



清華大學物理系 奈米物理特論 2013/0328 上課內容 (III)

半導體奈米結構之成長與光電特性

交通大學電子物理系 周武清 教授

大綱:

Part I: 半導體奈米結構成長-----

分子束磊晶(MBE, molecular beam epitaxy)

半導體奈米結構形貌研究---AFM

Part II: 半導體奈米結構光電特性---Photoluminescence

Part III: 半磁性半導體奈米結構之自旋磁光特性





What is “Spin”?

How to manipulate spin?

How can we use “spin” to fabricate useful devices?

Outline

1. Introduction to II-VI diluted magnetic semiconductor (DMS) quantum dots (QDs).

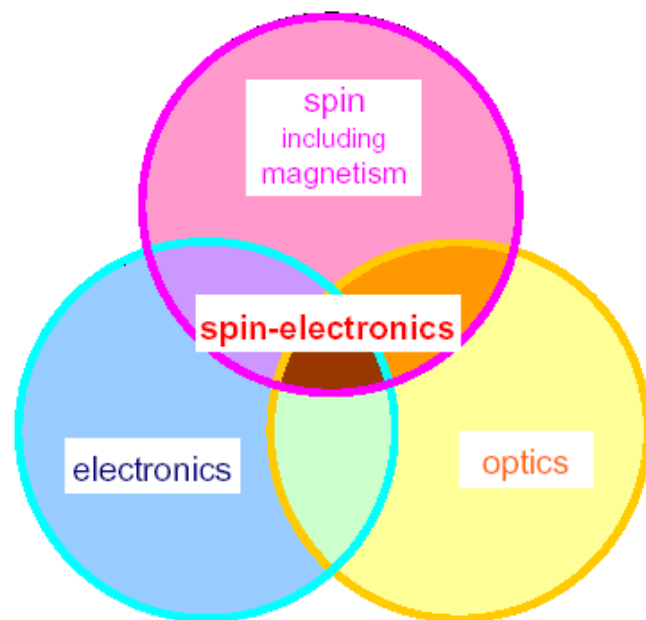
(比較III-V magnetic semiconductors)

2. Growth, structure and band alignment of ZnMnTe QDs.

3. Circular polarization measurement and spin dynamics.

4. Devices for spintronics

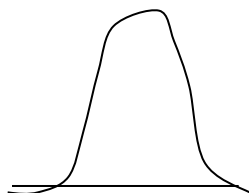
5. Conclusion.



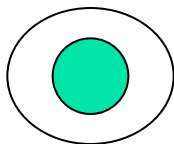


加入錳Mn後能帶的變化
有甚麼特殊的光電特性?

_____ $n=2$

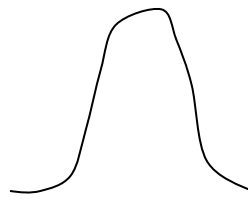


$n=1$

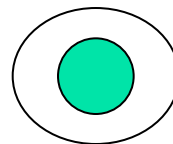


Isolated hydrogen atom

_____ $n=2$



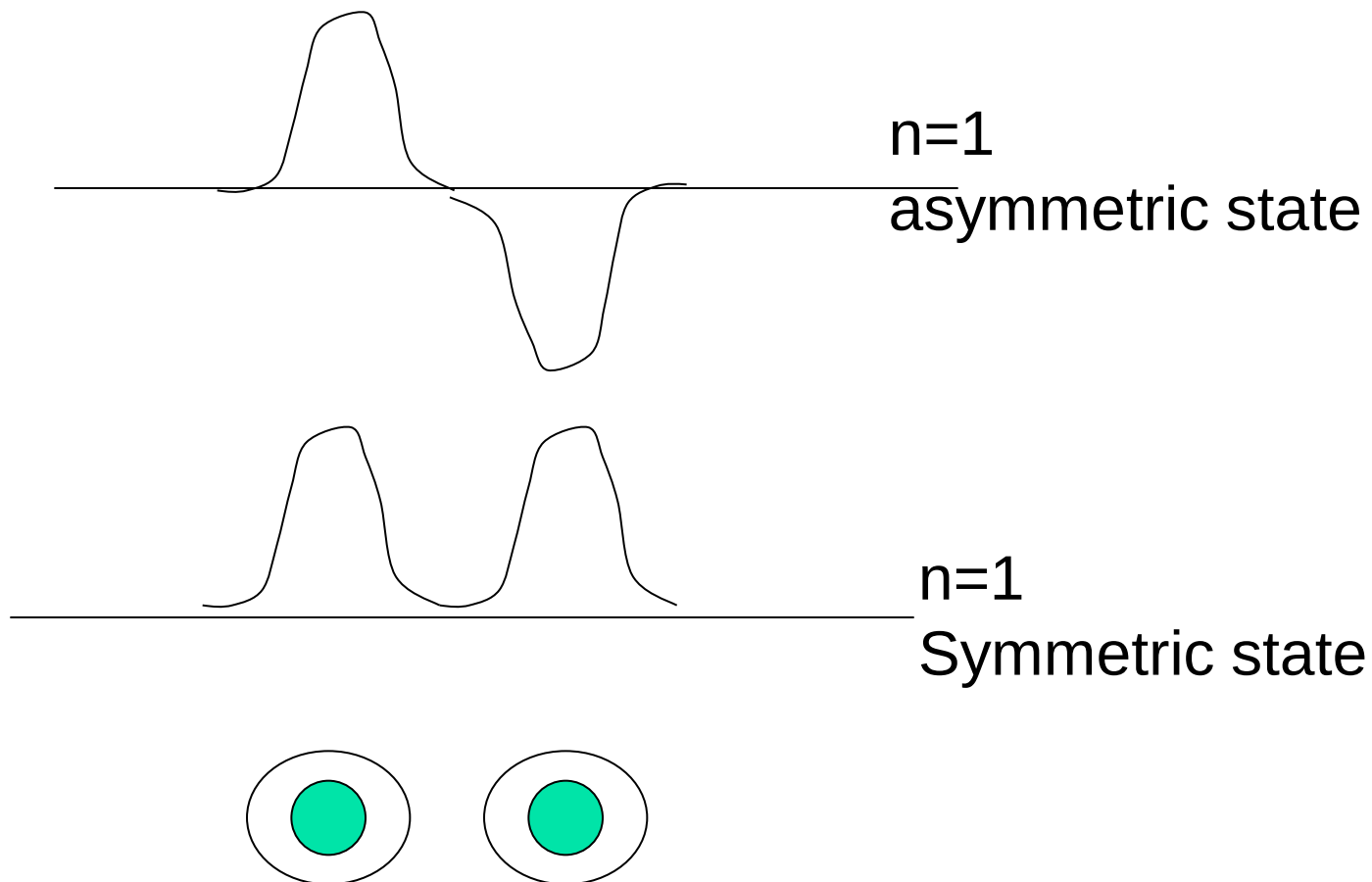
$n=1$



Isolated hydrogen atom

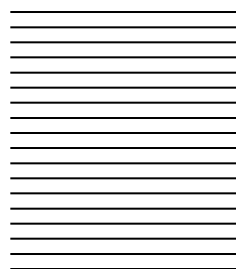


加入錳Mn後能帶的變化 有甚麼特殊的光電特性？

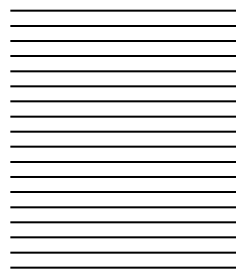




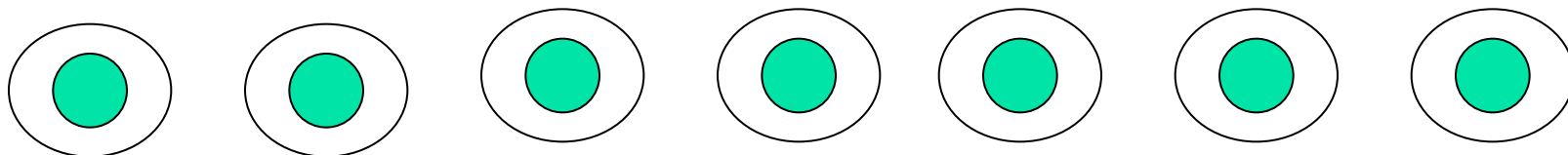
加入錳Mn後能帶的變化
有甚麼特殊的光電特性？



$n=2$

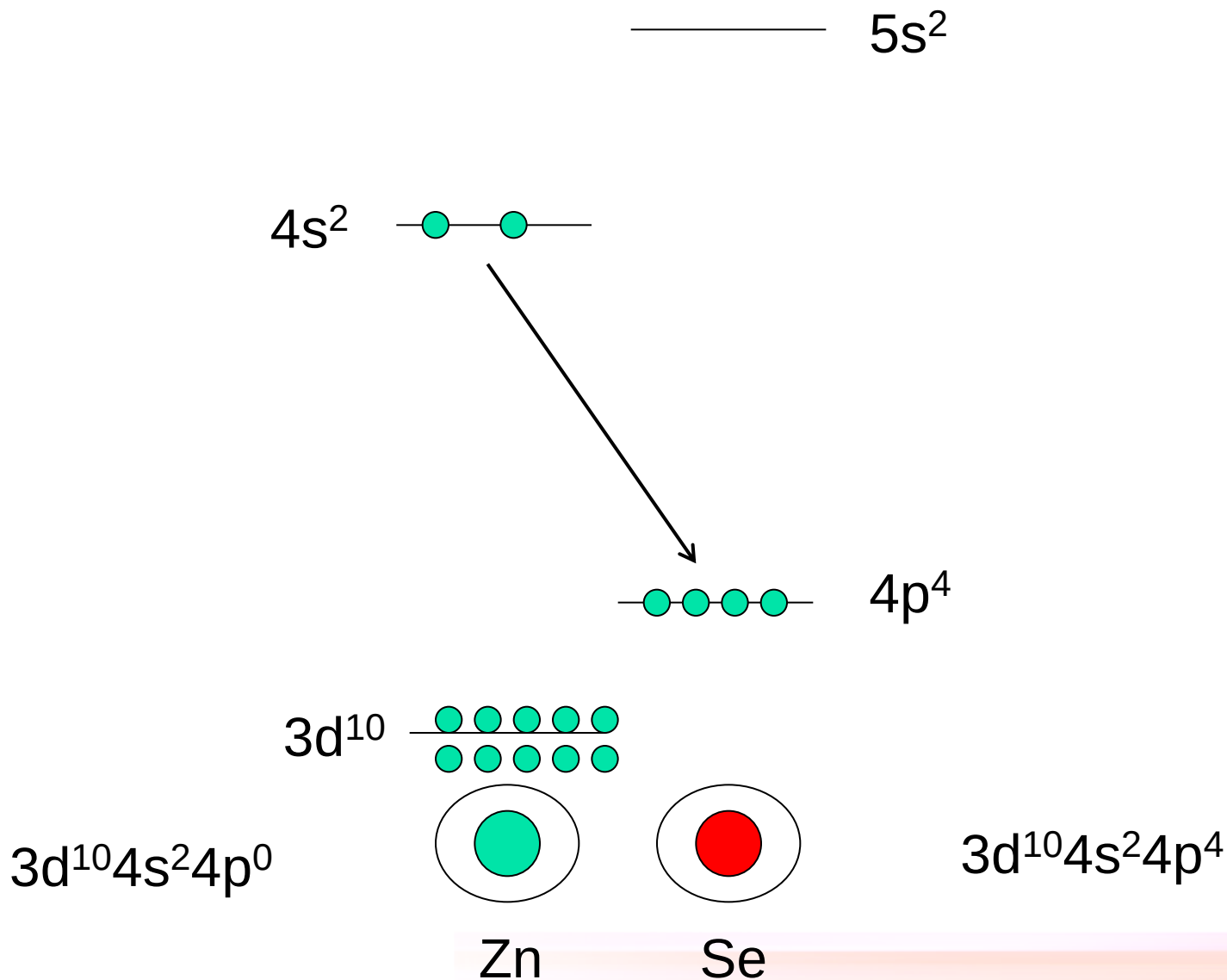


$n=1$





加入錳Mn後能帶的變化
有甚麼特殊的光電特性?





加入錳Mn後能帶的變化
有甚麼特殊的光電特性?

————— $5s^2$

$4s^0$ —————

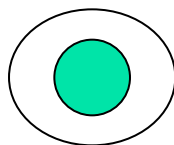
$L=0, S=1/2, J=1/2$

●●●●●● $4p^6$

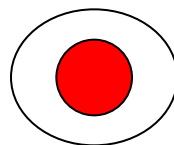
$L=1, S=1/2, J=1/2, 3/2$

$m_J=+1/2, -1/2$

$m_J=+3/2, +1/2, -1/2, -3/2$



Zn



Se

$3d^{10}4s^24p^0$ $3d^{10}4s^24p^4$



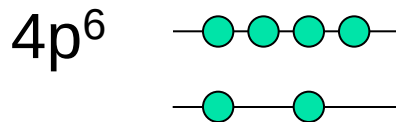
加入錳Mn後能帶的變化
有甚麼特殊的光電特性?

