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Resistance Imaging with a Scanning Electron Microscope

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# RESISTANCE IMAGING WITH A SCANNING ELECTRON MICROSCOPE

By

Qifu Zhu

# A DISSERTATION

Submitted to Michigan State University in partial fulfillment of the requirements for the degree of

# **DOCTOR OF PHILOSOPHY**

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#### ABSTRACT

# RESISTANCE IMAGING WITH A SCANNING ELECTRON MICROSCOPE

By

Qifu Zhu

This thesis describes two separate research projects. The first project is the development of a technique, called electron beam resistance imaging (EBRI), to measure electrical resistance variations along lithographic metal lines. With this technique, we have studied the onset of failure of metal lines by electromigration and local heating effects by imaging the resistance and then slowly increasing the external current to the failure, recording the evolution of the resistance at hot spots. A serious technical problem in the implementation of this technique is the formation of carbonaceous contamination on sample surfaces under the action of the electron beam in the scanning electron microscope. Several methods to prevent and remove this contamination have been compared. In the second project, a mechanically-controlled squeezable tunnel junction has been built to study single-electron tunneling phenomena through a vacuum barrier. Using a squeezable tunneling junction with a micron-sized sample, two-level fluctuations of the conductance in the tunneling regime has been observed at liquid nitrogen temperature.

To my parents

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# Chapter 1

# Introduction

This thesis describes two distinct research projects. The first project is the development of a new technique to image resistance variations in lithographically fabricated metal circuit lines with a scanning electron microscope (SEM). The second project is a study of single electron tunneling effects with two different mechanically-controlled, adjustable-gap tunnel junctions.

Scanning electron microscopy has been used in both the fabrication and testing of semiconductor devices since the development of integrated circuits in the 1960's. With an electron beam spot size as small as 1 nm, the scanning electron microscope (SEM) is now being used to fabricate sub-micron circuitry, producing computer chips with higher transistor density, that are faster, cheaper, and more energy efficient. The SEM can also be used to probe the function of a working integrated circuit. When the high energy electron beam (typically 1-50 keV) of an SEM strikes the surface of a metal or semiconductor, many reaction products are produced including x-rays, backscattered electrons, and secondary electrons. Secondary electrons have energies less than 50 eV and are emitted from parts of the sample within 5 nm of the surface. The flux of these low-energy secondaries is extremely sensitive both to the morphology of the sample surface and to local electric fields. A surface with a positive voltage tends to pull secondary electrons back to the surface and appears dark in a secondary electron image, while a negatively charged surface appears bright. This phenomena has been exploited in the technique of voltage contrast imaging, which is routinely used to test the function of chips. Circuit lines on the chip are seen to alternate dark and bright as the logic levels switch high and low. Although the 0-5V levels of standard TTL logic is easily imaged with voltage contrast electron microscopy, the IR voltage

drop along metal circuit lines is much too small to be seen with this technique. In this thesis, I describe the development of a technique to image this small IR voltage drop along metal lines, a technique which has been dubbed electron beam resistance imaging (EBRI). In this technique, the electron beam of the SEM injects current at a point along the metal sample line, and the resulting voltage drop is measured with external circuitry. This technique has a voltage resolution of a few nanovolts, a resistance resolution of a fraction of an ohm, and a spatial resolution approaching  $0.1 \mu m$ .

This technique was first used in 1965 by Watanabe and Munakata, who measured variations in the resistivity of bulk semiconductors.<sup>1</sup> Long and Slichter used a variant of the technique to study organic semiconductors in 1979.<sup>2</sup> However, both these groups studied non-metallic samples with very high resistivity. The primary goal of my work was to extend the sensitivity of the technique to allow characterization of metals. As a test, we studied the evolution toward failure of an aluminum circuit as it was stressed with a large external current. Electromigration is the primary cause of circuit failure in circuit lines and is a subject of active investigation in the electronic industry. The technique of EBRI may provide a powerful new probe of this phenomena.

One of the primary technical difficulties in implementing EBRI is the formation of carbonaceous contamination on the surface of the sample due to polymerization of contaminant molecules under the action of the energetic electron beam in a SEM. We tried several different ways to eliminate this contaminatin including replacement of the conventional diffusion pump oil by perfluoropolyethers (PFPE) type diffusion oil, the construction of a coldshroud around the sample and installation of a gas jet to clean the surface.

In the second research project, single-electron tunneling phenomenon were studied with a home-built, mechanically-controlled squeezable tunnel junction and a crack junction. The tunneling dynamics of a small junction with a very small capacitance has become a topic of intensive investigation in last several years, both theoretically and experimentally. In small capacitance junctions, there occurs a new regime of tunneling, single electron tunneling. Applications of single electron tunneling include the fabrication of precision current meters that can count electrons one-by-one.<sup>3</sup>

The remainder of this thesis is organized in the following way. In Chapter 2, I describe the procedures for use of photolithography and e-beam lithography to fabricate the micron-sized and submicron-sized samples used in the two research projects of this thesis. Chapter 3 and Chapter 4 describe the first research project -electron beam resistance imaging (EBRI)-a technique to measure electrical resistance variations along lithographic metal lines. A serious technical problem in the implementation of this technique is the formation of carbonaceous contamination on sample surfaces. In Chapter 3, I describe several methods employed to prevent and remove this contamination. In Chapter 4, I describe the principles of EBRI and the circuitry used to implement it, as well as some experimental results on resistance imaging of a metal line damaged by electromigration. In Chapter 5, I describe the second research project, development of two types of mechanically-controlled tunneling junctions, a squeezable junction and a crack junction. I also discuss some single-electron tunneling phenomena that were observed with these junctions.

<sup>1</sup> Hiroshi Watanabe and Chusuke Munakata, Japan. J. Appl., <u>4</u>, 250 (1965).

<sup>&</sup>lt;sup>2</sup> James P. Long and Charles P. Slichter, *Phys. Rev. B*, <u>21</u>, 4521 (1980).

<sup>&</sup>lt;sup>3</sup> L. J. Geerling, V. F. Anderegg, P. A. M. Holweg, and J. E. Mooij, *Phys. Rev. Lett.*, <u>64</u>, 2691 (1990).

# **Chapter 2**

# Sample Fabrication—Photolithography and E-beam Lithography

## 2.1 Introduction

Over the last several decades, the line width in integrated circuits has been pushed down from 3  $\mu$ m to 0.6  $\mu$ m. Many new technologies, such as electron beam lithography and soft x-ray lithography, have been simultaneously developed in order to make smaller, faster and more energy efficient chips. Electron beam lithography is widely used for the fabrication of master masks for contact lithography because it is capable of writing submicron lines directly on a quartz substrate coated with a Cr film. Contact lithography - a method for transferring patterns from a mask to a wafer - is still one of the most popular technologies in modern integrated circuit industry due to its unique capability of fabricating chips in a large scale. As a result, the CPU in modern personal computers, e.g. the "P6" processor manufactured by Intel Corp. using contact lithography, has 0.6  $\mu$ m lines, and has 5 million transistors in such a 0.5 inch by 0.5 inch area<sup>1</sup>.

In our lab, contact photolithography, projection photolithography and electron beam lithography have all been used together to fabricate samples for the studies of electromigration and the Coulomb blockade effect. These techniques have differing resolution. In our lab, we use contact photolithography to fabricate  $10\mu m$ lines using a photographic negative as a mask. We use projection photolithography to make  $1\mu m$  lines, a resolution which is limited by the wavelength of the Ultraviolet (UV) light source. And we have fabricated 0.1  $\mu m$ wide metal lines with e-beam lithography using a scanning electron microscope. Once a pattern is transferred from the mask to the photoresist, layers of thin metal films, such as  $Al/SiO_2/Al$  and Au/Cr, are deposited by thermal evaporation at a base pressure of  $1x10^{-7}$  torr. This chapter describes these procedures for sample fabrication in detail.

#### 2.2 Contact Lithography and Projection Lithography

#### 2.2.1 Photography of Mask Patterns

In our lab, contact photolithography is used to make  $10\mu m$  wide metal lines on silicon wafers, a line width which is limited by the minimum feature size in the mask. A photographic negative is used as the mask, and an ultraviolet light source is used to transfer the mask pattern to a silicon wafer which is coated with photoresist. The following procedure is used to make a mask.

First, a desired pattern is drawn and printed out by a laser printer with contrast and resolution as high as possible and with the pattern's edge as sharp as possible. The pattern's dimensions are calculated according to the desired line width in the final sample and the demagnification of the camera. For example, in order to produce a negative with a 50  $\mu$ m wide line using a camera with a demagnification of 20X, a line with a 1 mm width must be drawn on paper. Typically, laser printers have a resolution 300 dpi (dots per inch), with some as high as 600 dpi. These are ideal for making mask patterns. Before laser printers were available to us, we used an HP plotter with a 0.5 mm diameter, special dark ink pen. The hardcopies coming from such a plotter have two serious problems: (i) a line with a dimension smaller than a plotter pen's diameter of 0.5 mm cannot be drawn; (ii) even for a wide line, for example, 1 mm wide line, the edge is not sharp because the ink diffuses in the paper. Later, using a sharp razor, we cut dark paper into different shapes with the required dimensions and patched them together on white paper as the background. We also used transfer decals (trans-artype<sup>2</sup>) to produce fine lines with a width of 0.5 mm, because it is almost impossible to cut such narrow lines by hand with a razor. With the laser printers now available, it is much easier to make and modify a mask using software, such as DrawPerfect, HarvardGraphics, MicroSoftDraw, AutoCAD, etc. A laser printer produces satisfactory masks with high contrast, uniformly dark area and sharp edge definition, as desired.

The next step is to make a negative mask with a high contrast negative film, Kodalith Orth Film 9656, Type III (ASA 6), manufactured by Kodak Corp.. Kodalith Orth 9656 has the highest contrast among commercial negative films, with a resolution of a few µm's, which serves well for contact pads requiring dimensions larger than 10  $\mu$ m. When photographing the laser printer pattern, the paper pattern is covered by a large glass plate, in order to keep the entire pattern in the same focusing plane. (The paper tends to curl under the hot illumination lamps.) A Micro-Nikor 55 mm f/2.8 lens is used together with an FM2 Nikon camera body. For each pattern, several exposures are made by varying the exposure time, the aperture stop, and the position of the light source. The maximum demagnification that we use is 15X, limited by the maximum distance between the camera and the mask pattern allowed by our Polaroid MP4 camera With 10X demagnification and without the glass cover plate, the stand. appropriate exposure conditions are as follows: f/4(aperture)-1/8 sec. exposure time or f/5.6—1/4 sec., with two 150 watts tungsten-lamps illuminating the mask pattern. When 15X demagnification is used, the exposure time is somewhat longer with the same aperture and illuminating light because of the larger object distance. We found that use of the glass cover plate reduces the exposure time because the glass increases the reflection coefficient of the white paper. The best exposure

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time is 1/15 sec. with the aperture set at f/4 and four 150 W-lamps illuminating the sample. Shorter exposure times are preferred in order to reduce fuzzy edges caused by vibration of the table.

The recipe for developing the high contrast films is:<sup>3</sup>

Kodalith Super RT developer(Part A & B): 2 min. 45 sec.;Stop Bath:30 sec.;Kodak Fixer:3 min.;Water rinse:30 min.;Dry in air overnight.

A stop bath is a dilute acetic acid solution for stopping further development by the developer, and a fixer is used to stabilize the image. For developing one roll of film with 24 exposures, 8 oz (236 ml) of each solution is needed.

After developing the films, we view them with an optical microscope, looking at the silver grains under 1000X magnification with transmitted light illumination. This helps us to choose the best negative to use as a mask. We are now ready to transfer the mask pattern to a substrate by contact lithography.

# 2.2.2 Contact Photolithography

The most frequently used method for transferring a pattern from a mask to a sample substrate is contact lithography. This is a well-known technique, and there are many excellent descriptions of this technique in the literature.<sup>4,5</sup> Fig. 2.1 illustrates the basic procedure for contact photolithography.

A substrate is cleaned by a standard procedure:<sup>6</sup>

rinse in acetone for several seconds; rinse in methyl-alcohol (methanol) for several seconds; rinse in deionized water (DI) for two minutes;



Figure 2.1 This diagram shows the main steps of contact photolithography for a positive resist.

dry with nitrogen gas.

After being prebaked at about 200°C for 30 minutes in a hot plate oven to remove water from the surface, the substrate is coated with 1µm thick layer of positive photoresist S1811 from SHIPLEY company<sup>7</sup>, using a spinner at a speed of 5,000 rpm. A positive photoresist is a cross-linked polymer with chemical bonds that can be broken by exposure to UV light, and such an area with broken bonds can be dissolved by a special solvent, called a developer. Conversely, a negative photoresist has the opposite property, the exposed area is insoluble to the solvent. The resist-covered substrate then goes through a softbake procedure -- baking at 90-100°C controlled in the air for 30 min. to drive the carrier solvent out of the photoresist.

The crucial step for contact lithography is the UV exposure. A block diagram for this procedure is shown in Fig. 2.2. A 100W mercury lamp, with a collimating lens, is used as the UV source.<sup>8</sup> A simple camera shutter controls the exposure time in this home-built exposure system. In order to get uniform illumination in the 1 inch diameter of the field of view, a frosted glass plate is inserted into the light path, just above the shutter. It acts to scatter light and reduces the intensity of the UV light by half at the contact plane. With the frosted plate, the exposure time is typically 10 seconds. Optimum edge definition is achieved when the side of the negative coated with silver grains (the emulsion layer about  $0.2\mu$ m thick) faces the photoresist on the substrate. The negative film acetate base substrate (about 0.15 mm thick) scatters light and makes the edge profile fuzzy when the acetate is between the emulsion and the photoresist. In order to make good contact between the mask and the photoresist, a glass or quartz plate is placed on the mask held in place by two metal

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Figure 2.2 Block diagram for UV exposure system: (A) lamp housing, (B) 100W mercury lamp, (C) reflecting mirror, (D) quartz collimating lens, (E) reflecting mirror, (F) shutter, (G) stage with sample and mask; (I) glass or quartz cover plate, (II) metal bar held down with (III) 4-40 screws.

bars. This mimics the situation in a commercial mask aligner, in which a mechanical pump is used to create a vacuum between the mask and the photoresist, so that air pressure produces good contact. Of course, in this case, the mask must be on a solid substrate instead of a floppy negative film. In the integrated circuit industry, the mask is often made on fused silica (quartz). One side is coated by a Cr or Fe<sub>2</sub>O<sub>3</sub> film which is exposed with direct electron beam writing. Then the Cr or Fe<sub>2</sub>O<sub>3</sub> is etched during development so as to produce a master mask. In the most of my work, samples were fabricated using floppy negative film masks; some commercial Fe<sub>2</sub>O<sub>3</sub> film masks were also used.<sup>9</sup>



Figure 2.3 Pattern edge profiles: (i) over-cut; (ii) vertical-cut; (iii) under-cut. An under-cut profile is ideal for lift-off.

Right after being exposed to UV light, the substrate with photoresist is placed in Shipley Micro Developer 452 for 45 seconds, a development time which often produces an over-cut edge profile. The pattern edge profile is described as overcut, vertical-cut, or and under-cut, as shown in Fig. 2.3. Obviously, the pattern edge profile has to have an under-cut in order to produce clean patterns after liftoff. The lift-off technique is a process which removes the metal on the top of the photoresist by dissolving the photoresist underneath with a solvent, such as acetone. Achieving the desired under-cut profile requires a special treatment of the photoresist in chlorobenzene, as discussed in section 2.5.1.

When we need to do projection lithography after contact lithography in order to fabricate a 1 $\mu$ m sized samples, we partially develop the contact pattern for 2-3 seconds, just long enough so that a faint image of the pattern is barely visible with an optical microscope. After the projection lithography exposure, the photoresist is fully developed for 45 seconds. This procedure will be discussed in detail in the following section on projection photolithography.

