Project A: Fundamentals of scattering for industrial applications

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Complex Photonic Systems
University of Twente

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Workpackage A

A. Fundamentals of scattering for industrial applications	A1 Scattering in industrial regime
	A2 Anisotropic volume scattering
	A3 Scattering and conversion

Interaction of light with matter

We need understanding
We need predictability

Our matter is complex

Usually not transparent

Usually no symmetry present

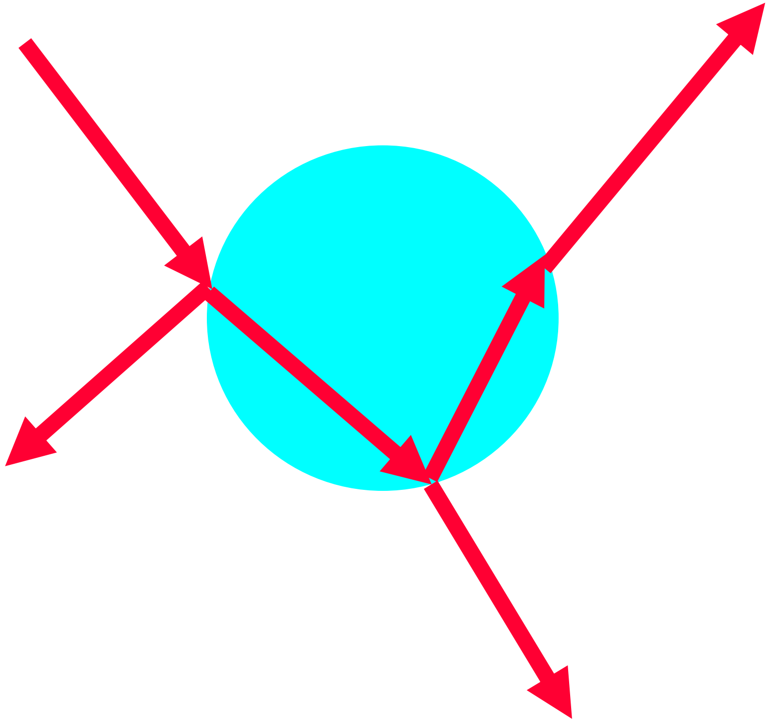
Autistic physicists

Physicists prefer samples with:
- no absorption
- isotropic scattering
- high symmetry

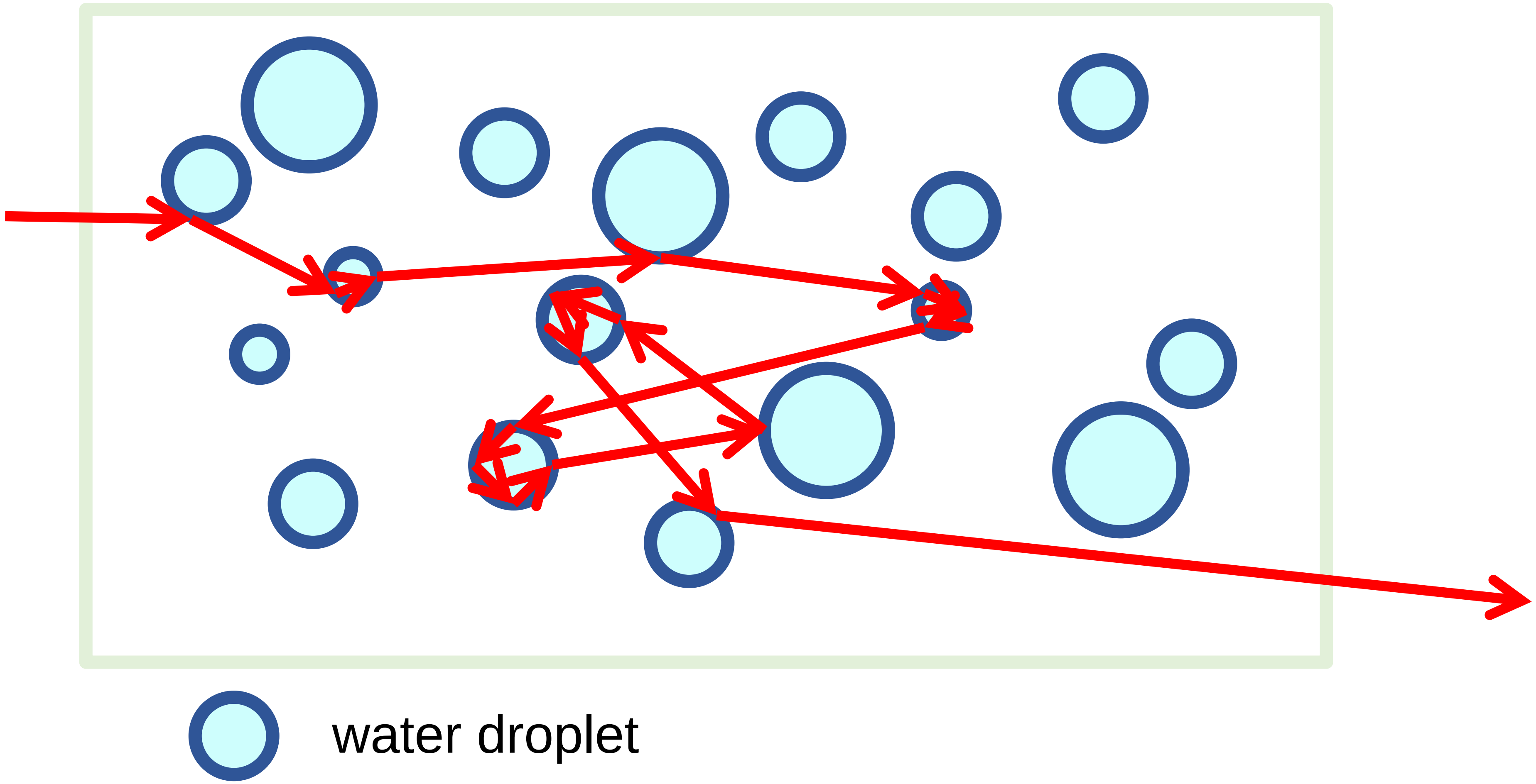
Real-world applications often involve materials with
- mild to strong absorption
- highly anisotropic scattering
- low symmetry

Rays in a droplet

contrast in index refraction

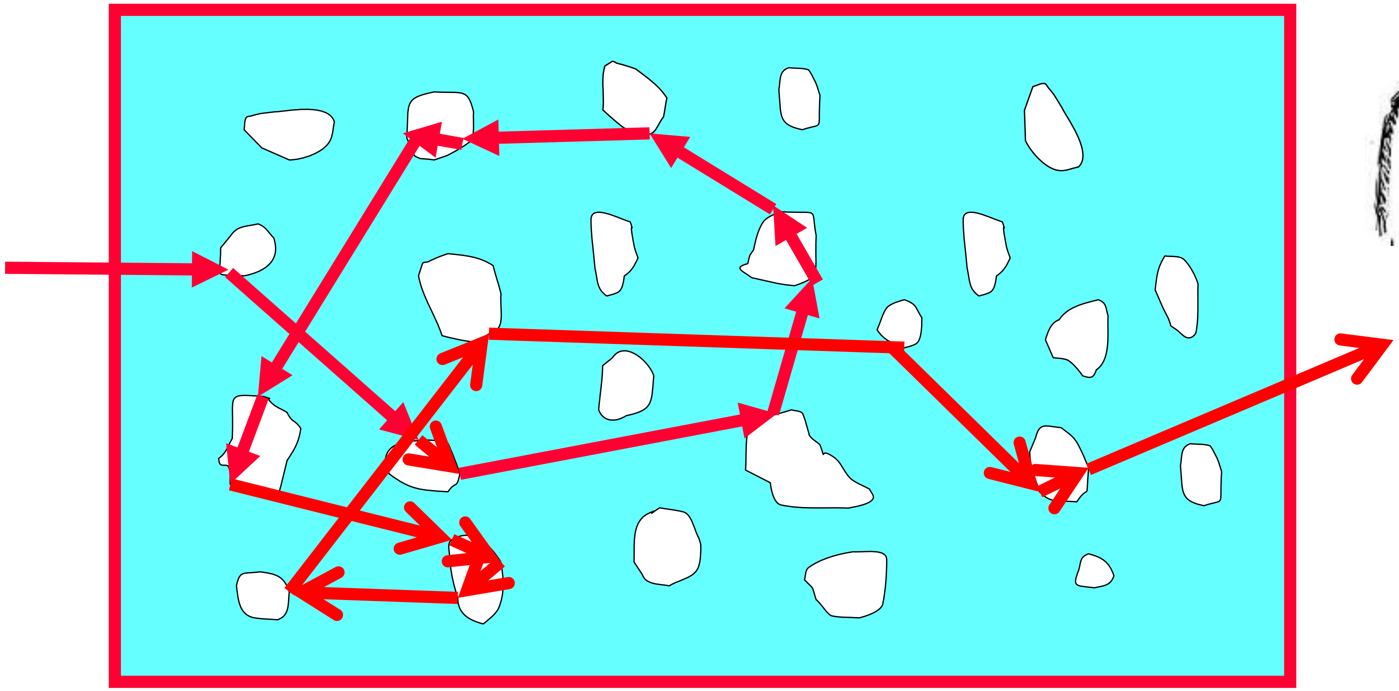


Multiple scattering in fog



Non-spherical obstacles

drunken sailor walk

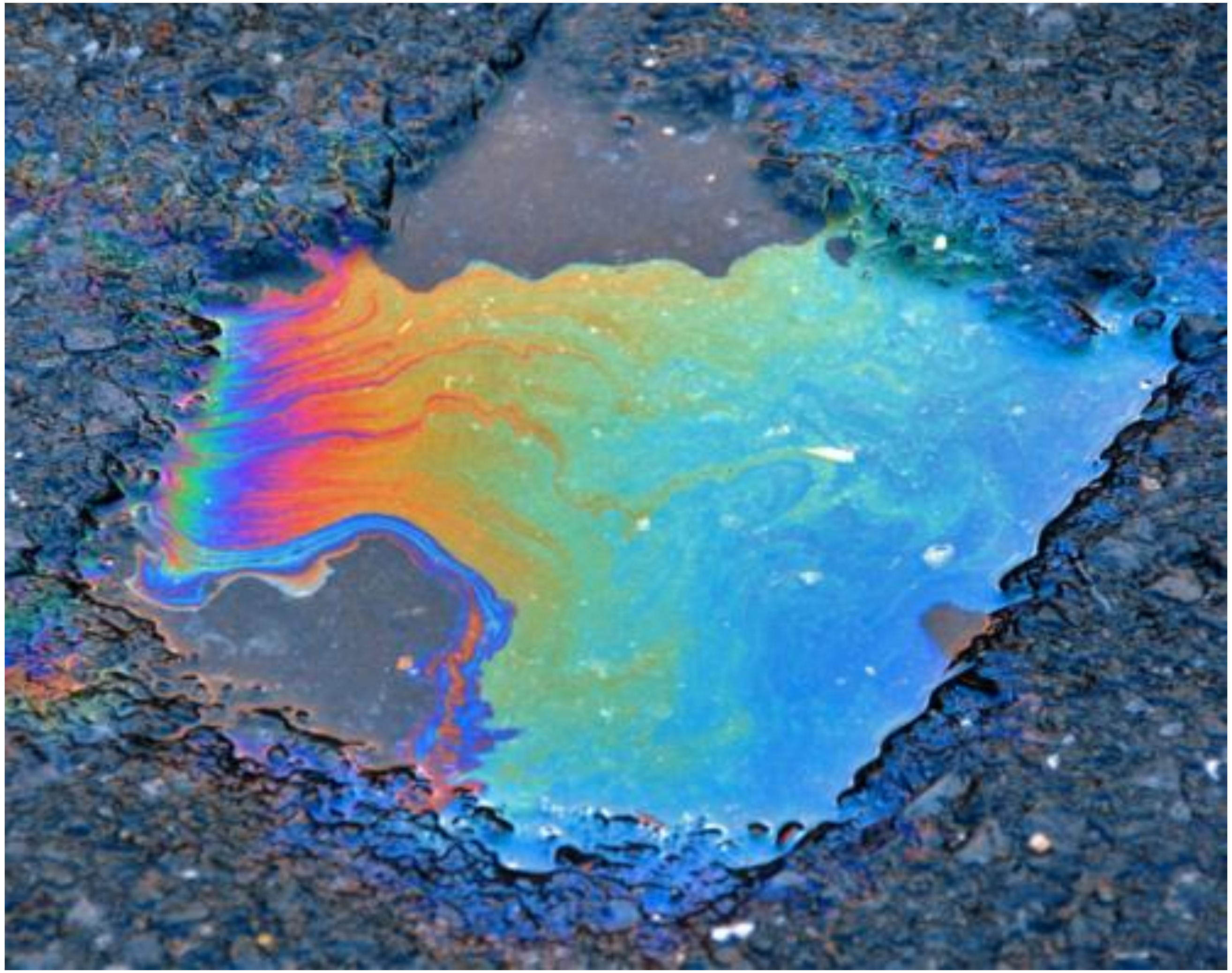


Witte suikerklontjes

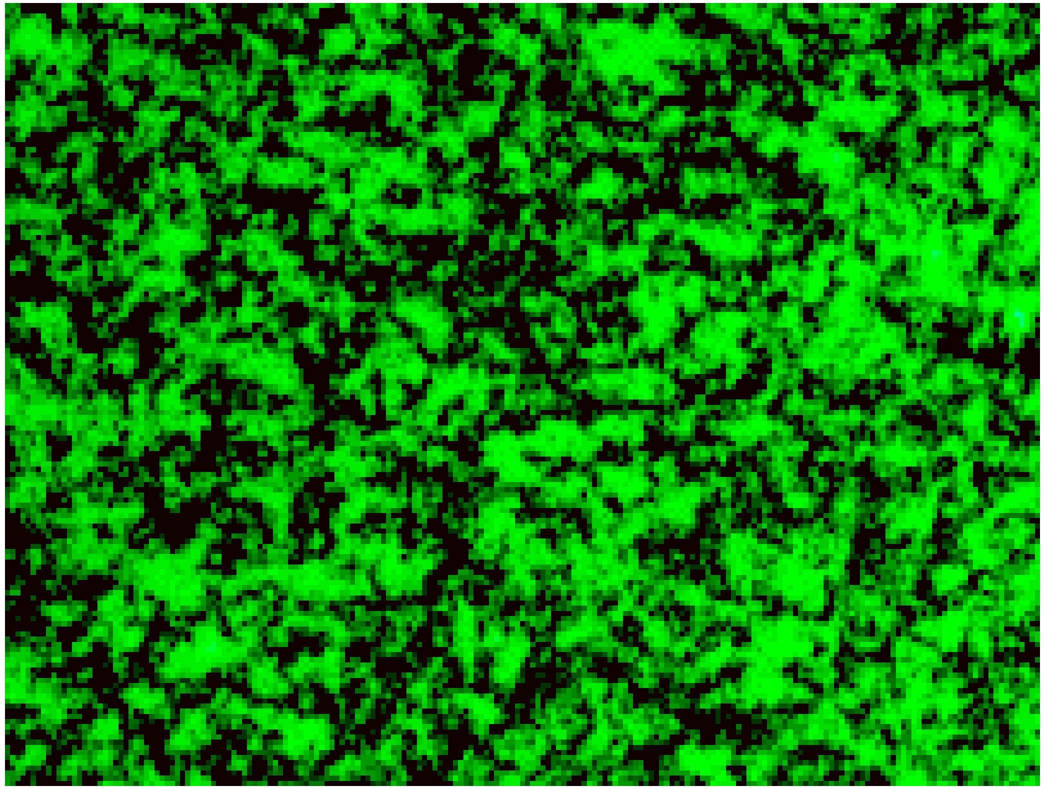
Rode suikerklontjes

Where is the interference?

