CCS-EPCC Workshop
Dc. 7-8, 2017

Ab-initio density functional simulation for nano-optics

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http://salmon-tddft.jp

We are developing a computational code, SALMON

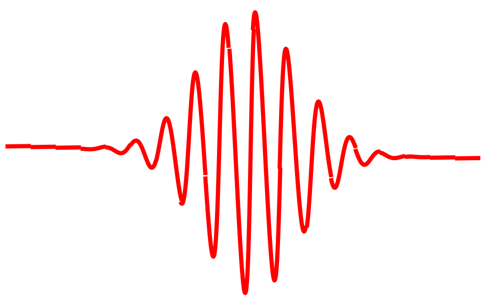
Scalable Ab-initio Light-Matter simulator for Optics and Nanoscience

It is capable of describing light-matter interactions starting with ab-initio electron dynamics calculations.

Two aspects of light-matter simulation

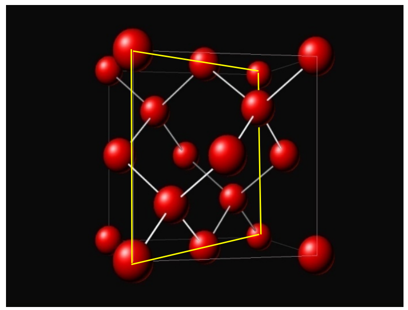
Light propagation usually described using Maxwell's equations
Macroscopic electromagnetism

Light wave of pulsed laser



Electron dynamics is induced by light electric field, described by Schrödinger equation
Quantum mechanics

Atoms, molecules, solids



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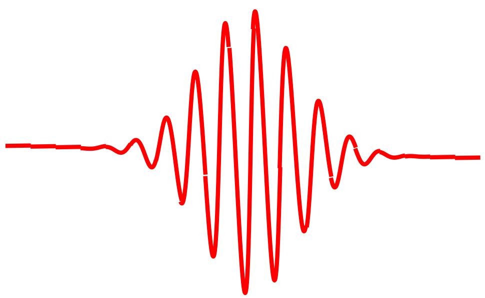

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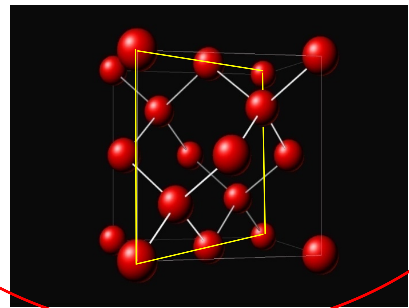
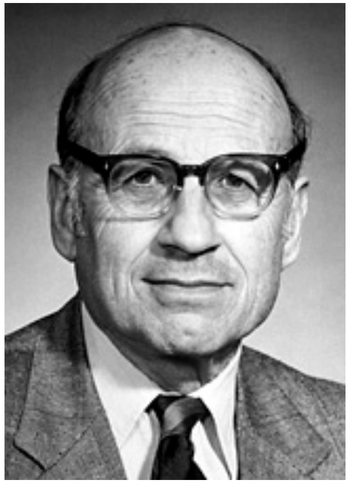


Popular tool in computational materials science:
Density functional theory (DFT)

$$\varepsilon_i \phi_i(r) = \left[\frac{1}{2m} p^2 + V_{ion}(r) + V_H(r) + V_{xc}(r)\right] \phi_i(r)$$



W. Kohn, 1998 Nobel prize in chemistry

Theory for ground state. Not applicable to photoexcitation

Extension to electron dynamics: Time-dependent DFT (TDDFT)

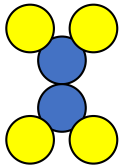
$$i\hbar \frac{\partial}{\partial t} \psi_i(r,t) = \left[\frac{1}{2m} p^2 + V_{ion}(r) + V_H(r,t) + V_{xc}(r,t) + V_{ext}(r,t)\right] \psi_i(r,t)$$

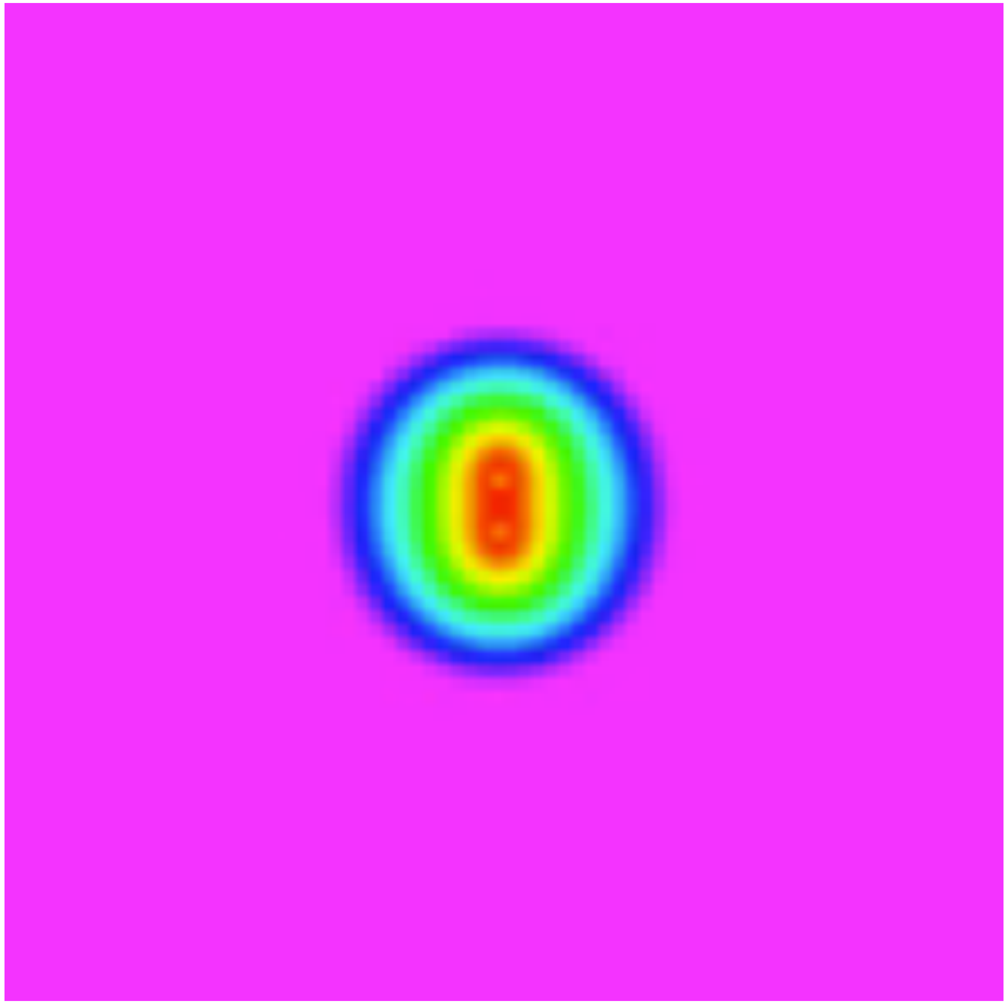
Electronic excited states, electron dynamics under external field, …

Real-time response: Optical response of Ethylene (C_2H_4) molecule

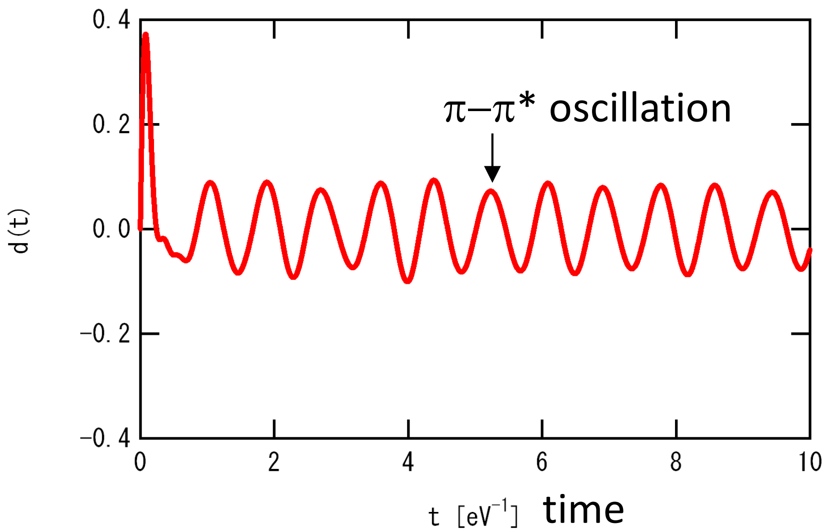
Electron dynamics in ethylene

(Applying an impulse at t=0)

