Thermoelectric signature of the excitation spectrum of a quantum dot

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An electon-heating technique has been used to measure the diffusion thermopower of a quantum dot in the Coulomb blockade (CB) regime. When the gate voltage defining the dot is swept, the thermovoltage across the dot oscillates about zero with a period equal to that of the CB oscillations in electrical conductance and with a magnitude consistent with a Landauer formulation. In addition, a reproducible fine structure on top of the CB oscillations in the thermovoltage was observed that appears to result from the N-particle excitation spectrum of the dot. [S0163-1829(97)52016-9]

Semiconductor quantum dots may be formed by using surface gates to electrostatically confine electrons to an isolated island of charge within a two-dimensional electron-gas (2DEG) layer.[1] Provided that tunneling between the dot and surrounding 2DEG reservoirs is sufficiently weak, electrical transport through the system is determined at low temperatures T by a combination of confinement and charging effects. The N-particle excitation spectra of the electron system can affect transport, but even when T is sufficiently low so that $k_BT<\Delta E$, the N-particle energy-level spacing, Coulomb blockade (CB) (Ref. 2) forbids tunneling through the excited states within linear response (LR). Beyond LR, when the voltage bias V is such that $k_BT<\Delta E<|eV|$, the electrons *can* tunnel through the excited states and their signature can be seen in the differential conductance dI/dV through the dot.[3,4]

Beenakker and Staring[5] have pointed out that the thermopower S is a far more sensitive probe of the transport mechanisms in a quantum dot than the conductance. The thermopower is defined by $S=\Delta V_{\rm th}/\Delta T$, where $\Delta V_{\rm th}$ is the thermovoltage generated by a temperature different ΔT, applied across the dot. They show[5] that S should exhibit a fine structure resulting from N-particle states even within LR. This structure is in addition to that resulting from electron charging, which leads to oscillations in S about zero as a function of gate voltage V_g with a period equal to that of the CB oscillations in electrical conductance G. CB oscillations in S have thus far been reported by two groups[6,7] but the magnitudes of S deduced in the two cases were considerably different.

Here we report measurements on a quantum-dot sample that shows clear CB oscillations in both G and S, and for which ΔT, and thus the magnitude of S, can be determined with reasonable accuracy. The thermopower is found to be far smaller than that predicted in Ref. 5 and a fine structure in S is observed that we believe represents the first thermoelectric signature of the excitation spectrum of a quantum dot.

The device used is depicted in Fig. 1. The 2DEG was formed at a GaAs/$Al_xGa_{1-x}As$ heterojunction 70 nm below the sample surface. The carrier density n and mobility μ were measured at 4.2 K and found to be $n=2.2\times10^{15}$ m^{-2} and $\mu=80$ m^2 V^{-1} s^{-1}, respectively, indicating a mean-free path of $l=6$ μm. Surface Schottky gates were used to define two quasiballistic 2DEG channels of width 10 μm and length 60 μm on either side of a central quantum dot, itself defined using gates T, BL, P, and BR. Four quantum-point contacts (QPC's) at the boundaries of the two channels were used as voltage probes for four-terminal measurements of the longitudinal resistance R_{xx}, which was ≈170 Ω at zero magnetic field B. The lithographic dimensions of the dot were 0.4×0.5 μm^2. Measurement of the period of Aharonov-Bohm oscillations in G as a function of B in the quantum-Hall regime, following the method of Ref. 8, gave an area of

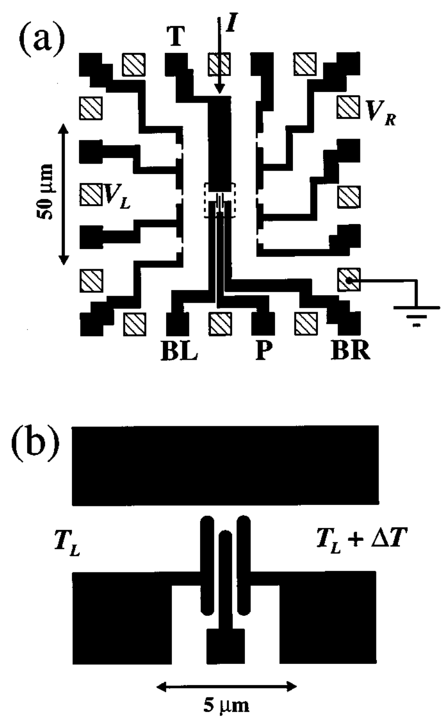

FIG. 1. (a) Schematic of the device. Dark regions represent Schottky gates, unshaded areas are regions of 2DEG, and the hatched squares represent Ohmic contacts to the 2DEG. (b) Detail of the dashed box in (a), showing the four gates used to define the quantum dot.

