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## Towards superconducting spintronics with RuO<sub>2</sub> and CrO<sub>2</sub> nanowires

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# 6

## Josephson Junctions with $\text{CrO}_2$ Nanowires

*Half-metallic systems such as  $\text{CrO}_2$  are an ideal candidate to investigate very long range proximity effects in S/F hybrids. Although  $\text{CrO}_2$  holds great potential for superconducting spintronics, its metastability at ambient temperatures has slowed research. In this work, we have fabricated  $\text{CrO}_2$  junctions with two distinct methods to address this issue. The first method involves removing the  $\text{Cr}_2\text{O}_3$  layer by standard Ar-etching of the top surface of  $\text{CrO}_2$ . We evaluated the impact of etching on interface transparency in over many ( $> 50$ ) devices and observed a very wide spread of interface resistance for the same etch parameters, indicating lack of control and consistency required to observe the desired effects. The second approach employs a protective layer of  $\text{RuO}_2$  which was grown in situ with  $\text{CrO}_2$  in CVD with customized arrangements. The  $\text{RuO}_2$  layer results in low contact resistances of around  $1 \Omega$ . However, we found the thickness of  $\text{RuO}_2$  to be above  $50 \text{ nm}$  for a very short growth time of  $5 \text{ sec}$ . Due to its short coherence length of  $12 \text{ nm}$ , the considerable thickness of  $\text{RuO}_2$  prevents the long range proximity effect in these devices. Further optimization of growth settings is needed to attain the target thickness of around  $5 \text{ nm}$ .*

## 6.1. Introduction

The interplay between conventional superconductivity and ferromagnetism in superconductor-ferromagnet (S/F) hybrid structures produces a novel form of equal spin triplet superconducting correlations [1]. The triplets, denoted as  $|\uparrow\uparrow\rangle$  and  $|\downarrow\downarrow\rangle$  in Dirac notation, have parallel spins. Consequently, they are less affected by the pair breaking due to exchange field in the ferromagnet. This facilitates the propagation of triplet pairs over significantly greater distances within the F layer [2]. The phenomenon known as long range proximity (LRP) effect was first proposed by Bergeret et al. in 2001. Since then, in the last two decades, this field has been studied extensively [3–15], primarily focusing on generation and control of superconducting triplet correlations. The key ingredient in conversion from singlet-to-triplet is the engineering of magnetic inhomogeneity through spin mixing and spin rotation at the S/F interface. Experimentally, magnetic inhomogeneity can be introduced through vertical multilayer stacks with different magnetic materials and magnetization directions (for eg. S/F<sub>0</sub>/F/F<sub>0</sub>/S) [16, 17] or magnetic textures like ferromagnetic domain walls [1, 18–20] or interfaces with magnetic disorder [21, 22]. Triplet supercurrents are by definition spin-polarized, and they bear great promise 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 potential applications include : use of the supercurrent to generate a spin torque on a nanomagnet and bring its magnetization into motion which can lead to high-frequency oscillators and cryogenic memories; use of the supercurrent to move domain walls; Josephson  $\varphi_0$ -junction-based phase batteries to provide phase shifts in superconducting circuits [23]; or Josephson diodes where the superconducting critical current of the junction depends on the direction of the current [24].

Half-metallic ferromagnets (HMF) like CrO<sub>2</sub> are particularly interesting due to their ability to sustain remarkably high supercurrents (of the order of  $10^{10}$  A/m<sup>2</sup>) over hundreds of nanometers [3, 12, 14]. This is in sharp contrast to conventional ferromagnets like Ni, Co, and Fe, which typically exhibit supercurrents over much shorter distances, in the range of a few tens of nanometers. In 2006, Keizer et. al. first reported spin triplet supercurrent in a Josephson Junction (JJ) device comprising two s-wave superconducting electrodes of NbTiN separated by distances of up to 1  $\mu\text{m}$  on a 100 nm thick film of CrO<sub>2</sub> grown on a TiO<sub>2</sub> substrate (see Fig. 6.1 (b)). However, they found large spread in the critical currents for different devices which suggested that the mechanism responsible for the singlet to triplet conversion was not very clear. In particular a quantitative analysis, comparing the critical current of HMF-based Josephson junctions with the existing theoretical framework [21], was not feasible. In 2010, Anwar *et al.* reported similar results on CrO<sub>2</sub> films grown on Al<sub>2</sub>O<sub>3</sub> substrates with MoGe as superconducting contacts. These devices again had

no built-in triplet generator, and the reproducibility was poor (less than 30%) [12]. Later in 2012, Anwar et al. observed long range supercurrents on  $\text{CrO}_2$  deposited on  $\text{TiO}_2$  substrates. A thin sandwich layer of Ni (2 nm)/Cu (5 nm) was used between  $\text{CrO}_2$  film and the superconducting MoGe to introduce the necessary magnetic inhomogeneity for spin triplet generation.

The above three work were conducted on ‘full-film’ devices which suffers from a significant limitation due to presence of grain boundaries and ill-defined current paths and an insufficient understanding of the magnetization state at the local level. Fig. 6.1(c) (top) shows the AFM image of the morphology of a  $\text{CrO}_2$  film in which the grain boundaries are clearly visible; while Fig. 6.1(c)(bottom) shows its corresponding MFM image. The contrast in the MFM image is due to stray field and indicates that the magnetic configuration over the film is not homogeneous. This may result in poorly controlled magnetic inhomogeneity at the S/F interface which is crucial for triplet generation. Fig. 6.1(d) shows an  $R(T)$  plot for two different junctions based on a  $\text{CrO}_2$  wire obtained by Ar etching the full film. One of the junction proximizes while the other junction does not, which suggests poor reproducibility of these junctions.

In 2016, Singh et. al. reported SA-grown  $\text{CrO}_2$  nanowires based lateral JJs. A sandwich layer of Cu (or Ag)/Ni at the interface of  $\text{CrO}_2$  and MoGe was used to provide the necessary magnetic non-collinearity. The junctions could sustain supercurrents of the order of  $10^9 \text{ A/m}^2$  below 5 K for a junction length of 500 nm [25]. The potential of  $\text{CrO}_2$  for superconducting spintronics is evident; however, advancements in this area have been hindered due to the metastability of  $\text{CrO}_2$  at room temperature.  $\text{CrO}_2$  reduces into insulating  $\text{Cr}_2\text{O}_3$  (see Sec. 6.2 for details) resulting in a poorly controlled S-F interface transparency which is critical for generation of spin triplets. In this study, we have fabricated junction devices with  $\text{CrO}_2$  nanowires using two distinct methods to address this issue. The first method employs Ar-etching of the top surface of  $\text{CrO}_2$  to remove  $\text{Cr}_2\text{O}_3$  before depositing the contacts. We have systematically analyzed the effects of etching on the interface transparency between  $\text{CrO}_2$  and the contact electrodes comprising Ag/Ni/MoGe trilayer stack by varying the etching time. Similar to the initial studies and contrary to report by Singh *et. al.* we found a lack of reproducibility across multiple measurements for the same growth and etch parameters. In the second method a thin layer of  $\text{RuO}_2$  is grown in situ on  $\text{CrO}_2$  before contacting the wires. This method demonstrates initial potential wherein very high transparency of interface is achieved. However, the growth of the  $\text{RuO}_2$  layer, specifically its thickness, needs further optimization to observe LRP effects in the devices. In the following sections both methods of junction fabrication are discussed and the results on them presented.

