Fabrication and characterization of Au island single-electron transistors with CrO_x step edge junctions

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Single-electron transistors fabricated using Au islands and CrO_x resistive microstrips are reported. To investigate the occurrence of Coulomb blockade in these devices, three types of device designs have been tested. Typical single-electron behavior, conductance modulation by the gate, is observed in the devices which had small overlap area with the gold island. Electron transport mechanism of CrO_x resistors is discussed and a hypothesis of the formation of step edge junctions at the edges of granular metal microstrips is given as the explanation for the experimental results. © 2004 American Vacuum Society. [DOI: 10.1116/1.1815310]

I. INTRODUCTION

The single-electron transistor (SET) (Refs. 1 and 2) is a device based on the Coulomb blockade effect. It consists of two tunnel junctions connected in series, with a gate electrode placed near the small island formed between the two junctions. There are two basic requirements³ to observe single-electron tunneling effects in a SET. First, the total capacitance of the island, C, must be small enough that the charging energy $E_C = e^2/2C$ is much greater than thermal energy k_BT . Second, the resistance of the tunnel barriers, R_T must exceed the resistance quantum $R_0 = h/e^2 = 25.8 \text{ k}\Omega$ to suppress quantum charge fluctuations. If these conditions are satisfied, the number of electrons on the island is fixed, and additional energy beyond thermal energy is needed to add even a single electron. This is referred to as the Coulomb blockade. In this device, discrete electrons tunneling through the junctions is controlled by the gate which is capacitivily coupled to the island.

Since the first successful experimental implementation of the single-electron transistor,¹ a number of different geometries, materials, and methods have been used for the fabrication of single electron devices^{4–6} were reported. Recently, Nazarov proposed a general theory of Coulomb blockade⁷ that can embrace tunnel junctions, quantum point contacts, diffusive conductors, and any type of scattering. According to this theory, tunnel junctions are not necessary to provide insulation required discrete charging effects and they may be replaced by an arbitrary scatterer. However, the scatterer of the same resistance as a tunnel junction suppresses Coulomb blockade exponentially by a factor of $\exp(-\alpha G/G_0)$, where α is a dimensionless coefficient depending on the type of the conductor, and $G_0 = 1/R_0$. This theory leaves open the effect of capacitance on Coulomb blockade (requirement 1), by assuming that this requirement is fulfilled.

Previous work in this area has produced SETs with resistive strips augmenting tunnel junctions,⁸ and more recently single-electron transistors with metallic microstrips instead of tunnel junctions were reported.⁹ The experiments by Lotkhov *et al.*⁸ also showed that microstrips with resistance nR_Q act as *n* tunnel junctions connected in series, strongly suppressing the cotunneling (a coherent quantum process consisting of multiple simultaneous tunneling events). In quantum-dot cellular automata (QCA) (Refs. 10 and 11) devices, cotunneling severely impairs the ability of cells to store information. To suppress cotunneling, complicated multiple-tunnel junction (MTJ) structures were used by Orlov *et al.*¹² Using resistive microstrips instead of MTJ, the number of junctions can be reduced. This eliminates the problem of junction random background charge compensation in extra dots of MTJs and greatly simplifies the design of cells.

The purpose of our work is to evaluate single-electron devices using resistive microstrips and to investigate its applicability for QCA fabrication. In this article, we present our work on the fabrication and characterization of SETs with gold islands and CrO_x resistive microstrip barriers. The electron transport mechanism of CrO_x resistors is also discussed and a hypothesis of the formation of step edge junctions is given as the explanation for the experimental results.

II. FABRICATION

The devices were fabricated by two steps of e-beam lithography and metal deposition, as shown in Fig. 1. Choosing Au instead of Al as the island metal eliminated the problem of parasitic junction formation caused by native Al oxide. Two versions of SET geometry, two CrO_x wires connected by a gold island [Fig. 1(a)] and one continuous CrO_{r} wire atop a gold island [Fig. 1(b)], were used in all of our experiments. Each version of geometry has three types of pattern design which will be expanded below. Surprisingly, no significant difference between the two versions of geometry was observed in the electrical characterizations. For simplicity, Fig. 2 shows only the three types of pattern using the design of Fig. 1(b). The first layer of metal (2 nm Ti and 10 nm Au) was patterned by e-beam lithography and lift-off to define the island and electrodes. A second e-beam lithography and deposition step was used to form the CrO_x resistive microstrips connecting the island to the electrodes. In

