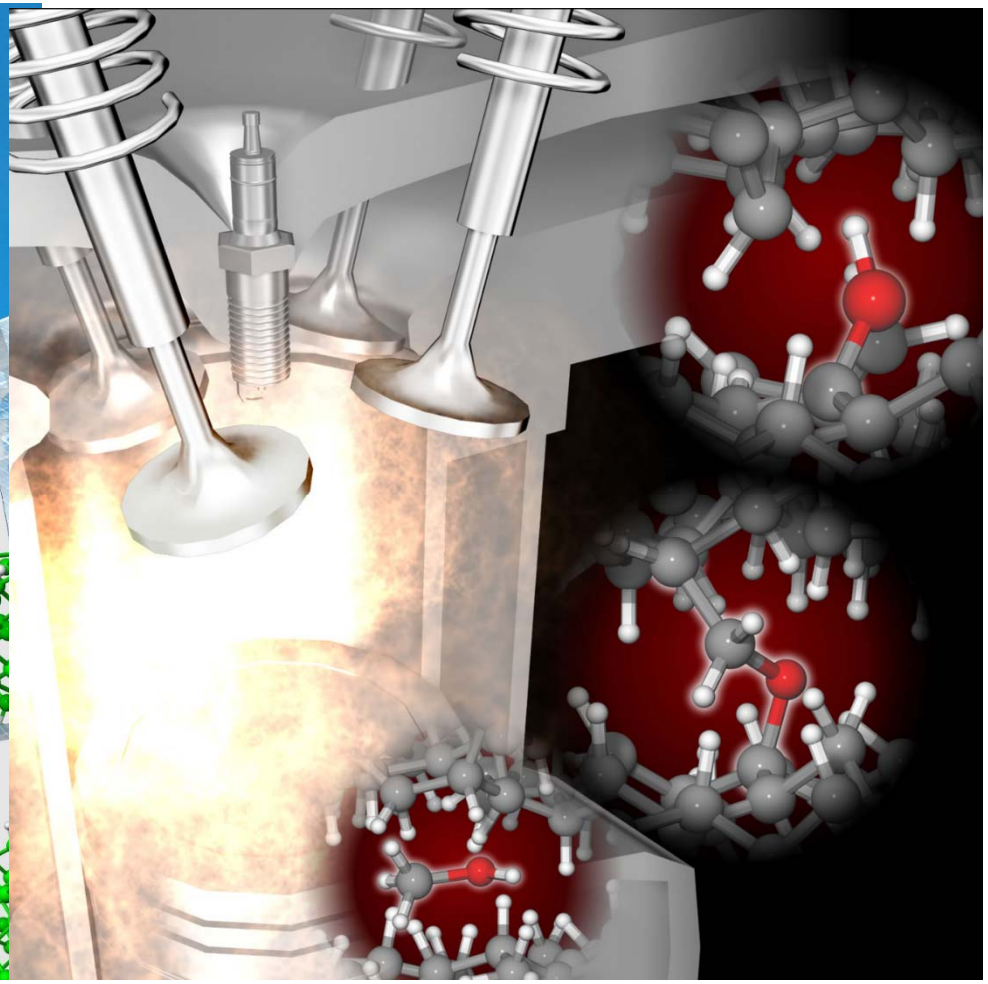
Multi-Physics Simulation for Material Design by Supercomputer

Institute for Materials Research, Tohoku University

Momoji Kubo

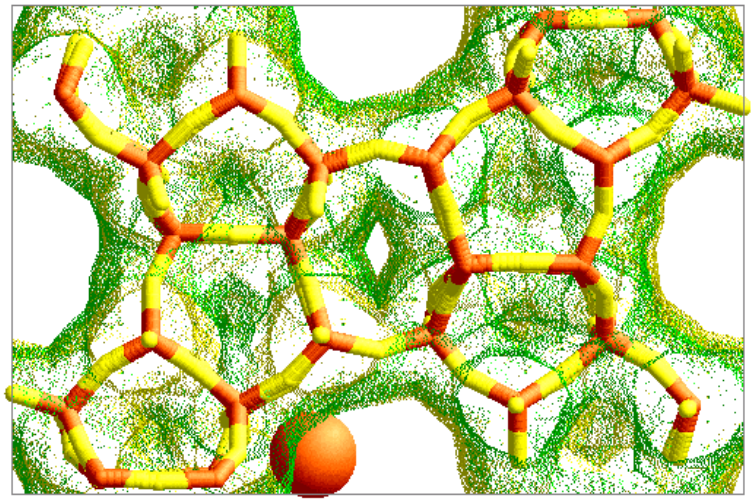


Momoji Kubo (Tohoku University)

Graduated from Hydrocarbon Chemistry Department

Associate Professor at Applied Chemistry Department

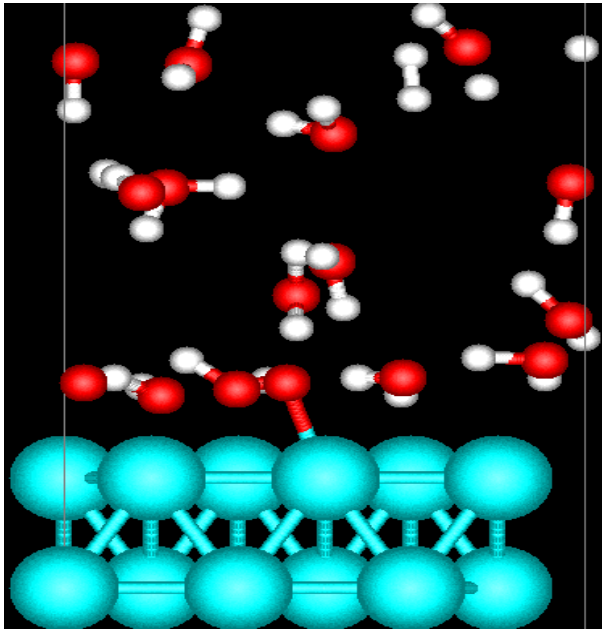
Zeolite Catalysts



(Electronic States of Zeolite)

Quantum Chemistry for Catalytic Activity of Zeolites

Fuel Cell Catalysts

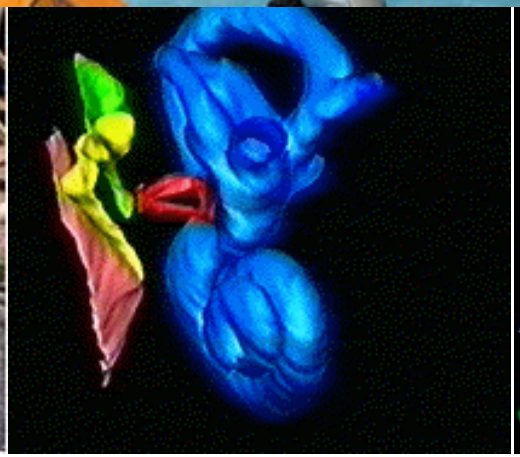


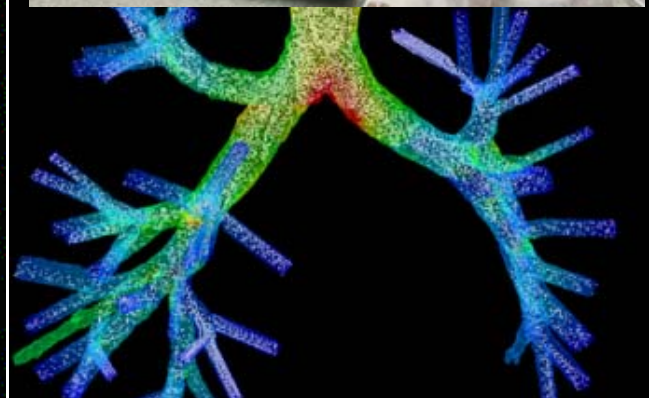
(Chemical Reaction Dynamics on Pt)

Quantum Chemical Molecular Dynamics

Professor at Department of Robotics from 2008

How does Quantum Chemistry Contribute to Robotics ?





Strategy of Kubo Laboratory

Design of Chemical-Reaction-Controlled Mechanical System

Massive Mechanical System

Advancement of Nanotechnology

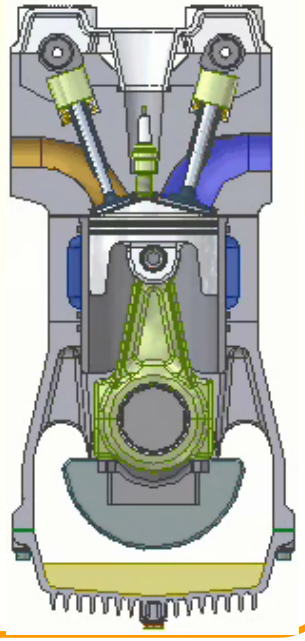
Paradigm Shift in Mechanical Engineering is essential

- ○Mechano-Chemical Reaction in Engine
- ○Chemical Wear in Robot Joint
- ○Chemical Mechanical Polishing

Chemical Reaction on Atomic Scale

Significant Influence

Mechanical Performance on Macro Scale



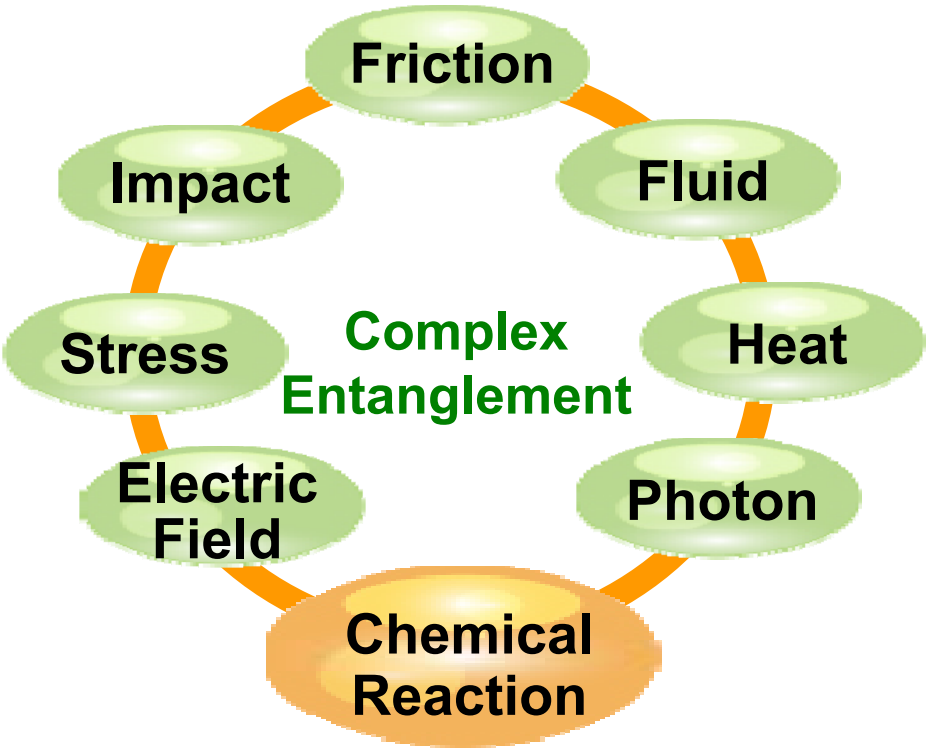
Strategy of Kubo Laboratory

Design of Chemical-Reaction-Controlled Mechanical System

Multi-Physics Process

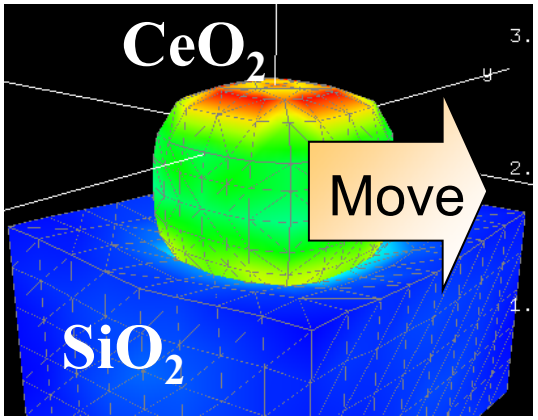
Massive Mechanical System

Its Function is Generated by "Motion"



Traditional Approach
- Finite Element Method

(1) Mechanical Polishing of Glasses

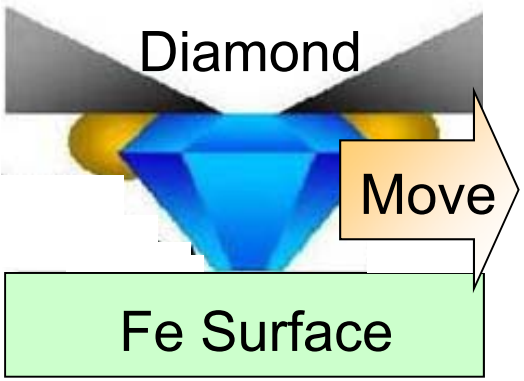


Soft CeO_2

Hard SiO_2 is polished.

Cannot Explain

(2) Mechanical Polishing of Metal

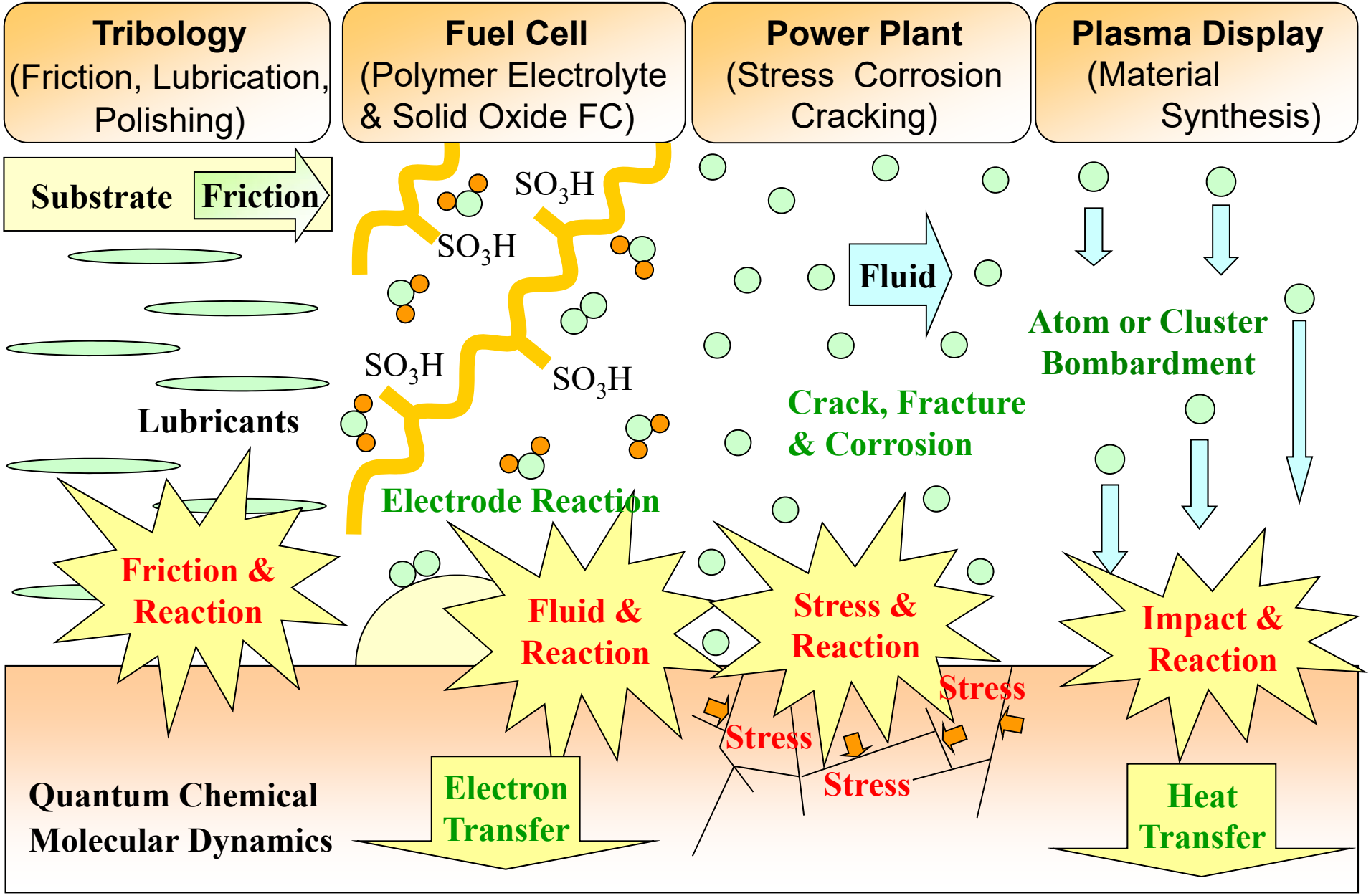


Hard Diamond

Soft Fe is not polished.

Cannot Explain

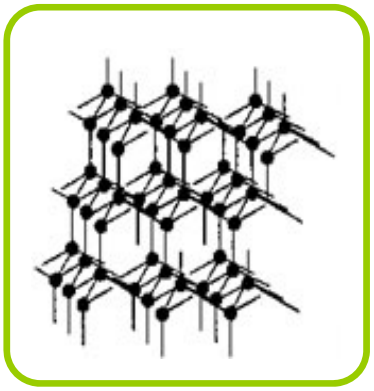
Development of Multi-Physics Simulator
Based on Quantum Chemical Molecular Dynamics



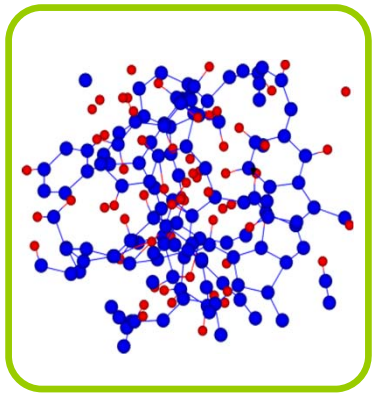
Diamond-Like Carbon (DLC)
for Super-Low Friction

Kubo Laboratory
Multi-Physics Computational Science

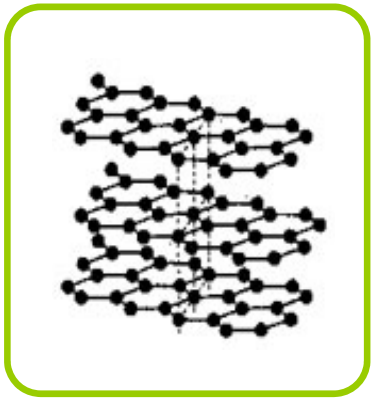
Diamond (sp3)



DLC



Graphite (sp2)



Diamond Like Carbon

- High strength
- Low friction
- Chemical stability
- Smoothness

Low Friction

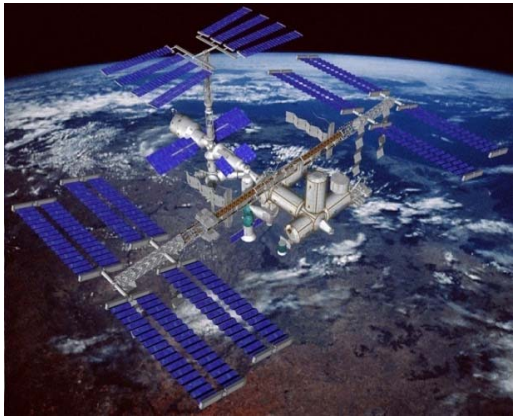
Engine

Hard Disk Drive

Solid Lubrication

MEMS

Use at Low Temperature

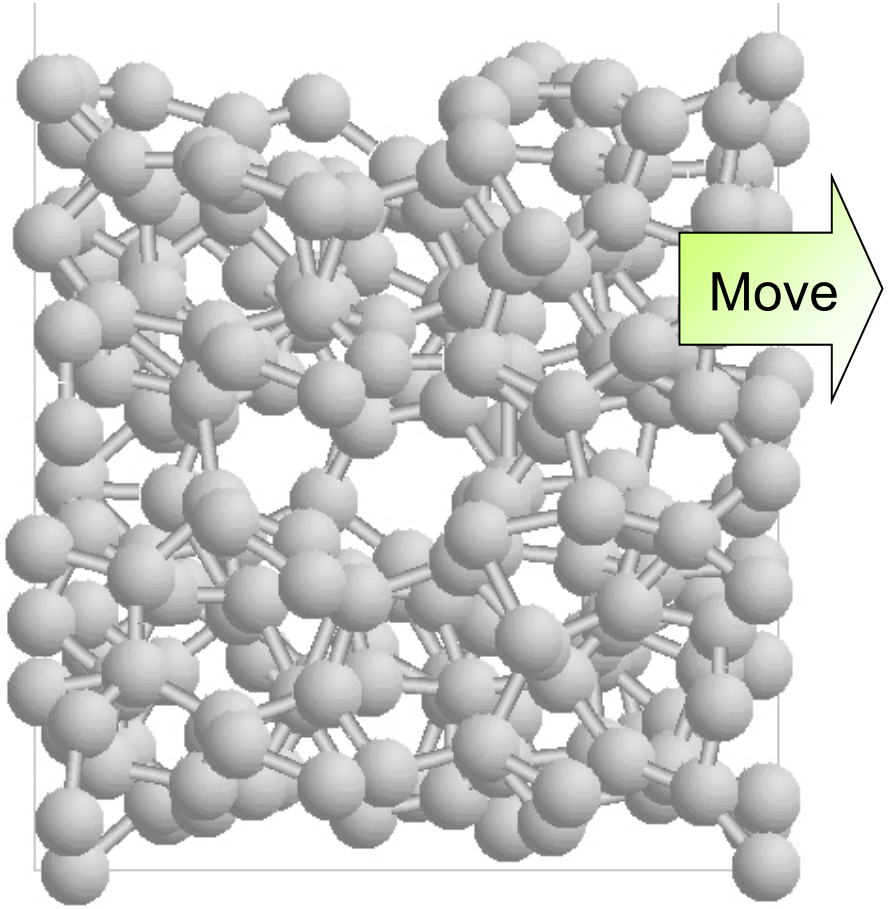


Space Machines

Tight-Binding Quantum Chemical Molecular Dynamics
Friction Simulation of Diamond-Like Carbon

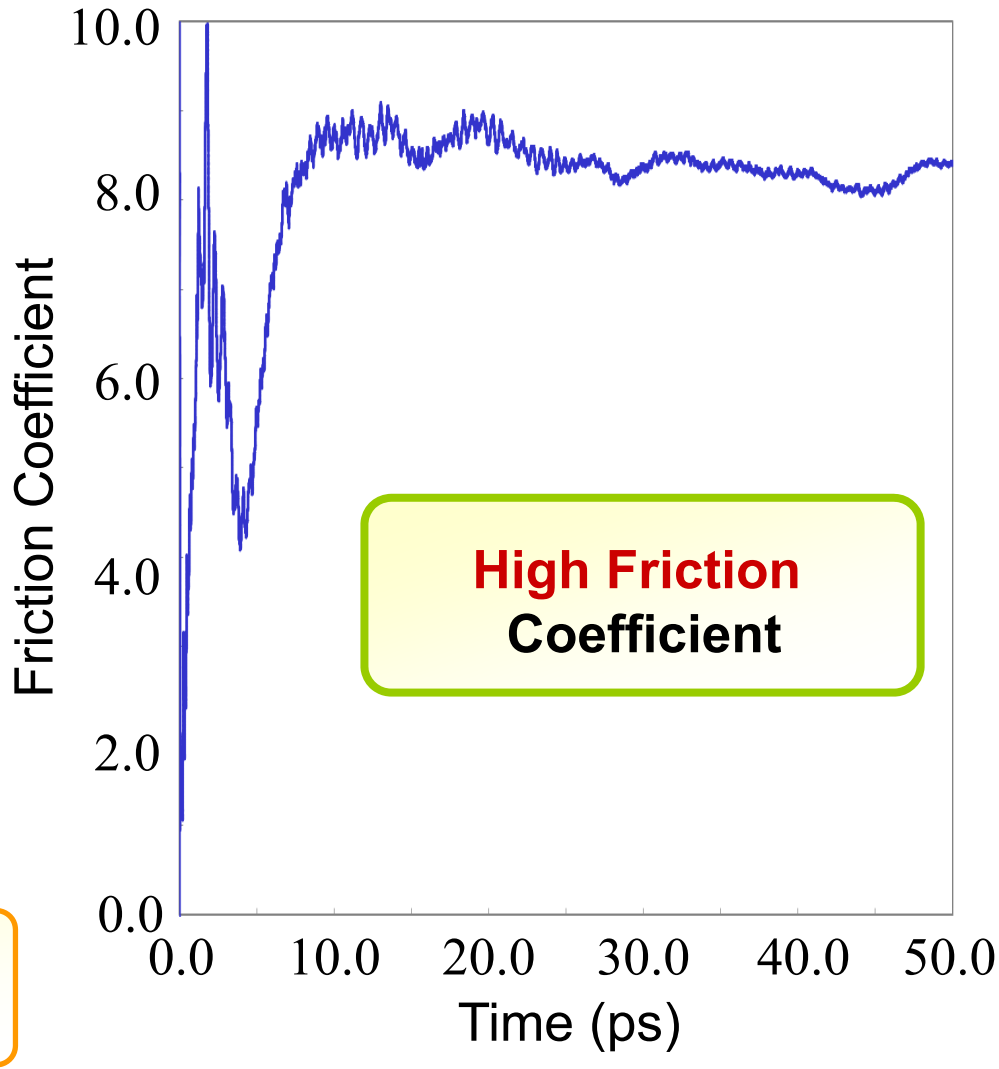
Kubo Laboratory
Multi-Physics Computational Science

