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Electronic structure study of double perovskite: A first principle approach

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ABSTRACT

The full potential Linearized augmented plane wave (FLAPW) method is employed to study the ground state and crystal properties of In_2MgTiO_6. The electronic-energy band structure, site and angular-momentum decomposed density of states and charge-density contours of double perovskite In_2MgTiO_6 are calculated by the FLAPW method with the generalized-gradient approximation using density-functional theory. From the analysis of density of states, we conclude that there is hybridization of Ti-d state with the O-p state, which implies that the interaction between the atoms of these two is highly covalent which is consistent with the calculation of electronic band structure as well as charge density studies. The equilibrium values, bulk modulus, and its pressure derivative have been estimated through optimization of the crystal structure of this material.

KEYWORDS

Electronic band structure;
Density of states;
FLAPW.

INTRODUCTION

Double perovskites with general formula $A_2B'B''O_6$ with A being an alkaline earth such as Ca, Ba, Sr or a lanthanide and B′, B″ are the transition metals are a matter of investigation since past decades[1-7]. Borges *et al.*[1] investigated the ferromagnetic metallic behaviour of Ca_2FeMoO_6, Sr_2FeMoO_6 and $Ba_2FeMoO_6$with Curie temperature of 345 – 426 K. Kang *et al.*[2] have studied the electronic structure of Ba_2FeMoO_6 using photoemission spectroscopy. Sarma[3] has pointed out the discrepancies between the manganites and the double perovskites and proposed the mechanism of occurrence of large T_C in double perovskites Sr_2FeMoO_6. This mechanism has been extended to many other systems by Kanamori and Terakura[4,5]. Moritomo *et al.*[6,7] shows a strong correlation between the curie temperature and the conductivity at room temperature which implies that the mobile conduction electrons mediate the exchange interaction between the local Fe^{3+} ions. In this model the hybridization of the Mo 4d (t_{2g}) and Fe 3d (t_{2g}) states plays the key role in stabilizing ferromagnetism at high curie temperatures[3-5,8] Based on band calculation using the Local Density approximation (LDA), Pickett[9] has proposed that the double perovskite La_2MnVO_6 can be a promising candidate for exhibiting half-metallic antiferromagnets (HM-AFM) characteristics. The electronic structure calculations were based on the assumption that both Mn and V ions are trivalent and more importantly a low spin state ($t_{2g}{}^4e_g{}^0$; S =1) resulted for all Mn^{3+} ions. Phillip *et al* studied the effect of structural changes and doping in

the double perovskite A_2CrWO_6 (A = Sr, Ba, Ca) on the magnetotransport, the magnetic and optical properties together with band structure calculations. Phillip *et al*. have also pointed out that the T_C in double perovskites was discussed to depend sensitively on band structure and band filling in contrast to experimental results[10,11].

In double perovskites having structural formula $A_2B'B''O_6$, each transition metal site (B′B″) is surrounded by an oxygen octahedron and the A atoms site are situated in the holes produced by eight adjacent oxygen octahedral. The properties of double perovskite compounds are determined by the relative sizes, valencies and ordering of the A and B site ions.

The study of elemental structures plays a pivotal role in solid-state physics[12]. The structural study gives insight into the properties of the elements. In the present article, the electronic structure (density of states, band structure) of In_2MgTiO_6 (IMT) are calculated by full potential linearized augmented plane wave (FLAPW) method[13] within the generalized gradient approximation (GGA)[14].

COMPUTATIONAL DETAILS

The ground-state properties of IMT are investigated employing the FLAPW method within the generalized gradient approximation (GGA). The structural parameters of the system were optimized at 3000-k points inside the irreducible Brillouin zone for integration. The full geometrical optimization gives an in-plane lattice constant a = b = c = 8.0 and c/a = 1.0 of pure In_2MgTiO_6. Accurate optimizations of crystal geometries have come to serve as a stringent test for electronic structure calculations. We use our optimized parameters for the study of electronic structure calculations taking muffin tin radius (RMT) as 2.0, 1.9, 1.8 and 1.6 for In, Mg, Ti and O respectively. The total energy has been minimized and calculated with respect to the volume. In our calculation, the crystal structure of IMT has the space group symmetry Fm-3m. The Birch-Murnaghan relation for equation of state (EOS) is used to get the static equilibrium volume V_0 (=940.39) as well as the bulk modulus B_0 (= 123.55 GPa) and its pressure derivative B′ (= 3.0873) at zero pressure.

