Abstract

Fundamental bulk properties of the isomorphic ferroelectrics lithium niobate ($LiNbO_3$, LN) and lithium tantalate ($LiTaO_3$, LT) are investigated from first principles. In the first part of the thesis the vibrational properties of stoichiometric $LiNbO_3$ and $LiTaO_3$ are analyzed within density-functional perturbation theory in order to obtain the complete phonon dispersion of the materials. The presence of phonons with imaginary frequencies for the paraelectric phases reveals that they do not correspond to a minimum-energy structure, which is compatible with an order-disorder-type phase transition. The calculated zero-point renormalization of the electronic Kohn-Sham eigenvalues of LN indicates a vibrational correction of the electronic band gap of 0.41 eV at 0 K, which is in excellent agreement with the extrapolated temperature-dependent measurements. Also, the additional temperature-dependent redshift of the band gap is evaluated and amounts up 0.1 eV at room temperature.

The second part of the thesis focuses on the optical properties of LN. The dielectric function of pristine, congruent and titanium-doped lithium niobate is calculated within time-dependent density-functional theory. A model long-range contribution is employed for the exchange-correlation kernel in order to account for the electron-hole binding. The effects of polarons that occur at isolated niobium defects and bipolarons that extend to the neighboring regular niobium atom are discussed in detail. Four different niobium point defects and simple defect structures are modeled and analyzed. The experimentally observed polaron absorption features can be best explained by the interstitial-vacancy pairs Nb_V–V_{Li}. Further studies cover the influence of substitutional titanium atoms primarily on lithium sites. It is shown that an increasing titanium concentration enhances the values of the refractive indices and the reflectivity.