time-dependent bias $v_{1,2}$ is applied across the two leads. We consider small amplitudes of $v_{1,2}$ so as to focus on the linear bias regime. Under the action of such a bias, the two capacitor plates develop their own frequency-dependent electric potential $U_{\mathrm{I,II}}(\omega)$. The charge on plate I is equal to the sum of the injected charge and induced charge: $Q_{\mathrm{I}}=Q_{\mathrm{I}}^{inj}+Q_{\mathrm{I}}^{ind}$. In the linear regime, the injected charge is proportional to bias $v_1(\omega)$: $Q_{\mathrm{I}}^{inj}=e^2D_{\mathrm{I}}(\omega)v_1(\omega)$, where $D_{\mathrm{I}}(\omega)$ is the generalized global density of states (DOS) of plate I at frequency ω. The induced charge, on the other hand, is in general related to a nonlocal Lindhard function[2] whose calculation is simplified by applying the Thomas-Fermi approximation. Within this approximation, the induced charge is proportional to the induced potential U_{I} on the plate: $Q_{\mathrm{I}}^{ind}=-e^2D_{\mathrm{I}}(\omega)U_{\mathrm{I}}$, where the minus sign indicates that the charge is induced. Putting things together, we obtain $Q_{\mathrm{I}}=e^2D_{\mathrm{I}}(\omega)(v_1-U_{\mathrm{I}})$. Clearly, the same charge Q_{I} can be calculated by the usual electrostatic geometric capacitance C_o: $Q_{\mathrm{I}}=C_o(U_{\mathrm{I}}-U_{\mathrm{II}})$. We therefore obtain a relationship: $C_o(U_{\mathrm{I}}-U_{\mathrm{II}})=e^2D_{\mathrm{I}}(\omega)(v_1-U_{\mathrm{I}})$. Applying the same argument to plate II, we similarly obtain $-C_o(U_{\mathrm{I}}-U_{\mathrm{II}})=e^2D_{\mathrm{II}}(\omega)(v_2-U_{\mathrm{II}})$. Finally, the same charge Q_{I} can also be obtained from the definition of the electrochemical capacitance: $Q_{\mathrm{I}}=C_\mu(\omega)(v_1-v_2)$. These three relations allow one to derive the following expression for $C_\mu(\omega)$:

$$\frac{e^2}{C_\mu(\omega)} = \frac{e^2}{C_o} + \frac{1}{D_{\mathrm{I}}(\omega)} + \frac{1}{D_{\mathrm{II}}(\omega)}. \tag{1}$$

This result resembles the one obtained by Büttiker for the static capacitance.[2] An important difference is that the frequency-dependent electrochemical capacitance in Eq. (1) is a complex quantity: its real part is a measure of the electrochemical capacitance and its imaginary part is proportional to the frequency-dependent charge relaxation resistance.

To proceed further, we need to calculate the frequency-dependent DOS $D_{\mathrm{I,II}}(\omega)$. Following Ref. 4, the generalized local DOS of plate α of the capacitor can be expressed in terms of Green's functions as follows:

$$\frac{dn_\alpha(\omega)}{dE} = \int \frac{dE}{2\pi}\frac{f-\bar{f}}{\hbar\omega}[\bar{G}^r\Gamma_\alpha G^a]_{xx}, \tag{2}$$

where the subscript x labels space coordinates and the global DOS is given by

$$D_\alpha(\omega) = \mathrm{Tr}\int \frac{dE}{2\pi}\frac{f-\bar{f}}{\hbar\omega}[\bar{G}^r\Gamma_\alpha G^a].$$

In Eq. (2), f is the Fermi function and $\bar{f}\equiv f(E_+)$, with $E_+\equiv E+\hbar\omega$; $G^{r,a}=G^{r,a}(E)$ is the retarded and/or advanced Green's function at energy E and $\bar{G}^r\equiv G^r(E+\hbar\omega)$; and Γ_α is the linewidth function describing the coupling strength between plate α and its lead. These quantities can be calculated in a straightforward manner when the Hamiltonian of the capacitor model is specified.[6,7]