RESULTS AND DISCUSSION

In Figure 1, we have calculated and minimized the total energy with respect to the volume. In our calculation, the crystal structure of IMT has the space group symmetry Fm-3m. The Birch-Murnaghan relation for equation of state (EOS) is used to get the static equilibrium volume V_0 (=940.39) as well as the bulk modulus B_0 (= 123.55 GPa) and its pressure derivative B′ (= 3.0873) at zero pressure. The pressure derivative of

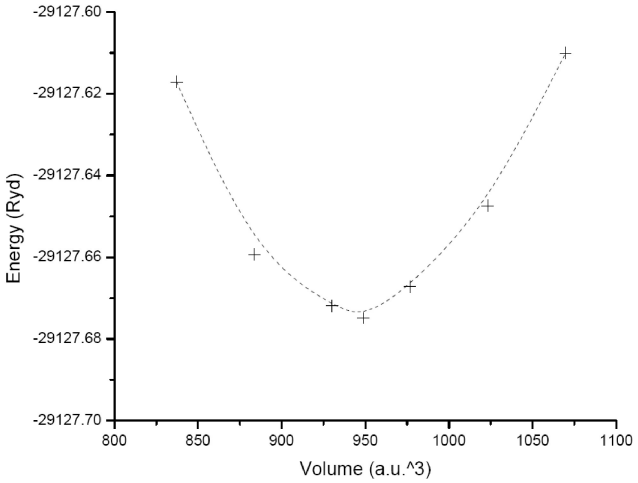

Figure 1 : Total energy of In_2MgTiO_6 as a function of cell volume

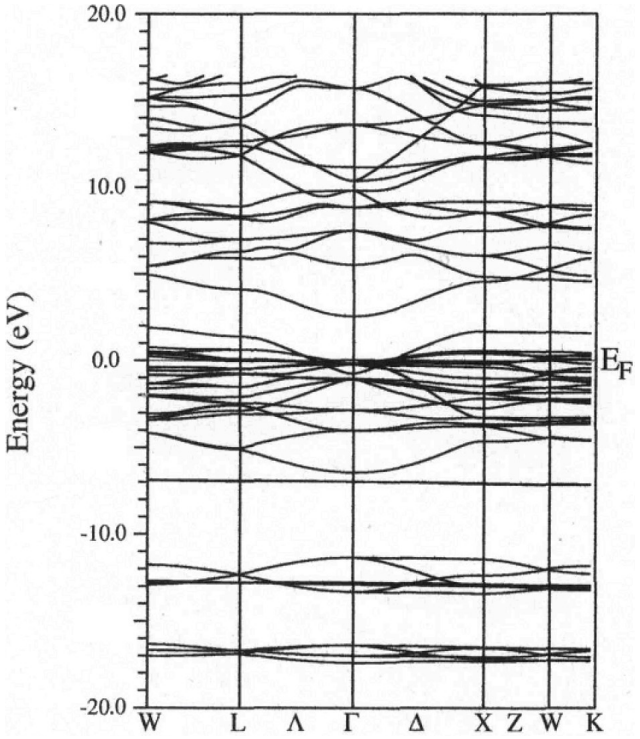

Figure 2 : The electronic band structure of In_2MgTiO_6 along high symmetry directions

