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

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(Dated: February 8, 2016)

I. THREE LAYER MODEL FOR SHG RADIATION

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

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

with the intensity

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

There are several ways to calculate R, one of which is the procedure followed by Cini ciniPRB91 [?]. 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 mizrahiJOSA88 [?], since the derivation of the three-layer-model is straightforward. In this scheme, we represent the surface by three regions or layers. The first layer is the vacuum region (denoted by v) with a dielectric function $\epsilon_v(\omega) = 1$ from where the fundamental electric field $\mathbf{E}_v(\omega)$ impinges on the material. The second layer is a thin layer (denoted by ℓ) of thickness d characterized by a dielectric function $\epsilon_\ell(\omega)$. Is in this layer where the second harmonic generation takes place. The third layer is the bulk region denoted by $b$and characterized by $\epsilon_b(\omega)$. Both the vacuum layer and the bulk layer are semiinfinite (see Fig. 3layer 1).

To model the electromagnetic response of the three-layer model we follow Ref. mizrahiJOSA88 ?, and assume a polarization sheet of the form

$$\mathbf{P}(\mathbf{r},t) = \boldsymbol{\mathcal{P}} e^{i\boldsymbol{\kappa}\cdot\mathbf{R}} e^{-i\omega t}\delta(z - z_\beta) + \text{c.c.}, \tag{3}$$ m31

where $\mathbf{R} = (x, y)$, $\boldsymbol{\kappa}$ is the component of the wave vector $\boldsymbol{\nu}_\beta$ paralel to the surface, and z_β is the position of the sheet within medium β (see Fig. 3layer 1). In Ref. sipeJOSAB87 ? it has been shown that the solution of the Maxwell equations for the radiated fields $E_{\beta,p\pm}$ and $E_{\beta,s}$ with $\mathbf{P}(\mathbf{r},t)$as a source can be written, at points $z \neq 0$, as

$$(E_{\beta,p\pm}, E_{\beta,s}) = (\frac{2\pi i\tilde{\omega}^2}{w_\beta}\,\hat{\mathbf{p}}_{\beta\pm}\cdot\boldsymbol{\mathcal{P}}, \frac{2\pi i\tilde{\omega}^2}{w_\beta}\,\hat{\mathbf{s}}\cdot\boldsymbol{\mathcal{P}}), \tag{4}$$ r2

where $\hat{\mathbf{s}}$ and $\hat{\mathbf{p}}_{\beta\pm}$ are the unitary vectors for the s and p polarization of the radiated field, respectively, and the $\pm$ refers to upward $(+)$ or downward $(-)$ direction of propagation within medium β, as shown in Fig. 3layer 1, and $\tilde{\omega} = \omega/c$. Also,

$$w_\beta(\omega) = \tilde{\omega}\big(\epsilon_\beta(\omega) - \sin^2\theta_0\big)^{1/2}, \tag{5}$$

r3

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

$$\hat{\mathbf{p}}_{\beta\pm}(\omega) = \frac{\kappa(\omega)\hat{\mathbf{z}} \mp w_\beta(\omega)\hat{\boldsymbol{\kappa}}}{\tilde{\omega} n_\beta(\omega)}, \tag{6}$$

r4

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

$$\hat{\boldsymbol{\kappa}} = \cos\phi\hat{\mathbf{x}} + \sin\phi\hat{\mathbf{y}}, \tag{7}$$

mc1

and

$$\hat{\mathbf{s}} = -\sin\phi\hat{\mathbf{x}} + \cos\phi\hat{\mathbf{y}}, \tag{8}$$

mmc2

where ϕ the angle with respect to the x axis.

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

$$\mathcal{P}_i(2\omega) = \chi_{ijk}(2\omega)E_j(\omega)E_k(\omega), \tag{9}$$

tres

where the tensor $\boldsymbol{\chi}(2\omega)$ is the surface nonlinear dipolar susceptibility and the Cartesian indices i, j, k are summed if repeated. `El rollo de la centrosimetria va en la introduccion` As it was done in Ref. mizrahiJOSA88 **?**, in presenting the results Eq. (r2 4)-(mmc2 8) we have taken the polarization sheet (Eq. (m31 3)) to be oscillating at some frequency ω. However, in the following we find it convenient to use ω exclusively to denote the fundamental frequency and $\boldsymbol{\kappa}$ to denote the component of the incident wave vector parallel to the surface. Then the nonlinear generated polarization is oscillating at $\Omega = 2\omega$ and will be characterized by a wave vector parallel to the surface $\mathbf{K} = 2\boldsymbol{\kappa}$. We can carry over Eqs. (m31 3)-(mmc2 8) simply by replacing the lowercase symbols $(\omega, \tilde{\omega}, \boldsymbol{\kappa}, n_\beta, w_\beta, \hat{\mathbf{p}}_{\beta\pm}, \hat{\mathbf{s}})$ with uppercase symbols $(\Omega, \tilde{\Omega}, \mathbf{K}, N_\beta, W_\beta, \hat{\mathbf{P}}_{\beta\pm}, \hat{\mathbf{S}})$, all evaluated at 2ω and we always have $\hat{\mathbf{S}} = \hat{\mathbf{s}}$.

