Frontiers in Intense Laser-Matter Interaction Theory (FILMITh)
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Ab-initio description for the interaction of intense laser pulses with solids

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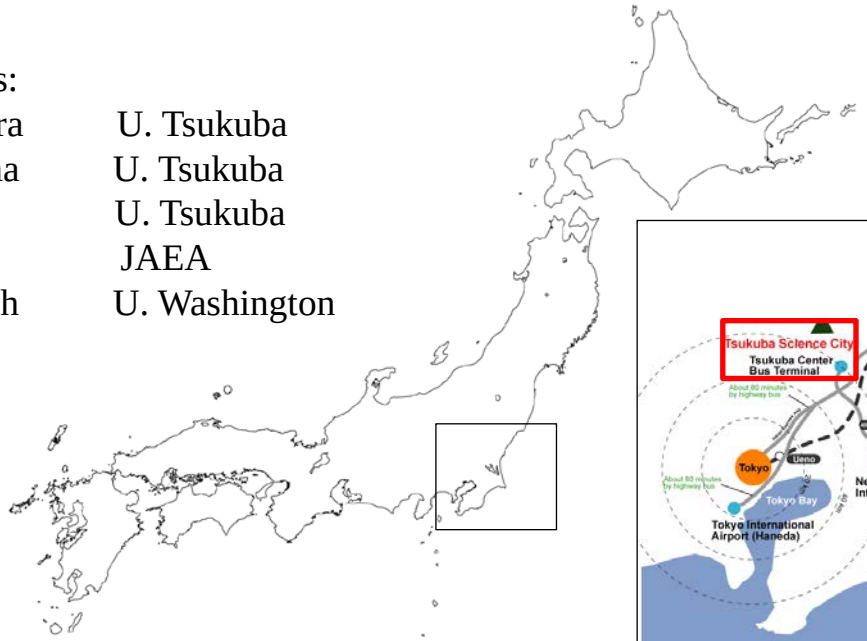


Theoretical description for intense laser pulse on solid from first-principles density functional calculation

--- From Quantum Mechanics to Macroscopic Electromagnetism ---

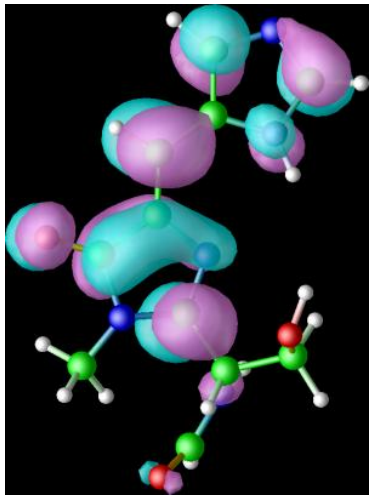
$10^{12} - 10^{15}\,\mathrm{W/cm^2}$

Laser intensity

Nonlinear optics | Coherent phonon | HHG | Electron-hole plasma, Optical breakdown | Laser acceleration | Vacuum breakdown

Nonrelativistic Quamtum mechanics

Relativistic Classical mechanics



Density functional theory

Successful theory to describe matters (atoms, molecules, solids, …)

Large scale computation

Massively parallel supercomputer (K-computer, Japan)

Theoretical approach for ordinary (weak) optical response

Electromagnetism:
Maxwell equation for macroscopic fields, *E*, *D*, *B*, *H*

Constitution relation

$$D = D[E] = \varepsilon(\omega)E$$

Quantum Mechanics:
Perturbation theory to calculate linear susceptibilities, $\varepsilon(\omega)$

As the light intensity increases, nonlinear effects appear

Quantum mechanics is still useful to calculate nonlinear susceptibilities.

$$D = D[E] = \varepsilon(\omega)E + 4\pi\chi^{(2)}E^2 + 4\pi\chi^{(3)}E^3 + \cdots$$

Extreme intensity of laser pulse

⇩

We need to solve couple Maxwell + Schroedinger dynamics.

Contents

1. Formalism I:
   Electron dynamics in a unit cell for a given electric field *E(t)*.

2. Formalism II:
   Coupled dynamics of laser pulse (macroscopic electric field) and electron dynamics in solid.

3. Example of applications:
   Dielectric properties of solid surface excited by intense laser pulse

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How to describe many-electron dynamics?

Density Functional Theory: First-principles calculation for electronic ground state

Widely used in chemistry and physics to describe atoms, molecules, and solids without any empirical parameters.

W. Kohn

1998, Nobel prize in chemistry

Time-Dependent Density-Functional Theory (TDDFT): An extension to describe electron dynamics

Time-dependent Kohn-Sham equation for electron orbitals

$$i\hbar\frac{\partial}{\partial t}\psi_i(\vec{r},t) =$$

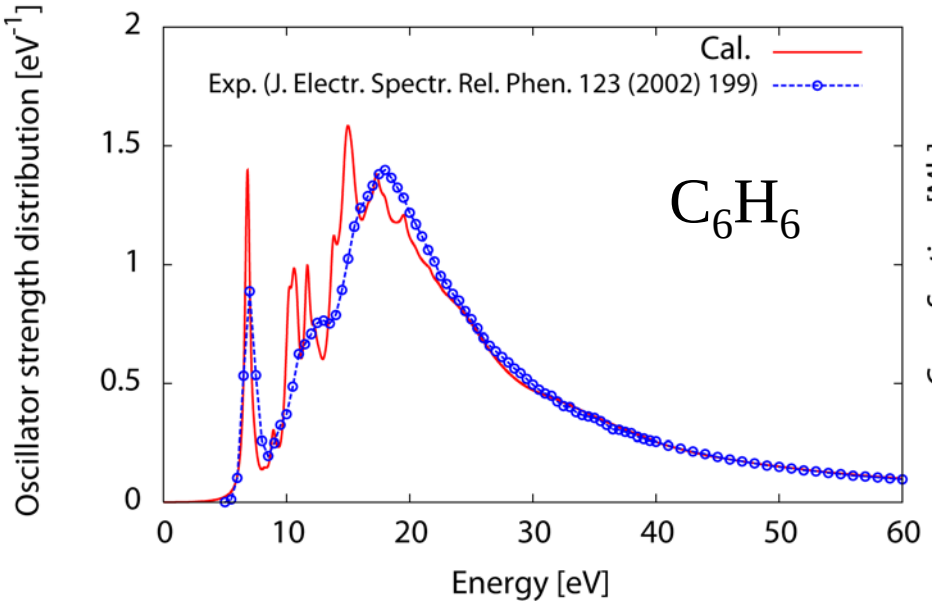
$$\left\{-\frac{\hbar^2}{2m}\vec{\nabla}^2 + \sum_a V_{ion}(\vec{r}-\vec{R}_a) + e^2\int d\vec{r}'\frac{n(\vec{r}',t)}{|\vec{r}-\vec{r}'|} + \mu_{xc}(n(\vec{r},t)) + V_{ext}(\vec{r},t)\right\}\psi_i(\vec{r},t)$$

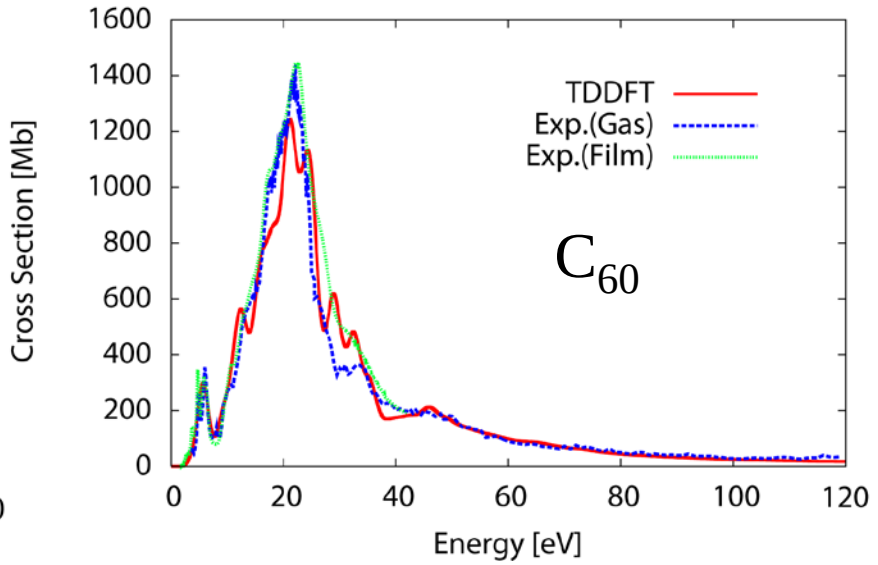
$$n(\vec{r},t) = \sum_i |\psi_i(\vec{r},t)|^2$$

Standard recipes: density functional, pseudopotential, real-space grid representation, …

Linear Response TDDFT is widely used for electronic excitations and optical responses of molecules

Oscillator strength (photoionization cross section) of molecules





Y. Kawashita, K. Yabana, M. Noda, K. Nobusada, T. Nakatsukasa, J. Mol. Struct. THEOCHEM 914 (2009) 130.

1. Formalism I:
Electron dynamics in a unit cell for a given electric field *E(t)*.

We assume a long wavelength limit

↓

Electron dynamics in a unit cell of crystalline solid caused by time-dependent, spatially uniform electric field

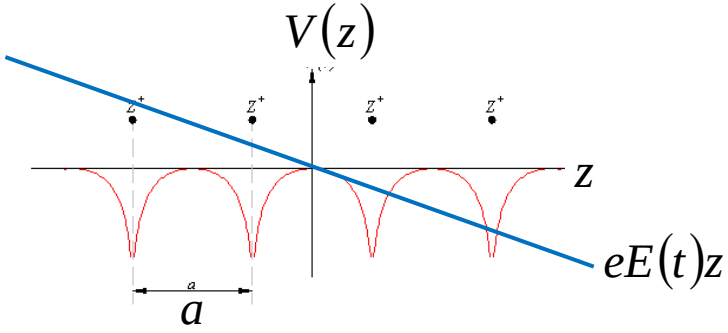
In practical calculations, following two points need to be considered.

