NORMAL AND SPIN POLARIZED TRANSPORT IN HIGH-TEMPERATURE SUPERCONDUCTOR TUNNELING JUNCTIONS

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ABSTRACT OF DISSERTATION

Mario Vadim Freamat

The Graduate School
University of Kentucky
2004
NORMAL AND SPIN POLARIZED TRANSPORT IN HIGH-TEMPERATURE SUPERCONDUCTOR TUNNELING JUNCTIONS

ABSTRACT OF DISSERTATION

A dissertation submitted in partial fulfillment of the requirements for the degree of Doctor of Philosophy in the College of Arts and Science at University of Kentucky

By

Mario Vadim Freamat

Lexington, Kentucky

Director: Dr. Kwok-Wai Ng, Professor of Physics and Astronomy

Lexington, Kentucky

2004

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ABSTRACT OF DISSERTATION

NORMAL AND SPIN POLARIZED TRANSPORT IN HIGH TEMPERATURE SUPERCONDUCTOR TUNNELING JUNCTIONS

One of the challenges facing condensed matter physics nowadays is to understand the electronic structure of high temperature superconductors. This dissertation compiles our contribution to the experimental information concerning this subject. Tunneling conductance spectroscopy – a technique capable of probing the electronic density of states in hybrid structures – was used to study the current and spin transport properties across junctions between metallic counterelectrodes and Bi₂Sr₂CaCu₂O₈₋ₓ (BSCCO) crystals. Since in these structures the transport is mediated by transmission channels depending on superconductive characteristics, the energy resolved density of states is a signature of the mechanism of superconductivity. For instance, one can observe the superconductive energy gap and the behavior of subgap bound states due to phase sensitive Andreev reflections at the junction interface. In particular, tunneling spectroscopy makes possible the observation of the LOFF state – characterized by the coexistence of superconductivity and magnetism.

Cuprates like BSCCO are highly anisotropic materials and their superconductivity is almost two dimensional, being confined in the CuO₂ planes. Therefore, our junctions combine monocrystals of underdoped samples of BSCCO with various thin film counterelectrodes – normal metal (Ag), conventional superconductor (Pb) and ferromagnetic metal (Fe) – deposited perpendicular onto the cuprate $ab$-plane (CuO₂ plane).

We performed measurements on Ag/BSCCO junctions for two current injection directions into the same crystal. We observed that, near the ⟨110⟩ crystal surface, the conductance spectra show a high zero bias peak (ZBCP) which is a manifestation of zero energy
Andreev bound states due to an anisotropic superconductive order parameter. Near the \(\langle 100\rangle\) surface, the ZBCP is largely suppressed. This is consistent with a predominantly \(d_{x^2-y^2}\)-wave pairing symmetry. In some cases, the ZBCP splits or decreases in amplitude at low temperatures. This is consistent with the existence of a subdominant \(s\)-wave (or \(d_{xy}\)) resulting in a mixed \(d+i s\) state which breaks time reversal symmetry (BTRS). Since we observe this phenomenon in the underdoped case, we do not confirm the possibility of a quantum critical point close to the optimal doping. Our Pb/BSCCO spectra contradict the theory explaining the BTRS by proximity effect. The Fe/BSCCO junctions measure the effect of spin polarization. We explain the recorded 4-peak asymmetric structure by the combined effect of a spin independent BTRS state and a spin filtering exchange energy in the barrier responsible for a large ZBCP splitting.

The LOFF state was observed in the proximity region induced on the ferromagnetic side of multilayered-Fe/Ag/BSCCO structures. As expected for the LOFF order parameter, the spectra develops coherent damped oscillations with the Fe layer thickness probing different regions. The magnitude and sign of the oscillation depends on the energy. The conductances at energy zero or equal to the superconductive gap are modulated in antiphase proving that the order parameters takes successively positive and negative values. Changing the junction orientation with \(\pi/4\), results in an opposite behavior for the same distance. The maximal amplitudes in one direction is replaced by minima, showing that, besides space, the LOFF state modulation depends on the phase of the high temperature order parameter inducing the proximity region.

KEYWORDS: \(d\)-wave superconductivity, tunneling spectroscopy, Andreev reflection, spin polarized tunneling, LOFF state.

Mario V. Freamat

June 28, 2004
NORMAL AND SPIN POLARIZED TRANSPORT IN HIGH-TEMPERATURE SUPERCONDUCTOR TUNNELING JUNCTIONS

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DISSERTATION

A dissertation submitted in partial fulfillment of the requirements for the degree of Doctor of Philosophy in the College of Arts and Science at University of Kentucky

By
Mario V. Freamat
Lexington, Kentucky

Director: Dr. Kwok-Wai Ng, Professor of Physics and Astronomy
Lexington, Kentucky
2004

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Dedicated to my wife and my daughter
Aknowledgments

This study owes a lot to my advisor, Dr. Kwok-Wai Ng, who inspired my dedication to experimental physics. I also substantially benefitted from my discussions with Dr. Joseph Brill.

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Introduction

It is widely accepted today that the progress in science is a result of a change in the scientific method, either according to Popper’s model of gradual growth or to Kuhn’s ideology of discrete, revolution based, evolution. In physics, an example of such a methodological change (not quite a change in paradigm but certainly a challenge to readapt) appeared when the perturbative methods - employed by condensed matter theories trying to explain cooperative effects in solids - failed to provide a microscopic picture or a lucrative description for the many-body ground state of a class of materials characterized by strong interaction between electrons. These compounds are commonly known as Strongly Correlated Systems, a theoretical framework including “unconventional” superconductors (cuprates, organic superconductors, heavy fermions), magnetic systems exhibiting ferromagnetism or antiferromagnetism (with the special case of the manganites), quantum Hall systems, one dimensional electron systems, etc. While manifesting a wide range of similar phenomena like the competition between different phases and an energy gap with a different origin than the band-gap, the properties of these materials still cannot be explained by a general theory. Consequently, while waiting for this scientific Godot, we have to accept a case by case approach or, faute de mieux, try to catch a glimpse of the unified explanation from the specific interactions between the different phases resulting from the respective strong correlations.

Consistent with this program, the main motivation of this study is to explore experimentally the electronic structure of high temperature superconductors via the symmetry of the electron-electron interactions leading to superconductivity, as well as to investigate the interaction between superconductivity and ferromagnetism. In particular, we observe the effect of a spin polarized current on the typical transport channels across superconductive tunneling junctions (like the Andreev bound states). We also probe an “exotic” state which arise when the superconductive Cooper pairs traveling in a ferromagnet dephase due to the presence of an exchange potential which tries to polarize the electronic spins. This rather elusive state - called LOFF, from the names in the two groups which independently predicted it theoretically in 1964 -
is qualitatively different from a conventional superconductive state described by the Bardeen-Cooper-Schrieffer (BCS) theory based on the phonon intermediated interactions between electrons forming Cooper pairs with long-range phase coherence. As example, while magnetism is mutually exclusive with a BCS state since an external magnetic field couples to orbital motion of the electrons, the superconductivity of a LOFF state can be even enhanced by a magnetic field, due to its coupling to the spins of the electrons.

The situation becomes more heuristic when the LOFF state arises from an unconventional type of superconductivity like the one characterizing some perovskite systems. This type of superconductivity, with critical temperatures as high as 135 K, was first discovered in appropriately doped cuprates by Bednorz and Müller in 1986 and, since then, despite the considerable progress, it continues to puzzle. For instance, in these systems there seems to be a phenomenological connection between magnetism and a flavor of non-BCS superconductivity with a pairing mechanism possibly based on spin fluctuations still not completely elucidated. The magnetic fluctuations may be responsible for the long-range phase coherence between the superconductive electron pairs which in unconventional superconductors are too small in size to communicate by the overlap of their wave functions as in the BCS theory. Particularly in the cuprates, the intimate relationship between magnetism and superconductivity also stems from the fact that the magnetic properties of these materials are inherited from their parent compounds which are antiferromagnetic Mott insulators. In fact, this apparent link between high-\(T_c\) superconductivity and magnetism was one of the reasons which led in the last years to a renewed interest in the LOFF state and the spin-transport effects in superconductive structures. Another motivation originates from a whole class of potential and concrete technological applications, including quantum-computational (qubits) and spintronic devices (spin valves, magnetic tunnel transistors, etc.).

This dissertation is a compilation of our experimental results collected on tunneling heterostructures with various architectures. Tunneling spectroscopy – a powerful technique probing the density of states of quasiparticle excitations in the superconductor – was employed to study superconductivity in Bi\(_2\)Sr\(_2\)CaCu\(_2\)O\(_{8-\delta}\) (BSCCO) crystals in proximity with different metallic counterelectrodes. The measurements and discussions regarding spin polarized tunnel junctions of the type ferromagnet/BSCCO are presented within the framework of the results obtained on nonmagnetic junctions such as normal/BSCCO metal and conventional superconductor/BSCCO.
In Chapter 1: Conceptual framework, we review succinctly the studied phenomena - superconductivity and ferromagnetism - with an emphasis on high temperature superconductivity and the methodological principles of tunneling spectroscopy applied to superconductors with anisotropic order parameter (predominantly $d$-wave). Besides a utilitarian introduction regarding superconductivity, ferromagnetism and the LOFF phase, we brief on some theoretical models explaining the spectra obtained on tunneling junctions – models used ubiquitously throughout this text. The concrete case of ferromagnet/superconductor junctions is discussed thoroughly.

Chapter 2: Experimental setup details the junction fabrication process and experimental procedures. The component materials (BSCCO crystals and thin film counterelectrodes) are characterized and the thin film reliability is evaluated.

In Chapter 3: Tunneling measurements and spin polarization effects, we present a series of our measurements performed on Ag/BSCCO, Pb/BSCCO and Fe/BSCCO tunnel junctions in the $ab$-plane of the cuprate crystal. We investigate a multitude of aspects of high temperature superconductivity like the pure $d$-wave or composite pairing symmetry and the energy shift of spectral peaks resulting from Andreev bound states. In the case of spin polarized current, we confirm the possibility of simultaneous peak splitting effects with different origins: on one hand due to a broken time reversal symmetry and, on the other hand, by spin filtering in the junction interface.

Chapter 4: Probing LOFF state deals with the LOFF phase in the proximity region of a multilayered BSCCO/Fe junction, a subject intensely studied theoretically but rather poorly investigated experimentally on cuprates. Some of the properties of its exotic order parameter – even more remarkable when arising from an unconventional superconductivity - will be observed in the proximity region of the ferromagnetic counterelectrode.

Finally, in the Appendix, the computer implementation of the models used throughout the dissertation will be briefly listed.
CHAPTER 1
Conceptual Framework

1.1 Superconductivity

Under a critical temperature, some materials undergo a phase transition to a superconductive state, phase characterized by zero electrical resistivity and perfect diamagnetism. The serendipitous experimental discovery of superconductivity by Kamerlingh Onnes in 1911 (Fig.1.1a) long preceded the theoretical means for understanding its microscopic mechanism – i.e. the quantum theory of normal metals. This is why, for decades, most of the knowledge about this state - described by Fritz London as a “quantum phenomenon on a macroscopic scale” [1] - came from experimental investigations or phenomenological theories. Year after year, its basic properties have been unveiled in a series of experiments and theoretical constructs intimately related to the developments in technology and paradigmatic revolutions in Physics. Eventually, understanding of conventional superconductivity culminated with the BCS theory. On the other hand, unconventional superconductivity is still a matter of intense debates. The next sections serve not only to point out to the main events throughout this saga, but also to introduce the concepts and theoretical framework necessary to understand the experiments presented in the following chapters. A special emphasis is placed on the interaction between magnetic fields and superconductors.

1.1.1 Phenomenon and developments. BCS theory.

Besides the discovery of the superconductive effect, Onnes [2] also observed that the superconductive state disappears under the effect of an external magnetic field exceeding a critical value $H_c$ characteristic to the material. Moreover, he noticed that a current larger than a critical value $I_c$ led to the same result, but Silsbee proved later that the two effects are related. In the following years, a multitude of materials were shown to present the superconductive transition, albeit at very small temperatures (e.g., Th ($T_c \approx 1.4$ K), Ta ($T_c \approx 4.4$ K), Pb ($T_c \approx 7.2$ K) or Nb ($T_c \approx 9.2$ K)). We owe the next significant discovery to Keesom and Van den Ende [3] who observed in 1932 a bump in the electronic specific heat at the transition temperature, feature
which hinted *avant la lettre* to the existence of a gap in the superconductive electronic density of states.

The set of fundamental properties of the superconductive state was completed next year, when Meissner and Ochsenfeld [4] established that the superconductors are not only perfect conductors but very good diamagnets as well. Thus, with the onset of superconductivity, in the presence of an external magnetic field lower than a critical value $H_c$, the magnetic flux is completely expelled from the interior of the material with exception of a thin penetration layer at the surface and, if the external field is removed, no flux or induced dipole is trapped inside. The constant flux is maintained by stable (or metastable) currents flowing in this penetration layer of thickness $\lambda \approx 500 \text{ Å}$. Denoting $F_s$ and $F_n$ the density of free energy in the superconductive and normal phases respectively, this *Meissner effect* implies that superconductivity is destroyed by a magnetic field $H_c$ such that:

$$F_s(T) - F_n(T) = -\frac{H_c^2(T)}{8\pi}. \quad (1.1)$$

Based on the Meissner effect, the materials in the realm of superconductors can be categorized into Type I (presenting a perfect diamagnetism) and Type II (mixed diamagnetism at

![Figure 1.1](image.png)

*Figure 1.1  a) The classical transition to perfect conductivity, as first observed by Onnes. b) Magnetic phase diagram for Type II superconductors (specifically for cuprates). Between $H_{c1}$ and $H_{c2}$ (Shubnikov phase), the uniform superconductive state is replaced with two complex vortex phases. At low temperatures, the transition to normal state is preceded by a narrow region of superficial superconductivity. Also, the penetration magnetic field is represented as a function of applied field for a cross-section in the phase diagram (that is a constant temperature value).*
high fields). The Type II superconductors (like Nb) have two critical magnetic fields $H_{c1}$ and $H_{c2}$: the flux is completely expelled from the bulk of the material under $H_{c1}$, but, above this value, it gradually penetrates the inside until the superconductivity is completely destroyed at $H_{c2}$ (Fig. 1.1b).

Meissner effect suggests that thermodynamics could be applied to the superconductive state. This had as an immediate consequence the first serious theoretical attempts to understand it: the two-fluid model developed by Gorter and Casimir [5] - which proved useful in describing the thermal properties of the equilibrium state -, followed by the semi-empirical theory developed by London brothers in 1935 [6]. The two-fluid model is an ansatz for the free energy of the superconductive state and suggests that this state can be described by the interplay between two phases characterizing the sea of conduction electrons: a normal one, represented by a density $n_n$ of electrons scattered ohmically as in a normal metal, and a superfluid phase, corresponding to a density $n_s$ of electrons less scattered, in principal affected by external electric fields. Hence, the total density of electrons is $n = n_n + n_s$. This model emphasize mainly the electric properties of the superconductive condensate and is nowadays known for inspiring the London theory which elaborates on the diamagnetic characteristics and actually proved to be a particular case of the modern microscopic BCS theory, as we shall see further below. Thermodynamically speaking, London theory is also based on the minimization of the free energy conveyed by the superfluid component in the two-fluid model, component which becomes dominant over the normal one in the superconducting phase.

London theory implies the Meissner effect by supplementing Maxwell equations with a set of electrodynamic equations particularly describing the penetration limits of the magnetic field into a superconductor. Thus, by assuming that, in the superconductive phase, the superfluid component of the two-fluid condensate transports a supercurrent with density $\mathbf{J}$ of $n_s$ carriers (electrons or holes, depending on the superconductor) encountering no resistance, London theory assumes that:

$$\frac{\partial \mathbf{J}}{\partial t} = \frac{n_s e^2}{m^*} \mathbf{E}, \quad (1.2)$$

$$\nabla \times \mathbf{J} = -\frac{n_s e^2}{m^* c} \mathbf{B}, \quad (1.3)$$
where \( m^* \) is the effective mass of the carriers. At low temperatures and assuming a slowly varying current which entails neglecting the displacement current (consistent with a good conductor), the supercurrent is

\[
J = -c/4\pi\lambda_L^2 A
\]  

(1.4)

and dominates the normal current so that, using Maxwell equation \( \nabla \times B = 4\pi J/c \) in expression (1.3), we obtain

\[
\nabla^2 B = \frac{1}{\lambda_L} B,
\]  

(1.5)

with \( \lambda_L \) being the London penetration depth, a characteristic length scale generally defined [7] by

\[
\lambda = B_0^{-1} \int B(r)dr,
\]  

(1.6)

where \( B_0 \) is a static magnetic field applied normal to the superconductive surface.

London’s theory gives at \( T = 0 \)

\[
\lambda_L = \sqrt{mc^2/4\pi e^2 n_s},
\]  

(1.7)

or \( \lambda_L \approx 100 \text{ Å} \). In practice, most of the measured penetrations are larger than this value due to the limitation in the original London theory as we shall discuss later.

The solution of (1.5) is an excellent qualitative illustration of the Meissner effect: as example, a constant external magnetic field \( B = (0, 0, B_0) \) applied perpendicular onto the \( xy \)-surface of a superconductor decays exponentially inside the material, with the superficial penetration being characterized by \( \lambda_L \):

\[
B(z) = B_0 e^{-z/\lambda_L},
\]  

(1.8)

a result phenomenologically consistent with the Meissner effect. Note that \( \lambda_L \to \infty \) when \( T \to T_c \) since \( n_s \to 0 \). Also, when \( T \to 0 \), \( n_s \to n \), and \( \lambda_L(T) = \lambda_L(0) = (mc^2/4\pi e^2 n)^{1/2} \). But, according to an empirical intuition of Gorter and Casimir, the dependency on temperature of the superfluid density takes the form

\[
n_s = n \sqrt{1 - T^4/T_c^4}.
\]  

(1.9)

Consequently, the temperature dependency of the penetration depth should be

\[
\lambda_L(T) = \lambda_L(0) / \sqrt{1 - T^4/T_c^4},
\]  

(1.10)
which is a qualitative result working fairly well with most conventional superconductors.

The next important development came with the discovery of vortices. In 1937, Shubnikov et al. [8] found a mixed phase around the critical magnetic field of a Type II superconductor. Whereas under the effect of a sufficiently strong external magnetic field a Type I superconductor exhibits a sharp transition to uniform normal state, a Type II superconductor is evolving from phase to phase between $H_{c1}$ and $H_{c2}$, being first permeated by a dense array of magnetic vortices, whirlpools of electric current swirling around islands of normal cores. If the external magnetic field increases, the normal regions gain terrain and merge together, up to the upper magnetic field value $H_{c2}$, where the entire material turns normal. Insofar as the current vortices are oriented parallel to the applied field, they repel each other like tiny bar magnets, so that they arrange themselves in a regular pattern, called the Abrikosov flux lattice after the theoretician who predicted it 20 years later [9].

A keystone in the edifice of conventional superconductivity was the discovery of the isotope effect [10, 11]. In 1950, two independent groups demonstrated that the critical field at zero temperature and the transition temperature $T_c$ vary with the isotope mass $M$:

$$H_c \approx T_c \approx M^{-\alpha},$$

(1.11)

with $\alpha \approx 0.45 \div 0.50$ for most superconductors. This hinted to a microscopic mechanism behind the appearance of the superfluid based on the interaction between electrons and quanta of lattice vibrations or phonons. Almost at the same time, the idea was being developed theoretically by H. Fröhlich [12] who, applying a perturbative approach to a Hamiltonian including interactions between electrons and phonons, found an instability of the Fermi surface provided the interaction is strong.

A significant theoretical contribution was due to V. Ginzburg and L. Landau [13] who used Landau’s theory of second-order phase transitions to extend London conjecture in order to include the spatial variations of the superfluid condensate around the critical temperature. Ginzburg-Landau (GL) theory is independent of the underlying mechanism of superconductivity and is limited to the normal-superconductor phase boundary. It employs Landau’s notion of a complex order parameter $\varphi$ characterizing the superconductive state as a function of temperature (vanishing above $T_c$), magnetic field and position, $\varphi = \varphi(T, \mathbf{B}, \mathbf{r}) = |\varphi|e^{i\phi}$. The difference between
the densities of free energy of the superconductive and normal phases can be expanded in terms of this order parameter $\phi$.

$$F_s - F_n = \alpha |\phi|^2 + \frac{1}{2} \beta |\phi|^4 + \frac{1}{2m} (-i\hbar \nabla - qA)^2 \phi^2 + \frac{1}{8\pi} |B|^2,$$  

where $\alpha$ and $\beta$ are phenomenological parameters dependent on temperature with $\alpha(T_c) = 0$ and $q$ is twice the charge of an electron, $|q| = 2|e|$. Since $\phi$ is given by the critical points of the energy functional, one can minimize this expression with respect to the fluctuations in $\phi$ and $A$, leading to the time-independent GL equations:

$$\frac{1}{2m^*} (-i\hbar \nabla - qA)^2 \phi + \alpha \phi + \beta |\phi|^2 \phi = 0$$  

(1.13)

$$J = -\frac{i\hbar q}{2m^*} (\phi^* \nabla \phi - \phi \nabla^* \phi^*) - \frac{q^2}{m^* c} \phi^* \phi A.$$  

(1.14)

The first equation describes the behavior of the order parameter $\phi$ in a magnetic field. The second equation, submitting supercurrent $J$, is almost similar to London’s expression (1.4), except that it is space dependent through $\phi$. One of the most important consequences of this model was the prediction of two length scales characterizing the superconductive state:

1) The penetration depth, which is a measure of the superfluid density, or the penetration scale of a magnetic field into the surface of a superconductor,

$$\lambda = \sqrt{m^* c^2 / 4\pi q^2 \phi_0^2},$$  

(1.15)

where

$$\phi_0^2 = |\phi_0(T)|^2 = |\alpha(T)|/\beta(T)$$  

(1.16)

is the zero-field equilibrium value of the order parameter which can be replaced in (1.14) to obtain – provided the sample is much larger than $\lambda$ - an expression similar to London’s supercurrent (1.4):

$$J = -\frac{q^2}{m^* c} |\phi_0|^2 A.$$  

(1.17)

Consistently, if we consider the order parameter as a measure of the coherence inside the superfluid condensate and so we set $\phi_0 = n_s/2$, (1.15) becomes identical to London’s penetration depth (1.7).
2) The *coherence length*, which provides a spatial scale for the superconductive phase fluctuations at the boundary with the normal state:

\[ \xi = \sqrt{\frac{\hbar^2}{2m^*|\alpha|}}. \]  

(1.18)

This quantity can be derived immediately by assuming a zero electromagnetic field and defining a space dependent solution for (1.13) of the form \( \phi(r) = \phi_0 + f(r) \). In this case, by (1.13), \( f(r) \) satisfies an ordinary differential equation:

\[ \frac{\hbar^2}{4m^*|\alpha|} \nabla^2 f - f = 0, \]  

(1.19)

which implies naturally that, at the superconducting-normal interface, there is a finite transition layer, a *proximity region*, where the order parameter decays into the normal zone along several space intervals given by \( \xi \). The difference between \( \lambda \) and \( \xi \) consists in that \( \lambda \) depends on the energy necessary to modify the magnetic field, while \( \xi \) measures the energy cost to modify the phase of the order parameter.

---

**Figure 1.2**  
(a) Local magnetic field penetration as predicted by London theory (dashed line) and the nonlocal BCS-Pippard version (solid line). As visible on the inset, the nonlocal profile is not exponential and has a reversed field region before decaying to zero.  
(b) Spatial distribution of magnetic field \( B \) and order parameter \( |\phi| \) flux lines in a Type II superconductor, calculated from Ginzburg-Landau equations for three \( \kappa = \lambda/\xi \) values. In the inset, the \( B \) and \( |\phi| \) profiles are represented at the boundary of a Type II superconductor with a normal metal. In the absence of \( B \), the order parameter decays smoothly in a proximity region of the normal metal. Otherwise, it grows inside the S region with the decaying \( B \).
The characteristic lengths $\lambda$ and $\xi$ depend on temperature and the relation (1.9), divergent at $T_c$, suggests a dependence of the form \( (1 - T^4/T_c^4)^{-1/2} \) which agrees with most experiments. Nevertheless, the ratio $\kappa = \lambda/\xi$, dubbed Ginzburg-Landau parameter, is approximately independent of temperature and it was shown that $\kappa$ can distinguish between the two types of Meissner effects: thus, $\kappa < 1/\sqrt{2}$ for Type I superconductors and $\kappa > 1/\sqrt{2}$ for Type II.

GL theory and the enhancement of London equations by A. B. Pippard [14] structured the understanding of superconductivity and inspired a plethora of theoretical developments and predictions. In particular, an improved notion of coherence length was useful in explaining the interaction between superconductivity and magnetic fields. Since London’s penetration depth (1.7) proved to be somewhat inaccurate, experimental observation usually measuring deeper magnetic field penetrations, Pippard proposed an adapted, non-local expression for the supercurrent density $\mathbf{J}(\mathbf{r})$ which, for almost constant magnetic fields, takes a form reminding London’s supercurrent:

$$
\mathbf{J}(\mathbf{r}) = -\frac{m^*}{e^2 n_s} \frac{\xi}{\xi_0} \mathbf{A}(\mathbf{r}). \tag{1.20}
$$

Pippard’s supercurrent assumes that $\mathbf{J}(\mathbf{r})$ at the position $\mathbf{r}$ inside the superconductor feels the perturbations in superfluid within a distance $\xi_0$ around that point, due to some sort of coherence manifested by the electrons in the volume of the “wave packet formed by the electronic states”. In other words, the wave function associated with the electrons in the superconducting state is not localized so that, at the transition temperature, only the electrons with energy within a gaplike interval $\approx k_B T_c$ from Fermi energy can participate in the transition, this energy corresponding by the uncertainty principle to a spatial smearing of the supercurrent response to a perturbation. Hence, any influence – like the application of an external magnetic field – is spatially scaled by an effective coherence length $\xi$ depending on the electron mean free path $\ell$

$$
\frac{1}{\xi} = \frac{1}{\xi_0} + \frac{1}{\ell}. \tag{1.21}
$$

Therefore, the electron correlation in the superfluid has a maximal spatial span $\xi \approx \xi_0$ in the case of low impurity density ($\ell \to \infty$) in the superconductive material. For pure metals at temperatures
$T \ll T_c$, Pippard used the uncertainty principle [15] to relate $\xi_0$ to the normal-superconducting transition temperature, $T_c$:

$$\xi_0 \approx a \frac{\hbar v_F}{k_B T_c}, \quad (1.22)$$

where $v_F$ is the Fermi velocity and $a \approx 0.18$ was obtained later from the microscopic theory.

Pippard’s supercurrent leads to a more intricate perspective about the behavior of the magnetic field in the penetration layer. Thus, knowing that the effect of a magnetic field on a superconductor is measured by the induced supercurrent density [16-18], one can define the Fourier transform of the generalized supercurrent $J(k)$ in terms of a tensorial electromagnetic response kernel $K(k)$ and the vector potential $A(k)$:

$$J(k) \propto K(k) A(k). \quad (1.23)$$

This kernel is connected to the applied magnetic field $B_0$ [7, 19] by:

$$B(r) \propto B_0 \int \frac{k \sin(kr)}{\mu_0 K(k) + k^2} dk \quad (1.24)$$

so that, substituting in the definition (1.6) for penetration depth, we see that

$$\lambda \propto K^{-1}. \quad (1.25)$$

Since the kernel corresponding to Pippard’s expression for supercurrent is always smaller than London’s kernel, we deduce that the penetration depth is larger than the value predicted by (1.7), which is approached only in certain conditions. Specifically, when $\xi \ll \lambda$,

$$\lambda \approx \lambda_L \sqrt{\frac{\xi}{\xi_0}} \quad (1.26)$$

allowing $\lambda \approx \lambda_L$ in pure Type II superconductors. Thus, Pippard’s conjecture contains London’s theory as a particular case. In this case, the Meissner decay of the magnetic field does not take into account the variation of $A$ in the negligible $\xi$ volume, so the description is still local and exponential. Alternatively, if $\xi \gg \lambda$, represented by Type I superconductors,

$$\lambda \approx 0.65 \left(\frac{\xi_0^2 \lambda_L^2}{\xi}\right)^{1/3}, \quad (1.27)$$

so that $\lambda$ does not depend on $\xi$, i.e., on the impurities in the material. In this case, the non-local decay (1.24) applies which involves a field reversal before the field tends to zero (Fig.1.2a) [19].
As noticed a few paragraph before, the destruction of superconductivity by magnetic field in a Type II superconductor is characterized by a transitional penetration of magnetic field between a lower critical field $H_{c1}$ and an upper field value $H_{c2}$. GL theory proved again its virtues being used by Abrikosov [9] to describes this mixed phase. He calculated a two-dimensional periodic solution of the GL equations interpreted as a lattice of flux lines or vortices, each vortex of circulating supercurrent carrying one quantum of magnetic flux $\Phi_0 = \frac{h}{2e} = 2.07 \times 10^{-15}$ Tm$^2$. In fact [7], as visible in Fig.1.2b, the vortex is a tube of radius $\lambda$ enclosing a core of radius of approximately $\xi$ with maximum magnetic field and order parameter $|\varphi(r)| = 0$. If the applied magnetic field $H_0 < H_{c1}$, the superconductor is in the Meissner state, with the magnetic only superficially penetrating the material. As $H_0$ ranges between $H_{c1} \approx \Phi_0 \ln(\sqrt{2})/4\pi\mu_0 \lambda^2$ and $H_{c2} = \Phi_0/\mu_0 2\pi\xi^2$, the vortex lattice gradually grows. Eventually, the flux tubes overlap and the order parameter throughout the material becomes zero everywhere, the superconductive state being completely eliminated. The flux lines can be displaced by pinning forces or thermal fluctuations and the elastic response of the vortex lattice becomes important when the Type II superconductor possesses a degree of anisotropy [17] as we shall see it is the case of high temperature superconductors.

