# 次世代エネルギーシステムを拓く新材料開発技術

## ～計算科学を用いたハイスループットスクリーニング～

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# 脱炭素化社会を目指して

産官学一体となった脱炭素化への機運が高まっている。

2019年 6月 「パリ協定に基づく成長戦略としての長期戦略」[1]

2019年12月 脱炭素社会の構想「チャレンジ・ゼロ」(経団連) [2]

再生可能エネルギーの大量導入など、技術的なブレークスルーとエネルギー業界の構造変革が求められている[3]

① 運輸 (電池, 燃料電池, バイオ燃料)
② 産業 (非化石燃料)
③ 業務/家庭 (IoT, ZEB, 熱利用, 水素利用)
④ **電力分野のイノベーション** (再エネ・既存発電, 蓄電・系統)

[1] https://www.env.go.jp/press/106869.html(環境省), [2] https://www.keidanren.or.jp/policy(経団連),
[3] https://www.enecho.meti.go.jp/committee/studygroup/ene_situation/006/pdf/006_011.pdf (経産省)



# 世界における電力分野への投資額

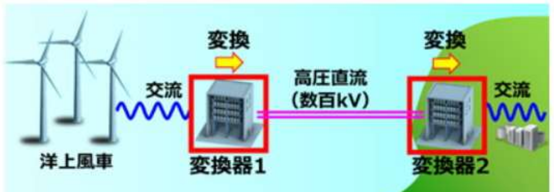

Source: IEA Flagship report May 2019 power sector, https://www.iea.org/reports/world-energy-investment-2019/power-sector#abstract



# 次世代電力ネットワークの要素(技術)

**電力機器の例**

- DC用ケーブル
- パワー半導体
- DC遮断器




https://www.nedo.go.jp/news/press/AA5_100919.html

**電気絶縁への要求**

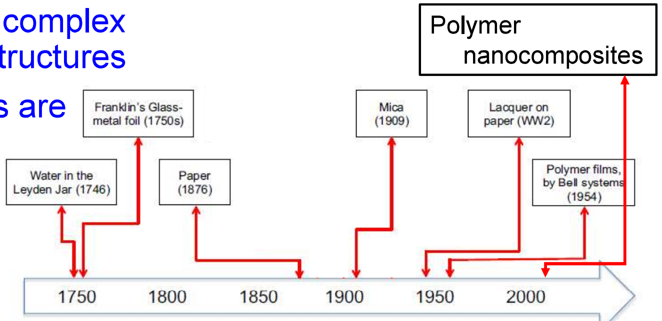
- 高電界化 (コンパクト化・高電圧化)
- 高耐熱・高熱伝導化
- 高環境調和化

(次世代交流-直流連系電力ネットワークには)
新絶縁材料(気体・液体・固体)が必須

## History of Electric Power Transmission
## ~ History of dielectric development

Dielectric materials development is slow because…

- dielectrics have complex morphological structures
- High field effects are complicated




T. D. Huan et al., Prog. Mat. Sci., 83, 236, (2016)

➡ **Need to accelerate materials development**

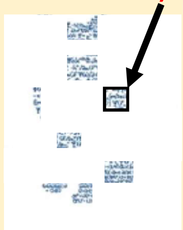
**We are in the industrial age of first principles modeling and Big-data analytics !**



## Computational materials design

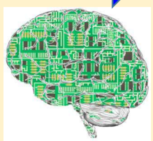
**High-throughput materials screening**

**First-principles (Deductive)**



https://www.slideshare.net/CanOz doruk/tera-chem-heatherkulik

**Machine learning (Inductive)**



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**Accelerated**
- **Materials discovery**
- **Materials design**

https://www.slideshare.net/CanOz doruk/tera-chem-heatherkulik

- Understanding the origin of the characteristic electrical properties (Basic research)
- Predict and design new materials (Applied science)



## Impact beyond academia
## -potential applications -

**Dielectrics**
- Electric power equipment
- Energy storage
- Remote wireless sensing device
- …

**Inorganic semiconductors**
- Water splitting
- Solar to Fuel
- …

**Organic semiconductors**
- Flexible device

http://flode-design.com/?p=1026

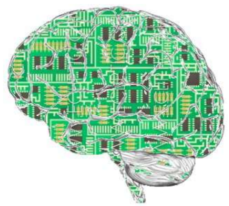
**Designing the electrical properties using first-principles calculations and machine learning**

