CENTRO DE INVESTIGACIONES
EN OPTICA, A.C.

Theoretical calculation of Second Harmonic Generation in Semiconductor Surfaces

Bernardo S. Mendoza

Acknowledge:

Sabbatical: Photonics Research Ontario and Department of Physics, University of Toronto, Canada
CONACYT-México (36033-E)

Topics

A. Introduction to second harmonic generation-SHG

B. Longitudinal Gauge Calculation of χ

- Gauge Invariance

C. Three-layer-model for SHG radiation $\mathcal{R}$

A. Introduction-SHG

Second harmonic generation SHG
is an optical technique that can be used as a surface probe
to characterize in situ
structural and electronic properties of surfaces

- no need of ultra high vacuum (UHV) conditions
- is nondestructive
- is noninvasive
- covers a wide spectral range
- buried interfaces

A. Introduction-SHG

Surface SHG is based on the fact that the surface and the bulk have different structural symmetry

$$P_i = \underbrace{\chi_{ijk} E_j E_k}_{\text{dipolar}} + \overbrace{\chi_{ijkl} E_j \nabla_k E_l}^{\text{quadrupolar}} + \cdots$$

Centrosymmetric system (Bulk): $\boldsymbol{r} \to -\boldsymbol{r}$

$$P_i'(-\boldsymbol{r}) = \chi_{ijk}' E_j'(-\boldsymbol{r}) E_k'(-\boldsymbol{r}) \to -P_i = \chi_{ijk}' (-E_j)(-E_k)$$

$$\chi_{ijk} = -\chi_{ijk}'$$

A. Introduction-SHG

but $\chi_{ijk} = \chi'_{ijk} \Rightarrow \chi^b_{ijk} = 0$

$\Rightarrow$ within dipole approximation **NO BULK CONTRIBUTION**

BUT the surface is not-centrosymmetric!!

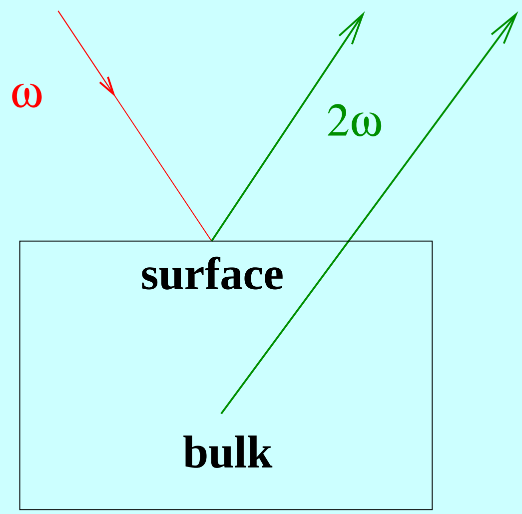
$\Rightarrow \chi^s_{ijk} \neq 0$

Surface $\Rightarrow$ $P^s_i(2\omega) = \chi^s_{ijk} E_j(\omega) E_k(\omega)$ (dipolar)

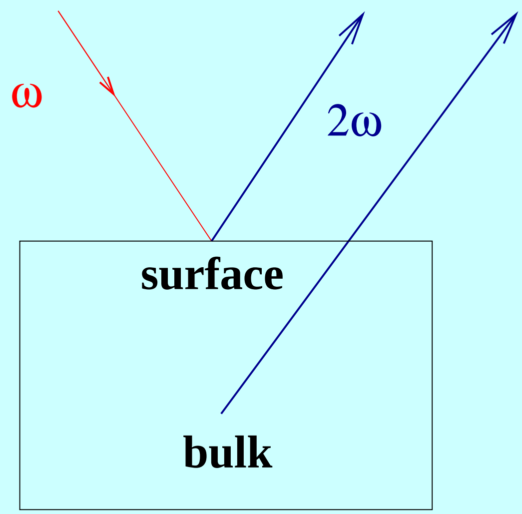
Bulk $\Rightarrow$ $P^b_i(2\omega) = \chi_{ijkl} E_j(\omega) \nabla_k E_l(\omega) \sim 0$ (quadrupolar)

$\Rightarrow$ SHG is surface sensitive

A. Introduction-SHG

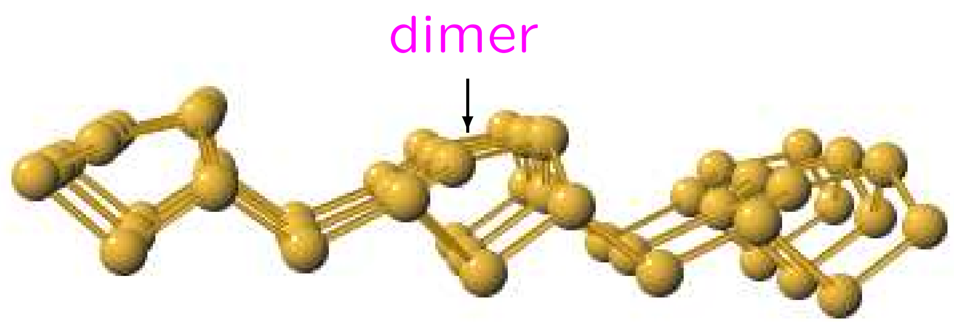


A. Introduction-SHG



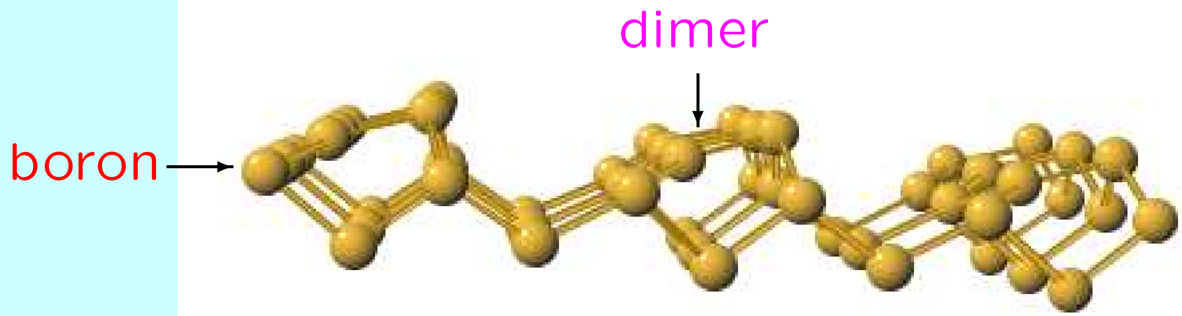
A. Introduction-SHG

A. Introduction-SHG



Clean Si(100)2×1 surface

A. Introduction-SHG

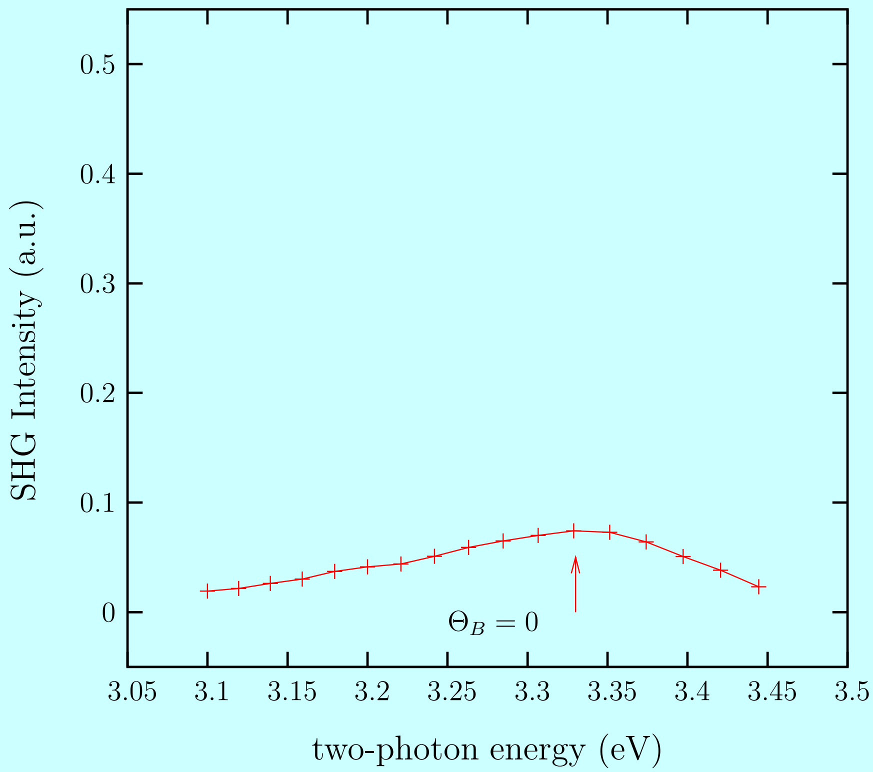


Clean Si(100)2 $\times$ 1 surface

A. Introduction-SHG

Boron-Reconstructed Si(100)