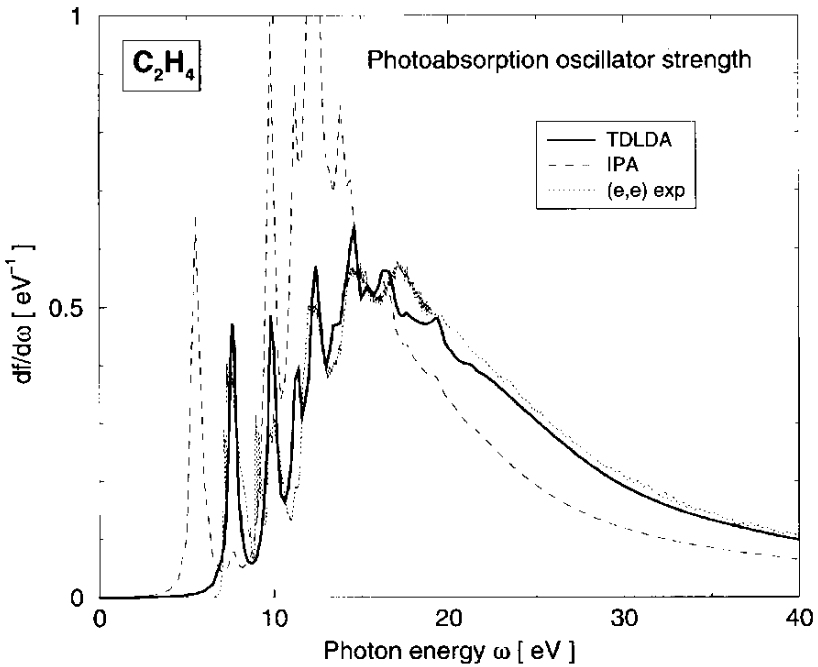




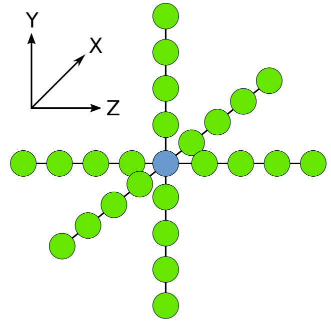
Dipole moment

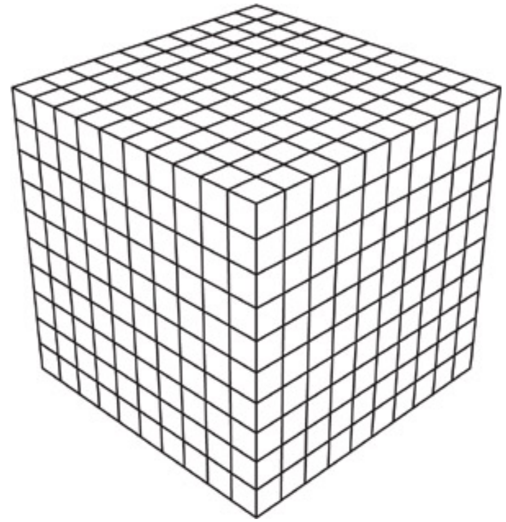


Photoabsorption cross section



Real-time and real-space method





Real-space grid representation
in 3D Cartesian coordinate
- High-order finite difference

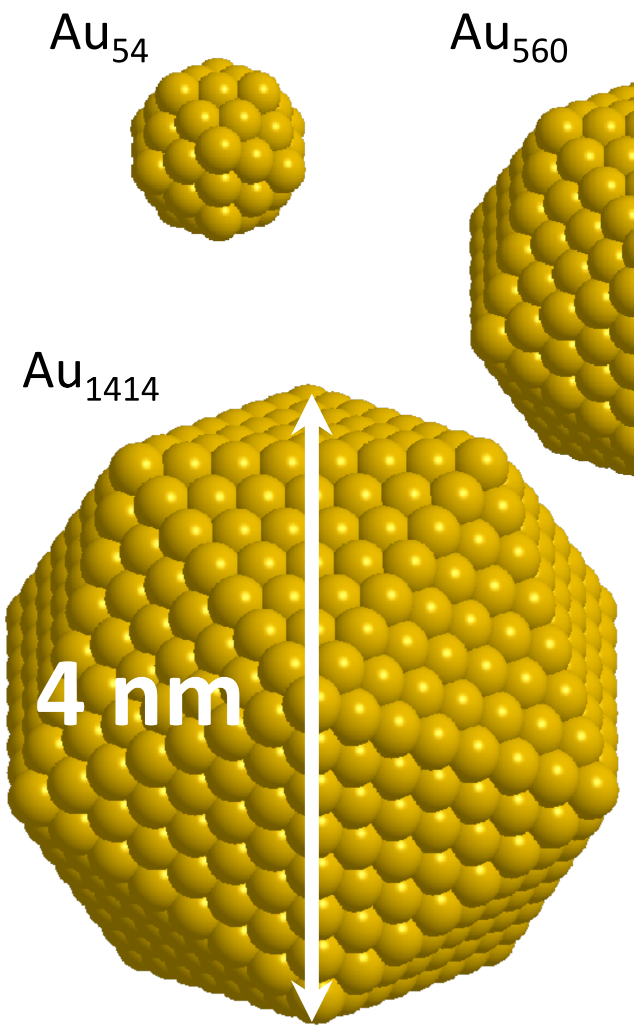
$$-\frac{\hbar^2}{2m}\left[\sum_{n_1=-N}^{N} C_{n_1}\psi(x_i+n_1h,y_j,z_k)+\sum_{n_2=-N}^{N} C_{n_2}\psi(x_i,y_j+n_2h,z_k)+\sum_{n_3=-N}^{N} C_{n_3}\psi(x_i,y_j,z_k+n_3h)\right]$$
$$+[V_{\text{ion}}(x_i,y_j,z_k)+V_H(x_i,y_j,z_k)+V_{\text{xc}}(x_i,y_j,z_k)]\psi(x_i,y_j,z_k)=E\psi(x_i,y_j,z_k)\ .$$

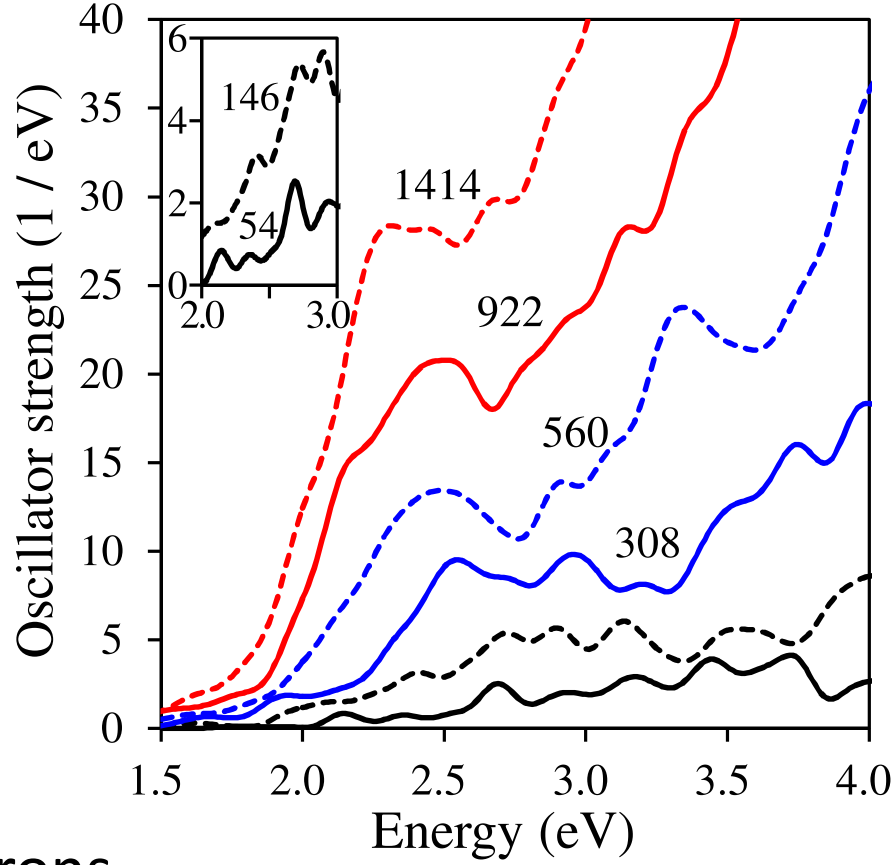
Time evolution calculation by explicit method
(Taylor expansion of 4th order)

$$\psi_i(t+\Delta t)=\exp\left[\frac{h_{KS}(t)\Delta t}{i\hbar}\right]\psi_i(t)\approx\sum_{k=0}^{N}\frac{1}{k!}\left(\frac{h_{KS}(t)\Delta t}{i\hbar}\right)\psi_i(t),\qquad N=4$$

Calculation of large systems using massively parallel supercomputers

Surface plasmon resonance of Au clusters



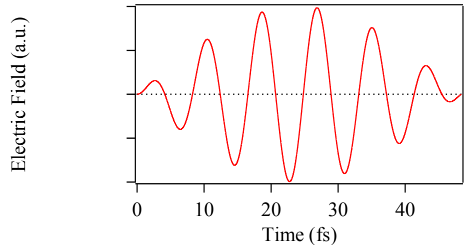


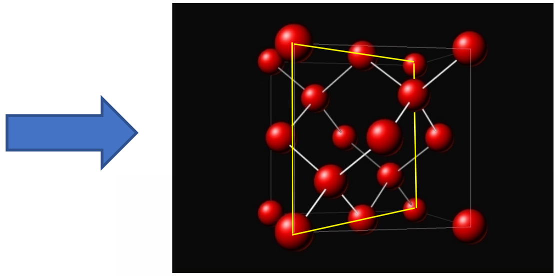
15,554 electrons

K. Iida, M. Noda, K. Ishimura, K. Nobusada, JPCA, 118, 11317 (2014)

(Group of Institute for Molecular Sciences, Okazaki)

Crystalline silicon under intense laser pulse





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

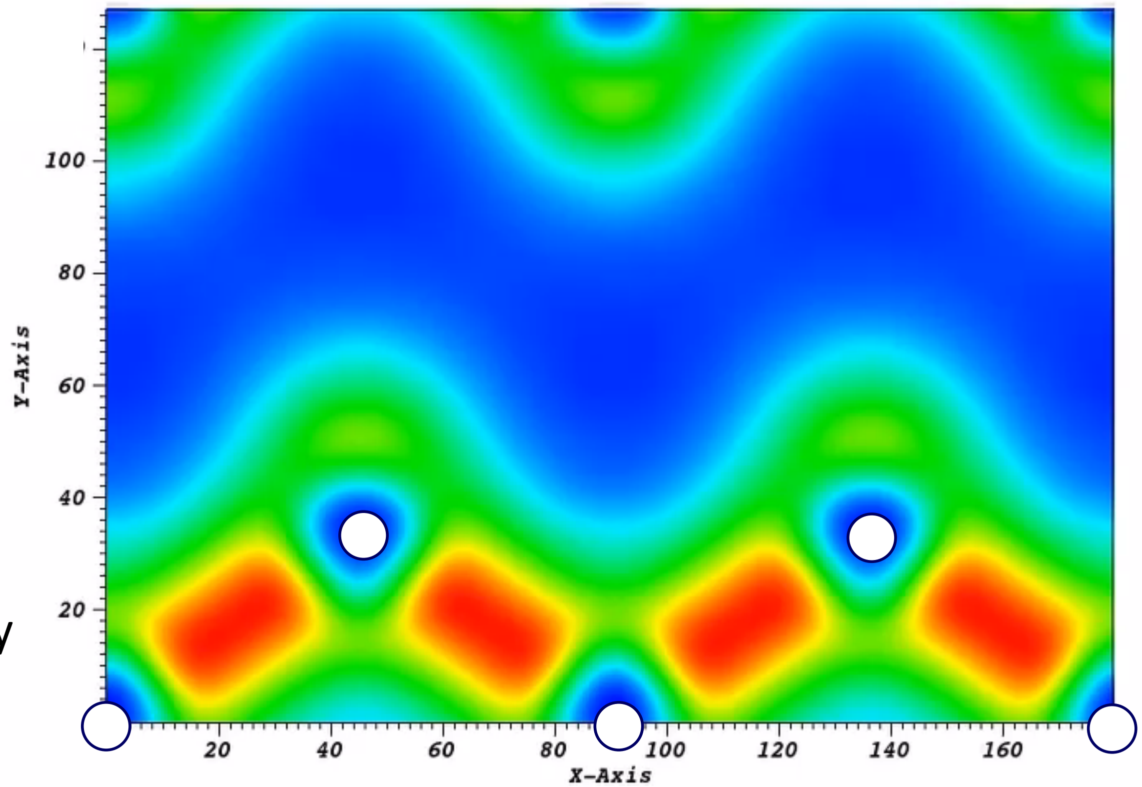
(direct bandgap 2.4 eV in LDA)

