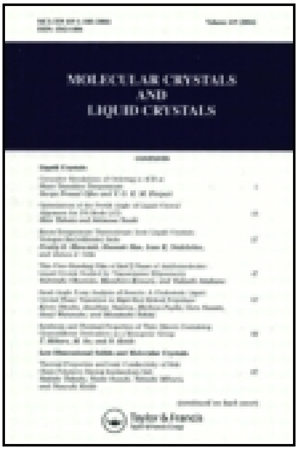

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Room Temperature Single Electron Tunneling in Nanoparticle-STM Tip Assemblies

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A novel approach to constructing double barrier tunneling junctions has been proposed for observing single electron tunneling(SET) phenomena at room temperature. PbS nanoparticles were directly synthesized on gold STM tip by self-assembly and *in-situ* chemical reaction techniques in this method. The SAM/PbS modified tip was used for STM-based current-voltage(*I-V*) measurements. Highly oriented pyrolytic graphite(HOPG) was used as substrate for electron tunneling. Clear Coulomb blockade and Coulomb staircase with equidistant voltage of ~110 mV were observed at room temperature with this novel assembly system. In addition, *I-V* curves also show an asymmetric shape at positive and negative voltages, suggesting a non-zero value of the fraction charge Q_0 existing on the Coulomb island.

Keywords: single electron tunneling; nanoparticle; tip assemblies

INTRODUCTION

Single electron tunneling (SET) phenomena, taking place in a system having extremely small capacitance, are of current interests due to the potential application in future nanoelectronic devices. A typical structure for observing SET phenomena is composed of a small metal or semiconductor island, the so-called Coulomb island, separated from two electrodes by two tunneling junctions. In such one double barrier tunneling junction (DBTJ) system, the addition of a single electron has an appreciable effect on the electrical potential across the junction, which leads to a quantized charging of the island when gradually increasing the bias potential. As a result, the Current-Voltage dependence shows a Coulomb blockade or Coulomb staircase effect, characteristic of a depression of current around zero bias or a stepwise increase of current, respectively. In the past few years, a great number of papers have appeared in this field, which cover both theoretical studies[1] and experimental approaches to fabricating various nano-sized tunneling junctions with different techniques[2,3].

In this paper, a new approach to forming ultrasmall double tunneling junctions is proposed. Our basic strategy is, using organic self-assembled monolayer (SAM) as the tunneling barrier and using the colloidal semiconductor nanoparticle immobilized on SAM as Coulomb island. In order to overcome the difficulty of Scanning Probe Microscope (STM) tip positioning on the nanoparticle, we directly synthesize PbS nanoparticles on the sharp end of STM gold tip using an *in-situ* chemical reaction, and then employ the STM technique to detect the tunneling current. The chemically functionalized tip here is incorporated into the tunneling barriers for room temperature SET studies.

EXPERIMENTAL

Au tips were prepared from 0.25mm-diameter gold wire(99.999% Nilaco Co.,Tokyo, Japan) by electrochemical etching in concentrated HCl at 1.6 V dc. After successive rinsing with de-ionized H_2O and ethanol, the tip was immersed in ~10^{-3} mol/L ethanolic solution of 11-mercaptoundecanoic acid (synthesized in our laboratory) for forming organic monolayer, followed by adsorbing lead ions (Pb^{2+}) for 6h in 10^{-4}mol/L Lead nitrate($Pb(NO_3)_2$) (Aldrich) solution. Thus treated tip was then exposed to H_2S atmosphere for 15 min to form Lead sulfide(PbS) nanoparticles on it. After structural characterization, the SAM/PbS modified Au tip was used for STM-based current-voltage (*I-V*) measurements.

Current-voltage measurements were carried out with a commercial NanoScope IIIa (Digital Instruments(DI) Inc.) in air at room temperature. The typical setpoint current was locked as 1nA and bias voltage was set as 1V. Once the tip was engaged under selected conditions, the feedback system was switched off and the *I-V* characteristics were measured. Highly oriented pyrolytic graphite(HOPG) was used as substrate for electron tunneling because of its easy cleaning in air and large areas of atomically flat plane.

