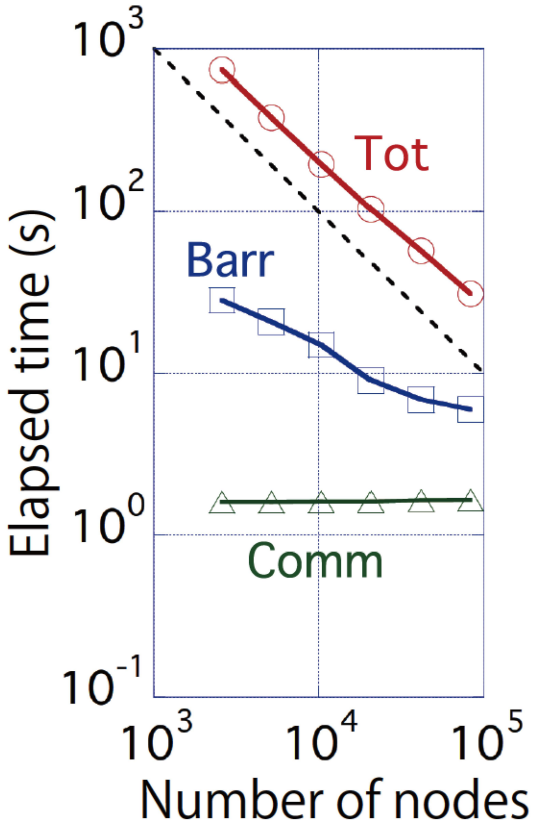
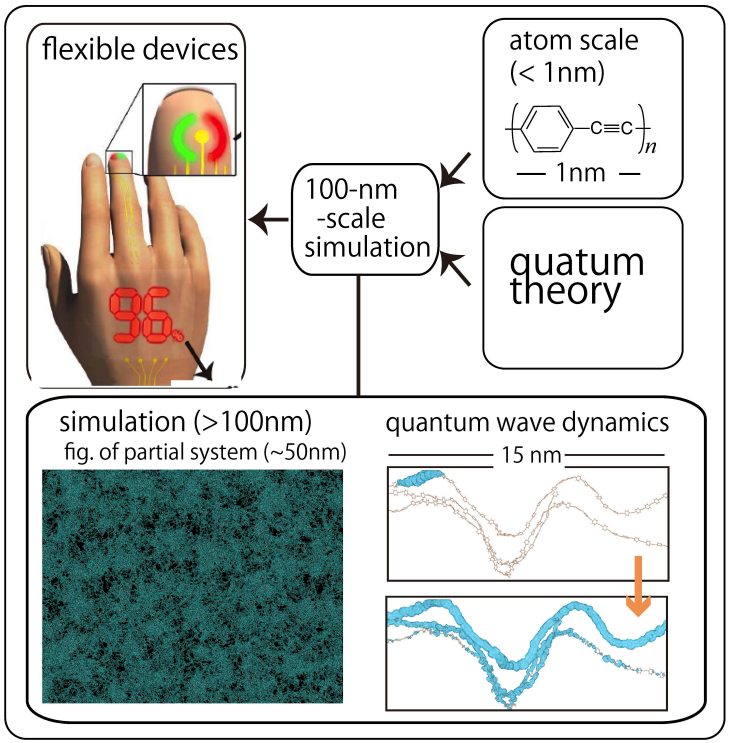
100nmスケール有機デバイス材料研究と数理

星健夫, 井町宏人, 梶貴美, 安部友樹也, 大平健太郎, 福本智哉(鳥取大)

1. アルゴリズム



2. アプリケーション



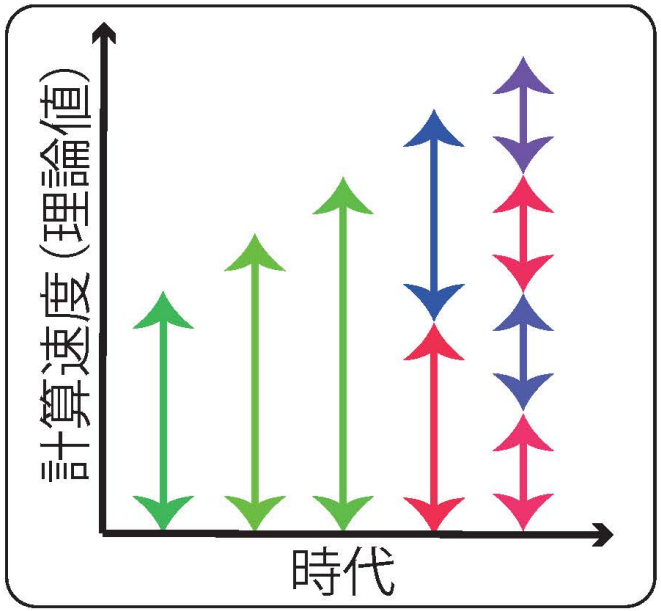
3. 現在進行形の話

(a) 光電子デバイス
(b) 機械学習との融合

1+2: T. Hoshi et al., in press;
http://arxiv.org/abs/1609.08377

共同研究(有機材料) : 多田朋史(東工大)・石田雅也(住友化学)
共同研究(コードチューニング) : 南一生など(理研計算科学研究機構)

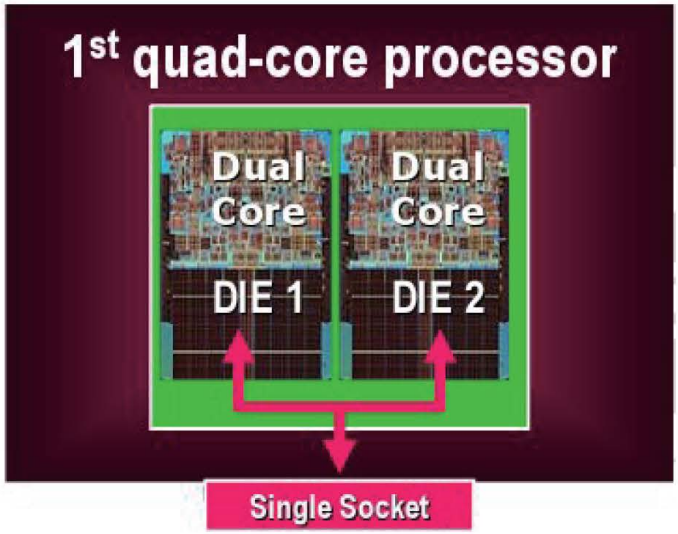
背景：並列計算へのパラダイムチェンジ(2000年代後半~)



→「手分け」して計算する設計
→計算手法にパラダイムチェンジを迫ることに

Herb Sutter
"The Free Lunch Is Over",
Dr. Dobb's Journal, 30(3), March (2005)

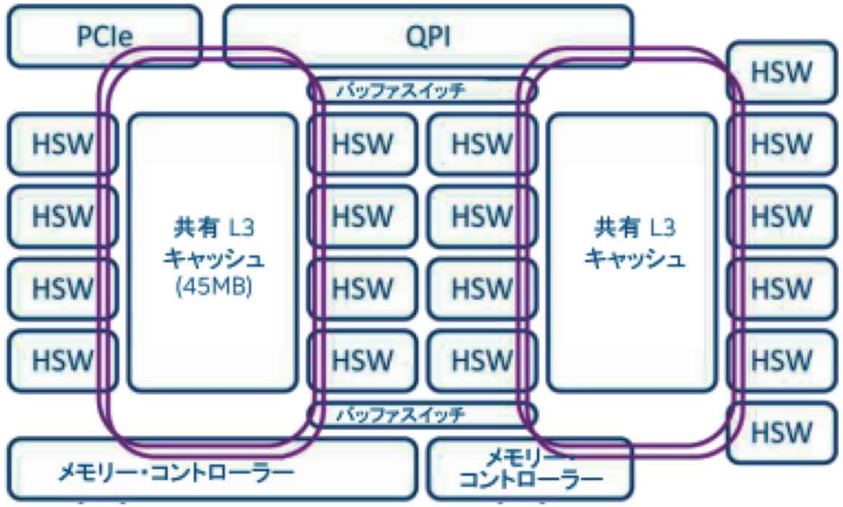
→物質科学と数理・情報科学の共同研究



インテル社のCPU

←Core 2 Quad(2006)

