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Supporting Information

Computation of Electromagnetic Properties of Molecular Ensembles**

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Supplemental Information for: Computation of electromagnetic properties of molecular ensembles

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I. CONNECTION

In this appendix we will prove Eq. (6) in the main text, which connects the T-matrix setting

$$\begin{bmatrix} c^\omega_{1-1} \\ c^\omega_{10} \\ c^\omega_{11} \\ d^\omega_{1-1} \\ d^\omega_{10} \\ d^\omega_{11} \end{bmatrix} = \begin{bmatrix} \underline{\underline{T}}^\omega_{NN} & \underline{\underline{T}}^\omega_{NM} \\ \underline{\underline{T}}^\omega_{MN} & \underline{\underline{T}}^\omega_{MM} \end{bmatrix} \begin{bmatrix} a^\omega_{1-1} \\ a^\omega_{10} \\ a^\omega_{11} \\ b^\omega_{1-1} \\ b^\omega_{10} \\ b^\omega_{11} \end{bmatrix}, \tag{S1}$$

to the polarizability tensor setting

$$\begin{bmatrix} \mathbf{p}^\omega \\ \mathbf{m}^\omega \end{bmatrix} = \begin{bmatrix} \underline{\underline{\alpha}}^\omega_{pE} & \underline{\underline{\alpha}}^\omega_{pH} \\ \underline{\underline{\alpha}}^\omega_{mE} & \underline{\underline{\alpha}}^\omega_{mH} \end{bmatrix} \begin{bmatrix} \mathbf{E}^\omega(\mathbf{0}) \\ \mathbf{H}^\omega(\mathbf{0}) \end{bmatrix}, \tag{S2}$$

and allows to build the T-matrix of a molecule to dipolar order using data obtained from quantum chemistry molecular simulations.

We will start from Eq. (S1) and transform it towards Eq. (S2). We begin by connecting the $\{a^\omega_{1m}, b^\omega_{1m}\}$ to $\{\mathbf{E}^\omega(\mathbf{0}), \mathbf{H}^\omega(\mathbf{0})\}$. The bottom line is that the $\{a^\omega_{1m}, b^\omega_{1m}\}$ are essentially the coordinates of $\{\mathbf{E}^\omega(\mathbf{0}), \mathbf{H}^\omega(\mathbf{0})\}$ in the spherical vector basis.

Let us start by writing expressions for the multipolar fields of well defined parity $\mathbf{M}^\omega_{jm}(\mathbf{r})$ and $\mathbf{N}^\omega_{jm}(\mathbf{r})$:

$$\begin{aligned} \mathbf{M}^\omega_{jm}(\mathbf{r}) =& j_j(k^\omega r)\mathbf{T}_{jjm}(\hat{\mathbf{r}}), \\ \mathbf{N}^\omega_{jm}(\mathbf{r}) =& \frac{\nabla \times \mathbf{M}^\omega_{jm}(\mathbf{r})}{k^\omega} = -i\sqrt{\frac{j}{2j+1}} j_{j+1}(k^\omega r)\mathbf{T}_{jj+1m}(\hat{\mathbf{r}}) + \\ & i\sqrt{\frac{j+1}{2j+1}} j_{j-1}(k^\omega r)\mathbf{T}_{jj-1m}(\hat{\mathbf{r}}). \end{aligned} \tag{S3}$$

where $k^\omega = \omega\sqrt{\epsilon^\omega \mu^\omega}$ is the frequency dependent wavenumber, $r = |\mathbf{r}|$, $\hat{\mathbf{r}} = \mathbf{r}/|\mathbf{r}|$, $j_l(\cdot)$ are spherical Bessel functions, and $\mathbf{T}_{jlm}(\hat{\mathbf{r}})$ are the vector spherical harmonics as defined in [1, Eq. (16.88)]. Importantly, the spherical Bessel functions contain all the radial dependence of the multipolar fields, and the $\mathbf{T}_{jlm}(\hat{\mathbf{r}})$ all their angular dependence. The second equality in the second line of Eq. (S3) follows from [1, Eq. (16.100)] and the relationships between spherical Bessel functions in [2, p. 172].

