



# Workshop on Complex Materials for Nonlinear Optics

29 - 31 January 2020, HIT E51, ETH Hönggerberg



Marie Skłodowska-Curie  
Actions



**MaP** Competence Center for  
Materials and Processes

## Program

*Wednesday 29 January 2020*

- 09.30**      **Registration Lab Tour**
- 10.30**      **Lab Tour I**
- 12.00**      **Registration & Lunch**
- 13.50**      **Opening**
- 14.00**      **Tailored Disorder I (Chair: R. Savo)**  
S. Skipetrov (14.00 - 14.35)  
R. Pierrat (14.35 - 14.55)  
G. Montemezzani (14.55 - 15.15)  
C. Denz (15.15 - 15.50)
- 15.50**      **Coffee Break**
- 16.30**      **Spatial, Temporal and Quantum Control I (Chair: M. Celebrano)**  
S. Gigan (16.30 - 17.05)  
A. Pasquazi (17.05 - 17.40)  
U. Tegin (17.40 - 18.00)
- 18.00**      **Poster Pitches**
- 18.30**      **Poster Session & Apéro**

*Thursday 30 January 2020*

- 09.00**      **From Bulk to 2D Systems I (Chair: R. Grange)**  
M. Scalora (09.00 - 09.20)  
A. Marini (09.20 - 09.55)  
A. Solntsev (09.55 - 10.15)  
M. Celebrano (10.15 - 10.35)
- 10.35**      **Coffee Break**
- 11.00**      **From Bulk to 2D Systems II (Chair: F. Setzpfandt)**  
D. Gerace (11.00 - 11.35)  
C. Cojocaru (11.35 - 12.10)  
R. Le Dantec (12.10 - 12.45)

- 12.45**            **Lunch**
- 14.00**            **Tailored Disorder II (Chair: S. Gigan)**  
M. Abbarchi (14.00 - 14.35)  
P. Yazhgur (14.35 - 15.10)  
L. Schertel (15.10 - 15.30)
- 15.30**            **Coffee Break**
- 16.10**            **Tailored Disorder III (Chair: S. Skipetrov)**  
L. Dal Negro (16.10 - 16.45)  
R. Sapienza (16.45 - 17.20)  
H. Galinski (17.20 - 17.55)
- 18.00**            **Poster Session**
- 18.50**            **Meeting point ETH Link Bus Stop (see map on last page)**
- 18.54**            **Transfer to ETH Zentrum**
- 19.30**            **Dinner**

*Friday 31 January 2020*

- 09.00**            **From Bulk to 2D Systems III (Chair: M. Scalora)**  
T. Ellenbogen (09.00 - 09.35)  
F. Setzpfandt (09.35 - 09.55)  
N. Cherroret (09.55 - 10.15)
- 10.15**            **Coffee Break**
- 10.45**            **Spatial, Temporal and Quantum Control II (Chair: R. Sapienza)**  
M. Chekhova (10.45 - 11.20)  
P. Goldner (11.20 - 11.55)  
N. Strkalj (11.55 - 12.15)
- 12.15**            **Closing**
- 14.15**            **Lab Tour II**

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## Tailored Disorder I (*Chair: R. Savo*)

### Second Harmonic Generation in the Regime of Anderson Localization

**Sergey E. Skipetrov**<sup>1\*</sup>

<sup>1</sup> Univ. Grenoble Alpes, CNRS, LPMMC, 38000 Grenoble, France

We compute the average intensity of the second optical harmonics generated by a strongly scattering random medium in which the fundamental wave experiences Anderson localization. Similar to the previous work [1] in which the problem was treated for a weakly disordered medium and the diffusion approximation was applied, we assume that the locally generated second-harmonic signal is proportional to the square of the fundamental-wave intensity, and that the phase matching is random. Our

calculation is based on the recent results for the intensity distribution inside a strongly disordered medium [2]. Second harmonic generation at a mobility edge is also discussed. Our results may be of practical interest for future nonlinear optical device exploiting strong disorder to increase the time of interaction between light and matter and thereby enhance the efficiency of nonlinear optical phenomena.

[1] Makeev, E.V. & Skipetrov, S.E., Opt. Commun. 224(1-3), 139 (2003)

[2] Skipetrov, S.E. & Sokolov, I.M. arXiv:1907.05299 (2019)

### Photon echoes in disordered media

**Romain Pierrat**<sup>1\*</sup>, **Rémi Carminati**<sup>1</sup>, **Jean-Louis Le Gouët**<sup>2</sup>

<sup>1</sup> ESPCI Paris, Université PSL, CNRS, Institut Langevin, 1 rue Jussieu, 75005 Paris, France

<sup>2</sup> Laboratoire Aimé Cotton, CNRS, Université Paris-Sud, ENS Cachan, Université Paris-Saclay, 91405 Orsay Cedex, France

Photon echoes are a non-linear light-matter interaction process that belongs to the four-wave mixing class such as CARS. There are routinely used for the optical dipole lifetime T<sub>2</sub> measurement in samples of high optical

quality. Substituting cheap and rough powders to these high-quality crystals would thus be of considerable practical interest. Moreover, the successful observation of photon echoes in rare-earth-ion-doped powders calls for a better understanding of the multiply scattered echo signal [1].

The goal of this work is to present a multiple scattering theory of photon echoes generation in strongly disordered powders based on a diagrammatic approach. In particular, analytical expressions for the average echo

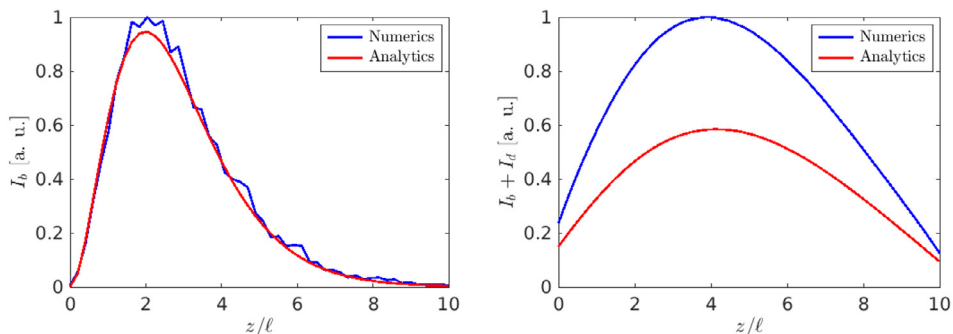


Figure 1. Ballistic (left) and total (right) intensity of the echo signal as a function of the normalized depth in a slab geometry illuminated by a plane-wave at normal incidence.  $\ell$  is the scattering mean-free path. The analytical model for the total intensity has a limited accuracy since it is derived in a large scale limit which explains its discrepancy compared to the numerics.

signal as well as for its correlation with the incident beam are derived and compared with ab-initio numerical simulations [2]. Figure 1 shows the typical behavior of echo signal in a slab geometry as a function of the depth. Since an important enhancement is observed compared to a homogeneous

medium, this work might also open the way to applications in energy conservation.

[1] Beaudoux, F., Ferrier, A., Guillot Noël, O., Chanelière, T., Le Gouët, J.-L. & Goldner, P. *Opt. Express* 19, 15236 (2011).

[2] Pierrat, R., Carminati, R. & Le Gouët, J.-L. *Phys. Rev. A* 97, 063816 (2018).

