## II. COMPUTATIONAL DETAILS

The nonmagnetic and ferromagnetic electronic structure calculations of $Sr_2CoO_4$ compound were carried out using LmtArt 6.61.[9] For calculating charge density, full-potential linearized Muffin-Tin orbital (FP-LMTO) method working in plane wave representation was employed. In the calculations, we have used the Muffin-Tin radii of 2.915, 1.965, 1.608, and 1.778 a.u. for Sr, Co, O1, and O2, respectively. The charge density and effective potential were expanded in spherical harmonics up to $l = 6$ inside the sphere and in a Fourier series in the interstitial region. The initial basis set included $5s$, $4p$, and $4d$ valence, and $4s$ semicore orbitals of Sr; $4s$, $4p$, and $3d$ valence, and $3p$ semicore orbitals of Co, and $2s$ and $2p$ orbitals of O. The exchange correlation functional of the density functional theory was taken after Vosko $et$ $al.$[10] and GGA calculations were performed following Perdew $et$ $al.$[11]

The effect of on-site Coulomb interaction ($U$) under GGA+$U$ formulation of the density functional theory is also considered in the calculations. The detailed description of the GGA+$U$ method implemented in the code can be found in Ref. [12]. The double counting scheme used in the code is normally called as fully localized limit in the literature. To study the role of orbital degrees of freedom, we have also included spin-orbit coupling (SOC) in the calculations. The Self-consistency was achieved by demanding the convergence of the total energy to be smaller than $10^{-5}$ Ry/cell. (10, 10, 10) divisions of the Brillouin zone along three directions for the tetrahedron integration were used to calculate the density of states (DOS).

## III. RESULTS AND DISCUSSIONS

The atomic arrangement in the unit cell is shown in Fig. 1, which displays bodycenter tetragonal lattice. The Co atoms occupy the corners and bodycenter positions and each Co atom is surrounded by six O atoms forming a distorted octahedron. In this structure there are two kinds of O represented by O1 and O2. O1 lies in the $ab$ plane and O2 along the $c$-axis. The bond distance between the corner and bodycenter Co atoms is almost 1.8 times larger than that between nearest Co atoms sitting at the corners. This suggests the quasi-two-dimensional nature of the system where its electronic properties are expected to be decided by transport of electrons in the $ab$ plane.

In order to know the crystal-field effect and nature of Co-O bonding in $Sr_2CoO_4$ we have plotted the partial density states (PDOS) of Co $3d$ and O $2p$ obtained from nonmagnetic solution in Fig. 2. In the octahedral symmetry Co $3d$ states split into $t_{2g}$ and $e_g$ states and the separation between these states is found to be $\sim$ 1.5 eV. The degeneracies of these states are further lifted in tetragonal symmetry as evident from Figs. 2(a)
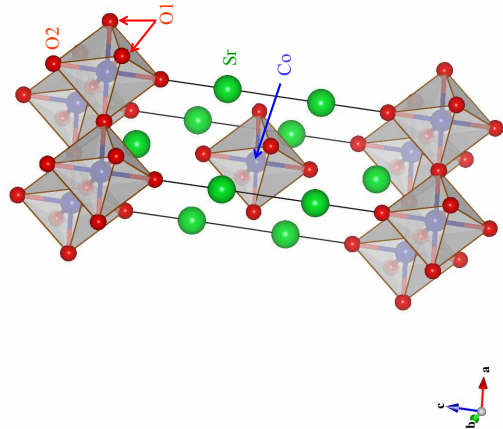


FIG. 1: (Color online) Atomic arrangements in the unit cell. Sr, Co and O atoms are represented by spheres with decreasing radii.

and (b). The triply degenerate $t_{2g}$ states split into doubly degenerate ($d_{xz}$, $d_{yz}$) states and nondegenerate $d_{xy}$ state. Similarly, doubly degenerate $e_g$ states split into nondegenerate $d_{x^2-y^2}$ and $d_{z^2-r^2}$ states. The O $2p$ orbitals also split in doubly degenerate ($p_x$, $p_y$) orbitals and nondegenerate $p_z$ orbital as evident from Figs. 2(c) and (d).

The $t_{2g}$ and $e_g$ sectors are spread over the energy range of about 5 and 10 eV, respectively. Roughly two times larger extent of the $e_g$ states is due to larger overlap between $e_g$ and $p$ orbitals in comparison to that between $t_{2g}$ and $p$ orbitals. In the present situation one can get maximum overlaps of $d_{x^2-y^2}$ and O1 ($p_x$, $p_y$) orbitals; and $d_{z^2-r^2}$ and O2 $p_z$ orbitals which can lead to larger bandwidth of $e_g$ symmetric states. The occupied $d_{z^2-r^2}$ states are mostly found in the narrow region of 1 eV whereas occupied $d_{x^2-y^2}$ states are spread over an energy window of about 4 eV. Total number of $d$ electrons is found to be $\sim$ 6.5, which is about 1.5 more than the expected nominal value for $Co^{4+}$ ion. The above observations can be considered as a signature of the covalent nature of the Co-O bonds. There are large Co $3d$ PDOS at the $E_F$ ($\sim$ 8.4 states/eV/atom) in the nonmagnetic solution which may be considered as a signature of ferromagnetic ground state based on the Stoner theory.