Photoinduced Phase Transitions
"DYCE" Optical Physics

Osaka PhoPs

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Contents:

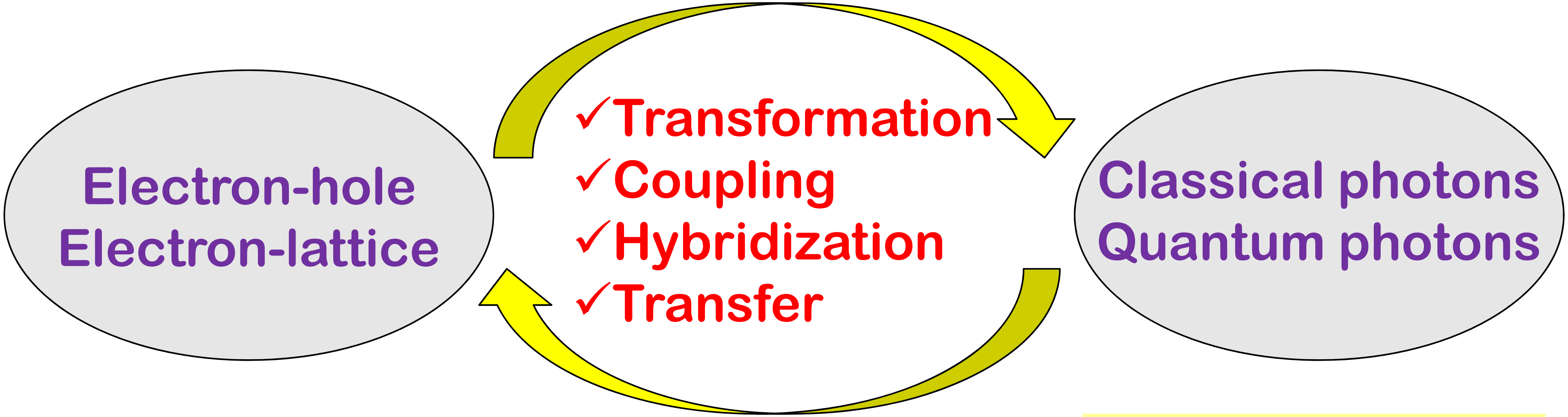
- Optical science using dynamically correlated electrons and holes (DYCE)
- Photoinduced **structural** phase transitions
  - Domino process
- Photoinduced **electronic** phase transitions
  - Exciton Mott transition
  - Quantum pair condensation: exciton BEC and e-h BCS condensation
- Cavity-polariton condensation and photon condensation (lasing)

DYCE
Optical Science of Dynamically Correlated Electrons

Ogawa Group in Dept. of Physics

17 July 2009@Building H, GSS, OU

From Matter to Light / From Light to Matter



Electron-hole
Electron-lattice

✓Transformation
✓Coupling
✓Hybridization
✓Transfer

Classical photons
Quantum photons

Cooperative phenomena in matter systems
“Photoinduced phase transitions (PIPT)”

Cooperative phenomena
in photon systems

Cooperative phenomena
in photon-matter coupled systems

Exciton Mott transition
e-h BCS

↕

Exciton BEC ⟷ **Cavity polariton BEC** ⟷ **Laser oscillation**

DYCE
Optical Science of Dynamically Correlated Electrons

PHOTOINDUCED PHASE TRANSITIONS (PIPT)

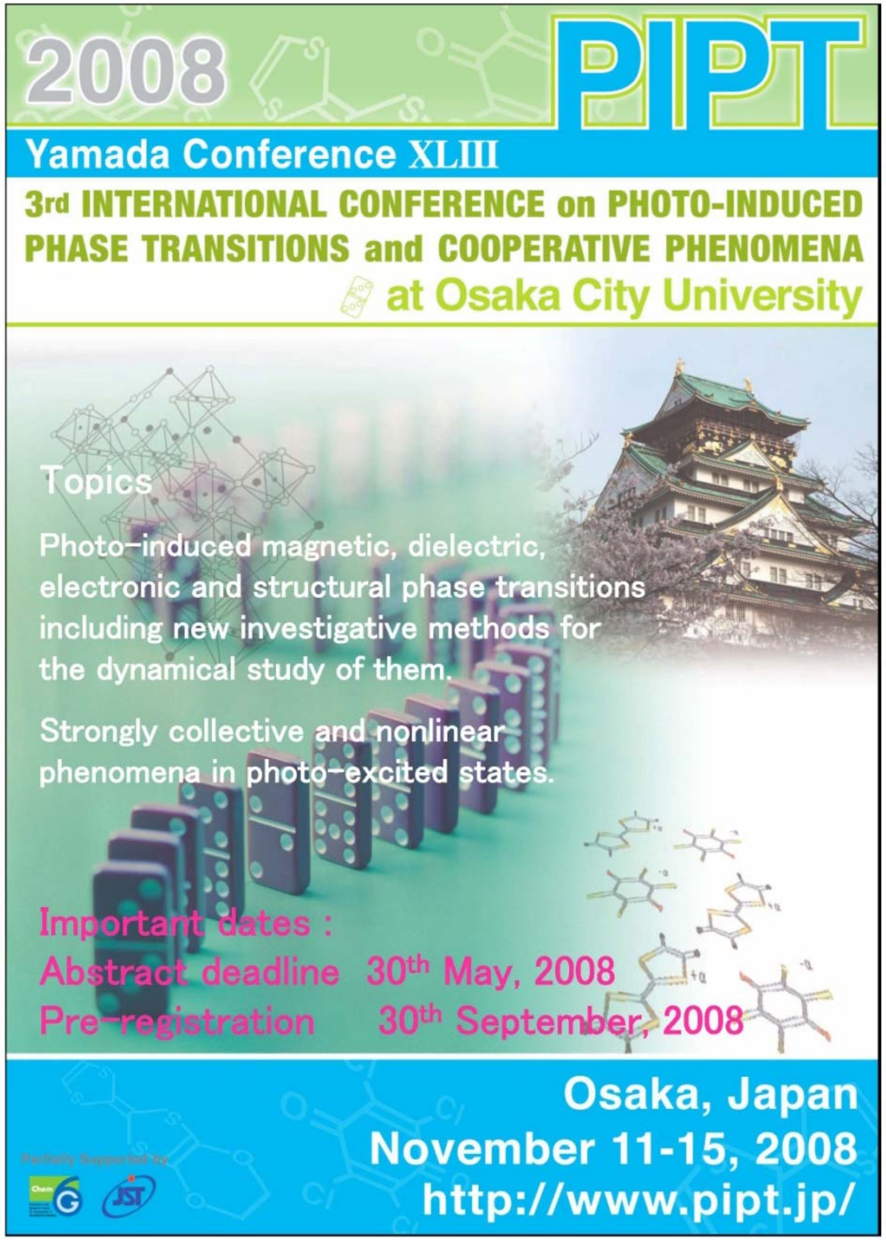
✓Light is not necessarily a probe in condensed-matter physics.

✓Creation and control of new states of matter by light irradiation:

- ➢<u>Electronic phase transitions</u>
- ➢Structural phase transitions

✓Two types of PIPT process:

- ➢<u>“Phase” transitions in photoexcited states</u>
- ➢Phase transitions via photoexcited states



2008 PIPT

Yamada Conference XLIII

3rd INTERNATIONAL CONFERENCE on PHOTO-INDUCED PHASE TRANSITIONS and COOPERATIVE PHENOMENA

at Osaka City University

Topics

Photo-induced magnetic, dielectric, electronic and structural phase transitions including new investigative methods for the dynamical study of them.

Strongly collective and nonlinear phenomena in photo-excited states.

Important dates :

Abstract deadline 30th May, 2008

Pre-registration 30th September, 2008

Osaka, Japan

November 11-15, 2008

http://www.pipt.jp/

Partially Supported by:

QUANTUM COOPERATIVE PHENOMENA

By changing temperature, pressure, particle density, interaction strength, …

Phase transition

ex)：gas-liquid-solid, metal-insulator, localization-delocalization, …

In e-h systems: Exciton Mott transition (e-h plasma – exciton gas)

Quantum condensation (“Macroscopic (long-range) quantum order”)

ex): Bose-Einstein condensation (BEC), superconductivity, superfluidity, …

In e-h systems: Exciton BEC, e-h BCS-like state, BEC-BCS crossover

One body⇔Many body

Importance of interparticle interaction: dynamical screening, X-X interaction

N-particle system ≠ 1-particle system × N → “nonlinearity”, “cooperation”

Classical⇔Quantum

Quantum statistics: Pauli exclusion, BEC（“interactionless phase transition”）

Quantum fluctuation (particle-wave duality): material coherence vs optical coherence, …

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    - Domino mechanism

- Photoinduced electronic phase transitions
    - Exciton Mott transition
    - Quantum pair condensation

- Cavity-polariton condensation

OUTLINE OF OUR STUDY

KEYWORDS:

Global (cooperative, nonlocal) phase changes
Photoinduced phenomena
Intersite (intermolecule) couplings

EXPERIMENTS:

Photoinduced structural transitions in PDA crystals
Photoinduced HS/LS transitions in spin-crossover compounds
Photoinduced ferromagnetism in (In,Mn)As/GaSb

MODEL SYSTEM & PHENOMENA:

A molecule (site) has two locally-stable structures: A and B
One-dimensional stacking
Intersite (intermolecule) elastic interaction
One-site excitation by irradiation (photoinduced nucleation)

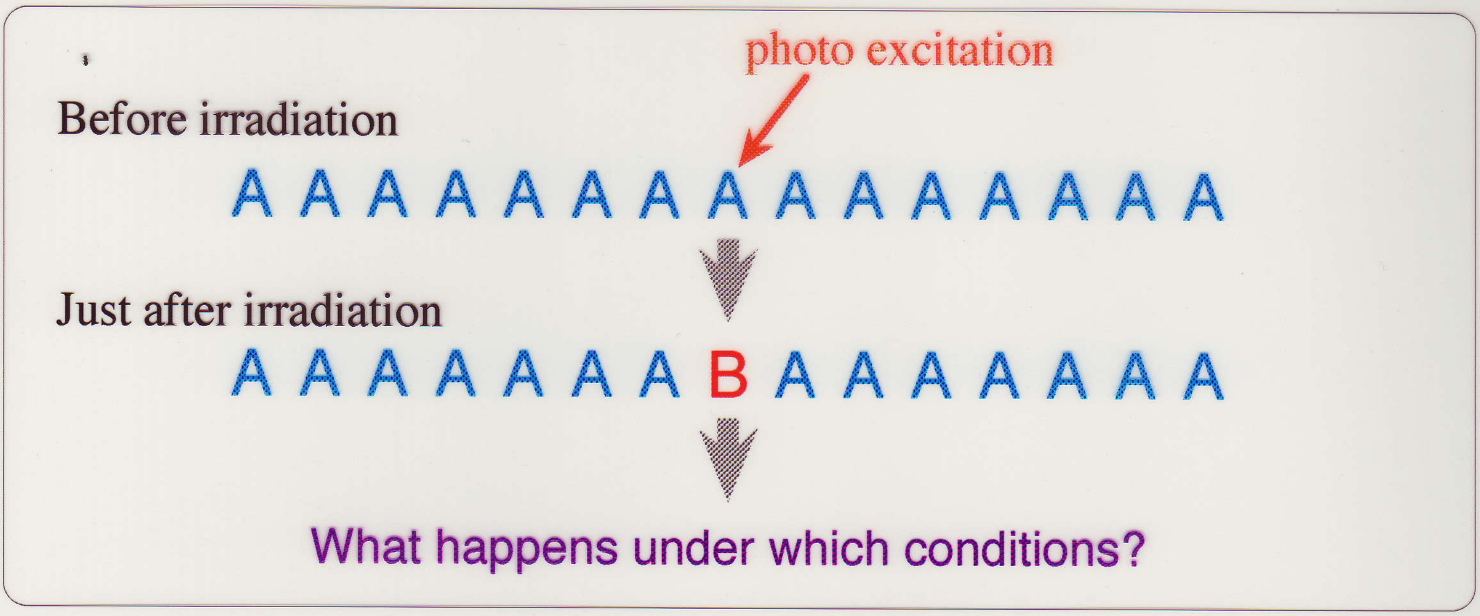


photo excitation

Before irradiation

A A A A A A A A A A A A A A A

Just after irradiation

A A A A A A A B A A A A A A A

What happens under which conditions?

