QMC study of bi-layer electron-hole system

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School of Information Science,
Semi Conductor Bilayer

(Termalization time~ 1ns)/(Lifetime~ Several \(\mu \)s)

\[ d \sim 4\text{-}5\text{nm} \sim 100 \text{ a.u.} \quad \text{e.g., AlAs/GaAs} \]

\[ \frac{m_h}{m_e} \sim 0.50 \text{ (heavier hole)} \]
\[ \sim 0.09 \text{ (lighter hole)} \]

L. V. Butov \textit{et al.}, PRL 73, 304(1994).

S. Yang, PRB 81, 115320(2010).

「Exciton formation」
Motivation
for Electron-Hole systems

- **DMC methodological**
  As a case with evaluations other than GS energy.

- **Foundation of Photonics**
  electronics → photonics
  Identifying where EH pairs stably exist.

- **Foundation of Solid state Physics**
  Electron Gas ; Exchange and Correlation
  Elec.-Hole Gas ; Localization v.s. Delocalization
    'Exciton' Binding '2-comp. plasma' Screening

Laser & Semiconductors providing tractable experiments for Mott Tr.
  Particle Density controlled by Laser Intensity
Two Component Plasma
in 3d-EH gas

Mott Criterion
\[ r_s^{cr.} = 4 \left( \frac{12}{\pi} \right)^\frac{2}{3} = 9.8 \]

(Exciton Bohr rad.) = (Screening length)

Keldish's droplet
Keldish/68

Mean-field Th.
Unstable Phase
Brinkman-Rice/73

Correlation Methods
Recover toward stability
Vashista et.al./73

Higher Diagram, STLS etc.

Realistic Materials
more Stable
Combescot-Nozieres/72

Anisotropy, Multi-Valley...
Motivation for Electron-Hole systems

- **DMC methodological challenge**

  As a case with evaluations other than GS energy.

  Analysis using *Density Matrices, Pair Correlation Functions*...

  Firstly establishing implementations

  for such a system that definitely shows the transition between 2C plasma and Exciton phases

"Bilayer System"

Pablo L. Rios/Thesis (2001)

Model Bilayer

- $(d, r_s)$-plane Phase Diagram
- $m_h/m_e = 1.0$; Mass ratio fixed in the present study
Phase Diagram

... as predicted

Neil Drummond, 2D-HEG.

$d = 0 + \varepsilon$

Fluid

Wigner X'tal

Pairing

Bi-exciton? Phase Separation?

$r_s$

1.0

20

31.0
Bi-exciton captured

VMC config snapshot

2C-Plasma
\(d = 3\)

Excitonic
\(d = 0.3\)

Bi-excitonic
\(d = 0\)

Red/Blue ; Elec./Hole

\(\Delta/\nabla\); \(\uparrow\) spin/ \(\downarrow\) spin

\[r_s = 4 / \text{a.u.}\]

Plasma

Wigner X’tal

Exiton

Bi-exiton

\(d\) vs. \(r_s\)

AF

AF
Previous Studies

Analytic Approaches

BCS-type WF, mainly by Mean-field approached


Difficult to describe global feature of Phase Diagram (as a matter of course)

   Excitonic phase predicted stable at all the region

   not able to reproduce 2C-Plasma at (d→large)
Study by QMC

Good at for Global Phase Diagram

→ Intermediate Regions about many-body correlations

  Numerical Variational Approach

  VMC & DMC

1）Phase Boundary identification

  Order parameter via Density Matrices

2）Internal spatial structure inside each phase

  Pair Correlation Functions
De Palo's prev. Results

G. Senatore Group, PRL 88, 206401 ('02)

Successfully Described 2C plasma/Excitonic Phase Boundary

Our works

Order Parameter/Pair Correlation Function analysis using Single Wave Function scheme

→ Successfully captured Biexcitonic phase.
Multi WaveFunc. Scheme

Phase Boundary as Intersection (conventional)

Energies

Excitionic trial WF

Plasma trial WF

Single WF scheme

bilayer spacing

SWF doesn't use 'energy intersection identification' for phase Boundary
Single WF scheme

Description using most general form of WF

possible to get Variational advantage, but

\[ E[\Psi_{Normal}] > E[\Psi_{Pairing}] \]

no such identification possible.

→ Require direct evaluation of Order Parameter
to identify the Phase Boundary

  McMillan, \(^4\)He VMC (Solid/Liquid phase transition)
**Single WF scheme**

*Phase Boundary by Order parameters*

- Make use of Backflow Tr.
  
  Distinction between trial WFs becomes unclear when BF is used.

