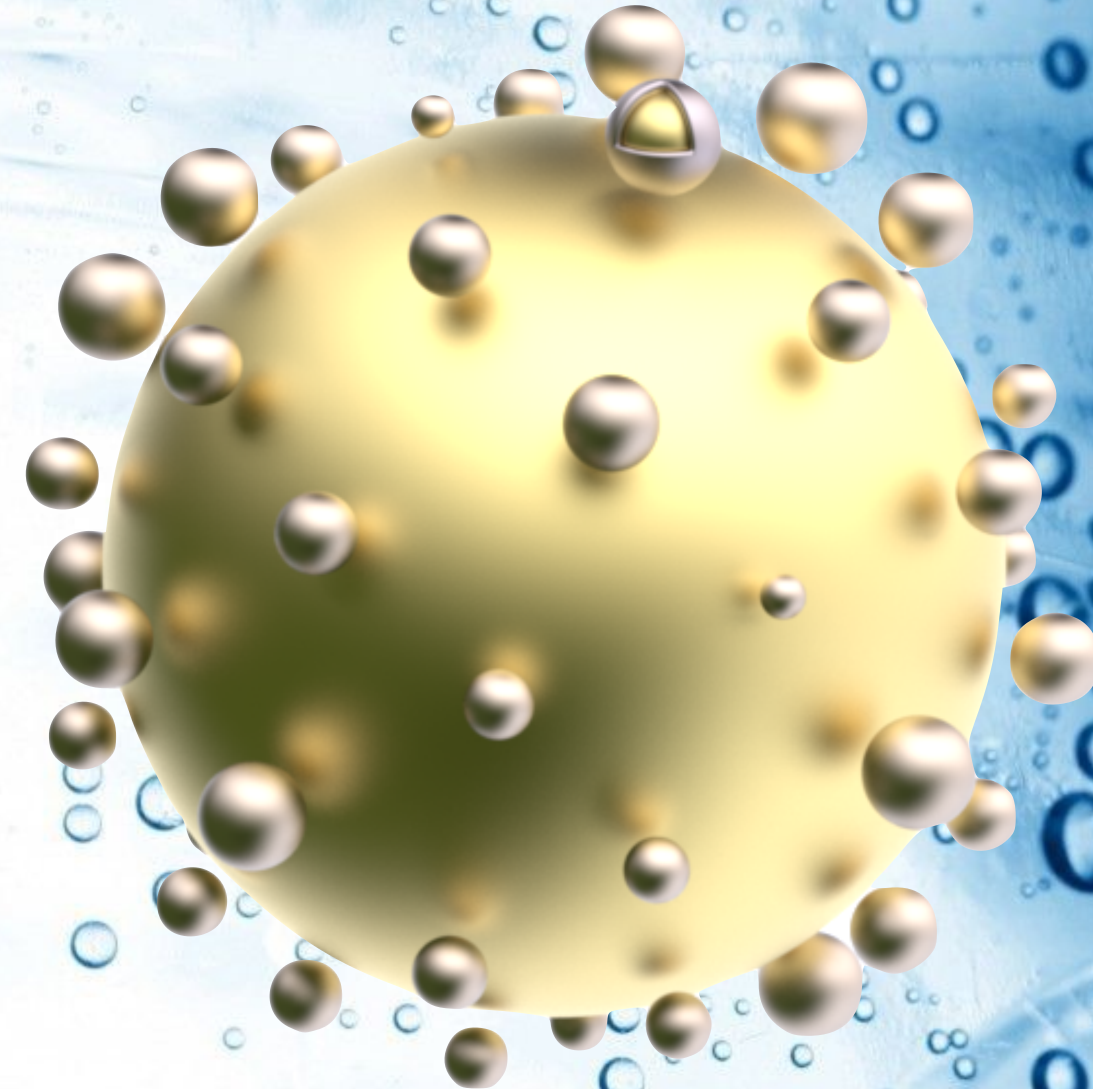


MODELLING LIGHT ABSORPTION IN HYBRID CORE-SATELLITE METAL NANOSTRUCTURES

M. HERRAN[1], A. SOUSA-CASTILLO[1,2], E. CORTÉS[1]

S. GLUKHOVA, E. LE RU, **B. AUGUIÉ[3]***

[1] Nanoinstitute Munich, Ludwig-Maximilians-Universitat, Germany

[2] Centro de Investigaciones Biomédicas, Universidade de Vigo, Spain

[3] School of Chemical & Physical Sciences, Victoria University of Wellington, Aotearoa NZ

OUTLINE

1. INTRODUCTION

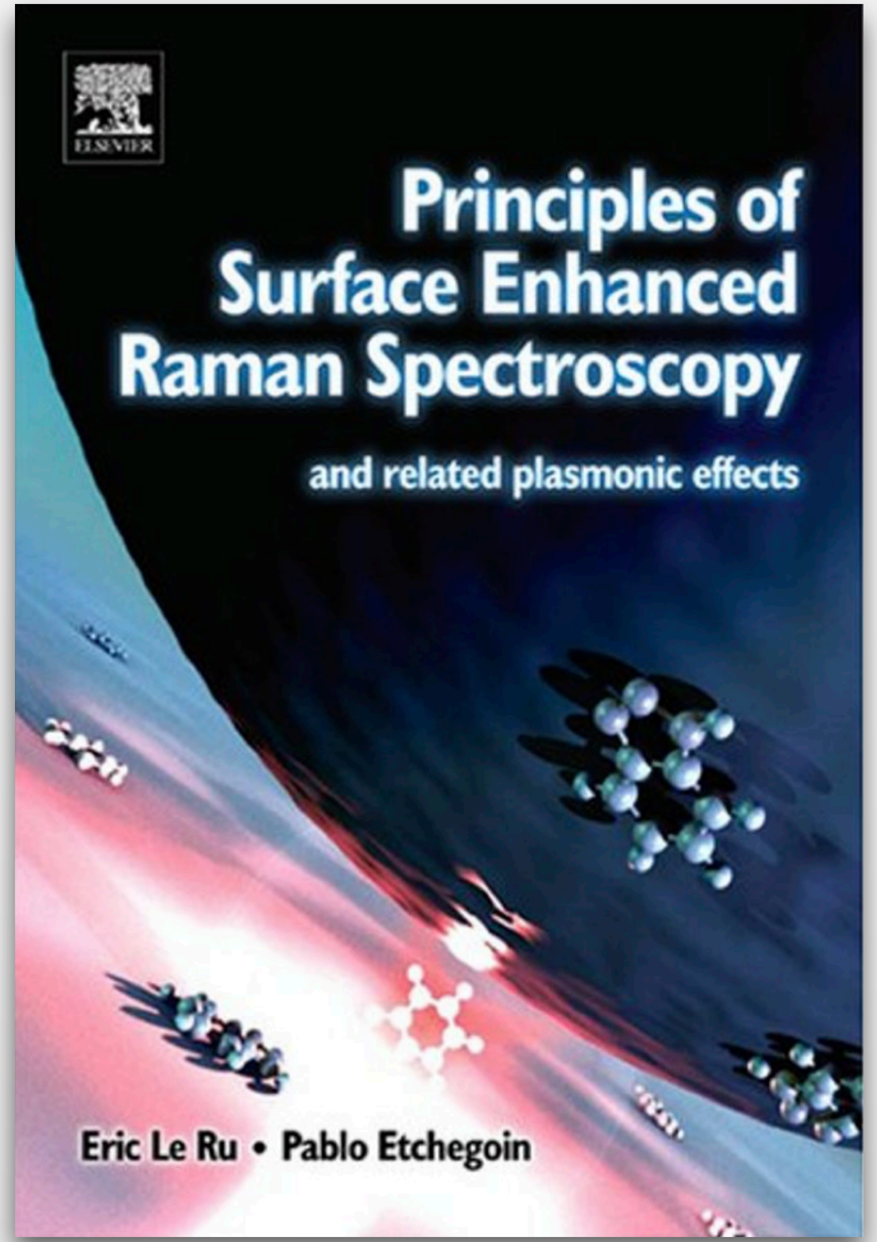
Nano-optics lab at Victoria University

T-matrix projects, lattice resonances, plasmonic chirality, …

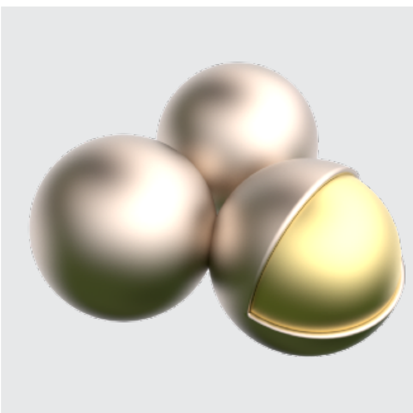
2. ABSORPTION IN CORE-SATELLITE NANOSTRUCTURES

Generalised coupled-dipole model for core-satellite nanostructures. Nanoscale, 2023

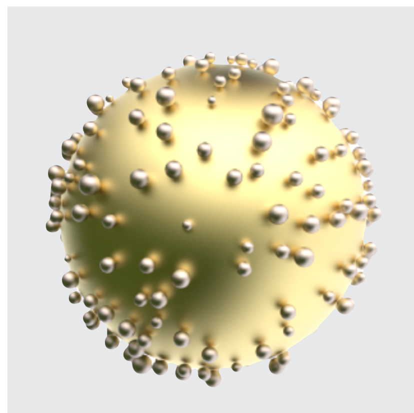
'RAMAN LAB' – PABLO ETCHEGOIN, ERIC LE RU



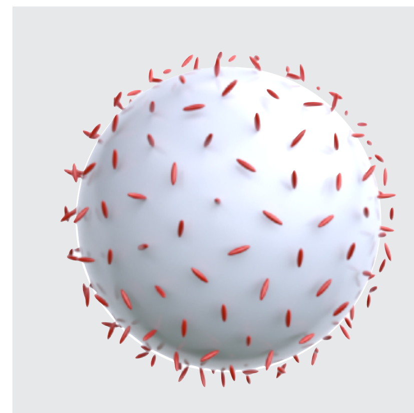
PRESENT / NOT YET FORGOTTEN RESEARCH INTERESTS