For the quantum capacitor of Ref. 5, plate I is a QD and plate II is a metal gate. Since the metal gate has much greater DOS, i.e., $D_{\mathrm{II}}\gg D_{\mathrm{I}}$, we can safely neglect the D_{II} term in Eq. (1). For a QD with one energy level E_0 and connected to one lead, its Green's function $G^r=1/(E-E_0+i\Gamma_L/2)$, where Γ_L is the linewidth function of the lead. With this Green's function, the frequency-dependent DOS can be easily calculated from Eq. (2), and we obtain

$$D_{\mathrm{I}}(\omega) = \frac{\Gamma_L}{2\pi\hbar\omega(\hbar\omega+i\Gamma_L)}\left[\frac{1}{2}\ln\frac{\Delta^2}{\Delta_+\Delta_-} - i\left(\arctan\frac{\Delta E-\hbar\omega}{\Gamma_L/2} - \arctan\frac{\Delta E+\hbar\omega}{\Gamma_L/2}\right)\right], \tag{3}$$

where $\Delta=\Delta E^2+\Gamma_L^2/4$, $\Delta_\pm=(\Delta E\pm\hbar\omega)^2+\Gamma_L^2/4$, and $\Delta E=E_F-E_0$. At resonance $\Delta E=0$, we obtain $\mathrm{Re}(D_{\mathrm{I}}(\omega))=[-x\ln(4x^2+1)+2\arctan(2x)]/[2\pi\Gamma_L x(x^2+1)]$, with $x=\hbar\omega/\Gamma_L$. Hence, $\mathrm{Re}(D_{\mathrm{I}}(\omega))$ is positive for small x and negative for large x, i.e., there is a sign change. A similar behavior is also found for the system away from the resonance. Due to this sign change of $\mathrm{Re}(D_{\mathrm{I}})$, from Eq. (1), the frequency-dependent electrochemical capacitance $C_R\equiv\mathrm{Re}[C_\mu(\omega)]$ can become negative.

To be more specific, we fix the classical capacitance of QD $C_0=1$ fF, which is a typical value for QD with an area of $\sim1\ \mu\mathrm{m}^2$. Figure 1 plots $C_R=\mathrm{Re}[C_\mu(\omega)]$ (real part) and $C_I=\mathrm{Im}[C_\mu(\omega)]$ (imaginary part) versus frequency for different values of Γ_L by setting ΔE and temperature to zero. We observe that C_R is positive at small frequency and becomes negative at larger frequency. For instance, C_R becomes negative at a "critical" frequency $\omega_c\sim10$ GHz for $\Gamma_L=10\ \mu$eV. This critical frequency can be smaller for smaller linewidth function Γ_L. We note that it is not difficult to achieve $\Gamma_L=10\ \mu$eV experimentally: in Ref. 8, Γ_L between 1 and 5 μeV has been realized. As will be discussed below, the effective Γ_L in the experiment of Ref. 5 is tunable by a gate voltage, so that the critical frequency at which the negative capacitance occurs can be even smaller. The inset of Fig. 1 also shows that as we increase ω, the imaginary part of $C_\mu(\omega)$ starts from zero, reaches a peak value around ω_c, and then decays to zero. The negative capacitance at large frequency can be understood as follows. For a classical capacitor, a charge is accumulated across the capacitor induced by an external voltage. The current and voltage have a fixed phase relationship: the voltage lags behind current with a phase $\pi/2$. For a quantum capacitor at low frequency, there exists a charge relaxation resistance $R_q=h/(2e^2)$ for a single channel plate; therefore, the charge buildup time is the RC time $\tau_{RC}=R_qC_\mu$. For $C_\mu=1$ fF and $R_q=h/(2e^2)$, this RC time is about $\tau_{RC}=13$ ps. If the external voltage reverses sign, the charge accumulation will follow the voltage and will also reverse sign in due time. When frequency is low, namely, when $\omega\ll1/\tau_{RC}=77$ GHz, the charge buildup follows the ac bias almost instantaneously just like a classical capacitor; thus, the capacitance is positive. When frequency is high, there is a phase difference between the ac bias and the charge buildup. For frequencies comparable to $1/\tau_{RC}$, the charge buildup cannot follow the ac bias; thereby, the capacitance may be negative.

