CCS-LBNL Collaborative Workshop
Mar. 5-6, 2018

Ab-initio Density Functional Simulation for Nano-Optics

K. YABANA

Division of Quantum Condensed Matter Physics
Center for Computational Sciences
University of Tsukuba

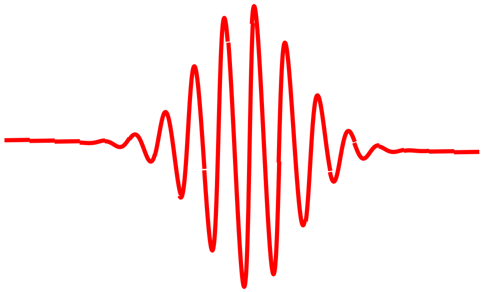
筑波大学
計算科学研究センター
Center for Computational Sciences

筑波大学
University of Tsukuba

Two kinds of simulations in optical science

Macroscopic Electromagnetism (EM)

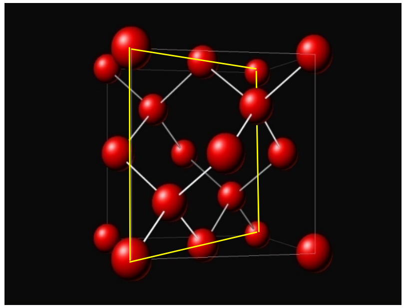
Light propagation described by Maxwell's equations



wave length ~ 1μm (10^{-6}m)

Quantum Mechanics (QM)

Electron dynamics described by Schrödinger equation



Atomic size ~ 1nm (10^{-9}m)

$$\nabla \cdot \boldsymbol{B} = 0$$
$$\nabla \times \boldsymbol{E} + \frac{\partial \boldsymbol{B}}{\partial t} = \boldsymbol{0}$$
$$\nabla \cdot \boldsymbol{D} = \rho$$
$$\nabla \times \boldsymbol{H} - \frac{\partial \boldsymbol{D}}{\partial t} = \boldsymbol{j}$$

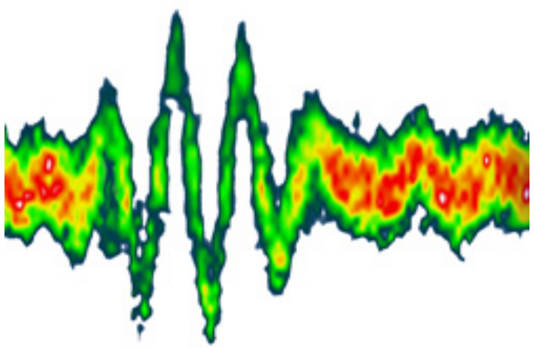
$$D = \varepsilon E$$

Constitutive relation

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

Forefront Extreme Optics requires simulations combining EM and QM

Ultrafast

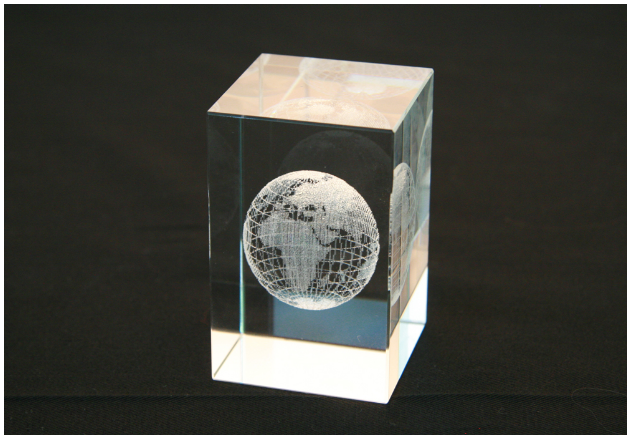


(Measurement of light electric field at Max Planck Inst. Quantum Optics)

Attosecond science

Measurements of electron dynamics in time domain

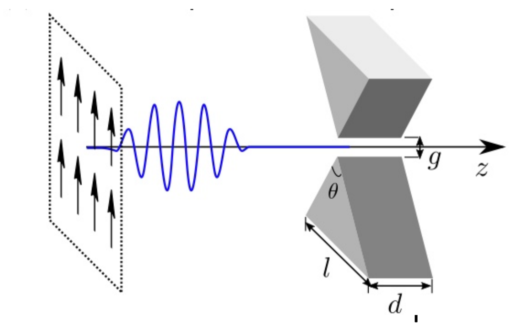
Nonlinear

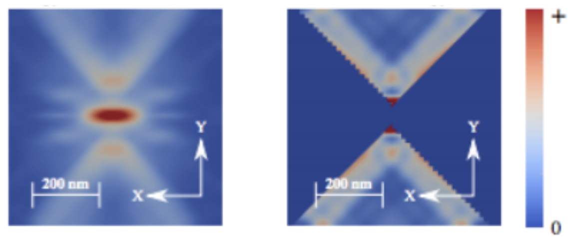


Laser processing

Transparent material absorbs light by nonlinear electron dynamics

Nonlocal





Nano-optics, near field

light wave length comparable to material size http://salmon-tddft.jp

We develop SALMON

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

Multi-scale modeling for light-matter interactions
starting with ab-initio electron dynamics calculations

Univ. Tsukuba + Inst. Molecular Sciences (Okazaki)

Supported by

Post-K priority issue 7
as one of 7 subjects on
“Creation of new functional devices and high-performance materials to support next-generation industries” (2014-2020)

2 post-docs

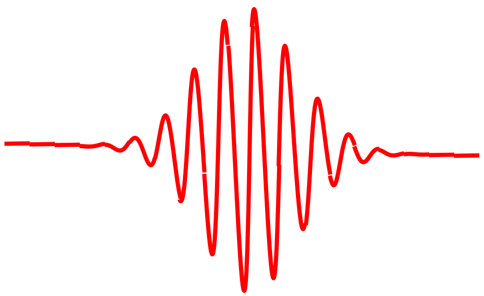
JST-CREST project
“Development and applications of first-principles software for unified photonic and electronic systems” (2016-2022)

3 post-docs + 1 PhD student (Hirokawa, CS)

Two kinds of simulations in optical science

Macroscopic Electromagnetism (EM)

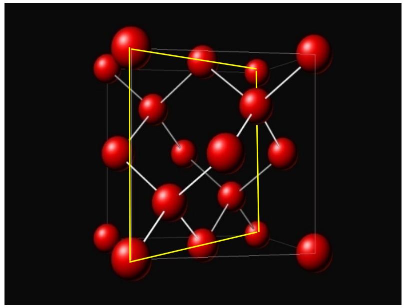
Light propagation described by Maxwell's equations



wave length ~ 1μm (10^{-6}m)

Quantum Mechanics (QM)

Electron dynamics described by Schrödinger equation



Atomic size ~ 1nm (10^{-9}m)

$$\nabla \cdot \boldsymbol{B} = 0$$
$$\nabla \times \boldsymbol{E} + \frac{\partial \boldsymbol{B}}{\partial t} = \boldsymbol{0}$$
$$\nabla \cdot \boldsymbol{D} = \rho$$
$$\nabla \times \boldsymbol{H} - \frac{\partial \boldsymbol{D}}{\partial t} = \boldsymbol{j}$$

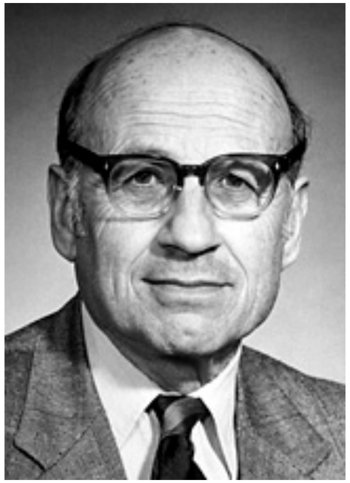
$$D = \varepsilon E$$

Constitutive relation

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

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

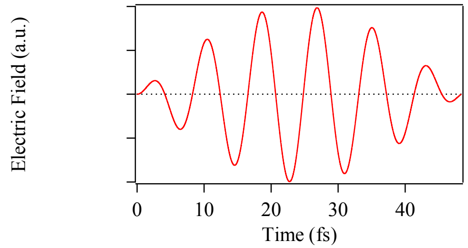
Theory for ground state. Not applicable to photoexcitation

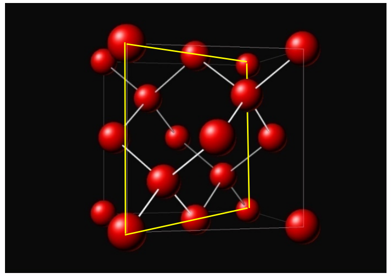
Extension to electron dynamics: Time-dependent DFT (TDDFT)