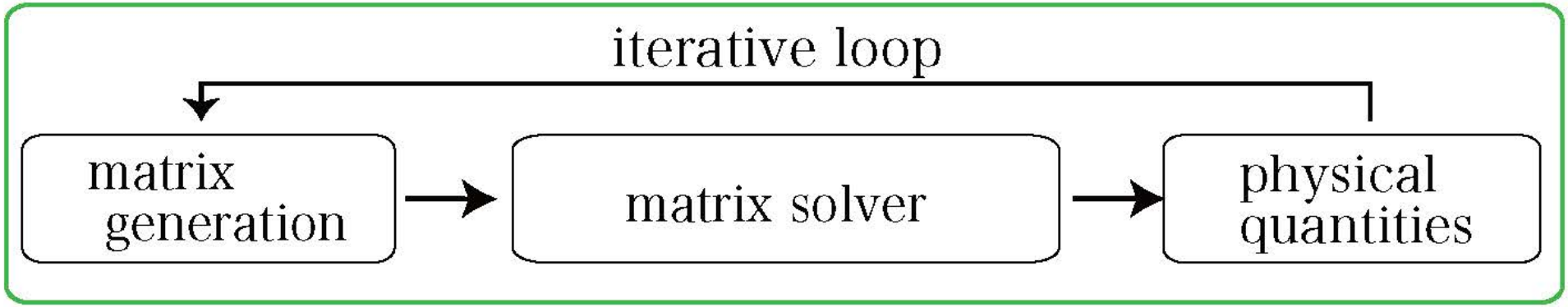
18-core Xeon (*)
(E5-2600-v3) (2014)→

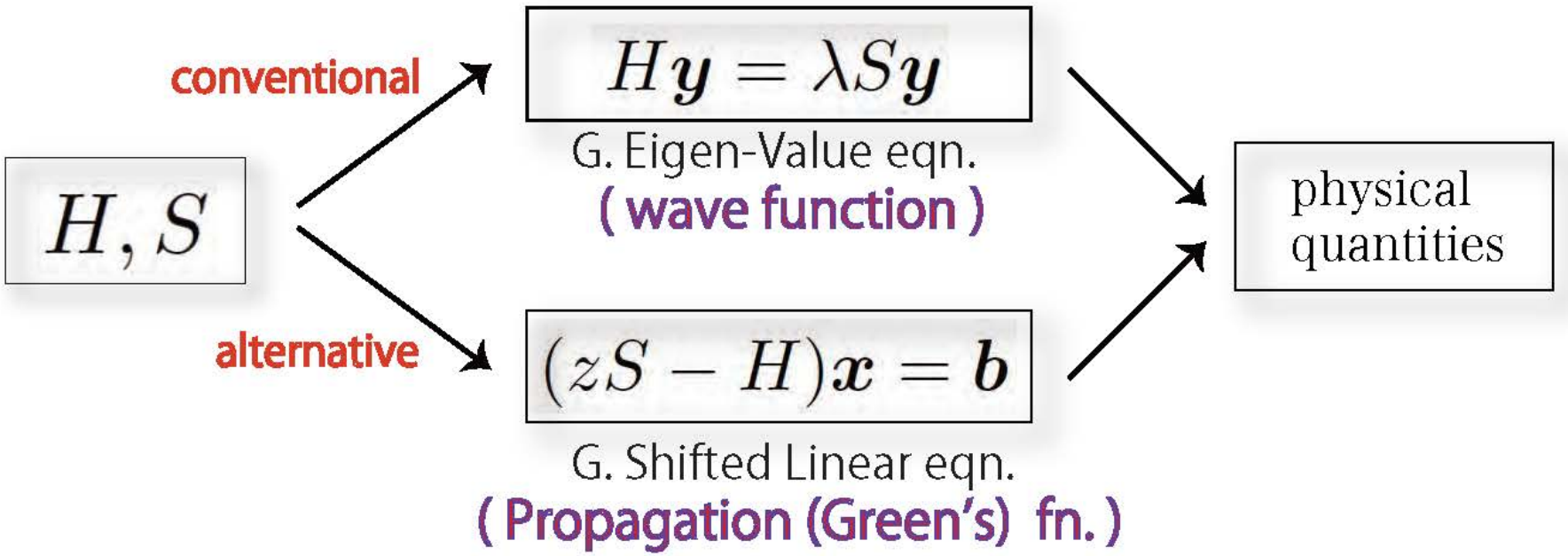


(*) http://pc.watch.impress.co.jp/docs/column/kaigai/20140909_665735.html

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

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

Physical quantity with a given matrix X

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

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

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

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

(β , μ : given parameters)

Physical quantity in trace form

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

with the density matrix

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

Decomposition of the trace form

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

'projected physical quantity' calculated in parallelism

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

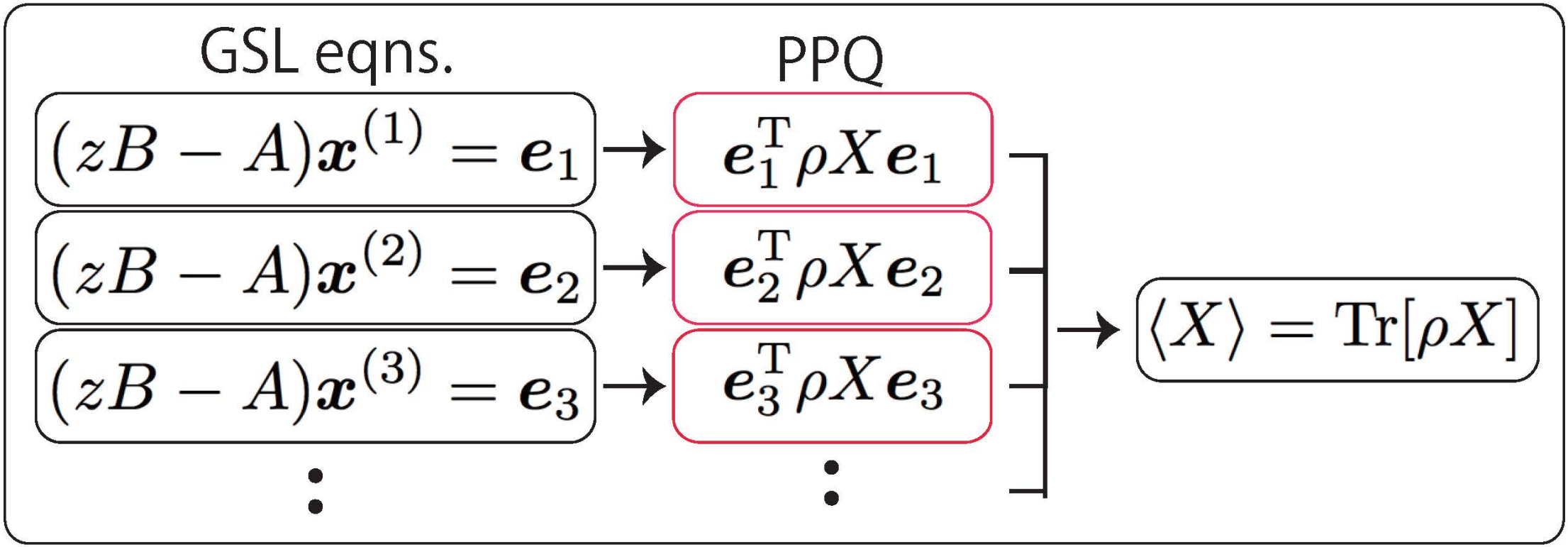
$$\boldsymbol{e}_j \equiv (0, \cdots 0, 1_j, 0, \cdots, 0)^{\mathrm{t}} \qquad (7)$$

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

Simplified explanation

- decomposition of the trace form
- calculation of trace elements
  as parallel computation

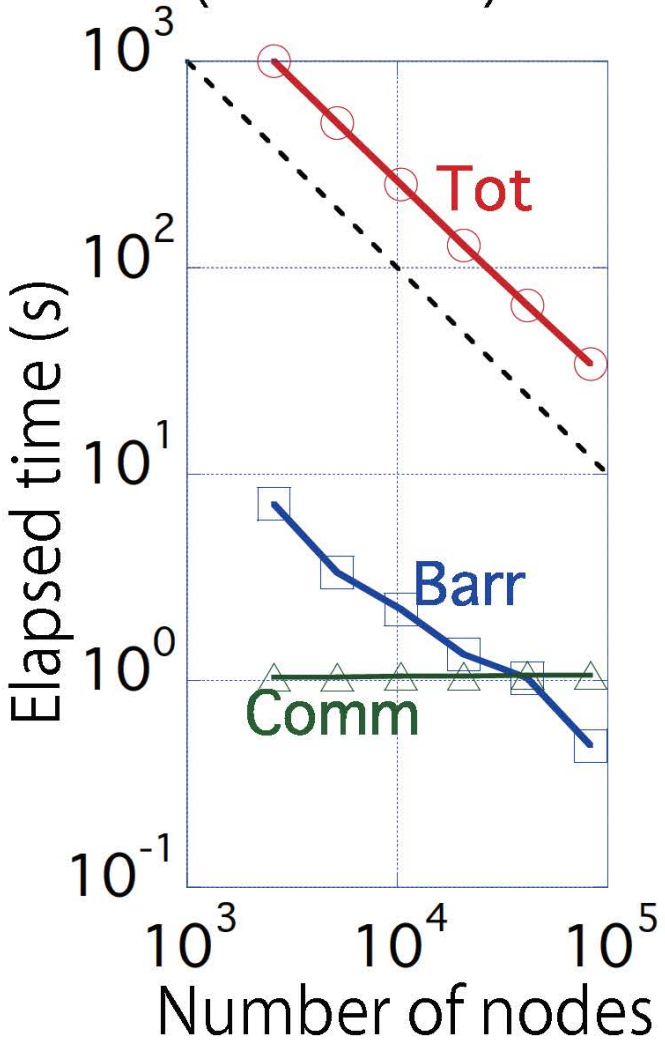
$$\mathrm{Tr}[A] = A_{11} + A_{22} + \ldots + A_{MM}$$

