

# Appendix A

## Units of Circular Dichroism

**Abstract** In chemical literature, different measures are evaluated when discussing the circular dichroism of a chiral analyte. We list the most important measures as a quick reference for the interpretation of circular dichroism spectra in this appendix.

In Sect. 2.2.3, the absorbance  $A$  is defined via the attenuation of the incident light beam. All the intensity of the incident beam is either transmitted, reflected, or absorbed. Therefore, one can write

$$1 = T + R + A \quad (\text{A.1})$$

with  $T$  being the transmittance and  $R$  the reflectance of the illuminated system. The differential absorbance is related to the differential transmittance via

$$\Delta A = -\Delta T - \Delta R. \quad (\text{A.2})$$

For an isotropic chiral medium described by the constitutive equations (2.48),  $\Delta R$  vanishes [1]. This is the common case for chiral liquids with isotropic orientation of the chiral molecules. For such systems, a measurement of  $\Delta T$  is sufficient to obtain information about  $\Delta A$ .

Artificial chiral media, such as chiral plasmonic nanostructures, cannot be described by absorption alone; scattering might play an important role as well [2].<sup>1</sup> Their response is, in general, anisotropic. Non-zero  $\Delta R$  can occur when the eigenpolarizations differ from CPL.<sup>2</sup> One must be careful when such anisotropic media are analyzed regarding their CD response via transmission measurements.<sup>3</sup>

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<sup>1</sup>There exist several techniques to describe chiral metamaterials via effective constitutive parameters [3-5]. This allows, in principle, to predict the chiroptical response without a full-field calculation of the underlying structure. However, a thorough optical characterization of the metamaterial is necessary for the parameter retrieval. Note that only structures much smaller than the wavelength can be described by an effective medium.

<sup>2</sup>This is the main mechanism of the helix-based circular polarizer discussed in [6]. A thorough theoretical discussion of the behavior of multiple helices is given in [7].

<sup>3</sup>Circular eigenpolarizations can be ensured via the arrangement. They are supported by structures with  $C_3$ ,  $C_4$ , or  $C_6$  rotational symmetry under normal incidence [8]. For all other symmetries, one should carefully check for differential reflectance.

In some chemical literature, CD is not defined via the differential absorbance  $\Delta A$ . Instead, they use Beer's law (in base 10 notation), which describes the attenuation of the light intensity  $I$  in the system:

$$I = I_0 10^{-\epsilon l c_{\text{mol}}}, \quad (\text{A.3})$$

where  $\epsilon$  is the molar extinction coefficient,  $l$  is the path length, and  $c_{\text{mol}}$  is the concentration of the chiral analyte in moles per liter. Then, one can identify the absorptivity  $\mathcal{A}$  as

$$\mathcal{A} := \epsilon l c = \log \left( \frac{I_0}{I} \right). \quad (\text{A.4})$$

CD is then defined as the differential absorptivity:

$$\Delta \mathcal{A} := \mathcal{A}^+ - \mathcal{A}^- = \log \left( \frac{I^-}{I^+} \right). \quad (\text{A.5})$$

This definition has the advantage that no reference beam is needed for the measurement [9].

Sometimes, also the difference of the extinction coefficients is given, because it is independent on path length and concentration. A transformation between the two definitions can be obtained by

$$\Delta \epsilon = \frac{1}{l c_{\text{mol}}} \Delta \mathcal{A}. \quad (\text{A.6})$$

In some chemical literature, the measured CD spectra do not report  $\Delta \mathcal{A}$  or  $\Delta \epsilon$  but ellipticity  $\theta$  (sometimes also  $\Psi$ ), which is defined as

$$\theta(\text{rad}) = \frac{\sqrt{I^-} - \sqrt{I^+}}{\sqrt{I^-} + \sqrt{I^+}}. \quad (\text{A.7})$$

