SHINING FROM ALL SIDES: ORIENTATION-AVERAGED OPTICAL PROPERTIES OF NANOPARTICLE ASSEMBLIES

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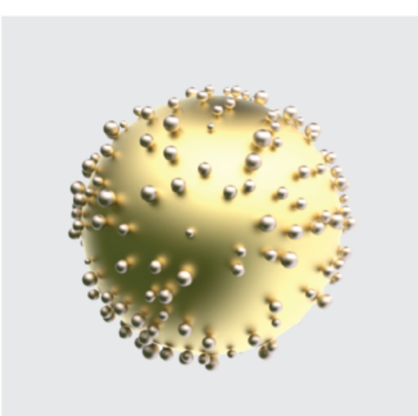
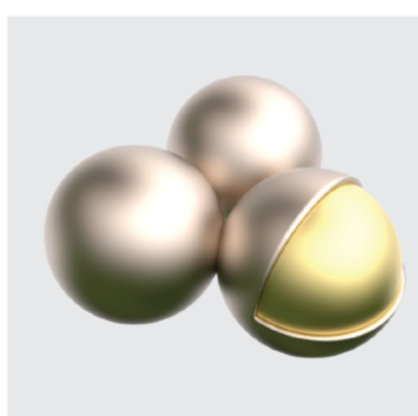
Victoria University of Wellington, New Zealand

