Ab initio TDDFT study of high-harmonic generation from dielectric thin films

December 12, 2022

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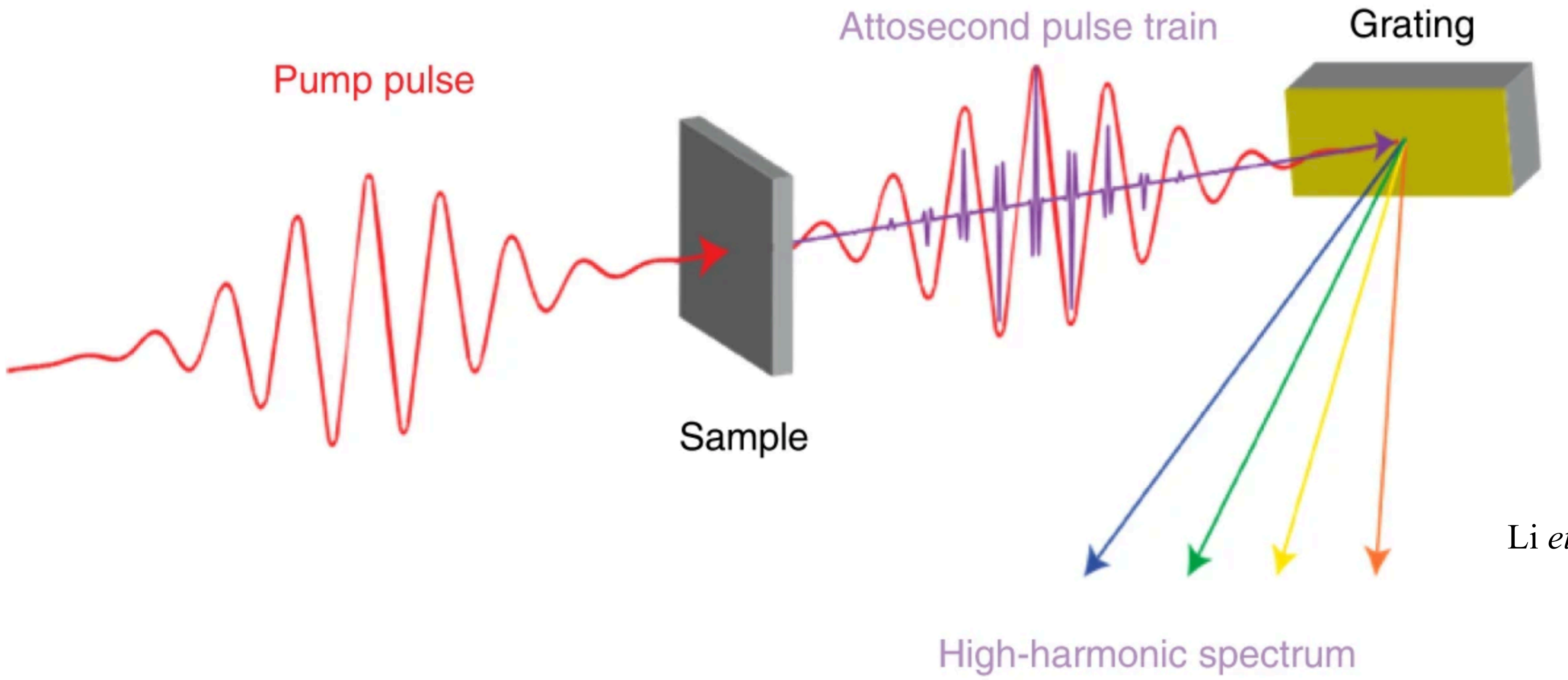
High-order Harmonic Generation (HHG) of solid materials



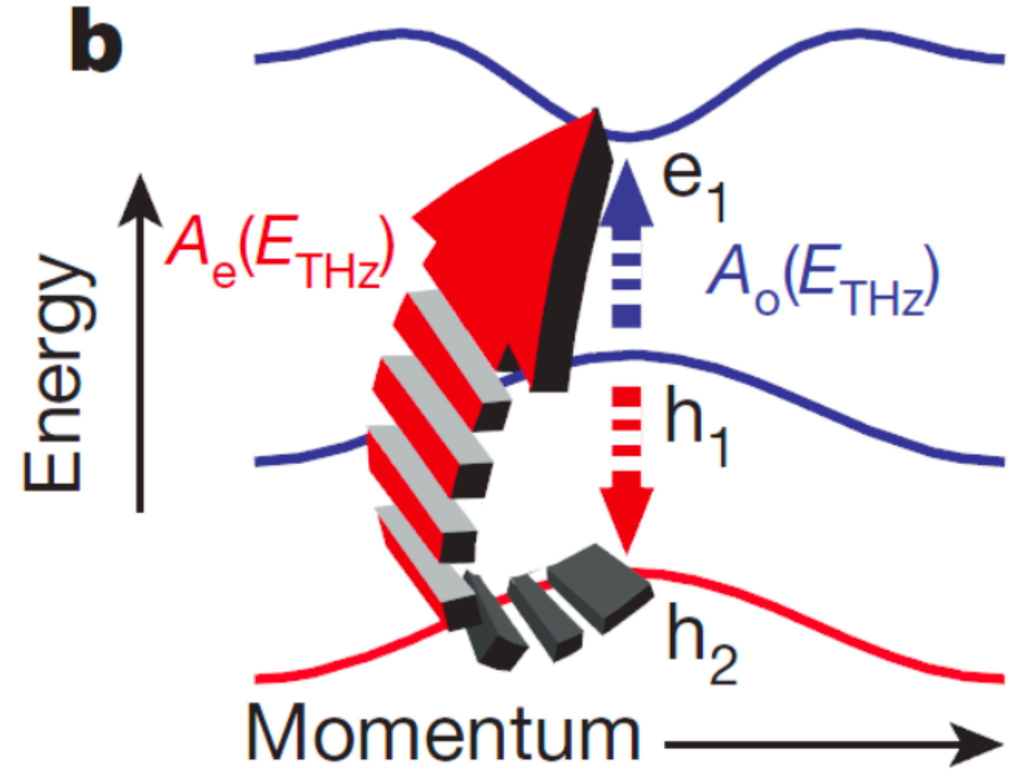
Li *et al.*, Nat. Commun. **11**, 2748 (2020).

- Under irradiation of a strong light pulse, a target (such as gas) emits light pulses including high-order harmonics of the fundamental wave.
- HHG is widely used as a coherent light source and it has many applications.
- HHG of solid materials is one of the hot topics in optical science, because of its improved harmonic intensities offered by the higher atomic densities of solids compared to gases or liquids.

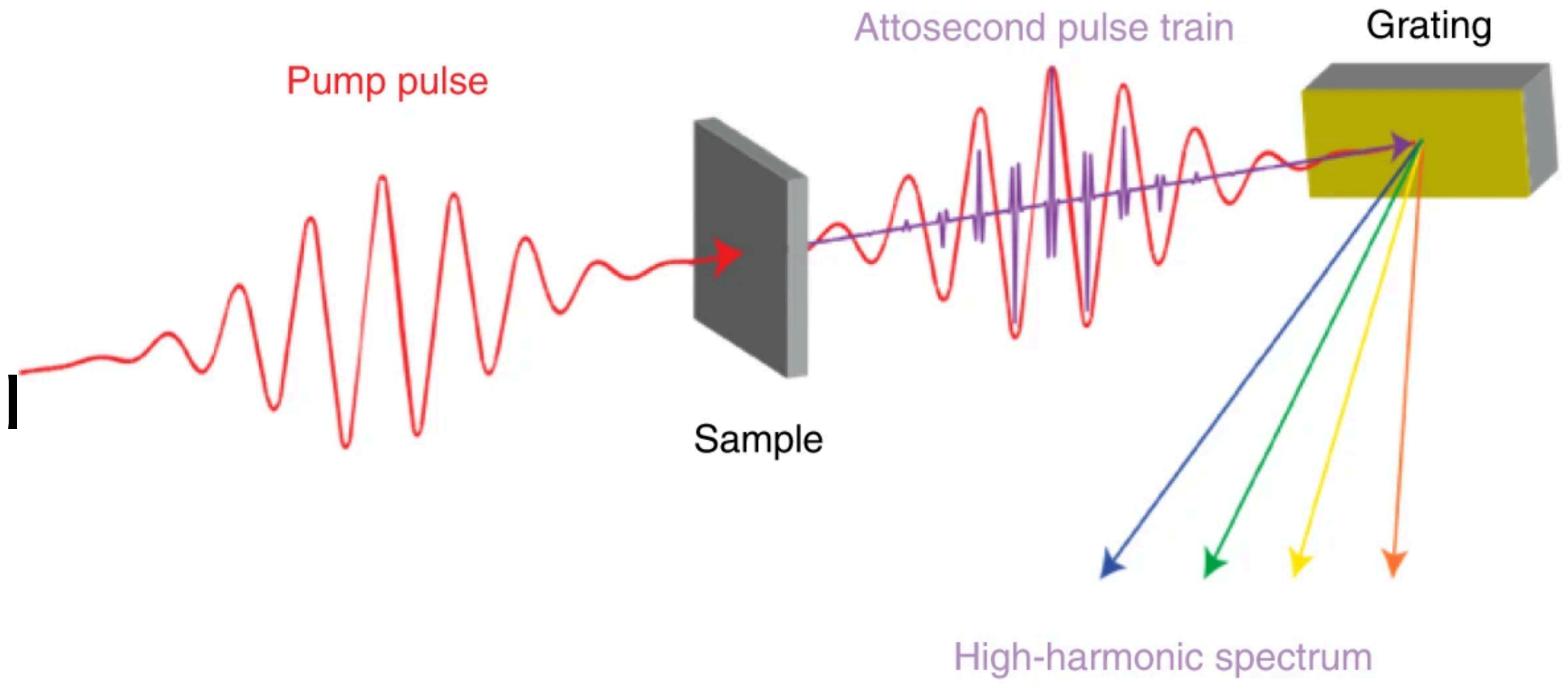
Theories for HHG in solids: Previous studies & Our study

- HHG is highly nonlinear phenomena → electron motion in materials is essential → Real-Time TDDFT study
- Most of the previous TDDFT studies for HHG in solids were limited to microscopic origin in a bulk system. The effect of light propagation is usually omitted.
- However, actual HHG spectra observed by experiments should be strongly modulated by the nonlinear light propagation effect.
- Question: what is the impact of light propagation on HHG in reflected & transmitted waves from solid thin films?
- To elucidate this effect, we use a combined method of Maxwell eqs. for light propagation & TDDFT for electron motion in solids.

