

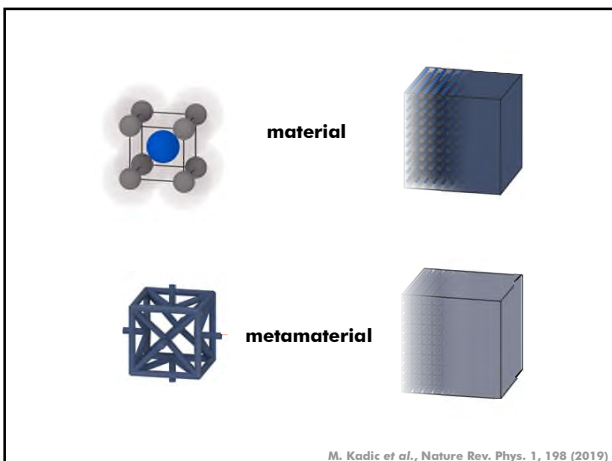
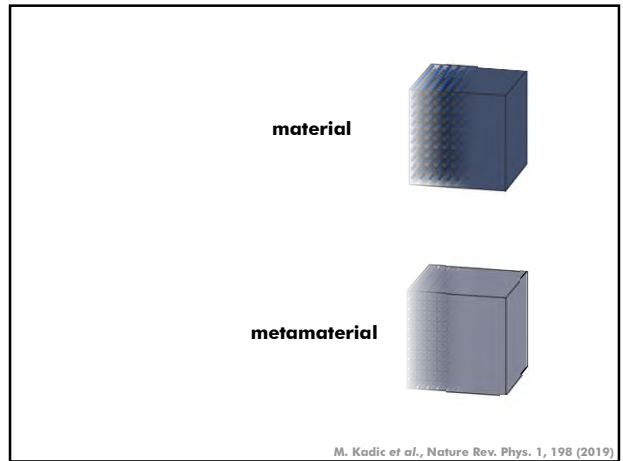
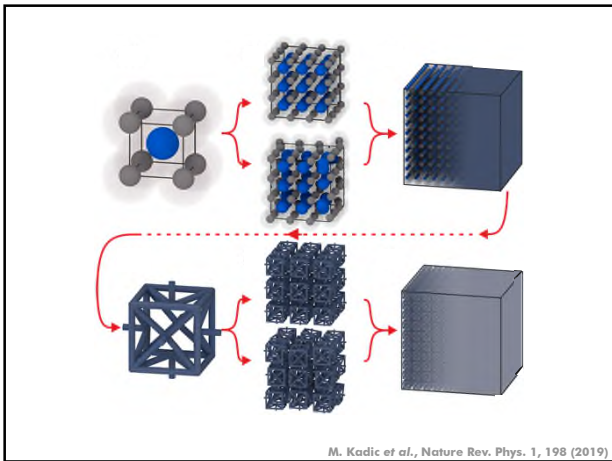
3D Mechanical Metamaterials Going Beyond Cauchy Elasticity

Martin Wegener
Karlsruhe Institute of Technology (KIT)

International Training School, Valencia (Spain), November 13-17, 2023

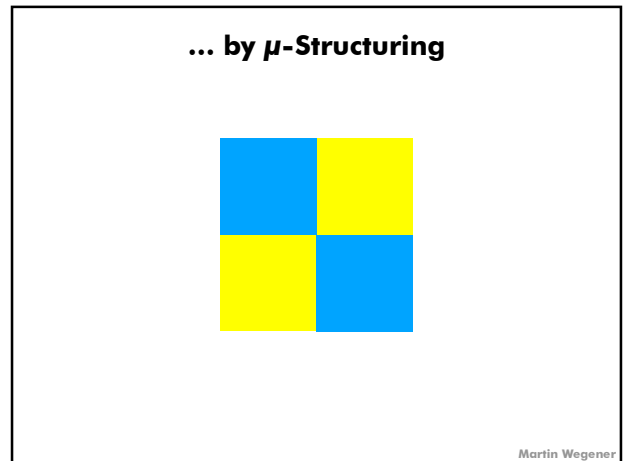
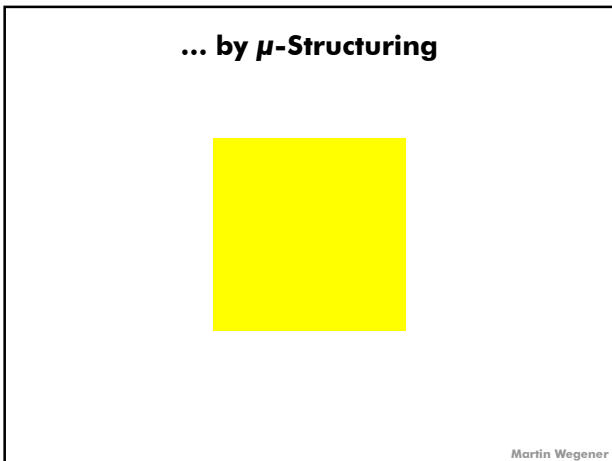
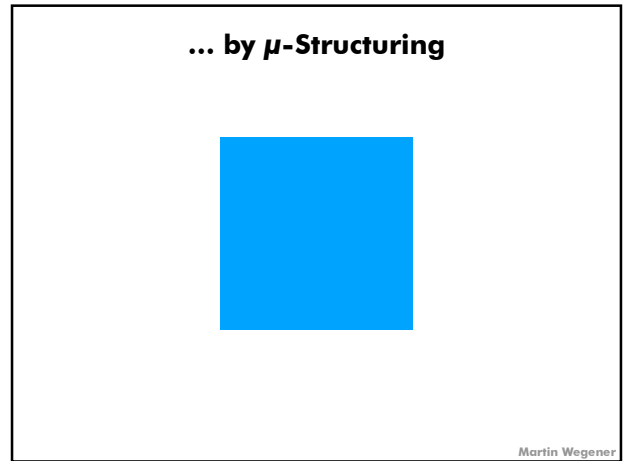
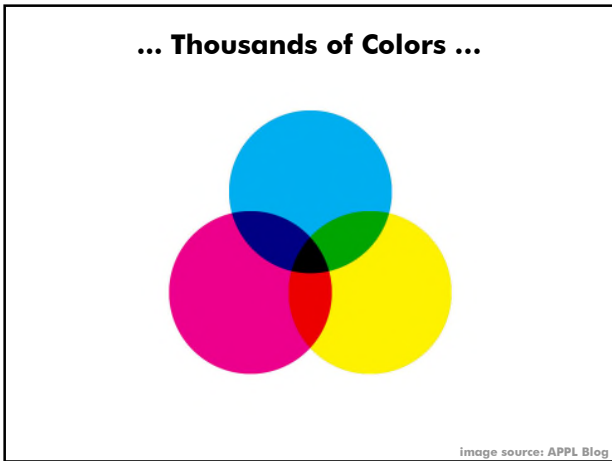
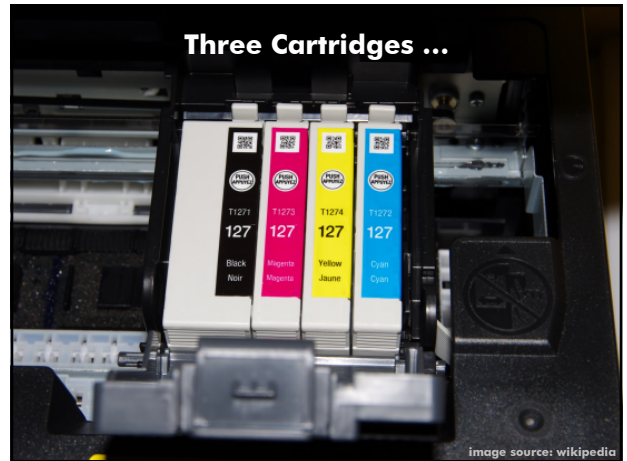
Metamaterials are **rationally designed** composites allowing for effective-medium properties that go qualitatively or quantitatively **beyond ("meta")** those of the bulk ingredients.

Martin Wegener

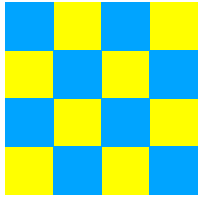


Bounds for Properties?

in some cases, metamaterials allow for unbounded behavior

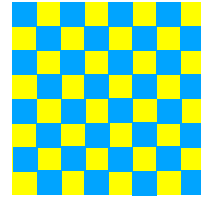


... by μ -Structuring



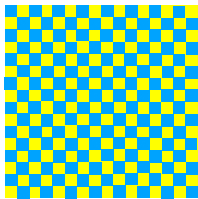
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... by μ -Structuring



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... by μ -Structuring



Martin Wegener

... by μ -Structuring



Martin Wegener

Today's 2D graphical printers allow for thousands of colors from just four cartridges.

By analogy, future 3D material printers might allow for thousands of effective (meta-)material properties from just a few "material cartridges".

(Faded text, likely a quote or reference)

M. Kadic et al., Nature Rev. Phys. 1, 198 (2019)

Metamaterials as "Meta-Inks"

(Faded text, likely a quote or reference)

Martin Wegener

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 - 5.1. Complex Band Structures and the Cauchy-Riemann Equations
 - 5.2. Examples

Martin Wegener

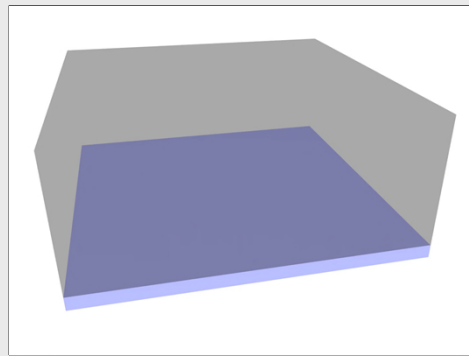
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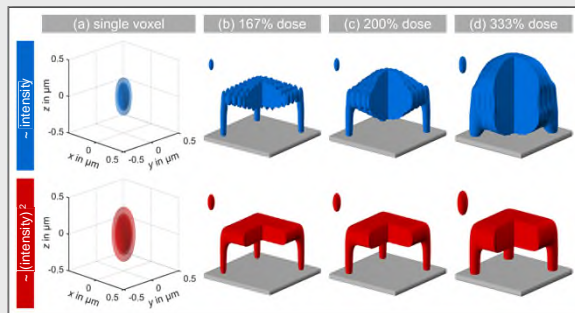
Martin Wegener

