Time resolved sum-frequency generation in lithium niobate tantalate nanoparticles

Jan Klenen,[1,2] Laura Vittadello,[1,2] Bjoern Bourdon[1,2],[2] Mirco Imlau[1,2]

[1]Department of Physics, Osnabrück University, Barbarastraße 7, Osnabrück, Germany
[2]Research Center for Cellular Nanoanalytics Osnabrück (CellNanOs), Osnabrück University, Barbarastraße 11, Osnabrück, Germany

Introduction

Polar oxide, nonlinear optical (NLO) nanoparticles are essential for the further understanding of light-matter interaction and light-induced charge transport phenomena in fundamental physics on the atomic scale, but also are promising for the development of novel NLO applications in the field of nanophotonics, such as nonlinear optogenetics in life sciences [1]. Apart from their pronounced NLO coefficients, polar oxide nanoparticles with diameters much below 100 nm enable spectral tuning by means of frequency conversion without the need for phase-matching. A much reported drawback, however, is their weak conversion efficiency, connected with low signal emission, that requires sophisticated detection technologies in spectroscopy and imaging applications. For this reason, ensemble measurements are attractive, i.e., the gathering of signals from a large number of particles. An important prerequisite is the reproducible preparation of pressed nanoparticle powder pellets [2, 3]. Furthermore, developing a thorough, experimentally validated understanding of the respective fundamental processes inside the pellet, such as the question about the complex optical paths taken in disordered media, represents a challenging task that has not been adequately addressed so far.

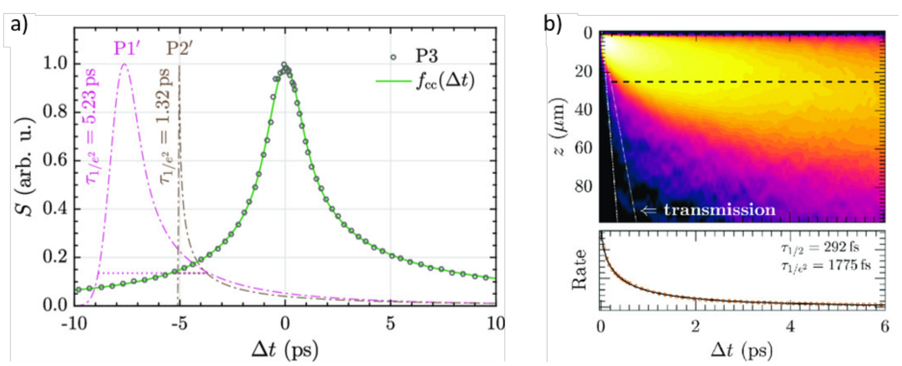

Figure 1: a) Measured SFG signal of two laser pulses ($P1$ and $P2$) in a HNP powder pellet as a function of temporal delay Δt between the two (circles) for pulse durations $\tau_{P1} = 1250$ fs and $\tau_{P2} = 44$ fs. b) Simulation of the scattering events per 60 fs time intervals at different depths inside the medium and the retrieved temporal pulse profile at a corresponding depth of 25 μm.

In this contribution, we focus on the particular case of ultrashort laser pulse propagation inside powder pellets of polar oxide, NLO nanoparticles and study the impact of composition in the solid solution $LiNb_{1-x}Ta_xO_3$ (LNT) with $0 < x < 1$) including the prominent edge compostions $LiNbO_3$ ($x = 0$) and $LiTaO_3$ ($x = 1$) [4, 5]. By recording the time-resolved sum-frequency generation (SFG) signal and by performing numerical studies incorporating multiple scattering and superdiffusive light propagation, we are able to retrieve temporal profiles of incident laser pulses on a sub-picosecond timescale (see Fig. 1).

This way, we are also able to predict the influence of pellets with differently sized particle distributions. Experimental validation is carried out within the framework of a series of measurements with particle sizes ranging from 50 to 400 nm. Moreover the influence of NLO properties on the pulse propagation is investigated. For instance, the pulses temporal evolution in pellets composed of LNT particles with different compositions x is monitored. We explore the possibility to apply our findings for short-pulse characterization, e.g., the determination of the chirp parameters usually determined from sophisticated femtosecond wave-mixing methods (see Fig. 2).

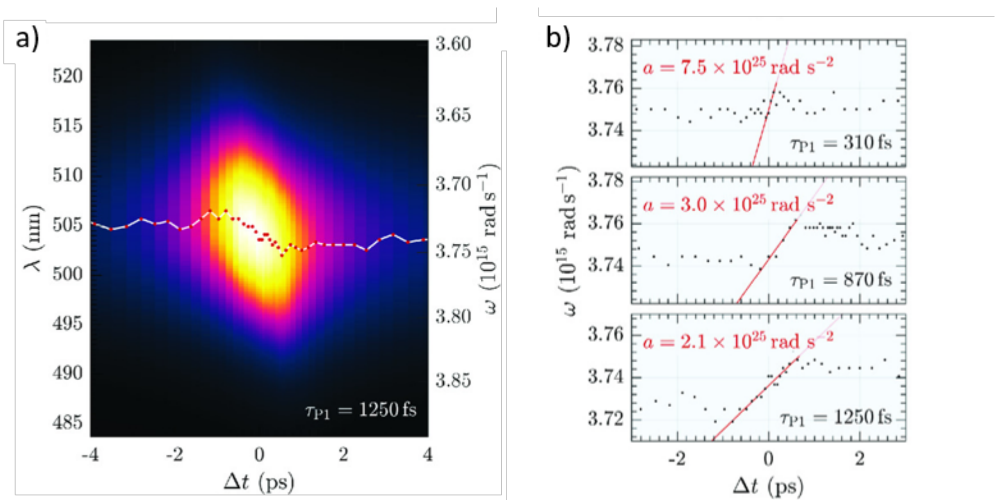

Figure 2: a) Instantaneous spectra of the SFG as a function of the temporal delay Δt between two differently colored pulses; respective maximums are highlighted by a dash-dotted line. b) Linear chirp parameter of the SFG with one initial pulse $P1$ being stretched to 310, 870, and 1250 fs.

Acknowledgments

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