To describe the propagation of the SH field, we see from Fig. 3layer 1, that it is refracted at the layer-vacuum interface (ℓv), and multiply reflected from the layer-bulk (ℓb) and layer-vacuum (ℓv)

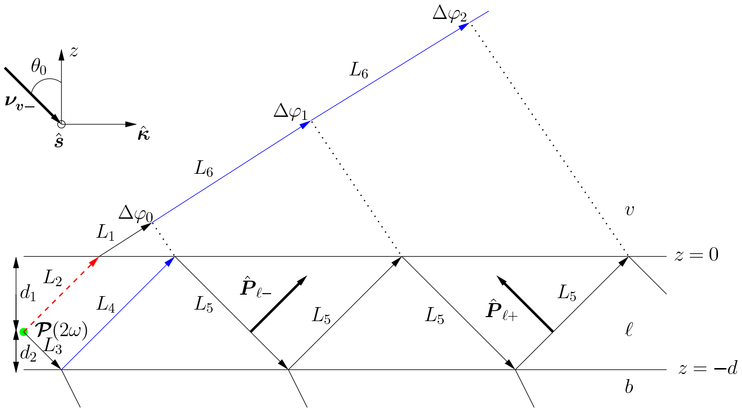

FIG. 1: (color on line) Sketch of the three layer model for SHG. Vacuum (v) is on top with $\epsilon_v = 1$; the layer ℓ, of thickness $d = d_1 + d_2$, is characterized with $\epsilon_\ell(\omega)$, and it is where the SH polarization sheet $\boldsymbol{\mathcal{P}}(2\omega)$ is located at $z_\ell = d_1$; The bulk b is described with $\epsilon_b(\omega)$. The arrows point along the direction of propagation, and the p-polarization unit vector, $\hat{\mathbf{P}}_{\ell-(+)}$, along the downward (upward) direction is denoted with a thick arrow. The s-polarization unit vector $\hat{\mathbf{s}}$, points out of the page. The fundamental field $\mathbf{E}(\omega)$ is incident from the vacuum side along the $z\hat{\boldsymbol{\kappa}}$-plane, with θ_0 its angle of incidence and $\boldsymbol{\nu}_{v-}$ its wave vector. $\Delta\varphi_i$ denote the phase difference of the multiply reflected beams with respect to the first vacuum transmitted beam (dashed-red arrow), where the dotted lines are perpendicular to this beam (see the text for details). 3layer

interfaces, thus we can define,

$$\mathbf{T}^{\ell v} = \hat{\mathbf{s}} T_s^{\ell v} \hat{\mathbf{s}} + \hat{\mathbf{P}}_{v+} T_p^{\ell v} \hat{\mathbf{P}}_{\ell+}, \tag{10}$$ r5

as the tensor for transmission from ℓv interface,

$$\mathbf{R}^{\ell b} = \hat{\mathbf{s}} R_s^{\ell b} \hat{\mathbf{s}} + \hat{\mathbf{P}}_{\ell+} R_p^{\ell b} \hat{\mathbf{P}}_{\ell-}, \tag{11}$$ r6

as the tensor of reflection from the ℓb interface, and

$$\mathbf{R}^{\ell v} = \hat{\mathbf{s}} R_s^{\ell v} \hat{\mathbf{s}} + \hat{\mathbf{P}}_{\ell-} R_p^{\ell v} \hat{\mathbf{P}}_{\ell+}, \tag{12}$$ r6b

as that of the ℓv interface. The Fresnel factors in uppercase letters, $T_{s,p}^{ij}$ and $R_{s,p}^{ij}$, are evaluated at 2ω from the following well known formulas

$$\begin{aligned} t_s^{ij}(\omega) &= \frac{2k_i(\omega)}{k_i(\omega)+k_j(\omega)}, & t_p^{ij}(\omega) &= \frac{2k_i(\omega)\sqrt{\epsilon_i(\omega)\epsilon_j(\omega)}}{k_i(\omega)\epsilon_j(\omega)+k_j(\omega)\epsilon_i(\omega)}, \\ r_s^{ij}(\omega) &= \frac{k_i(\omega)-k_j(\omega)}{k_i(\omega)+k_j(\omega)}, & r_p^{ij}(\omega) &= \frac{k_i(\omega)\epsilon_j(\omega)-k_j\epsilon_i(\omega)}{k_i(\omega)\epsilon_j(\omega)+k_j(\omega)\epsilon_i(\omega)}. \end{aligned} \tag{13}$$ e.f1

