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# Optically Isotropy In Scintillator Host Compounds $M_2LaCl_5$ (M=Rb, and Cs): Ab-initio Study

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**Abstract.** Full potential linearized augmented plane wave method (FP-LAPW) has been used to calculate the electronic structure and optical properties of high light output scintillator host compounds $M_2LaCl_5$ (M=Rb, and Cs) with in the Tran Blaha modified Becke Johnson (TB-mBJ) potential. These are wide band gap materials with the band gap of 4.75, and 4.72 eV for $Rb_2LaCl_5$, and $Cs_2LaCl_5$ respectively. From the calculated optical properties of these compounds, we find these compounds to be optically isotropic, though they are structurally anisotropic, which is an important criteria for the ceramic scintillators.

**Keywords:** Density Functional Theory, Scintillation, Optical constants.
**PACS:** 71.15.Mb, 78.70.Ps, 78.20.Ci.

## INTRODUCTION

Lanthanum halide scintillators are widely used for the x-ray and gamma ray detectors. These are good host materials having scintillating property with high energy resolution, fast scintillation decay, high yield and are less hygroscopic and much transparent. $K_2CeCl_5$, $K_2CeBr_5$, $Cs_2CeCl_5$ are one among these type of scintillating compounds with high light output (50,000 photons/Mev) [1,2]. $K_2LaCl_5$: Ce is a fast scintillator with decay time of 40 ns [3]. van Loef et al. [4] studied the scintillating property of $K_2LaX_5$:Ce (X=Cl, Br, and I) and observed $K_2LaCl_5$: 10% Ce to be the best scintillator with high light yield, fast decay time and high energy resolution and also the light yield increases from Cl to I. The spectroscopic and scintillating properties of pure and doped $K_2LaCl_5$ are studied and compared with that of high light yield scintillator [5]. There are only very few theoretical studies available on these compounds. Singh [6] studied the structural and optical properties of $K_2LaX_5$ (X=Cl, Br, and I) and reported the optical isotropy of these compounds despite being structurally anisotropic [6]. In the present paper, we are interested in studying the electronic structure and optical properties of these lanthanum halide based scintillator host compounds $M_2LaCl_5$ (M=Rb, and Cs).

## METHOD OF CALCULATIONS

The calculations were carried out using the Full potential linearized augmented plane wave method implemented in the WIEN2k code. As it is well known that Local Density Approximation (LDA), Generalized gradient approximation (GGA) generally underestimate the band gaps, we have used TB-mBJ functional for the electronic structure and optical properties of these compounds which give the band gap values comparable to the experimental values. Throughout our calculations, we have used the experimental lattice parameters [7] with optimized positions. The atomic positions of $K_2LaCl_5$ is used for optimizing the position of the remaining compounds due to the lack of experimental data. The position relaxations were carried out by using the Generalized gradient approximation of Perdew, Burke and Ernzerhof. The basis functions were expanded upto $R_{mt}xK_{max}=8$, where $R_{mt}$ is the smallest of all muffin tin sphere radii and $K_{max}$ is the plane wave cutoff. We have used 9x13x15 k-mesh with 280 k-points in the Irreducible Brillouine Zone (IBZ). Since the optical properties calculations need higher k-points, we used the k-mesh of 12x18x20 with 770 k-points in IBZ.

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## RESULTS AND DISCUSSIONS

### Electronic structure:

The electronic structure calculations are carried out using the TB-mBJ functional. These compounds crystallize in orthorhombic crystal structure with space group Pnma (62). We have used the experimental lattice parameters a=13.146, b=9.037, c=8.209 Å for $Rb_2LaCl_5$, and a=13.742, b=9.227, c=8.526 Å for $Cs_2LaCl_5$ compounds [7]. These are wide band gap insulators because of the large electronegativity difference between the cation, and halogen. The calculated band gap values are given in TABLE 1 with the GGA, and TB-mBJ functionals. We observed that the TB-mBJ functional is found to improve the band gap when compared to functional GGA, and the band gap values are found to decrease from $Rb_2LaCl_5$ to $Cs_2LaCl_5$. Singh [6] performed the electronic structure calculations of $K_2LaCl_5$ and reported two band gap values with and without the inclusion of additional unoccupied f derived bands in the calculation and the corresponding values are 4.59(4.46), 6.32(5.32) eV using the TB-mBJ (GGA) functional. We have also calculated these band gaps, our calculated band gap values with and without inclusion of f derived states are 4.75(4.55), 6.35(5.1) eV for $Rb_2LaCl_5$, 4.72(4.54), 6.31(5.1) eV for $Cs_2LaCl_5$ with TB-mBJ(GGA) functional, and they are in comparison with above values.