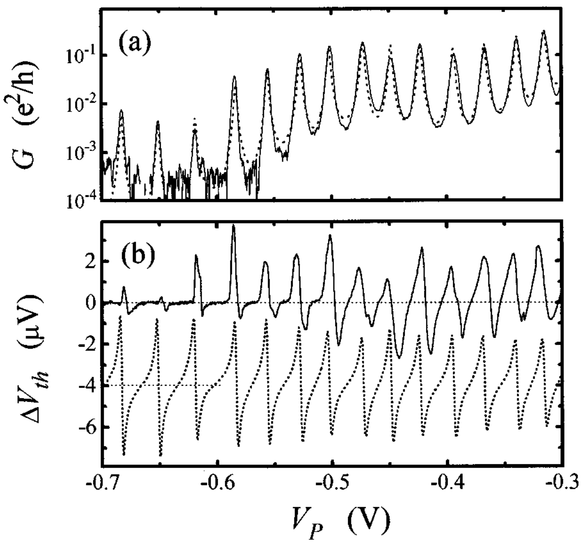

FIG. 2. (a) G (solid line) as a function of V_P at $T_L=50$ mK, within the other gates defining the dot at $V_T=-500$ mV, $V_{BL}=-830$ mV, and $V_{BR}=-900$ mV. The dashed line was calculated as described in the text with $\alpha=0.54$, $T=50$ mK, and $e^2/C=1.5$ meV. (b) Measured thermovoltage $\Delta V_{\rm th}$ (solid line) plotted against V_P with $I=30$ nA in the right-hand channel. The dotted line (offset) was calculated using $\alpha=0.54$, $\Delta T=100$ mK, $T_{\rm av}=100$ mK, and $e^2/C=1.5$ meV.

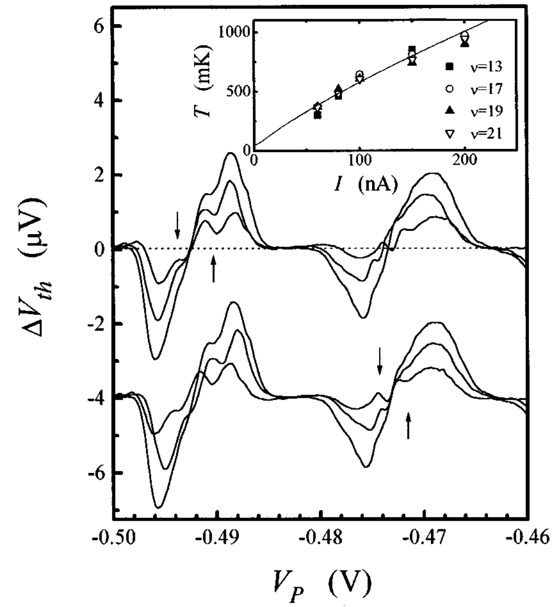

FIG. 3. $\Delta V_{\rm th}$ plotted against V_P with $I=30$, 40, and 50 nA, in order of increasing amplitude at $T_L=50$ mK. The data have been shifted in V_P to center on zero crossings in $\Delta V_{\rm th}$ at $V_P=-0.492$ V (top set of curves) and at $V_P=-0.473$ V (lower set). Arrows indicate structure associated with transport through excited states. Inset: Electron temperature T produced by a current I in the right-hand channel with $T_L=50$ mK, determined from a study of R_{xx} at various ν. The solid line is a fit to the data with $\gamma=2.5$ and $\beta=6.0\times10^{-18}$ WK$^{-2.5}$ (see text).

0.09 μm^2, indicating that the mean dot diameter was reduced to 0.3 μm by electrostatic depletion.