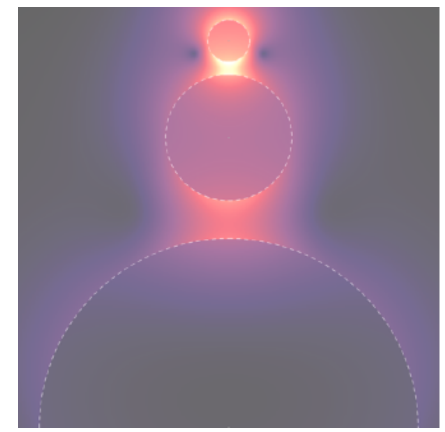
PARTICLE CLUSTERS



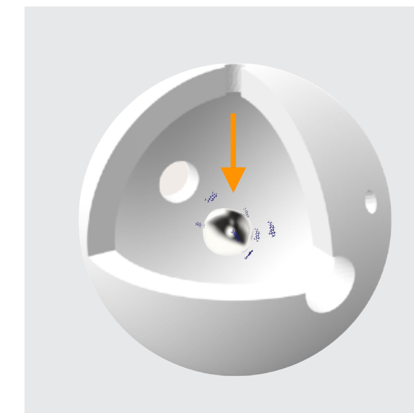
CORE-SATELLITES



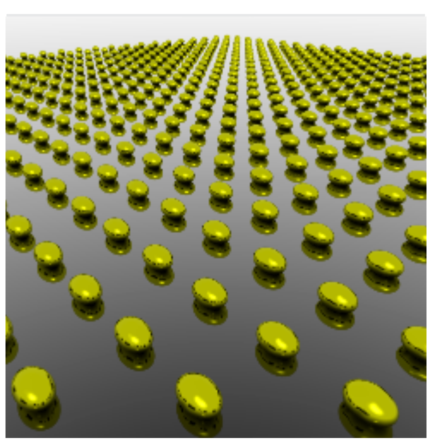
COUPLED DIPOLES



T-MATRIX

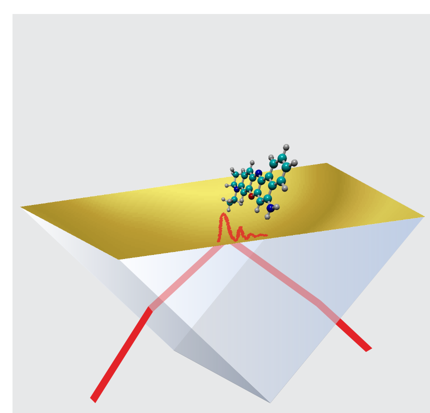


INTEGRATING SPHERE
UV–VIS SPECTROSCOPY

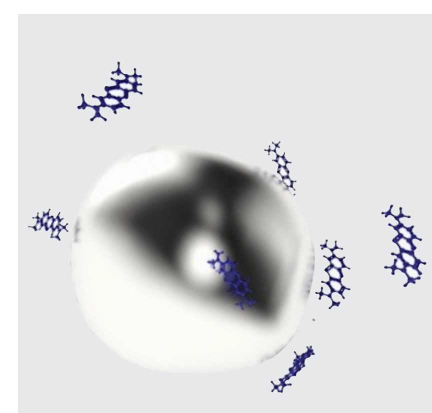


LATTICE RESONANCES

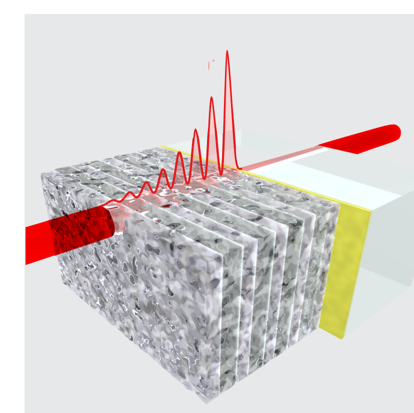
PLASMONIC CHIRALITY



SPR

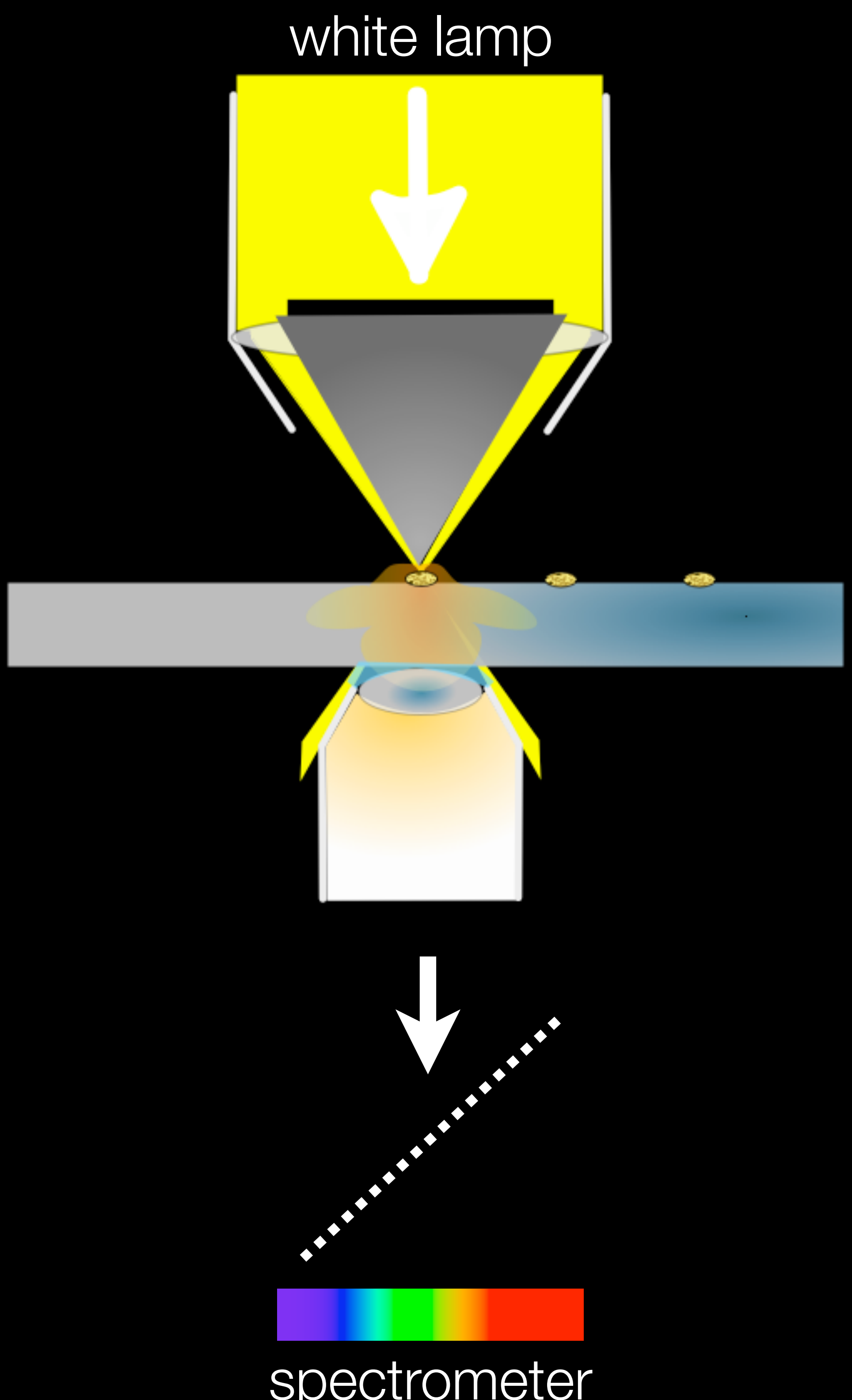


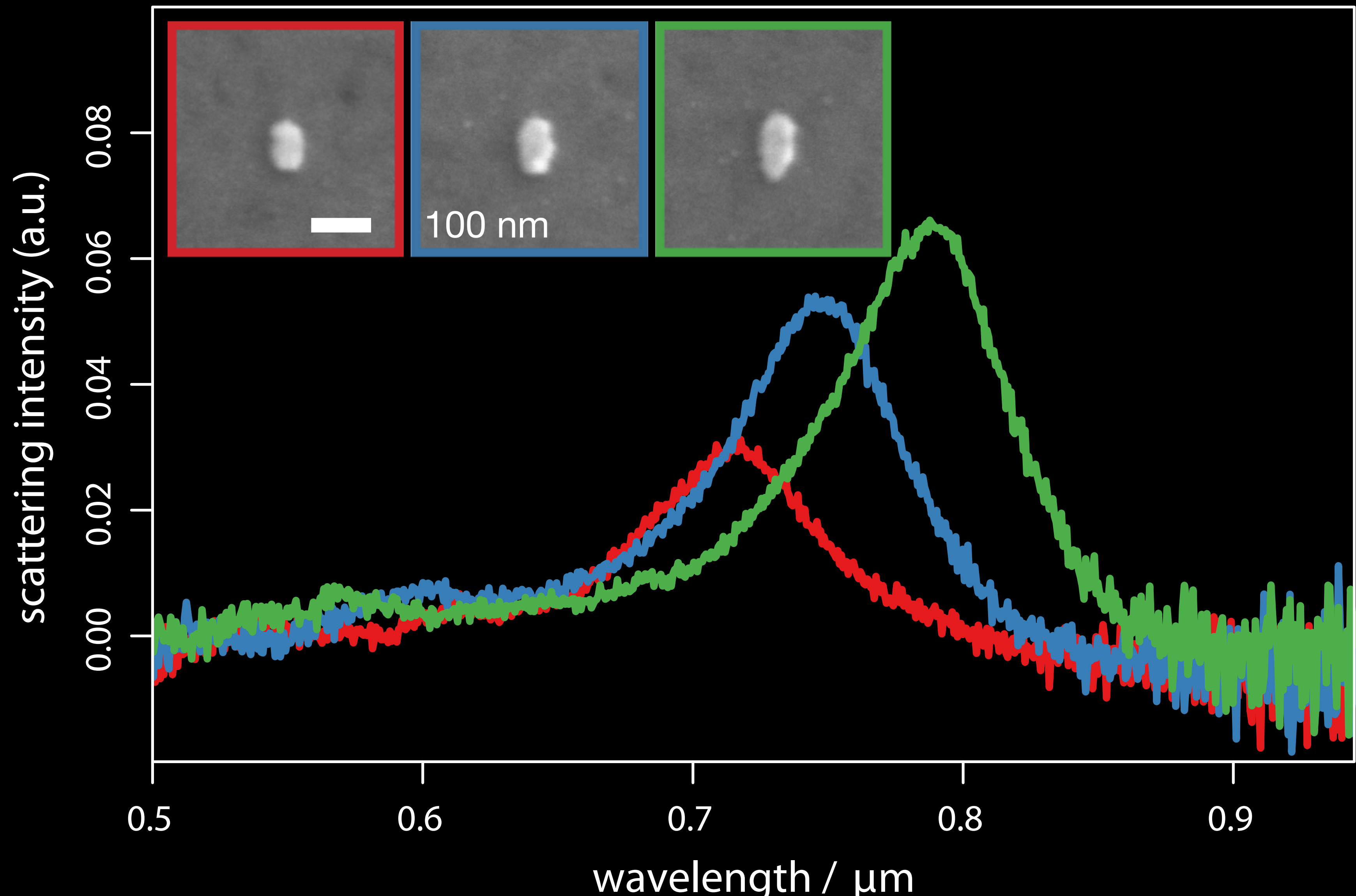
SERS

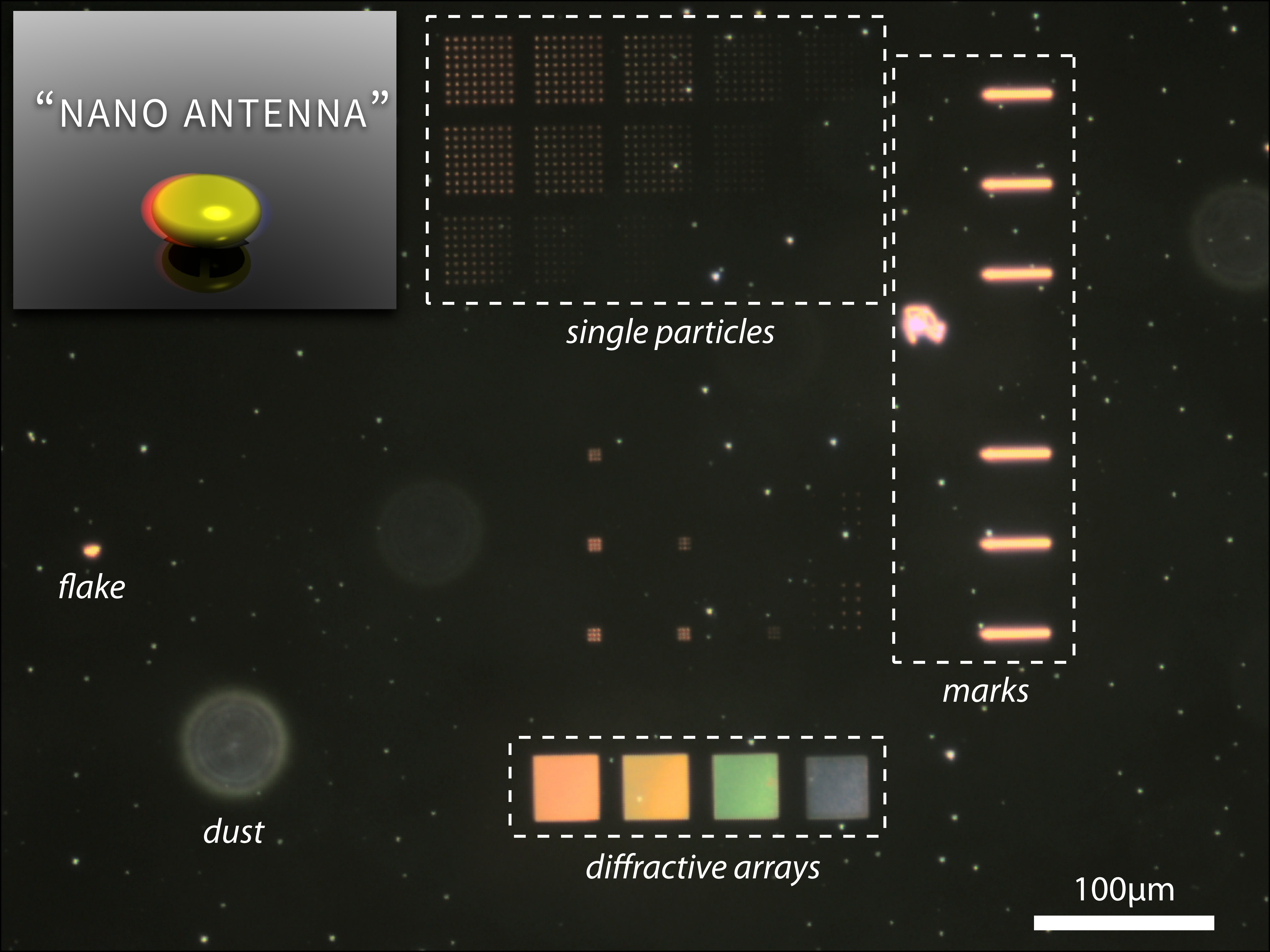


TAMM PLASMONS

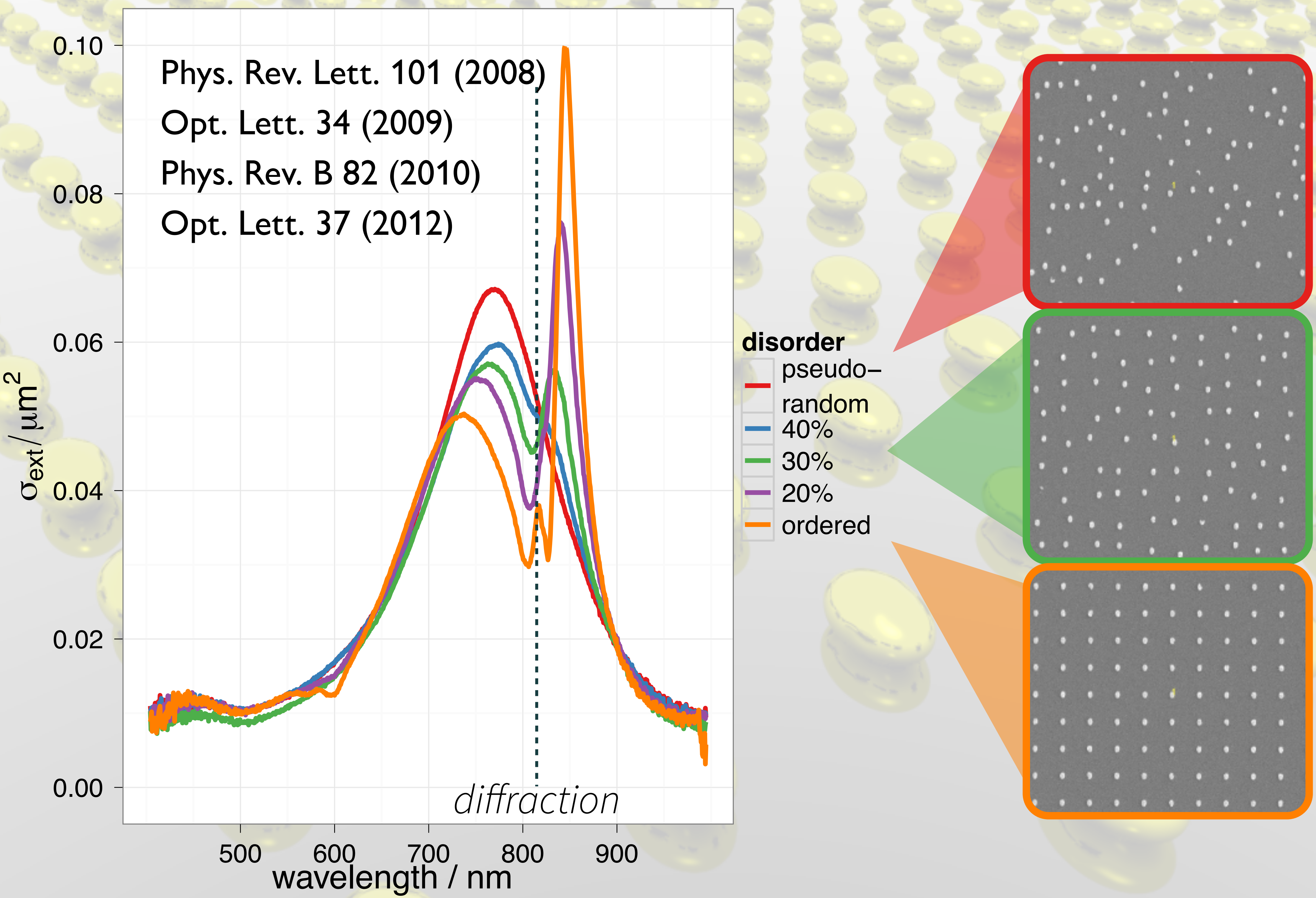
“NANO ANTENNAS”

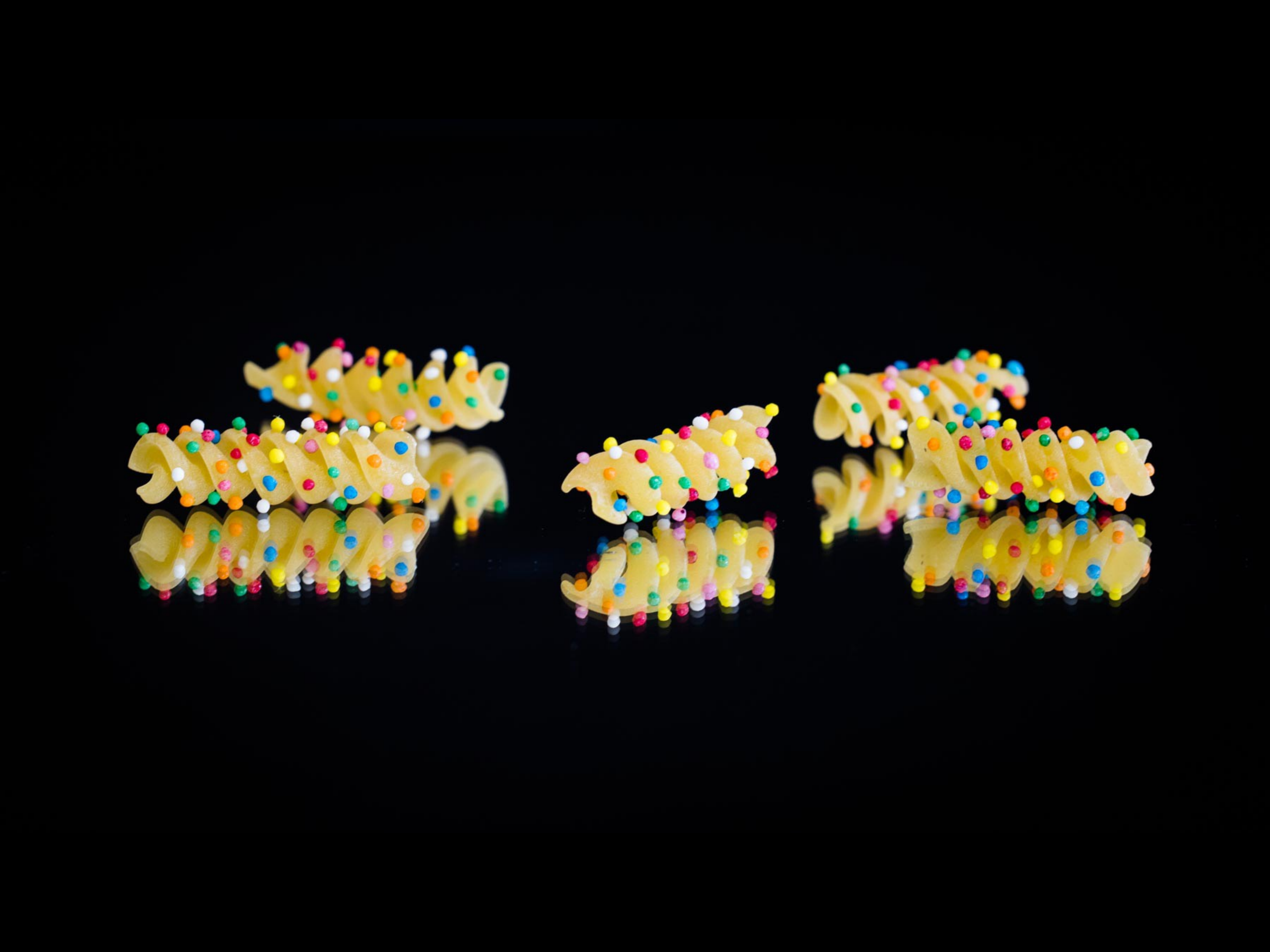





"NANO ANTENNA"
single particles
marks
flake
dust
diffractive arrays
100μm

2D array: lattice resonances

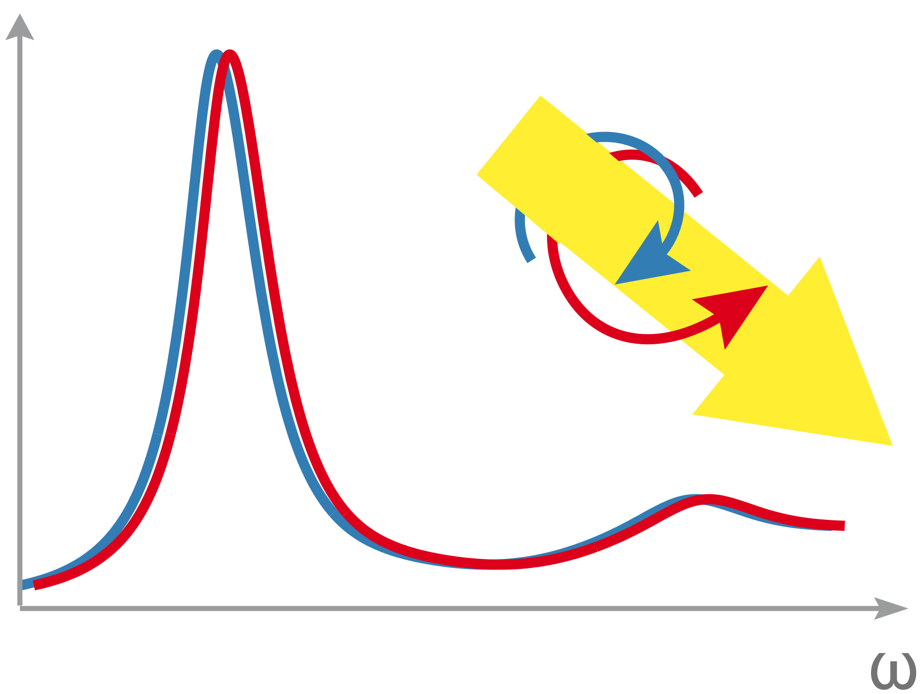




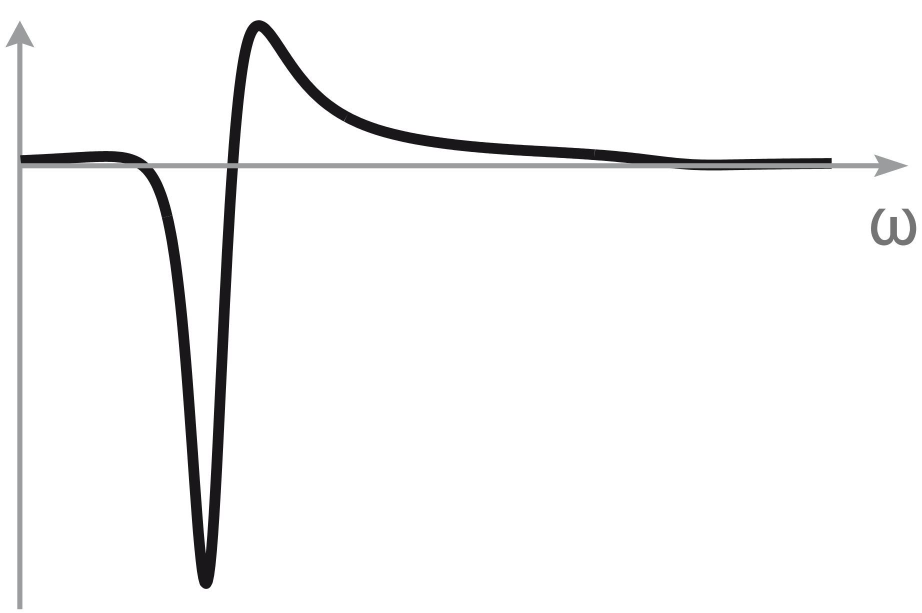
3D: PLASMONIC CHIRALITY

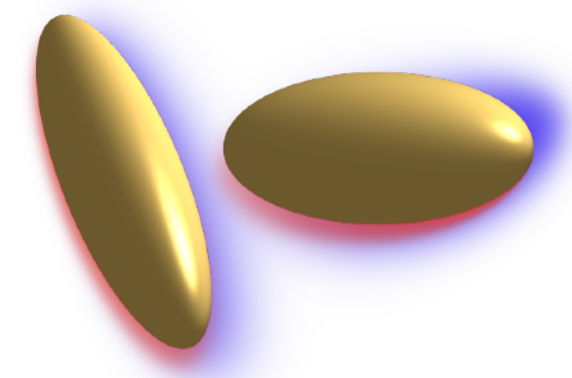
Angew. Chem. Int. Ed. 50 (2011)
J. Phys. Chem. Lett. 2 (2011)
Nano Today 6 (2011)