parallel computation

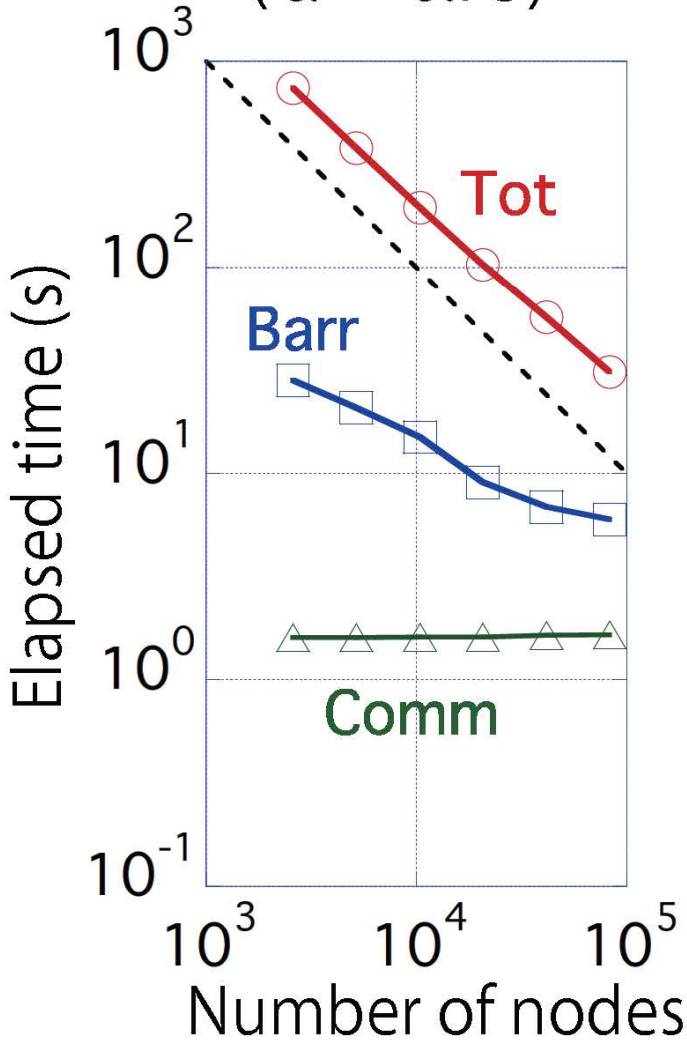
Parallel efficiency (strong scaling) on the full system of the K computer

100-nm-scale or 10^8 atoms calculations
parallel efficiency ratio α is determined with the reference data with 2,592 nodes.

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



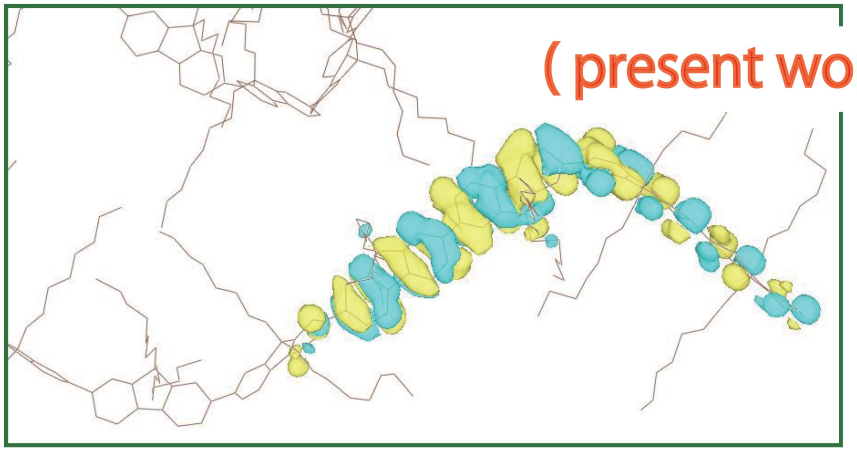
(b) condensed polymer ($\alpha = 0.75$)



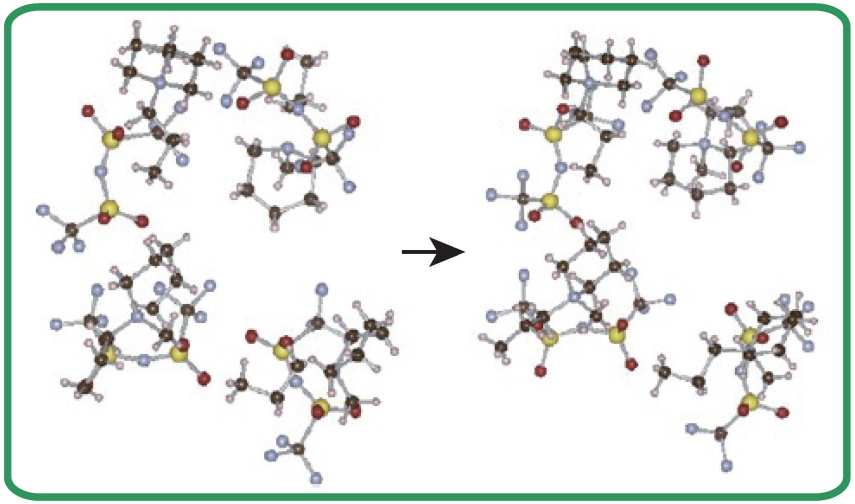
Tot : total elapsed time
Barr: barrier time
Comm: MPI communication time

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

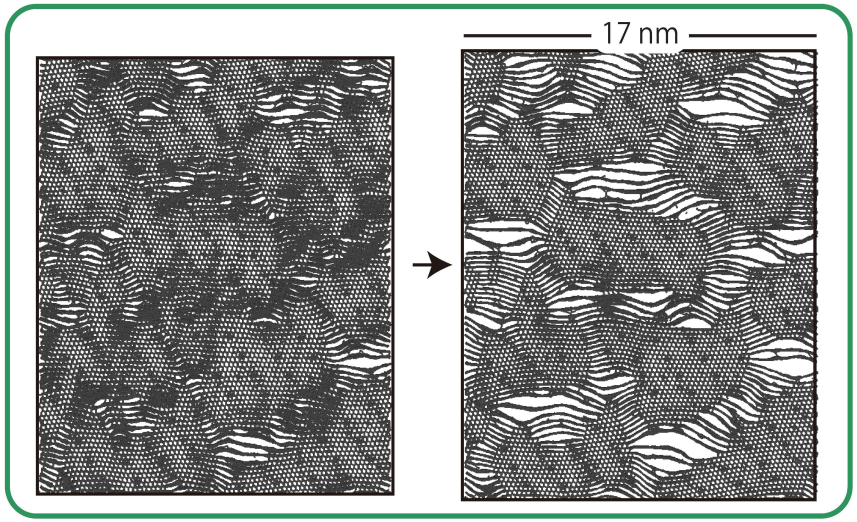
Organic device materials
(with Sumitomo Chemical Co.)



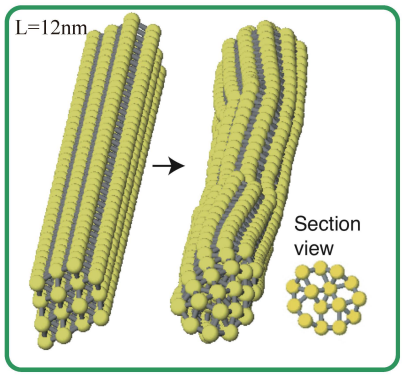
Battery material (with Toyota)



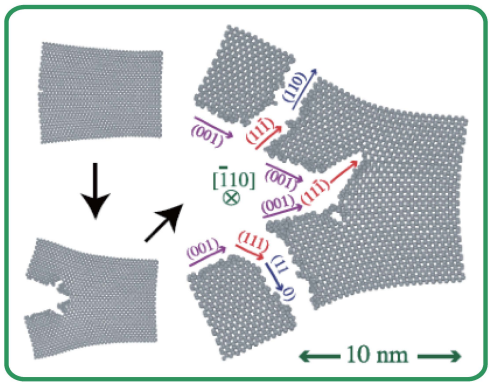
Ultra-hard diamond



helical metal
nanowire



silicon fracture



Detailed method:

Modelled (transferable tight-binding) theory based on first-principles calculations

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).