Inter- or intra-band motion in bulk



Hohenleutner *et al.* Nature **523**, 572 (2015)



Li *et al.*, Nat. Commun. **11**, 2748 (2020).

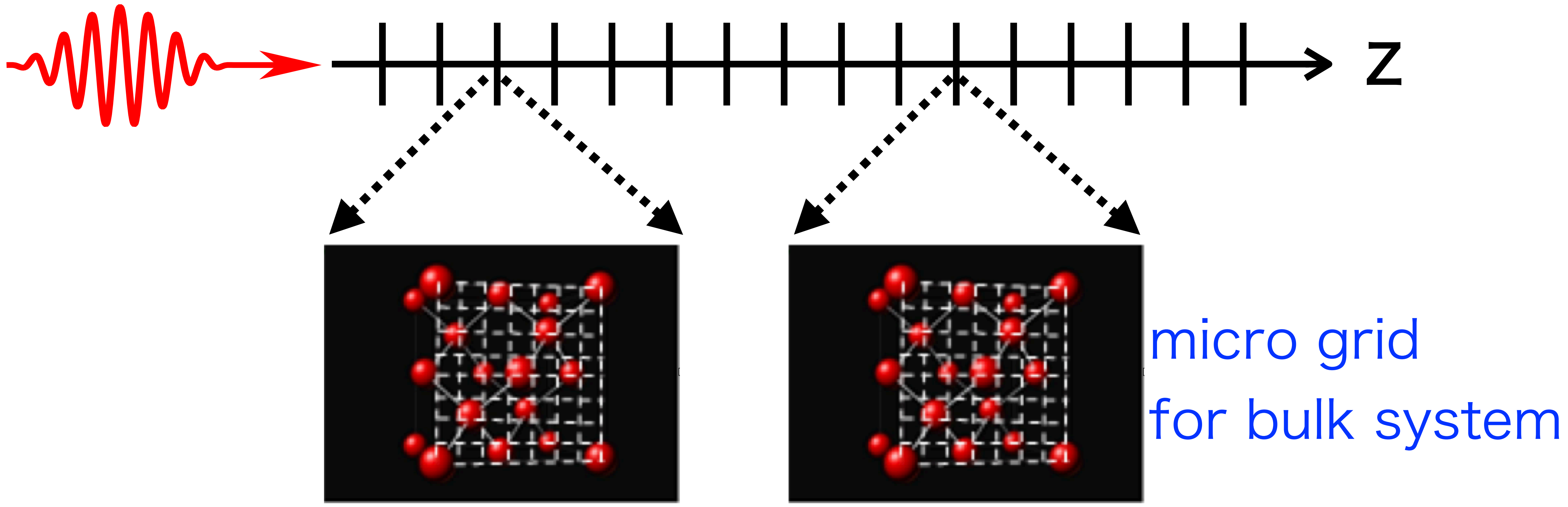
Method: Multiscale Maxwell-TDDFT

- For considering strongly coupled dynamics of light & electrons, we solve Maxwell eqs. (light) & TD Kohn-Sham eq (electrons) simultaneously
- Propagation of EM field is solved on a macroscopic grid.
- At each macro point inside a material, a bulk system for TDDFT is prepared.
- These TDKS systems are solved by parallel computation.

K. Yabana *et al.*, PRB **85**, 045134 (2012).

$$\left(\frac{1}{c^2}\frac{\partial^2}{\partial t^2}-\frac{\partial^2}{\partial Z^2}\right)\mathbf{A}_Z(t)=\frac{4\pi}{c}\mathbf{J}_Z(t)$$

macro grid



$$i\frac{\partial}{\partial t}u_{n\mathbf{k},Z}=\hat{H}_{\mathbf{k}+\frac{1}{c}A_Z(t)}u_{n\mathbf{k},Z}$$

$$\mathbf{J}_Z(t)=\int_\Omega\frac{d^3r}{\Omega}\sum_{n\mathbf{k}}^{\mathrm{occ}}u^*_{n\mathbf{k},Z}\left[i\mathbf{r},\hat{H}_{\mathbf{k}+\mathbf{A}_Z(t)/c}\right]u_{n\mathbf{k},Z}$$

$$i\frac{\partial}{\partial t}u_{n\mathbf{k},Z}=\hat{H}_{\mathbf{k}+\frac{1}{c}A_Z(t)}u_{n\mathbf{k},Z}$$

$$\mathbf{J}_Z(t)=\int_\Omega\frac{d^3r}{\Omega}\sum_{n\mathbf{k}}^{\mathrm{occ}}u^*_{n\mathbf{k},Z}\left[i\mathbf{r},\hat{H}_{\mathbf{k}+\mathbf{A}_Z(t)/c}\right]u_{n\mathbf{k},Z}$$

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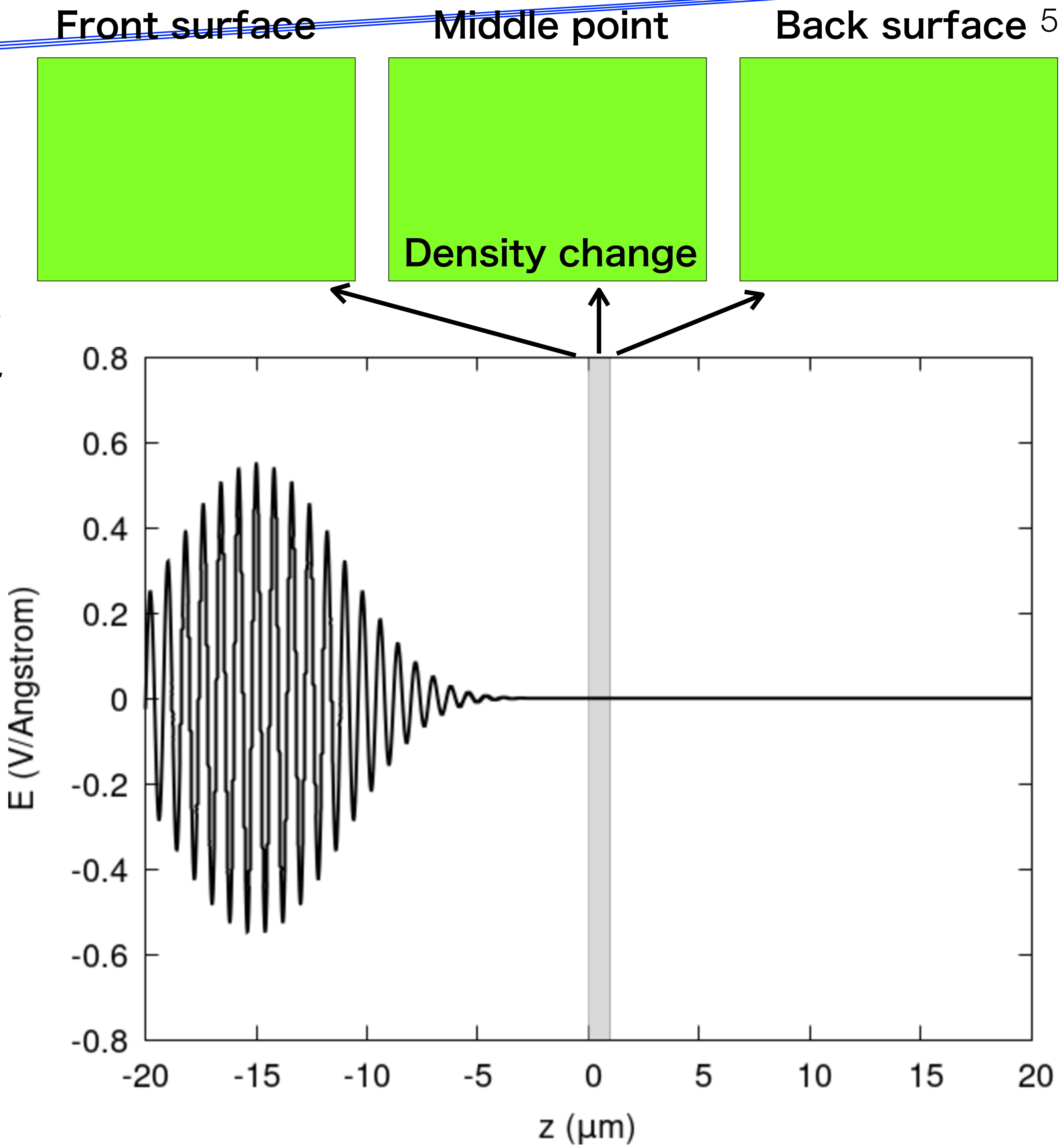
Method: Multiscale Maxwell-TDDFT

- Laser-irradiated Si thin film (thickness 1 μm)
- 1 μm Si film is represented by 160 macro points
- For each macro point, a bulk Si system for TDDFT is attached.

