7 th Time-Dependent Density Functional Theory
Benasque, Spain
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Maxwell + TDDFT multiscale descriptions for interactions of intense pulsed light with dielectrics

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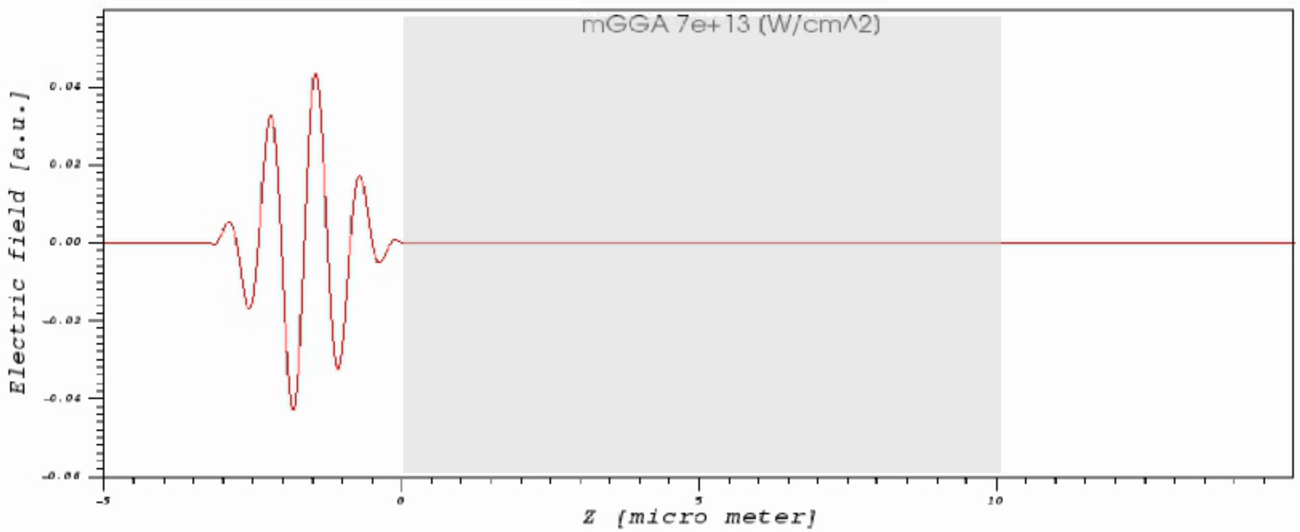
Maxwell + TDDFT multiscale simulation for intense and ultrashort laser pulse propagation in dielectrics

10 μm SiO_2 thin film

$\hbar\omega = 1.55\text{eV}$

λ=800nm

$I = 7 \times 10^{13} \text{W/cm}^2$



Laser electric field, red (strong), blue (weak)

Expensive calculation: 80,000 cores, 20 hours at K-Computer, Japan

- Why and when it is necessary?
- How it is done?
- How it works?

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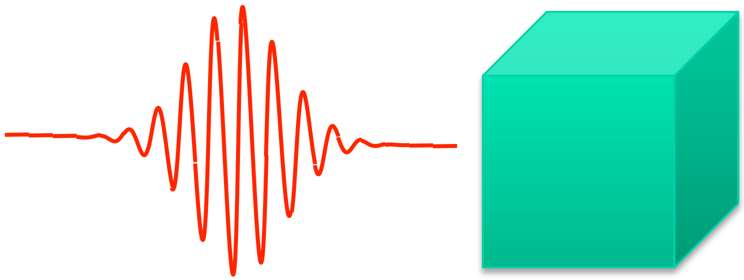
Ordinary theory and calculation for light-matter interactions



Macroscopic electromagnetism for light propagation in matter

Quantum mechanical calculation for dielectric function

$$D(\vec{r},t) = \int^{t} dt' \varepsilon(t-t') E(\vec{r},t')$$

Constitutive relation connects two descriptions

Intense and ultrashort laser pulse prohibits separation
but requires combination of two descriptions (EM and QM).

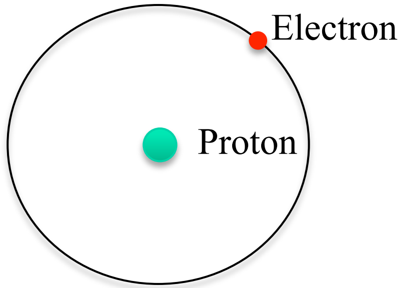
Intense and Ultrashort Laser Pulse

Atomic unit : $m_e = e = \hbar = 1$

Intense electric field

1 a.u. of electric field

= field felt by electron
in classical hydrogen model



Ultrafast motion

1(2π) a.u. of time

= Period of electron motion
in classical hydrogen model

Extremely nonlinear electron dynamics

Attosecond science:

Intense laser pulse on solids

Laser intensity

$$1\,\text{a.u.} = 3.51 \times 10^{16}\ \text{W/cm}^2$$

Solar constant
$0.1366\ \text{W/cm}^2$

$10^{13} - 10^{15}\ \text{W/cm}^2$

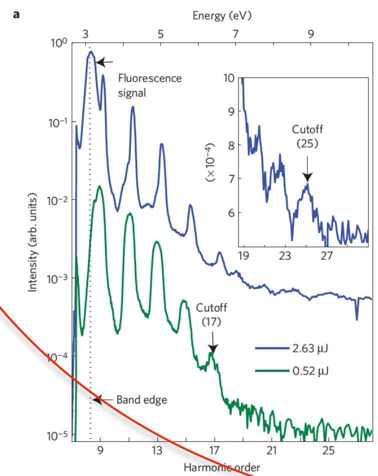
Strongest laser pulse
$10^{23}\ \text{W/cm}^2$

Perturbative
linear and nonlinear
responses

$$\varepsilon(\omega)$$

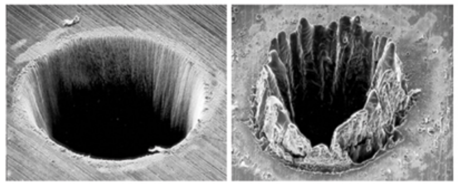
$$\chi^{(2)}(\omega_1,\omega_2),\ \chi^{(3)}(\omega_1,\omega_2,\omega_3)$$

Extreme
Nonlinear response



High Harmonics
Generation

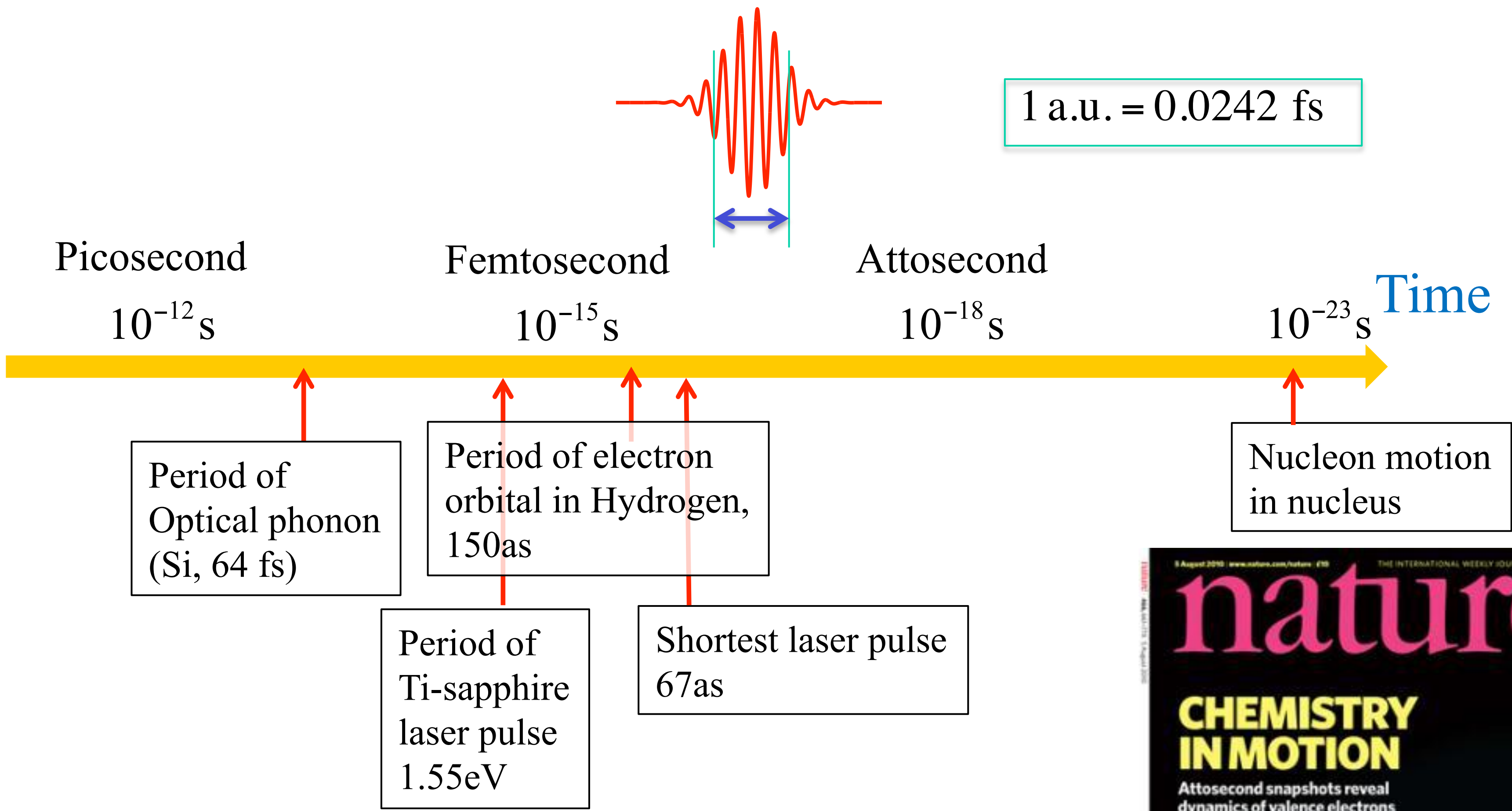
Laser Damage

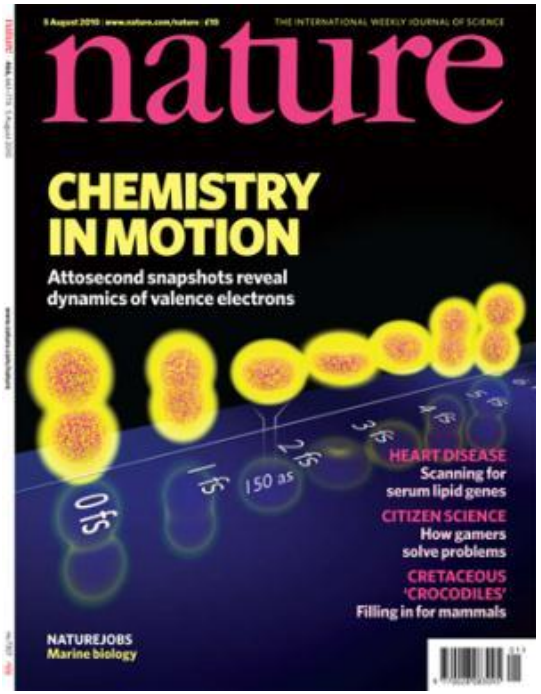


Nonthermal processing

Vacuum
breakdown
$10^{28}\ \text{W/cm}^2$

Ultrashort pulse : Snap shot of electron motion





Real-time observation of valence electron motion
E. Goulielmakis et.al, Nature 466, 739 (2010).

