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

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1 Three layer model for SHG radiation

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

$$R(\omega) = \frac{I(2\omega)}{I^2(\omega)}, \tag{1}$$

with the intensity

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

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). 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). We decompose the field into s and p polarizations, then the electric field radiated by a polarization sheet,

$$\mathcal{P}_i(2\omega) = \chi_{ijk}E_j(\omega)E_k(\omega), \tag{3}$$

is given by [2],

$$(E_{p\pm}, E_s) = (\frac{2\pi i\tilde{\omega}^2}{w}\,\hat{\mathbf{p}}_\pm \cdot \boldsymbol{\mathcal{P}}, \frac{2\pi i\tilde{\omega}^2}{w}\,\hat{\mathbf{s}} \cdot \boldsymbol{\mathcal{P}}), \tag{4}$$

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

$$k_i(\omega) = \sqrt{\epsilon_i(\omega) - \sin^2\theta_i}, \tag{5}$$

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

$$\hat{\mathbf{p}}_{i\pm} = \frac{\mp k_i(\omega)\hat{\mathbf{x}} + \sin\theta_i\hat{\mathbf{z}}}{\sqrt{\epsilon_i(\omega)}} \tag{6}$$

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), thus $\hat{\mathbf{s}} = -\hat{\mathbf{y}}$. The function $k_i(\omega)$ is the projection of the wave vector

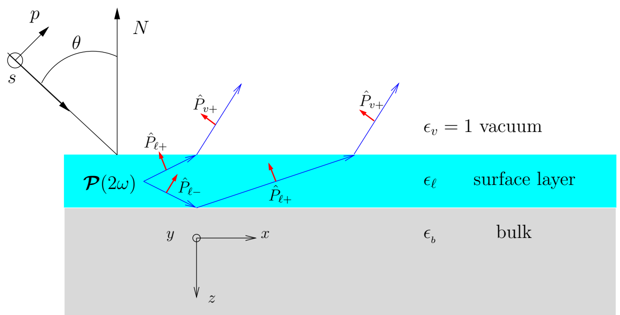

Figure 1: Sketch of the three layer model for SHG. Vacuum is on top with $\epsilon = 1$, the layer with nonlinear polarization $\mathbf{P}(2\omega)$ ischaracterized with $\epsilon_\ell(\omega)$ and the bulk with $\epsilon_b(\omega)$. In the dipolar approximation the bulk does not radiate SHG. The thin 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).

perpendicular to the surface. As we see from Fig. 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, $\mathbf{R}$, tensors as,

$$\mathbf{T}_{\ell v} = \hat{\mathbf{s}} T_s^{\ell v} \hat{\mathbf{s}} + \hat{\mathbf{P}}_{v+} T_p^{\ell v} \hat{\mathbf{P}}_{\ell+}, \tag{7}$$

and

$$\mathbf{R}_{\ell b} = \hat{\mathbf{s}} R_s^{\ell b} \hat{\mathbf{s}} + \hat{\mathbf{P}}_{\ell+} R_p^{\ell b} \hat{\mathbf{P}}_{\ell-}, \tag{8}$$

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

$$\begin{aligned}\mathbf{E}(2\omega) = {} & E_s(2\omega)\left(\mathbf{T}^{\ell v} + \mathbf{T}^{\ell v} \cdot \mathbf{R}^{\ell b}\right) \cdot \hat{\mathbf{s}} \\ & + E_{p+}(2\omega) \mathbf{T}^{\ell v} \cdot \hat{\mathbf{P}}_{\ell+} + E_{p-}(2\omega) \mathbf{T}^{\ell v} \cdot \mathbf{R}^{\ell b} \cdot \hat{\mathbf{P}}_{\ell-}.\end{aligned} \tag{9}$$

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

$$\mathbf{E}(2\omega) = \frac{2\pi i \tilde{\Omega}}{K_\ell} \mathbf{H}_\ell \cdot \boldsymbol{\mathcal{P}}(2\omega), \tag{10}$$

where,

$$\mathbf{H}_\ell = \hat{\mathbf{s}}\, T_s^{\ell v}\left(1 + R_s^{\ell b}\right) \hat{\mathbf{s}} + \hat{\mathbf{P}}_{v+} T_p^{\ell v}\left(\hat{\mathbf{P}}_{\ell+} + R_p^{\ell b} \hat{\mathbf{P}}_{\ell-}\right). \tag{11}$$

