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

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January 5, 2016

## 1 Three layer model for SHG radiation

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

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

with the intensity

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

There are several ways to calculate $R$, one of which is the procedure followed by Cini [1]. This approach calculates the nonlinear susceptibility and at the same time the radiated fields. However, we present an alternative derivation based in the work of Mizrahi and Sipe [2], since the derivation of the three-layer-model is straightforward. Within our level of approximation this is the best model that we can use. In this scheme, we assume that the SH conversion takes place in a thin layer, just below the surface, that is characterized by a surface dielectric function $\epsilon_\ell(\omega)$. This layer is below vacuum and sits on top of the bulk characterized by $\epsilon_b(\omega)$ (see Fig. 1). 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,

$$\mathcal{P}_i(2\omega) = \chi_{ijk}E_j(\omega)E_k(\omega), \tag{1}$$

is given by [2],

$$(E_{p\pm}, E_s) = \left(\frac{2\pi i\tilde{\omega}^2}{w}\,\hat{\mathbf{p}}_\pm \cdot \boldsymbol{\mathcal{P}}, \frac{2\pi i\tilde{\omega}^2}{w}\,\hat{\mathbf{s}} \cdot \boldsymbol{\mathcal{P}}\right),$$
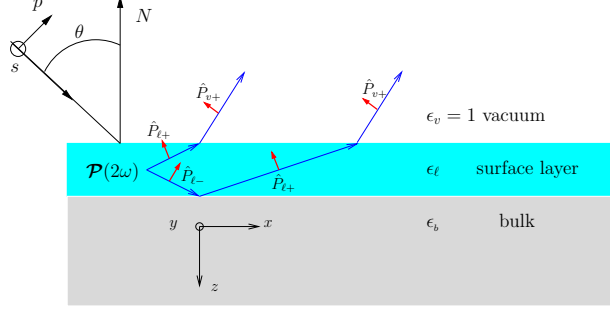
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Figure 1: Sketch of the three layer model for SHG. Vacuum is on top with $\epsilon = 1$, the layer with nonlinear polarization $\mathbf{P}(2\omega)$ ischaracterized with $\epsilon_\ell(\omega)$ and the bulk with $\epsilon_b(\omega)$. In the dipolar 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).

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

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

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

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

In the above equations $z$ is the direction perpendicular to the surface that points towards the vacuum, $x$ is parallel to the surface, and $\theta$ 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 perpendicular to the surface. As we see from Fig. 1, the SH field is refracted at the layer-vacuum interface $(\ell v)$, and reflected from the layer-bulk $(\ell b)$ interface, thus we can define the transmission, $\mathbf{T}$, and reflection, $\mathbf{R}$, tensors as,

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

and

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

where variables in capital letters are evaluated at the harmonic frequency $2\omega$. Notice that since $\hat{\mathbf{s}}$ is independent of $\omega$, 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 $\ell v$ or $\ell b$, 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 $(v)$, $\epsilon_\ell$ for the layer $(\ell)$, and $\epsilon_b$ for the bulk $(b)$. Therefore, the total radiated field at $2\omega$ is

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

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

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

where,

$$\mathbf{H}_\ell = \hat{\mathbf{s}}\,T_s^{\ell v}\left(1 + R_s^{\ell b}\right)\hat{\mathbf{s}} + \hat{\mathbf{P}}_{v+}T_p^{\ell v}\left(\hat{\mathbf{P}}_{\ell+} + R_p^{\ell b}\hat{\mathbf{P}}_{\ell-}\right). \tag{2}$$

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

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

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

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

where using

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

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

$$1 - R_p^{\ell b} = \sqrt{\frac{\epsilon_\ell(2\omega)}{\epsilon_b(2\omega)}}\frac{K_b}{K_\ell}T_p^{\ell b} \tag{3}$$

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

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

we can write

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

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where,

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

$$\hat{\mathbf{e}}^{\text{out}} \cdot \left[ \hat{\mathbf{s}} T_s^{v\ell} T_s^{\ell b} \hat{\mathbf{s}} + \hat{\mathbf{P}}_{v+} \frac{T_p^{v\ell} T_p^{\ell b}}{\epsilon_\ell(2\omega)\sqrt{\epsilon_b(2\omega)}} \left( \epsilon_b(2\omega)\sin\theta_{\text{in}}\hat{\mathbf{z}} - \epsilon_\ell(2\omega)K_b\hat{\mathbf{x}} \right) \right]. \quad (4)$$

