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Coulomb-Blockade Oscillations in the Thermopower of a Quantum Dot.

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Abstract. – The thermopower of a quantum dot, defined in the two-dimensional electron gas in a GaAs-$Al_xGa_{1-x}As$ heterostructure, is investigated using a current heating technique. At lattice temperatures $k_B T$ much smaller than the charging energy e^2/C, and at small heating currents, sawtoohlike thermopower oscillations are observed as a function of gate voltage, in agreement with a recent theory. In addition, a remarkable sign reversal of the amplitude of the thermopower oscillations is found in the non-linear regime at large heating currents.

Single-electron tunnelling [1] 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 $k_B T \ll e^2/C$, with C the capacitance of the dot, it leads the Coulomb-blockade oscillations in the conductance as a function of the voltage applied to a capacitively coupled gate electrode [2]. Whereas the conductance has been studied extensively, the thermo-electric properties of a quantum dot remain essentially unexplored. Amman *et al.* have studied theoretically the role of Coulomb interactions on thermo-electric effects in a single mesoscopic tunnel junction, and have used their results to interpret the thermopower of granular thin bismuth films [3]. Recently, a theory was developed for the thermopower of a quantum dot in the Coulomb-blockade regime [4]. This theory predicts sawtoothlike oscillations in the thermopower as a function of the Fermi energy in the reservoirs, with an amplitude that is determined by the charging energy and temperature only.

Here, we present an experimental study of the thermo-electric properties of a quantum dot, using the current-heating technique applied previously to study the thermovoltage across a quantum point-contact [5]. At low lattice temperatures and small heating currents,

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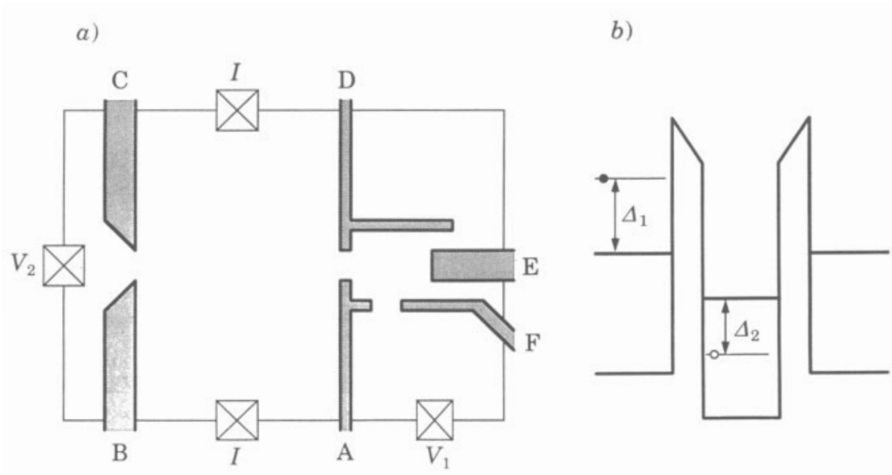

Fig. 1. – *a*) Schematic top view of the $(0.7 \cdot 0.8)\,\mu m^2$ quantum dot adjacent to a $2\,\mu m$ wide, $20\,\mu m$ long channel. Gates A, D and F (hatched) define individually adjustable tunnel barriers, and gate E controls the electrostatic potential of the dot; the gaps between gates D and E, and between gates E and F, are pinched off in the experiment. An a.c. 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. *b*) Schematic band diagram of the quantum dot. Due to the Coulomb blockade, transport through the dot is posible if an electron with excess energy Δ_1 is created in the reservoir, and a hole with excess energy Δ_2 is created in the dot, such that $\Delta_1 + \Delta_2 = \Delta$.

we observe sawtoothlike 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 are analysed in terms of the theory of ref. [4]. We find that the lineshapes of the Coulomb-blockade oscillations in both the thermopower and the conductance agree with the theory. In addition, we report a remarkable, counterintuitive sign reversal of the thermovoltage oscillations in the non-linear regime at large heating currents.