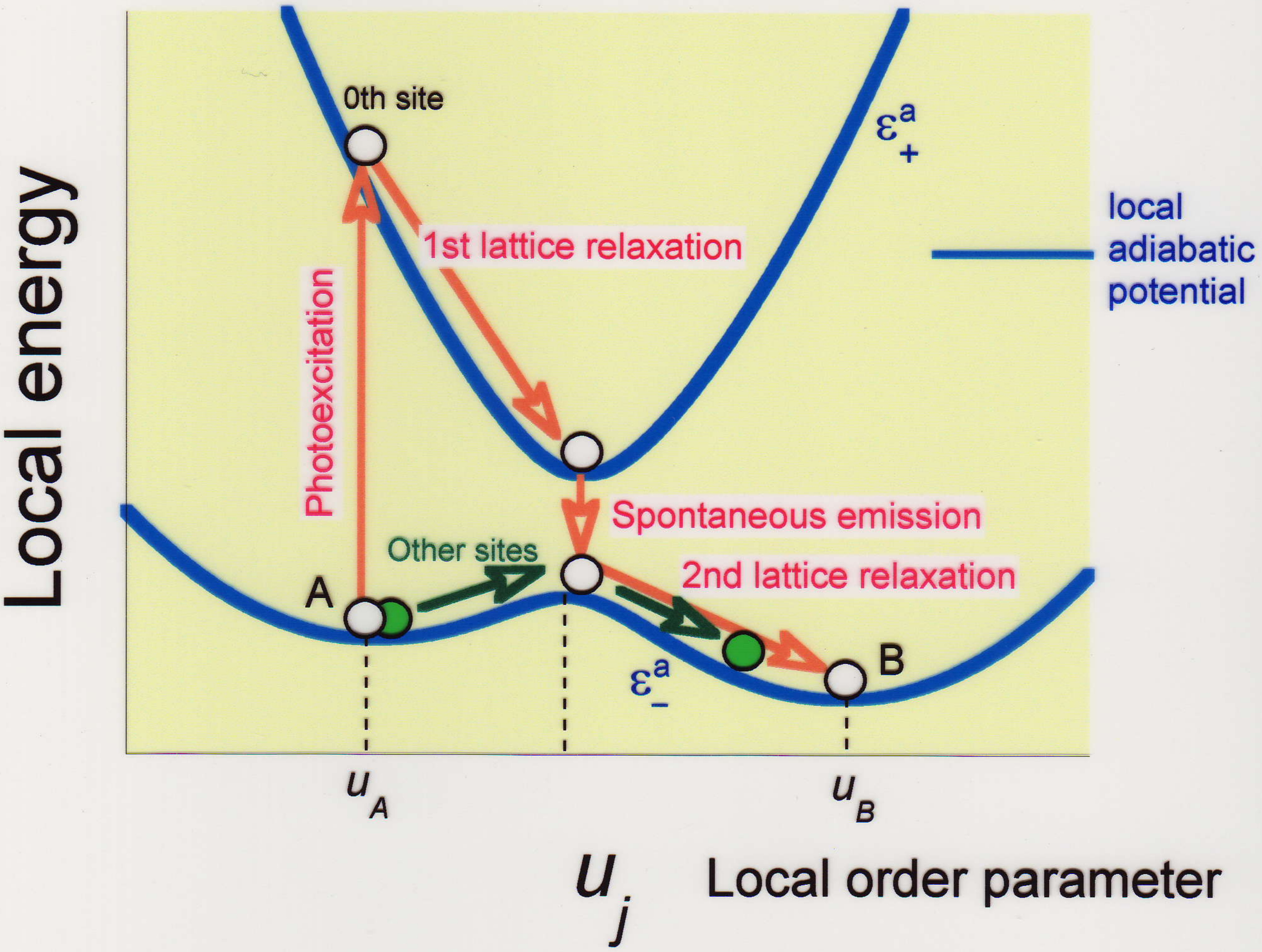
OUR GOAL:

To clarify spatiotemporal dynamics of photoinduced nucleation
To clarify the role of intersite interactions
To distinguish between adiabatic and diabatic regimes
To compare nucleation picture with mean-field description

Deterministic Domino mechanism

starting at the spontaneous emission

The strong friction case



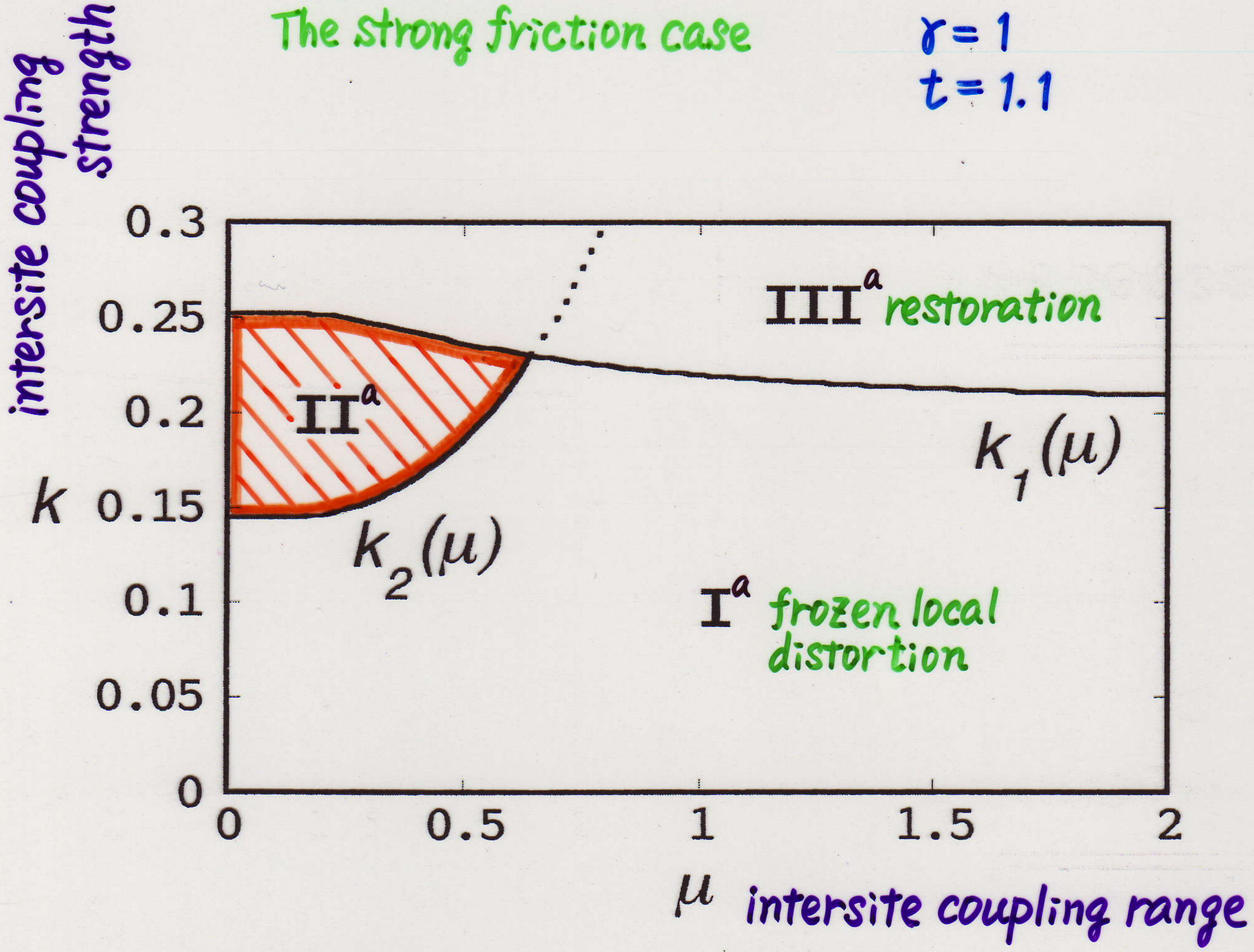
Phase Diagram on (μ, k) plane

Adiabatic limit

Single-site excitation

The strong friction case

$\varepsilon = -0.5$
$\gamma = 1$
$t = 1.1$



Only in Phase II^a, global structural transition is induced by single-site stimulation.

- Short-range intersite coupling
- Intermediate coupling strength

→ Deterministic (periodic) Domino mechanism

only one spontaneous emission

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Electron-hole system as an excited electronic state

