How to Enhance Nonlinear Optical Signals at the Nanoscale

Presented by:

OSA Nonlinear Optics Technical Group

The OSA Nonlinear Optics Technical Group Welcomes You!

WEBINAR ON HOW TO ENHANCE NONLINEAR OPTICAL SIGNALS AT THE NANOSCALE

10 January 2019 • 11:00 EST

OSA Nonlinear Optics Technical Group



Technical Group Leadership 2018



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Technical Group at a glance

Focus

- "Physics of nonlinear optical materials, processes, devices, & applications"
- **3800** members (**largest** in OIS, 3rd largest in OSA)
- Mission
- To benefit <u>YOU</u>
- webinars, e-Presence, publications, technical events, business events, outreach
- Interested in presenting your research? Have ideas for TG events? Contact us at
- Email: <u>TGNonlinearOptics@osa.org</u>
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- www.osa.org/NonlinearOpticsTG
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- LinkedIn: www.linkedin.com/groups/8302249



Today's webinar





How to enhance non-linear optical signals at the nanoscale

Speaker's short Bio:

Graduation at Indian Institute of Technology in New Delhi, India Ph.D. degree from ETH Zürich (CH) Postdoc at the EPFL in Lausanne (CH) Junior group leader at the Friedrich Schiller University, Jena (DE)

Prof. Dr. Rachel Grange

Research Group leader of Optical Nanomaterial Group Physics Department, Institute for Quantum Electronics, ETH Zürich, Switzerland grange@phys.ethz.ch

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Department of Physics | Institute for Quantum Electronics

Optical Nanomaterial Group

How to Enhance Nonlinear Optical Signals

at the Nanoscale?

Beyond Metal and Semiconductors: Nano-Oxides for Nonlinear Photonics Webinar

Rachel Grange Optical Nanomaterials Group Department of Physics ETH Zurich, Switzerland

grange@phys.ethz.ch @rachel_grange

www.ong.ethz.ch



How to Enhance Nonlinear Optical Signals

www.ong.ethz.ch

at the Nanoscale?



Alumni: Anton Sergeyev, Nick Hendricks, Andrea Steinbrück, Eugenie Kim, Claude Renaut







$$\vec{E}(t) = Acos(\omega t + kz)$$

No change under light

Induced dipole moment



Linear regime

Induced Polarization

$$\vec{P} = \varepsilon_0 \chi^{(1)} \vec{E}$$

dielectric constant of the vaccuum

$$\chi^{(1)}$$
 Electric susceptibility $n^2 = 1 + \chi$

no longer valid for intense light

Induced dipole moment



$$\vec{E}(t) = Acos(\omega t + kz)$$

Second Harmonic Generation



Nonlinear regime

Induced Polarization

$$\vec{P} = \varepsilon_0 \chi^{(1)} \vec{E} + \varepsilon_0 \chi^{(2)} \vec{E}^2 + \varepsilon_0 \chi^{(3)} \vec{E}^3 + \cdots$$

$$\int$$
Second order susceptibility

Second Harmonic Generation (SHG)

Franken et al., Phys Rev Lett, 7,4 1961



t al., 1 hys ivev Lett, 7,4 1301

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Green pointer



Laser for eye surgery





Second Harmonic Generation



Bioimaging



Hsieh, Grange et al, Opt. Exp. 2009

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Green pointer



Second Harmonic Generation



Laser for eye surgery



Bioimaging



Hsieh, Grange et al, Opt. Exp. 2009

Advantages

- + Photon interactions
- + Generation of new color: UV to NIR
- + Spectral filtering (no bleaching)
- + Coherent signal
- + Ultrafast response
- + Revealing crystalline structure

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$$\vec{P} = \varepsilon_0 \chi^{(1)} \vec{E} + \varepsilon_0 \chi^{(2)} \vec{E}^2 + \dots$$

Second Harmonic Generation







Second Harmonic Generation











Tensor coefficients

Material	$\chi^{(2)}$ coeff. (pm/V)
Silicon	0
SiO ₂ (Quartz)	0.335
BaTiO ₃	6.8 to 17
LiNbO ₃	6 to 34
GaAs	134 to 256



$$\vec{P} = \varepsilon_0 \chi^{(1)} \vec{E} + \varepsilon_0 \chi^{(2)} \vec{E}^2 + \dots$$

Second Harmonic Generation





 $\boldsymbol{\chi}^{(2)} = \mathbf{0}$







Tensor coefficients

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Silicon	0
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BaTiO ₃	6.8 to 17
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GaAs	134 to 256





Lithium niobate

Second Harmonic Generation



Tensor coefficients

2ω

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Silicon	0
SiO ₂ (Quartz)	0.335
BaTiO ₃	6.8 to 17
LiNbO ₃	6 to 34
GaAs	134 to 256

R. W. Boyd, Nonlinear Optics (Academic Press, 2008)

Why the metal-oxides ?