However, in the early 50’s, even with the subtle GL theory and the subsequent developments, the impulse for a much awaited microscopic approach came from the intuitions regarding the possibility of an interaction between super-electrons attractive in nature via an exchange of virtual acoustic phonons. This negative (attractive) term was already present in Fröhlich’s Hamiltonian [12] and became crucial in the work of J. Bardeen [20]. Equally fundamental was the suspicion enforced by some experimental data about the existence of an energy gap close from the Fermi surface [21]. Experimental evidence about this had been around since as early as 1932 in the rather inaccurate specific heat measurements – however, interpreted as such only much later. In the 40’s, an electronic quantum excitation possibly due to a gap was observed in a set of experiments dealing with microwave absorptivity of superconductors. Bardeen realized the importance of this concept and suggested [22] that Pippard’s electrodynamics most likely follows from an energy gap. Together with L.N. Cooper and J.R. Schrieffer, he initiated an effort to understand the mechanism of electron condensation within an
energy $k_B T_c$ from Fermi level. Conveniently, Landau’s theory of Fermi liquid was available as the standard model for metallic state based on adiabatic continuity. Adapting this theory, the starting point was to identify a ground state wave function as a superposition of normal state configuration consisting in *quasiparticles* – or fermions “dressed” by the medium in which they are immersed – excited above Fermi level at an energy $\nu_r \Delta p \approx \nu_F \hbar / \xi \approx k_B T_c$ consistent with Pippard’s coherence length. Their effort resulted in the theory of microscopic mechanism of Type I superconductivity called by their initials BCS theory [23]. Its central concept is the phonon intermediated pairing of electrons into *Cooper pairs*, the interaction between pairs being approached by Bardeen based on a previous model developed for polaron by D. Pines [24]. About $n \cdot 10^{-4}$ of the total number of electrons near Fermi surface bind into these pairs which act like giant, overlapping bosons condensed into a zero momentum state within an energy interval from Fermi level equal to the energy necessary to split a pair. The coherent superposition of paired quasiparticles form macroscopic wave packets able to transport unscattered charge and spins.

Concretely, Cooper [25] proved that, at low temperature, due to the balance between the electron-electron and electron-phonon interactions which becomes arbitrarily attractive for a range of electron frequencies, the Fermi surface becomes unstable with respect to the formation of bound states (in the momentum space) of paired electrons with zero total momentum and opposite spins and momenta, $(k_\uparrow, -k_\downarrow)$. Thus, if we consider a properly antisymmetrized wave function of the pair with the center of mass at $r = r_\uparrow - r_\downarrow$, containing the respective spin functions $\alpha_i, \beta_i$, 

$$
|\Psi_0(r)\rangle = (\alpha_1\beta_2 - \alpha_2\beta_1) \sum_k g_k \cos(kr),
$$

and we insert it into Schrödinger equation, we obtain:

$$
(E - 2\varepsilon_k) g_k = \sum_{k' > k_\uparrow} g_k' V_{kk'},
$$

where $\varepsilon_k = |k|^2/2m$ and $V_{kk'} = V_{k'k}$ is the phonon interaction potential [26] in momentum space. Since presumably most of the states involved in the superconducting transition are within a typical phonon energy (or Debye energy) $\hbar \omega_D$ from the Fermi energy $\varepsilon_F$, Cooper made the following assumption about the potential:
\begin{equation}
V_{k,k'} = \begin{cases}
-V & |\delta_k| < \hbar \omega_D,\ \delta_k = \epsilon_k - \epsilon_F, \\
0 & \text{otherwise}
\end{cases}, \quad (1.30)
\end{equation}

such that we can rearrange (1.29) to get:
\begin{equation}
\frac{1}{V} = \sum_{k>k_F} \frac{1}{2\epsilon_k - E}.
\end{equation}

Transforming the sum into an integral and introducing the pair density of states at the spherical Fermi surface \( N_0 = \frac{k_F}{\pi^2 v_F} \), it follows that
\begin{equation}
\frac{1}{V} = \frac{1}{2} N_0 \int_{\epsilon_F}^{\epsilon_F + \hbar \omega_D} \frac{d\epsilon}{2\epsilon - E} = \frac{1}{4} N_0 \ln \frac{2(\epsilon_F + \hbar \omega_D) - E}{2\epsilon_F - E}.
\end{equation}

Therefore, in the weak coupling approximation \( N_0 V \ll 1 \), the energy is
\begin{equation}
E = 2\epsilon_F - 2\hbar \omega_D \frac{1}{e^{\frac{\epsilon_F + \hbar \omega_D}{2\epsilon_F - E}} - 1} \approx 2\epsilon_F - 2\hbar \omega_D e^{-4N_0V}.
\end{equation}

That is to say, a bound state is formed with binding energy \( \epsilon_B = 2\hbar \omega_D e^{-4N_0V} \). Moreover, defining \( N = \sum g_k \), the wave function becomes:
\begin{equation}
|\Psi_0(r)\rangle = N \sum_{k>k_F} \frac{\cos (k \cdot r)}{2\delta_k + \epsilon_B}, \quad (1.34)
\end{equation}

so that the represented state decays rapidly with increasing \( \delta_k \). Since \( \epsilon_B \propto T_c \), the size of Cooper pairs is of the order of Pippard’s coherence length \( \xi \), as given by (1.22). However, since in the volume corresponding to \( \xi \) there are about \( 10^{11} \) electrons, the pair wave function was insufficient to explain the superconductive concentrate, so that the BCS theory took the next step by building a wave function for all electrons with energy close to the Fermi surface.

The BCS Hamiltonian represents the attractive pairing interaction [23, 27] as following:
\begin{equation}
H = \sum_{k\sigma} \epsilon_k c_{k\sigma}^\dagger c_{k\sigma}^\dagger + \sum_{k<k'} V_{kk'} b_{k}^\dagger b_{k'} + 2\sum_{k\sigma} \epsilon_k b_{k}^\dagger b_{k} + \sum_{k<k'} V_{kk'} b_{k}^\dagger b_{k}^\dagger, \quad (1.35)
\end{equation}

where \( c_{k\sigma}^\dagger, c_{k\sigma} \) are the creation and respectively destruction operators for single particle fermionic states of momentum \( k \) and spin \( \sigma = (\uparrow, \downarrow) \). Accordingly, the pairs are created by \( b_{k}^\dagger = c_{k\uparrow}^\dagger c_{-k\downarrow} \) and annihilated by \( b_{k} = c_{-k\downarrow} c_{k\uparrow}^\dagger \) which, unlike usual bosons, won’t allow two pairs be created (or annihilated) at the same \( k \) magnitude. Schematically, the BCS strategy was to apply a variational technique based on the trial wave function:
\[ |\psi_0\rangle = \prod_k \frac{1 + g_k b_k^\dagger}{\sqrt{1 + |g_k|^2}} |0\rangle \]  

(1.36)

which leads to an expectation value for the Hamiltonian:

\[ E = \langle \psi_0 | H | \psi_0 \rangle = 2 \sum_k \delta_k v_k^2 + \sum_{k,k'} V_{k,k'} u_k u_{k'} v_{k'} , \]  

(1.37)

where \( v_k^2 \) is the occupancy probability for a pair of electrons on the opposite sides of Fermi surface, \((k^\uparrow, -k^\downarrow)\):

\[ v_k = g_k / \sqrt{1 + g_k} , \quad u_k = 1 - v_k . \]  

(1.38)

By the variational principle, the best ground state of \( H \) is obtained using \( v_k^2 \) which minimizes \( E \), subject to \( \partial E / \partial (v_k^2) = 0 . \) Henceforth, considering a gap-like solution

\[ \Delta_k = \sum_{k'} V_{k',k} v_{k'} u_{k'} , \]  

(1.39)

minimizing equation (1.37) leads to:

\[ \delta_k = \frac{1}{2} \frac{\Delta_k (u_k^2 - v_k^2)}{v_k u_k} . \]  

(1.40)

Consequently, denoting \( E_k = \sqrt{\delta_k^2 + \Delta_k^2} \) the energy required to create a single quasiparticle of moment \( k \), we obtain the explicit identities of \( v_k^2 \) and \( u_k^2 \):

\[ v_k^2 = \frac{1}{2} \left( 1 - \delta_k / E_k \right) , \quad u_k^2 = \frac{1}{2} \left( 1 + \delta_k / E_k \right) , \]  

(1.41)

that is the ground state wave function of the Cooper pairs at zero temperature:

\[ |\psi_0\rangle = \prod_k \left( u_k + v_k b_k^\dagger \right) |0\rangle , \]  

(1.42)

and the gap equation becomes:

\[ \Delta_k = - \sum_{k'} V_{k,k'} \frac{\Delta_{k'}}{E_{k'}} . \]  

(1.43)

The presence of \( \Delta_k \) – a direct consequence of the pairing interaction – represents the onset of an energy gap in the paired quasiparticle excitation spectrum. Since the number of quasiparticles must be in a one-to-one correspondence with the number of Bloch states when there is no pairing, the quasiparticle density of states \( N(E) \) (Fig.1.3a) depends on the normal density of states \( N(0) \):

\[ 16 \]
\[ N(E) \approx N(0) \left( \frac{\partial E_k / \partial \delta_k}{\Delta} \right)^{-1} \approx N(0) E / \sqrt{E^2 - \Delta^2}, \]

provided \(|E| \geq \Delta\) and \(\delta\) is small (which assumes a \(\Delta\) approximately \(k\)-independent).

Remarkably, as shown by Gor’kov \cite{28}, the GL and BCS theories are equivalent and the order parameter (1.16) corresponds to the center-of-mass wave function of the \(N/2\) Cooper pairs and manifests an order of symmetry given in BCS by the form of the separable interaction potential \(V_{kk'} = \gamma \varphi_k \varphi_{k'}\), where \(\gamma\) is coupling strength between the single particle order parameter functions \(\varphi_k\). The singlet pairing present in the BCS Hamiltonian (1.37) is consistent both with s-wave and d-wave symmetries so that, up to this point, the theory has a degree of generality which is only lowered by the assumptions necessary to make predictions regarding normal metal superconductors.

Figure 1.3  a) BCS density of states for \(0 < T < T_c\). b) BCS type gap evolution with temperature. The curve was obtained in our lab on a double-gap MgB2 sample \cite{29}.

Specifically, returning to Cooper’s assumption (1.30), which is tantamount to assigning an s-wave pairing symmetry to the order parameter, and using (1.39), (1.41) and (1.44), one can calculate the BCS gap parameter and the expectation energy for \(|\delta_k| < \hbar \omega_p\), in the weak coupling limit \(N(0)V \ll 1\):

\[ E \approx N(0) \Delta^2 / 2, \]

\[ \Delta = \hbar \omega_p / \sinh \left[ \frac{1}{(N(0)V)} \right] \approx 2\hbar \omega_p e^{-1/(N(0)V)}. \]
Now, using this new energy scale much smaller than $V$, the classical superconductive parameters can be expressed in terms of a quantity related to the microscopic mechanism represented by the temperature dependent gap $\Delta = \Delta(T)$ represented in Fig.1.3b. Thus, going back to (1.1), the BCS theory predicts a critical magnetic field given by:

$$H_c = 2\sqrt{\pi N(0)} \Delta(0). \quad (1.47)$$

The theory can be easily extended to finite temperatures and it predicts an isotope effect (1.11) with $\alpha = 1/2$. The critical temperature can be explicitly related to the gap:

$$2\Delta(0) = 3.52 k_B T_c. \quad (1.48)$$

Subsequently, the coherence length:

$$\xi = \hbar v_F / 2\Delta. \quad (1.49)$$

Note that $\xi$ decreases with increasing $\Delta$ and $T_c$. The fact that $\xi$ is the characteristic size of the Cooper pairs transpires from Dyson’s real form for the BCS wave function:

$$|\Psi_0\rangle \propto \prod_i \Phi(r_{i+1} - r_i), \Phi = \sum_k g_k e^{ikr}. \quad (1.50)$$

From a physical point of view, a large $\xi$ means a weak pairing strength and brings about an easier correlation between the pair wave functions which in the BCS materials overlap.

In spite of the result (1.48), a finite value gap is not inherent to the superconductive state. Even if the amplitude of the GL order parameter is proportional to $\Delta$ [28], so that the gap can be incidentally regarded as an avatar of $\varphi$, the two concepts are different. Being proportional to the superfluid density of states, the GL order parameter is in fact a representation of the long range phase coherence between Cooper pairs (in the BCS case based on the overlap of wave functions), one of the fundamental ingredients of the superconducting state besides pairing itself, while the gap measures the strength of the pair bond. Thus, a vanishing order parameter signals out the disappearance of superconductivity, whereas a zero gap may be just a signature of unsteady condensate. Moreover, in some Type II superconductors, the pairs may preform before the superfluid condensation, the excitation spectrum presenting a so called pseudogap at temperatures exceeding $T_c$. However, in most of the applicability area of the classical BCS theory, the notions of order parameter and excitation gap can be legally interchanged.

A fundamental property of the BCS superconductive ground state is the fact that it is not disturbed by nonmagnetic disorder. This phenomenon is conceptualized by Anderson’s theorem
which notes that, being based on time-reversed pairs of single particles, the order parameter (and the transition temperature) is a solution of a mean field theory avoiding fluctuating conditions. *Inter alia*, the magnetic impurities, by breaking the time-reversal, can affect superconductivity and lead to the afore mentioned gapless superconductivity, since the pair coherence can be partially preserved such that the order parameter remains nonzero.

### 1.1.2 Post-BCS concepts of superconductive transport

After the BCS theory was published, its explanatory and predictive power continued to be adapted and stretched, on one hand in order to exhaustively explain the new experimental data related to the superconductive state, and, on the other hand, to take advantage from the newly developed theoretical tools. As example, as noticeable from the discussion above, the BCS works fine with systems of weakly bounded quasiparticles applicable to the conventional superconductors. However, it fails to explain the highly correlated systems like heavy-fermion compounds and (especially underdoped) cuprates. In such Type II superconductors, the order parameter ceases to be conformal with the energy gap which is already finite at the critical temperature. Also, the Cooper assumption about the pairing potential, although a good approximation for the conventional superconductors, evidently cannot explain the more complicated situations when the interaction leading to superconductivity is not constantly attractive in the vicinity of Fermi surface. Thus, when the effective potential is repulsive at short distances and attractive at larger distances, a *d-wave* channel is more favorable, that is the pairing potential and so the order parameter are anisotropic (as is the case of cuprates). This pairing symmetry is associated to a strong interaction potential or a much smaller pair size $\xi$ and, consequently, to a more rarefied pair distribution such that the wave functions cannot overlap and the inter-pair phase coherence is more difficult to explain.

In order to generalize the BCS theory, it proved especially useful to reformulate it using the quasiclassical Green’s function formalism in Nambu space [31] to tackle the problem as a many-body mesoscopic system. Shortly after the advent of BCS theory, A.B. Migdal [32] used such a technique to solve Fröhlich’s Hamiltonian for arbitrary strong electron-phonon couplings in metals but his results were inconclusive as to the origin of superconductivity. The clue was given by L.P. Gor’kov [28] who adapted the BCS microscopic scenario by considering the
spatial dependence of $\Delta_k$ and proved from a quantum-field perspective that the BCS theory is equivalent with GL at temperatures close to $T_c$. He developed a time-dependent version of the GL equations with the decisive collaboration of G.M. Eliashberg [33] who noticed that the superconductive state can be described using Migdal’s formalism with Nambu’s propagators and subsequently developed an extended microscopic theory of phonon intermediated superconductivity. Thus, Eliashberg’s equations provide a powerful tool to analyze metallic isotropic superconductive systems with strong coupling by detailing the frequency dependence of the pair potential contained in the directional spectral density $\alpha^2_k(\omega)F_k(\omega)$ which is basically the rate of scattering of an electron from state $k$ to $k'$, involving all possible phonon frequencies $\omega$. When the phonon frequencies are very large with respect to $k_BT_c$, the order parameter is independent of frequency, the coupling is said to be weak and Eliashberg’s construct reduces to the BCS theory.

Gor’kov’s pioneering strategy [34, 35] involves a functional approach where the pair potential arises as an “anomalous” off-diagonal pair energy in the equations for Green’s functions, and allows the necessary assumptions for strongly correlated superconductors. Specifically, albeit schematically, let’s consider a system of two species of fermions, say electron spin up and down participating in the Cooper pairs, with different Fermi energies, the difference being proportional to a perturbative influence like an applied magnetic field $H_a$: $\delta \mu \propto H_a$. (1.51) (This assumption will be necessary later, when we’ll introduce the LOFF state.) This split chemical potential adds an exchange field term to the Hamiltonian. In the lingo of Pauli matrices $\hat{\sigma}$, and Fermi fields expanded in a plane wave basis set through single fermion destruction operators $c_{k\sigma}$: $\varphi \equiv \varphi_{\sigma}(r,t) = \sum_k c_{k\sigma} e^{ikr}$, (1.52) the exchange term can be written as: $H_{exch} = \delta \mu \varphi^\dagger \hat{\sigma}_z \varphi$. (1.53) (Here the spin index $\sigma = (\uparrow, \downarrow)$ mustn’t be confused with the Pauli matrices.) Also, there will be four types of fermions (spin up and down electrons and holes, for a given chemical potential)
with four momenta \( k_i \), and the condensed ones are low lying with respect to the corresponding Fermi level. The zero spin condensate wave function depends on \( r \) in the case of inhomogeneous materials and can be expressed in coordinate space as

\[
\Gamma(r, r') = \langle \varphi(r, t) \hat{C} \varphi(r', t) \rangle,
\]

(1.54)

where \( \hat{C} = i \hat{\sigma}_z \), so that in the momentum space it becomes:

\[
\tilde{\Gamma}(k, k') = \langle \varphi(k) \hat{C} \varphi(k') \rangle.
\]

(1.55)

In the spirit of a mean field approach, Landau’s idea of Fermi liquid as a gas of nearly free electrons can be treated as an effective field theory [36]. In this case, the interaction term contained in the BCS effective action can be neglected so that the only term remaining is the one describing the fermion condensate:

\[
A_{\text{BCS}} \approx \int dt \prod_{i=1}^4 dk_i \left[ \tilde{\Gamma}(k_1, k_4) \varphi^\dagger(k_1) \hat{C} \varphi(k_4) - \tilde{\Gamma}^*(k_1, k_2) \varphi(k_1) \hat{C} \varphi^\dagger(k_2) \right] \delta(k_1 + k_2 - k_3 - k_4).
\]

(1.56)

On the other hand, using (1.53), the free action is

\[
A_0 = \int dt d\mathbf{k} \varphi^\dagger(\mathbf{k}) \left[ i \partial_t - \delta_k + \hat{\sigma}_3 \delta\mu \right] \varphi(\mathbf{k}),
\]

(1.57)

with \( \delta_k = \varepsilon_k - \mu \approx v_F (k - k_F) \) for fermions close from Fermi surface. Therefore, defining the gap function

\[
\Delta_{k,-k'} = V \int dp \tilde{\Gamma}(p, k + k' - p),
\]

(1.58)

and using the Nambu-Gor’kov basis:

\[
\chi(k) = \frac{1}{\sqrt{2}} \left( \begin{array}{c} \varphi(k) \\ \hat{C} \varphi^\dagger(k) \end{array} \right),
\]

(1.59)

the total action becomes

\[
A = A_0 + A_{\text{BCS}} = \int dt d\mathbf{k} d\mathbf{k}' \chi^\dagger(\mathbf{k}) \hat{G}^{-1}(\mathbf{k}, \mathbf{k}') \chi(\mathbf{k}'),
\]

(1.60)

where \( \hat{G}^{-1} \) is the Fourier transform of the inverse Gor’kov propagator:

\[
\hat{G}^{-1}(\mathbf{k}, \mathbf{k}') = \begin{pmatrix}
G_{0,\mathbf{k}}^{-1} - \Delta_{\mathbf{k},\mathbf{k}'} \\
-\Delta_{\mathbf{k},\mathbf{k}'} & G_{0,\mathbf{k}}^{-1}
\end{pmatrix},
\]

(1.61)
with \( G^{-1}_{0z} = \pm E - \delta_k \pm \sigma_3 \delta \mu + i0^+ \) being the free Green’s function in Nambu space. The Gor’kov propagator itself is then
\[
\hat{G} = \begin{pmatrix} G & -F \\ -F^T & G^T \end{pmatrix}, \tag{1.62}
\]
where the off-diagonal Green’s function \( F \) is the pair amplitude characteristic to the superconductive state. The Gor’kov equations of motion expressed in these terms is:
\[
\hat{G}^{-1} \hat{G} = 1, \tag{1.63}
\]
or, explicitly in configuration space,
\[
\begin{align*}
\left( E - E(-i\partial_r) + \sigma_3 \delta \mu \right) G(r, r', E) + \Delta(r) F(r, r', E) &= \delta(r - r'), \\
\left( -E - E(-i\partial_r) - \sigma_3 \delta \mu \right) G(r, r', E) - \Delta(r) F(r, r', E) &= 0,
\end{align*} \tag{1.64}
\]
Here the spatial derivative may include the contribution of an applied magnetic field: \( \partial_r = \nabla_r - ieA(r) \). The \textit{pair potential} or the gap equation at \( T = 0 \) is given by the off-diagonal terms:
\[
\Delta^*(r) = -iV \int dE \text{Tr} \left( F(r, r, E) \right), \tag{1.65}
\]
and it represents the part of the fermion-phonon self energy responsible for superconductivity. In a normal material \( V = 0 \), so that \( \Delta = 0 \), but the superconductivity can still exist in a proximity region due to the nonzero pair amplitude \( F \neq 0 \). At finite temperatures the form of the pair potential can be obtained by summation over Matsubara frequencies [37] \( \omega_n = (2n + 1)\pi T \):
\[
\Delta^*(r) = VT \sum_{n=-\infty}^{\infty} \left. \text{Tr} \left( F(r, r, E) \right) \right|_{E = i\omega_n}. \tag{1.66}
\]
In the case of homogeneous superconductors the pair amplitude \( F(r, r, E) \) is independent of the coordinate \( r \), so that one can infer from (1.66) a gap equation
\[
\Delta = V \Delta \int d\mathbf{k} \frac{1 - n^+(\mathbf{k}) - n^-(\mathbf{k})}{E(\mathbf{k}, \Delta)} \tag{1.67}
\]
where \( n^\pm(\mathbf{k}) = \left( \exp(\varepsilon_\mathbf{k} \pm \delta \mu) / k_B T + 1 \right)^{-1} \) are the equilibrium distributions for the two types of fermions and determine a cutoff temperature \( T_c \) above which \( \Delta \) vanishes.

In principle, Gor’kov equations can describe any mesoscopic situations but some subsequent quasiclassical reformulations proved especially useful in treating proximity systems of interest in
This thesis. The quasiclassical approximation basically assumes that particles in the superconductor move along definite trajectories. It is valid for pair potentials slowly varying in space which, knowing that the spatial variation is given by $\xi \sim 1/\Delta$, ultimately reduces to the condition $\Delta/\varepsilon_F \ll 1$. These quasiclassical theories dealt with problems like the inhomogeneous superconductors – first addressed by Eilenberger [38] and by Larkin and Ovchinnikov [39] –, the nonequilibrium superconductors – described by Keldysh Green’s function technique –, or the so called dirty limit where the impurity scattering in the superconductor dominates and a suitable model is provided by Usadel’s equations [40].

In most of the cases where superconductivity is independent of the single fermion wave function – inclusive the tunnel hybrid structures and proximity region experimentally studied in this thesis –, a lucrative quasiclassical approximation consists in integrating the Green’s functions over the relative coordinate $r_1 - r_2$, which results in a more workable function for problems involving exclusively paired-electron wave functions which depend on the center of mass coordinate $r$. This includes Andreev reflection [41] (a phenomenon, crucial in our experiments, which is to be detailed later) since it is based on the difference between electron-hole wave functions. Then, the quasiclassical form of Green’s function is

$$
\hat{g}(r, \varepsilon_F, E) = \frac{i}{\pi} \int d\delta_k \hat{G}(\delta_k, r, \varepsilon_F, E),
$$

(1.68)

and Gor’kov theory modifies into Eilenberger equation of motion

$$
-\left[\varepsilon_F \partial_r - iE\hat{\sigma}_3 + \hat{\Delta}(r, \varepsilon_F) + \langle \hat{g} \rangle_{\varepsilon_F}/2t, \hat{g}(r, \varepsilon_F, E)\right] = 0,
$$

(1.69)

where $\hat{\Delta}(r, \varepsilon_F)$ is the superconducting pair potential matrix and $\langle \hat{g} \rangle_{\varepsilon_F}/2t$ is the impurity scattering over time $t$, averaged over Fermi surface. The Green function $\hat{g}$ can be expressed likewise as a linear combination of Pauli matrices, essentially represented by

$$
\hat{g} = \begin{pmatrix}
g & f \\
-f & -g
\end{pmatrix},
$$

(1.70)

subject to the normalization condition $\hat{g}^2 = 1$.

The effective Schrödinger equation corresponding to Gor’kov expressions (in the clean limit) takes the form of Bogoliubov-de Gennes (BdG) equations [42, 43] which describes the quasiparticle states in superconductors with spatially varying pairing potential, states represented by the two-component wave function.
\( \varphi(r) = \begin{pmatrix} u(r) \\ v(r) \end{pmatrix} \).

Thus, using again the Fermi fields (1.52), the effective Hamiltonian

\[
H = \int dr \left[ \sum_\sigma \varphi^*_\sigma(r) H_0(r) \varphi_\sigma(r) + \Delta(r) \varphi^*_{\uparrow}(r) \varphi^*_{\downarrow}(r) + \Delta^*(r) \varphi_{\downarrow}(r) \varphi_{\uparrow}(r) \right],
\]

where \( H_0(r) = -(\hbar^2/2m) \partial^2_r + \sigma_\sigma \delta \mu \) and \( \Delta(r) \) is the pair potential given by the fermionic spin condensate (1.54) at position \( r \)

\[
\Delta(r) = -V \left\langle \varphi_{\downarrow}(r) \varphi_{\uparrow}(r) \right\rangle
\]

The Hamiltonian can be diagonalized using the so called Bogoliubov-Valatin transformation \([44]\) from fermion operators to \( n \) excited quasiparticle operators \( \gamma_{n\sigma} \),

\[
\begin{align*}
\varphi_{\uparrow}(r) &= \sum_n \left[ \gamma_{n\uparrow} u_n(r) - \gamma^*_{n\downarrow} v_n(r) \right], \\
\varphi_{\downarrow}(r) &= \sum_n \left[ \gamma_{n\downarrow} u_n(r) + \gamma^*_{n\uparrow} v_n(r) \right],
\end{align*}
\]

the diagonalized form being

\[
H = H_{\text{ground}} + \sum_{n,\sigma} E_{n\sigma} \gamma^*_{n\sigma} \gamma_{n\sigma},
\]

from where one can build up the BdG equations:

\[
\begin{pmatrix}
H_0 & \Delta(r) \\
\Delta^*(r) & -H_0^*
\end{pmatrix}
\begin{pmatrix}
u(r) \\ v(r)\end{pmatrix}
= E
\begin{pmatrix}
u(r) \\ v(r)\end{pmatrix}
\]

Consequently, the self consistency equation for the pair potential is

\[
\Delta(r) = V \sum_n v^*_n(r) u_n(r) (1 - 2 f_n),
\]

where \( f_n = \left( 1 + e^{E_n/k_B T} \right)^{-1} \) is the Fermi distribution.

### 1.1.3 High temperature superconductivity. Cuprates.

Following the triumphant BCS theory, the ample evolutions in the quantum theory of metals and Fermi liquid thermodynamics made possible an almost complete description of the conventional superconductivity. However, with the discovery of heavy-fermion compounds by Steglich \([45]\), in 1979, and the cuprate class of high temperature superconductors by Bednorz.
and Müller [46], in 1986, the harmonious picture faded due to the clearly non-BCS character of these materials. Unlike their conventional counterparts, the superconductivity in these compounds proved to originate from strong electron correlations and intrinsic magnetism seems to play a more constructive role in the mechanism, rather than the destructive effect predicted by BCS.

