Optimization of Spin Qubits in Double Perovskites

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Abstract:

The spin quantum bit (qubit) host Ba_2CaWO_6 presents promise as a quantum computing material, but the spin-spin relaxation time (T_2) has lots of room for improvement. Compensation doping, or the practice of introducing impurities to a material to change its properties, was utilized in an attempt to increase the T_2 for this double perovskite structure. Multiple series of doped samples were synthesized, grown into large single crystals, and then measured using Laue XRD and powder XRD to determine their identity and crystallinity. The process of doping onto Ba_2CaWO_6 was determined to be fairly efficient and effective, and therefore shows great promise for repetitive enhancement of this material, despite difficulties in crystal growth. Upon the completion and comparison of more electron paramagnetic resonance measurements, there should be a clear picture of how doping 4+ metallic ions onto Ba_2CaWO_6 can influence its T_2 value.

Introduction:

To propel the advancement of quantum information sciences, quantum materials must be created and optimized. Oxygen vacancies within double perovskite Ba_2CaWO_6 cause unpaired electrons to find a new home on the Tungsten site, changing the ion from W^{6+} to W^{5+}, which is a spin qubit host in this structure. However, interaction between these unpaired spins is too strong, yielding a small spin-spin relaxation time (T_2). To improve the T_2 for this material, 4+ metallic ions of similar size to Tungsten were added into the structure in a process known as compensation doping. By introducing an ion of a different charge, the unpaired spins have multiple atomic sites on which to reside, and therefore the amount of qubit sites within the material will decrease.

Methods:

The baseline material (Ba_2CaWO_6) was created by combining $BaCO_3$, $CaCO_3$ and WO_3 in a mortar and pestle, grinding them until homogeneous, and then baking them at 1200°C overnight. A series of samples of each dopant type(Zr^{4+} and Ge^{4+}) were created by adding the oxide form of each ion stoichiometrically to the original material's sample preparation in place of part of the WO_3 and then following the same procedure. Lattice parameters for each of the samples was determined via hkl refinements in Topaz using a Silicon standard to negate the effects of sample tilt and sample displacement. The shift in the lattice parameters for these samples can be directly attributed to the dopant on the B site, for the identity of the material remained the same and the change in lattice parameters showed a positive trend when compared to the B site doping. The results of these refinements can be seen below.

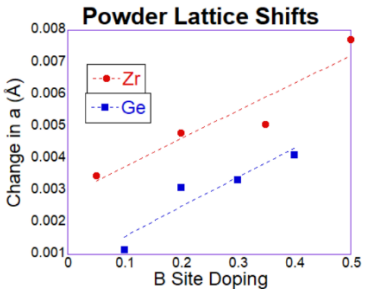


Figure 1. Graph of hkl refined lattice parameters of powdered samples versus intended B site doping

Once the success of the doping was determined, each doped sample was created in bulk following the original procedure. The bulk samples were then pressed into rods and sintered at 1200°C overnight, removed from the furnace at 400°C, and cooled under vacuum in the antechamber of the glovebox. These rods were then taken to the Laser Diode Floating Zone Furnace (LDFZ) where they were grown into crystals under comparable conditions.

Results and Discussion:

Both series of crystals grown in the LDFZ showed corresponding irregularities. 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. From my observations, the crystals themselves were not sensitive to the air, but as the sintered rods were exposed to the atmosphere, they began to disintegrate.

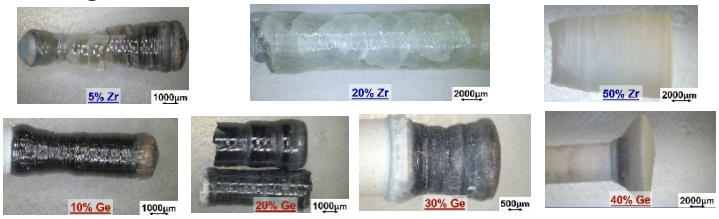


Figure 2. Doped crystals grown in the LDFZ labelled with their dopant percentage and size.

Based on the Laue XRD patterns, the opacity of the more doped crystals was most likely due to the materials growing in a polycrystalline fashion, for the patterns showed very few/no dots which would indicate a lack of crystalline symmetry. Powder XRD showed that all of the crystals did form the intended structure, which reinforced the conclusion that the opaque crystals were merely of a polycrystalline structure. The powder XRD patterns were again measured with a Silicon standard to ensure all lattice parameter changes were due purely to the dopant within the samples. When compared to the baseline Ba_2CaWO_6 crystal, all of the doped crystals did show a steady positive increase in lattice parameters with increasing dopant percentage.

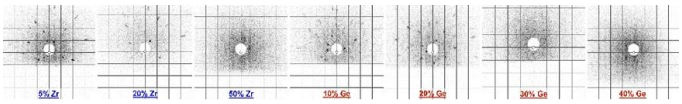
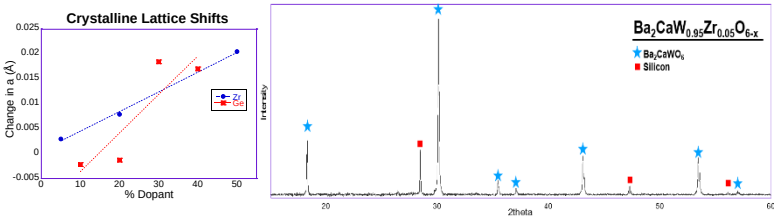


Figure 3. Laue XRD plots (above), example powder XRD and refined lattice parameters (below) for crystalline samples.



In addition, a continuous electron paramagnetic resonance (EPR) measurement on the undoped crystal was able to be performed. Unfortunately, the instrument went out of operation shortly after that sample was measured, and therefore no comparability of the crystalline qubits can be determined.

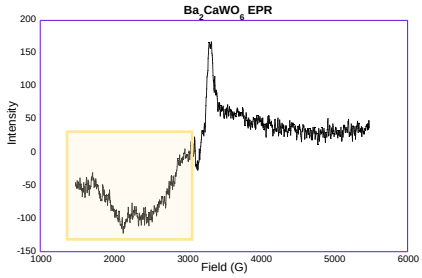


Figure 4: Continuous EPR scan on undoped Ba_2CaWO_6 with low field irregularities due to heavy spin-spin interactions.

Conclusions and Next Steps:

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.

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References:

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