Optical chirality enhancement with dielectric metasurfaces

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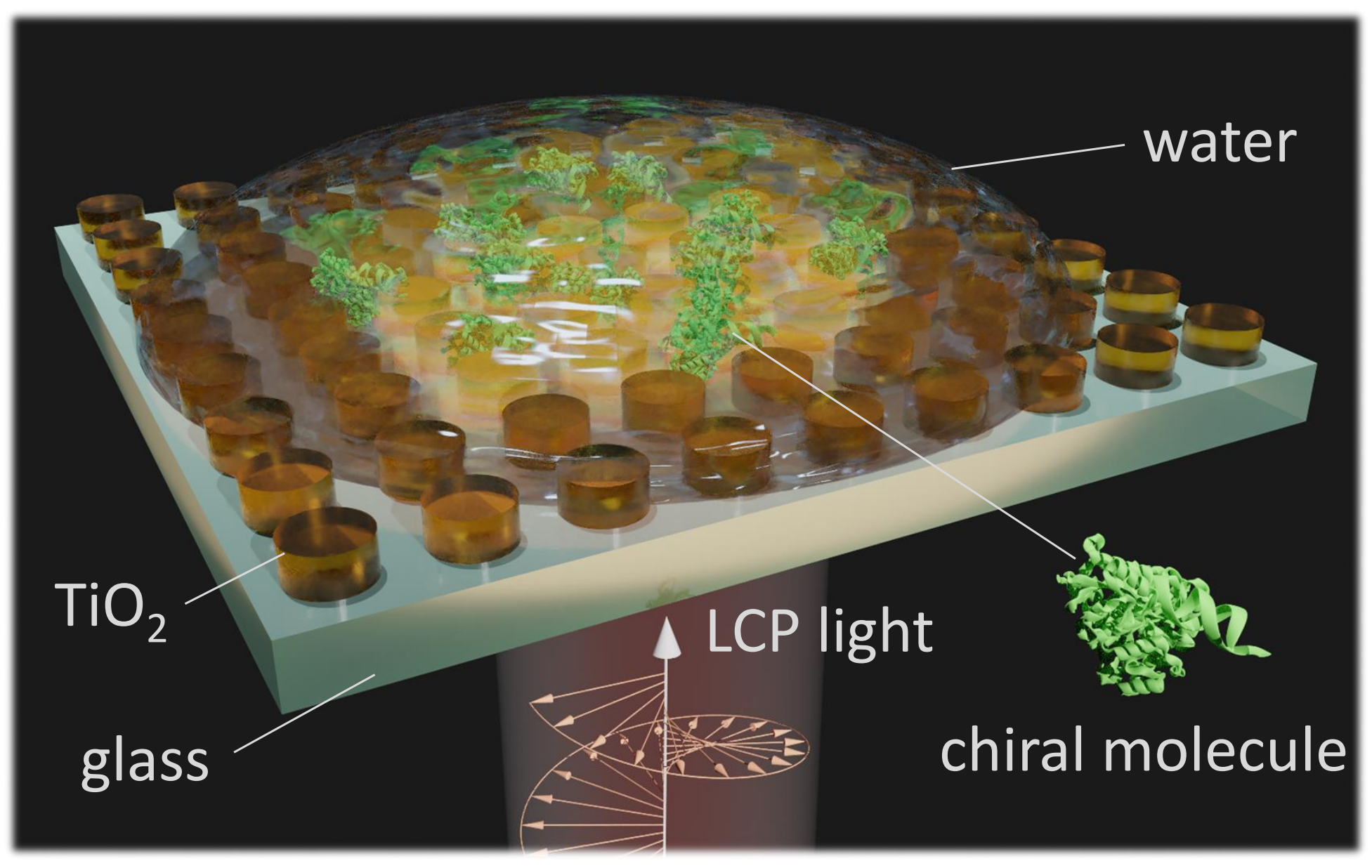
Motivation

Light-molecule interactions with metallic and dielectric nanostructures make it possible to effectively enhance their sensitivity. For example, right-handed (RCP) and left-handed (LCP) circularly polarized light interacts with chiral molecules in different ways that allow determining their handedness and structural information [1]. Circular dichroism (CD) spectroscopy (measures the different absorption of light from the left and right circular waves) is used as an effective tool to study chiral molecules. However, CD signal from molecules is very difficult to detect due to their weak internal chirality. These problems impose significant restrictions on the achievable measurement sensitivity. To enhance the CD response from low concentrations or even a single molecule, a new approach based on the formation of superchiral fields interacting with molecules to enhance their optical chirality has been used.



Optical chirality enhancement

The absorption of RCP (+) or LCP (−) light by a chiral molecule [2]:

$$A^{\pm} = \frac{\omega}{2}\left(\alpha''\left|\boldsymbol{E}^{\pm}\right|^2 + \chi''\left|\boldsymbol{B}^{\pm}\right|^2\right) \mp \frac{2}{\varepsilon_0} G'' C$$

non-magnetic enviroment $\downarrow$ $\chi''\left|\boldsymbol{B}^{\pm}\right|^2 \approx 0$

$$A^{\pm} = \frac{\omega}{2}\alpha''\left|\boldsymbol{E}^{\pm}\right|^2 \mp \frac{2}{\varepsilon_0} G'' C \longrightarrow C = -\frac{\varepsilon_0 \omega}{2} Im\left[\boldsymbol{E}^{*\pm} \cdot \boldsymbol{B}^{\pm}\right]$$

optical chirality

Circular dichroism:

$$CD \propto A^+ - A^- = \frac{\omega}{2}\alpha''\left(|\boldsymbol{E}^+|^2 - |\boldsymbol{E}^-|^2\right) - \frac{4}{\varepsilon_0} G'' C$$

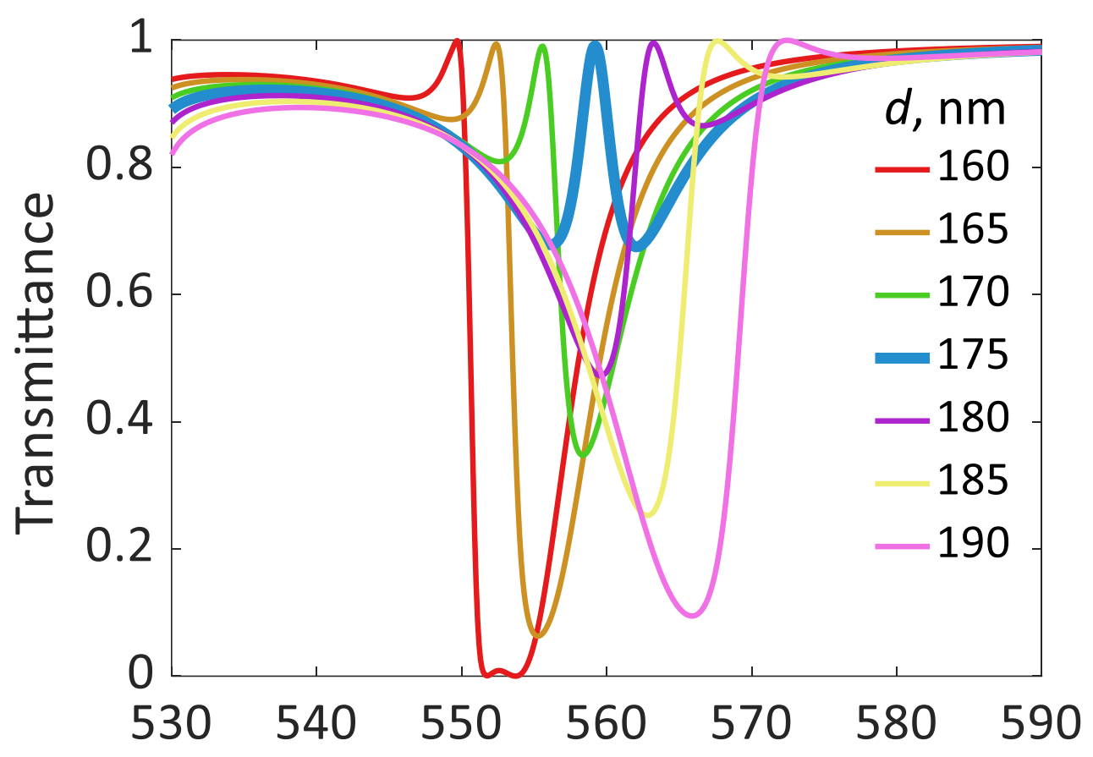
for achiral structure $\downarrow$ $|\boldsymbol{E}^+|^2 = |\boldsymbol{E}^-|^2$

$$CD \propto A^+ - A^- = \underbrace{-\frac{4}{\varepsilon_0}G''}_{const} C \longrightarrow CD = \frac{C}{C_{CPL}} \underbrace{CD_{CPL}}_{const}$$

