Spin-triplet supercurrents of odd and even parity in nanostructured devices

Lahabi, K.

1

INTRODUCTION

CONVENTIONAL ELECTRONICS relies on the motion of individual electrons in a conducting material. This type of charge transport is characterised by electron scattering, a dissipative process which results in finite electrical resistance. Good conductors such as gold or silver are characterized by fewer scattering events, and therefore smaller resistance. But what about superconductors? These are electronic systems with exactly zero electrical resistance below a critical temperature (T_c). This is however not the only (or the most profound) distinguishing quality of superconductors, compared to other electronic systems (e.g. normal metals or semiconductors), superconductors follow a fundamentally different set of rules. In fact, one can argue that in some respects superconductors have more in common with the vegetable cauliflower than they do with a good conductor like gold.

Elementary particles can be classified into two main categories based on their intrinsic angular momentum, or "spin". Those with half-integer spins (1/2, 3/2, etc.) are called Fermions while the ones with integer spins (0, 1, 2, etc.) are known as Bosons. The difference between these two classes however does not end with spin, they also follow entirely different distributions. In a system of fermions, each particle acts as an individual object with a unique quantum state which, in principle, can be distinguished from the rest[1]. In contrast, an indefinite number of bosons can share a single quantum state, and form a "condensate" of indistinguishable particles.

[1] In practice however, this can only be realised if there are a finite number of discrete states.

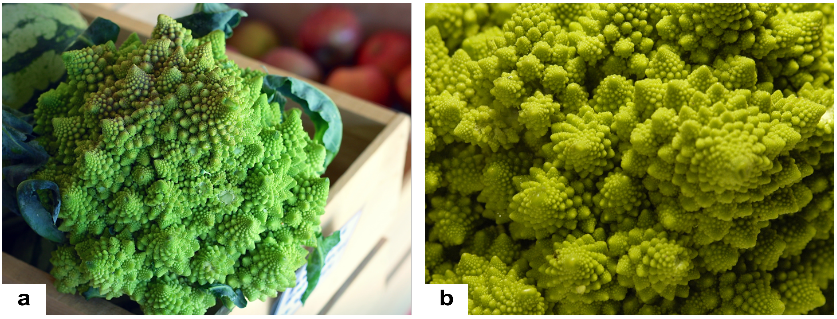

Figure 1.1: The Roman cauliflower (Romanesco). Without the help of the items in the background in **a**, it is practically impossible to guess the actual length scale in **b**. The self-similar construction of the buds makes their actual size irrelevant.

Superconductivity occurs by the condensation of pairs of electrons into a macroscopic quantum state. The paired electrons are called Cooper pairs[2] and, unlike electrons, which are spin 1/2 fermions, they are Bosonic in nature. This means that all paired electrons can share a single quantum state. As an interesting consequence, the wavefunction of a pair of electrons (referred to as the order parameter) can now elegantly describe the entire superconducting condensate, and vice versa. In this sense, a superconductor is rather analogous to the Romanesco cauliflower (shown in Figure 1.1), where the structure of each bud is indistinguishable from the ones it is made of.

In Figure 1.1 **a**, the Romanesco head is shown together with a number of other objects, while in Figure 1.1 **b** the buds appear by themselves. What is striking here is that the lack of reference objects has made it almost impossible to guess the actual length scale in **b**. One way to interpret this is that (at least to a large extent) the size of the system has lost its significance. If the sole function of a photograph is to help us identify objects, in the case of Romaneso the image can cover anywhere between a few microns to a few metres; and still produce the same result. This reasoning can also be applied to superconductivity, which is a macroscopic quantum phenomenon. In almost all other physical systems, the individual quantum states begin to smear out by increasing the number of interacting particles. As a result, all the intriguing aspects of quantum mechanics are typically observed in systems with no more than a few atoms. Superconductors and superfluids on the other hand, do not suffer this drawback, making them ideal platforms for exploring various quantum phenomena.

In analogy to Figure 1.1, whether we probe a single Cooper pair or a macroscopic

[2] named after Leon Cooper who developed the first microscopic theory of superconductivity with John Bardeen and John Robert Schrieffer (the BCS theory) in 1957 [1].

superconductor, there is only one order parameter with the same set of quantum characteristics. This however does not imply that the size of a superconducting system has no significance. Quite the contrary, as we will discuss throughout this thesis, mesoscopic systems can offer considerably better control over superconductivity, and are far more practical for device applications. Apart from their technological implications, mesoscopic systems can also be used to identify, and in certain cases even create, some of the rarest and most exotic quantum states in nature. Such insights are crucial to our understanding of the mechanisms involved in some of the most controversial phenomena of modern Physics.

