Optimized Software for Theoretical Nonlinear Optical Calculations

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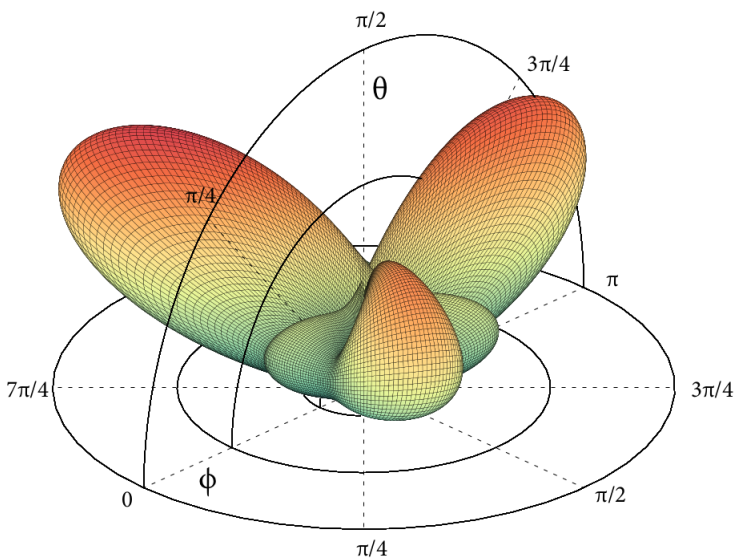
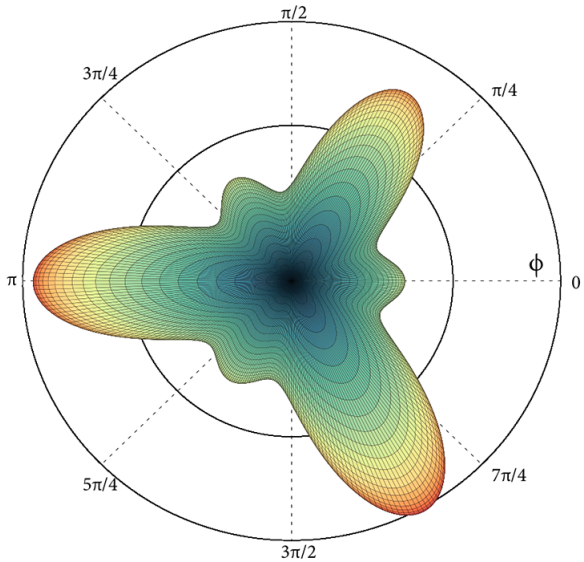
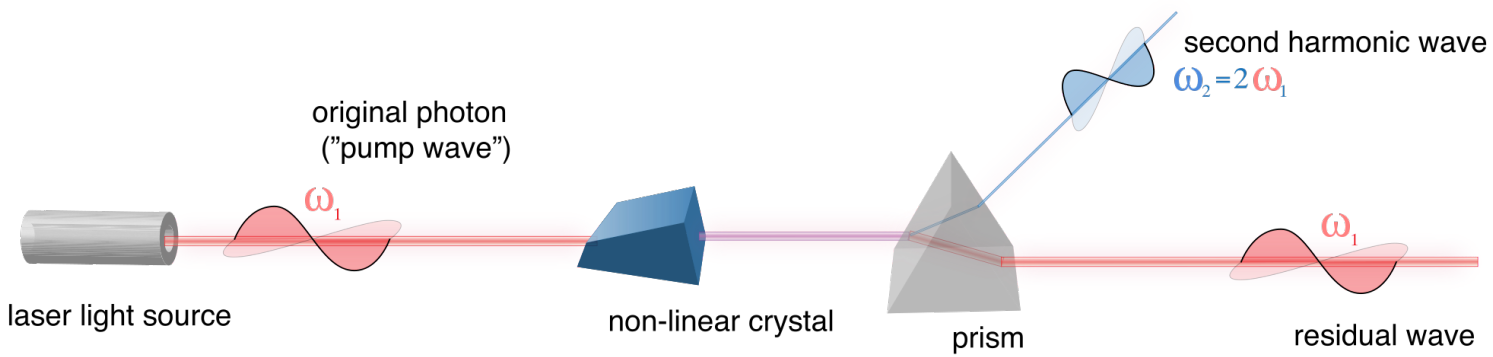




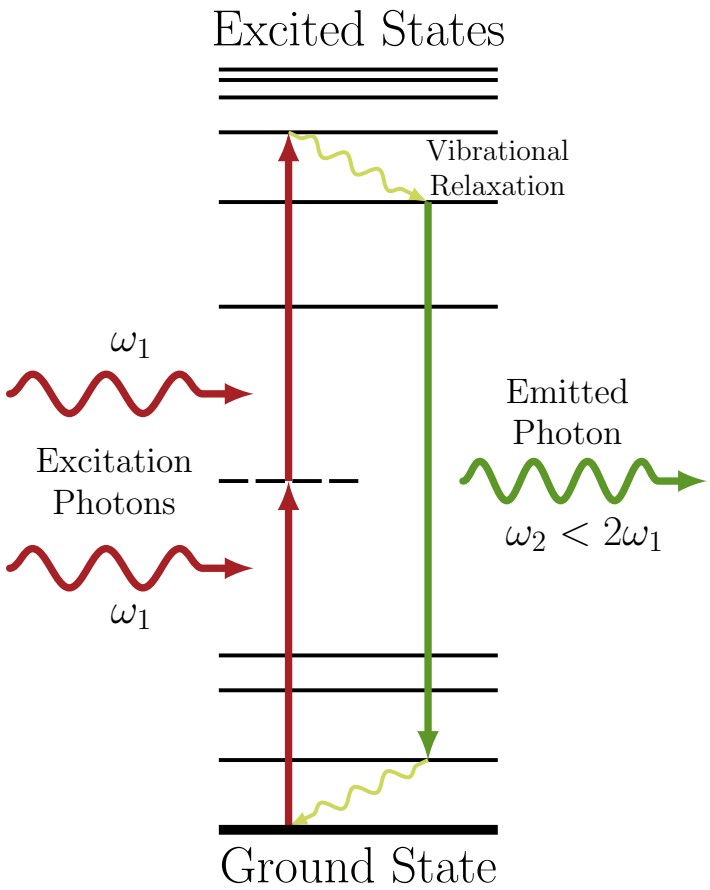
Second Harmonic Generation (SHG)

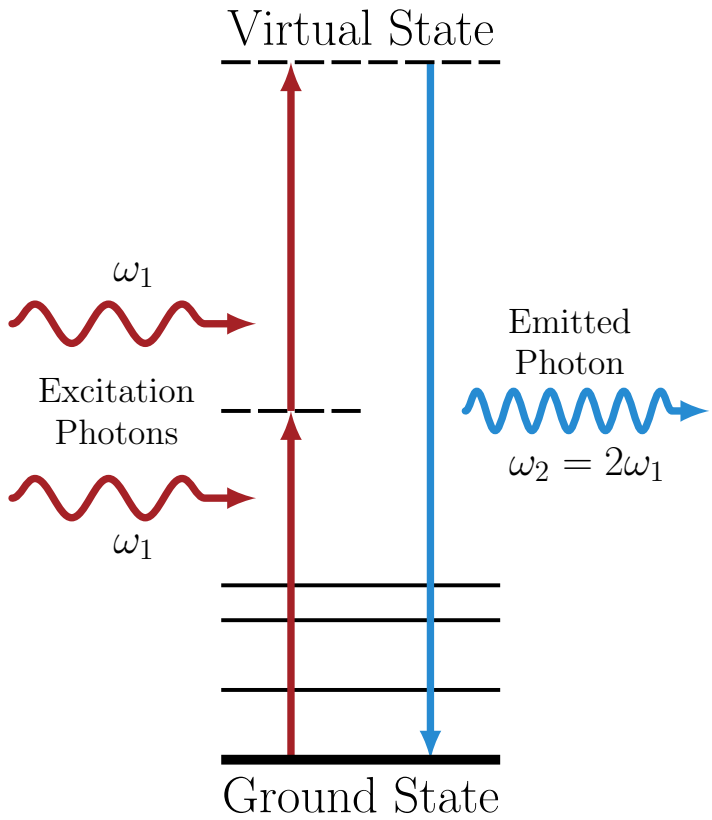
Characteristics[1]

- Two photons of the same frequency combine
- Create one photon of double the frequency



[1] Image: Jon Chui


Excited States
Vibrational
Relaxation
ω_1
Excitation
Photons
ω_1
Emitted
Photon
$\omega_2 < 2\omega_1$
Ground State


Virtual State
ω_1
Excitation
Photons
ω_1
Emitted
Photon
$\omega_2 = 2\omega_1$
Ground State

Second-order Nonlinear Effects

Early work[2] [3] demonstrated that second-order processes

- Are dipole forbidden in the bulk of centrosymmetric materials
- Are related to $\chi^{(2)}$, the nonlinear susceptibility
- Have bigger dipolar (surface) than quadrupolar contributions

Second-order processes are well studied for flat surfaces, but what about round materials like nanospheres?

[2] J.A. Armstrong et al. *Physical Review*, 127(6):1918–1939, Sep 1962.
[3] N. Bloembergen et al. *Physical Review*, 128(2):606–622, Oct 1962.

