Towards superconducting spintronics with RuO2 and CrO2 nanowires

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Introduction

Traditional electronic devices depend on the transport of electric charge carriers, specifically electrons, within a semiconductor such as silicon to perform their functions. Over the past five decades, there has been remarkable progress in semiconductor technology, characterized by significant advancements in performance and continuous efforts towards achieving greater miniaturization. Since the 1970s, there has been a trend observed in which the number of transistors in an integrated circuit (IC) doubles approximately every two years, an empirical observation that is commonly referred to as Moore's law. As the possibilities of silicon-based electronics reach their limits, physicists are actively exploring the potential of utilizing the inherent 'spin' property of electrons, in addition to their charge, to develop a new generation of devices known as 'spintronic' devices.

The term spintronics was first coined as an abbreviation for SPIN TRansport electrONICS. The origins of spintronics may be traced back to a series of discoveries and advancements in solid-state physics and electronics going even back to the 1970s, when Robert Meservey and Peter Tedrow conducted tunneling measurements on junctions between very thin superconducting aluminium films and ferromagnetic nickel films in a high magnetic field [1, 2]. Their experiments revealed a spin-dependent nature of the tunneling current. These experiments marked the first efforts to study spin-dependent electron transport phenomena. In 1975, Michel Julliere added to this groundwork by conducting initial experiments on magnetic tunnel junctions [3]. Another important milestone was the observation of spin-polarized electron injection from a ferromagnetic metal to a normal metal by Johnson and Silsbee in 1985 [4]. These early experiments laid the foundation for the development of spintronics. In 1988, Albert Fert and Peter Grünberg independently discovered Giant Magnetoresistance [5, 6]. In the 1990s, research in the field expanded to include other spin-based phenomena such as tunnel magnetoresistance (TMR) [7–11] and the manipulation of the spin of individual electrons using the technique of spin injection. In 2007, researchers demonstrated efficient electrical spin injection from a ferromagnetic metal contact into silicon, producing a large electron spin polarization in the silicon [12, 13]. These developments led to the creation of new types of devices such as spin valves and magnetic tunnel junctions, which are now widely used in data storage applications such as hard disk drives.

Spintronics offers several advantages over traditional electronics, including energy efficiency, higher storage density, improved durability, versatility etc [14]. There is a variety of spintronic devices that exploit the spin of electrons to store and process information, such as Magnetic Random Access Memory (MRAM), Spin Transfer Torque Random Access Memory (STT-RAM) [15], Spin Hall Effect (SHE) devices [16], and Spin-Orbit Torque (SOT) devices [17]. In recent years, research in spintronics has focused on the development of new materials and devices with suitable magnetic properties, such as a high degree of spin polarization and a high Curie temperature (T_C) — the temperature above which the material ceases to be a ferromagnet - that can be used in a variety of applications beyond data storage, including high-speed computing, energy-efficient electronics, and quantum computing. Of particular interest are magnetic materials that exhibit a very high spin polarization at the Fermi level. Prominent example of such materials include metallic oxides such as CrO_2, $La_{0.3}Sr_{0.7}MnO_3$ (LSMO); and Heusler alloys with the general composition X_2YZ (X = Co, Fe, Y = Mn, Z = Al, Si, Ge, Al, Sb). These Half-metals (HM) are a special type of ferromagnet that display 100% spin polarization due to their band structure, with one spin channel exhibiting metallic behavior due to finite electron density of states at the Fermi level, while the other spin channel behaves as an insulator (or semiconductor) due to an energy gap[18].

While spintronics technology has the potential to revolutionize the field of electronics, there are several challenges that are currently holding back its commercial use. In particular one of the biggest challenges is the large amount of dissipation that is generated by current-driven processes at the nanoscale [19, 20]. To address some of these shortcomings, a new field has emerged which combines spintronics with superconductivity, giving rise to superspintronics. Superconductors are, by nature, dissipationless. We can have dissipationless spin polarized currents which can exploit the intrinsically low switching energies and high switching frequencies of spintronics. Moreover, it is of great interest for the possibility of introducing quantum coherence phenomena in spintronic devices. Traditionally considered competing phenomena, when artificially juxtaposed, a wealth of physics at the interface between superconductors and ferromagnets emerges. Spin-polarised Cooper pairs are capable of surviving inside a ferromagnet over much longer distances than the regular (spin-singlet, anti-parallel) pairs. This new type of Cooper pair is the building block for super-spintronics; leading to a dissipationless spin-current combined with spintronic devices.