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

$$\mathbf{e}_v^{2\omega} = \hat{\mathbf{e}}^{\text{out}} \cdot \left[ \hat{\mathbf{s}} T_s^{vb} \hat{\mathbf{s}} + \hat{\mathbf{P}}_{v+} \frac{T_p^{vb}}{\sqrt{\epsilon_b(2\omega)}} \left( \epsilon_b(2\omega)\sin\theta_{\text{in}}\hat{\mathbf{z}} - K_b\hat{\mathbf{x}} \right) \right], \quad (5)$$

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

In the three layer model the nonlinear polarization is located in layer $\ell$, and then we evaluate the fundamental field required in Eq. (1) in this layer as well, then we write

$$\mathbf{E}_\ell(\omega) = E_0 \left( \hat{\mathbf{s}} t_s^{v\ell}(1 + r_s^{\ell b})\hat{\mathbf{s}} + \hat{\mathbf{p}}_{\ell-} t_p^{v\ell}\hat{\mathbf{p}}_{v-} + \hat{\mathbf{p}}_{\ell+} t_p^{v\ell} r_p^{\ell b}\hat{\mathbf{p}}_{v-} \right) \cdot \hat{\mathbf{e}}^{\text{in}} = E_0 \mathbf{e}_\ell^\omega, \quad (6)$$

and following the steps that lead to Eq. (4), we find that

$$\mathbf{e}_\ell^\omega = \left[ \hat{\mathbf{s}} t_s^{v\ell} t_s^{\ell b} \hat{\mathbf{s}} + \frac{t_p^{v\ell} t_p^{\ell b}}{\epsilon_\ell(\omega)\sqrt{\epsilon_b(\omega)}} \left( \epsilon_b(\omega)\sin\theta_{\text{in}}\hat{\mathbf{z}} + \epsilon_\ell(\omega)k_b\hat{\mathbf{x}} \right) \hat{\mathbf{p}}_{v-} \right] \cdot \hat{\mathbf{e}}^{\text{in}}. \quad (7)$$

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

$$\mathbf{e}_b^\omega = \left[ \hat{\mathbf{s}} t_s^{vb} \hat{\mathbf{s}} + \frac{t_p^{vb}}{\sqrt{\epsilon_b(\omega)}} \left( \sin\theta_{\text{in}}\hat{\mathbf{z}} + k_b\hat{\mathbf{x}} \right) \hat{\mathbf{p}}_{v-} \right] \cdot \hat{\mathbf{e}}^{\text{in}}, \quad (8)$$

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

With $\mathbf{e}^\omega$ we can write Eq. (1) as

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

and then from Eq. (1) we obtain that

$$|E(2\omega)|^2 = |E_0|^4 \frac{16\pi^2\omega^2}{c^2 K_v^2} \left| \mathbf{e}_\ell^{2\omega} \cdot \boldsymbol{\chi} : \mathbf{e}_\ell^\omega \mathbf{e}_\ell^\omega \right|^2$$

$$\frac{c}{2\pi}|E(2\omega)|^2 = \frac{32\pi^3\omega^2}{c^3 \cos^2\theta_{\text{in}}} \left| \mathbf{e}_\ell^{2\omega} \cdot \boldsymbol{\chi} : \mathbf{e}_\ell^\omega \mathbf{e}_\ell^\omega \right|^2 \left( \frac{c}{2\pi}|E_0|^2 \right)^2,$$

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

$$R(2\omega) = \frac{32\pi^3\omega^2}{c^3 \cos^2\theta_{\text{in}}} \left| \mathbf{e}_\ell^{2\omega} \cdot \boldsymbol{\chi} : \mathbf{e}_\ell^\omega \mathbf{e}_\ell^\omega \right|^2, \quad (9)$$

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

$$R(2\omega) = \frac{32\pi^3\omega^2}{c^3\cos^2\theta_{\rm in}} \left| \mathbf{e}_v^{2\omega} \cdot \boldsymbol{\chi} : \mathbf{e}_b^\omega \mathbf{e}_b^\omega \right|^2, \tag{10}$$