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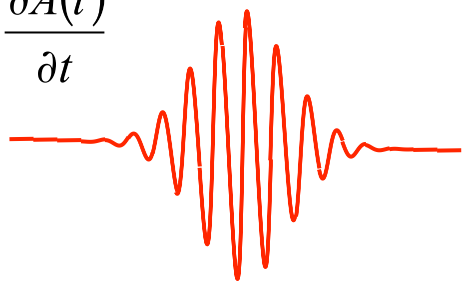
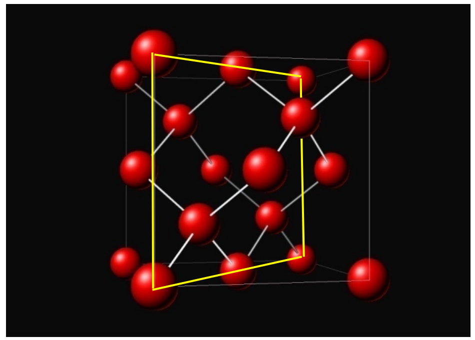
Real-time TDDFT for electron dynamics in a unit cell of crystalline solid under spatially uniform, time-dependent electric field

$$\vec{E}(t) = -\frac{1}{c}\frac{\partial \vec{A}(t)}{\partial t}$$





Time-dependent Kohn-Sham equation for Bloch orbitals

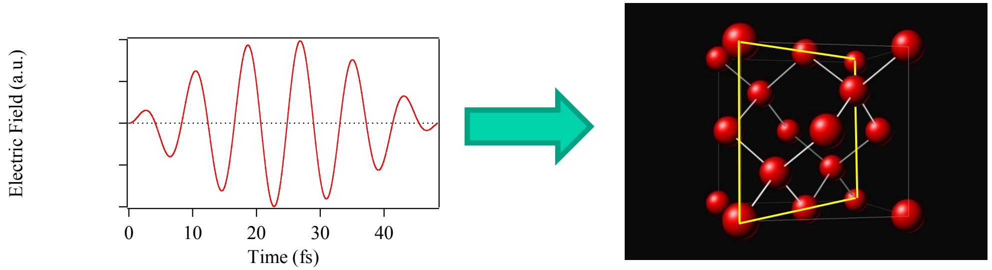
$$i\hbar \frac{\partial}{\partial t} u_{n\vec{k}}(\vec{r},t) = \left[\frac{1}{2m}\left(\vec{p} + \vec{k} + \frac{e}{c}\vec{A}(t) \right)^2 + \int d\vec{r}\,' \frac{e^2}{|\vec{r} - \vec{r}\,'|} n(\vec{r}\,',t) + \mu_{xc}\left[n(\vec{r},t) \right] \right] u_{n\vec{k}}(\vec{r},t)$$

$$n(\vec{r},t) = \sum_{nk} \left| u_{n\vec{k}}(\vec{r},t) \right|^2$$

G.F. Bertsch, J-I. Iwata, A. Rubio, and K. Yabana, Phys. Rev. B 62, 7998 (2000) 43.

Atoms are fixed at equilibrium positions

Example: crystalline silicon under intense laser pulse



$$i\hbar\frac{\partial}{\partial t}u_{n\vec{k}}(\vec{r},t)=\left[\frac{1}{2m}\left(\vec{p}+\vec{k}+\frac{e}{c}\vec{A}(t)\right)^2+\int d\vec{r}\,'\frac{e^2}{|\vec{r}-\vec{r}\,'|}n(\vec{r}\,',t)+\mu_{xc}\left[n(\vec{r},t)\right]\right]u_{n\vec{k}}(\vec{r},t)$$

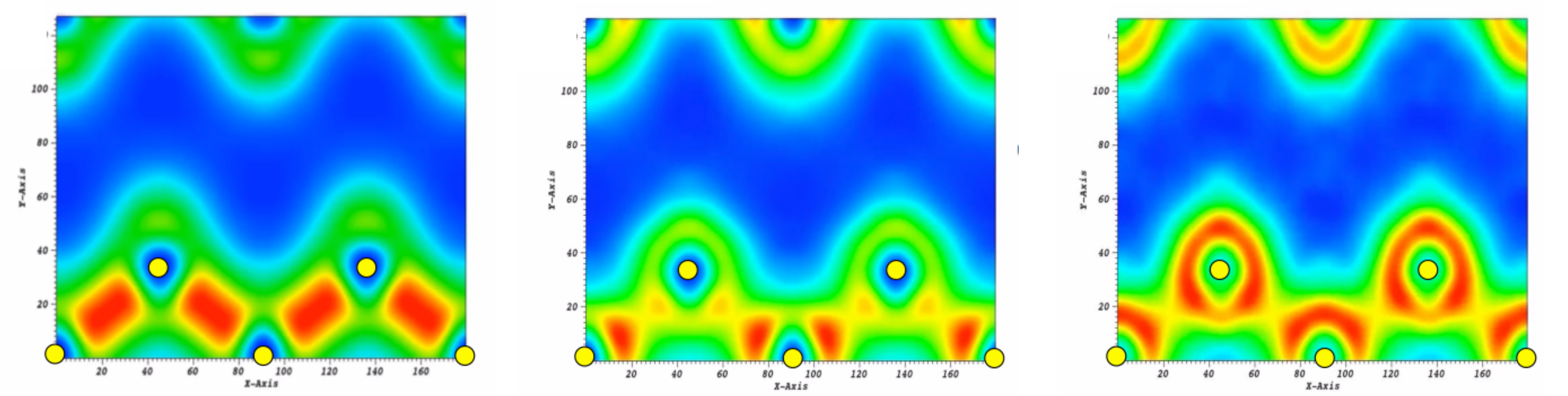
(direct bandgap 2.4 eV in LDA)

$E = 27.5$ V/nm

$\hbar\omega = 1.55$ eV

T_{FWHM} =7 fs

Electron density



Physical quantities from real-time TDDFT calculation

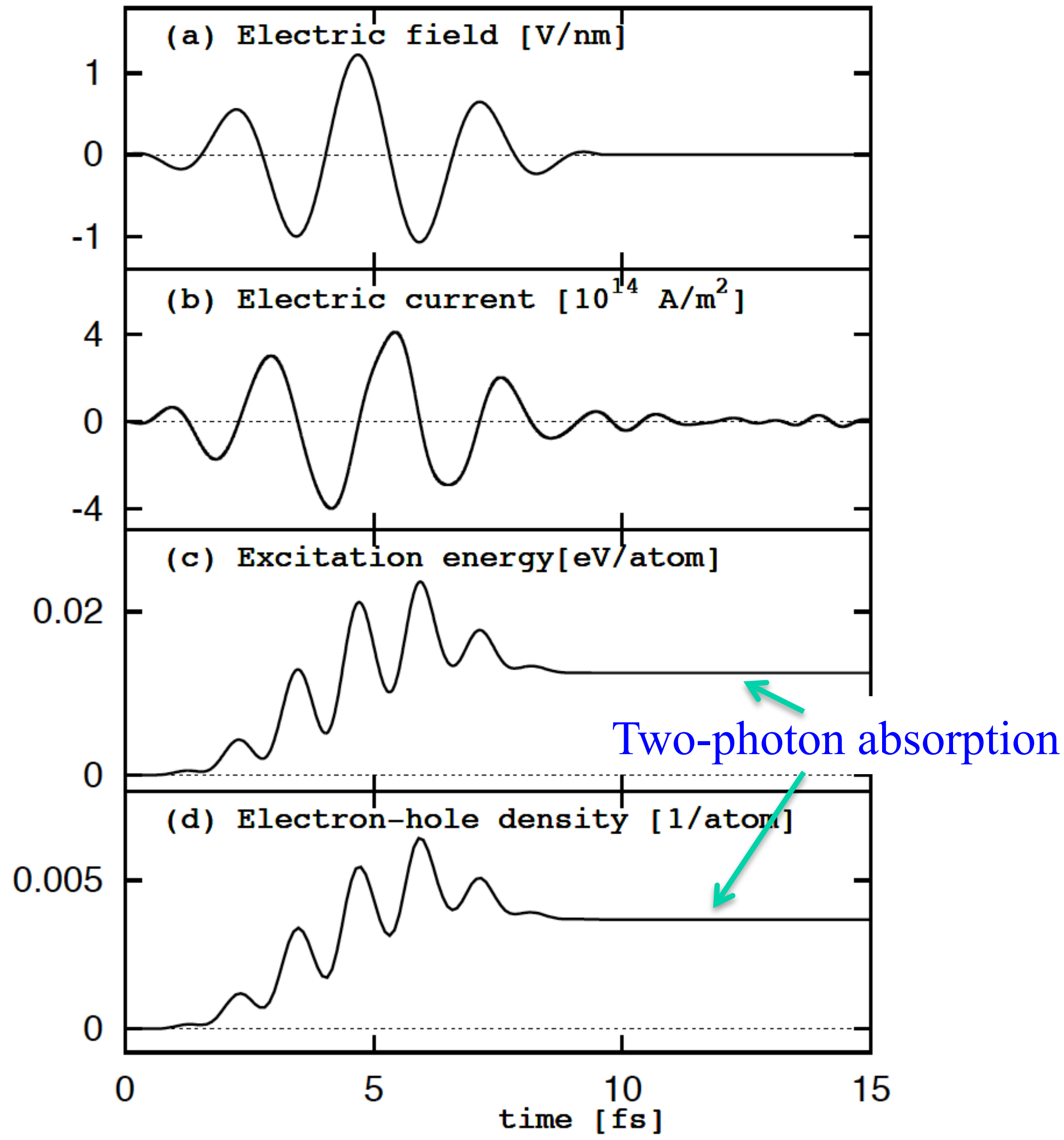
Silicon

(direct bandgap 2.4 eV in LDA)

$E = 1.23$ V/nm

$\hbar\omega = 1.55$ eV

T_{FWHM} =7 fs



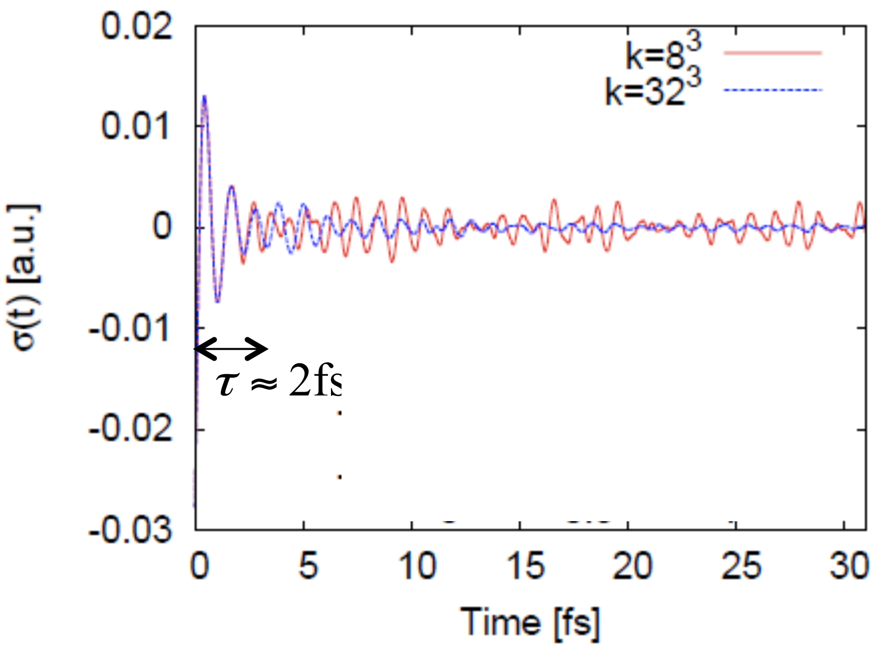
Linear response: Real−time TDDFT for dielectric function (Si, LDA)