$L=0, S=1/2, J=1/2$

空能階 $4s^0$

$5s^2$

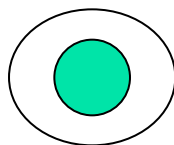
填滿



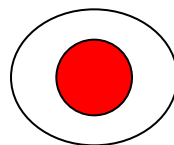
$L=1, S=1/2, J=1/2, 3/2$

$m_j=+3/2, +1/2, -1/2, -3/2$

$m_j=+1/2, -1/2$



Zn



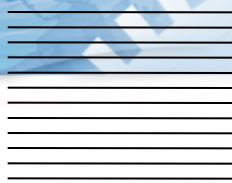
Se

$3d^{10}4s^24p^0$ $3d^{10}4s^24p^4$

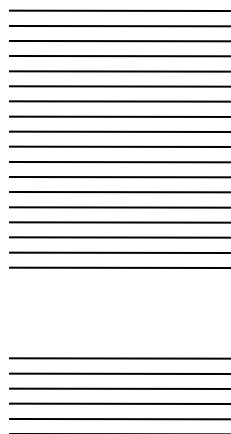


加入錳Mn後能帶的變化
有甚麼特殊的光電特性？

Conduction band (CB) 4s

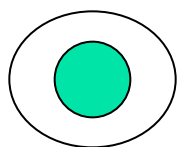


Valence band (VB) 4p

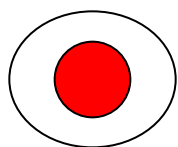


$m_j = +3/2, +1/2, -1/2, -3/2$
Heavy and light hole band

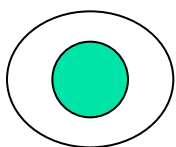
$m_j = +1/2, -1/2$
Spin orbital interaction band



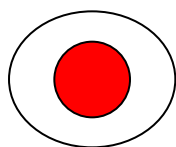
Zn



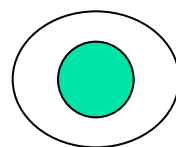
Se



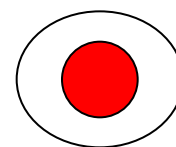
Zn



Se



Zn



Se

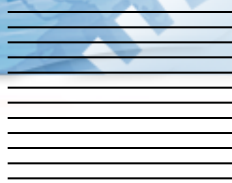
$3d^{10}4s^24p^0$ $3d^{10}4s^24p^4$



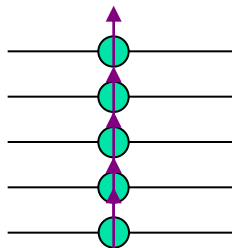
加入錳Mn後能帶的變化
有甚麼特殊的光電特性？

(CB)

4s²



3d⁵

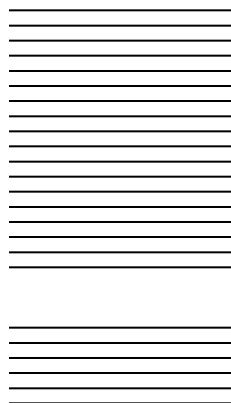


Hund's rule 

Spin alignment

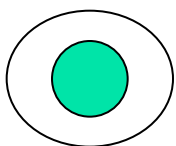
(VB) 填滿

4p

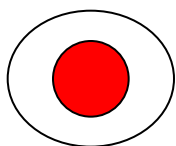


$m_j = +3/2, +1/2, -1/2, -3/2$
HH and LH band

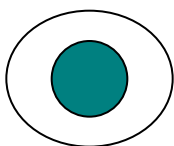
$m_j = +1/2, -1/2$ S-O band



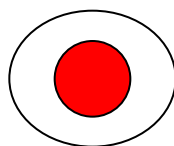
Zn



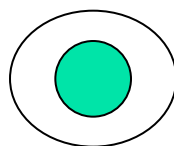
Se



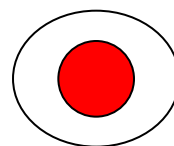
Mn



Se



Zn



Se

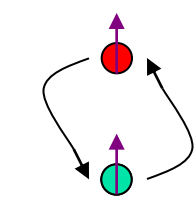
3d¹⁰4s²4p⁰

3d⁵4s²4p⁰

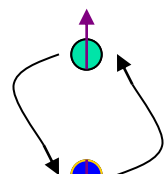
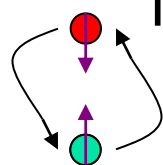
3d¹⁰4s²4p⁴



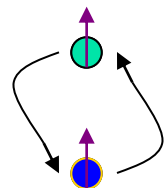
加入錳Mn後能帶的變化
有甚麼特殊的光電特性?



s-d exchange
interaction



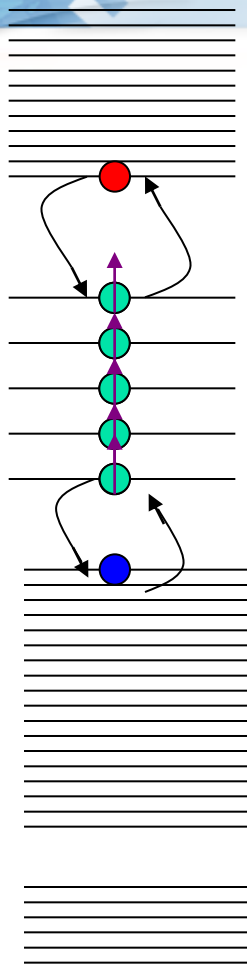
p-d exchange
interaction



4s²

3d⁵

4p



Hund's rule

$m_j = +3/2, +1/2, -1/2, -3/2$
HH and LH band

$m_j = +1/2, -1/2$ S-O band

sp-d exchange interaction的結果?

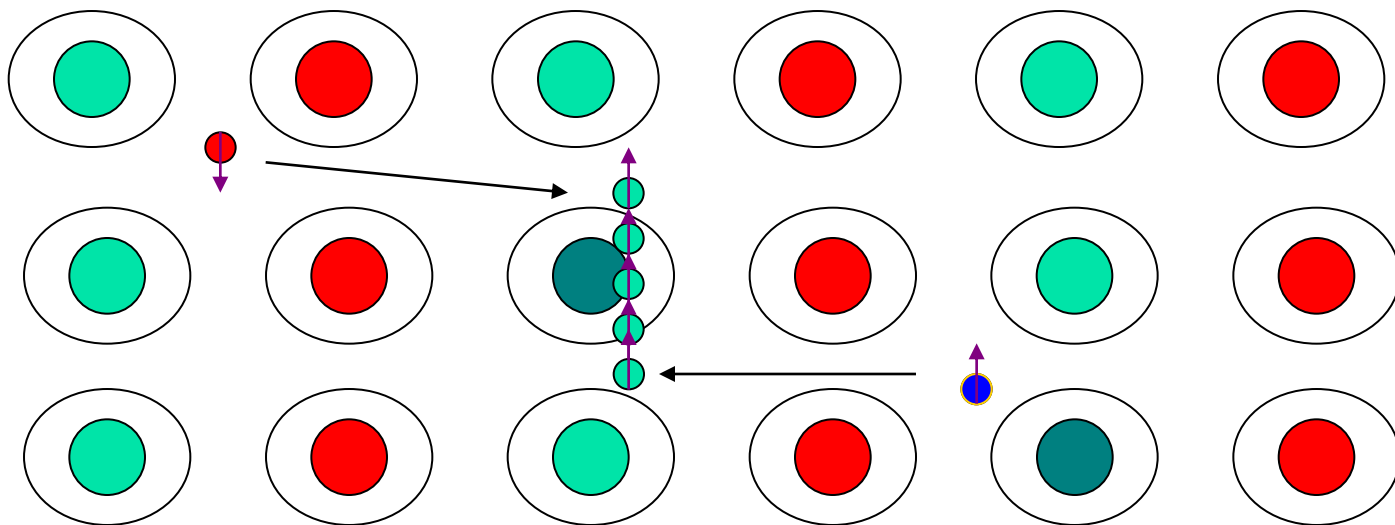
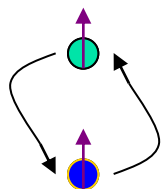
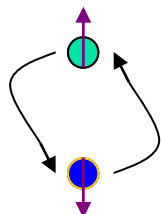
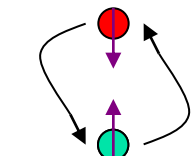
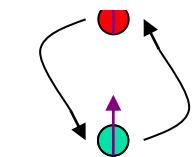


$$H_T = H_0 + H_{\text{ex}}$$

$$= H_0 + \sum_{\mathbf{R}_i} J^{sp-d}(\mathbf{r} - \mathbf{R}_i) \mathbf{S}_i \cdot \boldsymbol{\sigma},$$

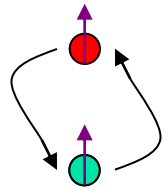
$$H_{\text{ex}} = \sigma_z \langle S_z \rangle \times \sum_{\mathbf{R}} J^{sp-d}(\mathbf{r} - \mathbf{R}),$$

$B//z$



$$H_T = H_0 + H_{ex}$$

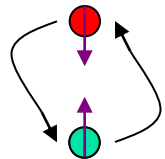
$$= H_0 + \sum_{\mathbf{R}_i} J^{sp-d}(\mathbf{r} - \mathbf{R}_i) \mathbf{S}_i \cdot \boldsymbol{\sigma}, \quad H_{ex} = \sigma_x \langle S_x \rangle x \sum_{\mathbf{R}} J^{sp-d}(\mathbf{r} - \mathbf{R}),$$



CB Γ_6

$$u_{10} = |\frac{1}{2}, \frac{1}{2}\rangle_{\Gamma_6} = S \uparrow,$$

$$u_{20} = |\frac{1}{2}, -\frac{1}{2}\rangle_{\Gamma_6} = S \downarrow;$$

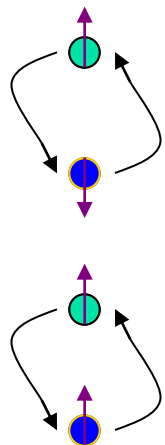


$$u_{30} = |\frac{3}{2}, \frac{3}{2}\rangle = (1/\sqrt{2})(X + iY) \uparrow,$$

$$u_{40} = |\frac{3}{2}, -\frac{3}{2}\rangle = i(1/\sqrt{2})(X - iY) \downarrow,$$

$$u_{50} = |\frac{3}{2}, \frac{1}{2}\rangle = (1/\sqrt{6})[(X - iY) \uparrow + 2Z \downarrow],$$

$$u_{60} = |\frac{3}{2}, -\frac{1}{2}\rangle = i(1/\sqrt{6})[(X + iY) \downarrow - 2Z \uparrow];$$

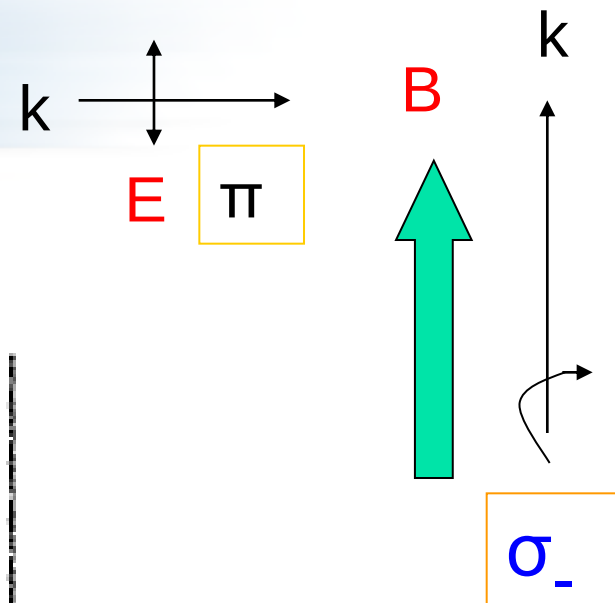


VB Γ_8

$$u_{70} = |\frac{1}{2}, \frac{1}{2}\rangle = -i(1/\sqrt{3})[(X - iY) \uparrow - Z \downarrow],$$

$$u_{80} = |\frac{1}{2}, -\frac{1}{2}\rangle = (1/\sqrt{3})[(X + iY) \downarrow + Z \uparrow].$$

Spin-orbital band Γ_7



$$\langle \Psi_{\Gamma_6} | H_{ex} | \Psi_{\Gamma_6} \rangle = \begin{vmatrix} 3A & 0 \\ 0 & -3A \end{vmatrix}$$

