Real-time electron dynamics in solids under strong electromagnetic fields

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TDDFT: First-principles tool for electron dynamics simulation

\[ i\hbar \frac{\partial}{\partial t} \psi_i(\mathbf{r}, t) = \{h_{KS}[\rho(\mathbf{r}, t)] + V_{ext}(\mathbf{r}, t)\}\psi_i(\mathbf{r}, t) \]

Linear response regime (Perturbation theory)

\[ \rho(\mathbf{r}, t) = \sum_i |\psi_i(\mathbf{r}, t)|^2 \]

Laser-solid interaction

Dielectric function

Absorption cross section

Electronic excitation

Collision of atoms and ions

Intense, ultra-short Laser pulses

Multiple ionization

Soft X ray (High harmonic generation)

Coulomb explosion

Conductance of nano-wire

Nonlinear, nonperturbative regime (Initial value problem)
1. A frontier in light - matter interaction : intense and ultrashort
   - Theories and computations required in current optical sciences -

2. Real-time and real-space calculation for electron dynamics in solid
   - time-dependent band calculation, treatment of boundaries. -

3. Electron and phonon dynamics in solid for a given electric field
   - Dielectric function, coherent optical phonon, optical breakdown, … -

4. Coupled Maxwell + TDDFT multiscale simulation
   - First-principles simulation for macroscopic electromagnetic field in intense regime,
     Dense electron-hole plasma induced by intense laser pulses -
Modern experiments on light-matter interactions

Requiring new theories and large-scale computations, beyond ordinary electromagnetism and quantum mechanics

- Nano-sized material
too large to calculate microscopically, too small to calculate macroscopically.
  \[\text{large-scale microscopic calculation}\]

- Near field optics (resolution beyond diffraction limit)
- Meta-material (negative refractive index)
- Surface plasmon polariton (coupled light-electron dynamics)
  \[\text{quantum effects (?)}\]

- Intense and ultrashort laser pulses (extreme nonlinear electron dynamics)
  requiring time-domain description, coupled dynamics of electrons and electromagnetic fields
Frontiers in Optical Sciences: Intense field

FIG. 1: Maximum laser intensity as a function of time and fields of research accessible with these intensities.
Intense laser pulse on solid

- Laser intensity: $10^{13} - 10^{15} \text{ W/cm}^2$
- Electron-hole plasma
- Optical breakdown
- Laser machining

Nonlinear optics | Coherent phonon | HHG | Laser acceleration | Vacuum breakdown

Nonrelativistic Quantum mechanics | Relativistic Classical mechanics

$eE(t)z$ Z

External electric field by laser pulse
$\approx$ Internal electric field by nuclei
Real-time observation of valence electron motion
E. Goulielmakis et al., Nature 466, 739 (2010).
Pump-Probe Measurement: Coherent Phonon

Intense 1\textsuperscript{st} pulse excites electrons and atoms.
Weak 2\textsuperscript{nd} pulse measure change of reflectivity.

\[ \frac{\Delta R(t)}{R} \propto e^{-rt} \sin(\Omega_{ph} t + \phi) \]

Optical microscope image of waveguides written inside bulk glass by a 25-MHz train of 5-nJ sub-100-fs pulses, C.B. Schaffer et.al, OPTICS LETTERS 26, 93 (2001).


Micromachining – waveguide- Nanosurgery

Optical microscope image of waveguides written inside bulk glass by a 25-MHz train of 5-nJ sub-100-fs pulses, C.B. Schaffer et.al, OPTICS LETTERS 26, 93 (2001)

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Calculation in real-time and real-space