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FIG. 1. Two versions of SET geometry, two CrO_x wires connected by a gold island (a), and one continuous CrO_x wire atop a gold island (b). The inset is the cross section at the overlap area of the Au layer and the CrO_x layer. The CrO_x wires are $0.375-2 \ \mu\text{m}$ long, 70 nm wide, and $6-10 \ \text{nm}$ thick. The thickness of the Au layer is 10 nm. The size of Au island is varied from 200 nm by 500 nm to 300 nm by 1 μ m.

the second evaporation, a thin film of Cr (8-10 nm) was evaporated in the oxygen ambient. Different values of sheet resistance of CrO_x film were achieved by changing the oxygen pressure in the chamber during evaporation.

III. CHARACTERIZATION OF CRO_X RESISTORS

To obtain high quality CrO_x resistive microstrips with controllable resistivity, we performed extensive characterization of the deposition process. *In situ* measurements were taken of the resistance of the microstrips during deposition of Cr in an O₂ ambient at different oxygen pressures. The results showed that the wires became conducting when the film thickness reached 2 nm. For 10 nm thick CrO_x , sheet resistances in the range of $1 \text{ k}\Omega/\Box$ to $10 \text{ k}\Omega/\Box$ were formed when the oxygen pressure was around 3.6×10^{-5} Torr in our evaporation system. A slightly higher ($\sim 4.5 \times 10^{-5}$ Torr)



FIG. 2. Schematic view of three types of pattern design. (a) Type #1: CrO_x layer consists of narrow lines (~70 nm) only. (b) Type #2: large tabs (wider than 300 nm in two dimensions) on both ends cover all of the steps where the two layers of metal overlap. (c) Type #3: large tabs only cover the steps of source and drain and no tabs appear on the island.



FIG. 3. Temperature dependence of a CrO_x resistor obeys the Schklovski– Efros (SE) law of the form $G(T)=G_0 \exp(T_0/T)^\nu$, where $\nu=1/2$. The top inset is conductance vs 1/T of CrO_x resistors in different resistance range. CrO_x wires with low resistance showed weak temperature dependence. As the resistance increases, temperature dependence became stronger. The bottom inset shows that the *IV* characteristic of the microstrip is nonlinear due to intrinsic Coulomb blockage.

oxygen pressure resulted in much higher sheet resistance and the films were completely insulating with even higher pressure. Although there is scatter in the sheet resistance for different depositions with similar oxygen pressure, there is a clear tendency of increasing resistance with increasing oxygen pressure.

Chromium has three common oxides.¹³ Cr_2O_3 is an insulator with stable chemical prosperities and has a green color, CrO_3 is an *n-type* semiconductor with a dark red color, and CrO_2 is a half-metal ferromagnet¹⁴ with a black color. When the temperature is higher than 250 °C, CrO_3 and CrO_2 decompose into Cr_2O_3 and O_2 . In our experiments, the color of sample holder was always green after deposition. Although there is a possibility that the green color was caused by interference effects due to thickness, with the additional information provided by the sheet resistance we obtained from deposition at different O_2 pressure, we conclude that the oxide deposited was Cr_2O_3 . Similar results were obtained by other groups using a similar deposition process.⁹

To study the transport mechanism of the CrO_x microstrips, we characterized strips with the same dimensions as those used in the single-electron transistor samples. We believe that the resistive microstrips consist of granules where each granule consists of a Cr core with a Cr_2O_3 shell. Electrons are tunneling from granule to granule through the Cr_2O_3 insulating layer, and higher oxygen content gives a thicker shell that changes the conductance behavior from metallic to insulating as intergranular oxide resistance approaches R_Q . It is commonly observed that conductance of the granular metal films composed of oxidized metal grains^{15,16} obeys the Schklovski–Efros (SE) law¹⁷

$$G(T) = G_0 \exp(-(T_0/T)^{1/2}), \qquad (1)$$

where parameter T_0 increases with increased with increase of oxygen content. We observe this type of temperature dependence for microstrips with sheet resistances $R > R_Q$ at room temperature (Fig. 3). *IV* characteristic of the microstrips in this case is strongly nonlinear due to intergranular Coulomb

blockage (bottom inset on Fig. 3).¹⁸ As was recently shown by Zhang and Shklovskii,¹⁷ the presence of charged impurities in the insulator smears the density of states at the Fermi level which explains the observation of SH law in temperature dependence in granular metal films. For microstrips with sheet resistance $R \ll R_Q$, weak temperature dependence and almost linear *IV* are observed down to 0.3 K.