Diamond-like Carbon Film

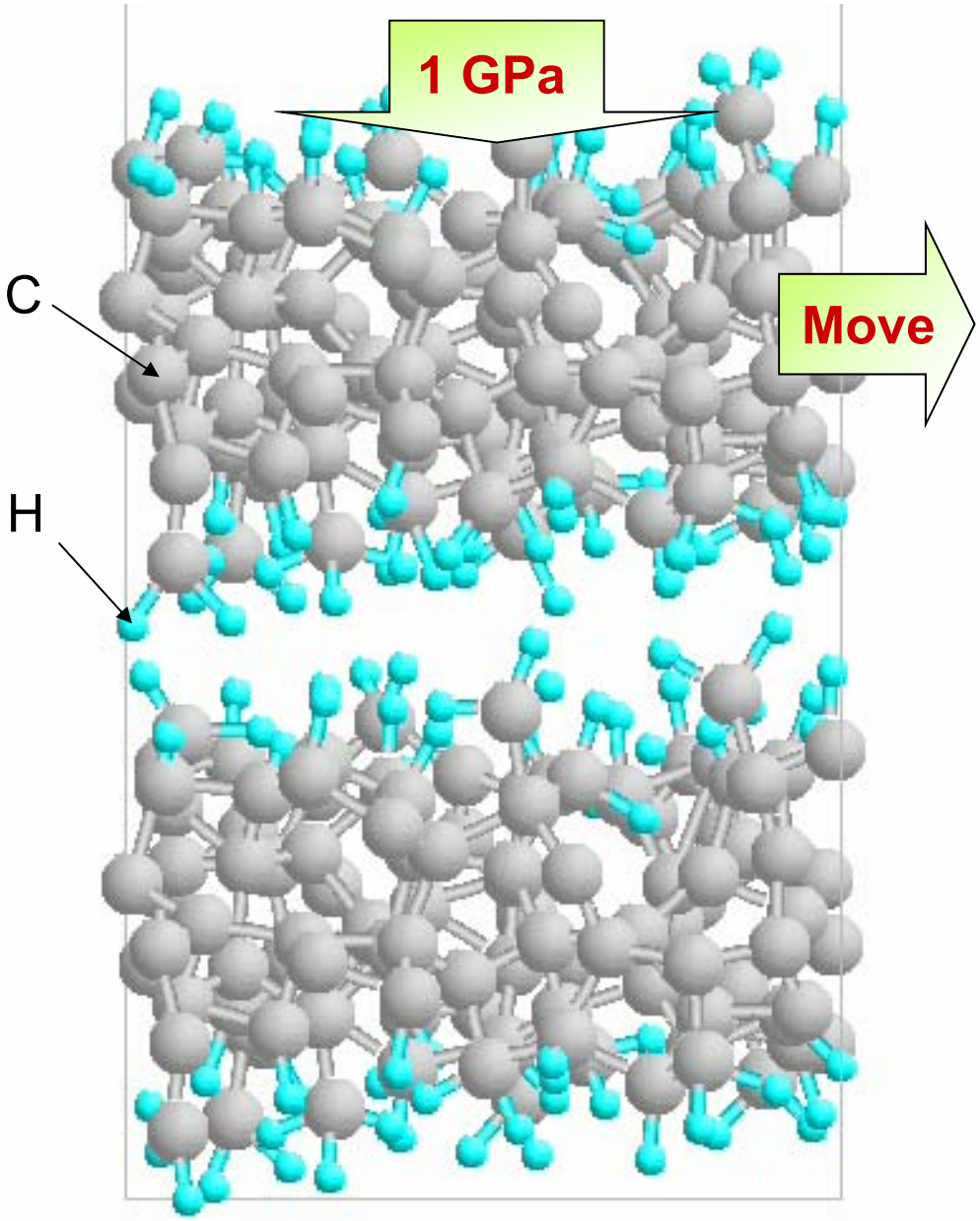


Repeated Generation and Dissociation of C-C Bonds

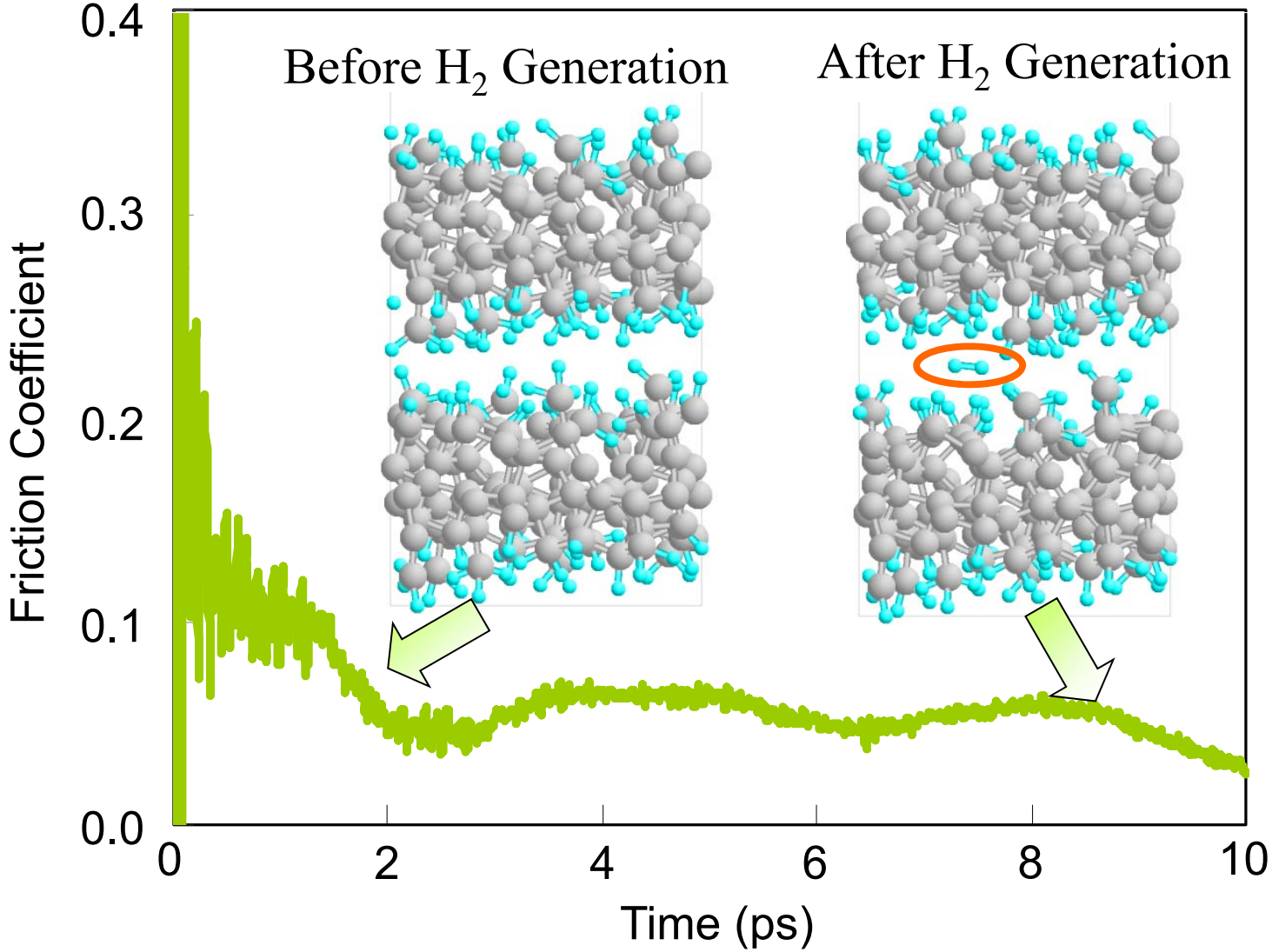
Friction Coefficient by Simulation



Tight-Binding Quantum Chemical
Molecular Dynamics Simulation on DLC

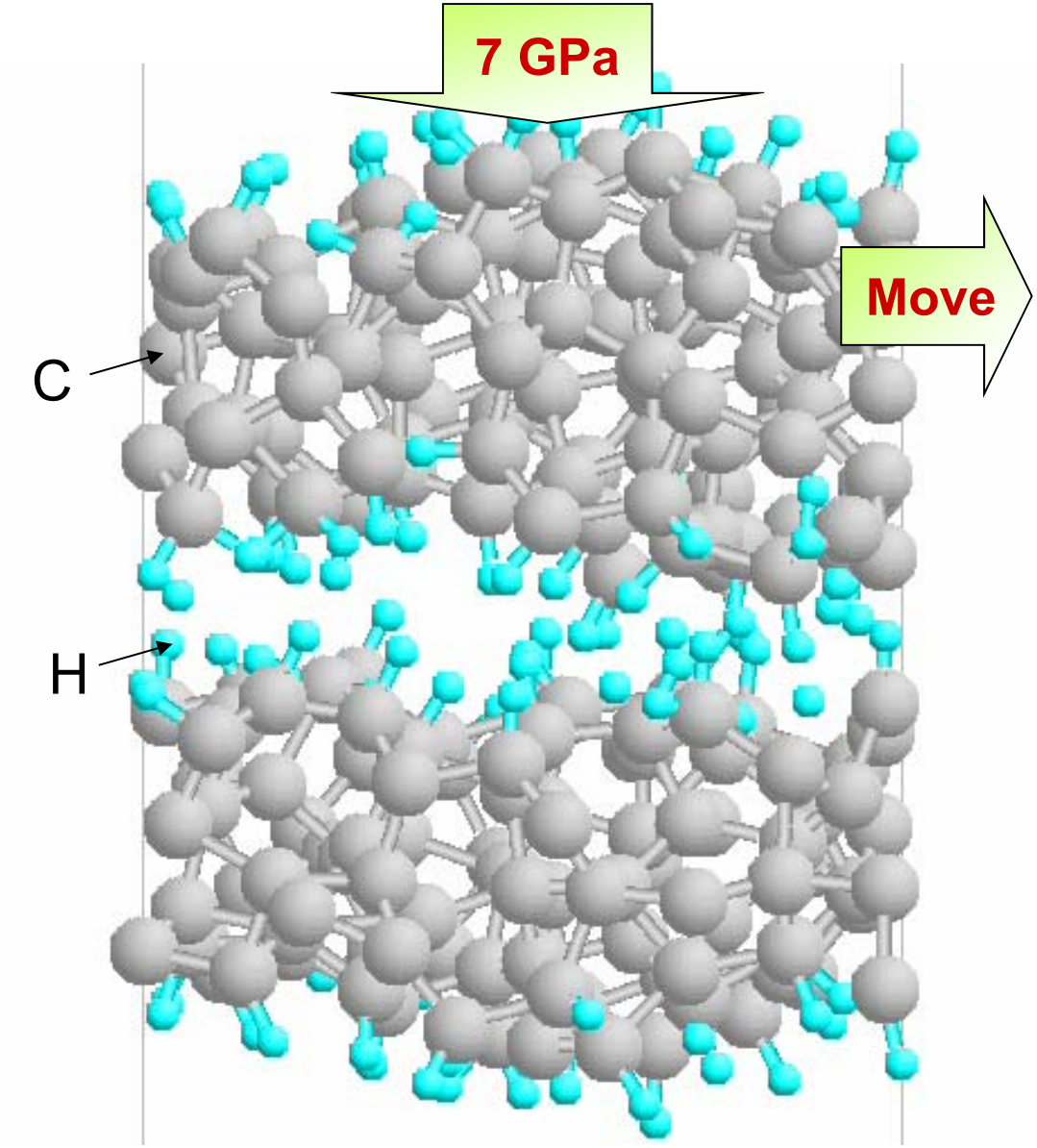


Friction Coefficient of Diamond-Like Carbon under 1 GPa



H_2 Generation Reaction ⇨ **Decrement of Friction Coefficient**

Tribochemical Reaction Dynamics of
Diamond-Like Carbon at Severe Condition (7 GPa)



Pressure Affect Chemical Reaction

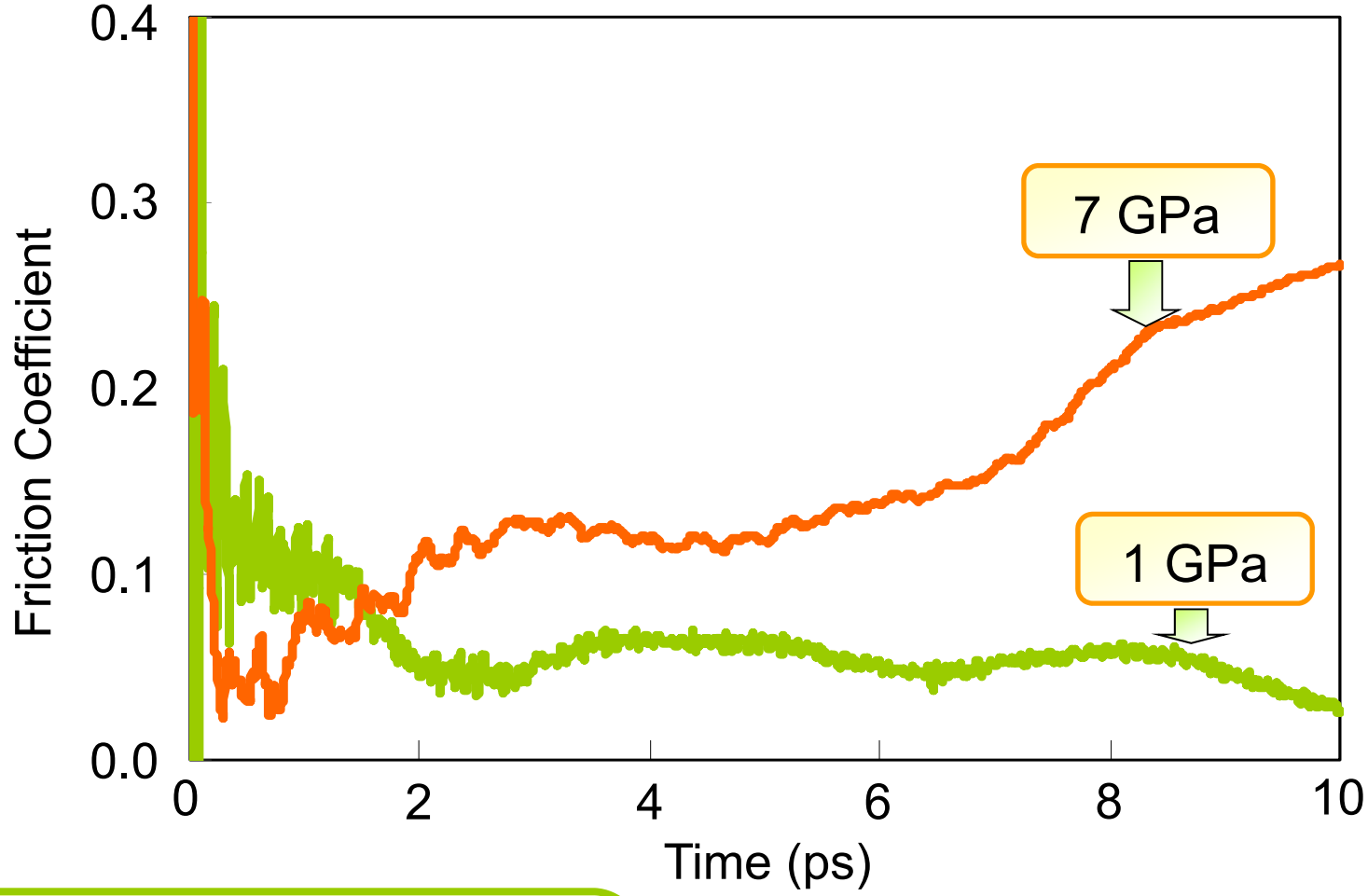
1 GPa

Generation of H_2

7 GPa

Generation of C-C Bond

Friction Coefficient of
Diamond-Like Carbon under 1 GPa and 7 GPa



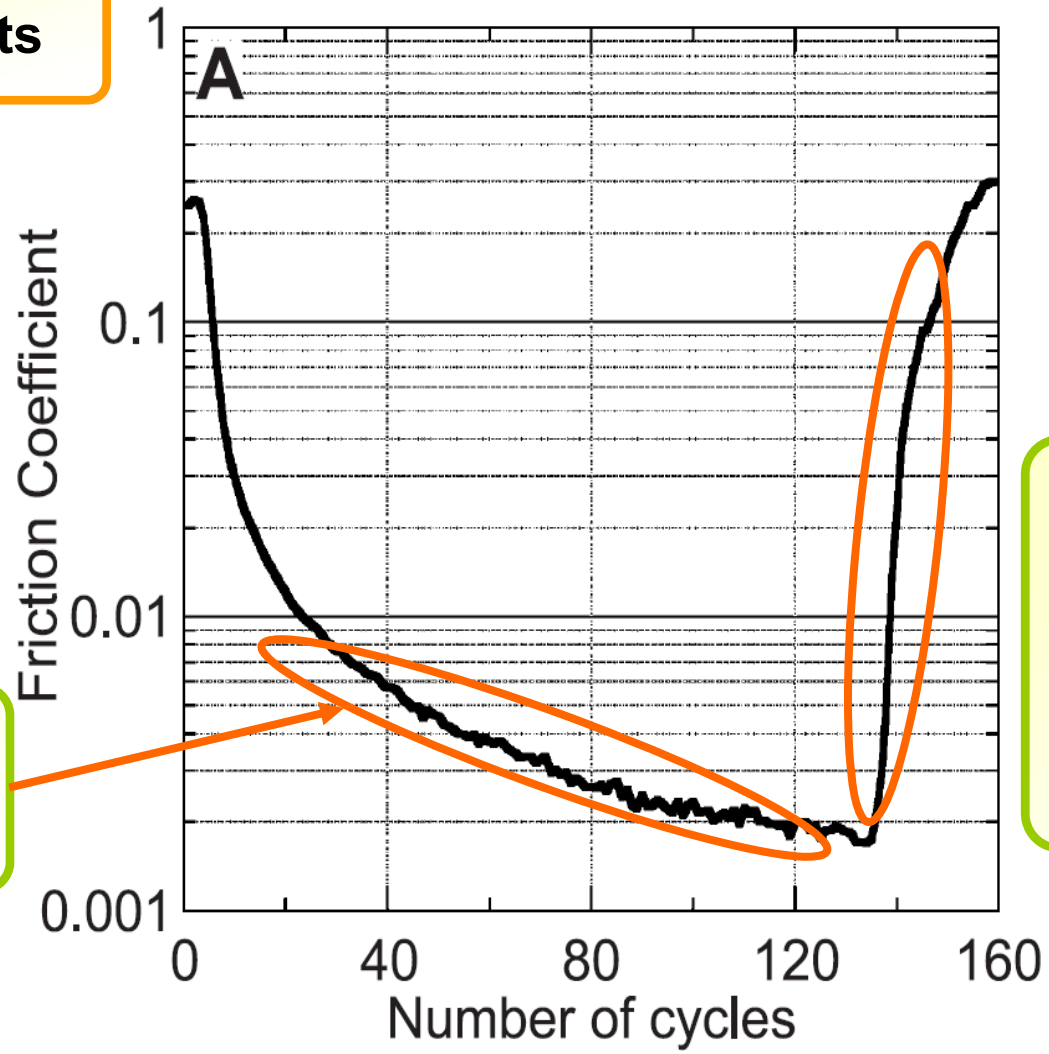
TEXT BOOK

Friction coefficient does not depend on load

Chemical reactions at interface change TEXT BOOK

Friction Property of Diamond-Like Carbon (DLC)

Experiments



Low Friction Property

Rapid Increase

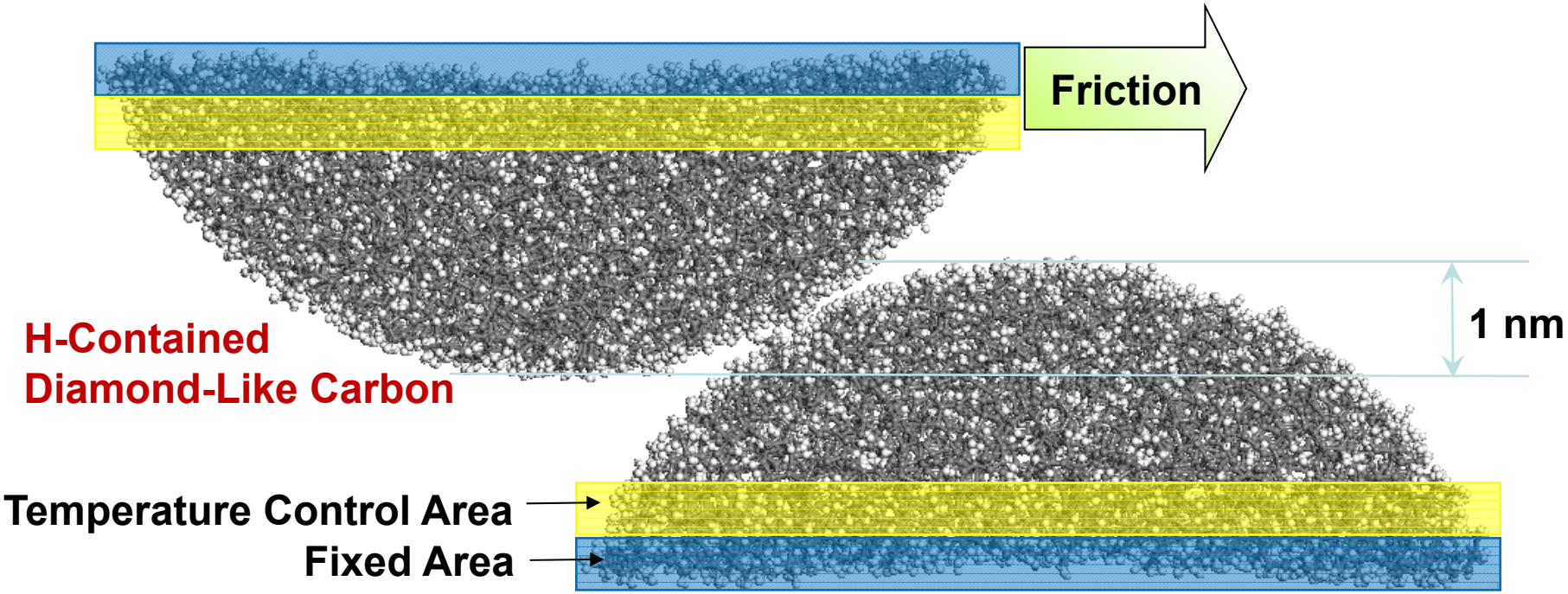
Instability of Friction Property

J.Fontaine et al., Thin Solid Films, 482, (2005) 99.

Large-Scale Molecular Dynamics Simulation for Friction of Diamond-Like Carbon

Reactive Force Field

Molecular Dynamics Simulation



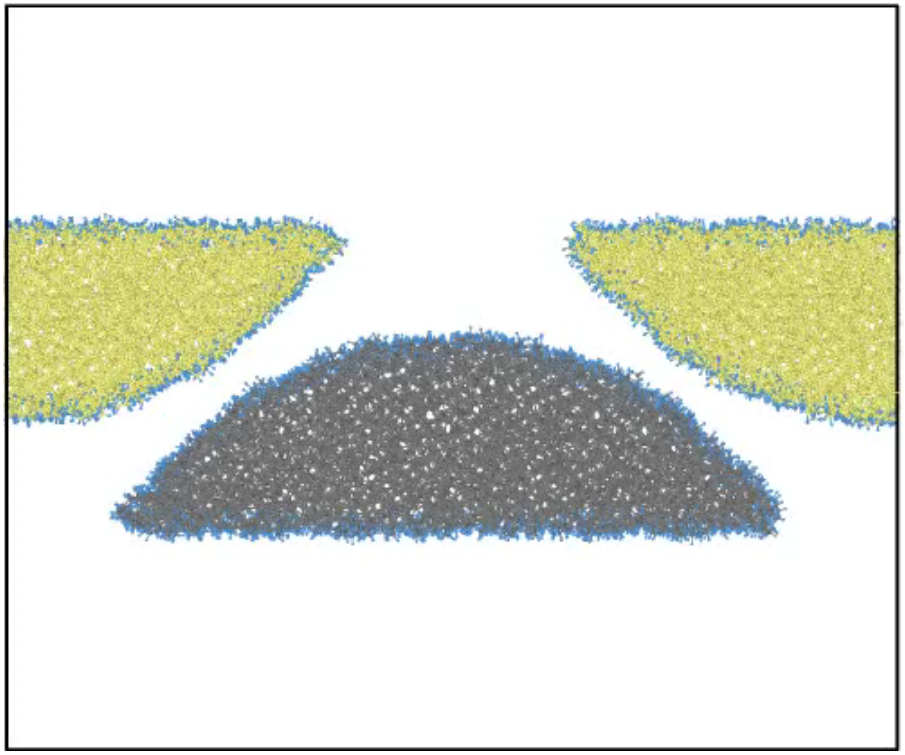
Large-Scale Calculation gives Paradigm Shift

Super-Low Friction → **Frictional Wear**

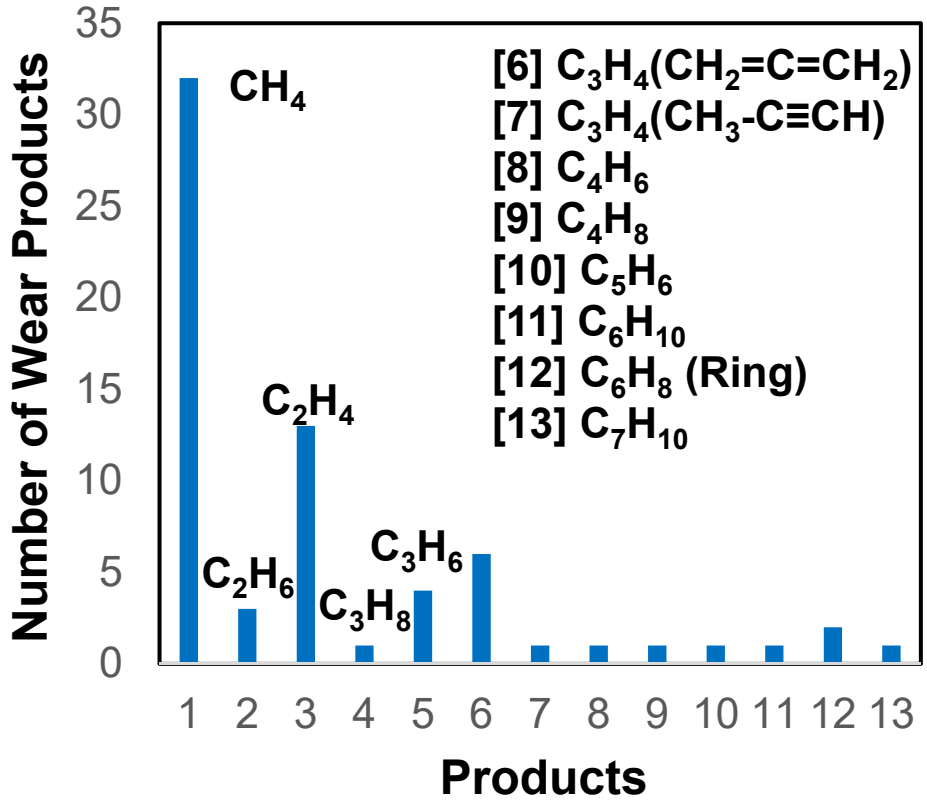
Tribo-Wear Process by Chemical Reactions on Diamond-Like Carbon

Kubo Laboratory
Multi-Physics Computational Science

Tribo-Chemical Reactions Lead to Tribo-Wear



Product Distribution by Tribo-Wear

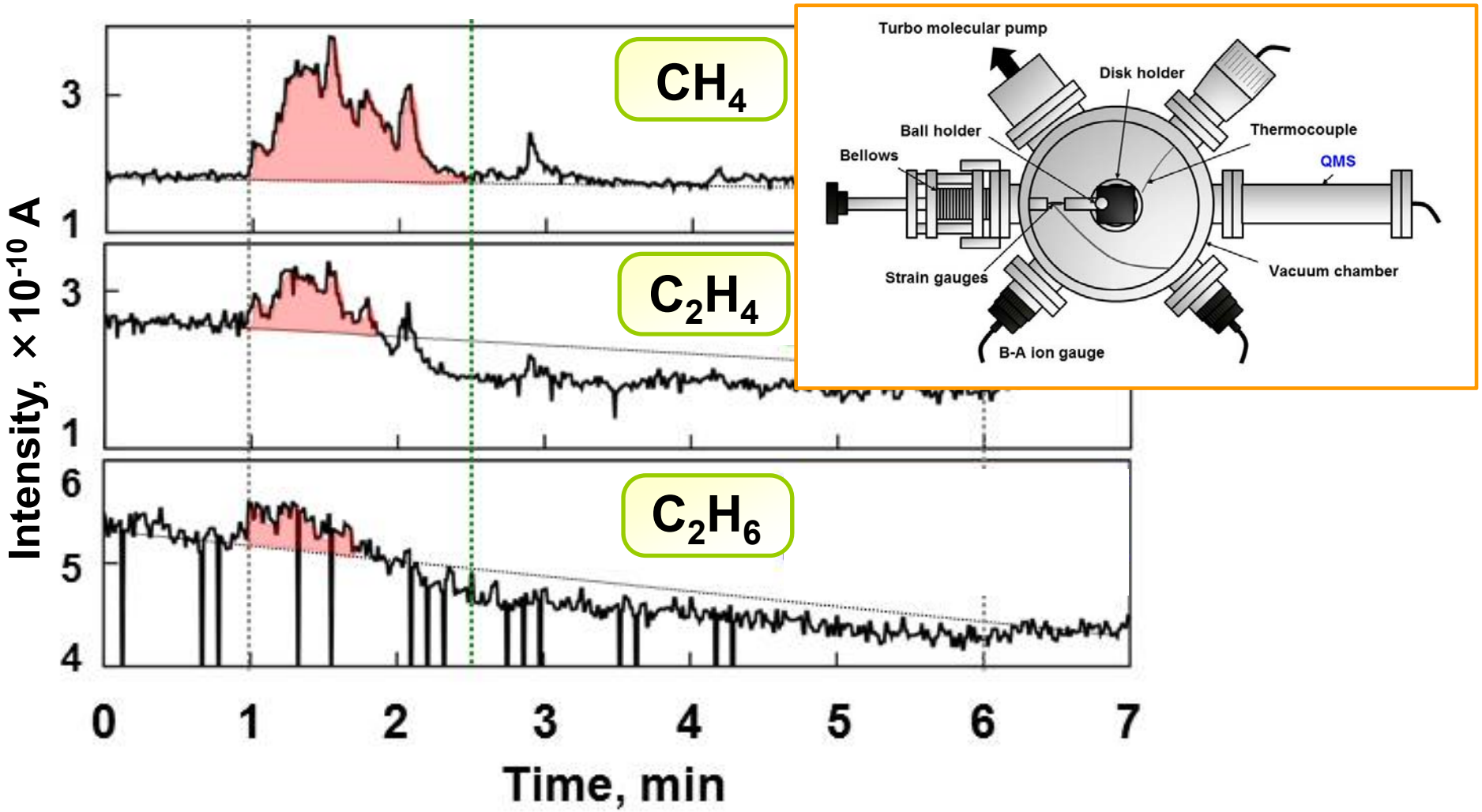


Product Distribution: CH_4 > C_2H_4 > C_2H_6

Unsaturated Hydrocarbon C_2H_4 is more than Saturated Hydrocarbon C_2H_6

Experiments by Adachi Lab. in Tohoku Univ.

