***The 10th US-Japan Joint Seminar** – Fundamental Issues and Applocations of Ultracold and Molecules -*

Collision-induced processes with super-cooled excitons

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Outline

1. Introduction
2. Excitation and detection of cold excitons in Cu_2O

   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

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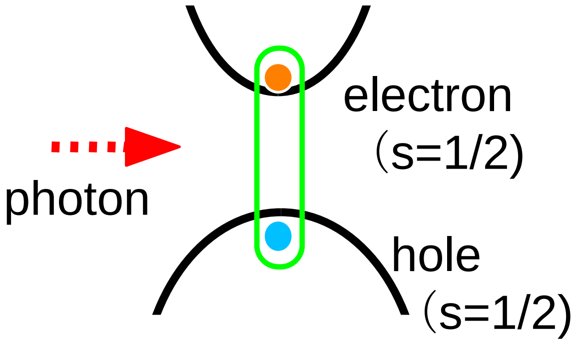
Acknowledgement
N. Naka (Univ. Tokyo)

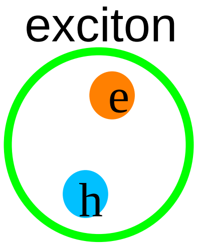
BEC of Excitons

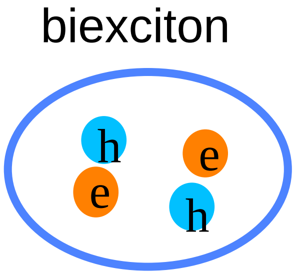
$$T_c = \frac{2\pi\hbar^2}{mk_B}\left(\frac{n}{2.612}\right)^{2/3}$$

Excitons in Semiconductors



quasi Bose particles

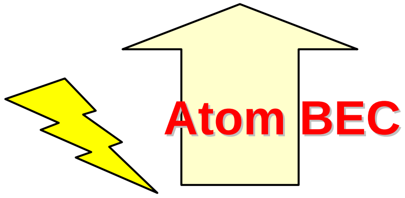




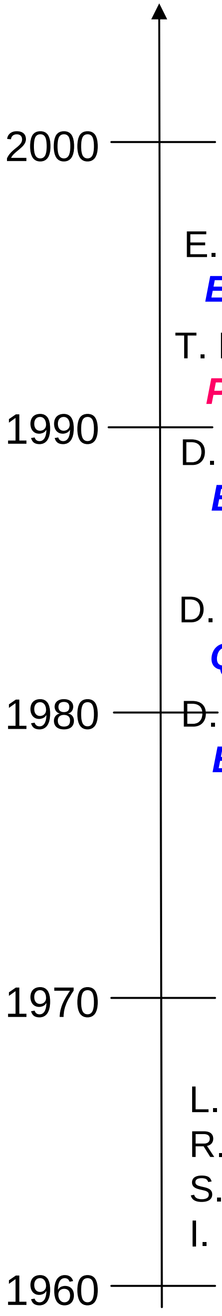
Small mass (less or comparable with the free electron) →high T_c
Density is easily controlled by light: boson-fermion crossover

	mass	n_c	T_c
^{87}Rb:	$\sim 10^5 \times m_e$	$\sim 10^{12}$ cm^{-3}	10^{-7} K
Cu_2O 1s-exciton:	$\sim 3 \times m_e$	10^{17} cm^{-3}	1.9 K

History of exciton BEC



Rb,Na BEC(95)



E. Fortin, E. Benson, and A. Mysyrowicz, Phys. Rev. Lett. **70**, 3951 (1993).

Excitonic superfluidity in Cu_2O

T. Fukuzawa, E. E. Merdez, and J. M. Hong, Phys. Rev. Lett. **64**, 3066 (1990).

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

D. W. Snoke, J. P. Wolfe, and A. Mysyrowicz, Phys. Rev. B **41**, 11171 (1990).

BEC of Cu_2O paraexcitons

Early Experiments on Cu_2O

D. Snoke, J. P. Wolfe, and A. Mysyrowicz, Phys. Rev. Lett. 59, 827 (1987).

Quqntum saturation of Cu_2O orthoexcitons

D. Hulin, A. Mysyrowicz, and C. Benoît à la Guillaume, Phys. Rev. Lett. **45**, 1970 (1980).

Bose statistics of Cu_2O orthoexcitons

biexciton BEC in CuCl

Theoretical Prediction

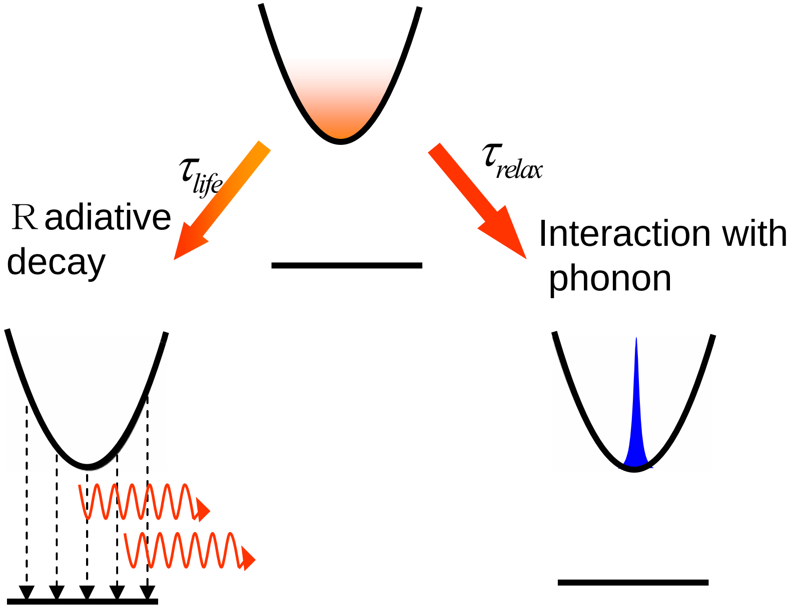
L. V. Keldysh and A. N. Kozlov, Sov. Phys. JETP **27**, 521 (1968).
R. C. Casella, J. Phys. Chem. Solids **24**, 19 (1963).
S. A. Moskalenko, Fiz. Trerd. Tela. **4**, 276 (1962) [Sov. Phys. Solid State **4**, 199 (1962)].
I. M. Blatt, K. W. Böer, and W. Brandt, Phys. Rev. **126**, 1691

Difficulties in Exciton BEC

Finite life time

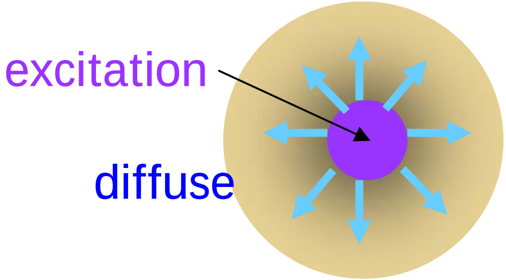
thermal relaxation versus
recombination lifetime

$k_B T_c$ versus $\eta / \tau_{\mathrm{life}}$



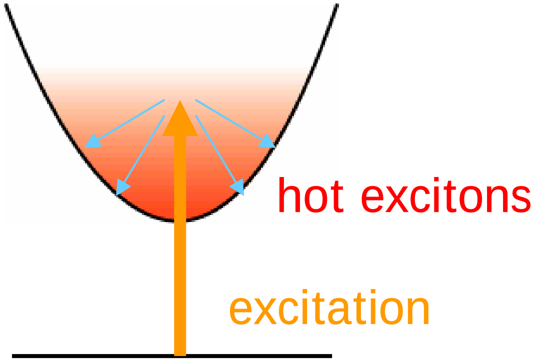
Open system

diffusion of exciton

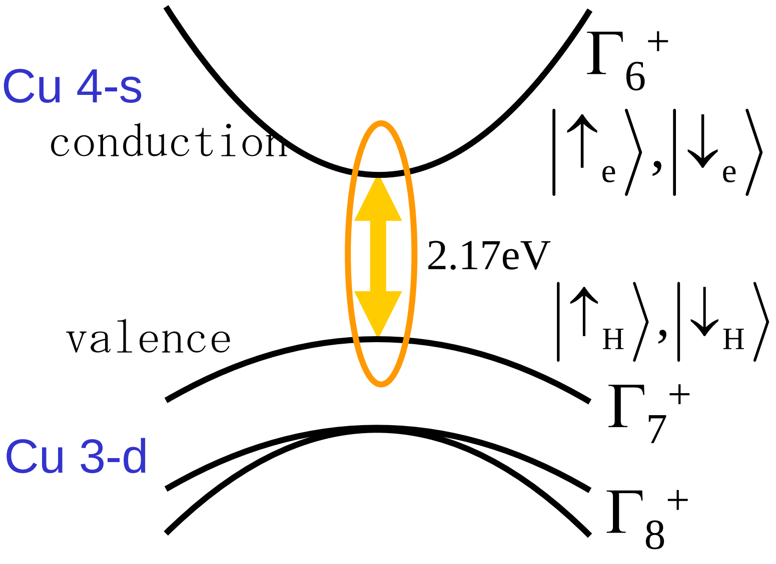


Heating

photo excitation
nonradiative recombination

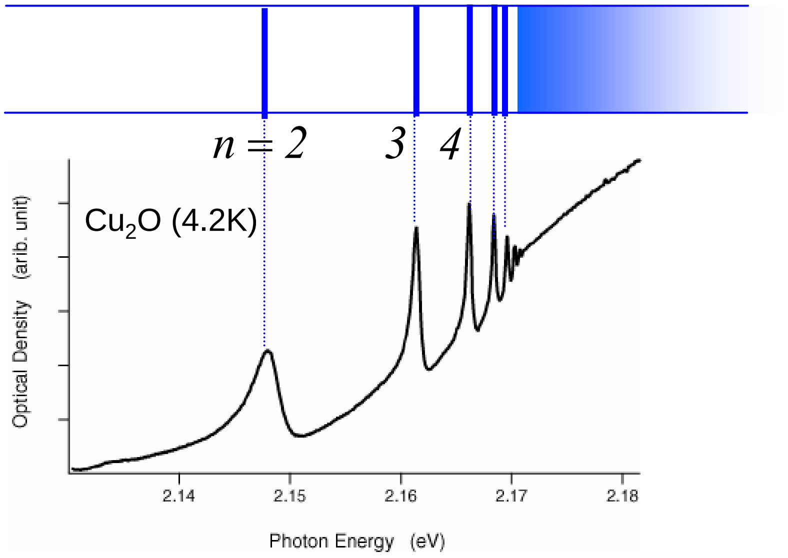


Excitons in Cu_2O



Yellow-series exciton

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



1s-excitons: electric dipole transition forbidden

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

Γ_5^+ orthoexciton: electric quadrupole transition allowed

$$\tau = 10\, n\,\mathrm{sec}$$
$$(\tau_{rad} > 300\, n\,\mathrm{sec})$$

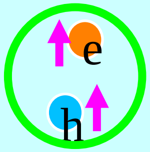
Γ_2^+ paraexciton: pure spin-triplet

→ optical transition is strictly forbidden,

extremely long life time $\tau = 10 \mu\mathrm{sec}$

Paraexcitons in Cu_2O

***J*=1: ortho** (Γ_5^+)

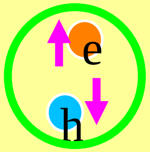


$$J_z = 1 : |\uparrow_e \uparrow_H\rangle$$

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

$$J_z = -1 : |\downarrow_e \downarrow_H\rangle$$

***J*=0: para** (Γ_2^+)



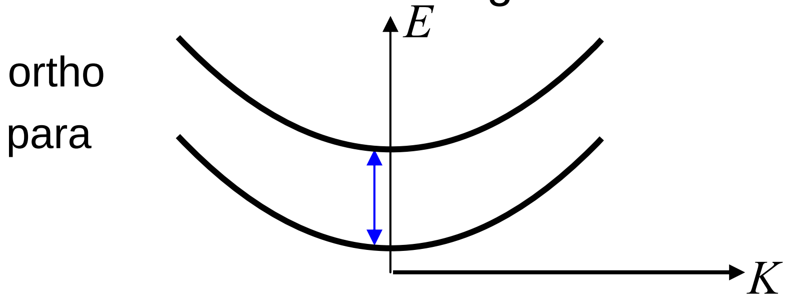
$$J_z = 0 : \frac{1}{\sqrt{2}}\left(|\uparrow_e \downarrow_H\rangle - |\downarrow_e \uparrow_H\rangle\right)$$

note;

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

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

electron-hole exchange interaction



$\Delta E_{ex}(K=0)=12\mathrm{meV}$

$$\Delta E_{\mathrm{ex}}(K) = \frac{2}{3}\int \Psi_{\mathbf{K}}(\mathbf{x},\mathbf{x})\frac{e^2}{\varepsilon_\infty |\mathbf{x}-\mathbf{x}'|}\Psi_{\mathbf{K}}{}^*(\mathbf{x}',\mathbf{x}')\,\mathrm{d}\mathbf{x}\mathrm{d}\mathbf{x}'$$

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

Para exciton state is purely spin-triplet state

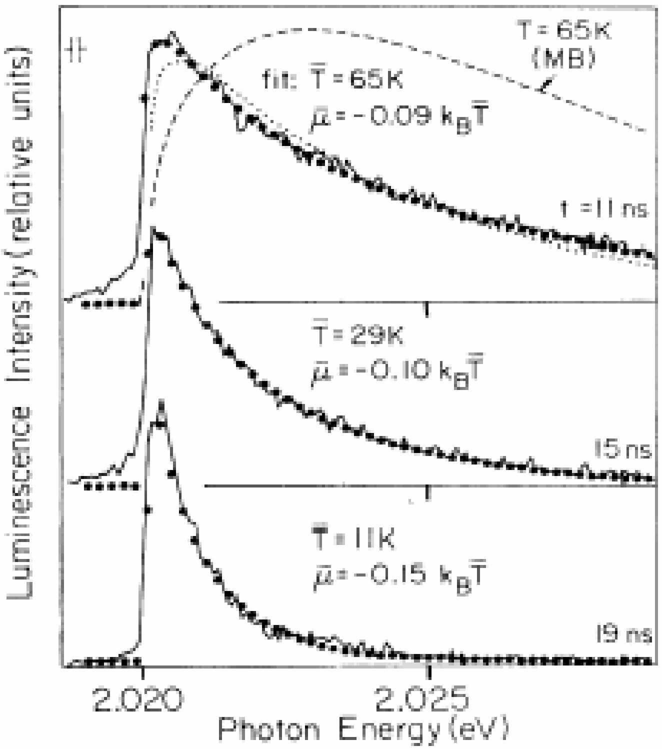
-> no direct optical processes

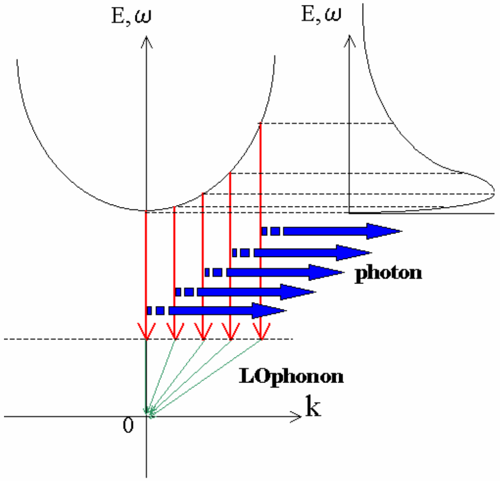
G. M. Kav

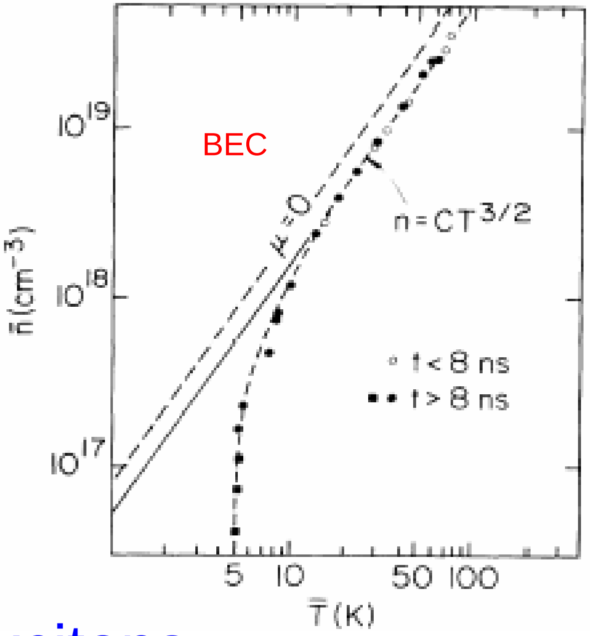
Experiments in Cu_2O so far: luminescence spectrum analysis

Phonon-assisted luminescence spectrum of orthoexciton (X_o-Γ_3^-) are well fitted with Bose-Einstein distribution function

D. W. Snoke *et al.*,
Phys. Rev. Lett. **59,** 828 (1987).







However, orthoexcitons remain in normal region.

