# Nanophotonics

#### 1. Basics

- **1.1. Analogy between photonics and electronics**
- **1.2. Paradigms of nanophotonics** 
  - **1.2.1. Evanescent waves & the limit of electrostatics**
  - **1.2.2.** Mie-scattering of nanoparticles
  - **1.2.3. Nanoscale metal particles: Particle plasmons**
  - 1.2.4. A nano-aperture in an ideal metal film: Bethe-Bouwkamp theory

#### 2. Photonic band gap (PBG) materials

- 2.1. One-dimensional Photonic Crystals (i.e., dielectric mirrors)
- 2.2. Two-dimensional Photonic Crystals
  - 2.2.1. Negative refraction and the superprism effect
  - 2.2.2. "Semiconductors for light": Waveguides and defect cavities
- 2.3. Three-dimensional Photonic Crystals
  - 2.3.1. Quantum optics: Modified Planck's law
  - 2.3.2. Fabrication: Overview and state-of-the-art
  - 2.3.3. Fabrication: Current efforts within the CFN

#### 3. Metamaterials

- 3.1. Left-handed or Veselago materials (LHM)
- 3.2. "Perfect lenses" made from LHM
- 3.3. Towards metamaterials @ optical frequencies (CFN activities)

#### 4. Plasmonics

- 4.1. Field-enhancement & surface-enhanced Raman scattering (SERS)
- 4.2. Surface-plasmon amplification by stimulated emission of radiation (SPASER)
- **4.3. Transmission through sub-wavelength hole arrays**

## **Textbooks**

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"Principles of Nano-Optics", L. Novotny and B. Hecht,
Cambride University Press (2006)
(best textbook available, in-depth mathematical discussion)
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"Nanophotonics", P.N. Prasad, John Wiley & Sons (2004) (recent comprehensive overview, nothing in depth, good for finding further references and original work)

"Photonic Crystals", J.D. Joannopoulos, R.D. Meade, J.N. Winn, Princeton University Press (nice textbook introduction into the theory, mostly 2D)

"Periodic Nanostructures for Photonics", K. Busch ... M. Wegener, Physics Reports 444, 101-202 (2007) (recent review on photonics crystals and metamaterials)

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## The solutions of problems from nanophotonics ...

## ... are "simply" solutions of the Maxwell equations



## The solutions of problems from nanophotonics ...

## ... are "simply" solutions of the Maxwell equations



see lectures within "Physik II"

## Remember: The free Maxwell equations are scalable.

## This means that if one replaces

 $\vec{r} \rightarrow s\vec{r}$ 

## and at the same time

 $t \rightarrow st$ 

## the solution remains the same.

see lectures within "Physik II"

## Both, electrons and light are waves.

But ... the (single-particle, 1D) Schrödinger equation

$$\mathrm{i}\hbar\frac{\partial}{\partial t}\psi(x,t) = \left(-\frac{\hbar^2}{2m_{\rm e}}\frac{\partial^2}{\partial x^2} + V(x)\right)\psi(x,t)$$

and the (scalar, 1D) wave equation for the electric field

$$\frac{\partial^2}{\partial x^2} E(x,t) - \frac{1}{c^2(x)} \frac{\partial^2}{\partial t^2} E(x,t) = 0$$

seem to be quite different mathematically at first sight.

see "Physik II" and "THEORIE D"

## Just a few simple manipulations ...

## With the usual ansatz for the Schrödinger equation

$$\psi(x,t) = \psi(x) \exp(-i E/\hbar t)$$

## and the ansatz for the wave equation

$$E(x,t) = E(x) \exp(-i\omega t) + c.c.$$

## ... and we get ...

## ... strictly the identical mathematical form

for the stationary Schrödinger equation

$$\frac{\partial^2}{\partial x^2}\psi(x) + \left(\frac{2m_{\rm e}}{\hbar^2}\left(\mathrm{E}-V(x)\right)\right)\psi(x) = 0$$

0

and for the light field

=: a(x)

$$\frac{\partial^2}{\partial x^2} E(x) + \left(\frac{\omega^2 n^2(x)}{c_0^2}\right) E(x) =$$

with the refractive index  $n(x) = \frac{c_0}{c(x)}$ .

## Beyond this "piecewise-constant", one-dimensional

& scalar case, the analogy between electron waves

and light waves is no longer complete because ...

## ... optical materials can be inhomogeneous

$$H(r,t) = H(r) \exp(-i\omega t) + \text{c.c.}$$
$$\frac{1}{\mu(\vec{r})} \vec{\nabla} \times \left(\frac{1}{\varepsilon(\vec{r})} \vec{\nabla} \times \vec{H}(\vec{r})\right) = \frac{\omega^2}{c_0^2} \vec{H}(\vec{r})$$

 $\rightarrow$ 

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## Beyond this "piecewise-constant", one-dimensional

- & scalar case, the analogy between electron waves
- and light waves is no longer complete because ...

- ... optical materials can be inhomogeneous
- ... light waves are vector waves (polarization)
- ... and have both an electric and a magnetic component
- ... optical materials can be anisotropic or birefringent

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With decreasing a, the decay is getting more rapid.



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Lowest diffracted order becomes evanescent for  $|a < \lambda|$  .



With decreasing a, the decay is getting more rapid.



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## **Consider total internal reflection.**



## **Consider total internal reflection.**



## **Consider total internal reflection.**





one can again apply the rules of electrostatics !

## Here, the wavelength can be considered as infinitely

## large. According to the dispersion relation of light,

$$c_0 = \frac{\omega}{|\vec{K}|} = \frac{2\pi f}{2\pi / \lambda} = f \lambda$$

the frequency is approximately zero.