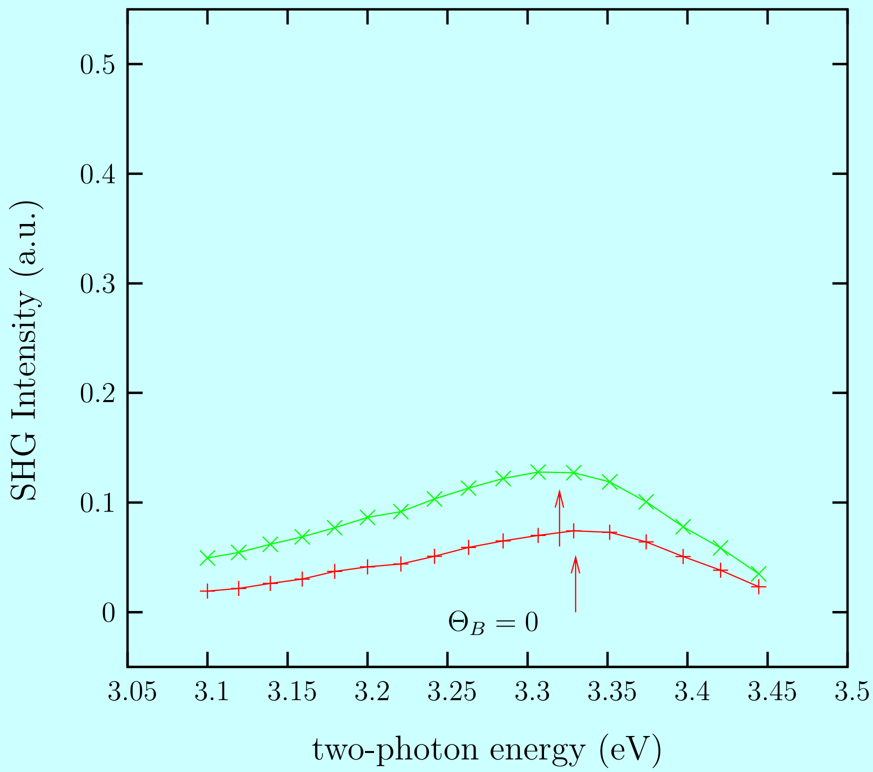
Downer, Mendoza et al. PRL **84**, 3406 (2000)



A. Introduction-SHG

Boron-Reconstructed Si(100)

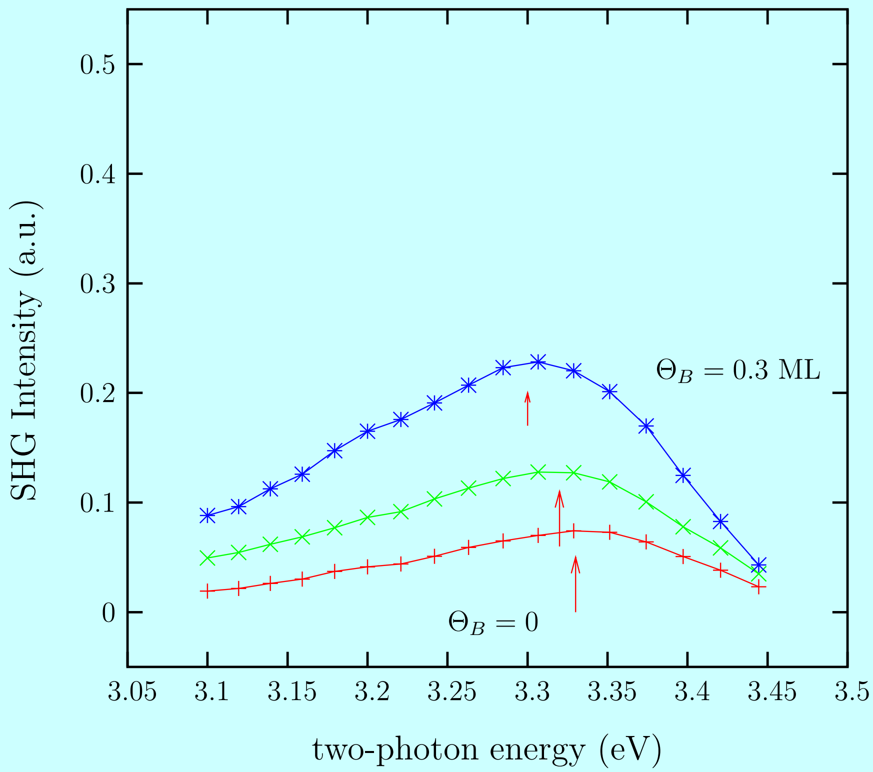
Downer, Mendoza et al. PRL **84**, 3406 (2000)



A. Introduction-SHG

Boron-Reconstructed Si(100)

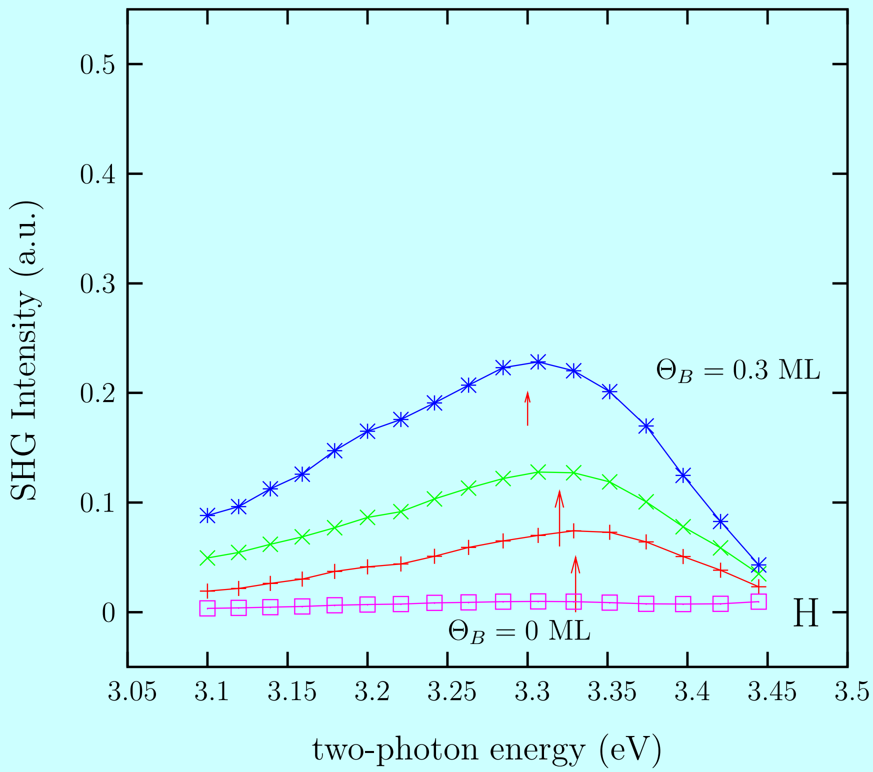
Downer, Mendoza et al. PRL **84**, 3406 (2000)