At the origin ($\mathbf{r} = \mathbf{0}$) the spherical Bessel functions $j_l(\cdot)$ are all zero except when $l = 0$: $j_0(0) = 1$. It follows that from all the multipolar fields in Eq. (S3), only the ones containing $j_0(0)$ are non-zero at the origin. Since $j = 1, 2, \ldots$, only electric dipolar fields

$$\mathbf{N}^\omega_{1m}(\mathbf{0}) = i\sqrt{\frac{2}{3}}\mathbf{T}_{10m}(\hat{\mathbf{r}}), \tag{S4}$$

where $m = [-1, 0, 1]$ are non-zero at the origin. Using [1, Eq. (16.88)] and a table of Clebsch-Gordan coefficients we obtain

$$\mathbf{T}_{10-1}(\hat{\mathbf{r}}) = \frac{\hat{\mathbf{e}}_{-1}}{\sqrt{4\pi}},\ \mathbf{T}_{100}(\hat{\mathbf{r}}) = \frac{\hat{\mathbf{e}}_{0}}{\sqrt{4\pi}},\ \mathbf{T}_{101}(\hat{\mathbf{r}}) = \frac{\hat{\mathbf{e}}_{1}}{\sqrt{4\pi}}. \tag{S5}$$

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where $\{\hat{\mathbf{e}}_{-1}, \hat{\mathbf{e}}_{0}, \hat{\mathbf{e}}_{1}\}$ are the spherical vector basis. We write a vector $\mathbf{w}$ in the spherical vector basis as:

$$\mathbf{w} = w_{-1}\hat{\mathbf{e}}_{-1} + w_0\hat{\mathbf{e}}_{0} + w_1\hat{\mathbf{e}}_{1}, \tag{S6}$$

with

$$\hat{\mathbf{e}}_{-1} = \frac{\hat{\mathbf{x}} - i\hat{\mathbf{y}}}{\sqrt{2}},\ \hat{\mathbf{e}}_{1} = -\frac{\hat{\mathbf{x}} + i\hat{\mathbf{y}}}{\sqrt{2}},\ \hat{\mathbf{e}}_{0} = \hat{\mathbf{z}}. \tag{S7}$$

This choice of basis induces the following relationship between the Cartesian and spherical coordinates of $\mathbf{w}$ in the spherical and Cartesian basis:

$$\begin{bmatrix} w_{-1} \\ w_0 \\ w_1 \end{bmatrix} = \begin{bmatrix} \frac{1}{\sqrt{2}} & \frac{i}{\sqrt{2}} & 0 \\ 0 & 0 & 1 \\ \frac{-1}{\sqrt{2}} & \frac{i}{\sqrt{2}} & 0 \end{bmatrix} \begin{bmatrix} w_x \\ w_y \\ w_z \end{bmatrix}. \tag{S8}$$

We can now use Eqs. (S4) and (S5) to write a more explicit version of Eq. (4) in the main text:

$$\begin{aligned} \mathbf{E}^\omega(0) &= i\sqrt{\frac{2}{12\pi}} \left[\sum_{m=-1}^{1} a^\omega_{1m}\hat{\mathbf{e}}_{\mathbf{m}}\right], \\ iZ^\omega\mathbf{H}^\omega(0) &= i\sqrt{\frac{2}{12\pi}} \left[\sum_{m=-1}^{1} b^\omega_{1m}\hat{\mathbf{e}}_{\mathbf{m}}\right], \end{aligned} \tag{S9}$$

which says that the a^ω_{1m} and b^ω_{1m} are respectively proportional to the coordinates of the incident electric and magnetic fields at the origin in the *spherical* vector basis coordinates.

We now turn to the left hand side of Eq. (S1). We need to relate the electric and magnetic dipole moments $\mathbf{p}^\omega$ and $\mathbf{m}^\omega$ to the multipolar coefficients c^ω_{1m} and d^ω_{1m} of their radiated fields. This task can be achieved using expressions from [3, Chap. 9]. The idea is to equate two different expressions of the magnetic(electric) field radiated by an electric(magnetic) dipole moment. One of the expressions involves $\mathbf{p}^\omega(\mathbf{m}^\omega)$ in [3, Eq. (9.19)]([3, Eq. (9.36)]), and the other the multipolar coefficients of the field [3, Eq. (9.149)](far field limit of [3, Eq. (9.122)] which can be obtained using [3, Eq. (9.89)]). One then uses an expression of the cross product in spherical coordinates

$$\sum_{m=-1}^{1} w_m \mathbf{X}_{1m}(\hat{\mathbf{r}}) = i\sqrt{\frac{3}{8\pi}}\mathbf{w}\times\hat{\mathbf{r}}, \tag{S10}$$

with the definition of $\mathbf{X}_{1m}(\hat{\mathbf{r}})$ in [3, Eq. (9.119)], and the scale factor differences between the coefficients in the multipole expansions that we are using [Eq. (2) in the main text] and those in [3, Eq. (9.122)] to conclude that:

$$\begin{bmatrix} c^\omega_{1-1} \\ c^\omega_{10} \\ c^\omega_{11} \end{bmatrix} = \frac{c^\omega Z^\omega (k^\omega)^3}{\sqrt{6\pi}}\mathbf{p}^\omega,\ \begin{bmatrix} d^\omega_{1-1} \\ d^\omega_{10} \\ d^\omega_{11} \end{bmatrix} = i\frac{Z^\omega (k^\omega)^3}{\sqrt{6\pi}}\mathbf{m}^\omega, \tag{S11}$$

where $c^\omega = 1/\sqrt{\epsilon^\omega\mu^\omega}$.