$E = 27.5$ V/nm

$\hbar\omega = 1.55$ eV

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

Electron density



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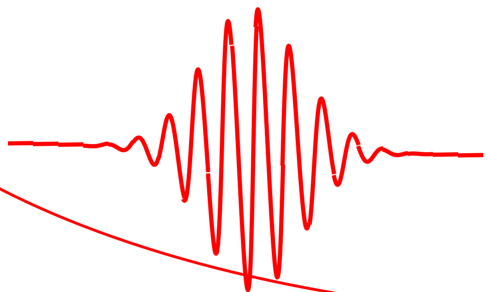

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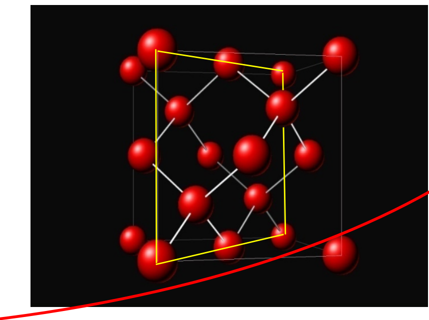


Usually, two simulations are separately used in optical sciences

Macroscopic Electromagnetism (EM)

Light propagation description by Maxwell equations. Materials' properties comes into through constitutive relations (dielectric constant).

$$\nabla \cdot \boldsymbol{B} = 0$$

$$\nabla \times \boldsymbol{E} + \frac{\partial \boldsymbol{B}}{\partial t} = \boldsymbol{0}$$

$$\nabla \cdot \boldsymbol{D} = \rho$$

$$\nabla \times \boldsymbol{H} - \frac{\partial \boldsymbol{D}}{\partial t} = \boldsymbol{j}$$

Quantum Mechanics (QM)

First-principles calculations for dielectric function.

Perturbation theory in quantum mechanics.

$$\varepsilon_r = 1 + \frac{2Ne^2}{\varepsilon_0 \hbar} \sum_j \frac{\omega_{j0} \left| \langle 0 | x | j \rangle \right|^2}{\omega_{j0}^2 - (\omega + i\gamma)^2}$$

$$D = \varepsilon E$$

Constitutive relation connects two theories

However, at the frontiers of optical science, unified simulation is required.

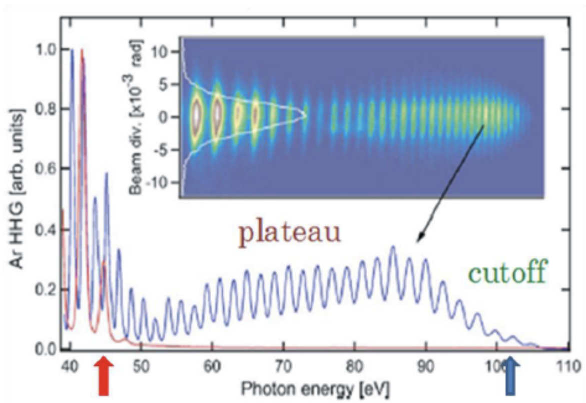
Ordinary electromagnetism

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

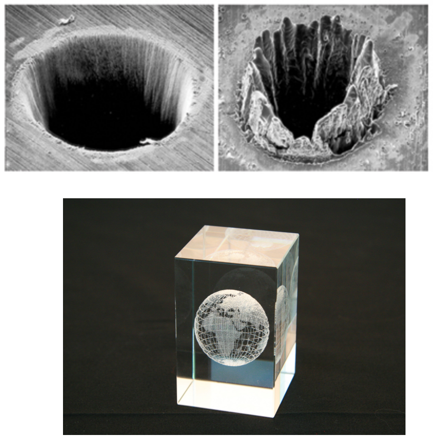
Linearity and locality

In forefront optical science, a theoretical and computational approach unifying Electromagnetism and Quantum Mechanics is required.

Extreme nonlinear optics

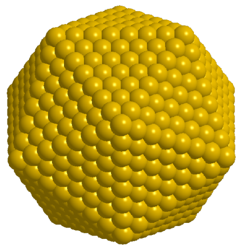


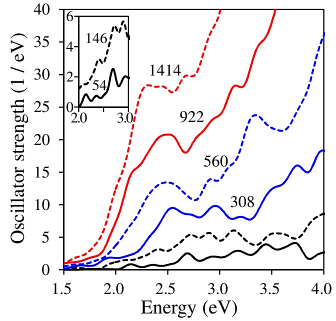
High harmonic generation from atoms and solids

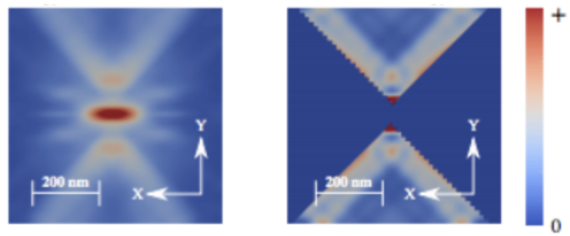


Nonthermal laser processing using femtosecond laser pulses

Nano-optics







Near field induces strong nonlinear interactions

第一原理電子ダイナミクス計算プログラム

ARTED

Ab-intio Real-Time Electron Dynamics symulator

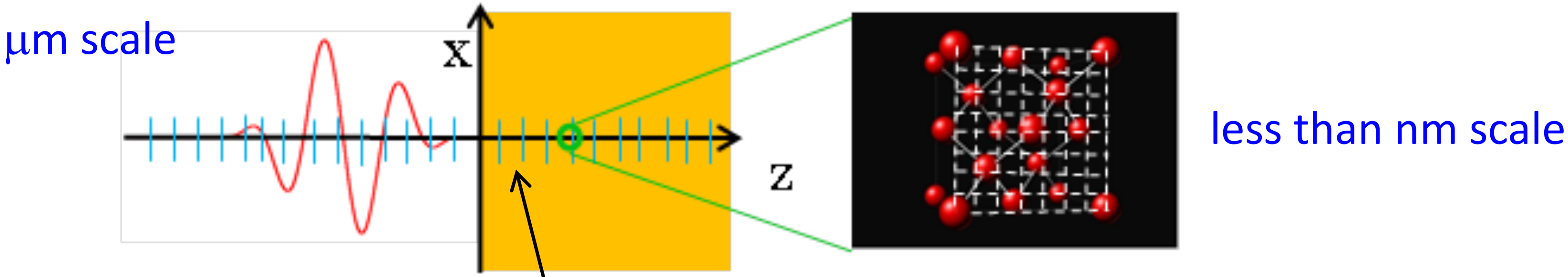
→ SALMON

Light propagation description: Coupling with Maxwell equations

'Multi-scale' coupling

Macroscopic grid
for Maxwell equation

Microscopic grid
for RT-TDDFT electron dynamics



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

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

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

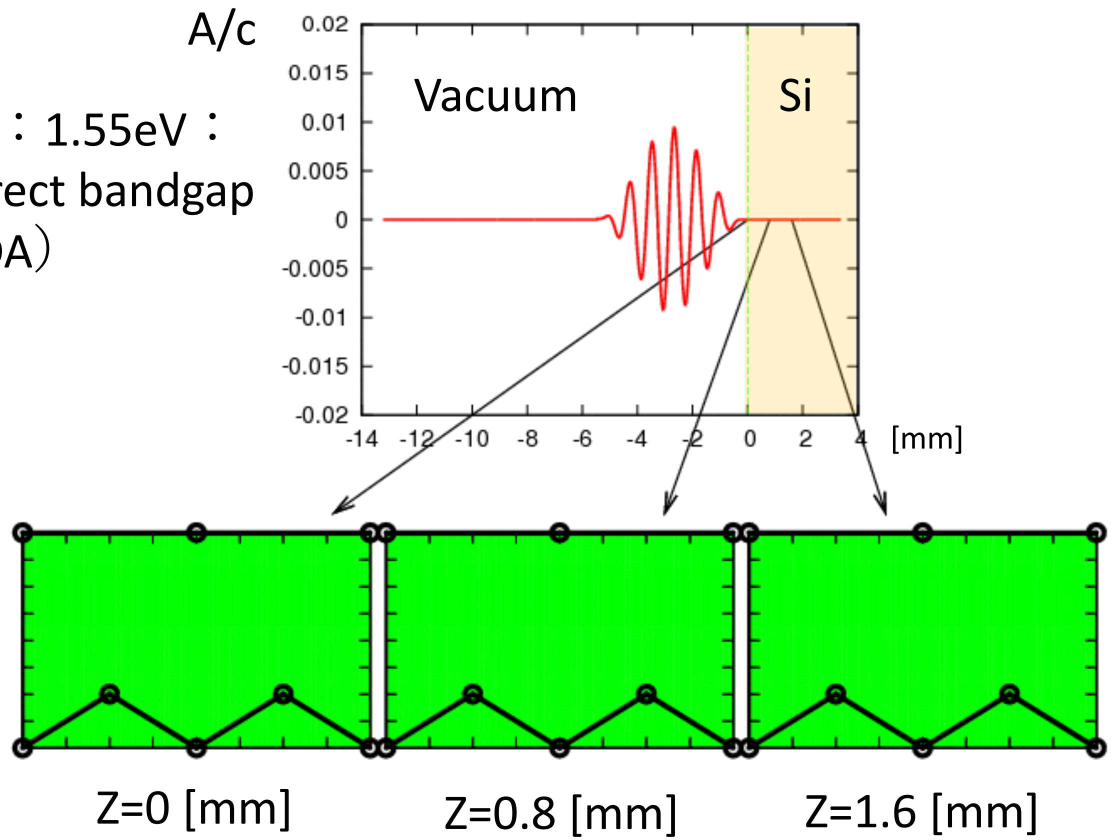
At each macroscopic grid point,
we solve real-time electron dynamics in parallel

