Chapter 6

Conclusions

In this thesis, we have presented the study of optical properties of solids within linear response regime. We have used generalised gradient approximation (GGA) which had been discussed in detail in chapter 2. In Chapter 5, we have presented the calculation of electronic band structures, density of states and optical properties by using FP-LAPW method. We applied it to the case of beryllium chalcogenides (BeS, BeSe and BeTe), lead chalcogenides (PbS, PbSe, and PbTe), zinc chalcogenides (ZnS, ZnSe, and ZnTe) and stibiotantalite (SbTaO$_4$). The band structures are plotted along various symmetry directions in the Brillouin zone. The imaginary part of the dielectric function ($\varepsilon_2$) was also calculated for each system.

The band structures of beryllium chalcogenides (BeS, BeSe and BeTe) show indirect band gaps. Band gaps are 3.1 eV, 2.6 eV and 1.9 eV respectively from $\Gamma$ to $X$ along $\Delta$-direction for BeS, BeSe and BeTe respectively. For BeS, BeSe and BeTe the energy peak at 6.8 eV, 6.2 eV and 5.1 eV in the $\varepsilon_2(\omega)$ arises from the interband transition between the maximum of valence band at $\Gamma$ - edge and the bottom most conduction band at $W$ - edge. In comparison with the experimental data we find that the energy gaps are underestimated which is attributed to our use of GGA. We also identified the microscopic origin of the main features in the optical spectra and found that transitions between highest lying valence band (HVB) at $\Gamma$ symmetry point and lowest lying conduction band (LCB) at $X$ symmetry point are responsible most of the optical absorption in these systems.
For lead salts, we find direct energy band gaps of 0.40 eV, 0.24 eV, and 0.60 eV for PbS, PbSe, and PbTe respectively at the L symmetry point. Our results were compared with the experimental results and found to agree and also with theoretical results of others using different methods. The main peaks in the spectra are situated at 3.1 eV, 2.6 eV and 2.2 eV respectively for PbS, PbSe and PbTe. The results for band structure and DOS show that the energy gap for lead chalcogenides changes when S is replaced by Se and Te. This trend is attributed to the increase in bandwidth of the conduction band on going from S to Se and Te. For PbS the energy peak at 3.1 eV in the $\varepsilon_2(\omega)$ arises from the interband transition between the valence band in -0.9 eV and the bottom most conduction band at W-edge. The sharp energy peak at 2.6 eV for PbSe is due to the interband transition from the highest valence band -0.7 eV to the lowest conduction band at W-edge. The energy peak at 2.2 eV for PbTe is due to the interband transition between the valence band in -0.3 eV and the conduction band in 1.9 eV.

The band structure for zinc chalcogenides also shows direct energy band gap at Γ symmetry point. The experimental band gaps are 3.8 eV, 2.8 eV, and 2.4 eV for ZnS, ZnSe and ZnTe respectively. Our calculated band gaps are 2.8 eV, 1.4 eV, 1.4 eV respectively for ZnS, ZnSe and ZnTe. Our analysis of the $\varepsilon_2(\omega)$ curves show that the first critical points of the dielectric function occurs at 2.8 eV, 1.3 eV and 1.25eV. These critical points are followed by small structure located at 5 eV in ZnS, 3.7 eV in ZnSe and 2.9 eV in ZnTe related to direct transition (L-L). The main peaks in the spectra are situated at 6.6eV, 5.5 eV and 4.5 eV respectively which are followed by pronounced peak situated at 7.9 eV, 7.1 eV and 5.9 eV. These peaks are primarily due to direct transition between valence band and conduction band above the Fermi energy at L-edge.
Our calculation of the electronic structure and DOS of ferroelectric SbTaO$_4$ in the orthorhombic phase using the FP-LAPW method shows that the fundamental gap of SbTaO$_4$ is indirect at $\Gamma$-R point with a band gap of 1.9 eV. From the DOS and band structure features, it can be concluded that the system is semiconductor. Further calculations of the x-ray spectra are to be investigated for the improvement of our results.

In the above studied systems, in comparison with the experimental data we find that the energy band gaps are underestimated which is attributed to our use of GGA. For optical calculation, a dense mesh of k points is required for accurate results. In our case, we took only 5000 k points. We are working on higher k points for the improvement of our results. Overall, we can conclude that FP-LAPW method is very appropriate for the calculation of electronic band structure and optical properties of solids.