ABSTRACT

Coupling between charge, orbital and spin degrees of freedom in transition metal oxides give rise to a plethora of exotic physical properties. The thesis is devoted to investigations on some titanate spinel (ATi_2O_4) compounds, which exhibit such intricate couplings. $MgTi_2O_4$ possesses a chiral ground state (which is very uncommon in inorganic compounds) which hosts a tetramer orbital-order (higher-order orbital ordering is very unconventional) along with a spin-singlet magnetic state. Combining experiments with density functional theory calculations, we establish a unique chiral ground state in another titanate spinel $MnTi_2O_4$ which, unlike $MgTi_2O_4$, contains a magnetic A site (the role of which is found crucial). $MnTi_2O_4$, which contains orbitally active Ti^{3+} ions, hosts a unique higher-order tetramer orbital order (qualitatively different from $MgTi_2O_4$) involving all three Ti t_{2g} orbitals. Further, a ferrimagnetic ground state (driven by antiferromagnetic Mn-Ti exchange interactions), where the Ti sublattice orders ferromagnetically, is realized in $MnTi_2O_4$. While both $MgTi_2O_4$ and $MnTi_2O_4$ are insulating in their orbital ordered ground state, we demonstrate a unique current or electric field induced metallic state (with a very low threshold electric field, thus, promising for multiple applications) in the corresponding V-doped compounds. A Joule-heating driven mechanism for the resistive transition is ruled out. V^{3+} (Jahn-Teller active ion) ion doping at the Jahn-Teller active Ti^{3+} site in $(Mg/Mn)Ti_2O_4$ induces a unique cooperation effect, where spontaneous electron transfer occurs from some of the Ti^{3+} ions to V^{3+} causing both the resultant ions (Ti^{4+} and V^{2+}) to lose their orbital degrees of freedom. While strain energy (due to much larger V^{2+} ion) prevents spontaneous transfer of electrons between all the Jahn-Teller active ion pairs, the same can be excited in presence of a large current flow (as suggested through calculated density-of-states) to establish a metallic state. Further, we also investigate the presence of a superconducting transition of a surface layer formed on a V-doped $MgTi_2O_4$ sample at reasonably high temperatures, thereby, highlighting the promise of this class of materials in hosting multiple functional properties.

Key Phrases: Jahn-Teller Effect; Orbital Ordering; Current induced metallic state.