Oil stain

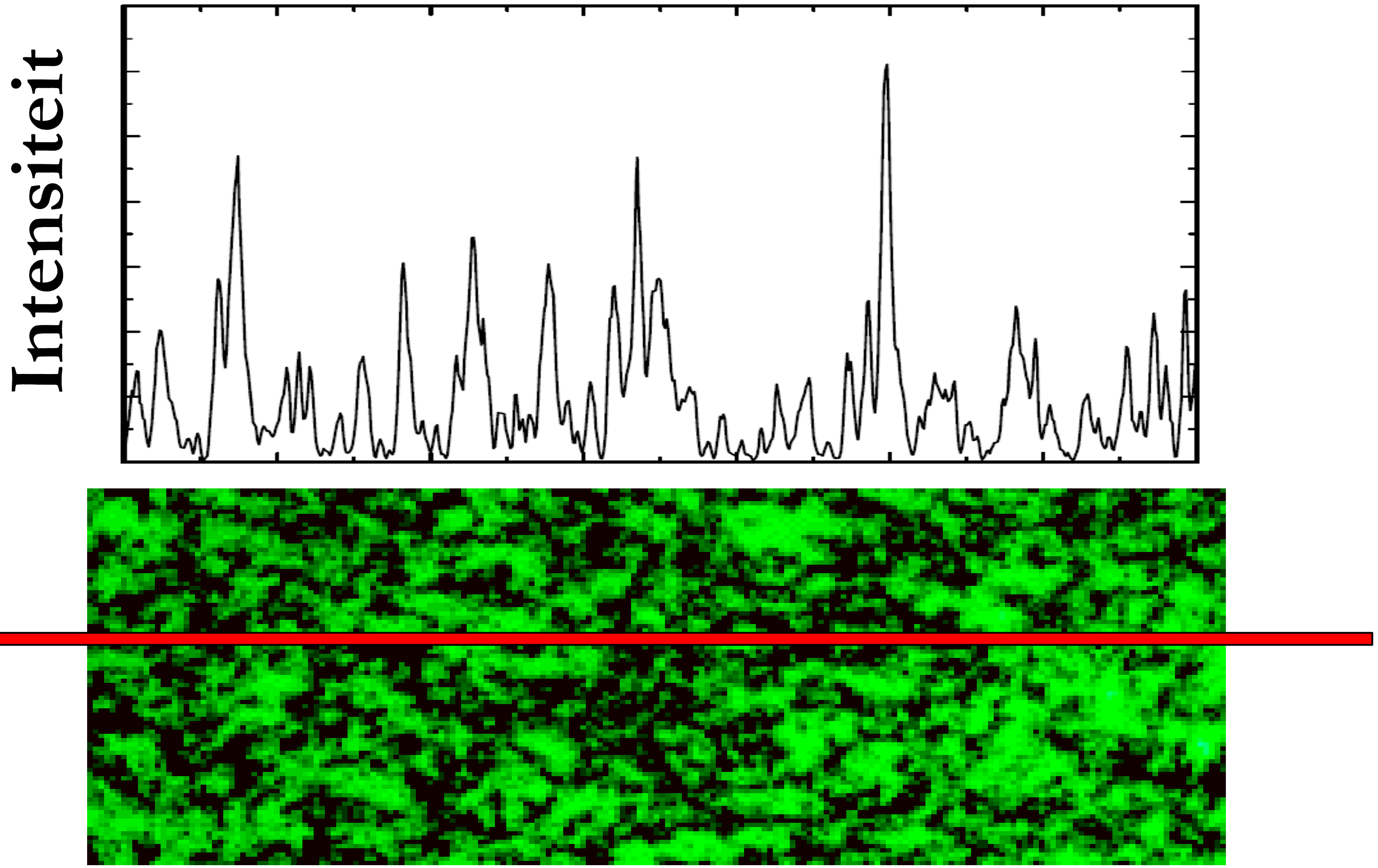


Speckle is interference



This dog is called Speckle,
because it is a speckled dog

Scanning a speckle



General solution: averaging

If you want to get rid of speckle: avegage
- geometric speckle -> shake your sample
- frequency speckle -> broad band light
- time speckle -> short pulses

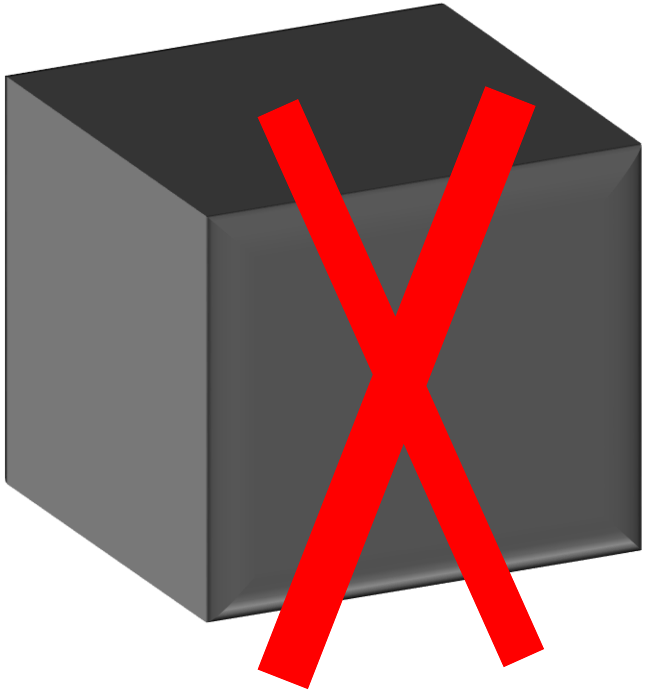
Average over realizations

Which exact theories do we have?

- homogeneous sphere (Mie solution)
- homogeneous slab
- collection of point scatterers (up to 10000)

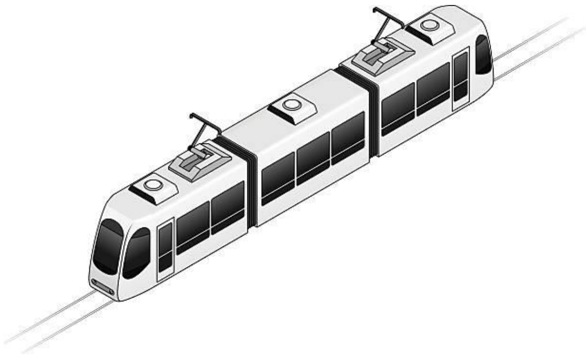
This far away from the materials
we are interested in

Not known exactly for a cube



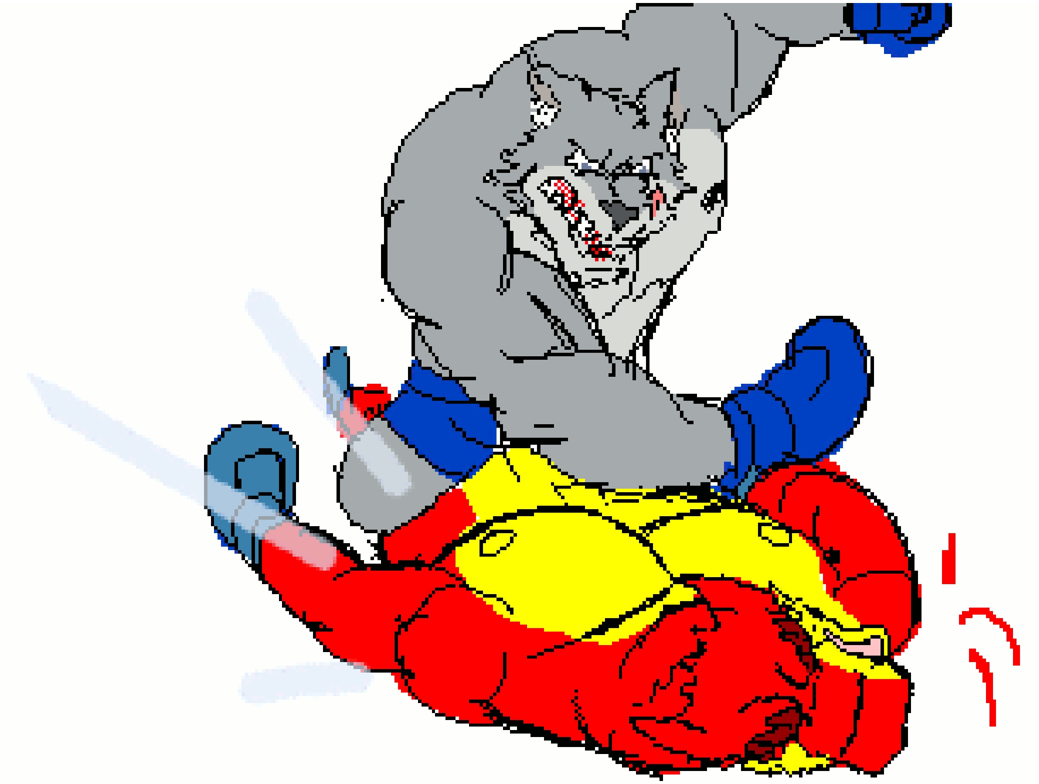
Approximations for light transport

- transport theory
- transport theory analytic
  - diffusion and beyond
- transport theory ray tracing



Ozan Akdemir

Bashing the diffusion approximation



Key diffusion parameters

- scattering **mean free path** ℓ_{scat}
- transport mean free path ℓ_{tr}
- **diffusion coefficients** D
- **system size** L
- absorption mean free path ℓ_{abs}

Crucial single particle properties

How much do the particles absorb?

What is the anisotropy of their scattering
(isotropic or very much forward)

Thomas Kotte

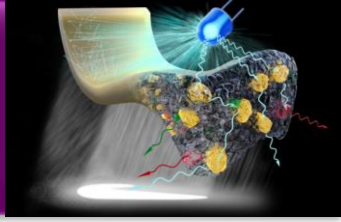
Exploiting interference

In the technique of wavefront shaping (invented in Twente) tens of thousands local phases of a wavefront are optimized to produce beneficial forms of scattering. This is a revolution in optics with many applications.

But not only exploiting scattering we can also exploit interference in the form of extinction (and so absorption) much to our advantage. For instance to measure shapes of scattering objects. (Alfredo Rates)

Have we delivered?

Project deliverables:
A1.1: Analytic model for transport in media with only scattering.
A1.2: Derive design rules from the model.
A2.1: Analytic model for transport in media with anisotropic scattering.
A2.2: Implementation of CSPs to obtain an optimized diffuser.
A3.1: Analytic model for transport in media with scattering, absorption, and conversion.
A3.2: Derive design rules from the model.
A1.3 & A3.3: Prototype of portable instrument to characterize mean free paths in industrial environments, input for models and design rules.

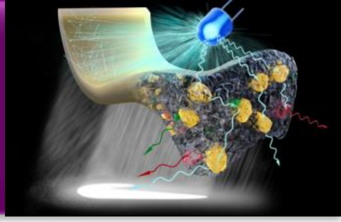


Breakdown of light transport models in photonic scattering slabs

Ozan Akdemir, Ad Lagendijk, Willem Vos

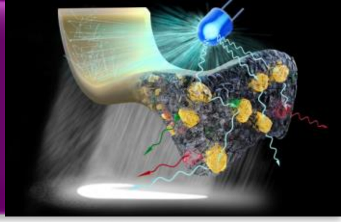
http://arxiv.org/abs/2110.15775

Outline



1. Transport theory & Radiative Transfer Equation for a slab
2. Unphysical ranges of P_N approximations
3. Comparisons to Monte Carlo Simulations
4. Summary & Outlook

Light transport through a scattering slab

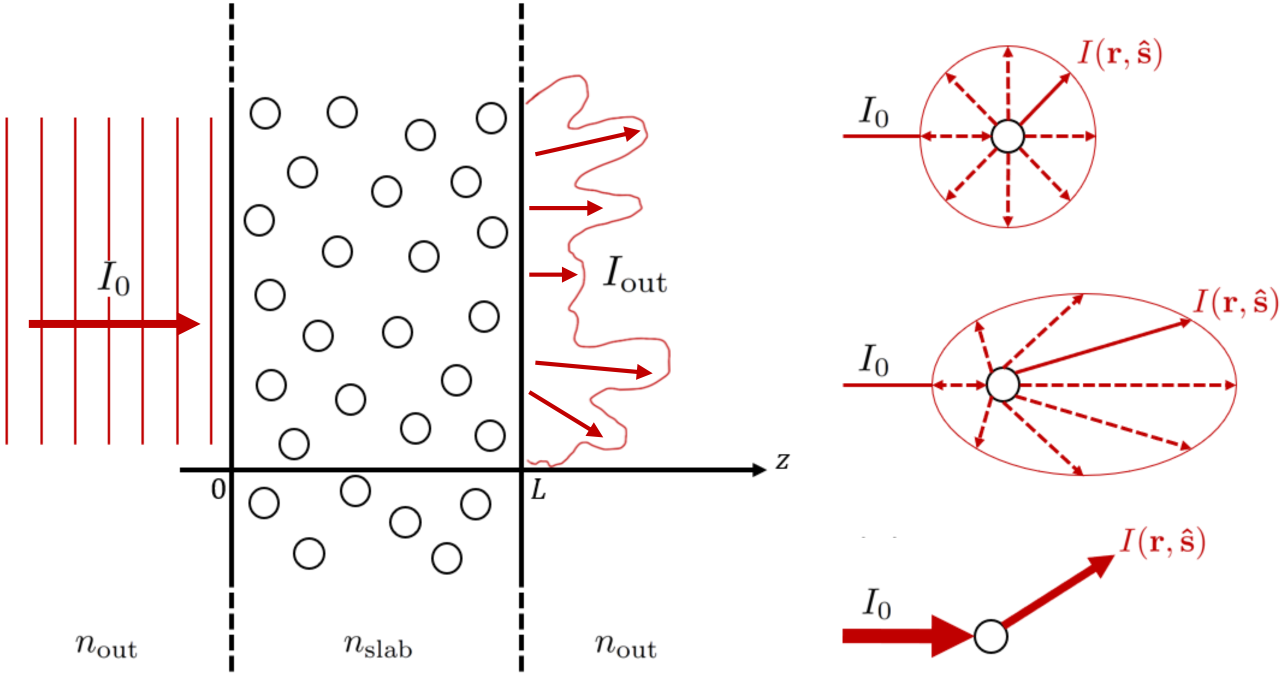


Monte Carlo: precise but tedious

Reliable analytical approximation needed, especially for industrial problems

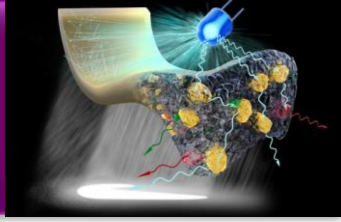
Transport theory: describe propagation of light in scattering media[1]

Analytic solutions not reliable for strongly anisotropic scattering and absorbing samples.