## 2.2.3 Projection lithography

With an acetate negative film used as a mask, contact lithography can only produce  $10\mu m$  wide lines due to the edge definition of the pattern on the negative film. Over the past decades, scientists have developed and refined projection lithography to reduce the dimensions in microlithography. Using an inorganic resist/polymer bilayer scheme, Tai et al. have successfully fabricated 0.5  $\mu m$  lines with a Perkin-Elmer projection printer photolithography system.<sup>10</sup> Another research group produced 0.2  $\mu m$  metal patterns using projection photolithography.<sup>11</sup> In the latter experiment, the substrate was transparent to light, the mask was projected

through an oil-immersion objective lens and the substrate onto a single photoresist layer. With this technique, the under-cut profile is obtained naturally.

The optical path in our commercial optical microscope<sup>12</sup> is shown in Fig. 2.4. The mask is an acetate negative held in a rigid frame. With a yellow filter, a conventional W lamp can be used as both a visible light source for mask alignment and as a UV source for exposure. The UV tail from a conventional W lamp is intense enough to expose photoresist if the light is focused down to a small enough area. There are three objective lenses in our microscope: 10X, 40X and 100X; the corresponding fields of view are 1800  $\mu$ m, 450  $\mu$ m and 180  $\mu$ m. For the 100X lens, the proper exposure time is 35~40 seconds when the voltage on the lamp power supply is set to 6V and aperture position is at its minimum position. For the 40X lens, the exposure time is 200s for the same mask with the same illuminating power. For the 10X lens, the pattern does not show up in photoresist in a reasonable time, because the UV intensity is not high enough to fully break the polymer's chemical bonds. All projection lithography in this work was done using the 100X objective lens.

If the film mask has a 100  $\mu$ m wide line feature, and a 100X lens is used for projection, then a fine line of  $100\mu$ m/<sub>100</sub> = 1 $\mu$ m is obtained in the resist. Usually this size is near the resolution limit of projection lithography set by the wavelength of the UV tail in the spectrum of a tungsten lamp. We routinely achieve 1 $\mu$ m resolution, a resolution which is perhaps limited by the thickness of the photoresist layer. We have found that the resolution of projection lithography is sensitive to the reflectivity of the substrate. The photoresist profile of the pattern on some transparent substrates, such as glass or quartz coated with Fe<sub>2</sub>O<sub>3</sub>, are well-controlled with sharp edge definition and good under-cut profile (shown in Fig. 2.3). However, the linewidth is slightly larger if



Figure 2.4 Optical path for projection lithography: (A) tungsten lamp, (B) collimating lens, (C) mask, (D) filter, (E) collimating lens, (F) objective lens, (G) prism, (H) eyepiece.

the substrate is a metal film with a high reflection coefficient. Just as in contact lithography, the pattern profile turns out better when the side of the negative coated with silver grains faces toward the substrate. Most of our samples are fabricated on oxidized silicon substrates. In the resistance image study [Sec. 3.3], the substrate is a strongly reflecting 2000 Å Al film. This substrate yields a good under-cut profile in 100X projection lithography, resulting in clean lift-off after metal film thermal evaporation.

In contrast to projection lithography, with contact photolithography, one cannot easily obtain the under-cut photoresist profile which is crucial to reliable sample fabrication. Intense research over the years has led to several recipes to achieve good undercuts with contact lithography. The most popular technique is the triplelayer method<sup>13</sup> which uses two photoresist layers and one thin metal film layer separating the two resist layers. In this technique, two UV exposures are required. The bottom resist layer is exposed to UV without a mask (first exposure) and then several hundred Å of Al or Cr is thermally evaporated onto it. A Cr film is preferred because photoresist developer  $452^2$ , which is a potassium hydroxide solution(KOH), etches Al. A thin top layer of photoresist is spun on following the metal thermal evaporation. Then the mask pattern is transferred to the top photoresist layer (second exposure), which then developed. The opaque metal film protects the bottom resist layer during this exposure. The Al / Cr film is then etched away and the bottom layer of photoresist which was previously exposed is developed to form an under-cut profile in this triple-layer system. The advantage of this method is that the under-cut profile can be well controlled by the length of the second development; the disadvantages are that it takes more steps than the single layer resist method and can leave more residues on the substrate surface after lift-off.

Instead of using the triple-layer method, we used a single photoresist layer and a "chlorobenzene soak" of the photoresist to achieve the under-cut on profile in contact lithography.<sup>12</sup> After UV exposure of the photoresist-covered substrate, the substrate is soaked for eight minutes in chlorobenzene, which hardens the positive photoresist which is not exposed to UV source, but has little effect on the exposed photoresist area. This hardening is proportional to the time which substrate is left in air after soaking and before its development. If it is developed immediately after the chlorobenzene soaking, only several seconds in the developer produces a faint image of the pattern; by contrast, it takes as long as 40 seconds to get a faint image if it is kept in air over night after chlorobenzene soaking. After this partial development, the sample is processed by projection lithography. The procedure for projection lithography is the same as described previously, except that the exposure time is 45~50 seconds for the 100X objective lens, 50% longer than before, because the photoresist surface is harder after chlorobenzene soaking. The photoresist is then developed for 60~70 seconds. Now that the mask pattern has been transferred to the photoresist layer on the substrate, it is ready for the next process—metalization.

#### 2.3 Metalization—Thin Film Deposition

#### 2.3.1 Substrate cleaning—Reactive Ion Etching(RIE)

In thin metal film deposition, one is often concerned about the adhesion of the evaporated metal film to the substrate. Usually metal films cannot form a strong chemical bond with the substrate due to interfacial contamination from residual photoresist. One solution is to install an ultra-high vacuum electron-beam evaporator with *in-situ* plasma cleaning facilities. However, in our lab, a thermal evaporator operating with a simple diffusion pump is used. Prior to the metal deposition the substrate is cleaned of photoresist residue in a reactive ion etching(RIE) system<sup>14</sup> for 30 seconds with O<sub>2</sub> base pressure = 50 mtorr and power = 19 watts. The working principle of RIE is that O<sub>2</sub> forms a reactive plasma and chemically reacts with residues on the substrate. There is no evidence that this short time RIE cleaning affects the photoresist profile. Within 10 minutes of RIE plasma cleaning, the substrate is put into the evaporation chamber. This cleaning process is crucial to the adhesion between the submicron-sized samples and substrates.

In order to further improve adhesion between Al or Au and a glass-like substrate, we find it necessary to put a thin Cr layer between the sample and the substrate. The Cr layer is usually about  $5 \sim 8$  nm for samples prepared in our thermal evaporation system, and this has a negligible effect on the electron transport properties measurement because it is much thinner than the sample thickness of 200 ~ 300 nm

#### 2.3.2 Metal Thermal Evaporation

In our lab, metalization for sample fabrication is done in a diffusion pumped vacuum system with a  $LN_2$  coldtrap. The basic structure is shown in Fig. 2.5. This thermal evaporator has one rotating stage with four source boat positions, so we can evaporate up to four different materials on the same substrate without breaking vacuum. In this way, we can fabricate samples while keeping their interfaces as clean as possible. With the coldtrap filled with liquid nitrogen, the base pressure is  $1 \times 10^{-7}$  torr. Usually the pressure rises to  $2 \times 10^{-6}$  torr during evaporation of metal films. Tungsten boats are used to evaporate Al and Au. A calibrated crystal thickness monitor, cooled by water, measures film thickness during deposition. A shutter is carefully positioned between the source boat and the sample so that the crystal monitor will never be blocked. Depending on the location of the monitor, the sample film thickness may differ from the thickness of the film on the monitor by a "tooling factor". The tooling factor is determined by comparing the reading of the crystal monitor and the thickness of a test sample, measured by a Dektak II surface profile. The deposition rate is about 1 nm/sec for Al and 0.1 nm/sec for Au or Cr. It is believed that fast deposition will produce purer metal films because less contamination can be incorporated into the film. However, fast deposition requires a higher boat temperature which raises the pressure during evaporation. The Cr source is a Cr-impregnated W-rod<sup>15</sup> so no boat is needed for the Cr evaporation.

During the deposition of Au, the substrate's temperature rises to about 100°C due to radiation heating from the hot boat without a Cu heatsink. Such a temperature change is not a concern in sample fabrication because STM and AFM's images indicate that metal film's morphology is not dramatically affected by it.<sup>16</sup>



Figure 2.5 (Top) Photograph of the thermal evaporator, Coating System E306A, made by Edwards High Vacuum International, Edwards, England. (Bottom) Schematic block diagram of the thermal evaporator.

#### 2.3.3 Thermal Deposition of Silicon Monoxide

In fabricating samples for electron beam resistance imaging, an insulating layer has to be laid down just underneath the metal sample, and this is done by thermal evaporation in the same vacuum chamber as the subsequent metal evaporation. Silicon dioxide is used as an insulating layer by thermally evaporating silicon monoxide in an  $O_2$  environment at a pressure of  $1 \times 10^{-4}$  torr.

A tungsten boat is used to evaporate silicon monoxide powder. Because silicon monoxide is an insulator with very poor thermal conductivity, radiation from the boat rather than conduction heats the silicon monoxide. This means that the tungsten boat is very hot and is an intense radiation source during the deposition. It is necessary to surround the boat with thermal shielding made of 0.01 inch thick stainless steel shim stock in order to keep the vacuum chamber cool. Also, a one inch diameter, half inch thick copper cylinder is placed on the back of the substrate as a heat sink to keep the substrate cool. Otherwise, the photoresist will crack due to overheating during silicon monoxide deposition. The deposition is carried out in an  $O_2$  atmosphere with the pressure just above  $1 \times 10^{-4}$  torr. Before admitting the O<sub>2</sub>, the silicon monoxide outgased for several minutes with the shutter covering the sample. The vacuum valve to the diffusion pump is then partially closed and O<sub>2</sub> gas is admitted through a needle valve. The pressure in the chamber is maintained at  $1 \times 10^{-4}$  torr by adjusting the value to the diffusion pump. The deposition rate is very slow, less than 0.1 nm/sec because of the difficulty in heating the silicon monoxide. As silicon monoxide vapor condenses on a cool substrate in an oxygen environment, it becomes silicon dioxide. As a result, this passive layer is 700~1000 Å thick SiO<sub>x</sub> with  $1 < x \le 2$ , which serves well as an

insulating layer with a resistance larger than 20 M $\Omega$  because the resistance of the sample is about 20  $\Omega$ .

It is dangerous to use a diffusion pump to pump  $O_2$  gas because the hot silicon or carbon-based oil can react explosively with the  $O_2$ . To be safe, one should use a PFPE type diffusion pump oil (a fluorine-based oil) which is inert to oxygen gas.

#### 2.4 Electron Beam Lithography by SEM

## 2.4.1 Comparison of E-beam Lithography and Photolithography

The principle of electron beam lithography is similar to that of contact photolithography schematically shown in Fig. 2.1 except that electron beam exposure is used instead of UV exposure and the resist layer is e-beam sensitive rather than photon sensitive. A UV photon is neutral and has kinetic energy of only a few eV, so its interaction with a polymer is relatively simple. However, an electron is charged and is accelerated to typically 30 keV in a SEM, so its interaction with an e-beam resist, such as polymethylmethacrylate (PMMA) is very complex, involving the production of X-rays, and backscattered and secondary electrons, as shown in Fig. 2.5. These differences between e-beam and photolithography determine their advantages and disadvantages, as well as their resolution and applications.

**Cost:** An electron with an energy of a several keV has a mean free path in air of a few microns. Therefore, a vacuum chamber is needed for the electron beam in a SEM. Also, an electron beam in a SEM is focused with an expensive magnetic lenses. In contrast, a UV photon can travel in air for long distances. An uv light rays are relatively easily focused and collimated with quartz or glass lenses. As a result, electron beam lithography often costs more than photolithography.
**Resolution:** The resolution of electron beam lithography is limited by the beam spot size of about 3 nm-20 nm and by the effective interaction volume in the sample, which depends on the substrate properties. If a substrate is electron-beam opaque, there will be large flux of the backscattered electrons and X-rays, producing an exposed region of resist about 100 nm diameter for a 30 kV electron beam. In order to achieve the minimum exposure spot size in e-beam lithography, a resist layer has to be thinner than 100 nm and the substrate has to be electron-beam transparent in order to reduce the back-scattering electron effect as much as possible. Photolithography's resolution is usually limited by the wavelength of UV photons to about  $0.5 \mu m$ .

Obviously, the lift-off technique requires that resist layer is thicker than the metal film. In photolithography, the resist layer is usually 1 $\mu$ m thick so that the minimum linewidth obtained with single layer resist technique is about 1 $\mu$ m. In electron beam lithography, the effective interaction region (~ 100 nm) is much greater than the spot size of electron beam (~ 6 nm), hence, the minimum linewidth obtained with single thick layer resist technique is about 100 nm. (Many people get 30 nm lines using thin layer resist with dry etching.) In addition, the effective interaction region is a droplet-like shape and naturally produces an undercut profile in electron beam lithography even using single resist layer method.

**Throughput:** E-beam lithography is a serial process while contact lithography is a parallel process. Electron beam lithography writes directly on resist without a mask. The disadvantage is that it takes a long time to write a large area with complex patterns; also, e-beam lithography requires pattern alignment of  $0.1\mu m$  for multipattern exposures. These are the challenges facing electron beam lithography specialists in the integrated circuit industry. Fortunately, in this thesis work, it is relatively easy to fabricate the samples needed because of the simple patterns used.



Figure 2.6 Schematic diagram showing the region of interaction between a high energy electron beam and 3000 Å thick e-beam resist layer, polymethylmethacrylate (PMMA). Such a droplet-shaped interaction volume produces a natural undercut profile for electron beam lithography if the e-beam resist layer is thin.

#### **2.4.2 Electron-Beam Exposure**

In our lab, electron beam exposure is done with a conventional scanning electron microscope (ISI model SX-40). Initially, a ramp generator was used to control the electron beam scanning range and speed for writing a single line. Later, a computer with a D/A board was used to control the x-y scanning coils and the electron beam blanker during writing of more complex pattern.

The production of sub-µm features by e-beam lithography requires careful attention to details. First, a well focused beam with small astigmatism is crucial for the production of a sub-micron line. Focusing of the electron beam is facilitated by a droplet of silver paint placed very close to sample writing area, which provides a focusing target. Second, the correct exposure dose of electrons is important. For the 496K, 4% polymethyl-methacrylate (PMMA) used in this work, the total charge deposited per unit area must be  $1.6 \times 10^{-4}$  C/cm<sup>2</sup>. The dose is determined by the primary electron beam current, the beam diameter, and the exposure time which is controlled by the appropriate writing speed. A third important factor is the SEM's accelerating voltage, which on our machine is adjustable from 1kV to 30 kV. Usually the maximum accelerating voltage is chosen because a higher accelerating voltage will give smaller beam spot size and better resolution. In addition, the sample stage should be clamped to the chamber wall in order to reduce vibration and drift caused by.

#### 2.5 Some Samples Fabricated

As discussed so far in this chapter, there are many techniques available for sample fabrication. The main concern is how to choose one technique or another, and how to wisely combine several techniques together. Here I present several recipes for samples fabricated for different purposes in this thesis.

## 2.5.1 Al/SiO<sub>2</sub>/Al and Au/SiO<sub>2</sub>/Al Samples for Resistance Imaging

A sample for the resistance imaging experiment with a scanning electron microscope is shown in Figs. 2.7 and 2.8. A 1 $\mu$ m wide metal line made with projection lithography is separated from an Al substrate by a 0.1  $\mu$ m thick thermally evaporated SiO<sub>2</sub> layer. The Al substrate consists of a 0.5 mm wide, 2500 Å thick Al stripe, which is embedded in a 0.8  $\mu$ m thick silicon dioxide layer of a silicon substrate. Projection lithography and two stages of contact photolithography are used in the sample fabrication.

The process begins with dry oxidation of Si substrates. Substrates are cut from a 3" diameter (100) p-type polished silicon wafer with a resistivity of 10  $\Omega$ -cm.<sup>17</sup> Using a diamond cutter, the wafer is cut into 0.5" by 0.5" square pieces. These are washed in an ultrasonic cleaner with acetone, methanol and de-ionized water in order, followed by drying with nitrogen gas.

