Collision-induced processes with super-cooled excitons

Makoto Kuwata-Gonokami

Department of Applied Physics, University of Tokyo
SORST, Japan Science and Technology Agency (JST)

http://www.gono.t.u-tokyo.ac.jp
1. **Introduction**

2. **Excitation and detection of cold excitons in Cu$_2$O**
   Direct excitation of *super-cooled* excitons by pulsed two-photon resonant excitation of 1s-ortho excitons
   Collision induced ortho to para transformation

3. **Quasi-steady state excitonic Lyman spectroscopy**
   Paraexcitons at quasi-equilibrium condition
   detected by CW excitonic Lyman spectroscopy with CO$_2$ laser
   Evaluation of density-dependent particle loss

4. **Future Prospects**
Coworkers

T. Tayagaki  
(Univ. Tokyo)  

K. Yoshioka  
(Univ. Tokyo)  

T. Ideguchi  
(Univ. Tokyo)  

A. Mysyrowicz  
(ENSTA,  
Ecole Polytechnique,  
France)  

M. Kubouchi  
(Univ. Tokyo)  

Y. Svirko  
(Joensuu Univ.  
Finland)  

R. Shimano  
(Univ. Tokyo)  

Acknowledgement  
N. Naka (Univ. Tokyo)
BEC of Excitons

\[ T_c = \frac{2\pi \hbar^2}{mk_B} \left( \frac{n}{2.612} \right)^{2/3} \]

Excitons in Semiconductors

small mass (less or comparable with the free electron) \(\rightarrow\) high \(T_c\)

Density is easily controlled by light: boson-fermion crossover

- **87Rb:** \(\sim 10^5 \times m_e\) \(\sim 10^{12} \text{ cm}^3\) \(10^{-7} \text{ K}\)
- **Cu_2O 1s-exciton:** \(\sim 3 \times m_e\) \(10^{17} \text{ cm}^3\) \(1.9 \text{ K}\)
History of exciton BEC

1960


I. M. Blatt, K. W. Börer, and W. Brandt, Phys. Rev. 126, 1691

1970


Bose statistics of Cu$_2$O orthoexcitons


BEC of Cu$_2$O paraexcitons

1980


Quantum saturation of Cu$_2$O orthoexcitons

1990


Phase transition to ordered state of indirect excitons in coupled quantum well


BEC of Cu$_2$O paraexcitons

1990


Excitonic superfluidity in Cu$_2$O

1990


Quantum saturation of Cu$_2$O orthoexcitons

1990


Phase transition to ordered state of indirect excitons in coupled quantum well


BEC of Cu$_2$O paraexcitons

Theoretical Prediction

1960

Theoretical Prediction

1960
Difficulties in Exciton BEC

**Finite life time**
thermal relaxation versus recombination lifetime
\[ k_B T_c \text{ versus } \eta / \tau_{\text{life}} \]

**Open system**
diffusion of exciton

**Heating**
photo excitation
nonradiative recombination
Excitons in Cu$_2$O

1s-excitons: electric dipole transition forbidden

$2\Gamma_6^+ + x 2\Gamma_7^+ + x \Gamma_1^+ = 3\Gamma_5^+ + \Gamma_2^+$

Γ$_5^+$ orthoexciton: electric quadrupole transition allowed

Γ$_2^+$ paraexciton: pure spin-triplet

→ optical transition is strictly forbidden,
extremely long life time $\tau = 10 \mu$s

$E_{ex} = E_g - \frac{R_y}{n^2}$

n = 2 3 4

Optical Density (arb. unit)

Cu$_2$O (4.2K)
Paraexcitons in Cu$_2$O

\[ J=1: \text{ortho} \quad (\Gamma_5^+) \]

\[ J_z = 1: \left| \uparrow_e \uparrow_H \right> \]

\[ J_z = 0: \frac{1}{\sqrt{2}} \left( |\uparrow_e \downarrow_H\rangle + |\downarrow_e \uparrow_H\rangle \right) \]

\[ J_z = -1: |\downarrow_e \downarrow_H\rangle \]