A. Multiple SH reflections

The SH field $\mathbf{E}(2\omega)$ radiated by the SH polarization $\boldsymbol{\mathcal{P}}(2\omega)$ will radiate directly into vacuum and also into the bulk, where it will be reflected back at the thin-layer-bulk interface into the thin layer again and this beam will be multiple-transmitted and reflected as shown in Fig. 1. As the two beams propagate a phase difference will develop between them, according to

$$\begin{aligned}\Delta\varphi_m &= \tilde{\Omega}\Big((L_3+L_4+2mL_5)n_\ell(2\omega) - \big(L_2 n_\ell(2\omega) + (L_1+mL_6)n_v(2\omega)\big)\Big)\\ &= \delta_0 + m\delta \quad m=0,1,2,\ldots,\end{aligned} \tag{14}$$

where

$$\delta_0 = 8\pi\left(\frac{d_2}{\lambda_0}\right)\sqrt{n_\ell^2(2\omega)-\sin^2\theta_0}, \tag{15}$$

$$\delta = 8\pi\left(\frac{d}{\lambda_0}\right)\sqrt{n_\ell^2(2\omega)-\sin^2\theta_0}, \tag{16}$$

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

To take into account the multiple reflections of the generated SH field in the layer ℓ, we proceed as follows. We show the algebra for the p-polarized SH field, the s-polarized field could be worked out along the same steps. The multiple-reflected $\mathbf{E}_p(2\omega)$ field is given by

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

From Eqs. (10) and (11) is easy to show that

$$\mathbf{T}^{\ell v}\cdot\Big(\mathbf{R}^{\ell b}\cdot\mathbf{R}^{\ell v}\Big)^n\cdot\mathbf{R}^{\ell b} = \hat{\mathbf{s}}T_s^{\ell v}\Big(R_s^{\ell b}R_s^{\ell v}\Big)^n R_s^{\ell b}\hat{\mathbf{s}} + \hat{\mathbf{P}}_{v+}T_p^{\ell v}\Big(R_p^{\ell b}R_p^{\ell v}\Big)^n R_p^{\ell b}\hat{\mathbf{P}}_{\ell-}, \tag{17}$$

then,

$$\mathbf{E}(2\omega) = \hat{\mathbf{P}}_{\ell+}T_p^{\ell v}\Big(E_{p+}(2\omega) + \frac{R_p^{\ell b}e^{i\delta_0}}{1+R_p^{v\ell}R_p^{\ell b}e^{i\delta}}E_{p-}(2\omega)\Big), \tag{18}$$

where we used $R_{s,p}^{ij} = -R_{s,p}^{ji}$. Using Eq. (4), we can readily write

$$\mathbf{E}(2\omega) = \frac{2\pi i\tilde{\Omega}}{K_\ell}\mathbf{H}_\ell\cdot\boldsymbol{\mathcal{P}}(2\omega), \tag{19}$$

where

$$\mathbf{H}_\ell = \hat{\mathbf{s}}\,T_s^{\ell v}\left(1+R_s^M\right)\hat{\mathbf{s}} + \hat{\mathbf{P}}_{v+}T_p^{\ell v}\left(\hat{\mathbf{P}}_{\ell+}+R_p^M\hat{\mathbf{P}}_{\ell-}\right). \tag{20}$$

and

$$R_l^M \equiv \frac{R_l^{\ell b} e^{i\delta_0}}{1 + R_l^{v\ell} R_l^{\ell b} e^{i\delta}} \quad l = s, p, \tag{21}$$ m61

is defined as the multiple reflection coefficient. To make touch with the work of Ref. mizrahiJOSA88 ? where $\boldsymbol{\mathcal{P}}(2\omega)$ is located on top of the vacuum-surface interface and only the vacuum radiated beam and the first (and only) reflected beam need to be considered, we take $\ell = v$ and $d_2 = 0$, then $T^{\ell v} = 1$, $R^{v\ell} = 0$ and $\delta_0 = 0$, with which $R_l^M = R_l^{vb}$. Thus, Eq. (mr9 20) coincides with Eq. (3.8) of Ref. mizrahiJOSA88 ? .