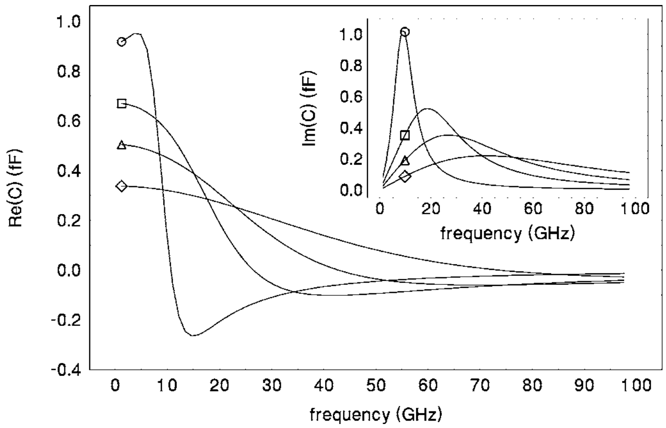

FIG. 1. Frequency-dependent capacitance $C_\mu(\omega)$ versus frequency. Main figure is for C_R and inset is for C_I. Here, $\Gamma_L=10\ \mu$eV (for the curve with the open circle), 50 μeV (open square), 100 μeV (open triangle), and 200 μeV (open diamond). Here, $C_o=1$ fF.

While the above argument explains why it is possible to have negative capacitance, it would indicate a critical frequency to be near 77 GHz. Our results (Fig. 1) show that the calculated ω_c is actually much smaller. This is because there exists a second relevant time scale in the QD, i.e., the dwell time τ_d which is the time spent by electrons inside the QD. The dwell time τ_d can be calculated for specific systems.[9] Importantly, at resonance, the electrons can dwell inside the

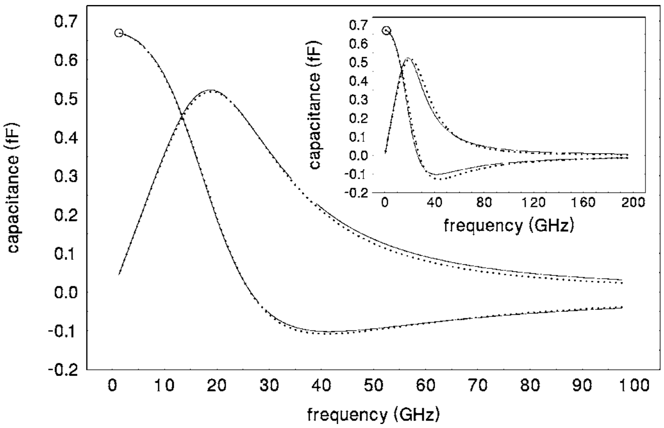

FIG. 2. Comparison of the full quantum capacitance $C_\mu(\omega)$ (solid line) to that obtained by the classical *RLC* circuit model (dotted line) for $\Gamma_L=50$ μeV. Here, $\mathrm{Re}[C_\mu(\omega)]$ is indicated by open circle. Inset: similar fit but using three constant parameters.

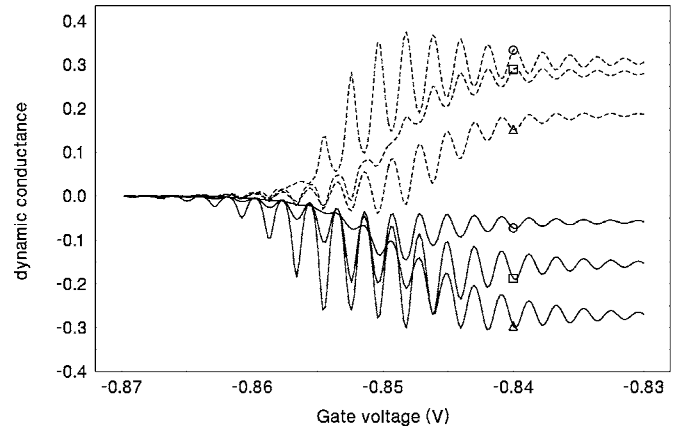

FIG. 3. The dynamic conductance $G(\omega)$ (unit e^2/h) versus gate voltage at different frequencies $\omega=1.2$, 3, and 5 GHz. Solid line, $\mathrm{Re}[G]/\omega$; dotted line, $\mathrm{Im}[G]/\omega$. Other parameters: $\Delta=500$ mK, $\alpha=3000$, $V_0=-0.85$, $\Delta V_0=0.003$, $C_0=4$ fF, and temperature 50 mK. For purpose of illustration, we divided $G(\omega)$ by ω. Inset: frequency-dependent R_q (unit h/e^2) vs frequency.

quantum dot for a long time. For instance, for our QD with $\Gamma_L=10$ μeV, we found $\tau_d=260$ ps while $\tau_{RC}=12$ ps (since $C_\mu=0.9$ fF). In other words, $\tau_d \gg \tau_{RC}$. Such a τ_d corresponds to a frequency of 4 GHz, much less than $1/\tau_{RC}$. In other words, when ac frequency is greater than $1/\tau_d$, the charges dwell inside the quantum dot and cannot respond to the ac voltage change. As a result, current lags behind voltage, leading to a negative capacitance.[10] This picture agrees very well with the numerical results (Fig. 1).