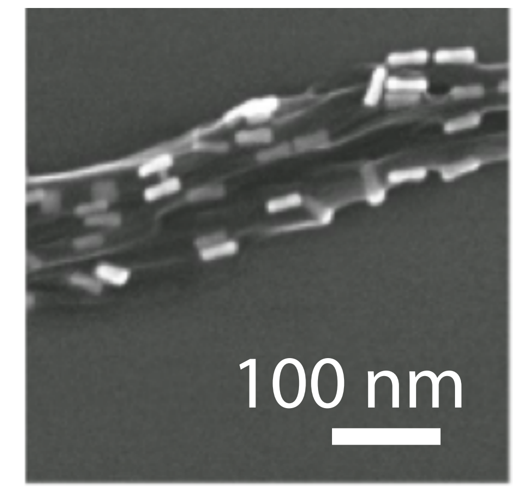
ABSORBANCE



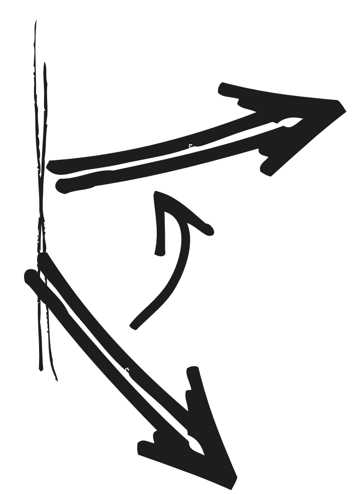
CIRCULAR DICHROISM

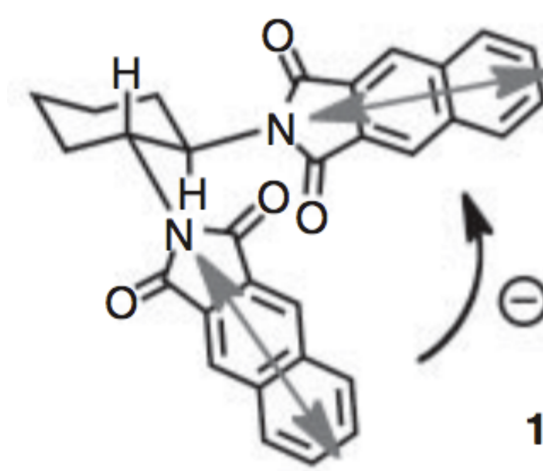






mimicking “*exciton coupling*”






THE T MATRIX
TWININGS
TWININGS
T 12
TWININGS
T 11
TWININGS
T 22
TWININGS
T 21

THE PROBLEM WITH SPHEROIDS

If we focus on a specific example, such as $n=5$, $k=1$, then the integral L_{51}^{1} takes the form

$$L_{51}^{1}=\int_{0}^{\pi}\mathrm{d}\theta\,\sin\theta\tau_5 d_1 x_\theta\left(\frac{\tilde{c}_0}{x^3}+\frac{\tilde{c}_1}{x}+\tilde{c}_2x+\tilde{c}_3x^3+\cdots\right). \quad (41)$$

For particles with a size parameter less than 1, the first few terms strongly dominate the integrand. However, as demonstrated numerically and analytically in Ref. [20], the x^{-3} term actually exactly integrates to zero in the special case of spheroids.

JQSRT 113 524–535 (2012)

SMARTIES https://github.com/nano-optics/smarties

SMARTIES

Table 3
Convergence study for an oblate spheroid with $s = 1.311 + 0i$. For each pair of size parameter $x_{\max}$ (columns) and aspect ratio h (rows), we list the convergence parameters N, N_θ, together with the relative error, only displayed if worse than 10^{-13}.

h:	$x_{\max}$:	0.01	0.1	1	2	3	4	5	6	7	8	10	12	15	20	25	30	40	50	60	70	80
	N	5	5	7	9	11	11	13	15	17	17	21	23	27	35	41	47	63	77	95	127	111
1.1	N_θ	6	6	6	7	7	8	8	9	9	10	15	15	15	20	25	35	40	40	80	80	1200
	Error	–	–	–	–	–	–	–	–	–	–	–	–	–	–	–	–	10^{-12}	10^{-11}	10^{-9}	10^{-7}	10^{-4}
	N	5	5	9	11	11	13	15	15	19	19	23	27	31	37	45	53	69	93	115		
1.3	N_θ	9	9	9	9	9	10	15	15	15	15	15	20	20	25	45	30	50	45	500		
	Error	–	–	–	–	–	–	–	–	–	–	–	–	–	–	–	10^{-12}	10^{-9}	10^{-7}	10^{-3}		
	N	5	7	11	13	15	15	19	19	21	23	25	27	33	41	49	61	75	103			
2	N_θ	20	20	20	20	20	20	20	20	25	25	25	25	30	35	45	35	50	45			
	Error	–	–	–	–	–	–	–	–	–	–	–	–	–	–	10^{-12}	10^{-11}	10^{-7}	10^{-4}			
	N	5	7	11	15	17	19	21	23	23	25	29	31	35	45	51	57	59				
4	N_θ	40	40	40	40	40	7	40	45	45	45	50	50	60	70	70	90	90				
	Error	–	–	–	–	–	10^{-3}	–	–	–	–	–	–	–	10^{-13}	10^{-11}	10^{-9}	10^{-4}				
	N	5	7	11	15	19	19	19	23	25	27	31	35	37	41	47	47					
7	N_θ	70	70	70	70	70	70	80	80	80	80	90	90	100	200	130	130					
	Error	–	–	–	–	–	–	–	–	–	–	–	–	–	10^{-11}	10^{-10}	10^{-6}					
	N	5	7	13	15	19	19	21	25	27	27	31	33	41	45	43	47					
10	N_θ	100	100	90	100	100	100	110	110	110	120	120	130	140	180	180	200					
	Error	–	–	–	–	–	–	–	–	–	–	–	–	–	10^{-11}	10^{-8}	10^{-6}					
	N	5	7	13	15	19	21	23	23	27	29	33	37	43	43	49	41					
20	N_θ	200	200	200	200	200	200	200	220	220	240	260	260	280	300	550	400					
	Error	–	–	–	–	–	–	–	–	–	–	–	–	–	10^{-12}	10^{-8}	10^{-4}					
	N	5	7	13	17	19	21	23	25	27	29	33	35	39	45	47	45					
50	N_θ	500	500	500	500	500	500	550	550	550	600	650	800	700	1100	900	1200					
	Error	–	–	–	–	–	–	–	–	–	–	–	–	10^{-13}	10^{-12}	10^{-9}	10^{-6}					
	N	5	7	13	17	19	21	23	25	27	27	33	35	47	53	47						
100	N_θ	1000	1100	1000	1100	1000	1100	1100	1100	1100	1100	1300	1500	1400	1600	2000						
	Error	–	–	–	–	–	–	–	–	–	–	–	–	–	–	10^{-9}						

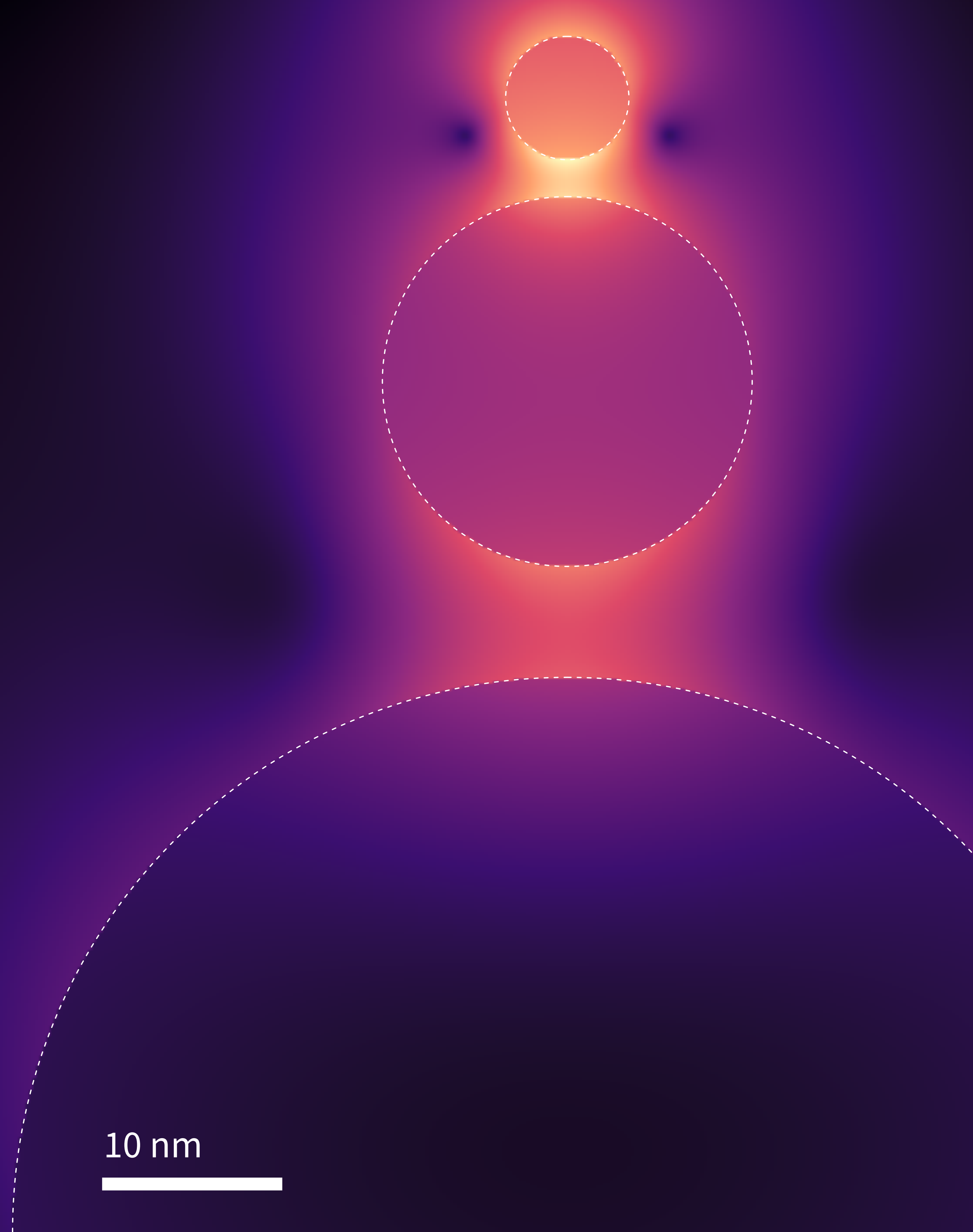
LONG WAVELENGTH APPROXIMATION

Taylor expansion of *T* (x^3) • Phys. Rev. A 99, 013853 (2019)

$$\mathbf{T}_{m=0} = \left[\begin{array}{cc|ccc} \frac{iK_{11}^{11}}{1-iK_{11}^{11}} & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 & 0 \\ \hline 0 & 0 & \frac{iK_0X^3}{1-\Omega_0X^2-iK_0X^3} & 0 & \frac{\sqrt{14}}{175}\frac{ie^2K_0X^5}{1-iK_0X^3} \\ 0 & 0 & 0 & \frac{iK_{22}^{22}}{1-iK_{22}^{22}} & 0 \\ 0 & 0 & \frac{\sqrt{14}}{175}\frac{ie^2K_0X^5}{1-iK_0X^3} & 0 & 0 \end{array}\right] + \mathcal{O}(X^7)$$

Generalised radiative correction • Phys. Rev. A 87, 012504 (2013)

$$(\boldsymbol{\alpha}^{\mathrm{RC}})^{-1} = (\boldsymbol{\alpha}_0)^{-1} - i\,\frac{k_1^3}{6\pi\,\epsilon_0\epsilon_1}\mathbf{I},$$



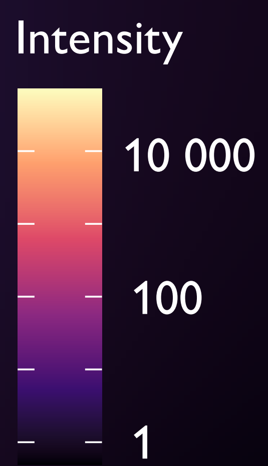
Dmitri
Schebarchov

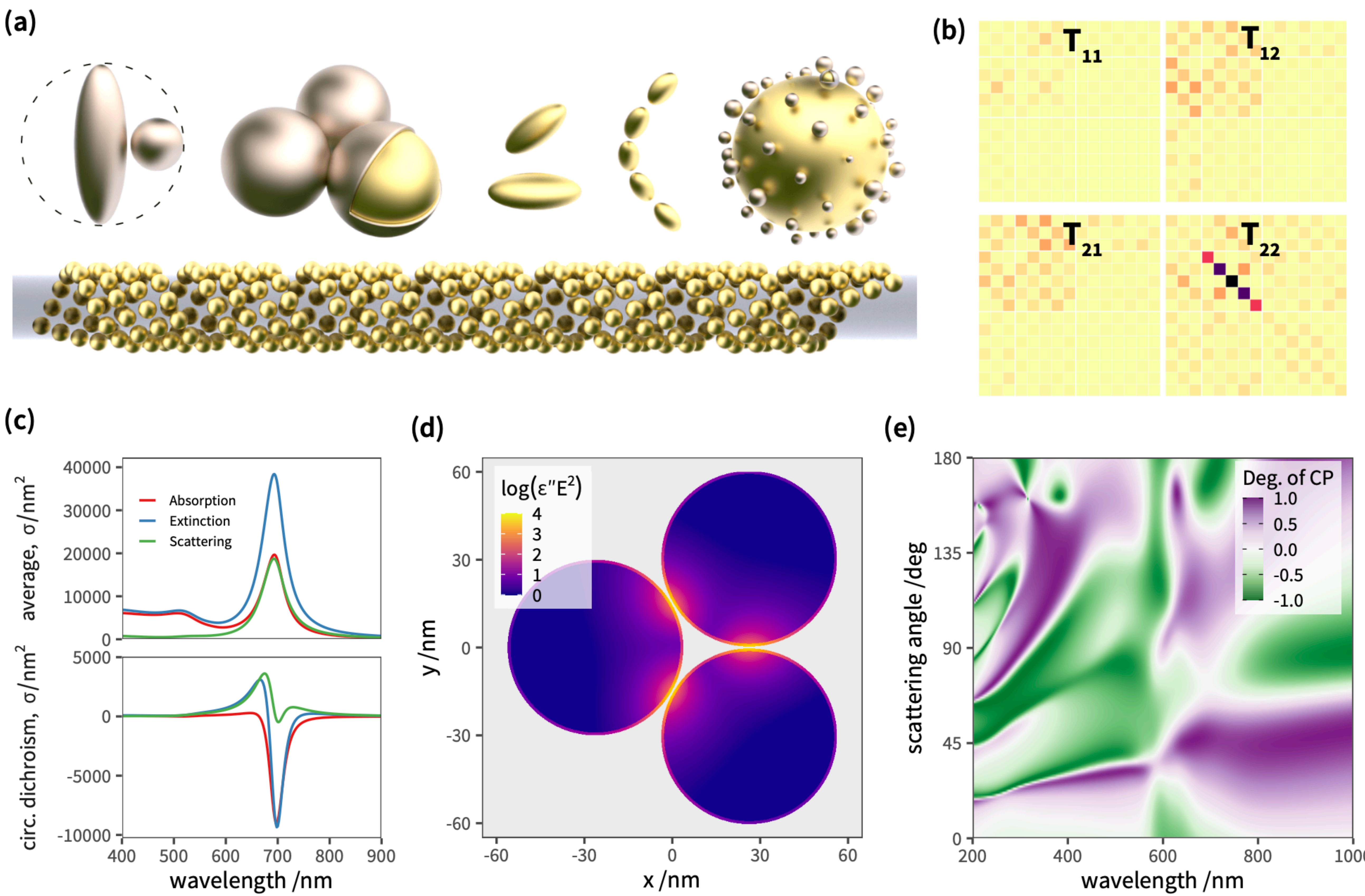
Atefeh
Fazel-Najafabadi

T.E.R.M.S.

