SUPERFLUIDITY IN ULTRATHIN CUPRATES AND NIOBIUM/FERROMAGNETIC HETEROSTRUCTURES

DISSERTATION

Presented in Partial Fulfillment of the Requirements for the Degree Doctor of Philosophy in the Graduate School of The Ohio State University

By

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2015

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The thrust of this dissertation involves superconductivity, or the lack of any resistance in an electric current, which has been of great interest since its discovery in 1911. Contemporary research has turned to ever more complex systems and has found fascinating interactions and new sources of the phenomenon. Our contribution comes from superfluid density, $\lambda^{-2}(0)$, measurements of Superconducting/Ferromagnetic (S/F) thin film bilayers and trilayers, as well as the high temperature superconducting ceramic cuprate Bi$_2$Sr$_2$CaCu$_2$O$_{8+\delta}$ (BSCCO or Bi-2212) films we grow by Pulsed Laser Deposition (PLD).

Superconductivity and ferromagnetism are opposing phenomena, resulting in complex interaction. We find the ferromagnet in S/F bilayers has a surprisingly small exchange energy, $E_{ex}$, and large interface resistance, $R_b$. This contradicts conventional understanding of these systems, but our study of the theory shows that it is entirely consistent. The theory proposed by Houzet and Meyer (H-M) expects normalized superfluid density, $\lambda^{-2}(0)/\lambda_0^{-2}$, as a function of normalized transition temperature, $T_c/T_{c0}$, to follow a narrow band, where $\lambda_0^{-2}$ and $T_{c0}$ are the parameters for an S monolayer. We show the data agree well with H-M theory and offer a plausible explanations for the exception.

When a second superconducting layer is added to make S/F/S trilayers, a second superconducting transition results. The second transition is visible by a kink in superfluid density and allows us to determine the contribution to $\lambda^{-2}(0)$ from each
S layer. We find strong evidence for the oscillation of the order parameter inside F, named the FFLO state after the authors who proposed it. We also find that $\lambda^{-2}(0)$ is suppressed much more than expected, which is possibly the result of the order parameter being of opposite sign in opposing S layers of the trilayer, known as a $\pi$-junction.

BSCCO films are exceedingly difficult to grow with PLD and before now there was little success growing very thin high quality films. We employ a novel growth technique, and substrates with better lattice matching, to construct the first BSCCO films with single-digit unit cell thicknesses using PLD. With our new thin films, we reconcile the previous factor-of-three difference between the scaling of two unit cell YBCO films and thick BSCCO, as well as detect the presence of a downturn in $\lambda^{-2}(T)$ indicative of a Kosterlitz-Thouless transition corresponding to the single CuO$_2$ bilayer, despite films consisting of several bilayers.
To my wife, Alice, my daughter, Delilah, and our future children.
This dissertation would not have been possible without the guidance and support of my advisor, Dr. Thomas R. Lemberger. I am grateful for his patience, understanding, and encouragement. I would also like to thank my candidacy and dissertation committee members, Dr. Louis F. DiMauro, Dr. Fengyuan Yang, and Dr. David Stroud. Additionally, I would like to thank previous members Dr. Julia Meyer and Dr. Nandini Trivedi.

My success in graduate school would have not been possible without the love and support of my wife Alice and the joy that is my daughter, Delilah. I also would not have reached this point if it wasn’t for Stanley Steers, who helped me greatly in the construction of this document, and Christopher Porter, whom I have known since the start of my journey at Ohio State.
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Jie Yong, MJ Hinton, A McCray, M Randeria, M Naamneh, A Kanigel, TR Lemberger ”Evidence of two-dimensional quantum critical behavior in the superfluid density of extremely underdoped Bi$_2$Sr$_2$CaCu$_2$O$_{8+x}$” Physical Review B 85(18): 180507, May 2012


**Fields of Study**

Major Field: Physics: Superconductivity
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Chapter 1

INTRODUCTION

1.1 Fundamentals of Superconductivity

Superconductors (S) manifest several unique properties, the most famous and useful of which is allowing the otherwise dissipative effects of current flow to vanish, meaning there is no resistance or energy loss that is typically found in classic conductivity. This is the phenomenon that indicated the discovery of superconductivity in 1911 by H. Kamerlingh Onnes when he supercooled mercury (Hg) with liquid helium (He)[1]. Since then, most of the early work and even much contemporary work involves simply measuring the resistivity (Figure 1.1) of superconductors and superconducting complexes. Resistivity measurements yield a superconducting transition temperature, $T_c$, as well as a residual resistivity, $\rho_0$, but contain little more information. Another major quality of superconductors is the Meissner effect, discovered in 1933 by Walther Meissner and Robert Ochsenfeld. The Meissner effect, shown in Figure 1.2, is the expulsion of magnetic field from inside the superconductor. A superconductor differs from the idea of a perfect conductor in that the latter is required to perpetuate a constant internal magnetic field no matter the change in the external field. The superconductor, through the Meissner Effect, is a perfect diamagnet. Any applied field is met with an induced field in the opposing direction through the induction of currents on the superconducting surface. This is of particular interest because it is
the effect we exploit in this work to measure the superfluid density and thereby probe the phenomenon between $T_c$ and 0 K.

Figure 1.1: Example of resistivity, the most common type of measurement done on superconductors. Shown here, the high-$T_c$ superconductor YBCO. At $T_c$, there is an abrupt drop to zero resistivity, which is unchanging for lower temperatures.

Since the discovery of superconductivity, many superconductors have been found, including several elements, of which Niobium (Nb) has the highest $T_c$ at 9.25 K. Much of the new physics contained in this dissertation involves Nb and Ferromagnetic (F) thin films in bilayer (F/Nb) and trilayer (Nb/F/Nb) form. The rest of the dissertation deals with the so-called high $T_c$ superconductors or “cuprates,” the most famous of which is yttrium-barium-copper-oxide, YBa$_2$Cu$_3$O$_{7-\delta}$ (YBCO). The cuprate involved in this work is Bi$_2$Sr$_2$CaCu$_2$O$_{8+\delta}$ (BSCCO or Bi-2212), another well studied cuprate
Figure 1.2: Depiction of the Meissner Effect. Magnetic field passes through the superconductor above $T_c$ (left). Below $T_c$, the magnetic field is repulsed (right), (from Wikipedia).

of which there is still much to learn. Bi-2212 is one of the most anisotropic cuprates—a crystal structure term for the distance between superconducting CuO$_2$ bilayers—while YBCO is the least anisotropic which allow us to compare these extremes.

1.2 BCS Theory

The first microscopic theory of superconductivity was completed in 1957 by Bardeen, Cooper, and Schrieffer in their seminal paper[2]. They used the hypothesis, and now well known fact, that superconductivity arises from electron-phonon interaction and calculated the resultant electron-electron interaction to show that the Fermi sea is unstable against the formation of at least one bound pair, as long as the interaction was attractive, even if of arbitrary small strength. This led to the formulation of Cooper pairs, the name given to the bound electrons that transport the superconducting current.
This is a quantum effect, but there is a way to imagine this in classical terms. A negatively charged electron sitting in a lattice of positively charged nuclei will attract those nuclei towards it through the Coulomb force. The higher concentration of positive charge around the electron will screen the repulsion from surrounding electrons. The altered lattice has the effect of attracting a long distance electron (typically thousands of Å). This second electron acts similarly, creating a mutual attractive force, allowing the electrons to move together as one entity (See Fig. 1.3).

Figure 1.3: The electrons (red) attract the lattice ions (yellow) which screens surrounding electrons, but creates an effective attractive force with another long distance electron, creating a Cooper Pair. The up and down arrows represent spin (Reproduced from J. Draskovic Thesis).

In BCS theory, the electrons in a Cooper pair have equal and opposite momenta with respect to their center of mass. Given the electron’s spin of 1/2, together they act as a boson with integer spin, and thus do not follow the Pauli exclusion principal. As a consequence, they exist in a Bose-Einstein condensate, with all Cooper Pairs occupying the same energy state. The condensed electrons leave behind a gap, \( \Delta \), in the available energy states which allows superconducting current to flow without
resistance. The gap is represented by:

\[ \Delta = \frac{\hbar \omega_c}{\sinh[1/N(0)V]} \approx \hbar \omega_c e^{-1/N(0)V} \]  

(1.1)

where the cutoff frequency, \( \omega_c \), is the Debye frequency, \( N(0) \) is the density of states at the Fermi surface when \( T = 0 \) K, and \( V \) is the attractive potential between electrons that gives rise to Cooper pairs. The gap is an important aspect of superconductivity. The full binding energy is \( 2\Delta \), because \( 2\Delta \) is required to excite two electrons above the gap and break the pair, and extends one \( \Delta \) above the Fermi level and one below. The Cooper pair electrons reside at the edge of the Fermi sea below the gap. For most conventional superconductors, \( \Delta \) is approximately 1 meV (\( \approx 10K \)), which is greater than most of the thermal energy vibrations at low \( T \). Since all excited states within the gap are disallowed, thermal energy is not able to excite most Cooper pairs above the gap, and thus current is able to flow without resistance as the Cooper pairs short circuit the normal conducting electrons to carry the current. At \( T = 0 \) K, BCS theory predicts a value for the gap of \( \Delta(0) = 1.76k_B T_c \) (meaning the full \( 2\Delta \) gap is \( 3.5k_B T_c \)), which is a good fit for most superconductors. This is one of the biggest successes of BCS theory.

The temperature dependence of the gap is flat near \( T = 0 \) K and follows a characteristic mean field theory near \( T_c \):

\[ \frac{\Delta(T)}{\Delta(0)} = 1.74 \left( 1 - \frac{T}{T_c} \right)^{1/2}, T \approx T_c \]  

(1.2)

As a function of normalized or “reduced” temperature, the gap looks like this (See Fig. 1.4):
where this plot is well approximated by:

$$\frac{\Delta(T)}{\Delta(0)} \approx \left[ \cos \left( \frac{\pi T^2}{2T_c^2} \right) \right]^{1/2}$$

(1.3)

which will become important as we talk about superfluid density in the next section. ($T_c$ is the superconducting transition temperature.)

Another important aspect of BCS theory is in the characteristic length called the coherence length, $\xi_0$. $\xi_0$ is the radius of the normal cores of the phenomenon of vortices, which are circulating superconducting currents surrounding a magnetic flux quantum that exist in Type-II superconductors. Type-II superconductors are characterized by a penetration depth, $\lambda$, that is much larger than its coherence length.
(\lambda \gg \xi_0). \lambda is the distance where an applied magnetic field penetrates the surface of S until it has dropped to 1/e of the external value. All the superconductors discussed here are Type-II superconductors. Additionally, \( \xi_0 \) represents the distance over which the superconducting order parameter can change appreciably in real space. \( \xi_0 \) is also important because, along with the mean free path, \( \ell \), it helps determine if we are in the “clean” (\( \xi_0 \gg \ell \)) or “dirty” (\( \xi_0 \ll \ell \)) limit of BCS. The temperature dependent superfluid density BCS fits we employ assume we are in the dirty limit.

### 1.3 Superfluid Density

The measurement most frequently discussed in this thesis is the superfluid density. This measurement will be carried out on a variety of thin superconducting films, Nb/F bilayers, Nb/F/Nb trilayers, and high \( T_c \) cuprates. Superfluid density, \( n_s \), is the number of superconducting electrons per unit volume and is proportional to the inverse-squared penetration depth \( n_s = \mu_0 \lambda^{-2}/mc \), and

\[
\lambda^{-2}(0) = \frac{\pi \mu_0 \Delta_0(0)}{\hbar \rho_0} \tag{1.4}
\]

where \( \lambda \) is the penetration depth, \( \mu_0 \) is permeability of vacuum, and \( \rho_0 \) is the residual resistivity of the film. We will use \( \lambda^{-2}(T) \) to represent \( n_s \) for the remainder of this thesis. As a superconductor becomes more superconducting it screens more magnetic field, decreasing the penetration depth, and increasing the measured superfluid density. Superfluid density decreases as we approach \( T_c \). The temperature dependence of \( \lambda^{-2}(T) \) in thin films is well fitted by dirty limit BCS theory:

\[
\frac{\lambda^{-2}(T)}{\lambda^{-2}(0)} = \frac{\Delta(T)}{\Delta(0)} \tanh \left[ \frac{\Delta(T)}{2k_B T} \right] \tag{1.5}
\]

where \( \frac{\lambda^{-2}(T)}{\lambda^{-2}(0)} \) was shown in Eq. 1.3. In the low frequency (\( \omega \ll \Delta/\hbar \)), thin-film limit (\( d \ll \xi \)), the areal superfluid density is proportional to the imaginary part, \( Y_2(T) \), of
the sheet conductivity $Y(T)$. Sheet conductivity is the local conductivity, $\sigma_2(\omega, z, T)$, integrated through the thickness of the superconducting film:

$$Y_2(\omega, T) = \int_0^{d_S} dz \, \sigma_2(\omega, z, T) \approx d_S \sigma_2$$  \hspace{1cm} (1.6)

for a film of thickness $d_S$. The $z$-axis is perpendicular to the bilayer for the films in this work. We directly measure $Y_2$, which when divided by $d_S$, is proportional to $\lambda^{-2}(T)$:

$$\lambda^{-2} \equiv \mu_0 \omega Y_2 / d_S$$  \hspace{1cm} (1.7)

We stay well below the gap energy of the system by measuring at frequencies far below the $T_c$ of the films by using very low AC current frequencies of $\omega = 50$ kHz (energy equivalent of 400 nK), placing us, for our purposes, in the DC regime.

