A treatise on phenomenological models of surface second-harmonic generation from crystalline surfaces

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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

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

with the intensity

$$I(\omega) = \frac{c}{2\pi} |E(\omega)|^2,$$

There are several ways to calculate R, one of which is the procedure followed by Cini [1]. 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 [2], since the derivation of the three-layer-model is straightforward. Within our level of approximation this is the best model that we can use. In this scheme, we assume that the SH conversion takes place in a thin layer, just below the surface, that is characterized by a surface dielectric function $\epsilon_{\ell}(\omega)$. This layer is below vacuum and sits on top of the bulk characterized by $\epsilon_{b}(\omega)$ (see Fig. 1.1). The nonlinear polarization immersed in the thin layer, will radiate an electric field directly into vacuum and also into the bulk. This bulk directed field, will be reflected back into vacuum. Thus, the total field radiated into vacuum will be the sum of these two contributions (see Fig. 1.1). We decompose the field into s and p polarizations, then the electric field radiated by a polarization sheet.

$$\mathcal{P}_i(2\omega) = \chi_{ijk} E_j(\omega) E_k(\omega), \tag{1.1}$$

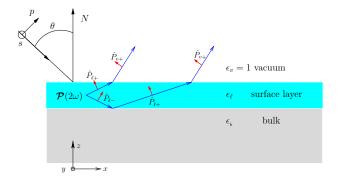


Figure 1.1: Sketch of the three layer model for SHG. Vacuum is on top with $\epsilon = 1$, the layer with nonlinear polarization $\mathbf{P}(2\omega)$ is characterized with $\epsilon_{\ell}(\omega)$ and the bulk with $\epsilon_{b}(\omega)$. In the dipolar approximation the bulk does not radiate SHG. The thin arrows are along the direction of propagation, and the unit vectors for p-polarization are denoted with thick arrows (capital letters denote SH components). The unit vector for s-polarization points along -y (out of the page).

is given by [2],

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

where $\hat{\mathbf{s}}$ and $\hat{\mathbf{p}}_{\pm}$ are the unitary vectors for s and p polarization, respectively, and the \pm refers to upward (+) or downward (-) direction of propagation. Also, $\tilde{\omega} = \omega/c$ and $w_i = \tilde{\omega} k_i$, with

$$k_i(\omega) = \sqrt{\epsilon_i(\omega) - \sin^2 \theta_i},$$

where $i = v, \ell, b$, with

$$\hat{\mathbf{p}}_{i\pm} = \frac{\mp k_i(\omega)\hat{\mathbf{x}} + \sin\theta_i\hat{\mathbf{z}}}{\sqrt{\epsilon_i(\omega)}}$$

In the above equations z is the direction perpendicular to the surface that points towards the vacuum, x is parallel to the surface, and θ is the angle of incidence, where the plane of incidence is chosen as the xz plane (see Fig. 1.1), thus $\hat{\mathbf{s}} = -\hat{\mathbf{y}}$. The function $k_i(\omega)$ is the projection of the wave vector perpendicular to the surface. As we see from Fig. 1.1, the SH field is refracted at the layer-vacuum interface (ℓv), and reflected from the layer-bulk (ℓb) interface, thus we can define the transmission, \mathbf{T} , and reflection,

1.1. THREE LAYER MODEL FOR SHG YIELD, WITHOUT MULTIPLE REFLECTIONS7

R, tensors as,

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

and

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

where variables in capital letters are evaluated at the harmonic frequency 2ω . Notice that since $\hat{\mathbf{s}}$ is independent of ω , then $\hat{\mathbf{S}} = \hat{\mathbf{s}}$. The Fresnel factors, T_i , R_i , for i = s, p polarization, are evaluated at the appropriate interface ℓv or ℓb , and will be given below. The extra subscript in $\hat{\mathbf{P}}$ denotes the corresponding dielectric function to be used in its evaluation, i.e. $\epsilon_v = 1$ for vacuum (v), ϵ_ℓ for the layer (ℓ) , and ϵ_b for the bulk (b). Therefore, the total radiated field at 2ω is

$$\mathbf{E}(2\omega) = E_s(2\omega) \left(\mathbf{T}^{\ell v} + \mathbf{T}^{\ell v} \cdot \mathbf{R}^{\ell b} \right) \cdot \hat{\mathbf{s}}$$

+ $E_{p+}(2\omega) \mathbf{T}^{\ell v} \cdot \hat{\mathbf{P}}_{\ell+} + E_{p-}(2\omega) \mathbf{T}^{\ell v} \cdot \mathbf{R}^{\ell b} \cdot \hat{\mathbf{P}}_{\ell-}.$

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

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

where,

$$\mathbf{H}_{\ell} = \hat{\mathbf{s}} T_s^{\ell v} \left(1 + R_s^{\ell b} \right) \hat{\mathbf{s}} + \hat{\mathbf{P}}_{v+} T_p^{\ell v} \left(\hat{\mathbf{P}}_{\ell+} + R_p^{\ell b} \hat{\mathbf{P}}_{\ell-} \right). \tag{1.2}$$

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

$$\hat{\mathbf{P}}_{\ell+} + R_p^{\ell b} \hat{\mathbf{P}}_{\ell-} = \frac{\sin \theta_{\text{in}} \hat{\mathbf{z}} - K_{\ell} \hat{\mathbf{x}}}{\sqrt{\epsilon_{\ell}(2\omega)}} + R_p^{\ell b} \frac{\sin \theta_{\text{in}} \hat{\mathbf{z}} + K_{\ell} \hat{\mathbf{x}}}{\sqrt{\epsilon_{\ell}(2\omega)}}$$

$$= \frac{1}{\sqrt{\epsilon_{\ell}(2\omega)}} \left(\sin \theta_{\text{in}} (1 + R_p^{\ell b}) \hat{\mathbf{z}} - K_{\ell} (1 - R_p^{\ell b}) \hat{\mathbf{x}} \right)$$

$$= \frac{T_p^{\ell b}}{\epsilon_{\ell}(2\omega) \sqrt{\epsilon_b(2\omega)}} \left(\epsilon_b(2\omega) \sin \theta_{\text{in}} \hat{\mathbf{z}} - \epsilon_{\ell}(2\omega) K_b \hat{\mathbf{x}} \right),$$

where using

$$1 + R_s^{\ell b} = T_s^{\ell b}$$

$$1 + R_p^{\ell b} = \sqrt{\frac{\epsilon_b(2\omega)}{\epsilon_\ell(2\omega)}} T_p^{\ell b}$$

$$1 - R_p^{\ell b} = \sqrt{\frac{\epsilon_\ell(2\omega)}{\epsilon_b(2\omega)}} \frac{K_b}{K_\ell} T_p^{\ell b}$$

$$T_p^{\ell v} = \frac{K_\ell}{K_v} T_p^{v\ell}$$

$$T_s^{\ell v} = \frac{K_\ell}{K_v} T_s^{v\ell},$$

$$(1.3)$$

we can write

$$E(2\omega) = \frac{4\pi i\omega}{cK_v} \hat{\mathbf{e}}^{\text{out}} \cdot \mathbf{H}_{\ell} \cdot \boldsymbol{\mathcal{P}}(2\omega) = \frac{4\pi i\omega}{cK_v} \mathbf{e}_{\ell}^{2\omega} \cdot \boldsymbol{\mathcal{P}}(2\omega).$$

where,

$$\mathbf{e}^{2\omega}_{\ell} = \hat{\mathbf{e}}^{\text{out}} \cdot \left[\hat{\mathbf{s}} T_s^{v\ell} T_s^{\ell b} \hat{\mathbf{s}} + \hat{\mathbf{P}}_{v+} \frac{T_p^{v\ell} T_p^{\ell b}}{\epsilon_{\ell}(2\omega) \sqrt{\epsilon_b(2\omega)}} \left(\epsilon_b(2\omega) \sin \theta_{\text{in}} \hat{\mathbf{z}} - \epsilon_{\ell}(2\omega) K_b \hat{\mathbf{x}} \right) \right].$$

$$(1.4)$$

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

$$\mathbf{e}_{v}^{2\omega} = \hat{\mathbf{e}}^{\text{out}} \cdot \left[\hat{\mathbf{s}} T_{s}^{vb} \hat{\mathbf{s}} + \hat{\mathbf{P}}_{v+} \frac{T_{p}^{vb}}{\sqrt{\epsilon_{b}(2\omega)}} \left(\epsilon_{b}(2\omega) \sin \theta_{\text{in}} \hat{\mathbf{z}} - K_{b} \hat{\mathbf{x}} \right) \right], \tag{1.5}$$

which agrees with Eq. (3.8) of Ref. [2].

In the three layer model the nonlinear polarization is located in layer ℓ , and then we evaluate the fundamental field required in Eq. (1.1) in this layer as well, then we write

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

$$\tag{1.6}$$

1.1. THREE LAYER MODEL FOR SHG YIELD, WITHOUT MULTIPLE REFLECTIONS9

and following the steps that lead to Eq. (1.4), we find that

$$\mathbf{e}_{\ell}^{\omega} = \left[\hat{\mathbf{s}} t_{s}^{v\ell} t_{s}^{\ell b} \hat{\mathbf{s}} + \frac{t_{p}^{v\ell} t_{p}^{\ell b}}{\epsilon_{\ell}(\omega) \sqrt{\epsilon_{b}(\omega)}} \left(\epsilon_{b}(\omega) \sin \theta_{\text{in}} \hat{\mathbf{z}} + \epsilon_{\ell}(\omega) k_{b} \hat{\mathbf{x}} \right) \hat{\mathbf{p}}_{v-} \right] \cdot \hat{\mathbf{e}}^{\text{in}}. \quad (1.7)$$

If we would like to evaluate the fields in the bulk, instead of the layer ℓ , we simply take $\epsilon_{\ell}(\omega) = \epsilon_b(\omega) (t_{s,p}^{\ell b} = 1)$, to obtain

$$\mathbf{e}_{b}^{\omega} = \left[\hat{\mathbf{s}} t_{s}^{vb} \hat{\mathbf{s}} + \frac{t_{p}^{vb}}{\sqrt{\epsilon_{b}(\omega)}} \left(\sin \theta_{\text{in}} \hat{\mathbf{z}} + k_{b} \hat{\mathbf{x}} \right) \hat{\mathbf{p}}_{v-} \right] \cdot \hat{\mathbf{e}}^{\text{in}}, \tag{1.8}$$

that is in agreement with Eq. (3.5) of Ref. [2].