## For very small length scales, i.e., for $|a| << \lambda|$ ,

## one can again apply the rules of electrostatics !





simulation for nano-brick with n=1.5, z=5nm, 60nm x 120nm x 40nm @ 633nm wavelength

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## Consider scattering off a small dielectric sphere in air.

# From a multipole expansion one gets, e.g., the scattering cross section (after a lenghty derivation)

$$C_{n} = \frac{2\pi}{K^{2}} \sum_{n=1}^{\infty} (2n+1)(|a_{n}|^{2} + |b_{n}|^{2})$$

$$E_{p}$$

$$K = 2\pi/\lambda$$

## with the coefficients

$$a_{n} = \frac{\sqrt{\varepsilon_{p}}\psi_{n}(\sqrt{\varepsilon_{p}}Kr_{p})\psi'_{n}(Kr_{p}) - \psi_{n}(Kr_{p})\psi'_{n}(\sqrt{\varepsilon_{p}}Kr_{p})}{\sqrt{\varepsilon_{p}}\psi_{n}(\sqrt{\varepsilon_{p}}Kr_{p})\xi'_{n}(Kr_{p}) - \zeta_{n}(Kr_{p})\psi'_{n}(\sqrt{\varepsilon_{p}}Kr_{p})}$$
  
$$b_{n} = \dots$$
  
**n-th order Riccati-Bessel functions**

G. Mie, Ann. d. Physik 25, 377 (1908)

## Consider scattering off a small dielectric sphere in air.

From a multipole expansion one gets the vectorial electric field as well ...

## A readable discussion can be found in



- C.F. Bohren and D.R. Huffman, "Absorption and scattering of light by small particles", John Wiley & Sons, 1983
- H.C. van de Hulst,
   "Light scattering by small particles",
   Dover, 1981

G. Mie, Ann. d. Physik 25, 377 (1908)

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## Consider a small metallic sphere in vacuum.

#### A reminder on the electrostatic depolarization factor:



## Consider a small metallic sphere in vacuum.

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### Consider a small metallic sphere in vacuum.

#### A reminder on the electrostatic depolarization factor:



### Consider a small metallic sphere in vacuum.

#### Light field excites within the long-wavelength limit:



**Mathematica graphics here** 

### Consider a small metallic sphere in vacuum.

The optical response of a bulk metal can be described by the Drude model.

For a metal nanoparticle, one rather gets a Lorentz oscillator response.

The position of this particle plasmon resonance is also determined by the embedding dielectric medium and by the particle size & shape.

$$\varepsilon(\omega) = -2\varepsilon_{\rm med}$$

### Measurements on gold nanoparticles on ITO/glass:



Stefan Linden, Institut für Nanotechnologie





**British museum** 





Ag and Au nanoparticles

## Glass doped with metal nanocrystals



church window from Sint Jan in Gouda (Netherlands)

## **Exercise:** Consider a thin (infinite) sheet of metal.

## Where does the resonance occur? For what polarization?





## **Exercise:** Consider a thin (infinite) sheet of metal.

### Where does the resonance occur? For what polarization?

# <u>Solution:</u> If the electric field is perpendicular to the plane of the sheet, we have

$$\hat{N}_{\rm dp} = 1 \Longrightarrow \varepsilon(\omega) = 0 \Longrightarrow \omega = \omega_{\rm pl}$$

## If the electric field lies in the plane: No resonance, as

$$\hat{N}_{\rm dp} = 0$$

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# Consider a plane wave impinging onto a small circular hole in an infinitely thin film of an ideal metal.

This problem was solved by Bethe in 1944. In 1950, technical mistakes were eliminated by Bouwkamp. The **"Bethe-Bouwkamp theory"** is the exact analytical solution of this model problem.

It is relevant for scanning near-field optical microscopy (SNOM), which uses apertures in metal films.

H.E. Bethe, Phys. Rev. 66, 163 (1944) & C.J. Bouwkamp, Philips Res. Rep. 5, 321 (1950)

# Consider a plane wave impinging onto a small circular hole in an infinitely thin film of an ideal metal.





aperture diameter = 50 nm
wavelength = 500 nm
movie from 50 nm to zero distance
the square modulus of the electric field is shown on a false-color scale

exponential decay versus z with a decay length given by 0.2 diameter
transmitted power scales with the sixth power of the diameter

animation by Andreas Naber, Institut für Angewandte Physik

## **Actual SNOM tips**



Ulrich Neuberth & Nicole Rau, Institut für Angewandte Physik

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## Silicon, a semiconductor crystal



## Silicon, a semiconductor crystal



## Silicon, a semiconductor crystal



## Is there such a thing as a "semiconductor for light" ?

S. John, Phys. Rev. Lett. 58, 2586 (1987) E. Yablonovitch, Phys. Rev. Lett. 58, 2059 (1987)

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In the 1D case with piecewise-constant refractive index, the analogy between the band structure of electrons and that of light is strict (see 1.1.).

see: Bloch waves for electrons

see: Kronig-Penney model for electrons

see: tight-binding model for electrons

see: nearly-free electron model for electrons

N.W. Ashcroft and N.D. Mermin, "Solid State Physics", Harcourt College Publishers

## Consider a 1D Photonic Crystal (a dielectric mirror).









$$\mathcal{E}_1 \approx \mathcal{E}_2$$







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## **2D Photonic Crystals in nature**



Taken from: A.R. Parker et al., Nature 409, 36 (2001)

## **2D** hexagonal PC structure



Taken from: R. Wehrspohn, U. Gösele et al., MPI in Halle

### Consider a Photonic Crystal with a 2D square lattice.











# One gets **complex iso-frequency curves** – especially for the higher bands at larger frequencies.



# One gets complex iso-frequency curves – especially for the higher bands at larger frequencies.



### for band structure calculations

One gets complex iso-frequency curves – especially for the higher bands at larger frequencies.