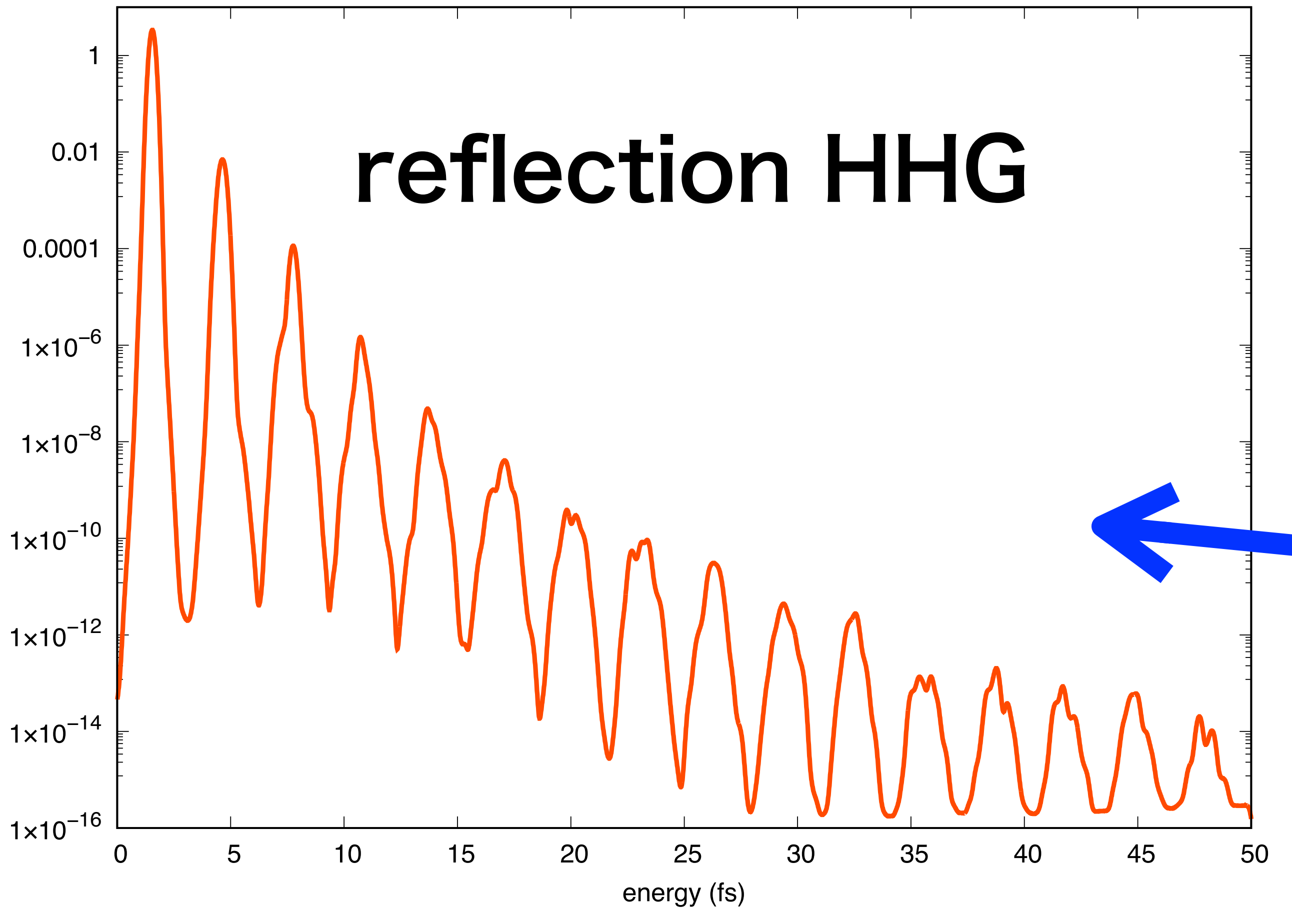
(Calculation conditions)

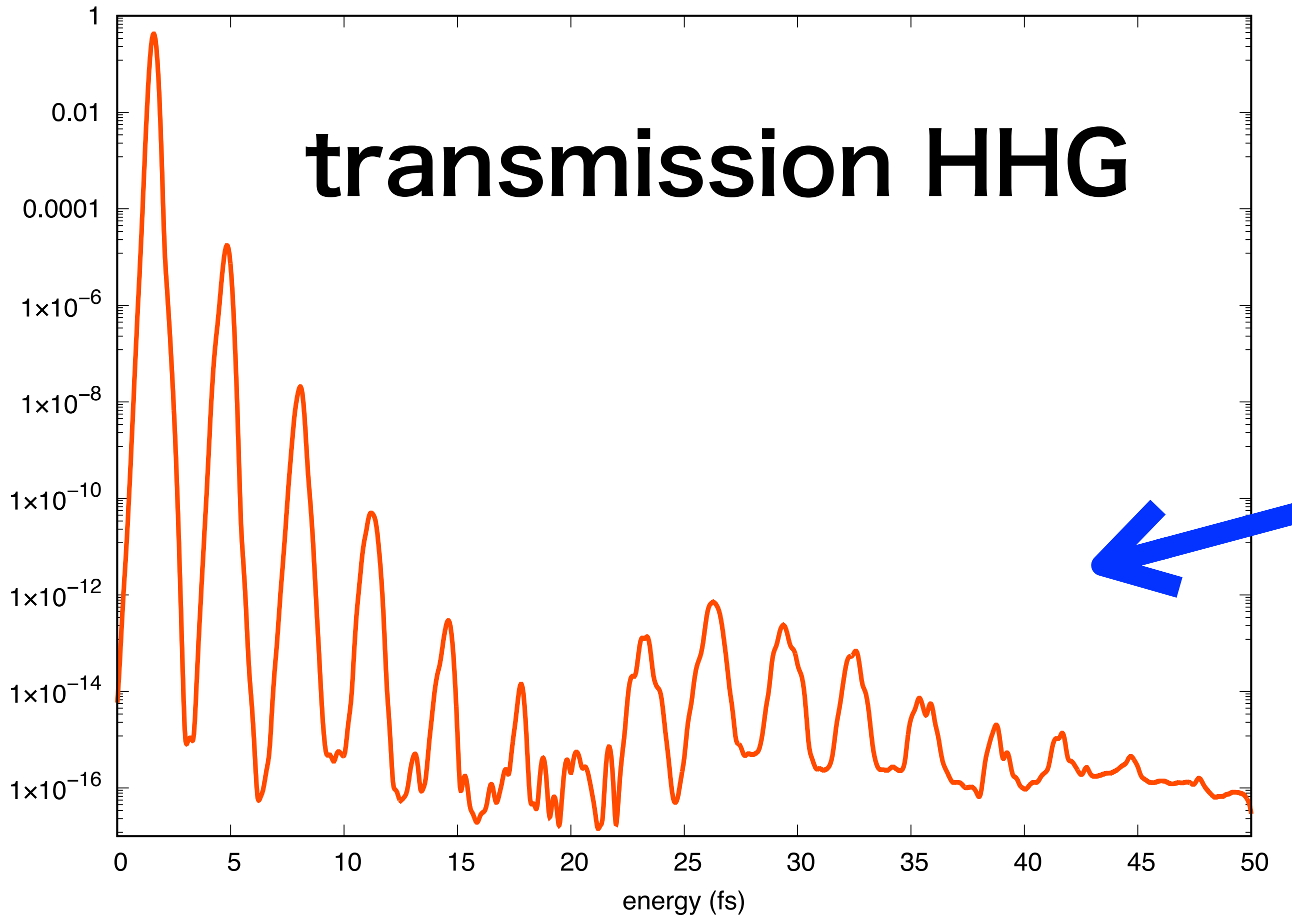
- SALMON-TDDFT code
- Wavelength 800 nm ($\hbar\omega$ = 1.55 eV)
- Intensity I = 4×10^{12} W/cm^2
- Total pulse duration T= 100 fs
- Nr = 16^3, Nk = 32^3, dt = 0.0025 fs
- ALDA, norm-conserving pp.