B. Radiation Terms

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

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

where using

$$\begin{aligned}
1 + R_s^{\ell b} &= T_s^{\ell b} \\
1 + R_p^{\ell b} &= \sqrt{\frac{\epsilon_b(2\omega)}{\epsilon_\ell(2\omega)}}T_p^{\ell b} \\
1 - R_p^{\ell b} &= \sqrt{\frac{\epsilon_\ell(2\omega)}{\epsilon_b(2\omega)}}\frac{K_b}{K_\ell}T_p^{\ell b} \\
T_p^{\ell v} &= \frac{K_\ell}{K_v}T_p^{v\ell} \\
T_s^{\ell v} &= \frac{K_\ell}{K_v}T_s^{v\ell},
\end{aligned} \tag{22}$$

we can write

$$E(2\omega) = \frac{4\pi i\omega}{cK_v}\hat{\mathbf{e}}^{\text{out}} \cdot \mathbf{H}_\ell \cdot \boldsymbol{\mathcal{P}}(2\omega) = \frac{4\pi i\omega}{cK_v}\mathbf{e}_\ell^{2\omega} \cdot \boldsymbol{\mathcal{P}}(2\omega). \tag{23}$$ r10

where,

$$\begin{aligned}
&\mathbf{e}_\ell^{2\omega} = \\
&\hat{\mathbf{e}}^{\text{out}} \cdot \left[\hat{\mathbf{s}}T_s^{v\ell}T_s^{\ell b}\hat{\mathbf{s}} + \hat{\mathbf{P}}_{v+}\frac{T_p^{v\ell}T_p^{\ell b}}{\epsilon_\ell(2\omega)\sqrt{\epsilon_b(2\omega)}}\left(\epsilon_b(2\omega)\sin\theta_{\text{in}}\hat{\mathbf{z}} - \epsilon_\ell(2\omega)K_b\hat{\mathbf{x}}\right)\right].
\end{aligned} \tag{24}$$ r12

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 $\ell = v$ ($T^{\ell v}_{s,p} = 1$), to get

$$\mathbf{e}^{2\omega}_v = \hat{\mathbf{e}}^{\rm out} \cdot \left[\hat{\mathbf{s}} T^{vb}_s \hat{\mathbf{s}} + \hat{\mathbf{P}}_{v+} \frac{T^{vb}_p}{\sqrt{\epsilon_b(2\omega)}} \left(\epsilon_b(2\omega) \sin\theta_{\rm in} \hat{\mathbf{z}} - K_b \hat{\mathbf{x}} \right) \right], \qquad (25) \quad \boxed{\texttt{r13}}$$

which agrees with Eq. (3.8) of Ref. mizrahiJOSA88 [?].

In the three layer model the nonlinear polarization is located in layer ℓ, and then we evaluate the fundamental field required in Eq. (tres 9) in this layer as well, then we write

$$\mathbf{E}_\ell(\omega) = E_0 \left(\hat{\mathbf{s}} t^{v\ell}_s (1 + r^{\ell b}_s) \hat{\mathbf{s}} + \hat{\mathbf{p}}_{\ell -} t^{v\ell}_p \hat{\mathbf{p}}_{v-} + \hat{\mathbf{p}}_{\ell +} t^{v\ell}_p r^{\ell b}_p \hat{\mathbf{p}}_{v-} \right) \cdot \hat{\mathbf{e}}^{\rm in} = E_0 \mathbf{e}^\omega_\ell, \qquad (26) \quad \boxed{\texttt{m2}}$$

and following the steps that lead to Eq. (r12 24), we find that

$$\mathbf{e}^\omega_\ell = \left[\hat{\mathbf{s}} t^{v\ell}_s t^{\ell b}_s \hat{\mathbf{s}} + \frac{t^{v\ell}_p t^{\ell b}_p}{\epsilon_\ell(\omega) \sqrt{\epsilon_b(\omega)}} \left(\epsilon_b(\omega) \sin\theta_{\rm in} \hat{\mathbf{z}} + \epsilon_\ell(\omega) k_b \hat{\mathbf{x}} \right) \hat{\mathbf{p}}_{v-} \right] \cdot \hat{\mathbf{e}}^{\rm in}. \qquad (27) \quad \boxed{\texttt{m12}}$$

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

$$\mathbf{e}^\omega_b = \left[\hat{\mathbf{s}} t^{vb}_s \hat{\mathbf{s}} + \frac{t^{vb}_p}{\sqrt{\epsilon_b(\omega)}} \left(\sin\theta_{\rm in} \hat{\mathbf{z}} + k_b \hat{\mathbf{x}} \right) \hat{\mathbf{p}}_{v-} \right] \cdot \hat{\mathbf{e}}^{\rm in}, \qquad (28) \quad \boxed{\texttt{m13}}$$

that is in agreement with Eq. (3.5) of Ref. mizrahiJOSA88 [?].