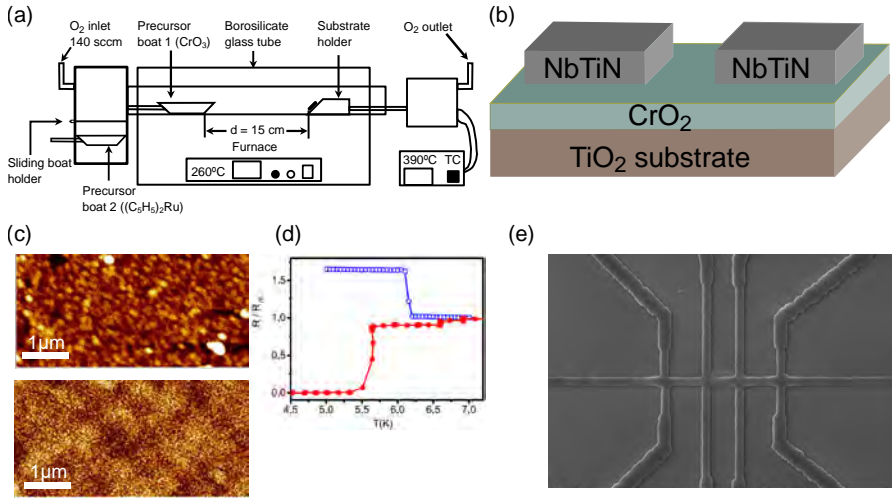


Figure 6.1: (a) Schematic of the CVD setup used for growth of CrO<sub>2</sub>. The same setup allows for in-situ deposition of a RuO<sub>2</sub> layer on top of CrO<sub>2</sub>. To grow RuO<sub>2</sub>, precursor boat1 containing CrO<sub>3</sub> is pulled back out of the glass tube and can be placed in the sealed chamber; and precursor boat2 containing ruthenocene (C<sub>5</sub>H<sub>3</sub>)<sub>2</sub>Ru is introduced in the glass tube. The temperature of the furnace when growing RuO<sub>2</sub> is kept at 80 °C. The substrate temperature is maintained at 390 °C for whole the duration of growth. (b) Schematic of the SFS junction fabricated by depositing superconducting NbTiN electrodes over CrO<sub>2</sub> film on a TiO<sub>2</sub> substrate with lateral gaps of the order of micron. Adapted from Ref. [3]. (c) Atomic force microscopy (AFM) image (top) and magnetic force microscopy (MFM) image (bottom) of a CrO<sub>2</sub> film. The contrast in the MFM image is due to stray fields and shows that magnetic configuration of the CrO<sub>2</sub> film is random, which may result in ill-defined magnetic inhomogeneity at the S-F interface. (d) Resistance vs temperature plot for two junctions based on a CrO<sub>2</sub> wire, obtained by Ar etching the full film, shows the poor reproducibility of these junctions. The success rate of CrO<sub>2</sub>-film junctions was less than 10%. Taken from Ref. [25]. (e) SEM image of a selective area grown CrO<sub>2</sub> nanowire. Making junctions based on SA-grown nanowires should result in more control over the local magnetization due to shape anisotropy. \*Schematic not drawn to scale.

## 6.2. Experimental details

As mentioned above, CrO<sub>2</sub> is metastable at ambient conditions and decomposes to a more thermodynamically stable Cr<sub>2</sub>O<sub>3</sub> state, which is antiferromagnetic and an insulator [26, 27]. Cheng et al. used photoemission and inverse photoemission to characterize the thickness of Cr<sub>2</sub>O<sub>3</sub> layer and they found it to be much thicker than the oxygen core level photoelectron mean free path (around 2 nm) for organometallic chemical vapor deposition grown CrO<sub>2</sub>. This insulating barrier prevents developing a good metallic contact on CrO<sub>2</sub> and complicates the fabrication process. From angle resolved x-ray photoemission (ARXPS) and ultraviolet photoemission experiments, it is known that the deposition of transition metal on CrO<sub>2</sub> surface leads to further oxidation and reduction, making the metal-CrO<sub>2</sub>

interface very inhomogeneous [28].

Since  $\text{CrO}_2$  is metastable, conventional methods for film growth such as sputtering, pulsed laser deposition, or molecular-beam epitaxy cannot be used. Instead chemical-vapor deposition (CVD) has to be employed which prevents in-situ deposition of a protective metal layer over  $\text{CrO}_2$ . Furthermore, cool-down in the CVD furnace is already an uncontrolled event that may start the  $\text{Cr}_2\text{O}_3$  conversion. In this work,  $\text{CrO}_2$  based lateral S–F–S Josephson junctions (JJs) devices were fabricated using two different approaches to work around the  $\text{Cr}_2\text{O}_3$  layer problem, which will be discussed in the following sections. Both methods start with the growth of epitaxial  $\text{CrO}_2$  wires along [001] direction on a  $\text{TiO}_2$  (100) substrate using the Selective Area growth technique via chemical vapor deposition (CVD). Schematic of the CVD set up is shown in Fig. 6.1 (a). and details of the growth process has been described in Chapter 3.2.

For the first method (see Fig. 6.2(a)),  $\text{CrO}_2$  wires were grown followed by patterning desired contacts using e-beam lithography. The top surface of  $\text{CrO}_2$  was then etched using an argon (Ar) plasma to remove the  $\text{Cr}_2\text{O}_3$  layer prior to contacts deposition. The tri-layer contacts comprising Ag (5 nm), Ni (2 nm) and the superconductor  $\text{Mo}_{70}\text{Ge}_{30}$  (150 nm; for simplicity called MoGe) were sputter deposited at a pressure of  $5 \times 10^{-3}$  mbar. This was followed by a lift-off process. The etching was carried out in the same sputtering system without breaking the vacuum by reversing the polarity of the plasma, using a pressure of  $5 \times 10^{-3}$  mbar and a bias voltage of 700 V. The Ag (or Cu) acts as a spacer layer and is used to decouple the two ferromagnets. Ni acts as a mixer layer and is used to achieve magnetic inhomogeneity at the interface. By ensuring Ni magnetization perpendicular to the  $\text{CrO}_2$  wire, maximum possible magnetic non-collinearity at the S-F interface can be achieved, resulting in an efficient triplet Cooper pair generation.