A. Introduction-SHG

Boron-Reconstructed Si(100)

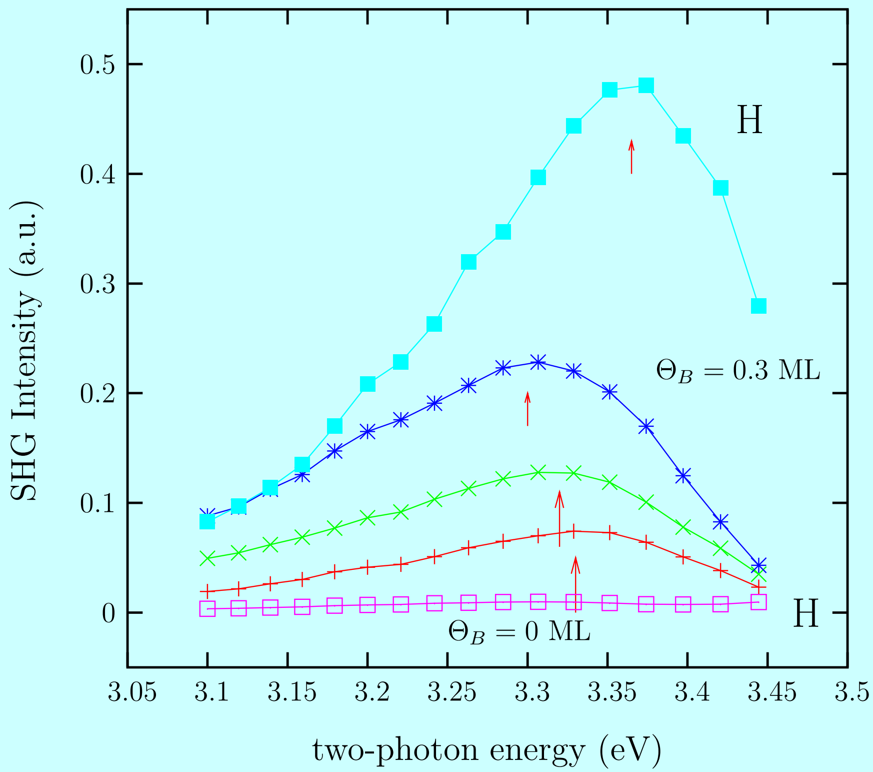
Downer, Mendoza et al. PRL **84**, 3406 (2000)



A. Introduction-SHG

Boron-Reconstructed Si(100)

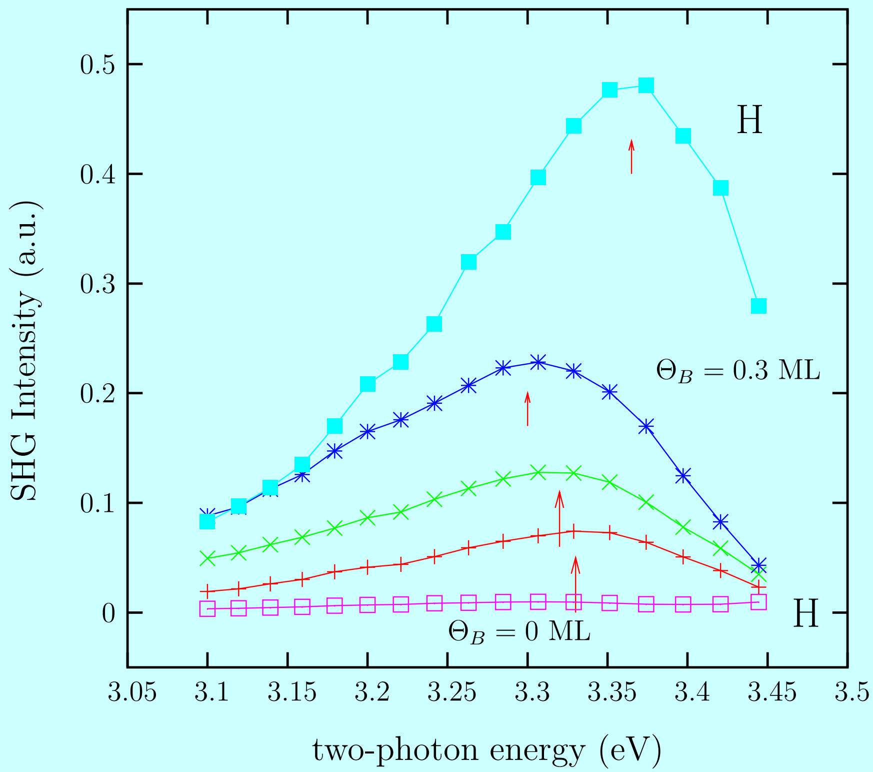
Downer, Mendoza et al. PRL **84**, 3406 (2000)

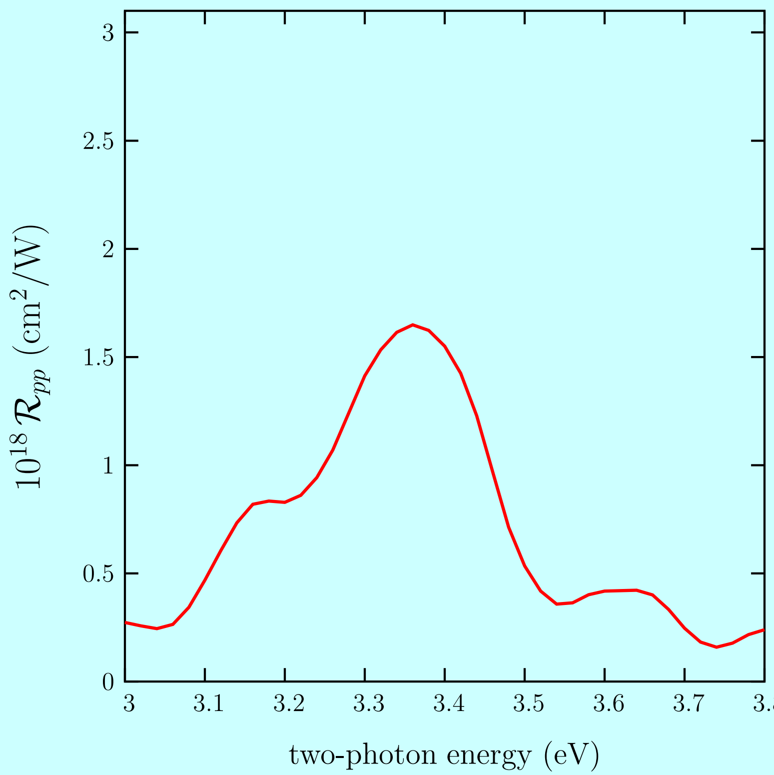


A. Introduction-SHG

Boron-Reconstructed Si(100)

Downer, Mendoza et al. PRL **84**, 3406 (2000)

