LECTURE 8

Exciton condensation

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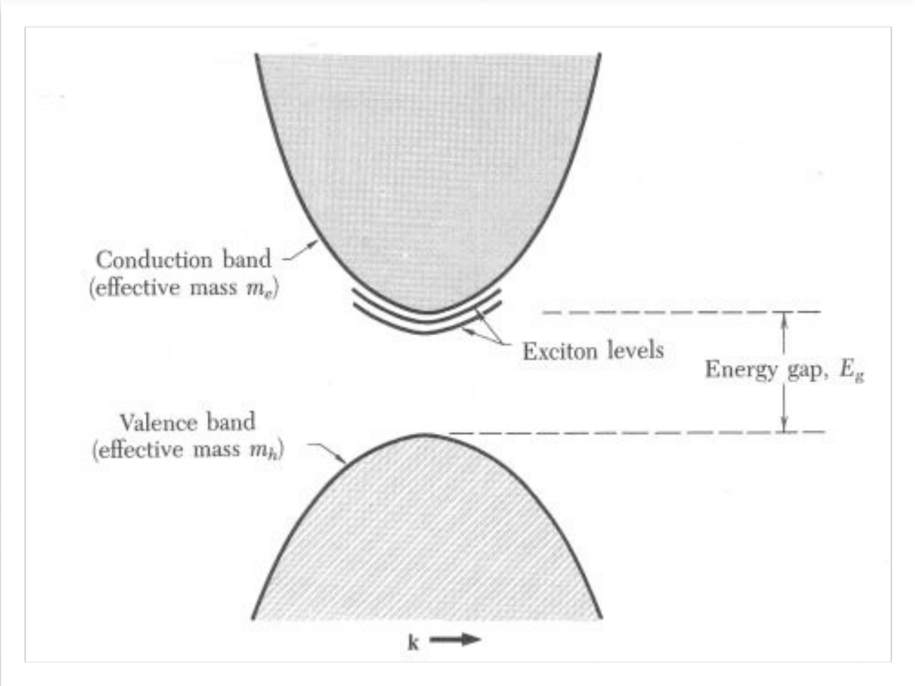
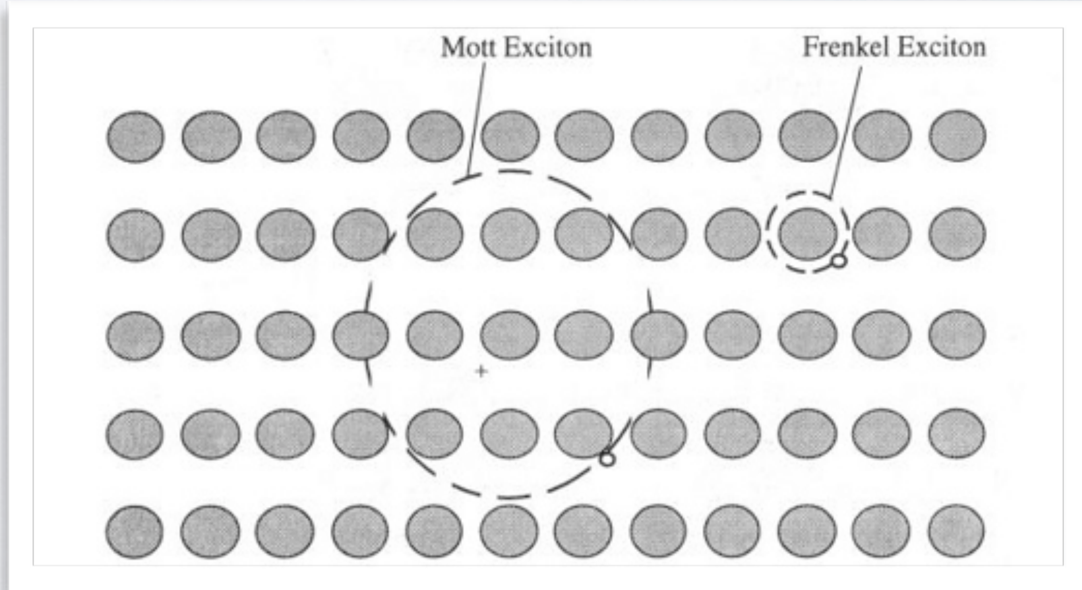
Excitons

An exciton is a bound state of an electron and an imaginary particle called an electron hole in an insulator or semiconductor.



- Quasiparticles: electrons and holes with half-integer spin
- The overall charge for this quasiparticle is zero.
- It carries no electric current.
- ! It is a composite BOSON !



http://electrons.wikidot.com/solar-cells

Excitons - 3D

Consider an electron-hole pair bound by the coulomb interactions:

$$-\frac{\hbar^2}{2\mu}\nabla^2 f(r) - \frac{e^2}{4\pi\varepsilon\varepsilon_0 r} f(r) = E f(r)$$

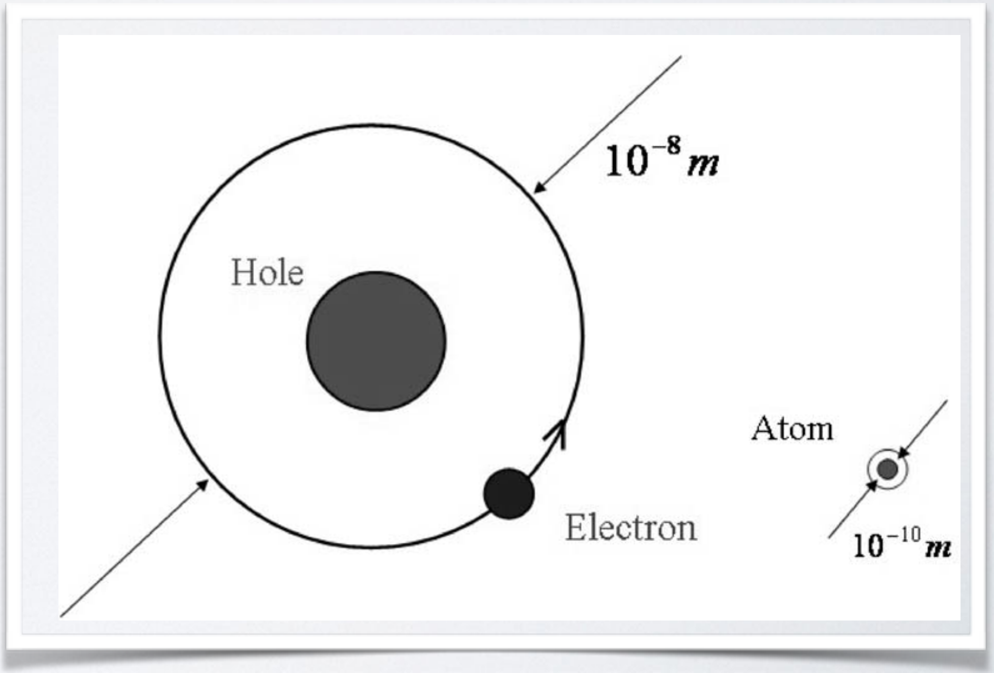
dielectric constant of a crystal

effective mass: $\mu = m_e m_h/(m_e + m_h)$

e-h distance: $r = \sqrt{x^2 + y^2 + z^2}$

Equation is analogous to Schrodinger equation for a hydrogen atom with the following renormalisations:

$$m_0 \to \mu, \qquad e^2 \to e^2/\varepsilon$$



A. Kavokin, et al. in *Microcavities*, Oxford Science Publications (2007)

Excitons - 3D

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$$-\frac{\hbar^2}{2\mu}\nabla^2 f(r) - \frac{e^2}{4\pi\varepsilon\varepsilon_0 r} f(r) = E f(r)$$

dielectric constant of a crystal

Bohr radius : $$a_{\mathrm{B}} = \frac{4\pi\hbar^2\varepsilon\varepsilon_0}{\mu e^2}$$

Binding energy of a ground state: $$E_{\mathrm{B}} = \frac{\mu e^4}{(4\pi)^2 2\hbar^2\varepsilon\varepsilon^2} = \frac{\hbar^2}{2\mu a_{\mathrm{B}}^2}$$

Wave-function of the 1*s* state: $$f_{1s} = \frac{1}{\sqrt{\pi a_{\mathrm{B}}^3}} e^{-r/a_{\mathrm{B}}}$$

A. Kavokin, et al. in *Microcavities*, Oxford Science Publications (2007)

Excitons - 3D

Semiconductor crystal	E_g (eV)	m_e/m_0	E_B (eV)	a_B (Å)
PbTe*	0.17	0.024/0.26	0.01	17 000
InSb	0.237	0.014	0.5	860
$Cd_{0.3}Hg_{0.7}Te$	0.257	0.022	0.7	640**
Ge	0.89	0.038	1.4	360
GaAs	1.519	0.066	4.1	150
InP	1.423	0.078	5.0	140
CdTe	1.606	0.089	10.6	80
ZnSe	2.82	0.13	20.4	60
GaN***	3.51	0.13	22.7	40
Cu_2O	2.172	0.96	97.2	38****
SnO_2	3.596	0.33	32.3	86****

Table 4.2 Strongly anisotropic conduction and valence bands, direct transitions far from the centre of the Brillouin zone.

* Strongly anisotropic conduction and valence bands, direct transitions far from the centre of the Brillouin zone.