The quantum dots used for the experiments are defined electrostatically in the two-dimensional electron gas (2DEG) in a GaAs-$Al_xGa_{1-x}As$ heterostructure (electron density $n_s = 3.7 \cdot 10^{11}\,cm^{-2}$ and mobility $\mu \approx 10^6\,cm^2/Vs$). The layout of the patterned Ti-Au gates is shown in fig. 1*a*). Gates A, D and F define two tunnel barriers with conductances of about $0.1e^2/h$ each, and two additional gates, B and C, define a narrow channel. A point-contact in the boundary of this channel is used as a reference voltage probe, opposite to the dot. The sample is immersed in the liquid helium in the mixing chamber of a dilution refrigerator, and measurements of the thermopower and conductance of the dot are made as a function of the voltage V_E applied to gate E. A thermovoltage across the dot is generated by heating the electron gas in the channel using a small low-frequency (13 Hz) a.c. current I. This leads to an electron temperature difference $\Delta T \propto I^2$ across the dot at low currents [5]. Lock-in detection at twice the frequency of the heating current is used to measure the thermovoltage $V_{th} \equiv V_1 - V_2$, which is equal to the difference in thermovoltages across the dot and reference point-contact,

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

where 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 constant offset voltage, which is minimized in our experiment by suitably adjusting the reference

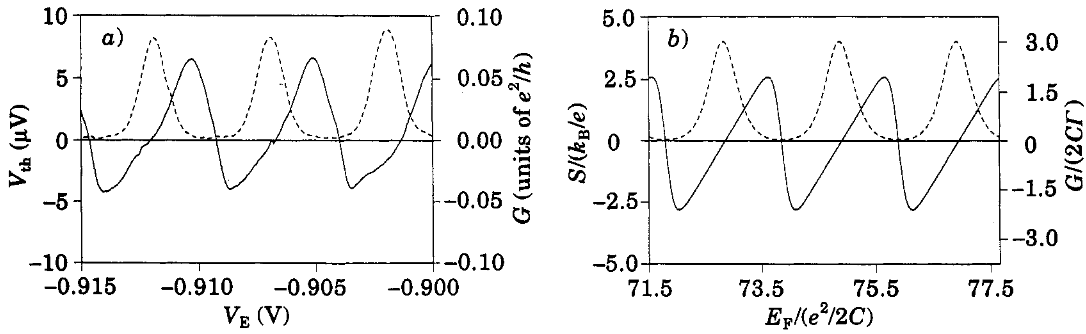

Fig. 2. – *a*) Thermovoltage V_{th} at a heating current of 58 nA (solid line) and conductance (dashed line) as a function of V_E at a lattice temperature of $T = 45$ mK. *b*) Calculated thermopower (solid line) and conductance (dashed line) of a quantum dot as a function of Fermi energy using the theory of ref. [4]. The parameters used in the calculations are discussed in the text; $k_B T = 0.065 e^2/C$.

point-contact [5]. Thus, variations in V_{th} as a function of V_E directly reflect changes in the thermopower of the dot. Separately, we have measured the two-terminal conductance of the dot using a standard lock-in technique with an excitation voltage across the dot below 9 μV. (No heating current is passed through the channel for this measurement.)

In fig. 2*a*) we compare measurements of the Coulomb-blockade oscillations as a function of V_E in the thermovoltage (solid) and conductance (dashed) of the dot, at a lattice temperature of $T = 45$ mK. Clearly, the thermovoltage V_{th} (and therefore S_{dot}) oscillates periodically. The period is equal to that of the conductance oscillations, and corresponds to depopulation of the dot by a single electron. In contrast to the conductance oscillations, which consist of a series of *symmetric* peaks separated by gate voltage regions where the conductance is suppressed, 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 between two conductance peaks. No shift in gate voltage is found when the thermovoltage and thermopower are measured alternatingly. These data comprise a clear experimental demonstration of the key characteristics of the thermopower oscillations of a quantum dot.

A detailed theory of the effect has been published elsewhere [4]. However, a *qualitative* understanding of the sawtooth lineshape of the thermopower oscillations can be obtained by considering the heat flow I_Q associated with a particle current J through the dot. The two are related through the Peltier coefficient $\Pi \equiv (\partial I_Q / e\, \partial J)_{\Delta T = 0}$, which may be interpreted as the energy that is transferred between the reservoirs per transferred carrier. In linear response, the Peltier coefficient is related to the thermopower through the Kelvin-Onsager relation (¹),

$$S = \frac{\Pi}{T} = \frac{1}{eT} \frac{\partial I_Q}{\partial J} \bigg|_{\Delta T = 0} . \tag{2}$$

We now estimate the amount of energy carried by a single electron. Due to the Coulomb

(¹) However, see ref. [3] for a discussion of the validity of the Kelvin-Onsager relation in the Coulomb-blockade regime.