- Exciton Mott Transition details/ possibility of cross-over

\[
\lim_{|r| \to \infty} \gamma_{eh}^{(2)} (\vec{r}_e, \vec{r}_h; \vec{r}_e + \vec{r}, \vec{r}_h + \vec{r})
\]

**Order Parameter**

**Exciton Formation occurs**

**Pair Corr. Func.**

 Exitonic phase

 層内は通常のガス

**Spatial structure the phase has**
Strong Scaling on K

Hydrogen atom on graphene sheet

Time taken to do N DMC stats accumulation samples

K computer, Riken Institute, Japan

Question: Does K do "genuine" asynchronous communication when doing non-blocking MPI operations? (some hardware/MPI combinations 'fake' this.) MDT

Mike Towler (2013).

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<th>Ratio</th>
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Scalability

Improved by replacing MPI_SEND --> MPI_ISEND

By Gillan/Towler/Alfe (2011)
Results
Global survey(1)

- Only for (PW and Poly)/VMC/N=116 (no PPW, Wigner...)

--> To get reference values, first of all.
Multi WaveFunc. Scheme

SWF doesn't use 'energy intersection identification' for phase Boundary
Global survey (3)

\[ \delta E(\text{hartree}) \]
\[ \text{(per Particle)} \]

\[ \delta E = E_{\text{poly}} - E_{\text{pw}} \]

\[ -d(\text{bohr}) \]

\[ r_s(\text{bohr}) \]

- Only for (PW and Poly)/VMC/N=116 (no PPW, Wigner...)
  --> To get reference values, first of all.
Global survey(4)

VMC Phase Boundary estimated by Fluid and Paring Trial WF.
Possibility of Wigner X'tal phase not taken into account here.
De Palo's work

- DMC value at (rs=1.0, d=0.0)

\[ E = -0.417(4) \quad \text{De Palo et al., Phys. Rev. Lett. 88, 206401 (2002).} \]

\[ E = -0.4236 (1) \quad \text{Our result by Paring WF.} \]

- No Paring at rs=1.0?

De Palo reports No Paring

while our DMC shows Paring at smaller distance.
Order Parameter
Condensate Fraction for Exciton formation
(normalized into [0,1])

Correlation between pairs located $\bar{r}$ distance

$$\lim_{|\bar{r}| \to \infty} \gamma^{(2)}_{eh}(\bar{r}_e, \bar{r}_h; \bar{r}_e + \bar{r}, \bar{r}_h + \bar{r})$$

[Graph showing the off-diagonal long-range order with condensate fraction indicated]
Phase Correlation

between different snapshots

Two-body Density Matrix

\[ \gamma_{eh}^{(2)}(\vec{r}_e, \vec{r}_h; \vec{r}_e + \vec{r}, \vec{r}_h + \vec{r}) = N_1 (N_2 - \delta_{12}) \]

\[ \frac{\int |\Psi(\vec{R})|^2 \frac{\Psi(\vec{r}_e + \vec{r}, \vec{r}_h + \vec{r}, \ldots, \vec{r}_N)}{\Psi(\vec{r}_e, \vec{r}_h, \ldots, \vec{r}_N)} \, d\vec{r}_3 \ldots d\vec{r}_N}{\int |\Psi(\vec{R})|^2 \, d\vec{R}} \]

Measure How much Cancellation
Quantum Condensation

Two-body DM
\[
\gamma_2(\vec{x}_1, \vec{x}_2; \vec{y}_1, \vec{y}_2) = \frac{1}{2} \gamma_1(\vec{x}_1, \vec{y}_1) \gamma_1(\vec{x}_2, \vec{y}_2) - \frac{1}{2} \gamma_1(\vec{x}_1, \vec{y}_2) \gamma_1(\vec{x}_2, \vec{y}_1) + \frac{1}{2} \chi^*(\vec{y}_2, \vec{x}_2) \chi(\vec{y}_1, \vec{x}_1)
\]