An electron-heating technique[9] was used to measure S, as employed in previous studies of mesoscopic thermopower.[10,11] When a current I was passed along one of the two quasiballistic channels, Joule heating in that channel created a differential ΔT in electron temperature across the dot [see Fig. 1(a)]. By measuring the resulting thermoelectric voltage $\Delta V_{\rm th}$ the thermopower could be determined from $S=\Delta V_{\rm th}/\Delta T$. Two QPC's opposing the dot, at the boundaries of the quasiballistic channels, were used as voltage contacts to determine the thermovoltage from $\Delta V_{\rm th}=(V_L-V_R)$. To increase sensitivity we employed an ac heating current I and phase-sensitive detection of $\Delta V_{\rm th}$, as used elsewhere.[7,12] Since the heating power varies as I^2, ΔT oscillates at *twice* the frequency $f=5$ Hz of the current I. It was thus necessary to phase lock to the $2f$ component of $\Delta V_{\rm th}$. The residual f component of the signal was below the noise threshold, as expected, since no current was passed through the dot. G was measured using a standard two-terminal configuration. It was necessary to switch between this circuit and the four-terminal thermovoltage circuit of Fig. 1(a), since earthing requirements precluded a simultaneous measurement.

In Fig. 2, G and $\Delta V_{\rm th}$ are plotted as a function of the voltage V_P on the central plunger gate P. The electrostatic barriers produced by gates BL and BR were biased into the tunneling regime, so that the individual conductances of the barriers were below e^2/h. In this regime we observed CB oscillations in G with gate voltage, which persisted almost all the way to $V_P=0$. The thermovoltage $\Delta V_{\rm th}$ exhibited oscillations about zero with the same period as those in G, as previously observed in similar quantum-dot systems.[6,7] As V_P was made more negative the tunnel barriers became more opaque with a resulting decrease in the amplitude of oscillations in G.

Figure 3 shows $\Delta V_{\rm th}$ as a function of V_P at three values of heating current I, for a different set of gate voltages to those in Fig. 2. The data show fine structure on a scale in V_P about an order of magnitude smaller than the CB oscillations. This structure appears to be due to the N-particle states of the dot, as predicted in Ref. 5, and was most pronounced when the dot was close to pinch-off, where $G<0.05e^2/h$. We have corrected the raw data in Fig. 3 for drifts of a few mV in gate voltage by centering on the zero crossings in $\Delta V_{\rm th}$, which correspond to CB peaks in G. The shifts in effective gate-voltage result from charge trapping and detrapping by impurities, commonly observed as random telegraph noise in GaAs/Al$_x$Ga$_{1-x}$As heterostructures.[14]

To independently study the N-particle states we performed nonlinear conductance measurements.[3,4] Figure 4 shows dI/dV plotted as a function of dc bias V across the dot at a variety of gate voltages. Both $dI/dV=0$ and $G=0$ in the regions around $V=0$ since the excess energy eV is not sufficient to break the CB. The peaks in dI/dV bordering this central region occur when the CB is breached and the widest separation $e\Delta V$ between peaks provides a direct measure of the charging energy e^2/C, where C is the capacitance between the dot and the surrounding charge. As the dot is taken closer to pinch-off it becomes increasingly isolated so that C decreases and e^2/C increases[15] from 1.3 meV at $V_P=-0.49$ V, to 2.2 meV at $V_P=-0.54$ V.

The secondary peaks in Fig. 4 represent tunneling through

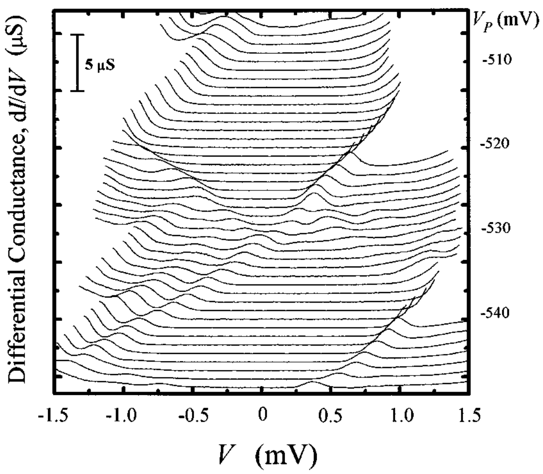

FIG. 4. dI/dV plotted against bias V, with $V_P = -504$ to -546 mV in 1-mV steps. Adjacent curves are offset by 0.75 μS for clarity. Here $V_T = -500$ mV, $V_{BL} = -921$ mV, $V_{BR} = -0.922$ mV, and $T_L = 50$ mK.

excited states in the dot that became accessible once the Coulomb gap has been breached.[3,4,15] We can directly measure the energy spacing ΔE from Fig. 4 and find typical values of around 250 μeV. Assuming that the fine structure in $\Delta V_{\rm th}$ of Fig. 3 results from the excited states we find an energy spacing of $\sim 0.1 e^2/C$, consistent with the data in Fig. 4. We note that the fine structure in $\Delta V_{\rm th}$ suffers thermal broadening at $I = 50$ nA, corresponding to $k_B \Delta T \sim 50$ μeV (see below), again consistent with it being due to excited states.