Impulsive electric field at t=0

$$E(t) = k\delta(t), \quad A(t) \propto \theta(t)$$

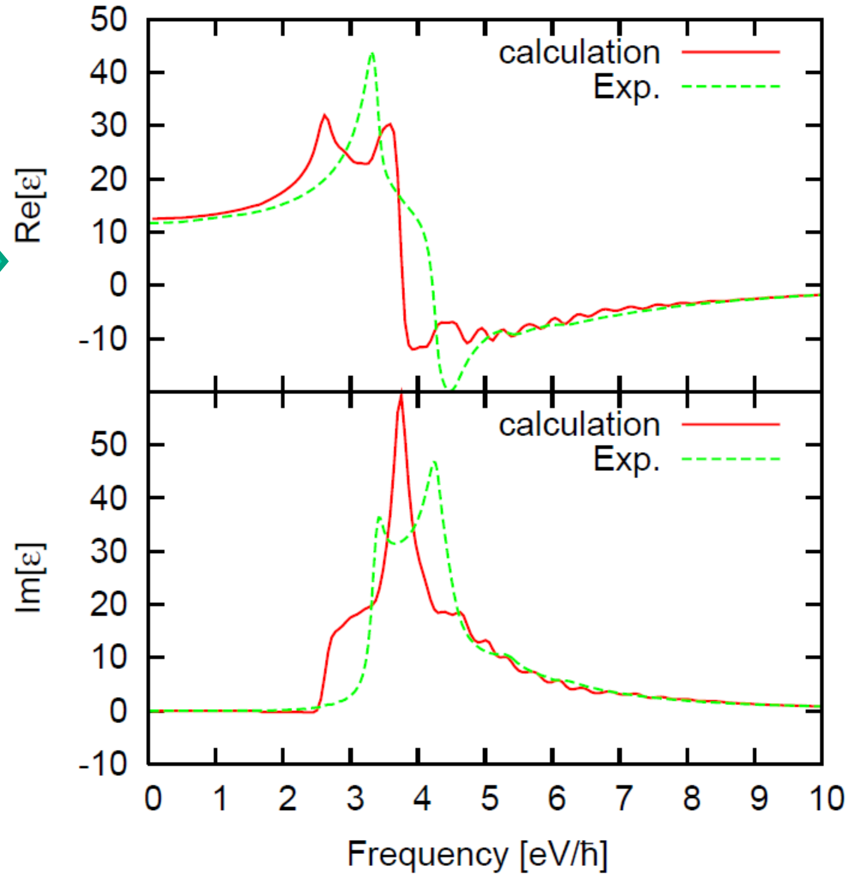
Induced current = conductivity in time domain

$$J(t) = k\sigma(t)$$



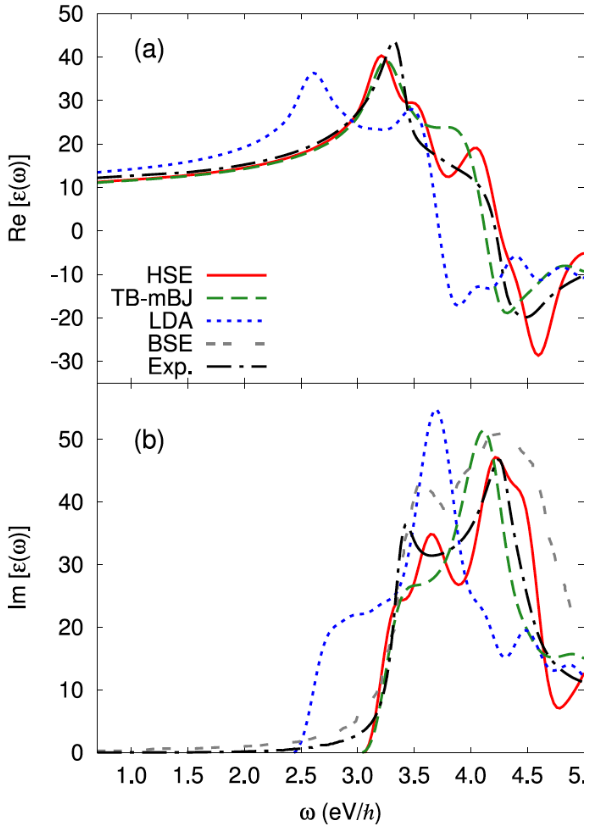
Time-frequency Fourier transformation

$$\sigma(\omega) = \frac{1}{k}\int dt\, e^{i\omega t} J(t) \qquad \varepsilon(\omega) = 1 + \frac{4\pi i \sigma(\omega)}{\omega}$$

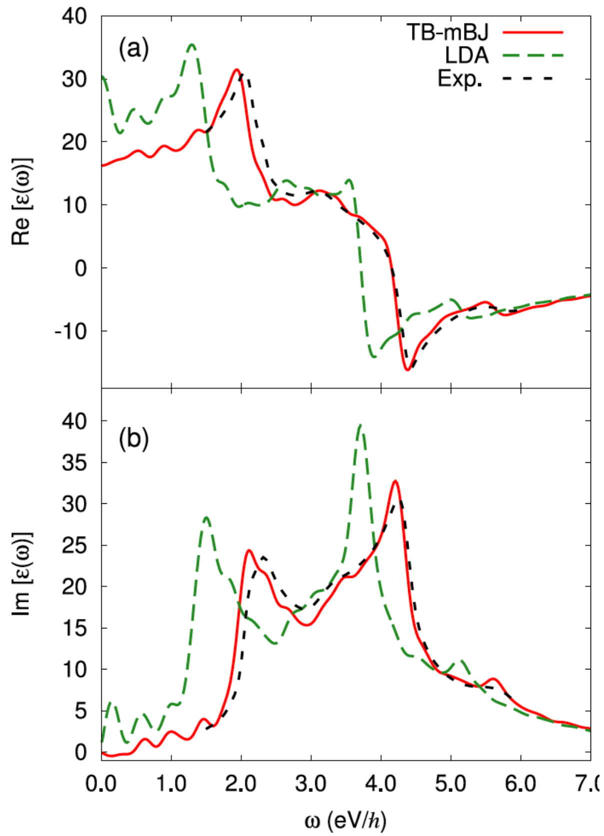


Quality of the exchange-correlation potential may be assessed by dielectric function

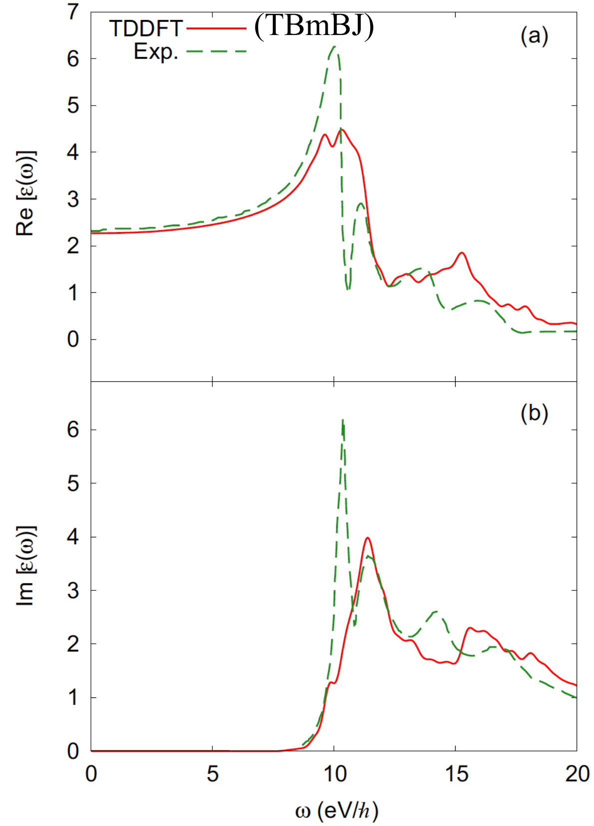
Meta-GGA potential of Tran and Blaha (TBmBJ)
reproduces band gap of insulators. PRL102, 226401 (2009)



Silicon



Germanium



SiO_2 (α-quartz)

S.A. Sato et.al, JCP143, 224116 (2015)

S.A. Sato et.al, PRB92, 205413 (2015)

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Electron dynamics calculation as Numerical constitutive relation

$$\vec{P}\left(\vec{R},t\right)=\vec{P}\left[\vec{E}\left(\vec{R},t\right)\right]$$

For a given electric field $E(t)=-\frac{1}{c}\frac{dA(t)}{dt}$

Electron dynamics calculation in a unit cell of solid

$$i\hbar\frac{\partial}{\partial t}u_{n\vec{k}}(\vec{r},t)=\left[\frac{1}{2m}\left(\vec{p}+\vec{k}-\frac{e}{c}\vec{A}(t)\right)^2+\int d\vec{r}'\frac{e^2}{|\vec{r}-\vec{r}'|}n(\vec{r}',t)+\mu_{xc}[n(\vec{r},t)]\right]u_{n\vec{k}}(\vec{r},t)$$

$$n(\vec{r},t)=\sum_{nk}\left|u_{n\vec{k}}(\vec{r},t)\right|^2$$

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

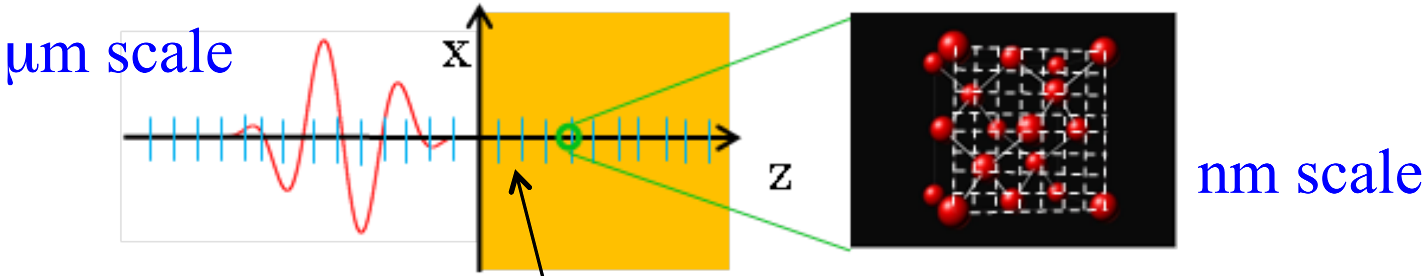
provides electric current and polarization

$$\vec{J}(t)=\frac{1}{\Omega}\int_\Omega d\vec{r}\,\vec{j}(\vec{r},t),\qquad \vec{P}(t)=-\int^t dt'\vec{J}(t')$$