Comparison with Beer's law leads to

$$\theta(\text{rad}) \approx \Delta \mathcal{A} \frac{\ln 10}{4} \quad (\text{A.8})$$

for small values of  $\Delta \mathcal{A}$ . Here,  $\theta$  is given in radians. A conversion to millidegrees results in

$$\theta(\text{mdeg}) \approx 32982 \cdot \Delta \mathcal{A}, \quad (\text{A.9})$$

which is a commonly used unit for CD spectra. The molar ellipticity  $[\theta]$  is given as

$$[\theta] = \frac{0.1 \cdot \theta(\text{mdeg})}{l c_{\text{mol}}} \approx 3298.2 \cdot \Delta \epsilon. \quad (\text{A.10})$$

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## Appendix B

# Details of the Simulations

**Abstract** All discussions of the chiral near-field response of plasmonic nanostructures in this book are based on numerical simulations. Depending on the demands, either an in-house implementation of the Fourier modal method or a commercially available finite element method code (CST Microwave Studio) has been used. In this appendix, we discuss the capabilities and limitations of both tools. Additionally, we provide further information regarding the calculation and normalization of optical chirality in CST Microwave Studio.

### B.1 Simulation Tools

We analyzed the chiral plasmonic near-field sources by numerical full-field calculations. These calculations are performed by two different tools. Firstly, a commercially available frequency domain solver using the finite element method, CST Microwave Studio (MWS) [1]. The second tool is an in-house implementation of the so-called Fourier modal method.<sup>4</sup>

This so-called Fourier modal method is well-suited for the simulation of systems with stacked periodic layers. It has been optimized for fast convergence by application of Fourier-factorization rules and adaptive spatial resolution for the interfaces between the nanostructures and the dielectric background [2]. The implementation is very versatile and can be adjusted for specific problems. For instance, the chiral constitutive equations (2.48) have been implemented. To obtain this, we extended the formulation of Onishi and collaborators [3] to bi-periodic gratings. Our implementation includes the correct factorization rules for chiral media in accordance to those of achiral media [4] to ensure optimum convergence. Therefore, not only chiral geometries but also effective chiral media can be efficiently simulated, which is not possible with most commercial software.

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<sup>4</sup>This method is also referred to as “rigorous coupled-wave analysis (RCWA)” in literature.

One drawback of the Fourier modal method approach is that it is restricted to stacked systems. Fully three-dimensional geometries cannot be tackled with this approach. Additional constraints to the geometry are due to the adaptive spatial resolution that can only be applied to a given set of structures, where the necessary coordinate transformations have been formulated. For all other geometries, the respective transformations must be found first, which might be very time-consuming [5]. Therefore, we use CST MWS for such more complex geometries. The underlying finite element method meshes the structure with a tetrahedral grid. In principle, it can be applied to arbitrary structures, but computing time and memory consumption increase for complex geometries.

The calculations in Chaps. 5 and 6 have been performed with CST MWS. The Fourier modal method has been used for the simulations described in Chap. 7. All these designs have been calculated in a periodic arrangement because most nanofabrication techniques result in a surface patterned with the desired structure. Additionally, single particle experiments are more complicated. We chose the size of the unit cells large enough that no near-field coupling is observed (except for the locally chiral plasmonic lattices, where coupling of all building blocks is crucial for the concept). As a consequence, the plasmonic modes are localized to single structures. We expect the respective results to be applicable to single structures as well.

In Sect. 8.2, single structure calculations have been performed because of the size and the complexity of the suggested design. Such calculations can only be performed with CST MWS because the Fourier modal method inherently requires a periodic arrangement. We have used plane wave illumination with perfectly matched layer boundary conditions.

A specialty are the calculations performed in Sect. 8.3. The geometry can be simulated with the Fourier modal method. However, it doesn't support PEC media. Therefore, the PEC simulations have been performed with CST MWS. The simulations for gold, however, have been performed with the Fourier modal method because CST MWS doesn't allow to incorporate chiral media. Additionally, an inclined unit cell has been used for the Fourier modal method simulations, while a rectangular unit cell has been used for CST MWS. The size of the inclined unit cell was  $160 \text{ nm} \times (550 \text{ nm} / \cos \zeta)$  with an inclination angle of  $\zeta = -\tan^{-1}(180 \text{ nm}/250 \text{ nm})$ . Comparison with simulations performed in CST MWS with a rectangular unit cell lead to similar results.

