



# The electronic and magnetic properties of $\text{Sr}_2\text{MnNbO}_6$ , $\text{Sr}_2\text{FeMoO}_6$ and $\text{Sr}_2\text{NiRuO}_6$ double perovskites: An LSDA + U + SOC study

Salah Eldin Ashamallah Yousif<sup>a</sup>, O.A. Yassin<sup>b,c,\*</sup>

<sup>a</sup> Department of Applied Physics, Faculty of Applied Sciences, P.O. Box 1343, Taibah University, Almadeenah Almonawarah, Saudi Arabia

<sup>b</sup> Department of Physics, Faculty of Science, P.O. Box 344, Taibah University, Almadeenah Almonawarah, Saudi Arabia

<sup>c</sup> School of Physics and Applied Physics, Al-Neelain University, Khartoum 11121, Sudan

## ARTICLE INFO

### Article history:

Received 13 June 2010

Received in revised form 3 July 2010

Accepted 6 July 2010

Available online 14 July 2010

### Keywords:

Magnetic oxides  
Electronic band structure  
Spin-orbit effects

## ABSTRACT

The electronic structure and magnetic properties of pseudo-cubic and tetragonal double perovskite oxides  $\text{Sr}_2\text{MnNbO}_6$ ,  $\text{Sr}_2\text{FeMoO}_6$  and  $\text{Sr}_2\text{NiRuO}_6$ , have been determined by using the full potential linear augmented plane wave method (FP-LAPW) employing the LSDA + U scheme. The spin-orbit coupling (SOC) has been taken into consideration. These calculations showed that all the three compounds in their two crystalline structures are half metallic. The values of the total energy have indicated that first two compounds prefer the tetragonal structures while the third one prefers the pseudo-cubic structure. The obtained magnetic moments indicate that  $\text{Sr}_2\text{MnNbO}_6$  and  $\text{Sr}_2\text{NiRuO}_6$  are ferromagnetic while  $\text{Sr}_2\text{FeMoO}_6$  has ferrimagnetic ordering. The effect of the spin-orbit coupling was seen to grow from quenched to weakly unquenched to unquenched as going from Nb to Ru and from Mn to Ni as signified by the values of the orbital magnetic moments of the 3d and 4d orbitals.  $\text{Sr}_2\text{NiRuO}_6$  shows half-metallic conduction and ferromagnetism in accordance with the superexchange interaction  $e_g^2 - 0 - e_g^0$  of Goodenough–Kanamori–Anderson (GKA).

© 2010 Elsevier B.V. All rights reserved.

## 1. Introduction

Double perovskite compounds, have attracted intense research activities in many theoretical and applied areas of solid state physics and materials science due to their interesting structural, magnetic and electronic properties [1,2]. The general chemical formula of double perovskite oxides can be expressed as  $\text{Sr}_2\text{BB}'\text{O}_6$ , where A is alkaline earth element such as Sr, Ca, Ba, etc. and B site is occupied by the first row of the 3d magnetic elements in the periodic table. The B' site is occupied by the 4d non-magnetic elements, with O atom located in between forming alternate  $\text{BO}_6$  and  $\text{B}'\text{O}_6$  octahedra and B–O–B' bonds [3,4]. The wide range of these compounds is due to variation of the magnetic and non-magnetic B and B' elements as well as the A-site cations.

Numerous double perovskite compounds were prepared. Their crystal structures vary from cubic, tetragonal to monoclinic, i.e. the most stable crystal structures. Forming of these types of crystal structures depend on the ionic size of the A, B and B' sites. For example when A is small, A = Ca, the obtained crystal structure vary from tetragonal to monoclinic [5]. On the other hand, most of the Sr

and Ba based compounds were found to be cubic or tetragonal. Kato et al. studied the crystal structure of  $\text{A}_2\text{MReO}_6$  (A = Ca, Sr, M = Mg, Sc, Cr, Mn, Fe, Co, Ni and Zn). They found that, the lattice with small A-site Ca is monoclinic and the Sr based compounds are tetragonal or cubic [5]. The crystal structure was found to affect the magnetic properties of the double perovskite oxides.

The half-metallic ground state character is a generic feature for a wide range of the ordered double perovskite oxides. This character is caused by the indirect 3d–O–4d–O–3d interactions such as Fe–O–Re–O–Fe, which is responsible for their magnetic identity [3]. This has opened up intensive investigations on the possible occurrence of the half-metallic properties as new conducting property that is of promising applications in technology.

