Chapter 3

Experiment

3.1 Samples Fabrication

All the tunnel junctions were fabricated by thermal evaporation on Corning glass substrates. First, several 0.5 mm thickness Corning glass substrates with size 9.6 mm×6 mm and a suitable amount of Al wires or pellets (99.999 at.%) were cleaned by Acetone followed by Alcohol. After cleaning, the substrates, Al wires or pellets, and tungsten basket coated with Al_2O_3 were put in the evaporating chamber followed by evacuating. We used the evaporating source coated with Al_2O_3 because the melted Al would alloy and wet Ta-, Mo-, or W-boats. The chamber was equipped with a turbo-evacuating system and its base pressure was about 1×10^{-6} torr.

3.1.1 Al Films Deposition

It took about 4 hrs to reach the pressure $\sim 3\times10^{-6}$ torr, and at or below this pressure, we started to deposit Al film. The deposition thickness and rate were monitored using a quartz crystal with 1 Å resolution in thickness and 0.1 Å/sec resolution in rate. At the beginning of the evaporation, the substrate shutter was

closed (to shelter the substrates from any deposition) and the heating power across the tungsten was slowly added until the Al melted (the melt could be observed through the window on the chamber). When the Al started to melted we temporarily stopped adding any more power to the evaporating source (at this time the heating power is fixed), and waited the Al to melt completely. Because the Al_2O_3 crucible has bad thermal conductivity, it needs more time to transfer the heat from the tungsten basket to the Al wires or pellets. The temporary stop would prevent too much heat transferred to the Al so that the melted Al jetted out from the crucible.

It took about 1 min for the Al to melt thoroughly, and then the rate displayed in the film thickness monitor would increase slowly from 0 to ~ 0.5 Å/sec. After the thorough melting we added the heating power slowly to increase the evaporation rate to the value we wanted and waited for about $30 - 60$ sec to cover the environment with a thin Al film under the situation that the shutter is closed. This could improve the quality of the Al film we would deposit subsequently to the substrate. After the pre-evaporating, we open the shutter and deposit the Al films onto the substrates to the thickness we needed. Then closed the shutter and slowly lowered the heating power to turn off. It is about 3 Å/sec in the evaporation rate and 250 Å in the thickness when we deposited Al. The resistivity of the Al films at room temperature under this deposition condition is about 15 $\mu\Omega$ cm .

3.1.2 The Growth of the AlO_x

The quality of AlO_x is crucial to a good tunnel junction. If the oxide layer is too thin, the probability of the existence of pinholes would increase. Increasing the thickness of the oxide layer would lower the probability of the existence of

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pinholes, but increase the impedance of the junction and the magnitudes of Johnson noise. In our experience, a junction with resistance $\gtrsim 1$ kΩ (at room temperature, junction cross-section ≈ 1mm^2) usually implies a low leakage (through pinholes) tunnel junction. To be convincing, the quality of the junction should be examined through measuring the superconducting gap of the Al film, which would be described in detailed in Section 4.1.

