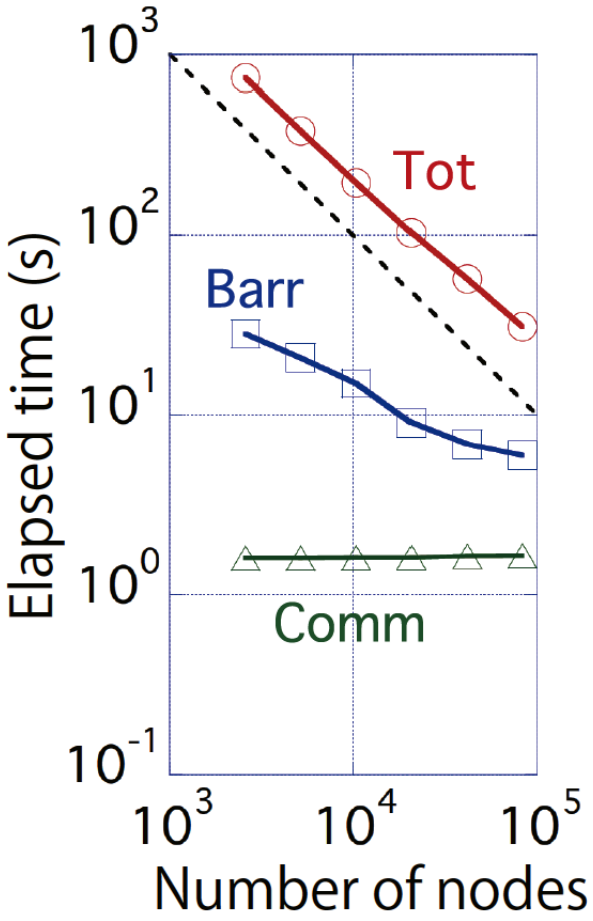
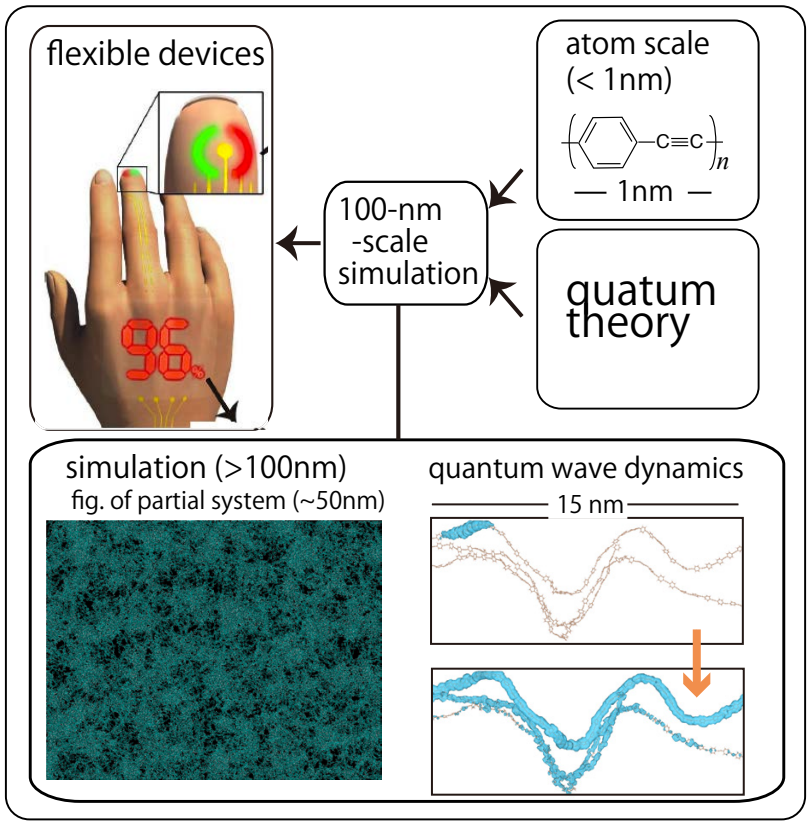
HPC Challenge in Material Science:
100-nano-meter-scale electronic state calculation for flexible device materials

T. Hoshi (Tottori U)

1. Algorithm



2. Application



Recent co-workers

- (ex-)student
  H. Imachi (D. Thesis, Mar. 2017; now in Preferred Networks Inc.)
  A. Kuwata, K. Kakuda, K. Oohira, Y. Abe
- Organic device material (industry)
  M. Ishida, S. Nishino (Sumitomo Chemical Co.)
- Organic device material (experiment)
  H. Matsui (Yamagata U.)
- HPC/numerical linear algebra
  Y. Yamamoto (UEC), T. Fukaya (Hokkaido U)
- Theory (quantum mechanics)
  T. Fujita (Inst. Mol. Sci.), Y. Mochizuki (Rikkyo U)
- Code tuning
  K. Kumahata, M. Terai, K. Miyamoto, K. Minami and F. Shoji (RIKEN R-CCS)

Acknowledgement for discussion
T. Imamura (RIKEN R-CCS), K. Nakajima (U Tokyo)

Projects

- The priority Issue 7 (New Device and Material) for the post-K computer;
  Subgroup G3 'Development of fundamental parallel algorithm'
  http://www.damp.tottori-u.ac.jp/~hoshi/post-K-algo.html
- other projects by japanase govement (KAKENHI)

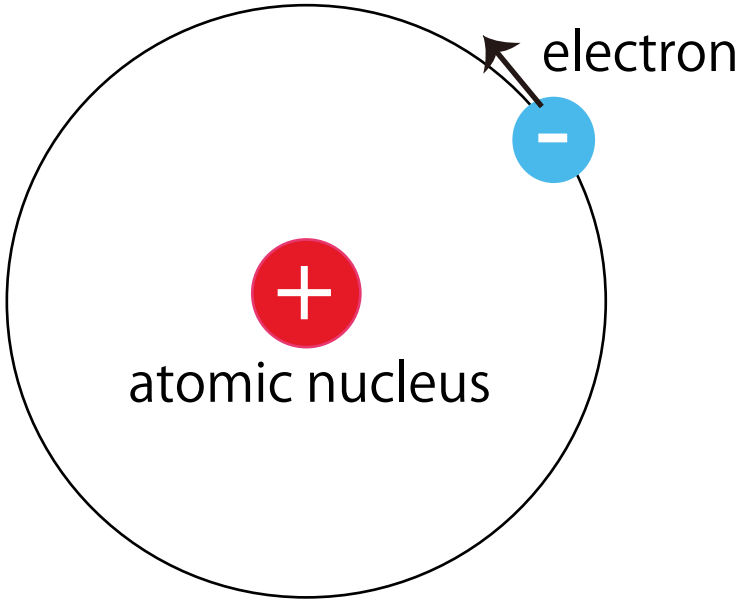
Tutorial on quantum material simulation (1/3)

Quantum mechanics (1925~)

- Electron is treated as 'wave'
  or the solution of Schroedinger equation
- Nowadays, small systems can be computed by non-experts
  with free or commercial software on laptop PC

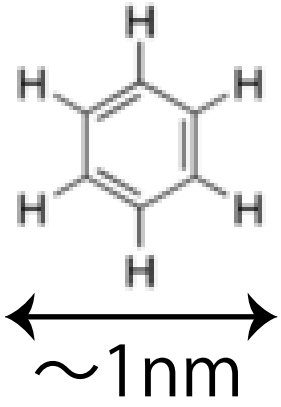
Before quantum mechanics
→electron as 'ball'

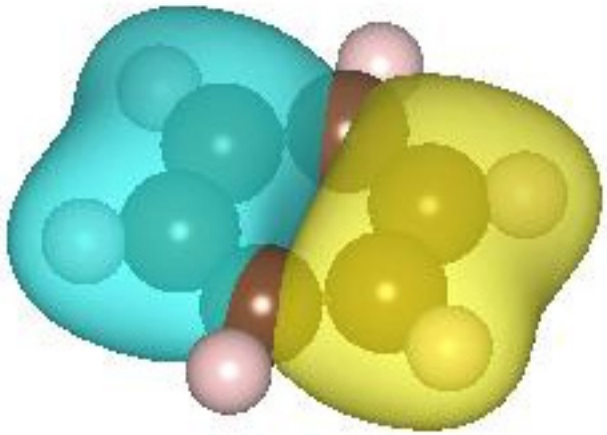
eg. hydrogen atom



In quantum mechanics
→electron as 'wave' (scalar function)

eg. benzene molecule (C_6H_6)