Time-dependent Kohn-Sham equation

\[ i\hbar \frac{\partial}{\partial t} \psi_i(\vec{r}, t) = \hbar_{KS}[n(\vec{r}, t)]\psi_i(\vec{r}, t) \quad n(\vec{r}, t) = \sum_i |\psi_i(\vec{r}, t)|^2 \]

\[ \hbar_{KS}[n(\vec{r}, t)] = -\frac{\hbar^2}{2m} \nabla^2 + V_{\text{ion}}(\vec{r}) + \int d\vec{r}' \frac{e^2}{|\vec{r} - \vec{r}'|} + \mu_{xc}[n(\vec{r}, t)] + V_{\text{ext}}(\vec{r}, t) \]

Norm-conserving pseudopotential

3D real-space grid representation, high order finite difference

\[ -\frac{\hbar^2}{2m} \left[ \sum_{n_1 = -N}^N C_{n_1} \psi(x_i + n_1 h, y_j, z_k) + \sum_{n_2 = -N}^N C_{n_2} \psi(x_i, y_j + n_2 h, z_k) + \sum_{n_3 = -N}^N C_{n_3} \psi(x_i, y_j, z_k + n_3 h) \right] \]

\[ + [V_{\text{ion}}(x_i, y_j, z_k) + V_{H}(x_i, y_j, z_k) + V_{\text{xc}}(x_i, y_j, z_k)]\psi(x_i, y_j, z_k) = E \psi(x_i, y_j, z_k) . \]

Time evolution: 4-th order Taylor expansion

\[ \psi_i(t + \Delta t) = \exp \left[ \frac{\hbar_{KS}(t) \Delta t}{i\hbar} \right] \psi_i(t) \approx \sum_{k=0}^N \frac{1}{k!} \left( \frac{\hbar_{KS}(t) \Delta t}{i\hbar} \right)^k \psi_i(t), \quad N = 4 \]
Treatment of optical electric field

We assume a long wavelength limit, lattice const. $\ll$ light wavelength

Electron dynamics in a unit cell under time-dependent, spatially uniform field

Two key issues

Use (spatially uniform) vector potential instead of (spatially linear) scalar potential to express macroscopic electric field.

Boundary effect (macroscopic shape effect) needs to be considered from outside of the theory.
Electron dynamics in crystalline solid under spatially uniform field

Employing a vector potential replacing the scalar potential, one may recover the periodicity of the Hamiltonian

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

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

\[ h(\vec{r} + \vec{a}) \neq h(\vec{r}) \]

Time-dependent, spatially uniform electric field

\[ \vec{E} = -\vec{\nabla} \phi - \frac{1}{c} \frac{\partial \vec{A}}{\partial t} \]

\[ \phi = E(t)z \quad \Leftrightarrow \quad \vec{A}(t) = c \int dt' E(t') \hat{z} \]

\[ h(\vec{r} + \vec{a}) = h(\vec{r}) \quad \psi_{nk}(\vec{r} + \vec{a}) = e^{ik\vec{a}} \psi_{nk}(\vec{r}) \]

Time-dependent Bloch function
Equation for time-dependent Bloch function

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

\[ n(\vec{r}, t) = \sum_{nk} |u_{nk}(\vec{r}, t)|^2 \]

\[ \psi_{nk}(\vec{r}, t) = e^{i\kappa r} u_{nk}(\vec{r}, t) \]

\[ u_{nk}(\vec{r} + \vec{a}, t) = u_{nk}(\vec{r}, t) \]
Electron dynamics in metallic clusters by TDDFT


Na$_{147}^+$

Li$_{147}^+$

Assume Icosahedral shape

Electron dynamics in metallic clusters by TDDFT

Collective electronic excitation: plasmon

Longitudinal plasmon

\[ \omega_p = \left( \frac{4\pi e^2 n}{m} \right)^{1/2} \]

\( n \): electron density

Mie plasmon
(Surface plasmon of spherical nanoparticle)

\[ \omega_{Mie} = \frac{\omega_p}{\sqrt{3}} \]
Choice of geometry (boundary condition)

- We solve electron dynamics in a unit cell inside a crystalline solid.
- The electric field in the unit cell depends on macroscopic geometry of the sample.
- We need to specify the ‘boundary condition’ in the calculation.

