2015年7月27日＠KEK

物質の新しい素励起発見を目指して

~ 光誘起相転移研究の立場から〜

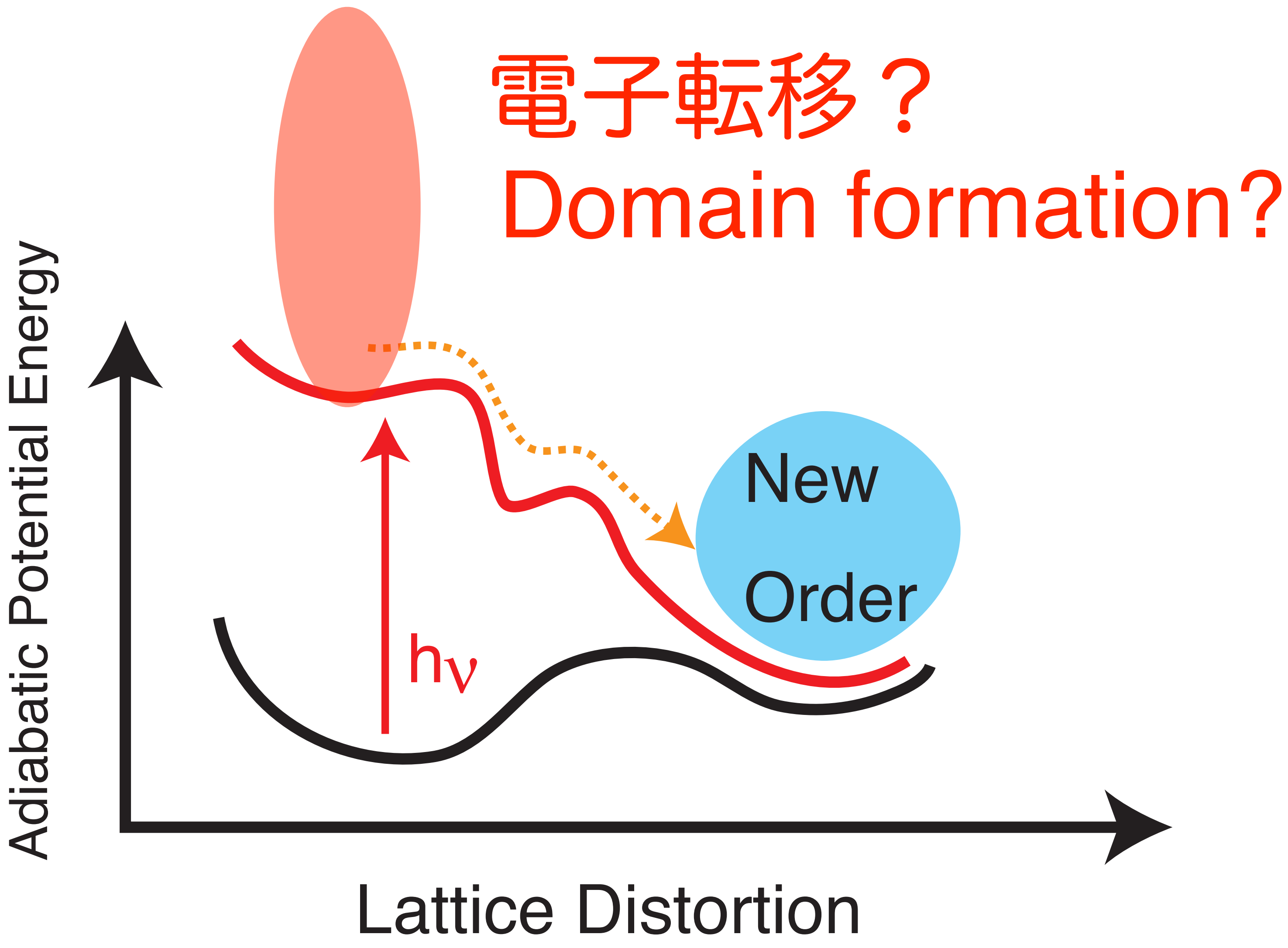
KEK物構研　　岩野薫

背景

光誘起相転移＝光で系の性質を巨視的に変化させる
本talkの興味→超高速(~ fs)なドメイン形成

1次元系：
理論◎、実験△

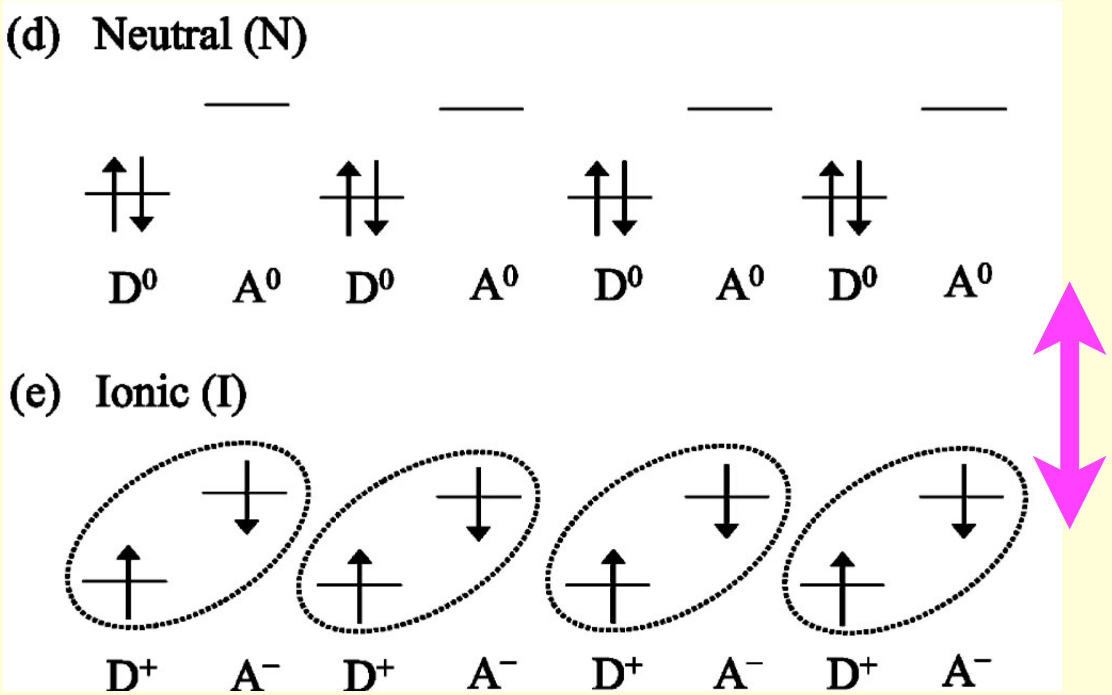
2次元系：
理論○、実験△



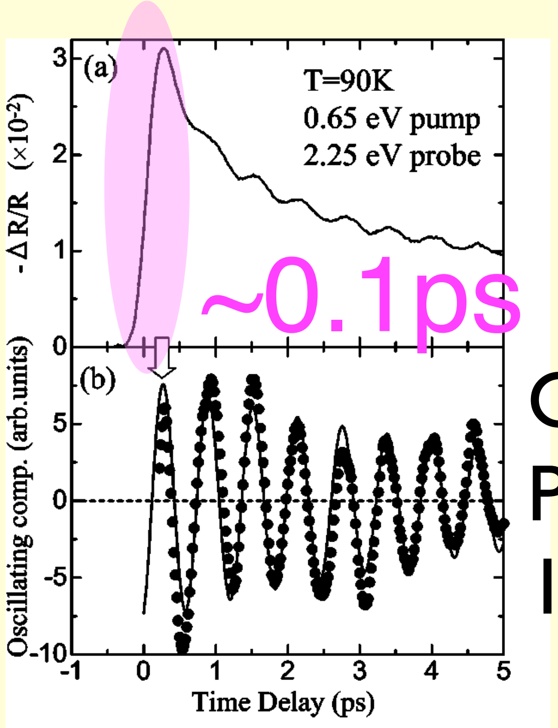
電子自由度と格子自由度の分離

光誘起相転移の超高速性

TTF-CA



PIPT

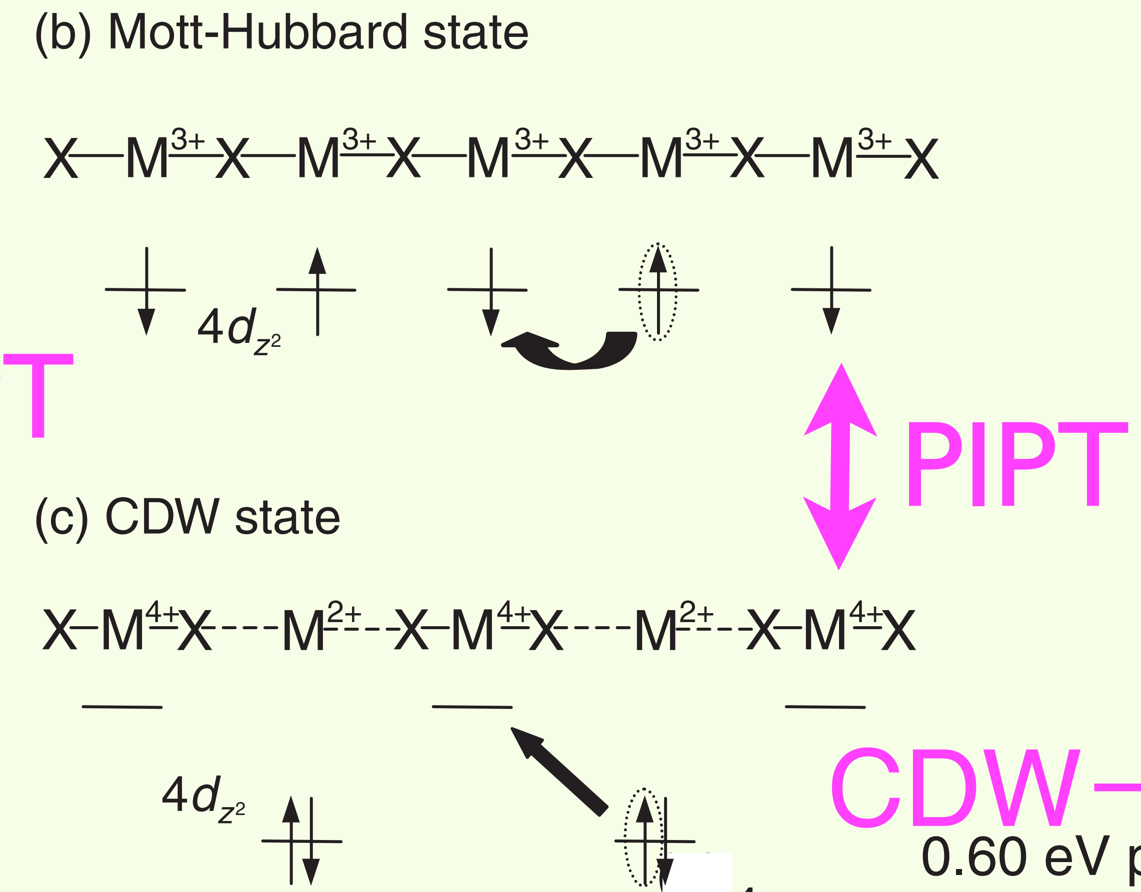


~0.1ps

N→I

Okamoto et al., PRB 70 (2004) 165202

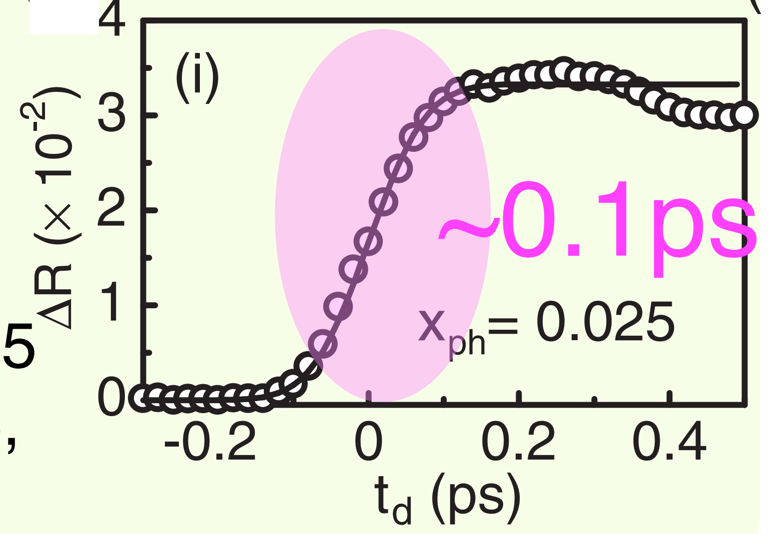
Br-bridged Pd complex



PIPT

CDW→MH

0.60 eV probe



~0.1ps

Matsuzaki et al., JPSJ 75 (2007)123701&PRL113, 096403 (2014)

Δeff=Δ-4V, and after some rewriting,

$$H = -t_0 \sum_{l\sigma} (C^{\dagger}_{l\sigma} C_{l+1\sigma} + H.c.)$$

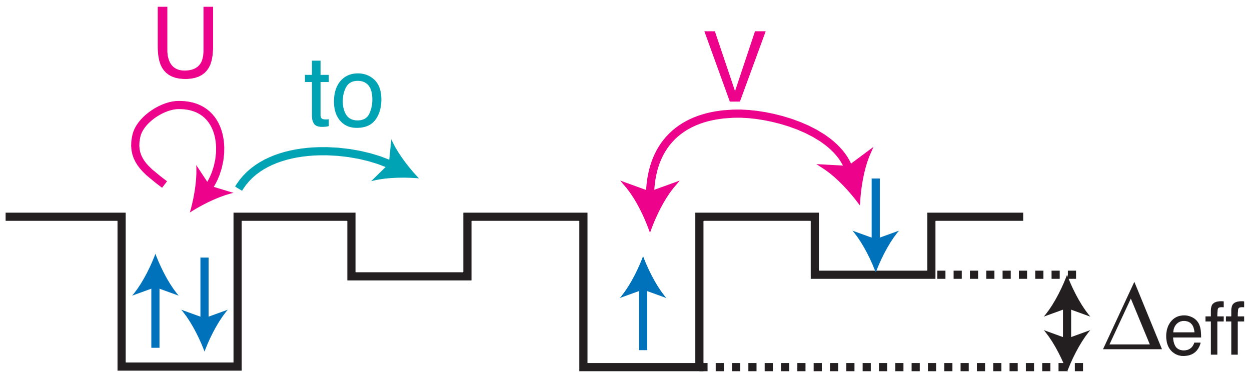
Electron Hopping

$$+ U \sum_{l} n_{l\uparrow} n_{l\downarrow} + V \sum_{l} n_l n_{l+1}$$

Coulombic Repulsions

$$+ \Delta_{eff} \sum_{l=even} n_l$$

Energy Difference between
Homo (TTF) and Lumo(CA)