Summary

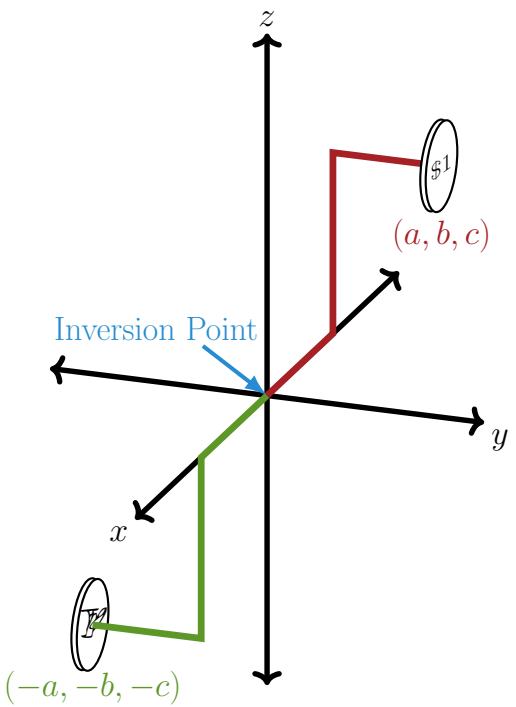
Nonlinear response depends on

- Nonlocal excitation of the electric dipole moment
- Local excitation of the electric quadrupole moment
- The strength of the incident beam and
- The form (plane wave, Gaussian beam, polarization, etc.)
- The quadrupolar $(\mathbf{E} \cdot \nabla)\,\mathbf{E}$ term

What's the best way to enhance this signal?

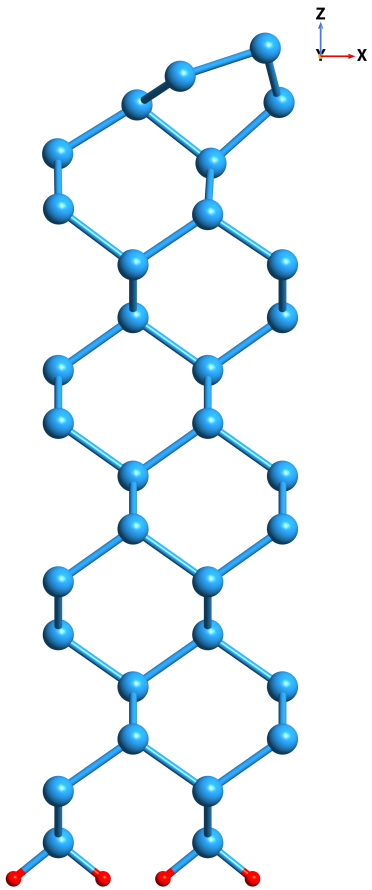
Centrosymmetric Materials

A centrosymmetric material is a material that displays inversion symmetry, such that $p(x, y, z) \rightarrow p(-x, -y, -z)$.

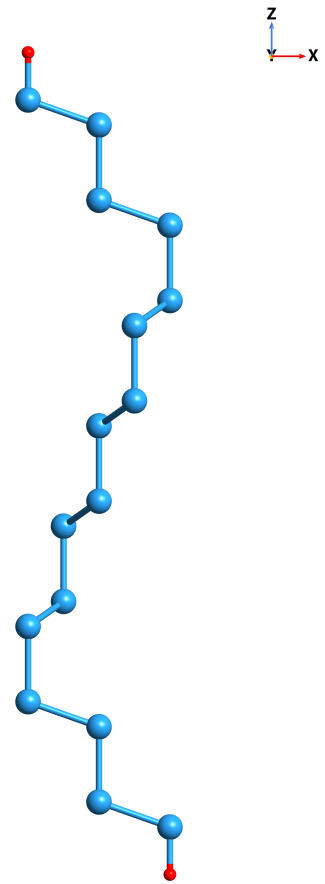


- Many nonlinear materials are centrosymmetric
- Nanospheres are definitely centrosymmetric
- The material in these nanoparticles is centrosymmetric