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

$$
\begin{aligned}
\hat{\mathbf{P}}_{\ell+} + R_p^{\ell b}\hat{\mathbf{P}}_{\ell-} &= \frac{\sin\theta_\ell \hat{z} - K_\ell \hat{x}}{\sqrt{\epsilon_\ell(2\omega)}} + R_p^{\ell b}\frac{\sin\theta_\ell \hat{z} + K_\ell \hat{x}}{\sqrt{\epsilon_\ell(2\omega)}} \\
&= \frac{1}{\sqrt{\epsilon_\ell(2\omega)}}\left(\sin\theta_\ell(1+R_p^{\ell b})\hat{z} - K_\ell(1-R_p^{\ell b})\hat{x}\right) \\
&= \frac{T_p^{\ell b}}{\epsilon_\ell(2\omega)\sqrt{\epsilon_b(2\omega)}}\left(\epsilon_b(2\omega)\sin\theta_\ell\hat{z} - \epsilon_\ell(2\omega)K_b\hat{x}\right),
\end{aligned}
\tag{12}
$$

where using

$$
\begin{aligned}
1 + R_s^{\ell b} &= T_s^{\ell b} \\
1 + R_p^{\ell b} &= \sqrt{\frac{\epsilon_b(2\omega)}{\epsilon_\ell(2\omega)}}T_p^{\ell b} \\
1 - R_p^{\ell b} &= \sqrt{\frac{\epsilon_\ell(2\omega)}{\epsilon_b(2\omega)}}\frac{K_b}{K_\ell}T_p^{\ell b} \\
T_p^{\ell v} &= \frac{K_\ell}{K_v}T_p^{v\ell} \\
T_s^{\ell v} &= \frac{K_\ell}{K_v}T_s^{v\ell},
\end{aligned}
\tag{13}
$$

we can write

$$
E(2\omega) = \frac{4\pi i\omega}{cK_v}\hat{\mathbf{e}}^{\mathrm{out}} \cdot \mathbf{H}_\ell \cdot \boldsymbol{\mathcal{P}}(2\omega) = \frac{4\pi i\omega}{cK_v}\mathbf{e}_\ell^{2\omega} \cdot \boldsymbol{\mathcal{P}}(2\omega). \tag{14}
$$

where,

$$
\mathbf{e}_\ell^{2\omega} = \hat{\mathbf{e}}^{\mathrm{out}} \cdot \left[\hat{\mathbf{s}}T_s^{v\ell}T_s^{\ell b}\hat{\mathbf{s}} + \hat{\mathbf{P}}_{v+}\frac{T_p^{v\ell}T_p^{\ell b}}{\epsilon_\ell(2\omega)\sqrt{\epsilon_b(2\omega)}}\left(\epsilon_b(2\omega)\sin\theta_\ell\hat{\mathbf{z}} - \epsilon_\ell(2\omega)K_b\hat{\mathbf{x}}\right)\right]. \tag{15}
$$

We mention that $n_v \sin\theta_{\mathrm{in}} = n_\ell \sin\theta_\ell$, from which $\sin\theta_\ell = \sin\theta_{\mathrm{in}}/n_\ell$ with $n_i = \sqrt{\epsilon_i(\omega)}$.