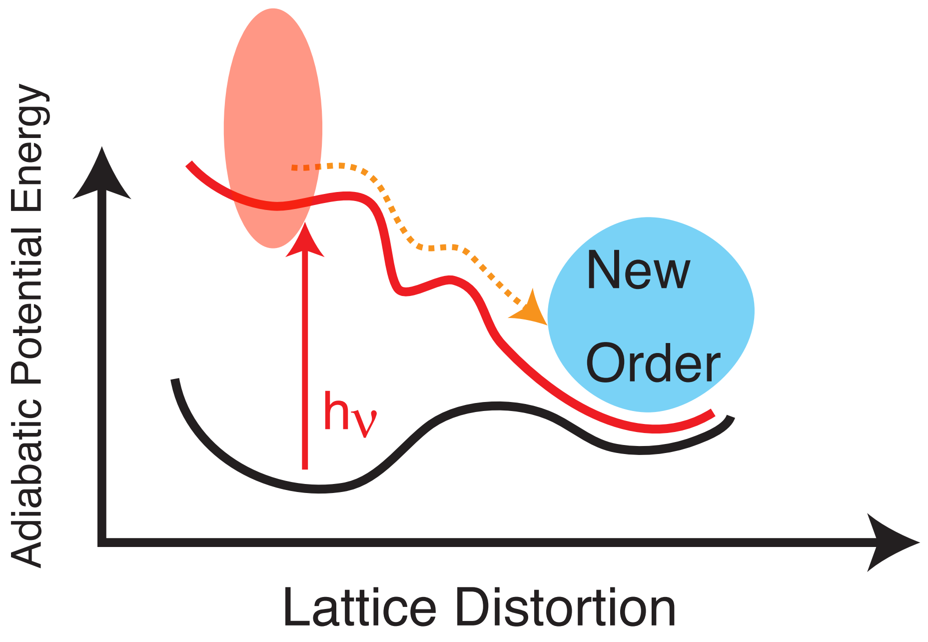


Method: Dynamical Density-Matrix Renormalization Group (DDMRG) technique

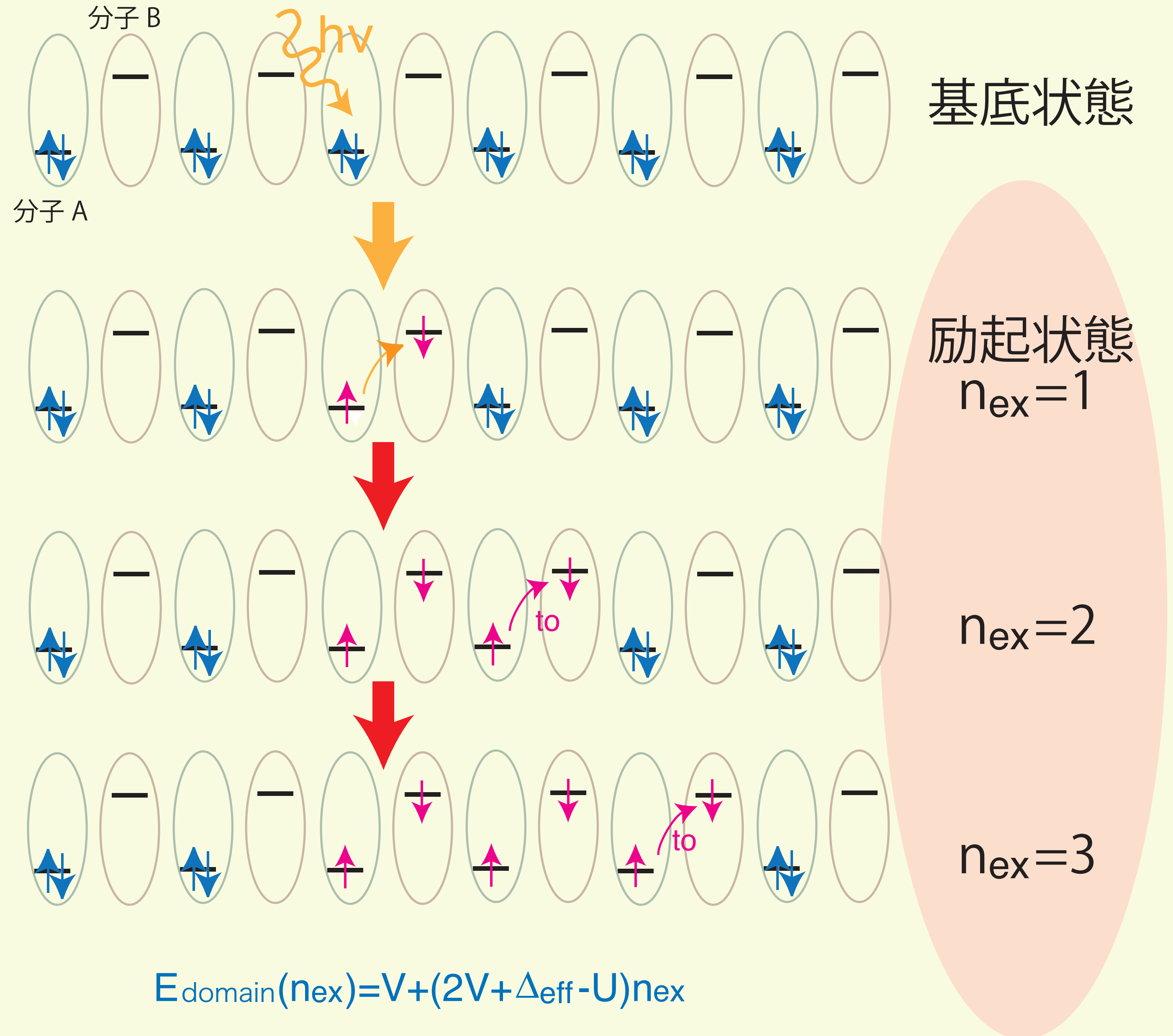
電子転移ダイナミックス（理論提案）

超高速電子ドメイン成長

Ultrafast
Domain Formation?



光励起によるイオン性相 (I 相) ドメインの生成



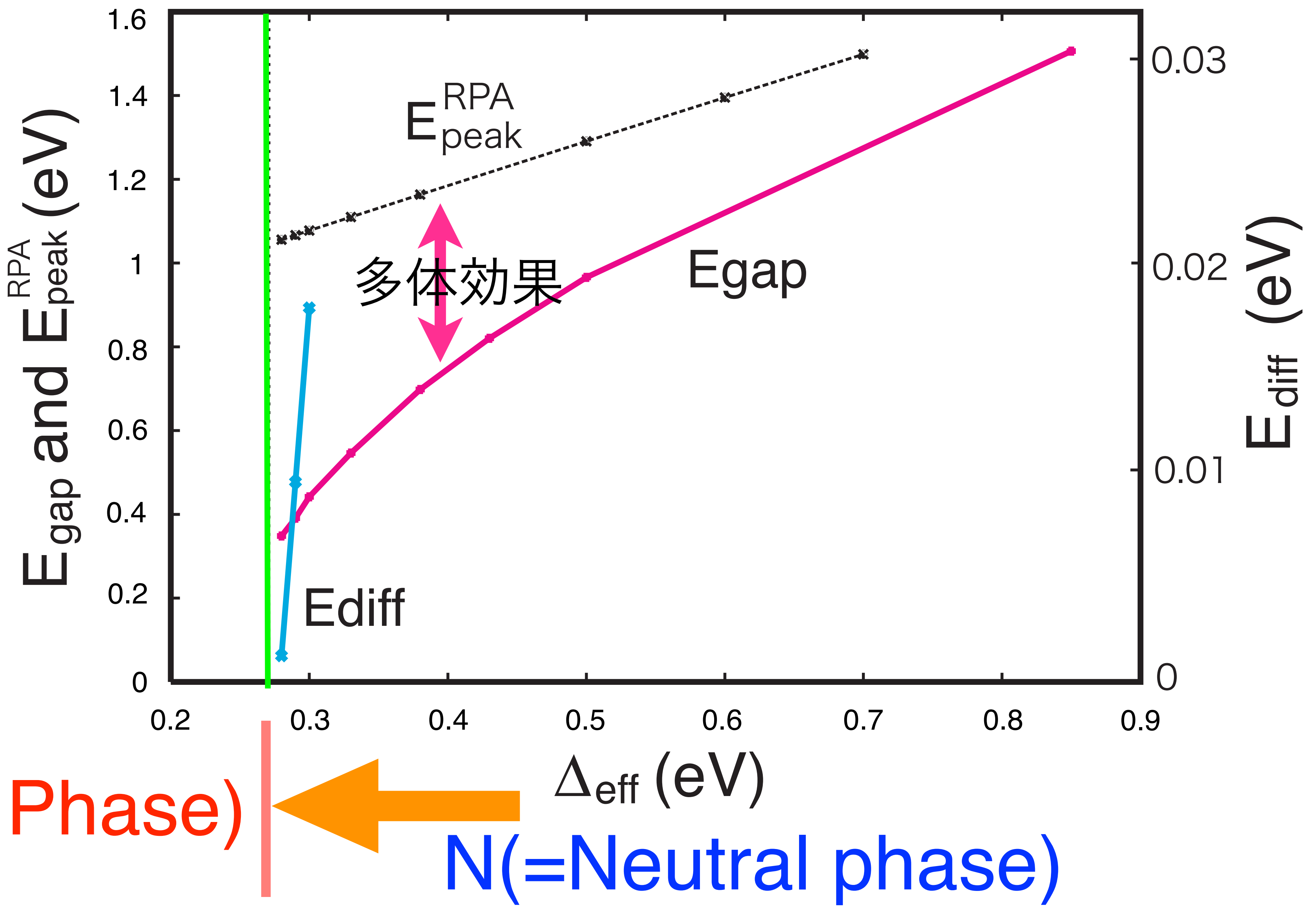
$E_{domain}(n_{ex})=V+(2V+\Delta_{eff}-U)n_{ex}$

$(2V+\Delta_{eff}-U) <<$ to @Phase boundary

相境界で縮退

Phase Diagram of N-I system

U=2.408 eV, V=1.070 eV, and to=0.17 eV



I(Ionic Phase)

N(=Neutral phase)

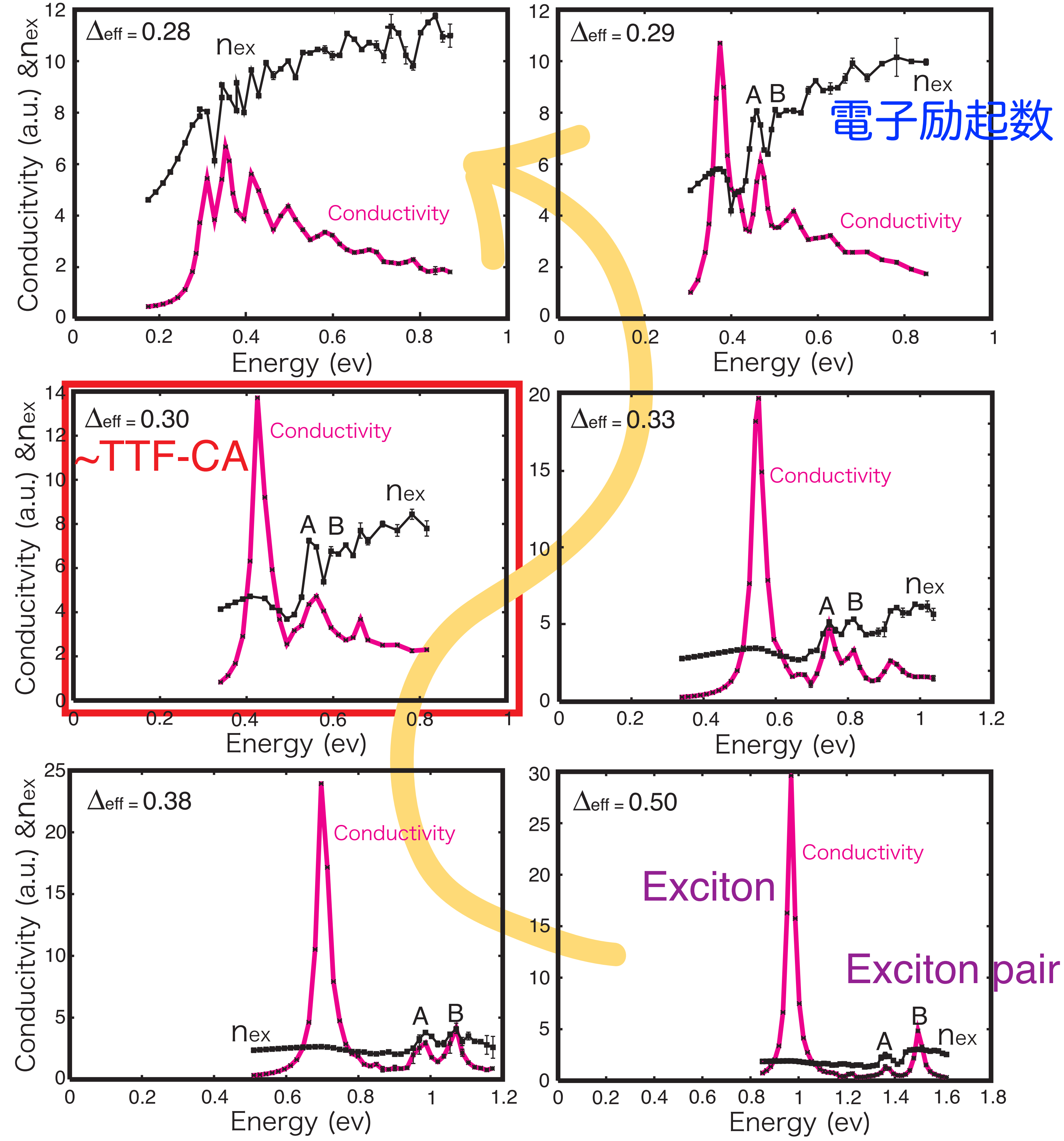
Approaching to phase boundary from the N side