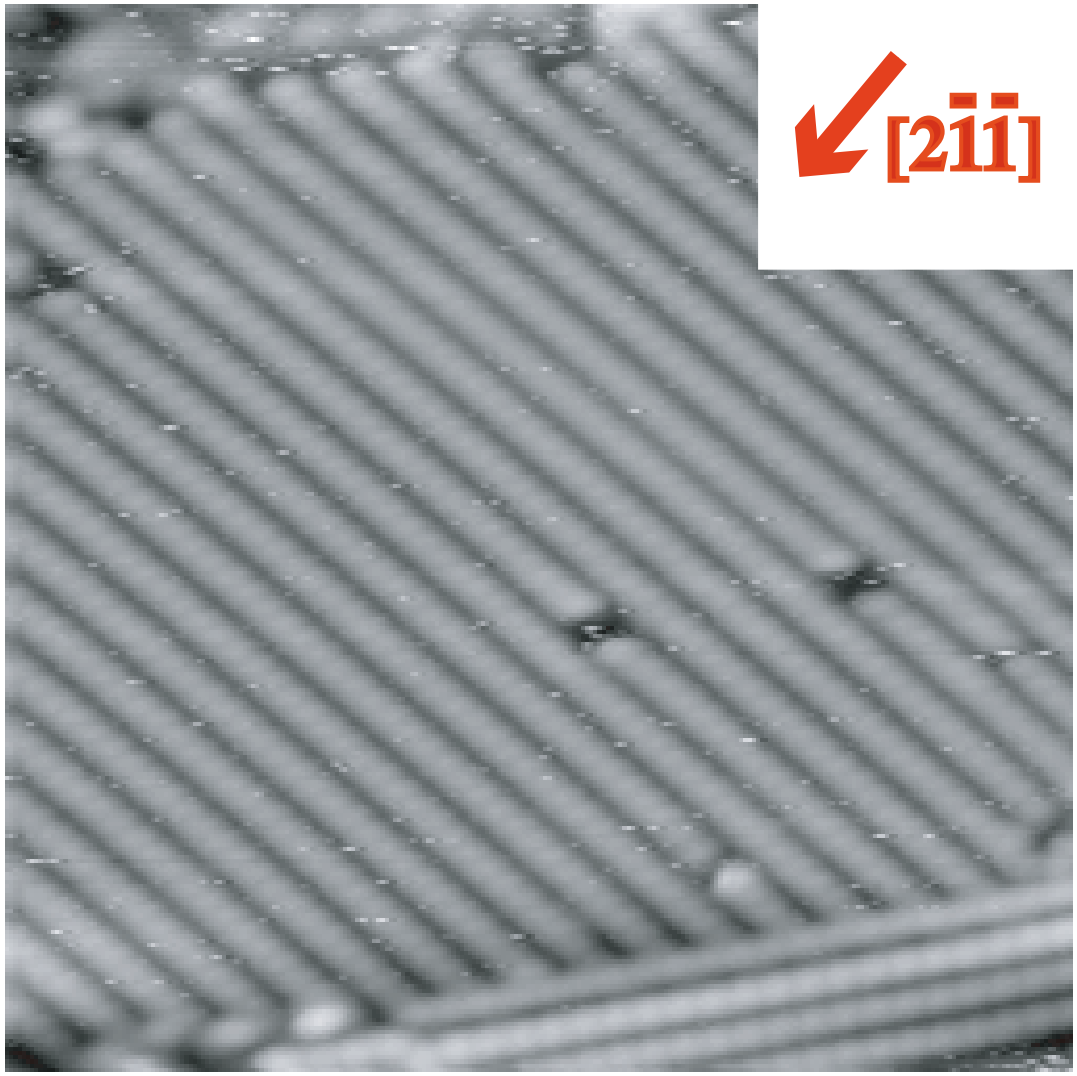




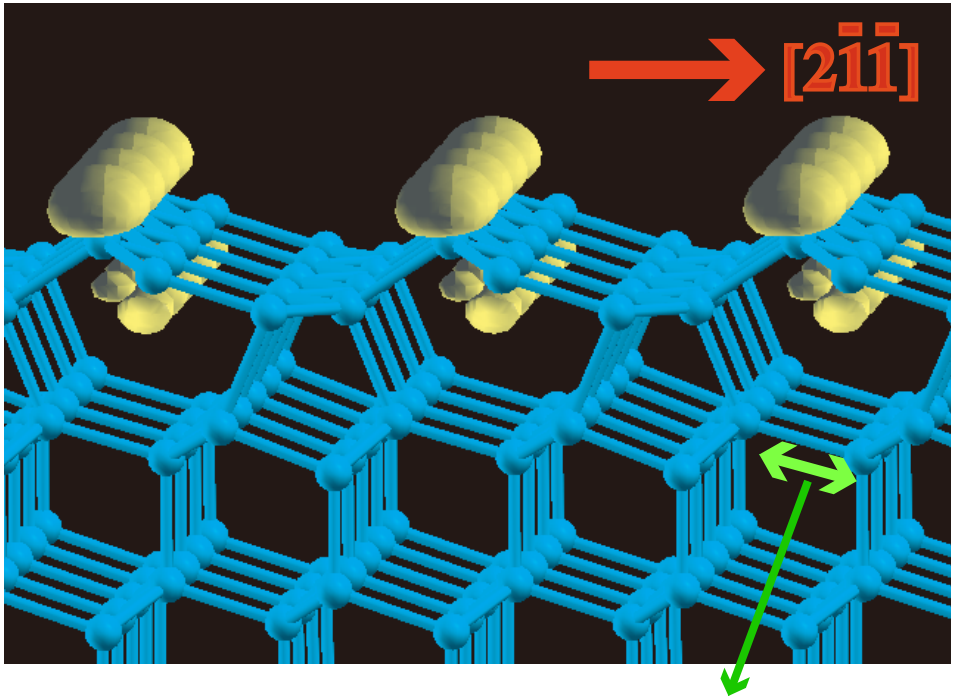
Tutorial on quantum material simulation (2/3)

Observation of electronic wavefuncion
by scanning tunneling microscope (STM)

15 nm



Calculated wavefunction



(inter-atomic distance) = 0.24 nm

← silicon surface ((111)-2x1 type)
Mera et al.,
Ultramicroscopy 42-44, 915 (1992)

Tutorial on quantum material simulation (3 / 3)

Numerical aspects

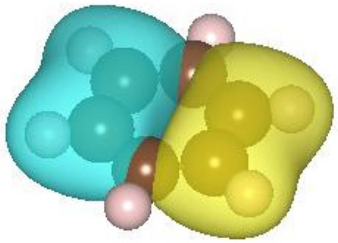
continuum space representation

$\phi(\boldsymbol{r})$ electronic wave

discretized vector

$\boldsymbol{u} = (u_1, u_2, \ldots u_M)^{\mathrm{T}}$

size = M



A typical discretization method (today's topic)
: linear combination
of by localized (atomic) basis functions

$$\phi(\boldsymbol{r}) := \sum_{j}^{M} u_j f_j(\boldsymbol{r})$$

$M \propto N$

bases # atoms

Note: Other types of basis functions are also used,
such as plane wave, mesh grid ...

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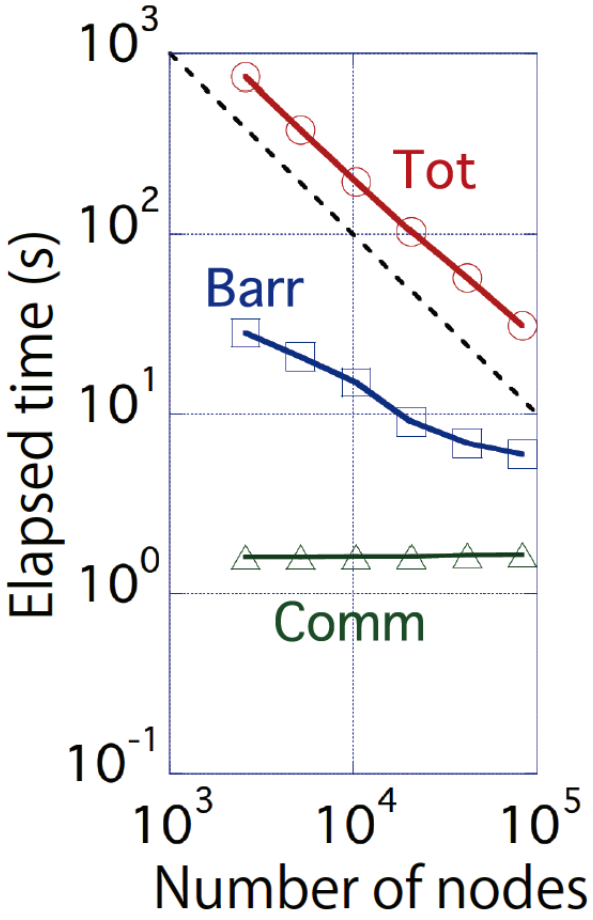

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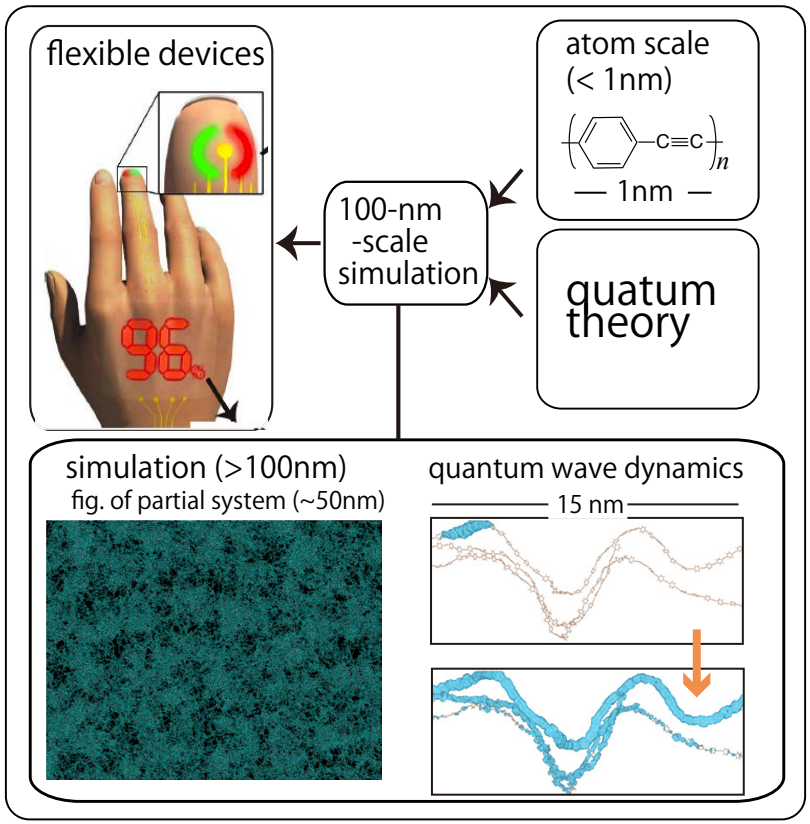
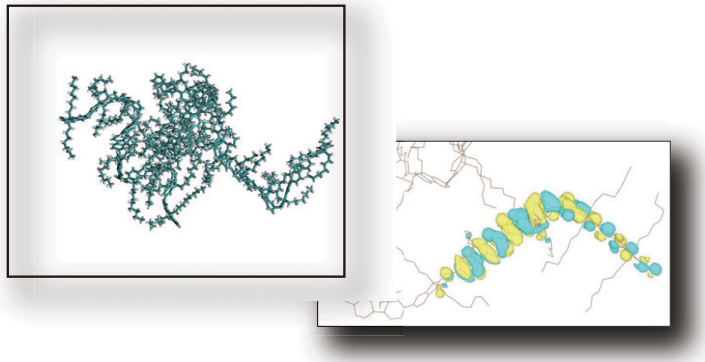


Overview

Application-Algotirhm-Architecture co-design

Application : Quantum material simulationn



$$Hy = \lambda Sy$$

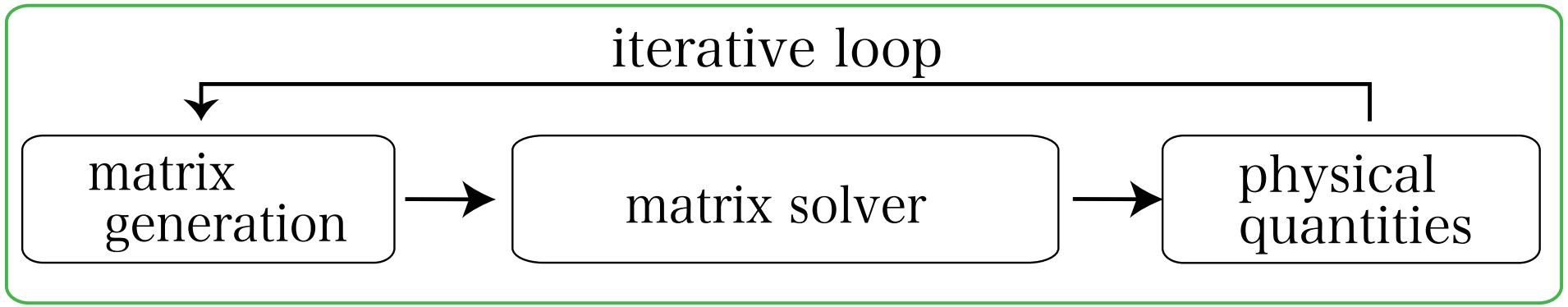
Algorithm : Numerical linear algebra

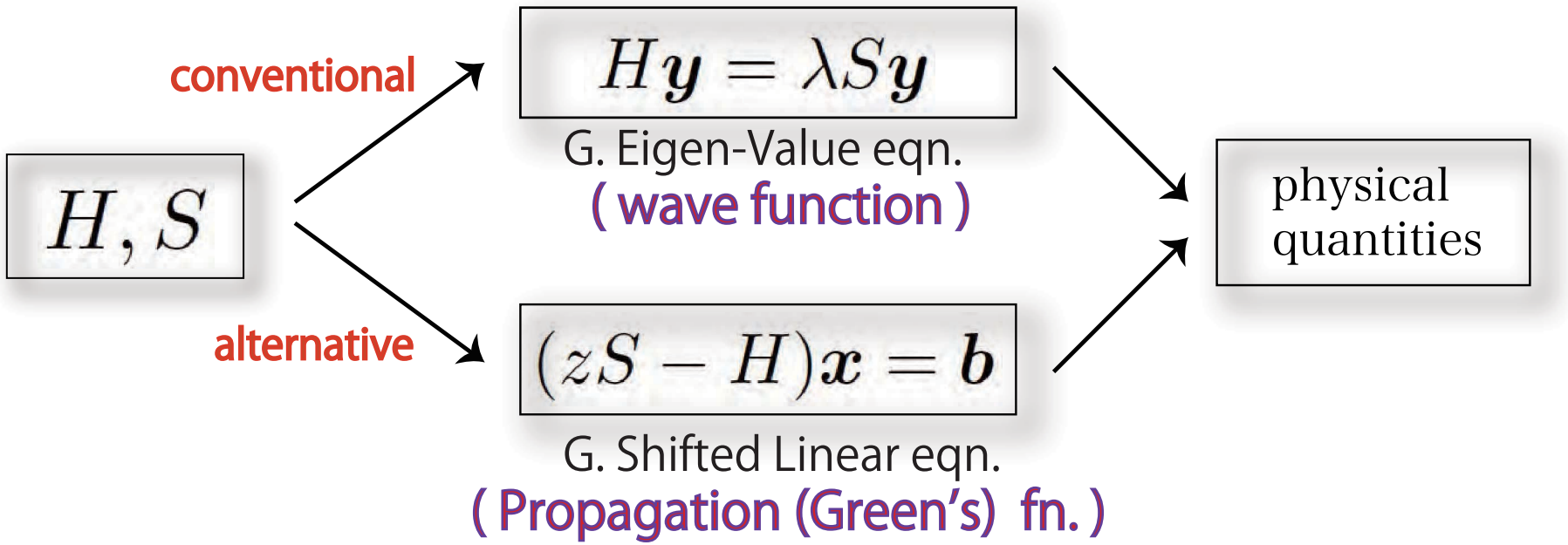
$$(zS - H)x = b$$

Architecture : The K computer (and beyond)