The behaviour of any quantum system is determined by its wavefunction. In superconductors, this corresponds to the pairing function of the electrons that form the Cooper pair. A superconductor can therefore be characterized by the type of symmetry that describes its pairing function. It is well established by the exclusion principle that fermions, such as electrons, can only be paired with each other if their combined wavefunction is antisymmetric i.e. can be represented by an *odd function* $f(-x) = -f(x)$. One way to satisfy this condition would be if the pairing occurred between electrons with opposite spins.This turns out to be the case in the overwhelming majority of all currently known superconductors (and superfluids). There is however no reason for this to be the only stable configuration. Cooper pairs can also form by electrons of equal spin.

Besides spin, a wavefunction has two other components that determine its pairing symmetry. These are space and time which, for practical reasons, are commonly represented in the form of momentum and frequency, respectively. Equal-spin pairing is allowed, as long as one of the two other components (but not both) corresponds to an odd function. The phenomenon is known as triplet superconductivity, and is the main subject of this thesis. There are two general categories of triplet superconductors based on their pairing symmetry: odd-momentum with even-frequency, and even-momentum with odd-frequency. It appears that both categories are extremely rare in nature. At present, we know only a handful of materials with odd-momentum triplet pairing, and odd-frequency triplet correlations have only been "generated" in carefully engineered superconductor-ferromagnet (S-F) hybrid systems. On the other hand, triplet Cooper pairs have become an ingredient in a multitude of newly emerging fields of condensed matter physics, with a growing number of applications in quantum computing, spintronics and superconducting electronics. This calls for a deeper understanding of the physics behind triplet superconductivity, and developing the means for its control so it can be utilized in upcoming device applications.

The research presented in this thesis extends into both categories of triplet superconductors. This involves both implementing S-F hybrids as the platform to explore odd-frequency triplet correlations, and investigating the unusual characteristics of strontium ruthenate Sr_2RuO_4, a leading candidate for odd-momentum triplet pairing. In each case we use *mesoscopic structures* as the principal tool to gain new insights into some of the most subtle and yet distinct characteristics of triplets, which otherwise would be very challenging to observe. Moreover, combining well-defined geometries with the substantial role of confinement in defining the free energy of a system makes mesoscopic structures the means to not merely observe, but gain effective control over the unique aspects of triplet pairing.

In S-F hybrids we control the odd-frequency triplet correlations by utilizing the shape of the ferromagnet to create a well-defined micromagnetic configuration. As triplet correlations are highly sensitive to the spin-texture the ferromagnet (or rather the exchange field gradient), micromagnetics can provide the means to control their amplitude, phase and even the location of their current path in the ferromagnet. The potential of such degree of control over superconductivity, and its implications in superconducting electronics are profound. A notable example of this is presented here in the form of a possible new type of non-volatile superconducting memory element, developed by combining the unique characteristics of triplet correlations with the controllable micromagnetic configuration of a disk-shaped Josephson junction.

As for Sr_2RuO_4, there are two main aspects to the use of mesoscopic structures in our studies. The first is related to the observation of an unusual state known as the half-quantum vortex (HQV). In the context of Sr_2RuO_4, the HQV is a result of equal-spin triplet pairing, and is also expected to be a host to the highly sought-after Majorana zero-modes[3] [3]. Unlike the ordinary (full-quantum) vortex, the HQV is accompanied by a spin current whose free energy grows logarithmically with the dimensions of system. Consequently, the HQV states become energetically less favourable, and unlikely to stabilise in macroscopic (bulk) systems. One solution to this is to reduce the size of the system; so that the spin current associated with the HQV can be contained within the geometrical boundaries of the system which, in our case is a micron-sized ring and is designed for field-dependent transport measurements.