We can now perform the connection between Eq. (S1) and (S2). First we use Eq. (S11) on the left hand side of Eq. (S1), and Eq. (S9) on the vector of its right hand side:

$$\frac{c^\omega Z^\omega (k^\omega)^3}{\sqrt{6\pi}} \begin{bmatrix} \mathbf{p}^\omega \\ \frac{i\mathbf{m}^\omega}{c^\omega} \end{bmatrix} = \begin{bmatrix} \underline{\underline{T}}^\omega_{NN} & \underline{\underline{T}}^\omega_{NM} \\ \underline{\underline{T}}^\omega_{MN} & \underline{\underline{T}}^\omega_{MM} \end{bmatrix} \begin{bmatrix} \mathbf{E}^\omega(0) \\ iZ^\omega\mathbf{H}^\omega(0) \end{bmatrix} (-i)\sqrt{\frac{12\pi}{2}} \tag{S12}$$

Re-arranging the scalar factors we obtain

$$\begin{bmatrix} \mathbf{p}^\omega \\ \frac{i\mathbf{m}^\omega}{c^\omega} \end{bmatrix} = \begin{bmatrix} \underline{\underline{T}}^\omega_{NN} & \underline{\underline{T}}^\omega_{NM} \\ \underline{\underline{T}}^\omega_{MN} & \underline{\underline{T}}^\omega_{MM} \end{bmatrix} \begin{bmatrix} \mathbf{E}^\omega(0) \\ iZ^\omega\mathbf{H}^\omega(0) \end{bmatrix} \frac{(-i)6\pi}{c^\omega Z^\omega (k^\omega)^3}, \tag{S13}$$

which, after modifying the 3×3 matrices to obtain the desired input and output vectors produces

$$\begin{bmatrix} \mathbf{p}^\omega \\ \mathbf{m}^\omega \end{bmatrix} = \begin{bmatrix} \underline{\underline{T}}^\omega_{NN} & iZ^\omega\underline{\underline{T}}^\omega_{NM} \\ -ic^\omega\underline{\underline{T}}^\omega_{MN} & c^\omega Z^\omega\underline{\underline{T}}^\omega_{MM} \end{bmatrix} \begin{bmatrix} \mathbf{E}^\omega(0) \\ \mathbf{H}^\omega(0) \end{bmatrix} \frac{(-i)6\pi}{c^\omega Z^\omega (k^\omega)^3}. \tag{S14}$$

And it follows form comparing Eq. (S2) with Eq. (S14) that

$$\begin{bmatrix} \underline{\underline{\alpha}}^\omega_{pE} & \underline{\underline{\alpha}}^\omega_{pH} \\ \underline{\underline{\alpha}}^\omega_{mE} & \underline{\underline{\alpha}}^\omega_{mH} \end{bmatrix} = \frac{(-i)6\pi}{c^\omega Z^\omega (k^\omega)^3} \begin{bmatrix} \underline{\underline{T}}^\omega_{NN} & iZ^\omega\underline{\underline{T}}^\omega_{NM} \\ -ic^\omega\underline{\underline{T}}^\omega_{MN} & c^\omega Z^\omega\underline{\underline{T}}^\omega_{MM} \end{bmatrix}. \tag{S15}$$

This result leads directly to Eq. (6) in the main text by recalling that in the typical case where the polarizabilities are available in the Cartesian basis, we must change the basis to spherical. The change of basis matrix $\underline{\underline{C}}$ in Eq. (6) in the main text is the one in Eq. (S8).

