Searching for Superconductivity in Ruthenate Thin Films Grown by Molecular Beam Epitaxy

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Abstract

Spin-triplet superconductors are unconventional superconductors in which the electrons form pairs with total spin moment magnitude S=1 while in conventional superconductors the Cooper pairs have total spin moment magnitude S=0. Such materials are possible platforms for realizing majorana fermions which can find applications in quantum computing. Originally, Sr_2RuO_4 was considered to be a spin-triplet superconductor, however, in 2019, a new study found it to not contain spin-triplet pairing using nuclear magnetic resonance (NMR) measurements. It has now been predicted that spin-triplet pairing may be possible in samples with the ideal amount of strain and chemical pressure. This work attempted to employ epitaxial strain using molecular beam epitaxy (MBE) as well as chemical pressure through the complete replacement of Sr atoms with Ba atoms within the same perovskite crystal structure to grow Ba_2RuO_4 thin films that display spin-triplet superconductivity. In this project, single-phase Ba_2RuO_4 thin films were successfully grown by MBE, however superconductivity was not observed in these films.

Introduction

Spin-triplet or topological superconductors are unconventional superconductors in which the electrons form pairs with total spin moment magnitude S=1 while in conventional superconductors the Cooper pairs have total spin moment magnitude S=0 [1]. Such materials are possible platforms for realizing majorana fermions which can find applications in quantum computing [2]. Originally, there was interest in Sr_2RuO_4 due to the possibility of it displaying spin-triplet, topological superconductivity. An initial nuclear magnetic resonance (NMR) study found spin-triplet pairing in Sr_2RuO_4, however, a second study found no evidence of spin-triplet pairing [3], [4].

It has now been predicted that with the proper amount of strain and chemical pressure spin-triplet superconductivity may still be possible in these ruthenate materials [5]. Further, angle-resolved photoemission spectroscopy (ARPES) has demonstrated the use of strain and chemical pressure to tune the topology of the Fermi surface in ruthenate thin films, showing that this is a possible route to spin triplet superconductivity [6].

This project aimed to use epitaxial strain and chemical pressure through the complete replacement of strontium with larger barium ions in Sr_2RuO_4 to grow single-phase, isostructural superconducting Ba_2RuO_4 thin films by MBE. We were successful in growing single-phase Ba_2RuO_4 films, however our samples did not display superconductivity.

Experimental

MBE was used to grow all Ba_2RuO_4 films in this project, and the growth conditions were selected using the Ellingham diagram shown in Figure 1. Three main growth parameters: the substrate temperature, oxygen partial pressure, and the Ru/Ba flux ratio were varied in an attempt to determine the ideal growth conditions for Ba_2RuO_4. The growths were performed using an excess of ruthenium due to its volatility, as to avoid ruthenium vacancies in the samples. The growth conditions for the samples discussed here are shown in Table 1 and are labeled as samples A, B, and C.

For this work, $DyScO_3$ (DSO) substrates oriented in the (110) direction were used. DSO has a lattice parameter of 3.94Å while that of Ba_2RuO_4 is 3.99Å leading to a 1.25% compressive strain in the films. Previously, Ba_2RuO_4 samples have been air sensitive, so an amorphous Dy_2O_3 capping layer was deposited over the films to keep them stable while they were characterized.

Sample	Growth Temperature (°C)	Oxygen Pressure (Torr)	Ru/Ba Flux Ratio
A	845	$5x10^{-7}$	0.7
B	850	$5x10^{-7}$	0.7
C	855	$7x10^{-7}$	0.8

Table 1: Growth Conditions for Ba_2RuO_4 samples A, B, and C

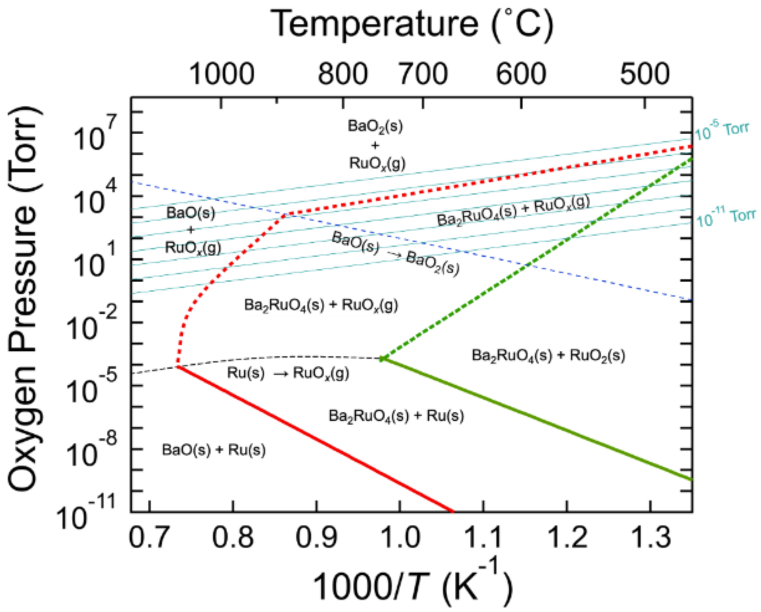

Figure 1: The Ellingham diagram for the various oxide phases containing ruthenium and barium

After growth, X-ray diffraction (XRD) was used to confirm the phase purity of the Ba_2RuO_4 films. Then, the resistance of the samples was measured along two directions of the substrate at temperatures ranging from 300K to 4K using a four-point probe with van der Pauw geometry in a liquid He dipper. An important parameter to check for superconductivity in ruthenate samples is residual reisistivity ratio (RRR) which is the ratio of resistance at 300K to resistance at 4K. For Sr_2RuO_4 samples to superconduct, a minimum RRR of 30 is required.