With e^{ω} we can write Eq. (1.1) as

$$\mathcal{P}(2\omega) = E_0^2 \chi : \mathbf{e}_\ell^\omega \mathbf{e}_\ell^\omega$$

and then from Eq. (1.1) we obtain that

$$|E(2\omega)|^{2} = |E_{0}|^{4} \frac{16\pi^{2}\omega^{2}}{c^{2}K_{v}^{2}} \left| \mathbf{e}_{\ell}^{2\omega} \cdot \boldsymbol{\chi} : \mathbf{e}_{\ell}^{\omega} \mathbf{e}_{\ell}^{\omega} \right|^{2}$$

$$\frac{c}{2\pi} |E(2\omega)|^{2} = \frac{32\pi^{3}\omega^{2}}{c^{3}\cos^{2}\theta_{\text{in}}} \left| \mathbf{e}_{\ell}^{2\omega} \cdot \boldsymbol{\chi} : \mathbf{e}_{\ell}^{\omega} \mathbf{e}_{\ell}^{\omega} \right|^{2} \left(\frac{c}{2\pi} |E_{0}|^{2} \right)^{2},$$

$$I(2\omega) = \frac{32\pi^{3}\omega^{2}}{c^{3}\cos^{2}\theta_{\text{in}}} \left| \mathbf{e}_{\ell}^{2\omega} \cdot \boldsymbol{\chi} : \mathbf{e}_{\ell}^{\omega} \mathbf{e}_{\ell}^{\omega} \right|^{2} I^{2}(\omega),$$

$$R(2\omega) = \frac{32\pi^{3}\omega^{2}}{c^{3}\cos^{2}\theta_{\text{in}}} \left| \mathbf{e}_{\ell}^{2\omega} \cdot \boldsymbol{\chi} : \mathbf{e}_{\ell}^{\omega} \mathbf{e}_{\ell}^{\omega} \right|^{2},$$

$$(1.9)$$

as the SHG yield. At this point we mention that to recover the results of Ref. [2] which are equivalent of those of Ref. [3], we take $\mathbf{e}_{\ell}^{2\omega} \to \mathbf{e}_{v}^{2\omega}$, $\mathbf{e}_{\ell}^{\omega} \to \mathbf{e}_{b}^{\omega}$ and then

$$R(2\omega) = \frac{32\pi^3 \omega^2}{c^3 \cos^2 \theta_{\rm in}} \left| \mathbf{e}_v^{2\omega} \cdot \boldsymbol{\chi} : \mathbf{e}_b^{\omega} \mathbf{e}_b^{\omega} \right|^2, \tag{1.10}$$

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 $\epsilon_b(\omega)$.

To complete the required formulas, we write down the Fresnel factors,

$$t_s^{ij}(\omega) = \frac{2k_i(\omega)}{k_i(\omega) + k_j(\omega)}, \qquad t_p^{ij}(\omega) = \frac{2k_i(\omega)\sqrt{\epsilon_i(\omega)\epsilon_j(\omega)}}{k_i(\omega)\epsilon_j(\omega) + k_j(\omega)\epsilon_i(\omega)},$$
$$r_s^{ij}(\omega) = \frac{k_i(\omega) - k_j(\omega)}{k_i(\omega) + k_j(\omega)}, \qquad r_p^{ij}(\omega) = \frac{k_i(\omega)\epsilon_j(\omega) - k_j\epsilon_i(\omega)}{k_i(\omega)\epsilon_j(\omega) + k_j(\omega)\epsilon_i(\omega)}.$$

1.2 \mathcal{R} for different polarization cases

1.2.1 \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 $\hat{\mathbf{e}}^{\text{in}} = \hat{\mathbf{p}}_{v-}$ in Eq. (1.7), and $\hat{\mathbf{e}}^{\text{out}} = \hat{\mathbf{P}}_{v+}$ in Eq. (1.4).

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

$$r_{pP}^{\ell} = \epsilon_b(2\omega)\sin\theta_{\rm in}\left(\epsilon_b^2(\omega)\sin^2\theta_{\rm in}\chi_{zzz} + \epsilon_\ell^2(\omega)k_b^2\chi_{zxx}\right)$$

$$-\epsilon_\ell(2\omega)\epsilon_\ell(\omega)k_bK_b\left(2\epsilon_b(\omega)\sin\theta_{\rm in}\chi_{xxz} + \epsilon_\ell(\omega)k_b\chi_{xxx}\cos(3\phi)\right),$$
(1.11)

and

$$\Gamma_{pP}^{\ell} = \frac{T_p^{\ell v} T_p^{\ell b}}{\epsilon_{\ell}(2\omega) \sqrt{\epsilon_b(2\omega)}} \left(\frac{t_p^{v\ell} t_p^{\ell b}}{\epsilon_{\ell}(\omega) \sqrt{\epsilon_b(\omega)}} \right)^2. \tag{1.12}$$

1.2.2 \mathcal{R}_{pS}

To obtain $R_{pS}(2\omega)$ we use $\hat{\mathbf{e}}^{\text{in}} = \hat{\mathbf{p}}_{v-}$ in Eq. (1.7), and $\hat{\mathbf{e}}^{\text{out}} = \hat{\mathbf{S}}$ in Eq. (1.4). We also use the unit vectors defined in Eqs. (A.1) and (A.2). Substituting, we get

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

where

$$r_{pS}^{\ell} = -\epsilon_{\ell}^{2}(\omega)k_{b}^{2}\sin 3\phi \chi_{xxx}, \qquad (1.13)$$

and

$$\Gamma_{pS}^{\ell} = T_s^{v\ell} T_s^{\ell b} \left(\frac{t_p^{v\ell} t_p^{\ell b}}{\epsilon_{\ell}(\omega) \sqrt{\epsilon_b(\omega)}} \right)^2. \tag{1.14}$$

In order to reduce above result to that of Ref. [2] and [3], 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,

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

$$\Gamma^b_{pS} = T^{vb}_s \left(\frac{t^{vb}_p}{\sqrt{\epsilon_b(\omega)}} \right)^2.$$

1.2.3 \mathcal{R}_{sP}

To obtain $R_{sP}(2\omega)$ we use $\hat{\mathbf{e}}^{\text{in}} = \hat{\mathbf{s}}$ in Eq. (1.7), and $\hat{\mathbf{e}}^{\text{out}} = \hat{\mathbf{P}}_{v+}$ in Eq. (1.4). We also use the unit vectors defined in Eqs. (A.1) and (A.2). Substituting, we get

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

where

$$r_{sP}^{\ell} = \epsilon_b(2\omega)\sin\theta_{\rm in}\chi_{zxx} + \epsilon_{\ell}(2\omega)K_b\chi_{xxx}\cos3\phi, \qquad (1.15)$$

and

$$\Gamma_{sP}^{\ell} = \frac{T_p^{\ell v} T_p^{\ell b} \left(t_s^{v\ell} t_s^{\ell b} \right)^2}{\epsilon_{\ell}(2\omega) \sqrt{\epsilon_b(2\omega)}}.$$
(1.16)

In order to reduce above result to that of Ref. [2] and [3], 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,

$$r_{sP}^b = \epsilon_b(2\omega)\sin\theta_{\rm in}\chi_{zxx} + K_b\chi_{xxx}\cos3\phi_{sp}$$

and

$$\Gamma^b_{sP} = \frac{T^{vb}_p(t^{vb}_s)^2}{\sqrt{\epsilon_b(2\omega)}}.$$

1.2.4 \mathcal{R}_{sS}

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

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

where

$$r_{sS}^{\ell} = \chi_{xxx} \sin 3\phi, \tag{1.17}$$

and

$$\Gamma_{sS}^{\ell} = T_s^{v\ell} T_s^{\ell b} \left(t_s^{v\ell} t_s^{\ell b} \right)^2. \tag{1.18}$$

In order to reduce above result to that of Ref. [2] and [3], 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,

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

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

iF	Γ_{iF}^ℓ	r_{iF}^{ℓ}
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	$T_s^{v\ell} R_s^{M+} \left(\frac{t_p^{v\ell} t_p^{\ell b}}{n_\ell^2 n_b} \right)^2$	$-n_{\ell}^{4}w_{b}^{2}\chi_{xxx}\sin 3\phi$ $R_{p}^{M+}\sin\theta_{0}\chi_{zxx}+R_{p}^{M-}W_{\ell}\chi_{xxx}\cos 3\phi$
sP	$rac{T_p^{v\ell}}{N_\ell} \left(t_s^{v\ell} t_s^{\ell b} ight)^2$	$R_p^{M+} \sin \theta_0 \chi_{zxx} + R_p^{M-} W_\ell \chi_{xxx} \cos 3\phi$
sS	$T_s^{v\ell}R_s^{M+}\left(t_s^{v\ell}t_s^{\ell b} ight)^2$	$\chi_{xxx} \sin 3\phi$