II. ABSORPTION

The absorption of an object upon a particular illumination is best analyzed using the S-matrix. Given the T-matrix, the S-matrix or scattering matrix can be computed as:

$$\underline{\underline{S}}^\omega = \underline{\underline{I}} + 2\underline{\underline{T}}^\omega, \tag{S16}$$

where I is the identity matrix. Notwithstanding the simple numerical relationship in Eq. (S16), there is an important physical difference between the two matrices. While the T-matrix relates incident and scattered fields, the S-matrix relates total incoming and outgoing fields. The total incoming(outgoing) fields are the total fields before(after) the interaction. The difference is that an incident field has a mixed incoming and outgoing character and exists before and after the interaction. Its incoming part is the total incoming field, its outgoing part plus the scattered field equals the total outgoing field. The fact that the S-matrix connects total fields allows for the simple following derivation of the absorption.

A. Oriented CD

Having assumed that the light-matter interaction does not change the frequency of the fields, we can just focus on a single frequency component. Let us consider an incoming field represented by its coordinates in the helicity basis $\underline{\alpha}^\omega$. Its norm squared is $\underline{\alpha}^{\omega\dagger}\underline{\alpha}^\omega$. The outgoing field is $\underline{\underline{S}}^\omega\underline{\alpha}^\omega$, with norm squared $\underline{\alpha}^{\omega\dagger}\underline{\underline{S}}^{\omega\dagger}\underline{\underline{S}}^\omega\underline{\alpha}^\omega$. The absorption must hence be the difference

$$\underline{\alpha}^{\omega\dagger}\underline{\alpha}^\omega - \underline{\alpha}^{\omega\dagger}\underline{\underline{S}}^{\omega\dagger}\underline{\underline{S}}^\omega\underline{\alpha}^\omega. \tag{S17}$$

After using the block decomposition of the T-matrix in Eq. (8) in the main text, Eq. (S17) becomes

$$\underline{\alpha}^{\omega\dagger}\underline{\alpha}^\omega - \underline{\alpha}^{\omega\dagger}\left(\underline{\underline{I}} + 2\begin{bmatrix}\underline{\underline{T}}^\omega_{++} & \underline{\underline{T}}^\omega_{+-}\\ \underline{\underline{T}}^\omega_{-+} & \underline{\underline{T}}^\omega_{--}\end{bmatrix}\right)^\dagger\left(\underline{\underline{I}} + 2\begin{bmatrix}\underline{\underline{T}}^\omega_{++} & \underline{\underline{T}}^\omega_{+-}\\ \underline{\underline{T}}^\omega_{-+} & \underline{\underline{T}}^\omega_{--}\end{bmatrix}\right)\underline{\alpha}^\omega. \tag{S18}$$

Particularizing $\underline{\alpha}^\omega$ to a plane wave of a given helicity with momentum direction $\hat{\mathbf{v}}$ quickly leads to Eq. (9) in the main text. Let us do it for an incoming plane wave of positive helicity, which has zero projection on the negative helicity multipoles $\mathbf{A}^\omega_{jm-}(\mathbf{r})$ [Eq. (7) in the main text], so half of the vector is filled with zeros:

$$\begin{bmatrix}\underline{\alpha}^\omega_+(\hat{\mathbf{v}})\\ 0\end{bmatrix}. \tag{S19}$$

Then Eq. (S18) can be written as:

$$\begin{aligned}&\underline{\alpha}^\omega_+(\hat{\mathbf{v}})^\dagger\underline{\alpha}^\omega_+(\hat{\mathbf{v}}) -\\ &\left(\begin{bmatrix}\underline{\alpha}^\omega_+(\hat{\mathbf{v}})\\ 0\end{bmatrix}^\dagger + 2\begin{bmatrix}\underline{\alpha}^\omega_+(\hat{\mathbf{v}})^\dagger\underline{\underline{T}}^{\omega\ \dagger}_{++} & \underline{\alpha}^\omega_+(\hat{\mathbf{v}})^\dagger\underline{\underline{T}}^{\omega\ \dagger}_{-+}\end{bmatrix}\right)\times\\ &\left(\begin{bmatrix}\underline{\alpha}^\omega_+(\hat{\mathbf{v}})\\ 0\end{bmatrix} + 2\begin{bmatrix}\underline{\underline{T}}^\omega_{++}\alpha_+(\hat{\mathbf{v}})\\ \underline{\underline{T}}^\omega_{-+}\alpha_+(\hat{\mathbf{v}})\end{bmatrix}\right),\end{aligned} \tag{S20}$$

which readily results into the first line of Eq. (9) in the main text after considering the following point. The vectors of coefficients $\mu_\pm(\hat{\mathbf{v}})$ in Eq. (9) in the main text represent *incident* plane waves, while the vectors of coefficients $\alpha_\pm(\hat{\mathbf{v}})$ represent the *incoming* part of the incident plane waves. The numerical relation

$$\alpha_\pm(\hat{\mathbf{v}}) = \frac{1}{2}\mu_\pm(\hat{\mathbf{v}}), \tag{S21}$$

can be deduced from the expansion of an incident plane wave into regular multipoles, featuring spherical Bessel functions, which can be written as the following sum of incoming and outgoing spherical Hankel functions: $j_l(\cdot) = [h^1_l(\cdot) + h^2_l(\cdot)]/2$.

