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DRESDEN concept

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Fakultät Mathematik und Naturwissenschaften - Fachrichtung Physik

Institute of Applied Physics / Photophysics / Nanooptics

Near-field Infrared Microscopy:

Near-field nano-optics at NIR to THz wavelengths using the FELBE free electron laser

Lukas M. Eng
IAPP
TU Dresden

503. Heraeus-Seminar
Bad Honnef, D
April 11-13 2012

lukas.eng@iapp.de

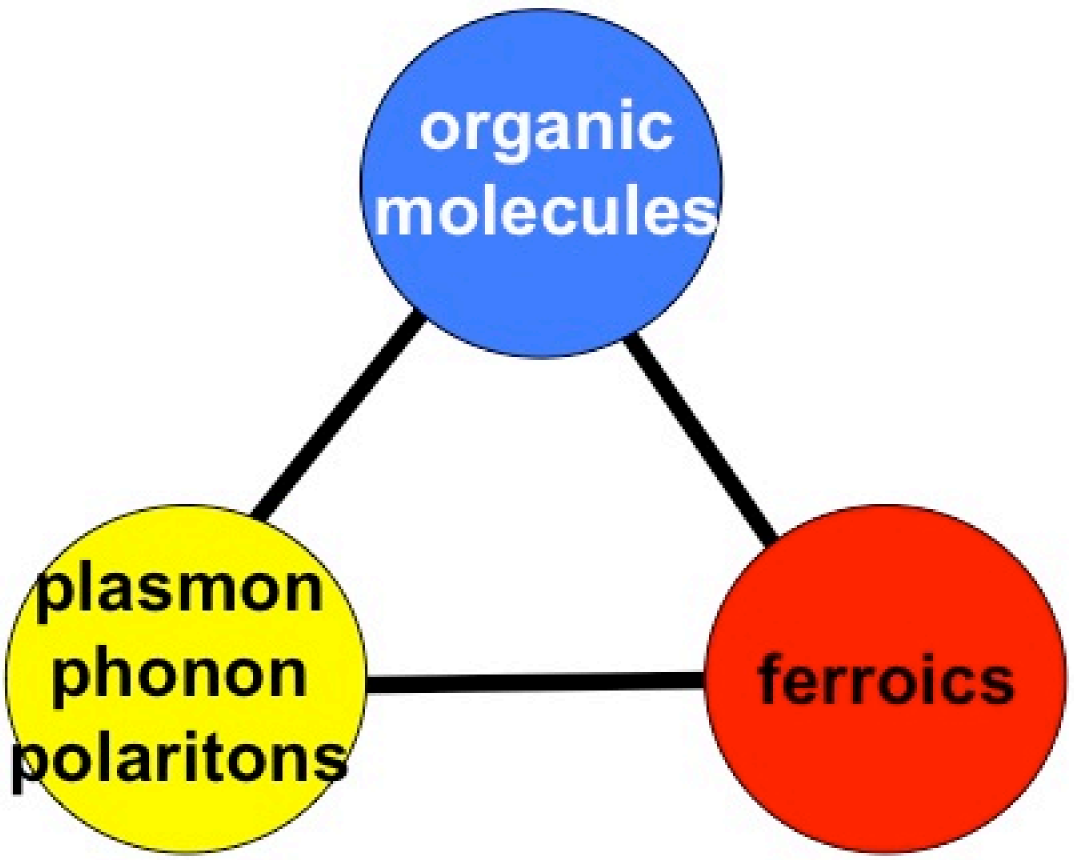
downtown Dresden

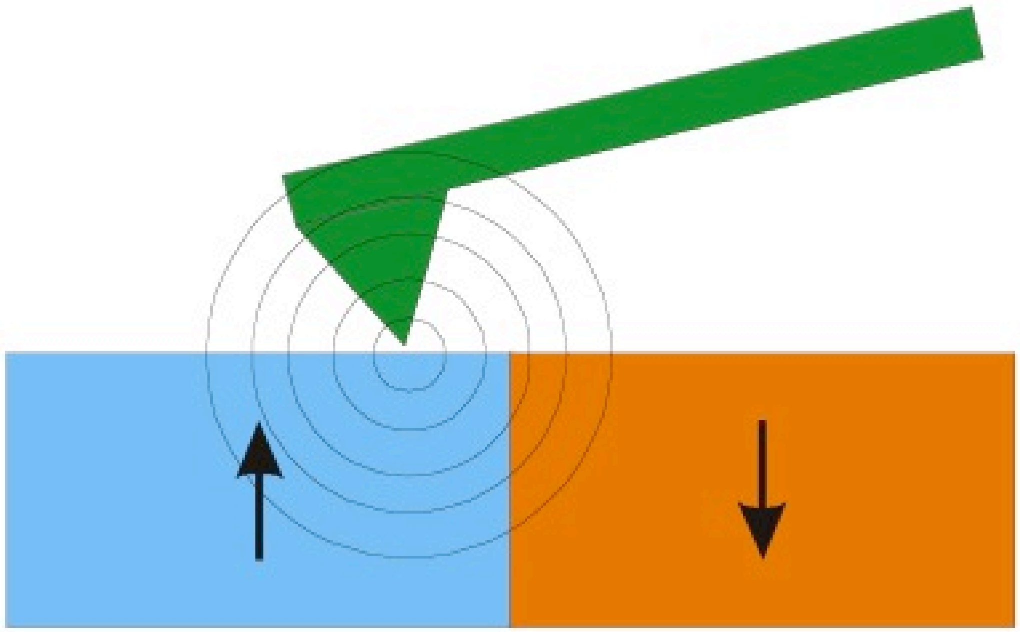
Goal

- IR-Spectroscopy
- IR-Microscopy
- “Local-scale” fingerprints
- Broad-band investigations (NIR to THz)
- Combine two key technologies:
  - free-electron laser
  - scanning force microscopy

Eng-group Research

Optical and electronic properties of novel and functional nanomaterials for applications in molecular electronics and optoelectronics





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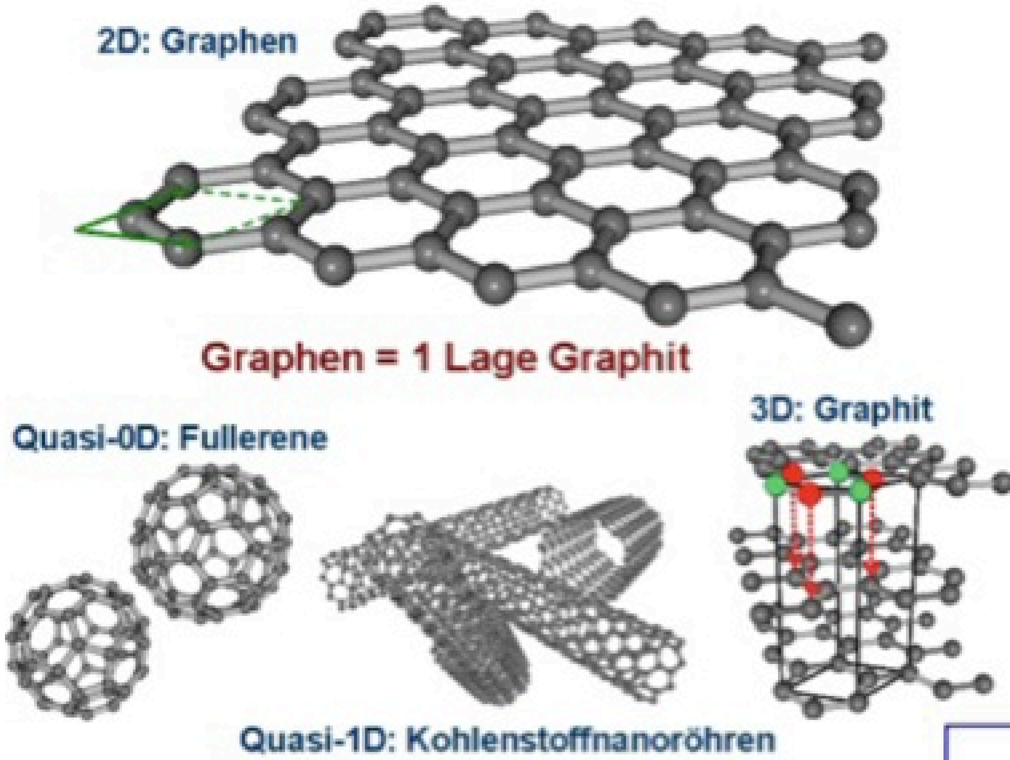
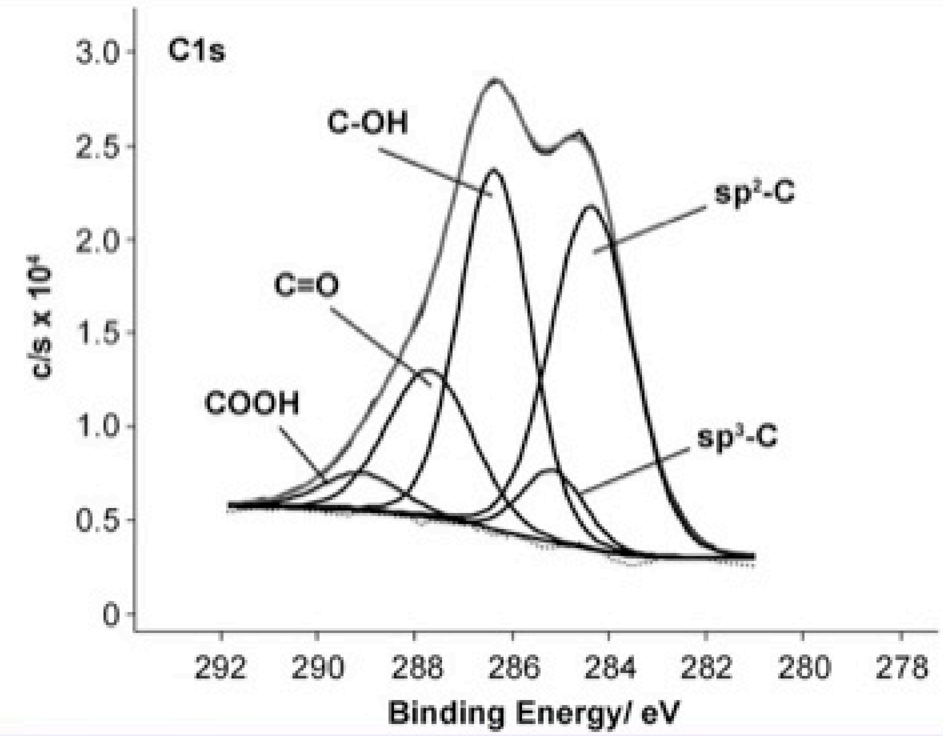
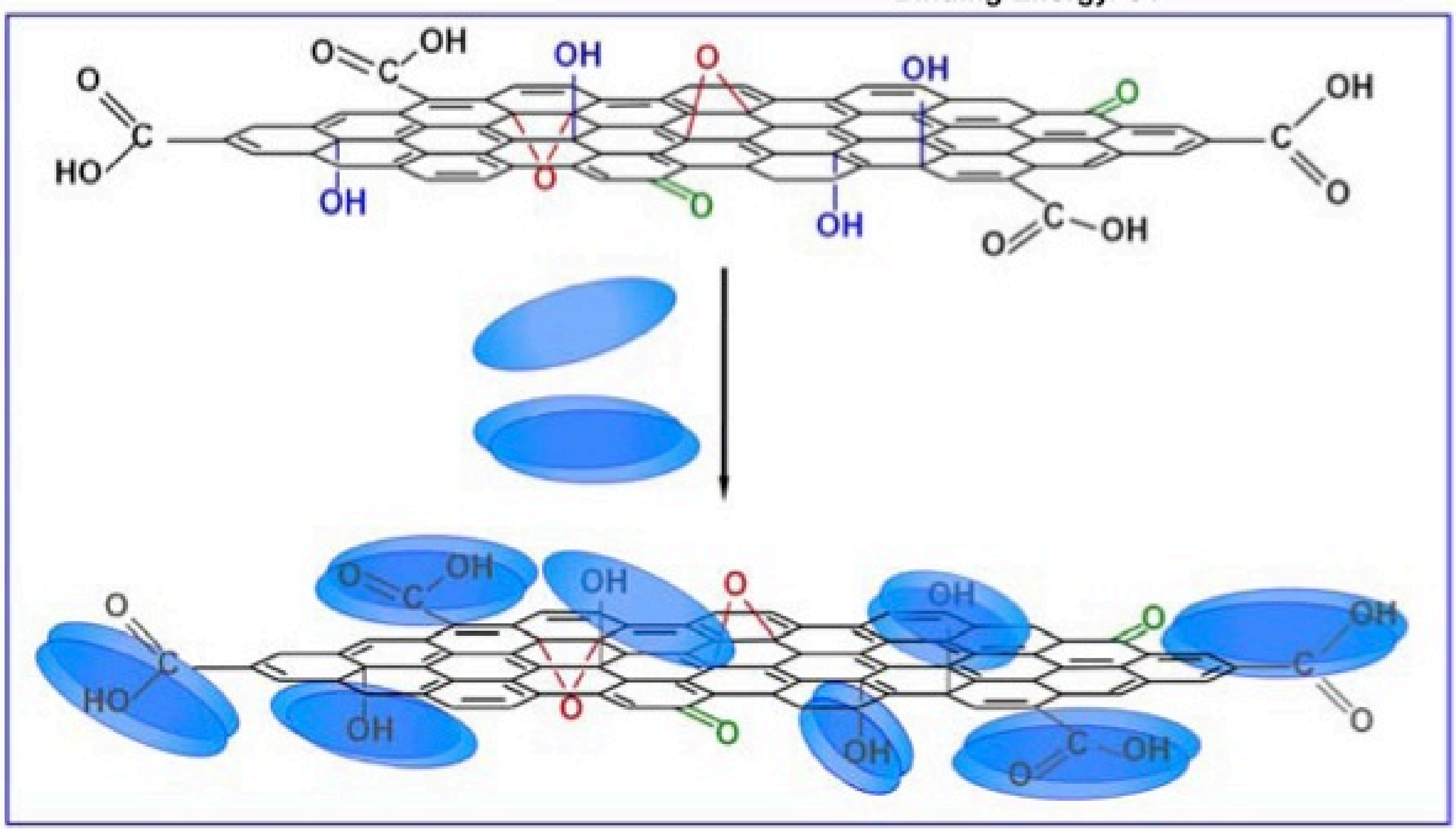
Functionalized Graphene