With $\mathbf{e}^\omega$ we can write Eq. (tres 9) as

$$\boldsymbol{\mathcal{P}}(2\omega) = E_0^2 \boldsymbol{\chi} : \mathbf{e}^\omega_\ell \mathbf{e}^\omega_\ell, \qquad (29) \quad \boxed{\texttt{m4}}$$

and then from Eq. (r10 23) we obtain that

$$\begin{aligned}
|E(2\omega)|^2 &= |E_0|^4 \frac{16\pi^2\omega^2}{c^2 K_v^2} \left| \mathbf{e}^{2\omega}_\ell \cdot \boldsymbol{\chi} : \mathbf{e}^\omega_\ell \mathbf{e}^\omega_\ell \right|^2 \\
\frac{c}{2\pi} |E(2\omega)|^2 &= \frac{32\pi^3\omega^2}{c^3 \cos^2\theta_{\rm in}} \left| \mathbf{e}^{2\omega}_\ell \cdot \boldsymbol{\chi} : \mathbf{e}^\omega_\ell \mathbf{e}^\omega_\ell \right|^2 \left(\frac{c}{2\pi} |E_0|^2 \right)^2, \\
I(2\omega) &= \frac{32\pi^3\omega^2}{c^3 \cos^2\theta_{\rm in}} \left| \mathbf{e}^{2\omega}_\ell \cdot \boldsymbol{\chi} : \mathbf{e}^\omega_\ell \mathbf{e}^\omega_\ell \right|^2 I^2(\omega), \\
R(2\omega) &= \frac{32\pi^3\omega^2}{c^3 \cos^2\theta_{\rm in}} \left| \mathbf{e}^{2\omega}_\ell \cdot \boldsymbol{\chi} : \mathbf{e}^\omega_\ell \mathbf{e}^\omega_\ell \right|^2, \qquad (30) \quad \boxed{\texttt{r01}}
\end{aligned}$$

as the SHG yield. At this point we mention that to recover the results of Ref. mizrahiJOSA88 [?] which are equivalent of those of Ref. sipePRB87 [?], we take $\mathbf{e}^{2\omega}_\ell \to \mathbf{e}^{2\omega}_v$, $\mathbf{e}^\omega_\ell \to \mathbf{e}^\omega_b$ and then

$$R(2\omega) = \frac{32\pi^3\omega^2}{c^3 \cos^2\theta_{\rm in}} \left| \mathbf{e}^{2\omega}_v \cdot \boldsymbol{\chi} : \mathbf{e}^\omega_b \mathbf{e}^\omega_b \right|^2, \qquad (31) \quad \boxed{\texttt{m69}}$$

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 $\epsilon_b(\omega)$.

C. One SH Reflection

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

$$\mathbf{E}(2\omega) = \frac{2\pi i\tilde{\Omega}}{K_\ell}\mathbf{H}_\ell\cdot\boldsymbol{\mathcal{P}}(2\omega), \tag{32}$$ r8

where,

$$\mathbf{H}_\ell = \hat{\mathbf{s}}\,T_s^{\ell v}\left(1 + R_s^{\ell b}\right)\hat{\mathbf{s}} + \hat{\mathbf{P}}_{v+}T_p^{\ell v}\left(\hat{\mathbf{P}}_{\ell +} + R_p^{\ell b}\hat{\mathbf{P}}_{\ell -}\right). \tag{33}$$ r9

II. $\mathcal{R}$ 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

$$\hat{\boldsymbol{\kappa}} = \cos\phi\hat{\mathbf{x}} + \sin\phi\hat{\mathbf{y}}, \tag{34}$$ mc1

and then

$$\hat{\mathbf{s}} = -\sin\phi\hat{\mathbf{x}} + \cos\phi\hat{\mathbf{y}}, \tag{35}$$ mc2

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

We develop five different scenarios for $\mathcal{R}_{pP}$ that explore different cases for where the polarization and fundamental fields are located. In all these scenarios, we use $\hat{\mathbf{e}}^{\mathrm{in}} = \hat{\mathbf{p}}_{v-}$ in Eq. (m12 27), and $\hat{\mathbf{e}}^{\mathrm{out}} = \hat{\mathbf{P}}_{v+}$ in Eq. (r12 24).