Gauge choice

Treatment of polarization

Gauge choice

Length gauge is not convenient in crystalline solid



Scalar potential $eE(t)z$ spoils spatial periodicity of the Hamiltonian

$$h(\vec{r}+\vec{a}) \neq h(\vec{r})$$

Velocity gauge: expressing uniform electric field by vector potential, one may recover the periodicity of the Hamiltonian

$$i\hbar\frac{\partial}{\partial t}\psi(t)=\left[\frac{1}{2m}\left(\vec{p}-\frac{e}{c}\vec{A}(t)\right)^2+V(\vec{r},t)\right]\psi(t) \qquad \vec{A}(t)=c\int^{t} dt' E(t')\hat{z}$$

$$h(\vec{r}+\vec{a},t)=h(\vec{r},t) \qquad \psi_{nk}(\vec{r}+\vec{a},t)=e^{i\vec{k}\vec{a}}\psi_{nk}(\vec{r},t)$$

At each time, we may define Bloch wave function.

➡ Time-dependent extension of first-principles band calculation.

Treatment of Polarization

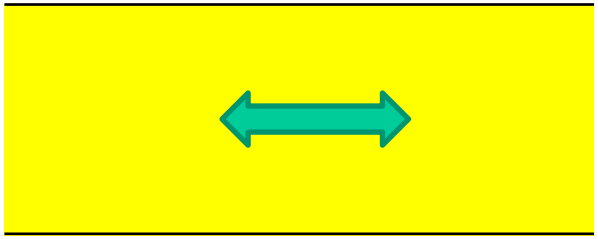
Electric field inside solid depends on macroscopic shape of solid

$$i\hbar\frac{\partial}{\partial t}\psi_{nk}(t)=\left[\frac{1}{2m}\left(\vec{p}-\frac{e}{c}\vec{A}(t)\right)^2+V(\vec{r},t)\right]\psi_{nk}(t)$$

$$A(t)=A_{ext}(t)+cA_{polarization}(t)$$

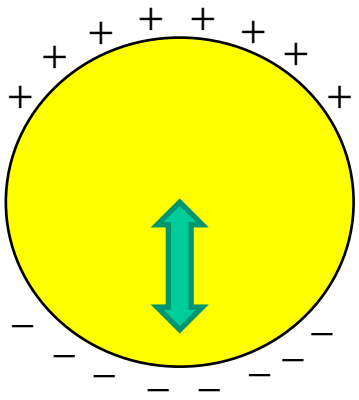
$$\frac{d^2\vec{A}_{polarization}(t)}{dt^2}=\frac{4\pi}{\Omega}\int_{\Omega(cell)}d\vec{r}\ \vec{j}\ (\vec{r},t)$$

Thin film: Transverse



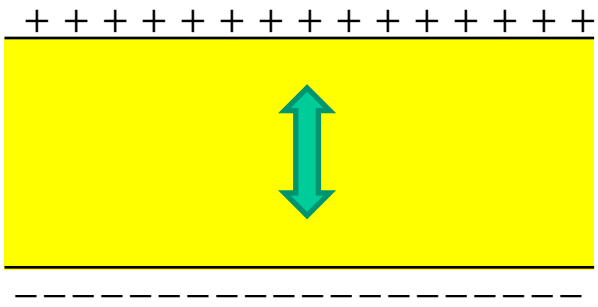
$$A(t)=A_{ext}(t)$$

Sphere



$$A(t)=A_{ext}(t)+\frac{1}{3}A_{polarization}(t)$$

Thin film: Longitudinal

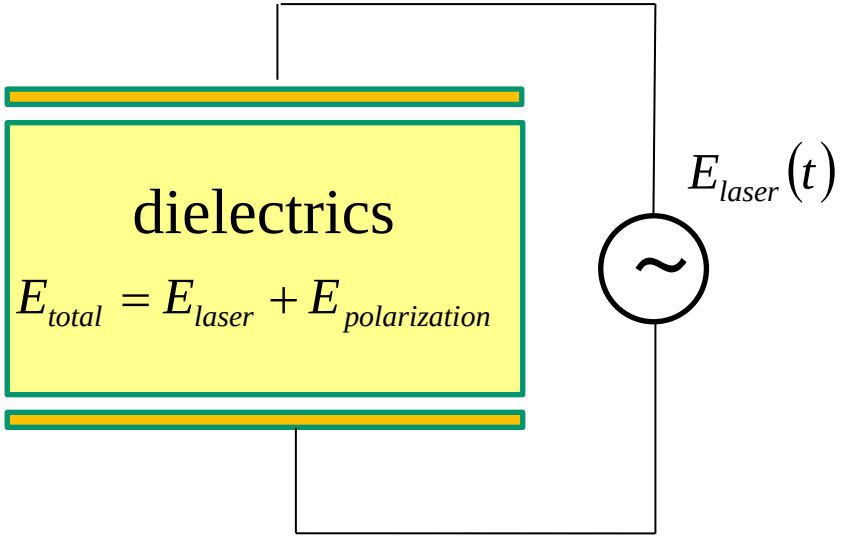


$$A(t)=A_{ext}(t)+A_{polarization}(t)$$

Electron dynamics in longitudinal geometry

Bertsch, Iwata, Rubio, Yabana, Phys. Rev. B62(2000)7998.

“parallel-plate-capacitor with dielectrics”.



$$i\hbar\frac{\partial}{\partial t}\psi_i = \frac{1}{2m}\left(-i\hbar\vec{\nabla} + \frac{e}{c}\vec{A}\right)^2\psi_i - e\phi\psi_i + \frac{\delta E_{xc}}{\delta n}\psi_i$$

$$n = \sum_i |\psi_i|^2 \qquad \vec{j} = \frac{1}{2m}\sum_i\left(\psi_i^*\left(\vec{p} + \frac{e}{c}\vec{A}\right)\psi_i - c.c.\right)$$

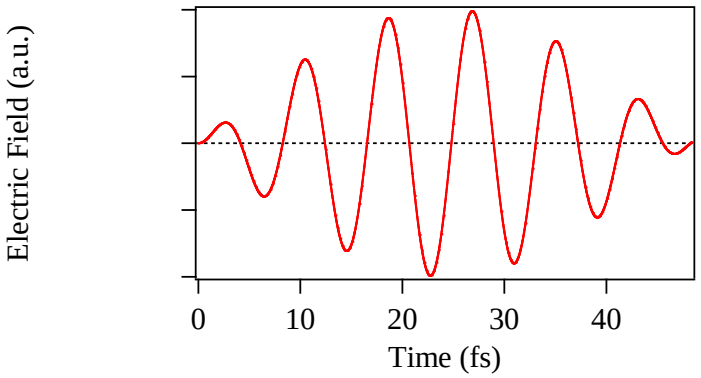
$$A(t) = A_{extr}(t) + A_{polarization}(t)$$

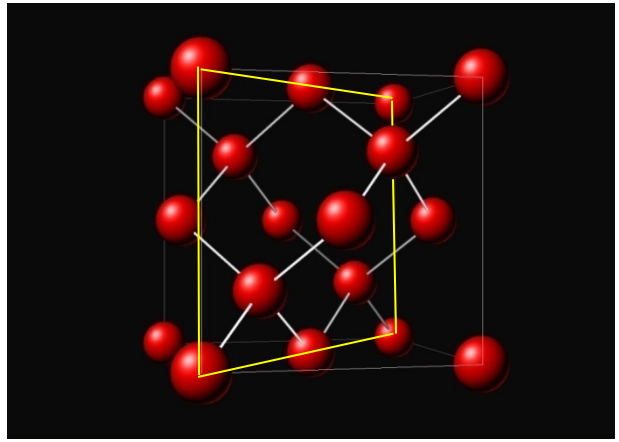
$$\frac{d^2\vec{A}_{polarization}(t)}{dt^2} = \frac{4\pi}{\Omega}\int_{\Omega(cell)} d\vec{r}\ \vec{j}(\vec{r},t)$$

Electron dynamics in bulk silicon under pulse light

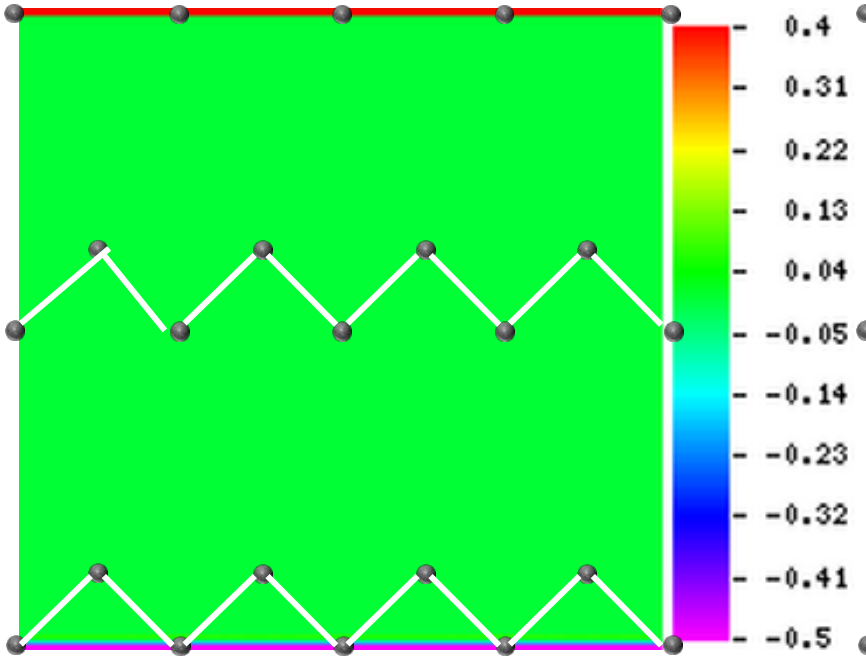
I= 3.5×10^{14} W/cm^2, T=50 fs, $\hbar\omega$=0.5 eV

Laser photon energy is much lower than direct bandgap.