All calculations have been carefully checked for convergence. Additionally, the correct representation of the geometry in the simulation tool must be taken care of. This is straightforward in the Fourier modal method: Due to the adaptive coordinates, every supported geometry is represented exactly in the method. However, the geometry is Fourier transformed, which induces truncation errors depending on the number of Fourier harmonics. For CST MWS, the quality of the mesh needs to be reviewed: Inaccuracies in the structure representation might have minor influence on far-field spectra, but can strongly disturb the near-field response [6]. In case of the calculations in Sect. 8.2, the automatic mesher had to be adjusted to realize round cross sections of the small wires.

**Table B.1** Intuitive handedness of the helix of the electric field vector in space and sign of OC density (calculated via (4.7)) for the different port modes in CST MWS. The second part of the table covers plane wave incidence for propagation in positive and negative  $z$ -direction

<i>Port</i>	<i>Helix</i>	<i>OC</i>
Zmin(1)	right	+
Zmin(2)	left	–
Zmax(1)	left	–
Zmax(2)	right	+
LCP (+ $z$ )	right	+
RCP (+ $z$ )	left	–
LCP (– $z$ )	right	+
RCP (– $z$ )	left	–

## B.2 Calculation of Normalized Optical Chirality in CST Microwave Studio

For the calculation of local OC in CST MWS, several differences must be taken into account. While we use the detector’s view convention, CST MWS uses the source view convention. Additionally, CST MWS uses the following time dependence for monochromatic plane waves

$$\mathbf{E} \propto e^{\hat{i}(\omega t - kz)} \quad (\text{B.1})$$

instead of (2.4). Therefore, the intuitive handedness of the helix of the electric field vector in space does not coincide with the nominal handedness of the light in CST MWS. However, the intuitive handedness is of major importance for our qualitative analysis of the behavior of geometrically chiral plasmonic nanostructures, e.g., the discussion of selectively excited modes in Born-Kuhn type structures. We give a mapping of the modes to the respective helices for Floquet ports<sup>5</sup> as well as plane wave incidence in Table B.1.

In addition, the difference in the time dependence results in a sign flip of the calculated values of OC density compared to the values obtained in (4.28) for CPL. Table B.1 shows the signs of OC obtained for the different ports. To be consistent with the conventions used in this book, the signs of local OC values obtained from CST MWS must be inverted.

The calculation of  $\hat{C}$  requires the values of  $E_0$ , which is the amplitude of the incident electric field, for the normalization (cf. (4.34)):

$$\hat{C} = \frac{\mu_0 c \Im(\mathbf{E}^* \cdot \mathbf{H})}{E_0^2}. \quad (\text{B.2})$$

<sup>5</sup>These ports are used by the frequency domain solver with unit cell boundary conditions.

Note that we both included the sign flip for  $C$  and accounted for the fact that CST MWS provides the magnetic field strength  $\mathbf{H}$  instead of the magnetic field  $\mathbf{B}$ .

For plane wave excitation, the amplitude of the electric field vector can be configured. Therefore, calculation of the normalized values is straightforward. Per default, CST MWS assumes  $E_0 = 1 \text{ V m}^{-1}$  for LPL and one obtains

$$\hat{C} = \frac{\mu_0 c}{1 \text{ V m}^{-2}} \Im(\mathbf{E}^* \cdot \mathbf{H}) \approx 376.730 \text{ m}^2 \text{ W}^{-1} \cdot \Im(\mathbf{E}^* \cdot \mathbf{H}). \quad (\text{B.3})$$