The compound  $\text{Sr}_2\text{FeReO}_6$  was suggested by Jeng and Guo to be ferrimagnetic [3] and existence of ferromagnetism in  $\text{A}_2\text{CrWO}_6$  (A = Sr, Ba, Ca) which is stabilized by a hybridization of states of the non-magnetic W site positioned in between the high spin Cr sites [6]. Terakura [7] and Kawanaka [8] had shown that  $\text{Sr}_2\text{FeWO}_6$  is antiferromagnetic system. The magnetic properties of these compounds depend on the electronic configurations of the 3d and 4d orbitals. Thus the double perovskite oxides (DPO) can have either ferromagnetic or antiferromagnetic coupling or both together. In general, the antiferromagnetic coupling in DPO is weak due to the long exchange path [9].

The spin-orbit coupling (SOC) is known to be very important in the calculations for total magnetic moment of the elemental

\* Corresponding author at: Department of Physics, Faculty of Science, P.O. Box 344, Taibah University, Almadeenah Almonawarah, Saudi Arabia. Tel.: +966 4 8460008; fax: +966 4 8470235.

E-mail address: [oyassin@ictp.it](mailto:oyassin@ictp.it) (O.A. Yassin).

**Table 1**

The lattice parameters of the pseudo-cubic and the tetragonal structures.

Compound	Lattice parameter of pseudo-cubic		Lattice parameter of tetragonal	
	a (Å)	c (Å)	a (Å)	c (Å)
Sr <sub>2</sub> MnNbO <sub>6</sub>	7.9767	7.9847	5.5672	7.9767
Sr <sub>2</sub> FeMoO <sub>6</sub>	7.9667	7.9747	5.5643	7.9667
Sr <sub>2</sub> NiRuO <sub>6</sub>	7.9347	7.9426	5.5554	7.9347

constituents of the double perovskite especially the 3d and 4d orbitals which play major roles in the predicted properties. It is well known that if the d orbital is more than half filled, the spin and orbital magnetic moments are parallel, otherwise they would be antiparallel [3,10]. Jeng and Guo, had found that the spin and orbital magnetic moments ( $\mu_l$ ) of Fe in the compound Sr<sub>2</sub>FeMoO<sub>6</sub> are parallel due to the over half-filled of the 3d orbital, while for the Cr element in Sr<sub>2</sub>CrWO<sub>6</sub> they are antiparallel due to the less than half filled 3d orbital according to the Hund's rule [3]. The values of the orbital magnetic moments are found in Sr<sub>2</sub>FeReO<sub>6</sub>, Sr<sub>2</sub>CrReO<sub>6</sub> (Re:  $\mu_l = 0.18 \mu_B$ ) and Sr<sub>2</sub>CrWO<sub>6</sub> (W:  $\mu_l = 0.12 \mu_B$ ) which resembles unquenched spin-orbit coupling of the 5d orbitals [3,11]. These three compounds have reported to be cubic while Sr<sub>2</sub>FeMoO<sub>6</sub> is tetragonal. The cubic structure of the DPO is very often distorted as a consequence of internal pressure due to cation size mismatch in both the A- and the B-sites that lower the symmetry to tetragonal or even to monoclinic (when the A-site is occupied by smaller cation, e.g. Ca). This distortion alters the B–O–B' angle and consequently changes the charge transfer and the exchange interaction between B and B' ions that govern the conduction and magnetic properties of the DPO.

In the present work, we study a series of compounds in which the selected B(3d) cations are Mn, Fe and Ni and the B'(4d) cations are Nb, Mo and Ru, with adopting the pseudo-cubic and tetragonal structures. To avoid the center of symmetry in cubic structure, the c-axis value has been increased by 0.1% along the c-axis. Since the tetragonal structure is reported experimentally to be one of the most stable structures for DPO, it would be wise to include this structure in the DOS calculations. This has resulted in the following compounds; Sr<sub>2</sub>MnNbO<sub>6</sub> (Mn<sup>3+</sup>(3d<sup>4</sup>:t<sub>2g</sub><sup>3</sup>e<sub>g</sub><sup>1</sup>)) and (Nb<sup>5+</sup>(4d<sup>0</sup>:t<sub>2g</sub><sup>0</sup>e<sub>g</sub><sup>0</sup>)), Sr<sub>2</sub>FeMoO<sub>6</sub> (Fe<sup>3+</sup>(3d<sup>5</sup>:t<sub>2g</sub><sup>3</sup>e<sub>g</sub><sup>2</sup>)) and (Mo<sup>5+</sup>(4d<sup>1</sup>:t<sub>2g</sub><sup>1</sup>e<sub>g</sub><sup>0</sup>)), and Sr<sub>2</sub>NiRuO<sub>6</sub> (Ni<sup>3+</sup>(3d<sup>7</sup>:t<sub>2g</sub><sup>5</sup>e<sub>g</sub><sup>2</sup>) and Ru<sup>5+</sup>(4d<sup>3</sup>:t<sub>2g</sub><sup>3</sup>e<sub>g</sub><sup>0</sup>)).