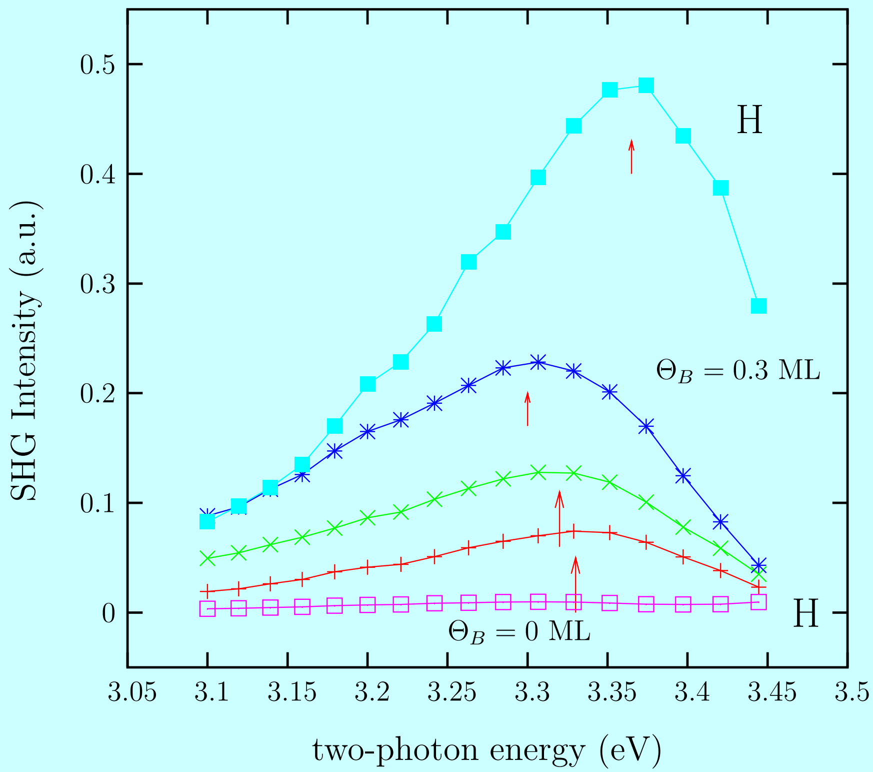


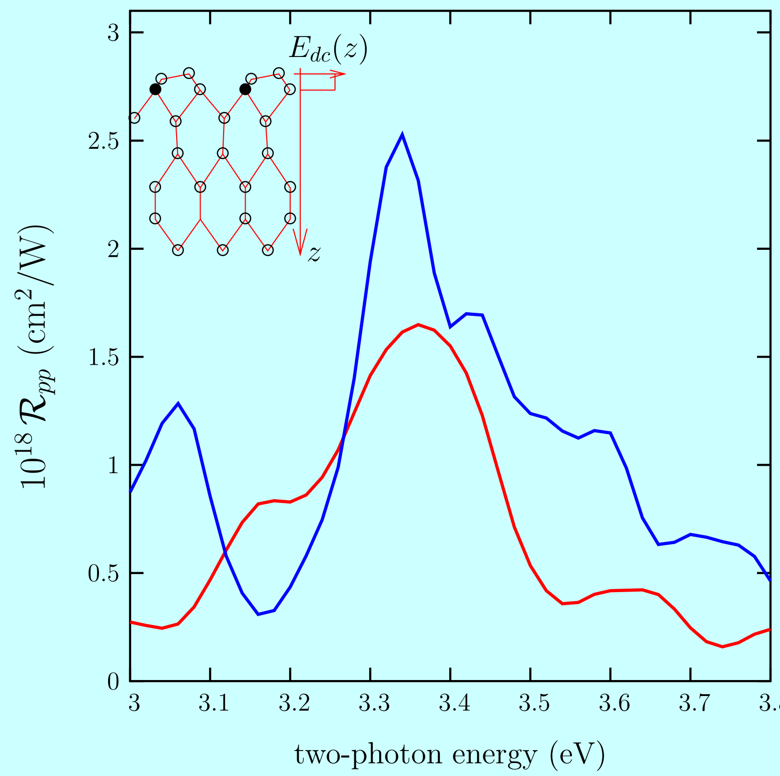


A. Introduction-SHG

Boron-Reconstructed Si(100)

Downer, Mendoza et al. PRL **84**, 3406 (2000)

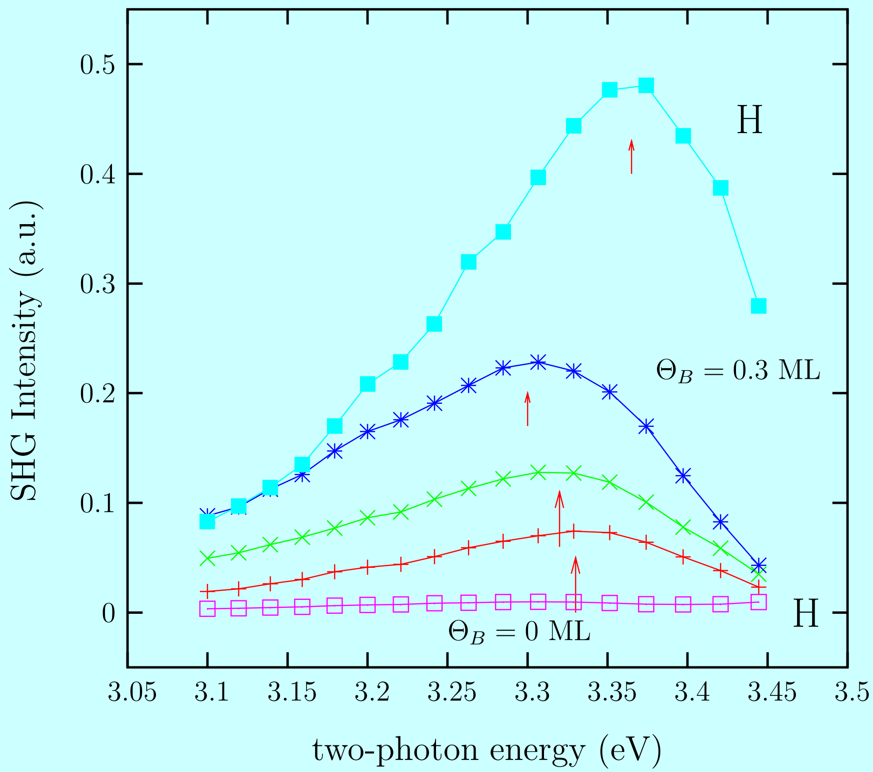


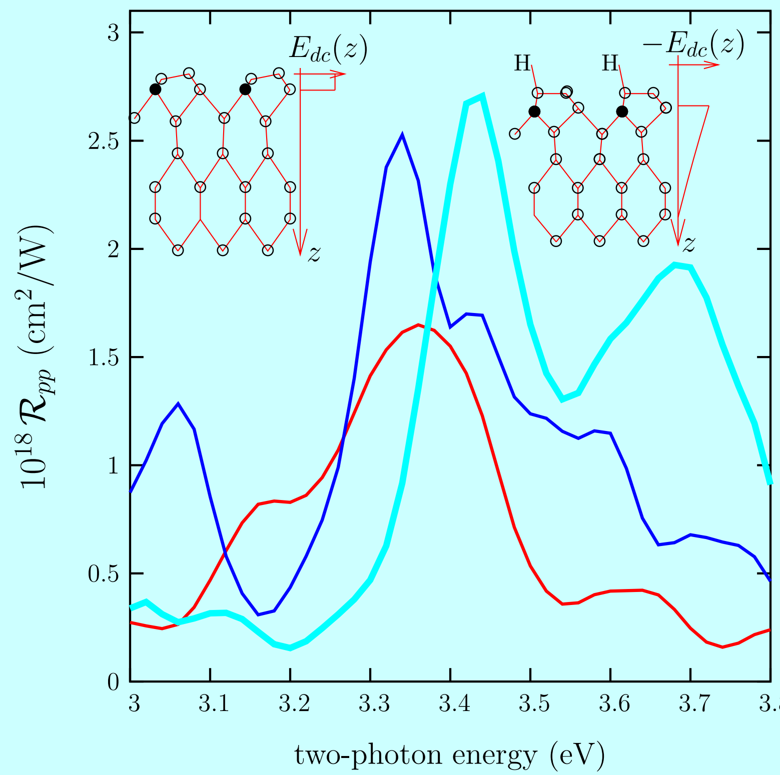


A. Introduction-SHG

Boron-Reconstructed Si(100)