Having determined the general behavior of $C_\mu(\omega)$ for the quantum capacitor, in the following, we determine how to simulate this quantity using a classical circuit. Expanding Eq. (1) into a Taylor series to second order in ω with the help of Eq. (3) at resonance, we obtain

$$C_\mu(\omega) = C_\mu + i\omega C_\mu^2 \frac{h}{2e^2} - \omega^2 C_\mu^3 \frac{h^2}{4e^4} + \omega^2 C_\mu^2 \frac{h^2}{12\pi\Gamma_L e^2}, \tag{4}$$

where $C_\mu=C_\mu(0)$ on the right-hand side is the static electrochemical capacitance. This result is equivalent to that of a classical *RLC* circuit as follows. For a classical *RLC* circuit with capacitance C_μ, resistance R_q, and inductance L_q, the dynamic conductance is

$$G(\omega) = -i\omega C_\mu/(1-\omega^2 L_q C_\mu - i\omega C_\mu R_q). \tag{5}$$

Expanding this expression in power series of ω, we obtain

$$G(\omega) = -i\omega C_\mu + \omega^2 C_\mu^2 R_q + i\omega^3 C_\mu^3 R_q^2 - i\omega^3 C_\mu^2 L_q. \tag{6}$$

Because for a capacitor $G(\omega)=-i\omega C_\mu(\omega)$, we obtain the result that our quantum capacitance [Eq. (4)] is equivalent to the classical *RLC* circuit model of Eq. (6). Comparing these two equations, we readily identify $R_q=h/(2e^2)$—a result first obtained by Büttiker *et al.*.[2] Importantly, another quantity—the equivalent inductance—is identified as $L_q=h^2/(12\pi e^2\Gamma_L)$. In terms of dwell time τ_d and charge relaxation resistance R_q, we obtain

$$L_q = R_q \tau_d/12, \tag{7}$$

where $\tau_d=4\hbar/\Gamma_L$.

What is the reason that a quantum capacitor at finite frequency needs to be modeled by a classical *RLC* circuit (instead of a *RC* circuit)? This is due to the role played by the large dwell time τ_d of the QD. When electrons dwell for long time τ_d inside the QD, the interaction between electrons becomes an important piece of physics which, in our theory, is modeled by the induced self-consistent potential $U_{\mathrm{I,II}}$ discussed above. Such an interaction gives rise to the physics of induction, resulting to the quantity L_q of Eq. (7). Indeed, the explicit dependence on τ_d by L_q also confirms the important role played by the dwell time. Because L_q is determined by τ_d, as well as fundamental constants h and e, it is of purely quantum origin and can be called quantum inductance.

Figure 2 compares the fitting of classical *RLC* circuit [Eq. (5)], with the full quantum result of Eq. (1). They compare very well for the entire range of the frequency—if we treat R_q as a function of ω. Indeed, while R_q has so far been a constant $h/(2e^2)$ as identified through the Taylor expanded equations [Eqs. (4) and (6)], it is actually a function of ω by the more general expression Eq. (5). The inset of Fig. 3 plots the general $R_q=R_q(\omega)$ obtained numerically, and we observe it to be a slowly increasing function of ω. As expected, in the small frequency limit, $R_q(\omega)$ recovers the result of half resistance quantum. For $\Gamma_L=50$ μeV, $R_q(\omega)$ deviates from $h/2e^2$ at about 5 GHz. We have also attempted using three *constant* parameters C_μ, L_q, and $R_q=h/(2e^2)$ into Eq. (5) to compare with the full quantum result of Eq. (1), and a reasonable agreement is obtained (inset of Fig. 2) although not as good as that shown in Fig. 2.