In the second approach, Ag was substituted with a layer of  $\text{RuO}_2$  which is a weak antiferromagnet and a metal.  $\text{RuO}_2$  is inert when exposed to typical atmospheric conditions, and can be deposited epitaxially on top of  $\text{CrO}_2$  (see Fig. 6.1 (a)) due to their similar lattice parameters. To grow  $\text{RuO}_2$  specific arrangements were made in the CVD set up to accommodate the two precursors boats which can be introduced into the glass tube sequentially without affecting the other precursor using the sliding holder. Initially,  $\text{CrO}_2$  is grown as shown in Fig. 6.1 using the precursor boat1 in the glass tube. The furnace temperature is set at 260 °C. After the growth of  $\text{CrO}_2$  is finished, precursor boat1 containing  $\text{CrO}_3$  is pulled back out of the glass tube and can be placed in a closed chamber by shifting the sliding holder up. The temperature of the furnace is brought down to 80 °C. After that, precursor boat2 containing ruthenocene is placed in the glass tube for  $\text{RuO}_2$  growth for around 5 sec before boat2 is also removed from the glass tube. The temperature of substrate holder is

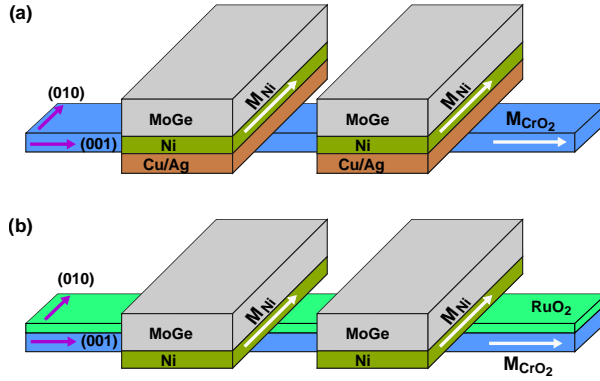


Figure 6.2: Schematic representation of the two different approaches used for fabrication of  $\text{CrO}_2$ -based lateral S–F–S Josephson junctions (JJs). White arrows indicate the magnetization direction of  $\text{CrO}_2$  ( $M_{\text{CrO}_2}$ ) and Ni ( $M_{\text{Ni}}$ ) while purple arrows show the crystal axis. (a) In first approach, the central  $\text{CrO}_2$  wire (blue) is first etched to remove the insulating oxide layer of  $\text{Cr}_2\text{O}_3$  and then tri-layer contacts comprising Cu or Ag/Ni/MoGe were deposited to make the device. (b) In the second approach, a thin layer of  $\text{RuO}_2$  was grown in situ on top of  $\text{CrO}_2$  wire and then bi-layer Ni/MoGe contacts were deposited without the need of etching. \*Schematic not drawn to scale.

maintained at  $390^\circ\text{C}$  for the whole duration of growth.  $\text{RuO}_2$  is inert at ambient conditions, so it prevents the formation of  $\text{Cr}_2\text{O}_3$  and protects the  $\text{CrO}_2$  layer, thus eliminating the need for further etching. Next, the contacts were patterned, followed by deposition and lift-off of bi-layer of Ni(2 nm)/MoGe(200 nm).

The fabrication process was followed by the characterization of the device. Initially, two-probe measurements were done at room temperature to check the contact leads, followed by a four-point connection (Current leads: 9-7, Voltage leads: 11-10) as shown in Fig. 6.3(b) (inset) that allowed a direct measurement of the central  $\text{CrO}_2$  wire resistance without the contact wire resistance. Further, electrical and magnetotransport measurements were performed in a Physical Property Measurement System (PPMS) that could be cooled down to a base temperature of 2 K. The magnetic field could be applied along z-axis up to 9 T. We used a Keithley model 6221 low Noise Precision AC/DC Current Source to provide dc current and a Keithley model 2182A digital nanovoltmeter to measure the voltage.

### 6.3. $\text{CrO}_2$ junctions fabricated by etching process

It is by now well established that a thin insulating layer of  $\text{Cr}_2\text{O}_3$  forms on top of the  $\text{CrO}_2$  surface exposed to atmospheric conditions. To get a comprehensive understanding of the impact of such a barrier on  $\text{CrO}_2$  junctions, our initial emphasis

### 6.3. CrO<sub>2</sub> junctions fabricated by etching process

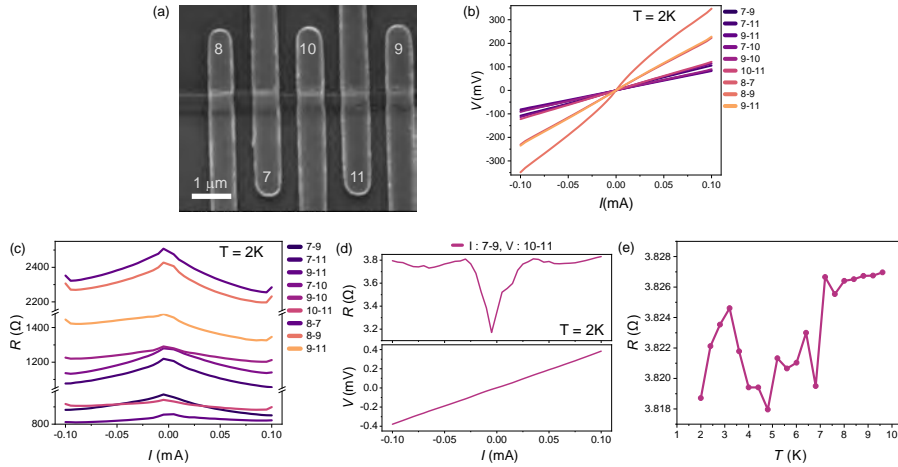


Figure 6.3: No-etch CrO<sub>2</sub> junctions. (a) SEM image of the device consisting of CrO<sub>2</sub> wire of width 370 nm along [001] and multiple contact leads comprising a Ag/Ni/MoGe stack across the CrO<sub>2</sub>. The contact leads have edge to edge separation of around 405 nm. (b) Two-point voltage vs current ( $I$ - $V$ ) plots for different contact configurations on device B measured at 2 K. (c) The corresponding 2 probe resistance as a function of current ( $R$ - $I$ ). (d) Four-point  $I$ - $V$  and the corresponding  $R$ - $I$  plot of the central CrO<sub>2</sub> wire. (e) Four-point  $R(T)$  measurement between 10 K and 2 K of the central CrO<sub>2</sub> wire between contacts 10-11. A small drop in resistance is observed around  $T_c$  of MoGe; after that it oscillates to finally reach 3.818  $\Omega$  at 2 K, without a visible proximity effect.

