Dispersion of the second harmonic generation from CdGa$_2$X$_4$ (X = S, Se) defect chalcopyrite: DFT calculations

Abstract

All electron full potential linear augmented plane wave method was used for calculating the nonlinear optical susceptibilities of CdGa$_2$X$_4$ (X = S, Se) within the framework of density functional theory. The exchange correlation potential was solved by recently developed modified Becke and Johnson (mBJ) approximation. The crystal structure of CdGa$_2$S$_4$ and CdGa$_2$Se$_4$ reveals a large uniaxial dielectric anisotropy ensuing the birefringence of −0.036 and −0.066 which make it suitable for second harmonic generation. The second order susceptibility $|\chi_{ijk}^{(2)}(\omega)|$ and microscopic first hyperpolarizability $\beta_{ijk}(\omega)$ were calculated. The calculated $|\chi_{123}^{(2)}(\omega)|$ and $|\chi_{312}^{(2)}(\omega)|$ static values for the dominant components found to be 18.36 pm/V and 22.23 pm/V for CdGa$_2$S$_4$ and CdGa$_2$Se$_4$. Both values shifted to be 60.12 pm/V and 108.86 pm/V at $\lambda = 1064$ nm. The calculated values of $\beta_{123}(\omega)$ is $6.47 \times 10^{-30}$ esu at static limit and $12.42 \times 10^{-30}$ esu at $\lambda = 1064$ nm for CdGa$_2$S$_4$, whereas it is $8.82 \times 10^{-30}$ esu at static limit and $20.51 \times 10^{-30}$ esu at $\lambda = 1064$ nm for CdGa$_2$Se$_4$. The evaluation of second order susceptibilities and first hyperpolarizabilities suggest that CdGa$_2$X$_4$ possess huge second harmonic generation.

Keywords; Defect chalcopyrites, Birefringence, Nonlinear optical susceptibilities