## 2. Computational details

The pseudo-cubic and tetragonal crystal structures are adopted for the present calculations. The structural data of the studied double perovskite oxides are generated from the Structure Prediction Diagnostic Software (SPuDS) [12] and are shown in Table 1.

It is well known that, the full potential linear augmented plane wave (FP-LAPW) is an accurate method in electronic structure calculations [13]. This method is implemented in WIEN2k software where no shape approximation in charge, potential and wave function expansion are considered [13]. In the present work, the electronic structure calculations were performed by using WIEN2k. The LSDA are used as the exchange correlation potential with inclusion of on-site coulomb interaction (U). Here U is set to 4.0 eV for

the 3d ions while the 4d ions are weakly correlated with U less than 1.0 eV. The  $R_{MT}K_{MAX}$  is 7.0 and the expansion was taken up to  $l = 10$ . The muffin-tin sphere used are given in Table 2. In the first Brillouin zone (BZ), 126 k-points are used for pseudo-cubic and 186 k-points for tetragonal in all compounds and the convergence of the total energy was better than  $10^{-4}$  Ry. The spin-orbit coupling (SOC) was included on the basis of the second variational method by using a scalar relativistic wave functions.

## 3. Results and analysis

The differences in the shapes of the DOS may be related to four factors; the first is the crystal structure and the second is electronic charge distribution in the energy levels, the third is coulomb interaction (U) and the fourth is the strength of the spin-orbit coupling. This is also valid to the 4d elements. The variations in the shape of the DOS are thus expected to be around the Fermi level.

### 3.1. The electronic structure in the pseudo-cubic crystal structure

Here, the pseudo-cubic structure is preferred than the cubic one so as to properly include the SOC to avoiding a center of inversion in the crystal lattice. The lattice parameters generated by SPuDS for a cubic structure is increased by 0.1% along the c-axis to construct the pseudo-cubic lattice. The calculated densities of states (DOS) of Sr<sub>2</sub>MnNbO<sub>6</sub>, Sr<sub>2</sub>FeMoO<sub>6</sub> and Sr<sub>2</sub>NiRuO<sub>6</sub>, in their ordered pseudo-cubic structure are shown in Fig. 1 for LSDA+U+SOC. In this scheme, the LSDA+U+SOC, the Fermi level is seen to shift to the high energy regions due to the increasing number of electrons in 3d and 4d bands. The shift of the Fermi level to the high energy, on going from Sr<sub>2</sub>MnNbO<sub>6</sub> to Sr<sub>2</sub>FeMoO<sub>6</sub> and to Sr<sub>2</sub>NiRuO<sub>6</sub> is estimated to be 1.88 eV and 1.03 eV, respectively.