This is simply the result of back-folding into the 1. Brillouin zone.

At the same time, one gets flat bands with small group velocities (e.g., along the  $\Gamma M$  direction)

$$\vec{v}_{\text{group}} = \vec{\nabla}_{\vec{K}} \, \omega$$
Actual band structure calculations have to solve the eigenvalue problem (see 1.1.).

$$\frac{1}{\mu(\vec{r})}\vec{\nabla} \times \left(\frac{1}{\varepsilon(\vec{r})}\vec{\nabla} \times \vec{H}(\vec{r})\right) = \hat{O}(\vec{r})\vec{H}(\vec{r}) = \frac{\omega^2}{c_0^2}\vec{H}(\vec{r})$$

### **Translational symmetry means that**

$$\mathcal{E}(\vec{r}) = \mathcal{E}(\vec{r} + \vec{T})$$
 and  $\mu(\vec{r}) = \mu(\vec{r} + \vec{T})$ 

### hence

$$\hat{O}(\vec{r}) = \hat{O}(\vec{r} + \vec{T})$$

often = 1

Actual band structure calculations have to solve the eigenvalue problem (see 1.1.).

$$\frac{1}{\mu(\vec{r})}\vec{\nabla} \times \left(\frac{1}{\varepsilon(\vec{r})}\vec{\nabla} \times \vec{H}(\vec{r})\right) = \hat{O}(\vec{r})\vec{H}(\vec{r}) = \frac{\omega^2}{c_0^2}\vec{H}(\vec{r})$$

### **Translational symmetry delivers the Bloch theorem**

$$\vec{H}(\vec{r}) = \vec{h}_{\vec{k}}(\vec{r}) \exp(i\vec{K}\cdot\vec{r}) \neq \vec{H}(\vec{r}+\vec{T})$$
$$\vec{h}_{\vec{k}}(\vec{r}) = \vec{h}_{\vec{k}}(\vec{r}+\vec{T})$$

Use, e.g., plane-wave expansion method



**Martin Wegener** 

# Actual band structure calculations using the plane-wave expansion method can be performed by everybody using the "MIT package".

### It can be downloaded (free of cost) from

http://ab-initio.mit.edu/mpb/

S.G. Johnson and J.D. Joannopoulos, Optics Express 8, 173 (2001)

### **Consider a 2D hexagonal Photonic Crystal.**

# For example, air cylinders in a dielectric, or vice versa (see sea-mouse or "Halle" structures).



#### **Important parameters:**

- r/a ratio
- refractive index (here 1.6)

### **Consider a 2D hexagonal Photonic Crystal.**

### (a) Real space lattice

### (b) 1. Brillouin zone





**Martin Hermatschweiler** 

### **Consider a 2D hexagonal Photonic Crystal.**

### Calculated band structure for H and for E-polarisation



#### Martin Hermatschweiler & the MIT package

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#### 2.2.1. Negative refraction and the superprism effect

- 2.2.2. "Semiconductors for light": Waveguides and defect cavities
- **2.3. Three-dimensional Photonic Crystals** 
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### **Photonic Crystal**





air/vacuum

### **Photonic Crystal**





 tangential component of the wavevector is conserved

\*\*\*\*\*

- frequency is conserved
- look at corresponding iso-frequency curve (analogy: Fermi surface)

Martin Wegener







# **Refraction at an interface** air/vacuum **Photonic Crystal** $\frac{\omega}{|\vec{K}|} = c_0$ ..... $\vec{E}$ $\vec{v}_{\text{group}} \parallel \vec{S}$ $\vec{K}, \vec{S}$ $\vec{B}$ $\vec{v}_{\text{group}} = \vec{\nabla}_{\vec{K}} \, \omega$ **Martin Wegener**

The result is negative refraction, i.e., refraction that looks as if the refractive index in Snell's law would be negative.

$$\frac{\sin(\alpha_{\rm vac})}{\sin(\alpha_{\rm med})} = n$$

The angle inside the medium can be a very sensitive function of the incident (vacuum) angle.

The angle inside the PC also sensitively depends on the frequency via the dependence of the shape of the iso-frequency curve on frequency.

The latter effect can be used as a "superprism".

H. Kosaka et al., Phys. Rev. B 58, R10096 (1998)

### The phenomenon of negative refraction ...

- ... can occur in Photonic Crystals with positive permittivity and unity permeability. It is a result of Bragg reflection (this section).
- ... can occur in left-handed metamaterials with negative permittivity and negative permeability. Bragg reflection plays no role (section 3.1.).

# **Self-collimation**



**Martin Wegener** 

# **Self-collimation**



**Martin Wegener** 

For example, for a tight-binding band structure and for a frequency in the middle of the band, the 2D iso-frequency contour can be a square.

A tight focus on the PC surface corresponds to a large spread of wave vectors of light (and of the Poynting vectors) in air.

Inside the PC, all Poynting vectors point in <u>one direction</u> – while the tight spatial focus remains (not possible in air due to diffraction).

This remarkable phenomenon is often refered to as self-collimation or as diffraction-less flow of light.

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# Guiding light around the corner



Kurt Busch

# **2D** hexagonal PC structure



Taken from: R. Wehrspohn, U. Gösele et al., MPI in Halle

# **2D PC slab waveguides**



Taken from: S. Noda et al., Nature 407, 608 (2000)

### Note that the physics of 2D PC slab waveguides ...

- ... is different from that of an ideal 2D PC as some of the modes can escape from the slab, others cannot (total internal reflection).
- ... is important because such structures are much more relevant for applications (light is guided in the third dimension as well).