In the early days, we made the AlO$_x$ layer by exposure of the evaporated film to air from the laboratory. By this method, the thickness of the oxidation layer and therefore the resistance of the junction is hard to controlled well in every fabrication. This was due to the oxidation rate is very sensitive to the degree of moisture in the atmosphere. Finally we tried growing the AlO$_x$ layer by glow discharge in an dilute O$_2$ atmosphere [34, 35]. The process is described as following. After finishing the Al film deposition, 30 mins was taken for the system to cool down. Then a small amount of O$_2$ flow was introduced into the chamber and at the same time a mechanical pump was used to evacuate the chamber. The amount of the O$_2$ flow could be tuned by a needle valve such that the pressure of the chamber in a steady state would be fixed at about 0.1 torr. The pressure 0.1 torr should be maintained for about 3 to 5 mins to let the distribution of O$_2$ gas in the chamber to become more steady. Then a negative voltage (~ -500 V) was applied to a cathode below the substrates, as shown in Fig. 3.1. This would result in electrons ionized from the cathode and accelerated towards the anode (in this system all components except the cathode are anode, including the sample holder). The energetic electrons would collide with O$_2$ molecules, transfer their energy to these molecules, and make the molecules to be at their excited or ionic states. The O$_2$ molecules in excited state
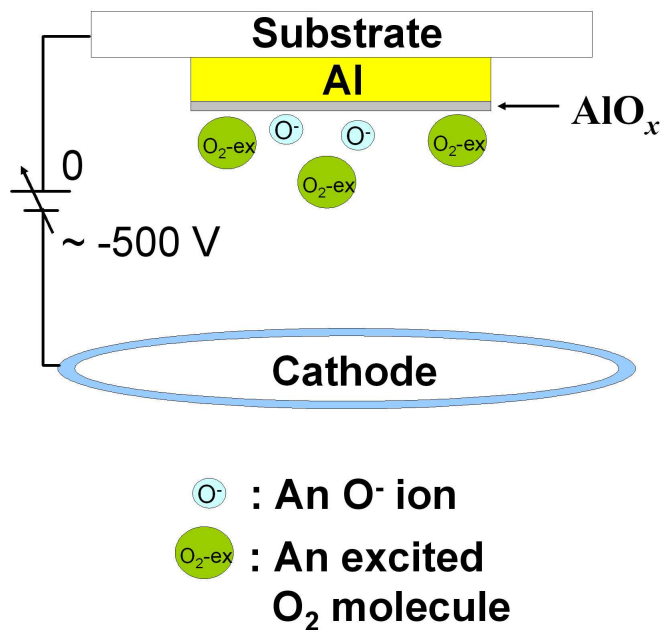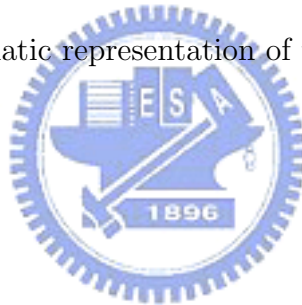
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Figure 3.1: A schematic representation of the glow discharge.

have higher efficiency to oxidize than in normal states. During the glow discharge process, O^+ ions would form, accelerate towards the cathode, collide the cathode and make particles which adhere to the cathode to be sputtered. Therefore initially we kept the shutter below the substrates in its closed state to protect the Al films from deposition from the sputtered particles. The sparkles at the cathode would disappear after 1–2 mins, and then we open the shutter to let the Al films to be oxidized for 3–5 mins. The barrier thickness and height under these condition was about 15 – 20 Å and 1.2 – 1.5 eV respectively, which would be discussed in Section 4.1.

3.1.3 Sc Films Deposition

After the growth of the oxide layer, we evaporate Sc films crossed to the Al films through the corresponding shadow mask. A tungsten boat was used as the evaporating source and the background pressure prior to evaporation was $\sim 3 \times 10^6$ torr. The deposition process was similar to that of Al films. After the deposition, about 1 hr was taken for cooling down. The finished junctions were shown in Fig. 2.1 (a), where Metal 1 is the Al film and Metal 2 is the Sc film.

3.1.4 Initial Test of the Junctions

After finishing the fabrication, we measured the junctions resistance initially by a simple multi-meter. The high impedance (low output current) mode was chosen for avoiding damaging the barrier. Junctions whose resistance were between 1–10 kΩ is appropriate for measurement.

3.2 Measurement Circuits

We measured the differential conductance dI/dV using lock-in techniques. A "send V measure I" circuit was used to measure the superconducting gap of the Al film while a "send I measure V" circuit was used to measure dI/dV in its normal state.

3.2.1 The Principles of the Lock-in Amplifier

Since we are interested in the dI/dV spectra of the tunnel junctions, we have two methods to obtain them. One is to measure the tunneling current I as a function of the bias voltage V, and then calculate the dI/dV. In practice, a set of discrete V_i are applied to the sample by a DC voltage source (here we consider the "sent

V measure I" as an example), and the corresponding responses I_i are measured by a DC current meter, and the dI/dV can be approximated to be $\Delta I/\Delta V$, where $\Delta V = V_{i+1} - V_i$ and $\Delta I = I_{i+1} - I_i$, if the intervals in the V_i we applied are sufficient small. This method is quite direct and simple, but the signal to noise ratio is small. The other method is to use an AC modulation and lock-in techniques, which has two advantages. One is it is suitable for low level measurement and the other is the dI/dV can easily obtained using this method. The principles of the method are described in the following, in which we consider the "send I measure V" as an example.