### 1.4 Two Coil Measurement

The two coil mutual inductance measurement is what we use to measure $\lambda^{-2}(T)$. Fig. 1.5 is a diagram of the two coil apparatus. AC current is supplied to the drive coil ($I_{drive}$), which induces a magnetic field inside the coil. When the film, placed in between the drive and pickup coils, goes superconducting, the Meissner effect expels most of the field from the film. The field that is not screened induces a voltage in the pickup coil ($V_{pickup}$). Due to the thinness of the film, the number of field lines turning around inside the film are negligible. As $T$ decreases, there are more superconducting pairs to contribute to superconducting currents, which expel more field and is recorded as an ever smaller effective mutual induction, $M$, between the drive and pickup coils:

$$M \equiv \frac{V_{pickup}}{\omega I_{drive}}$$  \hspace{1cm} (1.8)
where \( \omega = 2\pi f \) is the frequency of the applied magnetic field.

![Diagram of two coil apparatus showing configuration of drive and pickup coils. Coils are each about 2 mm in diameter on opposite sides of the superconducting film.](image)

Figure 1.5: Two coil apparatus showing configuration of drive and pickup coils. Coils are each about 2 mm in diameter on opposite sides of the superconducting film.

Even with perfect screening, some field will travel around the film. We find this “zero position” by using superconducting lead foil to maximize screening and then subtract the residual value from other mutual induction data, which allows us to assume our film has an infinite radius, simplifying the math. Our raw data are analyzed with a program that generates look-up tables based on the geometry of the coils to turn mutual induction values into the real and imaginary components of complex conductivity. As discussed in the last section, the imaginary conductivity is proportional to the superfluid density and related to the reduction in real mutual induction. When we talk about real and imaginary components, “real” refers to the correlation we would expect in typical electronic responses while “imaginary” is 90° out of phase to this expectation. As a result, superconductivity responds to voltage differently than resistive conductivity by trailing the change in voltage.

A raw data plot is shown in Fig. 1.6. Real mutual induction, \( M_{Re} \), falls by two orders of magnitude at \( T_c \), followed by a gradual reduction thereafter. There is a
negative spike in imaginary mutual induction, $M_{Im}$ as the film transitions. These data are converted into real, $\sigma_1$, and imaginary, $\sigma_2$, conductivities. $\sigma_2$ corresponds to $\lambda^{-2}(T)$ while $\sigma_1$ spikes at the transition due to a crossover of real and imaginary impedance. The resulting superfluid density is shown in Fig. 1.7. The peak in $\sigma_1$ is a reasonable indicator of $T_c$ and the width of the peak gives an idea of the homogeneity of the sample with smaller width indicating greater homogeneity.
1.5 Proximity Effect in Superconducting/Ferromagnetic Systems

Superconductivity most commonly exists in s-wave Cooper Pairs with anti-symmetric electron spins and is inherently incompatible with ferromagnetism, which consists of an array of aligned spins. This competition between states is unfavorable to superconductivity. When a superconductor is placed near a ferromagnet, Cooper pairs travel into the inhospitable state and induce superconductivity in the ferromagnet. Over time, the Cooper Pairs dephase until pairbreaking occurs, which destroys the pair and reduces the amount available in the superconductor, thereby shrinking the gap and lowering $T_c$ and $\lambda^{-2}(T)$. The proximity effect in a superconducting/normal metal system has a similar pair breaking effect but requires thousands of Angstroms
of normal metal to saturate, versus about 100 Å with a ferromagnet. It is possible for the Cooper Pairs to return unbroken, either from reflection off the back of the non-superconductor, off of impurities, or from spin-orbit scattering. Interestingly, we will see that at full saturation, the ferromagnet can send back more pairs than a non-ferromagnetic metal.

Figure 1.8: Change in the order parameter, $\Psi$, inside a normal metal (a.) and a ferromagnet (b.) In a ferromagnet, the order parameter may cross zero multiple times, leading to unusual effects.
In 1964, Peter Fulde and Richard A. Ferrell[4], and independently, Anatoly Larkin and Yuri Ovchinnikov[5] theorized about a unique response of the superconducting state inside a strong ferromagnet. In very high magnetic fields, which ferromagnets consist of on a microscopic level, the Zeeman energy, which augments the energy of a particle based on the particle’s magnetic moment, is strong enough to flip one of the Cooper pair electrons. If one instead considers the metallic state of the ferromagnet, the Zeeman energy imparts different Fermi surfaces to each spin. The effect on the up and down Cooper pair electrons gives the Cooper pair a non-zero momentum. They found that this leads to an order parameter that oscillates inside a ferromagnet (see Fig. 1.8). The phenomenon is called the Fulde–Ferrell–Larkin–Ovchinnikov (FFLO) state and has some interesting consequences. The oscillation in the order parameter may be seen in the $T_c$ and $\lambda^{-2}(T)$ of the films. Minimums have been seen in both parameters[6]. It is even possible for $T_c$ to fall to zero before again becoming finite as ferromagnetic thickness increases, which is called “reentrant superconductivity,”[7] an example of which is shown in Fig. 1.9. It is theoretically possible to see an increase in superconductivity for extremely thin ferromagnets as the additional conducting layer more than compensates for the reduction due to pair breaking.
Figure 1.9: Reentrant superconductivity in Ni$_{59}$Cu$_{41}$/Nb bilayers\cite{17}. All plots show a dip in $T_c$ around 5 to 10 nm. For some critical thicknesses of Nb, here 7.3 and 6.2 nm, $T_c$ drops to 0 K for a range of NiCu thickness. In both cases $T_c$ recovers, and in the case of $d_{Nb} \approx 6.2$ nm, $T_c$ returns to 0 K again around 37 nm.
1.6 Preview of New Physics

We have overviewed the basics of the physics inherent to this thesis. We will see the concepts of superfluid density, BCS theory, and S/F interaction throughout. Other concepts, such as the Kosterlitz-Thouless (K-T) transition, will be introduced in their relevant chapters. The next two chapters will consist of Nb films along side ferromagnets. We start with S/F bilayers, where we find a new interpretation of the dependence of $T_c$ on ferromagnetic thickness, $d_F$. Then, superfluid density is compared favorably to a theory attempting to explain such systems. We move on to S/F/S trilayers and find multiple transitions due to the advantages of superfluid density. In the case of ferromagnetic NiV, we see strong evidence for an FFLO state. We then shift to cuprates where we reveal the first superconducting ultra thin BSCCO films grown with PLD and see that its anisotropy leads to a transition associated with the thickness of a single unit cell and resolve the discrepancy between BSCCO and YBCO films near the quantum critical point (QCP).
Chapter 2

Unexpectedly Small Pair-breaking and Interface Resistance in F/S Bilayers

2.1 Introduction

The proximity effect in bilayers and multilayers of superconductors (S) like Nb and normal (N) metals like Au has been a subject of great interest for several decades[8, 9]. The focus over the last several years has been on multilayer structures of superconducting and ferromagnetic (F) metals because the exchange energy in ferromagnets leads to interesting new qualitative phenomena[9], such as a nonmonotonic dependence of $T_c$ on F layer thickness, $d_F$. In F/S systems, the proximity effect involves Cooper pairs leaking into F and dephasing rapidly because the exchange energy $E_{ex}$ gives different energies to spin-up and spin-down electrons in an s-wave pair, so their quantum phases evolve at different rates. The faster that pairs dephase, the fewer that return to S unbroken, and the lower the transition temperature $T_c$ of the F/S bilayer.

We argue that the dephasing rate may be an order of magnitude smaller than its currently understood value. Previous measurements of $T_c$ vs. $d_F$[10–12], were analyzed with the same free-electron theory used here, but with the assumption that
the F layer is “clean”, and they concluded that the dephasing rate is about $k_B T_C / \hbar$, where $T_C$ = Curie temperature. For example, Aarts et al.[10] find the clean-limit coherence length in F to be very short, 0.1 to 1 nm; Kim et al.[11] find $E_{ex} = 52$ meV and 99 meV for Ni and CoFe, respectively; Sidorenko et al.[12] find $\xi_F = 0.88$ nm in Ni, again very short. We argue that the dephasing rate may be an order of magnitude smaller than $k_B T_C / \hbar$, which is already a factor of 3 or so smaller than the measured exchange energies in 3d ferromagnets[13].

On the theoretical side, Tagirov[14] sounds a note of caution, expressing that before theory’s quantitative application to experiments is fully persuasive, theory should be advanced to include details such as electron correlations, and different Fermi velocities, densities of states, and mean-free-paths for majority and minority spins in F. Nevertheless, despite its shortcomings, current theory may still capture the essence of the physics, even quantitatively. Because ARPES finds short mean-free-paths in 3d ferromagnets[13], which is certainly reasonable for alloys like CoFe and Py, we prefer the dirty-F analysis. This analysis requires only two materials parameters to be taken from the literature – the densities of states at the Fermi level in S and F.

Finally, we analyze $T_c$ vs. $d_F$ taken from our superfluid density measurements on: Ni/Nb, Py/Nb [Py = Ni$_{0.81}$Fe$_{0.19}$], and CoFe/Nb [Co$_{0.5}$Fe$_{0.5}$] bilayers, and we analyze published data on Ni/Nb and CoFe/Nb bilayers.

### 2.2 Theory of Dephasing and Pairbreaking Rates

We explore theory at a level of detail sufficient to bring out our picture of the basic physics in the dirty-F regime. A key quantity is the dephasing angle, $\Gamma_F$, that a typical Cooper pair accumulates in F before returning into S. $\Gamma_F$ is proportional to $E_{ex} / \hbar$ and it grows as: $\Gamma_F = E_{ex} \Delta t / \hbar$, where $\Delta t$ is the typical dwell time of a pair.
in F. $\Delta t$ increases with F thickness, $d_F$, linearly when $d_F$ is smaller than an electron mean-free-path, $\ell_F$, because in that regime Cooper pairs bounce ballistically back and forth between the surfaces of F, striking the F/S interface, and possibly crossing it, on each round trip. (Note that $\ell_F$ is essentially the mean free path for electrons moving perpendicular to F.) When $d_F$ exceeds $\ell_F$, electron motion becomes diffusive, and $\Delta t$ and $\Gamma_F$ increase sublinearly with $d_F$, eventually becoming independent of $d_F$.

Exploration of theory shows that there are three fitting parameters: the dirty-limit coherence length, $\xi_F$, for Cooper pairs in F; $2N_S(0)R_b$, which is the electron density of states in S times the specific resistance $R_b$ of the F/S interface; and $2N_F(0)E_{ex}R_b$, where $2N_F(0)$ is the density of states in F. How to interpret $2N_F(0)$ is discussed below. Experimental values of $2N_S(0)R_b$ and $2N_F(0)E_{ex}R_b$, combined with the ratio $2N_F(0)/2N_S(0)$ from the literature, yield $E_{ex}$. Another point that emerges is that the shallow minimum in $T_c$ vs. $d_F$ that is sometimes observed occurs when the boundary condition for Cooper pairs in F changes from specular scattering at the back surface of F (when $d_F \ll \ell_F$) to diffusive scattering inside F (when $d_F \gg \ell_F$). Thus, existence of a single shallow minimum indicates that $\ell_F$ is much shorter than the thickness at which the minimum occurs.

The experimental dependence of $T_c$ on $d_F$ is illustrated in figures below. Briefly, $T_c$ decreases rapidly at first, then levels off and perhaps dips through a shallow minimum before becoming independent of $d_F$. The leveling-off point is the key piece of data. We follow the formalism of Houzet and Meyer (H-M), which treats both S and F in the dirty limit, but the picture is largely the same in different theoretical approaches, e.g., Tagirov[14], (a simplified version of Tagirov’s theory is found in Sidorenko et al.[12]) and Fominov[15]. H-M assumes that S is thin, so that the superconducting order parameter is uniform through its thickness.