In heavy fermion superconductors, the effective mass of the quasiparticles is greatly enhanced by the ferromagnetic interaction between electrons and the local momenta. Therefore, the ground state of these materials shows a magnetic ordering at very low temperatures so that it was suggested that the pairing may not be based on phonon exchange but on magnetic interactions between electron spins. Recently [47], the coexistence of magnetism and superconductivity manifested as a LOFF state was possibly identified in the heavy fermion superconductor CeCoIn$_5$ by interpreting the cascade of step in magnetization measurements as transitions into higher order Landau sub-phases favored by the LOFF interaction of orbital and paramagnetic effects.

The research on high temperature cuprates superconductors stirred a true revolution in the physics of superconductive state. Some of them are listed in Table 1.1, with the corresponding high critical temperatures at optimal doping.

<table>
<thead>
<tr>
<th>Cuprate</th>
<th>CuO$_2$ planes</th>
<th>Optimal doping $T_c$ [K]</th>
</tr>
</thead>
<tbody>
<tr>
<td>Nd$<em>{2.5}$Ce$</em>{0.5}$CuO$_4$</td>
<td>1</td>
<td>24</td>
</tr>
<tr>
<td>La$_{2.5}$Sr$_5$CuO$_4$</td>
<td>1</td>
<td>38</td>
</tr>
<tr>
<td>YBa$_2$Cu$<em>3$O$</em>{6+delta}$</td>
<td>2</td>
<td>93</td>
</tr>
<tr>
<td>Bi$_2$Sr$_2$CaCu$<em>2$O$</em>{8+delta}$</td>
<td>2</td>
<td>95</td>
</tr>
<tr>
<td>Tl$_2$Ba$_2$CaCu$_2$O$_8$</td>
<td>2</td>
<td>105</td>
</tr>
<tr>
<td>HgBa$_2$Ca$_2$Cu$_2$O$_8$</td>
<td>3</td>
<td>135</td>
</tr>
</tbody>
</table>

Unlike the conventional superconductors which are metals in their normal state, the cuprates are Mott insulators doped mostly by holes, rarely by electrons, characterized by the presence of at least one CuO$_2$ plane in a layered crystal structure leading to anisotropic conductance, almost two-dimensional for higher doping. The insulating property of the undoped parental perovskite...
originates from a small gap of about 2 eV between the 3d copper and 2p oxygen levels and Coulomb correlations among the copper states. These are responsible for the superexchange interaction and antiferromagnetic ordering of the undoped state and spin fluctuations in the superconducting state that may play a crucial role in the non-BCS pairing mechanism [48].

Figure 1.4  Generic hole-doping cuprate phase diagram and the commonly expected ab-plane resistivity dependence on temperature \( \rho_{ab} - T \) for different regions: semiconductive in the undoped region, then superconductive and metallic. The borders between the superconductive, pseudogap and ferromagnetic insulator regions are characterized by doping dependent critical temperatures \( T_c \), \( T^* \) and \( T_N \). The inset graph represents the phase diagram for Bi\(_2\)Sr\(_2\)CaCu\(_2\)O\(_{8+\delta}\) obtained experimentally using the characteristic \( \rho_{ab} - T \) dependency [49].

As noticeable in the generic phase diagram in Fig.1.4, cuprates satisfy one of the characteristics of strongly correlated systems: the specific behavior is the result of a competition between a multitude of very different phases. The antiferromagnetic insulator area covers a limited region at temperatures lower than Néel temperature \( T_N \) corresponding to the respective hole doping \( \delta < 0.05 \). As the hole doping is increased, the magnetic ordering is destroyed and the material transcends into a state similar to the spin liquid ground state predicted by Anderson [50] via his RVB scenario, followed at low temperature by a regime of randomly interacting
spins, a mixture of insulating and superconducting spots with an ambiguous ground state called spin glass. For temperatures $T < T_c$, where $T_c$ is a function of the respective doping $0.05 < \delta < 0.16$, the material enters in the underdoped superconductive state which is preceded, for $T^* > T > T_c$, by a zone dubbed pseudogap or spin gap region due to the onset at $T^*$ of a gaplike feature in the electronic low excitation spectrum. The pseudogap reshapes into the superconductive gap at $T_c$, even if the nature of this transition is still controversial [51]. As the doping enters the approximate range $0.16 < \delta < 0.27$, the superconductive state is overdoped and the pseudogap disappears. For temperatures $T > T^*$, the material reaches a “strange” metal non-Fermi liquid phase characterized by power-law transport properties. In the underdoped range, the resistivity vs. temperature curve bends down at $T^*$ and the decrease is steeper before the superconductive fall at $T_c$, a tendency which is not observable in the overdoped region (Fig.1.4).

Table 1.2  In plane coherence length, penetration depth and estimative upper critical magnetic field for some cuprates at optimal doping. The values reported in the literature vary around these numbers. Note that the very short coherence is itself an indication of the strong coupling and the large penetration depth points at the low superfluid density depending on the doping level [52]. Therefore, superconductivity in cuprates emerges from strongly interacting low density carriers condensed in small size pairs.

<table>
<thead>
<tr>
<th>Cuprate</th>
<th>$\xi_{ab}$ [Å]</th>
<th>$\lambda_{ab}$ [Å]</th>
<th>$B_{c2}^{ab}$ [T]</th>
</tr>
</thead>
<tbody>
<tr>
<td>YBa$_2$Cu$_3$O$_6$</td>
<td>16±5</td>
<td>1500</td>
<td>150</td>
</tr>
<tr>
<td>Bi$_2$Sr$_2$CaCu$_2$O$_8$</td>
<td>16±5</td>
<td>1800</td>
<td>120</td>
</tr>
<tr>
<td>HgBa$_2$Ca$_2$Cu$_3$O$_8$</td>
<td>22±5</td>
<td>1500</td>
<td>190</td>
</tr>
</tbody>
</table>

Consequently, there are important differences between the behavior of underdoped and overdoped samples. There are several theories regarding the nature of the pseudogap. As example, one of the most popular one is based on the idea that the Cooper pairs are preformed at $T^*$ but, due to large phase fluctuation of the order parameter, the pair coherence sets up only at the lower $T_c$. Another theory builds up on the particular way the doped holes arrange inhomogeneously in the CuO$_2$ planes along Cu-O bonds forming unidirectional charge density waves or stripes [53] of metallic hole-rich regions (half filled in the underdoped region) separated by magnetic stripes reminiscent from the antiferromagnetic insulator phase characterized by antiferromagnetic correlations strong enough to have a coherence length
spreading over two lattice constants, $\xi_{AF} \geq 2a$. In fact, the phenomena of occurrence, dynamics and evolution of charge stripes stimulated by the strong coupling is a signature of the doped antiferromagnetic insulator microscopic order. With increasing doping, the interstitial magnetic stripes fragment, up to the point where the charge distribution becomes almost 2D uniform and the material passes into a metallic phase.

As seen in Table 1.2, the cuprates are Type II superconductors, their coherence length being much shorter than the penetration. Therefore, the layered very anisotropic structure is a remarkable detail when it comes to explaining their properties, inasmuch as the lattice is unstable. This anisotropy also extends to the magnetic properties like the large discrepancy between the $ab$-plane and $c$-direction upper critical magnetic fields and penetration depths. For a concrete image of this situation, Fig.1.5 depicts a unit cell of the BSCCO tetragonal crystal structure. It has two CuO$_2$ layers flanked by the insulating SrO and semiconductor BiO planes. Other cuprates have similar external layers on the sides of 1-4 CuO$_2$ planes, layers considered as charge reservoir for the superconducting processes in the CuO$_2$ stack. For BSCCO the lattice constants are $c \approx 30.9$ Å and $a \approx b \approx 5.4$ Å, with a Fermi surface almost cylindrical, so that the in-plane conductivity is almost isotropic and in the order parameter $\Delta(k_f,r)$ the Fermi momentum $k_f$ can be considered two dimensional. However, this is not the case in general: other cuprates, like YBa$_2$Cu$_3$O$_{6+\delta}$ (YBCO), have a slight in-plane anisotropy and it is also possible for the cuprate to experience structural phase transitions between states with radically different superconductive characteristics due to the geometrical distinctiveness of CuO$_2$ plane. For example, the degree of orthorhombic distortion, the plane buckling or the distance between CuO$_2$ planes and the charge reservoirs play a decisive role in determining the optimal doping. This is one of the reasons why tuning external pressure can actually mimic for a given cuprate the effect of doping on the $T_c$ [54] and actually increase the $T_c$ as in HgBa$_2$Ca$_2$Cu$_3$O$_8$ which shows an impressive $T_c = 164$ K under a pressure of $\approx 30$ GPa. Particularly, with increasing doping, the structure changes from insulating tetragonal to metallic orthogonal.

As a signature of cuprate anisotropy, the conductivity in $c$ direction (diffusive rather than metallic) is much lower than the $ab$-plane conductivity and it diminishes with the increasing doping. The $ab$-plane component of the effective mass tensor, $m^*_a \approx 5m_{\text{electron}}$, is actually 30 (in YBCO) to 50000 (in BSCCO) times smaller than the $c$-direction component such that the
conductance is almost two directional. Related to this, the critical current is also anisotropic. Hence the main excitations are planar, so that most of the superconductive transport is indeed happening along the $ab$-plane. Therefore, being very sensitive to the formation of 1D conductance channel stripes, the transport may be an important source of information about the superconductive mechanism.

In many respects, the superconductivity in cuprates is similar to the conventional one. It is still carried by pairs, a gap opens in the energy spectrum, the heat capacity has a peak at $T_c$ and

![Figure 1.5 BSCCO crystal structure generated by the author in VRML v2.0. It is generally accepted that the superconductivity is confined in the CuO$_2$ planes even if, by proximity, it can spread to the neighboring semiconducting BiO planes [55]. The shadowed projection represents a qualitative distribution of the order parameter with a d-wave symmetry assuming this possible leak of superconductivity. On the left are represented the two subcomponents of the anisotropic pair potential symmetries emphasized in this thesis – singlet $d_{x^2-y^2}$ and s-wave – based on the quasi-cylindrical Fermi surface attached to the tetragonal crystal. On the right, the CuO$_2$ plane is represented with the antiferromagnetic arrangement of spins in undoped state.](image)
the isotope effect, albeit doping and impurities dependent, is still present [56] pointing out to a possible phonon, although unconventional, contribution to the pairing mechanism.

Table 1.3  Schematic comparison between several superconductive characteristics in conventional and unconventional superconductors [57]. Here $Q$ is the commensurate wave vector, $\omega_F$ and $\omega_f$ are the characteristic frequencies of spin fluctuations for Fermi liquid and lower doped cuprate respectively, and $\xi_{AF}$ is the antiferromagnetic correlation length.

<table>
<thead>
<tr>
<th>Characteristics</th>
<th>Metallic</th>
<th>Cuprate</th>
</tr>
</thead>
<tbody>
<tr>
<td>Resistivity</td>
<td>$\sim T^a$</td>
<td>$\sim T$</td>
</tr>
<tr>
<td>Quasiparticle life time</td>
<td>$aT^2 + b\omega^2$</td>
<td>$aT + b\omega$</td>
</tr>
<tr>
<td>Spin excitation spectrum</td>
<td>flat</td>
<td>peaks at $Q \sim (\pi/a, \pi/a)$</td>
</tr>
<tr>
<td>Spin excitation energy</td>
<td>$\sim \omega_F$</td>
<td>$\omega_f \ll \omega_F$</td>
</tr>
<tr>
<td>Magnetic correlations</td>
<td>none</td>
<td>strong, $\xi_{AF} \geq 2a$</td>
</tr>
<tr>
<td>Uniform susceptibility</td>
<td>flat</td>
<td>$\sim T^<em>$ maximum at $T^</em>$ in underdoped regime</td>
</tr>
<tr>
<td>Pairing symmetry</td>
<td>$s$-wave</td>
<td>$d$-wave or mixed $d+i$s at the surface</td>
</tr>
</tbody>
</table>

On the other hand, there are fundamental differences (see Table 1.3). These make necessary a substantially generalized approach which should in particular take into account the rather sensitive nature of high-temperature superconductivity with respect to perturbations otherwise without effect on conventional superconductors. Thus, as we saw in Section 1.1.1, while the BCS superconductivity is inert to the impact of nonmagnetic impurities, the cuprate superconductivity is decisively locally affected by such perturbations, very similar to the suppressing influence of magnetic impurities, even though the long range properties, like the doping effect and the pseudogap are not affected [58]. However, a magnetic impurity in CuO$_2$ planes has almost no effect locally, fact which backs the hypothesis of a magnetic mediated pairing in cuprates.

Another important difference is the size of Cooper pairs: while in the conventional case the size is larger than the inter-fermion distances, in the case of cuprates the size is much smaller (see Table 1.2 for the characteristic coherence lengths). A direct consequence is that in conventional superconductors the long range coherence is realized by the overlapping wave functions, or Josephson coupling, the condensate being immediately suitable for a mean-field treatment. In contrast, in cuprates the pairs don’t overlap, the phase stiffness is much weaker, and a Bose-Einstein distribution is more compatible with the situation. The pair condensation in this
case is possibly based on spin fluctuations in $c$-direction while the pairing itself may be still phonon intermediated, even if not in the same manner as in the conventional case. Above $T_c$, the sea of carriers in conventional superconductors transform quickly into a normal Fermi liquid, while it is apparent that in cuprates the pairs still persist up to a second energy scale as an incoherent distribution of composite bosons [59]. Moreover, whereas the energy scale of pairing and long range coherence is the same in metallic superconductors and converge to the same value in the overdoped region of cuprates, in the underdoped regime two energy scales lead to dissimilar results from gap measuring experimental techniques. Thus, procedures sensitive to single particle excitation energy (e.g. angle-resolved photoemission spectroscopy (ARPES) and tunneling spectroscopy) will measure a gap $\Delta_p$, an in-plane quantity associated with the pairing potential, while those sensitive to the coherence properties of the condensate (e.g., Andreev reflection) will measure the energy coherence range $\Delta_c$, characterizing the correlations along $c$-direction. The two energy scales are doping dependent and seemingly the only one surviving above the condensation energy in the underdoped regime is $\Delta_p$ under the form of the pseudogap.

The in-plane gap $\Delta_p$ shares the same symmetry with the order parameter and crystal lattice. In the cuprates, the most embraced opinion is that the pairing is anisotropic, less symmetric than the underlying crystal lattice, in principal with a singlet $d_{x^2-y^2}$ symmetry (see Fig.1.5) sufficiently supported theoretically [60-62] and experimentally [63, 64]. In conventional superconductors, the symmetry is nodeless singlet $s$-wave, i.e., constant $k$ and $\Delta_s(k,T)$ real and positive, which in turn entails an isotropic gap in the vicinity of Fermi surface and an antisymmetric spin contribution to the superconductive wave function which must be overall antisymmetric. In contrast, the $ab$-plane order parameter with singlet $d$-wave symmetry seems adequate for the cuprate Brillouin zone, which is approximately square for $c$ significantly larger than $a \approx b$. It is still real but it alternates signs 4 times around the essentially cylindrical Fermi surface. The sign changes in the four lobes of maximal order parameter amplitude (widest gap) separated by nodes where the gap vanishes and where quasiparticle excitation lead to unconventional low-energy spectra seen in tunneling, ARPES and thermal transport experiments. The gap is maximal near the $\langle 100 \rangle$ crystal surfaces and zero toward $\langle 110 \rangle$ surfaces. It can be written as
\[ \Delta_d(k, T) = \Delta_d(T) \left[ \cos(k_x a) - \cos(k_y a) \right], \] (1.77)

which satisfies the \( B_{1g} \) representation of the discrete point group \( D_{4d} \) symmetry required by the tetragonal crystal lattice: sign changing under 90° rotations and diagonal reflection. Also, the more complex pairing symmetry is consistent with the destructive effect of structural defects and impurities on high temperature superconductivity.

While the s-wave pairing is suppressed by the on-site Coulomb repulsion, the predominant d-wave pairing is enhanced by the antiferromagnetic ordering since the singlet pairing is favored along the Cu-O bonds and disfavored at \( \pi/4 \) with respect to this direction. Important for our discussion is the fact that, according to a series of tunneling experiments [65], the pure d-wave behavior may be confined in the bulk of the cuprate, whereas at the surface the symmetry tends to be mixed, with the predominant d-wave component being altered by a secondary s or rotated d subcomponent:

\[ \Delta = (1 - \alpha) \Delta_d(k/k_r) + i\alpha \Delta_x, \] (1.78)

where \( \alpha \) is the weight of the subdominant component. The mixing is consistent with the theory of symmetry, since the underlying crystal lattice may experience incommensurate orthorhombic structural distortions as functions of temperature and surface effects. In this case, the order parameter nodes disappear and the new group representation is \( B_{1g} + iA_{1g} \) which, unlike the pure s or d-wave case, breaks the time reversal symmetry, that is its excitation distribution depends on the direction of the current.

One may ask why a more complex singlet symmetry is favored in-plane. Theoretically, a possible answer stems from the simplest model allowing electron correlations, the Hubbard model [66, 67], which proved to be a good candidate for cuprate planes and can be shown to lead to \( d_{x^2-y^2} \) pairing in the strong antiferromagnetic coupling limit. The one-band Hubbard Hamiltonian is:

\[ H = -\sum_{i,j,\sigma} t_{ij} c_{i\sigma}^\dagger c_{j\sigma} + U \sum_j n_{i\uparrow} n_{i\downarrow}, \] (1.79)

where \( t_{ij} \) are the amplitudes for hopping processes between lattice sites, \( U \) is the on-site Coulomb repulsion and \( n_{i\sigma} \) are electronic number operators. The Mott insulating phase arises for the strong coupling \( U \gg t_{ij} \), and the Hubbard model takes the form of the so called \( t-J \) model.
In this case the site possibility of double-occupancy is eliminated due to the energy cost much larger than the kinetic energies and the density of states contains two Hubbard bands split by $U$. Therefore, the low energy charge transport is dominated by holes and, in the case of half-filled band (one particle per site), no hopping with energy less than $U$ is allowed and the system is an insulator well described by the Heisenberg antiferromagnetic model with long range Néel order. When the system is doped with holes over the half-filling, in spite of the frozen charge dynamics, the next-site electron spins can couple via antiferromagnetic spin fluctuations with exchange interaction strength given by $J = 4t^2/U$, the coupling term in the Hamiltonian taking the Heisenberg-Dirac form $J \mathbf{S}_i \cdot \mathbf{S}_j$, where $\mathbf{S}$ is a spin $1/2$ spin operator. Suggestively, in cuprates $J$ is very large, $J \approx 120-130$ meV. The pair binding is positive so that the model forms pairing bound states with dominant doping dependent $d$-wave singlet symmetry. The $t - J$ Hamiltonian per se is notoriously difficult to solve, but a number of methods were used more or less successfully to simplify the problem [67, 69, 70], most of them obtaining the $d$-wave superconducting instability induced by spin fluctuations close from half-filling, as was first predicted in the context of heavy-fermion superconductivity [71]. As example there are small periodic cluster numerical implementations [72, 73] (see Fig.1.6), quantum Monte Carlo methods [74] or mean-field adaptations like replacing the local no double-occupancy with a global constraint over large number of states per lattice site, so the local constraint is relaxed and the system can be treated in the weak coupling limit [75].

![Figure 1.6](image)  

**Figure 1.6** Pair correlation $D(r)$ with $d_{x^2-y^2}$ symmetry calculated using a $t - J$ ladder model. a) Dependence on doping density $\delta$, for $J/t = 0.35$ and 0.5. b) Dependence on distance for three fixed dopings and $J/t = 0.35$. 

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As a concrete instance of Hubbard model virtues, Fig. 1.6 shows the theoretical dependence on distance and doping of the $d_{x^2-y^2}^\pm$ pair correlation obtained by White and Scalapino [67, 72, 73] using a density-matrix renormalization group technique applied to the $t-J$ ladder model. If the pair field at $i$-th site is

$$\Delta_i^\uparrow = c_{i\uparrow}^\dagger c_{i\downarrow}^\dagger - c_{i\downarrow}^\dagger c_{i\uparrow}^\dagger,$$  (1.80)

then the pair correlation at distance $x$ can be written

$$D_x = \langle \Delta_{i,x}^\uparrow \Delta_i^\dagger \rangle.$$  (1.81)

Qualitatively, the behavior is the one expected: the correlation is maximum for an “optimal” doping and decreases with distance.

### 1.2 Ferromagnetism

Despite the relatively long history since its discovery, ferromagnetism and, in general, magnetic ordering in metals still defies an exhaustive and consistent description and are subject of intense research. As example, in recent years, the interest for materials showing inhomogeneous magnetic phase dynamics was greatly enhanced as a result of the discovery of the unusual magnetotransport properties of manganites [76]. These perovskites exhibit the so-called Colossal Magnetoresistance (CMR), that is their resistivity grows enormously under a small applied magnetic field. They provoked a great enthusiasm not only due to the possible applications but also due to the fascinating physics driving their behavior involving spin and orbital order closely related to the phase mechanisms in cuprates.

The theory of ferromagnetism is based on the concept of Weiss molecular field or exchange interaction [77] which is basically the mean field action on strongly coupled dipoles assumed to be proportional to magnetization. Under Curie temperature, microscopically it yields a collective alignment of spins along the same direction. The origin of the exchange field is in the overlap of wave functions in conjunction with Pauli exclusion principle: since the wave function for the interacting electrons must be antisymmetric and the spatial part changes symmetry with the relative alignment of spins (parallel or antiparallel), the spatial electron density is affected and this leads to a rigid shift in the majority and minority spin bands given by:

$$\hbar = \varepsilon_{k\downarrow} - \varepsilon_{k\uparrow}^\dagger,$$  (1.82)
Figure 1.7  a) The Bethe-Slater curve representing the magnitude of the direct exchange potential \( J \) function of the interatomic distance normalized by the radius of the 3d orbital. The interaction is antiferromagnetic for \( J < 0 \) and ferromagnetic for \( J > 0 \). b) Density of states in a ferromagnetic metal depicted schematically with the concrete example of iron. The 3d sub-bands are shifted one to respect to the other by the exchange interaction.

where

\[
h = -2J \sum_{i,j} S_i \cdot S_j \tag{1.83}
\]

is the pairwise exchange energy and \( J \) is a coupling constant dependent on the overlap of orbitals (\( J \) is positive for ferromagnetic coupling). The overlap of wave functions depends on the interatomic distance and hence the nature of the magnetic interaction is a function of the atomic size relative to the radius of the overlapping partially filled electronic shells (see Fig.1.7a). Thus, if the magnetic moments are close enough for the wave functions to overlap, there is a so called direct exchange which is the balance between Coulomb forces, kinetic energy of electron transfer and Hund’s rule for intra-atomic electrons. For moments further apart, the interaction sets via an indirect exchange intermediated by itinerant electrons, the nature of the magnetic ordering (ferromagnetic or antiferromagnetic) depending on distance due to the spatially damped oscillatory behavior of the coupling amplitude between positive and negative values. Most commonly, a 3d partially filled electronic shell in competition with the 4s one contributes the most to the electronic density of states at Fermi level resulted from overlapping (situation depicted in Fig.1.7b, with the inset specifically for iron). The overlap leads to an energy band
which splits in an external magnetic field $H$ into two spin bands, $\varepsilon_{k\downarrow}$ and $\varepsilon_{k\uparrow}$, according to (1.82). If we denote $N_{\downarrow}$ and $N_{\uparrow}$ the number of electrons in the respective bands, we have

$$N_{\uparrow\downarrow} = \int_{0}^{\varepsilon_{F}} G(\varepsilon) d\varepsilon,$$  

(1.84)

where $G(\varepsilon)$ is the density of states. Then, the spin polarization is defined by

$$P = \frac{N_{\uparrow} - N_{\downarrow}}{N}, \quad N = N_{\uparrow} + N_{\downarrow}. \quad (1.85)$$

These assumptions intimating the microscopic properties as a system of elementary magnets can be related to the phenomenological conjectures, like the identity of magnetic susceptibility and the specific form of the exchange energy dependency on magnetization, only by imposing mean field constraints meant to reduce the complexity of the problem. Thus, E.C. Stoner [78] proposed a summation manner in (1.83) taking into account only a time averaged action $\tilde{S}_{j}$ from all spins onto each spin $S_{j}$. In this case, the coupling term in the Hubbard Hamiltonian (1.79) in the strong Coulomb repulsion limit takes the form:

$$Jn_{\uparrow}n_{\downarrow} = J\left(\frac{1}{4}(n_{\uparrow} + n_{\downarrow})^{2} - \frac{1}{4}(n_{\uparrow} - n_{\downarrow})^{2}\right), \quad (1.86)$$

where only the second term depends on spin and gives the exchange energy:

$$h = -\frac{1}{4}J\left(N_{\uparrow} - N_{\downarrow}\right)^{2}. \quad (1.87)$$

Now, in these conditions and assuming a small band splitting so that $G(\varepsilon) \approx G(\varepsilon_{F})$, the total energy band can be written as:

$$E = \sum_{\sigma=\uparrow,\downarrow} \int_{0}^{\varepsilon_{F}} \varepsilon G(\varepsilon) d\varepsilon \approx \frac{N^{2}P^{2}}{4G(\varepsilon_{F})} - \mu_{0}\mu_{B}NPH. \quad (1.88)$$

Adding the exchange energy, the total energy becomes:

$$E = \frac{N^{2}P^{2}}{4} \left(\frac{1}{G(\varepsilon_{F})} - J\right) - \mu_{0}\mu_{B}NPH, \quad (1.89)$$

which yields the so called Stoner criterion of spontaneous magnetization:

$$JG(\varepsilon_{F}) \geq 1. \quad (1.90)$$
This criterion is satisfied by ferromagnetic metals like iron and rules that an external magnetic field is not necessarily needed to split the band. The local magnetic field is responsible for a spontaneous exchange field.

The physics of ferromagnetic state shows unexpected surprises when the structural geometry of the system has to be taken into account as is the case of thin films of ferromagnetic materials. These can be practically viewed as samples of two dimensional magnetization characterized by a dominance of short range order and uniaxial Ising-like behavior induced by anisotropy. As a result, in ultrathin films, the orientation of the spontaneous magnetization switches the easy-axis from perpendicular to in-plane direction at low temperatures [79].

1.3 LOFF State

In the middle of the sixties, it was shown that the apparent discrepant natures of superconductive state and magnetism can be reconciled in a special form of spatially inhomogeneous phase superconductivity – an “unexpected solution of the pairing equation” – which precedes the transition to normal state in the presence of a magnetic perturbation. This state was initially studied by two groups, on one hand P. Fulde and R.A. Ferrell [80] and, on the other hand, A.I. Larkin and Y.N. Ovchinnikov [39], so that it is commonly referred as LOFF state.

In the presence of an exchange field $h$, the unusual LOFF pairs contain electrons with unequal momenta for the two spin directions since the exchange interaction induces a difference of chemical potential $\delta \mu = h$ between the spin up and down electrons. Thus, as visible in Fig.1.8a, although the center-of-mass momentum $q$ of a normal Cooper pair $(k^+, -k^+_L)$ is zero, $h$ unpairs the electrons to $[(k + q/2)^+, (-k + q/2)_L^+]$, with $q$ having arbitrary direction but fixed magnitude $q = 2h/hv_F$. Consequently, the rotational and translation invariance is broken and the phase gap can be expressed in an inhomogeneous manner which was first considered by Fulde and Ferrell.

$$\Delta(r) = \Delta e^{iq \cdot r}. \quad (1.91)$$

Larkin and Ovchinnikov extended the discussion to multiple $q$ values. The shift in energy due to $\delta \mu$ is given by:
To further analyze the implications, we can go back to the gap equation (1.67) given in the context of Gor’kov formalism. Combining it with the BCS gap equation (1.43) in the weak coupling limit and denoting $\Delta_0$ the superconductive gap when $h = 0$, one can get a more explicit gap equation:

$$\Delta_0 = \delta\mu + \sqrt{\delta\mu^2 - \Delta^2}, \quad (1.93)$$

from where we see that, for $0 \leq \delta\mu \leq \Delta_0$, we either have $\Delta = \Delta_0$ (which gives a superconductive phase independent of $h$), or $\Delta^2 = (2\delta\mu - \Delta_0)\Delta_0$. To see which case is favored, we calculate the free energy difference at the normal-superconductor phase boundary using an expression justified in [34]:

$$F_s - F_n(\delta\mu) = -\frac{V}{2} \int_{\Delta(0)}^{\Delta_0(\delta\mu)} \frac{\Delta^2}{\Lambda_0} d\Delta_0, \quad (1.94)$$

and see that the minimum is obtained for the first solution:

$$F_s - F_n(0) = -\frac{V}{2} \left( \Delta_0^2 - 2\delta\mu^2 \right). \quad (1.95)$$

In conclusion, the superconductive condensate breaks only when the exchange potential $h$ leads to a split in chemical potential $\delta\mu = h \geq \Delta_0/\sqrt{2}$, this result being known as Clogson criterion for the onset of LOFF state.