Consider applying a dc current I together with a small modulation ac current $\Delta I \sin(\omega t)$ to a sample. The corresponding voltage developed in the sample due to the application of the current can be expressed as $V(I + \Delta I \sin(\omega t))$, and can be expanded as

$$
\begin{aligned}
V(I + \Delta I \sin(\omega t)) &= V(I) + \frac{dV}{dI} \Delta I \sin(\omega t) + \cdots \\
&\approx V(I) + \frac{dV}{dI} \Delta I \sin(\omega t).
\end{aligned} \tag{3.1}
$$

The approximation in (3.1) is based on the assumption that the modulation AC current is small. In addition to the response voltage, the total voltage across the sample contains the contribution of noise $V_{noise}(t)$, and hence the total voltage $V_{total}(t)$ can be expressed as

$$
\begin{aligned}
V_{total}(t) &\approx V(I) + \frac{dV}{dI} \Delta I \sin(\omega t) + V_{noise}(t) \\
&= V(I) + \frac{dV}{dI} \Delta I \sin(\omega t) \\
&\quad + \{a(0) + \sum_{\omega_n} [a(\omega_n) \cos(\omega_n t) + b(\omega_n) \sin(\omega_n t)]\}, \tag{3.2}
\end{aligned}
$$

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where the $V_{noise}(t)$ is expanded in Fourier series in (3.2). Then the total voltage $V_{total}(t)$ is introduced into a lock-in amplifier to process the signal further. On the other hand, two signals with the same frequency ω as used in the modulation are generated inside the lock-in amplifier, and they are

$$V_1(t) = V_L\sin(\omega t + \theta), \tag{3.3}$$

$$V_2(t) = V_L\sin(\omega t + \theta + \frac{\pi}{2}), \tag{3.4}$$

respectively, where θ is the phase relative to the modulation signal. Then the introduced signal $V_{total}(t)$ is multiplied by $V_1(t)$ and $V_2(t)$ to get $V_3(t)$ and $V_4(t)$ respectively:

$$
\begin{aligned}
V_3(t) &= V_{total}(t)V_1(t) \\
&= V(I)V_L\sin(\omega t + \theta) + \frac{dV}{dI}\Delta I\sin(\omega t)V_L\sin(\omega t + \theta) \\
&\quad + \{a(0) + \sum_{\omega_n}[a(\omega_n)\cos(\omega_n t) + b(\omega_n)\sin(\omega_n t)]\}V_L\sin(\omega t + \theta), \\
&= V(I)V_L\sin(\omega t + \theta) + \frac{1}{2}\frac{dV}{dI}\Delta I V_L[\cos\theta - \cos(2\omega t + \theta)] \\
&\quad + a(0)V_L\sin(\omega t + \theta) \\
&\quad + \sum_{\omega_n}\frac{1}{2}a(\omega_n)V_L\{\sin[(\omega_n + \omega)t + \theta] - \sin[(\omega_n - \omega)t - \theta]\} \\
&\quad + \sum_{\omega_n}\frac{1}{2}b(\omega_n)V_L\{\cos[(\omega_n - \omega)t - \theta] - \cos[(\omega_n + \omega)t + \theta]\},
\end{aligned}
$$

$$\tag{3.5}$$

and similarly,

$$
\begin{aligned}
V_4(t) &= V_{total}(t)V_2(t) \\
&= V(I)V_L\sin(\omega t + \theta + \frac{\pi}{2})
\end{aligned}
$$

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$$+\frac{1}{2}\frac{dV}{dI}\Delta IV_L[\cos(\theta+\frac{\pi}{2})-\cos(2\omega t+\theta+\frac{\pi}{2})]$$

$$+a(0)V_L\sin(\omega t+\theta+\frac{\pi}{2})$$

$$+\sum_{\omega_n}\frac{1}{2}a(\omega_n)V_L\{\sin[(\omega_n+\omega)t+\theta+\frac{\pi}{2}]-\sin[(\omega_n-\omega)t-\theta-\frac{\pi}{2}]\}$$

$$+\sum_{\omega_n}\frac{1}{2}b(\omega_n)V_L\{\cos[(\omega_n-\omega)t-\theta-\frac{\pi}{2}]-\cos[(\omega_n+\omega)t+\theta+\frac{\pi}{2}]\}.$$

$$(3.6)$$

$V_3(t)$ and $V_4(t)$ are passed through low-pass filters to get V_5 and V_6 respectively:

$$V_5 = \frac{1}{2}\frac{dV}{dI}\Delta IV_L\cos\theta+\frac{1}{2}a(\omega)V_L\sin\theta+\frac{1}{2}b(\omega)V_L\cos\theta, \qquad (3.7)$$

