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INVITED PAPER

Sawtooth-like thermopower oscillations of a quantum dot in the Coulomb blockade regime

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Abstract. Current heating is used to measure the thermopower of a quantum dot in the Coulomb blockade regime. We observe sawtooth-like oscillations as a function of gate voltage in the thermovoltage across the dot. These observations are compared with measured Coulomb blockade oscillations in the conductance, and with theory.

In the past few years our understanding of transport phenomena in semiconductor nanostructures has increased considerably. Most research has focused on purely electrical properties, and we refer the reader to [1] (quantum ballistic and quantum diffusive transport) and [2] (transport in the Coulomb blockade regime) for an overview of the transport properties encountered in these devices. In addition, experiments have started on thermal and thermoelectric quantum transport phenomena in semiconductor nanostructures, as documented in some recent reviews [3, 4]. In this paper we present some of our recent results [5, 6] on electric and thermoelectric transport phenomena of a quantum dot in the Coulomb blockade, or single-electron tunnelling, regime.

Single-electron tunnelling is the dominant mechanism governing the transport properties of a quantum dot that is weakly coupled to reservoirs by tunnel barriers. At temperatures T such that $k_BT \ll e^2/C$, with C the capacitance of the dot, it leads to novel transport phenomena, such as the 'Coulomb blockade oscillations' in the conductance in the linear, and the 'Coulomb staircase' in the nonlinear transport regime. In thermoelectric transport, the Coulomb blockade should lead to [7] sawtooth-like oscillations in the thermopower S ($S \equiv \Delta V_{thermo}/\Delta T$, where V_{thermo} is the thermovoltage induced by a temperature difference ΔT across the dot) as a function of the Fermi energy in the reservoirs. Examples of all of these phenomena follow below.

The samples used for the experiments are defined electrostatically in the two-dimensional electron gas (2DEG) of (Al,Ga)As heterostructures. We use electron-beam lithography to fabricate Ti–Au gates of dimensions down to 0.1 μm. The (Al, Ga)As wafer used here has an electron density $n_s \approx 3.7 \times 10^{11}$ cm^{-2} and a mobility $\mu \approx 10^6$ cm^2 V^{-1} s^{-1}. The layout of the patterned Ti–Au gates is shown in figure 1. Gates A, D and F define two adjustable tunnel barriers, and two additional gates, B and C, define a narrow channel. A point contact in the boundary of this channel, opposite to the dot, is used as a reference voltage probe in the thermopower measure-

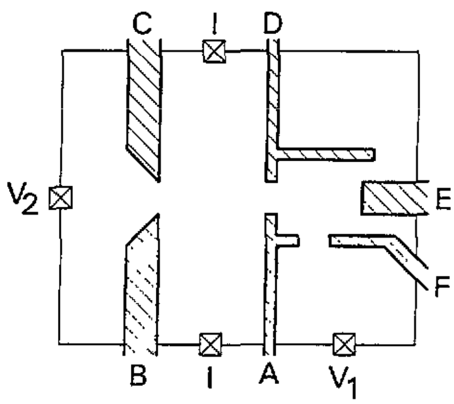

Figure 1. Schematic top view of the 0.7 × 0.8 μm^2 quantum dot adjacent to a 2 μm wide, 20 μm long channel. Gates A, D and F (hatched) define individually adjustable tunnel barriers, and gate E controls the electrostatic potential of the dot; the gaps between gates D and E, and between gates E and F, are pinched off in the experiment. For the thermovoltage experiment, an AC heating current I is passed through the channel and the thermovoltage $V_{th} \equiv V_1 - V_2$ is measured across the dot and the opposite reference point contact defined by gates B and C.

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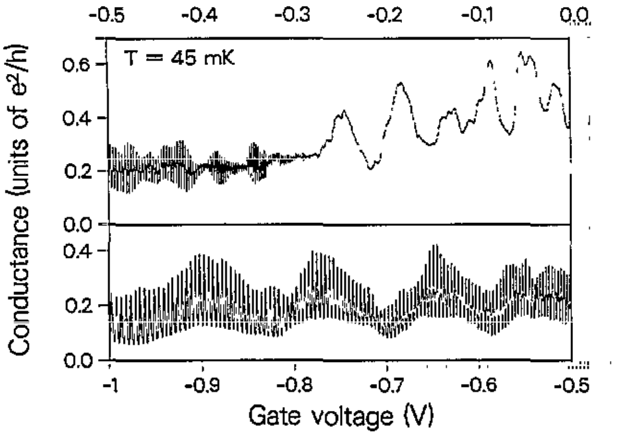

Figure 2. Electrical conductance of the quantum dot in the linear regime, as a function of the voltage applied to gate E. The excitation voltage was 9 μV.