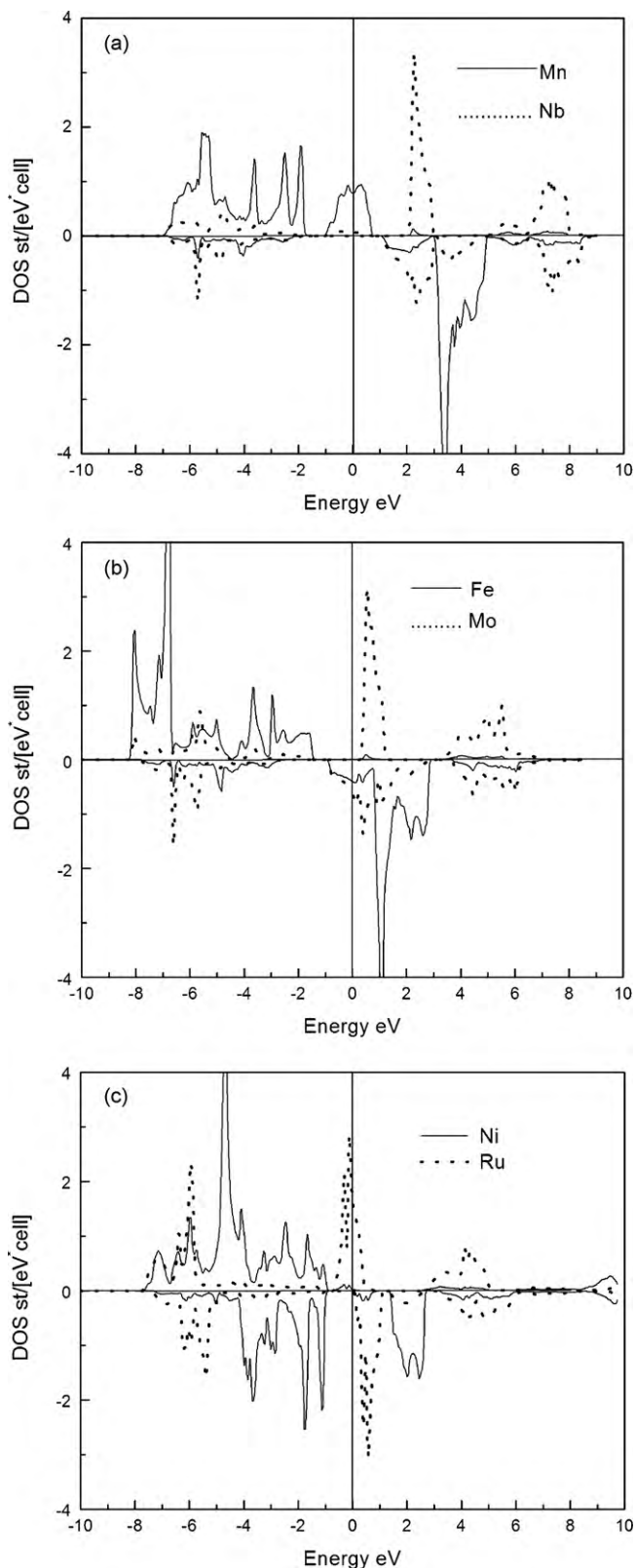
In all compounds, a hybridization between the 3d and 4d orbitals at Fermi level is seen between the Mn e<sub>g</sub>–Nb e<sub>g</sub> in the spin up channel, Fe t<sub>2g</sub>–Mo t<sub>2g</sub> in the spin down channel and Ni t<sub>2g</sub>–Ru t<sub>2g</sub> in the spin up channel. In Sr<sub>2</sub>MnNbO<sub>6</sub>, the 3d band is splitted due to the crystalline field. Upon the progress of the Fermi level to the high energy region, this band becomes below Fermi level with clear crystalline field splitting as shown in the DOS of Sr<sub>2</sub>FeMoO<sub>6</sub> and merges with the t<sub>2g</sub> bands of the spin up channel of Fe.

Interestingly, the spin down t<sub>2g</sub> band of 3d in all compounds is affected by the approach of the Fermi level to it. This band of Mn in Sr<sub>2</sub>MnNbO<sub>6</sub> spreads from 1.0 eV to 5.0 eV above Fermi level. In Sr<sub>2</sub>FeMoO<sub>6</sub> this band crosses Fermi level and it is seen to start splitting at  $\sim 1.7$  eV. In the DOS of the Sr<sub>2</sub>NiRuO<sub>6</sub> the 3d of Ni completely splits signifying the effect of the crystalline field effect on this band. The last observation is reported in Sr<sub>2</sub>CrReO<sub>6</sub> [14]. The width of 4d-t<sub>2g</sub> bands in the spin up channel of the Nb<sup>5+</sup>, Mo<sup>5+</sup> and