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

Electronic excited states, electron dynamics under external field, …

Crystalline silicon under an intense laser pulse





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

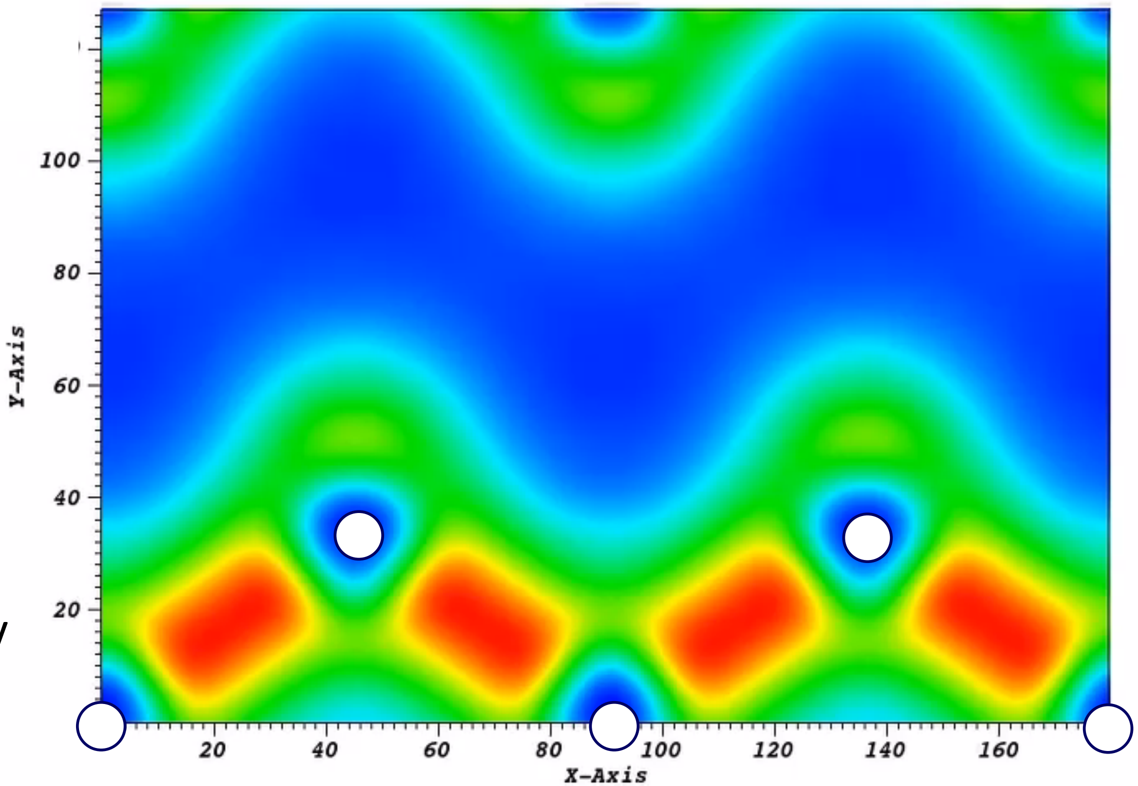
(direct bandgap 2.4 eV in LDA)

$E = 27.5$ V/nm

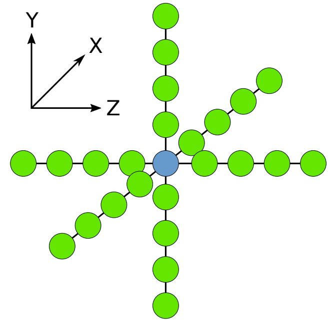
$\hbar\omega = 1.55$ eV

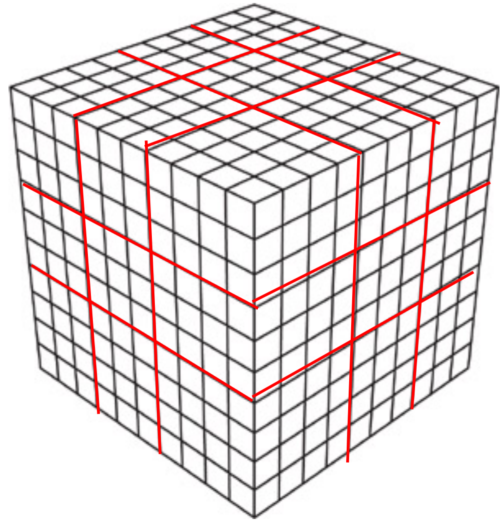
T_{FWHM} =7 fs

Electron density



Real-time and real-space finite difference method





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

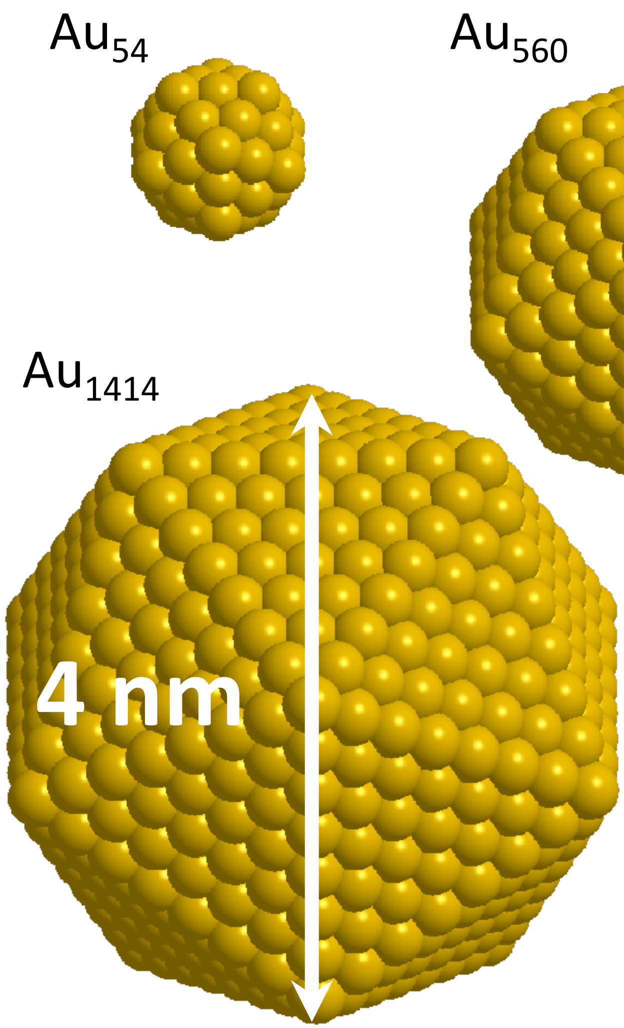
$$-\frac{\hbar^2}{2m}\left[\sum_{n_1=-N}^{N} C_{n_1}\psi(x_i+n_1h,y_j,z_k)+\sum_{n_2=-N}^{N} C_{n_2}\psi(x_i,y_j+n_2h,z_k)+\sum_{n_3=-N}^{N} C_{n_3}\psi(x_i,y_j,z_k+n_3h)\right]$$
$$+[V_{\text{ion}}(x_i,y_j,z_k)+V_H(x_i,y_j,z_k)+V_{\text{xc}}(x_i,y_j,z_k)]\psi(x_i,y_j,z_k)=E\psi(x_i,y_j,z_k)\ .$$

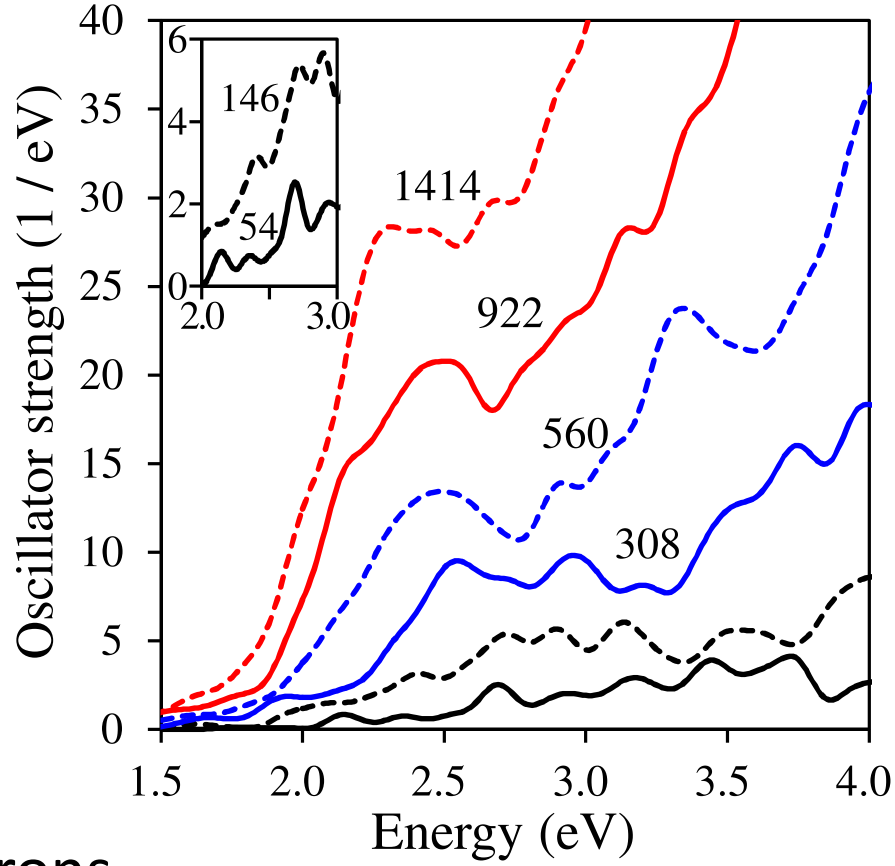
Time evolution calculation by explicit method
(Taylor expansion of 4^{th} order)