** In the presence of a magnetic field of 5 T.

*** An exciton in hexagonal GaN.

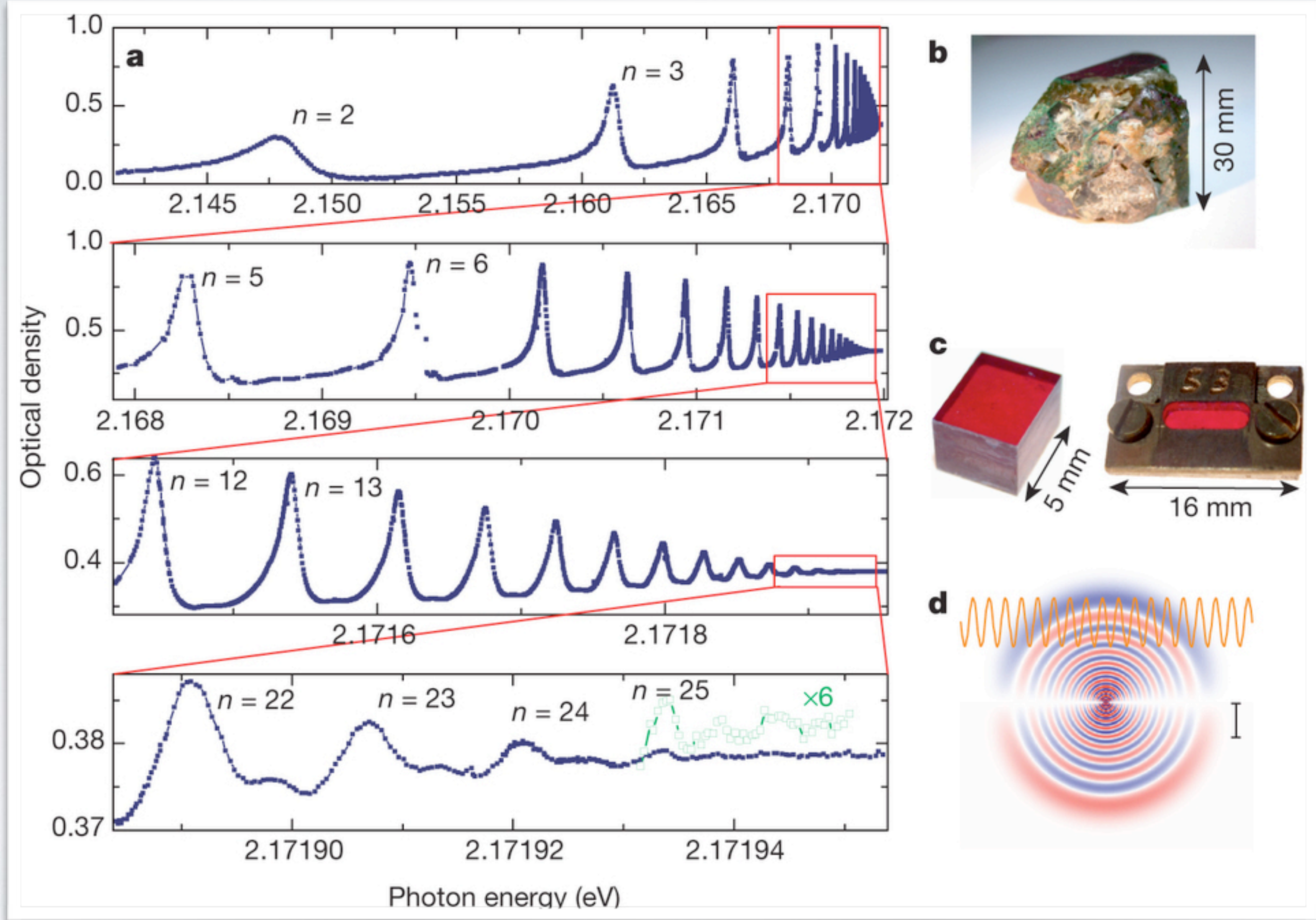
**** The ground-state corresponds to an optically forbidden transition, data given for $n = 2$ state.

A. Kavokin, et al. in *Microcavities*, Oxford Science Publications (2007)

Excitons - 3D

$$E_n = -\left(\frac{m^*}{m_0}\right)\frac{1}{\varepsilon_r^2} Ry \frac{1}{n^2}$$

Giant Rydberg excitons in the copper oxide Cu_2O



T. Kazimierczuk, D. Fröhlich, S. Scheel, H. Stolz & M. Bayer,
***Nature* 514, 343–347 (16 October 2014)**

Excitons - 2D (in quantum well)

The Schrödinger equation for an exciton in a quantum well (QW) reads:

$$\left(-\frac{\hbar^2}{2m_e}\nabla_e^2 - \frac{\hbar^2}{2m_h}\nabla_h^2 + V_e(z_e) + V_h(z_h) - \frac{e^2}{4\pi\varepsilon\varepsilon_0|\mathbf{r}_e - \mathbf{r}_h|}\right)\Psi = E\Psi$$

Solutions are again similar to 2D hydrogen atom:

Bohr radius : $a_{\mathrm{B}}^{2\mathrm{D}} = \frac{a_{\mathrm{B}}}{2}$

Binding energy of a ground state: $E_{\mathrm{B}}^{2\mathrm{D}} = 4E_{\mathrm{B}}$

Wave-function of the 1*s* state: $f_{1S}(\rho) = \sqrt{\frac{2}{\pi}}\frac{1}{a_{\mathrm{B}}^{2\mathrm{D}}}\exp(-\rho/a_{\mathrm{B}}^{2\mathrm{D}})$

Energies of the excited states: $E_n = -\frac{Ry^*}{\left(n-\frac{1}{2}\right)^2}$

Exciton condensation
few facts

BEC was predicted in the early1960's by Moskalenko, Blatt and coworkers and Casella (independently).

S.A. Moskalenko, Fiz. Tverd. Tela. 4, 276 (1962).
J.M. Blatt, K.W. Boer, and W. Brandt, Phys. Rev. 126, 1691 (1962).
Casella, R.C. Source: Journal of the Physics and Chemistry of Solids, v 24, p 19-26, Jan. 1963.

Necessary conditions for excitonic BEC

assumption: number of excitons is conserved

1. the lifetime of excitons is much longer than the thermalisation time
2. the exciton density is below the critical limit (in which the binding of the electrons and holes breaks down as in electron - hole plasma).

Possible description within weakly interacting Bose gas theory.

Exciton condensation

quantum coherence

Thermal de Broglie wavelength must be comparable or larger that the average distance between the particles:

$$\lambda_{dB} \sim r_s$$

Exciton condensation

quantum coherence

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de Broglie wavelength :

$$\frac{\hbar^2 k^2}{2m} = \frac{\hbar^2 (2\pi)^2}{2m\lambda_{dB}^2} \sim k_B T$$

which implies

$$\lambda_{dB} \sim \frac{2\pi\hbar}{\sqrt{2mk_B T}}$$

Exciton condensation

quantum coherence

Thermal de Broglie wavelength must be comparable or larger than the average distance between the particles:

$$\lambda_{dB} \sim r_s$$

Inter-particle distance:

$$r_s \sim n^{-1/3}$$

where n is the particle density.

The general condition for BEC is:

$$n \sim \frac{2^{3/2}}{(2\pi)^3} \frac{(mk_B T)^{3/2}}{\hbar^3}$$

or

$$T \sim \frac{(2\pi\hbar)^2}{2mk_B} n^{2/3}$$

In thermal equilibrium the standard calculation of statistical mechanics gives:

$$T_c = 0.17 \frac{(2\pi\hbar)^2}{2mk_B} n^{2/3}$$

E.g., K. Huang, Statistical Mechanics, 2nd ed., (Wiley, 1987)

Exciton condensation

particle mass significancy

$$T \sim \frac{(2\pi\hbar)^2}{2mk_B} n^{2/3}$$

Light mass implies Bose-Einstein effects at higher temperature !

	atoms	POLARITONS	EXCITONS
m	Rb: $10^4 m_e$	$10^{-4} m_e$	$10^{-2} m_e$
T_C	10^{-7}K	RT possible	~ 4 K possible
n	$10^{14}/cm^3$	$<10^{11}/cm^2$	limit: $10^{17}/cm^3$ or $10^{11}/cm^2$
lifetime	∞	10 ps	typically ~100 ns up to 1 ms in specially designed samples

Exciton condensation
lifetime

Many excitons are stable at room temperature

e.g. Cu_2O, CuCl, CdSe, ZnO, GaN

Exciton lifetime must be long enough to allow thermalisation (ps timescale).

and many semiconductors seemed to satisfy this requirement.

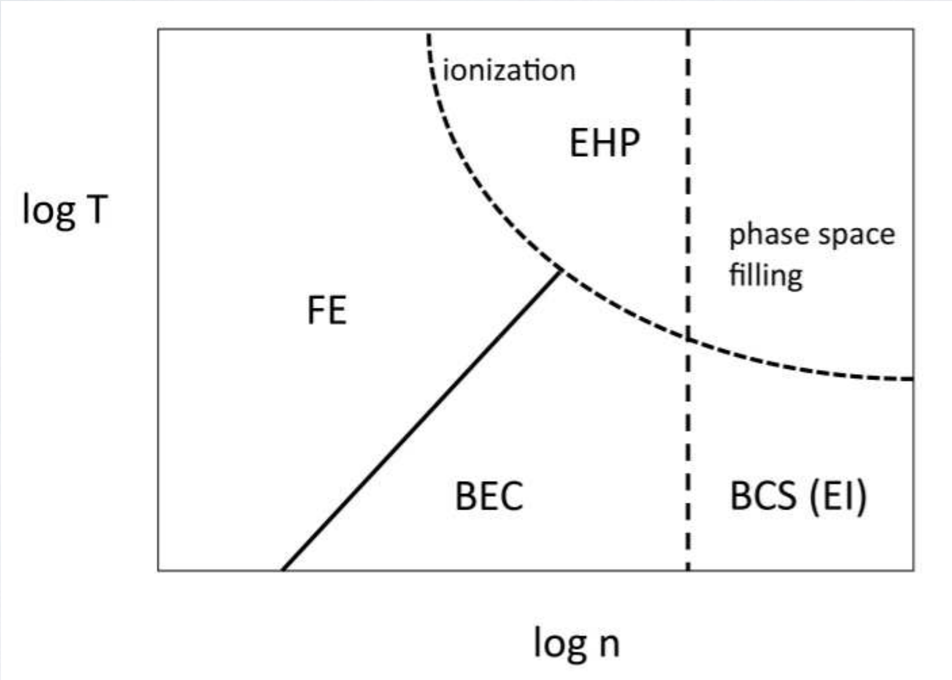
But the temperature - particle density relation has some important consequences.

