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Statistical properties of photon modes in random arrays of ZnO nano-needles

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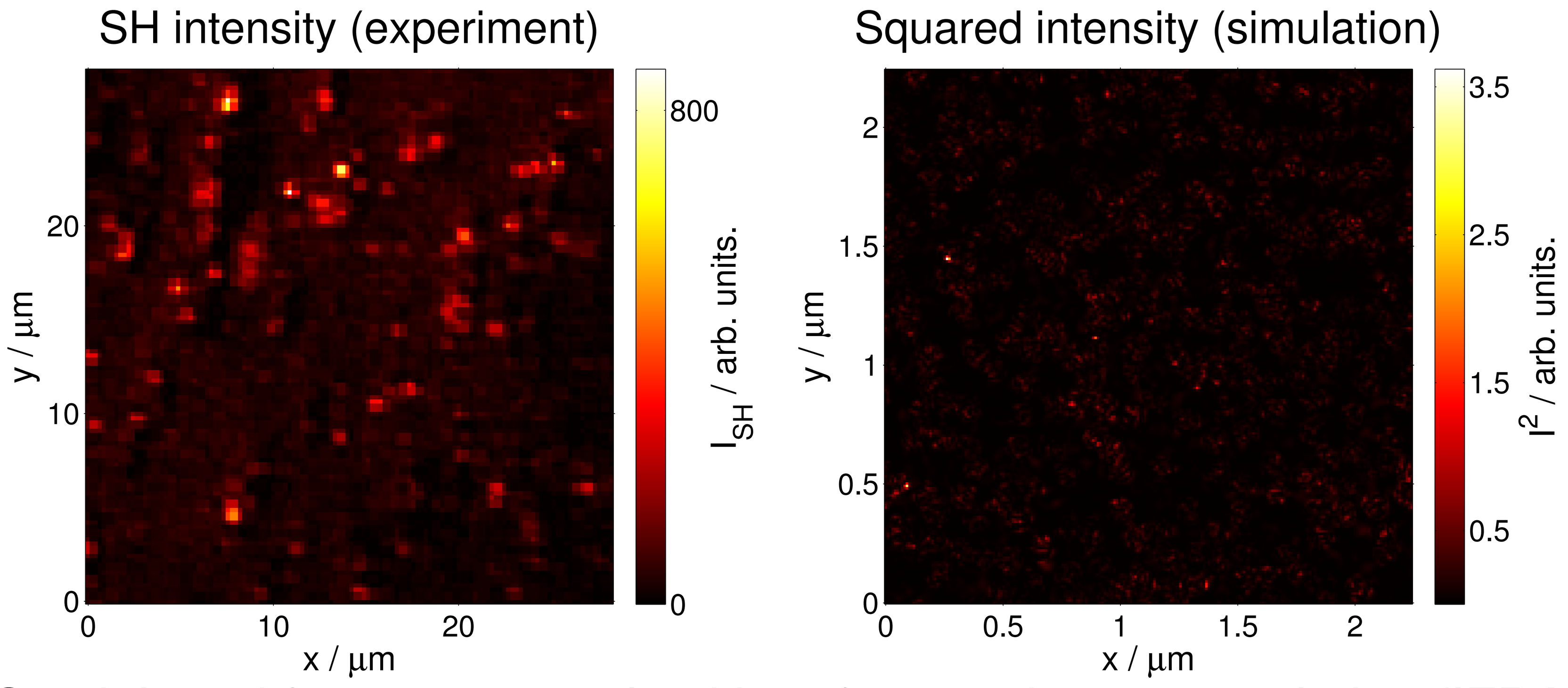
Introduction

Localization of electromagnetic waves in random media received renewed interest in the last years. Recent ultrafast optical experiments [1] indicate the existence of highly localized photon modes in a system of homogeneous, randomly distributed, vertically aligned ZnO nano-needles. In particular, hot spots in the spatial distribution of the second harmonic generation (SHG) were found.
In this work, we discuss the optical near field, which we obtain from full 3D solutions of Maxwell's equations of a model system in the time-domain. The spatial distribution of the electric near-field and the squared electric near-field intensity are investigated with statistical methods. The results are compared to the experimental findings and their time dependence is discussed.

