Optimization of Spin Qubits in Double Perovskites

Abby Neill, Thomas Whoriskey, Evan Crites, Tyrel M. McQueen
PARADIM Research Experience for Undergraduates

Background

Quantum information science (QIS) is a rapidly developing field, and quantum computing is the most advanced field within QIS[1]. Computation on the quantum scale is possible due to the existence of quantum bits or "qubits" which exist in a superposition of two distinct states. This project focuses on spin qubits that originate from unpaired electrons within the crystal structure of inorganic double perovskites. These unpaired electrons can exist in spin +1/2(up) and spin -1/2(down) states, as well as any superposition of these two states.

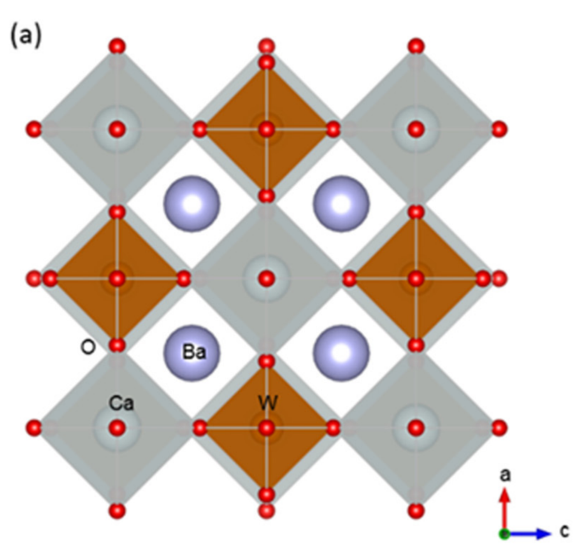


Fig 1: Crystal structure of Ba_2CaWO_6 from Sinha, M.; et al.

T_1: 310 ms
T_2: 4 μs
$T_1 \geq 2*T_2$

$Ba_2CaW_{1-y}M_yO_{6-x}$

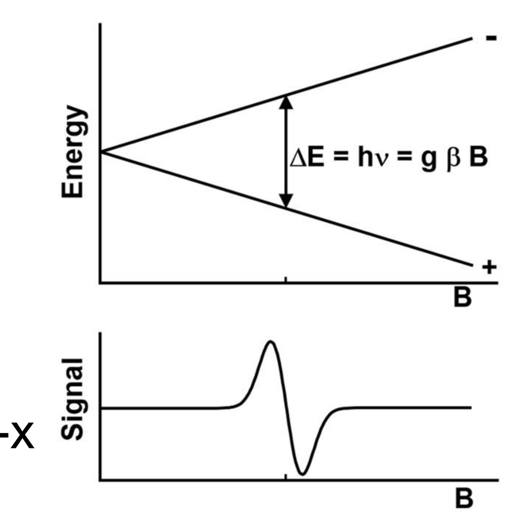


Fig 2: EPR Energy level diagram from Bogart, E.; et al.

Methodology

To improve the T_2 of Ba_2CaWO_6, the amount of W^{5+} spin centers needed to be decreased. This was accomplished by adding ions of similar size to Tungsten possessing a 4+ charge, in a process known as compensation doping. The ions that were utilized were Zr^{4+} and Ge^{4+}.

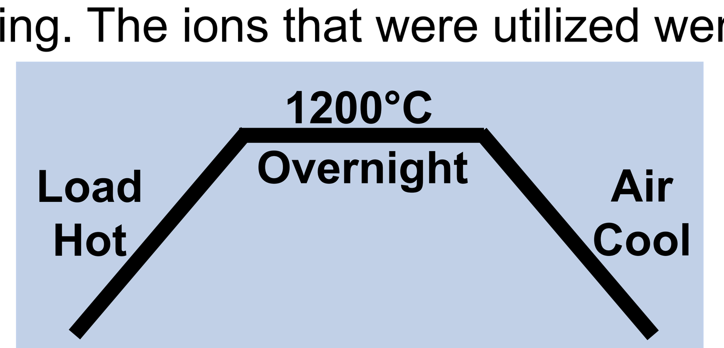


Fig 3: Heating profile for synthesis of Ba_2CaWO_6 and corresponding doped samples.