Exciton condensation

comment on temperature vs particle density dependence

$$T_c \sim n^{2/3}$$

D. Snoke and G. M. Kavoulakis, Reports on Progress in Physics 77 (2014)

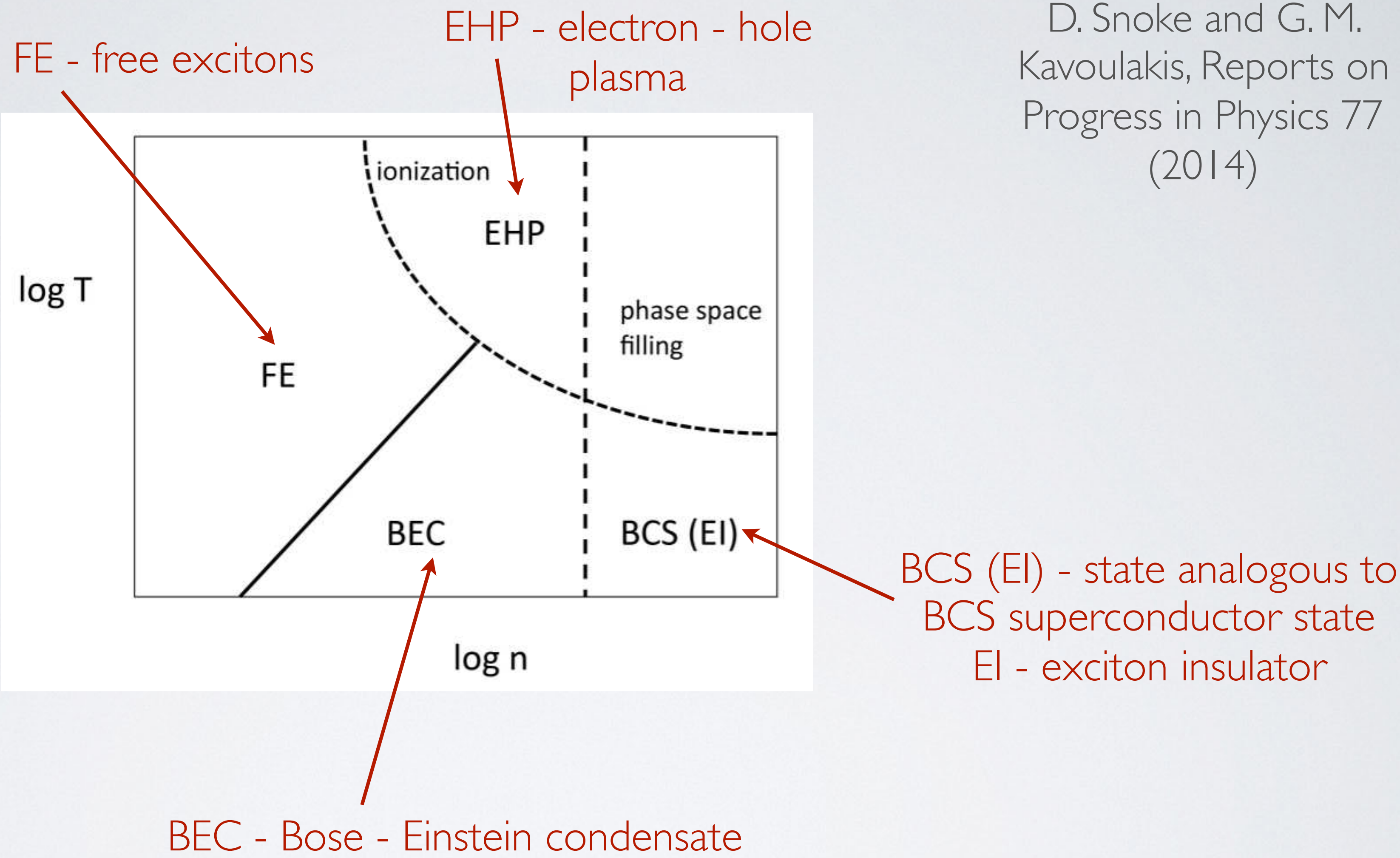


Problems:

1. competing phases of electron and holes system
2. density dependent recombination mechanisms

Exciton condensation

competing phases



D. Snoke and G. M. Kavoulakis, Reports on Progress in Physics 77 (2014)

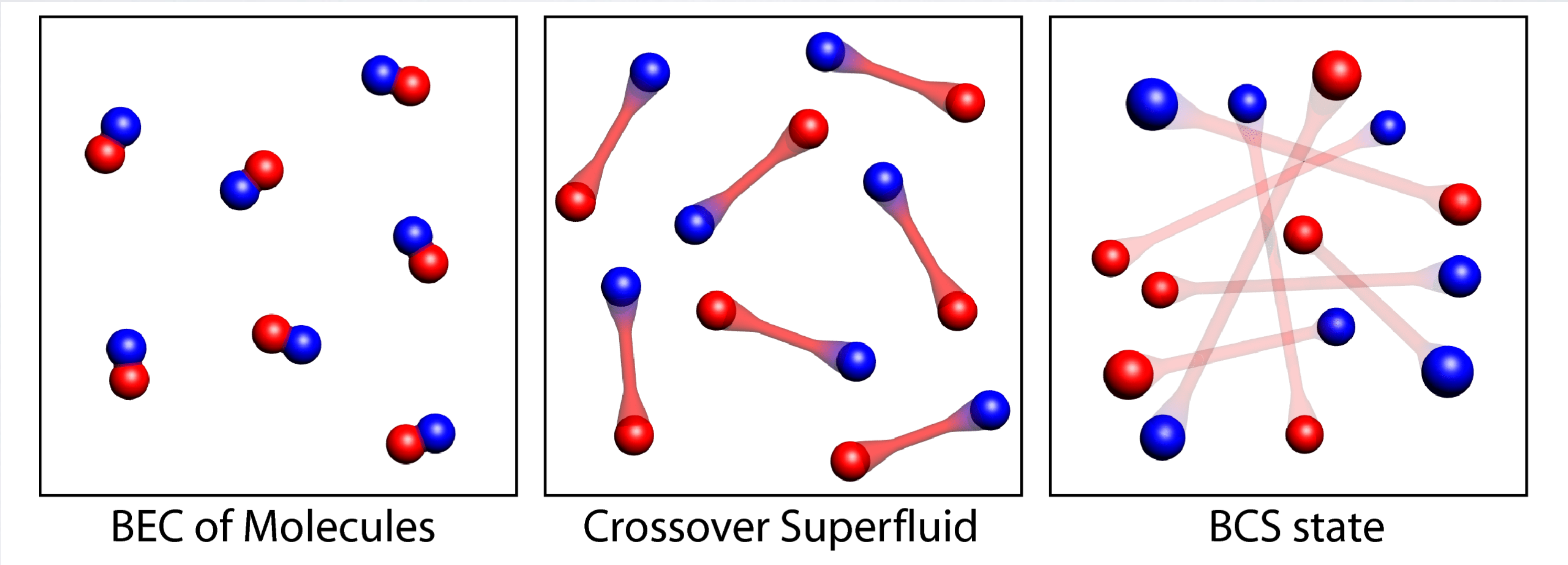
Exciton condensation

BCS state of excitons

Analog to BCS superconductor state first proposed by John **B**ardeen, Leon **C**ooper and John Robert **S**chrieffer („BCS”) in 1957

(Nobel Prize in Physics in 1972)

Superconductivity is a microscopic effect caused by a condensation of Cooper pairs into a boson-like state.



Strong pairing:
- atoms form molecules of up and down spin
- molecules are bosonic
- bosonic molecules condense into BEC

Weak pairing:
- atoms interact over a large distances
- BCS theory

Exciton condensation

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Superconductivity is a microscopic effect caused by a condensation of Cooper pairs into a boson-like state.

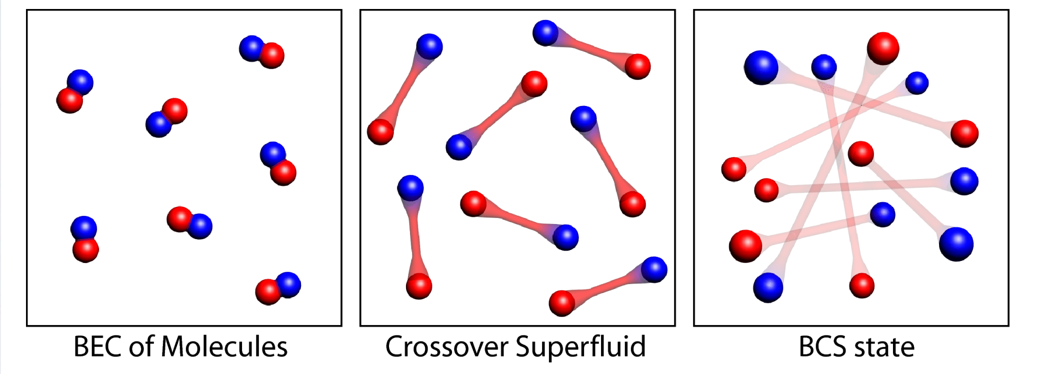
In a dense e-h system excitons are Cooper-pair-like Bose-particles and the exciton condensate is analogous to BCS superconductor state.

It is called excitonic insulator because the pairing occurs between electrons and holes and therefore the pair is neutral.

The transition between BEC and BCS state is smooth and the condensation has a mixed nature for intermediate densities.

Exciton condensation

BEC - BCS crossover



BEC - BCS crossover (and vice versa) was demonstrated for the first time for atomic condensates (of fermionic atoms) in 2003.

K. E. Strecker et al. , Phys. Rev. Lett 91, 080406 (2003)
M. Greiner et al., Nature 426, 537 (2003)
M. Bartenstein et al., Phys. Rev. Lett. 92, 203201 (2004)

The continuous crossover between BEC and BSC state is one of the fundamental problems in theoretical physics.

1969: David Eagles - BEC of fermion pairs for extreme concentrations
1980: Tony Leggett - problem described for T=0
1985: Phillip Nozieres and Stefan Smitt-Rink - generalisation of the problem for non-zero temperatures
1993: Mohit Randeria et al. - problem described within the integral trajectories
1995: Roman Micnas et al. - T matrices and Monte Carlo simulations

summary after T. Domański, IF UMCS Lublin

Exciton condensation

BCS state of excitons

Dense and diluted limit of e-h system determines the relation:

	BEC	BCS
r_s - inter-particle distance a_B - Bohr radius	$r_s > a_B$	$r_s < a_B$
	Fermi level of electrons (and holes) **small** compared to the intrinsic exciton binding energy	Fermi level of electrons (and holes) **large** compared to the intrinsic exciton binding energy
$E_F = \left(\frac{3\pi^2\hbar^3}{\sqrt{2}m^{3/2}}n\right)^{2/3}$ $\mathrm{Ry}_{ex} = \frac{e^2}{8\pi\epsilon a}$		$\frac{3\pi^2\hbar^3}{\sqrt{2}m^{3/2}}n > \left(\frac{e^2}{8\pi\epsilon a}\right)^{3/2}$ $\sim \left(\frac{4\pi\epsilon\hbar^2}{e^2 m}\right)^{3/2} n > \frac{1}{a^{3/2}}$

Exciton condensation

BCS state of excitons

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	Fermi level of electrons (and holes) **small** compared to the intrinsic exciton binding energy	Fermi level of electrons (and holes) **large** compared to the intrinsic exciton binding energy
n - density D - dimensionality	diluted $na_B^D << 1$	dense $na_B^D >> 1$

For excitonic BEC density of particles should be kept below $1/a^3$

Exciton condensation
BCS state of excitons

For excitonic BEC density of particles should be kept below $1/a^3$

In 2D case for excitons in GaAs QWs

$$n = 10^{10} cm^{-2}$$

$$m = 0.022 m_0$$

$$n < n_{mott} \sim \frac{1}{a_B^2} \sim 2 \cdot 10^{11} cm^{-2}$$

and in many semiconductors it is not a problem due to large binding energies of excitons.

Exciton condensation

EH plasma phase

Electron - hole plasma occurs for high temperatures and high densities.

Mechanism is analogous to classical ionisation of an atomic gas.

Analogue of „Mott" transition : conductor - insulator transition.

Screening of the electron-hole interaction by the number of ionised e and h.

Position of the phase boundary depends strongly on the binding energy of excitons: the higher the binding energy the higher the boundary is pushed to higher temperature.

Deeply bound excitons are preferable for BEC.

Exciton condensation

EH liquid phase

Electron-hole liquid (EHL) was first observed in Ge and Si in 1970's.

Electrons and holes are not bound into pairs as excitons or biexcitons but instead form two interpenetrating Fermi gases with the properties of classical liquid (with a surface tension).

EHL is a conductor.

EHL phase boundary scales with $T \sim n^{2/3}$

If EHL exists it prevents excitonic BEC at any temperature and density !

Exciton condensation

short summary

Electron - hole liquid

Ge

Si

Bose - Einstein condensate

Cu_2O ?

Indirect excitons in coupled quantum wells ??

excitonic insulator (BCS-like condensate)

electron bilayer in high magnetic fields at filling factor = 1

Exciton condensation

short summary

At low temperatures and low densities excitons can undergo BEC

Excitons with large binding energies and small Bohr radius are preferable for BEC.