**note:**

\[ |\uparrow_H\rangle = -\frac{1}{\sqrt{3}} \left[ (yz+i zx)|\downarrow_h\rangle + (xy)|\uparrow_h\rangle \right] \]

\[ |\downarrow_H\rangle = -\frac{1}{\sqrt{3}} \left[ (yz-i zx)|\uparrow_h\rangle - (xy)|\downarrow_h\rangle \right] \]

\[ |\uparrow_e \downarrow_H\rangle - |\downarrow_e \uparrow_H\rangle = -\frac{1}{\sqrt{3}} \left[ (yz-i zx)|\uparrow_e |\uparrow_h\rangle - (yz+i zx)|\downarrow_e \downarrow_h\rangle - xy \left( |\uparrow_e \downarrow_h\rangle + |\downarrow_e \uparrow_h\rangle \right) \right] \]

**Para exciton state is purely spin-triplet state**

\[ \Delta E_{\text{ex}}(K=0) = 12 \text{meV} \]

\[ \Delta E_{\text{ex}}(K) = \frac{2}{3} \int \Psi_K^*(x,x') \frac{e^2}{\epsilon_\infty |x-x'|} \Psi_K(x',x') \, dx \, dx' \]


\[ \rightarrow \text{no direct optical processes} \]
Phonon-assisted luminescence spectrum of orthoexciton ($X_o$-$\Gamma_3^-$) are well fitted with Bose-Einstein distribution function. However, orthoexcitons remain in normal region. Paraexciton BEC?
How to detect optically forbidden paraexcitons?

Very weak luminescence of paraexcitons

$$\eta_{\chi_0-\Gamma_3} : \eta_{\chi_p-\Gamma_5} \approx 500:1$$


Paraexcitons were detected by field ionization (Schottky barrier).


Ballistic transport of paraexcitons: super fluidity?

Spectroscopic information was lost.
Objection to exciton BEC in Cu$_2$O

Quantitative analysis of luminescence measurement

1) Luminescence spectrum can be reproduced by MB distribution with spatial inhomogeneity: not BE statistics

2) TA-phonon mediated ortho-para conversion rate:
   \( \tau_{o-p} = 3 \) ns \( (T=2 \) K) 
   \( \rightarrow \) too slow conversion rate to accumulate paraexcitons


3) Large Auger recombination rate
   (nonradiative two-body decay)
   \( A = 10^{-16} \) cm$^3$/ns : Excitons decay before reaching the critical density \( n_c = 10^{17} \) cm$^{-3}$

\[ \frac{dn}{dt} = -An^2 \]

No BEC of ortho nor paraexcitons!
Optical detection of paraexcitons by 1s-2p transition

Probing 1s-2p induced absorption
- electric dipole transition allowed for both *ortho*- and spin-triplet *para*-excitons
- distinguish paraexcitons from orthoexcitons
- \( m_{1s} > m_{2p} \rightarrow \) induced absorption spectrum reflects the distribution of excitons

Theoretical Prediction;

CW probe experiments; (1) M. Jorger & C. Klinghirn,
(2) K. Karpinska & P.H.M. von Loosdrecht (Groningen)

Exchange energy

Our approach:
Time resolved Excitonic Lyman Spectroscopy

Evaluation of Exciton density from 1s-2p induced absorption

\[
\left( \mathcal{N}(E, N_{ex}) \right)^2 = \varepsilon_b + \mathcal{N}(N_{ex}, E)
\]

\[
\mathcal{N}(N_{ex}, E) = N_{ex} \cdot \frac{2E_{1s-2p}}{\varepsilon_0} \cdot \frac{\left| \mu_{1s-2p} \right|^2}{(E^2 - E_{1s-2p}^2 - i\Gamma E)}
\]

\[
\Delta\alpha(E) = \frac{E}{\hbar c \sqrt{\varepsilon_b}} \cdot \text{Im}\left\{ \mathcal{N}(N_{ex}, E) \right\}
\]

\[
S \equiv \int \Delta\alpha(E) dE = N_{ex} \cdot \frac{\pi E_{1s-2p} \left| \mu_{1s-2p} \right|^2}{\hbar c \varepsilon_0 \sqrt{\varepsilon_b}}
\]

\[
N_{ex} = \frac{\hbar c \varepsilon_0 \sqrt{\varepsilon_b}}{\pi E_{1s-2p} \left| \mu_{1s-2p} \right|^2} \cdot S
\]
Sample