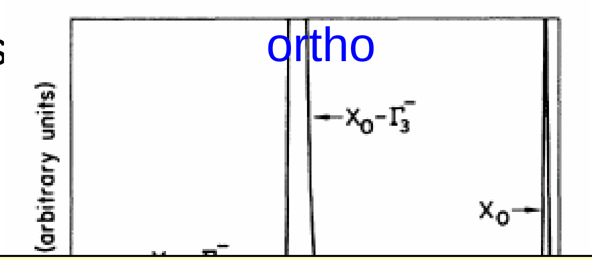
→ **Paraexciton BEC ?**

How to detect optically forbidden paraexcitons?

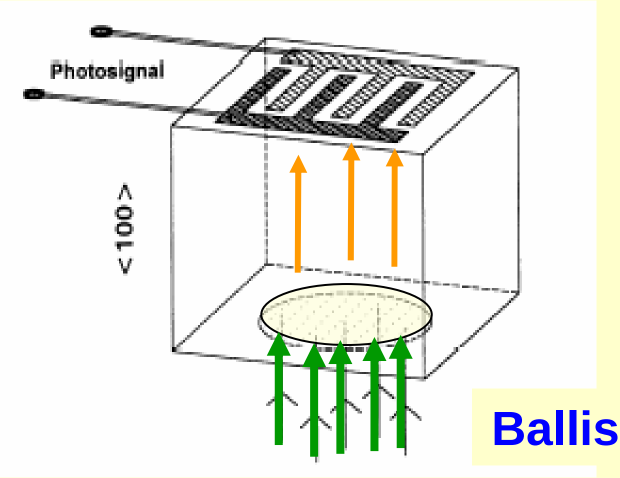
Very weak luminescence of paraexcitons

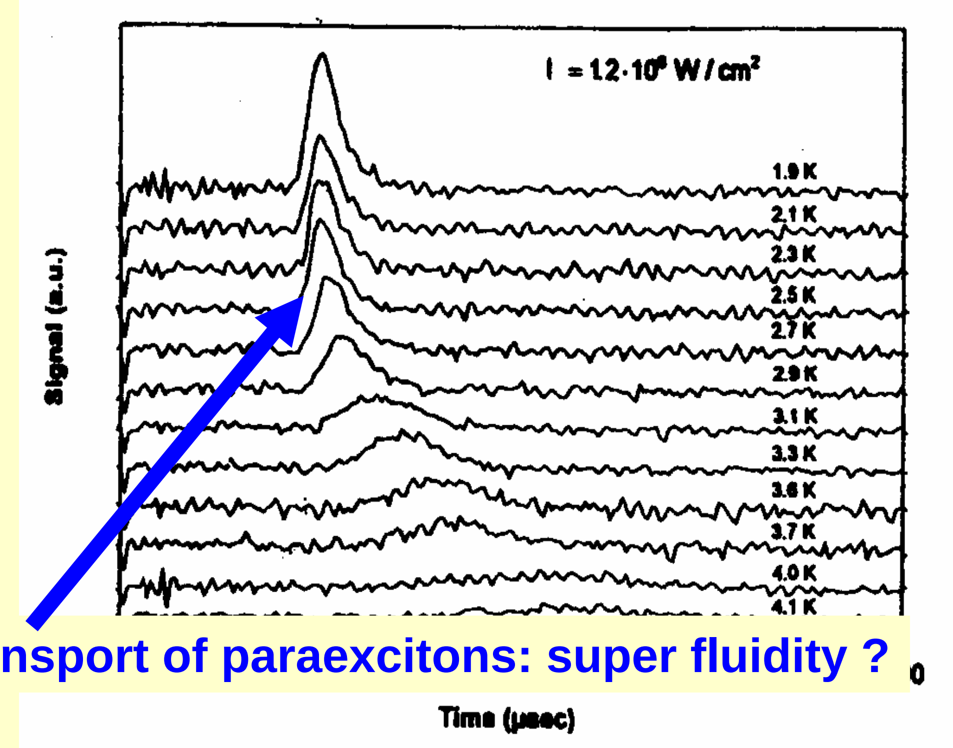
$\eta_{Xo\text{-}\Gamma3} : \eta_{Xp\text{-}\Gamma5} \sim 500:1$

D. W. Snoke *et al.*,
Phys. Rev. B **41**, 11171 (1990).



Paraexcitons were detected by field ionization (Schottky barrier) .





Ballistic transport of paraexcitons: super fluidity ?

Spectroscopic information was lost.

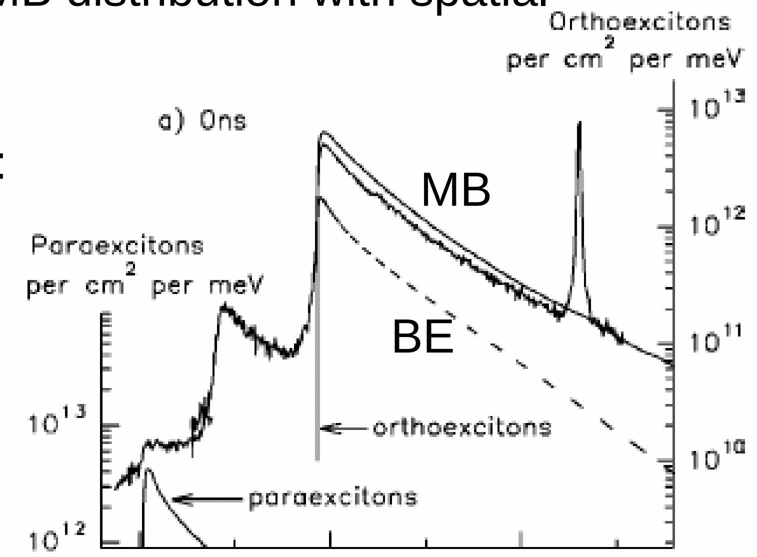
E. Fortin, E. Benson, and A. Mysyrowicz, Phys. Rev. Lett. **70**, 3951 (1993).

Objection to exciton BEC in Cu_2O

Quantitative analysis of luminescence measurement

K. E. O'Hara and J. P. Wolfe, Phys. Rev. B **62,** 12909 (2000).

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



2) TA-phonon mediated ortho-para conversion rate:

$\tau_{o\text{-}p}$=3 ns (T=2 K)

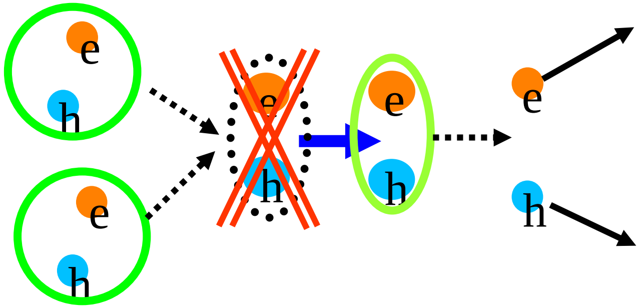
→ too slow conversion rate to accumulate paraexcitons

J. I. Jang et al. Phys. Rev. B **70,** 195205 (2004).

3) Large Auger recombination rate (nonradiative two-body decay)

$A=10^{-16}\,\mathrm{cm}^3/\mathrm{ns}$: Excitons decay before reaching the critical density $n_c=10^{17}\,\mathrm{cm}^{-3}$

$$\frac{\mathrm{d}n}{\mathrm{d}t} = -An^2$$



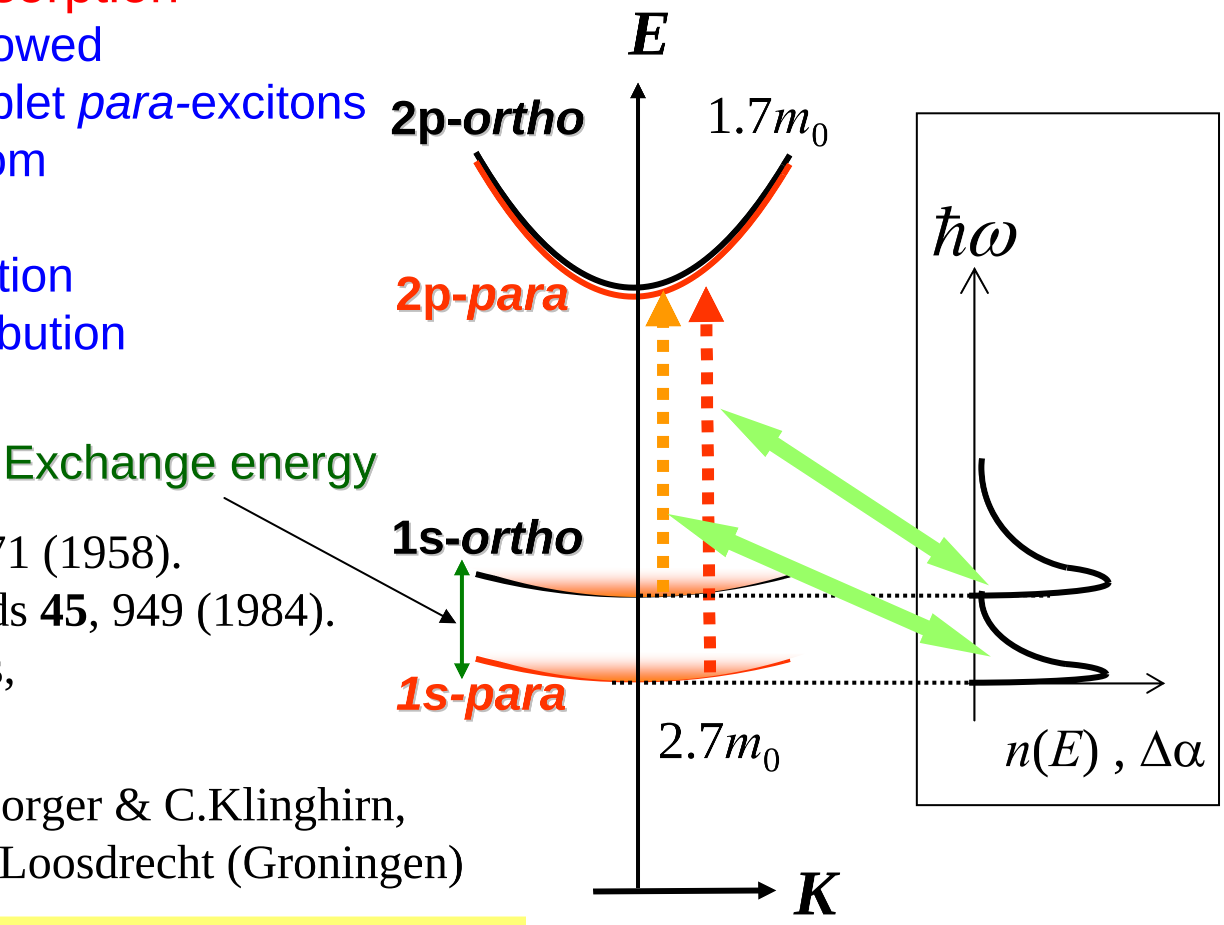
ionization due to recombination energy

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;
H. Haken, Fortschr. Phys. **38**, 271 (1958).
S. Nikitine, J. Phys. Chem. Solids **45**, 949 (1984).
K. Johnsen and G.M.Kavoulakis,
Phys. Rev. Lett. **86**, 858 (2001).

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

Our approach:
Time resolved Excitonic Lyman Spectroscopy

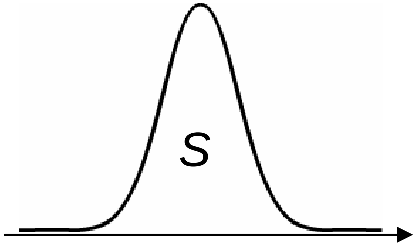
J. Phys. Soc. Jpn. 73, 1065 (2004).
Solid State Comm.134, 127 (2005).
Phys. Rev. Lett. 94, 016403 (2005).

