Optimising Chiral Dissymmetry in Plasmonic Nanostructures

Confirmation Report

Joel T. Collins
Supervisor: V. K. Valev

Department of Physics
University of Bath

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Abstract

Nonlinear optical processes provide a valuable technique for probing properties of chiral structures, those lacking mirror symmetry. Such structures are encountered frequently throughout organic chemistry, pharmacology, and biology, with a recent growing emphasis on sensitive characterisation of small quantities of chiral molecules.

It has been shown that chiroptical effects such as circular dichroism are significantly enhanced in second-harmonic generation. Additionally, plasmonic nanostructures can create local regions of high intensity, highly chiral electromagnetic fields that further enhance chiroptical interactions with molecules doped onto the material surface.

In this report, chiroptical and nonlinear optical properties of dielectric materials are reviewed in the context of both natural molecules and plasmonic nanostructures. Current work is outlined exploring the modal composition of strong chiroptical responses in plasmonic nanostructures. Simulations relating to an ongoing experiment are then detailed, showing how diffraction can reveal additional information about a the chirality of a periodic array of nanostructures. Progress on a collaborative project studying chiral molecular aggregates is also outlined. Future work will look to develop comprehensive nonlinear characterisation of deep sub-wavelength scale chiral metamaterials.

Acknowledgements

I'd like to thank LaTeXfor not being as horrible to use as MS Word.

Notes to self:

SyncTeX should be installed separately to function in VS Code.
Install from choco (https://chocolatey.org/packages/synctex),
or download from TeXLive (https://www.tug.org/svn/texlive/trunk/Master/bin/win32/) then place synctex.exe and kpathsea***.dll in /Program Files/MiKTeX 2.9/miktex/bin/x64/.

To-do:

- Adjust fancy headers
- Actually write a thesis

Chapter 1

Background and Previous Work

1.1 Chirality and Chiroptical Effects

1.1.1 Chirality

Chirality is an important property pervasive throughout nature, found in organic chemistry, and hence key to biological organisms [1], as well as throughout physics in systems such as spiral galaxies, fluid vortices, and particle angular momentum to name just a few. Chirality is also frequently encountered in photonics, though chiral electromagnetic fields. At its simplest, this manifests as the circular polarisation of light. Here, two orthogonal linearly polarised waves of equal amplitude, wave vector and frequency, but with a relative phase shift of $\pm\pi/2$ are superimposed (Figure 1.1). The sign of this phase shift determines the handedness of circularly polarised light (CPL), with the wave equations for left-circularly polarised (LCP) and right-circularly polarised (RCP) given by equation 1.1 [2, §8.1.2]).

$$\begin{aligned} \mathbf{E}_{RCP} &= E_0 \left[\hat{\mathbf{x}} \cos(\mathbf{k}z - \omega t) + \hat{\mathbf{y}} \sin(\mathbf{k}z - \omega t)\right] \\ \mathbf{E}_{LCP} &= E_0 \left[\hat{\mathbf{x}} \cos(\mathbf{k}z - \omega t) - \hat{\mathbf{y}} \sin(\mathbf{k}z - \omega t)\right] \end{aligned} \tag{1.1}$$

Due to the abundance of chiral molecules in organic chemistry, biology, and pharmacology, there is a growing emphasis on the importance of accurate, sensitive, and accessible characterisation of such molecules. The use of chiral light in sensing applications has become an active field of current research, due to the flexibility of chiral photonics. However, a common fundamental mechanism exploited is chiral dissymmetry in the excitation of molecules. If a molecule couples differently to one circular polarisation than the other, it will experience a chiral dissymmetry in rate or phase of excitation, equivalent to a difference in the complex refractive index of the molecular material between LCP and RCP. This difference in excitation rate and phase delay results in measurable circular dichroism (CD) and optical rotation (OR) [3, §1].

1.2 Chiral Structures

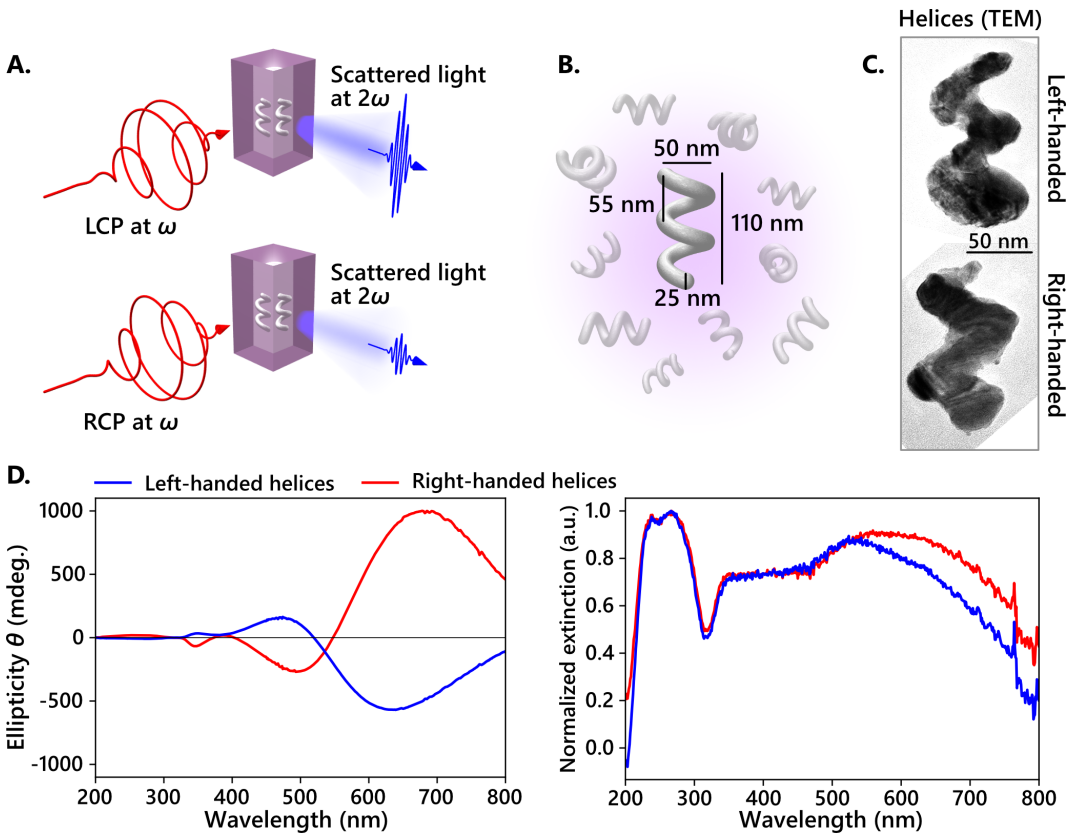

Figure 1.1: *A little test figure.*

Chapter 2

Experimental Techniques

Chapter 3

Experimental Results

Bibliography

[1] N. Berova, P. L. Polavarapu, K. Nakanishi, and R. W. Woody. *Comprehensive Chiroptical Spectroscopy*, volume 2. 2012.

[2] E. Hecht. *Optics: Pearson New International Edition*. Pearson Higher Ed USA, 4th edition, 2013.

[3] N. Purdie and H. Brittain. *Analytical Applications of Circular Dichroism*. Elsevier Science, 1993.

Appendix