NANO-OPTICS OF NANOPARTICLE ASSEMBLIES

NANO-OPTICS.AC.NZ/TERMS

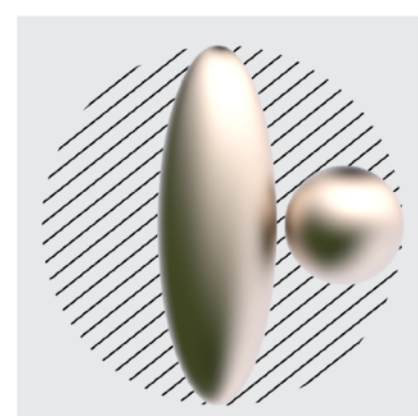


CORE-SATELLITES



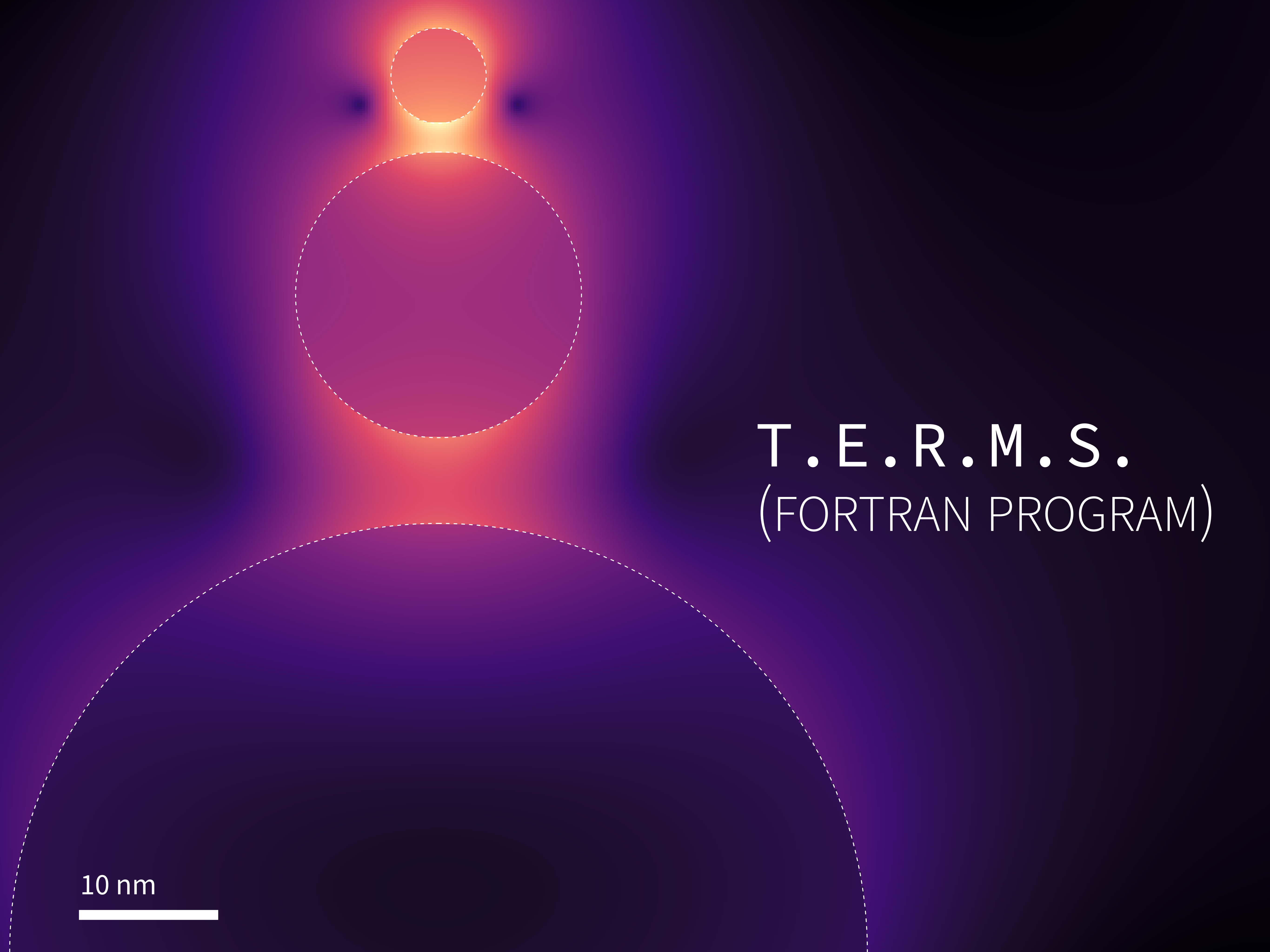
Au@Pd TRIMER

Au NANOROD HELIX

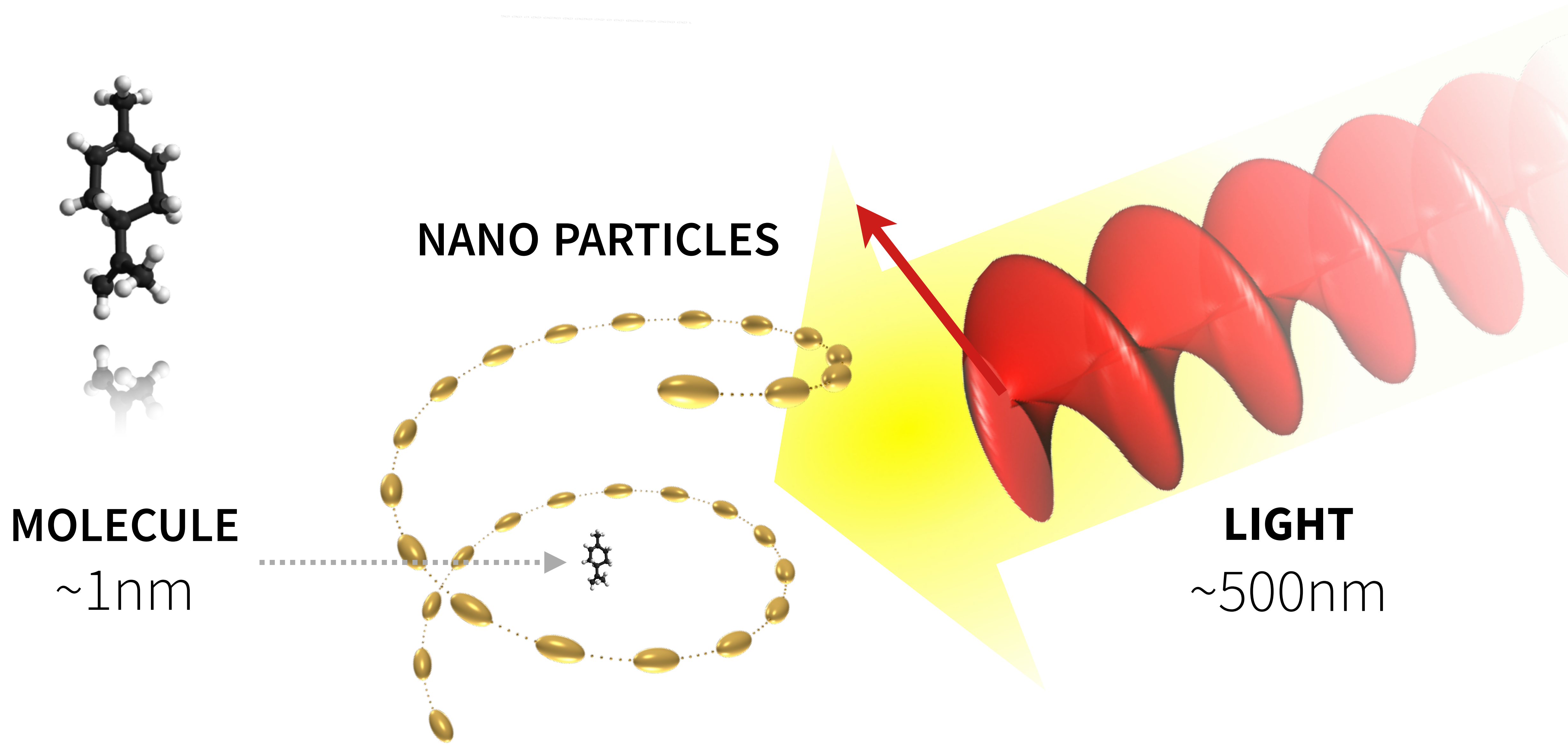


RAYLEIGH HYPOTHESIS

Au NANOSTAR

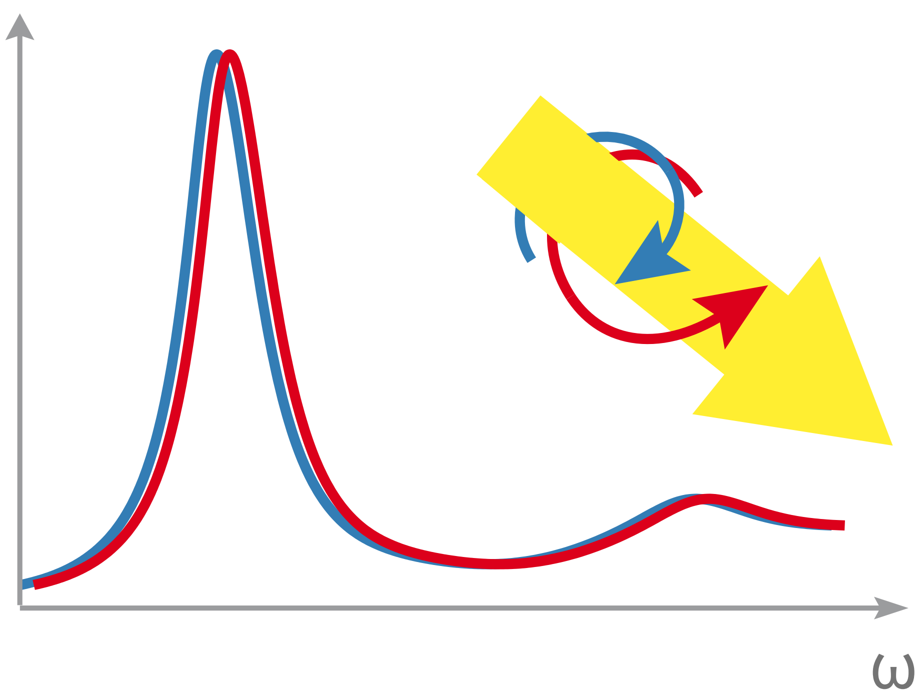

T.E.R.M.S.
(FORTRAN PROGRAM)
10 nm

EXPLORING CHIRALITY AT THE NANOSCALE

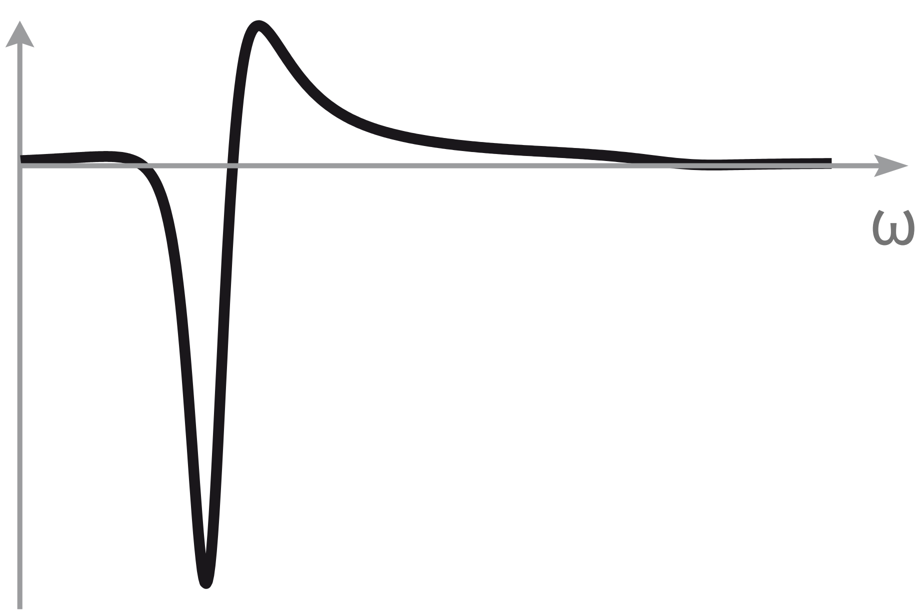


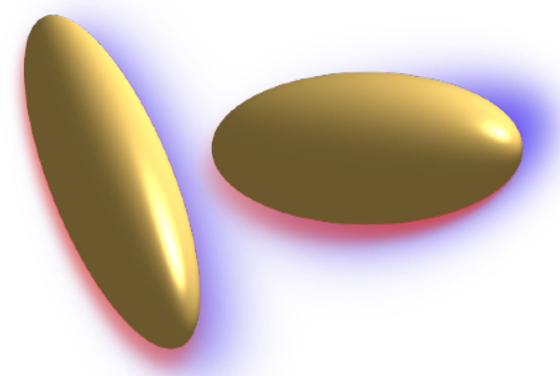
GIANT CIRCULAR DICHROISM WITH PLASMONIC ANTENNAS

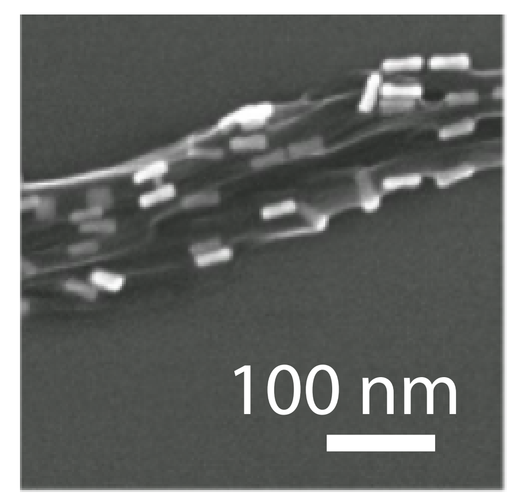
ABSORBANCE



CIRCULAR DICHROISM

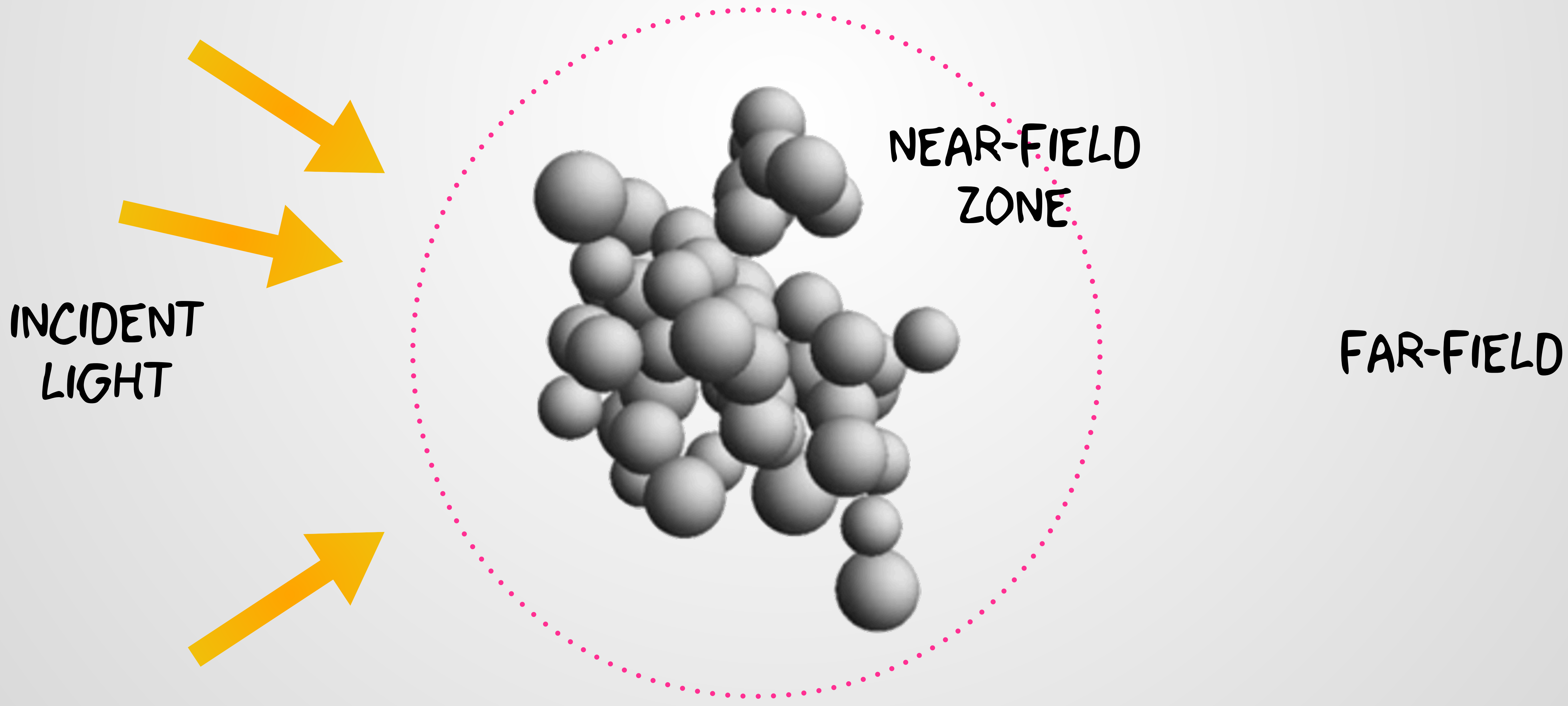




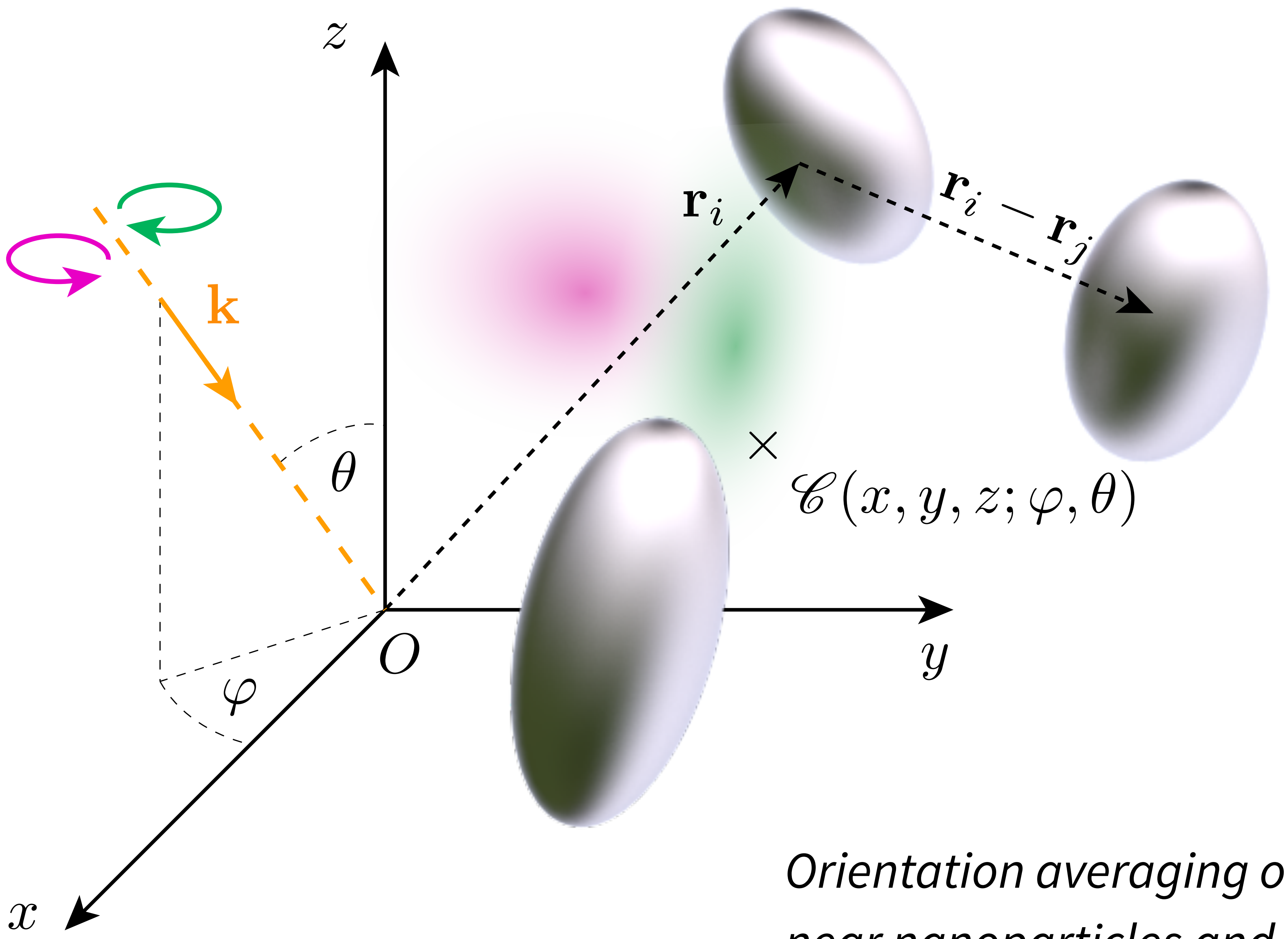


LIGHT SCATTERING BY COLLECTIONS OF PARTICLES

¿HOW MANY DIRECTIONS OF INCIDENCE?



LOCAL DEGREE OF OPTICAL CHIRALITY $\mathscr{C} \propto \Im(\mathbf{E}^* \cdot \mathbf{B})$



Orientation averaging of optical chirality near nanoparticles and aggregates
Phys. Rev. B 103, 115405 (2021)

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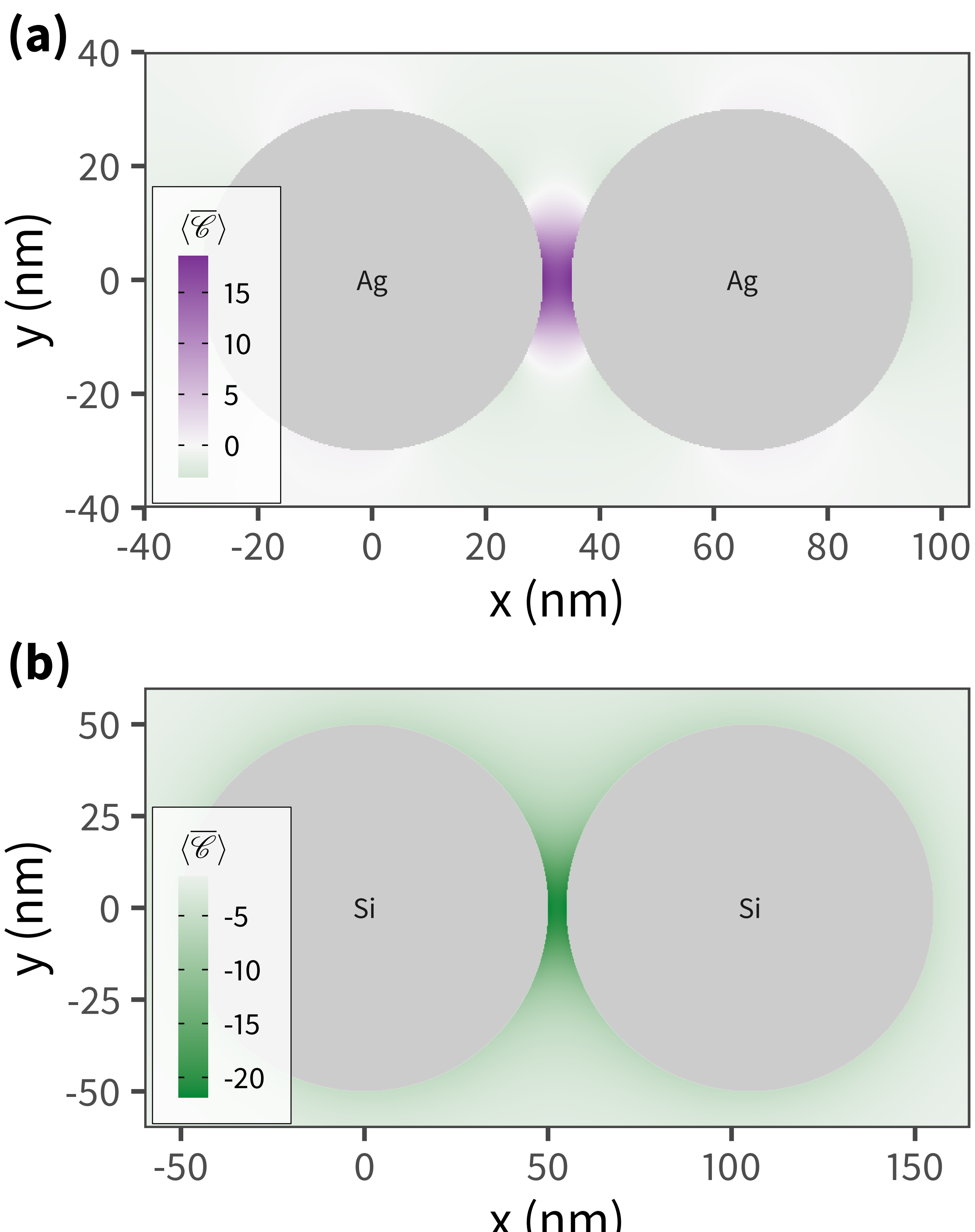

- NON-TRIVIAL SPATIAL DEPENDENCE
- INTERFERENCE BETWEEN E & B
- ANGLE-AVERAGING IMPORTANT

IN RELATION TO EXPERIMENTS

Orientation averaging of optical chirality near nanoparticles and aggregates
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LOCAL DEGREE OF OPTICAL CHIRALITY $\mathscr{C} \propto \Im(\mathbf{E}^* \cdot \mathbf{B})$

$$\langle \mathscr{C} \rangle = 2\pi k \varepsilon_0 E_0^2 \, \Re \left(A_0 + B_0 + C_0 + D_0 \right)$$

with,

For R polarisation:

$$A_0^{(\mathrm{R})} = -1/4\pi$$

$$B_0^{(\mathrm{R})} = \mathrm{Tr} \left(\sum_{j=1}^{N} \sum_{l=1}^{N} \widetilde{\mathbf{Z}}_R^\dagger(k\mathbf{r}_l) \left[\mathbf{Z}_L(k\mathbf{r}_j) T_{LR}^{(j,l)} - \mathbf{Z}_R(k\mathbf{r}_j) T_{RR}^{(j,l)} \right] \right)$$

$$C_0^{(\mathrm{R})} = \mathrm{Tr} \left(\sum_{j=1}^{N} \sum_{l=1}^{N} \left[-T_{LR}^{\dagger(j,l)} \mathbf{Z}_L^\dagger(k\mathbf{r}_j) - T_{RR}^{\dagger(j,l)} \mathbf{Z}_R^\dagger(k\mathbf{r}_j) \right] \widetilde{\mathbf{Z}}_R(k\mathbf{r}_l) \right)$$

$$D_0^{(\mathrm{R})} = \mathrm{Tr} \left(\sum_{j=1}^{N} \sum_{l=1}^{N} \sum_{i=1}^{N} \sum_{k=1}^{N} J_{RR}^{(k,l)} \left(T_{LR}^{\dagger(j,l)} \mathbf{Z}_L^\dagger(k\mathbf{r}_j) + T_{RR}^{\dagger(j,l)} \mathbf{Z}_R^\dagger(k\mathbf{r}_j) \right) \left(\mathbf{Z}_L(k\mathbf{r}_i) T_{LR}^{(i,k)} - \mathbf{Z}_R(k\mathbf{r}_i) T_{RR}^{(i,k)} \right) \right).$$

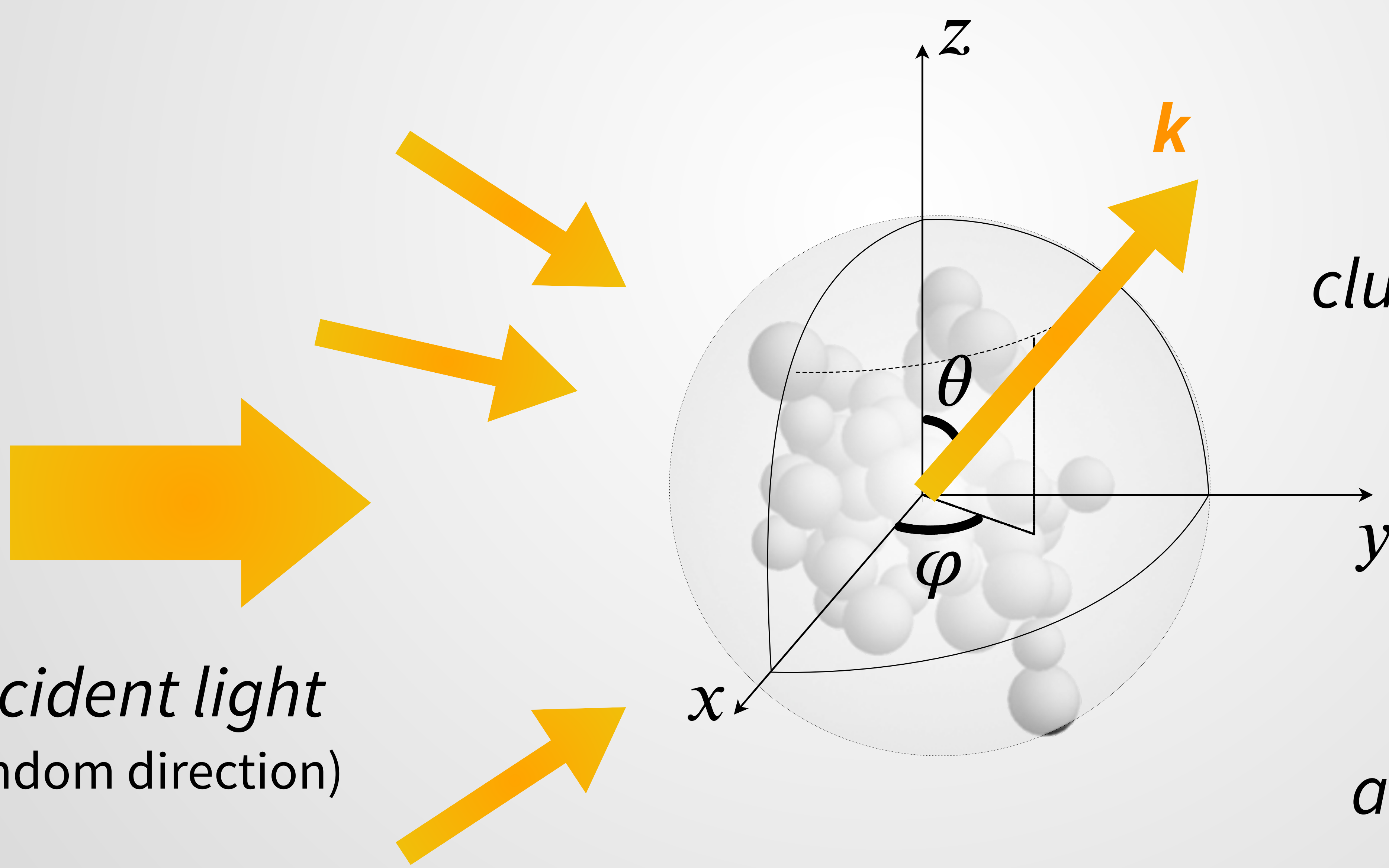
MOTIVATION

- NANO-OPTICS, MANY SAMPLES IN SOLUTION
- SIMULATIONS: FEM, FDTD, DDA, ETC.
- OFTEN ASSUMED 3 DIRECTIONS OF INCIDENCE ENOUGH
- T-MATRIX: ANALYTICAL ORIENTATION-AVERAGING

  – BUT IS IT ALWAYS BETTER?
- FAR-FIELD VS NEAR-FIELD
- AVERAGE VS CIRCULAR DICHROISM
- RULE OF THUMB?