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

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

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

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

# 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 $\chi_{ijk}$ different from zero are $\chi_{zzz}$, $\chi_{zxx} = \chi_{zyy}$, $\chi_{xxz} = \chi_{yyz}$ 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 $\phi$ with respect to the $x$ axis, and so $\hat{\mathbf{x}}$ should to be replaced by a unit vector $\hat{\kappa}$ such that

$$\hat{\boldsymbol{\kappa}} = \cos\phi\hat{\mathbf{x}} + \sin\phi\hat{\mathbf{y}}, \tag{11}$$

and then

$$\hat{\mathbf{s}} = -\sin\phi\hat{\mathbf{x}} + \cos\phi\hat{\mathbf{y}}, \tag{12}$$

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

To obtain $R_{pP}(2\omega)$ we use $\mathbf{e}^{\rm in} = \hat{\mathbf{p}}_{v-}$ in Eq. (7), and $\mathbf{e}^{\rm out} = \hat{\mathbf{P}}_{v+}$ in Eq. (4), 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

$$r_{pP}^\ell = \epsilon_b(2\omega)\sin\theta_{\rm in}\left(\epsilon_b^2(\omega)\sin^2\theta_{\rm in}\chi_{zzz} + \epsilon_\ell^2(\omega)k_b^2\chi_{zxx}\right) \tag{13}$$

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

and

$$\Gamma_{pP}^\ell = \frac{T_p^{\ell v}T_p^{\ell b}}{\epsilon_\ell(2\omega)\sqrt{\epsilon_b(2\omega)}} \left(\frac{t_p^{v\ell}t_p^{\ell b}}{\epsilon_\ell(\omega)\sqrt{\epsilon_b(\omega)}}\right)^2. \tag{14}$$

In order to reduce above result to that of Ref. [2] and [3], we take the $2\omega$ 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,

$$r_{pP}^b = \epsilon_b(2\omega)\sin\theta_{\text{in}}\Big(\sin^2\theta_{\text{in}}\chi_{zzz} + k_b^2 chi_{zxx}\Big)$$
$$- k_b K_b\Big(2\sin\theta_{\text{in}}\chi_{xxz} + k_b\chi_{xxx}\cos(3\phi)\Big),$$

and

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

### 2.1.1 Taking all fields in the bulk

To evaluate the $2\omega$ fields in the bulk, we take Eq. (2) considering that $\ell \to b$. We have already considered the $1\omega$ fields in the bulk in Eq. (8). After some algebra, we get that

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

where

$$r_{pP}^b = \sin^3\theta_{\text{in}}\chi_{zzz} + k_b^2\sin\theta_{\text{in}}\chi_{zxx} - 2k_b K_b\sin\theta_{\text{in}}\chi_{xxz} - k_b^2 K_b\chi_{xxx}\cos 3\phi,$$

and

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

### 2.1.2 Taking all fields in the vacuum

To evaluate the $1\omega$ fields in the vacuum, we take Eq. (6) considering that $\ell \to v$. We have already considered the $2\omega$ fields in the vacuum in Eq. (5). After some algebra, we get that

$$\mathbf{e}_v^{2\omega} \cdot \boldsymbol{\chi} : \mathbf{e}_v^\omega \mathbf{e}_v^\omega = \Gamma_{pP}^v r_{pP}^v$$

where

$$r_{pP}^v = \epsilon_b^2(\omega)\epsilon_b(2\omega)\sin^3\theta_{\text{in}}\chi_{zzz} + \epsilon_b(2\omega)k_b^2\sin\theta_{\text{in}}\chi_{zxx}$$
$$- 2\epsilon_b(\omega)k_b K_b\sin\theta_{\text{in}}\chi_{xxz} - k_b^2 K_b\chi_{xxx}\cos 3\phi$$

and

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

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

To obtain $R_{pS}(2\omega)$ we use $\mathbf{e}^{\text{in}} = \hat{\mathbf{p}}_{v-}$ in Eq. (7), and $\mathbf{e}^{\text{out}} = \hat{\mathbf{S}}$ in Eq. (4). We also use the unit vectors defined in Eqs. (11) and (12). Substituting, we get

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

where

$$r_{pS}^\ell = -\epsilon_\ell^2(\omega)k_b^2 \sin 3\phi \chi_{xxx}, \tag{15}$$

and

$$\Gamma_{pS}^\ell = T_s^{v\ell}T_s^{\ell b}\left(\frac{t_p^{v\ell}t_p^{\ell b}}{\epsilon_\ell(\omega)\sqrt{\epsilon_b(\omega)}}\right)^2. \tag{16}$$

