Exploiting symmetry-adapted structural distortions for electronic singularity engineering in $Sr_3Ru_2O_7$

Caitlin O'Neil[1,2], Maximilian T. Pelly[3], Ben Gade[3], Alexander J. Browne[3,4], Alexandra S. Gibbs[3,5], Andreas W. Rost[1]

1 SUPA, University of St Andrews, 2 IMPRS-CPQM, Dresden, 3 EaStCHEM, University of St Andrews, 4 EaStCHEM, Edinburgh, 5 ISIS, RAL

mp250@st-andrews.ac.uk

The orthorhombic n = 2 Ruddlesden-Popper compound $Sr_3Ru_2O_7$ is a prime example of a strongly correlated oxide, displaying unconventional electronic phases [1] associated with a quantum critical end point and an underlying Van Hove singularity (VHs) in the band structure [2, 3]. In the n = 1 relative, the unconventional superconductor Sr_2RuO_4, tuning this singularity through the Fermi level using uniaxial stress is associated with a significant increase in the superconducting transition temperature [4]. In $Sr_3Ru_2O_7$ the coupling of the VHs to uniaxial stress is weaker and it has not yet been possible to move the VHs to the Fermi energy using stress [5].

Here we instead utilise isovalent Ba doping on the Sr sites ($(Sr_{1-x}Ba_x)_3Ru_2O_7$) as a chemical pathway to tune the structure. Selectively doping on to the out of plane Sr sites this provides a clean way to drive the system closer to the tetragonal aristotype as realised in Sr_2RuO_4. We confirm this using high resolution neutron powder diffraction, required to accurately locate the relatively weakly scattering oxygen ions and resolve changes in internal modes. Three irreducible representations are activated by the doping - two Γ "breathing" modes and a combined rotation ($X_1^-\ B_1^{(a)}$) and rhombic distortion ($X_1^-\ B_2^{(a)}$) of the octahedra. Using band structure calculations we find that the rotation mode couples strongly to the VHs, driving it towards the Fermi level. We confirm this with detailed specific heat measurements, which show a shift towards 0 K of the low-temperature peak known in the undoped compound. We find the crossing to occur at approximately x = 0.08, with the peak position subsequently weakly increasing in temperature again.

This strong coupling means the rotational mode should soften as the VHs passes through the Fermi energy, just as has been recently observed for the bulk modulus in Sr_2RuO_4 [6]. Softening should occur regardless of how the movement in the VHs is achieved, and so could lead to significant activation of the mode as a function of magnetic field, leading to strong lattice feedback in the observed transitions. We note none of this is unique to $Sr_3Ru_2O_7$ and it is likely that such hard to detect internal modes are involved in many quantum phase transitions. We will present both our detailed study of $(Sr_{1-x}Ba_x)_3Ru_2O_7$ and our more general methodology of symmetry-mode-guided bandstructure engineering which is of relevance to a range of quantum materials.

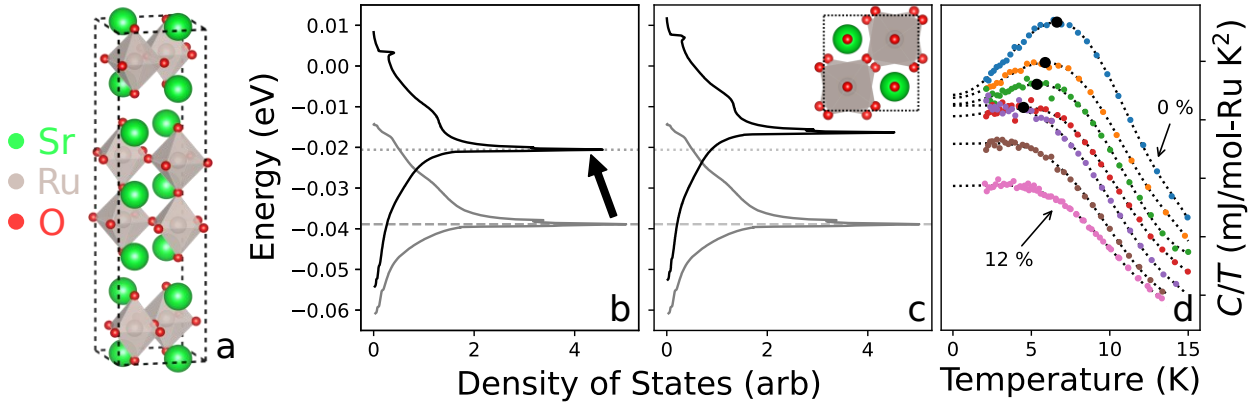


Figure 1. a: *The structure of $Sr_3Ru_2O_7$.* ***b:*** *Calculated density of states changes on adding 8 % Ba based on the HRPD derived structures.* ***c:*** *The effects of selectively activating only the rotational $X_1^-\ B_1^{(a)}$ mode (**inset**) showing it to be driving the observed changes.* ***d:*** *The measured electronic heat capacity highlighting the VHs peak position in black.*

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