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

Calculation of large systems using massively parallel supercomputers

Surface plasmon resonance of Au clusters





15,554 electrons

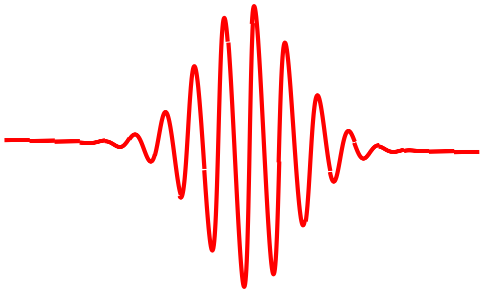
K. Iida, M. Noda, K. Ishimura, K. Nobusada, JPCA, 118, 11317 (2014)

(Group of Institute for Molecular Sciences, Okazaki)

Two kinds of simulations in optical science

Macroscopic Electromagnetism (EM)

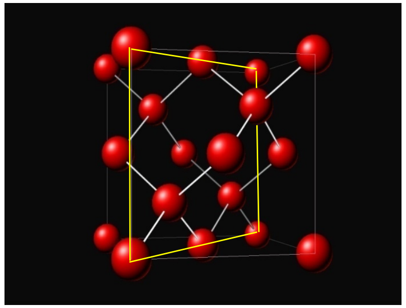
Light propagation described by Maxwell's equations



wave length ~ 1μm (10^{-6}m)

Quantum Mechanics (QM)

Electron dynamics described by Schrödinger equation



Atomic size ~ 1nm (10^{-9}m)

$$\nabla \cdot \boldsymbol{B} = 0$$
$$\nabla \times \boldsymbol{E} + \frac{\partial \boldsymbol{B}}{\partial t} = \boldsymbol{0}$$
$$\nabla \cdot \boldsymbol{D} = \rho$$
$$\nabla \times \boldsymbol{H} - \frac{\partial \boldsymbol{D}}{\partial t} = \boldsymbol{j}$$

$$D = \varepsilon E$$

Constitutive relation

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

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

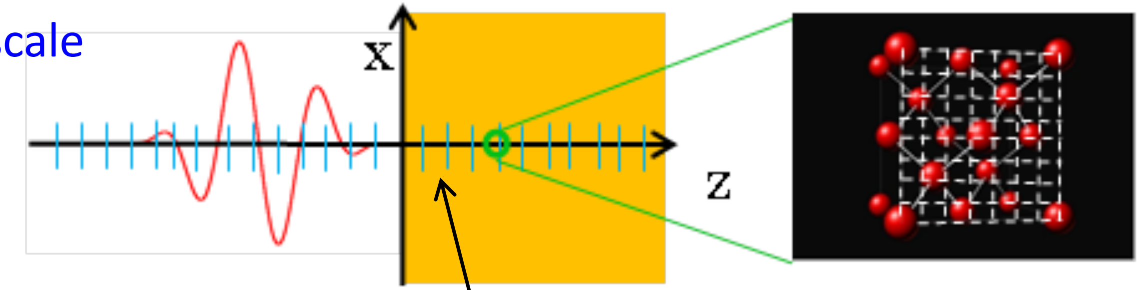
Light propagation description: Coupling with Maxwell equations

Multi-scale modeling

Macroscopic grid
for Maxwell equation

Microscopic grid
for RT-TDDFT electron dynamics

μm scale



less than nm scale

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

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

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

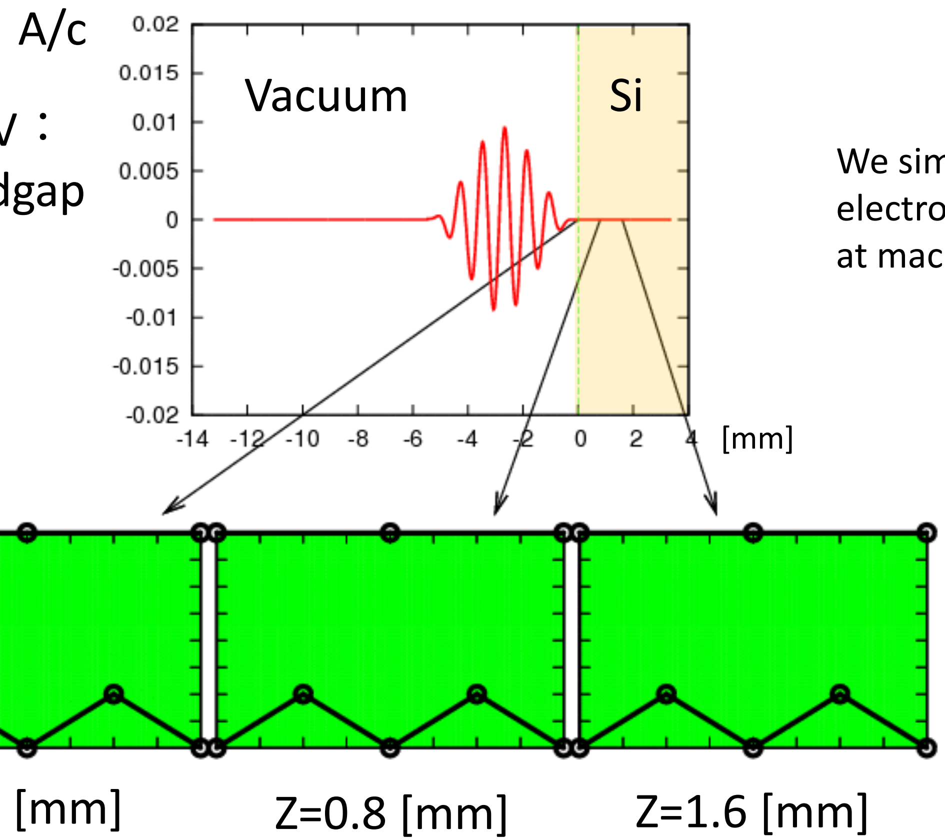
At each macroscopic grid point,
we solve real-time electron dynamics in parallel

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

Ab-initio simulation for light-wave propagation in Silicon

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

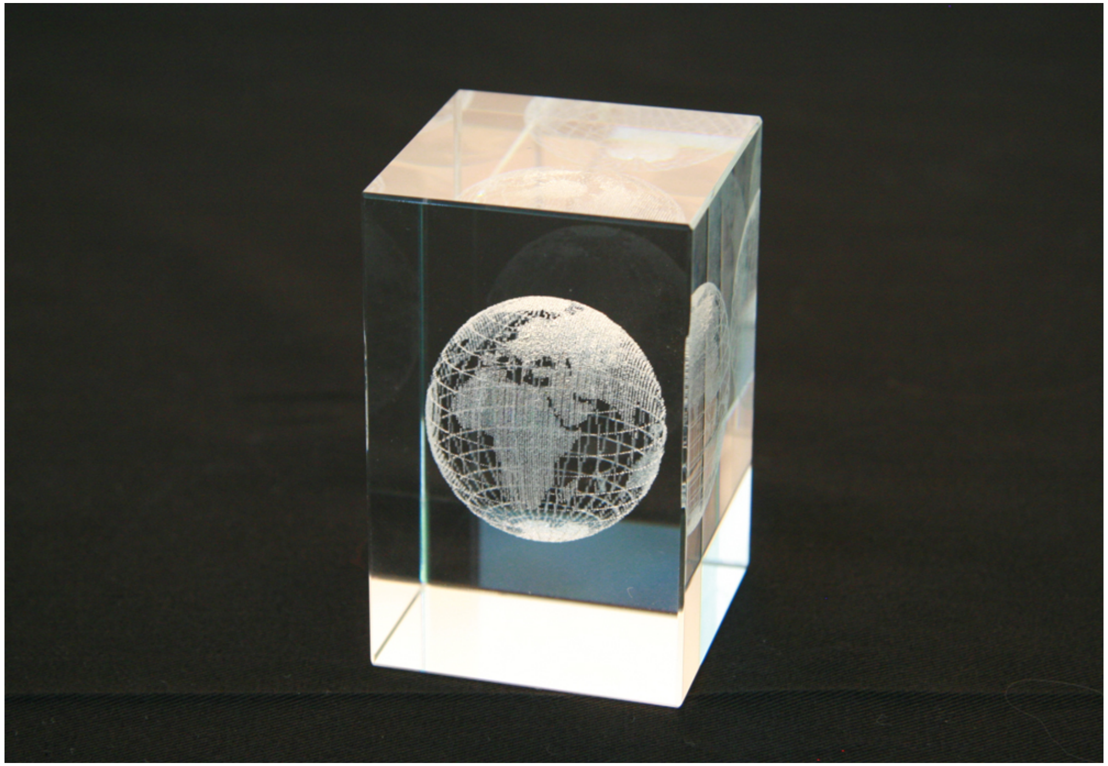
Laser frequency：1.55eV：
lower than direct bandgap
2.4eV(LDA)



We simultaneously calculate electron dynamics at macro-grid points (~200).