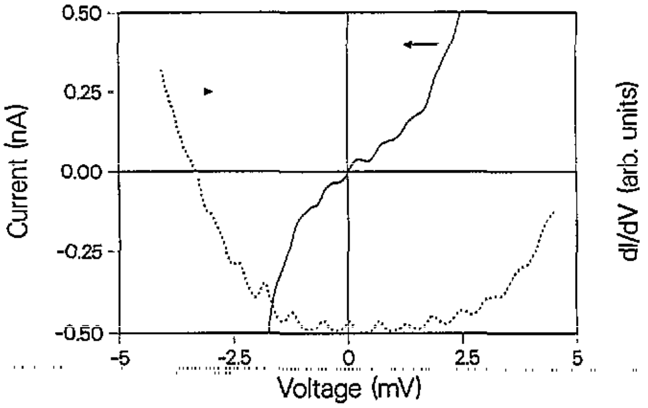

Figure 3. Coulomb staircase obtained at 45 mK for a strongly asymmetrically adjusted quantum dot. The full curve gives the measured current, the dotted curve the differential conductance, as a function of the applied bias voltage.

ment (see below). The sample is immersed in liquid helium in the mixing chamber of a dilution refrigerator at a temperature of 45 mK and at zero magnetic field. The signals are measured using low-frequency lock-in techniques.

Perhaps the most striking manifestation of the Coulomb blockade on the transport properties of a quantum dot is the occurrence of a periodic series of peaks in the conductance of the dot ('Coulomb blockade oscillations') as a function of the electrochemical potential of the dot, which in our case can be varied by changing the voltage on gate E. The peaks in the conductance occur for those gate voltages where the free energy of a dot containing N electrons equals that of a dot containing $N-1$ electrons. As an example, we show in figure 2 the Coulomb blockade oscillations observed for the dot used in the thermopower experiments. One observes a long series of peaks in the conductance, with consecutive peaks (for less negative gate voltages) corresponding to the addition of a single electron to the dot. At gate voltages > -0.3 V, the electron gas underneath gate E is not fully depleted, and we tentatively attribute the irregular structure in the conductance trace to Fabry–Pérot-type transmission resonances [8]. In the scan shown here, the entrance and exit tunnel barriers are both adjusted to a conductance of about $0.5e^2/h$; such relatively low barriers may enable (higher-order) co-tunnelling processes [9]. This explains, at least partly, the presence of a remanent conductance in the Coulomb blockade minima [5].

In the nonlinear regime, the Coulomb blockade can be suppressed by applying a bias voltage V_b across the dot so that $eV_b > e^2/C$. For a dot with asymmetrically adjusted tunnel barriers at entrance and exit this leads to a second transport peculiarity, namely the Coulomb staircase. An example of this type of I–V curve is given in figure 3, which was obtained for entrance and exit tunnel barrier conductance of $0.15e^2/h$ and $0.02e^2/h$, respectively. At each step in this curve, an extra electron is added to the dot.

From the theory of Coulomb blockade [10] it follows that the electrochemical potential of the dot in the single-electron tunnelling regime varies in a sawtooth fashion with the voltage on gate E. It has proved difficult to directly observe this sawtooth behaviour (however, see [11] for a recent experiment). In a recent paper, Beenakker and Staring [7] showed theoretically that one expects this sawtooth behaviour to be directly observable in the thermopower of the quantum dot. Therefore we designed an experiment to measure the thermopower of a quantum dot. It is at this point that the narrow channel, defined by gates B and D in our structure, becomes relevant: we use it to create a hot-electron reservoir. This is possible by virtue of the fact that in the 2DEG in an (Al, Ga)As heterojunction structure at low temperatures, the coupling between hot electrons and the lattice is much smaller (typical relaxation time $\lesssim 1$ ns) than the coupling within the electron system ($\sim$ps). Thus, by passing a current through a suitably dimensioned channel a reservoir of hot electrons is created. Using this technique, we have previously been able to observe the quantum size effects in the thermopower [12], Peltier coefficient and thermal conductance [13] of a quantum point contact.