$$V_6 = \frac{1}{2}\frac{dV}{dI}\Delta IV_L\cos(\theta+\frac{\pi}{2})+\frac{1}{2}a(\omega)V_L\sin(\theta+\frac{\pi}{2})$$

$$+\frac{1}{2}b(\omega)V_L\cos(\theta+\frac{\pi}{2}), \qquad (3.8)$$

In deriving (3.7) and (3.8), we note the non-zero frequencies are all filtered by the low-pass filter, and hence the contribution of the noise will be filtered except $\omega_n=\omega$. Generally speaking, $a(\omega)\ll(dV/dI)\Delta I$ and $b(\omega)\ll(dV/dI)\Delta I$ (the modulation frequency ω should be chosen to be away from the frequency at which the noise amplitude is large), and hence

$$V_5 \approx \frac{1}{2}\frac{dV}{dI}\Delta IV_L\cos\theta, \qquad (3.9)$$

$$V_6 \approx \frac{1}{2}\frac{dV}{dI}\Delta IV_L\cos(\theta+\frac{\pi}{2}). \qquad (3.10)$$

In the final stage, the lock-in amplifier will execute the following procedure:

$$\frac{2}{V_L}(V_5^2+V_6^2)^{1/2}. \qquad (3.11)$$

According to (3.9) and (3.10), (3.11) is equivalent to $(dV/dI)\Delta I$, and can be displayed on the screen of the lock-in amplifier. The magnitudes in which we are
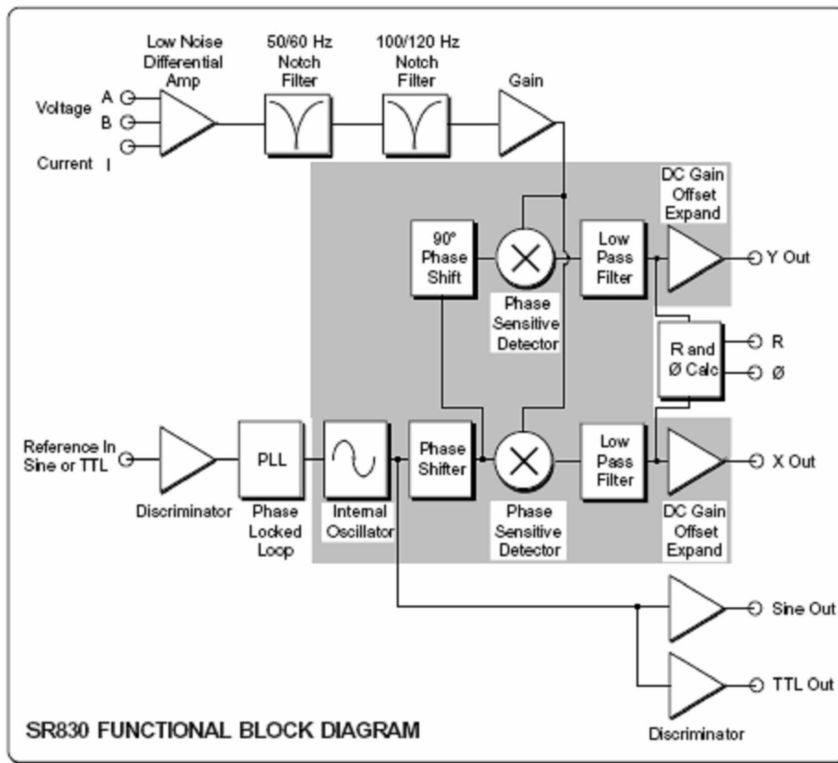
Figure 3.2: The functional block diagram of the SR830 lock-in Amplifier. This diagram is obtained from the user's manual of the SR830.

interested, dI/dV, can be obtained by dividing ΔI, which is known, by $(dV/dI)\Delta I$, which is displayed on the screen. By varying I, we can get the dI/dV at each bias voltage $V(I_i)$.