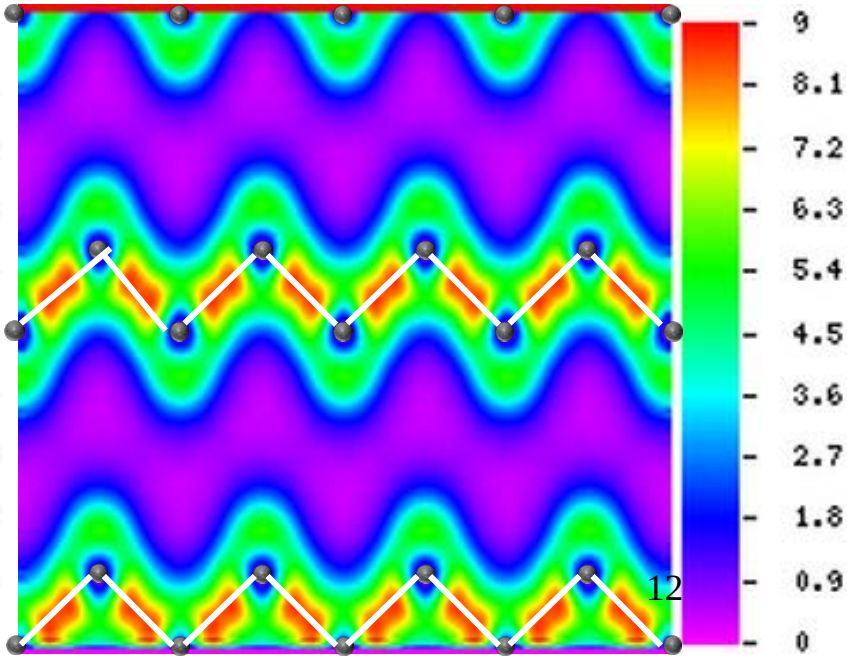




Density change from the ground state (110)



Ground state density (110)



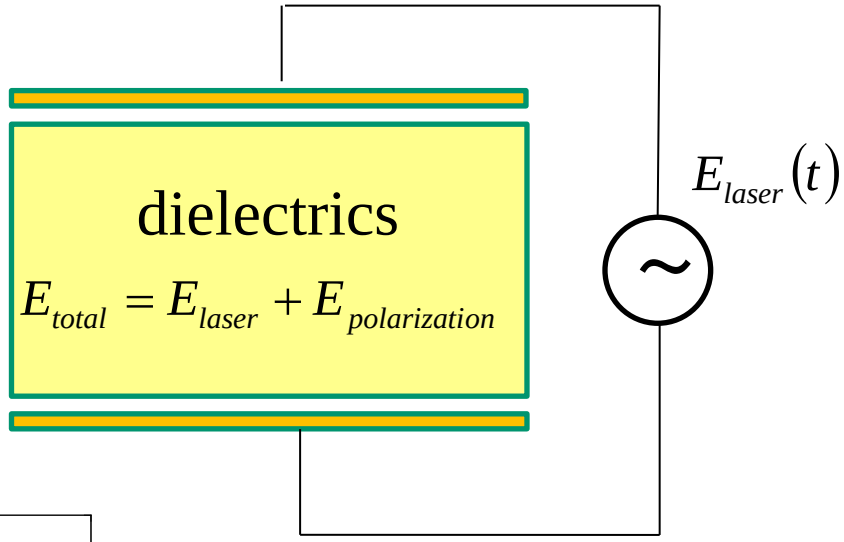
Red +

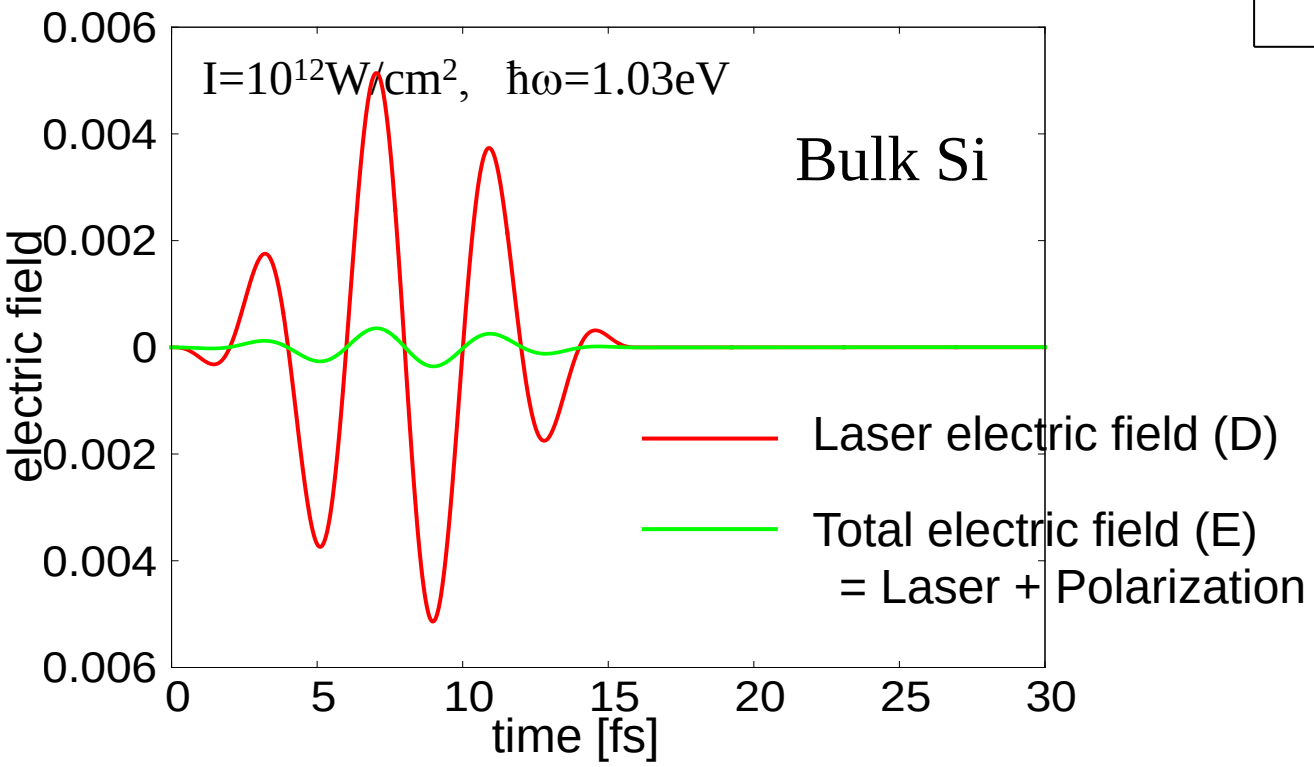
Green 0

Blue -

For weak field, dielectric screening

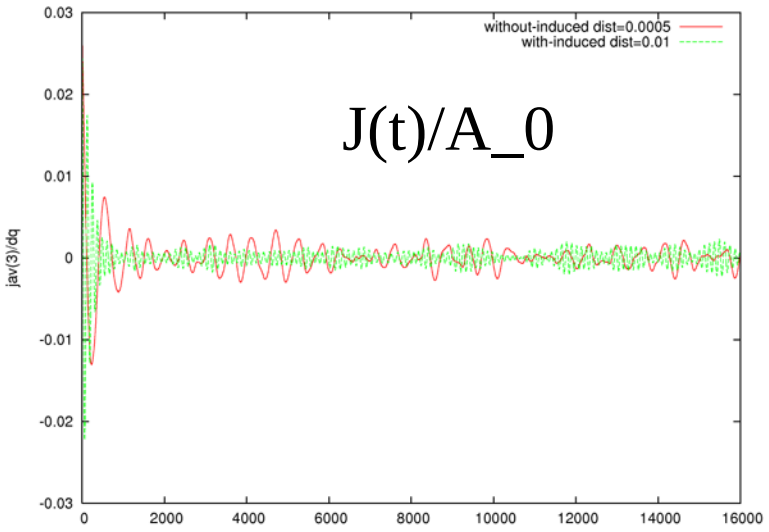
$$E_{total}(t) = \int^{t} dt' \, \varepsilon^{-1}(t-t') E_{ext}(t')$$

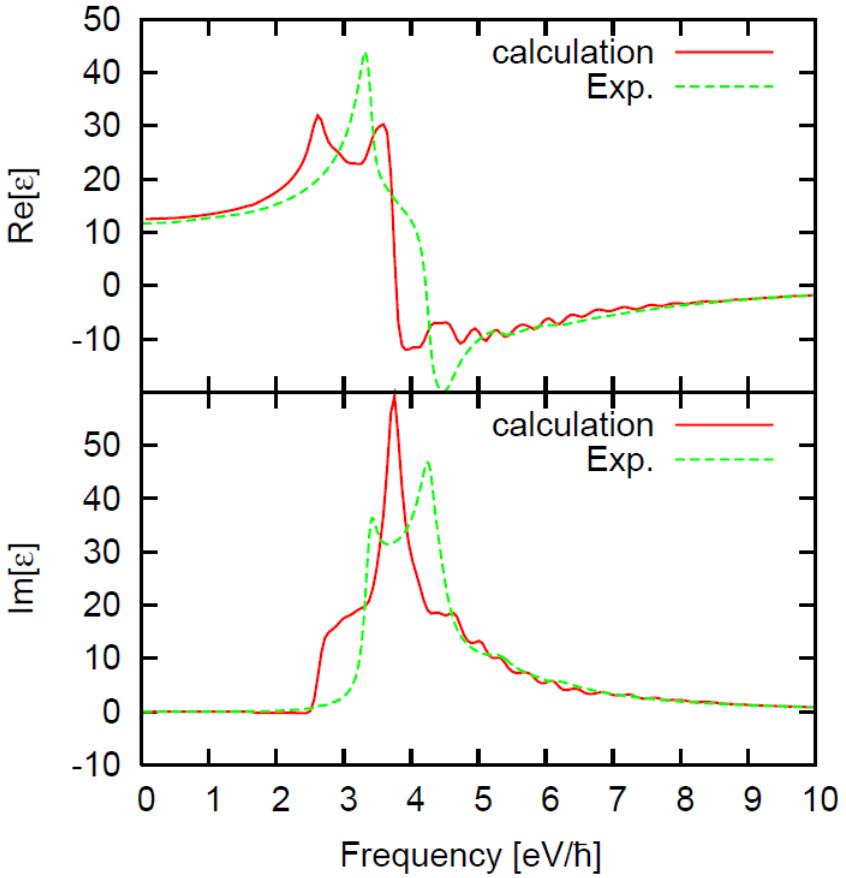




$$D = \varepsilon(\omega) E \qquad \varepsilon \approx 14 \quad (\text{TDDFT})$$

Dielectric function from real-time calculation: bulk Si





We employ simplest functiona, “adiabatic local density approximation”, though it does not give accurate enough result.