## Relaxing Phase-matching With Segmented Composite Approaches

**Germano Montemezzani<sup>1\*</sup>, Mouhamad Al-Mahmoud<sup>2</sup>, Virginie Coda<sup>1</sup>, Andon A. Rangelov<sup>2</sup>**

<sup>1</sup> Université de Lorraine, CentraleSupélec, LMOPS, F-57000 Metz, France

<sup>2</sup> Department of Physics, Sofia University, James Bourchier 5 Blvd., 1164 Sofia, Bulgaria

It is well known that phase-matching is crucial for any nonlinear-optical frequency

conversion processes in thick materials. The necessity to fulfill the phase matching condition generally limits the conversion to a narrow wavelength and temperature range, unsuitable to accommodate the spectrum of ultrashort pulses or widely tunable laser sources. Increasing the robustness of the process by relaxing the phase matching conditions is therefore crucial in such cases. The use of very short crystals increases

the bandwidth but suffers of a tradeoff with the conversion efficiency. While one of the alternative techniques in this context involves an adiabatic evolution of the phase matching condition along the propagation direction, here we will discuss an approach based on properly segmented crystals. This technique is inspired by similar robust schemes used in quantum dynamics of a two-level system (e.g. in nuclear magnetic resonance) to prepare a given quantum state in a fault-tolerant way [1]. The idea is to segment the nonlinear material in regions of different thickness and mutually reversed nonlinearity in such a way as to optimize the nonlinear conversion robustly over a wide range of parameter space. Specifically, the robustness is given with respect to the phase mismatch parameter  $\Delta k$  and the variation of the pump intensity [2]. This technique

can be easily combined with conventional quasi-phase matching (QPM) by domain reversal. It can be applied to sum frequency generation (SFG) [2], second-harmonic generation (SHG) [3] or other nonlinear optical frequency conversion processes such as optical parametric amplification (OPA) [4]. In the presentation the background for the composite technique will be elucidated and few recent examples will be given.

[1] Levitt, M. H., *Progr. Nucl. Magn. Reson. Spectrosc.*, 18, 61. (1986).

[2] Rangelov, A. A., Vitanov, N. V., Montemezzani, G., *Opt. Lett.*, 39(10), 2959. (2014).

[3] Ehrlich, Y., Rangelov, A. A., Montemezzani, G., Suchowski, H., *Opt. Lett.*, 44(15), 3837. (2019).

[4] Al-Mahmoud, M., Coda, V., Rangelov, A. A., Montemezzani, G., (to be submitted).

## From Caustic Photonic Lattices to Disordered Caustic Networks

**Cornelia Denz<sup>1\*</sup>, Alessandro Zannotti<sup>1</sup>, Daniel Ehrmanntraut<sup>1</sup>, Philip Menz<sup>1</sup>**

<sup>1</sup> Institute of Applied Physics and Center for Nonlinear Science (CeNoS), University of Muenster, Germany

Caustic light is key to many actual advances in structured light and random waves. On the one hand, tailored caustic beams serve as fabricating light for (nonlinear) material processing, transfer complex momentum flows for

advanced optical trapping, and enable novel high-resolution imaging methods. On the other hand, networks of caustics appear as high-intensity ramifications by focusing random light rays. This linear focusing, caused by strong wave front aberrations and denoted as branched flow, yields waves with extreme amplitudes – so called rogue waves, originally studied in oceanography. Optics has proven to be an excellent testbed to

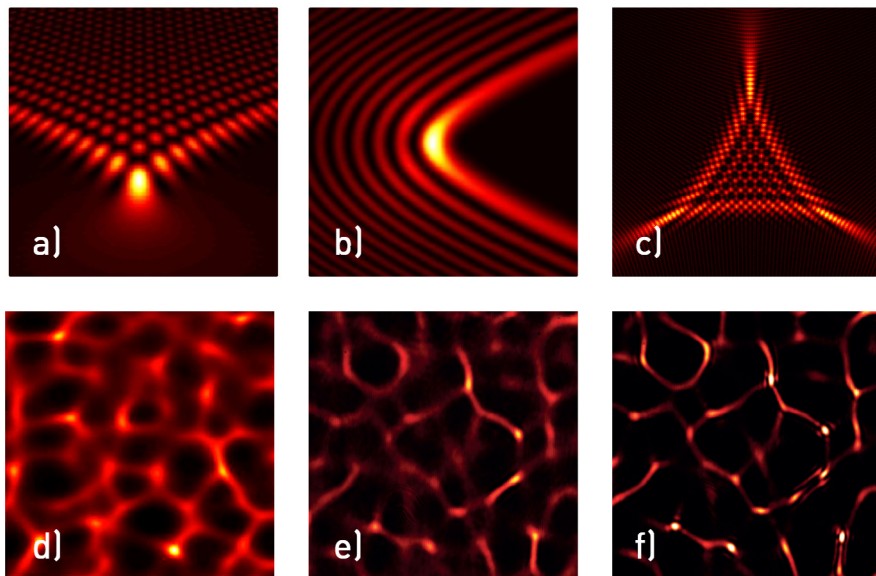


Figure 1. a)-c):Transverse optical caustics. a) Pearcey caustic b) swallowtail caustic, c) elliptic umbilic caustic, d)-e) Propagation of Gaussian random phase fields results in caustic networks of different sharpness. Phase hubs are d)  $\Delta\Phi$  e)  $2\Delta\Phi$  and f)  $4\Delta\Phi$ .



investigate different linear and nonlinear mechanisms as modulation instability for the formation of rogue waves as spatio-temporal wave phenomena originating from optical disorder.

In our contribution, we compare free space propagation of caustic structures [1,2] with propagation in a nonlinear refractive index material and demonstrate wave guiding as well as soliton formation by higher-order caustics [2]. We show that free-space branched flows of light caused by wave front distortions in form of correlated Gaussian random phase fields form caustic networks with controllable extension and sharpness.

This focusing is enhanced by nonlinear refractive index structures. Beyond propagating in homogeneous media, we fabricate tailored two-dimensional disordered photonic lattices in such a nonlinear medium to investigate the interplay of linear focusing and scattering. We find optimal conditions for enhanced focusing of waves with extreme intensities by controlling the size and strength of the disordered photonic refractive index structure.

With these tools in hand, we are able to link different mechanisms of rogue wave formation that are commonly studied separately and discuss their interplay. Our work demonstrates that non-linear focusing can enhance the formation of rogue waves, thereby introducing an

optical platform that allows exploring rogue waves beyond the optical realization and allows new insights into general spatio-temporal wave dynamics.

[1] Zannotti, A., Diebel, F., Denz, C. Dynamics of the optical swallowtail catastrophe. *Optica*, 1157-1162 (2017).

[2] Zannotti, A., Rüschenbaum, M., Denz, C. Pearcey solitons in curved nonlinear photonic caustic lattices. *J. Opt.* 19, 094001 (2017).

# Spatial, Temporal and Quantum Control I

*(Chair: M. Celebrano)*

## Light in Complex Media: From Imaging to Quantum Information Processing

**Sylvain Gigan**<sup>1\*</sup>

<sup>1</sup> Sorbonne Université

Light propagation in complex media is an extremely challenging problem in optics, due to the multiple scattering of light. It is also an important issue in biomedical imaging, to develop optical methods able to penetrate tissues. I will show how wavefront shaping has, in the last decade, revolutionized the field and allowed diffraction-limited imaging through or in complex media [1]. Beyond imaging, it has been realized that complex media in combination with wavefront shaping can be used for many other functions. In my talk, I will illustrate a emerging application: quantum information. I will show

how a complex medium, here a multimode fiber, can be used to emulate linear transformations, that we can subsequently use as a platform to propagate of indistinguishable photons pairs. I will show how this paradigm can be used for information processing, for emulating a coherent absorption experiment, and discuss its prospects for scalable photonic quantum computing [2].