## Today's outline

1. First principles based modeling of charge transfer in polymers (**main topic**)
   - Multi-scale modeling approach for charge transfer in polymers
   - Charge injection from electrodes to polymers

$$\hat{H}\Psi = E\Psi$$

2. Prediction of the physical quantities of gases
   - Computation of non-linear optical properties from first-principles
   - Prediction of electric breakdown strength of gases using first-principles calculation and machine learning



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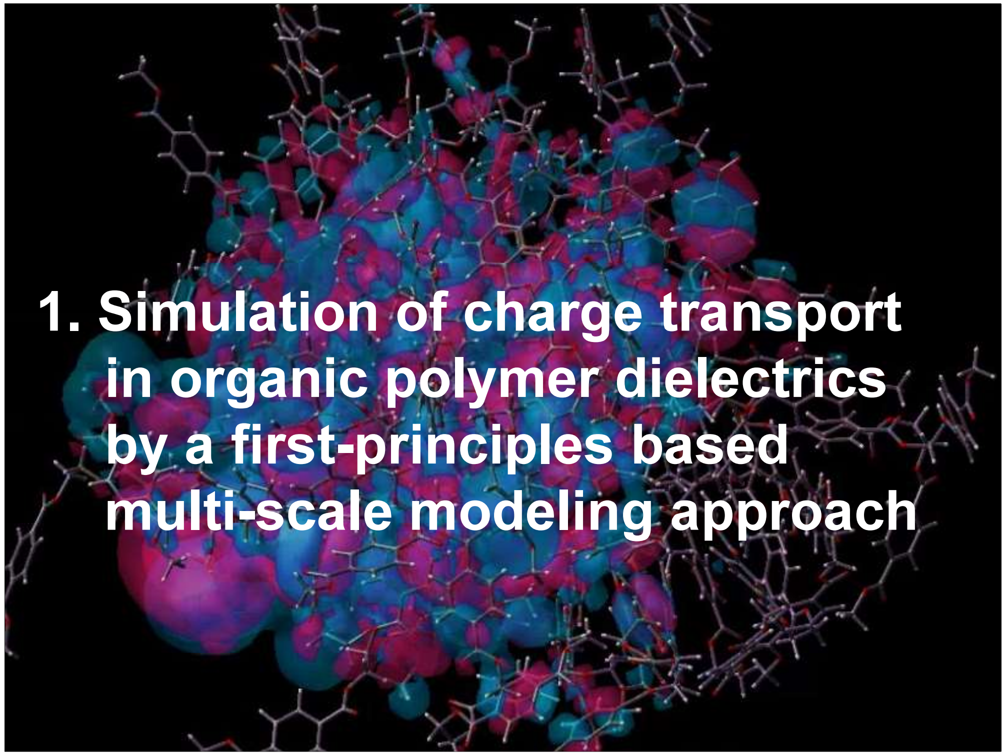
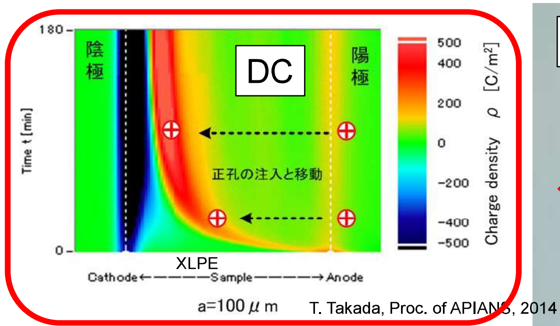
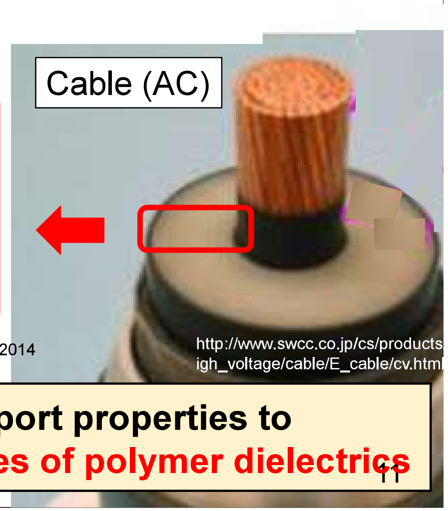