Prior to lithography, a thick native dioxide layer is grown on the silicon substrates by thermal oxidation in an oxygen environment at high temperature. The Si substrates are placed in a quartz tube through which oxygen from an O tank flows. The flow rate of O gas is monitored by bubbling the exhaust through a beaker of water, and the flow rate is set at about 5 standard cubic centimeters per second (sccm/sec.), which corresponds to several bubbles per second in the water. The quartz tube and substrates are heated in a tube furnace at 1100°C for 17 hours while oxygen flows continuously. This procedure produces a  $0.7 \sim 0.8 \ \mu m$  thick silicon dioxide layer which appears greenish under white light. The oxidation time can be shortened to 3 hours by a wet oxidation process, in which oxygen gas flows through 95°C hot water before flowing over the substrate. The silicon dioxide layer grows faster during wet oxidation than during dry oxidation because water molecules can diffuse through silicon dioxide layer more easily than oxygen gas at

the same temperature. In industrial processes, both wet oxidation and dry oxidation are used to grow high quality passive oxidation layers.

After oxidation, a patterned trench is etched in the SiO and filled with an Al film as shown in Fig. 2-9. The substrates are coated with Shipley S1811 (25% concentration) photoresist. The substrates are prebaked at 95°C for 30 minutes in air to drive the solvent out of the coating photoresist layer. Using an acetate film negative as a mask, a stripe line pattern is transferred to the photoresist with contact photolithography as shown in Fig. 2.2. After UV exposure, the photoresist is developed in Shipley Micro Developer 452 for 50 seconds. The substrate is then baked at 110°C for 30 minutes to harden the unexposed photoresist for further processing (the so-called "hardbake"). After cooling, the substrate is placed in a buffered HF solution for 3 minutes to partially etch away the silicon dioxide unprotected by photoresist. This chemical reaction is as follows:

$$SiO_2 + 4HF \implies SiF_4 + 2H_2O$$
 (2.1)

Buffered HF is made from one part 50% hydrofluoric acid and seven parts 50% NH<sub>3</sub>F; it etches SiO<sub>2</sub> at a rate of about 1000 Å per minute. As a result, 3000 Å SiO<sub>2</sub> are removed from a 7000 Å thick SiO<sub>2</sub> layer in three minutes. Only 3000 Å SiO<sub>2</sub> are removed because an Al film of that thickness will fill this trench. Chlorobenzene soaking of the photo-resist is not necessary here because isotropic etching with buffered HF indeed gives an undercut profile as shown in Fig. 2.9.

The resist-covered substrate is then coated with 2500 Å Al in a thermal evaporator. After lift-off, a 0.5 mm wide, 2500 Å thick Al stripe is embedded in the thicker silicon dioxide layer. This Al film forms the substrate for samples used in the resistance imaging experiment.

The reason for using such a metal stripe as a substrate will be explained in detail in Sec. 4.4.1. Briefly, it is because the high energy electron beam of a SEM interacts with silicon dioxide and generates electron-hole pairs. This pair



Figure 2.7 (a) An optical microscope photograph of a sample line under 400x magnification. The line width is  $1.6 \mu m$ . (b) A sample in the sample holder, with leads attached to the lithographically defined metal films on a  $1 \text{ cm}^2$  substrate.







Figure 2.8 Schematic diagram of the sample shown in Fig. 2.7 (b)

production generates a large substrate current, which ruins the resistance imaging measurements. A metal substrate eliminates such effects.

After the Al stripe is laid down in the SiO<sub>2</sub>, the contact photolithography procedure shown in Fig. 2.2 is followed. A 1 µm thick S1811 photoresist layer is spun on the Al stripe and substrate and is prebaked at 95°C for 30 minutes. An acetate negative mask with the contact pad pattern is then aligned on the photoresist, with the sample position over the 0.5 mm wide Al stripe. After a UV exposure, the substrate is soaked in chlorobenzene for 8 minutes to achieve an undercut profile. The substrate is baked at 75°C for 20 minutes to drive solvent out of photoresist layer instead of leaving it in air overnight. In doing so, the processing time is significantly shortened. After cooling, the contact pattern is partially developed in Shipley developer CD-30 for about 10 seconds until the contact pattern is barely visible. Then using projection lithography, a 2 µm wide and 160 µm long line is transferred to photoresist. As explained in sec. 2.2.3, the line width is larger than 1µm because the substrate is a highly reflecting Al film. After being fully developed for 60~70 seconds, the photoresist has an under-cut profile ideal for metalization. Prior to loading the substrate into the thermal evaporator, the substrate is cleaned by reactive ion etching in O<sub>2</sub> gas for 30 seconds with a power of 19 W. Then following the procedure for thermal evaporation of silicon dioxide and Cr/Al or Cr/Au described previously, 1000 Å SiO<sub>2</sub>, 50 Å Cr and 2500 Å Al or 2000 Å Au films are evaporated in sequence. (50 Å Cr is unneccessary for adhesion here since we are deporting Al or Au directly on fresh SiO<sub>2</sub>; it is for the consistence with the previous samples.) After lift-off, the sample shown in Figs. 2.7 and 2.8 is produced.



Figure 2.9 The procedure for laying down a metal stripe on a Si/SiO<sub>2</sub> substrate.

## 2.5.2 Samples for Squeezable Tunnel Junction

In order to study single electron tunneling effects, it is necessary to fabricate small tunnel junctions with small capacitance. To do this, we make a squeezable tunnel junction, which consists of two metal stripes on two different substrates, which are stacked over each other and separated by metal film spacers, forming a very small gap, adjustable from zero (contact) to  $0.5\mu$ m. The junction capacitance is related to the width of the metal stripes and their separation as follows:

$$C = \varepsilon_0 \cdot \frac{A}{d}$$
 (2.2)

Where  $\varepsilon_0$  is the dielectric constant, A is the surface area of the capacitor, approximately the square of the stripe line width, and d is the separation between the two metal surfaces. It is clear that A has to be small in order to reduce a junction's capacitance because d cannot be larger than 100Å for the purpose of electron tunneling through a vacuum barrier. Two metal stripes with micron and sub-micron sizes are shown in Fig. 2.9.

The substrates are 1 mm thick, optically flat, polished glass slides ordered from Esco Products, Oakridge, NJ. The micron sized sample is fabricated by contact lithography and projection lithography, using the procedures described in the previous sections. All samples were made with 1500 Å Au with a thin 50 Å Cr underlayer for improving adhesion. In the following section, I concentrate on the fabrication of sub-micron sized tunnel junctions using electron beam lithography.

As purchased, the original glass slides are one inch wide and three inches long. These are cut with a diamond saw into six sample substrates, each one inch long and 3/8 inch wide with precautions to avoid scratching the polished substrate surface. The substrates are cleaned in an ultrasonic cleaner with a detergent for glass, followed by cleaning with acetone and methanol. Next, the substrates



Figure 2.10 (a) An optical photograph of a micron-sized tunnel junction under 150x magnification; The junction is viewed through one of the transparent glass substrates. (b) a sub-micron sized bridge for a tunnel junction under 300x magnification, the sub-micron line at the center is 40  $\mu$ m long and 0.15  $\mu$ m wide.

are baked at 200°C for 30 minutes to remove water from their surfaces. Now they are ready for contact lithography and projection lithography to lie down contact pads for electron beam lithography samples.

Next, the glass substrate is covered with 1µm thick photo-resist layer (Shipley S1811), followed by a prebake at 95°C for 30 minutes to drive solvent out of photoresist polymer. An acetate negative is used as a mask in contact lithography. This mask defines two large contact pads leading to the junction area and four  $2 \times$ 2 mm<sup>2</sup> squares for the mechanical spacers. A chlorobenzene soak is used to harden the unexposed photoresist to get an under-cut profile for lift-off. After partial development of the resist for about 30 seconds, the contact pad area are visible. With projection lithography, a line 200 µm long and 2 µm wide, with a 40  $\mu$ m gap in its center, is registered with the contact pads and exposed. This 40  $\mu$ m gap is the writing field for e-beam lithography, and a sub-micron line written by ebeam lithography will fill this gap. After full development for 60 seconds, the substrate with its patterned resist is placed into the thermal evaporator for metalization. 50 Å Cr and 1500 Å Au are laid down as contact pads. After this metalization, the vacuum chamber is opened, and a mechanical mask made of stainless steel shim stock with four holes is placed against the substrate. The four holes match the positions of the four square mechanical spacers. A 0.4 µm Al film is evaporated and covers only the area of the spacers. After lift-off, the contact pads have 50 Å Cr and 1500 Å Au, while the four mechanical spacers have 50 Å Cr and 1500 Å Au plus 0.4  $\mu$ m Al film, much thicker than the contact pads.

Now, electron beam lithography is needed to place a sub-micron sized line between the two contact pads. For simplicity, a single layer resist method is used here.

After cleaning, the substrate with contact pads and mechanical spacers is spin coated with 300 nm polymethylmethacrylate (PMMA) at 4000 rpm for 60s. Then

it is prebaked at 170°C for 60 min. to drive the solvent out of the resist. Next, a 20 nm Al film is evaporated onto the PMMA to prevent charging during e-beam lithography. This step is required because the glass substrate is insulating. The thin Al film is nearly electron transparent and has little effect on SEM imaging of Au contact pads underneath the PMMA. Such a metal coating is not necessary if the substrate is a silicon wafer with silicon dioxide layer 1µm thick or less, because a 30 keV energy electron beam can penetrate through such an oxide into the Si substrate. A tiny droplet of silver paint is placed very close to the sample area for adjusting focus and astigmatism.

Now the substrate is loaded onto the SEM sample stage, which is clamped to the sample chamber's wall. After the pressure inside the chamber is below about  $10^{-5}$  torr, 30 kEV high voltage is turned on, and the SEM's filament warms up for one hour to stabilize the electron beam current before the sample writing. Final focusing the electron beam is done by imaging a filament of the silver paint droplet with the minimum beam spot size at high magnification, usually from 50 kX to 100 kX. This step is crucial for reliable electron beam lithography. The specimen current at minimum beam spot size is about 1.5 pA.

Sample alignment is achieved by imaging the two contact pads and the 40  $\mu$ m gap in the line connecting the pads. At a low magnification of 600X, the two contact leads are placed horizontally in the middle of the CRT viewing screen. Then at a magnification of 2100X, the ends of the two contact leads are visible on the two edges of the screen. This alignment procedure has to be done quickly in order to avoid an unwanted exposure of e-beam resist during imaging.

Now electron beam writing is carried out by switching the SEM to computer control. The computer contains the writing pattern(produced with HarvardGraphic and stored in a data file) and controls the electron beam position and its writing speed, i.e., its exposure time. A 40  $\mu$ m long. 0.2  $\mu$ m wide single line is drawn in



Figure 2.11 An atomic force microscope (ATM) image of an e-beam lithographic Au line on a glass substrate, imaged by Jeeseong Hwang with a NANOSCOPE-III.

9.3 seconds with the specimen current of 1.5 pA. The electron dose for this line is  $2x10^{-4}$  C/cm<sup>2</sup>, just over the critical exposure dose.

After electron beam exposure, the substrate is placed in 0.1 Mole NaOH solution to remove the 20 nm Al coating on the PMMA. Then the sample is developed for 65 seconds to get a clean profile of a sub-micron sized line. Before it is put into a evaporator, the substrate is cleaned in a reactive ion etching(RIE) system for 30 seconds. 5 nm Cr and 100 nm Au are thermally evaporated onto the substrate at a deposition rate of 1 Å per second. During lift-off, the tiny droplet of silver paint, as well as the PMMA resist, are removed by acetone. An AFM image of a sub-micron sample is shown in Fig. 2.11. This high resolution AFM image

shows that a high quality Au line with a smooth morphology and sharp edge definition is obtained by electron beam lithography and thermal evaporation.

### 2.5.3 Sample for Crack Junction

Among those samples fabricated lithographically, a sample used as a crack junction in the Coulomb blockade experiment is the simplest. The sample shown in Fig 2.13 is a single 30  $\mu$ m wide, 5 cm long line laid down by contact lithography. The substrate is cut from a 1.2 mm thick, 1" by 3/4" glass slide. There are two holes with a center to center separation of 3/16" which are drilled by 1/8" diameter ceramic tile drill. After a standard cleaning procedure, the pre-drilled substrate is coated with a 1  $\mu$ m photoresist layer. After a softbake at 95°C for 30 minutes, a pattern in an acetate negative mask is transferred to the photoresist by optical contact lithography. After development, the substrate is hardbaked at 115°C for 30 minutes, then loaded into the thermal evaporator for metalization. Again, 5 nm Cr film is used for improving the substrate adhesion before 45 nm Au film is deposited. After lift-off, a single 30  $\mu$ m wide, 5 cm long line stretches along the glass bridge between the two holes.

In the crack junction experiment, the metal line is broken in liquid nitrogen vapor by making a crack in the glass substrate perpendicular to the metal film line, connecting the two holes in the substrate; thus forming two electrodes for tunneling. As described in chapter 5, the crack is made by bending the substrate, and the broken metal line forms a tunnel junction whose gap can be adjusted by adjusting the stress on the substrate. A typical crack junction sample with a crack in the glass substrate is shown in Fig. 2.13. Because of the strong reflection from the metal film, the break in the metal line is invisible, even though the crack in the glass substrate is easily seen.



Figure 2.12 Schematic layout of a sample for a crack junction.



Figure 2.13 Optical microscope photograph of a broken Au line on a cracked glass substrate. The Au line is 30  $\mu$ m wide. Although the crack in the glass is clearly visible, the break in the Au line cannot been seen in this photo.

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# **Chapter 3**

## **E-beam Induced Contamination and Efforts to Stop It**

A sample surface bombarded by an energetic electron beam in a SEM quickly develops a contamination layer made up of polymerized hydrocarbons and other molecules from backstreaming diffusion pump oil and the chamber walls. This surface contamination is a dark, electrically conducting, tar-like substance which strongly absorbs secondary electrons and makes SEM images appear dark. In attempting to overcome this problem, we have tried several different techniques, including replacing the carbon-based diffusion pump oil with perfluoropolyethers (PFPE) fluids, adding a cold shroud around the sample, and installing a gas jet. Use of the fluorine-based pump oil and the cold shroud substantially reduced the rate of sample contamination, but did not eliminate the problem. Our results indicate that the gas jet eliminates the build-up of carbonaceous contamination, as evidenced by the appearance of SEM images and measurements of the secondary electron coefficient, but in place of the tar-like residue, the gas jet appears to produce a bright, non-conducting surface layer of unknown composition.

## 3.1 Contamination Problem and Decontamination Techniques

Hydrocarbon molecules on the sample surface in a scanning electron microscope (SEM) are polymerized under action of the electron-beam and form a dark, conducting contamination layer. The details of the chemistry of formation of such contamination remain unknown. Many researchers have made great efforts to eliminate such contamination using different strategies. Conru and Laberge report that contamination rates (determined by measuring the volume of contamination spots) decrease by a factor of 40 when perfluoropolyethers (PFPE) type diffusion pump oil replaces conventional silicon- and carbon-based diffusion pump oils.<sup>1</sup> Duerr and Ogilvie discovered a gas jet technique to efficiently remove light element contamination in an electron probe microanalyzer<sup>2</sup>. The review article by Miller gives a systematic description of different decontamination techniques attempted by a dozen different research groups.<sup>3</sup>

Contamination by polymerized hydrocarbon molecules is a problem in our resistance imaging experiment primarily because the contamination layer is conductive and shorts the sample to its metallic substrate when the sample is exposed to the electron beam. Here I describe some decontamination techniques used in our experiment and their effects on samples, in the order of their implementation.

### 3.2 **PFPE Type Diffusion Pump Oils**

The instrument used in this experiment is an International Scientific Instruments (ISI) Model SX-40 SEM, with a pumping system that consists of a mechanical roughing pump, a diffusion pump with a liquid nitrogen ( $LN_2$ ) coldtrap and a Zeolite baffle between the roughing pump and the diffusion pump to prevent backstreaming from the roughing pump.