In order to observe the thermopower of our quantum dot [6], we use the sample of figure 1, with the tunnel barriers defined by gates B, C, A and D, adjusted to conductances of about $0.1e^2/h$ each, and current heating provided by a small AC current passing through the channel defined by gates B, C, A and D. The current heating leads to a small difference in electron temperature ($\Delta T \propto I^2$) across the dot and across the opposite reference point contact (defined by gates B and C). Lock-in detection at twice the AC frequency is then used to measure a transverse thermovoltage $V_{th} = V_1 - V_2$, which equals the difference in thermovoltages across the dot and the reference point contact, as

$$V_{th} = (S_{dot} - S_{ref})\Delta T. \tag{1}$$

Here S_{dot} is the thermopower of the dot and S_{ref} is the thermopower of the reference point contact. The contribution of S_{ref} to V_{th} is independent of V_E and leads to a

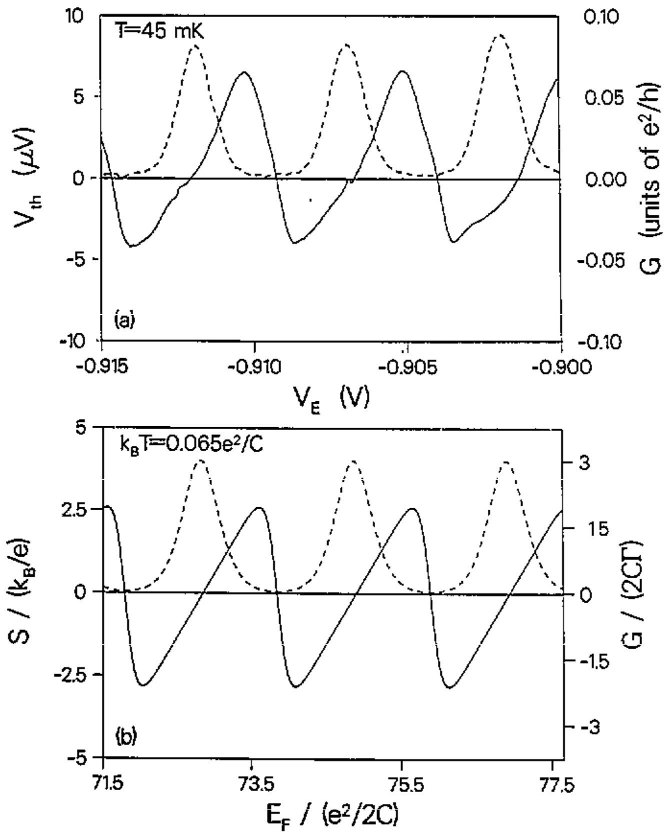

Figure 4. (*a*) Thermovoltage V_{th} at a heating current of 58 nA (full curve) and conductance (broken curve) as a function of gate voltage V_E at a lattice temperature of $T = 45$ mK. (*b*) Calculated thermopower (full curve) and conductance (broken curve) of a quantum dot as a function of Fermi energy using the theory of [7]. The parameters used in the calculations are discussed in the text.

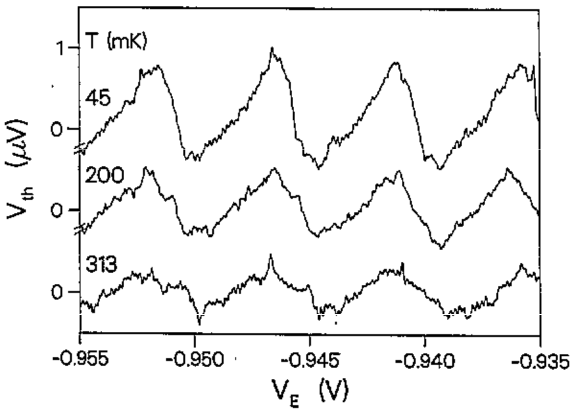

Figure 5. Thermovoltage V_{th} as a function of V_E at lattice temperatures of $T = 45$, 200 and 313 mK, obtained using a heating current of 18 nA.

constant offset voltage, which is minimized in our experiment by suitably adjusting the reference point contact [12]. Thus, variations in V_{th} as a function of V_E directly reflect changes in the thermopower of the dot.

In figure 4 we compare measurements of the Coulomb blockade oscillations as a function of V_E in the thermovoltage (full curve) and conductance (broken curve, obtained from a separate measurement) of the dot, at a lattice temperature of $T = 45$ mK. The heating current used in the thermovoltage experiment was 58 nA. Clearly, the thermovoltage $V_2 - V_1$ (and therefore the thermopower of the dot) oscillates periodically. The period is equal to that of the conductance oscillations, and thus corresponds to depopulation of the dot by a single electron. As expected, the thermovoltage oscillations have a distinct *sawtooth* lineshape. In addition, the conductance peaks are approximately centred on the positive slope of the thermovoltage oscillations, with the steeper negative slope occurring in between two conductance peaks. These data comprise a clear experimental demonstration of the key characteristics of the thermopower oscillations of a quantum dot.