Our starting point is the equation for $T_c$ as a function of the complex pair-breaking
rate, $1/\tau_S$, which depends on $\Gamma_F$, and thereby on $d_F$[16]:

$$\ln \left( \frac{T_c}{T_{c0}} \right) = \Psi \left( \frac{1}{2} \right) - \Re \left\{ \Psi \left( \frac{1}{2} + \frac{1}{2\pi T_c \tau_S} \right) \right\} = \Re \left\{ \sum_{n=0}^{\infty} \frac{-1/2\pi T_c \tau_S}{(n + 1/2)(n + 1/2 + 1/2\pi T_c \tau_S)} \right\},$$

(2.1)

where $\Psi(x)$ is the digamma function and $\Re$ denotes the real component. We use this equation to fit our data. But for simplicity, in this section we restrict the discussion to F/S bilayers in which the suppression of $T_c$ is small. In this regime, Eq. 2.1 simplifies to:

$$T_c \approx T_{c0} - \frac{\pi \hbar}{4k_B \Re \left\{ \frac{1}{\tau_S} \right\}}.$$

(2.2)

Using free-electron relations among density of states, diffusion constant, and resistivity, the pair-breaking rate in Eqs. 2.1 and 2.2 can be written:

$$\frac{1}{\tau_S} = \frac{1}{2\tau_{tun,S}} \frac{1}{1 + 1/\sqrt{2i}(R_b/\rho_F \xi_F) \tanh(\sqrt{2i}d_F/\xi_F)}.$$

(2.3)

The “tunneling” rate that appears in Eq. 2.3 is:

$$\frac{1}{2\tau_{tun,S}} \equiv \frac{1}{4N_S(0)e^2d_S R_b}.$$

(2.4)

$2\tau_{tun,S}$ is the time a typical Cooper pair spends in S before jumping into F, when S is thin[15]. It sets the scale for pair-breaking. The appellation “tunneling” arises because Eq. 2.4 also describes pair-breaking in superconductor-insulator-normal metal tunnel junctions[17–19]. $\rho_F$ in Eq. 2.3 is the resistivity of F. The question of whether $\rho_F$ is the resistivity parallel or perpendicular to the F layer is moot because $\rho_F$ will disappear in the end.

Ferromagnetism enters only through the coefficient of $1/2\tau_{tun,S}$ in Eq. 2.3. In the thin-F regime, $d_F \ll \xi_F$, the tanh in Eq. 2.3 roughly equals its argument, and the following simplification results:

$$\sqrt{2i} \frac{R_b}{\rho_F \xi_F} \tanh \frac{\sqrt{2i}d_F}{\xi_F} \approx 2i \frac{R_b}{\rho_F \xi_F} \frac{d_F}{\xi_F}.$$
introducing $\ell_F$ and $v_{FF}$ and rearranging,

$$2i \frac{R_b}{\rho_F \xi_F} \frac{d_F}{\xi_F} = 2i \left( \frac{3R_b \ell_F}{2\rho_F \ell_F} \right) \left( \frac{2d_F v_{FF}}{3v_{FF}} \right) \left( \frac{1}{\xi_F^2} \right) = i \left( \frac{4d_f}{v_{FF}} \right) \left( \frac{3R_b}{2\rho_F \ell_F} \right) \left( \frac{v_{FF} \ell_F}{3\xi_F} \right) \mbox{ (2.6)}$$

where, by definition, $\frac{v_{FF} \ell_F 3\xi_F^2}{\xi_F} = \frac{E_{ex}}{\hbar}$

$$i \left( \frac{4d_f}{v_{FF}} \right) \left( \frac{3R_b}{2\rho_F \ell_F} \right) \left( \frac{v_{FF} \ell_F}{3\xi_F} \right) = i \frac{E_{ex} 4d_f}{\hbar} \frac{3R_b}{v_{FF} 2\rho_F \ell_F} = i \Gamma_F \mbox{ (2.7)}$$

We identify this expression with $i\Gamma_F$ because it is the dephasing rate, $E_{ex}/\hbar$, times the dwell time in F, i.e., the time, $\Delta t \approx 4d_F/v_{FF}$, that a typical pair takes to bounce off the back of F and return to hit the F/S interface, multiplied by the typical number of hits needed to get through the F/S interface, $\approx 3R_b/2\rho_F \ell_F$[14, 20, 21]. Free-electron relations allow us to express $\Gamma_F$ as:

$$\Gamma_F = 2\tau_{tun,F} E_{ex}/\hbar, \mbox{ (2.8)}$$

where $2\tau_{tun,F} = 4N_F(0)e^2d_F R_b = \frac{d_F}{v_{FF}} \frac{3R_b}{\rho_F \ell_F}$ has the same form as $2\tau_{tun,S}$, but with $2N_F(0)d_F$ in place of $2N_S(0)d_S$. Thus, $2\tau_{tun,F}$ is the time a typical Cooper pair spends in F before jumping back into S, when F is thin.

Putting this all together, Eq. 2.2 for $T_c(\Gamma_F)$ becomes:

$$T_c \approx T_{c0} - \frac{\pi \hbar}{4k_B} \frac{1}{2\tau_{tun,S}} \frac{\Gamma_F^2}{\Gamma_F^2 + 1}, \mbox{ (2.9)}$$

in the thin-F regime $d_F \ll \ell_F$. From Eq. 2.9, $T_c$ decreases most rapidly when $\Gamma_F = 1/\sqrt{3} \approx 0.6$, then crosses to a slow decrease for $\Gamma_F \gtrsim 2$. That the crossover occurs at $\Gamma_F \approx 2$ makes quantitative sense if we suppose that pairs are broken when the dephasing angle exceeds $\pi$ or so. If the crossover happens to occur in the thin-F regime, $d_F \ll \ell_F$, where $\ell_F$ is the mean free path for electrons moving primarily perpendicular to F, then replacing $\Gamma_F \rightarrow 2$ and $d_F \rightarrow d_{F}^{cr}$ in Eq. 2.8 yields the fit parameter: $2N_F(0)E_{ex} R_b \approx \hbar/d_{F}^{cr} e^2$. Also, given that $\Gamma_F \approx 2$ at the crossover,
it follows that the measured suppression of $T_c$ at the crossover yields $1/2\tau_{\text{tun,S}}$: 
$k_B(T_{c0} - T_c) \approx 0.8\pi\hbar/8\tau_{\text{tun,S}}$. From $2\tau_{\text{tun,S}}$ we obtain the fit parameter, $2N_S(0)R_0$.

As mentioned above, the ratio of these fit parameters determines $E_{ex}$ to the extent that the ratio $2N_F(0)/2N_S(0)$ is known.

The foregoing applies for $\ell_F \ll \xi_{F0} \equiv \hbar v_{FF}/E_{ex}$, where $\xi_{F0}$ is the clean-F limit coherence length for Cooper pairs in F, and $v_{FF}$ is the Fermi velocity in F. As an aside, we note that in the clean-F regime, $\ell_F \gg \xi_{F0}$, theory\cite{12, 14} predicts a crossover in $T_c$ vs. $d_F$ at: $d_F \approx T_F\xi_{F0}$. If the transmission coefficient, $T_F$, across the F/S interface is much less than unity, which we believe it to be, then $1/T_F$ is approximately the number of times a Cooper pair has to hit the F/S interface to get through. While this expression differs from the dirty-F result, it corresponds to about the same value of $\Gamma_F$. That is, at this crossover thickness, the time it takes to reflect from the back of F and return to hit the F/S interface, repeated $1/T_F$ times, is: $\Delta t \approx 2d_F/T_Fv_{FF} = 2\xi_{F0}/v_{FF}$. Thus, $\Gamma_F = \Delta t E_{ex}/\hbar \approx 2$.

Returning to the dirty-F regime, we note that there is another, subtler, crossover above the one just described. This crossover is in the thick-F regime where motion inside F is diffusive rather than ballistic. Theory finds a crossover in $T_c$ vs. $d_F$ at: $d_F \approx \sqrt{\hbar D_F/E_{ex}} \equiv \xi_F$, where $D_F$ is the electron diffusion constant in F. When $d_F$ exceeds $\xi_F$, $T_c$ quickly becomes independent of $d_F$. Mathematically, this crossover is found in the $\tanh(\sqrt{2id_F}/\xi_F)$ in Eq. 2.3 that becomes unity for $d_F \gg \xi_F$. $\Gamma_F$ is slightly larger at this crossover. The time it takes an electron to diffuse to the backside of F, then back to the F/S interface is roughly: $\Delta t \approx (2d_F)^2/D_F$, so at $d_F = \xi_F$ the dephasing angle is: $\Gamma_F = E_{ex}\Delta t/\hbar \approx 4$. $\xi_F$ is the third fitting parameter, and its experimental value is essentially the value of $d_F$ at the shallow minimum in $T_c$ vs. $d_F$, above which $T_c$ finally becomes fully independent of $d_F$.

The origin of the minimum in $T_c$ vs. $d_F$ is interesting. We have neglected os-
oscillations in $T_c$ due to oscillations in the order parameter because we neglected the complex nature of the argument of the tanh in Eq. 2.3 for intermediate thicknesses of F. In the physics that we’ve kept, $\Gamma_F$ increases monotonically with $d_F$, so it seems that $T_c$ should decrease monotonically. However, the change in boundary condition at the back of the F layer, from specular reflection of electrons to diffusion, causes a change in the dependence of the pair-breaking rate on $\Gamma_F$. The new form is found from Eq. 2.3 in the thick-F limit where $\tanh(\sqrt{2i d_F}/\xi_F) = 1$:

$$\Re \left\{ \frac{1}{\tau_S} \right\} = \Re \left\{ \frac{\Gamma_F}{(\Gamma_F + 1 - i)} \frac{1}{2\tau_{tun}} \right\} = \frac{1}{2\tau_{tun,S}} \frac{\Gamma_F(\Gamma_F + 1)}{(\Gamma_F + 1)^2 + 1}.$$ (2.10)

For $\Gamma_F > 1$, this expression yields a slightly smaller pair-breaking rate than Eq. 2.9, hence there is a slight increase in $T_c$ as $\Gamma_F$ increases to its ultimate, thick-F value.

### 2.3 Experimental Results and Discussion

The H-M theory assumes that S is much thinner than the Ginzburg-Landau coherence length. We rely on experimental evidence to argue that our films are thin enough that the theory applies, that is, we show that the tunneling pair-breaking rate given in Eq. 2.4 applies for Nb films as thick as ours. Figure 2.1 shows $T_c$ vs. $d_S$ for symmetric Ni/Nb/Ni trilayers with $17 \text{ nm} \leq d_S \leq 52 \text{ nm}$, from Moraru et al.[22]. We fit the data by treating the trilayer as two independent bilayers, each with S thickness equal to half the Nb film thickness, and each in contact with an infinitely thick Ni film. We use the full theory, Eqs. 2.1 and 2.3. The good fit confirms the qualitative prediction that $1/2\tau_{tun,S} \propto 1/d_S$, as expressed in Eq. 2.4. Quantitatively, the best-fit value, $R_b = 2.7 f\Omega \cdot \text{m}^2$, compares well with the value, $R_b = 3.5 f\Omega \cdot \text{m}^2$, obtained by direct measurement[23]. We used $\Gamma_F = 4$ in the fit, per arguments in the preceding section, but the best-fit value of $R_b$ is insensitive to this choice as long as $\Gamma_F \gg 1$. The fitted curve in Fig. 2.1 comes from Eq. 2.1 with: $1/\tau_S = \frac{1}{2\tau_{tun,S}} \left( \frac{4}{5 - i} \right)$, arrived at by plugging
\( \Gamma_F = 4 \) into 2.10, and \( T_{c0} = 9.7 \) K. The experimental density of states in Nb is:

\[
2N(0) = 7.7 \times 10^{28}/\text{eV} \cdot \text{m}^3[24–26].
\]

Figure 2.1: \( T_c \) vs. \( d_{Ni} \) for Ni/Nb/Ni trilayers with thick Ni layers[22]. The good theoretical fit (solid curve) confirms that \( 1/2\tau_{\text{tun,S}} \propto 1/d_S \). The best fit finds a reasonable value for the specific resistance of the F/S interface: \( R_b = 2.7 \, \text{f}\Omega \cdot \text{m}^2 \).

Figure 2.2 shows \( T_c \) vs. \( d_F \) for Ni/Nb bilayers: (a) Ni/Nb(22.5 nm)[11] and (b) Ni/Nb(10 nm)[6]. Note the shallow dip extending from \( d_{Ni} \approx 2 \) nm up to \( d_{Ni} \approx 6 \) nm in the former. To highlight the insensitivity of fit parameters to details, we analyze \( T_c \) vs. \( d_F \) in two ways. The dashed curve in this figure, and in subsequent figures, is a fit using the highly-simplified two-parameter, thin-F, small-pair-breaking version of theory embodied in Eqs. 2.4, 2.8, and 2.9. The solid curve, which captures the dip, is calculated using the full theory (Eqs. 2.1 and 2.3) and includes the third fit parameter, \( \xi_F \). Best-fit parameters are given in the figure captions and in Table 2.1. Best-fit values of \( R_b \) are 2.5 and 4.1 \( \text{f}\Omega \cdot \text{m}^2 \), consistent with the fit in Fig. 1. Given \( R_b \), fitted values of \( 2N_{Ni}(0)E_{ex} \) are 0.66 and 0.88/\( \text{nm}^3 \).

