

Study of half-metallic behavior in Sr_2CoWO_6 perovskite by *ab initio* DFT calculations

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Abstract

In this work, we report several *ab initio* calculations performed for Sr_2CoWO_6 by means of the density functional theory and the linearized augmented plane wave method for both spin orientations. For calculations, the exchange and correlation potential were treated into the generalized gradient approximation, which permits to consider from the beginning the difference between the electronic densities for both up and down spin orientations. The densities of states are calculated by the histogram method and the positions of Fermi levels are found by integrating over the density of states for both spin configurations. Our results reveal that Sr_2CoWO_6 material behaves as insulators for the spin-up orientation and conductor for the spin down, as expected for the half-metallic systems. Results of partial densities of states permit to conclude that the conduction band has predominant contributions of $d_{x^2-y^2}$ and d_{xz+y^2} states of Co for the spin-down orientation. A magnetic moment of $3 \mu_B$ was calculated. From the Murnaghan equation state, we also calculate the cell dimensions that minimize the total energy for several configurations.

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1. Introduction

In the last decade, investigations of perovskite oxide materials have been increased because of their varied structure, composition and physical characteristics. These materials have attracted intense research activities in many applied and fundamental areas of solid-state science and advanced materials research due to the exotic properties such as high-temperature superconductivity [1], colossal magnetoresistance [2], half-metallicity [3] and magnetoelectricity [4]. The complex perovskite oxides generally have the formula $\text{A}_2\text{MM}'\text{O}_6$ and result from the ordering of M and M' cations on the octahedral site of the primitive perovskite unit cell. Magnetic complex perovskites $\text{A}_2\text{MM}'\text{O}_6$, where A is an alkaline earth and M, M' are magnetic and non-magnetic transition metals, respectively, were discovered by Longo and Ward in 1960s

[5]. However, the half-metallic feature of the $\text{Sr}_2\text{FeMoO}_6$ was only established by Kobayashi et al. in 1998 [3]. This exotic property is characterized by the differentiated conducting response of the spin-up and spin-down orientations.

The density of states as a function of energy clearly evidences that majority spin component shows a energy gap at the Fermi level, as the insulating materials, and the other spin orientation is continuous at the Fermi level, due to the strong hybridization of Fe-3d(t_{2g}) and O-2p states. The extensive half-metallicity studies in double perovskite materials are related with the probable technological applications in *spintronic* devices, such as spin valves, sources for spin polarized electrons and magnetic information storage systems. The aim of this work is to carry out a detailed *ab initio* theoretical study of the complex perovskite Sr_2CoWO_6 compound, which was experimentally analyzed and reported as an antiferromagnetic half-metallic material with low Néel temperature (24 K) [6].

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2. Calculation method

Calculation of band and electronic structure for the complex perovskite Sr_2CoWO_6 can be seen as a many body problem of ions and electrons. These were performed by employing the FP-LAPW method, in the framework of DFT and implemented in the WIEN97 code [7]. The FP-LAPW consists in the calculation of solutions for the Kohn–Sham by the first principles method. In the calculations reported here, we use a parameter $RMTK_{\text{max}} = 8$, which determines matrix size (convergence), where K_{max} is the plane wave cut-off and RMT is the smallest of all atomic sphere radii. We have chosen the muffin-tin radii (MT) for Sr, Co, W and O to be 2.5, 1.95, 1.95 and 1.73, respectively. The exchange and correlation effects were treated by using the generalized gradient approximation (GGA) [8]. This potential considers the difference between the electronic densities for the two distinct spin orientations from the beginning. The self-consistent calculations are considered to be convergent when the total energies of two successive iterations agreed to within 10^{-4} Ry. We adjusted the Fermi energy to zero. The integrals over the irreducible Brillouin zone are performed up to 196 k-points.