Tribo-Wear of Diamond-Like Carbon — QMS Measurement in Vacuum



Product Distribution: CH_4 > C_2H_4 > C_2H_6

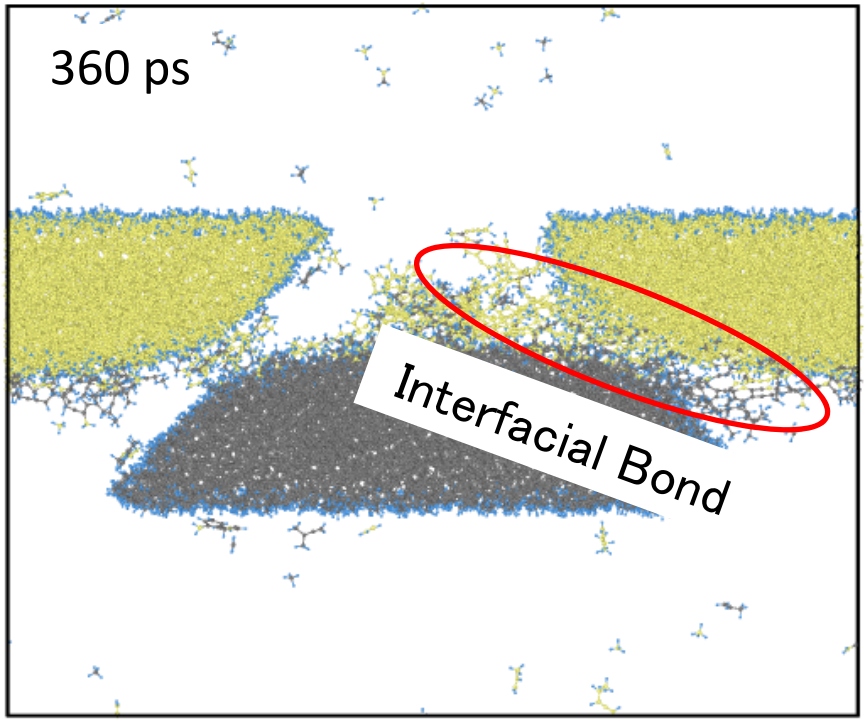
Good Agreement with Simulations

Effect of Environment can be Clarified

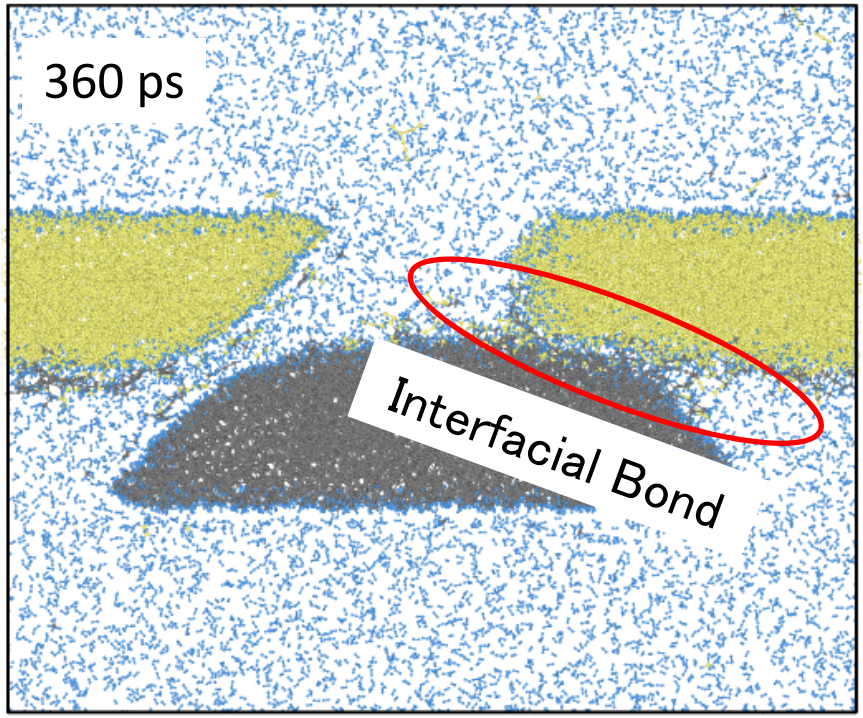
- H_2, O_2, H_2O, CH_3OH, etc.

Kubo Laboratory
Multi-Physics Computational Science

Vacuum



H_2 Environment (500 atm)

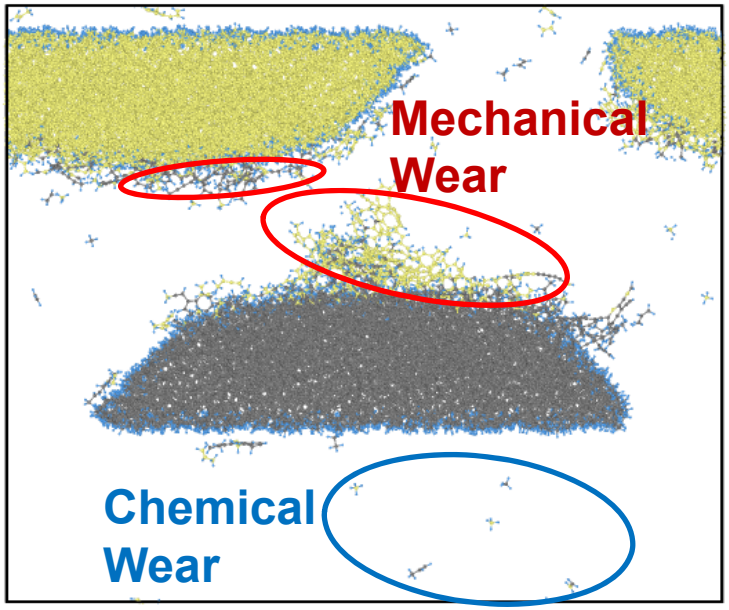


1) **Wear is Prohibited by H_2 Reaction** (H terminates Dangling Bond on C atom)
2) **Self-Healing by CH_3OH Reaction** (CH_3 adsorbs on C atom)

"Chemical Wear" versus "Mechanical Wear"
- Effect of H_2 Environment

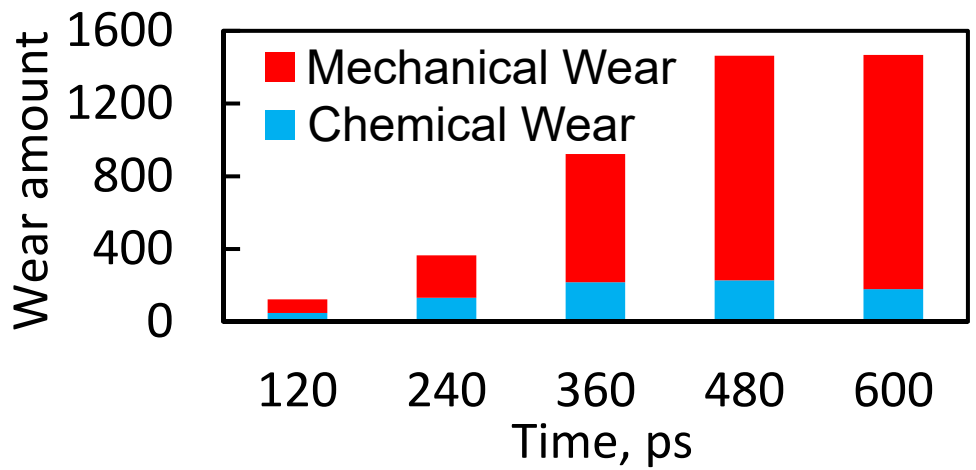
Kubo Laboratory
Multi-Physics Computational Science

Rising Process Simulation of DLC

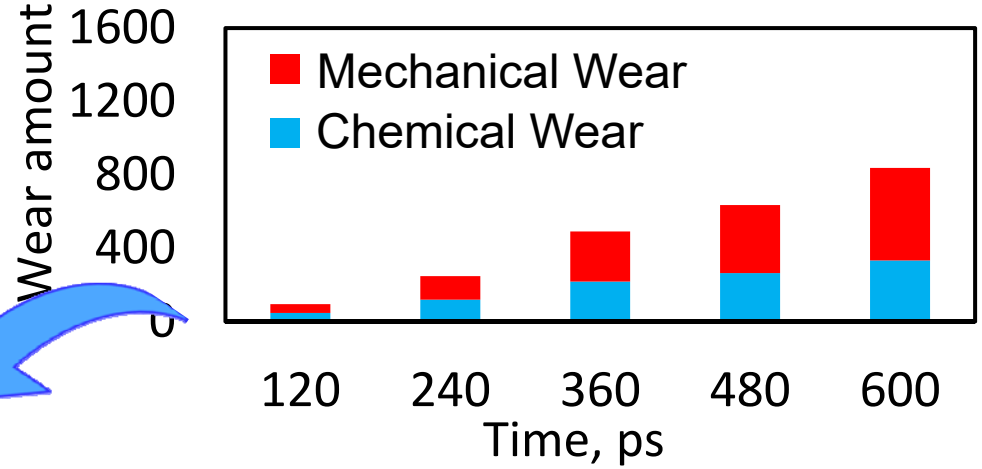


Wear = Mechanical Wear + Chemical Wear

Vacuum



H_2 Environment



H_2 Environment

- Mechanical Wear Decreases
- Chemical Wear Increases

Silicon-Based Solar Cell

Current status of Solar Cells

The production is increasing significantly.

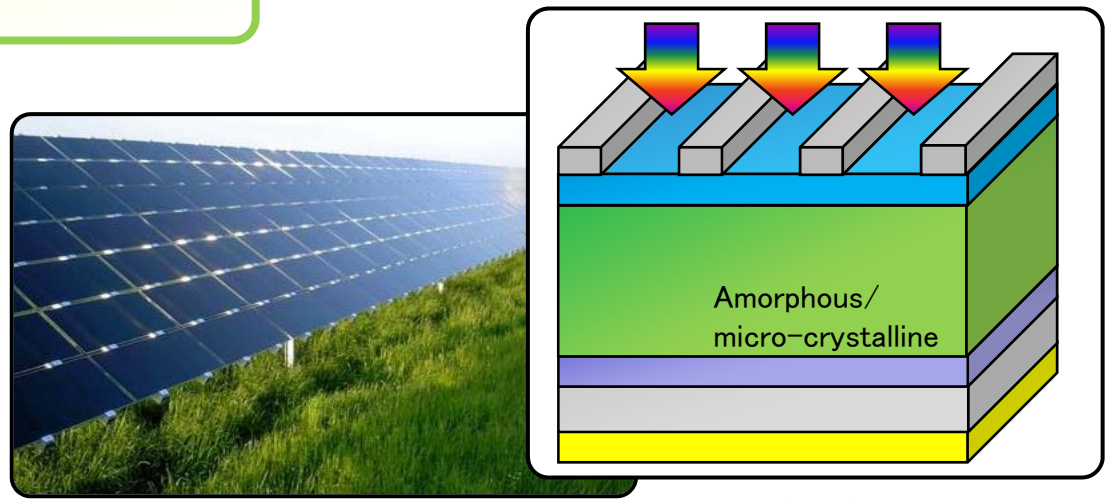
New materials of solar cells and the production technique should be developed for large-scale production.

Crystalline Silicon Solar Cells

*Tokyo electric power company

Thin-film Silicon Solar Cells

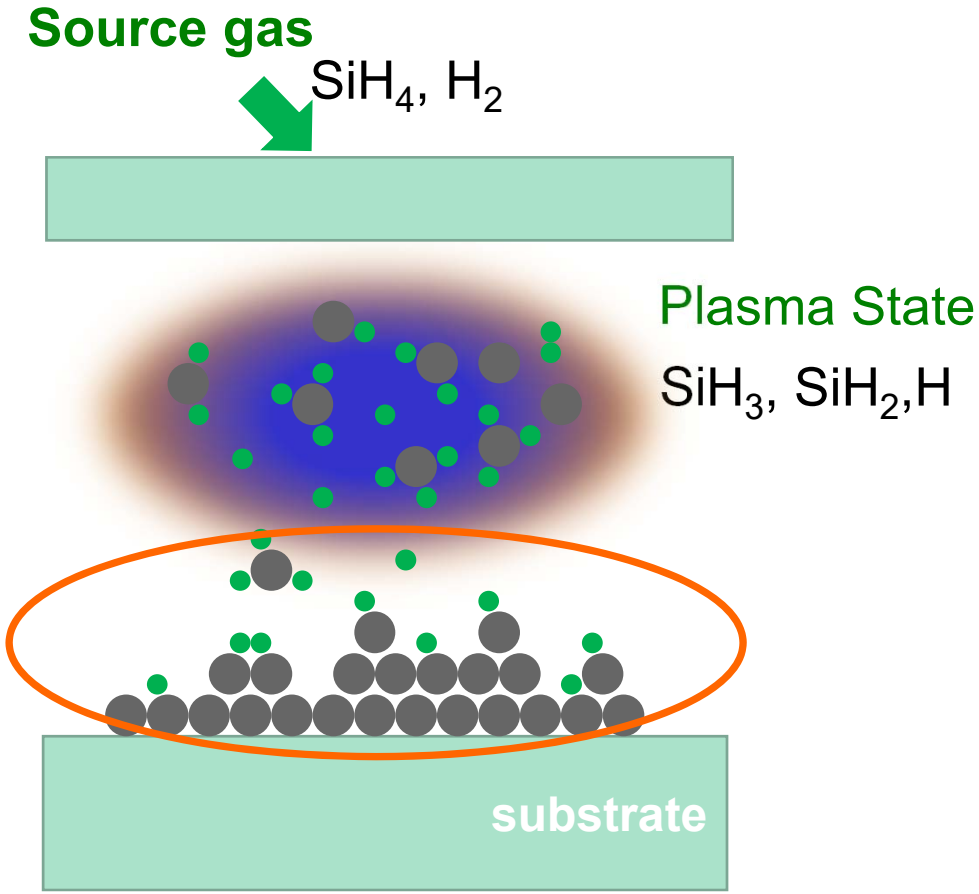
- Low-cost
- Saving resource
- Large-scale production
- Low efficiency
- Low-lifetime



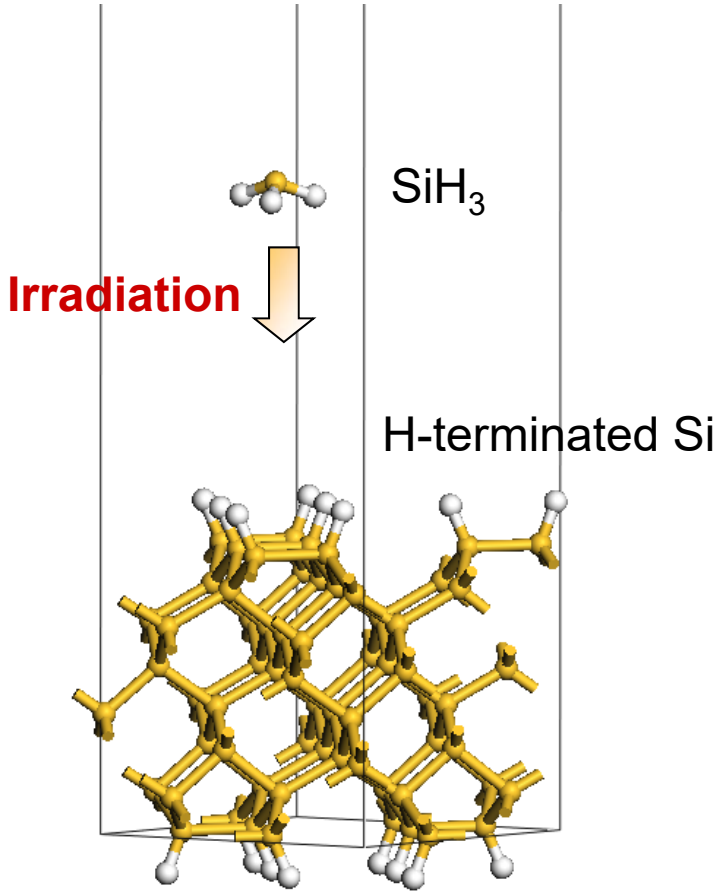
*PrimeStar Solar, Inc.

Quantum Chemical Molecular Dynamics Simulation
on Chemical Vapor Deposition

Plasma CVD



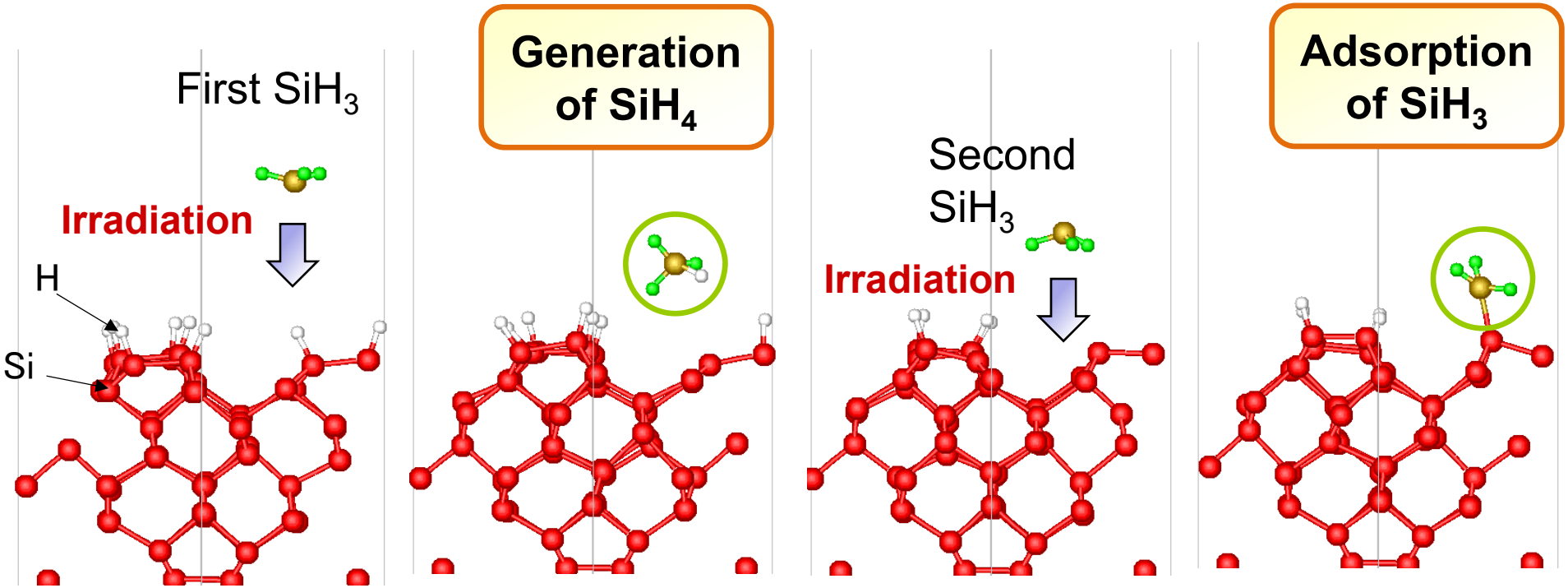
Crystal Growth Simulation with Chemical Reactions



Complicated Chemical Reactions

Quantum Chemistry is Essential

Chemical Vapor Deposition Process of Si(001) Surface by SiH_3

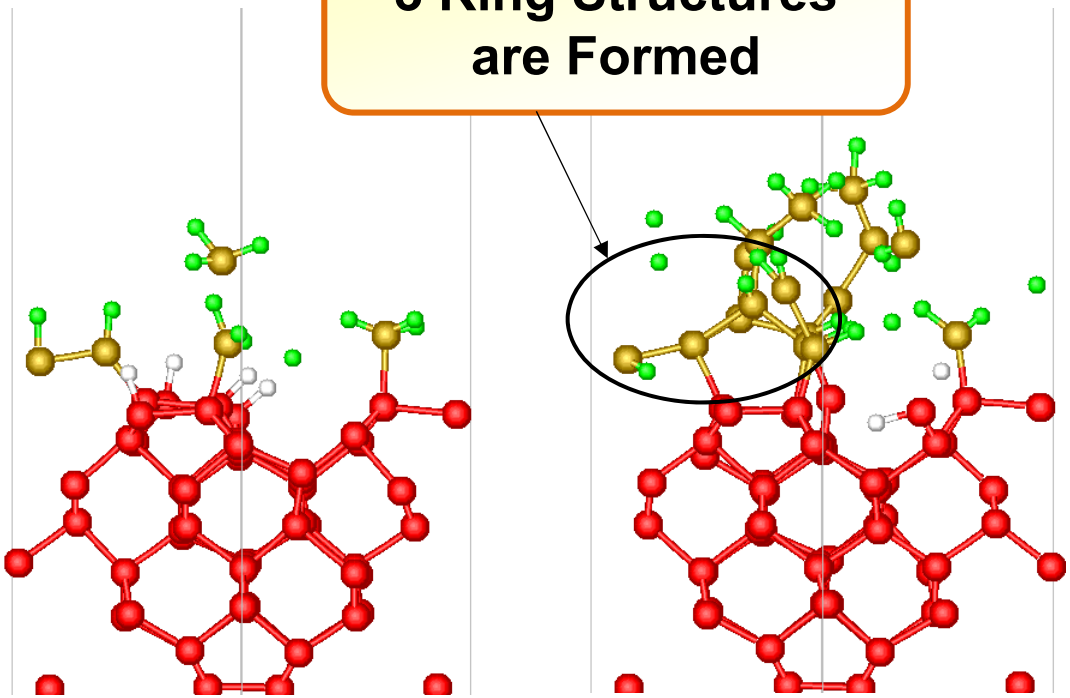


0.01 ps | 0.85 ps | 14.3 ps | 14.9 ps

Formation of New Si-Si Bond → 2 SiH_3 are essential.

