Time-Harmonic Optical Chirality for Classification of Scatterers

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CONSEJO SUPERIOR DE INVESTIGACIONES CIENTÍFICAS

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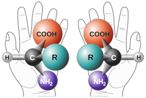
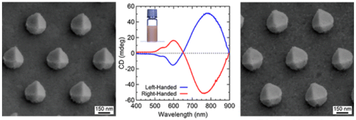
Chirality

- *Chirality is a geometric property of some molecules and ions.* [1]
- chiral molecules are non-superimposable on their mirror image



- different response to right-/left-handed circularly polarized light
- plasmonics enhance optical response [2]



Left-Handed Right-Handed

- optical chirality describes chiral excitation of molecule [3]

[1] https://en.wikipedia.org/wiki/Chirality_(chemistry), 01/11/17
[2] K. M. McPeak et al., *Nano letters*, 14(5):2934–2940, 2014
[3] Y. Tang et al., *Physical Review Letters*, 104(16):163901, 2010

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Conservation of Optical Chirality

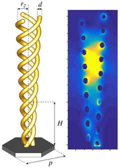
- *illumination*: both states of circularly polarized light (CPL)
- differential CPL excitation of molecule $\Delta A = 2\omega\,\mathrm{Re}\,(\alpha_{em})\,\mathfrak{X}$
- time-averaged optical chirality density $\mathfrak{X}$ in free space

$$\mathfrak{X} = -\frac{\varepsilon_0\omega}{2}\,\mathrm{Im}\,(\boldsymbol{\mathcal{E}}^* \cdot \boldsymbol{\mathcal{B}})$$

- continuity equation

$$\boxed{2i\omega\,(\mathfrak{X}_\mathrm{e} - \mathfrak{X}_\mathrm{m}) + \boldsymbol{\nabla} \cdot \boldsymbol{\mathfrak{S}} = \frac{1}{4}\left\{\boldsymbol{\mathcal{J}}^* \cdot (\boldsymbol{\nabla} \times \boldsymbol{\mathcal{E}}) + \boldsymbol{\mathcal{E}} \cdot (\boldsymbol{\nabla} \times \boldsymbol{\mathcal{J}}^*)\right\}}$$

(cf. conservation of energy: $2i\omega(\mathcal{U}_\mathrm{e} - \mathcal{U}_\mathrm{m}) + \boldsymbol{\nabla} \cdot \boldsymbol{\mathcal{S}} = -\frac{1}{2}\boldsymbol{\mathcal{J}}^* \cdot \boldsymbol{\mathcal{E}}$)



- optical chirality flux density $\boldsymbol{\mathfrak{S}}$
- rhs for localized source: $\frac{1}{2}\omega\boldsymbol{\mathcal{J}}^* \cdot \boldsymbol{\mathcal{B}}$

[4] P. Gutsche et al., In *SPIE 9756*, page 97560X, 2016, arXiv:1603.05011 [5] L. V. Poulikakos et al., *ACS Photonics*, 3(9):1619–1625, 2016

Chiral Maxwell's Equations and Mie Theory

- Maxwell's equations for reciprocal bi-anisotropic media [6]

$$\mathcal{D} = \varepsilon\mathcal{E} + i\kappa\mathcal{H}$$
$$\mathcal{B} = \mu\mathcal{H} - i\kappa\mathcal{E}$$

- basis of vector spherical harmonics for isolated scatterers [7]

$$\mathcal{E}_{\text{inc}}(\mathbf{x},t) = e^{-i\omega t}\sum_{mn} p_{mn}\mathbf{N}^{(1)}_{nm}(\mathbf{x}) + q_{mn}\mathbf{M}^{(1)}_{nm}(\mathbf{x}), \quad \mathcal{H}_{\text{inc}}(\mathbf{x},t) = -\frac{i}{Z}e^{-i\omega t}\sum_{mn} p_{mn}\mathbf{M}^{(1)}_{nm}(\mathbf{x}) + q_{mn}\mathbf{N}^{(1)}_{nm}(\mathbf{x})$$

$$\mathcal{E}_{\text{sca}}(\mathbf{x},t) = e^{-\omega t}\sum_{mn} a_{mn}\mathbf{N}^{(3)}_{nm}(\mathbf{x}) + b_{mn}\mathbf{M}^{(3)}_{nm}(\mathbf{x}), \quad \mathcal{H}_{\text{sca}}(\mathbf{x},t) = -\frac{i}{Z}e^{-i\omega t}\sum_{mn} a_{mn}\mathbf{M}^{(3)}_{nm}(\mathbf{x}) + b_{mn}\mathbf{N}^{(3)}_{nm}(\mathbf{x}).$$