Cu$_2$O naturally grown single crystal
3 × 5 mm
Thickness 200 μm

Mid-infrared linear absorption spectrum

transparent window near 1s-2p transition!
Experimental setup: mid-infrared pump-probe spectroscopy

- Ti: Sapphire Regen. (1 kHz, 150 fs)
- OPA + DFG (AgGaS$_2$, GaSe)
- OPA + SHG (BBO)
- pump: 1220 nm, 600 nm (width: 14 meV, 2 μJ)
- probe: ~10 μm

Sample (liq. He)

Spot size: 300 μm

Monochromator

HgCdTe detector

PC  I/O  Boxcar Integrator

Trigger

Delay

Chopper
Induced absorption spectra by one-photon (orthoexciton-phonon-sideband) excitation

1) Strong signal at para exciton resonance.
2) Spectrum narrowing with time.
3) Red shift of the absorption maximum.

Initially hot excitons

$T_{ex} \gg T_{lattice}$

Thermalization with lattice

Ortho

Para

Induced absorption spectra

Energy (meV)

Delta (cm$^{-1}$)

110 120 130

-10 ps

10 ps

30 ps

200 ps

800 ps

Excitation
Two-photon electric dipole transition of orthoexcitons is allowed.

\[
\Gamma_4 - x \Gamma_4 = \Gamma_1 + \Gamma_3 + \Gamma_4 + \Gamma_5
\]

orthoxcitons \( k_0 \sim 0 \)

Phase space compression of laser photons by resonant two-photon excitation

\[\text{Instantaneous preparation of Quantum degenerate orthoexcitons}\]

\[\text{Large phase space density of photons in ML-fs laser}\]

\[76\text{MHz repetition, } \delta \lambda \ 2\text{nm, } 1\text{mW}\]

\[\text{Photon number per mode; } n_\nu = 500\]

Resonant two-photon excitation of orthoexciton

No peak shift -> No excess heating

cold distribution around $k=0$

$T_{ex} < T_{lattice}$

$N_{ortho} \sim 10^{15} \text{ cm}^{-3}$

Rapid production of paraexcitons

$\Delta \alpha \text{ (cm}^{-1}\text{)}$

Energy (meV)

Resonant two-photon excitation of orthoexciton
Observation of Excitonic Lyman series

Low density regime: $N_{\text{ex}} < 10^{15}$ cm$^{-3}$

- Energy (meV)
  - $115$ to $135$
  - $2145$ to $2170$
- $\alpha$ (cm$^{-1}$)
  - $0$ to $400$
- $\Delta \alpha$ (cm$^{-1}$)
  - $0$ to $6$

- $\tau_{\text{delay}} = 4$ ns
  - $1s$-$2p$ para
- $\tau_{\text{delay}} = 5$ ps
  - ortho-exciton

- $12$ meV
- $129$ meV
- $116$ meV

- $2\omega = 2033$ meV
Thermalization dynamics of super-cooled 1s orthoexciton (4.2K)

\[
\Delta \alpha (\text{cm}^{-1}) = (\Delta E_{1s-4p})^2 (\gamma_{4p})^2 + ((m_{1s}/m_{4p}-1)f_{1s})^2
\]

Line shape analysis:

\[ n \sim 10^{15} \]

Super cooled state

Boltzmann fitting
Extraction of ortho-para conversion

n_{ex}(t\sim0)= 8\times10^{14} \text{ cm}^{-3}

\textbullet \ Total exciton density conserves

Ortho-para conversion: \ \tau_{o-p}=3 \text{ ns}

TA-phonon assisted spin transformation

1.8 K
Temporal evolution of excitons; High density excitation

- Nonlinear ortho-to-paraexciton conversion

- Linear ortho-para conversion: $\tau_{op} = 3$ ns

Temporal evolution of excitons

High density excitation

$n(t \sim 0) \sim 4 \times 10^{15}$ cm$^{-3}$
Collision induced ortho-para conversion


\[ \frac{dn}{dt} = -Cn^2 \]

\[ C \approx 5 \times 10^{-16} \text{ cm}^3 / \text{ns} \]
Temporal evolution of $n_{\text{ortho}}, n_{\text{para}}, n_{\text{total}}$

