# Exciton Mott transition and quantum pair condensation in electron-hole systems: Dynamical mean-field theory

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Contents:

- Electron-hole systems in quasi-thermal-equilibrium
  - Quantum cooperative phenomena
  - Modern theoretical methods and e-h Hubbard model
- Exciton Mott transition in high-*d*: DMFT and slave-boson MFT
  - Absorption spectra in the half-filled case
  - Role of inter-site interaction : Extended DMFT
- Quantum pair condensation and crossover in high-*d*: SCTMA
- Toward the complete phase diagram
- Absence of exciton Mott transition in 1-*d*: bosonization, RG, and CPT

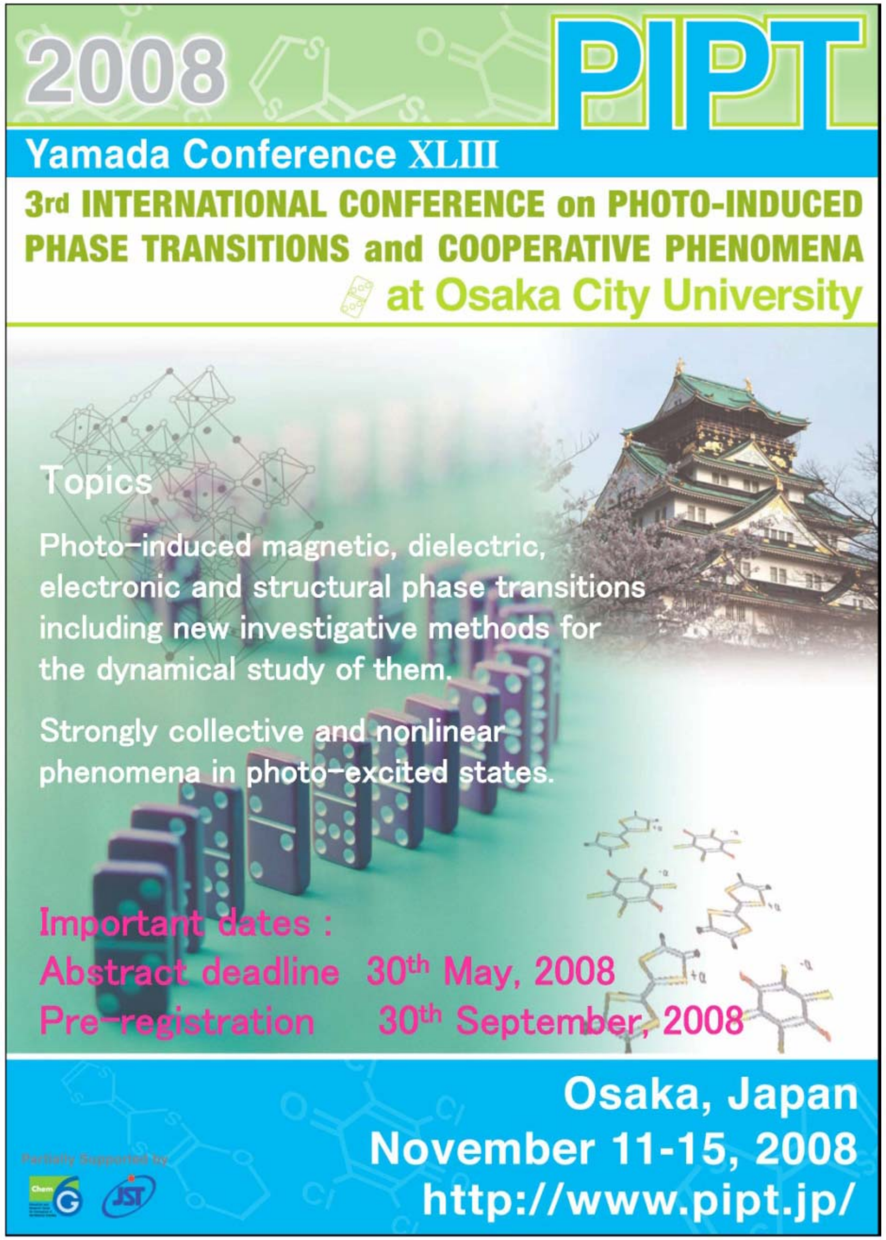
# PHOTOINDUCED PHASE TRANSITIONS (PIPT)

✓Creation and control of macroscopic states of matter by light:

- ➢Electronic phase transitions
- ➢Structural phase transitions

✓Two types of PIPT process:

- ➢"Phase" transitions in photoexcited states
- ➢Phase transitions via photoexcited states



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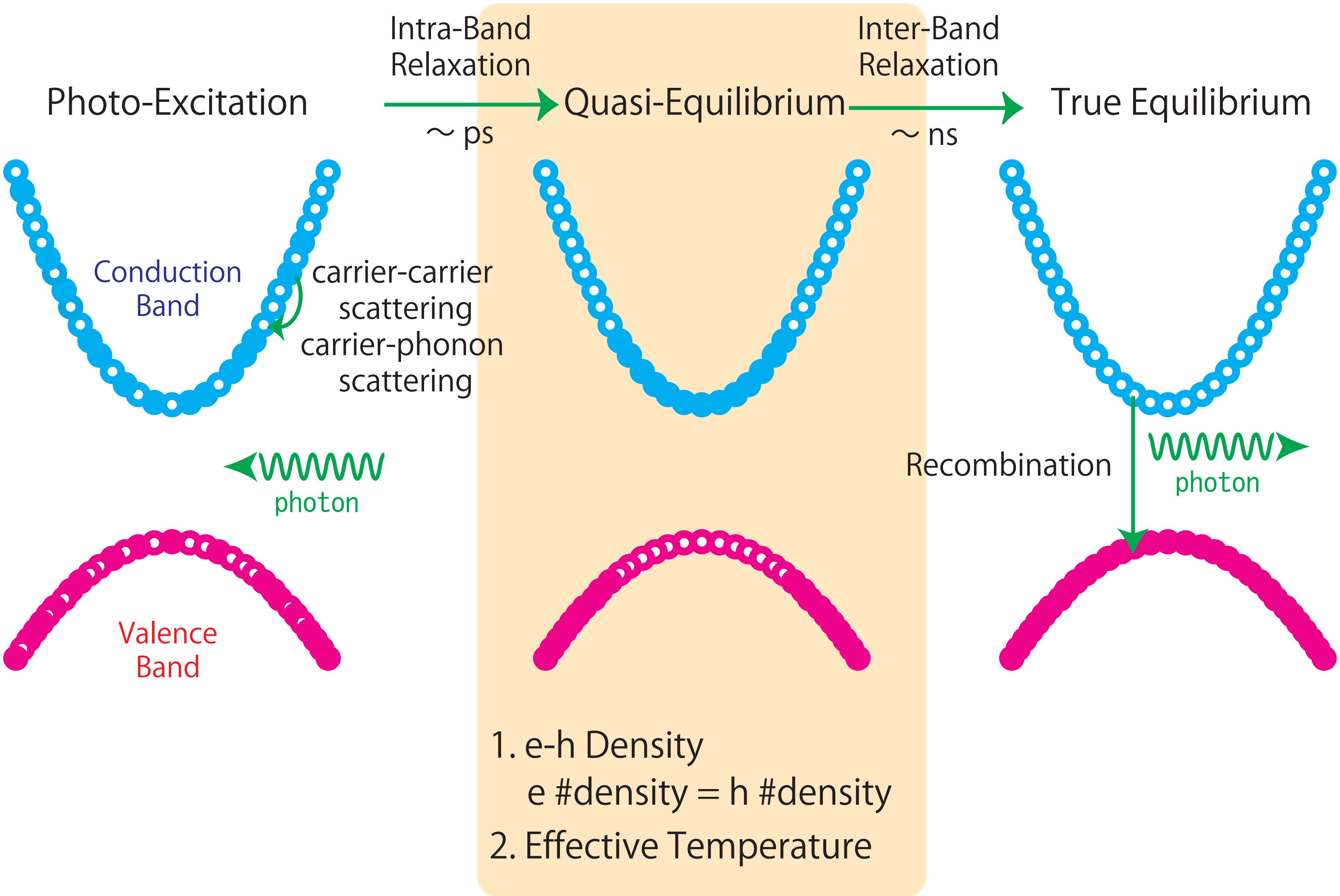
# Electron-hole system as an excited electronic state



# Characteristics of Electron-Hole Systems

2-component quantum many-body system

Opposite charges, different effective masses, mass anisotropy. Fermion→ Pauli blocking

Coexistence of repulsive and attractive Coulomb interactions

The same order of magnitude, Free-electron theory/Fermi liquid theory invalid

"Exciton" and "excitonic molecule" can be formed.

An exciton or a biexciton is (quasi)bosonic.

Tunable particle density by irradiation intensity

Not always # of electrons = # of holes→modulation doped system

Variable strength and range of Coulomb interaction

Static and dynamical screenings, dielectric confinement in low dimensions

Excited states with energy higher than thermal energy at RT

meV (Teraheltz, IR), eV (visible), keV (UV, X-ray): Wide range of light energy

Open system with phonon bath: Intraband energy relaxation

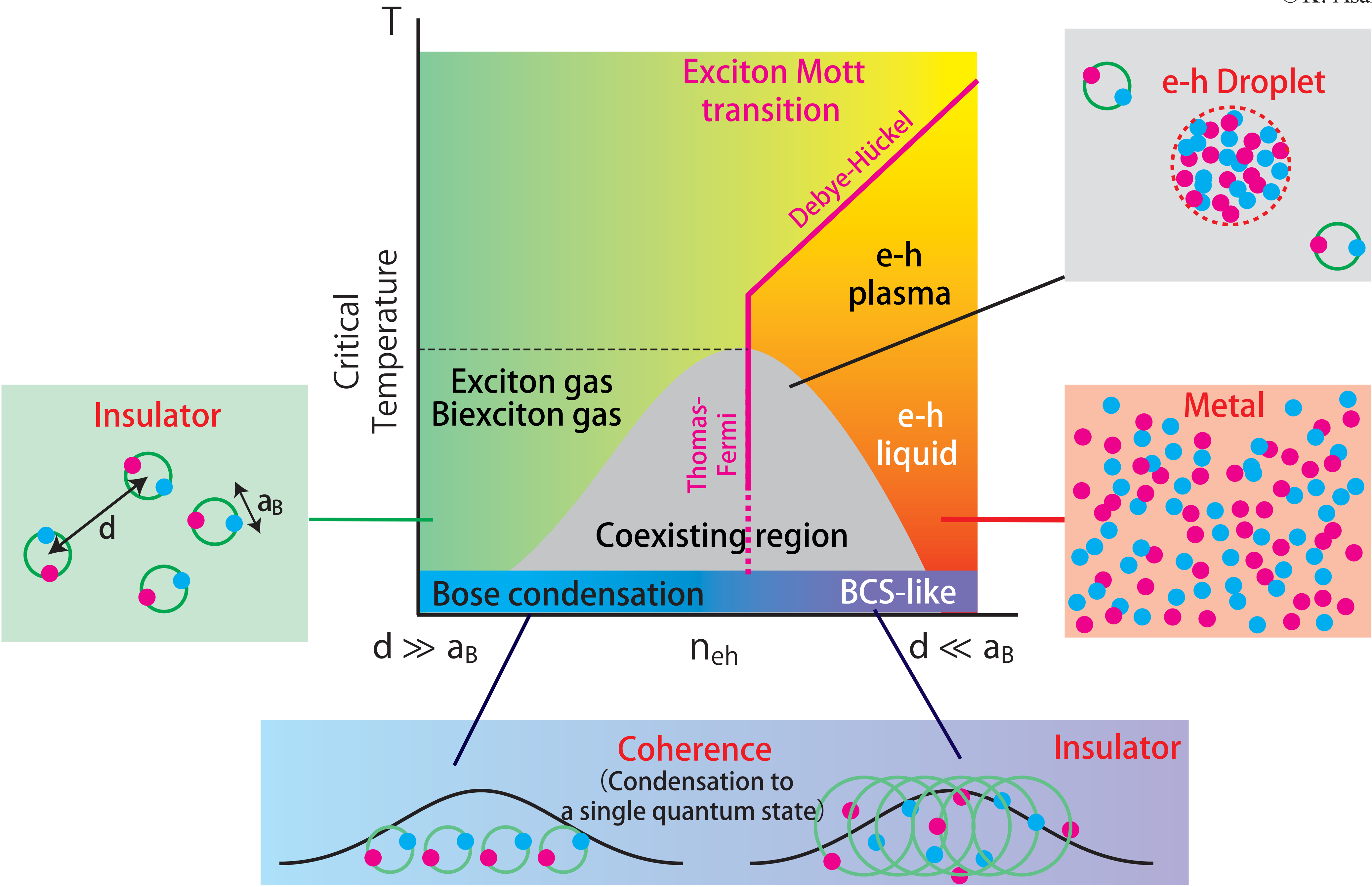
1$^{st}$ step of energy decay, open boundary problem

Open system with photon bath: Interband radiative recombination

2$^{nd}$ step of energy decay, cavity QED, cavity polaritons

# Schematic metastable phase diagram of a 3D e-h system





# PECULIARITY OF ELECTRON-HOLE SYSTEMS

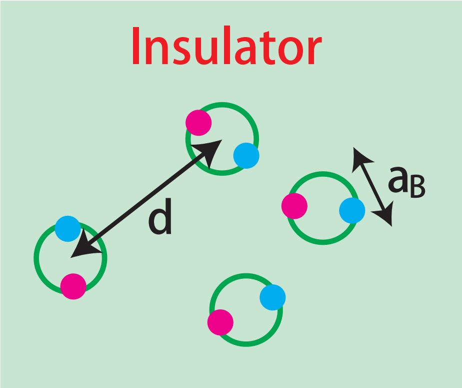
- ➢ DOUBLE-FACE ASPECTS OF BOSONIC AND FERMIONIC
- ➢ DOUBLE-FACE ASPECTS OF METALLIC AND INSULATING
  - ✓ Insulating, bosonic exciton/biexciton gas ⇔ Metallic, fermionic e-h plasma
- ➢ DOUBLE-FACE ASPECTS OF CLASSICAL AND QUANTUM
  - ✓ e-h plasma/liquid condensation ⇔ exciton BEC/e-h BCS condensation

