Density functional and dynamical mean-field study of molecular orbitals in correlated solids: an example of deficient spinel chalcogenides

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Formation of molecular orbitals in crystalline solids, such as √13 × √13 density wave ordering in van der Waals layered 1T-TaS_2 or V-V dimerization in VO_2, provides an interesting platform for the study of strong electron correlations in crystalline transition metal compounds. Lattice distortion and clustering of transition metal ions give rise to the formation of molecular orbitals, as well as renormalization of electronic bandwidth between neighboring molecular orbitals and the promotion of electron correlation effects. While electronically similar to conventional atomic orbitals, these molecular orbitals and the resulting correlated phases can provide an interesting pathway of controlling various degrees of freedom (charge, spin, orbital, and lattice) for the tuning of functional properties in condensed matter systems.

In this talk, I will present our recent density functional and dynamical mean-field studies on a series of deficient spinal chalcogenides GaM_4X_8 (*M* = V, Nb, Mo, Ta, *X* = S, Se, Te). Especially, I will focus on the role of internal energy scales, such as atomic spin-orbit coupling and Hund's coupling terms, in determining the molecular orbital degrees of freedom and the resulting ground state properties of deficient spinel compounds. First, in the 5*d* transition metal compounds $GaTa_4Se_8$, it will be shown that the atomic spin-orbit coupling in Ta *d*-orbital transforms into molecular spin-orbit coupling term, which makes this compound a promising candidate for topological superconductivity. Secondly, in the 3*d* counterpart GaV_4S_8, a nontrivial relation between the Hund's coupling and the V_4S_4 tetrahedral clustering will be suggested based on our recent cluster dynamical mean-field theory calculation results.