

Title: Molecular orbitals vs. relativistic orbitals in t_{2g} honeycomb lattices: SrRu_2O_6 as compared to Na_2IrO_3 , RuCl_3 , and Li_2RuO_3 .

Abstract: t_{2g} states on a honeycomb lattice tend to form non-dispersive localized states even if large intersite hopping is present. In the nonrelativistic case, these are molecular orbitals (MO) localized on metal hexagons, if the ligand-assisted nearest and next nearest neighbor hoppings, t_1' and t_2' , dominate, or dimers (DO), if the direct overlap, t_1 , dominates. In the ultrarelativistic limit t_{2g} form effective relativistic orbitals (RO), $j^{\text{eff}}=3/2$, which are atomically localized if t_1' is the dominant hopping. On the first glance, the three regimes are defined by the conditions $t_1' \gg t_1, \lambda$, $t_1 \gg t_1', \lambda$, and $\lambda \gg t_1, t_1'$. In reality, the latter condition is never fulfilled, especially in ruthenates, yet not only Na_2IrO_3 , but also RuCl_3 appear to be in a regime dominated by RO, even though the residual effect of MO critically influences magnetic interactions, while Li_2RuO_3 , not far removed from RuCl_3 in the parameter space, is firmly in the DO regime. Most surprisingly, SrRu_2O_6 , which is even closer to RuCl_3 , happens to be fully in the MO regime, with negligible spin-orbit effects. In this talk, we will show that an additional, decisive factor is the doping level per site. The principal difference between Na_2IrO_3 or RuCl_3 , Li_2RuO_3 , and SrRu_2O_6 is that the first two have one t_{2g} hole per site, the second one 2 holes, and the last 3 electrons. In particular, the total dominance of MO in the latter compound fully explains its unique and unexpected magnetic properties. Finally, I will discuss interesting spectroscopic signatures of the MO state.

References: (1) Localized itinerant electrons and unique magnetic properties of SrRu_2O_6 ; S. Streltsov, I. I. Mazin, and K. Foyevtsova. Phys. Rev. B92, 134408 (2015) (2) Spectroscopic signatures of molecular orbitals on a honeycomb lattice. Z.V. Pchelkina, S.V. Streltsov, and I.I. Mazin, arXiv:1607.08847