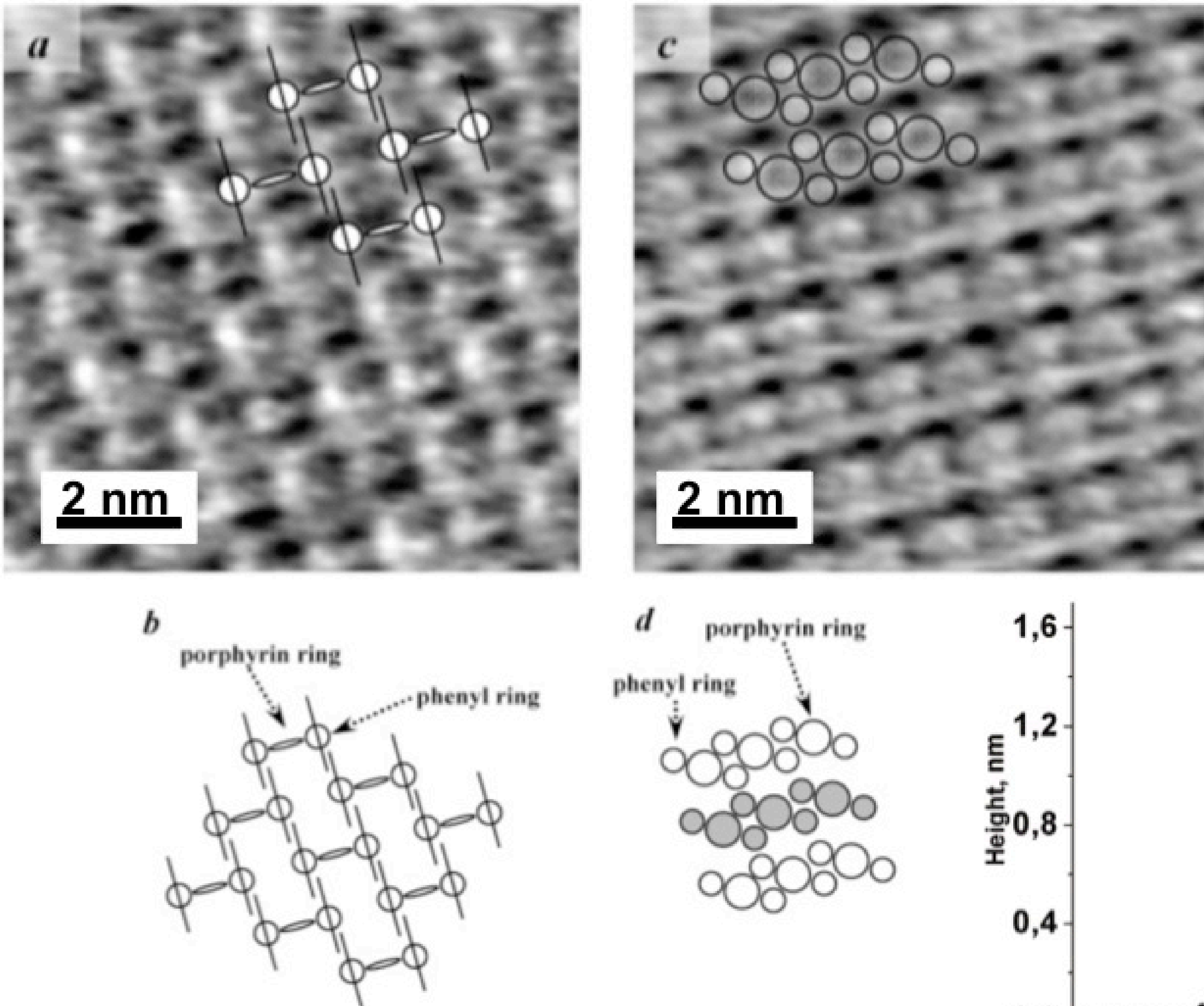


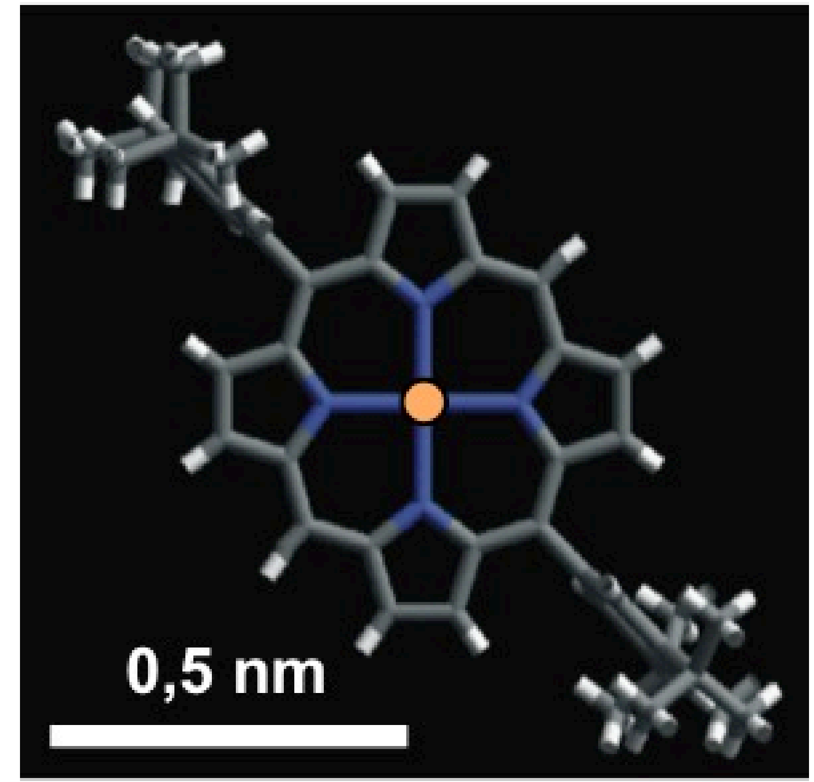


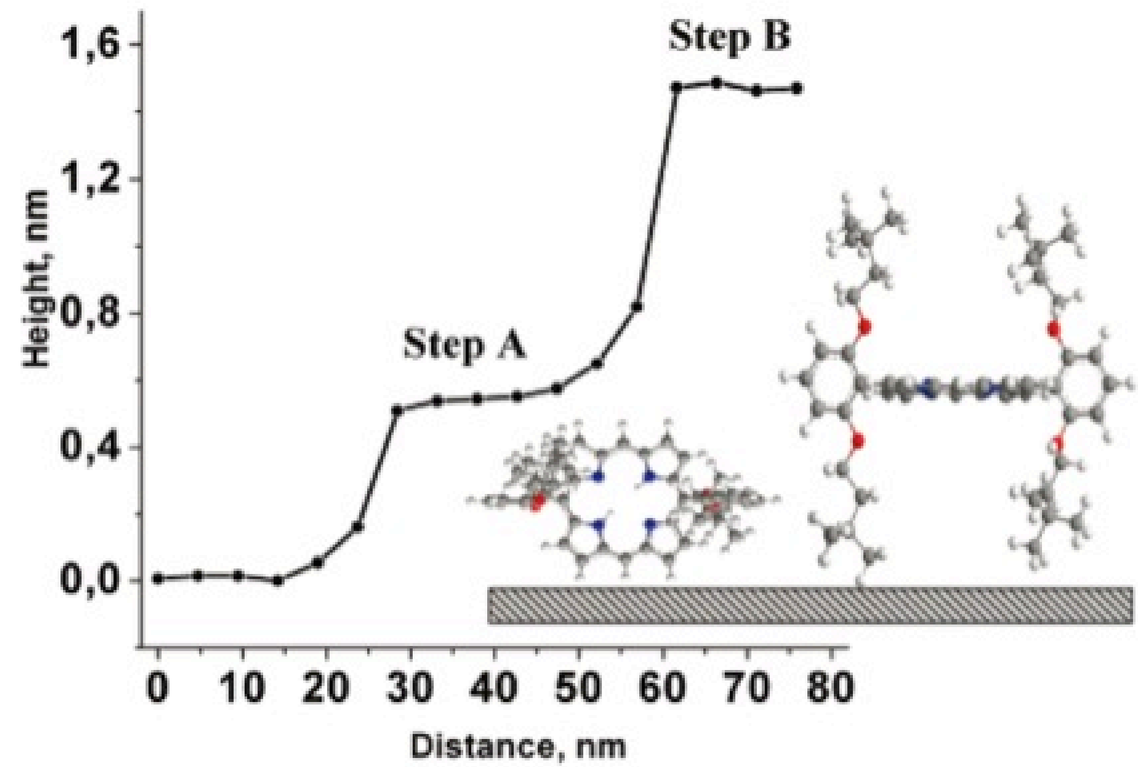


K. Haubner et al.,
CPC **11** (2010) 2131

Cu-dibutyl-porphyrine: Cu-DBPP



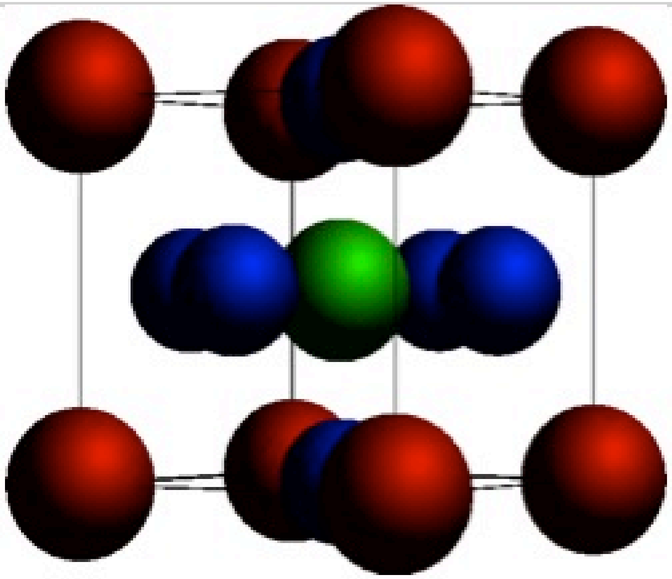




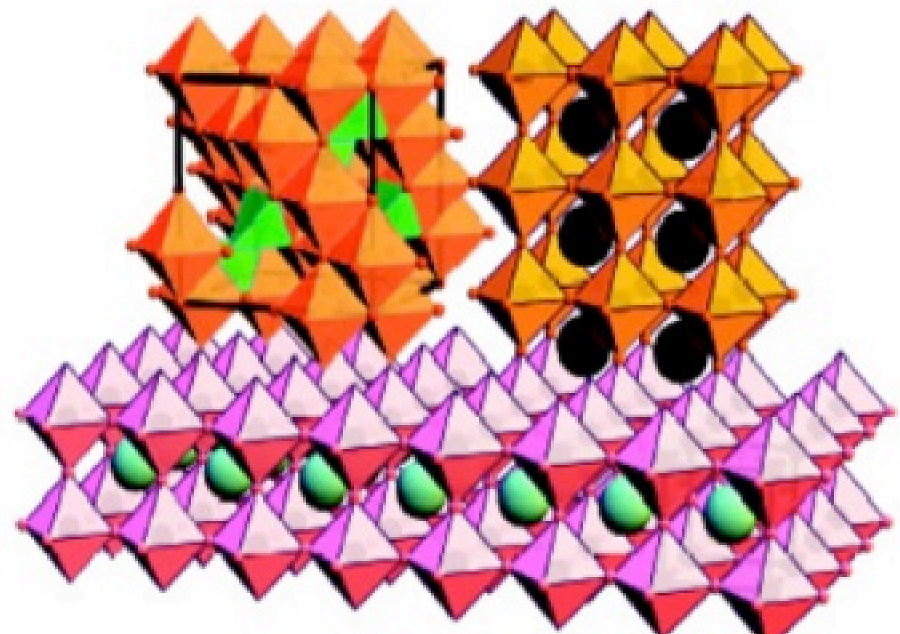
M. Nikiforov et al., *Nano Lett.* **8** (2008) 110

M. Nikiforov et al. *JAP* **106** (2009) 114307

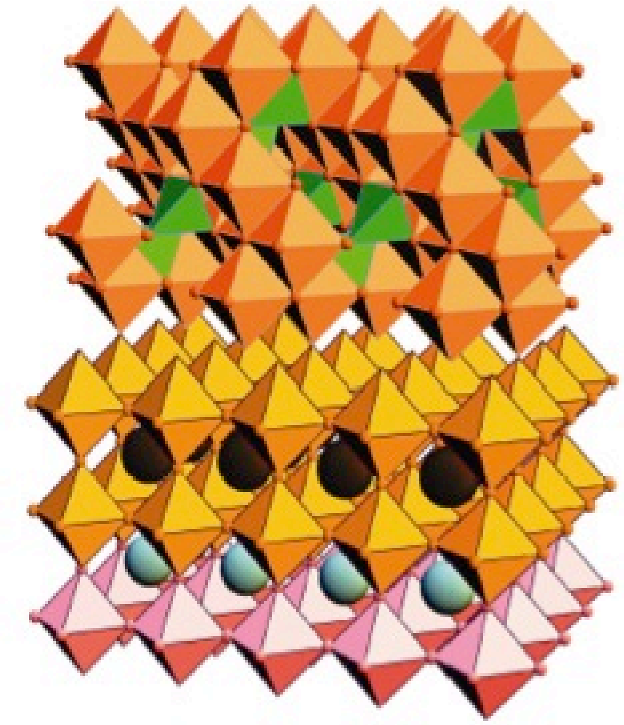
(Multi-) Ferroic Oxides



Single Phase Materials



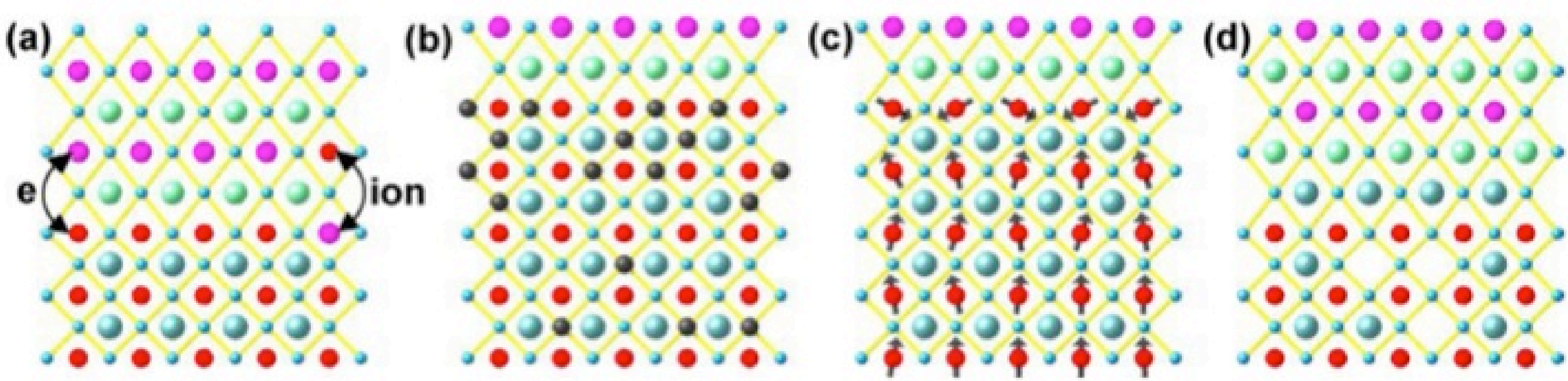
Horizontal Heterostructures



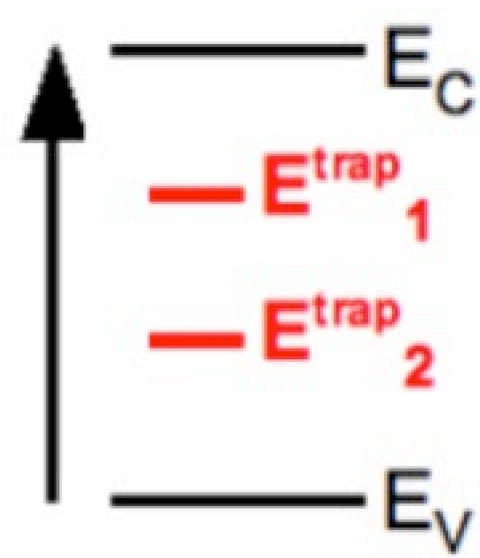
Vertical Heterostructures

"doping"

E. Beyreuther et al., *PRB* **73** (2006)



charge transfer | oxygen vacancies | magnetic/orbital disorder | cation segregation



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ε engineering

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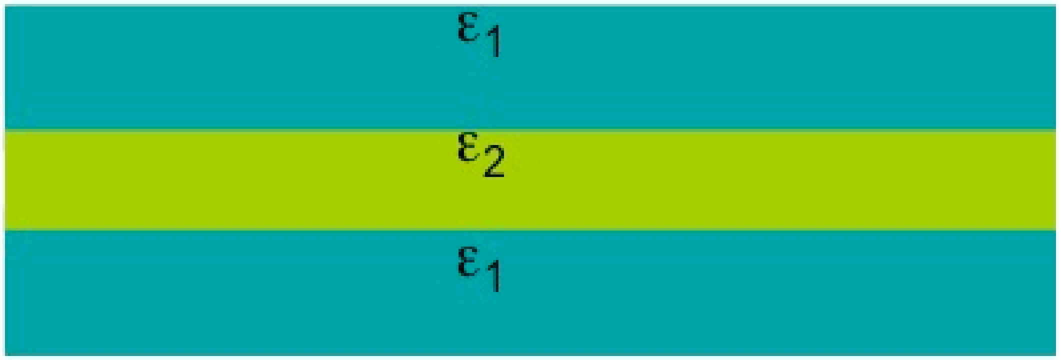
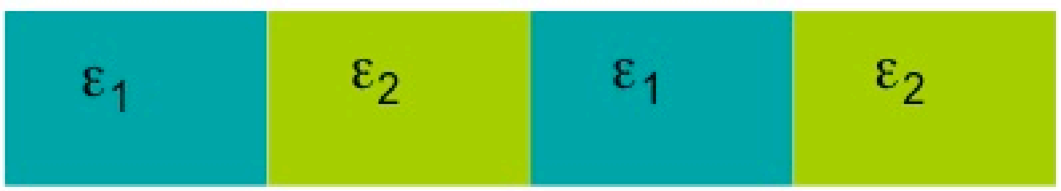
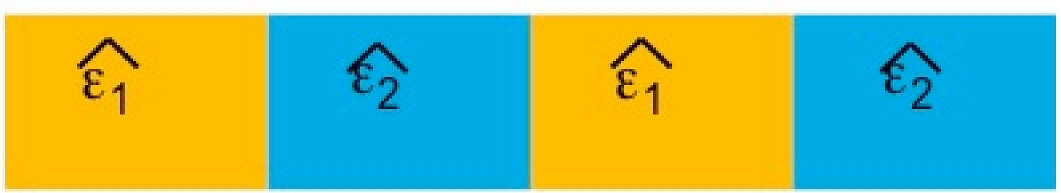


Vertically layered structure



Horizontally layered structure

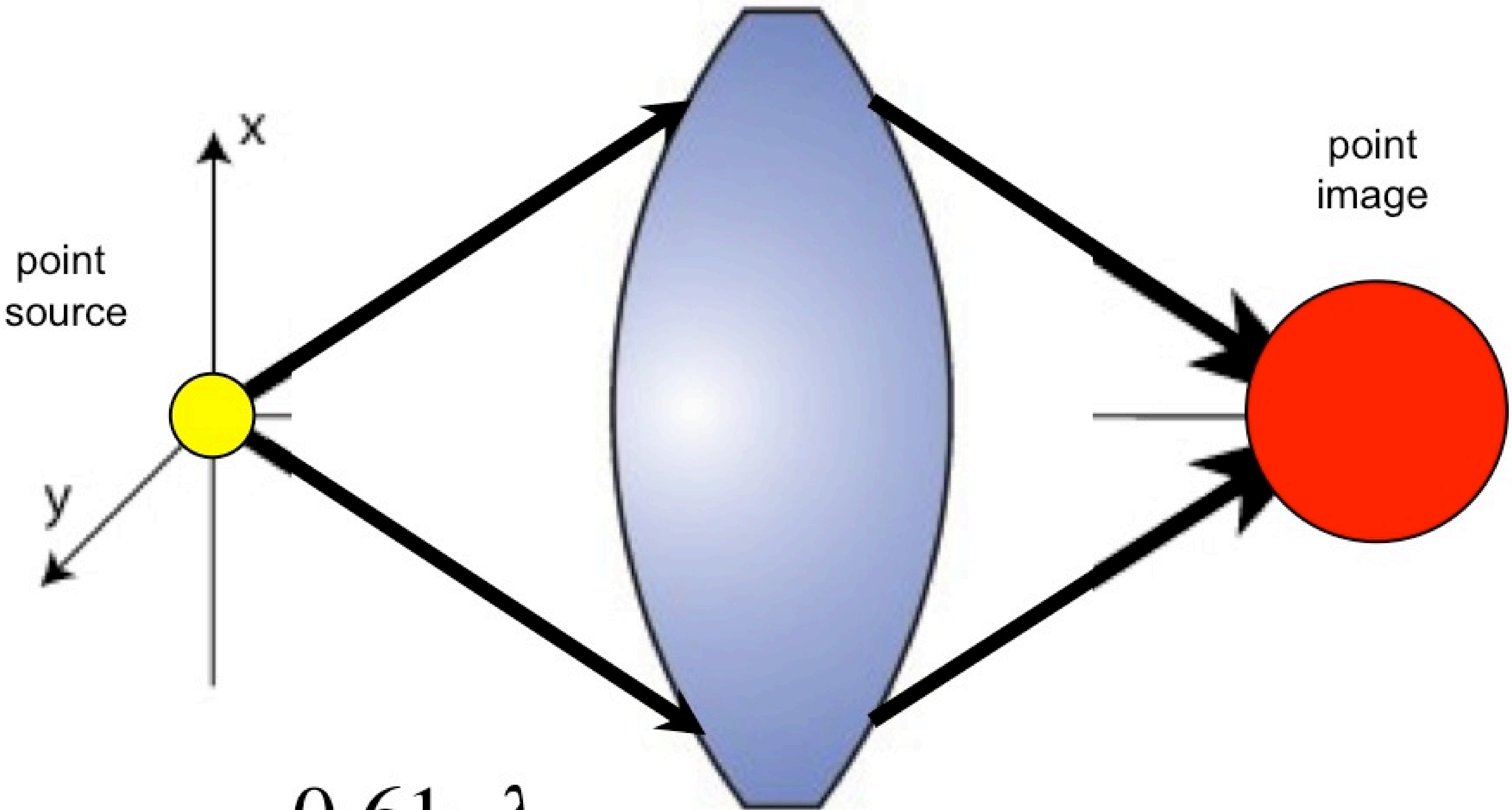


Horizontally layered, anisotropic structure

$\hat{\varepsilon}_i\,(\omega)$

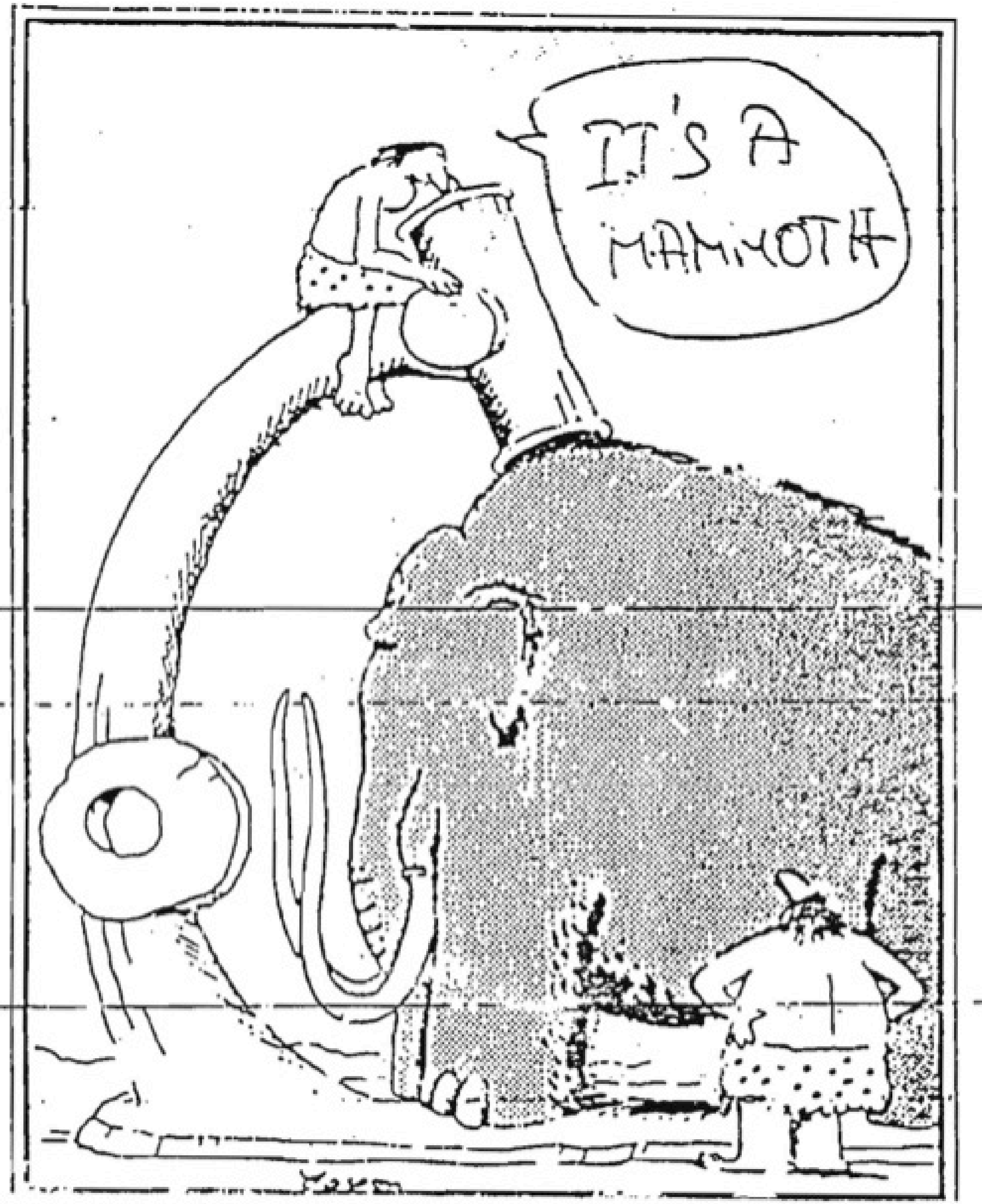
ω **most interesting @ NIR … THz**

Optical diffraction limit



$$\Delta x \approx \frac{0.61 \cdot \lambda}{N.A.}$$

λ: wavelength
N.A.: numerical aperture

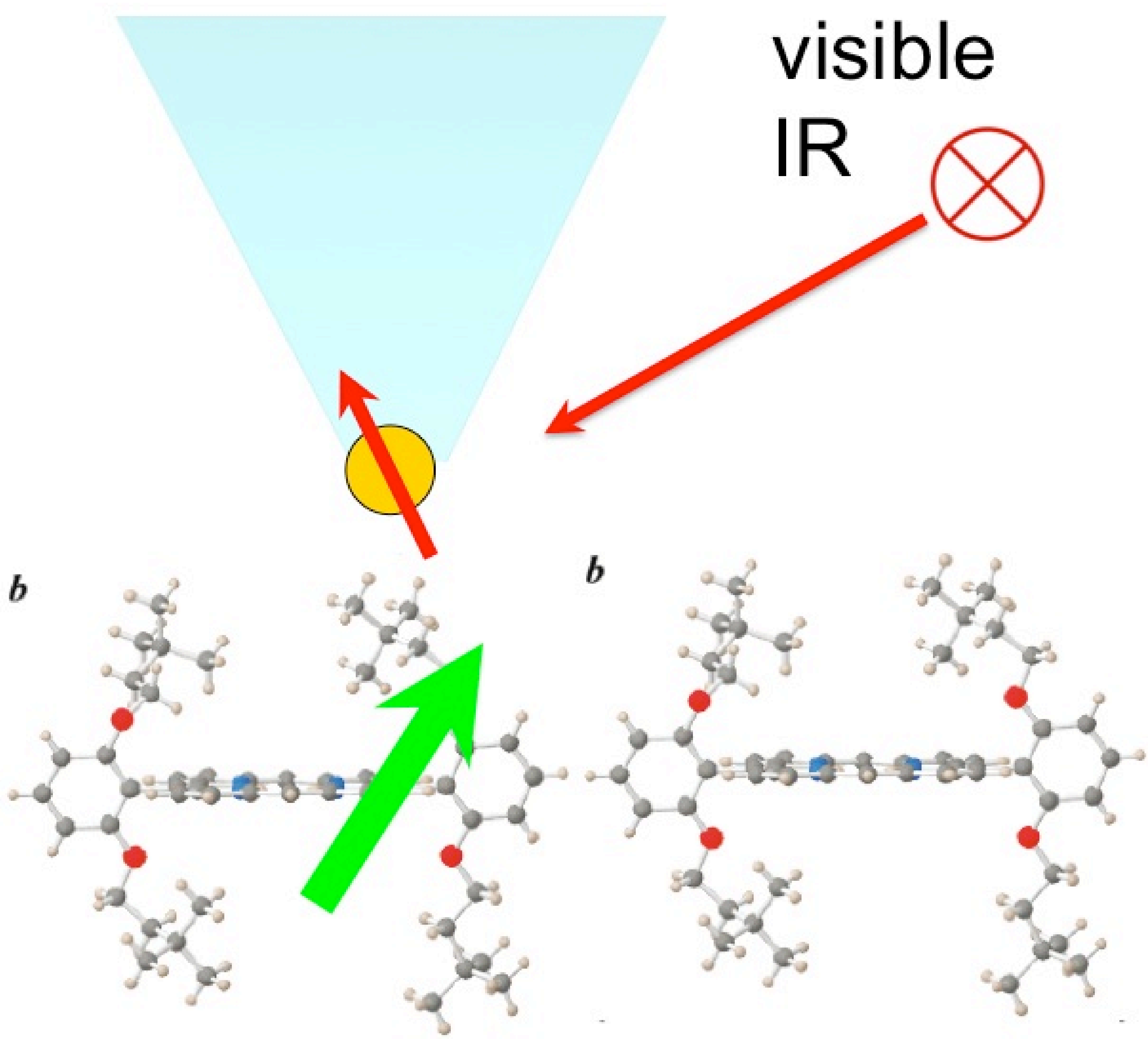


Early Microscope

Content

- Motivation
- IR scattering SNOM based on nc-AFM
- Examples:
  - Si/SiO_2
  - p-doped, buried Si
  - ferroelectric $BaTiO_3$
  - IR superlensing:
    ferroic metamaterials
- Summary