J.D. Joannopoulos et al., "Photonic Crystals", Princeton University Press

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## **3D Photonic Crystals in nature**





Morpho Rhetenor und Parides Sesostris

Overview: P. Vukusic and J.R. Sambles, Nature 424, 852 (2003)

## **3D Photonic Crystals in nature**



**Pachyrhynchus Argus** 

Overview: P. Vukusic and J.R. Sambles, Nature 424, 852 (2003)



# **Opals: 3D Photonic Crystals**





Taken from: eBay.com

# A closer look at an Opal





# Silicon-based inverse Opals



**Kurt Busch** 

Actual band structure calculations have to solve the eigenvalue problem (see 1.1. and 2.2.).

$$\frac{1}{\mu(\vec{r})}\vec{\nabla} \times \left(\frac{1}{\varepsilon(\vec{r})}\vec{\nabla} \times \vec{H}(\vec{r})\right) = \hat{O}(\vec{r})\vec{H}(\vec{r}) = \frac{\omega^2}{c_0^2}\vec{H}(\vec{r})$$

### **Translational symmetry delivers the Bloch theorem**

$$\vec{H}(\vec{r}) = \vec{h}_{\vec{K}}(\vec{r}) \exp(i\vec{K}\cdot\vec{r}) \neq \vec{H}(\vec{r}+\vec{T})$$
$$\vec{h}_{\vec{K}}(\vec{r}) = \vec{h}_{\vec{K}}(\vec{r}+\vec{T})$$

Use, e.g., plane-wave expansion method



Same mathematics as in 2D

### Today, <u>complete</u> three-dimensional photonic band gaps have been found for the following crystal symmetries:

- simple cubic (sc)
- base centered cubic (bcc)
- face centered cubic (fcc)
- diamond
- rhombohedral

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# **3D** metallic PC as light emitters



Idea & IR experiment: J.G. Fleming et al., Nature 417, 52 (2002)

# **3D** metallic PC as light emitters



Idea & IR experiment: J.G. Fleming et al., Nature 417, 52 (2002)

# **2.** law of thermodynamics ...

The PC can have 99.9% efficiency (opt. out/electr. in) rather than just 3% for an ideal black body at T=3000K.

However, it cannot emit more light in a spectral interval than an ideal black body held at the same temperature.



P. Würfel et al., Appl. Phys. Lett. 84, 1997 (2004)

# Consider a **3D** sc lattice of air cubes, each coated with a thin film of an ideal metal.

### Solutions are standing waves within the cubes

$$\mathsf{DOS}(\omega) \propto \sum_{N_x N_y N_z} \delta(\omega - \omega_{N_x N_y N_z})$$

### **Eigenfrequencies** from the dispersion relation of light:

$$c_0 = \frac{\omega_{N_x N_y N_z}}{|\vec{K}|} = \frac{\omega_{N_x N_y N_z}}{\sqrt{\left(N_x \frac{\pi}{a}\right)^2 + \left(N_y \frac{\pi}{a}\right)^2 + \left(N_z \frac{\pi}{a}\right)^2}}$$

**Martin Wegener**
# Consider a **3D** sc lattice of air cubes, each coated with a thin film of an ideal metal.

#### Solutions are standing waves within the cubes

$$\mathsf{DOS}(\omega) \propto \sum_{N_x N_y N_z} \delta(\omega - \omega_{N_x N_y N_z})$$

#### **Eigenfrequencies:**

$$\omega_{111} = \sqrt{3} c_0 \frac{\pi}{a}$$
;  $\omega_{112} = \omega_{121} = \omega_{211} = \sqrt{6} c_0 \frac{\pi}{a}$ 

**Martin Wegener** 

. . .

# Consider a **3D** sc lattice of air cubes, each coated with a thin film of an ideal metal.

#### Solutions are standing waves within the cubes

$$\mathsf{DOS}(\omega) \propto \sum_{N_x N_y N_z} \delta(\omega - \omega_{N_x N_y N_z})$$

**Eigenfrequencies**, e.g., in the visible (red):

$$\hbar \omega_{111} = 2 \text{ eV} (\lambda = 620 \text{ nm}) \implies a = \frac{\lambda \sqrt{3}}{2} = 536 \text{ nm}$$

**Martin Wegener** 

# Consider a 3D sc lattice of air cubes, each coated with a thin film of an ideal metal.

#### Solutions are standing waves within the cubes

$$\mathsf{DOS}(\omega) \propto \sum_{N_x N_y N_z} \delta(\omega - \omega_{N_x N_y N_z})$$

#### **Eigenfrequencies**, e.g., in the visible (red):



#### i.e., monochromatic emission!

**Martin Wegener** 

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## **3D woodpile structure**



Taken from: J.G. Fleming et al., Opt. Lett. 24, 49 (1999)

## **3D** silicon-based inverted opal



Taken from: A. Blanco et al., Nature 405, 437 (2000) & K. Busch

## **3D** silicon-based inverted opal



Taken from: D.J. Norris et al., Nature 414, 289 (2001)

## **3D** square-spiral structure



Taken from: S. John et al., Science 292, 1133 (2001)

#### **3D** square-spiral structure



Taken from: S.R. Kennedy, S. John et al., Nano Letters 2, 59 (2002)