blockade, an electron can be transferred through the dot at low temperatures only if an excitation energy of at least Δ is supplied to the system, where

$$\Delta = E_N + \left(N + \frac{1}{2}\right)\frac{e^2}{C} - e\phi_{\text{ext}} - E_F\,. \tag{3}$$

Here, N is the number of electrons in the dot, E_N is the energy of the N-th single-particle level in the dot, ϕ_{ext} is that part of the electrostatic-potential difference between dot and reservoirs that is due to external charges (in particular those on a nearby gate electrode), and E_F is the Fermi energy in the reservoirs. Minimization of $|\Delta|$ with respect to N results in a sawtooth dependence of Δ on the external gate voltage [2, 4]. The energy Δ may be used in part to excite an electron in the reservoir to an energy Δ_1 above the Fermi level (with probability $\exp[-\Delta_1/k_B T]$), and in part to create an empty state in the dot at an energy Δ_2 below the Fermi level (with probability $\exp[-\Delta_2/k_B T]$), see fig. 1*b*). The excited electron can then tunnel through the dot to the other reservoir via the empty state, so that the energy Δ_1 is transferred. This process occurs with probability $\exp[-\Delta/k_B T]$, independent of Δ_1. Therefore, the energy that is transferred per electron is on average $(1/2)\Delta$, so that $\Pi = \Delta/2e$, or $S = \Delta/2eT$. This explains the sawtooth lineshape of the thermopower oscillations.

Using the linear-response formulae of ref. [4], the thermopower and conductance can be calculated as a function of the Fermi energy. As shown in fig. 2*b*), the calculated lineshapes of the Coulomb-blockade oscillations in both the thermovoltage and the conductance reproduce the experimental lineshapes in fig. 2*a*) quite well. In the calculations, we assume a single-particle energy spectrum in the dot consisting of equidistantly spaced, twofold degenerate levels, with spacing $\Delta E = 0.05e^2/C$, and energy-independent tunnel rates Γ. The best match between the calculated and experimental lineshapes is obtained for a reduced temperature $k_B T/(e^2/C) \approx 0.065$. Experimentally, we find that the conductance oscillations vanish for $T \gtrsim 1.5$ K, so that we estimate $e^2/C \approx 0.3$ meV. This implies that the reduced temperature used in the calculations corresponds to about 0.23 K, which is five times as high as the actual lattice temperature in the experiment (45 mK). If we use a lower reduced temperature, the calculated conductance peaks are too narrow and the calculated thermopower oscillations are too skewed, compared to the experimental results. This anomalous broadening does not result from our current heating technique, since it is observed in *both* S_{dot} *and* G. Although we cannot completely exclude electron-gas heating due

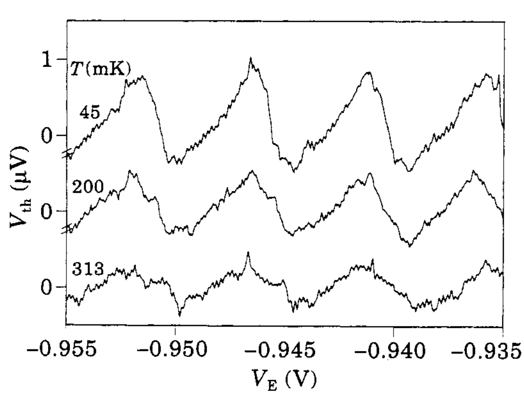

Fig. 3. – Thermovoltage V_{th} as a function of V_E at lattice temperatures of $T = 45$, 200 and 313 mK, obtained using a heating current of 18 nA.

to residual electrical noise, we suggest that the broadening may be intrinsic, and results from a finite width of the levels in the dot ($h\Gamma \gtrsim k_B T$). This is not taken into account in our calculations.

The lattice-temperature dependence of the thermovoltage oscillations is shown in fig. 3, where traces of V_{th} *vs.* V_E are given for three different temperatures (obtained using a heating current of $I = 18$ nA). The sawtooth lineshape of the thermopower oscillations is most pronounced at the lowest lattice temperature of $T = 45$ mK, and is gradually replaced by a more symmetric lineshape as T is increased to 313 mK. If the temperature is further increased to 750 mK, the oscillations are no longer observable using $I = 18$ nA, due to the large noise level. If larger heating currents are used, however, the oscillations can still be observed up to $T \gtrsim 1.5$ K (not shown).