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

Ab-initio simulation for light-wave propagation in Si

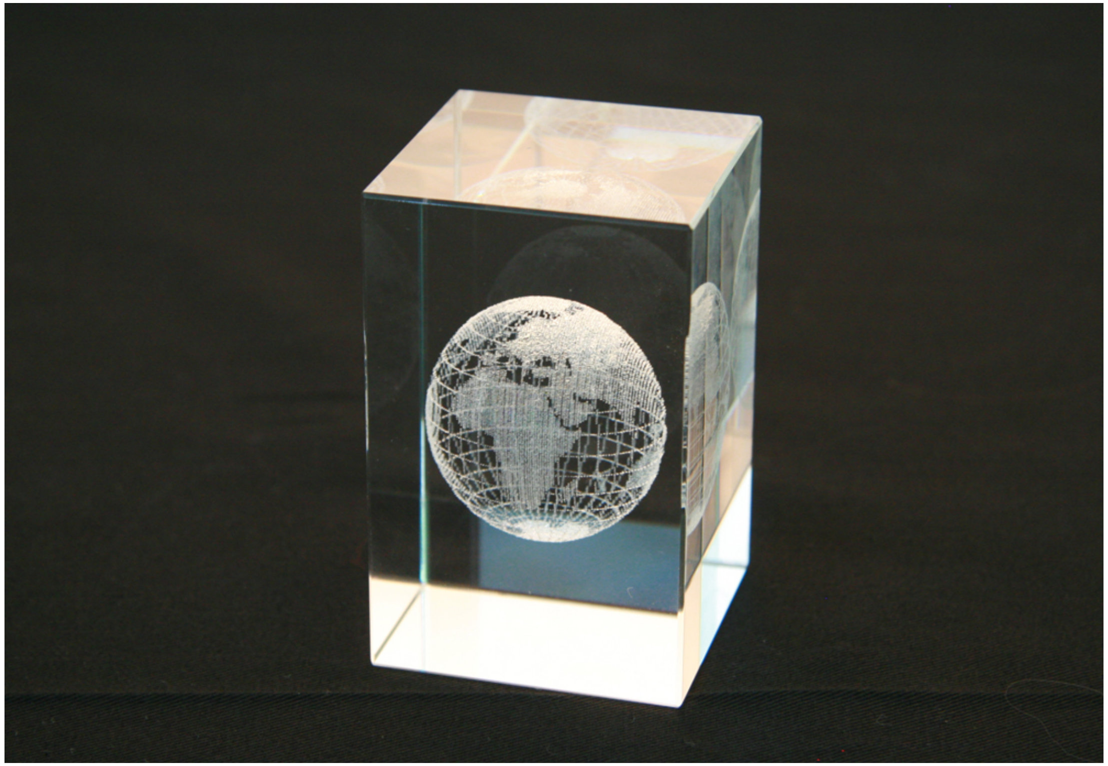
$I=10^{10} W/cm^2$

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

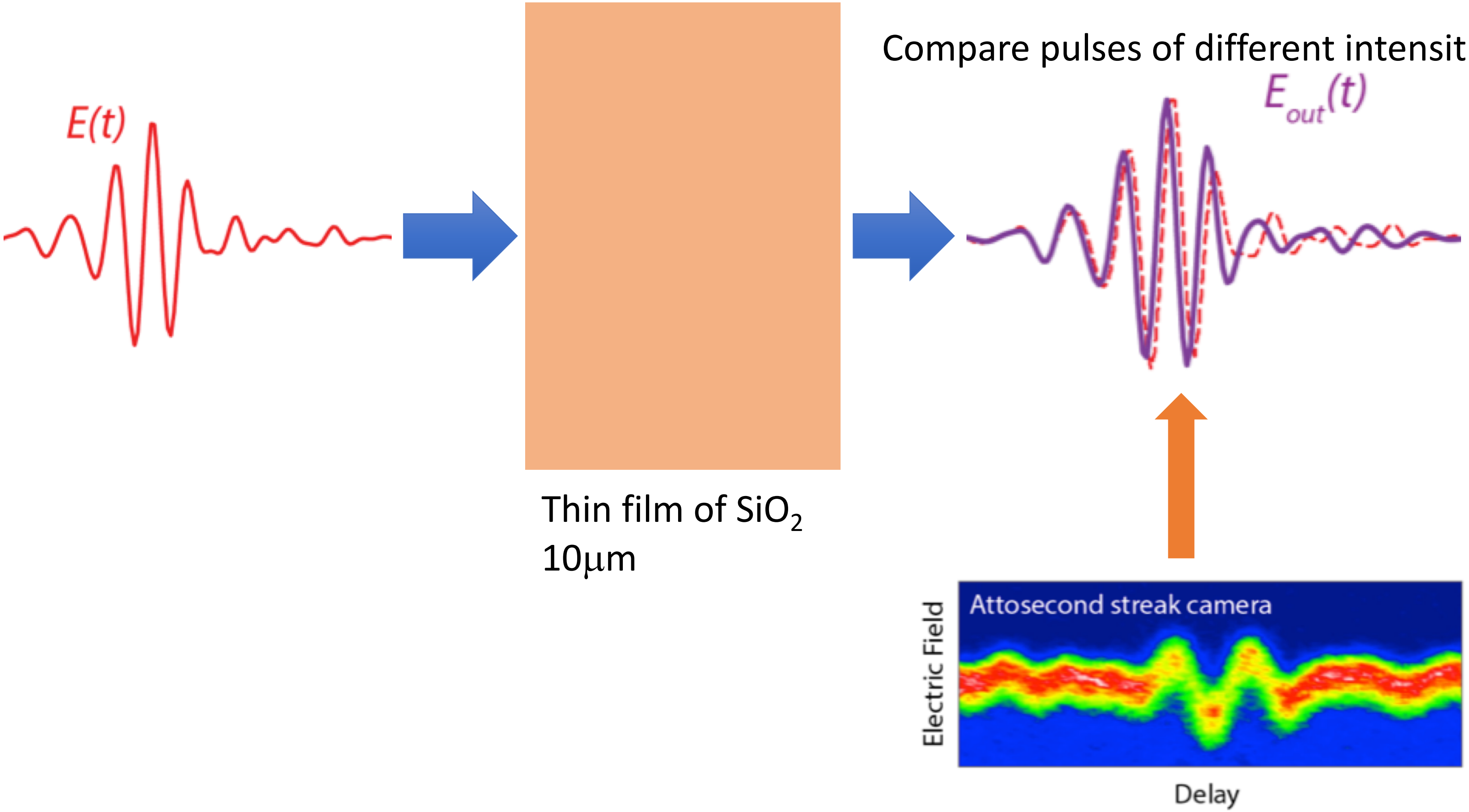


Towards Laser processing of transparent materials

At which intensity of light, glass changes from transparent to opaque material ?



Laser pulse propagation through SiO_2 10μm thin film



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

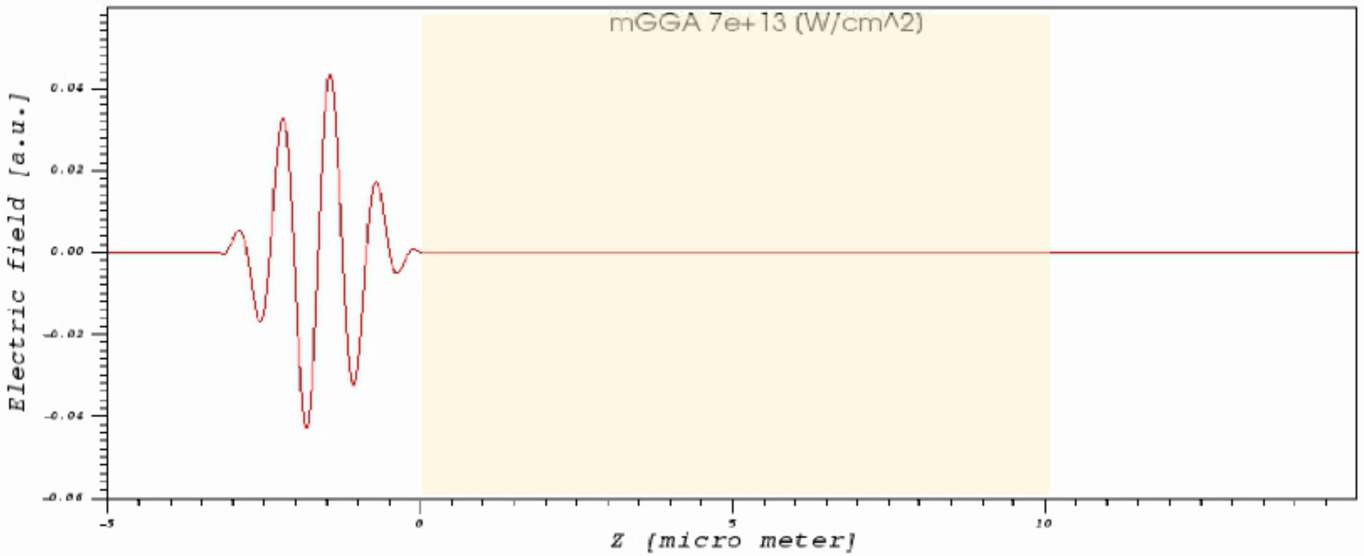
Maxwell + TDDFT multiscale simulation : 10 mm SiO_2