s-SNOM

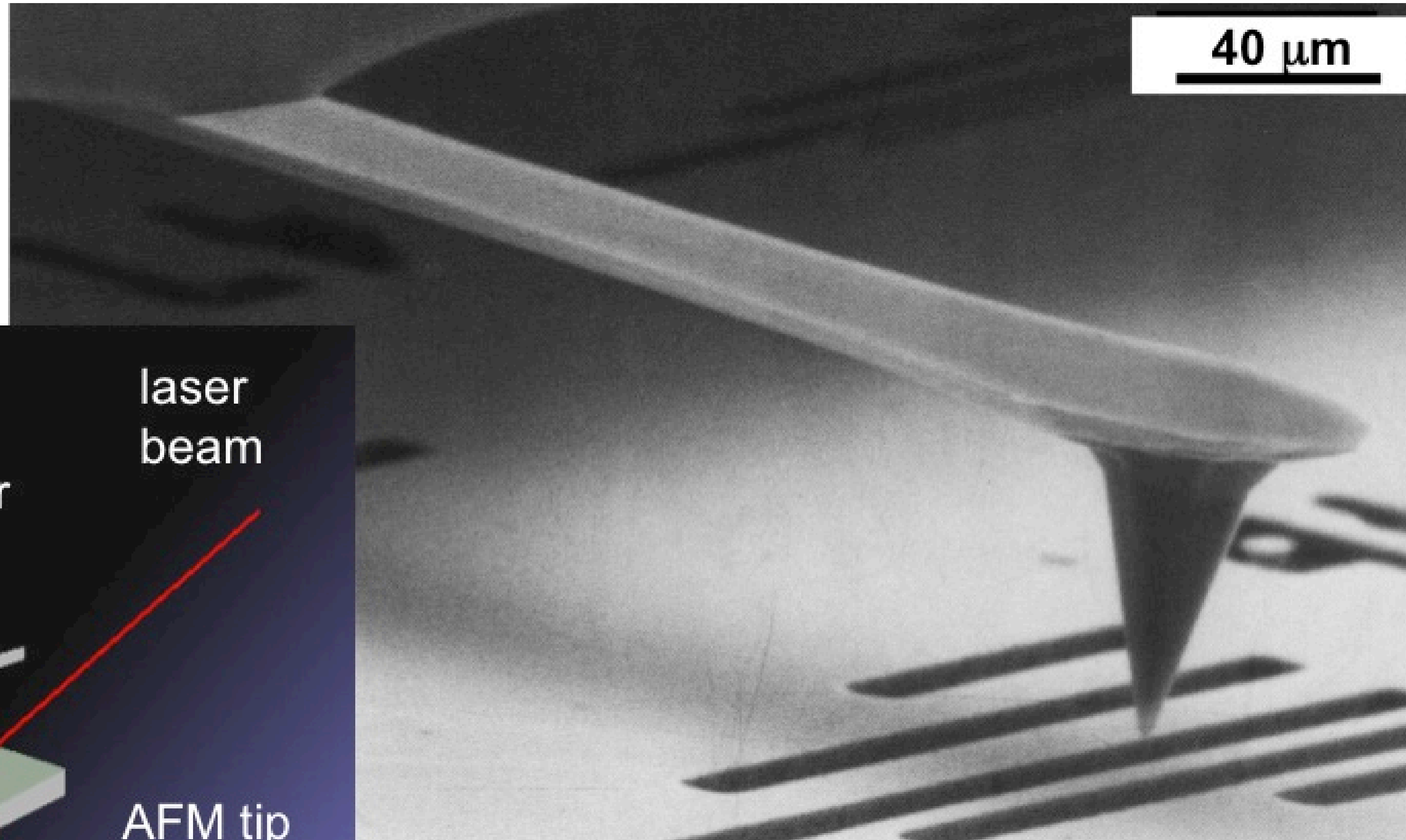


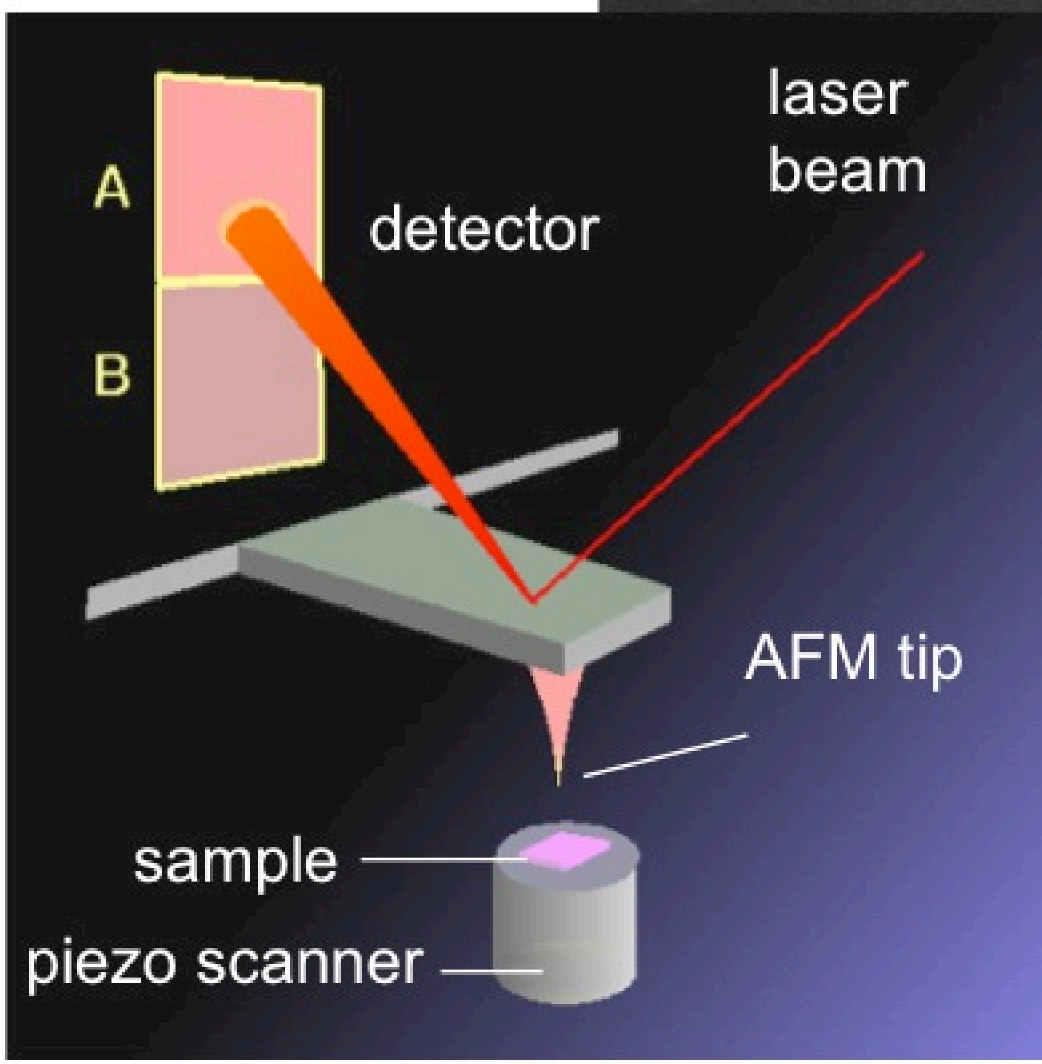
scattering near-field optical microscopy

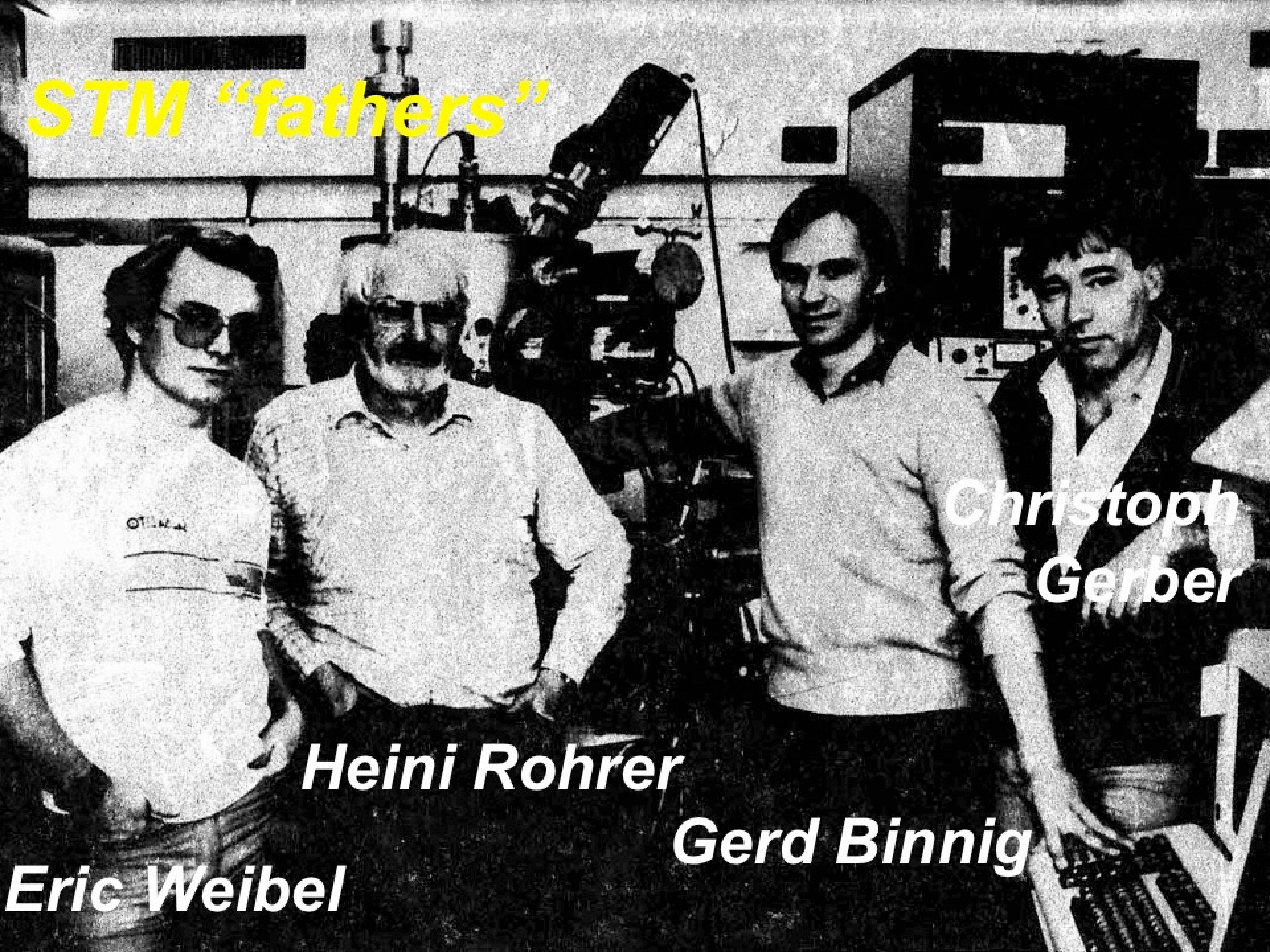
dipole-dipole interaction

M. Nikiforov et al., *JAP* **106** (2009) 114307

„Beam deflection“- AFM





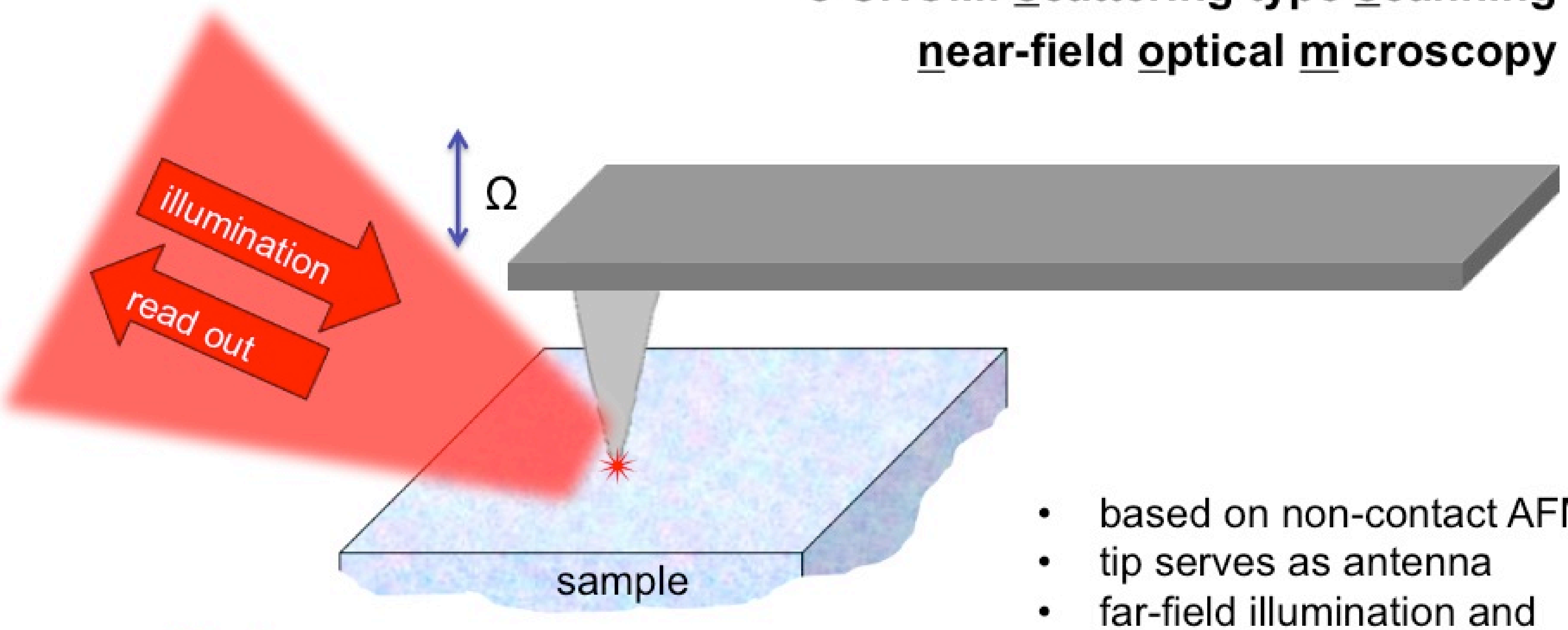

STM "fathers"
Christoph Gerber
Heini Rohrer
Gerd Binnig
Eric Weibel


AFM "fathers"
Heini Rohrer
Eric Weibel
Gerd Binnig
Christoph Gerber

"… feel the forces …"

s-SNOM principle

s-SNOM: <u>s</u>cattering type <u>s</u>canning <u>n</u>ear-field <u>o</u>ptical <u>m</u>icroscopy



- based on non-contact AFM
- tip serves as antenna
- far-field illumination and read out
- confined near field probes optical surface properties
- simultaneous AFM operation: topo, electrical, magnetic info

- **scattering**
- **Raman**
- **absorption**
- **fluorescence**
- **...**

Free-electron laser FELBE

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Free-electron laser FELBE

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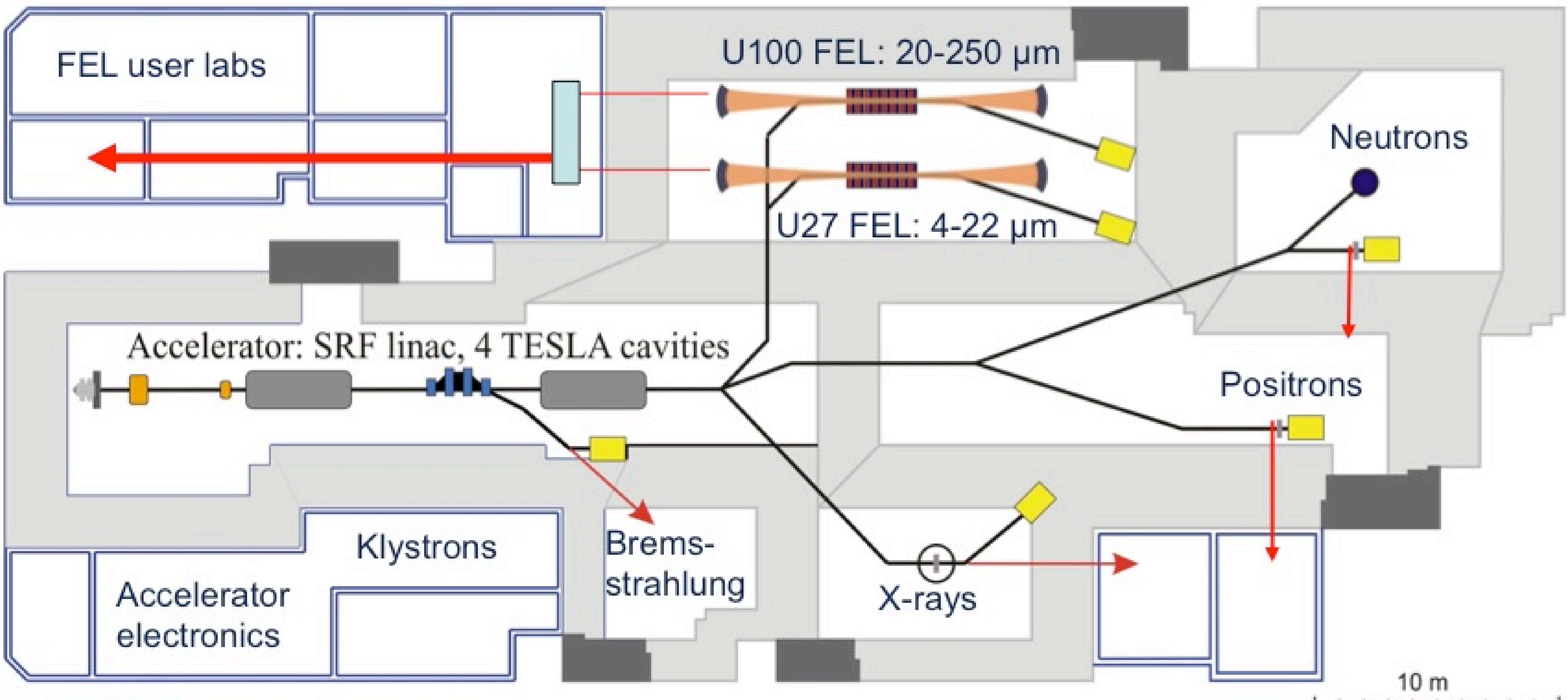
FELBE: Free Electron Linac with high Brilliance and low Emittance