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

1.2.5 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 κz plane, and define

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

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

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

and neglecting the multiple reflections reduces this expression to

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

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

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

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 $\hat{\mathbf{P}}_{v+}$,

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

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 $\hat{\mathbf{e}}^{\text{in}} = \hat{\mathbf{p}}_{v-}$,

$$\mathbf{e}_{\ell}^{\omega} \mathbf{e}_{\ell}^{\omega} = \left(\frac{t_{p}^{\nu\ell} t_{p}^{\ellb}}{n_{\ell}^{2} n_{b}}\right)^{2} \left(n_{\ell}^{4} w_{b}^{2} \cos^{2} \phi \hat{\mathbf{x}} \hat{\mathbf{x}} + 2n_{\ell}^{4} w_{b}^{2} \sin \phi \cos \phi \hat{\mathbf{x}} \hat{\mathbf{y}}\right)$$

$$+ 2n_{\ell}^{2} n_{b}^{2} w_{b} \sin \theta_{0} \cos \phi \hat{\mathbf{x}} \hat{\mathbf{z}} + n_{\ell}^{4} w_{b}^{2} \sin^{2} \phi \hat{\mathbf{y}} \hat{\mathbf{y}}$$

$$+ 2n_{\ell}^{2} n_{b}^{2} w_{b} \sin \theta_{0} \sin \phi \hat{\mathbf{y}} \hat{\mathbf{z}} + n_{b}^{4} \sin^{2} \theta_{0} \hat{\mathbf{z}} \hat{\mathbf{z}}\right),$$
(A.9)

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

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

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 $\chi(-2\omega;\omega,\omega)$.

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

The (111) surface

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

$$\chi_{xxx} = -\chi_{xyy} = -\chi_{yyx},$$

$$\chi_{xxz} = \chi_{yyz},$$

$$\chi_{zxx} = \chi_{zyy},$$

$$\chi_{zzz}.$$
(A.11)

The (110) surface

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

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

Case	$\hat{\mathbf{e}}^{\mathrm{out}}$	$\hat{\mathbf{e}}^{ ext{in}}$	$\mathbf{e}_{\ell}^{2\omega}$	$\mathbf{e}_{\ell}^{\omega}\mathbf{e}_{\ell}^{\omega}$
\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.

The (001) surface

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

$$\chi_{xxz} = \chi_{yyz},$$

$$\chi_{zxx} = \chi_{zyy},$$

$$\chi_{zzz}.$$
(A.13)

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

In this section, we derive the expressions 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.

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

$$\Upsilon_{pP}^{\text{MR}} = \Gamma_{pP}^{\text{MR}} \left[-R_p^{M-} W_\ell \left(n_\ell^4 w_b^2 \cos^3 \phi \chi_{xxx} + 2 n_\ell^4 w_b^2 \sin \phi \cos^2 \phi \chi_{xxy} \right. \right. \\ \left. + 2 n_\ell^2 n_b^2 w_b \sin \theta_0 \cos^2 \phi \chi_{xxz} + n_\ell^4 w_b^2 \sin^2 \phi \cos \phi \chi_{xyy} \right. \\ \left. + 2 n_\ell^2 n_b^2 w_b \sin \theta_0 \sin \phi \cos \phi \chi_{xyz} + n_b^4 \sin^2 \theta_0 \cos \phi \chi_{xyz} \right. \\ \left. - R_p^{M-} W_\ell \left(n_\ell^4 w_b^2 \sin \phi \cos^2 \phi \chi_{yxx} + 2 n_\ell^4 w_b^2 \sin^2 \phi \cos \phi \chi_{yxy} \right. \\ \left. + 2 n_\ell^2 n_b^2 w_b \sin \theta_0 \sin \phi \cos \phi \chi_{yxz} + n_\ell^4 w_b^2 \sin^3 \phi \chi_{yyy} \right. \\ \left. + 2 n_\ell^2 n_b^2 w_b \sin \theta_0 \sin^2 \phi \chi_{yyz} + n_b^4 \sin^2 \theta_0 \sin \phi \chi_{yzz} \right) \right. \\ \left. + R_p^{M+} \sin \theta_0 \left(n_\ell^4 w_b^2 \cos^2 \phi \chi_{zxx} + 2 n_\ell^4 w_b^2 \sin \phi \cos \phi \chi_{zxy} \right. \\ \left. + n_\ell^4 w_b^2 \sin^2 \phi \chi_{zyy} + 2 n_\ell^2 n_b^2 w_b \sin \theta_0 \cos \phi \chi_{zzx} \right. \\ \left. + 2 n_\ell^2 n_b^2 w_b \sin \theta_0 \sin \phi \chi_{zzy} + n_b^4 \sin^2 \theta_0 \chi_{zzz} \right) \right].$$

$$\left. (A.14) \right.$$

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

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

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

$$\Upsilon_{pP} = \Gamma_{pP} \left[-N_{\ell}^{2} W_{b} \left(+ n_{\ell}^{4} w_{b}^{2} \cos^{3} \phi \chi_{xxx} + 2n_{\ell}^{4} w_{b}^{2} \sin \phi \cos^{2} \phi \chi_{xxy} \right. \right. \\ \left. + 2n_{b}^{2} n_{\ell}^{2} w_{b} \sin \theta_{0} \cos^{2} \phi \chi_{xxz} + n_{\ell}^{4} w_{b}^{2} \sin^{2} \phi \cos \phi \chi_{xyy} \right. \\ \left. + 2n_{b}^{2} n_{\ell}^{2} w_{b} \sin \theta_{0} \sin \phi \cos \phi \chi_{xxz} + n_{b}^{4} \sin^{2} \theta_{0} \cos \phi \chi_{xyz} \right. \\ \left. + 2n_{b}^{2} n_{\ell}^{2} w_{b} \sin \theta_{0} \sin \phi \cos \phi \chi_{xyz} + n_{b}^{4} \sin^{2} \theta_{0} \cos \phi \chi_{xzz} \right) \\ \left. - N_{\ell}^{2} W_{b} \left(+ n_{\ell}^{4} w_{b}^{2} \sin \phi \cos^{2} \phi \chi_{yxx} + 2n_{\ell}^{4} w_{b}^{2} \sin^{2} \phi \cos \phi \chi_{yxy} \right. \\ \left. + 2n_{b}^{2} n_{\ell}^{2} w_{b} \sin \theta_{0} \sin \phi \cos \phi \chi_{yxz} + n_{\ell}^{4} w_{b}^{2} \sin^{3} \phi \chi_{yyy} \right. \\ \left. + 2n_{b}^{2} n_{\ell}^{2} w_{b} \sin \theta_{0} \sin^{2} \phi \chi_{yyz} + n_{b}^{4} \sin^{2} \theta_{0} \sin \phi \chi_{yzz} \right) \\ \left. + N_{b}^{2} \sin \theta_{0} \left(+ n_{\ell}^{4} w_{b}^{2} \cos^{2} \phi \chi_{zxx} + 2n_{\ell}^{4} w_{b}^{2} \sin \phi \cos \phi \chi_{zxy} \right. \\ \left. + n_{\ell}^{4} w_{b}^{2} \sin^{2} \phi \chi_{zyy} + 2n_{\ell}^{2} n_{b}^{2} w_{b} \sin \theta_{0} \cos \phi \chi_{zzz} \right. \\ \left. + 2n_{\ell}^{2} n_{b}^{2} w_{b} \sin \theta_{0} \sin \phi \chi_{zzy} + n_{b}^{4} \sin^{2} \theta_{0} \chi_{zzz} \right) \right],$$

$$\left. \left. \left(A.16 \right) \right.$$

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,

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

We reduce terms,

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

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

$$= \Gamma_{pP}^{\text{MR}} r_{pP}^{\text{MR},(111)},$$

where

$$r_{pP}^{\text{MR},(111)} = R_p^{M+} \sin \theta_0 (n_b^4 \sin^2 \theta_0 \chi_{zzz} + n_\ell^4 w_b^2 \chi_{zxx}) - R_p^{M-} n_\ell^2 w_b W_\ell (2n_b^2 \sin \theta_0 \chi_{xxz} + n_\ell^2 w_b \chi_{xxx} \cos 3\phi).$$
(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),

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

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and reduce,

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

where

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

$$\begin{split} \Upsilon_{pP}^{\text{MR},(110)} &= \Gamma_{pP}^{\text{MR}} \big[R_p^{M+} \sin \theta_0 \\ &\qquad \qquad (n_b^4 \sin^2 \theta_0 \chi_{zzz} + n_\ell^4 w_b^2 \cos^2 \phi \chi_{zxx} + n_\ell^4 w_b^2 \sin^2 \phi \chi_{zyy}) \\ &\qquad \qquad - 2 R_p^{M-} n_\ell^2 n_b^2 w_b W_\ell \sin \theta_0 (\cos^2 \phi \chi_{xxz} + \sin^2 \phi \chi_{yyz}) \big] \\ &= \Gamma_{pP}^{\text{MR}} \, r_{pP}^{\text{MR},(110)}, \end{split}$$

where

$$r_{pP}^{\text{MR},(110)} = R_p^{M+} \sin \theta_0 (n_b^4 \sin^2 \theta_0 \chi_{zzz} + n_\ell^4 w_b^2 \cos^2 \phi \chi_{zxx} + n_\ell^4 w_b^2 \sin^2 \phi \chi_{zyy}) - 2R_p^{M-} n_\ell^2 n_b^2 w_b W_\ell \sin \theta_0 (\cos^2 \phi \chi_{xxz} + \sin^2 \phi \chi_{yyz}).$$
(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),