**Table 2**

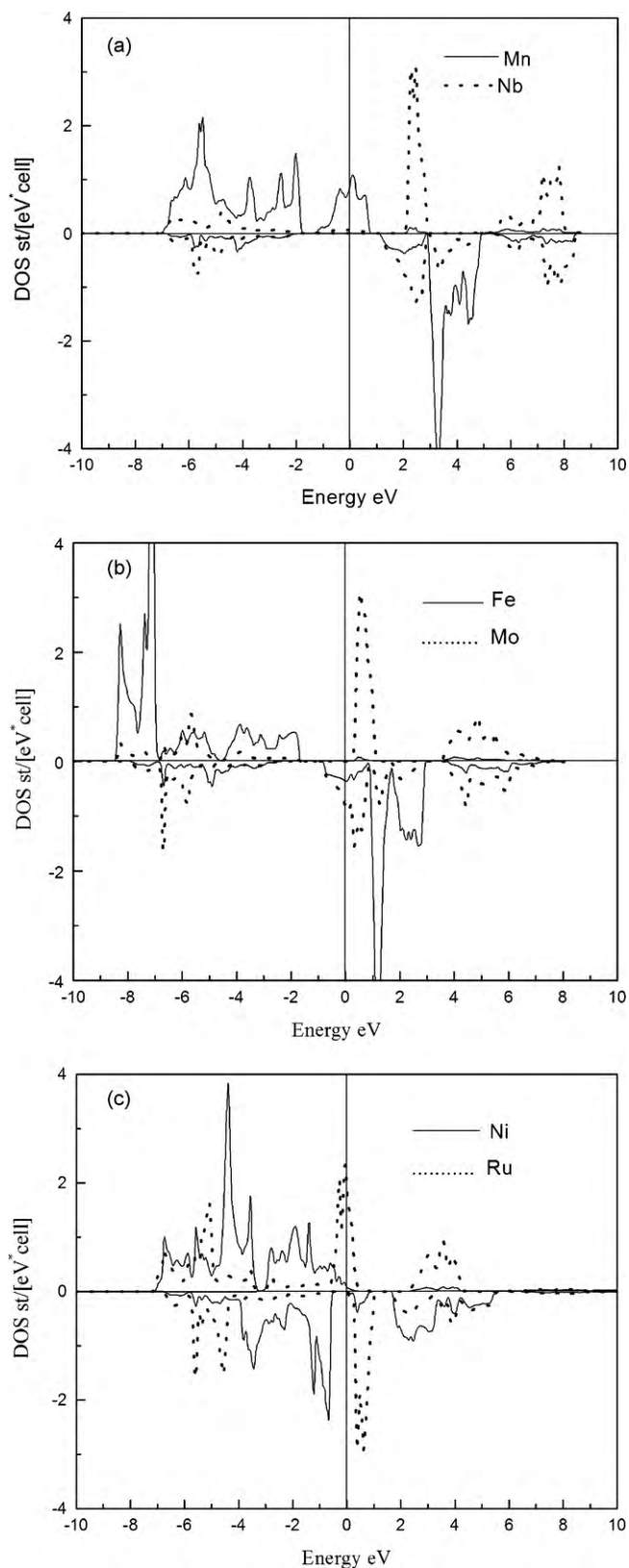
The muffin-tin spheres in atomic units (a.u) that were used in the present calculations.

Compound	Pseudo-cubic					Tetragonal				
	Sr	B	B'	O1	O2	Sr	B	B'	O1	O2
Sr <sub>2</sub> MnNbO <sub>6</sub>	2.5	1.94	1.91	1.69	1.69	2.36	1.94	1.91	1.69	1.69
Sr <sub>2</sub> FeMoO <sub>6</sub>	2.5	1.94	1.90	1.69	1.69	2.50	1.94	1.90	1.69	1.69
Sr <sub>2</sub> NiRuO <sub>6</sub>	2.5	2.00	1.81	1.61	1.61	2.37	1.93	1.90	1.68	1.68



**Fig. 1.** (a) The density of states of pseudo-cubic  $\text{Sr}_2\text{MnNbO}_6$ . (b) The density of states of pseudo-cubic  $\text{Sr}_2\text{FeMoO}_6$ . (c) The density of states of pseudo-cubic  $\text{Sr}_2\text{MnRuO}_6$ .

$\text{Ru}^{5+}$  in the vicinity of Fermi level varies from 0.82 eV to 0.87 eV, respectively. It crosses Fermi level in  $\text{Sr}_2\text{NiRuO}_6$  showing splitting not observed in the previous two compounds. These behaviors of the 3d and 4d bands which are seen to be affected by its closeness of the Fermi level resulting in observation of half-



**Fig. 2.** (a) The density of states of tetragonal  $\text{Sr}_2\text{MnNbO}_6$ . (b) The density of states of tetragonal  $\text{Sr}_2\text{FeMoO}_6$ . (c) The density of states of tetragonal  $\text{Sr}_2\text{NiRuO}_6$ .

metallic conduction in all compounds but through different bands and channels.

The calculated values of magnetic moments from the two schemes for the studied compounds are given in Table 3. It is clear

**Table 3**The values of the spin and orbital magnetic moments of the studied compounds in  $\mu_B$ .

	Pseudo-cubic				Tetragonal			
	Spin		Orbital		Spin		Orbital	
$\text{Sr}_2\text{MnNbO}_6$	Mn	Nb	Mn	Nb	Mn	Nb	Mn	Nb
LSDA + U + SOC	3.6833	0.044	-0.018	-0.002	3.6534	0.046	-0.0056	-0.0023
$\text{Sr}_2\text{FeMoO}_6$	Fe	Mo	Fe	Mo	Fe	Mo	Fe	Mo
LSDA + U + SOC	3.9498	-0.355	0.0395	0.044	3.9718	-0.36	0.0345	0.0398
$\text{Sr}_2\text{NiRuO}_6$	Ni	Ru	Ni	Ru	Ni	Ru	Ni	Ru
LSDA + U + SOC	1.7418	1.055	0.2009	-0.1055	1.7108	1.131	0.2201	-0.113