(SUPERPOSITION T-MATRIX)

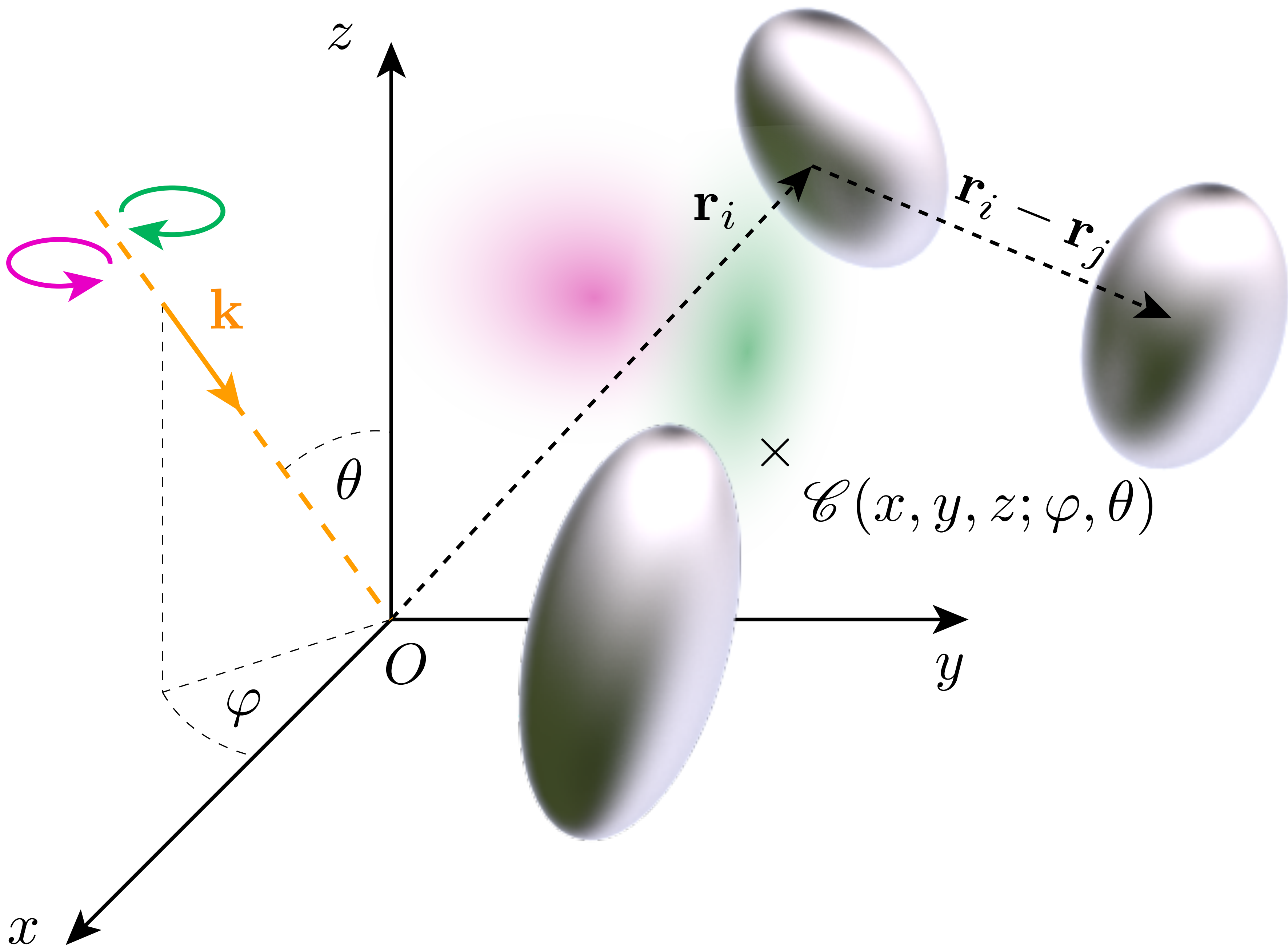
nano-optics.ac.nz/terms





nano-optics.ac.nz/terms

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



Phys. Rev. B 103, 115405 (2021)

ANALYTICAL AVERAGING, BUT…

Be careful what you wish for?

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

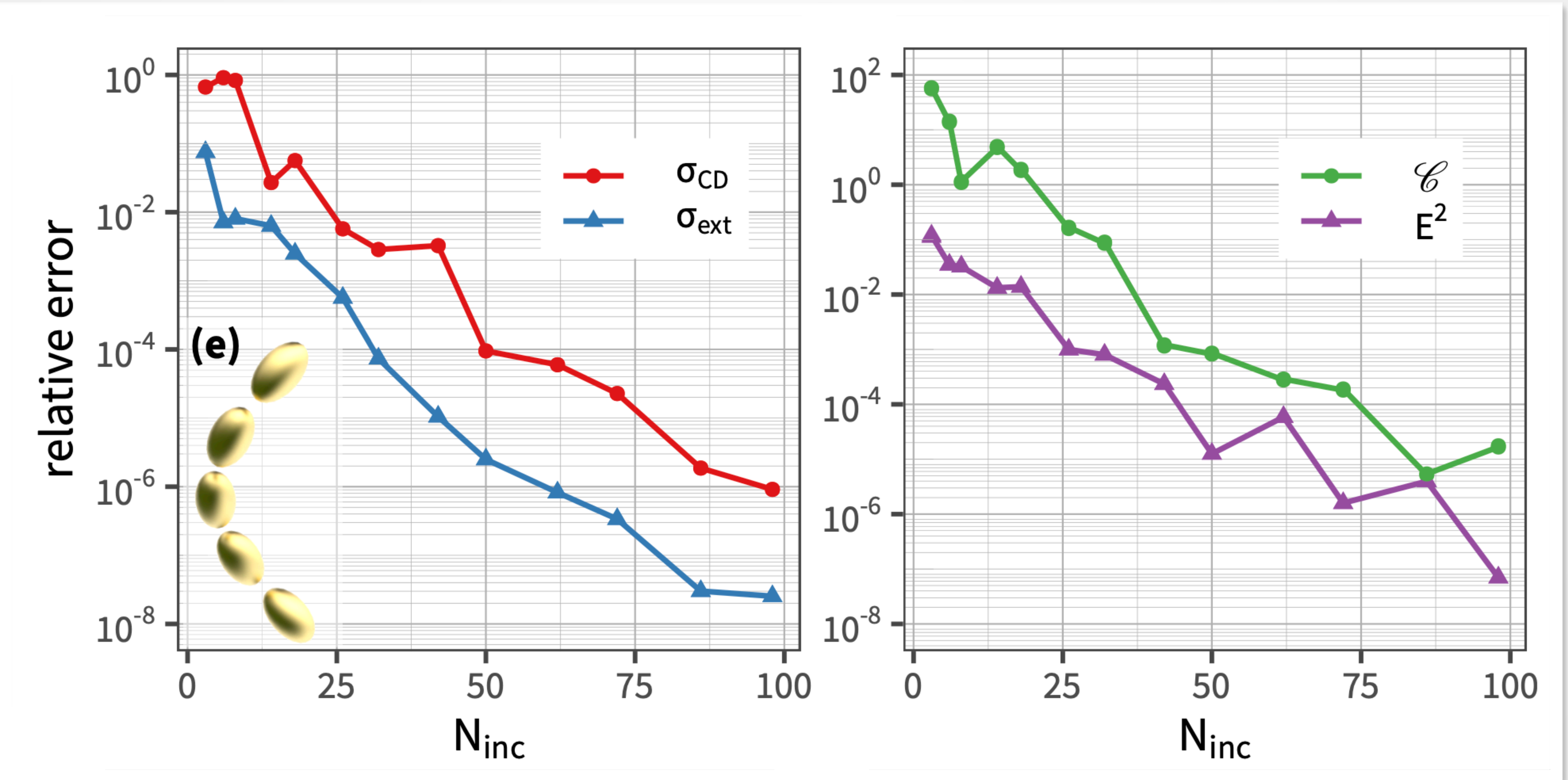
$$A_0^{(\mathrm{R})} = -1/4\pi,$$

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

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

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

ORIENTATION AVERAGING BY SPHERICAL CUBATURE

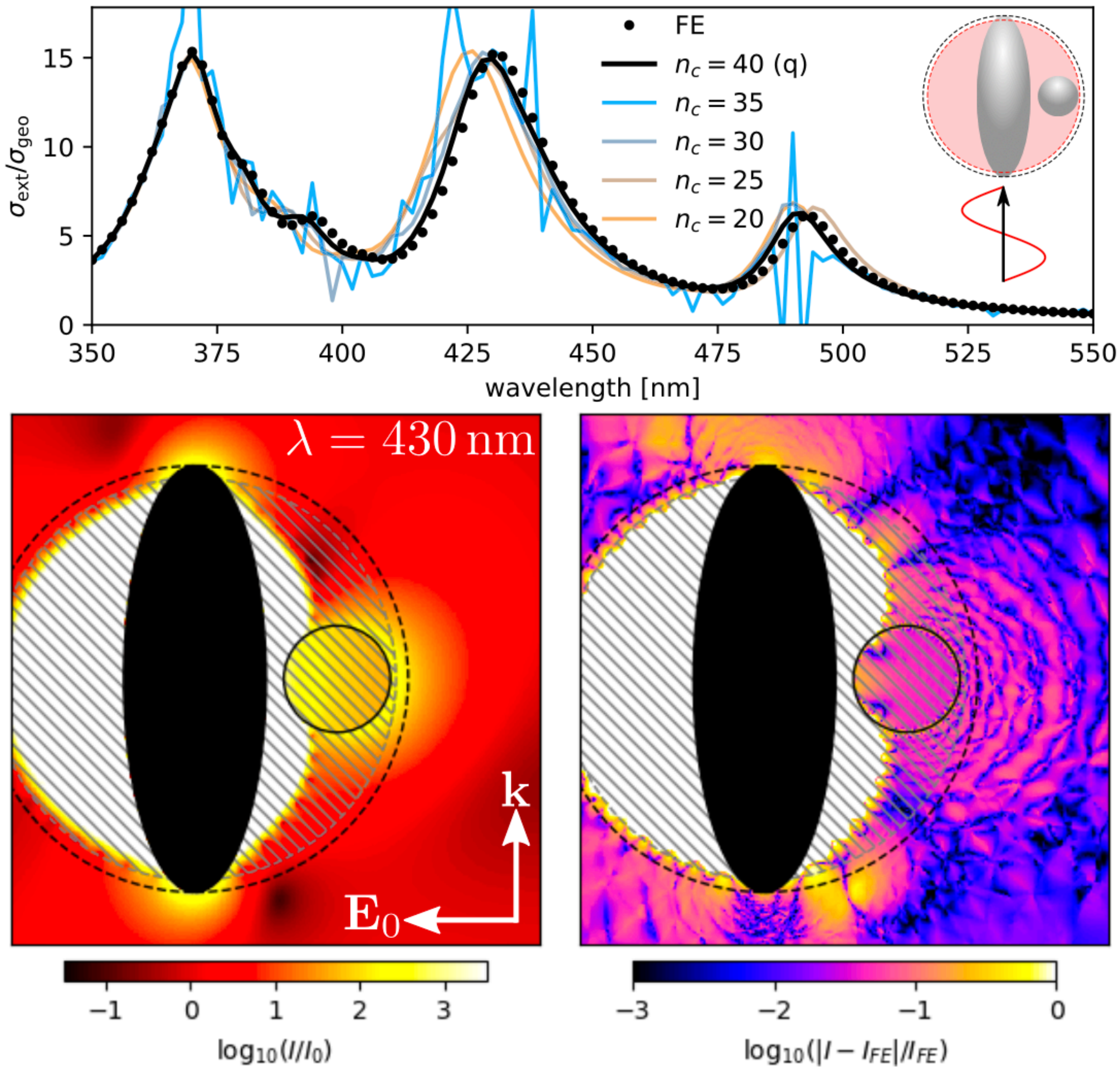


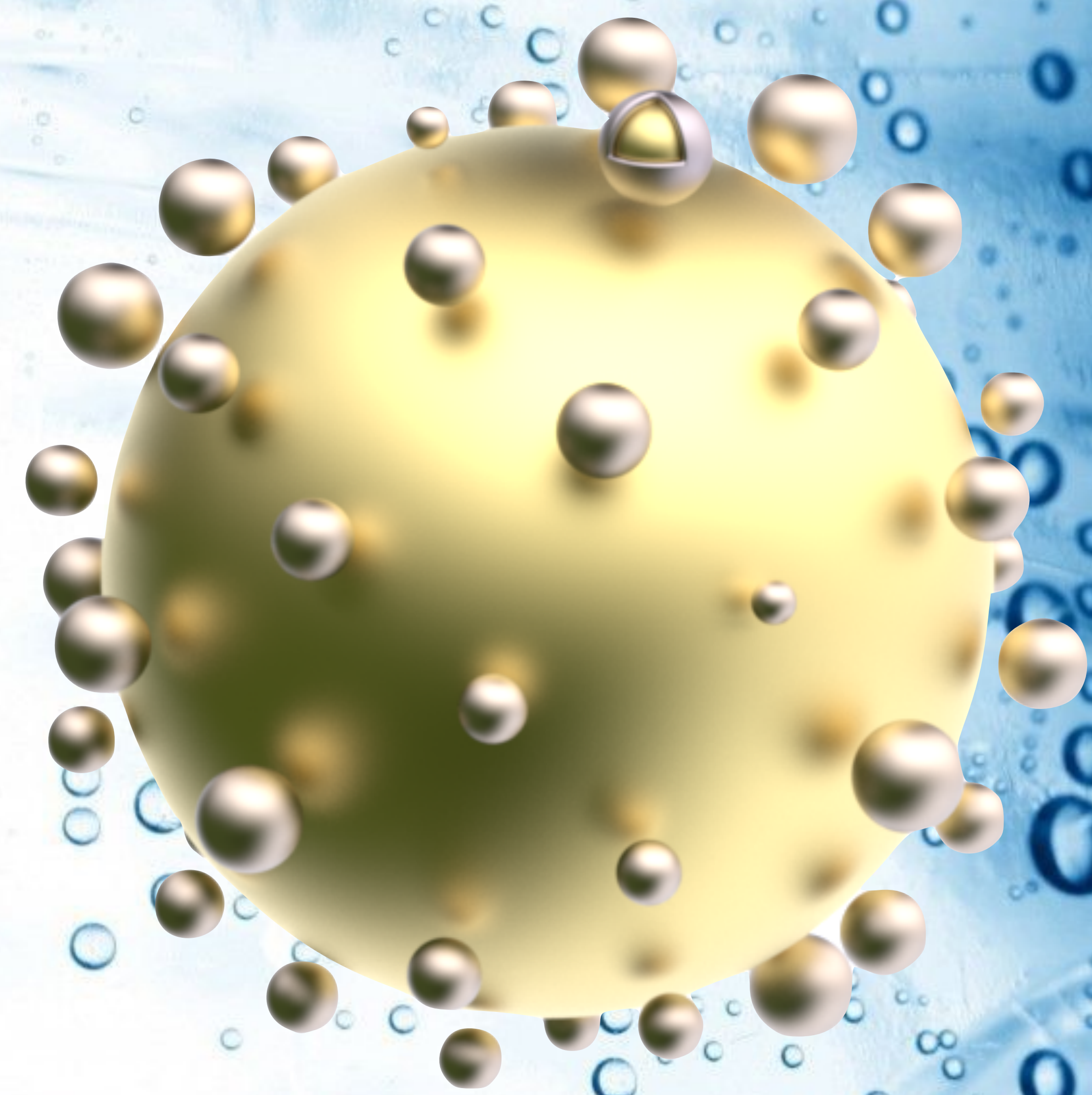
Mater. Adv. 3, 1547–1555 (2022)

JQSRT 286, 108197 (2022)

RAYLEIGH HYPOTHESIS

Opt. Exp. 27 35751 (2019)





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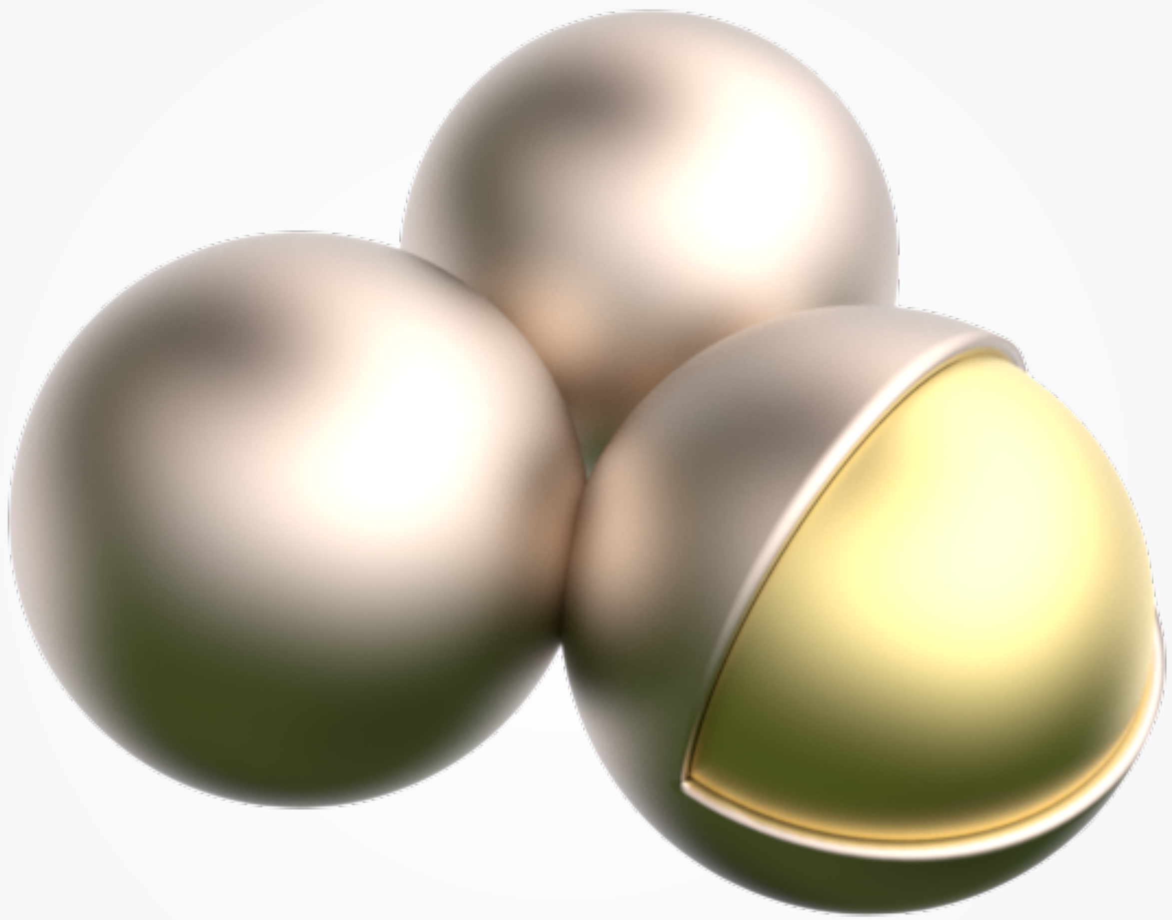
[1] Nanoinstitute Munich, Ludwig-Maximilians-Universitat, Germany