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

\[
\vec{E} = -\frac{1}{c} \frac{\partial \vec{A}}{\partial t}, \quad \vec{A}(t) = c \int dt' E(t') \hat{\mathbf{z}}
\]

Optical response of thin film

Transverse
\[
A(t) = A_{\text{ext}}(t)
\]

Longitudinal
\[
A(t) = A_{\text{ext}}(t) + A_{\text{polarization}}(t)
\]
**Longitudinal geometry:**

**Treatment of induced polarization**


“parallel-plate-capacitor with dielectrics”.

\[
A(t) = A_{\text{extr}}(t) + A_{\text{polarization}}(t)
\]

**Equation for polarization**

\[
\frac{d^2 \mathbf{A}_{\text{polarization}}(t)}{dt^2} = \frac{4\pi}{\Omega} \int_{\Omega_{(cell)}} d\mathbf{r} \mathbf{j}(\mathbf{r}, t)
\]

**Time-dependent Kohn-Sham equation**

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

\[
n = \sum_i |\psi_i|^2 \quad \mathbf{j} = \frac{1}{2m} \sum_i \left( \psi_i^* \left( \mathbf{p} + \frac{e}{c} \mathbf{A} \right) \psi_i - \text{c.c.} \right)
\]

\[
E(t) = -\frac{1}{c} \frac{\partial A(t)}{\partial t}
\]

\[
A(t) = -c \int E(t')dt'
\]
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Linear response:

Two methods to calculate dielectric function from real-time electron dynamics

**Transverse geometry**

\[ J(t) = \int dt' \sigma(t - t') \left( -\frac{1}{c} \frac{dA}{dt} \right) \]

\[ \sigma(\omega) = \int dt e^{i\omega t} \sigma(t) \]

\[ \varepsilon(\omega) = 1 + \frac{4\pi i \sigma(\omega)}{\omega} \]

**Longitudinal geometry**

\[ A_{tot}(t) = A_{ext}(t) + A_{pol}(t) \]

\[ \frac{d^2 A_{pol}(t)}{dt^2} = 4\pi J(t) \]

\[ A_{tot}(t) = \int dt' \varepsilon^{-1}(t-t')A_{ext}(t') \]

\[ \frac{1}{\varepsilon(\omega)} = \frac{\int dt e^{i\omega t} A_{tot}(t)}{\int dt e^{i\omega t} A_{ext}(t)} \]

For an impulsive external field,

\[ E(t) = k\delta(t), \quad A(t) = -kc \theta(t) \]

\[ J(t) = k\sigma(t) \]

\[ E_{tot}(t) = -\frac{1}{c} \frac{dA_{tot}}{dt} = k\varepsilon^{-1}(t) \]
Comparison of numerical results for dielectric function of Si by two methods
- Green: longitudinal
- Red: transverse

Spatial grid: \(16^3\)
K-points: \(8^3\)

Distortion strength:
\(A_0 = 0.01\) (with induced),
\(0.0005\) (without)

\[\text{Re}[\varepsilon]\]

\[\text{Im}[\varepsilon]\]

\[J(t)/A_0\]
Dielectric function of Si in TDDFT (ALDA)

Transverse calculation

Instantaneous pulse field is applied at $t=0$, the induced current as a function of time.

Dielectric function by TDDFT (ALDA) is not very good.
Too small bandgap (common problem with LDA).
Dots: experiment  
Dash-dotted: RPA  
Solid: Bethe-Salpeter  
(electron-hole interaction considered)

FIG. 5. Silicon absorption spectrum $[\text{Im}(\varepsilon_M)]$: •, experiment (Lautenschlager et al., 1987); dash-dotted curve, RPA, including local field effects; dotted curve, $GW\text{-RPA}$; solid curve, Bethe-Salpeter equation.