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

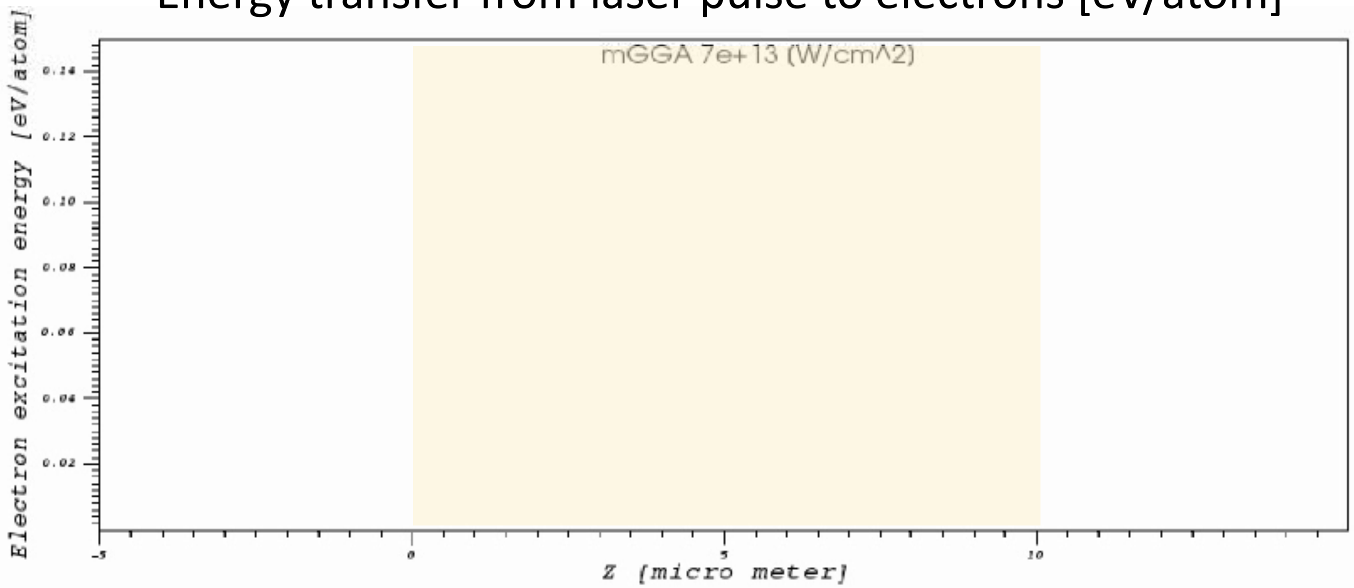
$\hbar\omega = 1.55\text{eV}$

$\lambda = 800\text{nm}$

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



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

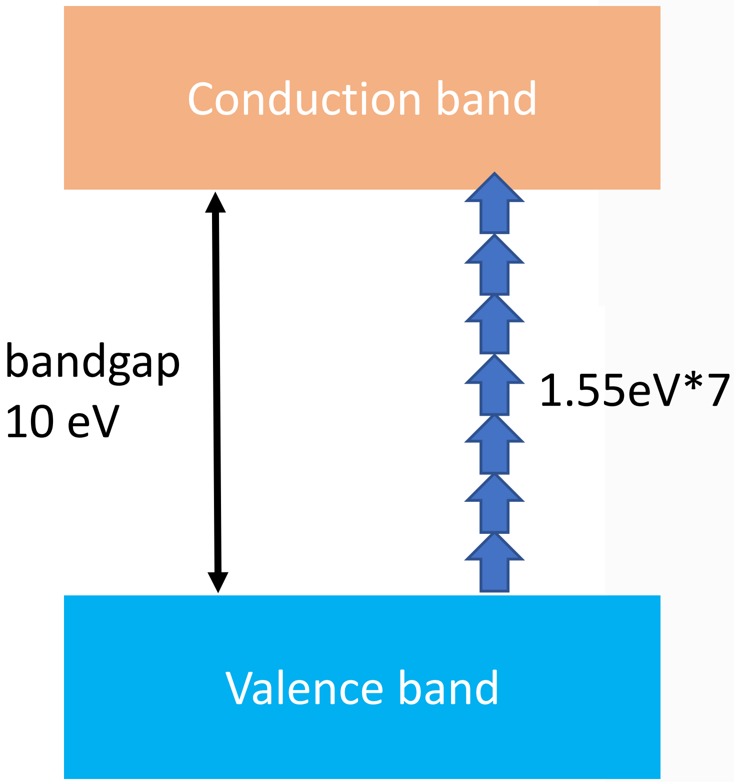


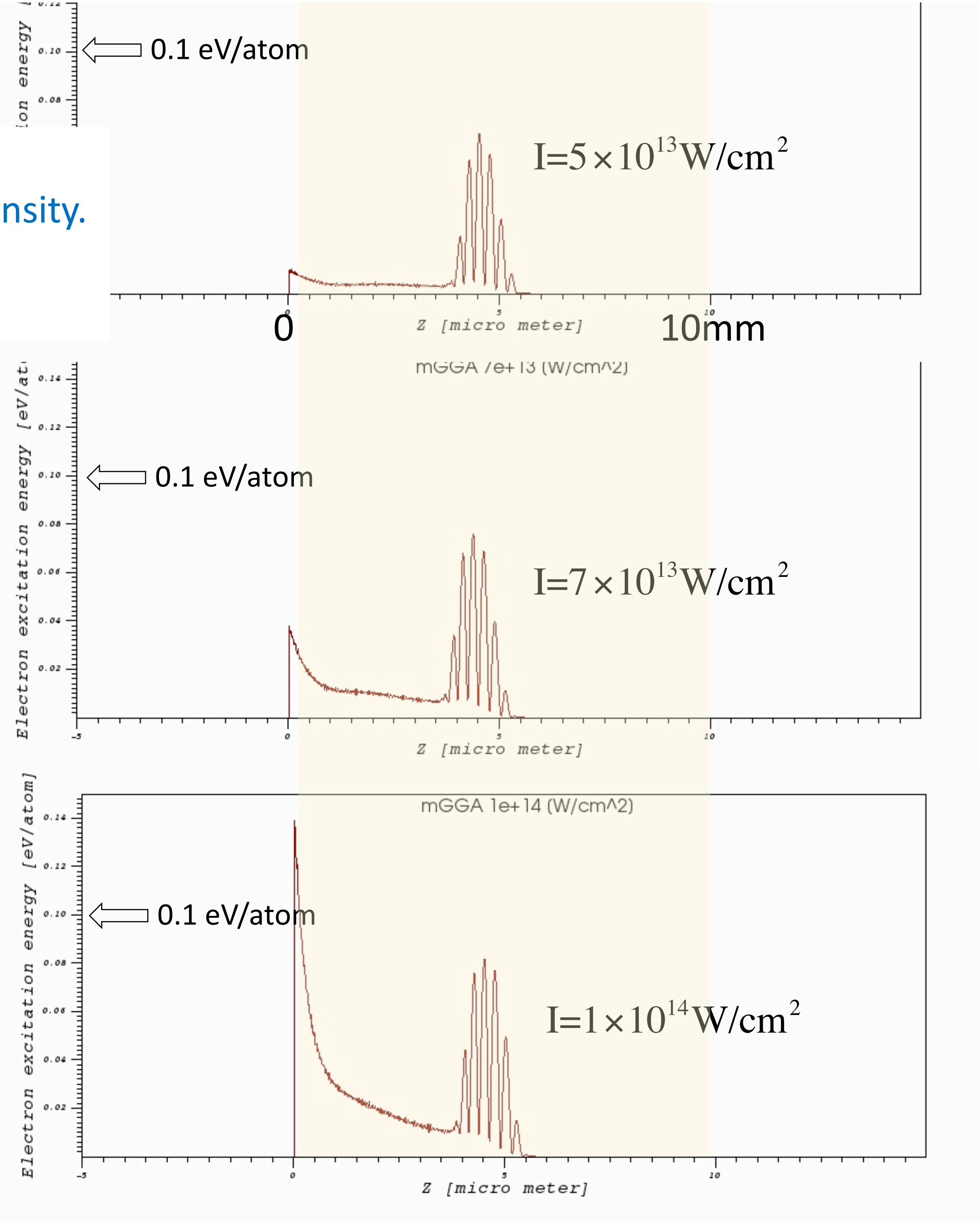
80,000 cores, 20 hours
at K-Computer, Kobe

Energy deposited to electrons in SiO_2 increases rapidly at a certain laser intensity.

