Chapter 1

SHG Yield

1.1 Three layer model for SHG yield, without Multiple Reflections

In this section we derive the formulas required for the calculation of the SHG yield, defined by

$$\mathcal{R}(\omega) = \frac{I(2\omega)}{I^2(\omega)}, \tag{1.1}$$

with the intensity in the MKS system is given by[1]

$$I(\omega) = 2n(\omega)\epsilon_0 c|E(\omega)|^2, \tag{1.2}$$

where $n(\omega) = \sqrt{\epsilon(\omega)}$ is the index of refraction with $\epsilon\omega$ the dielectric function, ϵ_0 is the vacuum permittivity, and c the speed of light in vacuum.

There are several ways to calculate R, one of which is the procedure followed by Cini [2]. This approach calculates the nonlinear susceptibility and at the same time the radiated fields. However, we present an alternative derivation based in the work of Mizrahi and Sipe [3], since the derivation of the three-layer-model is straightforward. In this scheme, we represent the surface by three regions or layers. The first layer is the vacuum region (denoted by v) with a dielectric function $\epsilon_v(\omega) = 1$ from where the fundamental electric field $\mathbf{E}_v(\omega)$ impinges on the material. The second layer is a thin layer (denoted by ℓ) of thickness d characterized by a dielectric function $\epsilon_\ell(\omega)$. Is in this layer where the second harmonic generation takes place. The third layer is the bulk region denoted by b and characterized by $\epsilon_b(\omega)$. Both the vacuum layer and the bulk layer are semiinfinite (see Fig. 1.1).

To model the electromagnetic response of the three-layer model we follow Ref. [3], and assume a polarization sheet of the form

$$\mathbf{P}(\mathbf{r},t) = \boldsymbol{\mathcal{P}} e^{i\boldsymbol{\kappa}\cdot\mathbf{R}} e^{-i\omega t}\delta(z - z_\beta) + \text{c.c.}, \tag{1.3}$$

where $\mathbf{R} = (x, y)$, $\boldsymbol{\kappa}$ is the component of the wave vector $\boldsymbol{\nu}_\beta$ paralel to the surface, and z_β is the position of the sheet within medium β (see Fig. 1.1). In Ref. [4] it has been shown that the solution of the Maxwell equations for the radiated fields $E_{\beta,p\pm}$ and $E_{\beta,s}$ with $\mathbf{P}(\mathbf{r}, t)$as a source can be written, at points $z \neq 0$, as

$$(E_{\beta,p\pm}, E_{\beta,s}) = (\frac{\gamma i \tilde{\omega}^2}{\tilde{w}_\beta}\, \hat{\mathbf{p}}_{\beta\pm} \cdot \boldsymbol{\mathcal{P}}, \frac{\gamma i \tilde{\omega}^2}{\tilde{w}_\beta}\, \hat{\mathbf{s}} \cdot \boldsymbol{\mathcal{P}}), \tag{1.4}$$

where $\gamma = 2\pi$ in cgs units and $\gamma = 1/2\epsilon_0$ in MKS units. Also, $\hat{\mathbf{s}}$ and $\hat{\mathbf{p}}_{\beta\pm}$ are the unitary vectors for the s and p polarization of the radiated field, respectively, and the $\pm$ refers to upward $(+)$ or downward $(-)$ direction of propagation within medium β, as shown in Fig. 1.1, and $\tilde{\omega} = \omega/c$. Also, $\tilde{w}_\beta(\omega) = \tilde{\omega} w_\beta$, where

$$w_\beta(\omega) = \left(\epsilon_\beta(\omega) - \sin^2\theta_0\right)^{1/2}, \tag{1.5}$$

where θ_0 is the angle of incidence of $\mathbf{E}_v(\omega)$, and

$$\hat{\mathbf{p}}_{\beta\pm}(\omega) = \frac{\kappa(\omega)\hat{\mathbf{z}} \mp \tilde{w}_\beta(\omega)\hat{\boldsymbol{\kappa}}}{\tilde{\omega} n_\beta(\omega)} = \frac{\sin\theta_0 \hat{\mathbf{z}} \mp w_\beta(\omega)\hat{\boldsymbol{\kappa}}}{n_\beta(\omega)}, \tag{1.6}$$

where $\kappa(\omega) = |\boldsymbol{\kappa}| = \tilde{\omega}\sin\theta_0$, $n_\beta(\omega) = \sqrt{\epsilon_\beta(\omega)}$ is the index of refraction of medium β, and z is the direction perpendicular to the surface that points towards the vacuum. We chose the plane of incidence along the $\boldsymbol{\kappa}z$ plane, then

$$\hat{\boldsymbol{\kappa}} = \cos\phi\hat{\mathbf{x}} + \sin\phi\hat{\mathbf{y}}, \tag{1.7}$$

and

$$\hat{\mathbf{s}} = -\sin\phi\hat{\mathbf{x}} + \cos\phi\hat{\mathbf{y}}, \tag{1.8}$$

where ϕ the angle with respect to the x axis.

In the three-layer model the nonlinear polarization responsible for the second harmonic generation (SHG) is immersed in the thin $\beta = \ell$ layer, and is given by

$$\mathcal{P}_i(2\omega) = \begin{cases} \chi_{ijk}(2\omega)E_j(\omega)E_k(\omega) & \text{(cgs units)} \\ \epsilon_0\chi_{ijk}(2\omega)E_j(\omega)E_k(\omega) & \text{(MKS units)} \end{cases}, \tag{1.9}$$

where the tensor $\boldsymbol{\chi}(2\omega)$ is the surface nonlinear dipolar susceptibility and the Cartesian indices i, j, k are summed if repeated. Also, $\chi_{ijk}(2\omega) = \chi_{ikj}(2\omega)$ is the intrinsic permutation symmetry due to the fact that SHG is degenerate in $E_j(\omega)$ and $E_k(\omega)$. As it was done in Ref. [3], in presenting the results Eq. (1.4)-(1.8) we have taken the polarization sheet (Eq. (1.3)) to be oscillating at some frequency ω. However, in the following we find it convenient to use ω exclusively to denote the fundamental frequency and $\boldsymbol{\kappa}$ to denote the component of the incident wave vector parallel to the surface. Then the nonlinear generated polarization is oscillating at $\Omega = 2\omega$ and will be characterized by a wave vector parallel to the surface $\mathbf{K} = 2\boldsymbol{\kappa}$. We can carry over Eqs. (1.3)-(1.8) simply by replacing the lowercase symbols $(\omega, \tilde{\omega}, \boldsymbol{\kappa}, n_\beta, \tilde{w}_\beta, w_\beta, \hat{\mathbf{p}}_{\beta\pm}, \hat{\mathbf{s}})$ with uppercase

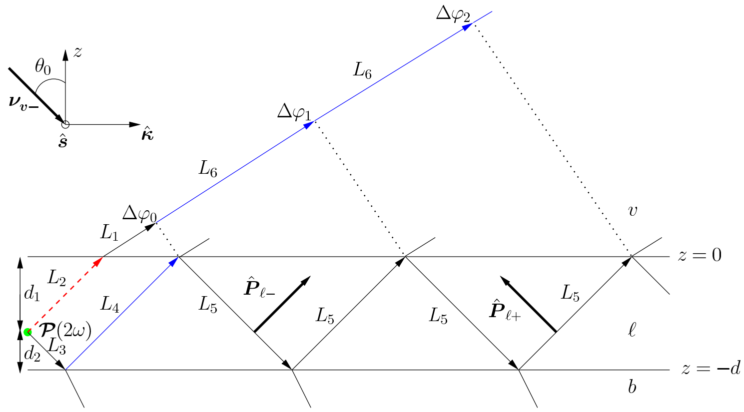

Figure 1.1: (Color Online) Sketch of the three layer model for SHG. Vacuum (v) is on top with $\epsilon_v = 1$; the layer ℓ, of thickness $d = d_1 + d_2$, is characterized with $\epsilon_\ell(\omega)$, and it is where the SH polarization sheet $\boldsymbol{\mathcal{P}}(2\omega)$ is located at $z_\ell = d_1$; The bulk b is described with $\epsilon_b(\omega)$. The arrows point along the direction of propagation, and the p-polarization unit vector, $\hat{\mathbf{P}}_{\ell-(+)}$, along the downward (upward) direction is denoted with a thick arrow. The s-polarization unit vector $\hat{\mathbf{s}}$, points out of the page. The fundamental field $\mathbf{E}(\omega)$ is incident from the vacuum side along the $\hat{\boldsymbol{\kappa}}z$-plane, with θ_0 its angle of incidence and $\boldsymbol{\nu}_{v-}$ its wave vector. $\Delta\varphi_i$ denote the phase difference of the multiply reflected beams with respect to the first vacuum transmitted beam (dashed-red arrow), where the dotted lines are perpendicular to this beam (see the text for details).

symbols $(\Omega, \tilde{\Omega}, \mathbf{K}, N_\beta, \tilde{W}_\beta, W_\beta, \hat{\mathbf{P}}_{\beta\pm}, \hat{\mathbf{S}})$, all evaluated at 2ω and we always have $\hat{\mathbf{S}} = \hat{\mathbf{s}}$.

To describe the propagation of the SH field, we see from Fig. 1.1, that it is refracted at the layer-vacuum interface (ℓv), and multiply reflected from the layer-bulk (ℓb) and layer-vacuum (ℓv) interfaces, thus we can define,

$$\mathbf{T}^{\ell v} = \hat{\mathbf{s}} T_s^{\ell v} \hat{\mathbf{s}} + \hat{\mathbf{P}}_{v+} T_p^{\ell v} \hat{\mathbf{P}}_{\ell +}, \tag{1.10}$$

as the tensor for transmission from ℓv interface,

$$\mathbf{R}^{\ell b} = \hat{\mathbf{s}} R_s^{\ell b} \hat{\mathbf{s}} + \hat{\mathbf{P}}_{\ell +} R_p^{\ell b} \hat{\mathbf{P}}_{\ell -}, \tag{1.11}$$

as the tensor of reflection from the ℓb interface, and

$$\mathbf{R}^{\ell v} = \hat{\mathbf{s}} R_s^{\ell v} \hat{\mathbf{s}} + \hat{\mathbf{P}}_{\ell -} R_p^{\ell v} \hat{\mathbf{P}}_{\ell +}, \tag{1.12}$$

as that of the ℓv interface. The Fresnel factors in uppercase letters, $T_{s,p}^{ij}$ and

$R^{ij}_{s,p}$, are evaluated at 2ω from the following well known formulas

$$\begin{aligned}
t^{ij}_s(\omega) &= \frac{2k_i(\omega)}{k_i(\omega)+k_j(\omega)}, \qquad & t^{ij}_p(\omega) &= \frac{2k_i(\omega)\sqrt{\epsilon_i(\omega)\epsilon_j(\omega)}}{k_i(\omega)\epsilon_j(\omega)+k_j(\omega)\epsilon_i(\omega)}, \\
r^{ij}_s(\omega) &= \frac{k_i(\omega)-k_j(\omega)}{k_i(\omega)+k_j(\omega)}, \qquad & r^{ij}_p(\omega) &= \frac{k_i(\omega)\epsilon_j(\omega)-k_j\epsilon_i(\omega)}{k_i(\omega)\epsilon_j(\omega)+k_j(\omega)\epsilon_i(\omega)}.
\end{aligned} \tag{1.13}$$

From these expressions one can show that,

$$\begin{aligned}
1 + r^{\ell b}_s &= t^{\ell b}_s \\
1 + r^{\ell b}_p &= \frac{n_b}{n_\ell} t^{\ell b}_p \\
1 - r^{\ell b}_p &= \frac{n_\ell}{n_b}\frac{w_b}{w_\ell} t^{\ell b}_p \\
t^{\ell v}_p &= \frac{w_\ell}{w_v} t^{v\ell}_p \\
t^{\ell v}_s &= \frac{w_\ell}{w_v} t^{v\ell}_s .
\end{aligned} \tag{1.14}$$

1.1.1 Multiple SH reflections

The SH field $\mathbf{E}(2\omega)$ radiated by the SH polarization $\boldsymbol{\mathcal{P}}(2\omega)$ will radiate directly into vacuum and also into the bulk, where it will be reflected back at the thin-layer-bulk interface into the thin layer again and this beam will be multiple-transmitted and reflected as shown in Fig. 1.1. As the two beams propagate a phase difference will develop between them, according to

$$\begin{aligned}
\Delta\varphi_m &= \tilde{\omega}\Big((L_3 + L_4 + 2mL_5)N_\ell - \big(L_2 N_\ell + (L_1 + mL_6)N_v\big)\Big) \\
&= \delta_0 + m\delta \quad m = 0, 1, 2, \ldots,
\end{aligned} \tag{1.15}$$

where

$$\delta_0 = 8\pi\left(\frac{d_2}{\lambda_0}\right)\sqrt{n^2_\ell(2\omega) - \sin^2\theta_0}, \tag{1.16}$$

$$\delta = 8\pi\left(\frac{d}{\lambda_0}\right)\sqrt{n^2_\ell(2\omega) - \sin^2\theta_0}, \tag{1.17}$$

where λ_0 is the wavelength of the fundamental field in vacuum, d the thickness of layer ℓ and d_2 the distance of $\boldsymbol{\mathcal{P}}(2\omega)$ from the ℓb interface (see Fig. 1.1). We see that δ_0 is the phase difference of the first and second transmitted beams, and $m\delta$ that of the first and third ($m = 1$), fourth ($m = 2$), etc. beams (see Fig. 1.1).

To take into account the multiple reflections of the generated SH field in the layer ℓ, we proceed as follows. We show the algebra for the p-polarized

SH field, the s-polarized field could be worked out along the same steps. The multiple-reflected $\mathbf{E}_p(2\omega)$ field is given by

$$\begin{aligned}\mathbf{E}(2\omega) &= E_{p+}(2\omega)\mathbf{T}^{\ell v}\cdot\hat{\mathbf{P}}_{\ell+} + E_{p-}(2\omega)\mathbf{T}^{\ell v}\cdot\mathbf{R}^{\ell b}\cdot\hat{\mathbf{P}}_{\ell-}e^{i\Delta\varphi_0} + E_{p-}(2\omega)\mathbf{T}^{\ell v}\cdot\mathbf{R}^{\ell b}\cdot\mathbf{R}^{\ell v}\cdot\mathbf{R}^{\ell b}\cdot\hat{\mathbf{P}}_{\ell-}e^{i\Delta\varphi_1}\\ &+ E_{p-}(2\omega)\mathbf{T}^{\ell v}\cdot\mathbf{R}^{\ell b}\cdot\mathbf{R}^{\ell v}\cdot\mathbf{R}^{\ell b}\cdot\mathbf{R}^{\ell v}\cdot\mathbf{R}^{\ell b}\cdot\hat{\mathbf{P}}_{\ell-}e^{i\Delta\varphi_2}+\cdots\\ &= E_{p+}(2\omega)\mathbf{T}^{\ell v}\cdot\hat{\mathbf{P}}_{\ell+} + E_{p-}(2\omega)\mathbf{T}^{\ell v}\cdot\sum_{m=0}^{\infty}\left(\mathbf{R}^{\ell b}\cdot\mathbf{R}^{\ell v}e^{i\delta}\right)^m\cdot\mathbf{R}^{\ell b}\cdot\hat{\mathbf{P}}_{\ell-}e^{i\delta_0}.\end{aligned} \tag{1.18}$$

From Eqs. (1.10)-(1.12) is easy to show that

$$\mathbf{T}^{\ell v}\cdot\left(\mathbf{R}^{\ell b}\cdot\mathbf{R}^{\ell v}\right)^n\cdot\mathbf{R}^{\ell b} = \hat{\mathbf{s}}T_s^{\ell v}\left(R_s^{\ell b}R_s^{\ell v}\right)^n R_s^{\ell b}\hat{\mathbf{s}} + \hat{\mathbf{P}}_{v+}T_p^{\ell v}\left(R_p^{\ell b}R_p^{\ell v}\right)^n R_p^{\ell b}\hat{\mathbf{P}}_{\ell-}, \tag{1.19}$$

then,

$$\mathbf{E}(2\omega) = \hat{\mathbf{P}}_{\ell+}T_p^{\ell v}\Big(E_{p+}(2\omega) + \frac{R_p^{\ell b}e^{i\delta_0}}{1+R_p^{v\ell}R_p^{\ell b}e^{i\delta}}E_{p-}(2\omega)\Big), \tag{1.20}$$

where we used $R^{ij}_{s,p} = -R^{ji}_{s,p}$. Using Eq. (1.4), we can readily write

$$\mathbf{E}(2\omega) = \frac{\gamma i\tilde{\Omega}}{W_\ell}\mathbf{H}_\ell\cdot\boldsymbol{\mathcal{P}}(2\omega), \tag{1.21}$$

where

$$\mathbf{H}_\ell = \hat{\mathbf{s}}\,T_s^{\ell v}\left(1+R_s^M\right)\hat{\mathbf{s}} + \hat{\mathbf{P}}_{v+}T_p^{\ell v}\left(\hat{\mathbf{P}}_{\ell+} + R_p^M\hat{\mathbf{P}}_{\ell-}\right). \tag{1.22}$$

and

$$R_l^M \equiv \frac{R_l^{\ell b}e^{i\delta_0}}{1+R_l^{v\ell}R_l^{\ell b}e^{i\delta}} \quad l = s,p, \tag{1.23}$$

is defined as the multiple (M) reflection coefficient. To make touch with the work of Ref. [3] where $\boldsymbol{\mathcal{P}}(2\omega)$ is located on top of the vacuum-surface interface and only the vacuum radiated beam and the first (and only) reflected beam need to be considered, we take $\ell = v$ and $d_2 = 0$, then $T^{\ell v} = 1$, $R^{v\ell} = 0$ and $\delta_0 = 0$, with which $R_l^M = R_l^{vb}$. Thus, Eq. (1.22) coincides with Eq. (3.8) of Ref. [3].