Downer, Mendoza et al. PRL **84**, 3406 (2000)





B. Longitudinal Gauge Calculation of χ

We follow the article by

C. Aversa and J. E. Sipe, Phys. Rev. B **52**, 14636 (1995)

A more recent derivation can also be found in

J. E. Sipe and A. I. Shkrebtii, Phys. Rev. B **61**, 5337 (2000)

W. R. L. Lambertch and S. N. Rashkeev, phys. stat. sol. (b) **217** 599 (2000)

B. Longitudinal Gauge Calculation of χ

$$H(\boldsymbol{r},t) = H_0(\boldsymbol{r}) \underbrace{-e\boldsymbol{r}\cdot\boldsymbol{E}(\boldsymbol{r},t)}_{\text{perturbation}} \tag{1}$$

$$H_0(\boldsymbol{r}) = p^2/2m + V(\boldsymbol{r}) \tag{2}$$

where $V(\boldsymbol{r}) = V(\boldsymbol{r}+\boldsymbol{R})$ is the periodic crystal potential

and the electric field

$$\boldsymbol{E}(\boldsymbol{r},t) = -\frac{1}{c}\frac{\partial}{\partial t}\boldsymbol{A}(\boldsymbol{r},t)$$

with $\boldsymbol{A}(\boldsymbol{r},t)$ the vector potential

B. Longitudinal Gauge Calculation of χ

$$H_0|n\boldsymbol{k}>=\hbar\omega_n(\boldsymbol{k})|n\boldsymbol{k}> \tag{3}$$

$$<\boldsymbol{r}|n\boldsymbol{k}>=e^{i\boldsymbol{k}\cdot\boldsymbol{r}}u_{n\boldsymbol{k}}(\boldsymbol{r}) \quad \text{Bloch States} \tag{4}$$

$u_{n\boldsymbol{k}}(\boldsymbol{r})=u_{n\boldsymbol{k}}(\boldsymbol{r}+\boldsymbol{R})$ is cell periodic and

$$<n\boldsymbol{k}|m\boldsymbol{k}'>=\delta_{nm}\delta(\boldsymbol{k}-\boldsymbol{k}') \tag{5}$$

B. Longitudinal Gauge Calculation of χ

$$
\begin{aligned}
< n\boldsymbol{k}|\boldsymbol{r}|m\boldsymbol{k}' > &= i\int d\boldsymbol{r} e^{-i\boldsymbol{k}\cdot\boldsymbol{r}} u^*_{n\boldsymbol{k}}(\boldsymbol{r}) \nabla_{\boldsymbol{k}'} e^{i\boldsymbol{k}'\cdot\boldsymbol{r}} u_{m\boldsymbol{k}'}(\boldsymbol{r}) \\
&= \int d\boldsymbol{r} e^{-i\boldsymbol{k}\cdot\boldsymbol{r}} u^*_{n\boldsymbol{k}}(\boldsymbol{r})(-\boldsymbol{r}) e^{i\boldsymbol{k}'\cdot\boldsymbol{r}} u_{m\boldsymbol{k}'}(\boldsymbol{r}) \\
&\quad + \int d\boldsymbol{r} e^{-i(\boldsymbol{k}-\boldsymbol{k}')\cdot\boldsymbol{r}} u^*_{n\boldsymbol{k}}(\boldsymbol{r})(i\nabla_{\boldsymbol{k}'}) u_{m\boldsymbol{k}'}(\boldsymbol{r}) \\
&= -i\nabla_{\boldsymbol{k}'} \int d\boldsymbol{r} < n\boldsymbol{k}|m\boldsymbol{k}' > \\
&\quad + i\sum_{\boldsymbol{R}} e^{-i(\boldsymbol{k}-\boldsymbol{k}')\cdot\boldsymbol{R}} \int_\Omega d\boldsymbol{\tau} e^{-i(\boldsymbol{k}-\boldsymbol{k}')\cdot\boldsymbol{r}} u^*_{n\boldsymbol{k}}(\boldsymbol{\tau}) \nabla_{\boldsymbol{k}'} u_{m\boldsymbol{k}'}(\boldsymbol{\tau}) \\
&= \delta(\boldsymbol{k}-\boldsymbol{k}')\xi_{nm}(\boldsymbol{k}) - i\delta_{nm}\nabla_{\boldsymbol{k}'}\delta(\boldsymbol{k}-\boldsymbol{k}')
\end{aligned}
\tag{6}
$$

Ω unit cell volume

$$
\xi_{nm}(\boldsymbol{k}) = i\frac{(2\pi)^3}{\Omega}\int_\Omega d\boldsymbol{\tau} e^{-i(\boldsymbol{k}-\boldsymbol{k}')\cdot\boldsymbol{r}} u^*_{n\boldsymbol{k}}(\boldsymbol{\tau}) \nabla_{\boldsymbol{k}'} u_{m\boldsymbol{k}'}(\boldsymbol{\tau}) \tag{7}
$$

B. Longitudinal Gauge Calculation of χ

$$\boldsymbol{r} = \overbrace{\boldsymbol{r}_e}^{\text{interband}} + \underbrace{\boldsymbol{r}_i}_{\text{intraband}} \tag{8}$$

$$< n\boldsymbol{k}|\boldsymbol{r}_e|m\boldsymbol{k}' >= (1 - \delta_{nm})\delta(\boldsymbol{k} - \boldsymbol{k}')\xi_{nm}(\boldsymbol{k}) \tag{9}$$

$$< n\boldsymbol{k}|\boldsymbol{r}_i|m\boldsymbol{k}' >= \delta_{nm}\left[\delta(\boldsymbol{k} - \boldsymbol{k}')\xi_{nn}(\boldsymbol{k}) + i\nabla_{\boldsymbol{k}}\delta(\boldsymbol{k} - \boldsymbol{k}')\right] \tag{10}$$

B. Longitudinal Gauge Calculation of χ

Polarization $\boldsymbol{P}(t)$

Independent particle approximation
electron density operator ρ

$$\frac{dP^a}{dt} = \frac{e}{m} \sum_{nm\boldsymbol{k}} p^a_{mn}(\boldsymbol{k}) \rho_{nm}(\boldsymbol{k}) \tag{11}$$

momentum operator $\boldsymbol{p}$

$$i\hbar \frac{d}{dt} \rho(t) = [H, \rho(t)]$$

B. Longitudinal Gauge Calculation of χ

interaction picture $\Rightarrow$

$$i\hbar\frac{d\tilde{\rho}}{dt} = [-e\tilde{\boldsymbol{r}}\cdot\boldsymbol{E}(t), \tilde{\rho}] \tag{12}$$

$$\tilde{O} = UOU^\dagger$$

$$U = \exp(iH_O t/\hbar)$$

solution by standard iterative scheme

B. Longitudinal Gauge Calculation of χ

unperturbed density operator

$$\tilde{\rho}^{(0)} \equiv \rho_0$$

$$< m\boldsymbol{k}|\rho_0|n\boldsymbol{k}' >= f_n(\hbar\omega_n(\boldsymbol{k}))\delta_{mn}\delta(\boldsymbol{k}-\boldsymbol{k}') \quad \text{Fermi-Dirac} \qquad (13)$$