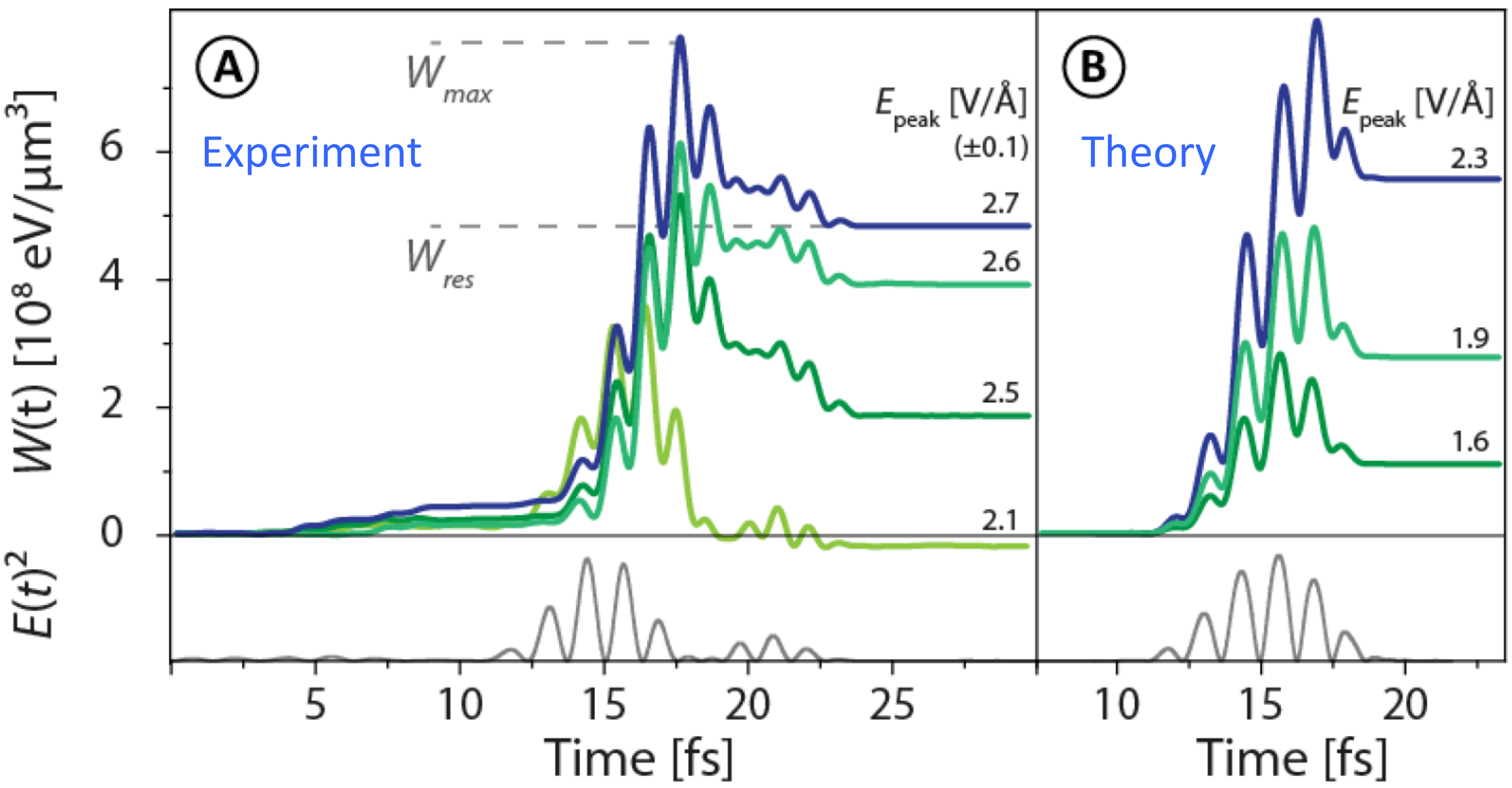
>> Damage threshold

Multiphoton/Tunneling excitation





Energy deposition from laser pulse to SiO_2 at mid point (5μm)



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

3D Maxwell + 3D RT-TDDFT simulation: a computational challenge

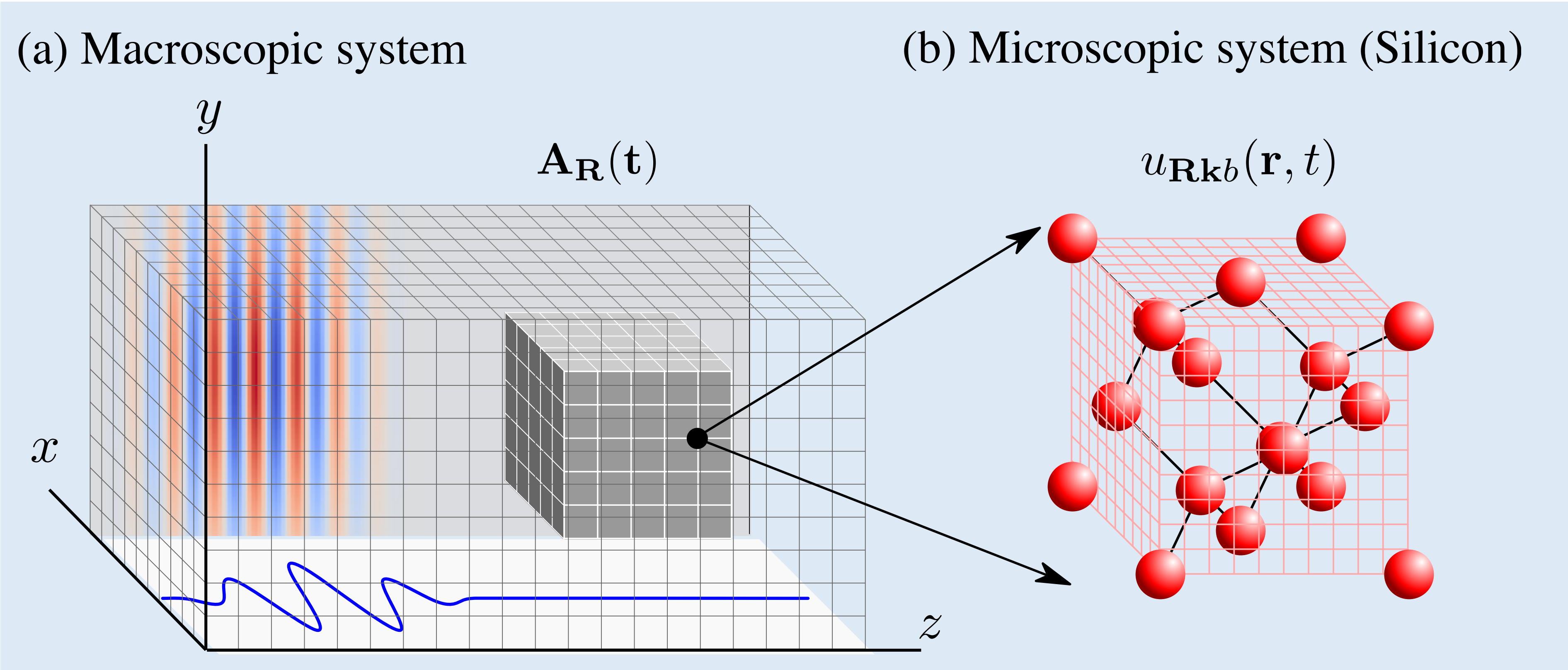
Our university started to operate supercomputer 'Oakforest-PACS' from 2016.11.

- 1st in Japan
- 9th in the world (Nov, 2017)

8208 nodes x 68 cores (Intel Xeon Phi 7250)