Initial process of nonthermal laser processing of transparent materials

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



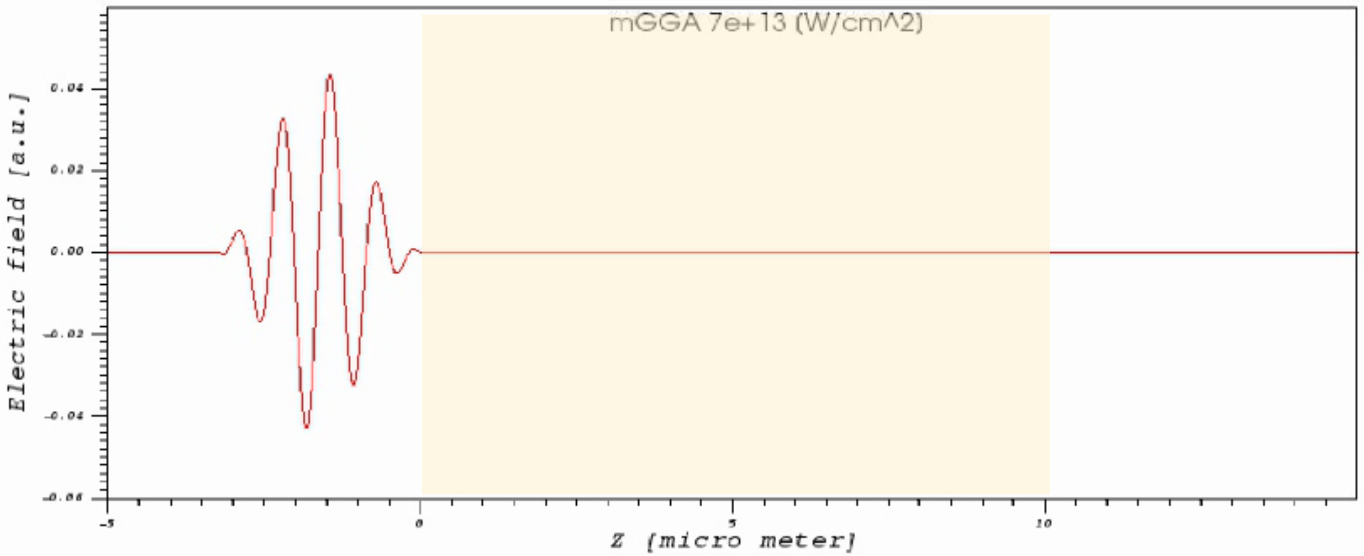
Maxwell + TDDFT multiscale simulation : 10 mm SiO_2

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

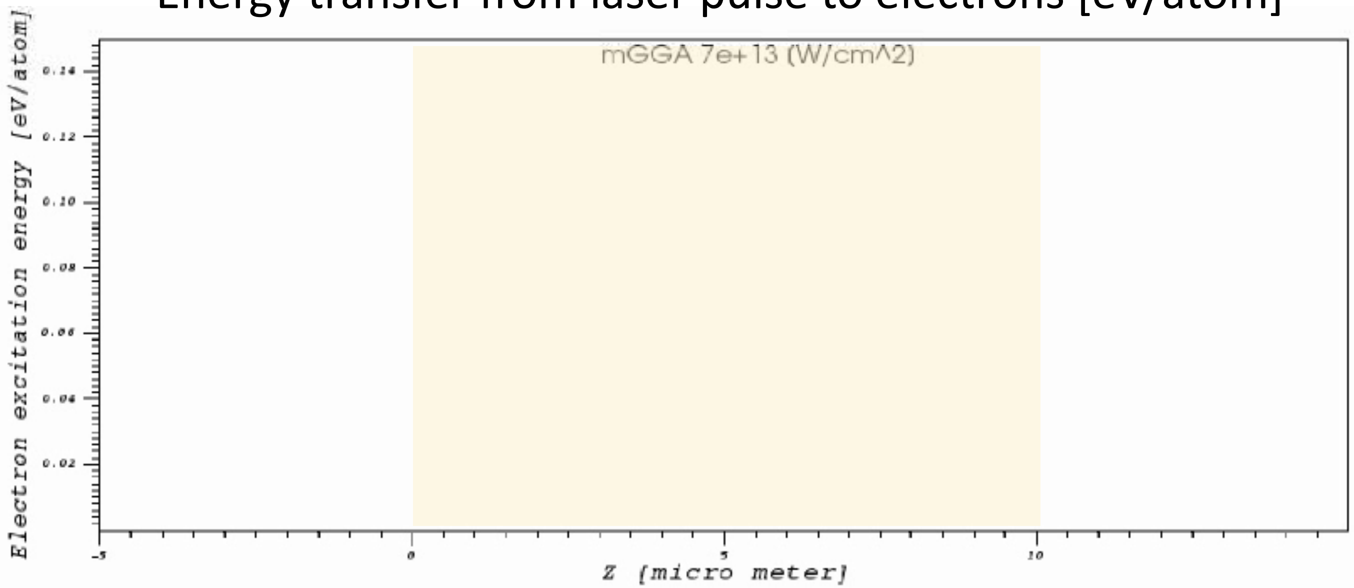
$\hbar\omega = 1.55\text{eV}$

$\lambda = 800\text{nm}$

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



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

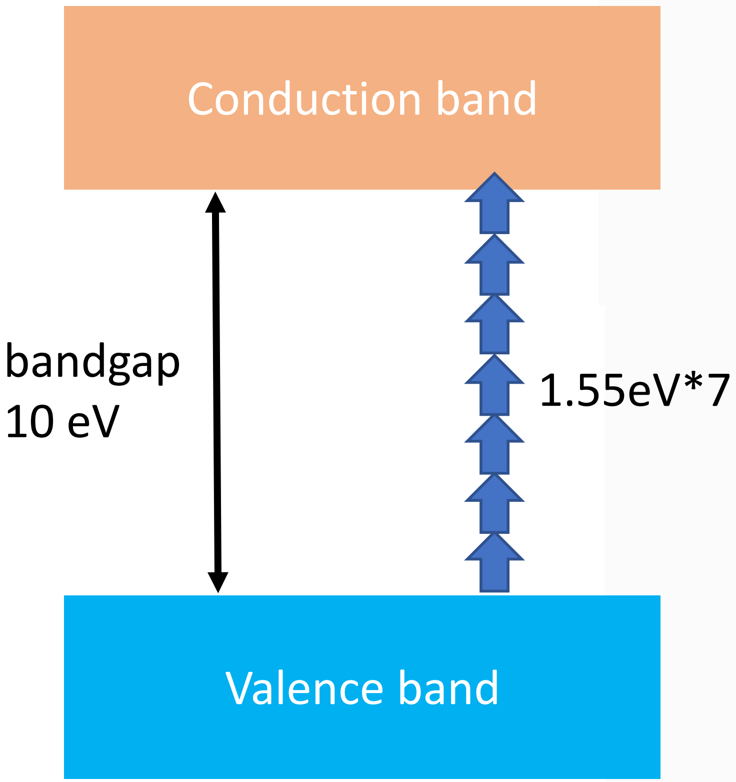


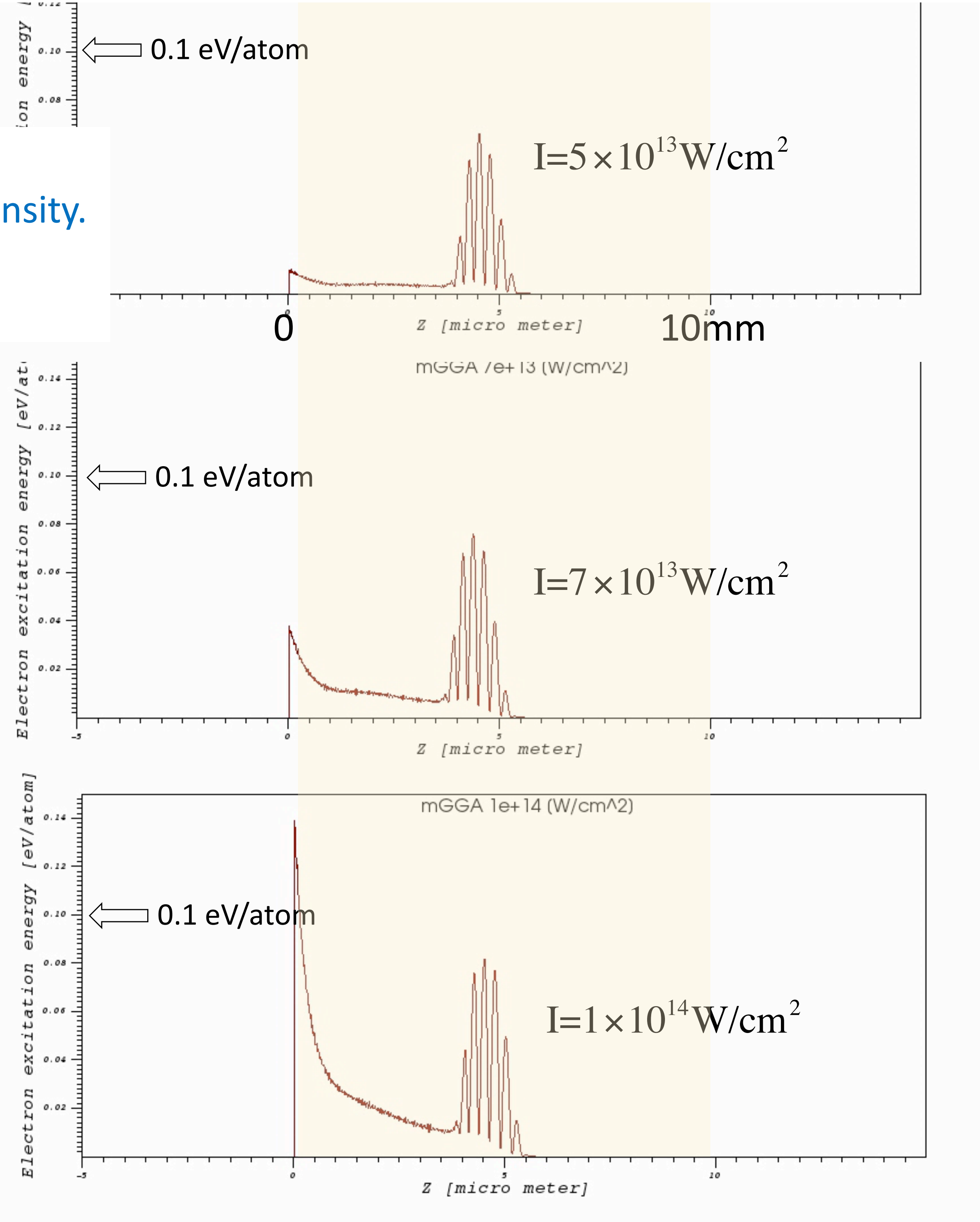
80,000 cores, 20 hours
at K-Computer, Kobe