that the LSDA + U + SOC scheme gives spin moments of  $3.683 \mu_B$  and  $0.044 \mu_B$  for Mn and Nb respectively, and  $3.949 \mu_B$  and  $-0.355 \mu_B$  for the Fe and Mo respectively while for the Ni and Ru the values are  $1.742 \mu_B$  and  $1.055 \mu_B$ . The values of the magnetic moments of Fe and Mo are consistent with the previously reported calculated ones [3]. The values of the magnetic moment are less than the theoretically expected values, due to the pd-hybridization across Fermi level. The magnetic moment of Sr is too small due to the diffusive 5s valence configuration and the ionic behavior of alkaline earth elements, where the magnetic moment of O atom is small due to the nearly closed 2p shells [3].

The magnetic ordering of the studied compounds can be understood in terms of the 3d and 4d band filling. This will be by adopting the valence and the spin states  $\text{Mn}^{3+}(3d^4:t_{2g}^3 e_g^1)$   $S=2$ ,  $\text{Nb}^{5+}(4d^0:t_{2g}^0 e_g^0)$   $S=0$ ,  $\text{Fe}^{3+}(3d^5:t_{2g}^3 e_g^2)$   $S=5/2$ ,  $\text{Mo}^{5+}(4d^1:t_{2g}^1 e_g^0)$   $S=1/2$ ,  $\text{Ni}^{3+}(3d^7:t_{2g}^5 e_g^2)$   $S=3/2$  and  $\text{Ru}^{5+}(4d^3:t_{2g}^3 e_g^0)$   $S=3/2$ .

In the compound  $\text{Sr}_2\text{MnNbO}_6$ , the Mn and Nb have the electronic configuration  $\text{Mn}^{3+}(3d^4:t_{2g}^3 e_g^1)$  and  $\text{Nb}^{5+}(4d^0:t_{2g}^0 e_g^0)$ . The calculated magnetic moments in Table 3 show that the Mn ions order ferromagnetically with respect to the Nb. The  $e_g$  orbitals of the  $\text{Mn}^{3+}$  and  $\text{Nb}^{5+}$  cross Fermi level in the spin up channel and the  $\text{Mn}^{3+}$ -O-Nb $^{5+}$  interaction is seen at Fermi level results in ferromagnetic ordering. In the extensively studied compound  $\text{Sr}_2\text{FeMoO}_6$ , the magnetic moment of Fe is aligned antiparallel to the one of Mo and the  $t_{2g}$  orbitals of  $\text{Fe}^{3+}$  and  $\text{Mo}^{5+}$  crosses the Fermi level in the spin down channel results in  $\text{Fe}^{3+}$ -O-Mo $^{5+}$  a ferrimagnetic ordering. The magnetic moments of Ni and Ru in  $\text{Sr}_2\text{NiRuO}_6$  are aligned parallel where the  $t_{2g}$  orbitals of  $\text{Ni}^{3+}$  and  $\text{Ru}^{5+}$  cross the Fermi level in the spin up channel which results in a ferromagnetic ordering.

The orbital magnetic moments of Fe and Ni 3d orbital are of the same sign as the spin magnetic moment indicating that the 3d orbital are over half filled in accordance with Hund's rule. But for the Mn ion, as well as for the other 4d orbitals, the orientation of orbital magnetic moments are antiparallel to the spin magnetic moment signifying these orbitals to be less than half filled in consistency with the Hund's rules. The values of the orbital magnetic moments for the 4d orbital of Nb, Mo and Ru are  $-0.002 \mu_B$ ,  $0.044 \mu_B$  and  $-0.105 \mu_B$ , respectively. These values indicate that the effect of the orbital magnetic moment vary from quenched to weakly unquenched to unquenched as going from Nb to Ru. A similar trend is observed in the 3d orbitals, see Table 3. For the Sr (5s) and O (2p) orbital, the orbital moments are negligibly small.

### 3.2. The electronic structure in the tetragonal crystal structure

The DOS of the compounds under the study in their tetragonal crystal structure are given in Fig. 2. The differences between these DOS's and those of the pseudo-cubic structure are highlighted in terms of the behavior of the bands near Fermi level in the valence and the conduction bands. In both structures the conduction character of the bands and the values of the magnetic moments are not varying very much. The sign of the spin and the orbital magnetic moments of the 3d and the 4d elements are not changing. This indicated that the magnetic order remains the same in both structures.