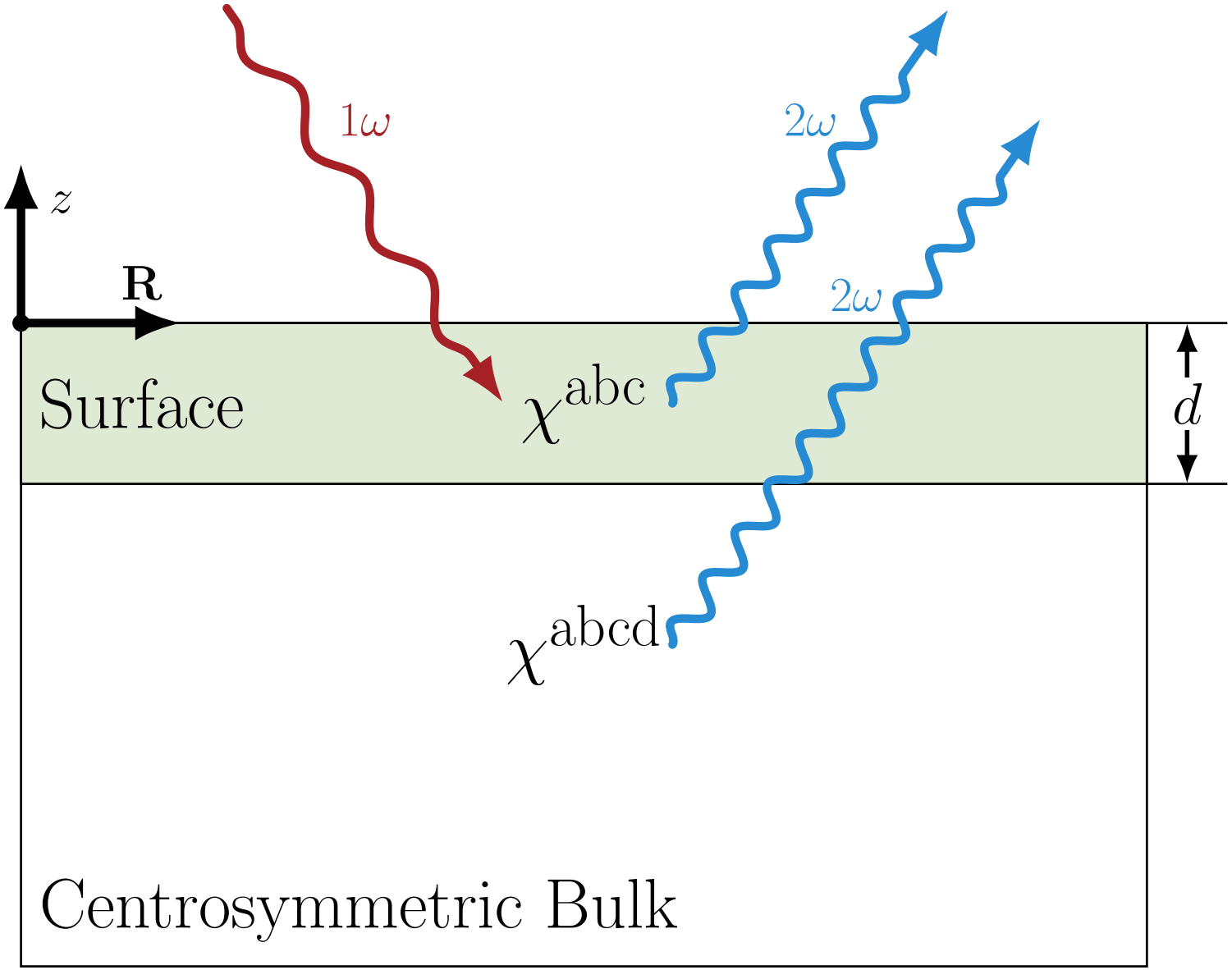
Test Cases

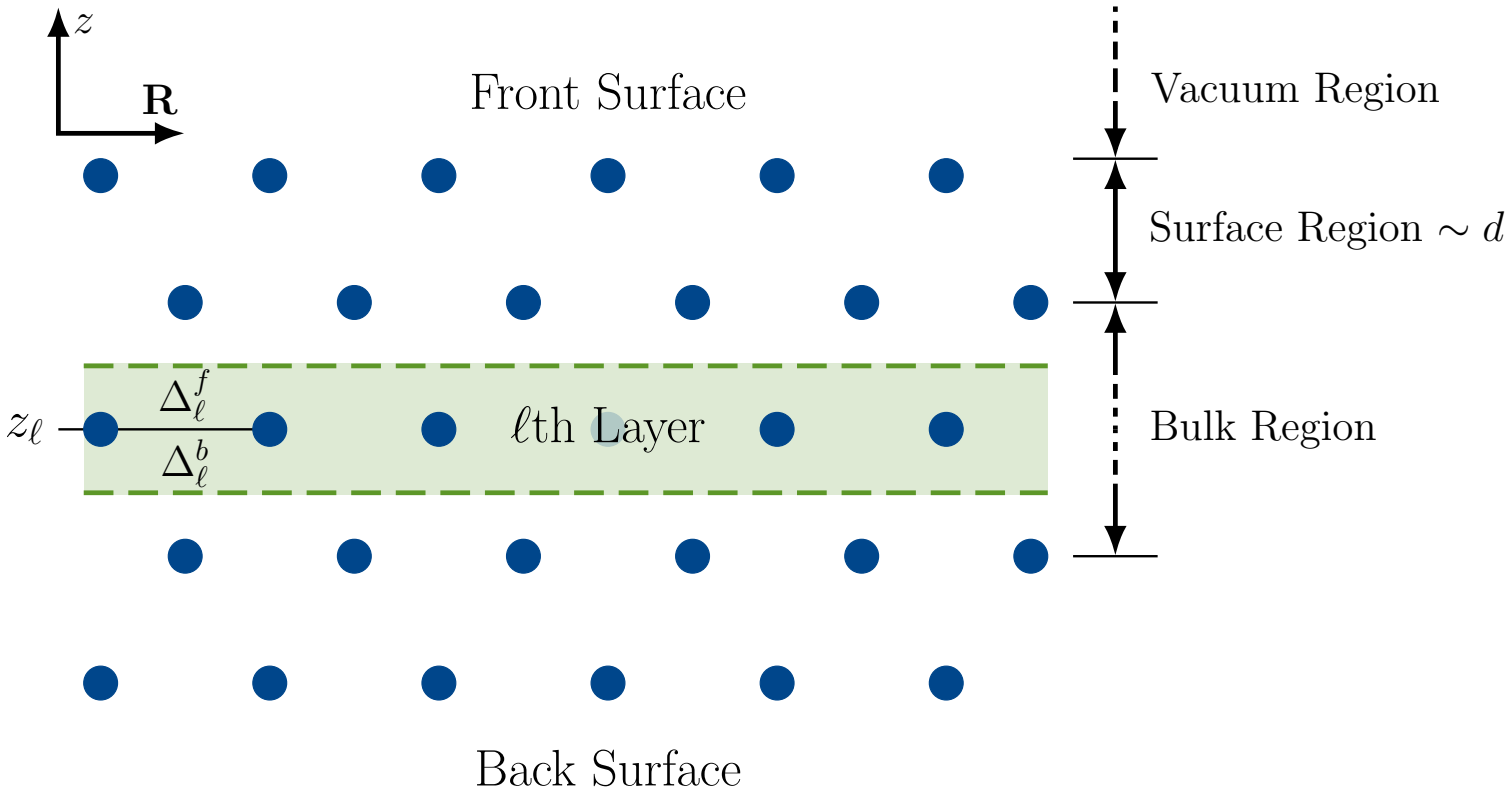


Si(001)(2×1)