[1] Ishimaru, Academic, Vol. I (1978)

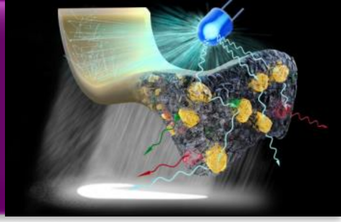
Radiative Transfer Equation for slab geometry



$$\mu \frac{dI_{\mathrm{dif}}(z,\mu)}{dz} = -\rho\sigma_{\mathrm{t}} I_{\mathrm{dif}}(z,\mu) + \frac{\rho\sigma_{\mathrm{t}}}{2}\int_{-1}^{1} a p(\alpha) I_{\mathrm{dif}}(z,\mu') d\mu' + \epsilon_{\mathrm{ri}}(z,\mu) + \epsilon(z,\mu)$$

Change in Specific intensity = Decrease + Gain + Source (outside) + Source (inside)

Outline



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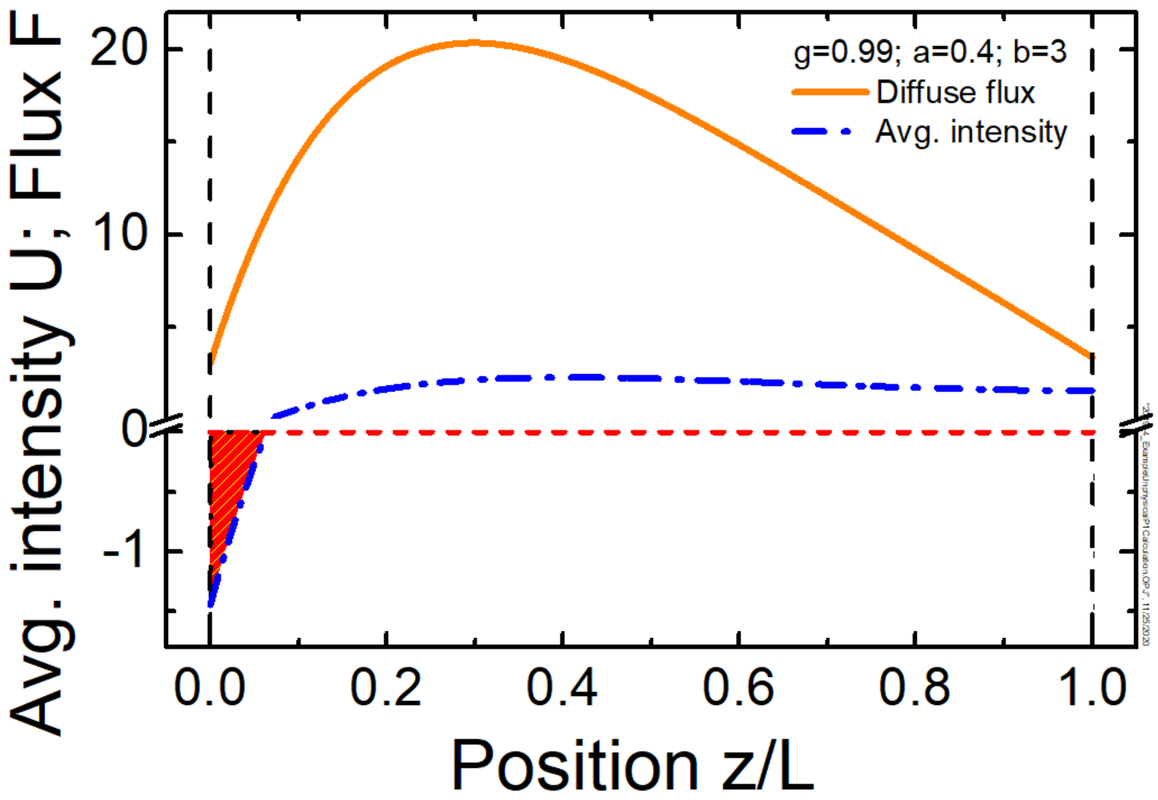
P_1, P_3 and $P_3 + \delta E(4)$

Unphysical calculation example

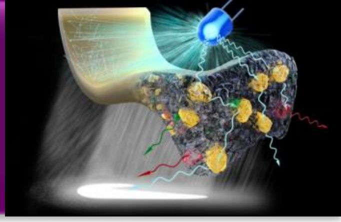
Calculated using P_1 (diffusion) approx.

Forward scattering & absorbing

Negative U → Unphysical



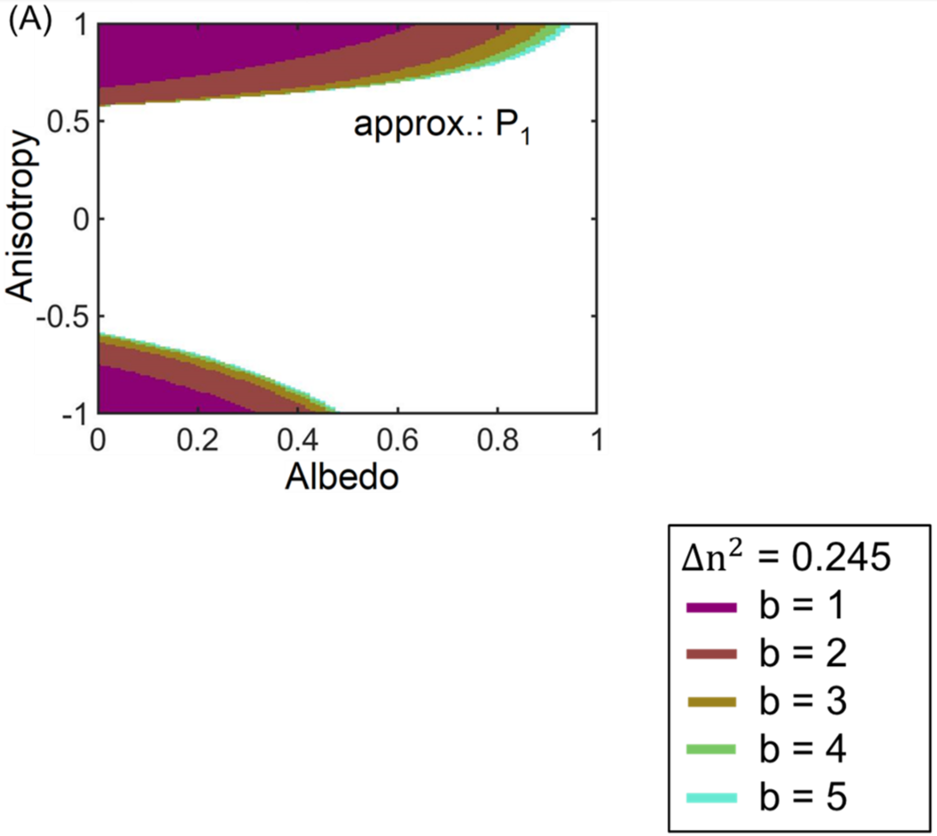
Unphysical ranges of P_1 approximation



Anisotropy (*g*) ⇒ $g \equiv < \cos\theta >$

Albedo (*a*) ⇒ $a = \frac{\sigma_s}{\sigma_t}$

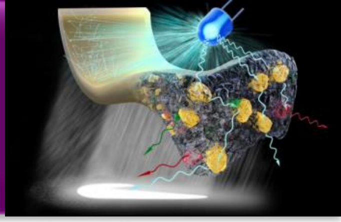
Optical thickness (*b*) ⇒ $b = \frac{L}{\ell_{\text{ext}}}$



Data points ⇒ Unphysical results

Would higher orders be better?

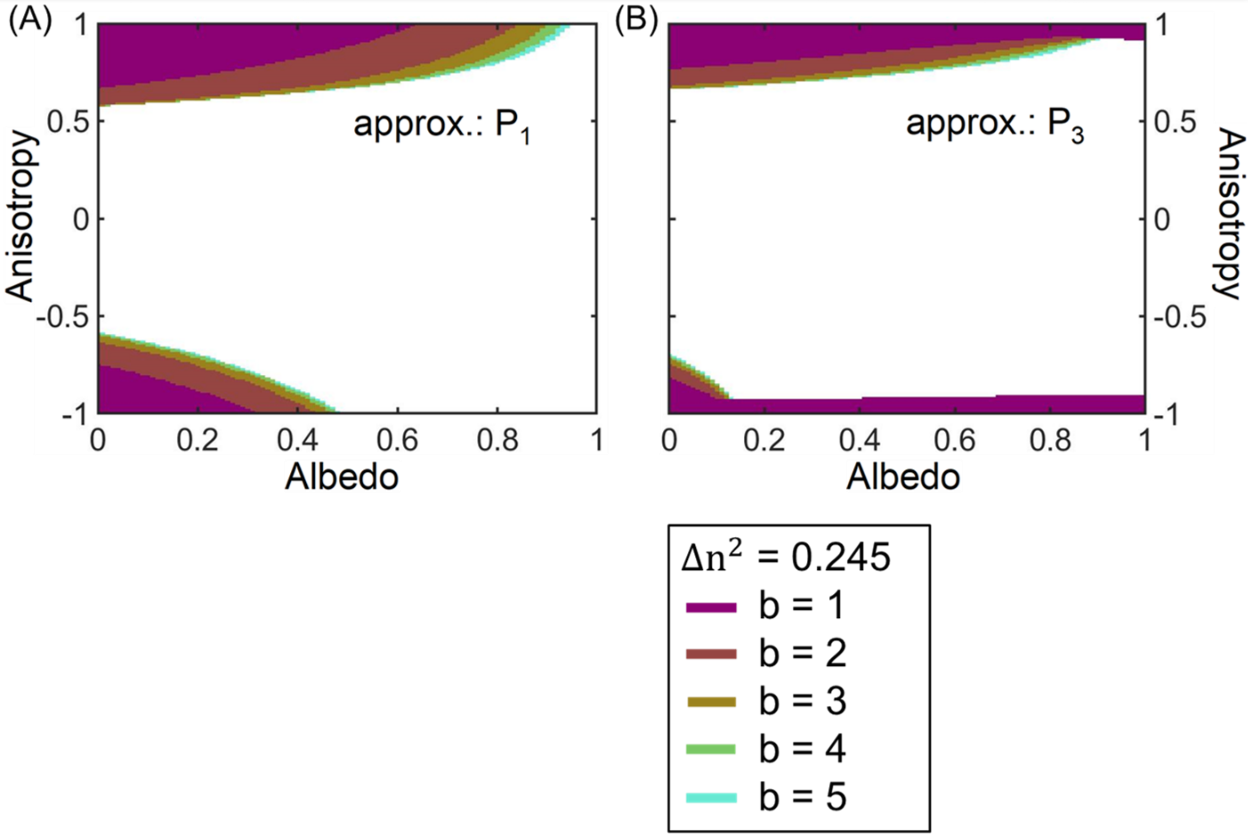
Unphysical ranges of P_3 approximation



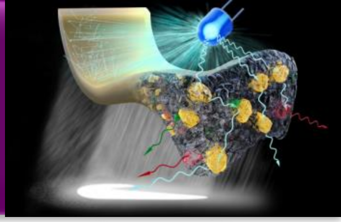
P_3 expected to improve
(Not really!)

Even worse for non-absorbing backscattering

Other options?



Unphysical ranges of $P_3 + \delta E(4)$ approximation

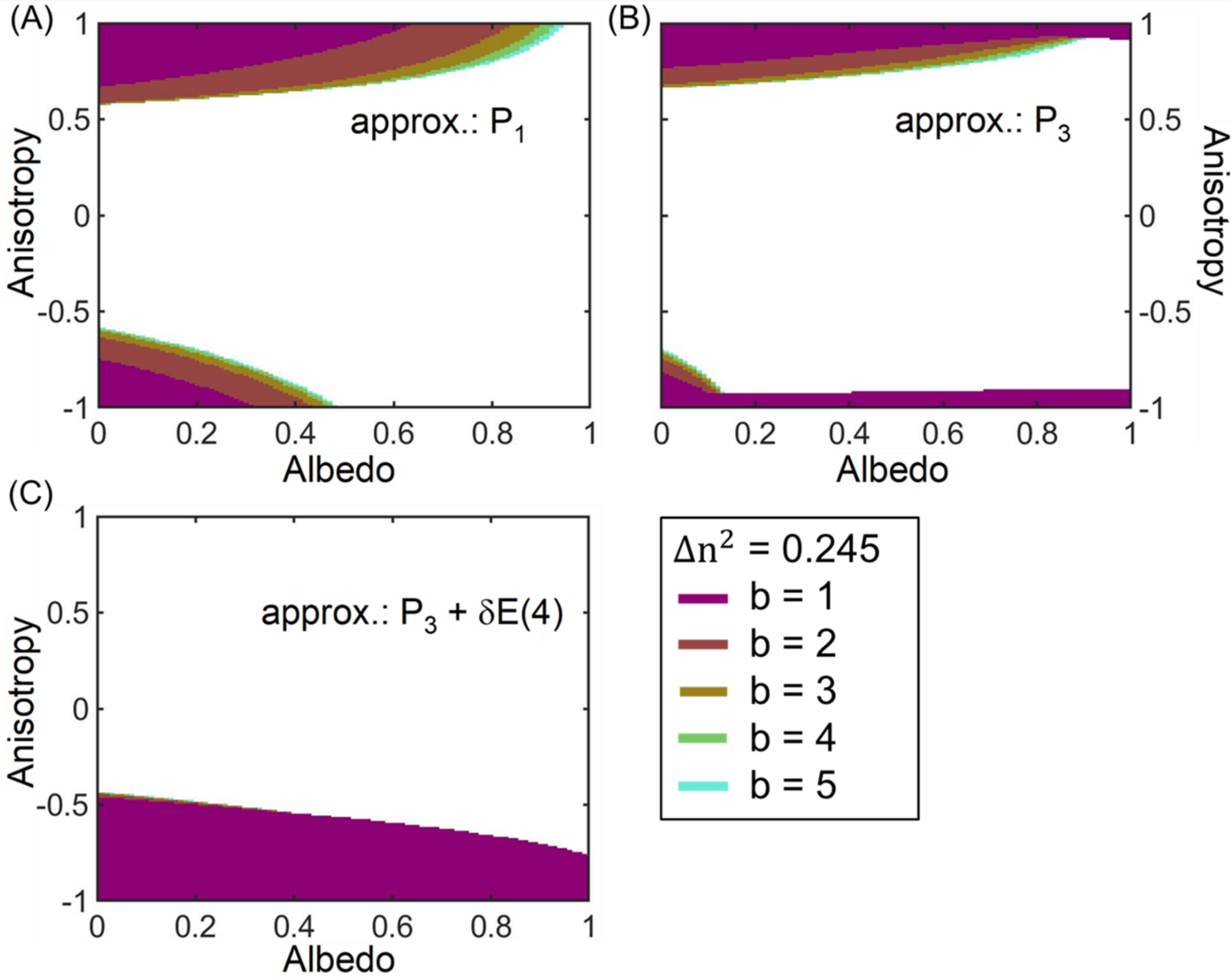


How about a correction for the forward scattering?