<u>Challenges</u> for realization of exciton condensates	**<u>To solve:</u> Find or design semiconductor structures where**
short lifetime	**excitons have long lifetimes >> cooling times**
competing ground states, e.g. EHL	**excitons form the lowest energy state**
exciton destruction, e.g. due to Mott transition	**excitons have large binding energy**
disorder	**disorder is weak**

after: http://butovgroup.ucsd.edu

Exciton condensation
short summary

At low temperatures and low densities excitons can undergo BEC

Excitons with large binding energies and small Bohr radius are preferable for BEC.

First experiments were performed on CdSe and CuCl bulk semiconductors.

H. Kuroda, S. Shionoya, H. Saito, and E. Hanamura, J. Phys. Soc. Japan 35, 534 (1973).
T. Goto, T. Anzai, and M. Ueta, J. Phys. Soc. Japan 35, 940 (1973)

Around the same time deeply bound excitons in Cu_2O were discovered.

Exciton condensation

condensation in **CuCl**

L.L. Chase, L.L. N. Peyghambarian, G. Grynberg, and A. Mysyrowicz, 42, 1231 (1979).
N. Peyghambarian, L.L. Chase, and A. Mysyrowicz, Phys. Rev. B 27, 2325 (1983)

Deeply bound biexciton state.

Binding energy of 26 *meV*.

Which is comparable to *kT* at room temperature.

But has also a strong polaritonic effect in which excitons and biexcitons strongly couple to light.

This leads to a short radiative lifetimes.

Exciton condensation

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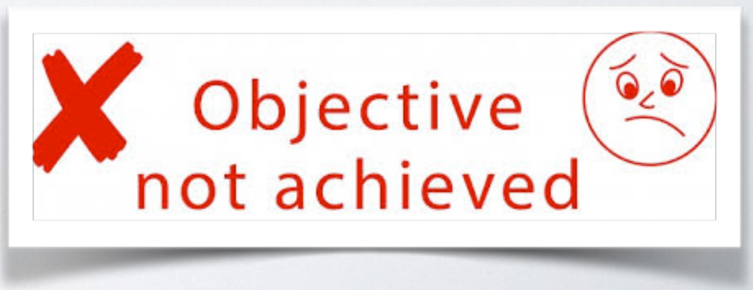
The question about the existence of bosonic stimulated scattering was not answered.



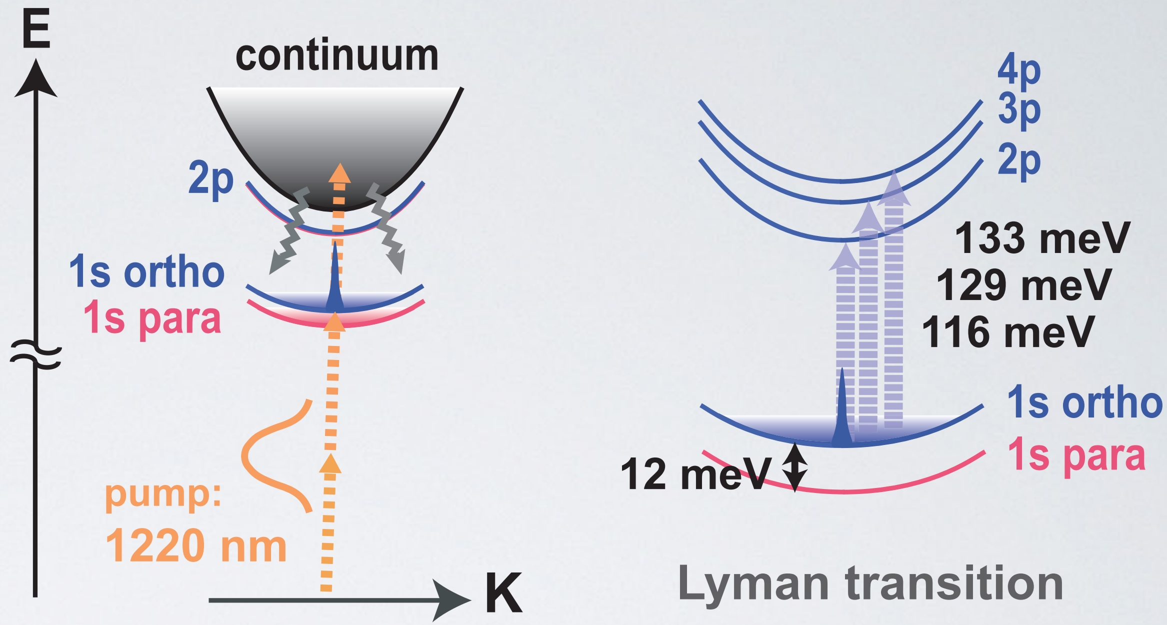
Exciton condensation

BEC in **Cu_2O**

K. Yoshioka, K. Miyashita, M. Kuwata-Gonokami, Optics Express 22, 3261 (2013)

Exciton binding energy of **150 *meV***.

Biexciton state weakly bound.



Two types of excitons:

orthoexcitons - spin triplet (parallel spins of electron and hole)

paraexcitons - spin singlet (anti-parallel spins of electron and hole)

The highest valence and the lowest conduction bands are formed from Cu states, the 3d and 4s orbitals, respectively.

Cu_2O is therefore a forbidden direct-gap semiconductor.

Exciton condensation

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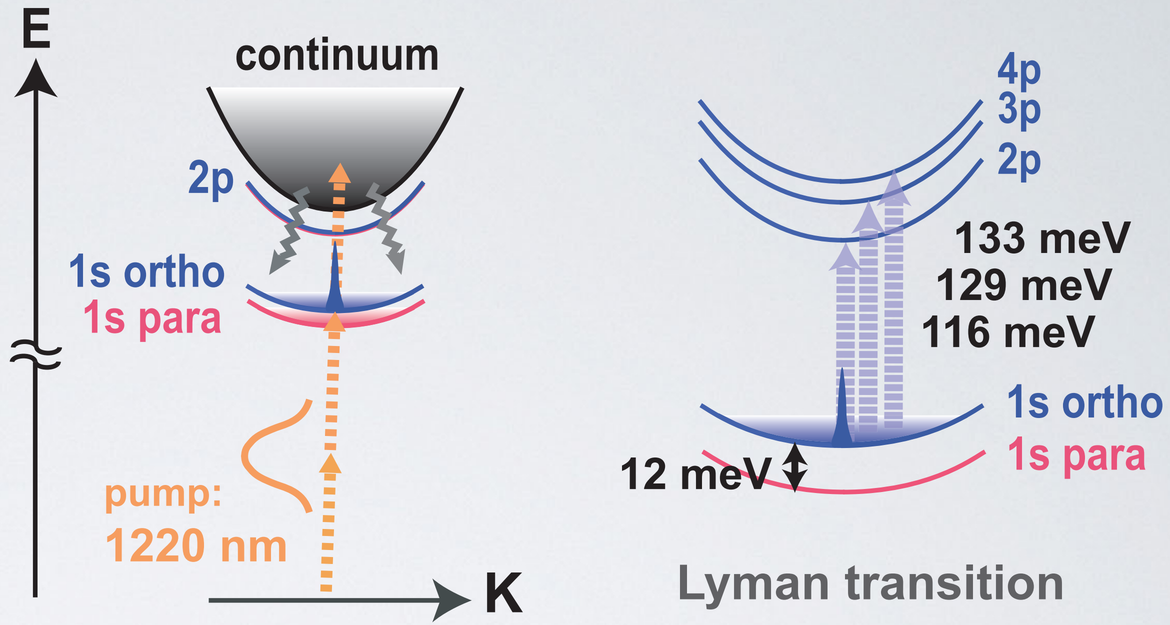


Two types of excitons:

orthoexcitons - spin triplet (parallel spins of electron and hole)
paraexcitons - spin singlet (anti-parallel spins of electron and hole)

Paraexciton is the ground state and due to the crystal lattice symmetry has zero oscillator strength for interaction with photons.

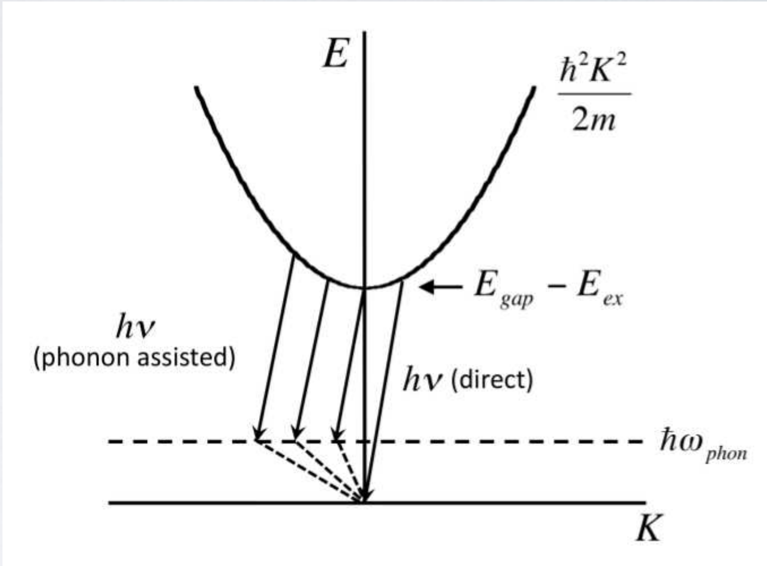
⟶ *(almost) infinite lifetime: 100 ns - 1 ms (weakly phonon allowed transitions, recombination on impurities or applied stress)*

Orthoexciton (spin-triplet) has a quadrupole allowed radiative recombination process (*also phonon assisted processes are allowed*).

Exciton condensation

BEC in Cu_2O

Phonon-assisted luminescence spectroscopy.

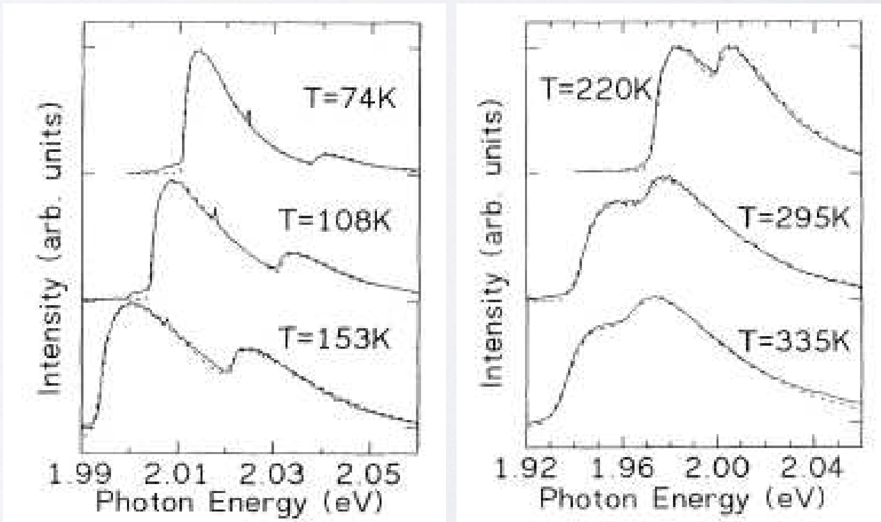


Exciton can recombine with the photon emission and a phonon taking up any excess momentum

$$E_{exciton} = E_{photon} + E_{phonon}$$

Can occur for orthoexcitons and paraexcitons.

