

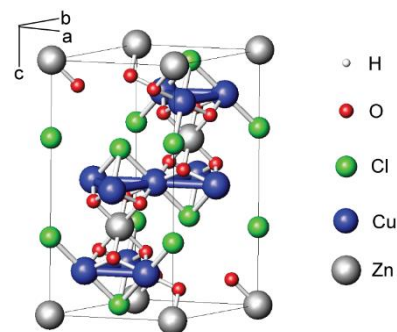
Can kitchen-chemistry help solve a cutting-edge physics problem?

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Magnetic materials are a nice playground for testing our understanding of physics, as relatively simple interactions build up to complex magnetic properties, depending on how the magnetic atoms are arranged in the material's crystalline lattice. If we arrange the magnetic atoms such that the overall interaction is frustrated, where pair-wise interactions can't all be satisfied at the same time, we often get unexpected ground-state properties.

One of these unusual magnetic ground-states, is a quantum-spin-liquid. The quantum spin-liquid state is predicted for a Kagome lattice of antiferromagnetically coupled low spin-state ions, but the exact properties are notoriously difficult to predict as they are strongly dependent on the method used for the theoretical calculations.

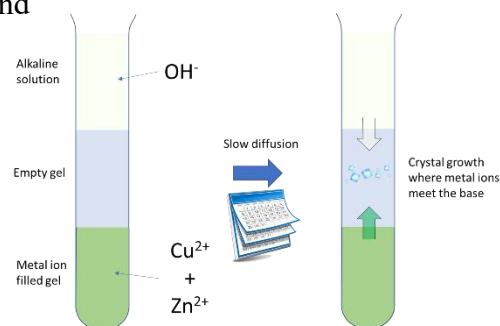
One material, that has the right arrangement of ions to test the model in real-life is the mineral Herbertsmithite, γ - $\text{Cu}_3\text{Zn}(\text{OH})_6\text{Cl}_2$. It can be synthesised as a single crystal by high-temperature hydrothermal recrystallisation, and has been extensively studied since it was first prepared in 2006.



Despite 18 years of intensive study and more than 136 papers written about Herbertsmithite, the ground-state still remains a contentious issue because of a small complication with the way the crystal samples are grown. During the crystallisation process, the formation of defects is inevitable (e.g. Zn^{2+} and Cu^{2+} swapping positions every now and again) [1,2]. These defects create a big problem in magnetic property measurements, because at very low temperatures they dominate the observed signals – masking the true physics of the Kagome lattice frustration and the properties of the quantum spin liquid ground-state. Defects form in higher concentrations when the material is crystallised at high temperatures, happening about once every 17 unit cells at the temperatures of the current crystal synthesis route. If the crystals can instead be grown at room temperature, the defect concentration is expected to be more than an order of magnitude smaller.

An old, and rarely used crystal growth technique – using slow diffusion through gels [3,4]– may be a solution to this problem. Initial tests using silicate gels hasn't worked due to sensitivity of the gel to the pH level needed for the crystallisation. Instead, this project will attempt to use gelatin based gels (identical to that use in food preparation) to achieve crystal growth. An unrelated study has found that food gelatine may be just right for growing the material we want [5]. The project will involve setting up an extensive series of tests using varying gel thickness and solution concentrations. The tests will be monitored to identify the setup that leads to the best crystals, and then will be used to prepare crystals with different Cu:Zn ratios.

The Cu:Zn ratio will be determined using crystallography and spectroscopic techniques, and defect concentrations will be determined using temperature dependent magnetometry.



References:

- [1] A. Olariu, P. Mendels, F. Bert, et al., Phys. Rev. Lett. **100**, 087202 (2008).
- [2] D. E. Freedman, T. H. Han, A. Prodi, et al., J. Am. Chem. Soc. **132**, 16185 (2010).
- [3] A. R. Patel and A. Venkateswara Rao, Bull. Mater. Sci. **4**, 527 (1982).
- [4] L. Dong, T. Besara, A. Henderson, et al., Cryst. Growth Des. **17**, 5170 (2017).
- [5] C. J. S. Ibsen, B. F. Mikkladal, U. B. Jensen, et al., Chem. - A Eur. J. **20**, 16112 (2014).