



## Full Potential Study of Electronic and Optical properties of Laves phase HoFe<sub>2</sub> Compound

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**Abstract:** In this work we presented optical properties and electronic structure of intermetallic compound. Our estimations are based on density functional theory (DFT) utilizing the Full Potential Linearized Augmented Plane Wave (FPLAPW) method with the inclusion of spin orbit coupling. Local spin density approximation with correlation energy (LSDA+U) was used for exchange-correlation potential. Band structure was estimated through calculation which shows the metallic character of HoFe<sub>2</sub> compound. We found that majority spin *d* states of Ho and Fe mainly contribute to the valence band while Minority spin states Ho-4*f*, 5*d* dominate in the conduction band from E<sub>F</sub> to 4 eV. We again found decent agreement accompanying experiment for the diagonal optical conductivity using LSDA+U approximations. The results are examined considering band to band transitions.

**Key Words:** Band Structure Calculations, FPLAPW Method, LSDA+U Approximation, Density of States, Optical Properties, HoFe<sub>2</sub> Compound

### Introduction

Study of Rare Earths Transition Metal (RE-TM) intermetallic compounds drawn much attention due to their outstanding magnetic, optical, electrical characteristics and stimulate many and varied uses of rare-earths in both civil and martial industries. Rare-earth magnetic materials are now in demand for applications including rare-earth permanent magnets, magnetocaloric and magnetostrictive materials (Clark, 1980). Among them RFe<sub>2</sub> compounds drawn the interest due to their two basis of magnetism – one is the Fe -3*d* and R – 5*d* electrons, and second is the localized 4*f* electrons illustrated by the Russell – Saunders scheme (Brooks et al 1991). Numerous studies on the electronic, optical and magneto-optical properties of RFe<sub>2</sub> compounds have been reported experimentally (Katayama and Hasegawa 1981, Mukimov et al 1985, Kravets et al 1988, Shripov et al 1990,

Lee et al 2000) and, to a lesser extent, theoretically (Brooks et al 1989, Coehoorn 1989) during last decade.

Brooks et al. (1989) considered the 4*f* electrons as part of a self-consistent outer core and calculated the electronic structure of RFe<sub>2</sub> (R= Gd-Lu) compounds. They conclude that total moment of electron conduction independent of R 4*f* spin in RFe<sub>2</sub> compounds. Rhee (1998) accomplished calculation of digital, optical and MO properties of RFe<sub>2</sub> (R= Y and Lu) using the tight-binding linear muffin-tin orbital (TB-LMTO) technique with in Local Spin Density Approximation (LSDA) and reproduced the magnetic moment of YFe<sub>2</sub> and LuFe<sub>2</sub> successfully. Rhee concluded that the self energy corrections to the calculated optical conductivity spectra led to a reasonable agreement between experiment and the theoretical calculations for both diagonal and



off-diagonal components of optical conductivity tensor. Lee et al. (2000) also presented a calculation of optical and MO properties of  $RFe_2$  compounds using the TB-LMTO method, treating  $4f$  electrons of rare earth as valence electrons and concluded that agreement between theory and experiment for optical conductivity tensors was poor except for  $LuFe_2$ , in which the  $4f$  shell is completely closed. Tanaka et al. (1992) performed the spin polarised of  $Gd_{33}Fe_{67}$  alloy using linear muffin-tin orbital (LMTO) method and found the resultant magnetic moment of Fe is in good agreement with the experiment. The Optical properties and electronic structure of  $GdFe_2$  compound has been calculated by Rhee (2003). FPLAPW method within the LSDA+U approximation has been adopted for the calculations and found that LSDA+U improve the structural and magnetic properties as compare with the experiment. However, application of the LDA+U method has rather insignificant effects on the optical and magneto-optical properties of  $GdFe_2$  in the 1.5 to 5.5-eV range. Electronic structure and magnetic properties of  $LuFe_2$  with C14, C15 and C36 structures have been calculated by Zhang et al. (2007) using Full potential linearized augmented plane wave (FPLAPW) method. They concluded with the remark that there exist both localized and itinerant  $d$  electrons, resulting in antiferromagnetic ordering in the three structures. Previously we (Saini et al 2007) have also theoretically investigated the optical and MO properties of  $GdFe_2$  and  $GdCo_2$  compounds and found that LSDA+U method improves the ground state properties as compared to LSDA. We have also found that in the energy range 0-3 eV, Kerr rotation and off- diagonal components of conductivity, LSDA method gives better results than the LSDA+U method. To the best of our knowledge there are no calculations performed on electronic and optical study of  $HoFe_2$  compound even though the experimental

data of optical conductivity is exist.