Remarkably, the LOFF phase occupies only a rather narrow area on the phase diagram $(T, h)$. As shown in Fig.1.8b, it lays at low temperatures, between the $q = 0$ superconductive region (first order phase transition border) and the normal regime (second order phase transition border), within a certain $\delta\mu$ range

$$\left[ \delta\mu_1 = \Delta_0/\sqrt{2}, \delta\mu_2 \approx 0.754\Delta_0 \right]. \quad (1.96)$$

The second limit $\delta\mu_2$ can be obtained by writing the gap equation (1.67) with $\epsilon_{k\uparrow\downarrow} \pm \delta\mu$ given by (1.92), followed by minimization with respect to $q$. Therefore, returning to equation (1.49) which yields $\Delta_0 = h\nu_F/2\xi$, we notice immediately that this condition makes the possibility of LOFF state in Type I superconductors very difficult due to the excessive pair size in these materials.
a) In the LOFF state the paired electrons have different Fermi levels so that, unlike the BCS case, the center of mass momentum $q$ is nonzero. b) The LOFF phase occupies a narrow strip at low temperatures separated from the conventionally paired superconductive phase by a first order transition border and by a second order boundary from the normal region.

Therefore, the best chance to observe it is in ferromagnetic proximity regions with Type II superconductors, where the pairs survive along several coherence lengths, $\xi \sim 1/h$, independent of the energy gap $\Delta$. We shall address this problem experimentally in Chapter 4.

Due to the finite center-of-mass momentum, the LOFF order parameter is spatially modulated such that at periodic nodal points in space its phase changes by $\pi$ and the state can be characterized as gapless superconductivity. Let’s see how this happens in a simplified picture where there is a unique $q$ and the state can be composed from the pairs with the same spin (along the magnetic field) but different momentum direction. Then, the Fulde-Ferrell wave function (1.91) adds up to:

$$\varphi(\mathbf{r}) = \varphi_0 \frac{1}{2} \left( e^{i q \cdot r} + e^{-i q \cdot r} \right) = \varphi_0 \cos(q \cdot \mathbf{r}), \quad (1.97)$$

so that the overall pair distribution is given for all angles $\theta$ by

$$\int_0^1 d \cos \theta \cos \left( \frac{2h}{v_F \cos \theta} r \right) \approx \sin \left( \frac{r}{\xi} \right) \frac{r}{r/\xi}, \quad (1.98)$$

which provides the coherent modulation of the order parameter in a strong exchange potential.

Certainly, the problem can be treated more rigorously by using Eilenberger equation of motion (1.69) in a clean proximity region [81-83] or Usadel equation in the dirty limit [83, 84]. As we observed several times up now, if a superconductor makes contact (or is within tunneling limit) with a normal medium, some superconductive correlation can penetrate into the normal
surface forming a proximity region where the pair amplitude has a finite value decaying along several coherence lengths. The same thing happens when the normal region is replaced with a ferromagnet. In this case, the depth of the proximity region is given by the exchange interaction $h$ which defines a ferromagnetic coherence length

$$\xi_F = \frac{\sqrt{hD}}{h} \text{ dirty limit,}$$

$$\xi_F = \frac{hV_F}{h} \text{ clean limit,}$$

(1.99)

where $D$ is the diffusion coefficient for electrons in the ferromagnet [85].

For instance, following [82], one can consider the quasiclassical case of such a ferromagnetic proximity region of thickness $d$ in contact with a clean superconductor (Fig.1.9a). The diagonal (normal) Green’s functions (1.68) can be expressed by

$$g_{\uparrow\downarrow}(k, E) = \frac{1 - e^{i\phi_{\uparrow\downarrow}}}{1 + e^{i\phi_{\uparrow\downarrow}}} \frac{\alpha(k, E)\alpha(k', E)}{\Delta(k, E)\Delta^*(k', E)}$$

(1.100)

where $\alpha(k, E) = E - \left(E^2 - |\Delta(k)|^2\right)$ and $\phi_{\uparrow\downarrow}(k, E) = 2(E \mp h)L/v_F$ (here and below all momenta are taken at Fermi level). Also, $L = 2dk/k$ is the length of a classical trajectory inside the ferromagnet, $k_i$ being the momentum component parallel to the interface. The order parameter is given by the off-diagonal Green functions:

$$\hat{\Lambda}(r, k) = \begin{pmatrix} 0 & i\hat{\sigma}_z\eta(k)\Delta^*(r) \\ -i\hat{\sigma}_z\eta(k)\Delta(r) & 0 \end{pmatrix}$$

(1.101)

where the coefficient $\eta(k_f)$ can be adapted to the type of pairing symmetry in the bulk of the superconductive reservoir:

$$\eta(k) = \begin{cases} 1 & s\text{-wave}, \\ \frac{(k_x^2 - k_y^2)/k^2}{k_x k_y / k^2} & d\text{-wave nodes}, \\ k_x k_y / k^2 & d\text{-wave anti-nodes.} \end{cases}$$

(1.102)

Using these quantities, the density of states in the ferromagnetic layer of thickness $d$ is

$$G(d, E) = \frac{1}{2} G_0 \text{ Re} \left(g_{\uparrow} + g_{\downarrow}\right).$$

(1.103)

In Fig.1.9b this theoretical oscillatory $d$-dependence of the density of states is plotted at $E = 0$, for a $d$-wave symmetric order parameter facing the junction interface with one of the four lobes ($\beta = 0$) or the lines of nodes ($\beta = 45^\circ$) (see Section 1.4.2). Note that for these two
orientations the oscillation is dephased. Thus, the maxima are located at $n\pi\xi_F$ with $n$ even for anti-node orientation or odd for the lines of nodes.

The available experimental evidence regarding the manifestation of LOFF state in the bulk of a superconductor is inconclusive [47]. However, convincing LOFF effects, naturally related to the pairing modulation, were obtained in the proximity region of tunneling heterostructures, even though any explanatory attempt should discriminate and explain the rich physics at the junction interface altering the state [86] (see Chapter 3 of this thesis) as well as to circumvent the technical difficulties due to the shallowness of the coherence layer [87]. As example, different groups reported spatial oscillation of the critical temperature and critical current [88, 89] in the vicinity of ferromagnet-conventional superconductor barriers. The order parameter oscillation in the proximity region was measured on Al/Al$_2$O$_3$/PdNi/Nb sandwich junctions [90]. It was clearly shown how the conductance spectra for different thicknesses $d$ of the proximity region flips upside down as the order parameter changes from negative values (0-state) to negative values (π-state). Even though the particularities of the LOFF state could be of real interest when the bulk superconductive state has a magnetic ingredient, as seems to be the case of cuprates, reports on similar experiments involving high temperature superconductors are at best scarce or oriented along the $c$-axis [91]. Part of the research at the root of this thesis, our group performed spin injection measurements into the $ab$-plane of a high temperature superconductor structure [92], as we shall see in the following chapters.
1.4 Tunneling spectroscopy

Shortly after the advent of BCS theory, I. Giaever proved the efficacy of tunneling spectroscopy [93], a simple but powerful technique to characterize the energy density of states (DOS) and hence the energy gap and the phonon distribution in superconductors [94]. This method is largely employed due to its high energy resolution and accessibility to external controlled influences like temperature, magnetic fields or pressure gradients. Although it measures only the surface DOS, its results can be easily extrapolated to the bulk in conjunction with other experimental procedures and theoretical models.

1.4.1 Principles

If two metallic electrodes are separated by a very thin insulating barrier (most commonly just an oxide layer) and a small voltage bias $V$ is applied across this junction, a current will pass through the barrier due to the quantum tunneling effect. If the electrode is a superconductor, only quasiparticles will tunnel since the tunneling probability for pairs is small. Hence, there is quantitative correspondence between the $I$-$V$ characteristic and the density of excitations $G(\varepsilon)$ in the electrodes. The $I$-$V$ characteristic will be nonlinear and, at low temperatures, its conductance normalized by the normal curve, $\sigma(V) = (dI/dV)_s / (dI/dV)_N$, will measures directly $G(\varepsilon)$. For instance, for a superconductor-insulator-normal (SIN) junction, if we assume that the superconductor is BCS type, the electronic density of states (DOS) is given by (1.44), so that the conductivity is

$$\sigma_{SN}(V) = \int_{-\infty}^{\infty} G(\varepsilon) \frac{\partial}{\partial(eV)} f(\varepsilon + eV) \, d\varepsilon \approx G(eV),$$

(1.104)

provided the temperature is low enough for the Fermi distribution $f(\varepsilon)$ to approximate a $\delta$-function. Likewise, if the junction connects two superconductors (SIS) with densities of states $G_1(\varepsilon + eV)$ and $G_2(\varepsilon)$ BCS type, the tunnel current is

$$j_{SS} \propto \int_{-\infty}^{\infty} \frac{\varepsilon + eV}{\sqrt{(\varepsilon + eV)^2 - \Delta_1^2}} \sqrt{\frac{\varepsilon}{\varepsilon^2 - \Delta_2^2}} \left[ f(\varepsilon) - f(\varepsilon + eV) \right] d\varepsilon.$$  

(1.105)
Fig. 1.10 exemplifies this behavior schematically using the so-called semiconductor model. We see that, while in the SIN the energy necessary for a tunnel quasiparticle to pair is the gap energy $\Delta$, in the SIS the threshold is $\Delta_1 + \Delta_2$. At finite temperatures the thermally excited quasiparticles can occupy the free states in the lower energy junction side up to a threshold bias $|\Delta_2 - \Delta_1|$ from where the current decreases with decreasing number of free states, to increase again at $\Delta_1 + \Delta_2$. For the ferromagnetic case, this model cannot really catch the difference from the normal case since the spin imbalance induced by polarization affects mainly the spin related effects like Andreev bound states discussed in the next section, depending on the thickness of the counterelectrode.

The junction fabrication is intrinsically an art, since the tunneling channel competes with other influences like the magnetic nanoparticles in the insulator or the Coulomb blockade [95]. The junctions can be built either by depositing thin film counterelectrodes (sandwich planar junctions), by touching the surface with a sharp tip (point contact junctions), by scanning the surface with an atomic size tip (Scanning Tunneling Microscopy aka STM) or by braking crystals and reconnecting them (break junctions).

1.4.2 Theoretical models. BTK model and Andreev bound states.

The tunneling scenario presented above is somewhat simplistic since the junction interface in reality is more that a exclusive tunnel vehicle for quasiparticles excited above the energy gap.
Thus, the conductivity profile generated by a bias voltage scan over a tunneling junction commonly presents a subgap finite value and a distribution of peaks sustained by the occurrence of an additional transmission channel: the so called Andreev reflection [41] leading to a transmitted electron and a retro-reflected hole (or vice-versa) in the vicinity of Fermi level. An electron-like excitation with energy less than the superconducting gap, incident from the normal side onto the junction barrier, cannot tunnel if it doesn’t pair with another electron beyond the interface. Once the pair is formed, a vacancy appears in the Fermi sea on the normal side, that is a spin flipped hole is reflected conserving the energy and momentum across the barrier (Fig.1.11a). Alternatively, with time reversal, a hole is absorbed, a pair is destroyed and an electron is retro-transmitted. The transmission of a pair into the superconductor translates into a two-fold resistance decrement so that the tunneling conductance spectrum may present a nonzero subgap distribution of states which can actually dominate the charge and spin transport through the junction, depending on the crystal-to-interface orientation.

The retro-reflectivity is characteristic to normal counterelectrodes and means that the incident quasiparticle and the reflected one travel along the same path albeit in different directions. However, if an exchange potential \( h \) acts inside the counterelectrode, the retro-reflectivity is lost which is a direct consequence of the difference between the momentum magnitude of the LOFF paired electrons. This can be seen on the junction diagram in Fig.1.12 where the injection trajectory making an angle \( \theta \) with the interface is characterized by the wave vector

\[
 k_{\pm,i} = \sqrt{2m/(\hbar^2)}(\varepsilon_F \pm h)
\]

and, since the momentum is conserved, the Andreev reflected quasiparticle is reflected under an angle \( \theta_\downarrow \) given by

\[
 \sin \theta_\downarrow = \frac{k_{\uparrow}}{k_{\downarrow}} \sin \theta, \quad (1.107)
\]

such that \( \theta_\downarrow \neq \theta \) unless \( h = 0 \). The transmitted Cooper pair and the reflected hole remain coherent a time proportional to the coherence length and the electron-hole conversion occurs within this distance directly related to the local superconductive pairing symmetry. Consequently, if the superconductor’s order parameter phase is \( \Phi \), the phase of the reflected
Figure 1.11 a) Schematic representation of a quasi-electron Andreev reflected into a quasi-hole. b) Quasiclassical image about the formation of Andreev bound states at the boundary between a superconductor and a thin normal layer. When the order parameter is anisotropic, the Andreev reflected quasiparticles feel different pairing potentials $\Delta_+ \neq \Delta_-$ unlike the s-wave case when $\Delta_+ = \Delta_-$. c) The expected spectra for s-wave order parameter calculated using the BTK model presented in the text. The curves were calculated by the author in order to fit temperature dependent spectra obtained on a double gap MgB$_2$ sample (compare with Fig.1.3b).

quasiparticle shifts by $\alpha(\varepsilon) \mp \Phi$, where the sign changes depending on the tunneling direction and $\cos \alpha = E/\Delta$ gives the reflection phase shift.

The Andreev reflection is responsible for the onset of the so called Andreev bound states (ABS) at the surface of superconductive surface and at the bulk grain boundaries [42] similar to bound states at a normal metallic surfaces. The states have energies corresponding to the constructive interference between the incident and reflected quasiparticle excitations. If the superconductor has an isotropic pairing symmetry, the states distribute symmetrically around the Fermi level, while the anisotropic pairing allows bound states to form at zero energy since the order parameter changes sign along the quasiclassical trajectory [61, 96, 97]. Specifically, let’s consider, as in Fig.1.11b, a superconductor in contact with a normal layer of thickness $d$ comparable to the coherence length $\xi$ (that is a proximity layer). In the quasiclassical picture, one bound state corresponds to each closed quasiparticle trajectory accumulating a total phase shift of $2\pi$ [98]. As visible in the figure, it contains contributions from two Andreev reflections, two specular reflections on the back wall given by the phase changes $\alpha_{1,2}(\varepsilon)$ and four ballistic
trajectories inside the normal layer corresponding to the net phase difference \( \chi(e) = 2d(k_f - k_i) \approx 4d e/(\Delta \xi \cos \theta) \). Therefore, the quantized total trajectory phase yields:

\[
-(\alpha_1 + \alpha_2) \pm (\Phi_2 - \Phi_1) + \chi = 2n\pi ,
\]  

(1.108)

where \( \Phi_{1,2} \) are the phases of the pairing potential as seen by Andreev reflected quasiparticles at different locations on the interface. If the order parameter is isotropic, we see that the \( \Phi \) is uniform at any point on the barrier and the bound state energy is given by a dispersion equation solutionless at \( e = 0 \):

\[
\frac{e}{\Delta} = \pm \cos \left( \frac{e}{\Delta} \frac{2d}{\Delta \xi \cos \theta} \right) ,
\]  

(1.109)

such that the bound state can exist even if \( d \) vanishes.

Figure 1.12  a) Qualitative model of transmission and reflection processes taking place at the junction boundary \( V\delta(x) \) between a metallic counterelectrode (normal or ferromagnetic) and a \( d \)-wave superconductor with a step-function order parameter \( \Delta(k)\Theta(x) \) (adapted from Refs. [99, 100]). Note that the Andreev retroreflectivity doesn’t hold in the ferromagnetic case. b) Theoretical spectra calculated using the anisotropic BTK-type model for a pure \( d \)-wave order parameter orientation \( \beta \) varying between \( \{100\} \) and \( \{110\} \) crystal surfaces.

On the other hand, if the order parameter is anisotropic and changes sign after each specular reflection as in the case of \( d \)-wave symmetry (1.77), the phase is orientation dependent, i.e., with the notations from Fig.1.12b,
\[ \Delta_z = \Delta_d \cos 2(\theta \mp \beta), \quad (1.110) \]

and extra quasiparticle states are added to satisfy the new boundary conditions. Remarkably, in this situation we have \(|\Phi_2 - \Phi_1| \geq 0\) with a maximal \(\Phi_2 - \Phi_1 = \pi\) when \(\beta = \pi/4\), so that equation (1.108) admits solutions at \(\varepsilon = 0\) for any \(-\pi/2 \leq \theta \leq \pi/2\), which corresponds to an unrestrictedly high zero bias conductance peak (ZBCP) in the tunneling spectra near \langle 110\rangle crystal surface. Nevertheless, when the current injection probes a surface with \(\beta = 0\) or \(\pi/2\), the zero energy states are prohibited.

The impact of Andreev reflection on the current distribution integrated into tunneling spectra has been studied theoretically using scattering methods as well as Green’s function techniques [101]. One of the most successful scattering formulation was given by G.E. Blonder, M. Tinkham and T.M. Klapwijk based on Bogoliubov-de Gennes equations [102] presented here in Section 1.1.2. Their formula, often called \textit{BTK model}, accounts for the scattering probabilities associated with the processes at the junction barrier seen as a \(\delta\)-function with a dimensionless strength \(Z\). The model is one-dimensional, thus ignoring the contribution of quasiparticles with momenta parallel to the interface. It also ignores the proximity effect and assumes that all particles incident on the barrier follow equilibrium Fermi distributions with energy difference \(eV\) between quasiparticles incoming from S and N sides. The net charge transport across the junction congregates the quasiparticles Andreev reflected, specularly reflected, normally transmitted and branch-crossing transmitted resulting in pairs. The respective probabilities, \(A(\varepsilon)\), \(B(\varepsilon)\), \(C(\varepsilon)\) and \(D(\varepsilon)\), were calculated using BdG equations (1.76), with Bogoliubov-Valatin transformations (1.74) suited to conserve particle number by the addition of two operators for pair creation and destruction. Then, the normalized junction conductance through the barrier for \(\varepsilon < \Delta\), where only the first two processes occur, writes

\[
\frac{\sigma_{SN}(V,Z,T)}{\sigma_{NN}} = (1 + Z^2) \int_{-\infty}^{\infty} \left[ 1 + A(\varepsilon) - B(\varepsilon) \right] \left[ \frac{\partial}{\partial \varepsilon} f(\varepsilon - eV) \right] d\varepsilon. \quad (1.111)
\]

The barrier parameter \(Z\) is only \textit{grosso-modo} the height of the barrier. In a generalized picture, it actually measures the strength of different influences upon the scattering mechanisms acting in the junction, like the S-N Fermi velocity mismatch [103] or the magnetic accumulation introducing an exchange amplitude in the barrier [86, 104, 105] (see Chapter 3).
The BTK formalism was extended in the last decade in order to treat anisotropic $d_{x^2-y^2}$-wave or mixed $(d,s)$-wave superconductors [97, 99, 106, 107], the respective theories showing considerable consistency with the tunneling experimental investigations [86, 108]. Here is the outline of the argument, using Fig.1.12 as a pictorial framework: let’s recall BdG equations (1.76) with $\delta\mu = 0$ so, in terms of the Hartree potential $U(r)$, $H_0(r) = -\left(\hbar^2/2m\right)\partial_r^2 - \mu + U(r)$ for a normal counterelectrode. The electronlike and holelike quasiparticle states $(u,v)$ in a superconductor with spatially varying pairing potential $\Lambda(r_1,r_2)$ must satisfy:

$$H_0(r_1)u(r_1) + \int dr_2\Delta(r_1,r_2)v(r_2) = \varepsilon u(r_1)$$

$$-H_0(r_1)v(r_1) + \int dr_2\Delta^*(r_1,r_2)u(r_2) = \varepsilon v(r_1).$$

(1.112)

The pair potential can be written in the center of mass coordinate $r = (r_1 - r_2)/2$ and relative vector $s = r_1 - r_2$. Therefore, its Fourier transform depends on momentum which, in the weak coupling limit, lays fixed on Fermi surface and depends only on direction $\theta$:

$$\Delta(k,r) = \int ds e^{-ik\cdot s} \Lambda(s,r) = \Delta(\theta, r).$$

(1.113)

In the quasiclassical approximation, the fast oscillating part of the wave function (1.71) can be separated:

$$\begin{pmatrix} \overline{u}(	heta, r) \\ \overline{v}(	heta, r) \end{pmatrix} = \begin{pmatrix} u(\theta, r) \\ v(\theta, r) \end{pmatrix} e^{ik\cdot r},$$

(1.114)

and equations (1.112) reduce to Andreev equations [41] which, for the geometry in Fig.1.12 with $x$-axis perpendicular on the barrier, take the form:

$$-iv_x k_F \partial_x \overline{u}(\theta, x) + \Delta(\theta) \overline{v}(\theta, x) = \varepsilon \overline{u}(\theta, x)$$

$$iv_x k_F \partial_x \overline{v}(\theta, x) + \Delta^*(\theta) \overline{u}(\theta, x) = \varepsilon \overline{v}(\theta, x).$$

(1.115)

We are interested only in the solution of this system in the region adjacent to the superconductor ($x < 0$) (the proximity layer). Then, when a current is injected under an angle $\theta$ with respect to the $x$-direction, the solution in this region is:

$$\varphi = \begin{pmatrix} 1 \\ 0 \end{pmatrix} e^{ik_x \cos \theta} + a(\varepsilon) \begin{pmatrix} 0 \\ 1 \end{pmatrix} e^{ik_x \cos \theta} + b(\varepsilon) \begin{pmatrix} 1 \\ 0 \end{pmatrix} e^{-ik_x \cos \theta},$$

(1.116)
where the ± signs mark the transmitted and Andreev reflected quasiparticles. The probability coefficients $|a|^2$ and $|b|^2$ can be calculated and one gets:

$$a(e) = \frac{4\Gamma_\Omega \Omega_+}{(1+\lambda)^2 + 4Z^2 - [(1-\lambda)^2 + 4Z^2] \Gamma_- \Omega_- / \Omega_+},$$

$$b(e) = \frac{-\left(1-\lambda^2 - 4iZ - 4Z^2\right)(1-\Gamma_- \Omega_- / \Omega_+)}{(1+\lambda)^2 + 4Z^2 - [(1-\lambda)^2 + 4Z^2] \Gamma_- \Omega_- / \Omega_+},$$

$$\lambda = k_s \cos \theta_s / k \cos \theta, \quad \Gamma_\pm = \left(e - \sqrt{e^2 - |\Delta_\pm|^2}\right) / |\Delta_\pm|, \quad Z = mV / (\hbar^2 k \cos \theta) \quad \text{and} \quad \Omega_\pm = \Delta_\pm / |\Delta_\pm|.$$

The effective pairing potentials experienced by quasi-electrons and quasi-holes are different since they have different wave vectors, $\Delta_s(\theta) = \Delta_s(\pi + \theta)$, determining a phase difference between $\Phi_{1,2}$ in (1.108). Subsequently, the BTK formula giving the conductance in direction $\theta$ is:

$$\sigma_\theta(e) = 1 + |a(e)|^2 - |b(e)|^2.$$ (1.118)

Since in a real junction the barrier is bombarded from multiple directions, the total conductance should integrate (1.118) over all possible incidence angles. Thus, assuming a uniform angle distribution, the 2D overall conductance is

$$\bar{\sigma}(e) = \int_{-\pi/2}^{\pi/2} \sigma_\theta(e) \cos \theta d\theta.$$ (1.119)

The $d_{x'y'}$-wave order parameter adapted to the geometry in Fig.1.12b is conveniently given by (1.110), while the mixed $d + is$-wave symmetry we are to consider in this thesis can be written:

$$\Delta(\theta) = \Delta_s \cos 2(\theta - \beta) + i\Delta_\perp.$$ (1.120)

In Fig.1.12a we plotted several curves predicted by this model for different junction orientation angles $\beta$. By varying $\beta$, the $ab$-plane current probes different regions of the $d_{x'y'}$-wave order parameter. We see that, for injection near $\langle 110 \rangle$ crystal surface ($\beta = 45^\circ$), the gap is almost filled by a ZBCP, while near the $\langle 100 \rangle$ surface ($\beta = 0$) the V-shaped gap has no ZBCP, consistent with Hu’s argument regarding the onset conditions of zero energy ABS [61], even though the BTK-type model doesn’t explicitly consider the surface bound states as explained above.
The BTK-type model in this form simplifies unrealistically the junction barrier by considering it perfectly flat, fact which entails the conservation of momentum component parallel with the surface and hence its specular character. The problem concerning the effect of barrier roughness was addressed either quasiclassically [109, 110] or based on the lattice simulations [111, 112].

The first approach introduces a randomizing, multi-channel scattering matrix in the boundary condition for the wave function of incident electrons at the barrier. It mainly predicts a wash-out of spectral features. The local DOS measured for injection perpendicular onto the node lines accumulates subgap states and may show a ZBCP due to a suppression of pairing potential. Conversely, the local DOS parallel with the node lines shows a ZBCP broader and shorter with increasing roughness due to an enhanced order parameter.

On the other hand, the lattice approach, based on Hubbard and $t-J$ models discussed in Section 1.1.3, circumvents the inadvertencies of the quasiclassical approximation which is not perfectly suited to describe the nonlocal pairing in cuprates where the coupling is between next-site electron spins via antiferromagnetic spin fluctuations. Also, the lattice models provide a realistic simulation of the atomic size defects which in high temperature superconductors are more disruptive due to the short coherence length. Among the predictions of these models are some anomalous features like the asymmetry and irregularity of local DOS spectra induced by atomic size defects, especially in the quasiparticle branch. Even if these features are smeared out by the macroscopic, angle averaged DOS measured by planar junctions, the sensibility of cuprate local distribution of states to microscopic accidents makes the interpretation of such cuprate tunneling spectra much more complex than in the case of conventional superconductors.

### 1.4.3 BTRS pairing

Even if the zero energy Andreev bound state is a solid feature in the energy spectrum of d-wave superconductors, their energy stability can be easily threatened by alterations in the pairing symmetries due to the sensitivity of the degenerate superposition of states to local Fermi level instabilities. One verified destabilizing procedure [105, 113, 114] is based on the application of a weak magnetic field $H$ parallel to the junction interface, provoking the so called Zeeman splitting of states with same spatial wave functions but opposite spin directions, $g\mu_B H \approx \Delta_s$. The “external” magnetic field mustn’t necessarily originate from outside of the structure: a
ferromagnetic counterelectrode or a distribution of resonant magnetic impurities in the insulating barrier can play the role.

However, the more intriguing case is when the split ABS is triggered spontaneously as reported in connection to some tunneling experiments apparently devoid of any external magnetic influences [65, 86, 108]. There are several theories about the mechanism possibly responsible for such broken time-reversal pairing symmetries (BTRS). Let’s briefly introduce four of them:

1) For instance, as we have already noticed in Section 1.1.3, the \( d + i s \ (B_{i}g + iA_{i}g) \) pairing potential, unlike its \( d (B_{g}) \) and \( s (A_{g}) \) singlet ingredients, breaks time-reversion symmetry and favors a lower energy configuration where the ABS energy locations are shifted away from Fermi level to finite values. In principle, this scenario is consistent with the fact that, by enhancing the amplitude of quasiparticles near Fermi level by pair breaking, the ABS aids to the formation of a nodeless gap. The possibility and constraints of this phenomenon have been studied within the milieu of Ginzburg-Landau theory [107], the quasiclassical transport perspective modeled by Eilenberger’s equations [106] or the \( t - J \) model [111]. Its presence in the tunneling spectra (at temperatures low enough for the subdominant order parameter to set-on) manifests as a splitting of ZBCP akin to the Zeeman splitting. Explicitly, by setting \( Z = 0 \) in the equation for Andreev reflection coefficient (1.117), one can derive a supplementary phase shift due to the \( s \)-wave component

\[
\vartheta_{k \sin \theta < 0} = \mp \arctan \left( \frac{\Delta_s}{\Delta_d} \right) \quad \text{at the } \beta = \pi/4 \quad \text{surface of a cuprate, so that expression (1.108) becomes}
\]

\[
-(\alpha_1 + \alpha_2) + 2\vartheta + \pi + \chi = 2n\pi,
\]

that is the bound state is \( \pm k \sin \theta \)-degenerate, where the sign of the transverse momentum component \( k \sin \theta \) depends on the quasiparticle energy under or above the Fermi level. The degeneracy is lifted if the state is shifted to the edge of the \( s \)-wave gap \( \Delta_s \). The onset of a \( d + is \)-wave admixture (as well as, for that matter, \( d - is \) or \( d + id \)) is associated with spontaneous surface currents carried by ABS (see Fig.1.11b) in the direction of \( k \sin \theta \), a net current being directly observable at low temperatures where only the \( k \)-state under Fermi level is occupied.

2) However, it was suggested that the surface currents can lead to spontaneous ZBCP splitting even on pure d-wave pairing surfaces due to the paramagnetic response of ABS to the magnetic field created by these currents. The bound states couple to this field perpendicular to the
surface by counterflowing screening supercurrents of momentum $k_e$ which introduce a Doppler shift term $\delta E = \Delta_0 k_e v_F \sin \theta$ in the excitation spectrum due to the coupling term in the quasiparticle transport equations. So, since the energy levels move to higher or lower energies $\pm \delta E$, depending on the quasiparticle moving parallel or antiparallel with respect to the supercurrent, the ABS surface currents alone can in principle remove the parallel momentum degeneracy and shift the bound states to finite energy locations and hence determine a Zeeman splitting of the ZBCP.