Chemical Vapor Deposition Process of Si(001) Surface by SiH_3

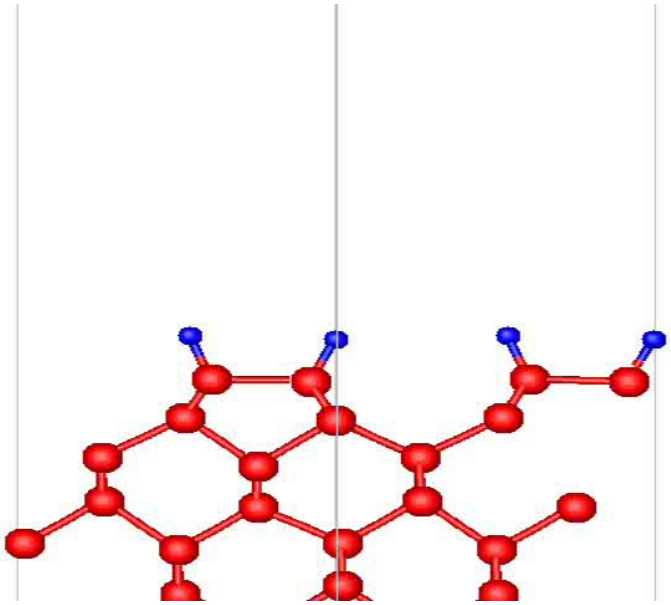
3 Ring Structures are Formed



44.2 ps

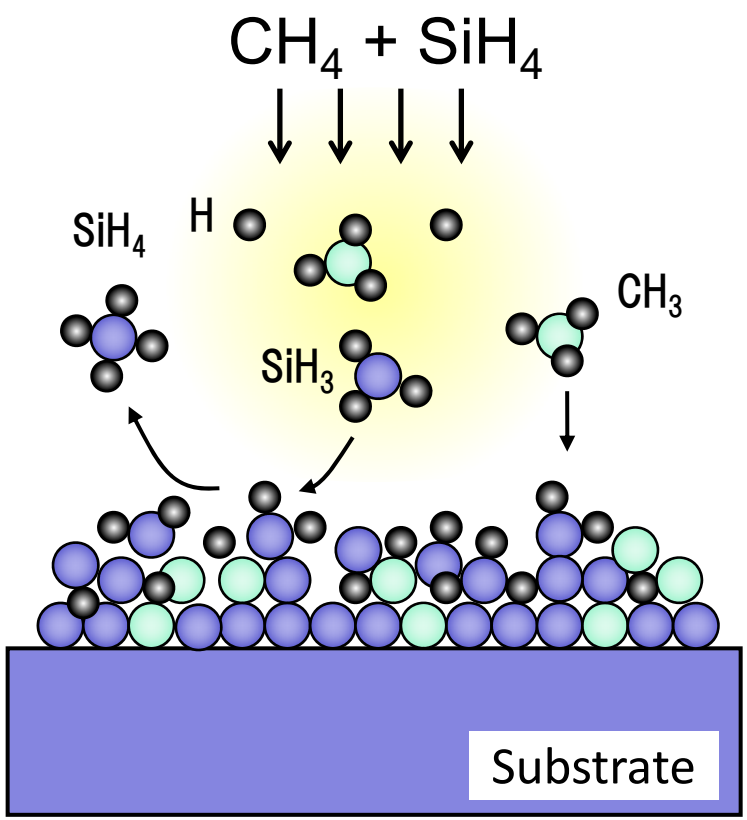
160.0 ps

Animation

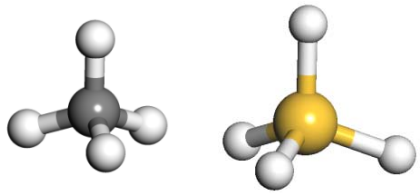


Continuous Formation of Si Rings → Epitaxial Growth of Si (001)

Chemical Vapor Deposition (CVD) of a-$Si_{1-x}C_x$:H



Source Gases



Methane Silane

Growth Mechanism of a-$Si_{1-x}C_x$:H
has not been understood

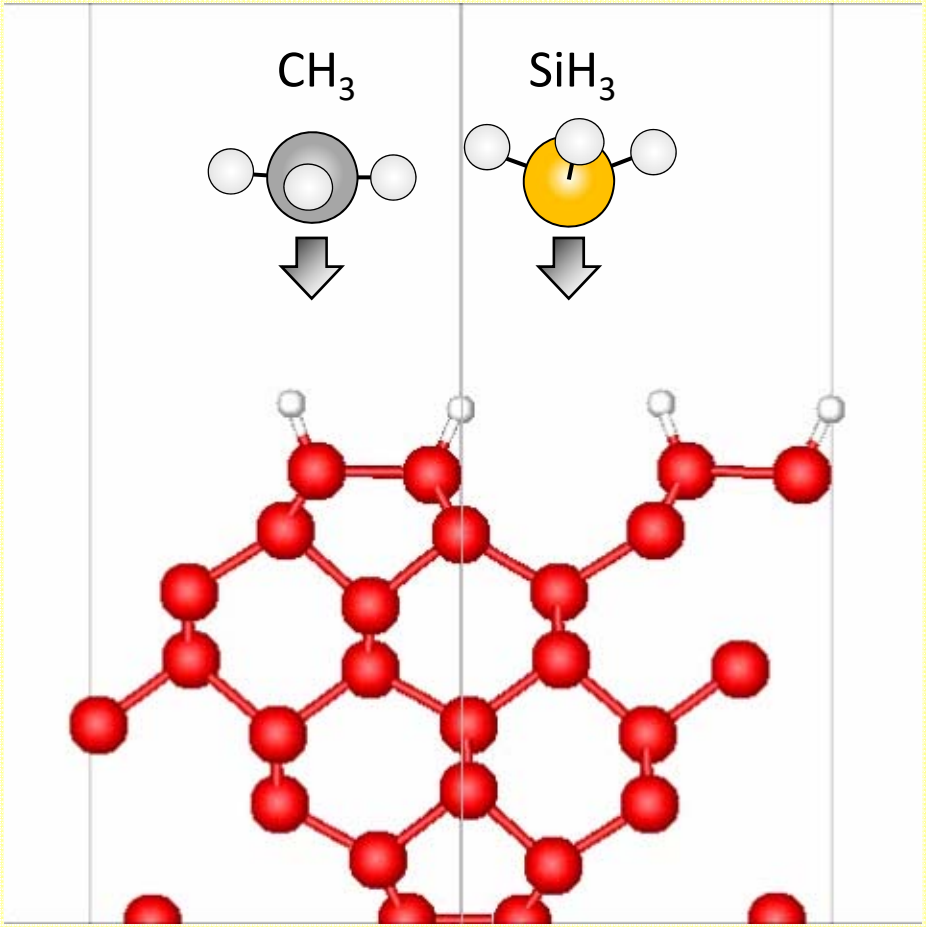
Quantum Chemical Molecular Dynamics
is effective for
understanding growth mechanisms

Purpose

(1) **Growth Mechanism** of a-$Si_{1-x}C_x$:H in CVD
(2) **Effect of $CH_3/(CH_3+SiH_3)$ Ratio** on Structures

CVD Simulation of a-$Si_{1-x}C_x$:H

$CH_3/(CH_3+SiH_3)$ Ratio of 0.5



Substrate: Si(001)-(2x1):H

Temperature: 500 (K)

Time step: 0.1 (fs)

Total simulation time: 160.0 (ps)

Impinging species:

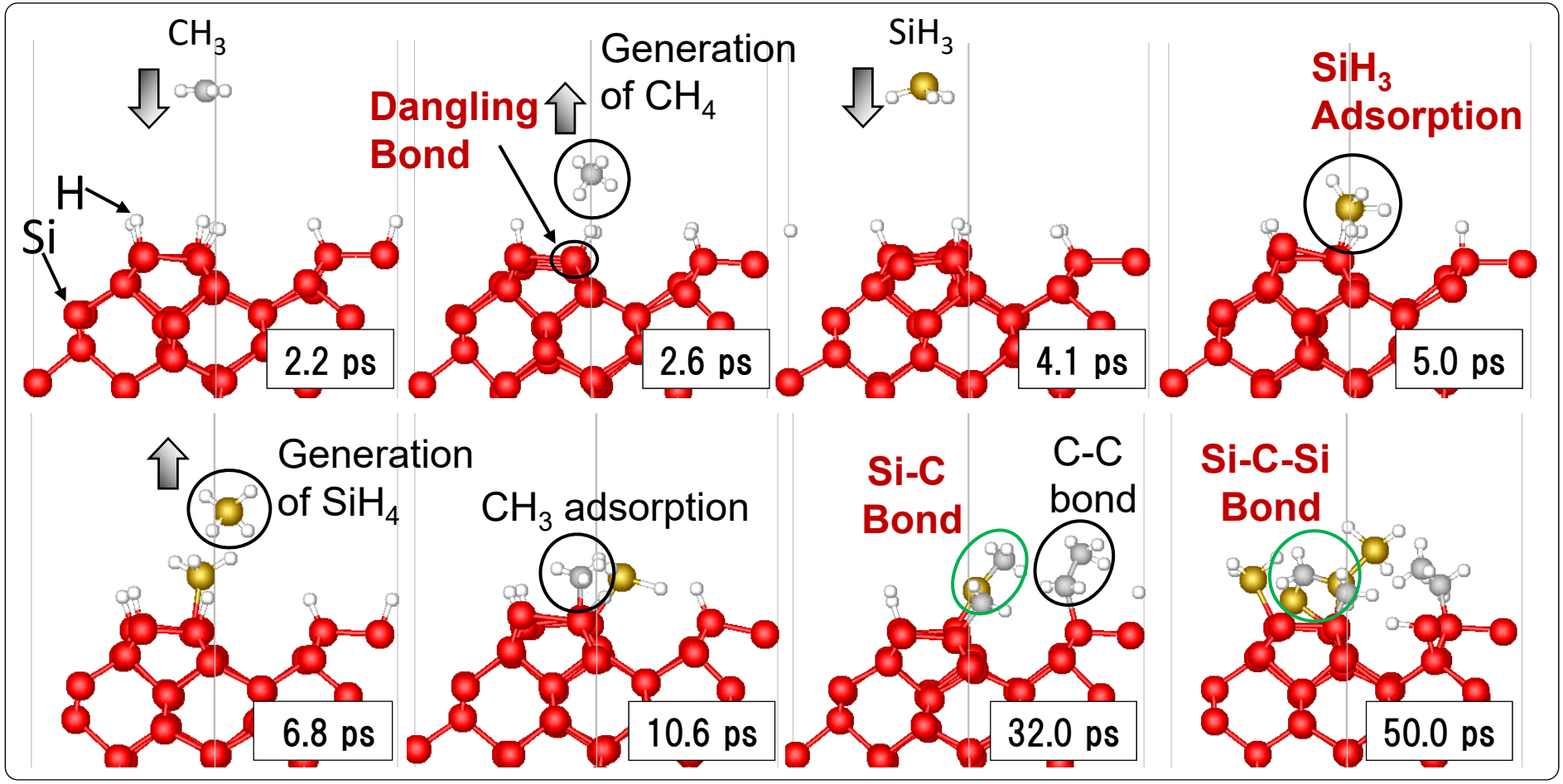
SiH_3 radical CH_3 radical

Time interval: 2.0 ps

Kinetic energy: 0.1 eV

CVD Simulation of a-$Si_{1-x}C_x$:H

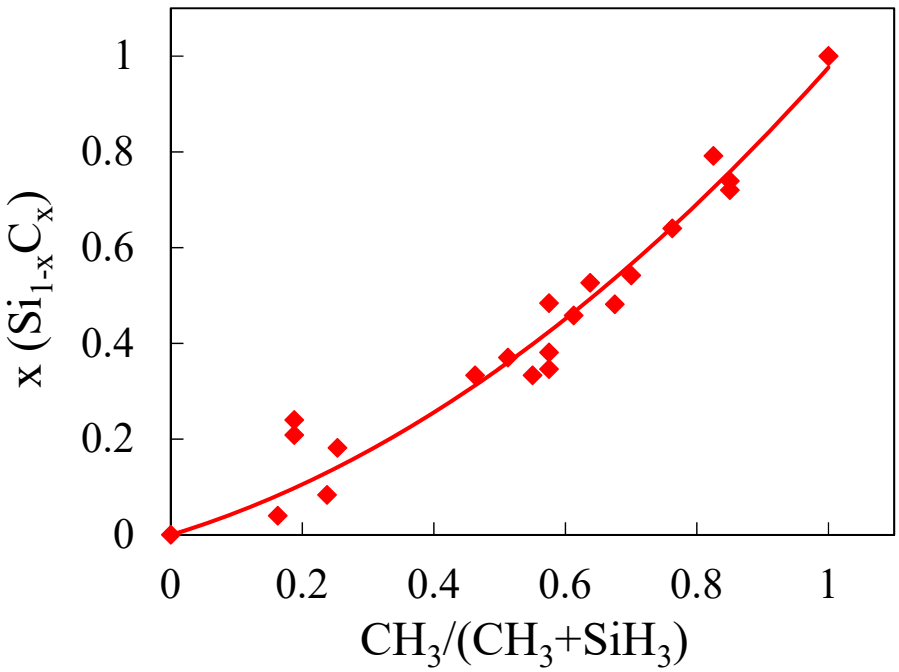
$CH_3/(CH_3+SiH_3)$ Ratio of 0.5



Via H-abstraction and adsorption, C and Si atoms are deposited.

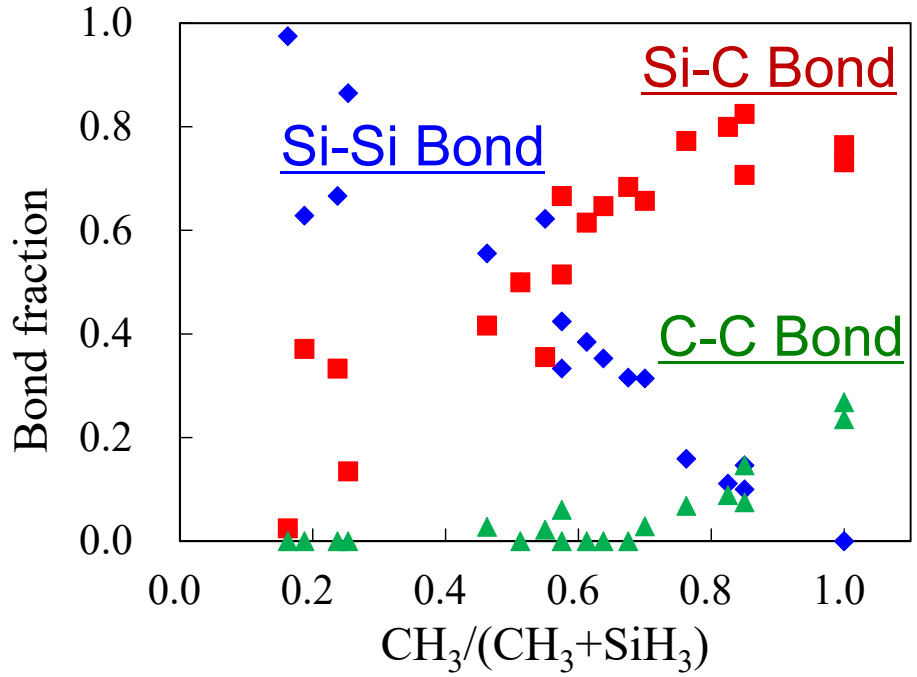
Effect of CH_3 Ratio on Structures

Carbon Atom Content



C atom content increases with increasing CH_3 ratio

Bonding States



Si-Si bonds decrease

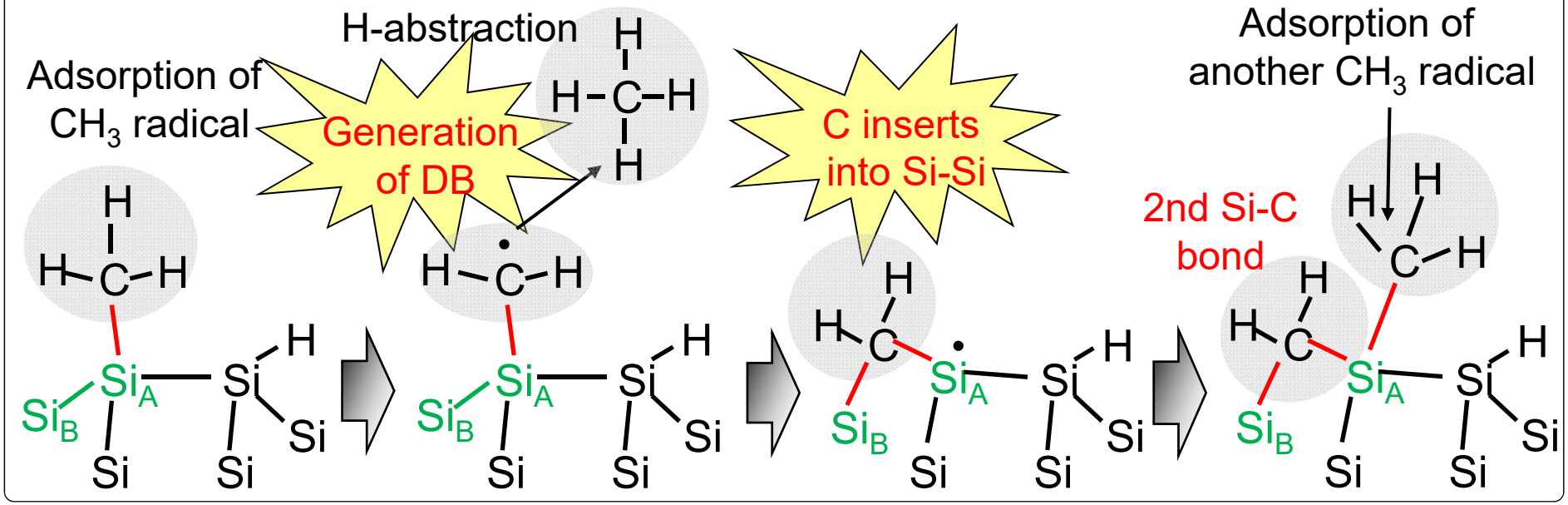
Si-C bonds increase

Few C-C bonds are generated

At high CH_3 ratio, heteronuclear Si-C bonds are preferentially formed

Why Few C-C Bonds are Formed in the a-$Si_{1-x}C_x$:H Films ?

Formation Mechanism of Si-C Bonds



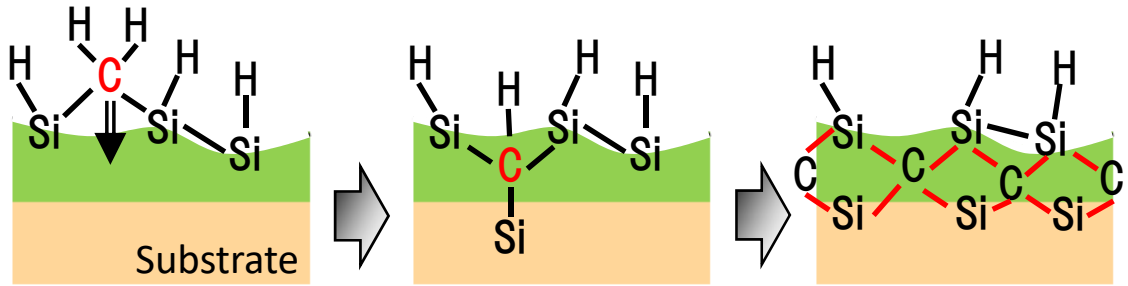
Bond Recombination

C inserts into Si-Si Bond

These suppress the formation of C-C bonds

C atoms penetrate into a film

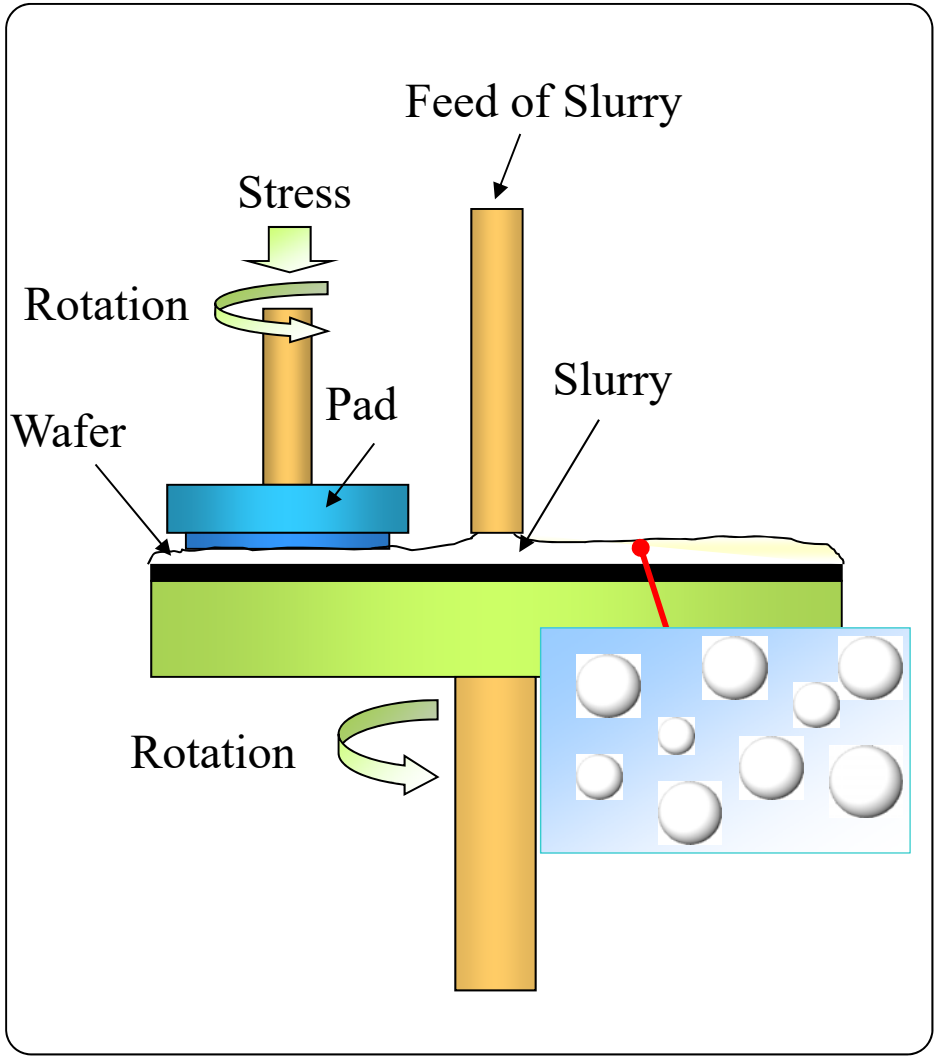
Formation of Si-C bond network



Multi-Physics Simulation
on "Friction" and "Chemical Reaction"

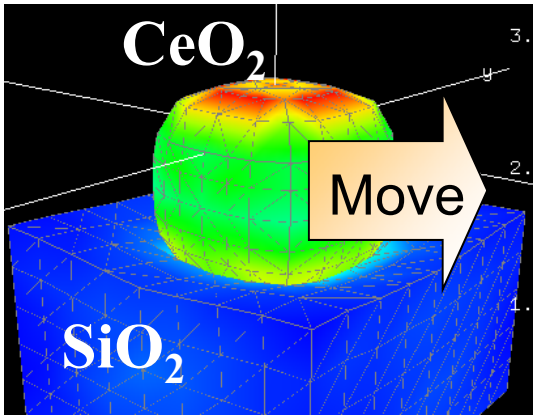
Kubo Laboratory
Multi-Physics Computational Science

Chemical Mechanical Polishing



Traditional Approach - Finite Element Method

(1) Mechanical Polishing of Glasses



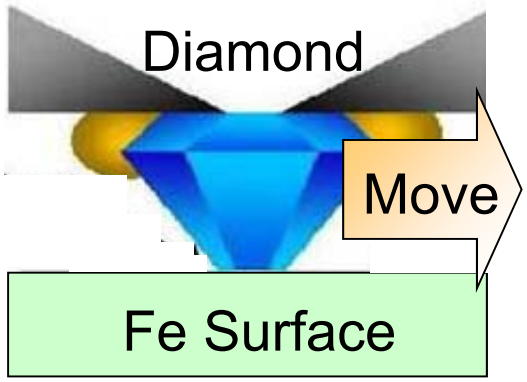
Soft CeO_2

↓

Hard SiO_2
is polished.

Cannot Explain

(2) Mechanical Polishing of Metal



Hard Diamond

↓

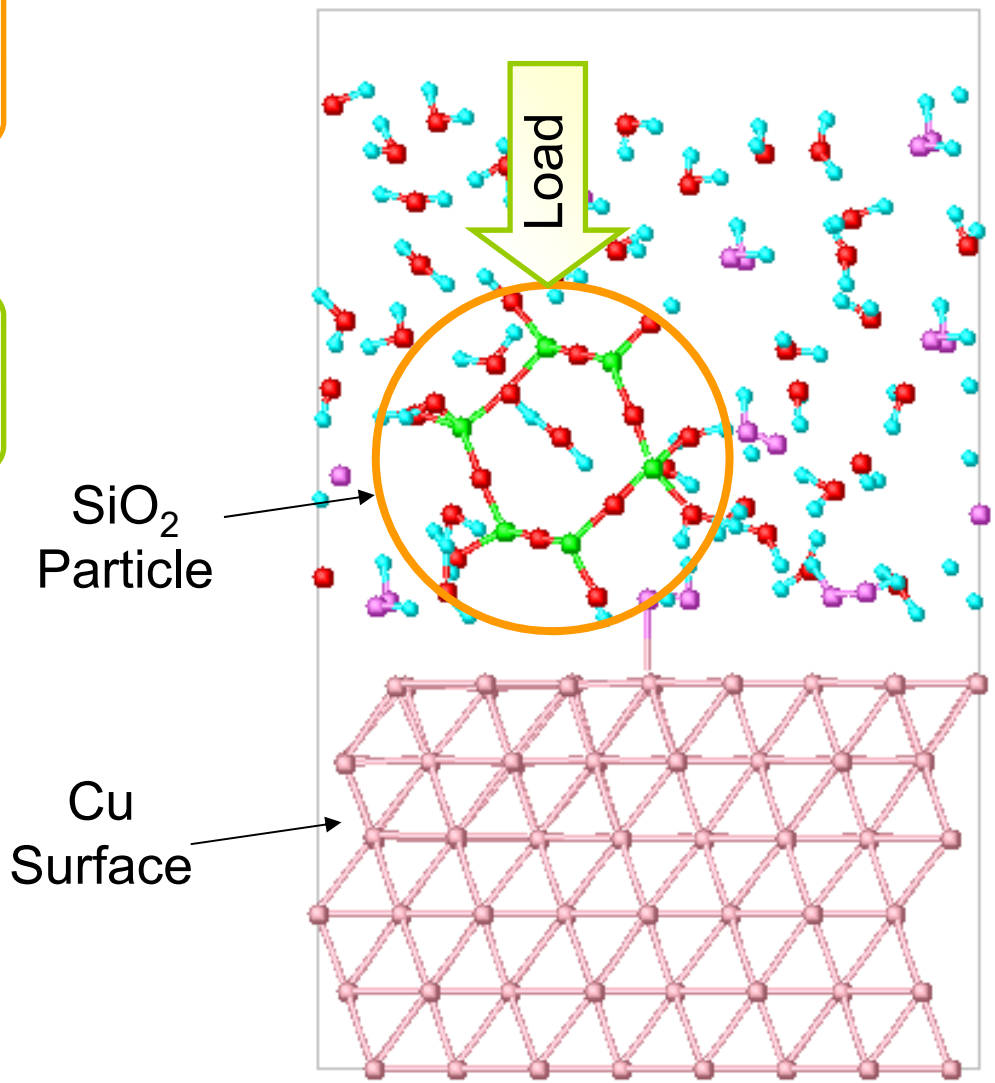
Soft Fe
is not polished.