Bilayers involving strongly ferromagnetic alloys Py and CoFe yield results similar to those of Ni despite their higher Curie temperatures and presumably shorter mean-free-paths. Our \( T_c \) vs. \( d_F \) data for Py/Nb and CoFe/Nb bilayers are shown in Figs. 2.3
Figure 2.2: $T_c$ vs. $d_{Ni}$ for Ni/Nb bilayers found in (a) Kim et. al.[11] and (b) Lemberger et al.[6]. Fitting values for exact [solid] (approximate [dashed]) fits are (a) $R_b = 2.5(3.3) f\Omega \cdot m^2$, $2N_F(0)E_{ex} = 0.88$ nm$^{-3}$, $\xi_F = 4.5$ nm; and (b) $R_b = 4.1(4.8) f\Omega \cdot m^2$, $2N_F(0)E_{ex} = 0.66$ nm$^{-3}$, and $\xi_F = 4.5$ nm.

Figure 2.3: $T_c$ vs. $d_{Py}$ for our Py/Nb bilayers. Fitting values for exact [solid] (approximate [dashed]) fits are $R_b = 2.2(3.1) f\Omega \cdot m^2$, $2N_F(0)E_{ex} = 0.67$ nm$^{-3}$, and $\xi_F = 6.5$ nm.

and 2.4a. For Py/Nb, best-fit values are: $R_b = 2.2 f\Omega \cdot m^2$, $2N_{Py}(0)E_{ex} = 0.67$/nm$^3$, and $\xi_F = 6.5$ nm. Clearly we don’t have enough data at large Py thickness to
determine $\xi_F$ – we show the fit only to show that $R_b$ and $2N_{Py}(0)E_{ex}$ change little when $\xi_F$ is included in the fit.

Figure 2.4: $T_c$ vs. $d_F$ for CoFe/Nb from (a) the present work, and (b) Kim et al. Fitting values for exact [solid] (approximate [dashed]) fits are (a) $R_b = 1.6(2.1) f\Omega \cdot m^2$, $2N_F(0)E_{ex} = 1.6 \text{ nm}^{-3}$, $\xi_F = 5.5 \text{ nm}$; and (b) $R_b = 2.3(2.6) f\Omega \cdot m^2$, $2N_F(0)E_{ex} = 3.8 \text{ nm}^{-3}$, $\xi_F = 5.5 \text{ nm}$.

For CoFe/Nb bilayers, Fig. 2.4, the shallow minimum in $T_c$ vs. $d_{CoFe}$ seems to be present in our data, Fig. 2.4a, and in the data of Kim et al., Fig. 2.4b. Per the argument above, this means that $\ell_F$ is much shorter than $\xi_F \approx 5.5 \text{ nm}$, unsurprising for an alloy. The initial drops in $T_c$ and the ultimate values of $T_c$ are different in the two data sets for unknown reasons. The detailed fits to our data and those of Kim et al. find $R_b = 1.6$ and $2.3 f\Omega \cdot m^2$, respectively, about half the value for Ni/Nb bilayers. Values of $2N_{CoFe}(0)E_{ex}$ differ by more than a factor of two from each other, while values of $\xi_F$ are the same, Table 2.1.
Having shown that the dirty-F data analysis provides good qualitative fits to the data, we turn to the key quantitative issue - the value of the effective exchange energy. To get experimental values for $E_{\text{ex}}$, we need to interpret the total density of states, “$2N_F(0)$”, that appears in the free-electron theory that we use. Two possible options are discussed below. In a more comprehensive theory that included ferromagnetic conduction bands, we might expect to find that $2N_F(0)$ is replaced by the total density of states, $N_{F\uparrow}(0) + N_{F\downarrow}(0)$, of majority and minority spin densities of states. Experimental total densities of states for Fe, Co, and Ni range from 1.54 to 2.07 / eV-atom[27]. Thus, at an accuracy appropriate for present purposes, the total density of states is about $1.8 \pm 0.3 / \text{eV}\cdot\text{atom} = 1.7 \pm 0.3 \times 10^{29} / \text{eV}\cdot\text{m}^3$ for all three F layers. Using this value to interpret our data, we find exchange energies given in Table 2.1. On the other hand, it is conceivable that in a better theory the smaller of $N_{F\uparrow}(0)$ and $N_{F\downarrow}(0)$ would create a bottleneck of sorts, and “$2N_F(0)$” would be replaced by two times the smaller of $N_{F\uparrow}(0)$ and $N_{F\downarrow}(0)$. Tunneling[28] and point-contact Andreev reflection[29, 30] experiments both find that the ratio of larger to smaller density of states in 3d ferromagnetic metals is about 2.4$\pm$0.3. In this case, “$2N_F(0)$” would be about half of $[N_{F\uparrow}(0) + N_{F\downarrow}(0)]$. As a consequence, exchange energies would be about double the values in Table 2.1.
Table 2.1: Parameters obtained from theoretical fits to $T_c$ vs. $d_F$ for three F/S bilayer systems. Fermi velocities $v_{FF}$ in F are from the literature. (See text for details.) $R_b$ was obtained from fit parameter $2N_S(0)R_b$ by using $2N_S(0) = 0.77 \times 10^{29}$/eV $\cdot$ m$^3$ for Nb[24–26]. Values of $R_b$ in parentheses are taken from the simple, approximate fits in the figures. The effective exchange energy, $E_{ex}$, was obtained from $2N_F(0)E_{ex}$ using $2N_F(0) = 1.7 \times 10^{29}$/eV $\cdot$ m$^3$. (See text.)

<table>
<thead>
<tr>
<th>F/S</th>
<th>Ref.</th>
<th>$R_b$ [f$\Omega$ $\cdot$ m$^2$]</th>
<th>$2N_F(0)E_{ex}$ [1/nm$^3$]</th>
<th>$\xi_F$ [nm]</th>
<th>$v_{FF}$ [$10^6$ m/s]</th>
<th>$E_{ex}$ [K] [meV]</th>
</tr>
</thead>
<tbody>
<tr>
<td>Ni/Nb</td>
<td>TRL</td>
<td>4.1 (4.8)</td>
<td>0.66</td>
<td>4.5</td>
<td>0.28</td>
<td>45</td>
</tr>
<tr>
<td>Ni/Nb</td>
<td>Kim</td>
<td>2.5 (3.3)</td>
<td>0.88</td>
<td>4.5</td>
<td>0.28</td>
<td>60</td>
</tr>
<tr>
<td>Py/Nb</td>
<td>Hinton</td>
<td>2.2 (3.1)</td>
<td>0.67</td>
<td>6.5</td>
<td>0.22</td>
<td>45</td>
</tr>
<tr>
<td>CoFe/Nb</td>
<td>Hinton</td>
<td>1.6 (2.1)</td>
<td>1.6</td>
<td>5.5</td>
<td>0.33</td>
<td>110</td>
</tr>
<tr>
<td>CoFe/Nb</td>
<td>Kim</td>
<td>2.3 (2.6)</td>
<td>3.8</td>
<td>5.5</td>
<td>0.33</td>
<td>260</td>
</tr>
</tbody>
</table>

Table 2.1 shows that effective exchange energies are about a factor of 10 smaller than $k_B T_C$, [$T_C = 627$ K[31], 871 K[32], and 1600 K[33], for Ni, Py, and CoFe, respectively.] This is our main result. For comparison, exchange energies from ARPES are several times larger than $k_B T_C$, about 0.25 eV for Ni and Py, and more than 0.5 eV for Co[13]. Our dirty-limit analysis is internally self-consistent in that the clean-limit coherence length in F is found to be more than 10 nm, which is surely much larger than electron mean-free-paths in alloys Py and CoFe.

We note that there is a second way to estimate $E_{ex}$, if one accepts the short electron mean-free-paths and Fermi velocities (Table 2.1) from ARPES[13]. (Experimental Fermi velocities are about the same for majority and minority spins.) For example, if we take $\ell_F = 1$ nm, $v_{FF} = 0.3 \times 10^6$m/s, and $\xi_F = 5$ nm, then the definition: $\xi_F \equiv (v_{FF}\ell_F\hbar/3E_{ex})^{1/2}$, leads to $E_{ex} = 8$ meV, which is about double the Table values for Ni and Py.

Our experimental specific boundary resistances, $R_b$, indicate that F/S interfaces are a significant barrier to electron transmission. Aarts and Geers studied Fe/Nb/Fe
trilayers[34] and V/V$_{1-x}$Fe$_x$ multilayers[10]. When we analyze their data, we find $R_b \approx 2 - 4 f \Omega \cdot \text{m}^2$ when we use $2N_V(0) = 2N_{Nb}(0)[35]$. We mention this because Aarts and Geers also concluded that transmission probabilities were small for most interfaces. In this context, we note that Aarts et al.[10], find a relatively large transmission rate for the $V/V_{1-x}\text{Fe}_x$ interface, but only when $V_{1-x}\text{Fe}_x$ is mostly $V$.

Measurements of superconducting transition temperature vs. ferromagnetic layer thickness in F/S bilayers can be analyzed assuming that F is either “dirty” ($\ell_F \ll \xi_{F0}$) or “clean” ($\ell_F \gg \xi_{F0}$). There is a quantitative difference. With the “dirty-F” assumption, the exchange energy is determined to be a factor of 5 to 10 smaller than $k_BT_C \approx 0.1$ eV. This means that Cooper pairs in strong ferromagnets dephase much less rapidly than expected from the experimental exchange energy, which is about 0.25 eV for Ni and Py and greater than 0.5 eV for CoFe from ARPES measurements. A possible reason is spin-orbit scattering, which has been considered theoretically[36]. The effect of the exchange energy on Cooper pairs is essentially the same as the pair-breaking Zeeman effect of a magnetic field, $B$, on Cooper pairs, which has a pair-breaking rate, $1/\tau_S = 2\mu_BB/h$, when spin-orbit scattering is negligible. But when spin-orbit scattering is strong, the pair-breaking rate is reduced by a factor of $\mu_BB/(h/\tau_{so})[1]$. We propose that this mechanism is the explanation, with $E_{ex}$ in place of $2\mu_BB$. Our analysis also finds that transmission of electrons through the F/S interface is relatively poor – Cooper pairs must strike the F/S interface at least several times before getting through.

2.4 Superfluid Density and H-M Theory

We look at superfluid density data for Ni/Nb, Permalloy Ni$_{0.81}$Fe$_{0.19}$/Nb (Py/Nb), and Co$_{0.5}$Fe$_{0.5}$/Nb (CoFe/Nb). Fig. 2.5 is $\lambda^{-2}(0)$ Ni/Nb bilayers with $d_{Nb} = 125 \text{ Å}$. To maximize consistency, most samples shown are grown with the growth chamber
vacuum unbroken. An example of what happens when vacuum is broken is found in Fig. 2.5. The two films with $d_{Ni} = 75$ Å differ by 50%, assuming $\xi_F = 4.5$ nm found in Ni/Nb films, the film with $T_c = 3.6$ K has $R_b = 3.15 f \Omega \cdot m^2$, and the film with $T_c = 2.6$ K has $R_b = 2.55 f \Omega \cdot m^2$. A smaller $R_b$ results in a lower $T_c$ by increasing the tunneling rate of Cooper Pairs into F.

d$_{Ni} = 20$ Å and 25 Å are suppressed less than we would expect from the literature and our experience[6, 11]. The simplest interpretation is for the effective Ni thickness to be much less than indicated. If these two films are assumed to have the average $R_b$ of $d_{Ni} = 75$ Å, 2.8 $f \Omega \cdot m^2$, their Ni thickness would be 6 Å and 11 Å. Alternatively, $R_b$ is extremely large. Regardless, all $\lambda^{-2}(0)/\lambda^{-2}_0(0)$ vs. $T_c/T_{c0}$ tracks with theory.

Figure 2.5: $\lambda^{-2}(T)$ of Ni/Nb films with $d_{Nb} = 125$ Å and $d_{Py} = 0 - 75$ Å. $d_{Ni} = 20$ and $d_{Ni} = 25$ have anomalously high $T_c$ and proportionally high $\lambda^{-2}(0)$. The two $d_{Ni} = 75$ Å indicate their $R_b$’s differ by 20%. Despite low effective F thicknesses, all films agree with theory. [See Fig. 2.8]

Figures 2.6 and 2.7 contain superfluid data for Py/Nb and CoFe/Nb, respectively,
Figure 2.6: $\lambda^{-2}(T)$ of Py/Nb bilayers with $d_{Nb} = 250$ Å and $d_{Py} = 5 - 30$ Å. $T_{c0} = 8.5$ K for $d_{Nb} = 250$ Å (not shown). Also not pictured, $d_{Py} = 100$ Å, which has identical $\lambda^{-2}(T)$ to $d_{Py} = 30$ Å.

