

DF 3: Optical and nonlinear optical properties, photonic

Time: Monday 9:30–12:10

Location: H26

DF 3.1 Mon 9:30 H26

Scanning nonlinear absorption in lithium niobate over the time regime of small polaron formation — HOLGER BADORRECK, STEFAN NOLTE, ●FELIX FREYTAG, PIA BÄUNE, and MIRCO IMLAU — School of Physics, Osnabrueck University, Barbarastr. 7, 49076 Osnabrueck, Germany

Nonlinear absorption is studied in presence of small polaron formation in lithium niobate using the *z*-scan technique and ultrashort laser pulses with pulse durations of 70 - 1.000 fs. A model for the analysis of the transmission loss as a function of pulse duration is introduced that considers (i) the individual contributions of two-photon and small polaron absorption, (ii) the small polaron formation time and (iii) an offset time between the optical excitation of free carriers by two-photon absorption and the appearance of small polarons. It is shown that the model allows for the analysis of the experimentally determined *z*-scan data with high precision over the entire range of pulse durations using a two-photon absorption coefficient of $\beta = (5.6 \pm 0.8) \text{ mm/GW}$. A significant contribution by small polaron absorption to the nonlinear absorption is uncovered for pulse durations exceeding the characteristic small polaron formation time of ≈ 100 fs. It can be concluded that the small polaron formation time is as short as (70 - 110) fs and the appearance of small polaron formation is delayed with respect to two-photon absorption by an offset of about 80 fs. Financial support by the DFG (IM37/5-2, INST 190/137-1 FUGG, INST 190/165-1) is gratefully acknowledged. [Baddoreck et al., Opt. Mater. Express 5(12) 2729-2741 (2015)]

DF 3.2 Mon 9:50 H26

Luminescence of undoped lithium niobate — ●SIMON MESSERSCHMIDT, ANDREAS KRAMPF, and MIRCO IMLAU — Department of Physics, Osnabrück University, Germany

Luminescence of lithium niobate (LN) was studied for the first time 1973 by Blasse [Blasse, G. Phys. Stat. Sol. (a) 20, K99–K102 (1973)]. He detected a broadband light emission of LN under UV-excitation at temperatures below 80 K and assumed that the origin of the luminescence lies in the NbO_6 -octahedron. Various authors subsequently have found a connection between the ratio of $[\text{Li}]/[\text{Nb}]$ and the emitted peak wavelengths so that the red-shift in lithium-poor LN is interpreted as a disturbance of the NbO_6 -octahedron (Nb_{Li} -antisite defect). The emitted light is independent of the excitation wavelength and has a large Stokes shift. Therefore, the luminescence is described in a model of a self-trapped exciton (STE) [Wiegel, M., et al., J. Phys. Chem. Solids 55, 773–778 (1994)]. Until now, the interplay of STE and small polarons as well as parameters such as duration of the excitation pulse or crystal morphology to control the probability of the luminescence is unknown which is the topic of this work. Two experimental setups are used and initial results are presented. In the first setup, ns-pulses were used to excite a sample cooled to $T = 30$ K whose emitted light is detected with a gated photon counter. In a second setup, a luminescence upconversion detector utilizing ultrashort femtosecond pulses is applied to detect the dynamics of the luminescence light. Financial support by the DFG (IM 37/5-2, INST 190/165-1 FUGG) is gratefully acknowledged.

DF 3.3 Mon 10:10 H26

Ultrafast holographic spectroscopy for the study of small polarons in lithium niobate — ●STEFAN NOLTE, BJOERN BOURDON, FELIX FREYTAG, and MIRCO IMLAU — School of Physics, Osnabrück University, Germany

The optical generation of small bound polarons in lithium niobate can be used to record holographic gratings with a single nanosecond laser pulse and a diffraction efficiency above 20% [M. Imlau et al. Opt. Express 19, 15322 (2011), H. Brüning et al. Opt. Express 20, 13326 (2012)]. This finding is explained via the linear electro-optic effect and the space-charge field generated from a small polaron number density modulation with $E \approx 17 \text{ kV/cm}$. For the long-range transport of small polarons, we assume cascaded optical excitation during light exposure, i.e. the pulse duration has a strong impact on the hologram recording process [cf. H. Badorreck et al. Opt. Mater. Express 5, 2729 (2015)].

In this contribution, we study the holographic buildup as a function of pulse duration in the femtosecond time regime. Thus, a four-wave pump-probe experiment with continuously varied pulse-durations be-

tween $\tau = (70 - 500) \text{ fs}$ is used to record the time-dependent diffraction efficiency from a mixed holographic grating, based on nearly instantaneous nonlinearities $\chi^{(3)}$ and polaron effects. The results are discussed under consideration of polaron formation.

Financial support by the DFG (IM 37/5-2, INST 190/165-1 FUGG) is gratefully acknowledged.