Energy spectrum (spectral function) gives the kinetic energy distribution of excitons (*not possible to observe for atoms and extensively explored for polaritons*).



D.W. Snoke, A. Shields, and M. Cardona, Phys. Rev. B 45, 11693 (1992)

Exciton condensation

BEC in Cu_2O

Spectral function

energy shift - real part of the self-energy

broadening - imaginary part of the self-energy - inverse of the scattering time of the excitons (exciton-exciton, exciton-phonon, exciton-impurity, ...)

Purpose:

1. To estimate the absolute density of the excitons

Early experiments show that orthoexcitons and paraexcitons exceeded the critical densities for BEC

2. Possibly fit the spectrum with the Bose-Einstein distribution

But the spectral broadening do not correspond to the presumed exciton densities.

Auger recombination process dominates !

Exciton condensation

BEC in Cu_2O

D.P. Trauernicht, J.P. Wolfe, and A. Mysyrowicz, Phys. Rev. 34, 2561 (1986)

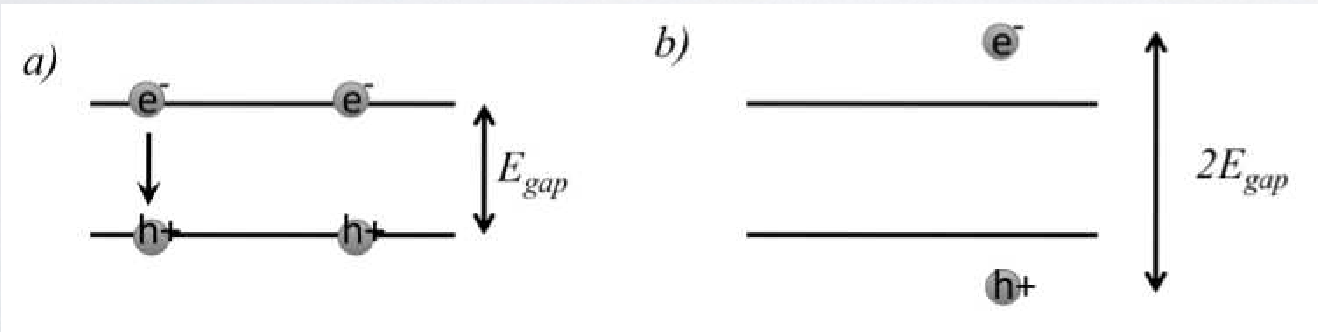
Auger recombination process dominates !

Two excitons collide, one recombines and instead of emitting photon the energy is given to ionise the second exciton.

Hot electron and hole loose energy by emitting phonons or by collisions and form an exciton again.

Returning exciton can be either ortho- or para-exciton.

Exciton lifetime is density dependent.



The Auger recombination process. In (a), the exciton on the left recombines, giving its energy to the exciton on the right, which leads to the final state (b) of a single ionised exciton.

Exciton condensation

BEC in Cu_2O

Conclusion:

Auger recombination process dominates and shortens the lifetime of excitons.

Only at sufficiently low temperature the time scale for thermalisation via phonons can be much shorter than the lifetime due to Auger recombination.

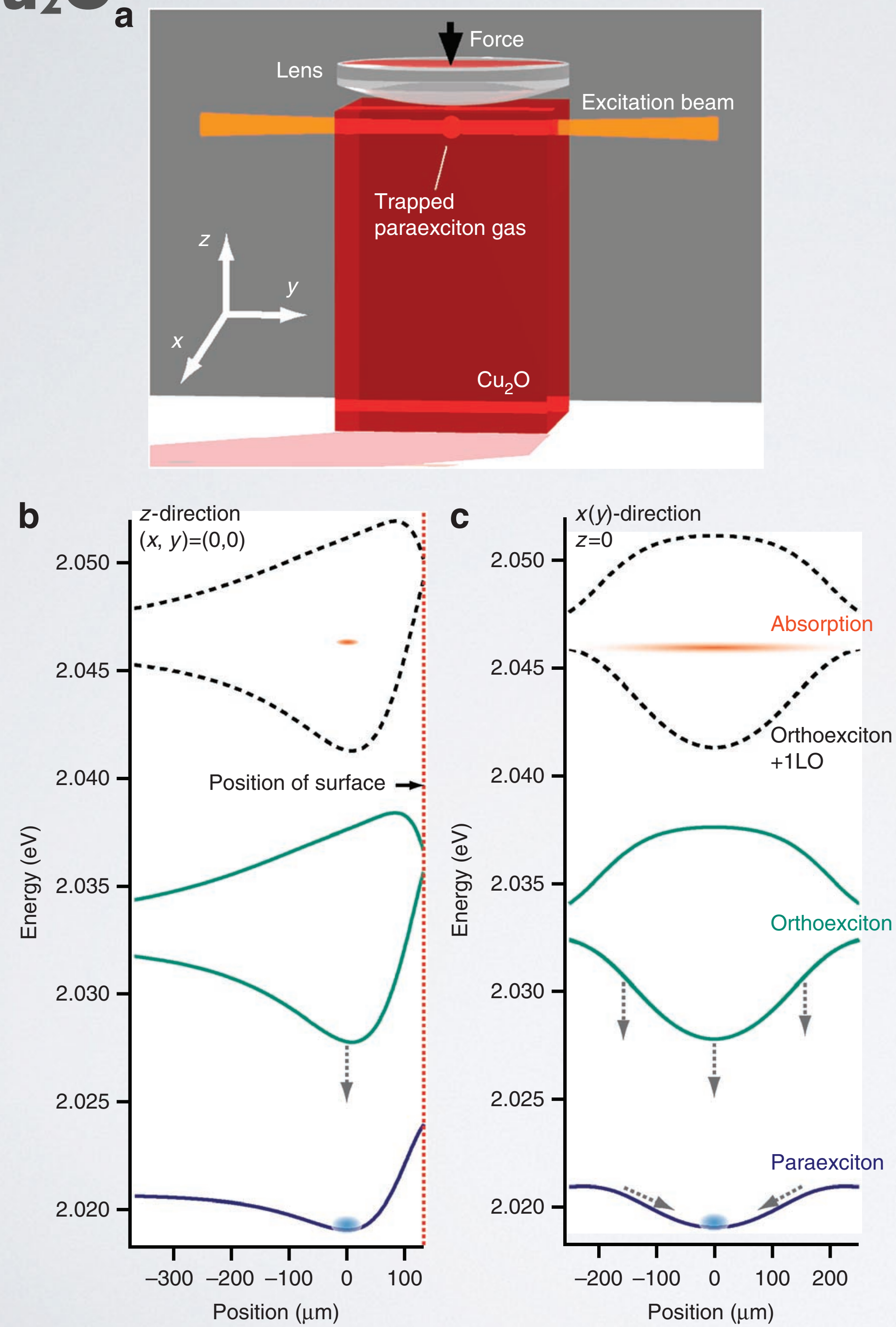
further experiments are performed at
hundreds of mK temperatures :-(

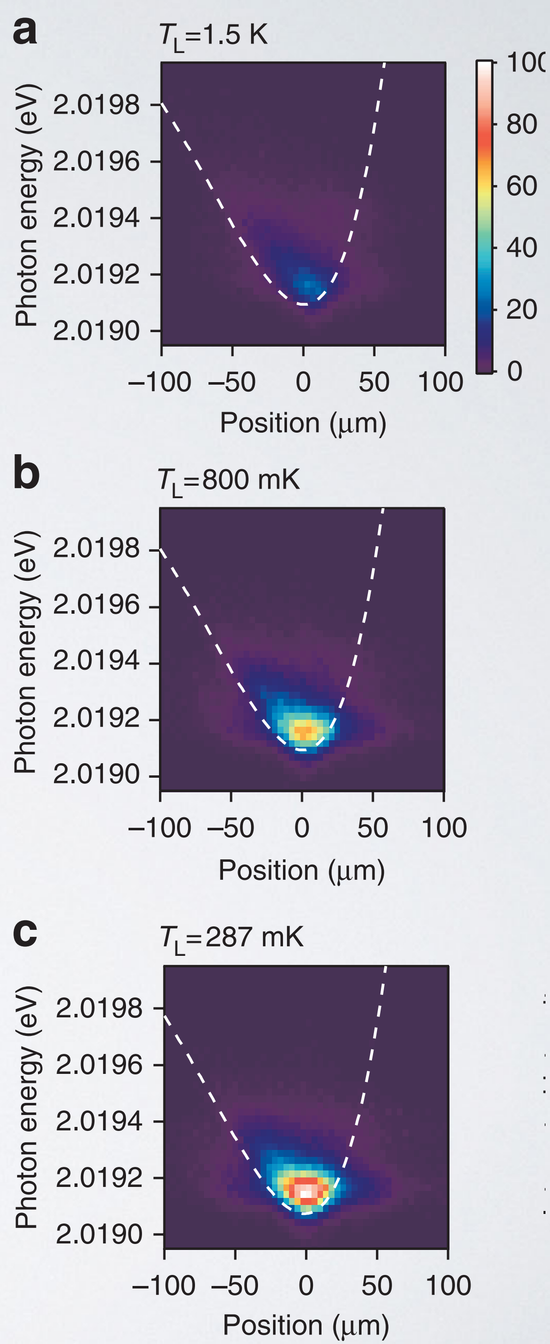
Stress is used to produce a three-dimensional harmonic trap for paraexcitons.