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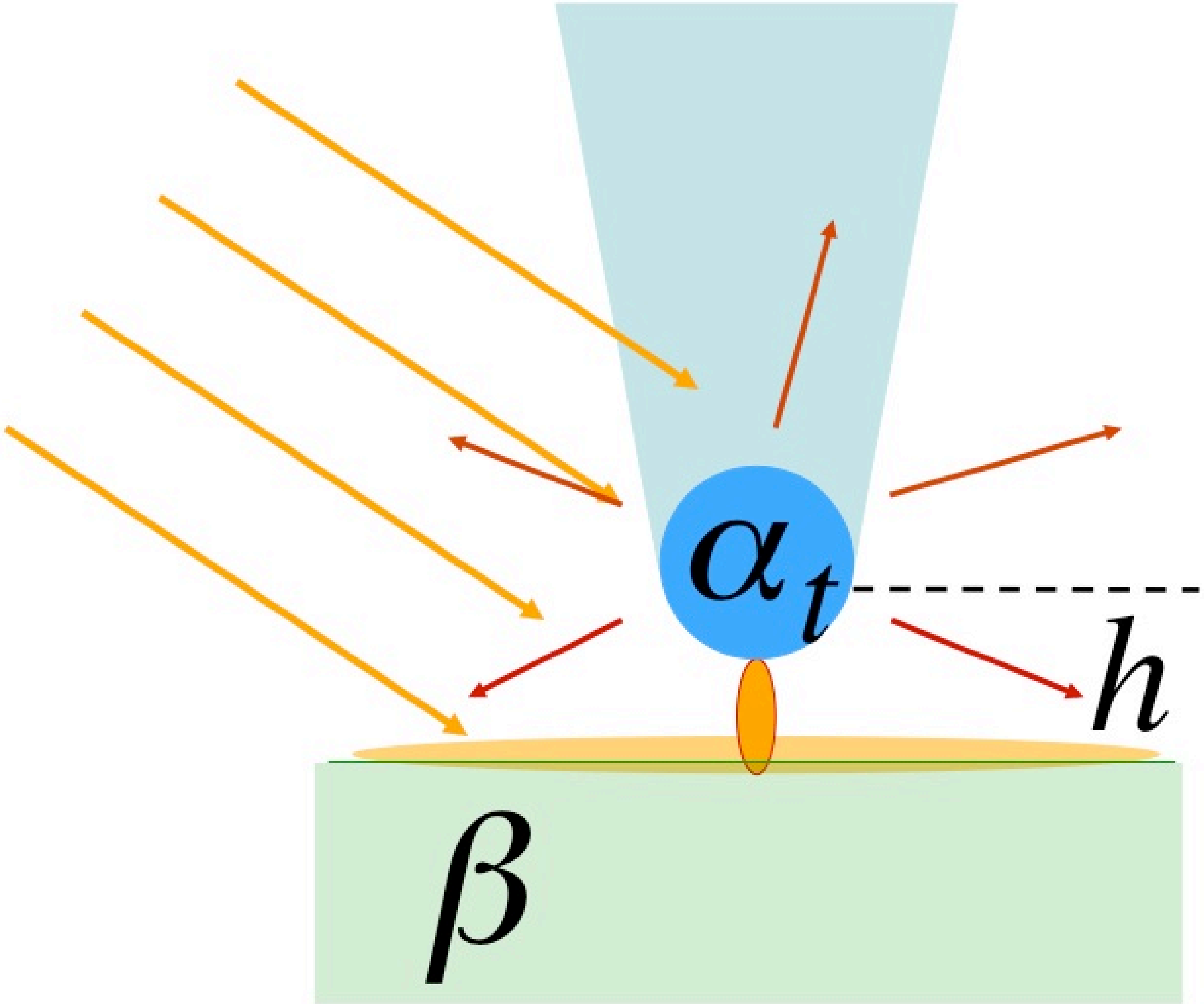
Resonant field enhancement



$\beta = \frac{\varepsilon_s - 1}{\varepsilon_s + 1}$

α_t tip polarizability
h tip-sample distance
β sample response
α_{eff} total polarizability

$$\alpha_{ij} = \frac{\delta \vec{P}_i}{\delta \vec{E}_j}$$

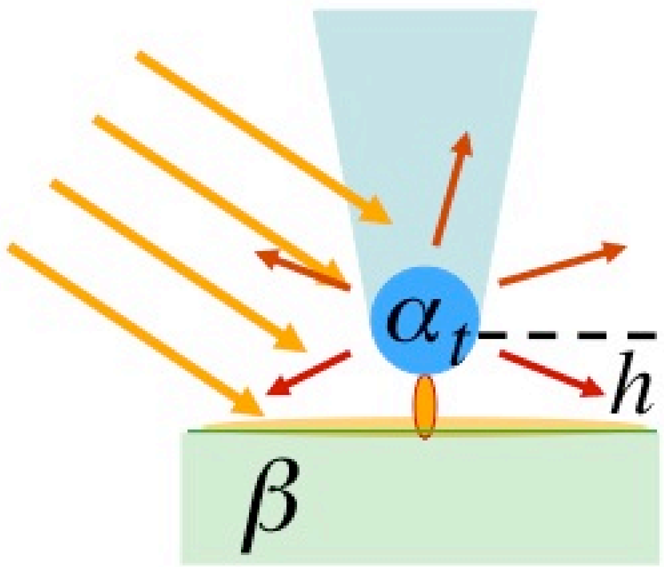
$$\alpha_{eff} = \frac{\alpha_t \cdot (1+\beta)}{1 - \frac{\alpha_t \cdot \beta}{16\pi h^3}}$$

p-polarization

$$\alpha_{eff} = \frac{\alpha_t \cdot (1-\beta)}{1 - \frac{\alpha_t \cdot \beta}{32\pi h^3}}$$

s-polarization

Resonant field enhancement



$$\alpha_{eff} = \frac{\alpha_t (1+\beta)}{1 - \frac{\alpha_t \cdot \beta}{16\pi h^3}}$$

p-polarization

α_t	tip polarizability
h	tip-sample distance
β	sample response
α_{eff}	total polarizability

$$\beta = \frac{\varepsilon_s - 1}{\varepsilon_s + 1}$$

Tip resonance
Plasmon polariton
(visible wavelengths)

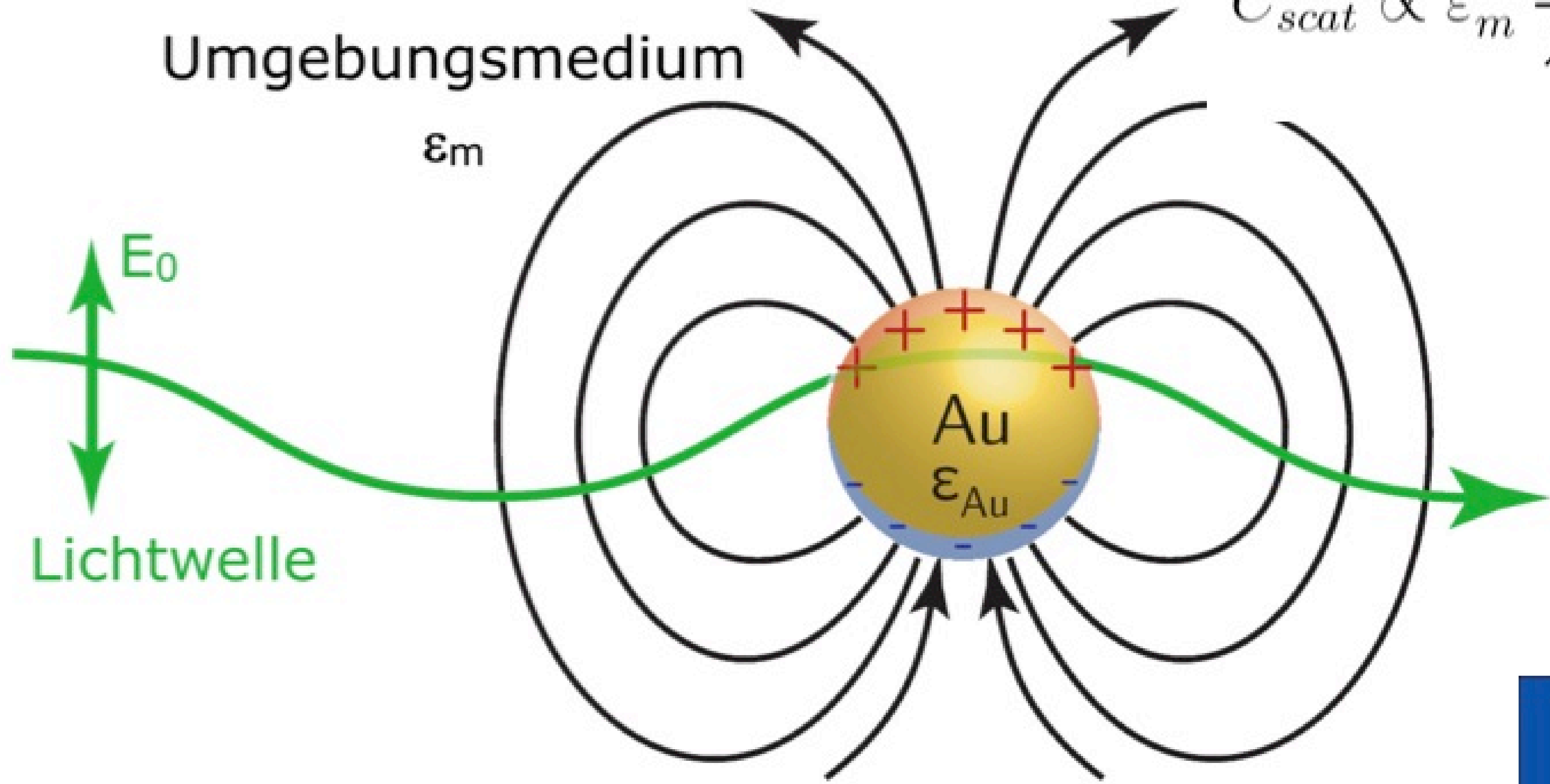
Sample resonance
Phonon polariton
(IR wavelengths)

Scattering: $$C_{sca} = \frac{k^4}{6\pi}\left|\alpha_{eff}\right|^2 = \frac{8\pi^3}{3\lambda^4}\left|\alpha_{eff}\right|^2$$

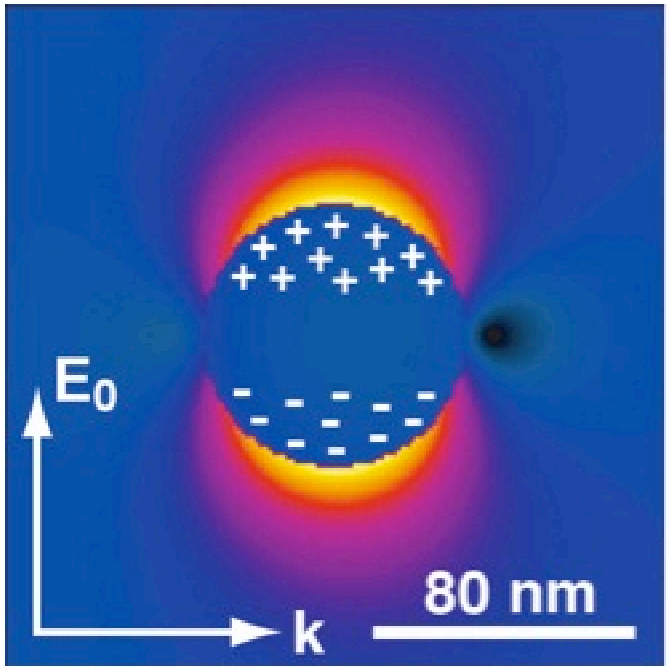
Absorption: $$C_{abs} = k \cdot \mathrm{Im}\left(\alpha_{eff}\right) = \frac{2\pi}{\lambda} \cdot \mathrm{Im}\left(\alpha_{eff}\right)$$

Plasmons in metal NPs

$$C_{scat} \propto \varepsilon_m^2 \frac{R^6}{\lambda^4} \cdot \left| \frac{\varepsilon_{Au}(\omega) - \varepsilon_m}{\varepsilon_{Au}(\omega) + 2\varepsilon_m} \right|^2$$



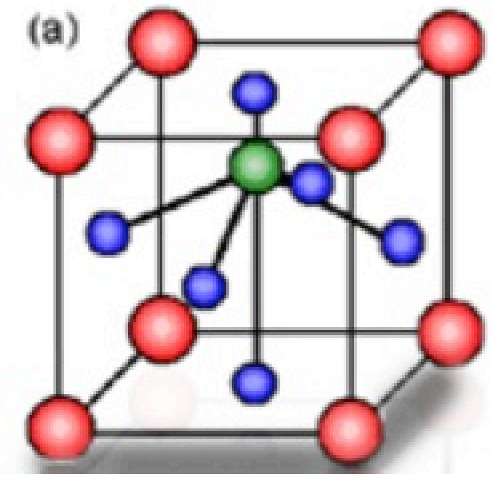
Mie scattering



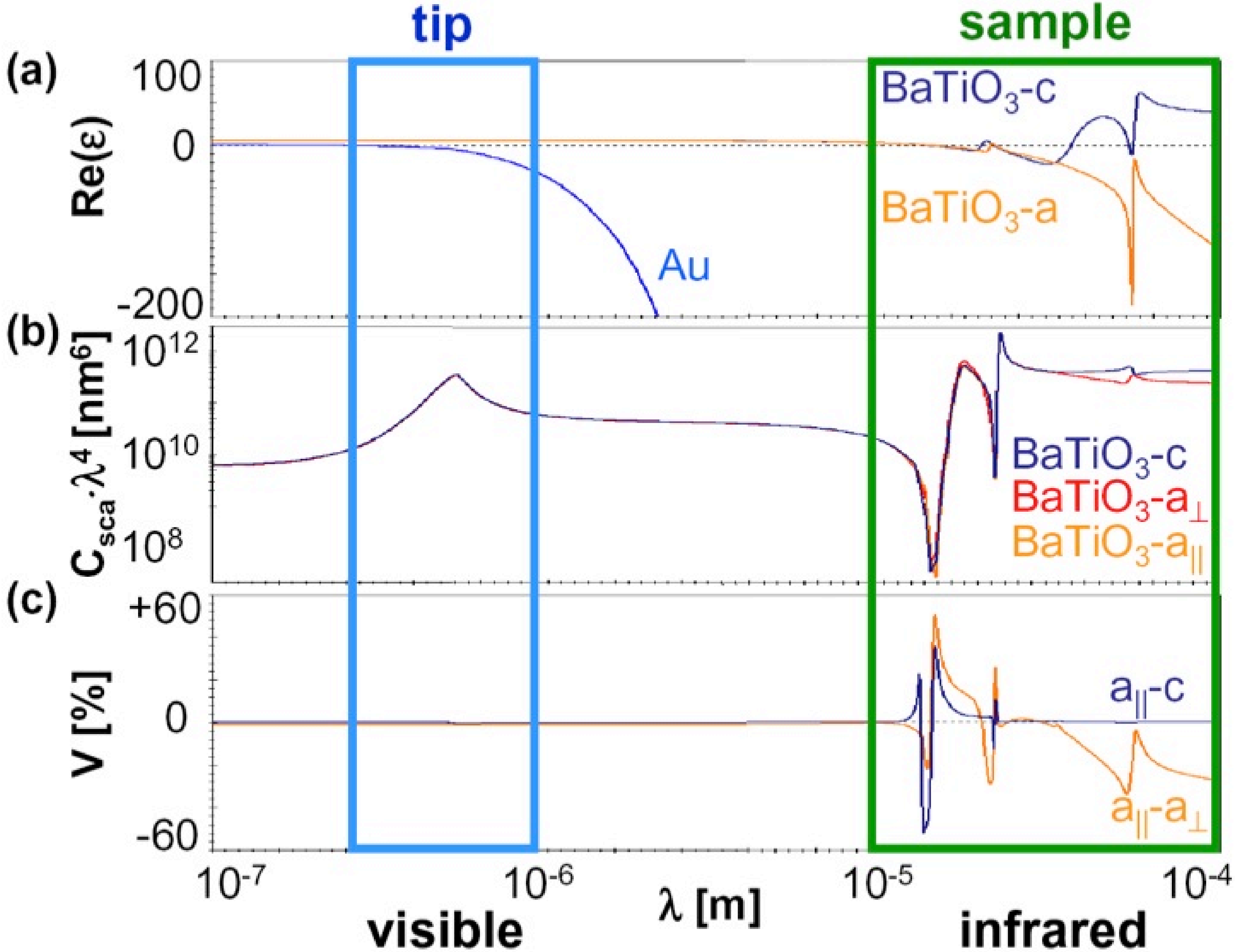
Limiting effects:
- small particle: d < 3 nm, damping
- large particle: d > λ/2, dephasing

Polaritons in Au and $BaTiO_3$

$BaTiO_3$
tetragonal

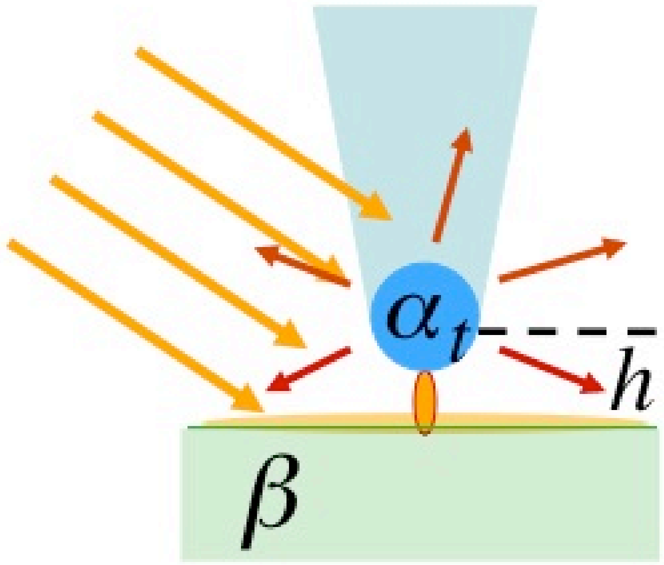


c-domain



S. Kehr, *PhD thesis*, IAPP (2007)

Resonant field enhancement



$$\alpha_{eff} = \frac{\alpha_t \cdot (1+\beta)}{1 - \frac{\alpha_t \cdot \beta}{16\pi h^3}}$$

p-polarization

α_t	tip polarizability
h	tip-sample distance
β	sample response
α_{eff}	total polarizability

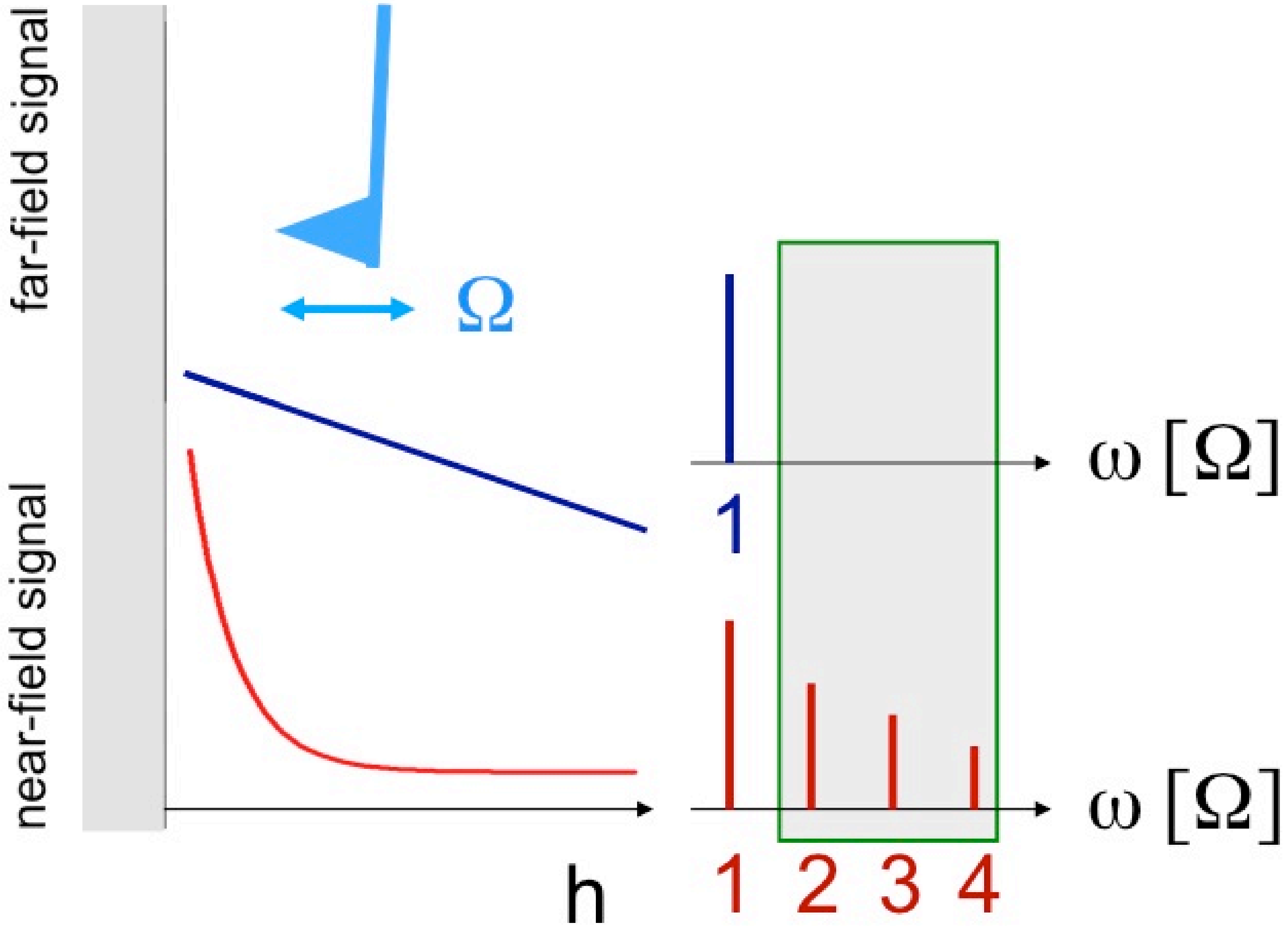
$$\beta = \frac{\varepsilon_s - 1}{\varepsilon_s + 1}$$