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

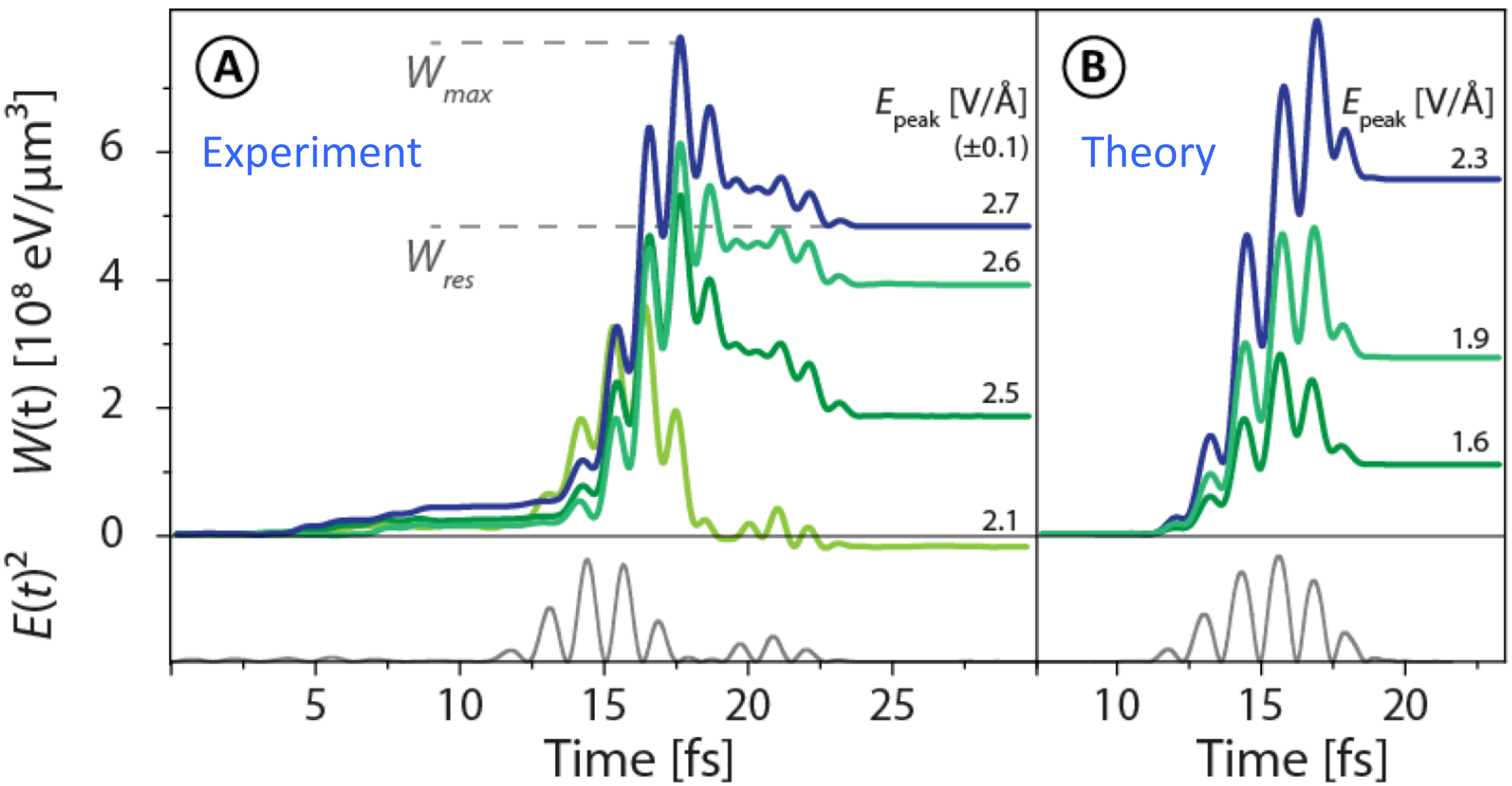
>> Damage threshold

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

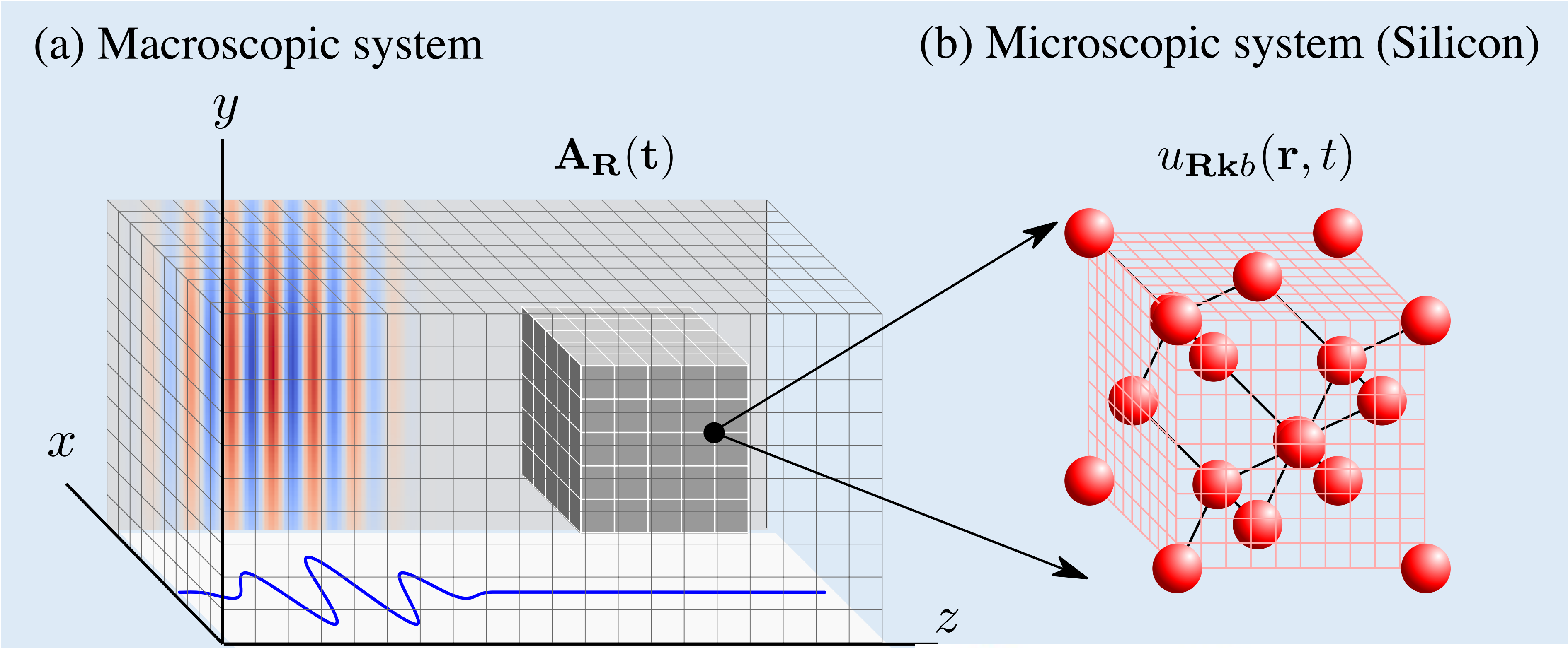
Oakforest-PACS

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

8208 nodes x 68 cores (Intel Xeon Phi 7250)

