Electronic, Magnetic Structure and Orbital Ordering in CdV_2O_4 from First Principles

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Abstract – First principles calculations using the APW+lo method, as implemented in the WIEN2k code, have been used to investigate the structural, electronic and magnetic properties of CdV_2O_4 within generalized gradient approximations (GGA), GGA+U and GGA+U+SO approach, especially as regards orbital ordering. The calculations were performed for paramagnetic, ferromagnetic and antiferromagnetic configurations using cubic and two kinds of magnetic structure (AFM-I and AFM-II, **Fig. 1**), respectively. The GGA results falsely gave ferromagnetic half-metallic ground state. Exciting, our GGA+U method rectify this error and revealed that the ground state is AFM-I configurations, which is Mott insulator with a staggered $d_{xy}^1 d_{xz}^1$ (V_2) and $d_{xy}^1 d_{yz}^1$ (V_1) orbital ordering. However, especially interesting, our GGA+U+SO method obtained AFM-II ground state with a staggered V_1 ($d_{xz}+id_{yz}$) and V_2 ($d_{xz}-id_{yz}$) orbital ordering. Analysis shows that electron correlation, spin-orbital coupling and co-operative Jahn-Teller distortions play a significant role in determining the orbital ordering.

Spin, charge, orbital and lattice degrees of freedom play important roles in electronic, magnetic and transport properties of transition metal-oxide. The origin of these complex natures is presently a debate subject, especially on geometrically frustrated lattice systems [**1**]. Vanadium spinel-type AV_2O_4 [**2-4**] belongs to frustrated antiferromagnets and show many interesting phenomena such as Heavy-Fermi behavior; two successive phase transitions, complicated charge and orbital ordering, which extensively enrich the physical properties of vanadium-oxide.

CdV_2O_4 is Mott insulator where magnetic ions V^{3+} are characterized by orbital degenerate due to partly occupancy of t_{2g} orbital ($n_{t2g} = 2$). These partially filled t_{2g} orbitals leave the orbital degrees of freedom open and have the possibility of orbital order. Otherwise, to the best of our knowledge, the experimental investigation was limited to a pseudotetramer proposition [**5**] and sporadic doped systems, no theoretical study have been performed on it so far, therefore first-principles calculations are urgent to understand structure and magnetic properties.

We have used FP-LAPW+lo within GGA, GGA+U and GGA+U+SO to make a detailed analysis of the densitiy of states (DOS) for this system in the paramagnetic (PM, cubic), ferromagnetic (FM,cubic), antiferromagnetic (AFM-I and AFM-II Fig. 1) states. The GGA results falsely gave ferromagnetic half-metallic ground state. Exciting, our GGA+U method rectify this error and revealed that the ground state is AFM-I configurations, which is Mott insulator with a staggered $d_{xy}^1 d_{xz}^1$ (V_2) and $d_{xy}^1 d_{yz}^1$ (V_1) orbital ordering. However, especially interesting, our GGA+U+SO method obtained AFM-II ground state with a staggered V_1 ($d_{xz}+id_{yz}$) and V_2 ($d_{xz}-id_{yz}$) orbital ordering (**Fig. 2**), which need experimental results to confirm.

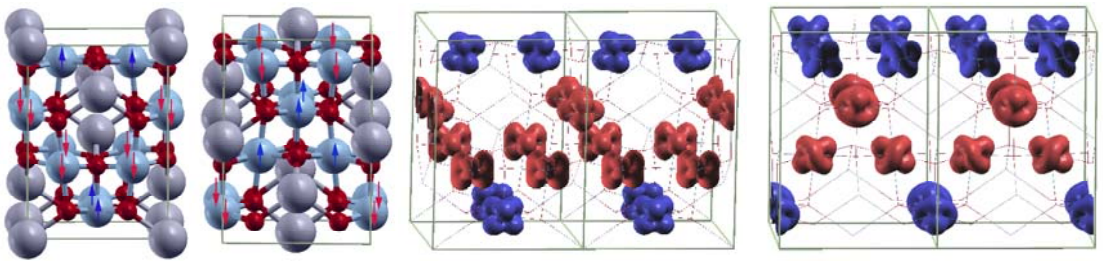

Fig.1. Two kinds of different magnetic structure of CdV_2O_4.

Fig.2. Spin density plot [isosurface at 0.1 e/Å^3 produced using XcrySden] red and blue denote spin-up and spin-down respectively.

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