- ➢ FINE TUNABILITY FROM WEAK TO STRONG COUPLING
  - ✓ Fine tuning of particle density, dynamical screening

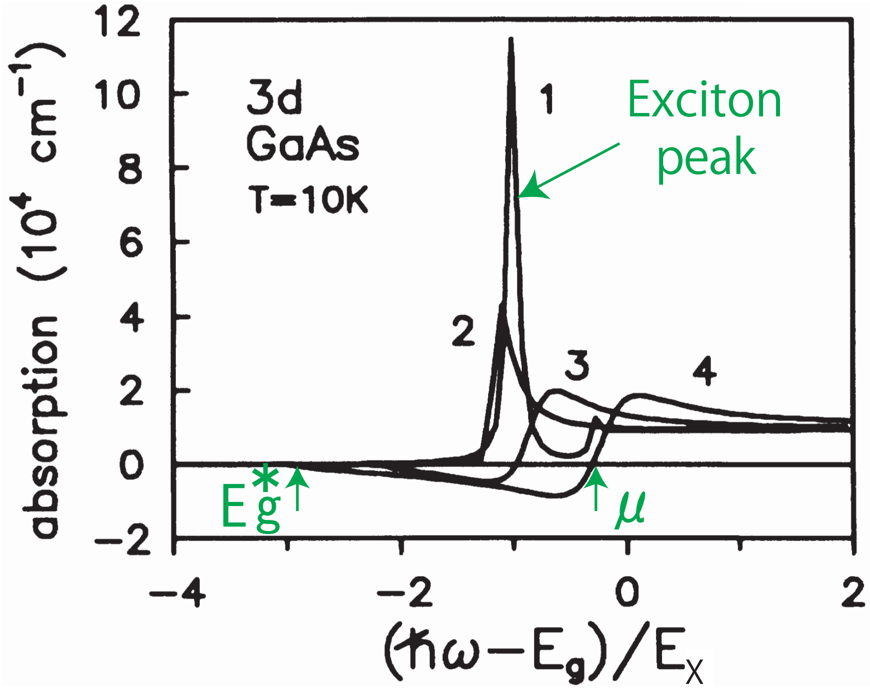
- ➢ FINE-CONTROLLED PREPARATION OF INITIAL STATES
  - ✓ Photo-pumping vs carrier-injection
  - ✓ cw pumping vs pulsed pumping
  - ✓ Tuning of energy profile, particle density, carrier balance, coherence, polarization
- ➢ FINE-CONTROLLED COUPLING TO PHOTON FIELDS
  - ✓ Coherence in material (e-h) system
  - ✓ Coherence in material (e-h) + photon systems, polariton-laser crossover
  - ✓ Cavity QED, feedback due to photons, control of interband recombination rate

# Relation between exciton Mott transition & optical gain/absorption

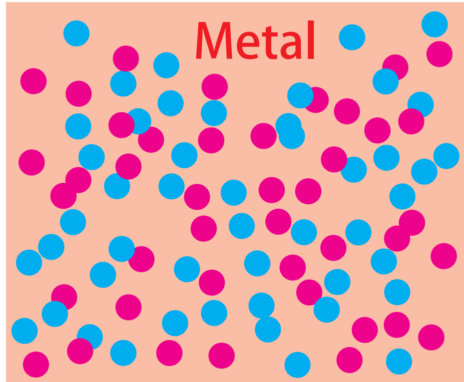
Low density ⟷ ? ⟷ High density



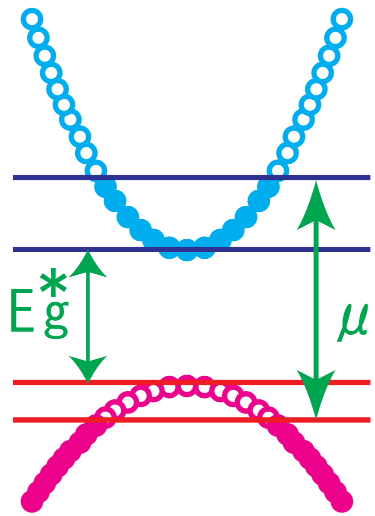

Exciton gas




1. $n_{eh}=0$
2. $n_{eh}=5\times10^{15}cm^{-3}$
3. $n_{eh}=3\times10^{16}cm^{-3}$
4. $n_{eh}=8\times10^{16}cm^{-3}$

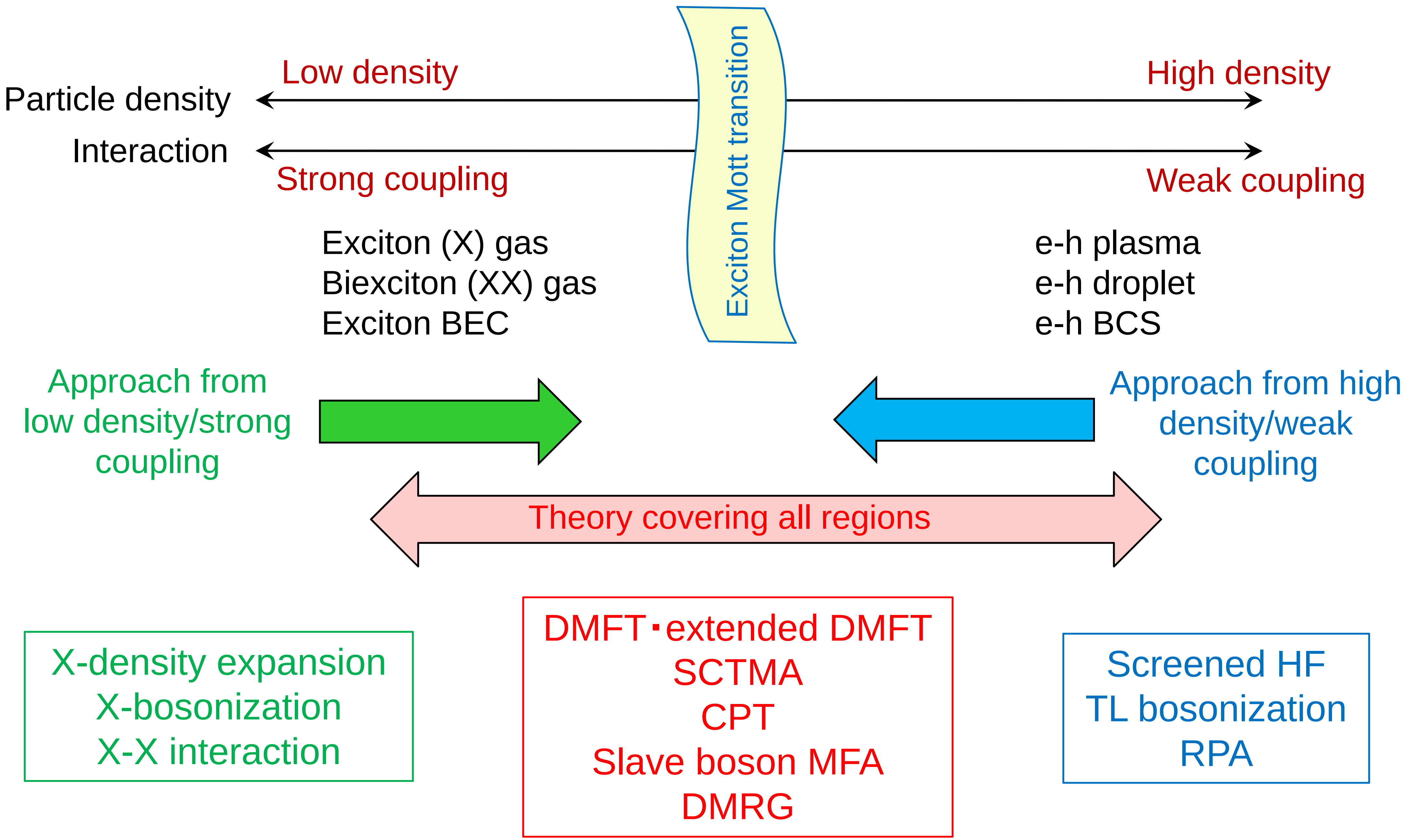



e-h plasma/liquid




Negative absorption = Gain

# MANY-FACETED and UNIFIED APPROACHES



# The e-h (two-band) Hubbard Model

Previous studies cover only local parts of the whole phase diagram.

$\Rightarrow$ We need to obtain globally the phase diagram.

$$\mathcal{H} = -\sum_{\alpha=\mathrm{e,h}}\sum_{\sigma=\uparrow\downarrow}\sum_{\langle ij\rangle} t_\alpha c^\dagger_{i\alpha\sigma} c_{i\alpha\sigma} + U\sum_{\alpha=\mathrm{e,h}}\sum_{i} n_{i\alpha\uparrow}n_{i\alpha\downarrow} - U'\sum_{\sigma\sigma'=\uparrow\downarrow}\sum_{i} n_{i\mathrm{e}\sigma}n_{i\mathrm{h}\sigma'}$$

Kinetic Term

Repulsive Interaction (Intraband)

Attractive Interaction (Interband)

Minimal model of the e-h system

the lattice fermion model (in the tight-binding picture)
- different masses of electron and hole
- screened on-site interactions as parameters independent of density
- Frenkel-type excitons without co-transfer
- modern theoretical tools applicable

cf.) the continuum-space model (in the nearly-free electron picture)
- effective-mass and envelope fn. approximations
- (long-range) Coulomb interactions dependent on density
- Wannier-type excitons with finite bandwidth

# MEAN-FIELD THEORIES for LATTICE FERMION MODELS

How can electronic correlation and quantum fluctuation be treated?



| ω -dependence / k-dependence | | Static MFA<br>No ω-dependence<br>GS at zero temperature | Dynamical MFA<br>ω-dependence<br>Finite temperature, DOS |
|---|---|---|---|
| Correlation neglected | One-body Approx.<br>No fluctuation | **Hartree Approx.**<br>No Mott transition<br>Only energy shift<br>→ Hartree-Fock approx. | **Coherent Potential Approx. (CPA)**<br>Mott insulator OK<br>Only upper/lower Hubbard bands<br>→ RPA, SCTMA |
| Exact correlation effects in a site<br>No k-dependence | Local correlation | **Slave-Boson MFA**<br>Equivalent to Gutzwiller variational<br>Mott transition with mass divergence:<br>Brinkmann-Rice picture OK<br>Only coherent peak | **Dynamical MF Theory (DMFT)**<br>Both coherent peak and U/L Hubbard bands<br>Exact in ∞ dimensions<br>Map to effective Anderson model<br>Numerical calculation → Finite size effects |
| Exact correlation effects in many sites (a cluster)<br>k-dependence | Nonlocal correlation | Cluster Slave-Boson MFA (?)<br>Not yet | **Quantum Cluster Theory (QCT)**<br>1 or 2 dimensions OK<br>Intersite interaction OK<br>CPT, cell-DMFT, DCA<br>Numerical calculation → Finite size effects |

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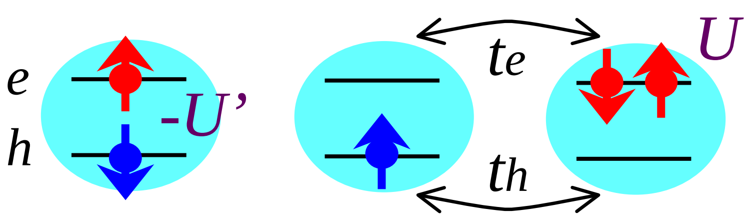
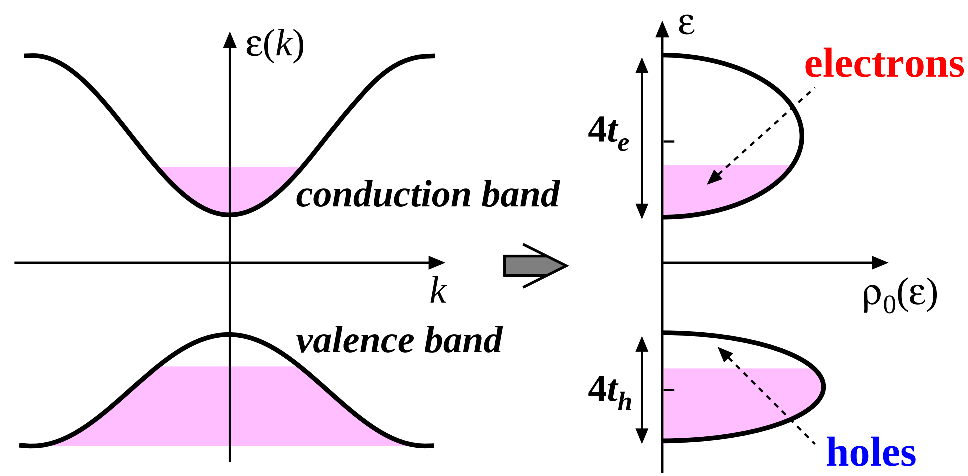
# 2-band Hubbard Model and bare DOS (Tomio)

A lattice fermion model (site representation)

$$
\begin{aligned}
H \;=\; & -\sum_{<ij>\sigma} t_e d^{e\dagger}_{i\sigma} d^{e}_{j\sigma} - \sum_{i\sigma} \mu_e d^{e\dagger}_{i\sigma} d^{e}_{i\sigma} \\
& -\sum_{<ij>\sigma} t_h d^{h\dagger}_{i\sigma} d^{h}_{j\sigma} - \sum_{i\sigma} \mu_h d^{h\dagger}_{i\sigma} d^{h}_{i\sigma} \\
& + U \sum_{i} n^{e}_{i\uparrow}\, n^{e}_{i\downarrow} + U \sum_{i} n^{h}_{i\uparrow}\, n^{h}_{i\downarrow} - U' \sum_{i\sigma\sigma'} n^{e}_{i\sigma} n^{h}_{i\sigma'}
\end{aligned}
$$

- $d^{e\dagger}_{i\sigma}$ : creation operator of electron
- $d^{h\dagger}_{i\sigma}$ : creation operator of hole
- $U$ : e-e (h-h) on-site repulsion
- $-U'$ : e-h on-site attraction