The diffusion pump oil originally used in the diffusion pump is Santovac-5, a five-ring polyphenyl ether fluid, whose structure is shown in Fig. 3.1.<sup>4</sup> It is well known that such benzene ring fluids are polymerized under bombardment of the electron beam in a SEM, leaving a tar-like residue on the specimen surface. This hydrocarbon contamination layer has a strong secondary electron absorption coefficient so that it looks dark in SEM images obtained from the secondary

electron signal. Moreover, such a contamination layer is conducting and can short out electrically conducting components of a sample..



Figure 3.1 Five-ring phenyl ether structure of Santovac-5 diffusion pump oil.

Conru and Laberge have demonstrated the seriousness of this oil contamination problem in a SEM. They found that a cone of contamination, a 1.6  $\mu$ m high and 0.4  $\mu$ m wide, built up on a cleaved Si wafer after 5 minutes in the spot mode (beam stationary on sample) with an electron beam of 25 keV and a specimen current of only 5 pA.<sup>1</sup>

Diffusion pump oils are the main source of contamination of the specimen in a SEM chamber, because such oils contain long-chain hydrocarbon molecules. In order to reduce hydrocarbon contamination in a SEM, alternative diffusion pump oils have been sought to replace the benzene-based oils. Fortunately, such alternative pump oils have been found. They are perfluoroakyl-polyethers, completely fluorinated fluids, which are called PFPE (perfluoropolyethers) for short. A typical PFPE structure, shown in **Fig. 3.2**, consists of 20 to 30 repeating  $C_3F_6O$  groups.

$$F - (CF - CF_2 - O)_n - C_2F_5$$

$$\int_{CF_3}^{CF_3} CF_3$$

Figure 3.2 Fluorochemical Structure, Krytox perfluoroalkylpolyether.

Holland et al. found that PFPE type fluids used in pumps are not polymerized under an energetic electron or ion beam in high vacuum, thus avoiding specimen contamination.<sup>5</sup> The claim is that the fluorine-based polymers unzip under the electron beam and form volatile species that desorb and are pumped away. All other fluids smeared on a metal substrate and placed under electron bombardment were found to form an insoluble tar-like residue. Moreover, even under ionized gas conditions, PFPE is not polymerized by oxygen or other gases, with the exception of pure hydrogen.

Because of the convincing evidence of the elimination of oil contamination by the use of PFPE fluids in a SEM, we decided to replace Santovac-5 used in our diffusion pump by a PFPE oil, Krytox 1625, whose structure is shown in Fig. 3.2.

Replacing the diffusion pump oil in our SEM required that the system be thoroughly cleaned in order to eliminate all traces of the old oil. The entire pumping system and sample chamber of the SEM was dissembled, and the diffusion pump was cleaned by trichlorethylene (TCE), acetone, and methanol; all pipes, valves, and chamber walls were cleaned thoroughly with acetone and methanol. Finally, the new pump oil Krytox 1625 was added and the system was reassembled.

A Micromaze foreline trap, made by Kurt J. Lesker company, was installed for blocking the backstreaming of hydrocarbon molecules from the mechanical pump. The Micromaze trap uses a ceramic, highly porous material which is non-metallic, inorganic and inert, and has a surface area of 200 m<sup>2</sup>/g with internal pores of diameter 40-60 Å. The Micromaze trap must be heated under vacuum at monthly intervals to purge it of oil.

After the SEM was reassembled, a test of the pumping speed of the new pump fluid was carried out by monitoring the pressure vs. pumping time. It was found that the diffusion pump with the PFPE fluid works as efficiently as with Santovac5, with no significant decrease in its pumping speed. Then, a contamination test was done with a fresh sample of Al film thermally evaporated on a silicon substrate. After exposure to a 2 keV electron beam for a couple of minutes with the beam in spot mode and a beam current of 10 nA (conditions similar to those of the resistance imaging experiments) a dark spot appeared on the metal film. Thus, the replacement of the old type diffusion pump oil had not eliminated the contamination problem. A likely reason for such an unhappy outcome is that there are other sources of hydrocarbon contamination such as outgassing of plastic and rubber materials in the instrument such as "O"-rings, specimen mounting material and electrical insulation, etc.. Therefore, other techniques were needed to further reduce hydrocarbon contamination. A cold shroud around the sample was used to reduce the partial pressure of hydrocarbon molecules and improve the vacuum near the specimen in the SEM chamber.

### 3.3 Cold Shroud

Our SEM's base pressure is usually in the range of  $10^{-5} - 10^{-6}$  torr when the liquid nitrogen coldtrap on the diffusion pump is filled. At such a pressure, a specimen surface is struck by 1- 10 monolayers of contaminant molecules every second. To eliminate hydrocarbon contamination altogether, one could switch to an ultra-high vacuum system. However, because of the high cost and complexity of an ultra-high vacuum system, this solution is seldom feasible.

Fortunately, some alternative solutions are available. One of them is to install a liquid-nitrogen-cooled cold plate very close to the specimen surface. Such a cold surface acts as a cryo-pump and reduces the partial pressure of hydrocarbon and water molecules, but has no effect on light, non-polar molecules such as  $N_2$ ,  $O_2$ , and  $CH_4$ . Flavio and Garuli demonstrated a striking reduction in contamination with such a cryopump.<sup>6</sup> Their results showed that almost no contamination could be detected with their design of cold trap in their electron microprobe.

Fig. 3.3 and Fig. 3.4 show the cold shroud, which consists of an  $LN_2$ -filled copper tube and an attached copper plate. The large cold surfaces help to improve vacuum. The ends of the coiled copper tube are soldered to two double-walled stainless steel dewar tubes which are welded to the chamber wall. Liquid nitrogen is poured into one dewar tubes and N<sub>2</sub> gas is vented from the other.

A copper plate, located above the specimen stage, is attached to the copper pipe by a copper adapter to ensure high thermal conduction between them. The electron beam passes through a small hole in the plate. In order to maximize the cold plate's solid angle as seen by the sample stage, one side of the plate is bent down around the specimen stage, without significantly blocking the secondary electron detector.

When the cold shroud and the diffusion pump coldtrap are filled with liquid nitrogen, the base pressure of the chamber obtained at a penning gauge located away from the specimen stage is  $1.0 \times 10^{-6}$  torr. We believe that the local pressure near the specimen stage is lower than the reading at the penning gauge. Moreover, the partial pressure of water and hydrocarbon molecules is believed to decrease by orders of magnitude because the temperature of the cold shroud (- 150 °C) is well below their freezing point. It takes approximately 20 minutes for the cold plate to achieve thermal equilibrium after pouring liquid nitrogen into the copper pipe, and the copper pipe must be topped off with LN<sub>2</sub> every 30 minutes. Prior to opening the sample chamber to air, the copper pipe must be warmed by blowing dry nitrogen through for several minutes, in order avoid condensation of water on the pipe.

The efficiency of the cold shroud as a cryopump was measured by monitoring the pressure while all valves connecting the chamber to the external pumps were



Figure 3.3 Arrangement of the cold shroud in the sample chamber of the SEM



Figure 3.4 Photograph of the cold finger installed in the wall of the sample chamber.

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closed. Pressure vs. time is plotted in Fig. 3.5, which shows that the presence of the cold finger reduces the pressure by a factor of two.

After installation of the cold shroud, another test for contamination was carried out with a fresh sample of Al film, 2000Å thick, thermally evaporated on a silicon substrate. The result was that the cold shroud significantly reduced, but did not eliminate the contamination. So far, we had replaced the diffusion pump oil by PFPE fluid, installed a foreline trap, and installed a cold shroud very close to the specimen surface. All of these measures helped to reduce the hydrocarbon contamination rate, but not enough to carry out the resistance imaging experiment. It was necessary to search for other solutions.

#### 3.4 Decontamination Using an Gas Jet Technique

Decontamination by the use of a gas jet directed at the point of impact of the electron beam on the specimen has been developed by other researchers.<sup>7,8</sup> The gas used may be  $O_2$ ,  $N_2$ , air, or Ar, and must be at a pressure  $<10^{-4}$  torr to be compatible with SEM operation. Castaining and Descamp directed a fine stream of  $O_2$  and  $N_2$  gas at the point of electron beam bombardment, and showed that the carbon concentration on the surface, measured by X-ray spectroscopy, decreased sharply once the gas jet was on, and remained at a low level<sup>9</sup>. When the gas jet was turned off, the carbon intensity rose linearly with time, indicating the buildup of contamination on the sample surface.  $O_2$  gas was the most effective. Their results showed clearly that the gas jet not only prevents the buildup of contamination but also removes any previously deposited material. Borile and Garulli found that oxygen and air are effective in preventing contamination, but not argon or nitrogen.<sup>6</sup> Other work suggests that nitrogen, room air, as well as other inert gases are effective, 10,11 and that water vapor has a strong



Figure 3.5 A plot of pressure vs. time while the SEM chamber is shut off from the pumping system.

decontamination effect.<sup>12</sup>

We installed a gas jet, similar to Castaining's, in our SEM as shown in Fig. 3.6. The flow of gas is controlled by a needle valve, a NUPRO B2-JNA with a 0.094 inch orifice and a 3-degree taper on the needle. The valve connects to a 1/8-inch copper pipe by a Swagelok fitting. A 22-gauge stainless steel syringe tube was glued into the other end of the 1/8" copper tube with a vacuum compatible epoxy. The position of the fine SS tube can be adjusted by sliding the Cu tube horizontally through a quickconnect vacuum seal. The fine SS tube is bent so that the gas jet is pointed at the point of impact of the electron beam on the specimen. Fig.3.4 also shows the gas jet tube inside the SEM chamber.



Figure 3.6 Schematic diagram illustrates the gas-jet technique: (A) 1" diameter clamp connector; (B) 1/8" quickconnect.

Again, a decontamination test was carried out on a fresh Al film on a silicon substrate. The build-up of surface contamination was monitored by measuring the specimen current while the electron beam was focused on a stationary point on the sample (spot mode) or a rastered over a small patch of the sample. As explained in Sec. 4.3.1, the specimen current  $I_{spec}$ , the primary beam current  $I_B$ , the secondary electron current  $I_{SE}$ , and the backscattered electron current  $I_{BS}$  are related by  $I_B = I_{spec}+I_{SE}+I_{BS}$ . Measurement of  $I_B$  and  $I_{spec}$  thus allows one to calculate the sum

of SE and BS coefficients  $\delta + \eta = \frac{I_{SE}}{I_B} + \frac{I_{BS}}{I_B} = \frac{I_B - I_{spec}}{I_B}$ . The SE coefficient  $\delta$  is extremely sensitive to the level of surface contamination. In general, clean surfaces have a larger  $\delta$  (appear bright in a SE image) and contaminated surfaces have a smaller  $\delta$  (appear dark).

The procedure for using the gas jet is first to pump the SEM chamber down to a base pressure of  $1 \times 10^{-6}$  torr with the cold shroud and diffusion pump cold-trap filled with liquid N<sub>2</sub>. Then, O<sub>2</sub> or Ar gas is admitted through the needle valve, which is adjusted until the pressure rises to  $4 \times 10^{-4}$  torr. The sum  $\delta + \eta$  for Ar-gas jet and O<sub>2</sub>-gas jet with a 2 keV electron beam is plotted vs. time in Fig. 3.7. The data show that the ratio rises more rapidly with a O<sub>2</sub> jet than with a Ar jet, even though the final values are very close. With the gas jet on,  $\delta + \eta$  remains high and darkened regions on the sample surface do not occur. With no gas jet,  $\delta + \eta$  falls continuously as the surface contaminates and the SEM image becomes more and more dark. We believe, therefore, that both the O<sub>2</sub> and Ar gas jets prevent carbonaceous contamination. However, with the gas jet, the ratio  $\delta + \eta$  becomes too high, higher that previously reported for clean surfaces (for clean Al,  $\delta + \eta = 1.2$ ), indicating that some kind of surface layer is forming, but not a conducting carbonaceous one. The bright layer is transparent when viewed with optical microscopy, and our resistance imaging data (Chap.4) indicates that the bright



Figure 3.7  $\delta+\eta$  vs. time for an Al film with a 2 keV electron beam at a pressure of  $4 \times 10^{-4}$  torr: (a) O<sub>2</sub>-jet; (b) Ar-jet

layer is non-conducting. One might argue that  $Al_2O_3$  is forming in the highly reactive oxygen plasma formed by the  $O_2$  jet and the e-beam, but then it is hard to understand why a similar bright layer is formed with a non-reactive Ar jet.

An SEM image of decontamination spots formed under the  $O_2$  gas jet is shown in Fig. 3.8. In Fig. 3.8 (a), which was imaged with a 1keV beam, the bright stripe is an Al film and the darker background is the oxidized silicon substrate (0.8 µm thick SiO<sub>2</sub>). Because the secondary coefficient  $\delta$  of Al is higher than that of silicon for a 1 keV electron beam, the Al film is brighter than the substrate in this image. The two bright squares were made by a 2 keV electron beam in scanning mode with the  $O_2$  jet on. The dark dot on the Al film is a hole in the film revealing the SiO<sub>2</sub> substrate. We believe that the dark regions surrounding the bright squares are hydrocarbon contamination. We do not know the composition of the surface in the dark regions and in the bright squares because of our lack of in situ analytical capability, such as Auger spectroscopy or X-ray spectroscopy. Transporting the sample for Auger analysis would probably yield inconclusive results because of contamination during transportation. In Fig.3.8 (b), another bright square was produced by a 2 keV electron beam with O<sub>2</sub> jet on the surface of the Si substrate, and this bright square is also surrounded by a dark region. This image was taken with a 10 keV electron beam, an energy at which  $\delta_{A1}$  is very close to  $\delta_{Si}$ , so that the Al stripe no longer appears brighter than the substrate. However, the bright squares appear similar in the two SEM images, indicating that the bright areas are not simply clean surfaces.

A highly undesirable result of the  $O_2$  gas jet technique is that the average lifetime of the tungsten filament of the electron gun is reduced from 100 hours to 10 hours due to oxidation of the filament. For this reason, we used the  $O_2$  jet on only a few occasions and then switched to the Ar jet. One solution to this problem



Figure 3.8 SEM images of decontamination spots: (a) Al film on an oxidized Si substrate (image taken with a 1keV beam) (a) One more spot made on the  $SiO_2$  substrate (image taken with a 10keV beam).

would be to keep the filament in a separate high vacuum chamber, as is done in field-emission SEM's.

No clear microscopic picture exists of how the gas jet modifies the surface chemistry. Borile<sup>7</sup> used ion milling and Auger analysis to measure the depth profile of oxygen and carbon in an iron sample that had been imaged while under an O<sub>2</sub> gas jet. He observed an enhanced oxygen concentration down a depth of 45Å. Evidently, use of the O<sub>2</sub> gas jet impregnated the sample surface with oxygen. Borile's conclusions are consistent with the speculation that the bright areas on our Al samples are Al<sub>2</sub>O<sub>3</sub>, and yet there remains the puzzle of why bright areas form with the Ar gas jet. In any case, the bright surface layer is non-conducting, and therefore, should not significantly affect the results of our resistance imaging experiments.

In summary, we have attempted to eliminate carbonaceous contamination in our SEM with increasingly heroic measures. We replaced the diffusion pump oil by a PFPE fluid, then installed a liquid  $N_2$  cold shroud around the sample, and finally installed a gas jet. Use of PFPE pump oil and the cold shroud reduced, but did not eliminate, the contamination. The gas jet apparently eliminated carbonaceous contamination, but produced a "bright" non-conducting contamination of unknown composition.

