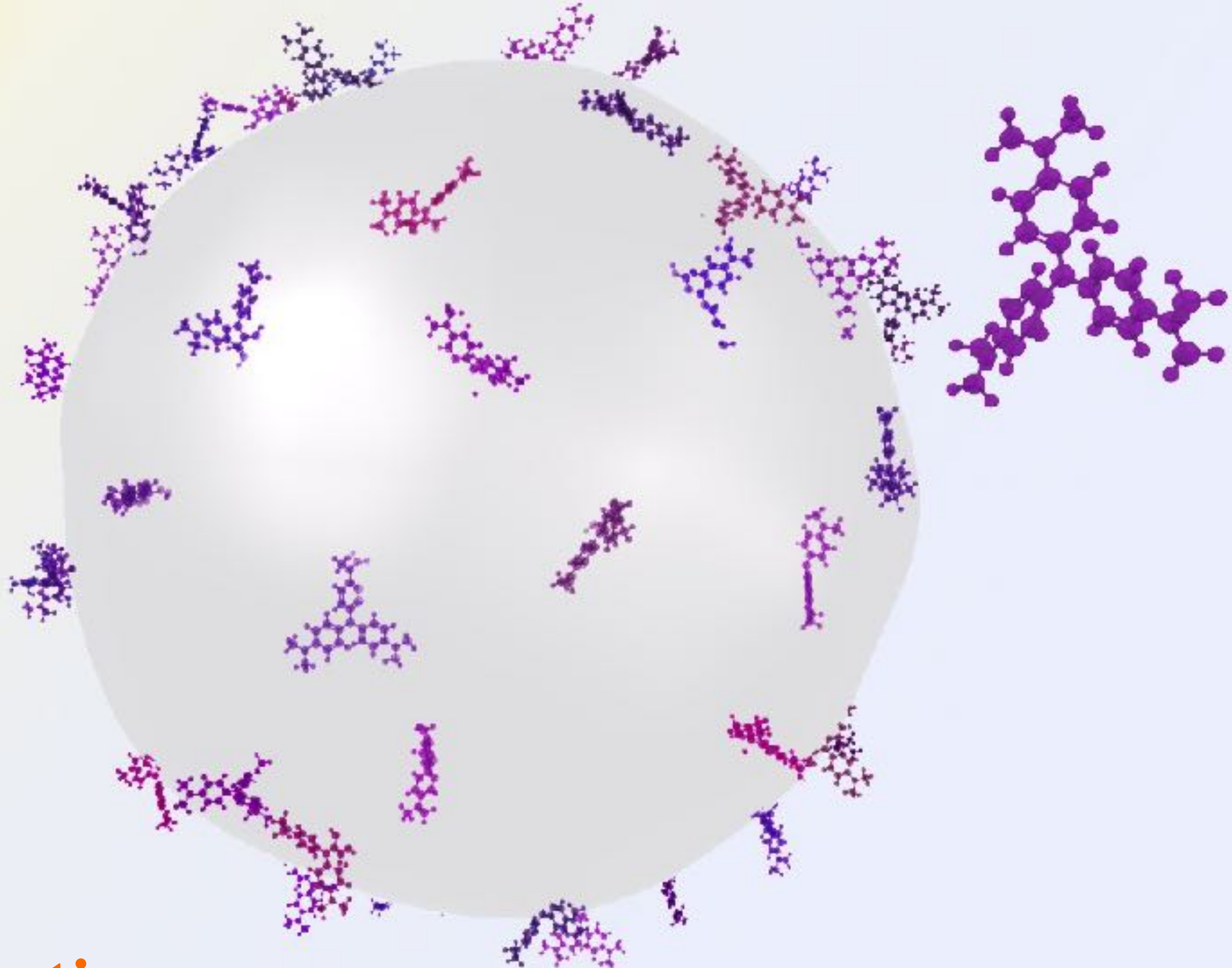
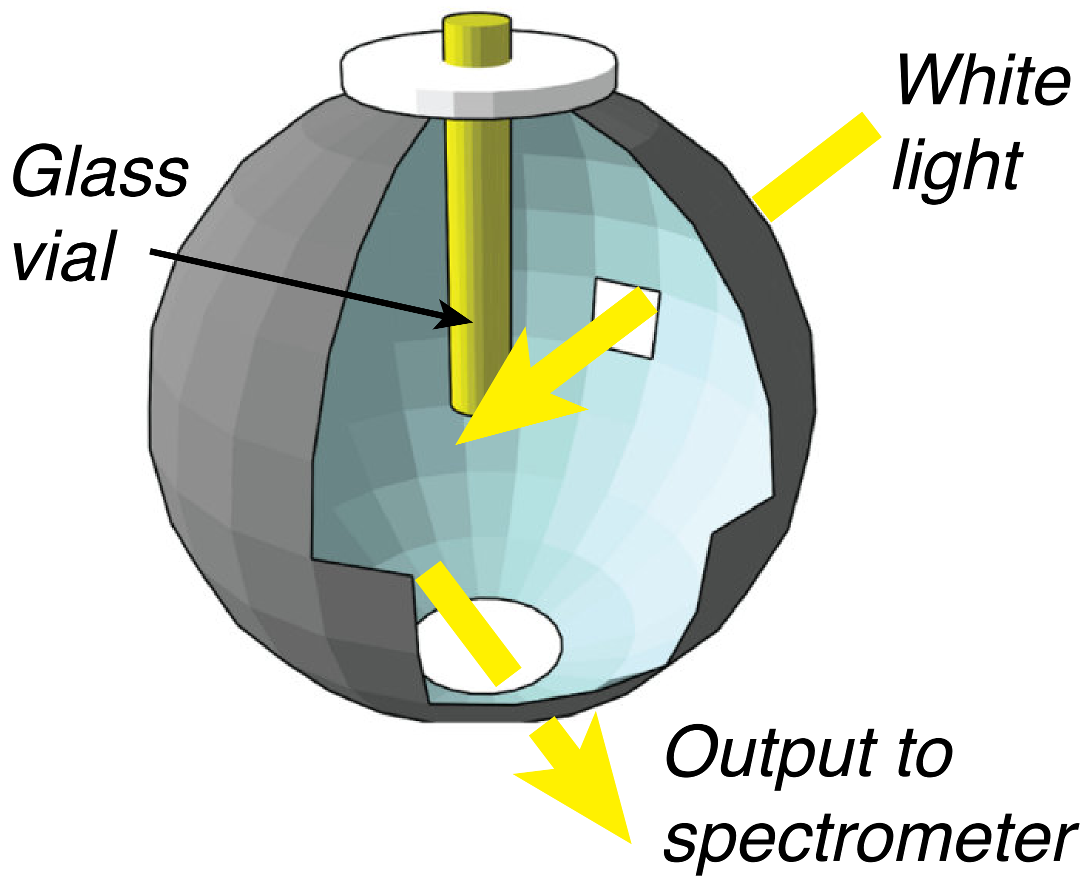
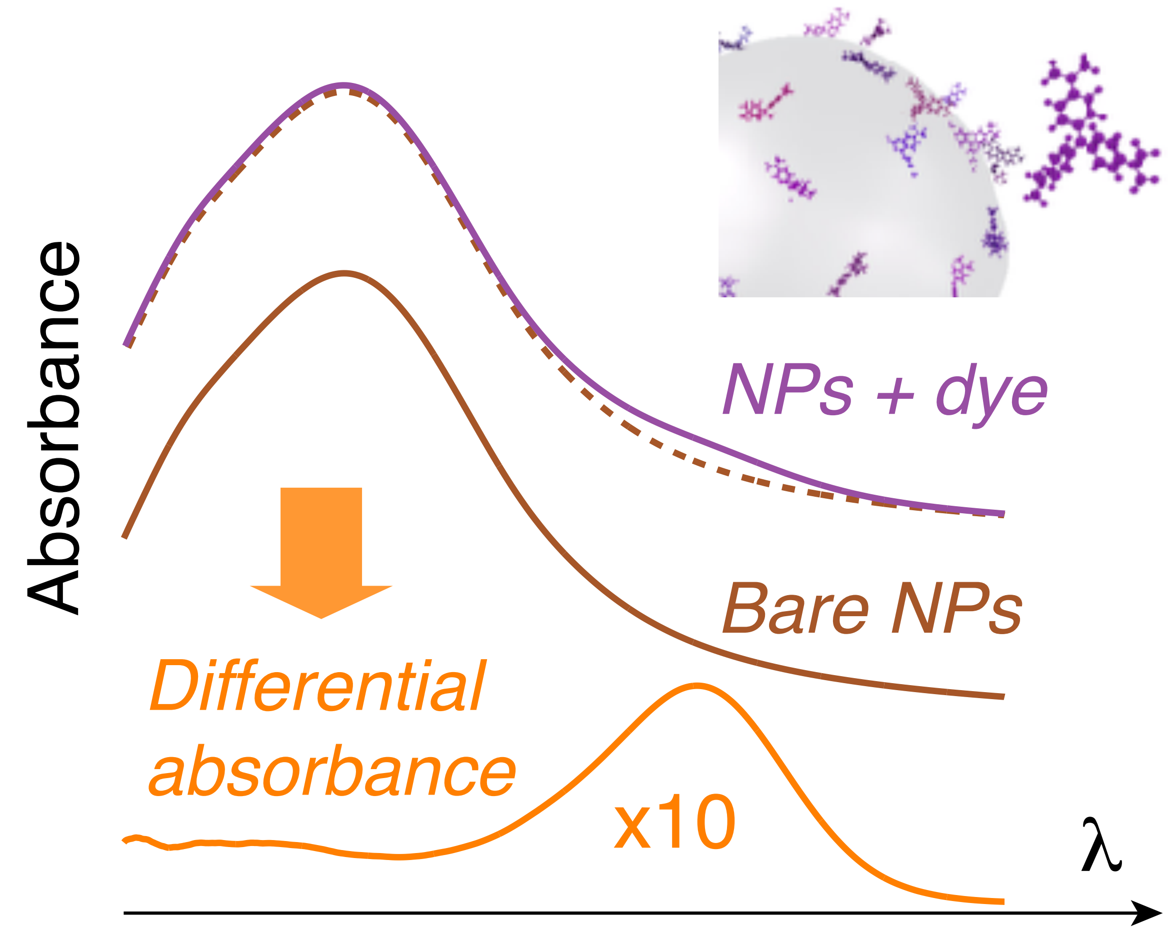