[2] Centro de Investigaciones Biomédicas, Universidade de Vigo, Spain

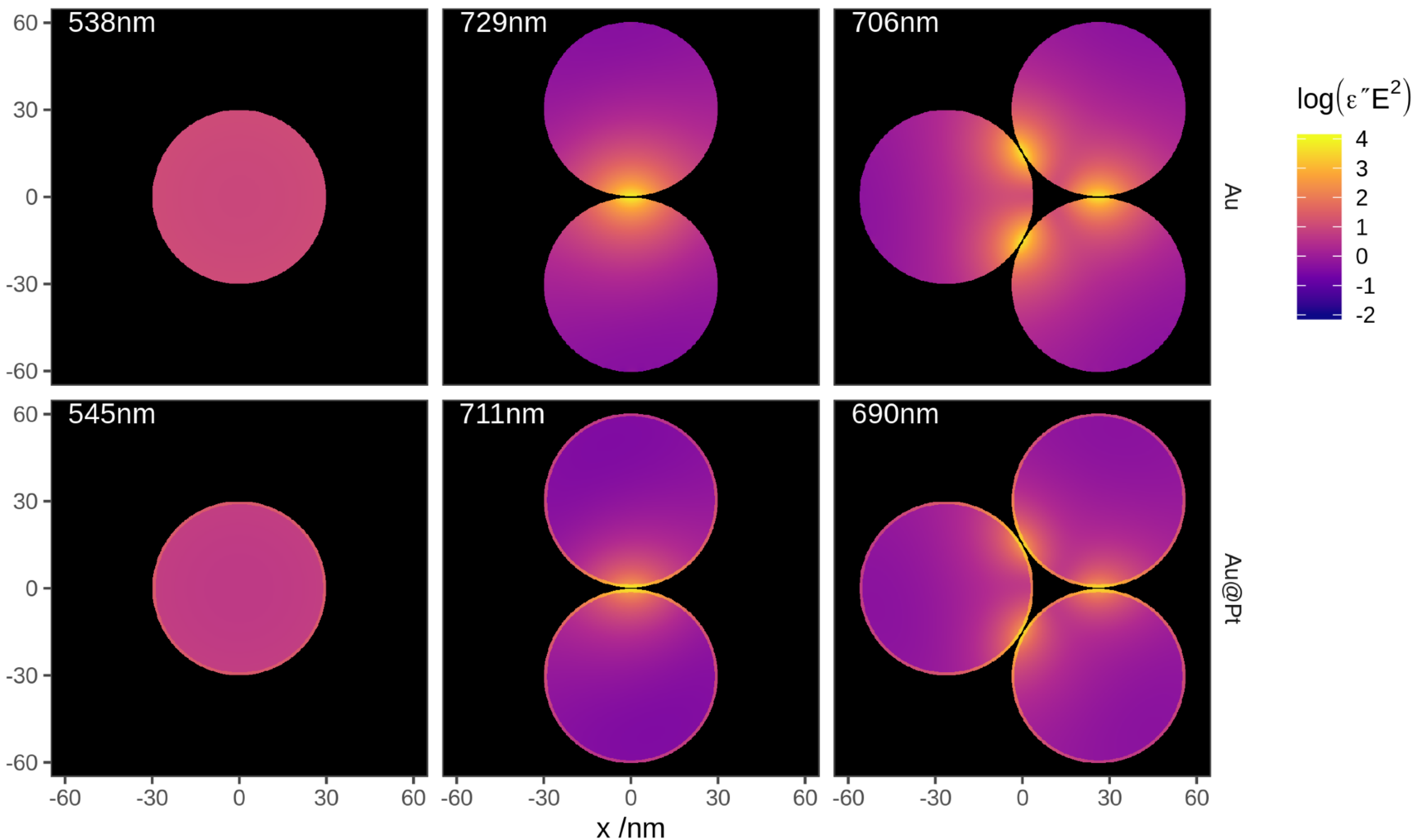
[3] School of Chemical & Physical Sciences, Victoria University of Wellington, Aotearoa NZ

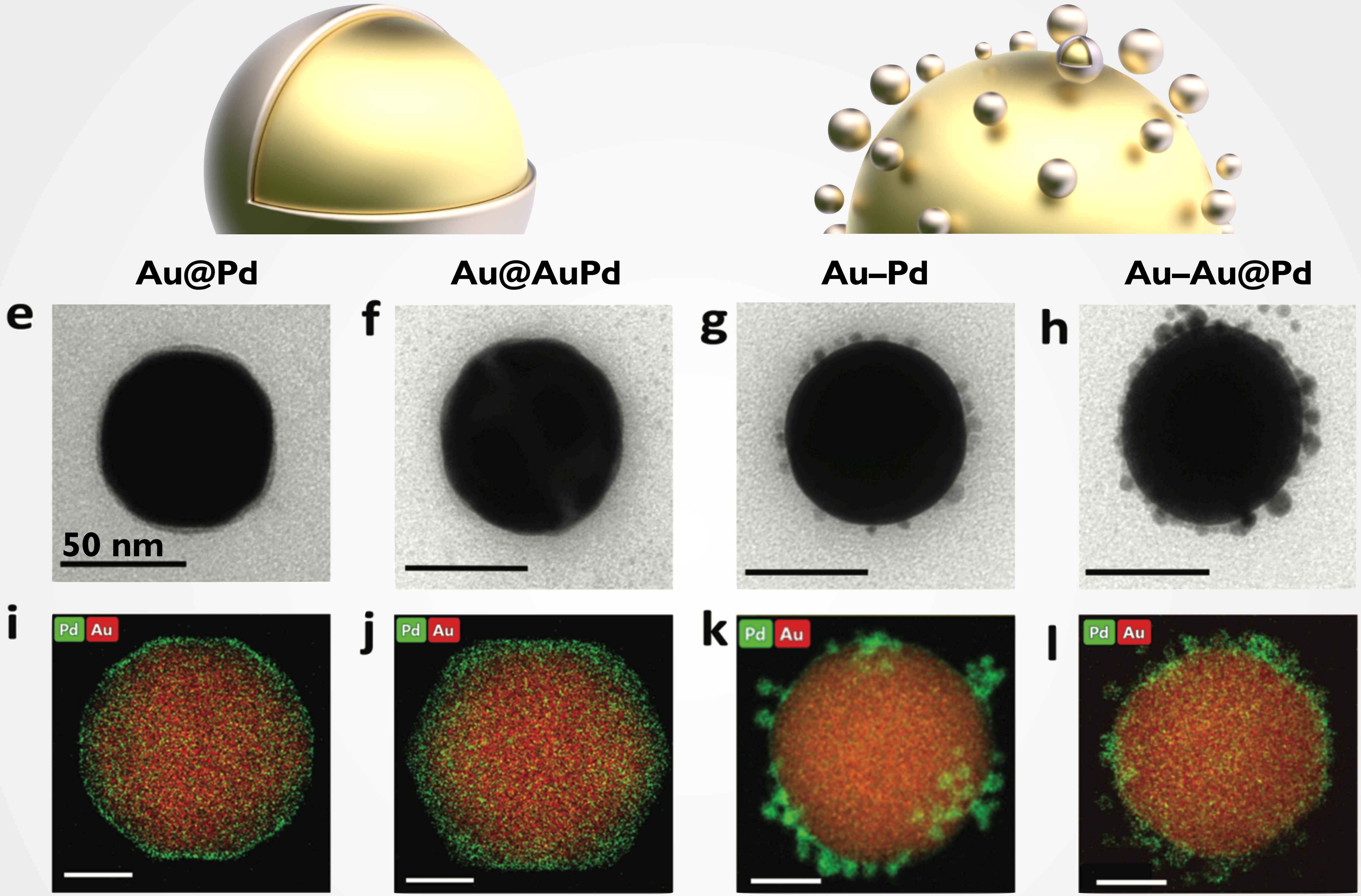
ABSORPTION OF AU@PD PHOTOCATALYSTS



ACS Energy Lett. 5, 12, 3881–3890 (2020)

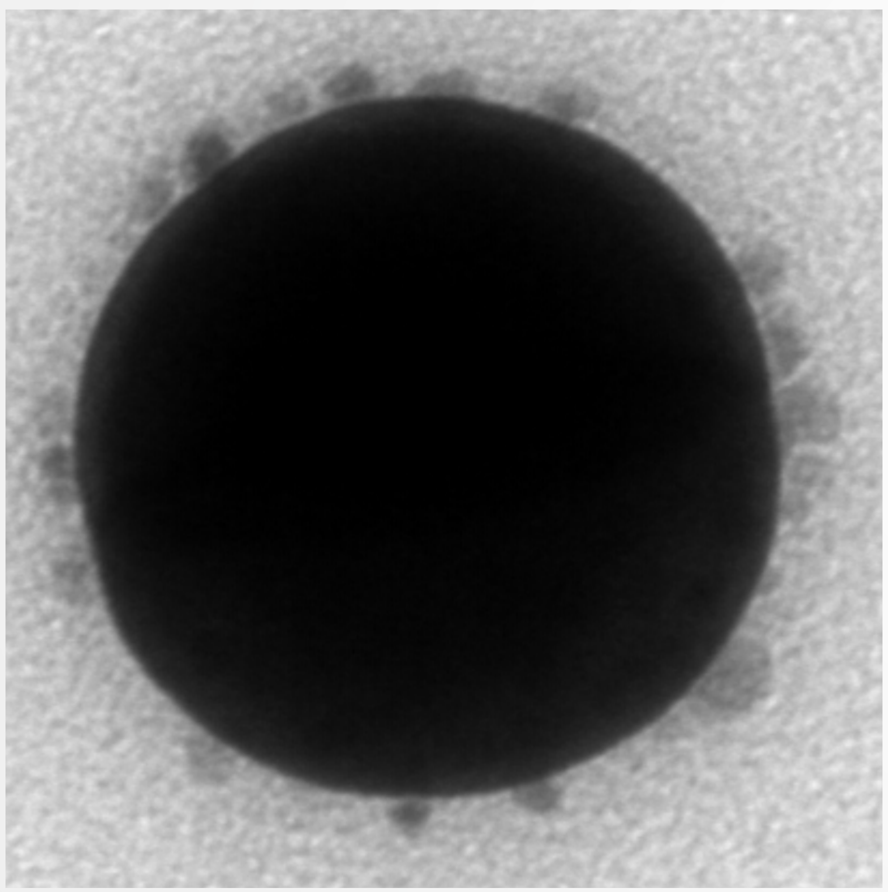
LOCAL ABSORPTION IN AU@PD NANO-TRIMERS



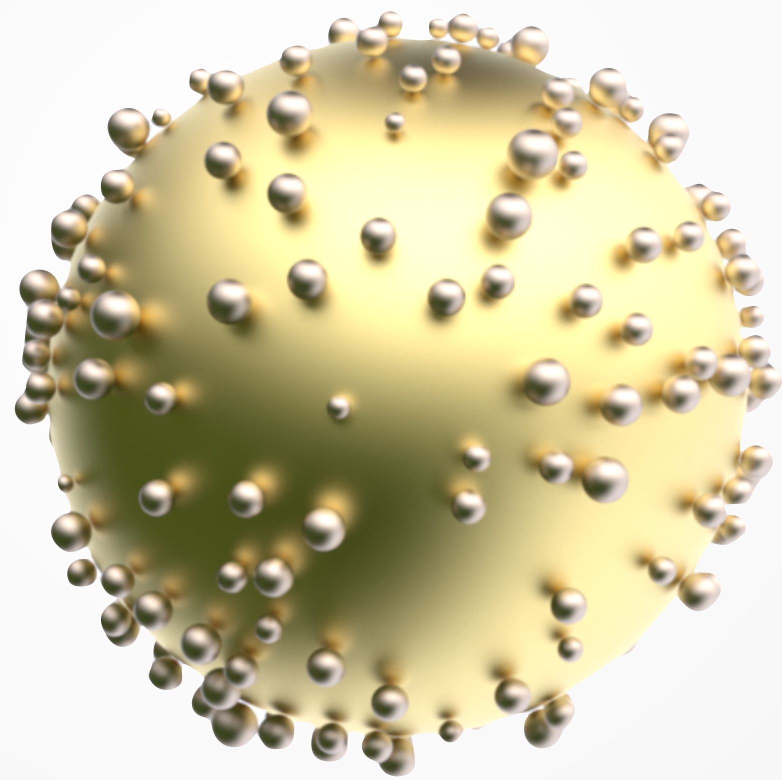


M. Herran, A. Sousa-Castillo, C. Fan, S. Lee, Wei Xie, M. Döblinger, B. Auguié and E. Cortés · *Adv. Func. Mat.* 32, 2203418 (2022)

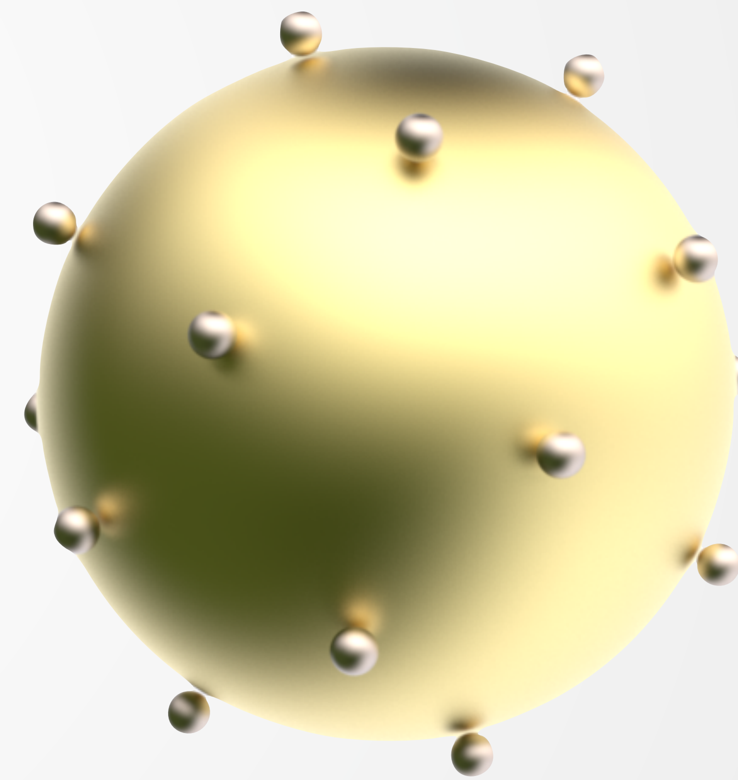
modelling Au–Pd core-satellite structures



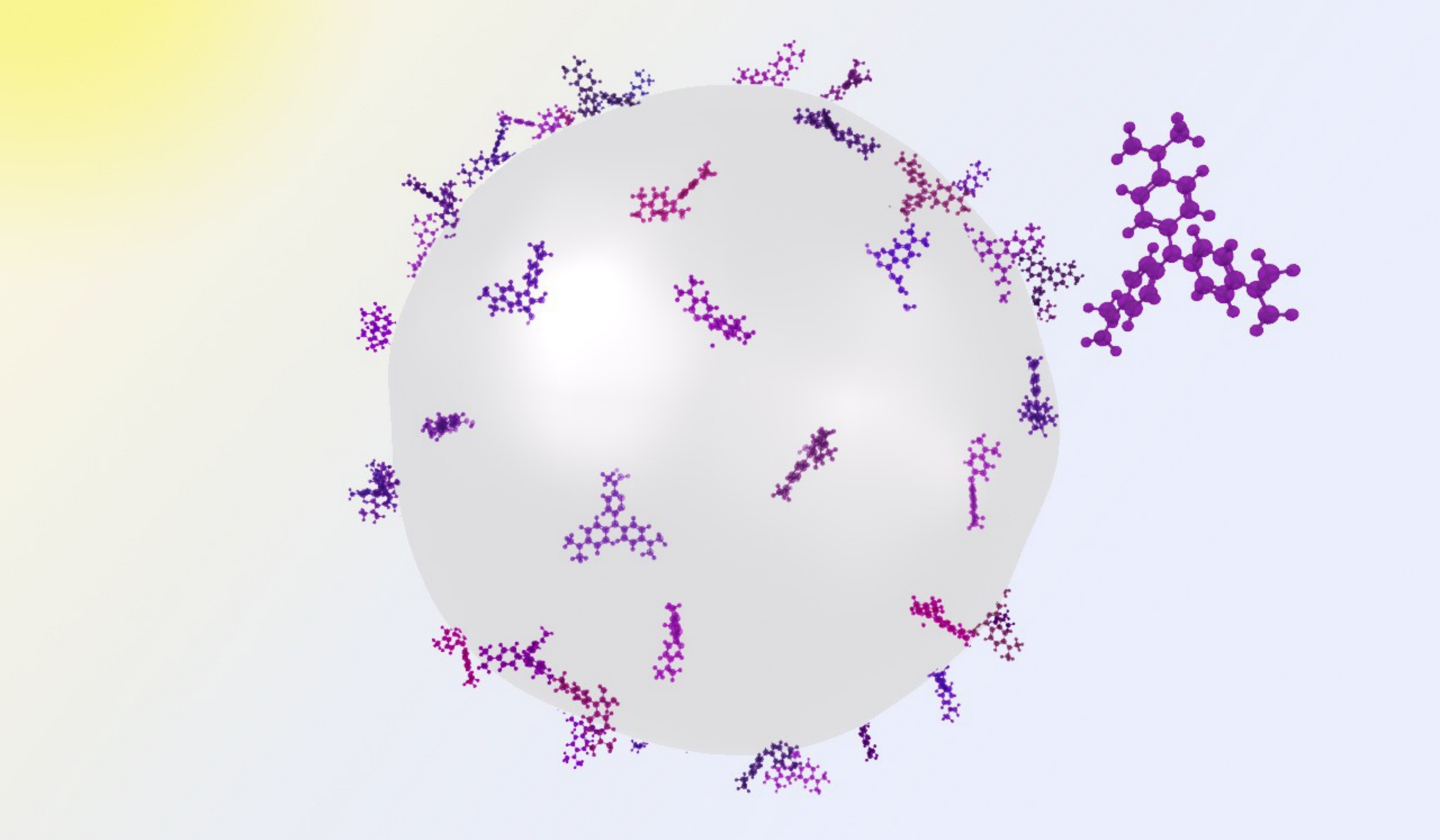
TEM



"model"