ORIENTATION AVERAGING

$$\langle f \rangle = \frac{1}{4\pi} \int_0^{2\pi} \int_0^{\pi} f(\varphi, \theta) \sin\theta \, \mathrm{d}\theta \mathrm{d}\varphi$$

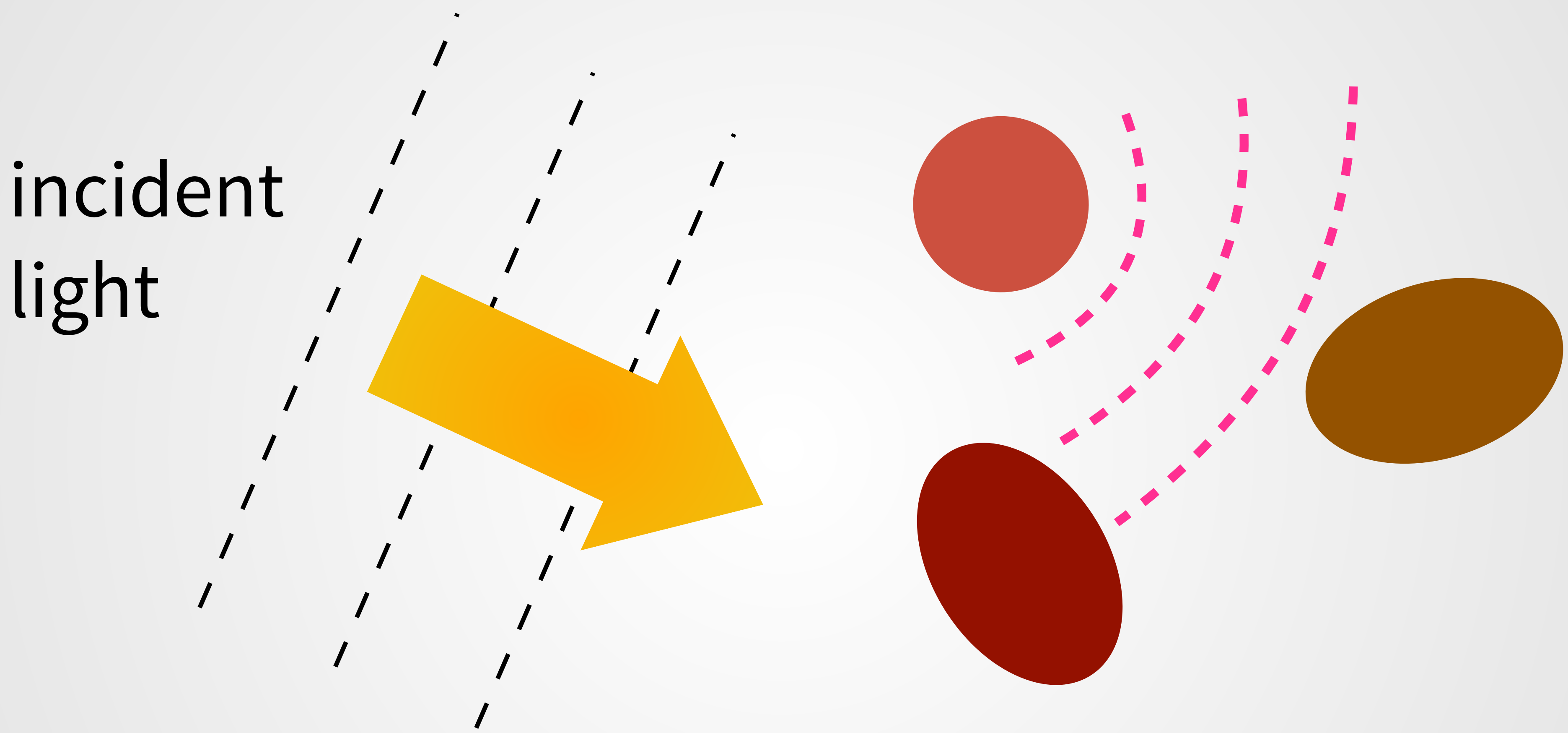


Incident light
(random direction)

Scatterer, cluster of particles

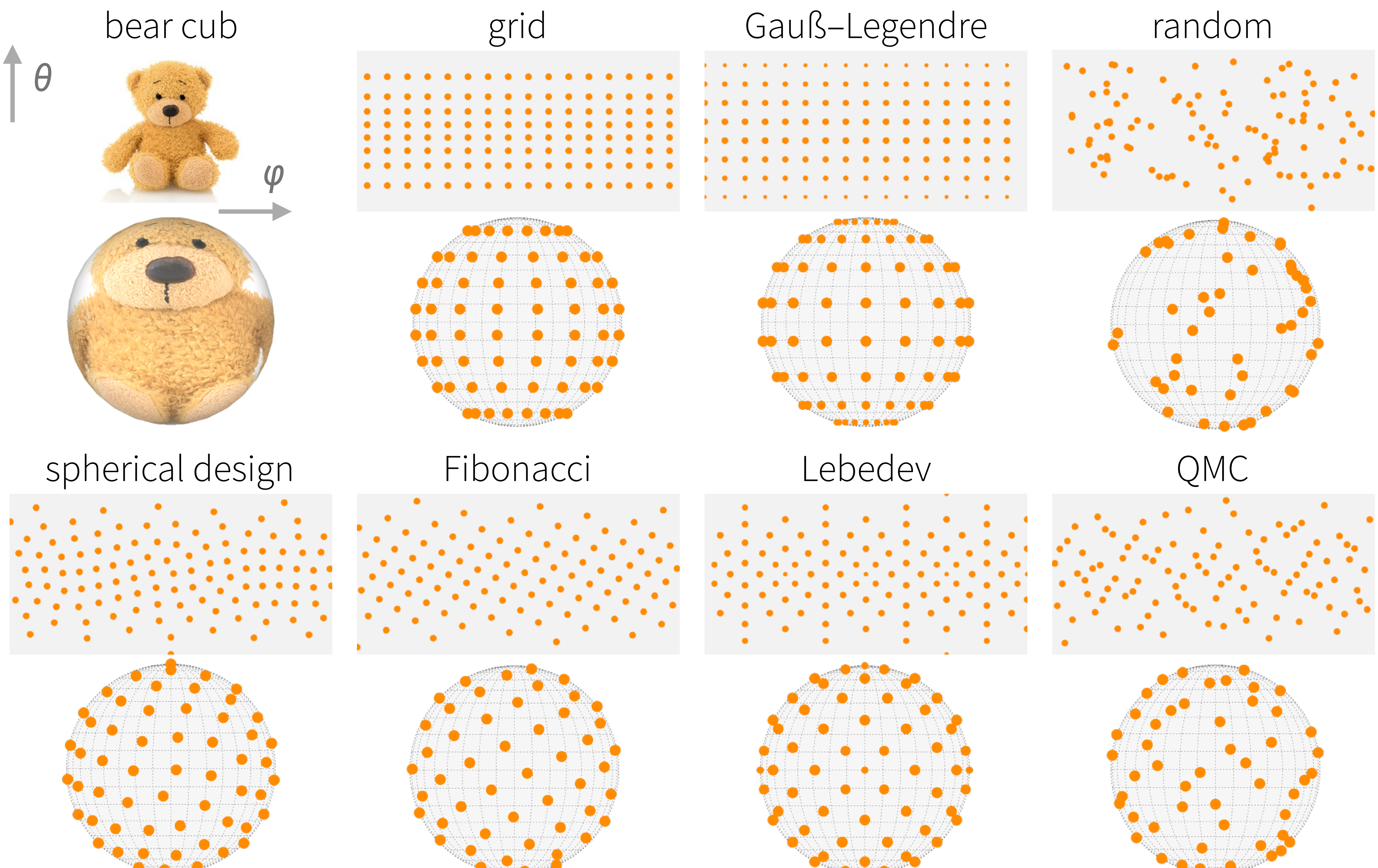
Scattering, absorption, etc.

SUPERPOSITION T-MATRIX METHOD

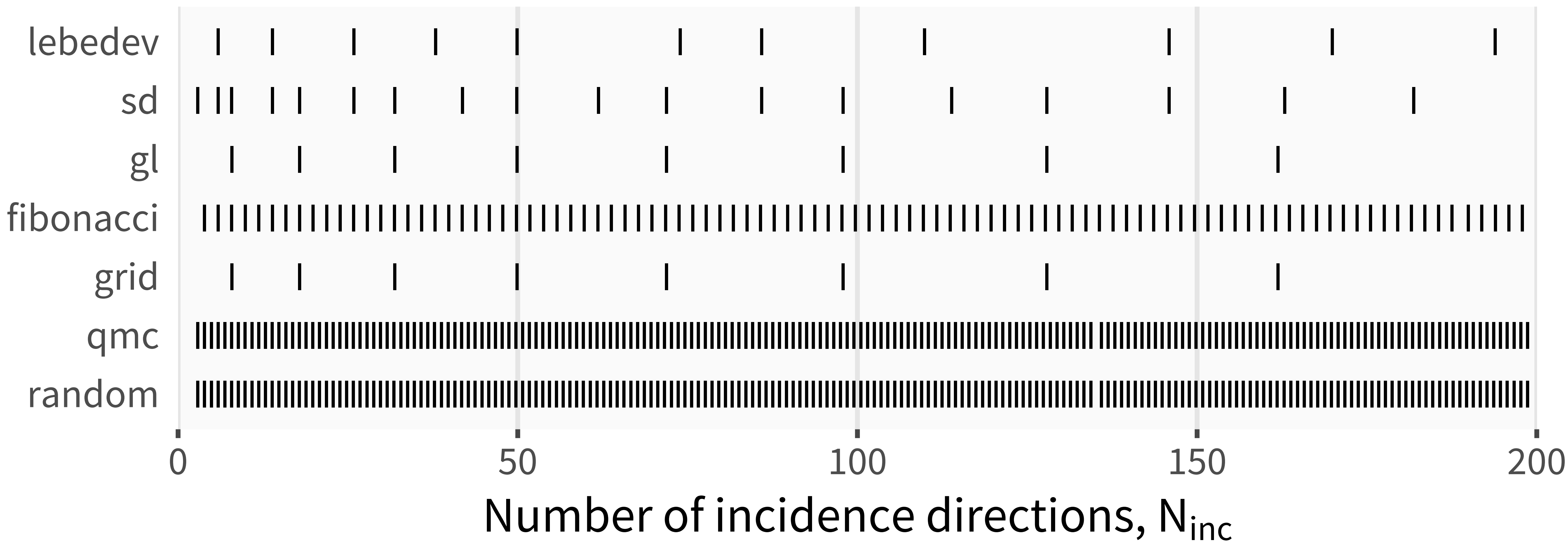


- EXPAND FIELDS IN **SPHERICAL WAVES** (MULTIPOLES)
- EXCITING FIELD = INCIDENT + SCATTERED
- LINEAR SYSTEM FOR N PARTICLES

SPHERICAL CUBATURE



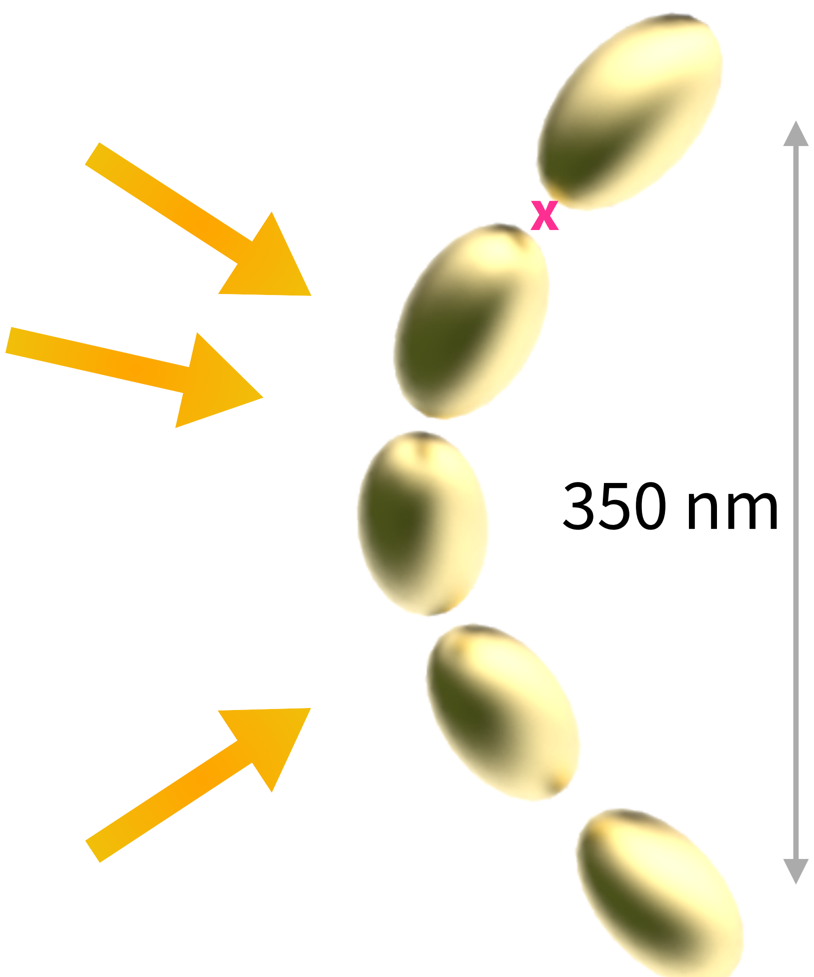
SPHERICAL CUBATURE METHODS

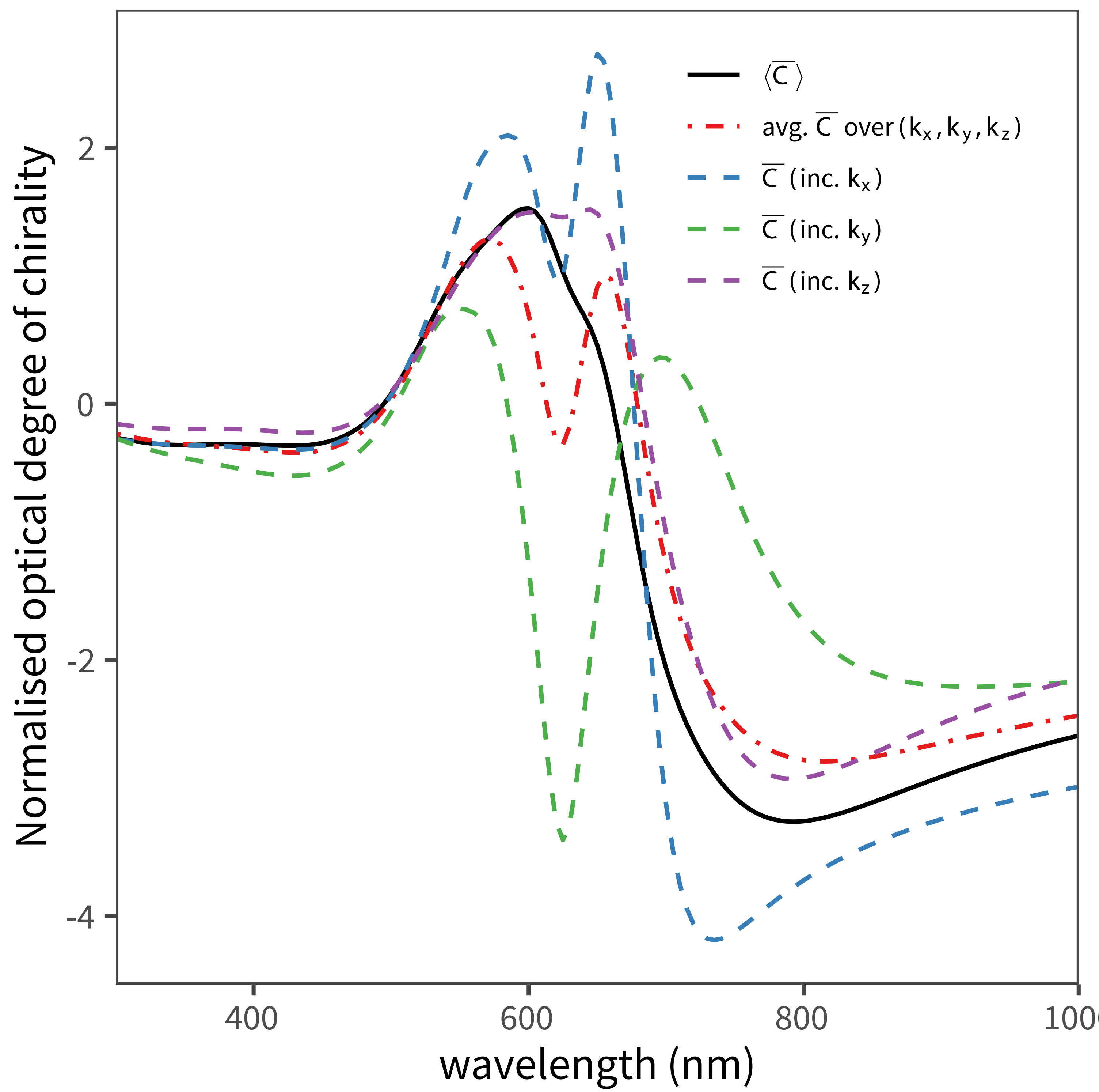


Optimal cubature on the sphere and other orientation averaging schemes
A. Penttila, K. Lumme JQSRT 112 (2011) 1741–1746