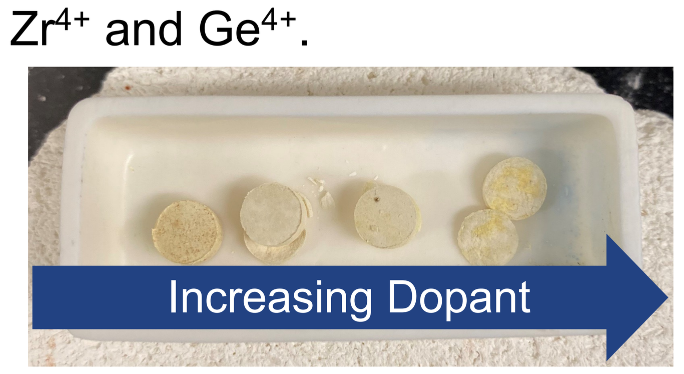


Fig 4: Series of samples (ANN019-022) where Ge was doped into Ba_2CaWO_6. Picture taken after reaction.

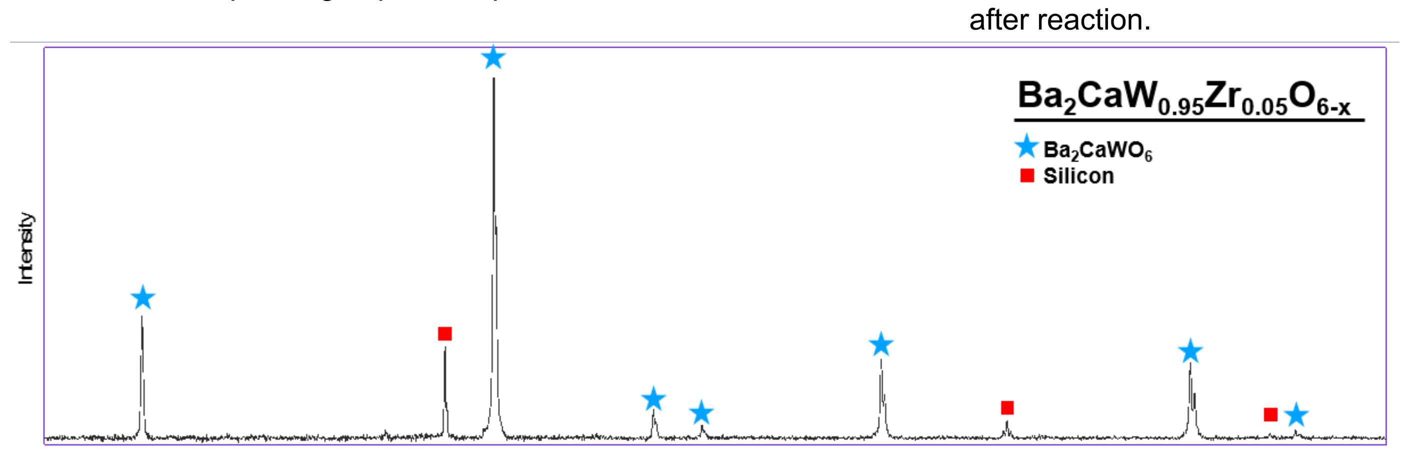


Fig 5: Powder X-Ray Diffraction data collected on ANN008 with labelled peaks for Ba_2CaWO_6 and the Si standard used to negate the effects of sample tilt and displacement in lattice parameter calculations.