# 1. Simulation of charge transport in organic polymer dielectrics by a first-principles based multi-scale modeling approach

## Why charge transport in polymers ?

- Charge transport in polymers degrade the material
- In DC applications, space charge formation leads to Field enhancement



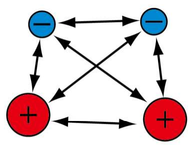




**Characterize the charge transport properties to tailor the electrical properties of polymer dielectrics**

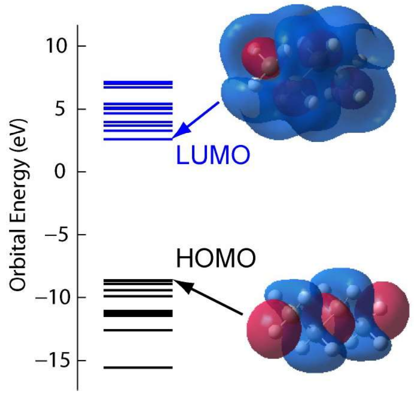


## What is First-principles calculation

$$\hat{H}\Psi = E\Psi$$



$$\hat{H}_{tot}(\boldsymbol{r},\boldsymbol{R}) = T_N + T_e + V_{NN} + V_{eN} + V_{ee} = \sum_{k=1}^{M}\frac{p_k^2}{2m_k} + \sum_{i=1}^{N}\frac{p_i^2}{2m_e} + \sum_{k>l}\frac{Z_k Z_l e^2}{R_{kl}} - \sum_{k,i}\frac{Z_k e^2}{r_{ki}} + \sum_{i>j}\frac{e^2}{r_{ij}}$$



