

CHAPTER

6

Part I

INVESTIGATION OF THE LINEAR AND NONLINEAR OPTICAL SUSCEPTIBILITIES OF KTiOPO_4 SINGLE CRYSTALS: THEORY AND EXPERIMENT

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 $\epsilon(\omega)$ dispersion, its zero-frequency limit $\epsilon_1(0)$ and the birefringence of KTiOPO_4 . The calculated birefringence at the zero-frequency limit $\Delta n(0)$ is equal to about 0.07 and $\Delta n(\omega)$ at 1.165 eV ($\lambda=1064$ nm) is 0.074. We also report calculations of the complex second-order optical susceptibility dispersions for the principal tensor components: $\chi_{113}^{(2)}(\omega)$, $\chi_{232}^{(2)}(\omega)$, $\chi_{311}^{(2)}(\omega)$, $\chi_{322}^{(2)}(\omega)$ and $\chi_{333}^{(2)}(\omega)$. The intra- and inter-band contributions to these susceptibilities are evaluated. The calculated total second order susceptibility tensor components $|\chi_{ijk}^{(2)}(\omega)|$ at $\lambda=1064$ 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 ($\lambda=1064$ 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 $\chi_{333}^{(2)}(\omega)$ is found to be 31.6×10^{-30} esu, at $\lambda=1064$ nm.

6.1 HISTORICAL REVIEW

Potassium titanyl phosphate KTiOPO_4 (KTP) is a Nonlinear Optical (NLO) single crystal which is commonly used for frequency doubling diode pumped solid-state lasers such as Nd:YAG and other neodymium doped lasers. The crystal structure of KTP was first determined by Tordjman *et al.* [1]. KTP has a large second order electronic dielectric constant [2], low thermal expansion coefficient and a slow variation of the optical constants with temperature (excellent thermal stability). These facts make KTP very favorable for use as second harmonic generators of near-infrared laser light [3-5]. It is highly transparent for spectral range between 350–2700 nm with a reduced transmission up to 4500 nm where the crystal is effectively opaque [4]. Its effective Second Harmonic Generation (SHG) coefficient is about three times higher than KDP (KH_2PO_4).

It has a Mohs hardness of about 5. At the same time KTP is an inexpensive ferroelectric material belonging to the family of isomorphic nonlinear optical crystals with the general formula MTiOXO_4 ($M=[\text{K}, \text{Rb}, \text{Tl}, \text{Cs}]$, $X=[\text{P}, \text{As}]$) and is used for electro-optic applications [6]. KTP possesses a high damage threshold ($\sim 9.6 \text{ GW/cm}^2$) which makes it very useful for application in high power laser light modulation [6]. However, it is prone to photochromic damage (called grey tracking) during high-power 1064 nm second harmonic generation which results in its limited use in low and mid-power systems. The ion exchange properties of KTP make it useful in waveguide applications [6]. Stucky *et al.* [4] have presented an extensive review of the crystal structure and physico-chemical properties of KTP and other isostructural compounds. KTP is also frequently used as an optical parametric oscillator for near IR generation up to 4 μm . It is particularly suited to high power laser light operation as an optical parametric oscillator due to its high damage threshold and large crystal aperture [4]. The high degree of birefringent walk off between the pump signal and idler beams present in this material limits its application as an