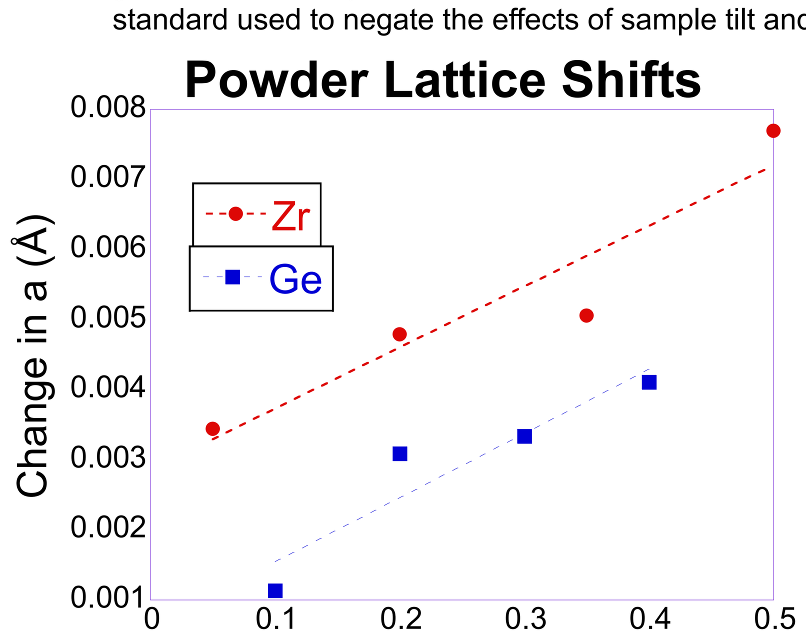


Fig 6: B site doping versus lattice parameter shift(determined via hkl calculations in Topaz).

Determination of dopant level within a given sample was determined via the lattice parameter shift. The addition of metallic ions of varying size on the B site of the perovskite will cause small, yet determinable changes in the lattice parameters, despite the identity of the compound remaining the same when evaluated via powder XRD.