Efficient numerical orientation averaging of light scattering properties with a quasi-Monte-Carlo method
Y. Okada JQSRT 109 (2008) 1719–1742

"LARGE" CLUSTER: HELIX OF NANORODS





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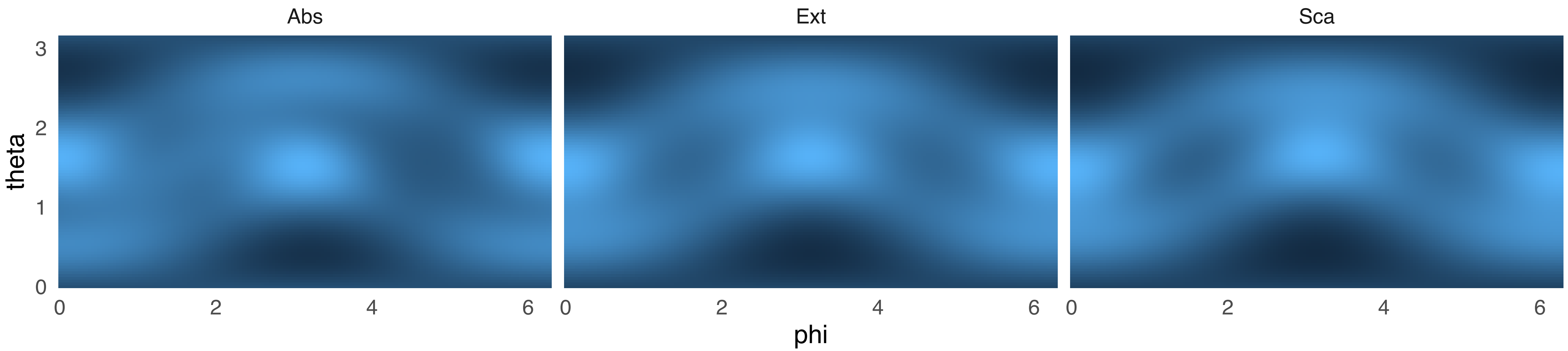
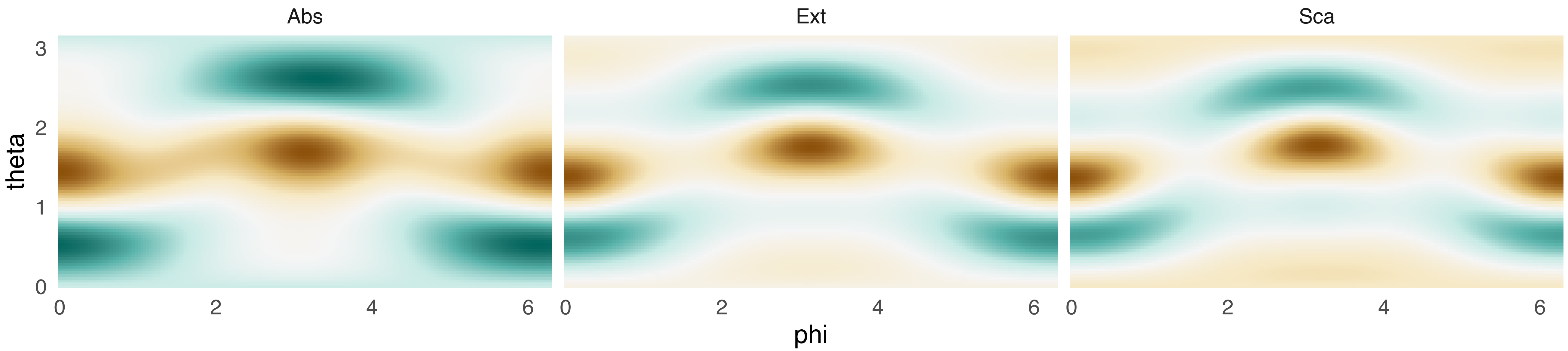
ANGULAR PATTERN: CROSS-SECTIONS & CIRCULAR DICHROISM

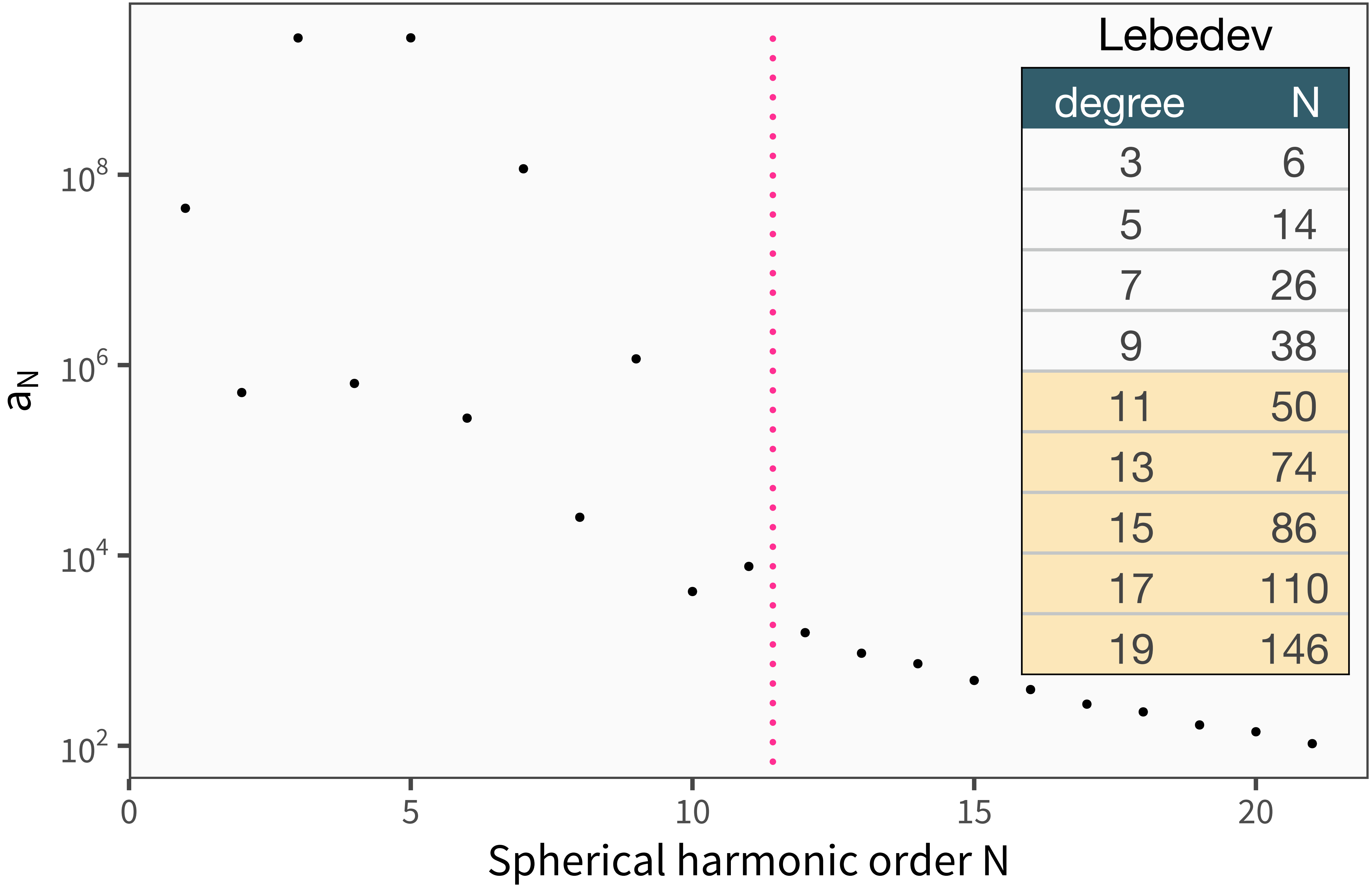
normalised average



normalised dichroism



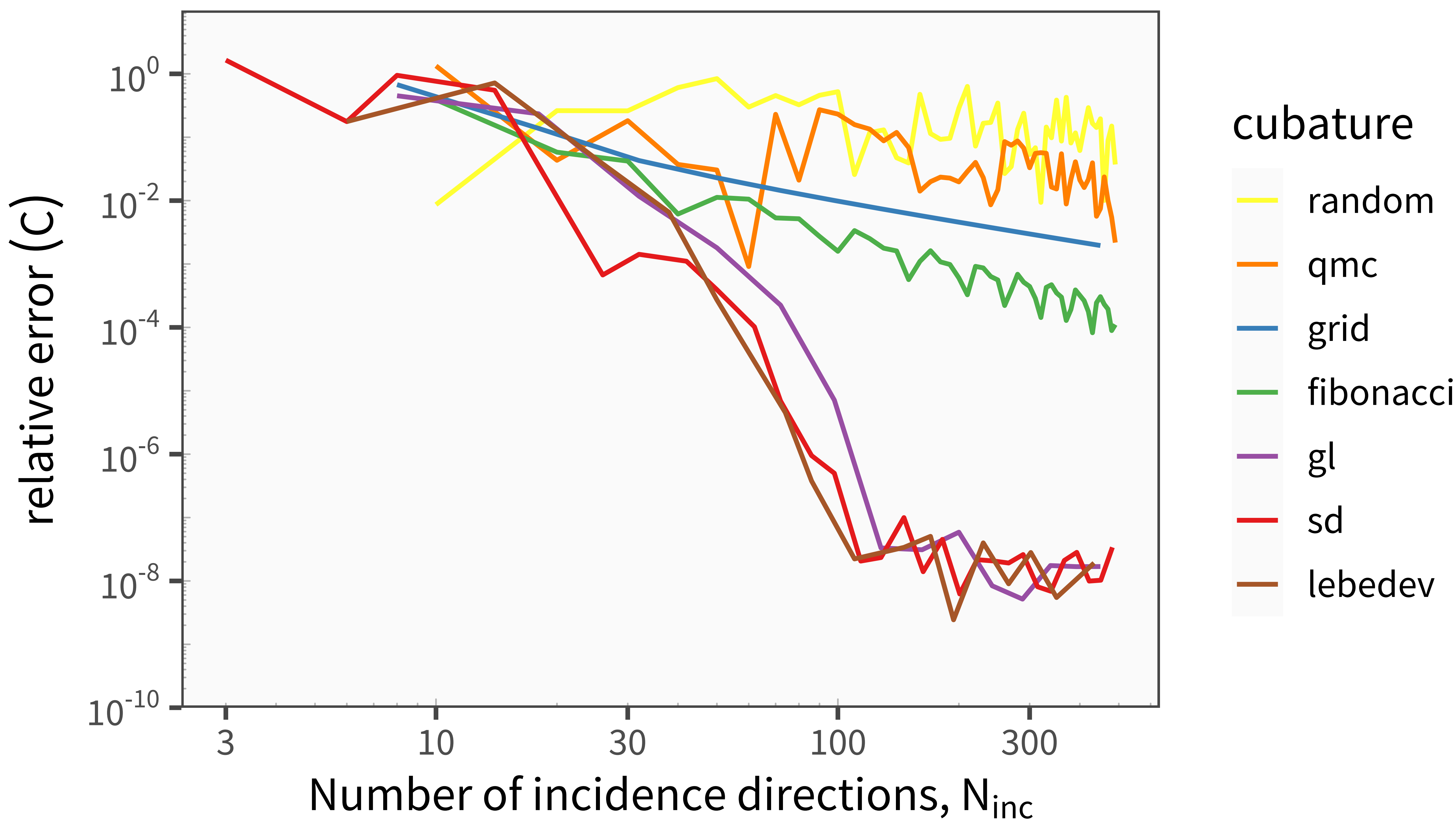
HOW MANY ANGLES? SPHERICAL HARMONIC DECOMPOSITION



degree	N
3	6
5	14
7	26
9	38
11	50
13	74
15	86
17	110
19	146

https://www.chebfun.org/examples/sphere/SphericalHarmonics.html

NANOROD HELIX: LOCAL DEGREE OF CHIRALITY



THANKS

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RUTHERFORD DISCOVERY FELLOWSHIPS

ROYAL SOCIETY TE APĀRANGI

The MacDiarmid Institute for Advanced Materials and Nanotechnology

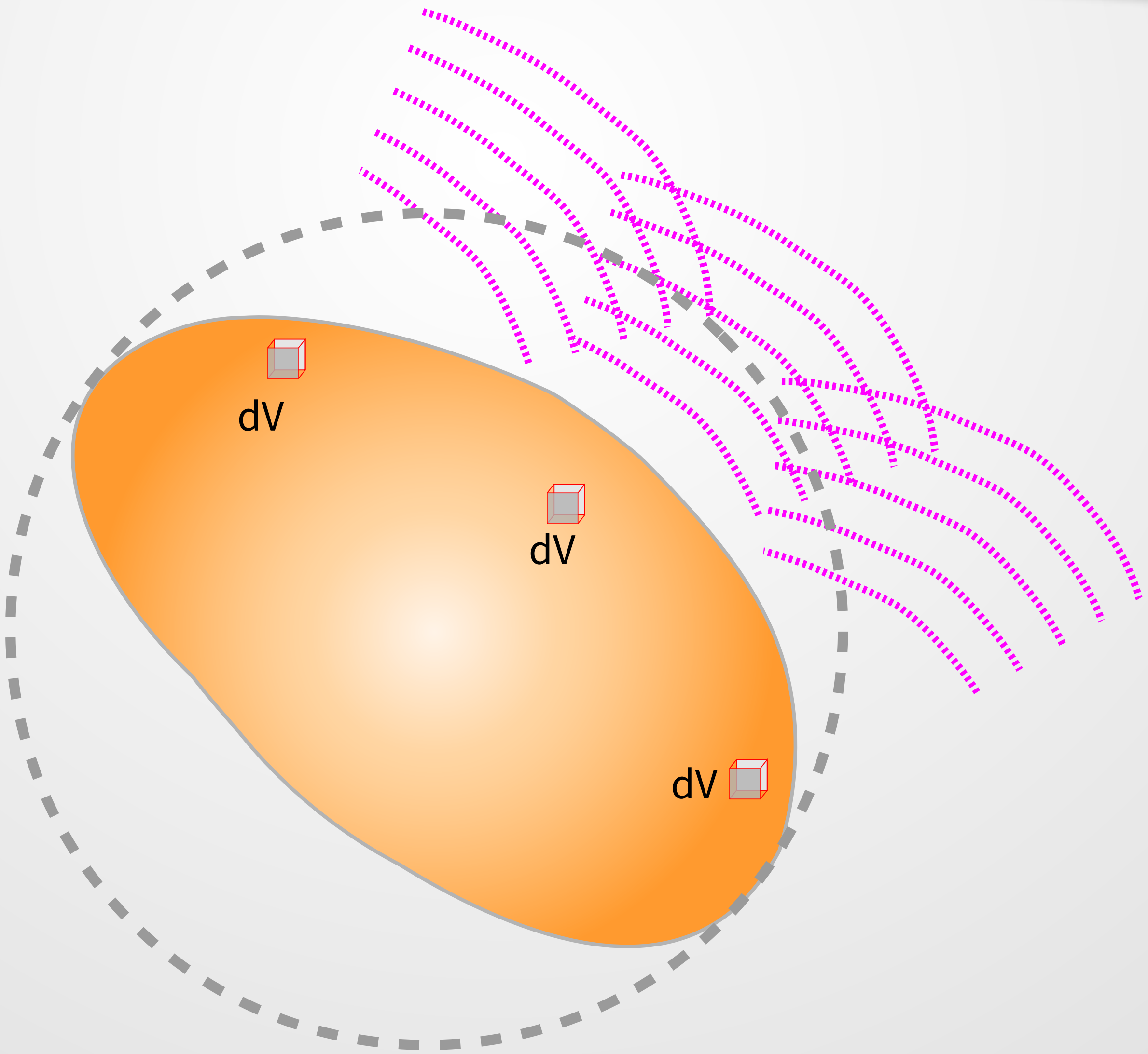
DODD-WALLS CENTRE for Photonic and Quantum Technologies

