Characterization of Superconducting Ruthenate Thin Films

Caroline Fedele

2019 PARADIM REU Intern @ Cornell

Intern Affiliation: Physics, University of North Florida

Program: 2019 Platform for the Accelerated Realization, Analysis, and Discovery of Interface Materials Research Experience for Undergraduates Program at Cornell University (PARADIM REU @ Cornell)

PARADIM REU Principal Investigator: Prof. Darrell Schlom, Materials Science and Engineering, Cornell University

PARADIM REU Mentor: Nathaniel Schreiber, Materials Science and Engineering, Cornell University

Primary Source of PARADIM REU Funding: Support for PARADIM is provided under NSF Grant # DMR-1539918 as part of the Materials Innovation Platform Program

Contact: N01259434@unf.edu, schlom@cornell.edu, ns835@cornell.edu

Website: http://cnf.cornell.edu/education/reu/2019

Abstract:

Superconducting materials are useful for a variety of applications, mainly quantum devices and powerful electromagnets. Strontium ruthenate, Sr_2RuO_4, is a complex oxide, which forms in the Ruddlesden-Popper structure. This material superconducts below a critical temperature (T_c) of about 1.5 K, and although this is considered a low T_c superconductor, we are interested in it because of the potentially exotic odd-parity pairing symmetry of the electron Cooper pairs. The proposed odd angular momentum quantum number of Sr_2RuO_4 corresponds to a triplet spin state, where the electron pairs have a combined spin of one (three states), instead of zero (one state) as with even-parity superconductors. With our group's success in optimizing the growth process of Sr_2RuO_4 [1], we decided to pursue another research route. Looking down the periodic table at the next alkali earth metal we aimed to synthesize a new material that will potentially exhibit odd-parity superconductivity as well. The goal of our research has been to synthesize Ba_2RuO_4, which is isostructural and isoelectronic to Sr_2RuO_4 [2]. Ba_2RuO_4 is also a metastable material [3], hexagonal in its most stable state but using epitaxial growth techniques we were able to grow a tetragonal polymorph of the material. Using molecular beam epitaxy, we have successfully grown metallic Ba_2RuO_4 thin films. After growth and initial characterization, we have further characterized the material using X-ray diffraction and electronic measurements, and thus far the results offer promise of a new spin-triplet superconductor.

Summary of Research:

The realization of superconducting Ba_2RuO_4 thin films provides many challenges that were not present in our previous Sr_2RuO_4 research. Not only is it a difficult material to synthesize in the tetragonal phase shown in Figure 1, but there are no reports of it being grown outside of our group and collaborators [2-4], so there is very limited information on its characteristics or best growth conditions. Based on our experience from growing Sr_2RuO_4 superconducting films previously, we suspect that Ba_2RuO_4 thin films will also be very sensitive to defects, meaning that we need a way of developing improved growth conditions to achieve superconducting Ba_2RuO_4.

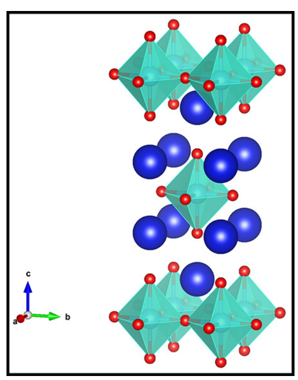

Figure 1: Tetragonal crystal structure of Ba_2RuO_4.

This project revolved around both the growth and characterization of Ba_2RuO_4 thin films, achieved by generating an optimization feedback loop. Using oxide molecular beam epitaxy (MBE) [4], our group grew the films under a variety of growth conditions, where the two main parameters were temperature and the barium-ruthenium flux ratio. Films were then measured using both X-ray diffraction and a helium "dipper" transport measurement system in order to characterize structural and electronic properties. This information was then fed back into the growth process to inform changes in the growth parameters to further optimize the quality of the films. We quantified the film quality using the residual resistivity ratio (*RRR*), which we obtained from the transport measurements and define as $RRR = \rho_{290k}/\rho_{4K}$ — the higher the *RRR*, the higher the quality of the film. The helium dipper transport measurement system uses a four-point probe technique in a square configuration known as the van der Pauw geometry shown in Figure 2.

The van der Pauw theorem for thin films was used to minimize error by allowing us to eliminate contact resistance and strictly measure the resistance of the metal [5]. This measurement also provided information on the isotropy of the film, which is another preliminary indicator of whether or not a film may superconduct (very anisotropic films of Sr_2RuO_4 were found to be non-superconducting).

A vitally important growth parameter in growing superconducting thin films is the substrate, which strains the film by some amount based on how closely the lattice parameters of the film and substrate are matched. Previously we have seen with Sr_2RuO_4 grown on various substrates that this deliberate mismatch allows us to improve the superconducting properties of the film, namely the T_c which can be optimized by finding this ideal lattice match.

One key challenge in the synthesis of Ba_2RuO_4 is how quickly the film decomposes after growth due to moisture, and therefore air exposure of the film surface is detrimental to any potential measurements. This degradation of the film could be observed clearly through X-ray diffraction measurements such as Figure 3 shows, which are used to determine the structure and fraction of the film that contains the desired Ba_2RuO_4 phase (phase purity) by analyzing film peak intensity (counts per second) and broadening (an indication of defect concentration in the material). With Ba_2RuO_4, the peak intensity decreases significantly within minutes of being removed from the growth chamber as it exposed to air. Several peaks are entirely gone within hours, forming probably what is a phase of different symmetry. To help improve the lifetime of the material we added a capping layer to the growth process.