was on a device without etching. This would help with the basic characterization of the device and thereafter serve as a reference point for correlating with the effect of etching of the CrO<sub>2</sub> surface. Subsequently, a series of devices was fabricated with varying etch times in order to determine the optimal time required for our devices. Etching plays a critical role in achieving a good metallic interface between CrO<sub>2</sub> and Ag/Ni/MoGe contact leads. The outcome of underetching is evident as it would result in partial removal of Cr<sub>2</sub>O<sub>3</sub> layer. Overetching is also detrimental as it would damage the underlying CrO<sub>2</sub> surface. It should be feasible to manipulate the interface transparency by varying the duration of etching in a systematic manner and identify an appropriate range of etch time for the fabrication of proximized CrO<sub>2</sub> JJs.

#### 6.3.1. Device without etching

In this section, we present the results of measurements conducted on CrO<sub>2</sub> junction device fabricated without etching step. Fig. 6.3(a) shows the scanning electron micrograph of a fabricated junction device consisting of a central CrO<sub>2</sub> wire of width around 370 nm along [001] and multiple contact leads comprising Ag/Ni/MoGe



stack across the CrO<sub>2</sub> wire. The contact leads have lateral edge to edge separation of around 405 nm. To characterize the interface between CrO<sub>2</sub> and contact leads, the device was measured in a 2-probe configuration. Fig. 6.3(b) shows the two-probe voltage vs current ( $I$ - $V$ ) plots for different contact lead configurations, with the contacts labeled in the SEM image. The measurement (e.g. 7-8 meaning from contact 7 to contact 8) gives the combined resistance of two interfaces. The measurements were at 2 K, which is well below the superconducting transition temperature ( $T_c$ ) of 6.5 K for MoGe. At 2 K, the measured resistance should have most of the contribution from the interface resistance. While certain  $I$ - $V$  curves may appear linear, leading one to assume an ohmic nature of the interfaces, the non-ohmic behavior becomes apparent in the resistance vs current plot (see Fig. 6.3(c)) which indicates that the interfaces are indeed not metallic and the CrO<sub>2</sub> layer is effectively decoupled from the contact leads. Each measurement shows a high resistance of the interfaces, the minimum being above 800  $\Omega$  and maximum around 2400  $\Omega$ . This suggests local variation among the five interfaces on the same device. We also measured the behavior of the central CrO<sub>2</sub> wire without the contribution coming from contact leads in 4-point geometry. Fig. 6.3(d) shows the  $I$ - $V$  plot (bottom) and the corresponding  $R$ - $I$  plot of CrO<sub>2</sub> wire between inner contact leads. The resistance was found to be around 3.8  $\Omega$  at 2 K which is typical for CrO<sub>2</sub> wires of such dimensions. However, the  $R$ - $I$  plot shows non-ohmic type behavior of the wire, the underlying cause of which remains unclear to us. Finally, we measured 4-point geometry resistance vs temperature measurement on a junction device between 10 K and 2 K as shown in Fig. 6.3(e). The resistance at 10 K is around 3.827  $\Omega$ . A small drop in resistance is observed around  $T_c$  of MoGe but after that it oscillates to finally reach 3.818  $\Omega$  at 2 K and as anticipated, no proximity effect was detected.

### 6.3.2. Device with etching

After measuring devices without etching, we made a series of devices with different etching times in our quest to find the optimal range. Fig. 6.4(a) shows the resistance vs etch time plot of different devices, measured in a 4-point geometry at 2 K, as the etching time is varied from 0 s up to 90 s. We fabricated multiple devices with the same etch time and in the Figure we chose a random device from the series to represent an etch time. We observed a non-monotonous behavior up to 40 sec of etching beyond which the resistance goes up sharply, suggesting that we are overetching and probably damaging the CrO<sub>2</sub> wire. We found the best results on our devices around 20-25 sec of etch time, but no clear window of the etch times was observed which led to proximization of the CrO<sub>2</sub> junction. This result is contrary to the earlier report by Singh et al. where they found a well defined etching time to see proximity effect [25].

### 6.3. CrO<sub>2</sub> junctions fabricated by etching process

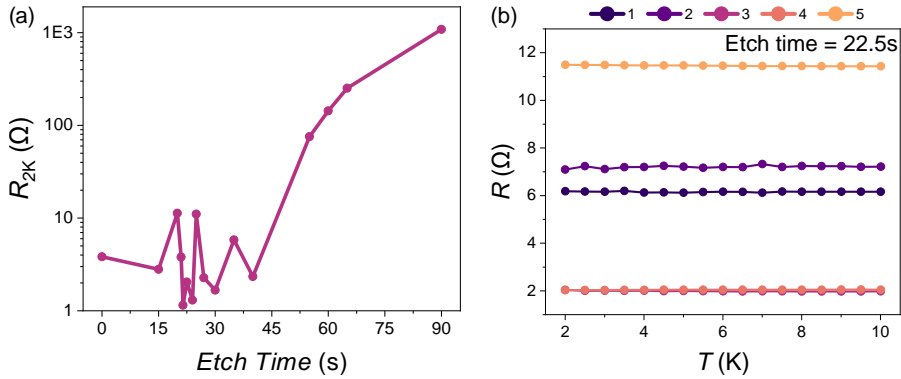


Figure 6.4: (a) Resistance vs etch time plot of different devices measured at 2 K as the etch time is varied from 0 s up to 90 s. Multiple devices were measured for the same etch times, out of which we chose one of the device randomly to be represented in the graph. (b) Four-point resistance vs temperature of CrO<sub>2</sub> wire for five different devices etched for same time of 22.5 s, showing small differences in the measured resistance.