In present communication we have presented a study of the electronic and optical properties of intermetallic  $HoFe_2$  compound with Ho ( $4f^7$ ). Our calculations thus demonstrate the effect on structural, electronic and optical properties of occupancy of  $f$  electron states since the Ho have partially filled  $f$  -states. The paper is organized as follow: In the Section 2 computational details are explained, third section devoted to discuss the results of the work, and finally the conclusions are presented in the fourth section.

### Computational details

Adopting the full potential linear muffin-tin orbital (FP-LMTO) method based on the Density Functional Theory (DFT) (Savrasov 1996, Savrasov and Savrasov 1992) calculations was done by us as applied in the recent version of WIEN2K code (Blaha et al 2001). Space is divided into non-overlapping muffin-tin (MT) spheres in(FP-LMTO) method centered at the atomic sites separated by an interstitial region (IR). Fourier series represents the basis functions in the IR region. The basis functions are expanded jointly with spherical harmonics functions within the MT spheres. Treated by the LSDA+U approximations, the exchange-correlation (XC) potential introduced by Anisimov et al. (1991, 1993) which explicitly comprise the onsite Coulomb interaction term in the conventional Hamiltonian. It is widely known that we have to include Coulomb interaction term in the Hamiltonian to explain the systems those contain transition metal (3d local electrons) or rare earth elements RE (localized  $4f$  electrons). These systems are known as strongly correlated systems. We used the intra-atomic correction energy (U) of 6.7eV for Gd given by Harmon et al. (1995). We used the parameters  $R_{MT}K_{max} = 7.0$  and  $G_{max} = 14$ , to perform the self consistent spin-polarized calculations. The cut-off energy is taken as

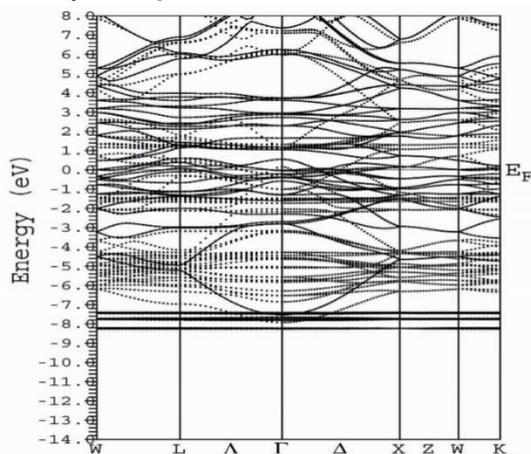


7.84Ry to separate the valence and core states. Our self-consistent calculations were finished with charge convergence of  $10^{-4}e$  and energy convergence of  $10^{-4}Ry$ . We used 8000 k points in the Brillouin zone for calculation of the optical properties as calculations of the optical properties require a dense k-space mesh.

## Results and Discussion

### Band Structure

The LSDA method is incapable to describe systems like electronic properties (Saini et al 2007) or strongly correlated system where the *f*-electrons are well localized, and where the spin orbital interactions cannot be neglected. The Hubbard term (*U*) which describes the *d-d* or *f-f* interaction is added to the LSDA in the LSDA+*U* approach. To explain the correct ground state properties of strongly correlated systems, this method has proven its effectiveness. From the LSDA+*U* calculations, the energy band structure of HoFe<sub>2</sub> compound along the higher symmetry direction in the Brillouin zone is shown in Fig.1. It is noted that the difference in the spin up and spin down bands is mainly due to the position of *f* states in HoFe<sub>2</sub> compounds which have partially filled *f*-orbitals.



**Fig.1 Spin-resolved band Structure of HoFe<sub>2</sub> for spin-up(solid line) and spin-down (broken line) states using LSDA+*U*.**

The occupied spin up and spin down Ho-*f* states lie at around 8.0eV and around 6.0 eV below Fermi energy(*E<sub>F</sub>*), respectively, while the unoccupied spin down Ho-*f* states lie around 3.0 eV above *E<sub>F</sub>*. We obtained more prominent splitting of Ho-*f* states as compared to Ho-*d* states.