The theoretical curve of the thermopower in figure 4(*b*) was calculated using the linear response formalism of [10]. In order to obtain the excellent agreement in both conductance and thermopower behaviour, we had to perform the calculations for an electron temperature in the dot of 230 mK, i.e. higher than the actual lattice temperature in the experiment. We encountered this problem in several different experiments on transport in quantum dots [5], and tentatively attribute the effect to a finite amount of lifetime broadening of the energy levels in the dot. However, we cannot rule out a certain amount of electron heating due to RF pick-up.

In figure 5 we show the behaviour of the thermovoltage oscillations for three different lattice temperatures T_{latt} ($T_{latt} = 45$, 200 and 315 mK, respectively), obtained for a heating current of 16 nA. One observes that sawtooth lineshape becomes more symmetric for higher lattice temperatures, owing to thermal smearing. For a quantitative comparison of the magnitude of the observed thermovoltage with the theoretical peak-to-peak value $\Delta V_{th} \approx (e/2CT_{latt})\Delta T$, one needs to know the self-capacitance C of the dot, and the increase in electron temperature ΔT in our experiment. From the temperature dependence of the conductance oscillations (not shown here) we find that $e^2/C \approx 0.3$ meV. From the 200 mK trace, we infer that this value for the self-capacitance implies that $\Delta T \approx 1$ mK for $I = 18$ nA. A convenient manner of independently determining ΔT is by making use of the quantized thermopower of the reference point contact BC (cf [12, 13]). However, this technique proved not to be viable for the present experiment, as the conductance of point contact BC does not exhibit well defined plateaus (probably due to quantum interference effects) at temperatures below about 1 K. Still another manner for estimating ΔT is by using the crude heat balance introduced in [12] for the current heating process. We have $c_v \Delta T = (I/W)^2 \rho \tau_{loss}$, with $c_v = (\pi^2/3)(k_B T/E_F) n k_B$ the heat capacity per unit area of the 2DEG, W the channel width, ρ the channel resistivity and τ_{loss} an energy relaxation time [12]. Substituting $\Delta T = 1$ mK, we obtain $\tau_{loss} = 2 \times 10^{-10}$ s. This is a reasonable number, and consistent with our earlier experiments on the thermopower of a quantum point contact defined in similar 2DEG material [12].

In conclusion, we have presented data on thermal and thermoelectric transport properties of a quantum dot in the Coulomb blockade regime. In contrast with the periodic peak structure ('Coulomb-blockade oscillations') of the conductance with varying gate voltage, the thermopower of the dot oscillates in a sawtooth manner. For both effects, the period corresponds to a period of one oscillation per electron added to the dot.

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References

[1] Beenakker C W J and van Houten H 1991 *Solid State Phys.* **44** 1
[2] van Houten H, Beenakker C W J and Staring A A M 1992 *Single Charge Tunnelling* (NATO ASI Series B vol 294) ed H Grabert and M H Devoret (New York: Plenum) pp 167–216
[3] Molenkamp L W, van Houten H and Beenakker C W J 1993 *Proc. CXVII-th Course Int. School of Physics 'Enrico Fermi': Semiconductor Superlattices and Interfaces* ed A Stella and L Miglio (Amsterdam: North-Holland) pp 365–78
[4] Gallagher B L and Butcher P N 1992 *Handbook on Semiconductors* ed T Moss vol 1 ed P T Landsberg (Amsterdam: Elsevier) p 721
[5] Staring A A M 1993 *PhD Thesis* Eindhoven University of Technology
[6] Staring A A M, Molenkamp L W, Alphenaar B W, van Houten H, Buyk O J A, Mabesoone M A A, Beenakker C W J and Foxon C T 1993 *Europhys. Lett.* **22** 57
[7] Beenakker C W J and Staring A A M 1992 *Phys. Rev.* B **46** 9667
[8] Smith C G, Pepper M, Frost J E F, Hasko D G, Newbury R, Peacock D C, Ritchie D A and Jones G A C 1991 *J. Phys.: Condens. Matter* **1** 9035
[9] Averin D V and Nazarov Yu V 1992 *Single Charge Tunnelling* (NATO ASI Series B vol 294) ed H Grabert and M H Devoret (New York: Plenum) pp 217–47
[10] Beenakker C W J 1991 *Phys. Rev.* B **44** 1646
[11] Field M, Smith C G, Pepper M, Ritchie D A, Frost J E F, Jones G A C and Hasko D G 1993 *Phys. Rev. Lett.* **70** 1311
[12] Molenkamp L W, van Houten H, Beenakker C W J, Eppenga R and Foxon C T 1990 *Phys. Rev. Lett.* **65** 1052
[13] Molenkamp L W, Gravier Th, van Houten H, Buyk O J A, Mabesoone M A A and Foxon C T 1992 *Phys. Rev. Lett.* **68** 3765