→ General numerical routine: 'Kω' https://github.com/issp-center-dev/Komega/ (2016)

with Yoshimi, Kawamura,Yamaji (U Tokyo), Sogabe (Nagoya U) et al.

ex. the use with the many-body calculation code 'HΦ' (Yamaji et al.)

[4] (ab initio transport calculation)

S. Iwase, T. Hoshi, T. Ono, Phys. Rev. E 91, 06330 (2015).

[5] (GW calculation)

F. Giustino, M. L. Cohen, S. G. Louie, Phys. Rev. B. 81, 115105 (2010)

[6] (nuclear physics)

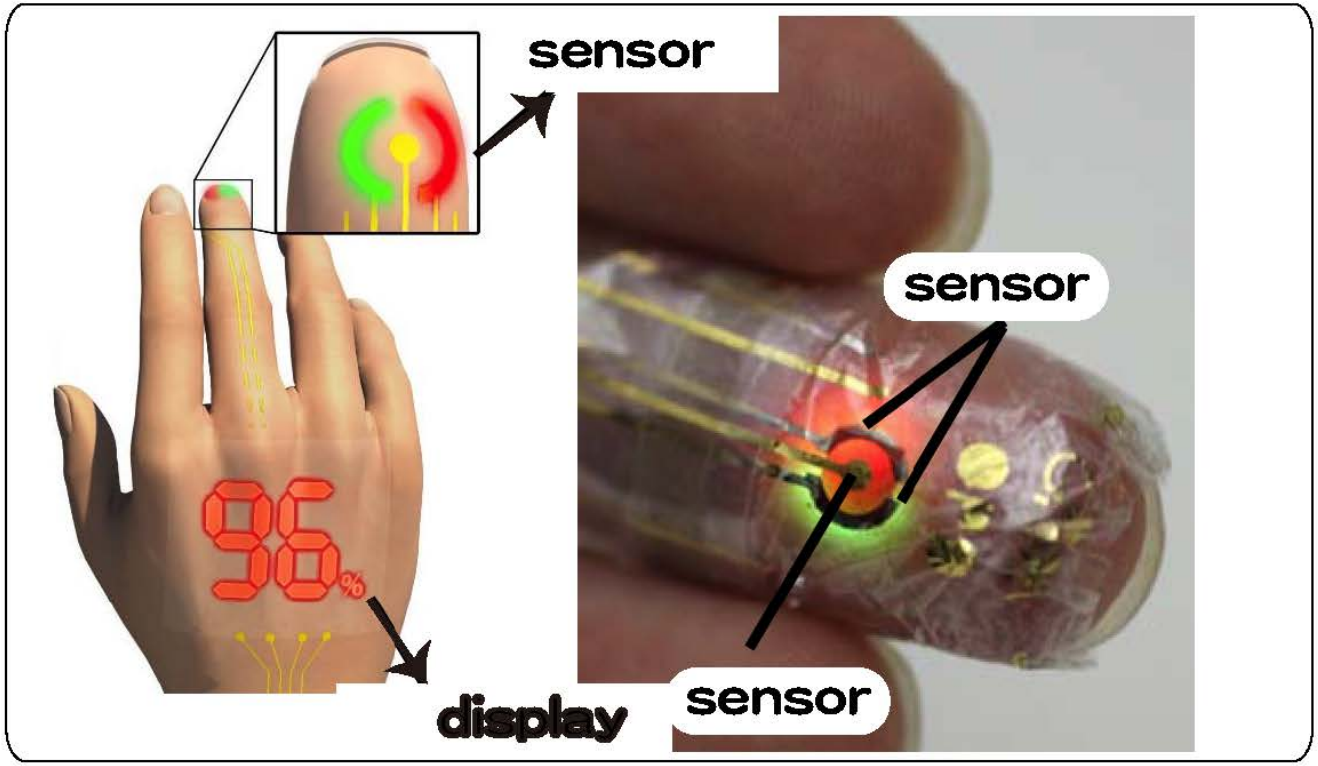
T. Mizusaki, K Kaneko, M. Honma, T. Sakurai, Phys. Rev.C 82, 024310 (2010)

Introduction : organic material for ultra-flexible devices

Organic material gives the foundation of ultra-flexible (wearable) devices of Internet-of-Things (IoT) producs, like display, and sensor

(thickness ≦ 1,000 nm)

Ex. 'Rollable' (ultra-flexible) display,
Sony, News Release, 26. May. (2010)



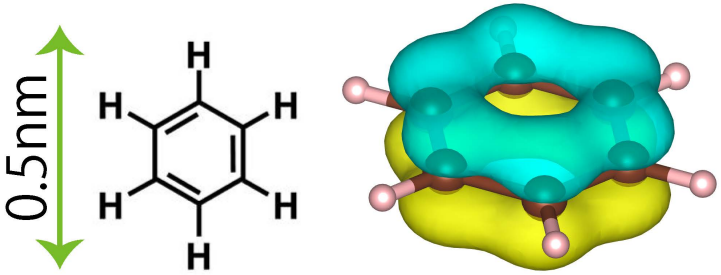
Ex. 'e-skin' (U Tokyo)
T. Yokota, et al., Sci. Adv. 2, e1501856 (2016)
(wearable device monitoring your health)

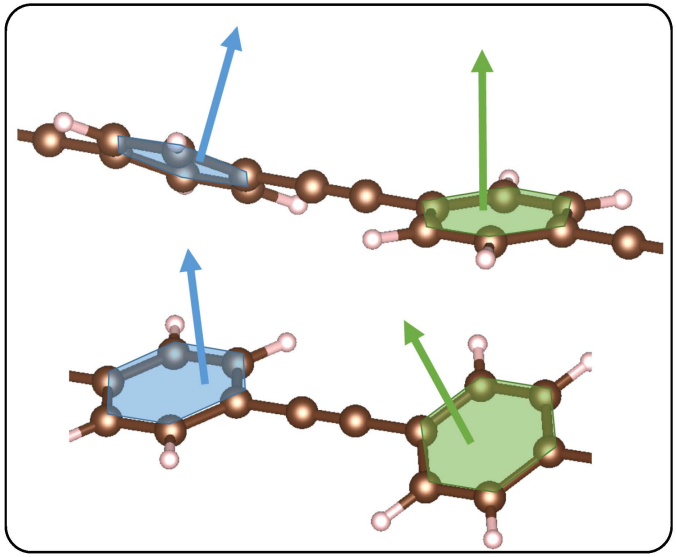
Organic device material and 100-nm-scale quantum simulation

Organic device material

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

→ strong anisotropy



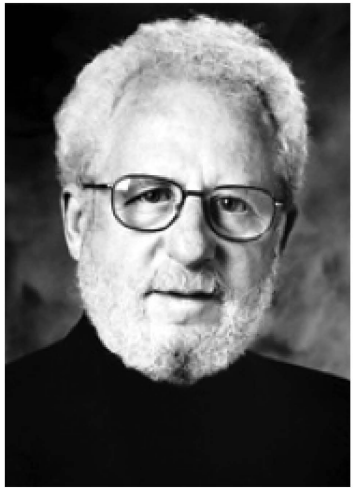


Pioneering experimental research in 1970's

↓

The Nobel Prize in Chemistry 2000:
'discovery and development of conductive polymers'

A. Heeger, A. G. MacDiarmid, H. Shirakawa



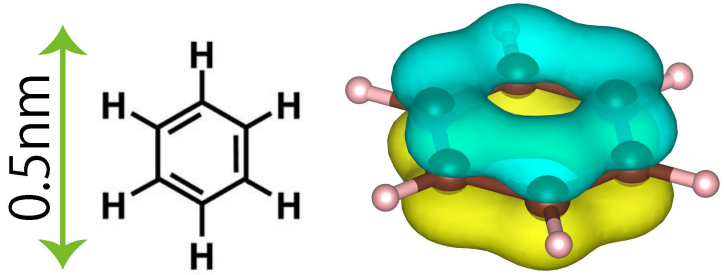
http://www.nobelprize.org/nobel_prizes/chemistry/laureates/2000/