[1] S. Rotter, S. Gigan. Rev. Mod. Phys. 89, 015005 (2017)

[2] S. Leedumrongwatthanakun, L. Innocenti, H. Defienne, T. Juffmann, A. Ferraro, M. Paternostro, S. Gigan. arXiv:1902.10678 [quant-ph]

## Microcombs Based on Laser Cavity Solitons

**Alessia Pasquazi**<sup>1\*</sup>, **Maxwell Rowley**<sup>1</sup>, **Pierre-Henry Hazard**<sup>1</sup>, **Hualong Bao**<sup>1</sup>, **Luana Olivieri**<sup>1</sup>, **Antonio Cutrona**<sup>1</sup>, **Juan Sebastian Toterogongora**<sup>1</sup>, **Sai T. Chu**<sup>2</sup>, **Brent E. Little**<sup>3</sup>, **Roberto Morandotti**<sup>4</sup>, **Gian-Luca Oppo**<sup>5</sup>, **David J. Moss**<sup>5</sup>, **Marco Peccianti**<sup>1</sup>

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<sup>3</sup> Xi'an Institute of Optics and Precision Mechanics, Chinese Academy of Science, Xi'an, China

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<sup>5</sup> SUPA, Department of Physics, University of Strathclyde, Glasgow, United Kingdom

Dissipative solitons are self-confined pulses which appear in driven and lossy systems when the phase dispersion is balanced by the nonlinear phase-shift. Temporal cavity-solitons belong to this class and have been largely studied in a 'driven' configuration, where an external pumping source is resonantly coupled in the nonlinear micro-resonator to sustain and excite the solitary pulses. More recently, we demonstrated that it is possible to generate localised pulses in a configuration where the micro-cavity is inserted in a fiber laser loop. We reported the observation of laser cavity-solitons [1], which have previously attracted large attention especially in spatial configurations. By merging their properties with the physics of both micro-resonators and multi-mode systems, this scheme represents a fundamentally new paradigm for the generation, stabilisation and control of solitary optical pulses in micro-cavities.

In general, laser cavity-solitons are a highly efficient class of cavity-solitons because they are intrinsically background-free. This is in stark contrast to cavity-solitons obtained in nonlinear Kerr cavity driven by an external source and described by the well-known Lugiato-Lefever equation. Currently, these self-localised waves form on top of

a strong background of radiation, usually containing 95% of the total power for bright configurations [2].

Our laser cavity-solitons cover a spectral bandwidth exceeding 50 nm and are induced with average powers more than one order of magnitude lower than those typically used in state-of-the-art soliton micro-combs [1]. Very importantly, in stark contrast to temporal cavity-solitons based Lugiato-Lefever systems, our bright laser cavity-solitons are background-free, and we achieve a mode-efficiency [2] above 75%, compared to typical 1% - 5% for bright solitons realised with standard approaches. Moreover, we can affect the soliton repetition-rate with a simple approach. The free-spectral range of the fiber cavity can be affected by a delay line that modifies the fibre cavity length and, hence, the mode-spacing. In turn this tunes the position of the mode of the system and the repetition rate of the micro-comb.

In this presentation, we will discuss the range of existence of Turing patterns and solitons and possible approaches to their generation and control in our system.

[1] H. Bao, et al. Laser Cavity-Soliton Microcombs. *Nat. Photonics* 13, 384 [2019].

[2] X. Xue, et al. Microresonator Kerr frequency combs with high conversion efficiency. *Laser Photonics*.

# Tailoring Spatiotemporal Nonlinearities in Multimode Fibers for Spectral and Spatial Shaping

**Uğur Teğin<sup>1,2\*</sup>, Babak Rahmani<sup>2</sup>, Eirini Kakkava<sup>1</sup>, Navid Borhani<sup>1</sup>, Christophe Moser<sup>2</sup>, Demetri Psaltis<sup>1</sup>**

<sup>1</sup> Optics Laboratory, École Polytechnique Fédérale de Lausanne, Lausanne, Switzerland

<sup>2</sup> Laboratory of Applied Photonics Devices, École Polytechnique Fédérale de Lausanne, Lausanne, Switzerland

Single-mode optical fibers have been used for various applications such as fiber lasers, optical imaging and nonlinear optics. Recently multimode fibers started to attract attention for the aforementioned applications by including the spatial degree of freedom. Specifically, in graded-index multimode fibers (GIMFs), ultrashort pulse propagation with complex modal interactions revealed new nonlinear dynamics for beam shaping, frequency conversion and ultrashort pulse generation [1,2]. Here, we present control of multimode nonlinearities with a machine learning approach for single-pass pulse propagation and generation of high-quality beam shapes by tailoring nonlinearities in spatiotemporal mode-locked fiber lasers. To understand and control the single-pass spatiotemporal nonlinear propagation in GIMF, we experimentally generate datasets by changing the initial modal excitation of the test fiber (GIMF with  $62.5\mu\text{m}$  core diameter) and record the spectral changes in for the peak powers capable to trigger cascaded Raman scattering and supercontinuum

generations. These collected results are employed to train deep neural networks to understand nonlinear propagation which later used for controlling the frequency generation [3].

So far, temporally Gaussian-shaped (dissipative soliton) pulses are generated in spatiotemporally mode-locked lasers with low beam quality. In our study, we aim to generate Kerr induced self-beam cleaning while spatiotemporal mode-locking achieved. To reach the experimentally reported power threshold of the self-beam cleaning we generated amplifier soliton pulses for the first time in a spatiotemporally mode-locked laser. Self-similar pulse propagation causes less dispersion than the total cavity thus after the gain fiber pulses with a short duration can reach high peak power to trigger self-beam cleaning in GIMF. We numerically and experimentally achieved sub-200 fs pulses with M2-value  $< 1.4$  from a multimode laser cavity [4].

In conclusion, we have presented a novel method to understand and control single-pass spatiotemporal nonlinearities in GIMFs with deep neural networks. With our laser studies, we have presented a way to generate high-quality output beams from a multimode fiber laser cavity by pulse shaping. Overall, the present approaches show paths to understand and tune complex nonlinear propagation in multimode fibers

and can be implemented for other spatio-temporal nonlinear effects.

[1] Krupa, K., Tonello, A., Shalaby, B. M., Fabert, M., Barthélémy, & Couderc, V. *Nature Photonics*, 11(4), 237. (2017).

[2] Teđin, U., & Ortaç, B. *Scientific Reports*, 8(1), 12470. (2018).

[3] Teđin, U., Rahmani, B., Kakkava, E., Borhani, N., Moser, C., & Psaltis, D. *arXiv:1909.10561*. (2019).

[4] Teđin, U., Kakkava, E., Rahmani, B., Psaltis, D., & Moser, C. *arXiv:1904.08456*. (2019). Accepted to *Optica*.