Model:

$$\frac{d}{dt} n_t = -An_t^2 \quad n(t) = \frac{n_0}{1 + An_0 t}$$

\[
\begin{align*}
\frac{d}{dt} n_O &= -\Gamma_{o-p} n_o - 2An_o n_t + \frac{3}{4} An_t^2 - Cn_o^2 \\
\frac{d}{dt} n_p &= \Gamma_{o-p} n_o - 2An_p n_t + \frac{1}{4} An_t^2 + Cn_o^2
\end{align*}
\]

($n_t = n_o + n_p$)

A: Dissociation process
C: Spin-flip process

If $C >> A$, we can accumulate paraexcitons before we lose excitons by Auger recombination.
Extraction of collision-induced spin-flip process

\[ \frac{d}{dt} n_{\text{para}} = -C n_{\text{ortho}}^2 \]

\[ \frac{d}{dt} n_{\text{total}} = -A n_{\text{total}}^2 \]

\[ (A=10^{-16} \text{ cm}^3/\text{ns}) \]


\[ C = 2.6 \times 10^{-16} \text{ cm}^3/\text{ns} \]

\[ C = 5 \times 10^{-16} \text{ cm}^3/\text{ns} \]

\[ C > A \]
Enhanced collision induced spin conversion of excitons: Virtual biexciton mediated resonant scattering?

Biexciton state in Cu$_2$O; Large e-h exchange makes Biexciton state unstable


Biexciton state in Cu$_2$O; Large e-h exchange makes Biexciton state unstable

Resonant scattering enhances collision cross section

Virtual biexciton state (nearly zero binding energy)
Paraexcitons generated via TPA of orthoexcitons

Summary of femtosecond experiments

We obtain paraexciton density of $10^{15}$ cm$^{-3}$ under orthoexciton excitation of $4 \times 10^{15}$ cm$^{-3}$

$T_{\text{para}} < 20$K

$C \sim 2.6 \times 10^{-16}$ cm$^3$/nsec

Questions:
- Mechanism of giant collision cross-section?
- Why did we obtain cold paraexcitons?

We need to
- Accumulate paraexcitons with continuous feeding at low lattice temperature.
- Precisely estimate Auger rate and paraexciton lifetime

CW based experiment
Excitonic 1s-$np$ transitions and CO$_2$ laser lines

Quasi-steady state measurements for long lived paraexcitons

Accidental coincidence – Single mode tunable CO$_2$ laser to probe 1s paraexcitons
Experimental Set-up: Steady-state excitonic Lyman spectroscopy

Pump: single-mode dye laser (orthoexcitons)
Probe: single-mode tunable CO₂ laser

Induced absorption (-ΔT/T) can be obtained by measuring f & 2f components of the probe beam using lock-in amplifier(s)
1s-2p absorption spectra of quasi-steady state paraexcitons

Temperature dependence of differential transmission spectra at 1s-2p paraexciton resonance

Due to the relative stability of the probe light, we are currently able to detect a transmission variation as small as 0.001 % (corresponds to $<10^{12} \text{ cm}^{-3}$)

Exactly match theoretical curves assuming Maxwell-Boltzmann distribution functions

We successfully detected 1s paraexcitons in a steady state regime!
Life time measurement of paraexcitons

Reported value of paraexciton lifetime:
Several hundred nanoseconds to milliseconds with luminescence measurements*

Lifetime measurement of 1s paraexcitons by CW Lyman spectroscopy

We measure the probe pulse transmission and evaluate the induced transmission change.

Excitation intensity dependence of paraexciton density

Sublinear dependence on excitation intensity

Auger effect is also observed in this steady-state regime

Temperature dependence of Auger recombination rate

**Numerical simulation**
- Temperature-dependent diffusion of excitons (2D)
- Nonlinear rate equations (Auger recombination)

**Temperature dependence of the nonlinear particle loss rate**

*Collision-induced spin-frip processes are not included*

\[ \tau^{-1} = \sigma v_{th} \cdot n_{ex} \]

**Independent of temperature**

The density-dependent particle loss rate cannot be explained by a classical hard-sphere model:

\[ v_{th} = \sqrt{\frac{8k_B T}{\pi m_{ex}}} \]
Conclusion

1) We proposed and demonstrated a scheme to detect paraexcitons by using the 1s-2p transition of excitons. This allows us to quantitatively study the temporal and spatial behavior of paraexcitons.