Electromagnetic interactions between molecules surrounding a metallic sphere

Baptiste Auguié • Eric Le Ru
Victoria University of Wellington
New Zealand

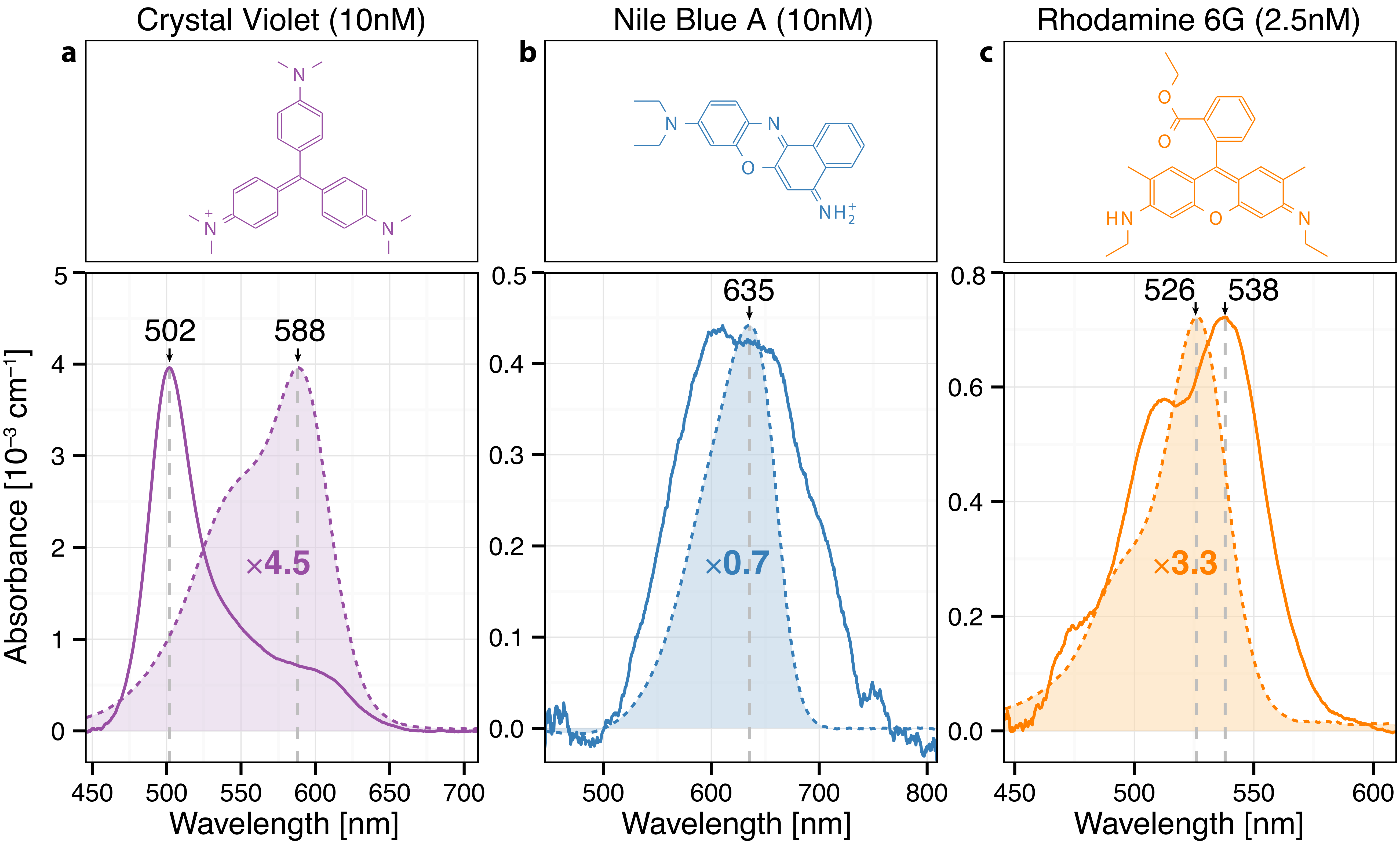
Integrating sphere for absorption measurements