The two previous studies of S for a quantum dot[6,7] used distinct models to fit their data. Staring *et al.*[7] used the model of Ref. 5, which predicts an amplitude $S_{p\text{-}p} = e/2CT$ that is generally much larger than that given by the Mott expression,[16] $S \sim -\ln[dG(E)/dE]$, as used in Ref. 6. To resolve this issue it was necessary to determine ΔT as accurately as possible so that S could be calculated from the $\Delta V_{\rm th}$ data in Fig. 2.

Previously[6,13] we have used the thermopower of a QPC near the heating channel as a probe of ΔT, allowing a comparison of the experimental $\Delta V_{\rm th}$ with theory. Here we used an independent method to determine $\Delta T(I)$. The amplitude of Shubnikov–de Haas (SdH) oscillations in R_{xx} along the heating channel was measured as a function of both T_L and the applied I. The damping of the SdH oscillations with increasing T_L agreed with standard theory[17] at a variety of filling factors ν for $50 < T_L < 800$ mK. The current-dependent $R_{xx}(I)$ amplitudes were then converted into electron temperatures T using this dependence. The inferred values of $T(I)$ are plotted in Fig. 3 (inset).

The energy-loss rate P from a 2DEG generally obeys $P = I^2 R = \beta(T^\gamma - T_L^\gamma)$. In a macroscopic GaAs/Al$_x$Ga$_{1-x}$As heterojunction below 1 K, energy loss is dominated by the emission of acoustic phonons via the piezoelectric interaction, leading to a $\gamma = 5$ dependence.[18] For $T \sim 500$ mK, the heated electrons relax to T_L over a distance $l_{e\text{-ph}} \sim 200$ μm, and as T is lowered further $l_{e\text{-ph}}$ can significantly exceed the sample size.[19] In this regime phonon emission is not efficient at cooling electrons in the channel region and heat is conducted out of the channel via the electrons before being dissipated as phonons in the contact regions.[19,20] Electron-electron scattering within the heated channel serves to redistribute energy, leading to a well-defined electron temperature profile. Using the Wiedemann-Franz law to calculate this T profile[21] one finds $\gamma = 2$. A best fit to the data in Fig. 3 (inset) gives $\gamma = 2.5$, indicating a crossover between electron out-diffusion and dissipation through direct phonon emission. For $T > 800$ mK our $T(I)$ values are within 20% of those calculated by Ma *et al.*,[18] who found $\gamma = 5$ and $\beta = 3.2 \times 10^{-18}$ WK^{-5} by assuming that P was determined solely by phonon emission.

For the data in Fig. 2(b) we have $\Delta T = 170$ mK from the $T(I)$ calibration. The model of Ref. 5 then predicts $\Delta V_{\rm th}^{p\text{-}p} = (e/2CT)\Delta T \approx 1$ mV. This is two orders of magnitude larger than our measured $\Delta V_{\rm th}$. Staring *et al.*[7] found agreement with Ref. 5 only by assuming $\Delta T \sim 1$ mK for values of I and T_L close to those used here. They did not have an independent measure of ΔT, but such a small rise in T is inconsistent with current theories of electron energy loss.[18–21]

The theory of Ref. 5 assumes the ''orthodox model' of CB (Ref. 22) in which $G \ll e^2/h$, so that virtual-tunneling processes (or *cotunneling*) are neglected. Away from maxima in G, transport through the dot can only occur by thermal activation. In our quantum-dot system we have $10^{-3} e^2/h < G < 10^{-1} e^2/h$ for most gate voltages, but judging from the small magnitude of the measured S the transport is not fully activated.