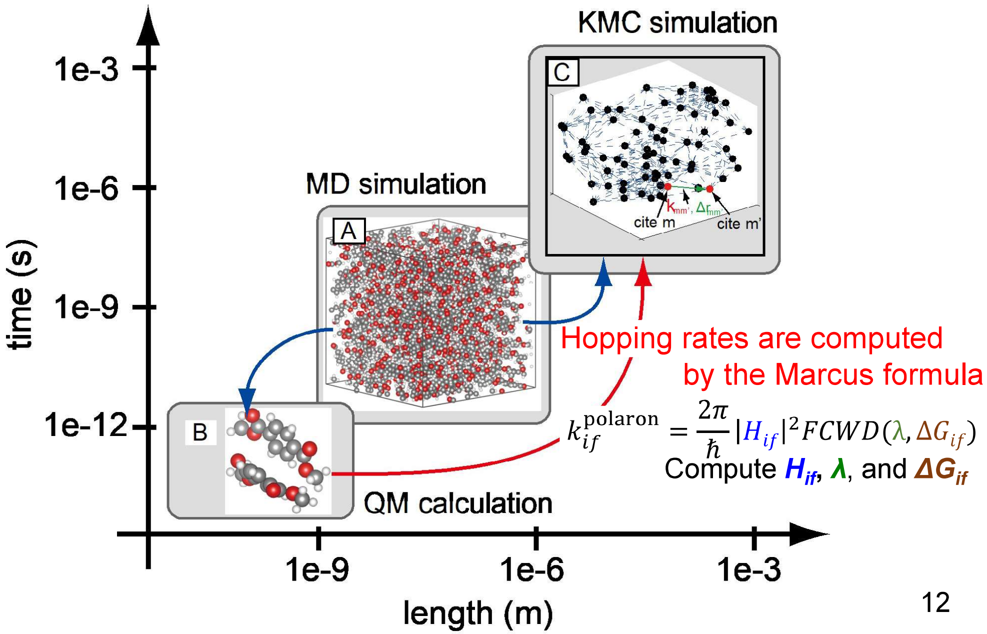

Input :
coordinates and number of the atom

Output :
Eigenvalues, eigenvectors

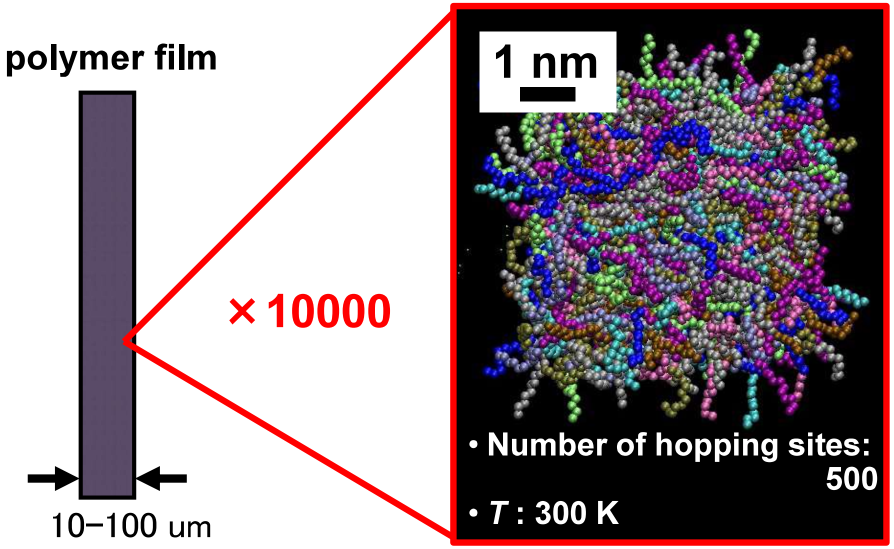
**No assumptions such as empirical model and fitting parameters**