Cannot Explain

Chemical Mechanical Polishing Simulation of Cu Surface by SiO_2 Abrasive Grain

Relaxation
Process

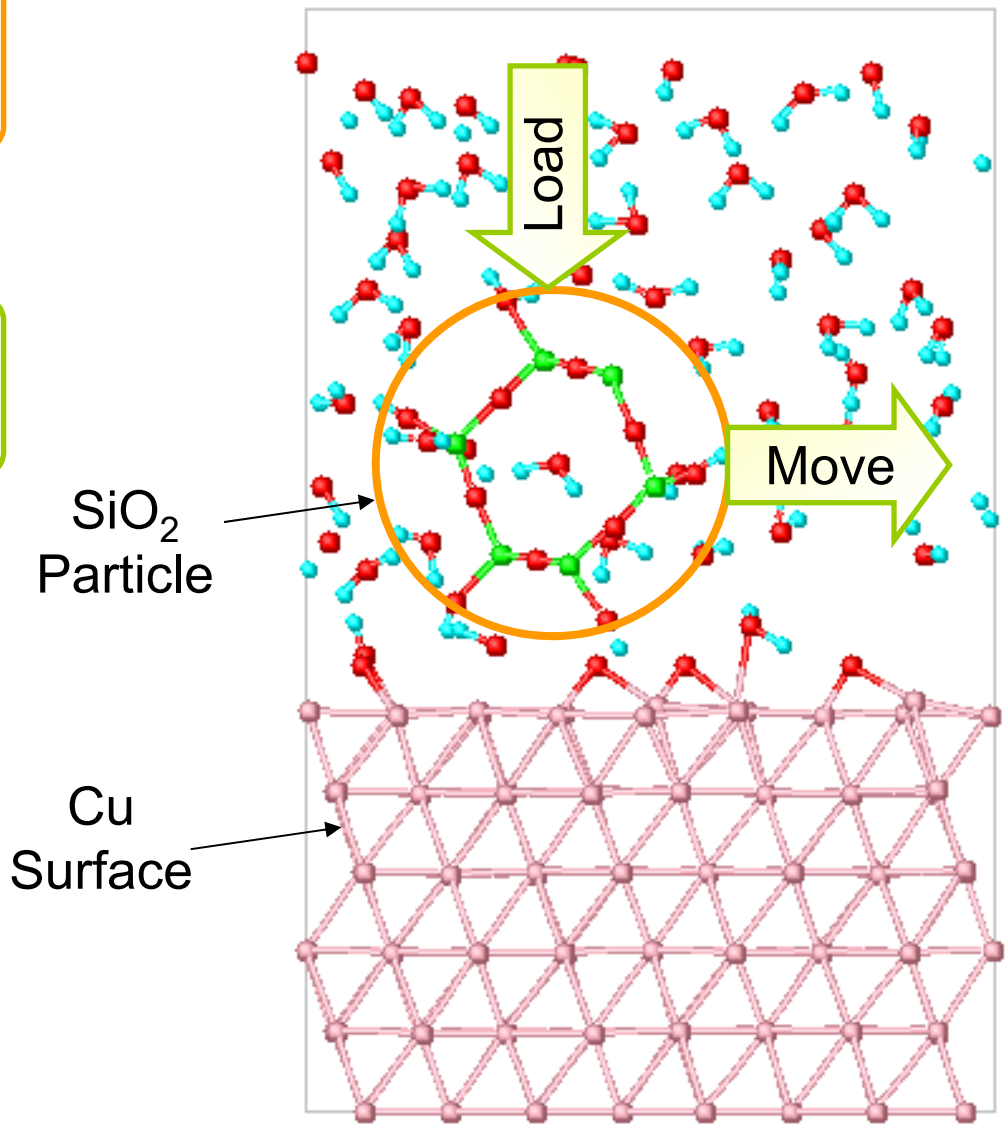
Under
$H_2O_2 + H_2O$



Chemical Mechanical Polishing Simulation of Cu Surface by SiO_2 Abrasive Grain

Cu Polishing
by SiO_2 Particle

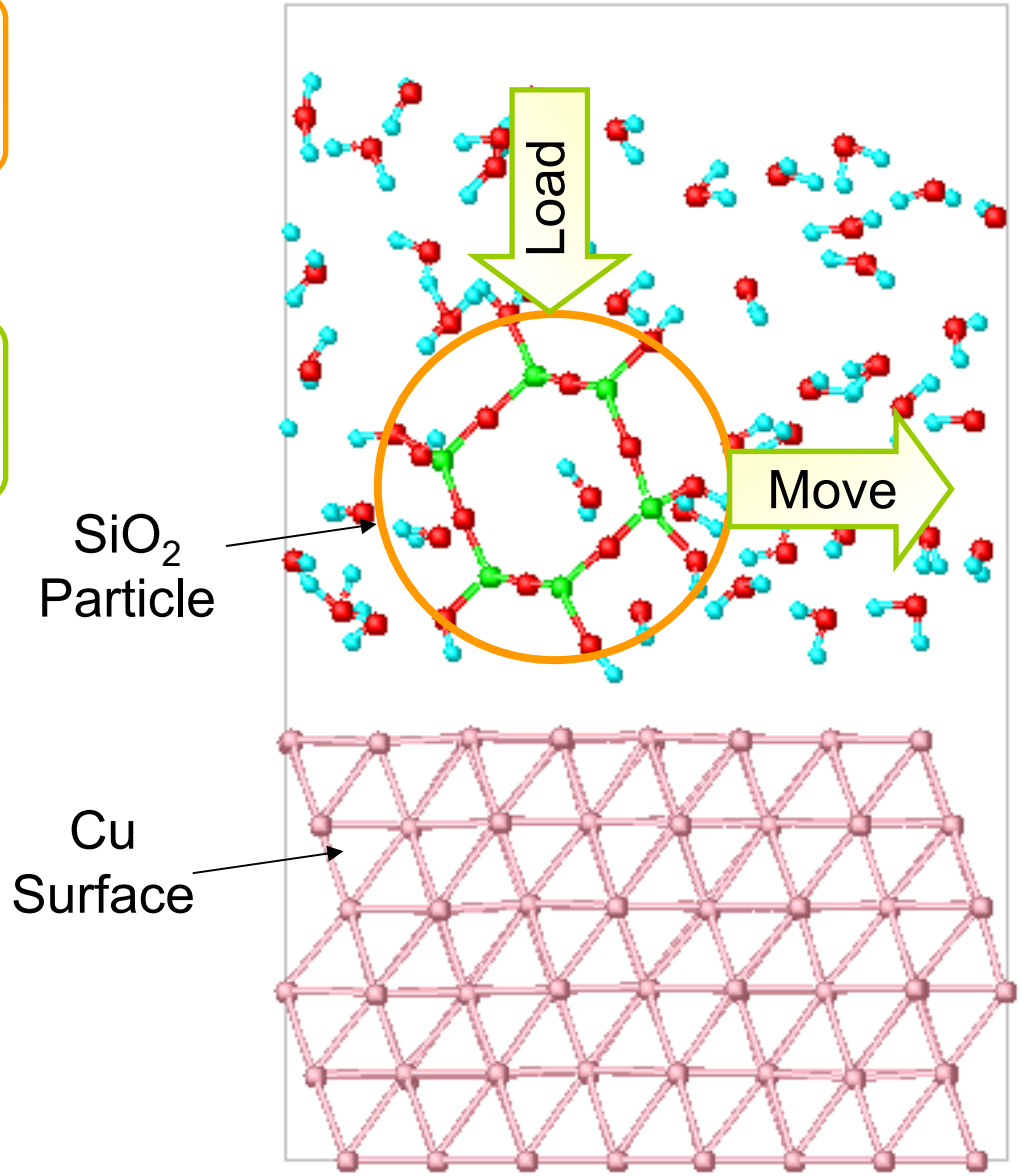
Under
$H_2O_2 + H_2O$



Chemical Mechanical Polishing Simulation of Cu Surface by SiO_2 Abrasive Grain

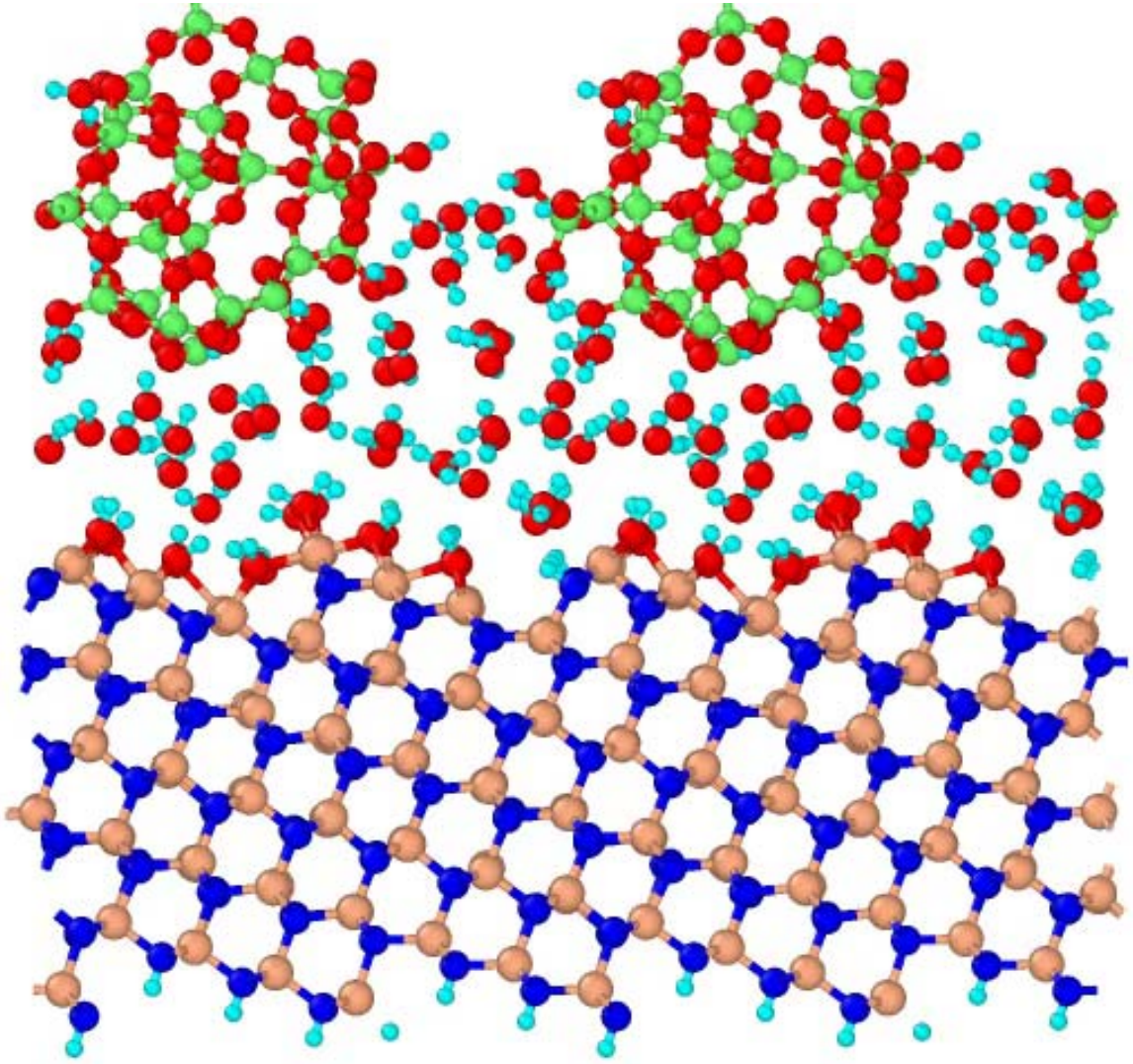
Cu Polishing
by SiO_2 Particle

Under
H_2O Only



Chemical Mechanical Polishing Simulation of GaN Surface

Kubo Laboratory
Multi-Physics Computational Science

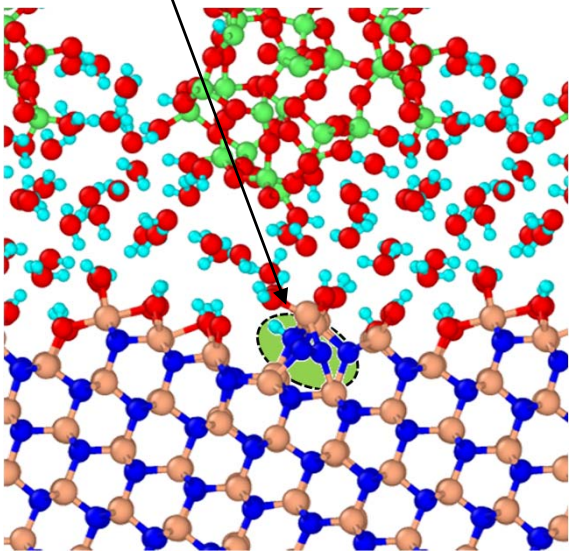


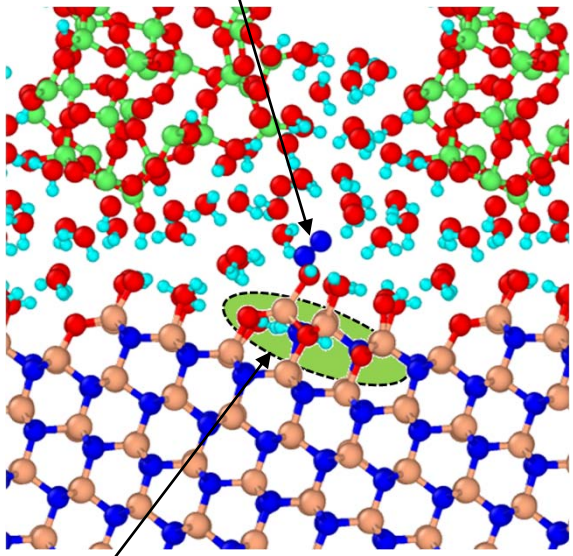
Chemical Mechanical Polishing Simulation of GaN Surface

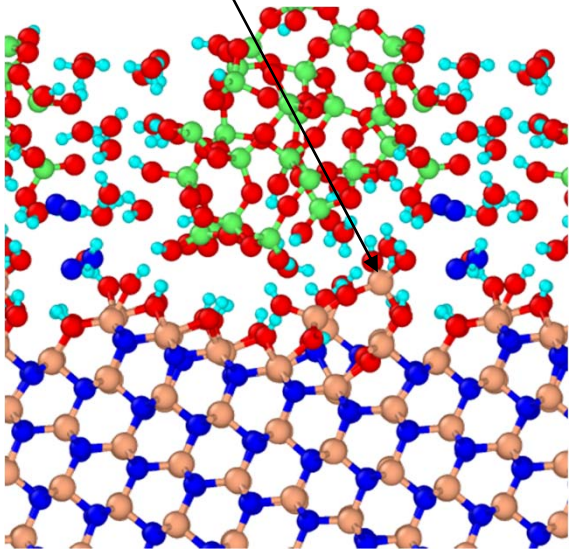
Generation of N-N Bond **at Step**

Evaporation of N_2 Molecule

Desorption of Ga as **$Ga(OH)_3$**







179.3 ps

Intrusion of O Atom **from Step**

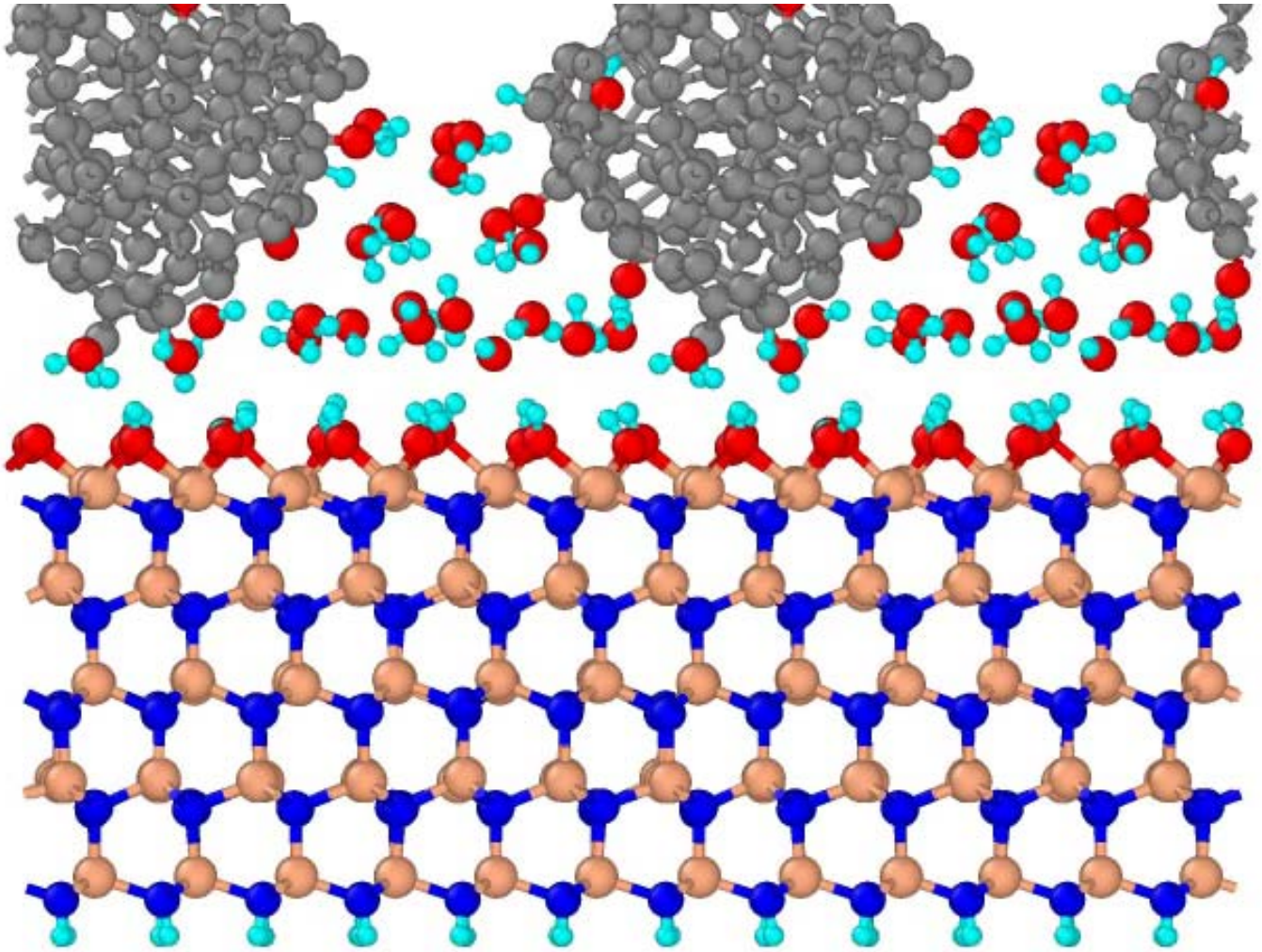
218.2 ps

600.2 ps

Oxidation and N_2 Evaporation Starts **from Step**

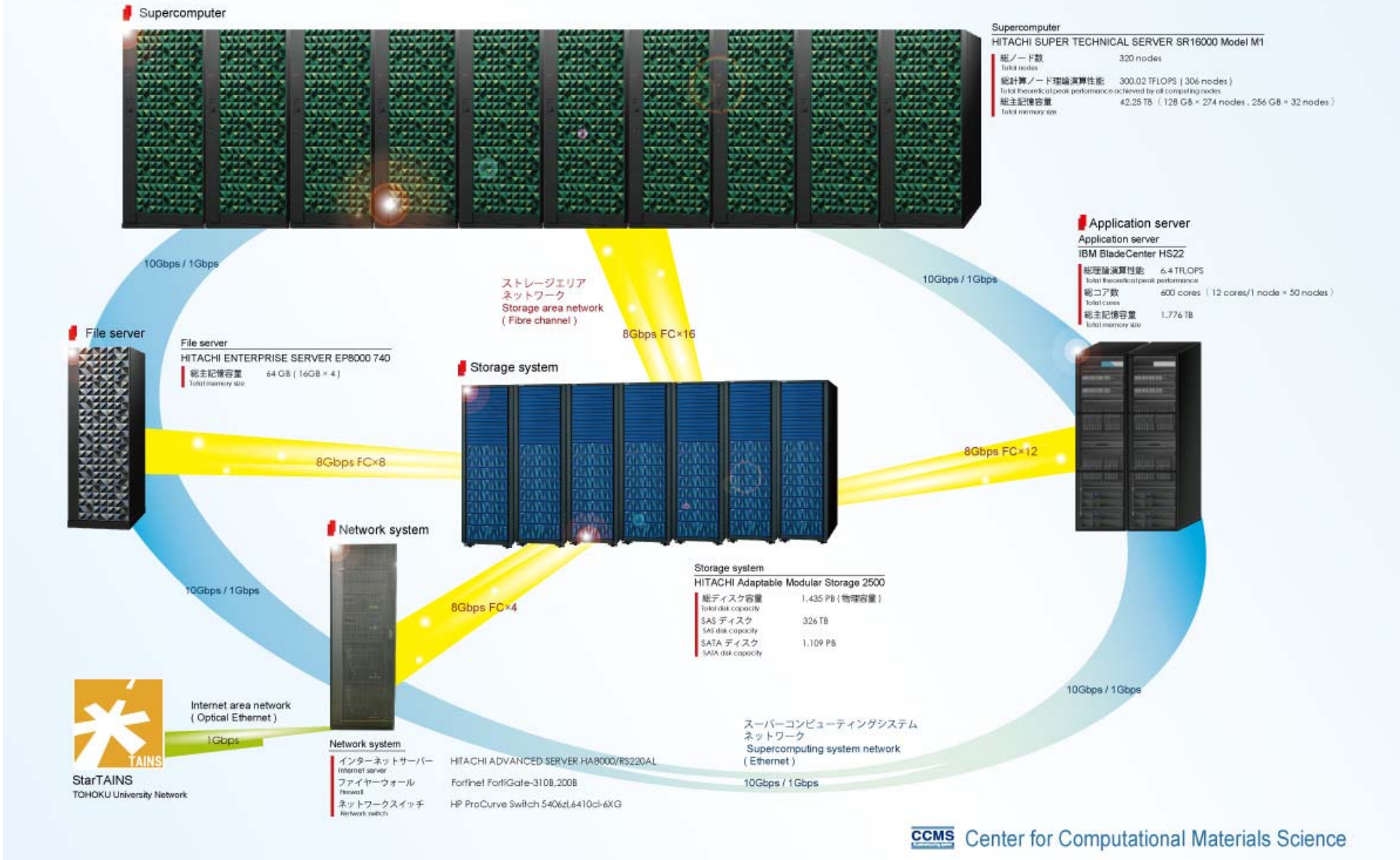
- CMP Proceeds Layer-by-Layer

Chemical Mechanical Polishing Simulation of GaN using Nano-Diamond



Damaged Layers are Generated because Nano-Diamond is Hard

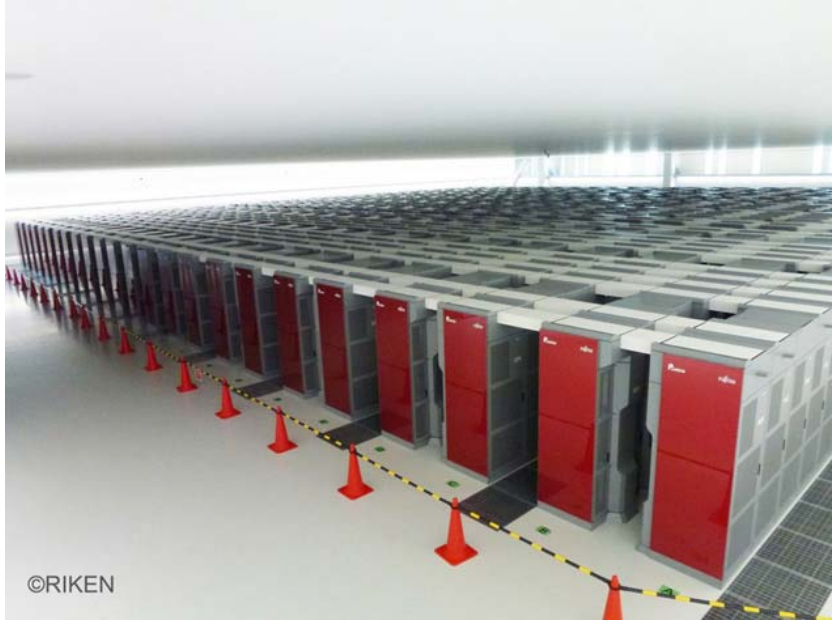
From 2017, Director of Supercomputer Center,
Institute for Materials Research, Tohoku University



Supercomputer Project in Japan
"Post-K"

Supercomputer "K" in Kobe

Completion on June, 2012
Start to share on Sep. 2012



80,000 Nodes, **700,000 Cores**
Speed：**10^{16} Calc./Sec. (京)**

Supercomputer World Ranking on June, 2011

TOP 10 Sites for June 2011

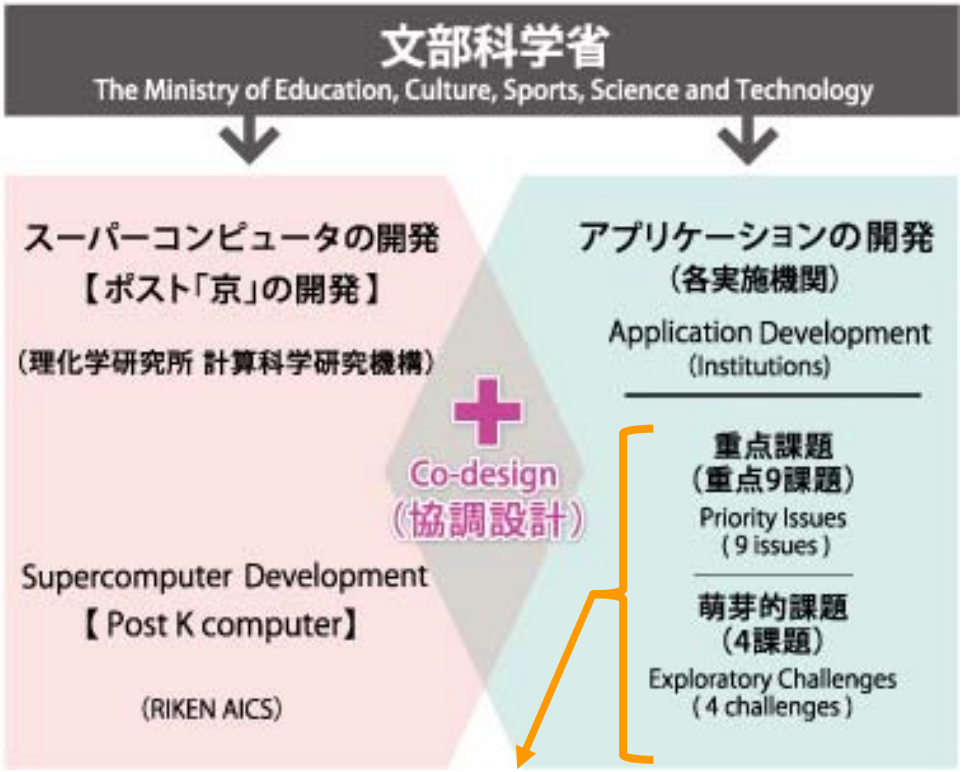
For more information about the sites and systems in the list, click on the links or view the complete list.