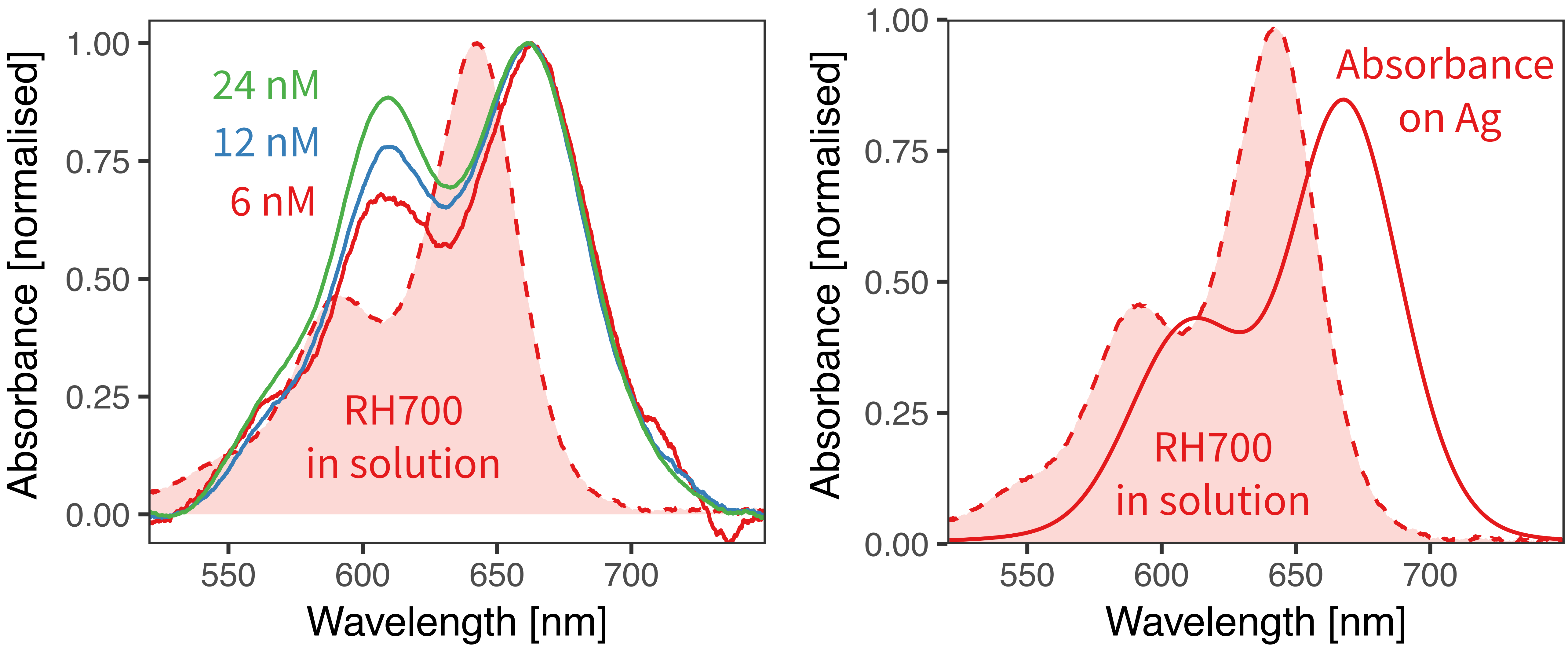


B. L. Darby, B. Auguié, M. Meyer, A. E. Pantoja, and E. C. Le Ru
Nat. Photonics 10.1 (2016)

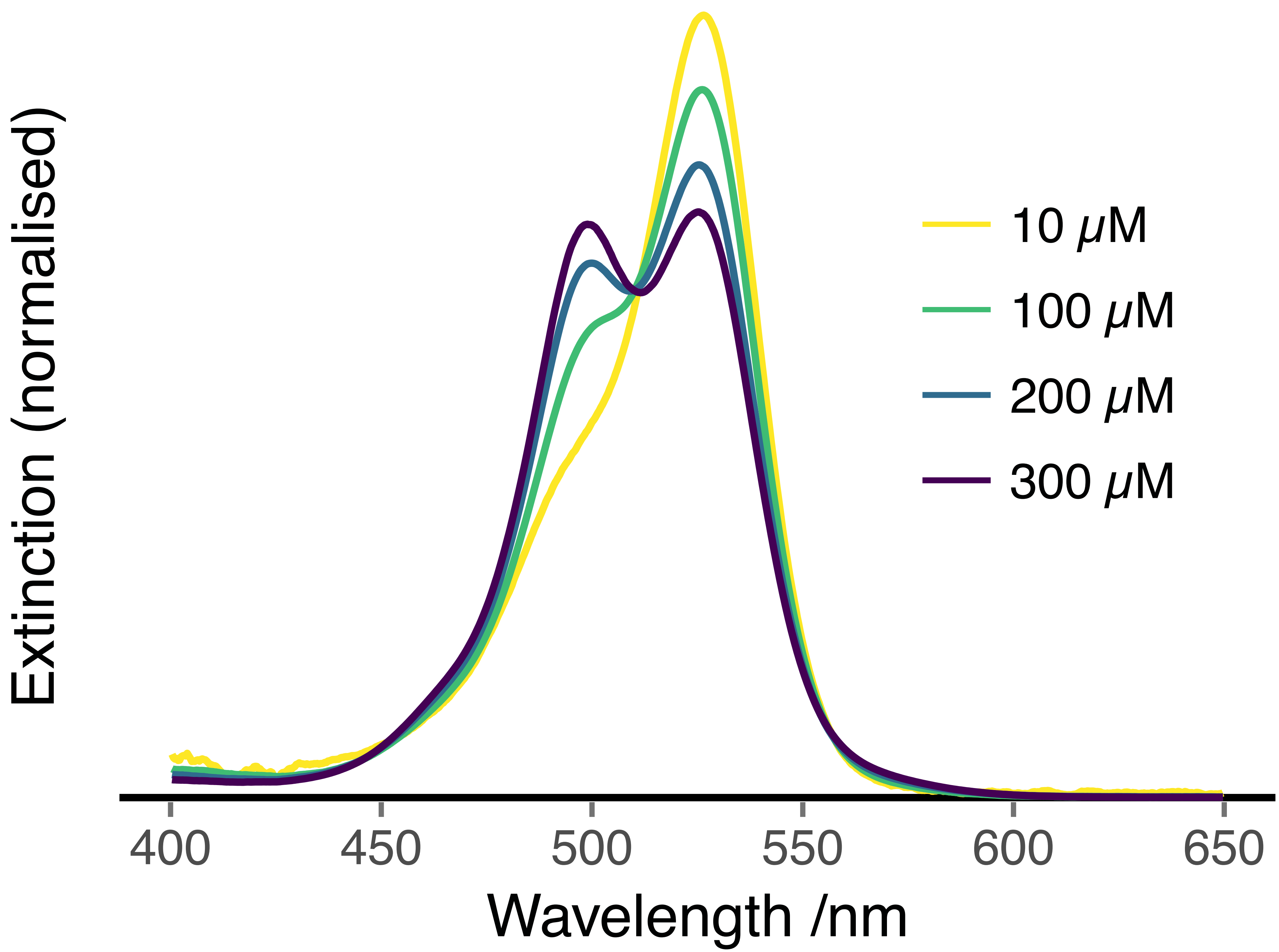
Different molecules



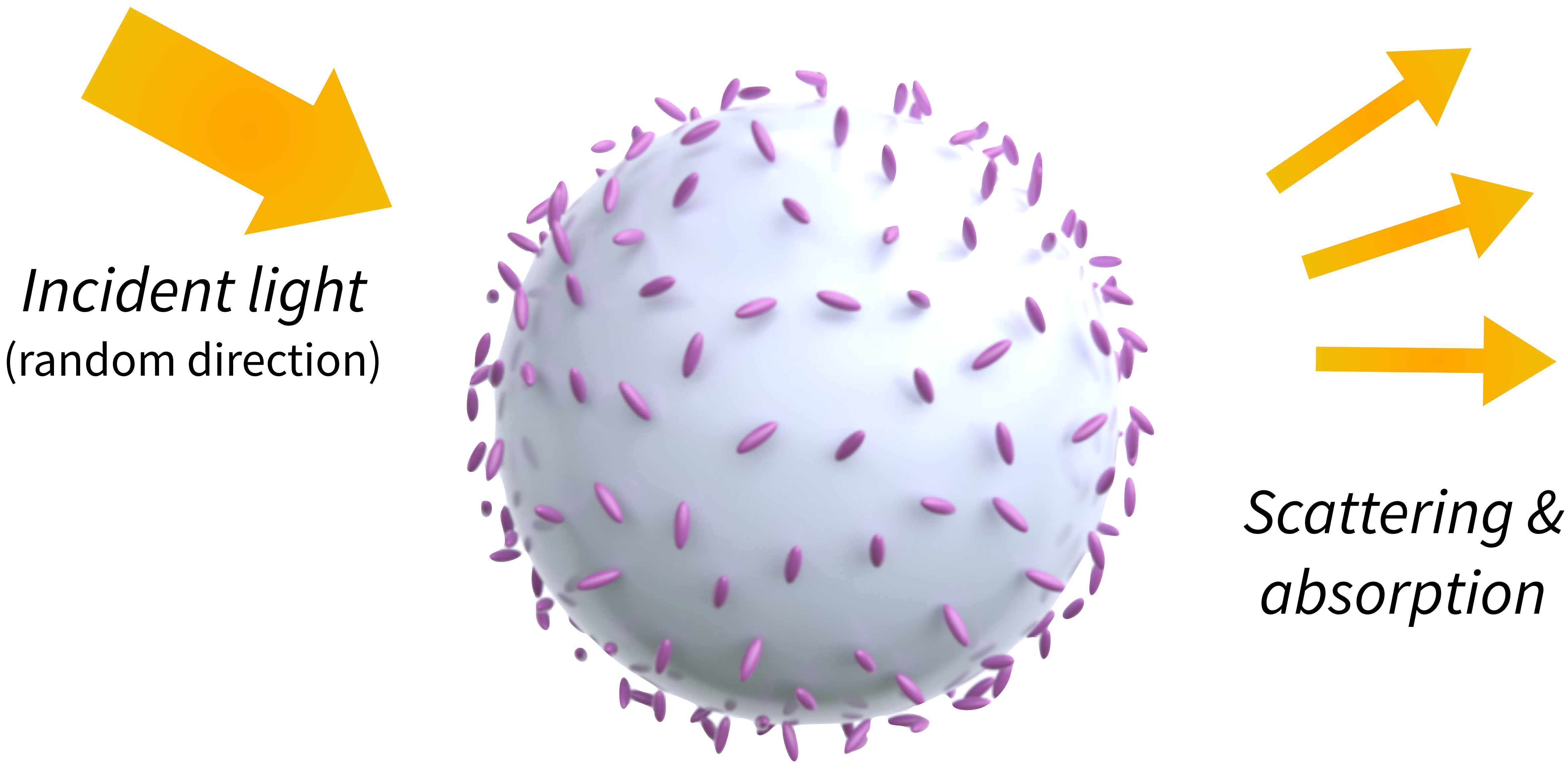
Concentration dependence



Dye–dye interactions (in solution)