The lock-in amplifier we used is a product of Stanford Research Co., SR830, and its functional block diagram is shown in Fig. 3.2.
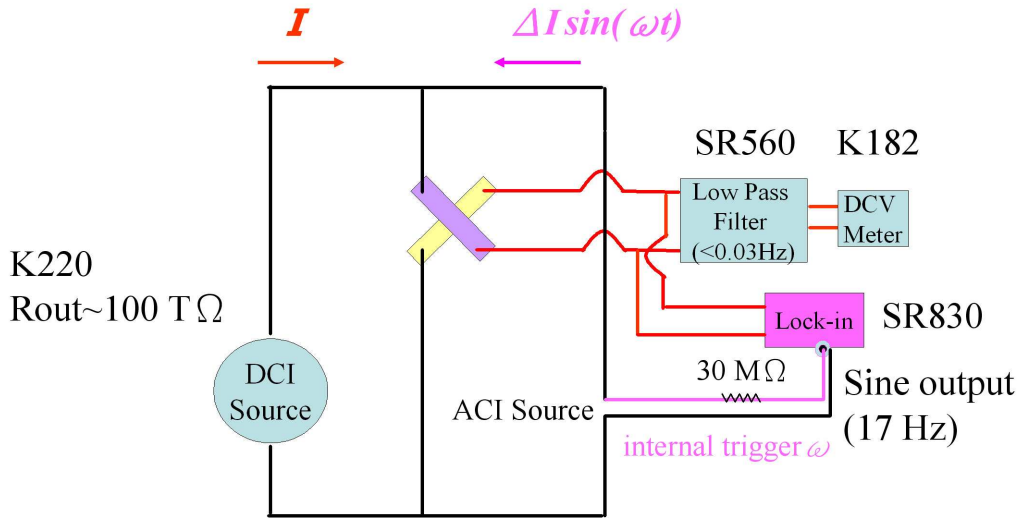
Figure 3.3: The "send I measure V" circuits.

3.2.2 The "Send I Measure V" Circuit

Fig. 3.3 show the "send I measure V" circuit we used. A dc current source Keithley 220 is used to output a dc current I, and a small ac modulation current $\Delta I \sin(\omega t)$ is acquired by connecting a large resistance resistor (30 MΩ) in series with the ac voltage out of the SR830. The frequency ω and the magnitude of the modulation current ΔI can be tuned by setting the corresponding parameters in the SR830. The dc current shunting to the SR830 can be neglected due to the large resistance resistor in the path, and the ac current shutting to the Keithley 220 can be neglected due to its large output resistance (~ 200 TΩ). Therefore, almost all I and $\Delta I \sin(\omega t)$ pass through the tunnel junction, and result in a response voltage $V(I + \Delta I \sin(\omega t))$, which contains a dc and an ac component. The dc component can be measured by introducing the response signal into a low-pass filter, SR560, to filter all the ac components, followed by a dc voltmeter, Keithley 182. The ac component can be
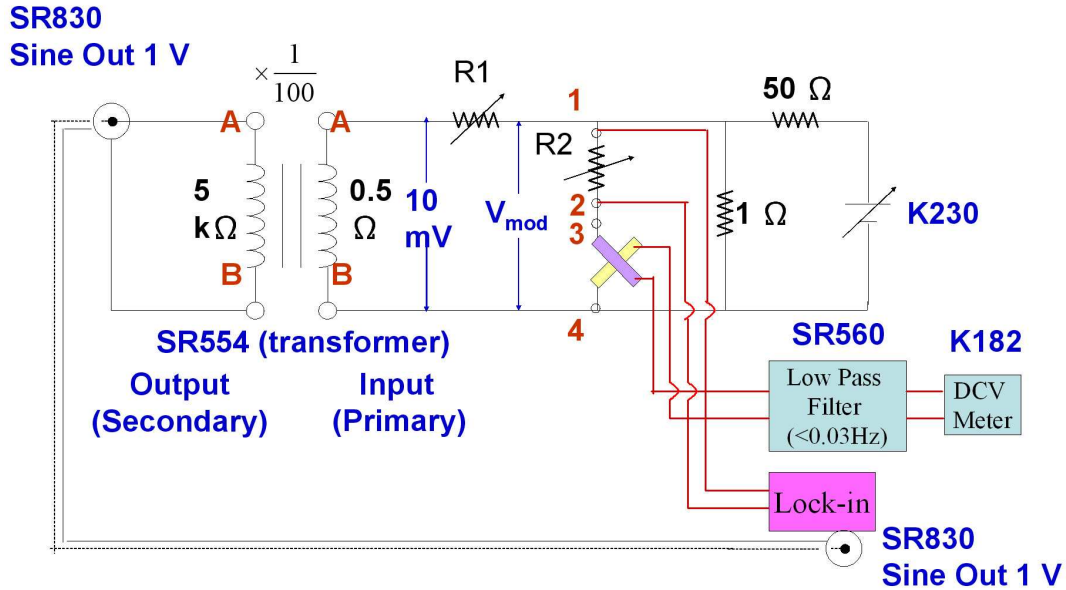
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Figure 3.4: The "send V measure I" circuits.

acquired by the lock-in amplifier, SR830, which uses the internal trigger to process the input signal, and the procedures have been described above. We should note we send current in two terminals and measure the voltage through the other two terminals which can avoid the contribution of the lead and the contact resistance.