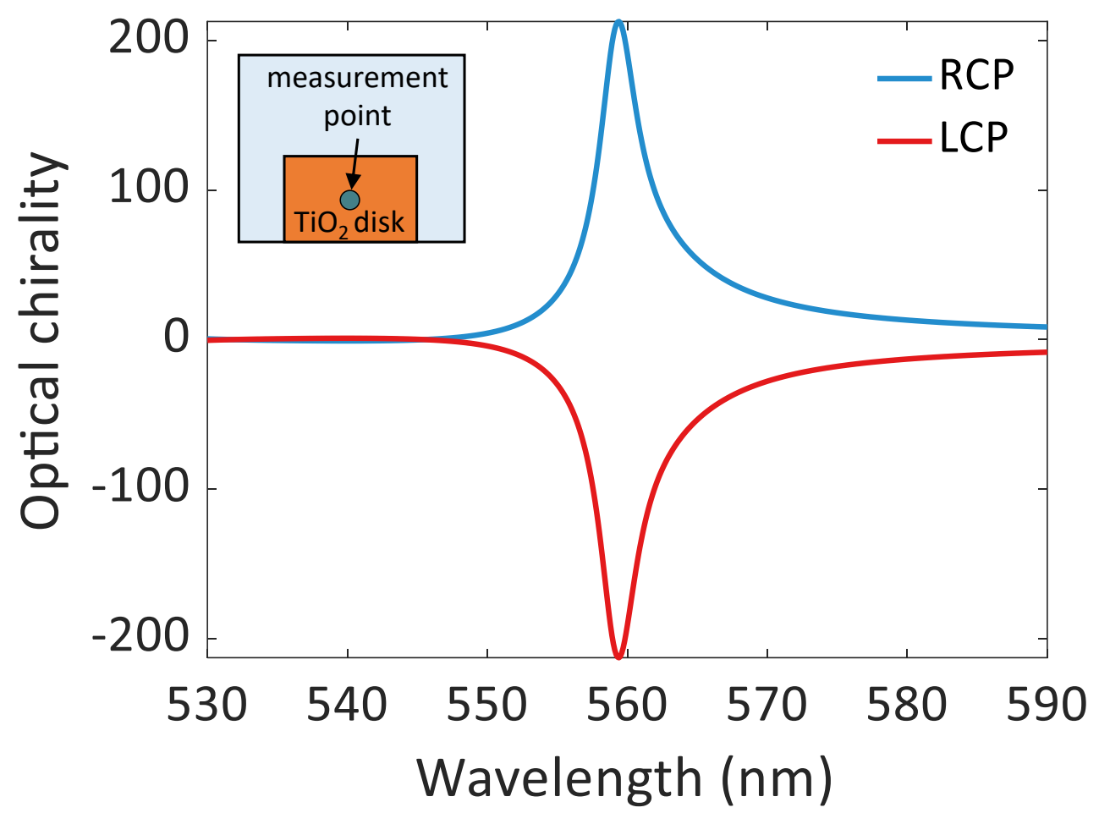
Decomposition of optical chirality [3]:

$$C/C_{CPL} = \underbrace{\Im\left(\frac{\boldsymbol{E_i}}{E_0} \cdot \frac{\boldsymbol{H_d^*}}{H_0}\right)}_{C(E_i, H_d)} + \underbrace{\Im\left(\frac{\boldsymbol{E_d}}{E_0} \cdot \frac{\boldsymbol{H_i^*}}{H_0}\right)}_{C(E_d, H_i)} + \underbrace{\Im\left(\frac{\boldsymbol{E_d}}{E_0} \cdot \frac{\boldsymbol{H_d^*}}{H_0}\right)}_{C(E_d, H_d)}$$

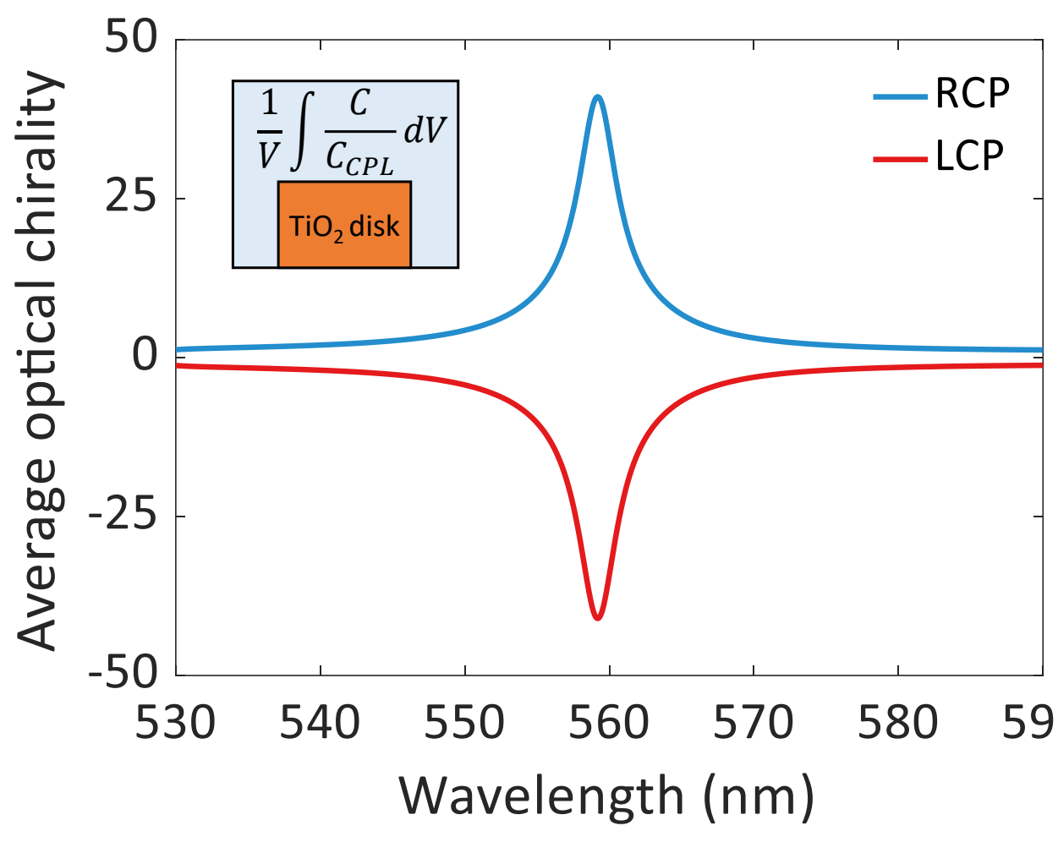
where $\boldsymbol{E_i}, \boldsymbol{H_i}$ are incident electric and magnetic field, $\boldsymbol{E_d}, \boldsymbol{H_d}$ are dipolar electric and magnetic fields.