Maxwell + TDDFT multiscale approach

Macroscopic grid
for Maxwell equation

Microscopic grid
for TDDFT electron dynamics



$$\frac{1}{c^2}\frac{\partial^2}{\partial t^2}A_Z(t)-\frac{\partial^2}{\partial Z^2}A_Z(t)=\frac{4\pi}{c}J_Z(t)$$

$$i\hbar\frac{\partial}{\partial t}u_{Z,n\vec{k}}(\vec{r},t)=h\left[\vec{A}_Z(t),n_Z(\vec{r},t)\right]u_{Z,n\vec{k}}(\vec{r},t)$$

$$\vec{J}_Z(t)=\frac{1}{V}\int_V \vec{j}_Z(\vec{r},t)$$

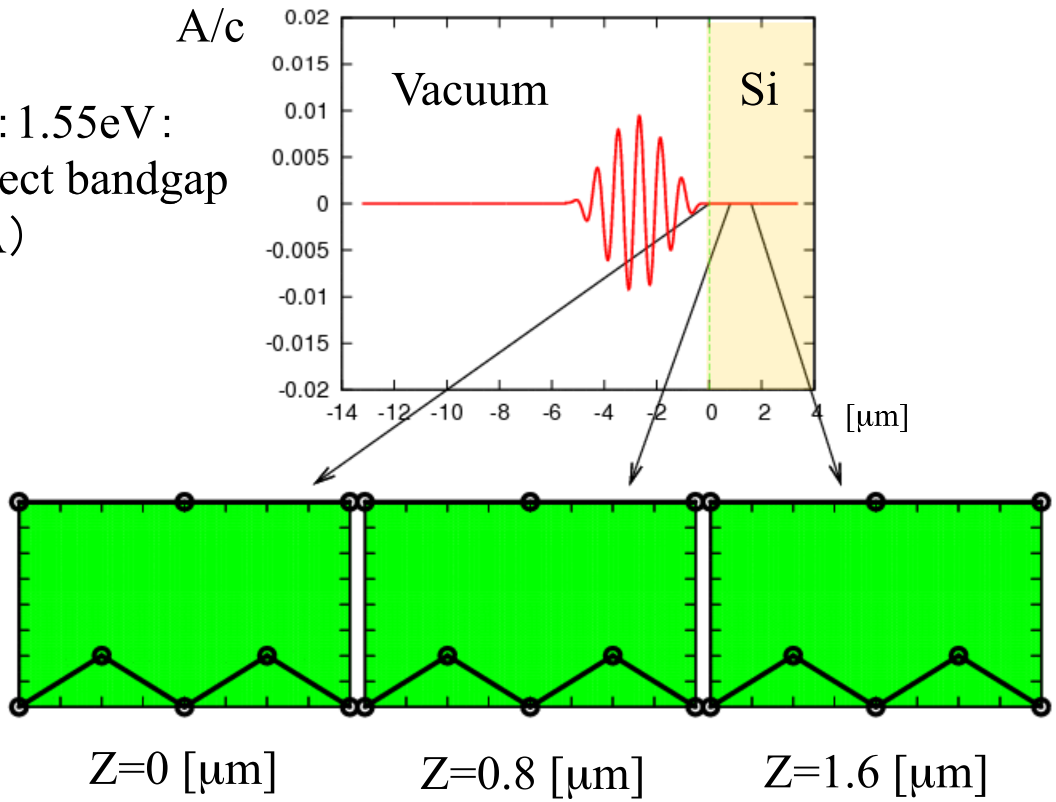
At each macroscopic grid point,
we solve real-time electron dynamics in parallel

K. Yabana et.al, Phys. Rev. B85, 045134 (2012).

Propagation of weak pulse (transparent)
(Linear response regime, ordinary linear optics applies)

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

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



Propagation of weak pulse (transparent)
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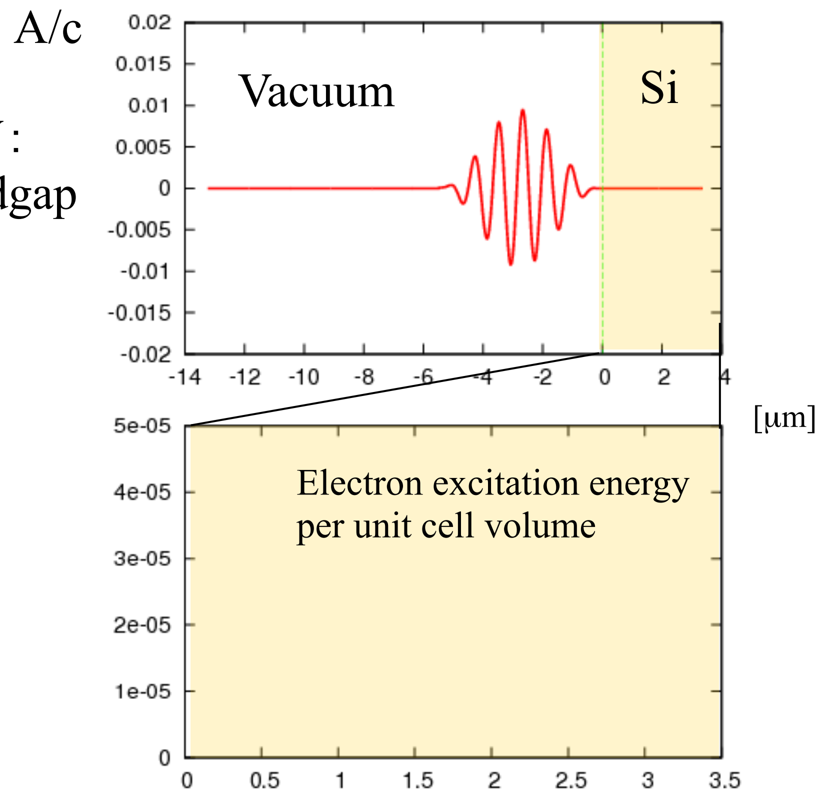
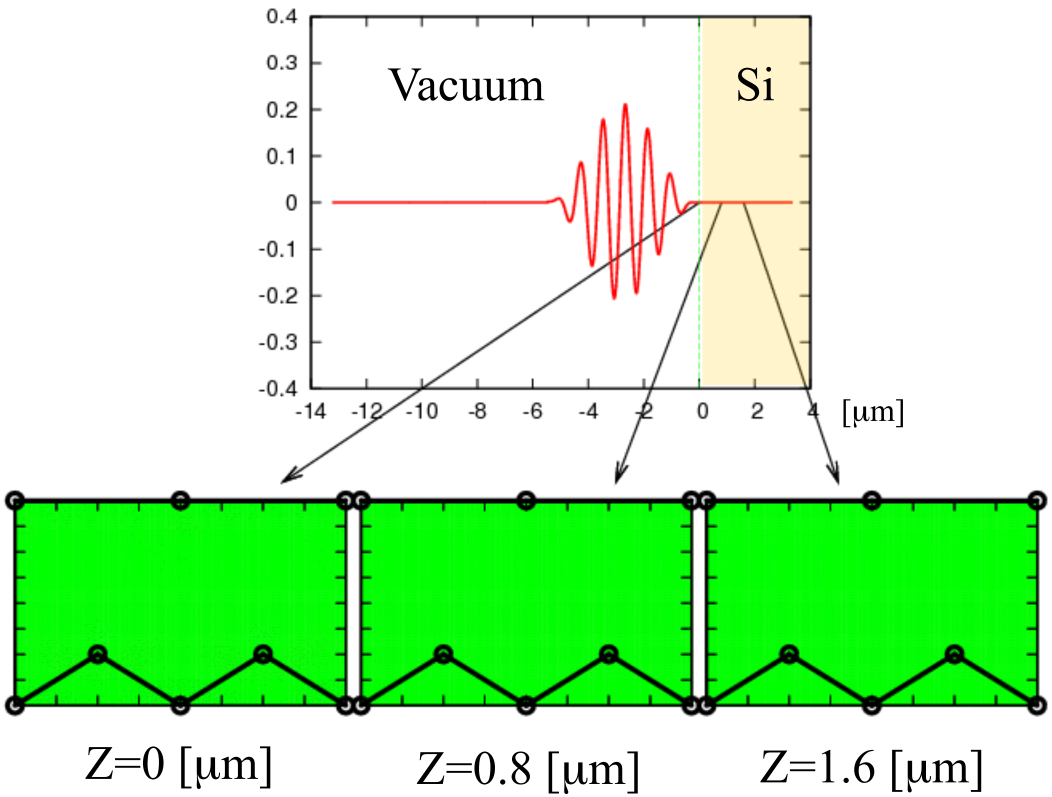


More intense laser pulse (absorptive)

Maxwell and TDKS equations no more separate.

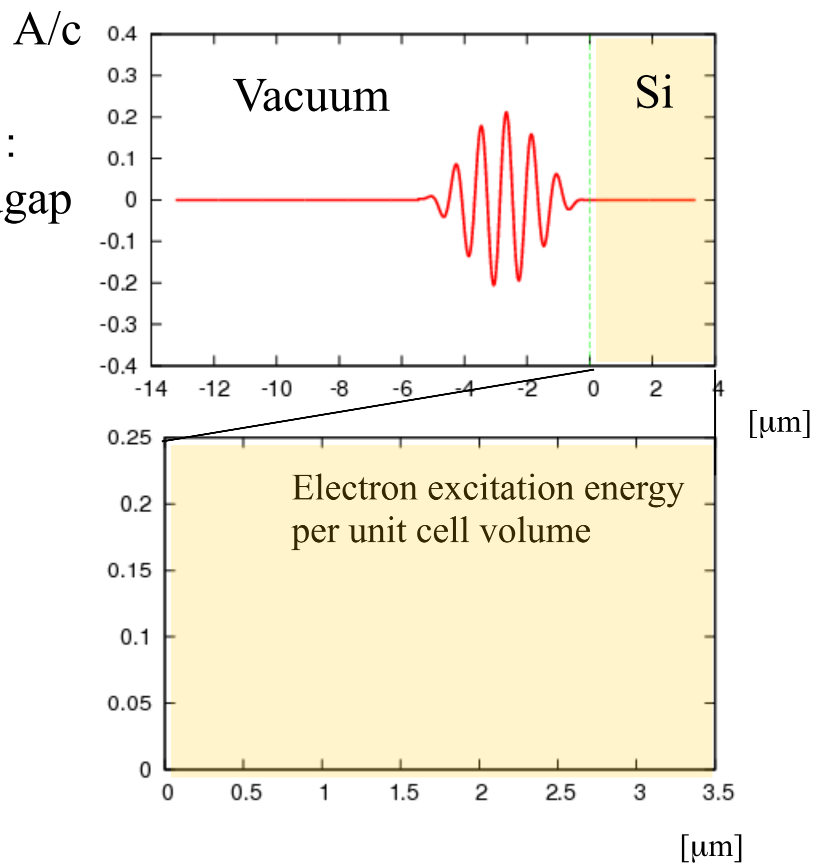
$I = 5 \times 10^{12} W/cm^2$



More intense pulse (absorptive)
(2-photon absorption dominates)