GW approx. + Bethe-Salpeter

Most successful but Beyond Kohn-Sham framework

Dots: experiment
Dash-dotted: RPA
Solid: Bethe-Salpeter

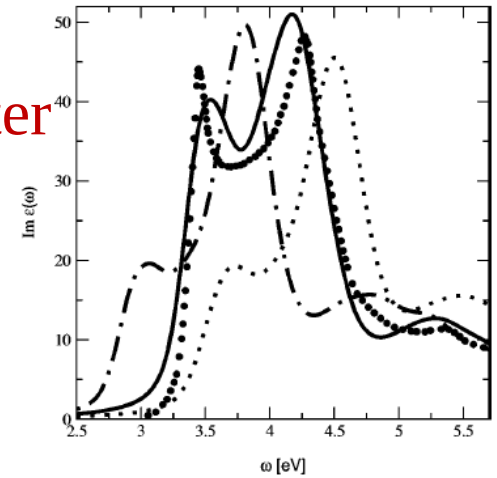
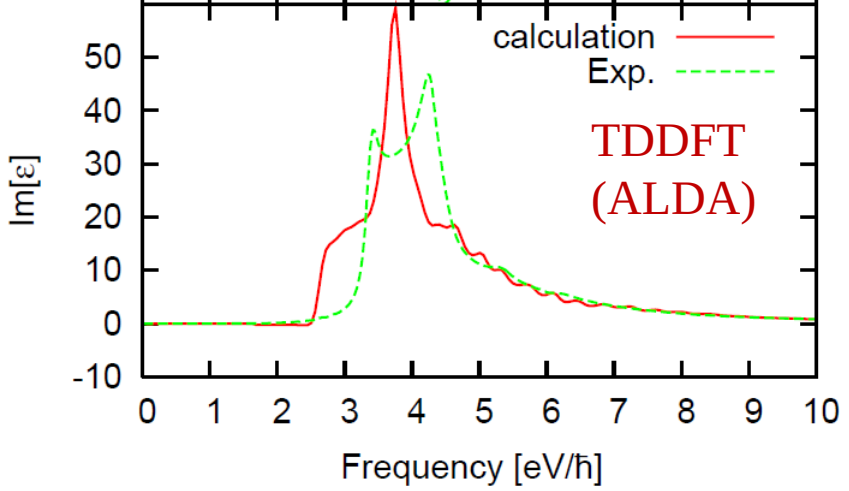
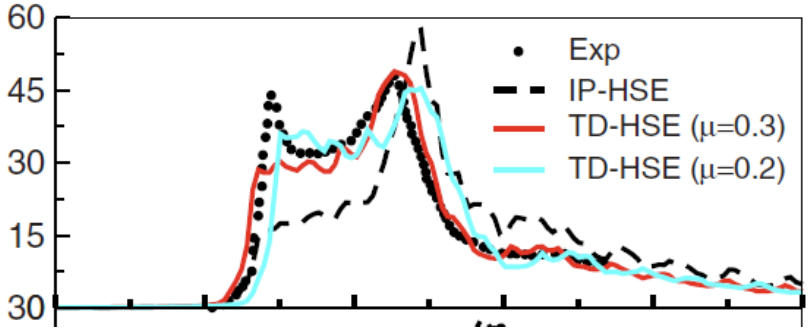


FIG. 5. Silicon absorption spectrum [Im(ε_M)]: ●, experiment (Lautenschlager *et al.*, 1987); dash-dotted curve, RPA, including local field effects; dotted curve, *GW*-RPA; solid curve, Bethe-Salpeter equation.

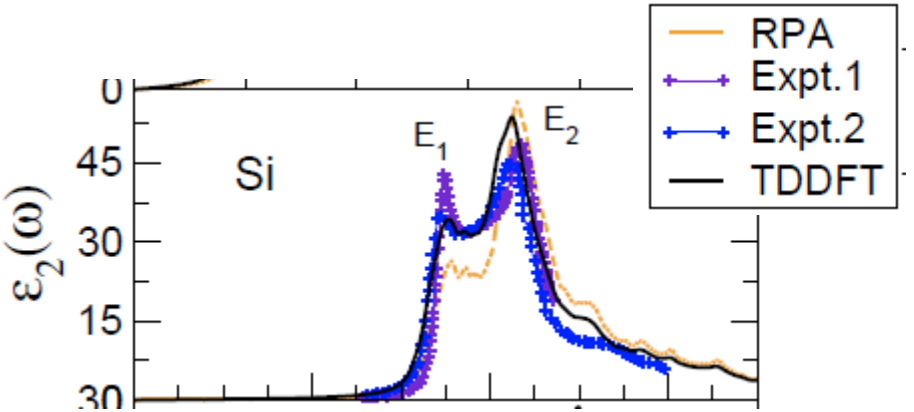
G. Onida, L. Reining, A. Rubio,
Rev. Mod. Phys. 74(2002)601.



TDDFT with hybrid (nonlocal) functional



Good results for small gap materials,
Less satisfactory for large gap materials.
J. Paier, M. Marsman, G. Kresse,
Phys. Rev. B78,121201 (2008)

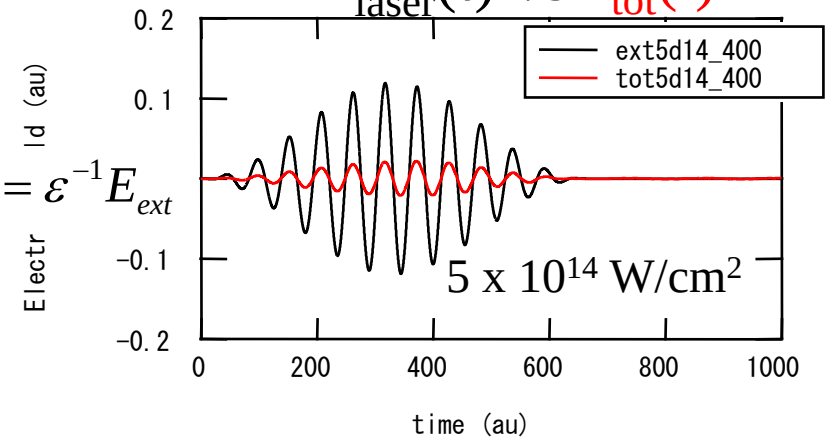


PRL107, 186401 (2011)
S. Sharma, J.K. Dewhurst, A. Sanna, E.K.U. Gross,
Bootstrap approx. for the exchange-correlation kernel
of time-dependent density functional theory

Limited to linear response regime.

As the laser intensity increases,
many electrons are excited.

$E_{laser}(t)$ vs $E_{tot}(t)$

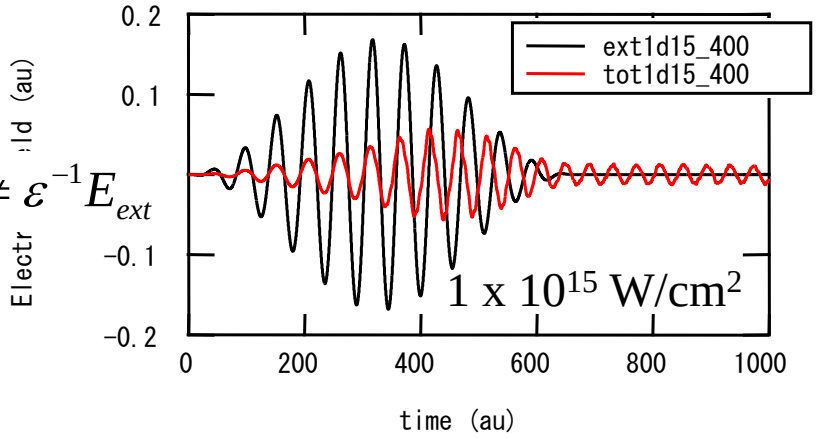


$$E_{tot} = \varepsilon^{-1} E_{ext}$$

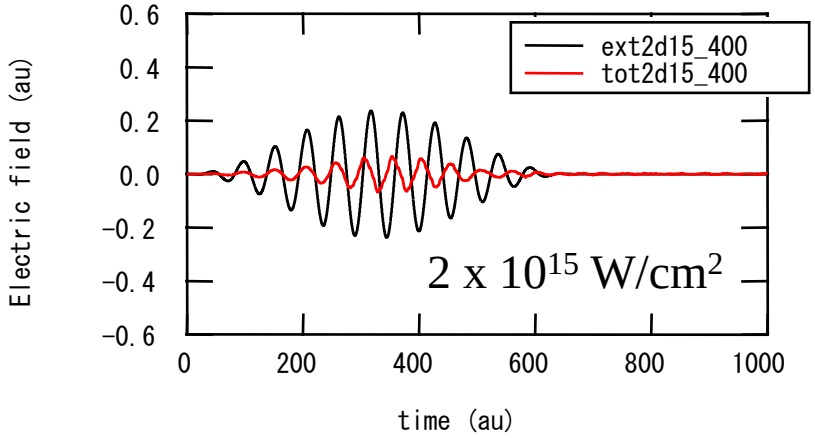
At a certain intensity,
we reach "critical density".

