

1 Preamble

Our interest is to calculate the nonlinear reflection coefficient, \mathcal{R} . We start with equation (2) of [1]

$$\mathcal{R}_{iF} = \frac{32\pi^3\omega^2}{(n_o e)^2 c^3 \cos^2 \theta} |T_F^{vs} T_F^{sb} (t_i^{vs} t_i^{sb})^2 r_{iF}|^2, \quad (1)$$

where $i = s$ or p is the incoming polarization at ω , $F = S$ or P the outgoing polarization at 2ω . θ is the angle of incidence, n_o is the electron density, e is the electron charge, and c is the speed of light. T and t are the Fresnel factors that give the transmitted fields at the vacuum-surface (vs) or the surface-bulk (sb) interfaces. They are given by

$$t_s^{vs}(\omega) = \frac{2 \cos \theta}{\cos \theta + k_{zs}(\omega)}, \quad (2)$$

$$t_p^{vs}(\omega) = \frac{2 \cos \theta}{\epsilon_s \cos \theta + k_{zs}(\omega)}, \quad (3)$$

$$t_s^{sb}(\omega) = \frac{2k_{zs}(\omega)}{k_{zs}(\omega) + k_{zb}(\omega)}, \quad (4)$$

$$t_p^{sb}(\omega) = \frac{2k_{zs}(\omega)}{\epsilon_b(\omega)k_{zs}(\omega) + \epsilon_s(\omega)k_{zb}(\omega)}. \quad (5)$$

where $k_{zj}(\omega) = (w/c) \times (\epsilon_j(\omega) - \sin^2 \theta)^{1/2}$ for $j = s$ (surface) or b (bulk). ϵ_b (ϵ_s) is the bulk (surface) dielectric function. The Fresnel factors for the outgoing fields are $T = t(2\omega)$. Finally, the r_{iF} terms are

$$\begin{aligned} r_{pP} = & \sin \theta \epsilon_b(2\omega) [\sin^2 \theta \epsilon_b^2(\omega) \chi_{\perp\perp\perp} + k_{zb}^2(\omega) \epsilon_s^2(\omega) \chi_{\perp\parallel\parallel\parallel}] \\ & + \epsilon_s(\omega) \epsilon_s(2\omega) k_{zb}(\omega) k_{zb}(2\omega) [-2 \sin \theta \epsilon_b(\omega) \chi_{\parallel\parallel\perp} \\ & + k_{zb}(\omega) \epsilon_s(\omega) \chi_{\parallel\parallel\parallel} \cos(3\phi)], \end{aligned}$$

$$r_{sP} = \sin \theta \epsilon_b(2\omega) \chi_{\perp\parallel\parallel\parallel} - k_{zb}(2\omega) \epsilon_s(2\omega) \chi_{\parallel\parallel\parallel\parallel} \cos(3\phi),$$

$$r_{pS} = -k_{zb}^2(\omega) \epsilon_s^2(\omega) \chi_{\parallel\parallel\parallel\parallel} \sin(3\phi),$$

$$r_{sS} = \chi_{\parallel\parallel\parallel\parallel} \sin(3\phi),$$

where ϕ is the azimuthal angle. χ is the second-order susceptibility tensor with the following relations¹,

¹for this symmetry group.

$$\begin{aligned}
\chi_{\perp\perp\perp\perp} &\equiv \chi_{zzzz}, \\
\chi_{\perp\parallel\parallel\parallel} &\equiv \chi_{zzxx} = \chi_{zyyy}, \\
\chi_{\parallel\parallel\parallel\perp} &\equiv \chi_{xxzz} = \chi_{yyyz}, \\
\chi_{\parallel\parallel\parallel\parallel} &\equiv \chi_{xxxx} = -\chi_{xyyy} = -\chi_{yyxx}.
\end{aligned} \tag{6}$$

Thus, we have four different cases for \mathcal{R}_{iF} : \mathcal{R}_{pP} , \mathcal{R}_{sP} , \mathcal{R}_{pS} , and \mathcal{R}_{sS} . Let us derive the most explicit equations for these cases.

