The three layer model

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

For practicality, we wish to review only the main results. From your document, we make the following observations:

1. Eq. (3) of your notes is now replaced by:

$$
\begin{aligned}
r_{pP} &= \sin\theta\epsilon_b(2\omega)\Big[\sin^2\theta\epsilon_b^2(\omega)\chi_{\perp\perp\perp} + k_{zb}^2(\omega)\epsilon_\ell^2(\omega)\chi_{\perp\parallel\parallel}\Big] \\
&\quad - \epsilon_\ell(\omega)\epsilon_\ell(2\omega)k_{zb}(\omega)k_{zb}(2\omega)\Big[2\sin\theta\epsilon_b(\omega)\chi_{\parallel\parallel\perp} + k_{zb}(\omega)\epsilon_\ell(\omega)\chi_{\parallel\parallel\parallel}\cos(3\phi)\Big],
\end{aligned}
\tag{1}
$$

   where the sign on the $\chi_{\parallel\parallel\parallel}$ term has been derived correctly, and is now negative as you pointed out in your notes.

2. Two layer limit: as it turns out, to go from the above expression to the two layer limit, it is a little bit more subtle. The reason being that in the papers by Sipe, Moss, and van Driel [1], and Mizrahi and Sipe [2] the second-harmonic polarization is put on top of the surface in the vacuum region, and the fundamental field is evaluated inside the bulk region.

   In order to reduce the three layer model r_{pP} of Eq. (1) to that of Refs. [2] and [1], we take the 2ω radiations factors for vacuum by taking $\ell = v$, thus $\epsilon_\ell(2\omega) = 1$, and the fundamental field inside medium b by taking $\ell = b$, thus $\epsilon_\ell(\omega) = \epsilon_b(\omega)$. With these choices,

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

   Please note that the $\epsilon_b^2(\omega)$ that is factorized from Eq. (1) is included into the Fresnel factors shown in Eq. (1) of your notes that we do omit here for brevity.

   We remark that Eq. (2) is identical to what one gets from completing the algebra from either Refs. [1] and [2] which both yield the same result.

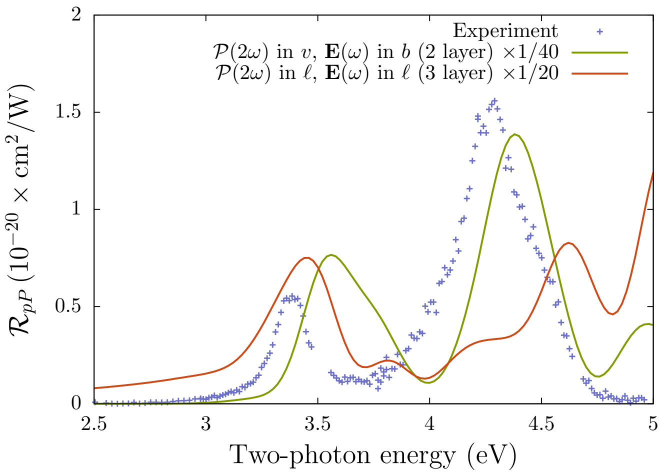

Figure 1: Comparison between the three and two layer models and experiment.

This is in contrast to Eq. 6 of your notes since that equation has an extra $\epsilon_b(2\omega)$ factor multiplying the $\chi_{\perp\|\|}$ term.

In summary, these results are consistent when derived from the three layer model, when derived directly from Ref. [2], and when derived directly from Ref. [1], and that is to say that the two layer model considers the nonlinear polarization on top of the surface in the vacuum region, and the fundamental field in the bulk region. When these considerations are taken into account, the three layer model reduces appropriately to the two layer model.

3. Having clarified the previous points, the three layer model is now capable of representing the following five cases:

Case 1 The three layer model where $\mathcal{P}(2\omega)$ is taken in the small layer below the surface (ℓ) characterized by $\epsilon_\ell(\omega)$ and the fundamental fields are evaluated in the same layer.