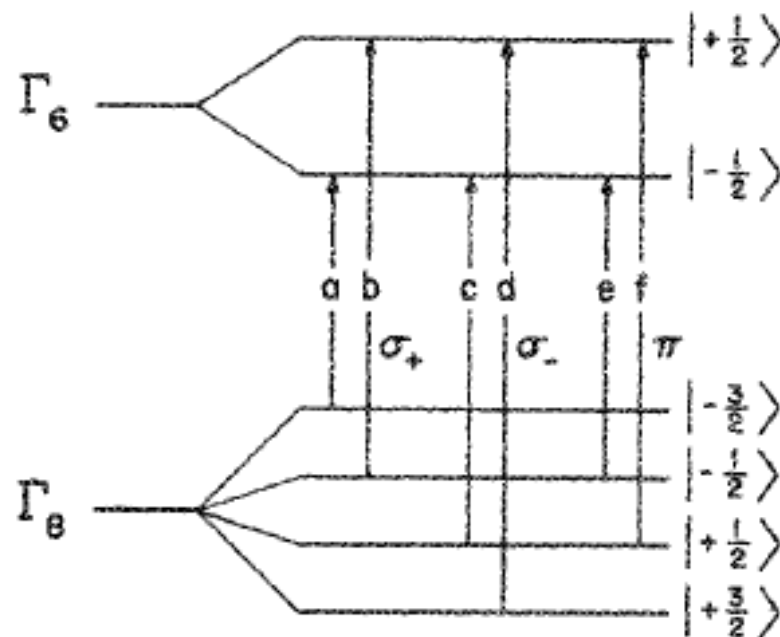
$$\langle \Psi_{\Gamma_8} | H_{ex} | \Psi_{\Gamma_8} \rangle = \begin{vmatrix} 3B & 0 & 0 & 0 \\ 0 & B & 0 & 0 \\ 0 & 0 & -B & 0 \\ 0 & 0 & 0 & -3B \end{vmatrix}$$

$$A = \frac{1}{6} N_0 \alpha x \langle S_z \rangle = -\frac{1}{6} \frac{\alpha M}{g_{Mn} \mu_B},$$

$$B = \frac{1}{6} N_0 \beta x \langle S_z \rangle = -\frac{1}{6} \frac{\beta M}{g_{Mn} \mu_B}$$

$$\alpha = \langle S | J^{sp-d} | S \rangle / \Omega_0,$$

$$\beta = \langle X | J^{sp-d} | X \rangle / \Omega_0.$$

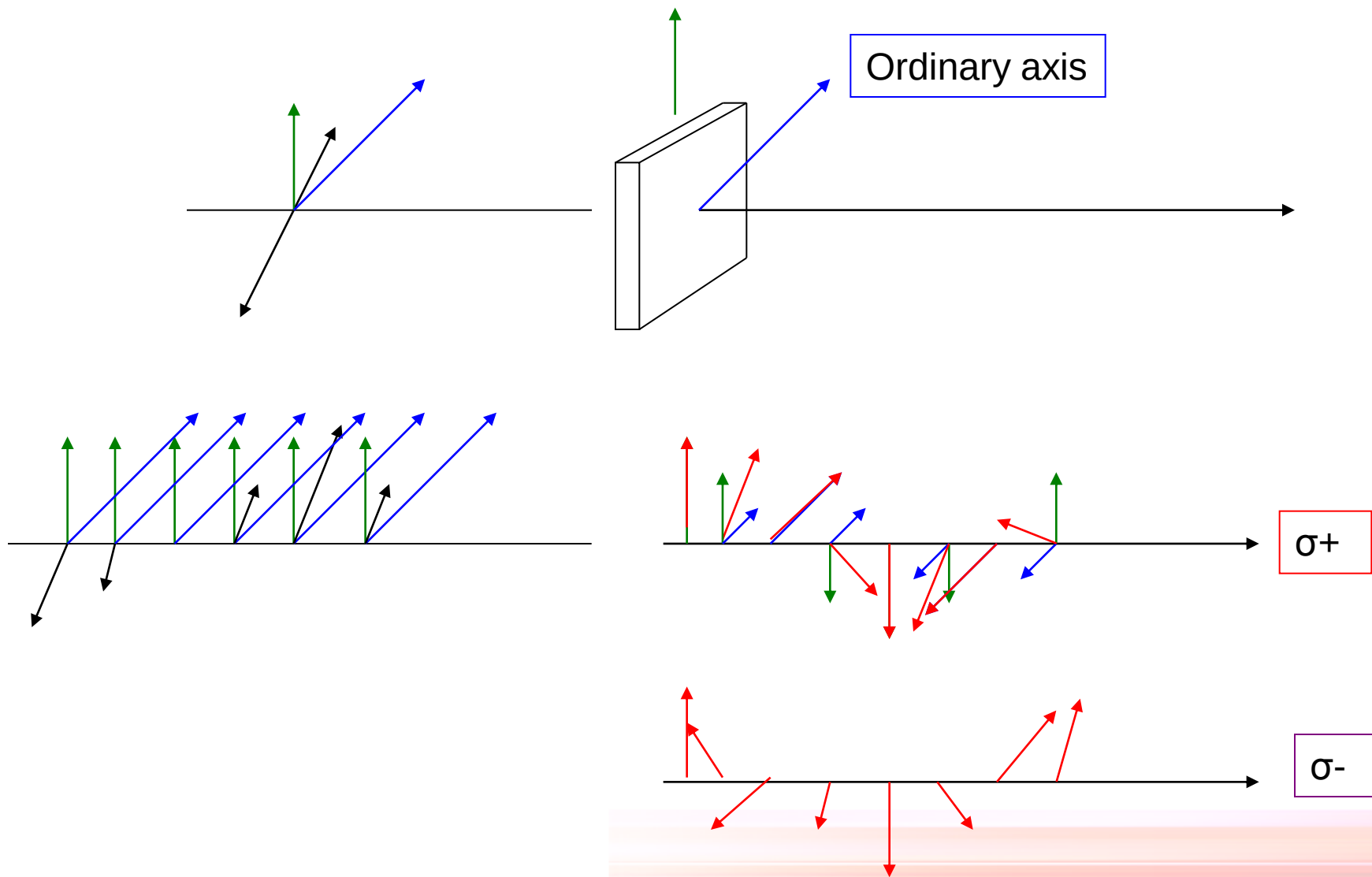




如何產生圓形極化光?

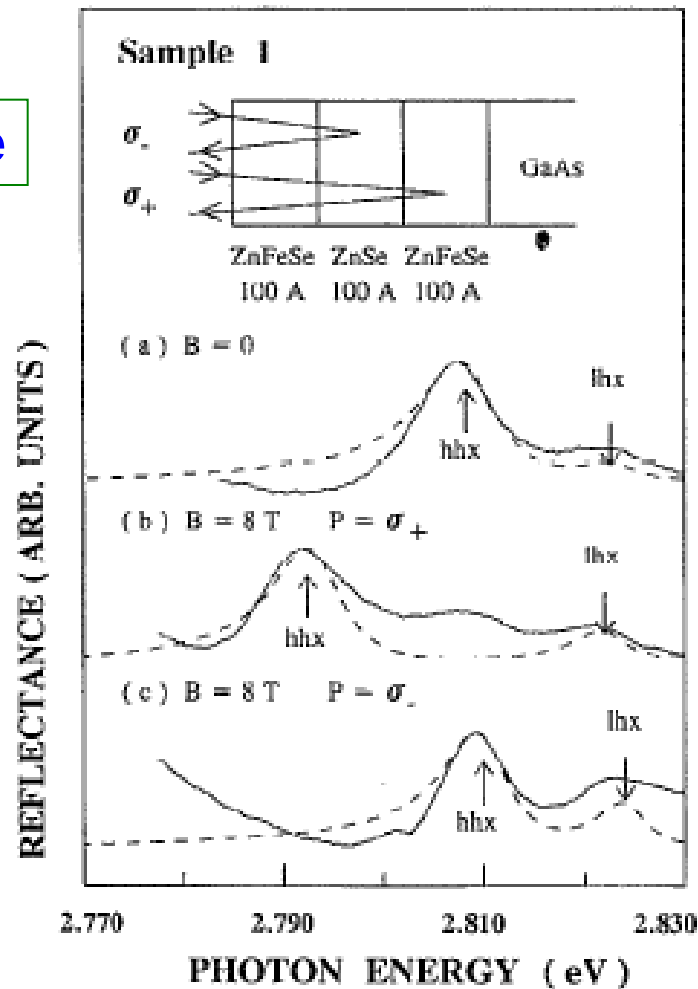
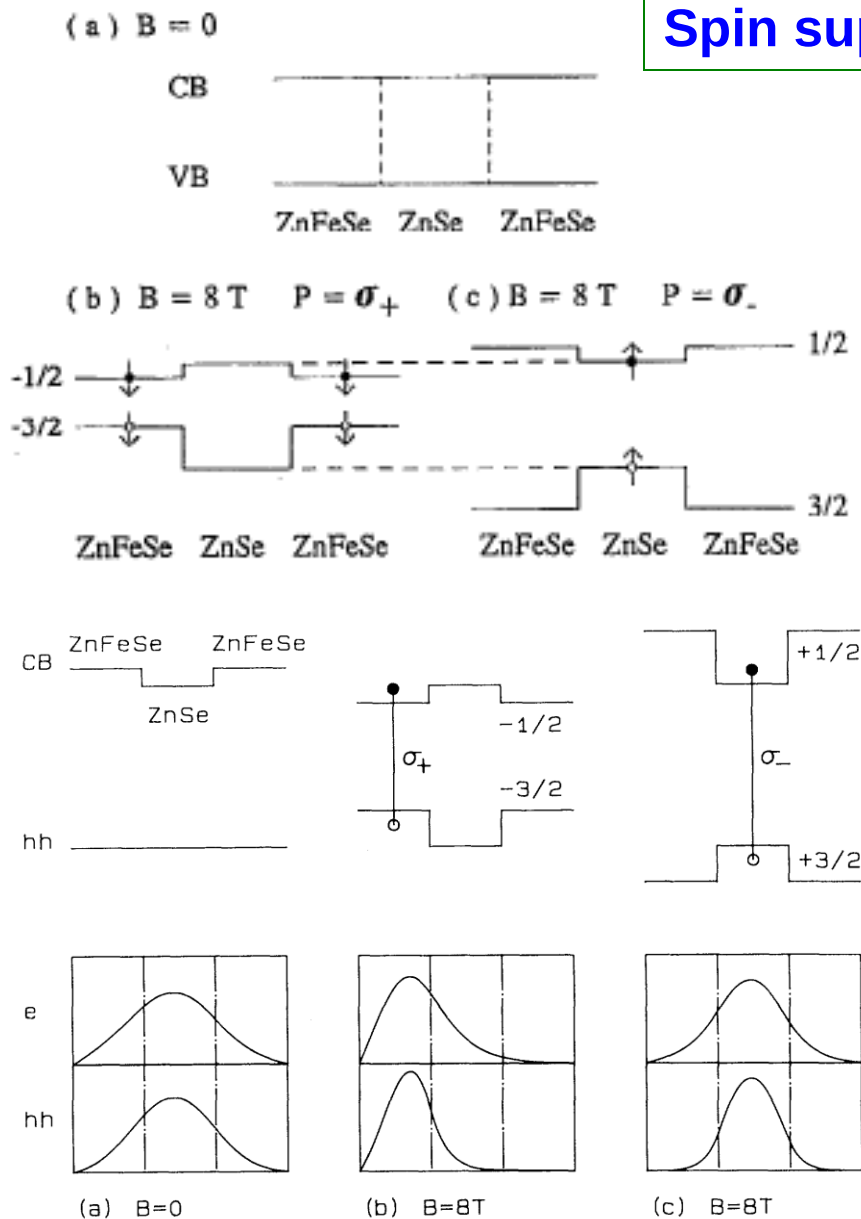
Extra-ordinary axis

Ordinary axis





Spin super-lattice



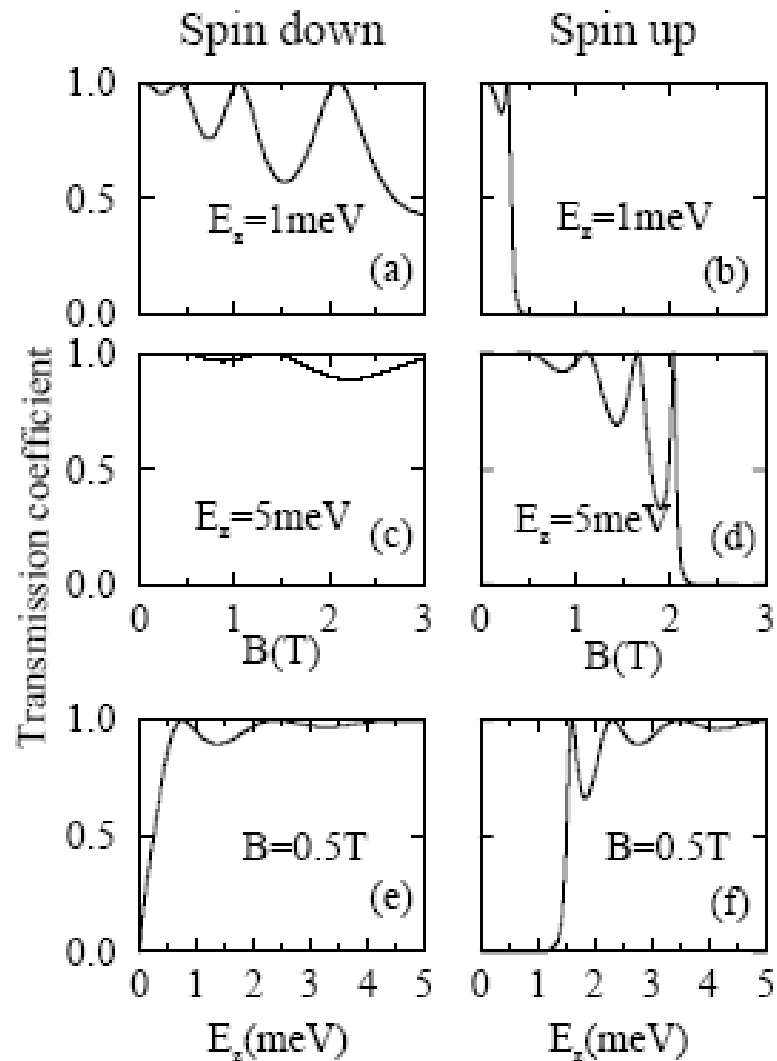
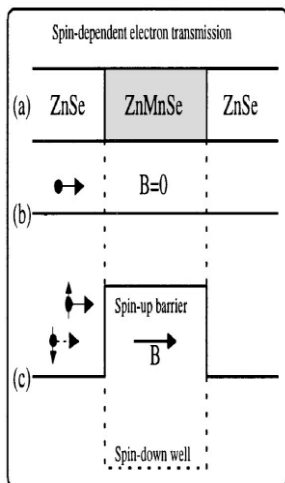
$$R = \left| \frac{E'_0}{E_0} \right|^2 \quad \epsilon = \epsilon_\infty + \sum_\alpha \frac{A_\alpha}{(E_\alpha^2 - E^2) - i\Gamma_\alpha E}$$

PRL 67, 3820 (1991),
JAP 75, 2988 (1994)



Spin-Dependent Perpendicular Magnetotransport through a Tunable ZnSe/Zn_{1-x}Mn_xSe Heterostructure: A Possible Spin Filter?