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

where

$$r_{pP}^{(110)} = N_b^2 \sin \theta_0 (n_b^4 \sin^2 \theta_0 \chi_{zzz} + n_\ell^4 w_b^2 \cos^2 \phi \chi_{zxx} + n_\ell^4 w_b^2 \sin^2 \phi \chi_{zyy}) - 2n_b^2 n_\ell^2 N_\ell^2 w_b W_b \sin \theta_0 (\cos^2 \phi \chi_{xxz} + \sin^2 \phi \chi_{yyz}).$$
(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,

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

where

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

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

where

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

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

Per Table 1.1, \mathcal{R}_{pS} requires Eqs. (A.8) and (A.9). After some algebra, we obtain that

$$\begin{aligned} \mathbf{e}_{\ell}^{2\omega} \cdot \boldsymbol{\chi} : \mathbf{e}_{\ell}^{\omega} \mathbf{e}_{\ell}^{\omega} &= T_{s}^{v\ell} R_{s}^{M+} \left(\frac{t_{p}^{v\ell} t_{p}^{\ell b}}{n_{\ell}^{2} n_{b}} \right)^{2} \left[-n_{b}^{4} \sin^{2}\theta_{0} \sin\phi \chi_{xzz} \right. \\ &\quad - n_{\ell}^{4} w_{b}^{2} \sin\phi \cos^{2}\phi \chi_{xxx} \\ &\quad - n_{\ell}^{4} w_{b}^{2} \sin^{3}\phi \chi_{xyy} \\ &\quad - 2n_{\ell}^{2} n_{b}^{2} w_{b} \sin\theta_{0} \sin\phi \cos\phi \chi_{xzx} \\ &\quad - 2n_{\ell}^{2} n_{b}^{2} w_{b} \sin\theta_{0} \sin^{2}\phi \chi_{xzy} \\ &\quad - 2n_{\ell}^{2} w_{b}^{2} \sin^{2}\phi \cos\phi \chi_{xxy} \\ &\quad + n_{b}^{4} \sin^{2}\theta_{0} \cos\phi \chi_{yzz} \\ &\quad + n_{\ell}^{4} w_{b}^{2} \cos^{3}\phi \chi_{yxx} \\ &\quad + n_{\ell}^{4} w_{b}^{2} \sin^{2}\phi \cos\phi \chi_{yyy} \\ &\quad + 2n_{\ell}^{2} n_{b}^{2} w_{b} \sin\theta_{0} \sin\phi \cos^{2}\phi \chi_{yzx} \\ &\quad + 2n_{\ell}^{2} n_{b}^{2} w_{b} \sin\theta_{0} \sin\phi \cos\phi \chi_{yzy} \\ &\quad + 2n_{\ell}^{2} n_{b}^{2} w_{b} \sin\phi \cos^{2}\phi \chi_{yxy} \right]. \end{aligned}$$

And neglection multiple refs,

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

$$T_s^{v\ell}T_s^{\ell b} \left(\frac{t_p^{\nu}t_p^{\ell b}}{\epsilon_{\ell}(\omega)\sqrt{\epsilon_b(\omega)}}\right)^2 \left[-\epsilon_b^2(\omega)\sin^2\theta_{\rm in}\sin\phi\chi_{xzz}\right. \\ \left. -2\epsilon_b(\omega)\epsilon_{\ell}(\omega)k_b\sin\theta_0\cos\phi\sin\phi\chi_{xxz} \right. \\ \left. -\epsilon_\ell^2(\omega)k_b^2\cos^2\phi\sin\phi\chi_{xxx} \right. \\ \left. -2\epsilon_\ell^2(\omega)k_b^2\cos\phi\sin^2\phi\chi_{xxy} \right. \\ \left. -2\epsilon_\ell^2(\omega)k_b^2\sin^3\phi\chi_{xyy} \right. \\ \left. -2\epsilon_b(\omega)\epsilon_{\ell}(\omega)k_b\sin\theta_0\sin^2\phi\chi_{xyz} \right. \\ \left. +\epsilon_b^2(\omega)\sin^2\theta_{\rm in}\cos\phi\chi_{yzz} \right. \\ \left. +\epsilon_b^2(\omega)\sin^2\theta_{\rm in}\cos\phi\chi_{yzz} \right. \\ \left. +2\epsilon_b(\omega)\epsilon_{\ell}(\omega)k_b\sin\theta_0\cos^2\phi\chi_{yxz} \right. \\ \left. +\epsilon_\ell^2(\omega)k_b^2\cos^3\phi\chi_{yxx} \right. \\ \left. +2\epsilon_\ell^2(\omega)k_b^2\cos^2\phi\sin\phi\chi_{yyy} \right. \\ \left. +2\epsilon_\ell^2(\omega)k_b^2\cos\phi\sin^2\phi\chi_{yyy} \right. \\ \left. +\epsilon_\ell^2(\omega)k_b^2\cos\phi\sin^2\phi\chi_{yyy} \right. \\ \left. +2\epsilon_\ell(\omega)k_b^2\cos\phi\sin^2\phi\chi_{yyy} \right].$$

and taking into account that $\chi_{xzz} = \chi_{xxy} = \chi_{xyz} = \chi_{yzz} = \chi_{yxz} = \chi_{yxx} = \chi_{yyy} = 0$, we have

$$\begin{split} &= \Gamma_{pS}^{\ell} \big[+ \epsilon_{\ell}^{2}(\omega) k_{b}^{2} \sin^{3} \phi \chi_{xxx} \\ &- 2 \epsilon_{\ell}^{2}(\omega) k_{b}^{2} \cos^{2} \phi \sin \phi \chi_{xxx} \\ &- \epsilon_{\ell}^{2}(\omega) k_{b}^{2} \cos^{2} \phi \sin \phi \chi_{xxx} \\ &+ 2 \epsilon_{b}(\omega) \epsilon_{\ell}(\omega) k_{b} \sin \theta_{0} \cos \phi \sin \phi \chi_{xxz} \\ &- 2 \epsilon_{b}(\omega) \epsilon_{\ell}(\omega) k_{b} \sin \theta_{0} \cos \phi \sin \phi \chi_{xxz} \big] \\ &= \Gamma_{pS}^{\ell} \big[\epsilon_{\ell}^{2}(\omega) k_{b}^{2} (\sin^{3} \phi - 3 \cos^{2} \phi \sin \phi) \chi_{xxx} \big] \\ &= \Gamma_{pS}^{\ell} \big[- \epsilon_{\ell}^{2}(\omega) k_{b}^{2} \sin 3\phi \chi_{xxx} \big]. \end{split}$$

We summarize as follows,

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

 $A.3. \mathcal{R}_{PS}$ 23

where

$$r_{nS}^{\ell} = -\epsilon_{\ell}^{2}(\omega)k_{b}^{2}\sin 3\phi \chi_{xxx},$$

and

$$\Gamma_{pS}^{\ell} = T_s^{v\ell} T_s^{\ell b} \left(\frac{t_p^{v\ell} t_p^{\ell b}}{\epsilon_{\ell}(\omega) \sqrt{\epsilon_b(\omega)}} \right)^2$$

In order to reduce above result to that of Ref. [2] and [3], we take the 2- ω 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,

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

and

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

A.3.1 For the (111) surface

For this surface we have that $\chi_{xzz} = \chi_{xzy} = \chi_{xxy} = \chi_{yzz} = \chi_{yxx} = \chi_{yyy} = \chi_{yzx} = 0$. We use the symmetry relations where necessary,

$$\begin{split} \Upsilon_{pP}^{\ell\ell} &= \Gamma_{pS}^{\ell} \big[-n_{\ell}^4 w_b^2 \sin\phi \cos^2\phi \chi_{xxx} \\ &+ n_{\ell}^4 w_b^2 \sin^3\phi \chi_{xxx} \\ &- 2n_{\ell}^2 n_b^2 w_b \sin\theta_0 \sin\phi \cos\phi \chi_{xxz} \\ &+ 2n_{\ell}^2 n_b^2 w_b \sin\theta_0 \sin\phi \cos\phi \chi_{xxz} \\ &- 2n_{\ell}^4 w_b^2 \sin\phi \cos^2\phi \chi_{xxx} \big], \end{split}$$

and reduce

$$\begin{split} \Upsilon_{pP}^{\ell\ell} &= \Gamma_{pS}^{\ell} \big[n_{\ell}^4 w_b^2 (\sin^3 \phi - 3 \sin \phi \cos^2 \phi) \chi_{xxx} \big] \\ &= \Gamma_{pS}^{\ell} \big[- n_{\ell}^4 w_b^2 \chi_{xxx} \sin 3\phi \big]. \end{split}$$

Finally, we express this final result in a more compact form,

$$\Upsilon_{pP}^{\ell\ell} = \Gamma_{pS}^{\ell} r_{pS}^{\ell}, \tag{A.24}$$

where

$$r_{pS}^{\ell} = -n_{\ell}^4 w_b^2 \chi_{xxx} \sin 3\phi, \qquad (A.25)$$

$$\Gamma_{pS}^{\ell} = T_s^{v\ell} R_s^{M+} \left(\frac{t_p^{v\ell} t_p^{\ell b}}{n_\ell^2 n_b} \right)^2. \tag{A.26}$$