## Multi-scale modeling method

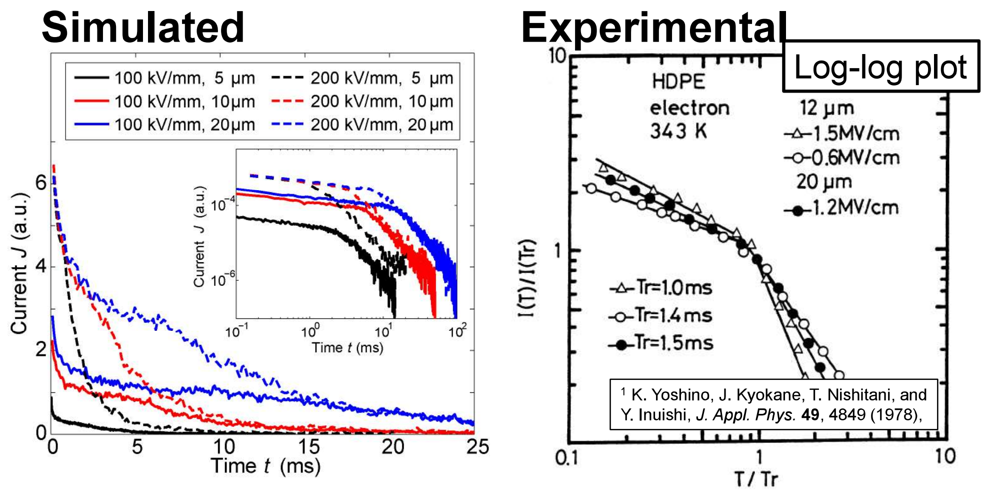


# Amorphous structure of PE





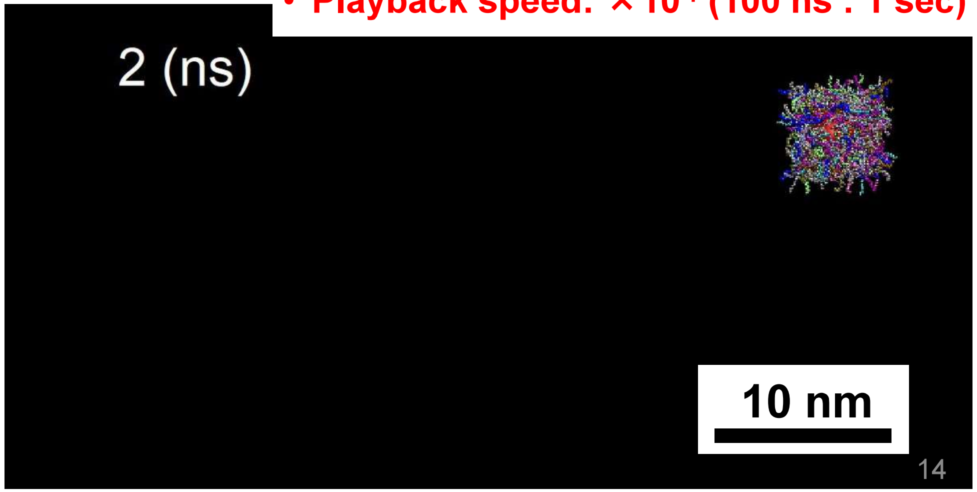

# Typical outcomes (Current waveform)




**Current waveforms can be simulated from first principles!**
**(without adopting empirical parameters)**

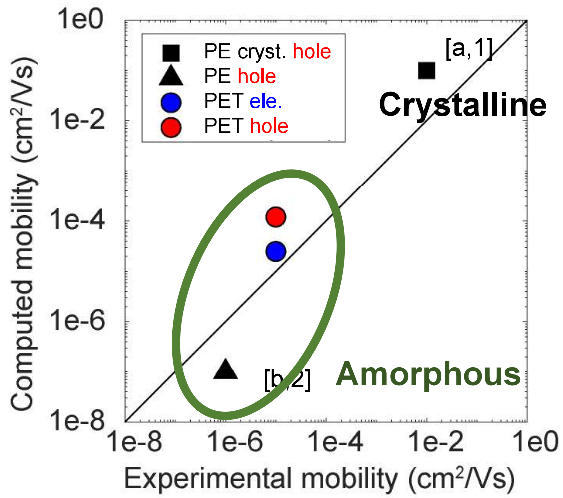
# Typical outcomes (KMC simulation)

- **Simulated distance : 50 nm**
- **50 kV/mm**
- **300 K**
- **Playback speed: ×$10^{-7}$ (100 ns : 1 sec)**






# Computed electron and hole mobilities



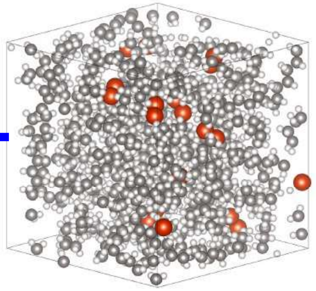

Experimental: [a] M. R. Belmont et al., *J. Phys. Chem. Solids* **46**, 607 (1985), [b] H. Neff and P. Lange, *J. Appl. Phys.* **72**, 4369 (1992), [c] K. Yoshino, J. Kyokane, T. Nishitani, and Y. Inuishi, *J. Appl. Phys.* **49**, 4849 (1978)

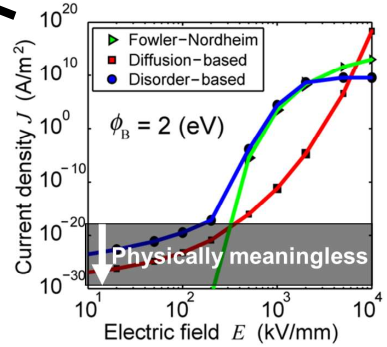
Computed (PE): [1] M. Sato, A. Kumada, K. Hidaka, T. Hirano, and F. Sato, *Appl. Phys. Lett.*, **110**, 092903, (2017), [2] M. Sato, A. Kumada, and K. Hidaka, *Phys. Chem. Chem. Phys.*, **21**, 1812-1819, (2019)

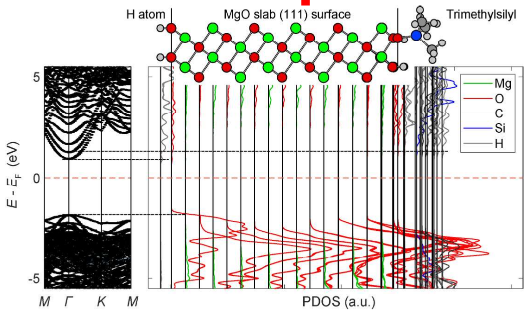
- Excellent agreement with experiments!
- The multi-scale simulation method **makes it possible to predict the carrier transfer properties**.