Second Harmonic Generation



Advantages of metal-oxides ($\chi^{(2)}$ materials)

- Barium titanate, Lithium niobate as bulk
- High nonlinearities
- Large band gap: 3-4 eV
- Refractive index > 2
- Electro-optic, ferroelectric,...

Material	$\chi^{(2)}$ coeff. (pm/V)
Silicon	0
SiO ₂ (Quartz)	0.335
BaTiO ₃	6.8 to 17
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GaAs	134 to 256



How is the interaction within the material?





How is the interaction within the material?





How is the interaction within the material?



How to deal with phase matching?

- Periodical poling
- Angle or temperature tuning
- In waveguides:
 - modal phase-matching
- **Reducing the size of the material**



Propagation distance



Limitations of current nonlinear optics

$$\vec{P} = \varepsilon_0 \chi^{(1)} \vec{E} + \varepsilon_0 \chi^{(2)} \vec{E}^2 + \cdots$$

Limitations

- High power -> high energy cost and damage
- Bulky material -> no integration
- Phase mismatch -> low output signal
- Small sizes -> reduced signal output

My goal



Limitations of current nonlinear optics



Limitations

- High power -> high energy cost and damage
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Limitations of current nonlinear optics



Limitations

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My goal

Resonance or confinement in particles and thin films to achieve compact devices

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²⁾ materials at small scale





²⁾ materials at small scale





²⁾ materials at small scale





Understanding building blocks



20 - 100s nm

Diffraction limit

$$d = \frac{\lambda}{2NA}$$



Super-resolution multiphoton polarimetric microscope

2018 competitors at the SPIE Europe Photonics Village in Strasbourg / Patent EP18193103.1





Diffraction limit

$$d = \frac{\lambda}{2NA}$$



2018 competitors at the SPIE Europe Photonics Village in Strasbourg / Patent EP18193103.1





Diffraction limit

$$d = \frac{\lambda}{2NA}$$

Special features

- Incident polarization rotation
- Wide field illumination
- Single pixel detection



2018 competitors at the SPIE Europe Photonics Village in Strasbourg / Bridge Funding









2018 competitors at the SPIE Europe Photonics Village in Strasbourg / Bridge Funding

Average SHG intensity of single particles

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Average SHG intensity of single particles



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Average SHG intensity of single particles





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E. Kim,...,and R. Grange, ACS Nano 7, 5343–5349, **2013.** M. Timofeeva,..., R. Grange Nano Lett.,16, 6290, 2016.





M. Timofeeva,..., R. Grange Nano Lett., 16, 6290, 2016.

A powerful characterization tool

SHG intensity is a fourth power dependence

$$I(\alpha) \propto |\chi^{(2)}: E(\alpha)E(\alpha)|^2$$







A powerful characterization tool



SHG intensity is a fourth power dependence

 $I(\alpha) \propto \left|\chi^{(2)}: E(\alpha)E(\alpha)\right|^2$

Decomposition into functions

 $I(\alpha)/I_0 \propto 1 + a_2 \cos 2\alpha + b_2 \sin 2\alpha + a_4 \cos 4\alpha + b_4 \sin 4\alpha$



Amplitudes

Orientations

$$I_{2} = \sqrt{a_{2}^{2} + b_{2}^{2}}, \quad I_{4} = \varepsilon_{4}\sqrt{a_{4}^{2} + b_{4}^{2}}$$
$$\varphi_{2} = \frac{1}{2} \arctan \frac{b_{2}}{a_{2}}, \quad \varphi_{4} = \frac{1}{4} \arctan \frac{b_{4}}{a_{4}}$$

Collaboration with Sophie Brasselet, Inst. Fresnel

Rendon, Timpu, Grange, Brasselet, Scientific report, Accepted, 2018

Super-resolution multiphoton polar microscope



Super-resolution multiphoton polar microscope

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Super-resolution polar multiphoton microscope

Super-resolution polar multiphoton microscope

2D materials layers orientation

Crystal structures along III-V nanowires

Zone III

Pure Zinc Blend

Grange, R et al. Nano letters 12, 2012 M. Timofeeva,..., R. Grange Nano Lett., 16, 6290, 2016.

Strategies to enhance nonlinear optical signals

with plasmonics

• with the $\chi^{(2)}$ material itself

What is plasmonics ?

60 nm gold nanoparticles

Stained glass, Notre Dame de Paris, 1250

Conduction electrons can oscillate at the surface of the metal...