A.3.2 For the (110) surface

A.3.3 For the (001) surface

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

Per Table 1.1, \mathcal{R}_{sP} requires Eqs. (A.7) and (A.10). After some algebra, we obtain that

$$\begin{split} \mathbf{e}_{\ell}^{2\omega} \cdot \boldsymbol{\chi} : \mathbf{e}_{\ell}^{\omega} \mathbf{e}_{\ell}^{\omega} &= \frac{T_p^{v\ell}}{N_\ell} \left(t_s^{v\ell} t_s^{\ell b} \right)^2 \left[+ R_p^{M+} \sin \theta_0 \sin^2 \phi \chi_{zxx} \right. \\ &\quad + R_p^{M+} \sin \theta_0 \cos^2 \phi \chi_{zyy} \\ &\quad - 2 R_p^{M+} \sin \theta_0 \sin \phi \cos \phi \chi_{zxy} \\ &\quad - R_p^{M-} W_\ell \sin^2 \phi \cos \phi \chi_{xxx} \\ &\quad - R_p^{M-} W_\ell \cos^3 \phi \chi_{xyy} \\ &\quad + 2 R_p^{M-} W_\ell \sin \phi \cos^2 \phi \chi_{xxy} \\ &\quad - R_p^{M-} W_\ell \sin^3 \phi \chi_{yxx} \\ &\quad - R_p^{M-} W_\ell \sin \phi \cos^2 \phi \chi_{yyy} \\ &\quad + 2 R_p^{M-} W_\ell \sin^2 \phi \cos \phi \chi_{yxy} \right]. \end{split}$$

We know that $\chi_{zxy} = \chi_{xxy} = \chi_{yxx} = \chi_{yyy} = 0$, so we can eliminate and reduce components,

$$\begin{aligned} \mathbf{e}_{\ell}^{2\omega} \cdot \boldsymbol{\chi} : \mathbf{e}_{\ell}^{\omega} \mathbf{e}_{\ell}^{\omega} &= \Gamma_{sP}^{\ell} \big[+ R_{p}^{M+} \sin \theta_{0} \sin^{2} \phi \chi_{zxx} \\ &+ R_{p}^{M+} \sin \theta_{0} \cos^{2} \phi \chi_{zxx} \\ &- R_{p}^{M-} W_{\ell} \sin^{2} \phi \cos \phi \chi_{xxx} \\ &+ R_{p}^{M-} W_{\ell} \cos^{3} \phi \chi_{xxx} \\ &- 2 R_{p}^{M-} W_{\ell} \sin^{2} \phi \cos \phi \chi_{xxx} \big] \\ &= \Gamma_{sP}^{\ell} \big[+ R_{p}^{M+} \sin \theta_{0} (\sin^{2} \phi + \cos^{2} \phi) \chi_{zxx} \\ &+ R_{p}^{M-} W_{\ell} (\cos^{3} \phi - 3 \sin^{2} \phi \cos \phi) \chi_{xxx} \big] \\ &= \Gamma_{sP}^{\ell} \big[+ R_{p}^{M+} \sin \theta_{0} \chi_{zxx} + R_{p}^{M-} W_{\ell} \chi_{xxx} \cos 3\phi \big]. \end{aligned}$$

Finally, we express this final result in a more compact form,

$$\mathbf{e}_{\ell}^{2\omega} \cdot \boldsymbol{\chi} : \mathbf{e}_{\ell}^{\omega} \mathbf{e}_{\ell}^{\omega} = \Gamma_{sP}^{\ell} r_{sP}^{\ell}, \tag{A.27}$$

 $A.4. \mathcal{R}_{SP}$ 25

where

$$r_{sP}^{\ell} = R_n^{M+} \sin \theta_0 \chi_{zxx} + R_n^{M-} W_{\ell} \chi_{xxx} \cos 3\phi, \tag{A.28}$$

and

$$\Gamma_{sP}^{\ell} = \frac{T_p^{v\ell}}{N_{\ell}} \left(t_s^{v\ell} t_s^{\ell b} \right)^2. \tag{A.29}$$

To obtain $R_{sP}(2\omega)$ we use $\hat{\mathbf{e}}^{\text{in}} = \hat{\mathbf{s}}$ in Eq. (1.7), and $\hat{\mathbf{e}}^{\text{out}} = \hat{\mathbf{P}}_{v+}$ in Eq. (1.4). We also use the unit vectors defined in Eqs. (A.1) and (A.2). Substituting, we get

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

for 2ω , and for the fundamental fields,

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

Therefore,

$$\mathbf{e}_{\ell}^{2\omega} \cdot \boldsymbol{\chi} : \mathbf{e}_{\ell}^{\omega} \mathbf{e}_{\ell}^{\omega} = \frac{T_{p}^{v\ell} T_{p}^{\ell b} \left(t_{s}^{v\ell} t_{s}^{\ell b}\right)^{2}}{\epsilon_{\ell}(2\omega) \sqrt{\epsilon_{b}(2\omega)}} \left[\epsilon_{b}(2\omega) \sin \theta_{0} \sin^{2} \phi \chi_{zxx} + \epsilon_{b}(2\omega) \sin \theta_{0} \cos^{2} \phi \chi_{zyy} - 2\epsilon_{b}(2\omega) \sin \theta_{0} \sin \phi \cos \phi \chi_{zxy} - \epsilon_{\ell}(2\omega) K_{b} \cos \phi \sin^{2} \phi \chi_{xxx} - \epsilon_{\ell}(2\omega) K_{b} \cos \phi \cos^{2} \phi \chi_{xyy} + 2\epsilon_{\ell}(2\omega) K_{b} \cos \phi \sin \phi \cos \phi \chi_{xxy} - \epsilon_{\ell}(2\omega) K_{b} \sin \phi \sin^{2} \phi \chi_{yxx} - \epsilon_{\ell}(2\omega) K_{b} \sin \phi \cos^{2} \phi \chi_{yyy} + 2\epsilon_{\ell}(2\omega) K_{b} \sin \phi \cos^{2} \phi \chi_{yyy} + 2\epsilon_{\ell}(2\omega) K_{b} \sin \phi \sin \phi \cos \phi \chi_{yxy}\right],$$

and taking into account that $\chi_{zxy} = \chi_{xxy} = \chi_{yxx} = \chi_{yyy} = 0$, we have

$$= \Gamma_{sP}^{\ell} \left[\epsilon_b(2\omega) \sin \theta_0 \sin^2 \phi \chi_{zxx} + \epsilon_b(2\omega) \sin \theta_0 \cos^2 \phi \chi_{zxx} \right.$$
$$\left. - \epsilon_{\ell}(2\omega) K_b \cos \phi \sin^2 \phi \chi_{xxx} + \epsilon_{\ell}(2\omega) K_b \cos^3 \phi \chi_{xxx} \right.$$
$$\left. - 2\epsilon_{\ell}(2\omega) K_b \sin^2 \phi \cos \phi \chi_{xxx} \right]$$

$$= \Gamma_{sP}^{\ell} \left[\epsilon_b(2\omega) \sin \theta_0 (\sin^2 \phi + \cos^2 \phi) \chi_{zxx} - \epsilon_{\ell}(2\omega) K_b(\cos \phi \sin^2 \phi - \cos^3 \phi + 2\sin^2 \phi \cos \phi) \chi_{xxx} \right]$$

$$= \Gamma_{sP}^{\ell} \left[\epsilon_b(2\omega) \sin \theta_0 \chi_{zxx} + \epsilon_{\ell}(2\omega) K_b(\cos^3 \phi - 3\sin^2 \phi \cos \phi) \chi_{xxx} \right]$$

$$= \Gamma_{sP}^{\ell} \left[\epsilon_b(2\omega) \sin \theta_0 \chi_{zxx} + \epsilon_{\ell}(2\omega) K_b \cos 3\phi \chi_{xxx} \right].$$

We summarize as follows,

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

where

$$r_{sP}^{\ell} = \epsilon_b(2\omega)\sin\theta_0\chi_{zxx} + \epsilon_{\ell}(2\omega)K_b\chi_{xxx}\cos3\phi,$$

and

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

In order to reduce above result to that of Ref. [2] and [3], we take the 2- ω 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,

$$r_{sP}^b = \epsilon_b(2\omega)\sin\theta_0\chi_{zxx} + K_b\chi_{xxx}\cos3\phi,$$

and

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

- A.4.1 For the (111) surface
- A.4.2 For the (110) surface
- A.4.3 For the (001) surface

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

$$\mathbf{e}_{\ell}^{2\omega} \cdot \boldsymbol{\chi} : \mathbf{e}_{\ell}^{\omega} \mathbf{e}_{\ell}^{\omega} = T_{s}^{v\ell} R_{s}^{M+} \left(t_{s}^{v\ell} t_{s}^{\ell b} \right)^{2} \left[-\sin^{3} \phi \chi_{xxx} - \sin \phi \cos^{2} \phi \chi_{xyy} + 2\sin^{2} \phi \cos \phi \chi_{xxy} + \sin^{2} \phi \cos \phi \chi_{xxy} + \sin^{2} \phi \cos \phi \chi_{yxx} + \cos^{3} \phi \chi_{yyy} - 2\sin \phi \cos^{2} \phi \chi_{uxy} \right].$$

 $A.5. \mathcal{R}_{SS}$ 27

As before, we know that $\chi_{xxy} = \chi_{yxx} = \chi_{yyy} = 0$, so we eliminate and reduce components,

$$\mathbf{e}_{\ell}^{2\omega} \cdot \boldsymbol{\chi} : \mathbf{e}_{\ell}^{\omega} \mathbf{e}_{\ell}^{\omega} = \Gamma_{sS}^{\ell} \left[-\sin^{3}\phi \chi_{xxx} + \sin\phi \cos^{2}\phi \chi_{xxx} + 2\sin\phi \cos^{2}\phi \chi_{xxx} \right]$$
$$= \Gamma_{sS}^{\ell} \left[(3\sin\phi \cos^{2}\phi - \sin^{3}\phi) \chi_{xxx} \right]$$
$$= \Gamma_{sS}^{\ell} \chi_{xxx} \sin 3\phi.$$