3.2.3 The "Send V Measure I" Circuit

For the superconducting gap measurement, it is better to use a "send V measure I" circuit rather than a "send I measure V" circuit. When one lead is in the superconducting state, the dI/dV of a tunnel junction for $|V| < \Delta = E_g/2$ is much smaller than that for $|V| > \Delta$. Therefore, the circuit should have the ability to apply V with nearly equal spacing to the junction when V sweeps from $|V| < \Delta$ to $|V| > \Delta$. For this purpose we devised the circuit as shown in Fig. 3.4.

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We want to apply $V + \Delta V \sin(\omega t)$ to the tunnel junction, and measure the response dc and ac component. The dc V is supplied by a dc voltage source, Keithley 230, and the ac voltage was supplied by the sine out of the SR830. A 50 Ω resistor is used in series with the output of the Keithley 230 to protect the voltage from an ac current shunt. A 1 Ω resistor is used to give the junction a nearly equal spacing bias whether $|V| < \Delta$ or $|V| > \Delta$, since the junction resistance is always much larger than 1 Ω whether $|V| < \Delta$ or $|V| > \Delta$, and the bias between node 1 and node 4 is fully pinned by the 1 Ω resistor. A transformer SR554 is used to isolate the SR830 sine out from the dc bias, and this can protect the sine out from a dc current shunt. The sine out is set to be 1 V and is introduced into the secondary part of the SR554, and the amplified signal ($\times$ 1/100) 10 mV can be obtained from the primary part. The resistor R_1 is used for three reasons: the first, is to act as a voltage divider to give an appropriate modulation voltage across node 1 and node 4, the second, is to protect the primary part of the SR554 from the dc current shunt, and the third, is to ensure that almost all the dc and ac current pass through the 1 Ω resistor so that the 1 Ω resistor can well control the voltage across node 1 and node 4. The dc voltage across the junction can be measured using the same method as described in the "send I measure V" subsection. A small resistance resistor R_2 is connect in series with the junction. The magnitude of the ac current passing through the junction, ΔI, can be obtained by measuring the ac voltage across R_2 and dividing it by R_2. Then, the dI/dV can be approximately calculated by dividing ΔI by ΔV, where ΔV is known if the values of R_1 and R_2 are known.

3.3 The ³He Cryostat

We measured the dI/dV spectra in low temperatures using an Oxford Heliox ³He cryostat with superconducting magnet, whose base temperature is about 240 mK, and the maximum magnetic field is 4 T.

3.3.1 A Brief Description

There are 5 thermometers in the cryostat, as shown in Fig. 3.5. One 100 Ω Allen-Bradley resistor on sorb, one 2200 Ω RuO_2 on 1 K plate, two 2200 Ω RuO_2 on ³He pot (one of these was calibrated from 4.2 K to 0.25 K), and one calibrated Cernox (from 325 K to 1.4 K) on ³He pot. There are 24 twisted pairs of experimental wires in this cryostat, and the diameter of all these wires is 0.1 mm. 12 pairs of these are constantan and the other are cooper. To avoid too much heat conducting from room temperature to 3He pot, we use only the constantan wires to measure and leave the copper wires untouched to the ³He pot.

