Optical response of strained $LiNbO_3$ crystals from first principles

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Introduction

The investigation of optical response study has become a widely used non-destructive method to characterize crystalline solids and nanostructures. X-ray diffraction measurements have shown that domain walls in lithium niobate (LN) behave like compressed bulk material [1]. Therefore, studying the optical response of LN as a function of compression may help to understand the optical response of the domain walls of LN. For this purpose, we have calculated the optical susceptibility, including second harmonic (SHG) and third harmonic generation (THG), of strained LN from first principles.

Methodology

In order to model strained LN, we have done calculations within density functional theory (DFT) as implemented in VASP [2]. For the exchange-correlation functional, the formulation of PBE [3] has been used. In a first approximation, compression was modeled by decreasing the lattice constants of the orthorhombic unit cell in x-, y- and z-direction, respectively, and minimizing the Hellmann-Feynman forces. This has been done in steps of 0.8 % up to 2.4 % with respect to the lattice constant.

The optical response has been calculated using time-dependent DFT (TDDFT) as implemented in Yambo [4]. The Bloch wave functions are calculated with QuantumEspresso using an $8 \times 5 \times 3$ k-point mesh, which corresponds to 61 irreducible k-points. A Kohn-Sham basis set consisting of the 100 highest valence bands and 100 lowest conduction bands has been considered for the calculation of the time-evolution of the dynamical polarization in Yambo.

Results

Our calculations reveal that structural strain strongly affects all the components of the second- and third-order polarizability tensor χ^2 and χ^3. Changes of the intensities and frequency shifts of the optical signatures occur. For instance, our calculations predict a roughly linearly increasing intensity for the $|\chi^2_{zzz}|$ component, as can be seen in Fig. 1 (a).

Due to the threefold rotational symmetry, unstrained LN has four independent χ^2 elements [5]. In contrast, when compressed in x- and y-direction, the degeneracy of some identical χ^2 components is lifted, as shown by our calculations. In particular, $|\chi^2_{yyy}|$ and $|\chi^2_{yxx}|$, which match in unstrained LN, split strongly upon compression in x-direction, as displayed in Fig. 1 (b) for a wavelength of 1035.90 nm. As shown in Fig.1 (b), $|\chi^2_{zyy}|$ and $|\chi^2_{zxx}|$ are also split upon compression in x-direction.

In addition, linear optical properties such as the refractive index as well as the birefringence are obtained from the calculated dielectric tensor as a function of compression. For instance, the birefringence for a wavelength of 1035.90 nm in dependence on compression in z-direction is shown in Fig. 1 (c). As can be seen, the birefringence is linearly decreasing with compression in z-direction.

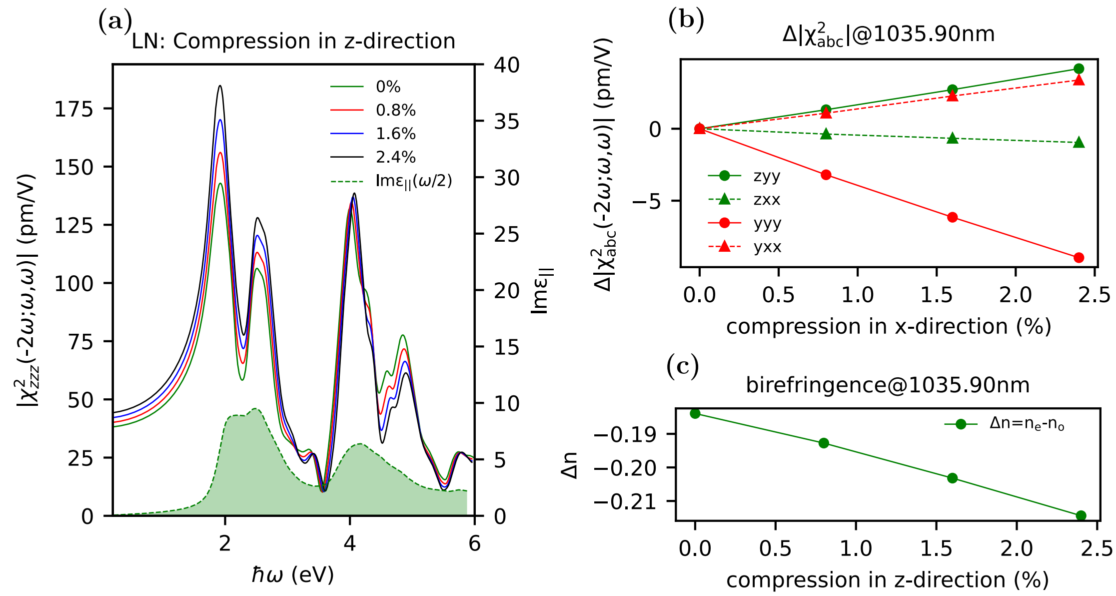

Figure 1: (a) The calculated SHG spectra of $|\chi^2_{zzz}|$ and the imaginary part of the extraordinary dielectric function of LN compressed in z-direction. (b) The relative change of the calculated intensity of $|\chi^2_{yyy}|$, $|\chi^2_{yxx}|$, $|\chi^2_{zyy}|$ and $|\chi^2_{zxx}|$ for a wavelength of 1035.90 nm of LN for compression in z-direction. (c) The calculated birefringence for a wavelength of 1035.90 nm of LN for compression in z-direction.

Conclusions

Both SHG and THG spectra of unstrained and strained LN are obtained from our calculations. Changes in the intensity and frequency shifts are yielded for all components. Due to symmetry reduction, identical χ^2components are lifted for compression in x- and y-direction. Our results may help to understand the optical response of domain walls in LN. Additionally, knowledge of the refractive index and birefringence is important for applications such as Ti waveguides.

References

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