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

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

and

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

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

To obtain $R_{sP}(2\omega)$ we use $\mathbf{e}^{\text{in}} = \hat{\mathbf{s}}$ in Eq. (7), and $\mathbf{e}^{\text{out}} = \hat{\mathbf{P}}_{v+}$ in Eq. (4). We also use the unit vectors defined in Eqs. (11) and (12). Substituting, we get

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

where

$$r_{sP}^\ell = \epsilon_b(2\omega)\sin\theta_{\text{in}}\chi_{zxx} + \epsilon_\ell(2\omega)K_b\chi_{xxx}\cos 3\phi, \tag{17}$$

and

$$\Gamma_{sP}^\ell = \frac{T_p^{\ell v}T_p^{\ell b}\left(t_s^{v\ell}t_s^{\ell b}\right)^2}{\epsilon_\ell(2\omega)\sqrt{\epsilon_b(2\omega)}}. \tag{18}$$

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

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

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

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

where

$$r_{sS}^\ell = \chi_{xxx} \sin 3\phi, \tag{19}$$

and

$$\Gamma_{sS}^\ell = T_s^{v\ell} T_s^{\ell b} \left( t_s^{v\ell} t_s^{\ell b} \right)^2. \tag{20}$$

In order to reduce above result to that of Ref. [2] and [3], we take the $2\omega$ 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

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

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

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

### A.1.1  Taking all fields in the bulk

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

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

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

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

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 $1\omega$ fields will still be evaluated inside the bulk, so we have Eq. (8)

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

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

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

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So lastly, we have that

$$\mathbf{e}_b^{2\omega} \cdot \boldsymbol{\chi} : \mathbf{e}_b^{\omega}\mathbf{e}_b^{\omega} = \frac{K_b}{K_v} \frac{T_p^{vb}\left(t_p^{vb}\right)^2}{\epsilon_b(\omega)\sqrt{\epsilon_b(2\omega)}} \big( \sin^3\theta_{\text{in}}\chi_{zzz}$$

$$+ k_b^2 \sin\theta_{\text{in}}\cos^2\phi\chi_{zxx}$$
$$+ k_b^2 \sin\theta_{\text{in}}\sin^2\phi\chi_{zyy}$$
$$+ 2k_b \sin^2\theta_{\text{in}}\cos\phi\chi_{zzx}$$
$$+ 2k_b \sin^2\theta_{\text{in}}\sin\phi\chi_{zzy}$$
$$+ 2k_b^2 \sin\theta_{\text{in}}\sin\phi\cos\phi\chi_{zxy}$$
$$- K_b \sin^2\theta_{\text{in}}\cos\phi\chi_{xzz}$$
$$- k_b^2 K_b \cos^3\phi\chi_{xxx}$$
$$- k_b^2 K_b \sin^2\phi\cos\phi\chi_{xyy}$$
$$- 2k_b K_b \sin\theta_{\text{in}}\cos^2\phi\chi_{xzx}$$
$$- 2k_b K_b \sin\theta_{\text{in}}\sin\phi\cos\phi\chi_{xzy}$$
$$- 2k_b^2 K_b \sin\phi\cos^2\phi\chi_{xxy}$$
$$- K_b \sin^2\theta_{\text{in}}\sin\phi\chi_{yzz}$$
$$- k_b^2 K_b \sin\phi\cos^2\phi\chi_{yxx}$$
$$- k_b^2 K_b \sin^3\phi\chi_{yyy}$$
$$- 2k_b K_b \sin\theta_{\text{in}}\sin\phi\cos\phi\chi_{yzx}$$
$$- 2k_b K_b \sin\theta_{\text{in}}\sin^2\phi\chi_{yzy}$$
$$- 2k_b^2 K_b \sin^2\phi\cos\phi\chi_{yxy}\big),$$