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#### - Direct laser writing

**Martin Wegener** 

#### - Direct laser writing

**Martin Wegener** 



G. Grynberg et al., Phys. Rev. Lett. 70, 2249 (1993)

$$I(\vec{r}) \propto \left| \sum_{n=1}^{4} \vec{E}_n \exp(i(\vec{k}_n \cdot \vec{r} - \omega t)) \right|^2$$

$$I(\vec{r}) \propto \left| \sum_{n=1}^{4} \vec{E}_n \exp(i(\vec{k}_n \cdot \vec{r} - \omega t))) \right|^2$$
$$= \sum_{m=1}^{4} \sum_{l=1}^{4} a_{ml} \exp(i \vec{G}_{ml} \cdot \vec{r})$$
$$a_{ml} = \vec{E}_m \cdot \vec{E}_l^* \qquad \vec{G}_{ml} = \vec{k}_m - \vec{k}_l$$



## **Iso-intensity surfaces**

# <u>500nm</u> 300nm Daniel Meisel & Martin Wegener

## Influence of exposure intensity





## **Different polarizations ...**



#### A look at the experimental setup

- frequency-tripled, single-mode
  Nd:YAG laser at 355nm wavelength
  (DFG-Leibniz)
- exposure of resist SU-8 with a single
  6ns pulse at 8mJ total energy
- about 1 cm<sup>2</sup> exposed area
- all four polarizations and intensities can be adjusted independently

Daniel Meisel, Juri Miklaev & Martin Wegener

## **Experimental results**



#### Daniel Meisel, Juri Miklaev & Martin Wegener

#### Working against refraction ...



M. Campbell et al., Nature 404, 53 (2000) X. Wang et al., Appl. Phys. Lett. 82, 2212 (2003)

## Working against refraction ...



J.M., D.M. & M.W., Patentanmeldung 10233309.2 (INT, FZK)

#### A fcc Photonic Crystal



#### A fcc Photonic Crystal



## A fcc Photonic Crystal



#### **Optical characterization**



#### **Comparison with theory**





#### "two-planes" and "umbrella-like" geometry



M. Maldovan et al., PRB 65, 165123 (2002) & S. John et al., PRL 92, 043905 (2004)



D.C. Meisel et al., Phys. Rev. B 70, 165104 (2004)

$$I(\vec{r}) \propto \left| \sum_{n=1}^{4} \vec{E}_{n} \exp(i(\vec{k}_{n} \cdot \vec{r} - \omega t))) \right|^{2}$$
  
=  $\sum_{m=1}^{4} \sum_{l=1}^{4} a_{ml} \exp(i \vec{G}_{ml} \cdot \vec{r})$   
 $a_{ml} = \vec{E}_{m} \cdot \vec{E}_{l}^{*}$   $\vec{G}_{ml} = \vec{k}_{m} - \vec{k}_{l}$ 

- fcc translational symmetry
- rhombohedral crystal symmetry
- 5.8% gap/midgap PBG between 2nd and 3rd band for 37% silicon
- resembles celebrated Yablonovite
- 10:1 interference contrast








#### Example II

- sc translational symmetry
- sc crystal symmetry
- 11% gap/midgap PBG between
  5th and 6th band for 24.4% silicon
- "very hard to make"



Daniel Meisel, Martin Wegener & Kurt Busch

#### - Holographic lithography

- Direct laser writing

**Martin Wegener** 

### **2D Electron-beam lithography**



used in, e.g., Science 306, 1351 (2004)

## polymer – SiO<sub>2</sub> – Si



N. Tétreault et al., Adv. Mater. 18, 457 (2006)

### Direct laser writing for waveguides ...







For DLW-technique see: S. Kawata et al., Nature 412, 697 (2001)

## Direct laser writing for waveguides ...

## The building block







Intensity (normalized

## The building block







Intensity (normalized









fcc for  $(c/a)^2=2$ , full gap for index contrast > 1.9, 25% gap for holes in Si

Proposal: C.M. Soukoulis et al., Solid State Commun. 89, 413 (1994)

## **Experimental results ...**















10 µm

M. Deubel et al., Nature Materials 3, 444 (2004)

## Analysis of transmission spectra



M. Deubel et al., Appl. Phys. Lett. 87, 221104 (2005)







## Laue diffraction (normal)

10 µm

anaanaanaanaanaan

## Lave diffraction (normal)



## Laue diffraction (side)



# $SiCl_4 + 2 H_2O \rightarrow SiO_2 + 4 HCl$

**Martin Hermatschweiler** 



11at

**Martin Hermatschweiler** 

# Resulting silicon woodpiles ...



N. Tétreault et al., Adv. Mater. 18, 457 (2006)

## ... or inverse silicon woodpiles



## ... or inverse silicon woodpiles



## ... or inverse silicon woodpiles



#### 14% PBG @ 2.5 µm achieved



## 20% PBG @ 1.5 µm in reach



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## "Normal" crystals ...



... have lattice constants much smaller than the wavelength of light

... are common optical materials; they have a refractive index n>0

Natur-Lexikon.com

### Metamaterials ...



... have lattice constants smaller than the wavelength of light

... can be left-handed, i.e., n<0, which is the basis for, e.g., "perfect lenses"

R.A. Shelby et al., Science 292, 77 (2001)

### Photonic Crystals ...



... have lattice constants comparable to the wavelength of light

... can be "semiconductors for light" (see section 2.)

S. John, Phys. Rev. Lett. 58, 2586 (1987) E. Yablonovitch, Phys. Rev. Lett. 58, 2059 (1987)

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### Plane waves ...

... are solutions of the Maxwell equations





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### Plane waves ...