Pump-Probe Measurement: Coherent Phonon

Intense 1\textsuperscript{st} pulse excites electrons and atoms.
Weak 2\textsuperscript{nd} pulse measure change of reflectivity.

\[ \Delta \Delta t \propto e^{-\Gamma t} \sin(\Omega_{ph}t + \phi) \]

\[ T_{ph} : \text{Period of optical phonon} \]

The two mechanisms for coherent phonon generation

**Virtual** electronic excitation  
(transparent material)

\[ V = \frac{1}{2} \chi |E(t)|^2, \quad F = -\frac{\partial V}{\partial q} \]

- Impulsive  
  Force appears only during laser irradiation

**Real** electronic excitation  
(opaque material)

\[ 1 - \cos(\Omega_{ph} t) \]

- Displacive  
  Change of equilibrium

\[ \Delta Q \]

---


Coherent phonon measurements in Sb

- Most popular material for coherent phonon generation
- Observed two modes: $A_{1g}$, $E_g$

Sb: crystal structure and phonon mode

- Semimetal
- Rhombohedral structure

Having C3 symmetry axis parallel to vertical line in following figure

Each atom have three bonds
Only one bond on plane
Other two bonds are out of plane

There are two modes:
A1g and Eg (doubly degenerate)
**Electron density under laser pulse**


During laser pulse: electrons moves atom to atom
After laser pulse ends: bonding electrons are reduced
Density change and force

Electron density at ground state

Density difference on plane-1

Density difference on plane-2

Electron density at ground state

Including electric field Out of electric field

Force is dominated by real-excitation (removal of bonding electrons)
Coherent phonon measurements in Sb

- Most popular material for coherent phonon generation
- Observed two modes: $A_{1g}$, $E_g$

However, calculation is not consistent with measurement. Experimentally, $A_{1g}$: displacive, $E_g$: impulsive.
TDDFT calculation for coherent phonon in Si


When laser frequency is below the direct gap, impulsive (Raman tensor) force is visible.
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Weak field regime: Standard approach for light-matter interaction

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

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

### Constitution relation

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

**Intense laser pulse induced nonlinear electron dynamics**

Quantum mechanics is useful to calculate nonlinear susceptibilities as well.

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

**Further increase of laser pulse intensity**

Perturbative expansion is no more useful.
Dense electron-hole plasma, Optical breakdown, etc.

We need to solve couple Maxwell + Schrodinger dynamics.
We solve coupled Maxwell + Schroedinger equations.

Maxwell equation describing electromagnetic field

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

Time-dependent density-functional theory describing electron dynamics

\[ \psi_i(\vec{r},t) \]

polarization

external field

[\text{\mu m}] \quad \text{Different spatial scales} \quad [\text{nm}]

Time-dependent Kohn-Sham eq.

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

\[ n = \sum_i |\psi_i|^2 \]

\[ j = \frac{1}{2im} \sum_i \psi_i^* \left( -i\hbar \vec{\nabla} + \frac{e}{c} \vec{A} \right) \psi_i - \text{c.c.} \]

Maxwell eq.

\[ \frac{1}{c^2} \frac{\partial^2 \vec{A}}{\partial t^2} - \vec{\nabla}^2 \vec{A} = \frac{4\pi}{c} \vec{j} \]

\[ \vec{\nabla}^2 \phi = -4\pi \{ e n_{ion} - e n_e \} \]

Multiscale simulation introducing two spatial grids
Laser pulse irradiation normal on bulk Si

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

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

Laser frequency below direct bandgap

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

Index of refraction

\( n = \sqrt{\varepsilon} \)

Reflectance

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

Velocity of wave

\[ v = \frac{c}{n} \]
Multiscale simulation


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

Macroscopic grid points (μm) to describe macroscopic vector potential

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

Exchange of information by macroscopic current and macroscopic vector potential.