Plasma freq. of excited electrons
coincides with incident laser freq.

$$E_{tot} \neq \varepsilon^{-1} E_{ext}$$



T. Otobe, M. Yamagiwa, J.-I. Iwata, K.Y. T. Nakatsukasa,
G.F. Bertsch, Phys. Rev. B77, 165104 (2008)



Calculation for diamond
laser frequency: 3.1eV,
below bandgap (4.8 eV)

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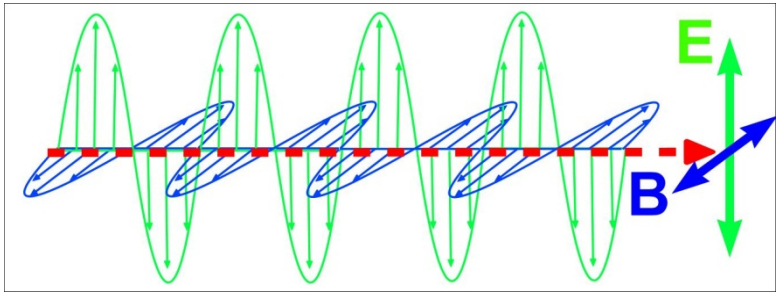
   Dielectric properties of solid surface excited by intense laser pulse

There are two spatial scales

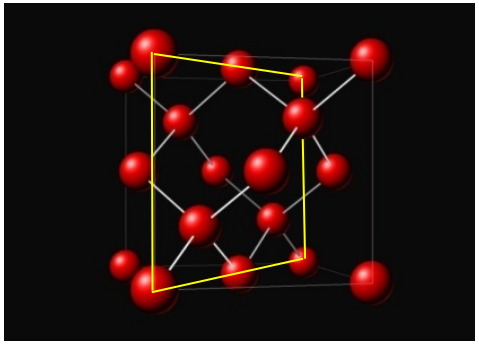
Laser pulse:
Macroscopic electromagnetic field

$$E(\vec{r},t), \quad B(\vec{r},t)$$



[μm]

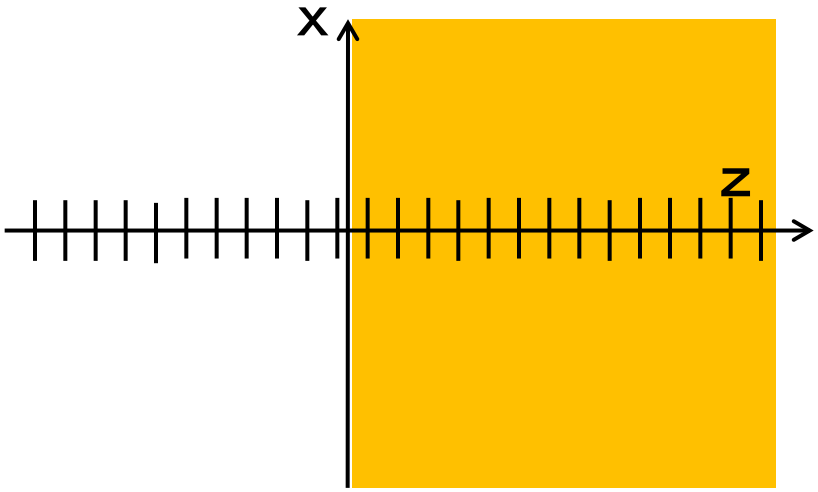
Microscopic electron dynamics:
Time-dependent Kohn-Sham orbitals



[nm]

We achieve “multiscale simulation”
introducing two kinds of spatial grids.

Consider a laser pulse
Irradiating normally on bulk Si surface



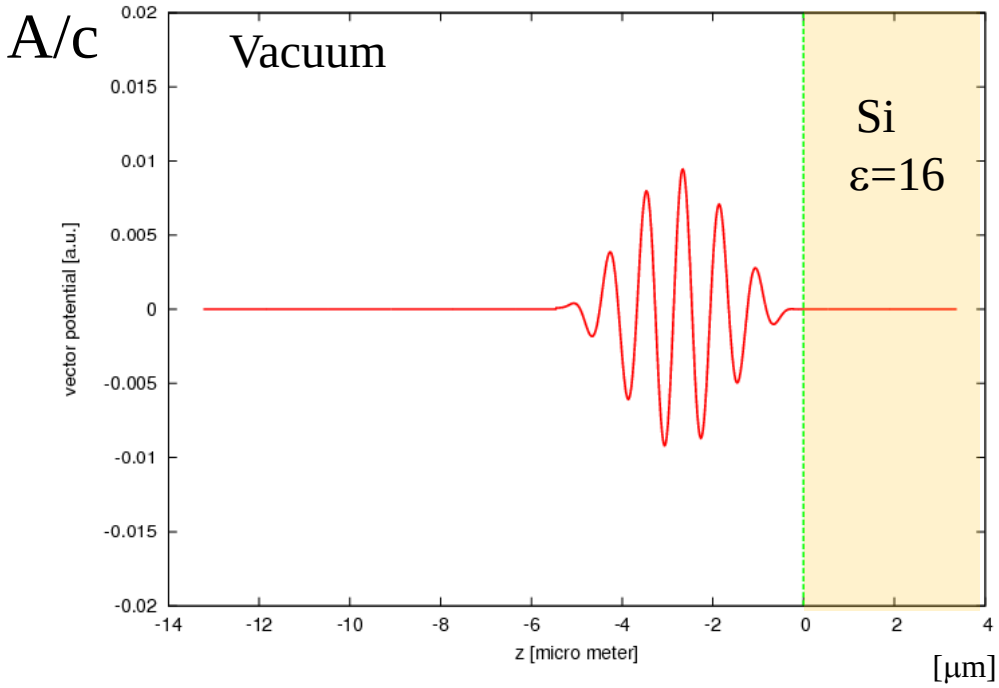
We solve Maxwell eq.

$$\frac{\varepsilon(z)}{c^2}\frac{\partial^2}{\partial t^2}A(z,t)-\frac{\partial^2}{\partial z^2}A(z,t)=0$$

Propagation z-direction, polarization x-direction

$$\vec{A}(\vec{r},t)=A(z,t)\hat{x}$$

$\lambda = 800\text{nm}, \quad \hbar\omega = 1.55\text{eV}$



Index of refraction

$$n=\sqrt{\varepsilon}$$

Reflectance

$$R=\left(\frac{1-n}{1+n}\right)^2$$

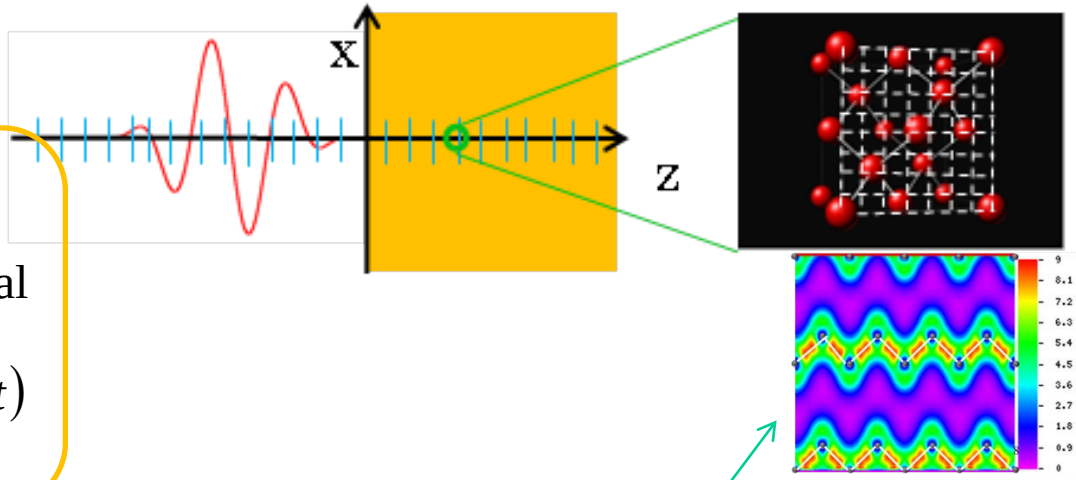
Velocity of wave

$$v=\frac{c}{n}$$

Multiscale calculation

At each macroscopic grid point,
We consider a unit cell and prepare microscopic grid.

K. Yabana, T. Sugiyama, Y. Shinohara, T. Otobe, G.F. Bertsch, Phys. Rev. B85, 045134 (2012).



Macroscopic grid points (μm)
to describe macroscopic vector potential

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

$J(Z,t)$

Exchange of information by macroscopic current and macroscopic vector potential.

$A(Z,t)$

$$J(Z,t)=\int_{\Omega} d\vec{r}\,\vec{j}_{e,Z}$$

$$\vec{j}_{e,Z}=\frac{\hbar}{2mi}\sum_i\left(\psi_{i,Z}^{*}\vec{\nabla}\psi_{i,Z}-\psi_{i,Z}\vec{\nabla}\psi_{i,Z}^{*}\right)-\frac{e}{4\pi c}n_{e,Z}\vec{A}$$

At each macroscopic points, Kohn-Sham orbitals $\psi_{i,Z}$ are prepared, and described in microscopic grids.

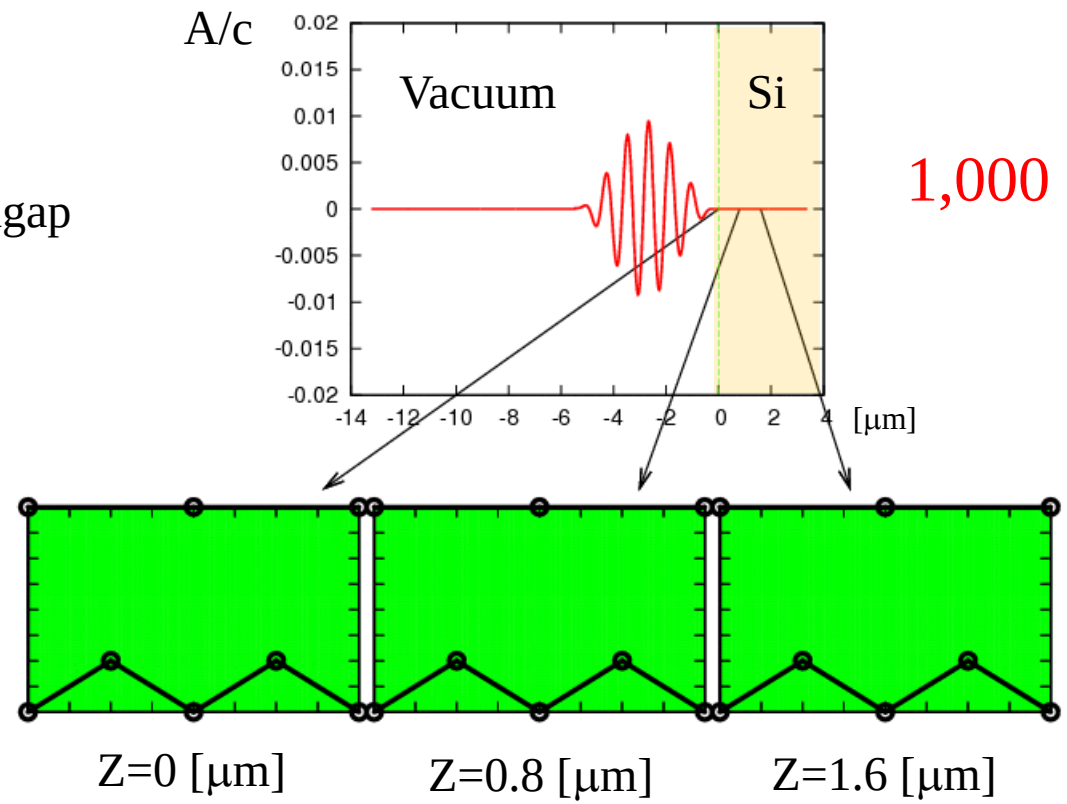
$$i\hbar\frac{\partial}{\partial t}\psi_{i,Z}=\frac{1}{2m}\left(-i\hbar\vec{\nabla}+\frac{e}{c}\vec{A}\right)^2\psi_{i,Z}-e\phi_Z\psi_{i,Z}+\frac{\delta E_{xc}}{\delta n}\psi_{i,Z}$$

$$\vec{\nabla}^2\phi_Z=-4\pi\left\{en_{ion}-en_{e,Z}\right\}$$

Propagation of weak pulse
Linear regime

$I=10^{10}\mathrm{W/cm^2}\quad \hbar\omega = 1.55\,\mathrm{eV}$

Laser frequency
below direct bandgap
2.4eV(LDA)