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

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



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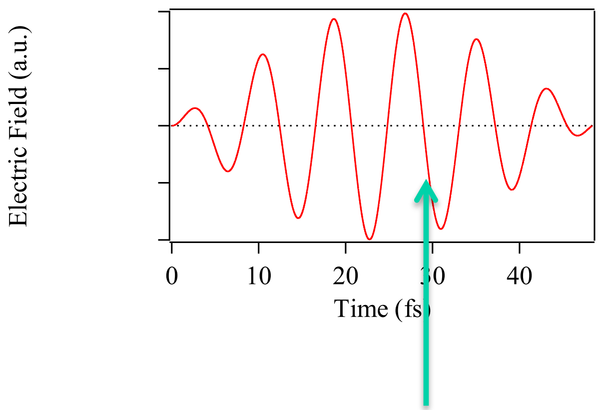
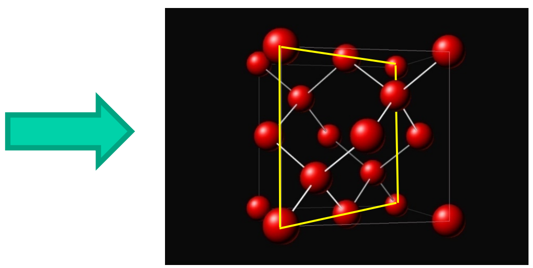
Application 1:
Change of dielectric property of solid under intense field



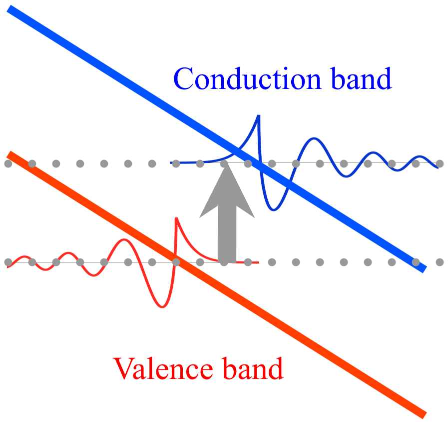


At each time T, how dielectric function changes?

$$\varepsilon(\omega, T)$$

Under a static electric field: Franz-Keldysh effect

W. Franz, Z. Naturforsch. 13, 484 (1958)
L.V. Keldysh, Sov. Phys. JETP 34, 788 (1958)



Analytic formula for a two-band model

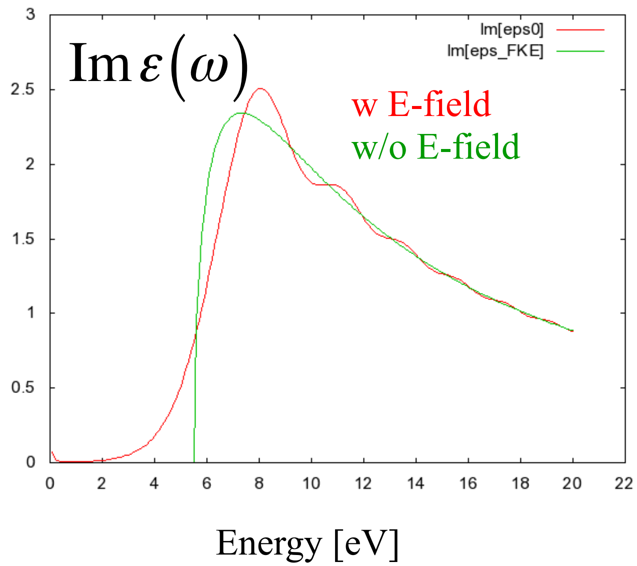
$$\mathrm{Im}\,\varepsilon(\omega, F) \propto \frac{\Theta^{1/2}}{\omega^2}\left\{\mathrm{Ai}'^2(\xi) - \xi \mathrm{Ai}^2(\xi)\right\}$$

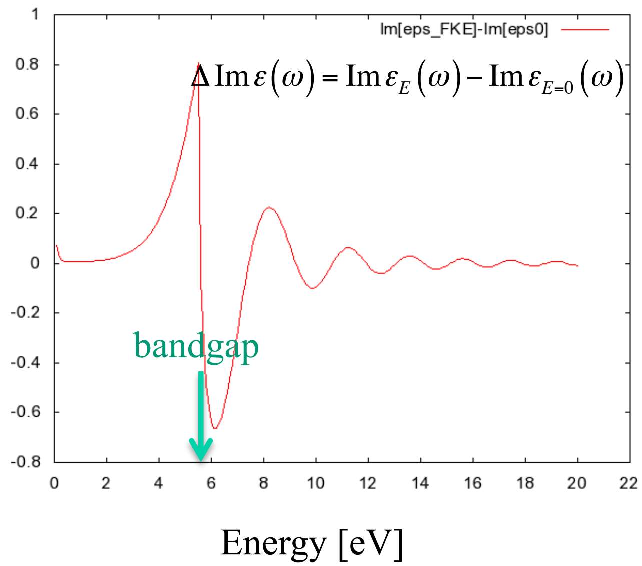
$$\xi = \frac{E_{gap} - \hbar\omega}{\Theta}$$

Airy function
→Quantum tunneling

$$\Theta = \left(\frac{e^2 F^2 \hbar^2}{2\mu}\right)^{1/3}$$

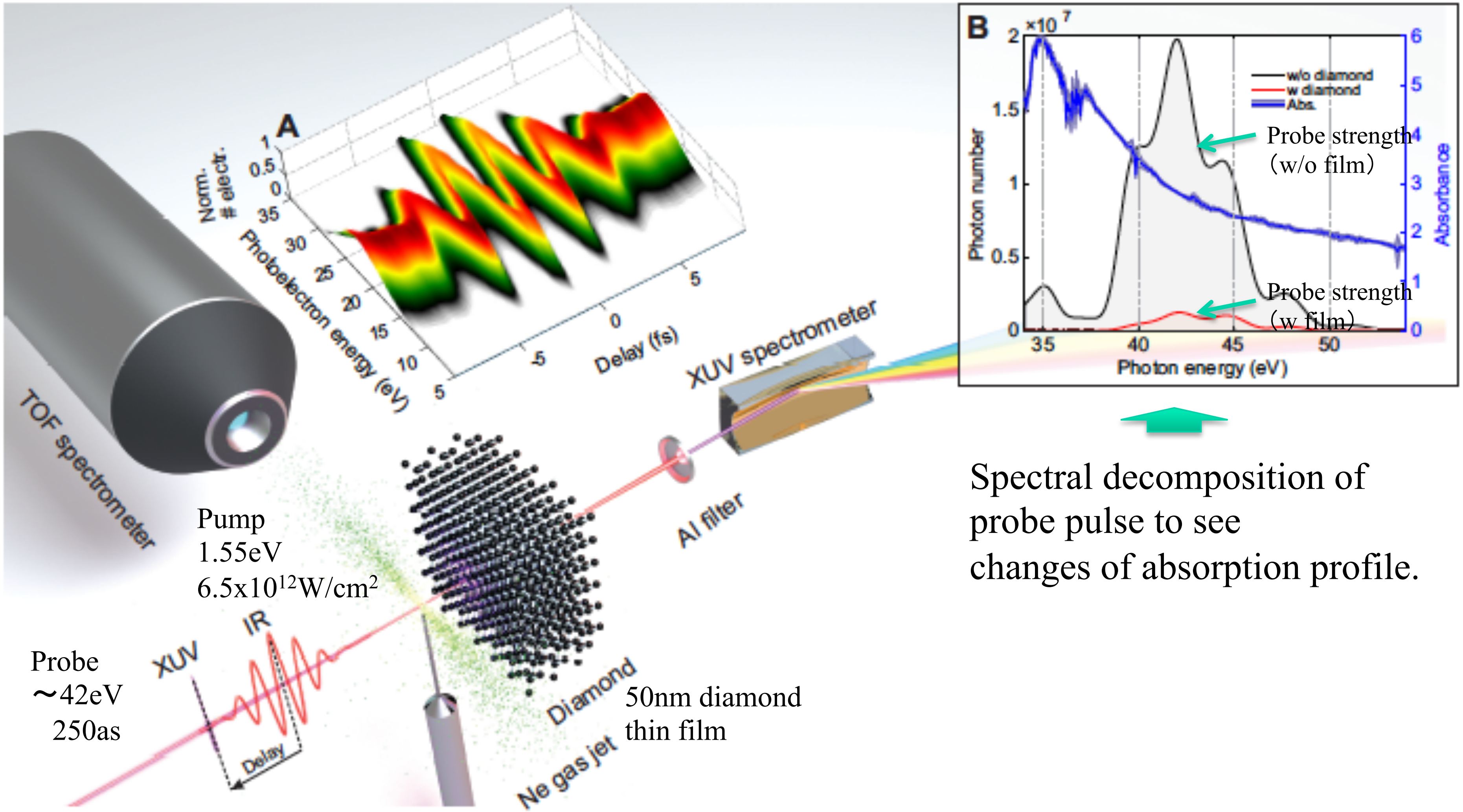
Electrooptical energy





Experiment (ETH group) and Calculation (Maxwell + TDDFT)
Pump-probe method for diamond thin film

- Irradiate IR strong pump-pulse (1.55 eV) and XUV weak probe pulse (42eV) simultaneously.
- Examine change of probe abosorption as a function of time-delay.



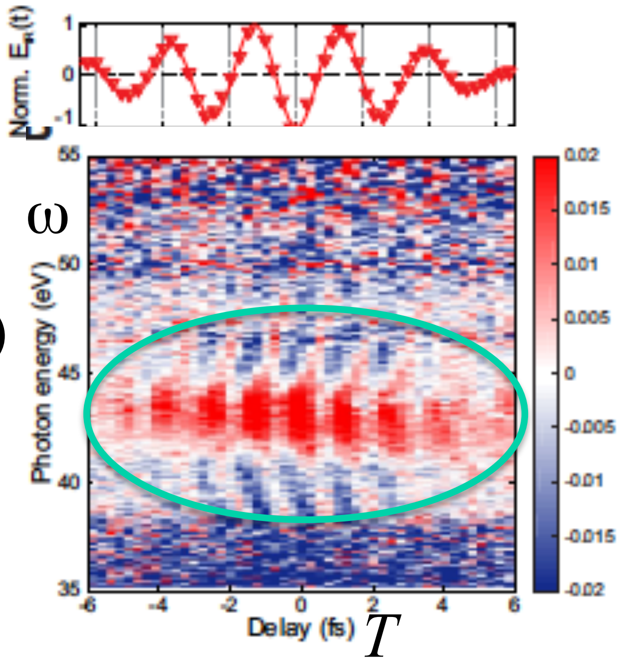
Spectral decomposition of
probe pulse to see
changes of absorption profile.

Results

M. Lucchini et.al, Science 353, 916-919 (2016)

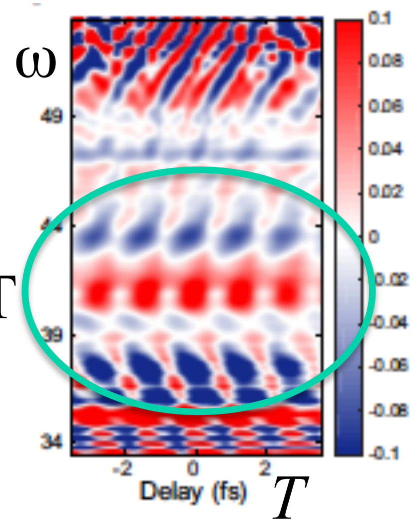
Pump electric field

$\Delta abs(\omega, T)$
Exp.