Eq. (12) $\Rightarrow$

$$\tilde{\rho}_{nm}^{(N+1)}(t) = \frac{ie}{\hbar}\int_{-\infty}^{t} dt' e^{i\omega_{nm}t'}\boldsymbol{E}(t')\cdot\left[\boldsymbol{R}_e^{(N)}(t') + \boldsymbol{R}_i^{(N)}(t')\right] \qquad (14)$$

$$\omega_{nm} = \omega_n - \omega_m$$

B. Longitudinal Gauge Calculation of χ

$$\boldsymbol{R}_e^{(N)} = < n\boldsymbol{k}|[\boldsymbol{r}_e, \rho^{(N)}]|m\boldsymbol{k}' > = \sum_{\ell} \left(\boldsymbol{r}_{n\ell}\rho_{\ell m}^{(N)} - \rho_{n\ell}^{(N)}\boldsymbol{r}_{\ell m} \right) \tag{15}$$

$$[H_0, \boldsymbol{r}] = \hbar\boldsymbol{p}/im \Rightarrow$$

$$\boldsymbol{r}_{nm} \equiv (1 - \delta_{nm})\xi_{nm} = \boldsymbol{p}_{nm}/im\omega_{nm} \quad (n \neq m)$$

$$\boldsymbol{R}_i^{(N)} = < n\boldsymbol{k}|[\boldsymbol{r}_i, \rho^{(N)}]|m\boldsymbol{k}' > = \nabla_{\boldsymbol{k}}\rho_{nm}^{(N)} - i\rho_{nm}^{(N)}(\xi_{nn} - \xi_{mm}) \tag{16}$$

Any operator $\mathcal{O}$

$$< n\boldsymbol{k}|[\boldsymbol{r}_i, \mathcal{O}]|m\boldsymbol{k}' > = \nabla_{\boldsymbol{k}}\mathcal{O}_{nm} - i\mathcal{O}_{nm}(\xi_{nn} - \xi_{mm}) \equiv (\mathcal{O}_{nm})_{;\boldsymbol{k}} \tag{17}$$

B. Longitudinal Gauge Calculation of χ

Harmonic Perturbation

$$\boldsymbol{E}(t) = \boldsymbol{E} \exp(-i\omega t)$$

$$\rho_{nm}^{(1)}(t) = B_{nm}^{a} E^{a} \exp(-i\omega t)$$

(sum over repeated Cartesian indices)

$$B_{nm}^{a} = \frac{e}{\hbar} \frac{f_{mn} r_{nm}^{a}}{\omega_{nm} - \omega} \tag{18}$$

$$f_{mn} = f_m - f_n$$

B. Longitudinal Gauge Calculation of χ

$(f_n)_{;\boldsymbol{k}} = 0$ for a semiconductor at $T = 0$

$$\Rightarrow \boldsymbol{R}_i^{(0)} = 0$$

The linear response has no contribution from intraband transitions!

$$P^b(\omega) = \chi_{ba}^{(1)} E^a(\omega)$$

Eq. 11 $\Rightarrow$

$$\chi_{ba}^{(1)}(-\omega;\omega) = \frac{e^2}{\hbar} \sum_{nm\boldsymbol{k}} \frac{f_{mn}(\boldsymbol{k}) r_{nm}^a(\boldsymbol{k}) r_{mn}^b(\boldsymbol{k})}{\omega_{nm}(\boldsymbol{k}) - \omega} \tag{19}$$

B. Longitudinal Gauge Calculation of χ

$\rho_{nm}^{(1)}$ in Eq. (14) $\Rightarrow$

$$\rho_{nm}^{(2)}(t) = \frac{e}{i\hbar}\frac{1}{\omega_{nm} - \omega_3}\left[-(B_{nm}^b)_{;\boldsymbol{k}} + i\sum_{\ell}\left(r_{n\ell}^c B_{\ell m}^b - B_{n\ell}^b r_{\ell m}^c\right)\right] E_1^b E_2^c e^{-i\omega_3 t} \tag{20}$$

$\omega_3 = \omega_1 + \omega_2$; $\boldsymbol{E}_i(\omega_i)$ for $i = 1, 2$

$$P^a(\omega_3) = \chi^{abc}(-\omega_3; \omega_1, \omega_2) E_1^b E_2^c$$

Eq. 11 $\Rightarrow$

B. Longitudinal Gauge Calculation of χ

interband

$$\chi_e^{abc} = \frac{e^3}{m\hbar^2\omega_3} \sum_{\ell mn\boldsymbol{k}} \frac{p^a_{mn}}{\omega_{nm} - \omega_3} \left(\frac{r^c_{n\ell} r^b_{\ell m} f_{m\ell}}{\omega_{\ell m} - \omega_2} - \frac{r^b_{n\ell} r^c_{\ell m} f_{\ell n}}{\omega_{n\ell} - \omega_2} \right) \tag{21}$$

intraband

$$\chi_i^{abc} = -\frac{e^3}{m\hbar^2\omega_3} \sum_{mn\boldsymbol{k}} \frac{p^a_{mn}}{\omega_{nm} - \omega_3} \left(\frac{f_{mn} r^b_{nm}}{\omega_{nm} - \omega_2} \right)_{;\boldsymbol{k}} \tag{22}$$

$$\chi^{abc} = \chi_e^{abc} + \chi_i^{abc}$$

symmetrized for intrinsic permutation symmetry

$$\chi^{abc}(-\omega_3;\omega_1,\omega_2) = \chi^{acb}(-\omega_3;\omega_2,\omega_1)$$

$$-\omega_3 + \omega_2 + \omega_1 = 0$$

SHG $\Rightarrow$ $\omega_1 = \omega_2 = \omega$ and $\omega_3 = 2\omega$

B. Longitudinal Gauge Calculation of χ

$$\left(\frac{f_{mn} r^b_{nm}}{\omega_{nm} - \omega_2}\right)_{;k^c} = \frac{f_{mn}}{\omega_{nm} - \omega}\left(r^b_{nm}\right)_{;k^c} - \frac{f_{mn} r^b_{nm}}{(\omega_{nm} - \omega)^2}\left(\omega_{nm}\right)_{;k^c} \quad (23)$$

$$(\omega_{nm})_{;k^a} = (\omega_n)_{;k^a} - (\omega_m)_{;k^a}$$

$$< n\boldsymbol{k}|[H_0, \boldsymbol{r}]|n\boldsymbol{k} >= -i\hbar\nabla_{\boldsymbol{k}}\omega_n(\boldsymbol{k}) = \hbar\boldsymbol{p}_{nn}/im$$

$$(\omega_{nm})_{;k^a} = (p^a_{nn} - p^a_{mm})/m \equiv \Delta^a_{nm}$$

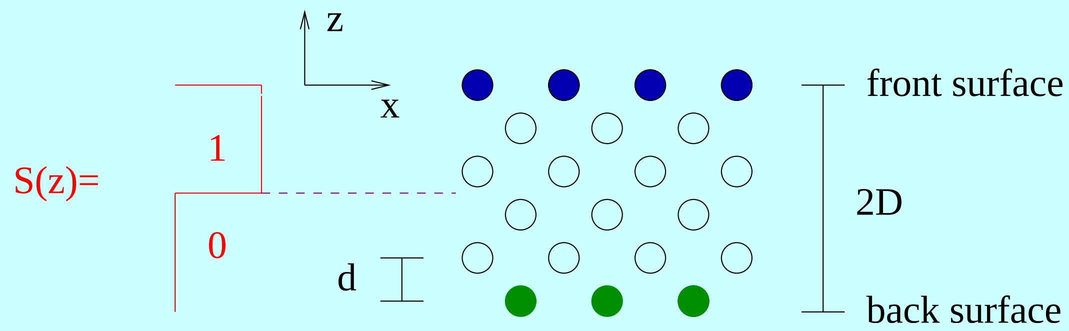
B. Longitudinal Gauge Calculation of χ

$$[r^a, p^b] = [r_i^a, p^b] + [r_e^a, p^b] = i\hbar\delta^{ab}$$

$$\Rightarrow$$

$$(r_{nm}^b)_{;k^a} = \frac{r_{nm}^a \Delta_{mn}^b + r_{nm}^b \Delta_{mn}^a}{\omega_{nm}} + \frac{i}{\omega_{nm}} \sum_\ell \left(\omega_{\ell m} r_{n\ell}^a r_{\ell m}^b - \omega_{n\ell} r_{n\ell}^b r_{\ell m}^a \right) \tag{24}$$