Organic device material and 100-nm-scale quantum simulation

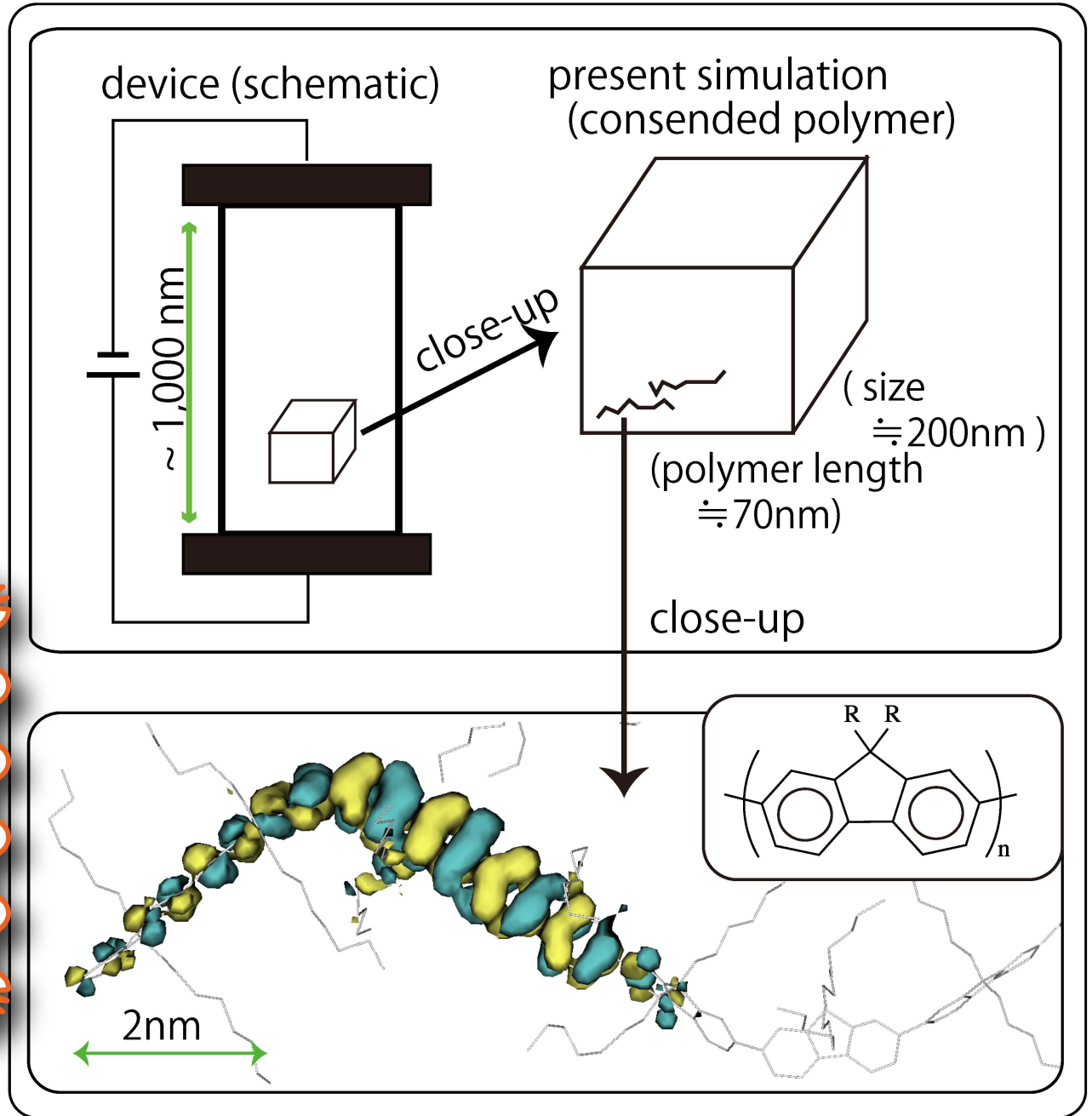
Organic device material → Scales between device and 100-nm-scale simulation

- semiconductor with ‘π-type’ electronic wave that lines in benzene rings and so on

→ strong anisotropy



Industrial materials are disoredered system in 100-nm-scale or with 100-million-atoms



Quantum (wavepacket) dynamics for device simulation

Non-equillibrium, non-stationary simulation

Solving an effective Schrodinger-type equation (details are not yet settled)

$$i\frac{\partial \psi}{\partial t} = H\psi \quad (1)$$

$$\psi \equiv \psi(\boldsymbol{r}, t) \quad (2)$$

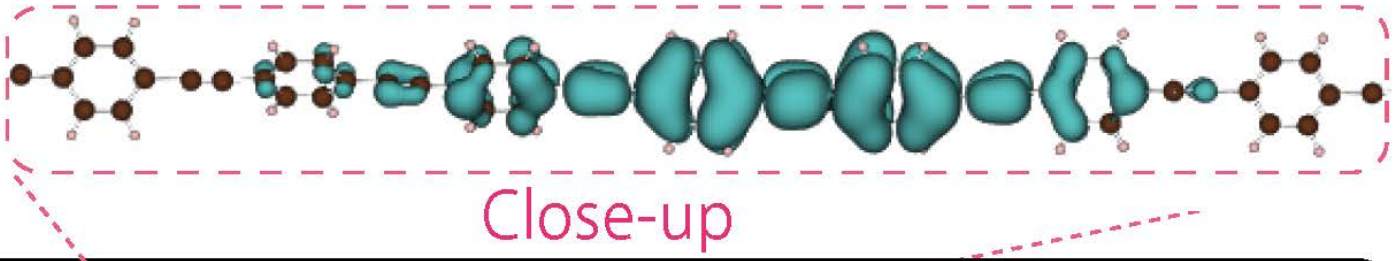
: electronic 'wave'
(ex. hole wave packet)

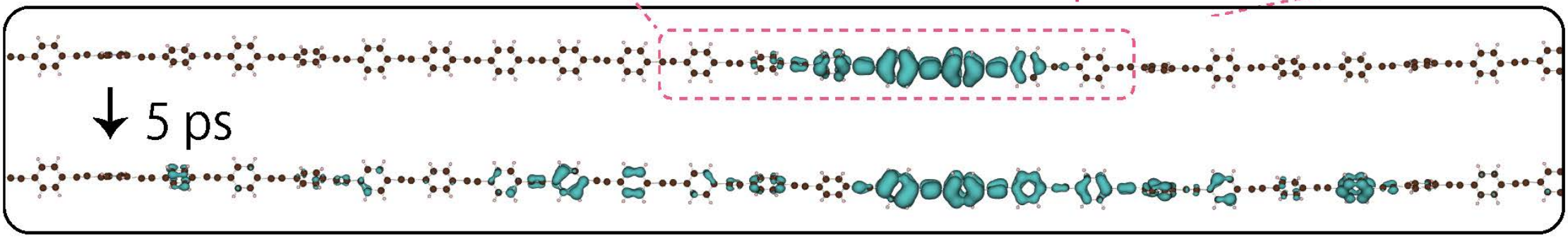
$$\rho(\boldsymbol{r}, t) \equiv |\psi(\boldsymbol{r}, t)|^2 \quad (3)$$

: charge distribution (real value)