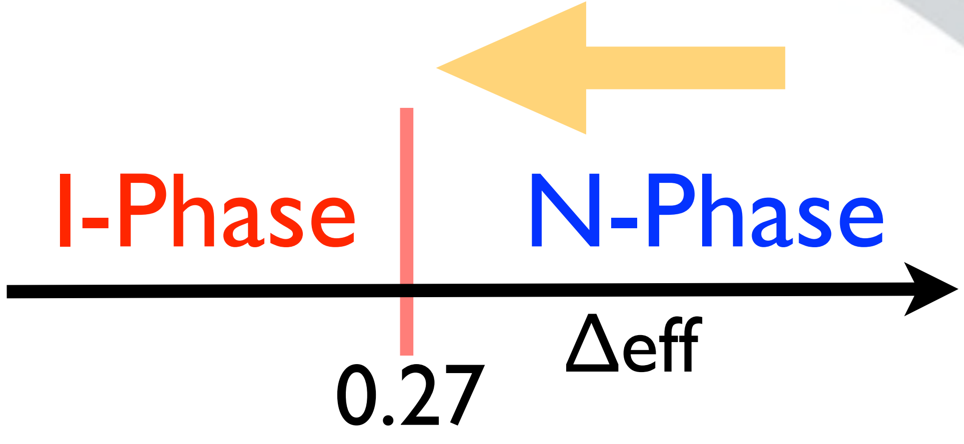
KI, PRL97, 226404 (2006).

Optical Conductivity spectra in a 60-site open system

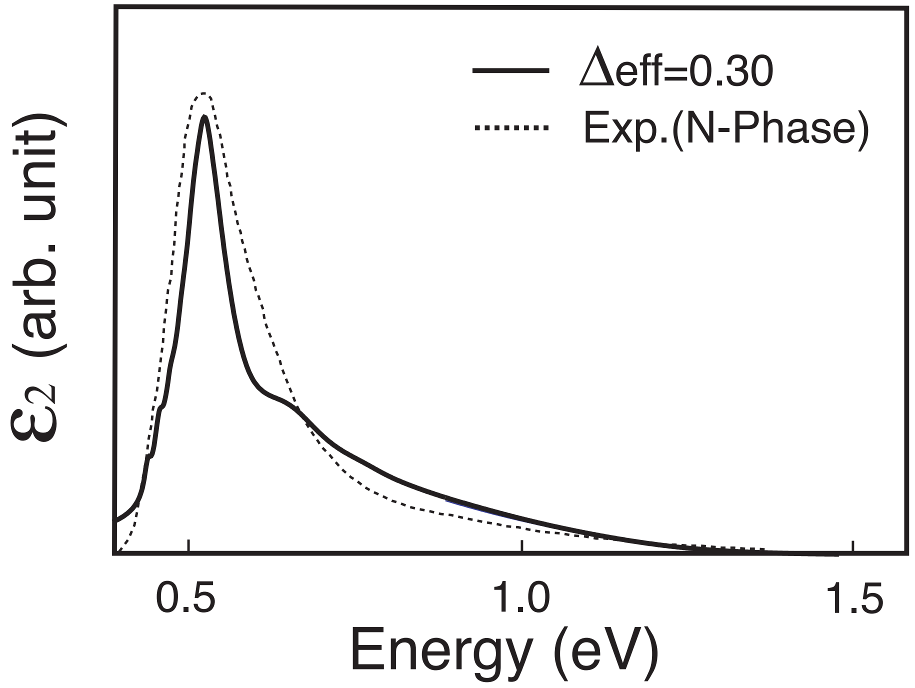
Large Spectral Anomaly

Almost on Phase Boundary





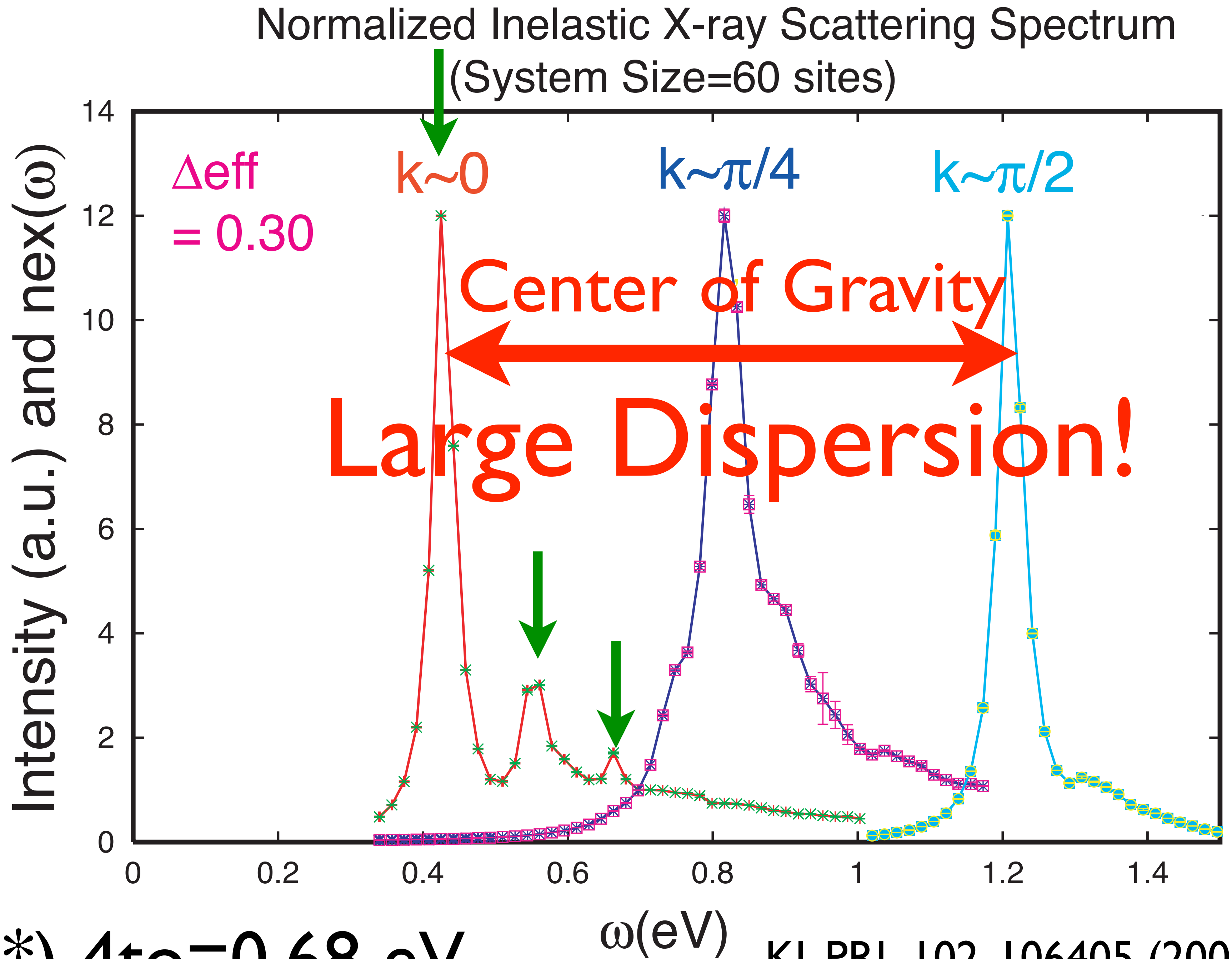
Comparison with Exp.



With further broadening assuming e-ph.

Inelastic X-ray scattering (non-resonant) = density-density correlation func.

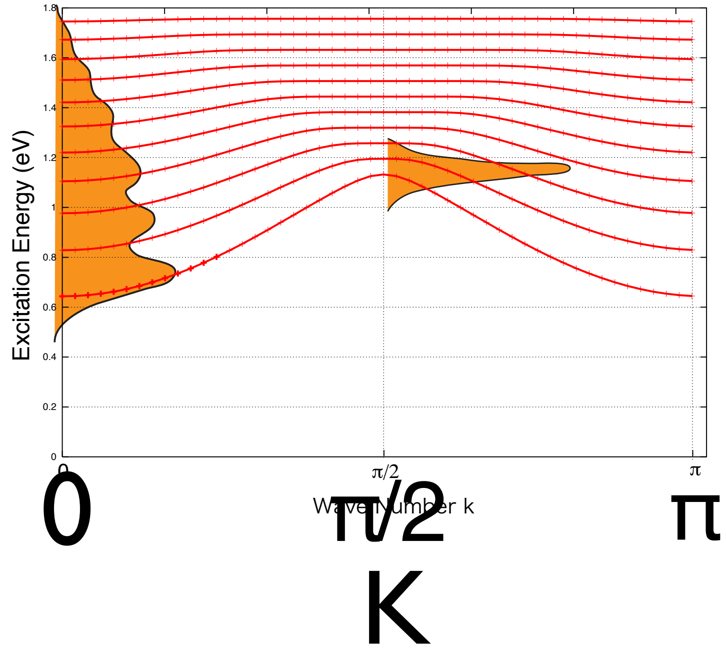
Dispersion of Domain & ON/OFF of "Domain-domain conversion"



Center of Gravity

Large Dispersion!