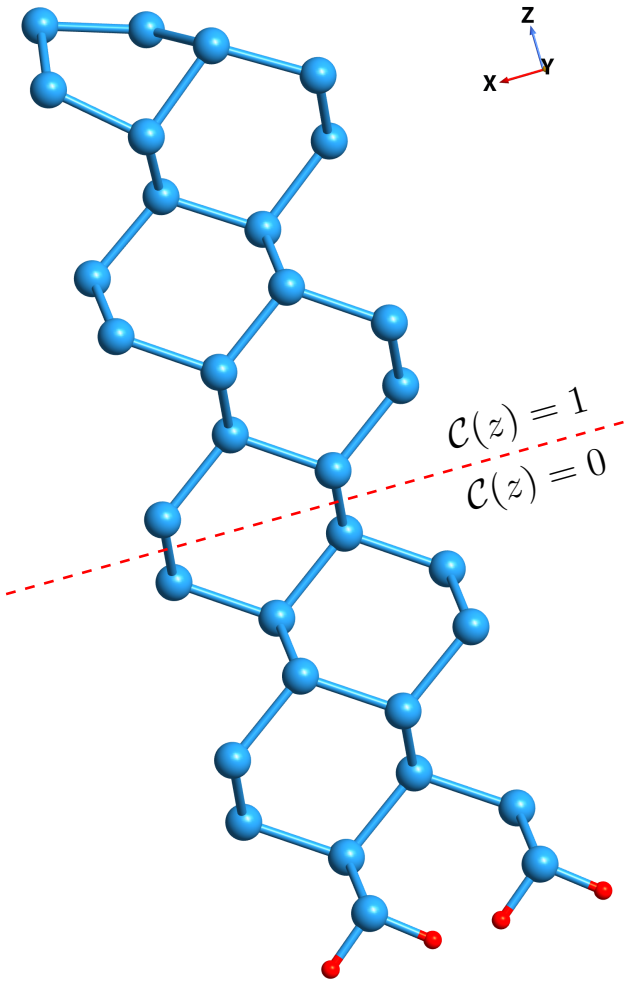


Si(111)(1×1):H

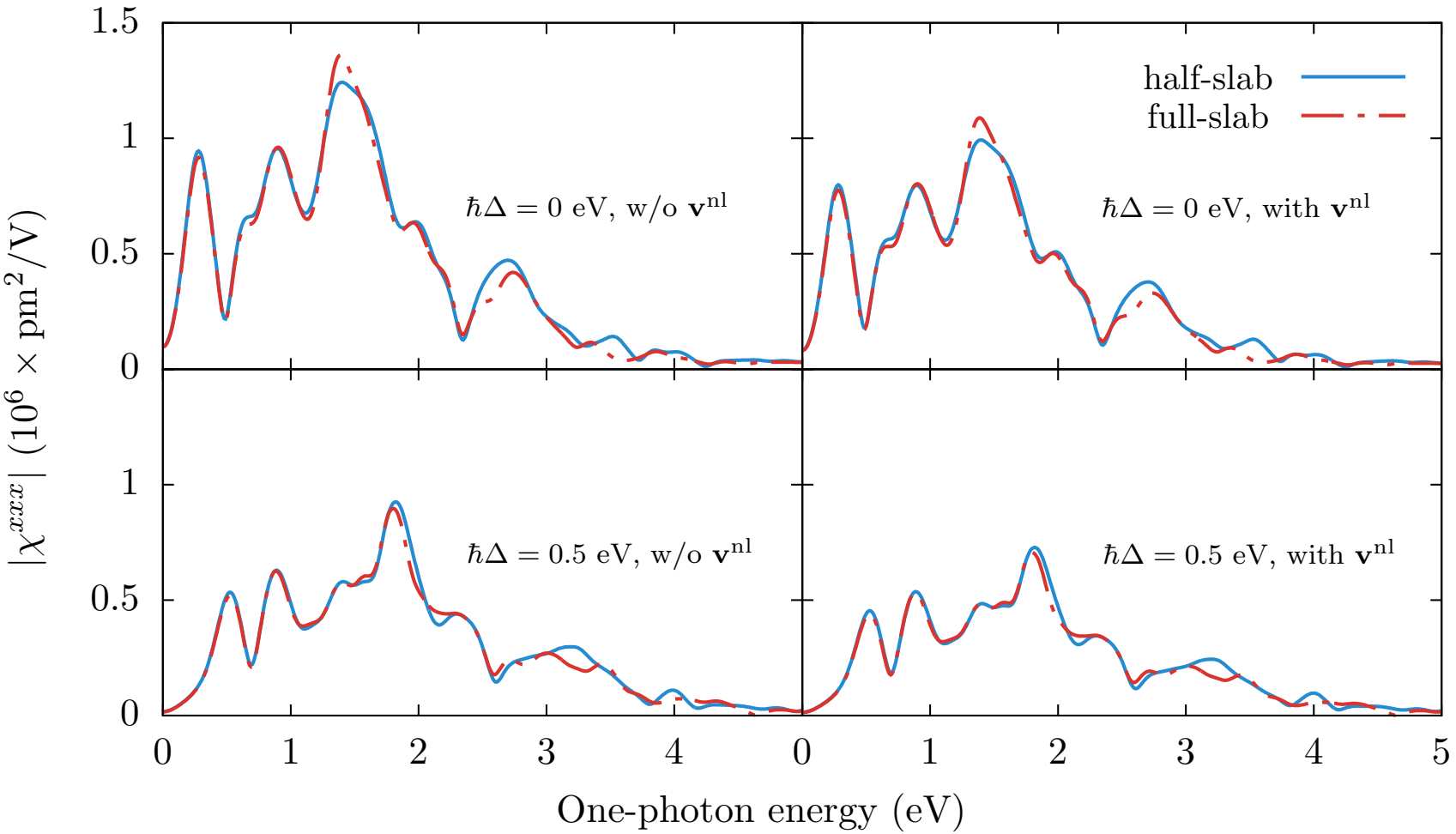

1ω
2ω
2ω
z
R
Surface
χ^abc
d
χ^abcd
Centrosymmetric Bulk

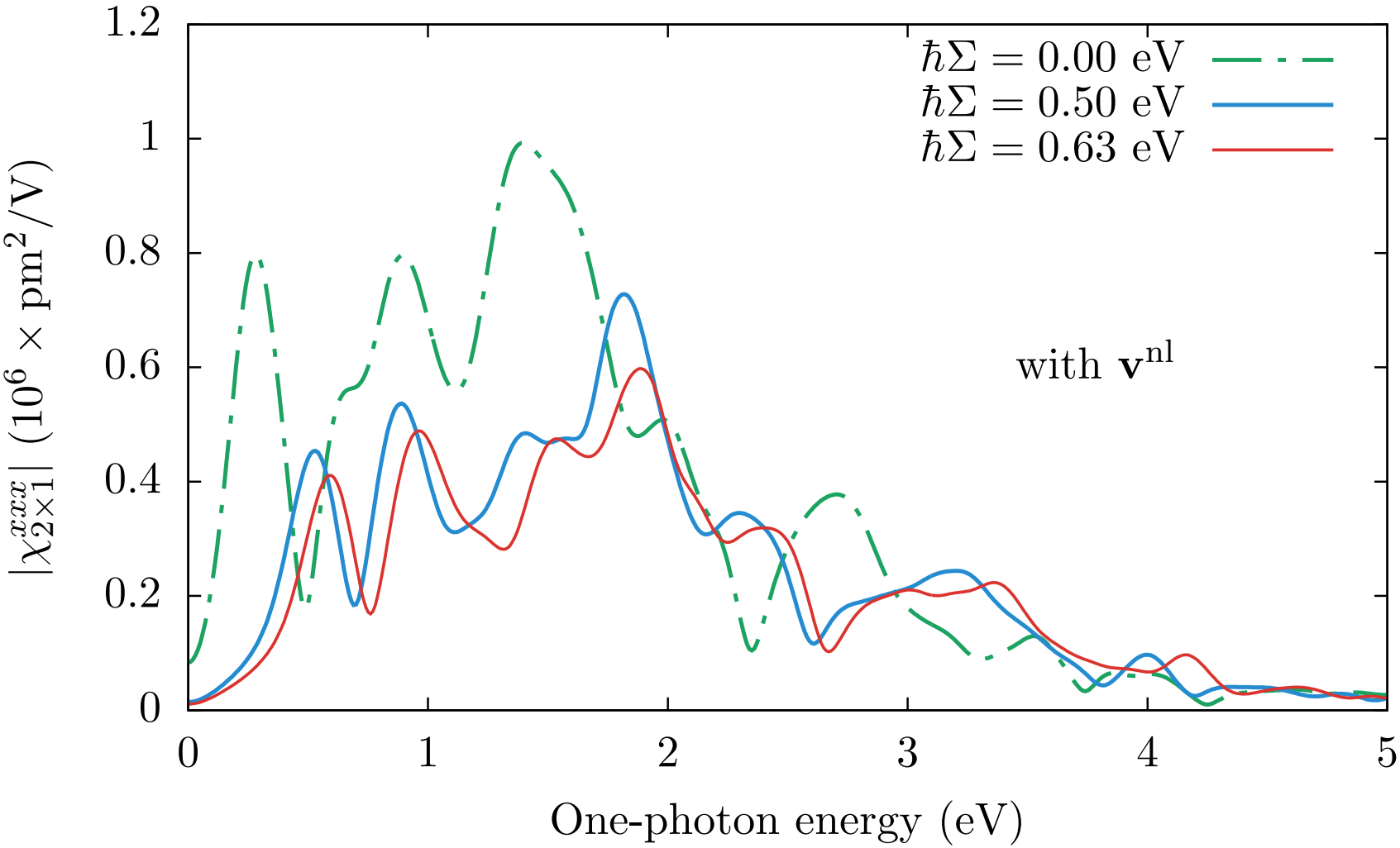

z
R
Front Surface
Vacuum Region
Surface Region ∼ d
Bulk Region
z_ℓ
Δ_ℓ^f
Δ_ℓ^b
ℓth Layer
Back Surface

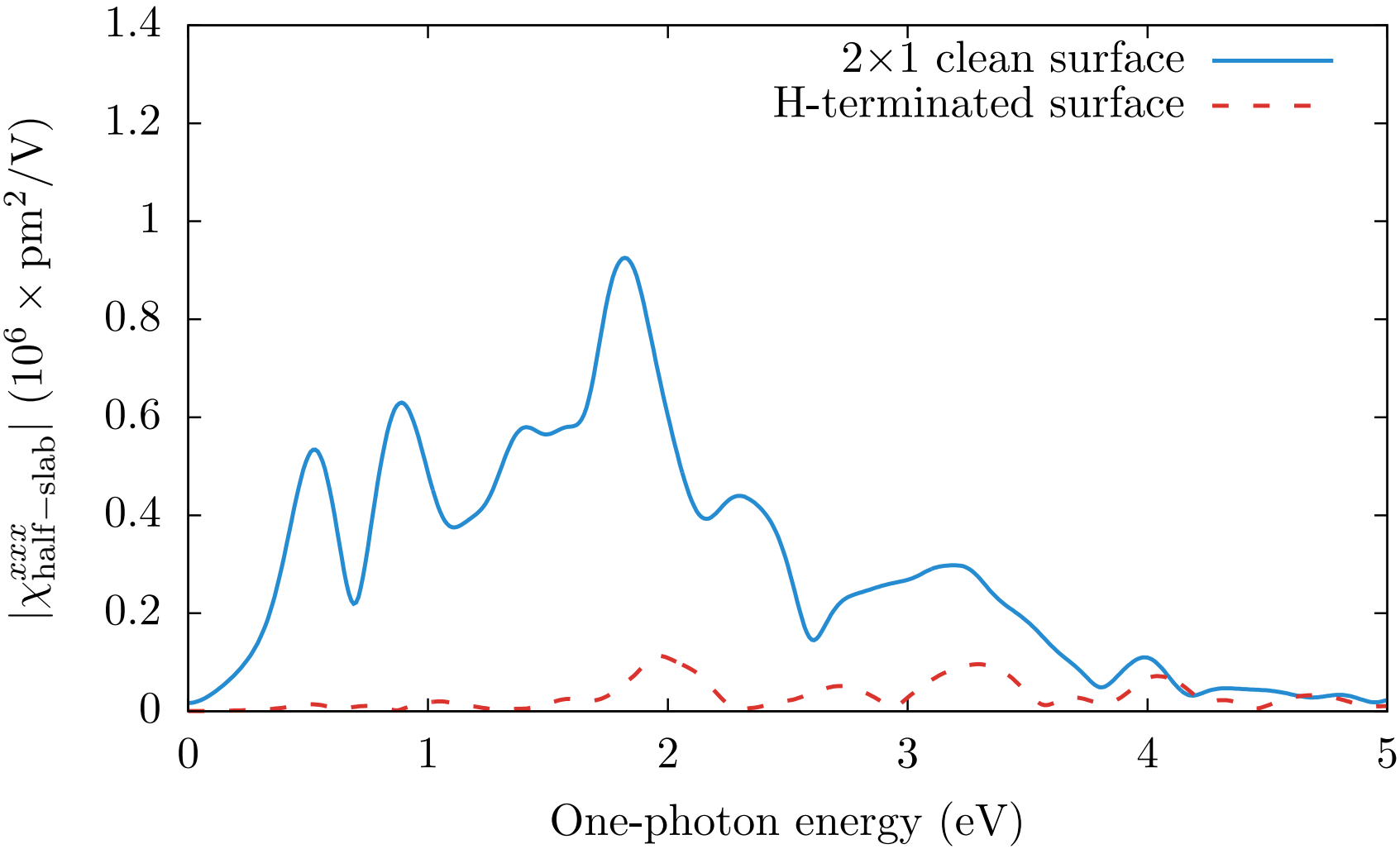
$2{\times}1$ reconstruction $\Rightarrow \chi^{xxx}_{2\times1} \neq 0$