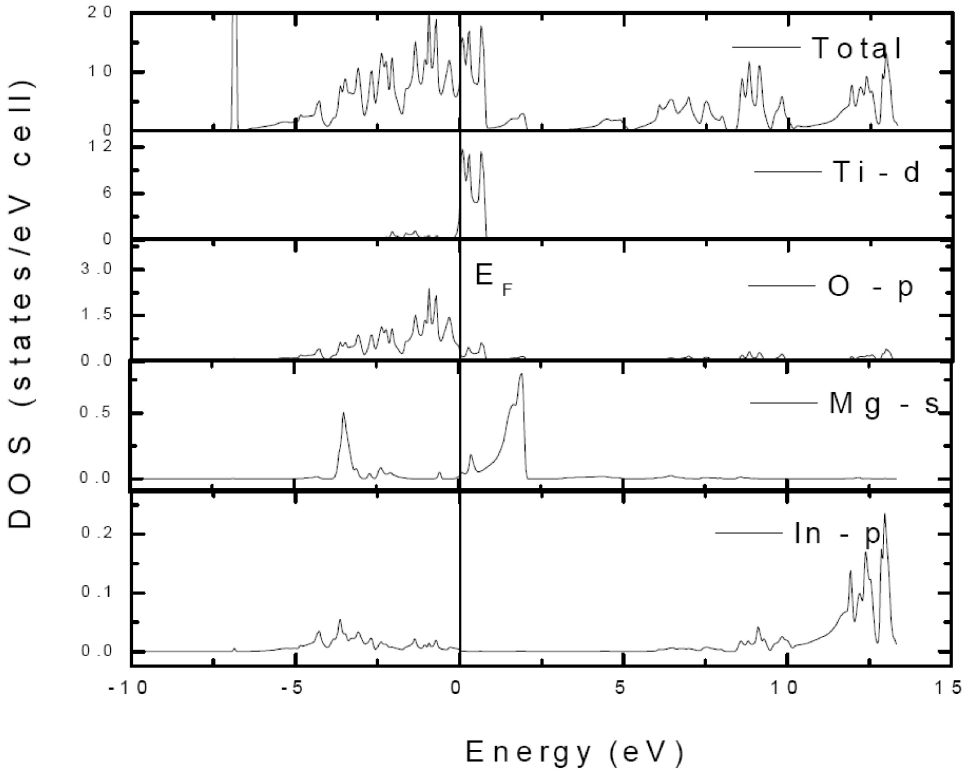

Figure 3 : Total DOS and PDOS around Fermi energy of In_2MgTiO_6 calculated for the optimized lattice constant

bulk modulus at zero pressure B_0' is a parameter of great physical significance in high pressure physics. It is related to a few other important thermo physical properties (like phase transitions, interphase energy, adsorption energy etc.)[14].

The calculated band structure for IMT in the high symmetry direction in the Brillouin zone is shown in Figure 2. In this figure, we find a large dispersion of the bands. It can be seen that there are many bands from – 3 eV to 1 eV. These bands are mainly contributed from Ti-d and O-p electrons. These bands also overlap each other at and around the Fermi level which indicates that there is hybridization between the Ti-d and O-p states. Comparing it with Figure 3, it supports our explanation.

The angular momentum projected densities of states were obtained by using 3000-k points inside the irreducible Brillouin zone for integration. The full geometrical optimization gives an in-plane lattice constant a = b = c = 8.0 and c/a = 1.0 of pure In_2MgTiO_6.

Figure 3 shows the total DOS along with partial DOS of Ti-d and O-p states of IZT. It is observed that the main contribution in the valence band comes from Ti-3d state. The Ti-d state hybridizes with O-p states near Fermi level. The hybridization between Ti-d and O-p states implies that the interaction between the atoms of these two is highly covalent.

CONCLUSION

We have performed and analyzed the FLAPW calculation of the double perovskite In_2MgTiO_6 (IMT). The ground state and electronic structural properties have been obtained. From the angular momentum projected densities of states at 3000-k points inside the irreducible Brillouin zone for integration, it can be concluded that Ti-d state hybridizes with O-p states near Fermi level and the interaction between the atoms of these two is highly covalent.

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