ドメイン状態
（有効モデルから）



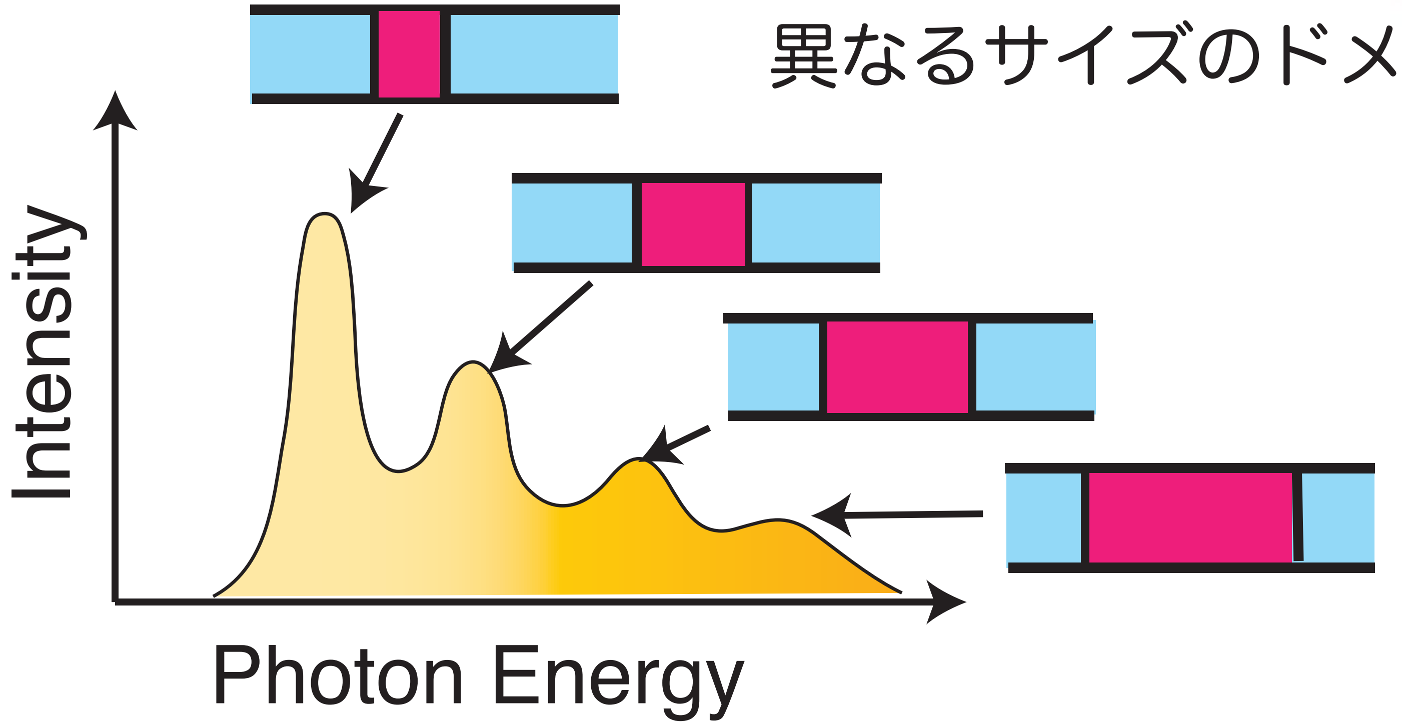
↓ Fine Structures

Quantization of size

*) 4to=0.68 eV

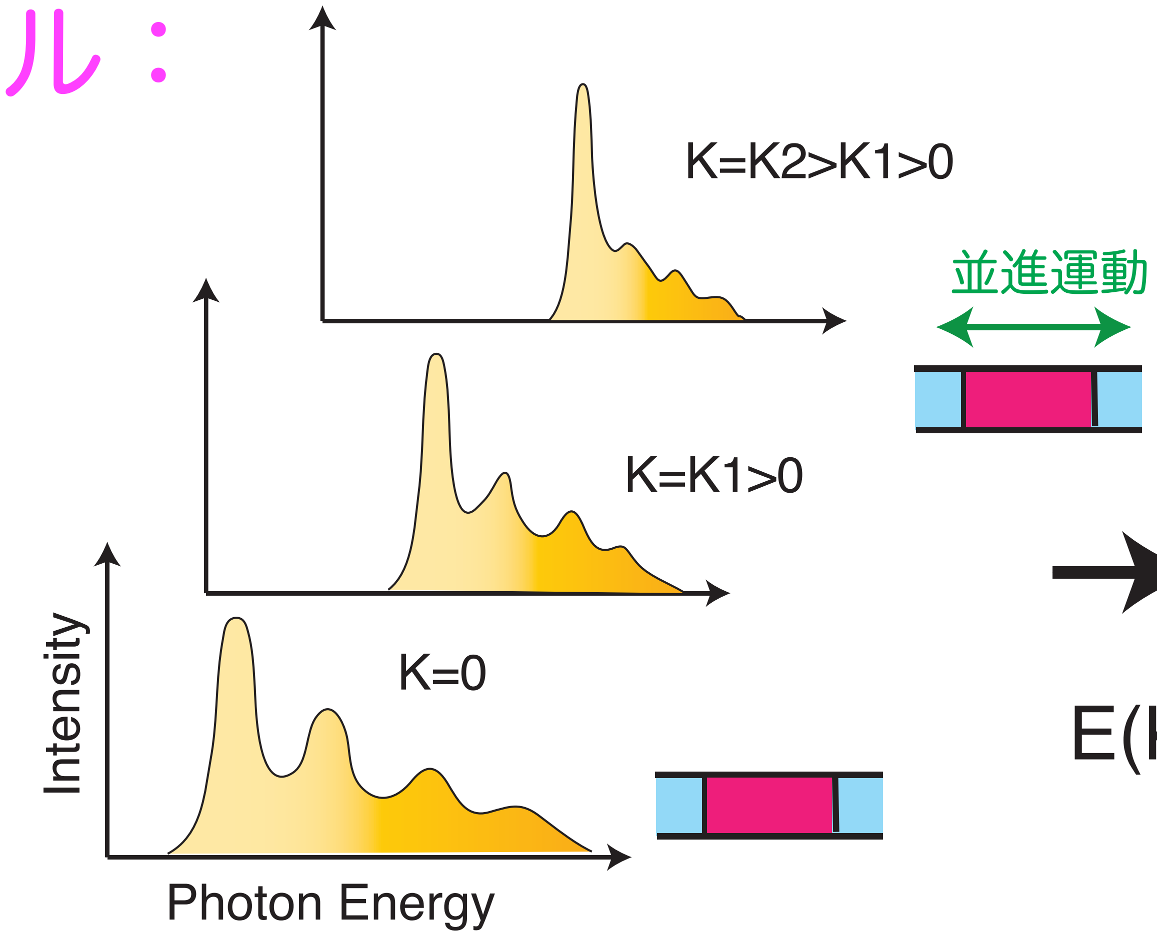
KI, PRL 102, 106405 (2009)