- scattered and extinction energy

$$W_{\text{sca}} = \frac{1}{2k^2Z}\sum_{mn}|a_{mn}|^2 + |b_{mn}|^2, \quad W_{\text{ext}} = -\frac{1}{2k^2Z}\sum_{mn}\text{Re}\left(p^*_{mn}a_{mn} + q^*_{mn}b_{mn}\right)$$

- scattered and extinction chirality

$$\boxed{X_{\text{sca}} = \frac{1}{2kZ}\sum_{mn}2\,\text{Re}\left(a^*_{mn}b_{mn}\right), \quad X_{\text{ext}} = -\frac{1}{2kZ}\sum_{mn}\text{Re}\left(p^*_{mn}b_{mn} + q^*_{mn}a_{mn}\right)}$$

[6] I. V. Lindell and A. H. Sihvola. Artech House, 1994

[7] M. Mishchenko, et al. Cambridge University Press, 2002

Non-Rayleigh Dipolar Particles

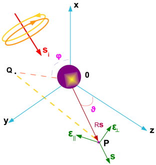
- scatterer as bi-isotropic dipolar particle [8]

$$\mathbf{p} = \alpha_e \boldsymbol{\mathcal{E}}_{\text{inc}} - \alpha_{em} \boldsymbol{\mathcal{B}}_{\text{inc}}$$
$$\mathbf{m} = \alpha_{me} \boldsymbol{\mathcal{E}}_{\text{inc}} + \alpha_m \boldsymbol{\mathcal{B}}_{\text{inc}}$$

(reciprocal: $\alpha_{em} = -\alpha_{me}$)

Helicity Optical Theorem:

$$\mathcal{W}^a_{\mathcal{H}} + \frac{8\pi c k^3}{3\varepsilon} \operatorname{Im}\left(\mathbf{p} \cdot \mathbf{m}^*\right) = 2\pi c \operatorname{Re}\left(-\frac{1}{n^2}\mathbf{p} \cdot \boldsymbol{\mathcal{B}}^*_{\text{inc}} + \mathbf{m} \cdot \boldsymbol{\mathcal{E}}^*_{\text{inc}}\right)$$



- lossless (achiral) particle

$$\operatorname{Re}\left(\alpha_e\right) = |\alpha_e|^2, \quad \operatorname{Re}\left(\alpha_m\right) = |\alpha_m|^2$$

- dual particle [9]

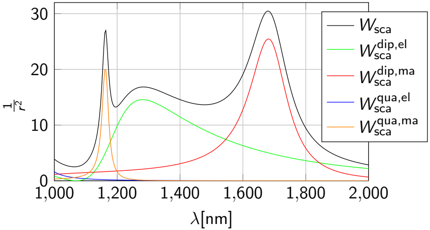
$$\not{d} = \max_{\pm}\left(\frac{|\alpha_e - \alpha_m|}{|\alpha_e \pm \alpha_{em}| + |\alpha_m \pm \alpha_{em}|}\right) = 0$$

- anti-dual particle: $\not{a} = \max_{\pm}\left(\frac{|\alpha_e + \alpha_m \pm 2\alpha_{em}|}{|\alpha_e \pm \alpha_{em}| + |\alpha_m \pm \alpha_{em}|}\right) = 0$
- helicity-annihiling particle: $\not{h}_a = \max_{\pm}\left(\left|\left|\mathbb{X}^{\pm}_{\text{sca}}\right|\right| / \left|\left|\mathbb{W}^{\pm}_{\text{sca}}\right|\right|\right) = 0$

[8] M. Nieto-Vesperinas, *Phys. Rev. A*, 92(2):023813, 2015
[9] I. Fernandez-Corbaton et al., *Physical Review X*, 6(3):031013, 2016

Achiral magnetodielectric sphere (Si particle)

- refractive index $n = 3.5$, radius $r = 230$nm
- incident (+) polarized plane wave



[10] A. García-Etxarri et al., *Opt. Express*, 19(6):4815–4826, 2011, arXiv:1005.5446