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

$$
\mathbf{e}_v^{2\omega} = \hat{\mathbf{e}}^{\mathrm{out}} \cdot \left[\hat{\mathbf{s}}T_s^{vb}\hat{\mathbf{s}} + \hat{\mathbf{P}}_{v+}\frac{T_p^{vb}}{\sqrt{\epsilon_b(2\omega)}}\left(\epsilon_b(2\omega)\sin\theta_{\mathrm{in}}\hat{\mathbf{z}} - K_b\hat{\mathbf{x}}\right)\right], \tag{16}
$$

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. (3) in this layer as well, then we write

$$\mathbf{E}_\ell(\omega) = E_0 \left(\hat{\mathbf{s}} t_s^{v\ell}(1 + r_s^{\ell b})\hat{\mathbf{s}} + \hat{\mathbf{p}}_{\ell -} t_p^{v\ell} \hat{\mathbf{p}}_{v-} + \hat{\mathbf{p}}_{\ell +} t_p^{v\ell} r_p^{\ell b} \hat{\mathbf{p}}_{v-}\right) \cdot \hat{\mathbf{e}}^{\mathrm{in}} = E_0 \mathbf{e}_\ell^\omega, \quad (17)$$

and following the steps that lead to Eq. (15), we find that

$$\mathbf{e}_\ell^\omega = \left[\hat{\mathbf{s}} t_s^{v\ell} t_s^{\ell b} \hat{\mathbf{s}} + \frac{t_p^{v\ell} t_p^{\ell b}}{\epsilon_\ell(\omega)\sqrt{\epsilon_b(\omega)}} \left(\epsilon_b(\omega) \sin\theta_\ell \hat{\mathbf{z}} + \epsilon_\ell(\omega) k_b \hat{\mathbf{x}}\right) \hat{\mathbf{p}}_{v-}\right] \cdot \hat{\mathbf{e}}^{\mathrm{in}}. \quad (18)$$

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

$$\mathbf{e}_b^\omega = \left[\hat{\mathbf{s}} t_s^{vb} \hat{\mathbf{s}} + \frac{t_p^{vb}}{\sqrt{\epsilon_b(\omega)}} \left(\sin\theta_{\mathrm{in}} \hat{\mathbf{z}} + k_b \hat{\mathbf{x}}\right) \hat{\mathbf{p}}_{v-}\right] \cdot \hat{\mathbf{e}}^{\mathrm{in}}, \quad (19)$$

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

With $\mathbf{e}^\omega$ we can write Eq. (3) as

$$\boldsymbol{\mathcal{P}}(2\omega) = E_0^2 \boldsymbol{\chi} : \mathbf{e}_\ell^\omega \mathbf{e}_\ell^\omega, \quad (20)$$

and then from Eq. (14) we obtain that

$$\begin{aligned}
|E(2\omega)|^2 &= |E_0|^4 \frac{16\pi^2\omega^2}{c^2 K_v^2} \left|\mathbf{e}_\ell^{2\omega} \cdot \boldsymbol{\chi} : \mathbf{e}_\ell^\omega \mathbf{e}_\ell^\omega\right|^2 \\
\frac{c}{2\pi}|E(2\omega)|^2 &= \frac{32\pi^3\omega^2}{c^3 \cos^2\theta_{\mathrm{in}}} \left|\mathbf{e}_\ell^{2\omega} \cdot \boldsymbol{\chi} : \mathbf{e}_\ell^\omega \mathbf{e}_\ell^\omega\right|^2 \left(\frac{c}{2\pi}|E_0|^2\right)^2, \\
I(2\omega) &= \frac{32\pi^3\omega^2}{c^3 \cos^2\theta_{\mathrm{in}}} \left|\mathbf{e}_\ell^{2\omega} \cdot \boldsymbol{\chi} : \mathbf{e}_\ell^\omega \mathbf{e}_\ell^\omega\right|^2 I^2(\omega), \\
R(2\omega) &= \frac{32\pi^3\omega^2}{c^3 \cos^2\theta_{\mathrm{in}}} \left|\mathbf{e}_\ell^{2\omega} \cdot \boldsymbol{\chi} : \mathbf{e}_\ell^\omega \mathbf{e}_\ell^\omega\right|^2, \quad (21)
\end{aligned}$$

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

$$R(2\omega) = \frac{32\pi^3\omega^2}{c^3 \cos^2\theta_{\mathrm{in}}} \left|\mathbf{e}_v^{2\omega} \cdot \boldsymbol{\chi} : \mathbf{e}_b^\omega \mathbf{e}_b^\omega\right|^2, \quad (22)$$