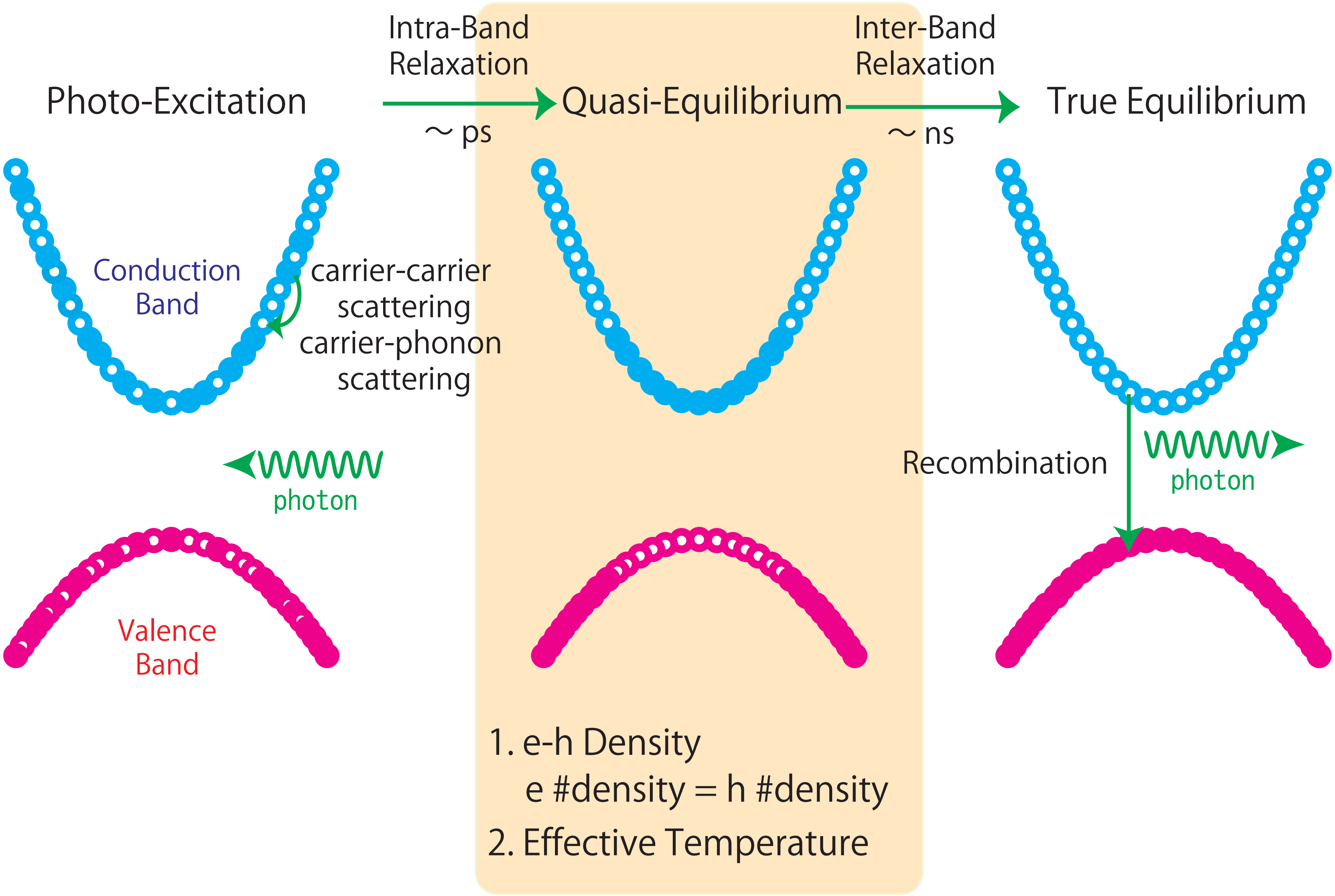


Photo-Excitation

Intra-Band
Relaxation

～ ps

Quasi-Equilibrium

Inter-Band
Relaxation

～ ns

True Equilibrium

Conduction
Band

carrier-carrier
scattering
carrier-phonon
scattering

photon

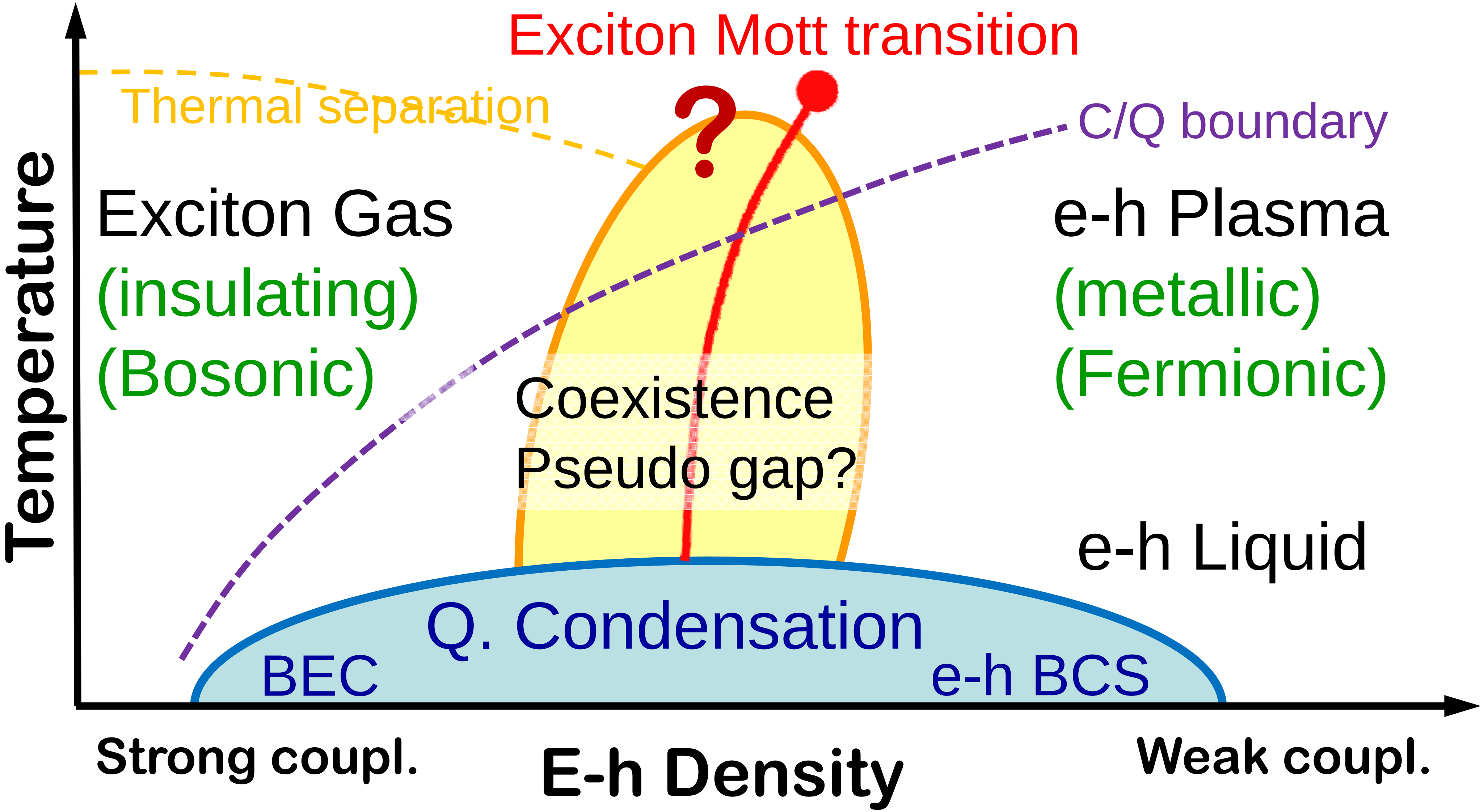
Valence
Band

Recombination

photon

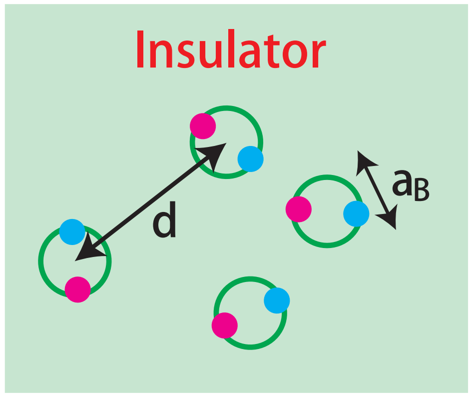
1. e-h Density
   e #density = h #density
2. Effective Temperature

Electron-hole systems

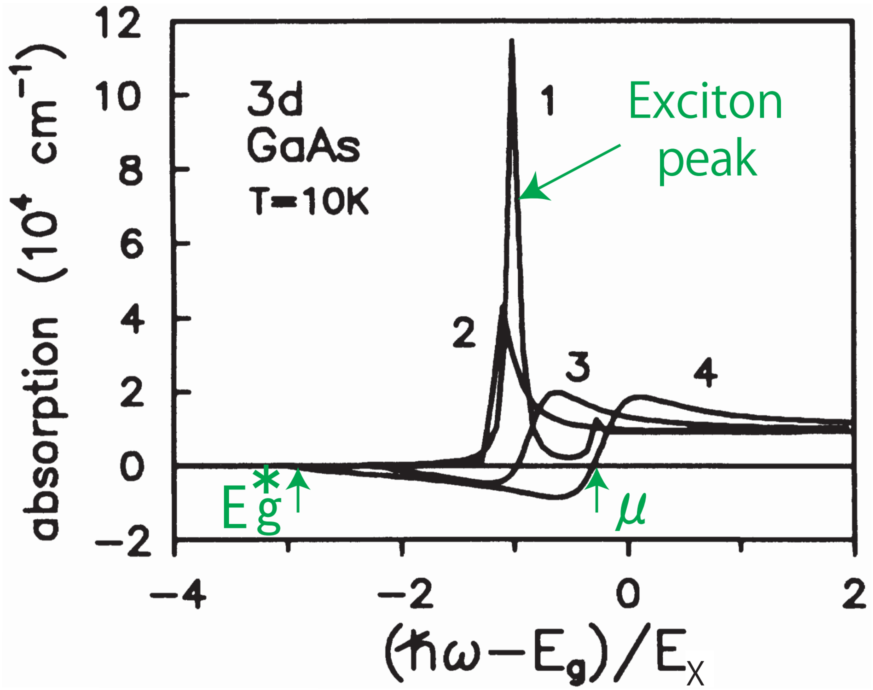


Relation between exciton Mott transition & optical gain/absorption

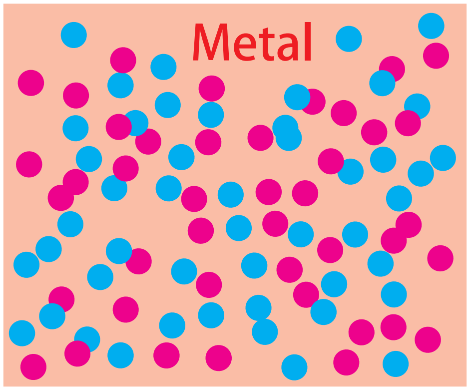
Low density ← ? → High density



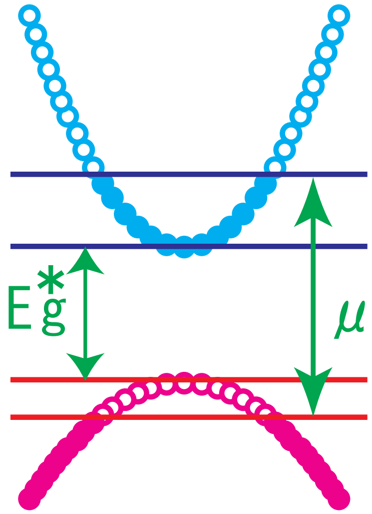
Exciton gas



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

The e-h (two-band) Hubbard Model

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

⇒ 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)

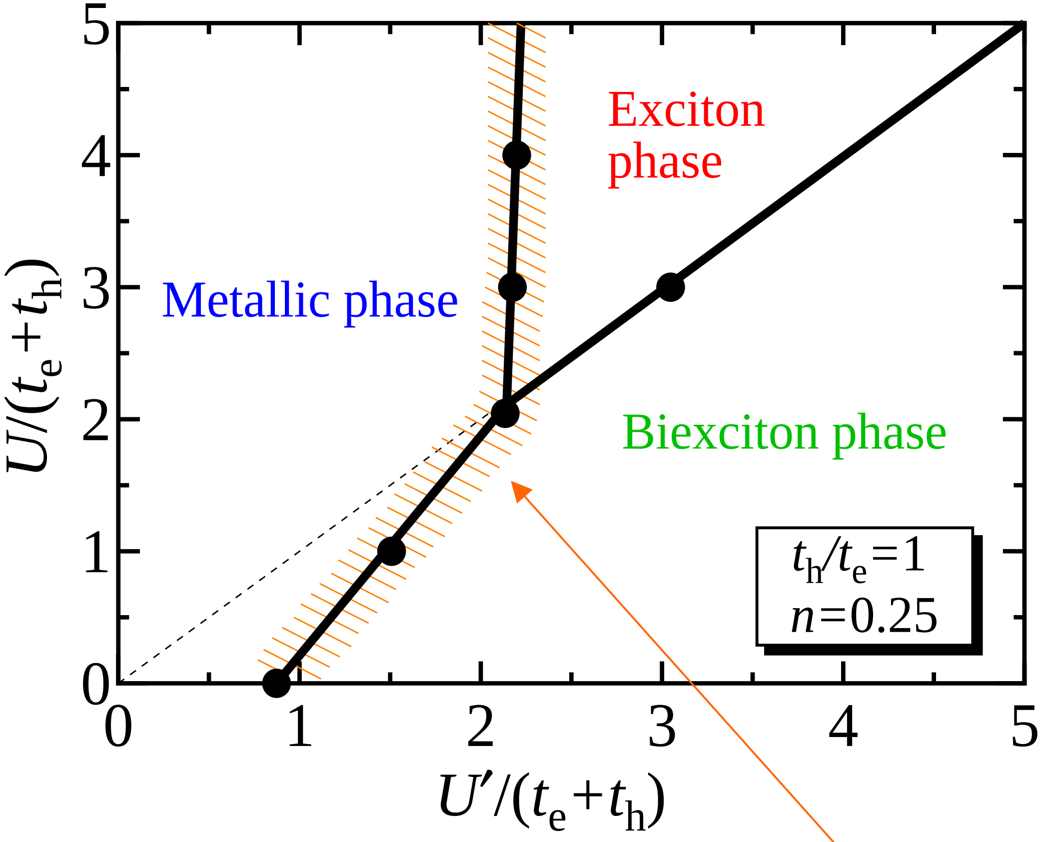
- different masses of electron and hole
- screened on-site interactions as parameters independent of density
- Frenkel-type excitons without co-transfer
- modern theoretical tools applicable

cf.) the continuum-space model (in the nearly-free electron picture)