Scattering: $$C_{sca} = \frac{k^4}{6\pi}\left|\alpha_{eff}\right|^2 = \frac{8\pi^3}{3\lambda^4}\left|\alpha_{eff}\right|^2$$

Absorption: $$C_{abs} = k \cdot \mathrm{Im}\left(\alpha_{eff}\right) = \frac{2\pi}{\lambda} \cdot \mathrm{Im}\left(\alpha_{eff}\right)$$

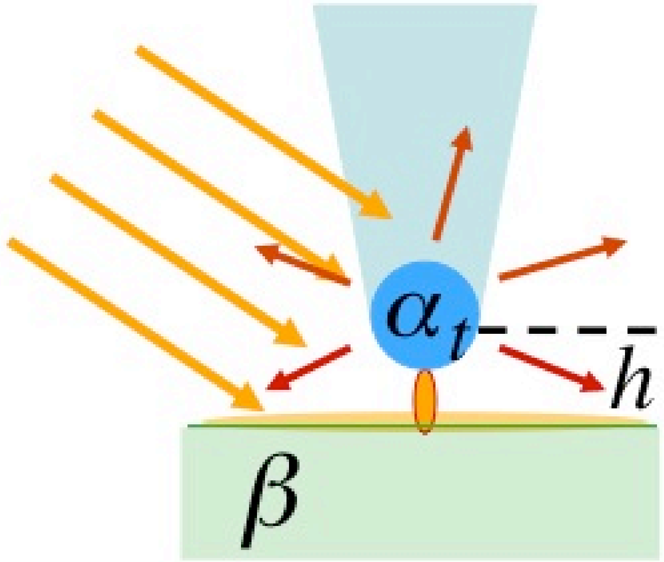
- **λ spectroscopy**
- **distance *h* spectroscopy**
- **imaging -> microscopy**

Background elimination



Fourier series de-convolution of non-linear near-field signal

4 Examples



$$\alpha_{eff} = \frac{\alpha_t (1+\beta)}{1 - \frac{\alpha_t \cdot \beta}{16\pi h^3}}$$

α_t tip polarizability
h tip-sample distance
β sample response

$$\beta = \frac{\varepsilon_s - 1}{\varepsilon_s + 1}$$

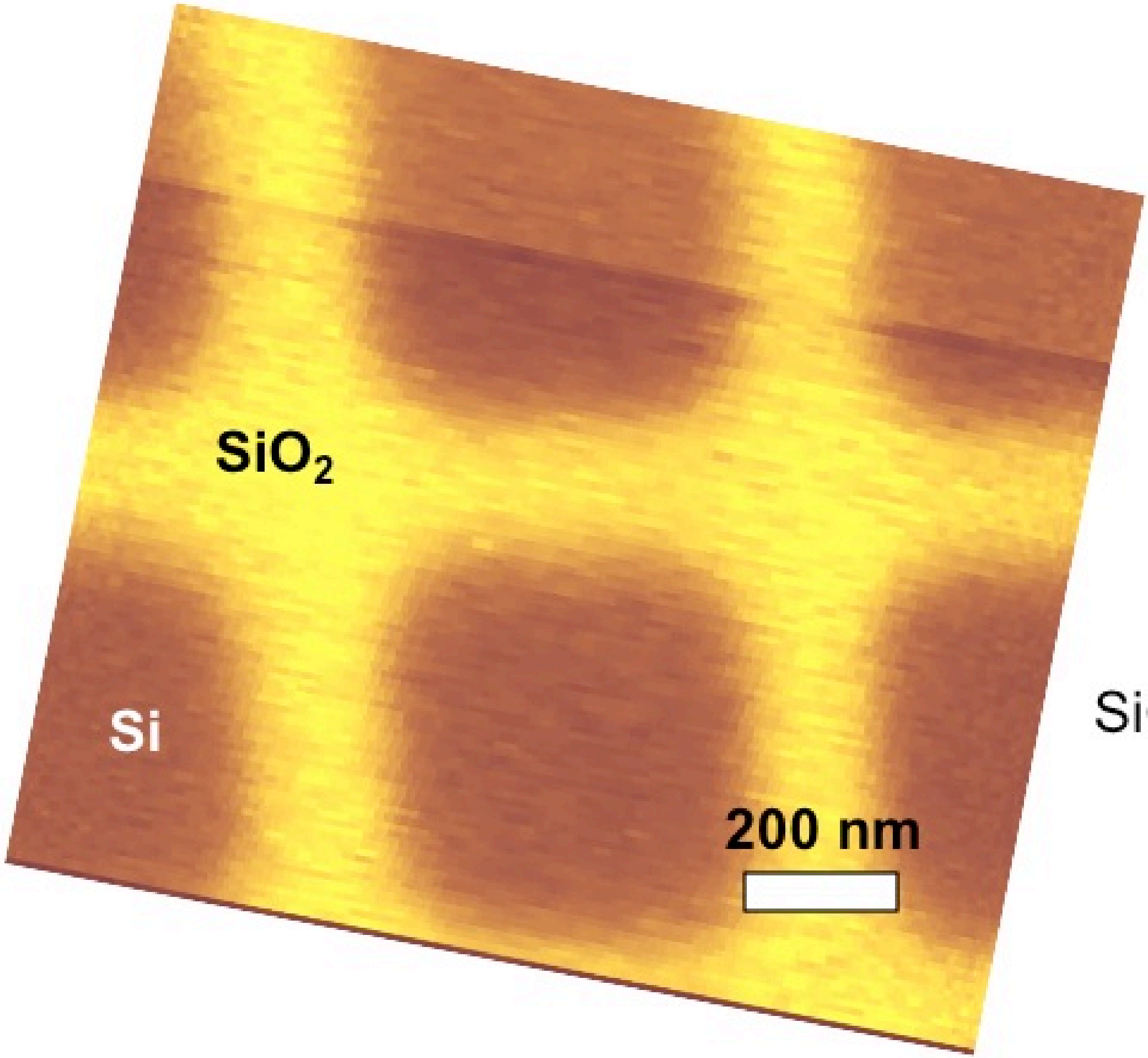
Tip resonance
Plasmon polariton
(visible wavelengths)

⇒ Signal increased

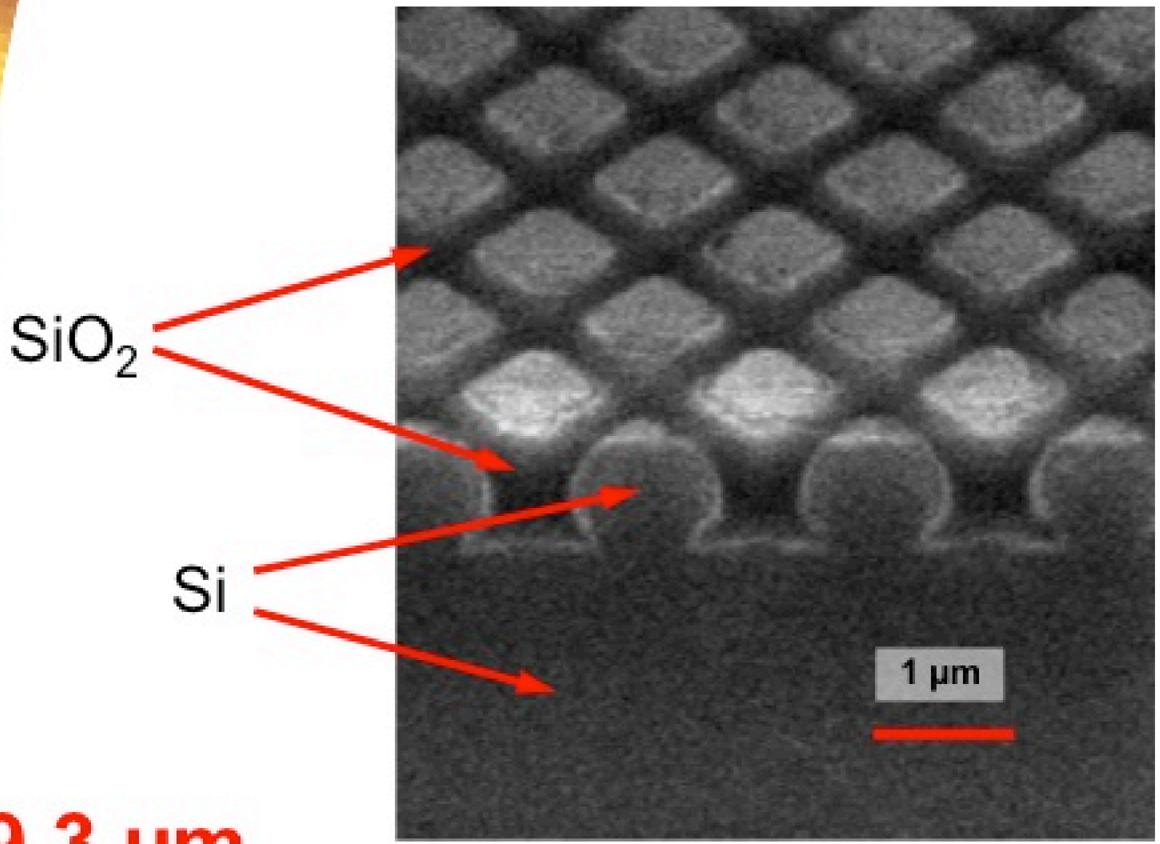
Sample resonance
Phonon polariton
(IR wavelengths)

⇒ Signal increased

#1: Si/SiO_2



- DRAM application
- Non-destructive technique
- Local dopand concentration

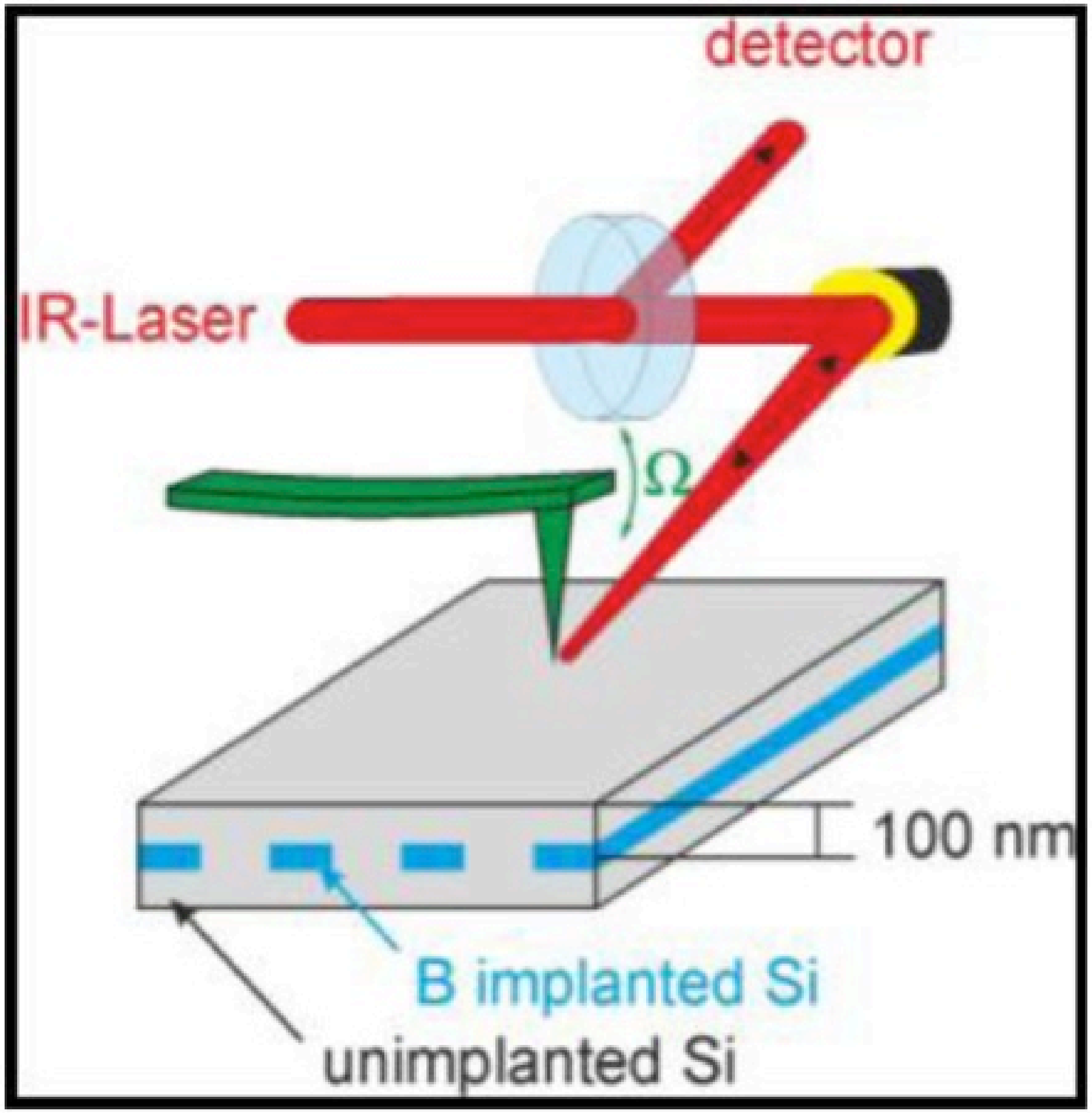


SiO_2 filled trenches, thickness 400 nm, width < 200 nm

➢ **Resolution of < λ/1000 @ 9.3 µm**

M.T. Wenzel, TUD (2011)

#2: p:Si

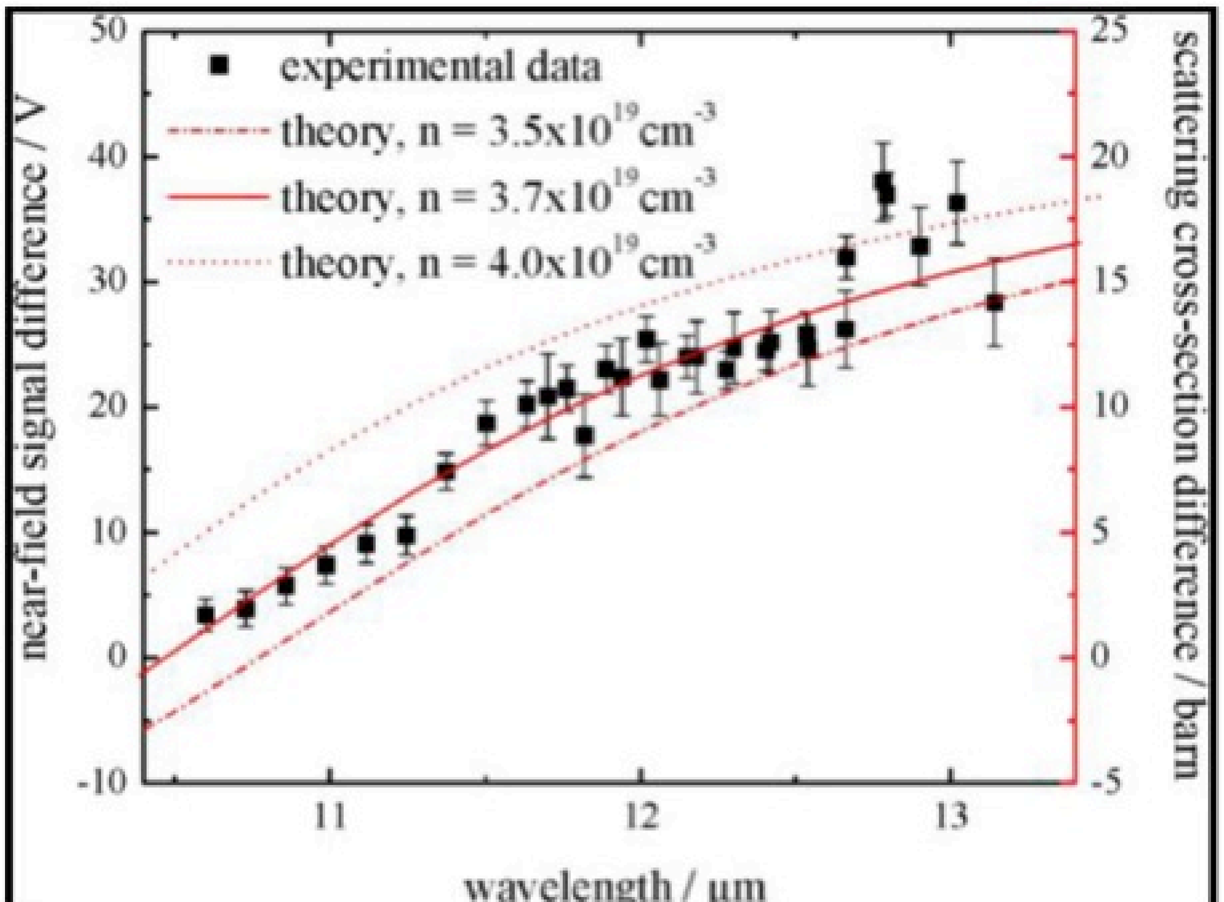


$$\varepsilon_s(\omega, N) = \varepsilon_\infty - \frac{\omega_p^2(N)}{\omega^2 + i\omega\gamma(N)}$$