Case 2 The two layer model where $\mathcal{P}(2\omega)$ is taken in the vacuum region above the surface (v) characterized by $\epsilon_v(\omega) = 1$ and the fundamental fields are evaluated in the bulk region with $\epsilon_b(\omega)$. We remark that this is the standard model used in the literature, Refs. [1] and [2].

We compare these two models in Fig. 1 with the experiment for $\mathcal{R}_{pP}$. As can be seen from the figure, the two layer model compares more favorably with the experimental results, while the three layer model does not reproduce the E_2 correctly. The intensity of both is higher than the experimental value.

The remaining cases are when

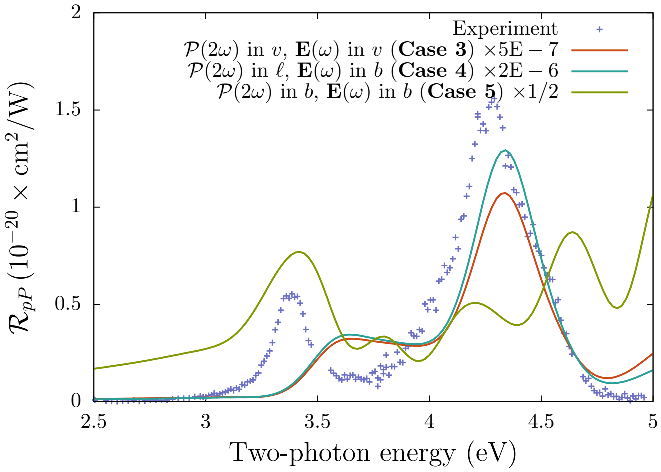

Figure 2: Comparison between the remaining cases for $\mathcal{R}_{pP}$ and experiment.

Case 3 $\mathcal{P}(2\omega)$ is taken in the vacuum region above the surface (v) characterized by $\epsilon_v(\omega) = 1$ and the fundamental fields are also evaluated in the same region.

Case 4 $\mathcal{P}(2\omega)$ is taken in the small layer region below the surface (ℓ) characterized by $\epsilon_\ell(\omega)$ and the fundamental fields are evaluated in the bulk region with $\epsilon_b(\omega)$.

Case 5 $\mathcal{P}(2\omega)$ is taken in the bulk region (b) characterized by $\epsilon_b(\omega)$ and the fundamental fields are also evaluated in the same region.

We compare these three cases in Fig. 2 with the experiment for $\mathcal{R}_{pP}$. As can be seen from the figure, the first two cases yield a similar lineshape where E_1 is not well represented and the intensity of both is 7 and 6 orders of magnitude higher than the experimental value, respectively. The last scenario gives a lineshape where E_1 is well represented but E_2 is incorrectly repdroduced. However, intensity for this case is much closer to the experimental value.

Therefore, we may conclude that the two layer model, where $\mathcal{P}(2\omega)$ is taken in the vacuum region above the surface (v) characterized by $\epsilon_v(\omega) = 1$ and the fundamental fields are evaluated in the bulk region with $\epsilon_b(\omega)$, is the best model for $\mathcal{R}_{pP}$.

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

For $R_{pS}(2\omega)$ we now have

$$r_{pS} = -\epsilon_\ell^2(\omega) k_{zb}^2(\omega) \chi_{\parallel\parallel\parallel} \sin 3\phi. \tag{3}$$

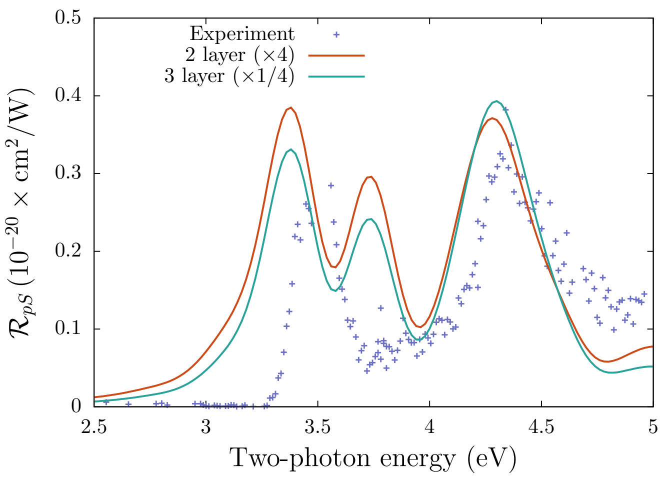

Figure 3: Comparison between the three layer and two layer models with experiment for $\mathcal{R}_{pS}$.