25 PFLOPS

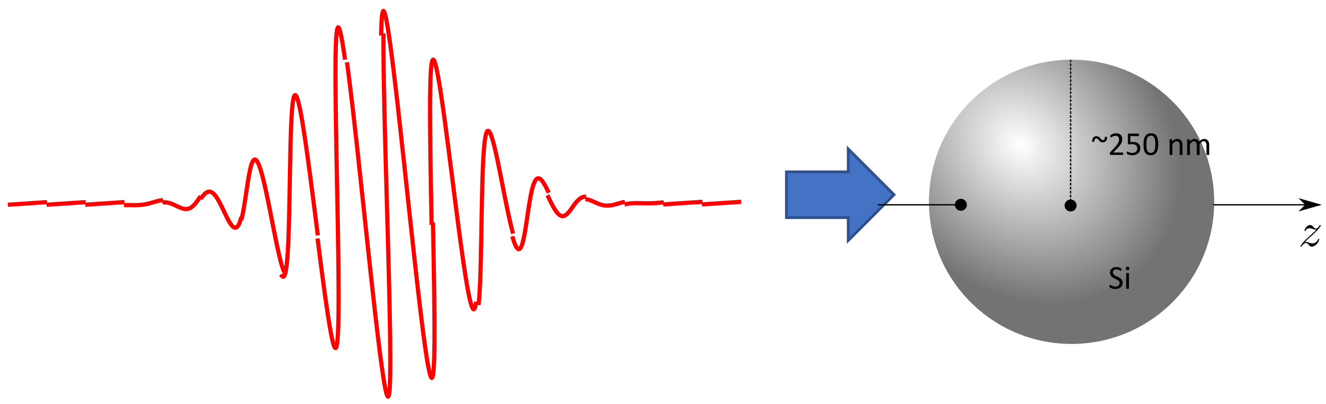
(Almost) full node calculation achieved on 2017.3.



$$-\nabla_{\mathbf{R}} \times \nabla_{\mathbf{R}} \times \mathbf{A}_{\mathbf{R}}(t) + \frac{1}{c^2}\frac{\partial^2}{\partial t^2}\mathbf{A}_{\mathbf{R}}(t) = \frac{1}{4\pi}\mathbf{J}_{\mathbf{R}}(t)$$

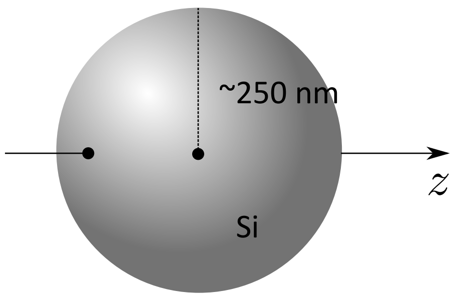
$$i\frac{\partial}{\partial t}u_{n\mathbf{k},\mathbf{R}}(\mathbf{r},t) = \hat{h}^{\mathrm{KS}}_{\mathbf{k},\mathbf{R}}(\mathbf{r},t;\mathbf{A}_{\mathbf{R}}(t))u_{n\mathbf{k},\mathbf{R}}(\mathbf{r},t)$$

Strong Laser Pulses on Silicon Nano-Sphere



Silicon nano-sphere of R ~ 250nm

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

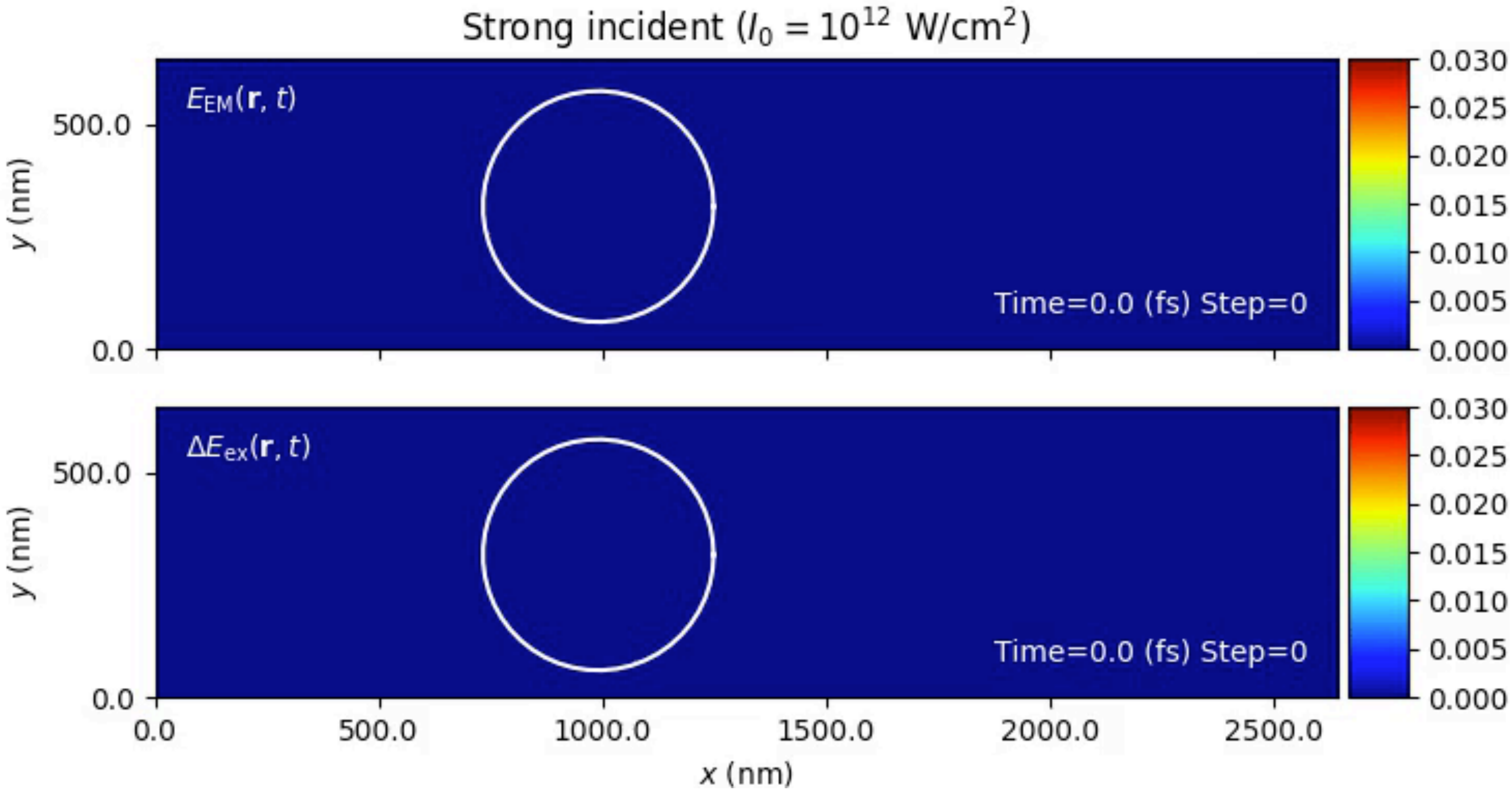


- 3D Maxwell + 3D TDDFT multiscale calculation
- 2.5 hourse by Oakforest-PACS 8188 nodes

Energy density of EM fields

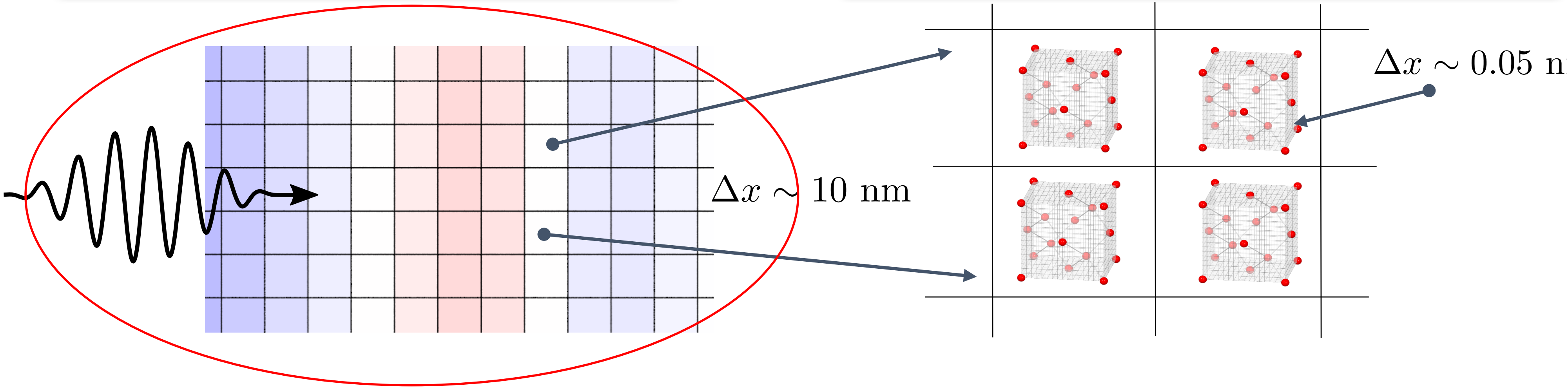
$$E^2 + B^2$$

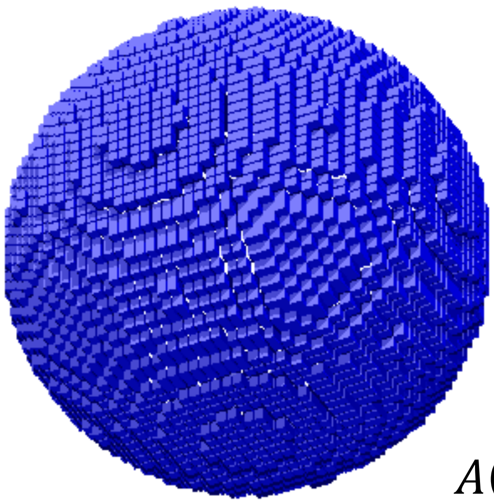
Electronic excitation energy (two photon absorption)



