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Single Electron Tunneling up to 300K

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A review is given of recent results obtained on single electron tunneling in ultra-small metallic nanostructures up to room temperature, using a scanning electron microscope. Additional results on semiconductor nanostructures will be presented at the conference.

1. Introduction

Recently, ultrasmall ($\lesssim$ 5nm in lateral diameter) double-barrier tunnel junctions have been realized using metal particles (2–5 nm in diameter) sandwiched in between a metallic substrate and the metallic tip of a scanning tunneling microscope (STM) [2]. The particles were either grown by e-beam evaporation or prepared using colloid chemistry.

Two electrical transport effects, in good agreement with the semi-classical theory of single-electron tunneling, have been found at room temperature: the Coulomb gap and the Coulomb staircase. The interpretation in terms of single-electron tunneling was further supported by liquid Helium temperature STM measurements on identical samples. In this abstract we review these results.

In the talk, but not in this abstract, also results will be presented of experiments on quantum dots defined in the two-dimensional electron gas in GaAs-AlGaAs heterostructures which prove that the amplitude of the Coulomb blockade oscillations reflects the details of the energy spectrum in the dot [3]. A review of this work is given in Ref. 4.

The most generic of the single-electron tunneling (SET) effects is the Coulomb gap: the suppression of the tunneling current at low voltages $\lesssim e/2C$. An additional manifestation is the Coulomb staircase, visible as a sequence of steps in the current-voltage characteristics, separated by voltage intervals equal to e/C. Each step corresponds to the addition of a single electron on the island.

So far, the observation of single-electron tunneling has been restricted to low-temperatures (typically $T \lesssim 4$K). The elementary charging energy $e^2/2C$, however, is inversely proportional to the capacitance C and can be enhanced by scaling down the structures to a smaller size. This is why it has been argued that single electron tunneling is the only known experimental phenomenon to be taken seriously as a possible basis for future room temperature electronics on the sub-10 nm scale (down to the realm of molecular electronics) [1].

2. Experiments

We have investigated double-barrier tunnel junctions that were realized by an STM tip situated above a small metal particle (tunnel junction 1 in Fig. 1a) which is separated by a tunnel barrier from the metallic substrate (tunnel junction 2) [2]. The two tunnel junctions are characterized by their resistances $R_i (i = 1, 2)$ and capacitances C_i. The capacitance relevant for the charging of the particle is $C = C_1 + C_2$. By measuring the electrical transport of such junctions we have recently observed the Coulomb gap and the Coulomb staircase at room temperature [2].

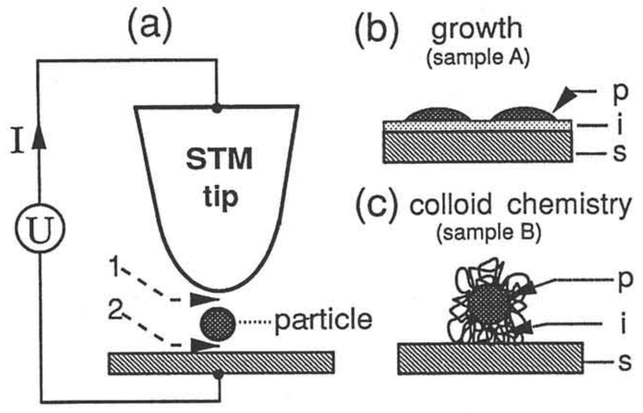

Fig. 1. Schematic arrangement of a double-barrier tunnel junction formed by a small metallic particle sandwiched in between an STM tip and a metallic substrate (a). The particle(p)-insulator(i)-substrate(s) system is either grown (b) or prepared by colloid chemistry (c).

Two types of particle-tunnel-barrier-substrate model systems have been investigated. Sample A (Fig. 1b) is grown by electron-beam evaporation and prepared as follows: First, a 100 nm thick Au film (conducting substrate) is grown epitaxially onto mica, followed by a 1 nm thick layer of ZrO_2 (oxide tunnel barrier). Finally, a 0.2 nm thick Au film is deposited. This film nucleates into small particles of approximately 4.5 nm in diameter. The capacitance C_2 of the particle-substrate junction is expected to be larger than C_1 due to the large dielectric constant $\epsilon_2 \sim 10$ of the oxide. We estimate $C = C_1 + C_2 \sim C_2 \sim 10^{-18}$F, corresponding to $e^2/2C \sim 70$ meV. This is well above kT at room temperature.

Sample B (Fig. 1c) is obtained using colloid chemical methods. An aqueous sol of Pd particles is prepared by electroless reduction of Pd^{2+} to metallic Pd in the presence of the water-soluble polymer polyvinylpyrrolidone (PVP). The polymer adsorbs onto Pd nuclei reducing their growth and preventing the flocculation of the sol by steric stabilization. By dipping a Au film (conducting substrate) into the sol, particles encapsulated by their polymer shell become adsorbed on the Au film. A tunnel barrier between the metal particle and the conducting substrate is now formed by the organic shell. This preparation method offers high flexibility: The particle coverage can be controlled over a wide range by varying the PVP concentration and the adsorption can be made specific by using surfactants on the substrate or by adding specific organic groups on the polymer chain (a detailed topographical STM investigation on such type of samples will be published elsewhere). The particle size distribution as obtained by transmission electron microscopy is quite monodisperse with an average diameter of 2 nm. The capacitance of the particle approximated by $C = 4\pi\epsilon_0\epsilon_r$ amounts to $\sim 4 \times 10^{-19}$F. This implies a charging energy as large as $e^2/2C \sim 0.28$eV.