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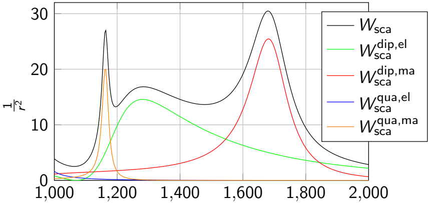
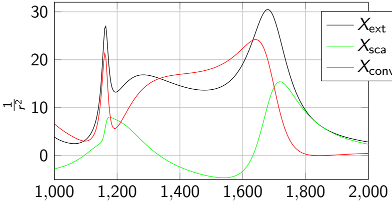


scattered energy in el./ma. multipolar basis



chirality extinction, scattering and conversion

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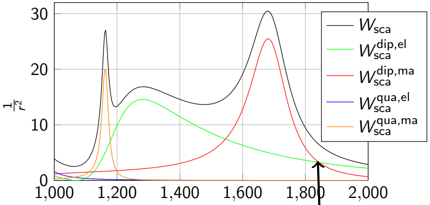

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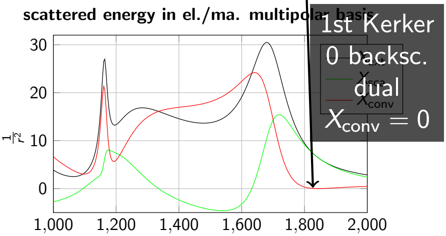

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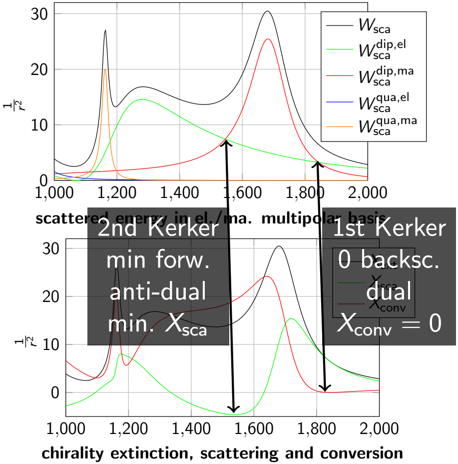

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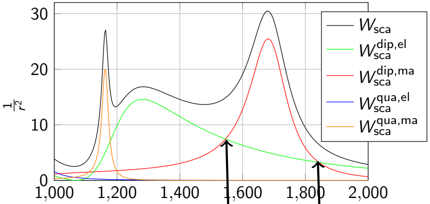

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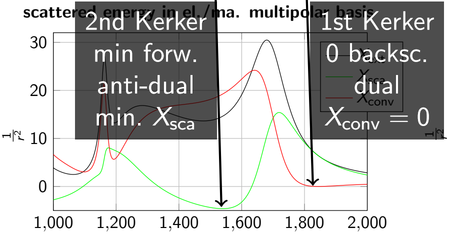

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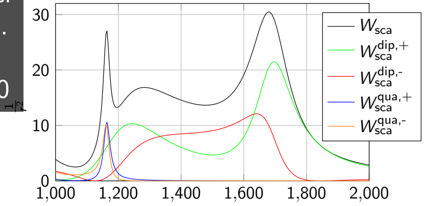

scattered energy in helicity multipolar basis (+/-)

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[11] M. Nieto-Vesperinas et al., *JOSA A*, 28(1):54–60, 2011
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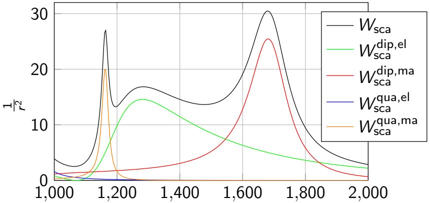

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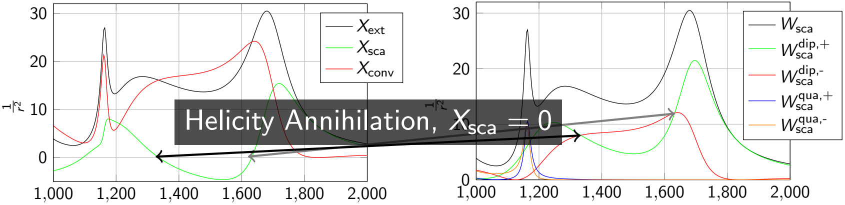

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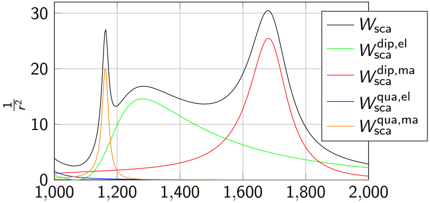