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

$$\left(\tilde{n}(E,N_{ex})\right)^2=\varepsilon_b+\tilde{\chi}(N_{ex},E)$$

$$\tilde{\chi}(N_{ex},E)=N_{ex}\cdot\frac{2E_{1s-2p}}{\varepsilon_0}\frac{\left|\mu_{1s-2p}\right|^2}{\left(E^2-E_{1s-2p}^2-i\Gamma E\right)}$$

$$\Delta\alpha(E)=\frac{E}{\mathrm{h}c}\frac{1}{\sqrt{\varepsilon_b}}\mathrm{Im}\left\{\tilde{\chi}(N_{ex},E)\right\}$$



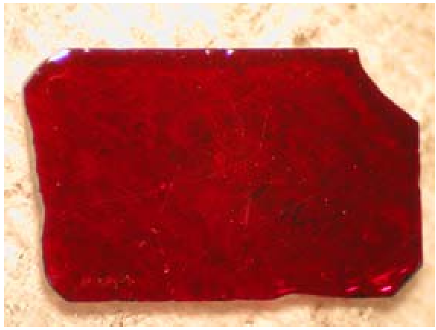
$$S\equiv\int\Delta\alpha(E)\,dE=N_{ex}\cdot\frac{\pi E_{1s-2p}\left|\mu_{1s-2p}\right|^2}{\hbar c\varepsilon_0\sqrt{\varepsilon_b}}$$

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

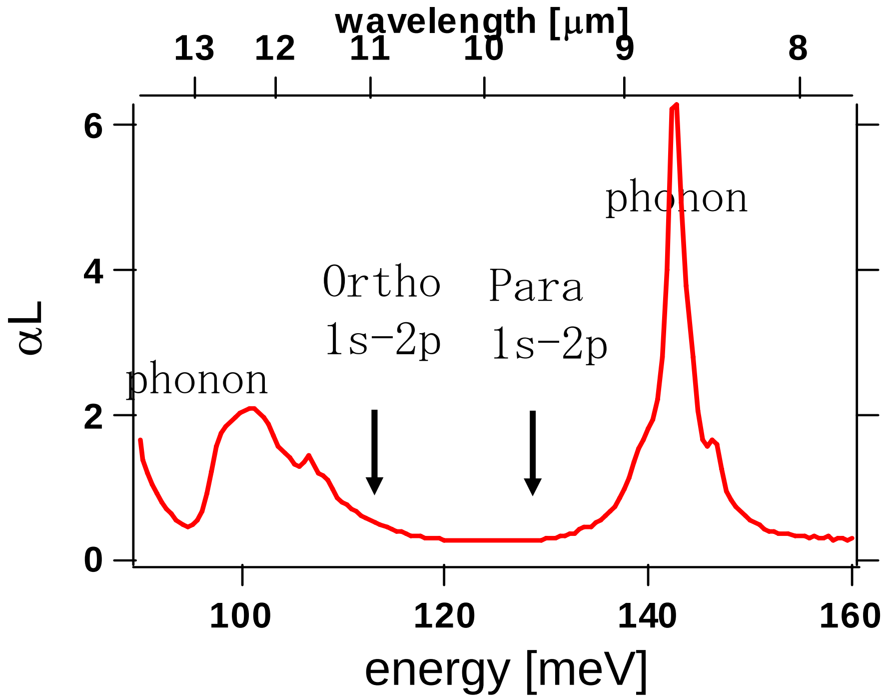
Sample

Cu_2O
naturaly grown
single crystal

3×5mm
Thickness 200 μm

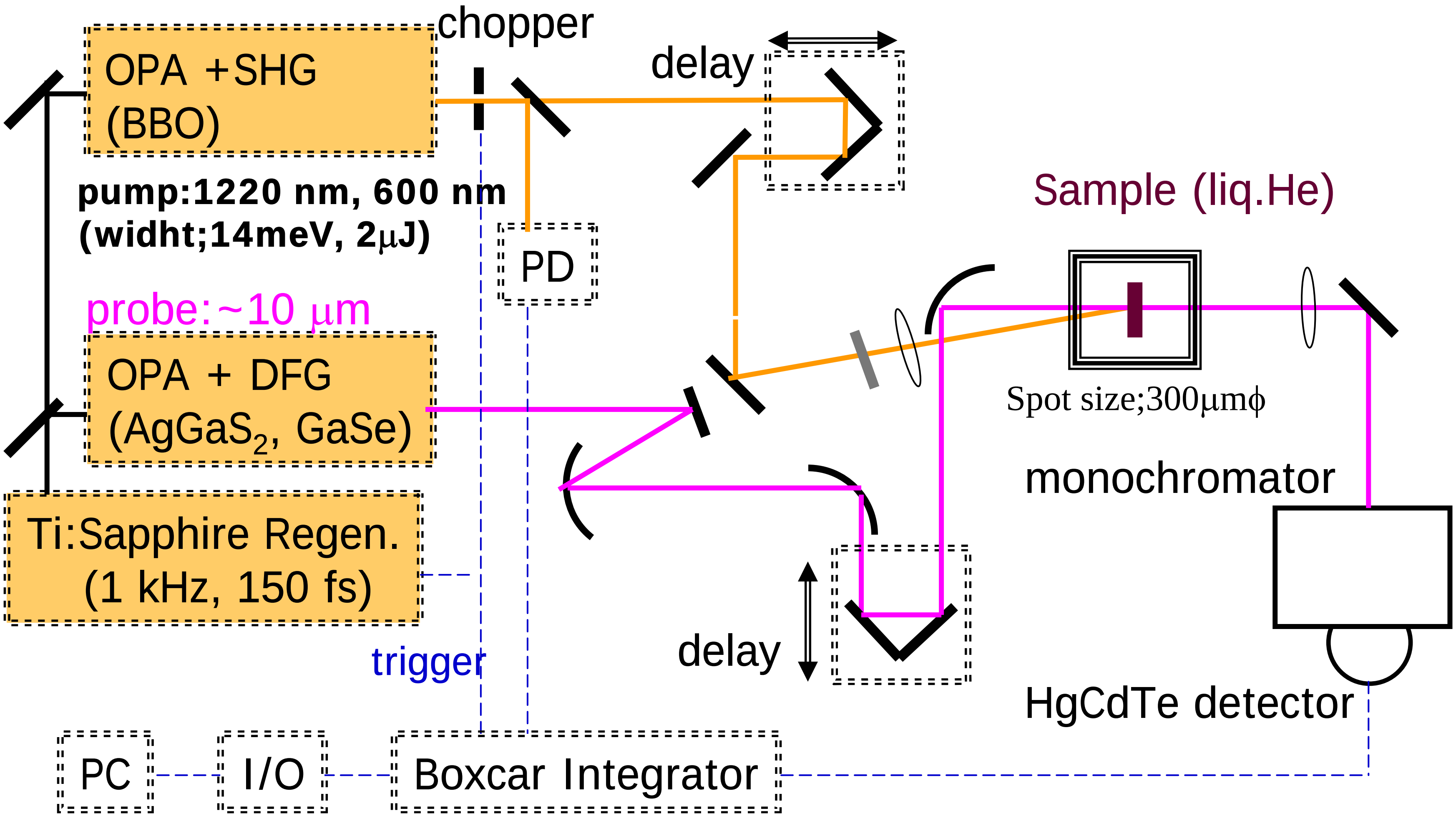


Mid-infrared linear absorption spectrum



transparent window near 1s-2p transition!

Experimental setup : mid-infrared pump-probe spectroscopy



OPA +SHG
(BBO)

chopper

delay

pump:1220 nm, 600 nm
(widht;14meV, 2µJ)

PD

Sample (liq.He)

probe:~10 µm

OPA + DFG
(AgGaS$_2$, GaSe)

Spot size;300µmϕ

monochromator

Ti:Sapphire Regen.
(1 kHz, 150 fs)

trigger

delay

HgCdTe detector

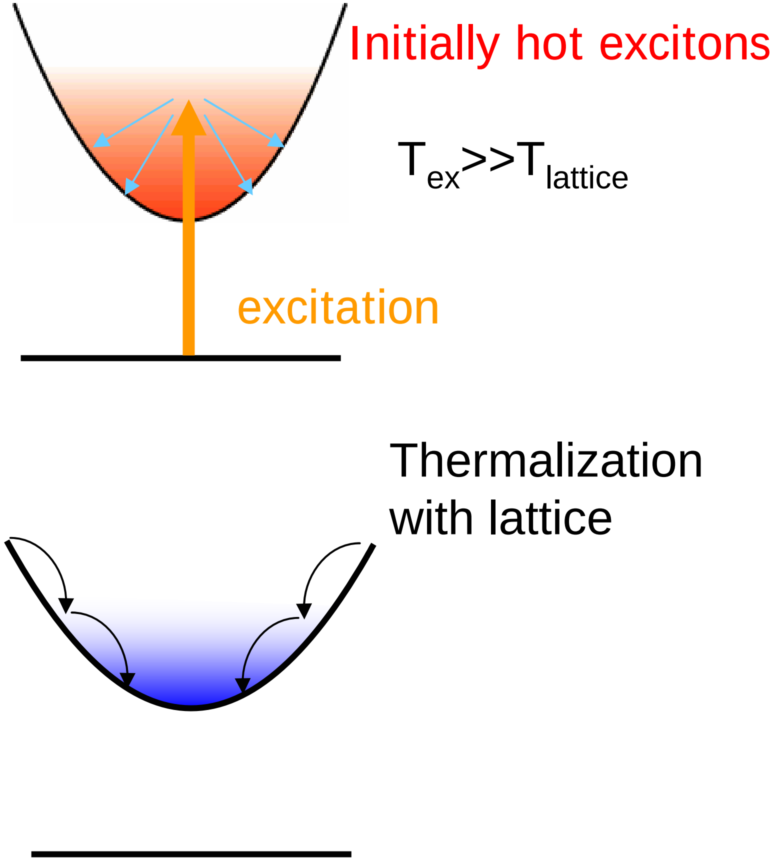
PC

I/O

Boxcar Integrator

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.

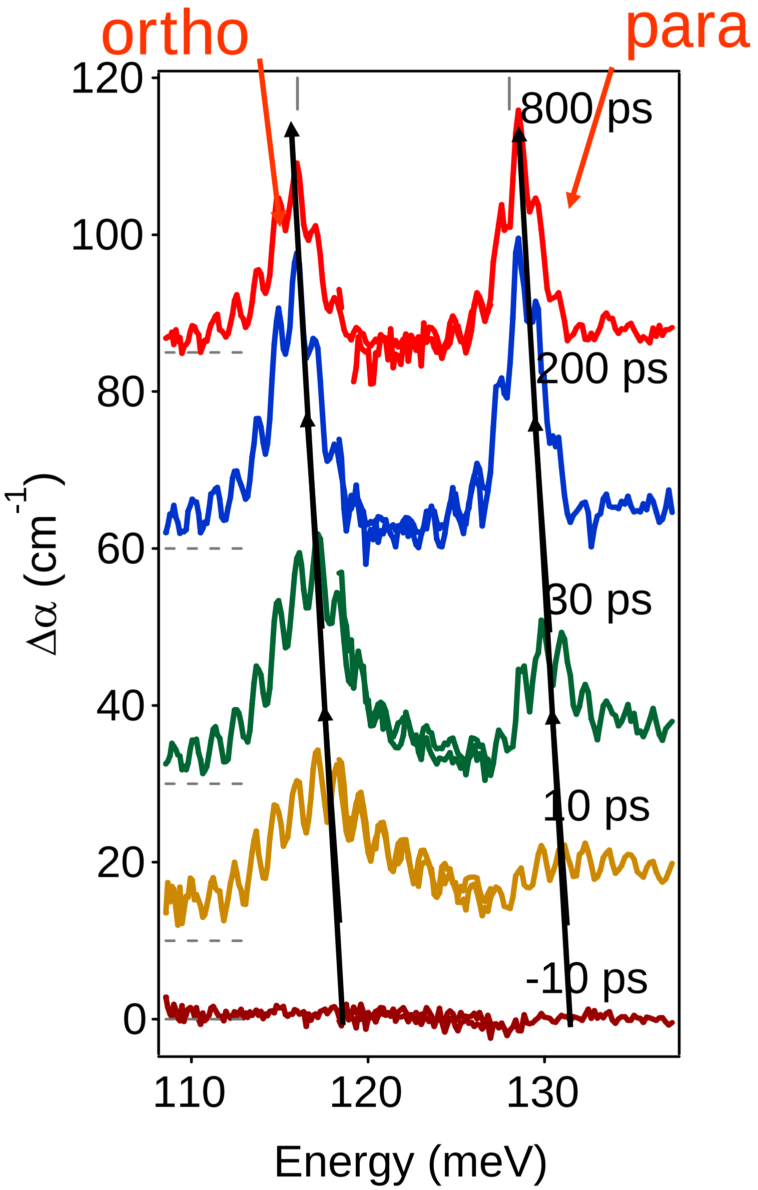


Initially hot excitons

$T_{ex} >> T_{lattice}$

excitation

Thermalization
with lattice

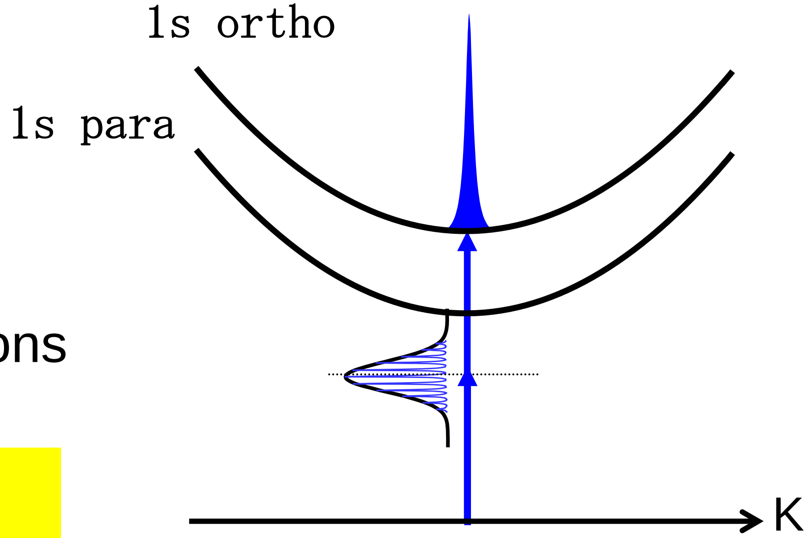


Direct excitation of cold orthoexcitons by TPA

Two-photon electric dipole transition of orthoexcitons is allowed.

$$\Gamma_4^- \times \Gamma_4^- = \Gamma_1^+ + \Gamma_3^+ + \Gamma_4^+ + \Gamma_5^+$$

orthoexcitons $k_o \sim 0$



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

⇨ Instantaneous preparation of Quantum degenerate ortho-excitons

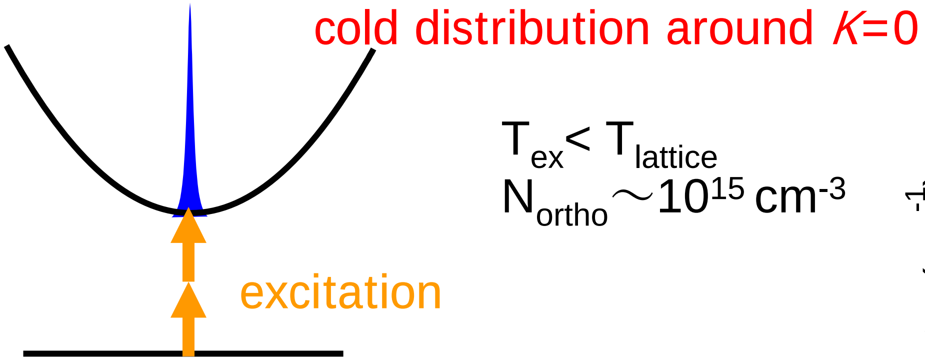
Large phase space density of photons in ML-fs laser

