Dispersion of the linear and nonlinear optical susceptibilities of disilver germanium sulfide from DFT calculations

Abstract

The dispersion of the linear and nonlinear optical susceptibilities is calculated for disilver germanium sulfide (Ag2GeS3) using the all-electron full potential linearized augmented plane wave (FP-LAPW) method. Calculations are performed with four exchange correlations namely local density approximation (LDA), general gradient approximation (GGA), Engel-Vosko generalized gradient approximation (EVGGA), and modified Becke-Johnson potential (mBJ). Our calculations give a band gap of 0.40 eV (LDA), 0.42 eV (GGA), 1.03 eV (EVGGA), and 1.30 eV (mBJ) in comparison with our measured gap (1.98 eV). The mBJ exchange correlation gives the best agreement with experiment. We find that the calculated linear optical susceptibilities of Ag2GeS3 show considerable anisotropy which is useful for second harmonic generation andoptical parametric oscillation. To analyze the spectra of the calculated χ 113 (2) (ω), χ 232 (2) (ω), χ 311 (2) (ω), χ 322 (2) (ω), and χ 333 (2) (ω), we have correlated the features of these spectra with the features of ε2(ω) spectra as a function of ω/2 and ω. From the calculated dominant component |χ 333 (2) (ω)|, we find that the microscopic second-order hyperpolarizability, β333, the vector components along the dipole moment direction is 41.2 × 10-30 esu at static limit and 222.9 × 10-30 esu at λ = 1064 nm.