B. Longitudinal Gauge Calculation of χ



surface calculation $\longrightarrow$ Slab

$$p^a \longrightarrow \frac{p^a S(z) + S(z) p^a}{2} \tag{25}$$

with $S(z)$ a suitable cut function to avoid the destructive interference from both surfaces of the slab

B. Longitudinal Gauge Calculation of χ

Gauge Invariance

$$H' = (-e/mc)\boldsymbol{A} \cdot \boldsymbol{p} \quad \text{Transverse Gauge}$$

Eq. (12) $\Rightarrow$

$$i\hbar(\tilde{\rho}_T(t) - \rho_0) = \frac{-e}{mc}\int_{-\infty}^{t} dt'[\tilde{\boldsymbol{p}}(t') \cdot \boldsymbol{A}(t'), \tilde{\rho}_T(t')] \tag{26}$$

(T→ transverse;integrating by parts)

$$\begin{aligned} i\hbar(\tilde{\rho}_T(t) - \rho_0) \;&=\; \frac{-e}{c}[\tilde{\boldsymbol{r}}(t) \cdot \boldsymbol{A}(t), \tilde{\rho}_T(t)] - e\int_{-\infty}^{t} dt'[\tilde{\boldsymbol{r}} \cdot \boldsymbol{E}, \tilde{\rho}_T(t')] \\ &\quad -\frac{e}{c}\int_{-\infty}^{t} dt'[\tilde{\boldsymbol{r}} \cdot \boldsymbol{A}, \dot{\tilde{\rho}}_T(t')] \end{aligned} \tag{27}$$

B. Longitudinal Gauge Calculation of χ

First order $\rightarrow$

$$\tilde{\rho}_T^{(1)}(t) = \tilde{\rho}_L^{(1)}(t) - \frac{e}{i\hbar c}[\tilde{\boldsymbol{r}}(t) \cdot \boldsymbol{A}(t), \rho_O] \tag{28}$$

(L$\rightarrow$ longitudinal)

$$\boldsymbol{v} = \frac{1}{m}\boldsymbol{p} - \frac{e}{mc}\boldsymbol{A} \quad \text{(T-gauge)}$$

Eq. (11) $\Rightarrow$

$$\chi_T^{(1)} = \chi_L^{(1)} + S^{(1)}$$

B. Longitudinal Gauge Calculation of χ

Harmonic Perturbation $\rightarrow$

$$S^{(1)} = \frac{-ie^2}{\omega^2 m\hbar} \sum_{mn} (C_1)_{mn} (\rho_0)_{nm} \tag{29}$$

$$C_1 = [r^a, p^b] - i\hbar\delta^{ab}$$

$$[r^a, p^b] = i\hbar\delta^{ab} \Rightarrow S^{(1)} = 0 \Rightarrow$$

$$\chi_T^{(1)} = \chi_L^{(1)}$$

B. Longitudinal Gauge Calculation of χ

$$\chi_T^{(N)} = \chi_L^{(N)} + S^{(N)}$$

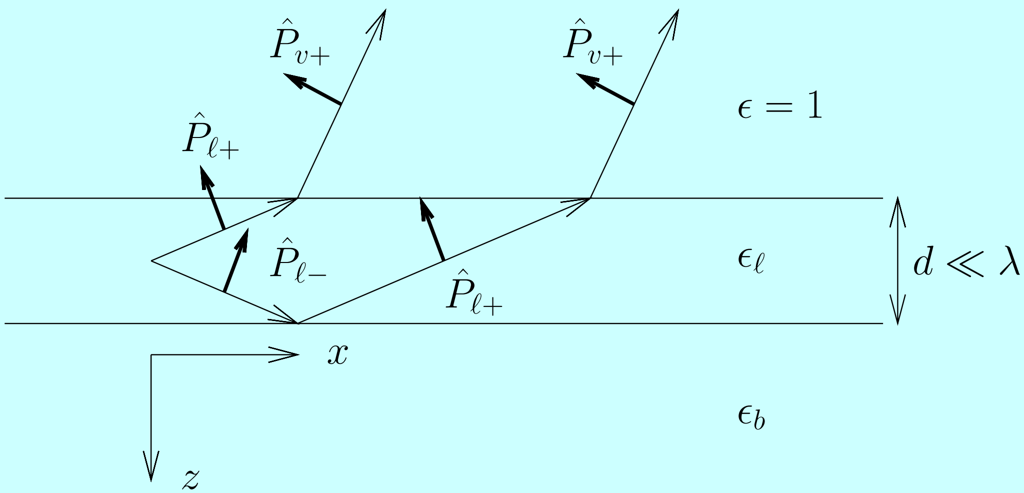
commutator identities $\Rightarrow S^{(N)} = 0$

$$\chi_T^{(N)} = \chi_L^{(N)}$$

$$([r^a, p^b])_{nn} = \sum_m \left(r^a_{nm} p^b_{mn} - p^b_{nm} r^a_{mn} \right) = \sum_m \frac{p^a_{nm} p^b_{mn} - p^b_{nm} p^a_{mn}}{i m \omega_{nm}}$$

$$\sum_m \frac{p^a_{nm} p^b_{mn} - p^b_{nm} p^a_{mn}}{\omega_{nm}} = m\hbar\delta^{ab} \qquad \forall\ n \neq m \qquad (30)$$

C. Three-layer-model for SHG Radiation



(V. Mizrahi and J. E. Sipe J. Opt. Soc. Am. B **5**, 660 (1988))

$$(E_{p\pm}, E_s) = (\frac{2\pi i\tilde{\omega}^2}{w}\,\widehat{p}_\pm \cdot \boldsymbol{P}, \frac{2\pi i\tilde{\omega}^2}{w}\,\widehat{s} \cdot \boldsymbol{P})$$

upward (+) or downward (−) direction of propagation

$$\tilde{\omega} = \omega/c \qquad w = \tilde{\omega} k_z \qquad k_z(\omega) = \sqrt{\epsilon(\omega) - \sin^2\theta}$$

$$\widehat{p}_\pm = \frac{\mp k_z \widehat{x} - \sin\theta\,\widehat{z}}{\sqrt{\epsilon}}$$

θ is the angle of incidence

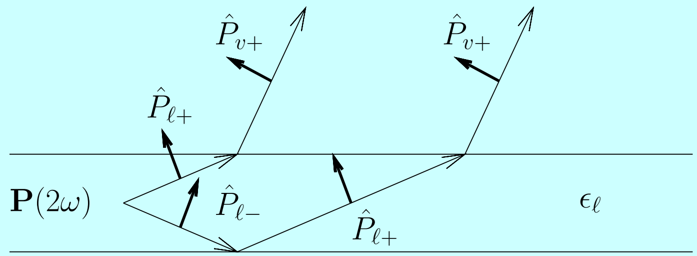
C. Three-layer-model for SHG Radiation

$$\mathcal{R}(\omega) = I(2\omega)/I^2(\omega) \tag{31}$$

$$I(\omega) = c/8\pi|E(\omega)|^2 \tag{32}$$

$$P_i = \chi_{ijk}E_j(\omega)E_k(\omega)$$

C. Three-layer-model for SHG Radiation



layer-vacuum interface (ℓv) transmission

$$\boldsymbol{T}_{\ell v} = \hat{s}T_s^{\ell v}\hat{s} + \hat{P}_{v+}\tilde{T}_p^{\ell v}\hat{P}_{\ell+} \tag{33}$$