1.1.2 SHG Yield

The magnitude of the radiated field is given by $E(2\omega) = \hat{\mathbf{e}}^{\mathrm{out}}\cdot\mathbf{E}(2\omega)$, where $\hat{\mathbf{e}}^{\mathrm{out}}$ is the polarization vector of the radiated field, for instance $\hat{\mathbf{s}}$ or $\hat{\mathbf{P}}_{v+}$. Then, we write

$$
\begin{aligned}
\hat{\mathbf{P}}_{\ell+} + R_p^M \hat{\mathbf{P}}_{\ell-} &= \frac{\sin\theta_0 \hat{\mathbf{z}} - W_\ell \hat{\boldsymbol{\kappa}}}{N_\ell} + R_p^M \frac{\sin\theta_0 \hat{\mathbf{z}} + W_\ell \hat{\boldsymbol{\kappa}}}{N_\ell} \\
&= \frac{1}{N_\ell}\left(\sin\theta_0 R_{p+}^M \hat{\mathbf{z}} - K_\ell R_{p-}^M \hat{\boldsymbol{\kappa}}\right),
\end{aligned}
\tag{1.24}
$$

where

$$
R_l^{M\pm} \equiv 1 \pm R_l^M \quad l = s, p. \tag{1.25}
$$

Using Eq. (1.14) we write Eq. (1.21) as

$$
E(2\omega) = \frac{2\gamma i\omega}{cW_\ell} \hat{\mathbf{e}}^{\mathrm{out}} \cdot \mathbf{H}_\ell \cdot \boldsymbol{\mathcal{P}}(2\omega) = \frac{2\gamma i\omega}{cW_v} \mathbf{e}_\ell^{2\omega} \cdot \boldsymbol{\mathcal{P}}(2\omega). \tag{1.26}
$$

$$
\mathbf{e}_\ell^{2\omega} = \hat{\mathbf{e}}^{\mathrm{out}} \cdot \left[\hat{\mathbf{s}} T_s^{v\ell} R_s^{M+} \hat{\mathbf{s}} + \hat{\mathbf{P}}_{v+} \frac{T_p^{v\ell}}{N_\ell}\left(\sin\theta_0 R_p^{M+} \hat{\mathbf{z}} - W_\ell R_p^{M-} \hat{\boldsymbol{\kappa}}\right)\right]. \tag{1.27}
$$

We pause here to reduce above result to the case where the nonlinear polarization $\mathbf{P}(2\omega)$ radiates from vacuum instead from the layer ℓ. For such case we simply take $\epsilon_\ell(2\omega) = 1$ and $\ell = v$ ($T_{s,p}^{\ell v} = 1$), to get

$$
\mathbf{e}_v^{2\omega} = \hat{\mathbf{e}}^{\mathrm{out}} \cdot \left[\hat{\mathbf{s}} T_s^{vb} \hat{\mathbf{s}} + \hat{\mathbf{P}}_{v+} \frac{T_p^{vb}}{\sqrt{\epsilon_b(2\omega)}} \left(\epsilon_b(2\omega)\sin\theta_0 \hat{\mathbf{z}} - W_b \hat{\boldsymbol{\kappa}}\right)\right], \tag{1.28}
$$

which agrees with Eq. (3.10) of Ref. [3].

In the three layer model the SH polarization $\surd(2\omega)$ is located in layer ℓ, where we evaluate the fundamental field required in Eq. (1.9). We write

$$
\mathbf{E}_\ell(\omega) = E_0\left(\hat{\mathbf{s}} t_s^{v\ell}(1 + r_s^{\ell b})\hat{\mathbf{s}} + \hat{\mathbf{p}}_{\ell-} t_p^{v\ell} \hat{\mathbf{p}}_{v-} + \hat{\mathbf{p}}_{\ell+} t_p^{v\ell} r_p^{\ell b} \hat{\mathbf{p}}_{v-}\right) \cdot \hat{\mathbf{e}}^{\mathrm{in}} = E_0 \mathbf{e}_\ell^{\omega}, \tag{1.29}
$$

where $\mathbf{e}^{\mathrm{in}}$ is the s ($\hat{\mathbf{s}}$) or p ($\hat{\mathbf{p}}_{v-}$) incoming polarization of the fundamental electric field. Above field is composed of the transmitted field and its first reflection from the ℓb interface for s and p polarizations. The fundamental field, once inside the layer ℓ will be multiply reflected at the ℓv and ℓb interfaces, however each reflection will diminish the intensity of the fundamental field, and as the SHG yield goes with the square of this field, the contribution of the subsequent reflections, other than the one considered in Eq. (1.29), could be safely neglected. From Eq. (1.14) we find that

$$
\mathbf{e}_\ell^{\omega} = \left[\hat{\mathbf{s}} t_s^{v\ell} t_s^{\ell b} \hat{\mathbf{s}} + \frac{t_p^{v\ell} t_p^{\ell b}}{n_\ell^2 n_b}\left(n_b^2 \sin\theta_0 \hat{\mathbf{z}} + n_\ell^2 w_b \hat{\boldsymbol{\kappa}}\right)\hat{\mathbf{p}}_{v-}\right] \cdot \hat{\mathbf{e}}^{\mathrm{in}}. \tag{1.30}
$$

Again, to touch base with Ref. [3], if we would like to evaluate the fields in the bulk, instead of the layer ℓ, we simply take $n_\ell = n_b$, ($t_{s,p}^{\ell b} = 1$), to obtain

$$
\mathbf{e}_b^{\omega} = \left[\hat{\mathbf{s}} t_s^{vb} \hat{\mathbf{s}} + \frac{t_p^{vb}}{n_b}\left(\sin\theta_0 \hat{\mathbf{z}} + w_b \hat{\boldsymbol{\kappa}}\right)\hat{\mathbf{p}}_{v-}\right] \cdot \hat{\mathbf{e}}^{\mathrm{in}}, \tag{1.31}
$$

that is in agreement with Eq. (3.5) of Ref. [3]. Then, we can write Eq. (1.9) as

$$\boldsymbol{\mathcal{P}}(2\omega) = \begin{cases} E_0^2\,\boldsymbol{\chi} : \mathbf{e}_\ell^\omega \mathbf{e}_\ell^\omega & \text{(cgs units)} \\ \epsilon_0 E_0^2\,\boldsymbol{\chi} : \mathbf{e}_\ell^\omega \mathbf{e}_\ell^\omega & \text{(MKS units)} \end{cases}, \tag{1.32}$$

where E_0 is the intensity of the fundamental electric field. Finally, with above equation we write Eq. (1.26) as

$$E(2\omega) = \frac{2\eta i\omega}{cW_v}\mathbf{e}_\ell^{2\omega}\cdot\boldsymbol{\chi} : \mathbf{e}_\ell^\omega \mathbf{e}_\ell^\omega, \tag{1.33}$$

where $\eta = 2\pi$ for cgs units and $\eta = 1/2$ for MKS units. To ease on the notation, we define

$$\Upsilon_{\mathrm{iO}} \equiv \mathbf{e}_\ell^{2\omega}\cdot\boldsymbol{\chi} : \mathbf{e}_\ell^\omega \mathbf{e}_\ell^\omega, \tag{1.34}$$

where i stands for the incoming polarization of the fundamental electric field given by $\hat{\mathbf{e}}^{\mathrm{in}}$ in Eq. (1.30), and O for the outgoing polarization of the SH electric field given by $\hat{\mathbf{e}}^{\mathrm{out}}$ in Eq. (1.27).

From Eqs. (1.1) and (1.2) we obtain that in the cgs units ($\eta = 2\pi$)

$$\begin{aligned}
|E(2\omega)|^2 &= |E_0|^4 \frac{16\pi^2\omega^2}{c^2 W_v^2}\,|\Upsilon_{\mathrm{iO}}|^2 \\
\frac{c}{2\pi}|\sqrt{N_v}E(2\omega)|^2 &= \frac{32\pi^3\omega^2}{c^3\cos^2\theta_0}\left|\frac{\sqrt{N_v}}{n_\ell^2}\Upsilon_{\mathrm{iO}}\right|^2\left(\frac{c}{2\pi}|\sqrt{n_\ell}E_0|^2\right)^2, \\
I(2\omega) &= \frac{32\pi^3\omega^2}{c^3\cos^2\theta_0}\left|\frac{\sqrt{N_v}}{n_\ell^2}\Upsilon_{\mathrm{iO}}\right|^2 I^2(\omega), \\
\mathcal{R}_{\mathrm{iO}}(2\omega) &= \frac{32\pi^3\omega^2}{c^3\cos^2\theta_0}\left|\frac{1}{n_\ell^2}\Upsilon_{\mathrm{iO}}\right|^2,
\end{aligned} \tag{1.35}$$

and in MKS units ($\eta = 1/2$)

$$\begin{aligned}
|E(2\omega)|^2 &= |E_0|^4 \frac{\omega^2}{c^2 W_v^2}\,|\Upsilon_{\mathrm{iO}}|^2 \\
2\epsilon_0 c|\sqrt{N_v}E(2\omega)|^2 &= \frac{2\epsilon_0\omega^2}{c\cos^2\theta_0}\left|\frac{\sqrt{N_v}}{n_\ell^2}\Upsilon_{\mathrm{iO}}\right|^2 \frac{1}{4\epsilon_0^2 c^2}\left(2\epsilon_0 c|\sqrt{n_\ell}E_0|^2\right)^2, \\
I(2\omega) &= \frac{\omega^2}{2\epsilon_0 c^3\cos^2\theta_0}\left|\frac{\sqrt{N_v}}{n_\ell^2}\Upsilon_{\mathrm{iO}}\right|^2 I^2(\omega), \\
\mathcal{R}_{\mathrm{iO}}(2\omega) &= \frac{\omega^2}{2\epsilon_0 c^3\cos^2\theta_0}\left|\frac{1}{n_\ell^2}\Upsilon_{\mathrm{iO}}\right|^2,
\end{aligned} \tag{1.36}$$

$$\mathcal{R}_{\mathrm{iO}}(2\omega) \begin{cases} \frac{32\pi^3\omega^2}{c^3\cos^2\theta_0}\left|\frac{1}{n_\ell^2}\Upsilon_{\mathrm{iO}}\right|^2 & \text{(cgs units)} \\ \frac{\omega^2}{2\epsilon_0 c^3\cos^2\theta_0}\left|\frac{1}{n_\ell^2}\Upsilon_{\mathrm{iO}}\right|^2 & \text{(MKS units)} \end{cases}, \tag{1.37}$$

as the SHG yield, where $N_v = 1$ and $W_v = \cos\theta_0$. In the MKS unit system $\boldsymbol{\chi}$ is given in m^2/V, since it is a surface second order nonlinear susceptibility, and $\mathcal{R}_{\mathrm{iO}}$ is given in m^2/W.

`tal vez esto al apendice` At this point we mention that to recover the results of Ref. [3] which are equivalent of those of Ref. [5], we take $\mathbf{e}_\ell^{2\omega} \to \mathbf{e}_v^{2\omega}$, $\mathbf{e}_\ell^{\omega} \to \mathbf{e}_b^{\omega}$, and then

$$\mathcal{R}(2\omega) = \frac{32\pi^3\omega^2}{c^3\cos^2\theta_0}\left|\mathbf{e}_v^{2\omega}\cdot\boldsymbol{\chi}:\mathbf{e}_b^{\omega}\mathbf{e}_b^{\omega}\right|^2, \tag{1.38}$$

will give the SHG yield of a nonlinear polarization sheet radiating from vacuum on top of the surface and where the fundamental field is evaluated below the surface that is characterized by $\epsilon_b(\omega)$.

1.2 One SH Reflection

Therefore, the total radiated field at 2ω is

$$\begin{aligned}\mathbf{E}(2\omega) = {} & E_s(2\omega)\left(\mathbf{T}^{\ell v} + \mathbf{T}^{\ell v}\cdot\mathbf{R}^{\ell b}\right)\cdot\hat{\mathbf{s}} \\ & + E_{p+}(2\omega)\mathbf{T}^{\ell v}\cdot\hat{\mathbf{P}}_{\ell +} + E_{p-}(2\omega)\mathbf{T}^{\ell v}\cdot\mathbf{R}^{\ell b}\cdot\hat{\mathbf{P}}_{\ell -}.\end{aligned}$$

The first term is the transmitted s-polarized field, the second one is the reflected and then transmitted s-polarized field and the third and fourth terms are the equivalent fields for p-polarization. The transmission is from the layer into vacuum, and the reflection between the layer and the bulk. After some simple algebra, we obtain

$$\mathbf{E}(2\omega) = \frac{2\pi i\tilde{\Omega}}{K_\ell}\mathbf{H}_\ell\cdot\boldsymbol{\mathcal{P}}(2\omega), \tag{1.39}$$

where,

$$\mathbf{H}_\ell = \hat{\mathbf{s}}\,T_s^{\ell v}\left(1 + R_s^{\ell b}\right)\hat{\mathbf{s}} + \hat{\mathbf{P}}_{v+}T_p^{\ell v}\left(\hat{\mathbf{P}}_{\ell +} + R_p^{\ell b}\hat{\mathbf{P}}_{\ell -}\right). \tag{1.40}$$

1.3 $\mathcal{R}$ for different polarization cases

1.3.1 $\mathcal{R}_{pP}$

We develop five different scenarios for $\mathcal{R}_{pP}$ that explore different cases for where the polarization and fundamental fields are located. In all these scenarios, we use $\hat{\mathbf{e}}^{\mathrm{in}} = \hat{\mathbf{p}}_{v-}$ in Eq. (1.30), and $\hat{\mathbf{e}}^{\mathrm{out}} = \hat{\mathbf{P}}_{v+}$ in Eq. (**??**).

This scenario involves $\mathcal{P}(2\omega)$ and the fundamental fields to be taken in a thin layer of material below the surface, which we designate as ℓ. Thus,

$$\mathbf{e}_\ell^{2\omega}\cdot\boldsymbol{\chi}:\mathbf{e}_\ell^{\omega}\mathbf{e}_\ell^{\omega} \equiv \Gamma_{pP}^{\ell}\,r_{pP}^{\ell},$$

where

$$\begin{aligned}r_{pP}^{\ell} = {} & \epsilon_b(2\omega)\sin\theta_{\mathrm{in}}\Big(\epsilon_b^2(\omega)\sin^2\theta_{\mathrm{in}}\chi_{zzz} + \epsilon_\ell^2(\omega)k_b^2\chi_{zxx}\Big) \\ & - \epsilon_\ell(2\omega)\epsilon_\ell(\omega)k_bK_b\Big(2\epsilon_b(\omega)\sin\theta_{\mathrm{in}}\chi_{xxz} + \epsilon_\ell(\omega)k_b\chi_{xxx}\cos(3\phi)\Big),\end{aligned} \tag{1.41}$$

and

$$\Gamma^{\ell}_{pP} = \frac{T_p^{\ell v} T_p^{\ell b}}{\epsilon_\ell(2\omega)\sqrt{\epsilon_b(2\omega)}} \left(\frac{t_p^{v\ell} t_p^{\ell b}}{\epsilon_\ell(\omega)\sqrt{\epsilon_b(\omega)}} \right)^2. \tag{1.42}$$

1.3.2 $\mathcal{R}_{pS}$

To obtain $R_{pS}(2\omega)$ we use $\hat{\mathbf{e}}^{\mathrm{in}} = \hat{\mathbf{p}}_{v-}$ in Eq. (1.30), and $\hat{\mathbf{e}}^{\mathrm{out}} = \hat{\mathbf{S}}$ in Eq. (**??**). We also use the unit vectors defined in Eqs. (A.1) and (A.2). Substituting, we get

$$\mathbf{e}_\ell^{2\omega} \cdot \boldsymbol{\chi} : \mathbf{e}_\ell^{\omega} \mathbf{e}_\ell^{\omega} \equiv \Gamma^{\ell}_{sP}\, r^{\ell}_{sP},$$

where

$$r^{\ell}_{pS} = -\epsilon_\ell^2(\omega) k_b^2 \sin 3\phi \chi_{xxx}, \tag{1.43}$$

and

$$\Gamma^{\ell}_{pS} = T_s^{v\ell} T_s^{\ell b} \left(\frac{t_p^{v\ell} t_p^{\ell b}}{\epsilon_\ell(\omega)\sqrt{\epsilon_b(\omega)}} \right)^2. \tag{1.44}$$