Macroscopic system (EM field)

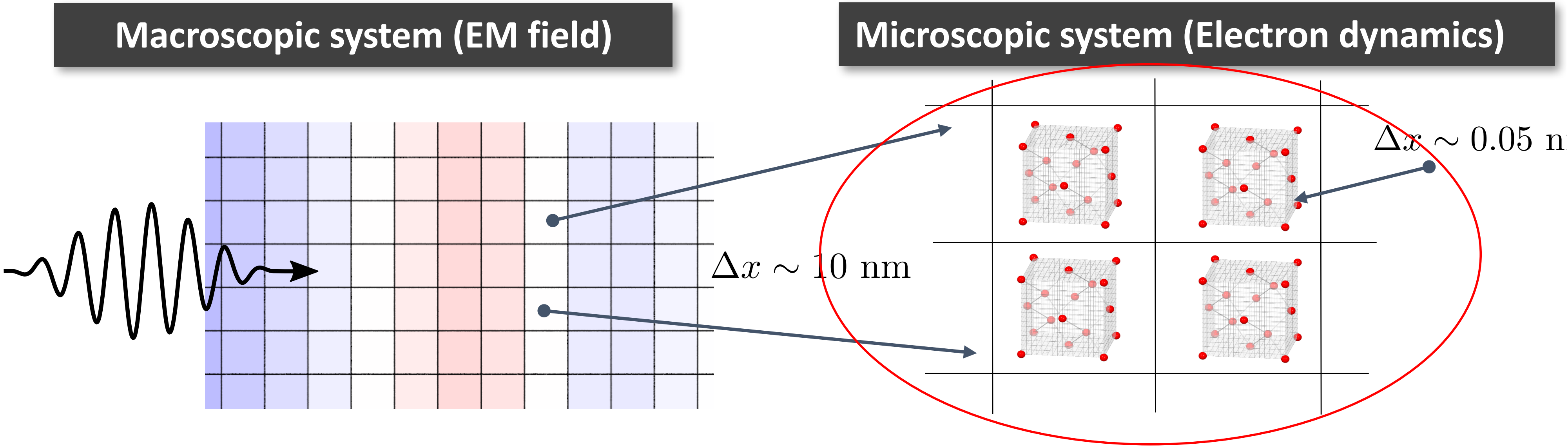
Microscopic system (Electron dynamics)





$A(X, t)$

Silicon nano-sphere is expressed by 32,752 macroscopic grid points.
Each node of Oakforest-PACS treats electron dynamics of 4 macro-points.
32,752 = 8,188 x 4



In one node, electron dynamics of 4 macro-points.

Each electron dynamics calculation uses

$$u_{X,nk}(r,t)$$

- Time step (t): 20,000
- Macro-grid points (X): 32,752
- band index (n): 16
- k-points (k): 8 x 8 x 8
- Micro-grid points (r): 16 x 16 x 16

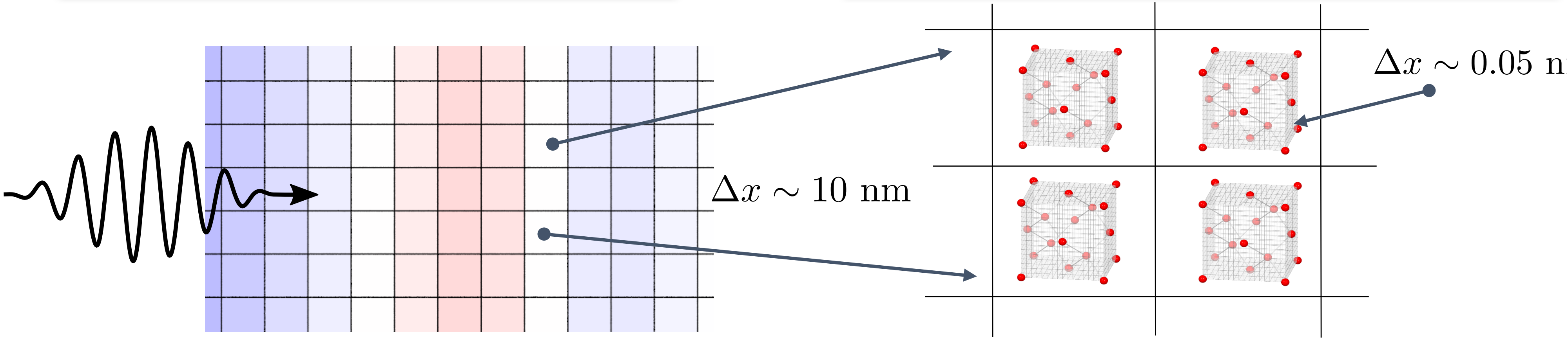
MPI parallelization = # of nodes (8188)

Other loops by OpenMP
- X=1,4 (macro grid)
- n=1,16 (band index)
- k=1,8^3 (k-points)
- r=1,16^3 (micro-grid)

Numerical aspects of Multiscale Maxwell-TDDFT calculation (Silicon case)

Macroscopic system (EM field)

Microscopic system (Electron dynamics)



$$-\nabla_{\mathbf{R}}\times\nabla_{\mathbf{R}}\times\mathbf{A}_{\mathbf{R}}(t)+\frac{1}{c^2}\frac{\partial^2}{\partial t^2}\mathbf{A}_{\mathbf{R}}(t)=\frac{1}{4\pi}\mathbf{J}_{\mathbf{R}}(t)$$

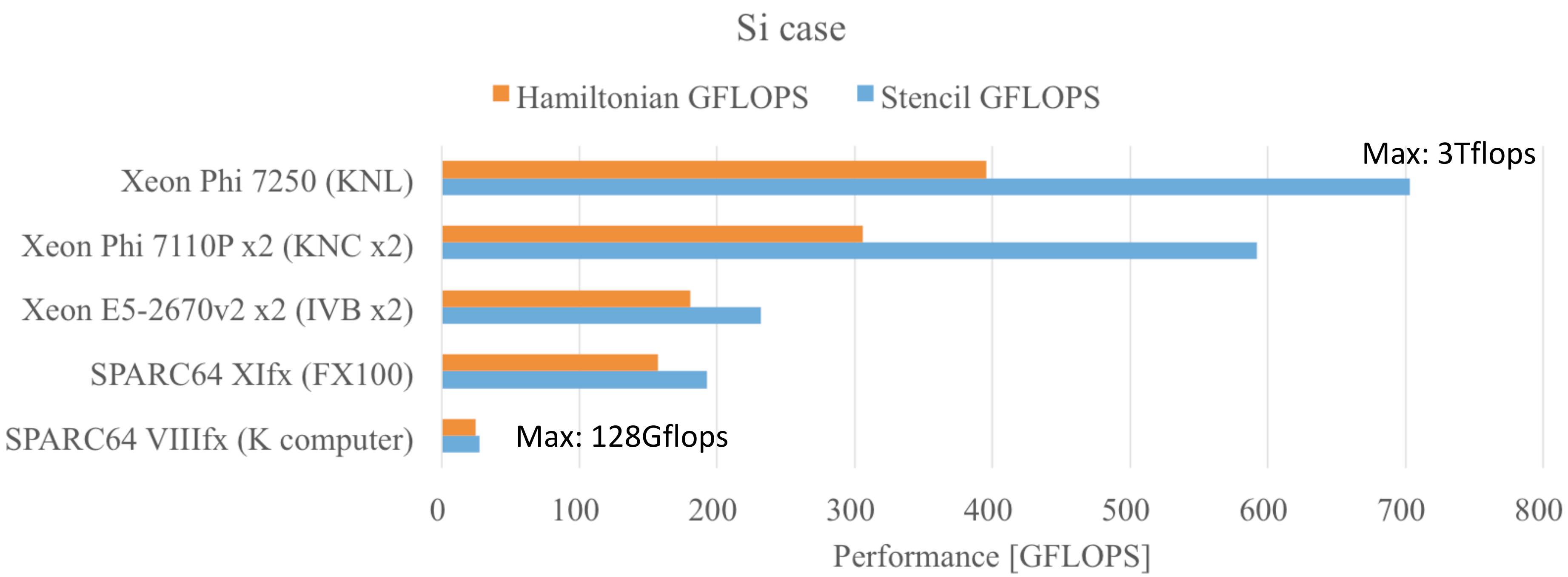
$$i\frac{\partial}{\partial t}u_{n\mathbf{k},\mathbf{R}}(\mathbf{r},t)=\hat{h}^{\mathrm{KS}}_{\mathbf{k},\mathbf{R}}(\mathbf{r},t;\mathbf{A}_{\mathbf{R}}(t))u_{n\mathbf{k},\mathbf{R}}(\mathbf{r},t)$$

EM – QM(electron dynamics) systems are connected only through $A_R(t)$ and $J_R(t)$.

Very small communication costs between nodes.