Exciton condensation

BEC in Cu_2O

K. Yoshioka, E. Chae, and M. Kuwata-Gonokami, Nature Comm. 2, 328 (2011)





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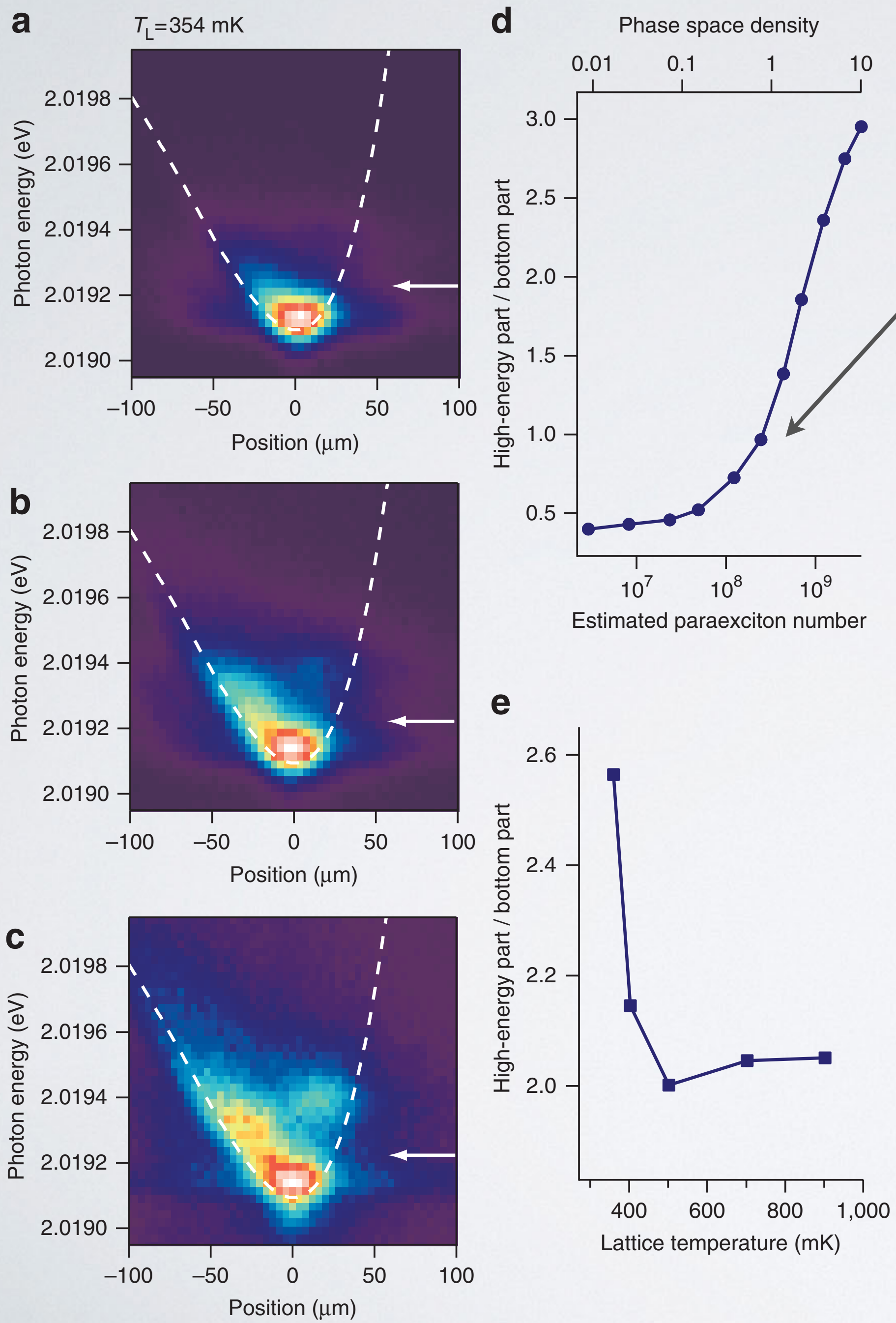


„Threshold like behaviour indicates the BEC."

„Explosion" of paraexcitons below a critical temperature

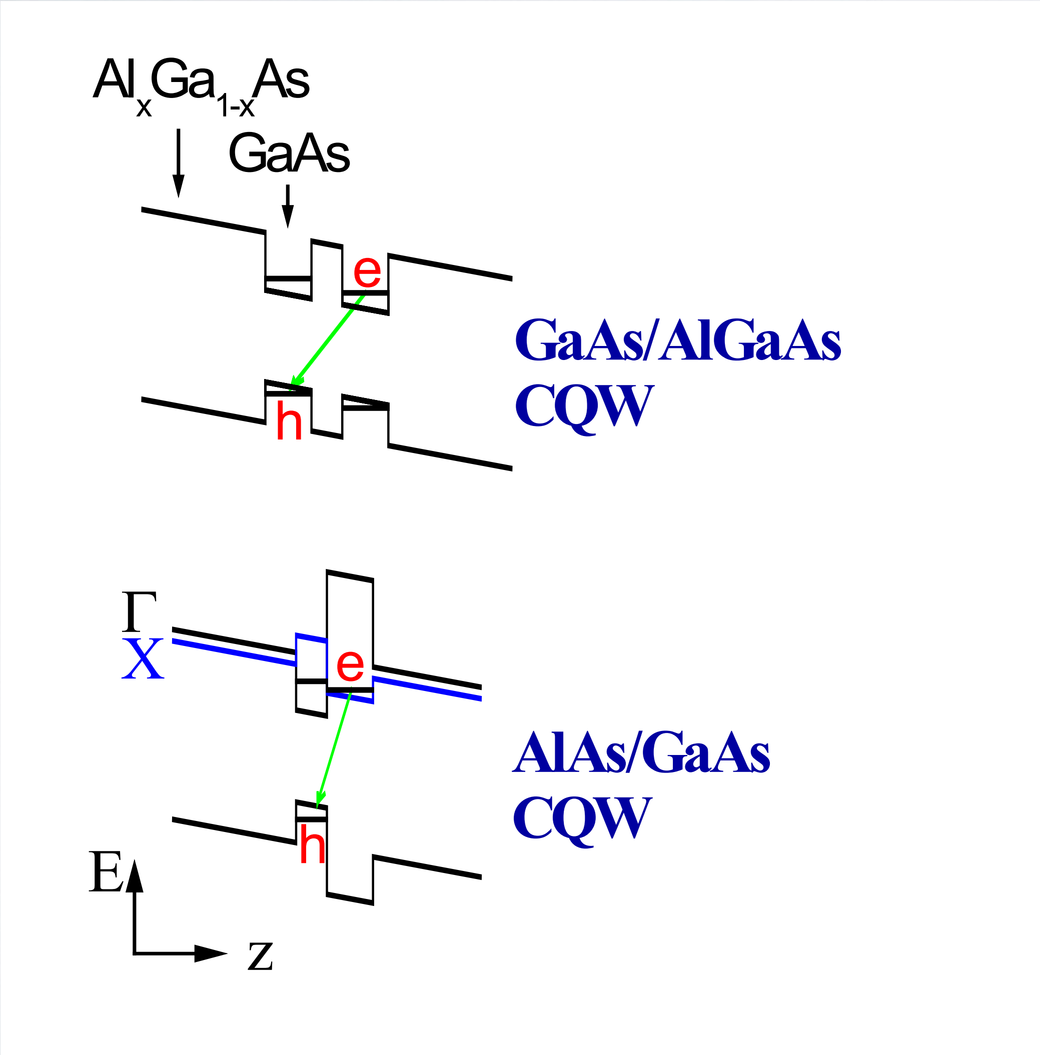
probably due to non-equilibrium effects.

Not observed by other groups, but the experimental conditions were different.

Exciton condensation

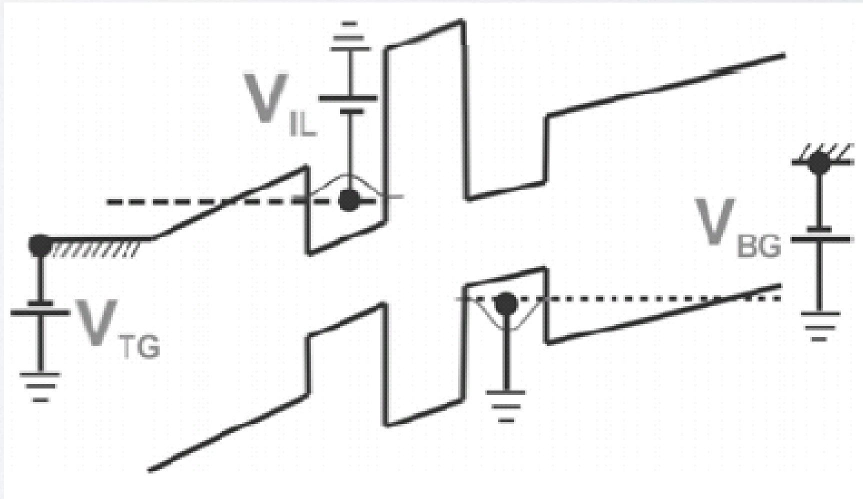
BEC of indirect excitons

Indirect excitons in coupled QW



Excitons act as oriented electric dipoles with repulsive interactions

Gated structures



Exciton condensation

BEC of indirect excitons

<u>10^3-10^6 times longer exciton lifetime</u> due to separation between electron and hole layers

realization of cold exciton gas in separated layers was proposed by
Yu.E. Lozovik, V.I. Yudson (1975); S. I. Shevchenko (1976);
T. Fukuzawa, S.S. Kano, T.K. Gustafson, T. Ogawa (1990)

<u>10^3 times shorter exciton cooling time</u> than that in bulk semiconductors

↓

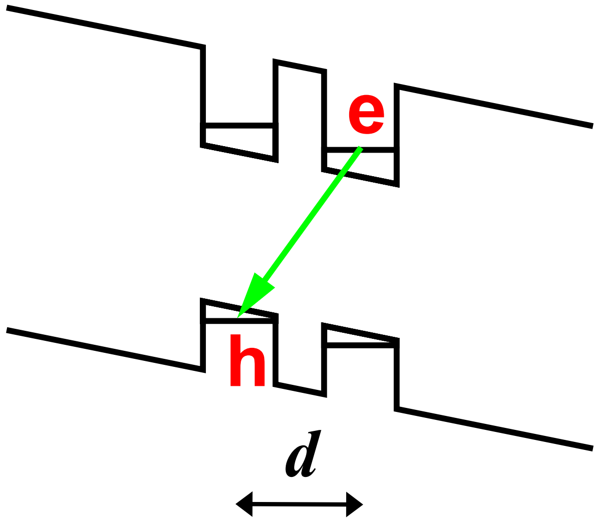
$$T_X \sim 100 \text{ mK}$$

has been realized experimentally 30 times below T_{dB}

L. V. Butov, et al. University of California at San Diego
www.butovgroup.ucsd.edu

Exciton condensation

BEC of indirect excitons ??



smaller *d*

↓

smaller exciton radius

↓

larger T_c at high n

in CQW with $L_B = 4$ nm, $L_{QW} = 8$ nm
$d \approx 12$ nm, exciton radius $a_B \approx 20$ nm

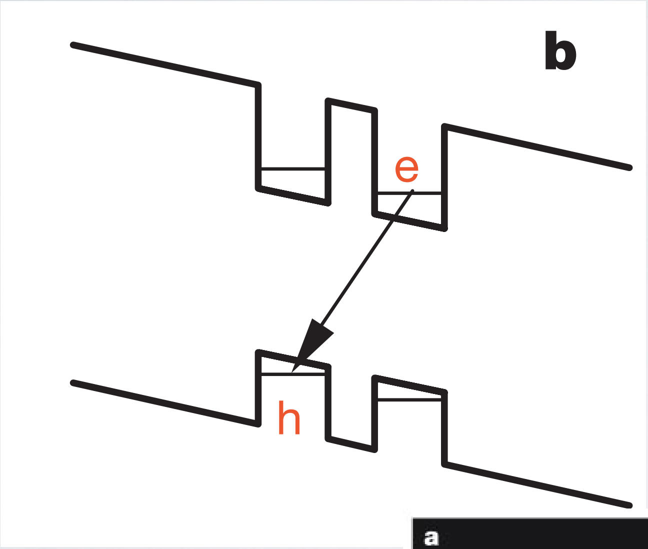
2D gas of excitons in GaAs QW
$n = 10^{10}$ cm^{-2}, $m_{exciton} = 0.2\, m_e \rightarrow T_{dB} \sim 3$ K