Interestingly however, the significance of mesoscopic structures for Sr_2RuO_4 goes well beyond the HQV. Unlike the ordinary superconductor, which is described by a single macroscopic quantum state, Sr_2RuO_4 is expected to have a twofold degenerate ground state; with different directions of orbital angular momentum for the condensate [4]. This breaking of time-reversal symmetry is associated with the so-called "chiral" superconducting states. Here, chirality refers to the direction-dependent phase of the superconducting order parameter. As the orbital phase can either wind clockwise or anticlockwise, there are two distinct chiral states (e.g. left or right) available to the order parameter. An interesting consequence of these degenerate

[3] also referred to as Majorana Fermions: a class of particles which, unlike protons and electrons, are their own antiparticles. While evidently rare in nature, in the past two decades Majorana Fermions have enjoyed substantial popularity for their potential in fault-tolerant quantum computing. More details on the topic can be found in [2].

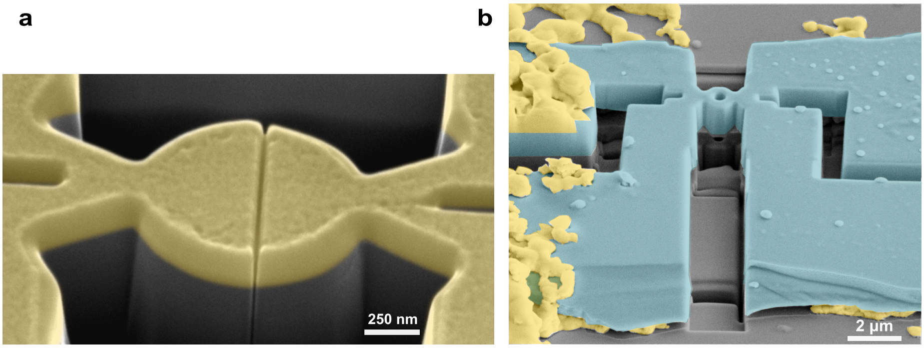

Figure 1.2: Examples of FIB milling used for structuring of the different systems discussed in this thesis (false coloured electron microscope images) . **a** Disk-shaped Josephson junction, structured from a multi-layer of Co/Cu/Ni/Nb. The junction is formed by the central trench. The gap is less than 20 nm wide, and cuts the top superconducting Cu/Ni/Nb layers in two halves — leaving only Co as a ferromagnetic barrier connecting them. **b**, a mesoscopic ring structured by milling a single Sr_2RuO_4 crystal (cyan), residing on a $SrTiO_3$ substrate, which is contacted by silver epoxy (gold) for electrical transport measurements.

ground states is the emergence of chiral superconducting domains, where the two chiral states are segregated in real space. Despite numerous efforts over the past two decades, a direct observation of such domains is still lacking. The vast majority of these experiments have been limited to bulk crystals of Sr_2RuO_4, typically hundreds of microns in dimension. This is partly due to the absence of thin superconducting Sr_2RuO_4 films. The domains are expected to be no more than a few microns in size [5]. Moreover, while the domains are expected to be pinned to random defects in the crystal, they also appear to be easily displaced under the influence of an applied current or magnetic field [6]. The arbitrary configuration of the domains in bulk systems introduces an element of uncertainty, which can be problematic when probing the local order parameter to demonstrate the spatial segregation of chiral states. This is where mesoscopic structures can provide a solution. It is known that the energy cost associated with a chiral domain wall, grows per area [7]. Hence, a domain wall would favour the most constricted parts of a given structure to reduce its energy. The situation is somewhat analogous to the magnetic domains inside a ferromagnet, where geometrical restrictions (e.g. a notch in a ferromagnetic wire), can be used as an effective mechanism for pinning the domain walls by lowering the free energy. This is a widely popular practice in spintronics and novel magnetic memory devices. This concept however has not been explored in the context of superconducting domains. This can partly be attributed to the material properties of Sr_2RuO_4, which put severe constraints on the fabrication of mesoscopic structures. This is also reflected in the fact that, while there have been a substantial number of experiments on Sr_2RuO_4 for over two decades, no more than a handful have examined mesoscopic structures.

This lack of studies on microstructures is not unique to Sr_2RuO_4. There is a growing family of exotic correlated electron systems which suffer the same drawback, as they can currently be prepared only as bulk-like crystals, due to their sensitivity to disorder. Many of these materials have highly unconventional magnetic and transport properties, which currently cannot be described by any existing theory. Understanding the mechanisms behind such correlated electron systems is one of the principal challenges of modern condensed matter physics. Here, we tackle this issue by utilizing a Ga^+ focused ion beam (FIB) to prepare mesoscopic structures out of bulk crystals of Sr_2RuO_4. The method can be described as "sculpting" the desired structure by shooting ions at a target to sputter away (or mill) the surrounding material. This provides a highly precise and versatile nanostructuring technique, and is implemented as the principal fabrication method throughout this work. In case of S-F devices, the use of exceptionally small ion currents (down to 1 pA) together with the spot-size of a carefully focused beam provided the means to obtain well-defined nanostructures, with the smallest features reaching below 20 nm (shown in Figure 1.2 **a**). As for Sr_2RuO_4, FIB enables us to cut through crystals that otherwise would be too thick to structure using conventional lithography and etching techniques (Figure 1.2 **b**). Furthermore, the arbitrary shape and dimensions of a crystal could carefully be accounted for while the milling took place. This allowed for precise adjustments to the sample design based on the unique structure of individual crystals.

