

Highlights

Innovative materials with strong interplay of spin orbital charge and topological degrees of freedom - 2018

Spin-Orbital Excitations in Ca_2RuO_4 Revealed by Resonant Inelastic X-Ray Scattering (RIXS)

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Spin-orbit coupling is a central thread in the search for novel quantum material physics. The strongly correlated insulator Ca_2RuO_4 is considered as a paradigmatic realization of both spin-orbital physics and a band-Mott insulating phase, characterized by orbitally selective coexistence of band and Mott gap. We present a high resolution oxygen K-edge resonant inelastic x-ray scattering study (Fig. 1 (a)) of the antiferromagnetic Mott insulating state of Ca_2RuO_4 . A set of low-energy (about 80 and 400 meV) and high-energy (about 1.3 and 2.2 eV) excitations are reported, which show strong incident light polarization dependence (Figs.1 (b,c)). Our results strongly support a spin-orbit coupled band-Mott scenario and explore in detail the nature of its exotic excitations (Fig.1 (d,e)). Guided by theoretical modeling, we interpret the low-energy excitations as a result of composite spin-orbital excitations. Their nature unveils the intricate interplay of crystal-field splitting and spin-orbit coupling in the band-Mott scenario (Fig.1 (f,g,h)). The high-energy excitations correspond to intra-atomic singlet-triplet transitions at an energy scale set by Hund's coupling. Our findings give a unifying picture of the spin and orbital excitations in the band-Mott insulator Ca_2RuO_4 .

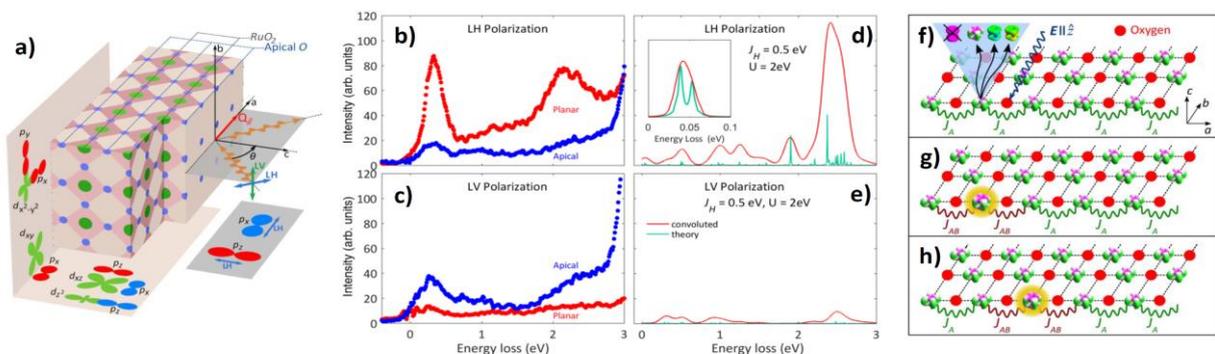


Fig. 1: (a) RIXS geometry with respect to the crystal lattice of Ca_2RuO_4 is displayed schematically. (b,c) RIXS spectra, with the elastic response subtracted, at the apical (blue lines) and planar (red lines) oxygen resonances for the respective light polarizations. (d,e) Calculated RIXS spectra for the planar site with respect to linear horizontal (b) and vertical (c) light polarization. (f) Schematic of an oxygen K-edge RIXS process creating a local excitation at the Ru site. Panels (g) and (h) illustrate propagation of the spin-orbital excitation.