76MHz repetition, $\delta\lambda$ 2nm, 1mW
Photon number per mode; $n_\nu = 500$

★ ***M. Kuwata-Gonokami, et al.,***
J. Phys. Soc. Jpn., 71, (2002) 1257

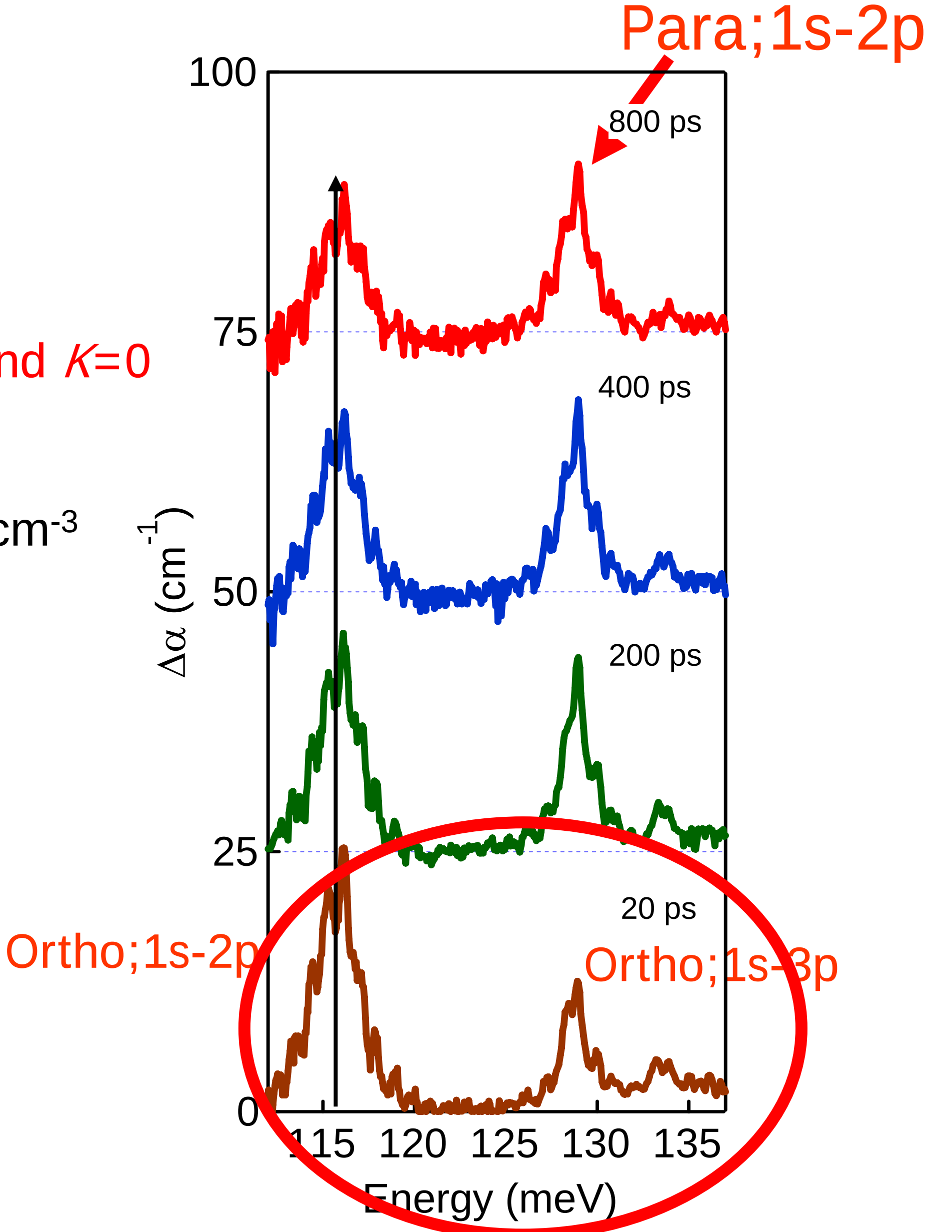
Resonant two-photon excitation of orthoexciton

No peak shift -> No excess heating



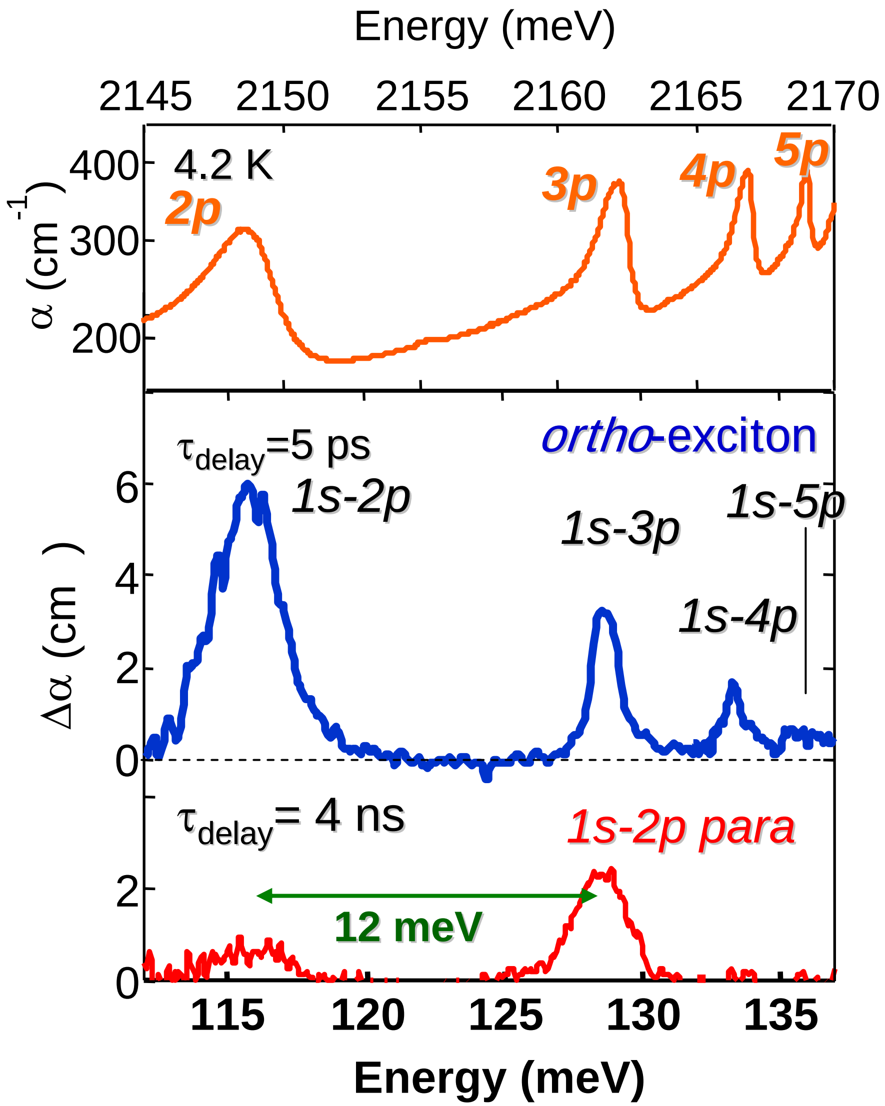
$T_{ex} < T_{lattice}$
$N_{ortho} \sim 10^{15}\ \mathrm{cm}^{-3}$

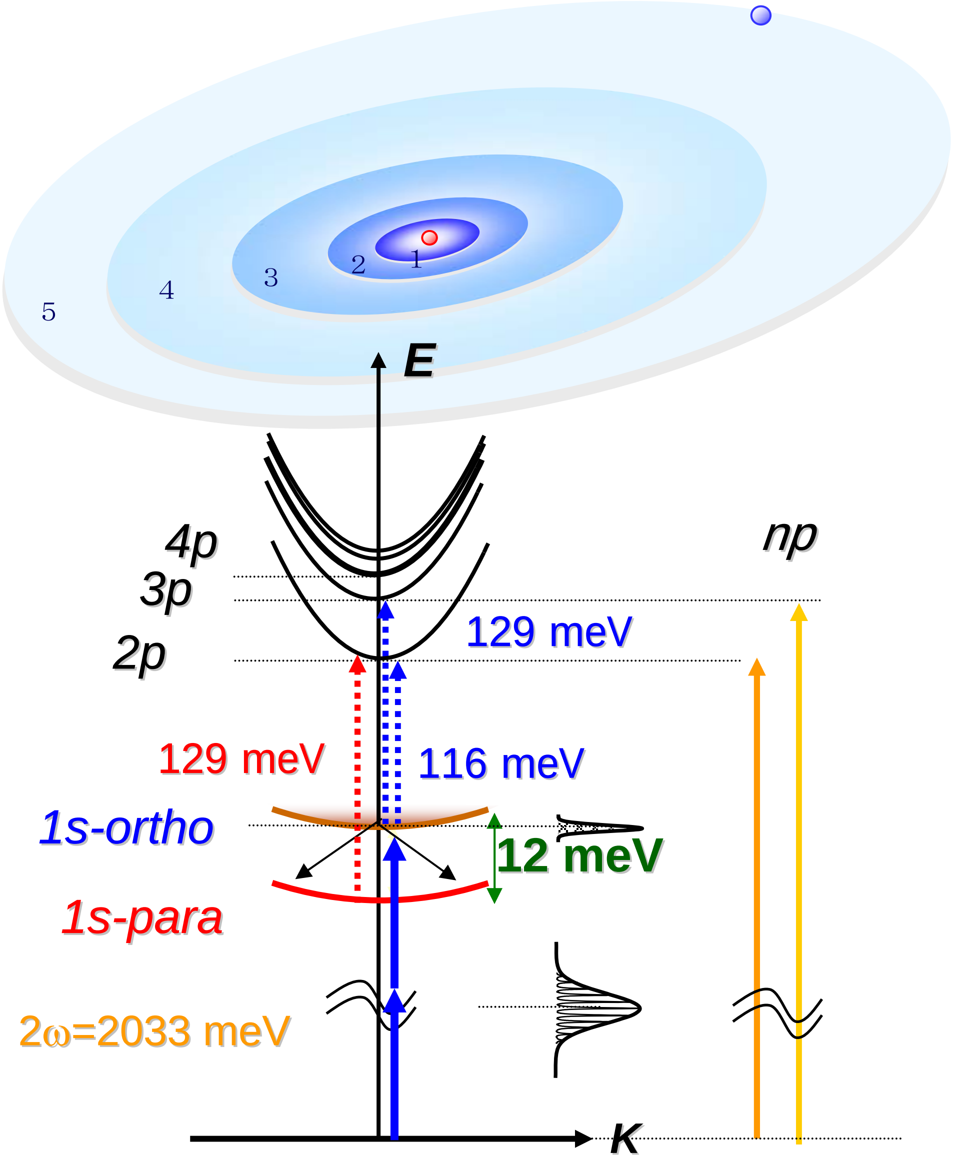
Rapid production of paraexcitons



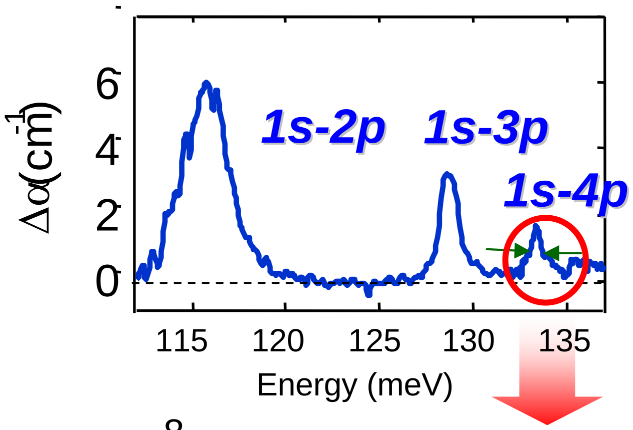
Observation of Excitonic Lyman series

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

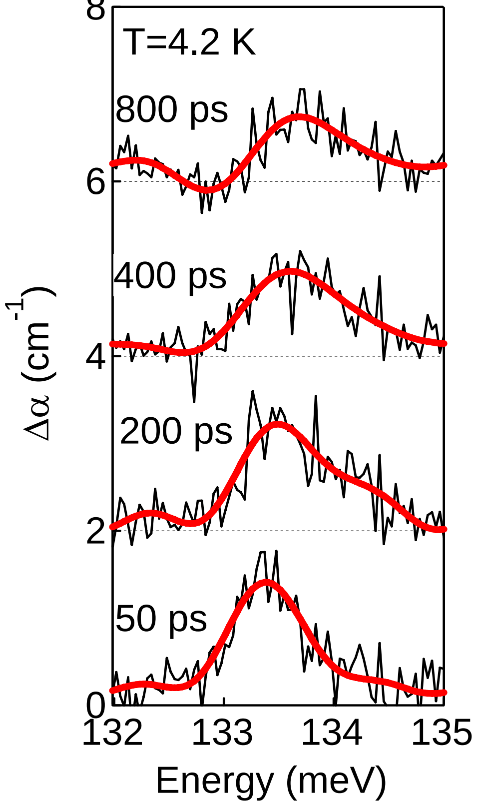




Thermalization dynamics of super-cooled 1s orthoexciton (4.2K)

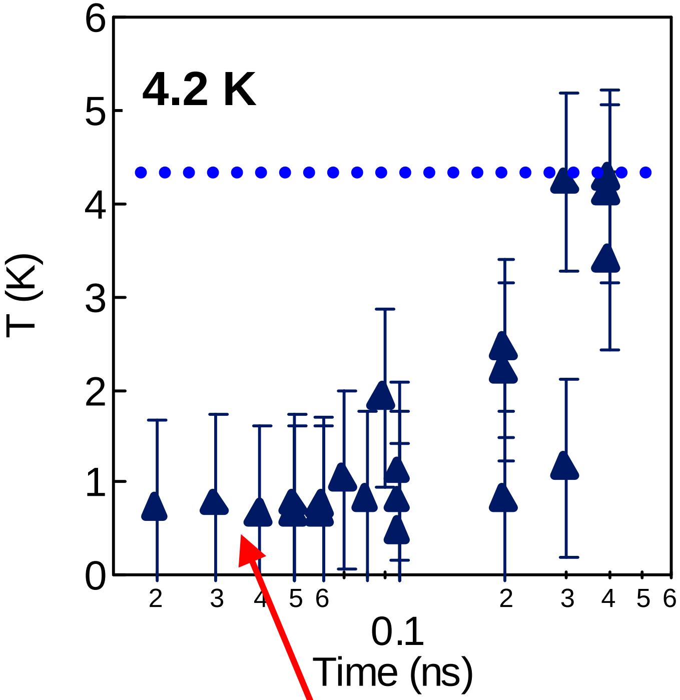


n~10^{15}



Line shape analysis:

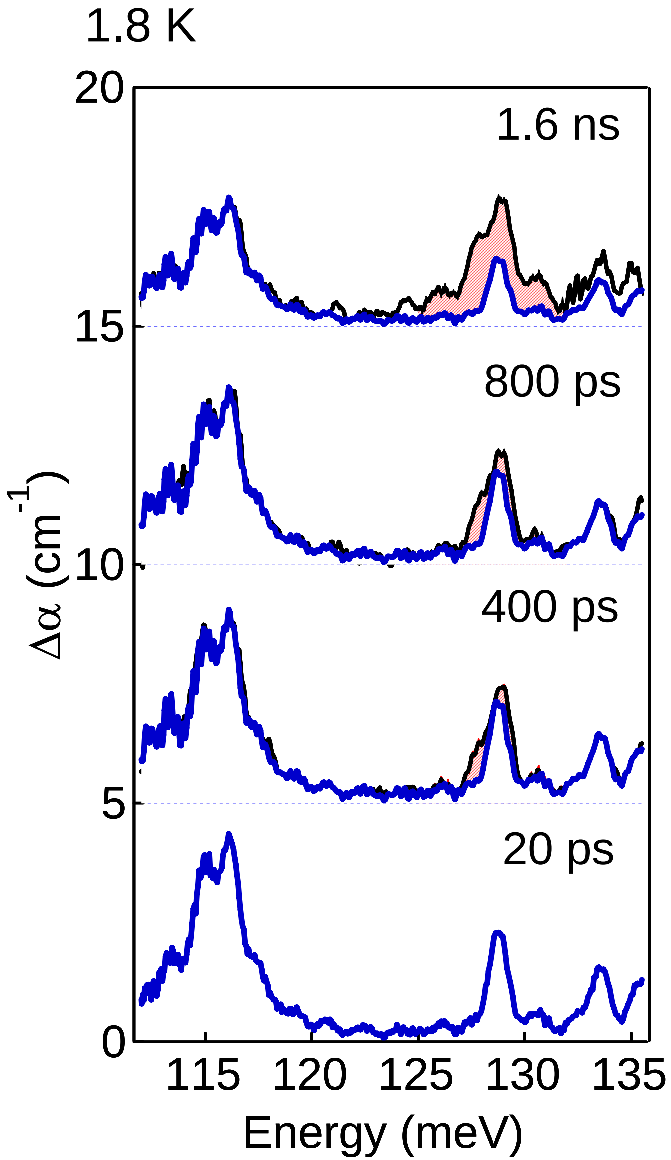
$$(\Delta E_{1s\text{-}4p})^2 \sim (\gamma_{4p})^2 + ((m_{1s}/m_{4p}-1)f_{1s})^2$$



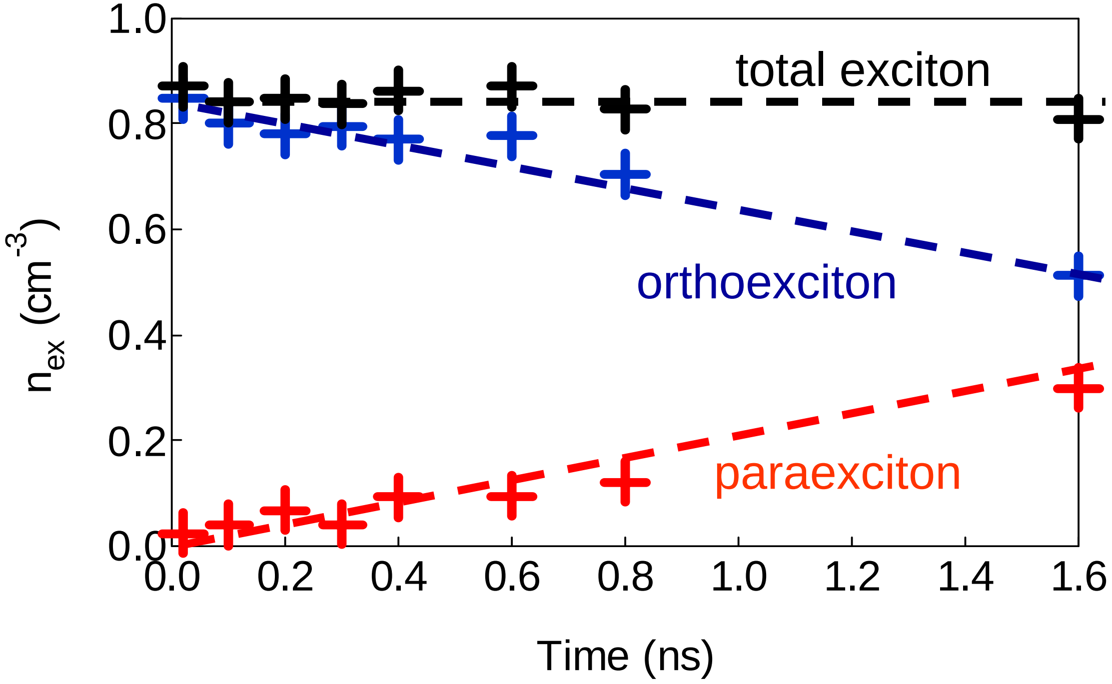
Boltzmann fitting

Super cooled state

Extraction of ortho-para conversion



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



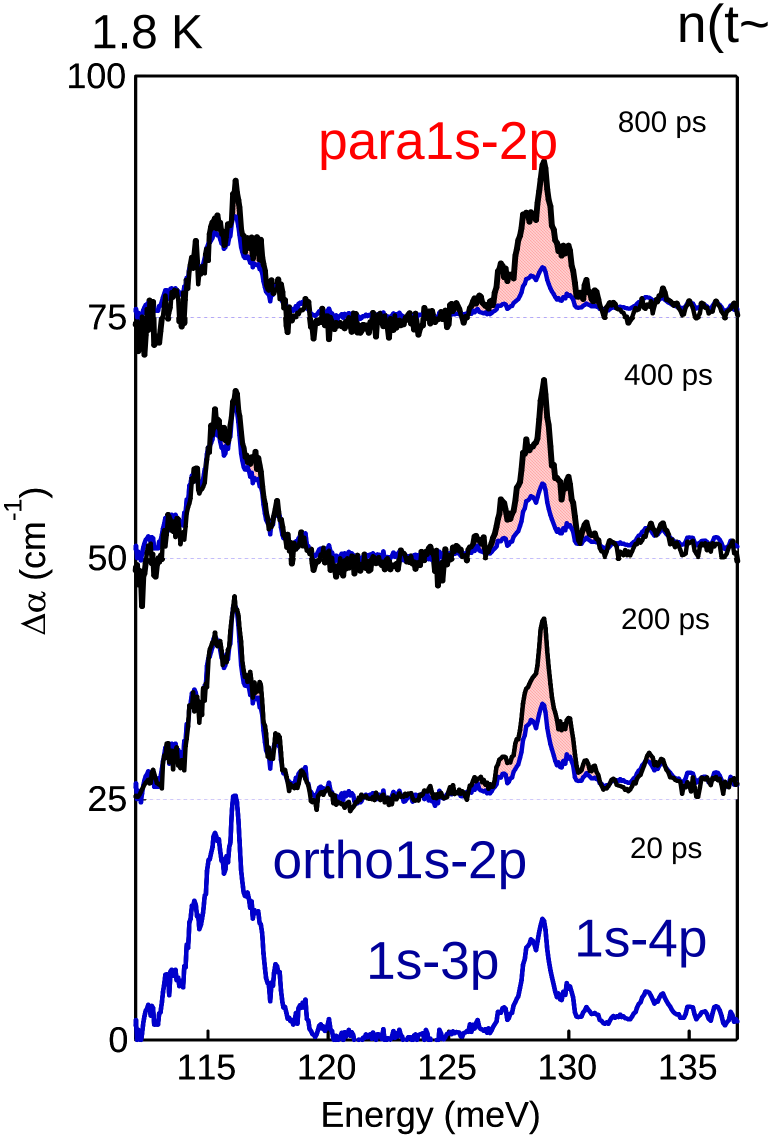
- Total exciton density conserves

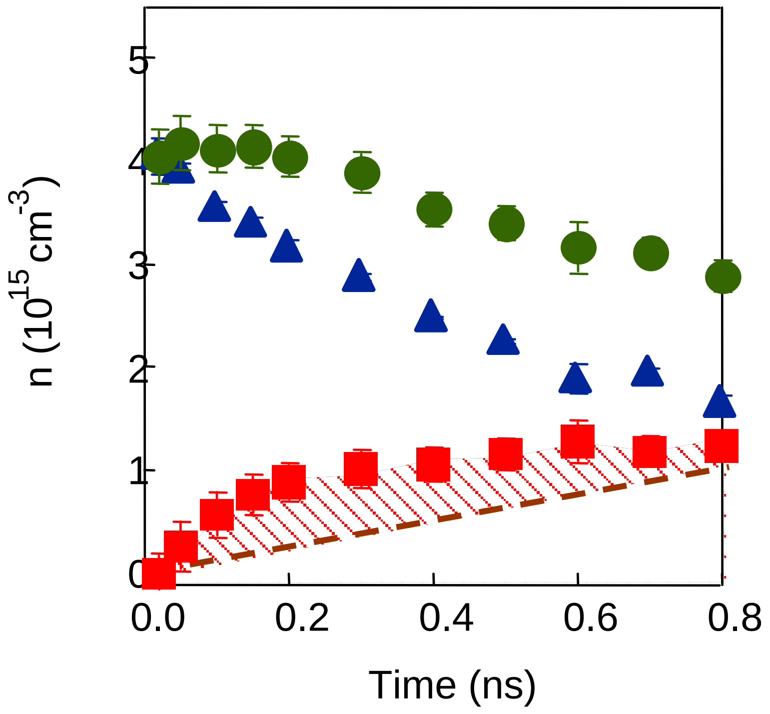
Ortho-para conversion: $\tau_{o\text{-}p}$=3 ns

TA-phonon assisted spin transformation

Temporal evolution of excitons ; High density excitation

$n(t\sim0)\sim4\times10^{15}\ \mathrm{cm}^{-3}$



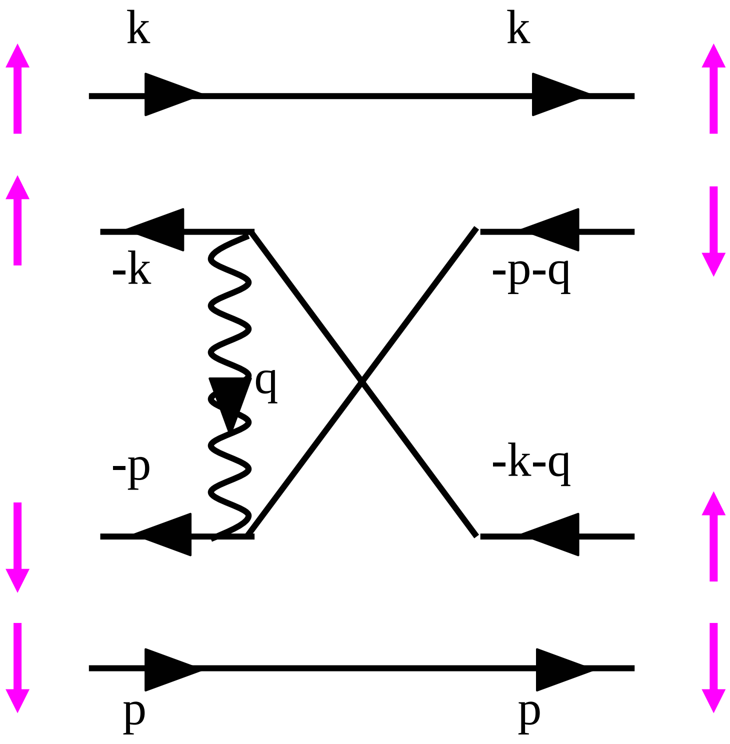


▨ conversion :Nonlinear ortho-to-paraexciton

– – – : Linear ortho-para conversion: $\tau_{o\text{-}p}=3$ ns

Collision induced ortho-para conversion

G. M. Kavoulakis and A. Mysyrowicz, Phys. Rev. B **61**, 16619 (2000).



$$\frac{\mathrm{d}n}{\mathrm{d}t} = -Cn^2$$

$$C \approx 5\times10^{-16}\, cm^3 / ns$$

Temporal evolution of n_{ortho}, n_{para}, n_{total}

Model:

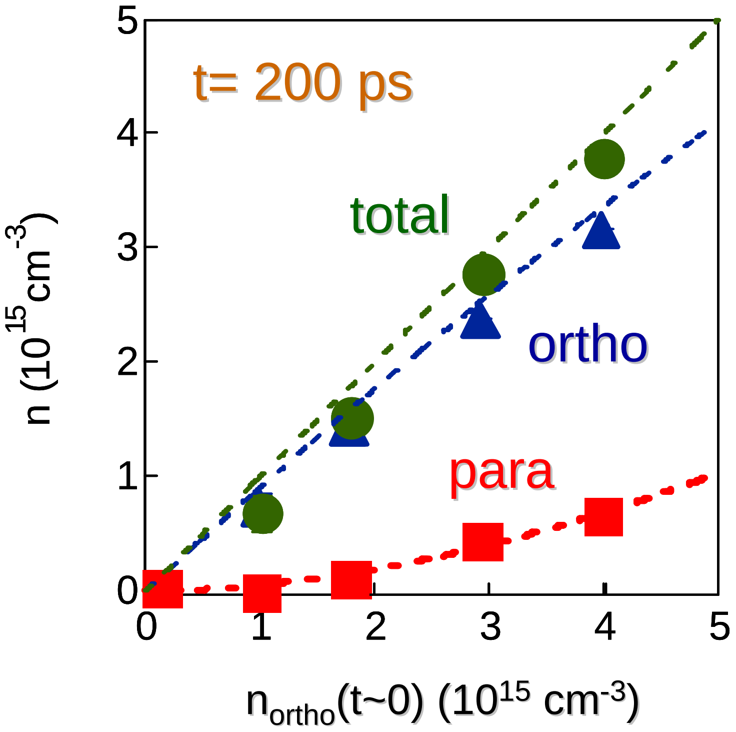
$$\frac{d}{dt}n_t = -An_t{}^2 \qquad n(t) = \frac{n_0}{1 + An_0 t}$$

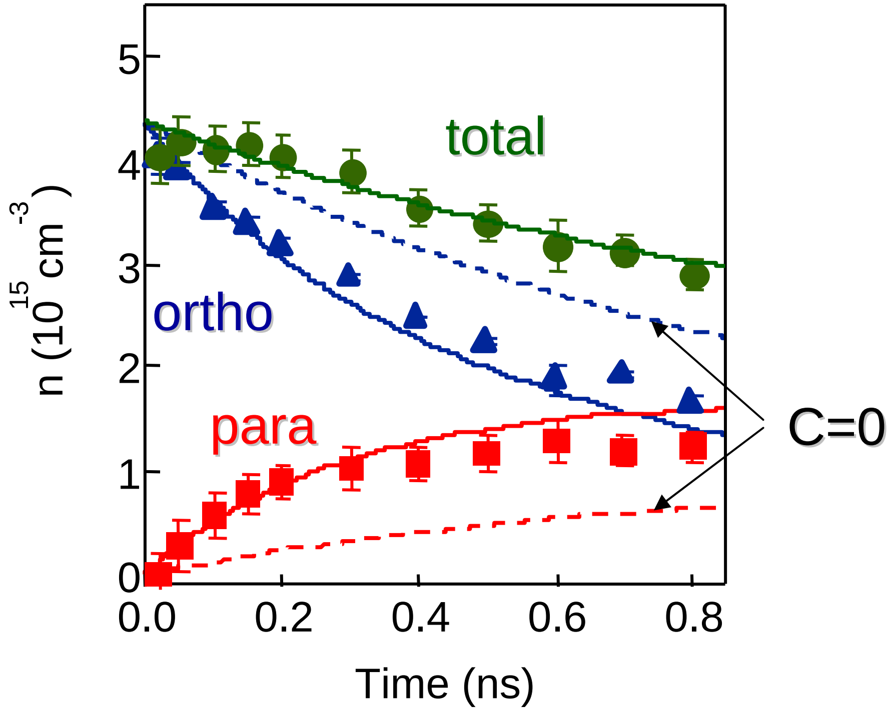
$$\begin{cases} \dfrac{d}{dt}n_O = -\Gamma_{o-p}n_o - 2An_on_t + \dfrac{3}{4}An_t^2 - Cn_o^2 \\ \dfrac{d}{dt}n_p = \Gamma_{o-p}n_o - 2An_pn_t + \dfrac{1}{4}An_t^2 + Cn_o^2 \\ (n_t = n_o + n_p) \end{cases}$$

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





collision-induced
spin-flip

$$\frac{d}{dt} n_{para} = -C n_{ortho}^{\ \ 2}$$

$$\frac{d}{dt} n_{toral} = -A n_{total}^{\ \ 2}$$

$(A=10^{-16}\ \mathrm{cm^3/ns})$

O'Hara and Wolfe, PRB 62, 12909 (2000).