Results and Conclusions:

We were successful in growing high quality films by repeating our growth optimization loop until we achieved record quality, as demonstrated by a *RRR* of 30 in our best film. This value is comparable with Sr_2RuO_4 films that clearly exhibited superconductivity. Because of the low T_c of ruthenate superconductors, however, this measurement could not give us definitive data on where this transition occurs along a resistivity versus temperature graph. For that these films need to be measured on a system that reaches ultra-low temperatures, namely a physical properties measurement system (PPMS), a system capable of reaching 0.3K. Ba_2RuO_4 was grown on $DyScO_3$, $GaScO_3$, and $TbScO_3$ over the course of this project. Our highest *RRR* films were grown on $TbScO_3$, including the sample in Figure 4.

We were also successful in synthesizing the capping layer to an extent that the film would last for days with little degradation, enough time to perform most required measurements. This was achieved by growing a $DyScO_3$ layer on top of Ba_2RuO_4, first in a crystalline structure to help prevent moisture from reaching the film followed by an amorphous layer to slow oxidation.

Future Work:

Some future work on this project will include gathering more data on the capping layer so that the lifetime of Ba_2RuO_4 thin films can be increased and more data can be collected on individual samples. This will mainly include taking X-ray scans on a film over time to see if the peaks degrade over time. We also want to definitively determine whether Ba_2RuO4 indeed superconducting and find the transition temperature if so. In addition to this, there can be further research that looks into how superconducting properties of this material can be manipulated by strain.

Acknowledgements:

Special thanks to Prof. Darrell Schlom, Nathaniel Schreiber, and others in the Schlom and Shen labs. Support for PARADIM was provided by the National Science Foundation under Cooperative Agreement No. DMR-1539918 as part of the Materials Innovation Platform Program.

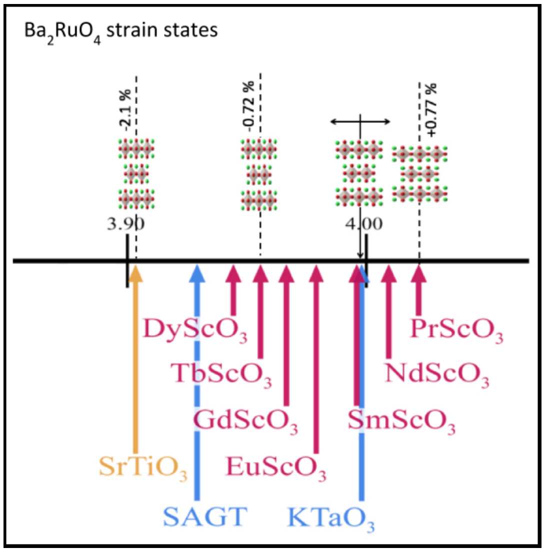

Figure 2: Ba_2RuO_4 strain states for various substrates.

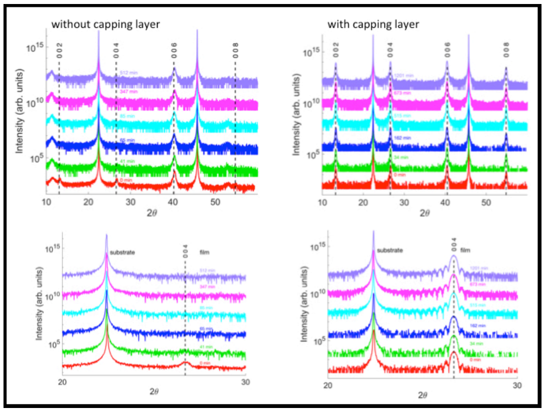

Figure 3: Degradation of Ba_2RuO_4 peaks without and with capping layer.

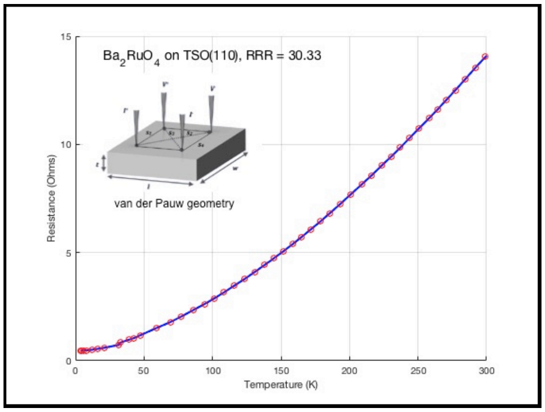

Figure 4: Best RRR Ba_2RuO_4 sample from the helium dipper.

References:

[1] Nair, et al. APL Mater. 6, 101108 (2018).
[2] Burganov, et al. Phys. Rev. Lett. 116, 197003 (2016).
[3] Jia, Y., et al. Appl. Phys. Lett. 74, 3830 (1999).
[4] Schlom, D. G., et al. J. Am. Ceram. Soc., 91 2429-2454 (2008).
[5] Miccoli, I., et al. J. Phys.: Condens. Matter 27, 223201 (2015).