Anomalous structural behaviour near a Mott transition in compressed Ca_2RuO_4

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Ca_2RuO_4 has a rich phase diagram, featuring a metal-insulator transition with a structural distortion upon cooling, antiferromagnetic ordering upon further cooling, and metallisation and ferromagnetic order, followed by a further structural transition, under pressure [1,2]. It is one end member of the $Ca_{2-x}Sr_xRuO_4$ series that includes the proposed triplet superconductor Sr_2RuO_4 at the other end.

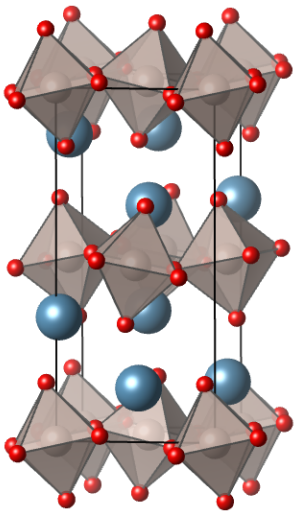

Figure 1. Crystal structure of Ca_2RuO_4.

Ca_2RuO_4 shows intriguing collective phenomena in its own right, such as electric-field-induced metallisation and current-induced diamagnetism, negative thermal expansion, and a Higgs mode [3-6]. Several of these properties are intricately linked to structural distortions of the RuO_6 octahedra, which in turn are susceptible to pressure-induced changes. In fact, Ca_2RuO_4 shows an unusual *c*-lattice expansion under hydrostatic compression [2]. However, while its electronic structure has been explored in several computational studies at ambient conditions [7,8], its structural and electronic properties under pressure have not been investigated in detail before.

Here, we study the high-pressure phase evolution of Ca_2RuO_4 using density functional theory (DFT) and DFT+U calculations, considering a wide range of on-site repulsion terms U to reflect the expected decrease of U with pressure. We show that the *c*-lattice expansion in the metallic high-pressure phase and the structural phase transition can be described well if the U-term is fine-tuned appropriately. The arrangement and distortions of the octahedra are found to be intimately connected to this unconventional behaviour. The sensitivity to the size of U highlights how close the metallic phase remains to the Mott insulating phase.

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