References for chapter 3

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## Chapter 4

# **Resistance Imaging with a SEM**

#### 4.1 Abstract

A technique, similar to one described by Long and Slichter<sup>1</sup>, has been developed to measure electrical resistance variations along lithographic metal lines. In this technique, called electron beam resistance imaging (EBRI), an electron beam from a conventional scanning electron microscope is used to inject current into a metal line sample which has one end attached to ground through a current meter and the other end connected to a voltmeter. Current I(x) and voltage drop V(x), where x is the distance from the point of current injection to the end grounded, are recorded simultaneously while the electron beam is scanning along the thin metal film line. The resistance R(x) is calculated by  $R(x) = \frac{V(x)}{I(x)}$ . With this technique, we have measured R(x) vs. x along Au and Al lines 1  $\mu$ m wide, 150  $\mu$ m long, and we have achieved a spatial resolution of 0.5  $\mu$ m and a resistance resolution of 0.2  $\Omega$ . We expect that the technique can achieve 0.1  $\mu$ m and 0.05  $\Omega$ resolution. We have studied the onset of failure of metal lines by electromigration and local heating effects by imaging the resistance and then slowly increasing the external current to the point of failure, recording the evolution of the resistance at hot spots. In addition to the linear ohmic voltage from the specimen current, we observed an extra voltage which we suspect is generated by the bombardment of the electron beam on the interface between the metal and semiconductor substrate (the barrier electron voltaic effect) at some pinholes produced by electromigration in the samples.

### 4.2 Introduction

The steadily increasing density of transistors in integrated circuits has been achieved by reducing the size of each transistor and the metallic circuit lines which connect them. The reliability of those conductor lines determines the lifetime of the chip. In order to fabricate more reliable conductor lines, it is necessary to understand the mechanism of the failure of those conductor lines.

When there is a high current density through a conductor line, there is a strong electron wind force in the direction opposite to the current. This electron wind strongly interacts with the ions or defects in the metal lattices and drives those defects away from their original positions. This phenomena is called electromigration, and it is the primary mechanism for the failure of conductor lines under the stress of a high current density. Scanning electron microscopy has been used to study electromigration by imaging the evolving morphology of conductor lines under the stress of high current density which causes the formation of voids and hillocks in the lines.

Here, I describe a technique called electron beam resistance imaging (EBRI), which uses an electron beam from a SEM as a movable injection current source, allowing measurements of local sample resistance with sub-micron resolution. Using this technique, we have measured the local resistance of micron-sized lithographically fabricated metal lines before and after the stress of a high current density.

In this chapter, a review of the interaction between an electron beam and metal samples is presented. The principles of EBRI and the details of the electronic circuitry are presented. Finally, some data from Al samples are discussed at the end of the chapter.

#### 4.3 Principle and Analysis of Measuring Circuits

### 4.3.1 Electron Beam Interaction with Metals and Semiconductors

A large number of complex interactions occur when a focused electron beam penetrates a specimen surface. Among the signals produced are secondary electrons, backscattered electrons, characteristic and continuum x-rays, Auger electrons, and photons of various energies. These signals are obtained from a specific interaction volume within the sample, and this interaction volume strongly depends on the electron beam energy  $E_0$  and the atomic number of the specimen Z. In fact the resolution of an image from a scanning electron microscope is primarily determined by the excitation volume and not by the electron beam size.

Electrons having kinetic energies in the range 1-50 keV exhibit very complex behavior as they impinge on the surface of a solid sample. The energetic electrons undergo elastic scattering (change of direction with negligible energy loss) and inelastic scattering (energy loss with negligible change in direction). Elastic scattering is caused mainly by close collisions with the nuclei of atoms and in this case significant deviations from the incident direction occur. Inelastic scattering is caused by interaction with the atomic nuclei and with the bound electrons..

Inelastic scattering is primarily responsible for producing signals other than backscattered electrons. The incident primary electrons interact with the Coulomb field of the nuclei of the atoms and lose energy by emitting continuum x-ray radiation. Inelastic collisions also occur between the loosely bound outer electrons and the incoming electrons, and in this case loosely bound electrons are ejected. The ejected electrons have an energy typically less than 50 eV and are called secondary electrons. If these secondary electrons are produced close to the sample surface and their energy is greater than the surface barrier energy ( a sample's work
function of 2-6 eV), then these secondary electrons have a high probability of escaping from the surface. In contrast, those secondary electrons produced at depths much larger than 100 Å from the surface of the sample are likely to lose their energy by inelastic collisions before reaching the surface. In some materials, if the secondary electrons recombine with the holes formed during the scattering process, a photon is produced in the visible or near-infrared range. Inelastic collisions can also result in the production of characteristic x-rays and Auger electrons when the K, L, or M shell electrons are ejected during collision.

Elastic scattering by the Coulomb field of atomic nuclei is the most probable mechanism of large-angle scattering of primary electrons. The scattering consists of two parts: (a) Rutherford scattering, whereby a single scattering event results in a large change of the direction (greater than 90°), and (b) multiple scattering, composed of many small-angle scattering events. Multiple scattering may also result in a large change of direction of the primary electrons. After changing direction, the primary electrons may travel back to the surface and escape. This is the process of backscattering. The backscattered electrons leave with somewhat reduced energy due to inelastic processes. At some depth within the target, the original direction of the electron beam is lost and the electrons diffuse through the material at random. The position at which this occurs is the depth of complete diffusion  $x_d$ . Cosslett and Thomas have given a rather complete discussion of scattering theory and experiments in a series of papers.<sup>2</sup>

In a high atomic number sample, there is considerable (single and multiple) scattering close to the sample surface and a large fraction of the incoming electrons are backscattered. In the case of heavy elements, such as gold, diffusion sets in much nearer the surface than for a light element, such as aluminum. In fact, the penetration depth (diffusion range) of 10 keV electrons is about 0.1  $\mu$ m, for Au

and 1  $\mu$ m for Al. Fig. 4.1 is a diagram showing the relationship among accelerating voltages, density of specimen, and diffusion length of electrons.

When the primary electron beam impinges on the surface of a specimen, there exists a secondary electron current and a backscattered electron current, while some electrons absorbed by the specimen flow to ground forming the specimen current. The diagram shown in Fig. 4.2 illustrates quantitative relationship among them.

To further characterize these relationships, two ratios  $\delta$  and  $\eta$  are defined as follows:

$$\delta = \frac{I_{SE}}{I_B}$$
(4.1)  
$$\eta = \frac{I_{BS}}{I_B}$$
(4.2)

The secondary electron coefficient  $\delta$  depends strongly on specimen surface topology and on surface contamination as well as on the primary beam energy.  $\delta$  has been used to characterize specimen surface contamination processes in a SEM. In contrast, the backscattered electron coefficient  $\eta$  is almost independent of the primary electron energy, is not very sensitive to sample topology, and increases gradually with increasing specimen atomic numbers. For Al,  $\eta = 0.15$  while for Au,  $\eta = 0.5$ .<sup>3</sup> for 30 keV electrons.

It is convenient to use the sum  $\eta + \delta$  to monitor the buildup of surface contamination.  $\eta + \delta$  can be determined very easily by directly measuring both the primary beam current and the specimen current.

$$\delta + \eta = \frac{I_{SE} + I_{BS}}{I_B} = \frac{I_B - I_{SPECIMEN}}{I_B}$$
(4.3)



Figure 4.1 Relationship of the diffusion range of electrons and the electrons' accelerating voltage and specimen's density. A line drawn between the density of a specimen and the accelerating voltage of the electron beam intersects the diffusion axis, giving the diffusion length. [Reproduced from the JEOL electron microscope service manual.]



Figure 4.2 Schematic illustration of the relationship between a primary beam current  $I_B$ , specimen current  $I_{SPECIMEN}$ , backscattered electron current  $I_{BS}$  and secondary electron current  $I_{SE}$  as a high voltage electron beam impinges on the surface of the sample:  $I_B = I_{BS} + I_{SE} + I_{SPECIMEN}$ . The shaded area indicates interaction volume.

Variations in  $\eta + \delta$  are due almost entirely to variations in  $\delta$ , since  $\eta$  is nearly independent of beam energy and sample surface conditions. Generally, metal surfaces that are contaminated with hydrocarbons have a low  $\delta$  and appear dark in SEM images, while atomically clean metal surfaces are more bright.

# 4.3.2 Principle of Electron Beam Resistance Imaging Technique

The goal of this research is to develop a practical technique for measuring resistance variations along a thin metal film line fabricated by lithography, and to study the failure of metal lines by electromigration.

Fig. 4.3 illustrates the principle of electron beam resistance imaging. An electron beam from a scanning electron microscope is used to inject current at a point x in a metal line. One end of the metal line is grounded through a current meter, and the other end is attached to a volt-meter. The specimen current created by the electron beam cannot flow through the high-impedance voltmeter, and instead flows to ground through the low-impedance current meter. By measuring specimen current and the voltage drop independently, the resistance R of the specimen between the point of current injection and ground is determined by R = V/I. As the beam is scanned along the line, the resistance R(x) is mapped out.

Note that when  $\delta + \eta = 1$ , then from eqn. 4.3 the specimen current is zero. Consequently, the voltage drop is zero and the R cannot be measured. Since  $I_{spec} = I_B - I_{se} - I_{Bs}$ , the specimen current may be either positive or negative, depending on whether  $\eta + \delta$  is greater than or less than one.  $\eta + \delta > 1$  means that for every electron entering the sample from the primary beam more than one electron leaves the surface as a secondary or backscattered electron. In this case, one should not think of the beam as a current injector, it is really a current extractor.



Figure 4.3 Principle of electron beam resistance imaging (EBRI): (a) Measuring the resistance between point x and ground by injecting current with a SEM; (b) A plot of resistance vs. position; (c) A plot of dR/dx vs. position.

### 4.3.4 Experimental Apparatus

We devised two different ac lock-in techniques for electron beam resistance imaging. In the first technique, which was used most often, the electron beam is chopped and the sample resistance from the point of beam penetration to ground is measured. In the second technique, which was used only a few times, the electron beam is not chopped. Instead, the position of the beam is dithered, and the position derivative of resistance dR/dx is measured.

The apparatus for the beam chopping technique is shown in Fig. 4.4. A conventional scanning electron microscope (ISI Model SX-40) is used as a current source. In most of our measurements, a relatively low accelerating voltage of 2 kV was chosen both to reduce sample damage and to maximize specimen current (see Fig. 4.6).

An ac lock-in technique is used for maximum sensitivity . The dc electron beam is chopped by a beam blanker with a frequency of 510 Hz, a frequency chosen to optimize the noise figure of the lock-ins and to be well away from 60 Hz and its multiples. A single phase lock-in amplifier (Stanford Research SR 510) with current preamplifier is used to measure the specimen current from the sample to ground. A dual-phase lock-in amplifier (SR 560) with a low-noise differential voltage preamplifier is used to measure the voltage drop from the beam impact spot to ground. The low-noise voltage preamplifier (SR 512) has a gain of 100 and a noise figure of 0.5 dB with a source impedance of 100  $\Omega$  in frequency range of 100 Hz-1 kHz. The resistance of a typical sample used in our experiments is about 10  $\Omega$ . An IBM compatible 286-class personal computer is interfaced to both lockin's through GPIB interface cards, and calculates the resistance at every point according to



Fig. 4.4 Experimental apparatus for resistance imaging with a scanning electron microscope. The dark line is a GPIB interface between computer and lock-in amplifiers.

$$R(x) = \frac{V(x)}{I(x)} \tag{4.4}$$

The computer also controls the location (x) of the electron beam by supplying the voltage of the scanning coils through a D/A board.

Obtaining a good signal-to-noise ratio with this scheme is not easy. Under normal operating conditions, a well-tuned SEM has a small electron beam current of 5 pA to 100 pA and a small electron beam spot diameter of 60 Å to 100 Å. With a 10  $\Omega$  sample, this small beam current produces a voltage of less than 1 nV. In order to increase the beam current, we have disabled the 2nd condenser lens and removed the final aperture of our SEM. These measures increase the beam current to 50-100 nA but greatly degrade resolution, producing a spot size of about 0.1  $\mu$ m. The spot size verses beam current is shown in Fig. 4.5. The size of the beam was measured from the secondary electron signal profile as the beam was scanned across the sharp edge of a Faraday cup.

A collection ring biased at +300V to ground was fixed over the sample to collect secondary electrons and prevent their return to the sample and current detection circuitry.<sup>1</sup> Without a collection ring for capturing stray secondary electrons, these secondaries can reenter the specimen at positions away from the beam spot, changing the specimen current, and causing errors in the measurement of the resistance. Watanabe and Munakata have performed an experiment similar to ours, using the electron beam of a SEM as a current injector and measuring the voltage drop along a series of resistors used as a test sample.<sup>4</sup> Their results show that stray secondary electrons have a strong effect on the measurement of the resistance. They found that, with the sample carefully shielded from stray secondaries, the relative errors in the resistance measurements were less than 2%, while without shielding, the results were wildly inaccurate, often incorrect by more than an order of magnitude. Fig. 4.6 is a plot of  $\eta + \delta$  vs. accelerating



Figure 4.5 A plot of spot sizes (diameter) of electron beams vs. measured beam currents.



Figure 4.6 A plot of the sum of backscattered and secondary coefficients  $\eta + \delta$  vs. the beam accelerating voltage for an Al film with and without secondary collection ring.

voltage for an Al film both with and without voltage applied to the collection ring, clearly showing the effects of stray secondaries.

Fig. 4.7 is the circuit we used to measure the position derivative of resistance. The primary beam is not chopped by the beam blanker. Instead, the beam's position along the sample is modulated with a small sinusoidal deflection voltage. As long as the sample current is constant, the voltage measured by the lockin is proportional to the position derivative of resistance.

The position x of the beam, controlled by voltages fed to the SEM scan coils, is given by

$$\mathbf{x} = \mathbf{x}_{0} + \Delta \mathbf{x} \sin(\omega t + \varphi) \tag{4.4.1}$$

where  $x_0$  is the mean position of the beam controlled by a large (0-5V) voltage from the computer's D/A converter and  $\Delta x$  is the amplitude of a small ac modulation generated by coupling a small ac voltage through a transformer to the scan coils of the SEM. The voltage drop along the sample is then given by

$$V(x) = V_{o} + \frac{dV}{dx} \cdot \Delta x + \frac{1}{2} \cdot \frac{d^{2}V}{dx^{2}} \cdot (\Delta x)^{2} + \cdots$$
$$= I \cdot R_{o} + \left(I \cdot \frac{dR}{dx} \cdot \Delta x\right) \sin(\omega t + \phi) + \left(\frac{1}{2} \cdot I \cdot \frac{d^{2}R}{dx^{2}} \cdot \Delta x^{2}\right) \cdot \frac{1 - \cos(2 \cdot (\omega t + \phi))}{2} + \cdots$$

where we have assumed that the specimen current I does not vary rapidly with position x, an assumption which is only valid if the sample has a relatively smooth topology and slow variations with position of the secondary electron coefficient  $\delta$ . The lock-in measures the voltage at frequency  $\omega$  which has amplitude  $I \cdot \frac{dR}{dx} \cdot \Delta x$ . The dc specimen current I is measured with a dc picoammeter, and the amplitude of the beam dither  $\Delta x$  can be computed from the known characteristics of the



Figure 4.7 Experimental apparatus for measuring the position derivative of resistance vs. position. (A) Low-noise voltage preamplifier; (B) DC current amplifier.

SEM's scan coils. Both current and voltage are recorded by the computer, which computes the derivative of resistance dR/dx as the beam is scanned along the sample, point by point, under computer control. Typically, for a 10 $\Omega$  sample and a beam current of 50 nA, the position dither  $\Delta x$  was 10 $\mu$ m and the  $\omega$ -voltage was 33 nV.

## 4.4 Experimental Results

# 4.4.1 Sample Preparation and Sample Holder

Our final samples consist of Al or Au lines 2000 Å thick, 1.9  $\mu$ m wide and 160  $\mu$ m long separated from an Al metal substrate by a 700 Å thick thermallyevaporated silicon dioxide(See Fig. 2.8). The SiO<sub>2</sub> layer exists only under the metal and is not exposed directly to the electron beam. The Al substrate is a thick Al film thermally evaporated onto a polished silicon wafer. The detailed procedure for the fabrication of these samples was presented in the section 2.5.2.