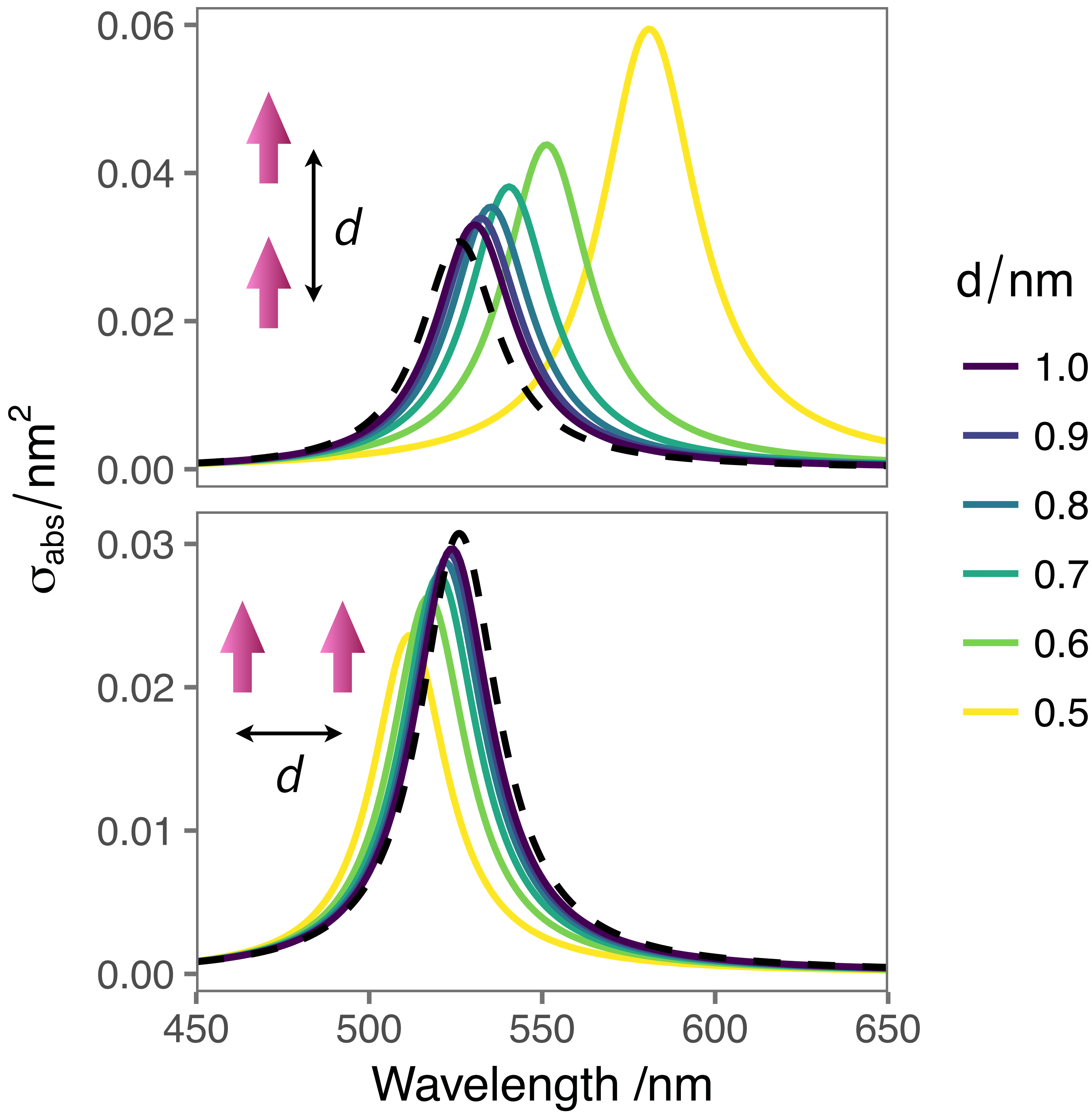


Light scattering problem



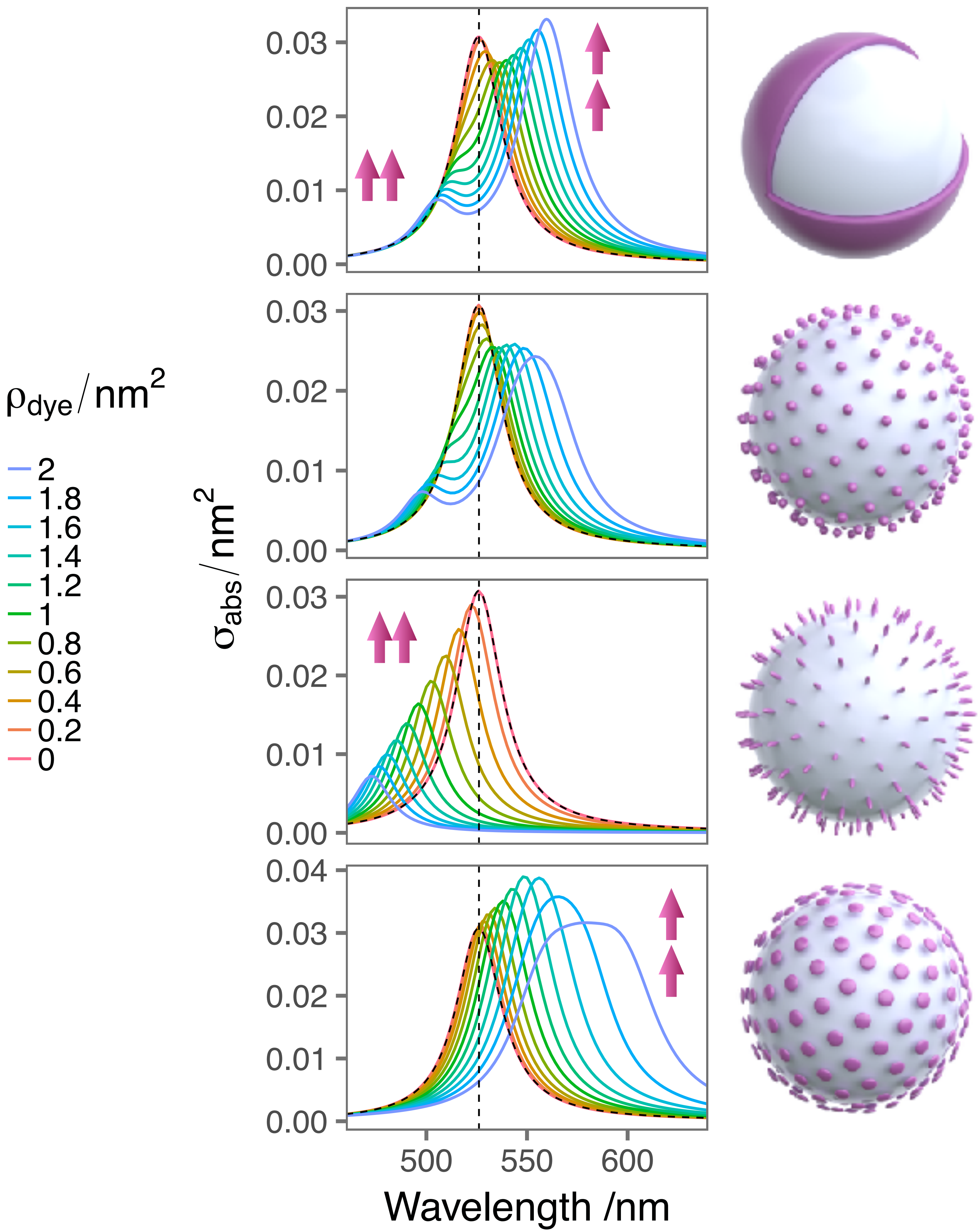
dipole-dipole and dipole-sphere interactions