可視光線形スペクトル：
運動量K=0



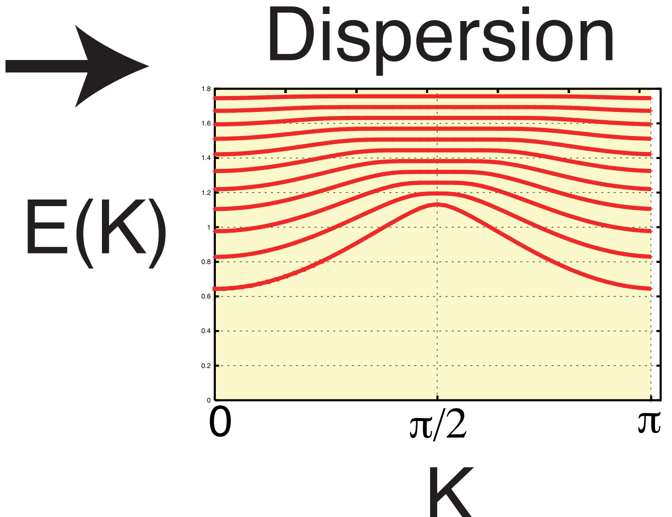
異なるサイズのドメイン状態

放射光スペクトル：
運動量K≠0

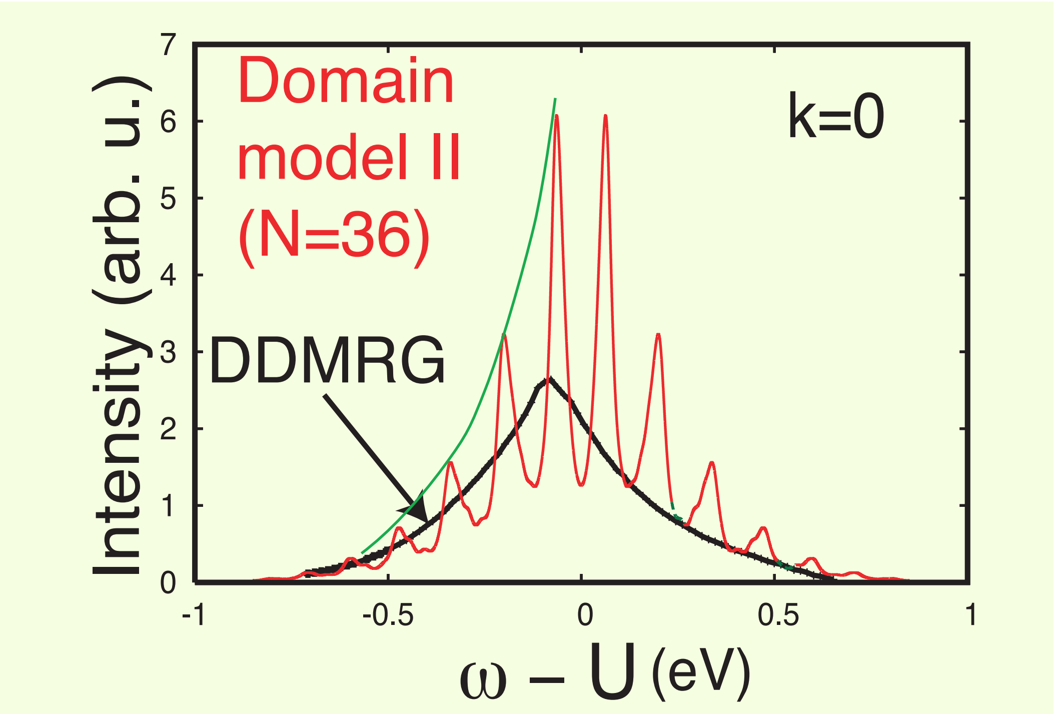


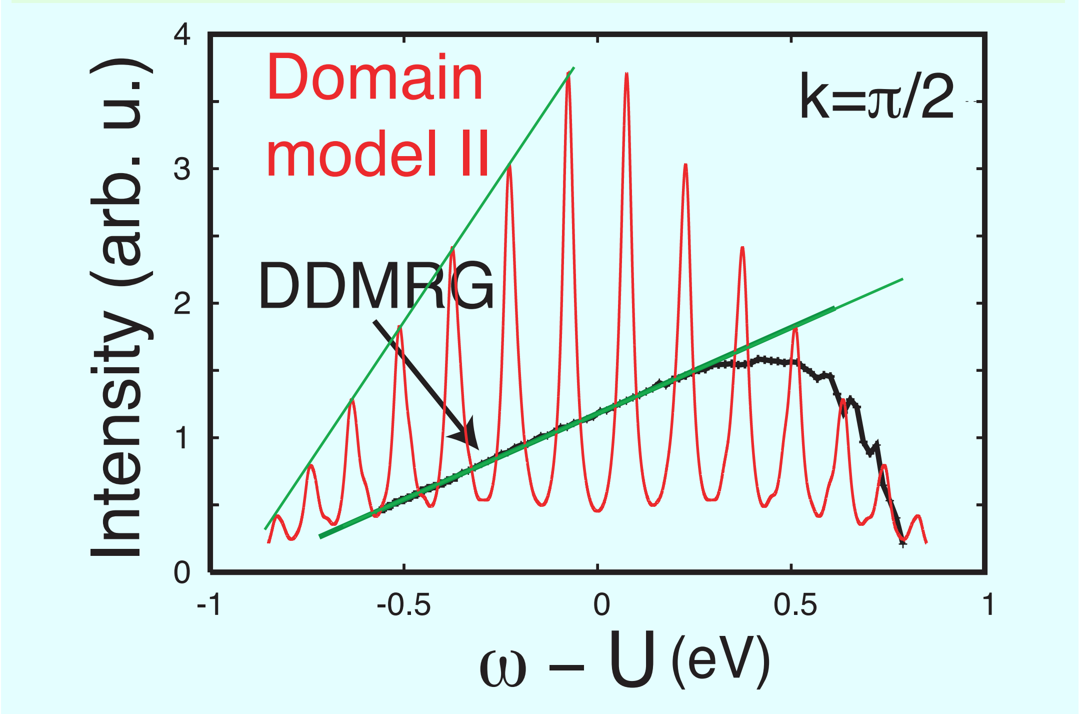
異なるサイズの
ドメイン状態の
埋め込まれ方が
Kに依る！

「粒子」としての
ドメインの分散

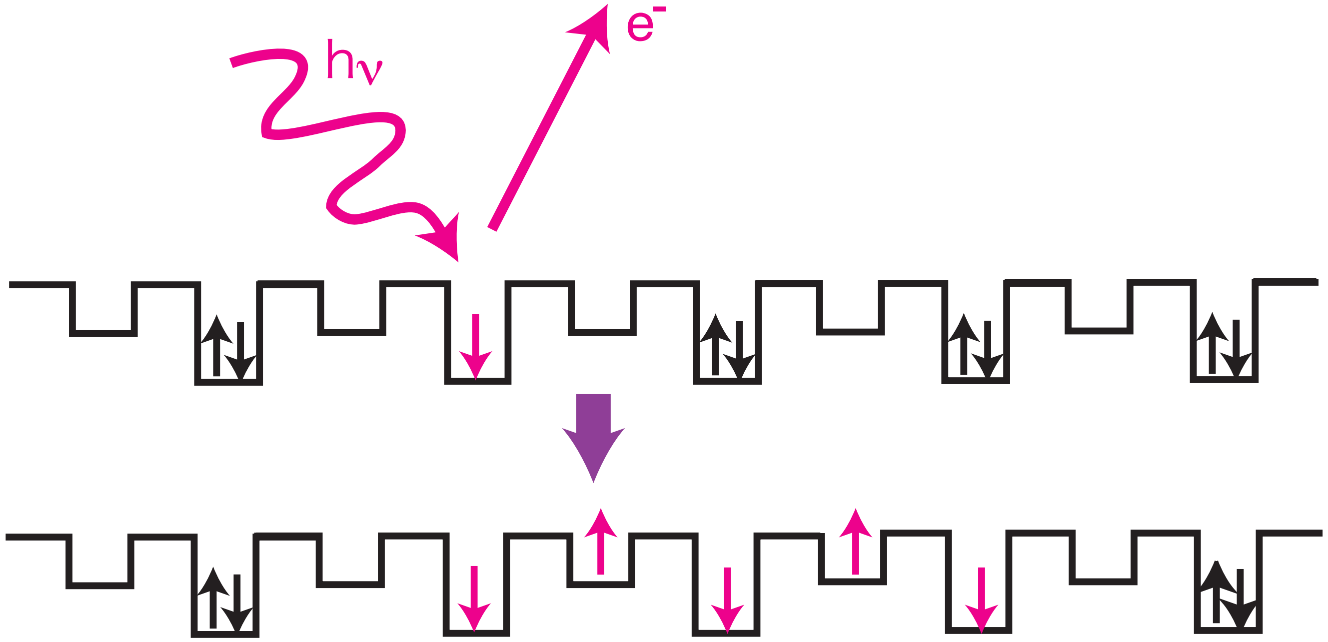


スピン自由度を陽に考える必要のある場合（角度分解光電子分光）





Photomemission



$E_{domain}(n_{ex})=n_{ex}*(2V+\Delta_{eff}-U)$

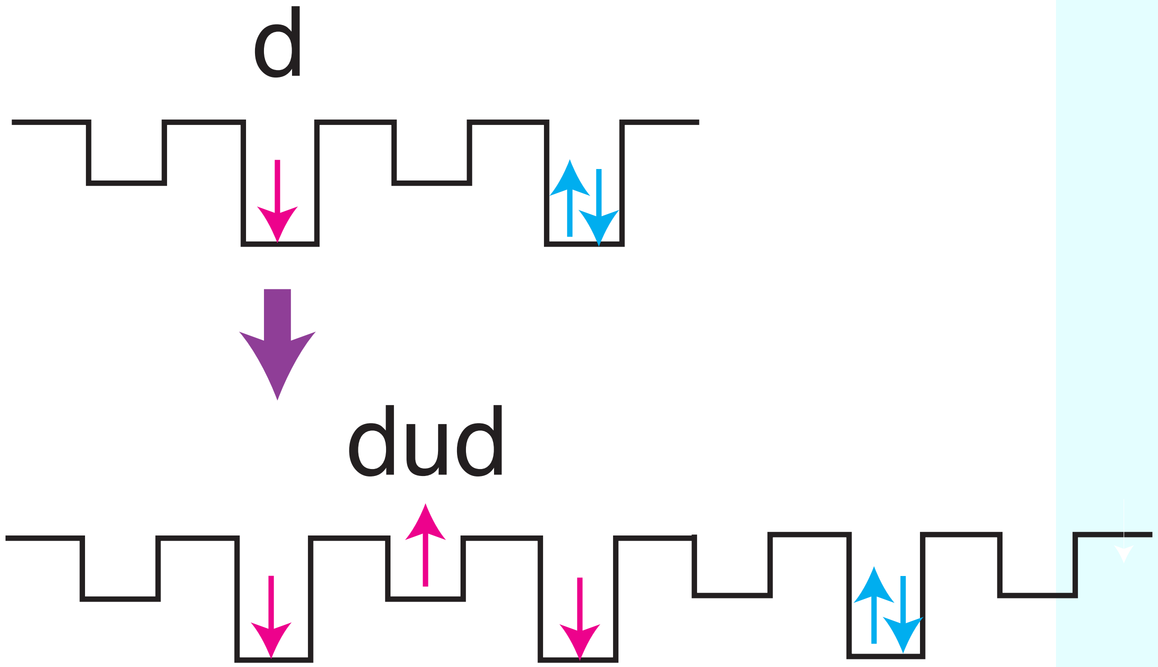
活性スピンの数＝奇数

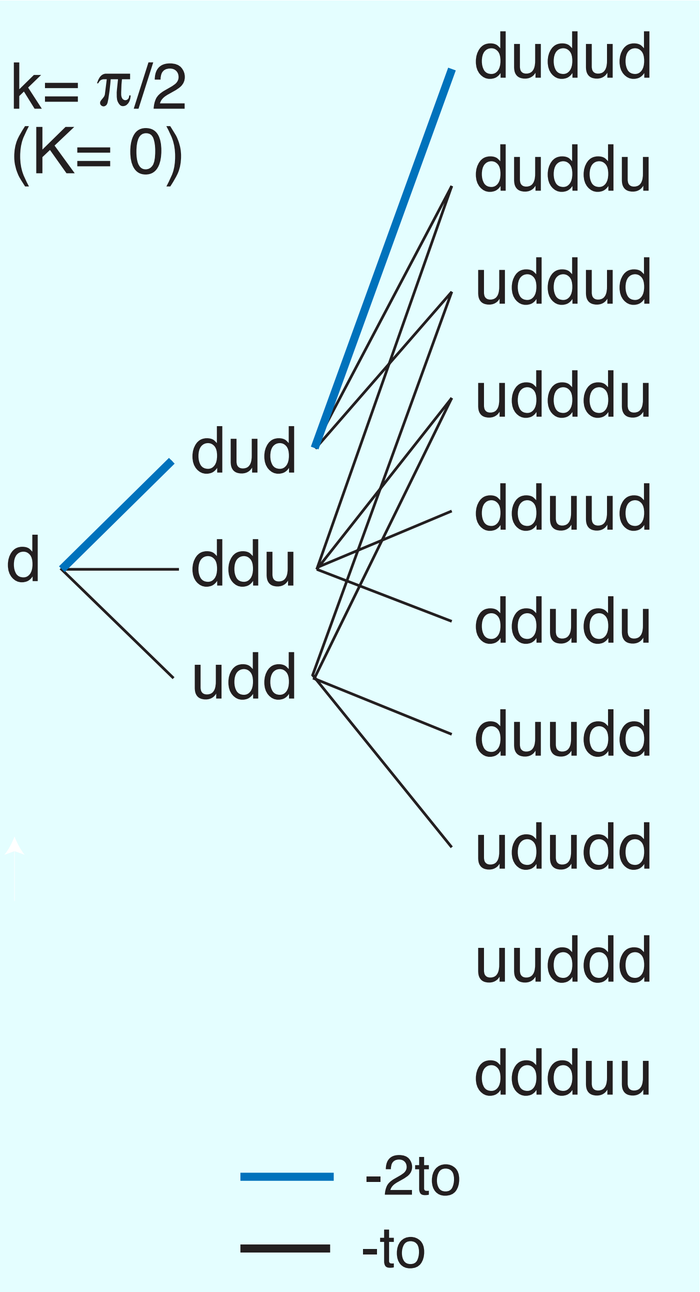
KI, PRL 102, 106405 (2009)

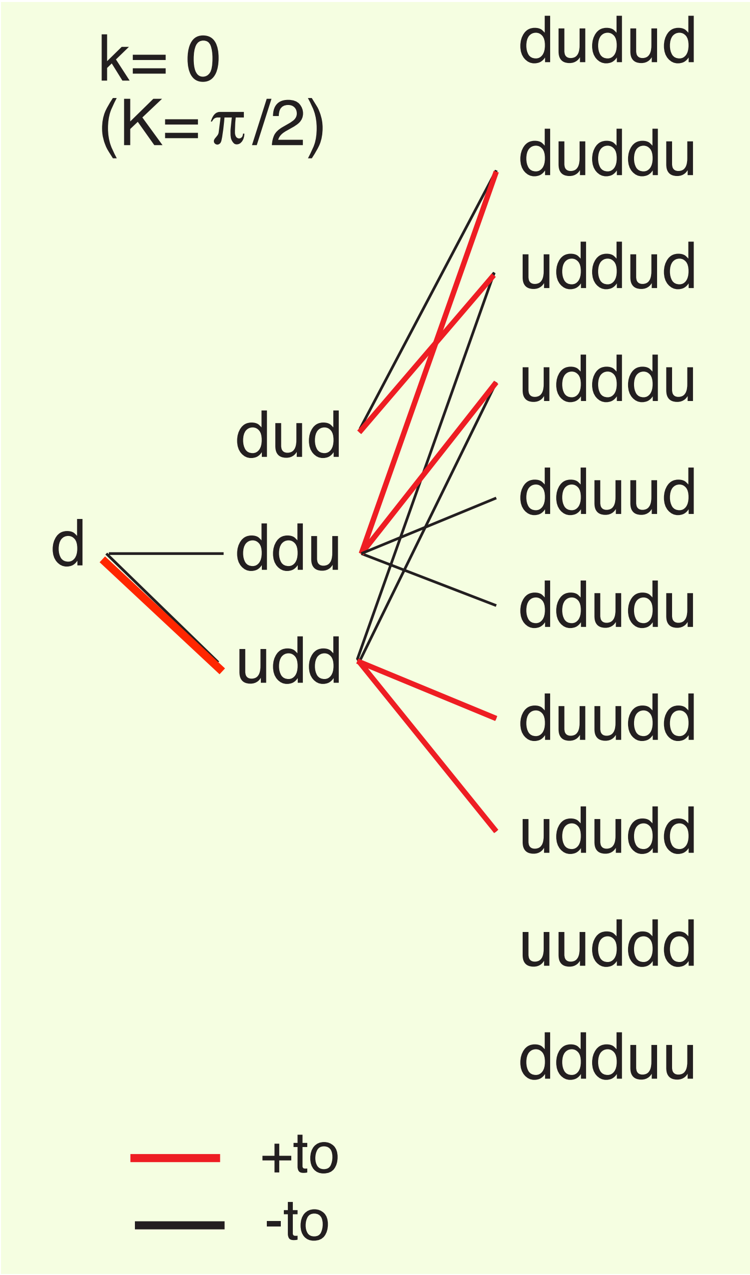
@相境界

Domain Model II — Involvement of spins

``Domain Tree”



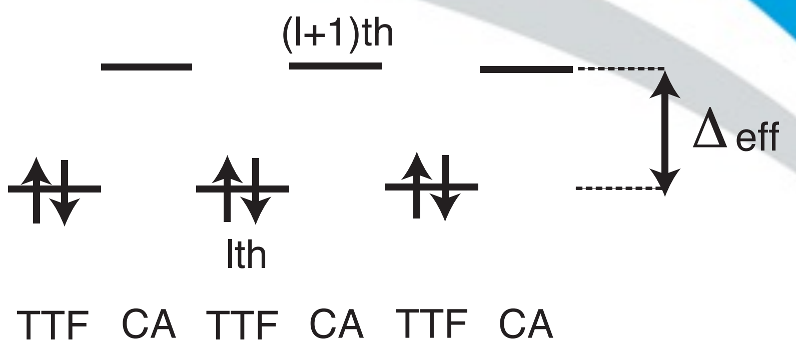




Domain Wall Properties

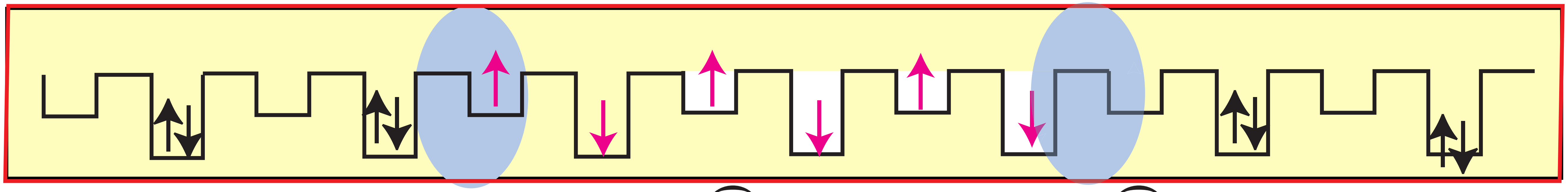
@phase boundary

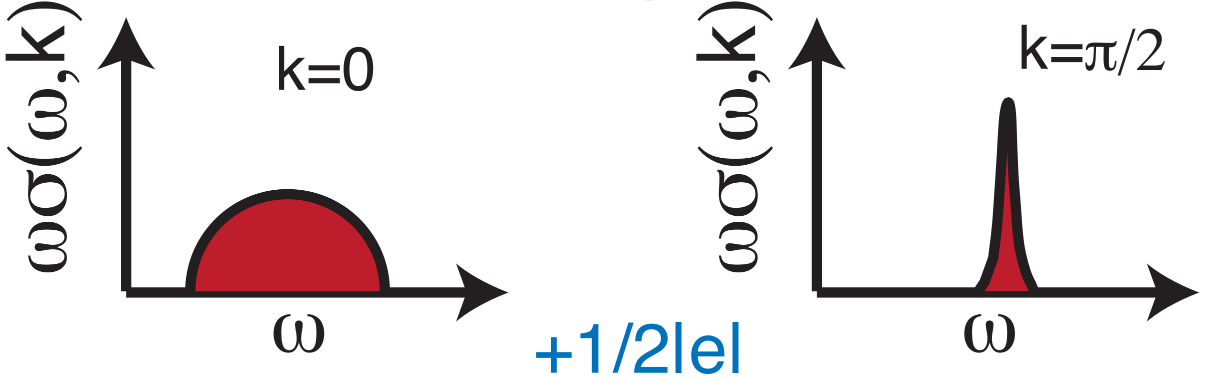
Ground State



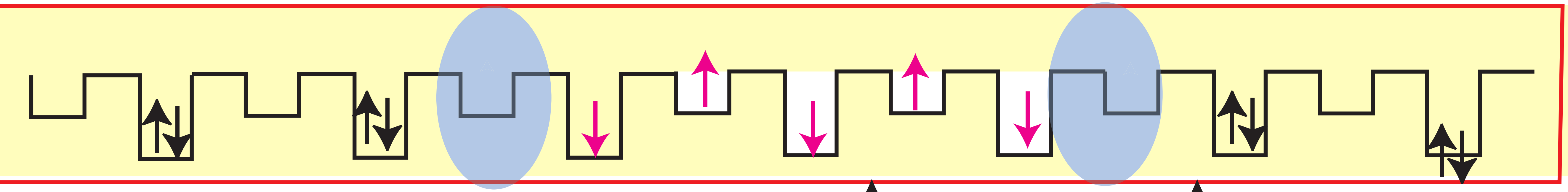
Fractional Charges

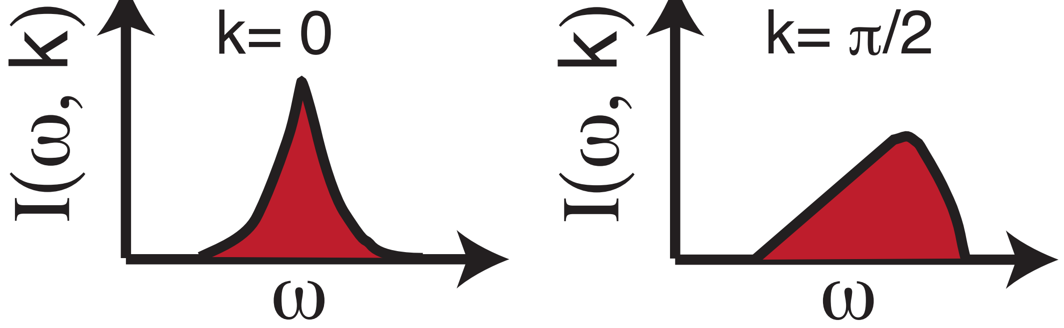
Valence Excitation -1/2lel +1/2lel



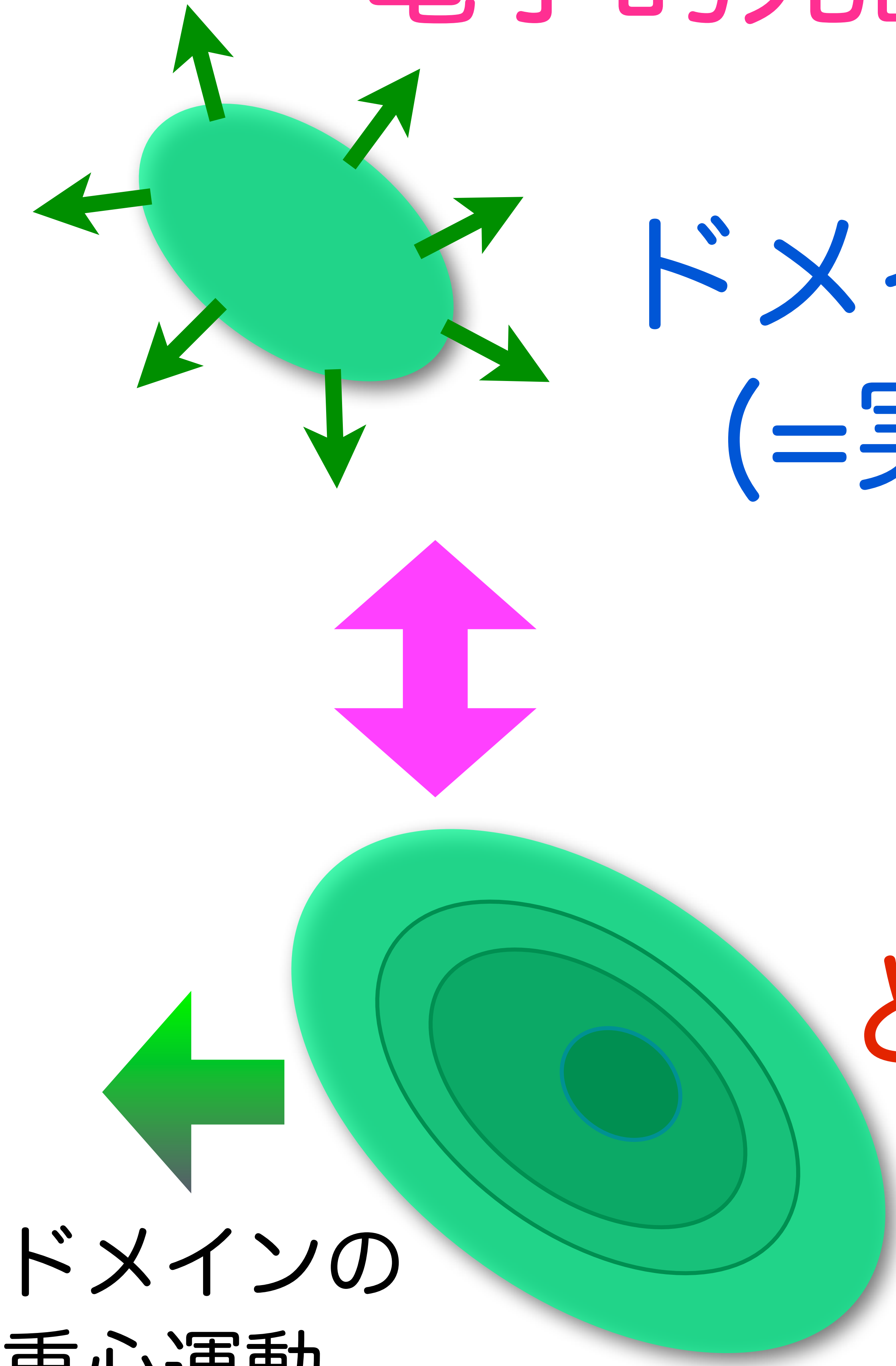


Photomemission +1/2lel +1/2lel





電子的光誘起相転移の考え方



ドメインの成長過程
(=実時間動力学)