1. *Three layer model*

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,

$$\mathbf{e}_\ell^{2\omega}\cdot\boldsymbol{\chi}:\mathbf{e}_\ell^{\omega}\mathbf{e}_\ell^{\omega}\equiv\Gamma_{pP}^{\ell}\,r_{pP}^{\ell},\tag{36}$$ m80

where

$$\begin{aligned}r_{pP}^{\ell}&=\epsilon_b(2\omega)\sin\theta_{\rm in}\Big(\epsilon_b^2(\omega)\sin^2\theta_{\rm in}\chi_{zzz}+\epsilon_\ell^2(\omega)k_b^2\chi_{zxx}\Big)\\&-\epsilon_\ell(2\omega)\epsilon_\ell(\omega)k_bK_b\Big(2\epsilon_b(\omega)\sin\theta_{\rm in}\chi_{xxz}+\epsilon_\ell(\omega)k_b\chi_{xxx}\cos(3\phi)\Big),\end{aligned}\tag{37}$$ m81

and

$$\Gamma_{pP}^{\ell}=\frac{T_p^{\ell v}T_p^{\ell b}}{\epsilon_\ell(2\omega)\sqrt{\epsilon_b(2\omega)}}\left(\frac{t_p^{v\ell}t_p^{\ell b}}{\epsilon_\ell(\omega)\sqrt{\epsilon_b(\omega)}}\right)^2.\tag{38}$$ m79

2. *Two layer model*

In order to reduce above result to that of Ref. mizrahiJOSA88 [?] and sipePRB87 [?], 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

$$\mathbf{e}_v^{2\omega}\cdot\boldsymbol{\chi}:\mathbf{e}_b^{\omega}\mathbf{e}_b^{\omega}\equiv\Gamma_{pP}^{vb}\,r_{pP}^{vb},\tag{39}$$ m800

where,

$$\begin{aligned}r_{pP}^{vb}&=\epsilon_b(2\omega)\sin\theta_{\rm in}\Big(\sin^2\theta_{\rm in}\chi_{zzz}+k_b^2 chi_{zxx}\Big)\\&-k_bK_b\Big(2\sin\theta_{\rm in}\chi_{xxz}+k_b\chi_{xxx}\cos(3\phi)\Big),\end{aligned}\tag{40}$$ m82

and

$$\Gamma_{pP}^{vb}=\frac{T_p^{vb}(t_p^{vb})^2}{\epsilon_b(\omega)\sqrt{\epsilon_b(2\omega)}}.\tag{41}$$ m78

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

To evaluate the 2ω fields in the bulk, we take Eq. (r9 33) considering that $\ell\to b$. We have already considered the 1ω fields in the bulk in Eq. (m13 28). After some algebra, we get that

$$\mathbf{e}_b^{2\omega}\cdot\boldsymbol{\chi}:\mathbf{e}_b^{\omega}\mathbf{e}_b^{\omega}=\Gamma_{pP}^{b}r_{pP}^{b}\tag{42}$$

where

$$r^b_{pP} = \sin^3\theta_{\rm in}\chi_{zzz} + k_b^2\sin\theta_{\rm in}\chi_{zxx} - 2k_bK_b\sin\theta_{\rm in}\chi_{xxz} - k_b^2K_b\chi_{xxx}\cos 3\phi, \tag{43}$$

and

$$\Gamma^b_{pP} = \frac{T_p^{vb}\left(t_p^{vb}\right)^2}{\epsilon_b(\omega)\sqrt{\epsilon_b(2\omega)}}. \tag{44}$$

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

To evaluate the 1ω fields in the vacuum, we take Eq. (26) considering that $\ell \to v$. We have already considered the 2ω fields in the vacuum in Eq. (25). After some algebra, we get that

$$\mathbf{e}_v^{2\omega}\cdot\boldsymbol{\chi}:\mathbf{e}_v^{\omega}\mathbf{e}_v^{\omega} = \Gamma^v_{pP}r^v_{pP} \tag{45}$$

where

$$\begin{aligned} r^v_{pP} = {} & \epsilon_b^2(\omega)\epsilon_b(2\omega)\sin^3\theta_{\rm in}\chi_{zzz} + \epsilon_b(2\omega)k_b^2\sin\theta_{\rm in}\chi_{zxx} \\ & - 2\epsilon_b(\omega)k_bK_b\sin\theta_{\rm in}\chi_{xxz} - k_b^2K_b\chi_{xxx}\cos 3\phi \end{aligned} \tag{46}$$

and

$$\Gamma^v_{pP} = \frac{T_p^{vb}\left(t_p^{vb}\right)^2}{\epsilon_b(\omega)\sqrt{\epsilon_b(2\omega)}}. \tag{47}$$