→ calculation of
device performance
or 'mobility' value

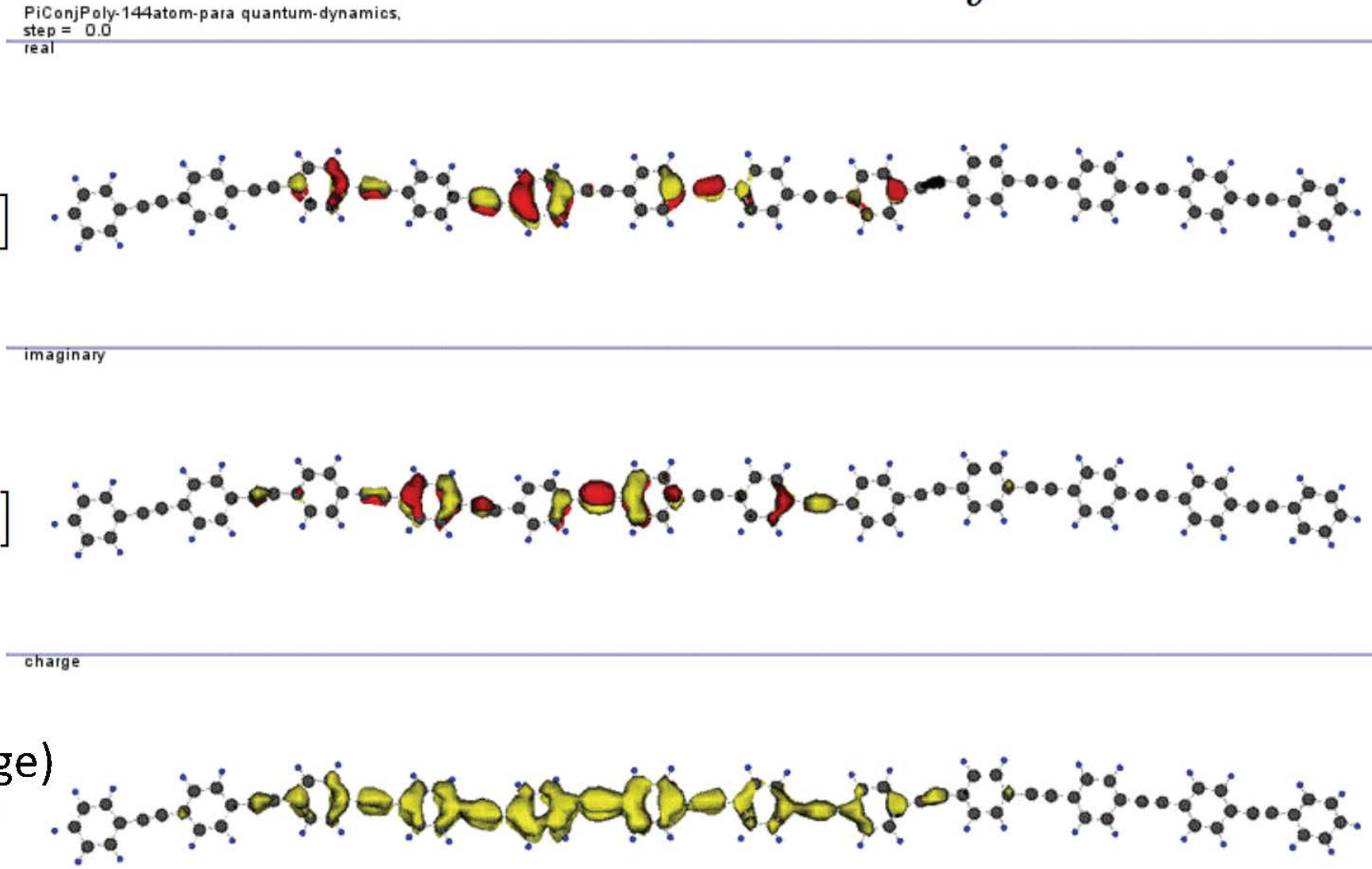
Example of a partial region of a polymer;





Example of wavepacket simulation

$$\partial_t \Psi = -iH\Psi$$

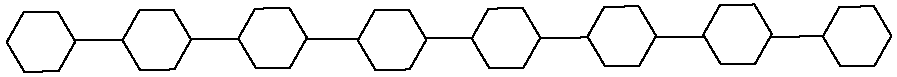


Comparizon between linear and zigzag chain structures

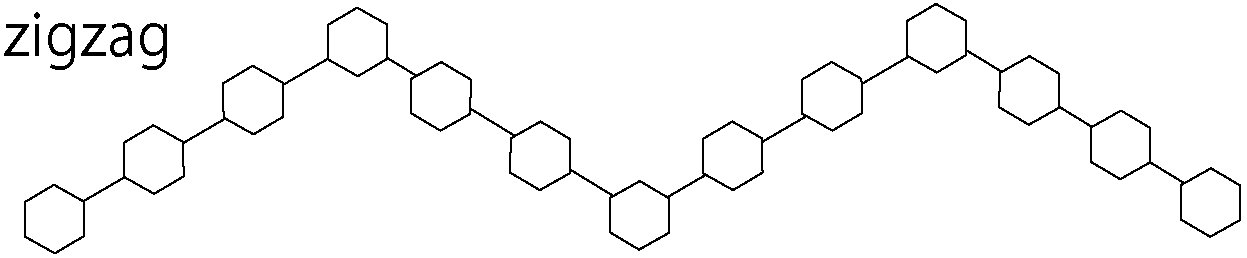
poly(phenylene–ethynylene) ([1] J. Terao, et al, Nature Comm. 4, 1691 (2013))

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

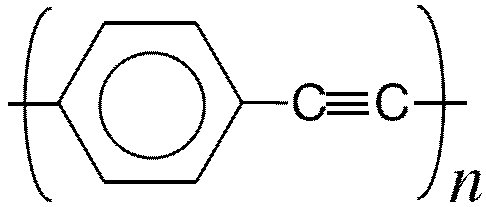
linear



zigzag



example (linear chain)



Result

mobility in ideal structure
(linear) > (zigzag)

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

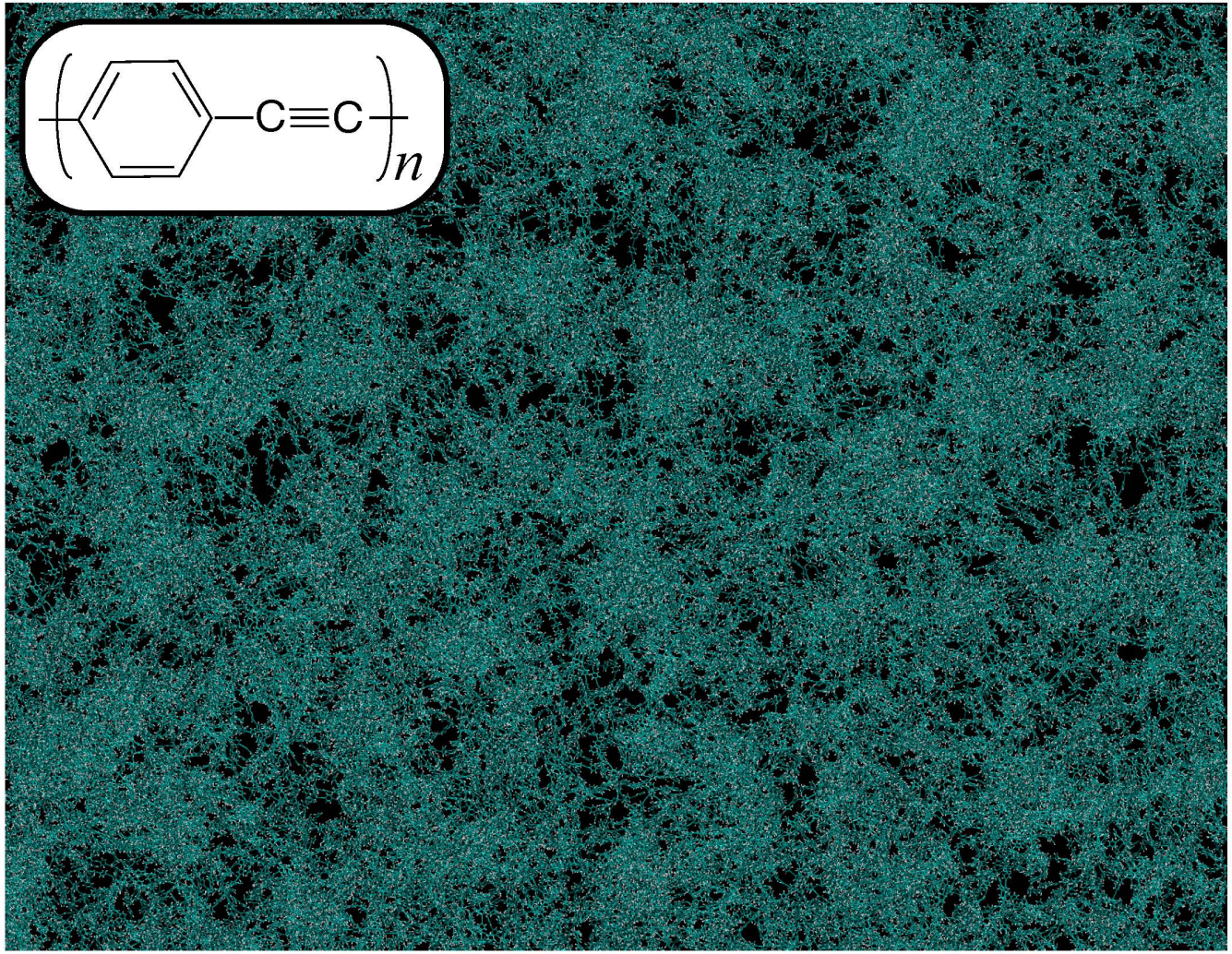
100-million-atom caulation of condensed organic polymers

Investigation on wave propagation mechanism (for device property)

(a) material：poly-(phenylene-ethynylene) (PPE)