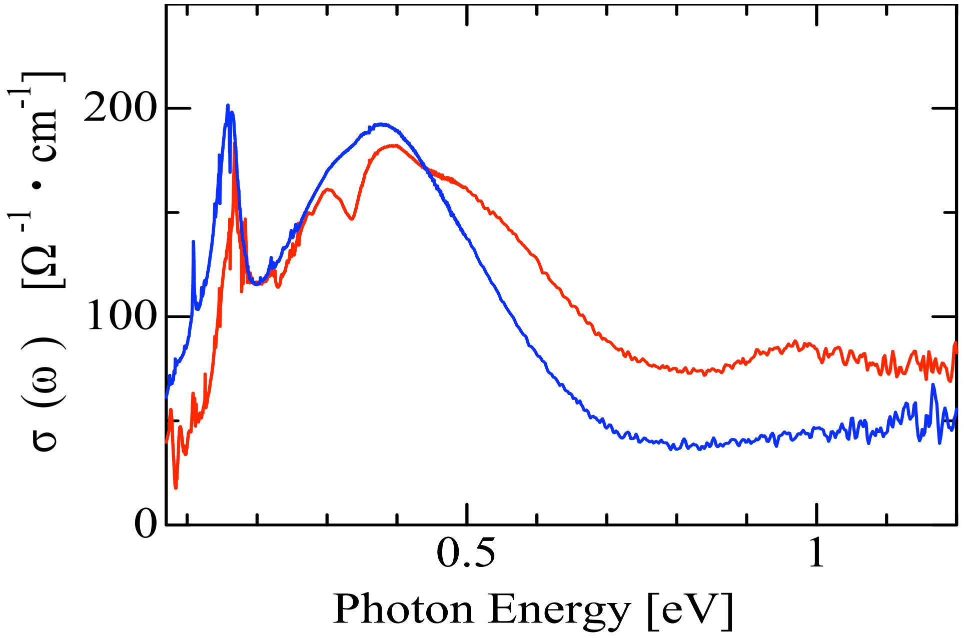
「量子状態(素励起)」
としての記述

ドメインの
重心運動

2-dimensional systems(1/4-filled system): Organic charge-transfer salts

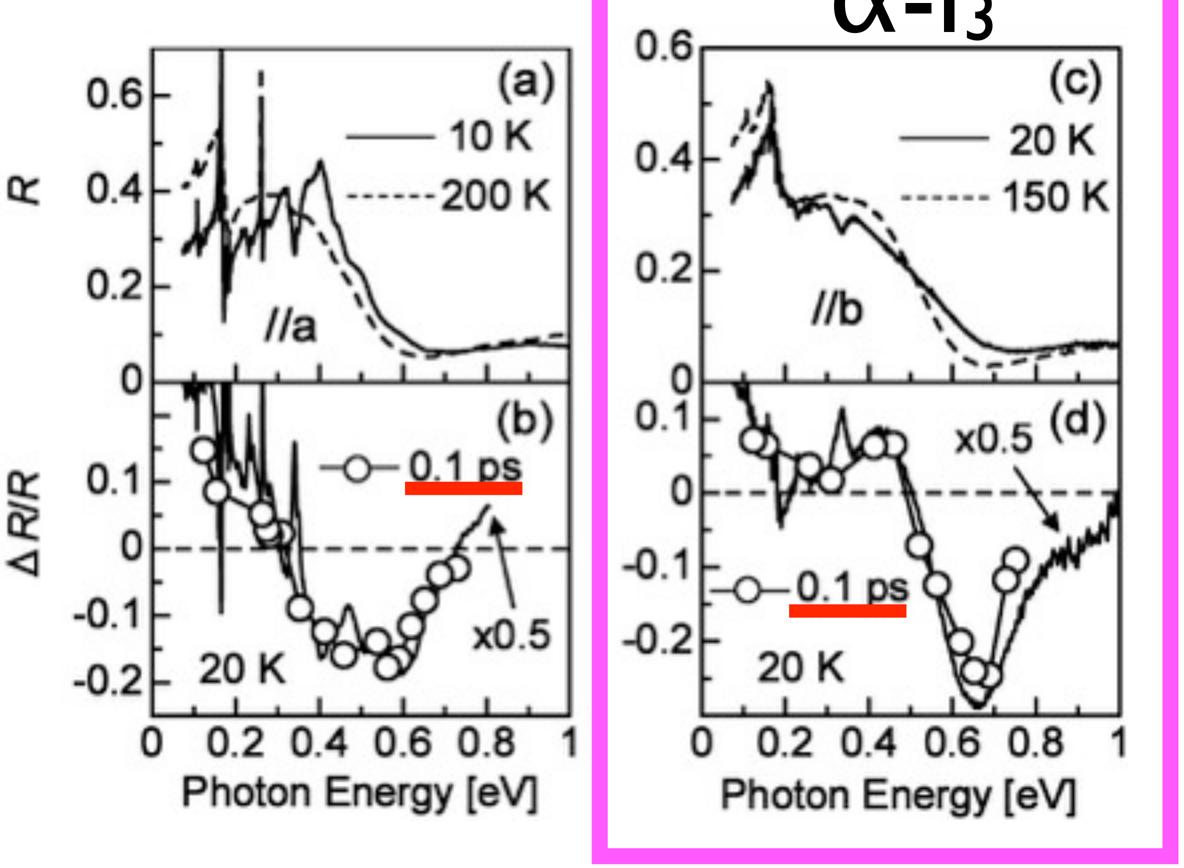
α-(BEDT-TTF $)_2I_3$

gap ~ as small as 0.1 eV.



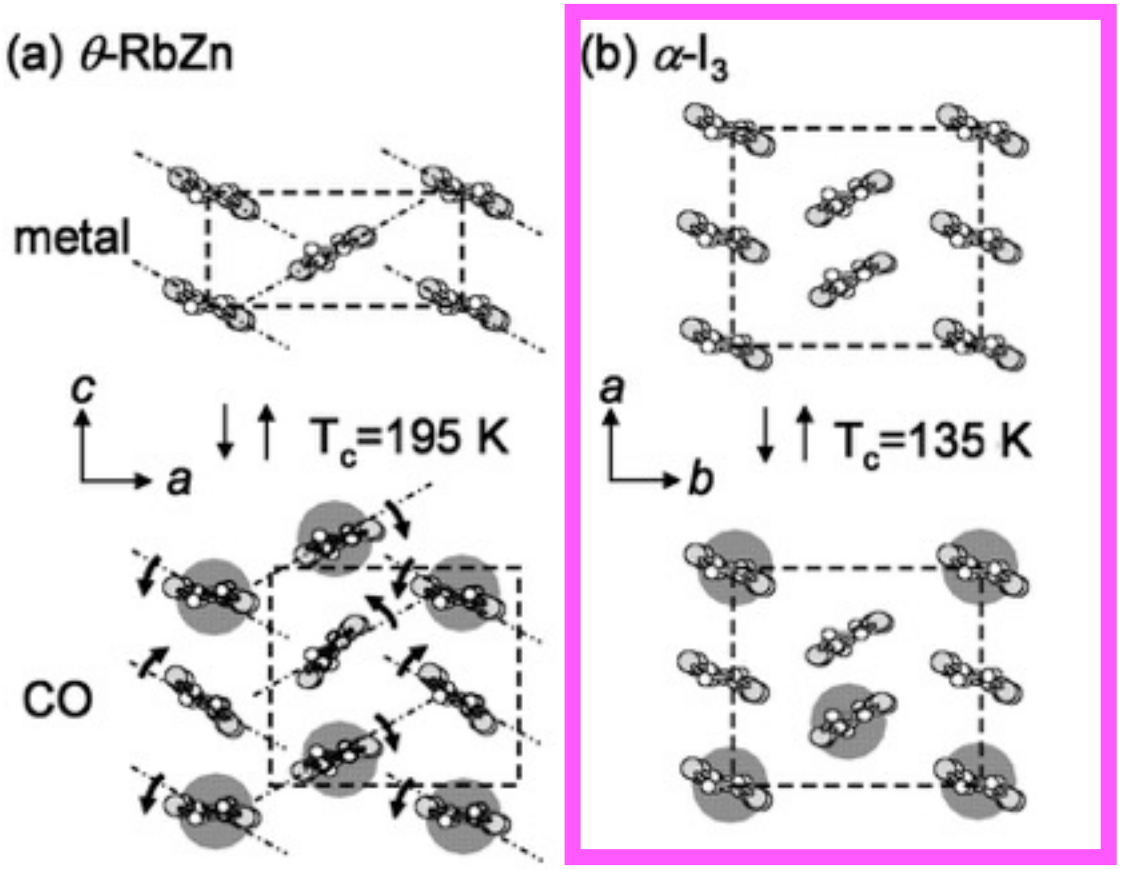
Photoinduced Phase Transition
100~250 molecules/photon!

α-I₃

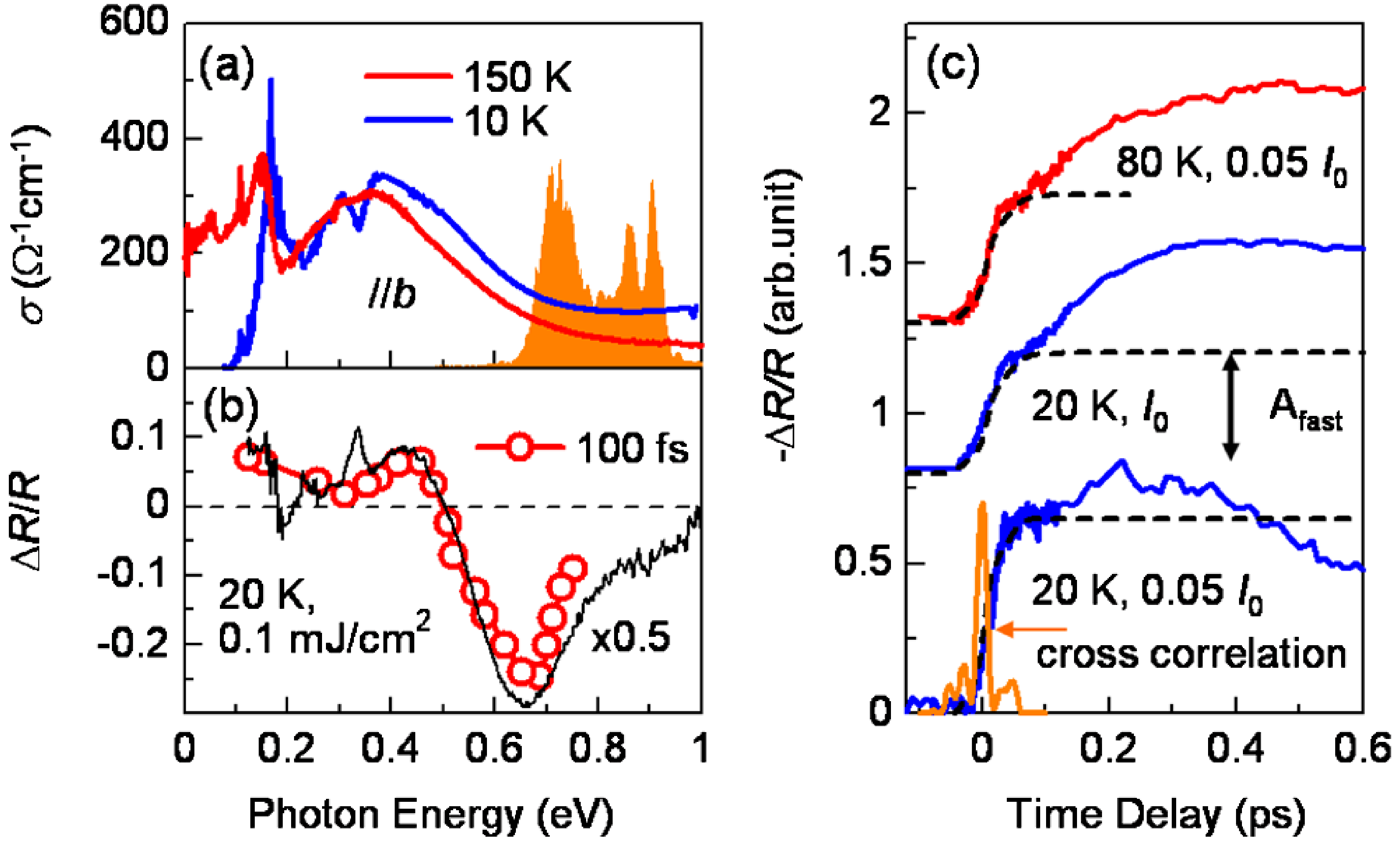


E_{ex}=0.89eV

Charge Ordering



By Iwai et al., PRL 98, 097402 (2007)



I_0= 0.03 mJ/cm^2

超高速！

The time scale of the initial rise in the ΔR/R signal (< 20 fs) is as fast as that of the correlated electron motion, as discussed later.