In order to reduce the above result to that of Refs. [1] and [2] (two layer model), we take the 2ω radiations factors for vacuum by taking $\ell = v$, thus $\epsilon_\ell(2\omega) = 1$, and the fundamental field inside medium b by taking $\ell = b$, thus $\epsilon_\ell(\omega) = \epsilon_b(\omega)$. With these choices,

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

Please note that the $\epsilon_b^2(\omega)$ of Eq. (3) is included into the Fresnel factors that we omit here for brevity. In Fig. 3 we show the comparison between the three layer and two layer models and experiment. Except for the intensity which is different, the lineshape is very similar between the two. This is understandable since it is only proportional to $\chi_{\parallel\parallel\parallel}$.

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

For $R_{sP}(2\omega)$ we have

$$r_{sP} = \sin\theta\epsilon_b(2\omega)\chi_{\perp\parallel\parallel} + \epsilon_\ell(2\omega)k_{zb}(2\omega)\chi_{\parallel\parallel\parallel}\cos 3\phi. \tag{4}$$

In order to reduce above result to that of Refs. [1] and [2] (two layer model), we take the 2ω radiations factors for vacuum by taking $\ell = v$, thus $\epsilon_\ell(2\omega) = 1$, and the fundamental field inside medium b by taking $\ell = b$, thus $\epsilon_\ell(\omega) = \epsilon_b(\omega)$. With these choices,

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

In Fig. 4 we show the comparison between the three layer and two layer models and experiment. The intensity of the two layer model is one order of magnitude

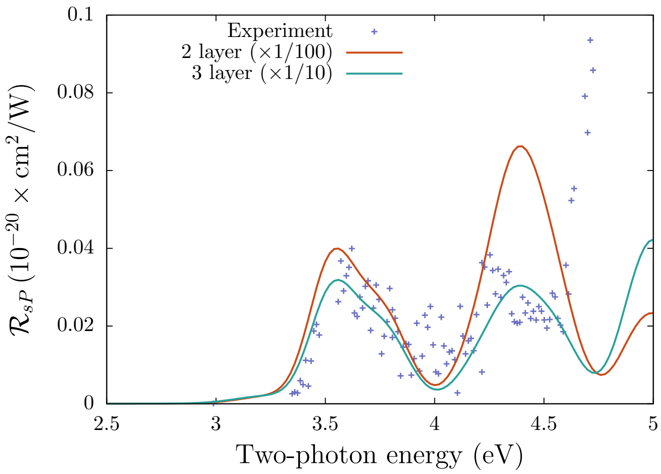

Figure 4: Comparison between the three layer and two layer models with experiment for $\mathcal{R}_{sP}$.

larger than that of the three layer model. The lineshape is similar between the two, with the two layer model producing a more prominent E_2 peak. Since, for the azimuthal angle used ($\phi = 30$) the contribution from the $\chi_{\parallel\parallel\parallel}$ term is zero, both lineshapes are quite similar as they only depend on $\chi_{\perp\parallel\parallel}$

4 Conclusions

What we conclude after this analysis is that the two layer model (**Case 2**) is more appropriate to represent $\mathcal{R}_{iF}$ in general. Therefore, we propose to only use this model and leave the three layer model (**Case 1**) and **Cases 3**, **4**, and **5** out of the manuscript and only present the two layer model.

So, if you agree let us know and we will proceed to change the manuscript accordingly for your next revision!

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.