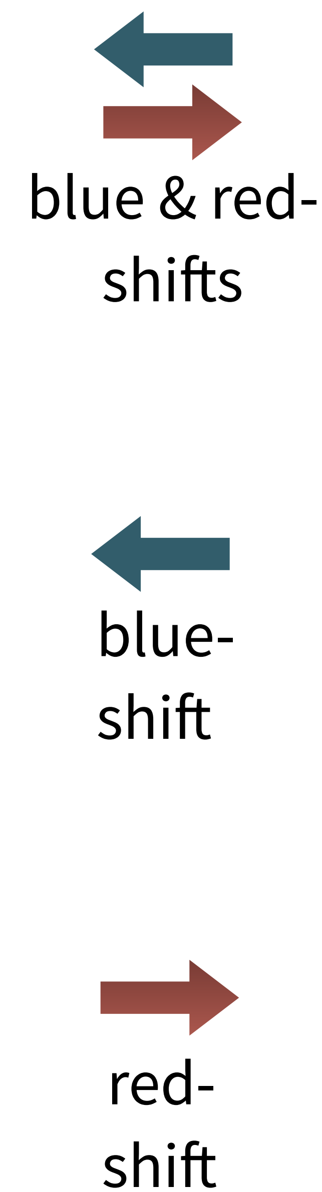
Dimers – hybridisation



Void shell results

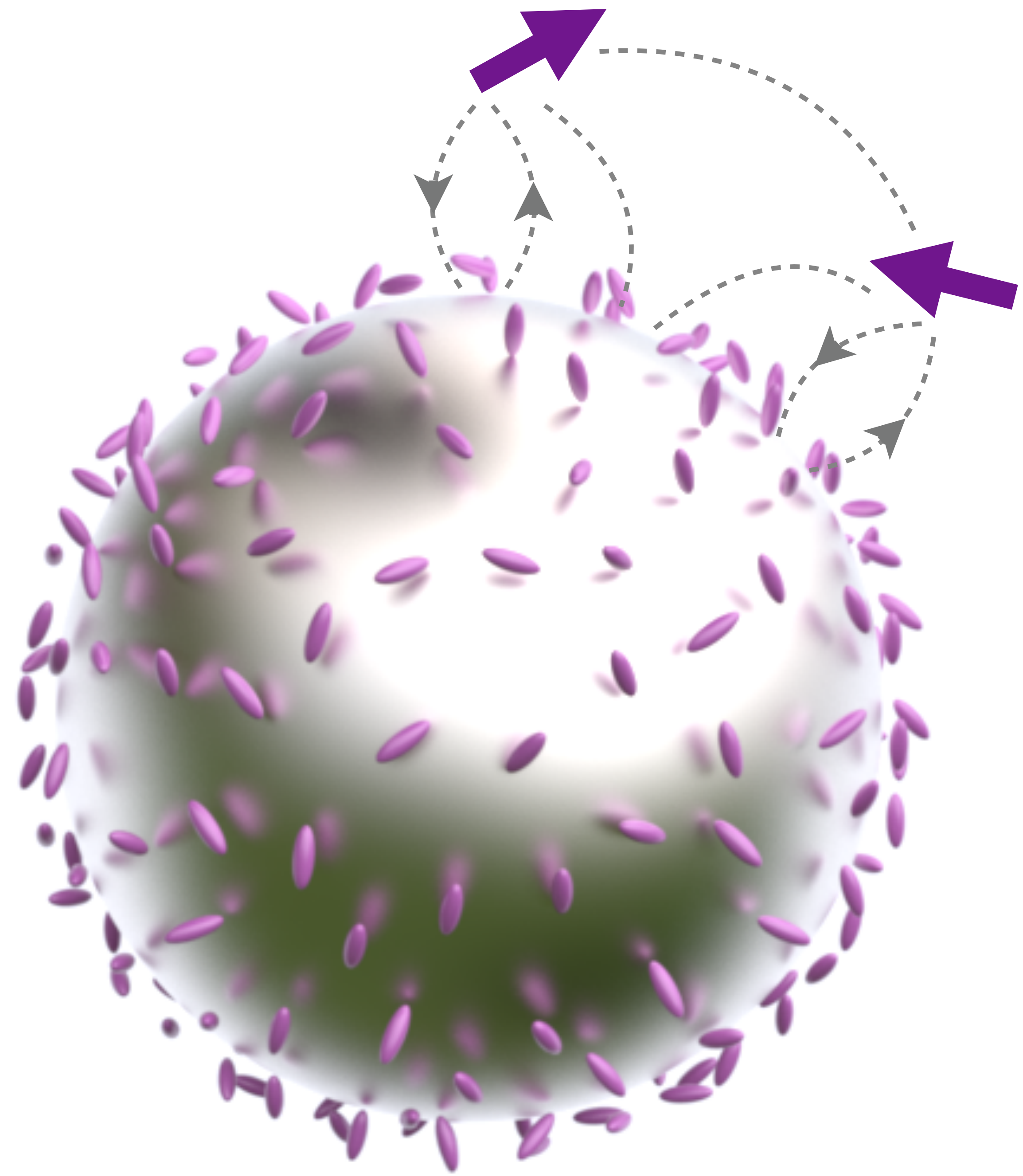
J. Phys. Chem.C 122, 33 (2018)
10.1021/acs.jpcc.8b05542





ADDING THE CORE PARTICLE

- Sphere-mediated coupling
- Self-reaction (“image” dipole)
- Additional excitation from sphere-scattered field



Technical challenges:

- Slow convergence of series*
- Easier when source along z-axis:
  ➥ *many rotations!*

* M. Majić, B. Auguié, E. Le Ru • Phys. Rev. E 95, 033307 (2017)

HYBRID COUPLED-DIPOLE – MIE THEORY

- Extended coupled-dipole system

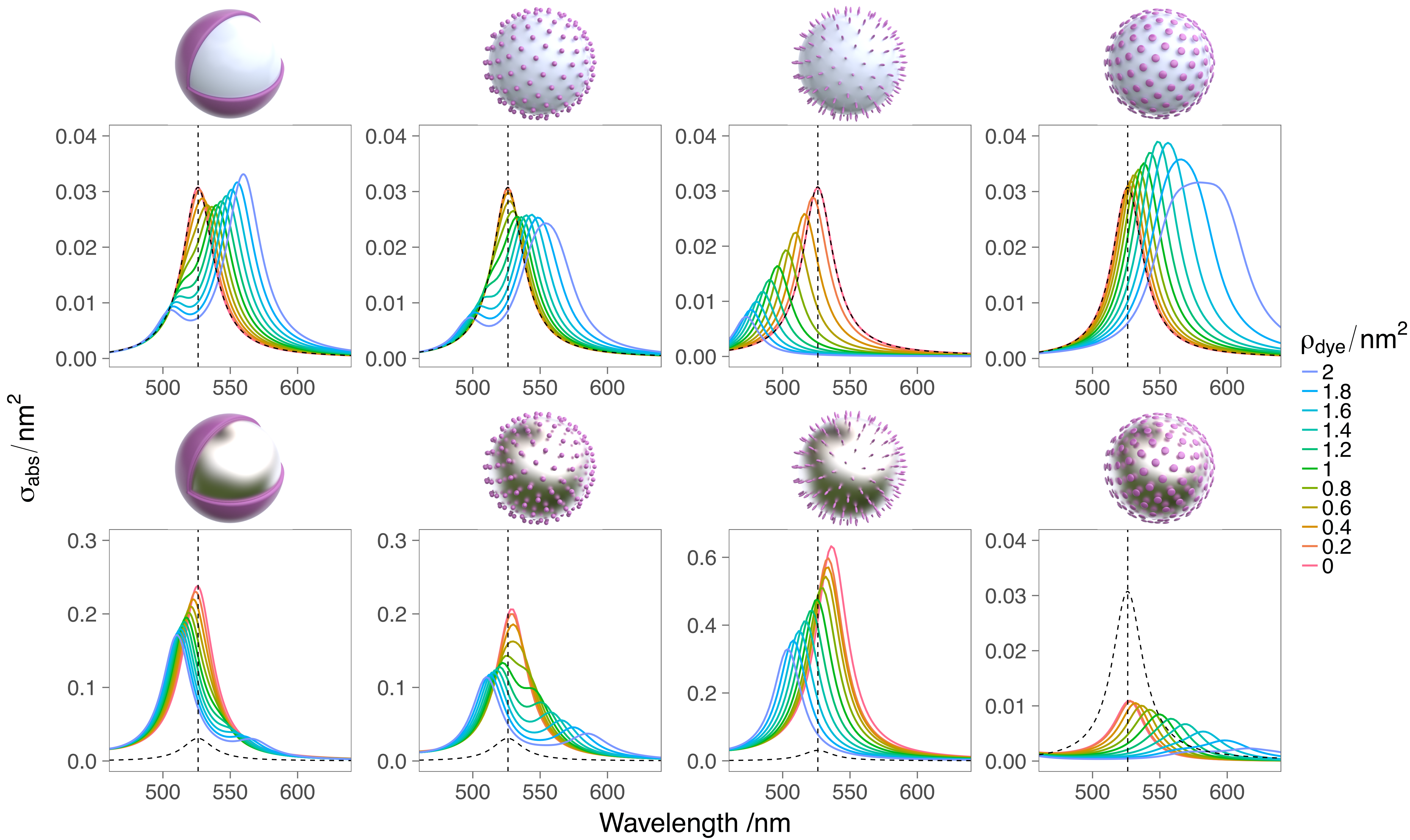
$$\mathbf{E}^i = \mathbf{E}^i_{\text{inc}} + \mathbf{E}^i_{\text{SPH}} + \sum_{j\neq i} \mathbb{G}_{ij}\alpha_j\mathbf{E}^j + \sum_{\forall j} \mathbb{S}_{ij}\alpha_j\mathbf{E}^j$$