Writing With a Pen of Light



scheme, not to scale; 3Dmattermatorder.kit.edu

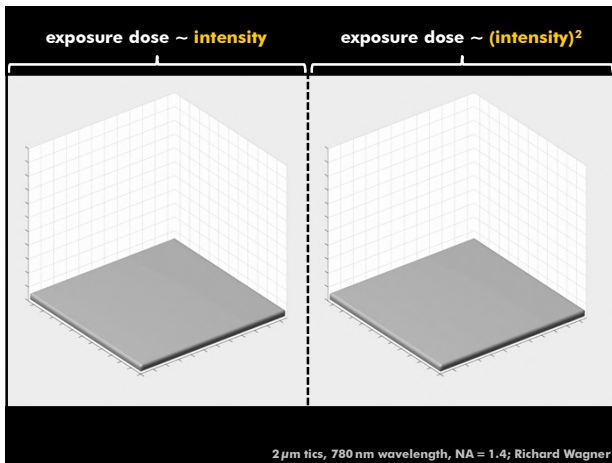
Proximity Effect



P. Kiefer et al., Adv. Opt. Mater. 8, 1907795 (2020)

Fictitious *In-Situ* Diagnostics

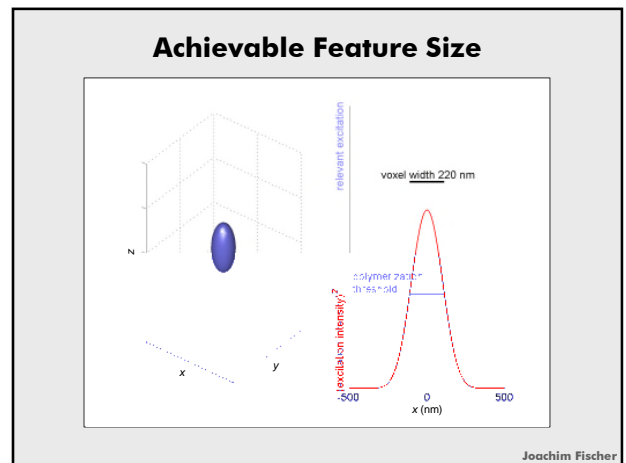
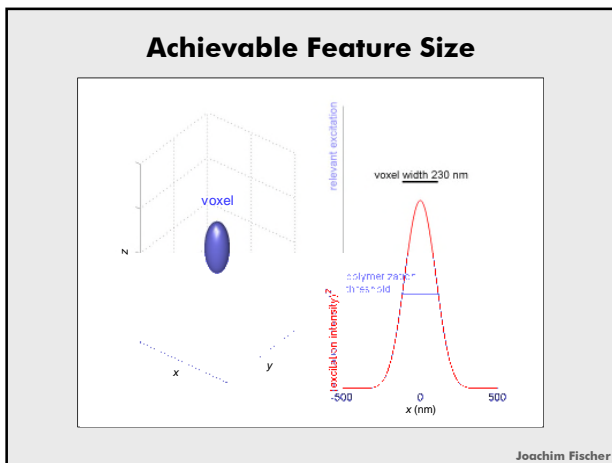
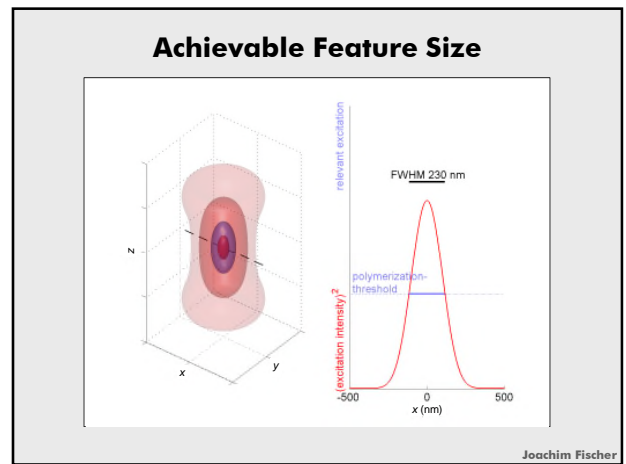
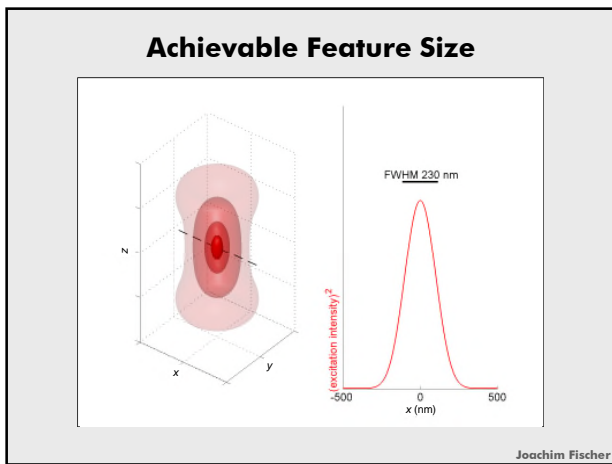
Martin Wegener



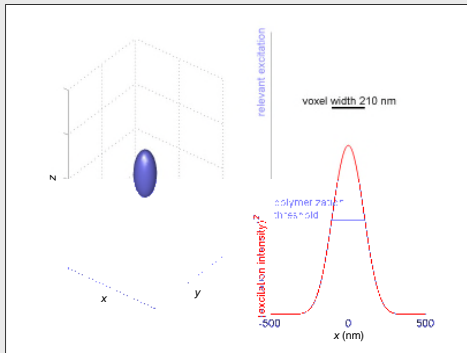
Optics Limit on Feature Size?

Optics limit on feature size?

Martin Wegener

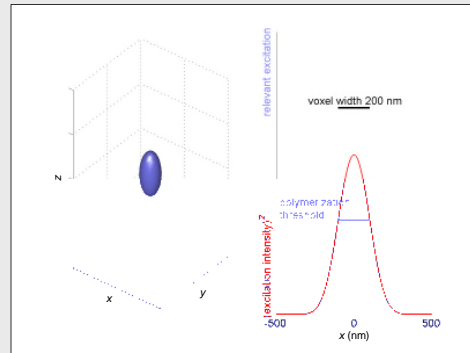


Achievable Feature Size



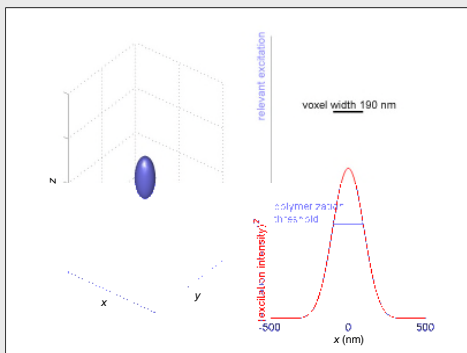
Joachim Fischer

Achievable Feature Size



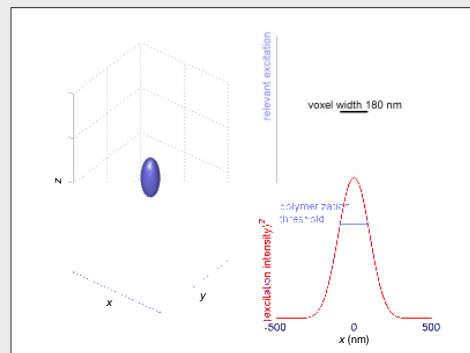
Joachim Fischer

Achievable Feature Size



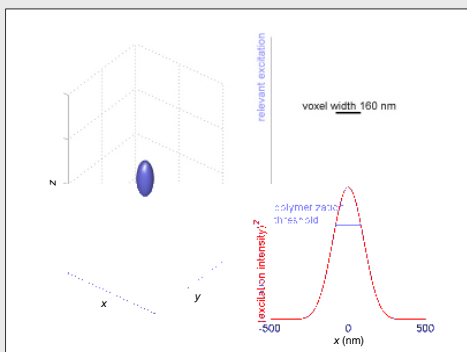
Joachim Fischer

Achievable Feature Size



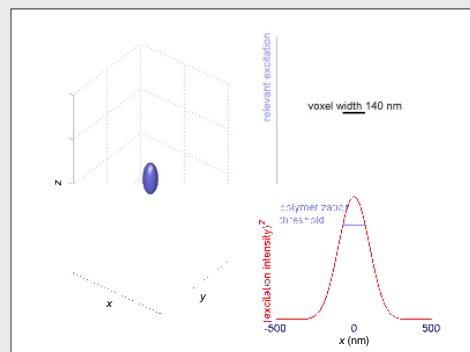
Joachim Fischer

Achievable Feature Size



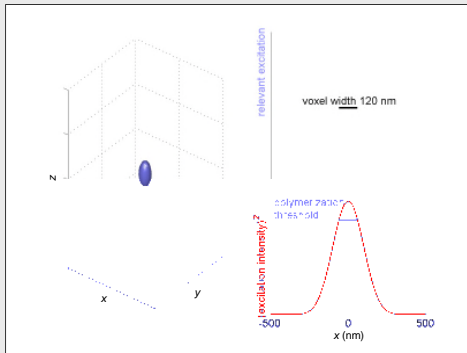
Joachim Fischer

Achievable Feature Size



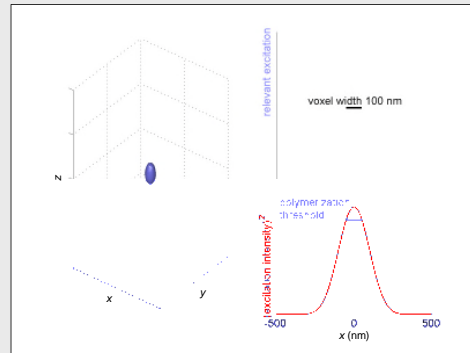
Joachim Fischer

Achievable Feature Size



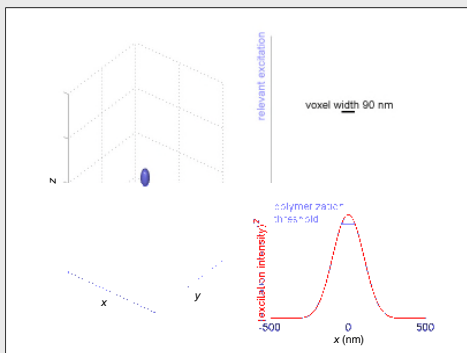
Joachim Fischer

Achievable Feature Size



Joachim Fischer

Achievable Feature Size



Joachim Fischer

No Optics Limit on Feature Size

No Optics Limit on Feature Size

Martin Wegener

Optics Limit on Resolution?

Optics Limit on Resolution?

