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Supporting Material

Molecular Order of Arterial Collagen Using Circular Polarization Second-Harmonic Generation Imaging

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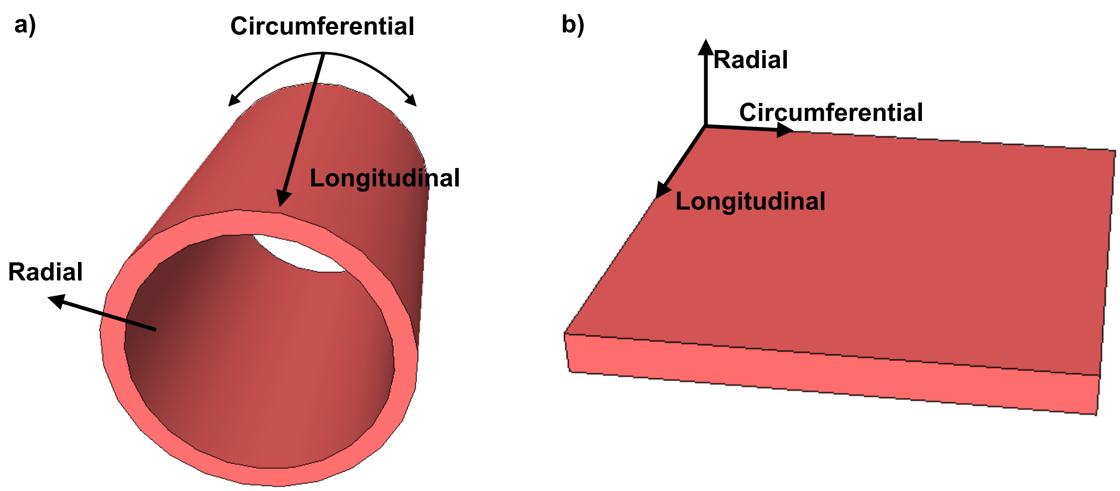

Supporting Fig 1 Diagram showing axial, circumferential, and radial directions in a) intact and b) cut-open arteries. Arteries are cut-open along the longitudinal direction.

Derivation of SHG Polarizability for collagen fibers

The general form of the second order polarizability is:[1]

$$\mathbf{P}^{(2)}_{\text{SHG}} = \epsilon_0 \chi^{(2)} \mathbf{E}, \tag{1}$$

where $\chi^{(2)}$ is the third rank tensor for the second order nonlinear susceptibility

$$\chi^{(2)} = \begin{bmatrix} \chi_{xxx} & \chi_{xyy} & \chi_{xzz} & \chi_{xxy} & \chi_{xxz} & \chi_{xyx} & \chi_{xzx} & \chi_{xyz} & \chi_{xzy} \\ \chi_{yxx} & \chi_{yyy} & \chi_{yzz} & \chi_{yxy} & \chi_{yxz} & \chi_{yyx} & \chi_{yzx} & \chi_{yyz} & \chi_{yzy} \\ \chi_{zxx} & \chi_{zyy} & \chi_{zzz} & \chi_{zxy} & \chi_{zxz} & \chi_{zyx} & \chi_{zzx} & \chi_{zyz} & \chi_{zzy} \end{bmatrix}, \tag{2}$$

and $\mathbf{E}$ is the general incident fields vector:

$$\mathbf{E} = \begin{bmatrix} E_x(\omega)E_x(\omega) \\ E_y(\omega)E_y(\omega) \\ E_z(\omega)E_z(\omega) \\ E_x(\omega)E_y(\omega) \\ E_x(\omega)E_z(\omega) \\ E_y(\omega)E_x(\omega) \\ E_z(\omega)E_x(\omega) \\ E_y(\omega)E_z(\omega) \\ E_z(\omega)E_y(\omega) \end{bmatrix}. \tag{3}$$

With c being a proportionality constant, the second-order polarizability is linked to the intensity by:

$$I = c[P^{(2)}_x P^{(2)*}_x + P^{(2)}_y P^{(2)*}_y + P^{(2)}_z P^{(2)*}_z], \tag{4}$$