Originally, we tried using samples consisting of metal lines separated from a Si substrate by a thermally grown 7000 Å thick silicon dioxide layer which covers the Si wafer everywhere. However, we encountered two serious problems with these samples. The thick silicon dioxide layer charged up under the electron beam, producing a large local electric field which distorted the focused beam and the SEM images. An even more serious problem was caused by interaction of the electron beam with the silicon substrate. When a high energy electron beam strikes a semiconductor such as Si, copious electron-hole pairs are generated. Only a few eV are required to make a single electron-hole pair in Si, so a high energy electron of several keV energy can create thousands of electron-hole pairs. Many of these carriers recombine after diffusing several micrometers (the diffusion distance in Si). However, those e-h pairs which are near the Si interface

are pulled apart by the strong electric field in the depletion layer and produce a spurious specimen current that may be many times larger than the beam current. We found specimen currents as large as 50 times greater than the beam current, and current multiplications of more than 1000 are reported in the literature. These huge currents were very noisy and always overloaded our lockin amplifiers. To solve this problem, we replaced the semiconducting substrate with a metal one, eliminating both the production of electron-hole pairs and the charging problem.

The sample holder consists of a 0.5" diameter aluminum cylinder surrounded by a tight fitting Teflon ring, as shown in Fig. 4.8. Electrical leads to the sample are held by eight SS syringe tubes (gauge #22) which fit snugly in vertical holes in the Teflon ring. Electrical leads from the SEM's electrical feedthrough are soldered to the SS tubes. Each SS tube contains a fine brass wire bent to form a simple clip which presses against the sample, making good electrical contact with both the metal film and the SS inner tube. This simple scheme allows rapid changing of samples with minimum damage to the sample's contact pads.





Side View

Figure 4.8 Sample holder.

#### 4.4.2 Result and Discussion

Fig. 4.9 is a plot of sample current and substrate current as the electron beam was scanned along the substrate, perpendicular to the sample line, crossing back and forth once over the sample line. The sample is an Al line is 1.9  $\mu$ m wide, separated from an Al substrate by a 700 Å thick SiO<sub>2</sub> layer. The 2 kV electron beam was chopped at 507 Hz and the average beam current was 51 nA. (The beam current was 113 nA with the AC modulation off.) A new data points was taken every 3 seconds and the lock-in's time constant was 300 ms. The secondary electron collection ring was biased at +300 V, and the O<sub>2</sub> gas jet was on with a pressure of 4x10<sup>-4</sup> torr for decontamination, as described in the section 3.3. In considering these data, it is well to recall that the specimen current is not, in general, equal to the beam current.

When electron beam is far from the sample, the sample current is negligible, compared with the substrate current, indicating that the silicon dioxide layer effectively insulates the sample from the substrate. However, when the beam is at the center of the sample, the substrate current does not decrease to zero although the sample current is at a maximum. Also, the current profile is very wide, much wider than the sample. A likely explanation of this is that the effective spot size is very large, more that 5  $\mu$ m. Apparently, the ac modulation of the beam blanker greatly increases the spot size. It is also possible that the SiO<sub>2</sub> layer becomes somewhat conductive when it is under bombardment by the electron beam, due to electron-hole production. Although the mean penetration depth of 2 kV electron in Al is about 0.1  $\mu$ m, shorter than the sample's thickness of 0.25  $\mu$ m, nevertheless, a portion of the beam current will reach the SiO<sub>2</sub> layer and may open a conducting channel.



Figure 4.9 A profile of current vs. position when a large current beam crossing the sample line.

Fig.4.10 is a plot of resistance to ground vs. position along a Al sample line, measured using the circuit in Fig. 4.5. The average sample current was 35.6 nA with the beam chopped at 507 Hz. The O<sub>2</sub> gas jet and +300 V collection ring were on, as usual. The data were taken while the beam swept forward and back once along the sample line, over a 5 minute period, which is about 3s for each data point.. The time constant for the lock-in's was 300 ms. The plot is nearly linear, as expected from Ohmic law. The two traces agree to within about 0.1  $\Omega$  This plot indicates that the total resistance of the sample is 10.1  $\Omega$ , which agrees well with a 2-terminal ohmmeter measurement of the resistance, which yielded 11  $\Omega$ and includes lead resistance.

In an effort to observe electromigration effects, the sample was then stressed with an external current of 1 mA for 24 hours while under vacuum in the SEM chamber at  $1 \times 10^{-5}$  torr. The current density was J =  $2.5 \times 10^{5}$  A/cm<sup>2</sup>. After 24 hours, the external current was removed, and the resistance vs. position was measured again. The data are shown in Fig. 4.11 and were taken with the gas jet off. (A single sweep of the sample does not result in significant contamination. Only when repeated sweeps of the beam across the sample are performed is the contamination severe and the gas jet necessary.)

The data of Fig.4.11 show very puzzling behavior. After the 24 hours,  $J=2.5\times10^5 \text{A/cm}^2$  stress, the computed resistance to ground vs. position is non-monatomic, showing two spots on the metal film line where R=V/I goes up and down. This behavior is inconsistent with  $R = \rho \frac{1}{A}$  and clearly shows that, in this sample, V/I is not equal to sample resistance. For this reason, we have relabeled the y-axis as V/I. The non-monotonic bumps reproduce well and, away from these bumps, the sample appears to show ohmic behavior with a total resistance of about 11.5  $\Omega$ , slightly greater than before the current stress.



Figure 4.10 A plot of resistance vs. position along the sample line.



Figure 4.11 (A) Resistance vs. position after J= $2.5 \times 10^5$ A/cm<sup>2</sup> for 24 hours, data taken without O<sub>2</sub> jet. (B) Resistance vs. position after 20 hour stress of J = 4.0  $\times 10^5$  A/cm<sup>2</sup> and 2 more hours of J = 2.5  $\times 10^6$  A/cm<sup>2</sup>, data taken with O<sub>2</sub> jet.

This same sample was then stressed with a current density of  $J = 4.0 \times 10^5$  A/cm<sup>2</sup> for 20 hours, followed by 2 hours of  $J = 2.5 \times 10^6$  A/cm<sup>2</sup>. After this treatment, the V/I of the sample was again mapped out, this time with the gas jet on. The data are plotted in Fig. 4.11, which shows a new huge anomalous peak appearing besides the previous two bumps. Obviously, this plot no longer indicates the resistance of the sample. We cannot explain this anomalous data entirely, however, we believe that the interaction between the energetic electrons and the SiO<sub>2</sub>/metal interface plays an important role here.

Recall that the resistance R = V/I is calculated from the measured voltage and sample current in this EBRI technique. The sample current profile for the most stressed sample of Fig. 4.11 is shown in Fig. 4.12. It is clear that the sample current is nearly constant with small fluctuations, except at the two ends of the sample line, where the larger contact pads increase the effective area for collecting electrons and cause a big jump in the sample current there. Since the sample current is almost constant, the anomalous resistance changes in Fig. 4.11 are primarily caused by the anomalous changes of the voltage signal.

Prior to current stressing, the Al sample line was smooth and of uniform width. After current stressing, the morphology of the Al line was rough and the line width was non-uniform as shown in Fig. 4.13. There were holes in the film (seen as dark areas to the right and left of center in Fig.4.13) whose positions corresponded to the locations of the anamolous voltage bumps in Fig.4.11. Clearly, much of the damage to the film was caused by electromigration during current stressing. However, some of the damage could have been caused by reaction with the oxygen plasma formed by the interaction of the O<sub>2</sub> gas jet and the electron beam. The holes in the Al film expose the SiO<sub>2</sub> underlayer. It is known that when a metal/semiconductor interface is exposed to an energetic electron beam, a voltage appears at the interface<sup>5</sup>. This phenomena, known as the barrier electron voltaic



Figure 4.12 Sample current vs. position.

effect, is probably the cause of the anamolous voltage. Note that the thickness of the Al film (250nm) is larger than the penetration depth of 2keV electrons in Al(150nm - see Fig.4.1) and so the Al/SiO<sub>2</sub> interface is not ordinarily exposed to the electron beam. Fig. 4.13 shows a SEM photo corresponding to the huge peak in Fig. 4.11.

Shown in Fig. 4.14, is a scan of the position derivative of resistance dR/dx of this same current stressed sample. The scan was made with the circuit shown in Fig.4.7, after the first current stress of  $2.5 \times 10^5$  A/cm<sup>2</sup> for 24 hours, but before the second larger current stress. Note the coincidence of the large voltage bumps in Figs.4.11 and 4.14.



Figure 4.13 SEM image of the void formed by electromigration corresponds to the huge peak in Fig. 4.11.



Figure 4.14 dR/dx vs. position.

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# Chapter 5

# **Squeezable Tunnel Junction and Crack Tunnel Junction**

A mechanically controlled squeezable tunnel junction and a crack tunnel junction, similar to Morland's "squeezable" and break junctions, have been built to probe single electron tunneling phenomena through a vacuum barrier. Both of these can work in the temperature range from 4.2 K to room temperature. In the squeezable and crack junctions, two electrodes are formed in two different ways: (i) by evaporating metal films onto two glass substrates in the squeezable tunnel junction; (ii) by breaking a narrow metal film by cracking a glass substrate in the crack junction. When the tunneling gap between the two electrodes, controlled by bending the glass substrates, is in the nanometer range, electrons can tunnel through the small gap. However, if the capacitance C of the junction is very small, single electron tunneling will be blocked by the Coulomb energy of  $e_{1/c}^{i}$  as long as the applied voltage is smaller than  $\frac{1}{2C}$  and the Coulomb energy  $\frac{e^2}{2C}$  is much larger than thermal energy kT. Such a Coulomb blockade effect is observed using a crack junction at liquid nitrogen temperature. Using a squeezable tunnel junction with micron-sized samples, two-level fluctuation of the conductance in the tunneling regime has been observed at liquid nitrogen temperatures.

# 5.1 Introduction

Since the experiment of electron tunneling through superconductor structures of  $Al-Al_2O_3$ -Al was done by Giaever in 1961,<sup>1</sup> it has been known that electrons can tunnel through an oxide barrier if two metal films are separated by a thin insulating layer, about 1~5 nm thick. Although it would be more interesting to study electron tunneling through an adjustable vacuum barrier instead of a fixed

thin oxide layer, this was not feasible until the scanning tunneling microscope (STM) was invented by Binnig, Rohrer et al.<sup>2</sup> in 1982. They implemented a negative feedback loop in the STM's electronic circuit to control the vacuum gap between the tunneling tip (one electrode) and the sample surface (the other electrode) as well as the tunneling current. In that feedback loop, the tunneling current from the tip to the sample is the feedback signal to control a high voltage output which either contracts or expands a piezoelectric material, thus controlling the vacuum gap between the tip and the sample surface, in turn, keeping the tunneling current constant as set.

The working principle of a scanning tunneling microscope is a quantum mechanical effect: if the gap between two electrodes is in the nanometer regime, wave functions of electrons in the two electrodes will overlap, and the electrons can tunnel through the vacuum gap. The probability of an electron tunneling depends exponentially on the vacuum barrier's height and its width, as well as electron's effective mass and charge. Because of such an exponential relationship between the tunneling current and the tunneling gap, a STM has an extremely high resolution and can often directly image atoms on a sample surface. It is one of the most effective instruments available to studies of surface science. Unfortunately, the tunneling tip and sample surface obviously have to be electrically-conducting materials, a serious limitation. In other words, a STM can only be used for observing surfaces of metal and semiconductor materials, not insulating materials, even not metals with a thick oxide layer. Fortunately, the atomic force microscope (AFM), developed after the STM, has no such limitation for observing material surfaces.

In addition, a STM can also serve as a spectroscopic tool if its feedback loop is turned off. For example, after an image is obtained the STM's tip is placed at an interesting position, at which a characteristic I-V curve can be recorded quickly while the feedback loop is turned off. An I-V curve contains information about electronic density of states (DOS) in the conduction band of the sample. The time for measuring an I-V characteristic has to be short because of the vibration of the STM body and the drifting of the tunneling tip while the feedback is turned off. Due to such a sensitivity to vibration, the STM has so far played a limited role as a spectroscopic tool for superconducting materials and normal metals. Therefore, it is desirable to develop a device with both an excellent stability and a mechanically-adjustable tunneling gap without the feedback loop in its electronic circuit.

Two years later after the invention of a STM, Morland and his coworkers designed a "squeezable" tunnel junction, which consists of two evaporated electrodes on two glass slides which are separated by four thin metal film spacers and pressed towards each other with an electro-magnetic squeezer until electrons can tunnel through the gap.<sup>3</sup> Later, They employed another scheme to form a clean tunneling interface by breaking Nb-Sn filaments in liquid helium.<sup>4</sup> Both the squeezable tunnel junction and the "break" junction have the extreme stability required for tunneling spectroscopy. Such a stability is achieved by sacrificing the transverse scanning capability. In this chapter, I describe a squeezable tunnel junction, similar to Morland's, with which both Coulomb blockade of single electron tunneling in a junction with an ultra-small capacitance and two level conductance fluctuation are observed.

### 5.2 Coulomb Blockade Theory

### **5.2.1 General Tunneling Theory**

In Quantum mechanics, it is well known that an electron can tunnel through a barrier with a finite probability, which depends on the barrier's height and width, as well as the electron's effective mass in the conduction band. For instance, when two electrodes are placed very close without contact, i.e., less than a few nm's, an electron in one electrode can tunnel through the vacuum gap and arrive at the opposite electrode if a small voltage is applied between the two electrodes, thus resulting in a current flowing through the vacuum gap between the two electrodes. This tunneling current is related to the applied voltage. Generally speaking, the relationship between the tunneling current and the bias voltage is determined by quantum mechanics and it is complicated. However, at a small bias voltage, the tunnel junction is nearly ohmic and can be characterized by a tunneling resistance  $R_t$ , without considering the effect of the junction's capacitance, which will be considered in the next section 5.2.2.

In Fig. 5.1, a schematic diagram shows the Fermi energy diagram in a basic tunneling process. The two electrodes can be two different metals with different work functions. The work function  $\varphi$  is defined as the energy required to remove one electron out of its conduction band to the vacuum. For most metals, the work function is in the range of 3-5 eV. When the two metals are placed very close, about 10 - 30 Å apart, electrons will tunnel between them, resulting in equalization of the Fermi levels of the two metals. [5.1(b)] With a bias V applied to the electrode on the left side, electrons on the right side electrode with the energy level higher than the opposite Fermi level can tunnel to the left, thus producing a current flowing in the circuit.



a) Two neutral metals separated by a large gap.



b) Two metals in equilibrium separated by a tunneling gap.



C) Tunnel junction with an applied bias voltage V.

Figure 5.1 a schematic diagram shows the Fermi energy diagram for a basic tunneling process.

By measuring the I-V characteristic, one can obtain information about the electron density of states (DOS) and the phonon spectrum. Usually, such experiments are done at low temperature because thermal energy smears out small non-linear features in the I-V characteristic. In the following, we are going to employ quantum mechanics to derive a quantitative expression for I-V in the simplest case: both electrodes are normal metal even at low temperature. Nevertheless, the result can be modified easily for a mixed structure with a superconducting electrode and a normal metal electrode. In cases involving superconducting electrodes, quasi-particle tunneling plays an important role. At low enough temperature, if both of the electrodes are superconducting and the tunneling gap t is smaller than about 20 Å, a Cooper pair can tunnel through the vacuum barrier, thus causing a supercurrent flow in a Josephson tunneling junction. For simplicity, this case will not be discussed here even though it is a very interesting research field now.

In Fig. 5.2, the electron density of states is plotted vs. the energy, and the Fermi levels are offset by an applied voltage V. We seek a quantitative expression



Figure 5.2 Energy diagram for a tunneling process, where a bias voltage V is applied to the left side electrode. Fermi level is set to zero as a reference point.

for the tunneling resistance in terms of physical quantities, such as the work function of the electrodes as well as the tunneling gap.

From "Fermi's golden rule number 2", the probability of an electron tunneling from the right side to the left side can be written:

$$w(t)_{r \to i} = (2\pi/\hbar) \Big| \langle L | H^{(1)} | R \rangle \Big|^2 \cdot N_i(\varepsilon)$$
(5.1)

where  $|\langle L|H^{(1)}|R\rangle|^{\epsilon}$  is the tunneling matrix element, denoted by  $|T_{rl}|^2$  and  $N_l(\epsilon)$  the density of states on the left.

