Dispersion of the second harmonic generation from $CdGa_2X_4$ (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 $CdGa_2X_4$ (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 $CdGa_2S_4$ and $CdGa_2Se_4$ 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 $CdGa_2S_4$ and $CdGa_2Se_4$. Both values shifted to be 60.12 pm/V and 108.86 pm/V at λ = 1064 nm. The calculated values of $\beta_{123}(\omega)$ is 6.47×10^{-30} esu at static limit and 12.42×10^{-30} esu at λ = 1064 nm for $CdGa_2S_4$, whereas it is 8.82×10^{-30} esu at static limit and 20.51×10^{-30} esu at λ = 1064 nm for $CdGa_2Se_4$. The evaluation of second order susceptibilities and first hyperpolarizabilties suggest that $CdGa_2X_4$ possess huge second harmonic generation.