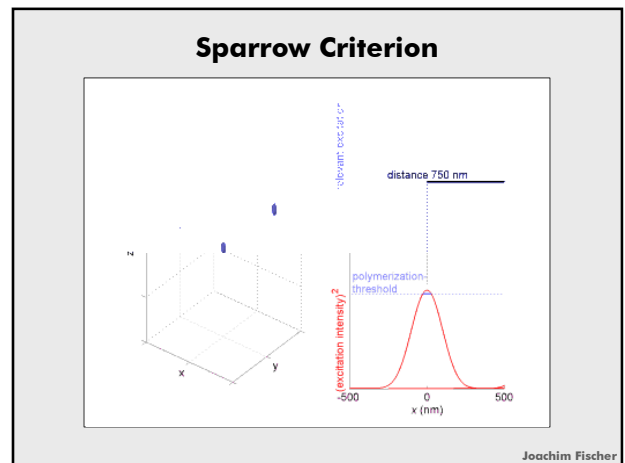
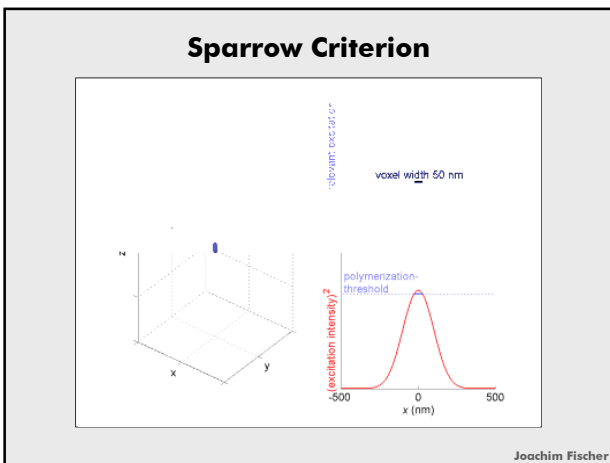
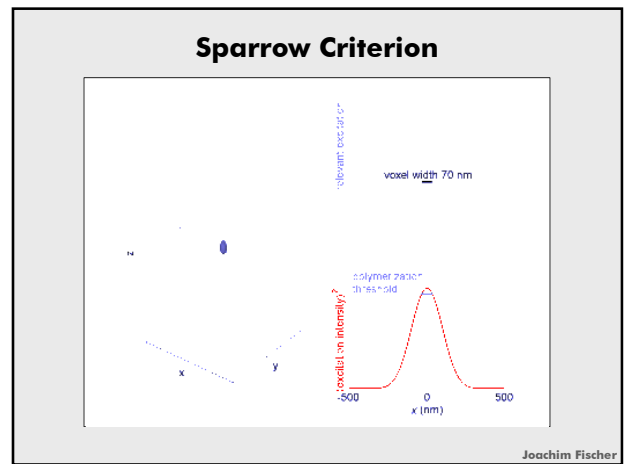
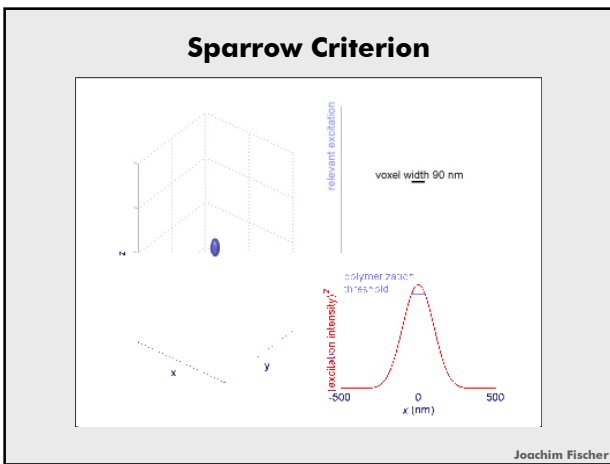
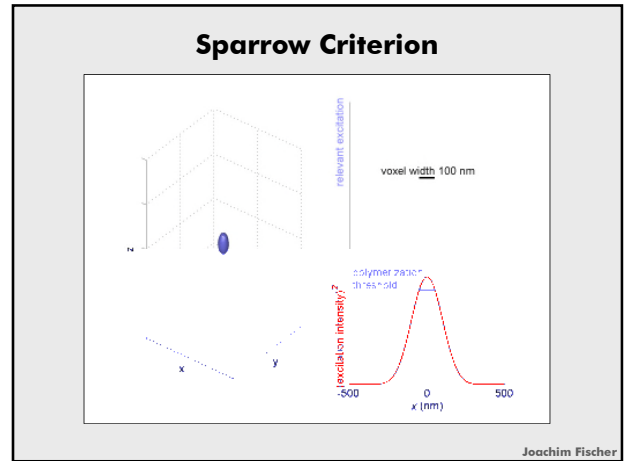
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Oxford Dictionary

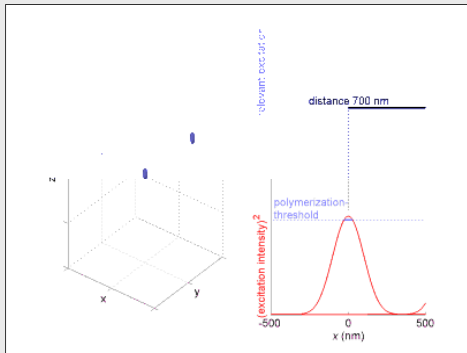
resolve

“... [with object] (of optical or photographic equipment) separate or distinguish between (closely adjacent objects). Hubble was able to resolve six variable stars in M31 ...”

Martin Wegener

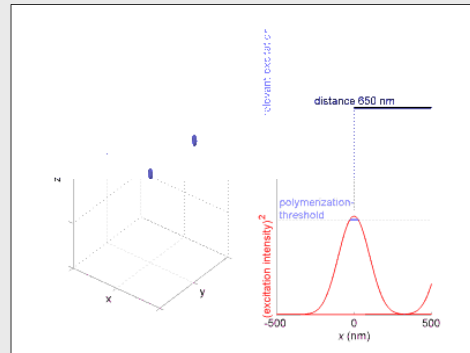


Sparrow Criterion



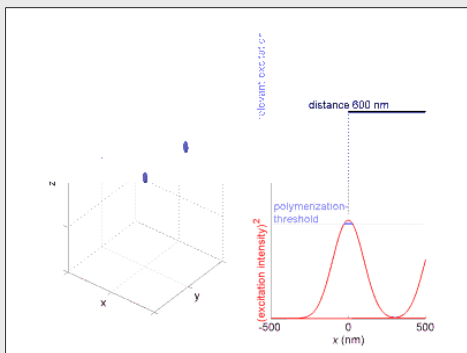
Joachim Fischer

Sparrow Criterion



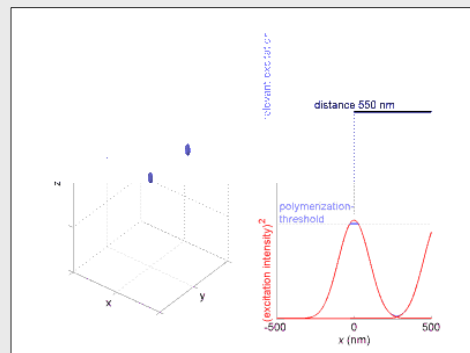
Joachim Fischer

Sparrow Criterion



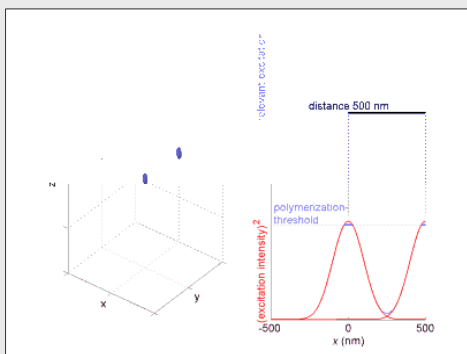
Joachim Fischer

Sparrow Criterion



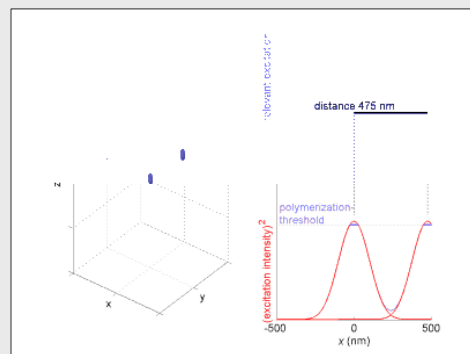
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Sparrow Criterion



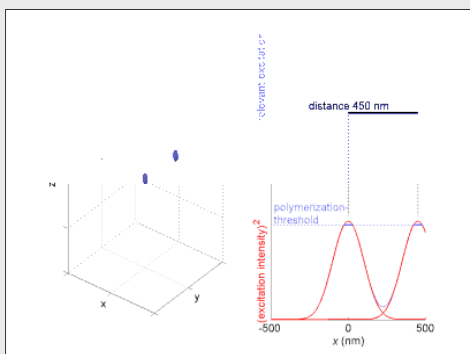
Joachim Fischer

Sparrow Criterion



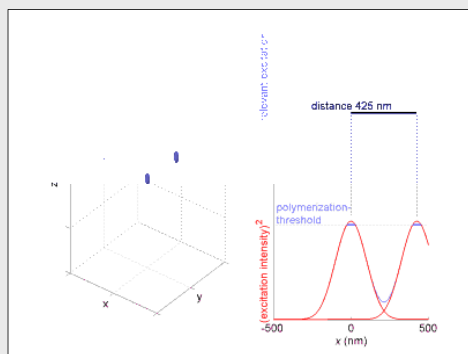
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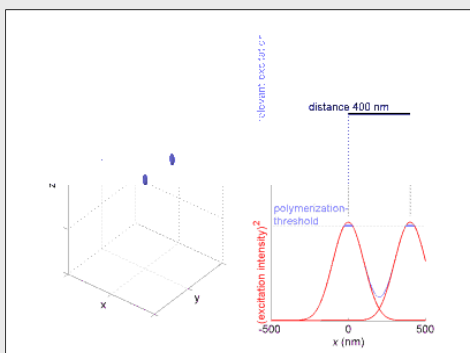
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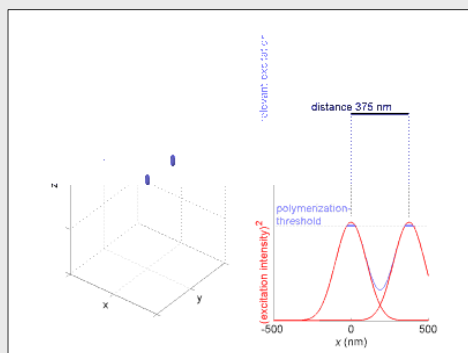
Joachim Fischer

Sparrow Criterion



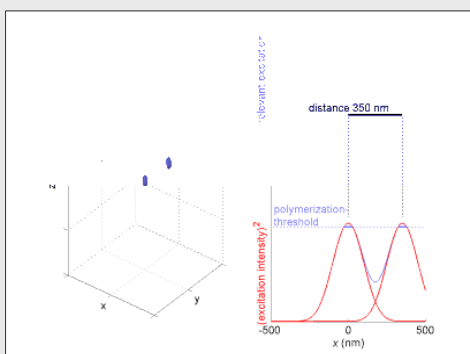
Joachim Fischer

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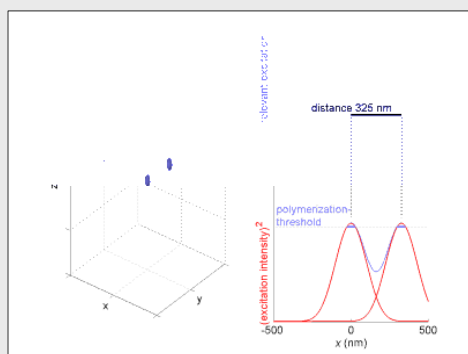
Joachim Fischer