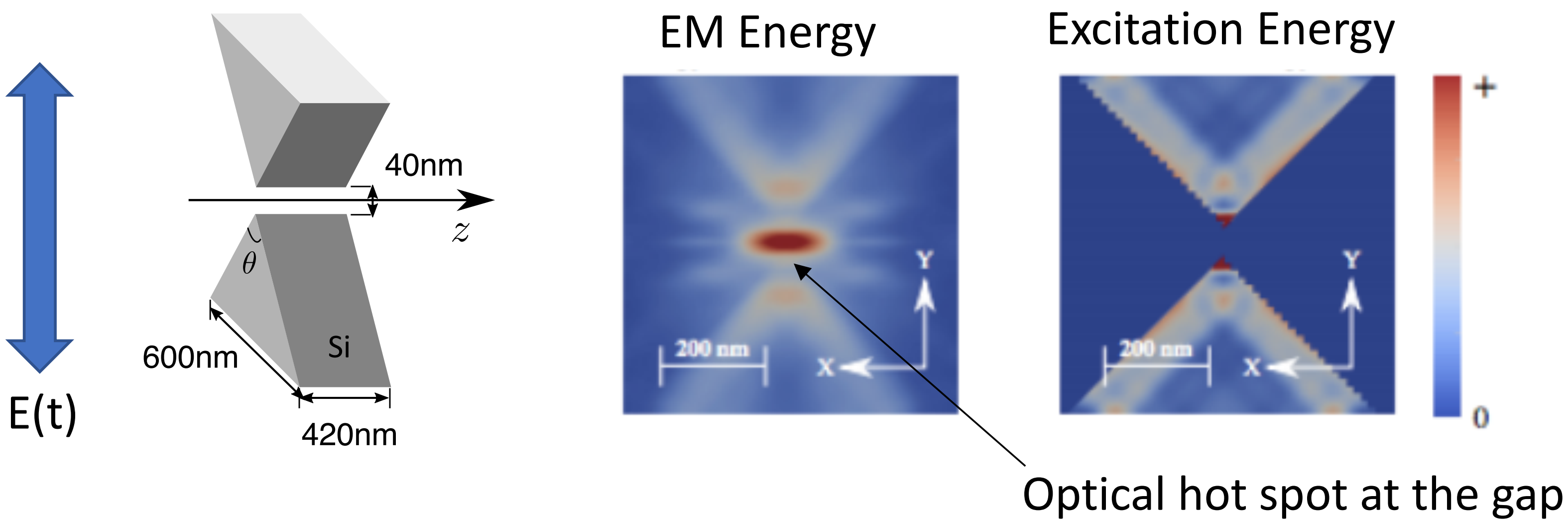
25 PFLOPS

2017.3 We had 3-day machine time to use full nodes.

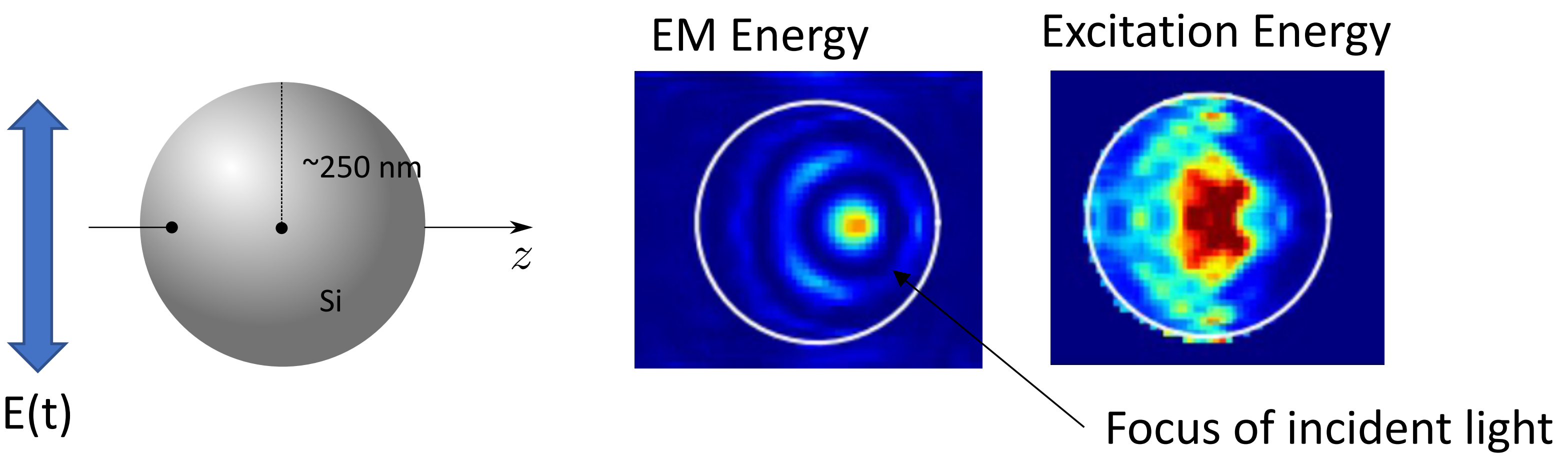


Strong laser pulses on Silicon nano-structures: Two test cases

- Silicon triangular prisms

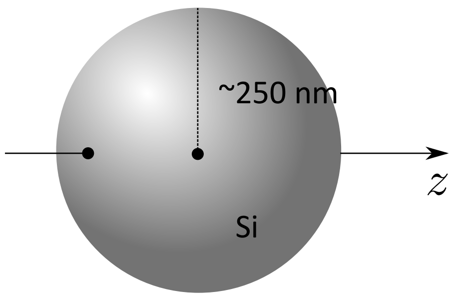


- Silicon nanosphere



We anticipated: dielectric response for weak, and metallic response for strong laser pulses.

Laser pulse (10^{12}W/cm^2, 1.55eV, 5fs) on silicon-nanosphere

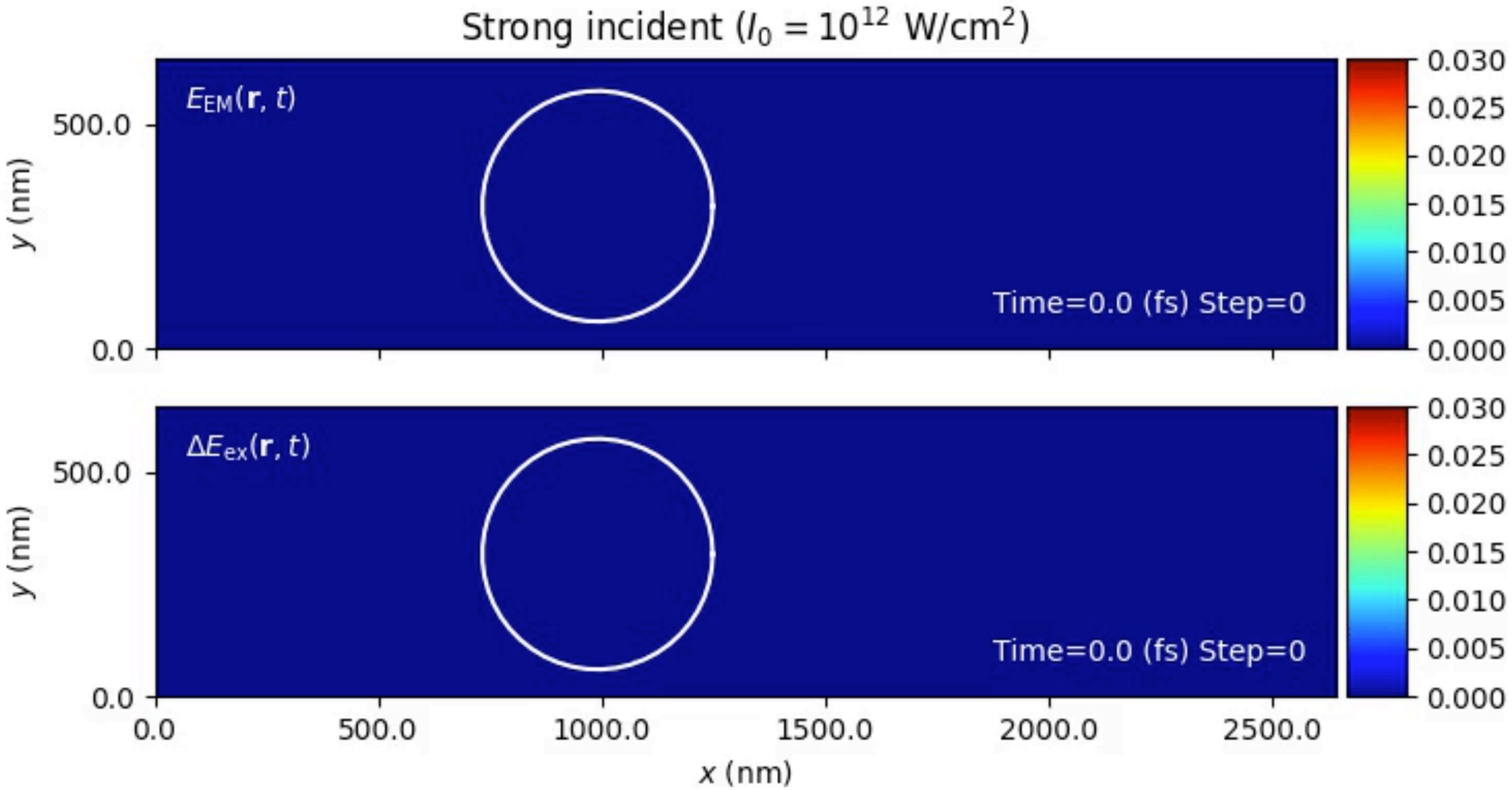


- 3D Maxwell + 3D TDDFT multiscale calculation
- Si sphere is expressed by 32,752 points
  (32,752 electron dynamics calculation in parallel)
- 2.5 hourse by Oakforest-PACS 8188 nodes

Energy density of EM fields

$E^2 + B^2$

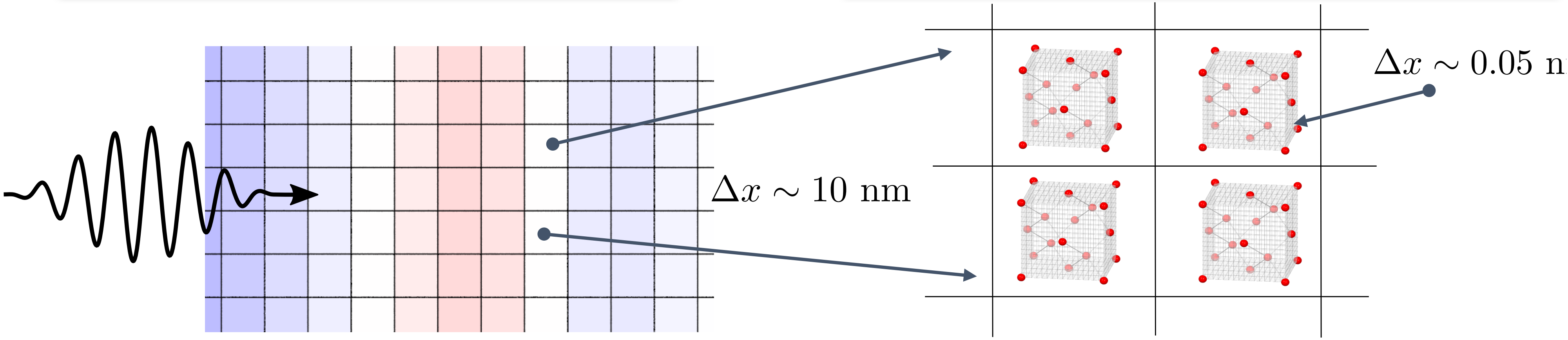
Electronic excitation energy (two photon absorption)



Multiscale Maxwell-TDDFT calculation

Macroscopic system (EM field)

Microscopic system (Electron dynamics)



FDTD calculation is inexpensive
and carried out redundantly in nodes.

1 node calculates 4 electron dynamics.
(32,752 electron dynamics by 8,188 nodes)