J. Carlos Egues, PRL80, 4578 (1998)



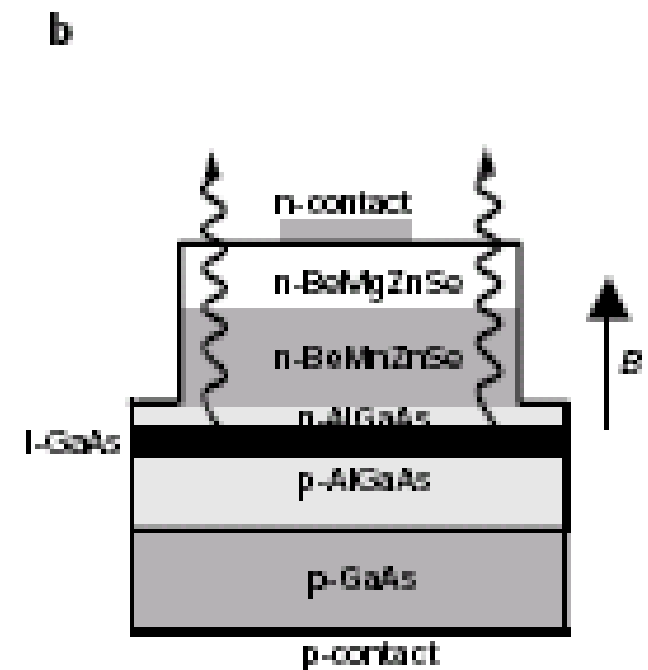
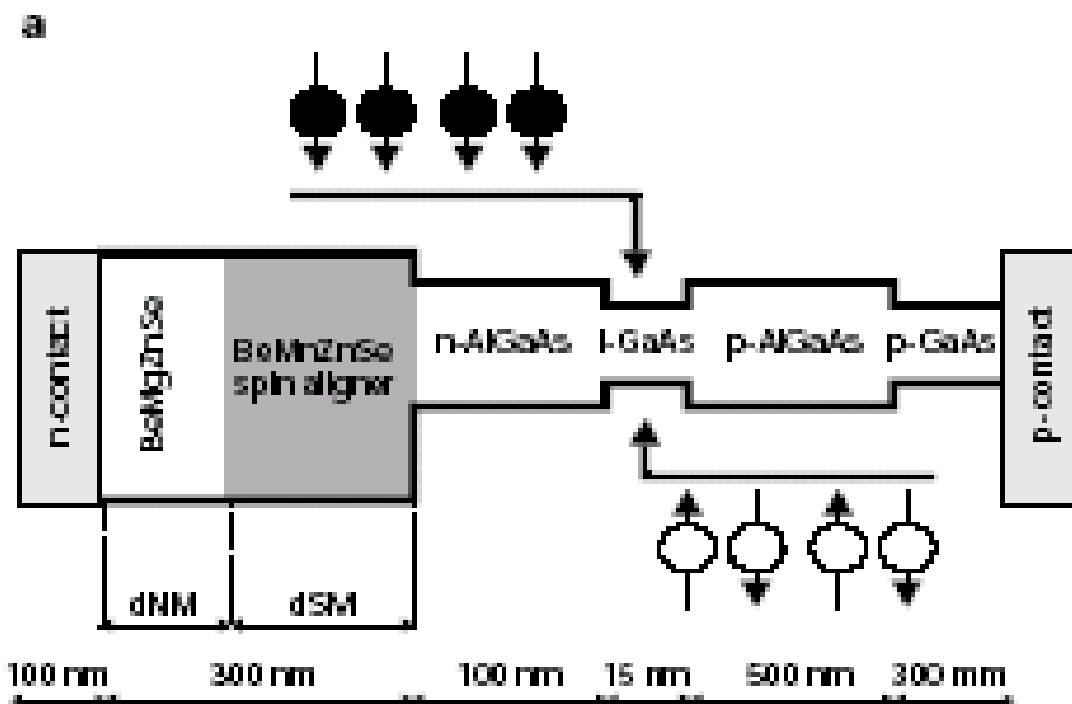
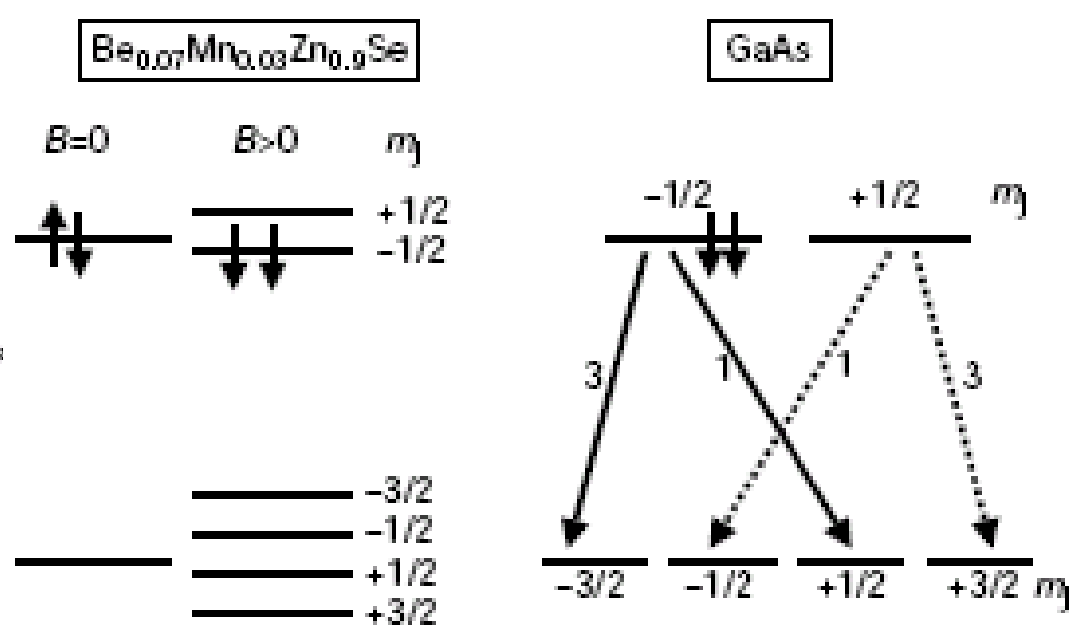
$$T_l(E_z, B) = \left\{ 1 + \frac{\sin^2 \left[\sqrt{\frac{2m_e^*(x|\langle S_z \rangle|N_0\alpha/2 + E_z)}{\hbar^2}} L \right]}{4 \left(\frac{E_z}{x|\langle S_z \rangle|N_0\alpha/2} \right) \left(\frac{E_z}{x|\langle S_z \rangle|N_0\alpha/2} + 1 \right)} \right\}^{-1} \quad (1)$$

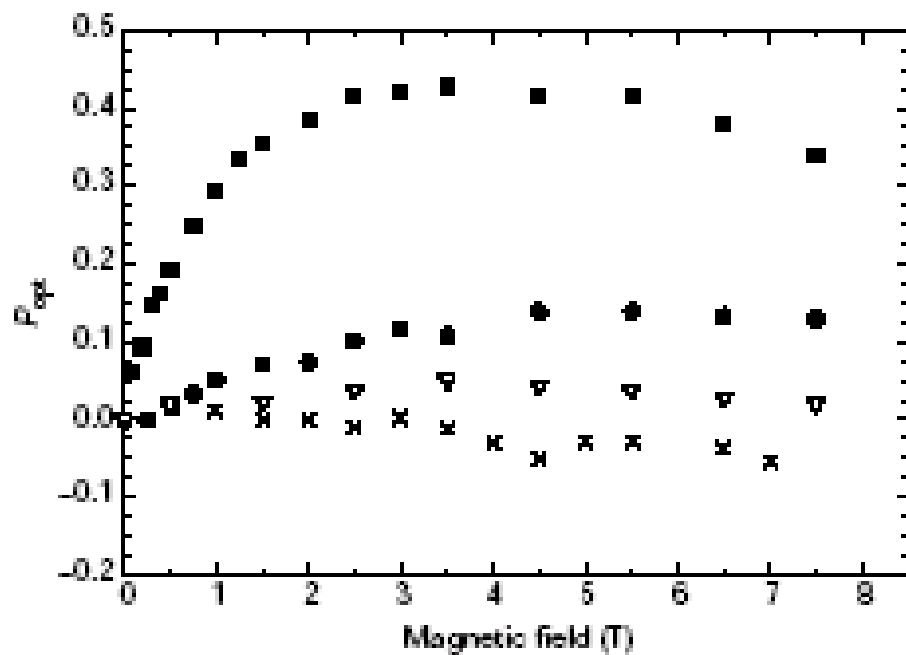
Injection and detection of a spin-polarized current in a light-emitting diode

R. Fiederling, M. Keim, G. Reuscher, W. Ossau, G. Schmidt, A. Waag & L. W. Molenkamp

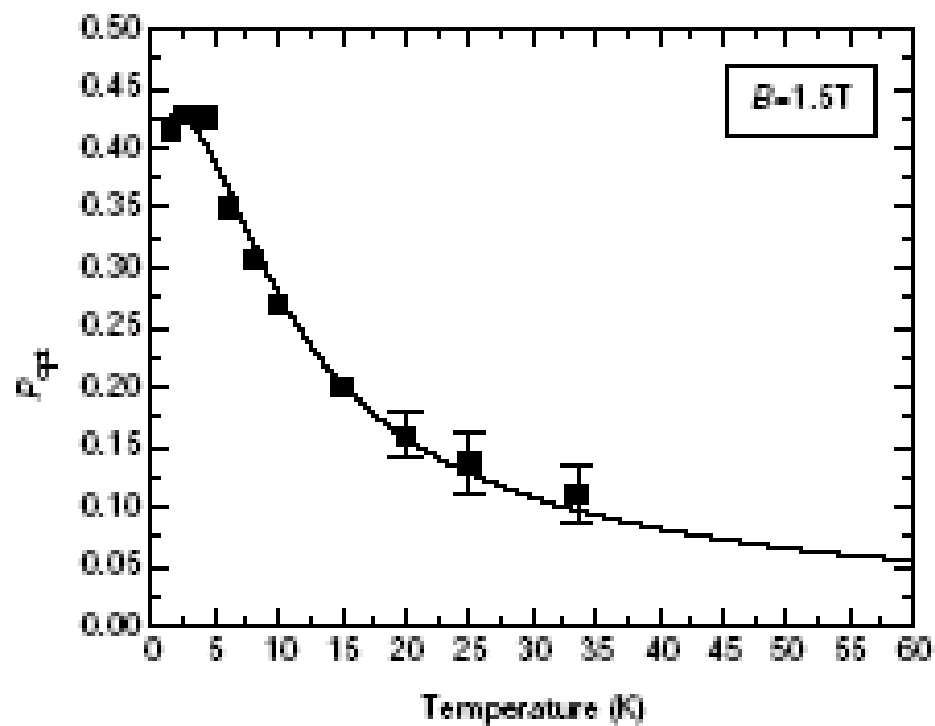
Physikalisches Institut, EP III, Universität Würzburg, 97074 Würzburg, Germany

Nature 402, p787 (1999)





Nature 402, p787 (1999)