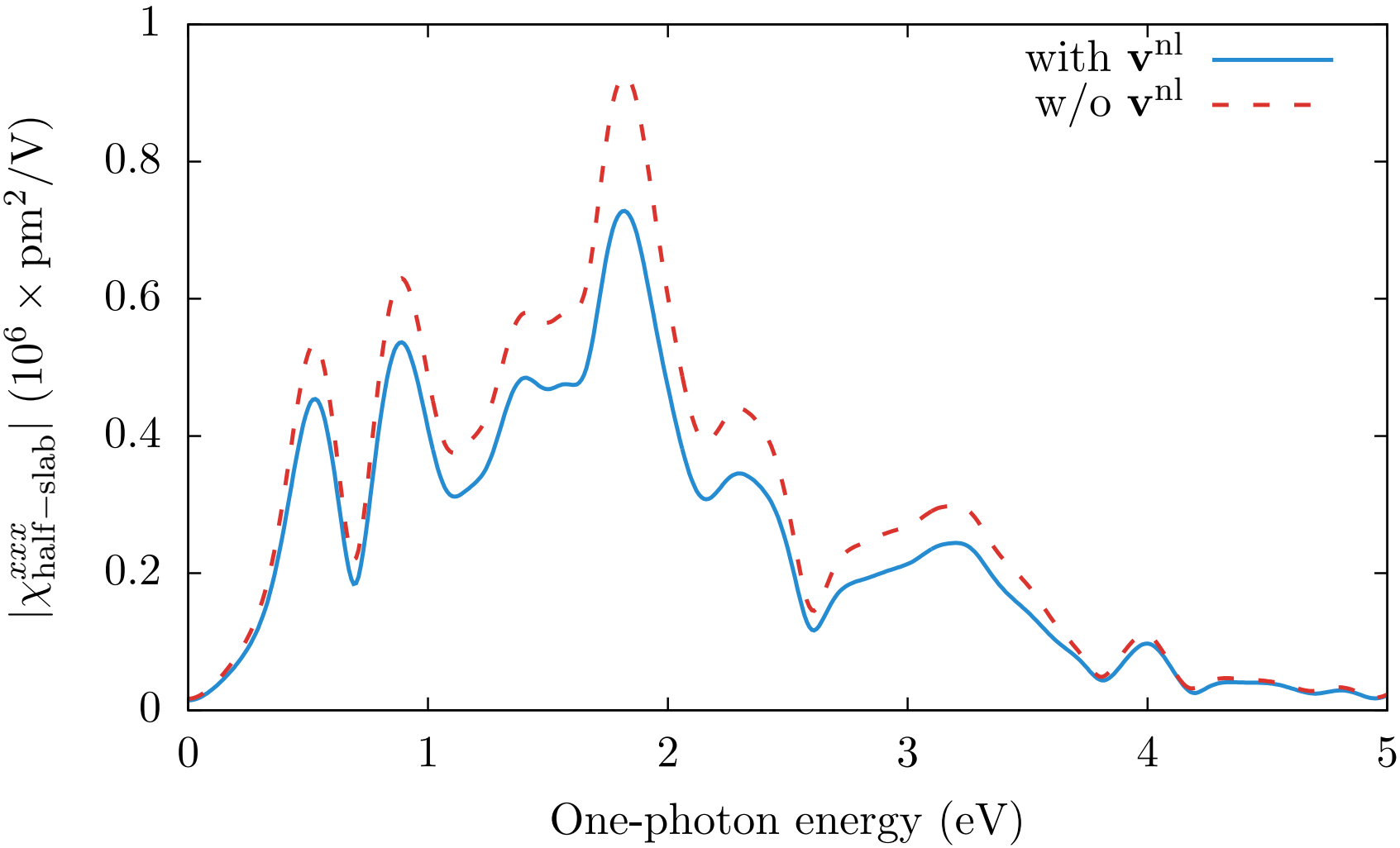


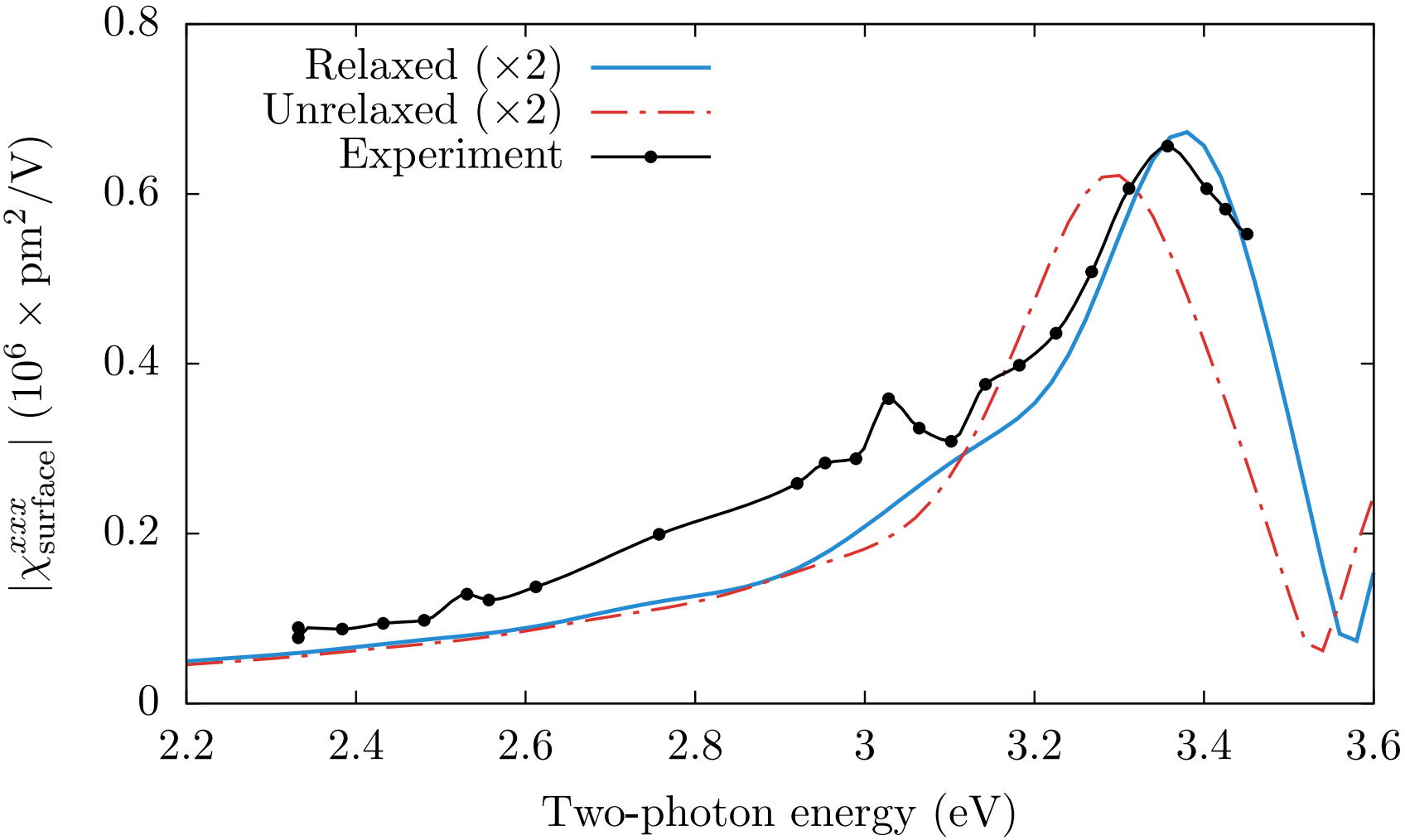
H-terminated $\Rightarrow \chi^{xxx}_{\mathrm{H}} = 0$

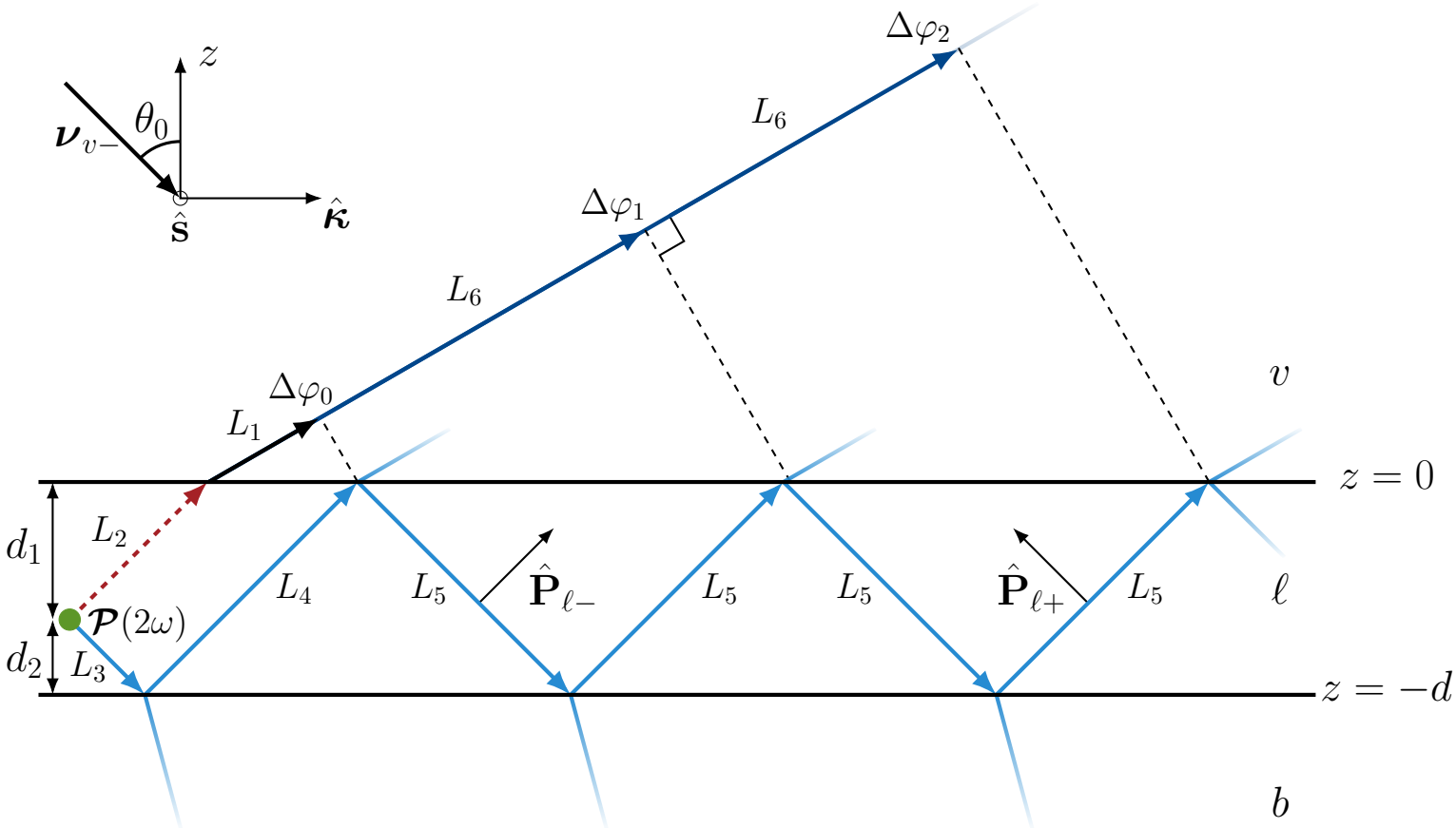

half-slab
full-slab
$\hbar\Delta = 0$ eV, w/o $\mathbf{v}^{\mathrm{nl}}$
$\hbar\Delta = 0$ eV, with $\mathbf{v}^{\mathrm{nl}}$
$\hbar\Delta = 0.5$ eV, w/o $\mathbf{v}^{\mathrm{nl}}$
$\hbar\Delta = 0.5$ eV, with $\mathbf{v}^{\mathrm{nl}}$
$|\chi^{xxx}|$ $(10^6 \times \mathrm{pm}^2/\mathrm{V})$
One-photon energy (eV)
0
0.5
1
1.5
0
1
2
3
4
5

