Design of an Experiment to Probe the Pseudogap State in High-$T_c$ Cuprate Superconductors with Mesoscopic Transport

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Abstract

In this thesis I propose the design and demonstrate the initial development of an experiment to perform mesoscopic transport measurements on high temperature cuprate superconductors. The goal is to determine the charge of normal state charge carriers in the pseudogap regime by measuring the magnetic field periodicity of the quantum correction to conductance in the Aharonov-Bohm effect. There are two fronts to the experiment: nanofabrication of complex oxides and measurement of mesoscopic effects in the transport of normal carriers only. The design and first trials with the nanofabrication technique are presented. The technique is based on pre-patternning the substrate with a focused ion beam and subsequently growing an epitaxial film of the high-$T_c$ material with pulsed laser deposition. The design of several measurement techniques is also presented. Two are based on high-frequency transport which allows to resolve the contribution to impedance of thermally excited normal carriers in the superconducting state. Another technique is based on standard low-frequency 4-point measurements on heavily underdoped nanostructures with $T_c$ suppressed to sub-Kelvin levels.
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Chapter 1

Motivation

Superconductors are materials that below a certain temperature exhibit the following phenomena: zero electrical resistance at DC, total exclusion of magnetic fields from the bulk (Meissner effect), and, most interestingly, macroscopic quantum coherence of all the electrons. These phenomena are very interesting from the standpoint of fundamental physics, but are also extremely useful in a great number of applications, such as extremely high quality resonators, high-field magnets, lossless power transmission, levitation, extremely sensitive magnetometers and phase-coherent superconducting electronics. Unfortunately, the applicability of superconductors has been limited by low superconducting transition temperatures. For conventional superconductors, these temperatures are below 20K, requiring expensive and cumbersome cryogenic cooling mechanisms involving liquid helium.

The microscopic mechanism of conventional superconductors is very well described by the Bardeen-Cooper-Shrieffer (BCS) theory and requires low transition temperatures. In the 1980’s, a whole new type of superconductivity was discovered, based on more complex crystals, with much higher transition temperatures (see figure 1.1). In these materials, superconductivity appears to be due to a completely different mechanism, still unknown, giving much hope for one day discovering or engineering room temperature superconductors. This goal and the interest in pure physics motivate the pursuit of this most tantalizing question in modern physics: that of the mechanism behind high $T_c$ superconductivity.

A number of models currently attempt to describe this mechanism, but no conclusive experimental evidence for any of them exists to this day. The most promising model is that of the BCS-BEC crossover, which seems to explain the observed pseudogap in the energy spectrum above
Figure 1.1: Highest $T_c$ known vs. year of material discovery

$T_c$. One direct way to verify this idea is to measure the charge of carriers in this pseudogap regime. This is what we seek to do by resorting to the ideas and techniques of mesoscopic physics.
Chapter 2

Background

2.1 Superconductivity

2.1.1 Phenomenology

Superconductivity is a ground-state phase that occurs in certain solids below some critical values of temperature $T_c$, magnetic field $H_c$ and current $J_c$. The critical values indicate the existence of a gap $\Delta$ in the electron energy spectrum, that at zero temperature, field and current is equal to the energy associated with each of the critical values. The superconducting phase exhibits the following phenomenology:

1. **Perfect electrical conductivity.**
   
   At DC, superconductors offer zero electrical resistance to the flow of current. At higher frequencies, there is a reactive component to the impedance associated with the inertia of the superconducting charge carriers, and a dissipative component associated with the inertial of thermally excited normal carriers. The latter disappears at $T = 0K$. These effects are key to the experiment proposed here and a quantitative analysis of them is included in section 4.3.1.

2. **Poor thermal conductivity.**
   
   While the superconducting charge carriers carry current, they carry no entropy, resulting in a thermal conductivity that is very poor and that goes to zero at $T = 0K$. This suggests some form of macroscopic ordering of the ground state phase.

3. **Meissner effect**
   
   Superconductors behave like perfect diamagnets, exponentially excluding all magnetic fields
from the bulk beyond the penetration depth $\lambda$. Unlike diamagnets, static fields are excluded as well, which is called the Meissner effect.

The macroscopic ordering hinted at in point 2 above is well described by the phenomenological Ginsburg-Landau theory \cite{1}. It assigns a complex order parameter $\Psi$ to the phase, which also acts as the quantum mechanical ground-state wavefunction of all the superconducting charge carriers. This means that all these carriers are phase-coherent in a quantum mechanical sense, leading to various observable interference phenomena, such as fluxoid quantization \cite{1}.

### 2.1.2 Bardeen-Cooper-Shrieffer theory

The Bardeen-Cooper-Shrieffer (BCS) theory of superconductivity is a microscopic theory that predicts extremely well the properties of the superconducting state for a limited class of superconductors. According to BCS, an arbitrarily weak attractive interaction will cause electrons at the Fermi surface to form bound states of pairs at an energy $\Delta$ below the Fermi energy $E_F$ \cite{1}. These so-called Cooper pairs are the superconducting carriers in question in the phenomenological theories. The interaction must be attractive and mediated by bosons. In many superconductors, phonons are suitable bosons, and attractive interaction occurs when the spread of electron energies $k_B T_c$ about $E_F$ becomes smaller than the Debye energy $k_B \Theta_D$, which characterizes the cutoff of the phonon spectrum. Physically, phonons correspond to the movement of the positive ion cores of the lattice, the displacement of which by one electron can attract another electron if the stiffness of the lattice (characterized by the phonon frequencies) is high enough given the speed of the electrons. One major success of the BCS theory is the prediction of the properties of the energy gap $\Delta$ observed to exist in the superconducting state and to vanish towards $T_c$.

### 2.1.3 High-$T_c$ superconductivity, pseudogap and BEC-BCS crossover

High-$T_c$ superconductors are a set of materials, usually complex oxides, with transition temperatures much higher than can be explained by the phonon-mediated Cooper pairing of the BCS theory. One qualitatively different feature of these superconductors is an energy gap in some regions of the Fermi surface in the normal state. This so-called pseudogap has been observed by a variety of techniques, such as angle-resolved photoemission spectroscopy (ARPES), tunneling spectroscopy, nuclear magnetic resonance, transport measurements, specific heat measurements, as well as Raman and neutron scattering \cite{2}. Another qualitatively different feature is the anisotropy, in
particular, the D-wave nature of the superconducting gap, varying as the cosine function around the Fermi surface (see figure 2.1). Averaging over all the directions results in non-zero excitation probabilities everywhere within the superconducting gap observed with the various spectroscopies mentioned above.