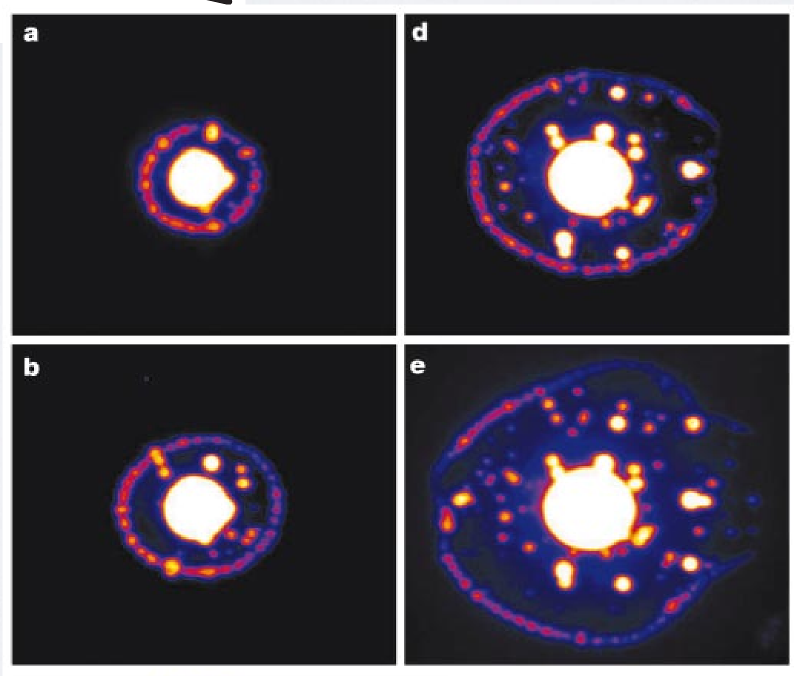
$n < n_{Mott} \sim 1/a_B^2 \sim 2\times10^{11}$ cm^{-2}

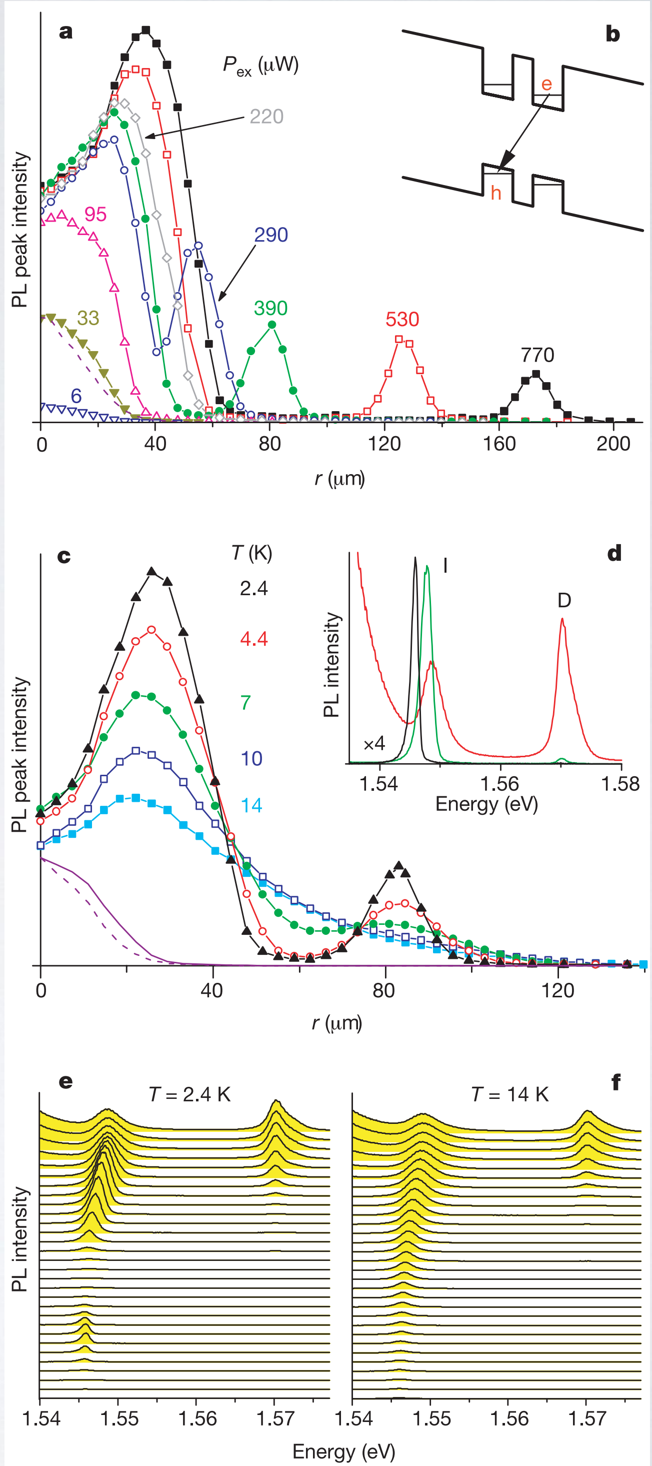
L. V. Butov, et al. University of California at San Diego
www.butovgroup.ucsd.edu

Exciton condensation

BEC of indirect excitons ??







L. V. Butov, A. C. Gossard, D. S. Chemla
Nature 418, 751 (2002)

Exciton condensation

BEC of dark excitons

„Grey" exciton condensates.

Excitons in semiconductor quantum wells are composed of electrons (±1/2 spin) and holes (±3/2 spin)

„bright" excitons (±1) : ∓1/2 electrons and ±3/2 holes
„dark" excitons (±2) : ±1/2 electrons and ±3/2 holes

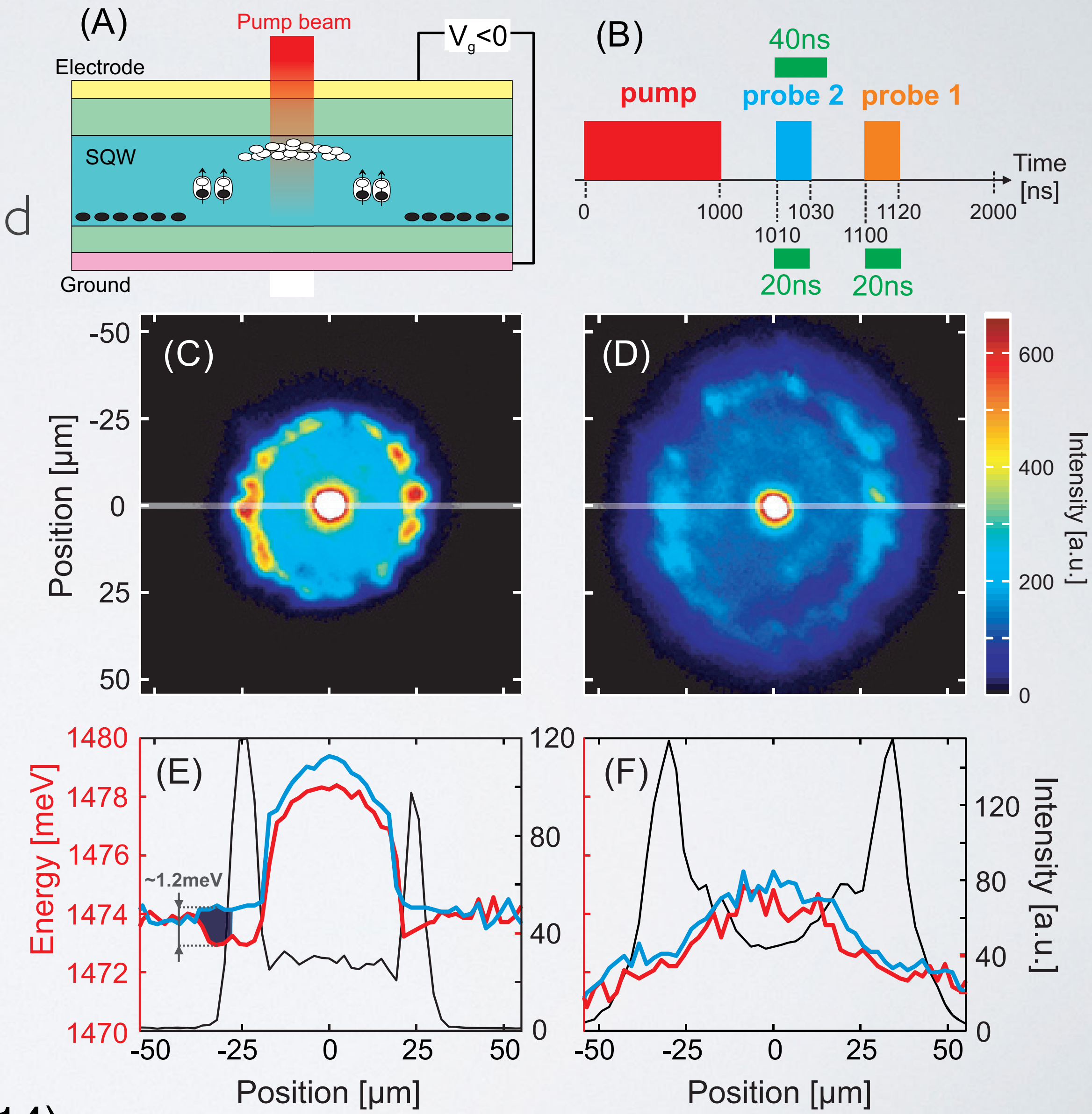
Dark excitons are ground state excitons.

Exciton condensation

BEC of dark excitons

„Gray" condensates of <u>dipolar</u> excitons.

Radiative lifetime ~ 20 ns.
Energy splitting between bright and dark states ~ 20 µeV.