B. Rotationally averaged CD

We now address the rotationally averaged CD, that is, the differential absorption averaged over all possible spatial directions of an incident plane wave:

$$\mathrm{CD}^\omega = \int d\hat{\mathbf{v}}\left[\underline{\mu}^\omega_+(\hat{\mathbf{v}})^\dagger\,\underline{\underline{A}}^\omega_+\underline{\mu}^\omega_+(\hat{\mathbf{v}}) - \underline{\mu}^\omega_-(\hat{\mathbf{v}})^\dagger\,\underline{\underline{A}}^\omega_-\underline{\mu}^\omega_-(\hat{\mathbf{v}})\right], \tag{S22}$$

where $\underline{\underline{A}}^\omega_\pm$ are defined in Eq. (9) in the main text. As per [2, Eq. (8.4-6)], each $\underline{\mu}^\omega_+(\hat{\mathbf{v}})$ and $\underline{\mu}^\omega_-(\hat{\mathbf{v}})$ can be obtained by a corresponding rotation of a reference vector representing a plane wave whose momentum is aligned with the $\hat{\mathbf{z}}$ direction: $\underline{\mu}_\pm(\hat{\mathbf{v}}) = \underline{\underline{R}}(\hat{\mathbf{v}})\underline{\mu}_\pm(\hat{\mathbf{z}})$. This allows us to write Eq. (S22) as

$$\mathrm{CD}^\omega = \underline{\mu}^\omega_+(\hat{\mathbf{z}})^\dagger\,\mathrm{Ro}\left\{\underline{\underline{A}}^\omega_+\right\}\underline{\mu}^\omega_+(\hat{\mathbf{z}}) - \underline{\mu}^\omega_-(\hat{\mathbf{z}})^\dagger\,\mathrm{Ro}\left\{\underline{\underline{A}}^\omega_-\right\}\underline{\mu}^\omega_-(\hat{\mathbf{z}}), \tag{S23}$$

where $\mathrm{Ro}\left\{\underline{\underline{F}}\right\} = \int d\hat{\mathbf{v}}\underline{\underline{R}}(\hat{\mathbf{v}})^\dagger\underline{\underline{F}}\,\underline{\underline{R}}(\hat{\mathbf{v}})$ is the rotational average of matrix $\underline{\underline{F}}$.

The rotationally averaged matrices $\mathrm{Ro}\left\{\underline{\underline{A}}^\omega_+\right\}$ and $\mathrm{Ro}\left\{\underline{\underline{A}}^\omega_-\right\}$ exhibit spherical symmetry, which means that they are diagonal in the multipolar basis of well defined total angular momentum, indexed by j, and angular momentum along the $\hat{\mathbf{z}}$ axis, indexed by m. Moreover, for each j subspace, the diagonal elements are equal for all $m \in [-j \dots j]$ (see [2, Eq. (7.5-13)]). The structure of the matrices is hence

$$\begin{bmatrix}c_1\underline{\underline{I}}_{3\times3} & \underline{\underline{0}}_{3\times5} & \cdots & \cdots\\ \underline{\underline{0}}_{5\times3} & c_2\underline{\underline{I}}_{5\times5} & \cdots & \cdots\\ \vdots & \vdots & \ddots & \vdots\end{bmatrix}. \tag{S24}$$

We now need the expansion of the circularly polarized incident plane waves $\underline{\mu}_\pm(\hat{\mathbf{z}})$ into regular multipolar fields. It can be found for example in [3, Eq. (10.55)]. In the notation used in this paper it reads:

$$\mathbf{E}(\mathbf{r})^\omega_\pm = \sum_{j=1}^{\infty} i^j\sqrt{(4\pi)(2j+1)}\left[\mathbf{M}^\omega_{jm=\pm1}(\mathbf{r}) \pm \mathbf{N}^\omega_{jm=\pm1}(\mathbf{r})\right]. \tag{S25}$$