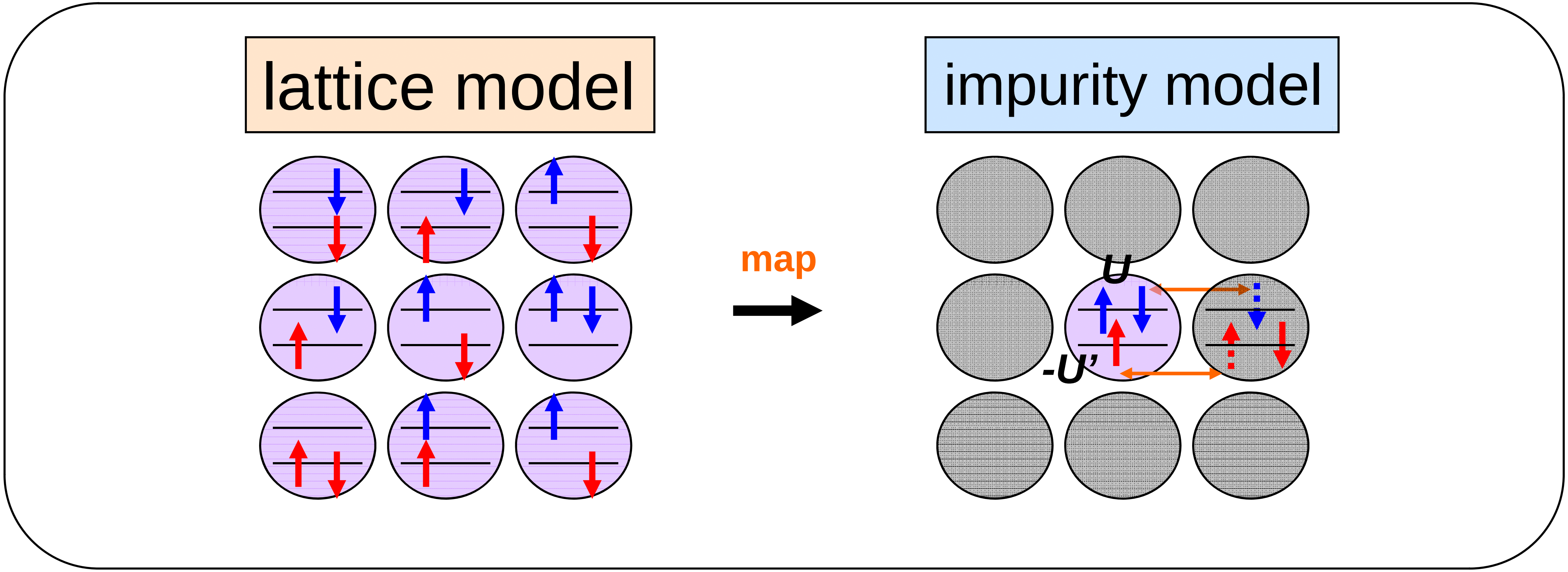

- **Elliptic DOS** assumed
- **Undoped** initially (#e=#h)

# Dynamical Mean-Field Theory (DMFT)

Georges, *et al.*, Rev. Mod. Phys. 68 (1996) 13

**lattice model**

mapping ⇒ **single-site impurity problem embedded in effective medium**



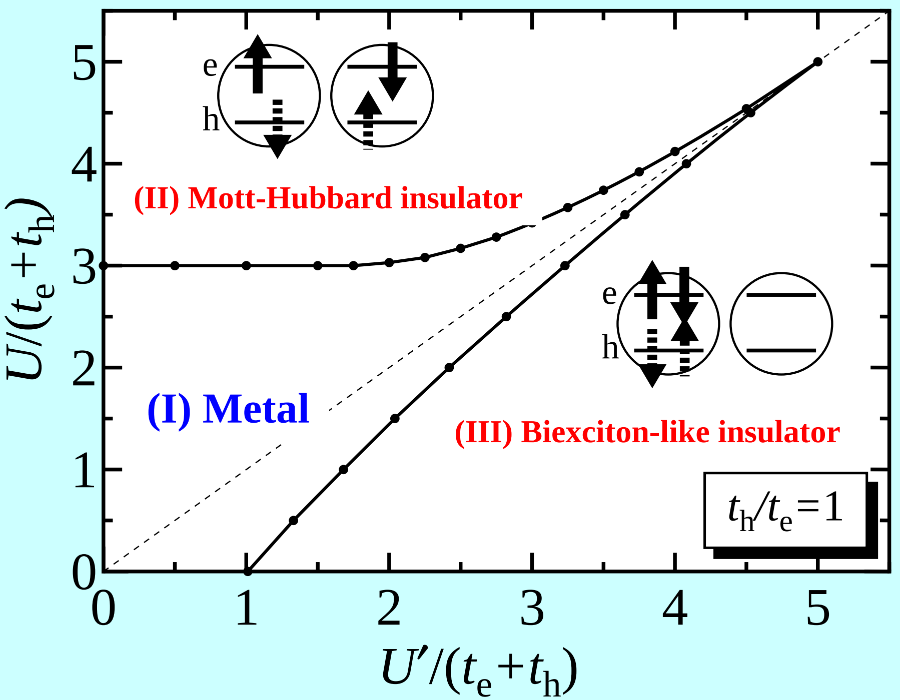

**DMFT requires only locality of self-energy**

**DMFT becomes exact in** $d \to \infty$ **and good approximation in** $d = 3$

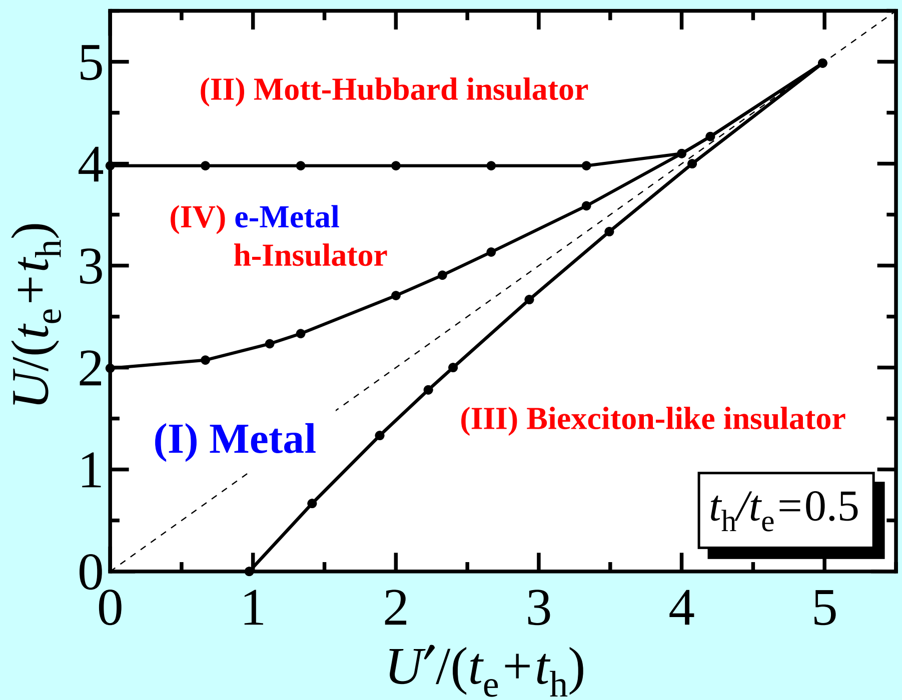
⇐ **DMFT includes full local correlations**

# Phase diagram at 1/2 filling ($n$=1)

$t_h/t_e = 1$




$t_h/t_e = 0.5$




(I)⟺(II): "band-selective" Mott-Hubbard transition except for $t_h/t_e = 1$

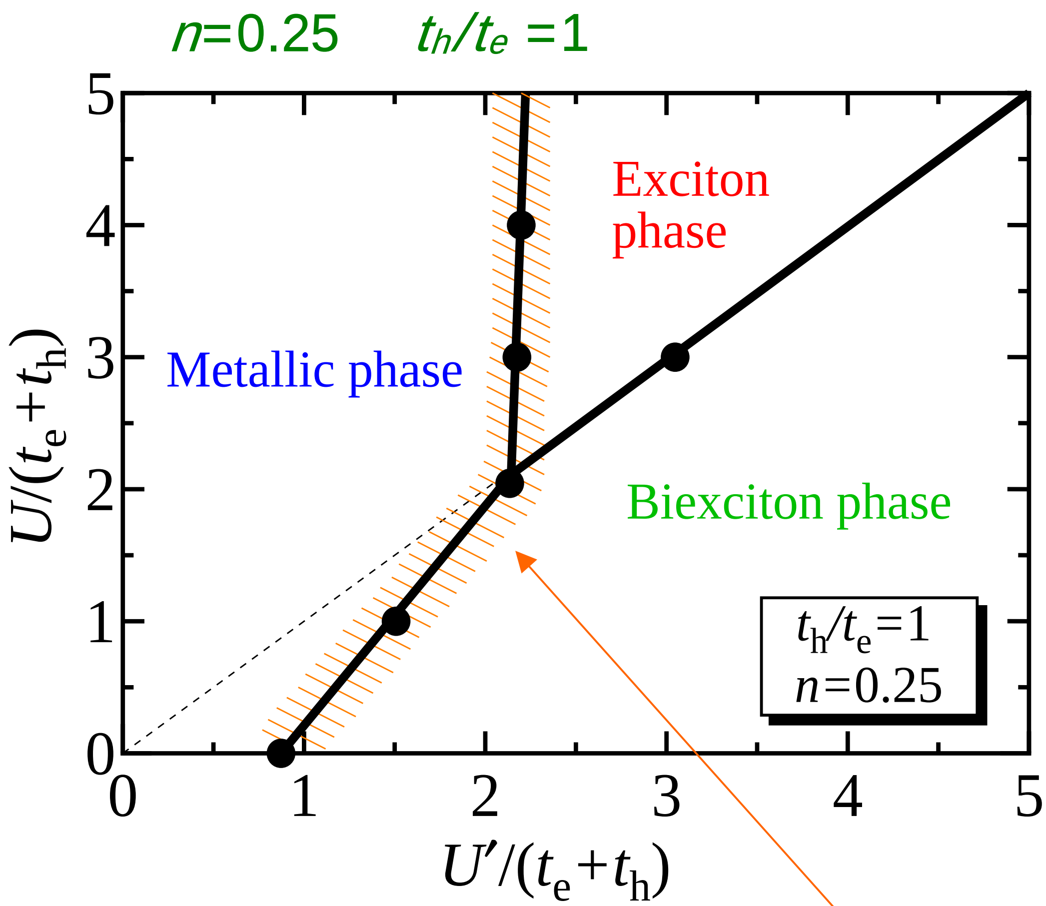
(I)⟺(III): no "band-selective" transition for any $t_h/t_e$

position of phase boundary is universal with regard to $t_h/t_e$

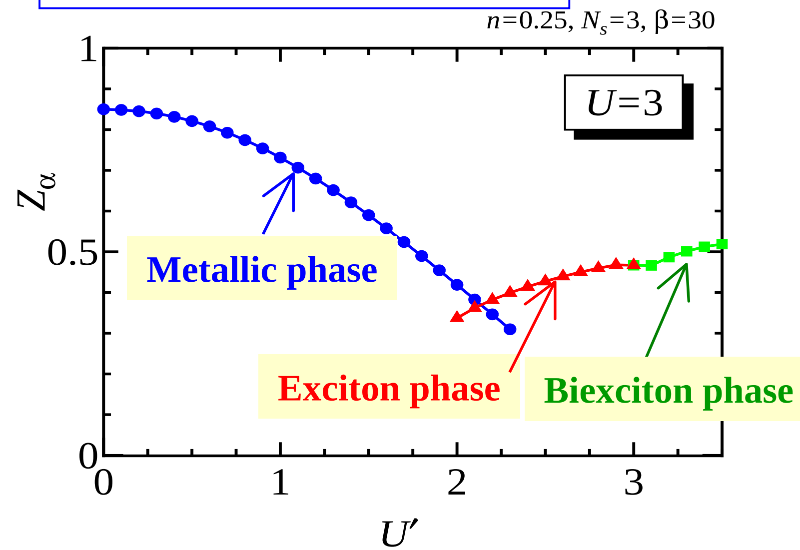
*competition of interactions and e-h relative motion*


cf. **2-orbital Hubbard model**: Koga, *et al.*, PRB 66 (2002) 165107,
**"orbital selective" Mott transition**: Koga, *et al.*, PRL 92 (2004) 216402

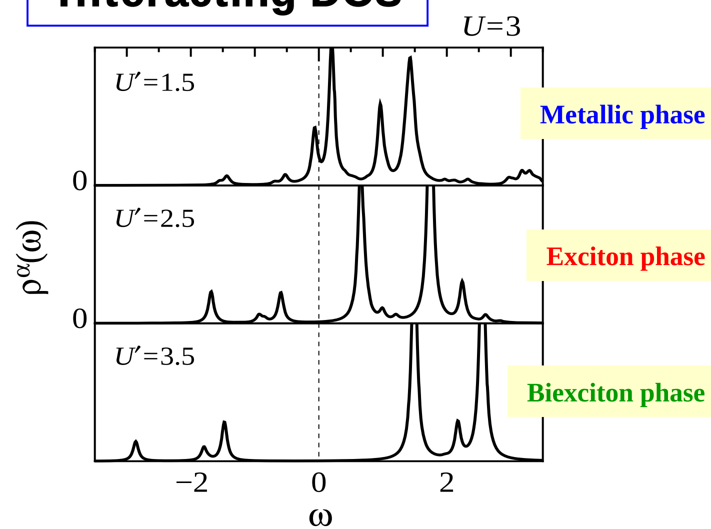
# Phase diagram for 2-band Hubbard model at $T$=0