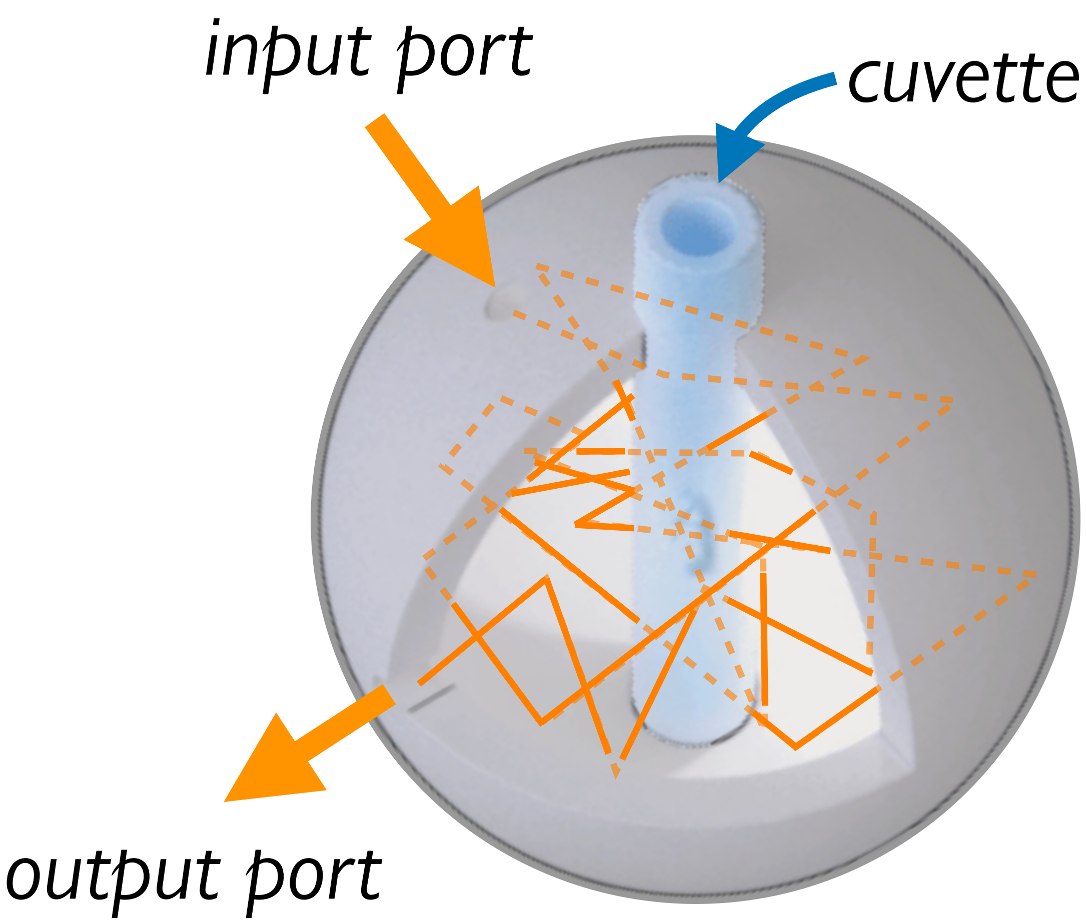


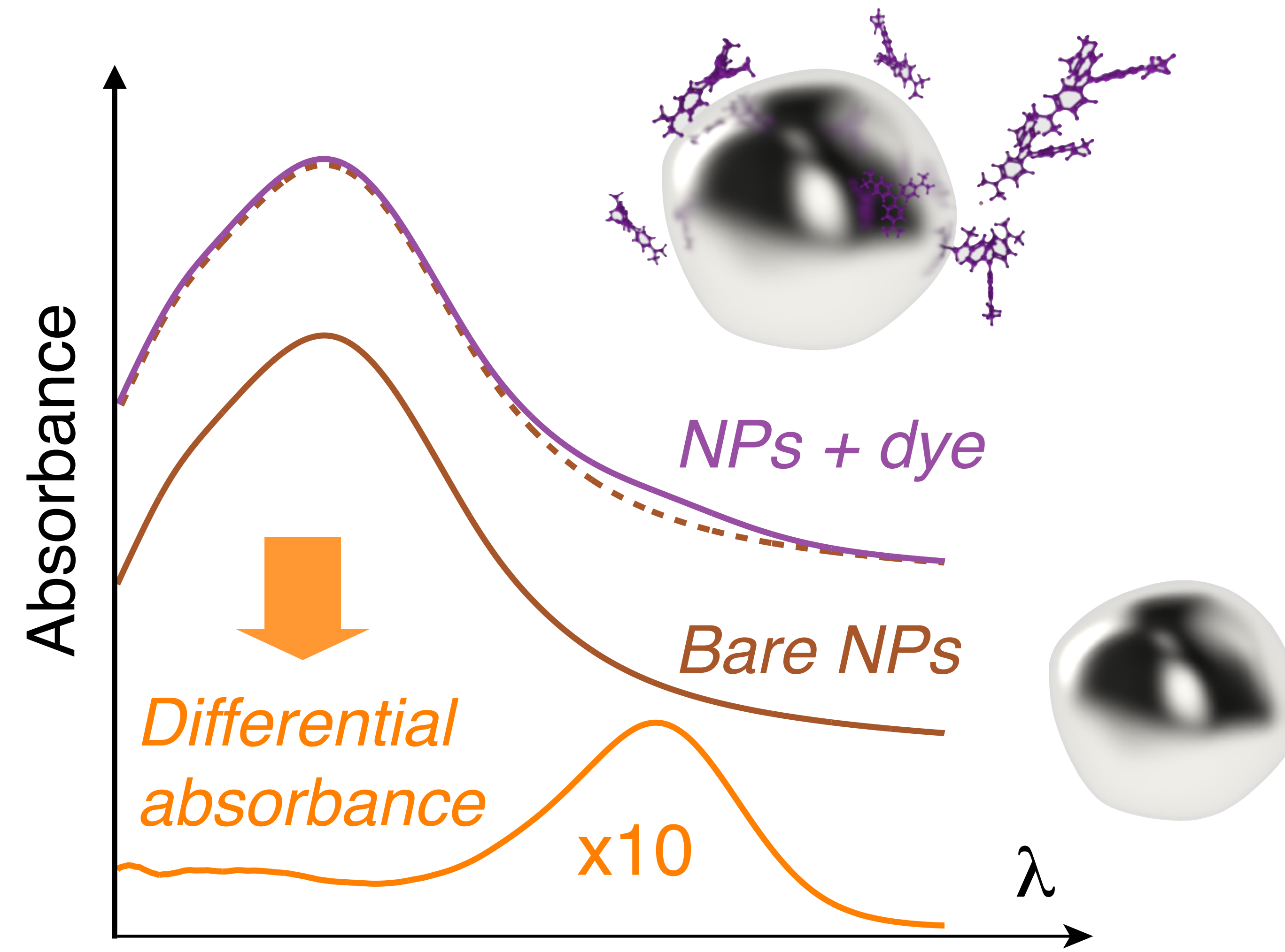
N=20



ELECTROMAGNETIC INTERACTIONS BETWEEN *MOLECULES* SURROUNDING A METALLIC SPHERE

EXPERIMENTAL BACKGROUND:
UV–VIS SPECTROSCOPY OF TURBID SAMPLES





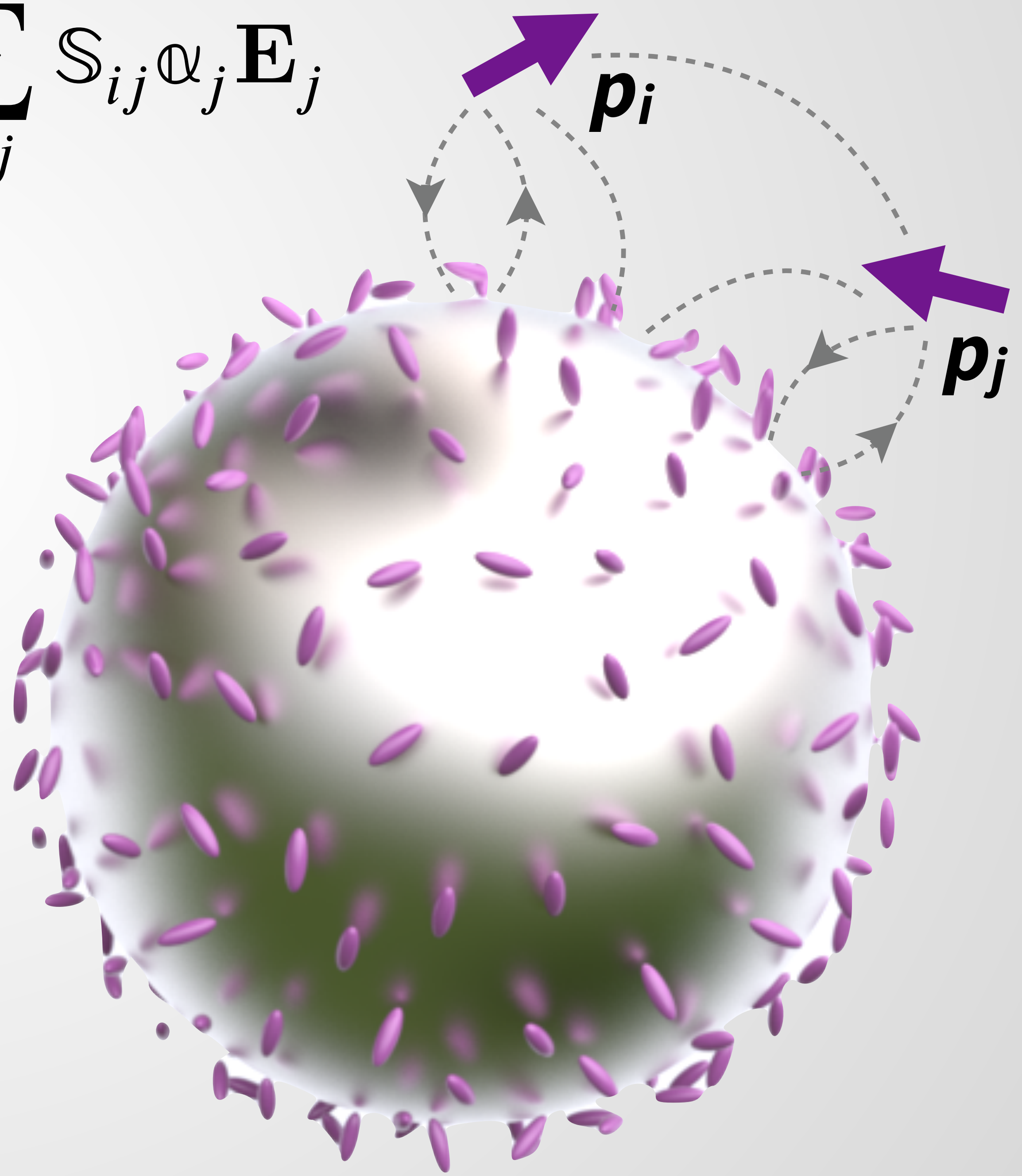
Modified optical absorption of molecules on metallic nanoparticles at sub-monolayer coverage – Nat. Photon. 10, 40–45 (2016)

MODIFIED COUPLED-DIPOLE EQUATIONS

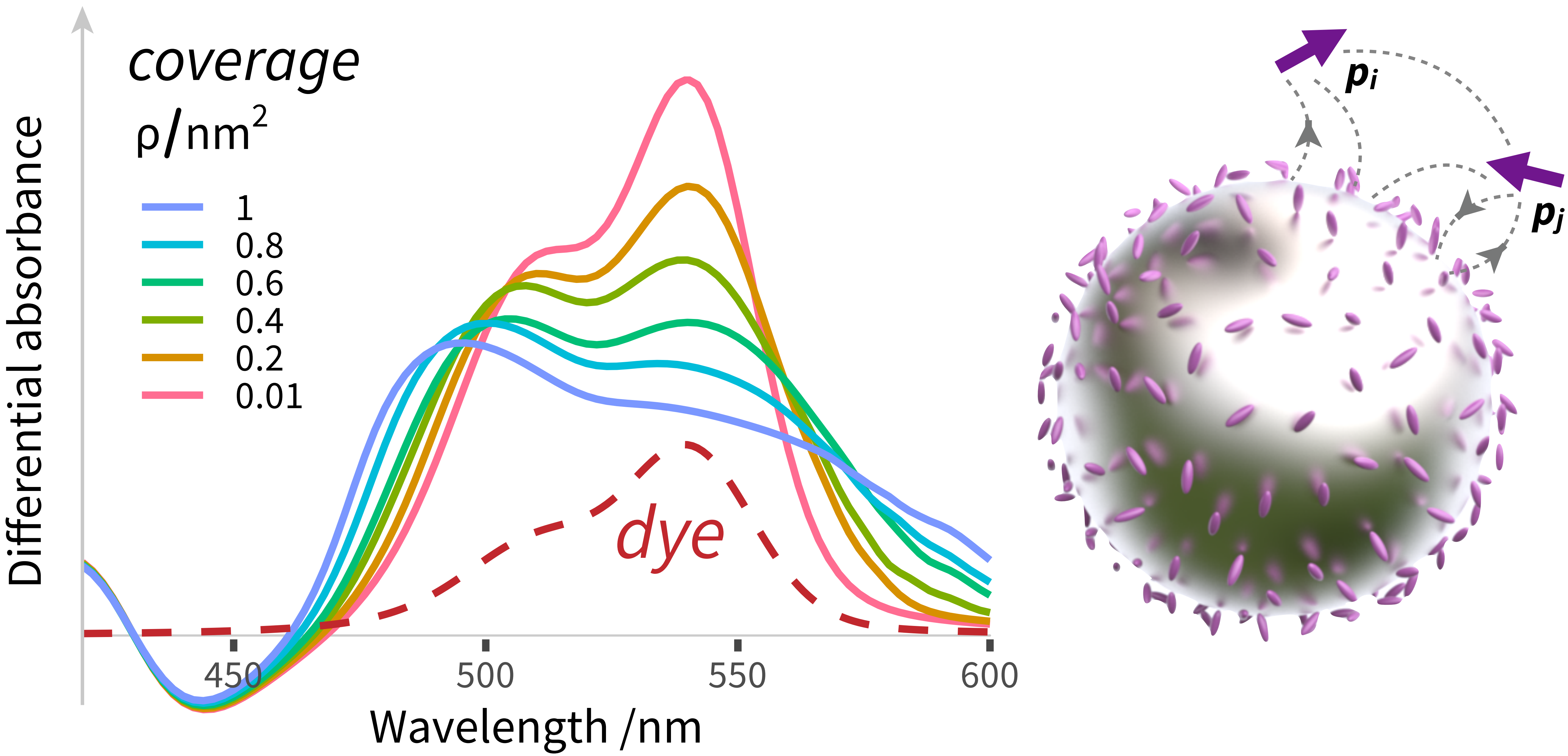
$$\mathbf{E}_i = \mathbf{E}_i^{\mathsf{INC}} + \mathbf{E}_i^{\mathsf{SPH}} + \sum_{j\neq i} \mathbb{G}_{ij}\alpha_j\mathbf{E}_j + \sum_{\forall j} \mathbb{S}_{ij}\alpha_j\mathbf{E}_j$$

$$\mathbb{A}\mathbf{E} = \mathbf{E}^{\mathsf{INC}} + \mathbf{E}^{\mathsf{SPH}}$$

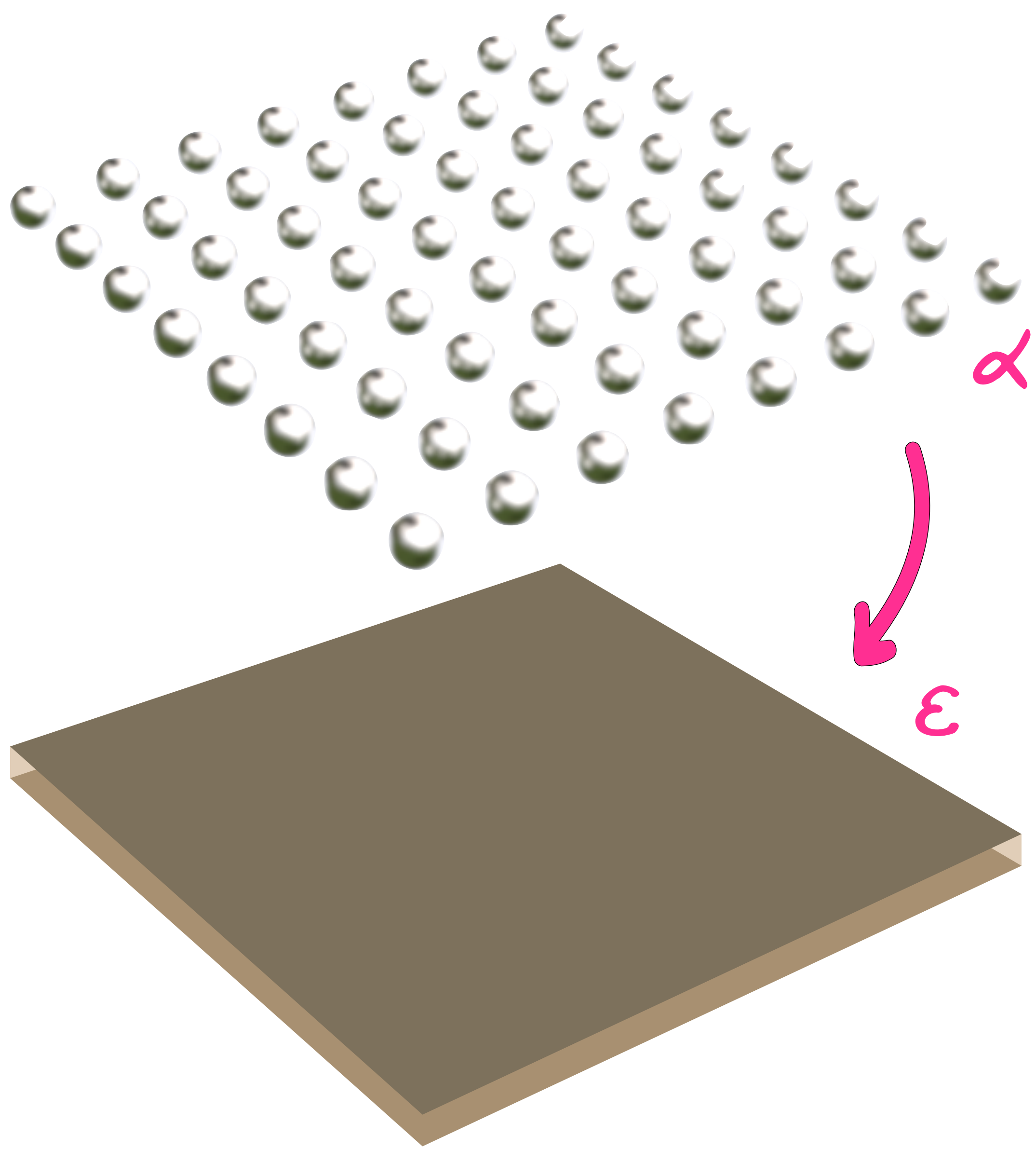
- Sphere-mediated coupling $\boldsymbol{S}_{ij}$
- Self-reaction ("image" dipole) $\boldsymbol{S}_{ii}$
- Additional excitation from sphere-scattered field $\boldsymbol{E}^{SPH}$