$C=2.6\mathrm{x}10^{-16}\ \mathrm{cm^3/ns}$

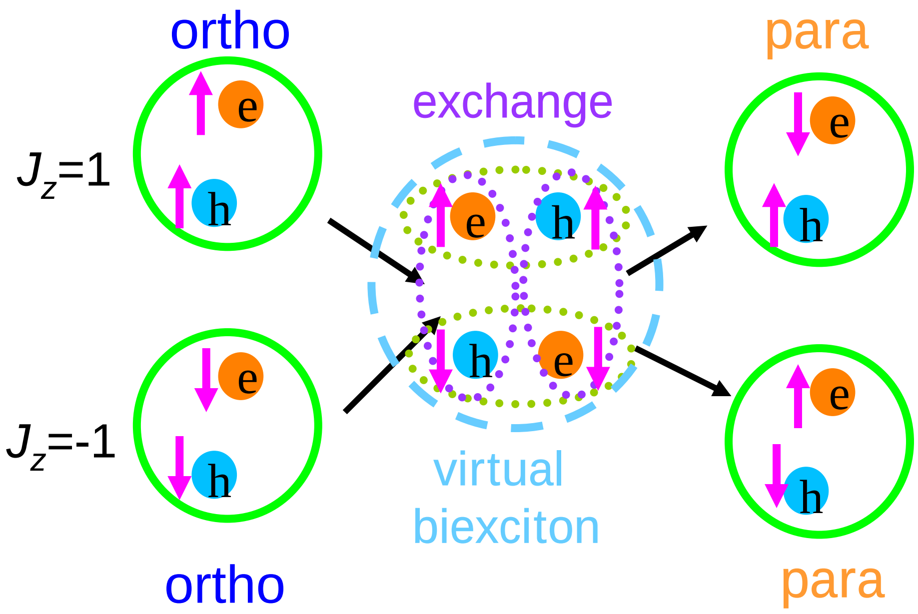
Kaovulakis *et al.* PRB 61, 16619 (2000).
$C=5\times10^{-16}\ \mathrm{cm^3/ns}$

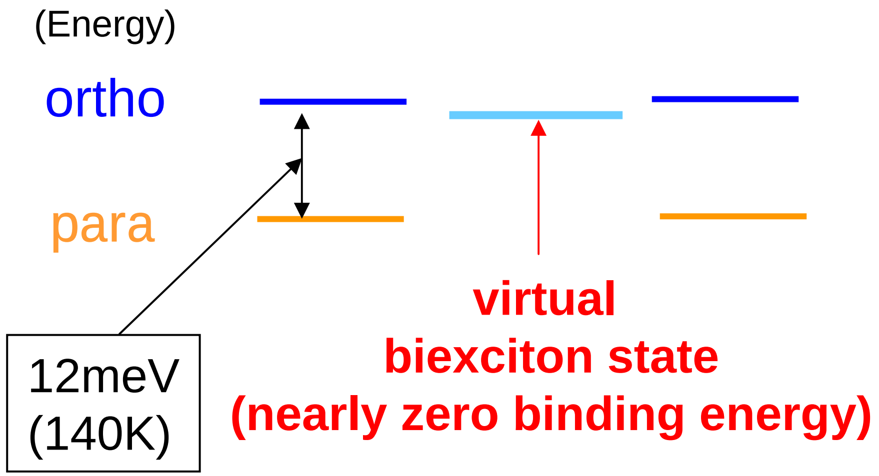
C > A

Enhanced collision induced spin conversion of excitons : Virtual biexciton mediated resonant scattering ?

Biexciton state in Cu_2O;
Large e-h exchange makes
Biexciton state unstable

F. Bassani and R. Rovere,
Solid State Commun., **19**, 887 (1976).

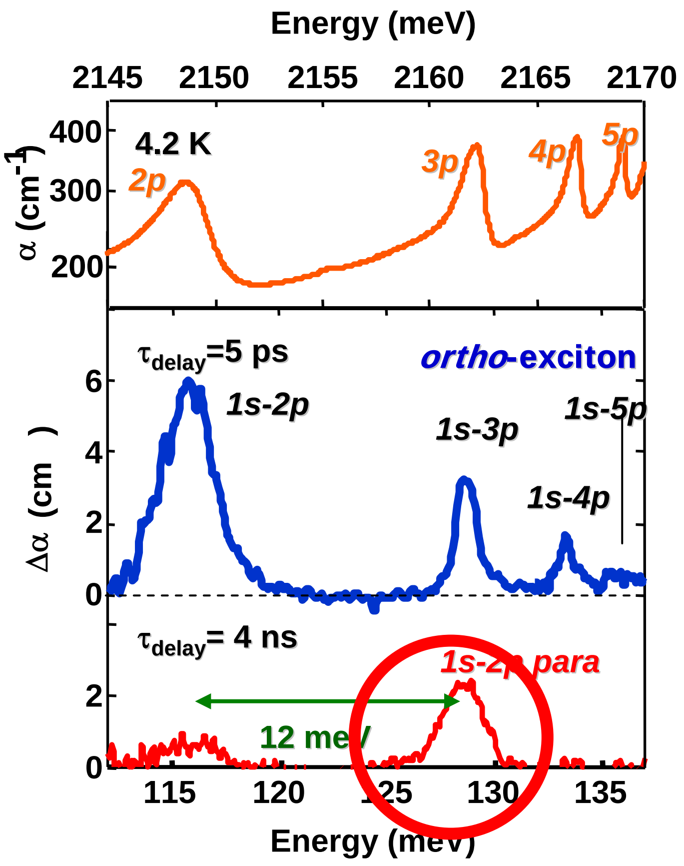




virtual
biexciton state
(nearly zero binding energy)

Resonant scattering
enhances collision cross section

Paraexcitons generated via TPA of orthoexcitons



Summary of femtosecond experiments

We obtain paraexciton density of 10^{15} cm^{-3} under orhtoexciton excitation of 4×10^{15} cm^{-3}

$T_{para} < 20$K

C ~ 2.6×10^{-16} cm^3/nsec

Questions:

Mechanism of giant collision cross-section ?

Why did we obtain cold paraexcitons ?

We need to

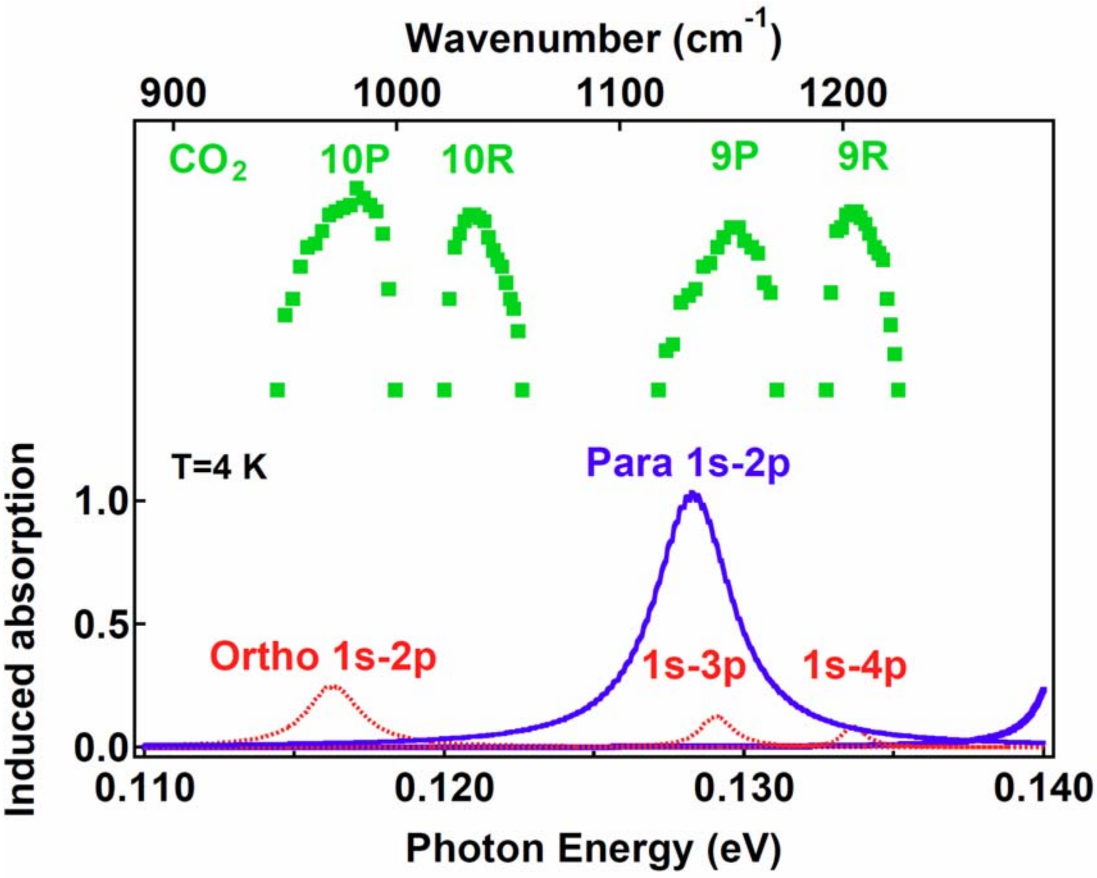
- Accumulate paraexcitons with continuous feeding at low lattice temperature.

- Precisely estimate
Auger rate and paraexciton life time

➡ **CW based experiment**

Excitonic 1*s*-*np* transitions and CO_2 laser lines

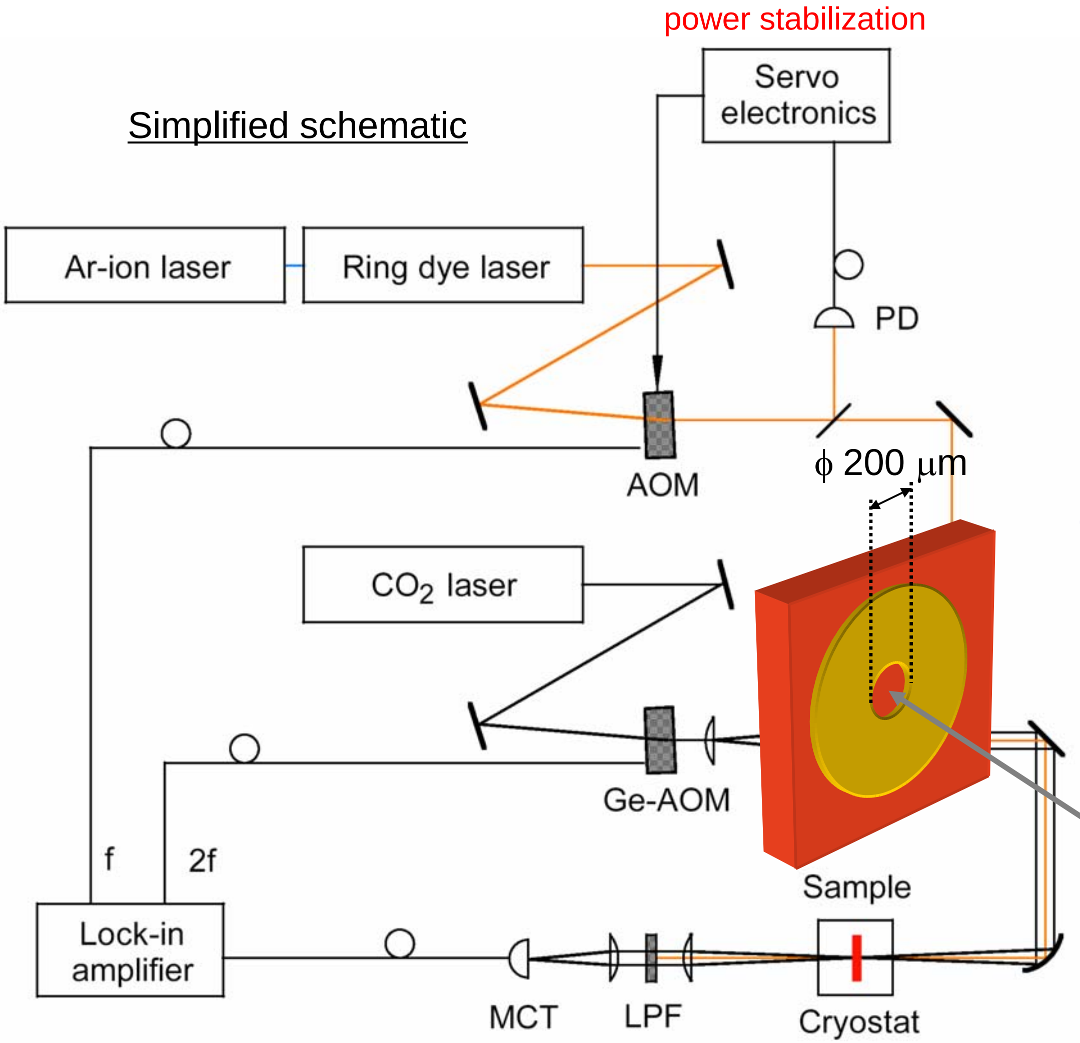
Quasi-steady state measurements for long lived paraexcitons



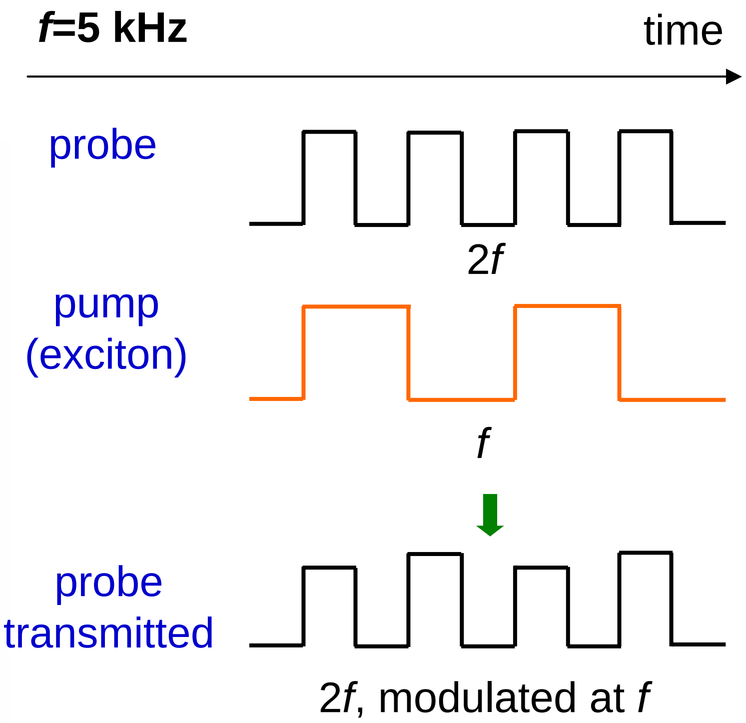
Accidental coincidence – Single mode tunable CO_2 laser to probe 1*s* paraexcitons