- Absorption increase at 42eV when pump field exists.
- "Fish-bone" structure (time delay in response change)

$\Delta abs(\omega, T)$
Maxwell-TDDFT



- Calculation reproduce measured features.
- Supports interpretation by dynamical Franz-Keldysh effect

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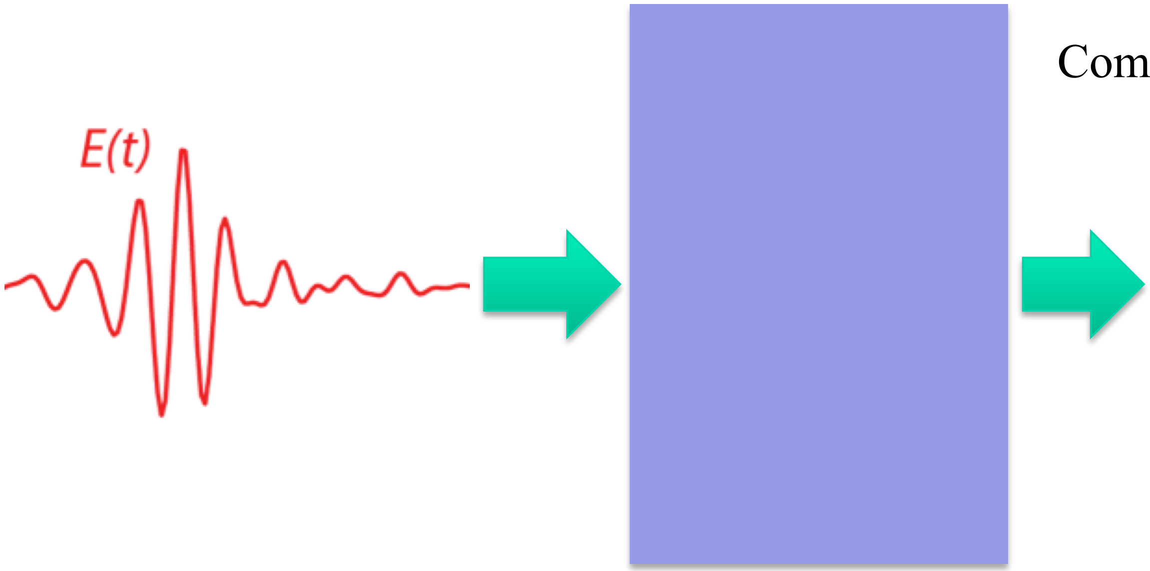
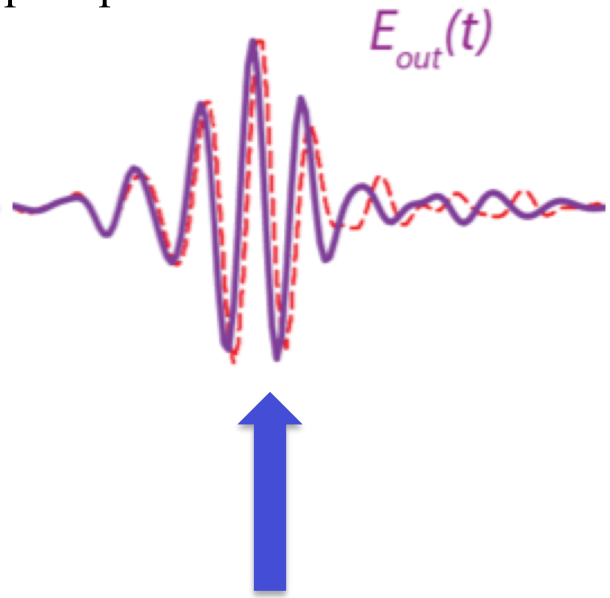
Application 2: laser pulse propagation through SiO_2 thin film

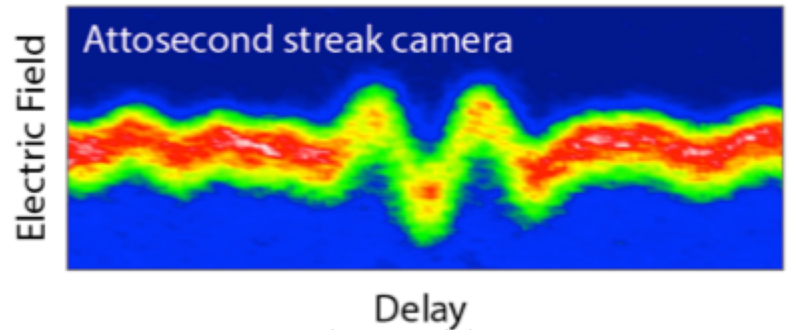
Energy deposition from laser pulse to dielectrics



Compare pulses of different intensities



Thin film of SiO_2
10μm



EXP: Attosecond streaking measurements
by Max-Planck Inst. Quantum Optics

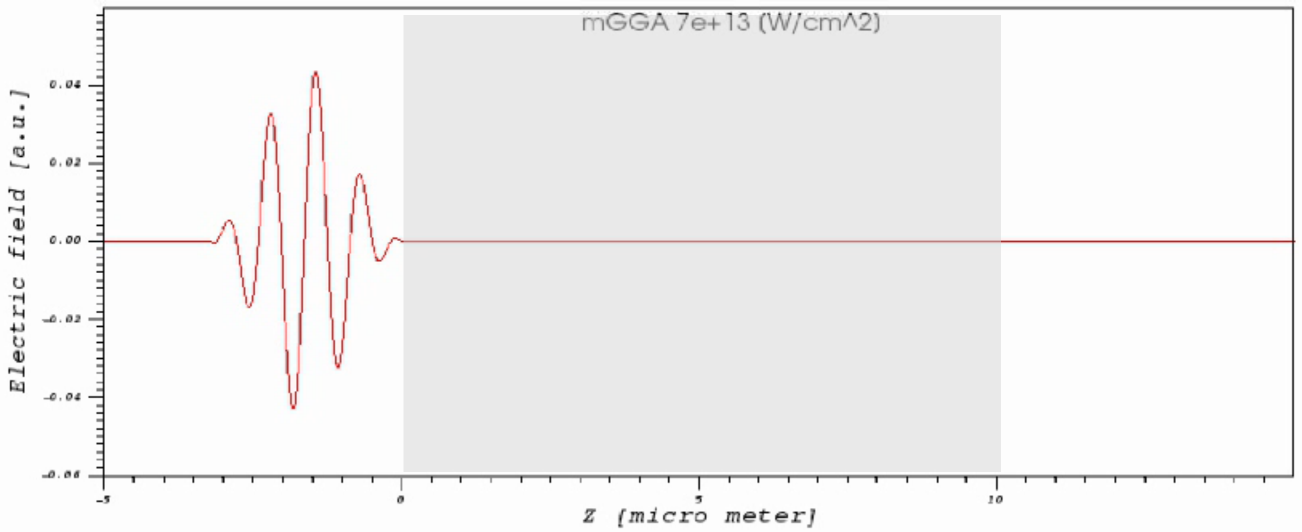
Maxwell + TDDFT multiscale simulation : 10 μm SiO_2

$\hbar\omega = 1.55\text{eV}$

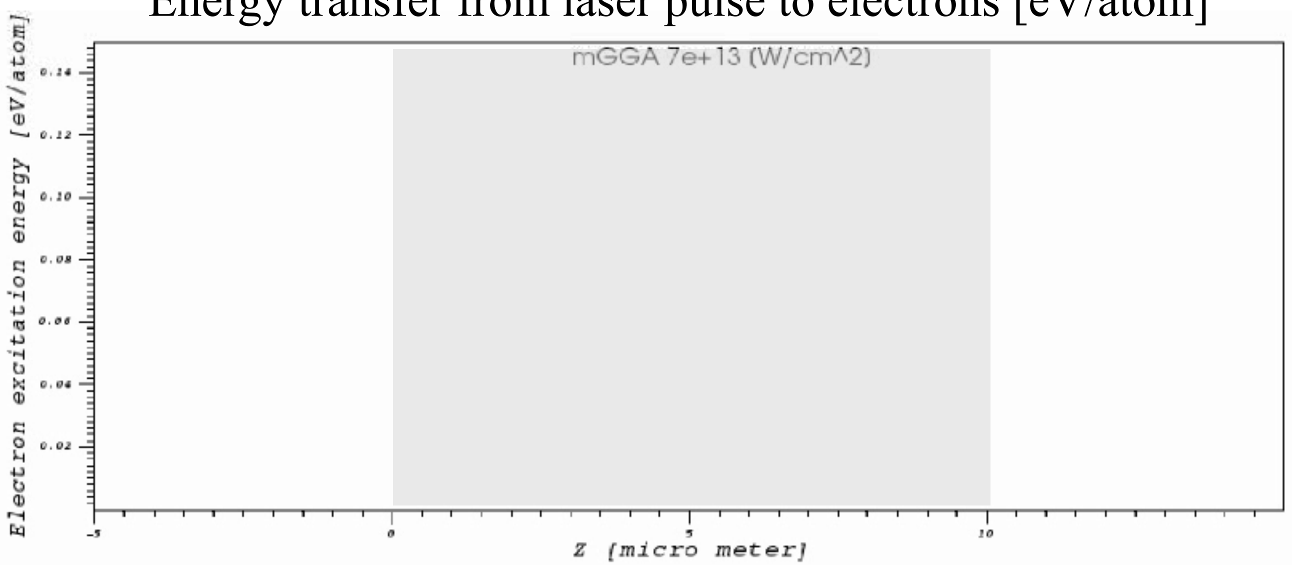
λ=800nm

$I=7\times10^{13}W/cm^2$

Laser electric field, red (strong), blue (weak)

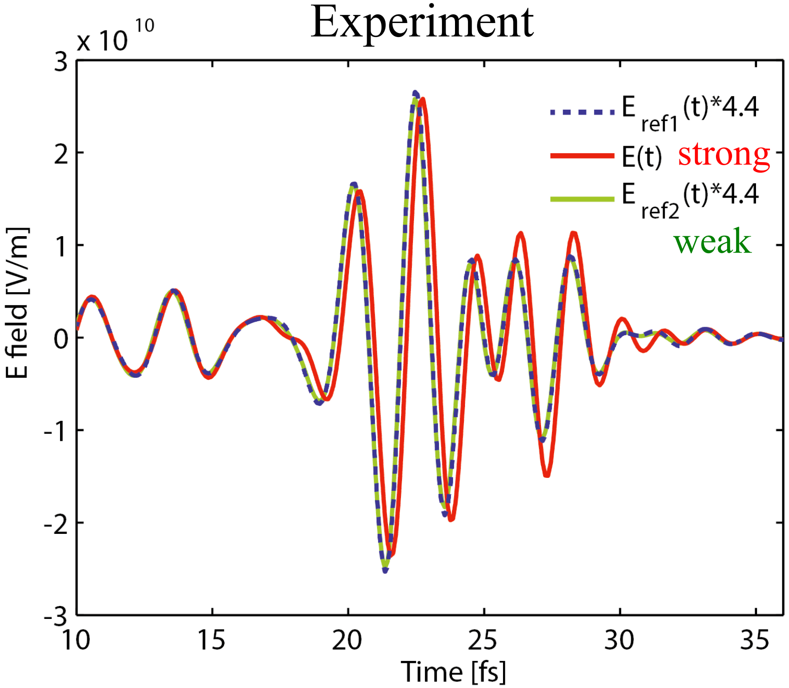


Energy transfer from laser pulse to electrons [eV/atom]