$$\omega_p^2(N) = \frac{q^2 N}{m^* \varepsilon_0}$$

Theory: $(4{,}0 \pm 1{,}0) \times 10^{19}\ \mathrm{cm}^{-3}$

Experiment: $(3{,}7 \pm 0{,}3) \times 10^{19}\ \mathrm{cm}^{-3}$

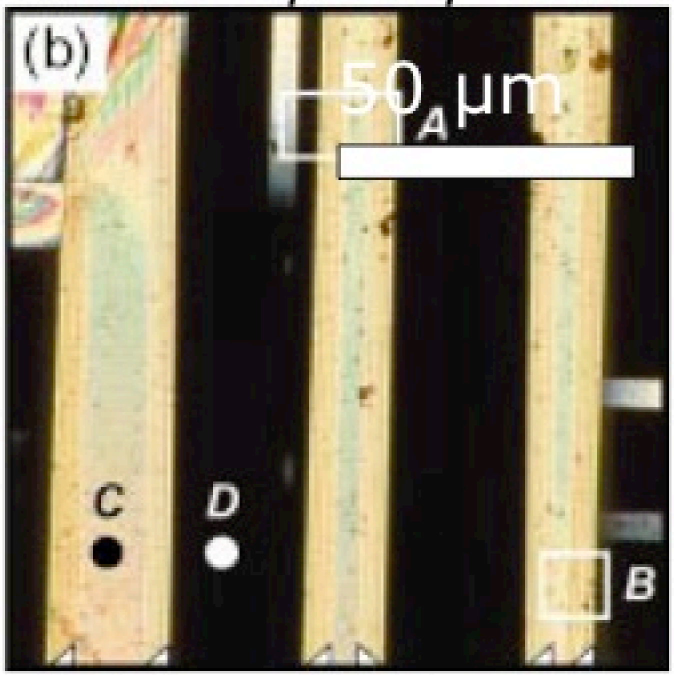


R. Jacob et al., *Optics Expr.* **18** (2010) 26206

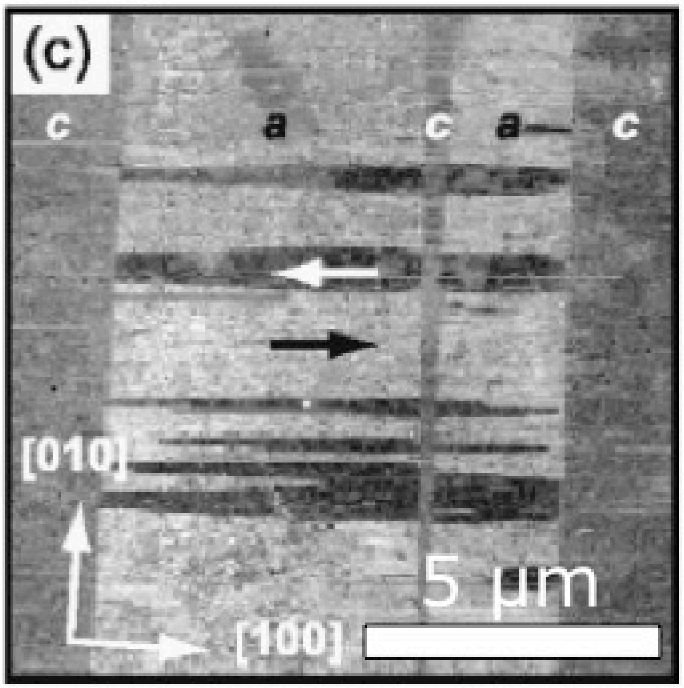
#3: $BaTiO_3$

- atomically flat surface
- anisotropic $\hat{\varepsilon}_s$
- domains

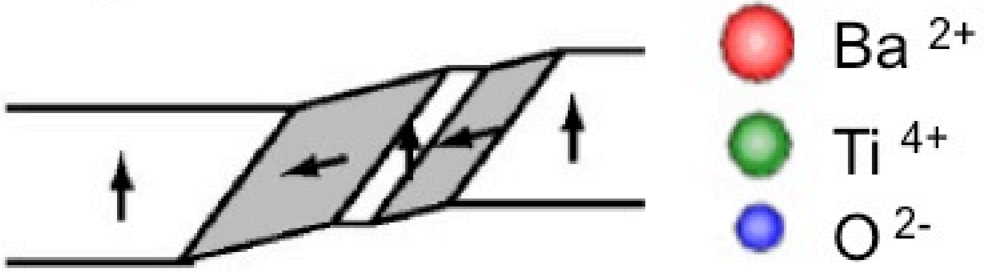
Confocal pol. optics

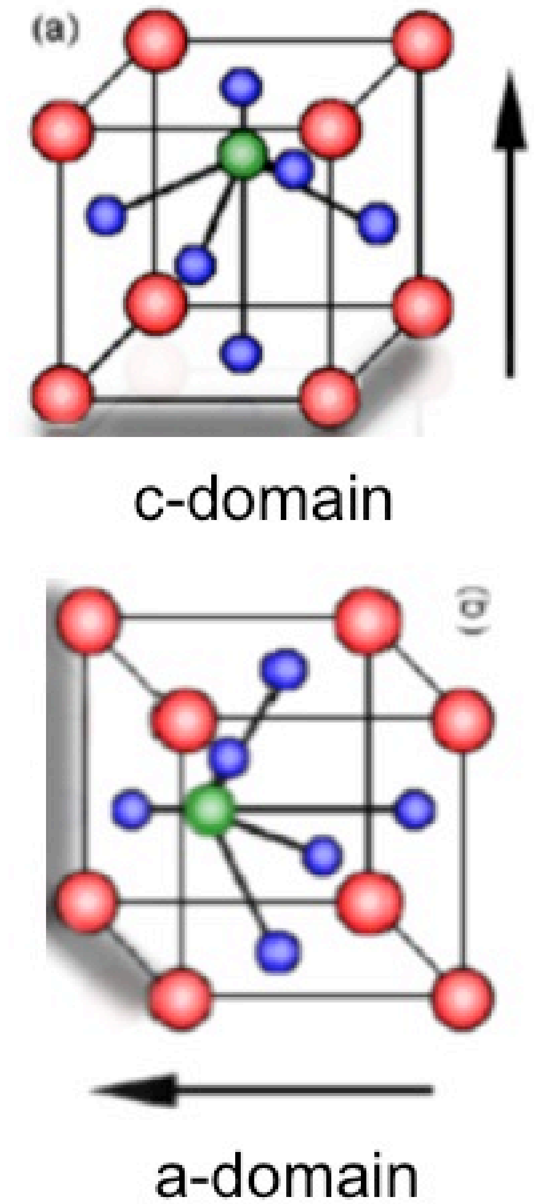


PFM

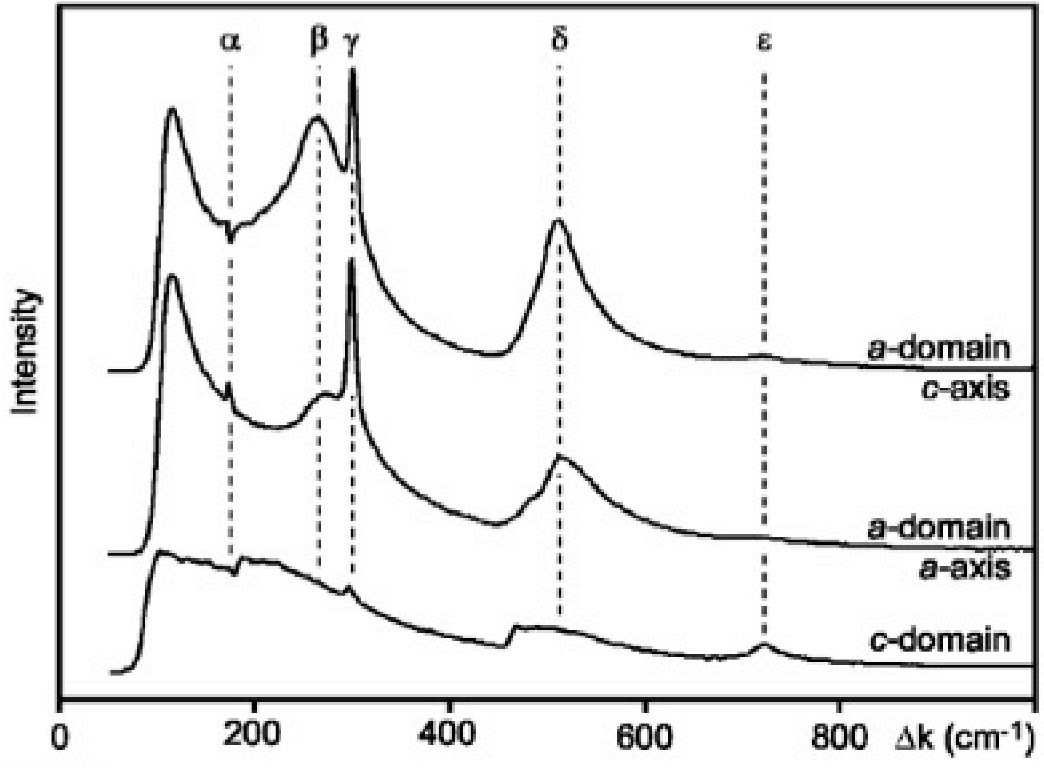


Region B



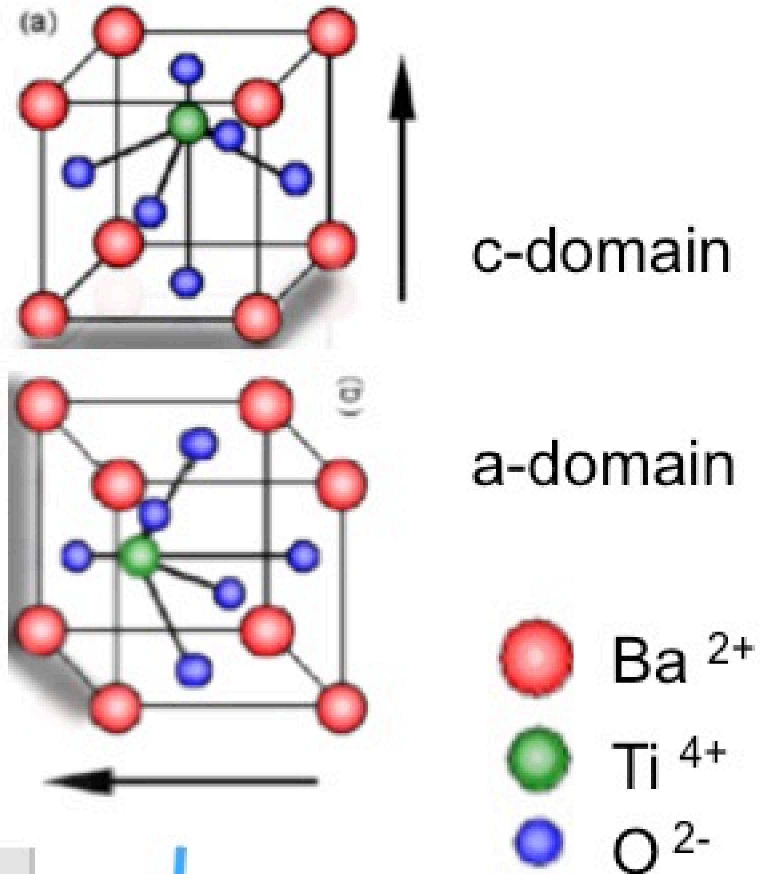


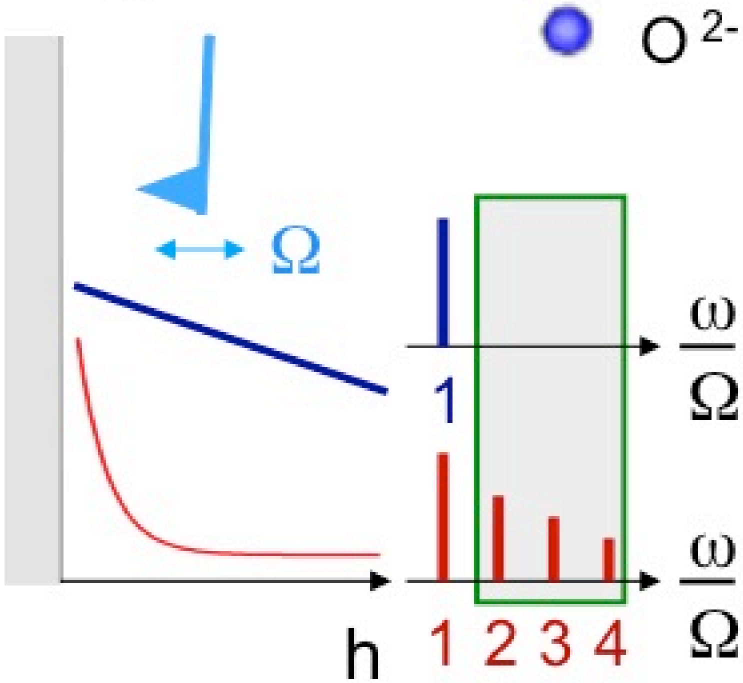
µ-Raman spectroscopy



G. Tarrach et al., *APL* (2001)

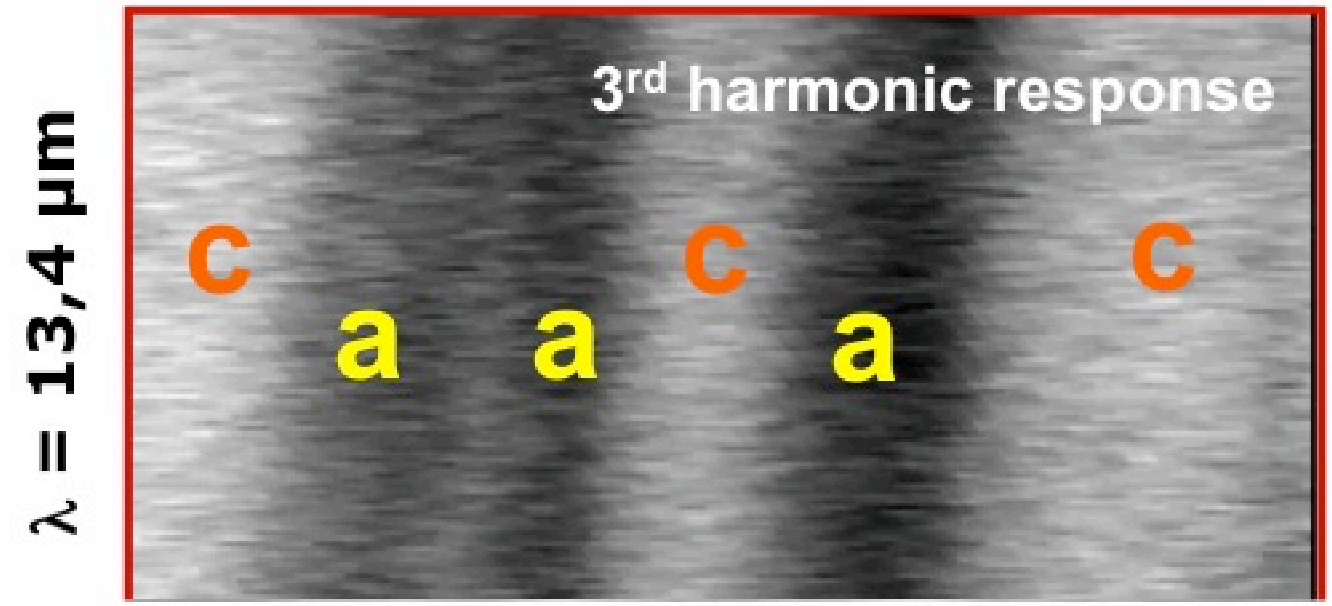
IR-s-SNOM microspectroscopy





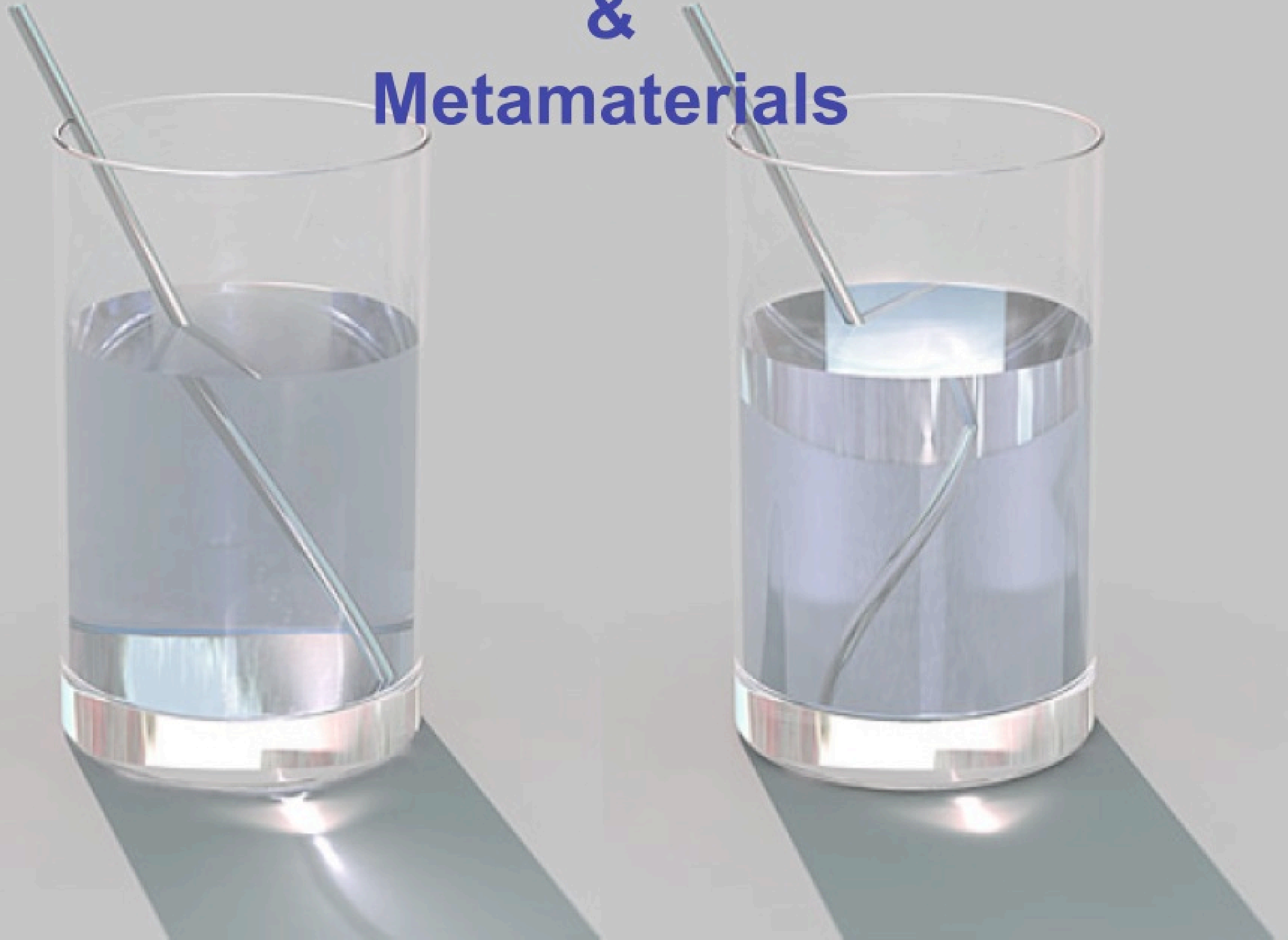
S. Kehr et al., *PRL* (2008)

$BaTiO_3$:
- atomically flat surface
- anisotropic $\hat{\varepsilon}_s$
- domains