$P_3 + \delta E(4)$ improves forward scattering

Worse for backscattering

Fourth parameter needed to fully characterize unphysical ranges!



Unphysical ranges of P_N approximations

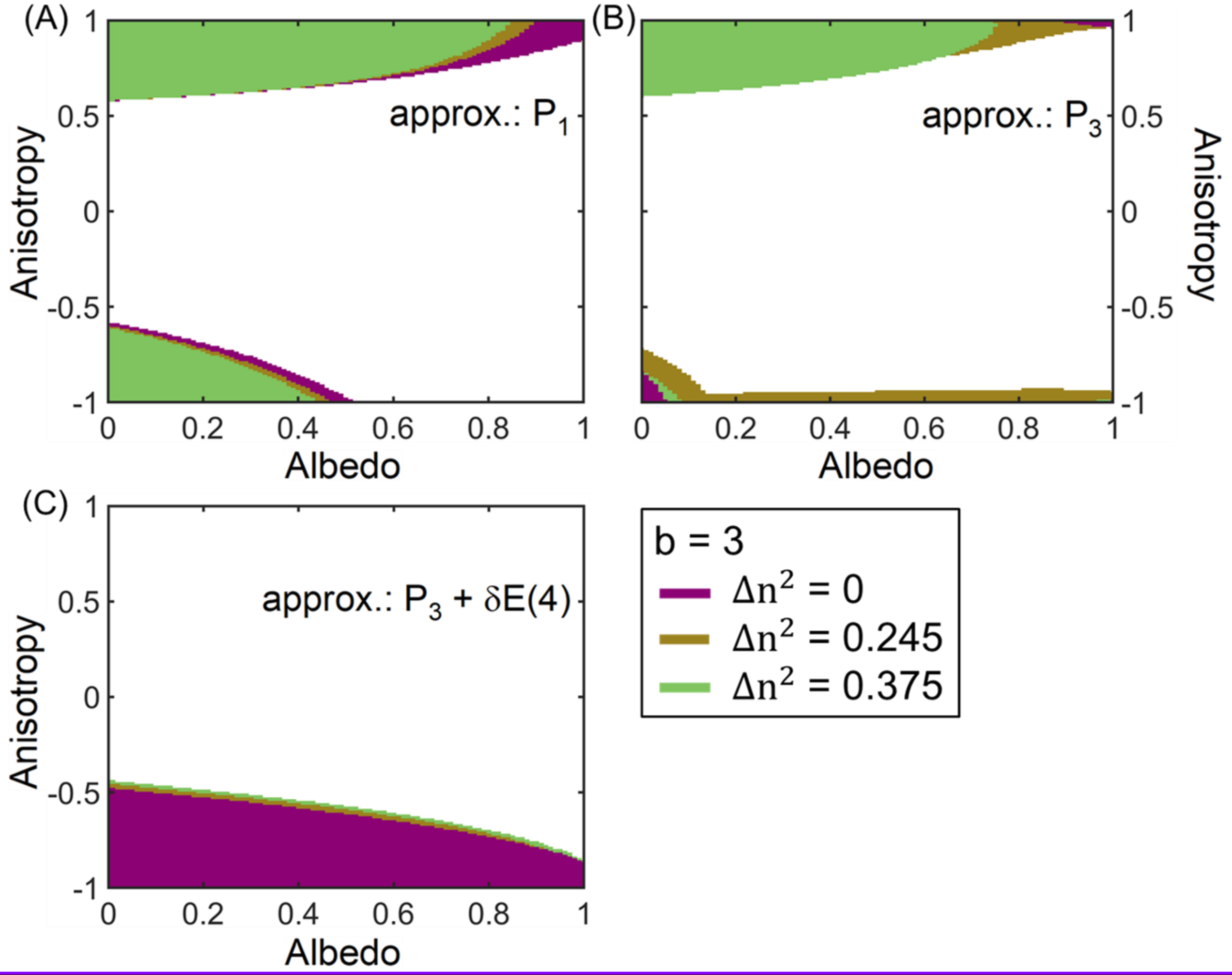
$\mathbf{n_{slab}}$: refractive index of the slab

$\mathbf{n_{outside}}$: refractive index of the surrounding medium

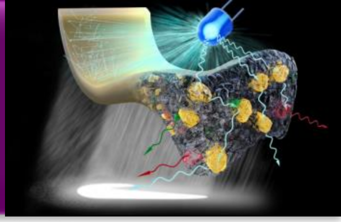
$\mathbf{\Delta n^2}$: Refractive index contrast

$$\Delta n^2 = \frac{(n_{slab}^2 - n_{outside}^2)}{2n_{slab}^2}$$

What about the physical regions?



Outline



1. Transport theory & Radiative Transfer Equation for a slab
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3. $\boxed{\text{Comparisons to Monte Carlo Simulations}}$
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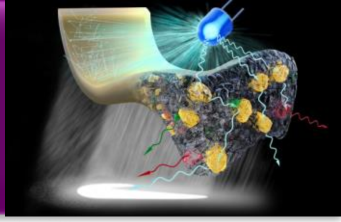
Physical & Unphysical

Practical example



A polymer slab with YAG:Ce^{3+} scatterers* (white LEDs) :

Absorbing case (λ= 460nm) :

a = 0.89 $\Rightarrow$ Absorbing

g = 0.72 $\Rightarrow$ Forward Scattering

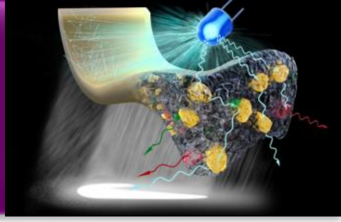
b = 3

Δn^2 = 0.245

$$\Delta \ell_{tr}^{P_1} = 37\%$$
$$\Delta \ell_{tr}^{P_3} = 6.66\%$$
$$\Delta \ell_{tr}^{\delta E(4)} \approx 0\%$$

* Meretska, Uppu, Vissenberg, Lagendijk, IJzerman, and Vos, Opt. Express 25, A906(2017).

Practical example



A polymer slab with YAG:Ce^{3+} scatterers* (white LEDs):

Non-absorbing case (λ=600nm) :

a = 1 ⇒ No Absorption

g = 0.82 ⇒ Forward Scattering

b = 3

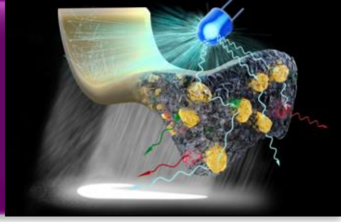
$\Delta n^2 = 0.245$

$$\Delta\ell_{tr}^{P_1} = 350\%$$
$$\Delta\ell_{tr}^{P_3} = 10\%$$
$$\Delta\ell_{tr}^{\delta E(4)} \approx 0\%$$

We can check any configuration

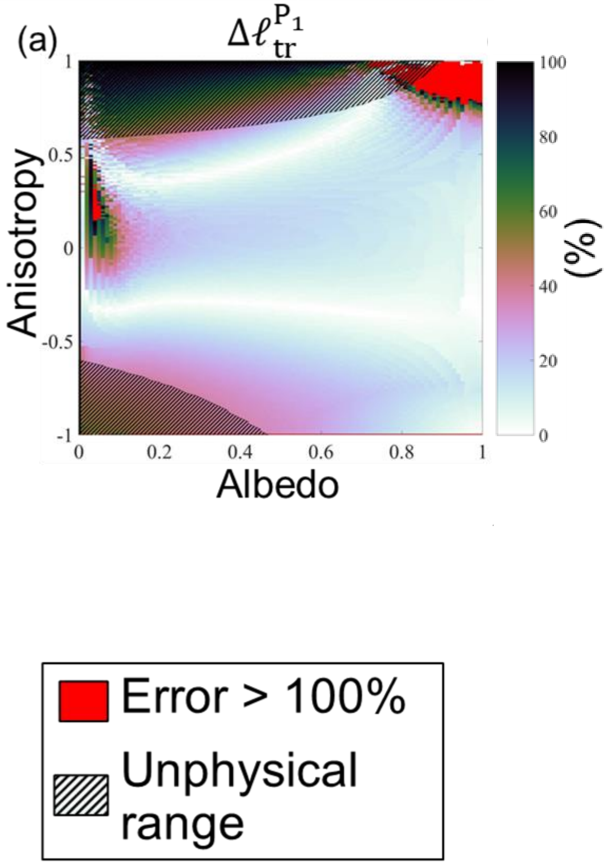
* Meretska, Uppu, Vissenberg, Lagendijk, IJzerman, and Vos, Opt. Express 25, A906(2017).

Relative error maps



Analytic methods compared to Monte Carlo simulations

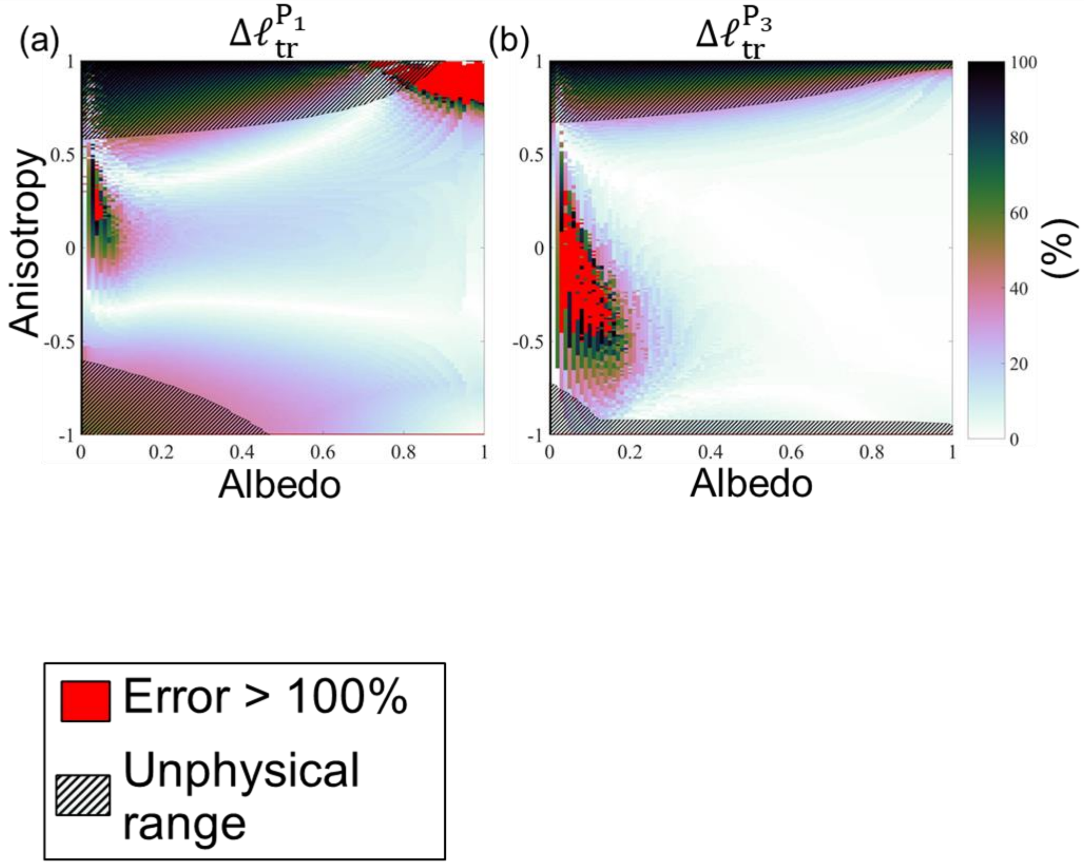
P_1 has large errors in both unphysical and physical!



Relative error maps

P_3 significantly improves the physical region

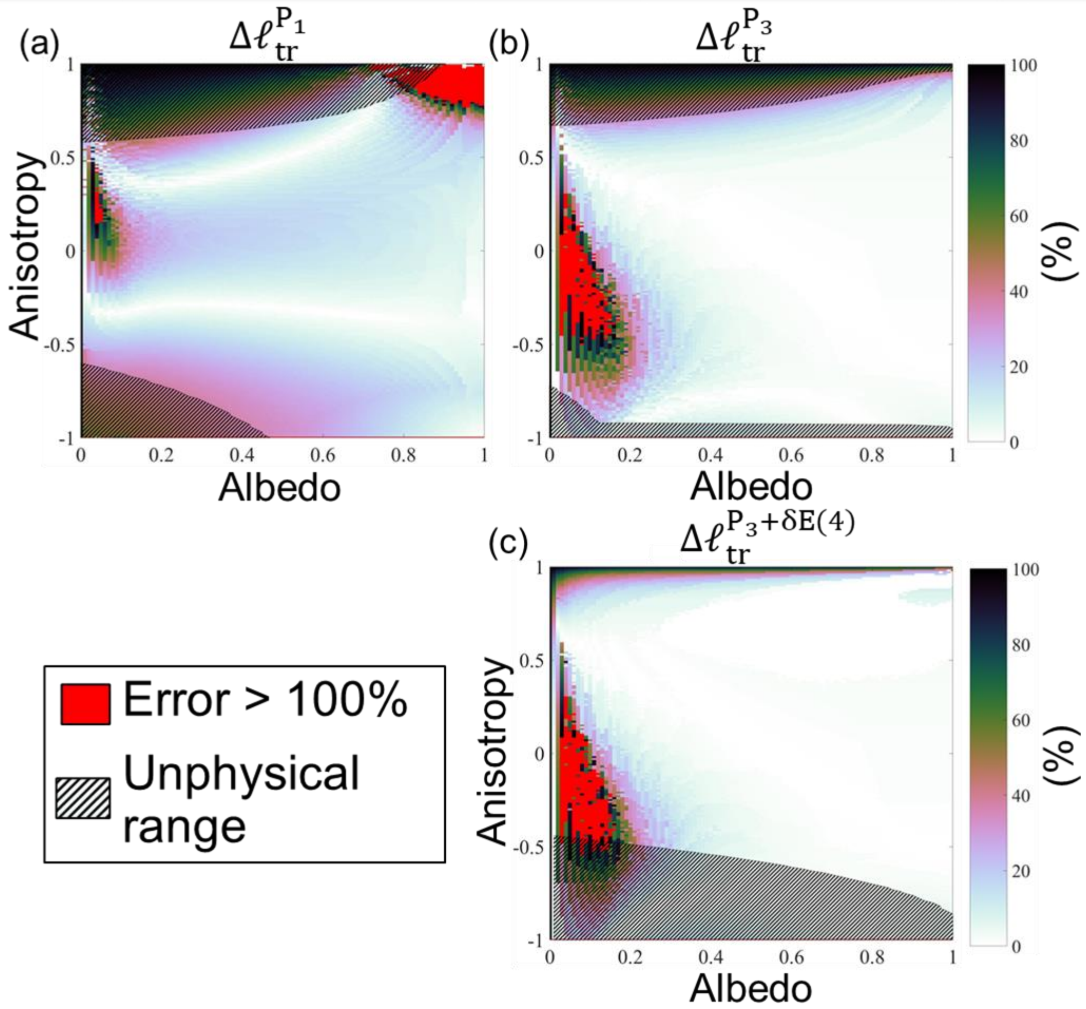
Large errors remain in forward scattering region and strongly absorbing region



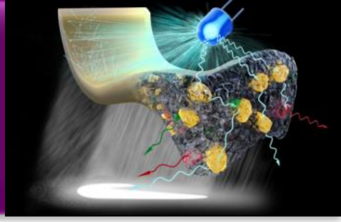
Relative error maps

$P_3 + \delta E(4)$ even more accurate in the physical range!