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

For this scenario, we have

$$\mathbf{e}_\ell^{2\omega}\cdot\boldsymbol{\chi}:\mathbf{e}_b^{\omega}\mathbf{e}_b^{\omega} = \Gamma^{\ell b}_{pP}r^{\ell b}_{pP} \tag{48}$$

where

$$\begin{aligned} r^{\ell b}_{pP} = {} & \epsilon_b(2\omega)\sin^3\theta_{\rm in}\chi_{zzz} + \epsilon_b(2\omega)k_b^2\sin\theta_{\rm in}\chi_{zxx} \\ & - 2\epsilon_\ell(2\omega)k_bK_b\sin\theta_{\rm in}\chi_{xxz} - \epsilon_\ell(2\omega)k_b^2K_b\chi_{xxx}\cos 3\phi, \end{aligned} \tag{49}$$

and

$$\Gamma^{\ell b}_{pP} = \frac{T_p^{v\ell}T_p^{\ell b}\left(t_p^{vb}\right)^2}{\epsilon_\ell(2\omega)\epsilon_b(\omega)\sqrt{\epsilon_b(2\omega)}}. \tag{50}$$

B. $\mathcal{R}_{pS}$

To obtain $R_{pS}(2\omega)$ we use $\hat{\mathbf{e}}^{\text{in}} = \hat{\mathbf{p}}_{v-}$ in Eq. (27), and $\hat{\mathbf{e}}^{\text{out}} = \hat{\mathbf{S}}$ in Eq. (24). We also use the unit vectors defined in Eqs. (34) and (35). Substituting, we get

$$\mathbf{e}_\ell^{2\omega} \cdot \boldsymbol{\chi} : \mathbf{e}_\ell^{\omega}\mathbf{e}_\ell^{\omega} \equiv \Gamma_{sP}^{\ell}\, r_{sP}^{\ell}, \tag{51}$$

where

$$r_{pS}^{\ell} = -\epsilon_\ell^2(\omega) k_b^2 \sin 3\phi \chi_{xxx}, \tag{52}$$

and

$$\Gamma_{pS}^{\ell} = T_s^{v\ell} T_s^{\ell b} \left(\frac{t_p^{v\ell} t_p^{\ell b}}{\epsilon_\ell(\omega)\sqrt{\epsilon_b(\omega)}} \right)^2. \tag{53}$$

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

$$r_{pS}^{b} = -k_b^2 \sin 3\phi \chi_{xxx}, \tag{54}$$

and

$$\Gamma_{pS}^{b} = T_s^{vb} \left(\frac{t_p^{vb}}{\sqrt{\epsilon_b(\omega)}} \right)^2. \tag{55}$$

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

To obtain $R_{sP}(2\omega)$ we use $\hat{\mathbf{e}}^{\text{in}} = \hat{\mathbf{s}}$ in Eq. (27), and $\hat{\mathbf{e}}^{\text{out}} = \hat{\mathbf{P}}_{v+}$ in Eq. (24). We also use the unit vectors defined in Eqs. (34) and (35). Substituting, we get

$$\mathbf{e}_\ell^{2\omega} \cdot \boldsymbol{\chi} : \mathbf{e}_\ell^{\omega}\mathbf{e}_\ell^{\omega} \equiv \Gamma_{sP}^{\ell}\, r_{sP}^{\ell}, \tag{56}$$

where

$$r_{sP}^{\ell} = \epsilon_b(2\omega) \sin\theta_{\text{in}} \chi_{zxx} + \epsilon_\ell(2\omega) K_b \chi_{xxx} \cos 3\phi, \tag{57}$$

and

$$\Gamma_{sP}^{\ell} = \frac{T_p^{\ell v} T_p^{\ell b} \left(t_s^{v\ell} t_s^{\ell b}\right)^2}{\epsilon_\ell(2\omega)\sqrt{\epsilon_b(2\omega)}}. \tag{58}$$

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

$$r^b_{sP} = \epsilon_b(2\omega)\sin\theta_{\rm in}\chi_{zxx} + K_b\chi_{xxx}\cos 3\phi, \tag{59}$$

and

$$\Gamma^b_{sP} = \frac{T_p^{vb}(t_s^{vb})^2}{\sqrt{\epsilon_b(2\omega)}}. \tag{60}$$