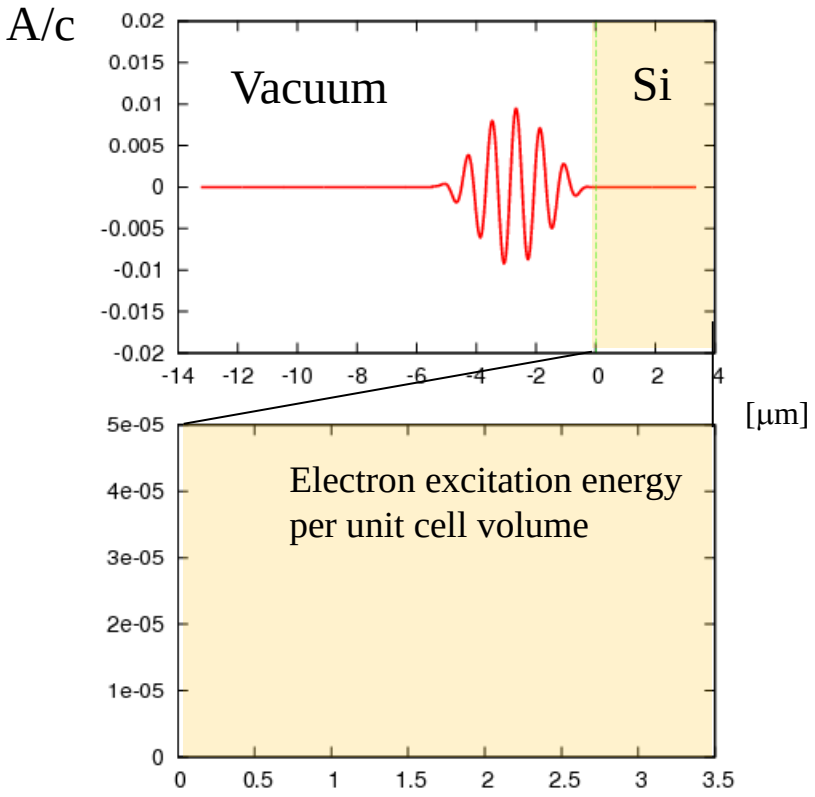


1,000 cores, 10 hours

Propagation of weak pulse
Linear regime

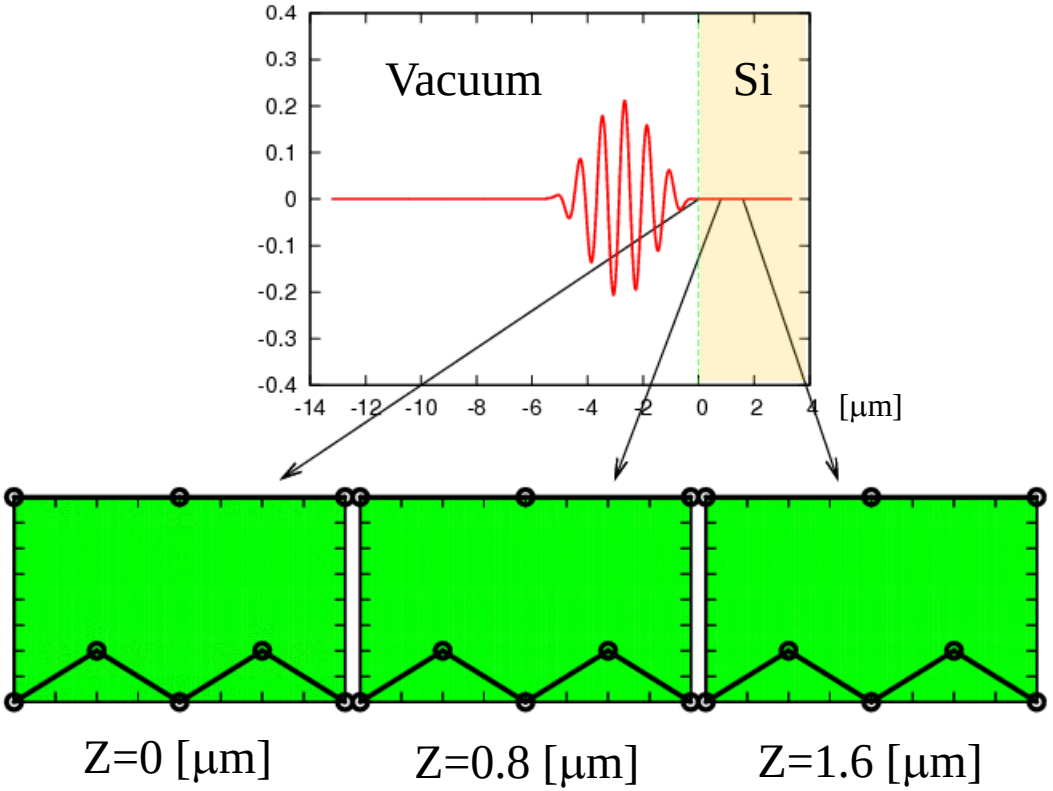
$I=10^{10}\mathrm{W/cm^2}\quad \hbar\omega = 1.55\,\mathrm{eV}$

Laser frequency
below direct bandgap
2.4eV(LDA)



More intense laser pulse
Maxwell and TDKS equations no more separate.

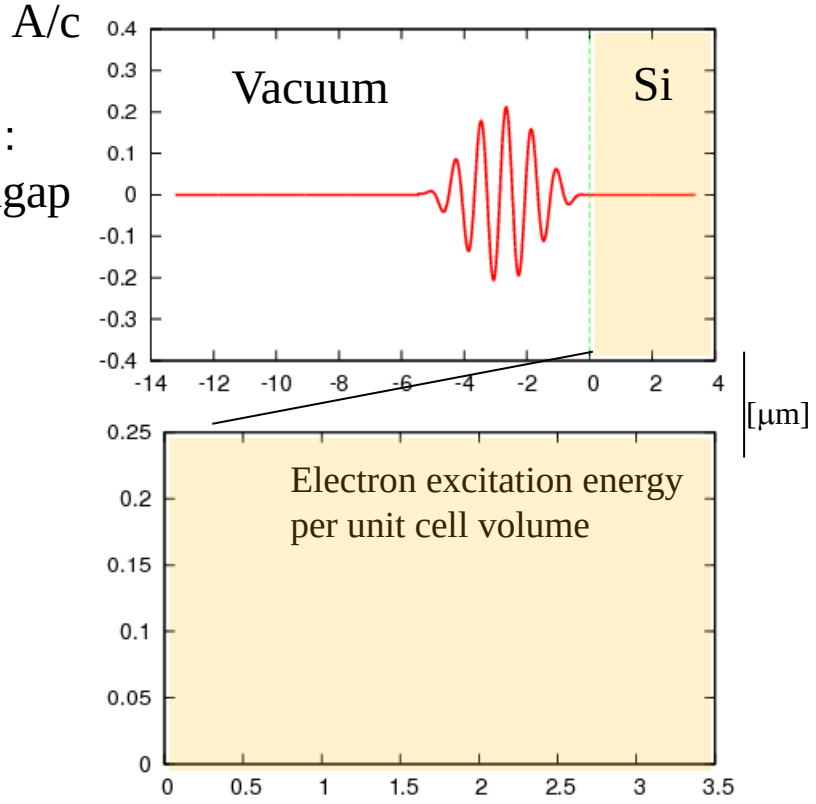
$I = 5 \times 10^{12} W/cm^2$



More intense pulse
(2-photon absorption dominates)