2) Excitonic Lyman series of super-cooled orthoexciton was observed. We found that high density cold paraexcitons are efficiently created by resonant two-photon excitation of orthoexcitons.

3) To examine the dynamics of long lived paraexcitons, we developed CW CO$_2$ laser-based Lyman spectroscopy. We measured a paraexciton lifetime longer than 20 micro seconds. We also obtained information on the collision-induced loss of paraexcitons under quasi-equilibrium condition.
Optical Trapping with Resonant Dressed Field

Exciton gas can be trapped by the Stark potential.

\[ \delta E = \frac{1}{2} \left( \sqrt{\Delta^2 + (\mu_{1s-2p} E)^2} - \Delta \right) \]
\[ \approx \frac{1}{2\Delta} (\mu_{1s-2p} E)^2 \propto I \]

\( \mu_{1s-2p} \) (transition dipole moment) = 4.2 eÅ
\( \Delta \) (detuning) = 1 meV
\( I \) (Intensity) = 20 MW/cm²

\( \delta E = 0.8 \) meV
(corresponds to 2K)
Sympathetic cooling under high-density excitation

Phonon scattering

\[ E \]

1s-ortho
1s-para

Phonon scattering rate:
\[ \gamma_{LA}^{-1} = 5 \text{ ns at 2K} \]

Experimental:
\[ \tau \approx 300 \text{ ps} \]

Sympathetic cooling

\[ E \]

1s-ortho
1s-para

Ortho-to-para exciton conversion

Hot paraexciton

Super-cooled ortho
How to cool paraexcitons?

Energy to be extracted from excitons \(10^{16} \text{ cm}^{-3}\) at \(T=2 \text{ K}\) in a \((100 \ \mu\text{m})^3\) box to cool down to \(T=1 \text{ K}\): \(4.12 \times 10^{-13} \text{ J}\)

- Sympathetic cooling with super cooled orthoexciton gas
  
  \[10^{16} \text{ excitons in } V=(100 \ \mu\text{m})^3 \quad T=0 \text{ K} \rightarrow T=1 \text{ K}\]
  
  *classical gas

- Heat exchange with the lattice
  
  \[V=(100 \ \mu\text{m})^3 \text{ lattice} \quad T=0.98 \text{ K} \rightarrow T=1 \text{ K}\]

Heat capacity of a phonon field is two-orders of magnitude larger than that of a cold exciton gas

We need to cool down the crystal.

How to reach excitonic BEC phase?

$n=1 \times 10^{17}$ cm$^{-3}$ \hspace{1cm} $T_c=2.3$ K

$n=1 \times 10^{16}$ cm$^{-3}$ \hspace{1cm} $T_c=500$ mK \hspace{1cm} ($m_{ex}=2.7 m_e$)

Exciton gas

Phase boundary

$^3$He refrigerator
Excitonic BEC created by spatial-temporal controlled optical pulses

- Efficient creation of ultracold excitons by chirp-controlled optical pulses (Frequency Control)
- Spatial confinement of ultracold excitons by polarization and wavevector controlled optical pulses (Spatial Control)

![Diagram of phase space compression scheme](attachment://phase_space_compression.png)

- Excitonic BEC
- Macroscopic Coherence

Energy vs. Wave number diagram with Biexciton and Energy levels:
- $2k_0$
- $E_{m}(2k_0)/2$
- $2.4 \mu eV$
- $10^2 \text{ cm}^{-1}$
- $12 \text{ meV}$

Phase diagram:
- Temperature (K)
- Density ($\text{cm}^{-3}$)
- BEC

CREST-JST: Oct. 2006~
Confinement of paraexcitons:
   Optical Trap by MIR field (1s-2p resonant exciton transition)
**Accumulation of para-excitons below 1K region;**
Cooling of para-excitons with super cooled ortho-excitons;
   …. Sympathetic cooling of excitons

**CW –based Experiment:**
   poster by Kousuke Yoshioka