The most remarkable differences between the DOS in the pseudo-cubic and the tetragonal lattices are noted in  $\text{Sr}_2\text{NiRuO}_6$ . A gap in DOS below Fermi level is observed and related to the crystal field splitting. Furthermore, the width and occupations of the conduction band, the band in the spin down channel and above Fermi level and the total energy are compared.

## 4. Discussion and conclusions

In  $\text{Sr}_2\text{MnNbO}_6$  and compared to the pseudo-cubic structure, the position of the Fermi level in the  $e_g$  band of Mn 3d and the band occupation are higher. Further, its band width is found to be 1.86 eV, larger by 0.11 eV than that of the pseudo-cubic. The width of the  $t_{2g}$  band above Fermi level in the spin down channel is 3.92 eV and also larger by 0.32 eV than that of the pseudo-cubic structure. A crystal field splitting gap is found to be about 0.52 eV which is less by 0.20 eV while the total energy in the tetragonal structure is found to be less than that of the pseudo-cubic structure by 0.20 eV. The values of the orbital magnetic moments for the Mn are listed in Table 3 and seen to be less than that of the pseudo-cubic structure while it is not changing for Nb 4d orbital. In  $\text{Sr}_2\text{FeMoO}_6$  the  $t_{2g}$  band crosses Fermi level as reported in earlier works [3,15,16]. The energy gap resulting from the crystal field splitting is found to be 0.87 eV with strong Hund's coupling in accordance with the calculations of Serrate et al. in  $\text{Sr}_2\text{FeMoO}_6$  [15]. Also here the total energy is less by 0.23 eV than that of the cubic structure. The SOC in both PC and T-structures is of the same order. Compared to  $\text{Sr}_2\text{MnNbO}_6$ , the spin-orbit coupling in the pseudo-cubic and tetragonal structures is becoming large but still of quenched character. The compound  $\text{Sr}_2\text{NiRuO}_6$  is seen to have different behaviors, as compared to  $\text{Sr}_2\text{MnNbO}_6$  and  $\text{Sr}_2\text{FeMoO}_6$ . It has no energy gap seen in the spin up channel while the width of the conduction band is extending over long energy scale amounts to 4.02 eV with strong Hund's coupling. The total energy in the pseudo-cubic structure is found to be less than that of the tetragonal structure by about 0.2 eV. Furthermore, the value of the orbital magnetic moment for Ni 3d and Ru 4d are found to be  $0.2201 \mu_B$  and  $0.105 \mu_B$ , respectively. These values are larger than those of the 3d and 4d orbitals of the two previously discussed compounds and are signature of unquenched spin-orbit coupling in this compound.

In  $\text{Sr}_2\text{MnNbO}_6$ , the 3d and 4d elements have the electronic configuration  $\text{Mn}^{3+}(3d^4:t_{2g}^3 e_g^1)$  and  $\text{Nb}^{5+}(4d^0:t_{2g}^0 e_g^0)$ . The calculated magnetic moment has also shown that Mn ions order antiferromagnetically with respect to Nb ions. In  $\text{Sr}_2\text{FeNbO}_6$  [18], the  $\text{Fe}^{3+}$  and  $\text{Nb}^{5+}$  have the electronic configuration  $\text{Fe}^{3+}(3d^5:t_{2g}^3 e_g^2)$  and  $\text{Nb}^{5+}(4d^0:t_{2g}^0 e_g^0)$  respectively similar to the electronic configuration of  $\text{Mn}^{2+}(3d^5:t_{2g}^3 e_g^2)$  and  $\text{Mo}^{6+}(4d^0:t_{2g}^0 e_g^0)$  in  $\text{Sr}_2\text{MnMoO}_6$  [1,17]. Both of these compounds are reported to be antiferromagnetic insulators [1,17,18]. Yang et al. have reported LSDA + U calculations on  $\text{Sr}_2\text{MnMoO}_6$  [16] in which the DOS of 4d of  $\text{Mo}^{6+}(t_{2g}^0 e_g^0)$  is similar to that of the 4d of  $\text{Nb}^{5+}$  of the  $\text{Sr}_2\text{MnNbO}_6$ . But in the present LSDA + U calculations on  $\text{Sr}_2\text{MnNbO}_6$  the  $e_g$  orbitals of the 3d of  $\text{Mn}^{3+}$  and 4d of  $\text{Nb}^{5+}$  cross Fermi level and the  $\text{Mn}^{3+}$ -O-Nb $^{5+}$  interaction is seen at Fermi level results in ferrimagnetic ordering.