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,

$$\begin{aligned}
t_s^{ij}(\omega) &= \frac{2k_i(\omega)}{k_i(\omega) + k_j(\omega)}, \qquad & t_p^{ij}(\omega) &= \frac{2k_i(\omega)\sqrt{\epsilon_i(\omega)\epsilon_j(\omega)}}{k_i(\omega)\epsilon_j(\omega) + k_j(\omega)\epsilon_i(\omega)}, \\
r_s^{ij}(\omega) &= \frac{k_i(\omega) - k_j(\omega)}{k_i(\omega) + k_j(\omega)}, \qquad & r_p^{ij}(\omega) &= \frac{k_i(\omega)\epsilon_j(\omega) - k_j\epsilon_i(\omega)}{k_i(\omega)\epsilon_j(\omega) + k_j(\omega)\epsilon_i(\omega)}.
\end{aligned} \quad (23)$$

2 $\mathcal{R}$ for different polarization cases

We obtain explicit relations for a C_{3v} symmetry characteristic of a (111) surface, for which the only components of χ_{ijk} different from zero are χ_{zzz}, $\chi_{zxx} = \chi_{zyy}$, $\chi_{xxz} = \chi_{yyz}$ and $\chi_{xxx} = -\chi_{xyy} = -\chi_{yyx}$ with $\chi_{ijk} = \chi_{ikj}$, where we have chosen the x and y axes along the [112] and [110] directions, respectively.

However, we have to remember that the plane of incidence so far was chosen to be the xz plane; the most general plane of incidence should be one that makes an angle ϕ with respect to the x axis, and so $\hat{\mathbf{x}}$ should to be replaced by a unit vector $\hat{\kappa}$ such that

$$\hat{\boldsymbol{\kappa}} = \cos\phi\hat{\mathbf{x}} + \sin\phi\hat{\mathbf{y}}, \tag{24}$$

and then

$$\hat{\mathbf{s}} = -\sin\phi\hat{\mathbf{x}} + \cos\phi\hat{\mathbf{y}}, \tag{25}$$

2.1 $\mathcal{R}_{pP}$

To obtain $R_{pP}(2\omega)$ we use $\mathbf{e}^{\mathrm{in}} = \hat{\mathbf{p}}_{v-}$ in Eq. (18), and $\mathbf{e}^{\mathrm{out}} = \hat{\mathbf{P}}_{v+}$ in Eq. (15), to obtain that for a C_{3v} symmetry characteristic of a (111) surface,

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

where

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

and

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

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 $\epsilon_\ell(2\omega) = 1$, $T_p^{\ell v} = 1$, $T_p^{\ell b} = T_p^{vb}$, and the fundamental field inside medium b by taking $\ell = b$, thus $\epsilon_\ell(\omega) = \epsilon_b(\omega)$, $t_p^{v\ell} = t_p^{vb}$, $t_p^{\ell b} = 1$ and $\theta_\ell = \theta_{\mathrm{in}}$. With these choices,

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

and

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

2.2 $\mathcal{R}_{pS}$

To obtain $R_{pS}(2\omega)$ we use $\mathbf{e}^{\text{in}} = \hat{\mathbf{p}}_{v-}$ in Eq. (18), and $\mathbf{e}^{\text{out}} = \hat{\mathbf{S}}$ in Eq. (15). We also use the unit vectors defined in Eqs. (24) and (25). Substituting, we get