1897 VICTORIA UNIVERSITY OF WELLINGTON TE HERENGA WAKA

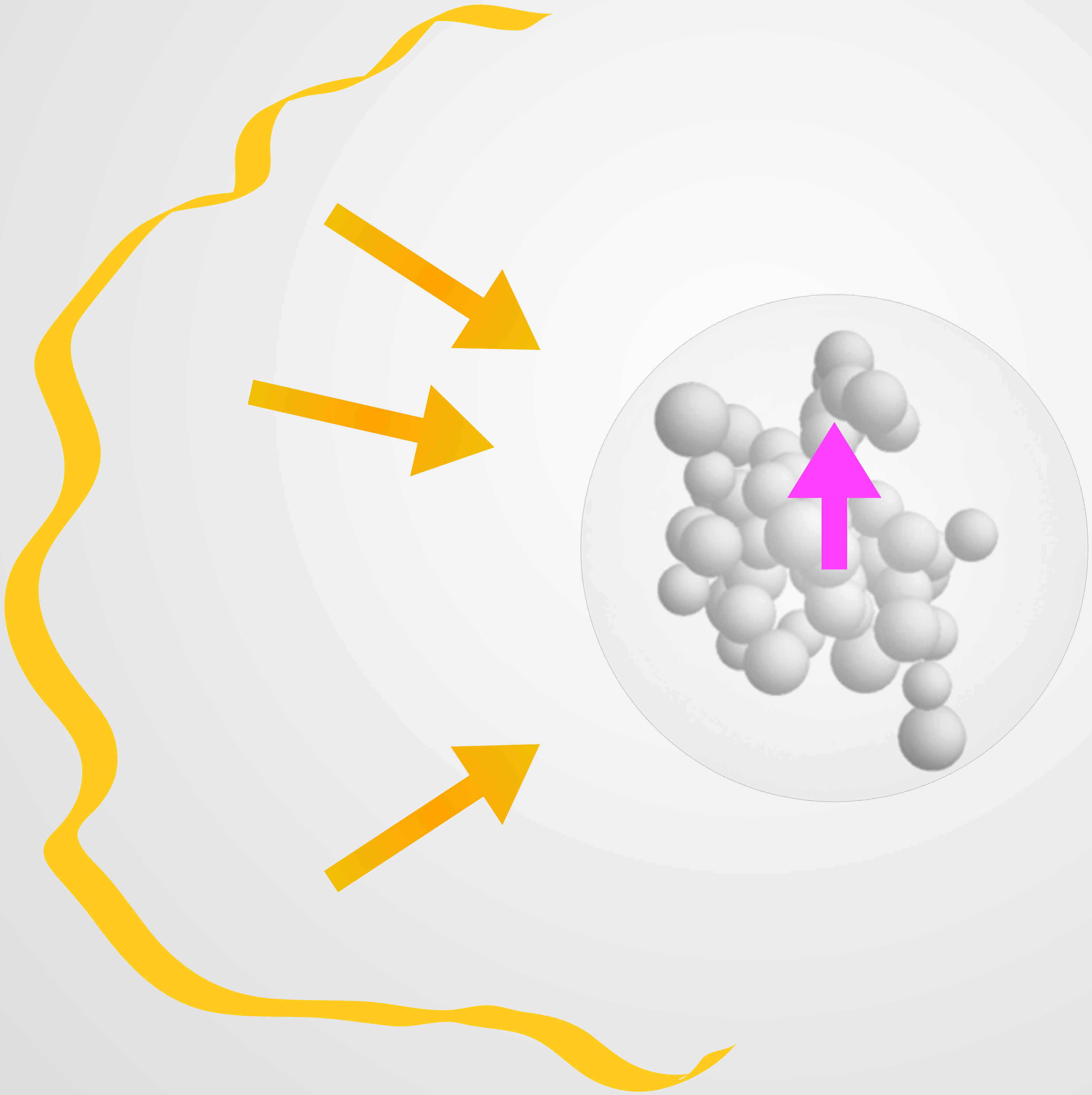
SUPPLEMENTARY SLIDES

RAYLEIGH'S HYPOTHESIS

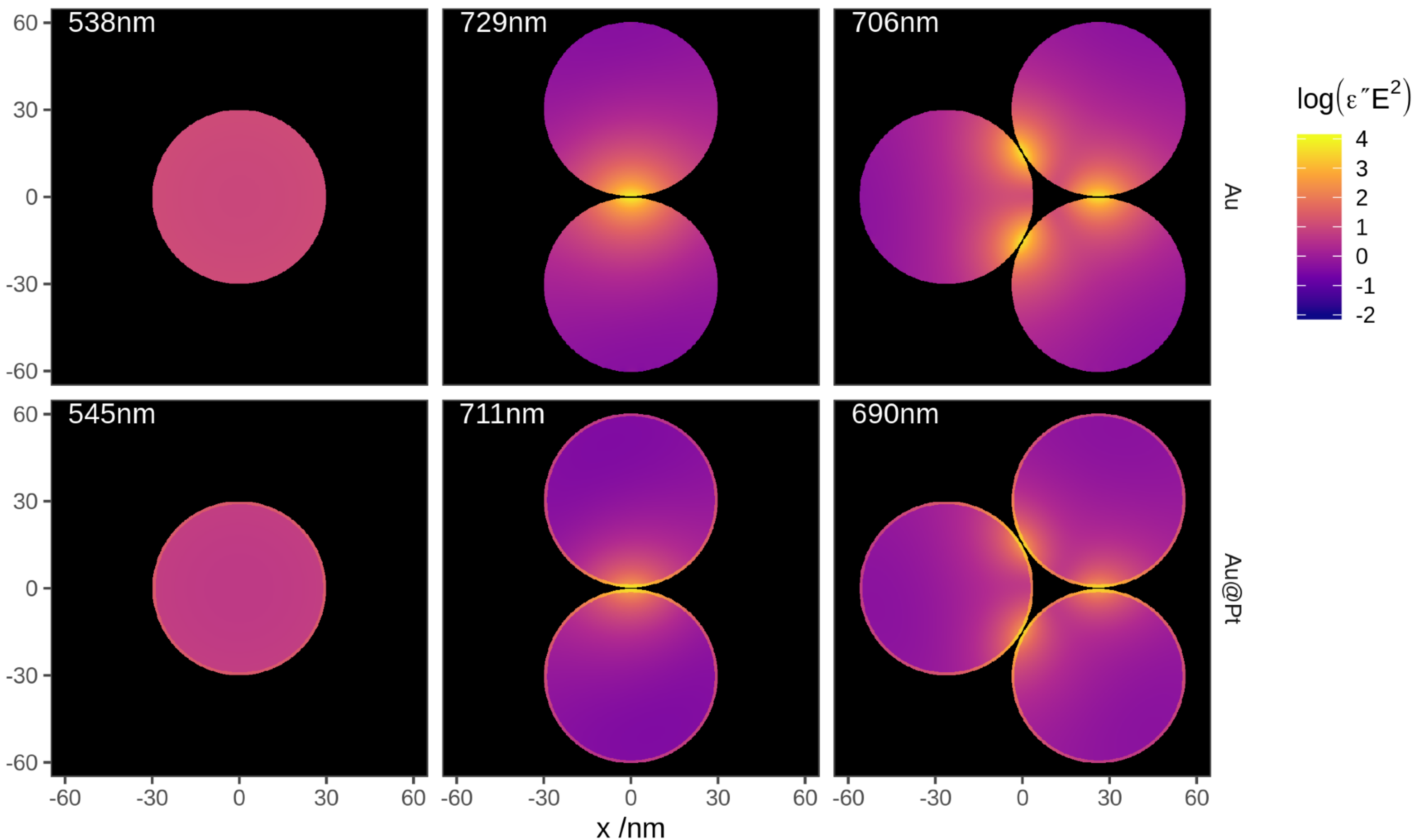
$$\mathbf{E}_{\text{sca}}(\mathbf{r}) = \sum_{n,m} p_{nm}\mathbf{M}_{nm}(k_1\mathbf{r}) + q_{nm}\mathbf{N}_{nm}(k_1\mathbf{r})$$



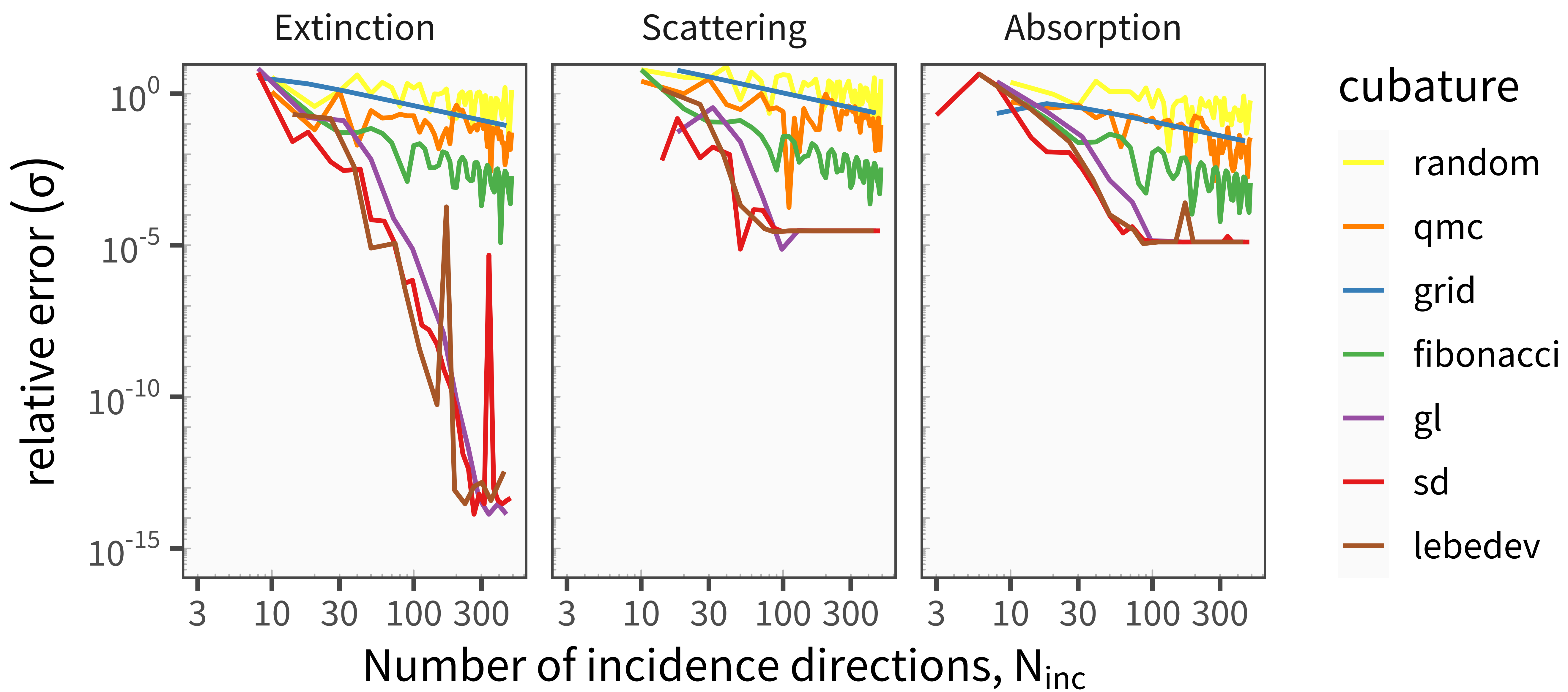
HOW MANY ANGLES ARE NEEDED? – RECIPROCITY¿