Since Zeeman splitting is a signature of a spin rather than orbital coupling between the ABS and the magnetic field, it confirms that the current carrying superconductive condensate at the junction barrier reacts paramagnetically to the effect of the magnetic field as opposed to the Meissner diamagnetic pair breaking which dominates the bulk in the Shubnikov phase. This is due to the size of the ABS which is of the order of coherence length, in cuprates much smaller than the penetration depth, such that the spin coupling dominates. However, it was shown that this surface interplay between the paramagnetic and diamagnetic contribution depends on the temperature [115], the paramagnetic contribution being suppressed with ABS broadening under a temperature in the clean limit given by

$$T_{cs} \propto T_{cd} \frac{\xi}{\lambda}, \quad (1.122)$$

where $T_{cd}$ is the critical temperature for the bulk d-wave superconductor. However, as we discussed in the previous section, the surface roughness is also expected to contribute to the ABS broadening and accordingly to the $T_{cs}$ value.

3) Another possible mechanism counts on a spin dependent surface DOS via a surface phase transition to a local spin density wave state. This alternative likewise summons the action of a magnetic field, but, unlike the previous mechanisms which lift the charge degeneracy of ABS, it removes the spin degeneracy through a spin imbalance among the quasiparticles involved in ABS. The magnetic field emergence on the surface and grains of the cuprate superconductor is assured by antiferromagnetic fluctuations. Honerkamp and Sigrist [116] discussed this scenario using a mean field BdG approach over a d-wave Hubbard lattice model. For finite onsite repulsion, the spin degeneracy is removed, a modulated magnetic ordering
decays with distance from the boundary and the ABS corresponding to spin up and down move apart to energies flanking Fermi level.

4) Nevertheless, magnetism may not be an indispensable drive in the onset of BTRS. Another scenario – which works even if the pairing is pure \( d \)-wave – is based on the proximity effect and the assumption of a junction barrier transparent enough to couple the two superconductive regions by a finite difference of phase calculated between values far away from the interface \( \delta \phi \neq n(0, \pi) \), \( n \) integer, corresponding to a minimum free energy at the interface with \( \pi/4 \)-misorientation \([117, 118] \). The ABS with this minimum energy is \( \pm \delta \phi \)-degenerate, the time reversal symmetry is spontaneously broken and the zero energy ABS are pushed to finite energies. Since a highly transparent barrier and precise crystal orientation entails the efficiency of this mechanism, it may be easily affected by nonuniform transmission and interface roughness. Also, it does not exclude the influence of an attractive \( s \)-wave channel added to the \( d \)-wave: at lower junction transparencies this additional time-reversal breaking influence may become dominant.

### 1.4.4 Modeling superconductor/ferromagnet tunnel junctions

The main differences between tunneling structures joining superconductors with normal and ferromagnetic counterelectrodes stem from the attributes of subgap channels, particularly Andreev reflection. Thus, let’s recall that Andreev reflection does not conserve spin, the transmitted and reflected quasiparticles having opposite spins. The normal metals have spin-rotation symmetry, meaning that the probability amplitude for the spin current is the same for spin up and down. Conversely, the exchange energy in the ferromagnet leads to an unbalanced availability in the two spin bands (i.e., creating a minority and a majority band) such that the band change associated with Andreev reflection is hindered and the respective channel is completely suppressed against highly polarized currents. For lower exchange potentials, the Andreev reflection survives with an amplitude diminished by \( 1 - P \), where \( P \) is the polarization defined by (1.85), and somewhat modified properties like the lost of retro-reflectivity (as we’ve already seen in Section 1.4.2) as well as the possibility of the so called virtual Andreev reflection (VAR) which still results in a transmitted pair but without the reflected quasiparticle. Moreover, the dynamics of ABS is substantially changed in the ferromagnetic vicinity of the barrier: in this
proximity region, ferromagnetism and superconductivity cooperate to form the afore discussed LOFF state, such that the distribution of ABS throughout the gap and hence the shape of DOS spectrum shows an oscillatory dependence on the distance from the interface. Also, in contrast to the normal case, the junction barrier itself is less inert [119, 120]; thus, the spin current may induce an exchange amplitude in the barrier sensitive to the spontaneous magnetic fields in the ferromagnet and with filtering effects decisive vis-à-vis the branch-symmetry of the spectra.

Tanaka and Kashiwaya’s reformulation of BTK model to include anisotropic pairing symmetries [99] can be once more adapted in order to describe high temperature superconductor-ferromagnetic junctions [100, 121]. The immediate modification consists in the introduction of the exchange potential \( h = \delta \mu \) in the BdG equations and redefining the \( \delta \)-function barrier function \( V_{\uparrow,\downarrow}(x) = (V \mp U) \delta(x) \) to include an exchange amplitude \( U \) intended to model a spin filtering ferromagnetic insulator. With these additions, the wave functions for the spin up and down quasiparticles in the counterelectrode aren’t equal anymore (see equation (1.106)), the respective conductance contributions \( \sigma_{\uparrow,\downarrow} \) are spin discriminated and the retro-reflectivity in Andreev reflection processes is lost that is, in Fig.1.12a, \( \theta \neq \theta_A \). Also, since the total reflection angle range is \( \theta > \arcsin(k_y/k_x) = \theta_1 \), the subgap DOS is fuelled only by contributions under this angle which is \( \approx \pi/2 \) in the normal case. For angles \( \theta \) in the range \( \theta_1 > \theta > \theta_2 = \arcsin(k_y/k_x) \), the Andreev reflection takes place without a reflected quasiparticle (VAR region). Only for \( \theta < \theta_2 \) the transport composition is qualitatively the same as in the normal case. In these conditions, the angle integrated total conductivity must take into account only the contributions from the two angle ranges with nonvanishing Andreev spin and charge currents (see Appendix A).

Besides these consequences, in the presence of a ferromagnet, the barrier feels differently the spin currents:

\[
Z_{\uparrow,\downarrow} = \frac{2m}{\hbar^2 k_y \cos \theta_A} (V \mp U), \tag{1.123}
\]

such that the majority carriers will have a higher contribution to the ABS and the amplitude of the respective spectral peak will be enhanced in comparison with the minority one by a ratio proportional to the ferromagnetic polarization. Therefore, the energy shift induced by a BTRS
pairing potential would result into an asymmetric spectrum which allows an estimation of the \( P \) value, since the net conductance, unlike the normal case (1.119), sums up the separate contributions \( \sigma_{\uparrow,\downarrow} \) weighted by respective polarization coefficients:

\[
\bar{\sigma}(E) = \bar{\sigma}_\uparrow(E) + \bar{\sigma}_\downarrow(E),
\]

\[
\bar{\sigma}_{\uparrow,\downarrow}(E) \propto \int_{-\pi/2}^{\pi/2} d\theta \cos \theta \sigma_{\uparrow,\downarrow}(E, \theta) P_{\uparrow,\downarrow} k_{\uparrow,\downarrow}.
\]

where, in the terms of this model,

\[
P_{\uparrow,\downarrow} = (\epsilon_e \pm U) / 2\epsilon_e.
\]

In fact, we use this “polarized” approach in all our anisotropic BTK fittings, modeling the normal situations by simply setting \( U = h = 0 \), which reduces to \( \sigma_\uparrow = \sigma_\downarrow \).

A problem with the BTK approach in the forms presented above is the step-function model of order parameter neglecting the proximity effect. Such a behavior is expected when the transmission probability of the interface is small enough to decouple the electrodes and suppress the proximity correlations. A more realistic picture in this respect is obtained if the pair amplitude beyond the barrier is obtained self-consistently from the gap functions emanated by an extended Hubbard square lattice model, which in mean fields quasiclassical approximation obeys the BdG equations [112]. In this context, the presence of the proximity effect is demonstrated by the presence of a subgap in the \( d \)-wave case and the pair amplitude in the proximity region decays monotonically in a normal counterelectrode and oscillatory in the LOFF environment on the ferromagnetic side. The local DOS shows the same properties predicted by the BTK formula: reduced subgap state distribution in the \( s \)-wave case and onset of zero energy ABS (enhanced by barrier strength) only for the \( d \)-wave pairing.

However, besides the BTK-like model, in this thesis we use yet another quasiclassical model [122-124] we found suitable to simulate our planar multilayered hybrid structures. It considers the junction structure as a ferromagnetic film of thickness \( d \) in contact with a superconductive volume (see Fig.1.13a), similar to the situation presented in Section 1.3. The ballistic condition allows a multitude of classical trajectories experiencing diffusive reflections on the sides of the metallic films and subgap Andreev reflections on the superconductive interface leading to a distribution of ABS directly related to the transport properties in the ferromagnetic layer with exchange potential \( h \). Thus, to each trajectory corresponds a density of states that can be
expressed as a distribution of lengths depending on $d$. The transport in the clean limit is described by Eilenberger’s equation (1.69) which can be solved along a trajectory between two successive superconductor interface contacts. Since the diagonal Green’s function $g_\sigma$ in (1.70) depends in the ferromagnetic layer only on the path length $l(\theta) = d/\cos \theta$, the solution for the DOS along a trajectory

$$N(\varepsilon, \theta) = \frac{N_0}{4} \sum_{\sigma=\uparrow, \downarrow} \text{Re} \left[ g_{\sigma}(\varepsilon, v_F, r) \right] = \frac{N_0}{2} \sum_{\sigma=\uparrow, \downarrow} \text{Re} \left[ -i \tan \left( \sum_{j=1,2} k_{\sigma}(l(\theta_j) + \arcsin \frac{\varepsilon}{\Delta(\theta_j)} \right) + \Theta \right],$$

(1.127)

where $N_0$ is the DOS when $h = 0$, the wave vector $k_{\uparrow, \downarrow} = (\varepsilon \pm h)/v_F$ and $\Theta = \Phi_2 - \Phi_1 = \frac{\pi}{\delta} \left( 1 - \Delta(\theta_1) \Delta(\theta_2) \right)$ is, as in (1.121), the difference between phases experienced by the quasiparticle at the end and beginning of a trajectory under the respective angles $\theta_{1,2}$. The order parameter $\Delta(\theta)$ is still given by (1.120). The total density of states is then the angular integral:

$$N(\varepsilon) = \int g(\theta_1, \theta_2) N(\varepsilon, \theta) d\theta,$$

(1.128)

Figure 1.13 a) Schematic representation of SIF junction with $d$-wave pairing. In the ferromagnetic thin film the ABS density is related in the quasiclassical approximation to the trajectory length distribution and thus to the geometrical properties of the film, in particular its thickness $d$. b) Trajectory length distribution for diffusive case for high and low barrier transparency $T$. Note that the two peaks corresponding to the shortest lengths dominate at high $T$. 

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where the correlation function \( g(\theta_1, \theta_2) \approx \delta(\theta_2 - \theta_1) \) for flat surfaces and is approximately constant for very rough surfaces. The correlation can be extended to account for the scattering inside the film represented by the mean free path \( \ell \) and, defining an interface roughness \( z \), we have in general \( g \propto \exp\left[-l/|\ell - (\theta_1 + \theta_2)|^2/z^2\right] \). Eventually, the DOS winds up being:

\[
N(\varepsilon) = \frac{N_0}{2} \sum_{\sigma = 1, k} \sum_{n=\infty}^{\infty} \Gamma_n(nk_\sigma),
\]

where

\[
\Gamma_n(q) = \int_{-\infty}^{\infty} g_{r_n}(\theta_1, q) g_n(\theta_2, q) e^{-i\sigma \theta} d\theta d\theta_2,
\]

provided

\[
\gamma_n(\theta, q) = \begin{cases} 
\exp\left[-i2q\ell + i\arccos\left(\varepsilon/|\Delta(\theta)|\right)\right], & |\varepsilon| \leq |\Delta(\theta)| \\
\exp\left[-i2q\ell - i\rho|a\cosh\left(\varepsilon/|\Delta(\theta)|\right)\right], & |\varepsilon| > |\Delta(\theta)|.
\end{cases}
\]

Graphically, the DOS spectrum given by this procedure predicts a decaying modulated behavior in antiphase for \( \beta = 0 \) and \( \beta = \pi/4 \), similar to the curves in Fig.1.9b.

To get a more intricate physical idea about the processes in the ferromagnetic layer, the average (1.128) can be retranslated into an integral over the trajectory length distribution \( p(l) \):

\[
N(\varepsilon) = \int p(l) N(\varepsilon, l) dl,
\]

expressing a subgap DOS composed of a superposition of ABS weighted by \( p(l) \).

Conversely, the DOS above the gap is formed exclusively by scattering resonances. Zareyan et al. [123] calculated the length distribution for a junction with transparency \( T \) and counterelectrode thickness \( d \) and it writes:

\[
p(l) = \frac{T}{2\pi} \int_{-\infty}^{\infty} \frac{\tilde{p}(q) e^{iql}}{1 - (1 - T) \tilde{p}(q)} dq,
\]

where \( \tilde{p}(q) = E_2^2(iq d/d/\ell)/E_2^2(d/\ell) \), \( E_2(x) = \int_{-\infty}^{\infty} dt \exp(-xt)/t^2 \) being the second order exponential integral. In Fig.1.13b the \( p(l) \) function for different transparencies is generated for \( d/\ell = 0.1 \). The curve shows two peaks at the shortest lengths and decays as \( \exp(-l/T) \) due to the scattering term \( d/\ell \) which determines the average length \( T \). At high transparencies, the
tunneling is dominated by the ABS corresponding to the shortest trajectories and the credibility of quasiclassical approach is enhanced.

For an isotropic gap, the DOS yields:

\[
N(\varepsilon) = \frac{N_0}{2} \sum_{\sigma=\uparrow,\downarrow} \sum_{n=-\infty}^{\infty} \tilde{p}(2nk_\sigma) e^{i2\pi n \text{arccos}(\varepsilon/\Delta)}, \quad \varepsilon \leq \Delta
\]

\[
N(\varepsilon) = \frac{N_0}{2} \sum_{\sigma=\uparrow,\downarrow} \sum_{n=-\infty}^{\infty} \tilde{p}(2nk_\sigma) e^{-|\varepsilon| \text{coth}|\varepsilon/\Delta|}, \quad \varepsilon > \Delta
\]

which disregards the $\theta$-dependency of the gap, but can offer qualitative predictions regarding the LOFF order parameter properties, as pictured in Fig.1.14. See Appendix B for a computer implementation and Chapter 4 for our experimental confirmation.

Figure 1.14 Reduced DOS ($N/N_0-1$) oscillations with counterelectrode thickness in the LOFF proximity area, as predicted by the ballistic model described in the text based on the Eilenberger’s equation in the clean limit.
CHAPTER 2
Experimental setup

In this chapter we introduce some of the most relevant procedural and technical elements concerning our experimental installation. Also, we present some specific characteristics of the cuprate samples and the ultrathin films fastidiously prepared for LOFF state detection.

2.1 Constructive details

We built a series of tunnel junction in principle based on the same idea, but in fact with substantially diverse purposes leading to various architectures. All our structures presented here involved crystals of BSCCO, more or less underdoped, forming junctions with metallic thin film counterelectrodes or sandwiches of thin films wired separately. As demonstrated by Giaever – the inventor of tunneling spectroscopy –, the insulating layer appears naturally by \textit{ex situ} surface oxidation. The measurements were performed in a four-point standard procedure, with voltage $V$ and current $I$ measured simultaneously. The conductivity was evaluated by numerically differentiating the $I$-$V$ characteristics, such that all our spectra $dI/dV$ represent only approximated DOS values over the true energy range which is sufficient for our purposes exclusively focused upon observing qualitative behavior and measuring energy gap values and other related quantities. The temperature variation was obtained by positioning the probe at different levels down the temperature gradient inside liquid helium dewars.

All our structures can be qualified as planar junctions. There are two main reason why such a constructive arrangement is most suitable for our experiments. First, since most of our measurements consist in sets of tunneling spectra collected at different temperatures, thermal stability is an important criterion. Unlike other similar experimental techniques, like STM or tunneling across mechanical junctions very sensible to structural modifications, planar junctions show a remarkable reliability in terms of thermal stability and mechanical strength even if they slowly degrade in time. Therefore, excluding instabilities, the transport properties in planar junctions depend only on phenomenologically intrinsic influences like the electronic,
nonstructural thermal, magnetic (spin) and geometrical characteristics of the electrodes, strength and filtering effects of the tunnel barrier and, of course, on the properly scaled bias voltage.

Second, recall that in search of LOFF state manifestations, we are interested in the proximity area presumably arising in the vicinity of the barrier on the ferromagnetic side. Admittedly, spin polarized current experiments are also possible using STM since the spin-dependent tunneling can be approached locally in the framework of theoretical models like the ones proposed by Slonczewski [125] or Julliere [126]. However, besides problems like magnetostriction leading to tip-ample separation uncertainty, there are phenomena crucial in the study of anisotropic superconductors, like the full range ABS dynamics, proximity effect and mixed pairing symmetries, which cannot be properly addressed using STM. The nonlocal, stable and versatile planar junctions seem to be more suited in this case.

As we have seen in the previous chapter, according to the most popular opinions, the \( d \)-wave superconductivity in cuprates is confined in the \( \text{CuO}_2 \) planes. Most experimental studies probe this direction on cuprate epitaxial thin films with different crystal orientations (ramp-edge junctions) but with a considerable interference from the \( c \)-axis conductivity due to the influence of roughness bringing about a different sort of pairing. On the other hand, in the case of experiments performed on single crystals, even though the effect of roughness cannot be avoided, the eventual averaging is done over the same type of order parameter favored by the higher crystal homogeneity. Also, monocrystals are more stable with respect to the oxygen doping level and mechanically more enduring. Consequently, we used in our measurements, monocrystals of \( \text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_{8-\delta} \) grown in our laboratory by the self-flux method [127]. Taking advantage from the extremely micaceous nature of BSCCO, the monocrystals were cleaved into rather large samples of 1-4 mm\(^2\) (see next section for comments). The respective \( I-V \) contacts were made under microscope through trails of silver epoxy binding the crystal to copper macroleads. However, to have \( ab \)-plane injection, the junction must be built on the edge of this extremely thin, very fragile piece. So, the lower side of the crystal is embedded in a mold of Varian Torr seal epoxy, a material very stable against temperature variation. Before the epoxy dries, the upper side is fixed between two quartz glass slabs treated with a thin spread of epoxy, such that the crystal is eventually caught between two smooth glass surfaces attached by thin walls of resin. After it solidifies, the whole structure is embedded in more epoxy. The purpose of the glass siding the crystal is to facilitate a smooth mechanical polish for thin film evaporation.
Subsequently, the mold is mechanically polished onto the side of the crystal using a Buehler Grinder in the direction parallel with the crystal $ab$-plane, first coarsely onto 600 grit grinding paper with nitrogen jetting away residue until the crystal edge is exposed. Then, a finer polishing is applied down to 0.3 $\mu$m, using alumina grinding powder. For convenience, a number of copper wires can be embedded in epoxy on the sides of the BSCCO crystal, such that the grinding exposes their cross-section. These can serve as macroleads for the metallic counterelectrodes which are thermally evaporated through a mask placed on the surface, in an evaporation chamber vacuum of $\approx 10^{-7}$ Torr.

![Figure 2.1](image_url)

**Figure 2.1** Pictures and schematic representations of tunnel junctions. *a)* Two junctions built on the same crystal under two orientations ideally differing by $\pi/4$. The scheme depicts the case analyzed in Chapter 4 where the two junctions contain a ferromagnetic interlayer between two normal layers. *b)* Multilayer hybrid structure intended to probe the LOFF order parameter behavior at five increasing distances from the junction barrier in the proximity region.

In this thesis we present a collection of tunneling spectra obtained on junctions using as counterelectrodes materials with different properties: normal metals (Pb above its $T_c$ and Ag), a conventional superconductor (Pb, $T_c \approx 7.2$ K), and a ferromagnetic metal (Fe). While for some of the junctions the thickness of the metallic thin film counterelectrode is irrelevant, this will become an important parameter in Chapter 4 when the presence of LOFF state will be investigated in the proximity region.
These basic procedures were changed to fit the necessities of each experiment. Two types of junction layouts used in this thesis are presented in Fig.2.1. First structure (a) contains in fact two junction on the same BSCCO crystal such that the tunneling characteristics can be probed in the same conditions under two angles \( \beta_2 - \beta_1 = \pi/4 \) (with the only uncontrollable parameter being the barrier strength \( Z \)). Therefore, essentially, for a \( d \)-wave anisotropic superconductor, the DOS measured into the two directions should sign out a different order parameter phase with exception of the antinode injection. This kind of junction is employed both in Chapter 3, to exemplify the phase dependency of ABS and in Chapter 4, to show that the phase and amplitude of LOFF order parameter oscillation change with the bulk order parameter orientation. Second structure (Fig.2.1b) is used in Chapter 4 to probe the onset of LOFF state at increasing distance from the interface. Toward this purpose, six 30 Å layers are deposited one on top of each other (Fig.2.3b). First layer is Ag, next are Fe thin films. It is practically the same junction (same \( Z \)) but with increasing counterelectrode thickness and increased in-film scattering rate which makes a simplistic quasiclassical approximation less and less applicable. Each lead (i.e., thickness) can be probed separately and the junction is reliable only once due to the destruction of interlayer interfaces. The main purposes of the first Ag layer (in all our kinds of ferromagnetic junctions) are as following: to provide a normal layer comparison for the ferromagnetic layers built onto the same crystal (i.e. with the same orientation and barrier strength), to minimize the barrier strength in order to increase transparency and thus aid to the applicability of quasiclassical models (see Fig.1.13b) and to avoid the appearance of a metastable spin-phase or disordered ferromagnetism at the junction interface [128].

2.2 Sample and thin film characterization

BSCCO is a perovskite compound with almost tetragonal unit cell (see Fig.1.5), the lattice constants being \( a \approx b \approx 5.4 \, \text{Å}, \ c \approx 30.89 \, \text{Å} \). It has two CuO\(_2\) layers intercalated by Ca atoms and flanked by symmetrical semiconducting BiO and insulating SrO layers. Cu atoms are surrounded by pyramids of in-plane and weaker bound apical oxygen atoms. Our BSCCO monocrystal samples were mechanically peeled off the batch, immediately before being processed. In general, one assumes that the BSCCO crystals cleave along the adjacent BiO planes connected by the longest and weakest bonds. However, STM studies [129] have shown that, while BiO planes are
most likely to be exposed, mechanical cleaving usually uncovers multiple terraces and step edges of any atomic plane (see Fig.2.3a). This is the reason why some of our resistivity measurements performed through a 4-point procedure with contacts on the surface of the crystal may contain some contribution from the c-axis directed current and some samples even show a semiconducting surface behavior due to BiO planes.

**Figure 2.2**  a) Temperature dependence of the ab-plane resistivity of some of our samples. All of them show the pseudogap transition at temperatures $T^*$ under the form of a downward deviation in resistivity decreasing with decreasing temperature. b) Critical temperatures $(T_c, T^*)$ – extracted by inspection from the $\rho_{ab} - T$ curves –, function of hole doping $\delta$ (oxygen content), delineating the phase diagram for our samples.

In order to establish the critical temperature $T_c$ and hence the doping range of our samples, we collected a set of ab-plane-resistivity versus temperature ($\rho_{ab} - T$) curves corresponding to the different BSCCO batches we used in our measurements. As shown in Fig.1.4, the $R - T$ characteristics are expected to take different shapes in the underdoped, optimally doped and overdoped regimes such that a one can place a curve corresponding to the same $T_c$ in the respective region by analyzing the shape. As one can see in Fig.2.2a, our curves present the pseudogap steeper downturn of slope at temperatures $T^*$ increasing with decreasing $T_c$ and oxygen content. This suggests a rather underdoped to optimal situation, with a range of critical temperatures between 72 K and 87 K. The relationship between hole doping $\delta$ and $T_c$ is similar for most cuprates [130] (with exceptions) and is given by the empirical formula
\[ T_c(\delta)/T_c^{\text{max}} \approx 1 - 82.6(\delta - \delta_{\text{optimal}})^2 \]  

where the optimal doping \( \delta_{\text{optimal}} \approx 0.16 \) and, in our case, the optimal critical temperature \( T_c^{\text{max}} \approx 90 \text{ K} \). We used this expression and the \( (T_c, T^*) \) values evaluated from the \( R-T \) characteristics to calculate a doping range between 0.11-0.16. As a pictorial result, Fig.2.2b depicts the corresponding phase diagram. Note the linear \( T \)-dependency in the ab-plane; since the temperature dependency of \( c \)-axis resistivity is almost insulating in the underdoped region, this reflects a two dimensional quasi-non-Fermi liquid behavior near the optimal doping level. The pseudogap transition line may be slightly underevaluated since the line between the pseudogap and non-Fermi liquid regions should regularly cross the boundary with the superconducting phase a bit above the optimal doping as the pseudogap is still possible in the slightly overdoped area.

The doping is important for our measurements in the light of some suggestions [131] that the onset of BTRS pairing is doping dependent, that is its occurrence is enhanced with increasing doping. This would explain the fact that only some of our junctions show the splitting of ZBCP, as we shall see in the next chapter.

The thickness of the thin films utilized in this thesis accommodates the purpose of the respective experiment. It varies from ultrathin 30 Å, in the case of proximity junctions, to 500-2500 Å, in the case of DOS spectra collected on normal and conventional superconductor counterelectrodes. The ultrathin films require special attention and, consequently, the deposition was carefully monitored using a calibrated quartz thickness monitor and the BSCCO crystal was sandwiched between two glass slabs such that most of the film superposition takes place on a smoother surface. Each layer is grown separately and one assumes a minimal diffusion at the Fe/Ag interface since Fe has a very low solubility. Fe was deposited at a rate of 0.1 Å/s which proved to to help surface uniformity [132]. The residual resistivity ratio (RRR) of the resulted films was \( RRR \approx R(300K)/R(40K) \approx 8.3 \), indicating a good deposition quality. As shown in Fig.2.3b, the Ag-Fe resistance at room temperature measured on different layers was slightly different by approximately equal amounts, which proves once more the thin film uniformity. A typical resistance on the first iron layer is \( R(300) \approx 450 \Omega \).
Figure 2.3  a) SEM picture of one of our BSCCO samples [133]. BSCCO is a very micaceous material so that monocrystals can be easily cleaved. However, note the terraces which may interfere with c-direction tunneling and resistivity measurements. b) Picture of the multilayered junction in Fig.2.1b and the normal temperature I-V characteristics measured between its Ag-Fe and Ag-BSCCO leads.

As expressed by relations (1.99), the correlation span in the proximity region depends on the transport properties of the thin film, that is on its diffusion coefficient or mean free path \( \ell \). Concurrently, the models presented in Sections 1.4.3-4 assume a quasiclassical description of conductance through a metallic film of thickness \( d \), provided \( \ell \leq d \), that is they work in the clean limit which allows quasiparticles to move along classical trajectories between the scatterings that lead to ABS formation. Let’s see how suitable is this description for our films. We can derive [92] an expression for \( \ell \) averaged over Fermi surface \( A_F \) from Pippard relations [132, 134]

\[
\sigma = \ell e^2 h^{-1} A_F / 12\pi^3, \quad \gamma = k_B^2 h^{-1} A_F / 12\pi v_F
\]

which combined give

\[
\ell v_F = (\pi k_B/e)^2 \left(\sigma/\gamma\right), \quad (2.3)
\]

where, in our case, iron film conductance \( \sigma \approx 4 \times 10^3 \) S/m\(^2\), electronic specific heat \( \gamma = 5 \times 10^{-3} \) J/K\(^2\) and Fermi velocity \( v_F = 1.98 \times 10^6 \) m/s. Therefore the average mean free path in our iron layers is

\[
\ell \approx 100 \ \text{Å}, \quad (2.4)
\]
such that the transport description based on quasiclassical models should be valid at least for the first three thicknesses. For the next two thicknesses the main inadvertency may appear in the assumed material dependencies of coherence length which is quite irrelevant since we extract our coherence length value from the period of the oscillatory behavior collected on the first three iron layers and the tunneling spectral main features should be qualitatively the same.

As indicated in the previous sections, besides iron, we also evaporated silver and lead thin films in Ag/BSCCO and Pb/BSCCO stand alone junctions. However, in these cases, the precise thickness of the film was irrelevant, although we tried to keep the same geometrical details throughout the experiments in order to enhance the reproducibility of the comparable spectral features.
CHAPTER 3
Tunneling measurements and spin polarization effects

As we saw in Section 1.4, tunneling spectroscopy on superconductor junctions has a long history of efficiency and, in the case of high temperature superconductor structures, it provided one of the most powerful instruments to probe their unconventional electronic properties. In this chapter, we investigate the ABS spectral features in their simplest form (N/BSCCO), subject only to spontaneous BTRS influence, and in perturbed conditions: either by an extrinsic $s$-wave pairing (S/BSCCO) or by spin polarization (F/BSSCO). Usually, the features exhibited by tunneling spectra on cuprates are especially rich and, due to the sensitivity of high-$T_c$ superconductivity on impurities and doping, may show spurious details and slightly differ even for spectra collected on the same junction. However, some characteristics connected to the nonlocal electronic properties should be common and interpreted as relevant. Consistently, in the case of our junctions, we present below only the features persistent and reproducible. We measured about 50 such junctions and each one of them belongs to the reproducible types discussed here.