3. Results and discussion

The tolerance factor (f) determines theoretically the perovskite structure by means of the ionic radii of each element in the compound [9]. In order to obtain the most accurate results, we have determined the optimal lattice parameters corresponding to the minimal energy value. This is the total energy as a function of volume, which is shown in Fig. 1. For the Sr_2CoWO_6 compounds total energy was calculated by fittings with the equation state of Murnaghan. From total energy as a function of volume we determine the ideal lattice parameters, a and c . Calcula-

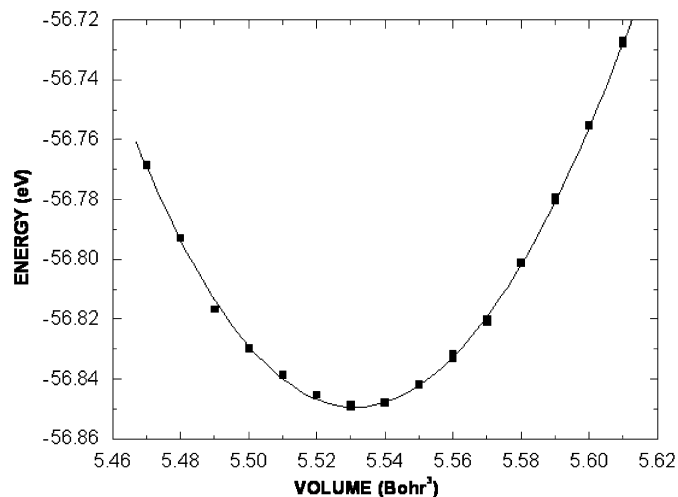


Fig. 1. Total energy as a function of volume for Sr_2CoWO_6 by considering the $I4/m$ space group.

tions were performed by using the tolerance factor and choosing the tetragonal crystallographic structure $I4/m$ (#87). We obtain lattice parameters $a = 5.531 \text{ \AA}$ and $c = 7.860 \text{ \AA}$, which are in accordance with experimental results reported in Ref. [6].

As shown in Fig. 2, for the total density of states close to the Fermi level, we observe the half-metallic nature of Sr_2CoWO_6 compound: the spin-down channel has an isolated behavior, with a gap of 1.07 eV through the Fermi level, while the spin-up evidences a conductor feature.

There are states of Co in the spin-up and -down channels of the valence band. However, in the conduction band, the spin-up state is relatively empty while the spin-down is continuous across the Fermi level and evidences an intragap of 0.9 eV on the valence band. Particularly, it is observed that the Co is responsible by the half-metallic effect in this perovskite compound. On the other hand, for W, there are not states close to the Fermi level for both valence and conduction bands. Calculations of partial DOS for Co-3d and W-5d levels were performed. For spin-down configuration of Co there are $d_{x^2-y^2}$ and d_{xz+yz} states in the conduction band between -1.09 eV and 0.28 eV , which have an important contribution to the total DOS; d_{z^2} and d_{xy} states are at the minor energy region. For spin-up