We observed a large spread in interface resistances for the same etching time not only across various devices but even on the same CrO<sub>2</sub> wire. We found interface resistances to be at their lowest for etch time around 20-25 sec. Around this time, the observed interface resistance ranged from as low as 20  $\Omega$  to as high as 10 K $\Omega$  (not shown in figure). Although the interfaces showed large variations in resistances, we found the 4-point geometry wire resistance (without interfaces) to be consistent. This indicates that the quality of CrO<sub>2</sub> wire is not a factor. Fig. 6.4(b) gives the four-point resistance vs temperature of CrO<sub>2</sub> wire in five different devices etched for same time of 22.5 s. These devices show small spread in the measured resistance which is probably due to small difference in the device dimensions, however none of them showed a proximity effect.

In Fig.6.5, we present the measurements of one of the CrO<sub>2</sub> junction device which was etched for 22.5 s. Fig.6.5 (a) shows the SEM image of the device comprising CrO<sub>2</sub> wire of width ( $w$ ) = 512 nm, thickness ( $t$ ) = 65 nm and multiple Ag/Ni/MoGe contact leads. The two inner contact leads are edge to edge separated by 350 nm. Fig.6.5 (b) gives the 2-point  $I$ - $V$  (top) and corresponding  $R$ - $I$  plots of the contact leads (without interface) measured at 10 K, well above  $T_c$ .  $R$ - $I$  plots confirm that the contact leads are ohmic and indeed metallic. Subsequently, we measured the device in the quasi 4-point geometry configuration. This configuration allows to measure the resistance of CrO<sub>2</sub> wire along the two interface between CrO<sub>2</sub> and Ag/Ni/MoGe on either side of the central CrO<sub>2</sub> wire (see Fig. 6.2). Fig. 6.5(c,d) show the  $I$ - $V$  and corresponding  $R$ - $I$  plots in different combinations measured at 2 K. We calculated the interface resistance at each of the four interfaces. Moving from left to right,

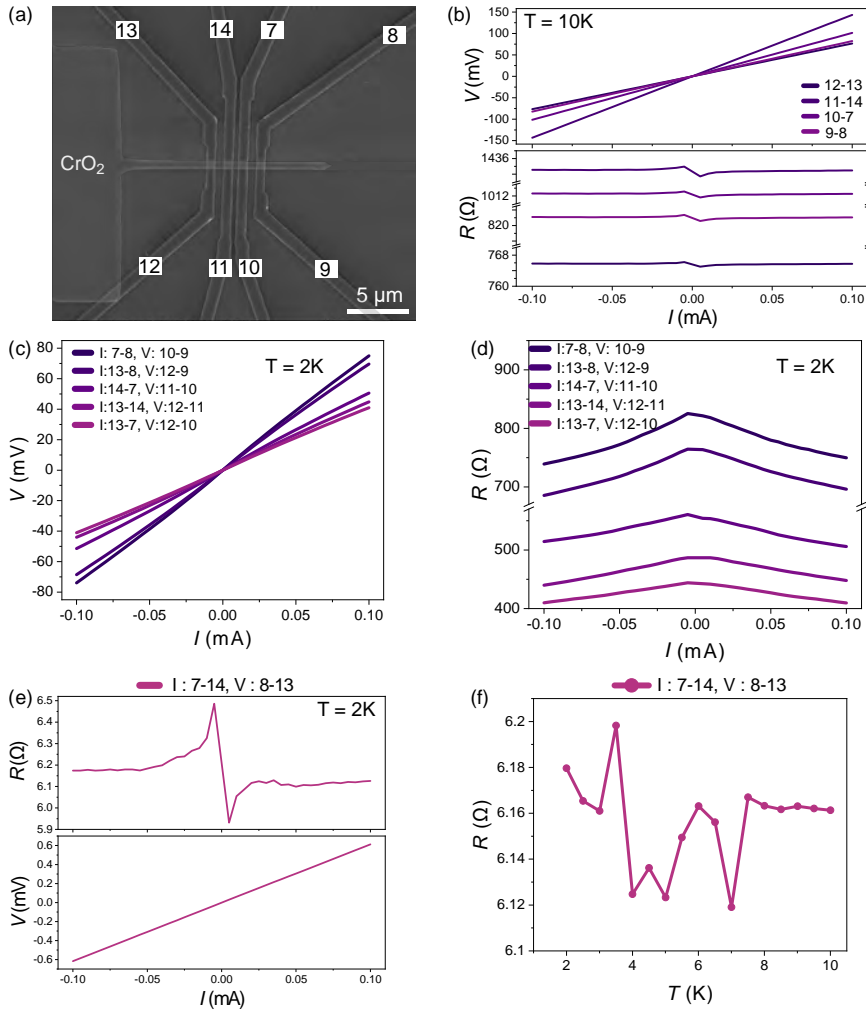


Figure 6.5: SEM image (false color) of one of the CrO<sub>2</sub> junctions fabricated using the first approach of etching the top surface of CrO<sub>2</sub> before depositing tri-layer (Ag/Ni/MoGe) contacts. The two inner contacts are laterally edge-to-edge separated by  $\sim 350$  nm. This device was etched for 22.5 s. (b) 2-point *I-V* and corresponding *R-I* plots of the contact leads (without interface) measured at 10 K before  $T_C$ . *R-I* shows good ohmic behavior. (c) Quasi 4-point *I-V* and the corresponding (d) *R-I* plot of the central CrO<sub>2</sub> wire and the two interfaces in different configurations measured at 2 K. The interfaces have resistances varying between 400 Ω and 800 Ω. (e) 4-point *I-V* and the corresponding *R-I* plot of the central CrO<sub>2</sub> wire without interface. (f) Resistance vs temperature plot of the device between 10 K and 2 K shows the resistance fluctuating around  $\sim 6.16$  Ω.

the resistances were roughly 180 Ω, 300 Ω, 250 Ω and 570 Ω, respectively. We can also calculate the resistance-area product (RA) for these interfaces. From the cross

sectional area of  $512 \times 65 \text{ nm}^2$  and interface resistance of  $180 \text{ } \Omega$ , we find an RA of  $0.6 \times 10^{-13} \text{ } \Omega \cdot \text{m}^2$ . Similar to the observation on earlier devices, we found a large local variation in resistance among each of the interfaces.

Next, we measured the 4-point  $I$ - $V$  and  $R$ - $I$  curves (see Fig. 6.5(d)) for the central CrO<sub>2</sub> wire at 2 K. We found that the wire showed typical ohmic characteristics. Finally, we took the resistance vs temperature measurement of the CrO<sub>2</sub> wire between 10 K and 2 K. As expected, a proximity effect was not observed. Instead we found that the resistance fluctuated around  $6 \text{ } \Omega$  after the superconducting transition at 6.5 K. Similar to the above junction device, we measured many ( $> 50$ ) devices but the interface transparency issue remained unresolved. The earlier studies on proximity effect reported the interface resistance to be around  $20 \text{ } \Omega$  [25]. However, we were not able to fabricate a device with such a low contact resistance, mostly because of the above-mentioned local variation in resistance.