Experiment and modeling

- ZnO nano-needles are grown on a sapphire substrate and produce a two-dimensional photonic crystal.
- Each nano-rod has an average diameter of 100nm and one very thin rod of 17.7nm on top; they are randomly distributed and do not overlap.
- Short laser pulses ($\approx 6fs$) with a center wavelength of about 800nm probe the sample.
- The experimental second harmonic (SH) intensity measured as a function of position can be compared to the calculated squared intensity, both as an indication of second order non-linear effects - showing hot spots:



- Spatial- and frequency-resolved interferometric autocorrelation (IFRAC) measurements prove the existence of slowly decaying fields.
- Note the difference of resolution in experiment and simulation with 300nm and 10nm, respectively.
- For the theoretical modelling, randomly distributed, 1μm long cylinders ($\varepsilon_{ZnO} = 4.016$) with a tailored correlation function are placed on top of a dielectric substrate ($\varepsilon_{sub} = 1.77$).
- Periodic boundary conditions with a unit cell size up to $5\mu m \times 5\mu m$ (≈ 1000 cylinders, limited by computational resources) have been chosen.
- Simulations have been done with the MIT Electromagnetic Equation Propagation (MEEP) package.

Log-normal intensity distribution

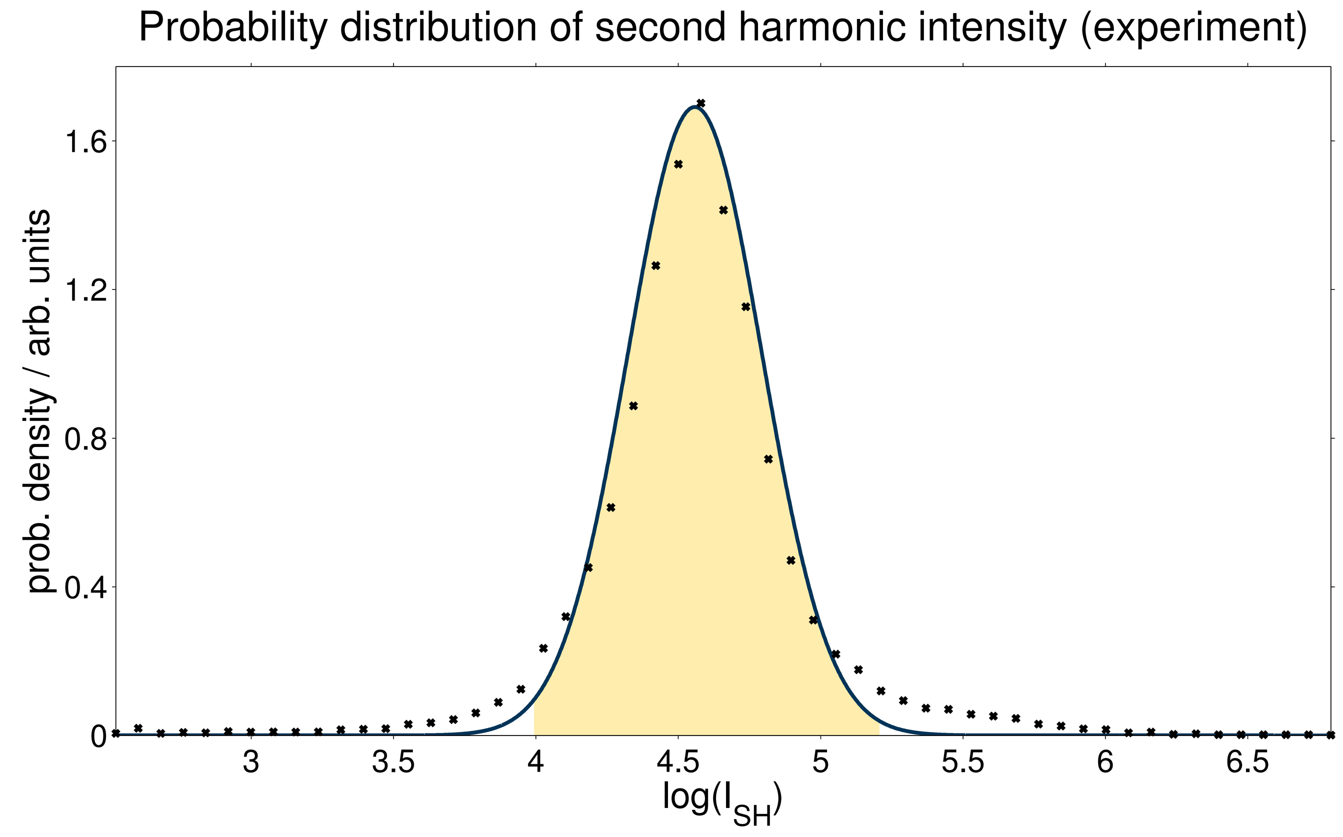
For anomalously localized states (ALS) [2] of waves in random media, log-normal distribution of the amplitude values are expected. In order to mathematically verify the presence of a log-normal distribution, mean μ and variance σ^2 are calculated and a chi-square-test has been done:

$$\chi^2 = \sum_{j=1}^{N} \frac{(O_j - E_j)^2}{E_j} \quad (1)$$

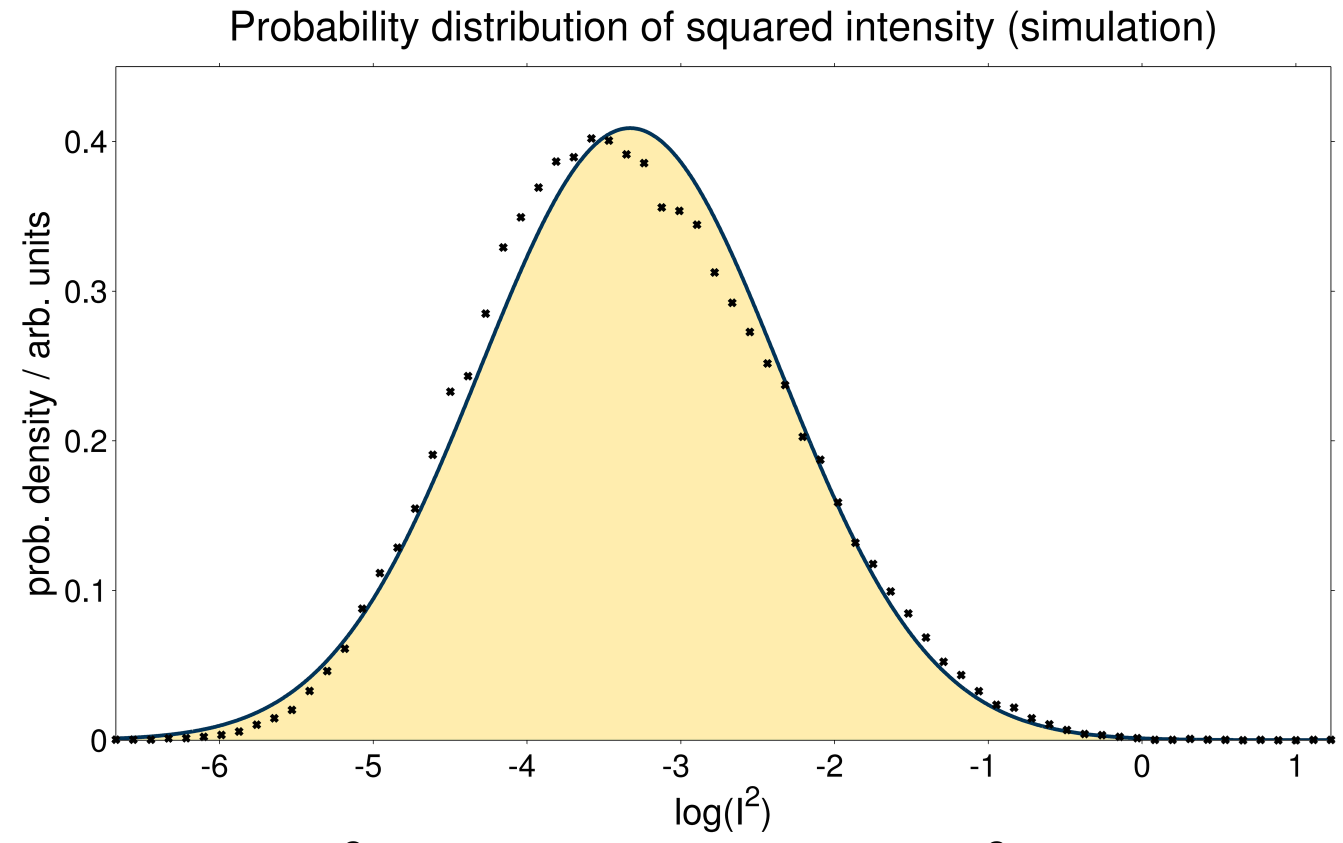
with observed counts O_j and frequencies E_j given by the log-normal distribution. χ^2 has to be less than the corresponding quantile with a defined statistical significance α of the chi-square-distribution with degree of freedom $d = N - 2$ (N ... number of intervals).