Large errors remain in forward scattering region and strongly absorbing region



Summary



Transport theory and Radiative Transfer Equation

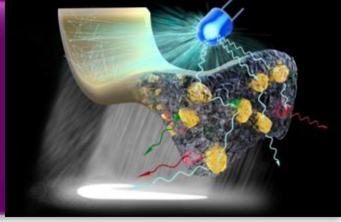
Unphysical ranges of P_1, P_3 and $P_3 + \delta$-E(4) approximations

Comparisons with Monte Carlo Simulations

$P_3 + \delta$-E(4) is better for forward scattering samples

Outlook

Experiments!



Anisotropic volume scattering

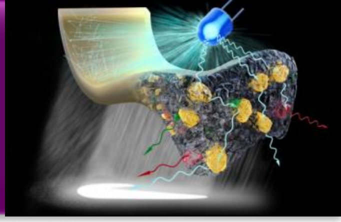
Thomas Kotte, Aurele Adam, Paul Urbach

TU Delft

NWO

Optics Research Group

Research goals

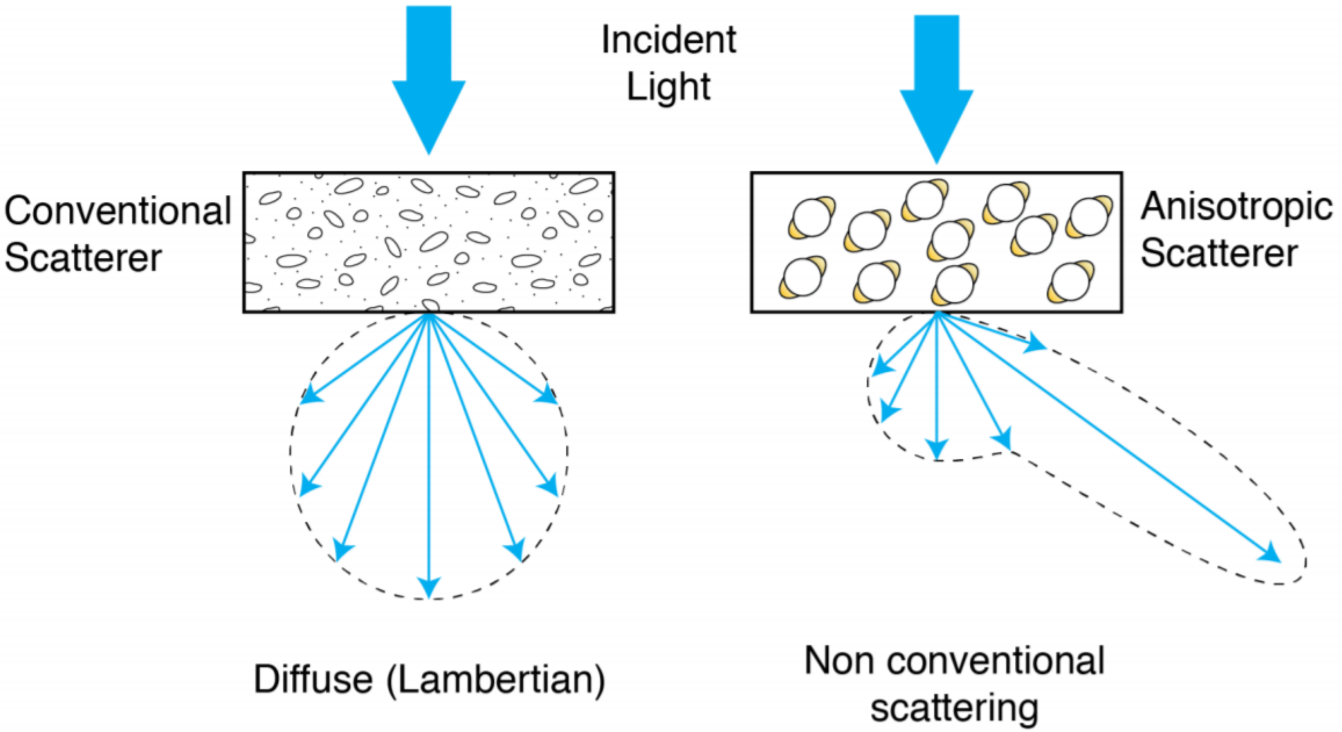


Realize a scattering volume with custom scattering behaviour

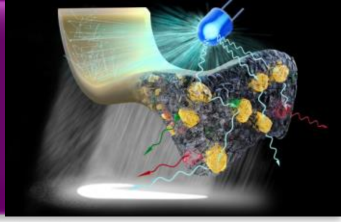
- Limited backscattering
- Scattering to the side

Applications:

- Custom diffusors
- Fluophore based LEDs



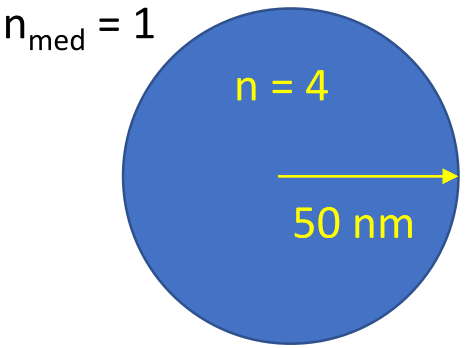
Scattering by homogenous particles

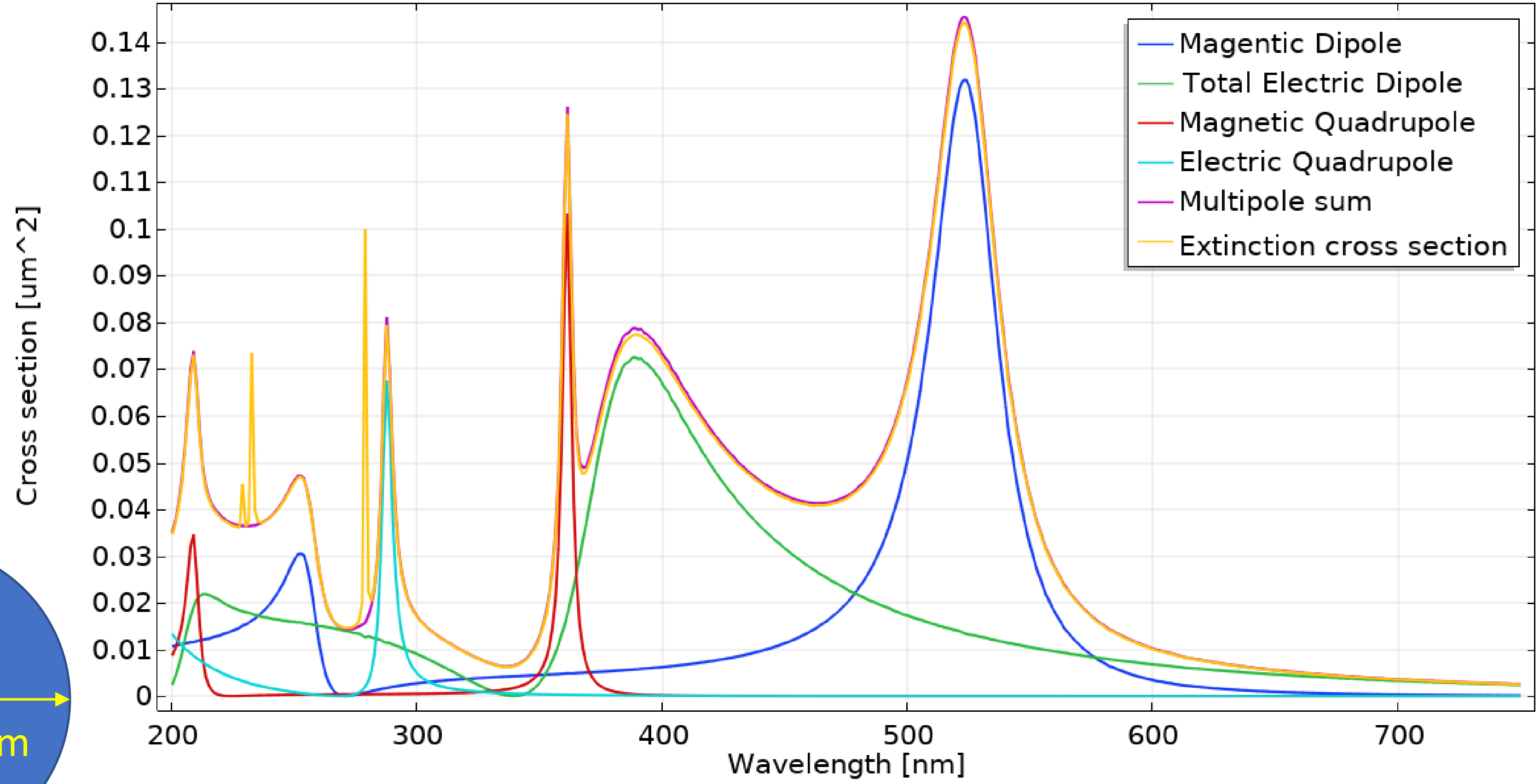


Multipole decomposition

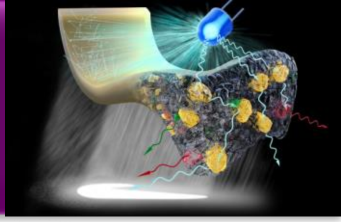
Kerker conditions

Directional scattering in small bandwidth





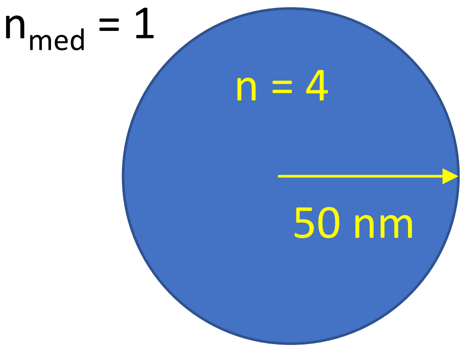
Scattering by homogenous particles

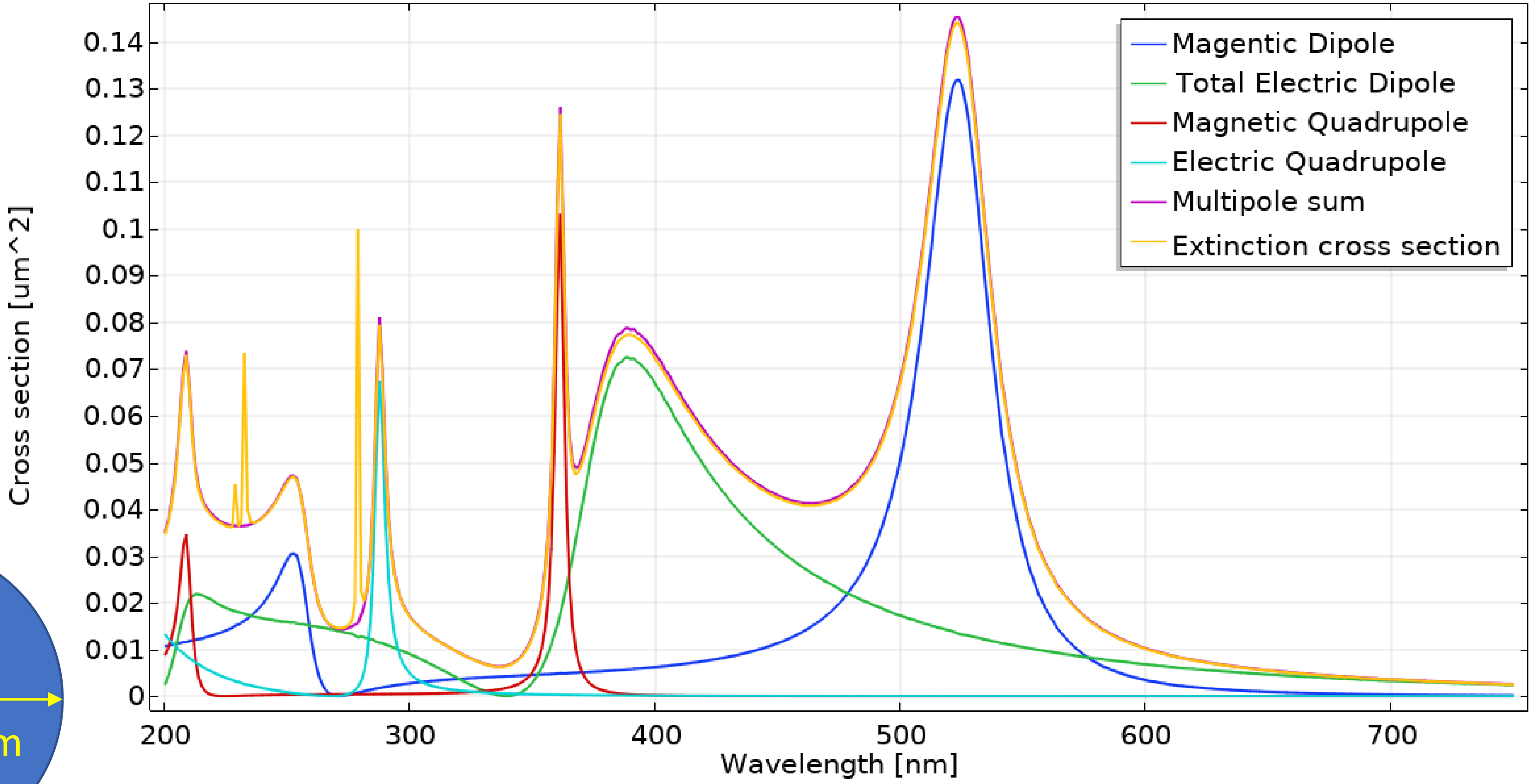


Multipole decomposition

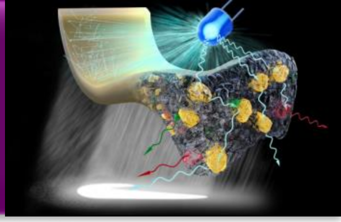
Kerker conditions

Directional scattering in small bandwidth





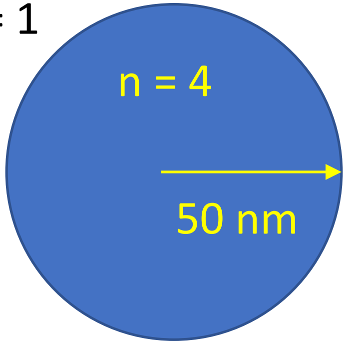
Scattering by homogenous particles

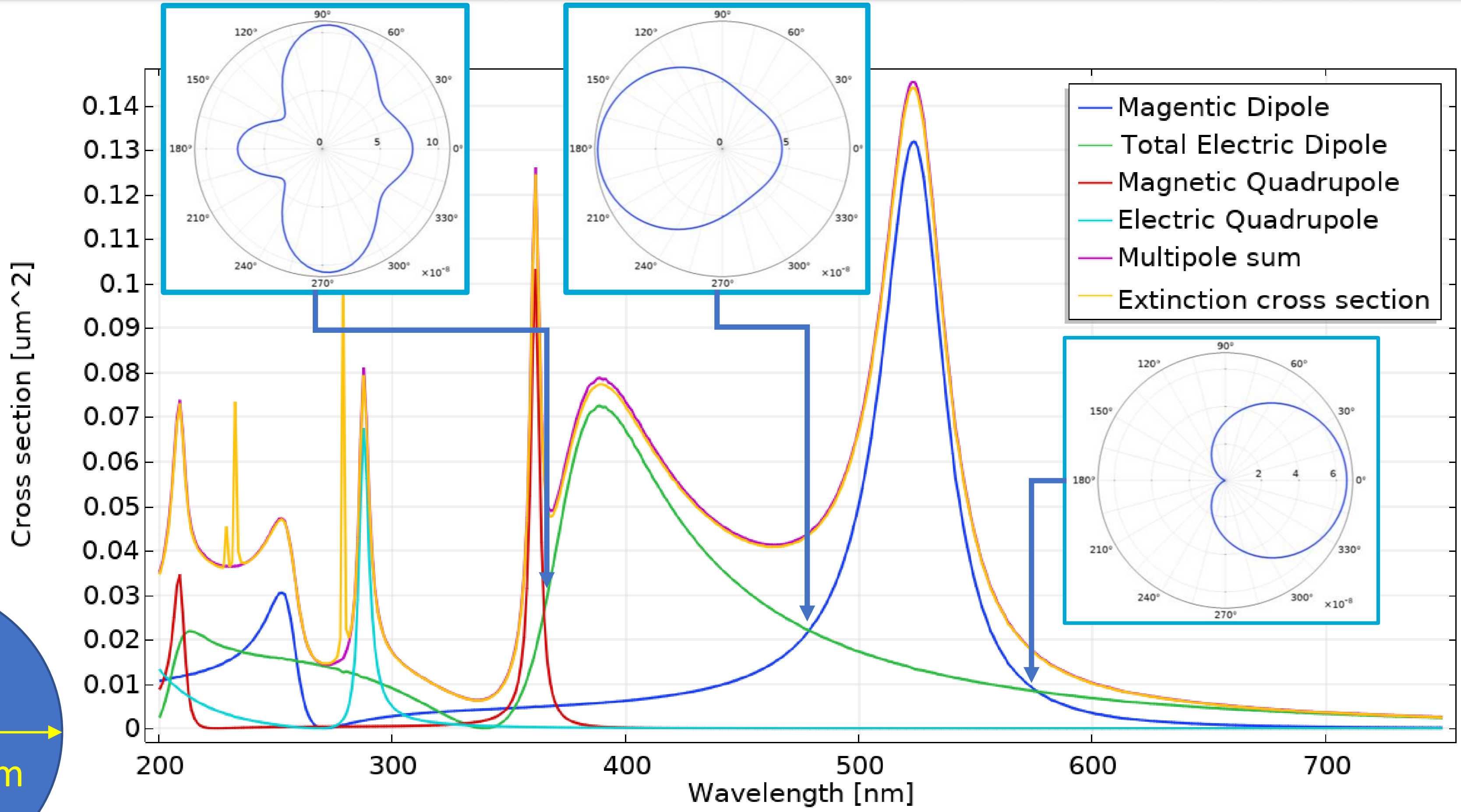


Multipole decomposition

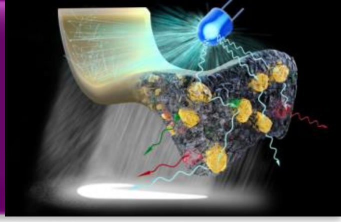
Kerker conditions

Directional scattering in small bandwidth





Anisotropic composite particles

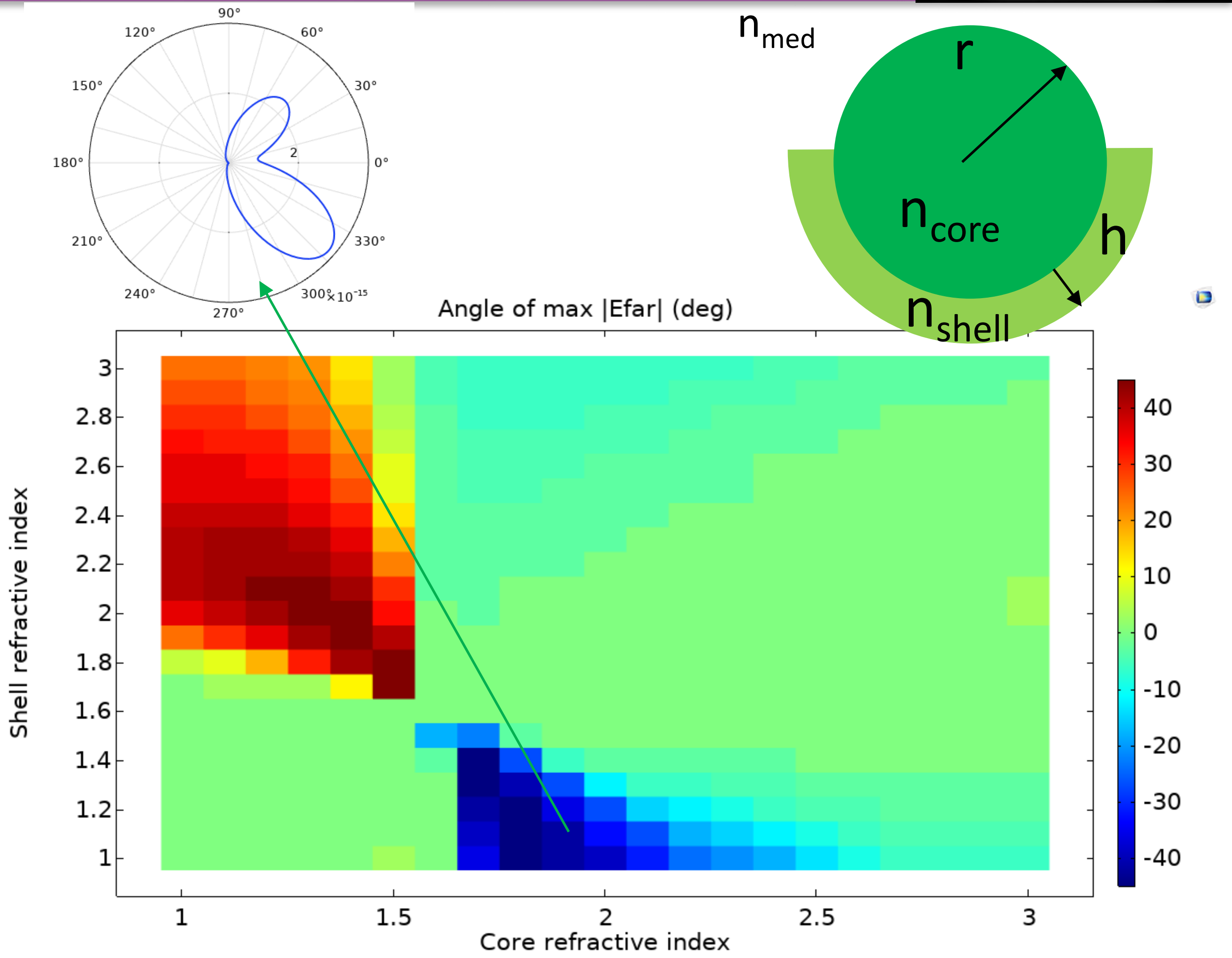


Directionality achieved by phase difference

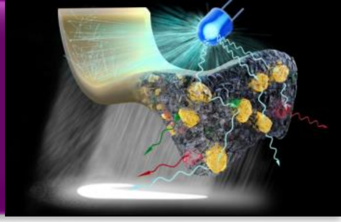
Can be induced by

- $n_{shell} > n_{med} > n_{core}$
- $n_{shell} < n_{med} < n_{core}$

$$\boldsymbol{P} = \varepsilon_0(\varepsilon_{core,shell} - \varepsilon_{med})\boldsymbol{E}$$

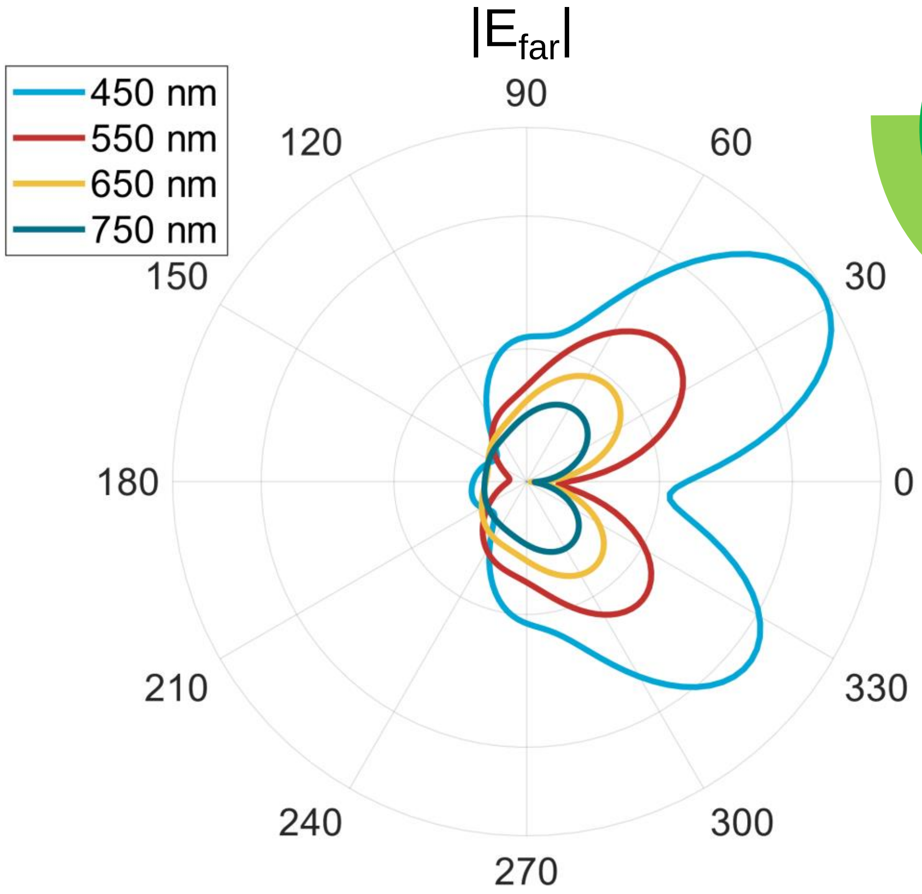


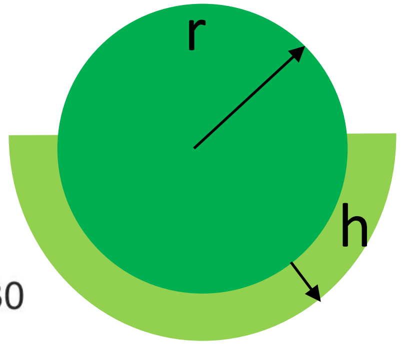
Dielectric design



SiO_2 core TiO_2 semi-shell in PS

r = 150 nm, h = 12 nm





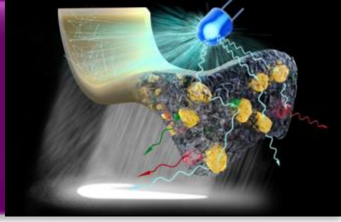
+ No losses

+ Side-scattering over entire visible spectrum

- Scattering decreases fast as function of wavelength

- Low refractive index contrast

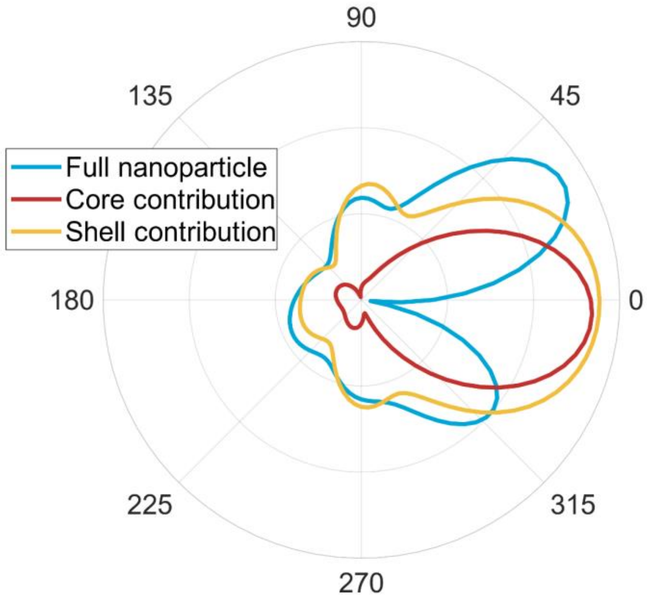
Contributions of core and shell



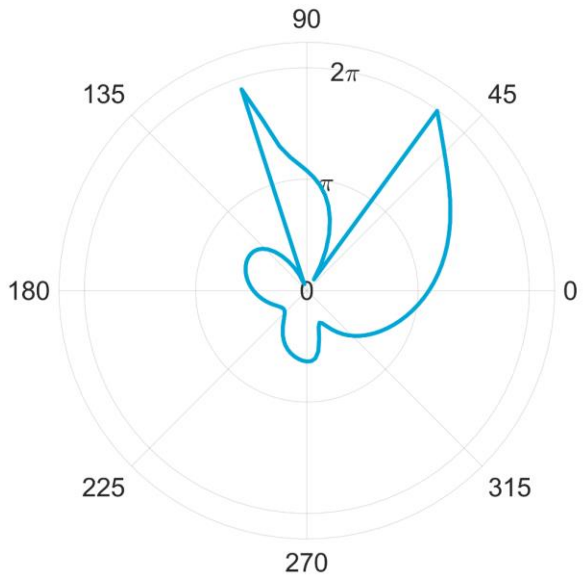
From simulations electric field inside core and shell are calculated