An interesting observation is that at $T_c$, the superconducting gap does not vanish, contrary to BCS predictions, but evolves into the normal state gap observed. Tunneling spectroscopy data shown in figure 2.2 clearly illustrates this phenomenon. Interestingly, the pseudogap persists to very high temperatures, much greater than $T_c$. Note also the non-zero spectrum in the superconducting gap below $T_c$.

By means of the various spectroscopies, the phase diagram of many high-$T_c$ materials has been mapped out; its typical temperature vs. doping profile is shown in figure 2.3.

A possible scenario that is believed to explain the evolution of the pseudogap from the superconducting gap is the so-called BCS-BEC crossover theory, where the fermionic chemical potential
changes from $E_F$ in the weak coupling limit (BCS regime) to $-E_F$ in the strong coupling limit (Bose-Einstein condensation, or BEC regime). This is illustrated in figure 2.4. In the BCS regime, electrons (or, more precisely Fermi liquid quasiparticles) exist as fermions, which form Cooper pairs and condense to the superconducting ground state simultaneously at $T_c$. In the BEC regime, electrons are paired up and form bosons, which Bose-condense at $T_c$. In the intermediate regime, there are two critical temperatures: $T^*$, at which the fermions form bound bosonic pairs, and $T_c$, at which the bosons condense to the ground state. Between the two temperatures is when the pseudogap is observed, associated with the energy of forming the bosonic pairs.
Figure 2.3: Typical temperature vs. doping phase diagram for high-$T_c$ superconductors \[3\].

Figure 2.4: Fermionic chemical potential at $T = 0$ defining the three regimes.
2.2 Mesoscopic Phenomena

2.2.1 The Aharonov-Bohm effect

Mesoscopic phenomena occur in solid state systems at length scales at which the phase coherence of the quantum mechanical wave functions describing the electrons is preserved. Consider a conductive ring with current passed through it and voltage measured across (impedance measurement) (see figure 2.5). The state of an electron moving through the ring is a superposition of states of the electron going through the top branch and through the bottom branch as follows [4]:

\[ |\psi\rangle = a_u|\psi_u\rangle + a_l|\psi_l\rangle \]  

(2.1)

Figure 2.5: An electron traveling through an Aharonov-Bohm ring.

For a strictly 1-D ring, we can project the state onto the x-coordinate and find the probability of transmission, or of finding the electron on the right hand side of the ring at \( x = x_r \):

\[ P_{\text{trans}} = |\psi(x_r)|^2 = |a_u\psi_u(x_r)|^2 + |a_l\psi_l(x_r)|^2 + 2Re[a_u^* a_l \psi_u(x_r)^* \psi_l(x_r)] \]  

(2.2)

The last term is the electron wave interference term analogous to the one in Young’s double slit experiment with coherent light. Similarly, coherence of electron waves is assumed here. Decoherence mechanisms, which arise from inelastic scattering events, and therefore increase with the size of the ring, suppress the interference pattern. For now, we assume perfect coherence. Assuming the structure is perfectly symmetric, we expect \( a_u = a_l = a \) and \( \psi_u(x_r) = \psi_l(x_r) \); the electron is as likely to go through the top branch as through the bottom branch. We now turn on a magnetic field \( B \) through the ring. From the path integral formulation of quantum theory [5], this field introduces a phase factor for each of upper and lower states as follows:
\[
a_u \rightarrow a_u \exp \left( \frac{iq}{\hbar} \int_{x_l}^{x_r} \vec{A} \cdot d\vec{r}_{\text{upper}} \right)
\]  
(2.3)

\[
a_l \rightarrow a_l \exp \left( \frac{iq}{\hbar} \int_{x_l}^{x_r} \vec{A} \cdot d\vec{r}_{\text{lower}} \right)
\]  
(2.4)

The phase factor is irrelevant when the amplitude is taken:

\[
|a_u \psi_u(x_r)|^2 = |a_l \psi_l(x_r)|^2 = \frac{1}{2} P_{\text{trans}}
\]  
(2.5)

However, in the interference term:

\[
a_u^* a_l = a^2 \exp \left( \frac{iq}{\hbar} \oint \vec{A} \cdot d\vec{r} \right)
\]  
(2.6)

Using Stokes' theorem and the definition of the vector potential:

\[
\oint \vec{A} \cdot d\vec{r} = \int_S (\nabla \times \vec{A}) \cdot d\vec{s} = \int \vec{B} \cdot d\vec{s} = \Phi
\]  
(2.7)

The interference term becomes

\[
P_{\text{trans}} = P_{\text{trans}}^{\text{class}} \left[ 1 + \cos \left( \frac{q}{\hbar} \Phi \right) \right]
\]  
(2.8)

Landauer's formula gives the conductance of a 1-D channel as conductance quantum times the probability of transmission [6]:

\[
G = \frac{2e^2}{h} P_{\text{trans}}
\]  
(2.9)

The conductance is therefore periodic in the magnetic flux through the ring, with period equal to the flux quantum, related to the charge \( q \) of the carrier as follows:

\[
\Phi_0 = \frac{\hbar}{q}
\]  
(2.10)

For a quasi-1D ring, there are multiple channels:

\[
G = \frac{2e^2}{h} \sum_i P_{\text{trans}_i}
\]  
(2.11)
Different channels acquire a slightly different phase from the vector potential. As a result, the oscillating quantum correction to the overall classical conductivity is suppressed as the ring becomes thicker. Furthermore, decoherence results in an exponential suppression to the interference pattern and thermal averaging results in linear suppression, such that the oscillation amplitude can be expressed as

$$\Delta G = K \frac{e^2}{h} \frac{L_T}{L} e^{-L/L_\Phi}$$

(2.12)

Here, $L$ is the circumference of the ring, $L_\Phi$ is the decoherence length, $L_T$ is the thermal diffusion length and $K$ is a constant of order unity that incorporates the effect of multiple-channel smearing.