Figure 2.7: $\lambda^{-2}(T)$ of CoFe/Nb bilayers with $d_{Nb} = 250$ Å and $d_{CoFe} = 5$ to 100 Å. $T_{c0} = 8.5$ K for $d_{Nb} = 250$ Å. $T_{c0} = 8.5$ K for $d_{Nb} = 250$ Å (not shown).
where $d_{Nb} = 250 \, \text{Å}$. A sample (not shown) with $d_{Py} = 100 \, \text{Å}$ had nearly identical data to $d_{Py} = 30 \, \text{Å}$ film, indicating little change after 30 Å. In Figure 2.8, the superfluid and $T_c$ values of Fig. 2.5, 2.6, and 2.7 were normalized to a Nb monolayer of the same $d_S$ used in the bilayers. The black line represents the expected drop in $\lambda^{-2}(0)$ due only to the reduction in $T_c$. The grey area marked “Theory” represents the allowed values given by Houzet-Meyer theory throughout all of parameter space. Ni and Py values stick closely to the theory while CoFe/Nb fall below expectations. We believe these films have less $\lambda^{-2}(0)$ rather than higher $T_c$, which could be caused by the large Nb thickness ($d_S = 250 \, \text{Å}$) combined with low $T_c$. Since $d_{Nb}$ is much larger than $\xi_{Nb} = 80 \, \text{Å}$, changes in the order parameter are possible. The low interface resistance means CoFe has more access to the superconducting state which could cause the OP to bend near the interface inside Nb, and compromise $\lambda^{-2}(0)$ as shown in Fig. 2.9. It is possible the same effect occurs in the Py films, but since $T_c$ never drops below $0.6T_{c0}$, the effect is not seen.

Superfluid density measurements have shown strong agreement between H-M theory and experiment. For Ni and Py, experiment is right in line with expectation, while CoFe has a lower $\lambda^{-2}(0)$ than is called for. H-M theory assumes that the thickness of $S$ is less than $\xi_S$. We were forced to use thicknesses several times $\xi_S$, which may explain what we observed. The thicker Nb makes it susceptible to variations in the order parameter. We did not see the same phenomenon in Py/Nb, even though it used equal $d_{Nb}$, but this could be explained by the lower overall effect Py had on $T_c$. Earlier, we showed that assumptions about the effective ferromagnetic exchange energy, $E_{ex}$, are likely incorrect and lead to underestimates of the interface resistance, $R_b$. We found values for these as well as the ferromagnetic coherence length, $\xi_F$, and showed how these results are consistent with our analysis of the theory.
Figure 2.8: $\lambda^{-2}(0)$ vs. $T_c$ normalized to a Nb monolayer of the same thickness used in the bilayers, compared to H-M theory (grey segment). The black line represents a one-to-one reduction in $\lambda^{-2}(0)$ expected from suppression in $T_c$. The Ni and Py data fit closely while $\lambda^{-2}(0)$ is lower than expected for CoFe below $0.6T_{c0}$. Fig. 2.9 offers a diagram of how CoFe may be causing this discrepancy through the order parameter.
Figure 2.9: Depiction of how the order parameter could differ at the F/S boundary in Nb/Ni and Nb/CoFe. (a.) Order parameter stays constant in Nb of Ni/Nb because $d_{Nb} = 125 \text{ Å}$ is only 50% more than $\xi_S \approx 80 \text{ Å}$[11, 15]. (b.) In this case, $d_{Nb} = 250 \text{ Å}$, which is several times $\xi_S$, was necessary to keep $T_c$ above 0 K. Thereby, the order parameter can dip inside Nb resulting in less superfluid. Concurrently, $T_c$ is reinforced by the Nb farthest from the F layer. This results in lower $\lambda^{-2}(0)$ than theory predicts.
Chapter 3
Evidence of FFLO State and π-Junction in Nb/NiV/Nb Trilayers

3.1 Introduction

S/F/S trilayers is the logical step after bilayers. When we transitioned from bilayers to trilayers, we attempted several ferromagnets, starting with the ones we used in the bilayers (Ni, Py, CoFe), but there were challenges. Our main obstacle was that a small amount of Py or CoFe had a large effect on $T_c$ that required thicker Nb, but thicker Nb saturated our signal. We had better luck with Ni, Ni$_{0.5}$Cu$_{0.5}$, and especially Ni$_{96}$V$_{04}$. The majority of this chapter deals with the latter.

3.2 Second Transition Analytic Methods

Trilayer data analysis poses issues because to determine the superfluid contribution of the layer that initially superconducts, $T_{c1}$, the data must be extrapolated far from the measured data to obtain $\lambda_1^{-2}(0)$, the contribution of this Nb layer to total $\lambda^{-2}(0)$ near $T_c$ of a Py/Nb bilayer that follows BCS. Thankfully, if one fits a portion of the data between $T_{c1}$ and $T_{c2}$, it extrapolates well to the rest of the data, and presumably to $\lambda^{-2}(0)$. 

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Lemberger et al. [37] studied the superfluid of Nb monolayers. In this paper, films with $d_{Nb} > 150$ Å were well fit with a gap value of $1.9 \, k_B T_c$ [38]. For films with $d_{Nb} < 125$ Å, $1.8 \, k_B T_c$ was more appropriate. In the subsequent paper involving Ni/Nb bilayers [6], approximately the lowest 10 - 20% $T$ data was fit with a two-term quadratic, to ensure a slope of zero at $T = 0$ K, since that is the “gapless” behavior that manifests with large pair breaking [39]. This occurs approximately when $T_c$ is suppressed by a factor of two from $T_{c0}$. For Nb, this is around 4 K, which is a reasonable cutoff for the point below which a BCS fit is no longer appropriate.

![Figure 3.1: Our fitting protocol, demonstrated on two bilayers, Py/Nb with $T_c \approx 6$ K and CoFe/Nb with $T_c \approx 3$ K. The yellow segment is the data selection used to generate the light blue BCS fits on both plots. For Py/Nb, the green line is the BCS fit using all the data. For CoFe/Nb the green line is a two-term quadratic fit from 1.5 K to 2.5 K. The two fitting methods differ by only 2% for Py/Nb but by 20% in the case of CoFe/Nb.](image)

Observing the Py/Nb and CoFe/Nb bilayers, it is obvious that films with $T_c$ above 5 K are still somewhat gapped (BCS-like) based on the flattening of $\lambda^{-2}(T)$ at low $T$. 35
When fit with the dirty-limit BCS curve from Eq. 1.5, values for $\Delta(0)/k_B T_c$ range from 1.5 to 1.9, which are plausible. A value of 1.9 is associated with Nb monolayers of $T_c > 8$ K and 1.8 is associated with $T_c < 8$ K, but most gap parameters in Py/Nb and CoFe/Nb were less than 1.8, consistent with the expected shrinking of the gap due to increased pair breaking. In Fig. 3.1, a high $T_c$ Py/Nb sample is contrasted with a low $T_c$ CoFe/Nb sample. In both cases, a BCS fit is employed over a small section of data near $T_c$. For Py/Nb, extrapolating the data to lower temperatures agrees with a BCS curve fit using all the data, with only a 2% deviation. This is how we find $\lambda^{-2}(0)$, by fitting the data where only that film is currently superconducting, and we see it should give us a close approximation of the contribution from a $T_c > 6$ K layer to $\lambda^{-2}(0)$. Whether it is more appropriate to fit films with $T_c = 4 - 5$ K with a quadratic or dirty-limit is something of an art, but it should be uncontroversial for 6 K and above. All NiV trilayers have $T_{c1} \geq 6$ K.

### 3.3 Ni and NiCu Trilayers

First, we remark on some of the results from Nb/Ni/Nb and Nb/NiCu/Nb trilayers where we found more obvious second transitions than the subtle ones we will tease out of NiV. Fig. 3.2 shows S/Ni/S trilayers. The bottom layer is Nb(125 Å) the middle is Ni(125 Å), and the top varies Nb(105.5, 125, and 150 Å). The second transition is revealed by a “kink” in superfluid density, seen consistently at $3.2 \pm 0.1$ K which is clear because the Nb layers are fully decoupled by 150 Å of Ni. The second transition is the bottom Nb layer, Nb(125 Å), transitioning in each trilayer. $T_{c1}$ increases with increasing $d_{Nb}$ of the top layer. As a consequence, Nb(105.5 Å) has a higher $T_c$ than Nb(125Å). Additionally, when both Nb layers are $d_{Nb} = 125$ Å, $T_{c1}$ is 50% higher than $T_{c2}$. Our conclusion is the upper layer is more insulated (larger $R_b$) from the ferromagnet, which probably means Ni grows more cleanly on Nb than Nb on Ni.
This is consistent with what we noticed working with Ni/Nb bilayers. Nb grown on top of Ni led to inconsistent results, which we speculate was caused by changes in interface roughness or Nb crystallinity with $d_{Ni}$.

Figure 3.2: Nb/Ni/Nb trilayers with top Nb thicknesses ranging from 105.5 Å to 150 Å, Ni thickness of 125 Å, and bottom Nb of 125 Å. The Ni is thick enough to completely isolate the Nb layers from each other. The result is two distinct transitions where the $d_{Nb} = 125$ Å film is stationary. The Nb layer grown above the Ni has a higher transition due to greater S/F interface resistance.

Before writing Lemberger et al. [40] (first half of Chapter 2), we believed the conventional wisdom that the strength of the ferromagnet was the dominant factor in determining $T_c$ rather than $R_b$. Thus, we were attracted to ferromagnets with low Curie temperatures in an effort to decrease required Nb thickness. The Curie temperature of Ni$_{0.5}$Cu$_{0.5}$ is $T_C = 110$ K. In Fig. 3.3, Nb(150 Å) is used on top and bottom. $d_{NiV}$ ranges from 15 Å to 25 Å. The film with NiCu(15 Å) has a single transition because the Nb films are fully coupled. All others act as if they are
completely decoupled. Films are grouped together with no correlation. NiCu(50, 150, and 200 Å) lie on top of each other, as does NiCu(75 and 100 Å), while NiCu(254 Å) has higher $\lambda^{-2}(0)$ than the rest.

In an attempt to explain this we note 100 Å and 250 Å were grown in the same series, as were 50, 75, 150, and 200 Å, though this does little to shed light on what happened. Instead, the inconsistency between samples is likely attributed to problems with the NiCu target. The sputtering process would spontaneously end, which makes growth thickness and quality inconsistent since it calls into question the sputtering energy and the actual growth time.

Absolute $R_b$ values cannot be determined with the given information, but we can estimate the ratio of $R_b$ between upper and lower Nb/NiCu interfaces. We do this by assuming the NiCu layer is in the thick limit. We averaged the fitted values of $T_{c1}$ and $T_{c2}$ for the closely bunched data. In the three resulting cases the ratio $T_{c2}/T_{c1} = 0.74 \pm 0.01$, and $R_{b2}/R_{b1} = 0.77 \pm 0.06$. These are tight error bars, indicating despite inconsistent $\lambda^{-2}(T)$, the ratio of $R_b$ between interfaces is consistent and insensitive to growth conditions. The interface resistance due to whether Nb is grown on NiCu or vice versa thus appears independent of other growth conditions.

### 3.4 Pi Junction in Nb/NiV/Nb

Since our NiCu target was not proving useful, we moved onto another alloy, Ni$_{0.96}$V$_{0.04}$, which led to our most successful results. This is enough vanadium to reduce the $T_C$ of Ni from 627 K to $\approx$ 400 K. We were then able to use Nb layers of 125 Å and 200 Å without completely screening the mutual inductance signal. In this section, NiV thickness goes from 20 Å to 55 Å in 5 Å increments. As a shorthand, each trilayer will be referred to by its NiV thickness. Observing Fig. 3.4, one thing is obvious: superfluid density decreases to a minimum at 35 Å and then increases from there.
Figure 3.3: Nb/NiCu/Nb trilayers with $d_{Nb} = 150$ Å on top and bottom. The kinks are obvious, which implies little thickness dependent change when $d_{NiCu} \geq 50$ Å. Therefore, 50 Å, 150 Å, and 200 Å, and separately, 75 Å and 100 Å are nominally equivalent, indicating these groupings were grown under similar conditions. The most likely reason is the target was poor, which caused $R_b$ to vary considerably though this did not affect the difference caused by Nb/NiCu growth order.

until 55 Å. Data for 30 Å and 45 Å were removed to improve readability, but are included in the rest of the analysis.