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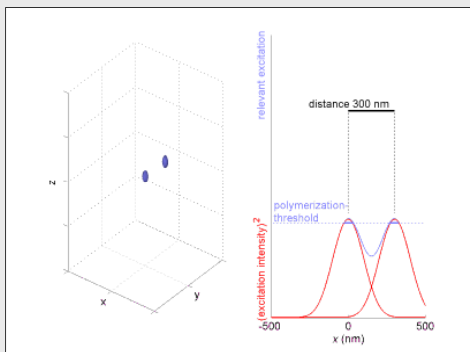
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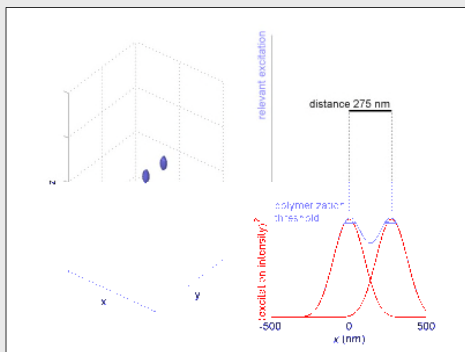
Joachim Fischer

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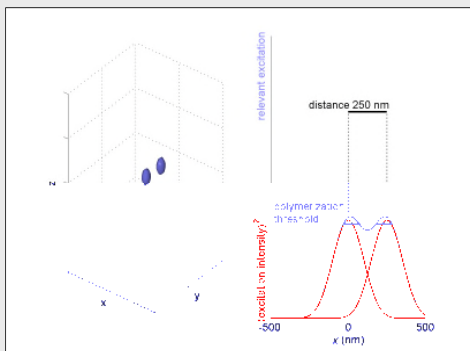
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Sparrow Criterion



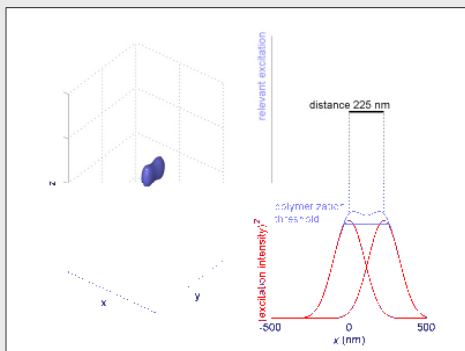
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Sparrow Criterion



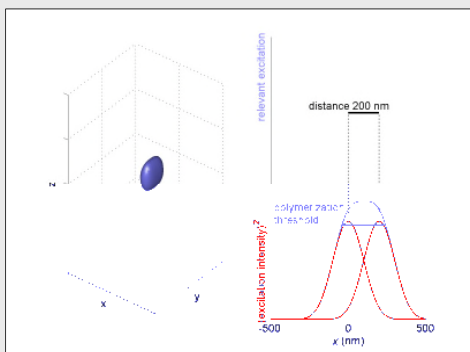
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Sparrow Criterion



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Sparrow Criterion



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Optics Limit on Resolution

Optics limit on resolution

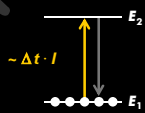
Sparrow criterion depends on nonlinearity of the process

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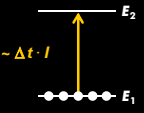


Albert Einstein
 1879 * Ulm (Germany)
 1905 Dissertation @ Zürich
Photoelectric effect
 1921 Nobel Prize in Physics
 1955 † Princeton




A. Einstein, Ann. Phys. 322, 132 (1905)

Probability?

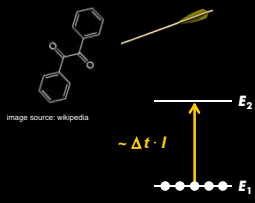


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Target area: m²



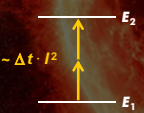
Molecule absorption cross section: 10⁻¹⁵ cm² (typ. @ RT)



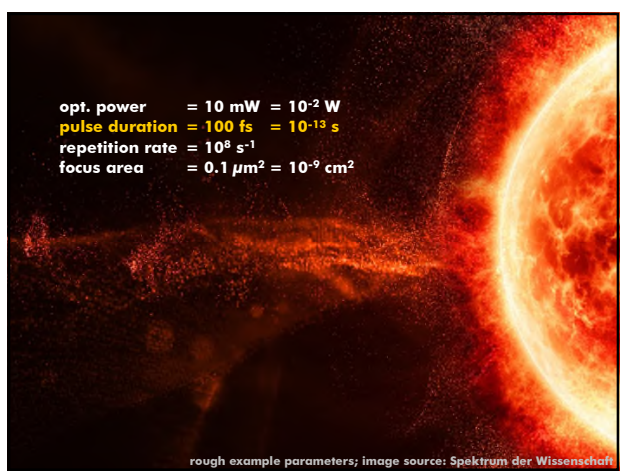
Martin Wegener



Marie Göppert-Mayer
 1906 * Kattowitz (Poland)
 1930 Dissertation @ Göttingen (Max Born)
 on **theory of two-photon absorption**
 1963 Nobel Prize in Physics
 1972 † San Diego



M. Göppert-Mayer, Ann. Phys. 401, 273 (1931)



opt. power = 10 mW = 10⁻² W
 pulse duration = 100 fs = 10⁻¹³ s
 repetition rate = 10⁸ s⁻¹
 focus area = 0.1 μm² = 10⁻⁹ cm²

rough example parameters; image source: Spektrum der Wissenschaft

opt. power = 10 mW = 10^{-2} W
 pulse duration = 100 fs = 10^{-13} s
 repetition rate = 10^8 s⁻¹
 focus area = $0.1 \mu\text{m}^2 = 10^{-9}$ cm²

leads to

opt. intensity = 1 TW cm⁻² = 10^{12} W cm⁻²
 flux density = 10^{31} photons s⁻¹ cm⁻²

rough example parameters; image source: Spektrum der Wissenschaft

opt. power = 10 mW = 10^{-2} W
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leads to

opt. intensity = 1 TW cm⁻² = 10^{12} W cm⁻²
 flux density = 10^{31} photons s⁻¹ cm⁻²

1000 GM = 10^{-47} cm⁴ s photons⁻¹ } → 10⁻¹⁶ cm² cross section

rough example parameters; image source: Spektrum der Wissenschaft

Theory of two-photon absorption
 M. Göppert-Mayer, *Ann. Phys.* 401, 273 (1931)

First experiment on two-photon absorption
 W. Kaiser and C.G.B. Garrett, *Phys. Rev. Lett.* 7, 229 (1961)

Two-photon laser microscopy
 W. Denk, J. Strickler, and W. Webb, *Science* 248, 73 (1990)

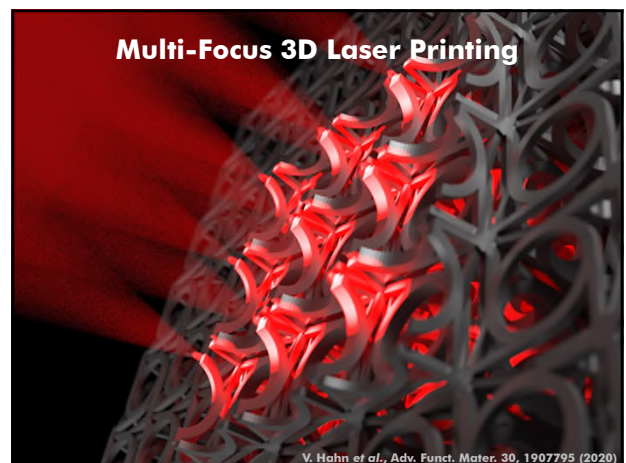
Two-photon laser lithography
 S. Maruo, O. Nakamura, and S. Kawata, *Opt. Lett.* 22, 132 (1997)

Martin Wegener

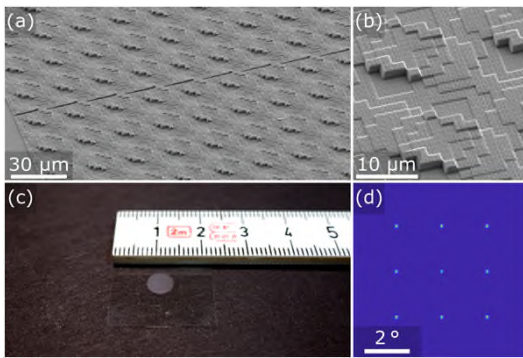
3D Laser Nanoprinting

User friendly software package
 Microscope docking station
 Optics cabinet with all necessary optics
 Turn-key NIR fiber laser
 Scanning unit based on pivoted galvo mirrors
 Positioning system (motorized stage + piezo stage)
 Microscope with autofocus system
 High-sensitivity microscope camera
 Self-leveling vibration isolation frame with optical breadboard
 Electronics rack with controllers and PC

www.nanoscribe.de (2014)