Consider now the interference of two time-reversed paths back at the origin, as shown in figure 2.6. The same arguments as above lead to a quantum correction to the conductivity with period

$$\frac{1}{2} \Phi_0 = \frac{h}{2e},$$

but smaller amplitude than the first harmonic by another factor of $e^{-L/L_\Phi}$.

![Figure 2.6: Interference of the time-reversed paths in the Aharonov-Bohm effect.](image)

### 2.2.2 Conductance fluctuations and weak localization

Consider now transport measurements on a mesoscopic quasi-1D wire. The charge carriers will scatter elastically from impurities, as shown in figure 2.7. At a given magnetic field, a given impurity configuration can result in constructive or destructive interference of two paths, similar to the Aharonov-Bohm effect. Since the set of such configurations for different fields is discrete and randomly distributed, the quantum correction to conductance as a function of applied field appears as reproducible random fluctuations, with an RMS value equal to the universal conductance fluctuations suppressed by decoherence and thermal averaging (in the regime $L_T \ll L_\Phi \ll L$).
as follows \[4\]:

\[
\Delta G \approx C e^2 / h \left( \frac{L_\Phi}{L} \right)^{3/2} \frac{L_T}{L_\Phi} 
\]  

(2.13)

Figure 2.8: Interference of real paths in a disordered conductor leading to conductance fluctuations.

Consider now the interference of time-reversed paths at the origin as the charge carrier scatters elastically from a given configuration of impurities, as shown in figure 2.8 (similar to the second harmonic of the Aharonov-Bohm effect). At zero field, the interference is constructive, and we obtain twice the classical return probability to the origin, meaning suppressed conductivity. As the magnetic field is increased from zero, time-reversal symmetry is broken because of the opposite sign of phase added to each path, and the quantum correction is reduced \[4\]:

\[
\frac{\delta \sigma}{\sigma} = -F_1 e^2 / h \sqrt{D\tau_\Phi} \quad \text{at zero field} 
\]  

(2.14)

\[
\frac{\delta \sigma}{\sigma} = -F_2 e^2 / h \sqrt{D\tau_H} \quad \text{for } \tau_\Phi >> \tau_H 
\]  

(2.15)

\[
\tau_H = 3h^2 / e^2 w^2 H^2 D 
\]  

(2.16)

Here \(F_1\) and \(F_2\) are constants, \(D\) is the diffusion constant, \(w\) is the width of the wire and \(H\) is the magnetic field. One can see that the magnitude of the conductivity dip at zero field is proportional

Figure 2.8: Interference of real paths in a disordered conductor leading to conductance fluctuations.
to the square root of the decoherence time, and that this dip is suppressed by the magnetic field as $1/H$ in the high-field limit.

### 2.2.3 Experiments with metallic mesoscopic structures

The fundamental harmonic of the Aharonov-Bohm oscillations was first observed in mesoscopic gold rings by Webb et al. [7]. In figure 2.9 taken from his original paper, one can clearly see the periodic oscillations in the field-domain data, and the Fourier spectrum shows the $h/e$ and $h/2e$ peaks. This data was taken at 0.01K. The inset shows the gold ring patterned with scanning transmission electron microscope (STEM), approximately 800nm in diameter.

![Figure 2.9: First observation of $h/e$ oscillations by Webb et al [7]. A smaller $h/2e$ peak can also be seen in the Fourier spectrum.](image)

Weak localization (figure 2.10) and conductance fluctuations (figure 2.11) have also been seen in quasi-1D gold and other metallic wires [4]. This has allowed the measurement of the decoherence time in metals as a function of temperature [5]. This dependence from a prototypical gold wire is shown in figure 2.12.
Figure 2.10: Weak anti-localization observed in gold wires [4].

Figure 2.11: Conductance fluctuations observed in gold wires [4].
Figure 2.12: Decoherence time vs. temperature measured from weak anti-localization in a 60µm×60nm×25nm gold wire [8].
Chapter 3

Niche and Experimental Goals

Mesoscopic effects are wiped out by decoherence. The structures must therefore be of length scales smaller than the decoherence length, typically in the tens to hundreds of nanometers at temperatures of a few K. Mesoscopic experiments in metals have relied on lithographic patterning. We are interested in observing these effects in high-$T_c$ superconductors, typically complex oxides, which are not amenable to standard nanofabrication techniques because of strict requirement on the correct stoichiometry and perovskite crystal structure. If nanofabrication is achieved, observation of the Aharonov-Bohm effect will allow to extract the charge of carriers. If the experiment is performed in the pseudogap regime, this would be the first direct verification of the BEC-BCS crossover model for high temperature superconductors, which predicts electron pairing prior to the superconducting transition at $T_c$. Unfortunately, mesoscopic effects in metals have previously been observed at temperatures of the order of 1K, when high-$T_c$ materials are superconducting. To probe the pseudogap means to perform transport measurements on the normal state carriers. This either requires the suppression of $T_c$ to around 1K in high-$T_c$ nanostructure, or using transport measurement techniques that are able to resolve the contribution from the thermally excited normal state quasiparticles only, neither of which has been done before. Therefore, the goals of the proposed experiment are as follows:

1. To develop a reliable nanofabrication technique for complex oxides, namely $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$ (YBCO).

2. To develop methods for doing low temperature transport measurements on the normal state carriers in high-$T_c$ nanostructures.

3. To measure weak localization and/or conductance fluctuations in high-$T_c$ nanowires to
extract the decoherence time $\tau_\phi$ as a function of temperature.

4. To measure the Aharonov-Bohm effect in high-$T_c$ nanorings to extract the charge of the pseudogap quasiparticles.
Chapter 4

Design Concept and Constraints

4.1 Experimental Concept

There are two aspects to the proposed experiment: nanofabrication and measurement.

The idea behind the developed nanofabrication technique is pre-patterning. In other words, the process has been designed such that the growth of the complex oxide material is left to the last step. This avoids any damage that might occur to the sample (in terms of crystal structure or stoichiometry) from any post-processing. Since quasi-1D structures of specific geometries need to be produced, we require three degrees of freedom in defining the structures. Drawing with a focused ion beam (FIB) on an \( \text{SrTiO}_3 \) substrate produces confinement in the plane of the substrate, allowing one to define the shape of the structure, such as an Aharonov-Bohm ring. Confinement in the third direction is achieved by deposition of an epitaxial film of the complex oxide on top of the pattern by using pulsed laser-ablated deposition (PLD).