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 $\boldsymbol{\chi}$,

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

$$+ k_b^2 \sin\theta_{\text{in}}\cos^2\phi\chi_{zxx}$$
$$+ k_b^2 \sin\theta_{\text{in}}\sin^2\phi\chi_{zxx}$$
$$- 2k_b K_b \sin\theta_{\text{in}}\cos^2\phi\chi_{xxz}$$
$$- 2k_b K_b \sin\theta_{\text{in}}\sin^2\phi\chi_{xxz}$$
$$- k_b^2 K_b \cos^3\phi\chi_{xxx}$$
$$+ k_b^2 K_b \sin^2\phi\cos\phi\chi_{xxx}$$
$$+ 2k_b^2 K_b \sin^2\phi\cos\phi\chi_{xxx}\big),$$

and reducing,

$$= \frac{K_b}{K_v} \Gamma_{pP}^b \big( \sin^3 \theta_{\text{in}} \chi_{zzz}$$
$$+ k_b^2 \sin \theta_{\text{in}} (\sin^2 \phi + \cos^2 \phi) \chi_{zxx}$$
$$- 2 k_b K_b \sin \theta_{\text{in}} (\sin^2 \phi + \cos^2 \phi) \chi_{xxz}$$
$$+ k_b^2 K_b (3 \sin^2 \phi \cos \phi - \cos^3 \phi) \chi_{xxx} \big)$$

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

where,

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

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

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

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

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

### A.1.2   Taking all fields in the vacuum

To consider the $1\omega$ fields in the vacuum, we start with Eq. (6) but substitute $\ell \to v$, thus

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

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

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

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

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

and

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

We also require the $2\omega$ fields evaluated in the vacuum, which is Eq. (5),

$$\mathbf{e}_v^{2\omega} = \hat{\mathbf{e}}^{\text{out}} \cdot \left[ \hat{\mathbf{s}} T_s^{vb} \hat{\mathbf{s}} + \hat{\mathbf{P}}_{v+} \frac{T_p^{vb}}{\sqrt{\epsilon_b(2\omega)}} \left( \epsilon_b(2\omega) \sin\theta_{\text{in}}\hat{\mathbf{z}} - K_b\hat{\boldsymbol{\kappa}} \right) \right], \qquad (21)$$

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

$$\mathbf{e}_v^{2\omega} = \frac{T_p^{vb}}{\sqrt{\epsilon_b(2\omega)}} \left( \epsilon_b(2\omega) \sin\theta_{\text{in}}\hat{\mathbf{z}} - K_b \cos\phi\hat{\mathbf{x}} - K_b \sin\phi\hat{\mathbf{y}} \right). \qquad (22)$$

So lastly, we have that

$$\mathbf{e}_v^{2\omega} \cdot \boldsymbol{\chi} : \mathbf{e}_v^{\omega} \mathbf{v}_v^{\omega} =$$

$$\frac{T_p^{vb}}{\sqrt{\epsilon_b(2\omega)}} \left( \frac{t_p^{vb}}{\sqrt{\epsilon_b(\omega)}} \right)^2 \Big[ \epsilon_b(2\omega) k_b^2 \sin\theta_{\text{in}} \cos^2\phi \chi_{zxx}$$

$$+ \epsilon_b(2\omega) k_b^2 \sin\theta_{\text{in}} \sin^2\phi \chi_{zyy}$$

$$+ \epsilon_b^2(\omega)\epsilon_b(2\omega) \sin^3\theta_{\text{in}} \chi_{zzz}$$

$$+ 2\epsilon_b(2\omega) k_b^2 \sin\theta_{\text{in}} \sin\phi\cos\phi \chi_{zxy}$$

$$+ 2\epsilon_b(\omega)\epsilon_b(2\omega) k_b \sin^2\theta_{\text{in}} \sin\phi \chi_{zyz}$$

$$+ 2\epsilon_b(\omega)\epsilon_b(2\omega) k_b \sin^2\theta_{\text{in}} \cos\phi \chi_{zxz}$$

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

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

$$- \epsilon_b^2(\omega) K_b \sin^2\theta_{\text{in}} \cos\phi \chi_{xzz}$$

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

$$- 2\epsilon_b(\omega) k_b K_b \sin\theta_{\text{in}} \sin\phi\cos\phi \chi_{xyz}$$

$$- 2\epsilon_b(\omega) k_b K_b \sin\theta_{\text{in}} \cos^2\phi \chi_{xxz}$$

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

$$- k_b^2 K_b \sin^3\phi \chi_{yyy}$$

$$- \epsilon_b^2(\omega) K_b \sin^2\theta_{\text{in}} \sin\phi \chi_{yzz}$$

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

$$- 2\epsilon_b(\omega) k_b K_b \sin\theta_{\text{in}} \sin^2\phi \chi_{yyz}$$

$$- 2\epsilon_b(\omega) k_b K_b \sin\theta_{\text{in}} \sin\phi\cos\phi \chi_{yxz} \Big],$$