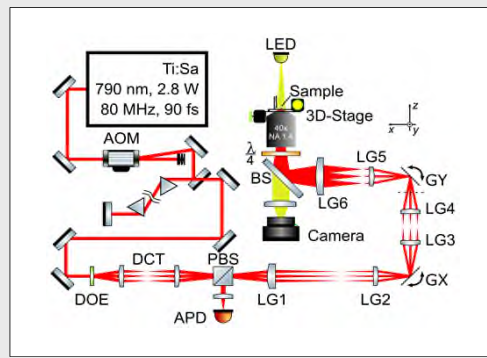


3D Printing the 3D Printer

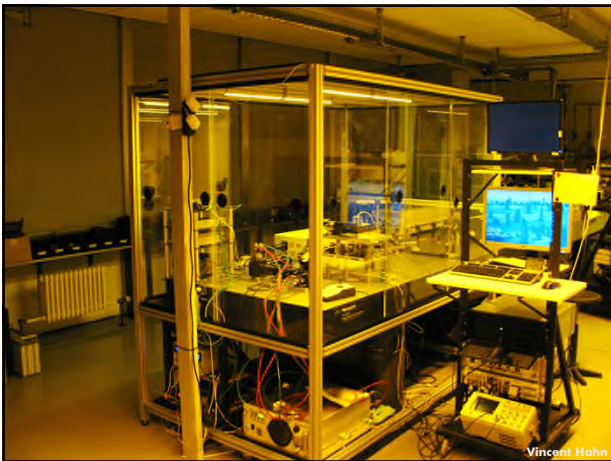


V. Hahn et al., Adv. Funct. Mater. 30, 1907795 (2020)

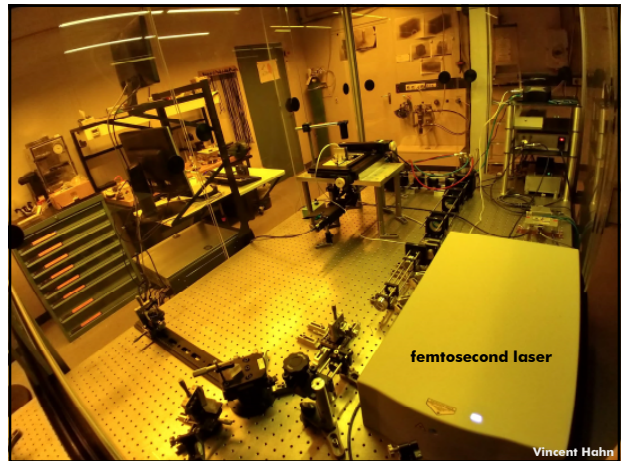
Multi-Focus 3D Laser Printing



V. Hahn et al., Adv. Funct. Mater. 30, 1907795 (2020)



Vincent Hahn



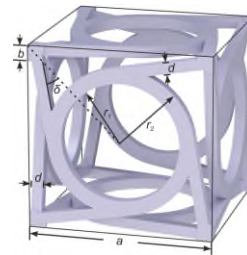
Vincent Hahn

Example

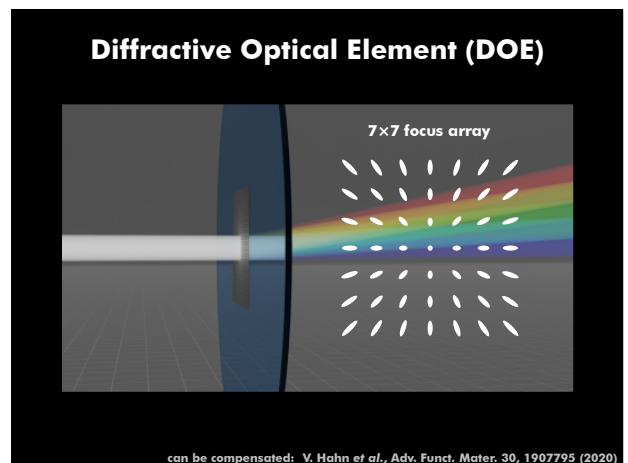
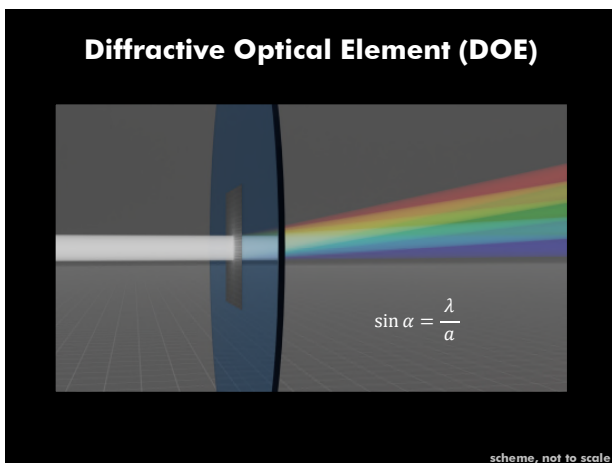
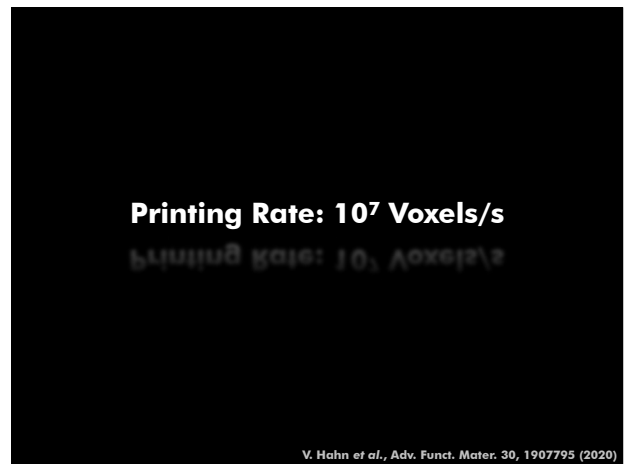
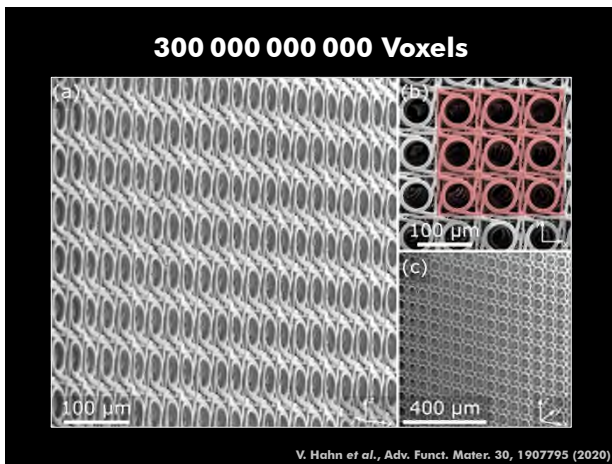
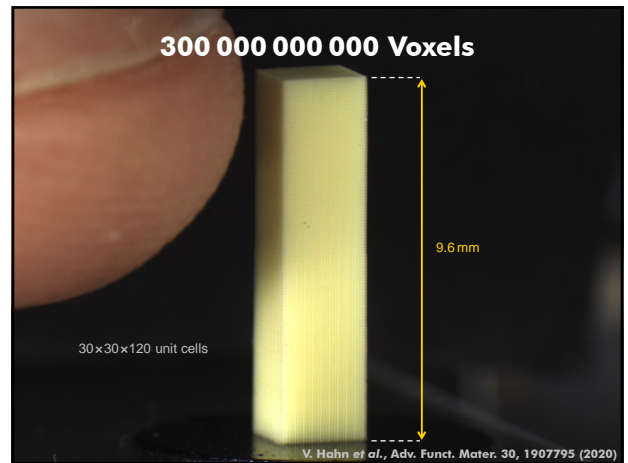
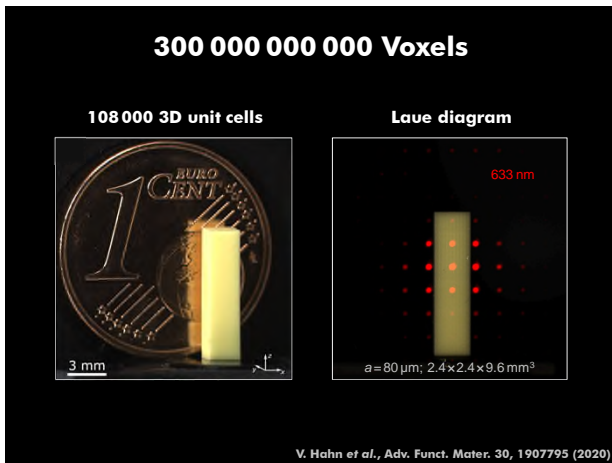
example

Martin Wegener

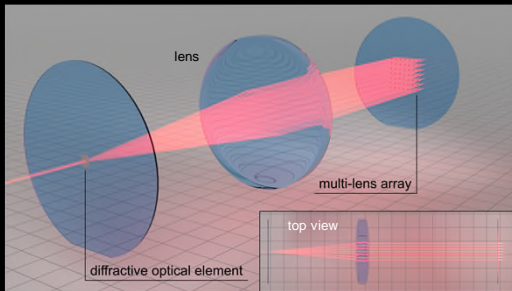
3D Chiral Cubic Unit Cell



T. Frenzel et al., Science 358, 1072 (2017)

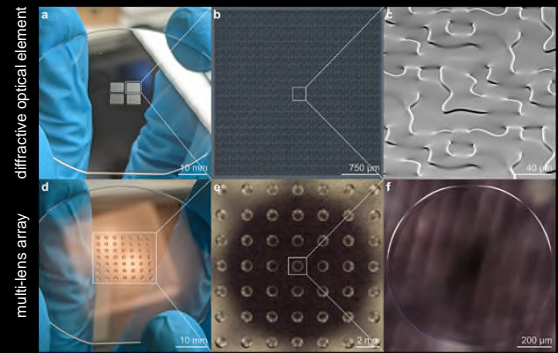


DOE and Multi-Lens Array (MLA)



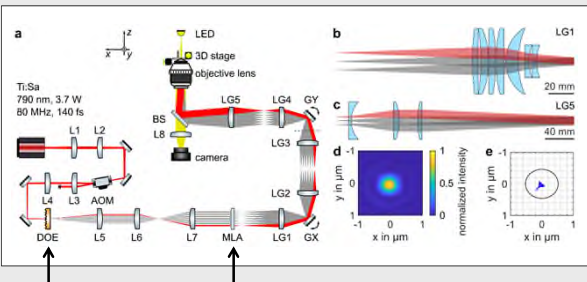
P. Kiefer *et al.*, submitted (2023)

Laser-Printed DOE & MLA



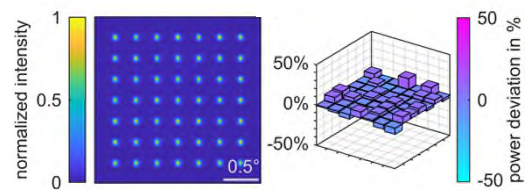
printed using Nanoscribe Quantum X

Multi-Focus Multi-Photon 3D Printer



P. Kiefer *et al.*, submitted (2023)

$7 \times 7 = 49$ Laser Foci



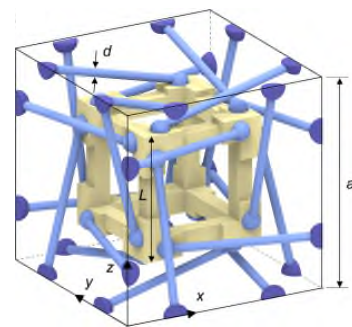
about 1/3 of the fs-laser power enters the entrance pupil of the microscope lens

