**Revisit to the molecular *J*eff ground state of lacunar spinel compounds: A charge-density functional plus *U* study**

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After the discovery of the *J*eff = 1/2 ground state in Sr2IrO4 [1], spin-orbit entangled *J*eff state has attracted a great deal of attention. This intriguing ground state can exhibit exotic phases of matter such as unconventional superconductivity [2] and quantum spin liquid [3]. The previous theoretical study predicted the molecular *J*eff ground state in the lacunar spinel compounds, GaM4X8 (M = Nb, Mo, Ta and W; X = S, Se and Te) [4]. Furthermore, RIXS (resonant inelastic X-ray scattering) experiment together with the first-principles calculations reported the *J*eff = 3/2 ground state in GaTa4Se8 [5]. Recently, it was reported that widely-used DFT+*U* functional based on the spin-density could easily lead to unphysical results due to the competition between the spin-density energy and the double counting term [6]. In order to avoid such an artifact, charge-density based DFT+*U* functional was suggested to be the desirable choice. Since all the previous studies of lacunar spinel compounds were performed within spin-density framework, it is strongly requested to re-investigate the previous results. In this study, by introducing a new parameter, *J*eff-ness, we analyzed the *J*eff electronic structure with respect to Hubbard *U* and Hund *J* within charge-densty framework. As a result, we confirmed the robustness of the molecule *J*eff ground states in the lacunar spinel compounds.

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