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

for 2ω, and for the fundamental fields,

$$\begin{aligned}
\hat{\mathbf{e}}_\ell^{\omega}\hat{\mathbf{e}}_\ell^{\omega} &= \left(\frac{t_p^{v\ell} t_p^{\ell b}}{\epsilon_\ell(\omega)\sqrt{\epsilon_b(\omega)}} \right)^2 \left(\epsilon_b(\omega) \sin\theta_\ell \hat{\mathbf{z}} + \epsilon_\ell(\omega) k_b \cos\phi \hat{\mathbf{x}} + \epsilon_\ell(\omega) k_b \sin\phi \hat{\mathbf{y}} \right)^2 . \\
&= \left(\frac{t_p^{v\ell} t_p^{\ell b}}{\epsilon_\ell(\omega)\sqrt{\epsilon_b(\omega)}} \right)^2 \big(\epsilon_b^2(\omega) \sin^2\theta_\ell \hat{\mathbf{z}}\hat{\mathbf{z}} + 2\epsilon_b(\omega)\epsilon_\ell(\omega) k_b \sin\theta_\ell \cos\phi \hat{\mathbf{z}}\hat{\mathbf{x}} \\
&\qquad + \epsilon_\ell^2(\omega) k_b^2 \cos^2\phi \hat{\mathbf{x}}\hat{\mathbf{x}} + 2\epsilon_\ell^2(\omega) k_b^2 \cos\phi \sin\phi \hat{\mathbf{x}}\hat{\mathbf{y}} \\
&\qquad + \epsilon_\ell^2(\omega) k_b^2 \sin^2\phi \hat{\mathbf{y}}\hat{\mathbf{y}} + 2\epsilon_b(\omega)\epsilon_\ell(\omega) k_b \sin\theta_\ell \sin\phi \hat{\mathbf{y}}\hat{\mathbf{z}} \big).
\end{aligned}$$

Therefore,

$$\begin{aligned}
&\hat{\mathbf{e}}_\ell^{2\omega} \cdot \boldsymbol{\chi} : \hat{\mathbf{e}}_\ell^{\omega}\hat{\mathbf{e}}_\ell^{\omega} = \\
&T_s^{v\ell} T_s^{\ell b} \left(\frac{t_p^{v\ell} t_p^{\ell b}}{\epsilon_\ell(\omega)\sqrt{\epsilon_b(\omega)}} \right)^2 \big[-\epsilon_b^2(\omega) \sin^2\theta_\ell \sin\phi \chi_{xzz} \\
&\qquad - 2\epsilon_b(\omega)\epsilon_\ell(\omega) k_b \sin\theta_\ell \cos\phi \sin\phi \chi_{xxz} \\
&\qquad - \epsilon_\ell^2(\omega) k_b^2 \cos^2\phi \sin\phi \chi_{xxx} \\
&\qquad - 2\epsilon_\ell^2(\omega) k_b^2 \cos\phi \sin^2\phi \chi_{xxy} \\
&\qquad - \epsilon_\ell^2(\omega) k_b^2 \sin^3\phi \chi_{xyy} \\
&\qquad - 2\epsilon_b(\omega)\epsilon_\ell(\omega) k_b \sin\theta_\ell \sin^2\phi \chi_{xyz} \\
&\qquad + \epsilon_b^2(\omega) \sin^2\theta_\ell \cos\phi \chi_{yzz} \\
&\qquad + 2\epsilon_b(\omega)\epsilon_\ell(\omega) k_b \sin\theta_\ell \cos^2\phi \chi_{yxz} \\
&\qquad + \epsilon_\ell^2(\omega) k_b^2 \cos^3\phi \chi_{yxx} \\
&\qquad + 2\epsilon_\ell^2(\omega) k_b^2 \cos^2\phi \sin\phi \chi_{yxy} \\
&\qquad + \epsilon_\ell^2(\omega) k_b^2 \cos\phi \sin^2\phi \chi_{yyy} \\
&\qquad + 2\epsilon_b(\omega)\epsilon_\ell(\omega) k_b \sin\theta_\ell \cos\phi \sin\phi \chi_{yyz} \big],
\end{aligned}$$

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

$$
\begin{aligned}
&= \Gamma^{\ell}_{pS}\big[+\epsilon^2_\ell(\omega)k_b^2 \sin^3\phi\chi_{xxx} \\
&\qquad -2\epsilon^2_\ell(\omega)k_b^2 \cos^2\phi\sin\phi\chi_{xxx} \\
&\qquad -\epsilon^2_\ell(\omega)k_b^2 \cos^2\phi\sin\phi\chi_{xxx} \\
&\qquad +2\epsilon_b(\omega)\epsilon_\ell(\omega)k_b \sin\theta_\ell\cos\phi\sin\phi\chi_{xxz} \\
&\qquad -2\epsilon_b(\omega)\epsilon_\ell(\omega)k_b \sin\theta_\ell\cos\phi\sin\phi\chi_{xxz}\big] \\
&= \Gamma^{\ell}_{pS}\big[\epsilon^2_\ell(\omega)k_b^2(\sin^3\phi - 3\cos^2\phi\sin\phi)\chi_{xxx}\big] \\
&= \Gamma^{\ell}_{pS}\big[-\epsilon^2_\ell(\omega)k_b^2\sin 3\phi\chi_{xxx}\big].
\end{aligned}
$$