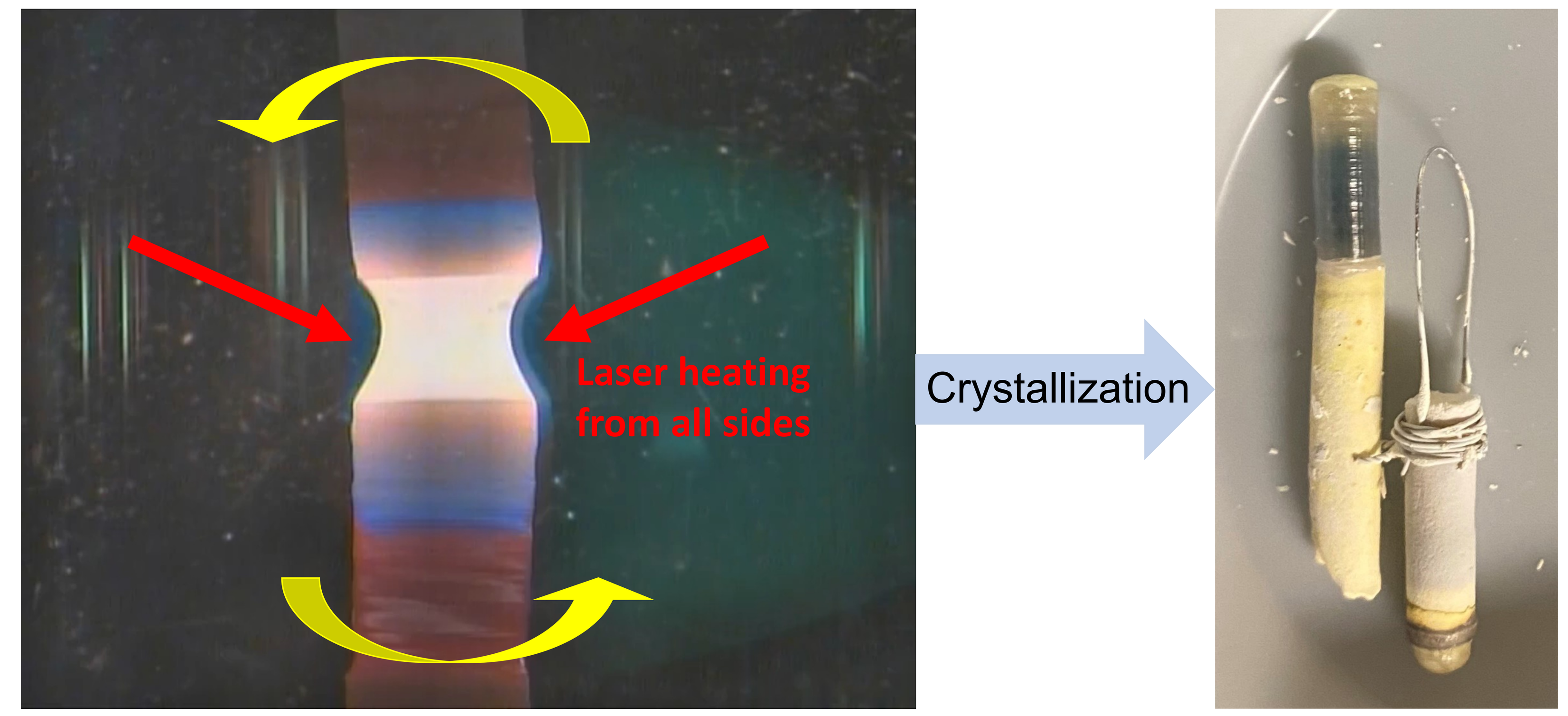


Fig 7: Sample ANN018 (pure Ba_2CaWO_6) during and after crystal growth. Image from inside the laser diode floating zone(LDFZ) during crystallization(left) and the crystal that resulted from said growth(right).

Results

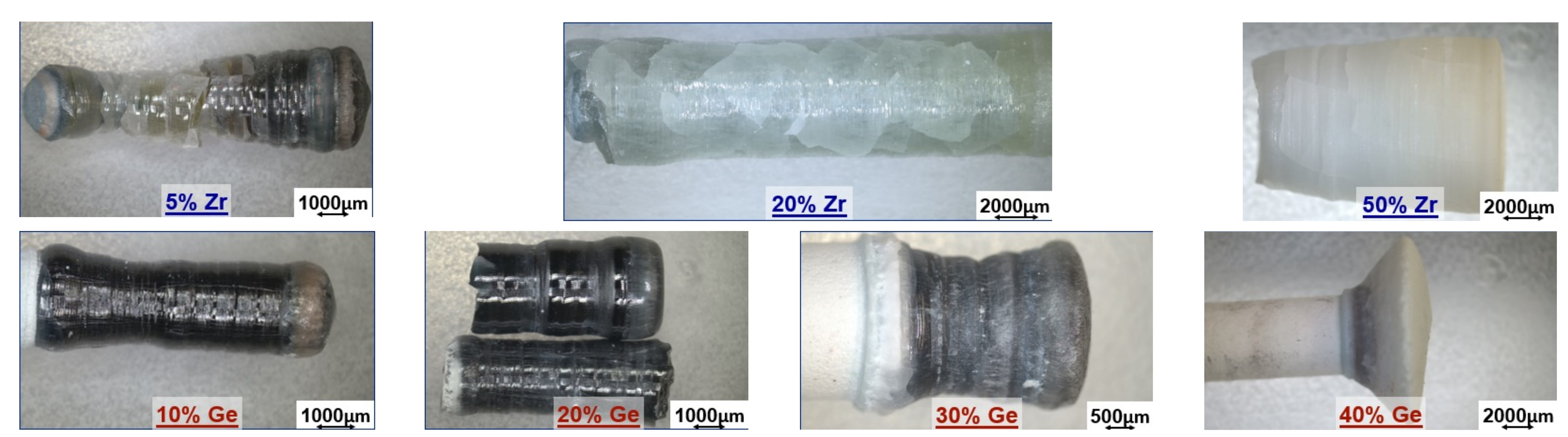


Fig 8: Doped crystals grown in the LDFZ labelled with their dopant percentage and size.

With increasing dopant percentage, the crystals became lighter and more opaque, and they had less stable molten zones in the growth process. In addition, the more dopant that was placed in the sample, the more sensitive that sample was to air.