At this point it is worth to mention that the net magnetization is becoming stronger while progressing in the present series. The last member of this series is  $\text{Sr}_2\text{NiRuO}_6$  which favors the ferromagnetic ordering where the magnetic moments of  $\text{Ni}^{3+}$  and  $\text{Ru}^{5+}$  aligned in the same direction of easy magnetization axis. This ferromagnetism is in accordance with Goodenough–Kanamori–Anderson (GKA) rules where  $e_g^2-0-e_g^0$  for the  $\text{Ni}^{3+}-\text{O}-\text{Ru}^{5+}$  superexchange interaction is designated to be strong. Stabilization of ferromagnetism in this compound is reported by Pardo and Pickett in their LSDA+U+SOC calculations [19].

In summary, the electronic structure and magnetic properties of pseudo-cubic and tetragonal structures of double perovskite oxides  $\text{Sr}_2\text{MnNbO}_6$ ,  $\text{Sr}_2\text{FeMoO}_6$  and  $\text{Sr}_2\text{NiRuO}_6$  have been investigated in terms of FP-LAPLW method. The calculations were performed within LSDA+U including the spin–orbit coupling as implemented in WIEN2k code. The results show that all the compounds have half-metallic behavior through different bands and channels. The values of the total energy indicate that  $\text{Sr}_2\text{MnNbO}_6$  and  $\text{Sr}_2\text{FeMoO}_6$  are stable in tetragonal structure while  $\text{Sr}_2\text{NiRuO}_6$  prefers the pseudo-cubic structure. The resultant values of the magnetic moments of the compounds  $\text{Sr}_2\text{MnNbO}_6$  and  $\text{Sr}_2\text{NiRuO}_6$  to be ferromagnetic through the  $\text{Mn}^{3+}-\text{O}-\text{Nb}^{5+}$  and  $\text{Ni}^{3+}-\text{O}-\text{Ru}^{5+}$  interactions, while  $\text{Sr}_2\text{FeMoO}_6$  is ferrimagnetic. The values of the orbital magnetic moments for the 4d orbital of Nb, Mo and Ru are  $-0.002 \mu_B$ ,  $0.044 \mu_B$  and  $0.105 \mu_B$ , respectively. These values indicate the progressive strength of the spin–orbit coupling effect of the orbital magnetic moment which varies from quenched to weakly unquenched to unquenched as going from Nb to Ru.

With the band filling a ferromagnetic half-metallic state has been observed in  $\text{Sr}_2\text{NiRuO}_6$  in accordance with the superexchange interaction  $e_g^2-0-e_g^0$  of Goodenough–Kanamori–Anderson (GKA).

### Acknowledgments

O.A. Yassin (Wien2k-1350 user) would like to thank Prof. K. Schwarz and Prof. P. Blaha, Inst. of Materials Chemistry, T U Vienna, Austria, for their kind support.

### References

- [1] I.V. Solovyev, J. Magn. Mater. 268 (2004) 194.
- [2] W. Prellier, et al., J. Phys. Condens. Matter 12 (2000) 965.
- [3] H. Jeng, et al., Phys. Rev. B 67 (2003) 094438.
- [4] Q.F. Li, et al., Phys. Lett. A 372 (2008) 2911.
- [5] H. Kato, et al., Phys. Rev. B 69 (2004) 184412.
- [6] J.B. Philipp, et al., Phys. Rev. B 68 (2003) 144431.
- [7] K. Terakura, Prog. Mater. Sci. 52 (2007) 388.
- [8] H. Kawanaka, et al., J. Phys. Soc. Jpn. 6 (1999) 2890.
- [9] J.B. Philipp, et al., Acta Physica Polonica A105 (2004) 7.
- [10] W. Song, et al., Chem. Phys. Lett. 482 (2009) 240.
- [11] G. Vaitheewaran, et al., J. Phys.: Conf. Ser. 29 (2006) 50.
- [12] M.W. Lufaso, P.M. Woodward, Acta Cryst. B 57 (2001) 725.
- [13] K. Schwarz, P. Blaha, Comput. Mater. Sci. 28 (2003) 259.
- [14] C.Q. Tang, et al., Solid State Commun. 133 (2005) 219.
- [15] D. Serrate, et al., J. Phys. Condens. Matter 19 (2007) 023201.
- [16] Z. Yang, L. Ye, J. Phys. Condens. Matter 17 (2005) 2035.
- [17] H. Wu, Phys. Rev. B 64 (2001) 125126.
- [18] X. Zhao, et al., Mater. Sci. Eng. B 111 (2004) 101, and references therein.
- [19] V. Pardo, W.E. Pickett, Phys. Rev. B 80 (2009) 054415.