Example

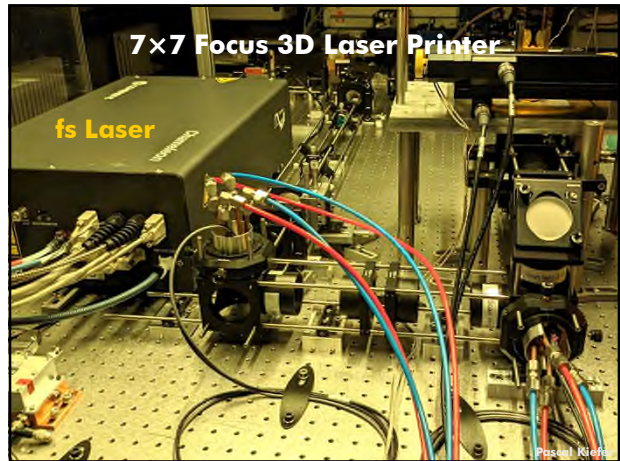
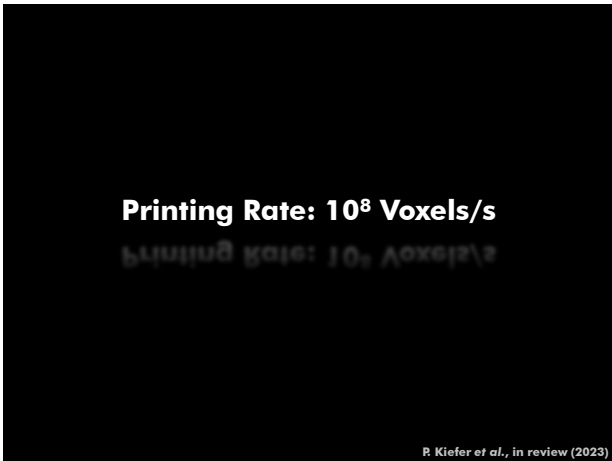
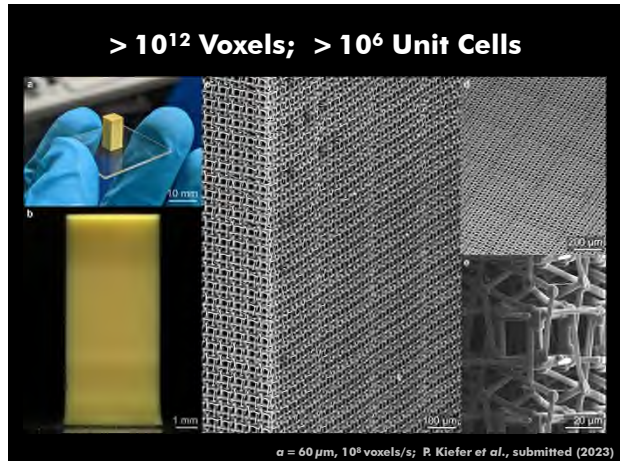
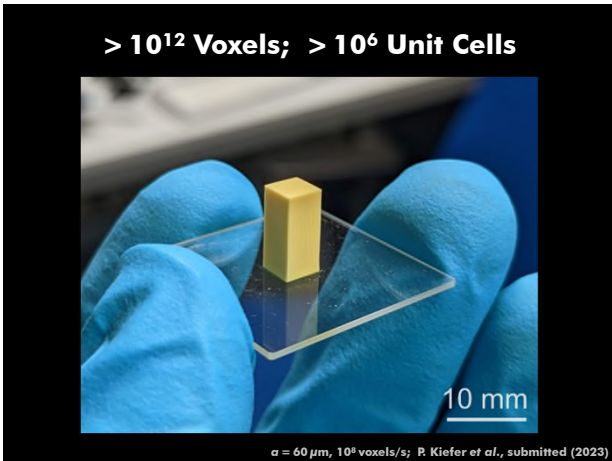
example

Martin Wegener

3D Chiral Cubic Unit Cell



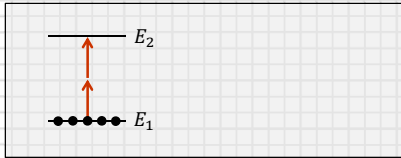
used parameters: $a = 185 \mu\text{m}$, $d/a = 0.04$, $L/a = 0.6$



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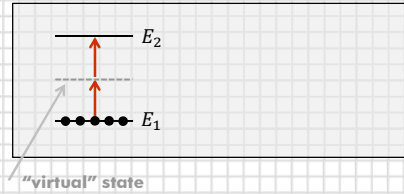


Two-photon absorption versus two-step absorption



M. Göppert-Mayer, Ann. Phys. 401, 273 (1931)

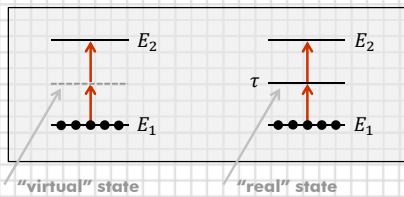
Two-photon absorption versus two-step absorption



"virtual" state

M.O. Scully and M.S. Zubairy, "Quantum Optics", Cambridge Univ. Press (1997)

Two-photon absorption versus two-step absorption



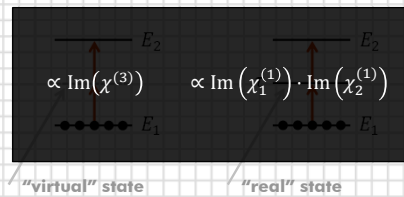
"virtual" state

"real" state

In both cases, one can obtain $N_2 \propto I^2$

P. Kiefer et al., Adv. Opt. Mater. 8, 1907795 (2020)

Two-photon absorption versus two-step absorption



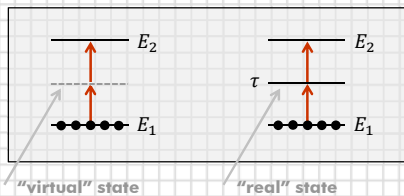
"virtual" state

"real" state

In both cases, one can obtain $N_2 \propto I^2$

P. Kiefer et al., Adv. Opt. Mater. 8, 1907795 (2020)

Two-photon absorption versus two-step absorption



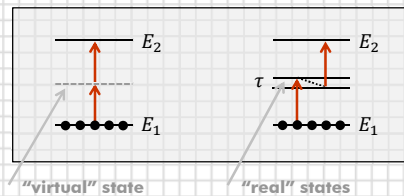
"virtual" state

"real" state

In both cases, one can obtain $N_2 \propto I^2$

P. Kiefer et al., Adv. Opt. Mater. 8, 1907795 (2020)

Two-photon absorption versus two-step absorption



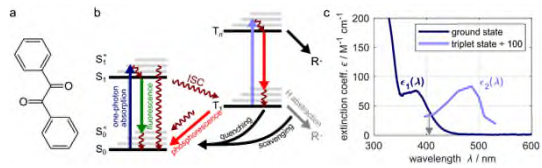
"virtual" state

"real" states

In both cases, one can obtain $N_2 \propto I^2$

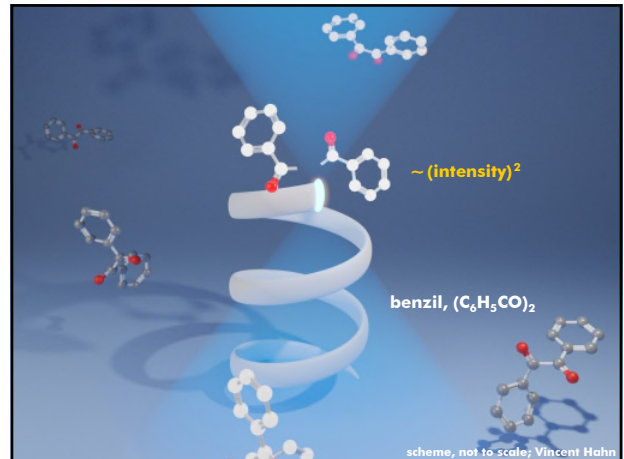
V. Hahn et al., Nature Photon. 15, 932 (2021)

Photoresist

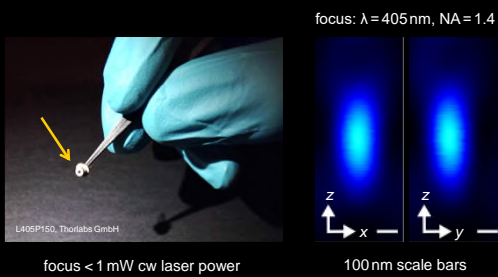


- benzil as two-step photoinitiator
- bis(2,2,6,6-tetramethyl-4-piperidyl-1-oxyl) sebacate (BTPOS) as quencher and scavenger
- pentaerythritol triacrylate (PETA) as monomer

V. Hahn *et al.*, Nature Photon. 15, 932 (2021)

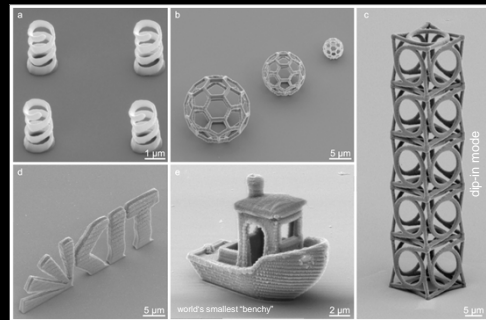


Use Inexpensive cw Laser Diode



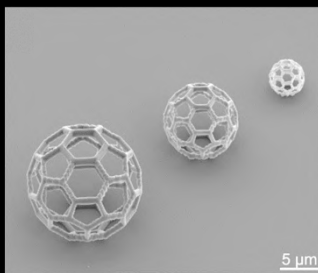
V. Hahn *et al.*, Nature Photon. 15, 932 (2021)

Two-Step 3D Nanoprinting



V. Hahn *et al.*, Nature Photon. 15, 932 (2021)

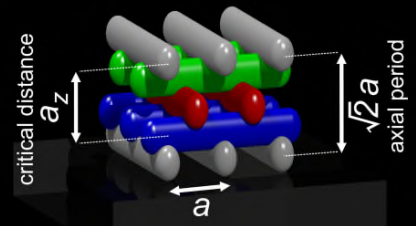
Two-Step 3D Nanoprinting



objective lens: NA = 1.4
 hatching: 30 nm
 slicing: 100 nm
 wavelength: 405 nm
 cw laser power: 320 μW
 scanning mode: galvo
 scan speed: 1.0 mm/s
 printing rate: 4800 voxel/s
 development: acetone

V. Hahn *et al.*, Nature Photon. 15, 932 (2021)

3D Woodpiles as Benchmark



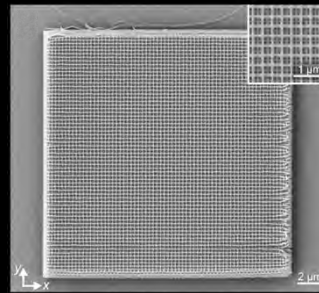
J. Fischer and M. Wegener, Laser Phot. Rev. 7, 22 (2013)

Two-Step 3D Nanoprinting



V. Hahn *et al.*, *Nature Photon.* 15, 932 (2021)

Two-Step 3D Nanoprinting

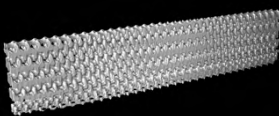


objective lens: NA = 1.4
 rod spacing: $a = 300$ nm
 # layers: 24
 wavelength: 405 nm
 cw laser power: 45 μ W
 scanning mode: piezo
 scan speed: 0.1 mm/s
 printing rate: 480 voxels/s
 development: acetone CPD

V. Hahn *et al.*, *Nature Photon.* 15, 932 (2021)

Two-Step 3D Nanoprinting

ultramicrotomy



objective lens: NA = 1.4
 rod spacing: $a = 300$ nm
 # layers: 24
 wavelength: 405 nm
 cw laser power: 45 μ W
 scanning mode: piezo
 scan speed: 0.1 mm/s
 printing rate: 480 voxels/s
 development: acetone CPD

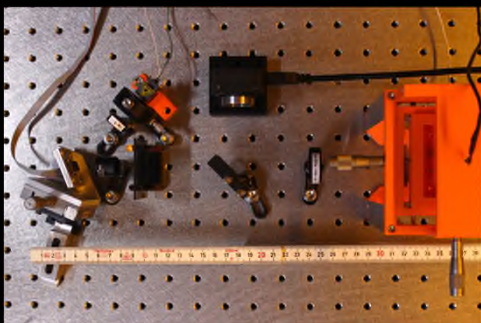
V. Hahn *et al.*, *Nature Photon.* 15, 932 (2021)