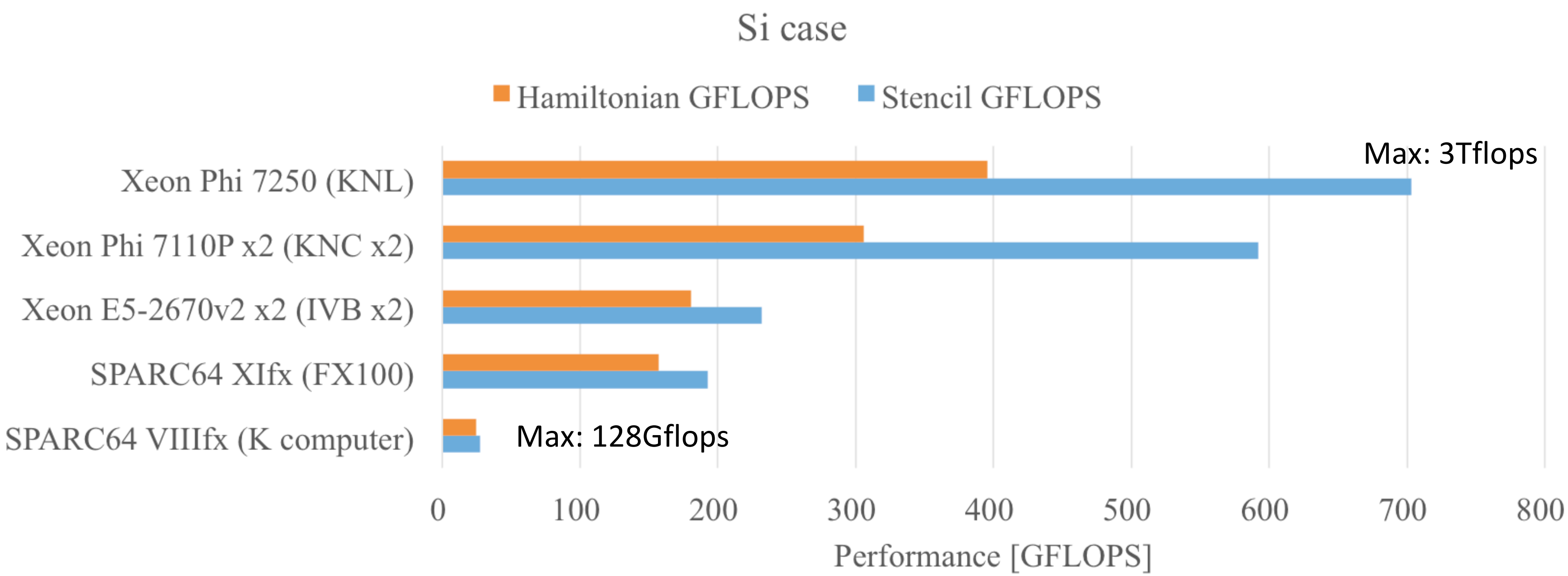
Each electron dynamics calculation uses

- 16x16x16 spatial grids
- 8x8x8 k-points
- 16 orbitals

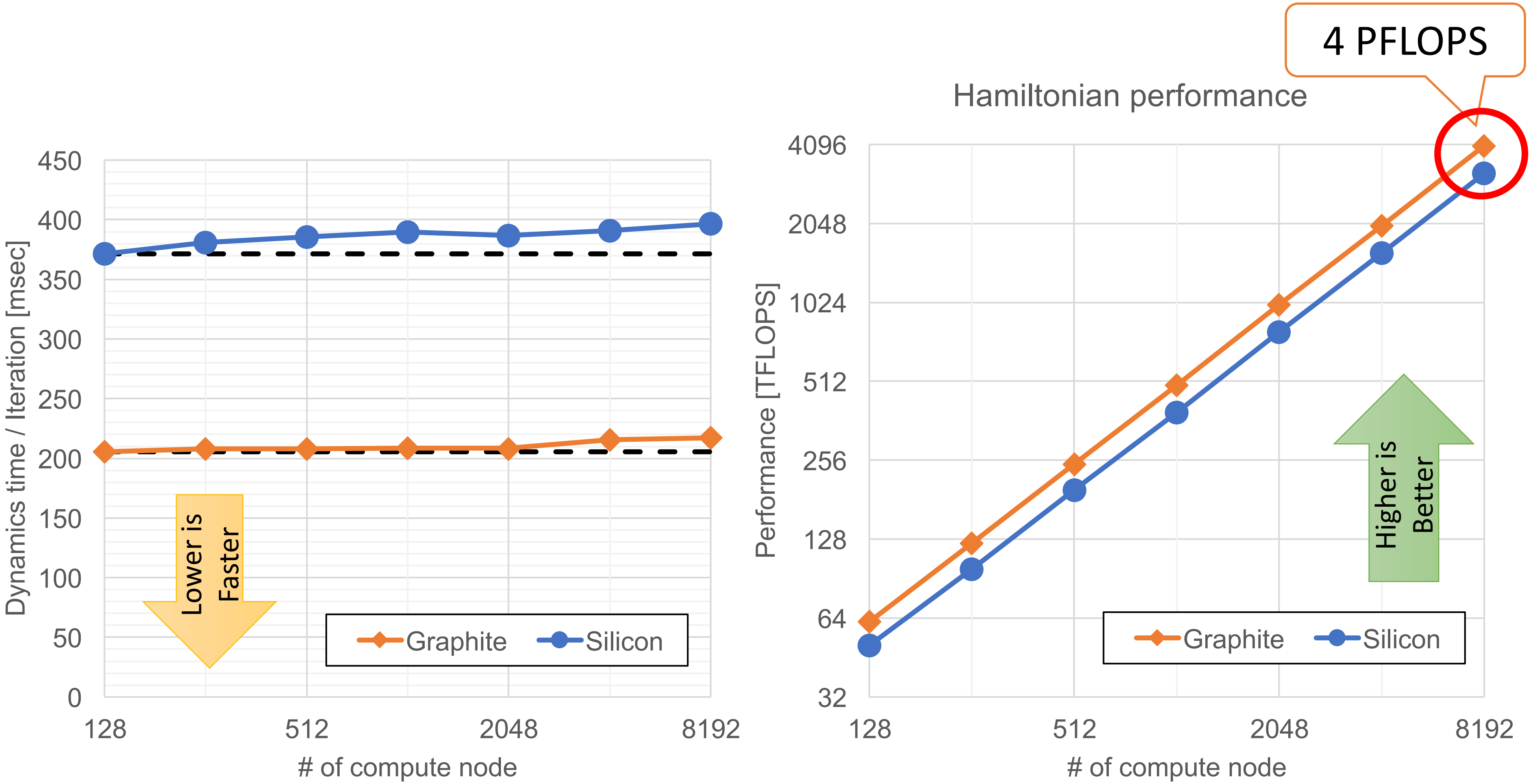
Total number of variables : 1.1 x 10^{12}
Number of time step : 20,000

Performance in various processors

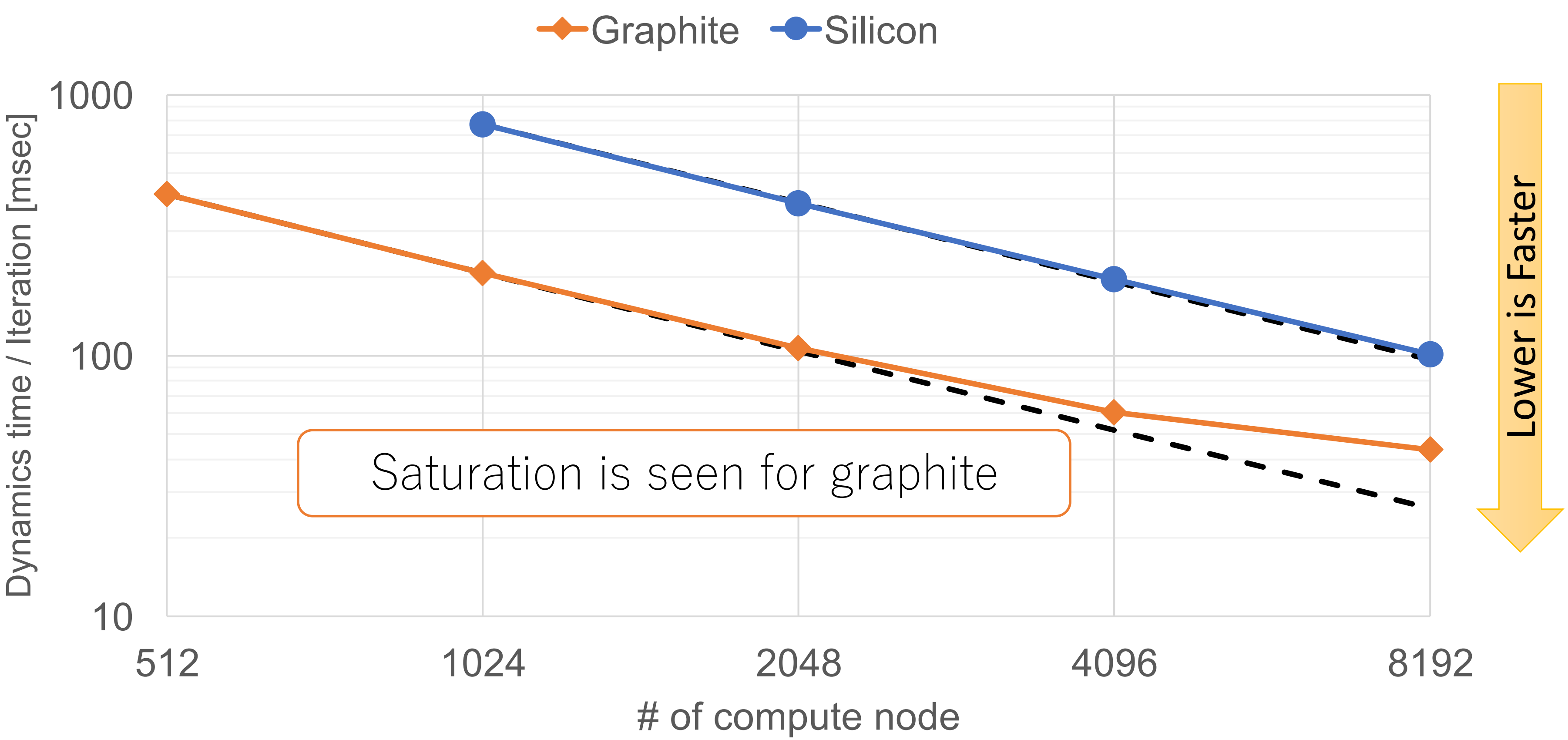
In-House collaboration with Computer Science group
(Prof. Boku, Ph.D student Hirokawa)



Weak scaling



Strong scaling



Summary

We are developing SALMON

- ab-initio calculation for light-matter interaction
- large-scale computing for Maxwell + TDDFT multiscale simulation

- in-house collaboration between application and computer researchers
- good scaling and performance using many-core processors

- provide numerical experiment platform for forefront optical science
- to be usable by experimental and company researchers

SALMON-TDDFT Code-Project

Scalable **A**b-initio **L**ight-**M**atter simulator for **O**ptics and **N**anoscience

Open-source, Real-time TDDFT (+Maxwell)

http://salmon-tddft.jp/

Acknowledgement

Collaborators

Univ. Tsukuba
- Mitsuharu Uemoto
- Yuta Hirokawa
- Taisuke Boku

Univ. Tokyo
- Yasushi Shinohara

Max-Planck Institute for Structure and Dynamics of Matter
- Shunsuke Sato

Univ. Washington
- George F. Bertsch

Max Planck Institute for Quantum Optics
- Annkatrin Sommer
- Martin Schultze
- Ferenc Krausz

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CREST 戦略的創造研究推進事業 Core Research for Evolutionary Science and Technology

CDMSI

科研費 KAKENHI

Supercomputers

京 K computer

JCAHPC