Abstract

Energy materials having versatile physical properties are the need of the present scenario and society. This by far intact competency promises the latest change in the field of renewable energy resources. This is possible by investigating the different functions into one highly crucial material. Ternary chalcopyrite structured semiconductor compounds with the general valence type I-III-VI have long been investigated due to their applications in optoelectronics, non-linear optics, and as light absorbers in solar cells. However, comparatively a little attention has been given on the II-IV-V chalcopyrite structured materials, which offer additional chemical flexibility and a suitable bandgap for a single-junction solar cell. Three different combinations of compounds including $LiGaX_2$ (X= S, Se, Te), $AuMX_2$ (M = B, Al, Ga, In; X = S, Se, Te), and $NaGaX_2$ (X= S, Se, Te) have been investigated to calculate the band gaps, absorption coefficient $\alpha(\omega)$, refractive index $n(\omega)$, extinction coefficient $k(\omega)$, energy loss spectrum $L(\omega)$, reflectivity $R(\omega)$, and optical conductivity $\sigma(\omega)$. Transport properties of $LiGaX_2$ (X= S, Se, Te) and $NaGaX_2$ (X= S, Se, Te) including See-beck coefficient (S), electrical (σ) and thermal (k) conductivities, the figure of merit (ZT), and power factor (PF) in the temperature range 100-900 K were also studied to compute the thermoelectric response of these compounds. This research work focuses on the investigation of physical properties of optoelectronic materials to recommend the highly efficient and new semiconductor materials that are more feasible for photovoltaic applications. Chalcopyrites with potential applications have already been highlighted by the researchers. The new chalcopyrite materials can be explored by tuning bandgap. In this thesis, we present the first principle study of bandgap engineering of ternary chalcopyrite structures within the framework of density functional theory (DFT) by employing the Full-Potential Linearized Augmented Plane Wave Plus Local Orbitals (FLAPW+LO) method. The modified Becke-Johnson (mBJ) approximation was used for the exchange-correlation energy in the strongly correlated electron systems. This functional is considered much better for bandgap prediction. Pseudopotential approximation was also included while calculating results using VASP. Therefore, ternary chalcopyrite structured semiconductor compounds having space group $I\bar{4}2d$ were designed and hence ground state structural, electronic (density of states and band structures), optical, and transport properties have been studied by using DFT based codes VASP and WIEN2K. Initially, we relaxed the crystal structures through optimization to obtain ground state structural

parameters including lattice constants. Afterwards, we used that optimized lattice constants for the Self-Consistent Field (SCF) cycle for energy convergence. The physical properties were investigated to probe the electronic, optical, and thermoelectric behaviour of materials using ground state parameters. The bandgap values for $LiGaS_2$, $LiGaSe_2$, and $LiGaTe_2$ are 3.39, 2.83, and 1.96eV respectively fall in near ultraviolet and visible portions of solar light, $NaGaSe_2$ (1.7eV) and $NaGaTe_2$ (1.2eV) systems are found to be smaller than $NaGaS_2$ (2.1eV), and in case of $AuBX_2$ (X = S, Se, Te) and $AuMTe_2$(M = Al, Ga, In) each and every alloy possess finite band gaps with mBJ+SOC, whereas $AuInTe_2$ and $AuGaTe_2$ are almost of metallic nature ($E_g \sim 0.05$ eV). $AuBS_2$ and $AuBSe_2$ show indirect band gaps. The tellurides are efficacious direct band-gap semiconductors, while $AuBS_2$ and $AuBSe_2$ show indirect band gaps. The bandgap of $AuBSe_2$ was found to fall in the optimal region of the Shockley-Queisser (S-Q) limit. We observed that the band gap decreases as the anion (X) are changed. Hence the anion has been playing an important role in narrowing electronic bandgap through valence electron contribution in all materials. However, the optimized lattice constants, the maximum of dielectric functions and the refractive index increase in the same order. It can also be seen that the prominent peaks of dielectric functions and the refractive index significantly shifted towards lower energy regions in the S, Se, and Te series in case of $LiGaX_2$, $NaGaX_2$, $AuBX_2$, and $AuAlTe_2$. The calculated results show that these compounds are feasible for the absorption of infrared, visible, and near ultraviolet regions of the solar spectrum as they have high $\alpha(\omega)$, and low $L(\omega)$ and $R(\omega)$. The scatter graph for the comparison of the absorption coefficient of predicted compounds on $AuBX_2$ (X = S, Se, Te) and $AuAlTe_2$.with other solar materials $CuGaSe_2$, $CuGaS_2$, $CuInSe_2$, CdTe and $CH_3NH_3PbI_3$ reveals that these materials are much efficient with very high absorption coefficients and having maximum absorption ability of solar energy in the infrared and visible range compared to the conventional compounds. Moreover, it was found that $AuBTe_2$ has the largest value of $\alpha(\omega)$ (upto 3.1eV) and $AuAlTe_2$ was the most optical anisotropic, suggesting potential candidates for nonlinear optical applications. Such type of structures with suitable band gaps have the ability to absorb a sufficient amount of light, which can be used as more efficient for solar cells to meet the requirements of energy. Therefore, this research work will help the experimental researchers to establish a newly designed ternary chalcopyrite structures with excellent features for the fabrication of solar cell.