For a plane wave of helicity ±1 the coefficients are zero except in the positions corresponding to angular momentum $m = \pm1$. In order to bring Eq. (S25) to our conventions, we first note that, as per [3, Eq. (10.46)], it corresponds to the expansion of $\mathbf{E}(\mathbf{r}) = (\hat{\mathbf{x}} \pm i\hat{\mathbf{y}})\exp(ikz)$, whose polarization vector is a factor of $\sqrt{2}$ larger than its corresponding unitary vector. After we divide Eq. (S25) by $\sqrt{2}$, we can take this factor into the multipolar functions to obtain

$$\frac{1}{\sqrt{2}}\left[\mathbf{M}^\omega_{jm=\pm1}(\mathbf{r}) \pm \mathbf{N}^\omega_{jm=\pm1}(\mathbf{r})\right], \tag{S26}$$

which we manipulate to match the definitions of multipolar fields of well defined helicity $\lambda = \pm1$ in Eq. (7) in the main text

$$\frac{\lambda}{\sqrt{2}}\left[\mathbf{N}^\omega_{jm=\lambda}(\mathbf{r}) + \lambda\mathbf{M}^\omega_{jm=\lambda}(\mathbf{r})\right] = \lambda\mathbf{A}^\omega_{jm=\lambda,\lambda}(\mathbf{r}), \tag{S27}$$

It then follows that the expansion coefficients $\underline{\mu}^\omega_\pm(\hat{\mathbf{v}})$ that we are looking for are

$$\mu^\omega_{jm\lambda}(\hat{\mathbf{z}}) = \lambda\left(i^j\right)\sqrt{(4\pi)(2j+1)}\delta_{\lambda m}, \tag{S28}$$

where $\delta_{\lambda m}$ is the Kronecker delta.

We can now evaluate the quadratic forms in Eq. (S23). For $\lambda = 1$, and recalling the diagonal structure of $\mathrm{Ro}\left\{\underline{\underline{A}}^\omega_+\right\}$ from Eq. (S24), we can write

$$\begin{aligned}&\underline{\mu}^\omega_+(\hat{\mathbf{z}})^\dagger\,\mathrm{Ro}\left\{\underline{\underline{A}}^\omega_+\right\}\underline{\mu}^\omega_+(\hat{\mathbf{z}}) =\\ &4\pi\mathrm{Tr}\left\{\mathrm{Ro}\left\{\underline{\underline{A}}^\omega_+\right\}_1\right\} + 4\pi\mathrm{Tr}\left\{\mathrm{Ro}\left\{\underline{\underline{A}}^\omega_+\right\}_2\right\} + \ldots +\\ &4\pi\mathrm{Tr}\left\{\mathrm{Ro}\left\{\underline{\underline{A}}^\omega_+\right\}_j\right\} + \ldots = 4\pi\mathrm{Tr}\left\{\mathrm{Ro}\left\{\underline{\underline{A}}^\omega_+\right\}\right\},\end{aligned} \tag{S29}$$

where $\mathrm{Tr}\left\{\underline{\underline{F}}\right\}_j$ is the trace of the submatrix of $\underline{\underline{F}}$ which connects multipoles of order j at the input to multipoles of the same order j at the output. Equation (S29) is reached by considering that, from Eq. (S24), the diagonal elements of $\mathrm{Ro}\left\{\underline{\underline{A}}^\omega_+\right\}$ are $c_j = \mathrm{Tr}\left\{\underline{\underline{F}}\right\}_j/(2j+1)$, and that the combined action of $\underline{\mu}^\omega_+(\hat{\mathbf{z}})^\dagger$ and $\underline{\mu}^\omega_+(\hat{\mathbf{z}})$ is to select a single element on the diagonal for each multipolar order, and, according to Eq. (S28), multiply it by $|\lambda\left(i^j\right)\sqrt{(4\pi)(2j+1)}|^2$.

The final step is the realization that it is not necessary to perform the rotational averages of the matrices because:

$$\mathrm{Tr}\left\{\mathrm{Ro}\left\{\underline{\underline{A}}^\omega_+\right\}_j\right\} = \mathrm{Tr}\left\{\underline{\underline{A}}^\omega_{+j}\right\}, \tag{S30}$$

and hence

$$\mathrm{Tr}\left\{\mathrm{Ro}\left\{\underline{\underline{A}}^\omega_+\right\}\right\} = \mathrm{Tr}\left\{\underline{\underline{A}}^\omega_+\right\}. \tag{S31}$$

Equations (S30) and (S31) follow from the facts that the trace is a rotationally invariant quantity, and that rotations do not mix the submatrices corresponding to different values of j.