Based on the above observations, we can certainly conclude that the large heterogeneity in resistance not only between the devices etched from same duration but also in the interface resistances of contact leads and CrO<sub>2</sub> wire separated by few hundreds of nm within a single device, limits the precise control required to fabricate a device with a defined resistance. Additionally, the growth of CrO<sub>2</sub> wires based on chemical vapor deposition limits the control of the final shape of the wire in the sense that the top surface of wire is not very flat. Instead we found surface roughness of about 10 nm along the width of the wire when measuring the topography with atomic force microscopy (see Appendix Fig. 6.7). This roughness in turn hinders with control of the etching process of CrO<sub>2</sub> and reliable metallic connection of contact electrodes with high interface transparency, a critical requirement for these junctions.

## 6.4. CrO<sub>2</sub> junctions fabricated with RuO<sub>2</sub>

The lack of control with etching and difficulty to achieve a low interface resistance on our junctions required us to find another approach of fabrication. This is where RuO<sub>2</sub> emerged as a choice of material. The main advantage is that it is a metal, inert in ambient conditions and it can also be grown epitaxially in situ with CrO<sub>2</sub> via CVD due to similar lattice parameters. Further details of growth of RuO<sub>2</sub> can be found in sec. 5.2 In this section, we present the results of measurements conducted on one of the CrO<sub>2</sub>/RuO<sub>2</sub> junction device.

Fig. 6.6 (a) shows the false colored SEM image of CrO<sub>2</sub>/RuO<sub>2</sub> wire (green) of width around 500 nm and four Ni/MoGe contact leads. The two inner contact leads are laterally edge to edge separated by  $\sim 300 \text{ nm}$ . Next, one of the contact lead,

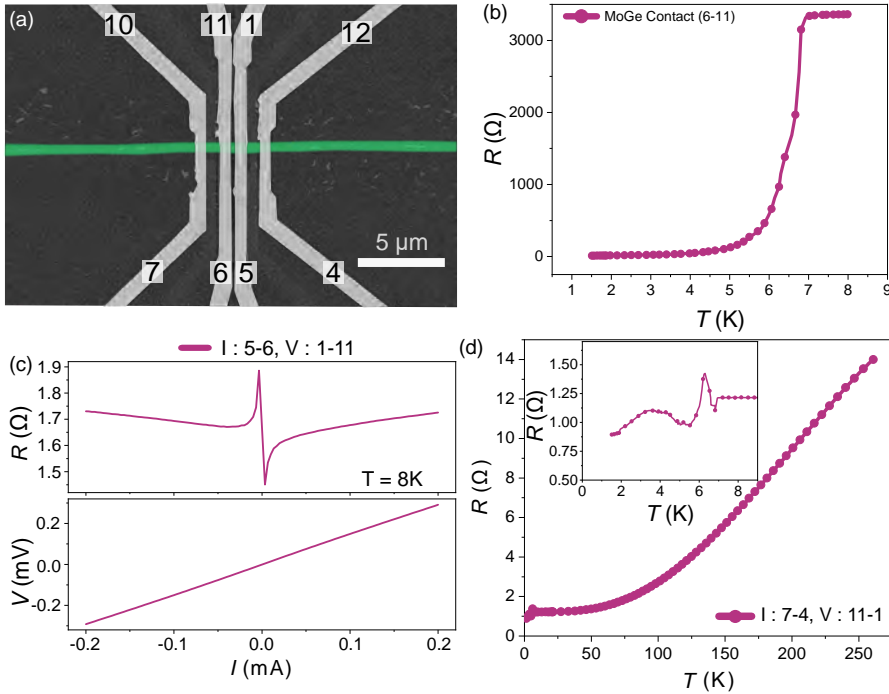


Figure 6.6: (a) SEM image (false color) of a CrO<sub>2</sub>/RuO<sub>2</sub> junction fabricated using the second approach where RuO<sub>2</sub> was grown in situ on top of CrO<sub>2</sub> wire, followed by deposition of contact leads comprising Ni/MoGe. The two inner contacts are laterally edge-to-edge separated by  $\sim 300$  nm. (b) 2-point resistance vs temperature plot of contact lead 6-11 shows a superconducting transition ( $T_c$ ) at  $T_c$  of  $\sim 6.5$  K. (c) Quasi 4-point  $I$ - $V$  and corresponding  $R$ - $I$  plot of the central CrO<sub>2</sub>/RuO<sub>2</sub> wire and the two interfaces measured at 8 K before  $T_c$ . (d)  $R(T)$  plot of the junction measured between 250 K and 1.5 K. The inset shows a small upturn and then a dip in the resistance near  $T_c$  of the contact leads. Below  $T_c$  the resistance fluctuates around 1  $\Omega$  up to 1.5 K.

6-11 was measured in a 2-probe configuration to observe the  $T_c$ . Fig. 6.6 (b) shows typical  $T_c$  of around 6.5 K. After that, we measured  $I$ - $V$  and the corresponding  $R$ - $I$  plot (Fig. 6.6(c)) of the central CrO<sub>2</sub>/RuO<sub>2</sub> wire and the two interfaces at 8 K. We observed a low resistance value of around  $\sim 1.7$   $\Omega$  and a good ohmic behavior from  $R$ - $I$  plot, indicating the transparent nature of the interface. Finally we measured  $R(T)$  of the junction in 4-point geometry between 270 K and 1.5 K (Fig. 6.6(d)). We observed that the resistance of the device became constant below 10 K (inset), with a values of 1.214  $\Omega$  at 7.5 K. When the MoGe contact leads became superconducting at 6.5 K, the resistance started to fall again (after a small dip-peak excursion) but did not reach 0  $\Omega$ , indicating that the CrO<sub>2</sub> wire did not proximize completely over the length of the junction. Instead, we observed that the resistance reduced to 0.953  $\Omega$  at 2 K, which was nearly 20% decrement from the normal state resistance ( $R_N$ ) at