2 Fresnel Factors

We start with $k_{zj}(\omega)$ and substitute into the Fresnel factors. From (2),

$$\begin{aligned}
t_s^{vs}(\omega) &= \frac{2 \cos \theta}{\cos \theta + k_{zs}(\omega)} = \frac{2 \cos \theta}{\cos \theta + (w/c)(\epsilon_s(\omega) - \sin^2 \theta)^{1/2}} \\
&= \frac{2 \cos \theta}{\cos \theta + (w/\sqrt{2c})(2\epsilon_s(\omega) + \cos 2\theta + 1)^{1/2}}.
\end{aligned}$$

From (3),

$$\begin{aligned}
t_p^{vs}(\omega) &= \frac{2 \cos \theta}{\epsilon_s \cos \theta + k_{zs}(\omega)} \\
&= \frac{2 \cos \theta}{\epsilon_s \cos \theta + (w/\sqrt{2c})(2\epsilon_s(\omega) + \cos 2\theta + 1)^{1/2}}.
\end{aligned}$$

From (4),

$$\begin{aligned}
t_s^{sb}(\omega) &= \frac{2k_{zs}(\omega)}{k_{zs}(\omega) + k_{zb}(\omega)} \\
&= \frac{2(\epsilon_s(\omega) - \sin^2 \theta)^{1/2}}{(\epsilon_s(\omega) - \sin^2 \theta)^{1/2} + (\epsilon_b(\omega) - \sin^2 \theta)^{1/2}} \\
&= \frac{2(2\epsilon_s(\omega) + \cos 2\theta + 1)^{1/2}}{(2\epsilon_s(\omega) + \cos 2\theta + 1)^{1/2} + (2\epsilon_b(\omega) + \cos 2\theta + 1)^{1/2}}.
\end{aligned}$$

Lastly, from (5),

$$\begin{aligned}
t_s^{sb}(\omega) &= \frac{2k_{zs}(\omega)}{\epsilon_b(\omega)k_{zs}(\omega) + \epsilon_s(\omega)k_{zb}(\omega)} \\
&= \frac{2(2\epsilon_s(\omega) + \cos 2\theta + 1)^{1/2}}{\epsilon_b(\omega)(2\epsilon_s(\omega) + \cos 2\theta + 1)^{1/2} + \epsilon_s(\omega)(2\epsilon_b(\omega) + \cos 2\theta + 1)^{1/2}}.
\end{aligned}$$

3 r_{iF} Terms

$$\begin{aligned}
r_{pP} = & \sin \theta \epsilon_b(2\omega) [\sin^2 \theta \epsilon_b^2(\omega) \chi_{\perp\perp\perp} + k_{zb}^2(\omega) \epsilon_s^2(\omega) \chi_{\perp\parallel\parallel}] \\
& + \epsilon_s(\omega) \epsilon_s(2\omega) k_{zb}(\omega) k_{zb}(2\omega) [-2 \sin \theta \epsilon_b(\omega) \chi_{\parallel\parallel\perp} \\
& + k_{zb}(\omega) \epsilon_s(\omega) \chi_{\parallel\parallel\parallel} \cos(3\phi)],
\end{aligned}$$

$$r_{sP} = \sin \theta \epsilon_b(2\omega) \chi_{\perp\parallel\parallel} - k_{zb}(2\omega) \epsilon_s(2\omega) \chi_{\parallel\parallel\parallel} \cos(3\phi),$$

$$r_{pS} = -k_{zb}^2(\omega) \epsilon_s^2(\omega) \chi_{\parallel\parallel\parallel} \sin(3\phi),$$

$$r_{sS} = \chi_{\parallel\parallel\parallel} \sin(3\phi),$$

References

- [1] J. E. Mejia, B. S. Mendoza, M. Palummo, G. Onida, R. Del Sole, S. Bergfeld, and W. Daum. Surface second-harmonic generation from Si (111)(1x1) H: Theory versus experiment. *Physical Review B*, 66(19):195329, 2002.