Miniaturized 3D Laser Nanoprinter

Miniaturized 3D Laser Nanoprinter

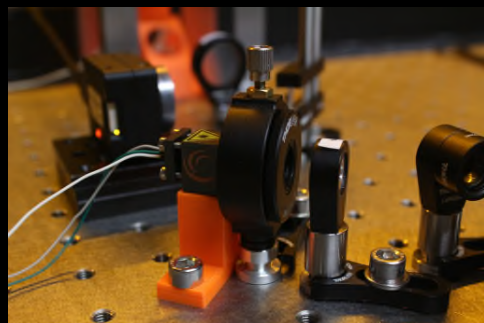
Martin Wegener

Shoobox Sized Nanoprinter



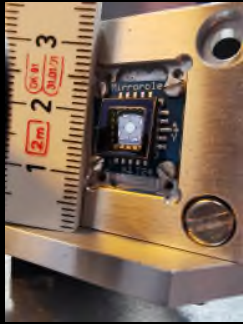
T. Messer *et al.*, *Light: Adv. Man.*, in review (2023)

Shoobox Sized Nanoprinter



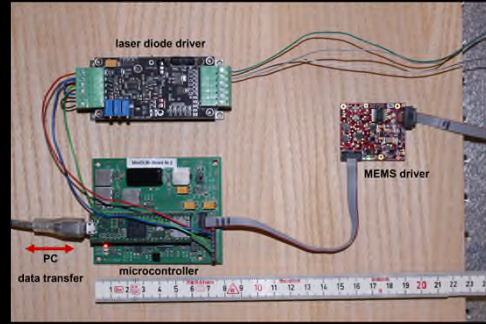
T. Messer *et al.*, *Light: Adv. Man.*, in review (2023)

Shoebbox Sized Nanoprinter



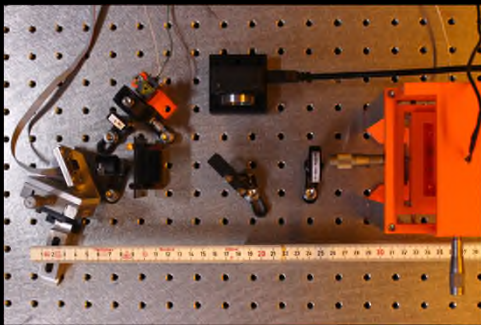
T. Messer et al., Light: Adv. Man., in review (2023)

Shoebbox Sized Nanoprinter



T. Messer et al., Light: Adv. Man., in review (2023)

Shoebbox Sized Nanoprinter



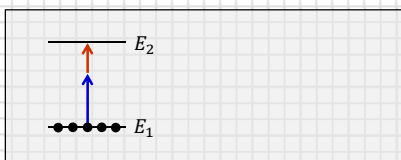
T. Messer et al., Light: Adv. Man., in review (2023)

Two Colors

Two Colors

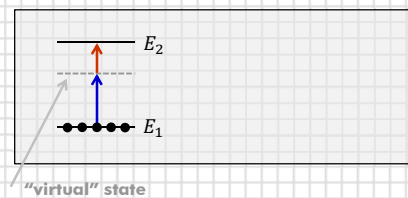
Martin Wegener

Two-photon absorption versus two-step absorption

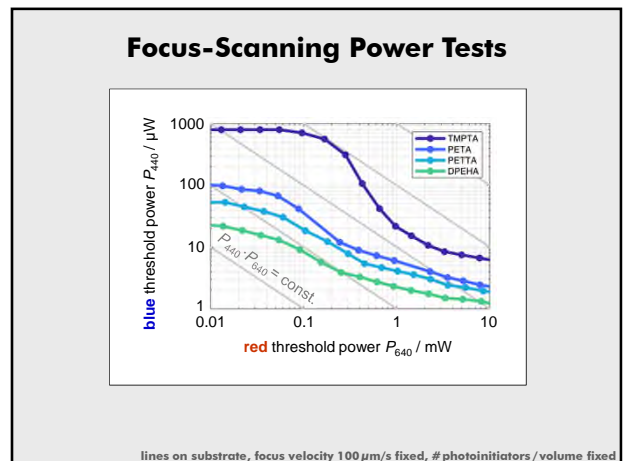
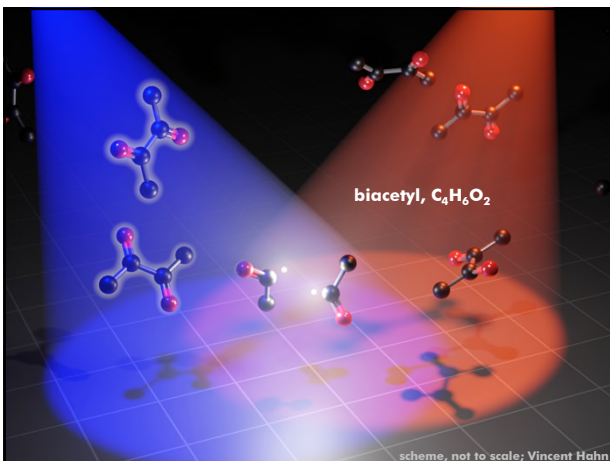
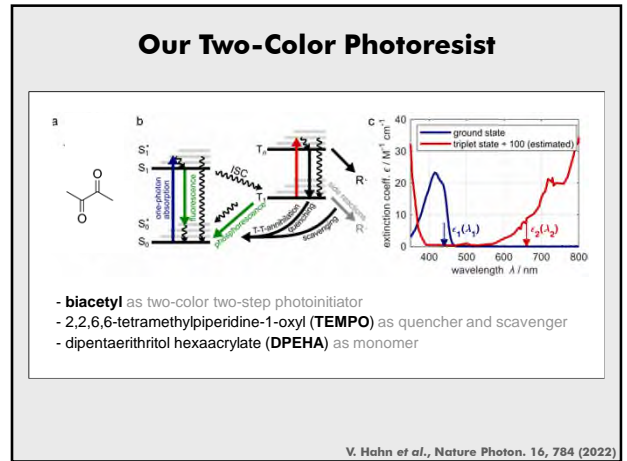
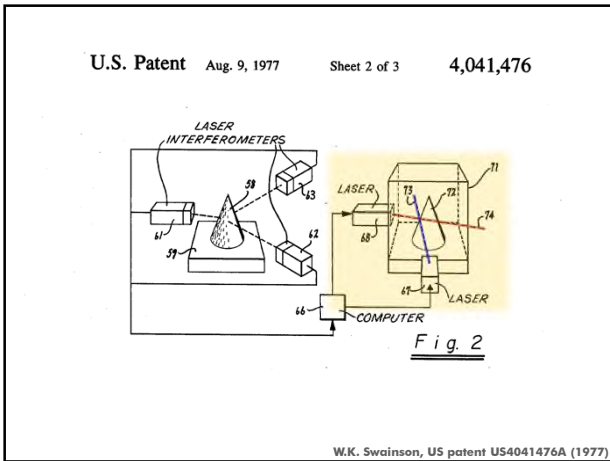
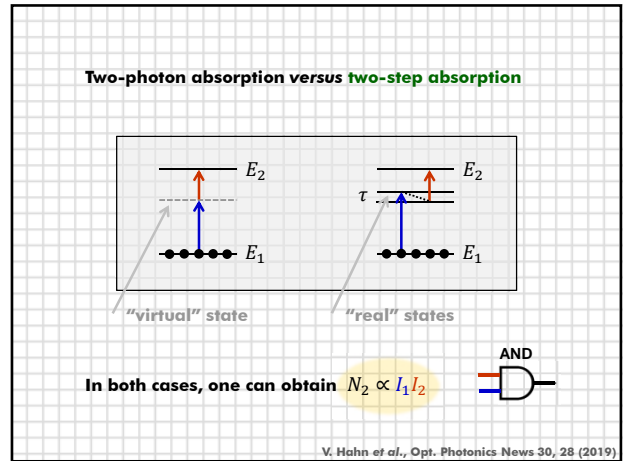
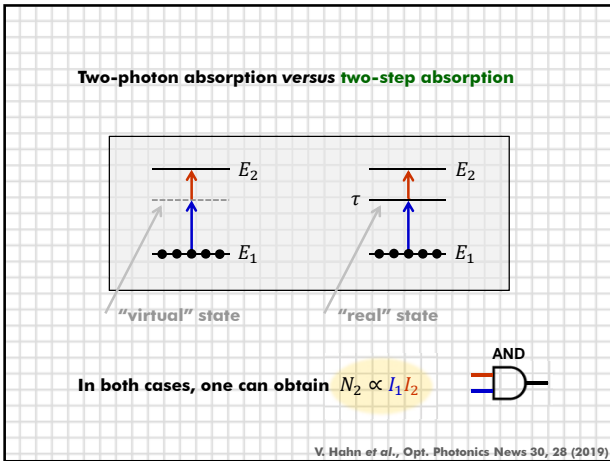


Martin Wegener

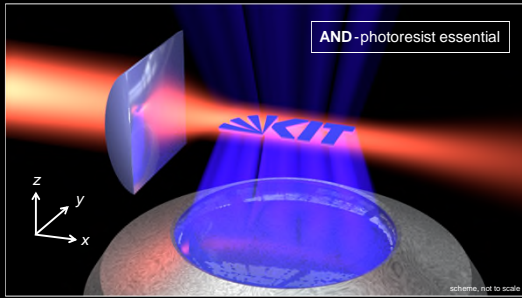
Two-photon absorption versus two-step absorption



Martin Wegener

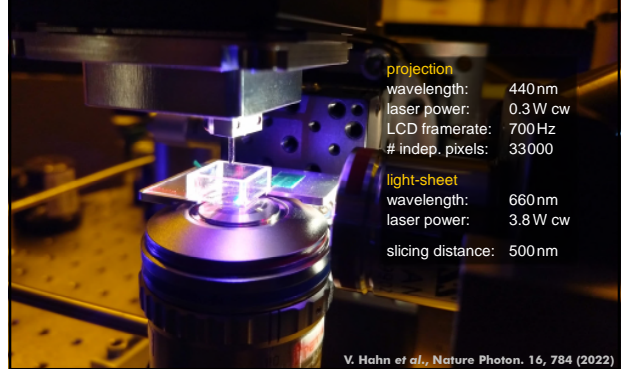


Light-Sheet 3D Laser Microprinting



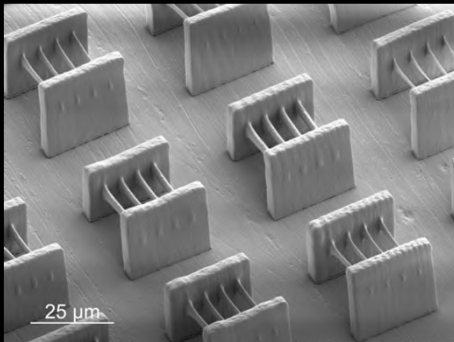
V. Hahn *et al.*, *Opt. Photonics News* 30, 28 (2019)