EXAMPLE: LOCAL ABSORPTION IN AU@PT NANO-TRIMERS

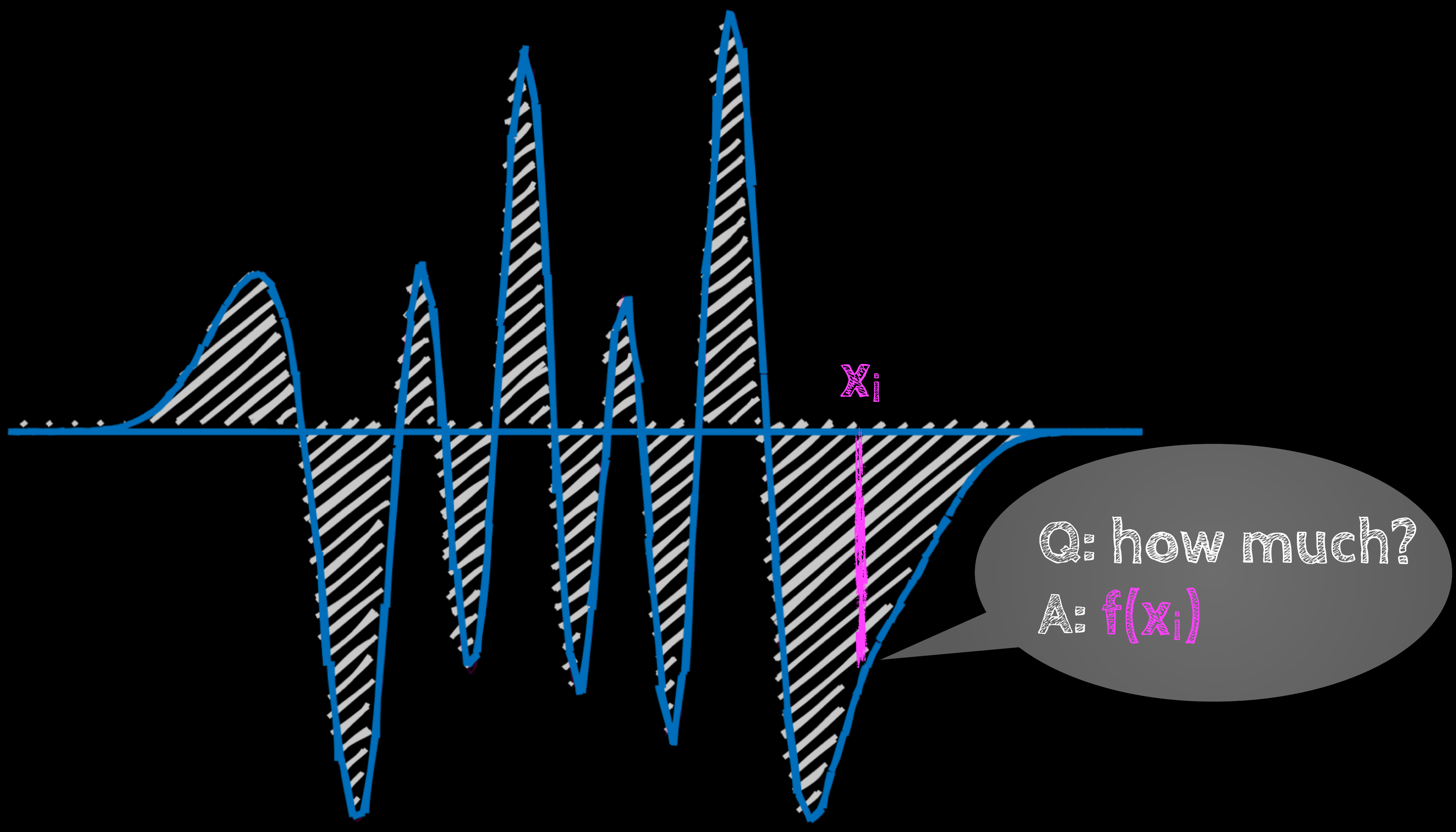


NANOROD HELIX: CIRCULAR DICHROISM



NUMERICAL QUADRATURE

$$\int_a^b f(x)\,\mathrm{d}x \approx (b-a)\sum_i w_i f(x_i)$$



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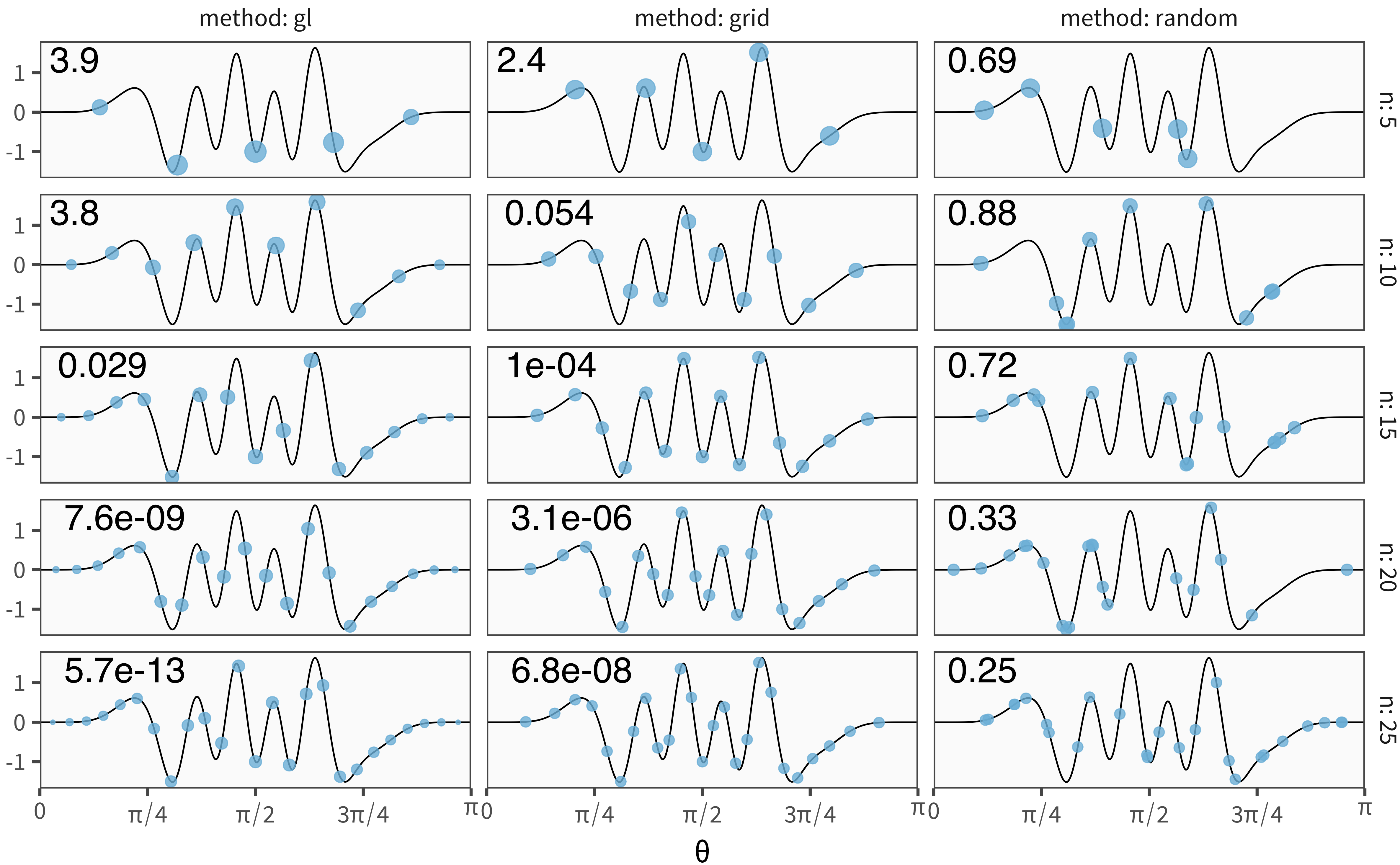
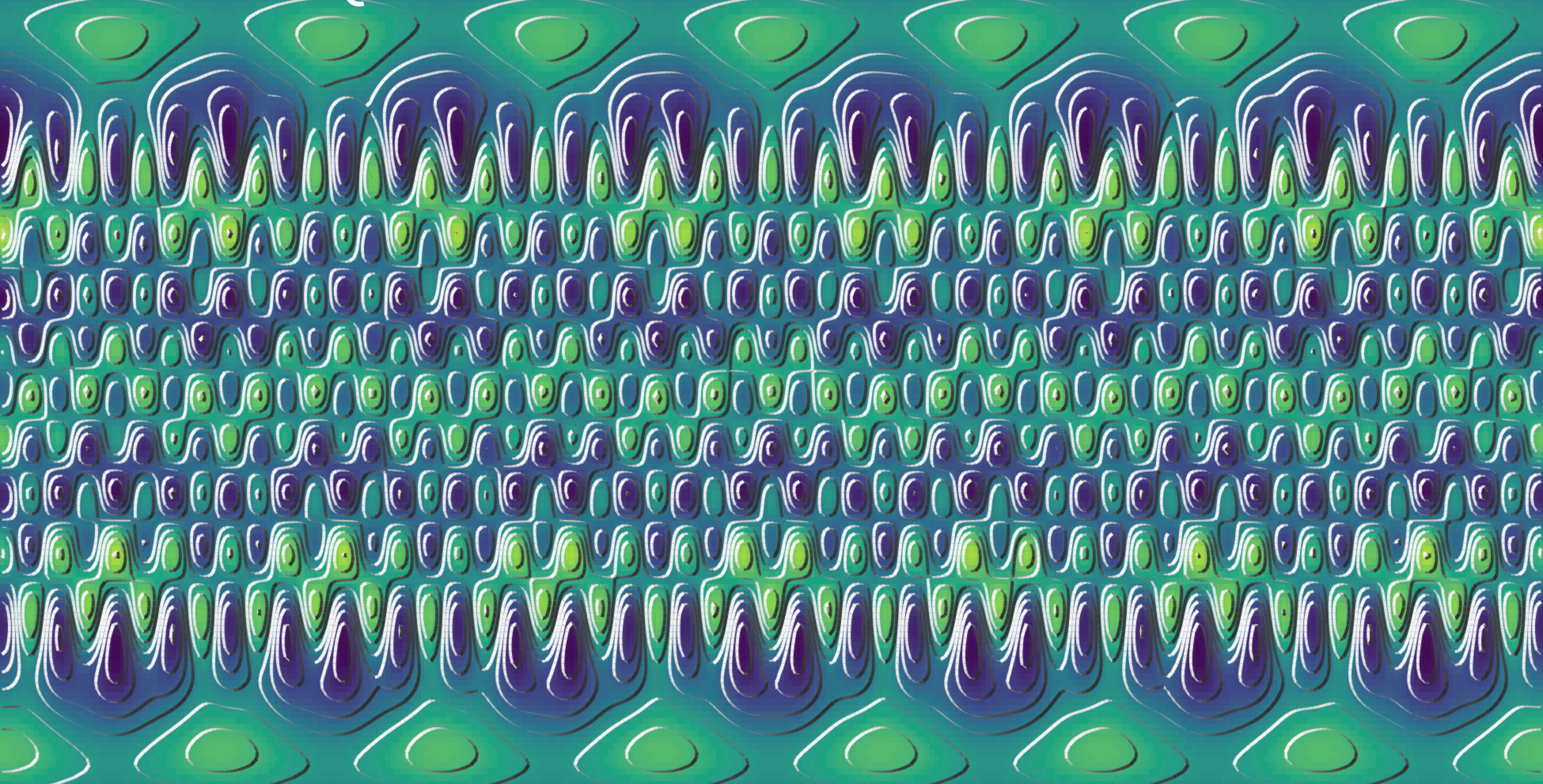


SPHERICAL QUADRATURE

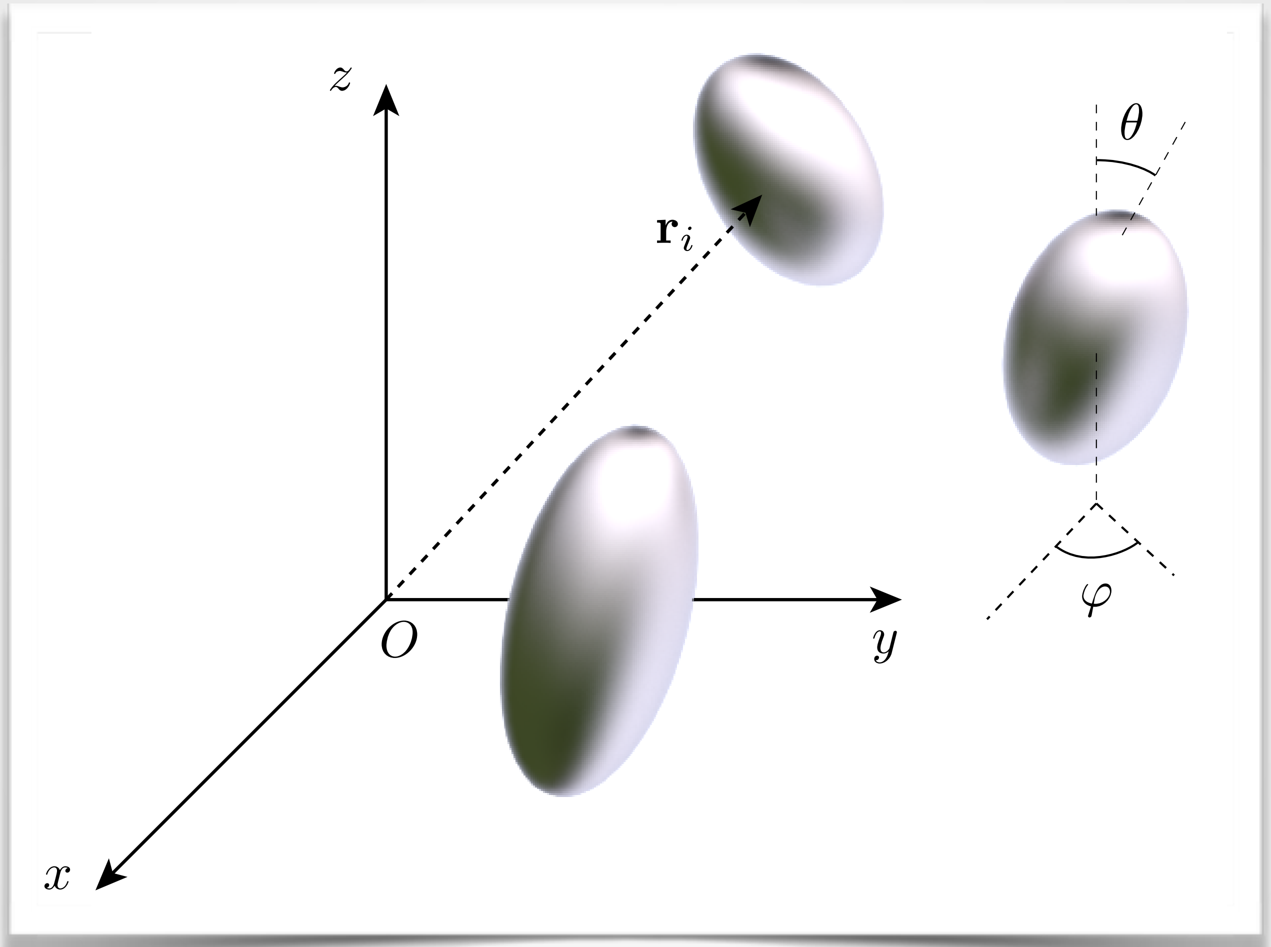


$$\frac{1}{4\pi}\int_0^{2\pi}\int_0^{\pi} f(\varphi,\theta)\sin\theta\,\mathrm{d}\theta\mathrm{d}\varphi \approx \sum_i w_i f(\varphi_i,\theta_i)$$

SPHERICAL HARMONIC (WHY L=35, M=27)

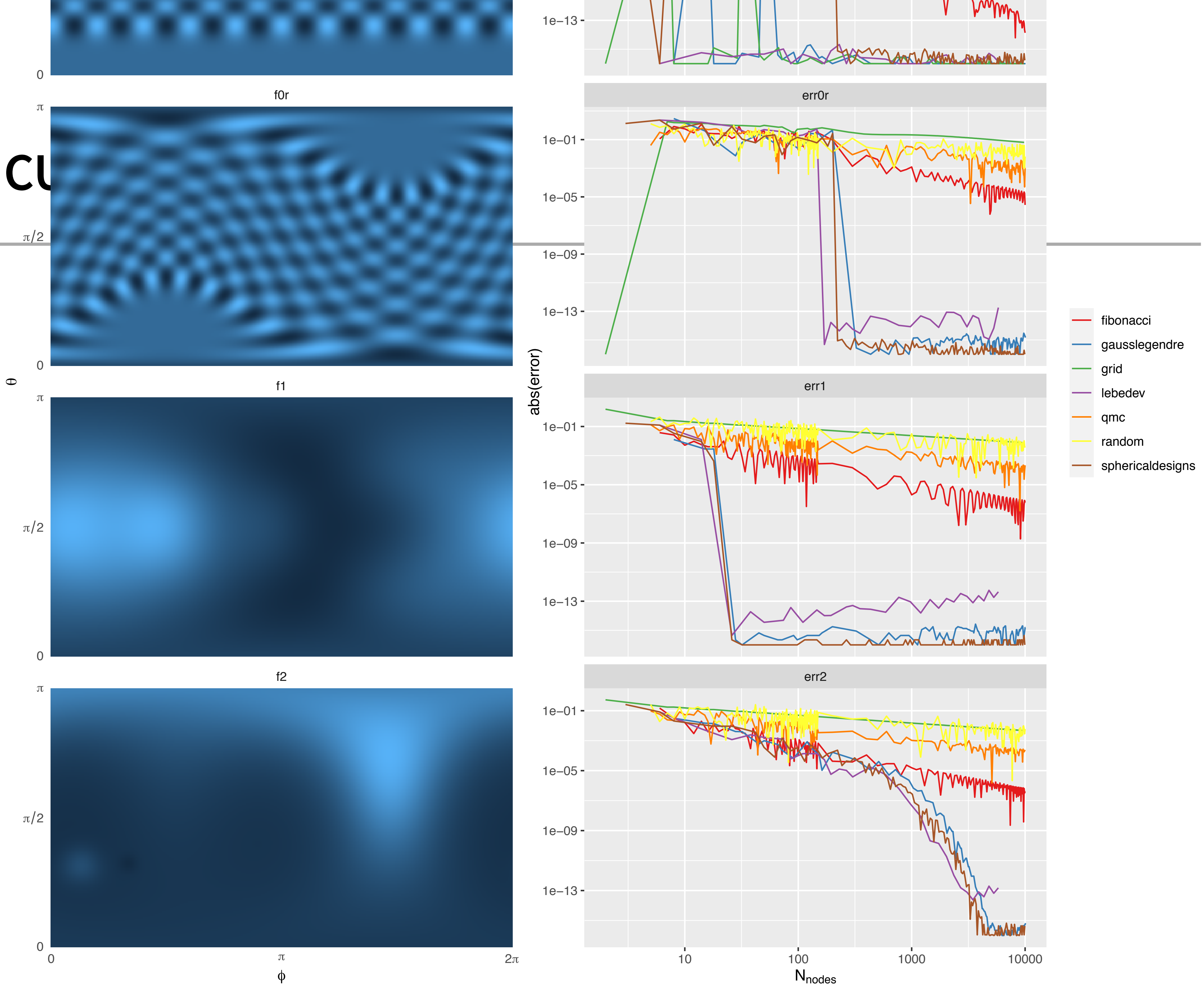
$$e^{27i\varphi}\sin^{27}(\theta)\left(180297\cos^{8}(\theta)-73164\cos^{6}(\theta)+8190\cos^{4}(\theta)-252\cos^{2}(\theta)+1\right)$$