A comparison of the temperature dependence of the thermovoltage oscillations with theory [4] requires knowledge of the electron temperature difference ΔT generated by the heating current in the channel. Previously [6], we have shown how ΔT can be inferred from the thermovoltage across a quantum point-contact in the boundary of the channel. However, this proves to be difficult in the present devices since the conductance of the reference point-contact does not exhibit well-defined quantized plateaus, probably due to quantum interference effects at temperatures below 1 K. Instead, we estimate ΔT using the theoretical result for the peak-to-peak amplitude of the thermopower oscillations, $\Delta V_{th} \approx \approx (e/2CT)\,\Delta T$ [4]. Inserting the measured ΔV_{th} (from the 200 mK trace in fig. 3) and using the above estimate for the charging energy, we obtain $\Delta T \approx 1.0$ mK. An independent expression for ΔT is the simple heat balance equation $c_v \Delta T = (I/W)^2 \rho \tau_\varepsilon$, with $c_v = (\pi^3/3)(k_B T/E_F)\, n_s k_B$ the heat capacity per unit area of the 2DEG, W the channel width, ρ the channel resistivity, and τ_ε an energy relaxation time associated with energy transfer from the electron gas to the lattice [5]. Substituting $\Delta T = 1$ mK, we obtain $\tau_\varepsilon = 2 \cdot 10^{-10}$ 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 [5].

We now turn to a discussion of an intriguing result found in experiments beyond the regime of linear response (²). In the inset of fig. 4 we show nine traces of thermovoltage oscillations using a second device, obtained for heating currents increasing stepwise from 0.1 to 0.9 μA. We find that the oscillations do not simply fade out, as one might expect from thermal smearing, but in addition exhibit a *sign reversal* of the oscillation amplitude (³). To study this effect in more detail, we have measured the dependence of the thermovoltage on the heating current I for two fixed gate voltages, one corresponding to a maximum in the (low heating current) thermovoltage oscillations, and the other corresponding to an adjacent minimum. Subtraction of these two data sets yields the trace of the peak-to-peak amplitude ΔV_{th} of the thermovoltage oscillations *vs.* I given in fig. 4, which clearly shows the sign reversal of ΔV_{th} at approximately 0.65 μA. Using the heat balance argument discussed above, we estimate that the electron gas temperature in the channel is as high as a few kelvin at this current level, so that $\Delta T \gg T$, $\Delta E/k_B$. A theoretical study of the thermovoltage of a quantum dot beyond the regime of linear response is called for.

In conclusion, we have presented an experimental study of the thermopower of a quantum dot in the Coulomb-blockade regime. The thermopower is observed to oscillate in a sawtooth

(²) Comparison of the experimental data in fig. 4 with the dashed curve, which corresponds to a quadratic increase of the amplitude with current, reveals that the linear-response regime ($\Delta V_{th} \propto \Delta T \propto I^2$) extends up to $I \lesssim 0.1$ μA.

(³) The average value of V_{th} in the inset of fig. 4 becomes more negative with increasing current. We attribute this to the increasing negative contribution of the reference point-contact to the measured thermovoltage (cf. eq. (1)).

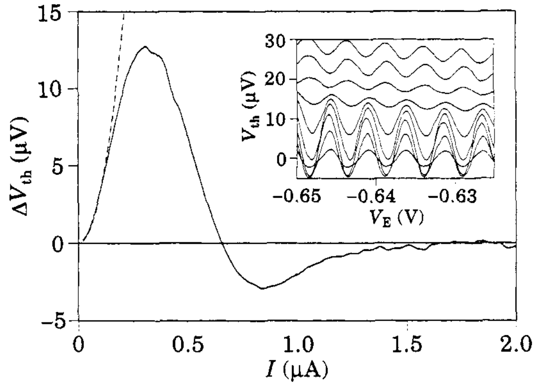

Fig. 4. – Peak-to-peak amplitude ΔV_{th} of the thermovoltage oscillations as a function of the heating current I, at a lattice temperature of $T = 45$ mK. The dashed line indicates quadratic increase of the amplitude with heating current. Inset: thermovoltage V_{th} *vs.* V_E at a lattice temperature of $T = 45$ mK. From top to bottom, the heating current is $0.1, 0.2, \ldots, 0.9$ μA.

fashion as the dot is depopulated, with a period equal to that of the Coulomb-blackade oscillations in the conductance. This is in agreement with a recent theory [4]. In the non-linear regime at large heating currents, a counterintuitive change of sign of the amplitude of the thermopower oscillations is observed, which remains to be understood.

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Additional remark. After we finished our manuscript, we received a preprint by Dzurak *et al.* reporting similar experiments.

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