7.5 K. This roughly translates to a proximity length of about 30 nm extending from contact leads on either side.

Further investigation into similar junctions fabricated with RuO<sub>2</sub> on top of CrO<sub>2</sub> revealed that the total height of the central CrO<sub>2</sub>RuO<sub>2</sub> wire stack is around 150 nm to 180 nm. From our experience with growing CrO<sub>2</sub> for similar duration of time we can safely assume that the thickness of the CrO<sub>2</sub> wire would be  $\leq 100$  nm. This would mean that the thickness of RuO<sub>2</sub> is  $\geq 50$  nm. Both CrO<sub>2</sub> and RuO<sub>2</sub> are good metal and have comparable low residual resistance below 10 K. Since the thickness of RuO<sub>2</sub> is quite close to the thickness of the CrO<sub>2</sub> wire, the current would flow through both the materials. However, we know that RuO<sub>2</sub> is a weak antiferromagnet with a short coherence length of around 12 nm (see sec. 5.4). This would hamper the observation of long range proximity effect in these junctions. Ideally, we want to deposit a thin layer ( $\sim 5$  nm) of RuO<sub>2</sub> on top of CrO<sub>2</sub> so that maximum current flows through CrO<sub>2</sub>. We found that the rate of growth of RuO<sub>2</sub> when growing above CrO<sub>2</sub> layer was very high. Even a growth for  $\sim 5$  s leads to a thick layer of RuO<sub>2</sub> around (50-80) nm. We tried to change the growth parameters by changing the temperature, oxygen pressure but we did not get the desired outcome. Further investigation into the growth conditions is required to obtain thin RuO<sub>2</sub> wires.

## 6.5. Summary and Outlook

In summary, CrO<sub>2</sub> based lateral Josephson junctions were fabricated using two different approaches. The first method involved the etching of the top surface of CrO<sub>2</sub> to remove insulating layer of Cr<sub>2</sub>O<sub>3</sub>. Following this, a stack of Ag/Ni/MoGe contacts was deposited. This method of fabrication demanded a significant level of precision and control throughout various stages, posing challenges in achieving the desired outcomes. Significant resistance variations observed across multiple measurements, without any parameter modifications, indicate a lack of reproducibility. This lack of consistency poses challenges in conducting systematic investigations on the junctions.

The second approach of fabrication using RuO<sub>2</sub> as a spacer layer demonstrate initial potential. The measured devices exhibited high degree of transparency in the interfaces, with interface resistance being as low as 1  $\Omega$ . However, the thickness of RuO<sub>2</sub> layer remained a significant concern as it plays a crucial role in the observation of the long-range proximity effect in these devices. Additional research is required to achieve successful control of the growth of thin layer of RuO<sub>2</sub> on top of CrO<sub>2</sub> surface.

## 6.6. Appendix

### 6.6.1. AFM image of CrO<sub>2</sub> wire after CVD growth

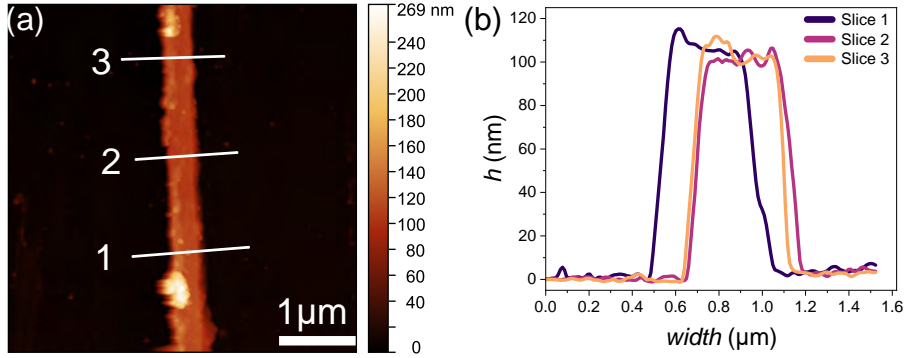


Figure 6.7: (a) Atomic force microscope image of the topography of a CrO<sub>2</sub> wire after CVD growth over an area of  $5 \times 5 \mu\text{m}^2$ . 3 slices were taken at random over a cross-section of the wire. (b) Corresponding height profiles of the 3 slices show the uneven top surface of the wire. The maximum roughness is around  $\sim 14$  nm.

The growth of CrO<sub>2</sub> wires using chemical vapor deposition (CVD) has limitations in controlling the ultimate wire shape, in contrast to alternative deposition processes like as sputtering and evaporation. These procedures allow for layer-by-layer deposition, often resulting in the formation of very smooth and flat films. Fig. 6.7(a) gives the atomic force microscope image of the topography of a CrO<sub>2</sub> wire of width ( $w$ )  $\approx 500$  nm. We took slices at three different positions on the wire (labeled 1, 2 and 3) and found that the top surface of the wire reveals a lack of flatness. Upon examination, a surface roughness of about 10 nm was detected over the width of the wire, as seen in the line profile of the three slices (Figure 6.7(b)). The presence of surface roughness poses a challenge to the precise control of the etching process of CrO<sub>2</sub> and the establishment of reliable metallic connections between contact electrodes, while maintaining a high level of interface transparency. This need is a critical requirement for the successful fabrication of these junctions.

## References

- [1] Bergeret, F. S., Volkov, A. F. & Efetov, K. B. Long-Range Proximity Effects in Superconductor-Ferromagnet Structures. Phys. Rev. Lett. **86**, 4096–4099 (2001). URL <https://link.aps.org/doi/10.1103/PhysRevLett.86.4096>.
- [2] Bergeret, F. S., Volkov, A. F. & Efetov, K. B. Odd triplet superconductivity and related phenomena in superconductor-ferromagnet structures. Rev. Mod. Phys. **77**, 1321–1373 (2005). URL <https://link.aps.org/doi/10.1103/RevModPhys.77.1321>.
- [3] Keizer, R. S. et al. A spin triplet supercurrent through the half-metallic ferromagnet CrO<sub>2</sub>. Nature **439**, 825–827 (2006). URL <https://doi.org/10.1038/nature04499>.
- [4] Khaire, T. S., Khasawneh, M. A., Pratt, W. P. & Birge, N. O. Observation of Spin-Triplet Superconductivity in Co-based Josephson Junctions. Phys. Rev. Lett. **104**, 137002 (2010). URL <https://link.aps.org/doi/10.1103/PhysRevLett.104.137002>.
- [5] Robinson, J. W. A., Witt, J. D. S. & Blamire, M. G. Controlled Injection of Spin-Triplet Supercurrents into a Strong Ferromagnet. Science **329**, 59–61 (2010). URL <https://www.science.org/doi/abs/10.1126/science.1189246>. <https://www.science.org/doi/pdf/10.1126/science.1189246>.
- [6] Robinson, J. W. A., Halász, G. B., Buzdin, A. I. & Blamire, M. G. Enhanced Supercurrents in Josephson Junctions Containing Nonparallel Ferromagnetic Domains. Phys. Rev. Lett. **104**, 207001 (2010). URL <https://link.aps.org/doi/10.1103/PhysRevLett.104.207001>.
- [7] Leksin, P. V. et al. Evidence for Triplet Superconductivity in a Superconductor-Ferromagnet Spin Valve. Phys. Rev. Lett. **109**, 057005 (2012). URL <https://link.aps.org/doi/10.1103/PhysRevLett.109.057005>.
- [8] Witt, J. D. S., Robinson, J. W. A. & Blamire, M. G. Josephson junctions incorporating a conical magnetic holmium interlayer. Phys. Rev. B **85**, 184526 (2012). URL <https://link.aps.org/doi/10.1103/PhysRevB.85.184526>.
- [9] Wang, Y., Pratt, W. P. & Birge, N. O. Area-dependence of spin-triplet supercurrent in ferromagnetic Josephson junctions. Phys. Rev. B **85**, 214522 (2012). URL <https://link.aps.org/doi/10.1103/PhysRevB.85.214522>.
- [10] Robinson, J. W. A., Chiodi, F., Egilmez, M., Halász, G. B. & Blamire, M. G. Supercurrent enhancement in Bloch domain walls. Scientific Reports **2**, 699 (2012). URL <https://doi.org/10.1038/srep00699>.