Now, taking into account the Fermi distribution for T $\pm 0$ , the flow rate of electrons from the right to the left is  $(F_e)_{r \rightarrow l}$ .

$$(F_{e})_{r \to i} = \left(\frac{2\pi A}{\hbar}\right) \int_{-\infty}^{\infty} |T_{rl}|^{2} \cdot N_{i}(\varepsilon) \cdot N_{r}(\varepsilon - eV) \cdot f_{i}(\varepsilon - eV) \cdot [1 - f_{i}(\varepsilon)] d\varepsilon$$
(5.2)

where A is the cross section area of the junction,  $\varepsilon$  the electron's energy above the Fermi level ( $\varepsilon = E - E_F$ ), and f is the Fermi-Dirac distribution function

$$f(\varepsilon) = \frac{1}{(1 + \exp(\beta \varepsilon))}$$
(5.3)

with  $\beta = 1/kT$ .

On the other hand, the flow rate of electrons from the left to the right is  $(F_e)_{l \rightarrow r}$ :

$$(F_{e})_{l \to r} = \left(2\pi \frac{4}{\hbar}\right) \int_{-\infty}^{+\infty} \left[T_{lr}\right]^{2} \cdot N_{l}(\varepsilon) \cdot N_{r}(\varepsilon - eV) \cdot f_{l}(\varepsilon) \cdot \left[1 - f_{r}(\varepsilon - eV)\right] d\varepsilon$$
(5.4)

So the current— the net electrons — flowing from the right to the left is:

$$I = -e[(F_e)_{r \to i} - (F_e)_{i \to i}] = (2\pi eA_h) \int_{-\infty}^{\infty} T_i^2 \cdot N_i(\varepsilon) \cdot N_r(\varepsilon - eV) \cdot [f_i(\varepsilon) - f_r(\varepsilon - eV)]d\varepsilon \quad (5.5)$$

where it is assumed that  $|T_{Ir}|^2 = |T_{rI}|^2 = |T|^2$ .

When eV (< 50 mV) is relatively small compared with Fermi level (4 eV), we have, approximately,

$$[f_{I}(\varepsilon) - f_{I}(\varepsilon - eV)] = [f(\varepsilon) - f(\varepsilon - eV)] = \frac{\partial}{\partial \varepsilon} \Big|_{\varepsilon} \cdot (eV)$$

Also, when the temperature is very low,  $\beta$  is very large so

$$\frac{\partial f}{\partial \varepsilon} = \delta(\varepsilon)$$
 (5.6)

is a good approximation, where  $\delta(\varepsilon)$  is the well-known Dirac Delta function. Then eq.(5.5) can be further simplified as follows:

$$I = \left(2 \frac{\pi e A}{\hbar}\right) \int_{-\infty}^{+\infty} |T|^2 \cdot N_1(\varepsilon) \cdot N_r(\varepsilon - eV) \cdot \frac{\partial}{\partial \varepsilon} \Big|_{\varepsilon = 0} \cdot (eV) d\varepsilon$$

$$I = \left(2\pi e A_{h}\right) \cdot (eV) \int_{-\infty}^{+\infty} |T|^{2} \cdot N_{i}(\varepsilon) \cdot N_{r}(\varepsilon - eV) \cdot \delta(\varepsilon) d\varepsilon$$

Finally,

$$I = \left(2\pi e^2 A V / \hbar\right) \cdot \left|T\right|^2 \cdot N_1(0) \cdot N_r(-eV) \qquad (5.7)$$

It is natural to characterize tunneling junction by a tunneling resistance  $R_t$  defined as:<sup>5</sup>

$$R_{t} = \frac{V_{I}}{2\pi e^{2}A} \cdot |T|^{-2} \cdot [N_{t}(0) \cdot N_{r}(-eV)]^{-1}$$
 (5.8)

where the tunneling matrix element can be written specifically:<sup>6</sup>

$$|T|^{2} \approx \frac{16E(V-E)}{V^{2}} e^{-\frac{2}{\hbar}\sqrt{2m(V-E)}}$$
 (5.9)

where E and V are the Fermi energy  $E_F$  and the barrier hight of the metal respectively. The work function of the metal is defined as:

$$\phi = V - E_F$$

 $\left|T\right|^{2} \approx \frac{16E_{F}\phi}{\left(E_{F}+\phi\right)^{2}} e^{-\frac{2}{\hbar}\epsilon\sqrt{2m\phi}}$ 

So we have:

$$|T|^{-2} \approx \frac{(E_{F} + \phi)^{2}}{16E_{F}\phi} e^{\frac{2}{\hbar}\sqrt{2m\phi}}$$
 (5.10)

Replacing  $|T|^{-2}$  in eqn. (5.8) with eqn. (5.10), we get

$$R_t = R_0 \exp(A' \sqrt{\phi} d) \tag{5.11}$$

Where  $A' = 1.025 eV^{-1/2} \text{ Å}^{-1}$ , and

$$R_{o} = \left(\frac{\hbar(E_{F} + \phi)^{2}}{32E_{F}\phi\pi e^{2}A}\right) \cdot [N_{I}(0) \cdot N_{r}(-eV)]^{-1}$$
(5.12)

In terms of conductance, this can be written as

$$\frac{I}{V} = G_0 = R_0^{-1} \propto [N_1(0) \cdot N_r(-eV)]$$
 (5.13)

It is clear that the first derivative of tunneling current with respect to bias voltage is proportional to the electron density of states in the conduction band. Furthermore, it can be shown that second derivative gives phonon spectroscopic information. So far, we have ignored any effect due to junction capacitance because this effect is negligible if the junction capacitance is  $\gtrsim 10^{-13}$  F. However, such an effect cannot be neglected if the junction cross section area is small, ( $\leq (1\mu m)^2$ ), and its capacitance is smaller than  $10^{-15}$  F. Such a small-capacitance

effect is the subject of the next section.

## 5.2.2 Coulomb Blockade Theory

In a current biased normal metal—Insulating layer—normal metal (N-I-N) junction with a junction capacitance C and tunnel resistance  $R_t$ , the initial charge Q is a classical, continuous variable, and can be a fraction of an electron charge e because it is treated as a polarized charge in a capacitor. When an electron tunnels through the junction, the charge Q is changed by an electron charge e, which is a discrete quantum number. In such a semi-classical theory, the Coulomb energy varies:

$$\Delta E = \frac{(Q \pm e)^2}{2C} - \frac{Q^2}{2C} = \frac{\pm e}{C} \cdot (Q \pm \frac{e}{2})$$

when the initial charge is in the following range,

$$-\frac{e}{2} < Q < \frac{e}{2} \tag{5.14}$$
the energy change is always positive for any sign of  $\Delta Q = \pm e$ ,

$$\Delta E > 0$$

Therefore, the tunneling process is forbidden at low temperature when its initial charge Q is between  $\pm e/2$ . This effect is called the Coulomb blockade of single electron tunneling. The physical origin of this blockade is electron's strong Coulomb interaction in a junction with a small capacitance C.

For such a junction, its I-V characteristic curve is strongly non-linear, because of the Coulomb blockade effect.<sup>7</sup> The junction I-V is parabolic at small bias voltage:

$$I = (2C/\pi R_e)V^2 \qquad \text{for } |\mathbf{V}| \leq e/2C \qquad (5.15a)$$

and approaches the linear asymptote

$$I \to G_i \cdot (V - \frac{e}{2C} signV)$$
 for  $|V| \gg e/2C$  (5.15b)

The above two equations show that the Coulomb blockade effect has dramatically changed the characteristic I-V curve in two regions: at small bias voltage, an I-V is parabolic and conductance of the *N-I-N* junction is linear to the applied bias voltage:

$$\frac{dI}{dV} = (4C/\pi R_t e)V \qquad (5.15a)^t$$

Actually, the dynamic conductance in eqn.(5.15a)' is smaller than the junction conductance without the Coulomb blockade effect. In an ordinary tunnel junction, the conductance is constant at a small bias voltage. At a large bias voltage, the junction I-V is linear to tunnel conductance, but with a voltage offset of e/2C. In fact, those features are experimental evidences for the existence of the Coulomb blockade of single electron tunneling.

Only under some appropriate conditions, can this Coulomb effect be observed in experiments. Three kinds of energy scales play roles in single electron tunneling junction: charging energy  $E_C \cong e^{-2}/2C$ ; thermal energy  $E_T = k_BT$ ; and quantum fluctuation energy  $\Delta E \sim 2\pi\hbar/\tau$ , where  $\tau$  is the discharging time and equals  $R_{\ell}C$  for the junction. The condition for observing the coulomb blockade effect in such an experiment is:

$$E_C \gg \Delta E \gg E_T$$

i.e.

$$e^{2/2C} \gg 2\pi\hbar/\tau = 2\pi\hbar/R_{t}C$$
$$R_{t} \gg 4\pi\hbar/e^{2} = R_{O} = 26 \text{ k}\Omega$$

where  $R_O$  is known as the quantum resistance.

For example, if a junction tunnel resistance is about 100 k $\Omega$  and its capacitance *C* is about  $3 \times 10^{-15}$  F with a cross section area  $A \approx 1 \ \mu\text{m}^2$ , the temperature has to be in  $\leq 1$  K so as to observe the Coulomb blockade effect in such a junction, in which the offset voltage is around  $e/C \approx 50 \ \mu\text{V}$ , large enough for being observed at 0.3 K. However, such a requirement of low temperature can be relaxed at certain situations. For instance, if a junction has a much smaller capacitance  $C \approx 3 \times 10^{-18}$ F, where the cross section area  $S \approx 0.001 \ \mu\text{m}^2$ , the offset voltage  $e/C \approx 50 \ \text{mV}$ , so that the Coulomb blockade effect can be measured even at room temperature!

The first experiment to show clear evidence of the Coulomb blockade effect was performed with a double tunnel junction structure by Fulton and Dolan.<sup>8</sup> So far most experiments have been done in similar structures with a multi-junction array. These results of Coulomb staircases as well as microwave coupling data demonstrate Coulomb blockade of single electron tunneling.<sup>9,10,11</sup> The reason for such a situation is because the electrodynamic environment profoundly effects the behavior of small-capacitance tunnel junction . The stray capacitance in an electronic circuit lead is usually about  $10^{-12}$  F, which lowers the offset voltage to 0.1  $\mu$ V, thus reducing the Coulomb blockade effect dramatically. In a double junction structure or a multi-junction arrangement, the central junction is isolated from its electrodynamic environment by the nearest neighbor junctions, those

neighbor junctions are shielding away the effect of the large stray capacitance. As a result, the Coulomb blockade effect has been observed most frequently in such multi-junction array structures. Instead of using junctions to reduce the environment's electrodynamic influence, a high resistance lead with a resistance larger than the quantum resistance 26 k $\Omega$  placed very close to a junction region has been shown to effectively isolate the junction from its electrodynamic environment both theoretically and experimentally.<sup>12,13</sup>

However, some controversial experimental results for a single tunnel junction should be mentioned here also. Using a STM as a probe above a stainless steel surface as well as a superconducting material, Bentum et. observed the Coulomb blockade effect of single electron tunneling in a voltage-biased single junction formed by the STM's tip and the conducting material surface.<sup>14</sup> Later, in a single tunnel junction formed by two tiny metal filaments, Gregory also reported experimental evident for the Coulomb blockade of single electron tunneling in such a voltage-biased junction.<sup>15</sup> In both cases, no higher resistive leads were used to isolate the single junction from its electrodynamic environment as suggested by the theory. There has been speculation that there could be a tiny metal defect embedded between the STM's tip and the clean metal surface in the former experiment or between the two filaments in the latter case, so that these single junction were actually double junction structure. In the section 5.4 of this chapter, I will present experimental evidence for the Coulomb blockade effect in a voltage-biased tunneling junction.

## 5.3 Apparatus and Techniques

A standard ac lock-in technique in the electronic circuit shown in Fig 5.3 was used to measure the differential conductance G = dI/dV of a single junction verse the biased voltage. The dc bias voltage supply is simply a ramp generator with an adjustable amplitude and ramp scanning time. A small ac modulation voltage is coupled to the dc bias voltage through a 1:1.25 transformer so as to break the ground loop between different power sources. The ac current signal is magnified with a current amplifier, then measured by a lock-in. Then the output from the lock-in goes to an x-y recorder, which has an inline buffer memory for storing data. Finally, a computer reads data from the recorder for further data processing. One 40 k $\Omega$  resistor is inserted in the circuit to limit the maximum current and protect the current amplifier in case the tunnel junction shorts.

The above circuit for measuring the differential conductance G = dI/dV is based on the following relationship between the current and the biased voltage.

$$V = V_0 + \Delta V \qquad (5.3.1)$$
  
$$\Delta V = v Sin(\omega t + \phi_0) \qquad (5.3.2)$$

where  $V_0$  is the dc biased voltage from the ramp generator, and  $\Delta V$  is the ac modulation voltage coupled through the transformer. The current I(V) in this circuit will vary with the voltage as:

$$I(V) = I_{0} + \frac{\partial}{\partial V} \cdot \Delta V + \frac{1}{2} \cdot \frac{\partial^{2} I}{\partial V^{2}} \cdot (\Delta V)^{2} + \cdots$$

$$= I_{0} + \frac{\partial}{\partial V} \cdot v \cdot Sin(\omega t + \phi_{0}) + \frac{1}{2} \cdot \frac{\partial^{2} I}{\partial V^{2}} \cdot (v \cdot Sin(\omega t + \phi_{0}))^{2} + \cdots$$

$$I(V) = G_{0} \cdot V_{0} + \left(\frac{\partial}{\partial V} \cdot v\right) \cdot Sin(\omega t + \phi_{0}) + \left(\frac{1}{2} \cdot \frac{\partial^{2} I}{\partial V^{2}} \cdot v^{2}\right) \cdot (Sin(\omega t + \phi_{0}))^{2} + \cdots$$

$$= G_{0} \cdot V_{0} + \left(\frac{\partial}{\partial V} \cdot v\right) \cdot Sin(\omega t + \phi_{0}) + \left(\frac{1}{2} \cdot \frac{\partial^{2} I}{\partial V^{2}} \cdot v^{2}\right) \cdot \frac{1 - Cos(2(\omega t + \phi_{0}))}{2} + \cdots$$

$$I(V) = G_{0} \cdot V_{0} + \left(\frac{1}{4} \cdot \frac{\partial^{2} I}{\partial V^{2}} \cdot v^{2}\right) + \left(\frac{\partial}{\partial V} \cdot v\right) \cdot Sin(\omega t + \phi_{0}) - \left(\frac{1}{4} \cdot \frac{\partial^{2} I}{\partial V^{2}} \cdot v^{2}\right) \cdot Cos(2\omega t + 2\phi_{0}) + \cdots$$
(5.3.4)



Figure 5.3 Apparatus for single electron tunneling experiment. 40 k $\Omega$  resistor limits current in the case of tunnel junction short.



Figure 5.4 A schematic diagram of the mechanically controllable squeezable tunneling junction. The diagram is not to scale.

The coefficients in front of the first harmonic  $\omega$  sine and the second harmonic  $2\omega$  cosine are proportional to differential conductance and its derivative respectively. As a result, using a lock-in amplifier to pick out the first harmonic  $\omega$  term or the second harmonic  $2\omega$  term, we can obtain either the first derivative of current verse voltage  $\frac{\partial}{\partial V}$  (differential conductance) or its second derivative  $\frac{\partial^2 1}{\partial^2 V}$  respectively.

The mechanical squeezer shown in Fig. 5.4 is the basic device for controlling the tunneling gap in our tunneling measurements. The squeezer consists basically of a spring and one piston. The piston is guided along a smooth cylinder and driven up or down with little friction by a 2-64 screw, thus compressing or relaxing the spring. The other open end of the spring is attached to an aluminum plug attached to a thin stainless steel shim, in which a 1/32" diameter ball bearing is embedded in one hexagonal screw. The 10 mil thick cross-shaped shim is so flexible that it can moves up and down easily. However, it prevents lateral movement of the spring because its four corners are attached to the body of the squeezer. A ball bearing is used for two reasons. First of all, it has a small contact area (point contact) with the glass substrates so that the applied force is concentrated on the junction area. Second, its spherical surface always keeps applied force vertical to the glass substrate, thus largely eliminating a horizontal force.