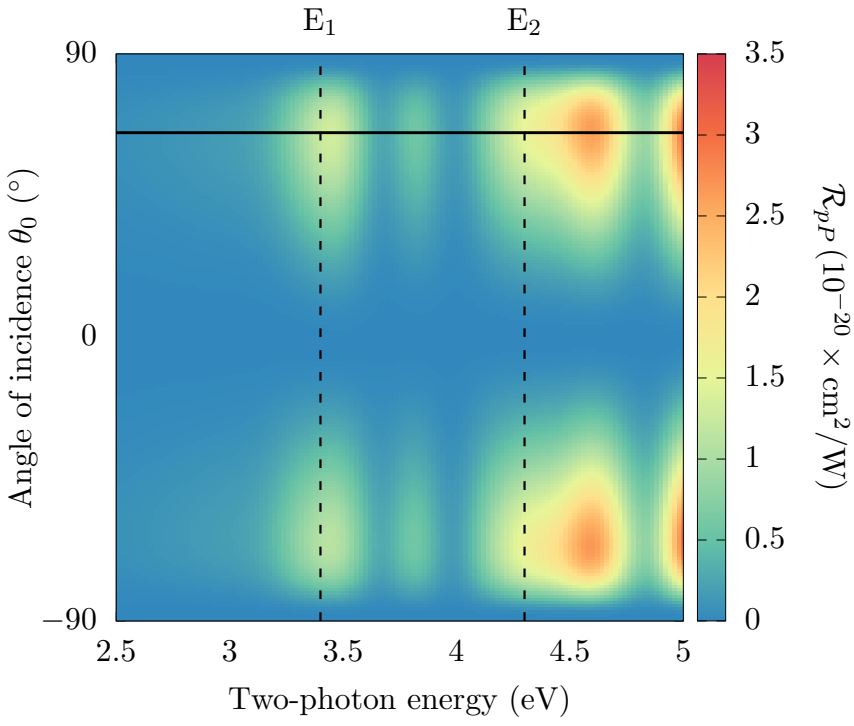

1.2
1
0.8
0.6
0.4
0.2
0
0
1
2
3
4
5
$|\chi^{xxx}_{2\times1}|$ ($10^6 \times$ pm^2/V)
One-photon energy (eV)
$\hbar\Sigma = 0.00$ eV
$\hbar\Sigma = 0.50$ eV
$\hbar\Sigma = 0.63$ eV
with $\mathbf{v}^{\mathrm{nl}}$

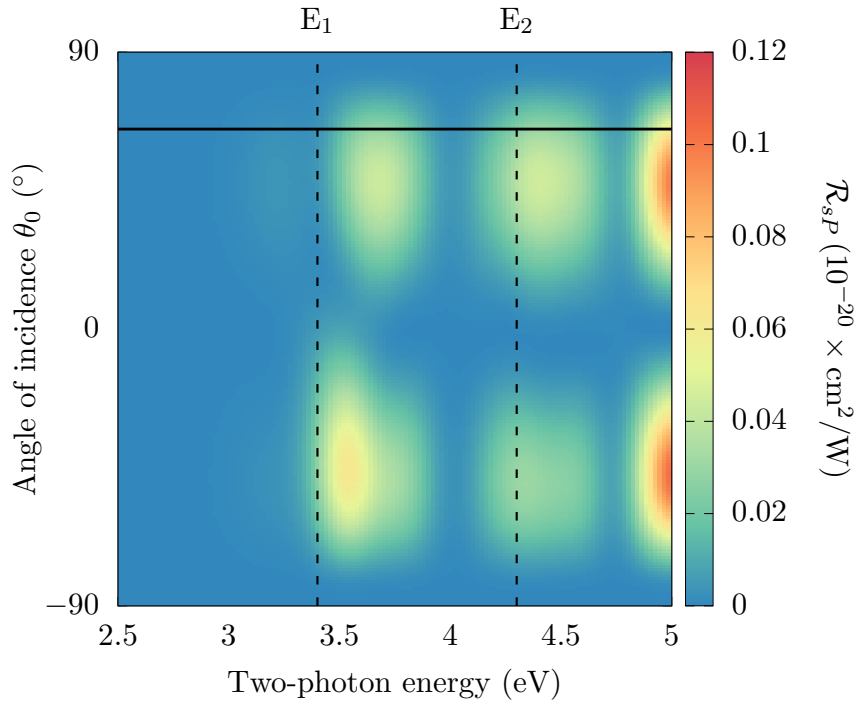

2×1 clean surface
H-terminated surface
$|\chi^{xxx}_{\mathrm{half-slab}}|$ ($10^6 \times \mathrm{pm}^2/\mathrm{V}$)
One-photon energy (eV)
0
0.2
0.4
0.6
0.8
1
1.2
1.4
0
1
2
3
4
5

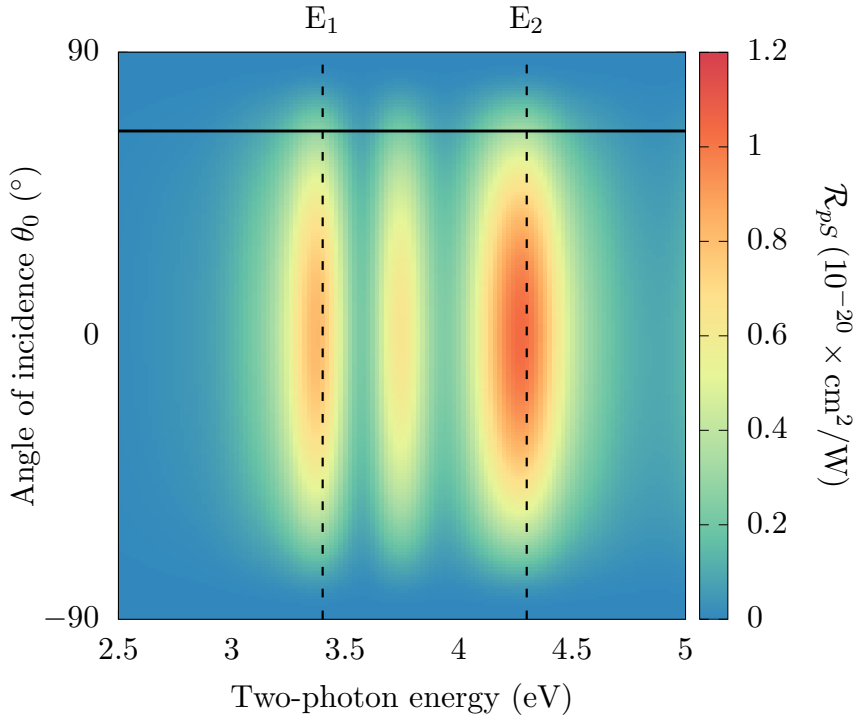

with $\mathbf{v}^{\mathrm{nl}}$
w/o $\mathbf{v}^{\mathrm{nl}}$
$|\chi^{xxx}_{\mathrm{half-slab}}|$ $(10^6 \times \mathrm{pm}^2/\mathrm{V})$
One-photon energy (eV)
0
0.2
0.4
0.6
0.8
1
0
1
2
3
4
5

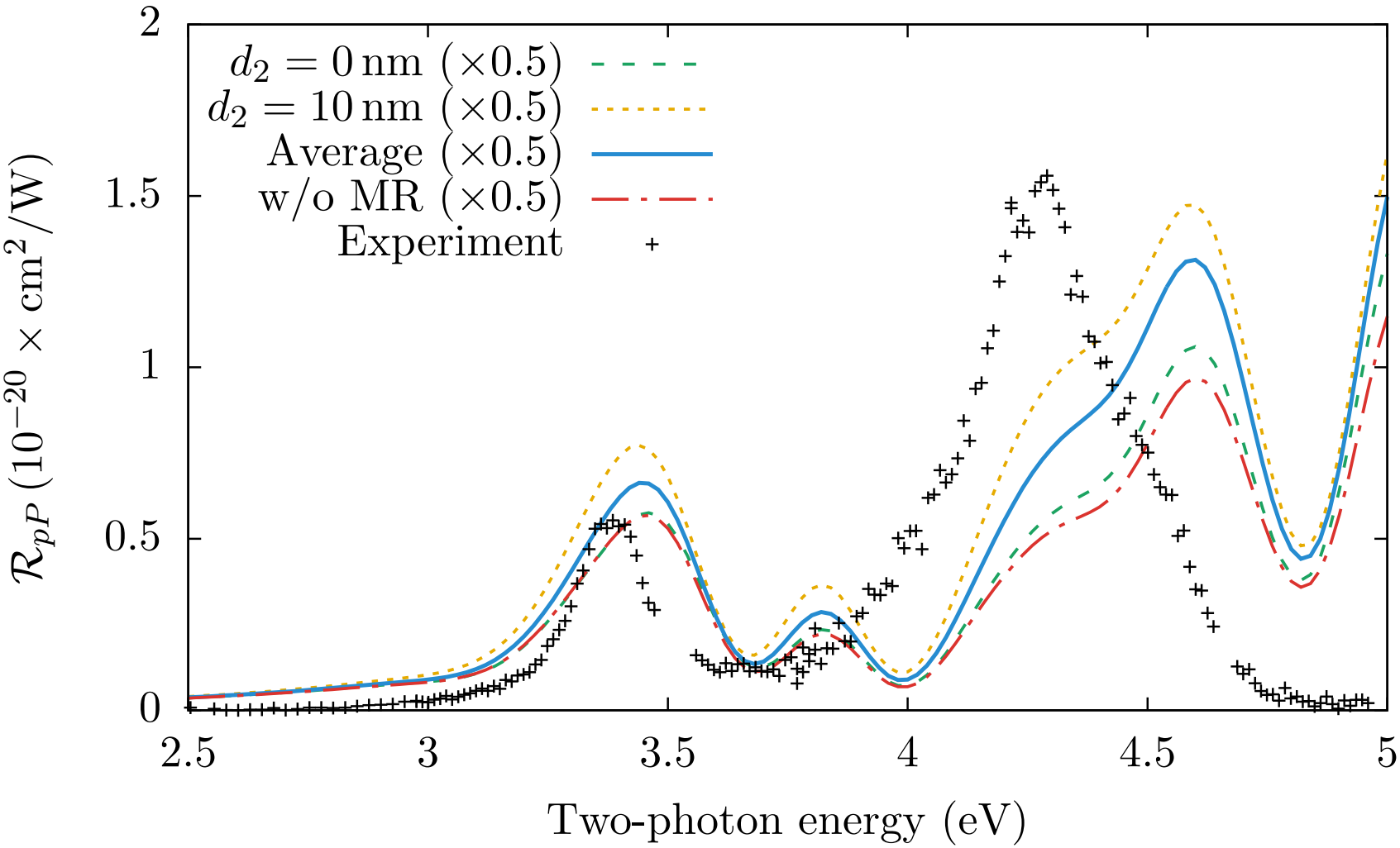

Relaxed (×2)
Unrelaxed (×2)
Experiment
$|\chi^{xxx}_{\text{surface}}|$ ($10^6 \times \text{pm}^2/\text{V}$)
Two-photon energy (eV)
0
0.2
0.4
0.6
0.8
2.2
2.4
2.6
2.8
3
3.2
3.4
3.6