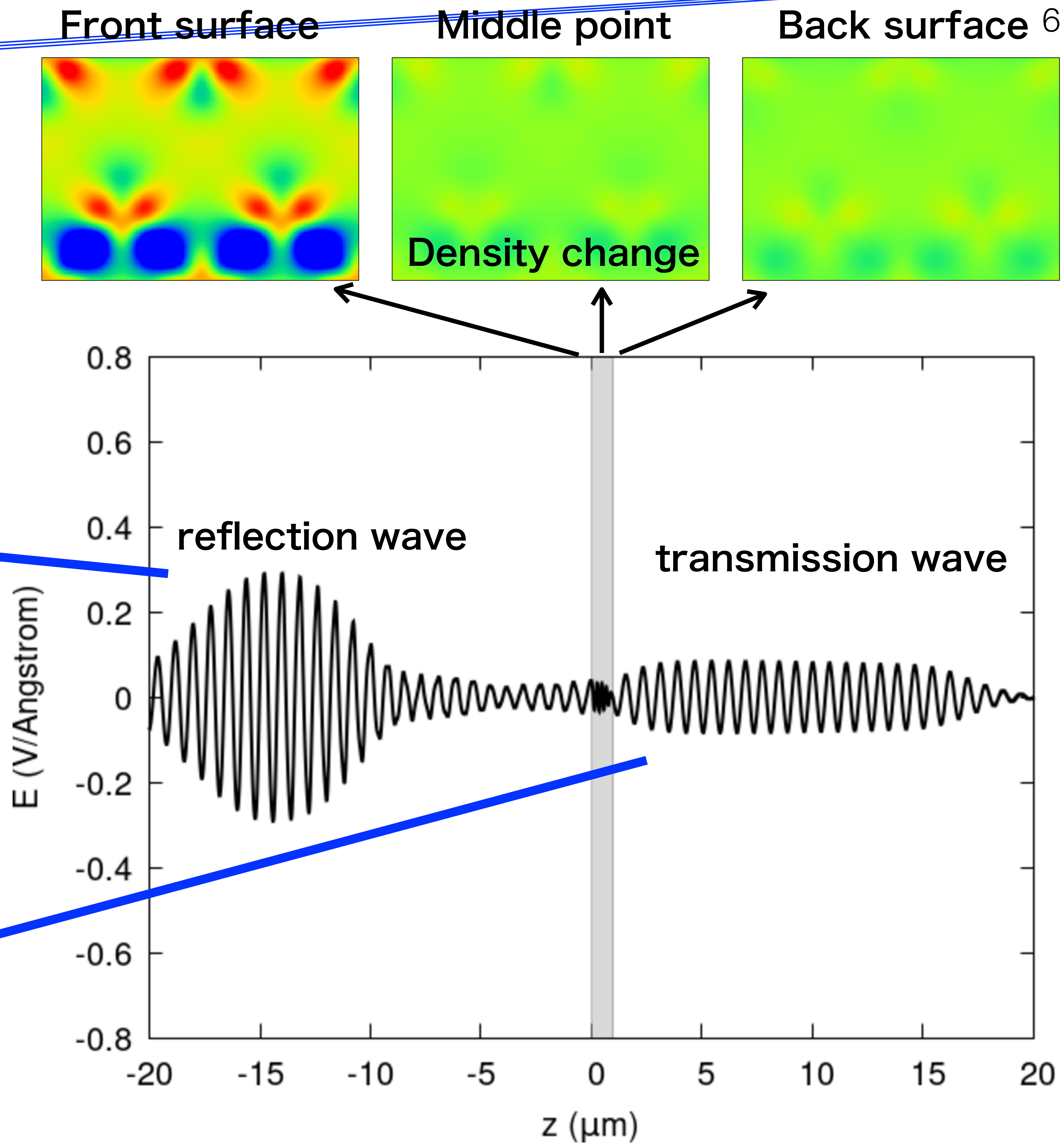


Method: Multiscale Maxwell-TDDFT

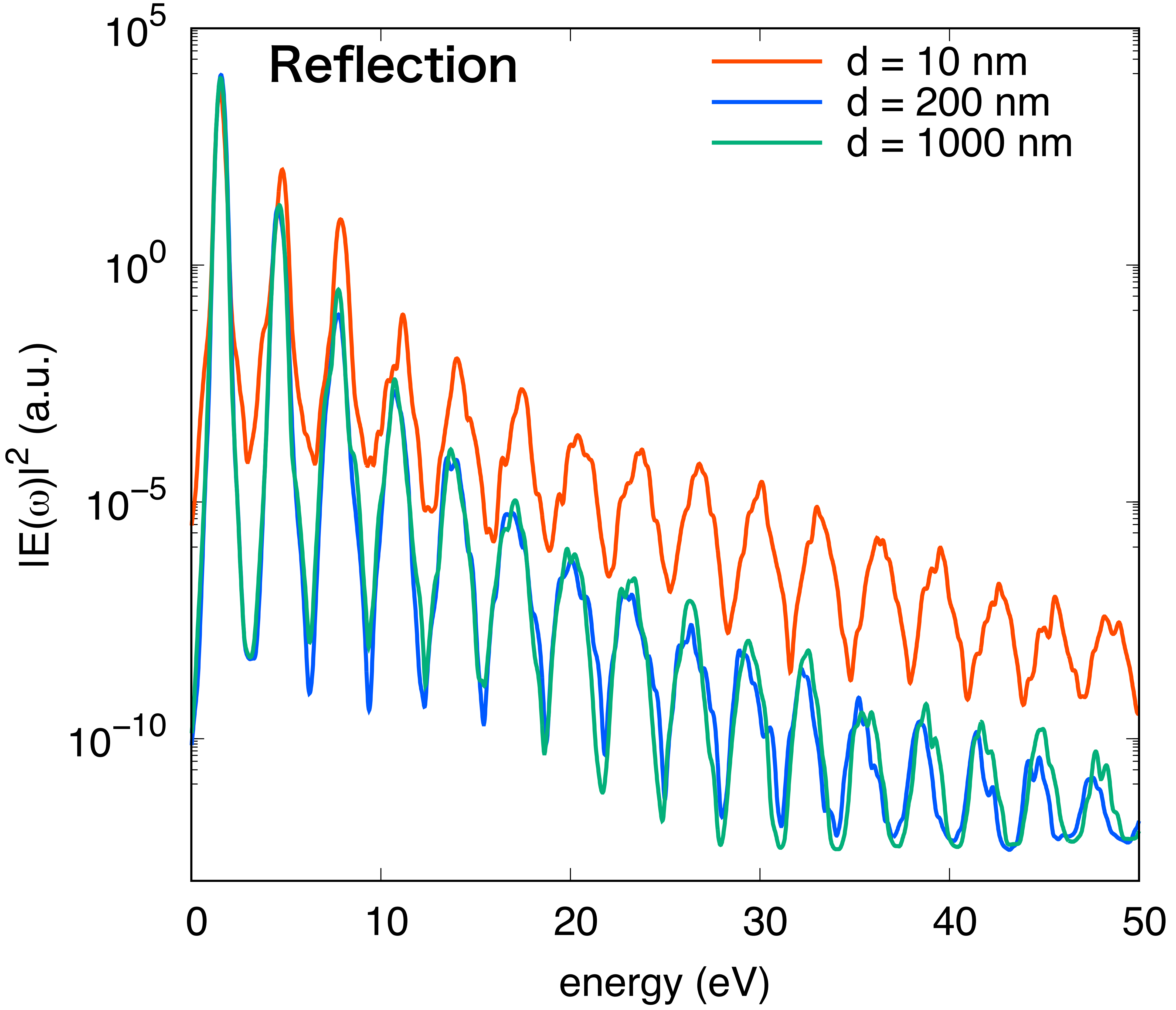
- After real-time calculation, HHG spectra for refection & transmission can be obtained by Fourier transforming the scattered waves.

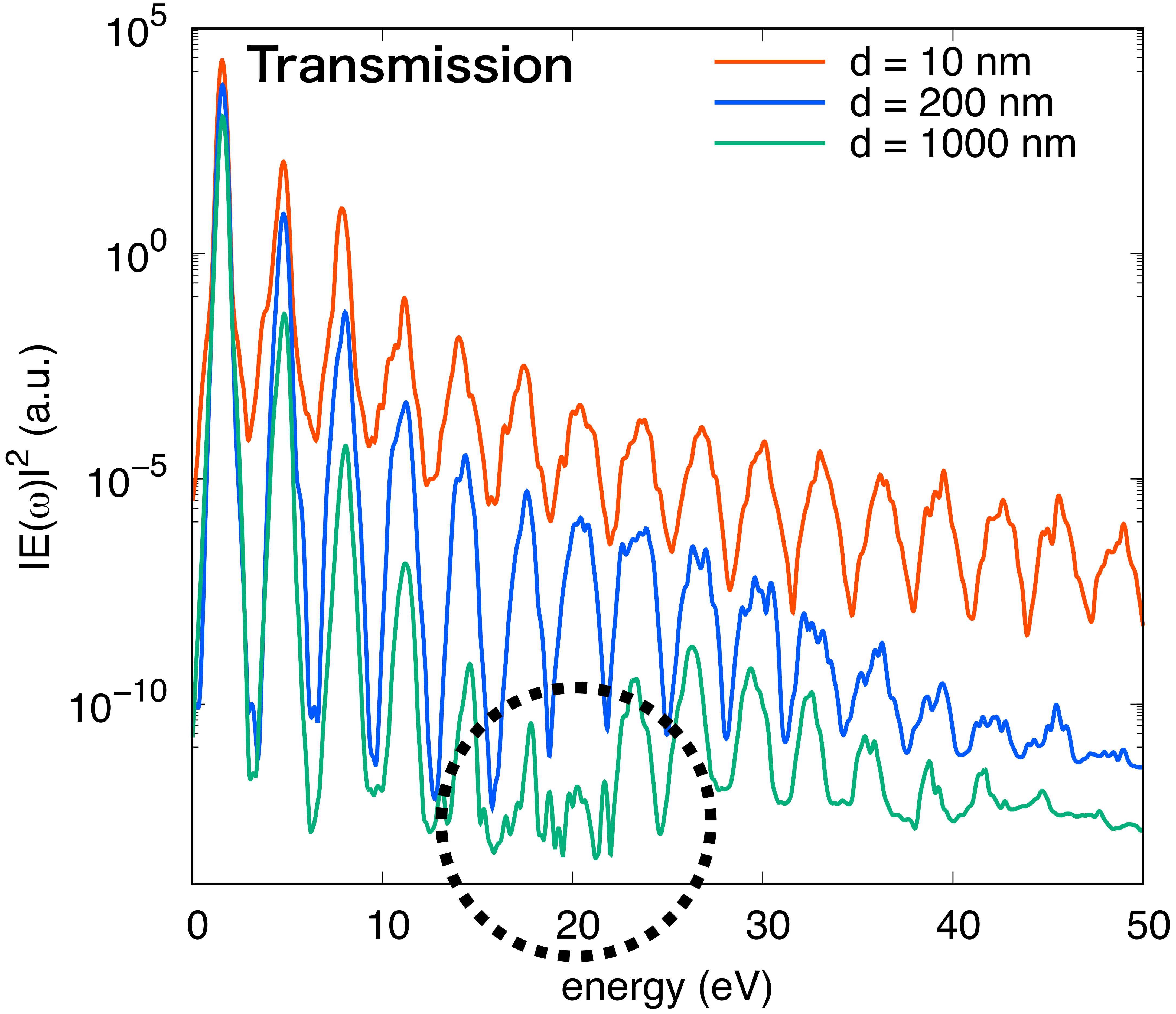






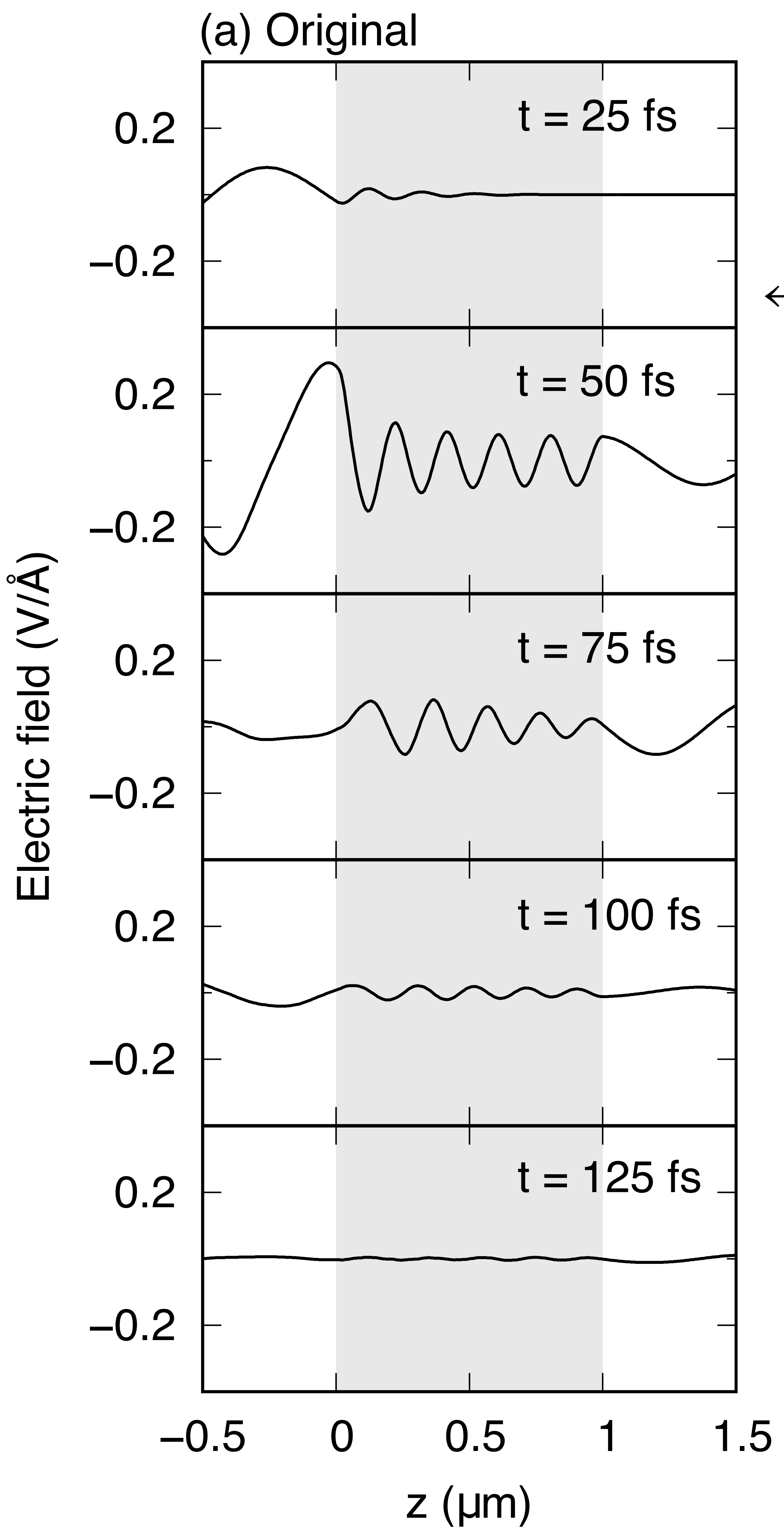
Reflection & Transmission HHG spectra





- Comparison of R & T HHG spectra with the thickness of 10, 200, & 1000 nm
- The d= 10 nm HHG is the strongest. Reflection HHG is almost saturated @ d= 200 nm.
- As the thickness increases, transmission HHG gradually decreases and a dip at 20 eV appears.