3.3.2 The Cool Down Process

After mounting the sample, soldering the leading wires, and a simple examination of the sample, the can was sealed and the IVC (inner vacuum chamber) was evacuated using a mechanical pump for at least 1 hr. After one hour pumping, the pressure in IVC should be below 10 mtorr. Then a small amount of exchange gas (⁴He, ∼15 mtorr) was introduced into the IVC (We introduced ∼ 80 mtorr ⁴He into the IVC and then pumped it to 15 mtorr). The cryostat at this moment was ready for cool down.
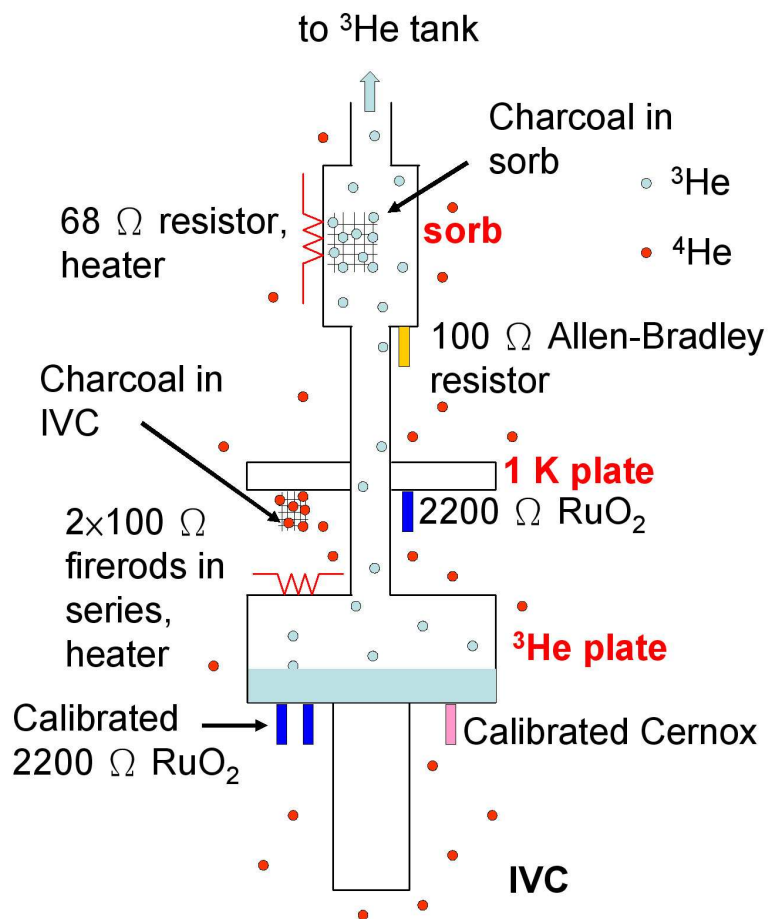
to ^{3}He tank

Charcoal in
sorb

^{3}He

^{4}He

68 Ω resistor,
heater

sorb

100 Ω Allen-Bradley
resistor

Charcoal in
IVC

1 K plate

2$\times$100 Ω
firerods in
series,
heater

2200 Ω RuO$_2$

^{3}He plate

Calibrated
2200 Ω RuO$_2$

Calibrated Cernox

IVC

Figure 3.5: The schematic representation of the IVC of the ^{3}He fridge.

From 300 K to 4.2 K

We used LN_2 (liquid nitrogen) to pre-cool the system. When the temperature is below 90 K, the cryostat was transferred into a ^{4}He dewar. The temperature of ^{3}He pot would decrease to ∼4.5 K in about 40 mins after the transferring. In this process the cooling down is carried out by heat exchange between the inner wall of the can (which is about 4.2 K) and the ^{3}He pot through the exchange gas.

From 4.2 K to 1.5 K

When the cryostat was near its equilibrium (the temperature of ^{3}He pot was about 4.5 K), we started to pump the 1 K plate using a mechanical pump and this would introduce a flow of liquid ^{4}He into the capillary. The 1 K plate and sorb would cool down to about 2.2 K due to their thermal contact to the capillary.

From 1.5 K to 240 mK

To cool down the temperature to lower, a condensation of ^{3}He and then a vaporization of it is utilized. In the condensation process, the temperature of sorb and 1 K plate are kept to be ∼30 K and ∼2.2 K respectively. The trapped ^{3}He molecules on the charcoal in sorb would be released due to the high temperature (∼30 K), and these ^{3}He molecules would condense in the ^{3}He pot due the heat exchange with 1 K plate (∼2.2 K). It would take about 30 mins for condensation.

After completing the condensation, the temperature of sorb was lowered and this would make the charcoal in the sorb to absorb ^{3}He molecules. The absorption would lower the pressure in the ^{3}He pot, and result in vaporization from the condensed ^{3}He. The vaporization will take away the heat and then lower the temperature of

^{3}He pot. The temperature of ^{3}He pot can be controlled through controlling the temperature of sorb and the lowest temperature can reach 240 mK.