As mentioned, transport of normal carriers only is of interest, and there are two approaches to doing the measurement. One approach is to underdope the sample in a controlled way as the last step of the fabrication procedure. The difficulty is to control \( T_c \) with a high degree of precision in the 1K region without radically altering the other properties of the material, such as crystal structure. If this is achieved, tradition 4-point impedance measurements with a lock-in amplifier can be done at low frequencies. Controlled underdoping is done by annealing the sample at high temperature and oxygen pressure, then following the phase stability line to room temperature and low pressure, effectively sucking the hole donor oxygen out of the sample. A powder of the complex oxide involved is used to cover the sample to prevent the structure from evaporating at
low pressures.

Underdoping quasi-1D structures of complex oxides is difficult and has not been done before. An alternative is to skip this step and measure optimally-doped structures, which are easy to produce. However, to do the transport measurements at low temperatures, when the decoherence length of normal carriers is long enough, but the sample is superconducting, one must resort to RF frequencies, at which the contribution from the normal carries becomes appreciable. An RF version of the 4-point impedance measurement technique has been developed, in which the 4 contacts are replaced by 4 RF ports. However, extremely high phase sensitivity is required in this case, difficult to achieve with currently available instruments. Another technique based on resonantly removing the superfluid contribution has been developed. It requires high frequencies and high frequency stability, but these are more easily achieved than high phase sensitivity. The difficulty here is in fabricating the reactive component able to match the superfluid kinetic inductance.

4.2 Length Scales

In mesoscopics, quantum corrections are exponentially suppressed by a factor of $e^{-L/L_\Phi}$, as shown in the section on mesoscopic theory. Therefore, for these corrections to be observable, sample size $L$ must be much smaller than the decoherence length $L_\Phi$, related to the decoherence time $\tau_\Phi$ through the diffusion coefficient $D$:

$$L_\Phi = \sqrt{D\tau_\Phi} \quad (4.1)$$

The decoherence time itself is temperature-dependent and has been measured for various metals, such as gold, by Mohanty et al. [8] (refer to section on mesoscopic experiments), giving decoherence lengths on the order of 1 $\mu$m at 4K. The thermal diffusion length, giving a linear suppression by a factor of $L_T/L$, is on the order of 100 nm at 4K. Because mesoscopic transport has not been measured in YBCO or other complex oxides, the decoherence length is not known. Although normal YBCO is a much poorer metal, meaning that more scattering shortens both $L_\Phi$ and $L_T$, 4K measurements on sub-micron structures is a good place to begin. We therefore seek to produce nanowires and nanorings of characteristic length scales of tens to hundreds of nm. If quantum corrections are not observed, next order experiments will be done at lower temperatures, down to tens of mK, accessible using the dilution refrigerator.

Another constraint on the length scale comes from the requirement to suppress flux quantization
through the ring induced by the superconducting electrons in the Aharonov-Bohm experiment. We seek to map out impedance as a function of flux through the ring and detect oscillation harmonics of fundamental h/e periodicity. A macroscopic superconducting ring will only allow superconducting flux through in quanta of h/2e [4], yielding discrete sample points below the Nyquist frequency. However, for rings small enough such that the penetration depth λ is comparable to the width of the ring, flux leaks through, and the sampling of the field in the experiment becomes more frequent. Calculations by Brandt and Clem [9] show this effect (figure 4.1) for various λ/b and a/b ratios, where a is the inner diameter and b is the outer diameter of the ring. By looking at the figure, we want as narrow and as small a ring as possible. At λ/b = 0.1 and a/b = 0.8, 80% of the flux leaks through, which is a reasonable target to set. In that case, a 1 µm diameter ring of 100 nm width would be an appropriate length scale, given the 120 nm penetration depth of YBCO at 4K. Making the ring smaller and narrower will clearly only improve the sampling.

![Figure 4.1: Leakage of field as a fraction of the applied field for different a/b and λ/b ratios.](image)

Figure 4.1: Leakage of field as a fraction of the applied field for different a/b and λ/b ratios.
4.3 Two-Fluid Model and AC Impedance

4.3.1 The two-fluid model

The simplest description of electron behaviour in a solid is the Drude model. The assumptions are that of a free electron gas, namely [12]:

1. The electrons are free (there is no external potential term in the Hamiltonian).
2. The electrons do not interact with each other.
3. Scattering from phonons, other electrons and impurities is instantaneous and can be modeled as a temperature-dependent average scattering time $\tau$.

While a naive picture for understanding the underlying physics, this model is very useful for estimating the conductivity of many materials.

In the case of a superconductor, we consider the two-fluid model, an extension of the Drude model. The assumption is that the normal state electrons and the superconducting electrons form two parallel channels, each obeying the Drude model, with the scattering time for the superconducting channel $\tau_{sc}$ going to infinity. This treatment holds at frequencies much smaller than the frequency of the superconducting energy gap. The normal and superfluid densities can then be assumed to take on their thermal equilibrium values. At frequencies higher than the gap frequency, superconducting electrons can be excited across the energy gap from their ground state, resulting in more complicated dynamics. We begin with the equation of motion of one particle in a free electron gas in an applied external field $\vec{E}$ [1]:

$$m \frac{d\vec{v}}{dt} = -e\vec{E} - \frac{m\vec{v}}{\tau}$$  \hspace{1cm} (4.2)

Using $\vec{J} = -ne\vec{v}$ and taking the Fourier transform with respect to time, we get:

$$\vec{J} = \sigma(0) \frac{1}{1 + i\omega\tau} \vec{E}$$ \hspace{1cm} (4.3)

$$\sigma(0) = \frac{ne^2\tau}{m} \quad \text{(Gaussian units)}$$ \hspace{1cm} (4.4)

This yields the dissipative (real) and reactive (imaginary) components of the frequency-dependent complex conductivity:

$$\sigma(\omega) = \sigma_1(\omega) - i\sigma_2(\omega) = \frac{\sigma(0)}{1 + \omega^2\tau^2} - i \frac{\sigma(0)\omega\tau}{1 + \omega^2\tau^2}$$ \hspace{1cm} (4.5)
For the superconducting (s) channel, we take the limit $\tau_s \to \infty$ (no scattering), which gives:

$$\sigma_s(\omega) = \frac{\pi n_s e^2}{2m} \delta(\omega) - i \frac{n_s e^2}{m\omega}$$  \hspace{1cm} (4.6)