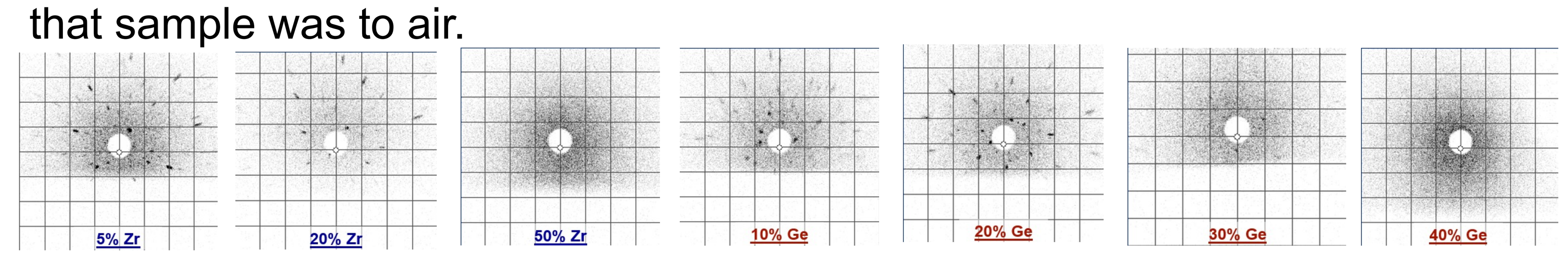
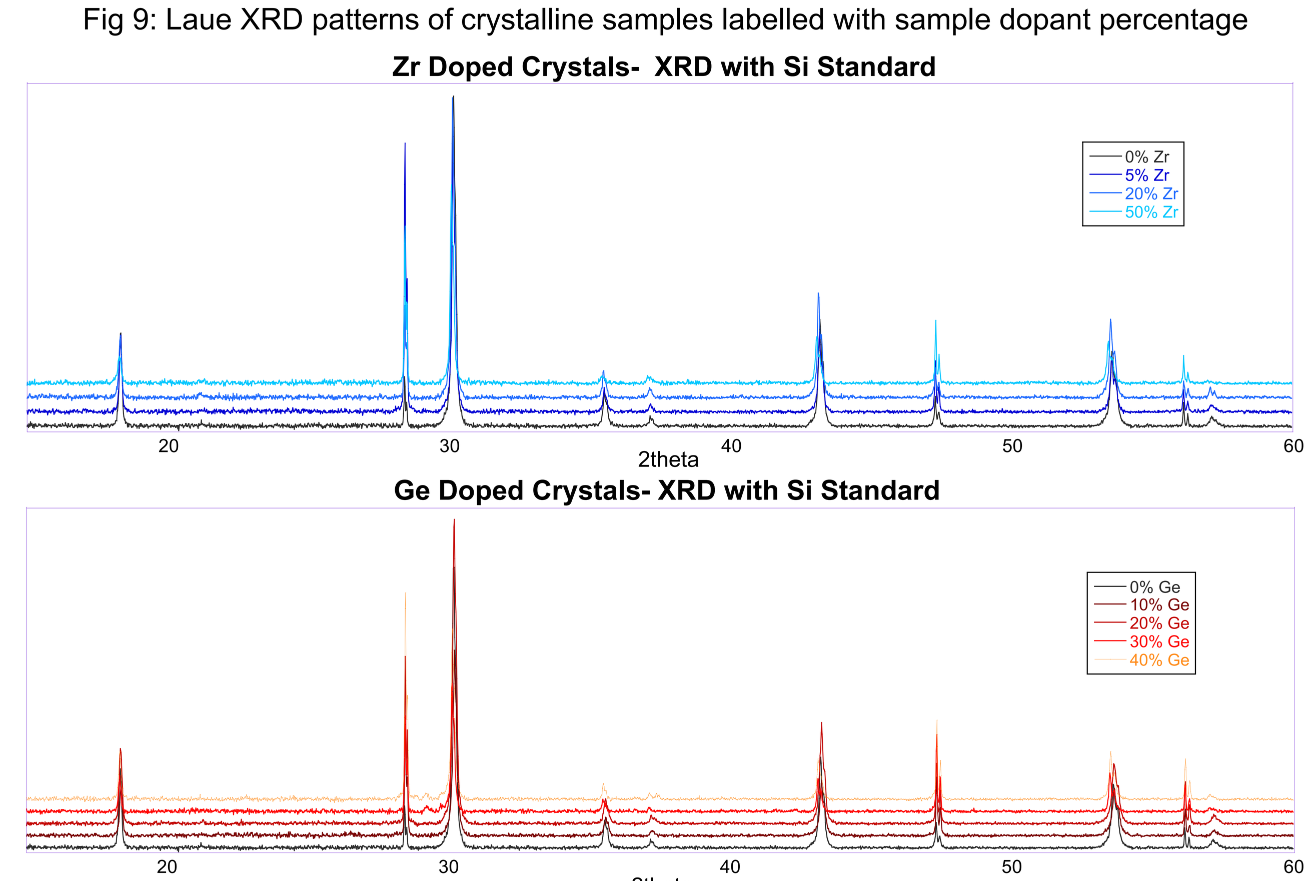