Propagation of high harmonics: Fourier analysis of EM field (d=1000 nm)



To understand the dip structure, we investigate the pulse propagation behavior separating frequency regions by Fourier analysis of the electric field

← Snapshots of the electric field for the d=1000 nm case

$$E(Z,\omega) = \int_0^{T_{\rm tot}} dt\, e^{i\omega t} f(t)\, E(Z,t)$$

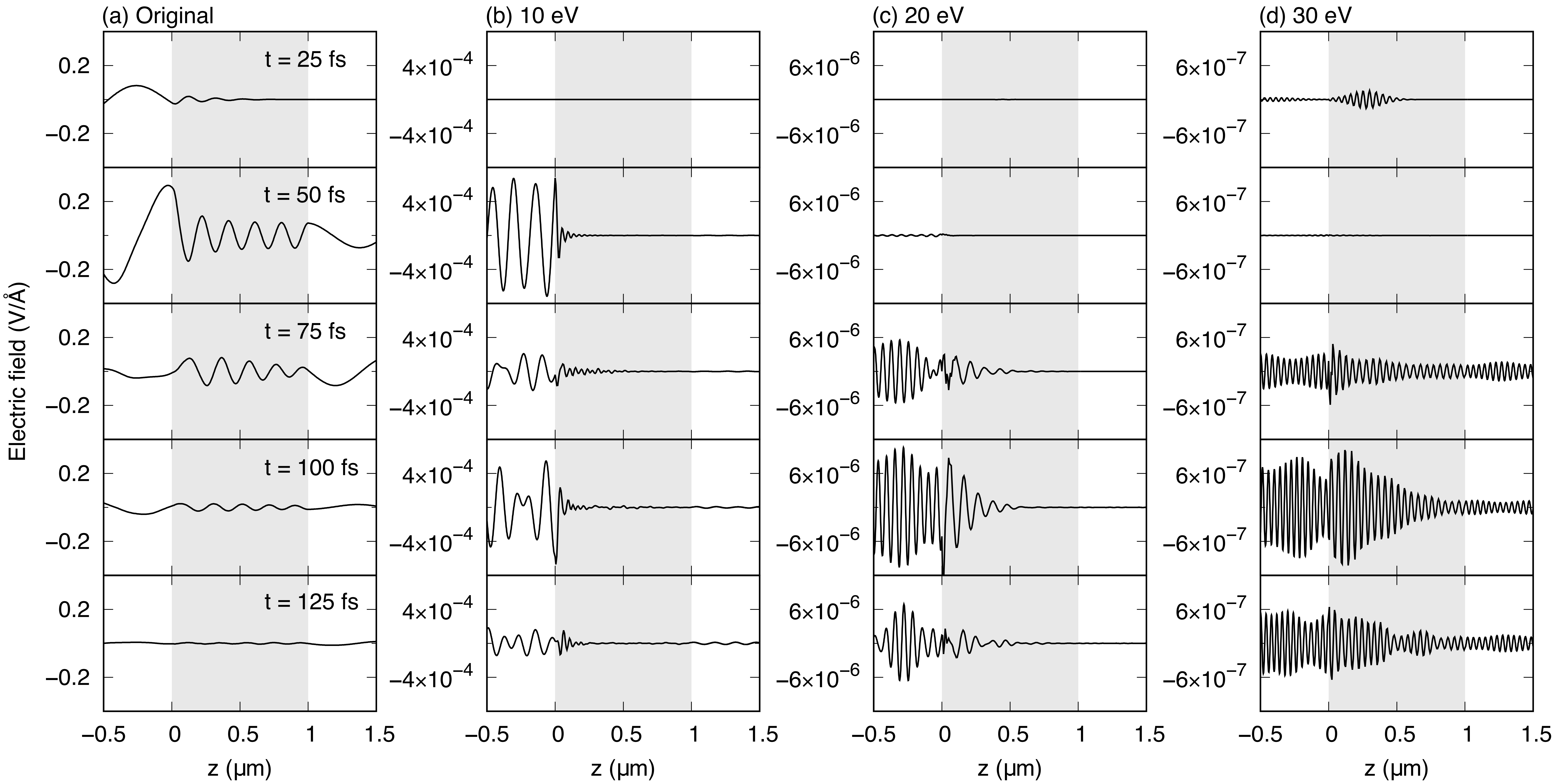
Time Fourier transform at each grid point

$$E_{\rm filtered}(Z,t) = \int_0^{\omega_{\rm max}} \frac{d\omega}{\pi} {\rm Re}\left[e^{-i\omega t} w(\omega-\omega')\, E(Z,\omega)\right]$$

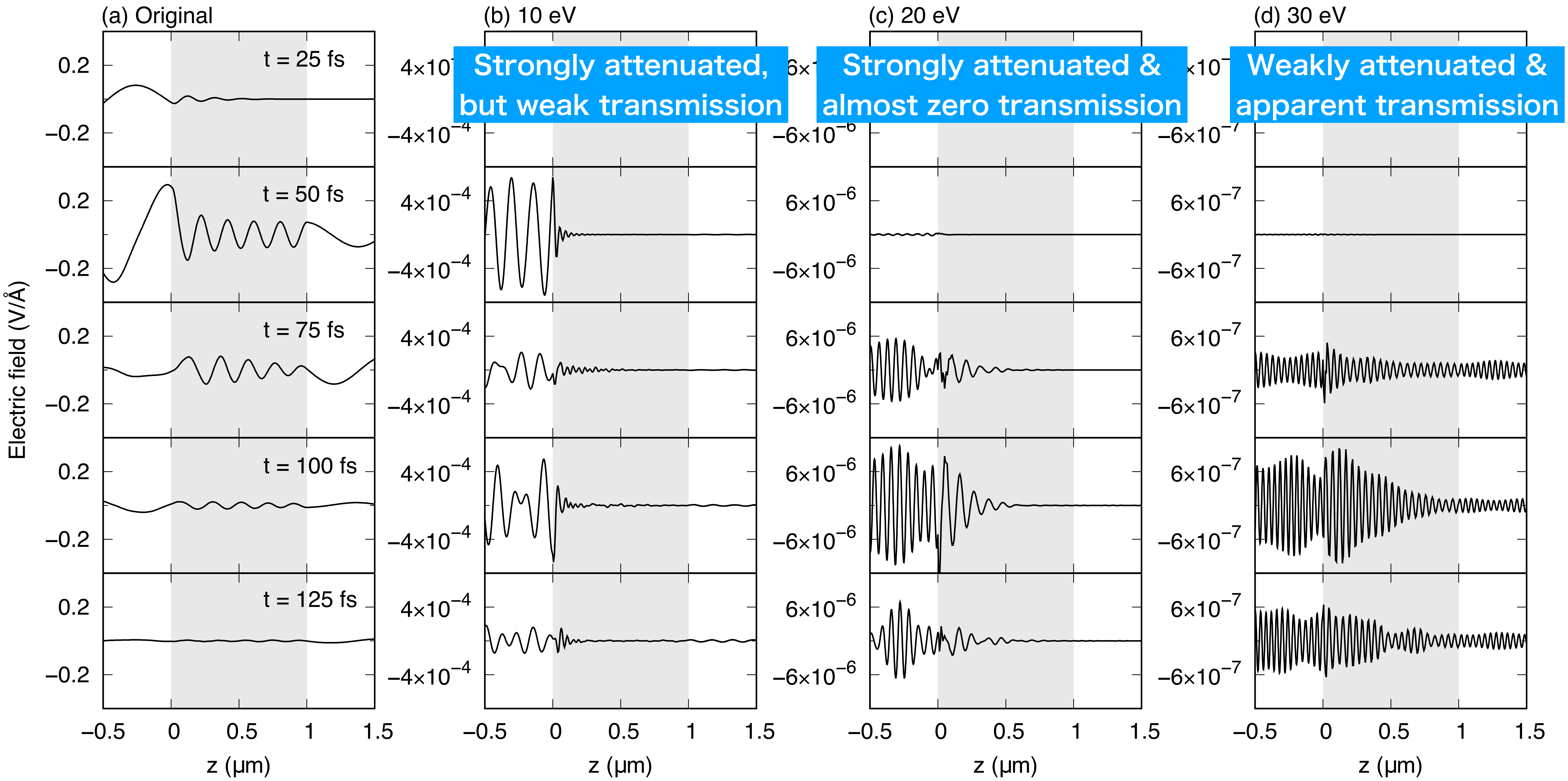
Inverse Fourier transform with a window function to extract oscillation in a certain frequency region

$w(\omega-\omega')$ is Blackman window function (width = 10 eV)