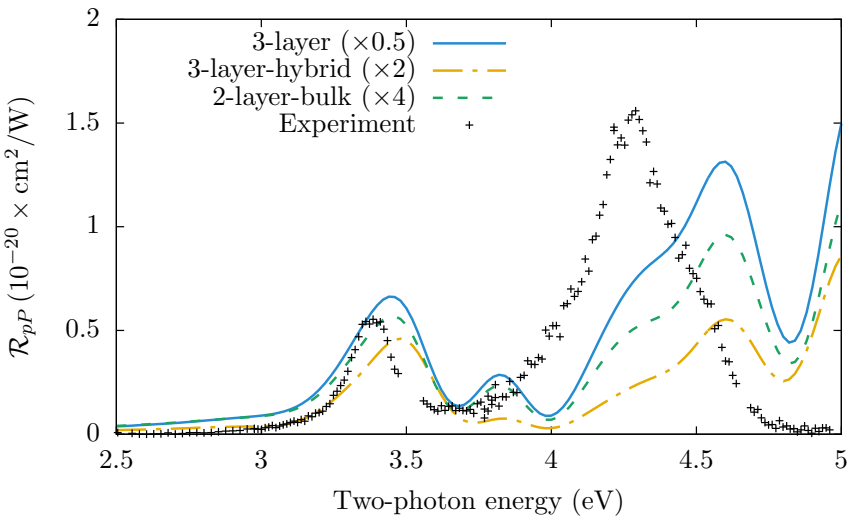

z
θ_0
$\boldsymbol{\nu}_{v-}$
$\hat{\mathbf{s}}$
$\hat{\boldsymbol{\kappa}}$
$\Delta\varphi_2$
L_6
$\Delta\varphi_1$
L_6
$\Delta\varphi_0$
L_1
v
$z = 0$
d_1
L_2
L_4
L_5
$\hat{\mathbf{P}}_{\ell-}$
L_5
L_5
$\hat{\mathbf{P}}_{\ell+}$
L_5
ℓ
$\boldsymbol{\mathcal{P}}(2\omega)$
d_2
L_3
$z = -d$
b

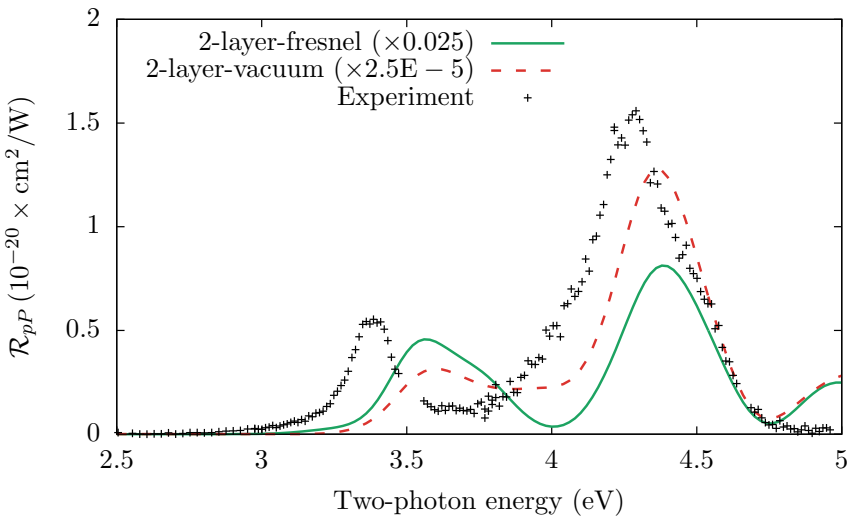

E1
E2
90
0
−90
Angle of incidence θ0 (°)
2.5
3
3.5
4
4.5
5
Two-photon energy (eV)
3.5
3
2.5
2
1.5
1
0.5
0
RpP (10−20 × cm2/W)

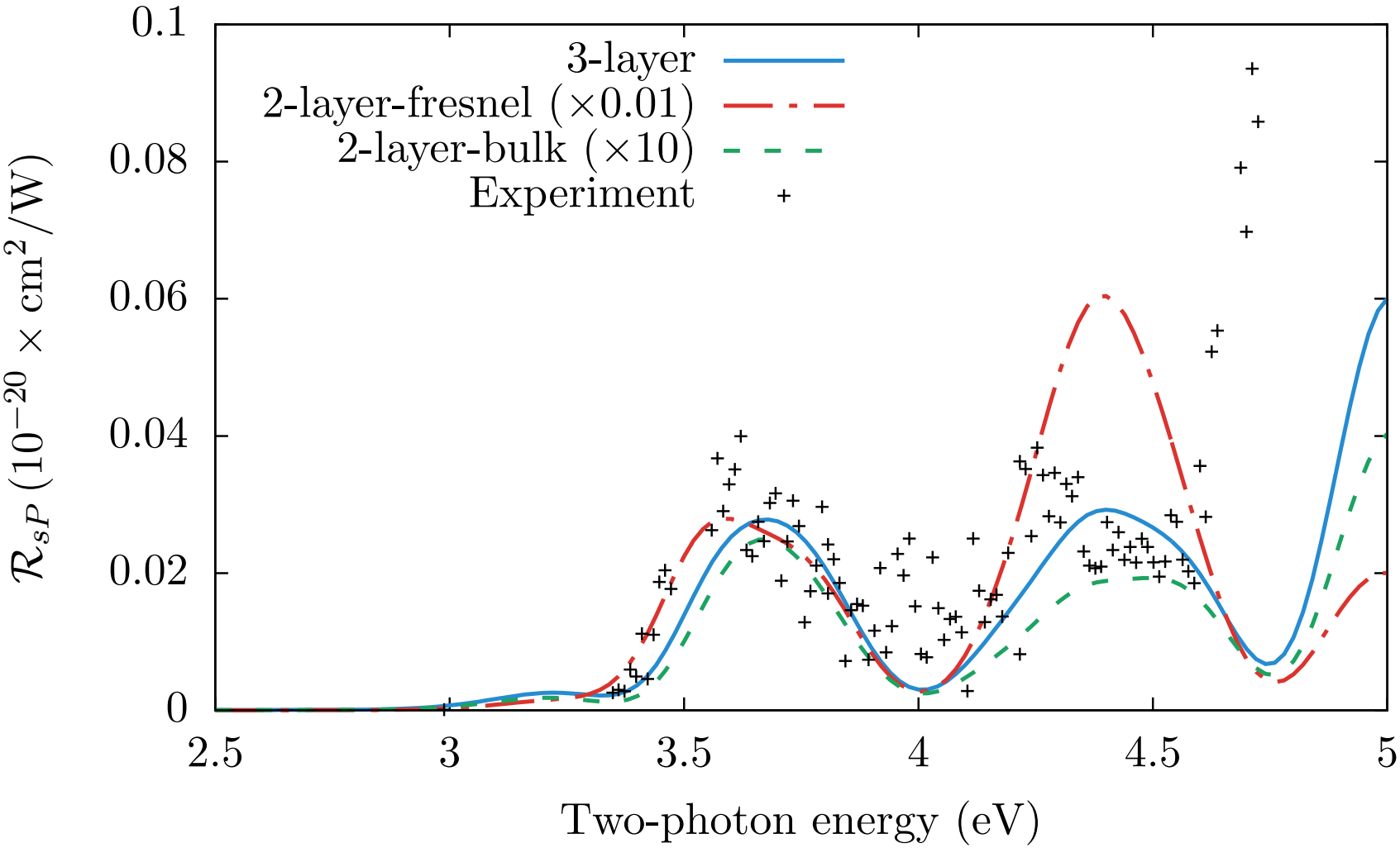

E1
E2
90
0
−90
Angle of incidence θ0 (°)
2.5
3
3.5
4
4.5
5
Two-photon energy (eV)
0.12
0.1
0.08
0.06
0.04
0.02
0
RsP (10−20 × cm2/W)