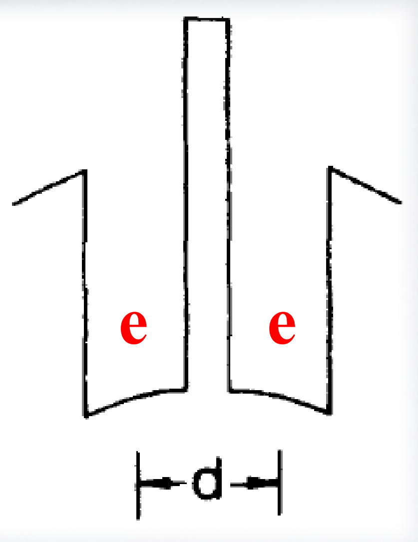


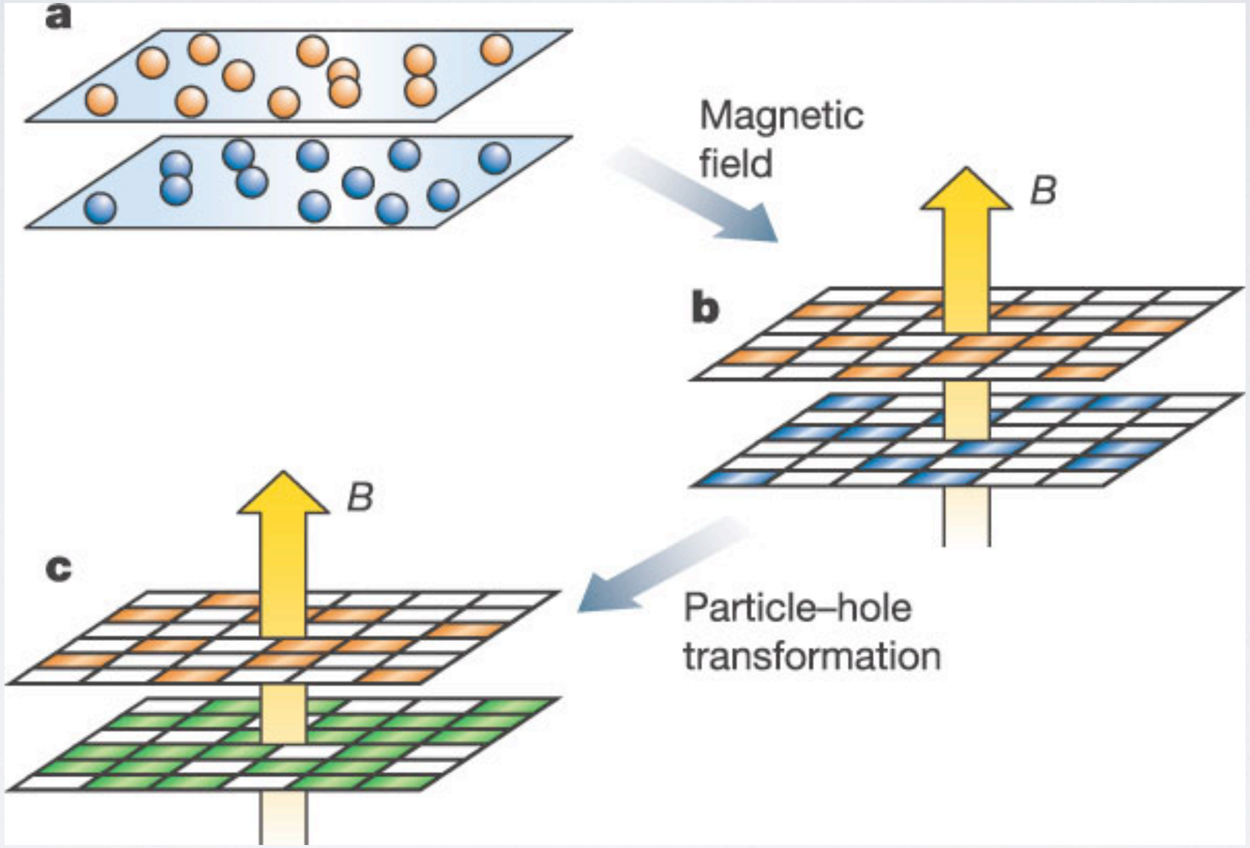
M. Alloing et al., EPL 107, 10012 (2014)

Exciton condensation

Quantum Hall bilayers at magnetic field

J.P. Eisenstein, A.H. MacDonald, Nature 432, 691 (2004)





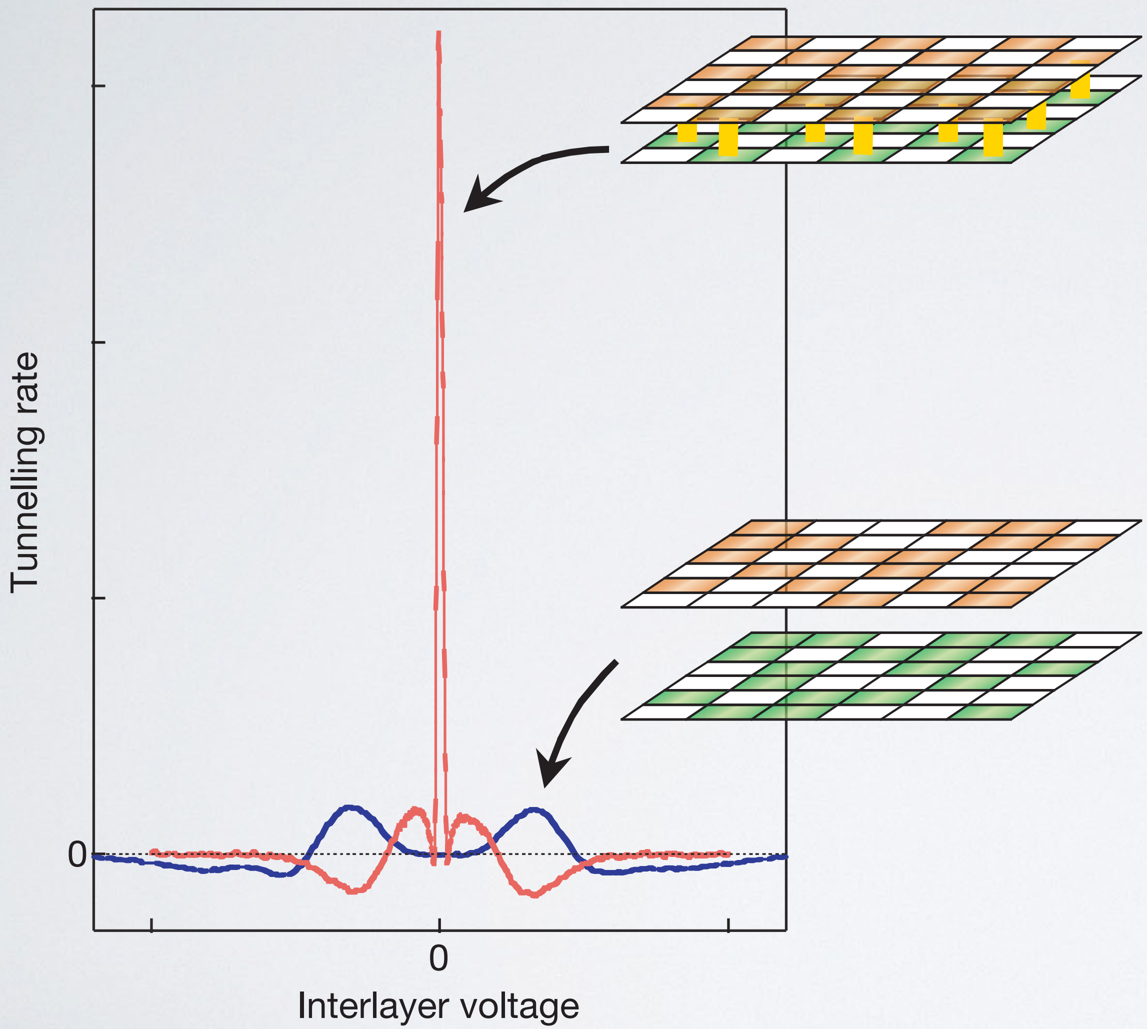
particle - hole transformation

collective electron state ⟶ exciton condensate

Exciton condensation

Quantum Hall bilayers at magnetic field

J.P. Eisenstein,
A.H. MacDonald,
Nature 432,
691 (2004)



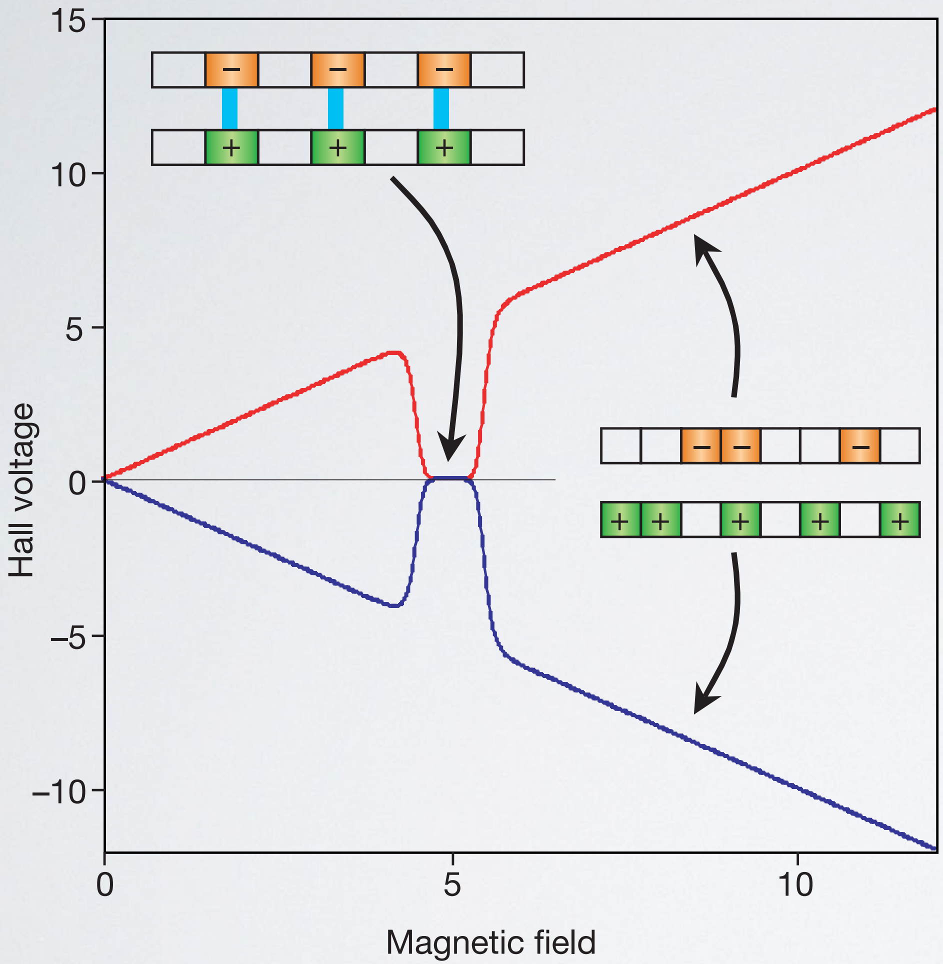
Enhancement of tunnelling rate
between electrons

no exciton above Tc
e - h pairing below Tc

Exciton condensation

Quantum Hall bilayers at magnetic field

J.P. Eisenstein,
A.H. MacDonald,
Nature 432,
691 (2004)



The currents are carried by independent charged particles in the two layers.

The currents are oppositely directed, these voltages have opposite signs in the two layers.

For exciton condensation, being charge-neutral, these excitons experience no Lorentz force and the Hall voltage is expected to vanish.

no exciton above Tc

e - h pairing below Tc

? BCS-like condensate ?

Exciton condensation

summary

„The Bose-Einstein condensation of excitons has a long history with seminal contributions from Leonid V. Keldysh. Despite numerous efforts, however, a compelling experimental evidence is still missing."

R. Zimmermann

for review:

BOSE-EINSTEIN CONDENSATION OF EXCITONS: PROMISE AND DISAPPOINTMENT
R. Zimmermann, Institut fur Physik der Humboldt-Universität zu Berlin, Newtonstrasse 15, 12489 Berlin, Germany

BOSE-EINSTEIN CONDENSATION OF EXCITONS IN CU_2O: PROGRESS OVER THIRTY YEARS
David Snoke, Department of Physics and Astronomy University of Pittsburgh,
and G. M. Kavoulakis, Technological Educational Institute of Crete