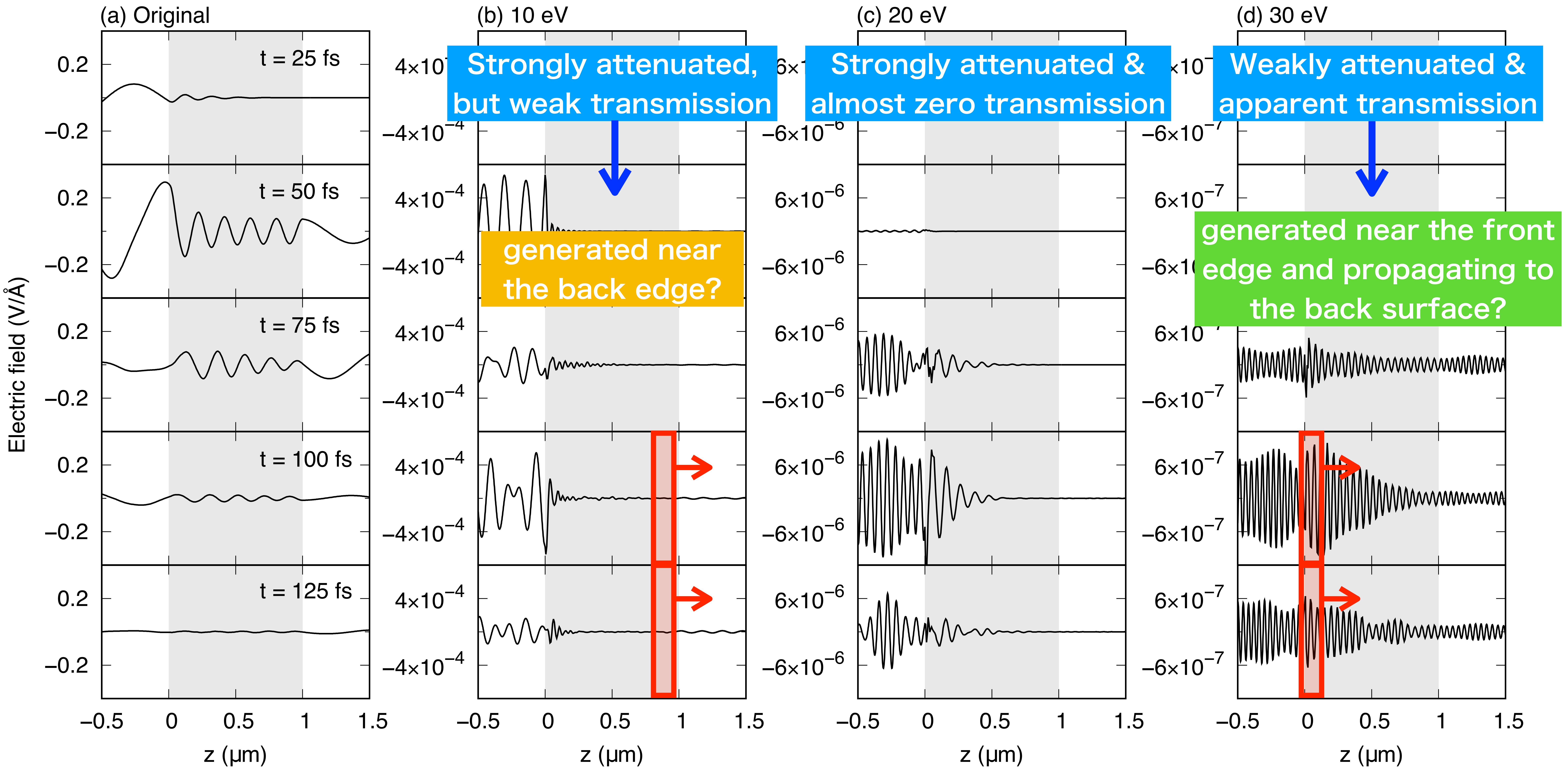
Propagation of high harmonics: Fourier analysis of EM field (d=1000 nm)



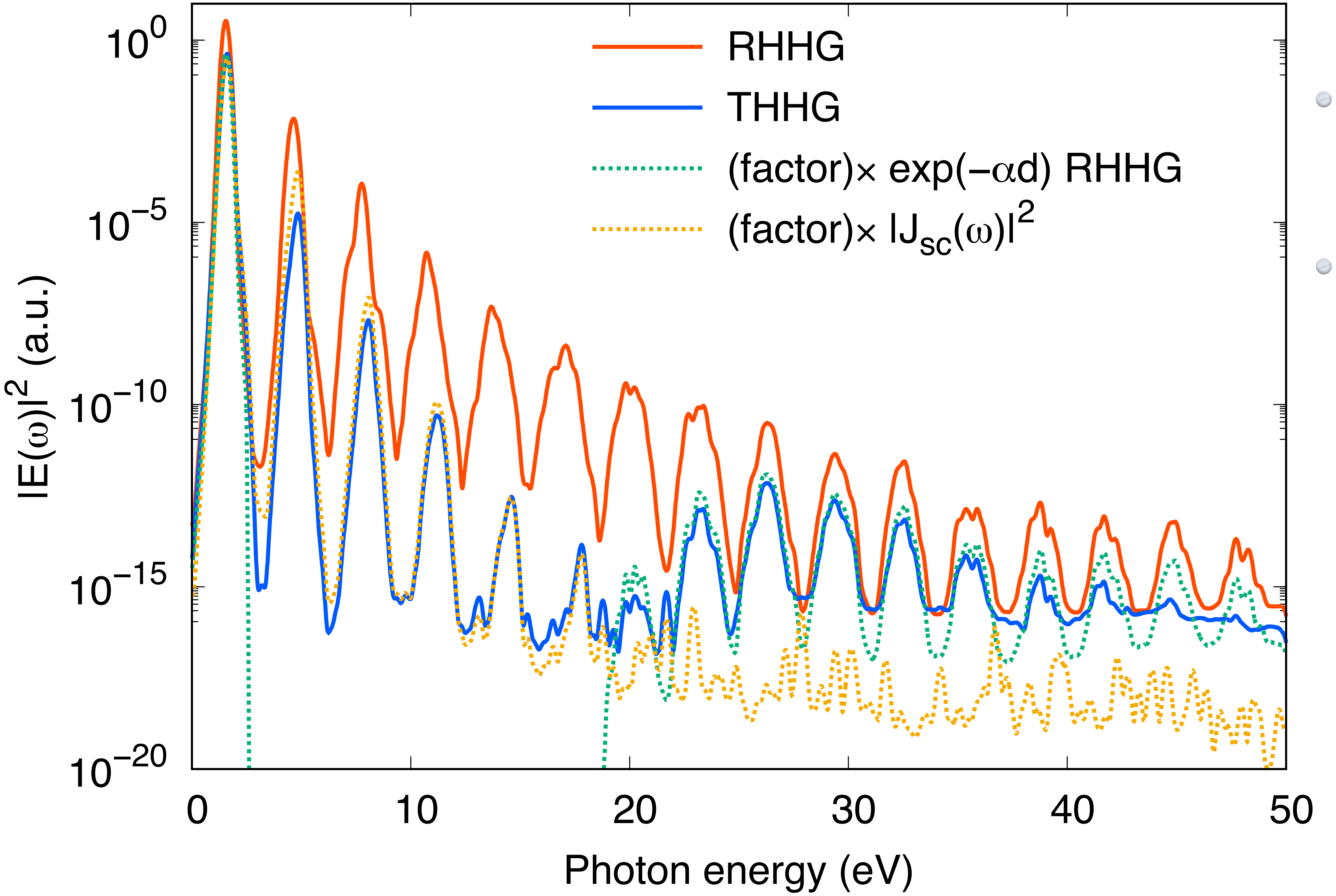
Propagation of high harmonics: Fourier analysis of EM field (d=1000 nm)



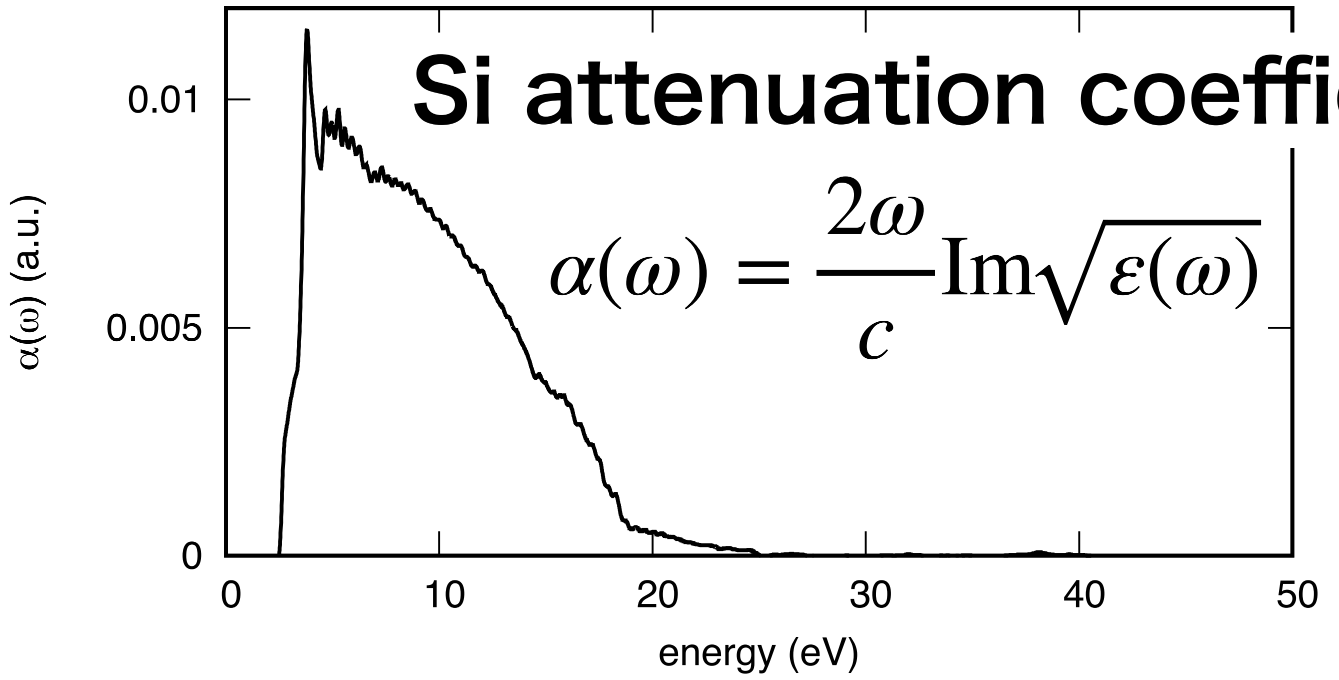
Propagation of high harmonics: Fourier analysis of EM field (d=1000 nm)



