Optoelectronic properties of barium titanate from first principles

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Linear electro-optical modulators based on barium titanate (BTO) integrated onto a silicon photonics chip can greatly improve the bandwidth of telecommunication devices and help reduce their power consumption. The underlying physics relies on the so-called Pockels effect, where the optical refractive index of a material can be modulated by an applied electric field.

In order to fabricate high-performance BTO modulators, a precise understanding of the Pockels effect and of its dependence on crystal defects is required. For this, we developed a framework to calculate the electronic and vibrational contributions to the Pockels tensor from first-principles using DFT, the Berry-phase formalism, and a finite-difference approach. Additionally, anharmonic dynamic instabilities of the BTO structure at room temperature are stabilised thanks to a supercell construction, where atomic degrees of freedom are enabled along the atomic displacements directions of the lower-temperature stable phase.

We show that the r13, r23 and r33 elements of the tetragonal BTO Pockels tensor can be accurately calculated within the Born-Oppenheimer approximation, as they exclusively depend on the electronic response to an external electric field. In contrast, the r42 and r51 tensor elements are subjected to strong vibrational contributions via the change in Raman polarizability, optical phonon modes polarities and frequencies. Since the tetragonal phase of BTO, which is stable within the range of 300K to 400K, is of high practical relevance, AIMD calculations must be introduced into our framework to correct for the effects of finite temperature.