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# From Bulk to 2D Systems I

*(Chair: R.Grange)*

## Electrodynamics of Conductive Oxides: Induced linear dielectric anisotropy, harmonic generation, and retrieval of the effective dielectric constant

**Michael Scalora<sup>1\*</sup>, Laura Rodriguez Sune<sup>2</sup>, Maria Antonietta Vincenti<sup>3</sup>, Domenico de Ceglia<sup>4</sup>, Zachary Coppens<sup>5</sup>, Neset Akozbek<sup>5</sup>, Jose Trull<sup>2</sup>, Crina Cojocaru<sup>2</sup>**

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<sup>2</sup> Universitat Politècnica de Catalunya, Physics Department, Rambla Sant Nebridi 22, 08222 Terrassa, Barcelona, Spain

<sup>3</sup> Department of Information Engineering – University of Brescia, Via Branze 38 25123 Brescia, Italy

<sup>4</sup> Department of Information Engineering – University of Padova, Via Gradenigo 6/a 35131 Padova, Italy

We undertake a detailed theoretical study of electromagnetic pulse propagation effects in an indium tin oxide nanolayer in the linear and nonlinear regimes. We use the constitutive relations to retrieve the effective dielectric constant and show that in the linear regime nonlocal effects induce a robust, anisotropic, dielectric response hitherto unknown: longitudinal and transverse effective dielectric functions are strongly and differently modulated along the propagation direction, and

display different epsilon-near-zero crossing points. In the nonlinear regime, at high intensities hot carriers are the dominant mechanism for third harmonic generation, induce a redshift of the plasma frequency and a corresponding translation of the effective nonlinear dispersion curves. The shift can then be used to accurately predict and quantify nonlinear refractive index changes as a function of incident peak power density. Our results suggest that large, nonlinear refractive index changes can occur without the need for epsilon-near-zero modes to couple with plasmonic resonators. We also predict the presence of additional absorption resonances that arise from longitudinal oscillations of the free electron gas, which give way to corresponding resonances in the second and third harmonic spectra, and the onset of optical bistability at sufficiently large intensities. The key to unraveling the basic physical mechanisms that play a fundamental role in the dynamics is a realistic propagation model able to extract accurate, basic information from the interaction as it occurs.

# Ultrafast Nonlinear Optical Response of Two-dimensional Materials

**Andrea Marini<sup>1\*</sup>**

<sup>1</sup> Department of Physical and Chemical Sciences, University of L'Aquila, Via Vetoio, 67100 L'Aquila, Italy

Optical nonlinearity in photonic materials enables a large number of applications such as frequency conversion, all-optical signal processing, and non-classical sources of light. The speed of solid-state electronic devices, determined by the temporal dynamics of charge carriers, could potentially reach unprecedented petahertz frequencies through direct manipulation by optical fields, consisting in a million-fold increase from state-of-the-art technology. In graphene, charge carrier manipulation is facilitated by exceptionally strong coupling to optical fields, from which stems an important back-action of photoexcited carriers. Here we report the instantaneous response of graphene to ultrafast optical fields, elucidating the role of hot carriers on sub-100 fs timescales. The observed nonlinear response and its dependence on interaction time and field polarization reveal the back-action of hot carriers over timescales commensurate with the optical

field. An intuitive picture is given for the carrier trajectories in response to the optical-field polarization state. We note that the peculiar interplay between optical fields and charge carriers in graphene may also apply to surface states in topological insulators with similar Dirac cone dispersion relations. Furthermore, we report harmonic generation and saturable absorption in graphene, discussing free-carrier generation and their ultrafast temporal dynamics in the atomically thin material. We further discuss the nonlinear optical properties of transition metal dichalcogenides, illustrating cavity-enhanced second-harmonic generation and parametric down-conversion and demonstrating that phase-matching free operation can be achieved in photonic micro-cavities embedding two-dimensional semiconductors as nonlinear optical media. Our results are promising for the development of integrated optical parametric oscillators and micro-sources of entangled photons.

[1] M. Baudisch et al., Nat. Commun. 9, 1018 (2018).

[2] A. Ciattoni et al., Light: Sci. & Appl, 7, 5 (2018).

## Second-harmonic Generation in Multilayer Hexagonal Boron Nitride

Sejeong Kim<sup>1</sup>, Johannes E. Fröch<sup>1</sup>, Augustine Gardner<sup>1</sup>, Chi Li<sup>1</sup>, Igor Aharonovich<sup>1</sup>, Alexander S. Solntsev<sup>1\*</sup>

<sup>1</sup>University of Technology Sydney, Ultimo, New South Wales, 2007, Australia

We report second-harmonic generation (SHG) from thick hexagonal boron nitride (hBN) flakes with approximately 109-111 layers. The resulting effective second-order susceptibility is similar to previously reported few-layer experiments [1], despite the expected drop-off with an increasing number of layers. This confirms that thick hBN flakes

can serve as a platform for nonlinear optics [2], which is useful because thick flakes are easy to exfoliate while retaining a large flake size. We also show spatial second-harmonic maps revealing that SHG remains a useful tool for the characterization of the layer structure even in the case of a large number of layers.

[1] Y. Li, Y. Rao, K. F. Mak, Y. You, S. Wang, C. R. Dean, and T. F. Heinz, *Nano Letters*, 13, 3329-3333 (2013).

[2] A. Autere, H. Jussila, Y. Dai, Y. Wang, H. Lipsanen, and Z. Sun, "Nonlinear Optics with 2D Layered Materials," *Adv. Mater.*, 30, 1705963 (2018).

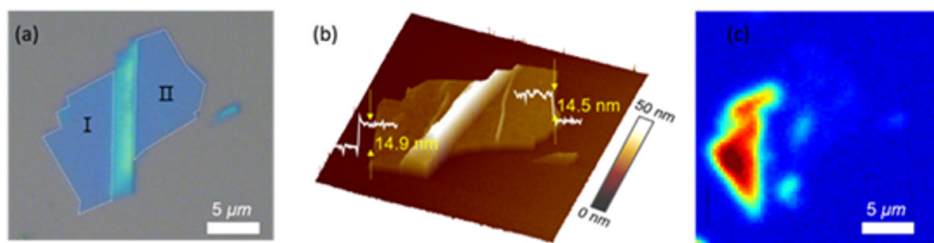


Figure 1. Spatial SHG mapping. (a) Optical image of hBN flake with areas of different layer number labeled as I and II. (b) AFM map of the flake. (c) Spatial SHG intensity mapping across hBN sample. Region I shows strong SHG signal while the signal from region II is negligible. This is because hBN layers are stacked in 2H order, where the sites for boron and nitrogen are alternating, and the inversion symmetry is only broken for an odd number of layers.



# Engineering Plasmonic Nanoantennas for Efficient Nonlinear Photon Conversion at the Nanoscale

**Michele Celebrano<sup>1\*</sup>, Andrea Locatelli<sup>2</sup>, Lavinia Ghirardini<sup>1</sup>, Attilio Zilli<sup>1</sup>, Giovanni Pellegrini<sup>1</sup>, Paolo Biagioni<sup>1</sup>, Xiaofei Wu<sup>3</sup>, Swen Grossman<sup>3</sup>, Luca Carletti<sup>2</sup>, Costantino De Angelis<sup>2</sup>, Lamberto Duò<sup>1</sup>, Bert Hecht<sup>3</sup>, Marco Finazzi<sup>1</sup>**

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<sup>2</sup> University of Brescia, Department of Information Engineering, 25123 Brescia, Italy

<sup>3</sup> University of Würzburg, Nano-Optics & Biophotonics Group - Department of Physics - Experimental Physics 5, 97074 Würzburg, Germany

The optimization of nonlinear optical processes at the nanoscale is a key challenge in state of the art nanoscience, given their potential key role in the development of building blocks for quantum-optical networks as well as in molecular sensing. In this context, plasmon-enhanced nonlinear effects are gaining ground thanks to the development of innovative nanoantenna designs and hybrid nanostructures. [1-2]

I will describe our approach to attain plasmon-enhanced second harmonic generation (SHG) by means of broken-symmetry nanoantenna working in the near infrared region of the electromagnetic spectrum. This nanoantenna behaves like a strongly coherent nanoscale light source, featuring extremely low multiphoton luminescence in the visible range and a marked third

harmonic generation (THG) along with an intense SHG [3].

Due to the relatively low nonlinear conversion efficiencies in these nanoscale systems, a widely accepted approach consists in treating different order nonlinear processes in plasmonic nanoantennas, such as, for instance, THG and SHG, as independent phenomena. We show that this paradigm can dramatically fail in these highly non-centrosymmetric nanoantennas, finding evidence of a SHG-mediated cascaded effect in THG (see Figure).