Our Light-Sheet Setup



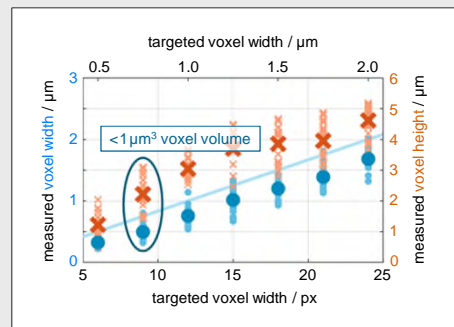
V. Hahn *et al.*, *Nature Photon.* 16, 784 (2022)

Voxel-Size Tests



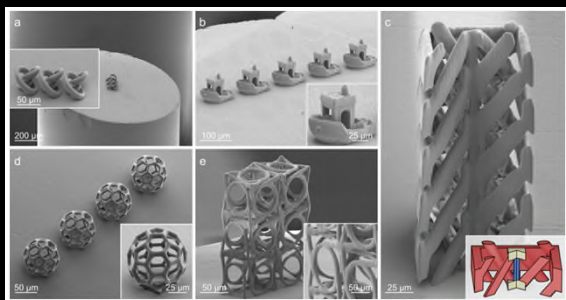
V. Hahn *et al.*, *Nature Photon.* 16, 784 (2022)

Voxel-Size Tests



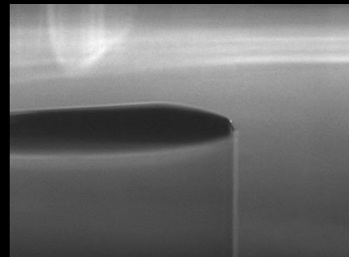
V. Hahn *et al.*, *Nature Photon.* 16, 784 (2022)

SEM Gallery of Examples



panel c, cf.: J.A. Iglesias Martinez *et al.*, *Science Adv.* 7, eabm2189 (2021)

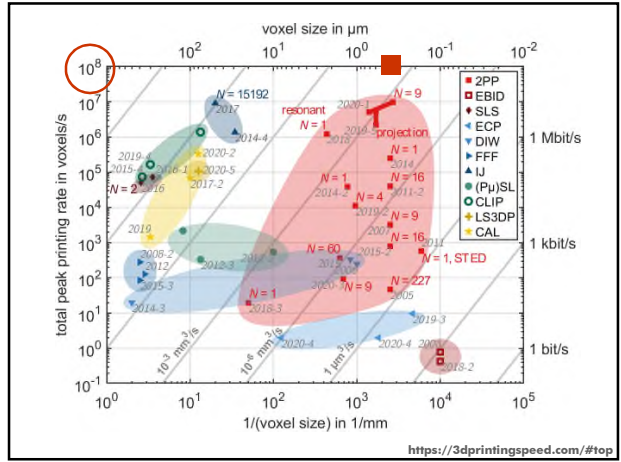
Real-Time Movie



eff. # LCD pixels: 33000
z-scan speed: 0.35 mm/s
printing rate: $7 \cdot 10^6$ voxels/s
time per boat: 266 ms
photoinitiator: biacetyl
scavenger: TEMPO
monomer: DPEHA
development: acetone

V. Hahn *et al.*, *Nature Photon.* 16, 784 (2022)

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More than 350 years ago, Robert Hooke wrote, "As the extension, so the force," when he appreciated how solids deform. This law of linear elasticity applies to all materials and as such constitutes the foundation of solid mechanics. [1]

$$F = Kx$$

Robert Hooke, 1635-1703

[1] C. Coucouzis, Science 358, 994 (2017)

A generalization of Hooke's law is **Cauchy elasticity**. It treats the (meta-) atoms in a (meta-) material as infinitesimally small volume elements, i.e., essentially as points ("point mechanics").

$$\vec{\sigma} = \vec{C} \vec{\epsilon}$$

Louis Cauchy, 1789-1857

up to 21 independent parameters of elasticity tensor

6 Eigenmodes of Deformation

within Cauchy elasticity in three dimensions (3D), also compare Voigt notation

N "Easy" Eigenmodes of Deformation

N	
6	hexamode material
5	pentamode material
4	tetramode material
3	trimode material
2	dimode material
1	monomode material
0	ordinary material

or "floppy" or "soft"; these are extremal materials within Cauchy elasticity

Hexamode metamaterials

Nihilicity, all components of the elasticity tensor are zero

Pentamode metamaterials

G.W. Milton *et al.*, *J. Eng. Mater. Technol.* 117, 483 (1995)

M. Kadic *et al.*, *Appl. Phys. Lett.* 100, 191901 (2012)

T. Bückmann *et al.*, *Nature Commun.* 5, 4130 (2014)

Tetramode metamaterials

M.F. Groß *et al.*, *Adv. Mater.* 35, 2211801 (2023)

Monomode metamaterials

M.F. Groß *et al.*, *Adv. Mater.*, in press (2023)

N=1 ... N=6 zero-mode metamaterials (static only)

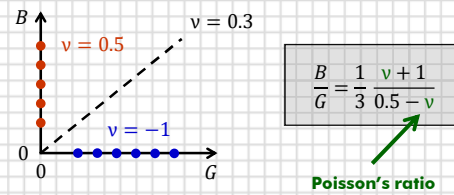
Z. Hu *et al.*, *Nature Commun.* 14, 1266 (2023)

Martin Wegener

Pentamode Metamaterials

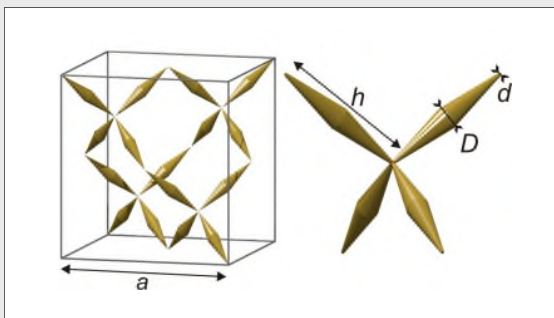
Martin Wegener

Bulk modulus B versus shear modulus G for isotropic pentamode, and auxetic materials ("Milton map")



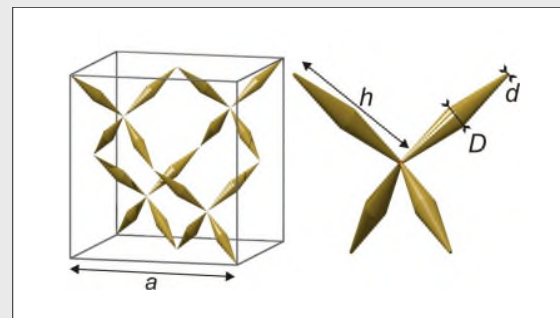
G.W. Milton, *J. Mech. Phys. Solids* 40, 1105 (1992)

Pentamode Metamaterials



















G.W. Milton and A. Cherkov, *J. Eng. Mater. Technol.* 117, 483 (1995)

Pentamode Metamaterials



G.W. Milton, "The Theory of Composites", Cambridge (2002)

Overview: FOM=B/G

	d=0.1μm	d=0.5μm	d=1μm	d=2μm
D=2μm	 47000	 1700	 440	 120
D=3μm	 42000	 1600	 390	 108
D=4μm	 32000	 1320	 350	 96
D=5μm	 29000	 1140	 310	 85

$h=16\ \mu\text{m}$, thus $a=37\ \mu\text{m}$, constituent material nearly irrelevant

Our numerical calculations can be summarized by the approximate **pentamode heuristic formula**

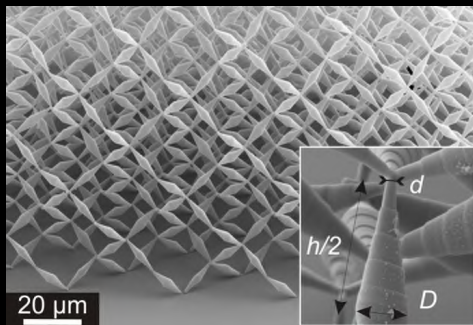
$$\text{FOM} = \frac{B}{G} \approx 0.63 \left(\frac{h}{d}\right)^2 \sqrt{\frac{h}{D}} ; d < D \ll a = \frac{4}{\sqrt{3}}h$$

$$\Rightarrow \nu \approx \frac{1}{2} \left(1 - \left(\frac{B}{G}\right)^{-1}\right)$$

for wavelengths $\lambda \in [\lambda_{\min}, \infty]$; $\lambda_{\min} \gg a$

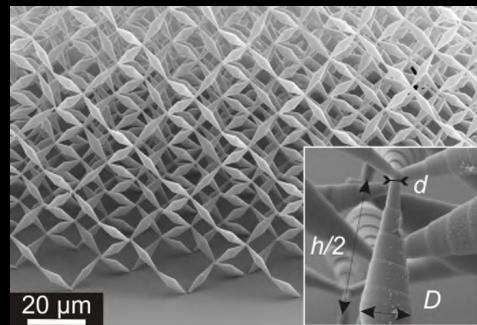
M. Kadic et al., Appl. Phys. Lett. 100, 191901 (2012)

Electron Micrographs



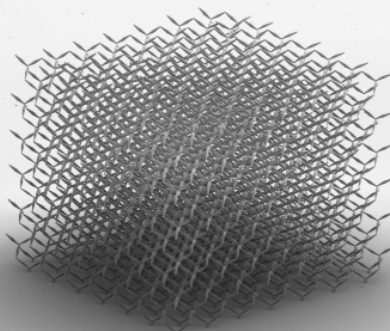
M. Kadic et al., Appl. Phys. Lett. 100, 191901 (2012)

Electron Micrographs



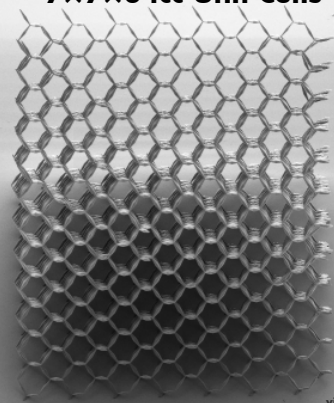
$d=0.55\ \mu\text{m}$, $D=3\ \mu\text{m}$, $h=16.15\ \mu\text{m}$, thus $a=37\ \mu\text{m}$, **volume fill fraction=1.5%**

7×7×6 fcc Unit Cells



$d=1\ \mu\text{m}$, $D=3\ \mu\text{m}$, $h=16\ \mu\text{m}$, thus $a=37\ \mu\text{m}$ and total volume $261\ \mu\text{m} \times 261\ \mu\text{m} \times 224\ \mu\text{m}$

7×7×6 fcc Unit Cells



view along (001) direction

Similar Hinges for Tetramodes ...

Martin Wegener

Hexamode metamaterials

Nihility, all components of the elasticity tensor are zero

Pentamode metamaterials

G.W. Milton *et al.*, *J. Eng. Mater. Technol.* 117, 483 (1995)

M. Kadic *et al.*, *Appl. Phys. Lett.* 100, 191901 (2012)

T. Bückmann *et al.*, *Nature Commun.* 5, 4130 (2014)

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