Ground algorithm design

Workflow of electronic structure calculations





Million dimensional generalized eigenvalue problem

- Real-symmatric generalized eigenvalue problem

$$A\boldsymbol{y} = \lambda B\boldsymbol{y}$$

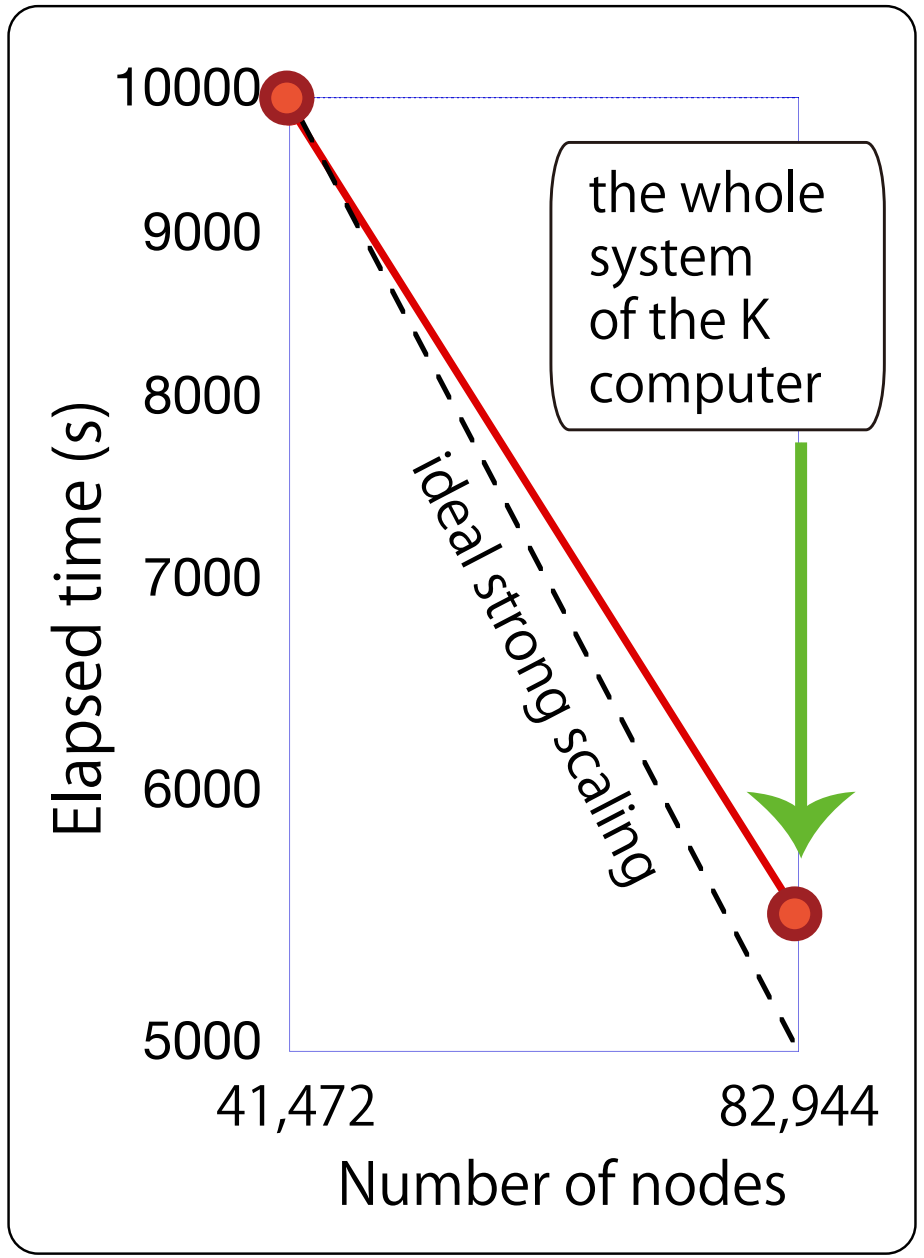
with the matrix dimension of $M=10^6$

→ • largest matrix size, strong scaling
  • 1.5 hour by the whole system of the K computer

- Solver library : 'EigenKernel'
  (https://github.com/eigenkernel/)

→A middleware-type dense-matrix solver

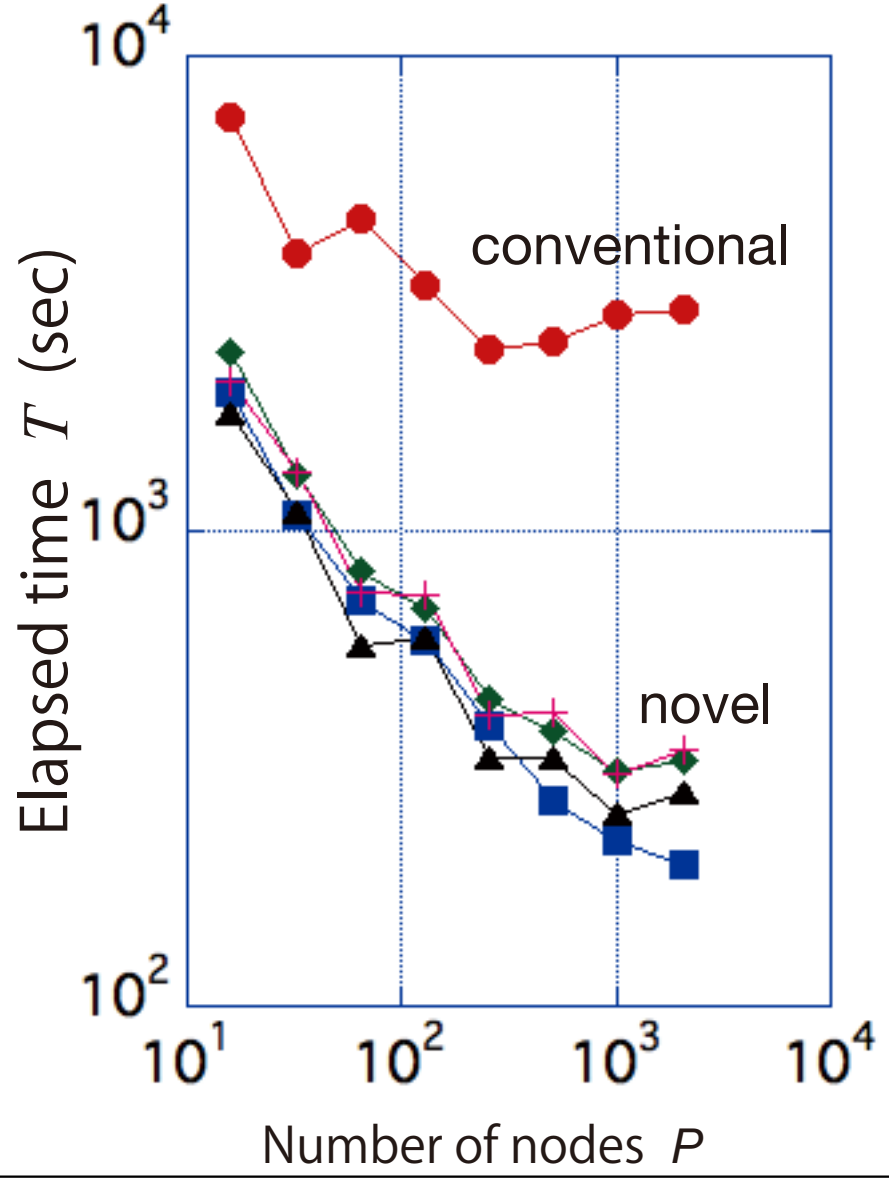
→ Hybrid workflow is possible among routines in scalable libraries of
  (i) ScaLAPACK
  (ii) ELPA (http://elpa.mpcdf.mpg.de/)
  (iii) EigenExa
  (http://www.aics.riken.jp/labs/lpnctrt/)



- H. Imachi and T. Hoshi, J. Inf. Proc. 24, 164 (2016)
- T. Hoshi, et al. Proc. ScalA16 in SC16, 33 (2016)
- K. Takana et al,
  preprint : http://arxiv.org/abs/1806.00741

Recent benchmark on Oakforest-PACS

→ strong scaling test with upto 2048 nodes,
a quater of the whole system



machine:
Xeon Phi, KNL

Conventional solver (ScaLAPACK)
→ severe problem in scalability

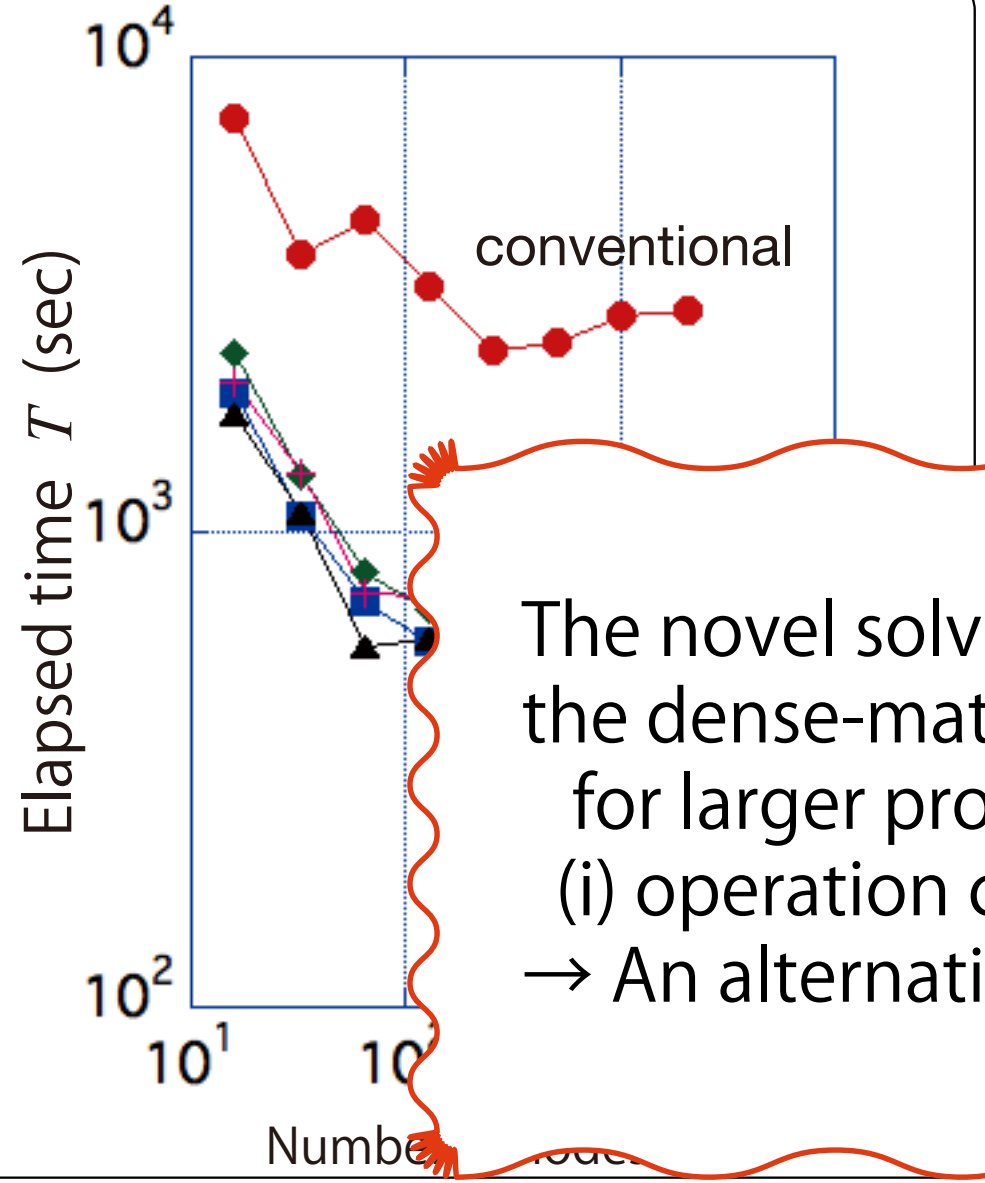
Novel solvers
(EPLA, EigenExa, and hybrids)
→ strong scaling property