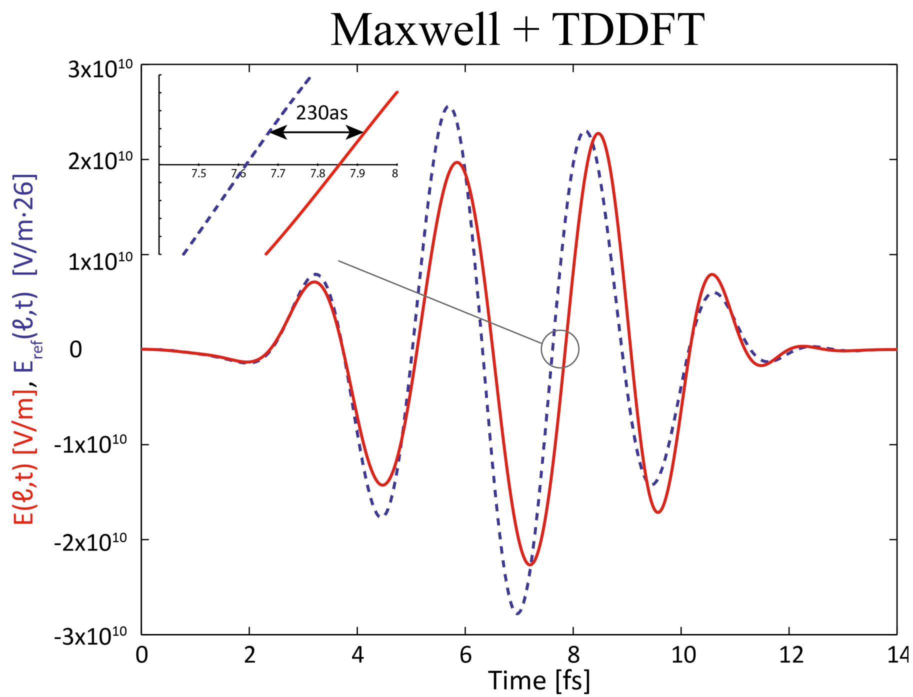


80,000 cores, 20 hours
at K-Computer, Kobe

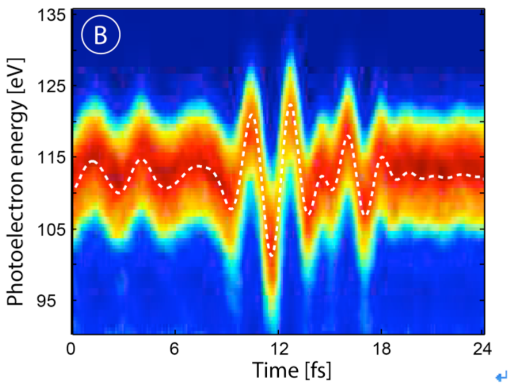
Comparison of pulse shape after passing through SiO_2 thin film



$I = 1.28 \times 10^{14} W/cm^2$

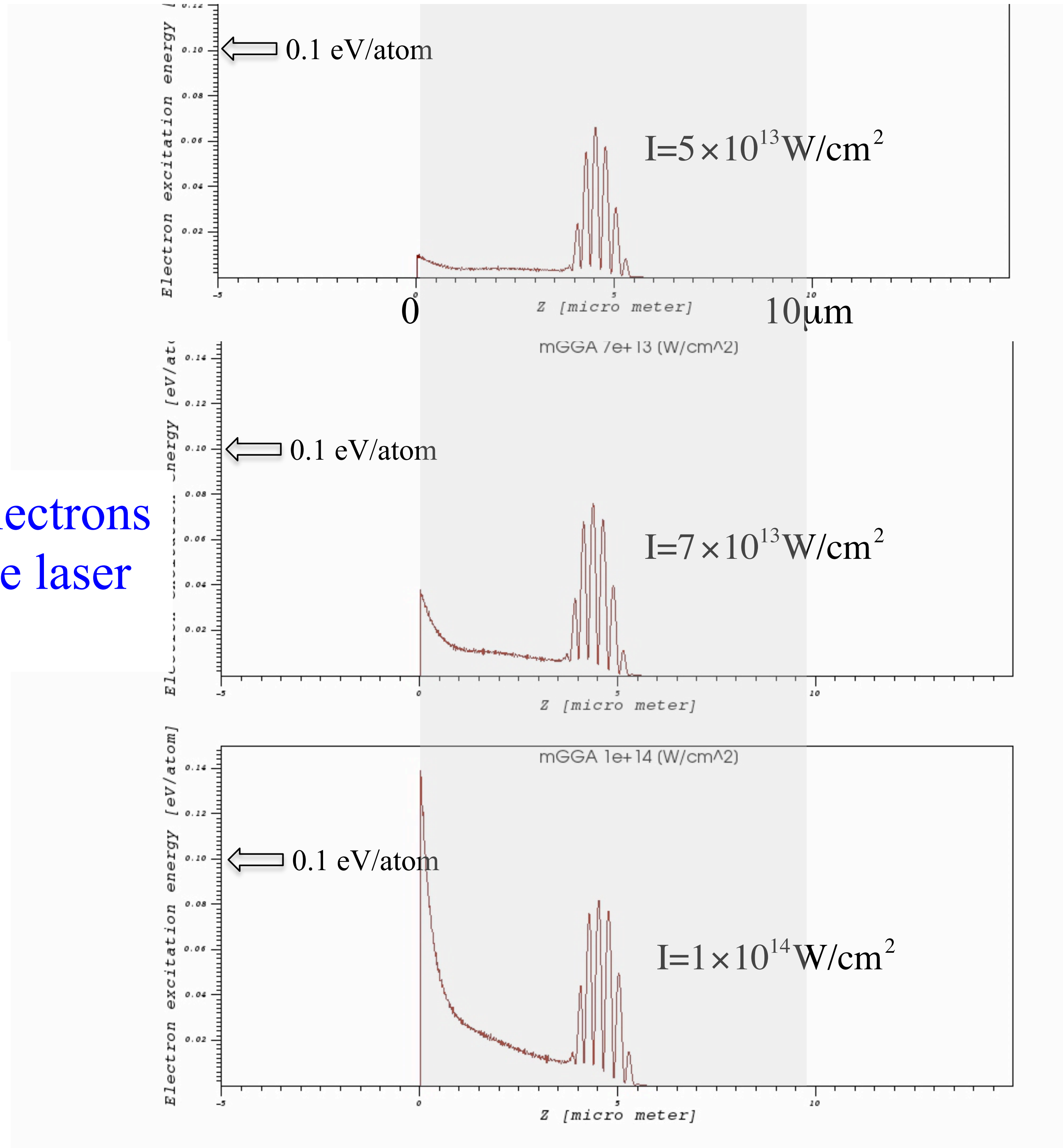


$I = 7 \times 10^{13} W/cm^2$

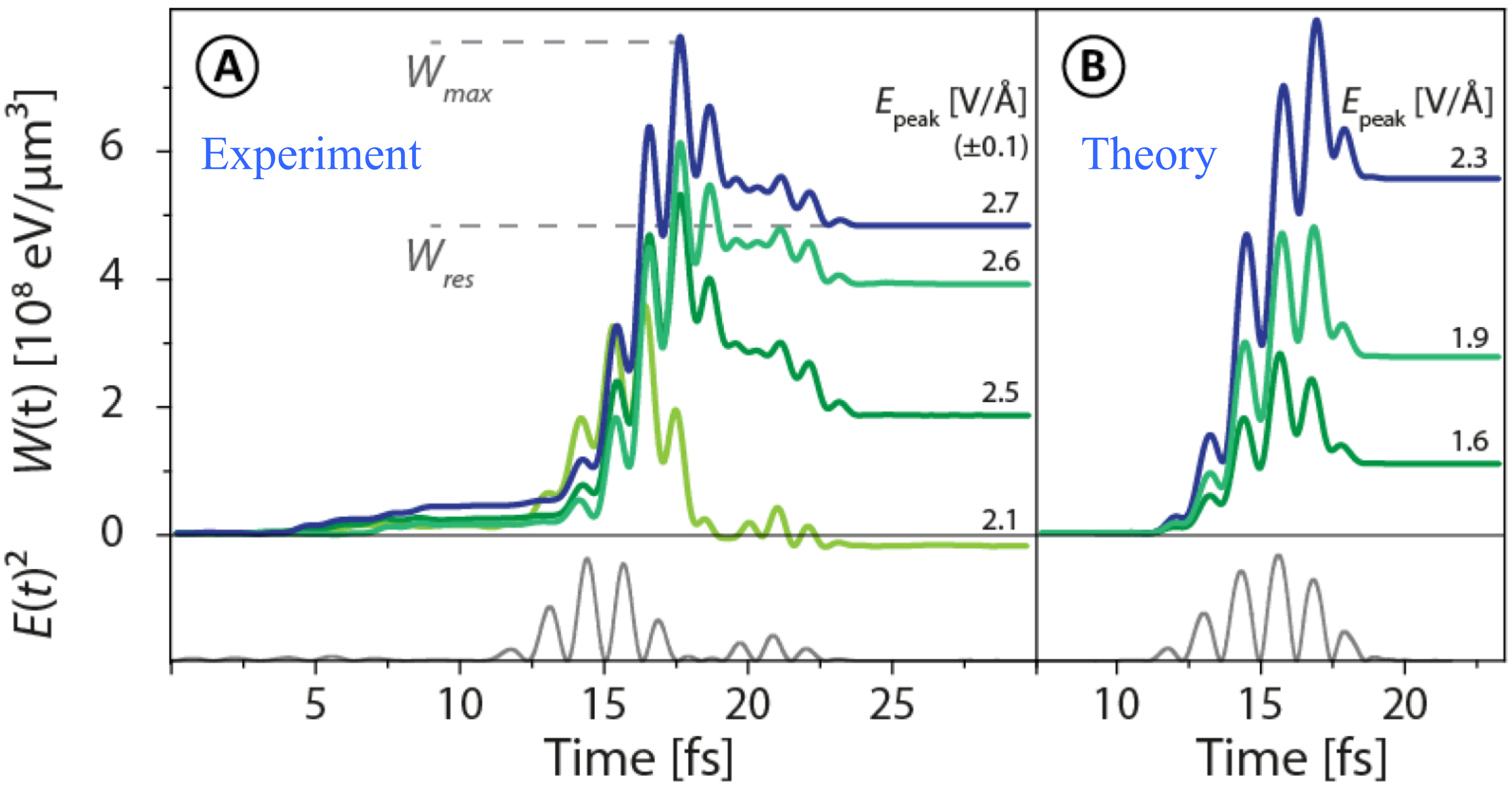


- Although very close to damage threshold, pulse shape is dominated by 3rd order nonlinearity.
- Pulse shape change, phase shift well described by Maxwell + TDDFT calculation.

Energy deposited to electrons increases rapidly as the laser intensity increases.



Energy deposition from laser pulse to SiO_2 at mid point (5μm) can be evaluated experimentally from the transmitted pulse.