In order to reduce above result to that of Ref. [3] and [5], we take the 2ω radiations factors for vacuum by taking $\ell = v$, thus $\epsilon_\ell(2\omega) = 1$, $T_s^{v\ell} = 1$, $T_s^{\ell b} = T_s^{vb}$, and the fundamental field inside medium b by taking $\ell = b$, thus $\epsilon_\ell(\omega) = \epsilon_b(\omega)$, $t_p^{v\ell} = t_p^{vb}$, and $t_p^{\ell b} = 1$. With these choices,

$$r^{b}_{pS} = -k_b^2 \sin 3\phi \chi_{xxx},$$

and

$$\Gamma^{b}_{pS} = T_s^{vb} \left(\frac{t_p^{vb}}{\sqrt{\epsilon_b(\omega)}} \right)^2.$$

1.3.3 $\mathcal{R}_{sP}$

To obtain $R_{sP}(2\omega)$ we use $\hat{\mathbf{e}}^{\mathrm{in}} = \hat{\mathbf{s}}$ in Eq. (1.30), and $\hat{\mathbf{e}}^{\mathrm{out}} = \hat{\mathbf{P}}_{v+}$ in Eq. (**??**). We also use the unit vectors defined in Eqs. (A.1) and (A.2). Substituting, we get

$$\mathbf{e}_\ell^{2\omega} \cdot \boldsymbol{\chi} : \mathbf{e}_\ell^{\omega} \mathbf{e}_\ell^{\omega} \equiv \Gamma^{\ell}_{sP}\, r^{\ell}_{sP},$$

where

$$r^{\ell}_{sP} = \epsilon_b(2\omega) \sin\theta_{\mathrm{in}} \chi_{zxx} + \epsilon_\ell(2\omega) K_b \chi_{xxx} \cos 3\phi, \tag{1.45}$$

and

$$\Gamma^{\ell}_{sP} = \frac{T_p^{\ell v} T_p^{\ell b} \left(t_s^{v\ell} t_s^{\ell b} \right)^2}{\epsilon_\ell(2\omega)\sqrt{\epsilon_b(2\omega)}}. \tag{1.46}$$

In order to reduce above result to that of Ref. [3] and [5], we take the 2ω radiations factors for vacuum by taking $\ell = v$, thus $\epsilon_\ell(2\omega) = 1$, $T_p^{v\ell} = 1$, $T_p^{\ell b} = T_p^{vb}$, and the fundamental field inside medium b by taking $\ell = b$, thus $\epsilon_\ell(\omega) = \epsilon_b(\omega)$, $t_s^{v\ell} = t_s^{vb}$, and $t_s^{\ell b} = 1$. With these choices,

$$r^{b}_{sP} = \epsilon_b(2\omega) \sin\theta_{\mathrm{in}} \chi_{zxx} + K_b \chi_{xxx} \cos 3\phi,$$

iF	Γ^{ℓ}_{iF}	r^{ℓ}_{iF}
pP	$\frac{T_p^{v\ell}}{N_\ell}\left(\frac{t_p^{v\ell}t_p^{\ell b}}{n_\ell^2 n_b}\right)^2$	$R_p^{M+}\sin\theta_0(n_b^4\sin^2\theta_0\chi_{zzz}+n_\ell^4 w_b^2\chi_{zxx})$ $-R_p^{M-}n_\ell^2 w_b W_\ell(2n_b^2\sin\theta_0\chi_{xxz}+n_\ell^2 w_b\chi_{xxx}\cos 3\phi)$
pS	$T_s^{v\ell}R_s^{M+}\left(\frac{t_p^{v\ell}t_p^{\ell b}}{n_\ell^2 n_b}\right)^2$	$-n_\ell^4 w_b^2\chi_{xxx}\sin 3\phi$
sP	$\frac{T_p^{v\ell}}{N_\ell}\left(t_s^{v\ell}t_s^{\ell b}\right)^2$	$R_p^{M+}\sin\theta_0\chi_{zxx}+R_p^{M-}W_\ell\chi_{xxx}\cos 3\phi$
sS	$T_s^{v\ell}R_s^{M+}\left(t_s^{v\ell}t_s^{\ell b}\right)^2$	$\chi_{xxx}\sin 3\phi$

Table 1.1: The expressions needed to calculate the SHG yield for the (111) surface, for each polarization case.

and

$$\Gamma^{b}_{sP}=\frac{T_p^{vb}(t_s^{vb})^2}{\sqrt{\epsilon_b(2\omega)}}.$$

1.3.4 $\mathcal{R}_{sS}$

For $\mathcal{R}_{sS}$ we have that $\hat{\mathbf{e}}^{\rm in}=\hat{\mathbf{s}}$ and $\hat{\mathbf{e}}^{\rm out}=\hat{\mathbf{S}}$. This leads to

$$\mathbf{e}_\ell^{2\omega}\cdot\boldsymbol{\chi}:\mathbf{e}_\ell^{\omega}\mathbf{e}_\ell^{\omega}\equiv\Gamma^{\ell}_{sS}\,r^{\ell}_{sS},$$

where

$$r^{\ell}_{sS}=\chi_{xxx}\sin 3\phi, \tag{1.47}$$

and

$$\Gamma^{\ell}_{sS}=T_s^{v\ell}T_s^{\ell b}\left(t_s^{v\ell}t_s^{\ell b}\right)^2. \tag{1.48}$$

In order to reduce above result to that of Ref. [3] and [5], we take the 2ω radiations factors for vacuum by taking $\ell=v$, thus $\epsilon_\ell(2\omega)=1$, $T_s^{v\ell}=1$, $T_s^{\ell b}=T_s^{vb}$, and the fundamental field inside medium b by taking $\ell=b$, thus $\epsilon_\ell(\omega)=\epsilon_b(\omega)$, $t_s^{v\ell}=t_s^{vb}$, and $t_s^{\ell b}=1$. With these choices,

$$r^{b}_{sS}=\chi_{xxx}\sin 3\phi,$$

and

$$\Gamma^{b}_{sS}=T_s^{vb}\left(t_s^{vb}\right)^2.$$

1.3.5 Summary

We present the final expressions for each polarization case in Table 1.1.

Appendix A

Derived expressions for the SHG yield

A.1 Some useful expressions

We are interested in finding

$$\Upsilon = \mathbf{e}_\ell^{2\omega} \cdot \boldsymbol{\chi} : \mathbf{e}_\ell^{\omega} \mathbf{e}_\ell^{\omega}$$

for each different polarization case. We choose the plane of incidence along the $\boldsymbol{\kappa} z$ plane, and define

$$\hat{\boldsymbol{\kappa}} = \cos\phi \hat{\mathbf{x}} + \sin\phi \hat{\mathbf{y}}, \tag{A.1}$$

and

$$\hat{\mathbf{s}} = -\sin\phi \hat{\mathbf{x}} + \cos\phi \hat{\mathbf{y}}, \tag{A.2}$$

where ϕ the angle with respect to the x axis.

A.1.1 2ω terms

Including multiple reflecions, the $\mathbf{e}_\ell^{2\omega}$ term is

$$\mathbf{e}_\ell^{2\omega} = \hat{\mathbf{e}}^{\mathrm{out}} \cdot \left[\hat{\mathbf{s}} T_s^{v\ell} R_s^{M+} \hat{\mathbf{s}} + \hat{\mathbf{P}}_{v+} \frac{T_p^{v\ell}}{N_\ell} \left(\sin\theta_0 R_p^{M+} \hat{\mathbf{z}} - W_\ell R_p^{M-} \hat{\boldsymbol{\kappa}} \right) \right], \tag{A.3}$$

and neglecting the multiple reflections reduces this expression to

$$\mathbf{e}_\ell^{2\omega} = \hat{\mathbf{e}}^{\mathrm{out}} \cdot \left[\hat{\mathbf{s}} T_s^{v\ell} T_s^{\ell b} \hat{\mathbf{s}} + \hat{\mathbf{P}}_{v+} \frac{T_p^{v\ell} T_p^{\ell b}}{N_\ell^2 N_b} \left(N_b^2 \sin\theta_0 \hat{\mathbf{z}} - N_\ell^2 W_b \hat{\mathbf{x}} \right) \right]. \tag{A.4}$$

We first expand these equations for clarity. Substituting Eqs. (A.1) and (A.2) into Eq. (A.3),

$$\mathbf{e}_\ell^{2\omega} = \hat{\mathbf{e}}^{\text{out}} \cdot \Bigg[\hat{\mathbf{s}} T_s^{v\ell} R_s^{M+} \left(-\sin\phi\hat{\mathbf{x}} + \cos\phi\hat{\mathbf{y}}\right) + \hat{\mathbf{P}}_{v+} \frac{T_p^{v\ell}}{N_\ell} \left(\sin\theta_0 R_p^{M+}\hat{\mathbf{z}} - W_\ell R_p^{M-}\cos\phi\hat{\mathbf{x}} - W_\ell R_p^{M-}\sin\phi\hat{\mathbf{y}}\right)\Bigg].$$

We now have $\mathbf{e}_\ell^{2\omega}$ in terms of $\hat{\mathbf{P}}_{v+}$,

$$\mathbf{e}_\ell^{2\omega} = \frac{T_p^{v\ell}}{N_\ell} \left(\sin\theta_0 R_p^{M+}\hat{\mathbf{z}} - W_\ell R_p^{M-}\cos\phi\hat{\mathbf{x}} - W_\ell R_p^{M-}\sin\phi\hat{\mathbf{y}}\right), \tag{A.5}$$

and in terms of $\hat{\mathbf{s}}$,

$$\mathbf{e}_\ell^{2\omega} = T_s^{v\ell} R_s^{M+} \left(-\sin\phi\hat{\mathbf{x}} + \cos\phi\hat{\mathbf{y}}\right). \tag{A.6}$$

Likewise, we do the exact same for Eq. (A.4), and get the following term for $\hat{\mathbf{P}}_{v+}$,

$$\mathbf{e}_\ell^{2\omega} = \frac{T_p^{v\ell} T_p^{\ell b}}{N_\ell^2 N_b} \left(N_b^2 \sin\theta_0\hat{\mathbf{z}} - N_\ell^2 W_b\cos\phi\hat{\mathbf{x}} - N_\ell^2 W_b\sin\phi\hat{\mathbf{y}}\right), \tag{A.7}$$

and $\hat{\mathbf{s}}$,

$$\mathbf{e}_\ell^{2\omega} = T_s^{v\ell} T_s^{\ell b} \left[-\sin\phi\hat{\mathbf{x}} + \cos\phi\hat{\mathbf{y}}\right]. \tag{A.8}$$

A.1.2 1ω terms

We posit that the effects of the multiple reflections can be neglected for the $\mathbf{e}_\ell^{\omega}$ term. This term is

$$\mathbf{e}_\ell^{\omega} = \left[\hat{\mathbf{s}} t_s^{v\ell} t_s^{\ell b}\hat{\mathbf{s}} + \frac{t_p^{v\ell} t_p^{\ell b}}{n_\ell^2 n_b} \left(n_b^2 \sin\theta_0\hat{\mathbf{z}} + n_\ell^2 w_b \hat{\boldsymbol{\kappa}}\right)\hat{\mathbf{p}}_{v-}\right] \cdot \hat{\mathbf{e}}^{\text{in}}.$$

For all cases, we require a $\mathbf{e}_\ell^{\omega}\mathbf{e}_\ell^{\omega}$ product. For brevity, we will directly list these terms for both polarizations. For $\hat{\mathbf{e}}^{\text{in}} = \hat{\mathbf{p}}_{v-}$,

$$\begin{aligned}\mathbf{e}_\ell^{\omega}\mathbf{e}_\ell^{\omega} = \left(\frac{t_p^{v\ell} t_p^{\ell b}}{n_\ell^2 n_b}\right)^2 \big(&n_\ell^4 w_b^2 \cos^2\phi\hat{\mathbf{x}}\hat{\mathbf{x}} + 2n_\ell^4 w_b^2 \sin\phi\cos\phi\hat{\mathbf{x}}\hat{\mathbf{y}} \\ &+ 2n_\ell^2 n_b^2 w_b \sin\theta_0\cos\phi\hat{\mathbf{x}}\hat{\mathbf{z}} + n_\ell^4 w_b^2 \sin^2\phi\hat{\mathbf{y}}\hat{\mathbf{y}} \\ &+ 2n_\ell^2 n_b^2 w_b \sin\theta_0\sin\phi\hat{\mathbf{y}}\hat{\mathbf{z}} + n_b^4 \sin^2\theta_0\hat{\mathbf{z}}\hat{\mathbf{z}}\big),\end{aligned} \tag{A.9}$$

and for $\hat{\mathbf{e}}^{\text{in}} = \hat{\mathbf{s}}$,

$$\mathbf{e}_\ell^{\omega}\mathbf{e}_\ell^{\omega} = \left(t_s^{v\ell} t_s^{\ell b}\right)^2 \left(\sin^2\phi\hat{\mathbf{x}}\hat{\mathbf{x}} + \cos^2\phi\hat{\mathbf{y}}\hat{\mathbf{y}} - 2\sin\phi\cos\phi\hat{\mathbf{x}}\hat{\mathbf{y}}\right). \tag{A.10}$$

We summarize these expressions in Table A.1. In order to derive the equations for a given polarization case, we refer to the equations listed there. Then it is simply a matter of multiplying the terms correctly and obtaining the appropriate components of $\boldsymbol{\chi}(-2\omega;\omega,\omega)$.

A.1.3 Nonzero components of $\boldsymbol{\chi}(-2\omega;\omega,\omega)$

For a (111) surface with C_{3v} symmetry, we have the following nonzero components:

$$\begin{aligned}
&\chi_{xxx} = -\chi_{xyy} = -\chi_{yyx},\\
&\chi_{xxz} = \chi_{yyz},\\
&\chi_{zxx} = \chi_{zyy},\\
&\chi_{zzz}.
\end{aligned} \tag{A.11}$$

For a (110) surface with C_{2v} symmetry, we have the following nonzero components:

$$\chi_{xxz}, \chi_{yyz}, \chi_{zxx}, \chi_{zyy}, \chi_{zzz}. \tag{A.12}$$

Lastly, for a (001) surface with C_{4v} symmetry, we have the following nonzero components:

$$\begin{aligned}
&\chi_{xxz} = \chi_{yyz},\\
&\chi_{zxx} = \chi_{zyy},\\
&\chi_{zzz}.
\end{aligned} \tag{A.13}$$

Case	$\hat{\mathbf{e}}^{\text{out}}$	$\hat{\mathbf{e}}^{\text{in}}$	$\mathbf{e}_\ell^{2\omega}$	$\mathbf{e}_\ell^{\omega}\mathbf{e}_\ell^{\omega}$
$\mathcal{R}_{pP}$	$\hat{\mathbf{P}}_{v+}$	$\hat{\mathbf{p}}_{v-}$	Eq. (A.5) or (A.7)	Eq. (A.9)
$\mathcal{R}_{pS}$	$\hat{\mathbf{s}}$	$\hat{\mathbf{p}}_{v-}$	Eq. (A.6) or (A.8)	Eq. (A.9)
$\mathcal{R}_{sP}$	$\hat{\mathbf{P}}_{v+}$	$\hat{\mathbf{s}}$	Eq. (A.5) or (A.7)	Eq. (A.10)
$\mathcal{R}_{sS}$	$\hat{\mathbf{s}}$	$\hat{\mathbf{s}}$	Eq. (A.6) or (A.8)	Eq. (A.10)

Table A.1: Polarization unit vectors for $\hat{\mathbf{e}}^{\text{out}}$ and $\hat{\mathbf{e}}^{\text{in}}$, and equations describing $\mathbf{e}_\ell^{2\omega}$ and $\mathbf{e}_\ell^{\omega}\mathbf{e}_\ell^{\omega}$ for each polarization case. When there are two equations to choose from, the former includes the effects of multiple reflections, and the latter neglects them.