and after eliminating components,

$$= \Gamma_{pP}^v \Big[ \epsilon_b^2(\omega)\epsilon_b(2\omega) \sin^3\theta_{\text{in}} \chi_{zzz}$$

$$+ \epsilon_b(2\omega) k_b^2 \sin\theta_{\text{in}} \cos^2\phi \chi_{zxx}$$

$$+ \epsilon_b(2\omega) k_b^2 \sin\theta_{\text{in}} \sin^2\phi \chi_{zxx}$$

$$- 2\epsilon_b(\omega) k_b K_b \sin\theta_{\text{in}} \cos^2\phi \chi_{xxz}$$

$$- 2\epsilon_b(\omega) k_b K_b \sin\theta_{\text{in}} \sin^2\phi \chi_{xxz}$$

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

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

$$= \Gamma_{pP}^v \Big[ \epsilon_b^2(\omega)\epsilon_b(2\omega) \sin^3\theta_{\text{in}} \chi_{zzz} + \epsilon_b(2\omega) k_b^2 \sin\theta_{\text{in}} \chi_{zxx}$$

$$- 2\epsilon_b(\omega) k_b K_b \sin\theta_{\text{in}} \chi_{xxz} - k_b^2 K_b \chi_{xxx} \cos 3\phi \Big],$$

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where

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

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

To obtain $R_{pS}(2\omega)$ we use $\mathbf{e}^{\text{in}} = \hat{\mathbf{p}}_{v-}$ in Eq. (7), and $\mathbf{e}^{\text{out}} = \hat{\mathbf{S}}$ in Eq. (4). We also use the unit vectors defined in Eqs. (11) and (12). 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 $2\omega$, and for the fundamental fields,

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

$$= \left(\frac{t_p^{v\ell} t_p^{\ell b}}{\epsilon_\ell(\omega)\sqrt{\epsilon_b(\omega)}}\right)^2 \left(\epsilon_b^2(\omega)\sin^2\theta_{\text{in}}\hat{\mathbf{z}}\hat{\mathbf{z}} + 2\epsilon_b(\omega)\epsilon_\ell(\omega)k_b\sin\theta_{\text{in}}\cos\phi\hat{\mathbf{z}}\hat{\mathbf{x}}\right.$$

$$+\epsilon_\ell^2(\omega)k_b^2\cos^2\phi\hat{\mathbf{x}}\hat{\mathbf{x}} + 2\epsilon_\ell^2(\omega)k_b^2\cos\phi\sin\phi\hat{\mathbf{x}}\hat{\mathbf{y}}$$

$$\left.+\epsilon_\ell^2(\omega)k_b^2\sin^2\phi\hat{\mathbf{y}}\hat{\mathbf{y}} + 2\epsilon_b(\omega)\epsilon_\ell(\omega)k_b\sin\theta_{\text{in}}\sin\phi\hat{\mathbf{y}}\hat{\mathbf{z}}\right).$$

Therefore,

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

$$T_s^{v\ell} T_s^{\ell b}\left(\frac{t_p^{v\ell} t_p^{\ell b}}{\epsilon_\ell(\omega)\sqrt{\epsilon_b(\omega)}}\right)^2\left[-\epsilon_b^2(\omega)\sin^2\theta_{\text{in}}\sin\phi\chi_{xzz}\right.$$

$$-2\epsilon_b(\omega)\epsilon_\ell(\omega)k_b\sin\theta_{\text{in}}\cos\phi\sin\phi\chi_{xxz}$$

$$-\epsilon_\ell^2(\omega)k_b^2\cos^2\phi\sin\phi\chi_{xxx}$$

$$-2\epsilon_\ell^2(\omega)k_b^2\cos\phi\sin^2\phi\chi_{xxy}$$

$$-\epsilon_\ell^2(\omega)k_b^2\sin^3\phi\chi_{xyy}$$

$$-2\epsilon_b(\omega)\epsilon_\ell(\omega)k_b\sin\theta_{\text{in}}\sin^2\phi\chi_{xyz}$$