First we find $T_c$ and $\lambda^{-2}(0)$ as if the films were bilayers with $d_{Nb} = 325$ Å and a single $T_c$. The results are in Fig. 3.5. We observe a recovery of 33% in $\lambda^{-2}(0)$, beginning at 30 Å. Since $T_c$ is essentially constant, the increase in $\lambda^{-2}(0)$ cannot be attributed to the Nb layers at first blush. Another option is to consider the additional NiV. The additional NiV increases the total trilayer thickness by only 7%. To account for the extra $\lambda^{-2}(0)$ from the extra conductive material, NiV would need to have almost five times the conductivity of Nb because the additional conductive material would be superconducting and thus add to the superfluid density. Since the conductivities are likely to be comparable, we need to account for the increase in
Figure 3.4: Nb/NiV/Nb trilayers with $d_{Nb} = 200$ Å and $d_{Nb} = 125$ Å and $d_{NiV} = 20 - 55$ Å in 5 Å increments excluding 30 Å and 45 Å. $\lambda^{-2}(T)$ and $T_c$ fall until $d_{NiV} = 35$ Å, and afterwards there is a recovery up to 55 Å.

another way.

Notice in Fig. 3.4, how the data tend to “fan out” at lower temperatures, starting below $\approx 3.5$ K, which may be an indicator of a second transition. These are more subtle than the kinks found in Nb/Ni/Nb and Nb/NiCu/Nb, which were separated by more ferromagnetic material. Using the method outlined earlier, we will decouple the contribution to $\lambda^{-2}(0)$ from each Nb layer. The 20 Å and 25 Å films are fully coupled, and are analyzed as such. To illustrate this, Fig. 3.6, which is the 20 Å film, is comfortably fit by a dirty-limit BCS curve with gap of $1.9k_BT_c$. Alternatively, Fig. 3.7 is the 55 Å film. The data corresponding to Nb(200 Å) is fit with a BCS curve. When the fit is extrapolated to low $T$, it departs below the data. $\lambda_1^{-2}(0)$ is recorded and the rest of the data is fit with a two-term quadratic, which obtains our value for $\lambda_2^{-2}(0)$. The quadratic fit invariably crosses the BCS fit, and we define this cross as $T_{c2}$. Quantitatively determining $\lambda_2^{-2}(0)$ requires subtracting $\lambda_1^{-2}(0)$ and renormalizing.
Figure 3.5: $\lambda^{-2}(0)$ and $T_c$ of NiV trilayers showing the rapid drop in $\lambda^{-2}(0)$ and subsequent recovery while $T_c$ stays effectively constant.

Figure 3.6: Nb/NiV/Nb with $d_{\text{NiV}} = 20$ Å fit with a BCS curve. The lack of deviation shows this trilayer is likely fully coupled.
Figure 3.7: Nb/NiV/Nb with $d_{\text{NiV}} = 55$ Å. Data above 4 K are fit with a BCS curve (blue) while data below 4K are fit with a two term quadratic (red). The failure of the BCS curve to fit all the data implies the second transition is real.

to $d = 125$ Å.

Now that we have obtained $\lambda^{-2}(0)$ and $T_c$ for each layer, Fig. 3.8 displays all four sets of data. As stated, the Nb layers in the NiV(20 Å) and NiV(25 Å) trilayers are fully coupled. The top two lines are $T_c$ and $\lambda^{-2}(0)$ for the 200 Å Nb layer and the bottom two are the $T_c$ and $\lambda^{-2}(0)$ for 125 Å. For the coupled data, it was necessary to calculate an equivalent Nb monolayer thickness using the sum of the areal superfluid $(\lambda^{-2}d)$ of a 200 Å and 125 Å Nb film, which has the same value as a Nb(246 Å) monolayer which has a corresponding $\lambda^{-2}(0) = 79.8 \mu$m$^{-2}$. This is more appropriate than using the sum of the thicknesses (325 Å) since splitting the monolayer with NiV would have a larger effect on $\lambda^{-2}(0)$ than adding NiV to one side. One striking result is the increase in $T_c^2$ and $\lambda_2^{-2}(0)$ from the minimums at $d_{\text{NiV}} = 35$ Å, even as $T_{c1}$ is flat and $\lambda_2^{-2}(0)$ grows only slightly. Additionally, the oscillatory nature of each of the graphs is indicative of the FFLO state one would expect in an F/S system.
Figure 3.8: Superfluid and $T_c$ attributed to the 200 Å and 125 Å Nb layers. Black represents fully coupled films. $T_c1$ (dark blue) and $\lambda_{c1}^{-2}(0)$ (red) are associated with the $d_{Nb} = 200$ Å. $T_c2$ (light blue) and $\lambda_{c2}^{-2}(0)$ (green) correspond to $d_{Nb} = 125$ Å. Lines are a guide for the eye. The oscillation of $T_c2$ and $\lambda_{c2}^{-2}(0)$ is what we expect from an FFLO state.

One might argue that the 45 Å or 50 Å data point holds too much weight in this determination, but it is worth pointing out $T_{c1}$ and $T_{c2}$ follow the same oscillatory pattern, implying the phenomenon is affecting both films similarly. This correlation bolsters the likelihood this is something real rather than an artifact of the fitting scheme.

The two films are partially coupled in most, if not all, of these films. This means, as Nb(200 Å) superconducts, the value of the order parameter at the interface of Nb(125 Å) will oscillate depending on the thickness of NiV. So after Nb(200 Å) is superconducting, Nb(125 Å) becomes “aware” it is also a superconductor, though without much superfluid response of its own. This could explain why the second “transition” is more subtle – Nb(125 Å) has already undergone normal to supercon-
ducting transition by virtue of its proximity to Nb(200 Å), which may be why there is a rise in $\lambda_1^{-2}(0)$ of Nb(200 Å) vs $d_{NiV}$. The oscillation in $T_{c2}$ and $\lambda_2^{-2}(0)$ of Nb(125 Å) is large – 25% in $T_{c2}$ and 75% in $\lambda_2^{-2}(0)$, peak to peak, which could mean $T_{c2}$ is highly dependent on the phase of the order parameter from Nb(200 Å) at the Nb(125 Å) interface. Thus, $T_{c2}$ and $\lambda_2^{-2}(0)$ fluctuate greatly while $T_{c1}$ is generally flat and $\lambda_1^{-2}(0)$ is gently increasing with little oscillation.

![Figure 3.9](image.png)

**Figure 3.9:** $\lambda^{-2}(0)/\lambda_0^{-2}(0)$ vs. $T_c/T_{c0}$ compared to H-M Theory (grey) for $d_{Nb} = 200$ Å (top right) and $d_{Nb} = 125$ Å (bottom left). Data point color corresponds to a specific trilayer and the numbers are the thickness of NiV in Å. Black line is the expected drop in $\lambda^{-2}(0)$ due to the drop in $T_c$.

In Fig. 3.9, we analyze each trilayer as two bilayers and compare to H-M theory. The number next to each data point is $d_{NiV}$ in Å. Points 20 and 25 are only shown once because there is no second transition. These points use the effective Nb thickness of 246 Å discussed earlier and drop considerably below the theory line. The Nb(200 Å) transition in NiV(30 Å) is near the NiV(25 Å) film, which increases confidence.
that the effective Nb thickness was the correct interpretation for points 20 and 25. Point 30 from the Nb(125 Å) transition is similarly below the theory line. From then on, the data are much closer to theory, which shows that the films are becoming less coupled and that our style of fitting the trilayers is likely correct.

Since the superfluid was considerably suppressed from $d_{NiV} = 20 - 30$ Å, where we would not expect much of an effect from thin NiV, we offer a π-junction as a possible explanation. The π-junction configuration would require the order parameter to pass through zero inside NiV. Having to change from the maximum value, inside Nb, down to zero across 10 - 15 Å is at odds with the superconducting coherence length $\xi_S \approx 80$ Å, even considering that the ferromagnetic coherence length, $\xi_F$, is likely around 25 Å. Thus, it is conceivable the order parameter dives inside Nb, resulting in a much lower superfluid density than expected.

Fig. 3.10 illustrates the consequences of a π-junction. Fig. 3.10a shows the small dip in $\Psi(x)$, where $\Psi^2(x) \propto \lambda^{-2}(0)$, that would occur from a very thin NiV layer. Fig. 3.10b shows the π-junction, likely appearing in the fully-coupled regime, NiV(20 Å) and NiV(25 Å), and also in the first decoupled trilayer, NiV(30 Å), as indicated by the H-M analysis. This is caused by $\Psi(x)$ being forced through zero without enough room to avoid dipping considerably inside the Nb layers, thus stealing from the main sources of $\lambda^{-2}(0)$. In Fig. 3.10c., $\lambda^{-2}(0)$ recovers overall, but $T_c$ and $\lambda^{-2}(0)$ drop in Nb(125 Å). At some point, NiV would become thick enough that the π-junction would no longer exist as the Nb layers become less coupled (Fig. 3.10d).

Finally, by fitting the $T_c$ vs $d_{NiV}$ as we did in Chapter 2, we find $E_{ex} = 5.2$ meV, approximately 14% of $T_C$, approximately the same proportion as in Ni, Py, and CoFe. The coherence length, $\xi_F = 25$ Å, is smaller than found in any of the bilayers. $R_b$ for the Nb(200 Å)/NiV interface is 1.6 $f\Omega\cdot m^2$, and 1.2 $f\Omega\cdot m^2$ for the Nb(125 Å)/NiV interface. These are similar to the interfaces of Py/Nb and CoFe/Nb, also alloys, and
Figure 3.10: (a.) The first bit of NiV causes a dip in the order parameter, $\Psi(x)$. (b.) The $\pi$-junction produces a considerable drop in $\lambda^{-2}(0)$ as the $\Psi(x)$ is forced to drop inside Nb. The value of the $\Psi(x)$ at the back of each Nb layer represent $T_c$ and $\Psi^2(x) \propto \lambda^{-2}(0)$. (c.) As $d_{NiV}$ increases, $T_c$ of Nb(200 Å) stays constant while overall $\lambda^{-2}(0)$ increases, though there is a simultaneous deep minimum in $T_c$ and $\lambda^{-2}(0)$ of Nb(125 Å). (d.) Eventually, they become nearly decoupled and the $\pi$-junction reverts to the standard configuration.

consistent with the difference between top and bottom S/F interface resistances seen in other trilayers.

In summary, we analyzed trilayers consisting of Nb/Ni/Nb and Nb/NiCu/Nb which displayed two transitions, and showed the strong role interface transparency plays in determining $T_c$. Then, by fitting sections of $\lambda^{-2}(T)$ in Nb/NiV/Nb with BCS and two-term quadratic functions, we unbiasedly analyzed the dependency of
$T_c$ and $\lambda^{-2}(0)$ as a function of NiV thickness and found unusually strong fluctuations indicative of an FFLO state in Nb/NiV/Nb trilayers. We also discovered evidence of a $\pi$-junction causing $\lambda^{-2}(0)$ to be suppressed to extremes that are otherwise difficult to explain by traditionally coupled trilayers. The H-M theory helped in visualizing these deviations and reinforced the legitimacy of the fitting methodology.
Chapter 4

**Single Bilayer K-T Transition in Ultrathin BSSCO**

4.1 Motivation for Studying Ultrathin BSCCO Films

A central question regarding cuprate superconductivity is the nature of the superconducting state in strongly underdoped materials. Part of the question concerns the pseudogap that appears at a temperature well above the superconducting $T_c$ [41, 42]. Does it diminish the portion of Fermi surface available for superconductivity, is it a superconducting fluctuation that persists to very high temperatures, or something else? Another aspect, the one of interest here, concerns the importance of quantum and thermal fluctuations of the superconducting order parameter below $T_c$, in the fully superconducting state.

To explore this issue, we measured $\lambda^{-2}(T)$ of strongly underdoped films of YBa$_2$Cu$_3$O$_{7-\delta}$ (YBCO) [43, 44], Ca-doped Y$_{1-x}$Ca$_x$Ba$_2$Cu$_3$O$_{7-\delta}$ (Ca-YBCO) [45], and Bi$_2$Sr$_2$CaCu$_2$O$_{8+\delta}$ (BSCCO or Bi-2212) [46]. These particular compounds were chosen for several reasons. One is that they both have been studied heavily at moderate underdoping, the former mostly by electron transport, optical spectroscopy, and thermodynamic measurements, the latter largely by surface techniques of Angle-Resolved PhotoEmission Spectroscopy (ARPES) and Scanning Tunneling
Microscopies (STM). Another is that the YBCO and Bi-2212 compounds stand at opposite ends of the spectrum of $ab$-plane vs. $c$-axis anisotropy. YBCO is less anisotropic with $\{a,b,c\} = \{3.82 \, \text{Å}, 3.89 \, \text{Å}, 11.68 \, \text{Å}\}$ while Bi-2212, is on the anisotropic extreme $\{a,b,c\} = \{3.82 \, \text{Å}, 3.82 \, \text{Å}, 30.85 \, \text{Å}\}$, though a Bi-2212 lattice contains double the copper oxide bilayers of YBCO, so it is more appropriate to think of its $c$-parameter as 15.42 Å. The final rationale is our ability to grow reasonably homogeneous, severely underdoped films of both materials.