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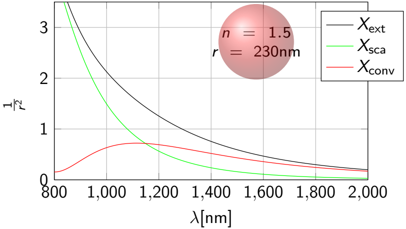

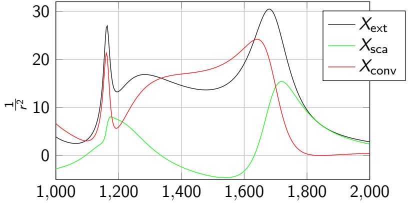

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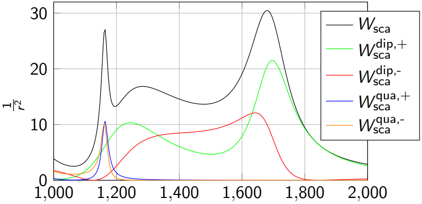

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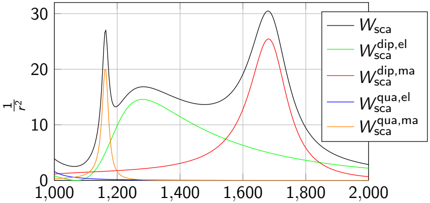

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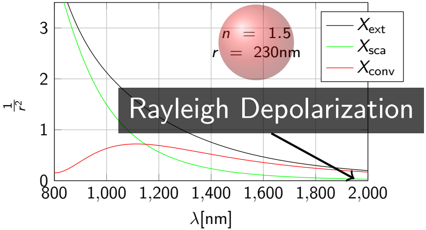

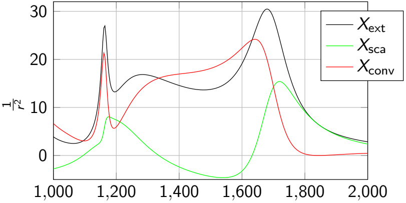

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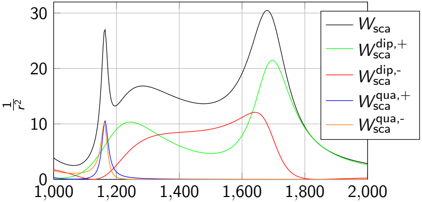

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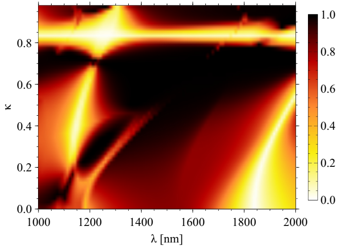
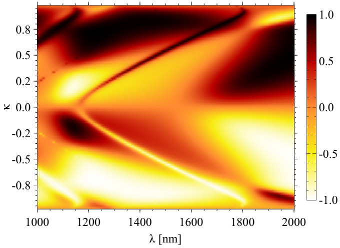
[12] X. Zambrana-Puyalto et al., *Opt. Express*, 21(15):17520–17530, 2013

Chiral Magnetodielectric Sphere

- refractive index $n = 3.5$, radius $r = 230$nm
- varying chirality parameter κ of material
- illumination-independent classification, e.g.
  - duality breaking: $\not{d} = \max_{\pm} \left(\frac{|\alpha_e - \alpha_m|}{|\alpha_e \pm \alpha_{em}| + |\alpha_m \pm \alpha_{em}|} \right)$
  - g-factor: $g = \frac{2\,\mathrm{Re}(\alpha_{em})}{\mathrm{Im}(\alpha_e + \alpha_m)}$



duality breaking $\not{d}$



g-factor

[13] P. Gutsche and M. Nieto-Vesperinas, *arXiv:1802.08029*, 2018

T-Matrix Formalism

- general isolated scatterers described by T-matrix

$$T\begin{pmatrix}\mathbf{p}\\ \mathbf{q}\end{pmatrix}=\begin{pmatrix}T_{ee} & T_{em}\\ T_{me} & T_{mm}\end{pmatrix}\begin{pmatrix}\mathbf{p}\\ \mathbf{q}\end{pmatrix}=\begin{pmatrix}\mathbf{a}\\ \mathbf{b}\end{pmatrix}$$