3.1 N/BSCCO junctions

In this section we present our measurements on N/BSCCO junctions in order to observe and understand the complex behavior of ABS in a nonmagnetic environment. The normal counterelectrode is Ag and the data was collected on hybrid structures as described in the previous chapter.

3.1.1 Results and discussions

Figure 3.1 shows two sets of typical DOS curves obtained for two different current injection angles from Ag deposited on the same BSCCO crystal with $T_c \approx 85$ K mounted as in Fig.2.1a. Let’s name the respective junctions A and B, as in the figure. Ideally, the angle difference should be $\pi/4$, such that the phase difference of the presumably $d$-wave order parameter probed by tunneling to be maximum.
Figure 3.1 Temperature dependent tunneling spectra obtained on Ag/BSCCO planar junctions A and B built on the same crystal at two different current injection angles. The lower planes are contour projections. a) In anti-nodal tunneling directions, the zero energy ABS is less probable due to the locally isotropic order parameter which leads to an unfavorable phase difference. b) In nodal direction, the gap is filled with ABS and a high amplitude ZBCP indicates the presence of zero energy ABS favored by the phase difference introduced by the $± d$-wave lobes.

However, most of our data, corroborated with the respective BTK extrapolations, fitted better a smaller angle difference, either due to the polishing procedure which, being done along the crystal sides, wears out more material close from the corner or due to the intrinsic
characteristics of the net tunneling current through planar junctions as formed as an angle average, with smaller or higher angle contributions being determined by interface roughness.

The superconductive structures are sometimes superimposed over a parabolic background conductance (Fig.3.2a) which could be associated either with an intrinsic effect leading to a more extreme V-shaped background [135] or with a normal junction dependency of barrier strength on the bias voltage [136]. The possible intrinsic effect assumes a breakdown of Fermi liquid theory due to the strong quasiparticle correlations enhanced by disorder which modifies the tunneling DOS by the singular correction $\delta N/N_0 \propto \ln(E)$ for two-dimensional systems. Consequently, the background depends linearly on the energy at high bias as in

$$N(E) = N_F(\alpha|E| + 1)$$

where the coefficient $\alpha$ for negative and positive $E$ differ by $\approx 10\%$, detail which may explain the sometimes asymmetric backgrounds. The temperature dependency of the DOS correction was one of the arguments used to explain the thermal broadening of spectral features [137]. However, the extrapolation of this model to cuprates proved problematic due to the lack of evidence concerning a $\ln(V)$ DOS dependence or a regular correlation between the background slope and disorder.

On the other hand, the alternative extrinsic explanation is based on the increasing tunneling probability due to the barrier distortion under increasing bias voltage. The current (1.105) can be adapted to include this effect and, with a proper choice of work functions, the idea can be applied to superconductive junctions.

In particular, when the barrier effect is considered, the resulted theoretical gap profiles reproduce the asymmetry of coherence peaks – a feature rather common in our spectra. However, the asymmetric coherence peaks can be explained alternatively by an anisotropic electronic structure (possibility ignored by most models), that is an imbalance between the number of filled states (quasielectrons) and empty states (quasiholes) leading to a violation of band half-filling and to the asymmetry of the conductance spectrum [138].

Subsequently, Kirtley and Scalapino [139] developed a model of inelastic tunneling into a two-dimensional strongly correlated system of uniformly distributed excitations $F(\omega) = \text{const.}$, accounting for a V-shaped background, $N(V) \propto F(\omega)eV/h$. This model, when extended to
cuprates, speculates that the fluctuations in the antiferromagnetic order at lower doping play a role in the linear background and hence this behavior should exclusively characterize underdoped high-$T_c$ superconductors or degraded crystal surfaces.

The superconductive features are superimposed over this parabolic or V-shaped background. Before discussing them, we notice that the sharp features assumable for an ideal junction are in real measurements smeared or even washed out by the redistribution of tunneling probability weights through different mechanisms. One source of distortion is the gap inhomogeneity ($r$-dependency) with a significant broadening effect when the coherence length is smaller than the junction surface. For instance, in cuprates, the gap can become inhomogeneous due to the proximity effect between the CuO$_2$ and BiO planes, as we pointed out in Section 1.1.3 and Fig.1.5. Hence, due to the short in-plane coherence length of the pairs in these materials (see Table 1.2), tunneling in planar junctions will likely result in broadened coherence peaks flanking the gap unlike the sharp peaks seen in STM measurements. Also, because of the gap anisotropy ($k$-dependency), the tunneling cone will be very flat and the spectra will be in fact averages over angle distribution of quasiparticle trajectories probing various local gaps. In this case, the morphology of junction interface plays a role as well. Another smearing factor is the Dynes’s
influence of quasiparticle lifetime, that is the nonequilibrium effects associated with the time $t$ spent by two quasiparticles before recombining into the superconductive condensate [140]. At temperatures much lower then $T_c$ the recombination rate is approximated by

$$\tau = \tau_0 T^{-1/2} e^{-\Delta/k_b T},$$

(0.2)

where $\tau_0$ is a parameter related to the electron-phonon strength. Therefore, this finite-lifetime effects lead to thermal broadening of features which can be modeled using a pair breaking *smearing factor* with dimension of energy, $\Gamma = \hbar/\tau$. For BSCCO crystals it was shown [141] that this factor depends on the temperature as

$$\Gamma(T) = 2.3 k_b T_c^{-2} T^{1/3}.$$  

(0.3)

Related to ABS, it was shown [142] that, if they are localized near a small transmission barrier, they become broadened due to the finite quasiparticle lifetime for the quasistationary bound state. Consequently, the smearing interferences are rather complex, but a more systematic consideration of their action is beyond the scope of this thesis, so that, in our theoretical extrapolations, we mainly emulate them collectively by convoluting the spectra with gaussian distributions of width $\Gamma$ which leads to a broadening slightly larger than the Dynes’s lifetime-broadening.

We see that the junction near the $(100)$ crystal surface probes a superconductive peak-to-peak evaluated gap $\Delta_d \approx 44$ meV at $T = 4.2$ K. This value is a signature of strong in-plane coupling inasmuch as the ratio $2\Delta_d/k_b T_c \approx 12$ is much larger than the weak coupling BCS value of 3.54. The experimental gap still retain a small ZBCP due to the contribution from higher angle tunneling directions probing small amplitude midgap ABS. Unlike the typically U-shaped conventional superconductor gaps, the high temperature gap presented here has a characteristic V-shaped profile unrelated to the shape of the background (Fig.3.1a and Fig.3.2). This gap profile has been consistently reported in literature [108, 137, 143]. With few irrelevant exception, all our measurements exhibiting a gap devoid of a significant zero energy ABS contribution show this V-shaped gap meaning that there is a linear dependency between the net subgap DOS and energy. As implied by the modified BTK model (Section 1.4.3), in the case of anisotropic order parameter symmetry this can be attributed to a DOS isotropically averaged over the Fermi surface (i.e., isotropic tunneling matrix) since in this situation the quasiparticle
excitations are possible at any finite energies resulting in a linear dependency $\sigma = \sigma(V)$ close from Fermi energy (that is the middle of the spectrum) due to the proportionality of quasiparticle scattering rate to the contributions of the states away from $k_F$. There are also indications that the V-shape may be connected to the surface morphology since a disordered surface may affect the conservation of the transverse component of the momentum (diffusion transport) which smears the $k$-direction dependency of tunneling probabilities by allowing equal weights for all trajectories as in the BTK model.

Another typical feature is a low bias subgap structure resembling a smaller gap of $\approx 35$ meV, a *minigap* marked with an arrow in Fig.3.1a and with arrow A in Fig.3.2a. The respective change of slope is also visible on the contour projection and in Fig.3.4a where it coincides with the $\pm 25$ meV position of the two more conspicuous *subgap peak features* on the sides of the ZBCP. These features – marked with arrows in Fig.3.4a – will be discussed in more detail further below and are not necessarily related to the minigap. In their most typical form, they are actually double-bump features, like smeared combination of two harmonics of the same resonance. It is important to mention that, in N/BSCCO junctions, we have rarely seen the subgap peaks without the ZBCP. Moreover, sometimes an extra pair of peaks appears (see Fig.3.3b) as the initial pair is shifted at higher energies with decreasing temperature (looking like a gap opens between them) while being replaced by new smaller bumps surging out from the slimmer but increasingly taller ZBCP. In the case of anti-nodal tunneling, these peaks are usually replaced by more subtle changes of $\sigma - V$ slope or by narrow steps. However, most of the time, we observed only a pair of peaks and only a minigap.

The origin of the minigap is not very clear. As example, if we recall from Section 1.4.4 that, from a quasiclassical perspective, the subgap DOS can be basically regarded as a distribution of ABS attached to the geometry dependent trajectory distribution [123], the disorder in the material and thin film thickness $d$ may provide an explanation. Thus, the length distribution of quasiparticle trajectories in the counterelectrode decays as $\exp(-l/\bar{T})$ (Fig.1.13), with the average length $\bar{T}$ depending on the mean free path and film thickness through $\ell/d$. Consequently, a degree of disorder sets an upper limit to the average length by cutting off the long trajectories, such that the spectrum is dominated by the short trajectories associated with higher energy ABS, while the lower energy ABS are suppressed below an energy threshold.
However, this scenario cannot explain the apparent independence of the minigap on the thickness and the presence in point contact and even break junction spectra. Also, this model disregards the dependence of ABS on order parameter phase such that it is not flexible enough to explicate why this feature is missing in tunneling experiments performed on conventional superconductors. Therefore, the explanation should be rather intrinsic and not related to the geometrical details.

Another speculation [48], tant mieux an intrinsic one, places the origin of the in-gap structures in the context of the theory of two energy-scales mainly in underdoped cuprates: $\Delta_c$ connected with the phase coherence of the condensate and a higher $\Delta_p$ related to pairing. Since the minigap has a temperature dependence similar to $\Delta_c$, it could be a (perhaps $g$-wave) subdominant harmonic, magnetic in nature, of the mechanism responsible for the onset of pair coherence based on spin fluctuations. In this case, the minigap should be half $\Delta_c$ and the respective higher peaks wouldn’t be necessarily related to the minigap itself but to the onset of two dips (at arbitrary positions) due to the interaction between normal electrons and the antiferromagnetic stripes in the cuprate. While this interpretation has its merits (e.g., it does explain the temperature evolution parallel with the $d$-wave gap), it cannot account for various positions of the minigap edge (for instance, in Fig.3.1a, $\Delta_p$ is less then twice the minigap) and neither does it account for the correlation which seems to exist between the subgap peaks and the zero energy ABS as described above.

The onset of minigap was also attributed [131] to the doping dependent onset of BTRS pairing due to an additional $s$ (or $d_{xy}$) subdominant pairing interaction with its own critical temperature, thus having the same nature with the splitting of ZBCP discussed in the next section. However, this hypothesis sets the quantum phase transition from pure $d$-wave to BTRS state in the vicinity of optimal doping so that the minigap shouldn’t be visible in the underdoped region. On the contrary, we do see the minigap (as well as the ZBCP splitting) in this region.

The inhomogeneity and the anisotropy of the gap can also determine a shape of the antinodal gap similar with our experimental spectra. Thus, as we discussed in Section 1.1.3, the superconductivity can leak from the CuO$_2$ planes into the BiO planes and this phenomenon can show up as a supplementary smaller gap even in the spectra collected in $ab$-planes, since some $c$-axis transport will inevitably occur in real conditions. In fact, any $c$-axis superconductivity should have the same effect. It was shown [144] that, if the gap anisotropy in the $ab$-plane takes
the \(d\)-wave form (1.77) and there is a \(k_z\)-dependent \(c\)-axis gap, one can write in terms of the interlayer distance \(c\) and the lattice parameter \(a\):

\[
\Delta(k_z) = \Delta_z \cos(k_z c),
\]

\[
\Delta(k_x, k_y) = \Delta_d \left[ \cos(k_x a) - \cos(k_y a) \right] \tag{0.5}
\]

and the \(ab\)-plane spectra should exhibit an intermediate change of slope due to the periodical change of Fermi surface between \(k_z c = 0\) and \(\pi\) (see Fig.3.2b). Specifically, this is a direct result of the van Hove singularities [145] near the Fermi energy arising in point 2 in Fig.3.2b, i.e., the saddle points corresponding to the energy spectrum of a single orbital tight binding Hamiltonian on a square lattice. This is a strong, structurally justified argument which explains \textit{inter alia} why a feature otherwise solid in cuprates does not appear on the DOS spectra of isotropic pairing superconductors. Nevertheless, it fails to account for the subgap finite energy peaks which need a separate discussion as following.

Usually, these side peaks appear at \(\approx \pm 10\) meV, at the lowest temperature attainable by our experimental arrangement, \(T = 4.2\) K. They flank a very sharp ZBCP. However, as we pointed out above, their energy locations move apart with decreasing temperature. Moreover, the width of the ZBCP may vary from junction to junction depending on factors such as the barrier strength, the current injection orientation, the degree of transport diffusiveness etc. Therefore, when the ZBCP is wide enough, it may even cover the “first order” side peaks such that the subgap DOS gets cleaner if it is not creased by higher energy subgap peaks as the case in Fig.3.1b where the subgap peaks occur at \(\approx \pm 25\) meV. In fact, the peaks at \(\pm 10\) meV may not be the lowest order peaks in \(ab\)-plane tunneling experiments: the fact that they are the first discernable peaks may be just a signature of the minimum strength of the ZBCP broadening influences acting inside our junctions. This is consistent with the fact that the ZBCP seem to have itself a structure, with small shoulders.

The subject of the diverse subgap unexpected features was tackled following the initial release of the BTK model, before the discovery of high temperature superconductivity. Thus, the “\textit{subharmonic gap structures}” [146] appearing in metallic weak links and low barrier tunnel junctions were explained in terms of multiple Andreev reflections. The subharmonic structures refer to a series of conductance dips located at periodic intervals which may originate in the band structure of the electronic distribution function with a periodicity determined by maxima in the
electronic transfer intermediated by multiple Andreev reflections and enhanced by normal scattering. The structures appear at energy intervals $2\Delta/n$, $n$ integer, where maxima with number of reflections occur. This is a natural scenario for low transparency junctions, including possible shorts across the barrier. The occurrence of the subgap peaks at sometimes periodic energy intervals pleads for such an explanation, even if, most of the times, we recorded only a pair of peaks located on the sides of the ZBCP (the rest of the gap being smooth) which suggests a correlation with the zero energy ABS.

Alternatively, the subgap features can be discussed in terms of non-zero energy ABS [142]. In Fig.3.3b we tentatively modeled the conductance spectra at $T = 6$ K using the anisotropic BTK (see Appendix A for the computer subroutines). Remarkably, this formula can emulate the side peak feature as two cusplike local maxima located at the bias voltages corresponding to the energy gap of the state probed by momenta perpendicular onto the junction interface (see Fig.1.12b and Fig.3.3c as well). More precisely, while the zero energy ABS (at the origin of the ZBCP) are allowed for all transversal momenta $k_y$ when the junction orientation is $\beta = \pi/4$, for less tilted interfaces only the ABS corresponding to $k_y \sin(\pi/4 - \beta) < k_y < k_y \sin(\pi/4 + \beta)$ can contribute to the ZBCP (see Section 1.4.2). The cusps appear for $k_y = 0$ at energies

$$E = \Delta(k_y = 0) = \pm \Delta_d \cos(2\beta),$$

and, close to nodal junction orientation ($\beta = \pi/4$), they join the ZBCP.

On the other hand, besides the contribution to ZBCP, the other allowed ABS are also responsible for the onset of two smooth, symmetric shoulders at maximum gap, that is the nominal $d$-wave gap $\Delta_d$, which develop into sharp coherence peaks as the cusplike features shift toward higher energies near anti-nodal junction orientation (where the ZBCP disappears). Therefore, the subgap peaks we observed in our experiments would be conveniently explained by this scenario: they are non-zero energy ABS defined by (0.6) – a correlation with lower or higher superconductive gaps $\Delta_d$ hence following the same temperature evolution. Note the good match between the energy location predicted by this model and the subgap peaks in our spectra: using the fitted values $\Delta_d = 44$ meV and $\beta = 30^\circ$, the calculated position is $\approx 26.5$ meV, in excellent agreement with the position of our experimental peaks.
Figure 3.3 a) Ag/BSCCO tunneling spectra collected for two different junction orientations on the same crystal (same with $T=6 \text{ K}$ spectra in Fig.3.1). b) The corresponding anisotropic BTK extrapolations in unpolarized conditions, $U = 0$, $h = 0$, with parameters $\Delta_d = 44 \text{ meV}$, $\Delta_s = 2 \text{ meV}$, $\beta = 7^\circ$, $Z = 3$, $\Gamma = 4 \text{ meV}$ ($\Gamma$ allocated by formula (3.3)) and $\Delta_d = 44 \text{ meV}$, $\Delta_s = 3 \text{ meV}$, $\beta = 30^\circ$, $Z = 5$, $\Gamma = 4 \text{ meV}$. c) Anisotropic BTK curves generated for fixed parameters except $\beta$ varying from 0 to $\pi/4$ in order to exemplify the distribution of subgap finite energy cusplike features.

Nevertheless, there still remains the question why sometimes we see more than one pair of such finite energy subgap peaks. A possible explanation is in the constructive specificity of our junctions and in general of the measurements performed along $ab$-plane using planar junctions. Thus, besides the interface roughness along the crystal, one may have a $c$-direction mismatch if the probed crystal contains several layers. In this case the resulting spectrum is in fact a sum of two or more slightly different curves and each finite energy experimental peak ultimately envelops several peaks. Consequently, the net peaks are broadened and, as we observed earlier,
they sometimes look split. Also, in extreme cases, the additional spectra would form completely separated peaks. For a graphic exemplification, in Fig.3.3c we generated the anisotropic BTK curves keeping fixed the parameters $\Delta_y = 40$, $\Delta_x = 0$, $Z = 5$ and $\Gamma = 2$ meV while varying the junction orientation $\beta$ from 0 to $\pi/4$ by small even steps. The subgap cusps (smeared by $\Gamma$) are indicated by arrows and, when contributed by slightly mismatched crystal layers, they may lead to a total spectrum exhibiting broadened and sometimes multiple subgap peaks.

3.1.2 Midgap ABS and BTRS splitting

As we discussed in Section 1.4.3, the onset of an isotropic subcomponent on top of the $d$-wave order parameter, leading to a broken time reversal symmetry, is a rather elusive and puzzling effect which complicates the already challenging question of pairing in high temperature superconductors. The presence of such a subdominant component should directly affect energy the distribution of ABS inasmuch as their existence is related to the phase of the order parameter and thus to its symmetry. Experimentally, this should manifest a splitting of the ZBCP due to the shift of zero energy ABS to finite energies – positive or negative for electronlike and holelike spectral branch respectively. We saw that such an effect is tantamount to a Zeeman splitting spontaneously induced by the paramagnetic response to the surface currents carried by ABS. We also noticed that it is not necessarily a magnetic effect, the splitting being explainable in the case of normal-cuprate hybrid structures by the proximity effect creating a thin $s$-wave region on the normal side. Then, the superconductive regions couple, reaching a minimum free energy when $\beta = \pi/4$, this minimum corresponding to a state with phase difference degeneracy which is lifted by the ABS shift to finite energies. Either way, the order parameter can be characterized by the nodeless $d + is$ admixture given by the formula (1.120), easy to integrate in the logic of anisotropic BTK formula based on angle resolved quasiclassical trajectories. Indeed, there are other types of symmetries breaking the time reversal, like the spin triplet $p_y$. However, this is improbable since it would split the ZBCP immediately under $T_c$. In our experiments, provided the magnetic field was zero, the ZBCP splitting always occurred at temperatures much lower than the BSCCO critical temperature.
Figure 3.4  a) Temperature dependence set of DOS spectra collected on an Ag/BSCCO junction showing BTRS induced splitting of ZBCP. The onset temperature of the respective subdominant s-wave component is \( \approx 20 \) K and its magnitude is 7.3\%. For a good resolution, only the central portion of the spectra is represented. b) The whole spectrum at 4.2 K, including the coherence peaks of the d-wave gap \( \Delta_d \approx 33 \) meV. Also, the spectrum at 100 K is depicted to show a pseudogap presence which places the sample in the underdoped phase.

Among our measurements performed on N/BSCCO junctions, there is certain diversity of results: some junctions, as the one presented in the preceding section, do not show a split ZBCP. Others show it. Concretely, in Fig.3.4a we plotted the temperature dependence of the DOS spectra obtained on such a junction, here named junction C. One can see that, at \( T_{cs} \approx 20 \) K, the ZBCP splits symmetrically. In Fig.3.5 the spectrum at 4.2 K is represented separately, with the corresponding BTK fit generated for \( \Delta_d = 33 \) meV, \( \Delta_s = 2.5 \) meV, \( \beta = 14^\circ \), \( Z = 2.5 \), \( \Gamma = 1.27 \) meV, \( U = 0 \), \( h = 0 \). Therefore, the s-wave subcomponent is about 7.3\%. Note that the BTK fit of the unsplit ZBCP (see Fig.3.3) also contains a nonzero \( \Delta_s \). However, the effect in that case is only a decreasing ZBCP amplitude. In fact, the splitting action of the s-wave subcomponent is just smeared out by the a \( \Gamma \) comparable or higher in magnitude than \( \Delta_s \), provided that the d-wave gap is large enough such that the ZBCP is relatively too sharp to accommodate the inner gaplike feature. A low energy resolution may also prevent the observation.
To quantify the sharpness of a ZBCP one can take advantage from the fact that the peak height is independent of the junction resistance and define a parameter $R$ equal to the ratio between the peak height (normalized by the background conductance) and the peak width at half maximum [133]. The ratio is to be calculated at temperatures before the onset of splitting influence. We calculated this parameter for the junctions B and C as following. Indeed, for junction B one can observe in Figs.3.1b (readable on the bottom contour) and Fig.3.6b (arrow) that the amplitude of the ZBCP registers a slight downturn at approximately 17.5 K which we interpret as the onset of the $s$-wave subcomponent leading to the BTRS state. Then, at 20 K the sharpness is given by $R \approx 2.8 \text{ meV}^{-1}$. On the other hand, as visible in Figs.3.4a and 3.6c, the midgap ABS of the DOS measured on junction C start to shift at about $T_{cs} \approx 20$ K and we calculated at 25 K a significantly smaller ratio $R \approx 0.75 \text{ meV}^{-1}$.

![Figure 3.5](image)

*Figure 3.5 Spectrum collected on the Ag/BSCCO junction (the 4.2 K curve in Fig.3.4a) where the BTRS induced splitting of ZBCP is obvious. The BTK fit (continuous line) ignores the peaks flanking the ZBCP and suggests $\Delta_d = 33 \text{ meV}, \Delta_s = 2.5 \text{ meV}$ (arrow). $\beta = 14^\circ, U = 0, h = 0, \Gamma = 1.27 \text{ meV}$.*

These values obey the rule found by our group [133] which in the past reported measurements on Pb/BSCCO junctions similar to the one we discuss in the next section: specifically, the ZBCP splitting always occurs if $R < 0.8 \text{ meV}^{-1}$ while the ZBCP stays unsplit but decreases in amplitude when $R > 1.1 \text{ meV}^{-1}$. This behavior is ultimately related to the effectiveness of our junctions via some smearing effects inherent to the respective structure due
Figure 3.6 Temperature dependence of the experimental conductance at different meaningful energies. a) At $E=0,\Delta_d$ for junction A. b) At $E=0,\Delta_d$ for junction B. Note the conductance downturn at $E=0$ in junction with the presumed onset of BTRS state. c) At energies $E=0,\pm\Delta_s$ for junction C. At about 20 K, the peak splits since the zero energy ABS shift to $\mp\Delta_s$.

to the fabrication limits. In other words, we believe that the splitting is still there, even though we see it indirectly, through the decreasing height of ZBCP.

Another interesting phenomenon is that the unsplit ZBCP, besides decreasing in amplitude under $T_c$, becomes broader and, in some cases, even develops prominent shoulders (see a good example discussed in the next section and illustrated in Fig.3.8a). This behavior becomes more suggestive if we trace back the evolution of ZBCP from immediately under$T_c$: thus, initially it is
just a very broad bump, partially due to the substantial thermal smearing, but also because it contains the precursors of the finite energy subgap peaks.

As the temperature decreases, these peaks shift at higher energies (parallel with the larger and larger $\Delta_d$ gap) while, in between, the ZBCP grows in amplitude and sharpens. Sometimes, for higher angle junction orientation, at lower energy a second pair of subgap peaks may leave the midgap complex. At lower temperatures, but above $T_{cs}$, the spectrum settles and the energy positions shift much slower. However, under $T_{cs}$, the curve starts again to show a more rapid evolution when ZBCP starts either to split or to diminish in amplitude, as described above. This is accompanied by broadening ZBCP similar in shape with the evolution under $T_c$ resulting in a peak with shoulders which may disconnect into separate peaks if the subdominant component is large enough (see next section). It looks like the junction goes through a second phase transition from a pure $d$-wave state to a mixed pairing phase. Note that this development is independent of the junction resistance such that, in Figs.3.1 and 3.4a, the evolution of subgap features is overposed but not related to a decrease in background conductance (also see Fig.3.6).

However, not all our N/BSCCO junctions show either the clear splitting or the decreasing peak. This kind of dichotomic behavior is widely reported in literature [65, 92, 108, 143, 147] and was explained mainly by the effect of surface roughness [148] and by doping dependence [131, 149, 150]. Moreover, it is possible for a large dominant order parameter to overwhelm the subdominant one such that, collaterally, the $s$-wave critical temperatures we found above by inspection may be underevaluated. Nevertheless, the fact that tunneling spectroscopy probes only the superficial DOS is for once an advantage: since the $d$-wave component is locally suppressed by perturbations like the vicinity of boundaries, whereas the $s$-wave component is inert with respect to inhomogeneities, the subdominant channel can manifest and be observed.

Knowing that the subdominant pairing interaction is likely a surface effect [149], the influence of surface roughness comes into play rather naturally. It can be modeled using a random scattering matrix where the roughness reduces the weight of specular reflections at the junction interface (conserved transversal momentum). The $d$-wave component, which is strongly suppressed at the boundary in the specular case, recovers when the surface is diffusive and inhibits the $s$-wave channel.
The onset of the BTRS state may be also doping dependent. This hypothesis is based on the existence close to the optimal doping of a critical doping associated with a quantum critical point (QCP) marking the transition from a pure $d_{x^2-y^2}$-wave superconducting state to a BTRS state. Hence, the $s$ (or $d_{xy}$) subcomponent would not be necessarily a surface effect but an instability of the $d$ phase in the overdoped region as opposed to the stability of the mixed state [150]. Accordingly, features like the minigap and the ZBCP splitting should appear and behave the same over the respective threshold doping located at high doping level. Although this scenario is sustained by some reported experimental studies [131] it seems to be contradicted by others [151].

Apparently, our results also do not seem to confirm the predicted doping dependence with a QCP in the middle of the superconductive phase. To show this, we represented in Fig.3.4b the tunnel spectrum collected on junction C at 4.2 K and 100 K, well above the $d$-wave critical temperature, where one can see that the “normal” spectrum presents the features of a pseudogap as does most of our data (see Section 2.2). Therefore, the sample is most probably undoped which, at least guesso modo, contravenes the doping ranges allowed by the theory for the ZBCP splitting. Indeed, as described several paragraphs above, the systems looks like going through a phase transition at $T_{cs}$ but, in our case, the respective QSP seems to placed at lower dopings. Certainly, the reason for this incongruency may hide behind the uncontrollable details of our junctions, mostly considering that one can detect at least an indirect connection between the doping and the subdominant component through the amplitude of the dominant one. For instance, the oxygen content and hence the doping level may be nonuniform at the junction surface. This is consistent with the reported inherent, disorder independent inhomogeneity of electronic phase at the surface of BSCCO measured using STM technique [152].

### 3.2 S/BSCCO junctions

Beside being important per se, investigating the electronic structure of cuprates using tunneling spectroscopy with conventional superconductor counterelectrodes can be especially helpful in understanding the ABS dynamics in the presence of mixed pairing potentials. Consequently, we studied Pb/BSCCO planar junctions with cuprate $ab$-plane injection, as
Figure 3.7 Temperature dependence of DOS spectra collected on two different Pb/BSCCO tunneling junctions (E and D) showing apparently discrepant behavior when Pb becomes superconductive: a) ZBCP split by a gap-like crevice equal to Pb gap $\Delta_{Pb} \approx 1.2$ meV and b) decreasing, broadening ZBCP.

described in Section 2.1. Pb is a conventional superconductor with bulk $T_c^{Pb} \approx 7.23$ K and makes excellent thin films.