The current-voltage (I-U) characteristics is measured with the STM tip situated above one particle for a fixed tip-particle separation. The measurements were either performed at room temperature with an STM operating in air, or with a low temperature STM operating in a helium gas atmosphere at 4.2 K.

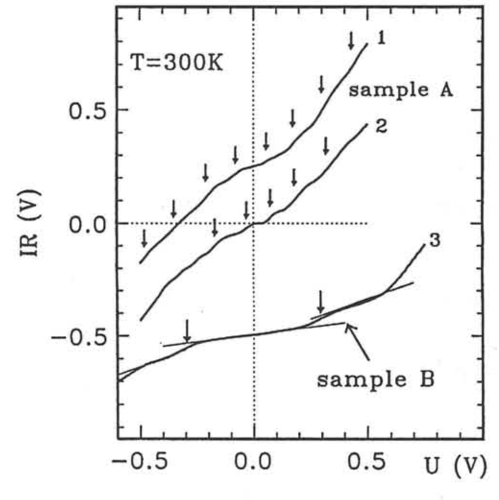

Fig. 2. Measured I-U characteristics at T = 300K showing the Coulomb staircase. Curve 1,2 were obtained on different particles on sample A and curve 3 on sample B. They are displaced vertically for clarity. Arrows indicate discontinuities in the measured current I. I is scaled by the asymptotic resistance R. ($R \sim 80, 80, 40\text{G}\Omega$ for curves 1-3, respectively).

A conclusive proof of single-electron tunneling in a double-barrier tunnel junction should include the observation of the Coulomb staircase which arises from incremental charging (by single electrons) of the intermediate particle. Fig. 2 shows three Coulomb staircases measured at room temperature. Curve 1,2 were obtained on sample A and curve 3 on sample B. Steps (indicated by arrows) in the I-U characteristics are clearly visible. Curve 1 was found to be in good agreement with the theory for asymmetric junctions taking into account the large temperature. Curve 2 appears to be displaced horizontally around zero bias. Such displacement has been explained by an "offset charge" q_0 (which is a continuous variable) induced on the particle.

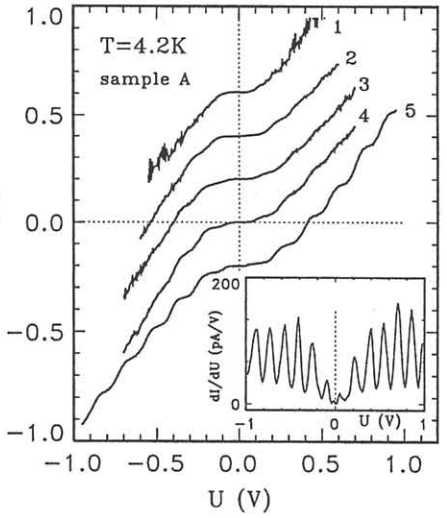

Fig. 3. Different types of I-U characteristics measured at $T = 4.2$K on different Au particles on sample A. Curves are displaced vertically for clarity. The current is scaled by the asymptotic resistance R ($R \sim 10, 1, 1, 0.5, 10\text{G}\Omega$ for curves 1-5, respectively). The inset shows the differential conductance for curve 5.

Fig. 3 shows several I-U curves obtained on different particles on sample A at 4.2 K. The most evident characteristic is the much stronger suppression of the zero-bias conductance which is due to the enlarged ratio $e^2/2CkT$. The size of the Coulomb gap and the periodicity of the staircase (curve 4 and 5) agree with the values found at $T = 300$K. This definitely proves that the observed effects at room temperature are single-electron tunneling effects. The sequence of curves in Fig. 4 demonstrates the different types of I-U characteristics that can be obtained depending on the resistance and capacitance ratios of the two tunnel junctions. Curve 1 represents a symmetric junction (Coulomb gap, but no incremental charging). At the other extreme, curve 5 represents an asymmetric junction (Coulomb gap and Coulomb staircase). The staircase is only observed on those particles for which $R_2 > R_1$. This is consistent with $C_2 >> C_1$ (large ϵ of the oxide).

3. Conclusion

In conclusion, the Coulomb blockade and the Coulomb staircase have been observed at room temperature as well as at 4.2K in ultrasmall double-barrier tunnel junctions consisting of an STM tip and a metallic substrate forming the two outer electrodes and an intermediate particle of size $2 - 4$ nm. This observation may stimulate serious considerations of the use of SET for future electronic devices. In addition, we have presented the first demonstration that a tunnel barrier may be realized by a polymer encapsulating a metallic particle.

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