Rank	Site	System	Cores	Rmax (TFlop/s)
1	RIKEN Advanced Institute for Computational Science (AICS) Japan	K computer, SPARC64 VIIIfx 2.0GHz, Tofu interconnect Fujitsu	548,352	8,162.0
2	National Supercomputing Center in Tianjin China	**Tianhe-1A** - NUDT TH MPP, X5670 2.93Ghz 6C, NVIDIA GPU, FT-1000 8C NUDT	186,368	2,566.0
3	DOE/SC/Oak Ridge National Laboratory United States	**Jaguar** - Cray XT5-HE Opteron 6-core 2.6 GHz Cray Inc.	224,162	1,759.0
4	National Supercomputing Centre in Shenzhen (NSCS) China	**Nebulae** - Dawning TC3600 Blade, Intel X5650, NVidia Tesla C2050 GPU Sugon	120,640	1,271.0
5	GSIC Center, Tokyo Institute of Technology Japan	**TSUBAME 2.0** - HP ProLiant SL390s G7 Xeon 6C X5670, Nvidia GPU, Linux/Windows NEC/HP	73,278	1,192.0
6	DOE/NNSA/LANL/SNL United States	**Cielo** - Cray XE6 8-core 2.4 GHz Cray Inc.	142,272	1,110.0
7	NASA/Ames Research Center/NAS United States	**Pleiades** - SGI Altix ICE 8200EX/8400EX, Xeon HT QC 3.0/Xeon 5570/5670 2.93 Ghz, Infiniband SGI	111,104	1,088.0

Ministry of Education, Culture, Sports, Science, and Technology
- Post K Project

Supercomputer World Ranking on Nov., 2017

Rank	Site	System	Cores	Rmax (TFlop/s)	Rpeak (TFlop/s)	Power (kW)
1	National Supercomputing Center in Wuxi China	**Sunway TaihuLight** - Sunway MPP, Sunway SW26010 260C 1.45GHz, Sunway NRCPC	10,649,600	93,014.6	125,435.9	15,371
2	National Super Computer Center in Guangzhou China	**Tianhe-2 (MilkyWay-2)** - TH-IVB-FEP Cluster, Intel Xeon E5-2692 12C 2.200GHz, TH Express-2, Intel Xeon Phi 31S1P NUDT	3,120,000	33,862.7	54,902.4	17,808
3	Swiss National Supercomputing Centre (CSCS) Switzerland	**Piz Daint** - Cray XC50, Xeon E5-2690v3 12C 2.6GHz, Aries interconnect , NVIDIA Tesla P100 Cray Inc.	361,760	19,590.0	25,326.3	2,272
4	Japan Agency for Marine-Earth Science and Technology Japan	**Gyoukou** - ZettaScaler-2.2 HPC system, Xeon D-1571 16C 1.3GHz, Infiniband EDR, PEZY-SC2 700Mhz ExaScaler	19,860,000	19,135.8	28,192.0	1,350
5	DOE/SC/Oak Ridge National Laboratory United States	**Titan** - Cray XK7, Opteron 6274 16C 2.200GHz, Cray Gemini interconnect, NVIDIA K20x Cray Inc.	560,640	17,590.0	27,112.5	8,209
6	DOE/NNSA/LLNL United States	**Sequoia** - BlueGene/Q, Power BQC 16C 1.60 GHz, Custom IBM	1,572,864	17,173.2	20,132.7	7,890
7	DOE/NNSA/LANL/SNL United States	**Trinity** - Cray XC40, Intel Xeon Phi 7250 68C 1.4GHz, Aries interconnect Cray Inc.	979,968	14,137.3	43,902.6	3,844
8	DOE/SC/LBNL/NERSC United States	**Cori** - Cray XC40, Intel Xeon Phi 7250 68C 1.4GHz, Aries interconnect Cray Inc.	622,336	14,014.7	27,880.7	3,939
9	Joint Center for Advanced High Performance Computing Japan	**Oakforest-PACS** - PRIMERGY CX1640 M1, Intel Xeon Phi 7250 68C 1.4GHz, Intel Omni-Path Fujitsu	556,104	13,554.6	24,913.5	2,719
10	RIKEN Advanced Institute for Computational Science (AICS) Japan	K computer, SPARC64 VIIIfx 2.0GHz, Tofu interconnect Fujitsu	705,024	10,510.0	11,280.4	12,660



Japan-Made Simulator Development Project

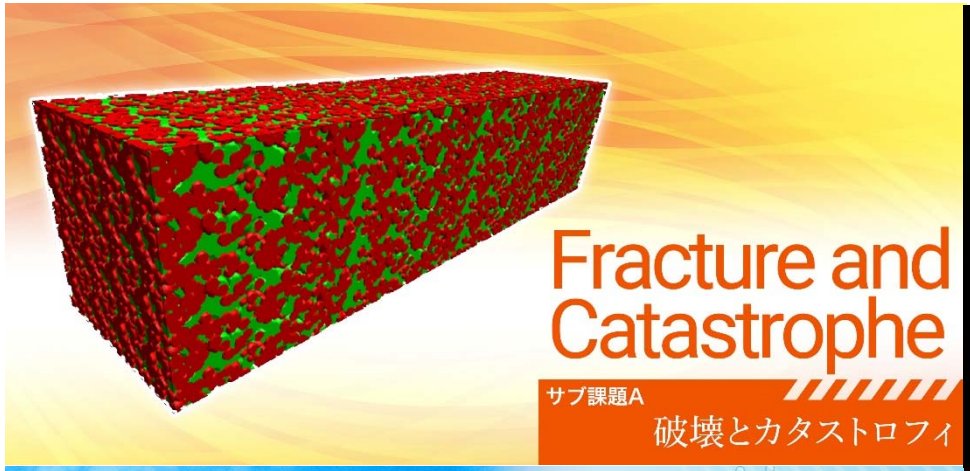
9 Priority Issues
(2015 - 2019)

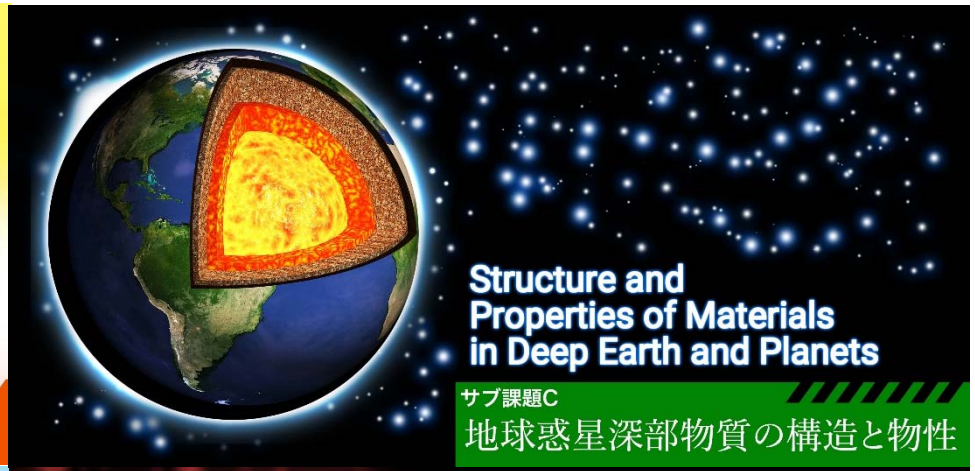
4 Challenge Issues
(2016 – 2019)

Supercomputer Post-K Project in Japan for Exploring Extremes through Multi-Physics and Multi-Scale Simulations

CBSM²
Challenge of Basic Science
– Exploring Extremes through Multi-Physics and Multi-Scale Simulations

Project Leader : Momoji Kubo





Post-K Project "Challenge of Basic Science"
- Exploring Extremes through Multi-Physics and Multi-Scale Simulations" (2016 - 2019FY)

Science for Exploring Extremes

1) Computational Science by "K"
2) Experiment and Observation by Large Facilities such as Spring-8

Significant Advancement of Individual Issues at Specific Scale

Objective of Our Post-K Project

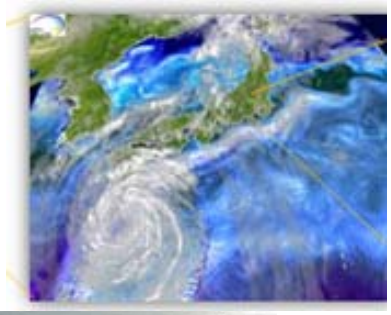
Unresolved Issues

Multi-Scale and Multi-Physics Problems

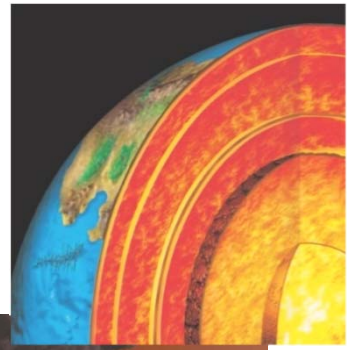
Micro-Scale Phenomena → Macro-Scale Property

Earthquake

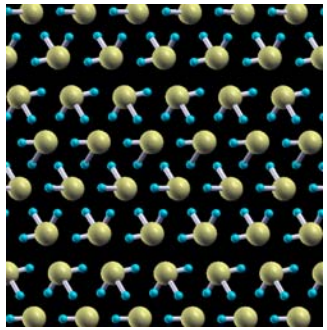
Fluctuation of Atmosphere



Magma



Extreme Physical Property



(Superconductor H_5S_2)

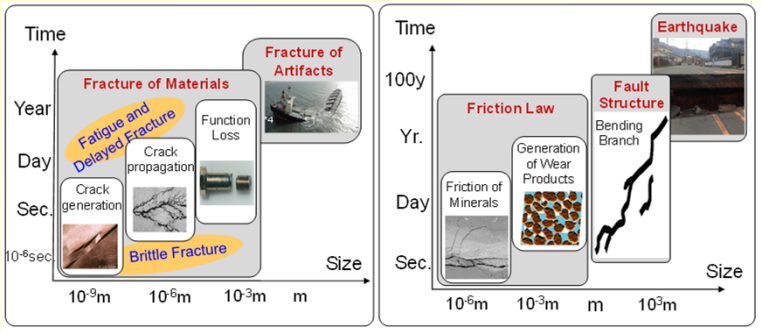
Fracture & Catastrophe

Volcano Eruption

Targets of 4 Sub-Groups

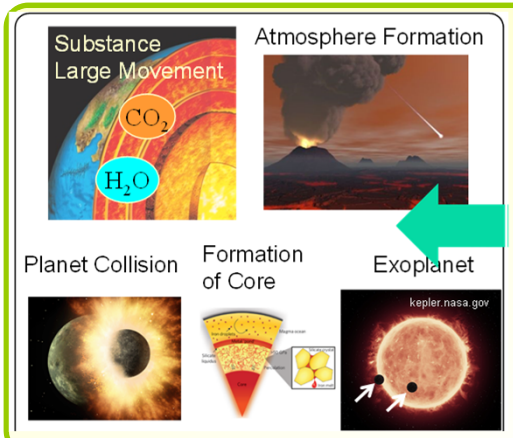
A) Fracture and Catastrophe

Fracture of Materials Fracture of Fault



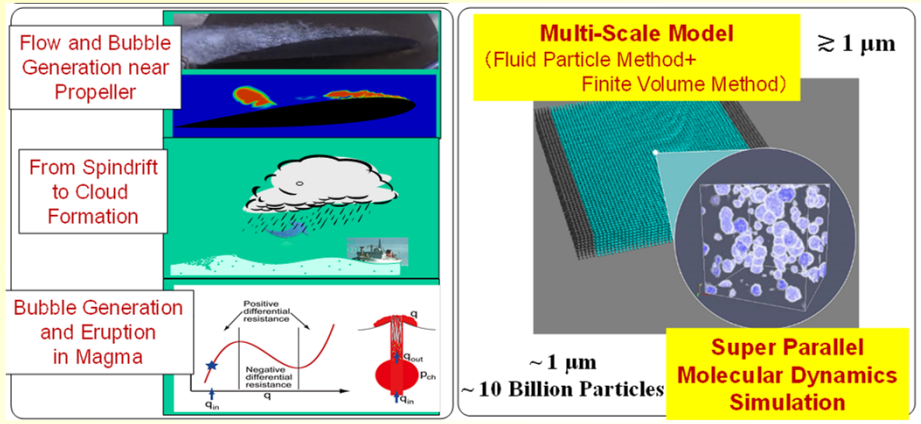
What Kind of Hierarchy Mechanism Works in "Fracture Process" ?

C) Structure and Properties of Materials in Deep Earth and Planets



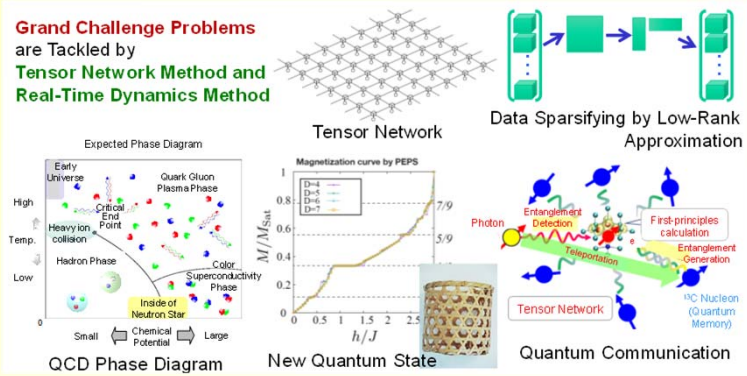
How Our Hometown "Earth" was Generated ?

B) Phase Transition and Flow



What Kind of Multi-Scale Mechanism Works in "Complex Multiphase Flow"

D) Fundamental Quantum Mechanics and Informatics



Development of Solver which can Solve Quantum Mechanics Many-Body Problems

Organization
(Leader: Momoji Kubo, Tohoku Univ.)

Sub-Issue A Leader
Momoji Kubo (Tohoku Univ., IMR)
"Simulation of Crack Generation and Chemical Reactions"

- JAEA Yamaguchi "Simulation of Crack Generation and Chemical Reactions"
- Osaka Univ. Ogata "Simulation of Cracking and Crack Propagation"
- Univ. Tokyo Hatano "Simulation of Fault Fracture Dynamics"
- Kanazawa Univ. Shimokawa "Common Statistical Law of Earthquake and Materials Fracture"

Sub-Issue B Leader
Toshihiro Kawakatsu (Tohoku Univ., Grad. Sci.)
"Development of MSSP and Simulation of Complex Flow"

- Univ. Tokyo Noguchi "Analysis of Complex Fluid by Molecular Dynamics Method"
- Kyushu Univ. Tsuda "Meso and macro Simulation of Internal Flow in Machine"
- JAMSTEC Onishi "Multi-Scale Simulation of Droplet Generation in Cloud Formation"

Sub-Issue C Leader
Toshiaki Iitaka (RIKEN)
"Simulation of Silicate Solution"

- Ehime Univ. Tsuchiya "Simulation of Hydorous minerals"
- Tokyo Institute of Technology Umemoto "Simulation of Inner and Outer Core"
- Osaka Univ. Ishikawa "Development of Crystal Structure Prediction Method"
- NIMS Miyazaki "Development of Large-Scale First-Principles Dynamics Method"

Sub-issue D Leader
Kawashima Naoki (Univ. Tokyo, ISSP)
"Innovation of Methods for Computational Materials Science

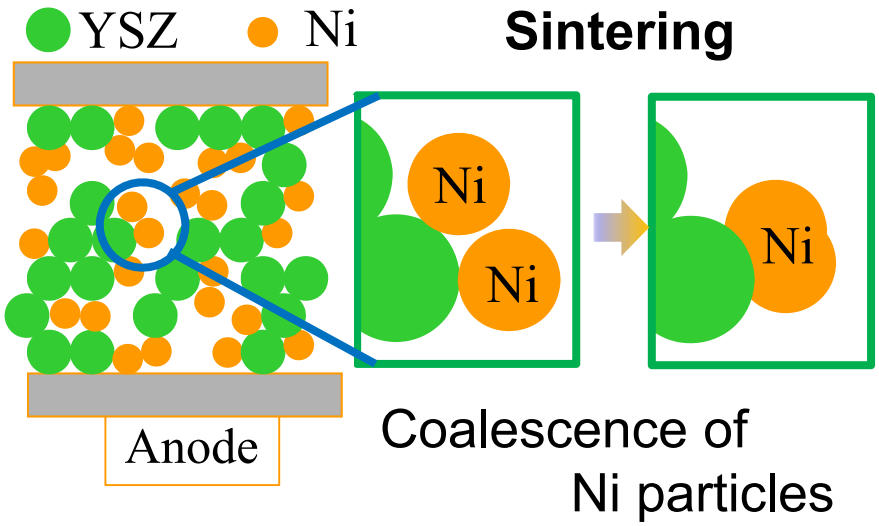
- Tsukuba Univ. Kuramashi "Application of Tensor Network Method to Particle Physics and Its Sophistication"
- Yokohama National Univ. Kosaka "Quantum Entanglement Network"

Members

Main Members： 11 Institutions, 56 Researchers
Cooperative Members： 56 Institutions, 111 Researchers

Solid Oxide Fuel Cell
Degradation by Ni Sintering

Degradation by Sintering



1) Ni Surface: H_2 Dissociation

⇒ Decrease

2) Triple Phase Boundary (TPB):

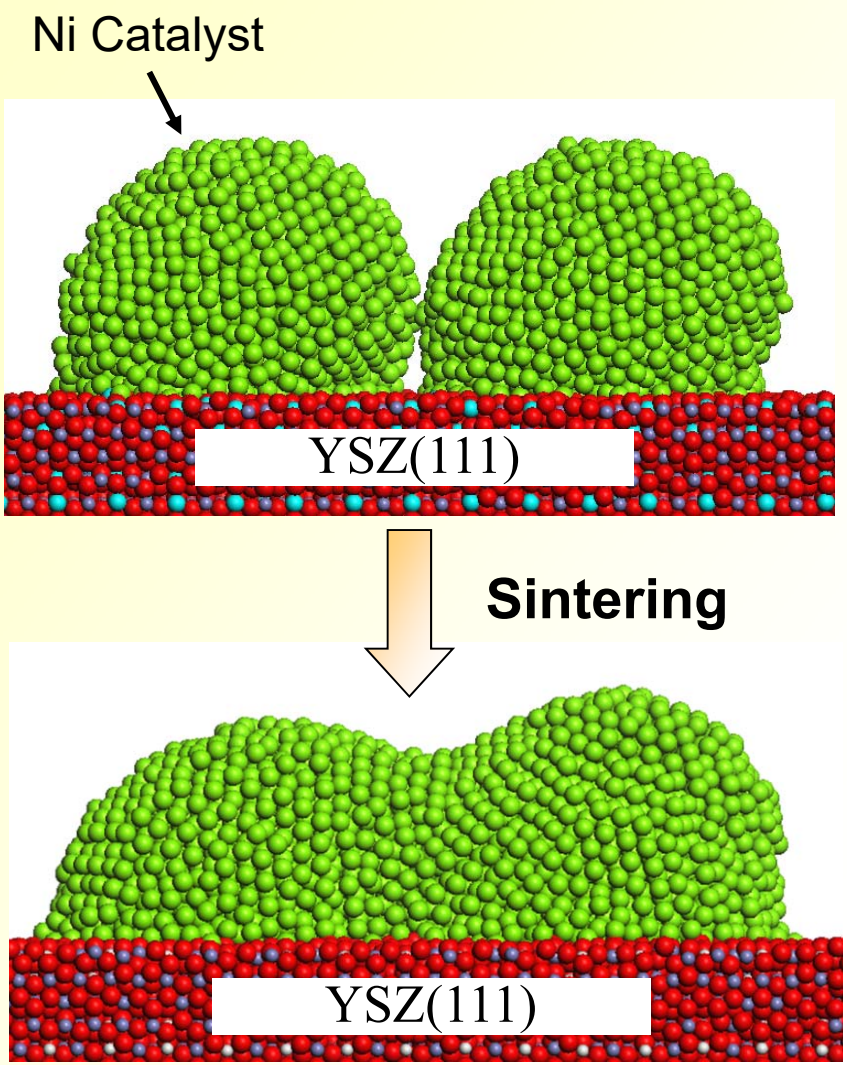
O^{2-} and H React

⇒ Decrease

3) Diffusion Path of H_2:

⇒ Decrease

Regular MD Simulation



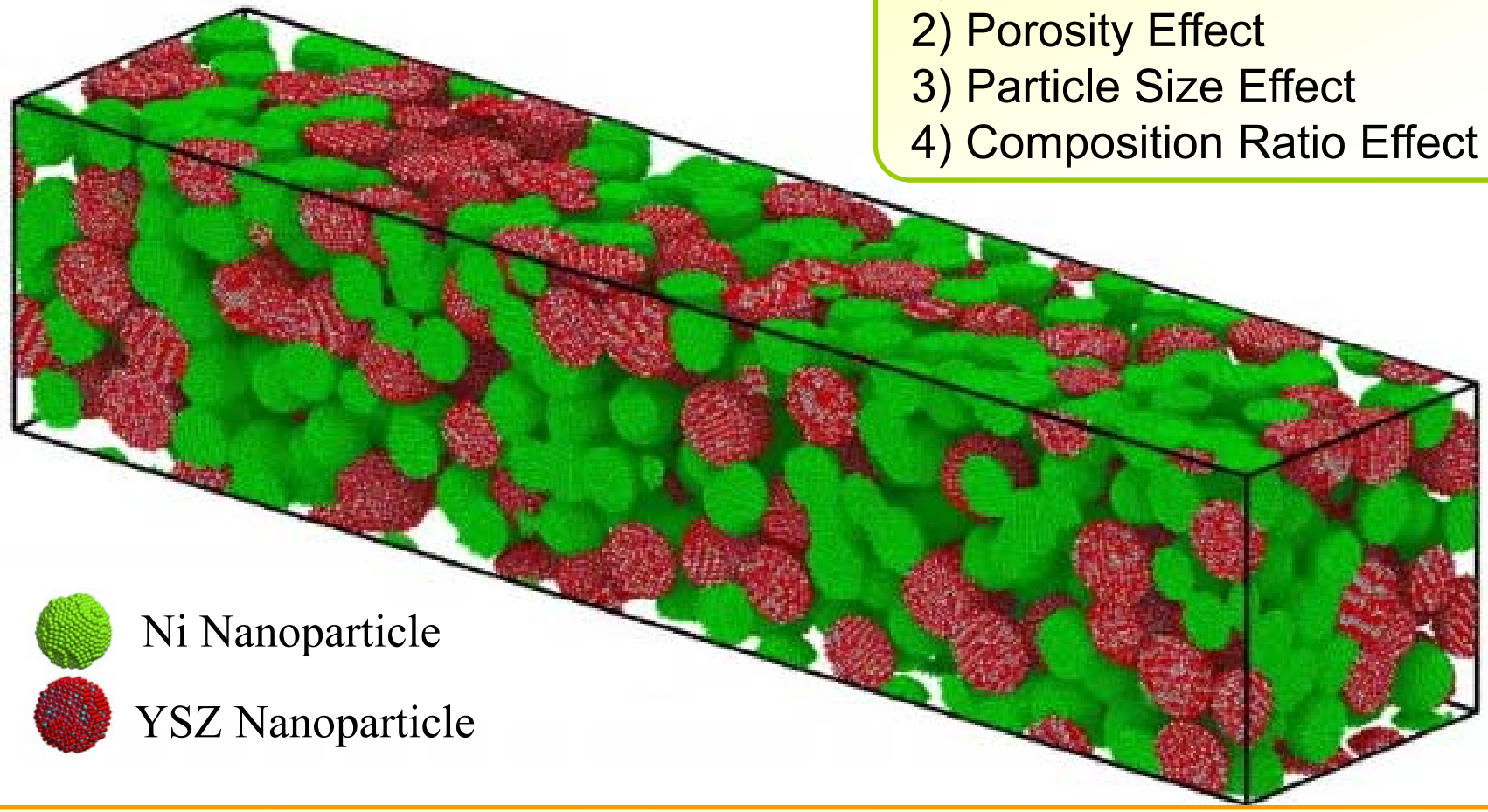
3 Million Atoms MD Simulation for Sintering

Kubo Laboratory
Multi-Physics Computational Science

YSZ: 60 Å

Change of Quality

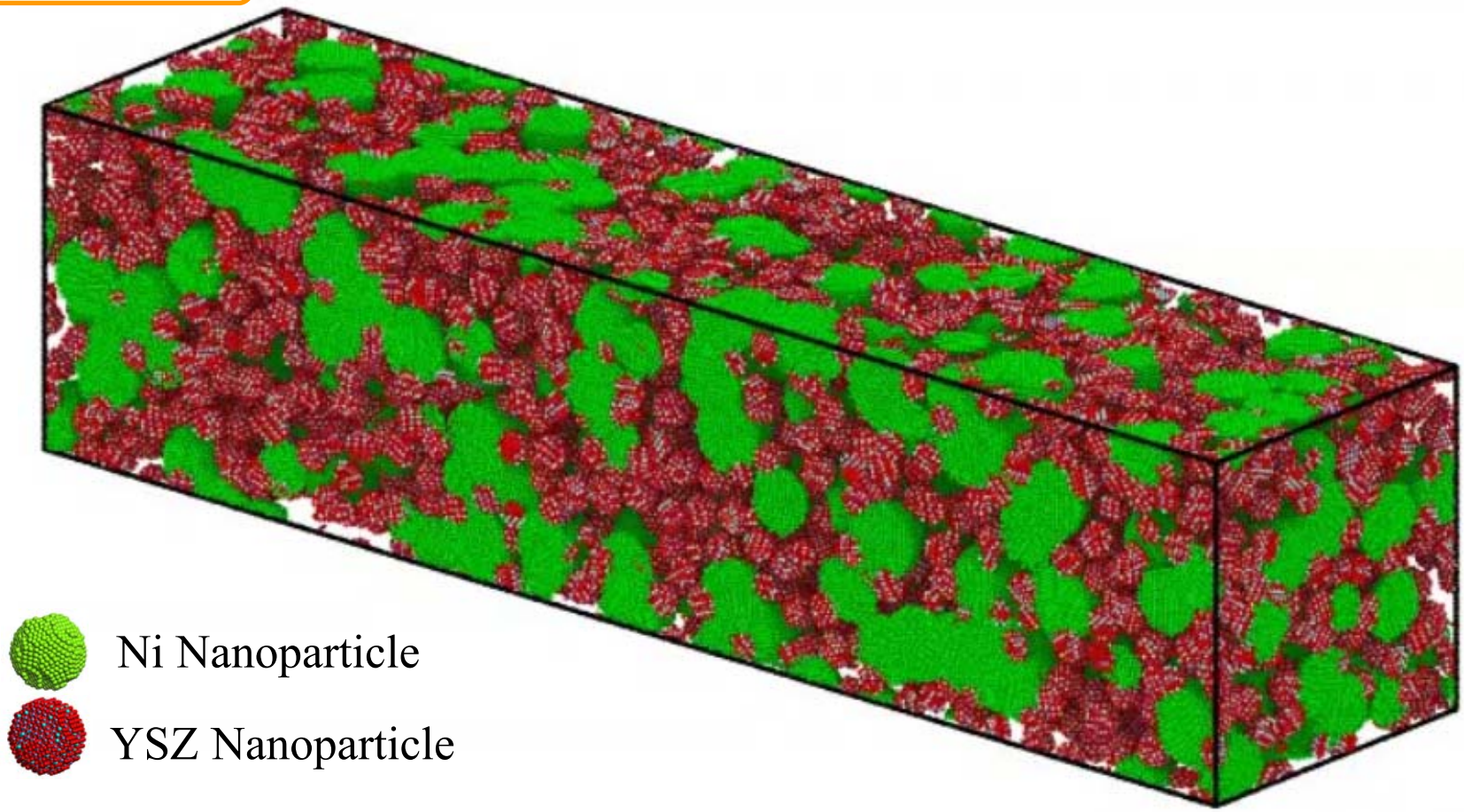
1) Framework Structure Effect
2) Porosity Effect
3) Particle Size Effect
4) Composition Ratio Effect



Sintering of Ni Particles Leads to **Dynamic Change of Porous Structure**, **which cannot be examined by Small Simulation**.

Effect of Particle Size
- Small Calculation does not Investigate it

YSZ: 20 Å

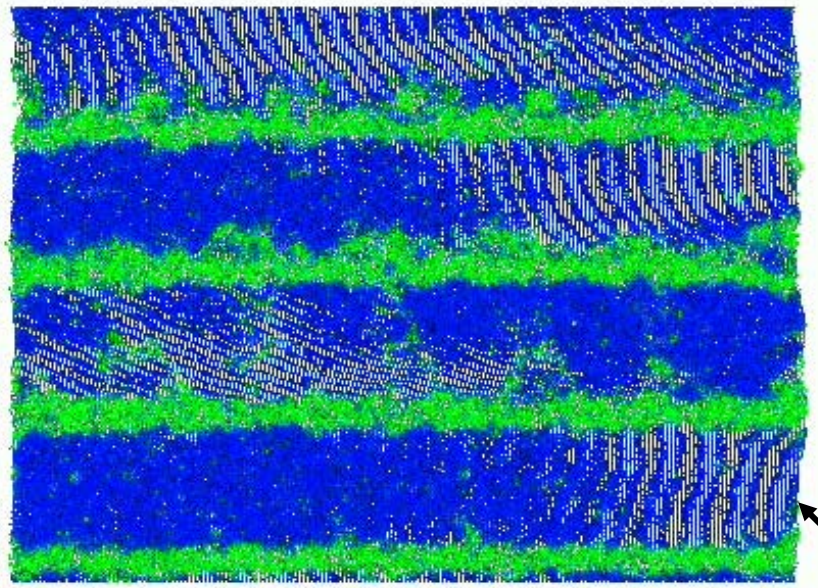


Sintering of Ni nanoparticles is not observed.

We clarified Effect of Particle Size, which can't be examined by Small Simulation

Destruction Process of
Plastic Bag in Convenience Store

Expand



Expand

6 Million Monomers Calculation

First, side is narrowed.

Second, hole is generated.