- Cross-sections: Mie theory with *N+1* sources

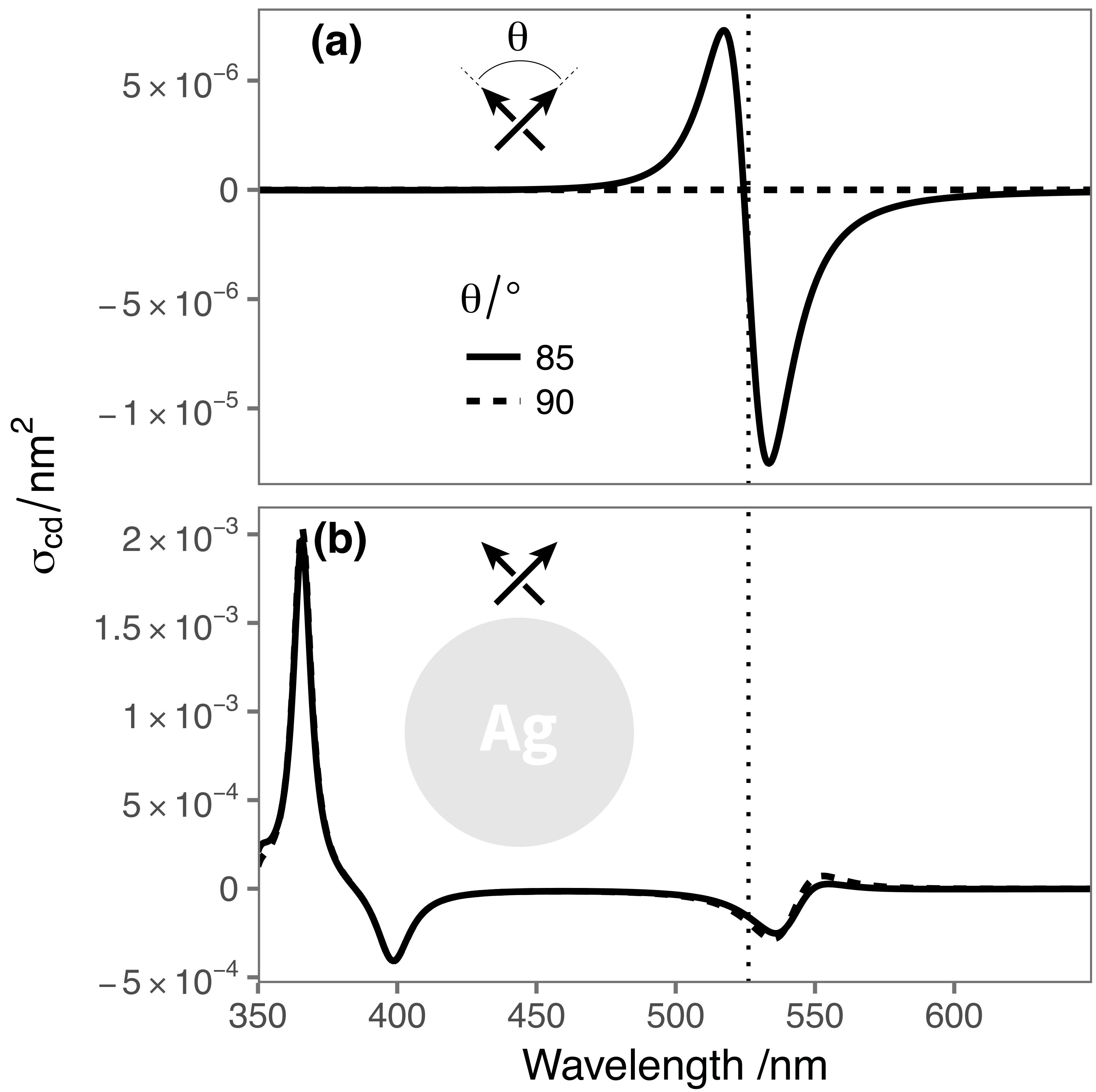
$$\begin{cases} \mathbf{E}_{\text{DIP}} = \sum_{n=1}^{\infty}\sum_{m=-n}^{n} a^{\text{DIP}}_{mn}\mathbf{M}^{(1)}(k_1,\mathbf{r}) + b^{\text{DIP}}_{mn}\mathbf{N}^{(1)}(k_1,\mathbf{r}), & r < r_{\text{dip}} \\ \mathbf{E}_{\text{DIP}} = \sum_{n=1}^{\infty}\sum_{m=-n}^{n} e^{\text{DIP}}_{mn}\mathbf{M}^{(3)}(k_1,\mathbf{r}) + f^{\text{DIP}}_{mn}\mathbf{N}^{(3)}(k_1,\mathbf{r}), & r > r_{\text{dip}} \end{cases}$$

- Orientation-averaging (multiple directions)

Effect of metal core – comparison

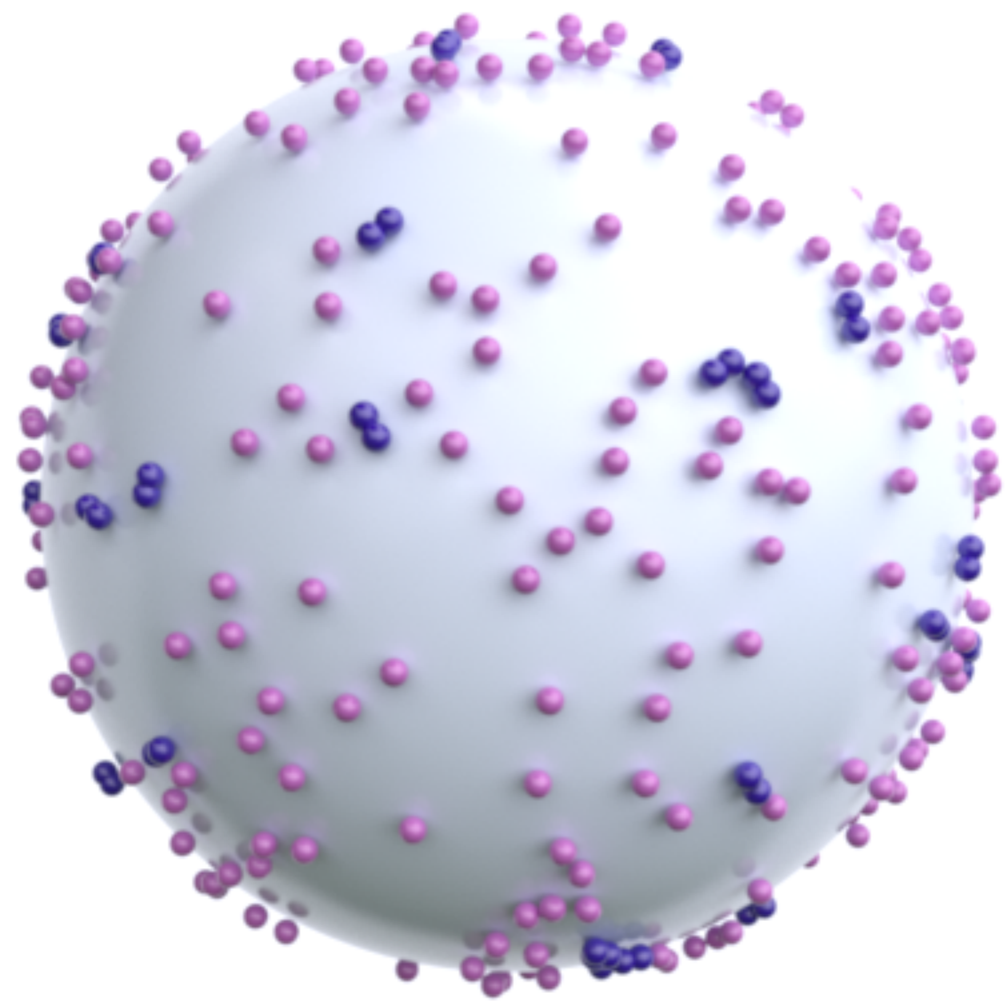


POLARISATION EFFECTS • CIRCULAR DICHROISM



ADSORBED MOLECULES: MANY UNKNOWNS

- "Random" landings?
- Surface diffusion?
- Dimerisation?
- Influence of ions
- Surface orientation



Modelling + experiments *might* shed some light

Perspectives

- ☑ **Effect of orientation, density, inhomogeneities, dipole image**
- ☐ Comparison with anisotropic shell models
- ☐ Shell of finite thickness, unusual coverage (e.g. Janus)
- ☐ Surface dimerisation

Less likely:

- ☒ *Chemical enhancement*, DFT predictions
- ☒ Super-radiance
- ☒ Non-spherical particles

Thank you!