**Quasiparticle weights**




**Interacting DOS**



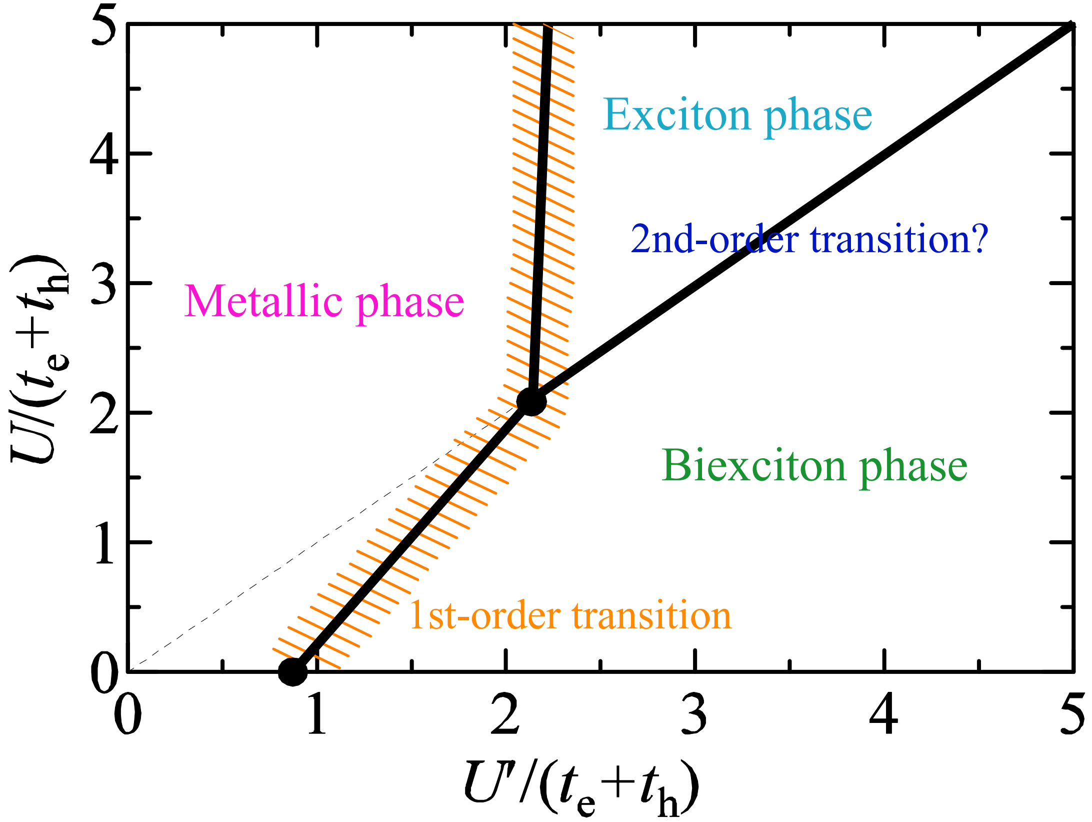

**Metal↔ Exciton / Biexciton phase :** 1st-order transition

**exciton Mott transition**

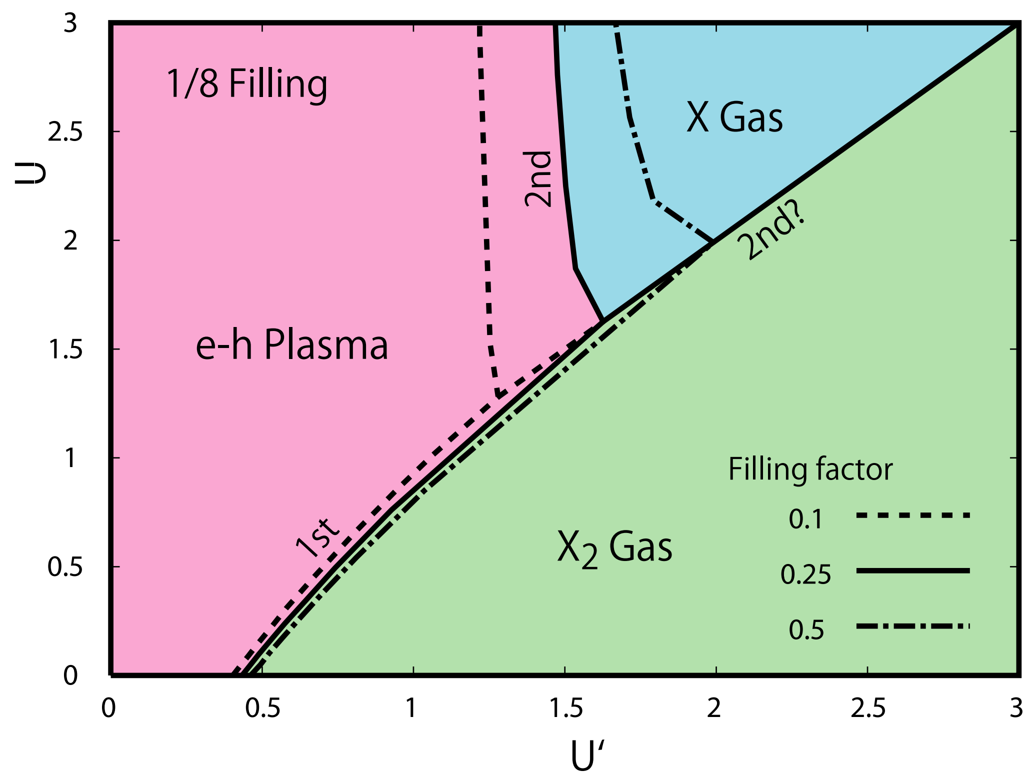
# Phase diagrams for 2-band Hubbard model at T=0