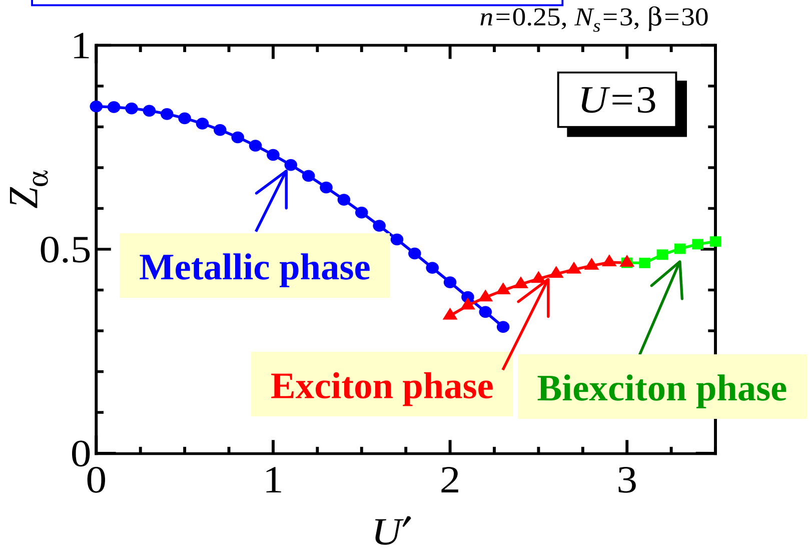
- effective-mass and envelope fn. approximations
- (long-range) Coulomb interactions dependent on density
- Wannier-type excitons with finite bandwidth

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

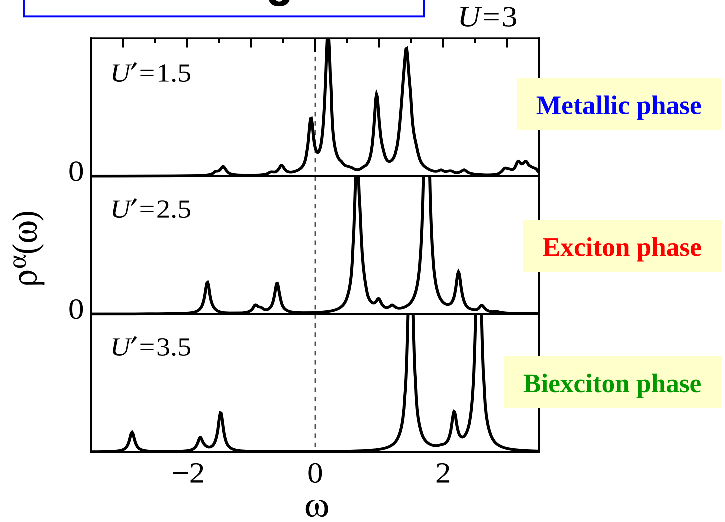
n=0.25 *t*$_h$/*t*$_e$ =1



Quasiparticle weights



Interacting DOS

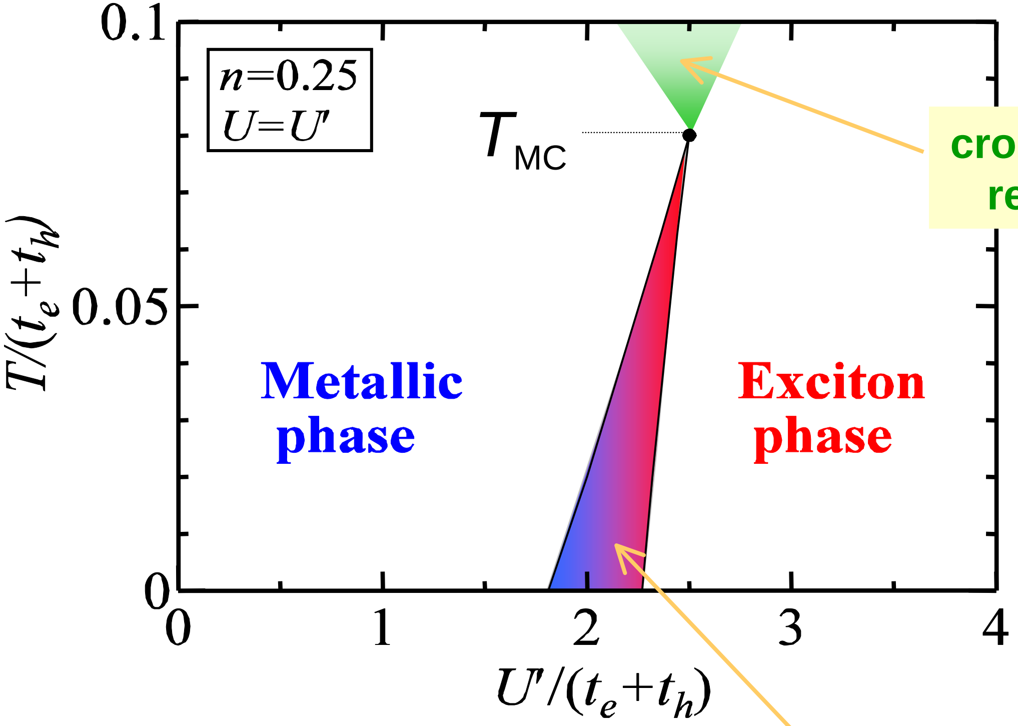


Metal↔ Exciton / Biexciton phase : 1st-order transition

exciton Mott transition

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

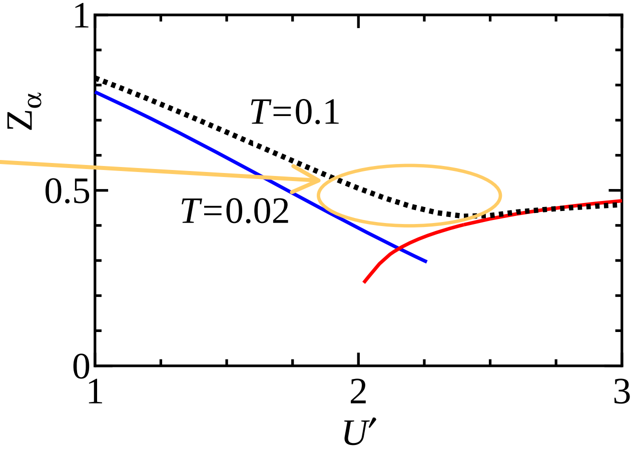
$U=U'$ $n=0.25$ $t_h/t_e=1$



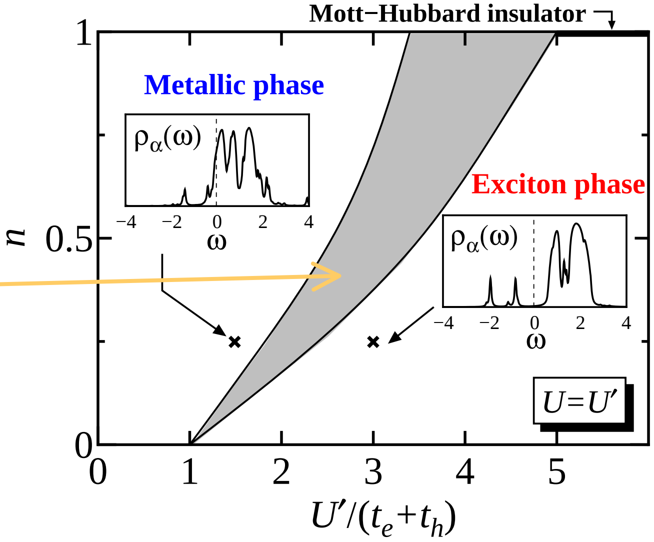
crossover region

coexistent region

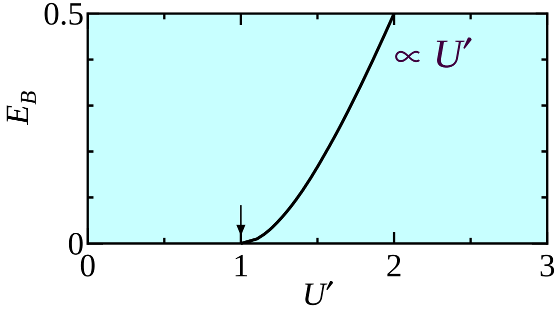
quasiparticle weights



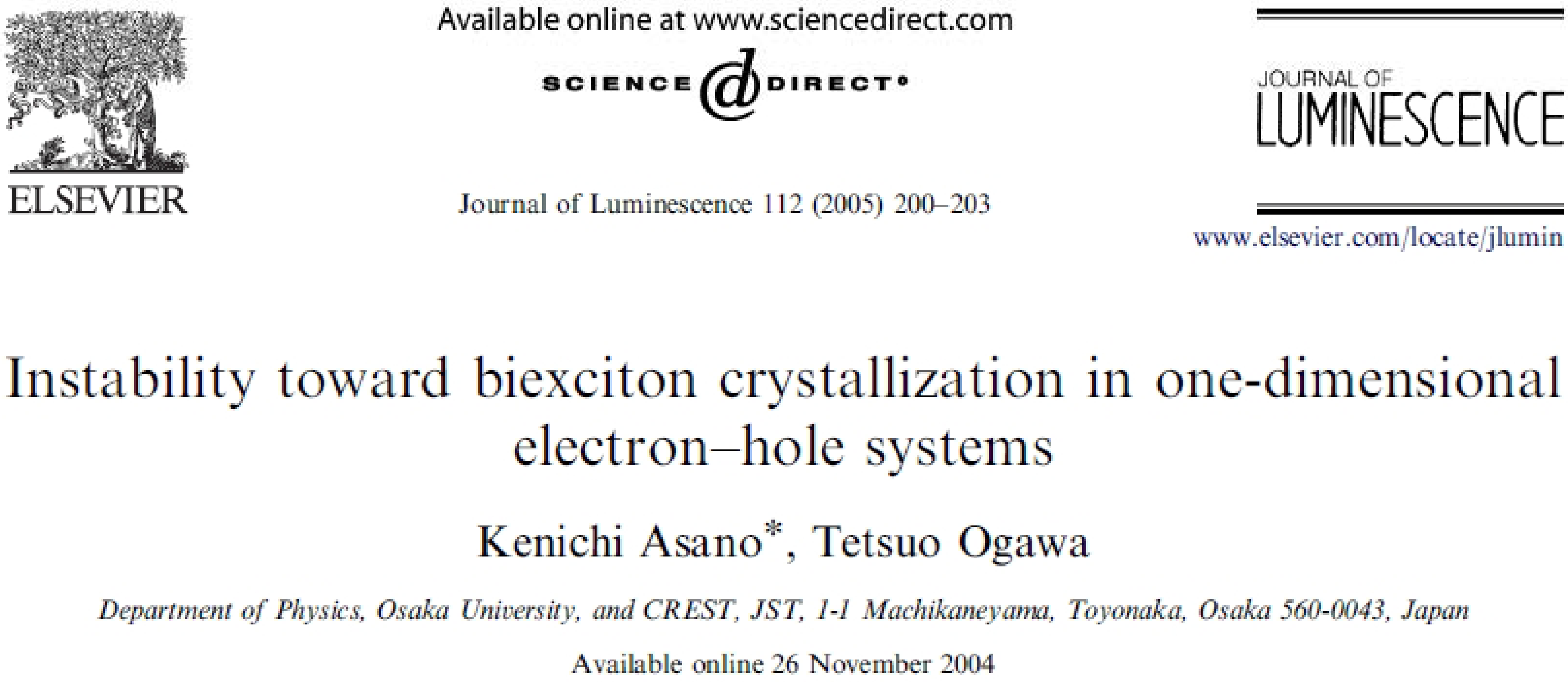
U'-n phase diagram at T=0



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



BIEXCITON CRYSTAL in 1D



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Journal of Luminescence 112 (2005) 200–203

JOURNAL OF LUMINESCENCE

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Instability toward biexciton crystallization in one-dimensional electron–hole systems

Kenichi Asano*, Tetsuo Ogawa

Department of Physics, Osaka University, and CREST, JST, 1-1 Machikaneyama, Toyonaka, Osaka 560-0043, Japan

Available online 26 November 2004

- ➢High-density electron-hole system in 1D
- ➢Bosonization
- ➢Forward and backward scatterings
- ➢Long-range Coulomb interaction