Several symmetry arguments can be made to simplified the $\chi^{(2)}$ tensor. These arguments depend on the properties of the nonlinear process, but also on sample. For SHG in collagen fibers, hexagonal symmetry applies and the only non-zero elements are d_{33}, d_{31}, d_{15}, and d_{14}.[1] d_{is} is defined as χ_{ijk} with $i, j, k = \{x, y, z\}$ and $s = 1, 2, 3, 4, 5, 6$ for $xx, yy, zz, \{yz, zy\}, \{xz, zx\}$, and $\{xy, yx\}$ respectively. Eq. (2) can thus be written as:

$$\chi^{(2)} = \begin{bmatrix} 0 & 0 & 0 & 0 & a & 0 & a & b & b \\ 0 & 0 & 0 & 0 & -b & 0 & -b & a & a \\ m & m & n & 0 & 0 & 0 & 0 & 0 & 0 \end{bmatrix}, \tag{5}$$

where $a = d_{15}$, $b = d_{14}$, $m = d_{31}$, and $n = d_{33}$.

Cylindrical symmetry is also often considered.[2] Assuming cylindrical symmetry ($b = 0$ and $a = m$) is equivalent to assuming Kleinman symmetry. The experimental a/m ratio is not exactly 1 and both terms are usually kept.[3] This allows to express Eq. (5) as:

$$\chi^{(2)} = \begin{bmatrix} 0 & 0 & 0 & 0 & a & 0 & a & 0 & 0 \\ 0 & 0 & 0 & 0 & 0 & 0 & 0 & a & a \\ m & m & n & 0 & 0 & 0 & 0 & 0 & 0 \end{bmatrix}. \tag{6}$$

Assuming y-axis propagation of excitation fields ($E_y = 0$), Eq. (1) can be written as:

$$\begin{bmatrix} P_x^{(2)}(2\omega) \\ P_y^{(2)}(2\omega) \\ P_z^{(2)}(2\omega) \end{bmatrix} = \epsilon_0 \begin{bmatrix} 0 & 0 & 0 & 0 & a & 0 & a & 0 & 0 \\ 0 & 0 & 0 & 0 & 0 & 0 & 0 & a & a \\ m & m & n & 0 & 0 & 0 & 0 & 0 & 0 \end{bmatrix} \begin{bmatrix} E_x(\omega)E_x(\omega) \\ 0 \\ E_z(\omega)E_z(\omega) \\ 0 \\ E_x(\omega)E_z(\omega) \\ 0 \\ E_z(\omega)E_x(\omega) \\ 0 \\ 0 \end{bmatrix}, \text{ or} \tag{7}$$

$$\begin{bmatrix} P_x^{(2)}(2\omega) \\ P_y^{(2)}(2\omega) \\ P_z^{(2)}(2\omega) \end{bmatrix} = \epsilon_0 \begin{bmatrix} \chi_{xzx}E_x(\omega)E_z(\omega)+\chi_{xzx}E_z(\omega)E_x(\omega) \\ 0 \\ \chi_{zxx}E_x(\omega)E_x(\omega)+\chi_{zzz}E_z(\omega)E_z(\omega) \end{bmatrix}. \tag{8}$$

Eq. (8) is valid only when hexagonal and cylindrical symmetries apply and can be used for any incident polarization state.

Intensity for linearly polarized light

The electric field for linearly polarization light can be expressed in terms of the Jones vector:

$$\mathbf{E} = |\mathbf{E}| Re\{|\psi\rangle exp[i(ky)]\}, \text{ where} \tag{9}$$

$$|\psi\rangle = \begin{bmatrix} cos(\theta) \\ sin(\theta) \end{bmatrix} exp[i\alpha]. \tag{10}$$

The electric field can therefore be decomposed as $E_z(\omega) = |E(\omega)|cos(\theta)$ and $E_x(\omega) = |E(\omega)|sin(\theta)$. For linearly polarized light, Eq. (8) can thus be expressed as:

$$\begin{bmatrix} P_x^{(2)}(2\omega) \\ P_y^{(2)}(2\omega) \\ P_z^{(2)}(2\omega) \end{bmatrix} = \epsilon_0 \begin{bmatrix} \chi_{xzx}|E(\omega)||E(\omega)|sin(\theta)cos(\theta)+\chi_{xxz}|E(\omega)||E(\omega)|sin(\theta)cos(\theta) \\ 0 \\ \chi_{zxx}|E(\omega)||E(\omega)|sin(\theta)sin(\theta)+\chi_{zzz}|E(\omega)||E(\omega)|cos(\theta)cos(\theta) \end{bmatrix}, \tag{11}$$