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

\[
J(Z,t) = \int d\vec{r} \vec{j}_{e,z}
\]

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

\[
\psi_{i,z} = \frac{1}{2m} \left( -i\hbar \vec{\nabla} + \frac{e}{c} \vec{A} \right)^2 \psi_{i,z} - e \phi_{z} \psi_{i,z} + \frac{\delta E_{xc}}{\delta n} \psi_{i,z}
\]

\[
\nabla^2 \phi_z = -4\pi \left\{ en_{ion} - en_{e,z} \right\}
\]
Each CPU calculates electron dynamics at different macroscopic position.

\[ J_{Z \pm 1}(t) \]
\[ A_{Z \pm 1}(t) \]

(We also parallelize with respect to k-points)

Macroscopic vector potential requires A and j of nearby points to evolve in time. (only tens of bytes exchanged)

Each CPU calculates nonlinear electron dynamics independently.
Propagation of weak pulse
(Linear response regime, separate dynamics of electrons and E-M wave)

$I = 10^{10} \text{W/cm}^2$

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

\[ Z = 0 \quad \mu \text{m} \]
\[ Z = 0.8 \quad \mu \text{m} \]
\[ Z = 1.6 \quad \mu \text{m} \]
Propagation of weak pulse
(Linear response regime, separate dynamics of electrons and E-M wave)

$I = 10^{10}$ W/cm$^2$

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

Electron excitation energy per unit cell volume

Vacuum

Si

A/c
Maxwell+TDDFT vs Maxwell only (constant $\varepsilon$)

Red = Maxwell + TDDFT

Green = Maxwell

$$\frac{\varepsilon}{c^2} \frac{\partial^2}{\partial t^2} A(z,t) - \frac{\partial^2}{\partial z^2} A(z,t) = 0 \quad \varepsilon = 16$$

In linear regime, dispersion of dielectric function Induces Phase velocity vs group velocity

$$v = \frac{c}{\sqrt{\varepsilon}} \quad v_g = \frac{c}{\sqrt{\varepsilon} \left( 1 + \frac{\omega}{2\varepsilon} \frac{d\varepsilon}{d\omega} \right)}$$

Chirp effect is also seen
More intense laser pulse
Maxwell and TDKS equations no more separate.

\[ I = 5 \times 10^{12} \text{W/cm}^2 \]
More intense pulse
(2-photon absorption dominates)

\[ I = 5 \times 10^{12} \text{W/cm}^2 \]

Laser frequency: 1.55eV: lower than direct bandgap 2.4eV (LDA)
Vector potential and electronic excitation energy
After the laser pulse splits into reflected and transmitted waves
Energy Conservation

\[ L[\psi, \phi, A] = \int dZ \left[ \sum_i \int_{\Omega(Z)} d\vec{r} \psi_{i,Z}^*(\vec{r}, t) i\hbar \frac{\partial}{\partial t} \psi_{i,Z}(\vec{r}, t) \right. \]

\[- \frac{1}{2m} \left| \left( -i\hbar \vec{\nabla} - \frac{e}{c} \vec{A}(t) \right) \psi_{i,Z}(\vec{r}, t) \right|^2 \]

\[ + \int_{\Omega(Z)} d\vec{r} \left\{ e \left( n_{ion,Z}(\vec{r}) - n_{e,Z}(\vec{r}, t) \right) \phi_Z(\vec{r}, t) - E_{xc}[n_Z(\vec{r}, t)] \right\} \]

\[ + \int_{\Omega(Z)} d\vec{r} \frac{1}{8\pi} (\vec{\nabla}_r \phi_Z(\vec{r}, t))^2 + \frac{\Omega}{8\pi c^2} \left( \frac{\partial^2 \vec{A}(t)}{\partial t^2} \right)^2 + \frac{\Omega}{8\pi} (\vec{\nabla}_Z \times \vec{A}(t))^2 \]

Our multiscale equations can be derived from the Lagrangian.