Problem with the matrix size of M~90,000

K. Takana et al,
preprint : http://arxiv.org/abs/1806.00741

Recent benchmark on Oakforest-PACS

→ strong scaling test with upto 2048 nodes,
a quater of the whole system



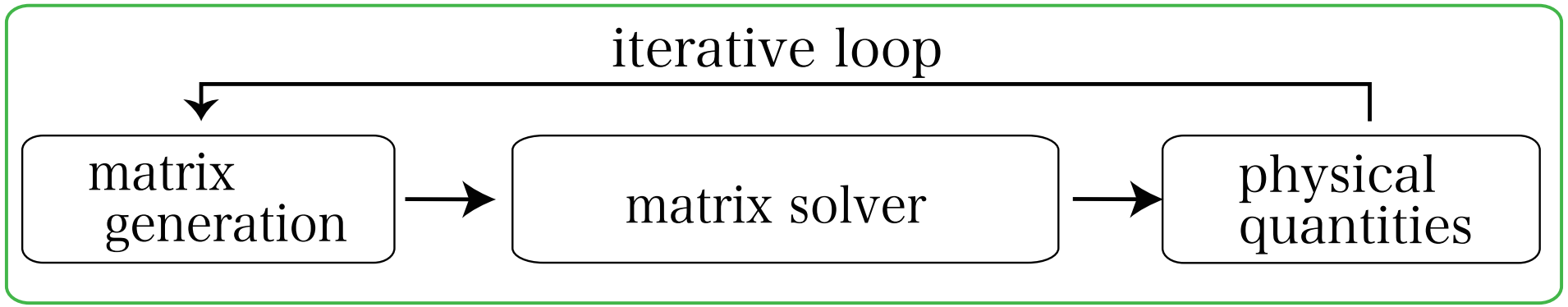
machine:
Xeon Phi, KNL

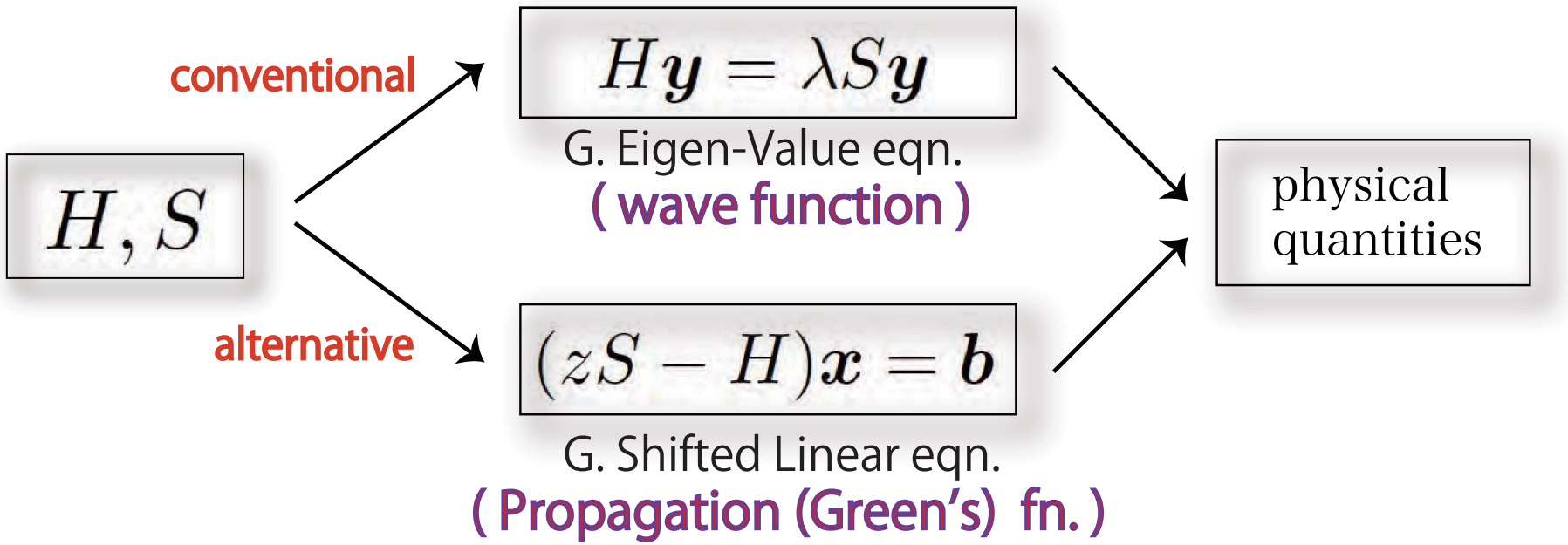
Conventional solver (ScaLAPACK)
→ severe problem in scalability

The novel solvers are fascinating but
the dense-matrix solvers have potential difficulty
for larger problems both in
(i) operation cost ($\propto M^3$) and (ii) scalability
→ An alternative approach is needed

Ground algorithm design

Workflow of electronic structure calculations





Basic equations

Generalized eigen-value (GEV) equation

wavefunction formulation

$$H\boldsymbol{y}_k = \varepsilon_k S\boldsymbol{y}_k$$

H, S : Hermitian, S: positive definite ($S \fallingdotseq I$)

$$G = \sum_k \frac{\boldsymbol{y}_k \boldsymbol{y}_k^{\mathrm{T}}}{z - \varepsilon_k}$$

Generalized shifted linear (GSL) equations

the propagation (Green's) function formulation

$$(zS - H)\boldsymbol{x} = \boldsymbol{b}$$ (z : complex energy)

non-Hermitian

$$\rightarrow \quad \boldsymbol{x} = G\boldsymbol{b}$$

with $G \equiv (zS - H)^{-1}$: the Green's function

Highly parallelizable mathematical structure

A pioneering work : W. Kohn, Phys. Rev. Lett. (1996) (W. Kohn won the Nobel Prize at 1998.)

Generalized eigen-value problem

$$H\boldsymbol{y}_k = \lambda_k S\boldsymbol{y}_k \quad (1)$$

Physical quantity with a given matrix X

(Ex. the case of $X = H$
--> Electronic structure energy)

$$\langle X \rangle \equiv \sum_k f(\lambda_k)\, \boldsymbol{y}_k^{\mathrm{t}} X \boldsymbol{y}_k \quad (2)$$

with a given weight funciton
('Fermi distribution function')

$$f(\lambda) \equiv \frac{1}{\exp(\beta(\lambda-\mu))+1} \quad (3)$$

(β, μ : given parameters)

Physical quantity in trace form

$$\langle X \rangle = \mathrm{Tr}[X\rho] \quad (4)$$

with the density matrix

$$\rho \equiv \sum_k f(\lambda_k)\boldsymbol{y}_k\boldsymbol{y}_k^{\mathrm{t}} \quad (5)$$

Decomposition of the trace form

$$\mathrm{Tr}[\rho X] = \sum_j \boldsymbol{e}_j^{\mathrm{t}}\rho X \boldsymbol{e}_j \quad (6)$$

'projected physical quantity' calculated in parallelism

with $\boldsymbol{e}_j$ (j-th unit vector)

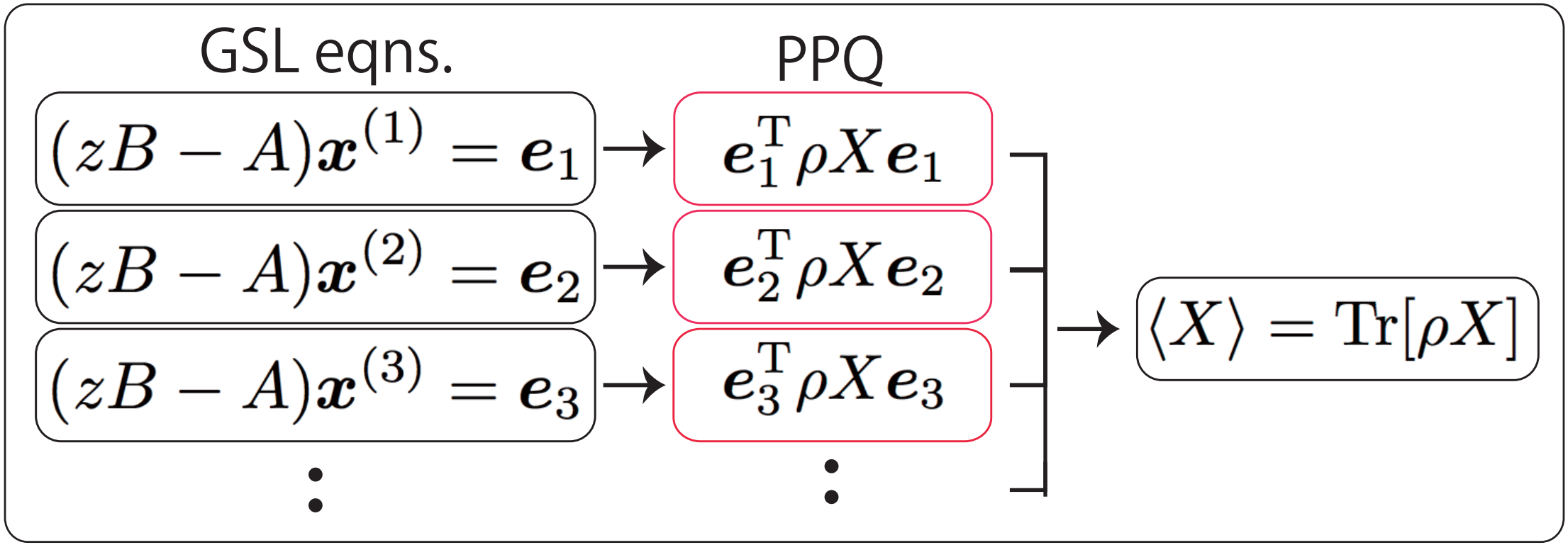
$$\boldsymbol{e}_j \equiv (0, \cdots 0, 1_j, 0, \cdots, 0)^{\mathrm{t}} \quad (7)$$

Highly parallelizable mathematical structure

The trace decomposition

$$\mathrm{Tr}[\rho X] = \sum_j \boldsymbol{e}_j^{\mathrm{t}} \rho X \boldsymbol{e}_j \qquad (6)$$

gives the generalized shifted linear (GSL) equations
for calcualtion of projected physical quantities (PPQ).