NEAR-FIELD AND RECIPROCITY



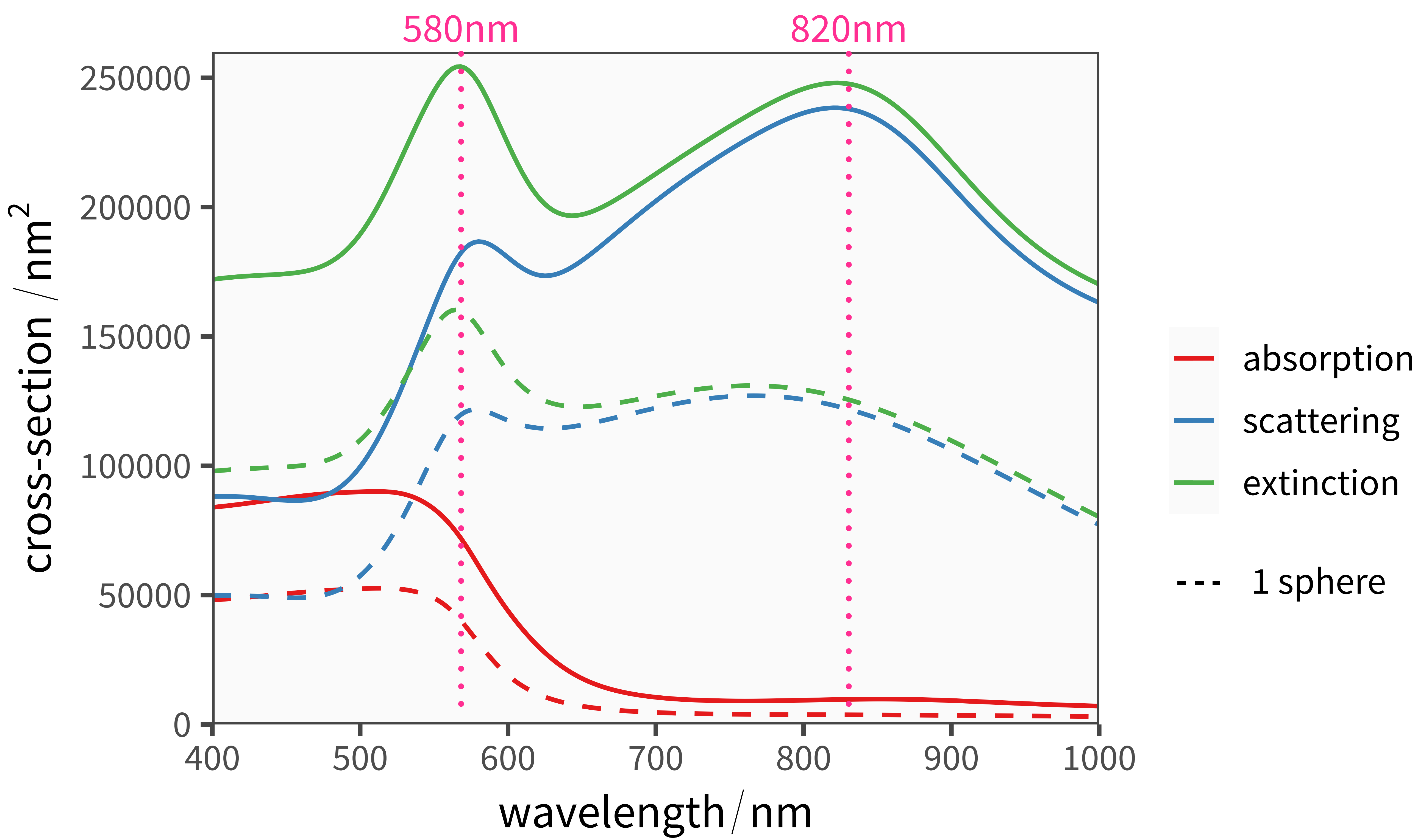
- NEAR-FIELD <-> FAR-FIELD RADIATION PATTERN OF A DIPOLE EMITTER
- DIPOLE SOURCE: COUPLES TO HIGH-ORDER MODES
- HIGH MULTIPOLE ORDER -> MANY LOBES IN THE RADIATION PATTERN

SPHERICAL CU



Casper Beentjes, "*Quadrature on a spherical surface*." Technical Report (2015)

SPHERICAL CUBATURE: FAR-FIELD CONVERGENCE



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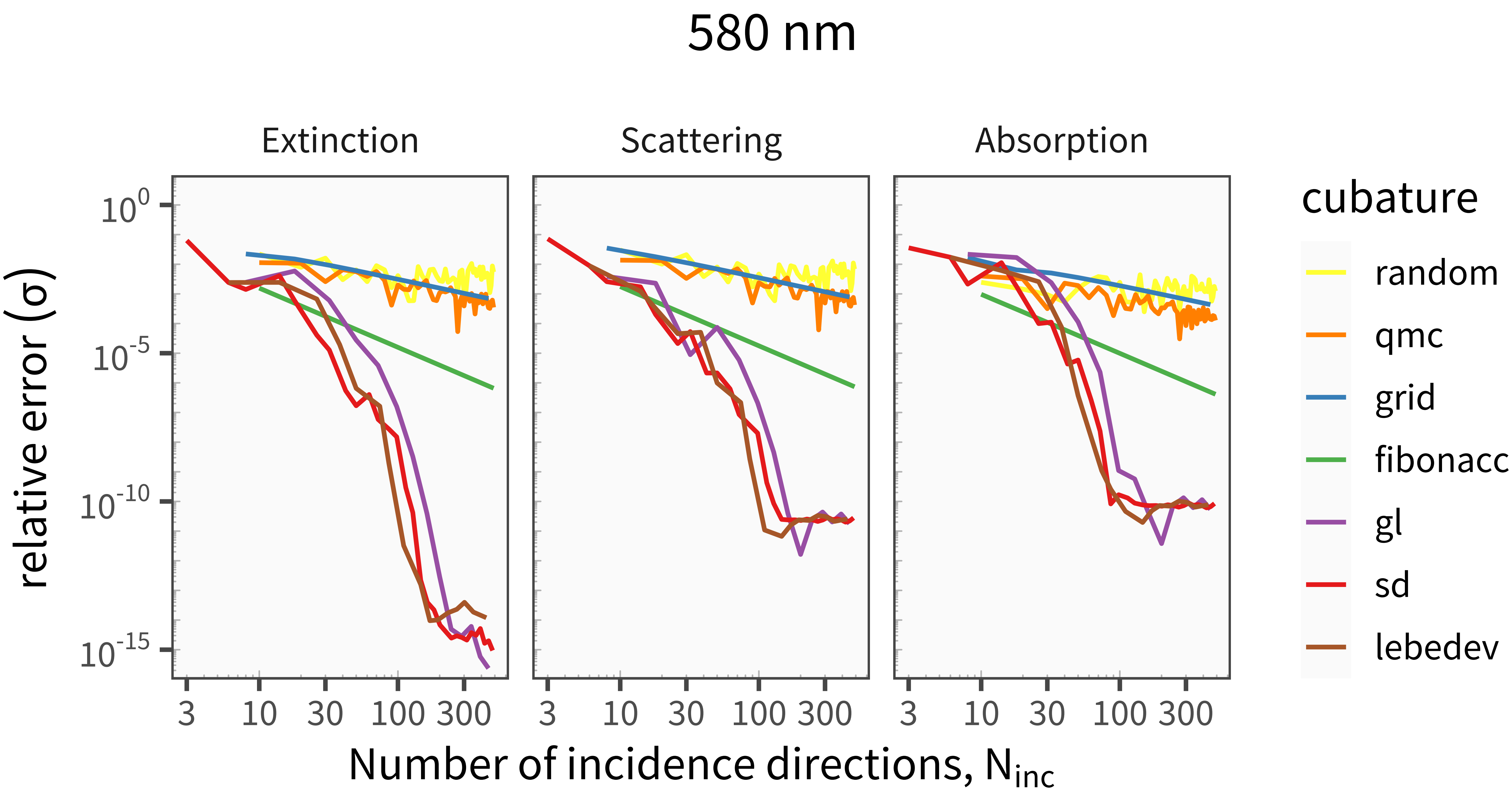

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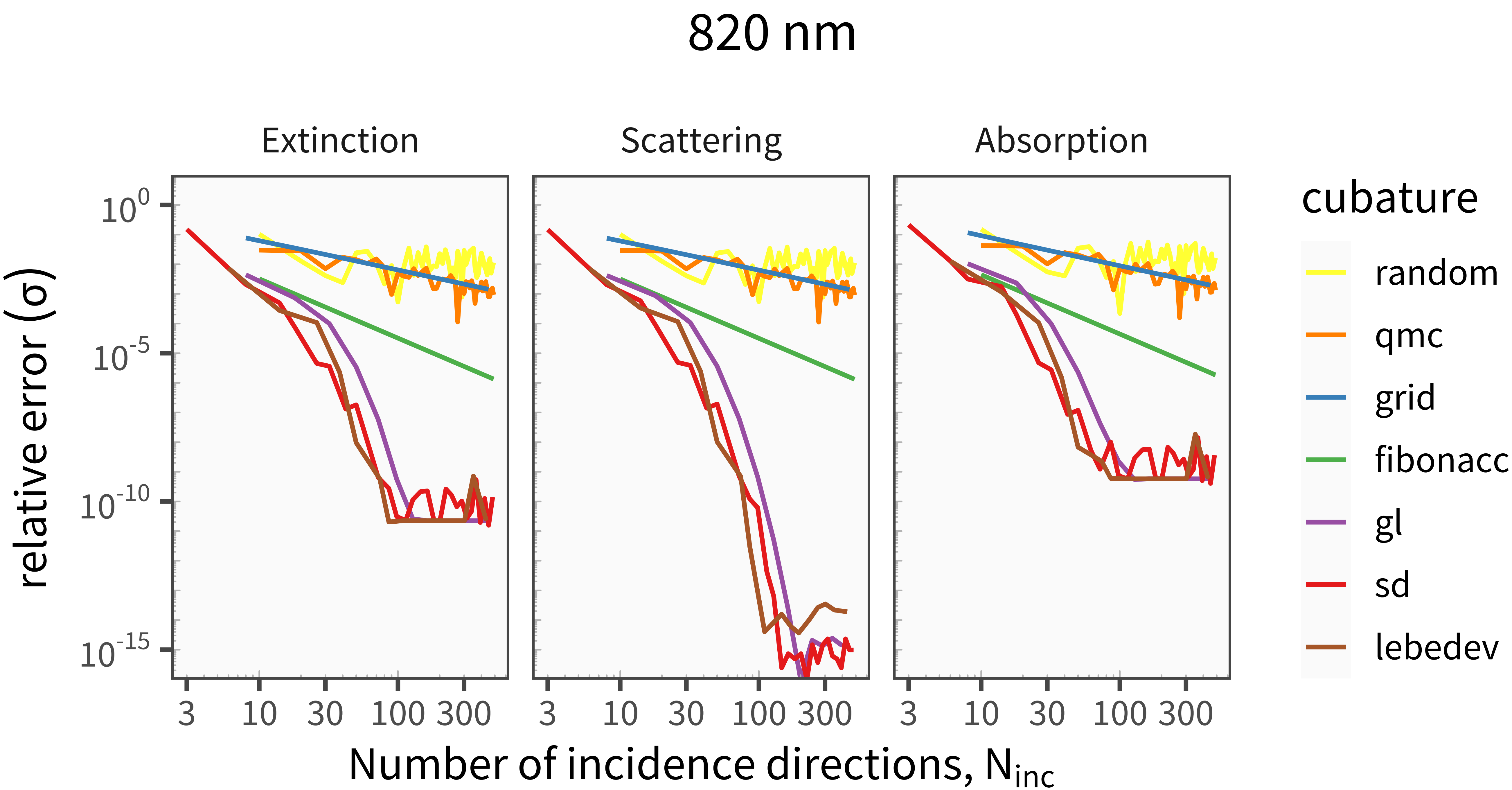
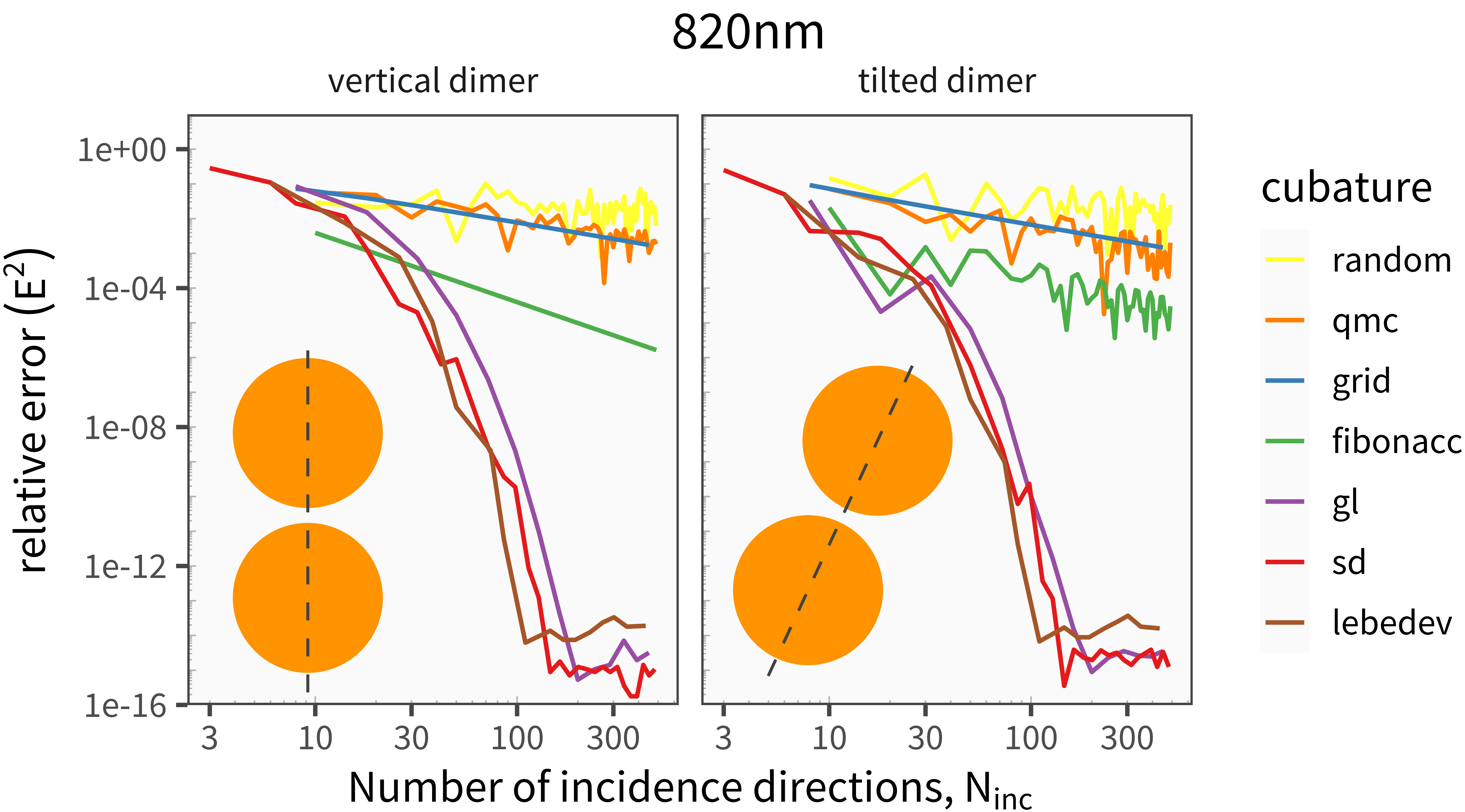