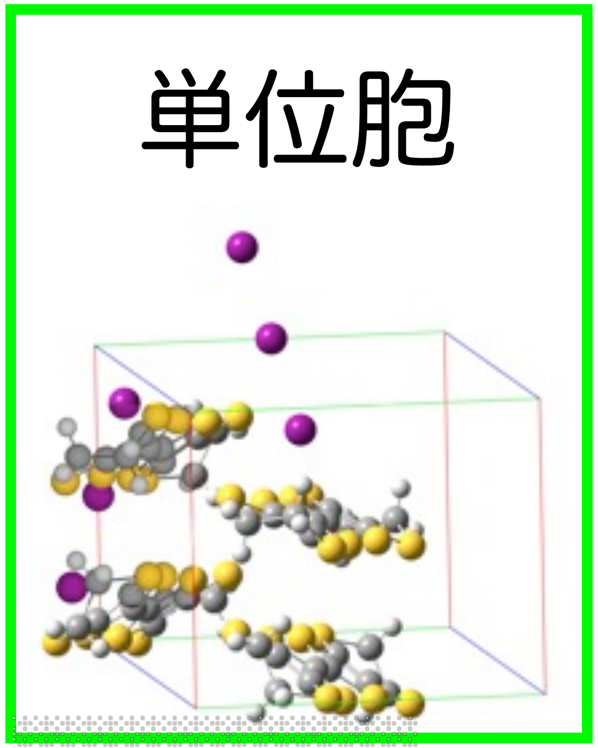
Kawakami et al., PRL 105, 246402 (2010)

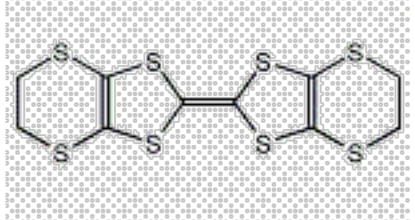
線形性！

The magnitude of ΔR/R observed at 0.1–0.8 eV for t_d = 0.1 ps increases linearly with I_{ex} up to 0.1 mJ cm^{-2}.

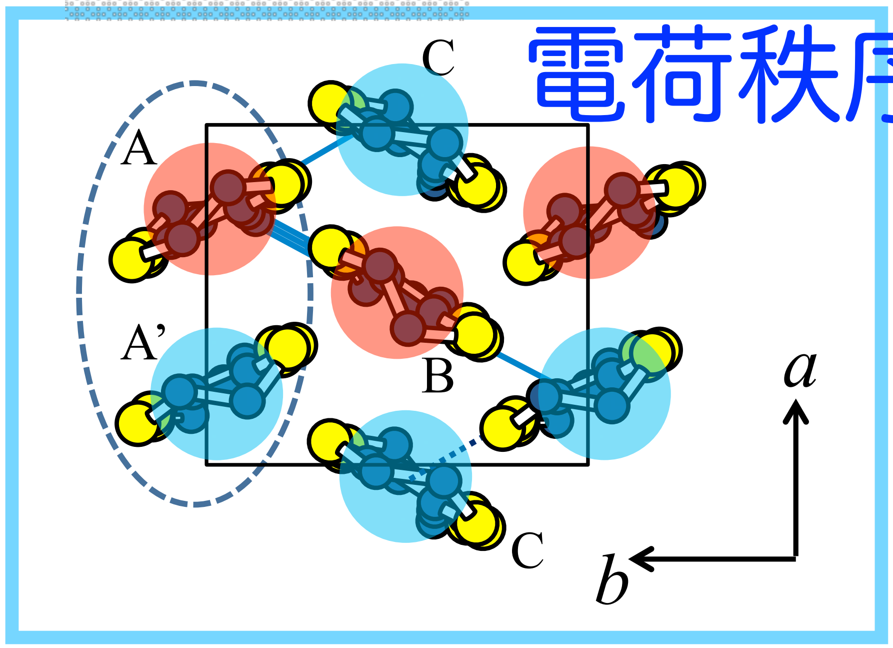
Iwai et al., PRL 98, 097402 (2007)

α-(BEDT-TTF)$_2$I$_3$

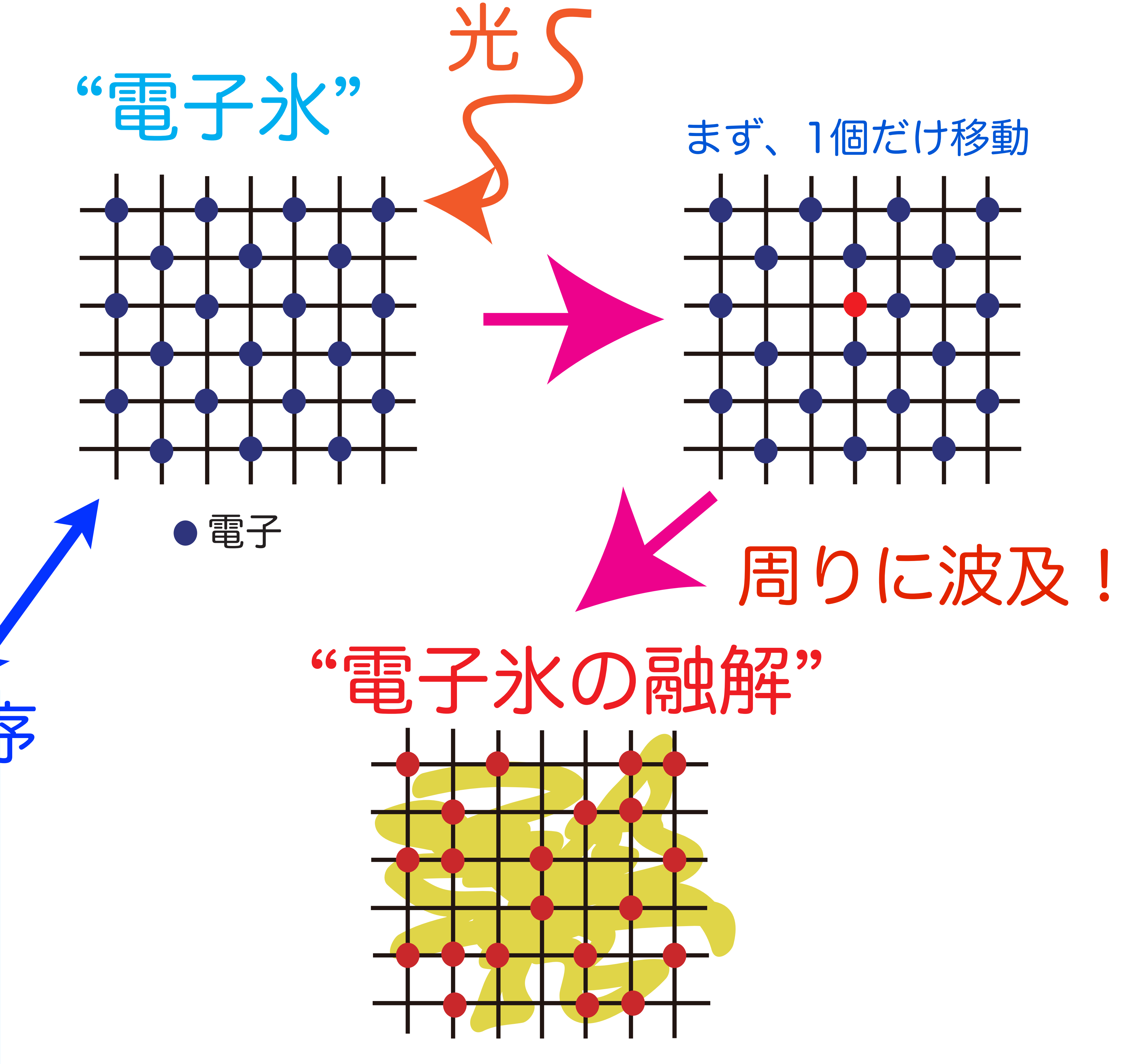




BEDT-TTF



電荷秩序



“電子氷”

まず、1個だけ移動

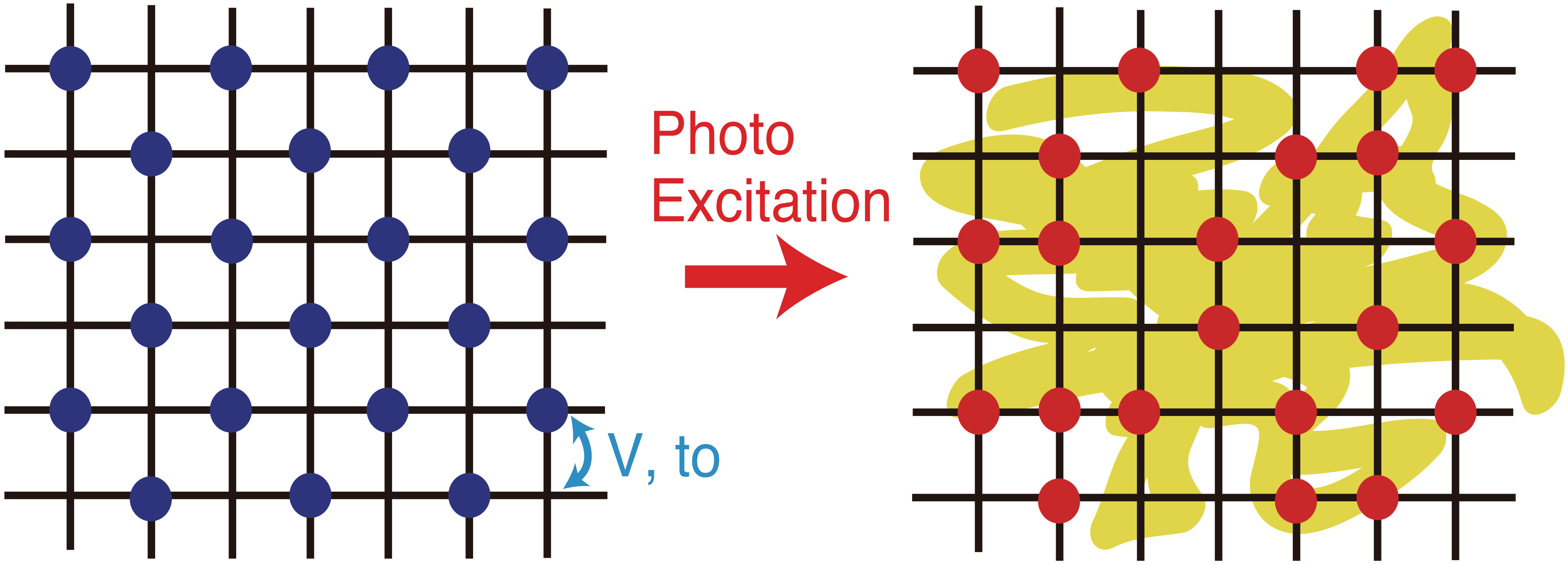
● 電子

周りに波及！

“電子氷の融解”

~20フェムト秒後

２次元系電荷秩序系



Charge Order (CO)

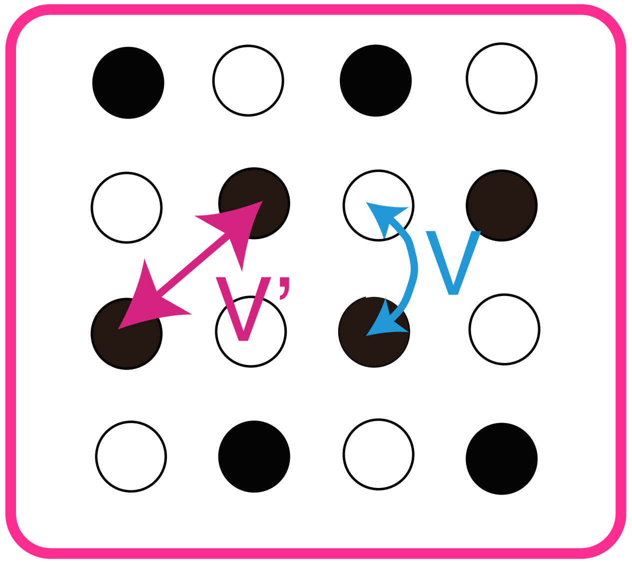
多電子励起？

Spinless Fermion Model

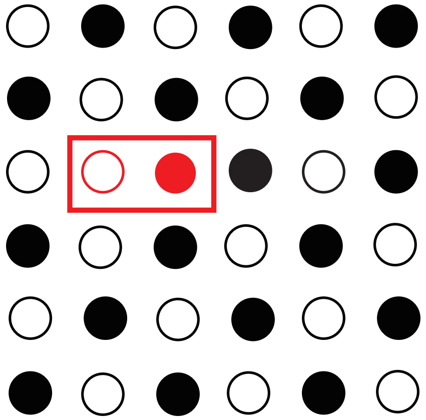
$$H = -t_0 \sum_{l} (C^{\dagger}_{l+1} C_l + h.c.) + \sum_{<l,l'>} V(l-l') n_l n_{l'}$$

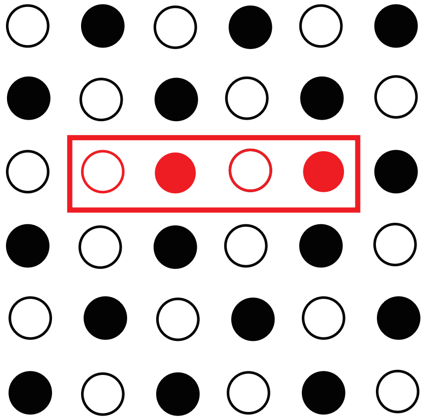
Ref. H. Hashimoto et al. JPSJ 83, 123703 (2014).

