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Optical Properties Of Halide And Oxide Compounds Including The Excitonic Effects

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Abstract. We have studied the optical properties of alkali halide and alkaline-earth oxide compounds including the excitonic effects by using the newly developed bootstrap kernel approximation for the exchange-correlation kernel of the Time-Dependent Density Functional Theory (TD-DFT) implemented in Full-Potential Linearized Augmented Plane Wave (FP-LAPW) method in the elk code. The bootstrap calculations are computationally less expensive and give results the same quality as the Bethe-Salpeter equation. We found improved results when compared to normal Density Functional Theory calculations, and observed results are comparable with the experiments. The lower energy peak of imaginary part of dielectric spectra shifts to lower energy regions as we move from MgO to BaO indicating the decrease in the band gap of these compounds from MgO to BaO. In all the studied compounds, the lower energy peak of the imaginary part of dielectric function is due to the transition from halogen p or oxide p states to metal derived s/d states.

Keywords: Optical Properties, Excitonic effect.

PACS: 78.66.Hf, 71.35.-y

INTRODUCTION

Density Functional Theory (DFT) is a powerful tool for the calculation of the ground state properties, but it does not include the excitonic effect which is very important in the calculation of the optical properties. The calculation of excitonic effects in solids is a major challenge in the solid state theory. The calculations of the optical properties including the excitonic effect involves three steps, first the electronic ground state is treated within DFT, second single particle spectrum of electrons-holes is obtained within the GW approximation, third electron-hole interaction is calculated and Bethe-Salpeter equation (BSE) is solved which gives the complete spectra, which is computationally very expensive. These excitonic effects are also effectively treated using TD-DFT. TD-DFT is nowadays widely used approach for the calculations of electronic excitation energies of molecules and solids, and are of reasonable accuracy with low computational cost. In our present calculations we have used the recently developed bootstrap kernel approximation¹ in order to include the excitonic effect. In the present work we have calculated the optical properties of the compounds CsCl, CsI, CsSrI₃², NaF, KF³⁻⁴, MgO, CaO, SrO, and BaO⁵ compounds including the excitonic effect.

COMPUTATIONAL DETAILS

We have used TD-DFT in order to include the excitonic effects by using newly developed bootstrap kernel approximation for the exchange correlation kernel which is implemented in the FP-LAPW method in elk code⁶, which is computationally cheap when compared to BSE calculations. We have used a 8x8x3 k-mesh for the CsSrI₃ compound and 25x25x25 for the other compounds. CsI is of CsCl type structure, NaF, KF, MgO, CaO, SrO, and BaO are of NaCl type structure and CsSrI₃ crystallize in orthorhombic structure. We have applied scissor correction for all these compounds based on the experimental band gap (TB-mBJ band gap values are considered where the experimental band gaps are not available) in order to correct the underestimation of DFT band gaps.

RESULTS AND DISCUSSION

The calculated imaginary parts of dielectric function for all the investigated compounds are shown in FIGURE. 1, FIGURE. 2, along with experimental and calculated RPA (Random Phase Approximation) spectra in order to compare with TD-DFT results. We observe improved spectra with the bootstrap kernel

approximation when compared to the normal DFT calculations. For the compounds CsCl, CsI, CsSrI₃ we observed the TD-DFT spectra to shift to low energy region when compared to RPA spectra which is clearly seen from FIGURE. 1. The lower edge of imaginary part of dielectric function shifts to lower energies when we move from CsCl to CsI because of decrease in the band gap. In the case of NaF, KF we observe additional peaks which might be due to the excitonic effect present in these compounds and is well comparable with the experimental spectra. These additional peaks are absent in the case of RPA spectra which fail to include the excitonic effect. In the higher energy region the RPA spectra is almost comparable with the TD-DFT results. For these compounds valence band is formed by the halide-*p* states and conduction band is formed by the alkali-*s/d* states and the lower energy peak in CsCl, CsI, NaF, KF is due to the transition from halide-*p* states to alkali-*s/d* states. TD-DFT spectra shows an enhanced height of lower energy peak for the compounds CsCl, CsI, CsSrI₃, NaF, and KF when compared to experimental and normal DFT calculations.

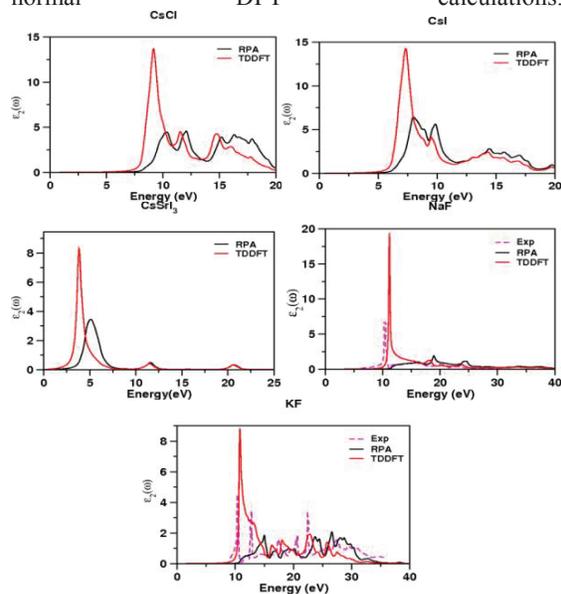


FIGURE 1. Imaginary part of dielectric function using elk code for the compounds (a) CsCl, (b) CsI, (c) CsSrI₃, (d) NaF, (e) KF including experimental spectra for the compounds NaF, KF (references are found in [7]).

We also studied the excitonic effects in MgO, CaO, SrO, and BaO compounds and are shown in FIGURE. 2, where we observe the experimental spectra to agree with the TD-DFT results. The low energy peak shifts further low as we move from MgO to BaO because of the decrease in the band gap. The experimental dielectric spectra of these compounds (CaO, SrO, and

BaO) shows a peak in the low energy region and it is well captured by TD-DFT calculations compared to normal DFT calculations indicating the importance of excitonic effect in these compounds. For the oxide compounds the valence band is formed due to oxide-*p* states and conduction band is formed due to the alkali-earth-*s/d* states. So the lower energy peak is due to transition from halide-*p* states to alkali earth -*d/s* states.

CONCLUSIONS

We have studied optical properties of CsCl, CsI, CsSrI₃, NaF and KF, MgO, CaO, SrO, and BaO using the newly developed bootstrap kernel approximation which includes the excitonic effects leading to improved results and the results agree well with experimental spectra.

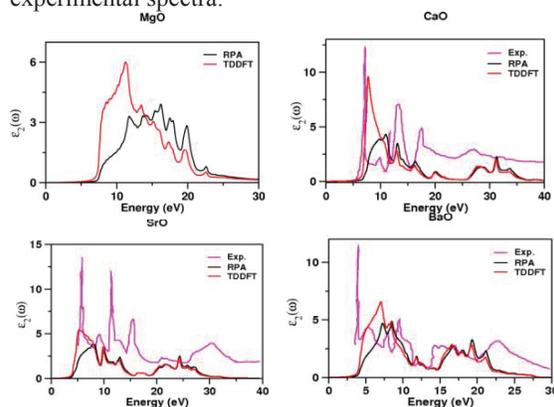


FIGURE 2. Imaginary part of dielectric function using elk code for the compounds (a) MgO, (b) CaO, (c) SrO, (d) BaO including experimental spectra (reference are found in [5]) for the compounds CaO, SrO, BaO.

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