A combined neutron and DFT study of the pressure-induced structural transition in Zabuyelite (Li₂CO₃)

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Zabuyelite, Li₂CO₃, is one of the largest global sources of lithium, used as a precursor in the synthesis of lithium-ion battery cathode materials, the supply of which is increasingly stressed with a growing demand for battery technology. This is a field where the use of high-pressure has yet to be fully investigated; it can, for example, be used to stabilise the synthesis of unusually doped cathode materials which can then be recovered to ambient conditions [1], or be used to control particle size, or alter synthesis efficiency for existing materials. In addition, Zabuyelite has been demonstrated to be effective as a solvent-catalyst for diamond growth, a process of understated industrial importance [2]. Despite this, there is relatively little known about the crystallography of Li₂CO₃ at high-pressure, beyond the existence of a hexagonal phase at elevated pressures [3].

I present a full structural analysis of Zabuyelite as a function of applied pressure, as measured using neutron powder diffraction with a large volume press (from the PEARL instrument at the ISIS Neutron and Muon Source), supported with Raman spectroscopy measurements, and density functional theory (DFT) analysis. The carbonate was found to show unusual high-pressure behaviour, differing to that reported in the literature from X-ray measurements, showing negative linear compressibility, leading to a distortive first-order phase-transition to a hexagonal structure above ~8 GPa, with a change to octahedral Li⁺ coordination.