Experimental Set-up: Steady-state excitonic Lyman spectroscopy

Pump: single-mode dye laser (orthoexcitons)
Probe: single-mode tunable CO_2 laser

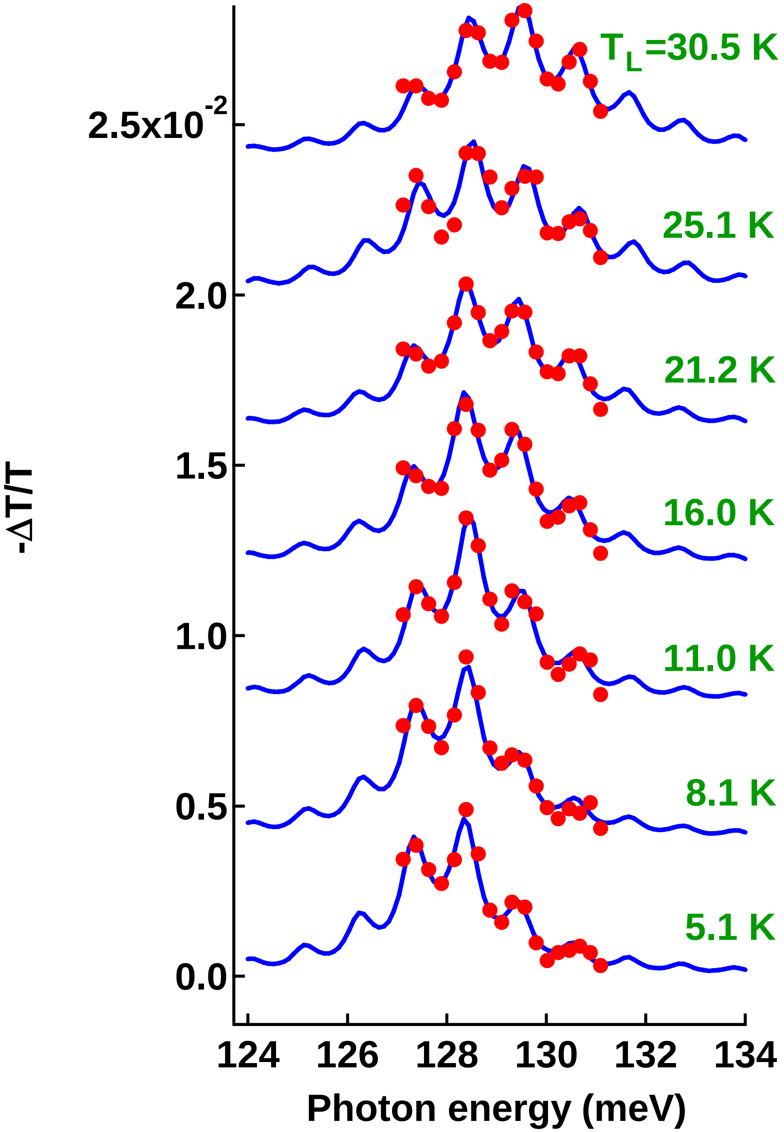


Simplified schematic



Induced absorption (-ΔT/T) can be obtained by measuring *f* & *2f* components of the probe beam using lock-in amplifier(s)

1*s*-2*p* absorption spectra of quasi-steady state paraexcitons



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

Exactly match theoretical curves assuming Maxwell-Boltzmann distribution functions

⬇

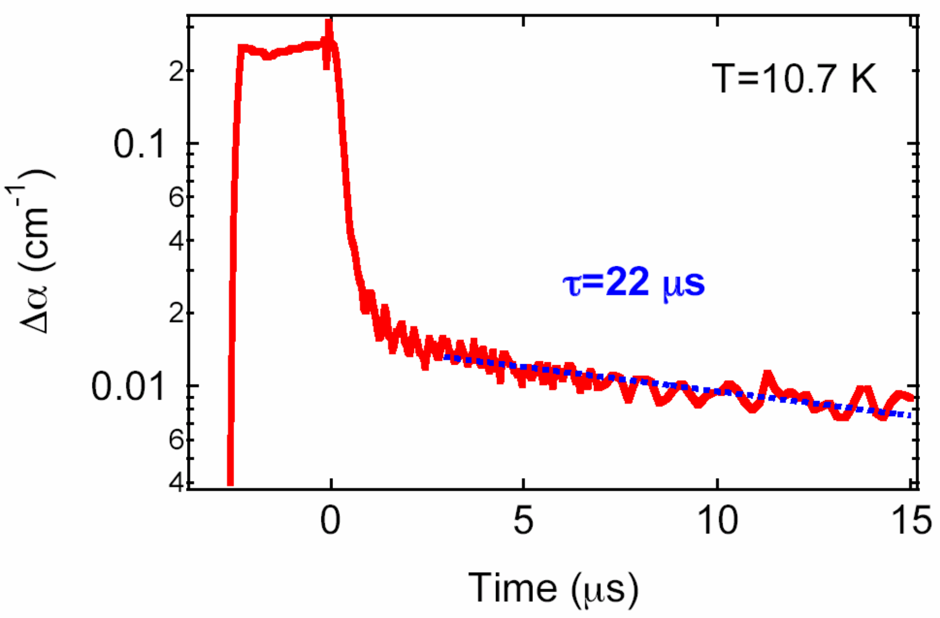
We successfully detected 1*s* paraexcitons in a steady state regime!

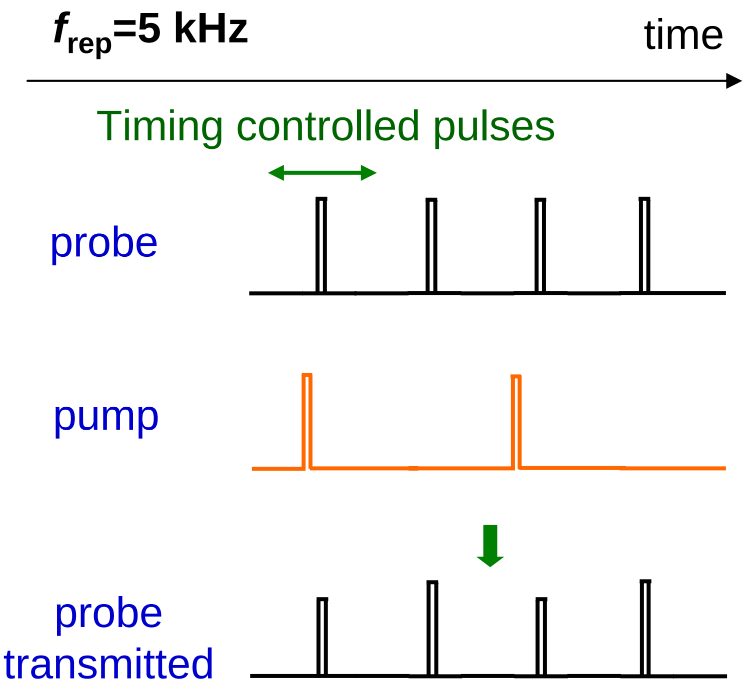
Due to the relative stability of the probe light, we are currently able to detect a transmission variation as small as 0.001 % (corresponds to <10^{12} cm^{-3})

Life time measurement of paraexcitons

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

Lifetime measurement of 1*s* paraexcitons by CW Lyman spectroscopy





***f*$_{rep}$=5 kHz**

time

Timing controlled pulses

probe

pump

probe transmitted

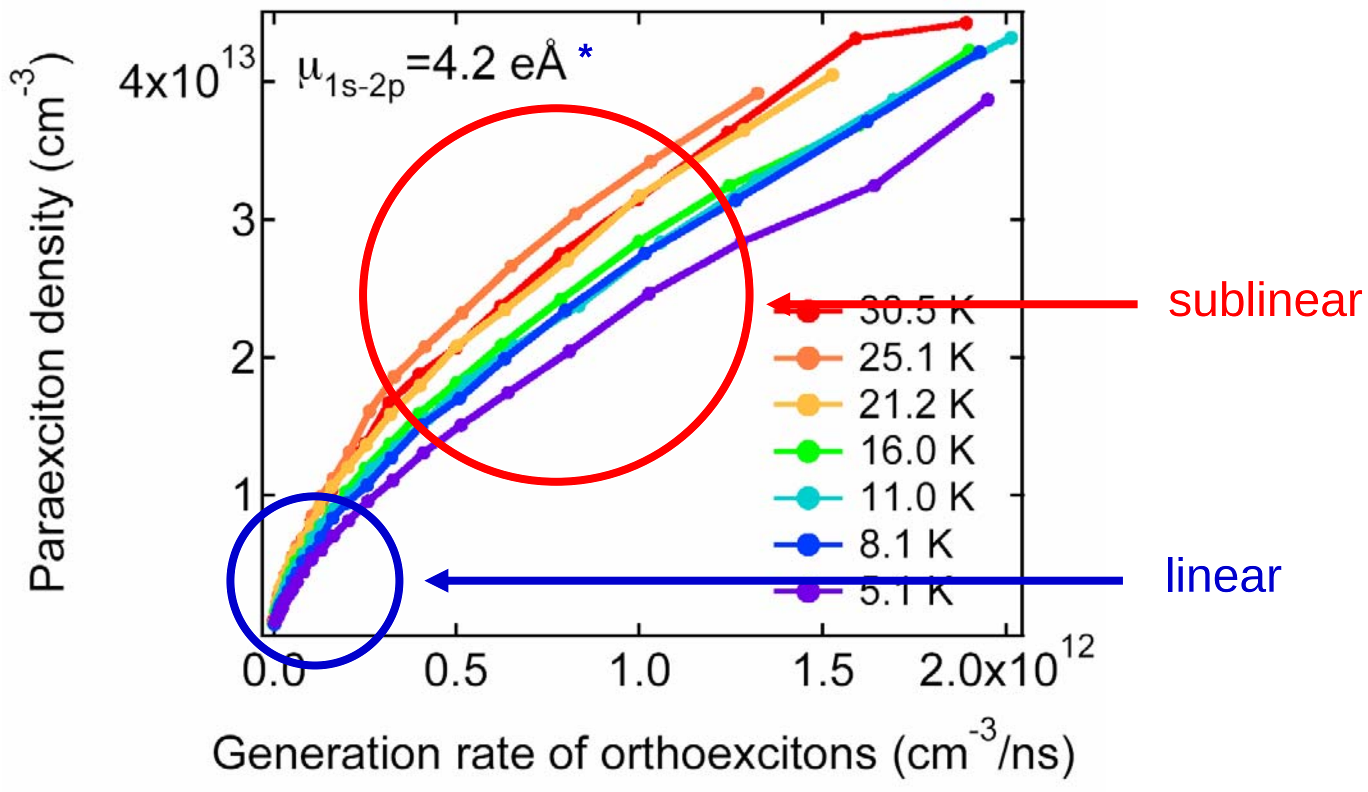
We measure the probe pulse transmission and evaluate the induced transmission change.

*S. Denev et al., Phys. Rev. B **65**, 085211 (2002).
A. Jolk *et al*., Phys. Rev. B **65**, 245209 (2002).
J. P. Wolfe *et al*., Solid State Commun. **134**, 143 (2005).

Excitation intensity dependence of paraexciton density

Sublinear dependence on excitation intensity

➡ Auger effect is also observed in this steady-state regime



* T. Tayagaki *et al*., J. Phys. Soc. Jpn. 74, 1423 (2005).

Temperature dependence of Auger recombination rate

Numerical simulation

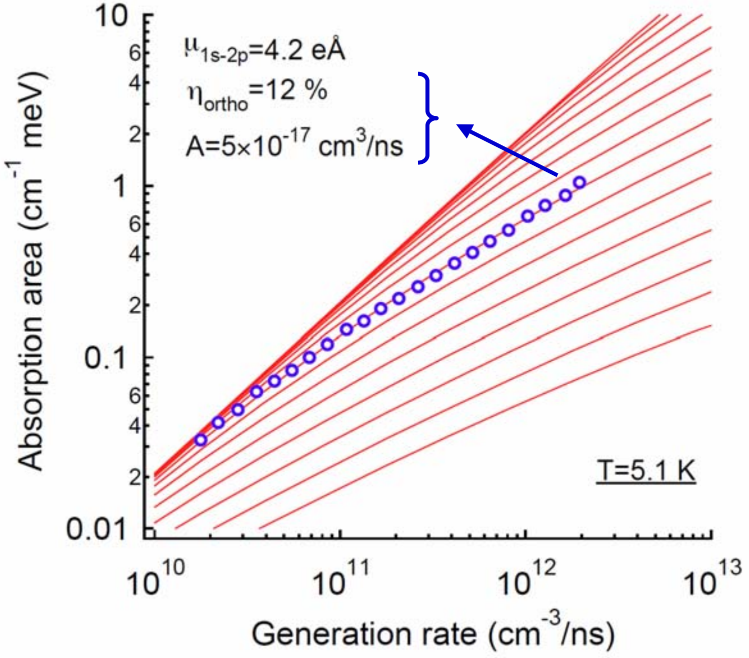
Temperature-dependent diffusion of excitons (2D)

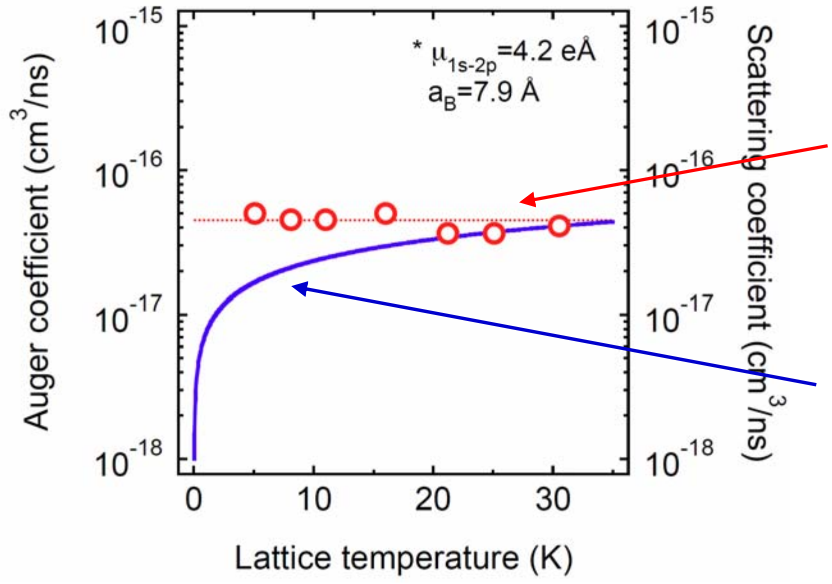
plus

Nonlinear rate equations (Auger recombination)

Temperature dependence of the nonlinear particle loss rate

*Collision-induced spin-frip processes are not included

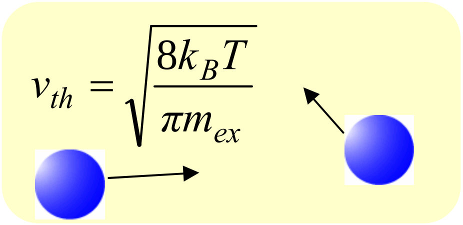




Independent of temperature

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

$$\tau^{-1} = \sigma v_{\mathrm{th}} \cdot n_{ex}$$



$$v_{th} = \sqrt{\frac{8k_B T}{\pi m_{ex}}}$$

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.