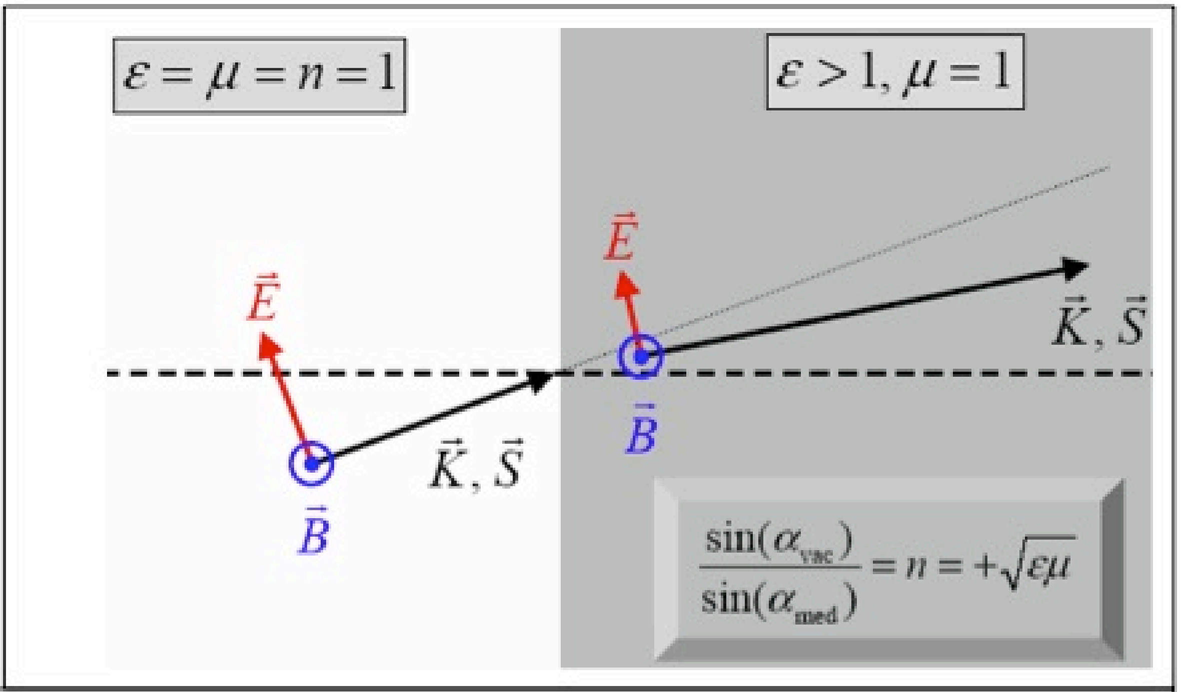
optical resolution: λ/500

#4: Perovskite Superlensing
&
Metamaterials

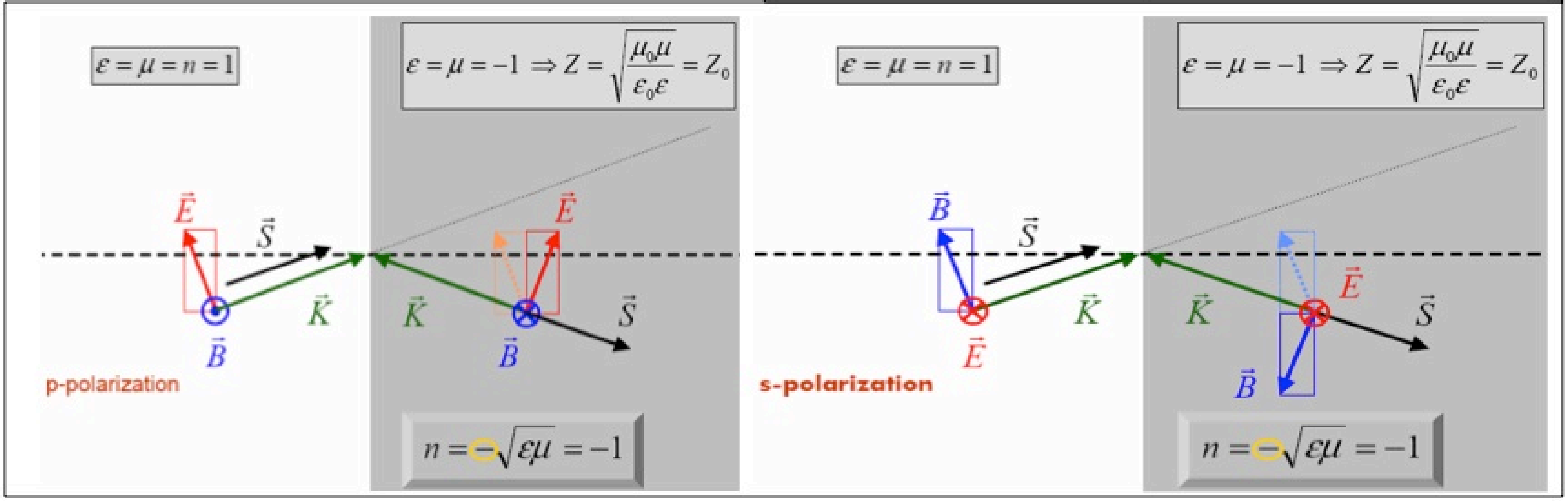


Optical refraction

RHM: right handed material
positive refractive materials



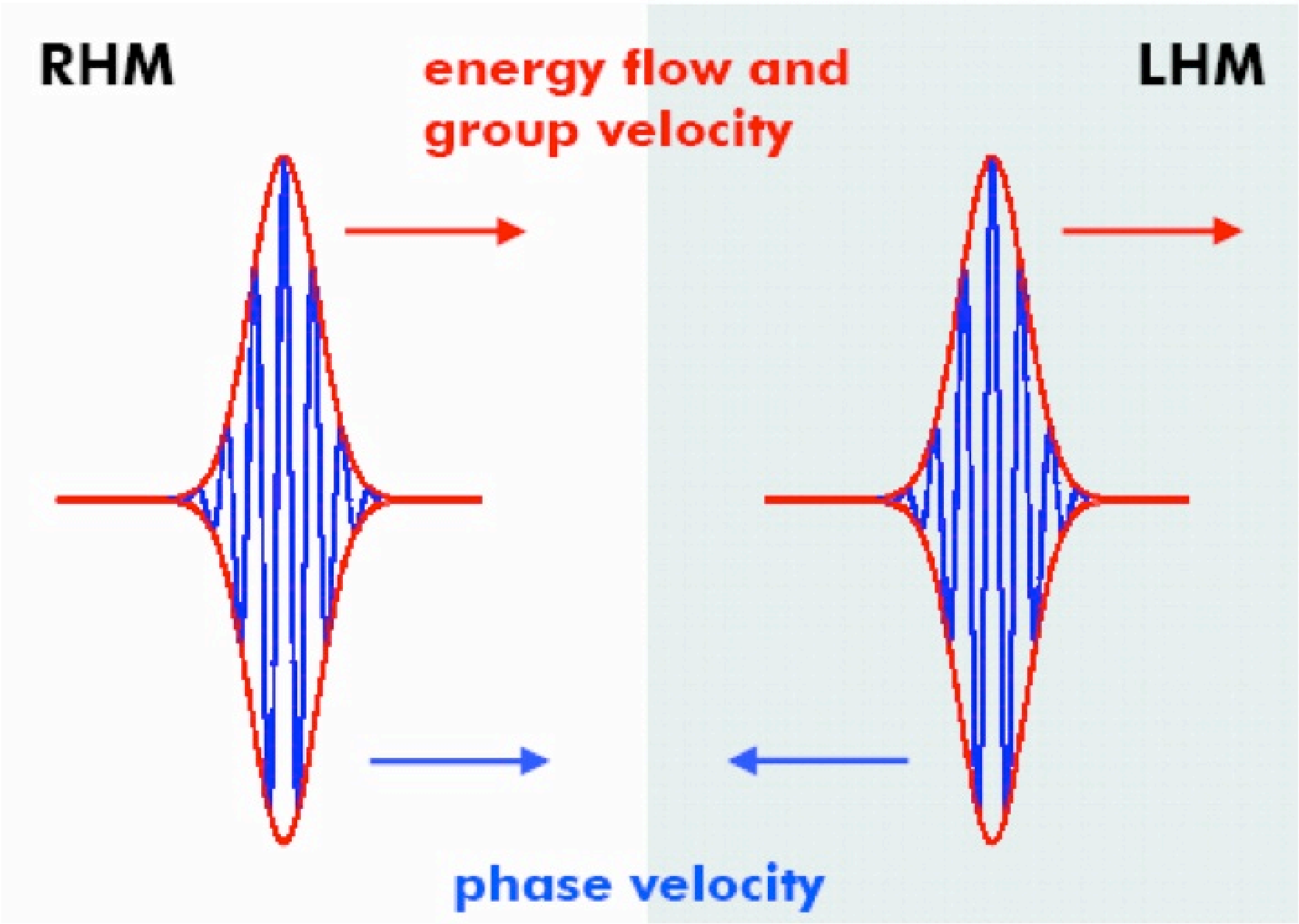
LHM:
left handed material
negative refractive material



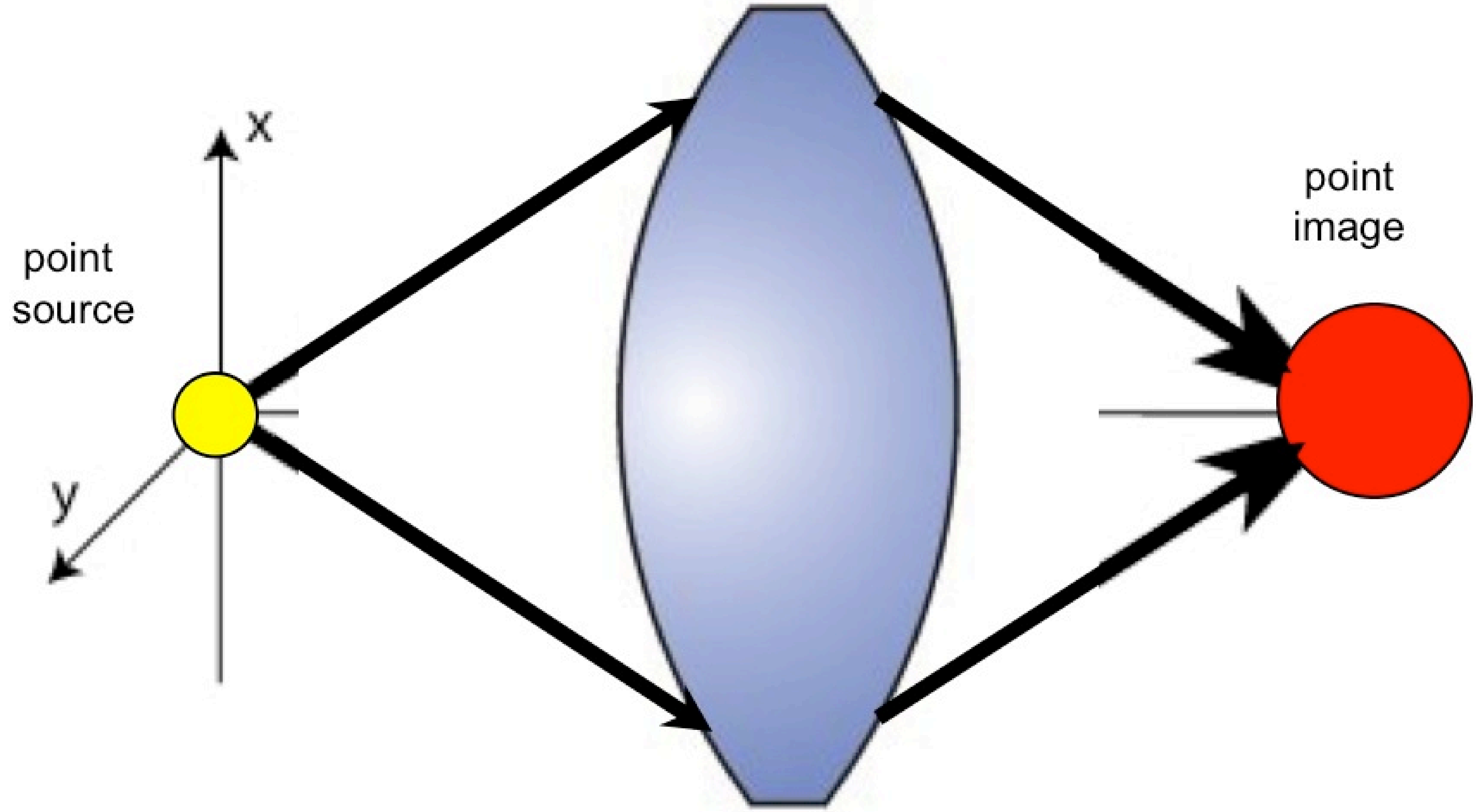
J.B. Pendry, *PRL* **85** (2000) 3966

Phase/group velocity in LHM

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

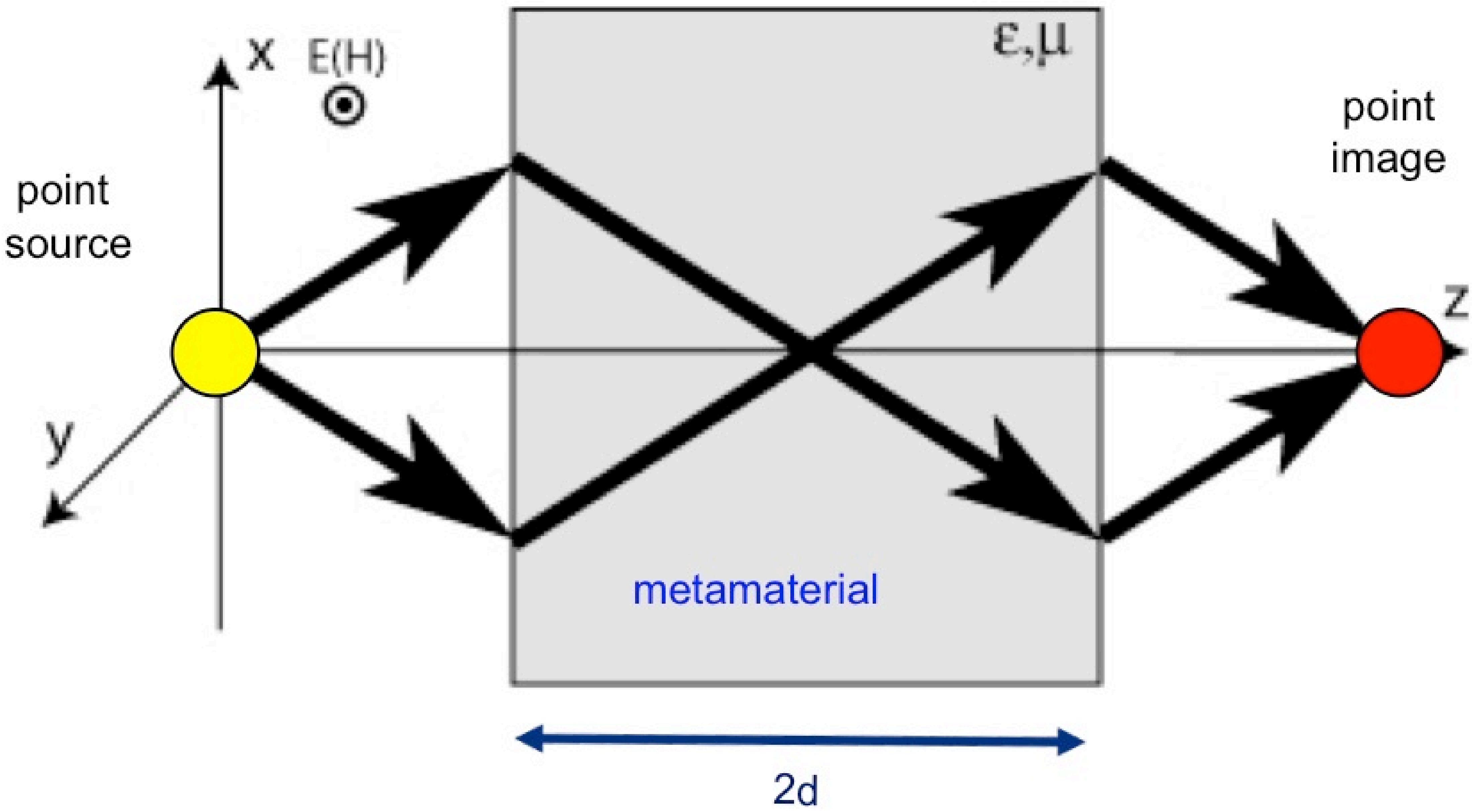


Superfocusing with RHMs



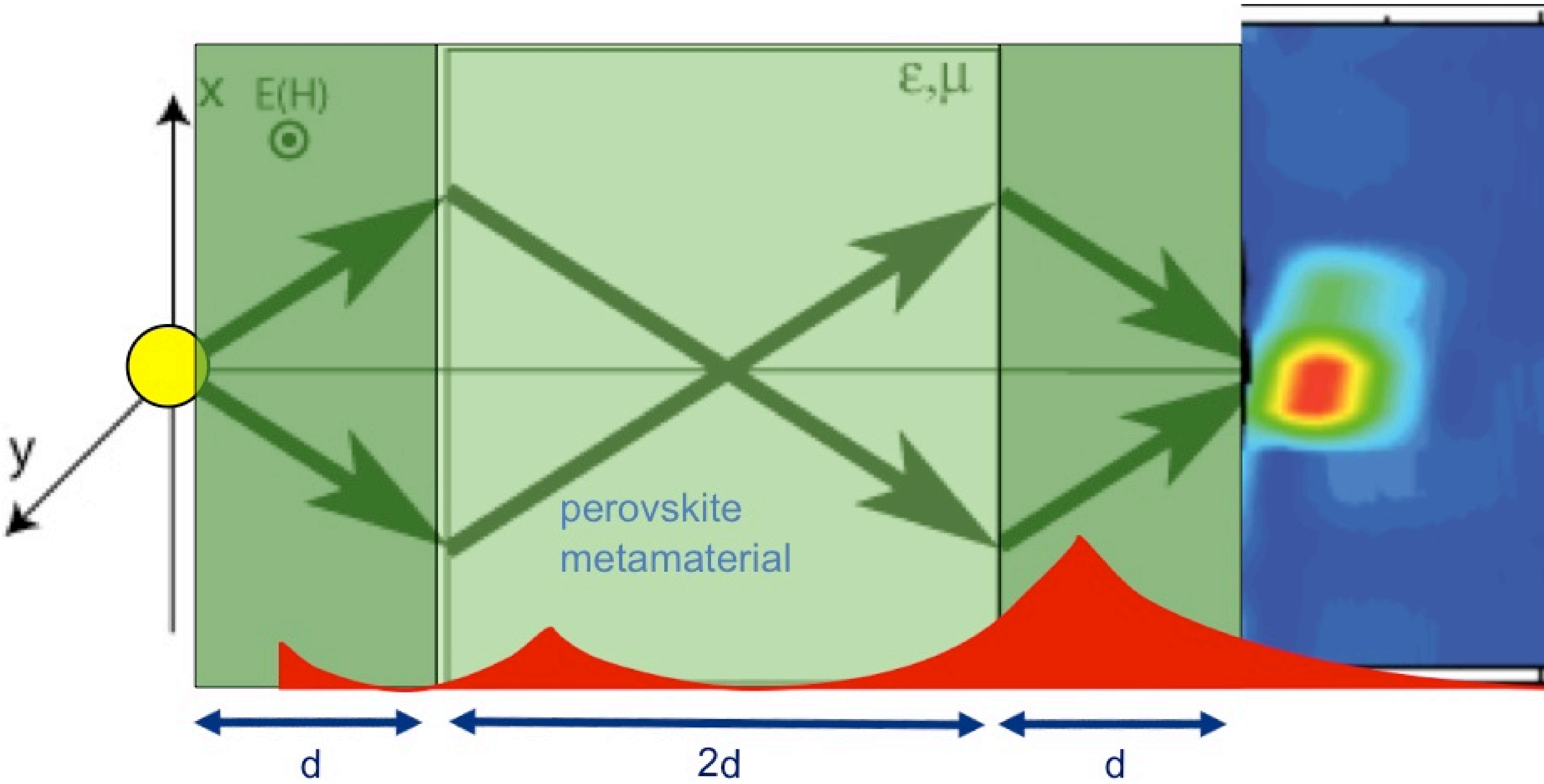
J.B. Pendry, *PRL* **85** (2000) 3966

Superfocusing with LHMs



J.B. Pendry, *PRL* **85** (2000) 3966

Superfocusing with LHMs



S.C. Kehr et al., *Nature Comm.* (2011)

IR-perovskite superlense

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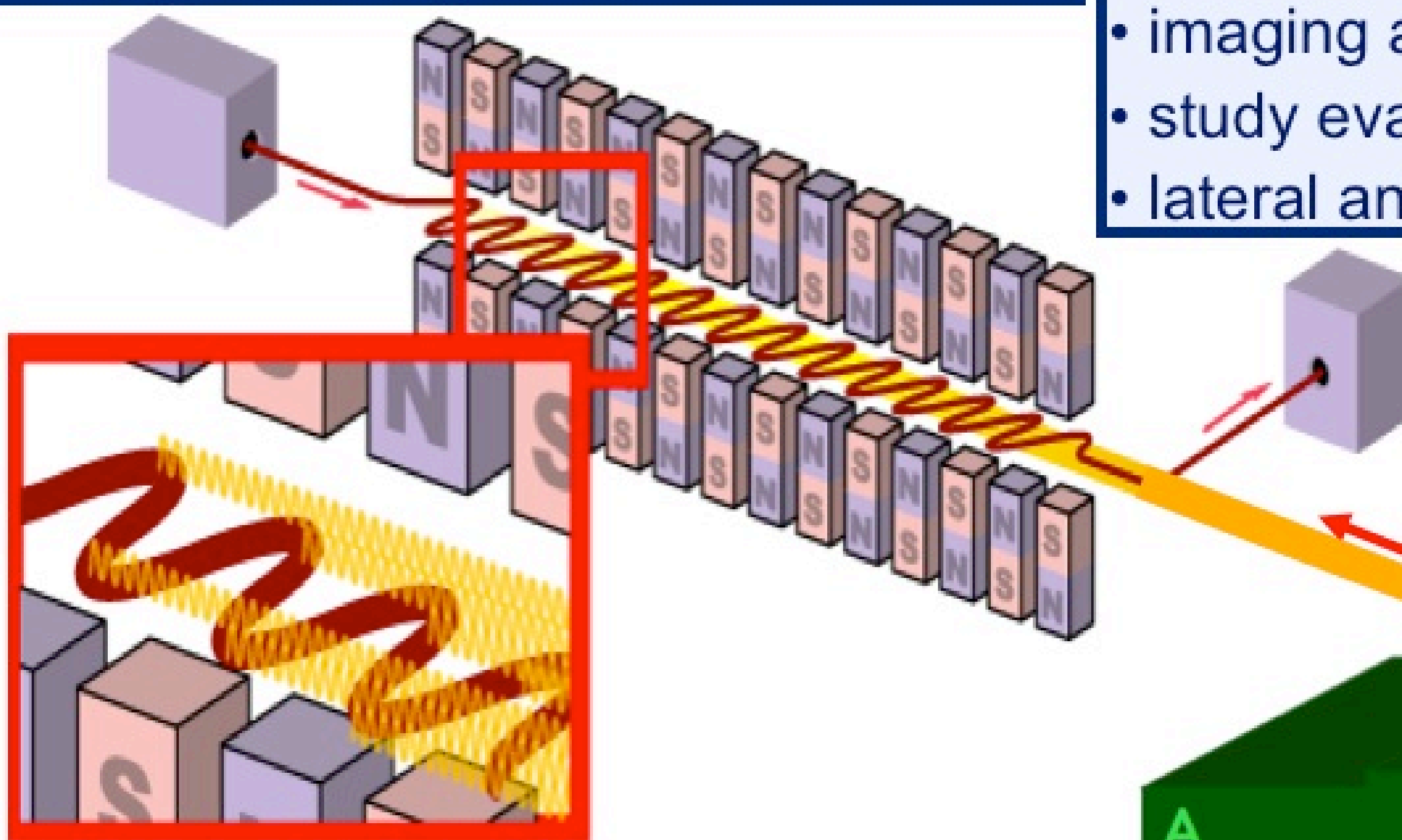
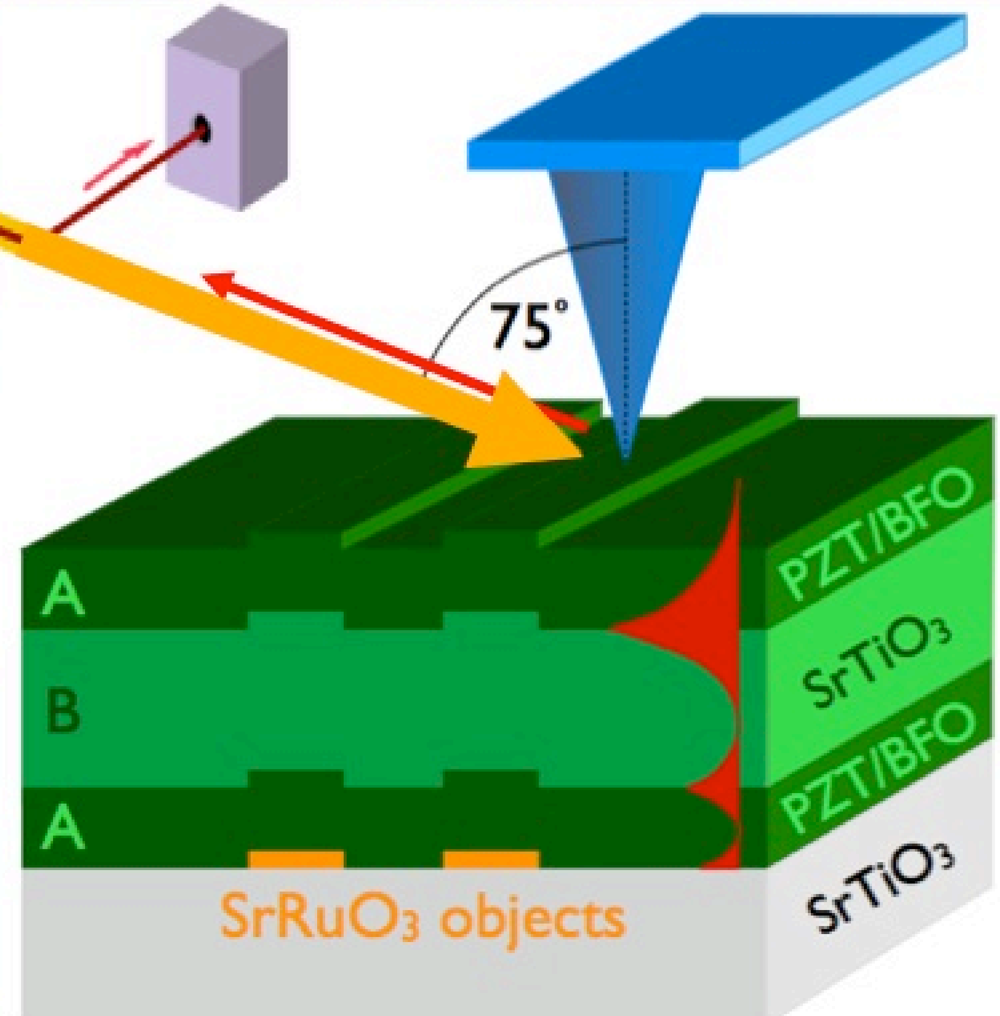
Free-electron laser

wavelength tunable from 4 to 250 µm
- ⇒ phonon excitation
- ⇒ spectral information