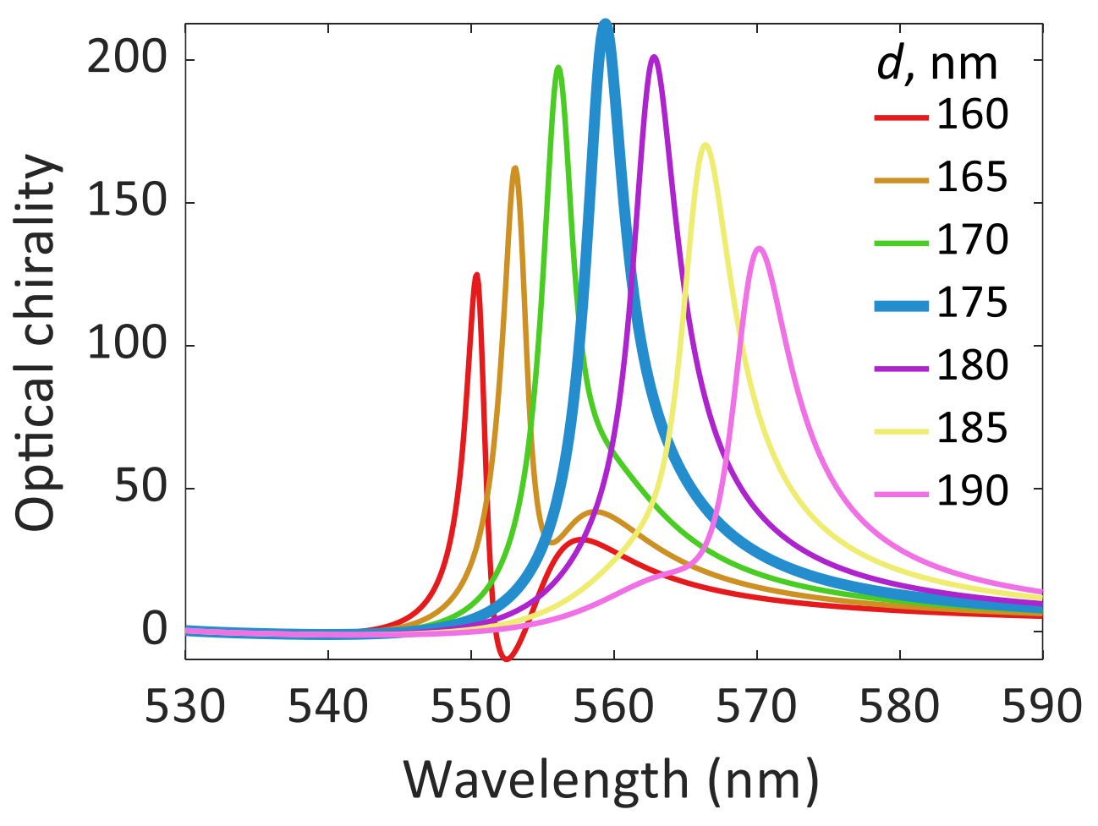
Transmittance at different diameter (d) of disk



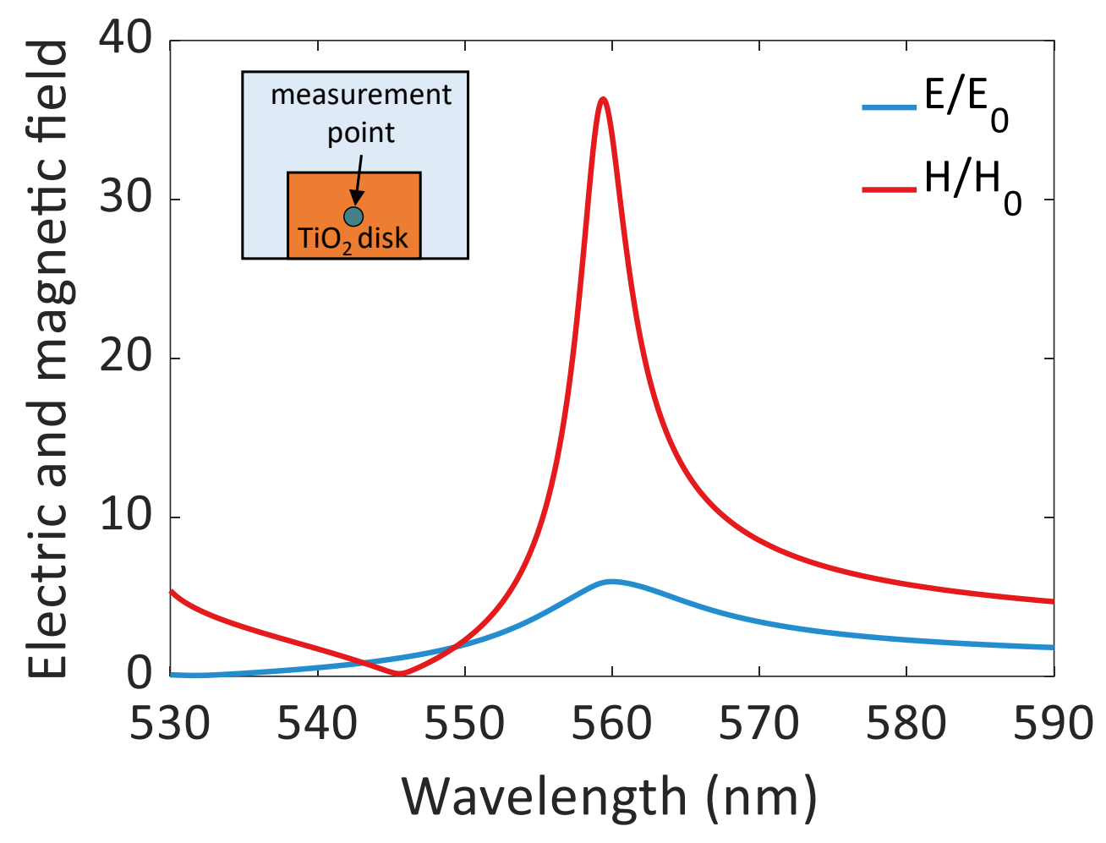
Optical chirality measured at the center of disk



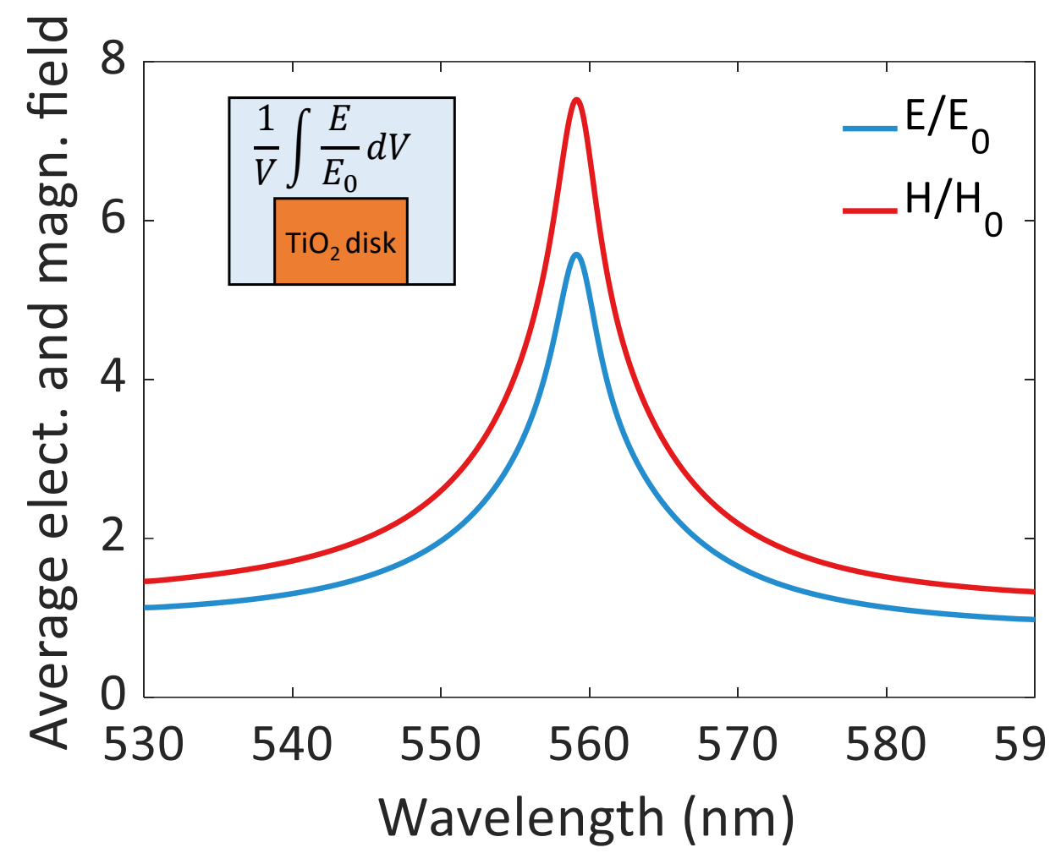
Integrated optical chirality by volume around disk