COUPLED DIPOLES AROUND A NANOSPHERE



FURTHER STEP: ANISOTROPIC EFFECTIVE MEDIUM

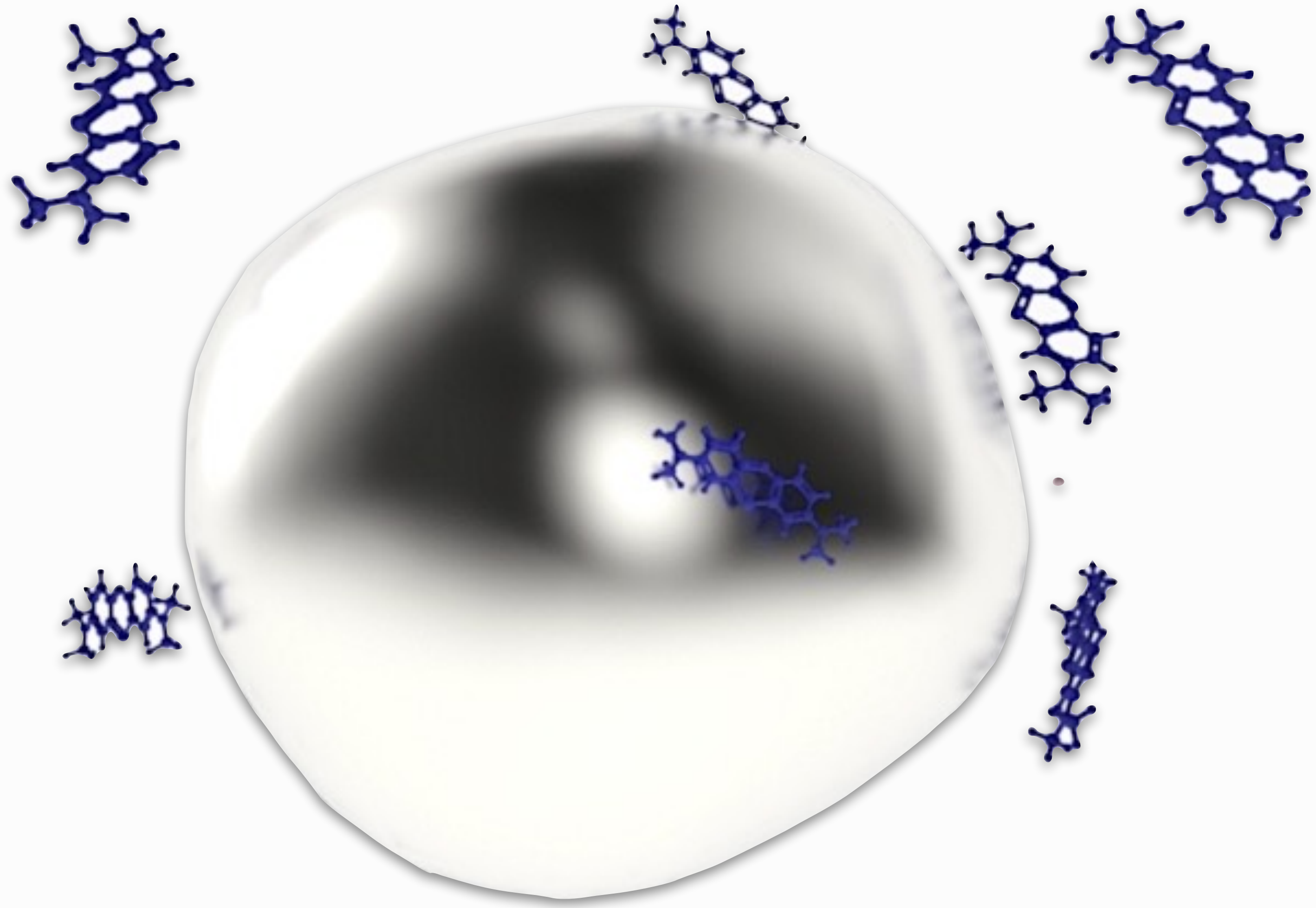


$$\alpha = a^3 \frac{\varepsilon - \varepsilon_m}{\varepsilon + 2\varepsilon_m} \begin{pmatrix} 1 & 0 & 0 \\ 0 & 1 & 0 \\ 0 & 0 & 1 \end{pmatrix}$$

$$\varepsilon_{xy} = \varepsilon_m + \frac{L_m^2 \frac{c_d \alpha}{\varepsilon_0}}{1 - \frac{\alpha}{8\pi\varepsilon_0} \frac{L_m^2}{\varepsilon_m} \rho^{3/2} (\xi_0 - \beta_m \xi_I)}$$

$$\frac{1}{\varepsilon_z} = \frac{1}{\varepsilon_m} - \frac{\frac{L_m^2}{\varepsilon_m^2} \frac{c_d \alpha}{\varepsilon_0}}{1 + \frac{\alpha}{4\pi\varepsilon_0} \frac{L_m^2}{\varepsilon_m} \rho^{3/2} (\xi_0 + \beta_m \xi_I)}$$

ANISOTROPIC EFFECTIVE MEDIUM FOR DYE SHELLS



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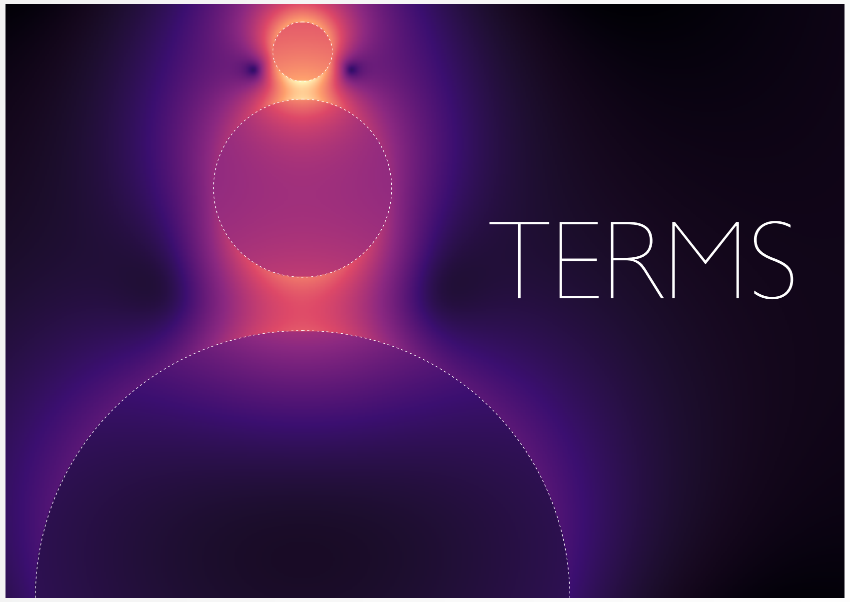
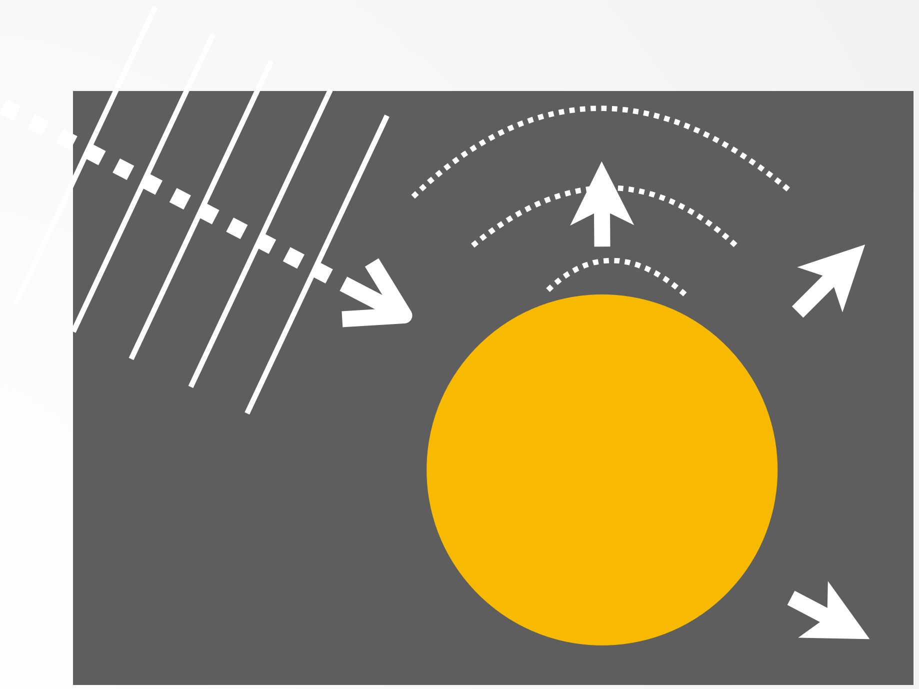
· *Phys. Rev. A* 104, 033502 (2021)

· *J. Phys. Chem. C*, 126, 24 (2022)