OUTLINE OF THE THESIS

- **Chapter** 2 (*Pairing symmetry*) begins with the general symmetry classes for Cooper pairs, with an emphasis on spin-triplet pairing. The discussion is then directed towards Sr_2RuO_4. By introducing the d-vector formalism, this chapter continues to describe possible pairing symmetries for Sr_2RuO_4. The likelihood of each case is evaluated as we review a number of key experiments.

- **Chapter** 3 (*Triplet Cooper pairs in magnetic hybrids*) is related to odd-frequency (even-parity) triplet correlations. The first section introduces the concept of long-range proximity effect, and addresses the challenges in utilising it in functional devices. The next section describes how we tackle these issues with the use of micromagnetic simulations. An example of this is provided in the last section, where we describe the magnetic patterns of CrO_2 nanowires, which we then implement to generate long-range triplet currents.

- **Chapter** 4 (*Controlling supercurrent and their spatial distribution in ferromagnets*) demonstrates how micromagnetic simulations can be used to control the path of spin-triplet supercurrents in a magnetic multilayer. This is realised in a disk-shaped planar Josephson junction with a Ni/Co/Ni barrier, where Co and Ni layers can have non-collinear magnetizations.

- **Chapter 5** (*Generating spin-triplet supercurrents with a ferromagnetic vortex*). Here we show that the magnetic pattern of a *single* ferromagnet can be implemented to generate and control long-range triplet currents. We also examine the *phase* of the triplet channels formed by a ferromagnetic vortex, and show that displacing the vortex core can produce widely different transport behaviours in the same device.

- **Chapter 6** (*Little-Parks oscillations with half-quantum fluxoid features in Sr_2RuO_4 micro rings*) is concerned with the half-quantum vortex in Sr_2RuO_4, and its possible signatures in magnetotransport measurements.

- **Chapter 7** (*Spontaneous emergence of Josephson junctions in single-crystal Sr_2RuO_4*) focuses on the behaviour of a *single* chiral domain wall, which is predicted to act as an unconventional Josephson junction. We investigate this using mesoscopic rings, structured entirely out of a single Sr_2RuO_4 crystal. Order parameter simulations predict a domain wall to cross the arms of the ring, forming a pair of parallel Josephson junctions. Our transport measurements show a clear critical current oscillation, similar to that of a DC SQUID with two symmetric junctions. This, together with a detailed analysis of current-voltage behaviour make a compelling case for the presence of a chiral domain wall.

REFERENCES

[1] J. Bardeen, L. N. Cooper, and J. R. Schrieffer. Theory of superconductivity. *Physical Review*, 108(5):1175, 1957.

[2] J. Alicea. New directions in the pursuit of Majorana fermions in solid state systems. *Reports on Progress in Physics*, 75(7):076501, 2012.

[3] N. Read and D. Green. Paired states of fermions in two dimensions with breaking of parity and time-reversal symmetries and the fractional quantum hall effect. *Physical Review B*, 61(15):10267, 2000.

[4] T. Rice and M. Sigrist. Sr_2RuO_4: an electronic analogue of ^{3}He ? *Journal of Physics: Condensed Matter*, 7(47):L643, 1995.

[5] A. Bouhon and M. Sigrist. Influence of the domain walls on the Josephson effect in Sr_2RuO_4. *New Journal of Physics*, 12(4):043031, 2010.

[6] F. Kidwingira, J. Strand, D. Van Harlingen, and Y. Maeno. Dynamical superconducting order parameter domains in Sr_2RuO_4. *Science*, 314(5803), 2006.

[7] M. Sigrist and D. F. Agterberg. The role of domain walls on the vortex creep dynamics in unconventional superconductors. *Progress of Theoretical Physics*, 102(5):965–981, 1999.