# 高分子材料中電荷輸送

1. 不純物や添加材の効果
2. 高分子材料の変性の効果
3. 電極からの電荷注入
4. ポリマーナノコンポジット材料の特性予測



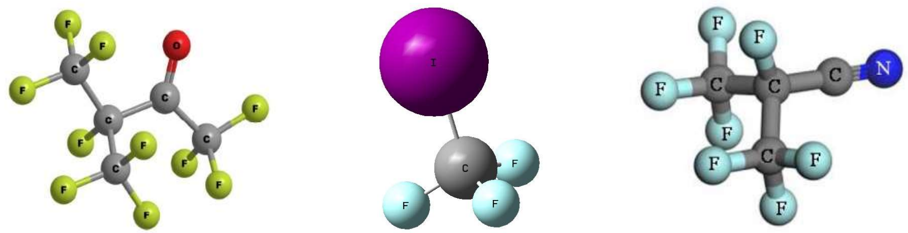




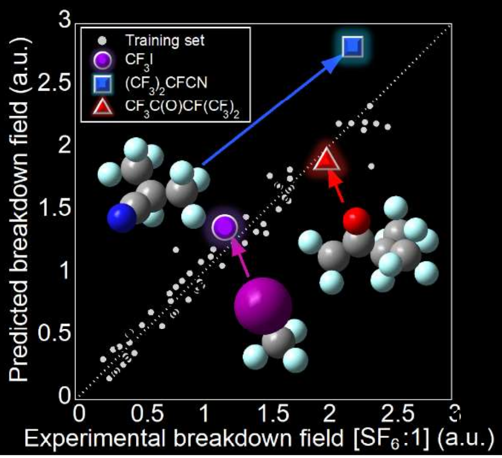

# $SF_6$ Alternatives

- $SF_6$ was included in the six greenhouse gases covered by the Kyoto Protocol
- The life-cycle cost of $SF_6$ insulation is subject to review
- $SF_6$ alternatives have been explored continuously.
- **There are vast number of gases, *in silico* materials screening is promising for seeking alternative gases.**

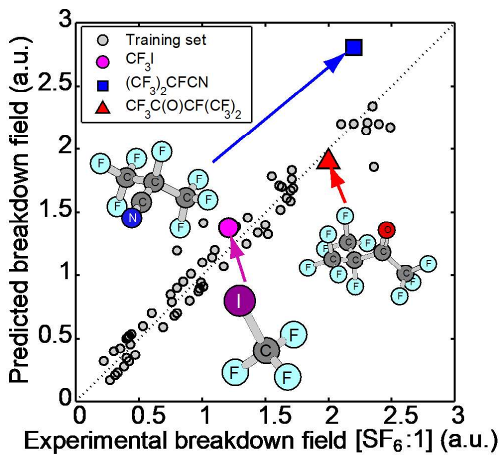


# 2. Prediction of electric breakdown strength and boiling point of gases using first-principles calculation and machine learning

絶縁破壊電界の
第一原理的計算は困難




# Prediction of $E_{BD}$ of $SF_6$ alternatives




Predictors $\alpha, \mu, \varepsilon_i^a, \varepsilon_a^v, \Delta\varepsilon_a$

(computed with the aid of first-principles calculations)

Molecules with N and I atoms are excluded from the training set

Training dataset size ~ 50

**高精度な絶縁破壊電界予測に成功！！**

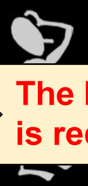
**$E_{BD}$ : 30%, Boiling point : 10 %**

# [3.補遺] 水分解を用いた水素製造
## -電極・触媒材料の計算機による設計に向けて-

(光)電極による水分解
- Carbon-neutral
- Can be isolated from the grid

① 運輸 (電池, **燃料電池**)
② 産業 (**非化石燃料**)
③ 業務/家庭 (IoT, **水素利用**)
④ 電力分野のイノベーション
(再エネ・既存発電, **蓄電・系統**)

How can we develop efficient and stable photocatalysts and (photo)electrodes



⇨ **The knowledge of the physical basis of PEC reactions is required to tailor the properties of photo-electrodes.**

# Characterization of semiconductor/electrolyte interface

**Experiment, Synthesis ⇄ Theory, Atomistic modeling**

| | Geometric Structure | Electronic Structure |
|---|---|---|
| Surface | TEM, SEM, AFM, LEED | UPS, XPS |
| Adsorption | FTIR | AP-XPS |
| Interface | FTIR | OCP |

DFT

**This talk will focus on DFT and AP-XPS results.**



# What we need to know -the known unknowns-

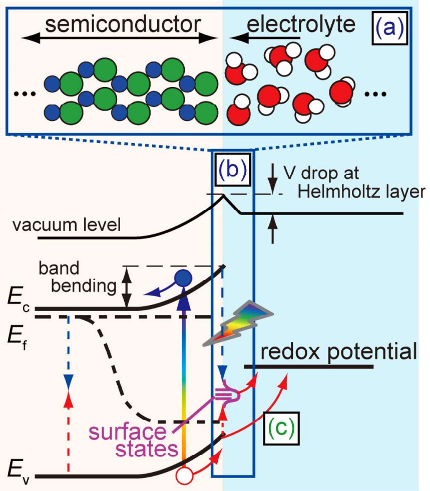
Prepare Photocatalyst ⇨ Geometric structure / Electronic structure ⇨ Charge transfer / Chemical reaction ⇨ Efficiency / Durability

?

