Spin and orbital structures in transition-metal compounds with face-sharing octahedra

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The study of correlated systems with orbital ordering (OO) is currently a very active field of research in solid state physics. OO is not only accompanied (or caused) by structural transitions, but it also largely determines magnetic properties of many materials, e.g. transition metal (TM) oxides.

For the doubly degenerate case in simple lattices as that of perovskites AMO_3 with MO_6 octahedra having common corner, the typical situation is such that ferro-orbital ordering gives rise to the antiferromagnetic spin exchange, whereas antiferro-OO is rather favorable for spin ferromagnetism. In many different situations in solids, with different geometries of the lattice, and this general paradigm (ferro-orbital–antiferro-spin and *vice versa*) may not, and does not work in other cases. One is that with neighboring TM ions having two common oxygens; for systems with MO_6 octahedra, this is the case of common edge, with ~90° M-O-M bonds.

There exists, however, the third, much less studied situation – that with neighboring MO_6 octahedra having common face. Here, the superexchange occurs via three oxygens. The situation with the orbital ordering in this case and the form of the resulting (spin and orbital) exchange is practically not studied. Nevertheless, experimentally there are many TM compounds with such geometry. Such are for example hexagonal crystals $BaCoO_3$ or $CsCuCl_3$, containing infinite columns of face-sharing octahedra; many other similar systems have finite face-sharing blocks, e.g. $BaIrO_3$ or $BaRuO_3$.

Here, we consider the form of the spin-orbital superexchange for TM with double or triple orbital degeneracy for neighboring TM ions with face-sharing octahedra. One surprising result of our study is that, whereas for doubly degenerate system of perovskite type with 180° M-O-M bonds, the form of orbital term in the Hamiltonian is rather complicated, for common face the symmetric model is realized, with the orbital interaction of the Heisenberg type[1].

Often in this geometry, the MO_6 octahedra have trigonal distortions. Such local distortions lead to splitting of t_{2g} orbitals into an a_{1g} singlet and e_g^π doublet. We show that for the partially filled e_g^π doublet, the resulting superexchange is very similar to the case of "real" e_g electrons, but here the orbital moment is not quenched and the real relativistic spin-orbit leads to rather non-trivial effects, which we also discuss in detail.

[1] Kugel K.I., Khomskii D.I., Sboychakov A.O., Streltsov S.V. (2015), *Phys. Rev. B*, 91, 155125