A.2 $\mathcal{R}_{pP}$

Per Table A.1, $\mathcal{R}_{pP}$ requires Eqs. (A.7) and (A.9). After some algebra, we obtain that

$$\begin{aligned}\Upsilon_{pP}^{\mathrm{MR}} = \Gamma_{pP}^{\mathrm{MR}}\Big[& -R_p^{M-}W_\ell\big(n_\ell^4 w_b^2\cos^3\phi\chi_{xxx} + 2n_\ell^4 w_b^2\sin\phi\cos^2\phi\chi_{xxy} \\ & \qquad + 2n_\ell^2 n_b^2 w_b\sin\theta_0\cos^2\phi\chi_{xxz} + n_\ell^4 w_b^2\sin^2\phi\cos\phi\chi_{xyy} \\ & \qquad + 2n_\ell^2 n_b^2 w_b\sin\theta_0\sin\phi\cos\phi\chi_{xyz} + n_b^4\sin^2\theta_0\cos\phi\chi_{xzz}\big) \\ & -R_p^{M-}W_\ell\big(n_\ell^4 w_b^2\sin\phi\cos^2\phi\chi_{yxx} + 2n_\ell^4 w_b^2\sin^2\phi\cos\phi\chi_{yxy} \\ & \qquad + 2n_\ell^2 n_b^2 w_b\sin\theta_0\sin\phi\cos\phi\chi_{yxz} + n_\ell^4 w_b^2\sin^3\phi\chi_{yyy} \\ & \qquad + 2n_\ell^2 n_b^2 w_b\sin\theta_0\sin^2\phi\chi_{yyz} + n_b^4\sin^2\theta_0\sin\phi\chi_{yzz}\big) \\ & +R_p^{M+}\sin\theta_0\big(n_\ell^4 w_b^2\cos^2\phi\chi_{zxx} + 2n_\ell^4 w_b^2\sin\phi\cos\phi\chi_{zxy} \\ & \qquad + n_\ell^4 w_b^2\sin^2\phi\chi_{zyy} + 2n_\ell^2 n_b^2 w_b\sin\theta_0\cos\phi\chi_{zzx} \\ & \qquad + 2n_\ell^2 n_b^2 w_b\sin\theta_0\sin\phi\chi_{zzy} + n_b^4\sin^2\theta_0\chi_{zzz}\big)\Big].\end{aligned} \tag{A.14}$$

We take this opportunity to introduce a quantity that will be repeated throughout this section,

$$\Gamma_{pP}^{\mathrm{MR}} = \frac{T_p^{v\ell}}{N_\ell}\left(\frac{t_p^{v\ell}t_p^{\ell b}}{n_\ell^2 n_b}\right)^2. \tag{A.15}$$

If we neglect the multiple reflections, as described in the manuscript, we have that

$$\begin{aligned}\Upsilon_{pP} = \Gamma_{pP}\Big[& -N_\ell^2 W_b\big(+ n_\ell^4 w_b^2 \cos^3\phi\chi_{xxx} + 2n_\ell^4 w_b^2 \sin\phi\cos^2\phi\chi_{xxy} \\ & \qquad + 2n_b^2 n_\ell^2 w_b \sin\theta_0\cos^2\phi\chi_{xxz} + n_\ell^4 w_b^2 \sin^2\phi\cos\phi\chi_{xyy} \\ & \qquad + 2n_b^2 n_\ell^2 w_b \sin\theta_0\sin\phi\cos\phi\chi_{xyz} + n_b^4\sin^2\theta_0\cos\phi\chi_{xzz}\big) \\ & -N_\ell^2 W_b\big(+ n_\ell^4 w_b^2 \sin\phi\cos^2\phi\chi_{yxx} + 2n_\ell^4 w_b^2 \sin^2\phi\cos\phi\chi_{yxy} \\ & \qquad + 2n_b^2 n_\ell^2 w_b \sin\theta_0\sin\phi\cos\phi\chi_{yxz} + n_\ell^4 w_b^2 \sin^3\phi\chi_{yyy} \\ & \qquad + 2n_b^2 n_\ell^2 w_b \sin\theta_0\sin^2\phi\chi_{yyz} + n_b^4\sin^2\theta_0\sin\phi\chi_{yzz}\big) \\ & +N_b^2\sin\theta_0\big(+ n_\ell^4 w_b^2 \cos^2\phi\chi_{zxx} + 2n_\ell^4 w_b^2 \sin\phi\cos\phi\chi_{zxy} \\ & \qquad + n_\ell^4 w_b^2 \sin^2\phi\chi_{zyy} + 2n_\ell^2 n_b^2 w_b \sin\theta_0\cos\phi\chi_{zzx} \\ & \qquad + 2n_\ell^2 n_b^2 w_b \sin\theta_0\sin\phi\chi_{zzy} + n_b^4\sin^2\theta_0\chi_{zzz}\big)\Big],\end{aligned} \tag{A.16}$$

and again we introduce a quantity that will be repeated throughout this section,

$$\Gamma_{pP} = \frac{T_p^{v\ell}T_p^{\ell b}}{N_\ell^2 N_b}\left(\frac{t_p^{v\ell}t_p^{\ell b}}{n_\ell^2 n_b}\right)^2. \tag{A.17}$$

A.2.1 For the (111) surface

We take Eqs. (A.14) and (A.11), eliminate the components that do not contribute, and apply the the symmetry relations as follows,

$$\begin{aligned}\Upsilon_{pP}^{\mathrm{MR},(111)} = \Gamma_{pP}^{\mathrm{MR}}\big[& + R_p^{M+} n_b^4 \sin^3\theta_0\chi_{zzz} \\ & + R_p^{M+} n_\ell^4 w_b^2 \sin\theta_0\cos^2\phi\chi_{zxx} \\ & + R_p^{M+} n_\ell^4 w_b^2 \sin\theta_0\sin^2\phi\chi_{zxx} \\ & - 2R_p^{M-} n_\ell^2 n_b^2 w_b W_\ell \sin\theta_0\cos^2\phi\chi_{xxz} \\ & - 2R_p^{M-} n_\ell^2 n_b^2 w_b W_\ell \sin\theta_0\sin^2\phi\chi_{xxz} \\ & - R_p^{M-} n_\ell^4 w_b^2 W_\ell \cos^3\phi\chi_{xxx} \\ & + R_p^{M-} n_\ell^4 w_b^2 W_\ell \sin^2\phi\cos\phi\chi_{xxx} \\ & + 2R_p^{M-} n_\ell^4 w_b^2 W_\ell \sin^2\phi\cos\phi\chi_{xxx}\big].\end{aligned}$$

We reduce terms,

$$
\begin{aligned}
\Upsilon_{pP}^{\mathrm{MR},(111)} &= \Gamma_{pP}^{\mathrm{MR}}\big[+ R_p^{M+} n_b^4 \sin^3\theta_0 \chi_{zzz} \\
&\qquad + R_p^{M+} n_\ell^4 w_b^2 \sin\theta_0 (\sin^2\phi + \cos^2\phi)\chi_{zxx} \\
&\qquad - 2R_p^{M-} n_\ell^2 n_b^2 w_b W_\ell \sin\theta_0 (\sin^2\phi + \cos^2\phi)\chi_{xxz} \\
&\qquad + R_p^{M-} n_\ell^4 w_b^2 W_\ell (3\sin^2\phi\cos\phi - \cos^3\phi)\chi_{xxx}\big] \\
\\
&= \Gamma_{pP}^{\mathrm{MR}}\big[R_p^{M+} \sin\theta_0 (n_b^4 \sin^2\theta_0 \chi_{zzz} + n_\ell^4 w_b^2 \chi_{zxx}) \\
&\qquad - R_p^{M-} n_\ell^2 w_b W_\ell (2n_b^2 \sin\theta_0 \chi_{xxz} + n_\ell^2 w_b \chi_{xxx} \cos 3\phi)\big] \\
\\
&= \Gamma_{pP}^{\mathrm{MR}}\, r_{pP}^{\mathrm{MR},(111)},
\end{aligned}
$$

where

$$
\begin{aligned}
r_{pP}^{\mathrm{MR},(111)} =& R_p^{M+} \sin\theta_0 (n_b^4 \sin^2\theta_0 \chi_{zzz} + n_\ell^4 w_b^2 \chi_{zxx}) \\
&- R_p^{M-} n_\ell^2 w_b W_\ell (2n_b^2 \sin\theta_0 \chi_{xxz} + n_\ell^2 w_b \chi_{xxx} \cos 3\phi).
\end{aligned}
\tag{A.18}
$$

If we wish to neglect the effects of the multiple reflections, we follow the exact same procedure but starting with Eq. (A.16),

$$
\begin{aligned}
\Upsilon_{pP}^{(111)} &= \Gamma_{pP}\big[+ n_b^4 N_b^2 \sin^3\theta_0 \chi_{zzz} \\
&\qquad + n_\ell^4 N_b^2 w_b^2 \sin\theta_0 \cos^2\phi \chi_{zxx} \\
&\qquad + n_\ell^4 N_b^2 w_b^2 \sin\theta_0 \sin^2\phi \chi_{zxx} \\
&\qquad - 2n_b^2 n_\ell^2 N_\ell^2 w_b W_b \sin\theta_0 \cos^2\phi \chi_{xxz} \\
&\qquad - 2n_b^2 n_\ell^2 N_\ell^2 w_b W_b \sin\theta_0 \sin^2\phi \chi_{xxz} \\
&\qquad - n_\ell^4 N_\ell^2 w_b^2 W_b \cos^3\phi \chi_{xxx} \\
&\qquad + n_\ell^4 N_\ell^2 w_b^2 W_b \sin^2\phi \cos\phi \chi_{xxx} \\
&\qquad + 2n_\ell^4 N_\ell^2 w_b^2 W_b \sin^2\phi \cos\phi \chi_{xxx}\big],
\end{aligned}
$$

and reduce,

$$
\begin{aligned}
\Upsilon_{pP}^{(111)} &= \Gamma_{pP}\big[+ n_b^4 N_b^2 \sin^3\theta_0 \chi_{zzz} \\
&\qquad + n_\ell^4 N_b^2 w_b^2 \sin\theta_0 (\sin^2\phi + \cos^2\phi)\chi_{zxx} \\
&\qquad - 2n_b^2 n_\ell^2 N_\ell^2 w_b W_b \sin\theta_0 (\sin^2\phi + \cos^2\phi)\phi\chi_{xxz} \\
&\qquad + n_\ell^4 N_\ell^2 w_b^2 W_b (3\sin^2\phi\cos\phi - \cos^3\phi)\chi_{xxx}\big] \\
\\
&= \Gamma_{pP}\big[N_b^2 \sin\theta_0 (n_b^4 \sin^2\theta_0 \chi_{zzz} + n_\ell^4 w_b^2 \chi_{zxx}) \\
&\qquad - n_\ell^2 N_\ell^2 w_b W_b (2n_b^2 \sin\theta_0 \chi_{xxz} + n_\ell^2 w_b \chi_{xxx} \cos 3\phi)\big] \\
\\
&= \Gamma_{pP}\, r_{pP}^{(111)},
\end{aligned}
$$

where

$$r_{pP}^{(111)} = N_b^2 \sin\theta_0 (n_b^4 \sin^2\theta_0 \chi_{zzz} + n_\ell^4 w_b^2 \chi_{zxx}) - n_\ell^2 N_\ell^2 w_b W_b (2 n_b^2 \sin\theta_0 \chi_{xxz} + n_\ell^2 w_b \chi_{xxx} \cos 3\phi). \tag{A.19}$$

A.2.2 For the (110) surface

We take Eqs. (A.14) and (A.12), eliminate the components that do not contribute, and apply the the symmetry relations as follows,

$$\begin{aligned}
\Upsilon_{pP}^{\mathrm{MR},(110)} &= \Gamma_{pP}^{\mathrm{MR}} \Big[R_p^{M+} \sin\theta_0 \big(n_b^4 \sin^2\theta_0 \chi_{zzz} + n_\ell^4 w_b^2 (\sin^2\phi \chi_{zyy} + \cos^2\phi \chi_{zxx}) \big) \\
&\quad - 2 R_p^{M-} n_\ell^2 n_b^2 w_b W_\ell \sin\theta_0 (\sin^2\phi \chi_{yyz} + \cos^2\phi \chi_{xxz}) \Big] \\
&= \Gamma_{pP}^{\mathrm{MR}} \Bigg[R_p^{M+} \sin\theta_0 \Bigg(n_b^4 \sin^2\theta_0 \chi_{zzz} \\
&+ n_\ell^4 w_b^2 \left(\frac{1}{2}(1-\cos 2\phi)\chi_{zyy} + \frac{1}{2}(\cos 2\phi + 1)\chi_{zxx} \right) \Bigg) \\
&- 2 R_p^{M-} n_\ell^2 n_b^2 w_b W_\ell \sin\theta_0 \left(\frac{1}{2}(1-\cos 2\phi)\chi_{yyz} + \frac{1}{2}(\cos 2\phi + 1)\chi_{xxz} \right) \Bigg] \\
&= \Gamma_{pP}^{\mathrm{MR}} \Bigg[R_p^{M+} \sin\theta_0 \Bigg(n_b^4 \sin^2\theta_0 \chi_{zzz} \\
&\quad + n_\ell^4 w_b^2 \left(\frac{\chi_{zyy} + \chi_{zxx}}{2} + \frac{\chi_{zyy} - \chi_{zxx}}{2} \cos 2\phi \right) \Bigg) \\
&\quad - 2 R_p^{M-} n_\ell^2 n_b^2 w_b W_\ell \sin\theta_0 \left(\frac{\chi_{yyz} + \chi_{xxz}}{2} + \frac{\chi_{yyz} - \chi_{xxz}}{2} \cos 2\phi \right) \Bigg] \\
&= \Gamma_{pP}^{\mathrm{MR}} r_{pP}^{\mathrm{MR},(110)},
\end{aligned}$$

where

$$\begin{aligned}
r_{pP}^{\mathrm{MR},(110)} &= R_p^{M+} \sin\theta_0 \Bigg[n_b^4 \sin^2\theta_0 \chi_{zzz} \\
&\quad + n_\ell^4 w_b^2 \left(\frac{\chi_{zyy} + \chi_{zxx}}{2} + \frac{\chi_{zyy} - \chi_{zxx}}{2} \cos 2\phi \right) \Bigg] \\
&\quad - 2 R_p^{M-} n_\ell^2 n_b^2 w_b W_\ell \sin\theta_0 \left(\frac{\chi_{yyz} + \chi_{xxz}}{2} + \frac{\chi_{yyz} - \chi_{xxz}}{2} \cos 2\phi \right).
\end{aligned} \tag{A.20}$$

If we wish to neglect the effects of the multiple reflections, we follow the

exact same procedure but starting with Eq. (A.16),

$$
\begin{aligned}
\Upsilon_{pP}^{(110)} &= \Gamma_{pP}\Bigg[N_b^2\sin\theta_0\Big(n_b^4\sin^2\theta_0\chi_{zzz} + n_\ell^4 w_b^2(\sin^2\phi\chi_{zyy} + \cos^2\phi\chi_{zxx})\Big) \\
&\qquad - 2n_b^2 n_\ell^2 N_\ell^2 w_b W_b \sin\theta_0\big(\sin^2\phi\chi_{yyz} + \cos^2\phi\chi_{xxz}\big)\Bigg] \\
&= \Gamma_{pP}\Bigg[N_b^2\sin\theta_0\Bigg(n_b^4\sin^2\theta_0\chi_{zzz} \\
&\qquad + n_\ell^4 w_b^2\left(\frac{\chi_{zyy}+\chi_{zxx}}{2} + \frac{\chi_{zyy}-\chi_{zxx}}{2}\cos 2\phi\right)\Bigg) \\
&\qquad - 2n_b^2 n_\ell^2 N_\ell^2 w_b W_b \sin\theta_0\left(\frac{\chi_{yyz}+\chi_{xxz}}{2} + \frac{\chi_{yyz}-\chi_{xxz}}{2}\cos 2\phi\right)\Bigg] \\
&= \Gamma_{pP}\, r_{pP}^{(110)},
\end{aligned}
$$

where

$$
\begin{aligned}
r_{pP}^{(110)} = N_b^2\sin\theta_0\Bigg[n_b^4\sin^2\theta_0\chi_{zzz} + n_\ell^4 w_b^2\left(\frac{\chi_{zyy}+\chi_{zxx}}{2} + \frac{\chi_{zyy}-\chi_{zxx}}{2}\cos 2\phi\right)\Bigg] \\
- 2n_b^2 n_\ell^2 N_\ell^2 w_b W_b \sin\theta_0\left(\frac{\chi_{yyz}+\chi_{xxz}}{2} + \frac{\chi_{yyz}-\chi_{xxz}}{2}\cos 2\phi\right).
\end{aligned}
\tag{A.21}
$$

A.2.3 For the (001) surface

We take Eqs. (A.14) and (A.12), eliminate the components that do not contribute, and apply the the symmetry relations as follows,

$$
\begin{aligned}
\Upsilon_{pP}^{\mathrm{MR},(001)} &= \Gamma_{pP}^{\mathrm{MR}}\big[R_p^{M+} n_b^4\sin^3\theta_0\chi_{zzz} \\
&\qquad + R_p^{M+} n_\ell^4 w_b^2\sin\theta_0\cos^2\phi\chi_{zxx} \\
&\qquad + R_p^{M+} n_\ell^4 w_b^2\sin\theta_0\sin^2\phi\chi_{zxx} \\
&\qquad - 2R_p^{M-} n_\ell^2 n_b^2 w_b W_\ell\sin\theta_0\cos^2\phi\chi_{xxz} \\
&\qquad - 2R_p^{M-} n_\ell^2 n_b^2 w_b W_\ell\sin\theta_0\sin^2\phi\chi_{xxz}\big] \\
&= \Gamma_{pP}^{\ell}\big[R_p^{M+}\sin\theta_0(n_b^4\sin^2\theta_0\chi_{zzz} + n_\ell^4 w_b^2\chi_{zxx}) \\
&\qquad - 2R_p^{M-} n_\ell^2 n_b^2 w_b W_\ell\sin\theta_0\chi_{xxz}\big] \\
&= \Gamma_{pP}^{\mathrm{MR}}\, r_{pP}^{\mathrm{MR},(001)},
\end{aligned}
$$

where

$$r_{pP}^{\mathrm{MR},(001)} = R_p^{M+}\sin\theta_0(n_b^4\sin^2\theta_0\chi_{zzz} + n_\ell^4 w_b^2\chi_{zxx}) - 2R_p^{M-}n_\ell^2 n_b^2 w_b W_\ell \sin\theta_0\chi_{xxz}, \tag{A.22}$$