In order to further understand the specific atomic contribution we have calculated the density of states, and the corresponding figures are shown in FIGURE 1. The valence band is mainly of Cl-p character in all the compounds lying around the energy range 0 to -1.98 eV for $Rb_2LaCl_5$, 0 to -1.85 eV for $Cs_2LaCl_5$ respectively. The lower part of the conduction band around 4.7 to 5 eV, and 4.6 to 5.0 eV is mainly of La-f states, and the states around 6.3 to 8 eV, and 6.2 to 7.9 eV is due to La-d states, and the higher energy levels are due to M-d (M=Rb, and Cs), Cl-d state for $Rb_2LaCl_5$, and $Cs_2LaCl_5$ respectively.

**TABLE 1.** The calculated band gap values of $M_2LaCl_5$ (M=Rb, and Cs) compounds.

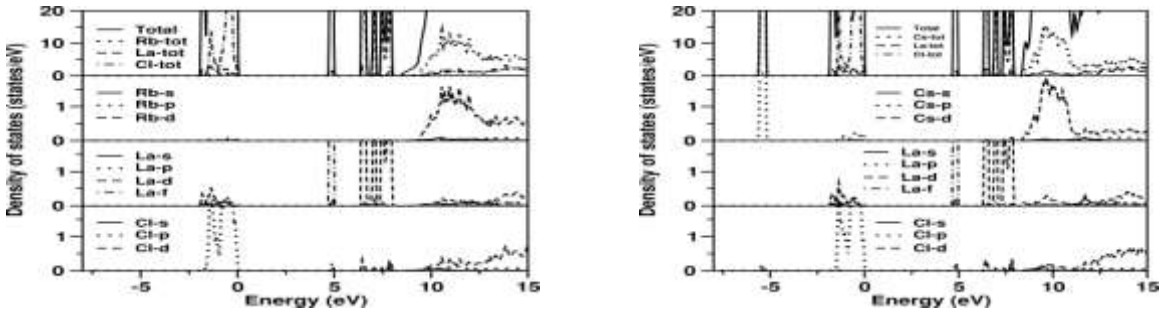
| Compound | Band gap (in eV) | |
|---|---|---|
| | GGA | TB-mBJ |
| $Rb_2LaCl_5$ | 4.55 | 4.75 |
| $Cs_2LaCl_5$ | 4.54 | 4.72 |

### Optical Properties

Optical properties are important in order to understand the scintillation mechanism. We have increased the k-points for analyzing the optical properties in order to ensure the convergence. We have also calculated the dielectric function of these compounds and the corresponding spectra are shown in FIGURE 2. The lower energy part of dielectric plot is due to the transition of electrons from the Cl-p states to La-f, d states and higher energy part is due to transition of electrons from Cl-p states to M-d (M=Rb, and Cs), Cl-d states. The calculated refractive index plots are shown in FIGURE 3 for all the compounds. Interesting point to note is the presence of isotropy in the refractive index, though the compounds are structurally anisotropic. The isotropy is found to decrease from $Rb_2LaCl_5$ to $Cs_2LaCl_5$. The static refractive index values of these compounds are given in TABLE 2, where we can observe the anisotropy of these compounds to be less than 1%. The optical isotropy of these compounds observed particularly in the low energy region, make them important to be used as ceramic scintillators.