The idea of combining superconductivity and ferromagnetism was first discussed in the seminal work in 1956 by Ginzburg. His theoretical analysis revealed that suppression of superconductivity could happen due to so called orbital effect : in the presence of magnetic field, the Lorentz force is exerted differentially on two electrons with opposite spin in Cooper pair. Furthermore, the Zeeman interaction, which arises from the coupling between spins and a magnetic field, promotes a parallel alignment of the spins. This implies that when the magnetic field is sufficiently high, the pairs of electrons become energetically unstable because one electron in the pair will undergo a spin-flip scattering process. Although rare, some unconventional superconductors with coexisting ferromagnetic order have been found like UGe [21], URhGe [22] and UCoGe [23].

The coexistence of superconductivity and ferromagnetism can also be engineered using conventional superconductors (S) and a ferromagnet (F) to create S/F hybrids. Due to the exchange-field E_{ex} of F, the up-spin and down-spin electrons of a Cooper-pair acquire a phase difference upon passing through a S/F interface. Consequently, Cooper pairs arrange themselves in a so called FFLO (Fulde-Ferrel-Larkin-Ovchinnikov) state within F. The coexistence of the spin-singlet $|\uparrow\downarrow - \downarrow\uparrow\rangle$ and spin-triplet components $|\uparrow\downarrow + \downarrow\uparrow\rangle$, both with zero spin projection, is observed. This mixed state gives rise to novel physical phenomena. Nevertheless, it is limited to a few nanometers inside the ferromagnetic material, rendering it somewhat impractical for most applications. The typical length scale over which the superconducting state survives inside the ferromagnet is given by the coherence length (in dirty limit) [1], $\xi_F = \sqrt{\hbar D_F / E_{ex}}$ where, $\hbar$ is the reduced Planck's constant, D_F is the electronic diffusion constant in F and E_{ex} is the exchange energy.

There is a way to overcome this problem of short range superconductivity in the F layer by exploiting anti-symmetrical pairing of the wave function describing the Cooper pair in the time domain. Although a Cooper pair behaves as a boson, its fundamental constituents are fermions, and therefore obey Pauli's exclusion principle. This means that the total wave function, which is a product of the spatial-orbital part, spin part and time (or frequency) has to be antisymmetric under an overall exchange of two electrons. Basically, allowing uncertainty in time as well as in space, and allowing 'negative times' or frequencies, an extra symmetry can be built into the wave function : even in frequency yields conventional s- or d-wave

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[1] in the dirty limit : $\xi_F < l_e$, where l_e is the mean free path of electron

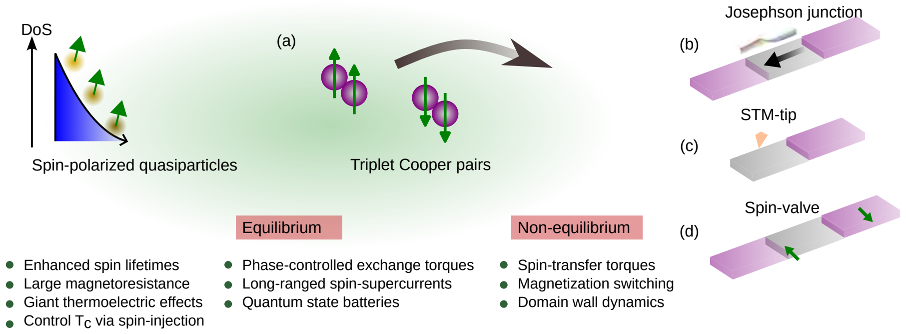

Figure 1.1: (a) Schematic overview of different ways to use superconducting spintronics by means of spin-polarized quasi-particles and triplet Cooper pairs, both in equilibrium and non-equilibrium settings. The fading colour of the quasi-particles in the superconducting region represents their loss of effective charge as they approach the gap edge. (b–d), Schematics for typical experimental set-ups used in superconducting spintronics, including Josephson junctions, bilayers and spin valves. [Taken from ref. [27]].

spin singlets $(|\uparrow\downarrow - \downarrow\uparrow\rangle)$ and p-wave spin triplets $(|\uparrow\downarrow + \downarrow\uparrow\rangle)$; while odd in frequency can yield s-type spin triplets $(|\uparrow\uparrow\rangle, |\downarrow\downarrow\rangle)$ or p-type spin singlets. The p-wave pairing is very sensitive to any external impurities or disorder and hence, is short-range in nature. However, s-type pairing does not suffer from any such limitations. These odd-frequency equal-spin pairing can coexist with a magnetic field as the Zeeman interaction due to the magnetization no longer has a pair-breaking effect. In other words, these triplets and the ensuing supercurrents can have a very long range, mainly determined by the temperature and by spin scattering.