Highly parallelizable mathematical structure

The trace decomposition

$$\mathrm{Tr}[\rho X] = \sum_j \boldsymbol{e}_j^{\mathrm{t}} \rho X \boldsymbol{e}_j \qquad (6)$$

gives the generalized shifted lin
for calcualtion of projected p

solved by Krylov subspace
→ called shifted
Krylov subspace method

GSL eqns.

$$(zB - A)\boldsymbol{x}^{(1)} = \boldsymbol{e}_1 \rightarrow$$
$$(zB - A)\boldsymbol{x}^{(2)} = \boldsymbol{e}_2 \rightarrow \boldsymbol{e}_2^{\mathrm{T}} \rho X \boldsymbol{e}_2$$
$$(zB - A)\boldsymbol{x}^{(3)} = \boldsymbol{e}_3 \rightarrow \boldsymbol{e}_3^{\mathrm{T}} \rho X \boldsymbol{e}_3$$
$$\vdots \qquad \vdots$$

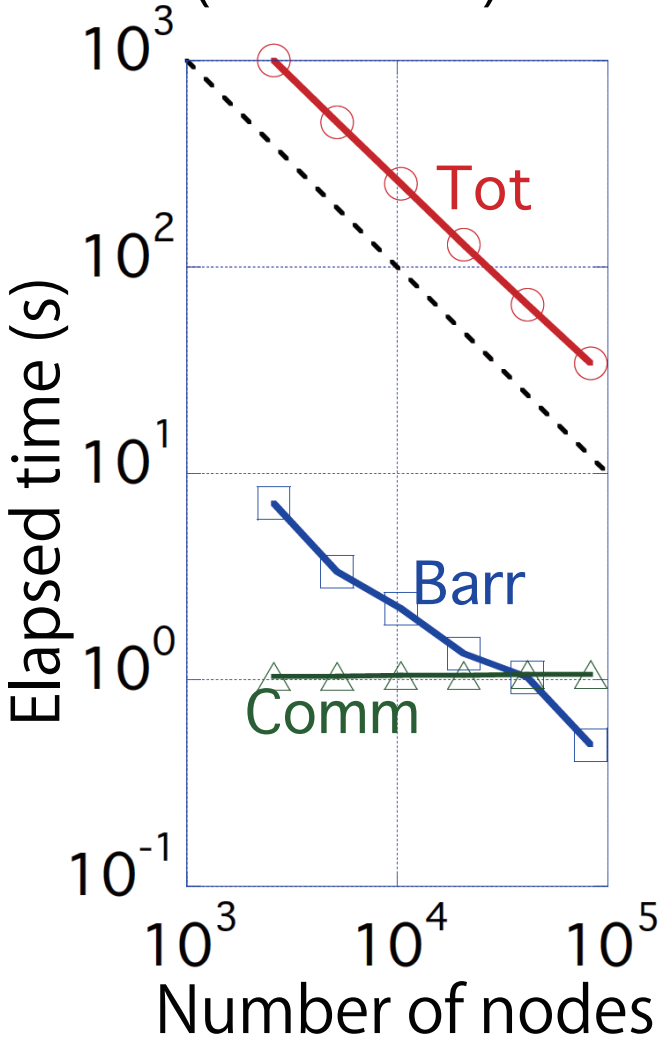
$$\rightarrow \langle X \rangle = \mathrm{Tr}[\rho X]$$

Parallel efficiency (strong scaling) on the full system of the K computer

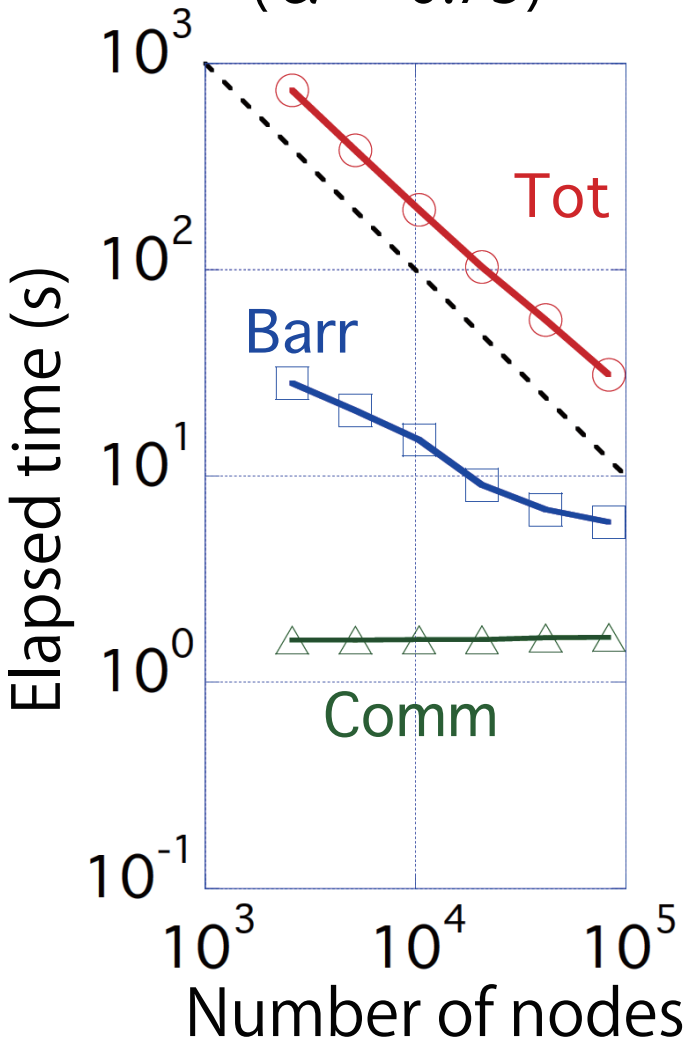
100-nm-scale or 10^8 atoms calculations

α : parallel efficiency ratio (α =1 for the ideal strong scaling) .

(a) ideal diamond crystal ($\alpha = 0.92$)



(b) condensed polymer ($\alpha = 0.75$)



Tot : total elapsed time

Barr: barrier time(*)

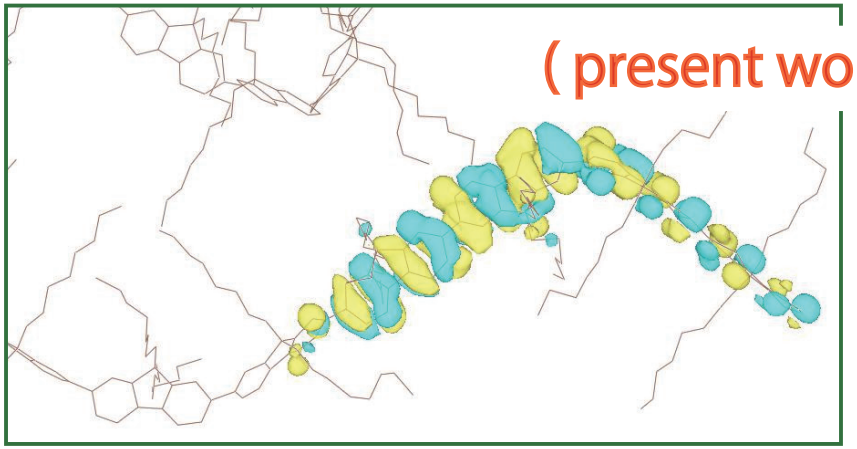
Comm: MPI communication time

(*)The barrier time includes the time to wait for other processors.

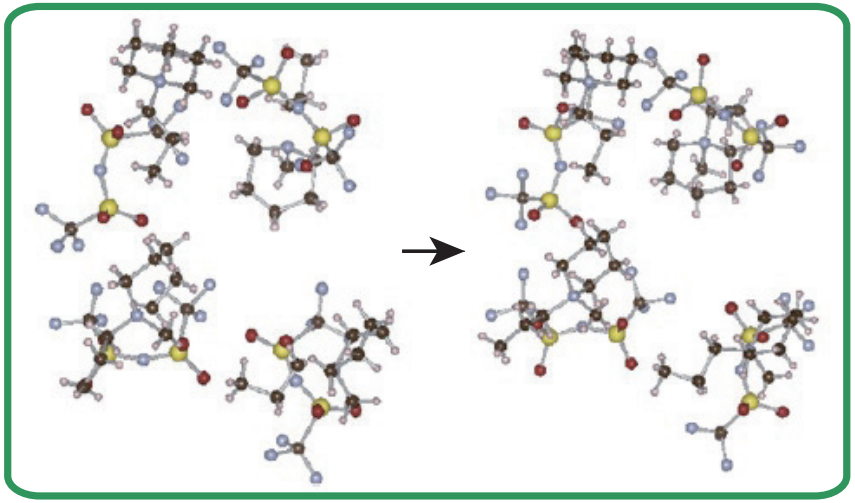
T. Hoshi, et al.
Proc. ScalA16
in SC16, 33 (2016)

Applications with ELSES (http://www.elses.jp), our software

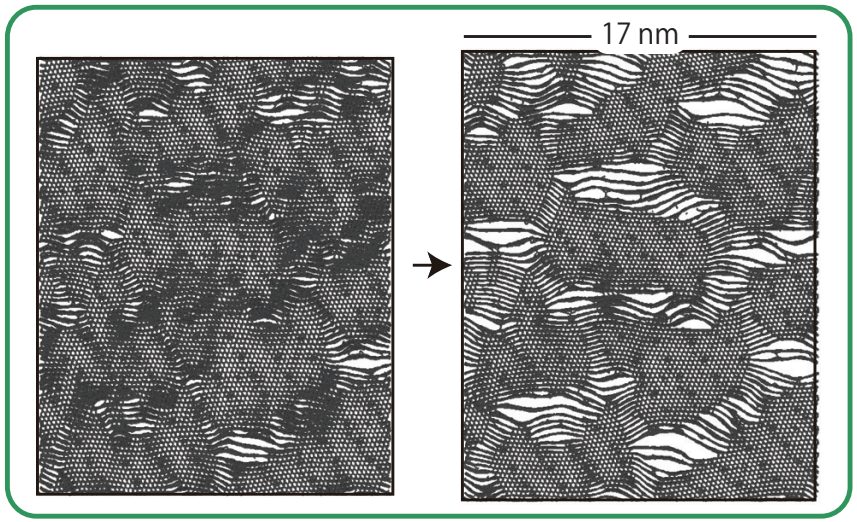
Organic device materials
(with Sumitomo Chemical Co.)