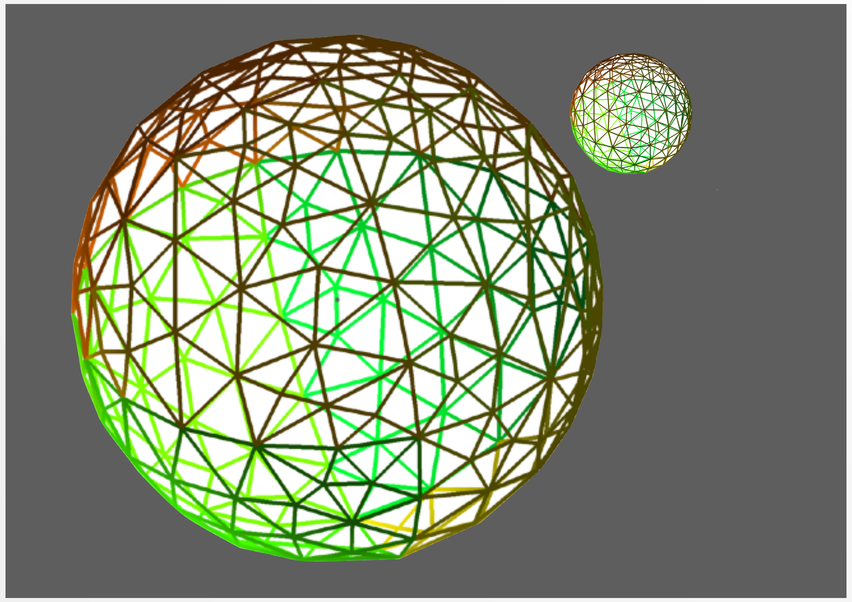
COMPARISON OF SIMULATION METHODS



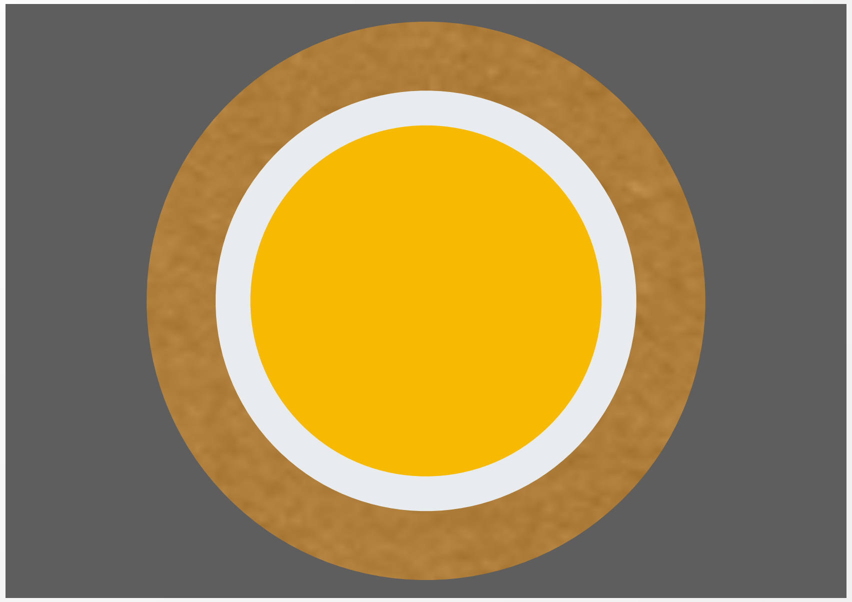
Superposition T-matrix



Generalised coupled-dipole

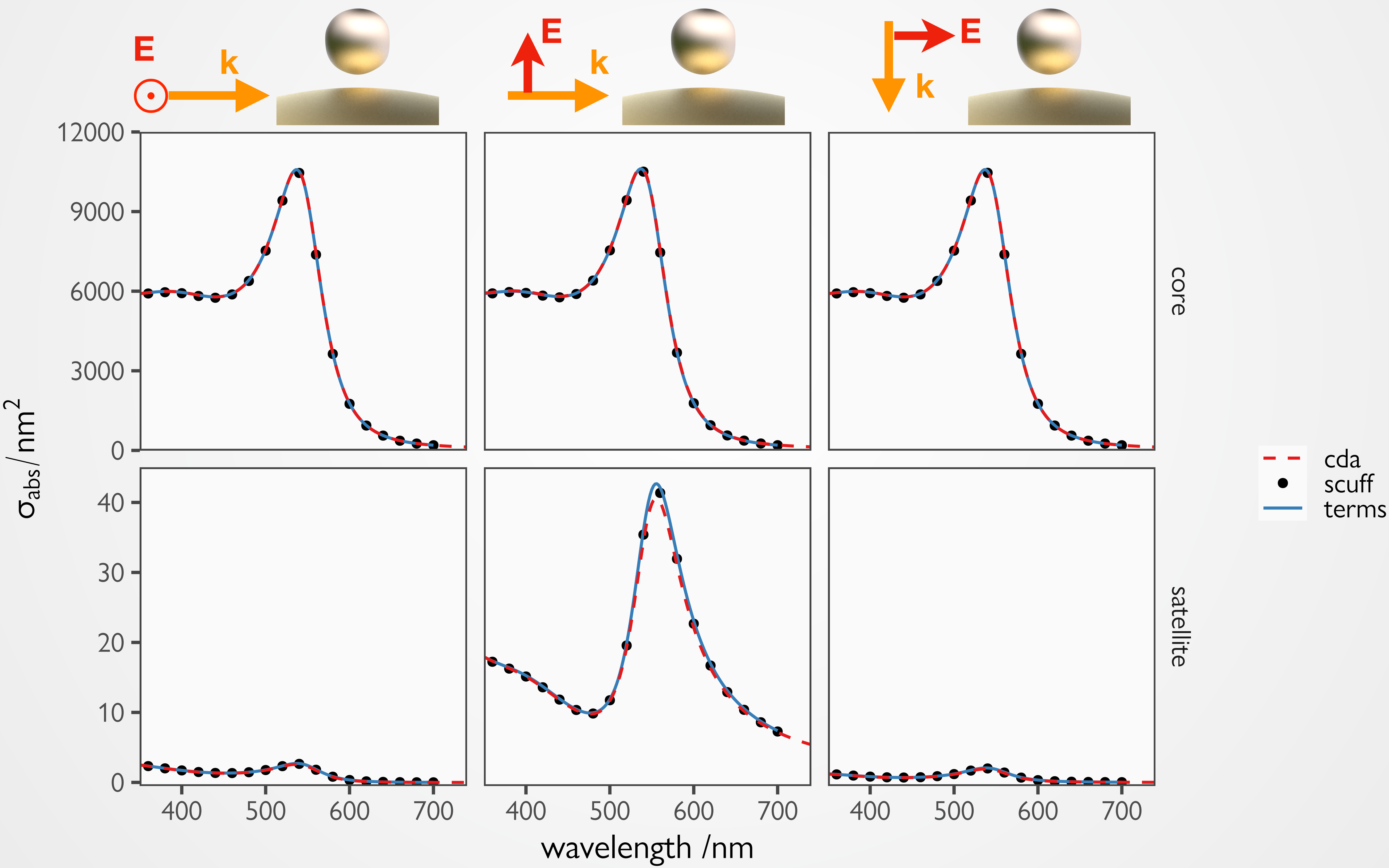


Boundary element method

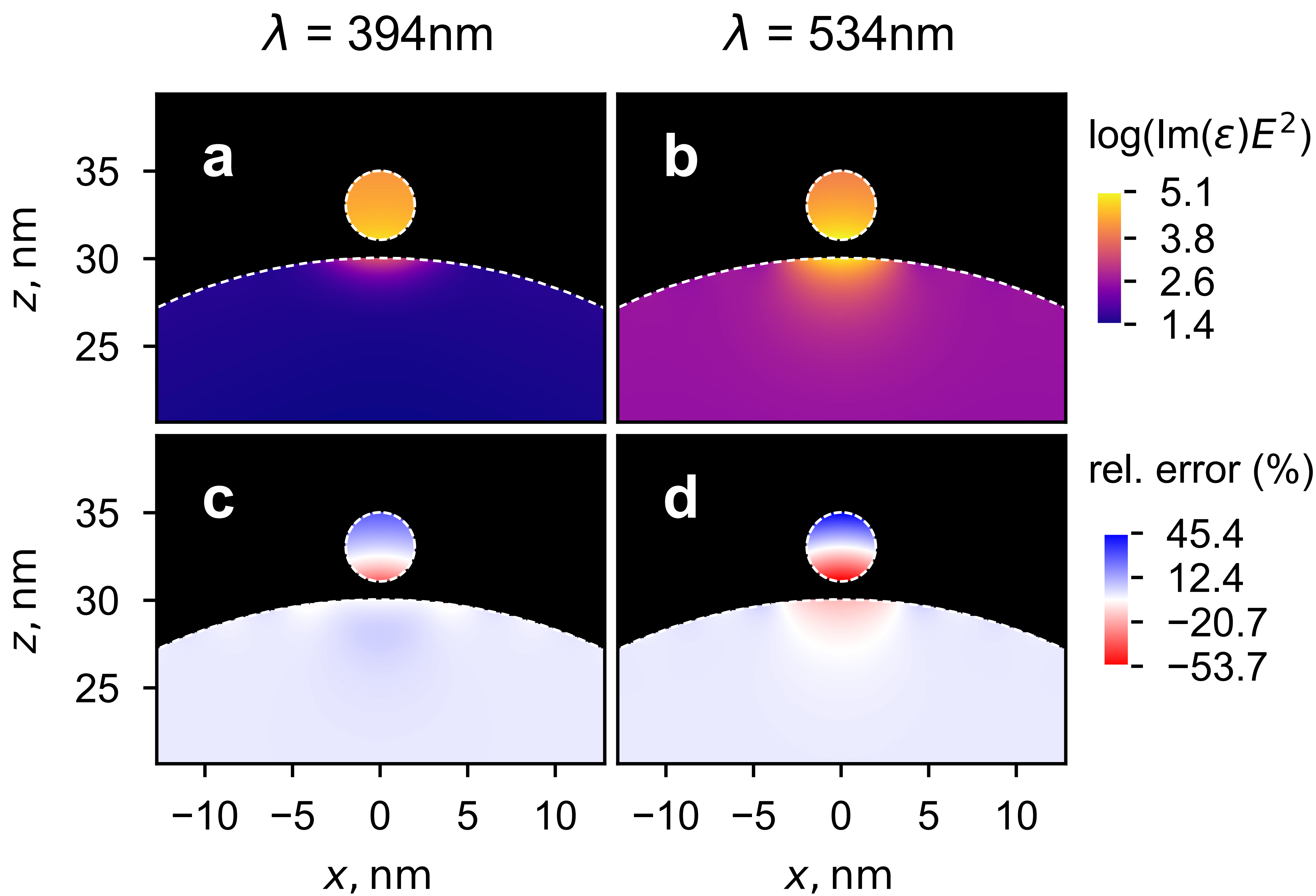


Effective medium + Mie

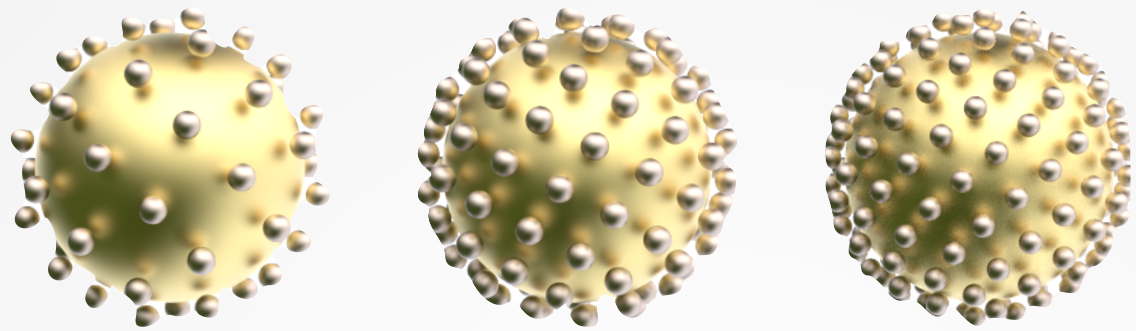
VALIDATION: FIXED INCIDENCE, SINGLE SATELLITE

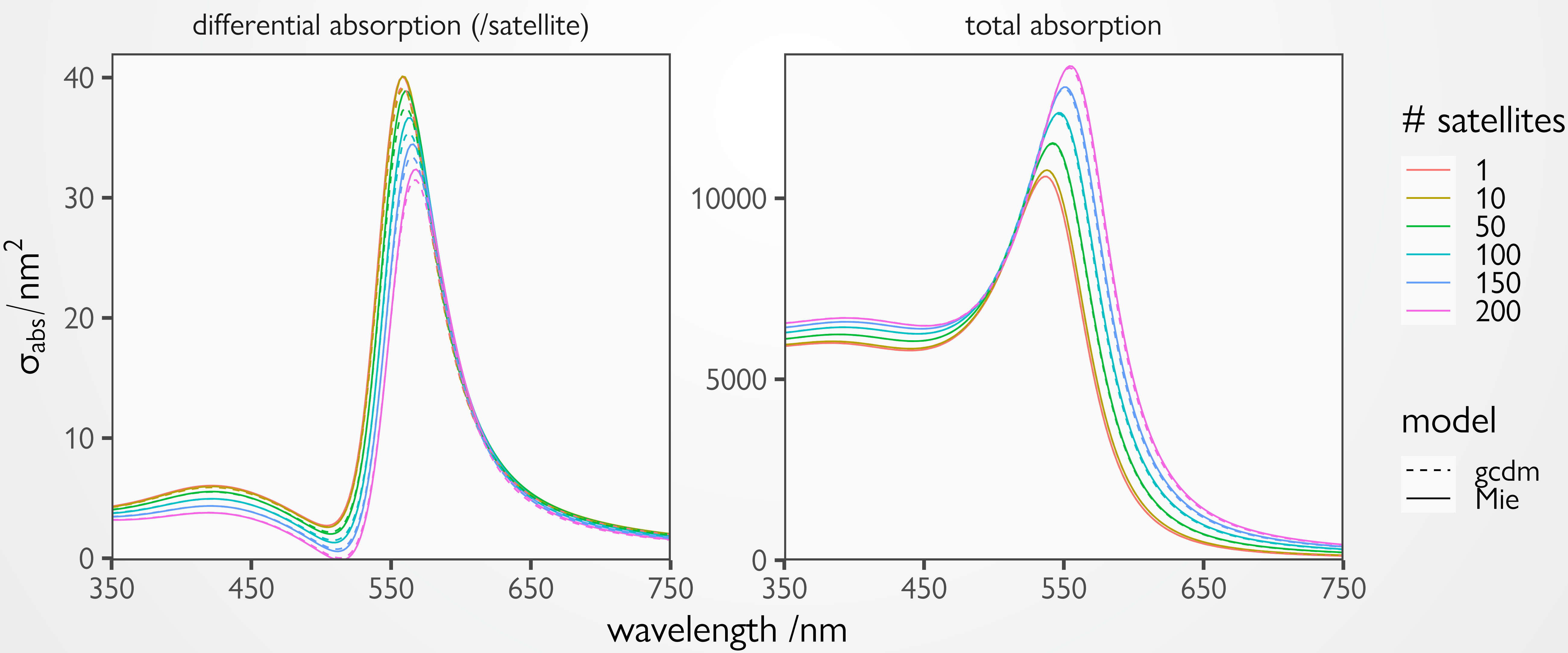


DIPOLE APPROXIMATION VS EXACT RESULTS

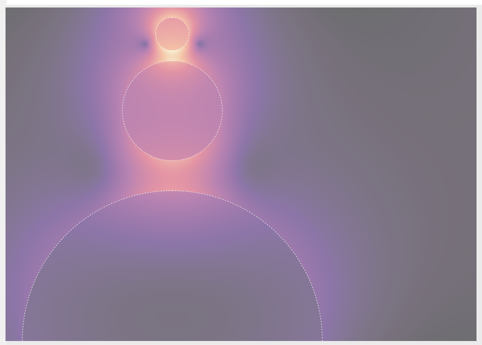
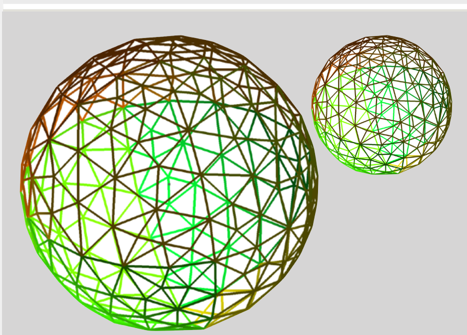
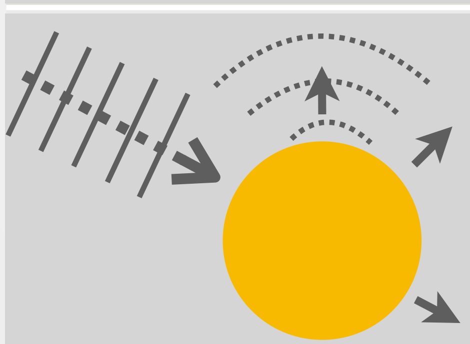
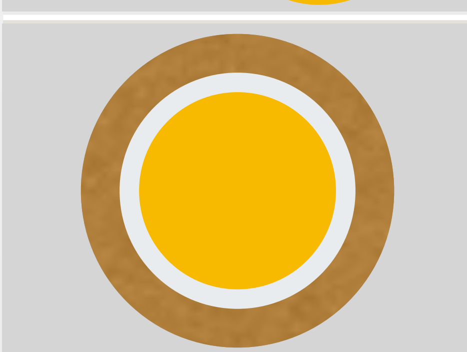


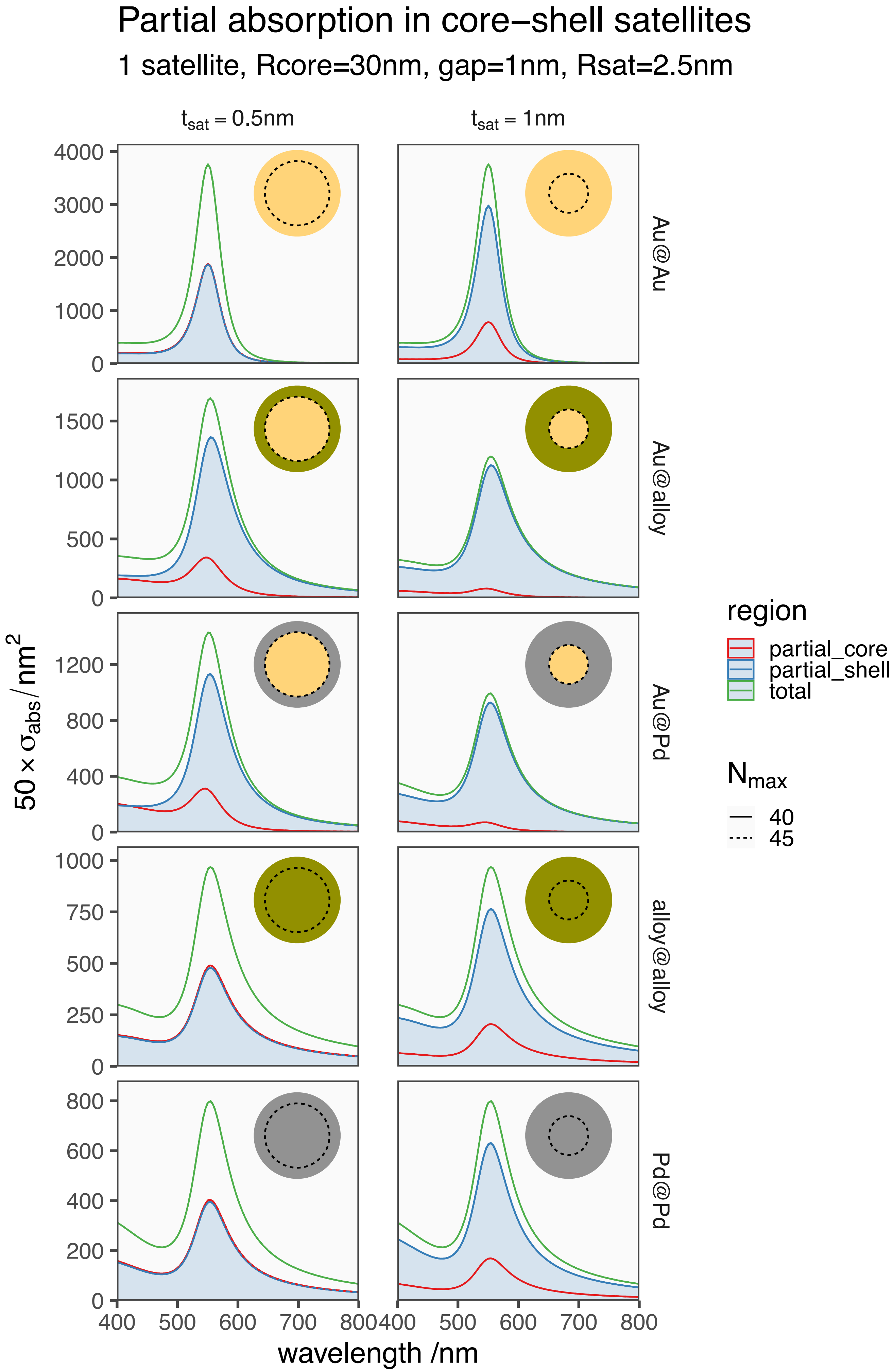
EFFECT OF SATELLITE CONCENTRATION

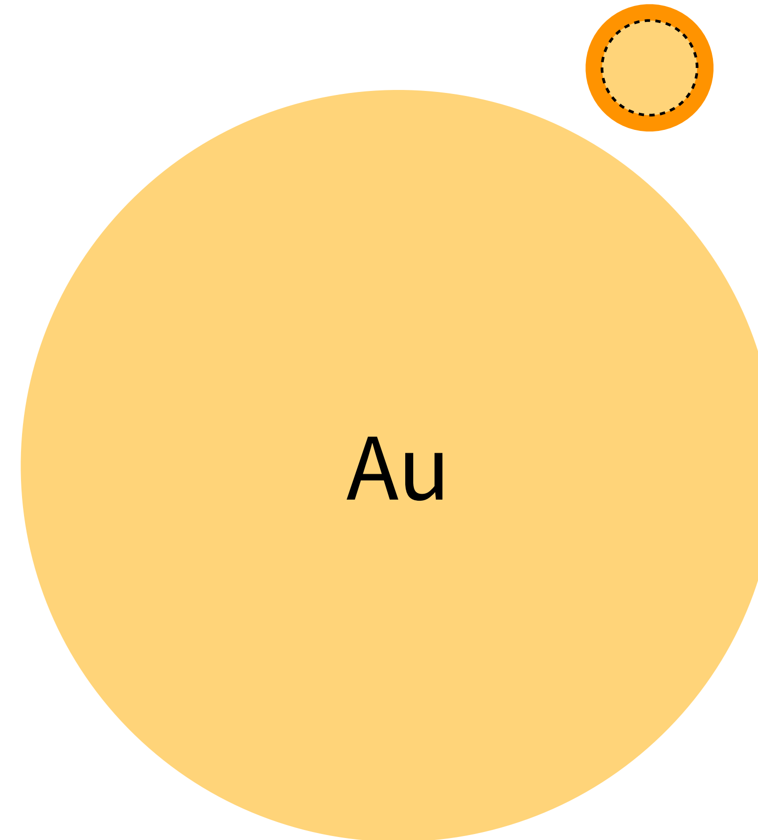


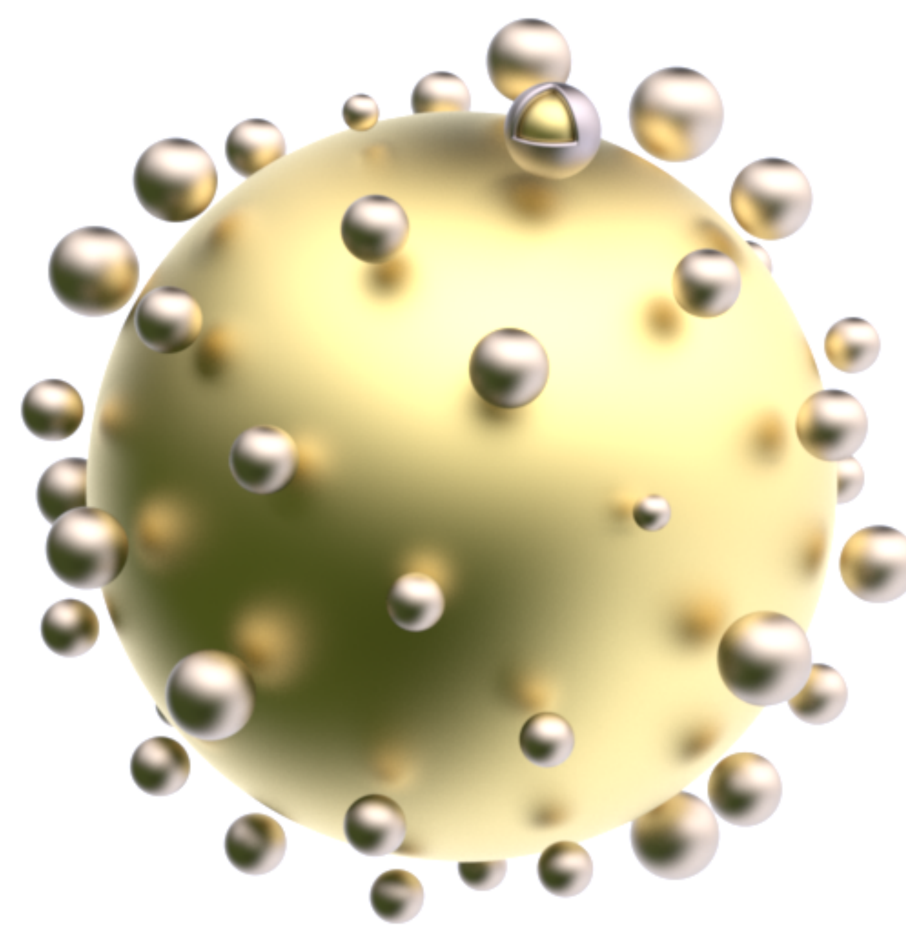


STRENGTHS AND WEAKNESSES 👍👌👎🤞

	METHOD	ACCURACY	TIME	SCALING	DETAILED INFO
	TERMS (T-matrix)	✔	✘	✘	✔
	SCUFF (surface int.)	✔	✘	≈	✔
	GCDM (coupled dip.)	✔	≈	≈	≈
	Mie	≈	✔	✔	✘


Partial absorption in core−shell satellites
1 satellite, Rcore=30nm, gap=1nm, Rsat=2.5nm
t_{sat} = 0.5nm
t_{sat} = 1nm
Au@Au
Au@alloy
Au@Pd
alloy@alloy
Pd@Pd
$50 \times \sigma_{abs} / nm^2$
wavelength /nm
400
500
600
700
800
region
partial_core
partial_shell
total
N_{max}
40
45


Au



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