At $\omega = 0$, the conductivity is infinite, as the superfluid is a perfect short at DC. At non-zero frequencies, the finite inertia of the superfluid results in non-dissipative out-of-phase conductivity. For the normal (n) channel, let us assume that $\omega \tau_n << 1$. In other words, the normal electrons scatter many times during one oscillation cycle. In this limit, the normal conductivity becomes:

$$\sigma_n(\omega) = \frac{n_n e^2 \tau_n}{m}$$  \hspace{1cm} (4.7)

Since the normal and superconducting fluids are in parallel, the total conductivity is $\sigma(\omega) = \sigma_n(\omega) + \sigma_s(\omega)$. In transport measurements, one measures the frequency-dependent complex impedance $z(\omega) = \frac{V(\omega)}{I}$ (measuring voltage at fixed current), where $z(\omega) = \frac{1}{\sigma(\omega)} = z_1(\omega) + iz_2(\omega)$. Under the assumptions above, and assuming also that $n_n \approx n_s$, this yields

$$z_1(\omega) \approx \frac{\sigma_n(\omega)}{[\sigma_s(\omega)]^2} = \frac{n_n \tau_n m}{n_s^2 e^2} \omega^2$$  \hspace{1cm} (4.8)

$$z_2(\omega) \approx \frac{1}{[\sigma_s(\omega)]} = \frac{m}{n_s e^2} \omega$$  \hspace{1cm} (4.9)

Since we are interested in the quantum correction to the normal channel conductivity, $z_1$ is what we seek to measure as a function of magnetic field. To extract $\sigma_n$, however, we need an independent measurement of $\sigma_s$, which can be done by measuring $z_2$. In other words, both the real and imaginary parts of the complex impedance of the sample must be measured (or alternatively, the amplitude and the phase) with great precision. At low frequencies, $z_2 >> z_1$ (reactive impedance due to the superfluid dominates) and $\sigma_1$ cannot be resolved. However, $z_2$ scales as $\omega$ and $z_1$ scales as $\omega^2$, so at high enough frequencies, one would expect that $z_1$ and $z_2$ become comparable.

### 4.3.2 Estimates of the AC impedance

Now we estimate the complex impedance of a YBCO ($T_c = 92K$) nanowire of typical dimensions near the limit of the nanofabrication techniques.

For the two-fluid model, we need $n_n$, $n_s$, and $\tau_n$, which are temperature-dependent. Note that $n_n$ here refers to the density of thermally excited quasiparticles, which behave as Drude electrons.
Since first-order experiments are to be done at liquid helium temperature, we take $T = 4\text{K}$. The London equations give the superfluid density as a function of the penetration depth $\lambda(T)$ [1]:

$$\lambda(T) \approx \frac{\lambda(0)}{1 - \left(\frac{T}{T_c}\right)^4} \quad (4.11)$$

$$n_s = \frac{m}{e^2 \lambda^2 \mu_0} \quad (4.10)$$

Empirically the temperature dependence of the penetration depth has been found to behave approximately as follows:

The zero-temperature London penetration depth is experimentally known to be $\lambda(0) \approx 120\text{nm}$ [13]. Using equations 4.11 and 4.10 we get $n_s = 1.9 \times 10^{27}\text{m}^{-3}$. The normal fluid density for a D-wave superconductor at low temperatures is:

$$n_n \approx n_s \frac{T}{T_c} \quad (4.12)$$

In our case, this gives $n_s = 8.7 \times 10^{25}\text{m}^{-3}$.

The scattering rate $1/\tau_n$ has been measured as a function of temperature by Hosseini et al. [14] using microwave spectroscopy on bulk YBCO. If we assume that it is the same for nanoscale YBCO, at 4K we get $\tau_n = 1.9 \times 10^{-11}\text{s}$.

Using equation 4.11 for the normal channel and 4.6 for the superconducting channel, and plugging in the relevant parameters, we plot the frequency dependence of the real and imaginary parts of impedance of a YBCO nanowire 10$\mu$m long and a 100nm x 50nm cross-section. A log-log plot is shown in figure 4.2.

The scaling behaviour is as expected for frequencies much less than 10 GHz and the $\tau_n$ given. At 10 GHz ($\omega \tau_n = 1$), $Z_1$ is still 2 orders of magnitude below $Z_2$. For frequencies much greater than 10 GHz, we observe $Z_2$ level off, while $Z_1$ continues to scale linearly with $\omega$. Physically, one can understand this as follows. At frequencies much greater than the scattering rate, normal electrons do not scatter over one period of motion induced by the electric field and therefore behave as superconducting electrons, contributing only to the imaginary part of the conductivity. As a result, for the given parameters in YBCO at 4K, $Z_2$ never reaches $Z_1$.

One could raise an objection to our treatment of YBCO, a nodal gap superconductor, using the two-fluid model, which, as stated in the previous section, applies only at frequencies below the gap.
frequency. In fact, the non-equilibrium effect could be helpful, since for higher and higher frequencies, the conductivity approaches that of the normal state sample, an effect that was measured by Ginsberg and Tinkham \cite{Ginsberg:1964p7027} in type I superconductors. For these superconductors, the gap is in the infrared, but for YBCO, where the gap is zero along certain directions, the effect might take place at much lower frequencies. However, this has been ruled out by Hosseini \textit{et al.} \cite{Hosseini:2016p12771} in bulk YBCO and Nguyen \textit{et al.} \cite{Nguyen:2016p12147} in YBCO thin films by surface resistance measurements up to tens of GHz. The frequency dependence of impedance is found to be well fit by the two-fluid model and the values agree very well with those estimated above.

### 4.4 Non-Equilibrium Effects.

Two non-equilibrium effects might be of concern when performing high-frequency transport measurements. The first is the suppression of the interference pattern at frequencies high than the inverse transit time of the electrons through the ring:
\[ f > \tau_L^{-1} = \frac{D}{I^2} \quad (4.13) \]

In this regime, electron paths do not close around the ring and therefore perhaps cannot interfere. This has been studies by Pieper and Price [10] in mesoscopic silver rings. They have found that the Aharonov-Bohm oscillation amplitude in the real part of the impedance remains constant from 0 to 1.2 GHz. For \( \omega \tau_L \geq 1 \), they also observe the oscillations in the imaginary part of impedance. In our experiment, the real part of impedance \( Z_1 \) is of interest and based on the experiments by Pieper and Price, we do not expect the quantum correction to be degraded by frequencies up to several GHz.

