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Phase diagram and charge-dynamics of electron-doped osmium based $Ba_2Na_{1-x}Ca_xOsO_6$ spin-orbit-coupled Mott insulator

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In recent years, 5d transition metal oxides have been the focus of increasing research interest, owing to their rich physics emerging from the interplay between electron correlations and strong spin-orbit coupling (SOC). Such SOC-induced insulating phases are frequently accompanied by the transition of the 5d ion to a magnetic state triggered by local structural distortions, in competition with ground states with exotic multipolar ordering [1].
Osmium based double perovskites $Ba_2Na_{1-x}Ca_xOsO_6$ (BNCOO), constitute a remarkable example of SOC-driven physics. In this system, electron doping of Os 7+
by etherovalent substitution of Na by Ca provides a further degree of freedom which strongly affects its magnetic ground state and raises TN from a few
kelvin (x = 0) up to ≈ 40 K (x = 1). 23Na NMR provides evidence that the onset of magnetic order is anticipated by local static distortions of the cubic perovskite cell, breaking the local point symmetry [1].
Here we report on a combined muSR and NMR experiment which allows us to draw the full phase diagram whith both the local magnetic and structural symmetry breaking and ordering phases. In addition unambiguous evidence for the slow diffusion of dynamic lattice distortions in Ca-substituted BNCOO at temperatures well above their magnetic transitions is reported. Their occurrence in conjunction with electron doping support their identification with small polarons [2], as predicted by recent theoretical studies. We argue that such polarons may play a role as the dynamic precursors of the low-temperature static symmetry-breaking distortions which, in turn, seemingly trigger the magnetism in the system.

[1] L. Lu, et al., Nature Communications, 2017, 8, 14407
[2] C. Franchini, M. Reticcioli et al. Nature Reviews Materials, 2021, 6, 560

Primary authors: SANNA, Samuele (University of Bologna); Prof. MITROVIC, Vesna (Brown University); Prof. DE RENZI, Roberto (University of Parma); Dr GARCIA, Erick (Department of Physics, Brown University, Providence, Rhode Island 02912, USA); Dr CONG, Rong (Department of Physics, Brown University, Providence, Rhode Island 02912, USA); TASSETTI, Anna (Dipartimento di Fisica e Astronomia "A. Righi", Università di Bologna, I-40127 Bologna, Italy); FORINO, Paola (Dipartimento di Fisica e Astronomia "A. Righi", Università di Bologna, I-40127 Bologna, Italy); FRANCHINI, Cesare (Dipartimento di Fisica e Astronomia "A. Righi", Università di Bologna, I-40127 Bologna, Italy); Prof. WOODWARD, Patrick (Department of Chemistry and Biochemistry, The Ohio State University, Columbus, Ohio 43210, USA); ALLODI, Giuseppe (Università di Parma)

Presenter: SANNA, Samuele (University of Bologna)

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