For CPL, both amplitudes  $E_0 J_x$  and  $E_0 J_y$  are set to  $1 \text{ V m}^{-1}$  per default. Therefore, one obtains  $E_0^2 = 2 \text{ V}^2 \text{ m}^{-2}$  and the normalization reads

$$\hat{C} \approx 188.365 \text{ m}^2 \text{ W}^{-1} \cdot \Im(\mathbf{E}^* \cdot \mathbf{H}). \quad (\text{B.4})$$

For Floquet ports, the whole port provides an input peak power of  $1 \text{ W}$ . Therefore, the absolute value of the Poynting vector  $\mathbf{S}$  is given as

$$|\mathbf{S}| = \sqrt{\frac{\varepsilon_0}{\mu_0}} E_0^2 = \frac{1 \text{ W}}{A_p}, \quad (\text{B.5})$$

where  $A_p$  is the area of the port in  $\text{m}^2$ . Solving for  $E_0^2$  and inserting in (B.2) yields

$$\hat{C} = \frac{A_p}{1 \text{ W}} \cdot \Im(\mathbf{E}^* \cdot \mathbf{H}). \quad (\text{B.6})$$

### B.3 Issues of Non-Enantiomorphic Field Pairs

All analysis of novel chiroptical spectroscopy techniques discussed in this book was based on the concept of enantiomorphic near-fields. However, arbitrary fields could be used in principle. In this case, the respective absorption rates must be directly calculated using (4.18).

However, this would result in a loss of generality. Any results for  $\Delta a$  would depend on the parameters  $\alpha$ ,  $\chi$ , and  $\xi$  of the chiral analyte. Particularly, the proportionality  $\Delta \hat{a} \propto \Delta \hat{C}$  would not be valid any more.

In addition, the simulation becomes more demanding because  $\omega U_e$  and  $\omega U_b$  must be considered together with  $C$ . However, a comparison of (4.16) with (4.28) leads to the estimate that  $\omega U_e$  supersedes  $C$  by eight orders of magnitude. Simulations in free space provide an estimate for the relative numerical error, which is in the order of  $10^{-4}$  for the configurations we used. Therefore, it would be necessary to increase the accuracy of the simulations by several digits to calculate the absorption rate via (4.18). Otherwise, the chiral contribution due to  $C$  would be hidden in the numerical noise of the achiral contributions.

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## Appendix C

# Volume Rendering with POV-Ray

**Abstract** The distribution of chiral near-fields for different plasmonic nanostructures is visualized by three-dimensional volume renderings. In this appendix, we show how such renderings can be obtained using the free ray tracing software POV-Ray. We provide scripts that can be directly adapted to obtain similar images.

We used POV-Ray (version 3.6.1 with MegaPOV 1.2.1) to visualize our results. We followed the guide given in [1]. The volume rendering was obtained by a transparent medium whose density and color was influenced by the local value of the OC as shown in Listing C.1.

At first, a cube with unity side length is created. This is necessary because media statements fill only this region, independent of the dimensions of the object. The box is rescaled afterwards to fit the dimensions of the exported fields.

The values are given via `df3` files. These are binary files with a simple file format [2]: At first, a header of three 16 bit unsigned integers describing the length of the three dimensions of the data set is needed. After this header, the data is expected as stream of unsigned integers in column major order, i.e., the first dimension varies the fastest. The size of the single integers is determined by the total size of the file. Note that all values are expected in big-endian byte order, while most Intel systems normally use little-endian encoding.<sup>6</sup> Therefore, the encoding must be set manually.

The simplified MATLAB code in Listing C.2 stores the negative values of a three-dimensional array in a `df3` file. A similar script can be used for the positive values. The values are rescaled to the maximum absolute value occurring in the initial data set. Of course, this can be changed to scale several data sets to the same value for comparison. Note that MATLAB stores arrays in column major order as well. Therefore, the array can be written to the file without further treatment.