The entire mechanical squeezer assembly was attached to one end of a long tube, and placed inside a liquid nitrogen Dewar. A long S.S. shaft is used to turn the 2-64 screw so as to compress or relax the spring for controlling the tunneling gap from outside of the Dewar.

## 5.3.1 Theoretical Calculation of Controlling Force vs. Tunneling Gap

A schematic diagram of our squeezable tunnel junction is shown in Fig 5.5. Two  $3/8"\times1"$  glass substrates are separated by four thin metal film spacers and forms a stiff spring. The lithographically-fabricated sample consists of two thermally-evaporated thin metal films on the glass substrates. The tunneling gap between those two electrodes is controlled by pressing the glass substrates with the squeezer shown in Fig. 5.4. The spring constant for such a system depends on its spacers' positions as well as on the glass substrate's thickness.



Figure 5.5 A schematic diagram of a squeezable tunnel junction sample with two electrodes on two glass substrates separated by four thin metal film spacers.

In order to calculate this spring constant, the sqeezable junction can be modeled as a beam which is supported on the two ends and pressed at the center by an external force F. The Young's Modulus Y of the beam is related to the force F and the deflection  $\varepsilon$ :

$$Y = \frac{Fl^3}{4st^3w} = \frac{Fl^2}{4st^3}$$

where *l* is the distance between the two supporting bars, or the two spacers, *w* is the length of the bar and equal to *l* approximately, *t* the thickness of the glass substrate. Its effective spring constant  $K_t$  is:

$$K_t = \frac{F}{s} = \frac{4Yt^3}{l^2}$$

Obviously,  $K_t$  is very sensitive to the thickness of the substrate and spacer's position.  $K_t$  is proportional to the cube of the thickness of the glass substrate, and inversely proportional to a square of the separation distance between two supporting bars.

In our experiment, the glass substrate t is 0.1 cm thick, and the four 0.2 cm square spacers are 0.4 cm apart from center to center,  $Y = 7.8 \times 10^{10}$  N/m<sup>2</sup>, the spring constant  $K_t$  for such a geometry is:

$$K_t = 3.2 \times 10^7 \, \text{N/m}$$

The spring constant for the steel spring in our sqeezer is  $K_t' = 28$  N/cm = 2800 N/m. the ratio for the displacements of these two springs is:

$$\gamma = \frac{\delta}{\delta'} = \frac{K'_t}{K_t} = \frac{2.8 \times 10^3}{3.2 \times 10^7} \approx 10^{-4}$$

The maximum deflection  $\varepsilon$  of the beam's center is:

$$\mathcal{E} = \frac{Fl^3}{48YI}$$

where I is the moment of inertia of the beam,

$$I=\frac{wt^3}{3}$$

where w and t are the length and the thickness of the bar respectively.

#### **5.3.2** Crack Junction

A diagram of a crack junction is shown in Fig. 5.6. The crack junction is similar to Morland's "Break junction".<sup>4</sup> A 30 µm wide Au line is fabricated by photolithography on a glass substrate with two predrilled holes. The detailed procedure for sample fabrication by photolithography was described previously in chapter 2, section 5.4. A force is provided by the same squeezer shown in Fig. 5.4, and redirected to one end of the glass substrate by a L-shape lever shown in Fig. 5.6, while the other end of the substrate is held by the base which is bolted solidly to the squeezer. An aluminum bar supports the substrate just underneath the narrow glass bridge between the two holes. When a small force is applied, the substrate will be bent just like the squeezable tunnel junction shown in Fig. 5.5. As the force increases and approaches a critical point, the glass substrate will crack along the supporting bar because a higher strain is built up there, thus breaking the metal film line above. Usually, such a crack line propagates through its glass substrate and reaches to the other side. However, by controlling the force through compressing the spring, it is possible that the crack line would stop at the middle of the glass substrate instead of going through. It is not crucial to the experiment.

After the thin metal film line is broken, it forms a tunnel junction with a very small capacitance because of the very small cross section of the thin metal film. By backing off the spring to reduce the force, the tunneling gap decreases until a tunnel current signal appears. Because the edges of the glass substrate are still continuous they produce a restoring force, just like in a bent beam, to overcount external force from the compressed spring.



Figure 5.6 Schematic diagram showing the controlling mechanism for a crack junction.

Unlike the squeezable tunnel junction, where the force is placed down at the center of the junction area, a crack junction has more complicated geometry arrangement and is more difficult to be modeled in a simple way. We have not calculated the tunneling gap verse the controlling force due to such complexities.

#### 5.4 Experimental Measurement and Results

#### 5.4.1 Coulomb Blockade with a Crack Junction

A photograph of a crack junction is shown in Fig. 5.7. The detailed procedure for sample fabrication by photolithography is described in section 2.5 of chapter 2. The sample is a thermally-evaporated 45 nm thick, 50  $\mu$ m wide, 1.2 cm long Au film above a 5 nm thick Cr layer for improving Au adhesion to the glass substrate, and has a 218  $\Omega$  resistance before cracking. The two electrical leads are attached to the two contact pads of the metal line by simply pressing indium (In) dots in the following procedure. First, a sample is cleaned by acetone and methanol; then, after a 1 mm diameter In wire is cut to several thin slabs by a razor, one piece of In is pressed down to the contact pad of the sample with the round end of a tweezers; then, with one electrical lead placed on the top of the pressed In pad, a second piece of In is placed over it, and pressed firmly, thus forming a solid electrical contact. This so-called cold-solder method produces excellent lead contacts for low temperature electronic measurements. Leads are 20 mil diameter Cu-Zn alloy wire.

After lead attachment, the squeezer with the sample is slowly lowered into the liquid nitrogen dewar. The sample is not immersed in the liquid nitrogen; instead, it remains in the cod nitrogen vapor, where T = 83 K at equilibrium.





Figure 5.7 Photograph of a crack junction with indium pads



Figure 5.8 Data for a crack junction at T= 77 K. Curve (A): plot of conductance G vs. bias voltage V; Curve (B): plot of tunneling current I vs. time t.

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The force on the sample is increased by turning the long screw driver of the sample stick until the metal line is broken, and the current decreases to zero in the electronic circuit, shown in Fig. 5.3.

A reference frequency of 510 Hz was chosen to minimize the Lock-in amplifier's intrinsic noise figure.<sup>16</sup> The root mean square (rms) of ac modulation voltage is about 0.7 mV, and the typical range of a ramp voltage is from -0.3 V to +0.3 V.

In Fig. 5.8, a conductance is plotted vs. ramp voltage as curve A, and curve B is a time trace of the tunneling current, both at T = 83 K. The ramp voltage rate is about 10 mV per second, while the time constant of the lock-in amplifier is 300 ms. From curve B, it can be seen that the tunneling current is very stable, with a fluctuation of  $\pm$  1%, at a low bias voltage region. For curve B, the full ranges of tunneling current and time axes are 1 nA and 120 seconds respectively.

From curve A, it is clear that conductance vs. bias voltage is linear at a low bias voltage in the range of  $|V| \le 0.1$  V. Above this range, the conductance signal becomes flat and more noisy at higher bias voltages. This can be explained by Coulomb blockade theory as mentioned previously in section 5.3. From curve A, the intercept of the linear line with plateau is about 0.12 V, where the tunneling current is 0.598 nA at the bottom and 0.8 nA at the intersection point. The corresponding conductances are 0.87  $\mu$ S and 1.16  $\mu$ S since the rms modulation voltage is 0.69 mV. The resistance R<sub>t</sub> of the plateau is 0.86 M $\Omega$ , so the capacitance C of the crack junction is, according to the eqn. (5.15a)':

$$\left[ \left( \frac{dI}{dV} \right)_{b} - \left( \frac{dI}{dV} \right)_{b} \right] / \Delta V = \frac{4C}{\pi R_{r}e}$$
$$\frac{4C}{\pi R_{r}e} = \left[ 1.16 - 0.87 \right] \mu S / 0.12V = 2.42 \mu S / V$$

$$\frac{e}{C} = \frac{4V}{\pi R_{t} (2.42 \mu S)} = 0.61V$$

This large offset voltage shows that the effective capacitance C of this crack junction is very small:

$$C = 3 \times 10^{-19} \, \mathrm{F}$$

However, this capacitance is much smaller than the capacitance calculated from the known geometry of the crack junction as follows:

$$C = \varepsilon \cdot \frac{A}{d}$$

where the cross section area is  $A = 0.05 \ \mu\text{m} \times 50 \ \mu\text{m} = 2.5 \ \mu\text{m}^2$ ,  $\varepsilon = 8.8 \times 10^{-12} \text{ s}^2\text{m}^{-2}$  is the permitivity of free space. Assuming a reasonable value of 20 Å for the tunneling gap d, we have:

$$C = \varepsilon \cdot \frac{A}{d} = 8.8 \cdot 10^{-12} \cdot \frac{2.5 \times 10^{-12}}{2 \times 10^{-9}} = 1.1 \times 10^{-14} \text{ F}$$

Gregory found a similar discrepancy in his data.<sup>15</sup> As pointed out before in section 5.2.2, this result is not expected for a single junction that is not isolated from its electromagnetic environment by nearby large lead resistance. A possible explanation for such a strange finding is that there is a tiny impurity defect in the tunneling gap, forming a double tunnel junction structure. However, we did not observe any Coulomb staircase as shown in other literature.<sup>13</sup>

Fig. 5.9 shows similar plot for another crack junction. The metal film line was broken in air at room temperature, and then the data were taken. This time, the linear regime obviously continues beyond the range of ramp voltage. At higher bias, tunneling current becomes more noisy. Moreland's squeezable tunnel junction showed the same noisy behavior at high bias voltage.<sup>9,12</sup> The reason for such behavior remains unknown.



Figure 5.9 Photograph of conductance vs. bias voltage for a crack junction at room temperature.

# 5.4.2 Two Level Conductance Fluctuation in a Squeezable Tunnel Junction

A photograph of an aligned sample for a squeezable tunnel junction is shown in Fig. 5.10. The sample is formed by two 1  $\mu$ m wide thermally evaporated metal lines facing each other on two glass substrates. The glass substrates are 1.2 mm thick, 1 inch long and 0.4 inch wide. It is a challenge to put such a pair of substrates together, and put the 1  $\mu$ m<sup>2</sup> junction area right underneath of the tip of the ball bearing. This is done with the help of an optical microscope.

The procedure for assembling the squeezable junction requires an inverted optical microscope. An inverted microscope is one in which the objective lens points up at a sample which faces down and is supported along its edges by the sample stage(see Fig.2.3). A 1" long, thin-walled Al cylinder with an inner



Figure 5.10 Photograph of a typical micron-sized squeezable junction

diameter of 3/4" fits snugly on the x10 objective lens of the microscope which has an X-Y-Z sample stage. The first substrate is placed on the top of the cylinder with the metal film sample facing upward and the cylinder is positioned on the objective lens so that the sample, viewed through the glass substrate, is in sharp focus. Then the second substrate, supported by the sample stage with its metal film sample facing down, is positioned over the first one and moved down slowly until both samples are in the focus, but still not touching. The sample stage and the top substrate are then positioned so that the two substrates are aligned with the four spacers on both substrates matching and the two metal stripes perpendicular to each other. Finally, the top substrate is lowered until the two substrates. Then, four electrical leads are attached to the four pads of the junction by pressing In slabs. No other better way has been found for attaching the leads after assembling the squeezer in our design.

Now, the junction with attached electrical leads has to be placed underneath the ball bearing of the squeezer in the mechanical squeezer shown in Fig. 5.4. This is done using a ruler and an optical microscope. A plastic ruler inserted into the path of illuminating light provides an one-dimensional coordinate in the field of the view. Using the ruler, the positions of the junction and the ball bearing can be located easily through the observing hole at the center of the bottom of the squeezer, even though the junction and the top of the ball bearing cannot be seen at the same time because of the limited depth of focus of the objective lens. As a result, the tunnel junction can be placed right at the center of the ball bearing's contacting area to the substrate by adjusting the junction's position carefully.

The mechanical squeezer is "top-loaded" in the following order: first, the aligned junction with electrical leads are placed onto the base of the squeezer; then the cross-shape stainless steel shim with the ball bearing is placed down along the four long bolts at the top; next, the spring is placed with piston at the top of the Al plug in the shim; finally, the top base with the smooth guiding cylinder and the # 2-64 screw is lowered down and four bolt nuts are tightened to make the entire squeezer solid.

The squeezer with the tunnel junction is then attached to one end of the sample stick, and lowered down into a liquid nitrogen dewar. Again, the tunnel junction is in the cold nitrogen vapor. It takes about two hours for the junction to come to equilibrium with the cold vapor. By turning a long S.S. shaft outside, the tunneling gap is decreased by the squeezer, until a tunneling current appears in the lock-in's monitor.

Fig. 5.11 shows the plot of conductance vs. bias voltage for a squeezable tunnel junction at 83 K. This curve is approximately parabolic, and varies slowly at low bias voltage, a behavior typical of a conventional tunnel junction with a large junction capacitance. At high bias voltage, the signal becomes noisy as mentioned previously in the crack junction's section. The reason for such a behavior remains unknown.

The tunnel current vs. time at low bias voltage is plotted in Fig. 5.12. This curve indicates that the mechanically-controlled squeezer has a very good stability,  $\pm 1\%$ . The curve in Fig. 5.13 shows the tunneling current trace vs. time while the tunneling gap is adjusted by the long S.S. shaft. First, the tunneling current is initially set to A1. After a while, the tunneling gap is decreased by turning the shaft, the tunneling current increases and goes off scale. However, the two electrodes do not contact. Then the screw shaft is turned to increase the tunneling gap, the tunneling current decreases, but it is still larger than the initial setting. A moment later, the tunneling gap is increased further, the tunneling current became smaller than the initial value. The horizontal axis is 2 minutes in full scale, the Y-axis is arbitrary unit. It is clear that our mechanical squeezer has good



Figure 5.11 Plot of conductance vs. bias voltage for a 1  $\mu$ m sample at T = 83 K.



Figure 5.12 Tunnel current vs. time for a 1  $\mu$ m sample at T = 83 K



Figure 5. 13 Trace of a tunneling current vs. time to show the controllable gap in the squeezable tunneling junction.



Figure 5.14 Two level conductance fluctuation of a sample in a tunneling junction.

controllability as well as stability during the entire process of the adjustment.

Finally, a two-level conductance fluctuation vs. time is show in Fig. 5.14. The data were taken at T = 83 K. Even though there is a background noise, it is clear that two level system shows up in the tunneling conductance fluctuation. We cannot identify the microscopic origin of this two-level conductance fluctuation yet. Nevertheless, it is speculated that there could be nanometer scale defect migrating near the tunneling area on the sample surface. When the defect jumps in or out of the tunneling area, it could modulate the tunneling gap, thus causing tunneling conductance fluctuation at two levels. Several minutes after the trace of Fig. 5.12 was taken, this two-level conductance fluctuation was gone. Either the tunneling area shifted away from the defect or the defect migrated away from the tunneling area.

It should be pointed out that we conducted such experiments for a large number of micron-sized squeezable tunnel junctions. However, none of them showed Coulomb blockade of single electron tunneling as the crack junctions did. Some effort was made to fabricate 0.1 µm tunnel junctions. Unfortunately, they were always destroyed by the lead attachments due to static electrical discharge because the sub-micron microbridges were not protected by shorting bridges. There was difficulty to scratch those lines in the squeezable tunneling junction because of the junction arrangement.

In conclusion, I have presented the design for the crack junction and the squeezable tunnel junction in this chapter. Both the crack junction and the squeezable tunneling junction demonstrated excellent mechanical controllability and stability. The experimental data from the crack junctions showed Coulomb blockade of single electron tunneling and the squeezable tunnel junctions showed two-level conductance fluctuation at T = 83 K.

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