Most of the studies performed on Pb/cuprate heterostructures probe the c-axis direction properties, mainly in relation to the occurrence of a Josephson coupling across the weak link
between in the superconductive electrodes [153]. Our junctions are built with the Pb thin films perpendicular onto the BSCCO $ab$-plane and, even if some of them presented features typical to Josephson coupling (e.g., extremely sharp and narrow ZBCP due to the surge of supercurrent), we discuss in this thesis only the uncoupled ones.

Between underdoped BSCCO $T_c$ and $T_c^{Pb}$ the temperature dependence of tunneling spectra is similar to the one measured on N/BSCCO junctions: a generally clear pseudogap above $T_c$ followed by the gradual amplitude increment of the midgap ABS on the bottom of the dipping $\Delta_d$ gap, the corresponding ZBCP being usually flanked by the subgap peaks discussed above, even if somewhat flatter than in the Ag/BSCCO case. In general, the spectra in this range are good candidates for anisotropic BTK extrapolation (see Fig.3.8c,d).

Under $T_c^{Pb} \approx 7.3$ K (value close from the generally accepted value [153]), the spectral behavior is an enhanced version of the N/BSCCO case under $T_{cs}$, even if the reason behind the changes may be different in nature. To illustrate this, we represented in Fig.3.7 the temperature evolution of energy resolved DOS spectra measured on two different Pb/BSCCO junctions (named E and D) showing more clearly the discrepant reaction to an additional $s$-wave potential observed on Ag/BSCCO junctions. On one hand, in Fig.3.7a, under, the top of the ZBCP folds inside and evolves rapidly into a gaplike crevice. On the other hand, the spectra in Fig.3.7b show a ZBCP perturbed but not split under $T_c^{Pb}$: it only decreases in amplitude and broadens developing shoulders reminding of the subgap peaks which grow under $d$-wave $T_c$ on the sides of ZBCP.

To categorize the ZBCP sharpness above the $s$-wave critical temperature, we can apply the same $R$-parameter quantitative approach as in the N/BSCCO case. Then, we get $R \approx 0.7$ for junction D and $R \approx 1$ for junction E which barely respects the rule outlined in the previous section. We see again that wide and sharp ZBCP tends to decrease in amplitude, washing out the effect of the $s$-wave component. Again we underline the fact that this is just a measure of the our junction resolution: the fact that we do not see the Pb gap directly in some cases (while there is no reason for a broader and sharper ZBCP to affect the confluence of the two DOS distributions) suggests that the BTRS can be also observed indirectly.
Figure 3.8  a) Tunneling conductance spectra of two Pb/BSCCO junctions at 5 K (shifted vertically for clarity). The upper curve is the result of a ZBCP decreasing and broadening under $T_{c}^{\text{Pb}} \approx 7.3$ K. The lower one is a splitting ZBCP. The split is enlarged in b) where the Pb gap and a resilient small ZBCP are visible. c) Junction D before splitting. The BTK fitting yields $\Delta_d = 21 \text{ meV}$, $\Delta_s = 0$, $\beta = 14^\circ$, $U = 0$, $h = 0$, $Z = 5$, $\Gamma = 0.8 \text{ meV}$. d) Junction E before ZBCP decrement. The fit corresponds to $\Delta_d = 41 \text{ meV}$, $\Delta_s = 0$, $\beta = 23.5^\circ$, $U = 0$, $h = 0$, $Z = 5$, $\Gamma = 1.2 \text{ meV}$.

Nevertheless, the similarities between the low temperature behavior of N/BSCCO and S/BSCCO junctions can be deceiving since the respective behaviors differs in their details. Recalling Section 1.4.3, we notice that the onset of BTRS state can be explained by the proximity effect creating on the normal side a superconducting region coupled with the original $d$-wave. In this case, the lead $s$-wave superconductivity would play the same role, leading to the same effect via the same mechanism. However, a simple observation refutes this argument. Thus, in Fig.3.7b we blew out the split upper side of ZBCP at about 5 K and one can see that a small ZBCP survives on the bottom of the gap-like crevice. We conclude that this split is an additional gap rather that the result of a zero energy ABS shift. Of course, we cannot eliminate
the possibility that the Pb layer has itself an effect on the BSCCO side. It is conceivable that the s-wave Pb superconductivity may induce by proximity a still anisotropic but nodeless state at the cuprate boundary where, mostly in clean junctions, the dominant $d$-wave component degrades due to inhomogeneity. Additionally, the BTRS state can still be induced through the other mechanisms, provoking a slight misalignment of the peaks resulted from the Pb contribution (visible at the edge of Pb gap in Figs.3.6a and 3.7b) as well as a complete shift of zero energy ABS leaving the bottom of the Pb gap without a ZBCP at lower temperatures (this is also detectable in Fig.3.6a where the small ZBCP disappears close to 4.2 K). This would be an example where the BTRS state is observable not in the splitting of the ZBCP but of the finite energy peaks resulted after the ZBCP was split by a different influence – in this case the presence of Pb.

### 3.3 F/BSCCO junctions

Even if spin polarized transport through tunnel junctions raised interest ever since the infancy of this technique [113], the subject gained a new impulse with the discovery of high temperature superconductivity – a phase intimately related to spin ordering and interacting with magnetism in a more subtle way than conventional superconductors. A ferromagnet put in contact with a non-magnetic metal may transpire the exchange interaction into the normal metal determining a nonequilibrium spin accumulation. Similarly, if the metal is a superconductor, the properties and availability of quasiparticle transport channels are fundamentally affected due to magnetic pair breaking. In particular, the density mismatch between carriers of different spin polarization lead to a suppression of the low energy channel intermediated by Andreev reflection and hence to an exotic spin dependence of ABS energy distribution near interfaces with anisotropic order parameter.

In this section we report our own spin polarized measurements performed on Fe/Ag/BSCCO tunneling junctions built as described in Chapter 2. In the first part we briefly address the spin filtering properties occurring in the ferromagnetic/superconductor barrier. In the second part, we discuss some interesting phenomena related to the ABS energy shift determined by the barrier spin filtering acting in parallel with the BTRS state induced splitting. The spin polarized transport across the tunneling junctions is an excellent test for the superconductive transmission channels sensitive with respect to the spin imbalance, like Andreev reflection.
3.3.1 Pair breaking barrier resistance

The subgap transport in superconductors is mediated by spinless pairs. On the other hand, in the ferromagnets, the current has a finite spin component. Ergo, when a tunneling junction combines a superconductor with a ferromagnet, a nonequilibrium spin density accumulates at the junction barrier in order to conserve the spin currents, phenomenon which can act as a spin filtering, pair breaking layer onto the tunnel current [154, 155]. This spin filtering can be viewed as the discriminating effect of a ferromagnetic exchange potential in the insulating interface: as depicted in Fig.3.8a (inspired by [154]) the current with spin up and down will feel a different barrier height. As far as the tunneling characteristics depend on the barrier height, the tunneling spectra will be radically modified, as we shall see further below. These modifications have been observed even without a specially grown ferromagnetic insulator [86, 156, 157] and were proven to be of magnetic origin: the polarized layer seems to appears naturally at the interface in the presence of a ferromagnetic counterelectrode, but the exact mechanism is not clear.

![Diagram of barrier resistance control by Andreev reflection and spin accumulation](image)

**Figure 3.9** a) Temperature dependence of normalized barrier resistance controlled by the competition between Andreev reflection and spin polarized accumulation. b) Schematic representation of a spin polarized junction interface leading to spin channel discrimination.

Another aspect which indicates a barrier polarization is the variation with temperature of the barrier resistance (Fig.3.8b). The resistance decreases or increases with decreasing temperature as a result of a competition between the Andreev reflection and the screening spin accumulation close to the interface [155, 158]. Being a pair transmission channel, the Andreev reflection tends to decrease the resistance, contrary to the pair breaking spin accumulation with an adverse effect.
The interplay between these two factors shapes up the transport properties of the barrier with a different profile in the specular and the diffusive regimes. In the case of a diffusive junction, the resistance first decreases and then increases to reach a plateau of equilibrium between the two contributions. In fact, this typical behavior was used in the last years in conjunction with the BTK model, as a remarkably precise technique to measure the spin polarization in metallic mesoscopic layers [120]. As visible in Fig.3.8b, our junction follows this directive, proving the existence of spin accumulation at the junction barrier.

### 3.3.2 Spin polarization effects on midgap ABS

One may have notice that in all our previous BTK fittings we carried with us the two exchange energy terms $U$ and $h$, even if they were zero. The scope was to underline in the spin independence of the measured DOS when the counterelectrode is a normal metal or conventional superconductor, as opposed to the F/BSCCO case presented in next few paragraphs. In other words, the charge contributions $\sigma_{q\uparrow\downarrow}$ are balanced. As seen in Section 1.4.4, the parameter $U$ models the barrier spin-filtering we discussed in the previous section, while $h$ is the exchange energy in the ferromagnetic counterelectrode – iron in our case. When these quantities are non-zero, the net charge current will be the spin resolved sum with unbalanced contributions from $\sigma_{q\uparrow\downarrow}$, weighted by the respective spin polarization $P_{\uparrow\downarrow}$ given by (1.126).

Our ferromagnetic junction, in the Fe/Ag/BSCCO arrangement, contains a 30 Å thin Ag layer which plays a rather passive role in our measurements: as discussed in Section 1.1.3, it prevents the onset of a spin glass disorder at the boundary between the ferromagnet and antiferromagnetically ordered stripes in the cuprate. The ultrathin iron layer has a thickness of $\approx 60$ Å where we expect to see a maximum of ABS amplitude, based on the space dependence of LOFF order parameter in the proximity region (see [92] and Chapter 4).

As usually, we plotted in Fig.3.10a the temperature dependence of the conductance spectra between about 80 K and 4.2 K collected on a junction we named here Junction F. The first detail one should notice is the huge peaks evolving almost equal in amplitude down to $\approx 32$ K, but asymmetric from that point, as visible in Fig.3.9a. The gap-like crevice in between the peaks does not contain a ZBCP, even if traces of a minigap are detectable (more clearly visible in Fig.3.10).
Figure 3.10  a) Temperature dependence of the conductance spectra of a Fe/Ag/BSSCO tunneling junction. The ZBCP is split by a very large gap-like structure due to the spin-filtering effect of the polarized barrier. b) Temperature dependence of the DOS at several meaningful energies: note that the ZBCP splitting occurs already at $T_c$ and the resulted peaks become asymmetric at $\approx 32$ K, where presumable the spins flip in the Fe layer.

We attribute the occurrence of this relatively large gap-like feature to the confluent action of two ZBCP splitting factors. On one hand, we have a spin independent effect due the existence of a subdominant component mixing with the $d$-wave state to create a BTRS state and, on the other hand, a Zeeman splitting due to the spin-filtering action of the polarized barrier which is capable to shift the zero energy ABS much more vigorously than the BTRS, such that the corresponding ZBCP splitting can invade the whole subgap space. Of course, in conformity with the definition (1.123), the shift is also dependent on the “normal” strength of the barrier, $V$, in the sense that, at very high $V$ (tunneling limit), the spin splitting doesn’t hold. The exchange energy $U$ in the interface filters the spins by lowering the barrier strength for spin up current and raising it for the spin down, such that, depending on the spin of the injected current originating either from the up or down spin bands, this shift is in fact a gain or respectively loss of energy suffered by the
midgap ABS during the tunneling process. The large splitting occurs already near $T_c$ (see the inset in Fig.3.10) and, in conjunction with the magnetic dependence we discuss further below, this excludes the possibility of an origin based on a BTRS order parameter, since the large magnitude of the subdominant component (large splitting) would be difficult to explain.

Figure 3.11 Experimental spectrum at 4.2 K (dots) and the corresponding anisotropic BTK fit (smooth lines) yielding $\Delta_d = 45$ meV, $\Delta_s = 7.2$ meV, $\beta = 45^\circ$, $U = 2.7$ (i.e., a difference of 5.4 between the barrier felt by spin up and down currents), $h = 0.086$, $\Gamma = 1.27$ meV. For distinctiveness, the fit includes besides the net current the spin up and down components shifted by the barrier spin-filtering. The spontaneous magnetization in iron determines the peak asymmetry. The spectra in the inset are taken at high and intermediate temperatures to show that the splitting appears near $T_c$ and the peaks are still symmetrical above $\approx 32$ K, as explained in the text.
The combined effect of these two perturbations is a four-peak structure represented in Fig.3.10 and marked with arrows A, B, C, D. The B-C splitting originates in the spin-filtering and the more modest A-B, C-D splitting appear due to the BTRS effect. A rather suggestive detail is the broadening of peaks with decreasing temperature attributable to the dependency of the spin-filtering on the quasiparticle trajectories, with the trajectories perpendicular on the interface experiencing the maximum splitting. Such a $k$-dependence was explained [157] by the exponential fall-off of the tunneling matrix elements with the thickness of the junction barrier.

A strong argument pleading for spin dependent origin of the large splitting is the specific dependence on magnetic field. Thus, as one can follow on Fig.3.9b, while at higher temperature the peaks are almost equal (with the quasielectron branch slightly higher as in most of our N/BSSCO junctions – see Section 3.1), at about 32 K the peaks become asymmetric with a higher and higher peak on the quasihole side and lower and lower peak on the other branch. It looks like the weight of carrier distribution is influenced by an external magnetic field. In order to explain this, let’s recall from Section 1.2 that iron satisfies the Stoner criterion of spontaneous magnetization. Therefore, we attribute this behavior to the ferromagnetism in the Fe layer creating a magnetic field.

The low temperature manifestation of this influence can be explained by a change of ferromagnetic phase – one of the phenomena which makes ferromagnetism a still surprising field – experienced by magnetization direction in ultrathin films. Thus, it was shown [79] that it can flip direction at critical temperatures much lower than Curie temperature, going through a reorientation transition from in-plane to perpendicular direction due to the temperature dependence of perpendicular anisotropy of thin films. Consequently, at high temperature the magnetization is mainly parallel with the junction interface such that the peaks show only the small asymmetry also detectable in normal junctions. Under a critical temperature the spins gradually switch to a direction perpendicular on the barrier, in the direction of spin injection, so that the DOS peaks start to become more and more asymmetric in the other direction. Remarkably, the peak ratio is proportional to the polarization of the iron film [159] and, in our case, we estimated at 4.2 K a value $P \approx 35\%$.

We used the anisotropic BTK model adapted to ferromagnetic environments (as discussed in Section 1.4.4) to evaluate the various parameters. The peak height is consistent with the nodal injection given by $\beta = 45^\circ$. The lateral shoulders on the peaks are interpreted as $\Gamma$-smeared
traces of the BTRS splitting due to a mixed order parameter with $\approx 6\%$ $s$-wave contribution to the predominant $d$-wave. BTK nicely emulates these shoulders and we see that the $s$-wave splitting is $\Delta_s = 7.2$ meV, significantly smaller than the spin splitting gap evaluated by peak-to-peak inspection, $\Delta_{BC} \approx 27$ meV. For a clearer image, we plotted the charge current contributions separately for spin up and down, so one can follow how the peak (split by BTRS) corresponding to $\sigma_{q\uparrow}$ is shifted to lower energy while the $\sigma_{q\downarrow}$ one moves to the right, gaining energy.

It was shown [160] that, since ABS form for all quasiparticle trajectories, at low temperatures their sensitivity to the exchange potential in the ferromagnetic counterlectrode becomes independent of the strength of the junction barrier. Therefore, the zero energy conductance is a direct measure of the exchange energy and its temperature dependence takes forms specific to the pairing symmetry of the superconductor. However, in our case (see Fig3.9b), the behavior expected for a $d_{x^2-y^2}$ pairing at zero energy (i.e. moderate growth with decreasing temperature) is shifted to negative finite energy, due to the splitting. A similar growth is expected from a $p_y$-wave symmetry but this is excluded by the reaction of the peaks to magnetic influences. Thus, the iron magnetization exaggerate the growth of the negative energy peak and distorts the growth of the positive energy peak.

### 3.4 Conclusions

We built a series of planar tunneling junctions in order to take advantage from the ability of this technique to probe the density of states and hence the properties of the superconductive order parameter – a quantum construct completely describing the state along its phase evolution and critical reactions against external perturbations. In high temperature superconductors, like cuprate perovskites, the details of this complex quantity are still incompletely understood.

Our junctions combined such a cuprate material, $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_{8-\delta}$ (BSCCO), with counterelectrodes of different natures – a normal metal (Ag), a conventional superconductor (Pb) and a ferromagnetic conductor (Fe). The carriers injected from these reservoirs have distinct characteristics, so that one expects them to integrate differently into the superconductive condensate or into the respective sea of quasiparticle excitations. The available tunneling channels are used differently. In particular, a transmission channel across the junction barrier typical for the presence of a superconductor is the Andreev reflection which can mediate the
onset of current carrying Andreev bound states (ABS) at the interface with high temperature superconductors. The ABS appear at the surfaces where the order parameter has a nonuniform phase and manifests as a subgap distribution of states. At zero energy, ABS leads to a zero bias conductance peak (ZBCP) with an amplitude proportional to the strength of order parameter anisotropy.

The Ag/BSCCO measurements (junctions A, B and C) probed the underdoped BSCCO crystal along its $ab$-plane under two angles. We observed that the amplitude of the ZBCP grew and the gap was filled with states when the junction orientation was near the $\langle 110 \rangle$ crystal surface. Alternatively, near the $\langle 100 \rangle$ surface, the BSCCO gap was clearly visible since the ABS formation is not favored. This pleads for a dominant $d_{x^2-y^2}$-wave symmetry order parameter. The rich subgap distribution of ABS also depend on the junction roughness, the respective features being more evenly angle-distributed but broadened when the transport is diffusive. We verified the several theoretical scenarios predicting the possibility of a subdominant $s$ or $d_{xy}$ on top of the $d_{x^2-y^2}$-wave pairing symmetry, the mixed state breaking the time reversal symmetry (BTRS) such that the zero energy ABS should shift to finite energies. We observed the ZBCP splitting associated with this shift at different critical temperatures much smaller than $d$-wave $T_c$.

It took place only in some cases, but we believe that the ZBCP decreasing in amplitude and broadening under a certain temperature is also a signature of BTRS, even if the splitting is relatively too small to be observed under the unavoidable smearing. Some junctions do not show the BTRS splitting or ZBCP decrement. We doubt the explanation based on doping dependence since our spectra have a pseudogap, so the crystals are in the underdoped regime. Therefore, if there is a quantum critical point, our measurement did not confirm its location close to the optimal doping. We reckon that the surface roughness is a more natural explanation.

The Pb/BSCCO structures (junctions D, E) behave consistently with the presence of two superconductors, the Pb gap manifesting as a ZBCP splitting of about 1.2 meV. However, much as in the case of BTRS splitting, the very sharp ZBCP just decreases in amplitude and broadens. The presence of a small ZBCP on the bottom of the Pb gap does not confirm the theory explaining the BTRS by coupling between superconductor and a proximity induced superconductive region on the normal side. Signs of an $s$-wave subcomponent – possibly induced
by Pb proximity onto the \textit{d}-wave side – are also visible in these spectra, the edges of the Pb being split.

The ferromagnetic structures in the Fe/Ag/BSCCO arrangement (junction F) probe the sensitivity of ABS to depairing influences like spin polarization. We measured a very large ZBCP splitting occurring immediately under \textit{d}-wave $T_c \approx 82$ K. The resulting finite energy peaks become gradually asymmetric at $T \approx 32$ K and develop shoulderlike lateral features. We interpret these developments as following: due to spinless character of the pair current intermediated by ABS as opposed to the spin polarized current on the ferromagnetic side, a spin accumulation appears at the junction barrier. This pair breaking distribution filters the charge currents according to the spin band from where they originate, thus determining the zero energy ABS to gain or loose energy depending on the spin direction, up or respectively down. Hence, the corresponding ZBCP splitting is proportional to the exchange interaction at the interface relative to the strength of the barrier. The spin origin of this splitting is proved by the effect of the spontaneous magnetization in the Fe layer: under a critical temperature the spins in the ultrathin film experience a reorientation from in-plane to perpendicular on the plane direction such that the magnetization start to affect the peak height symmetry. This unbalance is proportional to the polarization of the Fe film, about 35% in our case. In particular, this magnetic dependence excludes the possibility of a BTRS $p_y$-wave symmetry which would also split the ZBCP at $T_c$. We explain the shoulderlike features on the external sides of the peaks by a relatively small, BTRS inducing \textit{s}-wave subcomponent of about 6% which also splits the peaks resulting from the larger spin-filtering energy shift. Consequently, the spectrum has a 4-peak structure: a result of the combined effect of a spin dependent splitting – the spin-filtering barrier – and a spin independent factor – the BTRS state.

A heuristic continuation of the spin polarized measurements presented above should involve an external magnetic field oriented against the spin polarization of the junction. If the ZBCP is robust enough to survive a higher magnetic field, it should first be recovered as the peaks shift back to zero energy and then Zeeman split again. This would provide an undeniable proof regarding the spin dependence of this phenomenon.

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CHAPTER 4
Probing LOFF state

In Section 1.3 we saw that the coexistence of magnetism and superconductivity can create the LOFF state – an exotic superconductive state represented by Cooper pairs with non-zero total momentum. Since the transition to normal phase is regulated by Clogston criterion $h \geq \Delta/\sqrt{2}$, this is a very elusive state occupying a very narrow strip on the phase diagram at the boundary with the normal region. It was never unquestionably observed in the bulk of a superconductive material [161] due to the high exchange energies. However, if the superconductor is placed in contact with a ferromagnetic environment, the Clogston criterion can be relaxed and the pairs survive along several coherence lengths, $\xi_f = h/\Delta/2$ in the clean limit, across the proximity region. Consequently, most of the studies concerning LOFF state are done in hybrid junctions with a proximity region in the vicinity of the interface, where one can observe several exotic properties of the spatially inhomogeneous LOFF order parameter: oscillations of the density of states and gapless superconductivity [84, 90, 92], oscillations of $T_c$ and of the critical current $I_c$ with the thickness of the ferromagnetic layer in hybrid superconductive wafers [88, 89], etc.

In this chapter we present our own observations of LOFF state. In the first part, we observe the order parameter oscillations. Due to this oscillation, the conductance spectra show periodic inversions with increasing distance from the junction barrier. In the second part, we probe the dependence of this oscillation on the $d$-wave anisotropic symmetry.

4.1 Spatially modulated order parameter

As described in Section 1.3 and modeled in 1.4.4, in the proximity region the induced order parameter develops coherent damped oscillations as a function of distance from the barrier with the period proportional to the exchange energy and the finite momentum $q$ of the LOFF pair: $h/\Delta_f = 1/2\xi_f = q/2$. Formally, the oscillations can be regarded as the effect of a complex coherence length (which is real in a normal metal), the imaginary part being responsible for the modulation and the real part for the decay. A consequence and a test of this behavior, is the
DOS oscillation measurable using tunneling spectroscopy. In principle, the DOS in the proximity region depends on the distance from the barrier, such that, if it is resolved energetically through electrodes thin enough to confine the quasiparticle trajectories in this region, the gap should gradually vary up and down relative to the background until the superconductivity is either completely suppressed by a too strong $h$ or the counterelectrode becomes too thick and the measurement is just a diffusive average. Being a mirror of the order parameter successively taking positive and negative values (between the 0 and $\pi$ states) with increasing distance, the DOS oscillations are expected to show periodic complete flips along the current trajectory, that is spectral maxima (like the coherence peaks and ZBCP) must successively transform into minima and vice versa.

Of course, the high-$T_c$ subgap features associated with ABS are also affected since their amplitude and energy is different in the presence of an exchange field [162]. Thus, as we noticed before, the splitting of the energy bands in the ferromagnet lead to suppression of Andreev reflection. However, other phenomena are favored. Since in the LOFF state the spin up and down momenta are unequal, quantum interference is possible resulting in subgap resonances strongly affected by the magnitude of the exchange potential and the barrier scattering.

Based on these ideas, we build multilayered Fe/Ag/BSCCO junctions as pictured in Figs.2.1b and 2.3b. The six layers are probed successively, starting with the Ag layer which is supposed to offer information about the junction characteristics without an exchange potential even though, as we shall see, the layer may be slightly polarized itself, due to Fe migration. Each Fe layer measures the conductance spectra at increasing distances from the junction barrier, from 0 to 150 Å, by 30 Å. The pairs survive along a distance of several ferromagnetic coherence lengths. In our case, using the exchange energy $h \approx 170$ meV evaluated by fitting the experimental spectra (see below), we calculated $\xi_F \approx 36.8$ Å. Each layer was also measured within a temperature range, as represented in Fig.4.1 depicting the temperature dependence for the first three layers with the spectra normalized by the respective spectrum at 150 K.

We observed again the radical change of background conductivity at $T_c$. The difference in polarization is reflected in the opposite direction of this change: the conductivity drops in the Ag case (even if the very sharp drop may be just an accident) and increases in the Fe layer cases.
Figure 4.1  Temperature dependence of the conductance spectra collected on the first three layers. a) Ag layer; b) ≈ 30 Å Fe layer; c) ≈ 60 Å Fe layer.
The experimental data was theoretically extrapolated for each counterelectrode thickness $d$, using Zareyan’s quasiclassical model presented in Section 1.4.4 in the form disregarding the order parameter anisotropy. In Section 2.2 we estimated the mean free path $\ell \approx 100 \, \text{Å}$ for our Fe thin films, such that the quasiclassical condition $\ell < d$ should be satisfied for the first layers. Throughout the fitting process, the layer thickness was compared to $\xi_F$ by taking the distance parameter $d/\pi \xi_F$, a quantity related to the expected period of the DOS oscillation. The other free parameters were the superconductive gap $\Delta$, the exchange energy $h$ and the barrier transparency $Z$. We accounted for the interlayer irregularity by averaging spectra over a 10% distribution of thicknesses around each nominal $d$, a procedure which emulates the feature broadening and smearing observed in experimental spectra.

As visible in Fig.4.2, the spectra collected across the Ag/BSCCO junction indicate a DOS with a very typical cuprate gap opening at $T_c \approx 85$ K. The gap is well defined, without a ZBCP or other prominent subgap peaks, indicating a predominant injection near the (100) crystal surface, combined with a rarefied distribution of long trajectories due to the very low thickness of the film, determining a suppression of low energy states (see Section 1.4.4). The minigap discussed in Section 3.1.1 is also present, the coherence peaks are very broad bumps and a distinct pseudogap opens at $T^* \approx 160$ K. The peak-to-peak evaluated $d$-wave gap is $\Delta_d \approx 44$ meV at 40 K.

The first ferromagnetic layer, 30 Å of Fe, probes the first signs of an exchange interaction: the $\pm \Delta$ energies marked by maxima in the Ag case are now occupied by dips while the peaks move inside the gap at $\approx \pm 27$ meV when $T = 40$ K. The fit imposed an exchange energy $h = 25$ meV, sensibly lower than in the other Fe layers where $h = 170$ meV (see Fig.4.2b). Due to the interface mixing of the first Fe layer with the normal Ag layer, this evaluation seemed natural. Therefore, the exchange being very weak in this region ($0 < h < \Delta$), the only effect is the shift of the $\pm \Delta$ maxima to $\approx \pm h$. There is an upper limit for the trajectories due to disorder which may lead to the occurrence of minigap in the normal layer case (see the minigap discussion in Section 3.1.1). This constraint is enhanced by the exchange energy which makes the interference leading to ABS more difficult for oblique trajectories since the resonance is less probable for
Figure 4.2 Tunneling spectra obtained at different d-locations in the proximity region of a Fe/Ag/BSCCO junction. 

a) Spectra at low temperature, \( T = 5 \) K. Note the clear minigap. 

b) Spectra at \( T = 40 \) K and the corresponding fits. The parameters are \( \Delta = 44 \text{ meV}, \ h = 170 \text{ meV} (20 \text{ meV for the first } d = 30 \text{ Å}), Z = 0.51. \) The respective d values are represented in Fig.4.3b. 

c) Spectra at higher temperature, \( T = 60 \) K, where the superconductivity becomes even weaker with respect to \( h \) such that the LOFF oscillation is clearer.

quasiparticle experiencing gaps with various magnitudes (remember that the gap is anisotropic). 

Ergo, the states with low energy are suppressed up to a energy limit given by \( h \). Hence the conductance peaks at \( \pm h \).