IV. CHARACTERIZATION OF SINGLE-ELECTRON TRANSISTORS AND DISCUSSION

To form SETs, and study the origin of the Coulomb blockade in our devices, three types of patterns (Fig. 2) in each of the two versions of Fig. 1 were fabricated. In all of these, the first layer of metal (Au) is the same, defining the islands and electrodes. The difference is only in the second layer (CrO_x) at the overlap areas.

In the first type of design [type #1, Fig. 2(a)], the CrO_x layer consists of narrow lines (~70 nm) without tabs on the overlap areas. Only a small number (<20%) of devices showed finite conductance at room temperature and these resistances were distributed in a rather large range. Also most of the devices degraded rapidly to an open circuit when exposed to air. Therefore no low temperature data were obtained for these devices. A possible source of these failures is a step-coverage problem.

In the second type [type #2, Fig. 2(b)], there are large tabs (wider than 300 nm in two dimensions) on both ends of the lines which cover all of the steps where the two layers of metal overlap. Over 95% devices showed conductance at room temperature. The CrO_x films were rather uniform (variation ±20%) and lasted for a relatively long time when exposed to air. In the range of 2 k Ω/\Box to 7 k Ω/\Box , significant nonlinearities around zero bias and a temperature dependence characteristic of Schklovski–Efros law were observed (in the devices with sheet resistance greater than 7 k Ω/\Box , conductance was "freezing out" below 5 K); however, none of the devices exhibited Coulomb blockade oscillations. These experimental results indicate that the resistive microstrip itself does not provide Coulomb blockade of electrons on the island.

In the third type [type #3, Fig. 2(c)], large tabs cover only the steps of source and drain and no tabs appear on the island. The room temperature measurements showed that 95% devices were conducting, with good uniformity of resistance. However, among those devices having significant nonlinearity in I-V curves at 300 mK, only about 30% exhibited Coulomb blockade oscillations. From a study of the yield vs resistance (Table I), we found that Coulomb blockade oscillations were only observed when the resistance of devices was greater than 100 k Ω at 0.3 K, and devices with higher resistance were more likely to show Coulomb blockade oscillations. The value of charging energy was varying significantly for the devices of the same batch (0.01-1 meV). The two devices which have the largest charging energy (~0.4 meV) had about 5 M Ω resistances at the "open" state (when the Coulomb blockade is lifted) at 300 mK. The I-Vcurve [Fig. 4(a)] and $I-V_g$ modulation curve [Fig. 4(b)] of

TABLE I. Yield vs resistance.

Resistance range	$R \! < \! 100 \text{ k}\Omega$	$\begin{array}{c} 100 \text{ k}\Omega < R \\ < 200 \text{ k}\Omega \end{array}$	$\begin{array}{c} 200 \text{ k}\Omega < R \\ < 1 \text{ M}\Omega \end{array}$	$R > 1 M\Omega$
Total number of devices	12	9	5	4
Number of devices showed CBO	0	5	3	3
Yield	0%	56%	60%	75%

one of the devices clearly demonstrate single-electron transistor behavior with strong nonlinearity in the open state.

We also investigated the gate dependence of single microstrips without an island, but no change in conductance was observed over a large gate bias span (± 1 V). Since a single microstrip consists of a large number of granular islands with random offset charges, a common gate cannot synchronously control the Coulomb blockade of individual islands. The I-V curve therefore does not show any dependence of the gate bias.⁹

Based on these observations, we conclude that in the devices which show Coulomb blockade oscillations some sort of weak links with small intrinsic capacitance and $R > R_Q$ are formed at the edges of the CrO_x wires overlapping the Au islands. Indeed, the Coulomb blockade is observed only on those samples with narrow lines (~70 nm) overlapping the island (type #3) [Fig. 5(a)]. The CrO_x line must be thinner at the overlapping edges where it climbs onto the island. Taking into account the thickness of the island metal (12 nm) and the thickness of the CrO_x strips (8–10 nm) it is likely that the CrO_x at these edges has breaks or is more readily oxi-



FIG. 4. (a) I-V curves of SET in open state and blocked state. (b) $I-V_g$ modulation curve of the same SET of (a) measured at 300 mK showed deep modulation by the gate. Charging energy is about 0.4 meV.