After collecting these results, we obtain that Eq. (S23) can be written as

$$\boxed{\mathrm{CD}^\omega = 4\pi\mathrm{Tr}\left\{\underline{\underline{A}}^\omega_+ - \underline{\underline{A}}^\omega_-\right\}.} \tag{S32}$$

The quantity in Eq. (S32) can be seen as a differential absorption probability. We now show how to express it in the familiar CD units [liter/mol/cm]. We can achieve this in two steps: 1) Convert absorption probability to absorption cross-section, and 2) use the conversion factor between absorption cross-section and units of [liter/mol/cm]: $10N_A/\ln(10)$ where $N_A = 6.0221409\times 10^{23}$ is Avogadro's number. Step 1) can be achieved comparing the expression for the absorption cross-section of a sphere under a circularly polarized plane-wave illumination in [3, Eq. (10.61)]

$$\sigma^\omega_{abs} = \frac{\pi}{2(k^\omega)^2}\sum_j (2j+1)\left(2 - |\alpha^\omega_{j\pm}-1|^2 - |\beta^\omega_{j\pm}-1|^2\right), \tag{S33}$$

with our expression for the absorption probability

$$\mathrm{p}^\omega_{abs} = \underline{\mu}^\omega_\lambda(\hat{\mathbf{z}})^\dagger\underline{\underline{A}}^\omega_\lambda\underline{\mu}^\omega_\lambda(\hat{\mathbf{z}}). \tag{S34}$$

We start by exploiting the structure of the T-matrix of a sphere. First, it does not couple multipoles with different j or different m. Second, for each multipolar order j, it does not depend on m. And third, in the basis of multipoles of well defined helicity [Eq. (7) in the main text], the 2×2 submatrices relating incident and scattered multipoles of equal m and j read

$$-\frac{1}{2}\begin{bmatrix} a^\omega_j + b^\omega_j & a^\omega_j - b^\omega_j\\ a^\omega_j - b^\omega_j & a^\omega_j + b^\omega_j\end{bmatrix}, \tag{S35}$$

where a^ω_j and b^ω_j are the electric and magnetic Mie coefficients of the sphere, respectively.

Using the definitions of $\underline{\underline{A}}^\omega_\lambda$ in Eq. (9) in the main text, and Eqs. (S28) and (S35), one readily reaches from Eq. (S34)

$$\mathrm{p}^\omega_{abs} = 2\pi\sum_j(2j+1)\left(\mathbb{R}\left\{a^\omega_j + b^\omega_j\right\} - |a^\omega_j|^2 - |b^\omega_j|^2\right), \tag{S36}$$

where $\mathbb{R}\{\cdot\}$ takes the real part of its argument. We now need to relate the Mie coefficients (a^ω_j, b^ω_j) to the $(\alpha^\omega_{j\pm}, \beta^\omega_{j\pm})$ coefficients in Eq. (S33), which can be done using the expansion of the field scattered by a sphere upon illumination with a circularly polarized incident plane wave [3, Eq. (10.57)]. Particularized for incident helicity $\lambda = 1$, it reads:

$$\begin{aligned}\mathbf{E}^\omega_{sc}(\mathbf{r}) = \frac{1}{2}\sum_{j=1}^{\infty} i^j\sqrt{(4\pi)(2j+1)}\times&\\ \left[\alpha^+_j\hat{\mathbf{M}}^\omega_{jm=1}(\mathbf{r}) + \beta^+_j\hat{\mathbf{N}}^\omega_{jm=1}(\mathbf{r})\right]&,\end{aligned} \tag{S37}$$

where $\hat{\mathbf{M}}^\omega(\mathbf{r})$ and $\hat{\mathbf{N}}^\omega(\mathbf{r})$ are outgoing multipoles. We now change the basis, the position of the leading 1/2 factor, and divide by the previously discussed $\sqrt{2}$ factor:

$$\begin{aligned}\mathbf{E}_{sc}(\mathbf{r}) = \sum_{j=1}^{\infty} i^j\sqrt{(4\pi)(2j+1)}\times&\\ \left[\frac{\beta^+_j + \alpha^+_j}{4}\hat{\mathbf{A}}^\omega_{jm+} + \frac{\beta^+_j - \alpha^+_j}{4}\hat{\mathbf{A}}^\omega_{jm-}\right]&.\end{aligned} \tag{S38}$$