n=0.25 (1/8 filling)    $t_h/t_e=1$    normal phases only

## DMFT

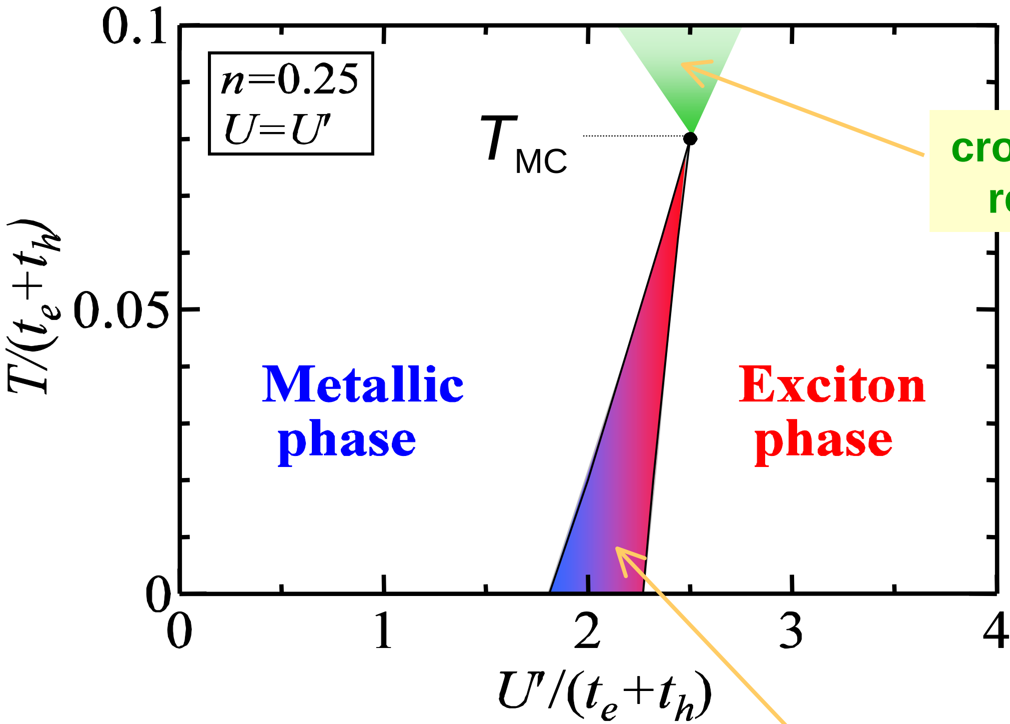



## Slave-boson MFT



# Phase diagram for 2-band Hubbard model at $T>0$ (Tomio)

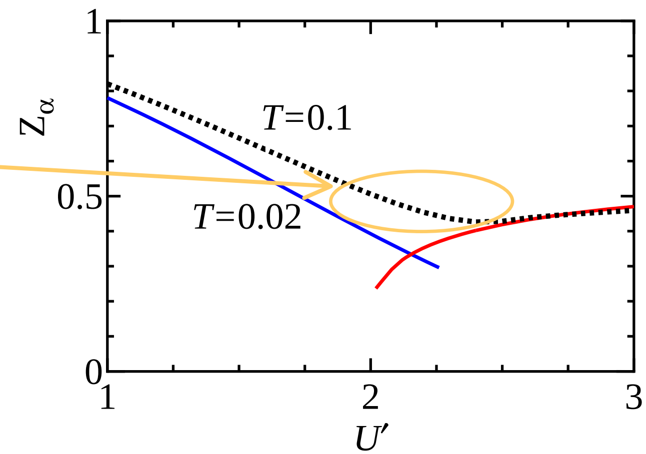
$U=U'$ $n=0.25$ $t_h/t_e=1$



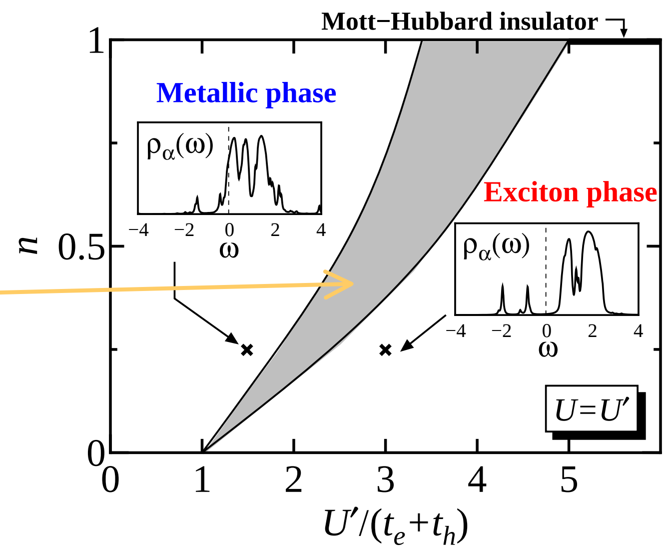

crossover region

coexistent region

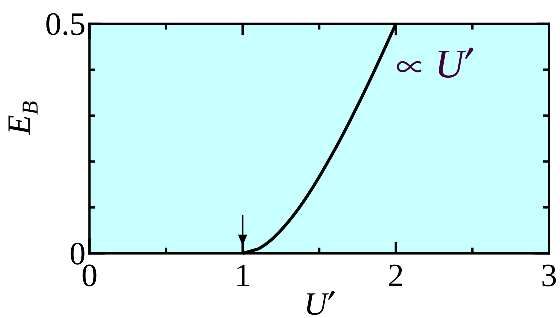
quasiparticle weights




$U'$-$n$ phase diagram at $T=0$




cf. binding energy of a free exciton
→ phase diagram in low-density limit



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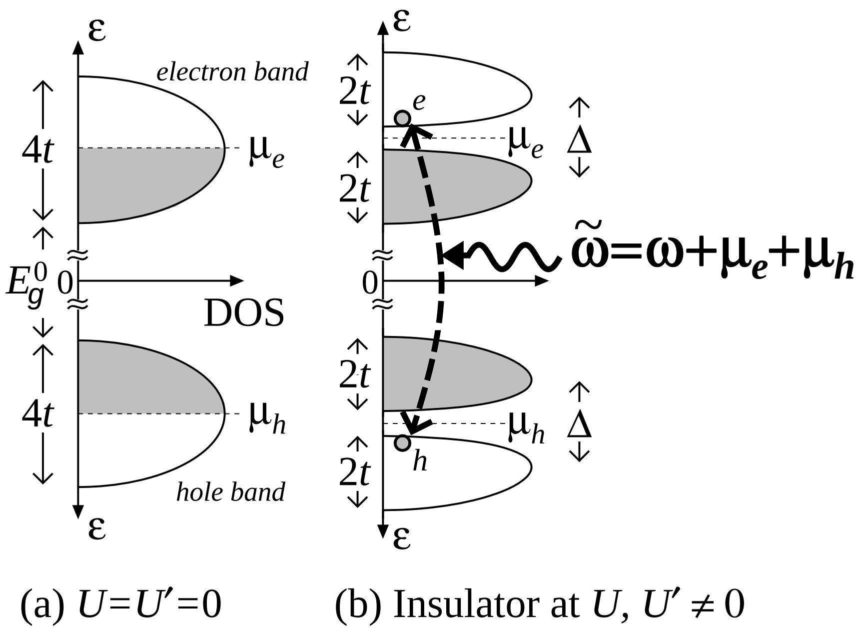
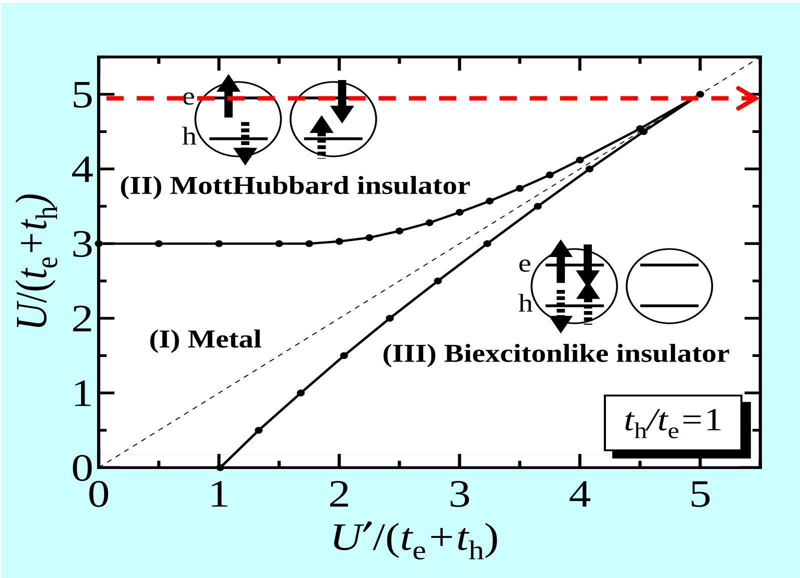
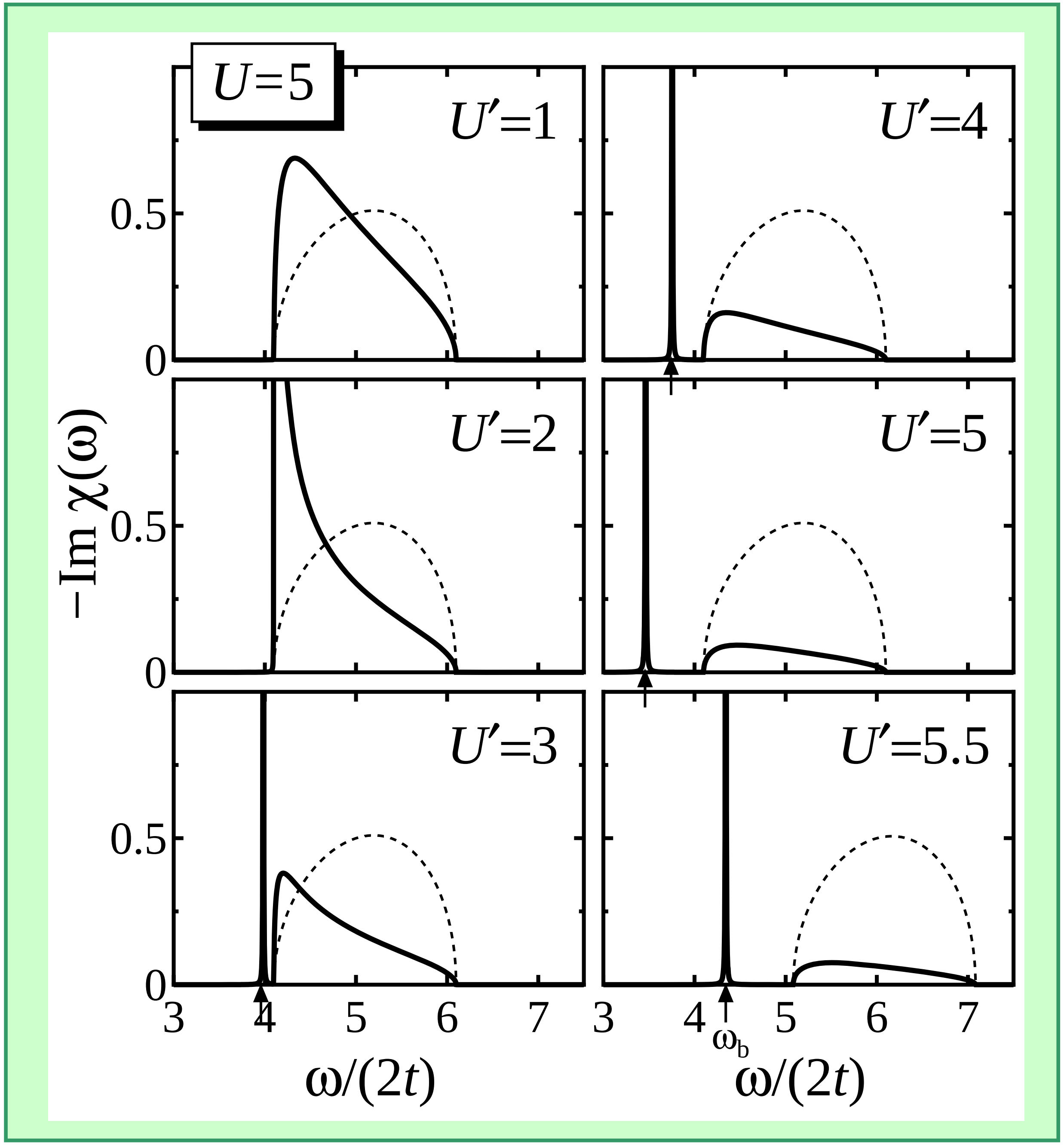
# Absorption spectra in MH / BX insulator



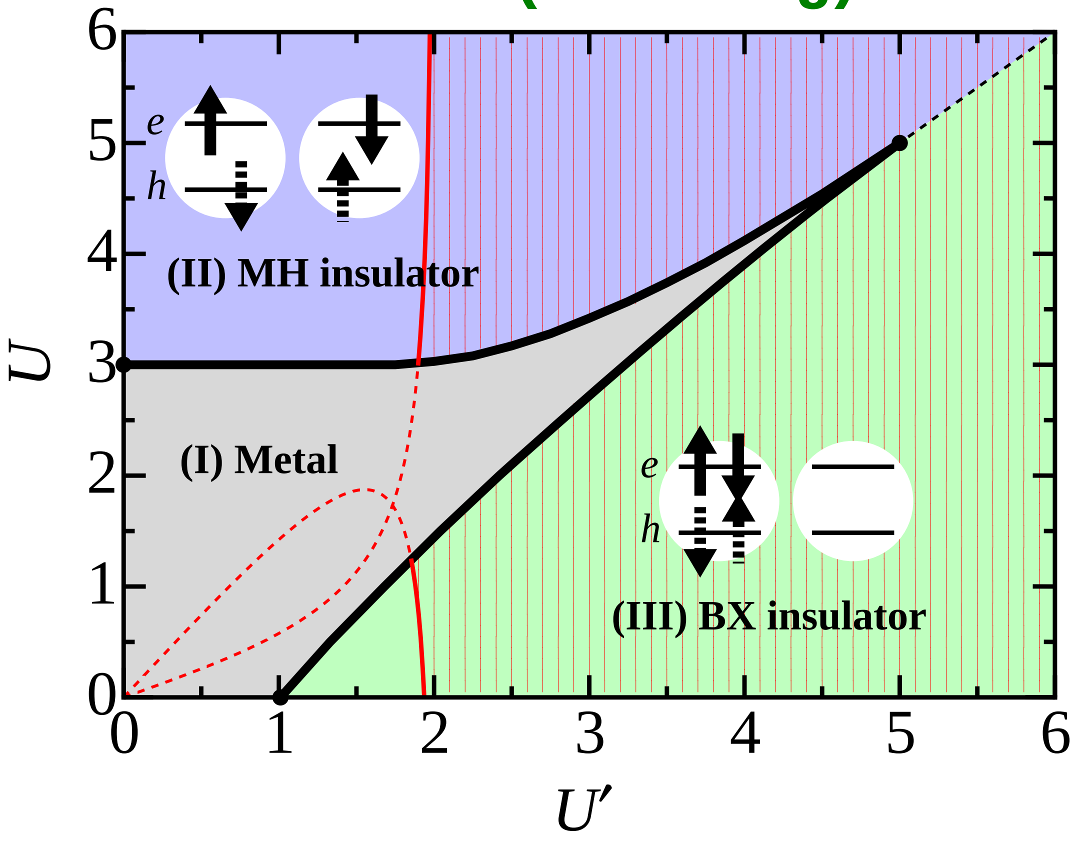



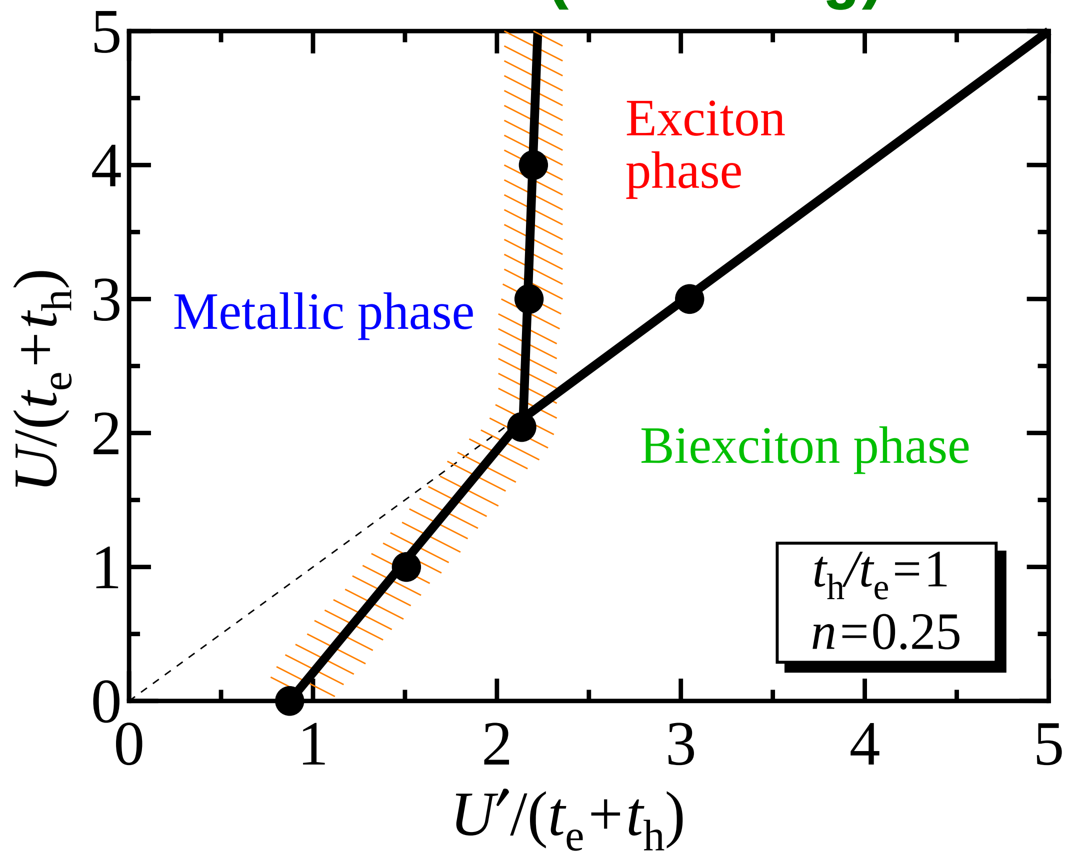
# Isolated absorption peak vs exciton-like insulator phase

$t_h / t_e = 1$

$n$=1.0 (half filling)




$n$=0.25 (1/8 filling)




Region for an isolated absorption peak in half filling
corresponds to the exciton-like insulator phase in not half filling.

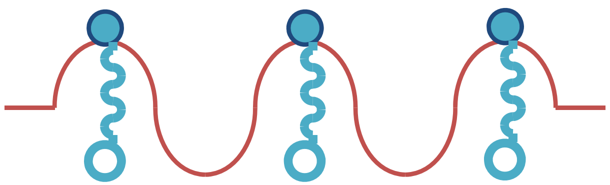
# Effects of inter-site interactions on EMT

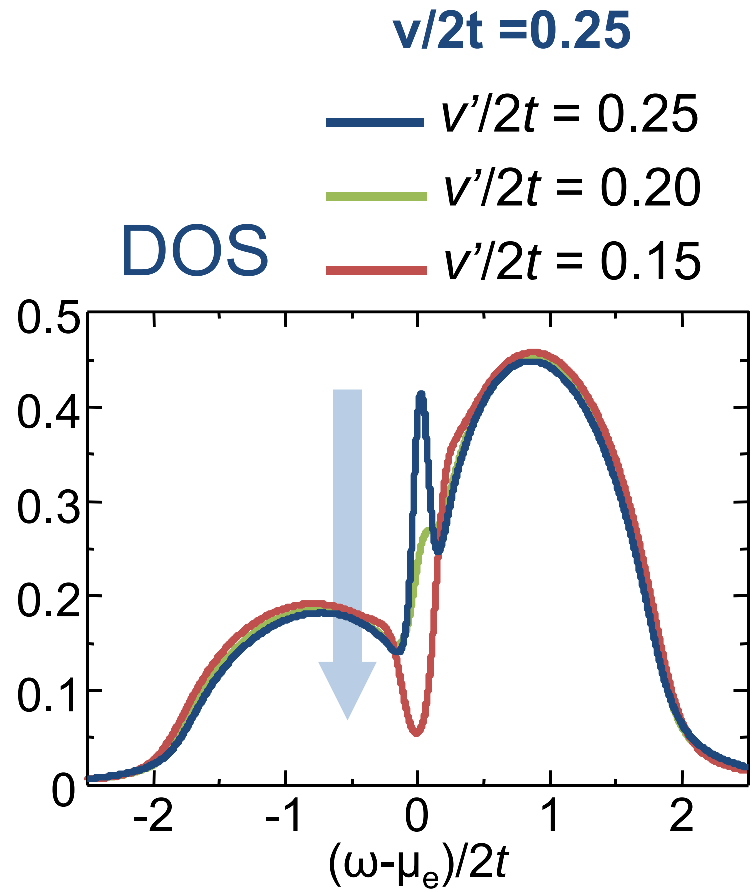
## Extended spinless Hubbard model

$$H = -t \sum_{\langle i,j\rangle,\alpha=e,h} c_{i\alpha}^{+} c_{i\alpha} - U' \sum_{i} c_{ie}^{+} c_{ie} c_{ih}^{+} c_{ih} + v \sum_{\langle i,j\rangle,\alpha=e,h} c_{i\alpha}^{+} c_{i\alpha} c_{j\alpha}^{+} c_{j\alpha} - v' \sum_{\langle i,j\rangle,\alpha=e,h} c_{i\alpha}^{+} c_{i\alpha} c_{j\bar{\alpha}}^{+} c_{j\bar{\alpha}}$$

## Extended Dynamical mean-field theory

- ( e-e & h-h repulsion v ) = ( e-h attraction v' )
  - does not change the nature of MIT
  - tends to stabilize the metallic e-h plasma
- ( e-e & h-h repulsion v ) > ( e-h attraction v' )
  - stabilizes the insulating state
  - enhances mass density wave fluctuations