Acknowledgements

TE WHARE WĀNANGA O TE ŪPOKO O TE IKA A MĀUI
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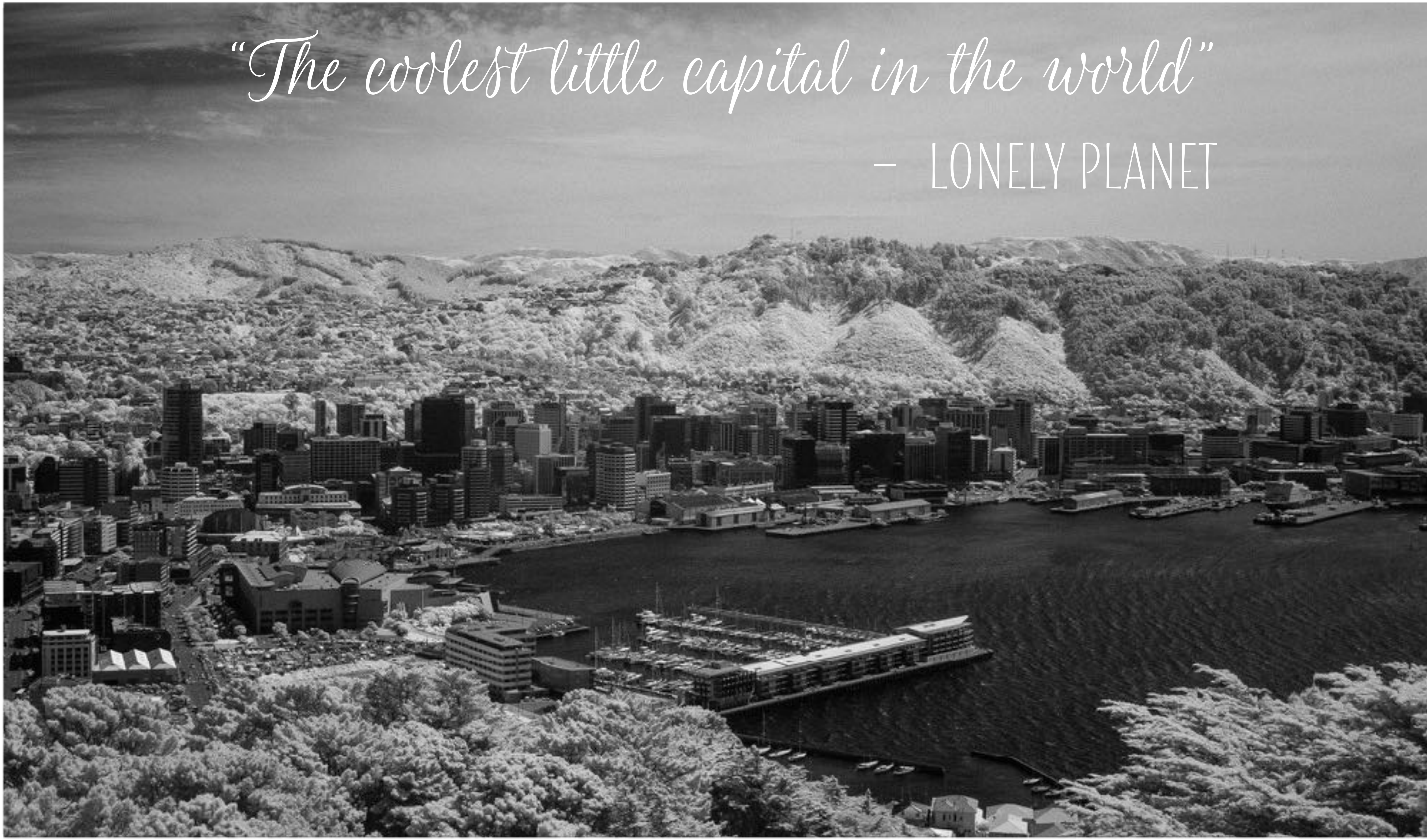
ROYAL
SOCIETY
TE APĀRANGI