Unmodified Au tips were characterized by scanning electron microscope(SEM) using an Amray 1919 Field emission microscope(USA) at an operation voltage of 5kV. SAM/PbS Modified Au tips were studied by transmission electron microscope (TEM) using a Hitachi H-9000 at an operation voltage of 300kV.

RESULTS AND DISCUSSION

The preparation of tunneling tips is the critical point of this study. For the initial electrochemical etching of gold wire, the current in the electrochemical cell was monitored as the gold wire necks down near the surface of concentrated HCl solution. A sharp drop in the current indicates the drop-off of the submerged portion of gold wire, leaving an optimally sharp upper tip. This technique enables us to routinely

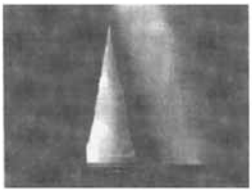

FIGURE 1 Field emission scanning electron micrograph of a made gold tip

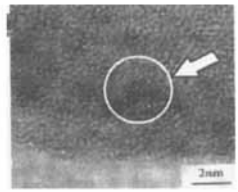

FIGURE 2 High resolution TEM image of a PbS nanoparticle on the tip apex

make tips with a radius of 10~20 nm evidenced by SEM image(Figure 1). Figure 2 shows a typical TEM image of the gold tip apex modified with SAM and PbS nanoparticle, on which a PbS nanoparticle with a size of ~ 4nm was observed clearly. The spacing between diffraction fringes of the nanocrystal is 0.34 nm in nice agreement with that of PbS{111}(d=0.3429nm). We found that ~70% thus prepared tips show clear SET phenomena, demonstrating the technical merit of the proposed fabrication method of double barrier tunneling junctions.

Figure 3 presents the single electron tunneling phenomena observed with the SAM/PbS modified Au tip. The tunneling current exhibits a stepwise increase with increasing the bias voltage. The dI/dV curve shown in Figure 3c gives a series of equidistant peaks, typical of Coulomb staircase phenomenon. The step-width of voltage is ~110 mV, corresponding to an electrostatic energy of ~55 meV, higher than thermal excitation energy at room temperature(~26meV). The equidistant voltage steps allow us to estimate the tunneling capacitance C_Σ, which was found to be ~1.4 aF, being of the same order(C= ~3.7aF,C= $4\pi\varepsilon_0\varepsilon_r r$, ε_r =17.9) as that estimated from simple calculations based on a spherical PbS nanoparticle with a radius of 2 nm. It is noted that the I-V dependence shows an asymmetric shape at positive and negative voltages, suggesting a non-zero value of the fraction charge Q_0 existing on the Coulomb island.

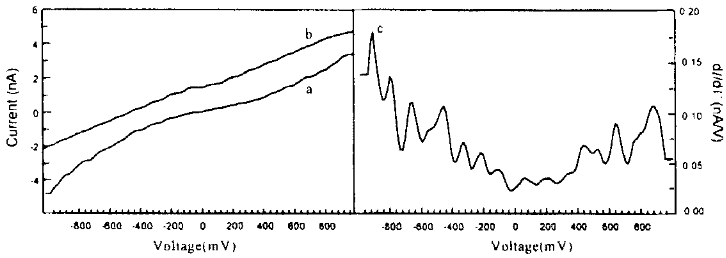

FIGURE 3 Coulomb staircase observed with SAM/PbS modified tip at 289K (a) experimental (b) theoretical and (c) dI/dV

Based on the semi-classical tunneling model[4], we performed computer simulation, which gives a theoretical fitting curve as shown in Figure 3b. Because of the asymmetric nature in *I-V* characteristic, we added a fractional charge Q_0 in the fitting process. The best fitting parameters are $C_1 = 1.38 \times 10^{-18}$ F, $C_2 = 3.50 \times 10^{-19}$ F, $R_1 = 160.0$ MΩ, $R_2 = 20.0$ MΩ, $Qo/e = -0.2$.

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