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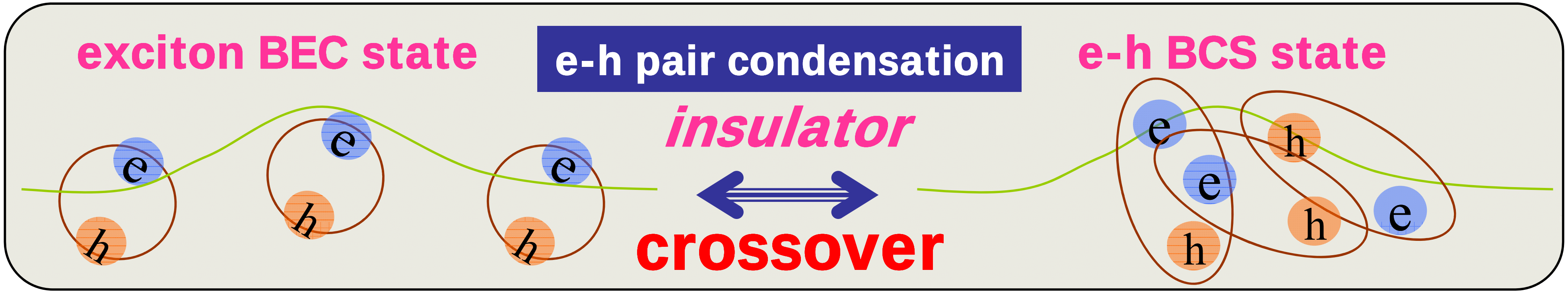
# Excitonic BEC-BCS crossover

**Quantum mechanical coherence at low temperatures**




| | | |
|---|---|---|
| $d >> a_B$ | | $d < a_B$ |
| **strong coupling** | ⟷ | **weak coupling** |
| bosonic feature | Coulomb correlation plays a key role in crossover problem. | fermionic feature |

## present study:

**properties of pair condensed phase** (BEC-BCS crossover & optical response)

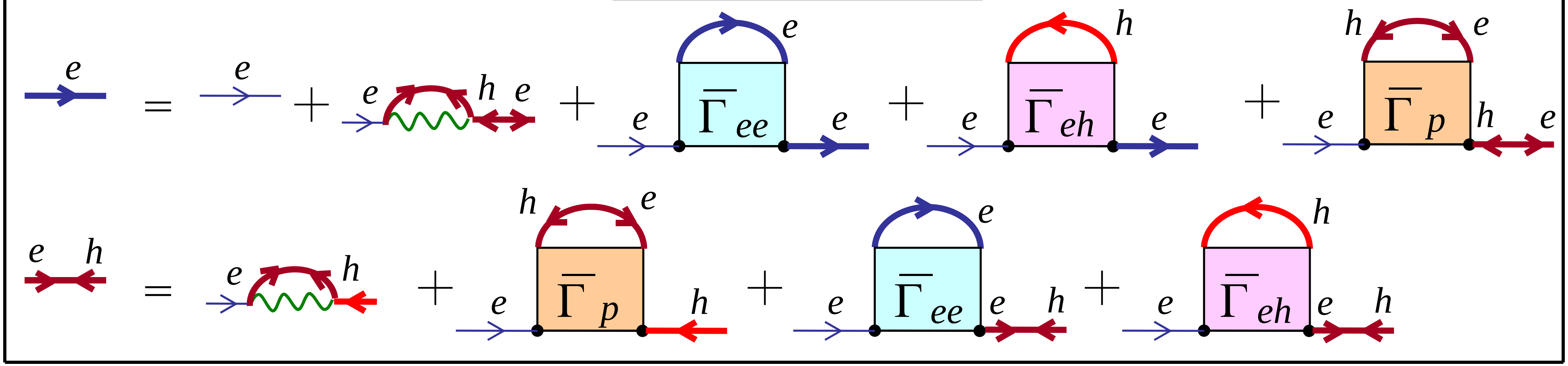
- **simplest model : 2-band Hubbard model**
- **method: self-consistent *t*-matrix approx. (SCTMA) + local approx. (LA)**

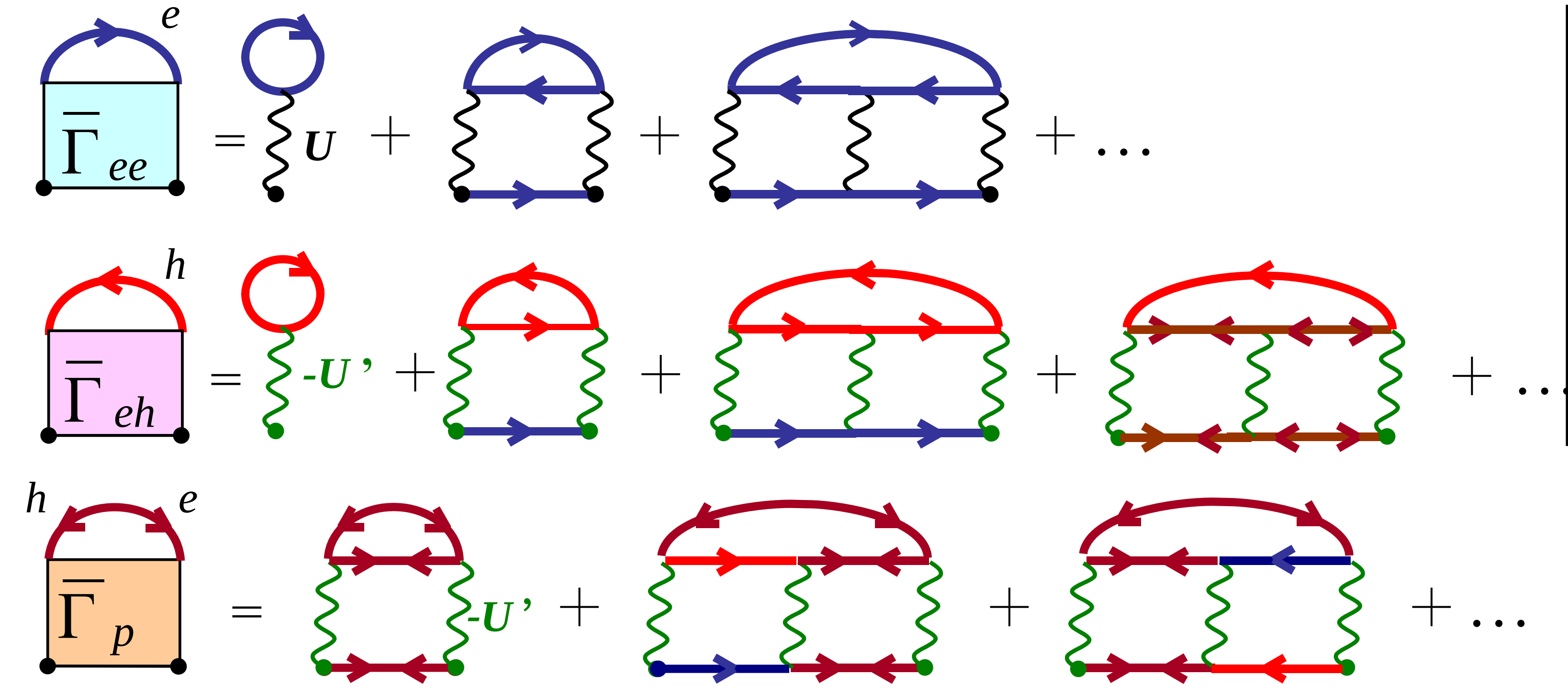
→ **systematic study on optical spectra and excitonic BEC-BCS crossover at finite temperatures**

# SCTMA for e-h pair condensed phase

SCTMA: self-consistent $t$-matrix approx.

## Dyson equation




**local approx. (LA)**

$$\Gamma(\boldsymbol{q}, i\omega_n) \Downarrow \overline{\Gamma}(i\omega_n)$$

**SCTMA+LA**:
valid for
**low-density** &
**high-dimension**

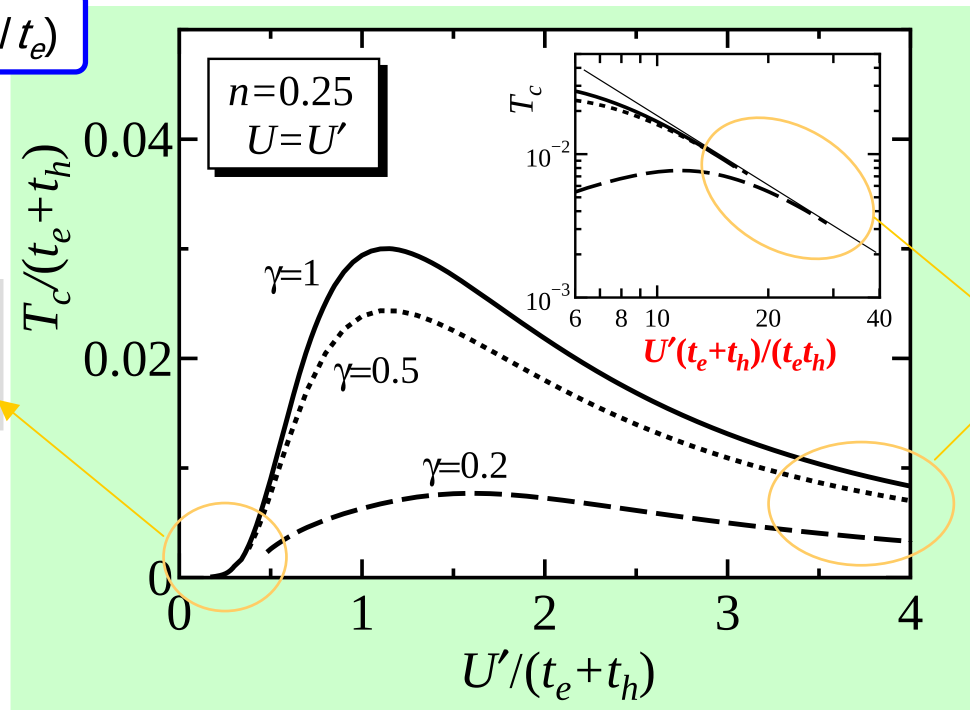
# Condensation temperature in BCS-BEC crossover (Tomio)

effect of mass difference ($\gamma = t_h / t_e$)

$$T_c = 1.13\sqrt{w_e w_h}\exp\left[-\frac{t_e + t_h}{2t_\alpha \rho_\alpha^0(\varepsilon_F^\alpha)U'}\right]$$

$w_\alpha$ : cutoff energy ~ $O(\varepsilon_F^\alpha)$

$$T_c^{BCS} \propto \frac{\sqrt{\gamma}}{1+\gamma}$$



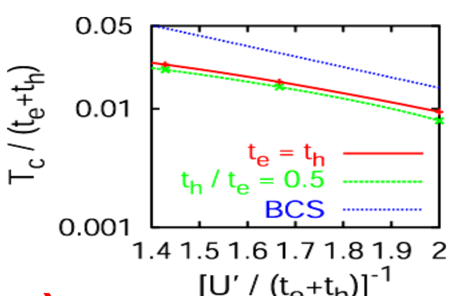

$$T_c = \frac{2t_e t_h}{U'}\frac{2n-1}{\ln[n/(1-n)]}$$

$$T_c^{BEC} \propto \frac{\gamma}{(1+\gamma)^2}$$

**weak-coupling region**

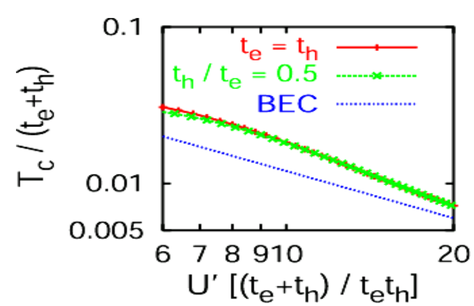
$$\frac{1}{t_e + t_h} \propto \frac{m_e m_h}{m_e + m_h}$$

reduced mass
→relative motion




**BCS-BEC crossover**

**strong-coupling region**




$$\frac{t_e + t_h}{t_e t_h} \propto m_e + m_h$$

motion of
center of mass

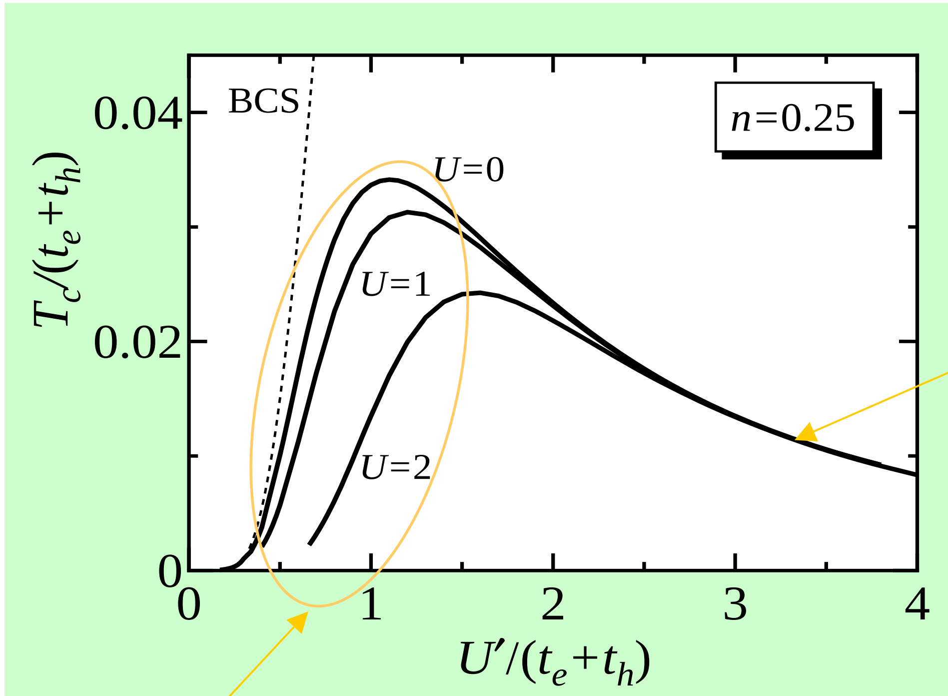
**e-h BCS (excitonic ins.)**

**exciton BEC**

# Condensation temperature in BCS-BEC crossover (Tomio)

effect of $U$

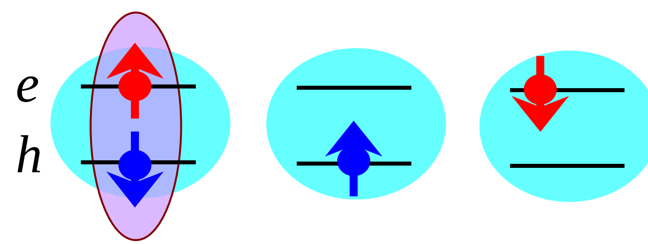
$\gamma = t_h / t_e = 1$




$T_c$ is insensitive to $U$ in BEC regime.