Finally, we express this final result in a more compact form,

$$\mathbf{e}_{\ell}^{2\omega} \cdot \boldsymbol{\chi} : \mathbf{e}_{\ell}^{\omega} \mathbf{e}_{\ell}^{\omega} = \Gamma_{sS}^{\ell} r_{sS}^{\ell}, \tag{A.30}$$

where

$$r_{sS}^{\ell} = \chi_{xxx} \sin 3\phi, \tag{A.31}$$

and

$$\Gamma_{sS}^{\ell} = T_s^{v\ell} R_s^{M+} \left(t_s^{v\ell} t_s^{\ell b} \right)^2. \tag{A.32}$$

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

$$\begin{split} \mathbf{e}_{\ell}^{2\omega} &= T_s^{v\ell} T_s^{\ell b} \left[-\sin\phi \hat{\mathbf{x}} + \cos\phi \hat{\mathbf{y}} \right], \\ \mathbf{e}_{\ell}^{\omega} \mathbf{e}_{\ell}^{\omega} &= \left(t_s^{v\ell} t_s^{\ell b} \right)^2 \left(\sin^2\phi \hat{\mathbf{x}} \hat{\mathbf{x}} + \cos^2\phi \hat{\mathbf{y}} \hat{\mathbf{y}} - 2\sin\phi\cos\phi \hat{\mathbf{x}} \hat{\mathbf{y}} \right). \end{split}$$

Therefore,

$$\begin{aligned} \mathbf{e}_{\ell}^{2\omega} \cdot \boldsymbol{\chi} : \mathbf{e}_{\ell}^{\omega} \mathbf{e}_{\ell}^{\omega} &= T_{s}^{v\ell} T_{s}^{\ell b} \left(t_{s}^{v\ell} t_{s}^{\ell b} \right)^{2} \left[-\sin^{3} \phi \chi_{xxx} - \sin \phi \cos^{2} \phi \chi_{xyy} + 2\sin^{2} \phi \cos \phi \chi_{xxy} \right. \\ &\quad + \sin^{2} \phi \cos \phi \chi_{yxx} + \cos^{3} \phi \chi_{yyy} - 2\sin \phi \cos^{2} \phi \chi_{yxy} \right] \\ &= T_{s}^{v\ell} T_{s}^{\ell b} \left(t_{s}^{v\ell} t_{s}^{\ell b} \right)^{2} \left[-\sin^{3} \phi \chi_{xxx} + 3\sin \phi \cos^{2} \phi \chi_{xxx} \right] \\ &= T_{s}^{v\ell} T_{s}^{\ell b} \left(t_{s}^{v\ell} t_{s}^{\ell b} \right)^{2} \chi_{xxx} \sin 3\phi \end{aligned}$$

Summarizing,

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

where

$$r_{sS}^{\ell} = \chi_{xxx} \sin 3\phi,$$

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

In order to reduce above result to that of Ref. [2] and [3], 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,

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

and

$$\Gamma^b_{sS} = T^{vb}_s \left(t^{vb}_s \right)^2.$$

- A.5.1 For the (111) surface
- A.5.2 For the (110) surface
- A.5.3 For the (001) surface

A.6 Some limiting cases of interest

A.6.1 The two layer model

In order to reduce above result to that of Ref. [2] and [3], 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,

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

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

A.6.2 Taking $\mathcal{P}(2\omega)$ and the fundamental fields in the bulk

To consider the 2ω fields in the bulk, we start with Eq. (1.2) 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

$$\begin{split} \mathbf{H}_{b} &= \hat{\mathbf{s}} \, T_{s}^{bv} \hat{\mathbf{s}} + \hat{\mathbf{P}}_{v+} T_{p}^{bv} \hat{\mathbf{P}}_{b+} \\ &= \frac{K_{b}}{K_{v}} \left(\hat{\mathbf{s}} \, T_{s}^{vb} \hat{\mathbf{s}} + \hat{\mathbf{P}}_{v+} T_{p}^{vb} \hat{\mathbf{P}}_{b+} \right) \\ &= \frac{K_{b}}{K_{v}} \left[\hat{\mathbf{s}} \, T_{s}^{vb} \hat{\mathbf{s}} + \hat{\mathbf{P}}_{v+} \frac{T_{p}^{vb}}{\sqrt{\epsilon_{b}(2\omega)}} (\sin \theta_{0} \hat{\mathbf{z}} - K_{b} \cos \phi \hat{\mathbf{x}} - K_{b} \sin \phi \hat{\mathbf{y}}) \right], \end{split}$$

and we define

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

For \mathcal{R}_{pP} , we require $\hat{\mathbf{e}}^{\text{out}} = \hat{\mathbf{P}}_{v+}$, 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. (1.8)

$$\mathbf{e}_{b}^{\omega} = \left[\hat{\mathbf{s}} t_{s}^{vb} \hat{\mathbf{s}} + \frac{t_{p}^{vb}}{\sqrt{\epsilon_{b}(\omega)}} \left(\sin \theta_{0} \hat{\mathbf{z}} + k_{b} \cos \phi \hat{\mathbf{x}} + k_{b} \sin \phi \hat{\mathbf{y}} \right) \hat{\mathbf{p}}_{v-} \right] \cdot \hat{\mathbf{e}}^{\text{in}},$$

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

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

$$\mathbf{e}_{b}^{\omega} \mathbf{e}_{b}^{\omega} = \frac{\left(t_{p}^{vb}\right)^{2}}{\epsilon_{b}(\omega)} \left(\sin\theta_{0}\hat{\mathbf{z}} + k_{b}\cos\phi\hat{\mathbf{x}} + k_{b}\sin\phi\hat{\mathbf{y}}\right)^{2}$$

$$= \frac{\left(t_{p}^{vb}\right)^{2}}{\epsilon_{b}(\omega)} \left(\sin^{2}\theta_{\text{in}}\hat{\mathbf{z}}\hat{\mathbf{z}} + k_{b}^{2}\cos^{2}\phi\hat{\mathbf{x}}\hat{\mathbf{x}} + k_{b}^{2}\sin^{2}\phi\hat{\mathbf{y}}\hat{\mathbf{y}}\right)$$

$$+ 2k_{b}\sin\theta_{0}\cos\phi\hat{\mathbf{z}}\hat{\mathbf{x}} + 2k_{b}\sin\theta_{0}\sin\phi\hat{\mathbf{z}}\hat{\mathbf{y}} + 2k_{b}^{2}\sin\phi\cos\phi\hat{\mathbf{x}}\hat{\mathbf{y}}$$

So lastly, we have that

$$\mathbf{e}_{b}^{2\omega} \cdot \boldsymbol{\chi} : \mathbf{e}_{b}^{\omega} \mathbf{e}_{b}^{\omega} = \frac{K_{b}}{K_{v}} \frac{T_{p}^{vb} \left(t_{p}^{vb}\right)^{2}}{\epsilon_{b}(\omega)\sqrt{\epsilon_{b}(2\omega)}} \left(\sin^{3}\theta_{\mathrm{in}}\chi_{zzz}\right. \\ + k_{b}^{2} \sin\theta_{0} \cos^{2}\phi\chi_{zxx} \\ + k_{b}^{2} \sin\theta_{0} \sin^{2}\phi\chi_{zyy} \\ + 2k_{b} \sin^{2}\theta_{\mathrm{in}} \cos\phi\chi_{zzx} \\ + 2k_{b} \sin^{2}\theta_{\mathrm{in}} \sin\phi\chi_{zzy} \\ + 2k_{b}^{2} \sin\theta_{0} \sin\phi\cos\phi\chi_{zxy} \\ - K_{b} \sin^{2}\theta_{\mathrm{in}} \cos\phi\chi_{xzz} \\ - k_{b}^{2}K_{b} \cos^{3}\phi\chi_{xxx} \\ - k_{b}^{2}K_{b} \sin^{2}\phi\cos\phi\chi_{xyy} \\ - 2k_{b}K_{b} \sin\theta_{0} \cos^{2}\phi\chi_{xzx} \\ - 2k_{b}K_{b} \sin\theta_{0} \sin\phi\cos\phi\chi_{xzy} \\ - 2k_{b}K_{b} \sin\theta_{0} \sin\phi\cos\phi\chi_{xzy} \\ - K_{b} \sin^{2}\theta_{\mathrm{in}} \sin\phi\chi_{yzz} \\ - K_{b} \sin^{2}\theta_{\mathrm{in}} \sin\phi\chi_{yzz} \\ - k_{b}^{2}K_{b} \sin^{3}\phi\chi_{yyy} \\ - 2k_{b}K_{b} \sin\theta_{0} \sin\phi\cos\phi\chi_{yzx} \\ - 2k_{b}K_{b} \sin\theta_{0} \sin\phi\cos\phi\chi_{yzx} \\ - 2k_{b}K_{b} \sin\theta_{0} \sin\phi\cos\phi\chi_{yzx} \\ - 2k_{b}K_{b} \sin\theta_{0} \sin^{2}\phi\chi_{yzy} \\ - 2k_{b}K_{b} \sin^{2}\phi\cos\phi\chi_{yzy} \right),$$

and we can eliminate many terms since $\chi_{zzx} = \chi_{zzy} = \chi_{zxy} = \chi_{xzz} = \chi_{xzz} = \chi_{xzy} = \chi_{yzz} = \chi_{yzz} = \chi_{yzx} = \chi_{yyz} = \chi_{yzx} = 0$, and substituting the