We summarize as follows,

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

where

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

and

$$\Gamma^{\ell}_{pS} = T^{v\ell}_s T^{\ell b}_s \left(\frac{t^{v\ell}_p t^{\ell b}_p}{\epsilon_\ell(\omega)\sqrt{\epsilon_b(\omega)}}\right)^2$$

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 $\epsilon_\ell(2\omega) = 1$, $T^{v\ell}_s = 1$, $T^{\ell b}_s = T^{vb}_s$, and the fundamental field inside medium b by taking $\ell = b$, thus $\epsilon_\ell(\omega) = \epsilon_b(\omega)$, $t^{v\ell}_p = t^{vb}_p$, $t^{\ell b}_p = 1$ and $\theta_\ell = \theta_{\text{in}}$. With these choices,

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

and

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

2.3 $\mathcal{R}_{sP}$

To obtain $R_{sP}(2\omega)$ we use $\mathbf{e}^{\text{in}} = \hat{\mathbf{s}}$ in Eq. (18), and $\mathbf{e}^{\text{out}} = \hat{\mathbf{P}}_{v+}$ in Eq. (15). We also use the unit vectors defined in Eqs. (24) and (25). Substituting, we get

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

for 2ω, and for the fundamental fields,

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

Therefore,

$$
\begin{aligned}
\hat{\mathbf{e}}_\ell^{2\omega}\cdot\boldsymbol{\chi}:&\hat{\mathbf{e}}_\ell^{\omega}\hat{\mathbf{e}}_\ell^{\omega} = \\
&\frac{T_p^{v\ell}T_p^{\ell b}\left(t_s^{v\ell}t_s^{\ell b}\right)^2}{\epsilon_\ell(2\omega)\sqrt{\epsilon_b(2\omega)}}\big[\epsilon_b(2\omega)\sin\theta_\ell\sin^2\phi\chi_{zxx}+\epsilon_b(2\omega)\sin\theta_\ell\cos^2\phi\chi_{zyy}\\
&-2\epsilon_b(2\omega)\sin\theta_\ell\sin\phi\cos\phi\chi_{zxy}-\epsilon_\ell(2\omega)K_b\cos\phi\sin^2\phi\chi_{xxx}\\
&-\epsilon_\ell(2\omega)K_b\cos\phi\cos^2\phi\chi_{xyy}+2\epsilon_\ell(2\omega)K_b\cos\phi\sin\phi\cos\phi\chi_{xxy}\\
&-\epsilon_\ell(2\omega)K_b\sin\phi\sin^2\phi\chi_{yxx}-\epsilon_\ell(2\omega)K_b\sin\phi\cos^2\phi\chi_{yyy}\\
&+2\epsilon_\ell(2\omega)K_b\sin\phi\sin\phi\cos\phi\chi_{yxy}\big],
\end{aligned}
$$

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

$$
\begin{aligned}
&=\Gamma_{sP}^{\ell}\big[\epsilon_b(2\omega)\sin\theta_\ell\sin^2\phi\chi_{zxx}+\epsilon_b(2\omega)\sin\theta_\ell\cos^2\phi\chi_{zxx}\\
&\quad-\epsilon_\ell(2\omega)K_b\cos\phi\sin^2\phi\chi_{xxx}+\epsilon_\ell(2\omega)K_b\cos^3\phi\chi_{xxx}\\
&\quad-2\epsilon_\ell(2\omega)K_b\sin^2\phi\cos\phi\chi_{xxx}\big]
\end{aligned}
$$

$$
\begin{aligned}
&=\Gamma_{sP}^{\ell}\big[\epsilon_b(2\omega)\sin\theta_\ell(\sin^2\phi+\cos^2\phi)\chi_{zxx}\\
&\quad-\epsilon_\ell(2\omega)K_b(\cos\phi\sin^2\phi-\cos^3\phi+2\sin^2\phi\cos\phi)\chi_{xxx}\big]
\end{aligned}
$$

