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NLO characterization of harmonic LNT nanoparticle pellets by means of nonlinear diffuse fs-pulse reflectometry

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Lithium niobate tantalate solid solutions ($\text{LiNb}_{1-x}\text{Ta}_x\text{O}_3$, LNT) represent a promising material system for nonlinear photonics and quantum technologies. It is because their nonlinear optical and photophysical properties can be selectively tuned by composition ($0 < x < 1$) from the properties of the edge compositions lithium niobate (LN, $x = 0$) and lithium tantalate (LT, $x = 1$). So far, the exact relationship between composition and material properties, and especially with respect to the nonlinear optical properties, is poorly understood. One of the reasons is the availability of LNT bulk crystals with high optical quality. Thus, it is particularly unclear so far whether LNT crystals can be used for frequency conversion at all.

To address this question, we have studied LNT nanopowder samples [provided by the Institute of Energy Research and Physical Technologies, Clausthal. See also: Vasylechko et al., *crystals* **2021**, 11, 755] using the method of nonlinear diffuse fs-pulse reflectometry. The advantage of this method is the possibility to obtain a quantitative measure, called 'harmonic ratio' about the frequency conversion properties of materials that are only available as powder samples. Thus, the fulfillment of a phase matching condition below a characteristic particle size is not required.

The paper shows the methodological requirements for this type of studies, the procedure of sample preparation, the obtained results for LN, LT and LNT crystals and explains the determination as well as the impact of the 'harmonic ratio' as a qualitative quality factor of the powder samples. The influence of the particle properties (size distribution, pellet thickness, etc.) but also of the pulse parameters (pulse duration, pulse peak intensity, etc.) is discussed. The results are compared with data from X-ray structure analysis. The possibility to tune second harmonic emission within the visible and near-infrared spectral range is successfully demonstrated.

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