equivalent components of χ ,

$$= \frac{K_b}{K_v} \Gamma_{pP}^b \left(\sin^3 \theta_{\text{in}} \chi_{zzz} \right.$$

$$+ k_b^2 \sin \theta_0 \cos^2 \phi \chi_{zxx}$$

$$+ k_b^2 \sin \theta_0 \sin^2 \phi \chi_{zxx}$$

$$- 2k_b K_b \sin \theta_0 \cos^2 \phi \chi_{xxz}$$

$$- 2k_b K_b \sin \theta_0 \sin^2 \phi \chi_{xxz}$$

$$- k_b^2 K_b \cos^3 \phi \chi_{xxx}$$

$$+ k_b^2 K_b \sin^2 \phi \cos \phi \chi_{xxx}$$

$$+ 2k_b^2 K_b \sin^2 \phi \cos \phi \chi_{xxx} \right),$$

and reducing,

$$= \frac{K_b}{K_v} \Gamma_{pP}^b \left(\sin^3 \theta_{\rm in} \chi_{zzz} \right.$$

$$+ k_b^2 \sin \theta_0 (\sin^2 \phi + \cos^2 \phi) \chi_{zxx}$$

$$- 2k_b K_b \sin \theta_0 (\sin^2 \phi + \cos^2 \phi) \chi_{xxz}$$

$$+ k_b^2 K_b (3 \sin^2 \phi \cos \phi - \cos^3 \phi) \chi_{xxx} \right)$$

$$=\frac{K_b}{K_v}\Gamma^b_{pP}\left(\sin^3\theta_{\rm in}\chi_{zzz}+k_b^2\sin\theta_0\chi_{zxx}-2k_bK_b\sin\theta_0\chi_{xxz}-k_b^2K_b\chi_{xxx}\cos3\phi\right),$$

where,

$$\Gamma_{pP}^{b} = \frac{T_{p}^{vb} (t_{p}^{vb})^{2}}{\epsilon_{b}(\omega) \sqrt{\epsilon_{b}(2\omega)}}.$$

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

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

A.6.3 Taking $\mathcal{P}(2\omega)$ and the fundamental fields in the vacuum

To consider the 1ω fields in the vacuum, we start with Eq. (1.6) but substitute $\ell \to v$, thus

$$\mathbf{E}_{v}(\omega) = E_{0} \left[\hat{\mathbf{s}} t_{s}^{vv} (1 + r_{s}^{vb}) \hat{\mathbf{s}} + \hat{\mathbf{p}}_{v-} t_{p}^{vv} \hat{\mathbf{p}}_{v-} + \hat{\mathbf{p}}_{v+} t_{p}^{vv} r_{p}^{vb} \hat{\mathbf{p}}_{v-} \right] \cdot \hat{\mathbf{e}}^{in},$$

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

$$\begin{aligned}
\mathbf{e}_{v}^{\omega} &= \left[\hat{\mathbf{s}} (1 + r_{s}^{vb}) \hat{\mathbf{s}} + \hat{\mathbf{p}}_{v-} \hat{\mathbf{p}}_{v-} + \hat{\mathbf{p}}_{v+} r_{p}^{vb} \hat{\mathbf{p}}_{v-} \right] \cdot \hat{\mathbf{e}}^{\text{in}} \\
&= \left[\hat{\mathbf{s}} (t_{s}^{vb}) \hat{\mathbf{s}} + (\hat{\mathbf{p}}_{v-} + \hat{\mathbf{p}}_{v+} r_{p}^{vb}) \hat{\mathbf{p}}_{v-} \right] \cdot \hat{\mathbf{e}}^{\text{in}} \\
&= \left[\hat{\mathbf{s}} (t_{s}^{vb}) \hat{\mathbf{s}} + \frac{1}{\sqrt{\epsilon_{v}(\omega)}} \left(k_{v} (1 - r_{p}^{vb}) \hat{\boldsymbol{\kappa}} + \sin \theta_{0} (1 + r_{p}^{vb}) \hat{\mathbf{z}} \right) \hat{\mathbf{p}}_{v-} \right] \\
&= \left[\hat{\mathbf{s}} (t_{s}^{vb}) \hat{\mathbf{s}} + \left(\frac{k_{b}}{\sqrt{\epsilon_{b}(\omega)}} t_{p}^{vb} \hat{\boldsymbol{\kappa}} + \sqrt{\epsilon_{b}(\omega)} \sin \theta_{0} t_{p}^{vb} \hat{\mathbf{z}} \right) \hat{\mathbf{p}}_{v-} \right] \cdot \hat{\mathbf{e}}^{\text{in}} \\
&= \left[\hat{\mathbf{s}} (t_{s}^{vb}) \hat{\mathbf{s}} + \frac{t_{p}^{vb}}{\sqrt{\epsilon_{b}(\omega)}} \left(k_{b} \cos \phi \hat{\mathbf{x}} + k_{b} \sin \phi \hat{\mathbf{y}} + \epsilon_{b}(\omega) \sin \theta_{0} \hat{\mathbf{z}} \right) \hat{\mathbf{p}}_{v-} \right] \cdot \hat{\mathbf{e}}^{\text{in}}.\end{aligned}$$

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

$$\mathbf{e}_{v}^{\omega}\mathbf{e}_{v}^{\omega} = \left(\frac{t_{p}^{vb}}{\sqrt{\epsilon_{b}(\omega)}}\right)^{2} \left[k_{b}^{2}\cos^{2}\phi\hat{\mathbf{x}}\hat{\mathbf{x}}\right]$$

$$+ k_{b}^{2}\sin^{2}\phi\hat{\mathbf{y}}\hat{\mathbf{y}}$$

$$+ \epsilon_{b}^{2}(\omega)\sin^{2}\theta_{\mathrm{in}}\hat{\mathbf{z}}\hat{\mathbf{z}}$$

$$+ 2k_{b}^{2}\sin\phi\cos\phi\hat{\mathbf{x}}\hat{\mathbf{y}}$$

$$+ 2\epsilon_{b}(\omega)k_{b}\sin\theta_{0}\sin\phi\hat{\mathbf{y}}\hat{\mathbf{z}}$$

$$+ 2\epsilon_{b}(\omega)k_{b}\sin\theta_{0}\cos\phi\hat{\mathbf{x}}\hat{\mathbf{z}}\right].$$

We also require the 2ω fields evaluated in the vacuum, which is Eq. (1.5),

$$\mathbf{e}_{v}^{2\omega} = \hat{\mathbf{e}}^{\text{out}} \cdot \left[\hat{\mathbf{s}} T_{s}^{vb} \hat{\mathbf{s}} + \hat{\mathbf{P}}_{v+} \frac{T_{p}^{vb}}{\sqrt{\epsilon_{b}(2\omega)}} \left(\epsilon_{b}(2\omega) \sin \theta_{0} \hat{\mathbf{z}} - K_{b} \hat{\boldsymbol{\kappa}} \right) \right], \quad (A.33)$$

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

$$\mathbf{e}_{v}^{2\omega} = \frac{T_{p}^{vb}}{\sqrt{\epsilon_{b}(2\omega)}} \left(\epsilon_{b}(2\omega)\sin\theta_{0}\hat{\mathbf{z}} - K_{b}\cos\phi\hat{\mathbf{x}} - K_{b}\sin\phi\hat{\mathbf{y}}\right). \tag{A.34}$$

So lastly, we have that

$$\begin{aligned} \mathbf{e}_{v}^{2\omega} \cdot \boldsymbol{\chi} : \mathbf{e}_{v}^{\omega} &= \\ \frac{T_{p}^{vb}}{\sqrt{\epsilon_{b}(2\omega)}} \left(\frac{t_{p}^{vb}}{\sqrt{\epsilon_{b}(\omega)}}\right)^{2} \left[\epsilon_{b}(2\omega)k_{b}^{2} \sin\theta_{0} \cos^{2}\phi \chi_{zxx} \right. \\ &+ \epsilon_{b}(2\omega)k_{b}^{2} \sin\theta_{0} \sin^{2}\phi \chi_{zyy} \\ &+ \epsilon_{b}^{2}(\omega)\epsilon_{b}(2\omega) \sin^{3}\theta_{\mathrm{in}}\chi_{zzz} \\ &+ 2\epsilon_{b}(2\omega)k_{b}^{2} \sin\theta_{0} \sin\phi \cos\phi \chi_{zxy} \\ &+ 2\epsilon_{b}(\omega)\epsilon_{b}(2\omega)k_{b} \sin^{2}\theta_{\mathrm{in}} \sin\phi \chi_{zyz} \\ &+ 2\epsilon_{b}(\omega)\epsilon_{b}(2\omega)k_{b} \sin^{2}\theta_{\mathrm{in}} \cos\phi \chi_{zxz} \\ &- k_{b}^{2}K_{b} \cos^{3}\phi \chi_{xxx} \\ &- k_{b}^{2}K_{b} \sin^{2}\phi \cos\phi \chi_{xyy} \\ &- \epsilon_{b}^{2}(\omega)K_{b} \sin^{2}\theta_{\mathrm{in}} \cos\phi \chi_{xzz} \\ &- 2k_{b}^{2}K_{b} \sin\phi \cos^{2}\phi \chi_{xxy} \\ &- 2\epsilon_{b}(\omega)k_{b}K_{b} \sin\theta_{0} \sin\phi \cos\phi \chi_{xyz} \\ &- 2\epsilon_{b}(\omega)k_{b}K_{b} \sin\theta_{0} \cos^{2}\phi \chi_{xxz} \\ &- k_{b}^{2}K_{b} \sin^{3}\phi \chi_{yyy} \\ &- \epsilon_{b}^{2}(\omega)K_{b} \sin^{2}\theta_{\mathrm{in}} \sin\phi \chi_{yzz} \\ &- 2k_{b}^{2}K_{b} \sin^{2}\phi \cos\phi \chi_{yxy} \\ &- 2\epsilon_{b}(\omega)k_{b}K_{b} \sin\theta_{0} \sin^{2}\phi \chi_{yyz} \\ &- 2\epsilon_{b}(\omega)k_{b}K_{b} \sin\theta_{0} \sin\phi \cos\phi \chi_{yxz} \Big], \end{aligned}$$