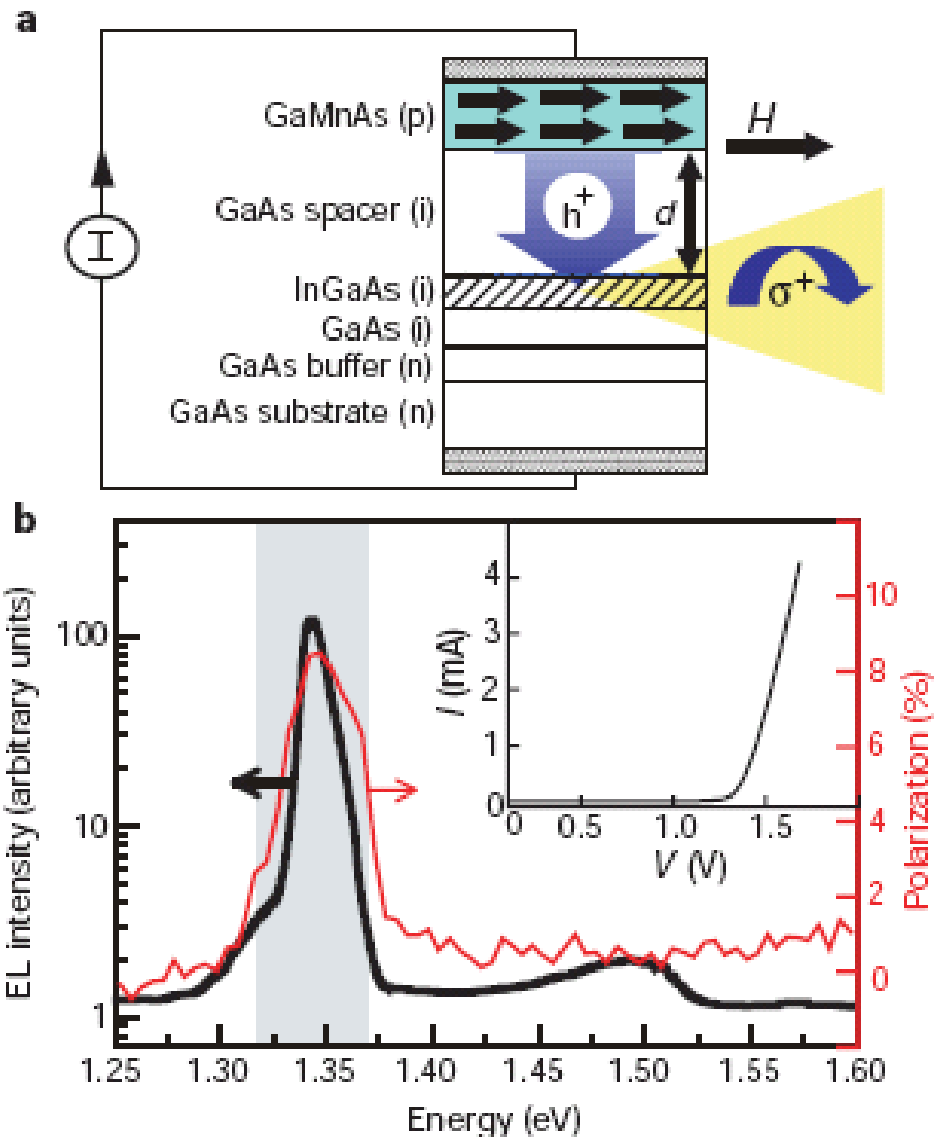
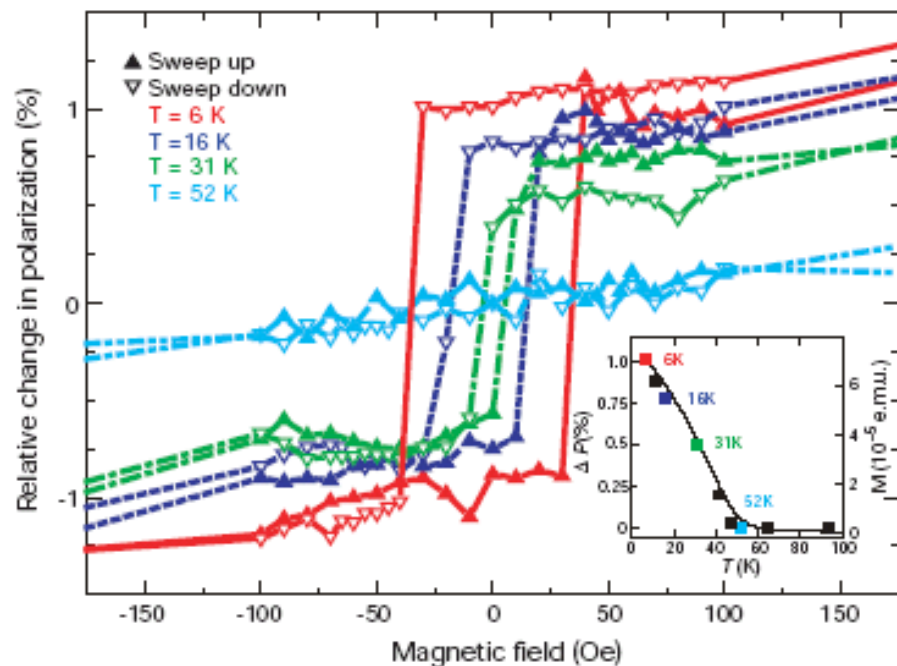
Electrical spin injection in a ferromagnetic semiconductor heterostructure

Y. Ohno*, D. K. Young†, B. Beschoten†, F. Matsukura*, H. Ohno*
& D. D. Awschalom†

* Laboratory for Electronic Intelligent Systems, Research Institute of Electrical
Communication, Tohoku University, Katahira 2-1-1, Aoba-ku, Sendai 980-8577,
Japan

† Center for Spintronics and Quantum Computation, Quantum Institute,
University of California, Santa Barbara, California 93106, USA

Nature 402, p790 (1999)



A magnetic-field-effect transistor and spin transport

R. N. Gurzhi, A. N. Kalinenko, A. I. Kopeliovich, and A. V. Yanovsky

B. Verkin Institute for Low Temperature Physics & Engineering of the National Academy of Sciences of the Ukraine, 47 Lenin Ave, Kharkov, 61103, Ukraine

E. N. Bogachek and Uzi Landman^{a)}

School of Physics, Georgia Institute of Technology, Atlanta, Georgia 30332-0430

(Received 19 May 2003; accepted 7 October 2003)

A magnetic-field-effect transistor is proposed that generates a spin-polarized current and exhibits a giant negative magnetoresistance. The device consists of a nonmagnetic conducting channel (wire or strip) wrapped, or sandwiched, by a grounded magnetic shell. The process underlying the operation of the device is the withdrawal of one of the spin components from the channel, and its dissipation through the grounded boundaries of the magnetic shell, resulting in a spin-polarized current in the nonmagnetic channel. The device may generate an almost fully spin-polarized current, and a giant negative magnetoresistance effect is predicted. © 2003 American Institute of Physics. [DOI: 10.1063/1.1630839]

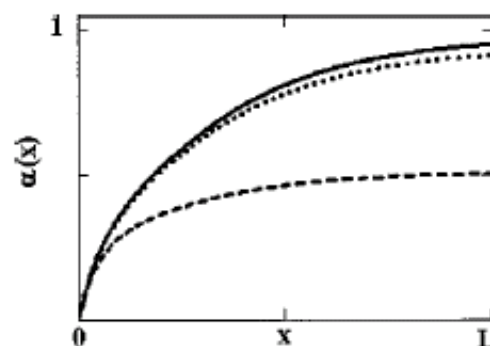
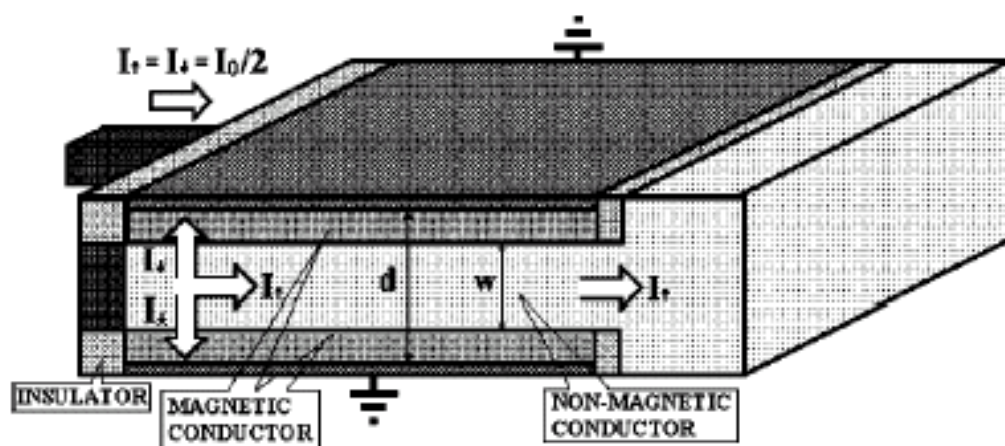
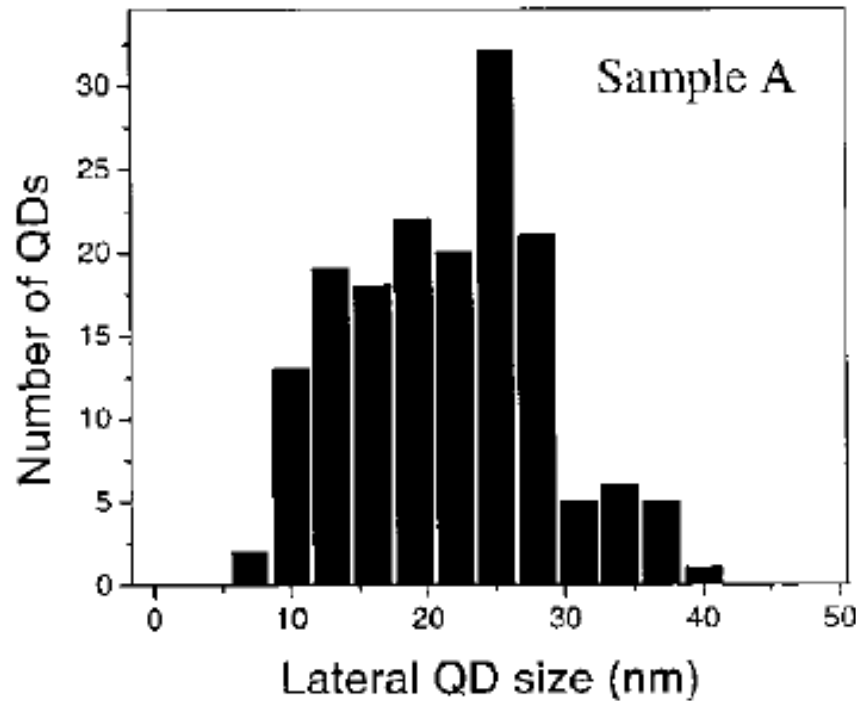


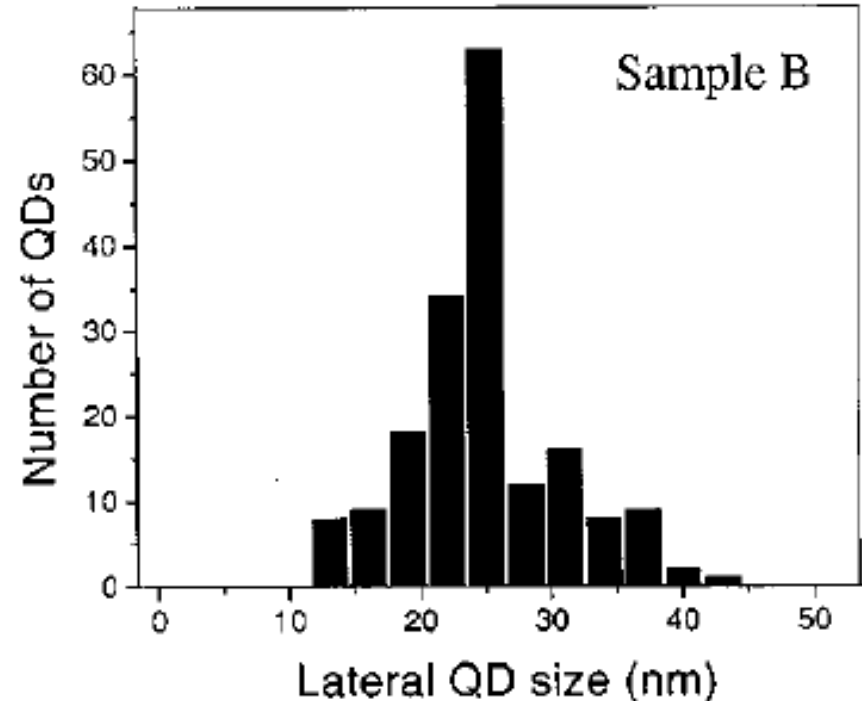
FIG. 2. The degree of SP (α) of the current plotted vs the longitudinal (x) coordinate along the spin guide; the curves were calculated from Eq. (7) with $\sigma_{M1}/\sigma_{M1}=0.3$, $\sigma_{M1}/\sigma_N=1$, $w/d=0.28$, $L=4d$, $w_M/\lambda_M=0.225$, and $w/\lambda_N=0.1$ (solid), $w/\lambda_N=0.5$ (dotted), $w/\lambda_N=0.7$ (dashed).