**a. Geometric structure**
- ➢Linked to the electronic structure

**b. Electronic structure**
- ➢Band alignment
(Potential shift, surface state pinning)

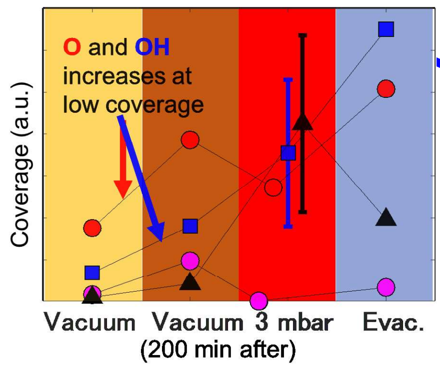
**c. CT, chemical reactions**
- ➢Reaction kinetics
(Selectivity, rate limiting step)




⇨ **Atomistic characterization of the "(a) Interface structure" and "(b) Band alignment"**

# 界面構造の実験結果と予測結果の比較

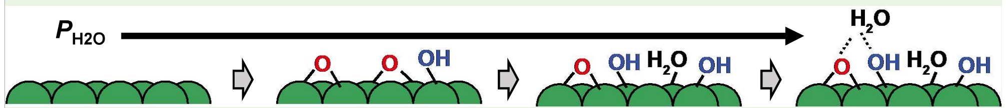
AP-XPS実験結果 (SPRING-8)




OH increases at higher coverage

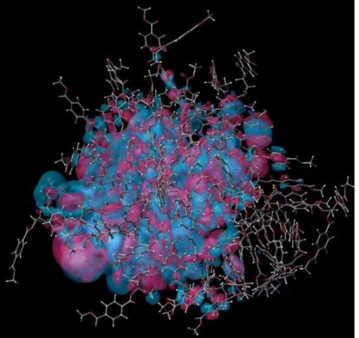
**計算機の中で、触媒/液体の界面構造予測に成功！**

第一原理計算による予測結果

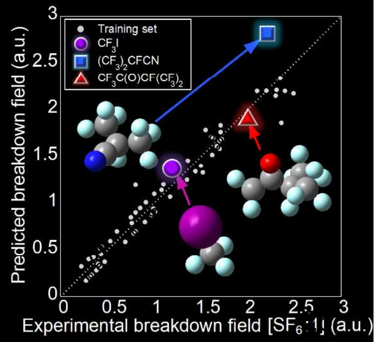


# Summary

- ◆ Charge transfer and injection was simulated from first-principles
  - The multi-scale simulation method makes it possible to predict the carrier transfer properties



- ◆ Successfully predicted the dielectric breakdown field of gases by combining Machine learning and First-principles calculation
  - The appropriate combination of first-principles approach and machine learning techniques will offer a rational approach for materials design.

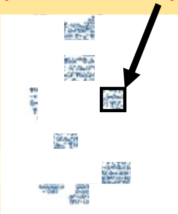



# Prospective

## -computational dielectric design-

**High-throughput materials screening**

**First-principles (Deductive)**



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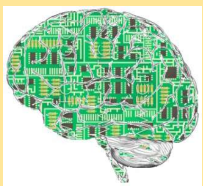
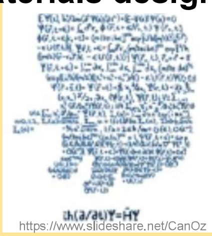
**Machine learning (Inductive)**



https://ja.wikipedia.org/wiki/%E4%BA%BA%E5%B7%A5%E7%9F%A5%E8%83%BD

**Accelerated**
- **Materials discovery**
- **Materials design**



https://www.slideshare.net/CanOzdoruk/tera-chem-heatherkulik

- **First-principles dielectric design has become realistic!**
- **Machine learning can be used together to beyond the limit.**

**Thank you for your kind attention!**