Fig 9: Laue XRD patterns of crystalline samples labelled with sample dopant percentage



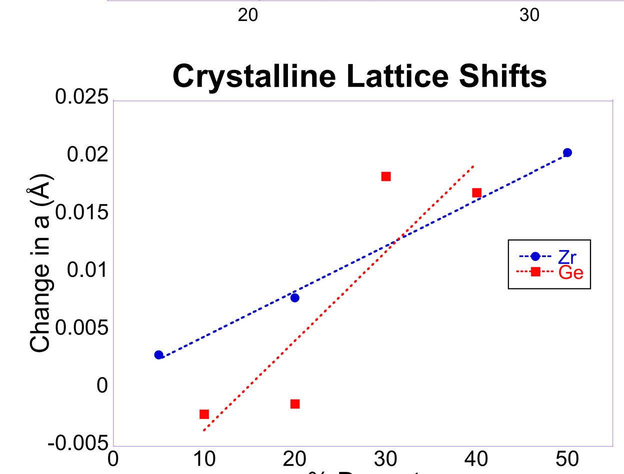


Fig 10 & 11: Figure 10 (above) shows powder XRD patterns of the LDFZ grown crystalline samples, and figure 11 (left) displays the calculated lattice parameter shifts (via hkl refinements in Topaz) plotted against intended dopant percentage.

Laue and powder XRD showed that the intended materials were created, but the crystallinity of said materials decreased with increasing dopant percentage.