If we wish to neglect the effects of the multiple reflections, we follow the exact same procedure but starting with Eq. (A.16),

$$\begin{aligned}\Upsilon_{pP}^{(001)} &= \Gamma_{pP}\big[N_b^2\sin\theta_0(n_b^4\sin^2\theta_0\chi_{zzz} + n_\ell^4 w_b^2\chi_{zxx}) \\ &\qquad - 2n_b^2 n_\ell^2 N_\ell^2 w_b W_b \sin\theta_0\chi_{xxz}\big] \\ &= \Gamma_{pP}\, r_{pp}^{(001)},\end{aligned}$$

where

$$r_{pP}^{(001)} = N_b^2\sin\theta_0(n_b^4\sin^2\theta_0\chi_{zzz} + n_\ell^4 w_b^2\chi_{zxx}) - 2n_b^2 n_\ell^2 N_\ell^2 w_b W_b \sin\theta_0\chi_{xxz}. \tag{A.23}$$

A.3 $\mathcal{R}_{pS}$

Per Table 1.1, $\mathcal{R}_{pS}$ requires Eqs. (A.6) and (A.9). After some algebra, we obtain that

$$\begin{aligned}\Upsilon_{pS}^{\mathrm{MR}} = \Gamma_{pS}^{\mathrm{MR}}\big[&- n_\ell^4 w_b^2\sin\phi\cos^2\phi\chi_{xxx} - 2n_\ell^4 w_b^2\sin^2\phi\cos\phi\chi_{xxy} \\ &- 2n_\ell^2 n_b^2 w_b\sin\theta_0\sin\phi\cos\phi\chi_{xxz} - n_\ell^4 w_b^2\sin^3\phi\chi_{xyy} \\ &- 2n_\ell^2 n_b^2 w_b\sin\theta_0\sin^2\phi\chi_{xyz} - n_b^4\sin^2\theta_0\sin\phi\chi_{xzz} \\ &+ n_\ell^4 w_b^2\cos^3\phi\chi_{yxx} + 2n_\ell^4 w_b^2\sin\phi\cos^2\phi\chi_{yxy} \\ &+ 2n_\ell^2 n_b^2 w_b\sin\theta_0\cos^2\phi\chi_{yxz} + n_\ell^4 w_b^2\sin^2\phi\cos\phi\chi_{yyy} \\ &+ 2n_\ell^2 n_b^2 w_b\sin\theta_0\sin\phi\cos\phi\chi_{yyz} + n_b^4\sin^2\theta_0\cos\phi\chi_{yzz}\big].\end{aligned} \tag{A.24}$$

We take this opportunity to introduce a quantity that will be repeated throughout this section,

$$\Gamma_{pS}^{\mathrm{MR}} = T_s^{v\ell}R_s^{M+}\left(\frac{t_p^{v\ell}t_p^{\ell b}}{n_\ell^2 n_b}\right)^2 \tag{A.25}$$

If we neglect the multiple reflections, as described in the manuscript, we have that

$$\begin{aligned}\Upsilon_{pS} = \Gamma_{pS}\big[&- n_\ell^4 w_b^2\sin\phi\cos^2\phi\chi_{xxx} - 2n_\ell^4 w_b^2\sin^2\phi\cos\phi\chi_{xxy} \\ &- 2n_\ell^2 n_b^2 w_b\sin\theta_0\sin\phi\cos\phi\chi_{xxz} - n_\ell^4 w_b^2\sin^3\phi\chi_{xyy} \\ &- 2n_\ell^2 n_b^2 w_b\sin\theta_0\sin^2\phi\chi_{xyz} - n_b^4\sin^2\theta_0\sin\phi\chi_{xzz} \\ &+ n_\ell^4 w_b^2\cos^3\phi\chi_{yxx} + 2n_\ell^4 w_b^2\sin\phi\cos^2\phi\chi_{yxy} \\ &+ 2n_\ell^2 n_b^2 w_b\sin\theta_0\cos^2\phi\chi_{yxz} + n_\ell^4 w_b^2\sin^2\phi\cos\phi\chi_{yyy} \\ &+ 2n_\ell^2 n_b^2 w_b\sin\theta_0\sin\phi\cos\phi\chi_{yyz} + n_b^4\sin^2\theta_0\cos\phi\chi_{yzz}\big],\end{aligned} \tag{A.26}$$

and again we introduce a quantity that will be repeated throughout this section,

$$\Gamma_{pS} = T_s^{v\ell} T_s^{\ell b} \left(\frac{t_p^{v\ell} t_p^{\ell b}}{n_\ell^2 n_b} \right)^2 . \tag{A.27}$$

We note that both Eqs. (A.24) and (A.26) are identical save for the different Γ_{pS} terms. Therefore, we can safely derive the equations only once, and then use $\Gamma_{pS}^{\mathrm{MR}}$ when we wish to include multiple reflections, or Γ_{pS} when we do not.

A.3.1 For the (111) surface

We take Eqs. (A.24) and (A.11), eliminate the components that do not contribute, and apply the the symmetry relations as follows,

$$\begin{aligned}
\Upsilon_{pS}^{\mathrm{MR},(111)} = \Gamma_{pS}^{\mathrm{MR}} \big[& + n_\ell^4 w_b^2 \sin^3\phi \chi_{xxx} \\
& - n_\ell^4 w_b^2 \sin\phi \cos^2\phi \chi_{xxx} \\
& - 2 n_\ell^4 w_b^2 \sin\phi \cos^2\phi \chi_{xxx} \\
& - 2 n_\ell^2 n_b^2 w_b \sin\theta_0 \sin\phi \cos\phi \chi_{xxz} \\
& + 2 n_\ell^2 n_b^2 w_b \sin\theta_0 \sin\phi \cos\phi \chi_{xxz} \big].
\end{aligned}$$

We reduce terms,

$$\begin{aligned}
\Upsilon_{pS}^{\mathrm{MR},(111)} &= \Gamma_{pS}^{\mathrm{MR}} \big[n_\ell^4 w_b^2 (\sin^3\phi - 3\sin\phi\cos^2\phi) \chi_{xxx} \big] \\
&= \Gamma_{pS}^{\mathrm{MR}} \big[- n_\ell^4 w_b^2 \chi_{xxx} \sin 3\phi \big] \\
&= \Gamma_{pS}^{\mathrm{MR}} \, r_{pS}^{\mathrm{MR},(111)},
\end{aligned}$$

where

$$r_{pS}^{\mathrm{MR},(111)} = -n_\ell^4 w_b^2 \chi_{xxx} \sin 3\phi. \tag{A.28}$$

As mentioned above,

$$r_{pS}^{(111)} = r_{pS}^{\mathrm{MR},(111)}, \tag{A.29}$$

so if we wish to neglect the effects of the multiple reflections, we simply use Γ_{pS} instead of $\Gamma_{pS}^{\mathrm{MR}}$.

A.3.2 For the (110) surface

We take Eqs. (A.24) and (A.12), eliminate the components that do not contribute, and apply the the symmetry relations as follows,

$$\begin{aligned}\Upsilon_{pS}^{\mathrm{MR},(110)} &= \Gamma_{pS}^{\mathrm{MR}}\big[2n_\ell^2 n_b^2 w_b \sin\theta_0 \sin\phi\cos\phi(\chi_{yyz}-\chi_{xxz})\big]\\ &= \Gamma_{pS}^{\mathrm{MR}}\big[n_\ell^2 n_b^2 w_b \sin\theta_0(\chi_{yyz}-\chi_{xxz})\sin 2\phi\big]\\ &= \Gamma_{pS}^{\mathrm{MR}} r_{pS}^{\mathrm{MR},(110)}.\end{aligned}$$

where

$$r_{pS}^{\mathrm{MR},(110)} = n_\ell^2 n_b^2 w_b \sin\theta_0(\chi_{yyz}-\chi_{xxz})\sin 2\phi. \tag{A.30}$$

As mentioned above,

$$r_{pS}^{(110)} = r_{pS}^{\mathrm{MR},(110)}, \tag{A.31}$$

so if we wish to neglect the effects of the multiple reflections, we simply use Γ_{pS} instead of $\Gamma_{pS}^{\mathrm{MR}}$.

A.3.3 For the (001) surface

We take Eqs. (A.24) and (A.12), eliminate the components that do not contribute, and apply the the symmetry relations as follows,

$$\begin{aligned}\Upsilon_{pS}^{\mathrm{MR},(001)} = \Gamma_{pS}^{\mathrm{MR}}\big[&-2n_\ell^2 n_b^2 w_b \sin\theta_0\sin\phi\cos\phi\chi_{xxz}\\ &+2n_\ell^2 n_b^2 w_b \sin\theta_0\sin\phi\cos\phi\chi_{xxz}\big]=0,\end{aligned}$$

and thus,

$$\Upsilon_{pS}^{\mathrm{MR},(001)} = \Upsilon_{pS}^{(001)} = 0. \tag{A.32}$$

A.4 $\mathcal{R}_{sP}$

Per Table 1.1, $\mathcal{R}_{sP}$ requires Eqs. (A.5) and (A.10). After some algebra, we obtain that

$$\begin{aligned}\Upsilon_{sP}^{\mathrm{MR}} = \Gamma_{sP}^{\mathrm{MR}}\Bigg[&R_p^{M-}W_\ell\big(-\sin^2\phi\cos\phi\chi_{xxx}+2\sin\phi\cos^2\phi\chi_{xxy}-\cos^3\phi\chi_{xyy}\big)\\ &+R_p^{M-}W_\ell\big(-\sin^3\phi\chi_{yxx}+2\sin^2\phi\cos\phi\chi_{yxy}-\sin\phi\cos^2\phi\chi_{yyy}\big)\\ &+R_p^{M+}\sin\theta_0\big(+\sin^2\phi\chi_{zxx}-2\sin\phi\cos\phi\chi_{zxy}+\cos^2\phi\chi_{zyy}\big)\Bigg].\end{aligned} \tag{A.33}$$

We take this opportunity to introduce a quantity that will be repeated throughout this section,

$$\Gamma_{sP}^{\mathrm{MR}} = \frac{T_p^{v\ell}}{N_\ell}\left(t_s^{v\ell}t_s^{\ell b}\right)^2 \tag{A.34}$$

If we neglect the multiple reflections, as described in the manuscript, we have that

$$\begin{aligned}\Upsilon_{sP} = \Gamma_{sP}\Big[&N_\ell^2 W_b\big(-\sin^2\phi\cos\phi\chi_{xxx} + 2\sin\phi\cos^2\phi\chi_{xxy} - \cos^3\phi\chi_{xyy}\big)\\ &+N_\ell^2 W_b\big(-\sin^3\phi\chi_{yxx} + 2\sin^2\phi\cos\phi\chi_{yxy} - \sin\phi\cos^2\phi\chi_{yyy}\big)\\ &+N_b^2\sin\theta_0\big(+\sin^2\phi\chi_{zxx} - 2\sin\phi\cos\phi\chi_{zxy} + \cos^2\phi\chi_{zyy}\big)\Big],\end{aligned} \tag{A.35}$$

and again we introduce a quantity that will be repeated throughout this section,

$$\Gamma_{pS} = \frac{T_p^{v\ell}T_p^{\ell b}}{N_\ell^2 N_b}\left(t_s^{v\ell}t_s^{\ell b}\right)^2. \tag{A.36}$$

A.4.1 For the (111) surface

We take Eqs. (A.33) and (A.11), eliminate the components that do not contribute, and apply the the symmetry relations as follows,

$$\begin{aligned}\Upsilon_{sP}^{\mathrm{MR},(111)} = \Gamma_{sP}^{\mathrm{MR}}\big[&+R_p^{M-}W_\ell\cos^3\phi\chi_{xxx}\\ &-R_p^{M-}W_\ell\sin^2\phi\cos\phi\chi_{xxx}\\ &-2R_p^{M-}W_\ell\sin^2\phi\cos\phi\chi_{xxx}\\ &+R_p^{M+}\sin\theta_0\sin^2\phi\chi_{zxx}\\ &+R_p^{M+}\sin\theta_0\cos^2\phi\chi_{zxx}\big].\end{aligned}$$

We reduce terms,

$$\begin{aligned}\Upsilon_{sP}^{\mathrm{MR},(111)} &= \Gamma_{sP}^{\mathrm{MR}}\big[R_p^{M-}W_\ell(\cos^3\phi - 3\sin^2\phi\cos\phi)\chi_{xxx}\\ &\qquad + R_p^{M+}\sin\theta_0(\sin^2\phi + \cos^2\phi)\chi_{zxx}\big]\\ &= \Gamma_{sP}^{\mathrm{MR}}\big[R_p^{M-}W_\ell\chi_{xxx}\cos 3\phi + R_p^{M+}\sin\theta_0\chi_{zxx}\big]\\ &= \Gamma_{sP}^{\mathrm{MR}}\, r_{sP}^{\mathrm{MR},(111)},\end{aligned}$$

where

$$r_{sP}^{\mathrm{MR},(111)} = R_p^{M+}\sin\theta_0\chi_{zxx} + R_p^{M-}W_\ell\chi_{xxx}\cos 3\phi. \tag{A.37}$$

If we wish to neglect the effects of the multiple reflections, we follow the

exact same procedure but starting with Eq. (A.35),

$$\begin{aligned}\Upsilon_{sP}^{(111)} = \Gamma_{sP}\big[&- N_\ell^2 W_b \sin^2\phi\cos\phi\chi_{xxx} \\ &+ N_\ell^2 W_b \cos^3\phi\chi_{xxx} \\ &- 2N_\ell^2 W_b \sin^2\phi\cos\phi\chi_{yyx} \\ &+ N_b^2 \sin\theta_0 \sin^2\phi\chi_{zxx} \\ &+ N_b^2 \sin\theta_0 \cos^2\phi\chi_{zxx}\big],\end{aligned}$$

and reduce,

$$\begin{aligned}\Upsilon_{sP}^{(111)} &= \Gamma_{sP}\big[N_\ell^2 W_b(\cos^3\phi - 3\sin^2\phi\cos\phi)\chi_{xxx} \\ &\qquad + N_b^2 \sin\theta_0(\sin^2\phi + \cos^2\phi)\chi_{zxx}\big] \\ &= \Gamma_{sP}\big[N_\ell^2 W_b\chi_{xxx}\cos 3\phi + N_b^2\sin\theta_0\chi_{zxx}\big] \\ &= \Gamma_{sP}\, r_{sP}^{(111)},\end{aligned}$$

where

$$r_{sP}^{(111)} = N_b^2 \sin\theta_0\chi_{zxx} + N_\ell^2 W_b\chi_{xxx}\cos 3\phi. \tag{A.38}$$

A.4.2 For the (110) surface

We take Eqs. (A.33) and (A.12), eliminate the components that do not contribute, and apply the the symmetry relations as follows,

$$\begin{aligned}\Upsilon_{sP}^{\mathrm{MR},(110)} &= \Gamma_{sP}^{\mathrm{MR}}\big[R_p^{M+}\sin\theta_0(\sin^2\phi\chi_{zxx} + \cos^2\phi\chi_{zyy})\big] \\ &= \Gamma_{sP}^{\mathrm{MR}}\left[R_p^{M+}\sin\theta_0\left(\frac{1}{2}(1-\cos 2\phi)\chi_{zxx} + \frac{1}{2}(\cos 2\phi + 1)\chi_{zyy}\right)\right] \\ &= \Gamma_{sP}^{\mathrm{MR}}\left[R_p^{M+}\sin\theta_0\left(\frac{\chi_{zyy}+\chi_{zxx}}{2} + \frac{\chi_{zyy}-\chi_{zxx}}{2}\cos 2\phi\right)\right] \\ &= \Gamma_{sP}^{\mathrm{MR}}\, r_{sP}^{\mathrm{MR},(110)},\end{aligned}$$

where

$$r_{sP}^{\mathrm{MR},(110)} = R_p^{M+}\sin\theta_0\left(\frac{\chi_{zxx}+\chi_{zyy}}{2} + \frac{\chi_{zyy}-\chi_{zxx}}{2}\cos 2\phi\right). \tag{A.39}$$

If we wish to neglect the effects of the multiple reflections, we follow the

exact same procedure but starting with Eq. (A.35),

$$\begin{aligned}
\Upsilon_{sP}^{(110)} &= \Gamma_{sP}\left[N_b^2 \sin\theta_0(\sin^2\phi\chi_{zxx} + \cos^2\phi\chi_{zyy})\right] \\
&= \Gamma_{sP}\left[N_b^2 \sin\theta_0 \left(\frac{1}{2}(1-\cos 2\phi)\chi_{zxx} + \frac{1}{2}(\cos 2\phi + 1)\chi_{zyy}\right)\right] \\
&= \Gamma_{sP}\left[N_b^2 \sin\theta_0 \left(\frac{\chi_{zxx}+\chi_{zyy}}{2} + \frac{\chi_{zyy}-\chi_{zxx}}{2}\cos 2\phi\right)\right] \\
&= \Gamma_{sP}\, r_{sP}^{(110)},
\end{aligned}$$

where

$$r_{sP}^{(110)} = N_b^2 \sin\theta_0 \left(\frac{\chi_{zxx}+\chi_{zyy}}{2} + \frac{\chi_{zyy}-\chi_{zxx}}{2}\cos 2\phi\right). \tag{A.40}$$