Using this, the contribution to the scattered far field can be determined (Lippmann-Schwinger integral)

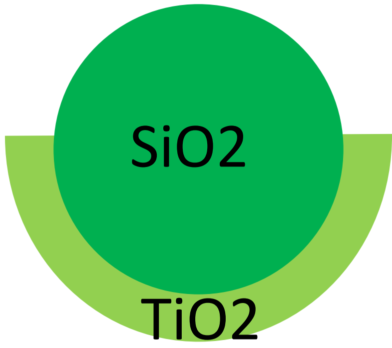
Magnitude



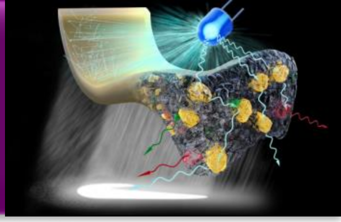
Phase difference



PS



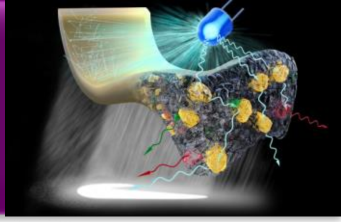
Design steps



1. Design core component: forward scattering, wavelength dependency, …
   (Mie theory and/or Kerker conditions can be used)
2. Add shell component (material dependent on core and medium material)
3. Use trial and error to optimize the design

Fabrication possibilities leading in design

Conclusions and outlook



Dielectric particles with asymmetric scattering can be realized

- Phase difference in materials cause asymmetry

Next steps:

- Additional designs
- Monte-Carlo simulations
- Fabrication of nanoparticles
- Measurements

Extracting scattering properties of materials with Mutual Extinction

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What is extinction of light?

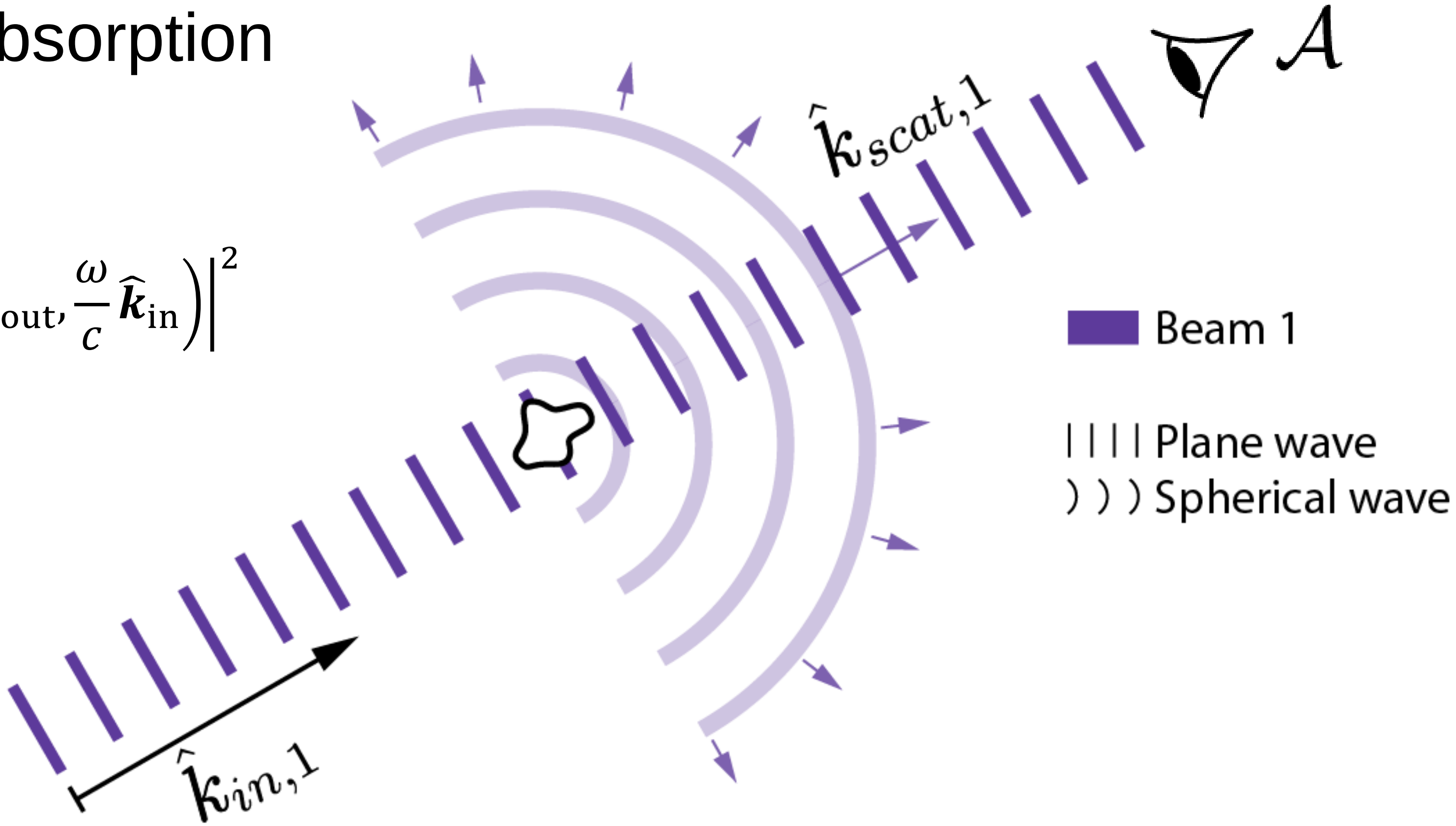
What is extinction of light?

- Extinction = Scattering + Absorption
- Optical theorem:

$$\frac{c}{\omega}\operatorname{Im} f\left(\frac{\omega}{c}\hat{\boldsymbol{k}}_{\text{out}}, \frac{\omega}{c}\hat{\boldsymbol{k}}_{\text{in}}\right) = \frac{1}{4\pi}\int \mathrm{d}\hat{\boldsymbol{k}}_{\text{out}} \left| f\left(\frac{\omega}{c}\hat{\boldsymbol{k}}_{\text{out}}, \frac{\omega}{c}\hat{\boldsymbol{k}}_{\text{in}}\right)\right|^2$$

- If $k_{\text{in}} = k_{\text{out}}$,

$$4\pi\frac{c}{\omega}\operatorname{Im} f(0) = \sigma_{\text{tot}}$$

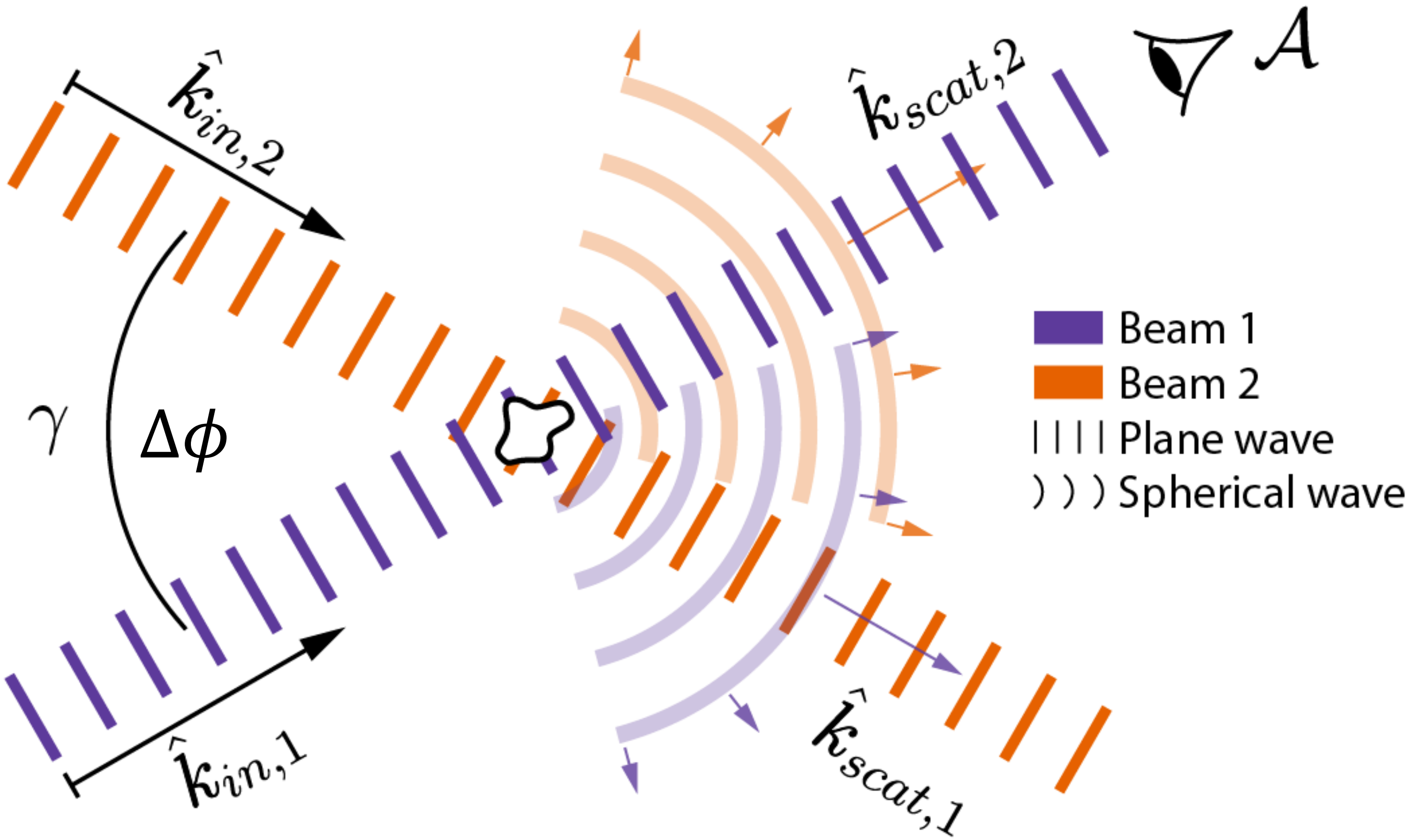


And Mutual Extinction?

Mutual Extinction:

Cross interference between the beams

We can control the total extinction (F^{TE})



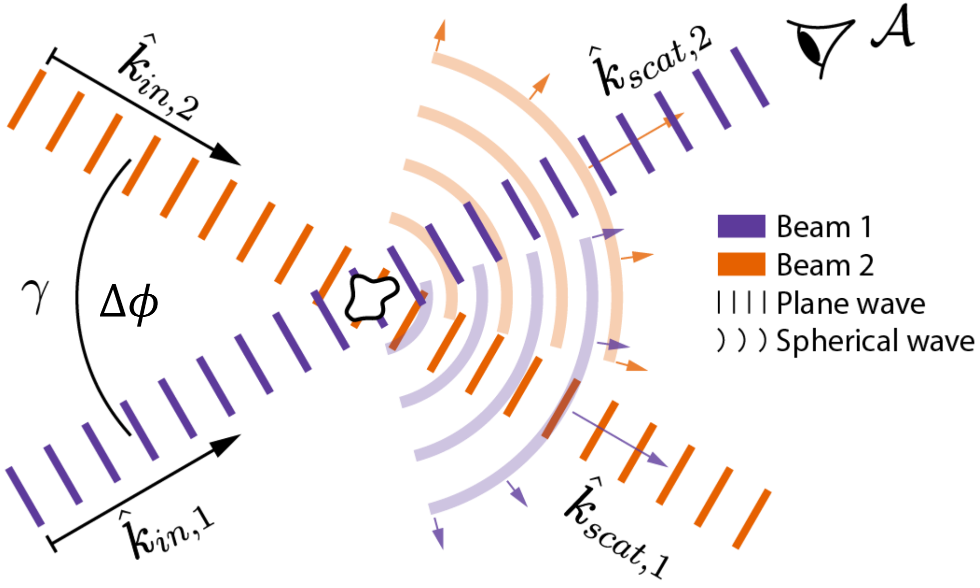
A. Lagendijk, A. P. Mosk, and W. L. Vos, *EPL* **130**, 34002 (2020)

Extinction and scattering amplitude

- Extinction $\propto \mathrm{Im}\{f\}$
- Intensity $\propto |f|^2$

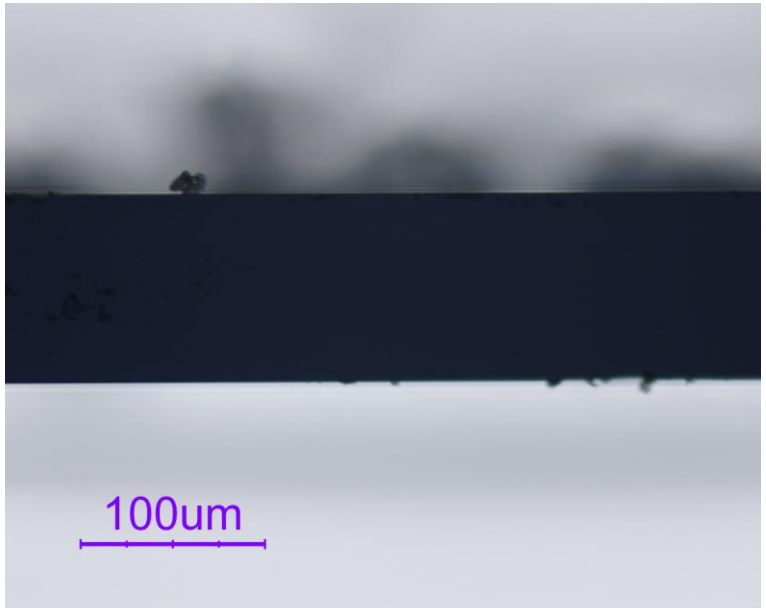
For each angle γ, how much light is extinguished?