### ... are characterized by their phase velocity c with

$$c^{2} = \frac{1}{\varepsilon_{0}\varepsilon\mu_{0}\mu} = \frac{c_{0}^{2}}{\varepsilon\mu} = \frac{c_{0}^{2}}{n^{2}}$$

 $n^2 = \varepsilon \mu \implies n = \pm \sqrt{\varepsilon \mu}$ 

### ... and by their impedance

$$Z = \sqrt{\frac{\mu_0 \mu}{\varepsilon_0 \varepsilon}} \qquad \qquad Z_0 = \sqrt{\frac{\mu_0}{\varepsilon_0}} \approx 376.7 \,\Omega$$

Institut für Angewandte Physik

### Planes waves in normal media

 $\vec{K}, \vec{S}$ 

 $\mathbf{E}$  $\vec{B}$ 



Martin Wegener

$$\varepsilon = \mu = n = 1$$

$$\overline{\varepsilon} > 1, \mu = 1$$

$$\overline{k}, \overline{S}$$

$$\overline{B}$$

$$\overline{K}, \overline{S}$$

$$\overline{B}$$

$$\overline{S}$$

$$\overline{S$$

**Snell's law of refraction** 

$$\varepsilon = \mu = n = 1$$

$$\vec{c} = \vec{c} = \vec{c}$$

Snell's law of refraction

$$\varepsilon = \mu = n = 1$$

$$\varepsilon = \mu = -1 \Rightarrow Z = \sqrt{\frac{\mu_0 \mu}{\varepsilon_0 \varepsilon}} = Z_0$$

$$\vec{E}$$

$$\vec{K}, \vec{S}$$

$$\vec{B}$$

$$\vec{K}, \vec{S}$$

$$\vec{R}$$

$$\vec{K}$$

$$\vec{B}$$

$$\vec{S}$$

$$\vec{R}$$

$$\vec{K}$$

$$\vec{F}$$

$$\vec{K}$$

$$\vec{F}$$

$$\vec{K}$$

$$\vec{F}$$

$$\vec{K}$$

$$\vec{F}$$

$$\varepsilon = \mu = n = 1$$

$$\varepsilon = \mu = -1 \Rightarrow Z = \sqrt{\frac{\mu_0 \mu}{\varepsilon_0 \varepsilon}} = Z_0$$

$$\vec{R}$$

$$\vec{K}, \vec{S}$$

$$\vec{K}$$

$$\vec{E}$$

$$\vec{K}$$

$$\vec{E}$$

$$\vec{K}$$

$$\vec{E}$$

$$\vec{S}$$

$$\vec{R}$$

# Materials with n>0 are right-handed

 $\vec{E}$ 

 $\vec{S}$ 

 $\vec{B}$ 

**Martin Wegener** 

# n<0: Left-handed materials (LHM)





V.G. Veselago, Sov. Phys. Usp. 10, 509 (1968)

### The phenomenon of negative refraction ...

- ... can occur in Photonic Crystals with positive permittivity and unity permeability. It is a result of Bragg reflection (section 2.2.1.).
- ... can occur in left-handed metamaterials with negative permittivity and negative permeability. Bragg reflection plays no role (this section).

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#### 3.2. "Perfect lenses" made from LHM

- 3.3. Towards metamaterials @ optical frequencies (CFN activities)
- 4. Plasmonics
  - 4.1. Field-enhancement & surface-enhanced Raman scattering (SERS)
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  - **4.3. Transmission through sub-wavelength hole arrays**

### A "perfect lens" from a LHM



### **First left-handed materials**

### 10 GHz (3 cm)



### Science magazine:

"LHM one of the top ten scientific breakthroughs in 2003"

**R.A. Shelby et al., Sci**ence **292, 77 (2001)** 

Editorial staff, Science 302, 2039 (2003)

Confirmation: A.A. Houck et al., Phys. Rev. Lett. 90, 137 (2003)

### **First left-handed materials**

### 10 GHz (3 cm)



### 1 THz (300 μm)



**R.A. Shelby et al., Sci**ence **292, 77 (2001)** 

**T.J.** Yen et al., Science 303, 1494 (2004)

**Review: D.R. Smith et al., Science 305, 788 (2004)** 

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### - Diamagnetism in optics

### - Negative refractive index

**Martin Wegener** 

### - Diamagnetism in optics

### - Negative refractive index

**Martin Wegener** 

# LC-circuits @ 100 THz (3 µm)



Stefan Linden, Christian Enkrich & Martin Wegener

# **Estimating the LC-resonance**



$$L = \mu_0 \frac{l^2}{t} = 5.6 \text{ pH}$$
$$C = \varepsilon_0 \varepsilon_C \frac{wt}{d} = 0.5 \text{ aF}$$
$$\omega_{LC} = \frac{1}{\sqrt{LC}} \approx 2\pi 100 \text{ THz}$$

$$\Rightarrow \lambda_{LC} = l \ 2\pi \sqrt{\varepsilon_C} \sqrt{\frac{w}{d}} \approx 3 \ \mu m$$

Stefan Linden, Christian Enkrich & Martin Wegener

# **Magnetic permeability**



$$U_{C} + U_{L} = U_{ind} = -\dot{\phi}$$
$$\Rightarrow I(t) \Rightarrow l^{2}I(t) \Rightarrow M(t)$$
$$0 \le F := \frac{l^{2}t}{a_{xy} a_{z}} \le 1$$

$$\Rightarrow \mu(\omega) = 1 + \frac{F \,\omega^2}{\omega_{LC}^2 - \omega^2}$$

**Martin Wegener** 

# **Polarization dependence**



Coupling to the LC-resonance & magnetic response but not accessible for normal incidence.