Epitaxially grown magnetic semiconductor nanostructures

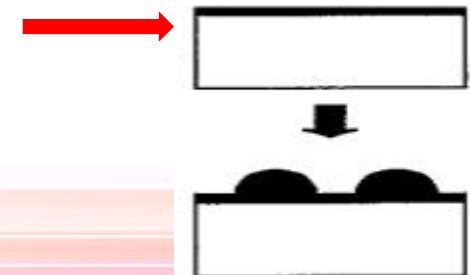
CdSe/ZnSe QDs



CdSe/ZnSe QDs

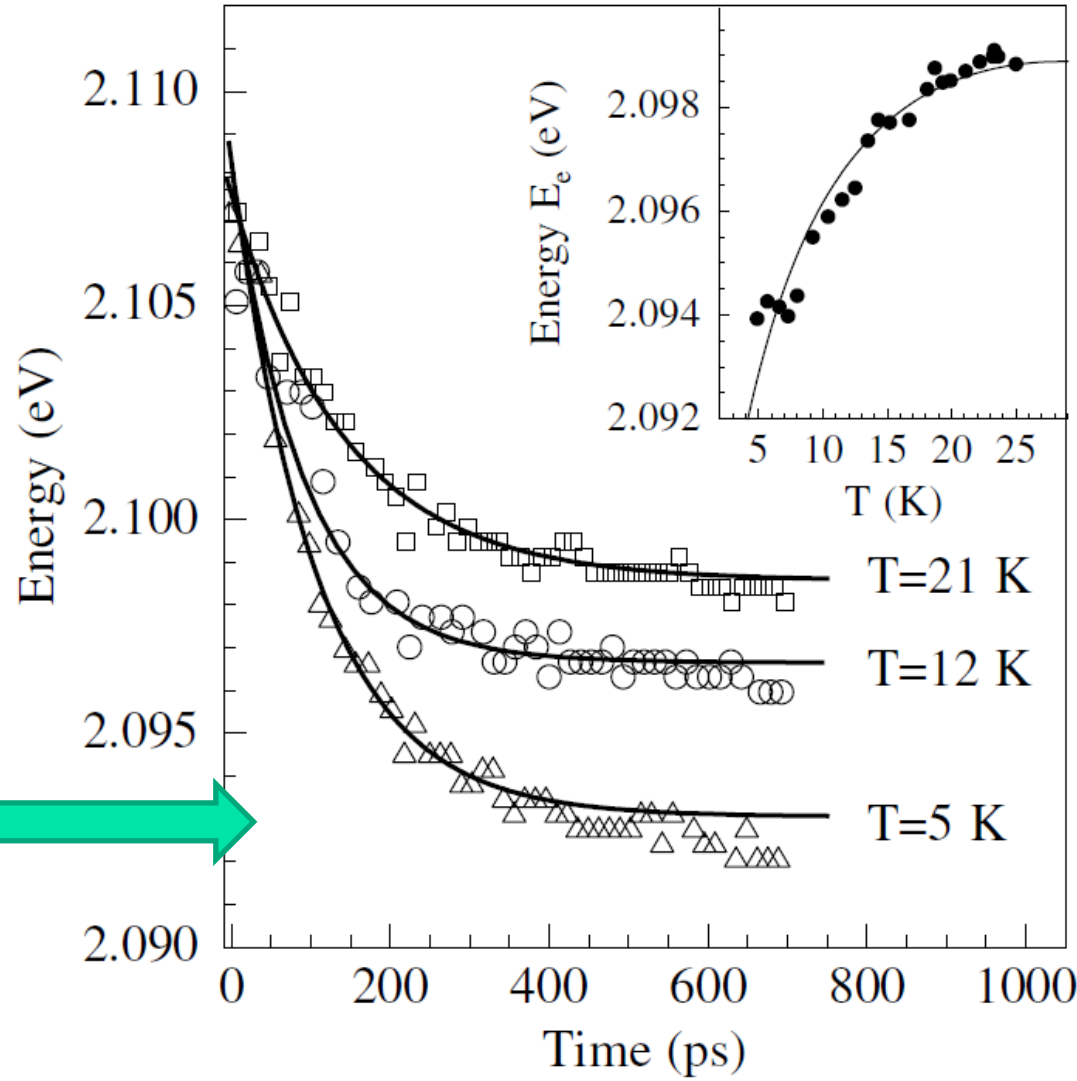


Magnetic CdSe-based quantum dots grown on Mn-passivated ZnSe



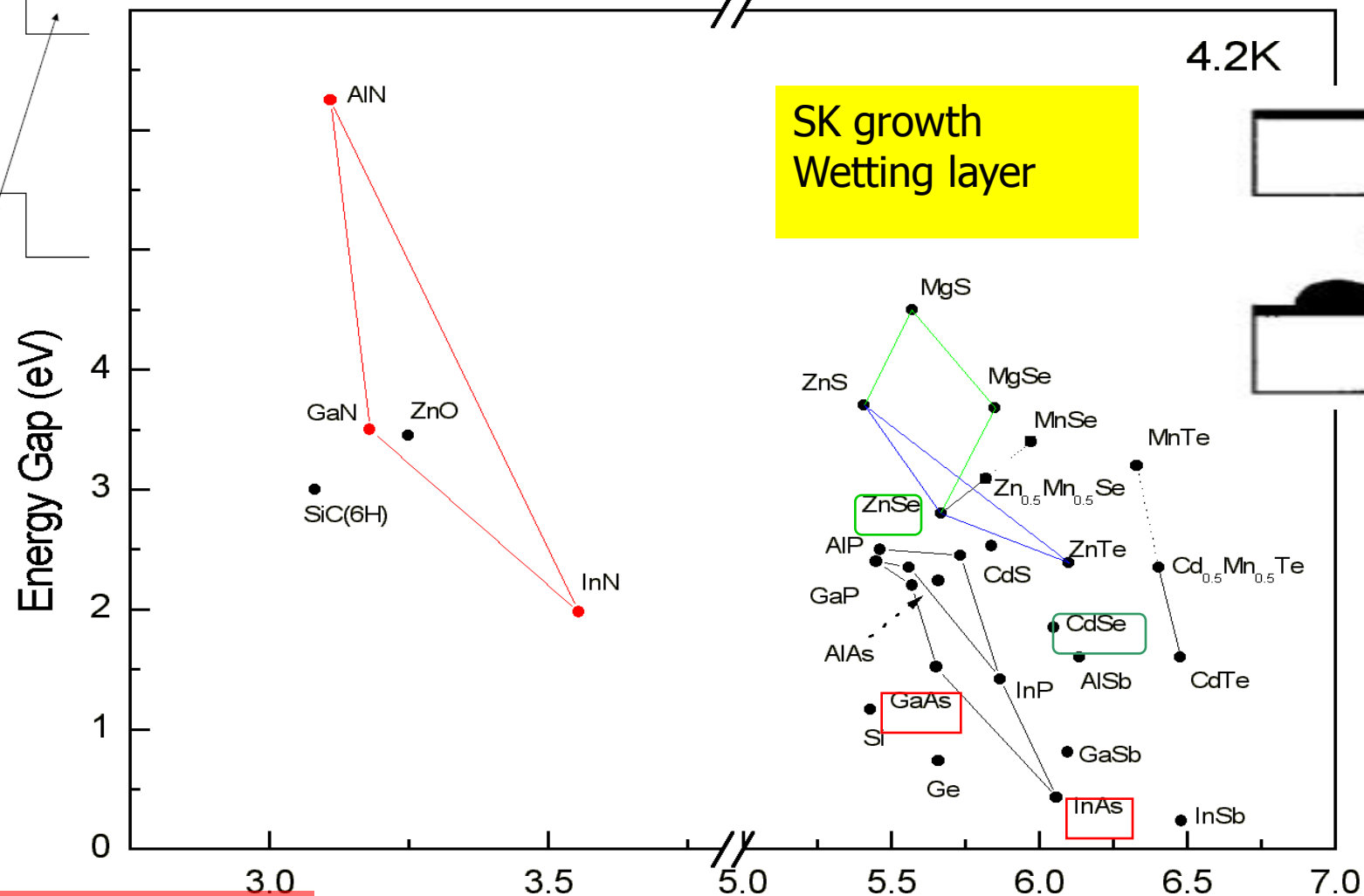
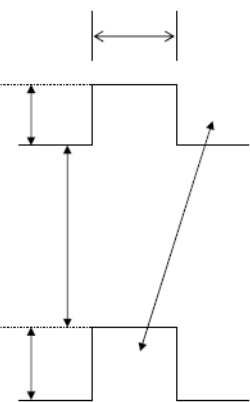
Dynamical Spin Response in Semimagnetic Quantum Dots
Prof. JK Furdyana's group

Formation of magnetic polaron in CdSe/ZnMnSe QDs



Their results from the magnetic semiconductor nanostructures are very fruitful.

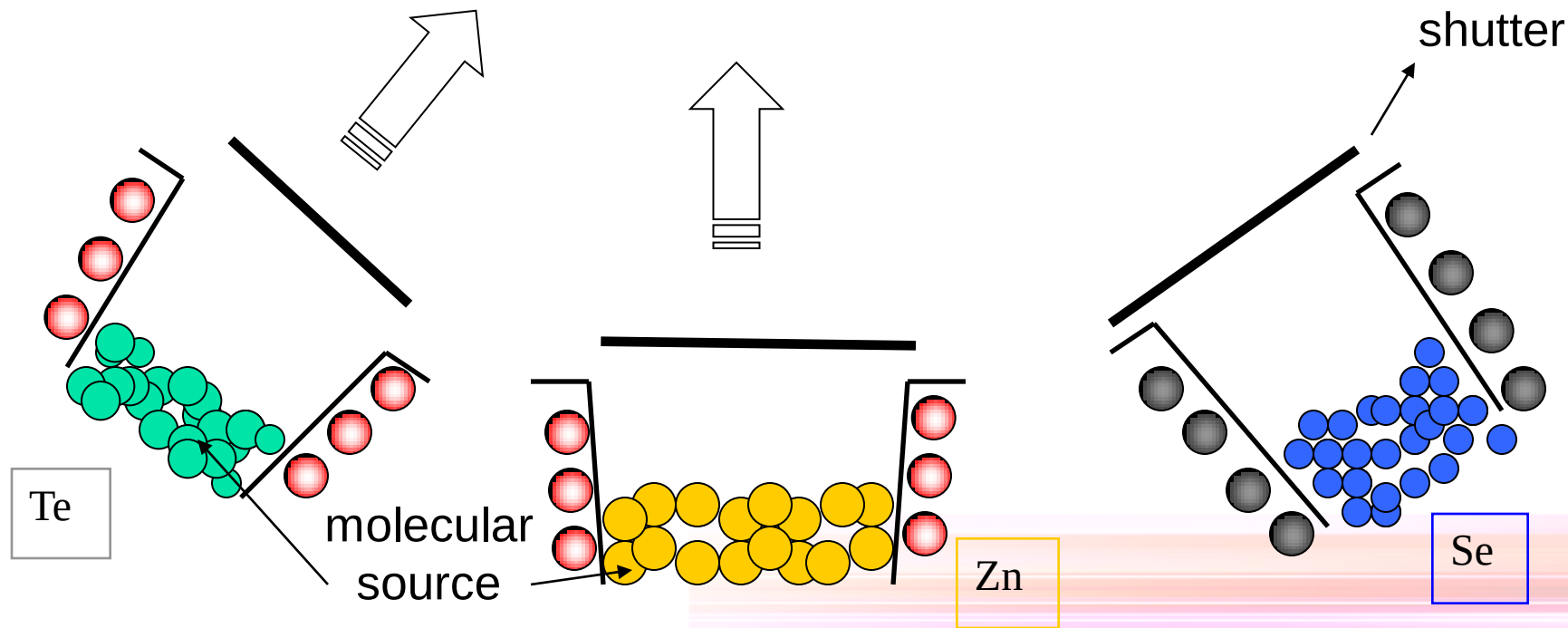
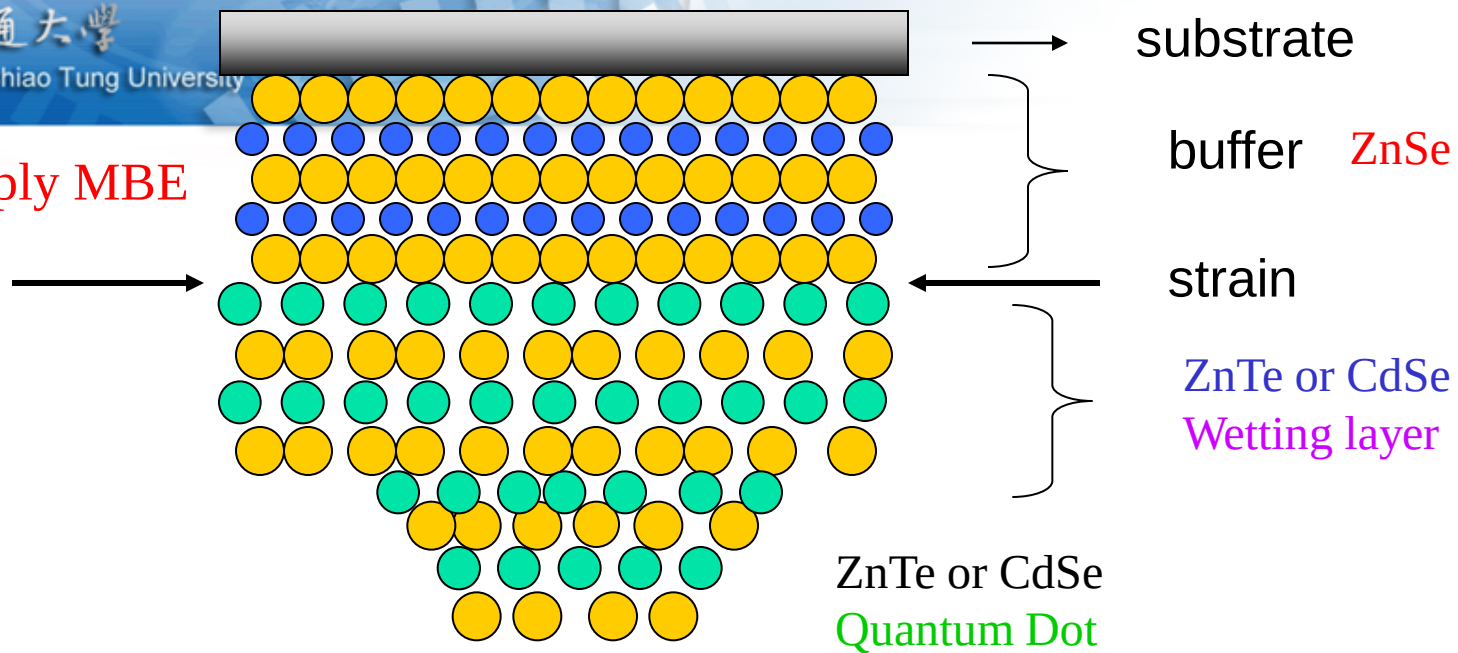
Energy Gap vs Lattice Constant



Type II ZnTe/ZnSe
GaSb/GaAs

InAs/GaAs, CdSe/ZnSe

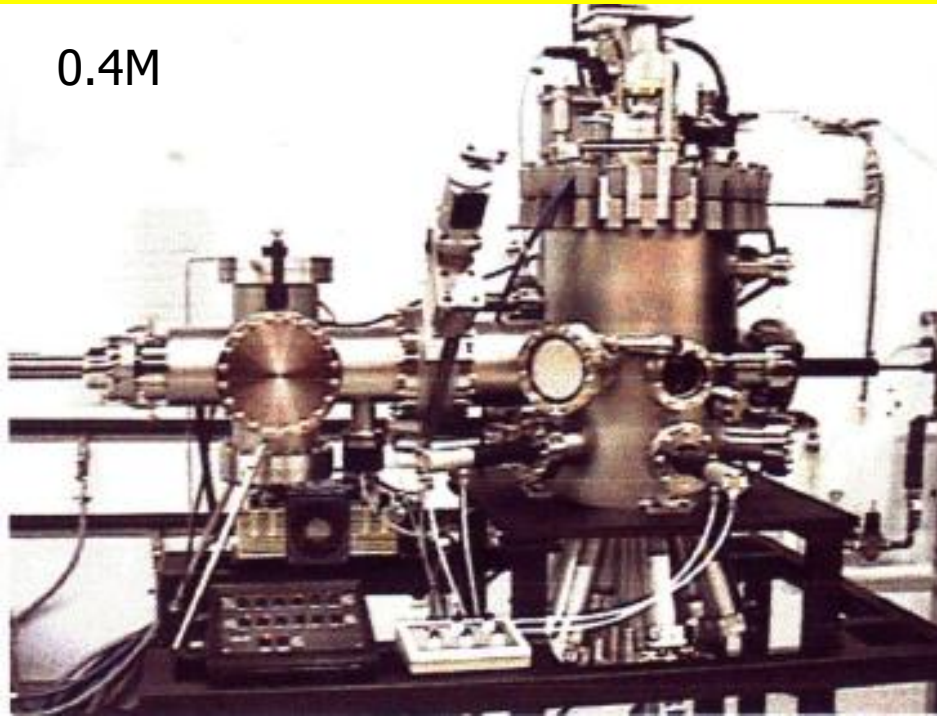
Alternating supply MBE



How to realize the growth of ZnMnTe QDs on ZnSe?