At the next distance, \( d \approx 60 \text{ Å} \), we recorded a more dramatic change. The zero energy states shoot up, yielding the amplitude maximum of our oscillation. The subgap states invade the spectrum and the behavior reminds of the spectra collected on Junction F, presented in the previous chapter. The quasiparticle branches and the peaks are very asymmetric at low temperature and there is a split in the ZBCP. One may consider that this splitting is connected with the DOS dependence on the distance in the LOFF regime, but it is arguable that it is still
due to the barrier spin-filtering as in the previous chapter. We tend to believe this since, for this thickness, the splitting is present at all temperatures and it is smeared off when the superconductivity is weakened (see Fig.4.2c, curve at $d \approx 150$ Å) thermally and due to $h$. Another smearing factor is the increased distance which allows longer trajectories and a low energy distribution of ABS filling the small gap-like crevice. With the next layer, $d \approx 90$ Å, the spectrum collapses back into a gap which deepens even more at the next distance, $d \approx 120$ Å. The “inverse gap” feature is again recorded on the last layer $d \approx 150$ Å. It shows a ZBCP, split only at low temperatures. Thus, a $d$-dependent oscillatory cycle is completed with smaller and

![Figure 4.3 a) Experimental oscillations with distance $d$ developed in the ferromagnetic proximity region by the reduced DOS, $\sigma/\sigma_0 - 1$, at $E = (0, \pm \Delta)$ and the corresponding theoretical curves. The respective “normal” Ag points are also represented. b) Experimental oscillation of the reduced DOS at the temperatures represented in Fig.4.2. c) The same oscillation at 40 K with the values of $d$ (empty circles) obtained from the fitting procedure (see Fig.4.2b) compared with the nominal values $d$ (full circles).](image)
smaller amplitudes. Even if it is unclear, one can discern an oscillation in the size of $\Delta$ by following the position of highest energy maxima.

At the next distance, $d \approx 60 \, \text{Å}$, we recorded a more dramatic change. The zero energy states shoot up, yielding the amplitude maximum of our oscillation. The subgap states invade the spectrum and the behavior reminds of the spectra collected on Junction F, presented in the previous chapter. The quasiparticle branches and the peaks are very asymmetric at low temperature and there is a split in the ZBCP. One may consider that this splitting is connected with the DOS dependence on the distance in the LOFF regime, but it is arguable that it is still due to the barrier spin-filtering as in the previous chapter. We tend to believe this since, for this thickness, the splitting is present at all temperatures and it is smeared off when the superconductivity is weakened (see Fig.4.2c, curve at $d \approx 150 \, \text{Å}$) thermally and due to $h$. Another smearing factor is the increased distance which allows longer trajectories and a low energy distribution of ABS filling the small gap-like crevice. With the next layer, $d \approx 90 \, \text{Å}$, the spectrum collapses back into a gap which deepens even more at the next distance, $d \approx 120 \, \text{Å}$. The up-trend oscillation is again recorded on the last layer $d \approx 150 \, \text{Å}$ which shows a ZBCP, split only at low temperature. Thus, a $d$-dependent oscillatory cycle is completed with smaller and smaller amplitude. Even if unclearly, one can discern an oscillation in the size of $\Delta$ by following the position of highest energy maxima.

In Fig.4.3 we extracted the conductance oscillations at $E = 0$ and $E = \Delta = 44 \, \text{meV}$ and plotted them against the theoretically predicted curves. The match is at, least qualitatively, very good considering the fragility of the structure and the effects introduced by an anisotropic order parameter. We see clearly that, as the superconductivity is suppressed with increasing distance from the junction interface, the reduced DOS, $\sigma/\sigma_0 - 1$, decays oscillatory. As visible on Fig.4.3, the maximum of experimental amplitude is reached at the extrapolated distance $d/\pi \xi_f \approx 0.45$. The normalized conductance peak is lower than the theoretical one (which is about 1.6) due to the ZBCP splitting. Consequently, the maxima at higher temperatures (Fig.4.3b) are higher and, in general, the higher temperature spectra correspond to a cleaner oscillation due to the decreasing superconductivity and predominance of LOFF effects. Notably, the conductances at $E = 0$ and $E = \Delta$ oscillate in antiphase showing that, indeed, the alternating order parameter
proceeds through negative and positive values as the system successively switches between 0 and $\pi$-states.

### 4.2 Effects of a $d$-wave pairing anisotropy

In the previous section, the main target was to see the LOFF modulation with distance from the junction barrier. In this section we shall measure the LOFF state behavior keeping the distance fixed, but changing the junction orientations $\beta$, in order to analyze the effect of an anisotropic order parameter.

Consequently, we built two junctions onto two planes of the same BSCCO crystal, exactly with the counterelectrode arrangement sketched in Fig.2.1a. Specifically, three metallic layers were deposited: a thin 30 Å Ag layer, an intermediate 70Å slab of Fe without any external contacts and another thicker Ag film. The first Ag layer was used to measure a “normal” tunneling spectrum, and the second Ag film to collect the LOFF spectrum at the upper surface of the Fe layer. Zareyan’s anisotropic $d$-dependent model presented in Section 1.4.4 will be used for theoretical evaluation.

The change in $\beta$ is expected to influence the phase, the amplitude and the period of the oscillation. Thus, for a ballistic junction, varying $\beta$ from 0 to $\pi/4$ should shift the maxima by $\pi$ while the amplitude is decreased (see Fig.1.13b). The maxima shift is due to change in the sign of the order parameter similar to the change induced by a variation in distance $d$ equal to a period. The inhomogeneous scattering onto the junction barrier leads to diffusive transmission and affects the antiphase character of the oscillations at 0 and $\pi/4$.

In Fig.4.4 we represented the variation with distance corresponding to the two angles. One can see that, close to the $\langle 110 \rangle$ plane (or $\beta = \pi/4$), the spectrum measured on the first Ag layer correctly shows a well defined albeit broad ZBCP. However, when the same angle is probed across the Fe layer, the curve is flipped upside down similar, the spectrum looking like a gap. We practically observe the same maxima we measured onto the 60 Å layer in the previous section. Likewise, the phenomenon happens for the injection closer to $\langle 100 \rangle$ crystal plane ($\beta = 0$): the initial well defined gap is inversed forming a noisy but well defined ZBCP.
Figure 4.4 Tunneling spectra collected onto the same BSCCO crystal under two different junction orientations, through a normal counterelectrode (Ag) and an ferromagnetic one (Fe). a) In the direction near the $\langle 110 \rangle$ plane, the ZBCP measured via Ag becomes a gap via Fe layer. b) Near the $\langle 100 \rangle$ surface, the gap Ag flips into a broad ZBCP when probed through Fe.

The dependence on the junction orientation is represented on the same graph in Fig.4.5a. Note that the maxima in one direction are replaced by minima in the other direction. The visual fitting couldn’t reproduce the tilted background but offered a good enough match of the distance $d$ dependence as seen in Fig.4.5b, $d_x \approx 68 \text{ Å}$, $d_y \approx 65 \text{ Å}$. Also, in evaluating the normalizing background conductance $\sigma_0$, we used the high energy quasiparticle conductance obtained from fitting the low energy spectral features. The difference between angles is sensibly lower than $\pi/4$, but the behavior opposite to the normal counterelectrode case is still observable. Thus, when the DOS spectrum shows a ZBCP feature we interpret it as being the $\pi$-state (due to the presumed positive value of the order parameter) and the angle is $\beta_\pi \approx 10^\circ$, that is the junction
orientation is close to the antinodal direction. On the other hand, for the 0-state gap-like spectrum, the extrapolated angle is $\beta_0 \approx 38^\circ$, that is the junction orientation is almost nodal. In Fig.4.5b we plotted the expected reduced DOS oscillation at $E = 0$, similar to the one represented in Fig.4.5a, but using the anisotropic order parameter model for $\beta_0$ and $\beta_\pi$. The exchange potential was higher that the one obtained in the previous section $h \approx 200$ meV for a maximum gap $\Delta \approx 70$ meV which is obviously a broadened value. This broadening is justified by the relatively high value of the barrier roughness parameter, $Z \approx 1.1$. Note again that the oscillation is again in antiphase in the two directions.

![Graph showing DOS oscillation](image)

**Figure 4.5** a) LOFF states with positive and negative order parameters ($\pi$ and 0-states) observed on the same BSCCO crystal under two different junction orientations, $\beta_\pi$ and $\beta_0$ with the values depicted obtained from the fits (smooth lines). b) Theoretically expected reduced DOS oscillation calculated at the experimental angles $\beta_{\pi,0}$. The points corresponding to the spectra represented in a) are placed on the modulated curves at the positions given by the fitted distance values, $d_{\pi,0}$. 

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4.3 Conclusions

In this final chapter we presented our own experimental survey regarding the properties of LOFF state: an exotic superconductive state combining the otherwise incompatible phenomenology of superconductivity and magnetism. The LOFF presence is difficult to detect in the bulk of materials due to the exchange energy much larger than the superconductive gap. On the other hand, tunneling spectroscopy proved to be a powerful tool suitable for LOFF detection since the nature of the constraints preventing the LOFF onset in the bulk can be tweaked in the vicinity of the superconductor, in the proximity region with a ferromagnetic material. The main phenomenon we investigated in this region was the oscillation of the LOFF order parameter with distance from the junction interface as well as the dependence of this oscillation on the anisotropy of the BSCCO pairing which (as we saw throughout the thesis) we have reasons to believe is predominantly $d_{x^2-y^2}$-wave.

Therefore, in the first part, we built planar Fe/Ag/BSCCO junctions, with the Fe counterelectrode imparted in layers of equal thickness to be probed separately. One of the jobs intended for the Ag layer was to measure the tunneling spectrum without an exchange field, while each Fe film was supposed to probe a different distance from the barrier. Since the Ag and Fe spectra are obtained in the same microscopic conditions (i.e., the same barrier roughness and junction orientation) the normal and ferromagnetic spectra could be considered as originating from the same junction with a better degree of confidence compared to the alternative solution: to repolish the junction after each distance measurement.

Temperature dependent spectra were collected on each counterelectrode and, indeed, we observed that the DOS characteristics oscillate with increasing distance, alternatively taking values above and under the background. At low temperatures and distances, the spectral features are affected by the barrier polarization similar to the effects observed in the spin polarized junction F presented in the previous chapter. The maximal amplitude is obtained at $d \approx 60 \ \text{Å}$ and we obtained a whole oscillation cycle matching well enough the theoretical expectation. Each curve was extrapolated at $T \approx 40 \ \text{K}$ using a ballistic quasiclassical model and we noticed that the spectral maxima shift sign at $E = 0$ and $E = \Delta$, proving that the regime successively passes through negative and positive order parameters – a phenomenon associated with the LOFF state.
In the second part, we performed a similar measurement, but keeping the distance \( d \) fixed and changing the junction orientation. We observed again the curve flipping at a distance corresponding to a maximum of amplitude in the previous experiment. However, this time, the initial ZBCP probed in the \( \beta = \pi/4 \) direction is transformed at the respective distance into a gap-like dip. The opposite spectra obtained on the normal counterelectrode are obtained again in the Fe region, but for different angles, proving that the change in \( d \)-wave phase introduces a phase shift of \( \pi \) in the LOFF oscillation. Subsidiarily, the oscillation dependency on the junction orientation can be considered as another test proving the anisotropy of the high-\( T_c \) order parameter. The curves were theoretically fitted using the anisotropic version of the model and we calculated the respective angles where the order parameter is positive \( (\beta_2 \approx 10^\circ) \) and negative \( (\beta_0 \approx 38^\circ) \). Subsequently, we could place the experimental extremal amplitudes on the same graph with the theoretical oscillation calculated at \( E = 0 \) for the extrapolated angles \( \beta_{\pi,0} \).

This last experiment should be continued to make sure that the respective behavior is not an experimental coincidence. Although it is reproducible, for complete confidence, the spectra should be evaluated at another distance to see if, indeed, the state measured is LOFF: the two distances per separate angle should show both an oscillation with distance and junction orientation.

In conclusion, we successfully probed the existence of LOFF state in the proximity region of a hybrid junction between a high \( T_c \) superconductor and a ferromagnetic thin film counterelectrode. The signature of the LOFF order parameter is the spatially damped oscillation of the subgap density of states. The oscillation sign and magnitude depends on the energy and the phase of the \( d \)-wave anisotropic pairing potential.
Appendices

Appendix A: Visual Basic implementation of the anisotropic BTK theoretical model developed by Kashiwaya and Tanaka [120] presented in Sections 1.4.2, 1.4.4 and used in Chapter 3. Subroutine Sigma sums the spin up and down contributions integrated over injection angle $\theta$. The separate contributions are denoted in the model by $\sigma_{\uparrow}$ and $\sigma_{\downarrow}$.

```vbnet
Const Rd = 0.01745329
Dim AAm, BBm, AAp, BBp, A, B As Double
Dim ReDm, ReDp, ImD As Double

'Total DOS = sum of spin Up and Down DOS
Public Sub Sigma(ByVal E As Double, ByRef SigmaValueForE As Double)
    Dim SUp As Double, SDn As Double, RN As Double
    Call SigmaUpAngleInt(E, -89.99, 89.99, NrTrapez, SUp)
    Call SigmaDnAngleInt(E, -89.99, 89.99, NrTrapez, SDn)
    SigmaValueForE = (SUp + SDn)
End Sub

'Defines the order parameter real and imaginary parts, minus (m) and plus (p)
Private Sub InitDelta(ByVal Angle As Double)
    ReDm = Delta1 * Cos(2 * (Angle + Rd * Beta))
    ReDp = Delta1 * Cos(2 * (Angle - Rd * Beta))
    ImD = Delta2
End Sub

Private Sub InitGamma(E As Double)
    Dim Am, Bm, Ap, Bp, ModDm, ModDp, Cm, Dm, Cp, Dp As Double
    'Gamma function complex components, simple [Am,Bm],[Ap,Bp] and cap
    ' [AAm, BBm],[AAp, BBp], minus (m) and plus (p), and function Gamma*Gamma (A,B)
    ModDm = (ReDm ^ 2 + ImD ^ 2) ^ 0.5
    If E ^ 2 - ModDm ^ 2 < 0 Then
        Am = E / ModDm
        Bm = (ModDm ^ 2 - E ^ 2) ^ 0.5 / ModDm
        Else
        Am = (E - (E ^ 2 - ModDm ^ 2) ^ 0.5) / ModDm
        Bm = 0
    End If
    AAm = (Am * ReDm - Bm * ImD) / ModDm
    BBm = (Am * ImD + Bm * ReDm) / ModDm
    ModDp = (ReDp ^ 2 + ImD ^ 2) ^ 0.5
    If E ^ 2 - ModDp ^ 2 < 0 Then
        Ap = E / ModDp
        Bp = (ModDp ^ 2 - E ^ 2) ^ 0.5 / ModDp
        Else
        Ap = (E - (E ^ 2 - ModDp ^ 2) ^ 0.5) / ModDp
        Bp = 0
    End If
    AAp = (Ap * ReDp + Bp * ImD) / ModDp
    BBp = (Bp * ReDp - Ap * ImD) / ModDp

    'Gammas take different forms for negative and positive energies.
    If E < 0 Then
        Cm = -AAm / (AAm ^ 2 + BBm ^ 2)
        Dm = -BBm / (AAm ^ 2 + BBm ^ 2)
        Cp = -AAp / (AAp ^ 2 + BBp ^ 2)
        Dp = -BBp / (AAp ^ 2 + BBp ^ 2)
        AAm = Cm
        BBm = Dm
        AAp = Cp
        BBp = Dp
    End If
    A = AAp * AAm - BBp * BBm
    B = AAp * BBm + AAm * BBp
End Sub
```
'Calculates the spin Up current for an angle Th
Private Function SigmaUp(E As Double, Th As Double) As Double
    Dim A1, B1, C1, T1 As Double
    Dim ThS As Double, ThA As Double, L1, L2, ZUp, ZDn As Double

'Initialize order parameter function and the Gammas
    InitDelta (Rd * Th)
    InitGamma (E)
    T1 = Sqr((1 + h) / (1 - h)) * Sin(Rd * Th)

'The DOS takes different values for different current injection angle (Theta) ranges
    If Abs(T1) <= 1 Then 'INTERVAL #1
        'Define the ThetaS and ThetaA angles and
        ThS = ArcSin(T1 * Sqr(1 - h))
        ThA = ArcSin(T1)

'Lambda functions, 1 and 2, and Z, up and down.
        L1 = (1 + h) ^ 0.5 * Cos(Rd * Th) / Cos(ThS)
        L2 = (1 - h) ^ 0.5 * Cos(ThA) / Cos(ThS)
        ZUp = (Z0 - U) / Cos(ThS)
        ZDn = (Z0 + U) / Cos(ThS)

'Angular density of states DOS
        B1 = (1 + L1) * (1 + L2) + ZUp * ZDn * (1 - A - (1 - L1) * (1 - L1) * A + B * (ZDn * (1 - L1) * (1 - L2) + ZUp * ZDn)
        C1 = ZDn * (1 + L2 + (1 - L2) * A) - ZDn * (1 + L1 + (1 - L1) * A) - B * ((1 - L1) * (1 - L1) * (1 - L2) + ZUp * ZDn)
        SigmaUp = A1 / (B1 * B1 + C1 * C1)
    Else
        If Abs(T1 * Sqr(1 - h)) <= 1 Then 'INTERVAL #2
            ThS = ArcSin(T1 * Sqr(1 - h))
            L1 = (1 + h) ^ 0.5 * Cos(Rd * Th) / Cos(ThS)
            L2 = -Sqr((1 - h) * (T1 * T1 - 1)) / Cos(ThS)
            ZUp = (Z0 - U) / Cos(Rd * ThS)
            ZDn = (Z0 + U) / Cos(Rd * ThS)
            A1 = 4 * L1 * (1 - (A ^ 2 + B ^ 2)) * (1 + (ZDn - L2) ^ 2) * (1 + (ZDn - L2) ^ 2)
            C1 = ZDn * (1 + L2 + (1 - L2) * A) - ZDn * (1 + L1 + (1 - L1) * A) - B * ((1 - L1) * (1 - L1) * (1 - L2) + ZUp * ZDn)
            SigmaUp = A1 / (B1 * B1 + C1 * C1)
        Else
            SigmaUp = 0 'INTERVAL #3
        End If
    End If
End Function

'Calculates the spin Down current for an angle Th
Private Function SigmaDn(E As Double, Th As Double) As Double
    Dim A1, B1, C1, T1 As Double
    Dim ThS As Double, ThA As Double, L1, L2, ZUp, ZDn As Double

'Initialize order parameter function and the Gammas
    InitDelta (Rd * Th)
    InitGamma (E)
    T1 = Sqr((1 + h) / (1 - h)) * Sin(Rd * Th)

'The DOS takes different values for different current injection angle (Theta) ranges
    If Abs(T1) <= 1 Then 'INTERVAL #1
        'Define the ThetaS and ThetaA angles and
        ThS = ArcSin(T1 * Sqr(1 - h))
        ThA = ArcSin(T1)

'Lambda functions, 1 and 2, and Z, up and down.
        L1 = (1 + h) ^ 0.5 * Cos(Rd * Th) / Cos(ThS)
        L2 = (1 - h) ^ 0.5 * Cos(ThA) / Cos(ThS)
        ZUp = (Z0 - U) / Cos(ThS)
        ZDn = (Z0 + U) / Cos(ThS)

'Angular density of states DOS
        C1 = ZDn * (1 + L2 + (1 - L2) * A) - ZDn * (1 + L1 + (1 - L1) * A) - B * ((1 - L1) * (1 - L1) * (1 - L2) + ZUp * ZDn)
        SigmaDn = A1 / (B1 * B1 + C1 * C1)
    Else
        SigmaDn = 0 'INTERVAL #3
    End If
End Function
If Abs(T1 * Sqr(1 - h)) <= 1 Then  'INTERVAL #2
  ThS = ArcSin(T1 * Sqr(1 - h))
  L1 = (1 - h) ^ 0.5 * Cos(Rd * Th) / Cos(ThS)
  L2 = Sqr((1 + h) * (T1 * T1 - 1)) / Cos(ThS)
  ZUp = (Z0 - U) / Cos(ThS)
  ZDn = (Z0 + U) / Cos(ThS)
  A1 = 4 * L1 * (1 - (A ^ 2 + B ^ 2)) * (1 + (ZDn - L2) ^ 2)
  B1 = 1 + L1 + (ZDn + L2) * ZUp - A * ((1 - L1) * (1 - ZDn) + ZUp * L2) + B * ((1 -
  L1) * L2 + ZUp * ZDn)
  C1 = ZUp - (L2 + ZDn) * (1 + L1) - B * ((1 - L1) * (1 - ZDn) + ZUp * L2) + A * ((1 -
  L1) * L2 + ZUp * ZDn)
  SigmaDn = A1 / (B1 * B1 + C1 * C1)
Else
  SigmaDn = 0  'INTERVAL #3
End If
End Function

'Angle integrations by trapeze method
'...for spin Up current
Public Sub SigmaUpAngleInt(ByVal E As Double, ByVal Angle1 As Double, ByVal Angle2 As Double,
ByVal n As Integer, ByRef SigmaUpAngleSum As Double)
  Dim j As Integer
  Dim Ah, AngleStep As Double
  Ah = (Angle2 - Angle1) / n
  SigmaUpAngleSum = 0
  AngleStep = Angle1 + Ah
  For j = 1 To (n - 1)
    SigmaUpAngleSum = SigmaUpAngleSum + SigmaUp(E, AngleStep) * Cos(Rd * AngleStep) * (1 + h)
    AngleStep = AngleStep + Ah
  Next
  SigmaUpAngleSum = 0.5 * Ah * (SigmaUp(E, Angle1) * Cos(Rd * Angle1) * (1 + h) ^ (3 / 2) / 2 +
  SigmaUp(E, Angle2) * Cos(Rd * Angle2) * (1 + h) ^ (3 / 2) / 2 + 2 * SigmaUpAngleSum)
End Sub

'...and for spin Down current
Public Sub SigmaDnAngleInt(ByVal E As Double, ByVal Angle1 As Double, ByVal Angle2 As Double,
ByVal n As Integer, ByRef SigmaDnAngleSum As Double)
  Dim j As Integer
  Dim Ah, AngleStep As Double
  Ah = (Angle2 - Angle1) / n
  SigmaDnAngleSum = 0
  AngleStep = Angle1 + Ah
  For j = 1 To (n - 1)
    SigmaDnAngleSum = SigmaDnAngleSum + SigmaDn(E, AngleStep) * Cos(Rd * AngleStep) * (1 - h)
    AngleStep = AngleStep + Ah
  Next
  SigmaDnAngleSum = 0.5 * Ah * (SigmaDn(E, Angle1) * Cos(Rd * Angle1) * (1 - h) ^ (3 / 2) / 2 +
  SigmaDn(E, Angle2) * Cos(Rd * Angle2) * (1 - h) ^ (3 / 2) / 2 + 2 * SigmaDnAngleSum)
End Sub
Appendix B: Visual Basic implementation of the quasiclassical ballistic model developed by Zareyan [122,123], presented here in Section 1.4.4 and used in Chapter 4. Note that the exponential integrals employed by the model are not calculated explicitly. Instead they were first evaluated in Maple for about 50000 discrete values and stored in two external files as explained below.

\[
\text{Const } n\text{Max} = 15, \text{ NrD} = 10, \text{ dWidth} = 1 / 10, \text{ d1} = 2
\]

The exponential integral files ReEI.dat and ImEI.dat contain 5 columns with the respective ‘real or imaginary parts. Each column is calculated for a different value of d/l_impurities, 'from 0 with increments of 0.1. The constant dl indicates which column is to be used in the 'following calculation.

\[
\text{Dim ReEI}(5, 10005) \text{ As Double, ImEI}(5, 10005) \text{ As Double, Cs}(5) \text{ As Double, } dT(2 * \text{ NrD + 1}) \text{ As Double}
\]

\[
\text{Dim EIRange As Double, NrP As Integer}
\]

Public Sub ReadEIFile()
Dim i As Integer
Open "ReEI.dat" For Input As #1
Open "ImEI.dat" For Input As #2
Erase ReEI, ImEI
i = 0
While EOF(1) = False
Input #1, ReEI(1, i), ReEI(2, i), ReEI(3, i), ReEI(4, i), ReEI(5, i)
i = i + 1
Wend
Close #1
EIRange = ReEI(1, 0)
NrP = ReEI(2, 0)
For i = 1 To 5
Cs(i) = ReEI(i, 1) * ReEI(i, 1)
Next
i = 0
While EOF(2) = False
Input #2, ImEI(1, i), ImEI(2, i), ImEI(3, i), ImEI(4, i), ImEI(5, i)
i = i + 1
Wend
Close #2
End Sub

Public Sub ExpI(ByVal ImIP As Double, ByRef ReOP As Double, ByRef ImOP As Double)
Dim i, Lin As Integer, Stp As Boolean
Dim xStep, xx As Double
If Abs(ImIP) <= EIRange Then
xStep = EIRange / (NrP - 1)
xx = 0
i = 2
Stp = False
While Stp = False And i <= NrP + 1
If Abs(ImIP) <= xx Then
ReOP = ReEI(d1, i)
ImOP = ImEI(d1, i)
Stp = True
End If
xx = xx + xStep
i = i + 1
Wend
Else
ReOP = 0
ImOP = 0
End If
End Sub
'DOS calculated for counterlectrode thickness d

Public Sub dDOS(ByVal dd As Double, ByVal E As Double, ByRef ReDOS As Double, ByRef ImDOS As Double, ByRef AbsDOS As Double)
Dim n As Integer
Dim rE2m As Double, iE2m As Double, rE2p As Double, iE2p As Double
Dim rP0, iP0, rP, iP As Double, rD, iD As Double
ReDOS = 0
ImDOS = 0
For n = -nMax To nMax
    Call ExpI(2 * n * dd * (E + h) / Delta, rE2p, iE2p)
    Call ExpI(2 * n * dd * (E - h) / Delta, rE2m, iE2m)
    If E <= Delta Then
        rP0 = (rE2p ^ 2 - iE2p ^ 2) / Cs(dl)
        iP0 = 2 * rE2p * iE2p / Cs(dl)
        rP = T * (rP0 * (1 - R * rP0) - R * iP0 * iP0) /
            ((1 - R * rP0) * (1 - R * rP0) + R * R * iP0 * iP0)
        iP = T * iP0 /
            ((1 - R * rP0) * (1 - R * rP0) + R * R * iP0 * iP0)
        rD = rP * Cos(2 * n * Arccos(E / Delta)) - iP * Sin(2 * n * Arccos(E / Delta))
        iD = iP * Cos(2 * n * Arccos(E / Delta)) + rP * Sin(2 * n * Arccos(E / Delta))
    End If
Else
    rP0 = (rE2m ^ 2 - iE2m ^ 2) / Cs(dl)
    iP0 = 2 * rE2m * iE2m / Cs(dl)
    rP = T * (rP0 * (1 - R * rP0) - R * iP0 * iP0) /
        ((1 - R * rP0) * (1 - R * rP0) + R * R * iP0 * iP0)
    iP = T * iP0 /
        ((1 - R * rP0) * (1 - R * rP0) + R * R * iP0 * iP0)
    rD = rP * Exp(-2 * Abs(n) * HArccos(Abs(E / Delta)))
    iD = iP * Exp(-2 * Abs(n) * HArccos(Abs(E / Delta)))
End If
ReDOS = ReDOS + rD
ImDOS = ImDOS + iD
Next n
ReDOS = ReDOS / 2
ImDOS = ImDOS / 2
AbsDOS = Sqr(ReDOS ^ 2 + ImDOS ^ 2)
End Sub

Public Sub ThicknessRange()
Dim dStep, dxx As Double, i As Integer, dW As Double
dW = dWidth * d
dStep = dW / (2 * NrD)
dxx = d - dW / 2
For i = 1 To 2 * NrD + 1
    dT(i) = d * Exp(-(dxx - d) ^ 2 / (2 * dW ^ 2))
dxx = dxx + dStep
Next i
End Sub

'Net DOS averaged over a Gaussian distribution of thicknesses

Public Sub DOS(ByVal EE As Double, ByRef ReDOS As Double, ByRef ImDOS As Double, ByRef AbsDOS As Double)
Dim i As Integer
Dim SumReD, SumImD, SumAbsD, SRD As Double, SID As Double, SAD As Double
SumReD = 0
SumImD = 0
SumAbsD = 0
For i = 1 To 2 * NrD + 1
    Call dDOS(dT(i), EE, SRD, SID, SAD)
    SumReD = SumReD + SRD
    SumImD = SumImD + SID
    SumAbsD = SumAbsD + SAD
Next
ReDOS = SumReD / (2 * NrD + 1)
ImDOS = SumImD / (2 * NrD + 1)
AbsDOS = SumAbsD / (2 * NrD + 1)
End Sub
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Vita

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I was born on August 23rd, 1968, in the city of Moinești, Romania. In 1994, I graduated with a Physics Diploma from University of Bucharest, Romania. Between 1994 and 1997, I’ve been working as a research assistant in the Institute of Physical-Chemistry of Romanian Academy. In 1997, I was accepted as a graduate student in the PhD program of the Department of Physics and Astronomy, University of Kentucky from where I received a MS degree in 2000.

Recent Journal Publications:


Selected Presentations:

M. Freamat and K.-W. Ng, “Exchange field effect on the Andreev bound state of high Tc Superconductors”, APS March Meeting, Montreal, Canada (2004)

M. Freamat and K.-W. Ng, “Spatial oscillations of the superconductive order parameter in ferromagnet/ (d+is)-wave superconductor planar junctions”, APS March Meeting, Austin, TX (2003)


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