N. Katsarakis et al., Appl. Phys. Lett. 84, 2943 (2004)

# **Polarization dependence**





N. Katsarakis et al., Appl. Phys. Lett. 84, 2943 (2004)

# **Polarization dependence**





N. Katsarakis et al., Appl. Phys. Lett. 84, 2943 (2004)

### FT-IR microscope-spectrometer

-Action

0 6

affer

EQUINOX 55

-

Funded by DFG-Leibniz award 2000

191

NANANA SUSS















**Theory: Iowa State** 



**Theory: Iowa State** 

# Numerical calculations (E-field)

### @ fundamental magnetic resonance



**3D-FDTD** calculations with CST Microwave Studio

### **Retrieved metamaterial parameters**



Jiangfeng Zhou, Thomas Koschny & Costas M. Soukoulis

# Focused-ion-beam writing (FIB)



FIB writing

**Christian Enkrich** 

# Focused-ion-beam writing (FIB)


# "Rapid prototyping" with FIB



20 min. fabrication time

### **Mie resonance - SRR**



C. Enkrich et al., Adv. Mater. 17, 2547 (2005)

## **Electron-beam lithography**



**Stefan Linden** 

### Magnetic resonance @ 1.5 µm

# инининии нининини **UNHUNNUN** LLLLLL 50 nm LLLLL 400 nm 200 nm

30nm Au on glass, e-beam lithography

## Magnetic resonance @ 1.5 µm



retrieval yields  $\mu < 0 @ 1.5 \mu m$ 

# SRR size scaling for ever?



$$L = \mu_0 \frac{l^2}{t} \propto \text{size}$$
$$C = \varepsilon_0 \varepsilon_C \frac{wt}{d} \propto \text{size}$$
$$\omega_{LC} = \frac{1}{\sqrt{LC}}$$
$$\Rightarrow \omega_{LC} \propto \frac{1}{\sqrt{LC}}$$

H.J. Schneider and P. Dullenkopf, Rev. Sci. Instrum. 48, 68 (1977)

size

# Limits of size scaling



$$E_{\rm kin} = N_{\rm e} \frac{m_{\rm e}}{2} v_{\rm e}^2 = \frac{1}{2} L_{\rm kin} I^2$$
$$L_{\rm kin} = \frac{m_{\rm e}}{n_{\rm e}} \frac{4(l-w) - d}{wt}$$

$$\Rightarrow \omega_{LC} \propto \frac{1}{\sqrt{\text{size}^2 + \text{const.}}}$$

J. Zhou et al., Phys. Rev. Lett. 95, 223902 (2005)

# Limits of size scaling



$$E_{\rm kin} = N_{\rm e} \frac{m_{\rm e}}{2} v_{\rm e}^2 = \frac{1}{2} L_{\rm kin} I^2$$
$$L_{\rm kin} = \frac{m_{\rm e}}{n_{\rm e}} \frac{4(l-w) - d}{wt}$$

$$\Rightarrow \omega_{LC}^{\max} \approx \omega_{\rm pl} \sqrt{\frac{d}{4l}}$$

A. Ishikawa et al., Phys. Rev. Lett. 95, 237401 (2005)

# **Ohmic damping & size scaling**



$$L = \mu_0 \frac{l^2}{t} \propto \text{size}$$
$$C = \varepsilon_0 \varepsilon_C \frac{wt}{d} \propto \text{size}$$
$$R = \frac{1}{\sigma} \frac{4(l-w) - d}{wt} \propto \frac{1}{\text{size}}$$

and

$$\ddot{I} + \frac{R}{L}\dot{I} + \frac{1}{LC}I = 0$$

# **Ohmic damping & size scaling**



$$L = \mu_0 \frac{l^2}{t} \propto \text{size}$$
$$C = \varepsilon_0 \varepsilon_C \frac{wt}{d} \propto \text{size}$$
$$R = \frac{1}{\sigma} \frac{4(l-w) - d}{wt} \propto \frac{1}{\text{size}}$$

$$\Rightarrow \frac{\text{damping}}{\text{frequency}} = \frac{R/L}{\frac{1}{\sqrt{LC}}} \propto \frac{1}{\text{size}}$$

# Size scaling, constant thickness



$$L = \mu_0 \frac{l^2}{t} \propto (\text{size})^2$$
$$C = \varepsilon_0 \varepsilon_C \frac{wt}{d} \propto \text{const.}$$
$$R = \frac{1}{\sigma} \frac{4(l-w) - d}{wt} \propto \text{const.}$$

$$\Rightarrow \frac{\text{damping}}{\text{frequency}} = \frac{R/L}{\frac{1}{\sqrt{LC}}} \propto \frac{1}{\text{size}}$$

### Limits of size scaling



M.W. Klein et al., Opt. Lett. 31, 1259 (2006)



### Limits of size scaling



M.W. Klein et al., Opt. Lett. 31, 1259 (2006)

#### Limits of size scaling



M.W. Klein et al., Opt. Lett. 31, 1259 (2006)

### **Metal plasma frequencies**

gold	Αυ	2081 THz	8.5 eV
silver	Ag	2182 THz	9.0 eV
aluminum	AI	3231 THz	13.2 eV
beryllium	Be	4800 THz	20 eV

Au & Ag from Drude fits to: P.B. Johnson et al., Phys. Rev. B 6, 4370 (1972) Al from Drude fit to: E.D. Palik, Handbook of Optical Constants, Academic Press

### ... brings us to cut-wire pairs



Theory: A.N. Lagarkov et al., Phys. Rev. B 53, 6318 (1996)

## **Calculated electric fields**

#### Short-wavelength resonance

#### Long-wavelength resonance





l=500nm, w=150nm, d=80nm











G. Dolling et al., Opt. Lett. 30, 3198 (2005)



G. Dolling et al., Opt. Lett. 30, 3198 (2005)

# **Alternative orientation**



A.N. Grigorenko et al., Nature 438, 335 (2005)