...generating strong resonance in the vicinity of the metal...

... at visible frequencies

El-Sayed, Atwater, Polman, Halas, Novotny, Fritsche, Aizpurua, ...

What is plasmonics ?

60 nm gold nanoparticles

Scattering cross-section

Conduction electrons can oscillate at the surface of the metal...

...generating strong resonance in the vicinity of the metal...

Single electric dipole resonance

El-Sayed, Atwater, Polman, Halas, Novotny, Fritsche, Aizpurua, ...

$\chi^{(2)}$ material core + gold shell

Core-shell BaTiO₃

Extinction measurement in suspension

Y. Pu, R. Grange, Ch.-L. Hsieh, and D. Psaltis, Phys. Rev. Lett., 104, **2010**.

Bioimaging

Hsieh, Grange et al, Opt. Exp. **2009** Hsieh, Grange et al, Biomaterials, **2010** Grange et al., Biomedical Opt Exp, **2011**

SHG enhancement factor > 500

$\chi^{(2)}$ material core + gold shell

Core-shell BaTiO₃

Extinction measurement in suspension

$KNbO_3$ nanowires

J. Richter,..., R. Grange Nanoscale, 6, 5200, 2014.

SHG enhancement factor > 250

$\chi^{(2)}$ material core + gold shell

Core-shell BaTiO₃

Extinction measurement in suspension

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KNbO₃ nanowires

J. Richter,..., R. Grange Nanoscale, 6, 5200, 2014.

SHG enhancement factor > 250

Markers as bright as quantum dots, no bleaching, and biocompatible

Sequential Capillarity-Assisted Particle Assembly

Timpu, F.; Hendricks, N. R.; Petrov, M.; Ni, S.; Renaut, C.; Wolf, H.; Isa, L.; Kivshar, Y.; Grange, R. **Nano Letters**, 17, 5381, 2017. SHG enhancement: 5-15

High-throughput and reduced fabrication complexity

Reshaping the Second-Order Polar Response

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Reshaping the Second-Order Polar Response

Drawback of metals : thermal losses

J. B. Khurgin, How to deal with loss in plasmonics and metamaterials, Nature Nanotechnology, 10, 2015.

Strategies to enhance nonlinear optical signals

with plasmonics

• with the $\chi^{(2)}$ material itself

Mie scattering Rayleigh scattering Multiple scattering

Mie resonances in material with n > 2 and size $\sim \lambda$:

electric and magnetic resonance have comparable strength

Magnetic n=3.44 Si dipole Electric cross-sec. (a.u. dipole Scatt. Magnetic quadrupole 600 800 500 700 900 Wavelength (nm)

Silicon nanopartices with 200 nm

• A. García-Etxarri et al., Opt. Express, 19, (2011)

Krasnok, A. et al., Opt. Express 20, 20599 (2012)

A. B. Evlyukhin et al. , Nano Lett. 12,3749, (2012)

Zywietz, U. et al. Nat. Commun. 5:3402 (2014)

Mie resonances in material with n > 2 and size $\sim \lambda$:

electric and magnetic resonance have comparable strength

Arseniy I. Kuznetsov et al. Science 2016, 354, 2472

Colors vary with sizes of the particle

Magnetic n=3.44 Si dipole Electric cross-sec. (a.u.) dipole Scatt. Magnetic quadrupole 600 500 700 800 900 Wavelength (nm)

Silicon nanopartices with 200 nm

A. García-Etxarri et al., Opt. Express, 19, (2011)

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Mie resonances in material with n > 2 and size $\sim \lambda$:

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Calculated field intensities at the magnetic dipole resonances

'Dielectric' material: electromagnetic field amplified in the volume of the nanoparticle

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GaAs	134 to 256

F. Timpu, A. Sergeyev, N. Hendricks, and R. Grange, ACS Photonics, 4, 2017.

Measured Scattering Spectrum

F. Timpu, A. Sergeyev, N. Hendricks, and R. Grange, ACS Photonics, 4, 2017.

Mie resonances enhance SHG from single nanoparticle

F. Timpu, A. Sergeyev, N. Hendricks, and R. Grange, ACS Photonics, 4, 2017.

Mie resonances in Lithium Niobate Nanocubes

Linear scattering

SHG Polar response

F. Timpu, et al, under review 2019

1.40

Mie resonances in III-V nanowires

Anapoles in Free-Standing III-V Nanodisks

M. Timofeeva,..., R. Grange, Nano Letters, 18,6, 2018

Mie resonances in III-V nanowires

Anapoles in Free-Standing III-V Nanodisks

M. Timofeeva,..., R. Grange, Nano Letters, 18,6, 2018

Anapoles in Free-Standing III-V Nanodisks

Anapoles in Free-Standing III-V Nanodisks

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Strong nonlinear response:

>10³ SHG enhancement

M. Timofeeva,..., R. Grange, Nano Letters, 18,6, 2018

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²⁾ materials at small scale

Perspective: Flat Nonlinear Photonic Devices

BaTiO₃ thin film for nonlinear metasurfaces

Pulse Laser Deposition of M. Trassin and M. Fiebig, Material sciences, ETHZ

Artificial nanostructured materials with exotic electromagnetic responses

Perspective: Flat Nonlinear Photonic Devices

Fabrication and etching challenges of oxides thin films

²⁾ materials at small scale





Bottom-up assembly for large surface area

Colloidal Solution Processing and nanoimprint lithography



Woodpile up to 8 layers





Marseille, France 08.28-31, Talk (2017).



Bottom-up assembly for large surface area

Colloidal Solution Processing and nanoimprint lithography



Woodpile up to 8 layers





Large surface area 2 x 2 cm Disposable sensors No etching

V. V. Vogler-Neuling, ... and R. Grange, Metamaterials Marseille, France 08.28-31, Talk (2017).



²⁾ materials at small scale



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Bottom-up processing

Wide surface area photonic crystal





Top-down fabrication

Flat photonic structures



Integrated devices



Integrated Photonics

Less weight and more transmitted data



Broadly transparent, low losses, fast, compact, robust



LiNbO₃ nanowaveguides by ion beam enhanced etching



A. Sergeyev,... R. Grange, Opt. Exp. 21, 19012, 2013.



LiNbO3 nanowaveguides by ion beam enhanced etching

400 nm diameter



Signal dependence on the incident laser power



A. Sergeyev,... R. Grange, Opt. Exp. 21, 19012, 2013.



LiNbO₃ nanowaveguides by ion beam enhanced etching

400 nm diameter

50 μm length

How to achieve phase-matching? In waveguides modal phase-matching



Sergeyev, A.; et al. ACS Photonics 2015, 2 (6), 687-691.



LiNbO3 nanowaveguides by ion beam enhanced etching

400 nm diameter



50 times increase with modal phase matching



Sergeyev, A.; et al. ACS Photonics 2015, 2 (6), 687-691.



LiNbO3 on insulator LNOI





LNOI ridge waveguides





Pockel's effect

Change in the refractive index linearly proportional to the electric field



Electro-optic tensor of LiNbO₃

Electro-optic tuning capabilities



Electric field has to be parallel to the z-crystal axis



Pockel's effect



Electro-optic tuning capabilities



Electric field has to be parallel to the z-crystal axis



Pockel's effect



Distributed Bragg reflector

Multilayers of alternating materials with varying n, each layer causes a partial reflection



Period : 384 nm Corrugation depth : 90 nm Propagation losses:

 α_{TE} =8.07 dB/cm α_{TM} =3 dB/cm





Distributed Bragg reflector

Multilayers of alternating materials with varying n, each layer causes a partial reflection



Period : 384 nm Corrugation depth : 90 nm Propagation losses: α_{TE} =8.07 dB/cm α_{TM} =3 dB/cm





Distributed Bragg reflector

Multilayers of alternating materials with varying n, each layer causes a partial reflection







- *R*_{peak} 93.5 %
- 14 dB in transmission suppression

Distributed Bragg reflector

Multilayers of alternating materials with varying n, each layer causes a partial reflection



Escalé, M. R.; Pohl, D.; Sergeyev, A.; Grange, R. Optics Letters, 43, 1515, 2018

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Wide tuning of the stop band





Wide tuning of the stop band



Insertion losses : 12.9 dB Extinction ratio 14dB

Footprint: 10×500 µm²

Bandwidth 2nm

33 x more tuning than state-of-the-art devices !



Wide tuning of the stop band



33 x more tuning than state-of-the-art devices !



²⁾ materials at small scale





Bottom-up processing

Wide surface area photonic crystal





Top-down fabrication

Flat photonic structures



Integrated devices



Escalé, M et al. Opt. Lett., 43, 1515, 2018



⁽²⁾ materials at small scale

$\chi^{(2)}$ Building blocks



Timpu, et al. ACS Photonics, 4, 2017. Renaut et al., Nano Lett., 2019

AlGaAs anapole



M. Timofeeva, et al. , Nano Letters, accepted May 17, 2018

Super-Polar microscope



Bottom-up processing Wide surface area photonic crystal





Top-down fabrication

Flat photonic structures



Integrated devices



Escalé, M et al. Opt. Lett., 43, 1515, 2018