Small Model Cannot Reproduce

This Structure

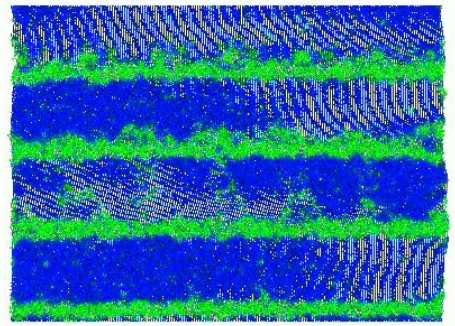
Amorphous Phase

Crystal Phase

Lamella Structure

Direct Comparison of Superlarge-Scale Molecular Dynamics with Electron Microscopy Image

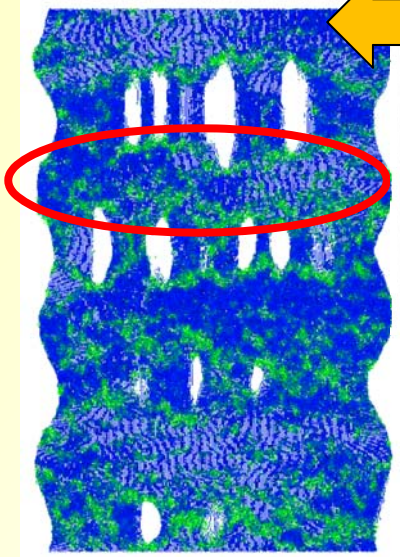
6 Million Monomers



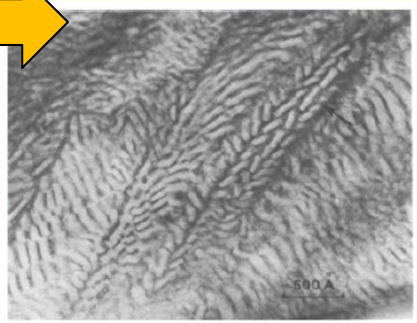
80 nm

Buckling

Electron Microscopy

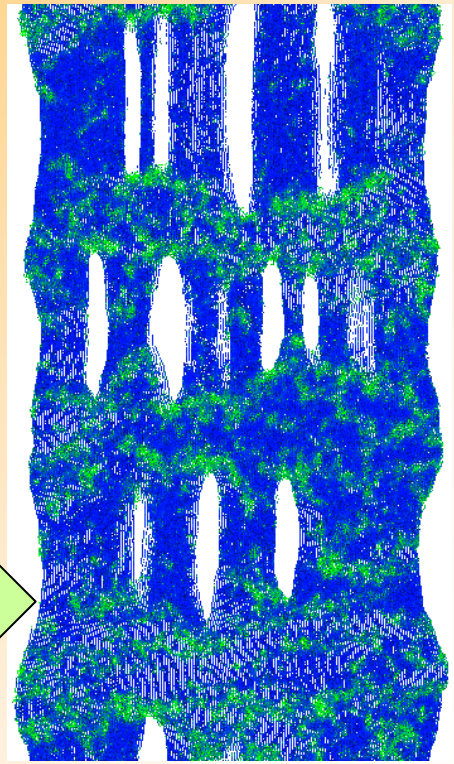


Strain=1.00



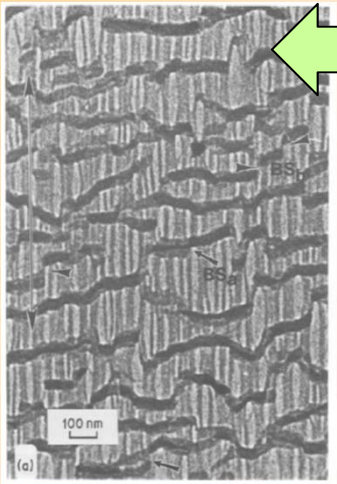
Burnay et al., J. Mater. Sci. 12, 1139 (1977).

Fragmentation



Strain=1.50

Electron Microscopy



Adams et al., J. Mater. Sci. 21, 2239 (1986).

Crystal Phase works as Hard Blocks.

After the Buckling,

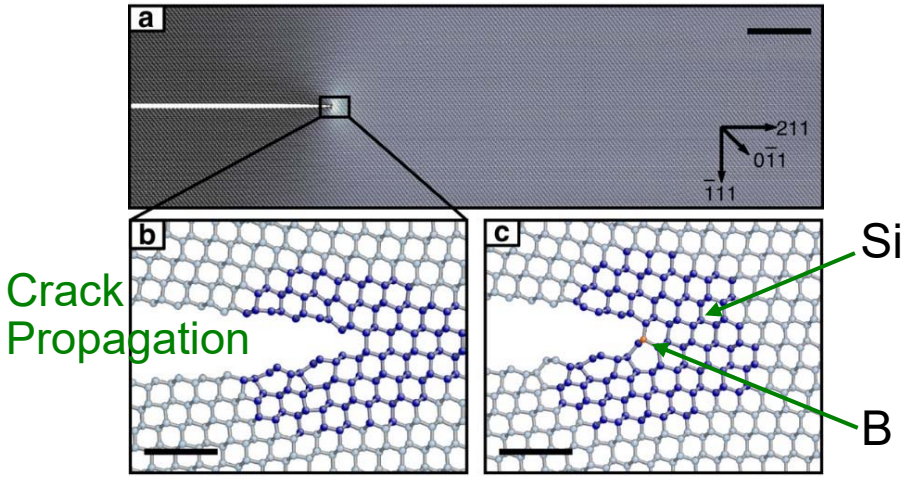
Fragmentation occurs

Fracture Process of Steels
- Crack Generation by Chemical Reaction

Kubo Laboratory
Multi-Physics Computational Science

Previous Large-Scale Molecular Dynamics Simulation

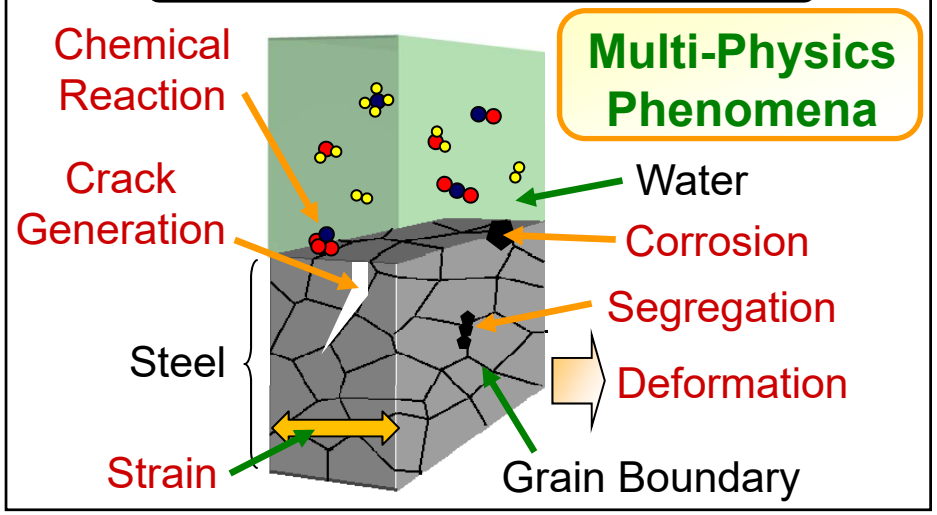
A. De Vita (King's College London)
Nature Commun., 4 (2013) 2441.



"Chemical reaction" and "Microstructure" are not considered

Our Targets

Fracture of Steels

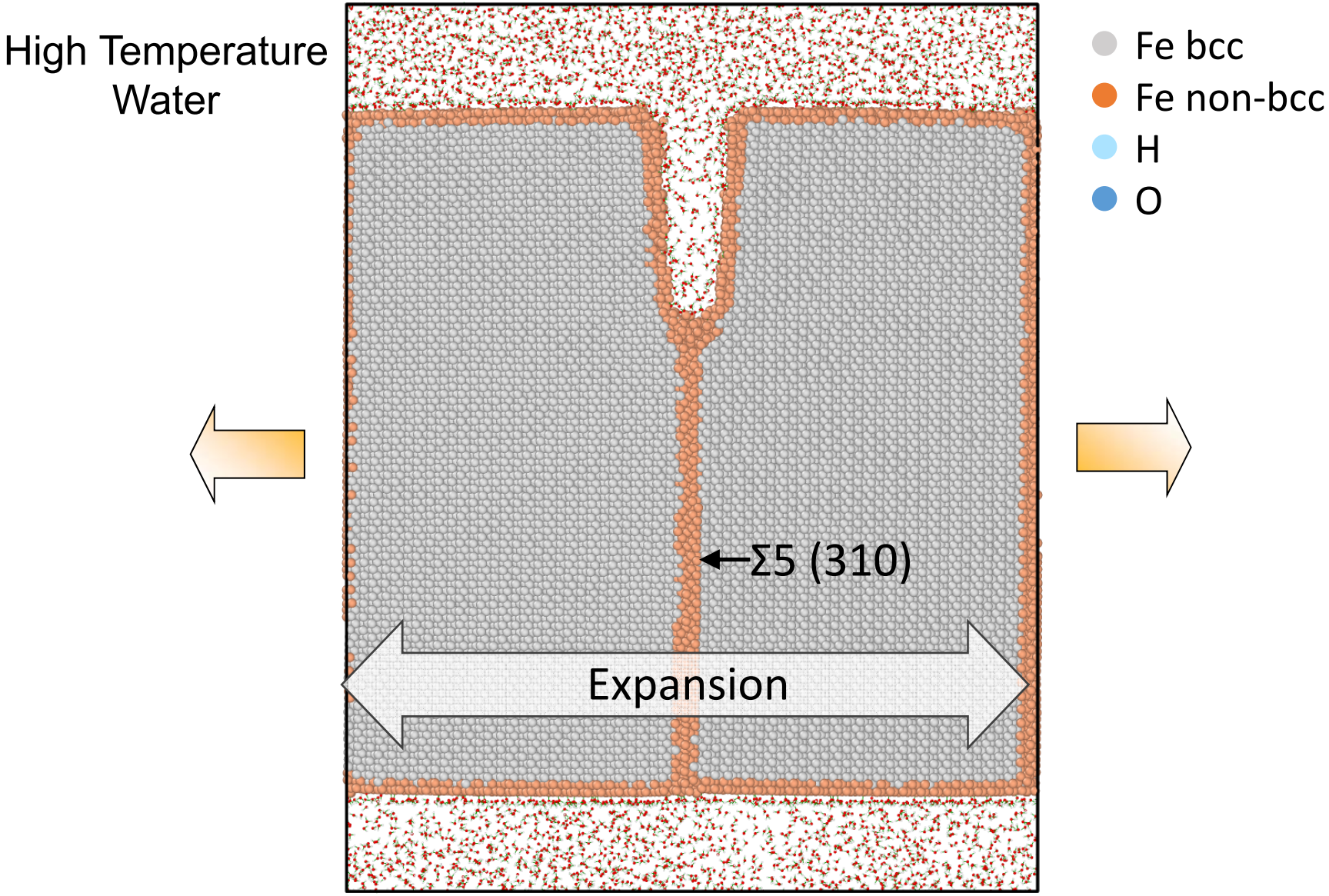


① Development of Molecular Dynamics Simulator for Chemical Reactions

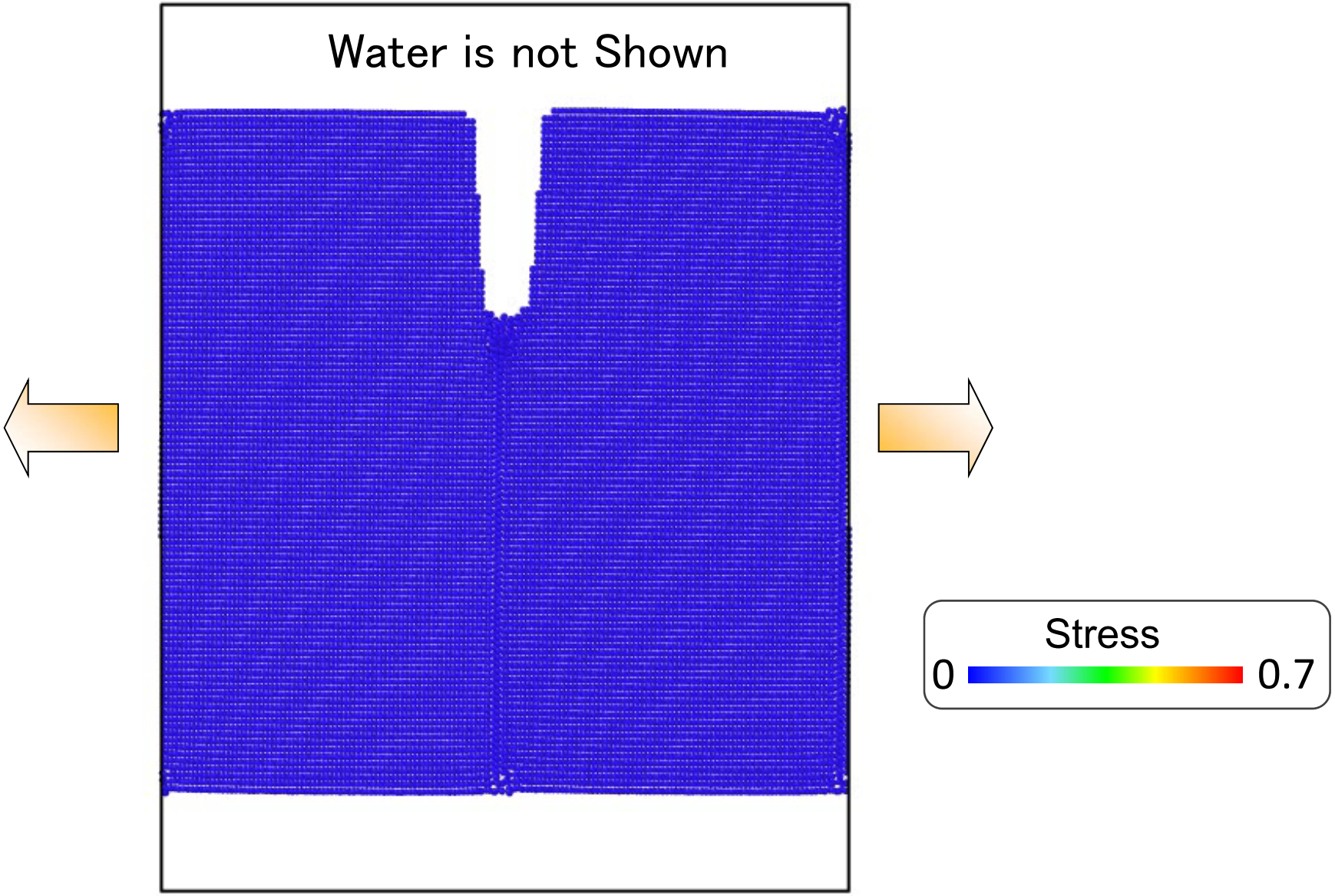
② Clarification of Chemical Reaction, Crack Generation, Corrosion, Deformation, etc.

③ Effect of External Factors (water, impurities, temperature, etc.)

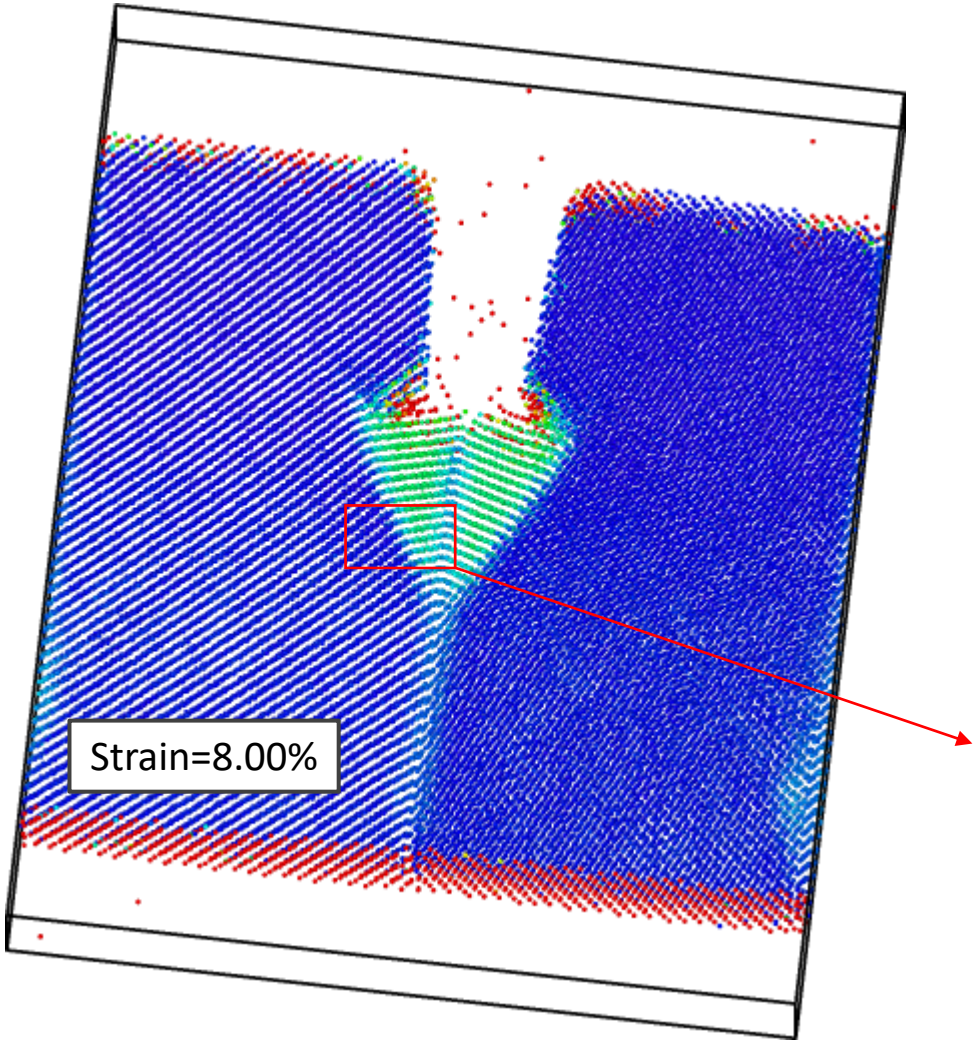
Destruction Process of Steel with Small Crack under Water Environment

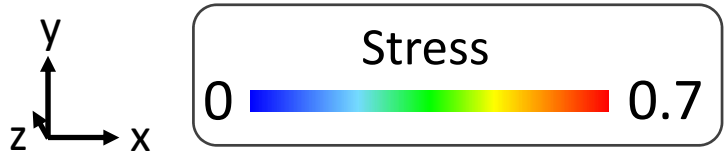


Destruction Process of Steel with Small Crack under Water Environment

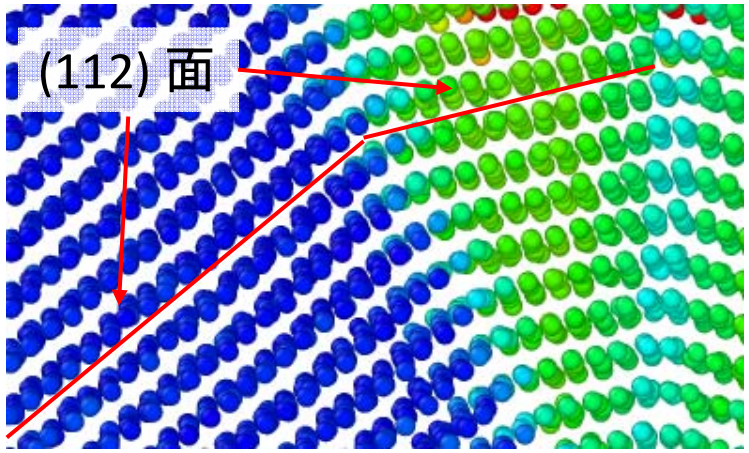


Plastic Deformation Mechanism of Steels with Pre-Crack





[111] / {112} Twin Microstructure

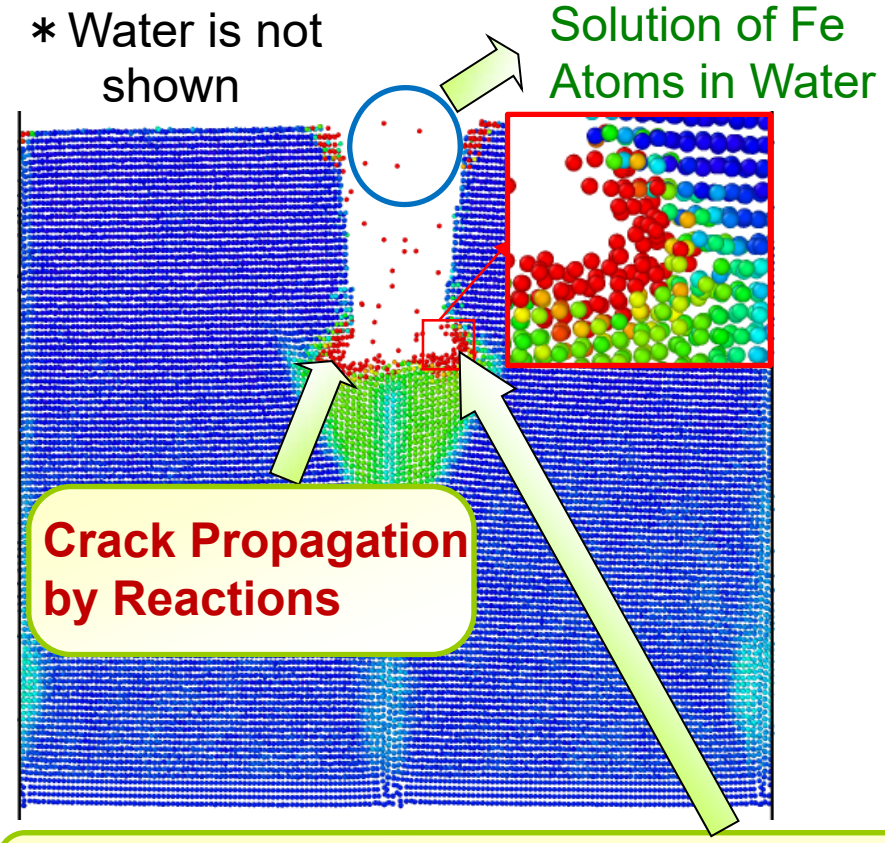


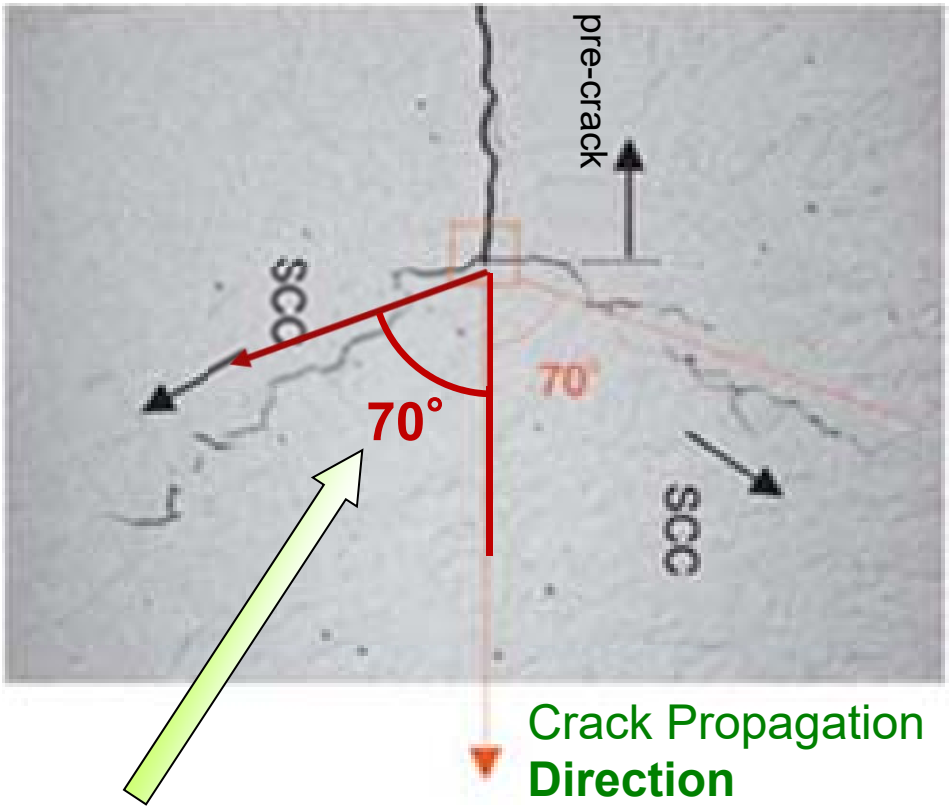
Generation of Twin Microstructure Leads to Generation of Small Crack

Crack Propagation Mechanism
from Pre-Crack of Steels

Stress Corrosion Cracking of Steels under Water

Experimental Observation - Stainless Crack



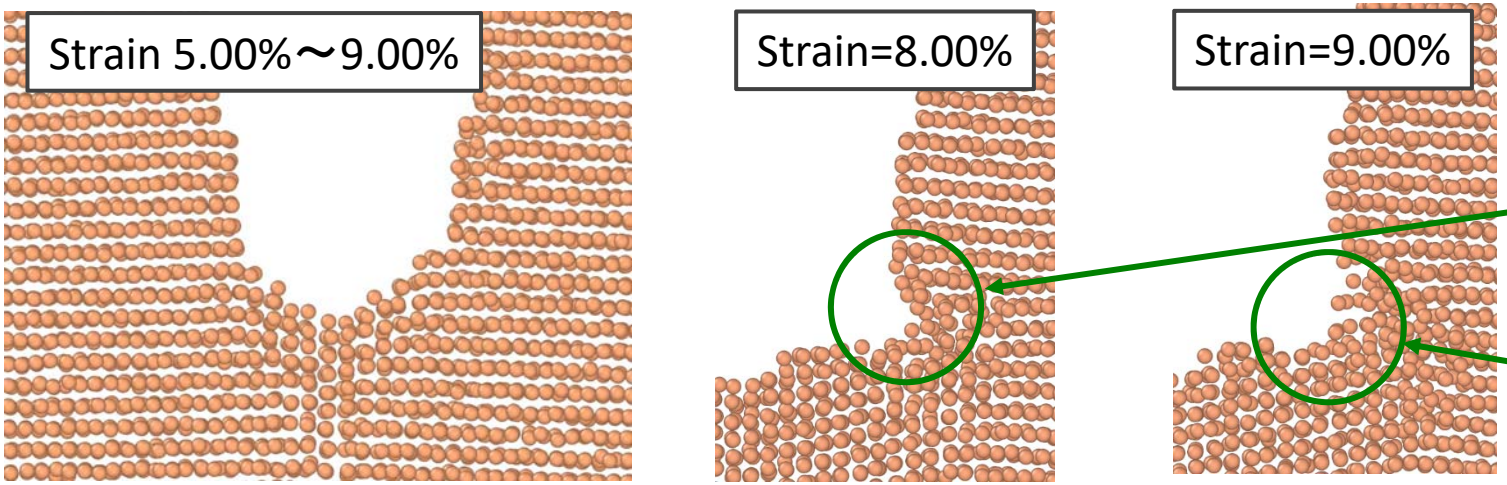


In both Simulation and Experiment,
New Crack Generation in 70° Direction was found.