Reflection & Transmission HHG spectra: the d=1000 nm case

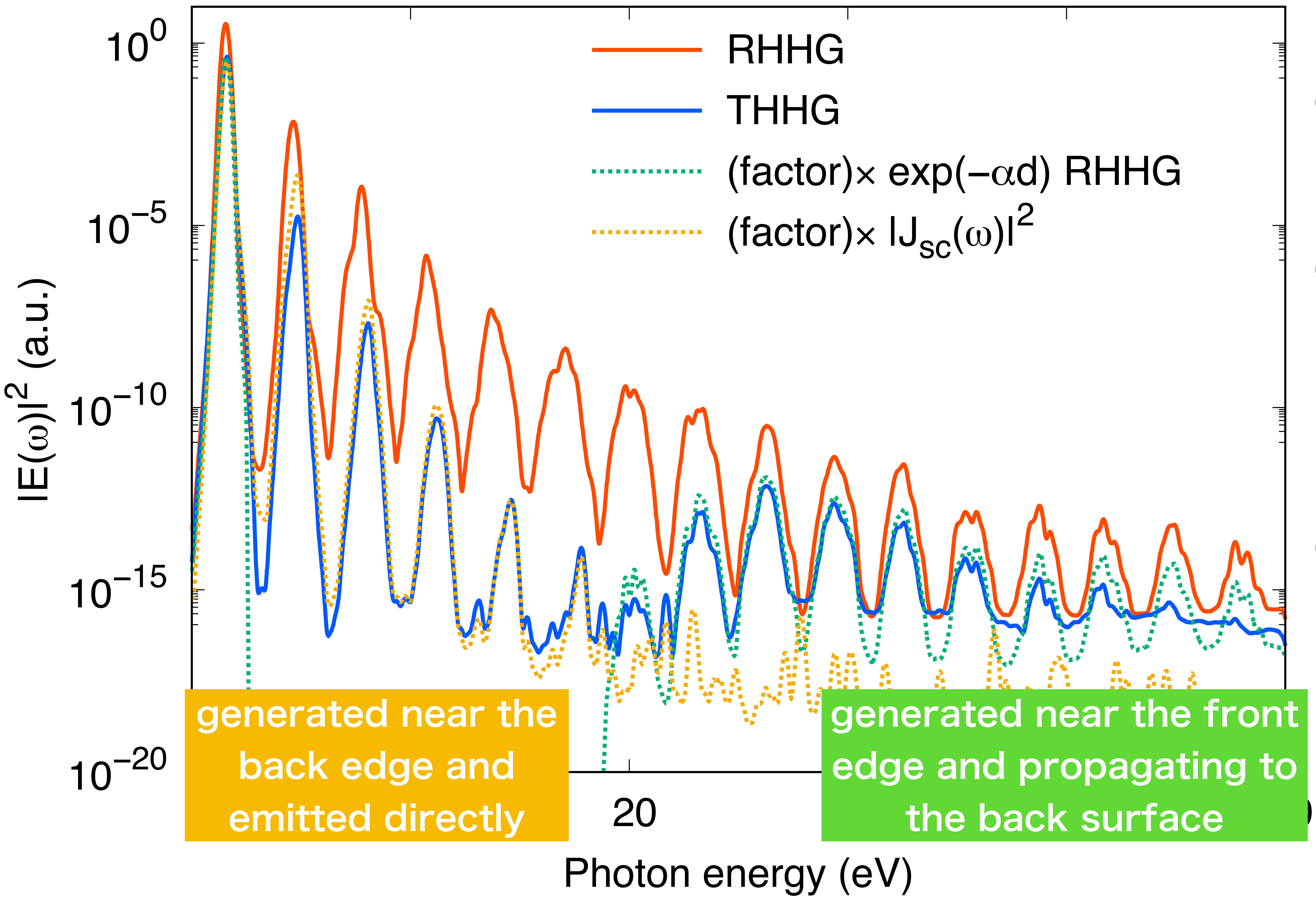


- We revisit HHG spectra for reflection (RHHG) & transmission (THHG)
- RHHG spectrum multiplied by exp(-αd) agrees with THHG above 20 eV
- It may be generated near the front edge and propagating to the back surface

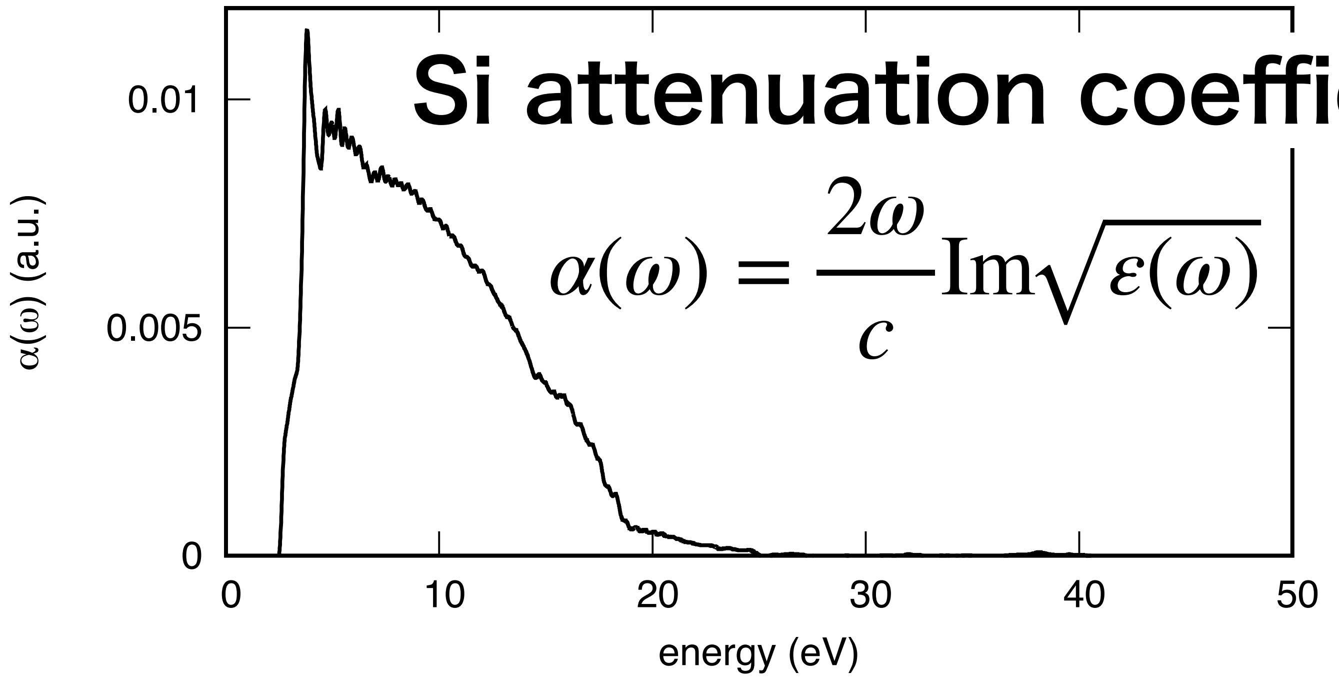


$$\alpha(\omega) = \frac{2\omega}{c}\mathrm{Im}\sqrt{\varepsilon(\omega)}$$

Reflection & Transmission HHG spectra: the d=1000 nm case

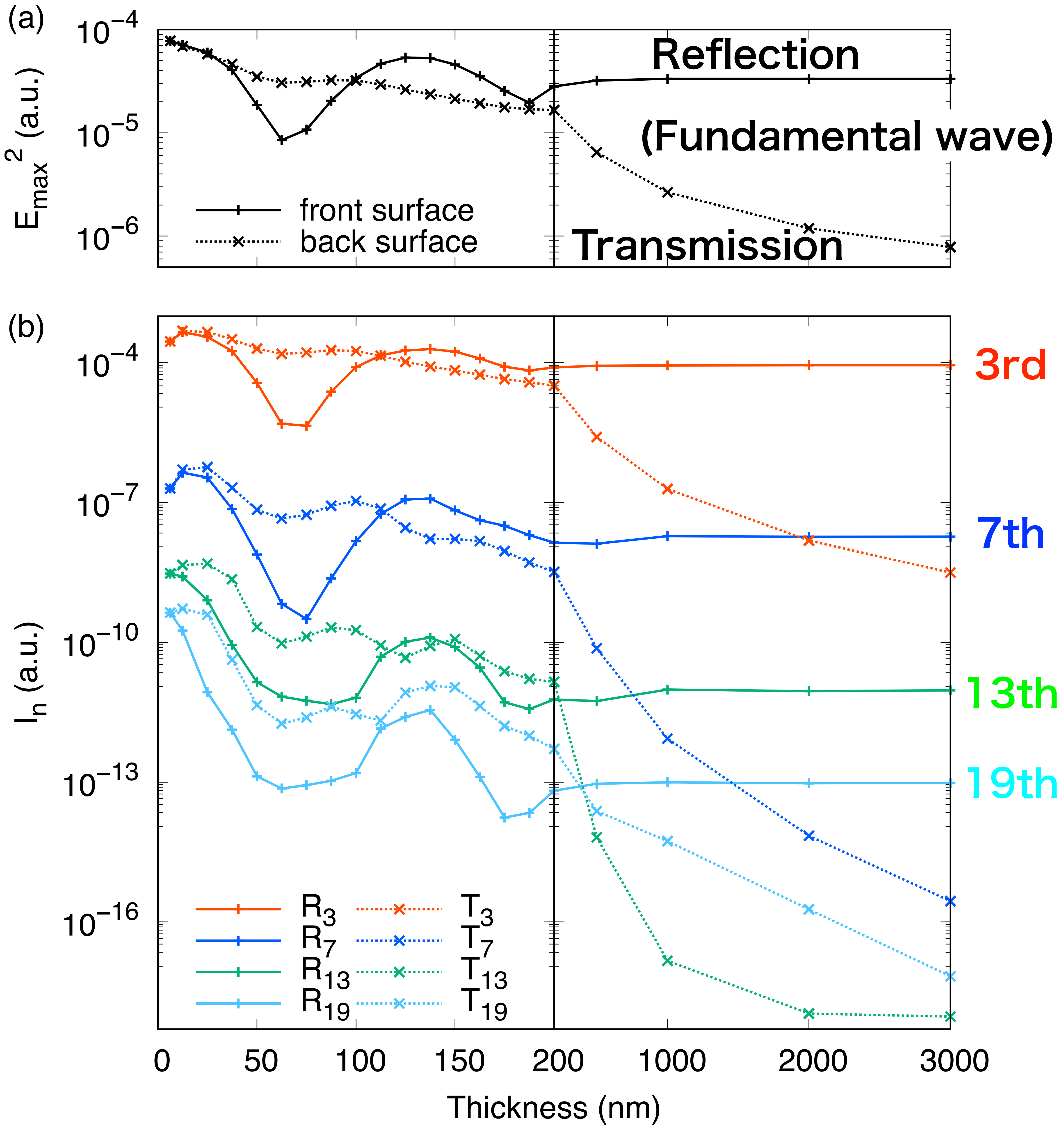


- We revisit HHG spectra for reflection (RHHG) & transmission (THHG)
- RHHG spectrum multiplied by exp(-αd) agrees with THHG above 20 eV
- Spectra of the current (J_{sc}) calculated by ordinary TDDFT using the transmitted fundamental wave (Fourier filtered) agrees with THHG below 20 eV
- <u>THHG originates from two different mechanisms below & above 20 eV</u>