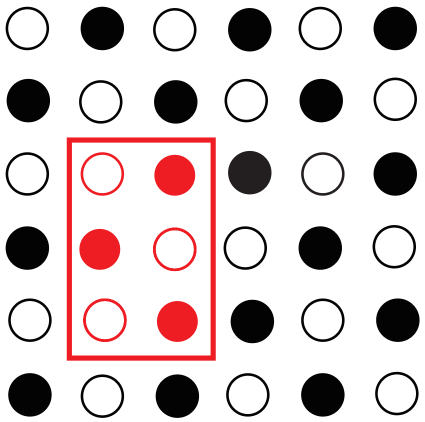
フラストレーション（V'）の効果



例えば、


1exciton


2excitons

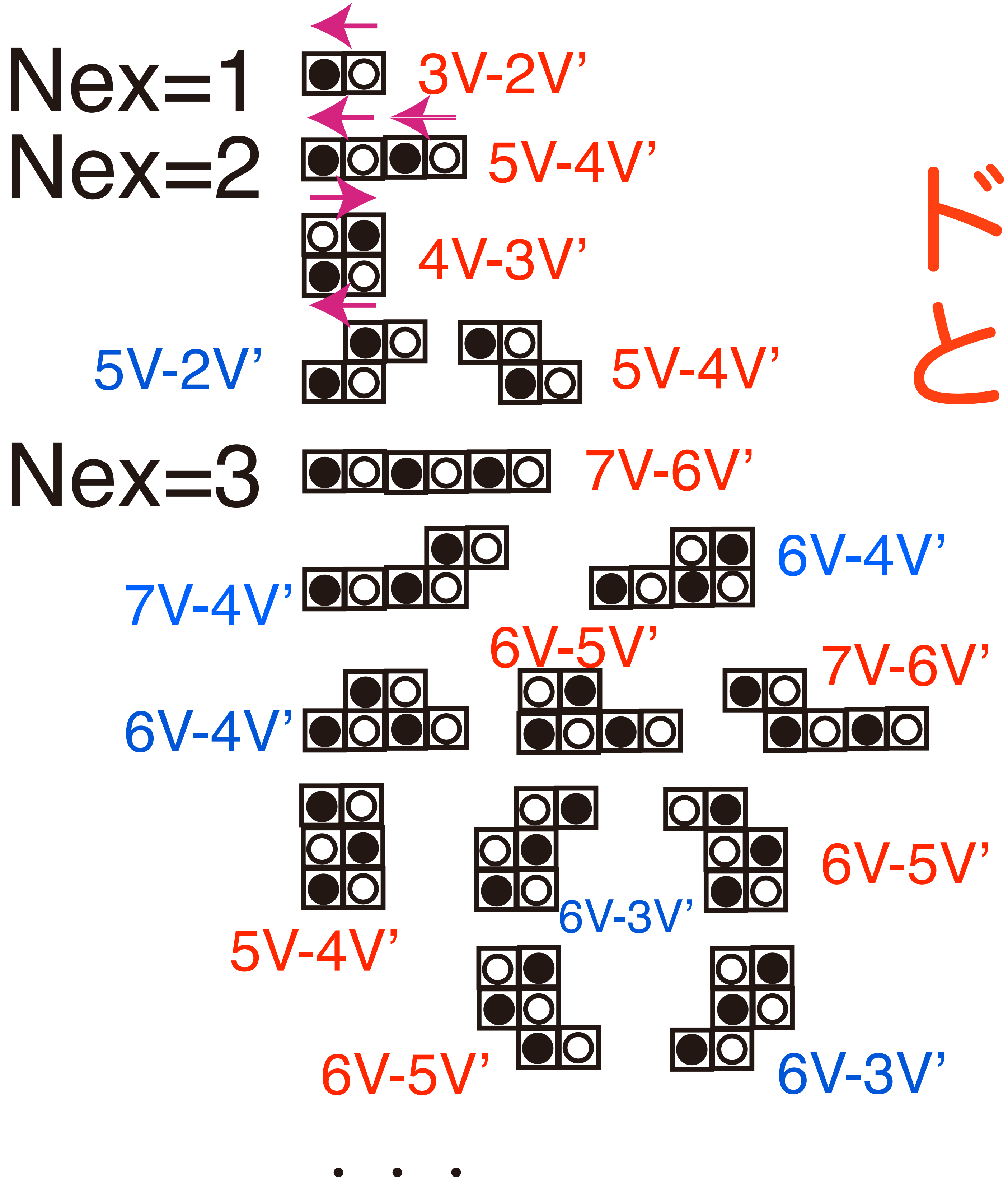

3excitons

Eex	3V-3V'	5V-4V'	5V-4V'
@V=V'	V	V	V

（Rem: V=V'は相境界）

◎相境界に近づくにつれて励起状態同志が縮退

“Domain-animals”

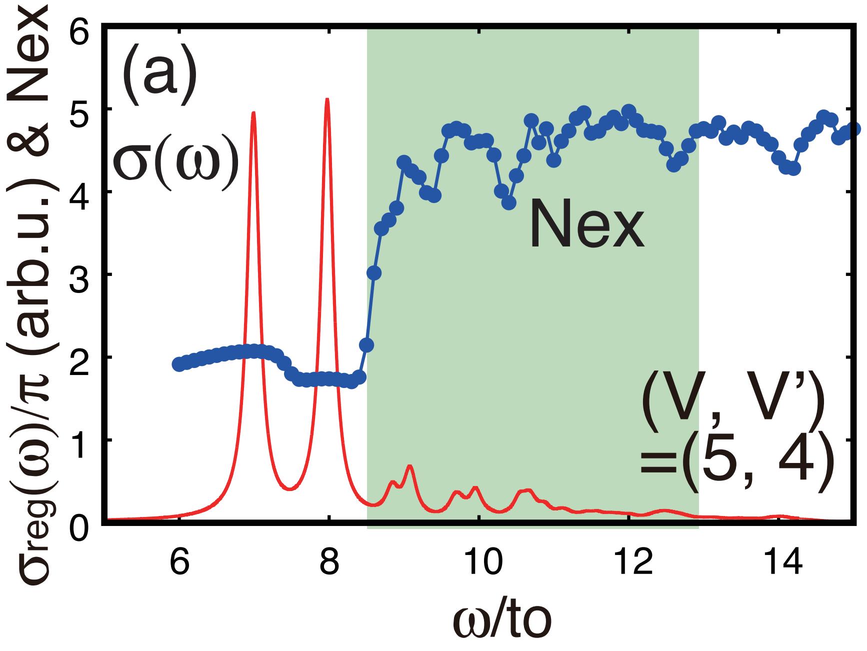


ドメイン形状
という自由度

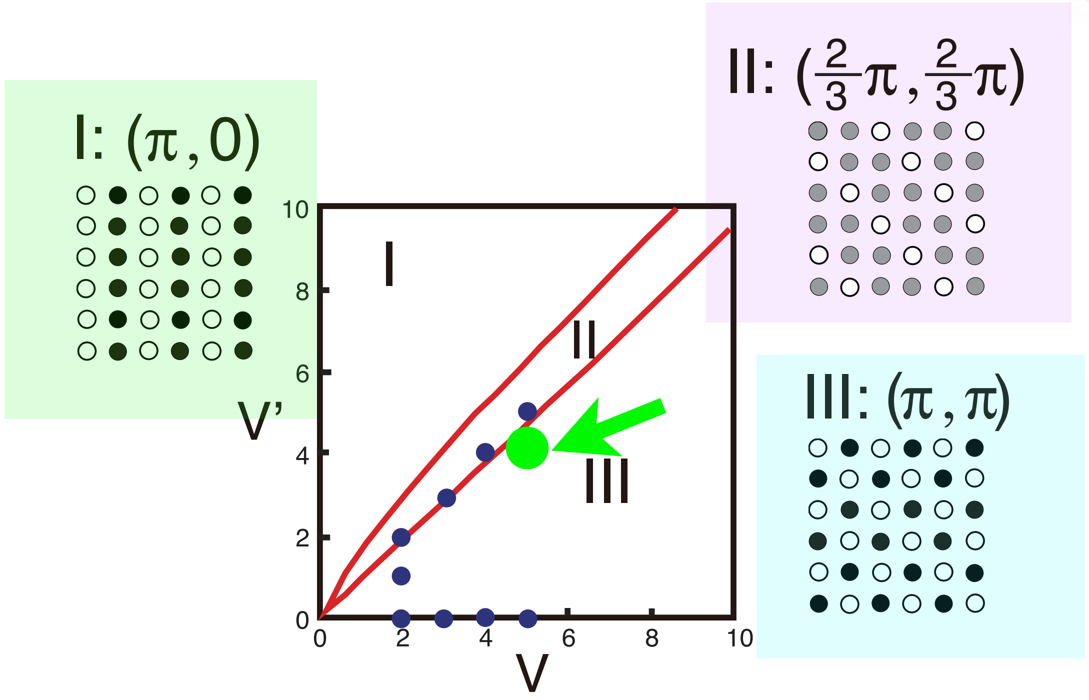
Rem.
相境界でも必ずしも
全ては縮退しない

6×6格子
光学伝導度スペクトル

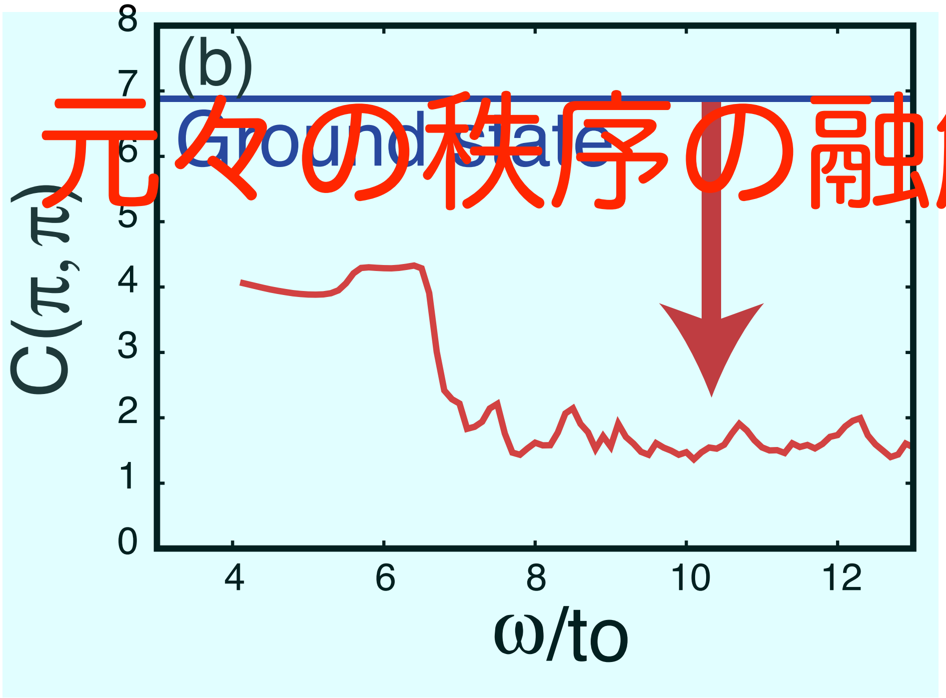
ドメイン状態
(Nex > 4
&有限な振動子強度)



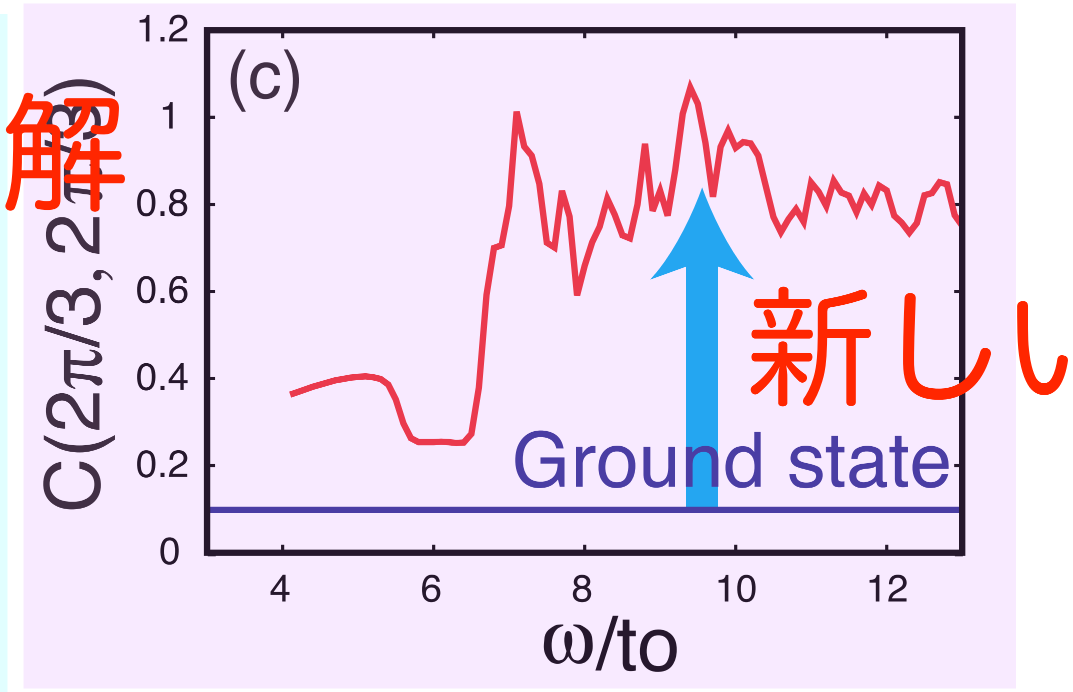
基底状態相図



Ref. C. Hotta et al., JPSJ 75, 123704 (2006). (Size=6×4)



元々の秩序の融解



新しい秩序の形成

KI, PRB 91, 115108 (2015).

まとめ

◎１，２次元系の一部の物質群でドメイン励起が観察できる可能性

ー2次元系の分散は理論でもまだ。

ー今までの理論は非共鳴で行った。
共鳴でも勿論観察可能と考えるが、
より複雑になると思われる。

ー物質は種類を問わない。
レーザー分光とのタイアップが望ましい。