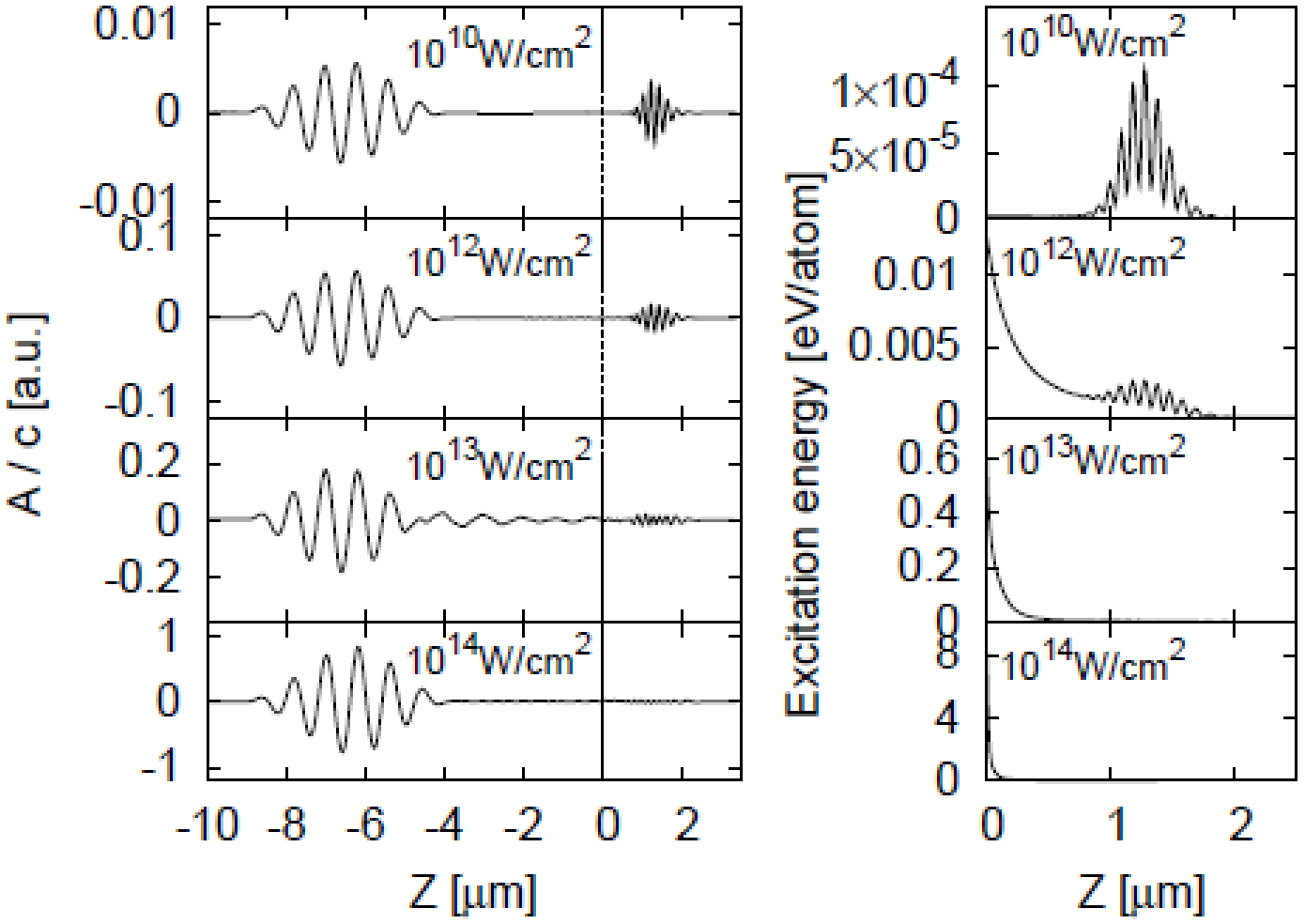
$I = 5 \times 10^{12} W/cm^2$

Laser frequency : 1.55eV :
lower than direct bandgap
2.4eV(LDA)



Reflected and transmitted waves

Excitation energy of electrons



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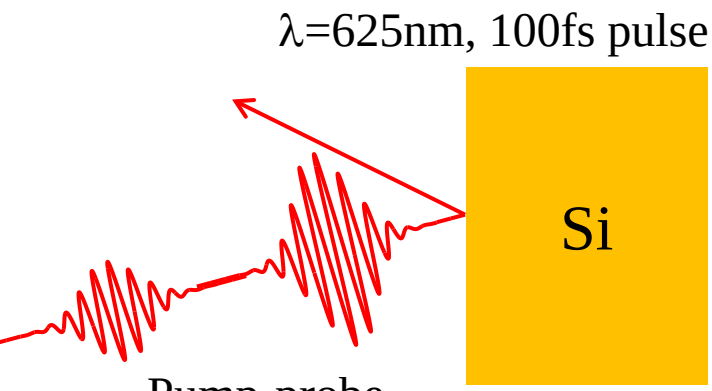
   Dielectric properties of solid surface excited by intense laser pulse

Dense electron-hole plasma generation at the surface modifies dielectric properties at the surface.

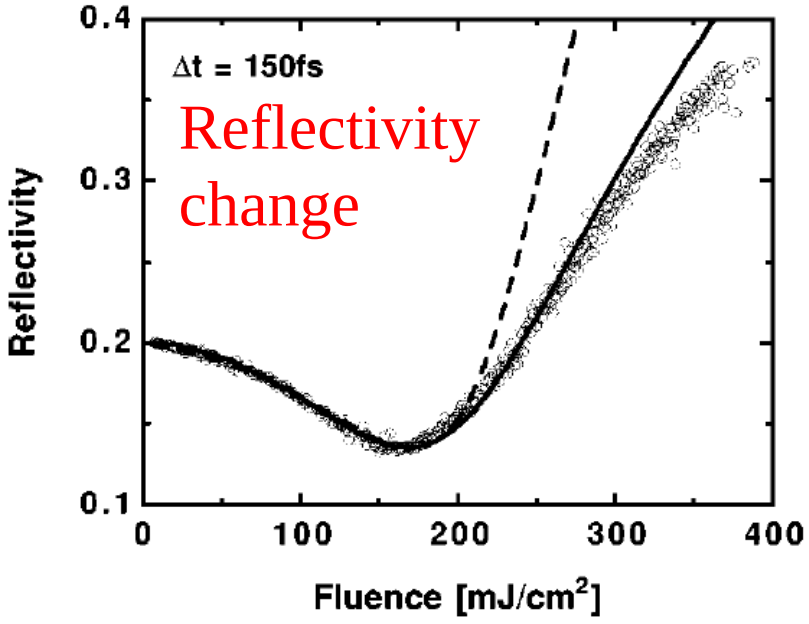


Pump-probe

Strong pump-pulse excites electrons at the surface, forming dense electron-hole plasma.

Probe-pulse measures change of dielectric property.

K. Sokolowski-Tinten, D. von der Linde, Phys. Rev. B61, 2643 (2000)



Pump-pulse intensity

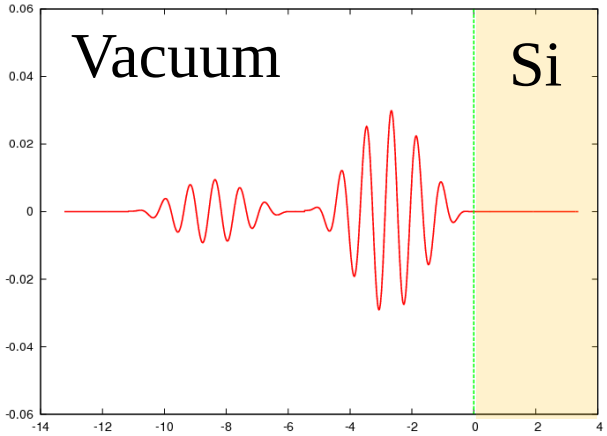
Drude model fit

$$\varepsilon\left(n_{ph}\right) = \varepsilon_{gs} - \frac{4\pi e^2 n_{ph}}{m^*} \frac{1}{\omega\left(\omega + \frac{i}{\tau}\right)}$$

m^*=0.18, τ=1fs

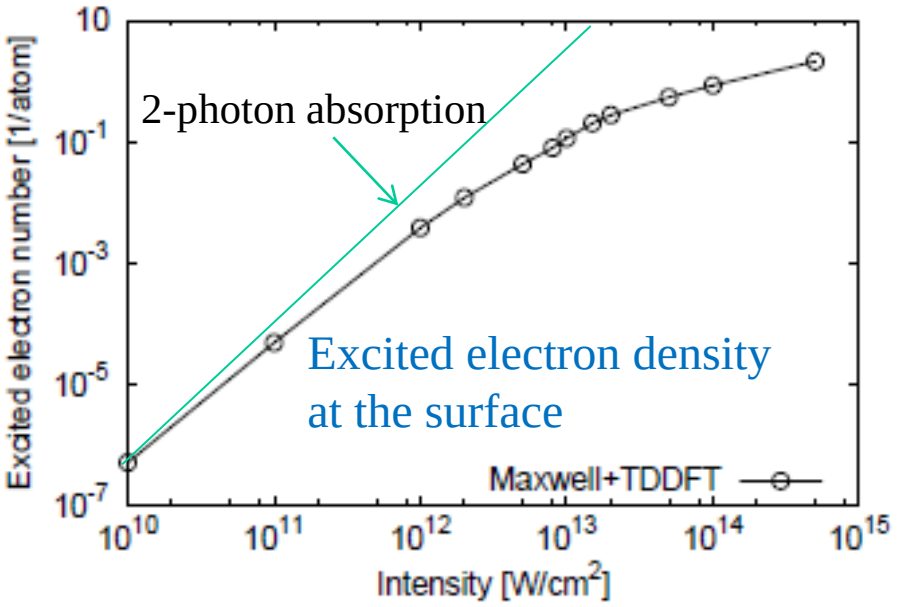
“Numerical Pump-Probe Experiment”

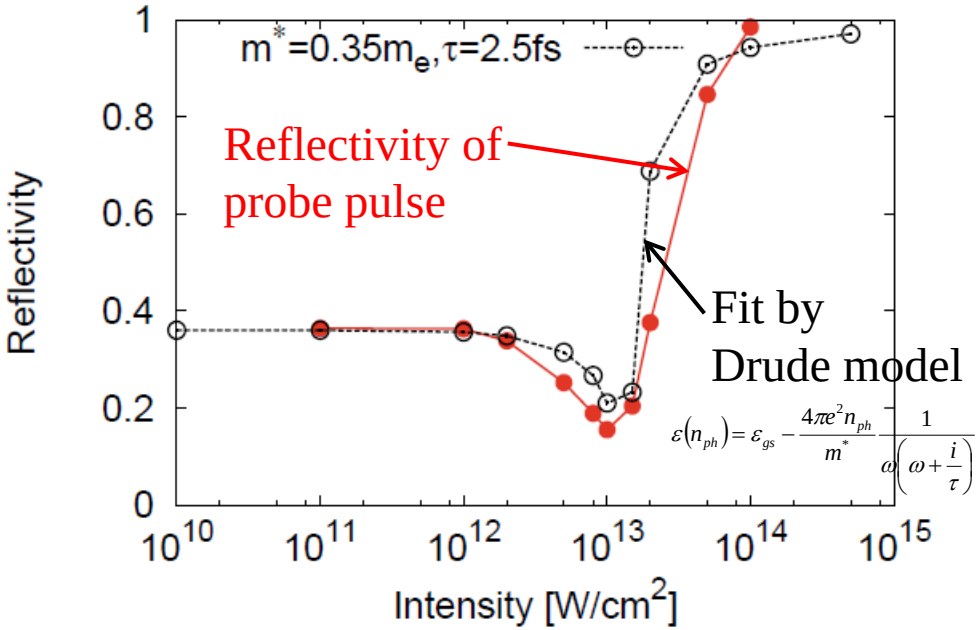
I=1x10^{11}W/cm^2,
$h\nu$=1.55eV



We can calculate

excited electron density at the surface by pump-pulse / reflectivity of probe pulse





