

Acentric nonlinear optical 2,4-dihydroxyl hydrazone isomorphous crystals with large linear, nonlinear optical susceptibilities and hyperpolarizability

Abstract

A systematic ab initio study of the linear, nonlinear optical susceptibilities, and hyperpolarizability of noncentrosymmetric-monoclinic 2,4-dihydroxyl hydrazone isomorphous crystals (DHNPH) within density functional theory in the local density approximation (LDA), general gradient approximation (GGA), the Engel-Vosko generalized gradient approximation (EV-GGA) and modified Becke-Johnson potential (mBJ) has been performed. The complex dielectric susceptibility dispersion, its zero-frequency limit and the birefringence are studied. Using scissors' corrected mBJ we find a large uniaxial dielectric anisotropy (~ 0.56) resulting in a significant birefringence (0.61). We also find that 2,4-DHNPH possess large second harmonic generation. The calculated second order susceptibility tensor components for the static limit $|\chi_{111}^{(2)}(0)|$ and $|\chi_{111}^{(2)}(\omega)|$ at $\lambda=1.9 \mu\text{m}$ (0.651 eV) and at $\lambda = 1.064 \mu\text{m}$ (1.165 eV) are 53, 91, and 209 pm/V, respectively. A remarkable finding, applying the scissors' correction has a profound effect on value, magnitude and sign of $\chi_{ijk}^{(2)}(\omega)$. In addition we have calculated the microscopic hyperpolarizability, β_{111} , vector component along the principal dipole moment directions for the dominant component. We find that the value of β_{111} equal to $47 \cdot 10^{-30}$ esu, in good agreement with the measured value ($48.2 \cdot 10^{-30}$ esu).