We can derive conserved energy which is a sum of:
- energy of EM field
- energy of electrons
- interaction energy between them.

\[ H = \int dZ \left[ \sum_i \int_{\Omega(Z)} \frac{1}{2m} \left| \left( -i\hbar \vec{\nabla} - \frac{e}{c} \vec{A}(t) \right) \psi_{i,Z}(\vec{r}, t) \right|^2 \right. \]

\[ + \left. \int_{\Omega(Z)} d\vec{r} \left\{ \frac{e}{2} \left( n_{ion} - n_{e,Z}(\vec{r}, t) \right) \phi_Z(\vec{r}, t) + E_{xc}[n_Z(\vec{r}, t)] \right\} \right\} \]

\[ + \frac{\Omega}{8\pi c^2} \left( \frac{\partial \vec{A}(t)}{\partial t} \right)^2 + \frac{\Omega}{8\pi} (\vec{\nabla}_Z \times \vec{A}(t))^2 \]
Energy conservation well satisfied in practical calculations.
Computational aspect

Microscopic (Schroedinger)
spatial grid: $16^3$
k-points : $8^3$ (reduced by symmetry to 80)
(too small)

Time step (common) = 16,000

Macroscopic (Maxwell)
spatial grid: 256

3.5$\mu$m, 256 points
$\Delta z = 13$nm

Computational costs:

1024 cores (MPI only), 10 hours at SGI Altix ICE 3800EX (ISSP, Univ. Tokyo)

20,480 cores (MPI+openmp), 20 min. at K-Computer
Dense electron-hole plasma generation at the surface modifies dielectric properties at the surface.

\[ \lambda = 625\text{nm, 100fs pulse} \]

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


Probe-pulse measures change of Dielectric properties.

Drude model fit

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

\[ m^* = 0.18, \tau = 1\text{fs} \]
Our Pump-Probe “Numerical Experiment”

\[ I = 1 \times 10^{11} \text{W/cm}^2, \quad h\nu = 1.55 \text{eV} \]

We can calculate excited electron density at the surface by pump-pulse / reflectivity of probe pulse.

Excited electron density

\[ n_e = \frac{4\pi e^2 n_{ph}}{m^*} \left( \frac{1}{\omega + i/\tau} \right) \]

Reflectivity of probe pulse

\[ R = \frac{1}{1 + \left( \frac{\omega}{\omega_c} \right)^2} \]

Fit by Drude model

\[ m^* = 0.35m_e, \tau = 2.5 \text{fs} \]
We may extract "dielectric function of excited surface" from numerical pump-probe calculation.

\[
A(t) = A_i(t) + A_r(t)
\]

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

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

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

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

Well described by Drude model (at least for real part).
Change of dielectric function as we increase the pump-pulse intensity.

Increase of electron-hole density enhances metallic response.

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

2-photon absorption

[Graph showing excited electron density at the surface vs. intensity]
Computational Aspects and Future Prospects

Large computational resources required for multi-scale calculation

At present
Normal incidence of linearly polarized pulse on Si crystal:
1,000 cores, 10 hours
30,000 cores, 20 min (K-computer, Kobe)

More resources to achieve
• More complex dielectrics (SiO₂, TiO₂, ZrO₂, …) x 10
• Oblique incidence (2-dim) x 50
• Self-focusing (3-dim) x 1000
• Circular polarization (3-dim) x 1000

K-Computer, Japan
- 640,000 cores, ¥100,000,000,000
Summary

Electron dynamics in bulk periodic solid by real-time TDDFT

- First-principles description of electron dynamics in femto- and atto-second time scale

- Applications to
  - dielectric function (linear response)
  - coherent phonon generation
  - optical breakdown

Coupled Maxwell + TDDFT multi-scale simulation

- Promising tool to investigate laser-matter interaction

- Requires large computational resources, a computational challenge

- Further developments necessary
  Surface treatment, longitudinal electromagnetic wave, 2D,3D propagation, collision effects (e-e, e-v)