To model our data we use a simple single-particle framework. Johnson *et al.*[3] showed that a single-particle Landauer formulation can model many aspects of CB even though it is a many-body charging effect. This is true because transport of the Nth electron through the dot is an energy-conserving process, where phase coherence is maintained even though the other $N-1$ electrons experience a Coulomb energy change. This approach has recently been shown valid for a general interacting mesoscopic system.[23] The transmission probability t through the dot is given by[3]

$$t = \frac{\Gamma_L^2 \Gamma_R^2}{1 + (1-\Gamma_L^2)(1-\Gamma_R^2) - 2[(1-\Gamma_L^2)(1-\Gamma_R^2)]^{1/2} \cos\phi}, \tag{1}$$

where $\Gamma_L = [\alpha + \Gamma[1-\alpha)]$ and $\Gamma_R = \Gamma^2/[\alpha + \Gamma(1-\alpha)]$ are the probability amplitudes for transmission through the left and right barriers and α is an adjustable parameter between zero and unity which determines asymmetry between the barriers. To obtain Γ as a function of V_P we averaged G (in units of e^2/h) over V_P to remove the oscillatory component, which is equivalent to setting $e^2/C = 0$. The phase ϕ is that acquired by an electron in one round trip between the barriers. To fit the data $\phi(V_P)$ was increased by 2π each time V_P passed through a peak in G, and intermediate values were obtained by linear interpolation. We do not suggest a physical mechanism through which ϕ varies with V_P, but note that the model provides a convenient form for the energy-dependent single-particle transmission $t(E)$ through the dot. We can then calculate both G and S using the standard Landauer formulas for thermoelectric transport.[24]

The predictions of this simple model are compared with experiment in Fig. 2. We have assumed $e^2/C = 1.5$ meV, which is inferred from Fig. 4 as the mean value of e^2/C over the range of the data in Fig. 2. A best fit to G was obtained using $\alpha=0.54$ and the resulting values are compared with experiment in Fig. 2(a). The thermovoltage data in Fig. 2(b) show qualitative agreement with theory for $V_P > -0.55$ V. At more negative V_P that data appear quenched. In particular, at V_P values corresponding to G minima, $\Delta V_{\rm th}$ falls rapidly to zero. This is also observed in the data of Fig. 3. Here the resistance R of the dot exceeds 200 MΩ and any leakage path of lower resistance will serve to short out the thermovoltage. We also calculated $\Delta V_{\rm th}$ using the Mott expression[16] and the experimental G data. This gave similar results to that obtained by calculating $t(E)$ from Eq. (1), but the G data were too noisy to obtain a satisfactory derivative with respect to V_P.

Typical values of I required to obtain a reliable $\Delta V_{\rm th}$ were $I \geqslant 10$ nA, giving $\Delta T \geqslant 100$ mK [see Fig. 3 (inset)]. Consequently it will be very difficult to obtain thermopower measurements in the LR regime ($\Delta T \ll T_L$) for T_L below a few hundred mK. However in calculations of nonlinear thermopower for QPC's (Ref. 13) and quantum dots[25] it has been shown that the thermopower can be well approximated by the LR value calculated at the *average* T of the two reservoirs, provided $k_B \Delta T$ is well below a characteristic energy of the system, being e^2/C for a dot.[25] For the data in Fig. 2(b) we have $\Delta T = 170$ mK from the $T(I)$ calibration, giving $e^2/C \approx 100 k_B \Delta T$, so it is valid to use the average T to calculate S. This gives an amplitude for $\Delta V_{\rm th}$ that is too large by a factor of 2.5. We find a best fit to $\Delta V_{\rm th}$ [dotted line in Fig. 2(b)] by setting $\Delta T = 100$ mK, giving an average $T = 100$ mK. Uncertainty in the values obtained from the $T(I)$ calibration and in our estimate of e^2/C are sufficient to account for this discrepancy.

In summary, we have used a current-heating technique to study CB oscillations in the thermopower of a quantum dot. The amplitude of SdH oscillations in R_{xx} for the 2DEG in the heated reservoir acted as an electron thermometer to determine the applied temperature differential ΔT. The magnitude of S was two orders of magnitude smaller than that predicted for a dot with negligible cotunneling. We found reasonable agreement to the data using a single-particle Landauer formulation indicating that even a small amount of cotunneling can have a significant effect on the form of S. Additional fine structure in the thermopower oscillations appears to be due to the effect of the N-particle excited states in the dot. This interpretation was supported by nonlinear electrical conductance measurements, which indicated an excitation spectrum with similar energy spacings to that determined from the thermopower.

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