The situation is somewhat different for quantum inductance L_q when the system is *off* resonance [$\Delta E \neq 0$ in Eq. (3)]. In this case, the dwell time τ_d becomes too small to be relevant and another time scale becomes important, namely, the tunneling time τ_t for electrons to go in and/or out of the

QD. The further away E is from E_0, the longer is τ_t. Hence, in Eq. (7), τ_d should be replaced by τ_t for off resonance. Our result shows that the fitting of full quantum capacitance $C_\mu(\omega)$ using classical parameters C_μ, L_q, and $R_q(\omega)$ is still quite good for off resonance. This further supports the conclusion that the frequency-dependent quantum capacitance can be described by a classical *RLC* circuit with static electrochemical capacitance, charge relaxation resistance, and a quantum inductance.

Note that our result [Eq. (7)] is obtained using Lorentzian line shape for the Green's function that is a good approximation at low frequency. Now, we compare our result with the high-frequency admittance for a single-channel wire against a macroscopic gate calculated using the Luttinger model.[13] The low-frequency expansion of gate capacitance is given in the last equation of Ref. 13, from which we can identify L_q to be

$$L_q = \frac{1}{3g^2} C_\mu R_q^2, \tag{8}$$

where the interaction parameter g is related to electrochemical capacitance via

$$C_\mu = \frac{Lg^2}{R_q v_F}, \tag{9}$$

with L the length of the wire and v_F the Fermi velocity. Substituting Eq. (9) into Eq. (8), we have

$$L_q = \frac{L}{3v_F} R_q. \tag{10}$$

We see that if we identify $\tau_d \sim 4L/v_F$, Eqs. (7) and (10) are the same. In another word, a spinless Luttinger liquid has a capacitance with a high-frequency inductance of the same form as in Eq. (7).

Finally, we perform a numerical calculation of the dynamic conductance for the device structure of Ref. 5. In terms of scattering matrix, the DOS of Eq. (2) for a capacitor can be rewritten as[4,6]

$$D_{\mathrm{I}}(\omega) = i \int \frac{dE}{2\pi\hbar^2\omega^2} \frac{f-\bar{f}}{} [1 - s_{LL}^\dagger(E_+) s_{LL}(E)], \tag{11}$$

with[5] $s_{LL}^\dagger(E) = (r - e^{i\phi})/(1 - re^{i\phi})$, $\phi = 2\pi E/\Delta$, $r^2 = 1 - T_{QPC}$, and $T_{QPC} = 1/[1 + \exp(-(V_g + V_0)/\Delta V_0)]$, which is the transmission coefficient of the QPC in the experimental setup.[5] Figure 3 shows the dynamic conductance $G(\omega)$ vs gate voltage V_g using our theory presented above. When $\omega = 1.2$ GHz (open circle), our results agrees very well[14] with the experimental data of Ref. 5. When $\omega = 3$ GHz (open square), our theory predicts that the imaginary part of $G(\omega)$, which is the electrochemical capacitance, goes to negative. For even larger frequency $\omega = 5$ GHz, the effect is more significant. To understand why one can observe a negative capacitance at small frequency such as 3 GHz, we note that since electron entering the QD has to first pass the QPC,[5] this QPC serves as a barrier (with an effective barrier height $1/\Gamma$) that is controlled by the gate voltage. At small gate voltage, T_{QPC} is nearly zero and goes to 1 at large V_g. Hence, the effective Γ for small gate voltage is quite large, making ω_c much smaller. Since the experiment of Ref. 5 is performed at $\omega = 1.2$ GHz, we assume that it is not too difficult to push the frequency to 3 GHz so that the effect of quantum inductance can be observed experimentally. Indeed, a single-wall carbon nanotube transistor operated at 2.6 GHz has been demonstrated[15] and measurement of current fluctuation at frequency from 5 to 90 GHz has been reported.[16]

In summary, we found that at finite frequency, a quantum capacitor consisting of a quantum dot and a large metal conductor is equivalent to a classical *RLC* circuit with three basic parameters: a static electrochemical capacitance C_μ, a charge relaxation resistance R_q, and a quantum inductance L_q. It is found that $L_q \sim R_q \tau$, where τ is the characteristic time scale for the quantum dot such as the dwell time τ_d or the tunneling time τ_t. Due to the phase delay by the quantum inductance, the dynamic current can lag behind the applied ac voltage, giving rise to a negative capacitance. Our numerical results show that this effect should be detectable experimentally using the present device technology.

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