D. $\mathcal{R}_{sS}$

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

$$\mathbf{e}^{2\omega}_\ell \cdot \boldsymbol{\chi} : \mathbf{e}^{\omega}_\ell \mathbf{e}^{\omega}_\ell \equiv \Gamma^\ell_{sS}\, r^\ell_{sS}, \tag{61}$$

where

$$r^\ell_{sS} = \chi_{xxx}\sin 3\phi, \tag{62}$$

and

$$\Gamma^\ell_{sS} = T_s^{v\ell} T_s^{\ell b}\left(t_s^{v\ell} t_s^{\ell b}\right)^2. \tag{63}$$

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

$$r^b_{sS} = \chi_{xxx}\sin 3\phi, \tag{64}$$

and

$$\Gamma^b_{sS} = T_s^{vb}\left(t_s^{vb}\right)^2. \tag{65}$$

Appendix A: Full derivations for $\mathcal{R}$ for different polarization cases

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

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

To consider the 2ω fields in the bulk, we start with Eq. (33) 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

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

and we define

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

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

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

The 1ω fields will still be evaluated inside the bulk, so we have Eq. (28)

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

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

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

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 $\boldsymbol{\chi}$,

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

and reducing,

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

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

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

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

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

To consider the 1ω fields in the vacuum, we start with Eq. (26) but substitute $\ell \to v$, thus

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

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

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

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. (25),

$$
\mathbf{e}_v^{2\omega} = \hat{\mathbf{e}}^{\mathrm{out}}\cdot\left[\hat{\mathbf{s}}T_s^{vb}\hat{\mathbf{s}} + \hat{\mathbf{P}}_{v+}\frac{T_p^{vb}}{\sqrt{\epsilon_b(2\omega)}}\left(\epsilon_b(2\omega)\sin\theta_{\mathrm{in}}\hat{\mathbf{z}} - K_b\hat{\boldsymbol{\kappa}}\right)\right], \tag{A1}
$$

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

$$
\mathbf{e}_v^{2\omega} = \frac{T_p^{vb}}{\sqrt{\epsilon_b(2\omega)}}\left(\epsilon_b(2\omega)\sin\theta_{\mathrm{in}}\hat{\mathbf{z}} - K_b\cos\phi\hat{\mathbf{x}} - K_b\sin\phi\hat{\mathbf{y}}\right). \tag{A2}
$$

So lastly, we have that

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

and after eliminating components,

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

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

where

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

c. 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. (24),

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

and Eq. (28),

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

Thus,

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

We eliminate and replace components,

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

so lastly

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

where

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

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

To obtain $R_{pS}(2\omega)$ we use $\hat{\mathbf{e}}^{\mathrm{in}} = \hat{\mathbf{p}}_{v-}$ in Eq. (27), and $\hat{\mathbf{e}}^{\mathrm{out}} = \hat{\mathbf{S}}$ in Eq. (24). We also use the unit vectors defined in Eqs. (34) and (35). Substituting, we get

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

for 2ω, and for the fundamental fields,

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

Therefore,

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

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

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

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

$$=\Gamma_{pS}^\ell\big[\epsilon_\ell^2(\omega)k_b^2(\sin^3\phi-3\cos^2\phi\sin\phi)\chi_{xxx}\big]$$

$$=\Gamma_{pS}^\ell\big[-\epsilon_\ell^2(\omega)k_b^2\sin 3\phi\chi_{xxx}\big].$$

We summarize as follows,

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

where

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

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. [?] and [?], 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 $\epsilon_\ell(\omega) = \epsilon_b(\omega)$, $t^{v\ell}_p = t^{vb}_p$, and $t^{\ell b}_p = 1$. With these choices,

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

and

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

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

To obtain $R_{sP}(2\omega)$ we use $\hat{\mathbf{e}}^{\rm in} = \hat{\mathbf{s}}$ in Eq. (27), and $\hat{\mathbf{e}}^{\rm out} = \hat{\mathbf{P}}_{v+}$ in Eq. (24). We also use the unit vectors defined in Eqs. (34) and (35). Substituting, we get

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

for 2ω, and for the fundamental fields,

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

Therefore,

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

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

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

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

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

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

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

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

and

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

In order to reduce above result to that of Ref. mizrahiJOSA88 [?] and sipePRB87 [?], we take the 2-ω radiations factors for vacuum by taking $\ell = v$, thus $\epsilon_\ell(2\omega) = 1$, $T^{v\ell}_p = 1$, $T^{\ell b}_p = T^{vb}_p$, and the fundamental field inside medium b by taking $\ell = b$, thus $\epsilon_\ell(\omega) = \epsilon_b(\omega)$, $t^{v\ell}_s = t^{vb}_s$, and $t^{\ell b}_s = 1$. With these choices,

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

and

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

4. $\mathcal{R}_{sS}$

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

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

Therefore,

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

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

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

and

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

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

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