This mechanism can be unveiled thanks to

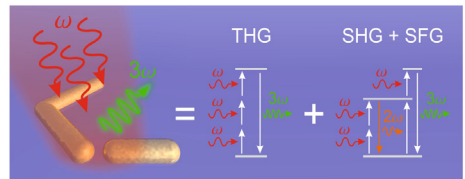


Figure 1. Pictorial description of the cascaded effect in broken-symmetry plasmonic nanoantennas.

the commensurate emission yields of SHG and THG processes in this nanostructure. We have disentangled this cascading effect from bulk  $X^{(3)}$ -mediated THG thanks to the unique nonlinear emission fingerprint of these processes, which can be identified through the analysis of the overall emission polarization state [4]. This result demonstrates the possibility of tailoring the coupling efficiency

between nonlinear processes in plasmonic nanoantennas and represents a crucial step towards the realization of nanoscale photon conversion and manipulation at room temperature, opening new possibilities in quantum optics and nonlinear optical sensing.

[1] Kauranen, M., Zayats, A. V. *Nature Photonics* 6 737-748 (2012).

[2] Butet, J. et al. *ACS Nano*, 9, 10545–10562 (2015).

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## From Bulk to 2D Systems II (*Chair: F. Setzpfandt*)

### Photonic Crystal Cavities for Enhanced Nonlinear and Quantum Optics

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Photonic crystal (PC) cavities allow to efficiently confine the electromagnetic field, achieving very long photon lifetimes (large Q-factor) in the smallest confinement volumes (diffraction-limited  $V$ ). Hence, any nonlinear process scaling with the  $Q/V$  ratio may be strongly enhanced in these systems. As an example, simultaneous second- (SH) and third-harmonic (TH) generation has been shown to occur under continuous wave excitation and unprecedented conversion efficiency from crystalline silicon PC cavities [1]. Similar results have later been obtained in different material platforms, such as GaN [2]. In order to further enhance the nonlinear generation processes, it would be highly desirable to design PC cavities with multiply resonant conditions, i.e. possessing cavity modes at fundamental frequency and at higher harmonics simultaneously. In practice, it has proven very difficult to engineer PC cavities fulfilling multiply resonant conditions. Here I will report on a recent strategy allowing to design doubly resonant PC cavities for enhanced SH generation [3]. The protocol is based on engineering a PC lattice supporting a photonic gap at

fundamental frequency, and a bound state in the continuum (BIC) at SH, thus allowing to relax the constraint of engineering a SH photonic band gap in order to achieve a doubly resonant PC cavity. Preliminary experimental results on GaN material platform will be reported, showing an unprecedented enhancement of SH generation as compared to the non-resonant case in [2]. Finally, the relevance of these results for a number of proposed applications in nonlinear and quantum optics will be discussed, such as low-power nonlinear optics in hybrid material nanophotonics [4], and the quest to ultimately achieve the regime of single-photon nonlinearities in passive materials exploiting doubly resonant cavities [5].

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# Harmonic Generation in the Opaque Region of GaAs: The Role of the Surface and Magnetic Nonlinearities

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Theoretical predictions show that if a beam is incident onto a generic nonlinear crystal, three components of harmonic beams are generated: one in reflection and two in transmission. One of the transmitted components travels with the group velocity and the absorption according to material dispersion at the harmonic wavelength. The second one, known as “phase locked” (PL) component, is trapped and dragged along with the pump. This PL harmonic component, having its origin in the inhomogeneous solution of the nonlinear equation, is generated very close to the surface and it propagates into the material with the same phase velocity and absorption coefficient as the fundamental beam, regardless of the dispersion at the harmonic frequency. These two harmonic components can only be distinguished when they are induced with short pulses and far from the usual phase-matching condition [1]. These predictions were experimentally validated first in transparent materials [2] and later it was demonstrated that phase locking leads to the inhibition of linear absorption of the harmonics in absorptive materials [3,4].

In spite of these studies, there is still a lack of fundamental knowledge regarding the generation of harmonics near the surface under these unusual conditions.

In this work we study in detail the generation of the PL SH and TH in the opaque region of GaAs, both in reflection and transmission. We show that measurement of the angular and polarization dependence of the observed harmonic components allows one to infer the different nonlinear mechanisms that trigger these processes, including not only the bulk nonlinearity, but also surface and magnetic Lorentz contributions, which usually are either hidden by the bulk contributions or assumed to be negligible. The experimental results are compared with a detailed numerical model that takes into account these different effects. Our results suggest that the intensity of the SH signal generated by the surface can be more intense than the signal generated by the bulk. Although the PL harmonic generation has a very low efficiency, these findings have significant repercussions and are consequential in nanoscale systems, which are usually investigated using only dispersionless bulk nonlinearities, with near-complete disregard of surface and magnetic contributions and their microscopic origins.

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## LiNbO<sub>3</sub> and BiFeO<sub>3</sub> Harmonic Nanoparticles

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Harmonic nanoparticles are inorganic oxide nanocrystals displaying a very rich nonlinear response because of their noncentrosymmetric crystal structure [1]. They have been proposed as new types of exogenous contrast agents for multiphoton imaging applications [2]. In this talk, we will present some of the routes concerning the preparation of BiFeO<sub>3</sub> and LiNbO<sub>3</sub> harmonic nanoparticles, among the most promising nanomaterials with high harmonic generation efficiency and low cytotoxicity. Recently, we used a low-cost precipitation technique that allows the preparation of almost phase-pure BiFeO<sub>3</sub>. For LiNbO<sub>3</sub> a solvothermal approach was developed and well-crystallized and phase-pure nanoparticles were obtained with low polydispersity.

We will also discuss about the nonlinear optical characterization of harmonic nanoparticles using second harmonic scattering (SHS). SHS is based on ensemble

measurements with colloidal suspensions, and this technique is better suited than multiphoton microscopy to retrieve quantitative parameters such as nonlinear optical susceptibilities. Thanks to the use of a tunable femtosecond laser, second harmonic spectroscopy on both LiNbO<sub>3</sub> and BiFeO<sub>3</sub> nanoparticles was performed in the 700 – 1300 nm range, showing a well-defined resonance for BiFeO<sub>3</sub> [3].

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## Tailored Disorder II (*Chair: S. Gigan*)

# Solid-state Dewetting of Si(Ge)-based Complex Nano-architectures and their Applications in Photonics

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Dewetting is a ubiquitous phenomenon in nature: many different thin films of organic and inorganic substances share this shape instability driven by surface tension and mass transport. This spontaneous phenomenon leads a thin film to break and drip in isolated islands. Here, I will address two distinct cases of solid-state dewetting: 1) templated dewetting of silicon and 2) spontaneous dewetting of silicon-germanium.

Templated solid-state dewetting can be used to frame complex nanoarchitectures, nanowires (up to 0.75 mm long) and connected circuits of monocrystalline silicon on insulator with unprecedented precision and reproducibility over large scales [1]. Phase-field simulations quantitatively benchmark

the experimental results revealing the dominant role of surface diffusion as a driving force for dewetting and the role of faceting in stabilizing the nanostructures. I will discuss the use of these ordered structures as dielectric Mie resonators for visible and NIR light manipulation [2,3].

Spontaneous dewetting of thick SiGe layers leads to the onset of spinodal-like structures as accounted for by the features of Minkowski-functionals and evolution of Betti numbers [4]. The formation of these disordered structures is interpreted in the framework of the Cahn-Hilliard-Cook theory of phase separation in analogy with spinodal dewetting of polymers and liquid-metals. I will discuss the possibility to exploit this bottom-up, self-assembly method to form hyper-uniform, dielectric metasurfaces at visible and near-infrared frequencies [5] over ultra-large scales.