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

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

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

$$
r_{sP}^{\ell}=\epsilon_b(2\omega)\sin\theta_\ell\chi_{zxx}+\epsilon_\ell(2\omega)K_b\chi_{xxx}\cos 3\phi,
$$

and

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

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 $\epsilon_\ell(\omega)=\epsilon_b(\omega)$, $t_s^{v\ell}=t_s^{vb}$, $t_s^{\ell b}=1$ and $\theta_\ell=\theta_{\mathrm{in}}$. With these choices,

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

and

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

2.4 $\mathcal{R}_{sS}$

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

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

Therefore,

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

3 Trials and tribulations

3.1 Taking the 2ω fields in the bulk

To consider the 2ω fields in the bulk, we start with Eq. (11) 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

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

and we define

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

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

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

The 1ω fields will still be evaluated inside the bulk, so we have Eq. (19)

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

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

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

and

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

So lastly, we have that

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

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 $\boldsymbol{\chi}$,

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

and reducing,

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

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

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

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

3.2 Taking the 1ω fields in the vacuum

To consider the 1ω fields in the vacuum, we start with Eq. (17) but substitute $\ell \to v$, thus

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

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

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

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

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

and

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

We also require the 2ω fields evaluated in the vacuum, which is Eq. (16),

$$
\mathbf{e}_v^{2\omega} = \hat{\mathbf{e}}^{\mathrm{out}}\cdot\left[\hat{\mathbf{s}}T_s^{vb}\hat{\mathbf{s}} + \hat{\mathbf{P}}_{v+}\frac{T_p^{vb}}{\sqrt{\epsilon_b(2\omega)}}\left(\epsilon_b(2\omega)\sin\theta_{\mathrm{in}}\hat{\mathbf{z}} - K_b\hat{\boldsymbol{\kappa}}\right)\right], \tag{26}
$$

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

$$
\mathbf{e}_v^{2\omega} = \frac{T_p^{vb}}{\sqrt{\epsilon_b(2\omega)}}\left(\epsilon_b(2\omega)\sin\theta_{\mathrm{in}}\hat{\mathbf{z}} - K_b\cos\phi\hat{\mathbf{x}} - K_b\sin\phi\hat{\mathbf{y}}\right). \tag{27}
$$

So lastly, we have that

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

and after eliminating components,

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

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

where

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

4 Two layer model for SHG radiation

In this treatment we follow the work of Ref. [3]. They define the following for all polarizations;

$$\begin{aligned} f_s &= \frac{\kappa}{n\tilde{\omega}} = \frac{\kappa}{\sqrt{\epsilon(\omega)}\tilde{\omega}}, \\ f_c &= \frac{w}{n\tilde{\omega}} = \frac{w}{\sqrt{\epsilon(\omega)}\tilde{\omega}}, \\ f_s^2 &+ f_c^2 = 1, \end{aligned} \tag{28}$$

where

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

$$w_0 = \sqrt{\tilde{\omega} - \kappa^2} = \tilde{\omega}\cos\theta, \tag{29}$$

$$w = \sqrt{\tilde{\omega}\epsilon(\omega) - \kappa^2} = \tilde{\omega}k_z(\omega). \tag{30}$$

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,

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

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

$$\begin{aligned} A_p &= \frac{4\pi\tilde{\Omega}\sqrt{\epsilon(2\omega)}}{W_0\epsilon(2\omega) + W}, \\ A_s &= \frac{4\pi\tilde{\Omega}}{W_0 + W}. \end{aligned} \tag{32}$$

4.1 $\mathcal{R}_{pP}$

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

$$\frac{E^{(2\omega)}(\|,\|)}{E_p^2 A_p} = a_{\|,\|} + c^{(3)}_{\|,\|}\cos 3\phi. \tag{33}$$