Results and Discussion

As shown in Table 1, sample A was grown at a substrate temperature of 845°C. The theta-2theta XRD scan for the sample can be seen in Figure 2(a). Sample A was single-phase Ba_2RuO_4, however the broadness of the diffraction peaks indicates poor crystallinity. Additionally, as shown in Figure 2(b), resistance measurements found the sample had a RRR of 6 in the vertical direction and only 5 in the horizontal direction across the substrate, demonstrating that this sample would not be superconducting.

Sample B was grown at a higher temperature of 850°C. The XRD pattern shown in Figure 2(c) confirmed that it was single-phase, and the peaks were sharper than in sample A, indicating better crystallinity. However, the film was still not superconducting as the RRR was only 6 in the vertical diraction and 4 in the horizontal direction as seen in Figure 2(d).

Sample C was grown at 855°C and once again was single-phase as shown in Figure 2(e). Sample C was also not superconducting as indicated by its RRR of 6 in both the horizaontal and vertical directions displayed in Figure 2(f).

Although phase-pure Ba_2RuO_4 films were able to be grown by MBE in this project, none of them were found to be superconducting. The lack of superconductivity can most likely be attributed to carbon impurities in the samples. The MBE used to grow these samples had difficulties with carbon outgassing from the silicon carbide substrate heater during the growths, particularly at high temperatures. Superconductivity in Sr_2RuO_4 is incredibly sensitive to impurities, so the carbon in the chamber likely lead to the poor electrical transport in Ba_2RuO_4 and unobservance of superconductivity.

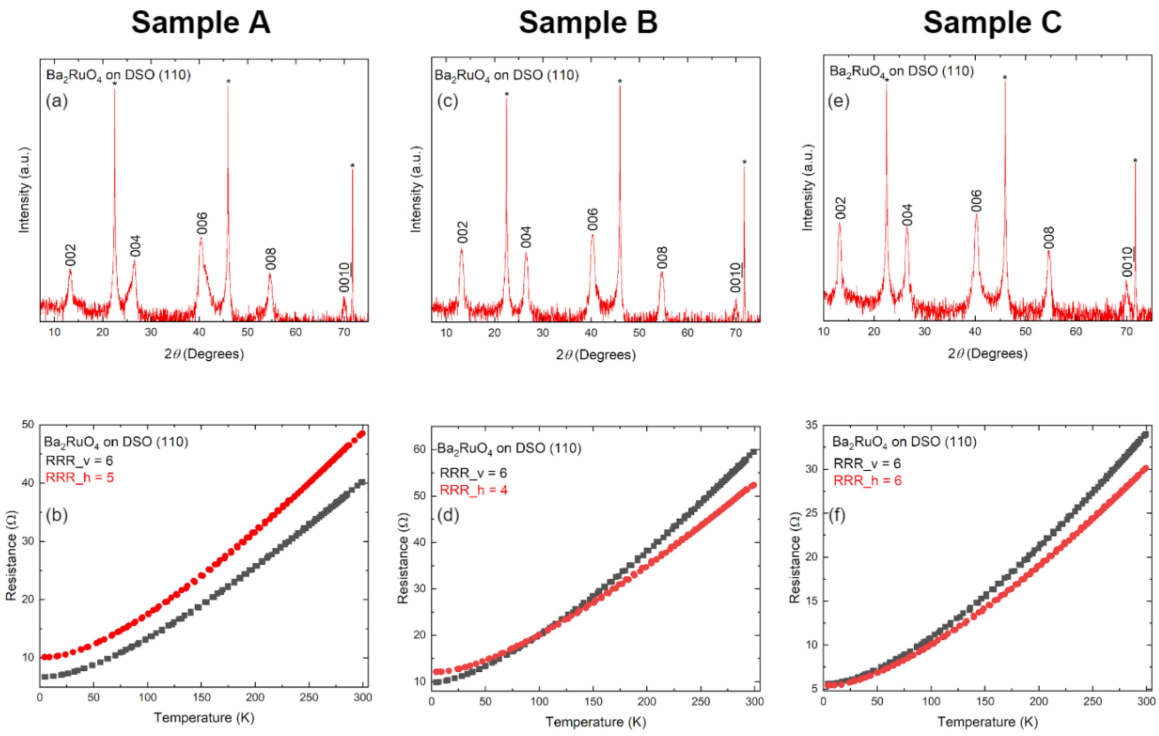

Figure 2: (a) and (b) show the X-ray diffraction pattern and resistance data respectively for sample A, (c) and (d) show the X-ray diffraction pattern and resistance data respectively for sample B, and (e) and (f) show the X-ray diffraction pattern and resistance data respectively for sample C

Conclusion

In this project, single-phase Ba_2RuO_4 thin films were successfully grown by MBE. However, the films did not display a superconducting transition, possibly due to impurities in the samples. In the future, we hope to grow superconducting Ba_2RuO_4 thin films by further optimizing the growth conditions and decreasing the number of impurities within the samples.

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