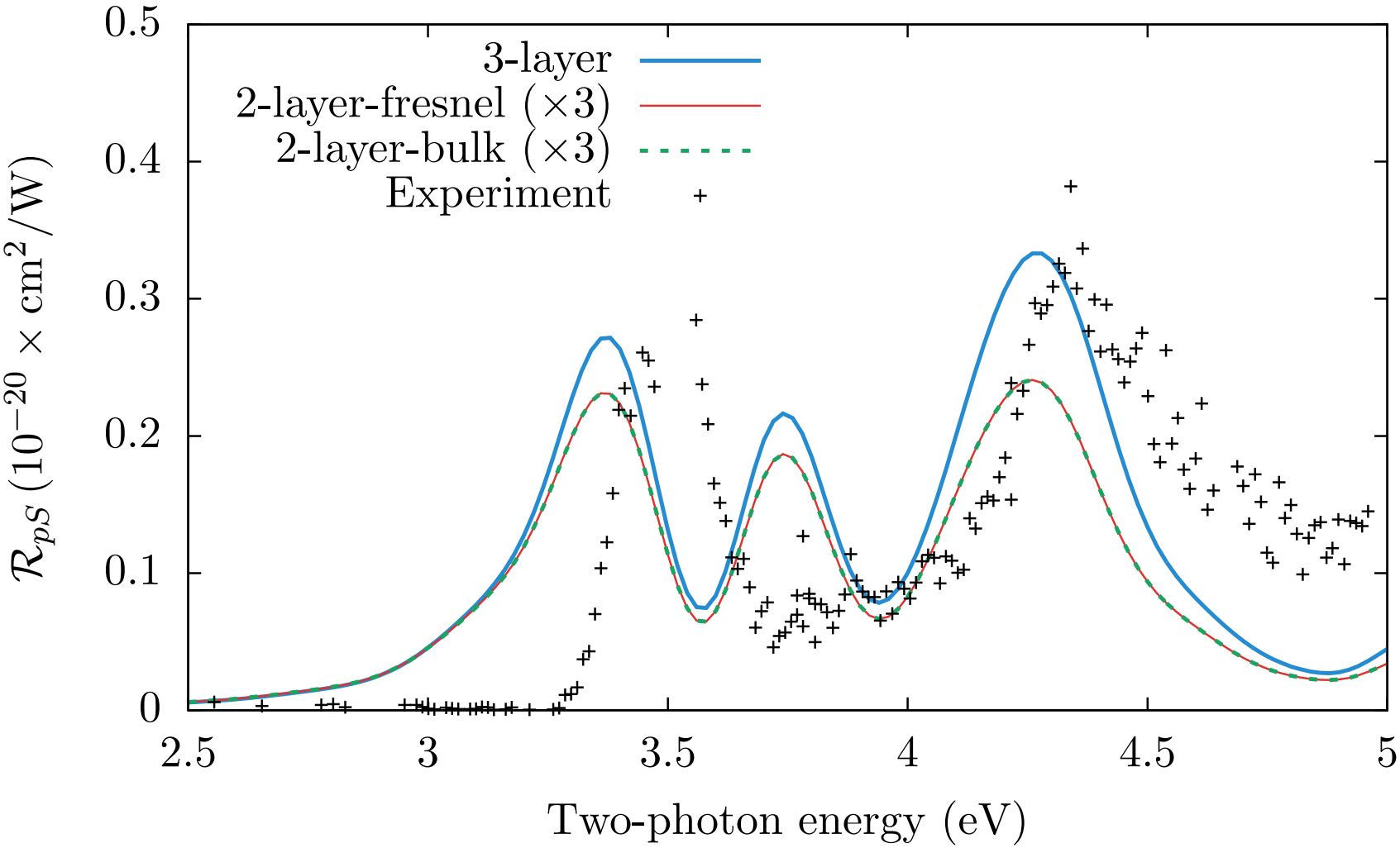

E1
E2
90
0
−90
Angle of incidence θ0 (°)
2.5
3
3.5
4
4.5
5
Two-photon energy (eV)
1.2
1
0.8
0.6
0.4
0.2
0
RpS (10−20 × cm2/W)


$d_2 = 0\,\mathrm{nm}$ (×0.5)
$d_2 = 10\,\mathrm{nm}$ (×0.5)
Average (×0.5)
w/o MR (×0.5)
Experiment
$\mathcal{R}_{pP}$ (10^{-20} × cm^2/W)
Two-photon energy (eV)
0
0.5
1
1.5
2
2.5
3
3.5
4
4.5
5


3-layer (×0.5)
3-layer-hybrid (×2)
2-layer-bulk (×4)
Experiment
$\mathcal{R}_{pP}\,(10^{-20}\times\mathrm{cm}^2/\mathrm{W})$
Two-photon energy (eV)
0
0.5
1
1.5
2
2.5
3
3.5
4
4.5
5


2-layer-fresnel (×0.025)
2-layer-vacuum (×2.5E − 5)
Experiment
$\mathcal{R}_{pP}\,(10^{-20}\times\mathrm{cm}^2/\mathrm{W})$
Two-photon energy (eV)
0
0.5
1
1.5
2
2.5
3
3.5
4
4.5
5


3-layer
2-layer-fresnel (×0.01)
2-layer-bulk (×10)
Experiment
$\mathcal{R}_{sP}$ (10^{-20} × cm^2/W)
0.1
0.08
0.06
0.04
0.02
0
2.5
3
3.5
4
4.5
5
Two-photon energy (eV)


3-layer
2-layer-fresnel (×3)
2-layer-bulk (×3)
Experiment
$\mathcal{R}_{pS}$ (10^{-20} × cm^2/W)
Two-photon energy (eV)
0
0.1
0.2
0.3
0.4
0.5
2.5
3
3.5
4
4.5
5

Title

- Item 1
- Item 2
- Item 3

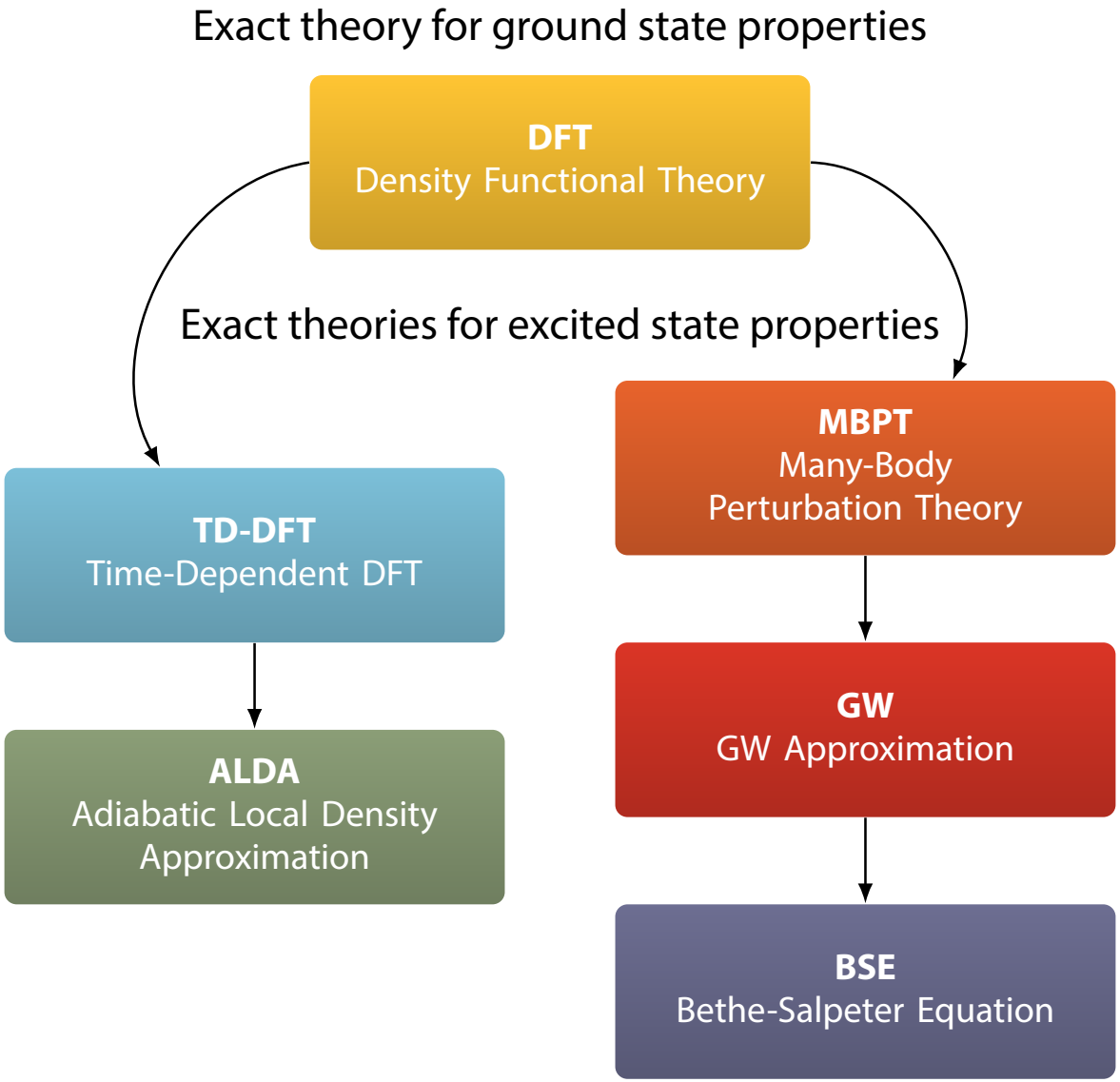
Some starting text,

$$E = mc^2, \tag{1}$$

and more text.

A block

- Item 1
- Item 2


Exact theory for ground state properties
DFT
Density Functional Theory
Exact theories for excited state properties
TD-DFT
Time-Dependent DFT
ALDA
Adiabatic Local Density Approximation
MBPT
Many-Body Perturbation Theory
GW
GW Approximation
BSE
Bethe-Salpeter Equation