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# Amorphous Photonic Band Gap Materials Fabricated by Colloidal Self-assembly

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Most of photonic band gap (PBG) materials fabricated and studied today are periodic nanostructures. Recently discovered well-engineered disordered photonic materials not only are argued to be more convenient in fabrication, but also offer isotropy, allowing, for example, create free-form wave guides, displays or non-iridescent ultra-resistant pigments. Surprisingly, the fabrication of such amorphous photonic materials is completely dominated by the top-down approach and suffer from its severe limitations (cost, time scale, defects, resolution, etc.).

To overcome this bottleneck, I will present some concepts of all-dielectric disordered photonic materials, which can be potentially fabricated by colloidal self-assembly. Using microfluidic-assisted assembly of droplets, we experimentally produce disordered 2D

materials with very low level of long-range density fluctuations - hyperuniformity. Hyperuniformity has been shown as a guideline for creating disordered PBG materials and fabrication of hyperuniform materials is often considered as a first step to create amorphous PBG materials. By drying these 2D emulsions, we get a material with even lower long-range density fluctuations and stronger short-range order, which finally open a PBG for TE (electric field in plane) polarization. The obtained results suggest that photonic foams are potentially excellent candidate for disordered 3D PBG materials. Finally, I will present some results concerning photonic properties of small disordered nanoparticle assemblies (photonic balls), which are useful to create structural colors.

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# Scattering in Advanced Photonic Glasses: Why Isotropic Structural Color Will Never be Pure!

**Lukas Schertel<sup>1,2\*</sup>, Gianni Jacucci<sup>1</sup>,**

**Lukas Siedentop<sup>2</sup>, Janne-Mieke**

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Colors of materials usually originate from a combination of wavelength dependent absorption and scattering. Controlling the color without the use of absorbing dyes is of practical interest, not only because of undesired bleaching but also regarding the use of biopolymer based environmental-friendly materials. Color control without dyes can be achieved by tuning the material's scattering properties in controlling size, spatial arrangement and shape of scatters.

Periodically ordered colloidal structures such as opals, where iridescent colors arise from Bragg diffraction, are well established. Their colors are, however, strongly dependent on the angle of view because of the periodicity of interparticle distance giving rise to narrow diffraction maxima. For certain applications in coatings, paints, as well as cosmetics, this angular dependency is undesirable. The disorder counterpart, photonic glasses (PGs), have become of interest as these possess isotropic properties and can produce angle-independent structural colors. Still, a limited range of colors was produced so far by self-assembly

of monodisperse spheres to PG as well as strong color purity has not been achieved in these materials.

A recently developed quantitative scattering model [1] allows us to fully capture the observed isotropic structural colors of PGs produced so far. The model predicts the reflectance spectra and the expected sample color solely from the scatter size, refractive index, filling fraction, structural order and sample thickness [2]. To compare the reflectance spectra predicted by the model we prepare optical thin films of PGs by random aggregation of non-absorbing, monodisperse colloidal polystyrene (PS) spheres and measure their spectral reflectance over the entire visible spectrum. By varying the colloidal scatter size we can obtain blue, green, red and purple colors repeatably, as predicted by the model. In addition, changing the effective sample thickness enables us to tune between vivid colors and white broadband reflectance. The good agreement between the model and experiment for PS and TiO<sub>2</sub> PGs enables us to map the parameter space and to predict the range of color hues for different refractive index PGs. Still, for PGs made of assemblies of monodisperse colloids color purity is limited.

Experimentally, more advanced scatter morphologies have been shown to improve the achievable color purity. Thus, we use finite difference time domain simulations to



explore the limitations of color purity and saturation of isotropic structural colors in advanced scatter morphologies such as inverse photonic glasses or hollow sphere glass structures. Clearly, this study points out that high color saturation cannot be combined with isotropy in scattering and appearance, especially for red hues.

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## Tailored Disorder III (*Chair: S. Skipetrov*)

### Engineering Hyperuniformity: Wave Transport & Localization in Nanoparticle Arrays

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The ability to design complex wave fields and electromagnetic interactions in optical structures with aperiodic geometries provides novel opportunities to advance nanophotonics and metamaterials technologies. In particular, recent progress in the theory, fabrication, and characterization of dielectric nanostructures with tailored disorder and hyperuniform geometries establish a novel strategy to enhance the linear and nonlinear optical responses of engineered optical media with unique scattering and wave localization properties. In this talk, I will discuss our recent work on the design and engineering

of hyperuniform and anti-hyperuniform arrays of scattering nanoparticles. Specifically, I will present the experimental demonstration of novel scattering arrays of dielectric nanoparticles, named **isotropic scattering arrays (ISAs)**, with intensity optimized using the collective coordinate control method, which give rise to targeted ring-shaped regions in Fourier space. Enhanced light extraction and directional light emission are demonstrated experimentally on large-scale (1 mm<sup>2</sup>) ISAs arrays fabricated atop LED ceramic converter materials. Finally, using the rigorous dyadic Green's matrix spectral method with electric and magnetic scattering contributions, we describe the multiple scattering and wave localization problems in both hyperuniform and anti-hyperuniform media with unbounded density fluctuations and address their potential for critical device applications to solid-state lighting, lasing, and optical spectroscopy using Si-based optical materials.

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# Nanophotonic Lasers on a Graph

**Riccardo Sapienza**<sup>1\*</sup>

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Random lasing — for which disorder is exploited to enhance stimulated emission — has emerged as a paradigmatic phenomenon of complex lasers [1]. In this talk I will discuss our recent efforts to control laser light emission in unconventional laser architectures, where light flows and get amplified on a random graph, supported by a material nanostructured sub-wavelength networks [2]. The network is composed of interconnected polymer nanofibers that are doped with a laser dye (Rhodamine-6G). Network lasers offer the advantage of strong modal competition for the gain, a property that can be exploited for control and tuning. In particular networks can be modelled with a graph description of Maxwell's equations, and the lasing action by a steady-state ab

initio laser theory (netSALT) [3], which includes nonlinear mode interaction.

We demonstrate lasing from the network, over a wide range of frequencies corresponding to many Anderson localised network modes. I will also discuss how unbalancing the mode competition is an effective strategy for spectral control and signal processing. The complexity of the photonic network lasers brings the opportunity to single-out a lasing frequency out of the spectral haystack by selectively illuminating a very small subset of the network links.

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[2] A nanophotonic laser on a graph, M.Gaio, D. Saxena, J. Bertolotti, D. Pisignano, A. Camposo, R. Sapienza, Nature Comm. 10, 226 (2019)

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# Large-area Disordered Metamaterials

**Henning Galinski**<sup>1\*</sup>

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Today, optical metamaterials and metasurfaces are produced using lithography techniques and typically cannot be scaled above the gram level. In addition, the optical response of metamaterials is often designed for a specific polarization, which makes

them unsuitable for many real-world applications using unpolarized light. In this talk, I will present two examples of our ongoing efforts to use disorder as a design parameter to create scalable optical metamaterials: (i) Network Metamaterials are metamaterials fabricated via dealloying, a versatile wet-etching technique, that allows to nanostructure large areas in a fast and

cost-effective fashion. Despite the lack of both symmetry and long-range order, these networks possess complex light-matter interactions, generating an ensemble of surface plasmon (SP) waves that, due to nano-scaled disorder, are not able to propagate and acquire the character of localized waves. The local subwavelength confinement of light leads to the formation of near-perfect absorbing states, which can be used for plasmon-assisted energy conversion. (ii) Metal induced crystallization in quasi-random metasurfaces is a new design approach to create large-area epsilon near-zero materials. Here, the preferential crystallization of an amorphous semiconductor at the metal/semiconductor interface is used as design

parameter. The induced crystallization generates new epsilon near-zero states corresponding to a hybridized plasmonic mode emerging from selective coupling of light to the Ångstrom-sized crystalline shell of the semiconductor.