- scattering response given by matrices $\left(q=(\mathbf{pq})^T\mathbb{Q}(\mathbf{pq})\right)$
- chiral light of well-defined helicity [14]: $\mathbf{q}^{\pm}=\pm\mathbf{p}^{\pm}$
- simplification of matrices: $(T_e^{\pm}=T_{ee}\pm T_{me},\, T_m^{\pm}=T_{mm}\pm T_{em})$

scattered energy: $\mathbb{W}_{\text{sca}}^{\pm}=+\left\{\left(T_{\mathbf{e}}^{\pm}\right)^H T_e^{\pm}+\left(T_{\mathbf{m}}^{\pm}\right)^H T_m^{\pm}\right\}$

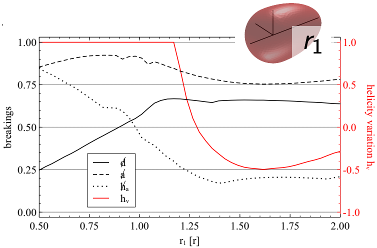
scattered chirality: $\mathbb{X}_{\text{sca}}^{\pm}=\pm\left\{\left(T_{\mathbf{m}}^{\pm}\right)^H T_e^{\pm}+\left(T_{\mathbf{e}}^{\pm}\right)^H T_m^{\pm}\right\}$

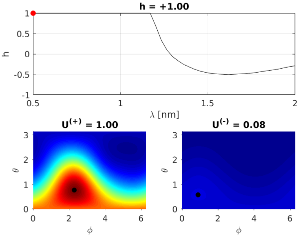
en./ch. extinction: $\mathbb{W}_{\text{ext}}^{\pm}=\pm\mathbb{X}_{\text{ext}}^{\pm}=-\frac{1}{2}\left\{T_e^{\pm}+\left(T_e^{\pm}\right)^H+T_m^{\pm}+\left(T_m^{\pm}\right)^H\right\}$

[14] I. Fernandez-Corbaton, PhD thesis, Macquarie University, Department of Physics and Astronomy, 2014

Achiral Ellipsoid (Si particle)

- achiral ellipsoid with $n = 3.5$ and $r = 230\text{nm}$
- helicity variation: $h_v = \frac{1}{2}\left(\frac{\sum_i \lambda_i^{\mathbb{X}^+_{\text{sca}}}}{\sum_i \left|\lambda_i^{\mathbb{X}^+_{\text{sca}}}\right|} - \frac{\sum_i \lambda_i^{\mathbb{X}^-_{\text{sca}}}}{\sum_i \left|\lambda_i^{\mathbb{X}^-_{\text{sca}}}\right|}\right)$ from scattered chirality $\mathbb{X}_{\text{sca}}$ [13]
- averaged near-field behaviour of scattered field
- electromagnetic energy $\mathcal{U}^{\pm}$ of positive and negative helicity [15]



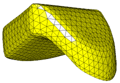


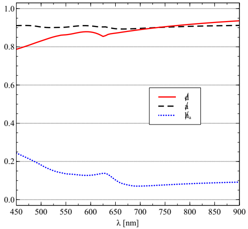
[13] P. Gutsche and M. Nieto-Vesperinas, *arXiv:1802.08029*, 2018

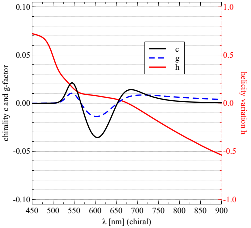
[15] P. Gutsche et al., *Photonics*, 3(4):60, 2016

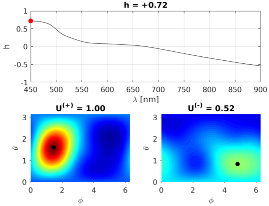
Chiral Nanoparticle

- chiral gold particle [2]
- exhibits large circular dichroism
- complex near-fields





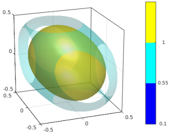




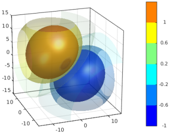
[2] K. M. McPeak et al., *Nano letters*, 14(5):2934–2940, 2014
[13] P. Gutsche and M. Nieto-Vesperinas, *arXiv:1802.08029*, 2018

Helicity Enhancement Factor

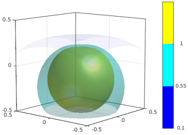
- electromagnetic or helicity basis for circularly polarized (CP) dipoles