We may extract "dielectric function of excited surface" from numerical pump-probe calculation

$$A(t) = A_i(t) + A_r(t)$$

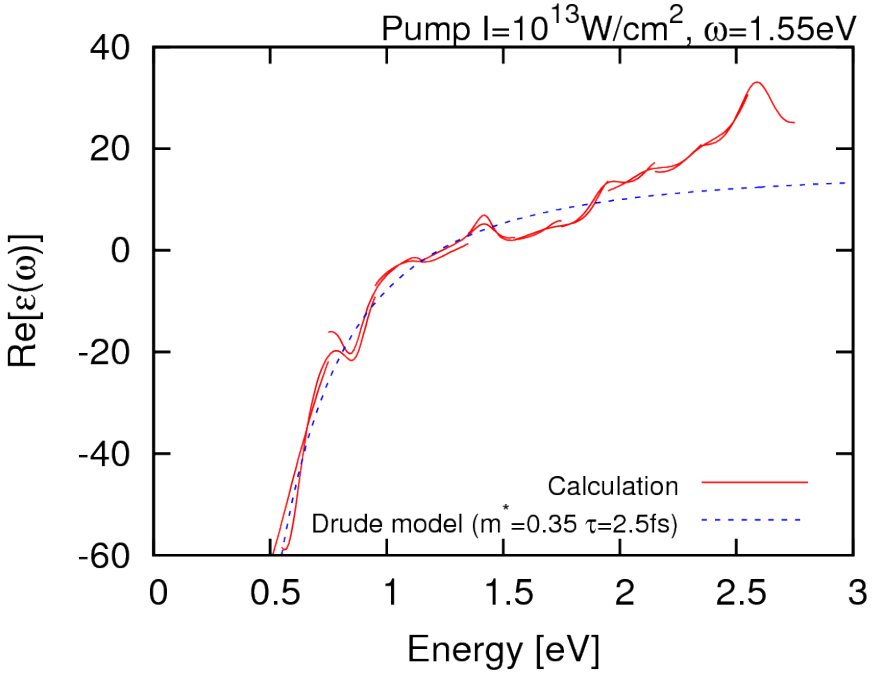
$$R(\omega) = \frac{\int dt\, e^{i\omega t} A_r(t)}{\int dt\, e^{i\omega t} A_i(t)}$$

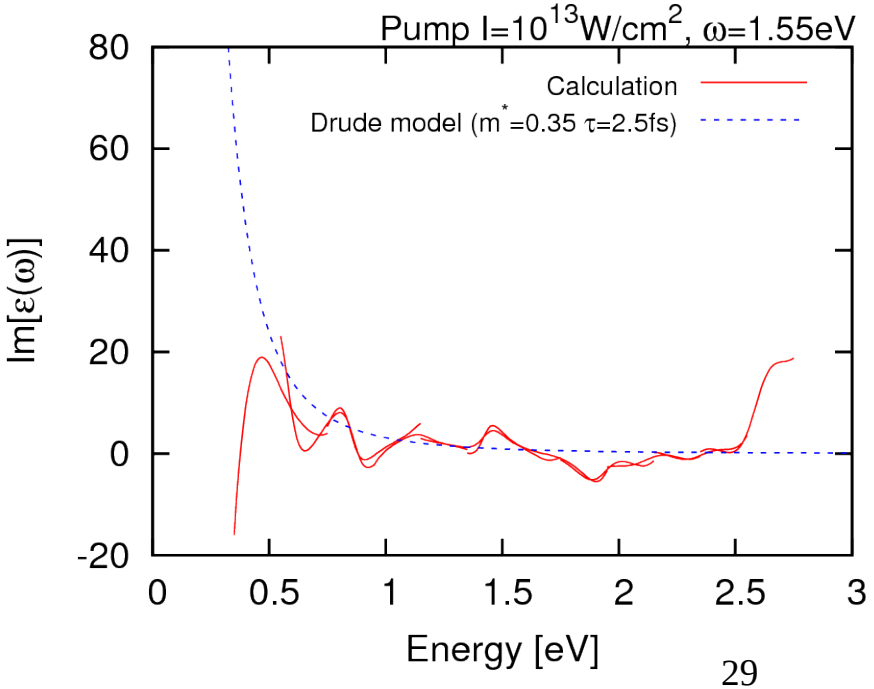
Decompose vector potential at the surface into incident and reflected components.

Ratio of two Fourier components gives us a reflectivity as a function of frequency.

$$R(\omega) = \frac{1-\sqrt{\varepsilon(\omega)}}{1+\sqrt{\varepsilon(\omega)}}$$

Dielectric function at the surface when irradiated by 10^{13}W/cm^2 pulse





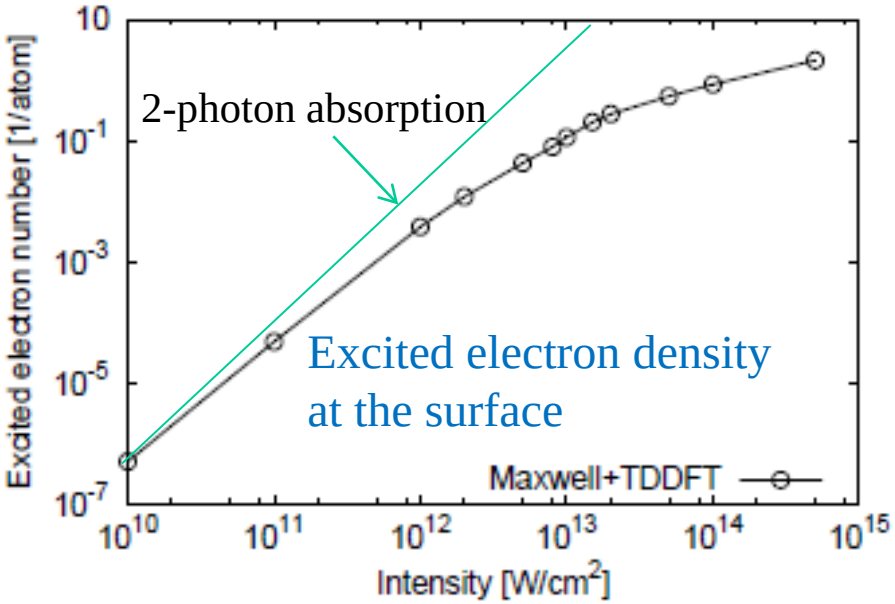
Real-part is well described by Drude model.

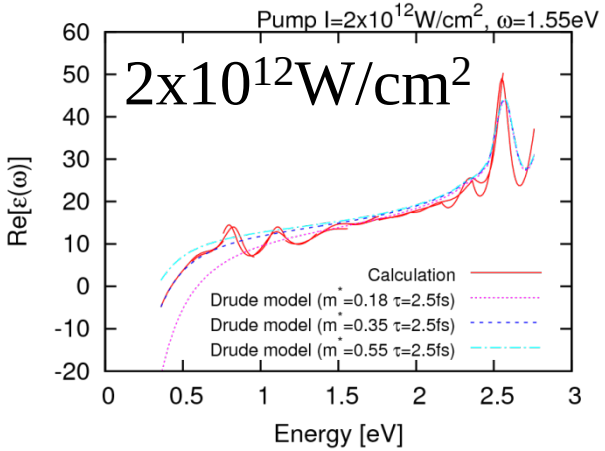
Change of dielectric function as we increase the pump-pulse intensity.

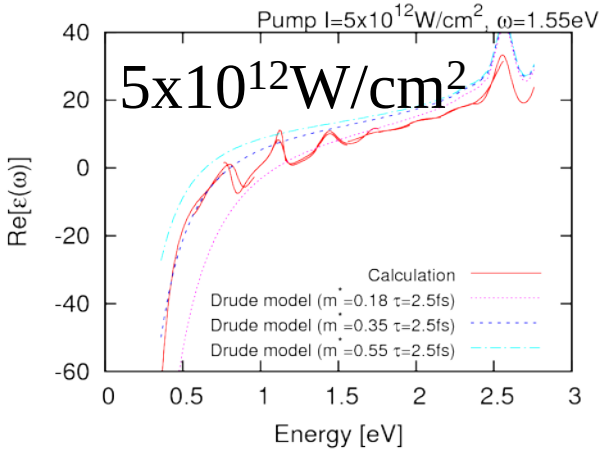
Poster by S.A. Sato

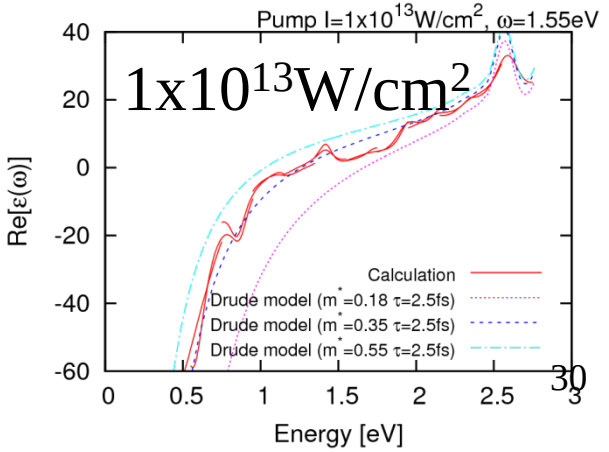
Increase of electron-hole density enhances metallic response.

$$\varepsilon(n_{ph}) = \varepsilon_{gs} - \frac{4\pi e^2 n_{ph}}{m^*} \frac{1}{\omega\left(\omega + \frac{i}{\tau}\right)}$$









Summary

Electron dynamics in bulk periodic solid by real-time TDDFT

- First-principles description is possible for electron dynamics
  in femto- and atto-second time scale

Coupled Maxwell + TDDFT multi-scale simulation

- Promising tool to investigate laser-solid interaction around breakdown threshold
- Requires large computational resources, a computational challenge

Further developments necessary

- Better energy functional to increase accuracy of results
- How to treat collision effects (electron-electron, electron-phonon)
- Extension towards
  surface electron dynamics, core-electron dynamics, …