Superfluid density informs us of fluctuations in two ways. Its $T$-dependence reveals critical thermal fluctuations as an abrupt downturn in $\lambda^{-2}(T)$ as $T$ approaches $T_c$. If the sample is reasonably homogeneous, i.e., the spread in $T_c$’s in the sample is small, then the details of the downturn reveal the dimensionality of thermal fluctuations. For example, YBCO crystals near optimal doping show critical scaling, $n_s(T) \propto (1 - T/T_c)^{2/3}$, for $T/T_c$ greater than about 0.9. The exponent, 2/3, indicates 3-D thermal fluctuations [47]. Two-unit-cell thick Ca-YBCO films show an abrupt downturn in $n_s(T)$ at about the same place that 2-D theory predicts a discontinuous drop to zero [45]. The presence of power-law scaling of $\lambda^{-2}(0)$ with $T_c$ over a wide range of $T_c$’s indicates we are near a quantum critical point; the scaling exponent indicates the dimensionality of the fluctuations. Linear scaling points to 2-D fluctuations.

A recent paper [46] reports superfluid density measurements on a series of underdoped Bi-2212 films. The paper shows that thermal critical behavior is absent from these films, even though there is strong evidence for 2-D quantum critical fluctuations. On one hand, this is surprising, considering that the only other strongly-underdoped system that shows strong 2-D quantum critical fluctuations (ultrathin Ca-YBCO films [45]) also exhibits clear 2-D thermal critical fluctuations. On the other hand, superfluid densities of strongly underdoped “thick” YBCO films [46] and YBCO crystals [48] display 3-D quantum fluctuations but do not exhibit critical ther-
mal fluctuations of any dimensionality, so the Bi-2212 result is at least consistent with the less-anisotropic YBCO on this point.

This chapter extends the work in [46] to include recently grown, thinner, smoother, Bi-2212 films. These are interesting because they reconcile a quantitative discrepancy in 2-D quantum scaling between ultrathin Ca-YBCO and Bi-2212, as well as exhibit a downturn in $\lambda^{-2}(T)$ for a single CuO$_2$ bilayer where theory expects. For a detailed discussion of the growth process see the Appendix.

4.2 Growth Technique

The Bi-2212 films are grown on LaAlO$_3$ (LAO) by pulsed laser deposition (PLD). General details of the process are written in Yong et al. [46] or found in Appendix A. Where this system differs from Yong is our films were grown off-axis using a pulse-and-wait growth pattern. The previous films had been grown on-axis, with constant pulse separated by 3 s on MgO substrates. (“On-axis” means that the plume of material from the PLD target flies directly toward the substrate; “off-axis” means the plume is parallel to the substrate.) The pulse and wait system means material equivalent to half a Bi-2212 unit cell, including a single CuO$_2$ bilayer, was ablated at 30 Hz followed by a 90 s pause.

In addition to changing the growth technique, we went through an iteration (see Fig. 4.1) of substrates and buffer layers before arriving at LAO. Films were typically grown on MgO despite the massive lattice mismatch of over 9%. First we attempted a buffer layer of another form of BSCCO (Bi-2201), forcing it to take the brunt of the mismatch, and had success making the Bi-2212 film much thinner (See Fig. 4.2). Turning our efforts to substrates, SrTiO$_3$ (STO) allowed us to lower the lattice mismatch between each interface to 1%, and finally it was realized that LAO eliminated any need for a buffer layer.
LAO has an advantage over MgO due to its much closer lattice match with Bi-2212. A considerable amount of the increased superfluid and smoothness is likely attributed to this. The off-axis films were about 7 times thinner (15 nm vs. 100 nm). The earlier films had rough surfaces, as did films grown by sputtering at Technion in Israel. While the PLD and sputtered films agreed with each other remarkably well in both magnitude and T-dependence of \( \lambda^{-2}(T) \), and both displayed 2-D quantum scaling, \( T_c \propto \lambda^{-2}(0) \), there was a factor-of-three difference from ultra-thin Ca-YBCO films. That is, for a given \( T_c \), \( n_s(0) \) for Bi-2212 was three times smaller than for Ca-
Figure 4.2: 7 nm thick Bi-2212 film with 25 nm thick Bi-2201 buffer layer. The transition of the Bi-2201 layer can be seen at low $T$. Blue data is $\sigma_1$.

YBCO. It is plausible that the discrepancy would be addressed by making thinner, and thus smoother Bi-2212 films, since the bumps probably carry little super current and may obscure thermal fluctuations.

Figure 4.3 shows Atomic Force Microscopy (AFM) images of the surfaces of on-axis and off-axis PLD-grown films. Horizontal and vertical scales are the same in the two images. The off-axis films are ten CuO$_2$ bilayers thick (about 15 nm) or less, while the on-axis films were about 100 nm thick. Taking the difference in thickness into account, it is still apparent that the off-axis films are smoother, except for occasional small aspersions, while the on-axis films are quite rough. We emphasize that $\theta - 2\theta$ x-ray scans found only Bi-2212 peaks for both types of films.
Figure 4.3: AFM images of the surfaces of films of Bi-2212 grown by PLD on-axis (upper) and off-axis (lower). Horizontal and vertical scales are the same.

4.3 Single Bilayer K-T Transition

Vortices, which we mentioned in the first chapter, occur in vortex-antivortex pairs. The difference between the two is the direction of the rotating supercurrent and magnetic flux quantum residing in its core. The nature of the pairs causes attraction between them and thus a binding energy. This binding energy can be overcome by thermal fluctuations. This can only occur in two dimensions (2D) and is called
the Kosterlitz-Thouless (K-T) transition\cite{Kosterlitz1973}. Since it occurs in a 2D environment, observing the transition requires a sufficiently thin, homogeneous film.

Figure 4.4: 2 u.c. YBCO films from Hetel et al. \cite{Hetel2020} showing the abrupt downturn, indicative of a K-T transition. The dashed line represents the superfluid density value where we would expect the K-T transition to occur, which bisects the downturn in these plots.

In Hetel et al. \cite{Hetel2020}, the K-T transition was found in 2 u.c. YBCO. This is seen in Fig. 4.4 as a downturn in $\lambda^{-2}(T)$. For higher $T_c$ films, the K-T line cuts through the center of the downturn while in the lowest $T_c$ samples, it crosses at the start of the transition. In YBCO, the slope of the K-T transition line depends on the thickness of the film. As thickness increases, the slope of the K-T line decreases proportionally. This is why extremely thin films were needed to see the downturn.
Figure 4.5: Thick Bi-2212 data from Yong et al. [46]. In near-optimally doped films (top), a downturn appears. The red line, corresponding to two CuO$_2$ bilayers, crosses at the downturn. The black line for a single CuO$_2$ bilayer fails to show any action where it crosses the data. (bottom) Data from above, zoomed in at low $T$, showing a linear behavior and absence of K-T transition.
Figure 4.6: (a.) Superfluid density and real conductivity, $\sigma_1$, vs. $T$ for off-axis-grown Bi-2212 films of 4, 5, and 10 CuO$_2$ bilayers. Black line indicates expected 2D transition for single copper oxide bilayer. Thicknesses of samples shown in terms of number of CuO$_2$ bilayers. (b.) Zoomed in on the samples with the lowest doping, showing the K-T line hits right at the downturn.
The hope in Bi-2212 was that the large anisotropy would reveal K-T transitions at the single CuO$_2$ bilayer level, despite films consisting of many unit cells. In Yong et al., our group found evidence for such a transition in near-optimally doped films, but strangely, it occurred at the double CuO$_2$ level. Underdoped films show quasi-linear superfluid response also found in underdoped YBCO crystals. (See Fig. 4.5) The off-axis films we grew in Fig. 4.6 are not linear at low temperatures, and display what could be a K-T like drop in $\lambda^{-2}(T)$.

The data have similar curvature to 2 unit cell YBCO films. The line indicates where theory predicts a 2D transition should occur, assuming that individual CuO$_2$ bilayers fluctuate independently, which is visible as a kink in some films and occurs at an inflection point in others. Also shown in Fig. 4.6a is the real conductivity, $\sigma_1(T)$, measured at 50 kHz, which has a peak near $T_c$. The width of the peak is an upper limit on the inhomogeneity of $T_c$ within the area probed by the two-coil apparatus. This peak occurs near the point where the K-T line crosses the superfluid data in each film (near the lower of the two peaks in the highest $T_c$ film), lending further evidence to the conclusion that we are seeing the K-T transition in a single copper oxide bilayer of Bi-2212. This differs from the downturn seen in thick sputtered films, which happens at the K-T line corresponding to the thickness of two CuO$_2$ bilayers.

Figure 4.7 shows $T_c$ vs. superfluid density, $\lambda^{-2}(0)$, on a log-log plot. The black squares are from thick, rough Bi-2212 films and Bi-2212 powders [46, 50]. The dashed line through these data indicate linear scaling over the range 7 K $\leq T_c \leq$ 50 K. The open circles in the lower data are from ultrathin Ca-YBCO films. They also show linear scaling over a significant range, 3 K $\leq T_c \leq$ 15 K. The solid diamonds overlaid on the Ca-YBCO data are from the new, smooth Bi-2212 films. Their improved smoothness has removed the apparent quantitative discrepancy with ultrathin Ca-YBCO. The quantitative agreement is evidence that the mechanism that connects $T_c$
Figure 4.7: Log-log plot of $T_c$ vs. superfluid density at $T = 0$. Upper data (black squares) are for thick, on-axis Bi-2212 films and Bi-2212 powder [50]. Lower data are for 2-unit-cell thick Ca-YBCO films (red circles) and the off-axis-grown Bi-2212 films (blue diamonds) shown in Fig. 4.6a. Dashed lines indicate linear scaling.

to superfluid density is the same in both systems, despite their structural differences.

In regards to thermal fluctuations, for our Bi-2212 films K-T theory predicts a transition at specific superfluid densities (indicated by black line in Fig. 4.6) assuming that individual CuO$_2$ bilayers fluctuate independently. The widths of the peaks in $\sigma_1$ indicate that the inhomogeneous spread in $T_c$ across the sample is a few percent to about 25%. The spread in $T_c$’s likely softens the abrupt downturn, but are not inhomogeneous enough to eliminate it altogether. This indicates that interlayer coupling is weak enough the thermal fluctuations are 2-D. Thus, the anisotropy in Bi-2212 is large enough to see 2D thermal critical behavior on the individual CuO$_2$ bilayer level when the films are smooth and thin enough to avoid smearing of $T_c$’s.

Finally there is the issue of the overall $T$-dependence of $\lambda^{-2}(T)$. The paucity of
curvature in films, and the very-nearly-linear $T$-dependence in YBCO crystals [48] immediately suggest that the $d$-wave gap dominates. In carefully grown crystals, one might believe that the elastic scattering rate is so small, i.e., less than a few Kelvins, that the $d$-wave linear-in-$T$ behavior might not be obscured by elastic scattering. Otherwise, it is unlikely that the elastic scattering rate is less than tens of Kelvins. A possible explanation for the linearity of thick Bi-2212 films lies in the notion that the underdoped superconducting state is highly correlated and thereby relatively insensitive to elastic scattering [51]. In our ultrathin Bi-2212 films elastic scattering appears to dominate, contributing to non-linear curvature of $\lambda^{-2}(T)$.

In this chapter, we catalogued the first ultra thin Bi-2212 films grown by PLD. We explored the physics of these films as they varied from thick Bi-2212 in some key ways. We showed $\lambda^{-2}(0)$ vs. $T_c$ was reconciled with ultra thin YBCO to prove both have the same dependency near the quantum critical point. By making Bi-2212 much thinner, we increased superfluid density by a factor of three for films of identical $T_c$. Finally, we uncovered the KT-transition at the single CuO$_2$ bilayer level in low $T_c$ films, unseen in thick Bi-2212.
Chapter 5
CONCLUSIONS

This dissertation represents several significant contributions to the field of superconductivity. In Chapter 2 we explored a different interpretation of $T_c$ vs. $d_F$ in S/F bilayers, which showed that the effective exchange energy $E_{ex}$ is nearly an order of magnitude smaller than the literature assumes, which means Cooper pairs dephase in the ferromagnet much less rapidly than expected. We also found higher interface resistance, $R_b$, meaning the tunneling rate of pairs between the S and F layers is less than previously thought. Then we turned to superfluid density, $\lambda^{-2}(T)$, to show that the rate of reduction in $\lambda^{-2}(0)$ with $T_c$ is as expected in the cases of Ni and Py, but CoFe causes a reduction in superfluid in an Nb film more than expected, likely due to $d_{Nb} << \xi_S$.