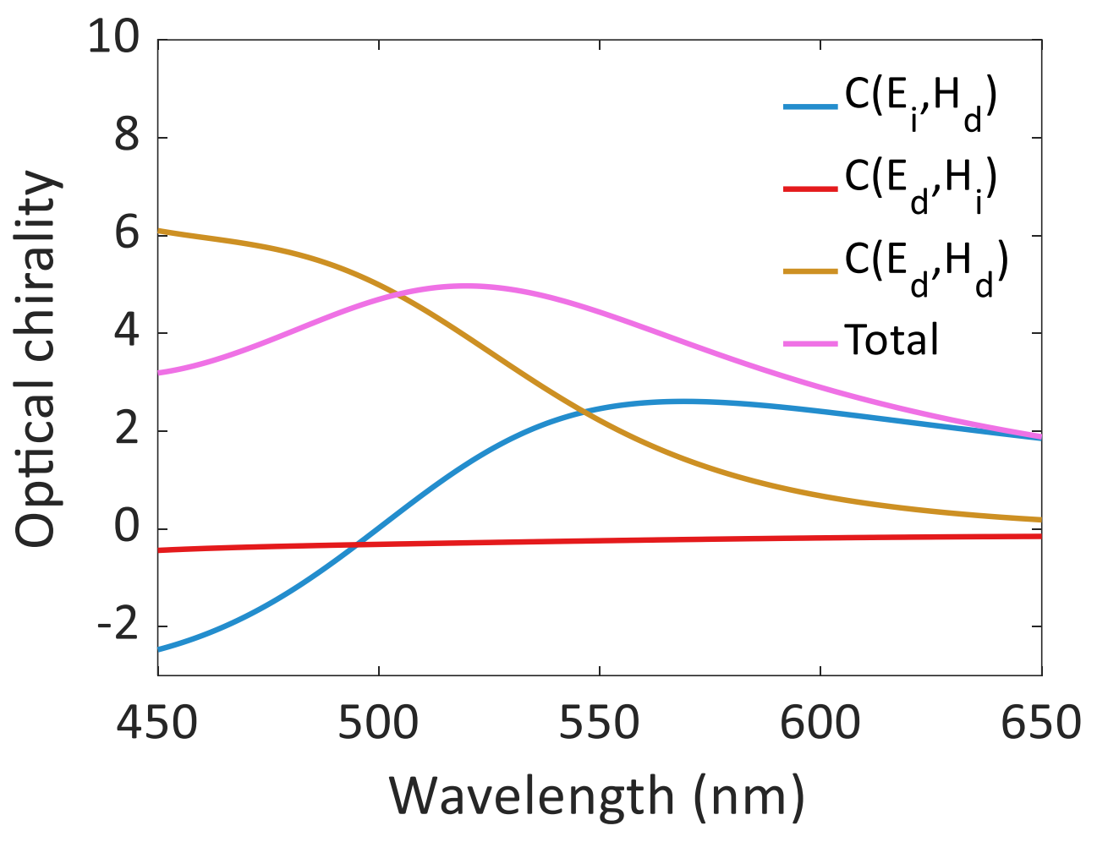
Optical chirality at various disk diameters



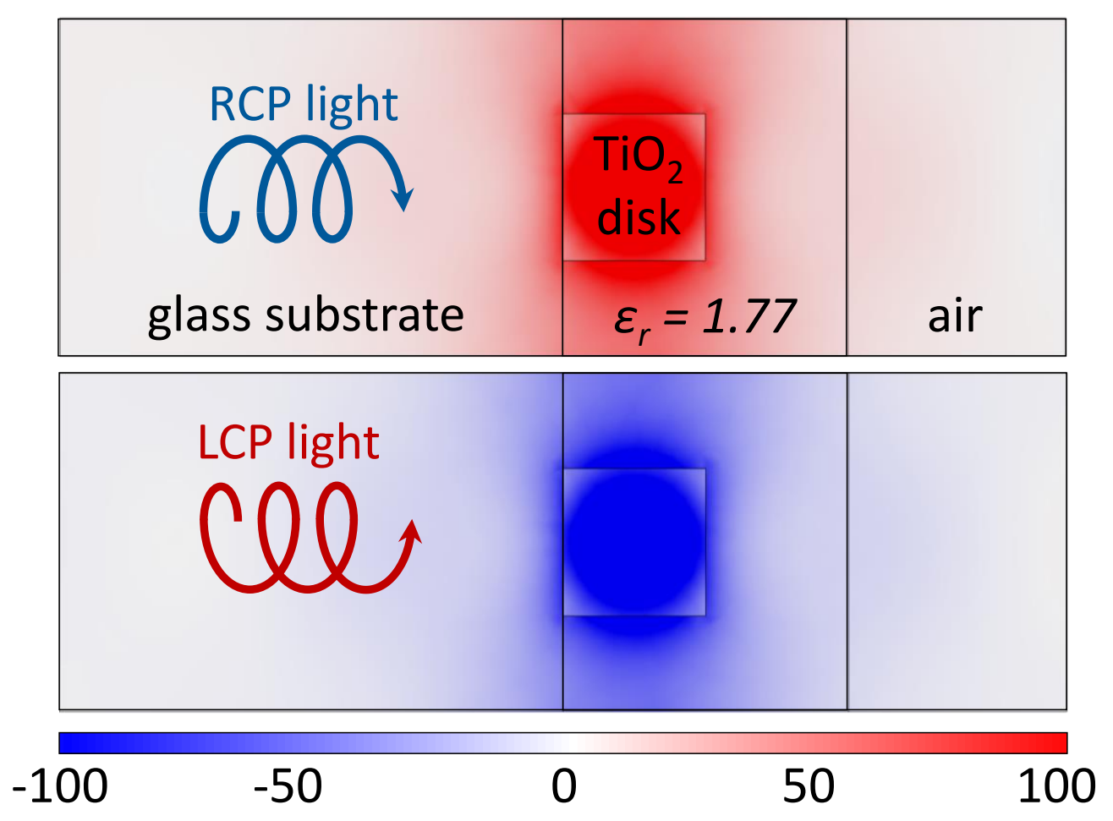
Electric and magnetic fields (at the center of disk)