I will chart a course how these two approaches can be used in nonlinear optics, e.g. for photonic materials with extremely large optical nonlinearities or second harmonic generation.

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## From Bulk to 2D Systems III (Chair: M. Scalora)

### Nonlinear metasurfaces for THz generation

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The last two decades have witnessed dramatic advances in generation, manipulation and detection of THz radiation that forecast various desirable applications including optical characterization of various materials, biomedical imaging, molecular spectroscopy, and faster wireless communication to give just a few examples. However, despite the intensive efforts to tame the THz band, there is still a substantial lack of simple methods to generate, control and measure THz waves. Recently it was shown that nonlinear metasurfaces can be used to generate broadband THz radiation. Here I will present recent advancements in the ability of nonlinear metasurfaces to generate THz waveforms by demand.

I will show that the ability to engineer the properties of each single nonlinear dipole emitter on the metasurface, provides unprecedented controllability over the spatiotemporal structure of the THz waves that are emitted from the nonlinear metasurfaces [1]. I will demonstrate the direct generation of single-cycle Hermite-Gauss (1,0) wavepackets. Intriguingly, the radiation

from such metasurface shows a single-cycle quadrupole field propagating in free space. This type of single-cycle wavepacket cannot be achieved by standard phase-plate based beam shaping techniques. Moreover, I will show how to generate dispersed [1] or focused few-cycles THz pulses [2] with engineered dispersion and tunable carrier frequency. I will also show that the carrier envelop can be further controlled by the spatial design of the metasurface [3]. Moreover, I will introduce a new waveguide configuration platform [4], which can be used to generate broadband THz radiation directly into the waveguide, and to tune the frequency of the generated THz. These unprecedented control capabilities open the door to develop new and improved THz spectroscopy and imaging tools. In addition, they motivate more fundamental research of new ways to further enhance and control THz generation by nanostructured materials.

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# Lithium Niobate Metasurfaces for Parametric Three-wave Mixing

**Anna Fedotova<sup>1</sup>, Mohammadreza Younesi<sup>1</sup>, Jürgen Sautter<sup>1</sup>, Reinhard Geiss<sup>1</sup>, Thomas Pertsch<sup>1,2</sup>, Isabelle Staude<sup>1</sup>, Frank Setzpfandt<sup>1\*</sup>**

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Lithium niobate (LN) is an attractive material for nonlinear photonics due to its wide transparency range and high second-order nonlinearity. However, almost all applications of LN to date were using bulk LN crystals or waveguides. Although isolated Mie-type nanoresonators in lithium niobate have been shown very recently [1], nonlinear functional metasurfaces based on densely packed arrangements of such nanoresonators have not yet been demonstrated.

In our contribution, we will show experimental results from nonlinear metasurfaces consisting of LN nanocubes realized in a LN thin film on a SiO<sub>2</sub> substrate. Compared to other demonstrations of nonlinear metasurfaces featuring Mie-type resonances [2], LN has a comparatively low refractive index of only

2.1 at the wavelength of 1  $\mu\text{m}$ . Nevertheless, electric dipole (ED) and magnetic dipole (MD) Mie-type resonances could still be observed. We could demonstrate, that the ED and MD resonances enhance the conversion efficiency for second-harmonic generation, that the generated second harmonic is predominantly emitted in the forward direction, and that the specific structure of the nonlinear tensor of LN leads to significantly different dependences of the generated light on the polarization of the excitation for the ED and MD resonances.

With these results, we show that resonant metasurfaces based on lithium niobate are a suitable platform for parametric three-wave mixing. Due to the large transparency range of LN, a wide spectral range from the UV to the mid-IR can be addressed and our work is a step towards using the potential of LN metasurfaces for nonlinear applications.

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# Simulating Quantum Mechanics with Light in Disordered and Nonlinear Materials

**Tamara Bardon-Brun<sup>1</sup>, Dominique Delande<sup>1</sup>, Nicolas Cherroret<sup>1\*</sup>**

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A current exciting question deals with the realization of photonic setups allowing to reproduce phenomena usually encountered in quantum systems. Recent examples include the observations of light superfluidity in nonlinear materials or, in disordered media, of Anderson localization of light, optical Hall effects and even photonic topological insulators.

In this context, an interesting strategy consists in propagating light beams through materials – e.g. photonic waveguide arrays or 2D optical fibers – displaying refractive-index variations in two transverse directions but homogeneous along the optical axis. In the paraxial limit, light then mimics the behavior of a quantum matter wave, the wave equation mapping onto a Schrödinger equation where the optical axis coordinate plays the role of time. In the presence of transverse disorder, this mapping was first exploited to demonstrate 2D Anderson localization of optical wave packets, analogous to what is achieved in cold-atomic systems. In the presence of an index nonlinearity, light can even behave effectively like a quantum

fluid of interacting particles and display, e.g., superfluidity.

In my talk, I will show that nearly paraxial light propagation in dielectric materials can be exploited to achieve optical analogues of two important quantum mechanical phenomena. The first is the *spin Hall effect*, which can be observed with polarized light by using of transverse disorder [1]. The optical spin Hall effect manifests itself by helicity-dependent shifts of beam trajectories, in complete analogy with electrons subjected to spin-orbit interactions. The second phenomenon is the spontaneous emergence of *quasi long-range spatial coherence of light* [2]. This property can be observed in Kerr nonlinear materials, and reproduces the behavior of interacting quantum Bose condensates in low dimensions.

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# Spatial, Temporal and Quantum Control II

(Chair: R. Sapienza)

## Microscale spontaneous parametric down-conversion

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Nanoscale generation of biphotons via spontaneous parametric down-conversion (SPDC) is a challenging goal. At the same time, it offers unique radiative characteristics: the frequency angular spectrum generated is extremely broad and as such it promises a two-photon correlation width, in time and position, narrower than anything observed before. Additionally, it gives an insight into the subwavelength resonances for vacuum fluctuations. However, SPDC is a relatively weak effect, linear in the pump power, and its nanoscale observation is masked by fluorescence.

In our recent works [1,2] we make an important step towards this goal, demonstrating SPDC from an effective length of 1.4 micrometer. Using correlation methods, we retrieve the spectral and polarization properties despite the strong fluorescence background.

We use a 6.8  $\mu\text{m}$  layer of lithium niobate in a geometry where the strongest nonlinear tensor component is fully used but the phase

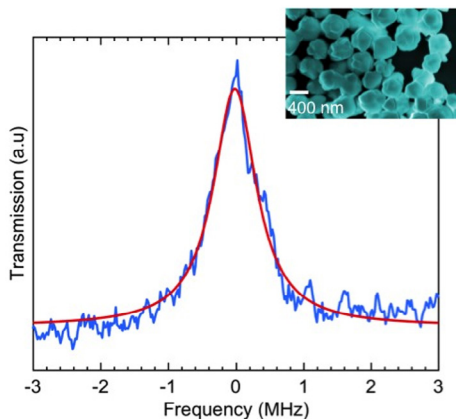


Figure 1. Ultra-narrow optical line in  $\text{Eu}^{3+}:\text{Y}_2\text{O}_3$  nanoparticles.

matching is not satisfied. Photon pairs are generated effectively from a single nonlinear coherence length (1.4-3  $\mu\text{m}$ , depending on the pump wavelength). Under 170 mW CW pumping at 500 nm, we register 1 kHz of photon pairs.

