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

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## Contents

# 1 SHG yield per Sipe, Moss, and van Driel [1]

In this treatment, they define the following for all polarizations;

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

where

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

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

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

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

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

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

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

## 1.1 $\mathcal{R}_{pP}$

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

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

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

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

We substitute these in Eq. (6),

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

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

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

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

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

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

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

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

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

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

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

and we can finally complete the expression,

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

where

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

### 1.2 $\mathcal{R}_{pS}$

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

$$
\frac{E^{(2\omega)}(\parallel,\perp)}{E_p^2 A_s} = b_{\parallel,\perp}^{(3)}\sin 3\phi, \qquad (8)
$$

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

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

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

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

As before, we must calculate

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

to obtain the final expression,

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

where

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

### 1.3 $\mathcal{R}_{sP}$

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

$$\frac{E^{(2\omega)}(\perp,\|)}{E_s^2 A_p} = a_{\perp,\|} + c^{(3)}_{\perp,\|}\cos 3\phi, \qquad (10)$$

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

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

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

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

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

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

where

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

## 1.4 Summary

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

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

The necessary factors are summarized in Table 1.

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

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

# 2 SHG yield per Mizrahi and Sipe [2]

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

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

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

## 2.4 Summary

# 3 SHG yield per Mendoza [3]

## 3.1 $\mathcal{R}_{pP}$

## 3.2 $\mathcal{R}_{pS}$

## 3.3 $\mathcal{R}_{sP}$

## 3.4 Summary

# References


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

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

[3] B. S. Mendoza. Longitudinal Gauge Theory of Second Harmonic Generation at Semiconductor Surfaces. In *Epioptics-7: Proceedings of the 24th Course of the International School of Solid State Physics: Erice, Italy, 20-26 July 2002*, volume 23, page 52, 2004.