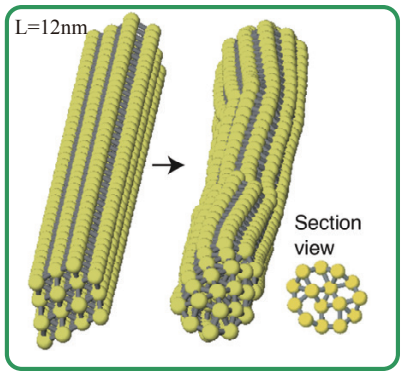
Battery material (with Toyota)



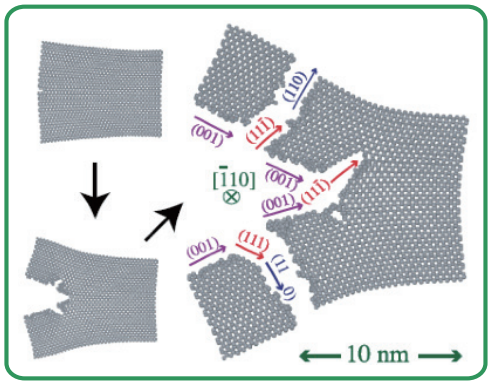
Ultra-hard diamond



helical metal
nanowire



silicon fracture



Detailed method: ref: Hoshi et al, J. Phys.: Cond. Matt. 24, 165502 (2012)

- Modelled (transferable tight-binding) theory based on first-principles calculations
- Local-area approximation ('real-space projection') on the matrices in (zS-H)x=b

The algorithmic strategy of the use of the shifted linear eqns is general and was applied to many scientific areas with large computation, for examale ..

$$(zI - A)\boldsymbol{x} = \boldsymbol{b}$$

[1] (**QCD**) A. Frommer, Computing 70, 87 (2003)

[2] (**large scale electronic structurre calc.**)

R. Takayama, T. Hoshi, T. Sogabe, S.-L. Zhang, and T. Fujiwara, PRB 73, 165108 (2006)

[3] (**many-body wavefunction theory**)

S. Yamamoto, T. Sogabe, T. Hoshi, S.-L. Zhang and T. Fujiwara, JPSJ 77,114713 (2008).

→ spectrum for $La_{3/2}Sr_{1/2}NiO$, matrix size：M=64,000,000
(Multi-orbital extended Hubbard model)

$$A(\omega) = -\frac{1}{\pi}\mathrm{Im}[\langle\psi_0|\hat{O}^\dagger \frac{1}{\omega + E_0 + i\epsilon - \hat{H}}\hat{O}|\psi_0\rangle] \quad = x$$

→ 'Kω' https://github.com/issp-center-dev/Komega/ (2017); paper in preparation

[4] (**GW**) F. Giustino, M. L. Cohen, S. G. Louie, Phys. Rev. B. 81, 115105 (2010)

[5] (**shell model calculation**)T. Mizusaki, et al., Phys. Rev.C 82, 024310 (2010)

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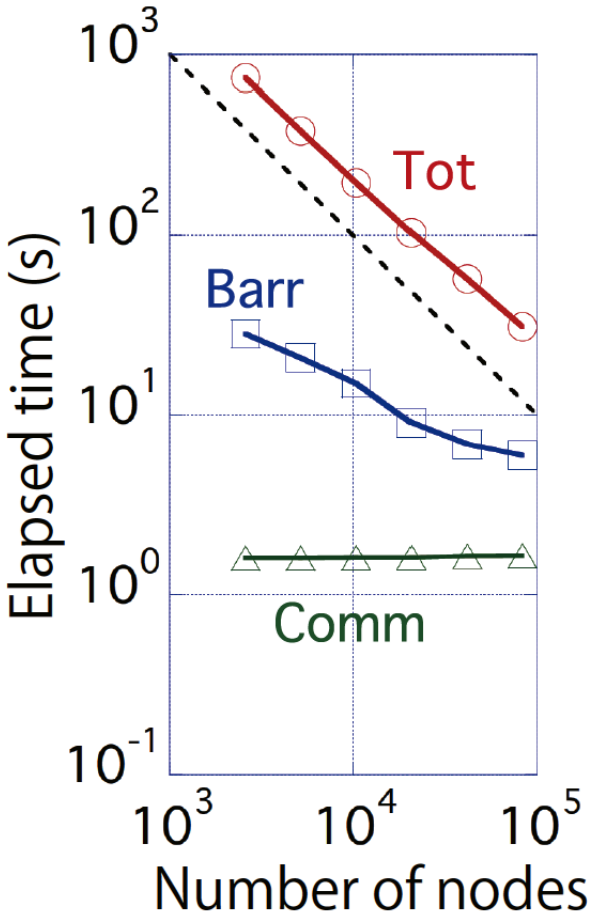
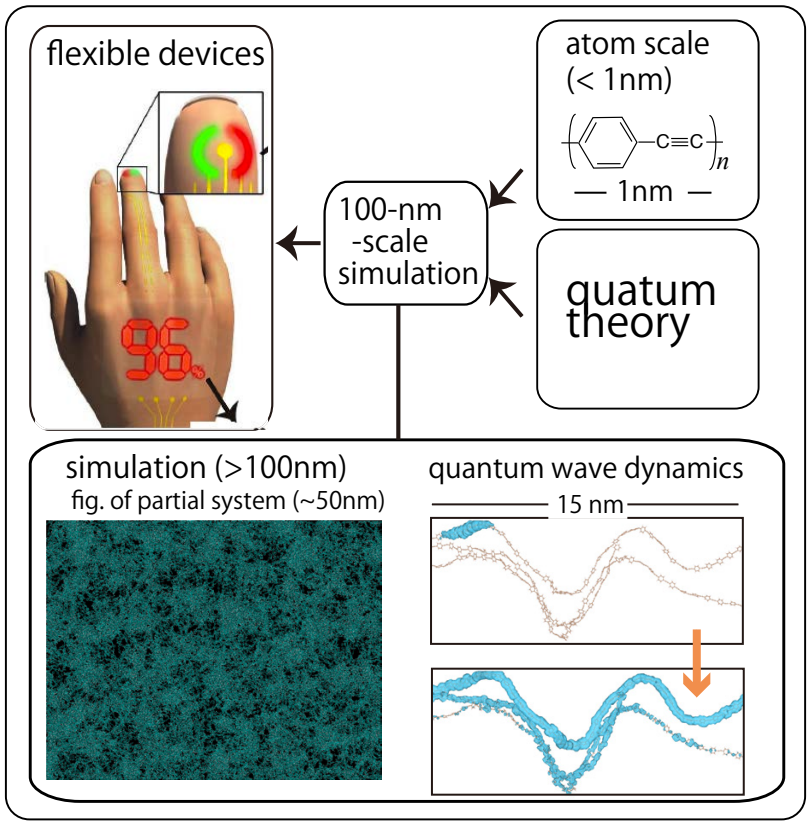


2. Application



Organic material for ultra-flexible devices

- next-generation Internet-of-Things (IoT) producs
- ultra-thin (≦ 1,000 nm) •low fabrication cost by printing technique

Current device
ex. iPhone X

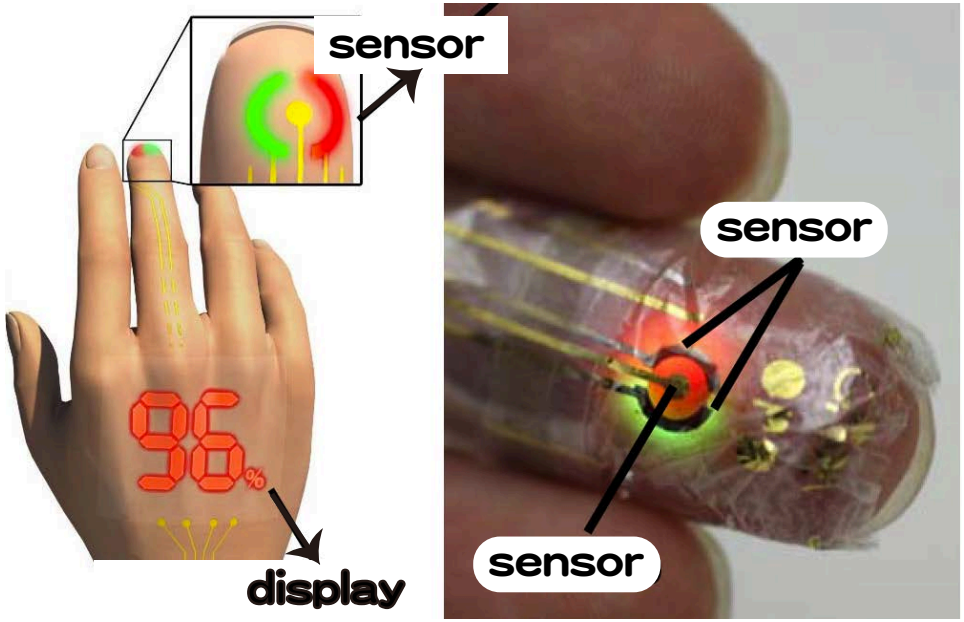
http://www.apple.com/

Ex. future ultra-flexibile opt-electronic devices

(a) display (NHK RD)

J. Soc. Inf. Display, 24, 3 (2016)

(b) 'e-skin' health monitor* (U Tokyo)



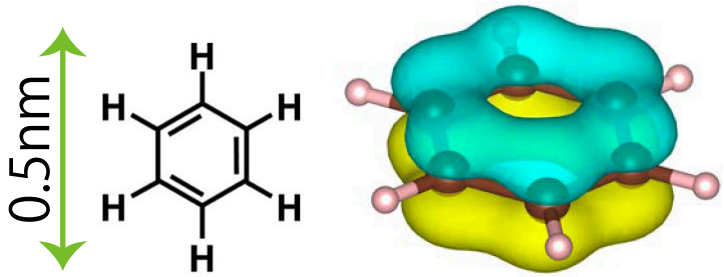
Sci. Adv. 2, e1501856 (2016)

*pulse oximeter

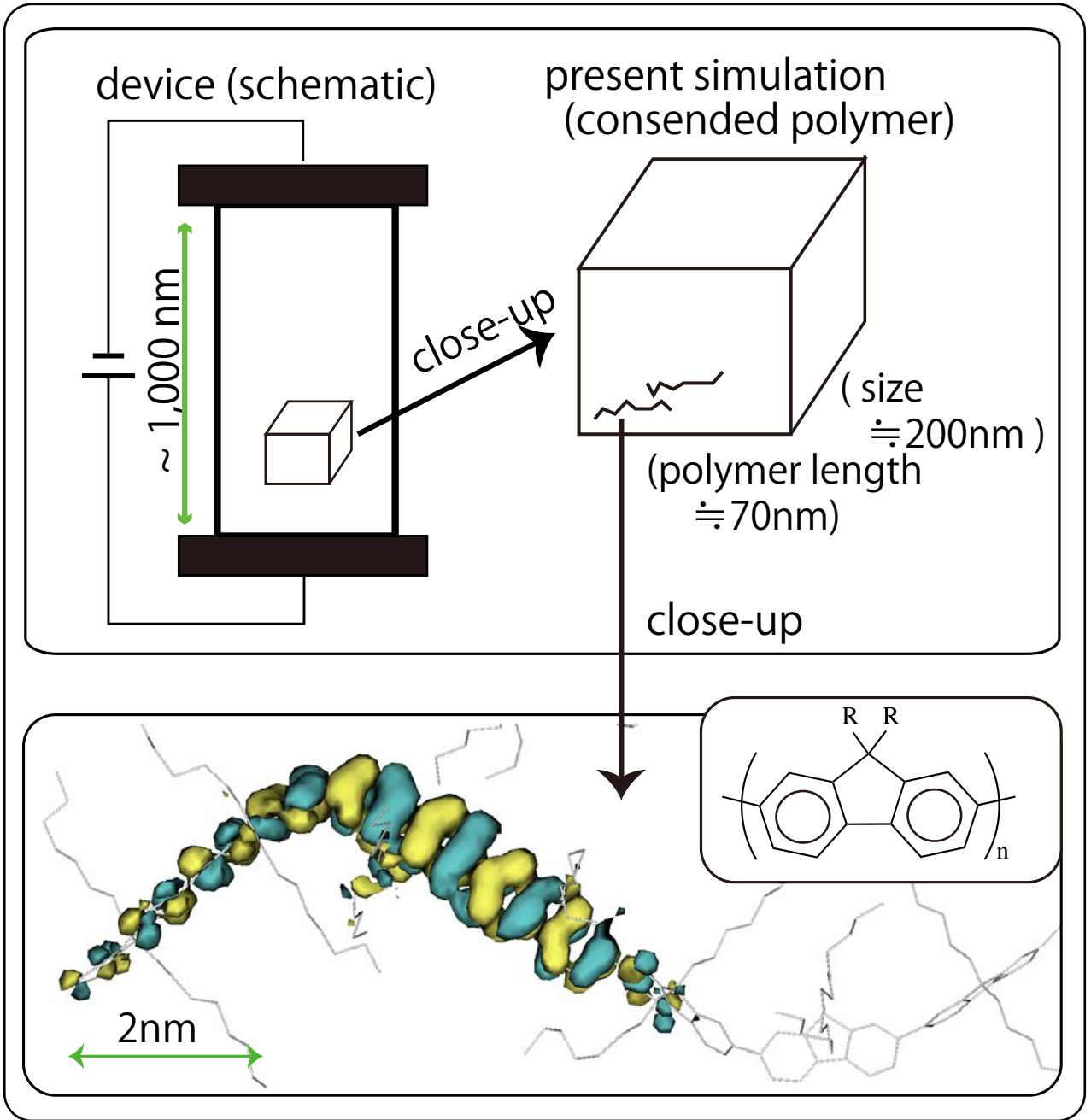
Organic device material and 100-nm-scale quantum simulation