### Density of States

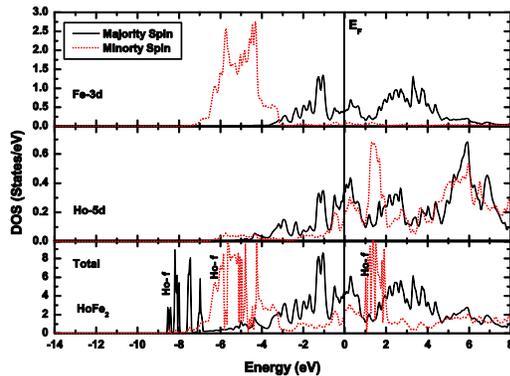
Total and partial DOS results of HoFe<sub>2</sub> using LSDA+*U* calculations are shown in Fig.2 which shows the contributions of different atoms and orbital. Ho-*f* states are clearly identified by the sharp peaks in figure. It can be notice that majority spin electrons form the top of the valence band up to the -3 eV below *E<sub>F</sub>*. These states are dominating by Ho-5*d* and Fe-3*d* states. These states cross the *E<sub>F</sub>* showing the metallic character of the HoFe<sub>2</sub> compound. Ho-*f* states lie well below around -8 eV from *E<sub>F</sub>* and retain their atomic character. We can also found the splitting of Ho-*f* states which is due to the inclusion of spin orbit interaction in the calculations. On the other hand minority spin dominate well above the *E<sub>F</sub>* and form the conduction band. Minority spin states Ho-4*f*, 5*d* mainly contribute to the conduction band from *E<sub>F</sub>* to 4 eV. We notice that DOS are almost similar to those for majority and minority spin expect Ho-*f* states. These states are lie around -8 eV below *E<sub>F</sub>* in majority spin while lie at around 2 eV above *E<sub>F</sub>* in minority spin. We found that the spin-down Ho-*p*, -*d* and Fe- *p* states hybridize with the spin-down Ho-*f* bands on the other hand the spin-up Ho-*f* bands remains unhybridized. From DOS one can also find that *d* states of Ho and Fe dominate in the energy range -3 to 3 eV and significantly contribute to DOS at *E<sub>F</sub>* i.e. *N(E<sub>F</sub>)*.

### Optical conductivity

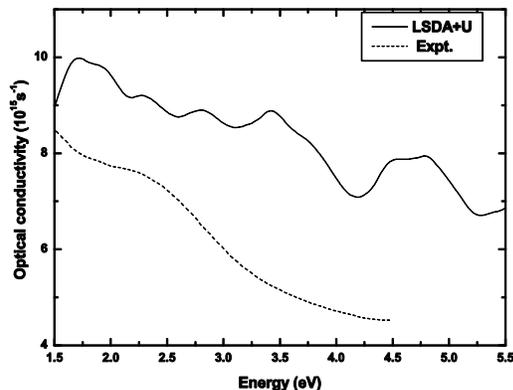
The diagonal component of interband optical



conductivity tensor calculated by the use of LSDA+U calculations along with experimental data are shown in Fig.3. Various values of broadening were attempted to reproduce experimental broadening.



**Fig. 2: The Total and partial d-DOS of HoFe<sub>2</sub> using LSDA+U method.**



**Fig. 3: The diagonal ( $\sigma_{1xx}$ ) optical conductivity of HoFe<sub>2</sub> using LSDA+U, along with experimental data**

Large values of broadening shows the calculated results much close to the experiment but the structure was lost. Small values give a peaky spectrum. We have used a moderate broadening of 0.5 eV. We found isotropy (i.e.  $\sigma_{1xx} = \sigma_{1zz}$ ) nature of HoFe<sub>2</sub> compound. The experimental spectrum is quite smooth and a broad structure is at around 2.5eV while calculations show broad peaks around 1.75 and 3.4eV using LSDA+U calculations. The magnitude of calculated optical conductivity is larger than that obtained

from experiment (Lee et al 2000). Other calculations also show similar structure this could be due to high reactivity of the surface of rare earth compounds. The structures around 1.75 and 3.4 eV could be due to the transitions from occupied Fe *p*, *d* states to unoccupied Ho *p*, *d* states.

## Conclusions

Electronic structure and optical properties of HoFe<sub>2</sub> compound has been calculated. We find majority Ho-*f* states lie well below from  $E_F$  using LSDA+U calculation and retain their atomic character. On the other hand minority Ho-*f* states lie around 1.5 eV above  $E_F$ . Splitting of Ho-*f* states is also obtained with the inclusion of spin orbit interaction. The calculated optical conductivity spectra is in reasonable agreement with the experimental results in 1.5 -5.5 eV range. Transitions from occupied Fe *p*, *d* states to unoccupied Ho *p*, *d* states contribute to the main structures in the optical conductivity. A review is also available in literature (Zegaou et al 2014, Bhatt et al 2018) and references therein.

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