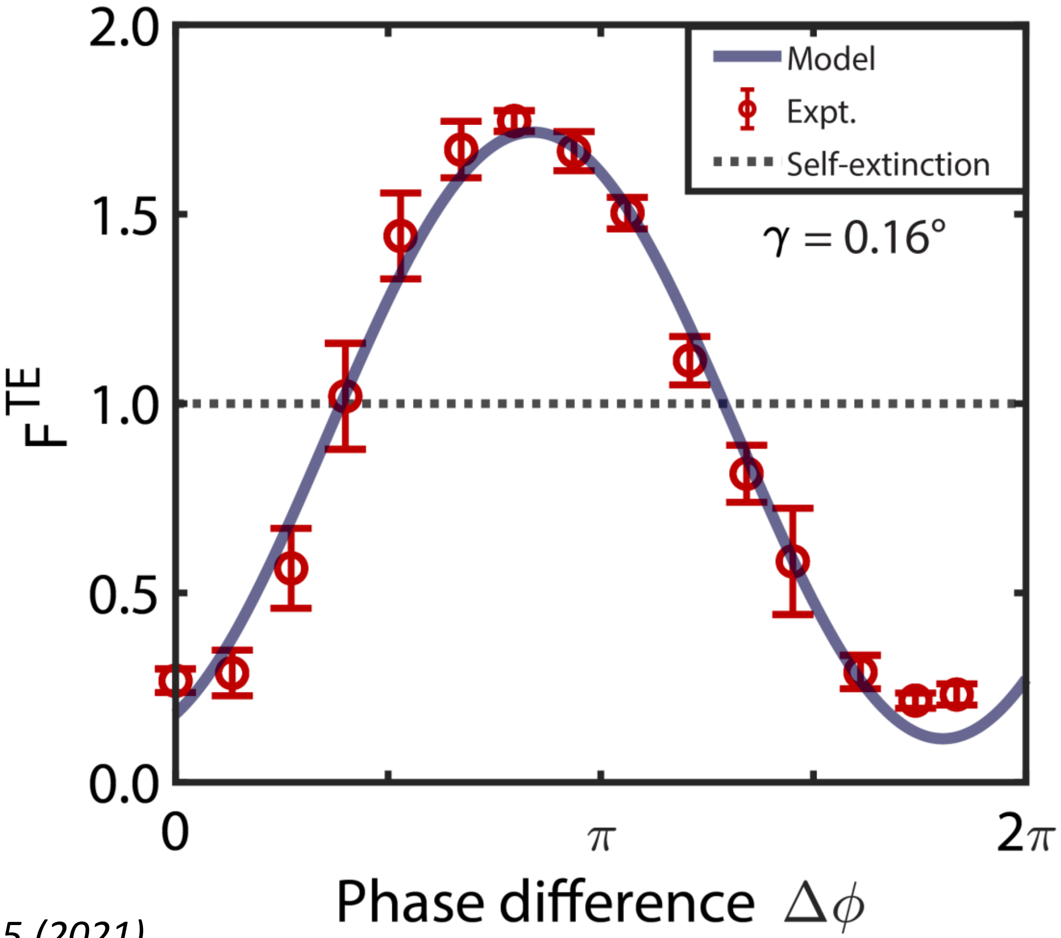
➢ Amplitude of $\mathrm{Im}\{f\}$



Experimental results - phase

- Sample: Silicon bar
- Phase dependency of total extinction

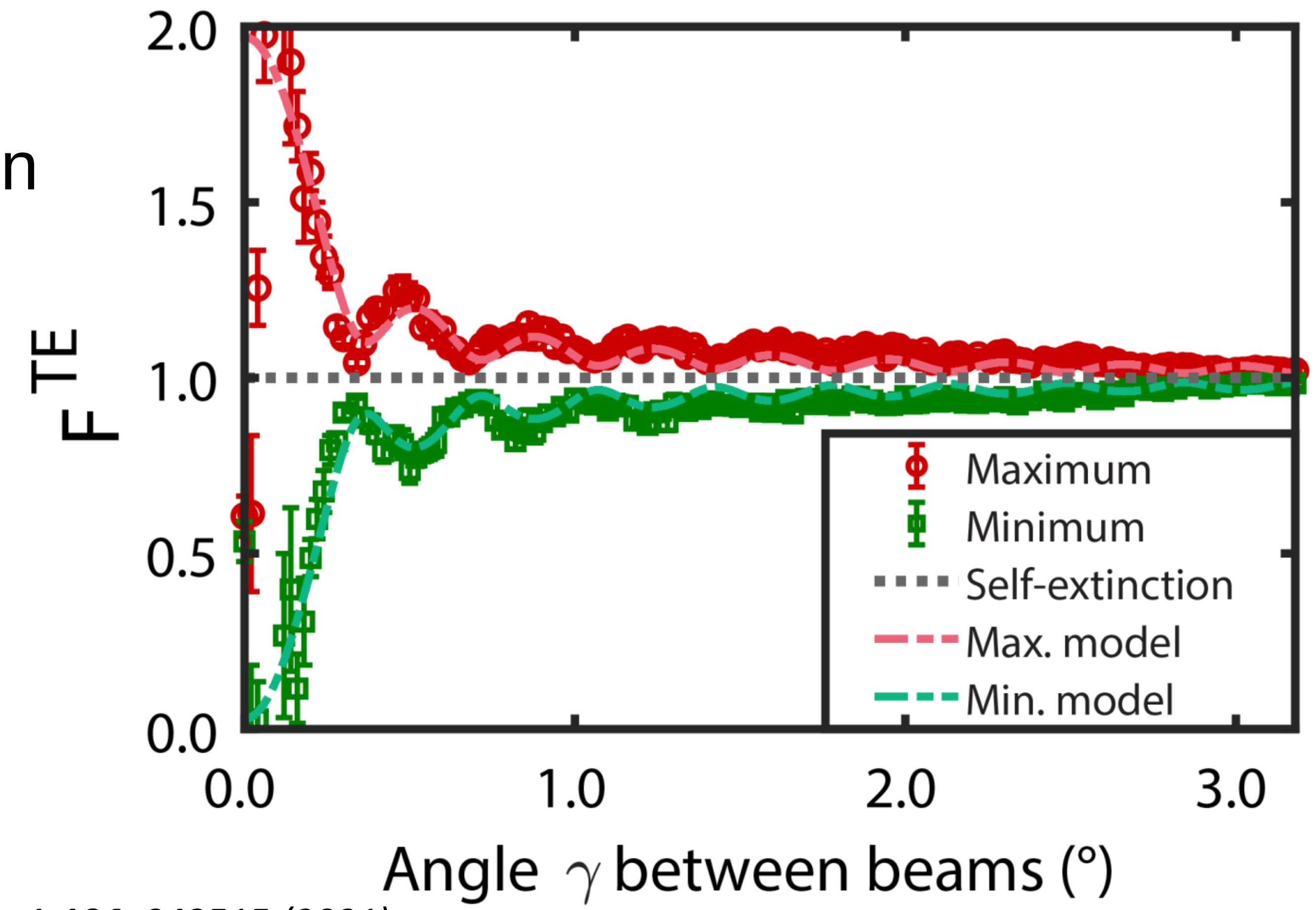




A. Rates, A. Lagendijk, O. Akdemir, A. P. Mosk, and W. L. Vos, *Phys. Rev. A* ***104****, 043515 (2021)*

Experimental results - amplitude

- Sample: Silicon bar
- Angle dependency of total extinction
- Analytical model

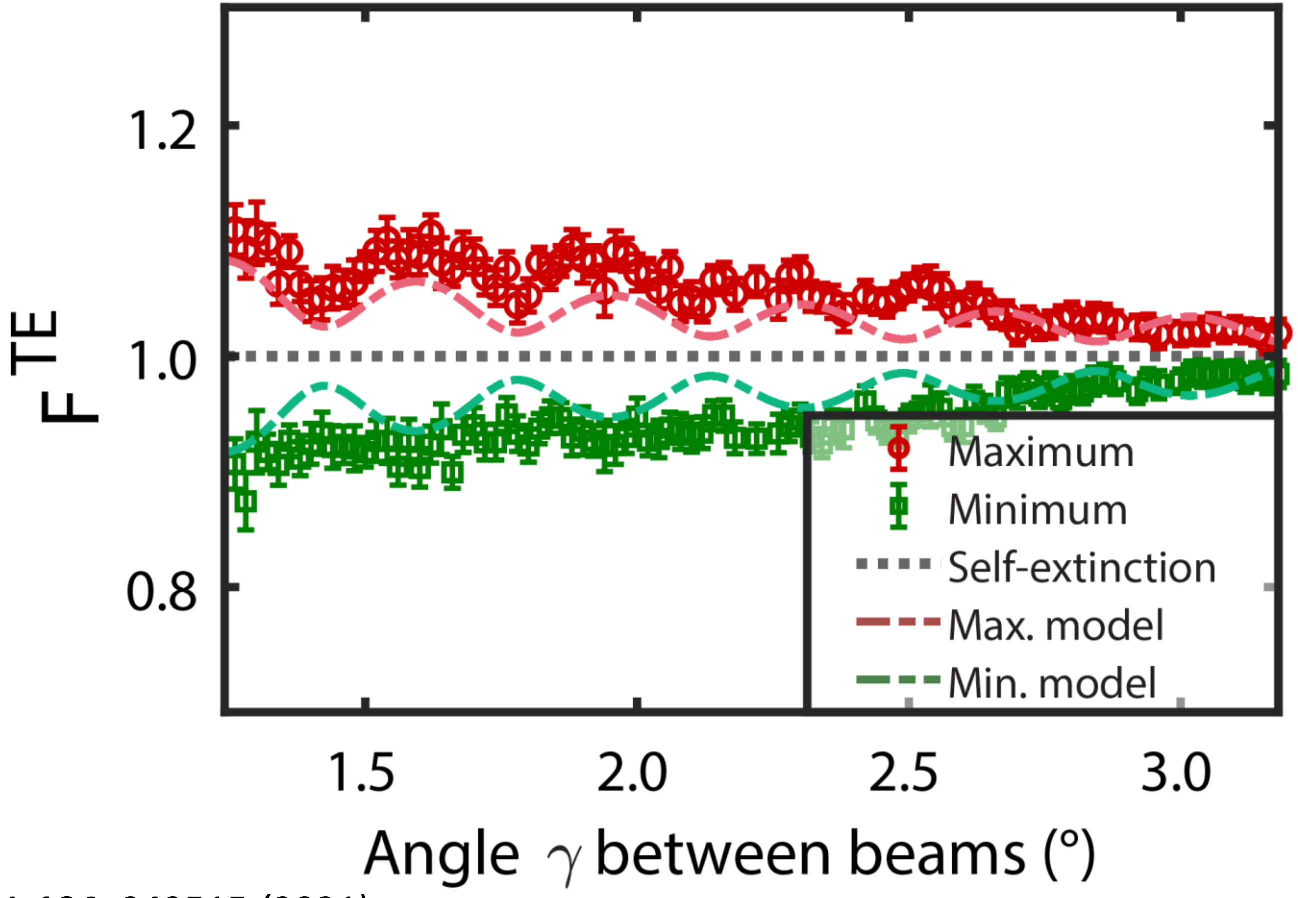


A. Rates, A. Lagendijk, O. Akdemir, A. P. Mosk, and W. L. Vos, *Phys. Rev. A* ***104**, 043515 (2021)*

Experimental results - amplitude

- Random variations
- Model fails to describe data

➢ *Angular speckle*

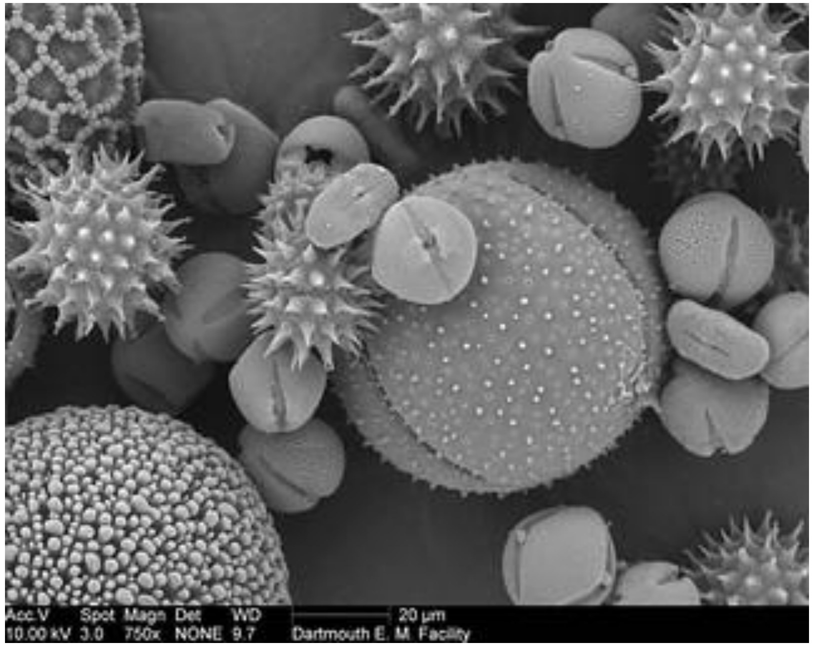


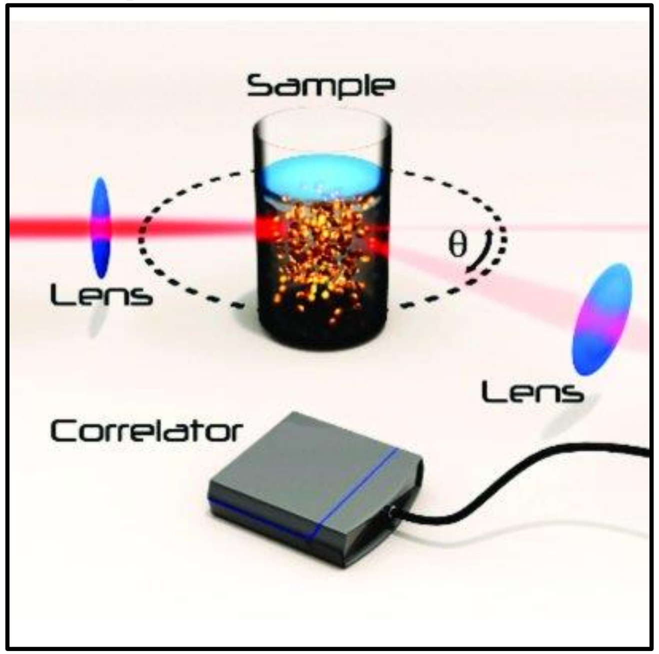
A. Rates, A. Lagendijk, O. Akdemir, A. P. Mosk, and W. L. Vos, *Phys. Rev. A* ***104****, 043515 (2021)*

Possible applications

1. Extract scattering properties of object
2. Measure shape with high precision
3. Dynamic Light Scattering with absorbing samples

Darthmout College





LS Instruments

Conclusions

- Large control over total extinction
- Mutual Extinction gives us information that scattering intensity does not
- Angular speckle regime for large angles

Next steps

- Phase information of the scattering amplitude