A.4.3 For the (001) surface

We take Eqs. (A.33) and (A.12), eliminate the components that do not contribute, and apply the the symmetry relations as follows,

$$\begin{aligned}
\Upsilon_{sP}^{\mathrm{MR},(001)} &= \Gamma_{sP}^{\mathrm{MR}}\left[R_p^{M+}\sin\theta_0(\sin^2\phi + \cos^2\phi)\chi_{zxx}\right] \\
&= \Gamma_{sP}^{\mathrm{MR}}\left[R_p^{M+}\sin\theta_0\chi_{zxx}\right] \\
&= \Gamma_{sP}^{\mathrm{MR}}\, r_{sP}^{\mathrm{MR},(001)}.
\end{aligned}$$

where

$$r_{sP}^{\mathrm{MR},(001)} = R_p^{M+}\sin\theta_0\chi_{zxx}. \tag{A.41}$$

If we wish to neglect the effects of the multiple reflections, we follow the exact same procedure but starting with Eq. (A.35),

$$\begin{aligned}
\Upsilon_{sP}^{(001)} &= \Gamma_{sP}\left[N_b^2\sin\theta_0(\sin^2\phi + \cos^2\phi)\chi_{zxx}\right] \\
&= \Gamma_{sP}\left[N_b^2\sin\theta_0\chi_{zxx}\right] \\
&= \Gamma_{sP}\, r_{sP}^{(001)},
\end{aligned}$$

where

$$r_{sP}^{(001)} = N_b^2\sin\theta_0\chi_{zxx}. \tag{A.42}$$

A.5 $\mathcal{R}_{sS}$

Per Table 1.1, $\mathcal{R}_{sS}$ requires Eqs. (A.8) and (A.10). After some algebra, we obtain that

$$\begin{aligned}\Upsilon_{sS}^{\mathrm{MR}} = \Gamma_{sS}^{\mathrm{MR}}\big[&-\sin^3\phi\chi_{xxx} + 2\sin^2\phi\cos\phi\chi_{xxy} - \sin\phi\cos^2\phi\chi_{xyy} \\ &+ \sin^2\phi\cos\phi\chi_{yxx} - 2\sin\phi\cos^2\phi\chi_{yxy} + \cos^3\phi\chi_{yyy}\big].\end{aligned} \tag{A.43}$$

We take this opportunity to introduce a quantity that will be repeated throughout this section,

$$\Gamma_{sS}^{\mathrm{MR}} = T_s^{v\ell} R_s^{M+} \left(t_s^{v\ell} t_s^{\ell b}\right)^2 \tag{A.44}$$

If we neglect the multiple reflections, as described in the manuscript, we have that

$$\begin{aligned}\Upsilon_{sS} = \Gamma_{sS}\big[&-\sin^3\phi\chi_{xxx} + 2\sin^2\phi\cos\phi\chi_{xxy} - \sin\phi\cos^2\phi\chi_{xyy} \\ &+ \sin^2\phi\cos\phi\chi_{yxx} - 2\sin\phi\cos^2\phi\chi_{yxy} + \cos^3\phi\chi_{yyy}\big],\end{aligned} \tag{A.45}$$

and again we introduce a quantity that will be repeated throughout this section,

$$\Gamma_{sS} = T_s^{v\ell} T_s^{\ell b} \left(t_s^{v\ell} t_s^{\ell b}\right)^2. \tag{A.46}$$

We note that both Eqs. (A.43) and (A.45) are identical save for the different Γ_{sS} terms. Therefore, we can safely derive the equations only once, and then use $\Gamma_{sS}^{\mathrm{MR}}$ when we wish to include multiple reflections, or Γ_{sS} when we do not.

A.5.1 For the (111) surface

We take Eqs. (A.43) and (A.11), eliminate the components that do not contribute, and apply the the symmetry relations as follows,

$$\begin{aligned}\Upsilon_{sS}^{\mathrm{MR}} &= \Gamma_{sS}^{\mathrm{MR}}\big[(3\sin\phi\cos^2\phi - \sin^3\phi)\chi_{xxx}\big] \\ &= \Gamma_{sS}^{\mathrm{MR}}\big[\chi_{xxx}\sin 3\phi\big] \\ &= \Gamma_{sS}^{\mathrm{MR}} r_{sS}^{\mathrm{MR},(111)},\end{aligned}$$

where

$$r_{sS}^{\mathrm{MR},(111)} = \chi_{xxx}\sin 3\phi. \tag{A.47}$$

As mentioned above,

$$r_{sS}^{(111)} = r_{sS}^{\mathrm{MR},(111)}, \tag{A.48}$$

so if we wish to neglect the effects of the multiple reflections, we simply use Γ_{sS} instead of $\Gamma_{sS}^{\mathrm{MR}}$.

A.5.2 For the (110) surface

When considering Eqs. (A.43) and (A.12), we see that there are no nonzero components that contribute. Therefore,

$$\Upsilon_{pS}^{\mathrm{MR},(110)} = \Upsilon_{pS}^{(110)} = 0. \tag{A.49}$$

A.5.3 For the (001) surface

When considering Eqs. (A.43) and (A.12), we see that there are no nonzero components that contribute. Therefore,

$$\Upsilon_{sS}^{\mathrm{MR},(001)} = \Upsilon_{sS}^{(001)} = 0. \tag{A.50}$$

Appendix B

Some limiting cases of interest

In this section, we derive the expresions for $\mathcal{R}_{pP}$ for different limiting cases. We evaluate $\mathcal{P}(2\omega)$ and the fundamental fields in different regions. It is worth noting that the first case, the three layer model, can be reduced to any of the other cases by simply considering where we want to evaluate the 1ω and 2ω terms.

B.1 The two layer model

In order to reduce above result to that of Ref. [3] and [5], we now consider that $\mathcal{P}(2\omega)$ is evaluated in the vacuum region, while the fundamental fields are evaluated in the bulk region. To do this, we take the 2ω radiations factors for vacuum by taking $\ell = v$, thus $\epsilon_\ell(2\omega) = 1$, $T_p^{\ell v} = 1$, $T_p^{\ell b} = T_p^{vb}$, and the fundamental field inside medium b by taking $\ell = b$, thus $\epsilon_\ell(\omega) = \epsilon_b(\omega)$, $t_p^{v\ell} = t_p^{vb}$, and $t_p^{\ell b} = 1$. With these choices

$$\mathbf{e}_v^{2\omega} \cdot \boldsymbol{\chi} : \mathbf{e}_b^{\omega}\mathbf{e}_b^{\omega} \equiv \Gamma_{pP}^{vb}\, r_{pP}^{vb},$$

where,

$$\begin{aligned} r_{pP}^{vb} = {} & \epsilon_b(2\omega)\sin\theta_0\Big(\sin^2\theta_0\chi_{zzz} + k_b^2\chi_{zxx}\Big) \\ & - k_b K_b\Big(2\sin\theta_0\chi_{xxz} + k_b\chi_{xxx}\cos(3\phi)\Big), \end{aligned}$$

and

$$\Gamma_{pP}^{vb} = \frac{T_p^{vb}(t_p^{vb})^2}{\epsilon_b(\omega)\sqrt{\epsilon_b(2\omega)}}.$$

B.2 Taking $\mathcal{P}(2\omega)$ and the fundamental fields in the bulk

To consider the 2ω fields in the bulk, we start with Eq. (1.40) but substitute $\ell \to b$, thus

$$\mathbf{H}_b = \hat{\mathbf{s}}\, T_s^{bv} \left(1 + R_s^{bb}\right) \hat{\mathbf{s}} + \hat{\mathbf{P}}_{v+} T_p^{bv} \left(\hat{\mathbf{P}}_{b+} + R_p^{bb} \hat{\mathbf{P}}_{b-}\right).$$

R_p^{bb} and R_s^{bb} are zero, so we are left with

$$\begin{aligned}
\mathbf{H}_b &= \hat{\mathbf{s}}\, T_s^{bv} \hat{\mathbf{s}} + \hat{\mathbf{P}}_{v+} T_p^{bv} \hat{\mathbf{P}}_{b+} \\
&= \frac{K_b}{K_v} \left(\hat{\mathbf{s}}\, T_s^{vb} \hat{\mathbf{s}} + \hat{\mathbf{P}}_{v+} T_p^{vb} \hat{\mathbf{P}}_{b+}\right) \\
&= \frac{K_b}{K_v} \left[\hat{\mathbf{s}}\, T_s^{vb} \hat{\mathbf{s}} + \hat{\mathbf{P}}_{v+} \frac{T_p^{vb}}{\sqrt{\epsilon_b(2\omega)}} (\sin\theta_0 \hat{\mathbf{z}} - K_b \cos\phi \hat{\mathbf{x}} - K_b \sin\phi \hat{\mathbf{y}})\right],
\end{aligned}$$

and we define

$$\mathbf{e}_b^{2\omega} = \frac{K_b}{K_v}\, \hat{\mathbf{e}}^{\mathrm{out}} \cdot \left[\hat{\mathbf{s}}\, T_s^{vb} \hat{\mathbf{s}} + \hat{\mathbf{P}}_{v+} \frac{T_p^{vb}}{\sqrt{\epsilon_b(2\omega)}} (\sin\theta_0 \hat{\mathbf{z}} - K_b \cos\phi \hat{\mathbf{x}} - K_b \sin\phi \hat{\mathbf{y}})\right].$$

For $\mathcal{R}_{pP}$, we require $\hat{\mathbf{e}}^{\mathrm{out}} = \hat{\mathbf{P}}_{v+}$, so we have that

$$\mathbf{e}_b^{2\omega} = \frac{K_b}{K_v} \frac{T_p^{vb}}{\sqrt{\epsilon_b(2\omega)}} (\sin\theta_0 \hat{\mathbf{z}} - K_b \cos\phi \hat{\mathbf{x}} - K_b \sin\phi \hat{\mathbf{y}}).$$

The 1ω fields will still be evaluated inside the bulk, so we have Eq. (1.31)

$$\mathbf{e}_b^{\omega} = \left[\hat{\mathbf{s}} t_s^{vb} \hat{\mathbf{s}} + \frac{t_p^{vb}}{\sqrt{\epsilon_b(\omega)}} \left(\sin\theta_0 \hat{\mathbf{z}} + k_b \cos\phi \hat{\mathbf{x}} + k_b \sin\phi \hat{\mathbf{y}}\right) \hat{\mathbf{p}}_{v-}\right] \cdot \hat{\mathbf{e}}^{\mathrm{in}},$$

and for our particular case of $\hat{\mathbf{e}}^{\mathrm{in}} = \hat{\mathbf{p}}_{v-}$,

$$\mathbf{e}_b^{\omega} = \frac{t_p^{vb}}{\sqrt{\epsilon_b(\omega)}} \left(\sin\theta_0 \hat{\mathbf{z}} + k_b \cos\phi \hat{\mathbf{x}} + k_b \sin\phi \hat{\mathbf{y}}\right),$$

and

$$\begin{aligned}
\mathbf{e}_b^{\omega} \mathbf{e}_b^{\omega} &= \frac{\left(t_p^{vb}\right)^2}{\epsilon_b(\omega)} \left(\sin\theta_0 \hat{\mathbf{z}} + k_b \cos\phi \hat{\mathbf{x}} + k_b \sin\phi \hat{\mathbf{y}}\right)^2 \\
&= \frac{\left(t_p^{vb}\right)^2}{\epsilon_b(\omega)} \big(\sin^2\theta_0 \hat{\mathbf{z}}\hat{\mathbf{z}} + k_b^2 \cos^2\phi \hat{\mathbf{x}}\hat{\mathbf{x}} + k_b^2 \sin^2\phi \hat{\mathbf{y}}\hat{\mathbf{y}} \\
&\qquad + 2k_b \sin\theta_0 \cos\phi \hat{\mathbf{z}}\hat{\mathbf{x}} + 2k_b \sin\theta_0 \sin\phi \hat{\mathbf{z}}\hat{\mathbf{y}} + 2k_b^2 \sin\phi \cos\phi \hat{\mathbf{x}}\hat{\mathbf{y}}\big)
\end{aligned}$$

So lastly, we have that

$$
\begin{aligned}
\mathbf{e}_b^{2\omega}\cdot\boldsymbol{\chi}:\mathbf{e}_b^{\omega}\mathbf{e}_b^{\omega} = \frac{K_b}{K_v}\frac{T_p^{vb}\left(t_p^{vb}\right)^2}{\epsilon_b(\omega)\sqrt{\epsilon_b(2\omega)}}\big(&\sin^3\theta_0\chi_{zzz}\\
&+k_b^2\sin\theta_0\cos^2\phi\chi_{zxx}\\
&+k_b^2\sin\theta_0\sin^2\phi\chi_{zyy}\\
&+2k_b\sin^2\theta_0\cos\phi\chi_{zzx}\\
&+2k_b\sin^2\theta_0\sin\phi\chi_{zzy}\\
&+2k_b^2\sin\theta_0\sin\phi\cos\phi\chi_{zxy}\\
&-K_b\sin^2\theta_0\cos\phi\chi_{xzz}\\
&-k_b^2K_b\cos^3\phi\chi_{xxx}\\
&-k_b^2K_b\sin^2\phi\cos\phi\chi_{xyy}\\
&-2k_bK_b\sin\theta_0\cos^2\phi\chi_{xzx}\\
&-2k_bK_b\sin\theta_0\sin\phi\cos\phi\chi_{xzy}\\
&-2k_b^2K_b\sin\phi\cos^2\phi\chi_{xxy}\\
&-K_b\sin^2\theta_0\sin\phi\chi_{yzz}\\
&-k_b^2K_b\sin\phi\cos^2\phi\chi_{yxx}\\
&-k_b^2K_b\sin^3\phi\chi_{yyy}\\
&-2k_bK_b\sin\theta_0\sin\phi\cos\phi\chi_{yzx}\\
&-2k_bK_b\sin\theta_0\sin^2\phi\chi_{yzy}\\
&-2k_b^2K_b\sin^2\phi\cos\phi\chi_{yxy}\big),
\end{aligned}
$$

and we can eliminate many terms since $\chi_{zzx}=\chi_{zzy}=\chi_{zxy}=\chi_{xzz}=\chi_{xzy}=\chi_{xxy}=\chi_{yzz}=\chi_{yxx}=\chi_{yyy}=\chi_{yzx}=0$, and substituting the equivalent components of $\boldsymbol{\chi}$,

$$
\begin{aligned}
=\frac{K_b}{K_v}\Gamma_{pP}^{b}\big(&\sin^3\theta_0\chi_{zzz}\\
&+k_b^2\sin\theta_0\cos^2\phi\chi_{zxx}\\
&+k_b^2\sin\theta_0\sin^2\phi\chi_{zxx}\\
&-2k_bK_b\sin\theta_0\cos^2\phi\chi_{xxz}\\
&-2k_bK_b\sin\theta_0\sin^2\phi\chi_{xxz}\\
&-k_b^2K_b\cos^3\phi\chi_{xxx}\\
&+k_b^2K_b\sin^2\phi\cos\phi\chi_{xxx}\\
&+2k_b^2K_b\sin^2\phi\cos\phi\chi_{xxx}\big),
\end{aligned}
$$

and reducing,

$$
\begin{aligned}
= \frac{K_b}{K_v}\Gamma^b_{pP}\big(&\sin^3\theta_0\chi_{zzz}\\
&+ k_b^2\sin\theta_0(\sin^2\phi+\cos^2\phi)\chi_{zxx}\\
&- 2k_bK_b\sin\theta_0(\sin^2\phi+\cos^2\phi)\chi_{xxz}\\
&+ k_b^2K_b(3\sin^2\phi\cos\phi-\cos^3\phi)\chi_{xxx}\big)
\end{aligned}
$$

$$
= \frac{K_b}{K_v}\Gamma^b_{pP}\big(\sin^3\theta_0\chi_{zzz} + k_b^2\sin\theta_0\chi_{zxx} - 2k_bK_b\sin\theta_0\chi_{xxz} - k_b^2K_b\chi_{xxx}\cos 3\phi\big),
$$

where,

$$
\Gamma^b_{pP} = \frac{T_p^{vb}\left(t_p^{vb}\right)^2}{\epsilon_b(\omega)\sqrt{\epsilon_b(2\omega)}}.
$$

We find the equivalent expression for $\mathcal{R}$ evaluated inside the bulk as

$$
R(2\omega) = \frac{32\pi^3\omega^2}{c^3K_b^2}\left|\mathbf{e}_b^{2\omega}\cdot\boldsymbol{\chi}:\mathbf{e}_b^{\omega}\mathbf{e}_b^{\omega}\right|^2,
$$

and we can remove the K_b/K_v factor completely and reduce to the standard form of

$$
R(2\omega) = \frac{32\pi^3\omega^2}{c^3\cos^2\theta_0}\left|\mathbf{e}_b^{2\omega}\cdot\boldsymbol{\chi}:\mathbf{e}_b^{\omega}\mathbf{e}_b^{\omega}\right|^2.
$$