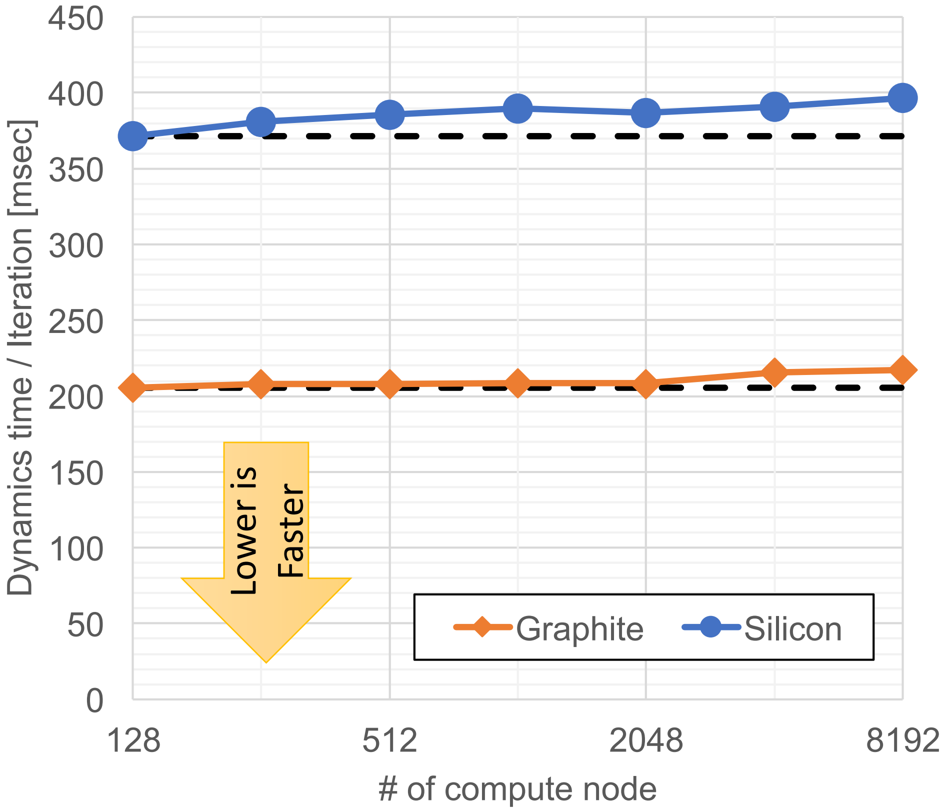
Performance in various processors

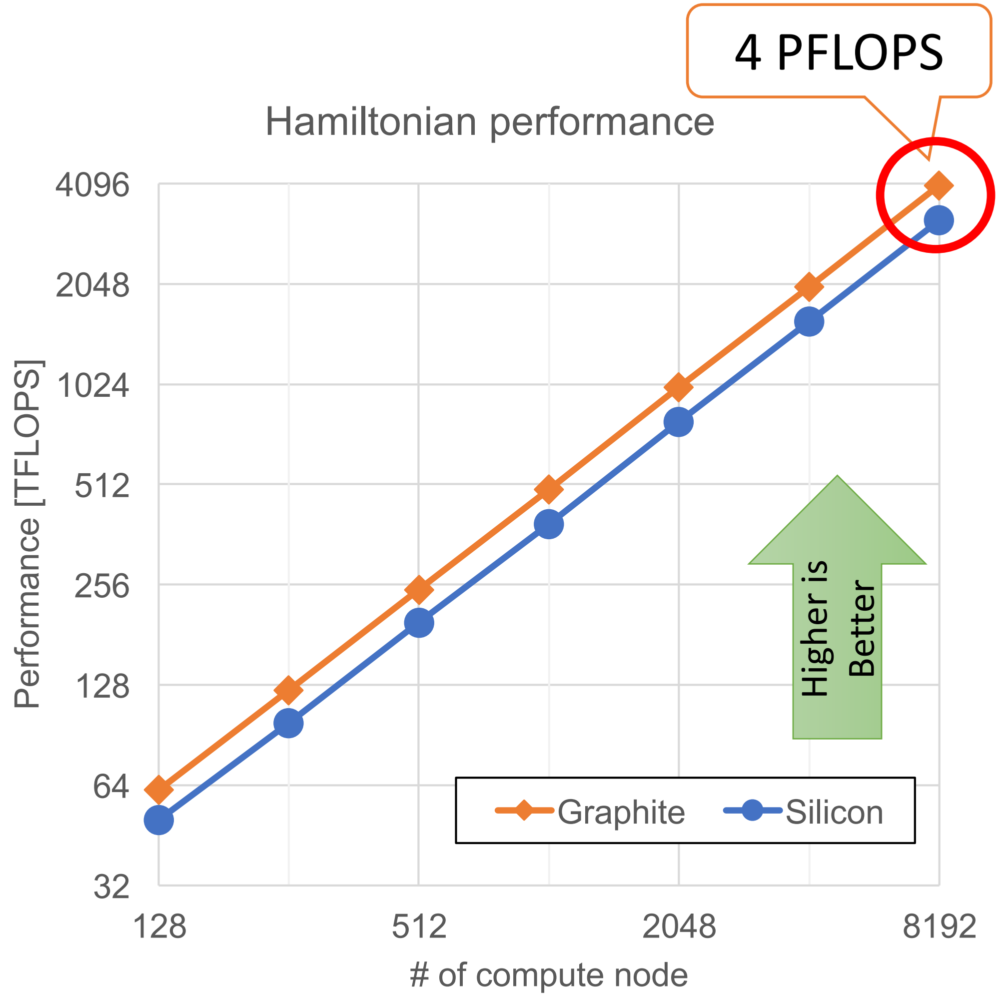
In-House collaboration with Computer Science group
(Prof. Boku, Ph.D student Hirokawa)



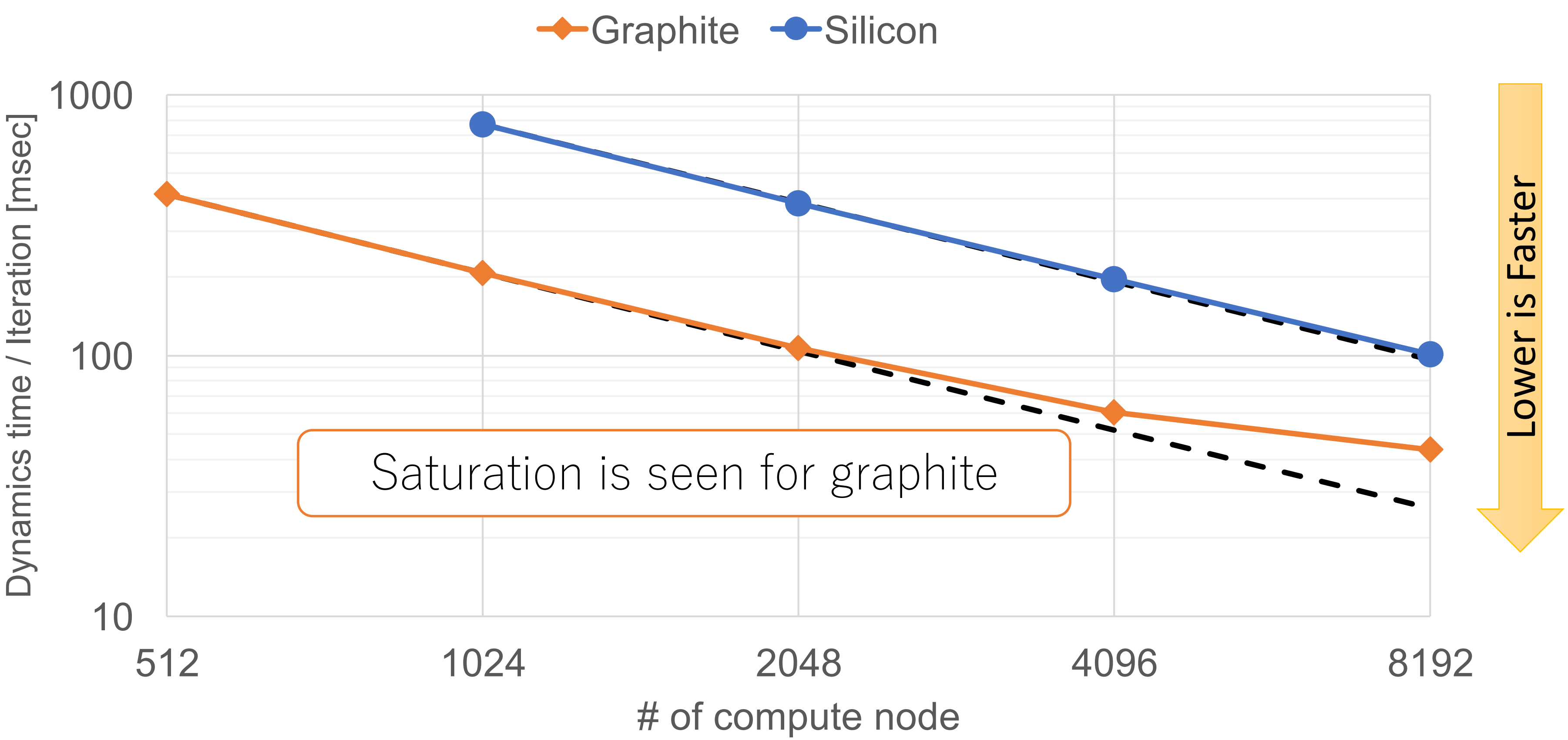
Weak scaling

12% of theoretical peak for silicon,
16% for graphite.





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

SALMON

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

Financial supports

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

K computer

JCAHPC