Investigation of the linear and nonlinear optical susceptibilities of $KTiOPO_4$ single crystals: Theory and experiment

Abstract

Experimental and theoretical studies of the linear and nonlinear optical susceptibilities for single crystals of potassium titanyl phosphate $KTiOPO_4$ are reported. The state-of-the-art full potential linear augmented plane wave method, based on the density functional theory, was applied for the theoretical investigation. The calculated direct energy band gap at γ, using the Engel-Vosko exchange correlation functional, is found to be 3.1 eV. This is in excellent agreement with the band gap obtained from the experimental optical absorption spectra (3.2 eV). We have calculated the complex dielectric susceptibility ε(Ω)dispersion, its zero-frequency limit $\varepsilon_1(0)$ and the birefringence of $KTiOPO_4$. The calculated birefringence at the zero-frequency limit δn(0) is equal to about 0.07 and δn(Ω) at 1.165 eV (Λ = 1064 nm) is 0.074. We also report calculations of the complex second-order optical susceptibility dispersions for the principal tensor components: $\xi_{113}^{(2)}(\Omega)$, $\xi_{232}^{(2)}(\Omega)$, $\xi_{311}^{(2)}(\Omega)$, $\xi_{322}^{(2)}(\Omega)$, and $\xi_{333}^{(2)}(\Omega)$. The intra- and interband contributions to these susceptibilities are evaluated. The calculated total second order susceptibility tensor components $|\xi_{ijk}^{(2)}(\Omega)|$ at Λ = 1064 nm for all the five tensor components are compared with those obtained from our measurements performed by nanosecond Nd:YAG laser at the fundamental wavelength (Λ = 1064 nm). Our calculations show reasonably good agreement with our experimental nonlinear optical data and the results obtained by other authors. The calculated the microscopic second order hyperpolarizability, β_{333}, vector component along the dipole moment direction for the dominant component $\xi_{333}^{(2)}(\Omega)$ is found to be 31.6 - 10^{-30} esu, at Λ = 1064 nm.