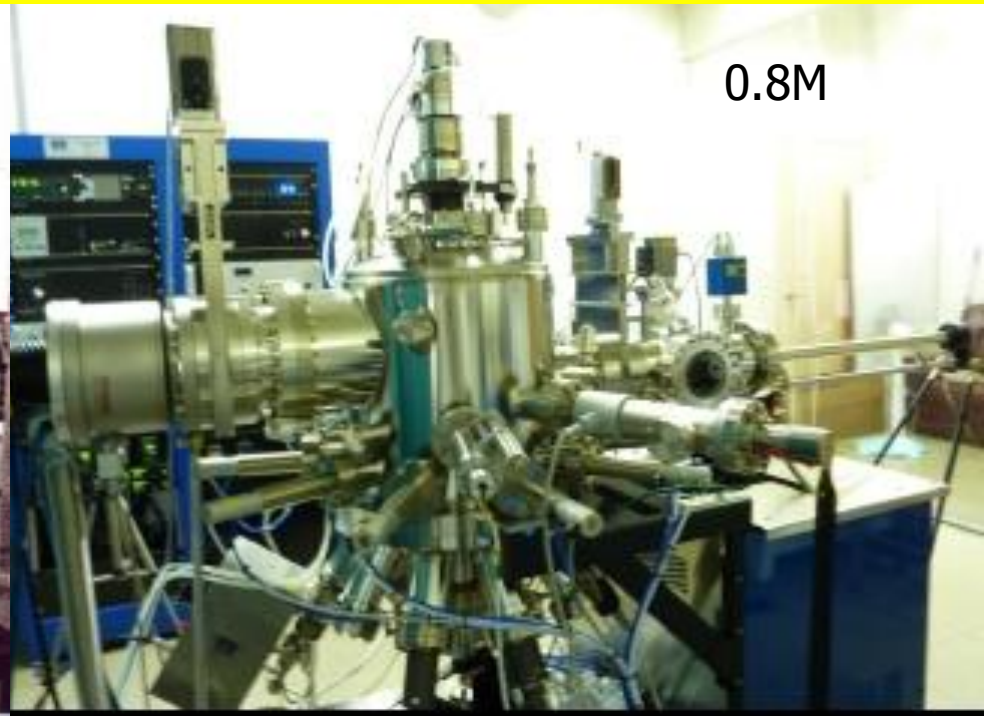
What kind of type-II quantum dots (QDs)? How to grow them?

0.4M



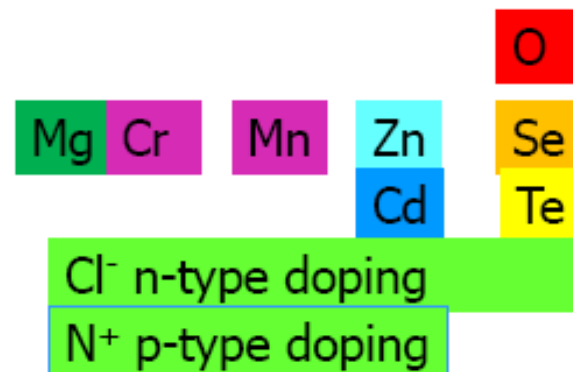
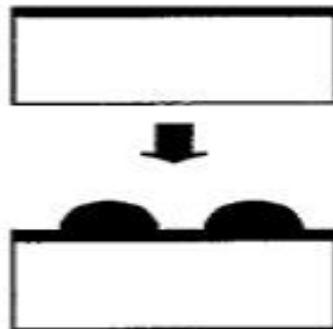
Veeco EPI 620
molecular beam epitaxy
(MBE) system

0.8M



SVT (MBE) system

Self-assembled
Quantum dots (QDs)

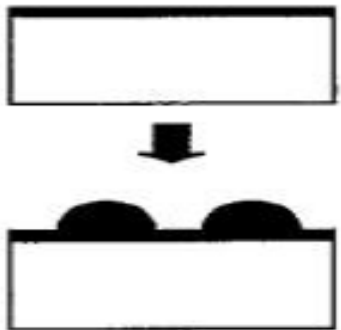
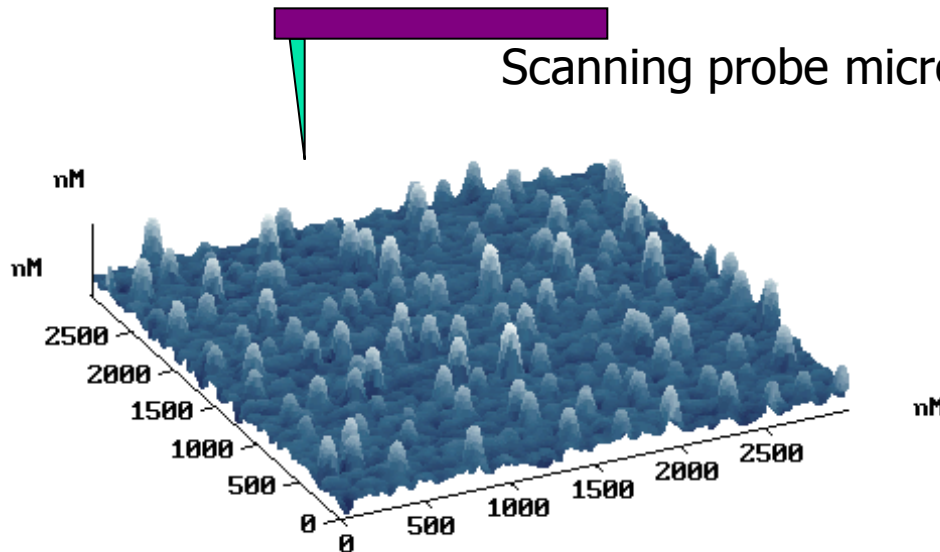


It's very important to have a smooth buffer layer.

National Chiao Tung University

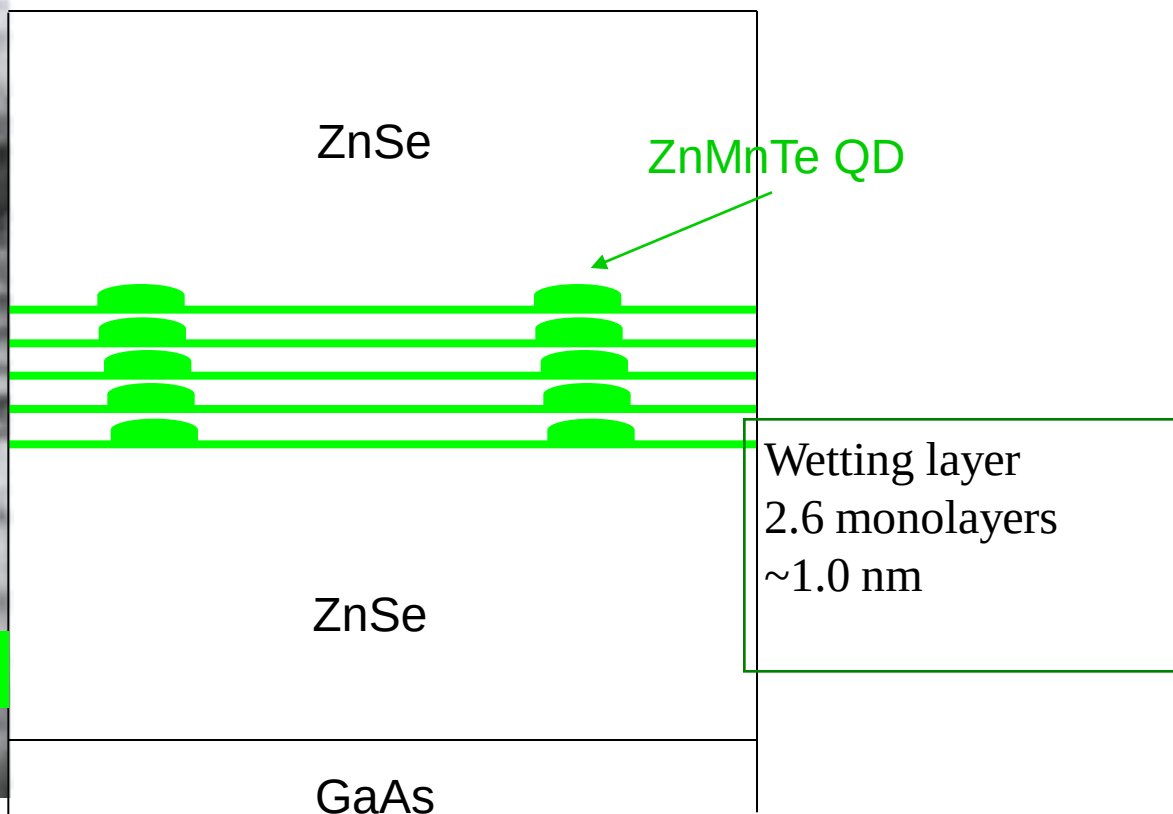
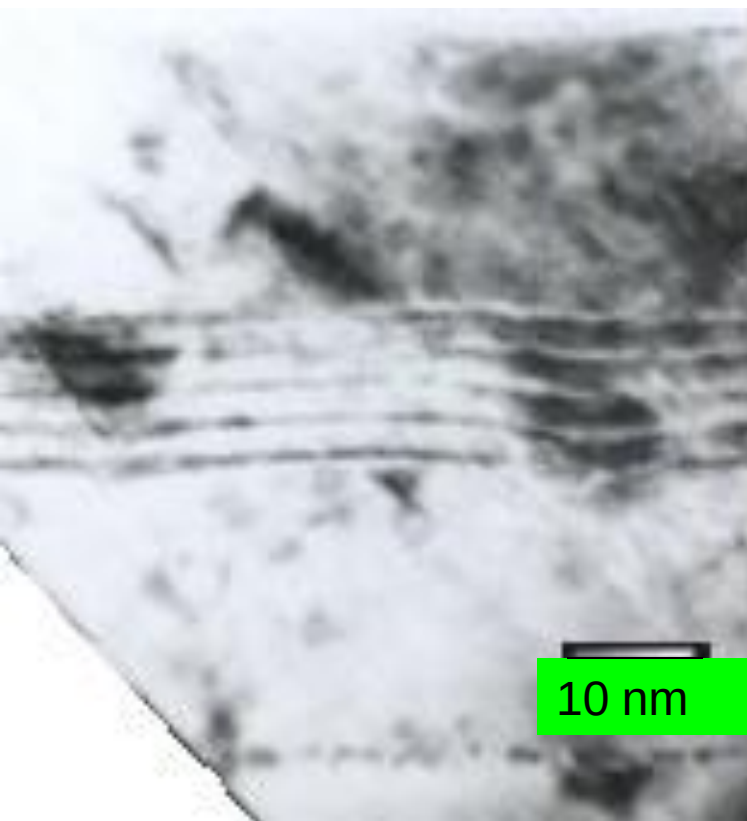
Atomic force microscopy, AFM

Scanning probe microscopy





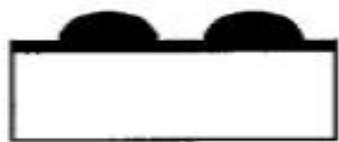
Cross-section TEM of 2.6 ML ZnMnTe MQDs



Stranski-Krastanow (SK), 2-D to 3D (0D) growth mode

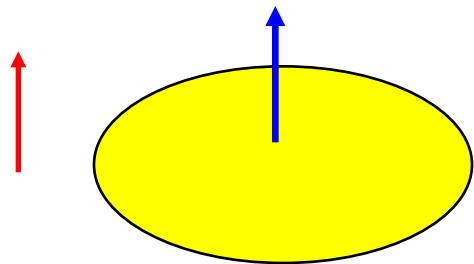
Vertical correlation.