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- [11] Sprungmann, D., Westerholt, K., Zabel, H., Weides, M. & Kohlstedt, H. Evidence for triplet superconductivity in Josephson junctions with barriers of the ferromagnetic Heusler alloy  $\text{Cu}_2\text{MnAl}$ . *Phys. Rev. B* **82**, 060505 (2010). URL <https://link.aps.org/doi/10.1103/PhysRevB.82.060505>.
- [12] Anwar, M. S., Czeschka, F., Hesselberth, M., Porcu, M. & Aarts, J. Long-range supercurrents through half-metallic ferromagnetic  $\text{CrO}_2$ . *Phys. Rev. B* **82**, 100501 (2010). URL <https://link.aps.org/doi/10.1103/PhysRevB.82.100501>.
- [13] Wang, J. et al. Interplay between superconductivity and ferromagnetism in crystalline nanowires. *Nature Physics* **6**, 389–394 (2010). URL <https://doi.org/10.1038/nphys1621>.
- [14] Anwar, M. S., Veldhorst, M., Brinkman, A. & Aarts, J. Long range supercurrents in ferromagnetic  $\text{CrO}_2$  using a multilayer contact structure. *Applied Physics Letters* **100**, 052602 (2012). URL <https://doi.org/10.1063/1.3681138>.
- [15] Jiang, J. et al. Long-range superconducting proximity effect in nickel nanowires. *Phys. Rev. Res.* **4**, 023133 (2022). URL <https://link.aps.org/doi/10.1103/PhysRevResearch.4.023133>.
- [16] Houzet, M. & Buzdin, A. I. Long range triplet Josephson effect through a ferromagnetic trilayer. *Physical Review B* **76**, 060504 (2007).
- [17] Halterman, K., Valls, O. T. & Barsic, P. H. Induced triplet pairing in clean *s*-wave superconductor/ferromagnet layered structures. *Phys. Rev. B* **77**, 174511 (2008). URL <https://link.aps.org/doi/10.1103/PhysRevB.77.174511>.
- [18] Fominov, Y. V., Volkov, A. F. & Efetov, K. B. Josephson effect due to the long-range odd-frequency triplet superconductivity in SFS junctions with Néel domain walls. *Phys. Rev. B* **75**, 104509 (2007). URL <https://link.aps.org/doi/10.1103/PhysRevB.75.104509>.
- [19] Volkov, A. F. & Efetov, K. B. Odd triplet superconductivity in a superconductor/ferromagnet structure with a narrow domain wall. *Phys. Rev. B* **78**, 024519 (2008). URL <https://link.aps.org/doi/10.1103/PhysRevB.78.024519>.
- [20] Buzdin, A. I., Mel'nikov, A. S. & Pugach, N. G. Domain walls and long-range triplet correlations in sfs Josephson junctions. *Phys. Rev. B* **83**, 144515 (2011). URL <https://link.aps.org/doi/10.1103/PhysRevB.83.144515>.

- [21] Eschrig, M. & Löfwander, T. Triplet supercurrents in clean and disordered half-metallic ferromagnets. *Nature Physics* **4**, 138–143 (2008). URL <https://doi.org/10.1038/nphys831>.
- [22] Linder, J., Yokoyama, T. & Sudbø, A. Theory of superconducting and magnetic proximity effect in s/f structures with inhomogeneous magnetization textures and spin-active interfaces. *Phys. Rev. B* **79**, 054523 (2009). URL <https://link.aps.org/doi/10.1103/PhysRevB.79.054523>.
- [23] Robinson, J. W. A., Samokhvalov, A. V. & Buzdin, A. I. Chirality-controlled spontaneous currents in spin-orbit coupled superconducting rings. *Phys. Rev. B* **99**, 180501 (2019). URL <https://link.aps.org/doi/10.1103/PhysRevB.99.180501>.
- [24] Ando, F. et al. Observation of superconducting diode effect. *Nature* **584**, 373–376 (2020). URL <https://doi.org/10.1038/s41586-020-2590-4>.
- [25] Singh, A., Jansen, C., Lahabi, K. & Aarts, J. High-Quality CrO<sub>2</sub> Nanowires for Dissipation-less Spintronics. *Physical Review X* **6**, 041012 (2016).
- [26] Dai, J. et al. Characterization of the natural barriers of intergranular tunnel junctions: Cr<sub>2</sub>O<sub>3</sub> surface layers on CrO<sub>2</sub> nanoparticles. *Applied Physics Letters* **77**, 2840–2842 (2000). URL <https://doi.org/10.1063/1.1320845>. [https://pubs.aip.org/aip/apl/article-pdf/77/18/2840/10185525/2840\\_1\\_online.pdf](https://pubs.aip.org/aip/apl/article-pdf/77/18/2840/10185525/2840_1_online.pdf).
- [27] Cheng, R. et al. Characterization of the native Cr<sub>2</sub>O<sub>3</sub> oxide surface of CrO<sub>2</sub>. *Applied Physics Letters* **79**, 3122–3124 (2001). URL <https://doi.org/10.1063/1.1416474>. [https://pubs.aip.org/aip/apl/article-pdf/79/19/3122/10190307/3122\\_1\\_online.pdf](https://pubs.aip.org/aip/apl/article-pdf/79/19/3122/10190307/3122_1_online.pdf).
- [28] Cheng, R. et al. Oxidation of metals at the chromium oxide interface. *Applied Physics Letters* **81**, 2109–2111 (2002). URL <https://doi.org/10.1063/1.1506942>.