Character of the Ground State

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

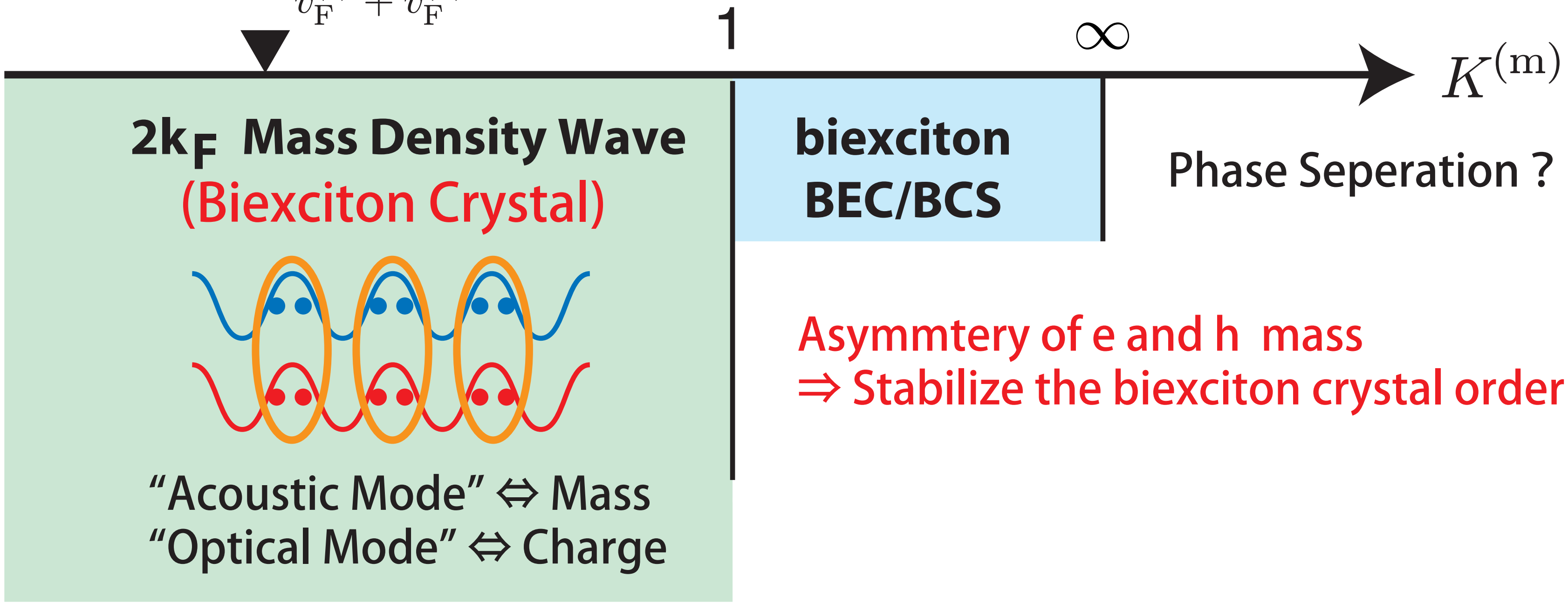
$$\mathcal{H}_\rho^{(\mathrm{m})} = \frac{v^{(\mathrm{m})}}{2\pi}\int dx \left[K^{(\mathrm{m})}\left(\partial_x\Theta_\rho^{(\mathrm{m})}\right)^2 + \frac{1}{K^{(\mathrm{m})}}\left(\partial_x\Phi_\rho^{(\mathrm{m})}\right)^2\right]$$

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

⇒ 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)



2k$_\mathrm{F}$ Mass Density Wave

(Biexciton Crystal)

"Acoustic Mode" ⇔ Mass
"Optical Mode" ⇔ Charge

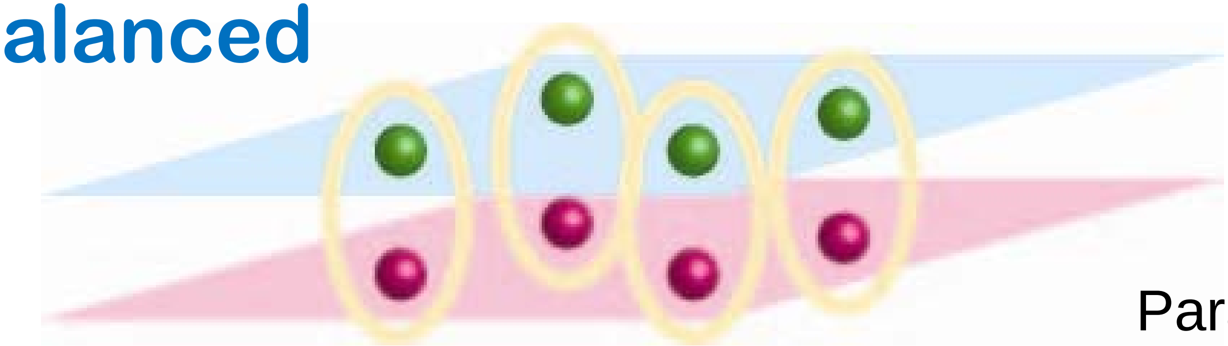
biexciton
BEC/BCS

Phase Seperation ?

Asymmtery of e and h mass
⇒ Stabilize the biexciton crystal order

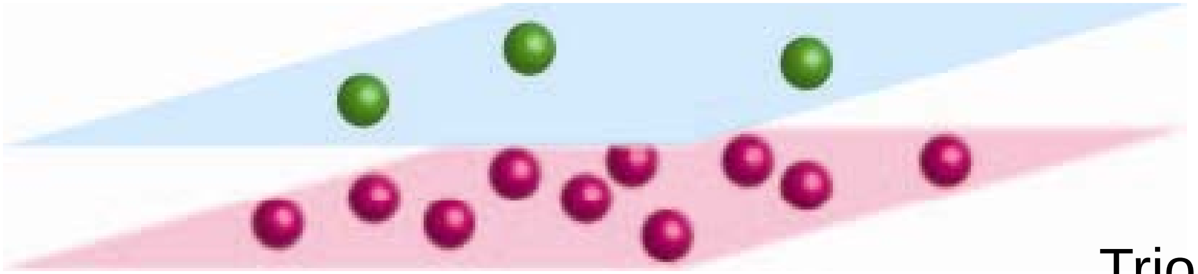
Double layer e-h systems

Charge balanced



Parallel dipoles
Repulsive?
Exciton BEC?

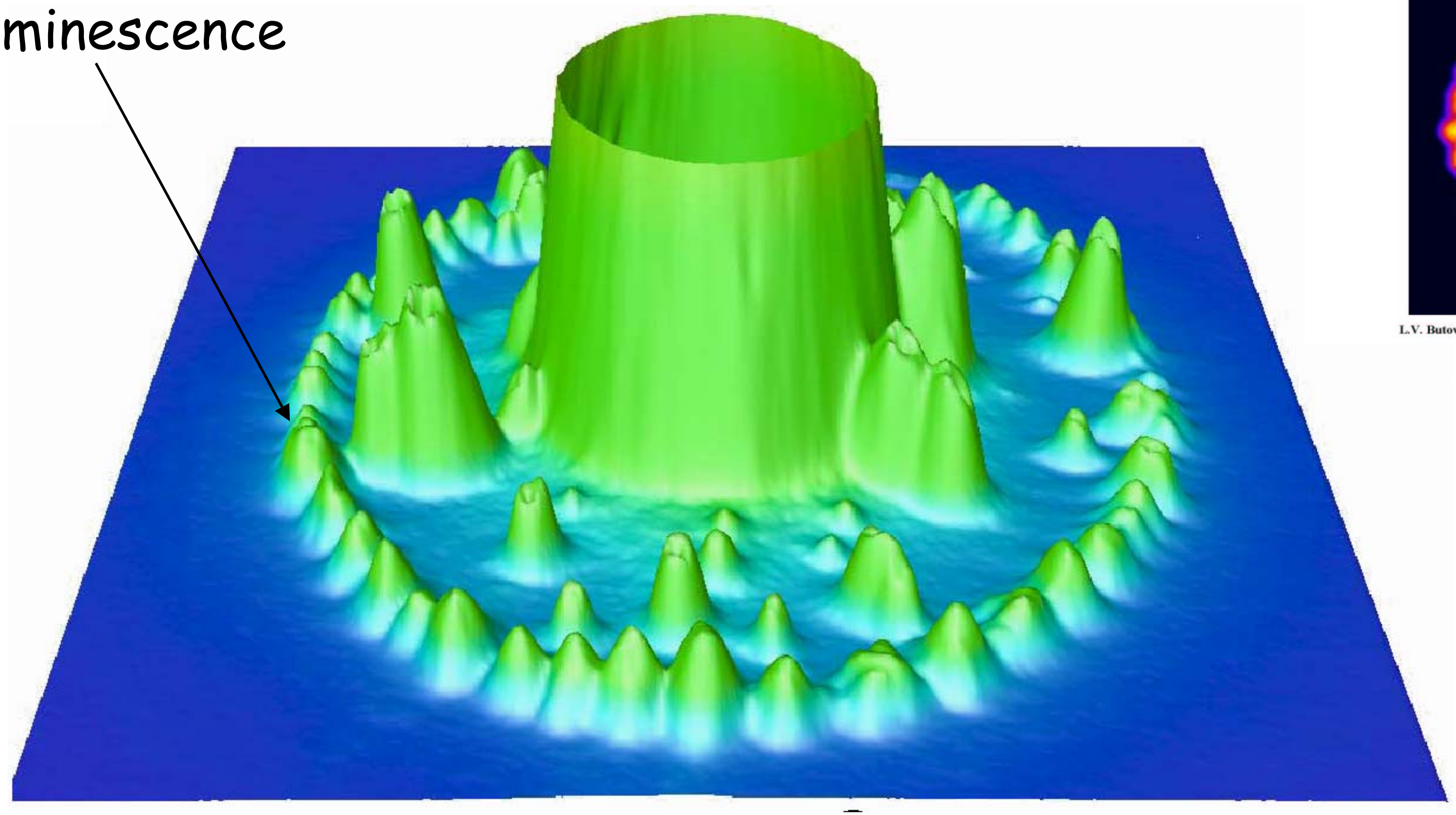
Charge imbalanced



Trions
FFLO?