Working in the regime that is free of phase matching results in an ultrabroad frequency angular biphoton spectrum. We measure a frequency spectrum around 150 THz and an angular spectrum of 30 degrees. As a result, two-photon correlations are expected to be subwavelength in space and nearly subcycle



in time. Meanwhile, transverse wavevector and energy are strictly conserved, which leads to the photon pairs displaying an extremely high degree of continuous-variable entanglement. We demonstrate this using stimulated-emission tomography.

The efficiency can be increased by passing to materials with huge nonlinearity, i.e. GaAs, GaP or other semiconductors. The length can then be reduced to the nanoscale, without a reduction in the coincidence rate.

Further developments include: integration into optical chips and nanostructuring the nonlinear material.

[1] C. Okoth, A. Cavanna, T. Santiago-Cruz, and M. V. Chekhova, arXiv:1902.11218v2 [quant-ph].

[2] C. Okoth, E. Kovlakov, F. Bönsel, A. Cavanna, S. Straupe, S. P. Kulik, and M. V. Chekhova, arXiv:1907.09888v1 [quant-ph].

## Rare Earth Doped Crystals for Optical Quantum Technologies

### **Philippe Goldner<sup>1\*</sup>**

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Quantum technologies are developed to overcome classical limits in communication and processing but also in new areas like sensing, imaging and simulations. They will impact all aspects of life by allowing e.g. ultra-secure communications, simulation of complex drug molecules, or new bio-medical imaging techniques.

Rare earth doped crystals have been recently considered for these applications because of their long-lived quantum states, or narrow linewidths, for optical and spin transitions. This unique property in the solid-state, observed at low temperature, make these materials very promising for quantum light-matter interfaces.

Recent results obtained in rare earth doped crystals include quantum teleportation, record-long optical storage and optical or superconducting resonator enhanced interactions.

In this talk, we will review recent results obtained in our group on long optical and spin coherence lifetimes in bulk and nano-structured crystals. These results have been obtained by careful material choice and development combined with rare earth decoupling from noise using clock transitions or dynamical techniques [1-2].

[1] Serrano, D., Karlsson, J., Fossati, A., Ferrier, A., Goldner, P., Nat. Commun. 9, 2127 (2018).

[2] Ortu, A., Tiranov, A., Welinski, S., Fröwis, F., Gisin, N., Ferrier, A., Goldner, P., Afzelius, M., Nat. Mater. 17, 671–675 (2018).

# Probing Ultrathin Ferroelectricity in Perovskite Thin Films Using Nonlinear Optics

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The development of low-energy-consuming electronics based on ferroelectrics is hindered by the loss of net polarization in the ultrathin regime caused by polar discontinuities at the interfaces. So far, either the charge screening of the bottom or the top interface has been engineered to set the polarization domain state and direction. The lack of techniques to disentangle the contributions from each interface renders elusive

the understanding of their joint impact governing the final polarization state. Taking  $\text{PbTiO}_3$  as a model system, we investigate the polarization dynamics during the synthesis and isolate each interface contribution to a thin film polarization using in situ optical second harmonic generation (ISHG) [1,2]. By tailoring the interface chemistries towards a collaborative configuration, we stabilize a polarization enhancement and a robust polarization state in a multilayer architecture. Exploring the uncharted territory of the competition and collaboration of interfaces opens new degrees of freedom for the integration of ferroelectrics in oxide electronics.

[1] De Luca, G., Strkalj, N. et al., Nat. Commun., 8, 1419. (2017).

[2] Strkalj, N., De Luca, G. et al., Phys. Rev. Lett., 123, 147601. (2019).

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## List of Posters

**Bohn Justus** - *University of Exeter*  
Optical Switching of an Epsilon-Near-Zero Plasmon Resonance in Kretschmann Configuration

**Clementi Marco** - *Università degli Studi di Pavia*  
Photonic Crystal Cavities with Comb-like Spectrum for Integrated Nonlinear Optics in Silicon

**Devaud Louisiane** - *Laboratoire Kastler Brossel*  
Eigen-modes of the Time-gated Transmission Matrix in Complex Media for Temporal Energy Control

**Heugel Toni** - *ETH Zurich*  
Parametric Oscillators in the Nonlinear Regime

**Jacucci Gianni** - *University of Cambridge*  
Role of Anisotropy and Refractive Index in Scattering and Whiteness Optimization

**Morandi Andrea** - *ETH Zurich*  
Disordered Lithium Niobate Assemblies for Second Harmonic Generation

**Müller Jolanda** - *ETH Zurich*  
Numerical Analysis of Random Quasi-Phase Matching in Assemblies of Birefringent Materials

**Savo Romolo** - *ETH Zurich*  
Random Quasi-Phase Matching is Disordered Optically Resonant Micro-structures

**Wang Xiyue (Sissi)** - *ETH Zurich*  
Lithium Niobate as a Platform for Integrated Photonic Devices with Complex Geometry

**Zilli Attilio** - *Politecnico di Milano*  
Sum Frequency Generation from Individual AlGaAs Nanocylinders

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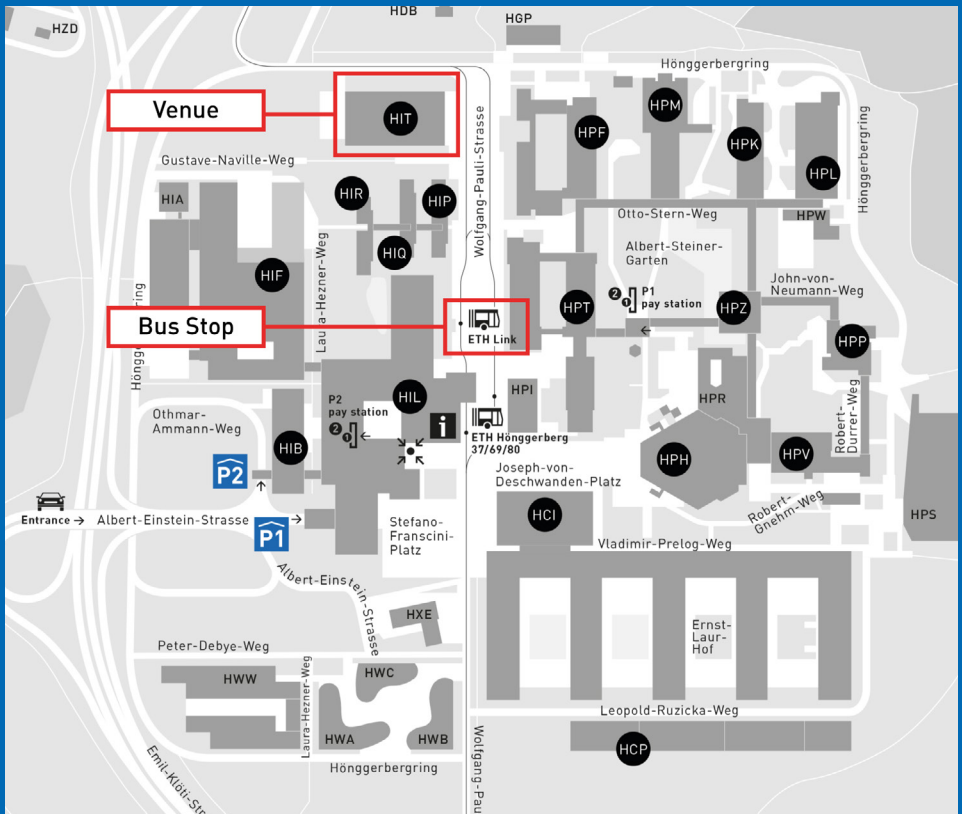
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- Newport (Thursday coffee break).



# Siteplan ETH Höggerberg

## Venue

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## WiFi access

- connect to **public** or **public-5** SSID
- open browser and follow the landing page, which appears automatically
- request internet access by inserting your mobile phone number
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