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and after eliminating components,

$$= \Gamma_{pP}^{v} \left[\epsilon_{b}^{2}(\omega) \epsilon_{b}(2\omega) \sin^{3}\theta_{\text{in}} \chi_{zzz} \right.$$

$$+ \epsilon_{b}(2\omega) k_{b}^{2} \sin\theta_{0} \cos^{2}\phi \chi_{zxx}$$

$$+ \epsilon_{b}(2\omega) k_{b}^{2} \sin\theta_{0} \sin^{2}\phi \chi_{zxx}$$

$$- 2\epsilon_{b}(\omega) k_{b} K_{b} \sin\theta_{0} \cos^{2}\phi \chi_{xxz}$$

$$- 2\epsilon_{b}(\omega) k_{b} K_{b} \sin\theta_{0} \sin^{2}\phi \chi_{xxz}$$

$$+ 3k_{b}^{2} K_{b} \sin^{2}\phi \cos\phi \chi_{xxx}$$

$$- k_{b}^{2} K_{b} \cos^{3}\phi \chi_{xxx} \right]$$

$$= \Gamma_{pP}^{v} \left[\epsilon_{b}^{2}(\omega) \epsilon_{b}(2\omega) \sin^{3}\theta_{\text{in}} \chi_{zzz} + \epsilon_{b}(2\omega) k_{b}^{2} \sin\theta_{0} \chi_{zxx} \right.$$

$$- 2\epsilon_{b}(\omega) k_{b} K_{b} \sin\theta_{0} \chi_{xxz} - k_{b}^{2} K_{b} \chi_{xxx} \cos 3\phi \right],$$

where

$$\Gamma_{pP}^{v} = \frac{T_{p}^{vb} (t_{p}^{vb})^{2}}{\epsilon_{b}(\omega) \sqrt{\epsilon_{b}(2\omega)}}.$$

A.6.4 Taking $\mathcal{P}(2\omega)$ in ℓ and the fundamental fields in the bulk

For this scenario with $\hat{\mathbf{e}}^{\text{in}} = \hat{\mathbf{p}}_{v-}$ and $\hat{\mathbf{e}}^{\text{out}} = \hat{\mathbf{P}}_{v+}$, we obtain from Eq. (1.4),

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

and Eq. (1.8),

$$\mathbf{e}_{b}^{\omega}\mathbf{e}_{b}^{\omega} = \frac{\left(t_{p}^{vb}\right)^{2}}{\epsilon_{b}(\omega)} \left(\sin^{2}\theta_{\mathrm{in}}\hat{\mathbf{z}}\hat{\mathbf{z}} + k_{b}^{2}\cos^{2}\phi\hat{\mathbf{x}}\hat{\mathbf{x}} + k_{b}^{2}\sin^{2}\phi\hat{\mathbf{y}}\hat{\mathbf{y}}\right) + 2k_{b}\sin\theta_{0}\cos\phi\hat{\mathbf{z}}\hat{\mathbf{x}} + 2k_{b}\sin\theta_{0}\sin\phi\hat{\mathbf{z}}\hat{\mathbf{y}} + 2k_{b}^{2}\sin\phi\cos\phi\hat{\mathbf{x}}\hat{\mathbf{y}}.$$

Thus,

$$\begin{split} \mathbf{e}_{\ell}^{2\omega} \cdot \mathbf{\chi} : \mathbf{e}_{b}^{\omega} \mathbf{e}_{b}^{\omega} &= \frac{T_{p}^{v\ell} T_{p}^{\ell b} \left(t_{p}^{vb} \right)^{2}}{\epsilon_{\ell}(2\omega)\epsilon_{b}(\omega)\sqrt{\epsilon_{b}(2\omega)}} \Bigg[+ \epsilon_{b}(2\omega)\sin^{3}\theta_{\mathrm{in}}\chi_{zzz} \\ &+ \epsilon_{b}(2\omega)k_{b}^{2}\sin\theta_{0}\cos^{2}\phi\chi_{zxx} \\ &+ \epsilon_{b}(2\omega)k_{b}^{2}\sin\theta_{0}\sin^{2}\phi\chi_{zyy} \\ &+ 2\epsilon_{b}(2\omega)k_{b}\sin^{2}\theta_{\mathrm{in}}\cos\phi\chi_{zzx} \\ &+ 2\epsilon_{b}(2\omega)k_{b}\sin^{2}\theta_{\mathrm{in}}\sin\phi\chi_{zzy} \\ &+ 2\epsilon_{b}(2\omega)k_{b}^{2}\sin\theta_{0}\sin\phi\cos\phi\chi_{zxy} \\ &- \epsilon_{\ell}(2\omega)\sin^{2}\theta_{\mathrm{in}}K_{b}\cos\phi\chi_{xzz} \\ &- \epsilon_{\ell}(2\omega)k_{b}^{2}K_{b}\sin^{2}\phi\cos\phi\chi_{xxx} \\ &- \epsilon_{\ell}(2\omega)k_{b}^{2}K_{b}\sin^{2}\phi\cos\phi\chi_{xyy} \\ &- 2\epsilon_{\ell}(2\omega)k_{b}K_{b}\sin\theta_{0}\sin\phi\cos\phi\chi_{xzy} \\ &- 2\epsilon_{\ell}(2\omega)k_{b}K_{b}\sin\theta_{0}\sin\phi\cos\phi\chi_{xxy} \\ &- 2\epsilon_{\ell}(2\omega)k_{b}^{2}K_{b}\sin\phi\cos^{2}\phi\chi_{xxy} \\ &- \epsilon_{\ell}(2\omega)k_{b}^{2}K_{b}\sin^{2}\theta_{\mathrm{in}}\sin\phi\chi_{yzz} \\ &- \epsilon_{\ell}(2\omega)k_{b}^{2}K_{b}\sin^{3}\phi\chi_{yyy} \\ &- 2\epsilon_{\ell}(2\omega)k_{b}K_{b}\sin\theta_{0}\cos\phi\sin\phi\chi_{yzx} \\ &- 2\epsilon_{\ell}(2\omega)k_{b}K_{b}\sin\theta_{0}\sin^{2}\phi\chi_{yzy} \\ &- 2\epsilon_{\ell}(2\omega)k_{b}K_{b}\sin\theta_{0}\sin^{2}\phi\chi_{yzy} \\ &- 2\epsilon_{\ell}(2\omega)k_{b}K_{b}\sin\theta_{0}\sin^{2}\phi\chi_{yzy} \\ &- 2\epsilon_{\ell}(2\omega)k_{b}K_{b}\sin^{2}\phi\cos\phi\chi_{yxy} \Bigg]. \end{split}$$

We eliminate and replace components,

$$\begin{split} \mathbf{e}_{\ell}^{2\omega} \cdot \boldsymbol{\chi} : \mathbf{e}_{b}^{\omega} \mathbf{e}_{b}^{\omega} &= \Gamma_{pP}^{\ell b} \bigg[+ \epsilon_{b}(2\omega) \sin^{3}\theta_{\mathrm{in}} \chi_{zzz} \\ &+ \epsilon_{b}(2\omega) k_{b}^{2} \sin\theta_{0} \cos^{2}\phi \chi_{zxx} \\ &+ \epsilon_{b}(2\omega) k_{b}^{2} \sin\theta_{0} \sin^{2}\phi \chi_{zxx} \\ &- 2\epsilon_{\ell}(2\omega) k_{b} K_{b} \sin\theta_{0} \cos^{2}\phi \chi_{xxz} \\ &- 2\epsilon_{\ell}(2\omega) k_{b} K_{b} \sin\theta_{0} \sin^{2}\phi \chi_{xxz} \\ &- \epsilon_{\ell}(2\omega) k_{b}^{2} K_{b} \cos^{3}\phi \chi_{xxx} \\ &+ \epsilon_{\ell}(2\omega) k_{b}^{2} K_{b} \sin^{2}\phi \cos\phi \chi_{xxx} \\ &+ 2\epsilon_{\ell}(2\omega) k_{b}^{2} K_{b} \sin^{2}\phi \cos\phi \chi_{xxx} \bigg], \end{split}$$

so lastly

$$\mathbf{e}_{\ell}^{2\omega} \cdot \boldsymbol{\chi} : \mathbf{e}_{b}^{\omega} \mathbf{e}_{b}^{\omega} = \Gamma_{pP}^{\ell b} \left[\epsilon_{b}(2\omega) \sin^{3}\theta_{\mathrm{in}} \chi_{zzz} + \epsilon_{b}(2\omega) k_{b}^{2} \sin\theta_{0} \chi_{zxx} - 2\epsilon_{\ell}(2\omega) k_{b}K_{b} \sin\theta_{0} \chi_{xxz} - \epsilon_{\ell}(2\omega) k_{b}^{2} K_{b} \chi_{xxx} \cos 3\phi \right],$$

where

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

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