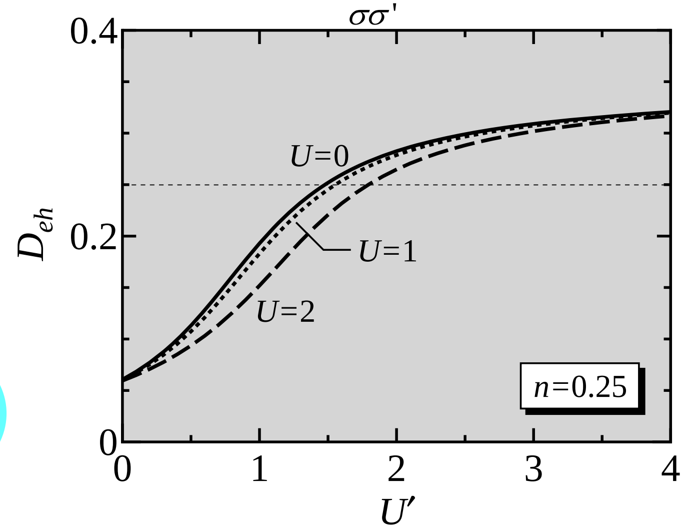
In BCS regime,
$T_c$ is strongly suppressed by $U$.

suppression of
excitonic correlation




**density of occupied sites**

$$D_{eh} = \sum_{\sigma\sigma'} \langle n^e_{i\sigma} n^h_{i\sigma'} \rangle$$



Contents:

- Electron-hole systems in quasi-thermal-equilibrium
  - Quantum cooperative phenomena
  - Modern theoretical tools and e-h Hubbard model
- Exciton Mott transition in high-*d*: DMFT and slave-boson MFT
  - Absorption spectra in the half-filled case
  - Role of inter-site interaction : Extended DMFT
- Quantum pair condensation and crossover in high-*d*: SCTMA
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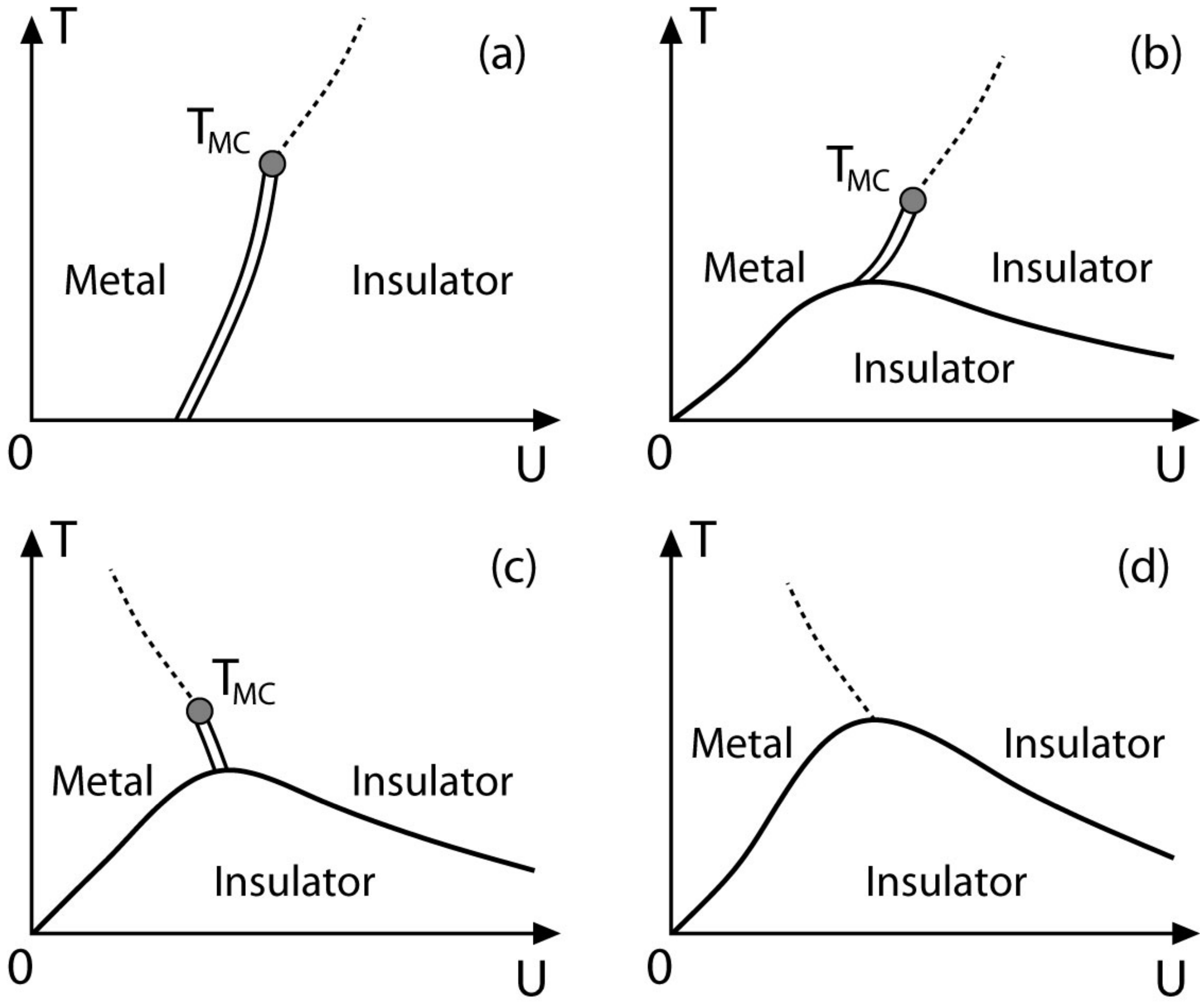
- Absence of exciton Mott transition in 1-*d*: bosonization, RG, and CPT

# TOPOLOGY OF COMPLETE PHASE DIAGRAM




EMT vs QC

Low-density regime:
QC is harder to emerge.
EMT can be observed.
→ (b) or (c)

High-density regime:
QC is easier to emerge.
EMT is buried in QC.
→ (d)

Role of repulsion:
QC is suppressed.
→EMT is easier to be observed.
cf.) Attractive Hubbard model

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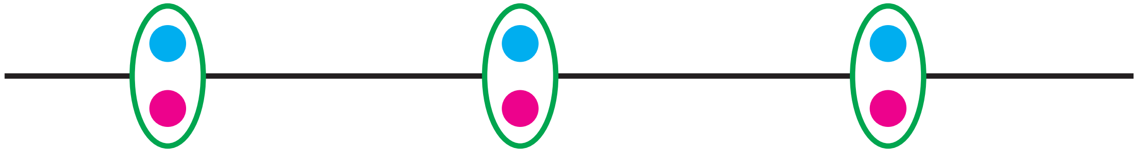
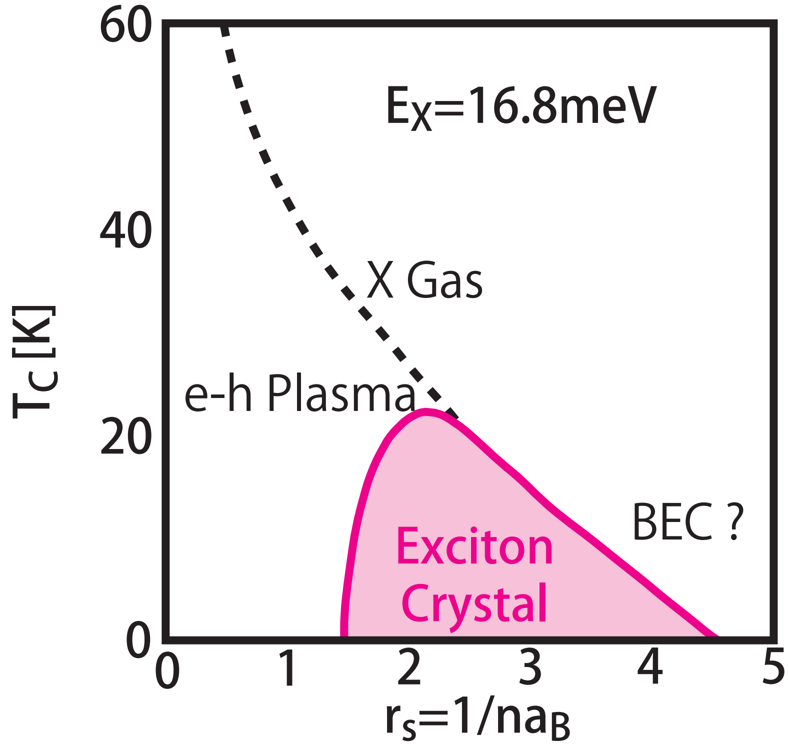
# Exciton Crystal

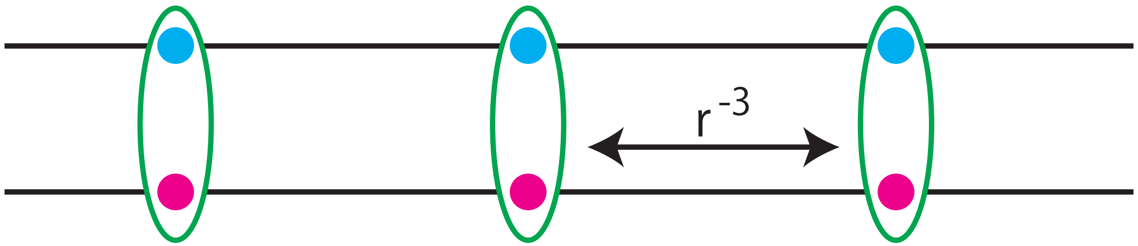
*Ivanov and Haug PRL71(1993) 3182.*



Spinless electrons and holes
⇒ Repulsive X-X interaction
**Heitler-London Method**
**Mott's Criterion at T=0**