Exciton luminescence from quantum wells: central spot and fragmented ring

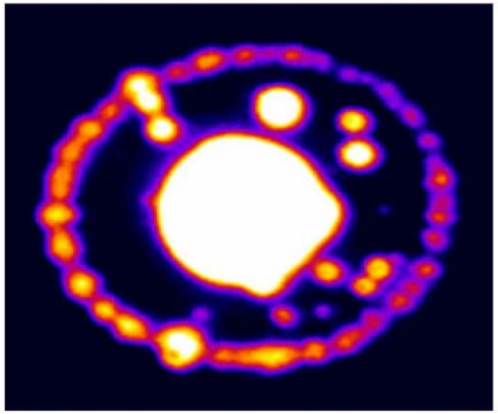
fragments of luminescence



260 μm

T=1.8 K
Pex=390 μW

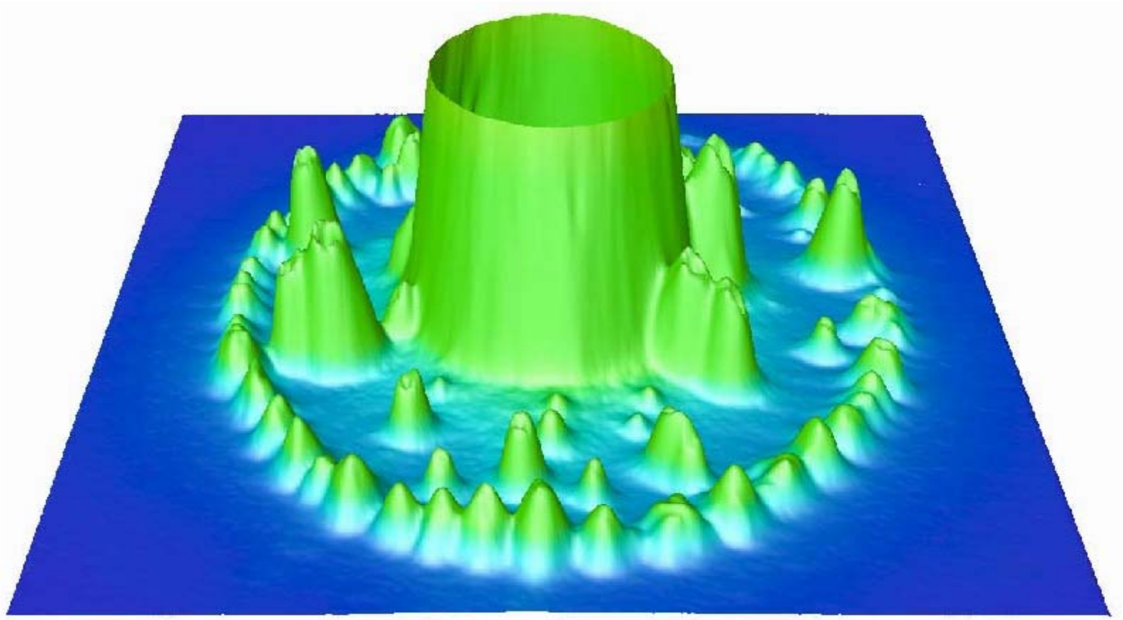
2D image of indirect exciton PL vs P_{ex}

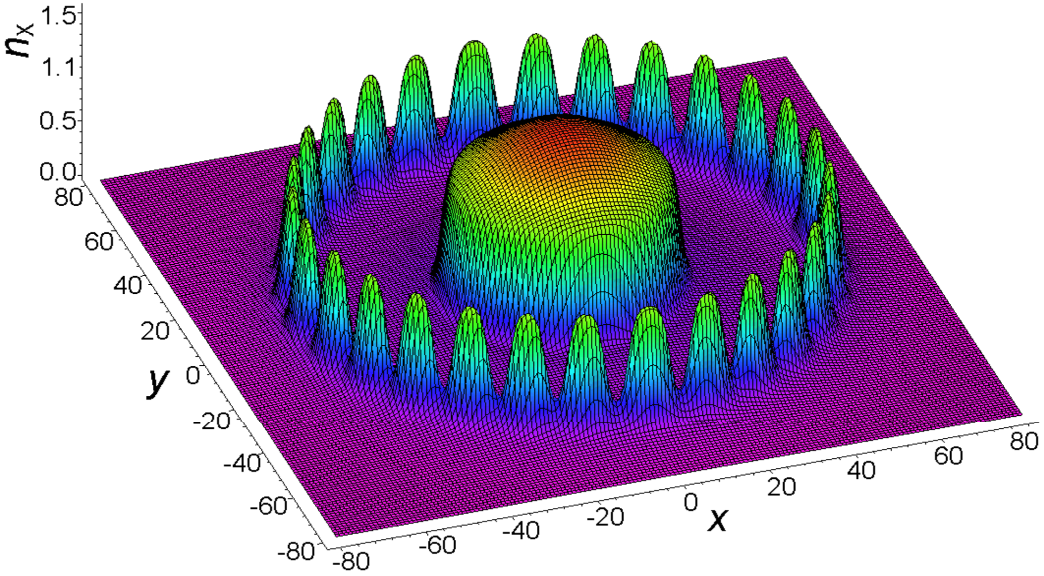


L.V. Butov, A.C. Gossard, and D.S. Chemla, cond-mat/0204482 [Nature 418, 751 (2002)]

View from above

L. Butov et al.
Nature 418 (2002),
PRL 92 (2004)





Exciton luminescence **Butov et al.,**

Nature 418, 751 (2002)

Chernyuk A.A., Sugakov V.I.:

PRB 74, 085303 (2006),

PHYSICAL REVIEW B **81**, 115320 (2010)

Exciton front propagation in photoexcited GaAs quantum wells

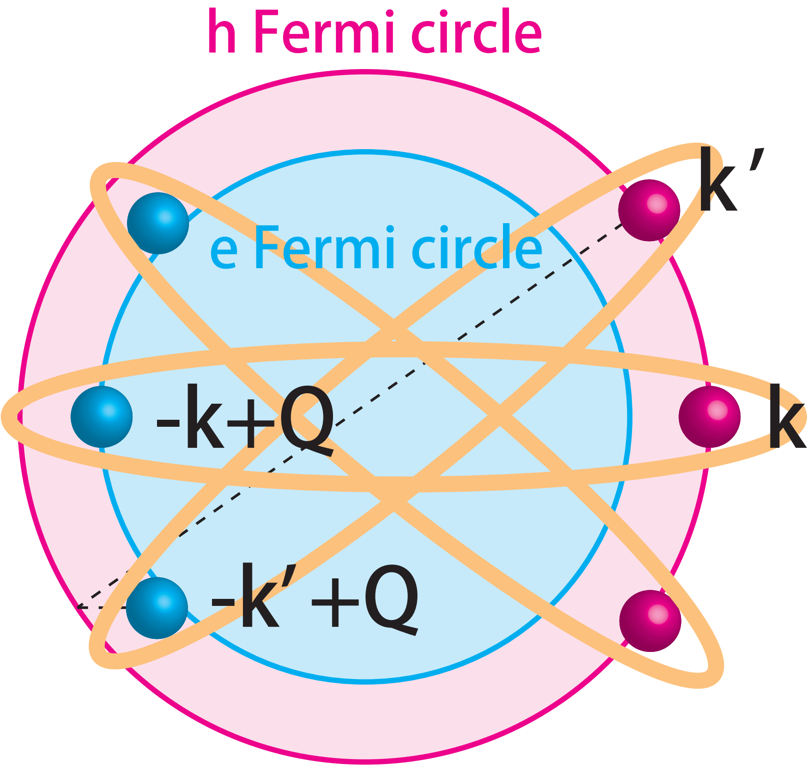
Sen Yang,[1] L. V. Butov,[1] L. S. Levitov,[2] B. D. Simons,[3] and A. C. Gossard[4]
[1]*Department of Physics, University of California at San Diego, La Jolla, California 92093-0319, USA*
[2]*Department of Physics, Massachusetts Institute of Technology, Cambridge, Massachusetts 02139, USA*
[3]*Cavendish Laboratory, Madingley Road, Cambridge CB3 OHE, United Kingdom*
[4]*Materials Department, University of California at Santa Barbara, Santa Barbara, California 93106-5050, USA*
(Received 20 August 2009; revised manuscript received 10 February 2010; published 16 March 2010)

We report on the study of spatiotemporal self-organization of carriers in photoexcited GaAs quantum wells. Propagating interfaces between electron-rich and hole-rich regions are seen as expanding and collapsing exciton rings in exciton emission patterns. The interfaces preserve their integrity during expansion, remaining as sharp as in the steady state, which indicates that the dynamics is controlled by carrier transport. The front propagation velocity is measured and compared to theoretical model. The measurements of expanding and collapsing exciton rings afford a contactless method for probing the electron and hole transport.

DOI: 10.1103/PhysRevB.81.115320 PACS number(s): 78.67.De, 71.35.−y, 72.20.−i, 78.55.Cr

Quantum Condensations in Imbalanced e-h Systems

Fulde-Ferrell Phase



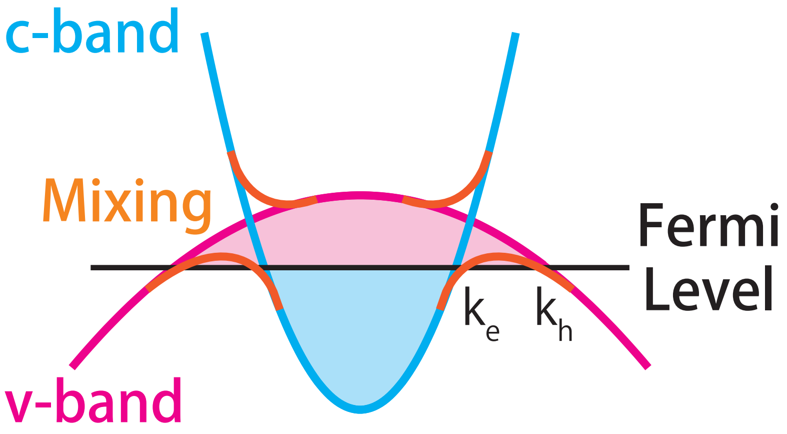
e-h pair with CM momentum Q

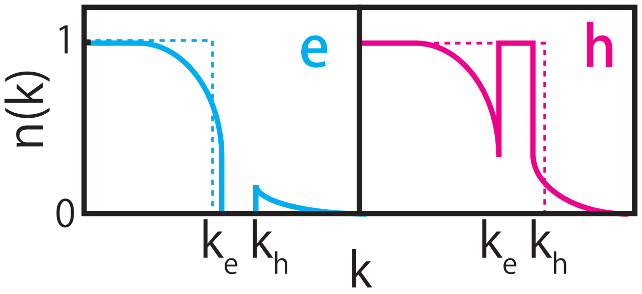
Fulde and Ferrell: PR**135**, 705(1964).

c.f. Inhomogeneous solution:
A. I. Larkin and Y. N. Ovchinnikov,
Sov. Phys. JETP 20, 762 (1965).

Sarma Phase

(Breached pair phase)





Condensation of e-h pair with Q=0
+ Normal hole liquid

Sarma: J. Phys. Chem. Sol. **24**, 1029 (1963).
W. V. Liu and F. Wilczek: PRL90, 047002 (2003).

Phase Diagram at Zero Temperature

Parameters

Mass ratio

$$\frac{m^{(h)}}{m^{(e)}} = 4.3$$

Interlayer distance

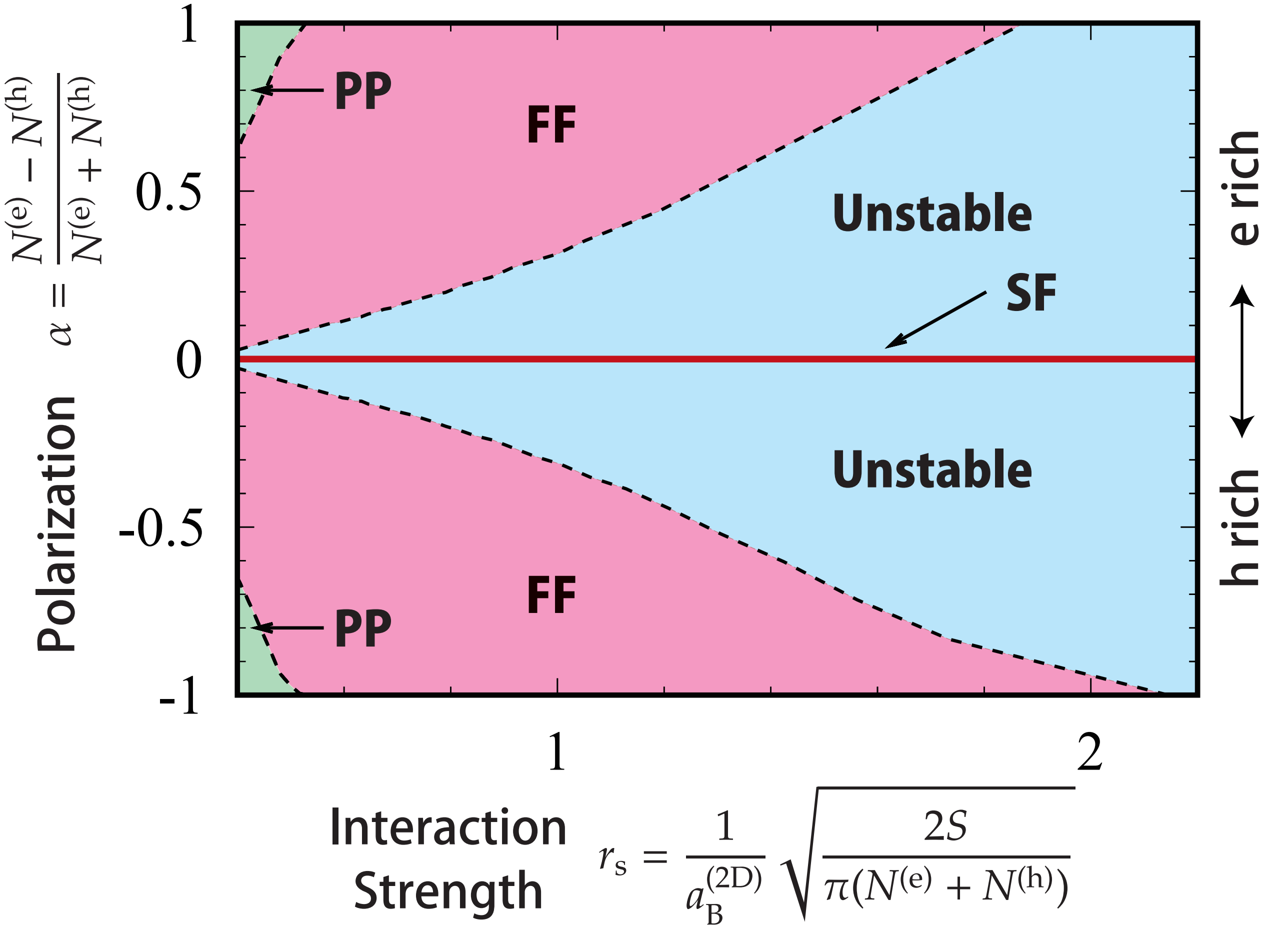
$$d = 2a_B^{(2D)} = \frac{\epsilon}{e^2 m_r}$$

