Dispersion of linear and nonlinear optical susceptibilities and the hyperpolarizability of 3-methyl-4-phenyl-5-(2-pyridyl)-1,2,4-triazole

Abstract

As a starting point for our calculation of 3-methyl-4-phenyl-5-(2-pyridyl)- 1,2,4-triazole we used the XRD data obtained by C. Liu, Z. Wang, H. Xiao, Y. Lan, X. Li, S. Wang, Jie Tang, Z. Chen, J. Chem. Crystallogr., 2009 39 881. The structure was optimized by minimization of the forces acting on the atoms keeping the lattice parameters fixed with the experimental values. Using the relaxed geometry we have performed a comprehensive theoretical investigation of dispersion of the linear and nonlinear optical susceptibilities of 3-methyl-4-phenyl-5-(2-pyridyl)-1,2,4-triazole using the full potential linear augmented plane wave method. The local density approximation by Ceperley-Alder (CA) exchange-correlation potential was applied. The full potential calculations show that this material possesses a direct energy gap of 3.4 eV for the original experimental structure and 3.2 eV for the optimized structure. We have calculated the complex's dielectric susceptibility $\varepsilon(\omega)$ dispersion, its zero-frequency limit $\varepsilon_1(0)$ and the birefringence. We find that a 3-methyl-4-phenyl-5-(2-pyridyl)-1,2,4-triazole crystal possesses a negative birefringence at the low-frequency limit $\Delta n(0)$ which is equal to about -0.182 (-0.192) and at λ = 1064 nm is -0.193 (-0.21) for the non-optimized structure (optimized one), respectively. We also report calculations of the complex second-order optical susceptibility dispersions for the principal tensor components: $\chi(2)123(\omega)$, $\chi(2)231(\omega)$ and $\chi(2)312(\omega)$. The intra- and inter-band contributions to these susceptibilities are evaluated. The calculated total second order susceptibility tensor components at the low-frequency limit $\chi(2)ijk(0)$ and $\chi(2)ijk(\omega)$ at λ = 1064 nm for all the three tensor components are evaluated. We established that the calculated microscopic second order hyperpolarizability, β_{ijk}, the vector component along the dipole moment direction, at the low-frequency limit and at λ = 1064 nm, for the dominant component $\chi(2)123(\omega)$ is 4.99×10^{-30} esu (3.4×10^{-30} esu) and 7.72×10^{-30} esu (5.1×10^{-30} esu), respectively for the non-optimized structure (optimized structure).