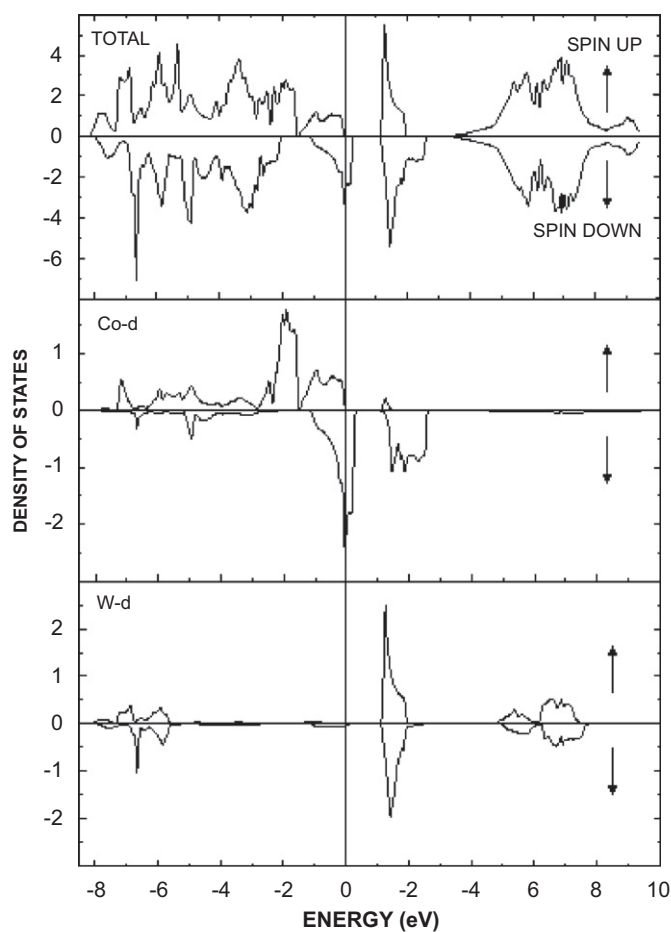


Fig. 2. Total density of states for up (above) and down (below) spin configurations, on the Sr_2CoWO_6 compound.

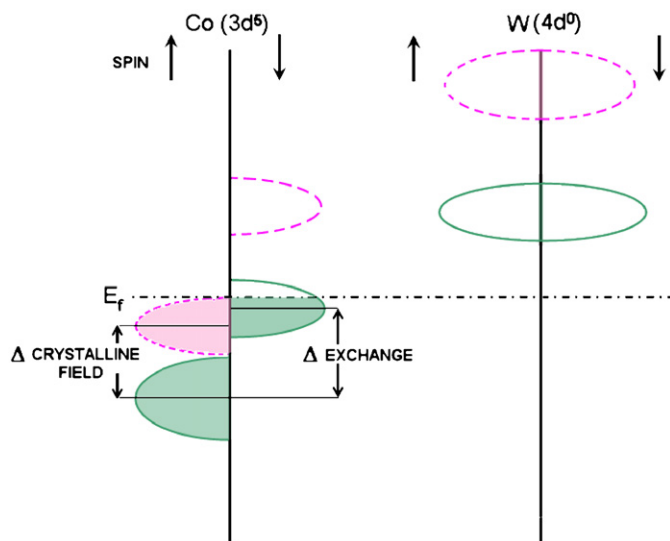


Fig. 3. Distribution of Co-d and W-d states for the up and down spin configurations, on the Sr_2CoWO_6 material. Fill and dot lines represent minor and main energy states, respectively.

orientation, d_{z^2} and d_{xy} states are distributed from -1.49 to 0 eV, close to Fermi level. Other states ($d_{x^2-y^2}$ and d_{xz+yz}) are localized at the minor energy regime. Results for W show that this element has not a significant spin-up or -down contribution to the DOS. Moreover, there are not occupied states by the W-5d level. Then, we assume that appropriated valences of these elements are Co^{2+} and W^{6+} .

In the case of Co, we observe a crystalline-field splitting of 1.25 eV for spin-up channel and 1.91 for spin-down orientation. Additionally, exchange splitting of 0.97 eV and 1.94 were determined for the high and low energy states, respectively. For W, a great crystalline-field splitting of 5 eV was observed in the up and down spin channels. These results are summarized in the scheme of Fig. 3, which shows Co-d and W-d levels for both up and down spin orientations. Calculations permitted to obtain the effective magnetic moment of system to be $3 \mu_B$, which is lower when compared with the experimental report [6].

4. Conclusions

Theoretical study of electronic properties of Sr_2CoWO_6 perovskite was carried out by the FP-LAPW method. Crystallographic parameters of the tetragonal perovskite were optimized by minimization of energy as a function of volume. Results are in agreement with experimental reports. By introducing spin polarization, calculation results reveal the half-metallic character of material, evidencing a conductor behavior for the spin-down, spin-channel and isolating feature for spin-up orientation. About the conduction mechanism, we conclude that the large splitting between spin-down states of Co and W make difficulties for the electronic hopping between them. For this reason, we consider that the intermediation of oxygen plays an important role in the electronic conduction because the hybridization between Co and W is small.

Acknowledgments

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