Organic device material → Scales between device and 100-nm-scale simulation

- semiconductor with 'π-type' electronic wave that lines in benzene rings and so on
  → strong anisotropy



Pionnering experiments at '70 by H. Shirakawa et al.
→ The Nobel Prize in Chemistry 2000
'Discovery and development of conductive polymers'



General issue of organic opt-electronic device material

- complicated disordered structure
  → large-scale electronic state calculations is crucial
  (cf. silicon material : ideal crystal with perfect periodicity)
- (Eigenvector) ⇆ (spatial extent of wavefunction) ⇆ (device property)
- Fundamental conflicting aspects in material design

electrical property (electrical current)	→ Extend wavefunction is preferable. (→ easy to propagate)
optical property (display, sensor, soler cell battery)	→ Localized wavefunction is preferable. (→ easy to interact with light)

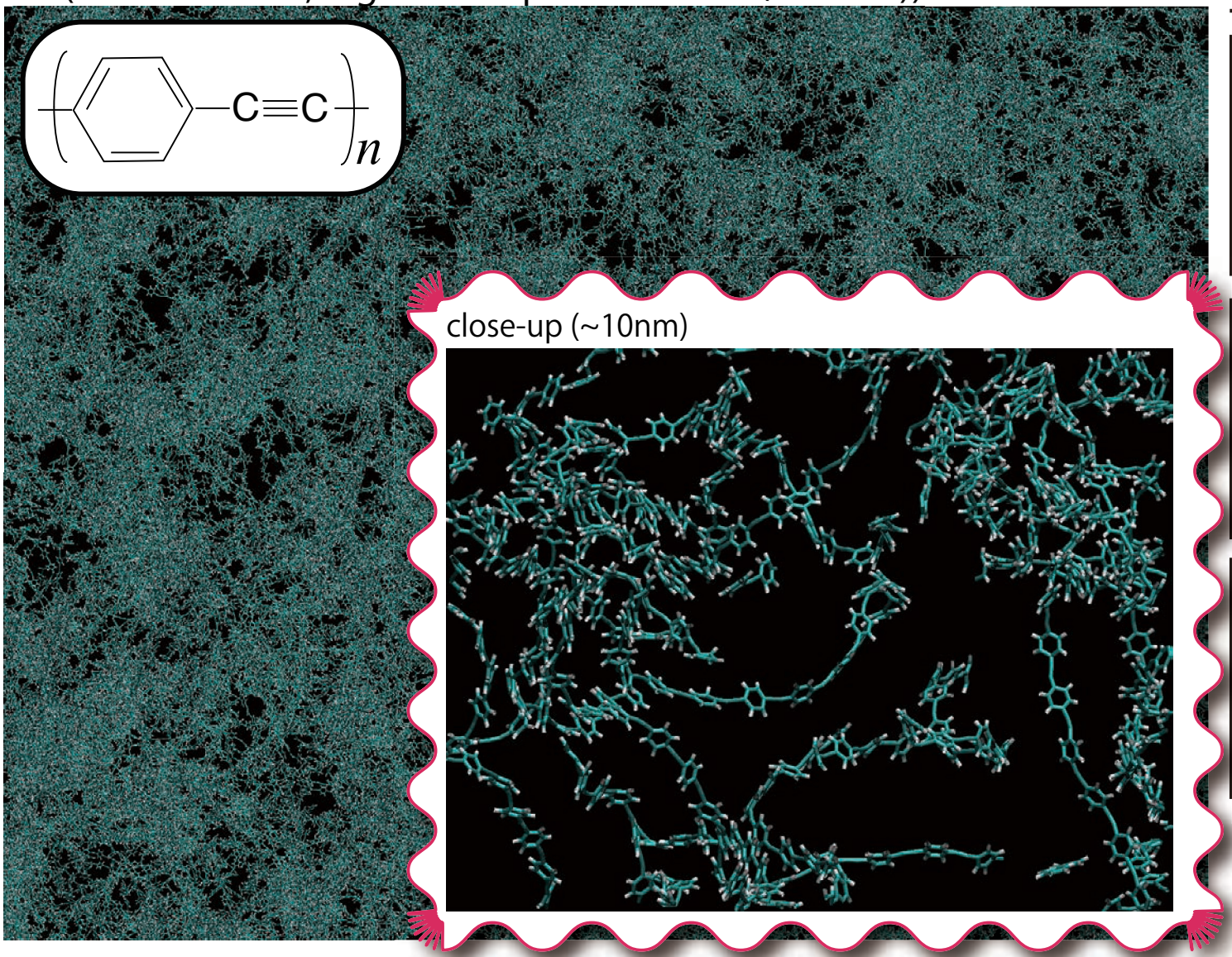
Note: other issues: device lifetime, fabrication costs

Hoshi et al., Proc. ScalA16 in SC16, 33 (2016).

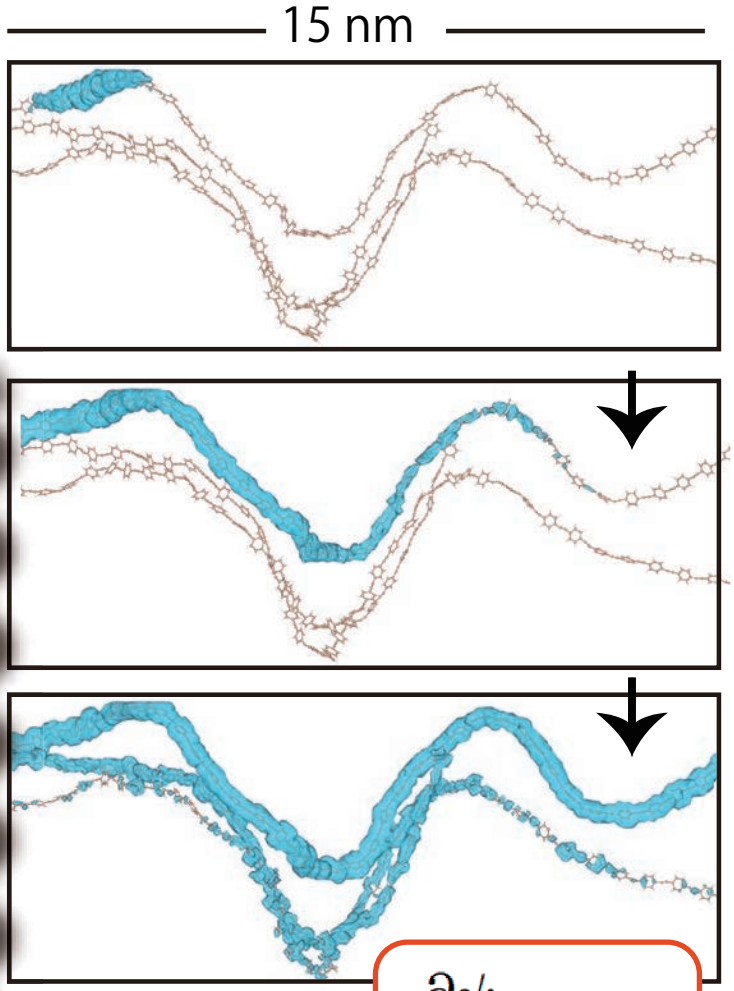
100-million-atom simulation of condensed organic polymers

→ suggests a quantum trasnport mechanism through dynamical polymer networks

(a) poly-(phenylene-ethynylene) (PPE)
(size≒200nm; figure of a partial retion (~50nm))



(b) quantum dynamics with 1 ps

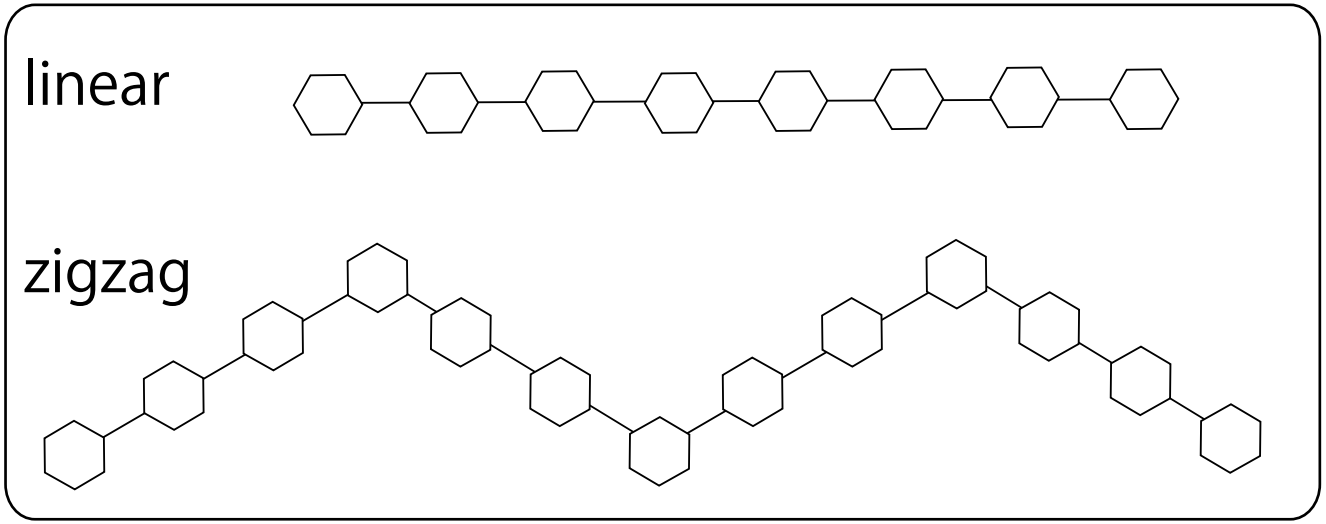


$$i\frac{\partial \psi}{\partial t} = H\psi$$

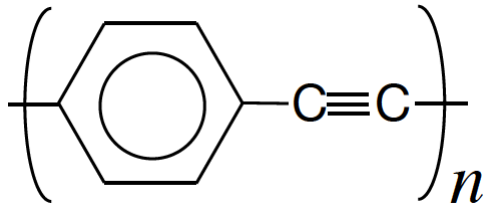
Research on realistic (complicated disordered) polymer

poly(phenylene–ethynylene) ([1] J. Terao, et al, Nature Comm. 4, 1691 (2013))

→ The static and dynamical disorders
are essential for device performance (mobility value)



example (linear chain)



Result

mobility in ideal structure
(linear) > (zigzag)
mobility in realistic (disordered) structure
(linear) < (zigzag)

mobility (cm^2/Vs)