Another concern is the maximum current that can be applied. Firstly, it must not exceed the critical current, which is \( 10^7 A/cm^2 \) for YBCO at 4K, which translates to 0.5 mA for a cross-section of 100 nm by 50 nm. Secondly, electron heating must be avoided. In the experiments by Pieper and Price, electron heating began to affect the amplitude of the oscillations at dissipated RF powers higher than 70 pW [10]. For \( Z_1 = 1.3 m\Omega \) at 1 GHz in the YBCO nanowire, this corresponds to a current of 0.23 mA, in the same order of magnitude as the critical current. Therefore, we stipulate that the current used in the transport measurements do not exceed 100 \( \mu \)A.
Chapter 5

Nanofabrication

5.1 Substrate Patterning

The shape of the structure is defined on the surface of an $SrTiO_3$ (STO) substrate with $\overline{001}_\parallel$ crystal orientation by digging tranches with a focused ion beam (FIB). This step of the process is done by our collaborators from Academia Sinica in Taiwan. Figure 5.1 shows a scanning electron microscope (SEM) image of some of the first patterns of an Aharonov-Bohm ring defined this way [11].

![SEM images of first micron-sized Aharonov-Bohm rings produced with FIB](image)

Figure 5.1: SEM images of first micron-sized Aharonov-Bohm rings produced with FIB [11].

A problem has been found with this first-order technique of digging straight trenches. While the heat from FIB destroys the crystallinity of the substrate, ideally preventing epitaxial growth of YBCO in the trenches and on the walls, YBCO is grown at high temperatures, which cause recrystallization of the substrate [17]. As a result, shorts are possible across the trenches, destroying
the defined topology of the sample.

An alternative patterning technique has been designed. It involves off-axis FIB and the process is illustrated in figure 5.2. When YBCO is deposited, even if all of it is epitaxial, it will not connect across the trenches if they are deep enough and at a shallow enough angle. Note that the Cr layer is part of the standard FIB procedure. It is necessary to conduct away the charge of the ions from the beam deposited on the substrate.

Figure 5.2: Trenches dug in STO at an angle with off-axis FIB [11].

Figure ?? shows a transmission electron microscope (TEM) cross-sectional image of a nanowire defined in STO using off-axis FIB [11]. The Cr layer has not been removed yet. The width of the wire shown is 466 nm, but wires as narrow as 200 nm have also been successfully made using this technique.

Using this technique introduces one major difficulty. To pattern trenches running in perpendicular directions, the FIB beam needs to be tilted about different axes. In reality, it is the stage where the sample sits that gets tilted, introducing an error in the position of the beam on the substrate of the order of 5 µm, which is much greater than the typical sample size.

For patterning nanowires, this problem can be solved by careful design of the geometry of the wire and contact pads and of the sequence of FIB steps. One method, illustrated in figure 5.4, has been successfully implemented. The pattern is shown on the SEM image in figure 5.5 [11].

First, the stage is tilted about the x-axis and the nanowire is drawn in the fine setting, but with the sides offset by 50 microns as shown (step 1). Then the rest of the horizontal lines of the
Figure 5.3: Cross-sectional TEM image of a nanowire in STO defined using off-axis FIB [11].

Figure 5.4: Procedure for defining a nanowire in STO using off-axis FIB.
pattern are drawn in the coarse setting. Then the stage is rotated back and tilted about the y-axis. The vertical lines of the pattern are drawn, including the middle lines connecting onto the wire. This way, the large offsets allow for a 5 micron error in positioning when the tilt is changed.

The same approach does not work for more complicated structures such as rings. In that case, landmarks crosses are drawn, such as the ones shown in figure 5.4, which allow to re-position the beam by imaging with it after the tilt axis is changed.

5.2 YBCO Thin Film Deposition

After nanopatterning the substrate, a thin epitaxial film of YBCO is deposited using pulsed laser-ablated deposition (PLD). The set-up is shown in figure 5.6. A KrF excimer laser is used to strike a target of YBCO sitting in vacuum across from the substrate attached to a heater plate. The laser energy is transferred to the kinetic energy of YBCO nanoparticles ejected from the target and deposited on the substrate.

The controllable parameters in the growth process are oxygen pressure, substrate temperature, length of deposition time, laser energy per pulse, and pulse frequency. The recipe for growing optimally doped YBCO has been developed. First, the chamber is pumped down to $10^{-6}$ Torr. The substrate is heated to 760°C and molecular oxygen pressure is stabilized at 250mTorr. Ablation is performed at a laser energy of 480mJ per pulse, at 5 Hz pulse frequency, which gives a film growth rate of 8nm/min. After ablation, oxygen pressure is raised to 760 Torr and the sample is cooled down to 300°C in 45 minutes, which freezes in the oxygen configuration.

A YBCO nanowire was deposited on the patterned substrate shown in figure 5.5. Its resistance
vs. temperature was measured using the low-frequency 4-point lock-in technique. Figure 5.7 shows a sharp superconducting transition at 90K and nice metallic behaviour above $T_c$.

A cross-sectional TEM image of the same wire was taken; it is shown in figure 5.8. Careful analysis of the image showed that the tilted trenches managed to separate the YBCO regions [11].
Figure 5.7: Resistance vs. temperature of YBCO nanowire fabricated by off-axis FIB and PLD.

Figure 5.8: Cross-sectional TEM image of the nanowire fabricated using tilted trenches [11].
5.3 Underdoping YBCO Films

Thin films of YBCO have been underdoped down to oxygen content \( x_n = 6.6 \), corresponding to \( T_c = 25K \), by Osquiguil et al. [18]. Resistance versus temperature for a 200nm thick film at different doping levels is shown in figure 5.9.

![Normalized resistance vs. temperature for a 200nm thick YBCO film](image)

Figure 5.9: Normalized resistance vs. temperature for a 200nm thick YBCO film [18].

