Stress-dependent Bulk Photovoltaic Effect in Donor-doped $LiNbO_3$: Relation Between Defect Structure, Band Structure and Dielectric Properties

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Lithium niobate ($LiNbO_3$, LN) is a prototypical material for the study of the many of the properties of ferroelectrics. Single crystals of high quality can be grown by the Czochralski method, resulting in superior optical properties. It also displays a pronounced 'anomalous' or 'bulk' photovoltaic effect (BPVE); unlike the typical photovoltaic effect due to potential differences at interfaces such as Schottky barriers or p-n-junctions, the BPVE is based on long-range charge transport relying on the non-centrosymmetric crystal structure of the ferroelectric and can create voltages in the order of hundreds of volts in bulk crystals.

Perhaps most importantly, LN is characterized by a rich intrinsic defect structure that can be further modified either by doping with a wide range of cations during the growth process or post-growth high-temperature annealing in a defined atmosphere. Most dopants are incorporated as donors, creating well-defined optical absorption lines. Additionally, they significantly decrease the width of the energetic band gap. While the optical properties of differently doped materials are well studied, there is little knowledge about the interrelation between the energetic position of the donor center in the band gap, the width of the band gap, the dielectric response of the system and the bulk photovoltaic charge transport mechanism.

To elucidate the physical mechanism driving the bulk photovoltaic charge transport, this presentation investigates the stress-dependence of the BPVE of LN single crystals doped with different donor elements such as Fe, Mn, Er and Co. First, it is shown how the light-induced short-circuit current through the crystals changes under uniaxial compressive stress. By varying the crystal orientation and light polarization direction, all accessible components of the bulk photovoltaic tensor are measured. It is demonstrated how the symmetry breaking induced by the mechanical stress causes the appearance of tensor components that are zero in the unstressed system for symmetry reasons. The observed stress-induced changes can be ascribed either to changes in the light-induced excitation of charges from the donor centers or the subsequent directed movement of the charges before thermalization. To separate these contributions, the optical absorption of the unstressed and stressed systems is measured, as the number of photoexcited electrons is proportional to the optical absorption. It is shown that the optical absorption changes notably under stress, but that this change is not sufficient to account for the observed variation of the BPVE. The directed movement of photoexcited charge carriers is proposed to be based on hopping between neighboring Nb_{Nb}^{4+} polaronic states. To get a rough estimate of the modification of this hopping process by stress, the dielectric permittivity measured with and without stress are compared and related to changes in the phonon spectrum that govern the polaron behavior. Finally, the results are discussed by comparing the differences between differently doped systems.