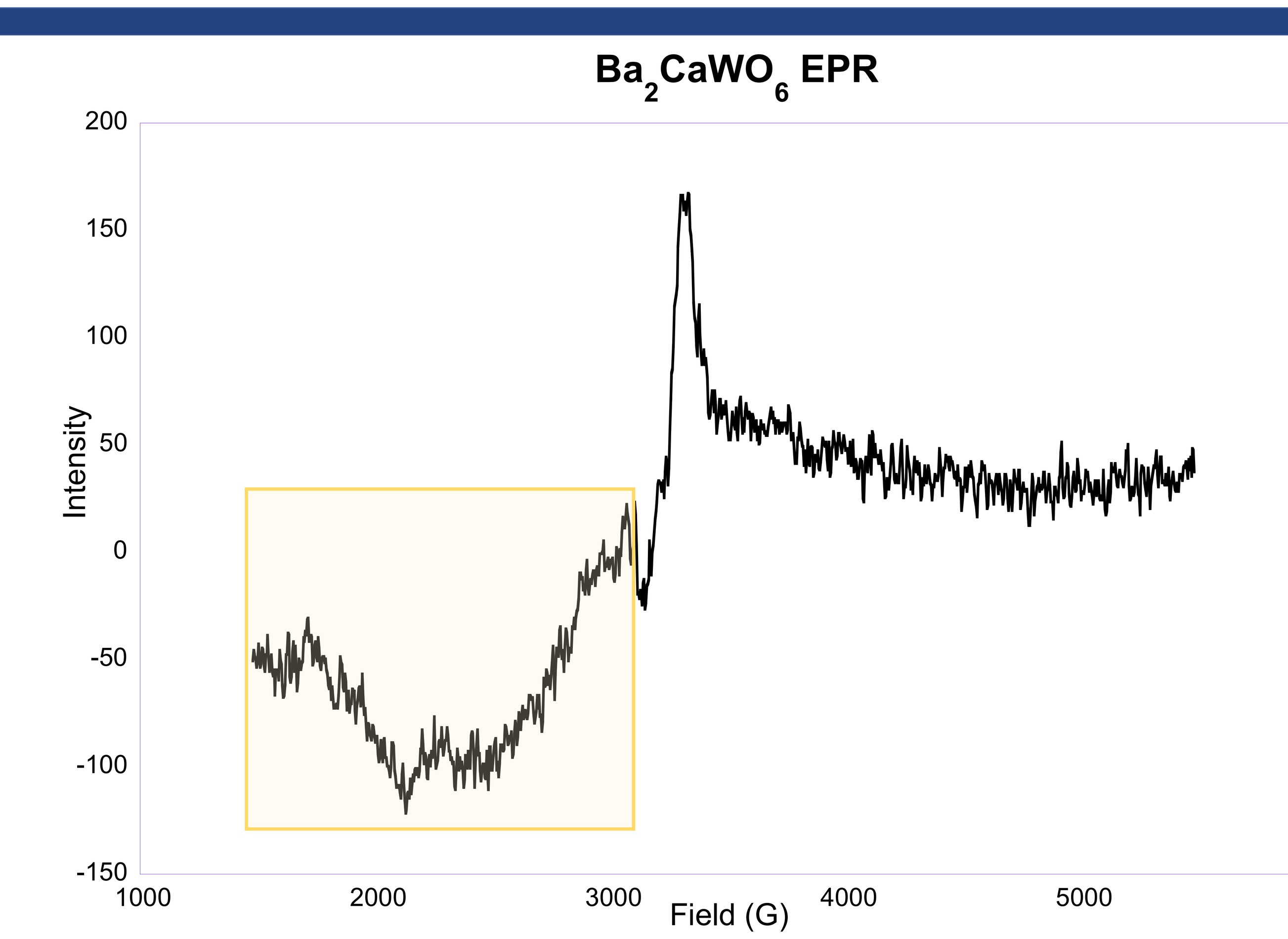


Fig 8: Continuous EPR scan on undoped Ba_2CaWO_6 with low field irregularities due to heavy spin-spin interactions.

Discussion

The compensation doping of 4+ ions onto the Tungsten site of Ba_2CaWO_6 proves to be a fairly painless procedure with very few drawbacks. However, several different roadblocks are introduced when these powders are to be made into crystals. Sintered rods, especially those of high dopant concentration, are air sensitive and must be treated with more care than their less doped counterparts. These higher doped samples also have less stable floating zones and extremely high evaporation rates in the LDFZ crystalline growths. Difficulties in growth require more attention and a more experienced furnace operator. The crystals produced from these growths also decrease in crystallinity as dopant concentration increases, which could present complications in future properties measurements.

The next step in this project is to find a functioning EPR on which to acquire measurements on the seven remaining crystals I grew during my time at JHU. Once these measurements are complete, an accurate conclusion can be drawn about the effect the dopant concentration has on the quality of the spin qubits of this material. Once these conclusions are made, I hope to write a paper on this material in order to disperse this information to the quantum materials community.

Sources & Acknowledgements

1. Leon, N. P. d.; et al., Materials Challenges and Opportunities for Quantum Computing Hardware. *Science* **2021**, *372*.

2. Bogart, E.; et al.,The Noninvasive Analysis of Paint Mixtures on Canvas Using an EPR Mouse. *Heritage* **2020**, 3, 140-151.

3. Sinha, M.; et. al, Introduction of Spin Centers in Single Crystals of Ba_2CaWO_6. *Phys. Rev. Mater.* **2019**, *3*.

4.Mirinioui, F.; et al., Sequence of Phase Transitions Induced by Chemical Composition and High Temperature in $[Ba_2CaWO_6]_{(1-x)}[Sr_2CaWO_6]_x$ Double Perovskite Tungsten Oxides. *J. Solid State Chem.* **2015**, *232*, 182-192.

Thank you to Dr. Tyrel M. McQueen, Thomas Whoriskey, Lucas Pressley, Satya Kushwaha, Austin Ferrenti, Brandon Wilfong, Shannon Bernier, and Evan Crites for all their knowledge and assistance in this incredible educational experience. Also, thank you to PARADIM and the National Science Foundation for making my research possible. This work is part of a PARADIM user project funded by the U.S. Department of Energy, Office of Science, National Quantum Information Science Research Centers, Co-design Center for Quantum Advantage (C2QA) under contract number DE-SC0012704.