Figure 5.10 shows \( T_c \) vs. \( x_n \) for bulk YBCO and for films made by Osquiguil et al. [18]. One can observe that for films, \( T_c \) is a linear function of \( x_n \). If the doping could be lowered to 6.4, \( T_c \) very close to zero could be achieved. This is what we seek to do here, by imitating the procedure used by Osquiguil et al. [18].

To do this, a set-up has been built for re-annealing YBCO films under precise control of temperature and oxygen pressure. It is shown in figure 5.11. The furnace is temperature-programmable. The two needle valves allow to control the pumping rate and the flow of oxygen, giving the capability to set the desired oxygen pressure in the quartz tube between atmospheric and 1 mTorr. The pressure is monitored on both sides by TC pressure gauges. The sample itself sits in the quartz tube in the middle of the furnace, covered with YBCO sand in order to maintain high vapour pressure of the cations above the film, thus preventing it from evaporating at low pressures.

To underdope a sample, one starts with an optimally doped sample at a high temperature (depending on the final doping level designed) and atmospheric pressure, then slowly lowers the
Figure 5.10: $T_c$ vs. $x_n$ for bulk YBCO (empty points) and for films made by Osquiguil et al. [18] (filled points).

temperature and pressure following the stability line in the pressure-temperature phase diagram until room temperature is reached. The phase diagram has been measured by a number of groups, such as Kim and Gaskell [19]. Osquiguil et al. had to go down to 70 mTorr oxygen pressures. For lower doping levels, lower pressures might be required. The set-up in figure 5.11 has been tested down to 1 mTorr.
5.4 Contact Making

Making contact to the four measurement leads is an important element for successful measurements. Areas for the pads are defined using rough FIB, as shown in figure 5.4. Silver pads are deposited in those areas by means of magnetron sputtering. The rest of the sample must be masked. Masks are fabricated using a laser micromachining facility, shown in figure 5.12. A typical mask is shown in figure 5.13. The mask is aligned with the FIB pattern using a microscope and a specially designed mask aligner (figure 5.14), which holds the mask above the sample on top of two whiskers and is able to move the mask in three directions by means of micrometers. A microscope image of silver pads deposited onto the FIB pattern is shown in figure 5.15.
Figure 5.12: Micromachining set-up. A tightly focused laser beam (right) cuts into sheet metal, sitting on a computer-controlled movable stage.

Figure 5.13: Sputtering mask fabricated for the off-axis FIB nanowire design.
Figure 5.14: Mask aligner with 3 degrees of freedom for positioning.

Figure 5.15: Silver pads (seen as dark shadows) deposited on patterned YBCO.
Chapter 6

Measurement Techniques

6.1 Low Frequency 4-Point Technique

The 4-point technique is a standard way to measure low impedances, configured to remove the effect of measurement leads and of contact resistance. The idea is illustrated in figure 6.1.

Figure 6.1: 4-point impedance measurement configuration.

A current source drives current through the sample via one set of contacts. The voltage measurement is done via a second set of contacts. Since the input impedance of the voltage measurement circuit is very high (ideally, infinity), no current flows through the voltage contacts. As a result, there is no voltage drop across the contact interface or across the leads. In practice, the current source is typically and AC voltage source with a large series resistor that sets the AC current level. Voltage is measured at the source frequency by amplifying the signal and mixing it with part of the source signal, then low-pass filtering. The instrument that does this is called a lock-in amplifier. The set-up is illustrated in more detail in figure 6.2. There are three purposes of doing the measurement at AC: to reject noise at other frequencies, particularly to reduce the 1/f noise, to average over any thermal gradients that might exist across the sample, and to get a complex
impedance for a range of low frequencies.

Figure 6.2: 4-point measurement configuration with a lock-in amplifier.

6.2 RF 4-Point Technique

In view of the estimated values of \(Z_1\) and \(Z_2\) for superconducting YBCO at 4K (see figure 4.2), we want to do transport measurements at frequencies in the GHz range. Furthermore, this has to be done with extreme phase sensitivity. Leads no longer work at high frequencies, so one has to work with transmission lines (TL), each TL representing one port. One can then think of extending the 4-point impedance measurement technique to the 4-port RF impedance measurement technique as shown in figure 6.3. Note that each TL must be matched to 50\(\Omega\) on the side of the sample and reference (whose impedance is negligible compared to 50\(\Omega\)).

Consider the superconducting sample of interest (blue) and a normal wire for reference (orange). The sample and the reference are driven at the same frequency and amplitude, but with opposite polarity. The impedance of the reference must be chosen to be comparable to \(Z_2\) of the sample. The signal from the reference is then attenuated and phase shifted around 90° with high precision, then
Figure 6.3: RF balanced 4-point measurement configuration.

subtracted from the signal across the sample by the differential broadband amplifier. The amplified
difference then goes to the high frequency lock-in detection scheme. By tweaking the variable
attenuator and phase shifter one can achieve a purely real signal at the output, corresponding to
$Z_1$; then $Z_2$ has been canceled out by the reference.

The specifications of this system are quite challenging. Consider trying to measure a quantum
correction to $Z_1$, which will typically be $Z_1/1000$ [4]. Assuming we can achieve precision in the
phase and amplitude of the read signal to be $\Delta \theta = 0.001^\circ$ and $\Delta |Z|/Z = 0.001$. Then the relative
error in $Z_1$ versus frequency is shown in figure 6.4. The resolution in $Z_1$ barely makes it to the 1
/ 1000 level required to see the quantum corrections, for a limited set of frequencies in the several
GHz range. Therefore, we system must satisfy:

$$\Delta \theta < 0.001^\circ \quad (6.1)$$

$$\frac{\Delta |Z|}{Z} < 0.001 \quad (6.2)$$

The variable phase shifter therefore must have at 0.001 degree phase resolution, and the dif-
fferential amplifier must not introduce any spurious phases. Consider now a current of 100 $\mu$A
at 1 GHz. The voltage $V_1 = IZ_1 = 0.13\mu V$, so the required resolution is 0.13 nV. Furthermore,
the common mode rejection ratio of the differential amplifier must be at least 53 dB at the GHz
frequencies.
6.3 Resonant Technique

For the two-fluid model with conductivities given by equations 4.6 and 4.7 one can construct an equivalent circuit (figure 6.5) composed of a resistance in parallel with an inductance (the kinetic inductance of the superfluid), given by:

\[ R = \frac{m}{n_s e^2 \tau_n} \frac{l}{A} = \frac{1}{\sigma_n} \frac{l}{A} \]  \hspace{1cm} (6.3)

\[ L_k = \frac{m}{n_s e^2} \frac{l}{A} \]  \hspace{1cm} (6.4)

We are interested in the value of \( R \), since it is inversely proportional to \( \sigma_n \). However, at low frequencies \( L_k \) acts as a short. Going to higher frequencies allows one to resolve the value of \( R \) better and better. Unfortunately, as we have already seen, even at the highest frequencies at which this approximation holds, current flows preferentially through the \( L_k \) channel by several orders of magnitude. Resorting to RF circuit theory, one can get rid of the reactive component due to \( L_k \) through resonance, by matching it with an external capacitance \( C \), as shown in figure 6.6.