$$\alpha(\omega) = \frac{2\omega}{c}\mathrm{Im}\sqrt{\varepsilon(\omega)}$$

Thickness dependence of reflection & transmission HHG

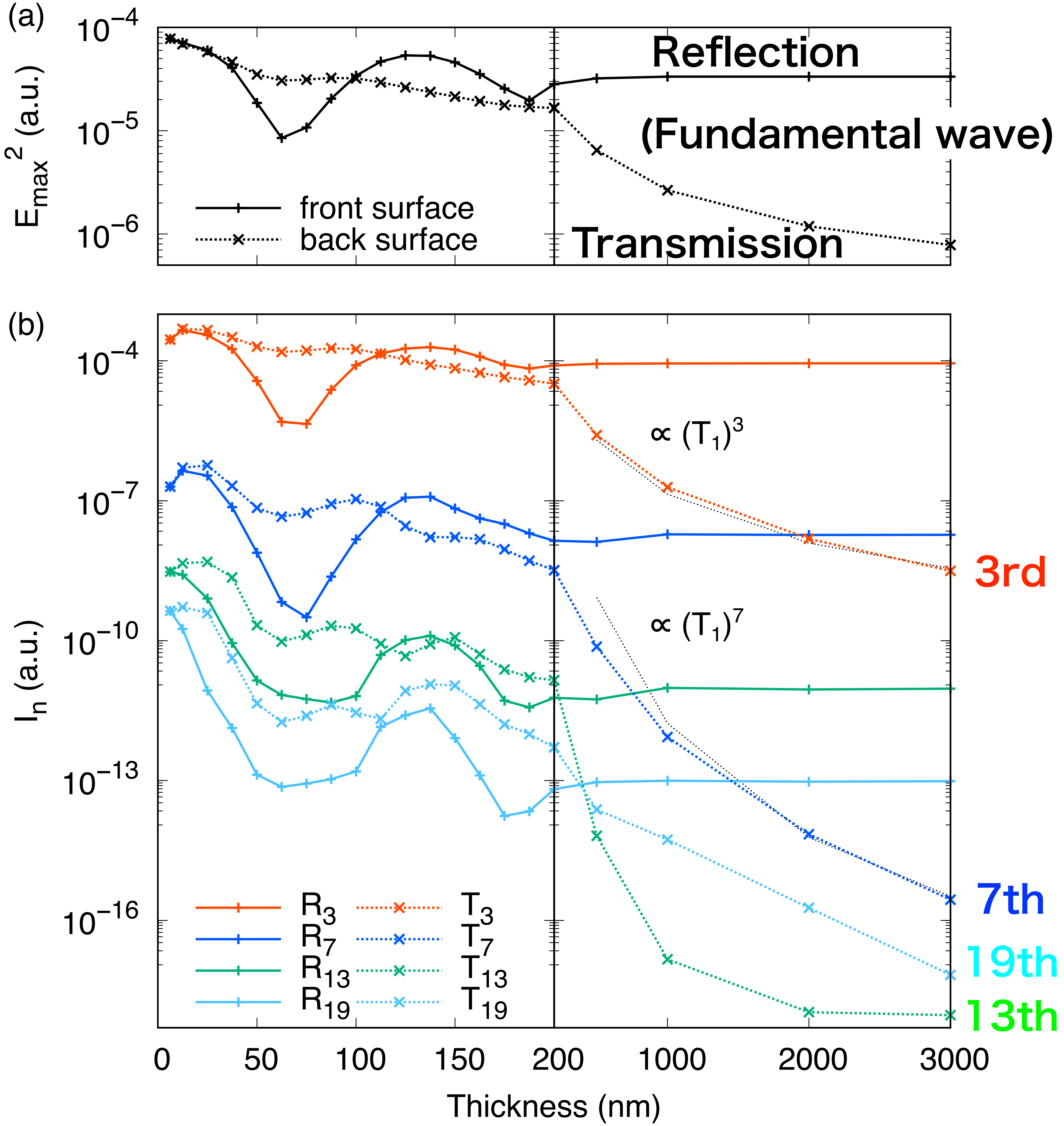


- Thickness dependence of the intensities of the fundamental wave, 3rd, 7th (~ 10eV), 13th (~ 20eV), & 19th (~ 30eV) high harmonics.
- Below 200 nm: oscillatory behavior as a function of thickness originates from the interference effect.*
- Very thin case: RHHG = THHG, and HHG has the maximum @ few tens of nm.*

* cf. S. Yamada and K. Yabana, PRB **103**, 155426 (2021).

- RHHG (solid lines) becomes constant from d= 200 nm.
- THHG (dotted lines) gradually decreases.

Thickness dependence of reflection & transmission HHG



- 3rd & 7th (~ 10eV) THHGs follow 3rd & 7th powers of the transmitted fundamental wave
  → generated near the back edge
- 19th (~ 30eV) THHG linearly decreases as exp(-αd)
  → generated near the front edge
- 13th (~ 20eV) THHG: “valley” of the two mechanisms

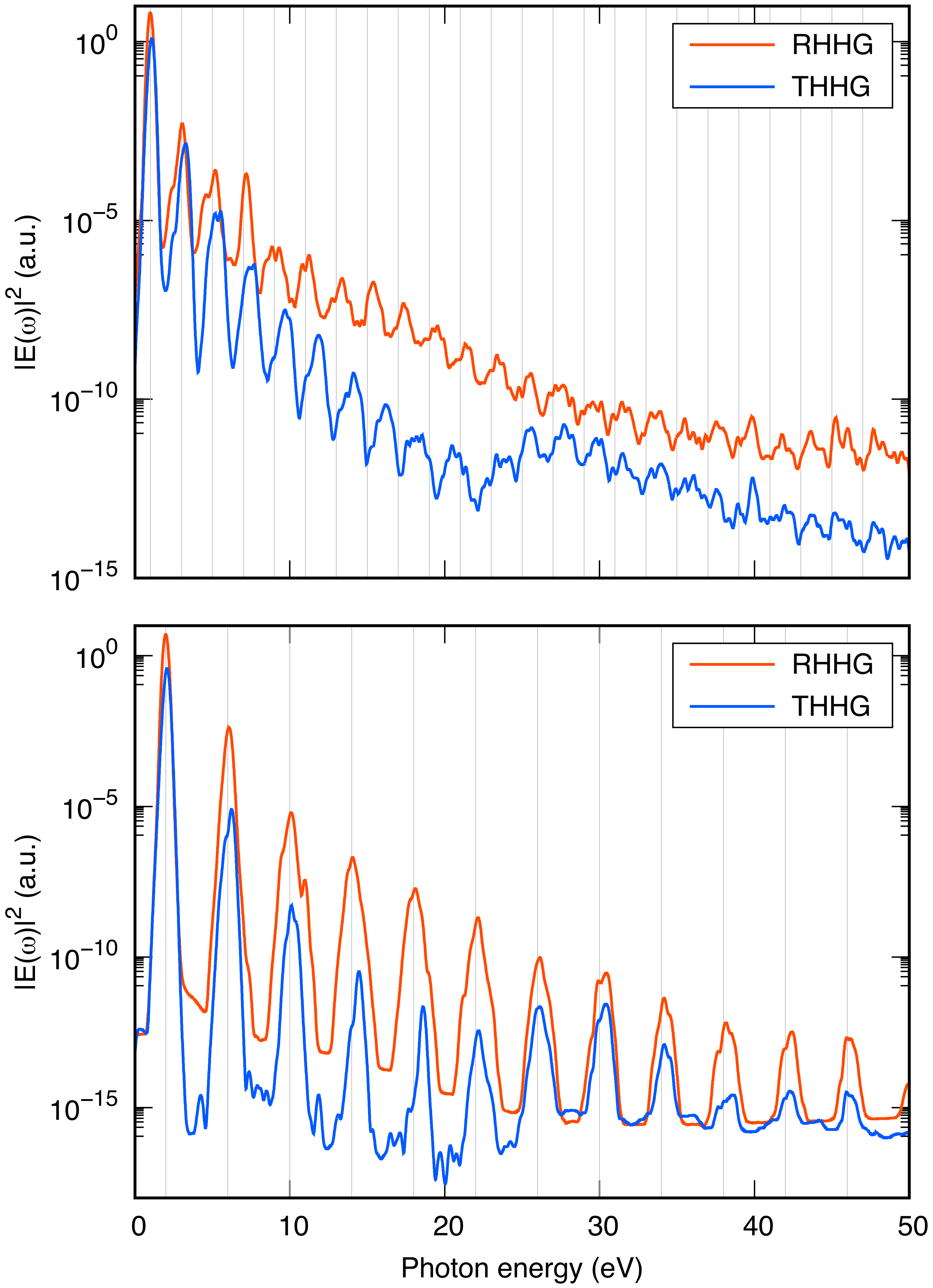
Other fundamental frequencies: the d=1000 nm case

- $\hbar\omega$ = 1 eV

(I= 6x10[12] W/cm[2])

- $\hbar\omega$ = 2 eV

(I= 5x10[12] W/cm[2])



- RHHG and THHG for the fundamental frequencies of 1 eV and 2 eV.
- THHGs have similar dips @ 20 eV as expected.
- These results show that the position of the dip at 20 eV in THHG is independent of the fundamental frequency.
- This is natural because the dip originates from the Si attenuation coefficient.

Summary

“Propagation effects in high-harmonic generation from dielectric thin films”, accepted, PRB, arXiv:2210.14430

- The first-principles simulations are performed of the process in which an intense pulsed light irradiates Si thin films up to 3 μm thickness.
- The intensity of transmission HHG (THHG) gradually decreases with the thickness, while the reflection HHG (RHHG) becomes constant from a certain thickness.
- THHG has two different origins for frequency below & above 20 eV.
- THHG below 20 eV is generated near the back edge from the transmitted fundamental wave.
- THHG above 20 eV is generated near the front edge and propagates from there to the back surface.