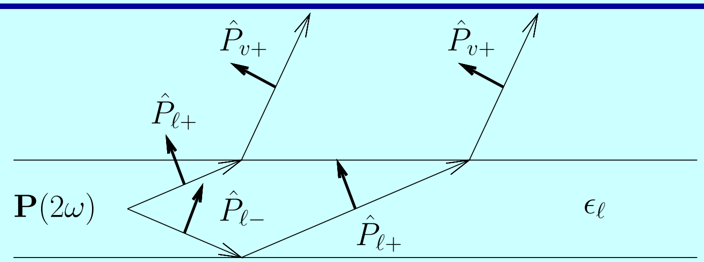
layer-bulk interface (ℓb) reflection

$$\boldsymbol{R}_{\ell b} = \hat{s}R_s^{\ell b}\hat{s} + \hat{P}_{\ell+}R_p^{\ell b}\hat{P}_{\ell-} \tag{34}$$

Capital Letters $\Rightarrow 2\omega$

$\hat{P}_{i\pm}(\epsilon_i)$ $i = v, \ell, b$ $R's$ & $T's$ Fresnel Factors

C. Three-layer-model for SHG Radiation



Total radiated field at 2ω

$$\boldsymbol{E}(2\omega) = E_s(2\omega)\,(\boldsymbol{T}_{\ell v} + \boldsymbol{R}_{\ell b}\cdot\boldsymbol{T}_{\ell v})\cdot\hat{s} + E_{p+}(2\omega)\boldsymbol{T}_{\ell v}\cdot\hat{P}_{\ell+} + E_{p-}(2\omega)\boldsymbol{T}_{\ell v}\cdot\boldsymbol{R}_{\ell b}\cdot\hat{P}_{\ell-} \quad (35)$$

first (third) term is the transmitted s (p)-polarized field

second (fourth) term is the reflected and then transmitted s (p)-polarized field

C. Three-layer-model for SHG Radiation

$$\boldsymbol{E}(2\omega) = \frac{4\pi i\tilde{\omega}}{K_{z\ell}}\boldsymbol{H}\cdot\boldsymbol{P} \tag{36}$$

$$\boldsymbol{H} = \hat{s}\,T_s^{\ell v}\left(1 + R_s^{\ell b}\right)\hat{s} + \hat{P}_{v+}\tilde{T}_p^{\ell v}\left(\hat{P}_{\ell+} + R_p^{\ell b}\hat{P}_{\ell-}\right) \tag{37}$$

$$E(2\omega) = \hat{e}^{out}\cdot\boldsymbol{E}(2\omega)$$

$\hat{e}^{out} = \hat{s}$, $\hat{P}_{v+}$ or any combination

$$E(2\omega) = \frac{4\pi i\omega}{c}\boldsymbol{e}^{2\omega}\cdot\boldsymbol{P} \tag{38}$$

C. Three-layer-model for SHG Radiation

$$\boldsymbol{e}^{2\omega} = \frac{1}{\cos\theta}\widehat{e}^{out}\cdot\Big[\widehat{s}T_s^{v\ell}T_s^{\ell b}\widehat{s} - \widehat{P}_{v+}T_p^{v\ell}T_p^{\ell b}\left(\epsilon_\ell(2\omega)K_{zb}\widehat{x} + \epsilon_b(2\omega)\sin\theta\widehat{z}\right)\Big] \tag{39}$$

$$T_s^{\ell v} = \frac{K_{z\ell}}{\cos\theta}T_s^{v\ell} \qquad \tilde{T}_p^{\ell v} = \frac{\sqrt{\epsilon_\ell(2\omega)}K_{z\ell}}{\cos\theta}T_p^{v\ell} \tag{40}$$

$$1 - R_p^{\ell b} = \frac{\epsilon_\ell(2\omega)K_{zb}}{K_{z\ell}}T_p^{\ell b} \qquad 1 + R_p^{\ell b} = \epsilon_b(2\omega)T_p^{\ell b} \tag{41}$$

C. Three-layer-model for SHG Radiation

Eq. (38) $\Rightarrow$

$$E_s(2\omega) = \frac{4\pi i\omega}{c\cos\theta} T_s^{v\ell} T_s^{\ell b} \chi_{yij} E_i(\omega) E_j(\omega) \tag{42}$$

$$E_p(2\omega) = \frac{-4\pi i\omega}{c\cos\theta} T_p^{v\ell} T_p^{\ell b} \left[\epsilon_\ell(2\omega) K_{zb} \chi_{xij} + \epsilon_b(2\omega) \sin\theta \chi_{zij}\right] E_i(\omega) E_j(\omega) \tag{43}$$

$E_i(\omega)$ is the incident field given by the properly screened external field

C. Three-layer-model for SHG Radiation

$$\boldsymbol{E}_s(\omega) = E_o t_s^{v\ell}\left(1 + r_s^{\ell b}\right)\hat{y} \tag{44}$$

$$\boldsymbol{E}_p(\omega) = E_o\left[\tilde{t}_p^{v\ell}\left(1 - r_p^{\ell b}\right)\cos\theta_\ell\hat{x} - \tilde{t}_p^{v\ell}\left(1 + r_p^{\ell b}\right)\sin\theta_\ell\hat{z}\right] \tag{45}$$

E_o is the incoming amplitude

θ_ℓ is the angle of refraction in the layer

Eqs. (40-41) $\Rightarrow$

$$\boldsymbol{E}_s(\omega) = E_o t_s^{v\ell} t_s^{\ell b}\hat{y} \tag{46}$$

$$\boldsymbol{E}_p(\omega) = E_o t_p^{v\ell} t_p^{\ell b}\left(\epsilon_\ell(\omega) k_{zb}\hat{x} - \epsilon_b(\omega)\sin\theta\hat{z}\right) \tag{47}$$

C. Three-layer-model for SHG Radiation

Eqs. (31), (32), (42), (43), (46), (47) $\Rightarrow$

$$\mathcal{R}_{iF}(2\omega) = \frac{32\pi^3\omega^2}{c^3\cos^2\theta}\left|T_F^{v\ell}T_F^{\ell b}(t_i^{v\ell}t_i^{\ell b})^2\, r_{iF}\right|^2 \tag{48}$$

i (lower case) initial polarization and F (upper case) final polarization

$$r_{iP} = \Big(K_{zb}\chi_{xjk}(2\omega) + \sin\theta\,\chi_{zjk}(2\omega)\Big)\, E_j^i(\omega)E_k^i(\omega) \tag{49}$$

$$r_{iS} = \chi_{yjk}(2\omega)E_j^i(\omega)E_k^i(\omega) \tag{50}$$

$\boldsymbol{E}^s = \hat{y}$ and $\boldsymbol{E}^p = \epsilon_\ell(\omega)k_{zb}\hat{x} - \epsilon_b(\omega)\sin\theta\hat{z}$

C. Three-layer-model for SHG Radiation

Fresnel factors:

$$t_s^{v\ell}(\omega) = \frac{2\cos\theta}{\cos\theta + k_{z\ell}(\omega)} \qquad t_p^{v\ell}(\omega) = \frac{2\cos\theta}{\epsilon_\ell(\omega)\cos\theta + k_{z\ell}(\omega)} \tag{51}$$

$$t_s^{\ell b}(\omega) = \frac{2k_{z\ell}(\omega)}{k_{z\ell} + k_{zb}(\omega)} \qquad t_p^{\ell b}(\omega) = \frac{2k_{z\ell}(\omega)}{\epsilon_b(\omega)k_{z\ell}(\omega) + \epsilon_s(\omega)k_{zb}(\omega)} \tag{52}$$

Three-Layer Model

$$\begin{cases} \epsilon_\ell = 1 & \Rightarrow \text{ No screening} \\ \epsilon_\ell = \epsilon_b & \Rightarrow \text{ Fresnel screening} \end{cases}$$

This has been a

Microsoft-free

presentation!!

THANKS!