$\mathcal{U}$ of achiral CP (el.) dipole



$\mathfrak{X}$ of achiral CP (el.) dipole



$\mathcal{U}$ of chiral CP (el.&ma.) dipole

- enhancement of spontaneous emission rate (Purcell factor, 2D)

$$F_P = 1 + \frac{1}{2W_0} \left\{ \frac{1}{\omega\mu} \operatorname{Im}\left(\mathbf{p}^* \cdot \boldsymbol{\mathcal{E}}_{\text{sca}}\right) + k \operatorname{Im}\left(\mathbf{m}^* \cdot \boldsymbol{\mathcal{H}}_{\text{sca}}\right) \right\}$$

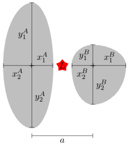
- helicity enhancement factor (analogous to Purcell factor for energy)

$$F_{\mathcal{H}} = 1 + \frac{1}{X_0} \left\{ \operatorname{Re}\left(\mathbf{p}^* \cdot \boldsymbol{\mathcal{H}}_{\text{sca}}\right) - \frac{1}{Z} \operatorname{Re}\left(\mathbf{m}^* \cdot \boldsymbol{\mathcal{E}}_{\text{sca}}\right) \right\}$$

[16] M. Nieto-Vesperinas, *Phil. Trans. R. Soc. A*, 375(2090):20160314, 2017

Magnetodielectric Dimer

- emission enhancement in dimer
- 9-dimensional parameter space
- optimization of objective function
  $\widetilde{\mathbf{x}} = \text{argmin}_{\mathbf{x}\in\mathcal{X}} f(\mathbf{x})$

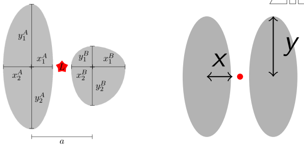


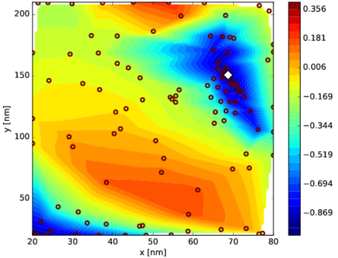
- Bayesian optimization: stochastics based on previous results

[17] P.-I. Schneider et al., In *Digital Optical Technologies 2017*, volume 10335, page 103350O, 2017

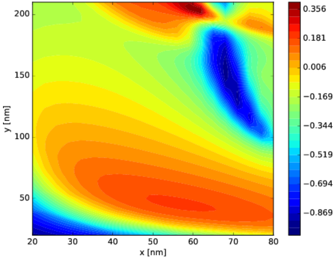
Magnetodielectric Dimer

- emission enhancement in dimer
- 9-dimensional parameter space
- optimization of objective function
  $\widetilde{\mathbf{x}} = \text{argmin}_{\mathbf{x} \in \mathcal{X}} f(\mathbf{x})$
- Bayesian optimization: stochastics based on previous results
- example: mirror symmetric dimer ($x_A = x_B, y_A = y_B$ and fixed a)



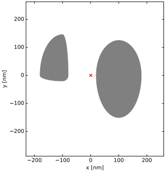
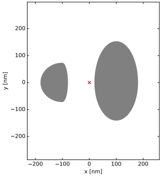
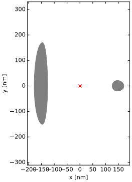
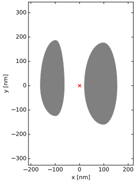


optimization of helicity $h = \frac{X_{\text{ext}}}{kW_{\text{ext}}}$ of (−) dipole



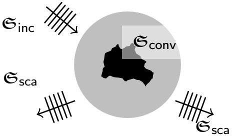
[17] P.-I. Schneider et al., In *Digital Optical Technologies 2017*, volume 10335, page 103350O, 2017
[18] P. Gutsche et al., In *Journal of Physics: Conference Series*, volume 963, page 012004. IOP Publishing, 2018