DODD-WALLS CENTRE
for Photonic and Quantum Technologies



AMN9 conference

Advanced Materials & Nanotechnology

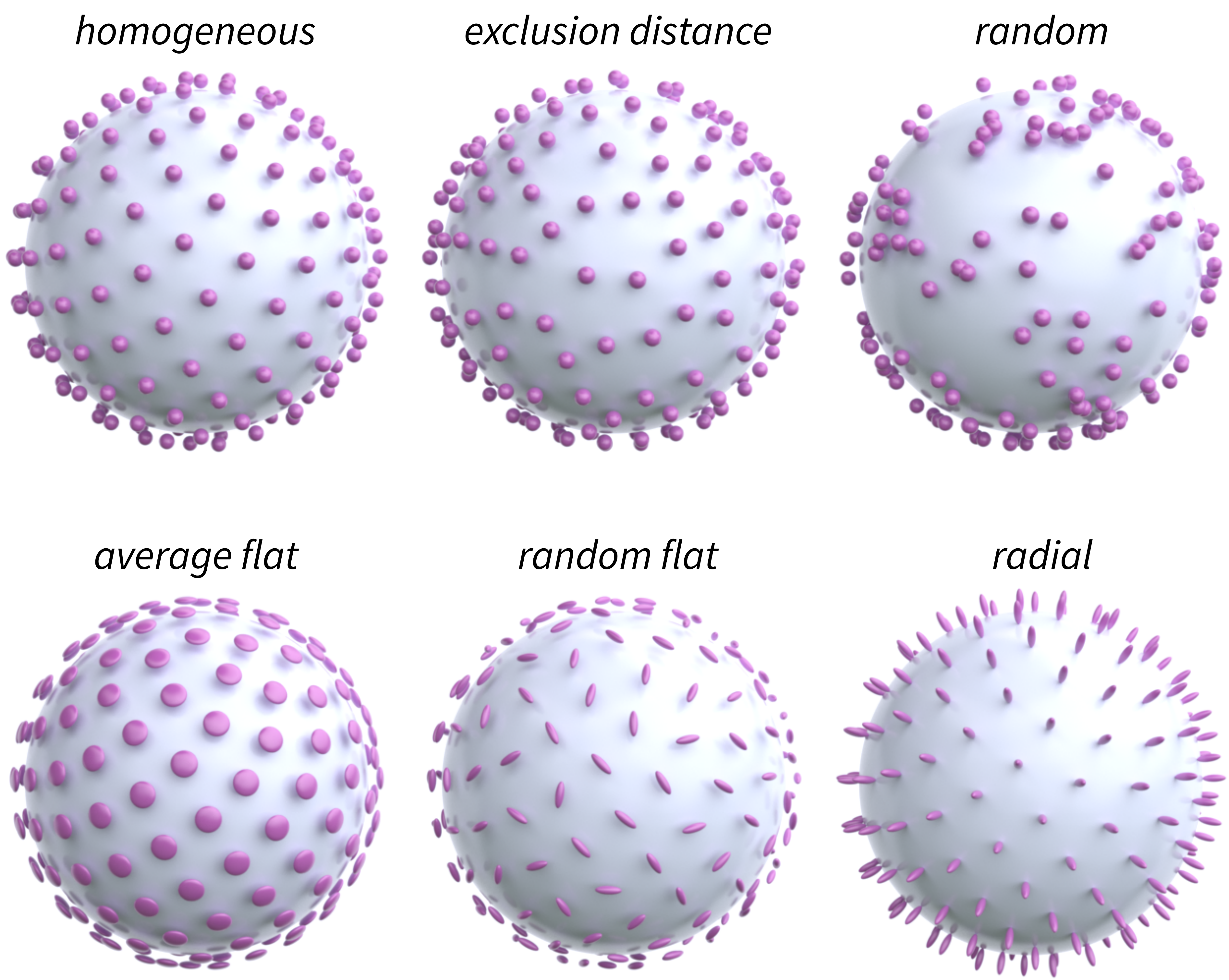
Wellington, New Zealand

10–14 February 2019

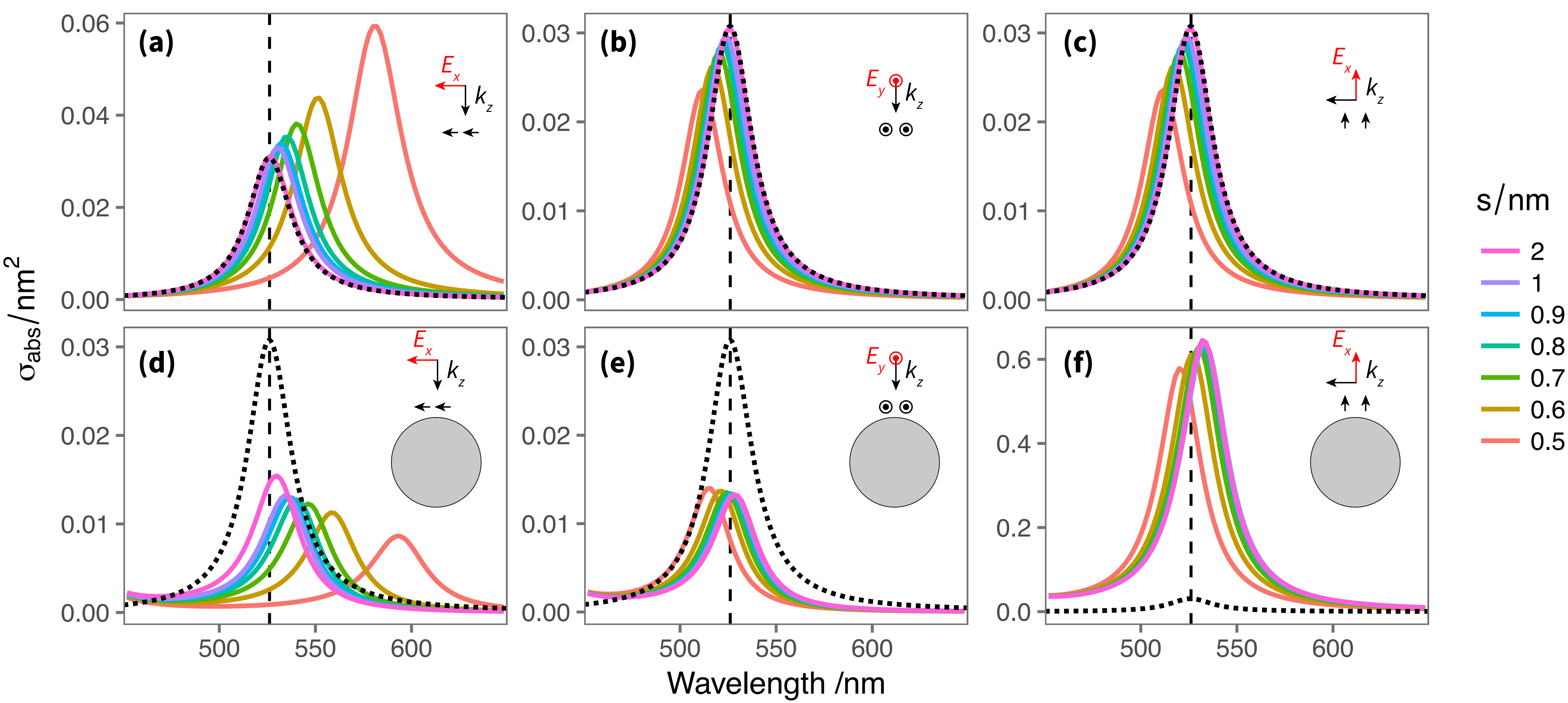
Supplementary Material

BEYOND HOMOGENEOUS ISOTROPIC SHELLS

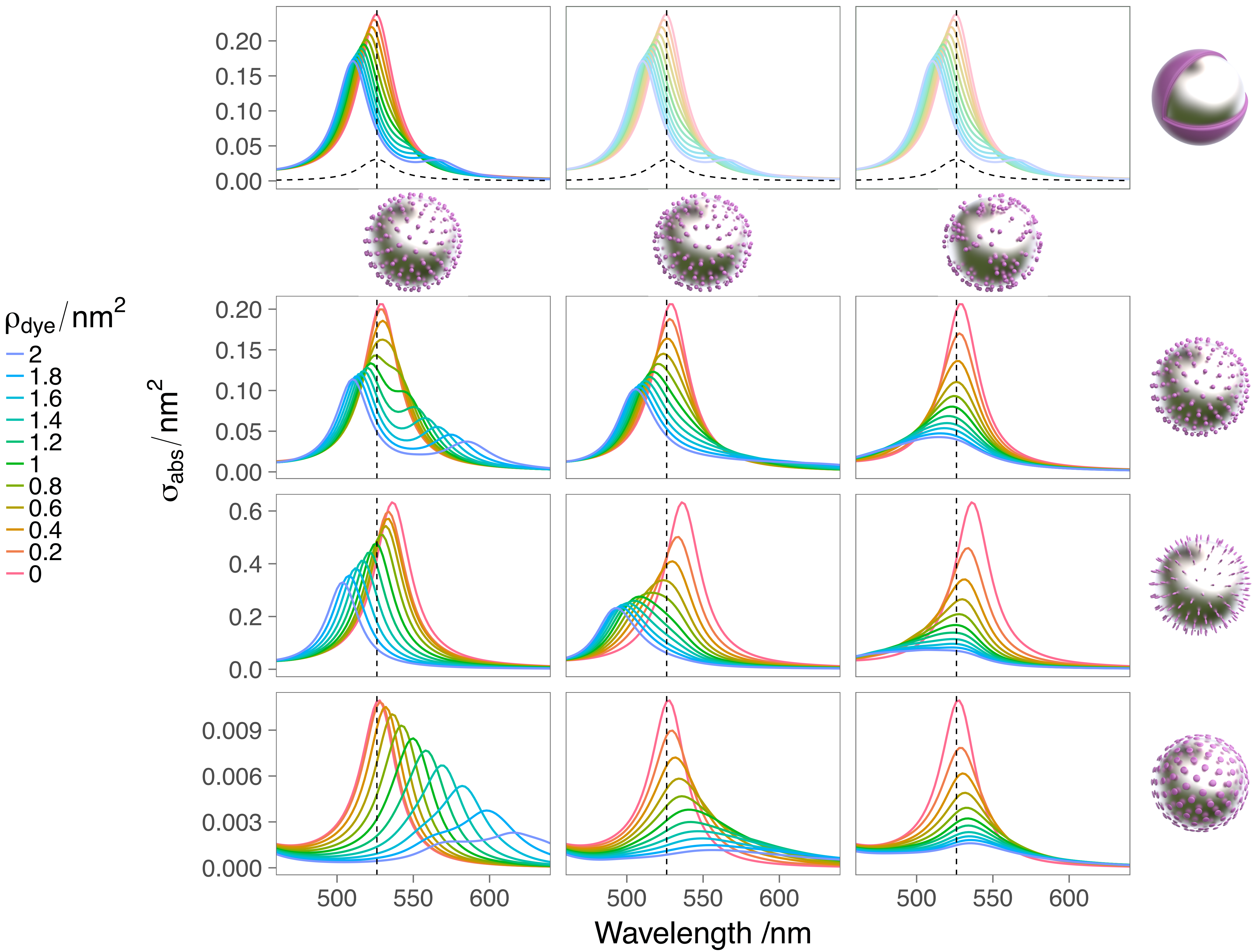
- Position
- Orientation



Dimer



(a)
(b)
(c)
(d)
(e)
(f)
0.06
0.04
0.02
0.00
0.03
0.02
0.01
0.00
0.6
0.4
0.2
0.0
500
550
600
σ_{abs}/nm^2
Wavelength /nm
E_x
E_y
k_z
s/nm
2
1
0.9
0.8
0.7
0.6
0.5



ρ_{dye}/nm^2
2
1.8
1.6
1.4
1.2
1
0.8
0.6
0.4
0.2
0
σ_{abs}/nm^2
0.00
0.05
0.10
0.15
0.20
0.0
0.2
0.4
0.6
0.000
0.003
0.006
0.009
500
550
600
Wavelength /nm