Comparison of intensity distributions

- A normal distribution (blue solid line) has been fit to the frequencies of the logarithmic non-linear intensity intervals (black points).
- Mean μ, variance σ^2 and skewness s as well as the results of the chi-square-test, χ^2 and d are specified below each plot.



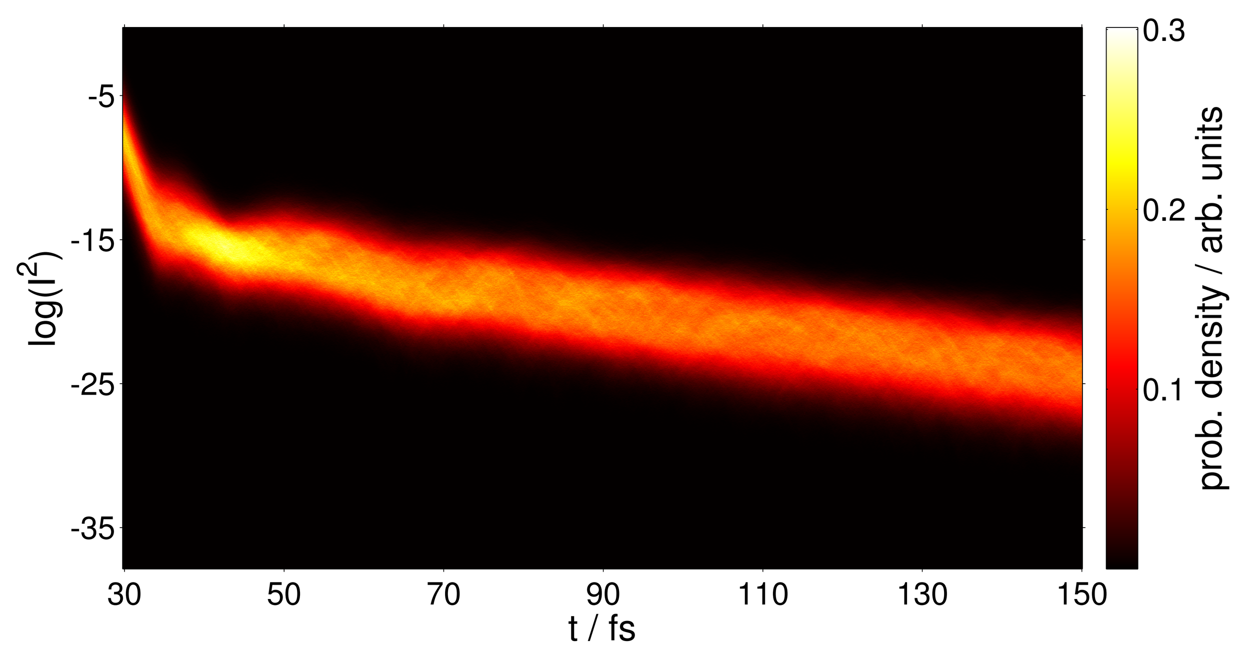
$\mu = 4.558, \quad \sigma^2 = 0.056, \quad s = 0.029, \quad \chi^2 = 371.1, \quad d = 15$



$\mu = -3.330, \quad \sigma^2 = 0.952, \quad s = 0.206, \quad \chi^2 = 574.9, \quad d = 61$

- In order to avoid artefacts from dark currents and noise, the calculation with the experimental data is restricted to $4.0 \leq \log(I_{SH}) \leq 5.2$ (orange).
- The well-fitted high-intensity tails enables us to study anomalously localized light modes in random media.

Temporal evolution of probabilities



- After the initial pulse passage ($t \leq 40fs$) the squared intensity distribution preserves its log-normal shape, drifts approximately linearly to lower average values μ, and broadens only weakly.

References & acknowledgements

[1] Experiments by Manfred Maschek, et al. from the Carl von Ossietzky Universität Oldenburg and Takashi, et al. from the University of Tokyo.

[2] V. M. Apalkov, M. E Raikh, B. Shapiro: "Anomalously Localized States in the Anderson Model", *Phys. Rev. Lett.* **92**, 066601 (2004).

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