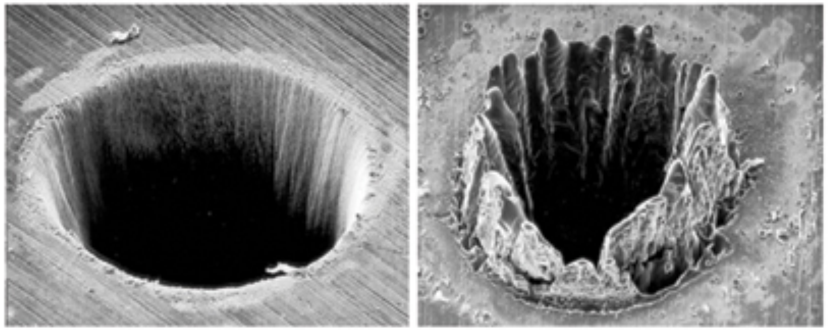


A. Sommer et.al, Nature 534, 86 (2016).
(EXP: Max Planck Institute for Quantum Optics)

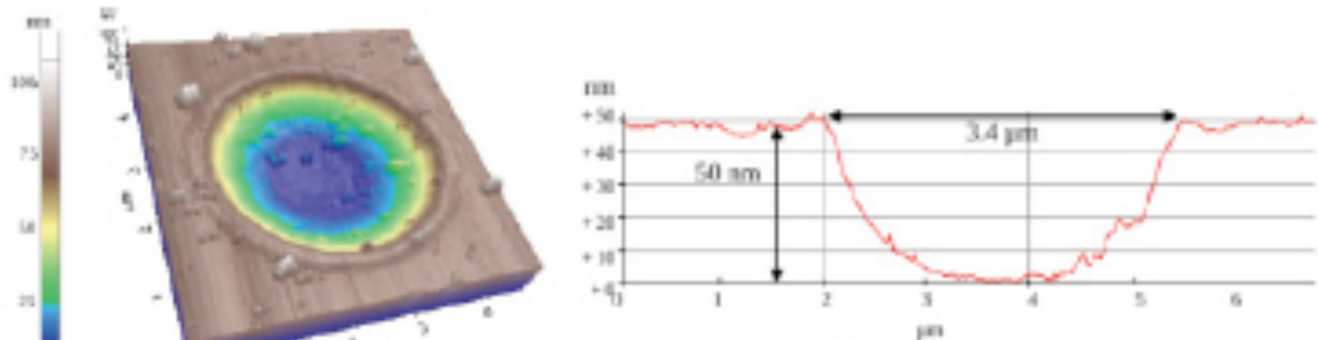
CONTENTS

Application 3: Estimate laser-ablation threshold and depth

Femtosecond laser pulse expected for nonthermal laser processing.



Single shot laser pulse create 'crator'.



1.1 $F_{th,abl}$ (0.5 μJ) - 7 fs

Maxwell + TDDFT multiscale calculation for α-quartz (SiO_2)

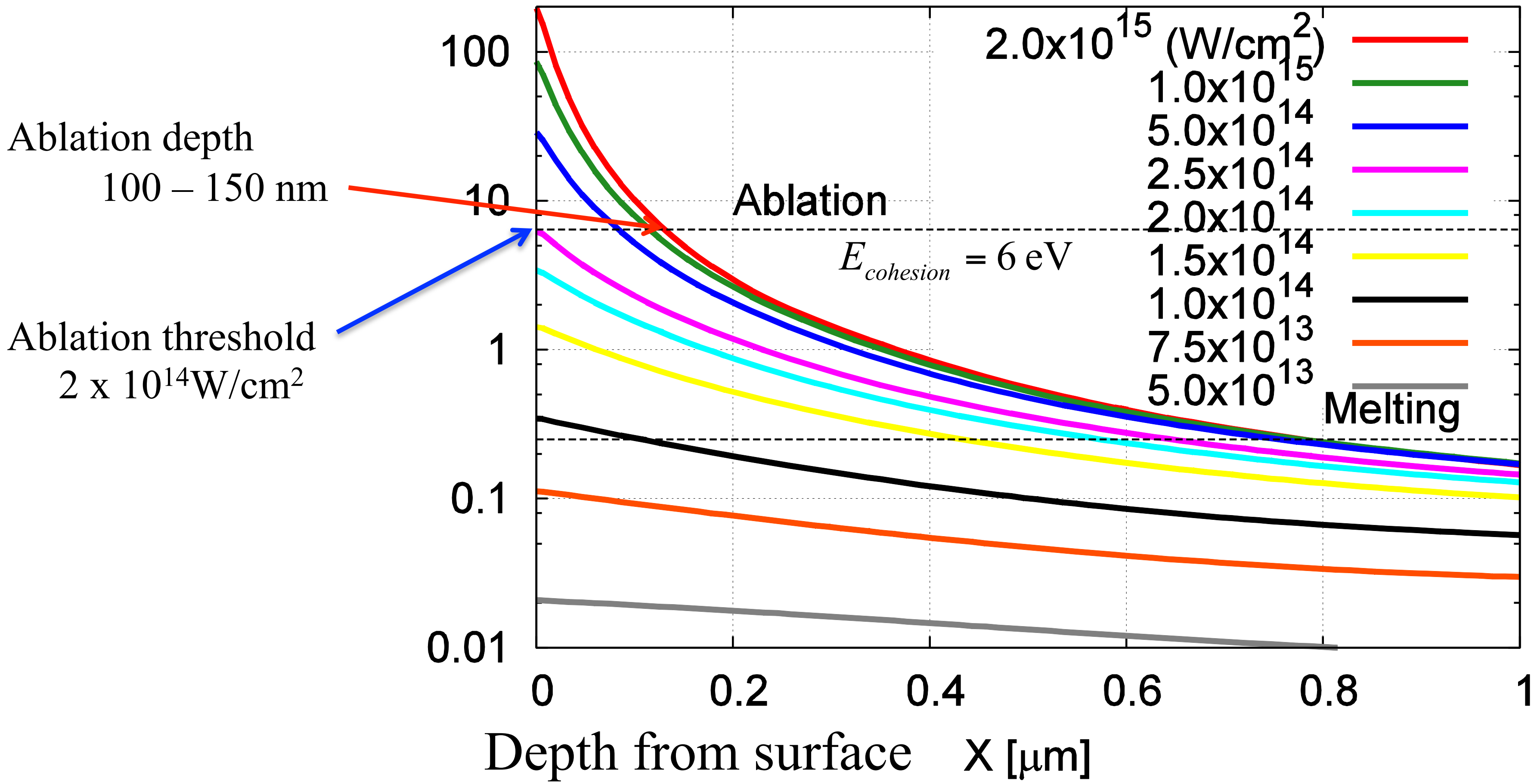
$\hbar\omega = 1.55\text{eV}\ (\lambda = 800\text{nm}),$

$T_{FWHM} = 7\text{fs}$

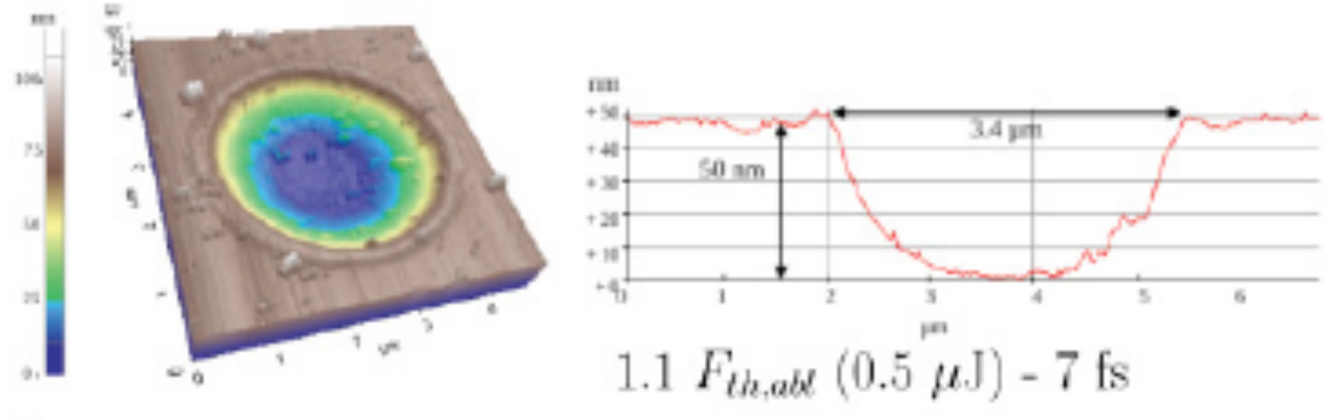
$E_{gap} = 9.0\,\text{eV}$

Energy transfer from pulsed electric field to electrons [eV/atom]

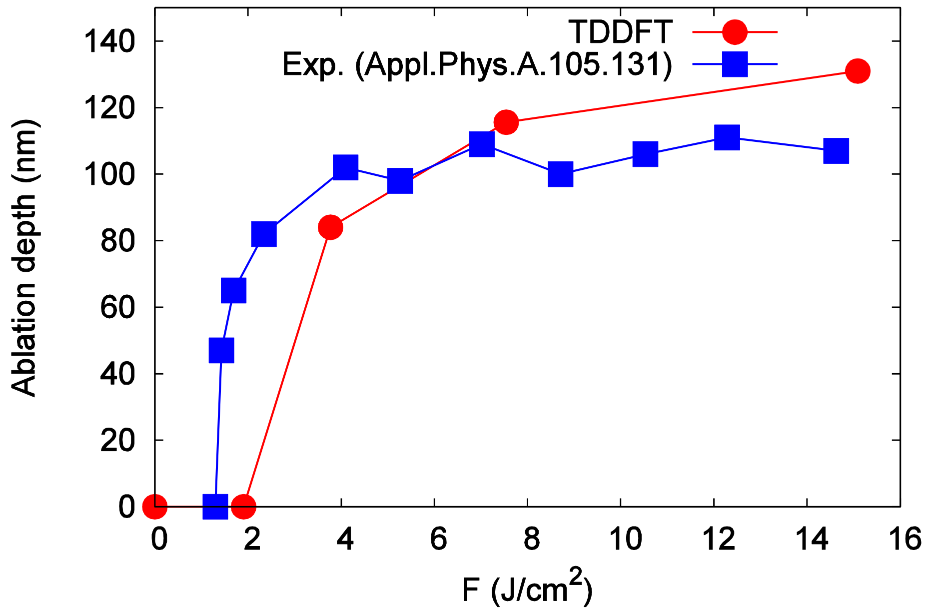
S.A. Sato et.al, Phys Rev. B92, 205413 (2015)



Ablation: threshold and depth, comparison with measurements



Crater formation: measurement, B. Chimier et al, Phys. Rev. B84, 094104 (2011)



S.A. Sato et.al, Phys Rev. B92, 205413 (2015)

Threshold and depth reasonably estimated.

Summary

Real-time TDDFT in crystalline solid

is useful to describe nonlinear and nonperturbative electron dynamics induced by intense and ultrashort laser pulse.

Maxwell + TDDFT multiscale simulation

provides numerical experiment platform for optical science frontiers

- can describe propagation of intense laser pulse in the medium.
- can provide physical quantities which can be compared with cutting-edge optical measurements.
- will open first-principle investigation of nonthermal laser processing
- is computationally challenging requiring cutting-edge supercomputers

Future problems

- collision (e-e, e-phonon), dephasing effects
- improved functional (excitons, …)
- 2D, 3D light pulse propagation (self-focusing, filamentation, light vortex…)