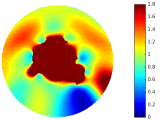
Optimal Designs

				
goal	F_P contrast	F_P contrast and high F_P^-	helicity $h = \frac{X_{\text{ext}}}{kW_{\text{ext}}}$: $(-)$ enhancement and $(+)$ annhilation	$(-)$ helicity enhancement and high F_P^-
$f(\mathbf{x})$	$g_P = 2\frac{F_P^+ - F_P^-}{F_P^+ + F_P^-}$	$\left\{\frac{1}{4}g_P + \frac{1}{2}\right\} + \frac{1}{F_P^- + 1}$	$g_{\text{far}} = 2(\lvert h^+\rvert - \lvert h^-\rvert)$	$\left\{\frac{1}{4}g_{\text{far}} + \frac{1}{2}\right\} + \frac{1}{F_P^- + 1}$
$g_{P/\text{far}}(\tilde{\mathbf{x}})$	-1.34	-1.28	-1.54	-1.36
F_P^+	1.65	2.09	0.88	1.66
F_P^-	8.38	9.56	1.45	3.57

Optical Chirality for Classification of Scatterers

- conserved optical chirality for detailed scattering response
- chiral quantities in Mie Theory
- classification based on incident light of well-defined helicity
- scalar quantification of properties of particles:
  - duality breaking
  - anti-duality breaking
  - helicity-annihilation breaking
  - helicity variation
- averaged circular dichroism c and g-factor
- helicity enhancement factor for optimization of scatterers





enhanced chirality $\mathrm{Re}\,(\mathfrak{X})$ by dual scatterer

Thank you!

Conservation of Optical Chirality

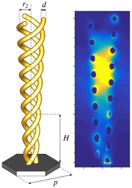
- *illumination*: both states of circularly polarized light (CPL)
- differential CPL excitation of molecule $\Delta A = 2\omega \, \mathrm{Re}\,(\alpha_{em})\, \mathfrak{X}$
- time-averaged optical chirality density $\mathfrak{X}$ in free space

$$\mathfrak{X} = -\frac{\varepsilon_0 \omega}{2} \,\mathrm{Im}\,(\boldsymbol{\mathcal{E}}^* \cdot \boldsymbol{\mathcal{B}}) \qquad \mathfrak{X} \propto |\mathbf{A}^+|^2 - |\mathbf{A}^-|^2 \text{ with CPL amplitudes } \mathbf{A}^\pm$$

- conservation law

$$2i\omega\,(\mathfrak{X}_\mathrm{e} - \mathfrak{X}_\mathrm{m}) + \nabla \cdot \mathfrak{S} = \frac{1}{4}\left\{\boldsymbol{\mathcal{J}}^* \cdot (\nabla \times \boldsymbol{\mathcal{E}}) + \boldsymbol{\mathcal{E}} \cdot (\nabla \times \boldsymbol{\mathcal{J}}^*)\right\}$$

(cf. conservation of energy: $2i\omega(\mathcal{U}_\mathrm{e} - \mathcal{U}_\mathrm{m}) + \nabla \cdot \boldsymbol{\mathcal{S}} = -\frac{1}{2}\boldsymbol{\mathcal{J}}^* \cdot \boldsymbol{\mathcal{E}}$)



- optical chirality flux density $\mathfrak{S}$
- rhs for localized source: $\frac{1}{2}\omega \boldsymbol{\mathcal{J}}^* \cdot \boldsymbol{\mathcal{B}}$

electric $\mathfrak{X}_\mathrm{e}$ and magentic $\mathfrak{X}_\mathrm{m}$ chirality

$$\mathfrak{S} = 1/4\left\{\boldsymbol{\mathcal{E}} \times (\nabla \times \boldsymbol{\mathcal{H}}^*) - \boldsymbol{\mathcal{H}}^* \times (\nabla \times \boldsymbol{\mathcal{E}})\right\}$$

$$\mathfrak{X}_\mathrm{e} = 1/8\left\{\boldsymbol{\mathcal{D}}^* \cdot (\nabla \times \boldsymbol{\mathcal{E}}) + \boldsymbol{\mathcal{E}} \cdot (\nabla \times \boldsymbol{\mathcal{D}}^*)\right\}, \mathfrak{X}_\mathrm{m} = 1/8\left\{\boldsymbol{\mathcal{H}}^* \cdot (\nabla \times \boldsymbol{\mathcal{B}}) + \boldsymbol{\mathcal{B}} \cdot (\nabla \times \boldsymbol{\mathcal{H}}^*)\right\}$$

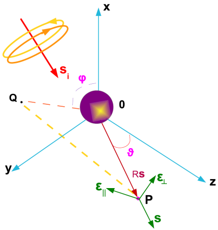
[4] P. Gutsche et al., In *SPIE 9756*, page 97560X, 2016, arXiv:1603.05011 [5] L. V. Poulikakos et al., *ACS Photonics*, 3(9):1619–1625, 2016