$$\begin{bmatrix} P_x^{(2)}(2\omega) \\ P_y^{(2)}(2\omega) \\ P_z^{(2)}(2\omega) \end{bmatrix} = \epsilon_0 |E(\omega)|^2 \begin{bmatrix} 2\chi_{xzx}sin(\theta)cos(\theta) \\ 0 \\ \chi_{zxx}sin^2(\theta)+\chi_{zzz}cos^2(\theta) \end{bmatrix}, \text{ or} \tag{12}$$

$$\begin{bmatrix} P_x^{(2)}(2\omega) \\ P_y^{(2)}(2\omega) \\ P_z^{(2)}(2\omega) \end{bmatrix} = \epsilon_0 |E(\omega)|^2 \begin{bmatrix} \chi_{xzx}sin(2\theta) \\ 0 \\ \chi_{zxx}sin^2(\theta)+\chi_{zzz}cos^2(\theta) \end{bmatrix}. \tag{13}$$

Using Eq. (13) in Eq. (4), we obtain the commonly used expression for the SHG intensity in polarimetric measurements:[3]

$$I_{SHG}^{Lin}(2\omega, \theta) = c\epsilon_0^2 |E(\omega)|^4 \left[(\chi_{xzx}sin(2\theta))^2 + (\chi_{zxx}sin^2(\theta) + \chi_{zzz}cos^2(\theta))^2 \right]. \tag{14}$$

A standard notation for Eq. (14) uses $\rho_1 = \chi_{zzz}/\chi_{zxx}$ and $\rho_2 = \chi_{xzx}/\chi_{zxx}$:[3]

$$I_{SHG}^{Lin}(2\omega, \theta) = c\epsilon_0^2 \chi_{zxx}^2 |E(\omega)|^4 \left[(\rho_2 sin(2\theta))^2 + (sin^2(\theta) + \rho_1 cos^2(\theta))^2 \right]. \tag{15}$$

Intensity for circularly polarized light

The linear polarization dependence has been presented on multiple occasions.[2] Here, we also introduce the equivalent expression for circularly polarized incident light. The electric field for circularly polarization light can be expressed in terms of the Jones vector:

$$\mathbf{E} = |\mathbf{E}| Re\{|\psi\rangle exp[i(ky)]\}, \text{ where} \tag{16}$$

$$|\psi\rangle = \tfrac{1}{\sqrt{2}} \left[\begin{smallmatrix} 1 \\ \pm i \end{smallmatrix} \right] exp[i\alpha]\}. \tag{17}$$

The electric field can therefore be decomposed as $E_z(\omega) = \frac{1}{\sqrt{2}}|E(\omega)|$ and $E_x(\omega) = \frac{\pm i}{\sqrt{2}}|E(\omega)|$. For circularly polarized light, Eq. (8) can thus be expressed as:

$$\begin{bmatrix} P_x^{(2)}(2\omega) \\ P_y^{(2)}(2\omega) \\ P_z^{(2)}(2\omega) \end{bmatrix} = \epsilon_0 \begin{bmatrix} \frac{\pm i}{2}\chi_{xzx}|E(\omega)||E(\omega)| + \frac{\pm i}{2}\chi_{xzx}|E(\omega)||E(\omega)| \\ 0 \\ \frac{-1}{2}\chi_{zxx}|E(\omega)||E(\omega)| + \frac{1}{2}\chi_{zzz}|E(\omega)||E(\omega)| \end{bmatrix}, \text{ or} \tag{18}$$

$$\begin{bmatrix} P_x^{(2)}(2\omega) \\ P_y^{(2)}(2\omega) \\ P_z^{(2)}(2\omega) \end{bmatrix} = \frac{\epsilon_0}{2}|E(\omega)|^2 \begin{bmatrix} \pm i2\chi_{xzx} \\ 0 \\ \chi_{zzz} - \chi_{zxx} \end{bmatrix}. \tag{19}$$

Using Eq. (19) in Eq. (4), we obtain an expression for the SHG intensity with circularly polarized incident light.

$$I_{SHG}^{Cir}(2\omega) = c\frac{\epsilon_0^2}{4}|E(\omega)|^4 \left[(4\chi_{xzx}^2 + (\chi_{zzz} - \chi_{zxx})^2\right]. \tag{20}$$

Using the same notation as in Eq. (15), Eq. (20) can be written as:

$$I_{SHG}^{Cir}(2\omega) = c\frac{\epsilon_0^2}{4}\chi_{zxx}^2|E(\omega)|^4 \left[(4\rho_2^2 + (\rho_1 - 1)^2\right]. \tag{21}$$

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