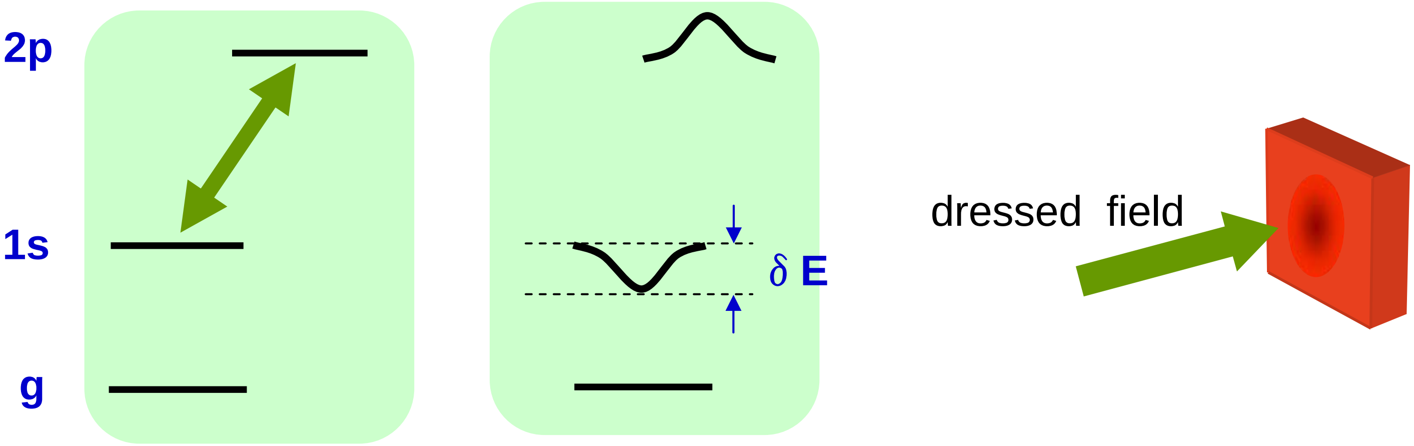
2) Excitonic Lyman series of super-cooled orthoexciton was observed.

   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 .



$$\delta E = \frac{1}{2}\left(\sqrt{\Delta^2 + (\mu_{1s-2p}E)^2} - \Delta\right)$$

$$\approx \frac{1}{2\Delta}(\mu_{1s-2p}E)^2 \propto I$$

μ_{1s-2p} (transition dipole moment) = 4.2 eÅ

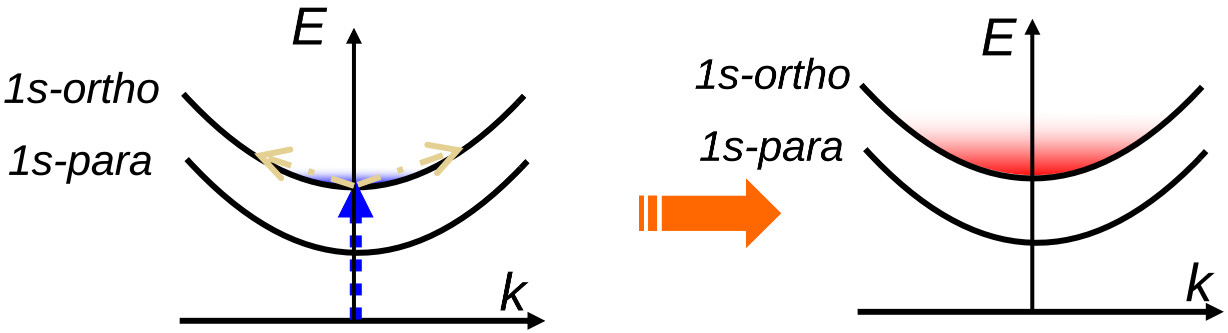
Δ (detuning) = 1 meV

I (Intensity) =20MW/cm^2

→ **δE = 0.8 meV**
(corresponds to 2K)

Sympathetic cooling under high-density excitation

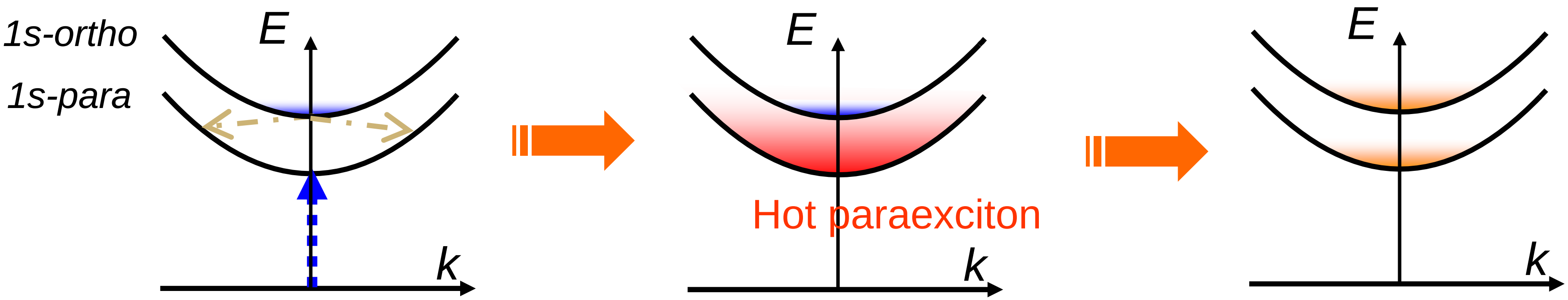
Phonon scattering



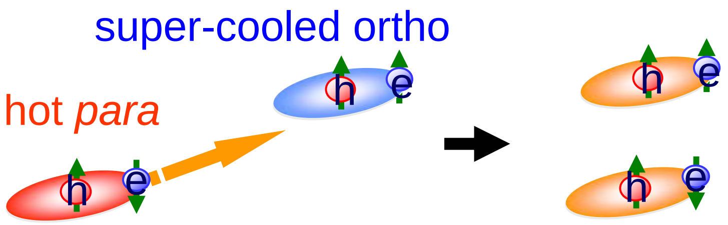
experimental:
$\tau \sim 300$ ps

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

Sympathetic cooling

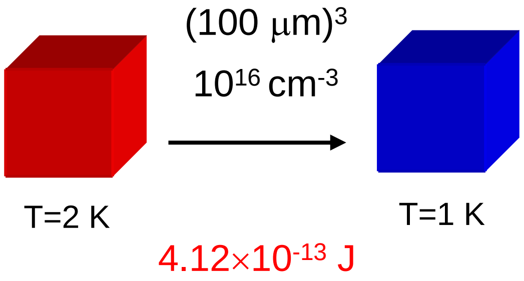


ortho-to-paraexciton conversion



How to cool paraexcitons ?

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



(100 μm)^3
10^{16} cm^{-3}

T=2 K

T=1 K

4.12×10^{-13} J

- Sympathetic cooling with super cooled orthoexciton gas

10^{16} excitons in V=(100 μm) ^{3} T=0 K → T=1 K

*classical gas

Specific heat
$3k_B/2\ N_{ex}$

- Heat exchange with the lattice

V=(100 μm) ^{3} lattice T=0.98 K → T=1 K

Specific heat
$^*36T^3$ Jm^{-3}K^{-4}

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

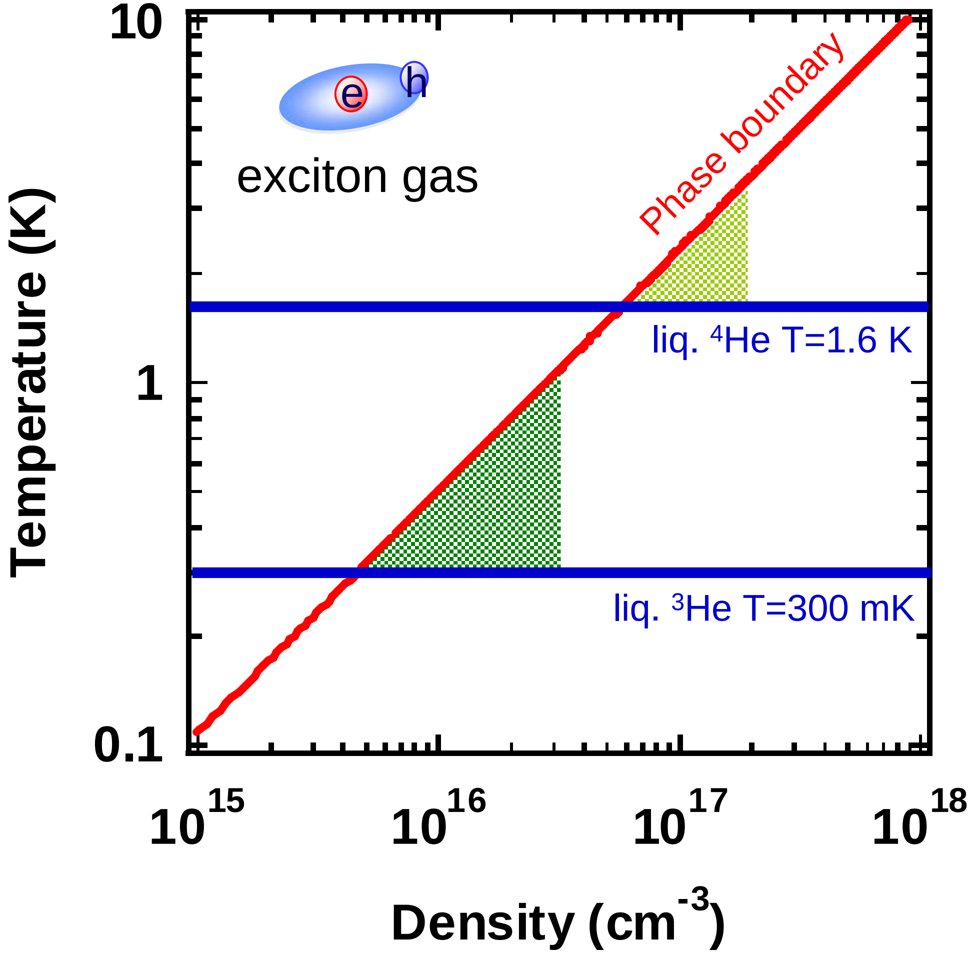
We need to cool down the crystal.

*L. V. Gregor, J. Phys. Chem. 66, 1645 (1962)., K. E. O'Hara and J. P. Wolfe, Phys. Rev. B 62, 12909 (2000).

How to reach excitonic BEC phase ?

$n = 1\times10^{17}\ \mathrm{cm^{-3}}$ $T_C = 2.3$ K

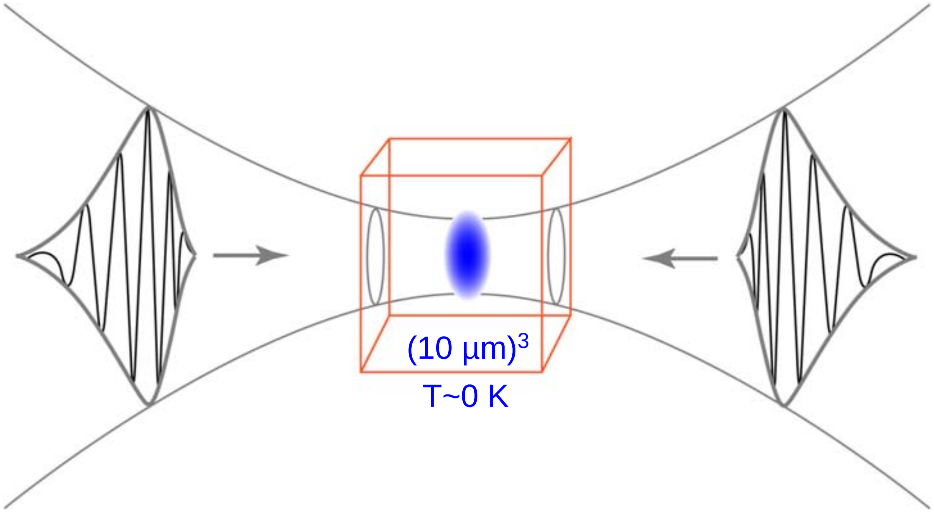
$n = 1\times10^{16}\ \mathrm{cm^{-3}}$ $T_C = 500$mK $(m_{ex} = 2.7m_e.)$



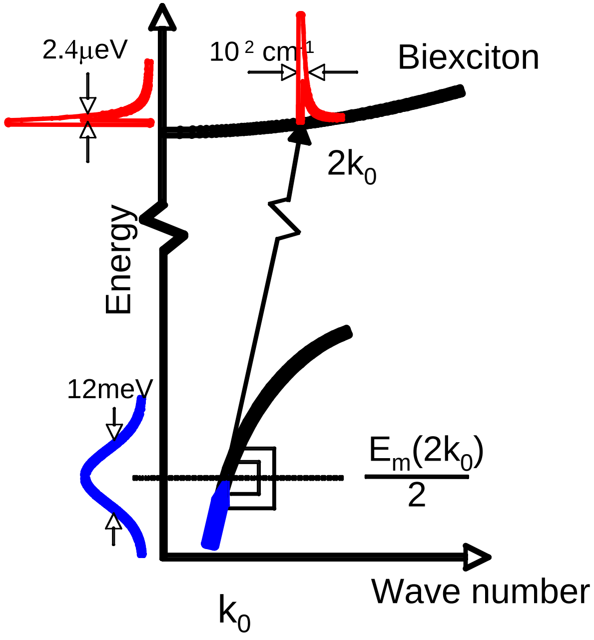
^{3}He refrigerator

Excitonic BEC
created by spatial-temporal controlled optical pulses

CREST-JST: Oct. 2006~



＜Phase space compression scheme＞



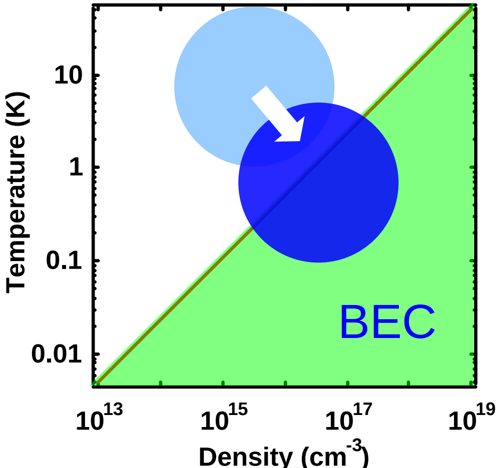
Efficient creation of ultracold excitons by chirp-controlled optical pulses
（Frequency Control）

Spatial confinement of ultracold excitons by polarization and wavevectror controlled optical pulses
（Spatial Control）

Excitonic BEC

・

Macroscopic Coherence



Next:

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