DF 3.4 Mon 10:30 H26

OH^- as local, vibrational probe for the inspection of small, strong-coupling polarons — ●FELIX FREYTAG, PHILLIP BOOKER, and MIRCO IMLAU — School of Physics, Osnabrueck University, Barbarastr. 7, 49076 Osnabrueck, Germany

A *small polaron* is defined by the unit of a self-trapped carrier and the associated displacement of atomic equilibrium positions confined to a single lattice site [Emin, *Polarons*, Cambridge University Press (2013)]. There's an increasing attention to the structural distortion parameters of small polarons for nonlinear optical processes in polar oxides on the ultrafast time-scale [Imlau et al., Applied Physics Reviews 2, 40606 (2015)]. Even though, a multitude of knowledge has been gathered and a variety of experimental techniques like EPR/NMR measurements were applied, there is still no possibility for direct experimental access to the structural aspects of small polarons within a continuum and at elevated temperatures. Here, we propose a vibrational method to experimentally achieve local information about small polarons at room temperature. The absorption change of the OH^- stretching vibration is applied as local probe for optically excited small polarons with lithium niobate as an example; detected by time-resolved mid-infrared pump-probe spectroscopy. The change is explained straightforwardly by a local change of the lattice environment induced by small polaron formation. By analysis of the vibrational potential of the OH^- -stretching bond, we are able to estimate the polaron induced lattice distortion. Financial support by the DFG (IM 37/5-2, INST 190/165-1 FUGG) is gratefully acknowledged.

20 min. break

DF 3.5 Mon 11:10 H26

Quadrupolar Bulk SHG from First Principles — ●KLAUS-DIETER BAUER and KURT HINGERL — Zentrum für Oberflächen- und Nanoanalytik, Johannes Kepler Universität, Linz, Austria

Second harmonic generation (SHG) has been established as surface sensitive probe by exploiting symmetry-suppression of the otherwise dominant bulk dipole term in centrosymmetric systems. However, bulk quadrupole contributions are not suppressed and can be on the same order of magnitude as surface effects, yet abinitio calculations so far have neglected this term for bulk solids.

We discuss the formal relation of the optical quadrupole SHG response to a microscopic second-order response function $\chi^{(2)}(\mathbf{q}_0; \mathbf{q}_1, \mathbf{q}_2)$ and report our progress towards an implementation in the Vienna Ab-initio Simulation Package (VASP).

DF 3.6 Mon 11:30 H26

Nonlinear optical spectroscopy of niobate nanopowders — ●CHRISTIAN KIJATKIN¹, ANKE DUETTSMANN^{1,2}, KARSTEN KOEMPE², LAURA OLAH³, EVA TICHY-RACS³, ZSUZSANNA SZALLER³, and MIRCO IMLAU¹ — ¹School of Physics, Osnabrück University, Germany — ²Institute of Chemistry of New Materials, Osnabrück University, Germany — ³Wigner Research Centre for Physics, Budapest, Hungary

Nonlinear optical (NLO) nanocrystals (NCs) excel in a wide spectrum of applications, most notably as optical probes in high-resolution microscopy. This field has incited a multitude of NLO characterization approaches with the ultimate goal of providing comprehensive analysis of NCs. The plethora of available NLO materials in conjunction with different measurement techniques raises the question of comparability. A peculiar case is frequency-doubling which may be attributed to surface- or volume-second harmonic generation (SHG) as well as Hyper-Rayleigh scattering or higher-order multipole contributions. Extensive studies have been performed with little interconnections to each other. We present the results of our examinations on the individual NLO contributions arising from their respective origins by assessing the SHG behavior of niobate nanopowders (NaNbO_3 , $\text{LiNbO}_3:\text{Yb}$). Transmittive and reflective techniques are performed to

elucidate intensity and dispersion relation. In combination with linear diffuse reflectance scans, a prospective method of comparing NLO investigations is introduced. Financial support (DFG INST 190/165-1 FUGG, DAAD 57139940) is gratefully acknowledged.

DF 3.7 Mon 11:50 H26

Quasi-solitons and rogue waves generation in optical fibers

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Optical rogue waves are sharp, rare and extremely high power pulses. Experimental and numerical data suggest that these giant waves are due to at least two mechanisms of amplification, modulation instability (that allows the creation of quasi-solitons) and multiple inelastic quasi-soliton collisions. Using a generalized nonlinear Schrödinger equation we calculate the probability distribution function (PDF) for the intensity inside a fiber. We investigate pair-wise quasi-soliton collisions and propose a formula for the energy transfer. This allows us to implement an effective cascade model. The PDFs calculated using the two models are very similar. Our results suggest that once localized pulses are formed because of modulation instability an important contribution to rogue waves generation is quasi-soliton inelastic scattering.