One-body vanishes for Fermion

\[
\chi(\vec{x}, \vec{x}') = \langle \Psi_0 | \psi(\vec{x}') \psi(\vec{x}) | \Psi_0 \rangle
\]

should be non-zero for Condensation

For Slater Det. \(\Psi\), it vanishes and then \(\gamma_2\) is decoupled by \(\gamma_1\)

\[\rightarrow\] No Condensation

Quantum Condensation requires many-body description

beyond Slater Determinant

e.g., Geminal WF \[\rightarrow\] HFB theory
Density Matrix Sampling

many-body WF form

One-body DM: \[ \gamma^{(1)}(\vec{r}_i; \vec{r}_i') = N_1 \frac{\int \Psi^*(\vec{r}_1, \vec{r}_2, \ldots, \vec{r}_N) \Psi(\vec{r}_1', \vec{r}_2, \ldots, \vec{r}_N) d\vec{r}_2 \ldots d\vec{r}_N}{\int |\Psi(\vec{R})|^2 d\vec{R}} \]

\[ = N_1 \frac{\int |\Psi(\vec{R})|^2 \frac{\Psi(\vec{r}_1', \vec{r}_2, \ldots, \vec{r}_N)}{\Psi(\vec{r}_1, \vec{r}_2, \ldots, \vec{r}_N)} d\vec{r}_2 \ldots d\vec{r}_N}{\int |\Psi(\vec{R})|^2 d\vec{R}} \]

Two-body DM: \[ \gamma^{(2)}_{12}(\vec{r}_1, \vec{r}_2; \vec{r}_1', \vec{r}_2') = N_1 (N_2 - \delta_{12}) \frac{\int \Psi^*(\vec{r}_1, \vec{r}_2, \ldots, \vec{r}_N) \Psi(\vec{r}_1', \vec{r}_2', \ldots, \vec{r}_N) d\vec{r}_3 \ldots d\vec{r}_N}{\int |\Psi(\vec{R})|^2 d\vec{R}} \]

\[ = N_1 (N_2 - \delta_{12}) \frac{\int |\Psi(\vec{R})|^2 \frac{\Psi(\vec{r}_1', \vec{r}_2', \ldots, \vec{r}_N)}{\Psi(\vec{r}_1, \vec{r}_2, \ldots, \vec{r}_N)} d\vec{r}_3 \ldots d\vec{r}_N}{\int |\Psi(\vec{R})|^2 d\vec{R}} \]
Order parameter

Condensate Fraction of Exciton

\[ \lim_{|r| \to \infty} \gamma_{eh}^{(2)}(\bar{r}_e, \bar{r}_h; \bar{r}_e + \bar{r}, \bar{r}_h + \bar{r}) \]

Order Parameter

Exciton Formation occurs
Order Parameter

Revised on 13 Sep. 2012.
Phase Diagram

$d(\text{bohr})$

Fluid

Pairing

Fluid/Pairing boundary?

Stronger Order parameter?

Inset graph:

Coordination Fraction

$d = 0.5$

$d = 1.0$

$r_s$
Dying-off behavior

Condensate Fractions

d=0.00

d=0.01

d=0.05

d=0.10

d=0.15

d=0.20

d=0.40

condensate fraction vs rs (hartree)
Dying-off behavior

\[ d(\text{bohr}) \]

Fluid  Pairing  Wigner X'tal

Plasma  Exiton

AF

AF

\[ r_s \]

p.09
Dying-off

$d(\text{bohr})$

Fluid
Pairing

0.4
0.2
0.1
0.1

Bi-exciton Phase

$r_s$

Plasma
Wigner X'tal
Exiton
AF
AF

$p.09$
Pair Correlation Functions
Different behaviors in Eu-Ed distributions.

**Exitonic phase**

**Biexitonic phase**

Correlation Hole

Exchange Hole

Molecule formed within layer

Biexiton Radius $R_{BE}$ characteristic profile appears with peak.
VMC config snapshot

2C-Plasma
\( d = 3 \)

Excitonic
\( d = 0.3 \)

Bi-excitonic
\( d = 0 \)

Red/Blue; Elec./Hole

\( \Delta/\nabla \); ↑ spin/↓ spin

\( d \)

\( r_s = 4 \text{ / a.u.} \)

Plasma

Exiton

Bi-exciton

Wigner X’tal

AF

AF

1.0 3.0 20 31.0
Biexciton Radius

Revised on 20 May. 2012.

rs=0.0/rs=0.6/N=116/d=0.00
rs=4, Eu-Ed
rs=6, Eu-Ed (x1/2)

\( r_s = 4 \)
\( r_s = 6 \)

Finer definition for \( R_{BE} \) required?

Based on \( \int dr \cdot w(r) \cdot g(r) \)
Inter-Biexciton length

Revised on 18 May. 2012.

rs=04.0/rs=06.0/N=116/d=00.00

Pair Correlation Functions

Distance (bohr)

2nd peak (rs=6)

rs=6, Eu-Ed (x1/2)

2nd peak (rs=4)

rs=4, Eu-Ed

$L_{BE}(r_s = 6) \sim 13$

$L_{BE}(r_s = 4) \sim 10$

Biexciton Gas