[Experiment] T. Terada, aesj2007s meeting, 391, Stress Corrosion Cracking of Stainless Steel under Deaerated High-Temperature Water

Crack Propagation Mechanism
from Pre-Crack of Steels

Vacuum Condition



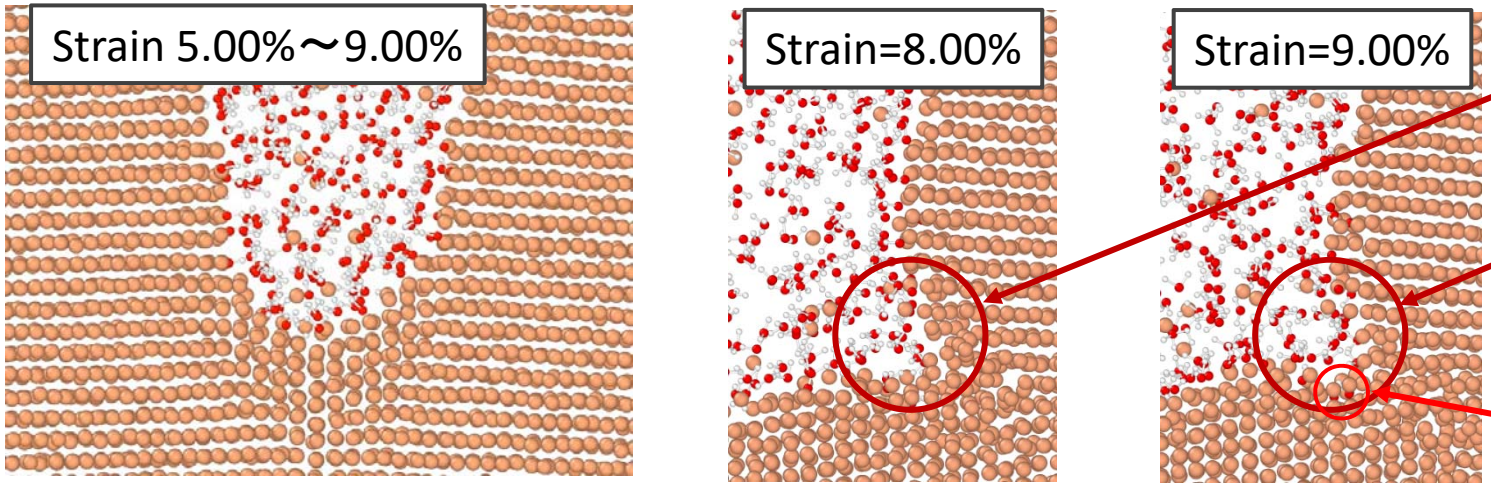
Fe
H
O

Crack Generation by Twinning

Structure Relaxation

Crack Disappears

Water Environment



Crack Generation by Twinning

Generation of Fe-OH Bond

No Structure Relaxation

Chemical Reactions Inhibit the Structure Relaxation of Crack,
Leading to Advancement of Crack Propagation

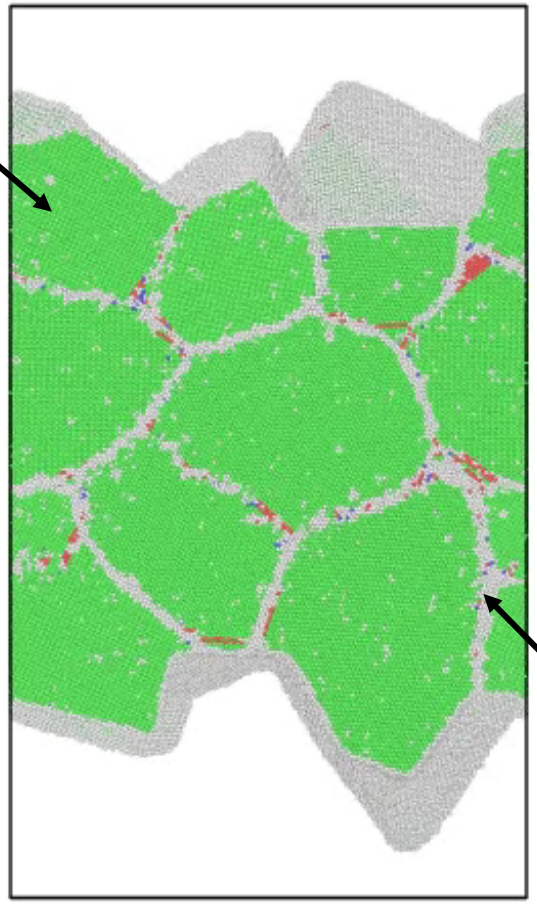
Chemical Reactions Assisted
Stacking Fault Generation in Ni Polycrystalline

Kubo Laboratory
Multi-Physics Computational Science

Vacuum | **Water Environment**

2 Million Atoms

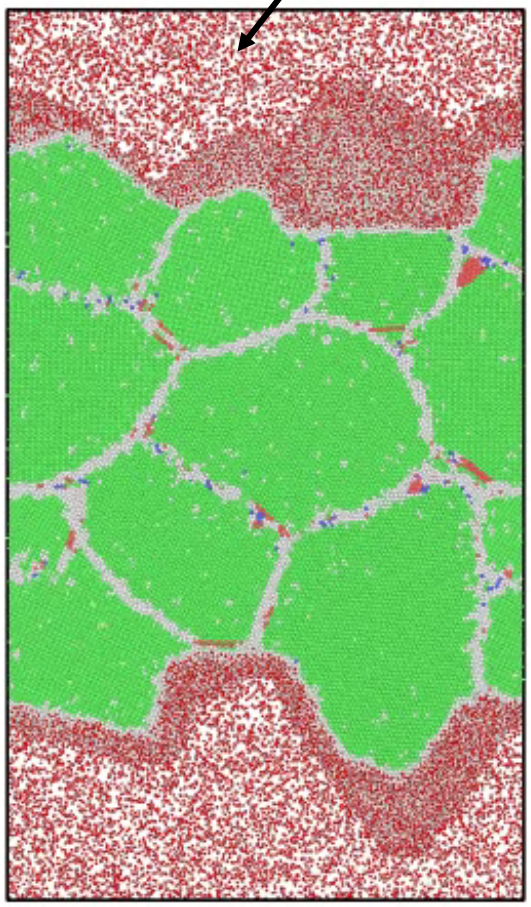
Ni



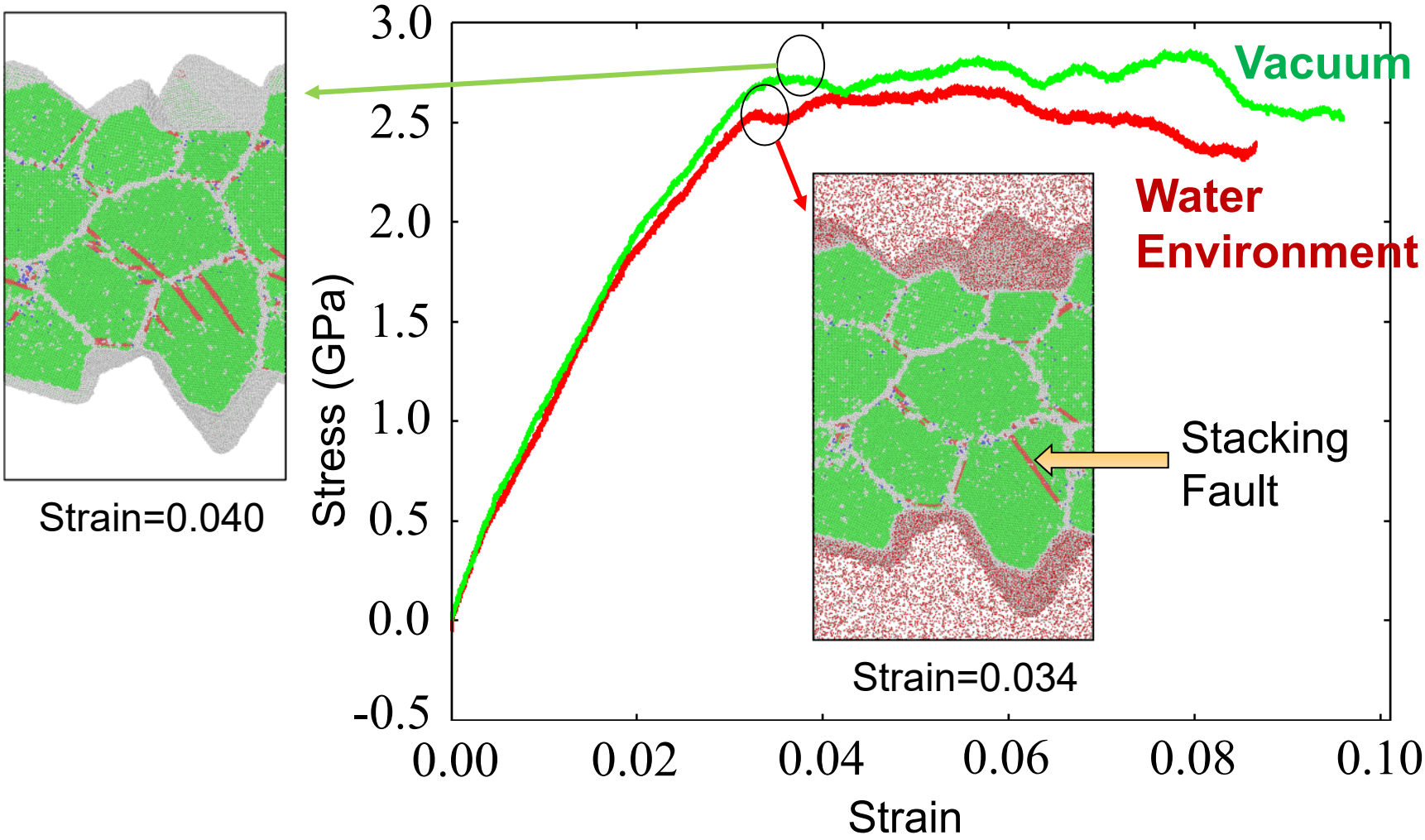
Grain Boundary

Extension

Water



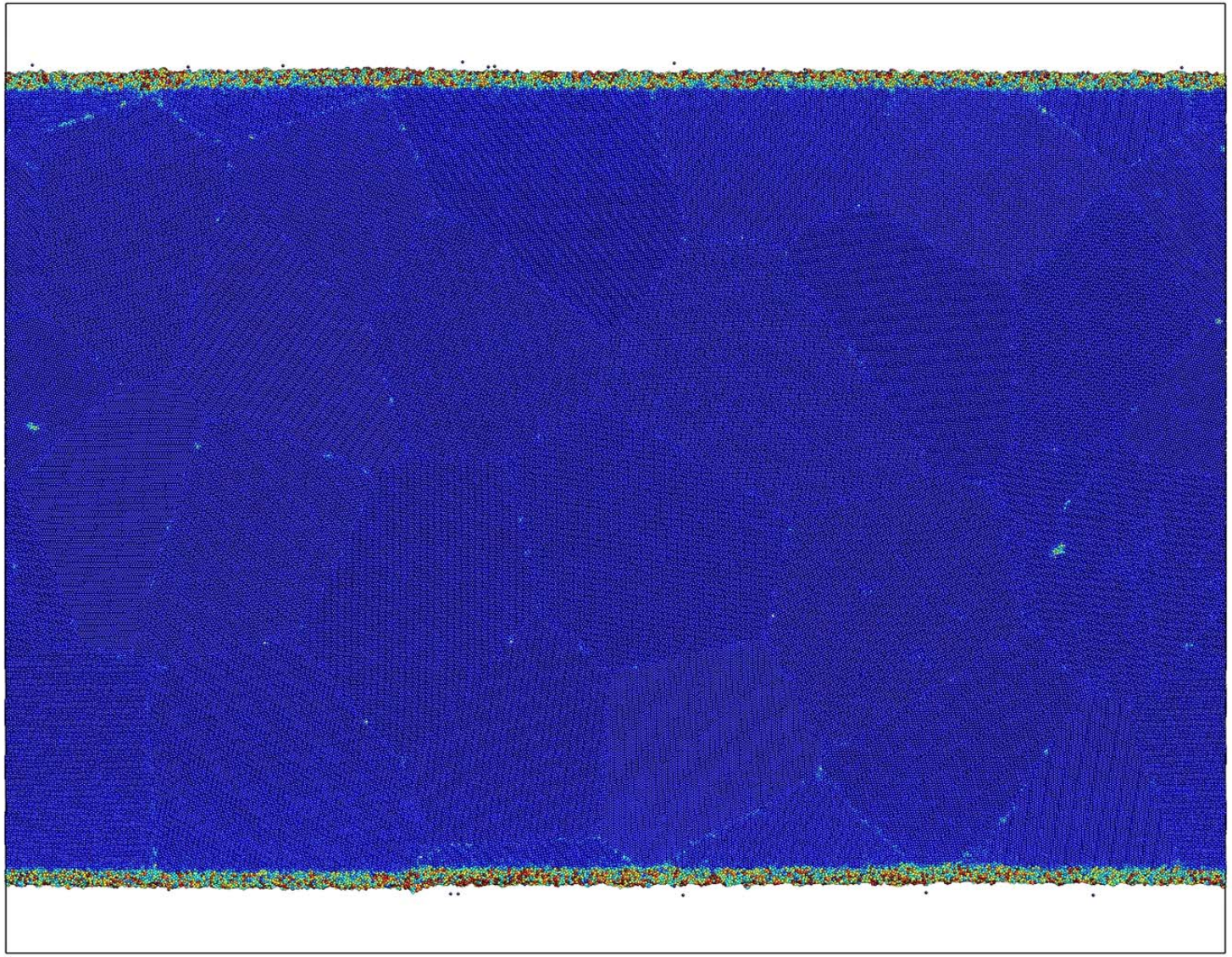
Chemical Reactions Affect
Stress-Strain Curve in Ni Polycrystalline



Stacking fault generation and yielding point is earlier
in water environment

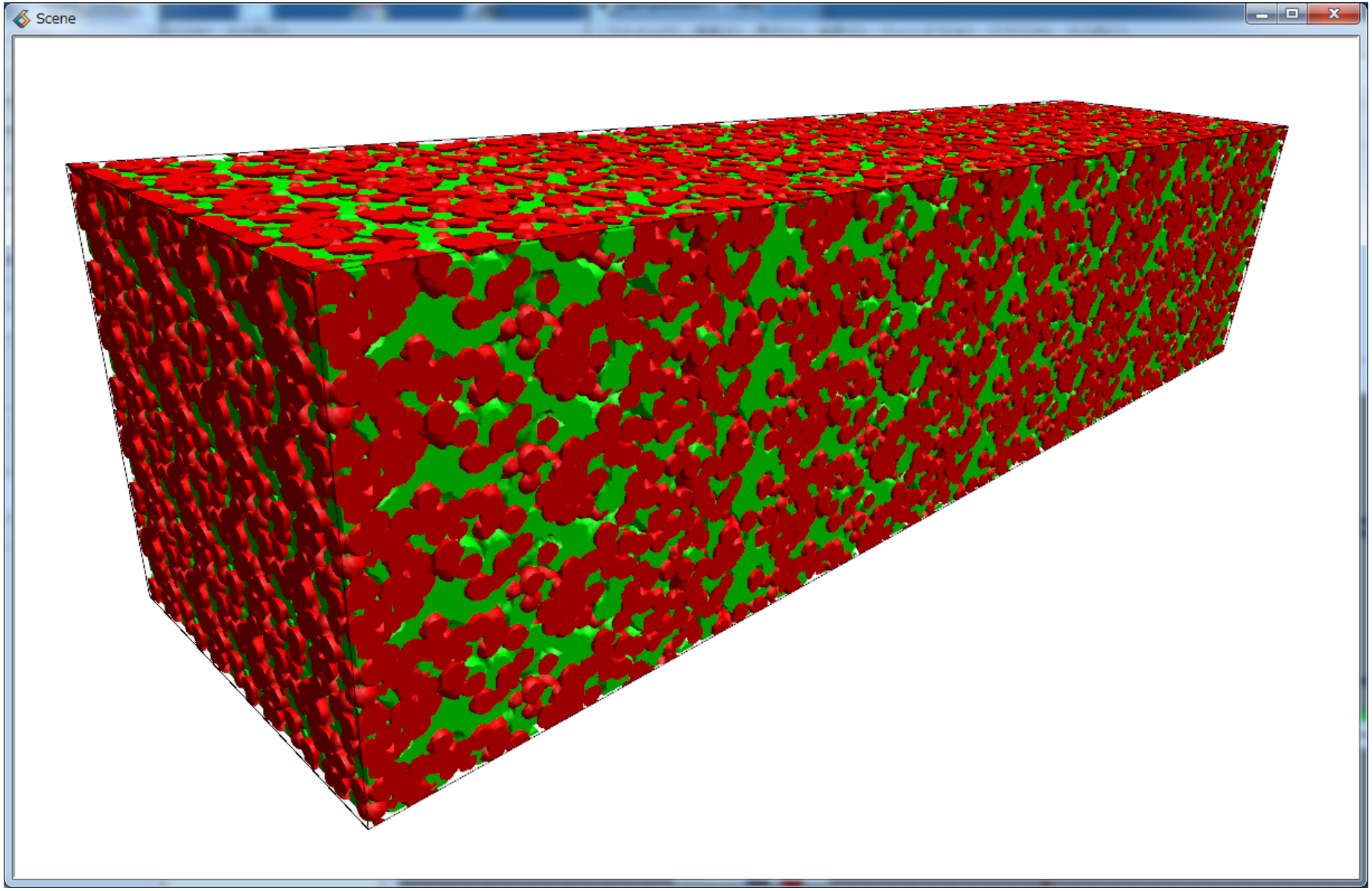
2 Million Atoms → 10 Million Atoms Simulation for Chemical-Reaction-Assisted Stacking Fault Generation of Ti Polycrystalline

Kubo Laboratory
Multi-Physics Computational Science

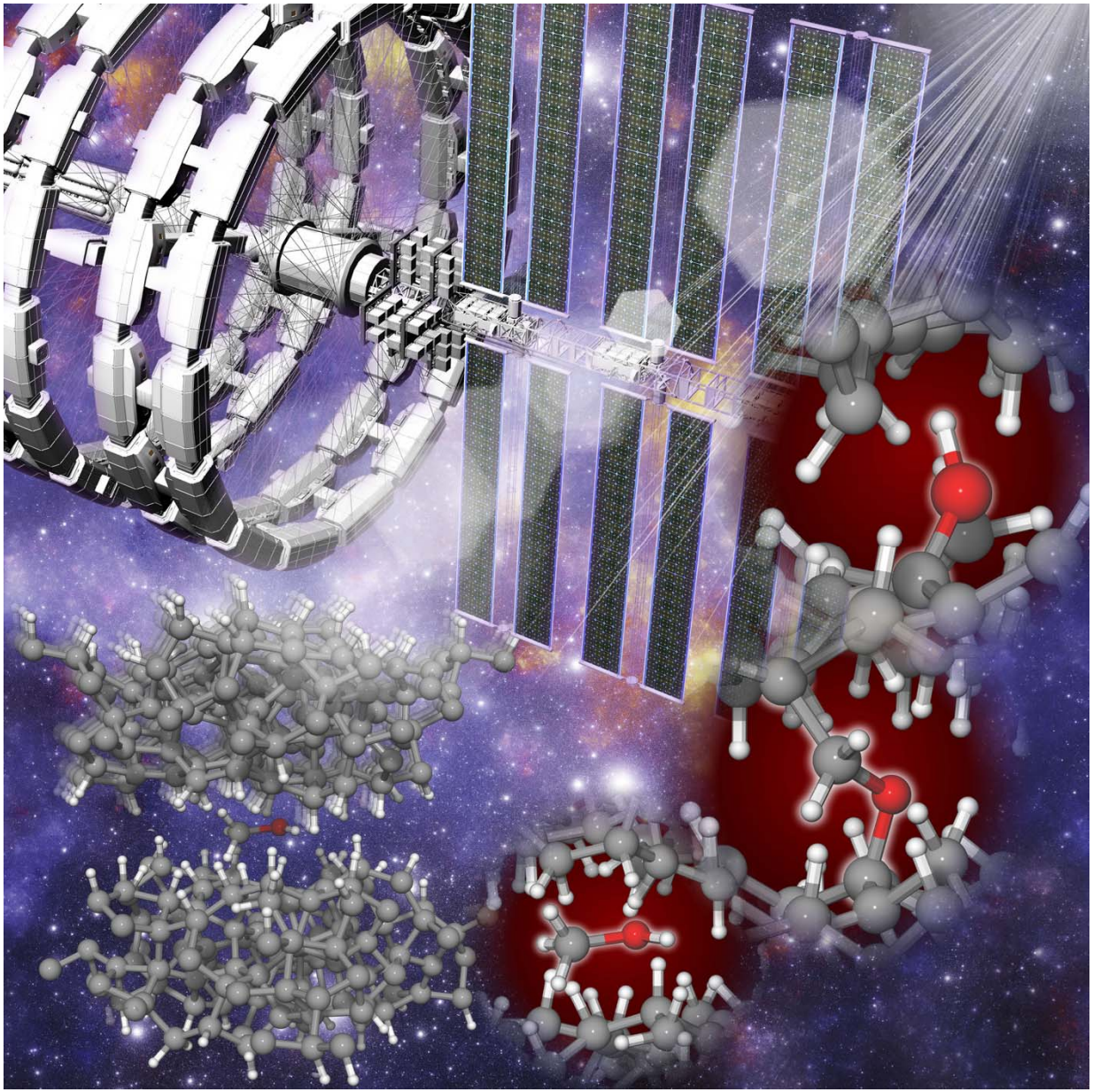


10 Million Atoms → 200 Million Atoms Simulation for Ni/YSZ Sintering

Kubo Laboratory
Multi-Physics Computational Science



Conceptual Figure
Design of Massive Equipment from Atomic-Scale



Cover Picture

K. Hayashi, M. Kubo et al.,
Faraday Discussions.

Chemical reactions at Nano-scale affect Macro-scale Material Property.

Sendai

Masamune Date
(1567-1636) - Samurai

Statue of Masamune Date

Aoba Castle (1601)

Sendai Old
Samurai Residence

Osaki Hachimangu
Shrine (1604)

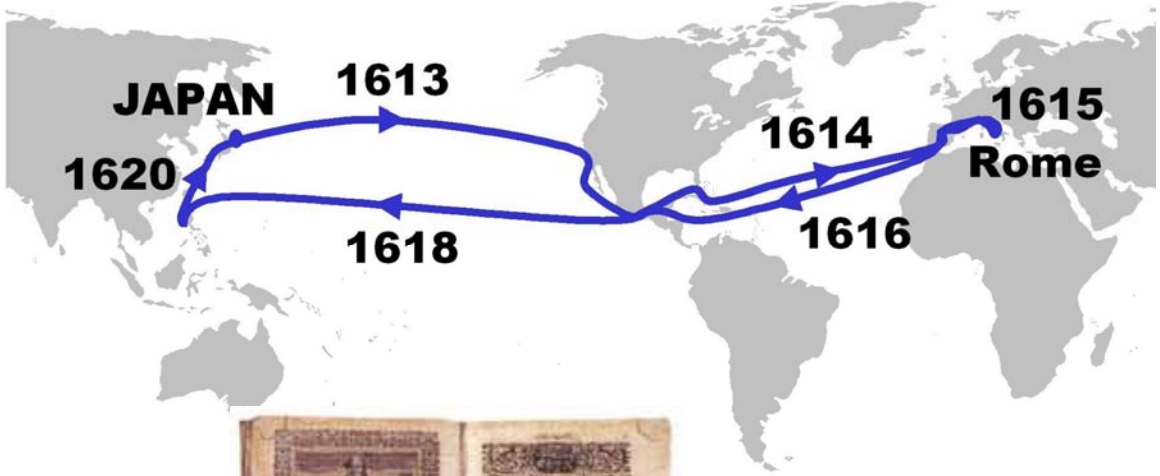
Tohoku University (1907-)
(3rd Public Univ.)

Ambition of Masamune Date for Dominating Japan

Samurai (Retainer)
- Tsunenaga Hasekura

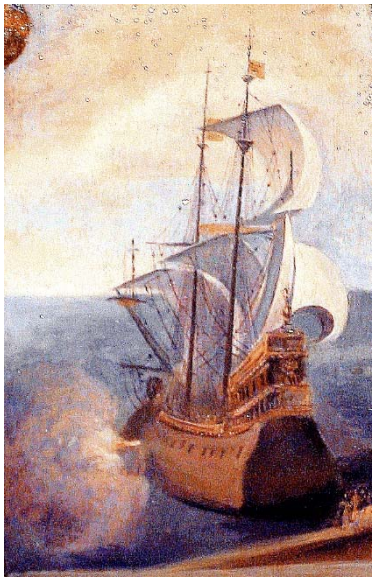
Portrait at Rome

Travel Route of Tsunenaga Hasekura
(Objective is Military Alliance to be SHOGUN)



He met the
King of Spain

Ship - San Juan Bautista



He met the Pope at Rome

Supercomputer in IMR, Tohoku University

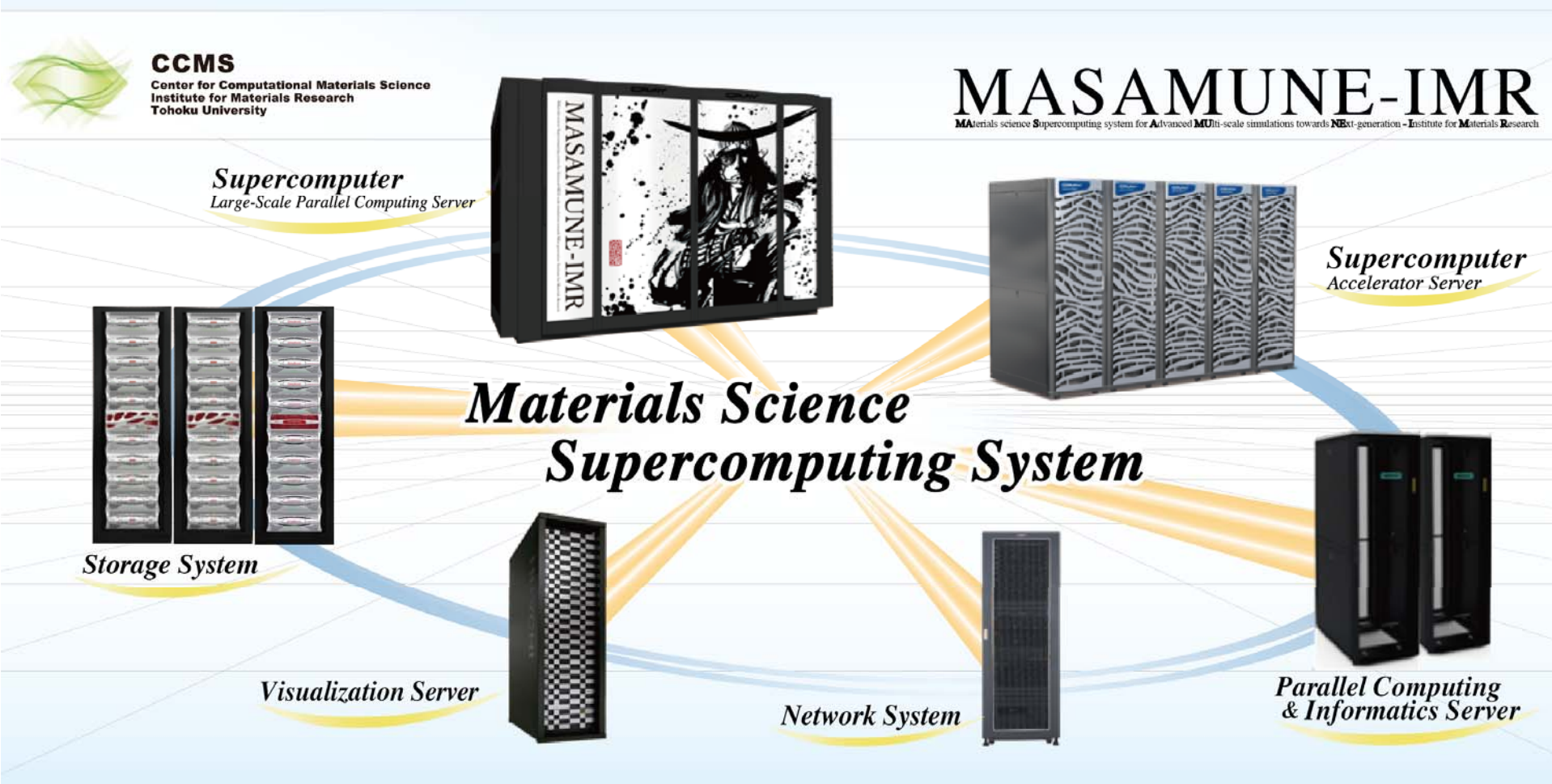
MASAMUNE-IMR

MAterials science **S**upercomputing system
for **A**dvanced **MU**lti-scale simulations towards **NE**xt-generation
- **I**nstitute for **M**aterials **R**esearch

Starts Running on
August 1st, 2018

Supercomputer in IMR, Tohoku University

MASAMUNE-IMR



Supercomputer

Cray XC50-LC & Cray CS-Storm 500 GT

Computing Performance 3.03 PFLOPS

CPU: 11,592 Core

GPU: 1,484,800 Core

TV Broadcasting of Our Simulation Results and New Supercomputer MASAMUNE-IMR

NHK – Nippon Television (July 12th, 2018)

Sendai Television (July 12th, 2018)

East Japan Television (September 20th, 2018)