B.3 Taking $\mathcal{P}(2\omega)$ and the fundamental fields in the vacuum

To consider the 1ω fields in the vacuum, we start with Eq. (1.29) but substitute $\ell\to v$, thus

$$
\mathbf{E}_v(\omega) = E_0\left[\hat{\mathbf{s}}t_s^{vv}(1+r_s^{vb})\hat{\mathbf{s}} + \hat{\mathbf{p}}_{v-}t_p^{vv}\hat{\mathbf{p}}_{v-} + \hat{\mathbf{p}}_{v+}t_p^{vv}r_p^{vb}\hat{\mathbf{p}}_{v-}\right]\cdot\hat{\mathbf{e}}^{\text{in}},
$$

t_p^{vv} and t_s^{vv} are one, so we are left with

$$
\begin{aligned}
\mathbf{e}_v^{\omega} &= \left[\hat{\mathbf{s}}(1+r_s^{vb})\hat{\mathbf{s}} + \hat{\mathbf{p}}_{v-}\hat{\mathbf{p}}_{v-} + \hat{\mathbf{p}}_{v+}r_p^{vb}\hat{\mathbf{p}}_{v-}\right]\cdot\hat{\mathbf{e}}^{\text{in}}\\
&= \left[\hat{\mathbf{s}}(t_s^{vb})\hat{\mathbf{s}} + (\hat{\mathbf{p}}_{v-}+\hat{\mathbf{p}}_{v+}r_p^{vb})\hat{\mathbf{p}}_{v-}\right]\cdot\hat{\mathbf{e}}^{\text{in}}\\
&= \left[\hat{\mathbf{s}}(t_s^{vb})\hat{\mathbf{s}} + \frac{1}{\sqrt{\epsilon_v(\omega)}}\big(k_v(1-r_p^{vb})\hat{\boldsymbol{\kappa}} + \sin\theta_0(1+r_p^{vb})\hat{\mathbf{z}}\big)\hat{\mathbf{p}}_{v-}\right]\\
&= \left[\hat{\mathbf{s}}(t_s^{vb})\hat{\mathbf{s}} + \left(\frac{k_b}{\sqrt{\epsilon_b(\omega)}}t_p^{vb}\hat{\boldsymbol{\kappa}} + \sqrt{\epsilon_b(\omega)}\sin\theta_0 t_p^{vb}\hat{\mathbf{z}}\right)\hat{\mathbf{p}}_{v-}\right]\cdot\hat{\mathbf{e}}^{\text{in}}\\
&= \left[\hat{\mathbf{s}}(t_s^{vb})\hat{\mathbf{s}} + \frac{t_p^{vb}}{\sqrt{\epsilon_b(\omega)}}\left(k_b\cos\phi\hat{\mathbf{x}} + k_b\sin\phi\hat{\mathbf{y}} + \epsilon_b(\omega)\sin\theta_0\hat{\mathbf{z}}\right)\hat{\mathbf{p}}_{v-}\right]\cdot\hat{\mathbf{e}}^{\text{in}}.
\end{aligned}
$$

For $\mathcal{R}_{pP}$ we require that $\hat{\mathbf{e}}^{\mathrm{in}} = \hat{\mathbf{p}}_{v-}$, so

$$\mathbf{e}_v^{\omega} = \frac{t_p^{vb}}{\sqrt{\epsilon_b(\omega)}}\left(k_b \cos\phi\hat{\mathbf{x}} + k_b \sin\phi\hat{\mathbf{y}} + \epsilon_b(\omega)\sin\theta_0\hat{\mathbf{z}}\right),$$

and

$$\begin{aligned}\mathbf{e}_v^{\omega}\mathbf{e}_v^{\omega} = \left(\frac{t_p^{vb}}{\sqrt{\epsilon_b(\omega)}}\right)^2 \big[&k_b^2\cos^2\phi\hat{\mathbf{x}}\hat{\mathbf{x}}\\ &+ k_b^2\sin^2\phi\hat{\mathbf{y}}\hat{\mathbf{y}}\\ &+ \epsilon_b^2(\omega)\sin^2\theta_0\hat{\mathbf{z}}\hat{\mathbf{z}}\\ &+ 2k_b^2\sin\phi\cos\phi\hat{\mathbf{x}}\hat{\mathbf{y}}\\ &+ 2\epsilon_b(\omega)k_b\sin\theta_0\sin\phi\hat{\mathbf{y}}\hat{\mathbf{z}}\\ &+ 2\epsilon_b(\omega)k_b\sin\theta_0\cos\phi\hat{\mathbf{x}}\hat{\mathbf{z}}\big].\end{aligned}$$

We also require the 2ω fields evaluated in the vacuum, which is Eq. (1.28),

$$\mathbf{e}_v^{2\omega} = \hat{\mathbf{e}}^{\mathrm{out}} \cdot \left[\hat{\mathbf{s}}T_s^{vb}\hat{\mathbf{s}} + \hat{\mathbf{P}}_{v+}\frac{T_p^{vb}}{\sqrt{\epsilon_b(2\omega)}}\left(\epsilon_b(2\omega)\sin\theta_0\hat{\mathbf{z}} - K_b\hat{\boldsymbol{\kappa}}\right)\right], \tag{B.1}$$

and with $\hat{\mathbf{e}}^{\mathrm{out}} = \hat{\mathbf{P}}_{v+}$ we have

$$\mathbf{e}_v^{2\omega} = \frac{T_p^{vb}}{\sqrt{\epsilon_b(2\omega)}}\left(\epsilon_b(2\omega)\sin\theta_0\hat{\mathbf{z}} - K_b\cos\phi\hat{\mathbf{x}} - K_b\sin\phi\hat{\mathbf{y}}\right). \tag{B.2}$$

So lastly, we have that

$$
\begin{aligned}
&\mathbf{e}_v^{2\omega}\cdot\boldsymbol{\chi}:\mathbf{e}_v^{\omega}\mathbf{e}_v^{\omega} = \\
&\quad\frac{T_p^{vb}}{\sqrt{\epsilon_b(2\omega)}}\left(\frac{t_p^{vb}}{\sqrt{\epsilon_b(\omega)}}\right)^2\big[\epsilon_b(2\omega)k_b^2\sin\theta_0\cos^2\phi\chi_{zxx} \\
&\qquad + \epsilon_b(2\omega)k_b^2\sin\theta_0\sin^2\phi\chi_{zyy} \\
&\qquad + \epsilon_b^2(\omega)\epsilon_b(2\omega)\sin^3\theta_0\chi_{zzz} \\
&\qquad + 2\epsilon_b(2\omega)k_b^2\sin\theta_0\sin\phi\cos\phi\chi_{zxy} \\
&\qquad + 2\epsilon_b(\omega)\epsilon_b(2\omega)k_b\sin^2\theta_0\sin\phi\chi_{zyz} \\
&\qquad + 2\epsilon_b(\omega)\epsilon_b(2\omega)k_b\sin^2\theta_0\cos\phi\chi_{zxz} \\
&\qquad - k_b^2K_b\cos^3\phi\chi_{xxx} \\
&\qquad - k_b^2K_b\sin^2\phi\cos\phi\chi_{xyy} \\
&\qquad - \epsilon_b^2(\omega)K_b\sin^2\theta_0\cos\phi\chi_{xzz} \\
&\qquad - 2k_b^2K_b\sin\phi\cos^2\phi\chi_{xxy} \\
&\qquad - 2\epsilon_b(\omega)k_bK_b\sin\theta_0\sin\phi\cos\phi\chi_{xyz} \\
&\qquad - 2\epsilon_b(\omega)k_bK_b\sin\theta_0\cos^2\phi\chi_{xxz} \\
&\qquad - k_b^2K_b\sin\phi\cos^2\phi\chi_{yxx} \\
&\qquad - k_b^2K_b\sin^3\phi\chi yyy \\
&\qquad - \epsilon_b^2(\omega)K_b\sin^2\theta_0\sin\phi\chi_{yzz} \\
&\qquad - 2k_b^2K_b\sin^2\phi\cos\phi\chi_{yxy} \\
&\qquad - 2\epsilon_b(\omega)k_bK_b\sin\theta_0\sin^2\phi\chi_{yyz} \\
&\qquad - 2\epsilon_b(\omega)k_bK_b\sin\theta_0\sin\phi\cos\phi\chi_{yxz}\big],
\end{aligned}
$$

and after eliminating components,

$$
\begin{aligned}
= \Gamma_{pP}^{v}\big[&\epsilon_b^2(\omega)\epsilon_b(2\omega)\sin^3\theta_0\chi_{zzz} \\
&+ \epsilon_b(2\omega)k_b^2\sin\theta_0\cos^2\phi\chi_{zxx} \\
&+ \epsilon_b(2\omega)k_b^2\sin\theta_0\sin^2\phi\chi_{zxx} \\
&- 2\epsilon_b(\omega)k_bK_b\sin\theta_0\cos^2\phi\chi_{xxz} \\
&- 2\epsilon_b(\omega)k_bK_b\sin\theta_0\sin^2\phi\chi_{xxz} \\
&+ 3k_b^2K_b\sin^2\phi\cos\phi\chi_{xxx} \\
&- k_b^2K_b\cos^3\phi\chi_{xxx}\big]
\end{aligned}
$$

$$
\begin{aligned}
= \Gamma_{pP}^{v}\big[&\epsilon_b^2(\omega)\epsilon_b(2\omega)\sin^3\theta_0\chi_{zzz} + \epsilon_b(2\omega)k_b^2\sin\theta_0\chi_{zxx} \\
&- 2\epsilon_b(\omega)k_bK_b\sin\theta_0\chi_{xxz} - k_b^2K_b\chi_{xxx}\cos 3\phi\big],
\end{aligned}
$$

where

$$\Gamma^{v}_{pP} = \frac{T^{vb}_{p}\left(t^{vb}_{p}\right)^{2}}{\epsilon_b(\omega)\sqrt{\epsilon_b(2\omega)}}.$$

B.4 Taking $\mathcal{P}(2\omega)$ in ℓ and the fundamental fields in the bulk

For this scenario with $\hat{\mathbf{e}}^{\rm in} = \hat{\mathbf{p}}_{v-}$ and $\hat{\mathbf{e}}^{\rm out} = \hat{\mathbf{P}}_{v+}$, we obtain from Eq. (**??**),

$$\mathbf{e}^{2\omega}_{\ell} = \frac{T^{v\ell}_{p}T^{\ell b}_{p}}{\epsilon_\ell(2\omega)\sqrt{\epsilon_b(2\omega)}}\left(\epsilon_b(2\omega)\sin\theta_0\hat{\mathbf{z}} - \epsilon_\ell(2\omega)K_b\cos\phi\hat{\mathbf{x}} - \epsilon_\ell(2\omega)K_b\sin\phi\hat{\mathbf{y}}\right),$$

and Eq. (1.31),

$$\begin{aligned}\mathbf{e}^{\omega}_{b}\mathbf{e}^{\omega}_{b} = \frac{\left(t^{vb}_{p}\right)^{2}}{\epsilon_b(\omega)}\big(&\sin^2\theta_0\hat{\mathbf{z}}\hat{\mathbf{z}} + k_b^2\cos^2\phi\hat{\mathbf{x}}\hat{\mathbf{x}} + k_b^2\sin^2\phi\hat{\mathbf{y}}\hat{\mathbf{y}}\\ &+ 2k_b\sin\theta_0\cos\phi\hat{\mathbf{z}}\hat{\mathbf{x}} + 2k_b\sin\theta_0\sin\phi\hat{\mathbf{z}}\hat{\mathbf{y}} + 2k_b^2\sin\phi\cos\phi\hat{\mathbf{x}}\hat{\mathbf{y}}\big).\end{aligned}$$

Thus,

$$\begin{aligned}\mathbf{e}^{2\omega}_{\ell}\cdot\boldsymbol{\chi}:\mathbf{e}^{\omega}_{b}\mathbf{e}^{\omega}_{b} = \frac{T^{v\ell}_{p}T^{\ell b}_{p}\left(t^{vb}_{p}\right)^{2}}{\epsilon_\ell(2\omega)\epsilon_b(\omega)\sqrt{\epsilon_b(2\omega)}}\Bigg[&+\epsilon_b(2\omega)\sin^3\theta_0\chi_{zzz}\\
&+\epsilon_b(2\omega)k_b^2\sin\theta_0\cos^2\phi\chi_{zxx}\\
&+\epsilon_b(2\omega)k_b^2\sin\theta_0\sin^2\phi\chi_{zyy}\\
&+2\epsilon_b(2\omega)k_b\sin^2\theta_0\cos\phi\chi_{zzx}\\
&+2\epsilon_b(2\omega)k_b\sin^2\theta_0\sin\phi\chi_{zzy}\\
&+2\epsilon_b(2\omega)k_b^2\sin\theta_0\sin\phi\cos\phi\chi_{zxy}\\
&-\epsilon_\ell(2\omega)\sin^2\theta_0K_b\cos\phi\chi_{xzz}\\
&-\epsilon_\ell(2\omega)k_b^2K_b\cos^3\phi\chi_{xxx}\\
&-\epsilon_\ell(2\omega)k_b^2K_b\sin^2\phi\cos\phi\chi_{xyy}\\
&-2\epsilon_\ell(2\omega)k_bK_b\sin\theta_0\cos^2\phi\chi_{xzx}\\
&-2\epsilon_\ell(2\omega)k_bK_b\sin\theta_0\sin\phi\cos\phi\chi_{xzy}\\
&-2\epsilon_\ell(2\omega)k_b^2K_b\sin\phi\cos^2\phi\chi_{xxy}\\
&-\epsilon_\ell(2\omega)K_b\sin^2\theta_0\sin\phi\chi_{yzz}\\
&-\epsilon_\ell(2\omega)k_b^2K_b\cos^2\phi\sin\phi\chi_{yxx}\\
&-\epsilon_\ell(2\omega)k_b^2K_b\sin^3\phi\chi_{yyy}\\
&-2\epsilon_\ell(2\omega)k_bK_b\sin\theta_0\cos\phi\sin\phi\chi_{yzx}\\
&-2\epsilon_\ell(2\omega)k_bK_b\sin\theta_0\sin^2\phi\chi_{yzy}\\
&-2\epsilon_\ell(2\omega)k_b^2K_b\sin^2\phi\cos\phi\chi_{yxy}\Bigg].\end{aligned}$$

We eliminate and replace components,

$$
\begin{aligned}
\mathbf{e}_\ell^{2\omega}\cdot\boldsymbol{\chi}:\mathbf{e}_b^\omega\mathbf{e}_b^\omega = \Gamma_{pP}^{\ell b}\Big[&+\epsilon_b(2\omega)\sin^3\theta_0\chi_{zzz} \\
&+\epsilon_b(2\omega)k_b^2\sin\theta_0\cos^2\phi\chi_{zxx} \\
&+\epsilon_b(2\omega)k_b^2\sin\theta_0\sin^2\phi\chi_{zxx} \\
&-2\epsilon_\ell(2\omega)k_bK_b\sin\theta_0\cos^2\phi\chi_{xxz} \\
&-2\epsilon_\ell(2\omega)k_bK_b\sin\theta_0\sin^2\phi\chi_{xxz} \\
&-\epsilon_\ell(2\omega)k_b^2K_b\cos^3\phi\chi_{xxx} \\
&+\epsilon_\ell(2\omega)k_b^2K_b\sin^2\phi\cos\phi\chi_{xxx} \\
&+2\epsilon_\ell(2\omega)k_b^2K_b\sin^2\phi\cos\phi\chi_{xxx}\Big],
\end{aligned}
$$

so lastly

$$
\begin{aligned}
\mathbf{e}_\ell^{2\omega}\cdot\boldsymbol{\chi}:\mathbf{e}_b^\omega\mathbf{e}_b^\omega = \Gamma_{pP}^{\ell b}\Big[&\epsilon_b(2\omega)\sin^3\theta_0\chi_{zzz}+\epsilon_b(2\omega)k_b^2\sin\theta_0\chi_{zxx} \\
&-2\epsilon_\ell(2\omega)k_bK_b\sin\theta_0\chi_{xxz}-\epsilon_\ell(2\omega)k_b^2K_b\chi_{xxx}\cos 3\phi\Big],
\end{aligned}
$$

where

$$
\Gamma_{pP}^{\ell b} = \frac{T_p^{v\ell}T_p^{\ell b}\left(t_p^{vb}\right)^2}{\epsilon_\ell(2\omega)\epsilon_b(\omega)\sqrt{\epsilon_b(2\omega)}}.
$$

Appendix C

The two layer model for SHG radiation from Sipe, Moss, and van Driel

In this treatment we follow the work of Ref. [5]. They define the following for all polarizations;

$$
\begin{aligned}
f_s &= \frac{\kappa}{n\tilde{\omega}} = \frac{\kappa}{\sqrt{\epsilon(\omega)}\tilde{\omega}}, \\
f_c &= \frac{w}{n\tilde{\omega}} = \frac{w}{\sqrt{\epsilon(\omega)}\tilde{\omega}}, \\
f_s^2 &+ f_c^2 = 1,
\end{aligned}
\tag{C.1}
$$

where

$$
\kappa = \tilde{\omega}\sin\theta,
$$

$$
w_0 = \sqrt{\tilde{\omega} - \kappa^2} = \tilde{\omega}\cos\theta, \tag{C.2}
$$

$$
w = \sqrt{\tilde{\omega}\epsilon(\omega) - \kappa^2} = \tilde{\omega}k_z(\omega). \tag{C.3}
$$