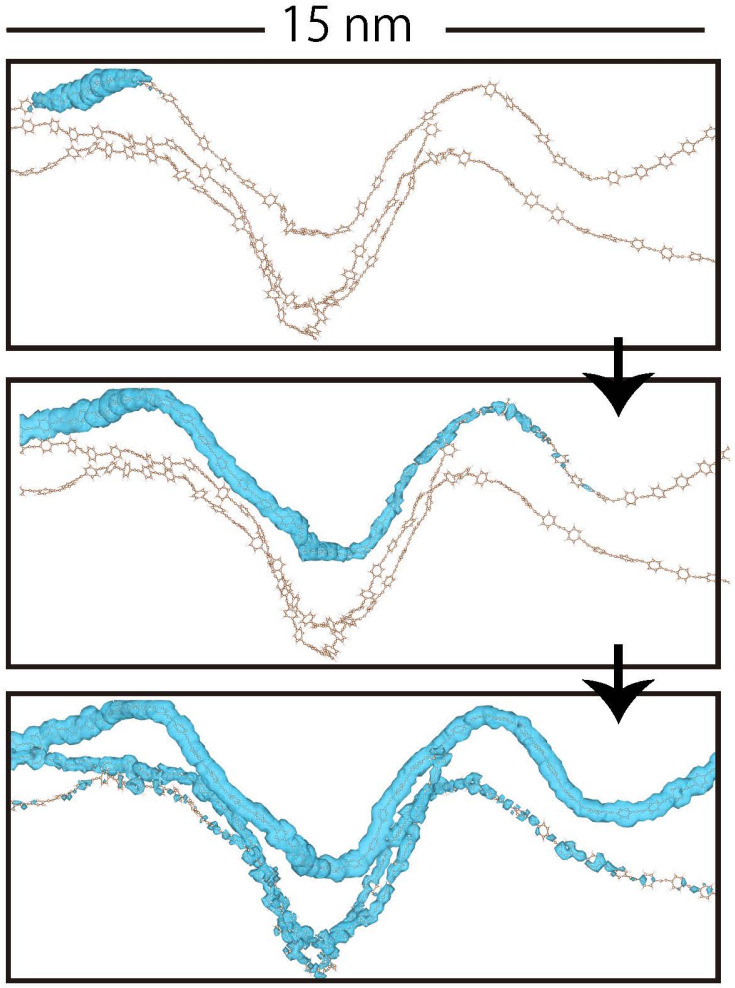
(size≒200nm, P=100,000 polymers)

Figure of a partial retion (~50nm)



(b) Quantum dynamics

Fig: Charge dynamics with 1 ps



Opt-electronics device simulations

with T. Fujita (IMS) and Y. Mochizuki (Rikkyo U)

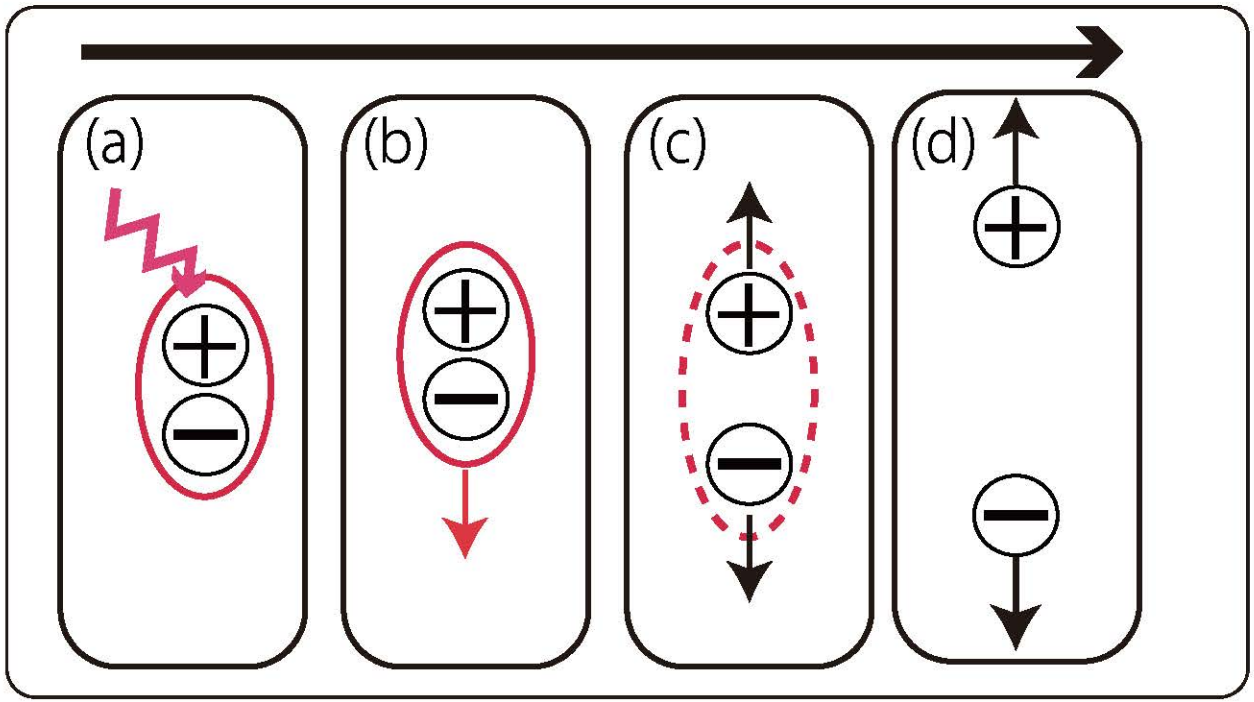
Four elementary processes

(a) Creation of exiton (electron-hole pair) by light absorption

(b) Exciton dynamics (diffusion)

(c) Dissociation of exiton into carriers (hole and electron)

(d) Diffusion of carriers



Two types of device

(i) sensor type
(light→current),

(ii) display type
(current→light)

Opt-electronics device simulations

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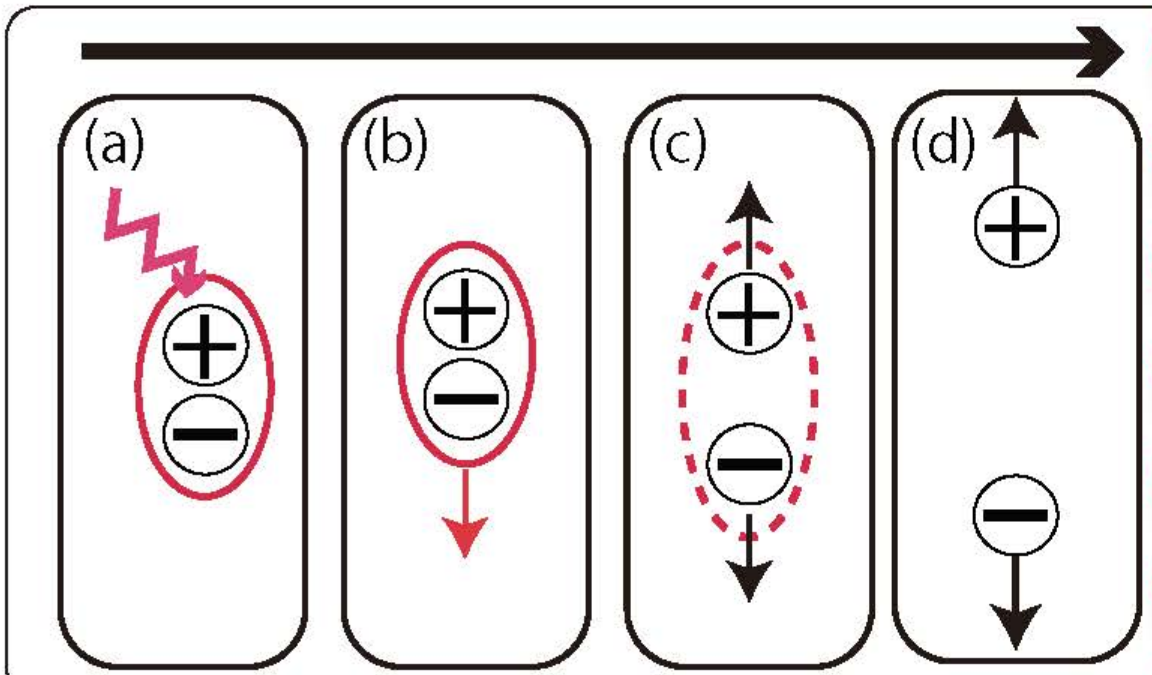