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FIG. 5. (a) AFM image of a CrO_x wire deposited on the edge of the Au island. (b) The AFM image revealed that only two edges were covered by large tabs in the sample with a pattern shift.

dized than along the strip, forming weak links to the island. The number of oxidized granules in the 70 nm wide line is about 10-20 [grain size 5-15 nm (Ref. 16)]. These weak links, which consist of a few oxidized Cr granules, having small capacitance with resistance $\sim R_Q$ comprise the lowcapacitance junctions needed for Coulomb blockade of electrons on the island. The single period of $I-V_g$ modulation curves indicated there are only two junctions (one island) in those devices. An undesirable property of this fabrication technique is that the junctions are formed randomly. A significant number of devices did not show single-electron behaviors even though they had the same pattern design and sheet resistance as those that showed Coulomb blockade effects. In devices with the large tabs covering the steps it is likely that shorting bridges are formed across the steps and/or that the capacitance was sufficiently large to suppress Coulomb blockade effects at a temperature of 0.3 K. The charging energy $(E_C = e^2/2C)$ depends only on the total capacitance of the island which is dominated by the capacitances of the tunnel junctions. Large variation in oxide thickness of weak link junctions leads to large variation in junction capacitance. Thus, the value of charging energy varied significantly for the devices of the same batch. Exposure to air after fabrication leads to further oxidation of the devices. Oxidation of the CrO_x granules at the weak links causes the observed degradation of devices. Type 1 devices have two more weak links than type 3, and one weak link's failure degrades the whole device. Thus, type 1 devices should degrade faster than type 3 devices. In addition, type 1 devices have higher resistivity than the other two types of

FIG. 6. Multiple frequencies in $I-V_g$ modulation curves of the devices with a very rough surface of CrO_x films indicate the formation of multiple islands in the devices.

devices and our experiments showed that devices with higher resistivity degraded faster. We also fabricated wires with width 1 to 100 μ m with all of which showed a small variation (~10%) in the resistance over time (several weeks).

The above results were obtained from "normal" devices, where the patterns in two steps of lithography are properly aligned to each other and CrO_x films are uniform and have a relatively smooth surface. The results from "abnormal" devices also support our conclusions. Single period oscillations were also observed in devices having big tabs on all edges, but with a pattern alignment shift between two layers. The AFM images [Fig. 5(b)] revealed that only two edges were actually covered by large tabs while the other two were not. In this case, two junctions were formed at the edge of island and one electrode, respectively; the island consists of the gold island and one arm of the CrO_x wire. It is worth nothing that the CrO_x film and Au island acts as a single entity. Multiple frequencies of $I-V_g$ modulation curves (Fig. 6) were also observed in the devices which had a very rough surface of CrO_x films. Multiple frequencies are the indication of multiple islands in the devices. Strong thickness fluctuations along the wire could lead to formation of the thicker segments of CrO_x wire acting as islands.

V. SUMMARY

We demonstrated that single-electron transistors can be fabricated using two steps of e-beam lithography and deposition of CrO_x microstrip resistors in oxygen ambient. Although the electron transport mechanism needs to be studied further, we show that resistive microstrips by itself can not fulfill the first requirement for Coulomb blockade oscillations $(E_C = e^2/2C \gg k_B T)$. However, in the devices where "weak link" junctions are formed with small capacitances between microstrips and islands, both requirements for R_T and C are fulfilled and Coulomb blockade oscillations are observed. It is likely that weak link junctions or parasitic oxide layers were formed at Al/CrO_x interface in Ref. 9, leading to the observed SET behavior. Although resistive microstrips cannot by themselves be used to produce singleelectron transistors, they still are useful in suppressing the cotunneling effects.

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