SF: superfluid phase (excitonic insulator)

FF: Fulde-Ferrell phase

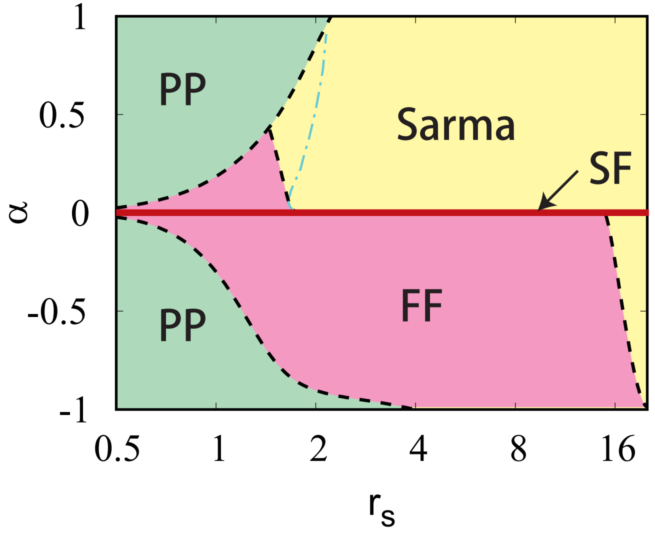
PP: partially polarized normal phase

Unstable: no uniform solution



c.f. Previous calculation

Instability of Sarma phase toward FF phase



Pieri et al. PRB75,113301 (2007).

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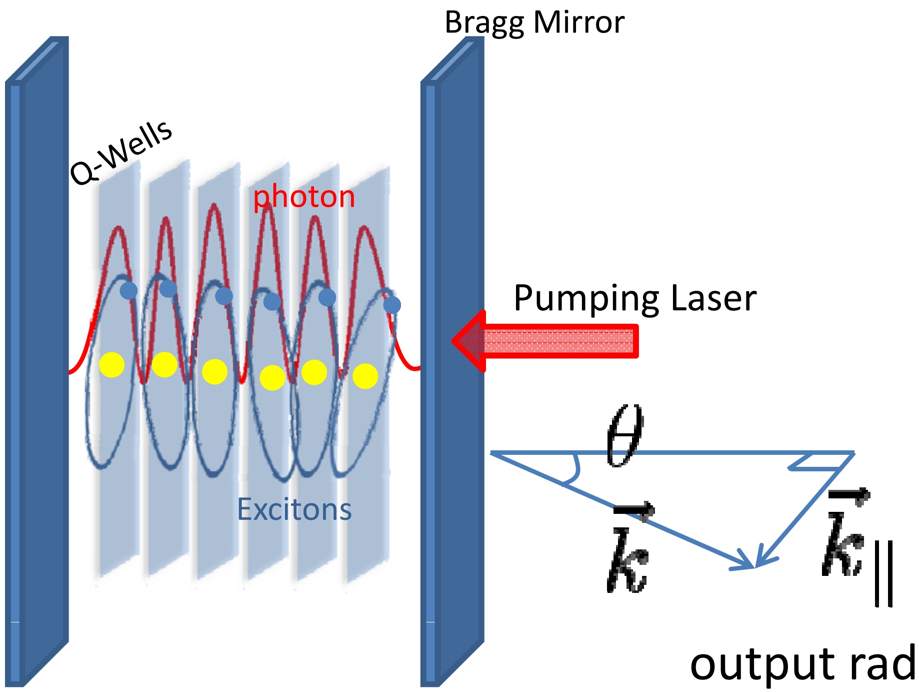
Polariton BEC in a microcavity

N-layers of semiconductor quantum wells in distributed Bragg reflectors

- single photon mode
- strong coupling with excitons $g \propto \sqrt{N}$



output radiation → detection

Observation of the BEC

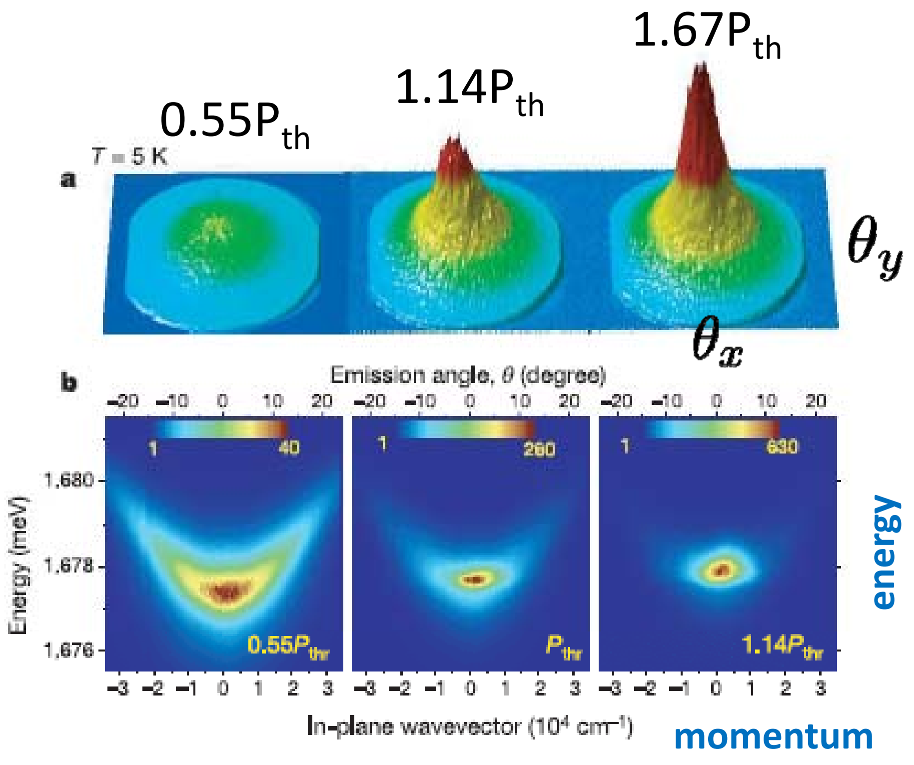
J. Kasprzak et al, Nature 443, 409 (2006).

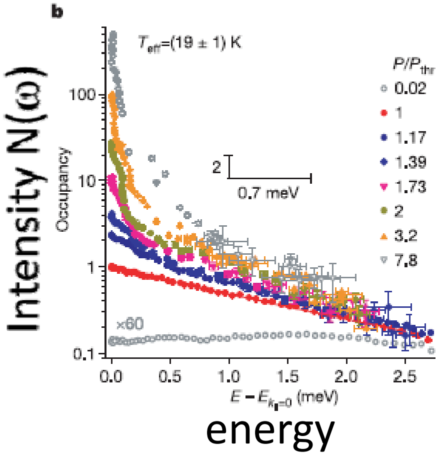
Condition: non-resonant pumping + 16 CdTe Q-wells
Detection: angle resolved emission spectrum (far-field)

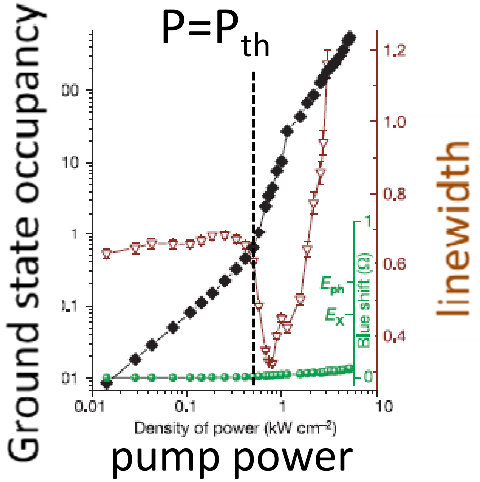
Bose narrowing in the momentum and energy space

Angular distribution

Emission Spectrum

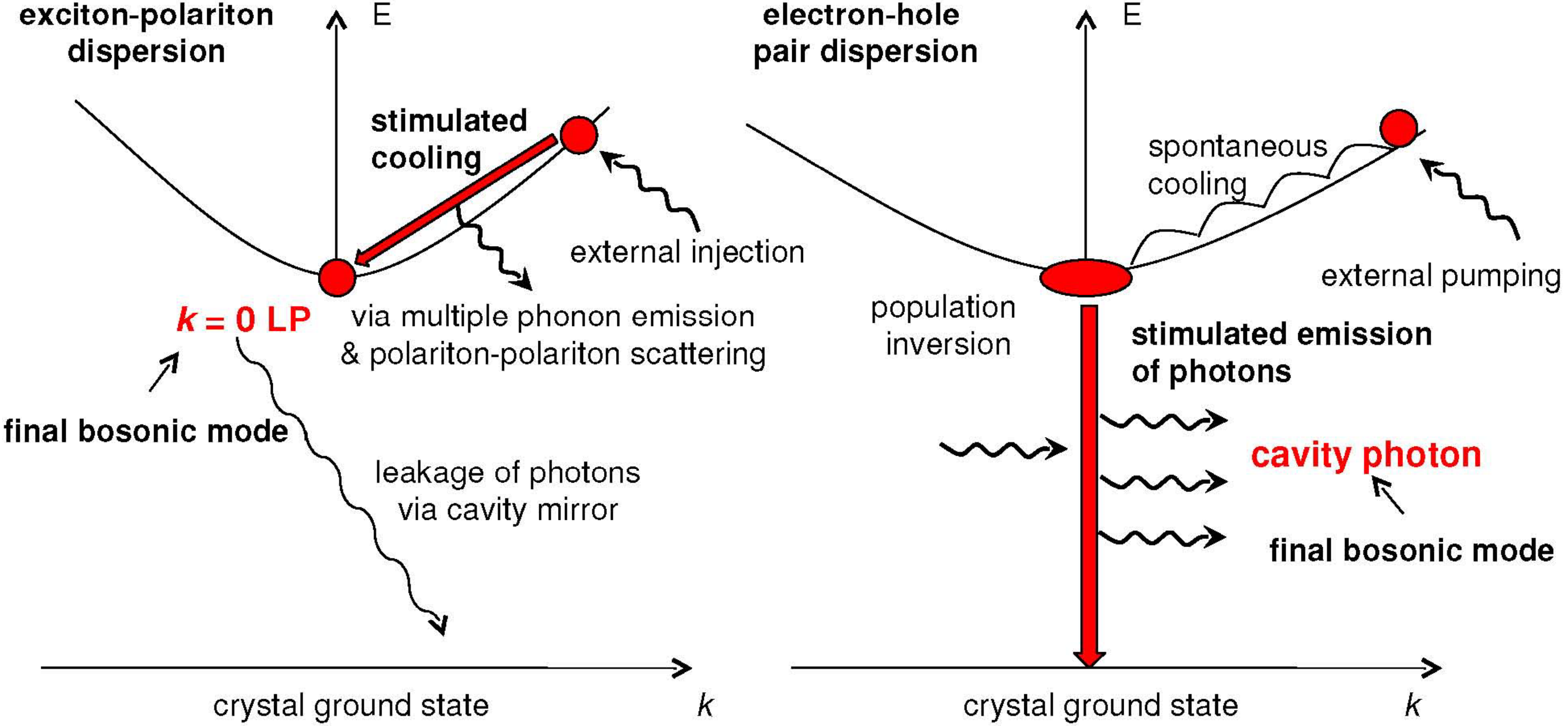






Polariton BEC is similar to Bose-Einstein Condensation of atomic gases in a thermal equilibrium!