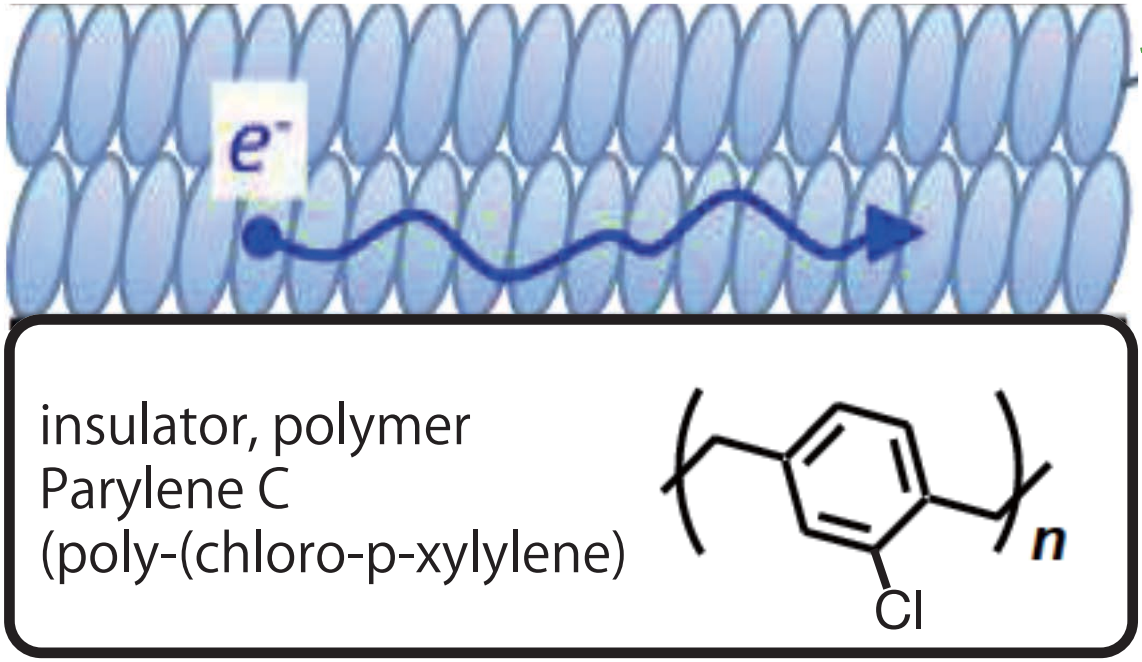
	linear	zigzag
present calc. [2]	0.98	1.4
exp. [1]	0.75	2.1
moldel only w/ π orbitals [1]	5.4	7.2

[2] Imachi et al., AIP CP 1790, 020010 (2016)
(with T. Tada (Tokyo Tech.))

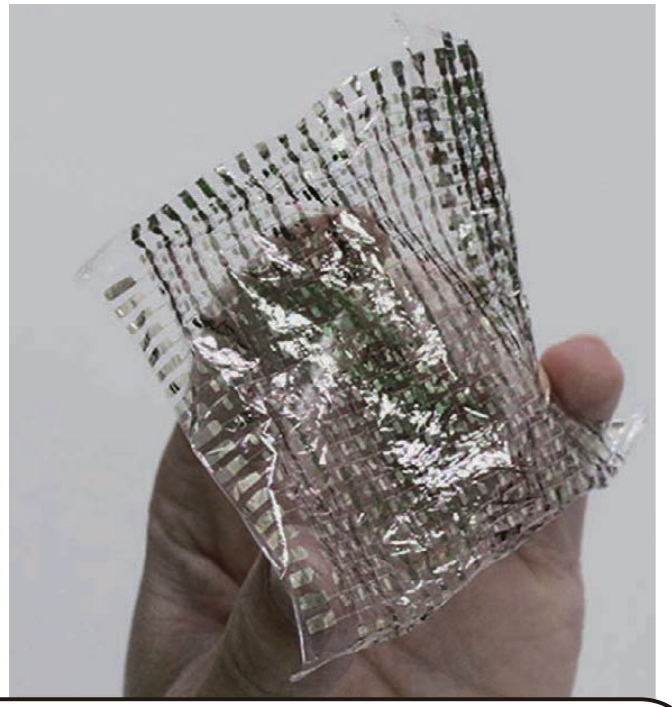
Preliminary result of 2D organic material (1)

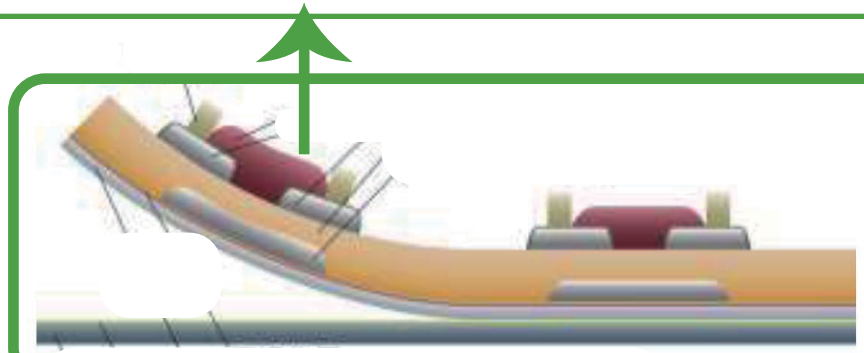
- Target device: ultra-flexible organic transistor
- Collaboration with H. Matsui (Yamagata U).

Schematic figure (side view)



Pentacene





close up

Flexible, ultra-thin device
by Yamagata U.;
Nature Comm. 5. 4147 (2014)

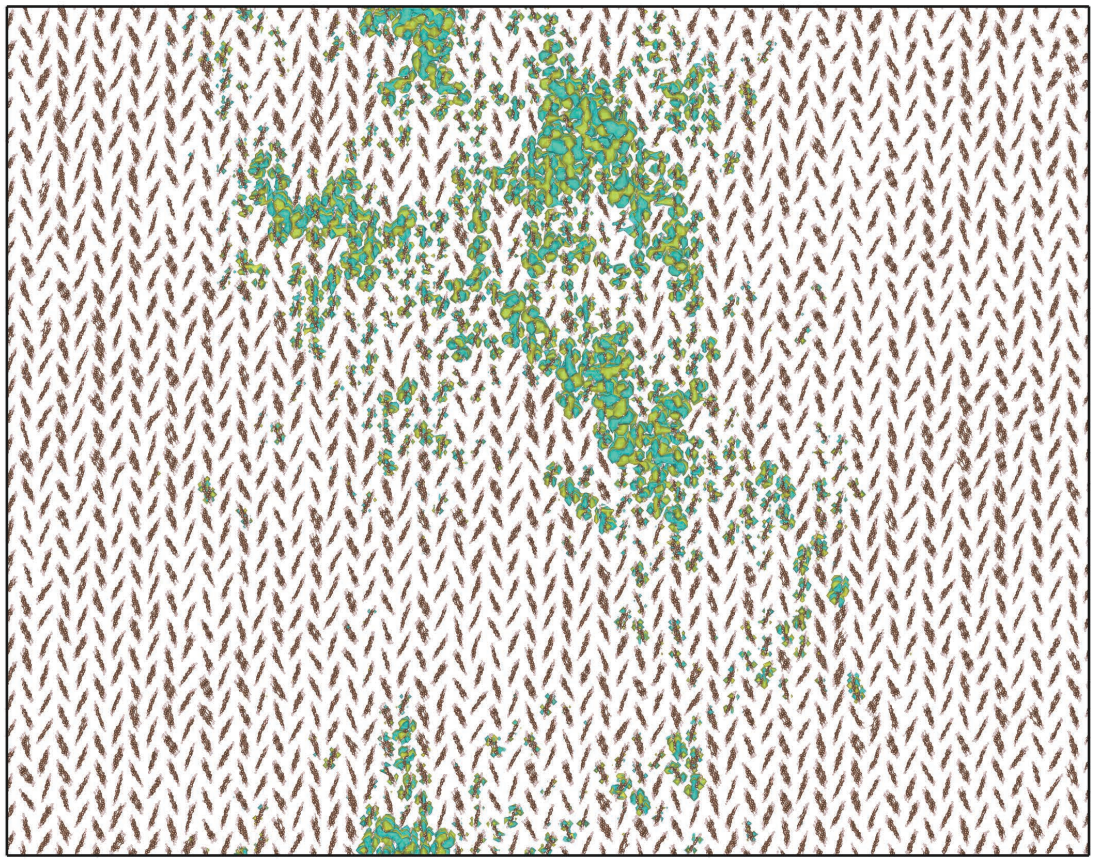
Preliminary result of 2D organic material (2)

Experimental observation of 'semi-localized' states over $\leqq 10^2$ molecules

→Electron Spin Resonance experiment

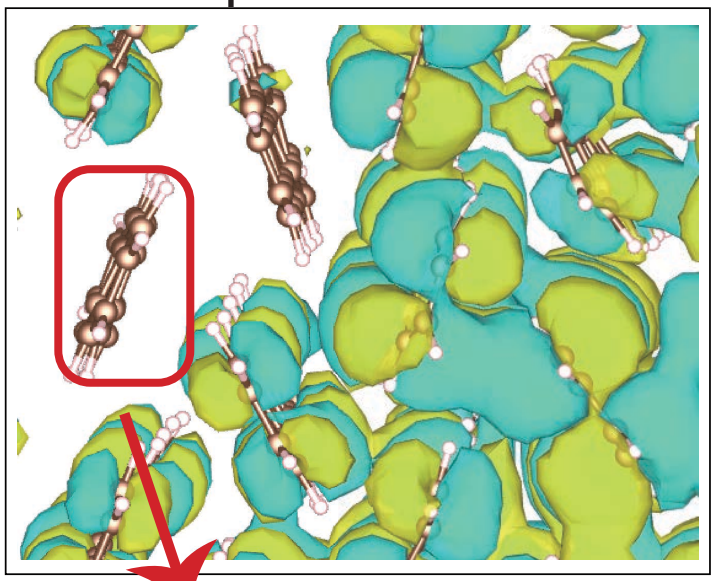
by H. Matsui (Yamagata U.), Phys. Rev. Lett. 104, 056602 (2010)

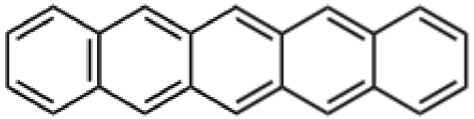
Calculated 'semi-localized' wavefunctions in disordered pentacene thin film (top view)



23 nm

close-up





~ 1 nm

calculated sample:
1,800 molecules (64,800 atoms)

Remarks for exascale computation

1. application-algorithm-architecture co-design
→ parallel algorithm
for specific problem and/or architecture
→ codes may be shared among applications
as numerical library or mini-application

2. possible target
: organic materials for flexible device
→ condensed organic molecules or polyemers
in 100-nm-scale complicated disordered structure

Other (skipped) recent topics

1. Performance prediction of parallel eigenvalue solver
   by Bayesian inference
   K. Tanaka et al., http://arxiv.org/abs/1806.00741

2. Novel numerial algoritm for calculating partial eigenpairs
   with Sylvester's theorem of inertia and sparse-direct solver
   D. Lee et al., J. Comp. Phys. 371, 618 (2018)
   https://github.com/lee-djl/k-ep

3. Data-scientific analysis (principal component analysis)
   of disordered organic polmers
   with electronic state calculation
   T. Hoshi, et al., in SIAM-PP18, Mar. 2018;
   Paper in preparation.

Summary

- 3-page tutorial on quantum material simulations
  → fundamental theory, numerical aspects
- A novel linear algebraic algorithm for massively parallel computaion
  with generalized eigenvalue equation
  or generalized shifted linear equations
  → an extreme scalabilityon on the whole system of the K computer
- Application to flexible device materials,
  with organic molecule or polymers,
  for next generation
  Internet-of-Things (IoT) products,
  such as flexible display, sensor
  and battery.