*Arkhipov et al. cond-mat 0505700.*




**Bosons with dipole-dipole interaction**

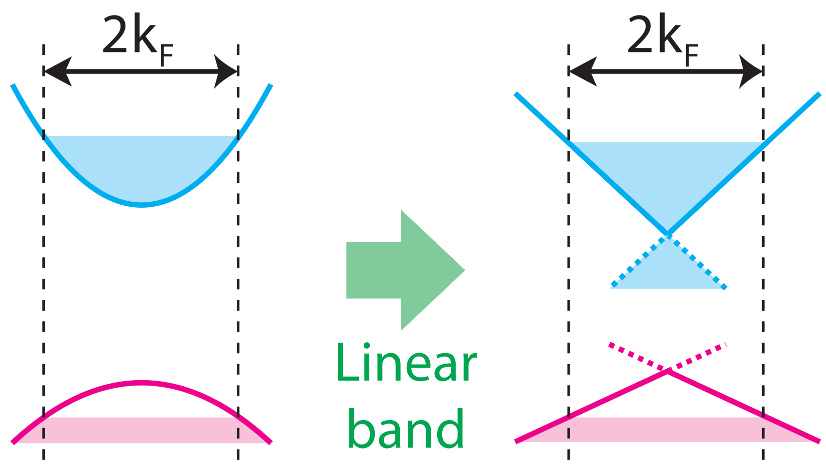
**Spin ⇒ Biexciton Crystal ? ?**

# g-ology in 1D e-h systems

$$g^{(ij)}_{2,4}(q) \sim g^{(ij)}_{2,4} + g_L \ln(1/q)$$

Short Range

Long-range Coulomb




Backward scatterings

Forward scatterings ⇒ Excactly solvable

Scaling dimension

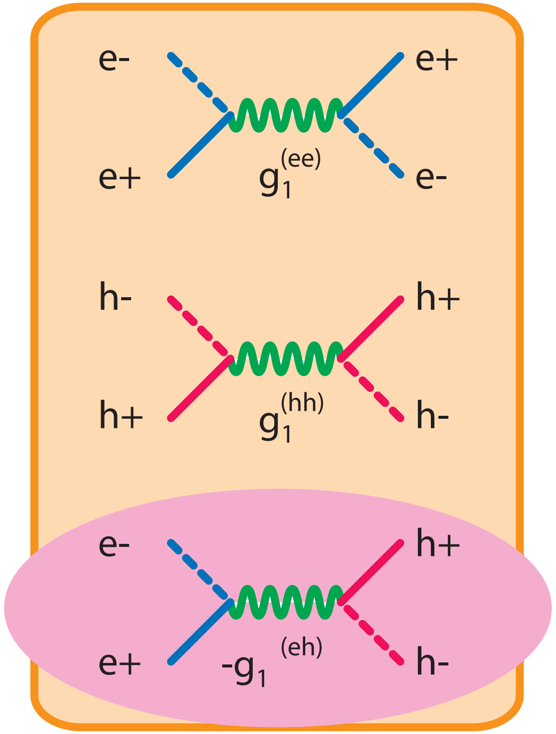
**Intraband BS**

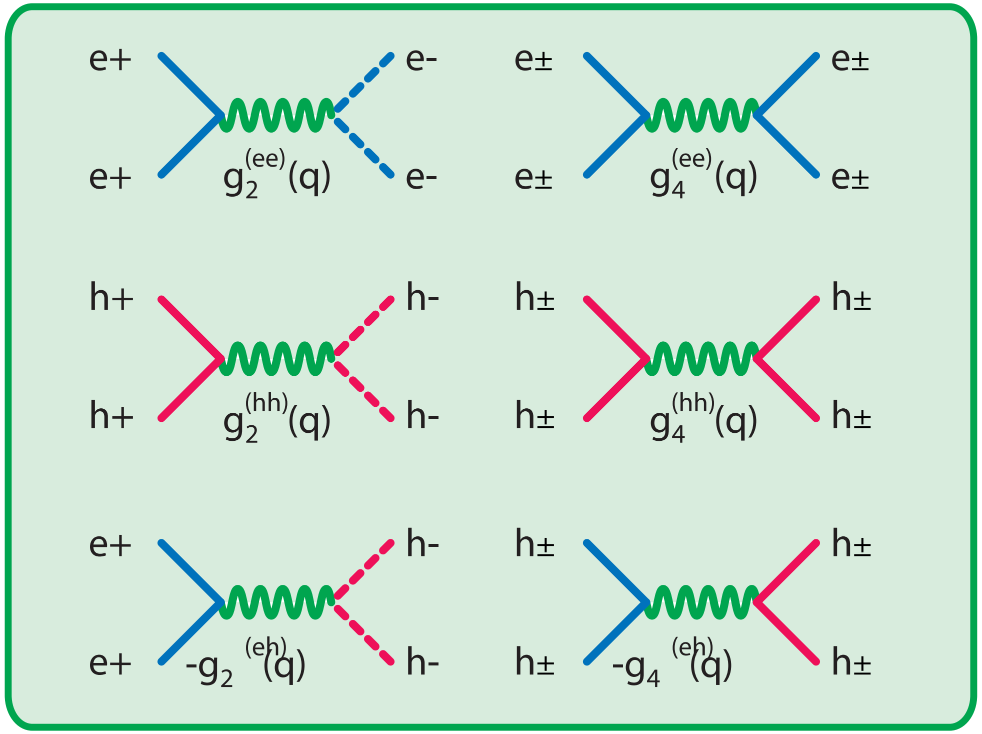
V

**Interband BS**

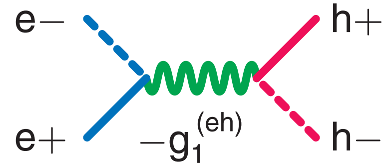
exists only in nondoped case

$k_F^{(e)} = k_F^{(h)}$



**Backward Scattering** RG / SCHA ⇒ Relevancy




**TL Hamiltonian** **Phase Lock**

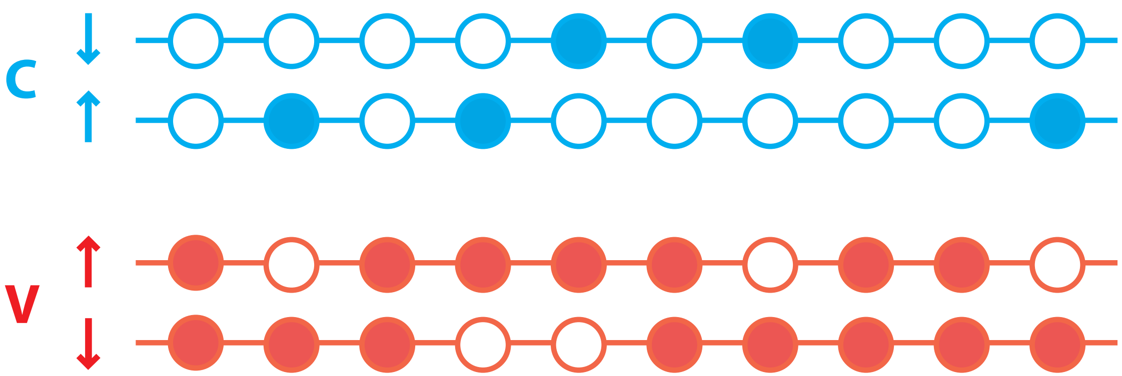
$$\mathcal{H}_\rho^{(c)} + \mathcal{H}_\sigma + \cancel{\mathcal{H}_{bs}^{(ee)} + \mathcal{H}_{bs}^{(hh)}} + \mathcal{H}_{bs}^{(eh)}$$

$$\mathcal{H}_{bs}^{(eh)} = -\frac{8g_1^{(eh)}}{(2\pi\alpha)^2}\int dx \cos\sqrt{2}\Phi_\rho^{(c)} \cos\sqrt{2}\Phi_\sigma^{(e)} \cos\sqrt{2}\Phi_\sigma^{(h)}$$

**Relevant** ⇒ $\Phi_\rho^{(c)} = 0 \quad \Phi_\sigma^{(e)} = 0 \quad \Phi_\sigma^{(h)} = 0$

| | |
|---|---|
| Charge | Massive |
| Mass | Massless |
| e-Spin | Massive |
| h-Spin | Massive |

Insulator even at the high density limit
⇔ No metal-insulator transition




**"Commensurate Filling"**

# Character of the Ground State

Mass Density ⇔ $\Phi_\rho^{(\mathrm{m})} \propto m^{(e)}\Phi_\rho^{(\mathrm{e})} + m^{(h)}\Phi_\rho^{(\mathrm{h})}$

$$\mathcal{H}_\rho^{(\mathrm{m})} = \frac{v^{(\mathrm{m})}}{2\pi}\int dx \left[K^{(\mathrm{m})}\left(\partial_x \Theta_\rho^{(\mathrm{m})}\right)^2 + \frac{1}{K^{(\mathrm{m})}}\left(\partial_x \Phi_\rho^{(\mathrm{m})}\right)^2\right]$$

$g_\rho = g_\rho^{(\mathrm{e})} + g_\rho^{(\mathrm{h})}$

⇒ Short-range part

$$K^{(\mathrm{m})} = \sqrt{\frac{\bar{v}}{v_\mathrm{F}^{(\mathrm{e})} + v_\mathrm{F}^{(\mathrm{h})} - g_\rho/\pi}} \qquad v^{(\mathrm{m})} = \sqrt{\bar{v}\left[v_\mathrm{F}^{(\mathrm{e})} + v_\mathrm{F}^{(\mathrm{h})} - g_\rho/\pi\right]} \qquad \bar{v} = \frac{v_\mathrm{F}^{(\mathrm{e})} v_\mathrm{F}^{(\mathrm{h})}}{v_\mathrm{F}^{(\mathrm{e})} + v_\mathrm{F}^{(\mathrm{h})}}$$

$$K_\mathrm{free}^{(\mathrm{m})} = \frac{\sqrt{v_\mathrm{F}^{(\mathrm{e})} v_\mathrm{F}^{(\mathrm{h})}}}{v_\mathrm{F}^{(\mathrm{e})} + v_\mathrm{F}^{(\mathrm{h})}}$$ ~ 0.3 (Bulk GaAs)

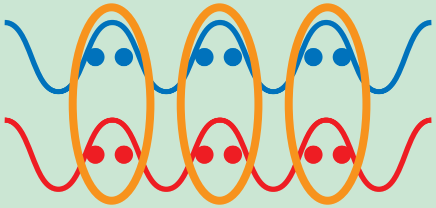
1

∞

$K^{(\mathrm{m})}$

**$2k_F$ Mass Density Wave**

(Biexciton Crystal)



"Acoustic Mode" ⇔ Mass

"Optical Mode" ⇔ Charge

**biexciton**

**BEC/BCS**

Phase Seperation ?

Asymmtery of e and h mass

⇒ Stabilize the biexciton crystal order

**for comparison**

## Phases in a high-densiy 1D e-h system

<table>
<tr><th>T < Δ<sub>gap</sub></th><th colspan="2">T > Δ<sub>gap</sub><br>e-h backward scattering: negligible</th></tr>
<tr><td rowspan="2">Strong instability toward<br>biexciton crystallization<br>(or biexciton BEC)<br><br>Always insulating<br>NO exciton Mott transition</td><td>Short-range<br>Interaction</td><td>h-CDW/SDW<br>(or exciton BEC)<br>Biexcitonic correlation<br>is suppressed.</td></tr>
<tr><td>Long-range<br>Coulomb</td><td>"biexciton liquid"<br>(bad metallic)</td></tr>
</table>

$\Delta_{gap}$ : a charge gap induced by e-h backward scattering

# SUMMARY

Objects:
- Balanced electron-hole systems in quasi-thermal-equilibrium
- In high dimensions and in 1 dimension

Subjects:
- Exciton Mott transition (EMT): X gas / e-h plasma
- Quantum condensation (QC): exciton BEC / e-h BCS

Theoretical tools and models:
- e-h Hubbard model, DMFT, slave-boson MFA
- extended e-h Hubbard model, extended DMFT
- Self-consistent T-matrix approx. (SCTMA)
- e-h Tomonaga-Luttinger model, bosonization, RG, SCHA
- Cluster perturbation theory (CPT)

Important findings:
- Two insulating phases: exciton-like and biexciton-like
- EMT: $1^{st}$-order transition
- MC temperature vs QC temperature
- XX crystal and absence of exciton Mott transition in 1*d*
- …

# PROBLEMS TO BE SOLVED

1) From lattice fermion model to continuum-space fermion model

DMFT for continuum models (Ohashi, Ueda, TO)

Excitation modes: collective – individual

Comparison with hydrogen metals, positronium condensates, superconductivity

2) Description of low-density regime (X gas, XX gas)

Self-consistent exciton-gas theory (Hanamiya, Asano, TO: Physica E **40**, 1401 (2008)).

Biexciton Auger scattering (Watanabe, Asano, TO)

3) Description of intermediate-density regime (e-h-X-XX mixture, crossover)

Dynamical screening

4) Dynamics for/against condensation

Photo excitation vs Current injection, e-h transport, Relaxation/condensation dynamics

5) Imbalanced e-h systems

FFLO order (Yamashita, Asano, TO)

6) Dimensionality

Cluster perturbation theory (Asano, Nishida, TO), Dynamical DMRG (Asano, TO)

7) Photon physics

Two-particle Green function, diagrammatic extension of DMFT (Ohashi, TO)

Cavity effects, Temporal/spatial coherence of PL, Polariton condensation vs lasing,

Nonlinear optical responses, Lasing process

# SIMILAR SYSTEMS

$T=10^{12}$ K Hadron QCD systems:

quark-gluon plasma (QGP) vs hadron gas/liquid vs color SC

$T=10^{5}$ K Warm dense matter:

weakly-coupled plasma vs condensed matter

T=1 K e-h systems (condensed matter):

e-h plasma/liquid vs exciton gas vs exciton BEC, e-h BCS

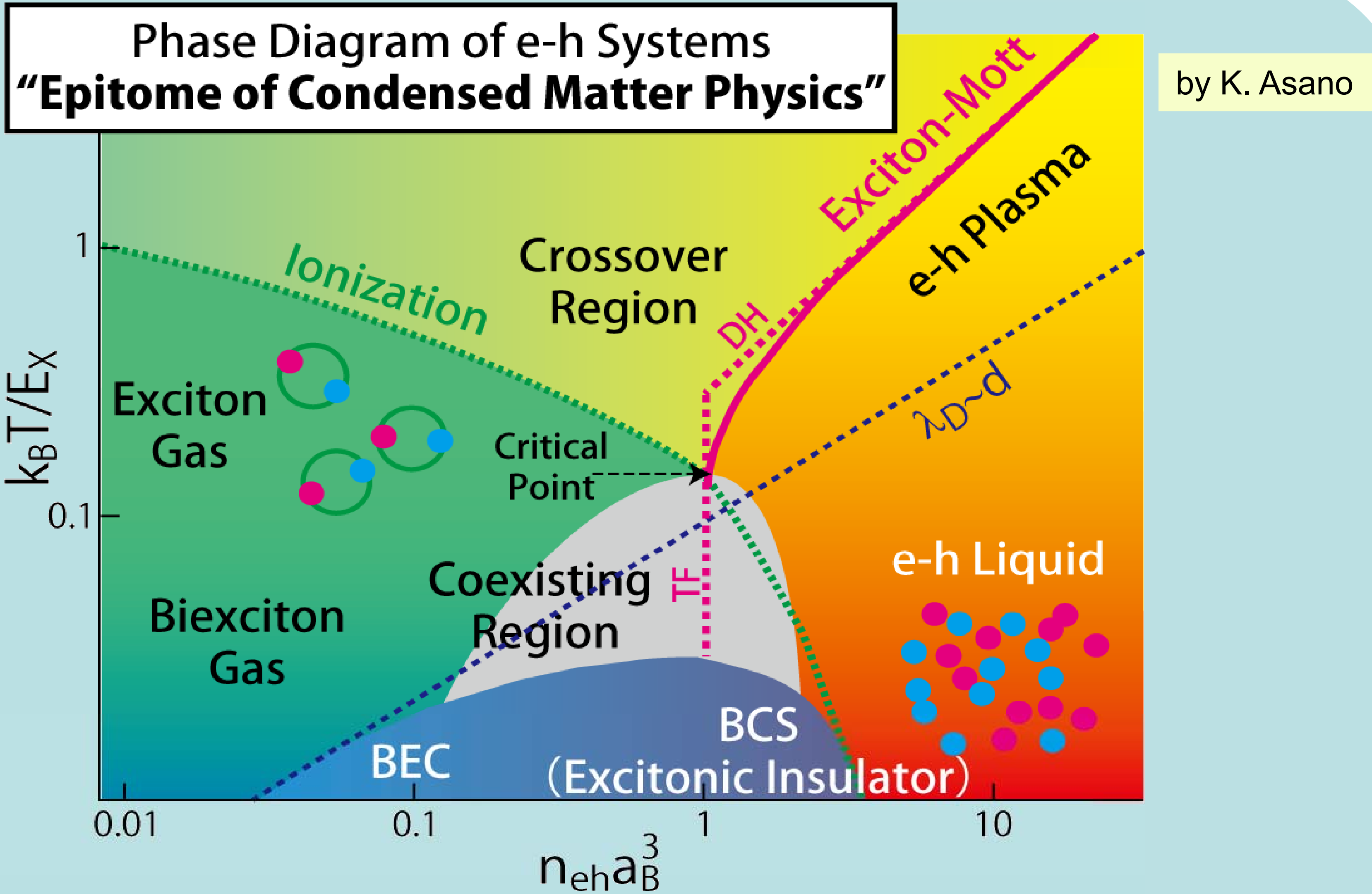
$T=10^{-6}$ K Cooled atoms and optical lattices:

fermi gas vs molecular Bose gas vs superfluid, supersolid, spinor BEC

Common features:

**Competing several order parameters**

- Strong-coupling regime - weak-coupling regime
- Pairing - antipairing, confinement - deconfinement
- Quantum coherence - classical coherence
- Finite temperature effects




Still just schematic !

Three boundaries:

✓Exciton Mott transition (EMT)
✓Quantum condensation (QC)
✓Thermal ionization

➡

✓Changing particle density
✓Unified treatment for EMT and QC
✓Continuum-space model
✓Optical responses