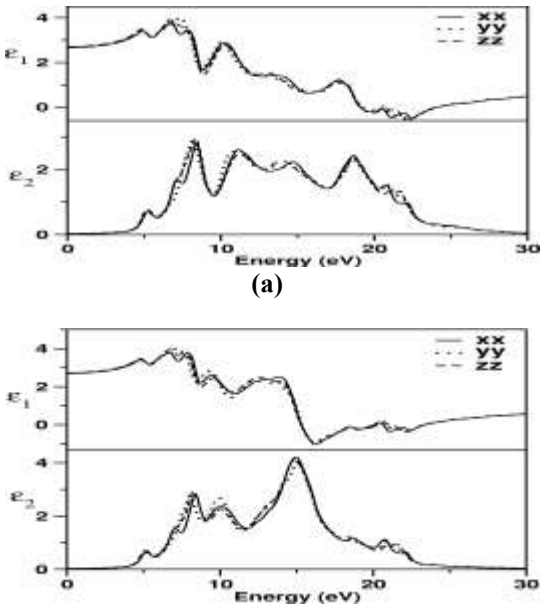
## CONCLUSIONS

We have studied the electronic structure and optical properties of $M_2LaCl_5$ (M=Rb, and Cs) scintillator host compounds which are having high light yield and fast decay time. From the calculated optical properties of these compounds we could find optical isotropy of these compounds though they are structurally anisotropic, which is an important criteria for the ceramic scintillators.



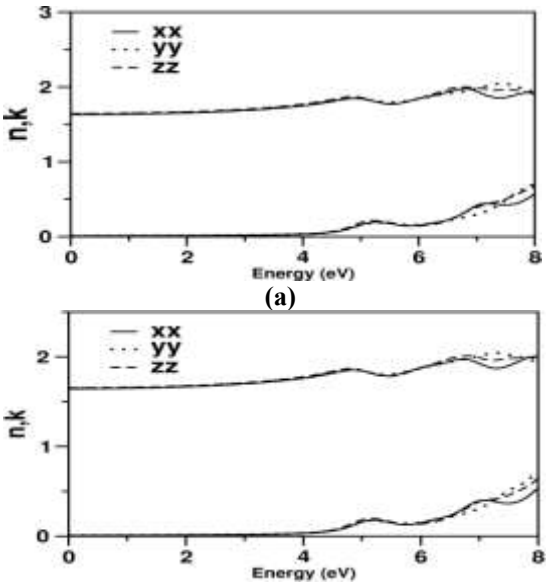

**(a)** **(b)**

**FIGURE 1.** The density of states plots of a) $Rb_2LaCl_5$ b) $Cs_2LaCl_5$ compounds using the TB-mBJ functional.




**(a)** **(b)**

**FIGURE 2.** The real and imaginary part of dielectric function of a) $Rb_2LaCl_5$ b) $Cs_2LaCl_5$ compounds using the TB-mBJ functional.




**(a)** **(b)**

**FIGURE 3.** Refractive index of the a) $Rb_2LaCl_5$ b) $Cs_2LaCl_5$ compounds using the TB-mBJ functional.

**TABLE 2.** The calculated static refractive index (n(0)) values of $M_2LaCl_5$ (M=Rb, and Cs) compounds.

| Compound | n(0) | | |
|---|---|---|---|
| | xx | yy | zz |
| $Rb_2LaCl_5$ | 1.633 | 1.639 | 1.644 |
| $Cs_2LaCl_5$ | 1.644 | 1.649 | 1.655 |

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## REFERENCES

1. U. N. Roy, M. Groza et al., *Nucl. Inst. Meth. Phyc. Res. A* **579**, 46-49 (2007).
2. R. Hawrami, A. K. Batra et al., *J. of Cry.Grow* **310,** 2099-2102 (2008).
3. J. C. Vant Spijker, P. Dorenbos et al., *Radiation measurments* **24**, 379-381 (1995).
4. E. V. D. van Loef, P. Dorenbos et al., *Nucl. Inst. Meth. Phys. Res.* A **537**, 232-236 (2005).
5. E. V. D. van Loef, P. Dorenbos et al., *Phys. Re.* B, **68**, 045108 (2003).
6. David J. Singh, *Phys. Rev. B* **82**, 155145 (2010).
7. H. J. Seifert, H. Fink, and G. Thiel *J. of. Less common metals*, **110**, 139-147 (1985).