ABSTRACT

Various physical phenomena and ground state properties of two important correlated electronic systems, Vanadate spinel oxides (FeV_2O_4, MnV_2O_4) and the Oxygen-deficient layered perovskite compound ($YBaCuFeO_5$) have been studied in this thesis. These phenomena are primarily driven by the interplay between orbital, spin and lattice degrees of freedom.

A long-range orbital order as a function of doping in Mn-doped FeV_2O_4 is investigated by using first-principles density functional theory calculations in the presence of Coulomb correlations and spin-orbit interactions. Through a detailed analysis of corresponding Wannier orbital projections of the vanadium d-bands, we have established that for $x \leq 0.6$, the orbital order at V sites is $d_{xz} \pm d_{yz}$ type. On the contrary, for $x > 0.6$, it is the d_{xz} or d_{yz} orbital that orders at V sites in the successive ab planes along c direction (so-called A-type ordering). At the Fe sites, an ordering of $d_{x^2-y^2}$ orbitals for $x \leq 0.6$ and d_{z^2} orbitals for $x > 0.6$ is found. The effect of spin-orbit interaction on orbital ordering is found to be not significant.

Next, we study the effects of magnetic ordering and Coulomb correlation on the lattice vibrations of the low-temperature tetragonal structure of MnV_2O_4 and calculate the phonon dispersion relations in two different magnetic orders (ferrimagnetic and ferromagnetic). Further, we have calculated the Raman intensities to understand the polarization-dependence of the B_g modes already observed experimentally. Finally, we have estimated the spin-phonon coupling across the Brillouin zone. Temperature dependence of the Raman intensity peaks has been analyzed from the strong spin-phonon coupling, observed at the high symmetry points of the Brillouin zone.

Experimental results on $YBaCuFeO_5$, in its incommensurate magnetic phase, appear to disagree on ferroelectric response. By using first-principles calculations for the parent compound within LSDA + U + SO approximation, we reveal the nature of the spiral state in $YBaCuFeO_5$. The helical spiral is found to be more stable below the transition temperature as spins prefer to lie in the ab plane. While the Dzyaloshinskii-Moriya (DM) interaction turns out to be negligibly small, the spin current mechanism does not work in the helical spiral state ruling out an electric polarisation. We also investigate the magnetic transition in $YBa_{1-x}Sr_xCuFeO_5$ for the entire range ($0 \leq x \leq 1$) of doping. The exchange interactions are estimated as a function of doping and our quantum Monte-Carlo (QMC) calculations on the resulting effective spin Hamiltonian show that the paramagnetic to commensurate phase transition temperature increases with doping till $x = 0.5$ and decreases beyond. This conforms well to the extant experimental observation.