From this point on, all capital letters and symbols indicate evaluation at 2ω. Common to all three polarization cases studied here, we require the nonzero components for the (111) face for crystals with C_{3v} symmetry,

$$
\begin{aligned}
\delta_{11} &= \chi^{xxx} = -\chi^{xyy} = -\chi^{yyx}, \\
\delta_{15} &= \chi^{xxz} = \chi^{yyz}, \\
\delta_{31} &= \chi^{zxx} = \chi^{zyy}, \\
\delta_{33} &= \chi^{zzz}.
\end{aligned}
\tag{C.4}
$$

Lastly, the remaining quantities that will be needed for all three cases are

$$
\begin{aligned}
A_p &= \frac{4\pi\tilde{\Omega}\sqrt{\epsilon(2\omega)}}{W_0\epsilon(2\omega)+W},\\
A_s &= \frac{4\pi\tilde{\Omega}}{W_0+W}.
\end{aligned}
\tag{C.5}
$$

C.1 $\mathcal{R}_{pP}$

For the (111) face ($m=3$), we have

$$
\frac{E^{(2\omega)}(\|,\|)}{E_p^2 A_p} = a_{\|,\|} + c^{(3)}_{\|,\|}\cos 3\phi. \tag{C.6}
$$

We extract these coefficients from Table V, noting that $\Gamma=\gamma=0$ as we are only interested in the surface contribution,

$$
\begin{aligned}
a_{\|,\|} &= i\tilde{\Omega}F_s\epsilon(2\omega)\delta_{31} + i\tilde{\Omega}\epsilon(2\omega)F_s f_s^2(\delta_{33}-\delta_{31}) - 2i\tilde{\Omega}f_s f_c F_c\delta_{15},\\
c^{(3)}_{\|,\|} &= -i\tilde{\Omega}F_c f_c^2\delta_{11}.
\end{aligned}
$$

We substitute these in Eq. (C.6),

$$
\begin{aligned}
\frac{E^{(2\omega)}(\|,\|)}{E_p^2 A_p} = {} & i\tilde{\Omega}F_s\epsilon(2\omega)\delta_{31} + i\tilde{\Omega}\epsilon(2\omega)F_s f_s^2(\delta_{33}-\delta_{31})\\
& - 2i\tilde{\Omega}f_s f_c F_c\delta_{15} - i\tilde{\Omega}F_c f_c^2\delta_{11}\cos 3\phi
\end{aligned}
$$

and reduce (omitting the $(\|,\|)$ notation),

$$
\begin{aligned}
\frac{E^{(2\omega)}}{E_p^2} &= A_p i\tilde{\Omega}\left[F_s\epsilon(2\omega)(\delta_{31}+f_s^2(\delta_{33}-\delta_{31})) - f_cF_c(2f_s\delta_{15}+f_c\delta_{11}\cos 3\phi)\right]\\
&= A_p i\tilde{\Omega}\left[F_s\epsilon(2\omega)(f_s^2\delta_{33}+(1-f_s^2)\delta_{31}) - f_cF_c(2f_s\delta_{15}+f_c\delta_{11}\cos 3\phi)\right]\\
&= A_p i\tilde{\Omega}\left[F_s\epsilon(2\omega)(f_s^2\delta_{33}+f_c^2\delta_{31}) - f_cF_c(2f_s\delta_{15}+f_c\delta_{11}\cos 3\phi)\right].
\end{aligned}
$$

As every term has an $f_i^2F_i$, we can factor out the common

$$
\frac{1}{\tilde{\omega}^2\tilde{\Omega}\epsilon(\omega)\sqrt{\epsilon(2\omega)}}
$$

factor after substituting the appropriate terms from Eq. (C.1),

$$
\begin{aligned}
\frac{E^{(2\omega)}}{E_p^2} &= \frac{A_p i}{\epsilon(\omega)\sqrt{\epsilon(2\omega)}\tilde{\omega}^2}\left[K\epsilon(2\omega)(\kappa^2\delta_{33} + w^2\delta_{31}) - wW(2\kappa\delta_{15} + w\delta_{11}\cos 3\phi)\right] \\
&= \frac{A_p i\tilde{\Omega}}{\epsilon(\omega)\sqrt{\epsilon(2\omega)}}\big[\sin\theta\epsilon(2\omega)(\sin^2\theta\delta_{33} + k_z^2(\omega)\delta_{31}) \\
&\qquad - k_z(\omega)k_z(2\omega)(2\sin\theta\delta_{15} + k_z(\omega)\delta_{11}\cos 3\phi)\big] \\
&= \frac{A_p i\tilde{\Omega}}{\epsilon(\omega)\sqrt{\epsilon(2\omega)}}\big[\sin\theta\epsilon(2\omega)(\sin^2\theta\chi^{zzz} + k_z^2(\omega)\chi^{zxx}) \\
&\qquad - k_z(\omega)k_z(2\omega)(2\sin\theta\chi^{xxz} + k_z(\omega)\chi^{xxx}\cos 3\phi)\big].
\end{aligned}
$$

We substitute Eq. (C.5) to complete the expression,

$$
\begin{aligned}
\frac{E^{(2\omega)}}{E_p^2} &= \frac{4i\pi\tilde{\Omega}^2}{\epsilon(\omega)(W_0\epsilon(2\omega) + W)}[\cdots] \\
&= \frac{4i\pi\tilde{\Omega}}{\epsilon(\omega)(\epsilon(2\omega)\cos\theta + k_z(2\omega))}[\cdots] \\
&= \frac{4i\pi\tilde{\omega}}{\cos\theta}\frac{1}{\epsilon(\omega)}\frac{2\cos\theta}{\epsilon(2\omega)\cos\theta + k_z(2\omega)}[\cdots].
\end{aligned}
$$

However, our interest lies in $\mathcal{R}_{pP}$ which is calculated as

$$
\mathcal{R}_{pP} = \frac{I_p(2\omega)}{I_p^2(\omega)} = \frac{2\pi}{c}\left|\frac{E^{(2\omega)}(\|,\|)}{E_p^2}\right|^2,
$$

and we can finally complete the expression,

$$
\begin{aligned}
\mathcal{R}_{pP} &= \frac{2\pi}{c}\left|\frac{4i\pi\tilde{\omega}}{\cos\theta}\frac{1}{\epsilon(\omega)}\frac{2\cos\theta}{\epsilon(2\omega)\cos\theta + k_z(2\omega)}r_{pP}\right|^2 \\
&= \frac{32\pi^3\tilde{\omega}^2}{c\cos^2\theta}\left|t_p(\omega)T_p(2\omega)r_{pP}\right|^2 \\
&= \frac{32\pi^3\omega^2}{c^3\cos^2\theta}\left|t_p(\omega)T_p(2\omega)r_{pP}\right|^2,
\end{aligned}
\tag{C.7}
$$

where

$$
\begin{aligned}
t_p(\omega) &= \frac{1}{\epsilon(\omega)}, \\
T_p(2\omega) &= \frac{2\cos\theta}{\epsilon(2\omega)\cos\theta + k_z(2\omega)}, \\
r_{pP} &= \sin\theta\epsilon(2\omega)(\sin^2\theta\chi^{zzz} + k_z^2(\omega)\chi^{zxx}) \\
&\quad - k_z(\omega)k_z(2\omega)(2\sin\theta\chi^{xxz} + k_z(\omega)\chi^{xxx}\cos 3\phi).
\end{aligned}
$$

C.2 $\mathcal{R}_{pS}$

We follow the same procedure as above. For the (111) face ($m = 3$),

$$\frac{E^{(2\omega)}(\|, \perp)}{E_p^2 A_s} = b_{\|,\perp}^{(3)} \sin 3\phi, \tag{C.8}$$

and we extract the relevant coefficient from Table V with $\Gamma = \gamma = 0$,

$$b_{\|,\perp}^{(3)} = i\tilde{\Omega} f_c^2 \delta_{11}.$$

Substituting this coeffecient and Eq. (C.5) into Eq. (C.8),

$$\begin{aligned}
\frac{E^{(2\omega)}(\|, \perp)}{E_p^2} &= A_s i\tilde{\Omega} f_c^2 \delta_{11} \sin 3\phi \\
&= \frac{A_s i\tilde{\Omega}}{\tilde{\omega}^2 \epsilon(\omega)} w^2 \delta_{11} \sin 3\phi \\
&= \frac{A_s i\tilde{\Omega}}{\epsilon(\omega)} k_z^2(\omega) \delta_{11} \sin 3\phi \\
&= \frac{A_s i\tilde{\Omega}}{\epsilon(\omega)} k_z^2(\omega) \chi^{xxx} \sin 3\phi \\
&= \frac{4i\pi\tilde{\Omega}^2}{W_0 + W} \frac{1}{\epsilon(\omega)} k_z^2(\omega) \chi^{xxx} \sin 3\phi \\
&= 4i\pi\tilde{\Omega} \frac{1}{\epsilon(\omega)} \frac{1}{\cos\theta + k_z(2\omega)} k_z^2(\omega) \chi^{xxx} \sin 3\phi \\
&= \frac{4i\pi\omega}{c\cos\theta} \frac{1}{\epsilon(\omega)} \frac{2\cos\theta}{\cos\theta + k_z(2\omega)} k_z^2(\omega) \chi^{xxx} \sin 3\phi
\end{aligned}$$

As before, we must calculate

$$\mathcal{R}_{pS} = \frac{2\pi}{c} \left| \frac{E^{(2\omega)}(\|, \perp)}{E_s^2} \right|^2,$$

to obtain the final expression,

$$\begin{aligned}
\mathcal{R}_{pS} &= \frac{2\pi}{c} \left| \frac{4i\pi\omega}{c\cos\theta} \frac{1}{\epsilon(\omega)} \frac{2\cos\theta}{\cos\theta + k_z(2\omega)} k_z^2(\omega) \chi^{xxx} \sin 3\phi \right|^2 \\
&= \frac{32\pi^3\omega^2}{c^3\cos^2\theta} \left| \frac{1}{\epsilon(\omega)} \frac{2\cos\theta}{\cos\theta + k_z(2\omega)} k_z^2(\omega) \chi^{xxx} \sin 3\phi \right|^2 \\
&= \frac{32\pi^3\omega^2}{c^3\cos^2\theta} \left| t_p(\omega) T_s(2\omega) k_z^2(\omega) r_{pS} \right|^2,
\end{aligned} \tag{C.9}$$

where

$$\begin{aligned}
t_p(\omega) &= \frac{1}{\epsilon(\omega)},\\
T_s(2\omega) &= \frac{2\cos\theta}{\cos\theta + k_z(2\omega)},\\
r_{pS} &= k_z^2(\omega)\chi^{xxx}\sin 3\phi.
\end{aligned}$$

C.3 $\mathcal{R}_{sP}$

We follow the same procedure as above for the final polarization case. For the (111) face ($m = 3$),

$$\frac{E^{(2\omega)}(\perp,\|)}{E_s^2 A_p} = a_{\perp,\|} + c_{\perp,\|}^{(3)}\cos 3\phi, \tag{C.10}$$

and we extract the relevant coefficients from Table V with $\Gamma = \gamma = 0$,

$$\begin{aligned}
a_{\perp,\|} &= i\tilde{\Omega}F_s\epsilon(2\omega)\delta_{31},\\
c_{\perp,\|}^{(3)} &= i\tilde{\Omega}F_c\delta_{11}.
\end{aligned}$$

Substituting this coeffecient and Eq. (C.5) into Eq. (C.10),

$$\begin{aligned}
\frac{E^{(2\omega)}(\perp,\|)}{E_s^2} &= A_p(i\tilde{\Omega}F_s\epsilon(2\omega)\delta_{31} + i\tilde{\Omega}F_c\delta_{11}\cos 3\phi)\\
&= A_p i\tilde{\Omega}(F_s\epsilon(2\omega)\delta_{31} + F_c\delta_{11}\cos 3\phi)\\
&= \frac{A_p i\tilde{\Omega}}{\sqrt{\epsilon(2\omega)}}(\sin\theta\epsilon(2\omega)\delta_{31} + k_z(2\omega)\delta_{11}\cos 3\phi)\\
&= \frac{A_p i\tilde{\Omega}}{\sqrt{\epsilon(2\omega)}}(\sin\theta\epsilon(2\omega)\chi^{zxx} + k_z(2\omega)\chi^{xxx}\cos 3\phi)\\
&= \frac{4i\pi\tilde{\Omega}^2}{W_0\epsilon(2\omega) + W}(\sin\theta\epsilon(2\omega)\chi^{zxx} + k_z(2\omega)\chi^{xxx}\cos 3\phi)\\
&= \frac{4i\pi\tilde{\Omega}}{\epsilon(2\omega)\cos\theta + k_z(2\omega)}(\sin\theta\epsilon(2\omega)\chi^{zxx} + k_z(2\omega)\chi^{xxx}\cos 3\phi)\\
&= \frac{4i\pi\omega}{c\cos\theta}\frac{2\cos\theta}{\epsilon(2\omega)\cos\theta + k_z(2\omega)}(\sin\theta\epsilon(2\omega)\chi^{zxx} + k_z(2\omega)\chi^{xxx}\cos 3\phi).
\end{aligned}$$

iF	$t_i(\omega)$	$T_F(2\omega)$	r_{iF}
pP	$\frac{1}{\epsilon(\omega)}$	$\frac{2\cos\theta}{\epsilon(2\omega)\cos\theta + k_z(2\omega)}$	$\sin\theta\epsilon(2\omega)(\sin^2\theta\chi^{zzz} + k_z^2(\omega)\chi^{zxx})$ $-k_z(\omega)k_z(2\omega)(2\sin\theta\chi^{xxz}+k_z(\omega)\chi^{xxx}\cos 3\phi)$
pS	$\frac{1}{\epsilon(\omega)}$	$\frac{2\cos\theta}{\cos\theta + k_z(2\omega)}$	$k_z^2(\omega)\chi^{xxx}\sin 3\phi$
sP	1	$\frac{2\cos\theta}{\epsilon(2\omega)\cos\theta + k_z(2\omega)}$	$\sin\theta\epsilon(2\omega)\chi^{zxx} + k_z(2\omega)\chi^{xxx}\cos 3\phi$

Table C.1: The necessary factors for Eq. (C.12) for each polarization case.

And we finally obtain $\mathcal{R}_{sP}$,

$$\begin{aligned}
\mathcal{R}_{sP} &= \frac{2\pi}{c}\left|\frac{E^{(2\omega)}(\perp,\|)}{E_s^2}\right|^2 \\
&= \frac{2\pi}{c}\left|\frac{4i\pi\omega}{c\cos\theta}\frac{2\cos\theta}{\epsilon(2\omega)\cos\theta + k_z(2\omega)}(\sin\theta\epsilon(2\omega)\chi^{zxx} + k_z(2\omega)\chi^{xxx}\cos 3\phi)\right|^2 \\
&= \frac{32\pi^3\omega^2}{c^3\cos^2\theta}\left|\frac{2\cos\theta}{\epsilon(2\omega)\cos\theta + k_z(2\omega)}(\sin\theta\epsilon(2\omega)\chi^{zxx} + k_z(2\omega)\chi^{xxx}\cos 3\phi)\right|^2 \\
&= \frac{32\pi^3\omega^2}{c^3\cos^2\theta}\left|t_s(\omega)T_p(2\omega)r_{sP}\right|^2, \qquad \text{(C.11)}
\end{aligned}$$

where

$$\begin{aligned}
t_s(\omega) &= 1, \\
T_p(2\omega) &= \frac{2\cos\theta}{\epsilon(2\omega)\cos\theta + k_z(2\omega)}, \\
r_{sP} &= \sin\theta\epsilon(2\omega)\chi^{zxx} + k_z(2\omega)\chi^{xxx}\cos 3\phi.
\end{aligned}$$

C.4 Summary

We unify the final expressions for the SHG yield, Eqs. (C.7), (C.9), and (C.11), as

$$\mathcal{R}_iF = \frac{32\pi^3\omega^2}{c^3\cos^2\theta}\left|t_i(\omega)T_F(2\omega)r_{iF}\right|^2. \qquad \text{(C.12)}$$

The necessary factors are summarized in Table C.1.

Bibliography

[1] Robert W. Boyd. *Nonlinear Optics*. AP, New York, 2007.

[2] Michele Cini. Simple model of electric-dipole second-harmonic generation from interfaces. *Physical Review B*, 43(6):4792–4802, February 1991.

[3] V. Mizrahi and J. E. Sipe. Phenomenological treatment of surface second-harmonic generation. *J. Opt. Soc. Am. B*, 5(3):660–667, 1988.

[4] J. E. Sipe. New Green-function formalism for surface optics. *Journal of the Optical Society of America B*, 4(4):481–489, 1987.

[5] J. E. Sipe, D. J. Moss, and H. M. van Driel. Phenomenological theory of optical second- and third-harmonic generation from cubic centrosymmetric crystals. *Phys. Rev. B*, 35(3):1129–1141, January 1987.