Helicity Optical Theorem (HOT)

- scatterer (*molecule*) as bi-isotropic dipolar particle

$$\mathbf{p} = \alpha_e \boldsymbol{\mathcal{E}}_{\text{inc}} - \alpha_{em} \boldsymbol{\mathcal{B}}_{\text{inc}}$$
$$\mathbf{m} = \alpha_{me} \boldsymbol{\mathcal{E}}_{\text{inc}} + \alpha_m \boldsymbol{\mathcal{B}}_{\text{inc}}$$

(chiral: $\alpha_{em} = -\alpha_{me}$)



- standard optical theorem for energy: $W_{\text{ext}} = W_{\text{sca}} + W_{\text{abs}}$

$$\frac{\omega}{2} \operatorname{Im}\left(\mathbf{p} \cdot \boldsymbol{\mathcal{E}}^*_{\text{inc}} + \mathbf{m} \cdot \boldsymbol{\mathcal{B}}^*_{\text{inc}}\right) = \frac{ck^4}{3n} \left\{ \frac{1}{\varepsilon} |\mathbf{p}|^2 + \mu |\mathbf{m}|^2 \right\} + \mathcal{U}_{\text{abs}}$$

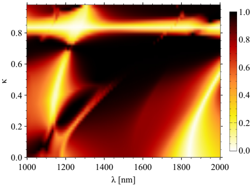
- **helicity** optical theorem for chirality: $X_{\text{ext}} = X_{\text{sca}} + X_{\text{conv}}$

$$\boxed{\frac{2\pi c}{\mu} \operatorname{Re}\left(-\frac{1}{\varepsilon} \mathbf{p} \cdot \boldsymbol{\mathcal{B}}^*_{\text{inc}} + \mu \mathbf{m} \cdot \boldsymbol{\mathcal{E}}^*_{\text{inc}} \right) = \frac{8\pi c k^3}{3\varepsilon} \operatorname{Im}\left(\mathbf{p} \cdot \mathbf{m}^*\right) + \mathfrak{X}_{\text{conv}}}$$

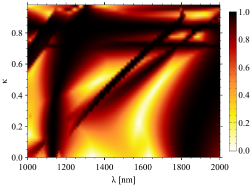
[8] M. Nieto-Vesperinas, *Phys. Rev. A*, 92(2):023813, 2015

Chiral Sphere

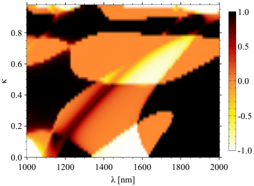
- refractive index $n = 3.5$, radius $r = 230$nm
- illumination-independent classification



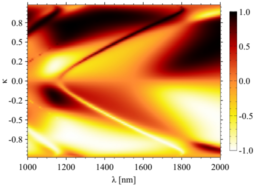
duality breaking $\not d$



helicity annihilation breaking $\not h_a$



helicity variation h



g-factor

T-Matrix Formalism

- general isolated scatterers described by T-matrix

$$T\begin{pmatrix}\mathbf{p}\\\mathbf{q}\end{pmatrix}=\begin{pmatrix}T_{ee} & T_{em}\\ T_{me} & T_{mm}\end{pmatrix}\begin{pmatrix}\mathbf{p}\\\mathbf{q}\end{pmatrix}=\begin{pmatrix}\mathbf{a}\\\mathbf{b}\end{pmatrix}$$

- scattering response given by matrices $\left(q = (\mathbf{pq})^T\mathbb{Q}(\mathbf{pq})\right)$

$$\begin{aligned}
\mathbb{W}_{\text{sca}} &= T^H T\\
\mathbb{X}_{\text{sca}} &= \frac{1}{2}\begin{pmatrix} T_{ee}^H T_{em} + T_{em}^H T_{ee} & T_{ee}^H T_{mm} + T_{em}^H T_{me}\\ T_{mm}^H T_{ee} + T_{me} T_{em}^H & T_{me}^H T_{mm} + T_{mm}^H T_{me}\end{pmatrix}\\
\mathbb{W}_{\text{ext}} &= \frac{1}{2}(T^H + T)\\
\mathbb{X}_{\text{ext}} &= \frac{1}{2}\begin{pmatrix} T_{em} + T_{em}^H & T_{ee}^H + T_{mm}\\ T_{ee} + T_{mm}^H & T_{me} + T_{em}^H\end{pmatrix}
\end{aligned}$$