The odd-frequency pairing in a ferromagnet was first outlined in two seminal papers by Bergeret et al. [24] and Kadigrobov et al. [25] in 2001. Crucial in converting s-wave singlet pairs in the superconductor to s-wave triplets in the ferromagnet is the engineering of well-defined magnetic inhomogeneity (the 'generator') at the interface with the superconductor. This was demonstrated experimentally by Keizer et al. on the half-metal CrO_2 films over distances up to 1 μm in 2006 [26]. Since the triplets supercurrents are by definition spin-polarized, they offer great potential for a new kind of superconducting electronics, in which not only the charge and the superconducting phase, but also the spin is utilised. Some of the possible area of applications of superconducting electronics as shown in Fig.1.1 include magnetization switching, magnetization precession, spin-transfer torque, or domain wall motion due to spin-polarized supercurrents; injection of spin-triplet pairs into superconductors, superconducting spin valves; Josephson φ-junction for phase batteries; and Josephson diodes. The major perspective and challenge here is to develop a framework for nonequilibrium transport that can account for dynamic interactions involving spin-triplet pairs and ferromagnetic layers.

1.1. Outline of this thesis

This thesis is concerned with whether and how in particular the half-metal CrO_2, with its full spin polarization, can be utilized as a building block in superspintronics. For this we study the epitaxial growth of CrO_2 and RuO_2 nanowires and investigate their transport properties under the influence of magnetic field. Further, Josephson Junctions devices were fabricated using these wires and it was attempted to find long range supercurrents, in particular in CrO_2. The thesis is structured as follows:

- **Chapter 2** consists of two parts. The first part of this chapter treats the necessary concepts of superconductivity and gives the background on long range spin triplet superconductivity and Josephson physics. In the second part, we introduce the various individual energy contributions that lead to the magnetization state of a ferromagnet, and in competition result in the spontaneous formation of domain walls (DWs) in magnetic materials. Different types of DWs can occur, that are based on the geometry and size. We also introduce the basics of magnetization dynamics through the Landau-Lifshitz-Gilbert equation that describes the dynamic behavior of magnetic moments under the influence of magnetic fields and currents.

- **Chapter 3** describes the selective area growth of CrO_2 nanowires on a TiO_2 substrate along both the substrate *c*-axis (easy axis) and the substrate *b*-axis (hard axis). We investigate the morphology of these nanowires by high-resolution transmission electron microscopy (TEM) and measure their transport properties, in particular magnetoresistance (MR) and the Anomalous Hall Effect (AHE). TEM images show the difference in morphology of the wires grown along the two axes, which is supported by the MR measurements.

- **Chapter 4** focuses on the pulse measurements on CrO_2 nanowires with the purpose of removing a domain wall from an artificially made constriction in the wire. For this we design a high frequency pulse setup and then use Py wires to develop and optimize the setup to study current driven DW motion. Then we demonstrate current induced DW motion in CrO_2 in the same setup. We show that the critical depinning current in CrO_2 is comparable to Py despite the high spin polarization of the former, and show it to be very sensitive to small changes in magnetic field and the dimension of the constriction (a 'notch').

- **Chapter 5** studies the growth of RuO_2 nanowires followed by characterization of these wires through electrical and magnetotransport measurements. Then we focus on making Josephson junctions (JJs) in which superconducting MoGe are contacted on top of RuO_2 nanowires with varying lateral gaps and present the results on these junctions. From the data, we extract the coherence length of the superconducting correlations in the RuO_2 wire.

- **Chapter 6** presents the nanofabrication of Josephson Junctions using CrO_2 nanowires. Two approaches were used to make these devices. We start with using the traditional etching of insulating Cr_2O_3 on CrO_2 surface before depositing tri-layer contact (Ag/Ni/MoGe). The interface transparency is studied systematically by varying the etch times on multiple samples (> 50). Despite multiple attempts, we did not succeed in producing the required high transparency. In the second method, RuO_2 is deposited in-situ with CrO_2 to protect the surface of CrO_2 before depositing Ni/MoGe contacts and the results on these junctions are discussed.

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