In Chapter 3, we analyzed Nb/Ni/Nb, Nb/NiCu/Nb and Nb/NiV/Nb trilayers containing two transitions. The $R_b$ for the top Nb/Ni interface was much larger than the bottom, such that $T_c$ for the top Nb layer was always larger than the bottom, even when the top layer was thinner. With Nb/NiV/Nb trilayers, BCS theory was employed to extrapolate $\lambda^{-2}(T)$ to $T = 0$ to obtain $\lambda^{-2}(0)$ to high accuracy. $\lambda^{-2}(0)$ for the thicker S layer was found with two-term quadratics. By analyzing the dependencies of $T_c$ and $\lambda^{-2}(0)$ on $d_{NiV}$, without bias, we found large oscillations; which we associate with oscillations in the order parameter known as an FFLO state. We also
found evidence of a $\pi$-junction causing superfluid to be suppressed to extremes that are otherwise difficult to explain by traditionally coupled trilayers.

Finally in Chapter 4, we looked at the physics near the Quantum Critical Point (QCP) of the first ultra thin BSCCO films grown by PLD. They had a higher $\lambda^{-2}(0)$ than thick BSCCO films. This resolved the discrepancy between the scaling of 2 uc YBCO films and thick BSCCO near the QCP. We were able to see 2D effects in BSCCO. These films were several times thicker than 2 uc YBCO, but because of the anisotropy of BSCCO, each superconducting layer was sufficiently decoupled that we found the K-T transition in single CuO$_2$ bilayers.
Bibliography


Appendix A

Collaborative Efforts

A.1 Introduction

Our superfluid measurement is of general interest to others in the thin film superconducting community. We forge collaborations around the world as researchers seek out more information about the quality and strength of their grown superconducting systems. Here we show results from Las Alamos and NTT Basic Research Laboratories in Japan. One involves combinations of YBCO with a non-superconducting BaZrO$_3$ (BZO) which spontaneously forms into alternating layers. The other is Nd$_2$CuO$_4$, which they were especially interested in finding the quality of their films.

A.2 Self Assembled YBCO:BZO Multilayers

Figure A.1 is data on two self assembled multilayers. The transition, shown in red and corresponding to the higher $T_c$ film is quite sharp. Thus, even though we are measuring several independent layers separated by insulating material, each YBCO layer has relatively uniform properties. The $T_c$’s are close to the maximum for YBCO, implying near-optimal doping. The films were grown on STO and are about 1500 Å thick. Since they contain 50% YBCO and 50% BZO and BZO is insulating, we divided by 750 Å for the effective thickness in determining the superfluid density.
Figure A.1: $\lambda^{-2}$ for layered YBCO$_{0.5}$/BZO$_{0.5}$ films of 150 nm thickness. A thickness of 75 nm was used to find the superfluid from the sheet conductivity. $\sigma_1$ (red) is relatively sharp for such a potentially inhomogenous film.

The slight increase beginning at around 35 K is an anomaly due to large screening. So values of $\lambda^{-2}(0)$ are about 10% less than indicated. The data at temperatures above the anomaly can be considered most accurate. For sample HY-184, $\lambda^{-2}(0) = 3.25 \mu m^{-2}$, and for sample HY-184, $\lambda^{-2}(0) = 4.0 \mu m^{-2}$, which are an order of magnitude below the expected value for YBCO with a $T_c$ of 85 K, which is around $30 \mu m^{-2}$. These two values are within the margin of error of the measurement, but there are reasons the samples could differ. If one looks at the TEM image in Figure A.2, while the YBCO and BZO make clear distinct layers, the interfaces are visibly rough and the layers are of varying size. These are not ideal crystal growth conditions, and yet, the layers are ordered enough that they transition as uniformly as a pure YBCO film.
A.3 Film Quality of Nd$_2$CuO$_4$

Michio Niato sent films of Nd$_2$CuO$_4$ grown on STO. They arrived in sealed glass containers that were carefully cut open with a file. Both films came with resistivity data which is shown in Figures A.3 and A.4. The first film measured had a lower residual resistivity, and so Niato expected this to be the higher quality of the two films. This turned out to be an excellent example of the benefits of superfluid density measurements beyond those of resistivity. When the film went superconducting at 25 K, there was no more information to be garnered from resistivity. Though this was the first instance of zero resistance in the film, superconductivity on the bulk scale did not begin until below 15K, and became substantial below 5 K. This observance is explained by the multiple peaks in $\sigma_1$. With each new peak more of the film transitions, contributing to the overall superfluid density.

The second film, Figure A.4, arrived with a resistivity three times higher than the
Figure A.3: $\lambda^{-2}$ (red), $\sigma_1$ (blue), and resistivity (green) of a Nd$_2$CuO$_4$ film. Despite having a lower resistivity value just above the transition, superfluid density reveals multiple transitions and the lack of substantial superfluid until 10 K.

first, but with a similar $T_c$ of 26 K. In this case, the bulk superfluid turned on at 24 K and increased steadily. There appears to be a single major transition and superfluid grows linearly with decreasing $T$. As a result, $\lambda^{-2}(0)$ is three times higher than the low resistivity film. Thus, values for $T_c$ and $\rho_0$ are not necessarily good proxies for the quality of a film, especially when we are dealing with complex cuprates that suffer from issues of crystallinity, interface roughness, and homogeneity more than pure metal and alloy films.
Figure A.4: λ−2 (red), σ₁ (blue), and resistivity (green) of a Nd₂CuO₄ film. Unlike Figure A.3, this film has a higher resistivity value just above the transition, superfluid density is non-neglegable at 24 K, only two degrees from the resistive transition. A much more robust film reviled by superfluid measurement.
Appendix B

Pulsed-Laser Deposition

There are a few major types of thin film growth employed throughout physics, chemistry, and material science including, sputtering, molecular beam epitaxy, and pulsed-laser deposition. We cover the last in this section.

Figure B.1 shows the general set up of the PLD system. The laser is supplied with a power source, cooling water, and gas mixtures. After a pulse leaves the laser, it hits a set of two mirrors that reflects the beam at 90° and rotates the elongated shape from horizontally oriented to vertically oriented. The pulse then passes through a
beam aperture, shaping it to a rectangular shape and allowing only the highest, most homogeneous part of the pulse through.

The beam travels to a lens near the entrance of the chamber. A power meter is placed before the lens to read the laser power. One of the mirrors that redirects and rotates the beam is adjusted until we obtain a reading of 180 mJ with the beam centered on the lens. With the power meter removed, the lens focuses the beam down to a size of 2.5 x 3.0 mm, yielding an energy density of 2.4 J/cm$^2$ at the target inside the growth chamber.

Most growth is done with a conventional “on-axis” geometry which places the sample substrate directly in the path of the plume. In figure B.2, an alternative off-axis configuration is shown, which we used to grow the first ultra thin BSCCO films made with PLD.

Traditional on-axis growth uses a slower pulse rate, usually 3 Hz. For off-axis growth, the maximum rate of the laser, 30 Hz was employed. The goal of high repetition growth is to put down a unit-cell quantity of material as quickly as possible followed by a delay, 60 - 120 s, to let the material spontaneously order into a crystal. As we saw in Chapter 4, this type of growth leads to tremendously smooth films compared to traditional growth.

In preparation for growth, the heater surface is sanded to remove old silver paint and deposited material from the previous growth. The substrate, usually STO, MgO, or LSCO is attached to the heater with silver paint. A drop of paint is allowed to fall from an application tool such as the non-cotton end of a cotton-tipped applicator. Then the substrate is dropped from tweezers a short distance above the paint, before the paint is allowed to thicken, ideally resulting in the paint wicking out across the majority of the bottom of the substrate and without the substrate sliding much from its intended position. Two separate substrates are used to lightly press down on the
corners of the substrate on the heater to allow for more even and complete contact.

The substrate is covered with a small crucible to protect from dust. The heater controller is manually increased from room temperature to 60 °C over the course of about 90 seconds, then the program increases at 0.1 °C/s up to 100°C, pauses for 10 minutes, and then cools to room temperature. Before the heater is placed into the chamber, a Kim-wipe with methanol is run across the surface, once, to remove any accumulated debris.

Ideally, the substrate is directly in front of the plume for on axis and as near to
the plume as possible for off axis. This is to prevent gradients and allow for uniform growth.

When preparing for growth, we run a routine for cleaning the surface of the target. This is done with substrate in place and the shield engaged. This routine is from 1000 to 3000 pulses and ablates the upper layer in hopes of removing condensed impurities. Most films use more than one target and our system allows for the transition between targets which is useful for more complex growth. Targets are periodically removed and sanded down to prevent indentations that would deflect the plume away from the substrate.

We employ a Lambda Physik LPX 135i Excimer Laser in our growths. The laser uses a Krypton Fluorine (KrF) mixture to yield pulses of wavelength 248 nm. Repetition rates between 1 Hz and 50 Hz are possible manually, while the program used for complex growth management is capped at 30 Hz. Pulse duration last about 100 ns.

Pulses enter our vacuum chamber which is pumped to pressures from $1 \times 10^{-7}$ torr to $2 \times 10^{-6}$ torr with no noticeable difference in superconducting properties of the grown films. Growth occurs in O$_2$ atmospheres of 300 mtorr flowing gas, with a replacement time of about 90 s. Pulses ablate a set of targets of various complex oxide superconductors and structurally similar buffering compounds.

Y$_1$Ba$_2$Cu$_3$O$_{8-\delta}$ films are grown at thicknesses between 2 uc (23.4 Å) and 170 uc (2000 Å). Single unit cell films are successfully superconducting much less often than 2 uc with little change in two dimensional superconducting properties.
Appendix C

Numerically Solving for $R_b$

The equation for the theory for finding $T_c$ is transcendental, and thus can only be solved numerically:

$$\ln \left( \frac{T_c}{T_{c0}} \right) = \Psi \left( \frac{1}{2} \right) - \Re \left\{ \Psi \left( \frac{1}{2} + \frac{1}{2\pi T_c \tau_S} \right) \right\} \quad (C.1)$$

This was a challenge when we wanted to solve it for many values of $d_F$ in order to fit plots. The solution was to write a program which solved each value to reasonable precision and with relative timeliness. We employed Mathematica to carry out the task.

To find $T_c$ as a function of $d_F$, for some $d_F$ we solve C.1 beginning at some fraction of $T_{c0}$, typically 1% is sufficient, though 0.1% is used at times to improve precision. The program solves the equation starting with $0.01 T_c$ and then checks if the output value is greater than the input value. If it is not, $0.02 T_c$ is attempted. When the condition is met, that value is outputted to the plot. $d_F$ iteration is also controlled for x-axis precision, typically at the 0.1 Å level. Depending on the plot we wish to fit, we have between 500 and 1000 data points, each of which take from 1 to 100 attempts to solve. Depending on desired precision, one run takes from 10 s to 600 s.

This leads to a trial and error system of finding the values that most closely match the data. Some values are fixed when fitting begins while others are adjusted. We
assume we know $v_F$ of the ferromagnet. Any error in $v_F$ will lead mainly to a equal magnitude error in $E_{ex}$. We obviously fix $T_{c0}$ since this is the unsuppressed Nb transition temperature of thickness $d_S$, which is also used in calculation. The values we calculate are $R_b$, $E_{ex}$, and $\xi_F$.

For instance, we look at a fit for data by Sidorenko in Fig. C.1. The $T_{c0}$ used to fit is higher than the data point shown because as far as we can tell this data point was not measured and it is unusually small for Nb. The fit gives values of $R_b = 2.8 \, f \Omega \cdot m^2$, $E_{ex} = 92$ K, and $\xi_F = 15$ Å, which are consistent with most measurements on other Ni/Nb bilayers though with a somewhat smaller $\xi_F$.

To get an idea how altering the values changes the shape of the fit, in Fig. C.2, we double the $R_b$ value which most notably raised the ultimate value of $T_c$. An intuitive response because a more opaque interface insulates the pair breaking effects of the ferromagnet. In Fig. C.3, the effective dephasing rate is halved, leading to a softer
Figure C.2: Same fit as Fig. C.1 with $R_b$ doubled.

Figure C.3: Same fit as Fig. C.1 with $E_{ex}$ halved.
bend in $T_c$ and a smaller increase in final $T_c$ than when we increased $R_b$ by the same factor. This is attributed to more Cooper pairs surviving to return to S unbroken. Finally, in Fig C.4, when we double $\xi_F$ the minimum extends out to nearly that value and the width of the minimum increases considerably. A larger $xi_F$ will tend to make the superconductor more “dirty” as it is more likely to be greater than the mean free path. This leads to a broadening of the ferromagnetic contribution to pair breaking.