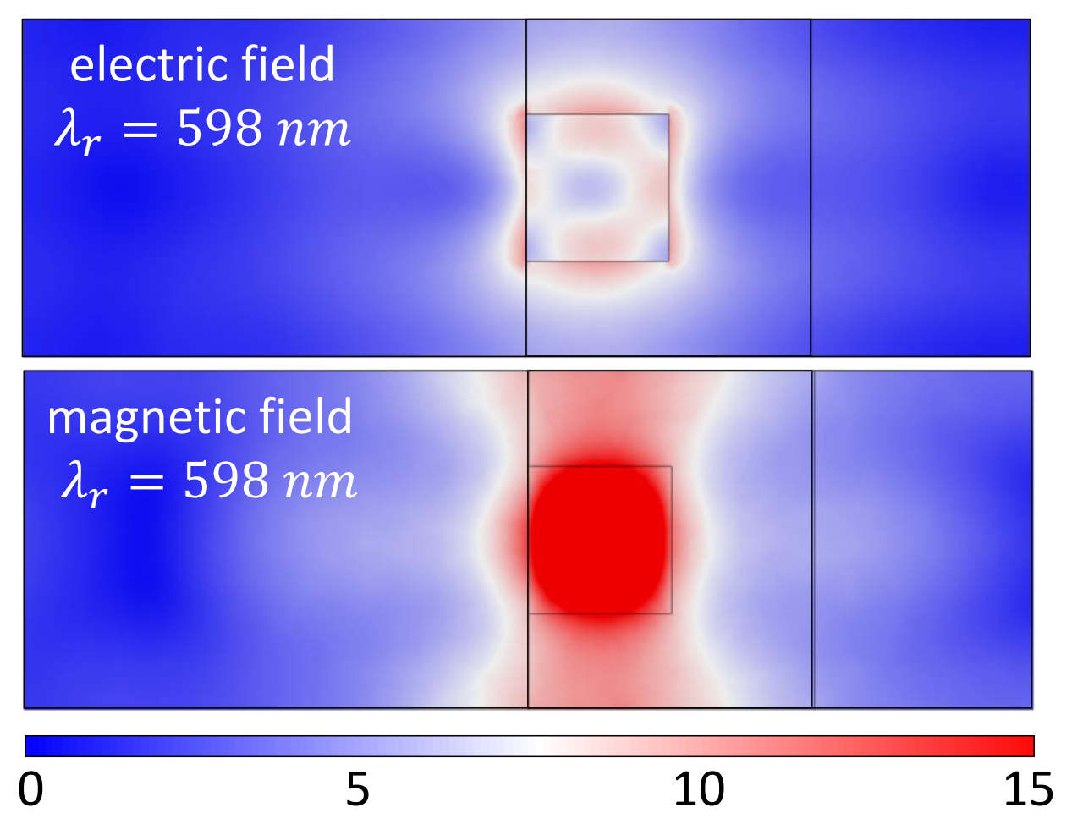
Average electric and magnetic fields (around disk)



Optical chirality decomposition of single TiO_2 disk



Optical chirality enhancement



Electric and magnetic fields enhancement

Modelling chiral medium

The constitutive relations:

$$\boldsymbol{D} = \varepsilon_0 \varepsilon_r \boldsymbol{E} + i\kappa \boldsymbol{H}/c$$
$$\boldsymbol{B} = \mu_0 \mu_r \boldsymbol{H} - i\kappa \boldsymbol{E}/c$$

The dispersion of chiral molecules [4]:

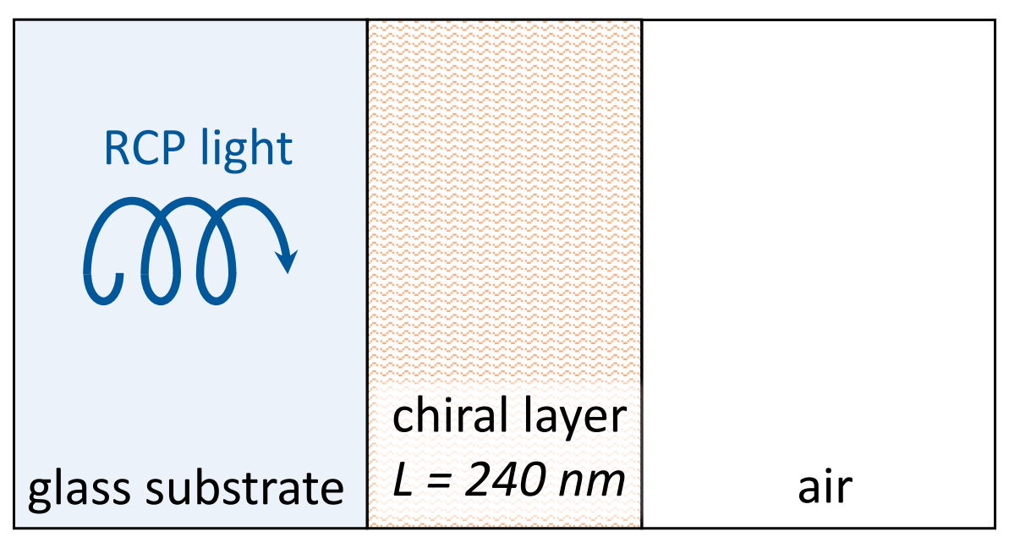
$$\varepsilon_{mol} = \varepsilon_r + Nd\omega_0\{f(\omega) + ig(\omega)\}$$
$$\kappa_{mol} = Nr\omega\{f(\omega) + ig(\omega)\}$$

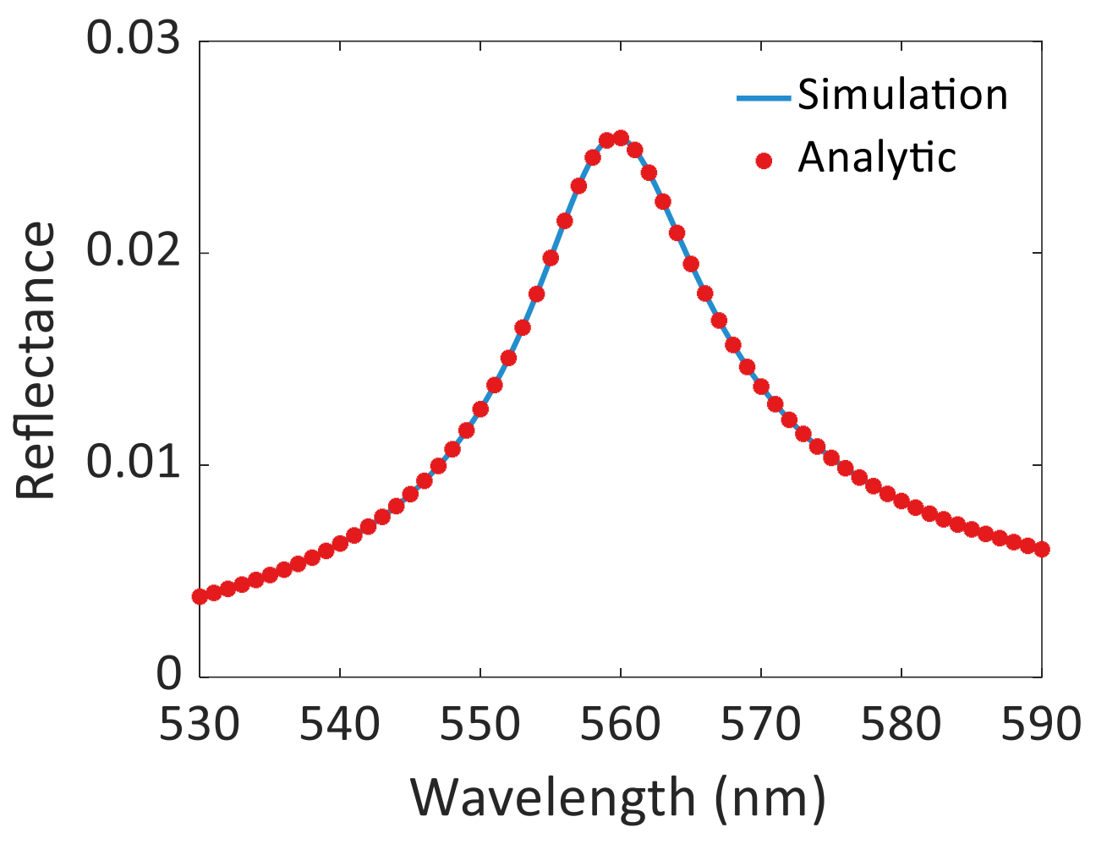
The dispersion and absorption line shape functions $f(\omega)$ and $g(\omega)$ are given by:

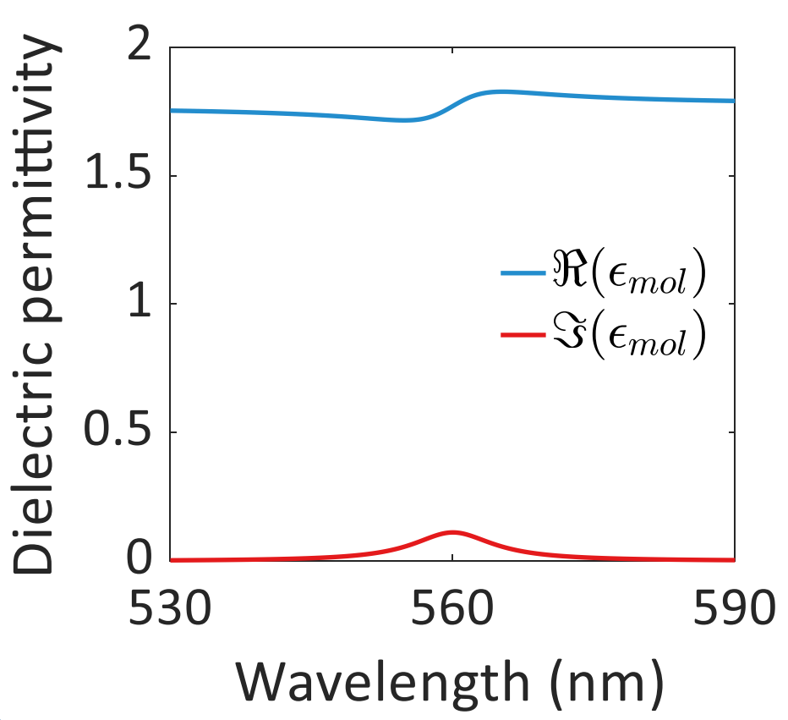
$$f(\omega) = \frac{\omega_0^2 - \omega^2}{\left(\omega_0^2 - \omega^2\right)^2 + \omega^2\Gamma^2}$$

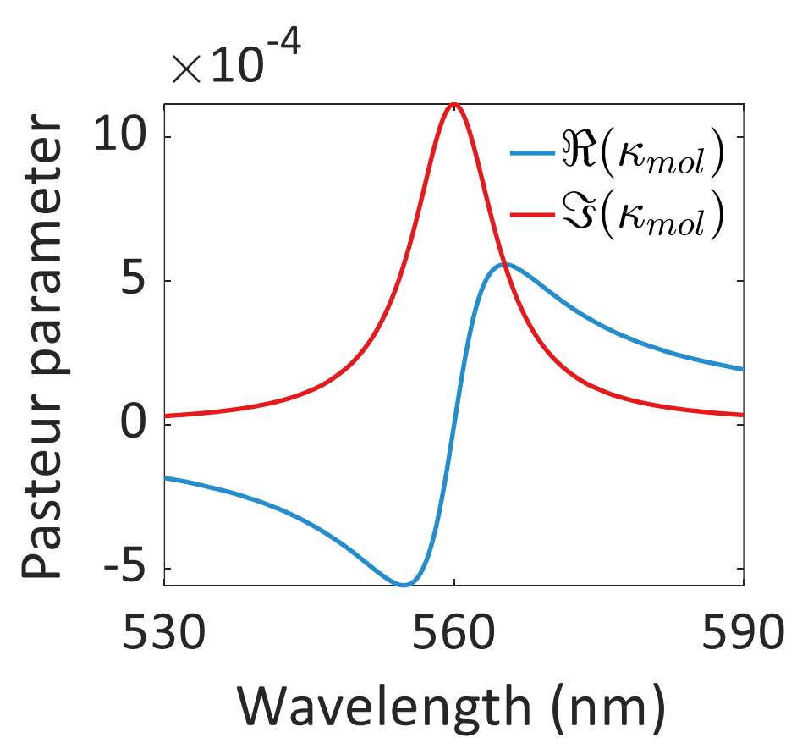
$$g(\omega) = \frac{\omega\Gamma}{\left(\omega_0^2 - \omega^2\right)^2 + \omega^2\Gamma^2}$$

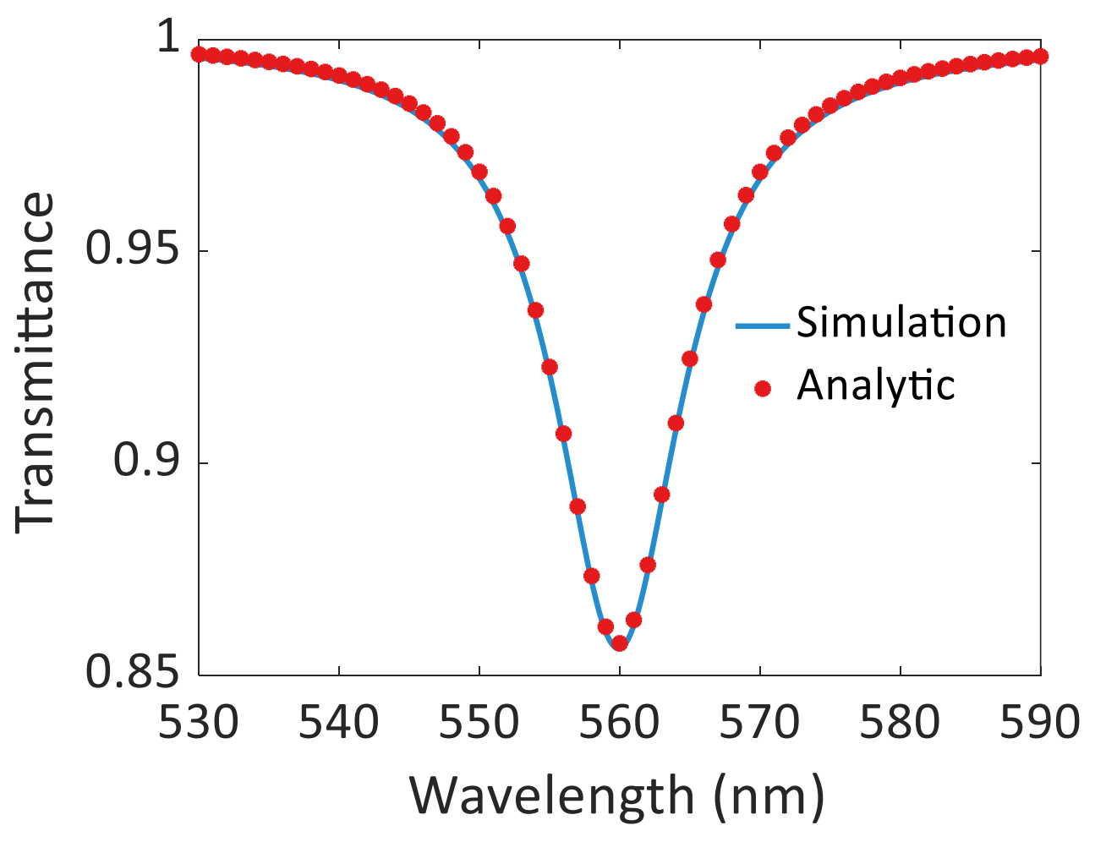
$\varepsilon_r = 1.77, \omega_0 = 2\pi c/560nm, \Gamma = 2\pi c/30\mu m,$
$Nd = 0.7 \times 10^{13}\ s^{-1}, Nr = 0.7 \times 10^{13}\ s^{-1}$