(b) Two-body wave dynamics

$$\dot{\boldsymbol{u}} = -iH\boldsymbol{u}$$

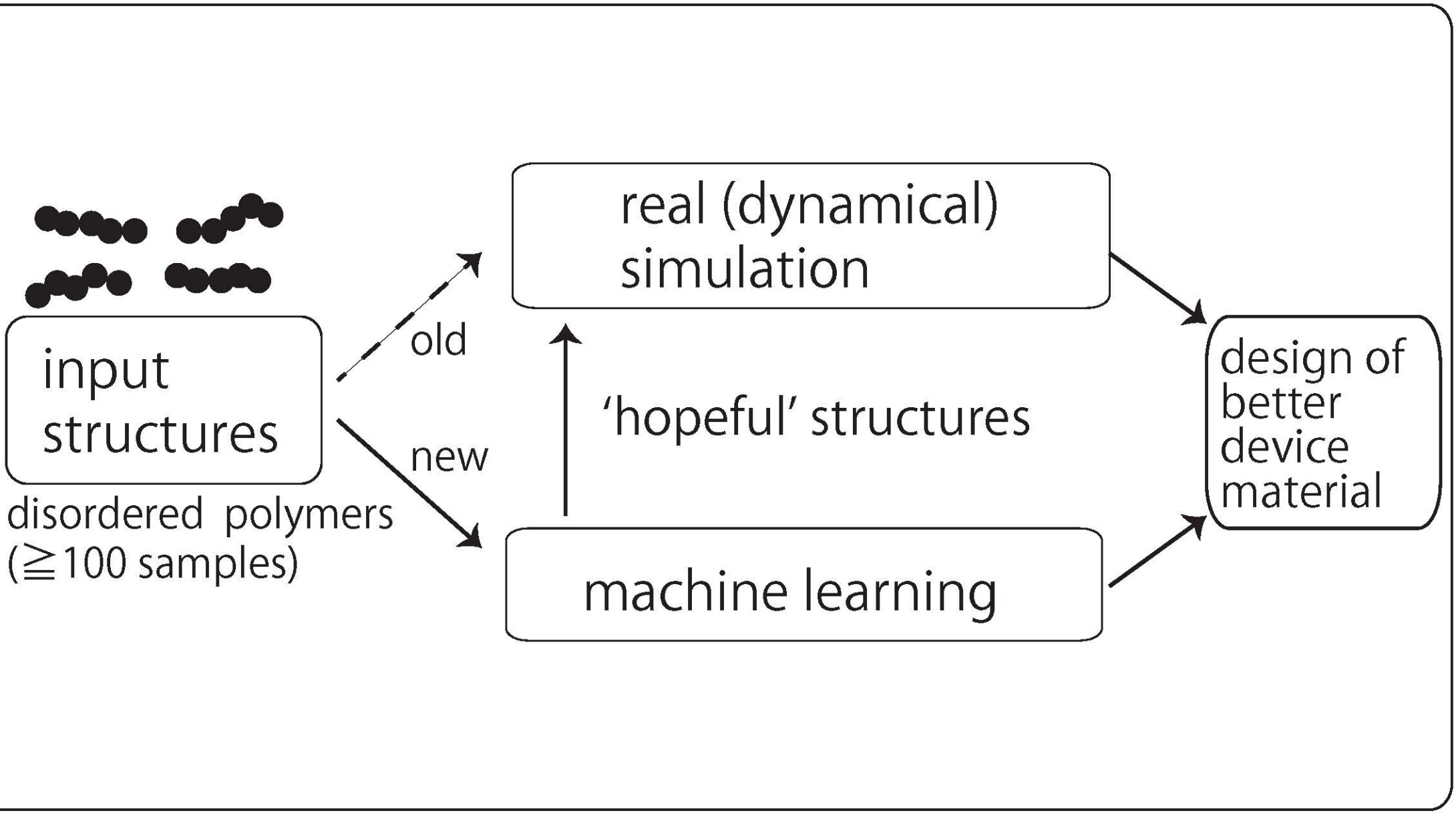
(matrix size) $\propto$ (# atom)^2

(d) One-body wave dynamics
(in previous section)

(matrix size) $\propto$ (# atom)

Machine learning analysis on organic polymers (1/2)

with Weichung Wang's group (National Taiwan U)



Machine learning analysis on organic polymers (2/2)

with Weichung Wang's group (National Taiwan U)

200 samples of disordered polymers
→ classification for higher or lower device performace (mobility value)

samples
(200 structures)

↓ Eigen value analysis

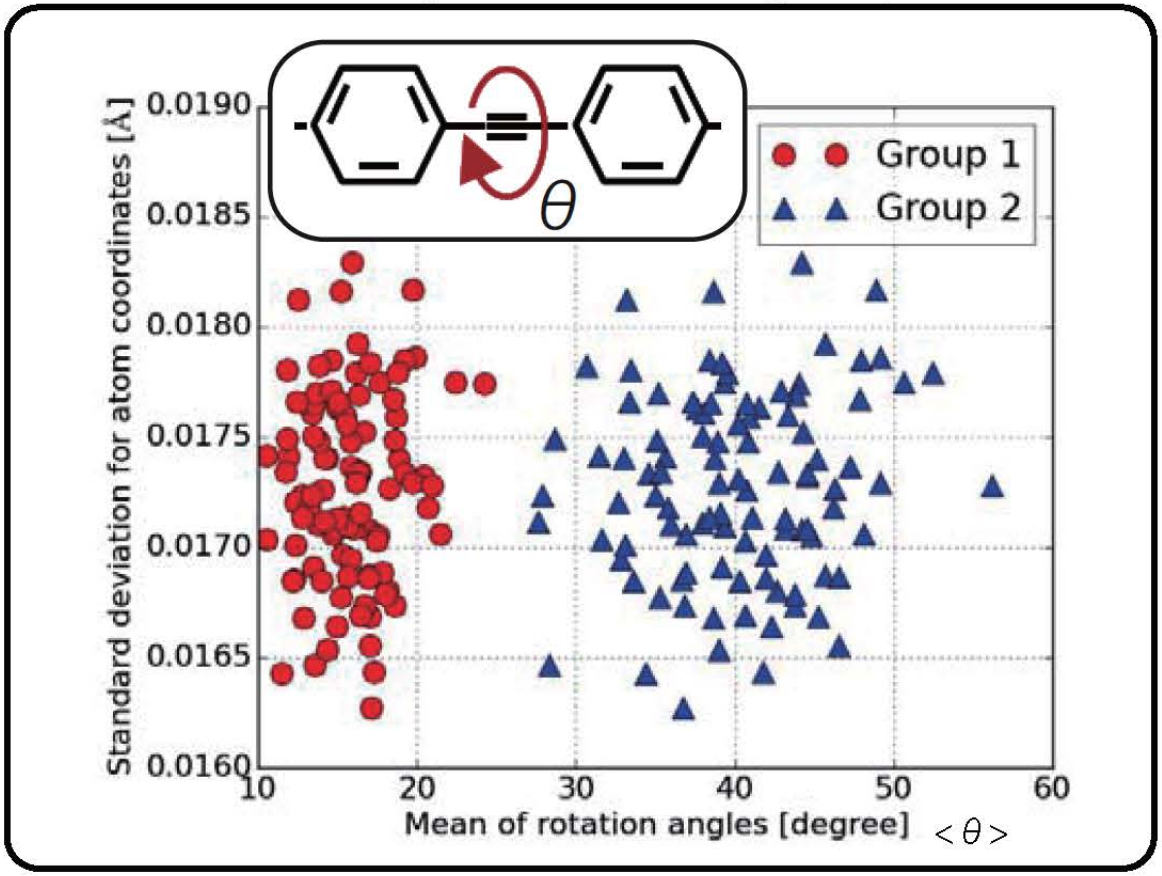
Participation Ratio
diagram

↓ Use as descriptor

Machine learning

(k-means clustering method)

Classification of 200 samples into two groups successfully



本発表のまとめ：有機デバイス材料の量子シミュレーション

1. アルゴリズム
   →応用・数理系研究者で、問題・コードの共有
2. アプリケーション
3. 現在進行形の話(*)
   ・光電子デバイス、機械学習の融合

→ 行列方程式

$$A\boldsymbol{y} = \lambda B\boldsymbol{y}$$
$$(zB - A)\boldsymbol{x} = \boldsymbol{b}$$
$$\dot{\boldsymbol{u}} = -iH\boldsymbol{u}$$

(*) 12/6-7,ポスト「京」重点課題7研究会(東大物性研)等で発表

大局的なまとめ

1. ニーズ志向型解法
   - 俯瞰的視点：　物理理論→計算系(行列など)→数値計算→応用
     (例：本講演での高分子集合体→「ほぼ」ブロック対角行列)
   - 数理構造がガイドライン
2. 大規模シミュレーションと機械学習の融合
   - コードレベルの融合：すべてをスパコン上でオンライン処理
     大規模シミュレーション(big data生成)→記述子抽出→機械学習
   - 精度可変型可変型数値計算：新しい需要
     「少数の高精度計算」⇔「多数の低精度計算」