---

<sup>6</sup>Big-endian and little-endian refer to different orderings of the bytes in multibyte values. More details can be found in [3].

```

1  box {
2    <0,0,0>, <1,1,1>
3    pigment { rgbt 1 }
4    interior {
5      media {
6        intervals 100
7        samples 1,20
8        emission 1/50
9        absorption 1/10
10       density {
11         density_file df3 "values-negative.df3"
12         interpolate 1
13         color_map { CMAP_NEGATIVE }
14       }
15     }
16     media {
17       /* same statement, but with
18        values-positive.df3 and CMAP_POSITIVE */
19     }
20   }
21   hollow
22   translate <-0.5,-0.5,-0.5>
23   /* sizes must be set to the dimensions
24   of the exported data in the df3 files */
25   scale <SIZE_X, SIZE_Y, SIZE_Z>
26 }

```

**Listing C.1** POV-Ray statements to render transparent media with color and density controlled by numerical values.

```

1  /* assumes values to save given in 'Data'
2
3  % prepare output file
4  out = fopen('values-negative.df3', 'w', 'ieee-be');
5
6  % header
7  fwrite(out, size(Data), 'uint16', 0, 'ieee-be');
8
9  % rescale values
10 Data_df3 = uint32((2^32-1)*abs(Data)/max(abs(Data(:))));
11
12 % delete positive values
13 Data_df3(Data > 0) = 0;
14
15 % output
16 fwrite(out, Data_df3, 'uint32', 0, 'ieee-be');
17 fclose(out);

```

**Listing C.2** MATLAB code to save a given array in a df3 file for use in POV-Ray.

For the scaling, values that exceed the absolute value of OC in all data sets that should be compared have been used. However, numerical noise can lead to extraordinary high values. Those unphysical responses would destroy the scaling. Therefore, we filtered all data with a  $3 \times 3 \times 3$  median filter. This is a nonlinear filter that is used for noise reduction in image processing. It is the optimal filtering technique to remove single outstanding values.

We use two media statements in the POV-Ray code because `df3` files can only contain unsigned values. Therefore, we separated positive and negative values into different files. It would be possible to rescale the whole range to positive values. However, the density is lowest for the value 0, which would result in minimal medium density for the strongest negative OC.

The color of each pixel is controlled by the respective color map. We used the maps in Listing C.3. Note that, due to the `absorption` keyword, the colors given in the color map have to be inverted. `<1, 1, 0>`, for example, absorbs red and green resulting in the blue color for negative values. The second entry in each color map controls the value up to which the OC is not shown at all. This is necessary because the incident light can also carry non-zero OC, which should not be visualized. Otherwise, the whole domain would be colored. Dismissing these values results in no loss of information because we are only interested in enhanced values above the OC of CPL. We used different cutting values depending on the normalization.

For most plots displaying OC, the cutting value 0.025 has been used. This allows for a simple comparison of the respective designs. However, the plots in Chap. 8 have been rendered with 0.075; additionally, a value of 0.6 instead of 0.5 was used in the third line of the color maps. Figure 7.2 was rendered using a very small cutting value of 0.003 due to the logarithmic scale used in this plot. The plots of the electric energy density enhancement were rendered with 0.05 (Fig. 5.2) and 0.01 (Fig. 7.1b).

```

1 #declare CMAP_NEGATIVE = color_map {
2   [0 rgb 0]
3   [0.025 rgb 0]
4   [0.5 rgb <1,1,0>]
5   [1 rgb <3,0,0>]
6 }
7
8 #declare CMAP_POSITIVE = color_map {
9   [0 rgb 0]
10  [0.025 rgb 0]
11  [0.5 rgb <0,1,1>]
12  [1 rgb <0,0,3>]
13 }

```

**Listing C.3** POV-Ray color maps to obtain the color scheme used for volume rendering.

Due to the transparency, the color of one distinct point depends not only on the actual OC value at this point but also on the color of the region behind. Therefore, the plots should not be used for quantitative analysis. However, this is a general problem of all volume rendering tools. The resulting plots are only useful to obtain information about the general shape and location of the chiral near-fields, which is important for the general design of chiral near-field sources. We provide slice plots for quantitative analysis wherever necessary.

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