# Holographic lithography

N. Feth et al., Opt. Express, in press (2006)

# Holographic lithography

N. Feth et al., Opt. Express, in press (2006)

## One incident beam does the job



exposure @ 532 nm

## Billion "magnetic atoms"



N. Feth et al., Opt. Express, in press (2006)

#### - Diamagnetism in optics

#### - Negative refractive index

# A double-negative metamaterial

Proposal: S. Zhang et al., Opt. Express 13, 4922 (2005)














**G. Dolling et al., Science 312, 892 (2006)** 





G. Dolling et al., Opt. Lett. 31, 1800 (2006)





FOM=0.5 @ Re(n)=-1 & 2 µm & Au: S. Zhang et al., Phys. Rev. Lett. 95, 137404 (2005)

FOM=0.1 @ Re(n)=-0.2 & 1.5 µm & Au: V.M. Shalaev et al., Opt. Lett. 30, 3356 (2005)

FOM=1-2 @ Re(n)≈-2 & 1.7 µm & Au: S. Zhang et al., J. Opt. Soc. Am. B 23, 434 (2006)

FOM=0.7 @ Re(n)=-1 & 1.5 µm & Au: G. Dolling et al., Science 312, 892 (2006)

FOM=3 @ Re(n)=-1 & 1.4 µm & Ag: G. Dolling et al., Opt. Lett. 31, 1800 (2006)

**Martin Wegener** 

FOM=0.5 @ Re(n)=-0.6 & 780 nm & Ag G. Dolling et al., submitted (2006)

## **Ag-based visible** metamaterial



s=17 nm, t=40 nm,  $a_x = a_y = 300$  nm,  $w_x = 102$  nm,  $w_y = 68$  nm, e=8 nm

## **Ag-based visible** metamaterial



G. Dolling et al., Opt. Lett., in press (2007)

# Michelson interferometer

@ 780 nm

CART II



G. Dolling et al., Opt. Lett., in press (2007)

## Towards **3D** metamaterials

So far, we have only demonstrated metamaterial <u>mono</u>layers.

However, it is well known that monolayers can have properties distinct from the bulk.

So what about several functional layers?

Theoretical proposal: S. Zhang et al., Opt. Express 14, 6778 (2006)

## N=3 functional layer sample



magnetic field and current @ 1410-nm wavelength where  $\mu < 0$ 



#### 500-nm scale bar

## **Towards 3D** metamaterials



G. Dolling et al., Opt. Lett., in press (2007)

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### Field-enhancement effects in metal nano-optics ...

- ... are analogous to the physics of lightning-rods
- ... can enhance local fields by factors >10<sup>3</sup>, leading to intensity enhancements of >10<sup>6</sup> and enhancements of >(10<sup>6</sup>)<sup>N</sup> for a N-th order nonlinear optical process
- ... for example, allow to detect Raman scattering from single molecules near to a metal nanostructure (started with rough metal films, hence the name)

J.J. Lasema ed., "Modern Techniques in Raman spectroscopy", John Wiley & Sons (1996)

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## LASER nm-analoge: SPASER



D.J. Bergmann and M.I. Stockman, Phys. Rev. Lett. 90, 027402 (2003)

## **Eigenmodes of silver "V"**

dark mode @ 1.63 eV

luminous mode @ 1.56 eV



D.J. Bergmann and M.I. Stockman, Phys. Rev. Lett. 90, 027402 (2003)

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#### **4.3. Transmission through sub-wavelength hole arrays**

# Consider a real metal film, perforated with a periodic array of sub-wavelength holes.



Naively, one would expect a transmission <u>smaller</u> than anticipated from geometrical optics (see section 1.2.4.).

In sharp contrast to this, experiments have shown much larger transmission for particular wavelengths.

T.W. Ebbesen et al., Nature 391, 667 (1998)

### The key are surface plasmon waves:



### The ansatz (air) for the electric field

$$E_y = E_y^0 \exp(i(K_x^{sp}x + K_y^{sp}y - \omega t)) + c.c.$$

### together with the boundary conditions leads to

$$K_x^{\rm sp} = \sqrt{\frac{\varepsilon(\omega)}{1 + \varepsilon(\omega)}} \frac{\omega}{c_0} > \frac{\omega}{c_0} \Longrightarrow (K_y^{\rm sp})^2 = \left(\frac{\omega}{c_0}\right)^2 - (K_x^{\rm sp})^2 < 0$$

#### i.e., to evanescent waves.



### Usually, one <u>cannot</u> couple from vacuum because of wavevector and frequency conservation.

For the arrays, reciprocal lattice vectors can be added

$$K_{x} = \frac{\omega}{c_{0}} \sin(\alpha) = \sqrt{\frac{\varepsilon(\omega)}{1 + \varepsilon(\omega)}} \frac{\omega}{c_{0}} \pm \left(N_{x} \frac{2\pi}{a}\right)$$

hence, the conservation laws <u>can</u> be fulfilled.



The coupling to the surface plasmons via reciprocal lattice vectors leads to enhanced transmission.

This qualitative explanation has been confirmed by detailed numerical calculations.

### **Measured spectra on perforated silver:**



**T.W. Ebbesen et al., Nature 391, 667 (1998)** 













### **Measured spectra on perforated silver:**



**T.W. Ebbesen et al., Nature 391, 667 (1998)** 



But! The holes are not essential for obtaining enhanced transmission through the metal film.

Any periodic modulation allows for coupling to the surface plasmon polaritons via reciprocal lattice vectors.

U. Schröter and D. Heitmann, Phys. Rev. B 60, 4992 (1999) I.R. Hooper and J.R. Sambles, Phys. Rev. B 67, 235404 (2003)

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# The End