We keep Eq. (S38) for future use and we now use the relationship between the coefficients of the incident plane wave $\underline{\mu}^\omega_+(\hat{\mathbf{z}})$, and the scattered field $\underline{\rho}^\omega(\hat{\mathbf{z}})$ [Eq. (8) in the main text]

$$\begin{bmatrix}\underline{\rho}^\omega_+\\ \underline{\rho}^\omega_-\end{bmatrix} = \begin{bmatrix}\underline{\underline{T}}^\omega_{++} & \underline{\underline{T}}^\omega_{+-}\\ \underline{\underline{T}}^\omega_{-+} & \underline{\underline{T}}^\omega_{--}\end{bmatrix}\begin{bmatrix}\underline{\mu}^\omega_+\\ \underline{0}\end{bmatrix}, \tag{S39}$$

and Eqs. (S28) and (S35) to write:

$$\begin{aligned}\rho^\omega_{jm+} &= -\frac{(a^\omega_j + b^\omega_j)}{2}(i)^j\sqrt{(4\pi)(2j+1)}\delta_{1m},\\ \rho^\omega_{jm-} &= -\frac{(a^\omega_j - b^\omega_j)}{2}(i)^j\sqrt{(4\pi)(2j+1)}\delta_{-1m}.\end{aligned} \tag{S40}$$

Comparing Eqs. (S38) and (S40) shows that:

$$\alpha^\omega_{j+} = -2b^\omega_j,\ \beta^\omega_{j+} = -2a^\omega_j, \tag{S41}$$

and allows to change Eq. (S33) into:

$$\sigma^{\omega}_{abs} = \frac{2\pi}{(k^{\omega})^2} \sum_j (2j+1) \left(\mathbb{R}\left\{a^{\omega}_j + b^{\omega}_j\right\} - |a^{\omega}_j|^2 - |b^{\omega}_j|^2 \right). \tag{S42}$$

The same result is obtained using the opposite circular polarization.

Now, we note that the definition of the absorption cross-section includes the division by the incident flux. This means in particular that the $1/\sqrt{2}$ factor that we have been compensating for does not change it. We can hence, finally, compare Eqs. (S42) and (S36) to reveal that

$$\sigma^{\omega}_{abs} = \frac{\mathrm{p}^{\omega}_{abs}}{(k^{\omega})^2}. \tag{S43}$$

Therefore, in units of [liter/mol/cm], the frequency dependent circular dichroism as a function of the T-matrix reads:

$$\overline{\mathrm{CD}}^{\omega} = \frac{10N_A}{\ln(10)} \frac{4\pi}{(k^{\omega})^2} \mathrm{Tr}\left\{ \underline{\underline{A}}^{\omega}_{+} - \underline{\underline{A}}^{\omega}_{-} \right\}. \tag{S44}$$

III. PARITY TO HELICITY CHANGE

The change of basis of Eq. (7) in the main text

$$\mathbf{A}^{\omega}_{jm+}(\mathbf{r}) = \frac{\mathbf{N}^{\omega}_{jm}(\mathbf{r}) + \mathbf{M}^{\omega}(\mathbf{r})}{\sqrt{2}},\ \mathbf{A}^{\omega}_{jm-}(\mathbf{r}) = \frac{\mathbf{N}^{\omega}_{jm}(\mathbf{r}) - \mathbf{M}^{\omega}_{jm}(\mathbf{r})}{\sqrt{2}}, \tag{S45}$$

induces the relationships

$$\begin{aligned} \sqrt{2}\rho^{\omega}_{jm\pm} &= c^{\omega}_{jm} \pm d^{\omega}_{jm} \\ \sqrt{2}\mu^{\omega}_{jm\pm} &= a^{\omega}_{jm} \pm b^{\omega}_{jm} \end{aligned} \tag{S46}$$

and

$$2\begin{bmatrix} \underline{\underline{T}}^{\omega}_{++} & \underline{\underline{T}}^{\omega}_{+-} \\ \underline{\underline{T}}^{\omega}_{-+} & \underline{\underline{T}}^{\omega}_{--} \end{bmatrix} = \begin{bmatrix} \underline{\underline{I}} & \underline{\underline{I}} \\ \underline{\underline{I}} & -\underline{\underline{I}} \end{bmatrix} \begin{bmatrix} \underline{\underline{T}}^{\omega}_{NN} & \underline{\underline{T}}^{\omega}_{NM} \\ \underline{\underline{T}}^{\omega}_{MN} & \underline{\underline{T}}^{\omega}_{MM} \end{bmatrix} \begin{bmatrix} \underline{\underline{I}} & \underline{\underline{I}} \\ \underline{\underline{I}} & -\underline{\underline{I}} \end{bmatrix}, \tag{S47}$$

where $\underline{\underline{I}}$ is the identity matrix.

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[3] J. D. Jackson, *Classical Electrodynamics* (Wiley, New York City, 1998).