The equivalent impedance is given by equation ?? and equals \( R \) at the resonance frequency given by equation 6.6.
By choosing a correct aspect ratio for the nanosample, one can adjust $R$ to be close to 50 Ω. At this nominal impedance, a single-port reflection measurement with a vector network analyzer can yield a very sensitive complex impedance measurement. If one sweeps the frequency and finds the resonance, then measures the impedance exactly on resonance, one can get a very precise reading of the normal channel conductivity.

As a suitable example, let us go back to the $1 \mu m \times 100 \text{nm} \times 50 \text{nm}$ YBCO nanowire. It has the equivalent values $R = 50 \Omega$ and $L_k = 3 \text{pH}$. A plot of the magnitude and phase of the equivalent impedance as a function of frequency is shown in figure 6.7. The resonance is at 1 GHz and is extremely narrow. In fact, the width of the resonant peak (full width at half power) is given by equation ??, which gives a 60 kHz resonance width in our case.
There are two challenges in implementing this technique. The first is doing a reflection measurement at GHz frequencies with a source that is stable within tens of kHz. Furthermore, the measurement must be done around 50 pW incident power and have a sensitivity of 1/1000, or 50 mΩ. An instrument for doing just that has been designed and used by Pieper and Price [20], and used in their work on high-frequency mesoscopic transport [10]. A schematic is shown in figure 6.8. The accessible frequency range is 0.3-1.0 GHz, and the impedance measurement resolution of a nominally 50 mΩ load is $10^{-5}$, at 50 pW incident power. This instrument will be rebuilt for our experiments.

The second challenge is fabricating a relatively large capacitance ($C = 300$ nF assuming $L_k = 3$ pH to get $\omega_0 = 1$ GHz) with virtually no parasitic inductances between it and the sample. The parasitic inductances must be smaller than $L_k$, and since 3 pH is extremely tiny, it is very difficult to connect this capacitance to the sample because all wires carry geometric inductance.

$$FWHP = \frac{L_k}{R} \omega_0^2$$

(6.7)
A suggested procedure for fabricating a 300 nF capacitor on the sample to avoid parasitic leads is shown in figure 6.9. This is a multilayer PLD process that allows to define a parallel-plate capacitor, one plate being the YBCO layer that makes up the nano-sample, and the other plate being a layer of niobium. The dielectric is amorphous STO deposited using the same PLD process. Note that masking during the PLD process is required and that the outline of the bottom capacitor plate is defined using FIB.
Figure 6.9: Steps for fabricating an on-chip 300 nF capacitor using multi-step PLD process.
6.4 Cryomagnetic Facilities

The transport measurements need to be done at low temperatures and as a function of magnetic field. A 4K cryostat with a 4T magnet is available for first-order measurements. If subsequent measurements need to be done at even lower temperatures, a He-3 cryostat and a dilution refrigerator are also available. For 4K measurements, a special RF probe has been assembled, as shown in figure 6.10. Stainless steel coaxial cables are installed in it, acting as 50Ω transmission lines for RF measurements. Stainless steel is used for reducing the heat load on the sample because stainless steel is a poor conductor of heat.

Figure 6.10: The 4K cryostat with a 9T magnet and a high frequency probe.
Chapter 7

Conclusion

Striving towards the goal of measuring mesoscopic transport of normal state carriers in high-$T_c$ cuprate superconductors, and in particular using the Aharonov-Bohm effect to probe the pseudogap state, significant progress has been made in developing necessary experimental techniques.

On the nanofabrication side, a YBCO nanowire with contact pads has been successfully fabricated and characterized by resistance vs. temperature and cross-sectional TEM. To fabricated the nanowire, it was first defined on an STO substrate using off-axis FIB, then a thin epitaxial layer of YBCO was deposited using PLD. Contact silver pads were sputtered with the help of a micromachined mask. The next step on this front of the experimental development is to nanofabricate and characterize YBCO Aharonov-Bohm rings. Another direction which will be pursued is to underdope the fabricated nano-structures to sub-Kelvin transition temperatures by means of the underdoping apparatus, which has been designed and assembled.

On the measurement side, two high-frequency transport techniques have been designed and are yet to be implemented and tested on the actual nanostructures, at low temperatures and in magnetic field. One technique is an extension of the standard low-frequency 4-point lock-in impedance measurement to RF frequencies. It is a 4-port bipolar balanced measurement of a sample with respect to a reference, with precise phase and amplitude control of the reference signal, which allows to distinguish the real part of the sample impedance only.

The second technique is based on resonantly matching out the kinetic inductance due to the superfluid by fabricating an on-chip capacitor, then measuring the normal channel impedance at the resonance frequency. A reflection measurement using a homodyne reflectometer that will be rebuilt after that of Pieper and Price [20], can then achieve the necessary frequency, power level
and measurement sensitivity to observe quantum correction to the normal fluid impedance only. The challenge yet to be overcome on this front is fabrication of such capacitor with parasitic inductances much smaller than the kinetic inductance of the superfluid.

On the cryomagnetics front, cryogenic facilities with 4 K and 10 mK temperature capabilities and 9 T magnetic field capabilities are in place, and an RF low-temperature transport measurement probe has been assembled and tested.

While the end goal of this experiment is still some ways in the future, this thesis demonstrates the feasibility and scientific worthiness of it, and lays down baseline experimental development work on which to build in pursuing it further.
Bibliography