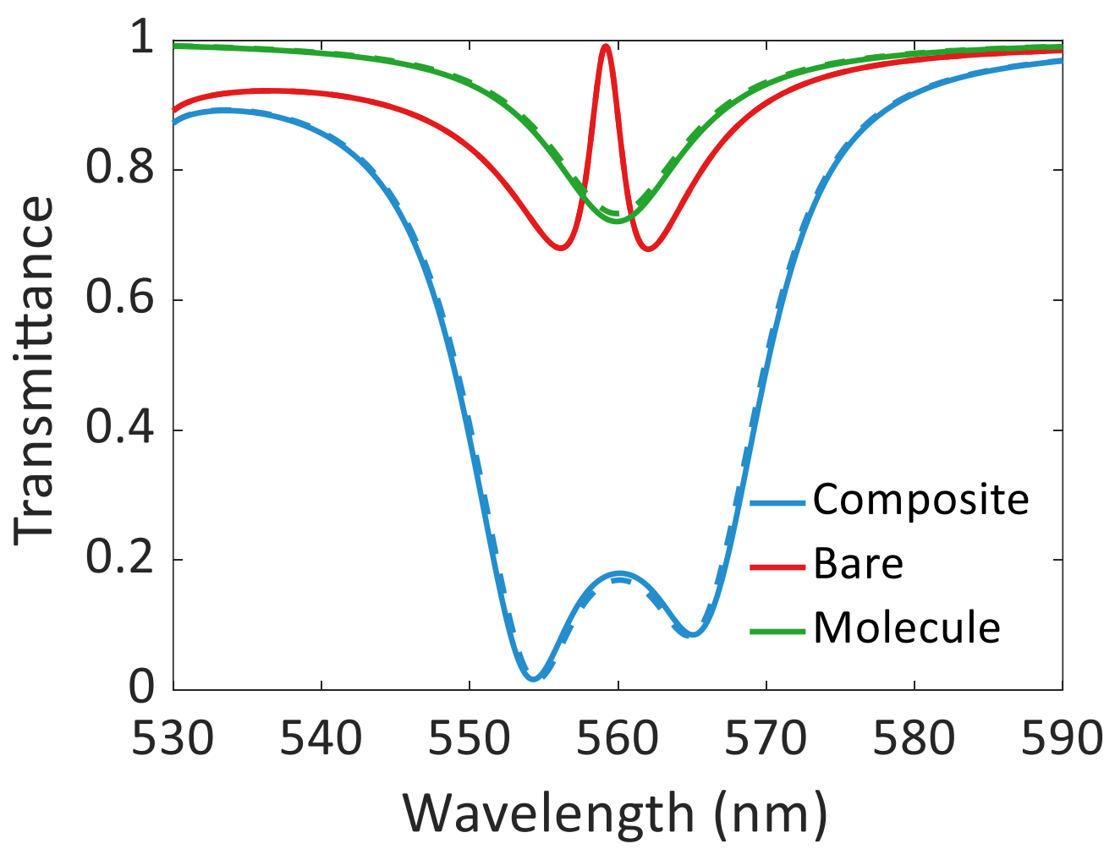




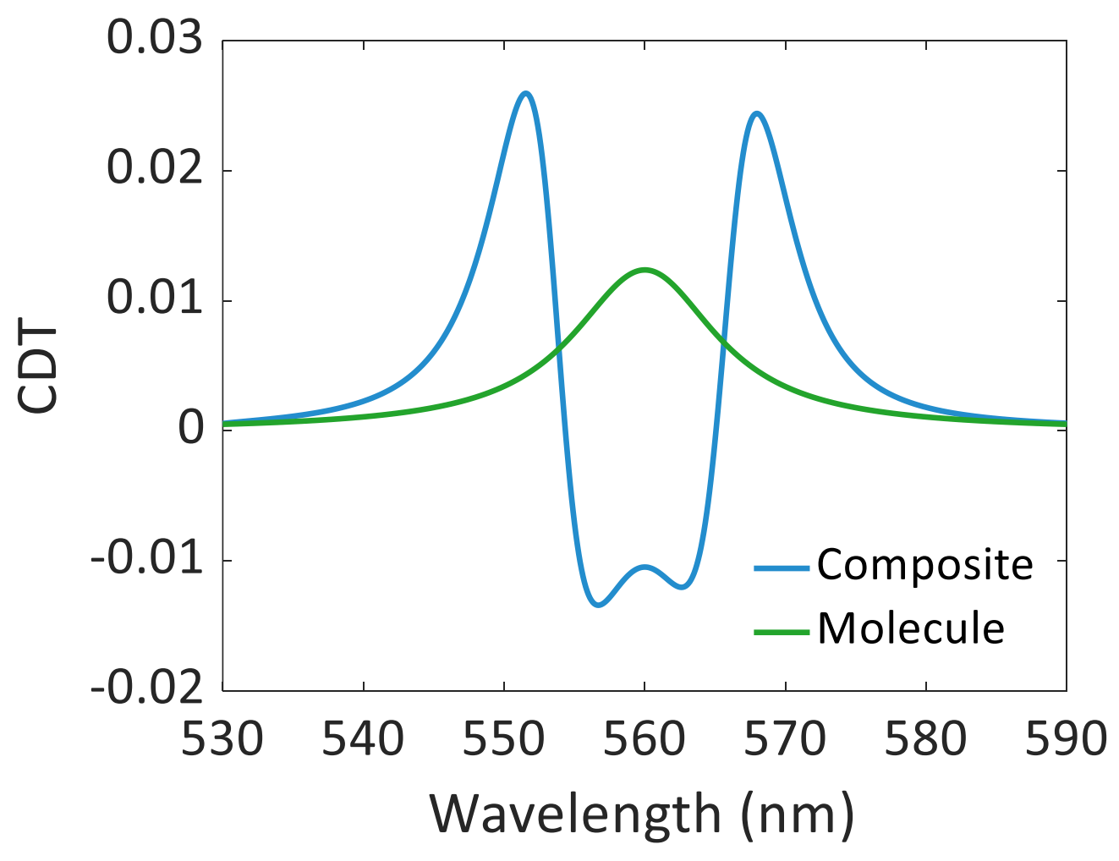




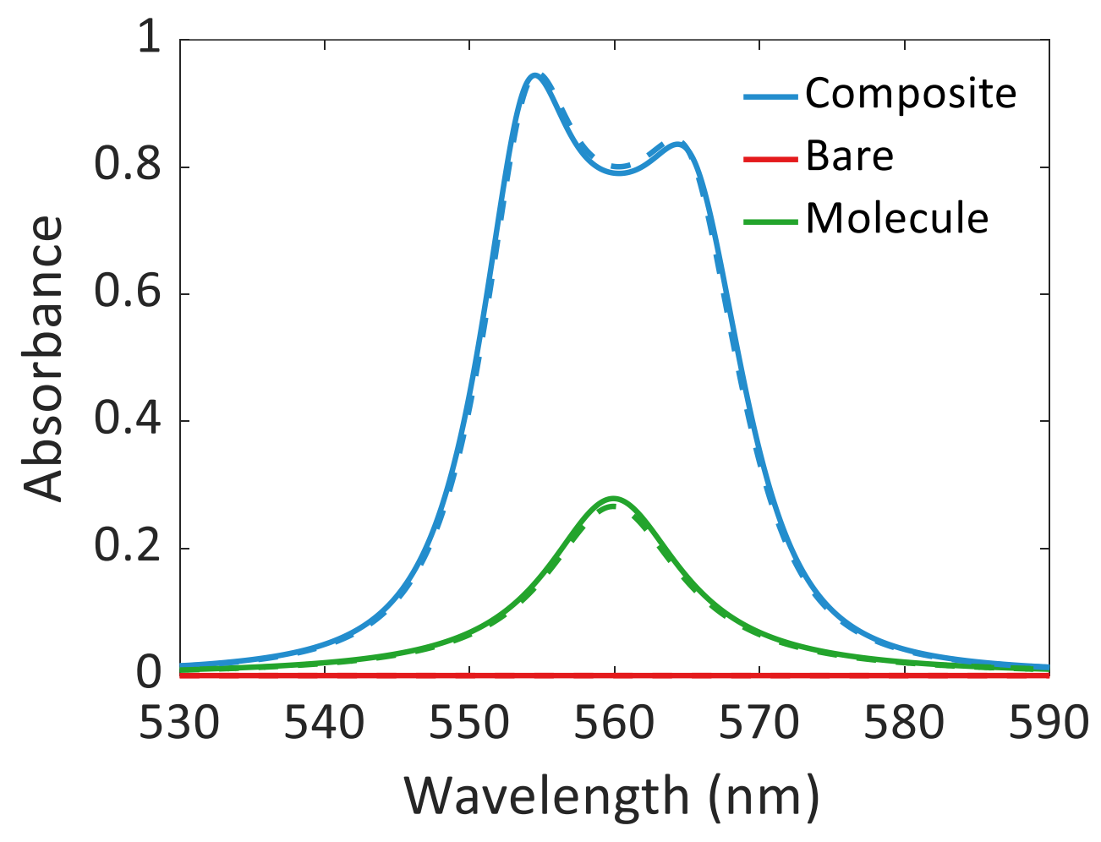
CD enhancement of chiral molecules



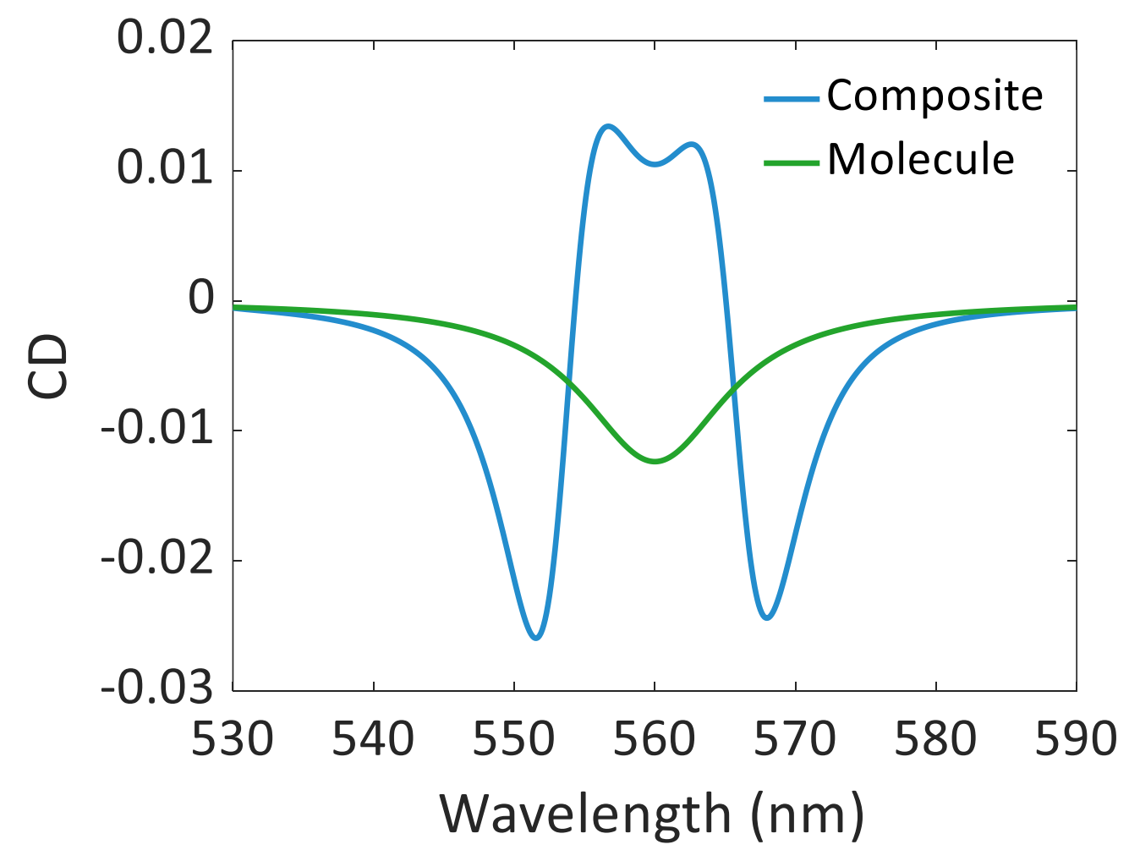
Transmittance for composite, bare and molecules



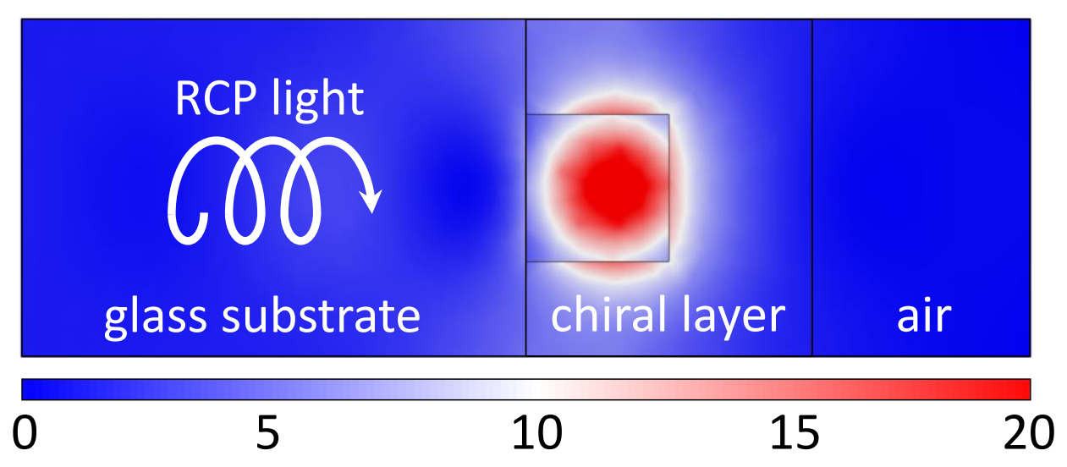
Chiral differential transmittance spectra



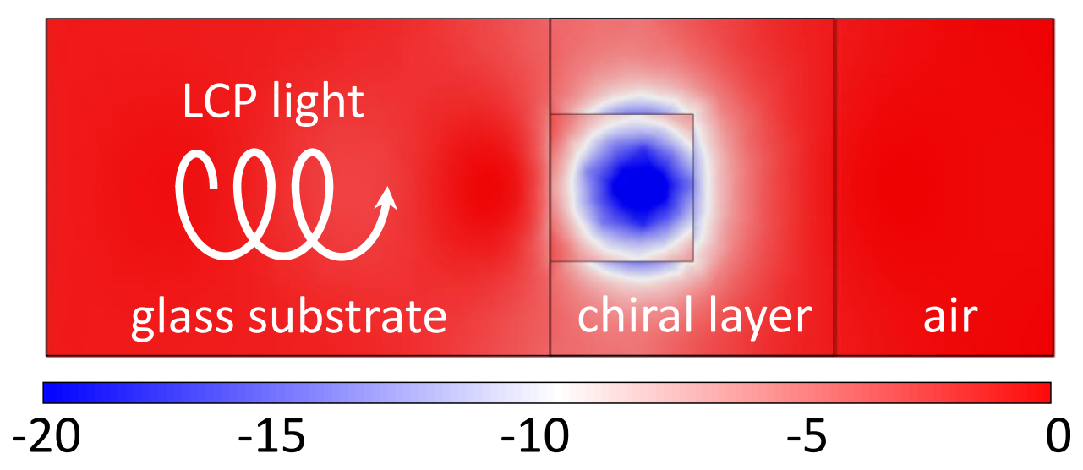
Absorption comparison for composite, bare and molecules



CD spectra of composite and chiral layer



Optical chirality enhancement



Optical chirality enhancement

Conclusion

- Optical chirality enhancement by about 200 folds
- The same sign of optical chiral field in the entire volume
- Implementation of the chiral medium in the simulation (COMSOL)
- CDT and CD enhancement of thin chiral molecular layer

References

[1] L. D. Barron, Cambridge University Press, Cambridge, England, 2004.
[2] Y. Tang, and A. E. Cohen *PRL*, 104, 163901 (2010).
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[4] K. Yao and Y. Liu *Nanoscale*, 10(18), 8779-8786 (2018).

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