Exciton-Polariton Condensation vs. Photon Laser



exciton-polariton dispersion

E

stimulated cooling

external injection

via multiple phonon emission & polariton-polariton scattering

k = 0 LP

final bosonic mode

leakage of photons via cavity mirror

crystal ground state

k

electron-hole pair dispersion

E

spontaneous cooling

external pumping

population inversion

stimulated emission of photons

cavity photon

final bosonic mode

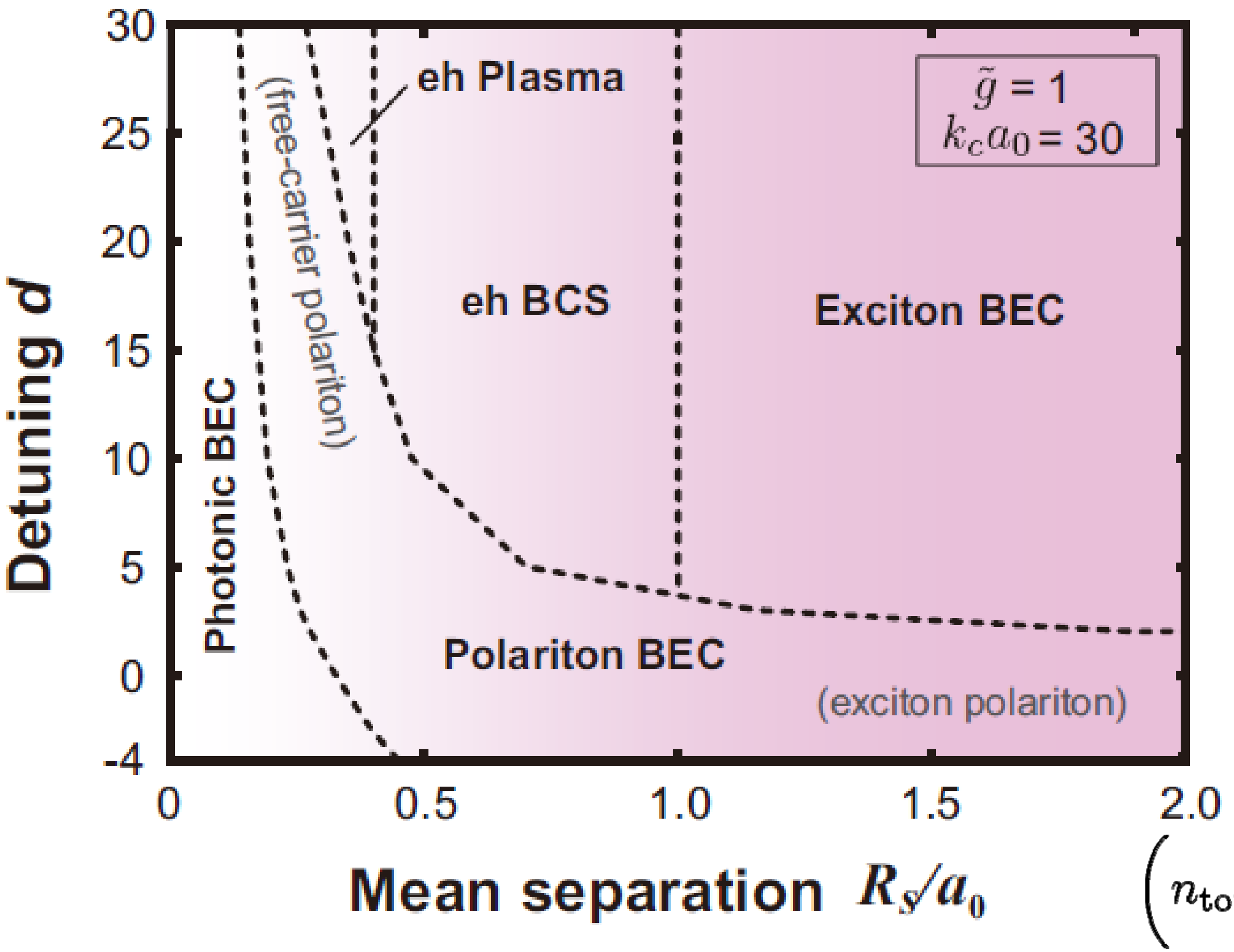
crystal ground state

k

Exciton-Polariton Condensation

Photon Laser

Phase diagram



$$\left(n_{\text{tot}} = n_{\text{ph}} + \frac{n_{\text{e}} + n_{\text{h}}}{2} \right)$$

Different ground states are expected to occur depending on the detuning and the pump power (total excitation density n_{tot}).

Composite many-body systems

exciton eh
charged exciton eeh
biexciton eehh
polariton eh+b

Cooper pair ee
positronium eē
metal hydrogen HH
Bose atom mol. gas AA

meson qq̄
baryon qqq
tetraquark qqq̄q̄
pentaquark qqqqq̄
hexaquark qqqqqq

diquark qq
anti-baryon q̄q̄q̄

Even-odd mixture in imbalanced systems

- Exciton-trion mixture
- Meson-baryon mixture
- Atom-molecule mixture

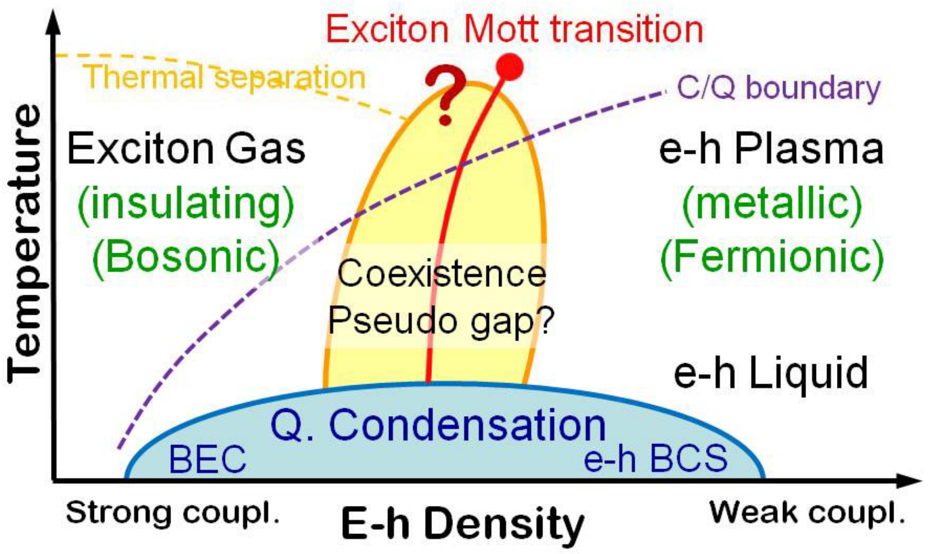
Even-even mixture in balanced systems

- “Unitary limit” of BEC-BCS crossover
- Exciton-biexciton mixture
- Meson-tetraquark mixture

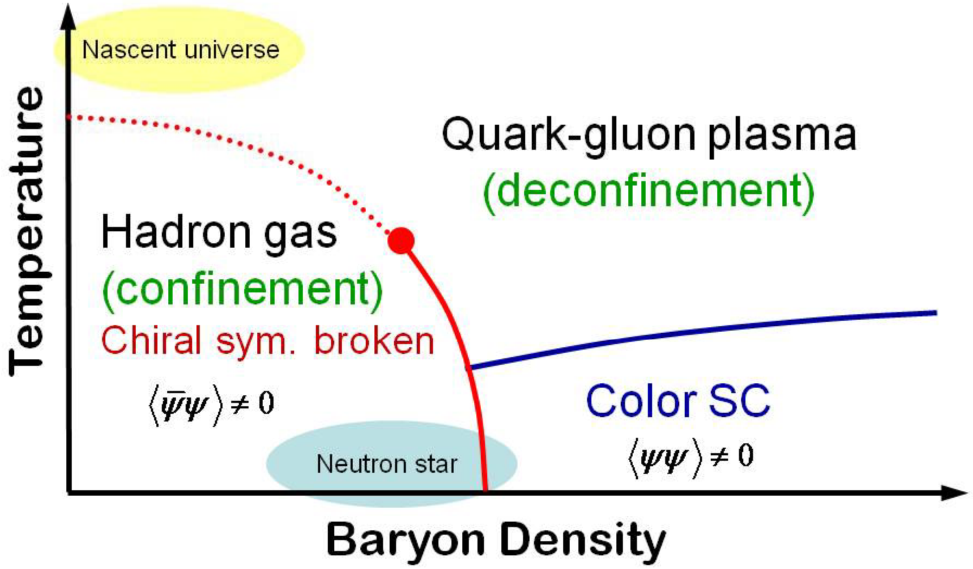
Charge-balanced/imbalanced electron-hole systems are good stages for exploiting the roots of matter.

Comparison of electron-hole systems with

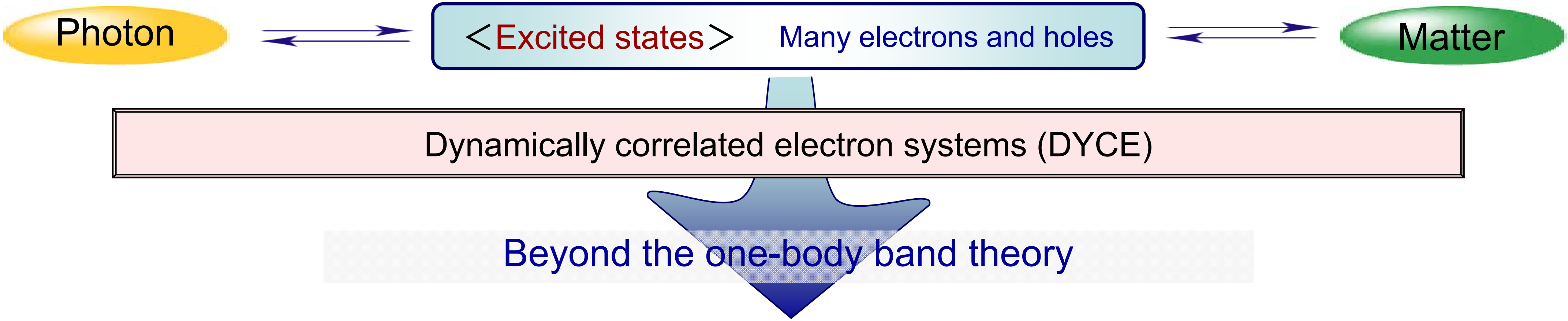
Electron-hole systems



Hadron QCD systems



Toward COE of DYCE Optical Physics



Photon

＜Excited states＞ Many electrons and holes

Matter

Dynamically correlated electron systems (DYCE)

Beyond the one-body band theory

New Optical Science, Materials Science, and Electronics

Ogawa group in OU is the unique and best place for theoretical study of DYCE optical physics to combine quantum many-body physics and quantum nonlinear optics.

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