Janh-Teller polaron in the spin-orbit multipolar magnetic oxide Ba_2NaOsO_6

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Complex oxides hosting 5d electrons present a variety of exotic phases arising from spin-orbital (SO) interactions and electronic correlation (EC) [1]. In the Mott insulator Ba_2NaOsO_6, a canted antiferromagnet with multipolar interactions [2], strong EC together with Jahn-Teller (JT) lattice activity pave the way for bridging polarons and SO coupling, distinct quantum effects that play a critical role in charge transport and spin-orbitronics [3, 4]. Polarons are quasiparticles originating from strong electron-phonon interaction and are ubiquitous in polarizable materials, especially in 3d transition metal oxides [3]. Despite the more spatially delocalized nature of 5d electrons, we demonstrate the formation of *Jahn-Teller spin-orbital polarons* in electron doped $Ba_2Na_{1-x}Ca_xOsO_6$ by combining ab-initio calculations with nuclear magnetic resonance and muon spin rotation measurements. The polaronic charge trapping process converts the Os $5d^1$ spin-orbital $J_{eff} = 3/2$ levels, characteristic of pristine BNOO, into a $5d^2$ $J_{eff} = 2$ manifold, leading to the coexistence of different J-effective states in a single-phase material. Moreover, we suggest that polaron formation creates robust in-gap states that prevent the transition to a metal phase even at ultrahigh doping, thus preserving the Mott gap across the entire doping range from d^1 BNOO to d^2 Ba_2CaOsO_6.

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