$$+\epsilon_b^2(\omega)\sin^2\theta_{\text{in}}\cos\phi\chi_{yzz}$$

$$+2\epsilon_b(\omega)\epsilon_\ell(\omega)k_b\sin\theta_{\text{in}}\cos^2\phi\chi_{yxz}$$

$$+\epsilon_\ell^2(\omega)k_b^2\cos^3\phi\chi_{yxx}$$

$$+2\epsilon_\ell^2(\omega)k_b^2\cos^2\phi\sin\phi\chi_{yxy}$$

$$+\epsilon_\ell^2(\omega)k_b^2\cos\phi\sin^2\phi\chi_{yyy}$$

$$\left.+2\epsilon_b(\omega)\epsilon_\ell(\omega)k_b\sin\theta_{\text{in}}\cos\phi\sin\phi\chi_{yyz}\right],$$

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

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

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

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

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

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

and

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

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

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

and

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

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

To obtain $R_{sP}(2\omega)$ we use $\mathbf{e}^{\mathrm{in}} = \hat{\mathbf{s}}$ in Eq. (7), and $\mathbf{e}^{\mathrm{out}} = \hat{\mathbf{P}}_{v+}$ in Eq. (4). We also use the unit vectors defined in Eqs. (11) and (12). Substituting, we get

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

for $2\omega$, and for the fundamental fields,

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

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Therefore,

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

$$\frac{T_p^{v\ell} T_p^{\ell b} \left(t_s^{v\ell} t_s^{\ell b}\right)^2}{\epsilon_\ell(2\omega)\sqrt{\epsilon_b(2\omega)}} \Big[\epsilon_b(2\omega)\sin\theta_{\rm in}\sin^2\phi\chi_{zxx} + \epsilon_b(2\omega)\sin\theta_{\rm in}\cos^2\phi\chi_{zyy}$$

$$- 2\epsilon_b(2\omega)\sin\theta_{\rm in}\sin\phi\cos\phi\chi_{zxy} - \epsilon_\ell(2\omega)K_b\cos\phi\sin^2\phi\chi_{xxx}$$

$$- \epsilon_\ell(2\omega)K_b\cos\phi\cos^2\phi\chi_{xyy} + 2\epsilon_\ell(2\omega)K_b\cos\phi\sin\phi\cos\phi\chi_{xxy}$$

$$- \epsilon_\ell(2\omega)K_b\sin\phi\sin^2\phi\chi_{yxx} - \epsilon_\ell(2\omega)K_b\sin\phi\cos^2\phi\chi_{yyy}$$

$$+ 2\epsilon_\ell(2\omega)K_b\sin\phi\sin\phi\cos\phi\chi_{yxy}\Big],$$

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

$$= \Gamma_{sP}^\ell \Big[\epsilon_b(2\omega)\sin\theta_{\rm in}\sin^2\phi\chi_{zxx} + \epsilon_b(2\omega)\sin\theta_{\rm in}\cos^2\phi\chi_{zxx}$$

$$- \epsilon_\ell(2\omega)K_b\cos\phi\sin^2\phi\chi_{xxx} + \epsilon_\ell(2\omega)K_b\cos^3\phi\chi_{xxx}$$

$$- 2\epsilon_\ell(2\omega)K_b\sin^2\phi\cos\phi\chi_{xxx}\Big]$$

$$= \Gamma_{sP}^\ell \Big[\epsilon_b(2\omega)\sin\theta_{\rm in}(\sin^2\phi + \cos^2\phi)\chi_{zxx}$$

$$- \epsilon_\ell(2\omega)K_b(\cos\phi\sin^2\phi - \cos^3\phi + 2\sin^2\phi\cos\phi)\chi_{xxx}\Big]$$

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

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

We summarize as follows,

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

where

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

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-$\omega$ radiations factors for vacuum by taking $\ell = v$, thus $\epsilon_\ell(2\omega) = 1$, $T_p^{v\ell} = 1$, $T_p^{\ell b} = T_p^{vb}$, and the fundamental field inside medium $b$ by taking $\ell = b$, thus $\epsilon_\ell(\omega) = \epsilon_b(\omega)$, $t_s^{v\ell} = t_s^{vb}$, and $t_s^{\ell b} = 1$. With these choices,

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

and

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

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

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

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

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

Therefore,

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

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. [2] and [3], we take the $2\omega$ 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

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

# References

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