SPHERICAL CUBATURE: NEAR-FIELD CONVERGENCE



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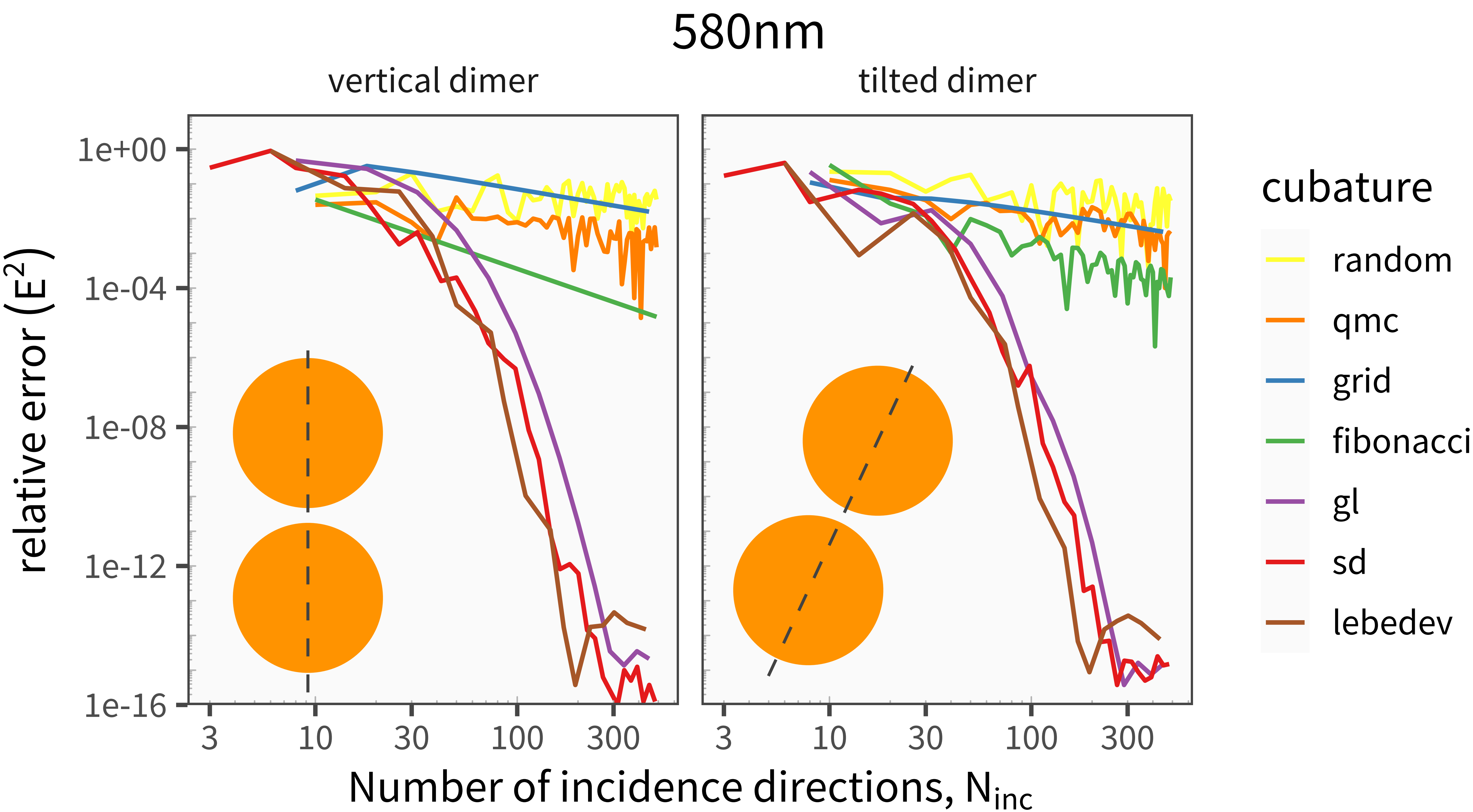
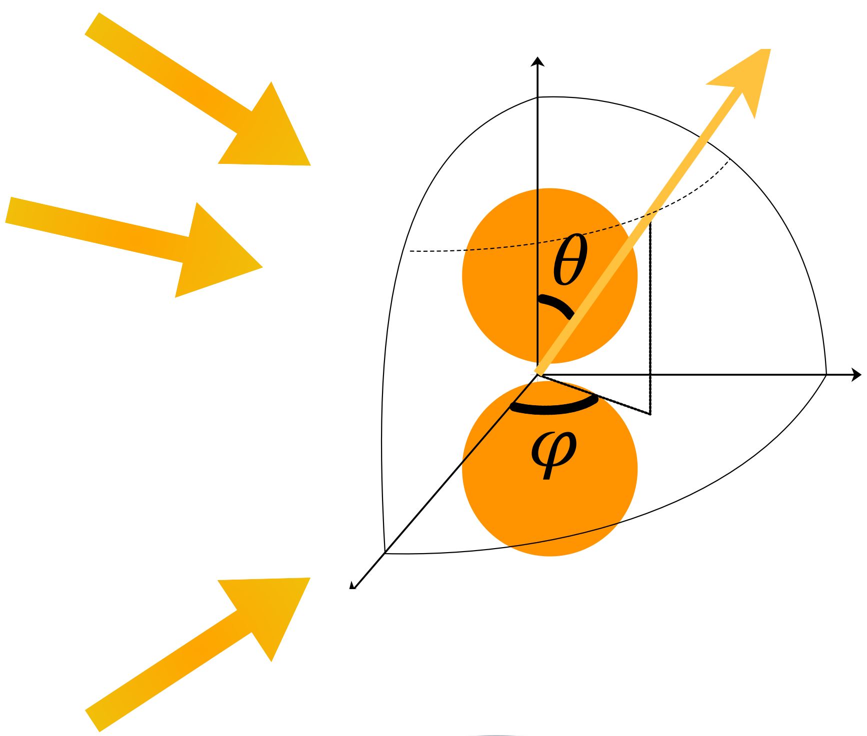


DIFFERENTIAL CROSS-SECTIONS – WHAT IS THE PATTERN?



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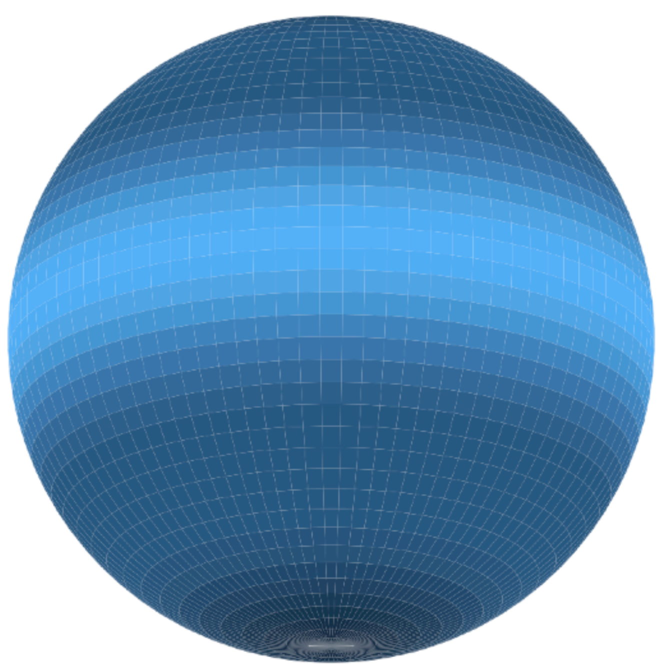
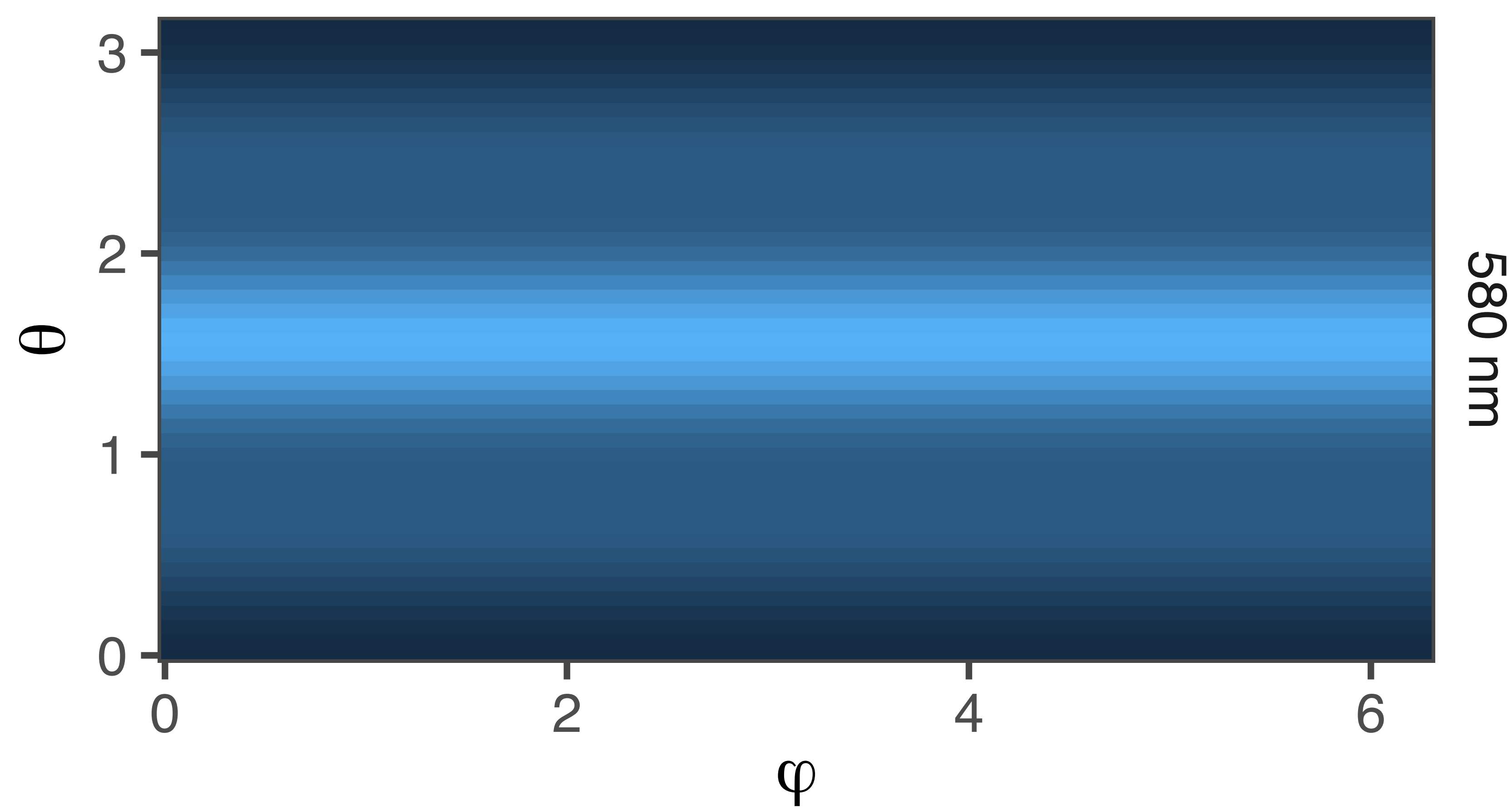
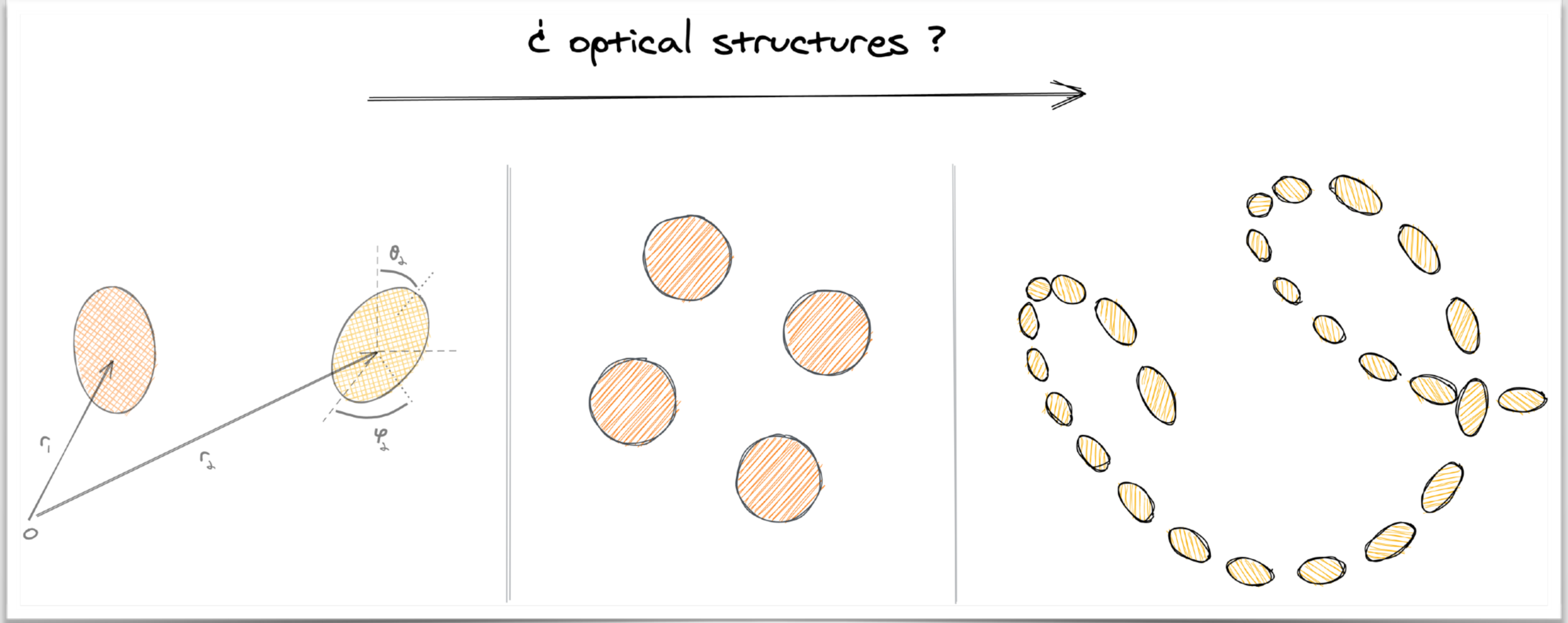




GEOMETRY OPTIMISATION – FROM DIMER TO HELIX (?)



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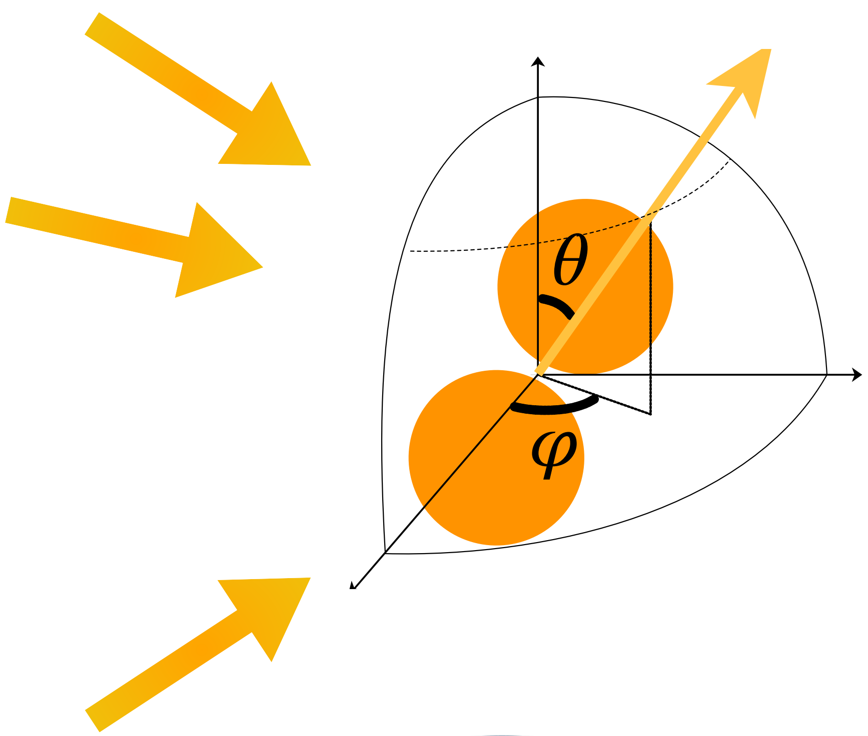

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