Scattering-type NSOM

- high resolution below diffraction limit
- imaging and spectroscopy
- study evanescent fields only
- lateral and vertical field distribution





Perovskite oxide-based superlens

- low absorption
- epitaxial growth: reduced scattering
  - ⇒ low loss

Superlens Sample Design

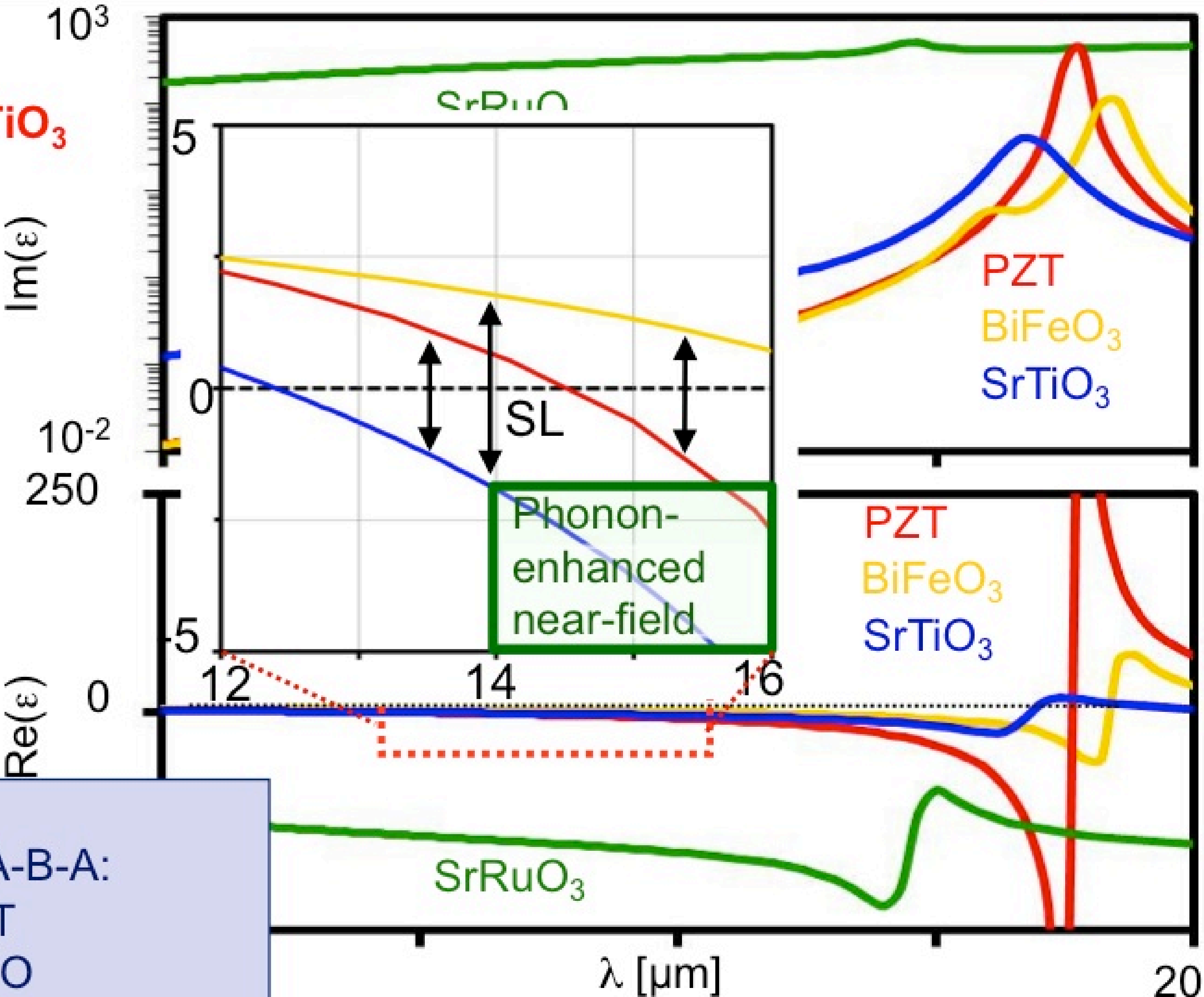
Layered perovskite thin film oxides: (d = 200 nm)

- Epitaxial growth
- Pairs with **Re(ε_A)= - Re(ε_B)**
- Im(ε) small

S.C. Kehr et al., *Nature Comm.* (2011)

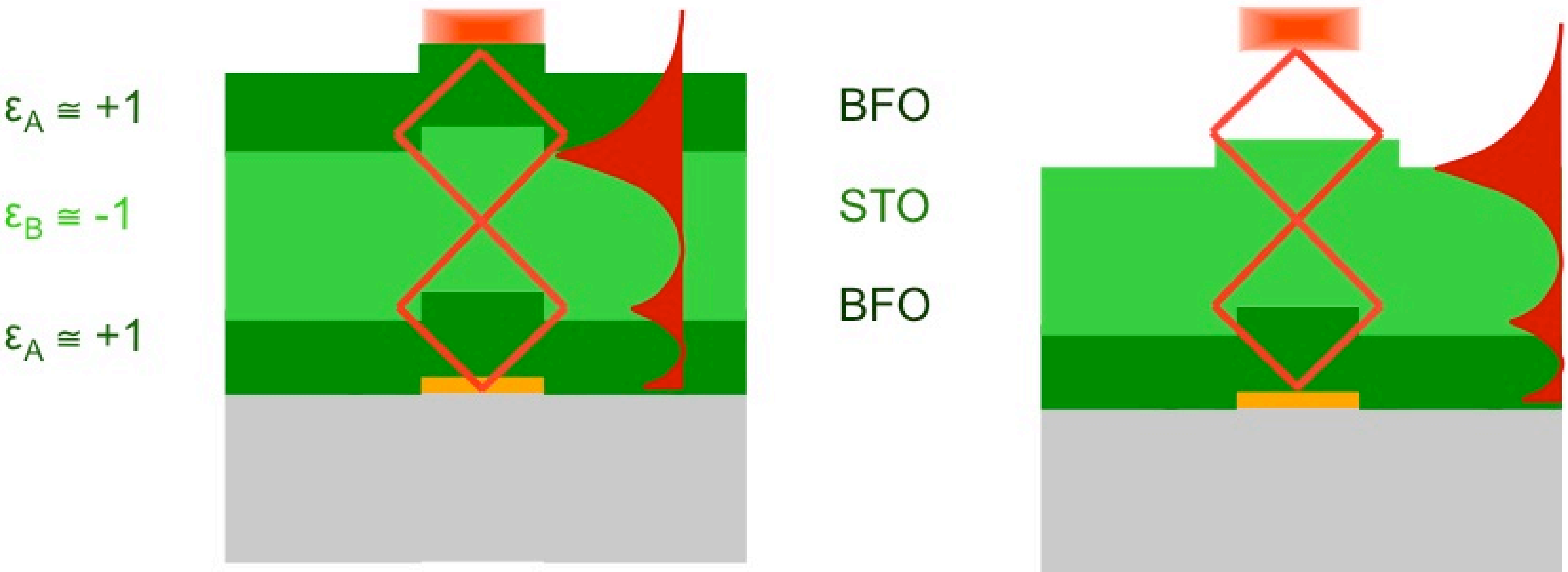
Superlens Sample Design

PZT: $PbZrO_3/PbTiO_3$
BFO: $BiFeO_3$
STO: $SrTiO_3$



Possible pairs A-B-A:
- PZT-STO-PZT
- BFO-STO-BFO
- BFO-PZT-BFO

Sample Design



<u>3-layer superlens</u>

- Image plane at sample surface

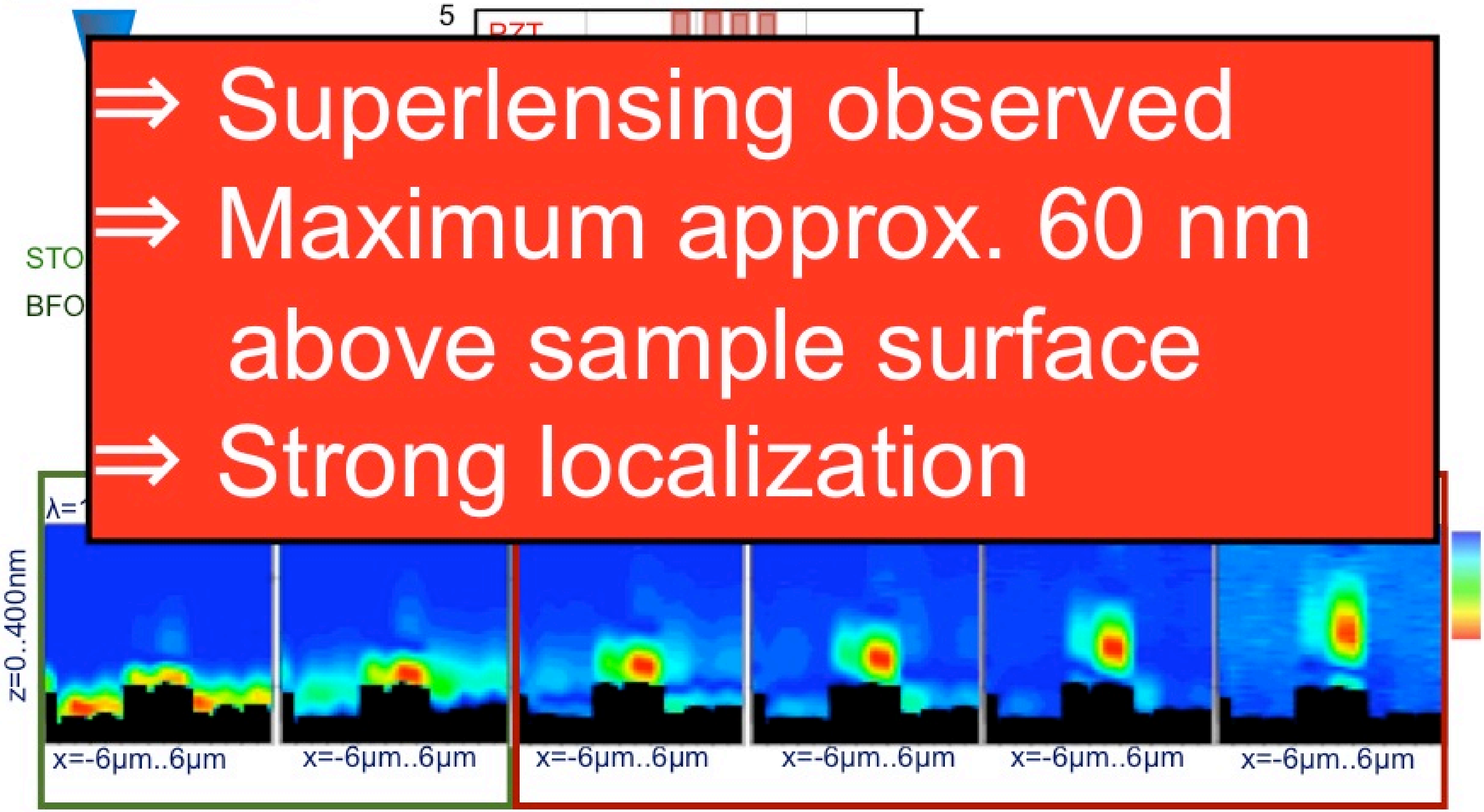
<u>2-layer superlens</u>

- Focus at certain distance above sample
- Polariton mode study

S.C. Kehr et al., *Nature Comm.* (2011)

Cross sections

2-layer superlens:



⇒ Superlensing observed

⇒ Maximum approx. 60 nm above sample surface

⇒ Strong localization

S.C. Kehr et al., *Nature Comm.* (2011)

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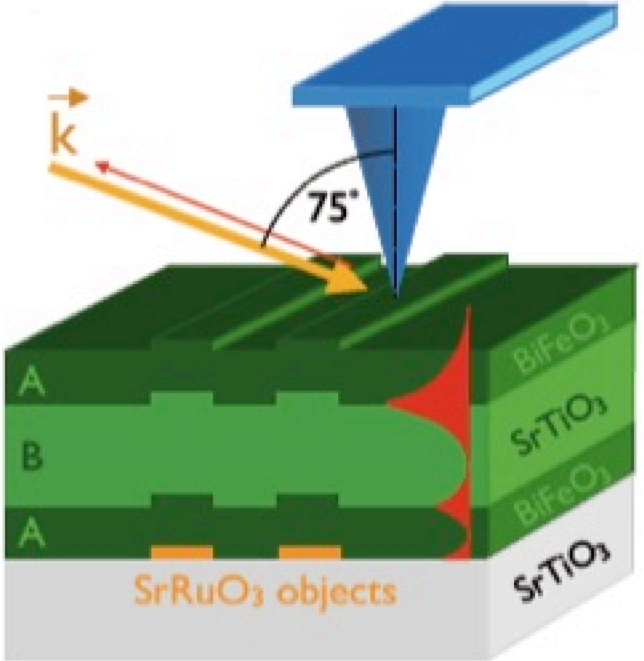
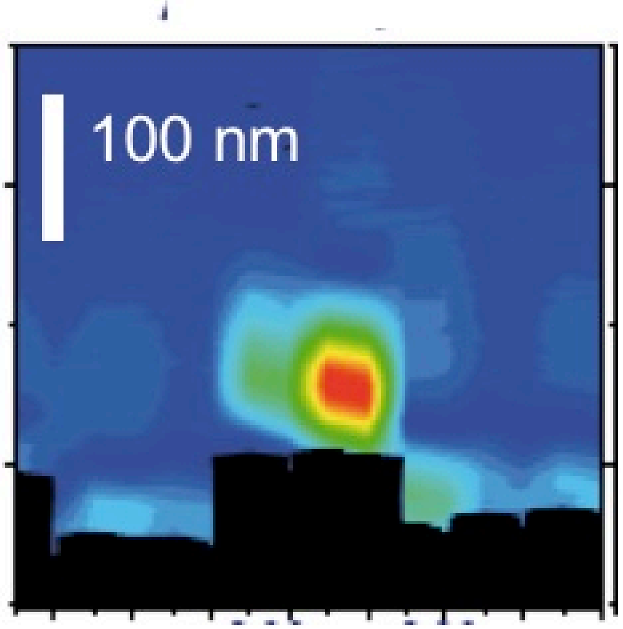
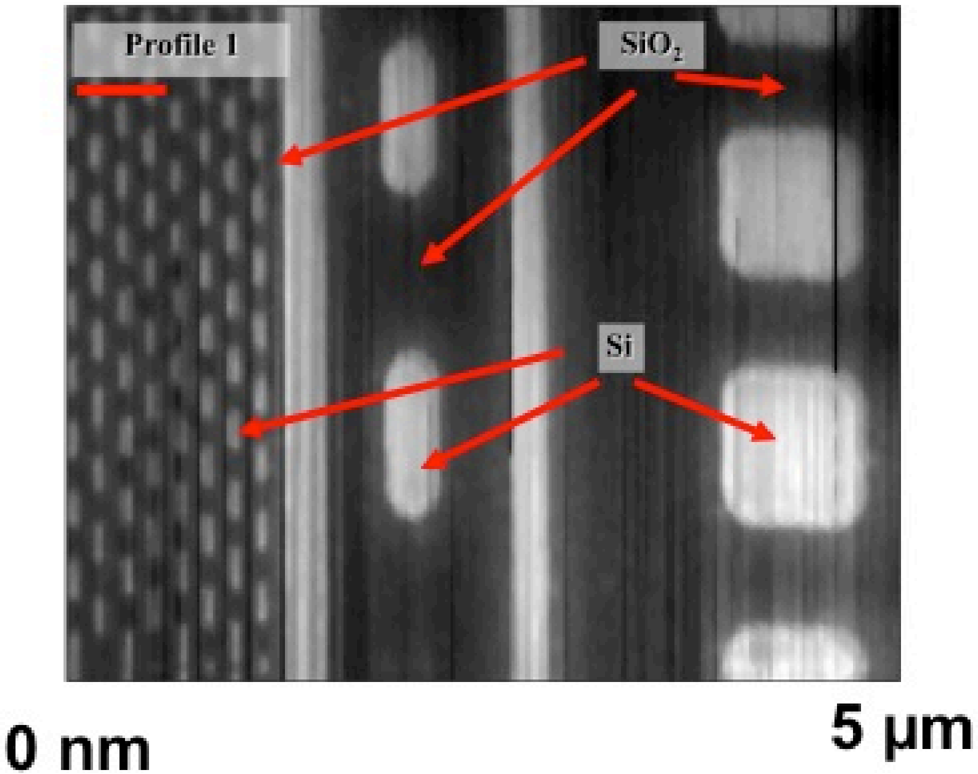
Conclusions

iapp

- scattering SNOM bases on nc-AFM
- optical resolution < λ/1000
- IR, visible, UV, ...
- technological applications & fundamental science



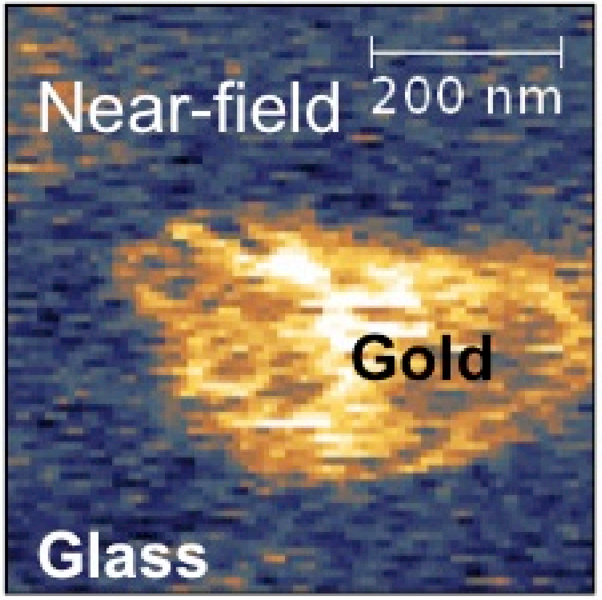




Outlook

- **THHz-IR-s-SNOM**

➢ **Resolution: λ/5000 @ 1,3 THz**



- **use time structure of FEL pulses**
- **push optical resolution to 1 nm**
- **application to molecules and organic structures**

Acknowledgment

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