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

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

We substitute these in Eq. (33),

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

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

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

As every term has an $f_i^2 F_i$, 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. (28),

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

We substitute Eq. (32) 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,

$$\begin{aligned}\mathcal{R}_{pP} &= \frac{2\pi}{c}\left|\frac{4i\pi\tilde{\omega}}{\cos\theta}\frac{1}{\epsilon(\omega)}\frac{2\cos\theta}{\epsilon(2\omega)\cos\theta + k_z(2\omega)} r_{pP}\right|^2 \\ &= \frac{32\pi^3\tilde{\omega}^2}{c\cos^2\theta}\left|t_p(\omega)T_p(2\omega)r_{pP}\right|^2 \\ &= \frac{32\pi^3\omega^2}{c^3\cos^2\theta}\left|t_p(\omega)T_p(2\omega)r_{pP}\right|^2,\end{aligned} \tag{34}$$

where

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

4.2 $\mathcal{R}_{pS}$

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

$$\frac{E^{(2\omega)}(\|,\perp)}{E_p^2 A_s} = b_{\|,\perp}^{(3)}\sin 3\phi, \tag{35}$$

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. (32) into Eq. (35),

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

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,

$$
\begin{aligned}
\mathcal{R}_{pS} &= \frac{2\pi}{c} \left| \frac{4i\pi\omega}{c\cos\theta} \frac{1}{\epsilon(\omega)} \frac{2\cos\theta}{\cos\theta + k_z(2\omega)} k_z^2(\omega) \chi^{xxx} \sin 3\phi \right|^2 \\
&= \frac{32\pi^3\omega^2}{c^3\cos^2\theta} \left| \frac{1}{\epsilon(\omega)} \frac{2\cos\theta}{\cos\theta + k_z(2\omega)} k_z^2(\omega) \chi^{xxx} \sin 3\phi \right|^2 \\
&= \frac{32\pi^3\omega^2}{c^3\cos^2\theta} \left| t_p(\omega) T_s(2\omega) k_z^2(\omega) r_{pS} \right|^2, \qquad (36)
\end{aligned}
$$

where

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

4.3 $\mathcal{R}_{sP}$

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

$$
\frac{E^{(2\omega)}(\perp,\|)}{E_s^2 A_p} = a_{\perp,\|} + c_{\perp,\|}^{(3)} \cos 3\phi, \qquad (37)
$$

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

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

Substituting this coeffecient and Eq. (32) into Eq. (37),

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

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

$$
\begin{aligned}
\mathcal{R}_{sP} &= \frac{2\pi}{c}\left|\frac{E^{(2\omega)}(\perp,\parallel)}{E_s^2}\right|^2\\
&= \frac{2\pi}{c}\left|\frac{4i\pi\omega}{c\cos\theta}\frac{2\cos\theta}{\epsilon(2\omega)\cos\theta + k_z(2\omega)}(\sin\theta\epsilon(2\omega)\chi^{zxx} + k_z(2\omega)\chi^{xxx}\cos 3\phi)\right|^2\\
&= \frac{32\pi^3\omega^2}{c^3\cos^2\theta}\left|\frac{2\cos\theta}{\epsilon(2\omega)\cos\theta + k_z(2\omega)}(\sin\theta\epsilon(2\omega)\chi^{zxx} + k_z(2\omega)\chi^{xxx}\cos 3\phi)\right|^2\\
&= \frac{32\pi^3\omega^2}{c^3\cos^2\theta}\left|t_s(\omega)T_p(2\omega)r_{sP}\right|^2, \qquad (38)
\end{aligned}
$$

where

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

4.4 Summary

We unify the final expressions for the SHG yield, Eqs. (34), (36), and (38), as

$$
\mathcal{R}_i F = \frac{32\pi^3\omega^2}{c^3\cos^2\theta}\left|t_i(\omega)T_F(2\omega)r_{iF}\right|^2. \qquad (39)
$$

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 1: The necessary factors for Eq. (39) for each polarization case.

The necessary factors are summarized in Table 1.

References

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

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