Magnetic semiconductor
QD (type-II QD)

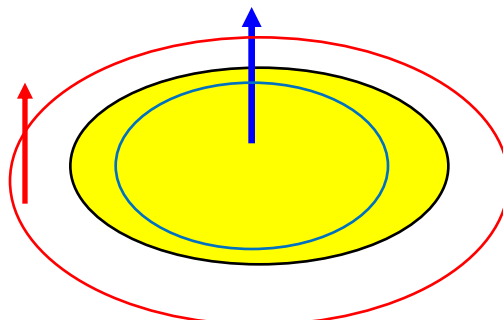


electron

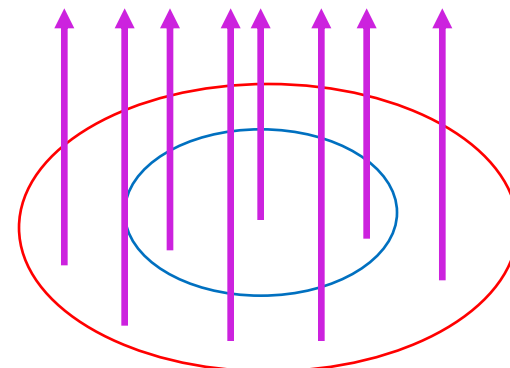
hole



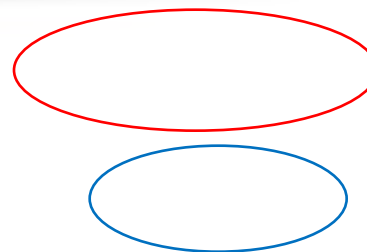
Magnetic field $B=0$



Magnetic field $B>0$



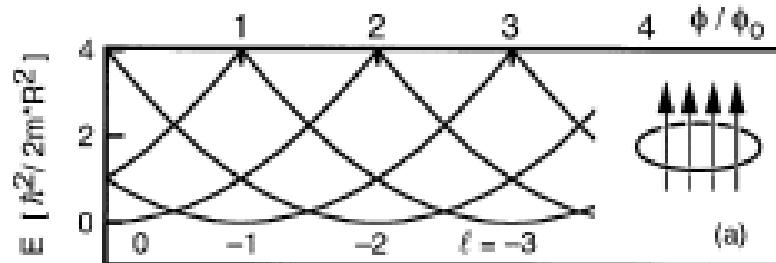
Magnetic field $B>0$



Electron orbit

Hole orbit

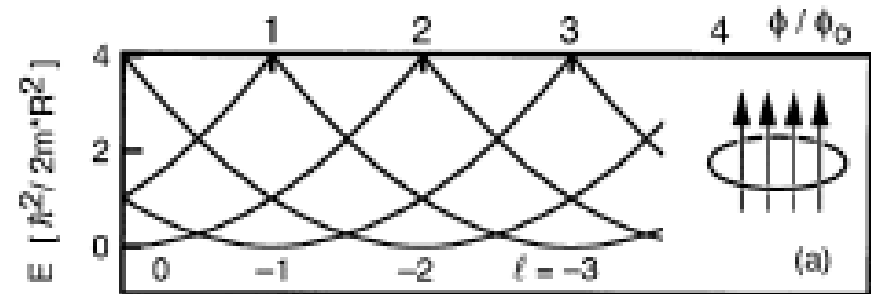
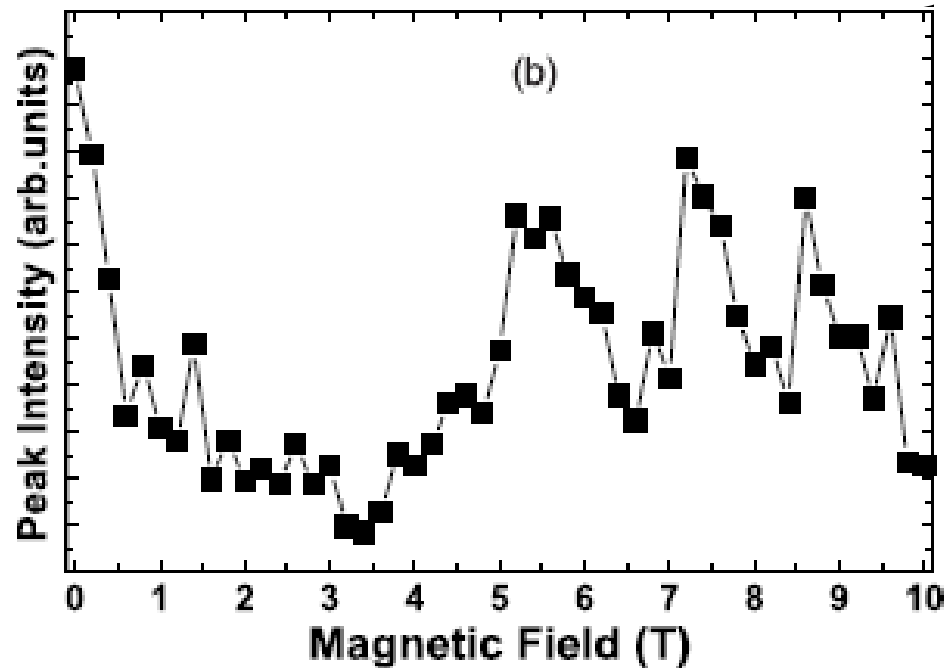
Magnetic flux $\Delta\phi$
 ϕ_0 = flux quanta



$$E_{\text{exc}} = E_g + \frac{\hbar^2}{2MR_0^2} \left(L + \frac{\Delta\Phi}{\Phi_0} \right)^2,$$

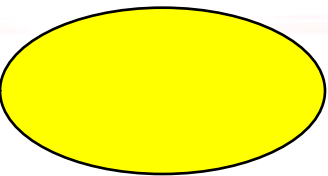
Observation of optical Aharonov-Bohm oscillation

Coherent Aharonov-Bohm oscillation in ZnMnTe/ZnSe quantum dots PRB77, 241302(2008)

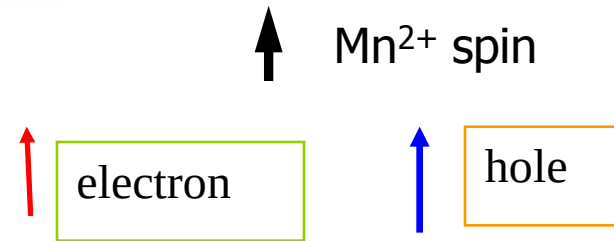


$$E_{\text{exc}} = E_g + \frac{\hbar^2}{2MR_0^2} \left(L + \frac{\Delta\Phi}{\Phi_0} \right)^2$$

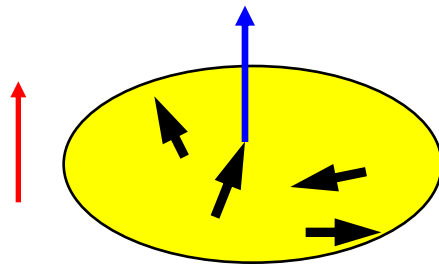
Observation of optical Aharonov-Bohm oscillation



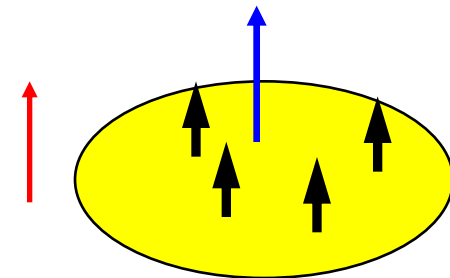
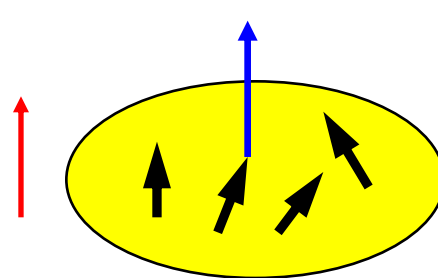
Magnetic semiconductor
QD



Time resolved photoluminescence is sensitive to probe MP.



initially $t=0$



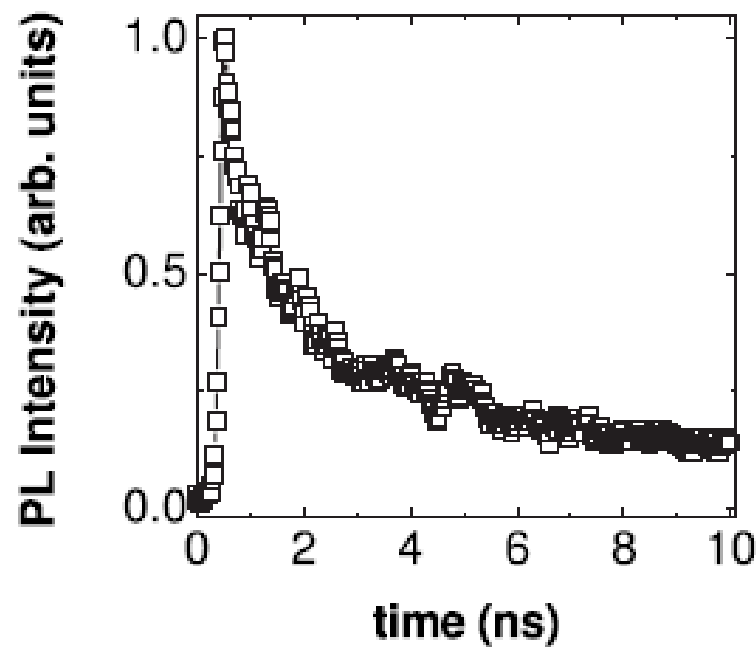
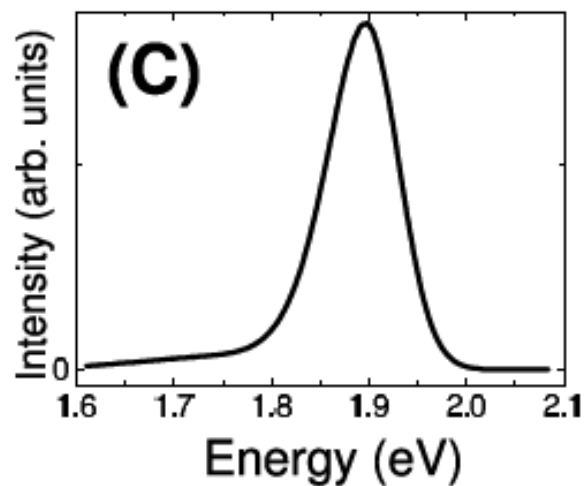
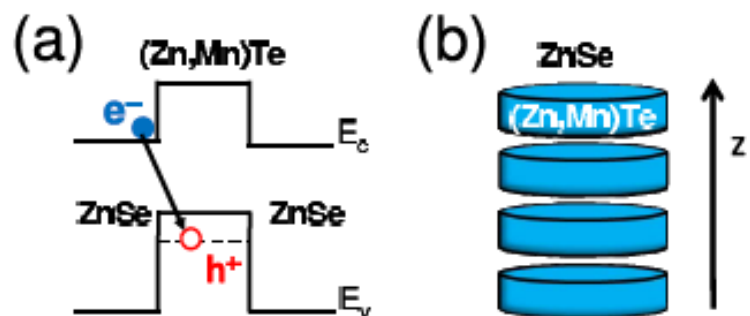
$t>0$

Right after pulse laser
excitation

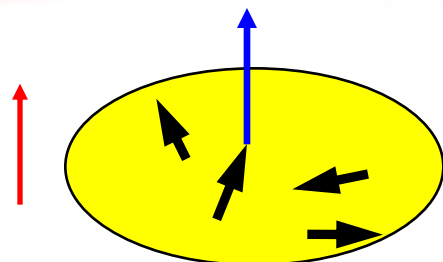
Exchange interaction between hole spin and
manganese ion spin

Exciton emission energy decreases with time.

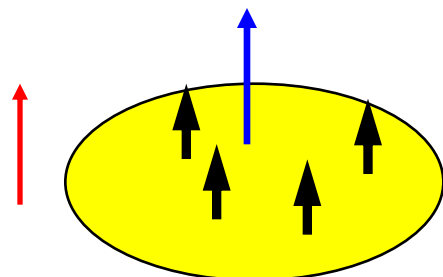
Formation of magnetic polaron (MP).



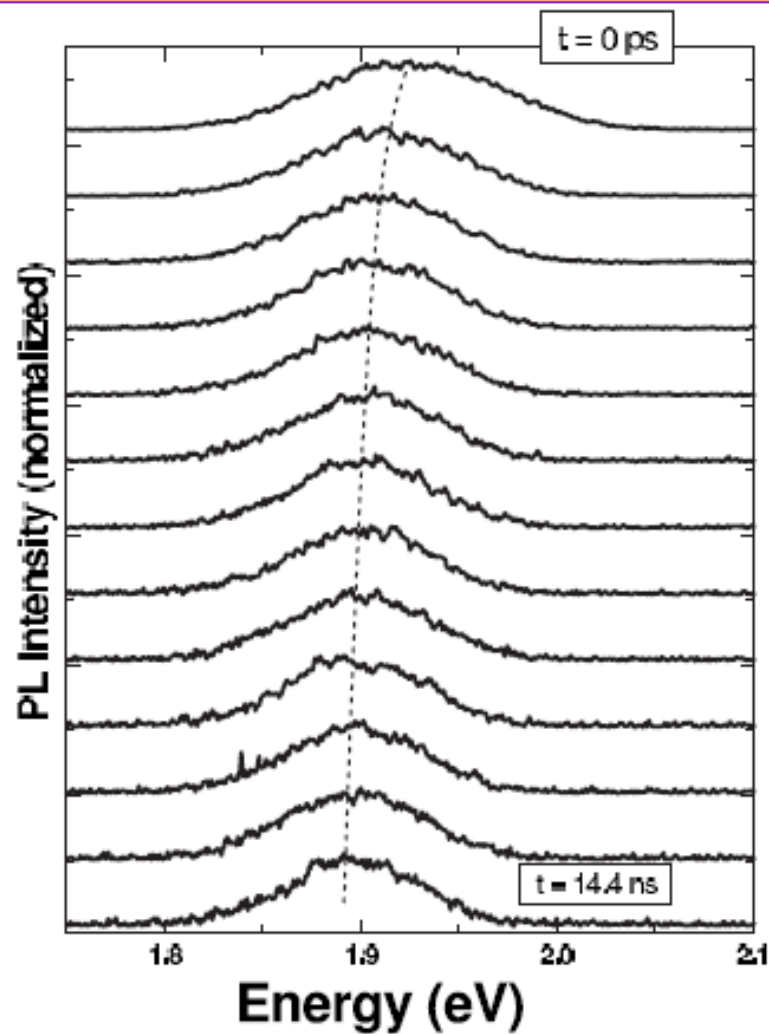
Photoluminescence (PL) spectrum of ZnMnTe/ZnSe QDs.



initially $t=0$

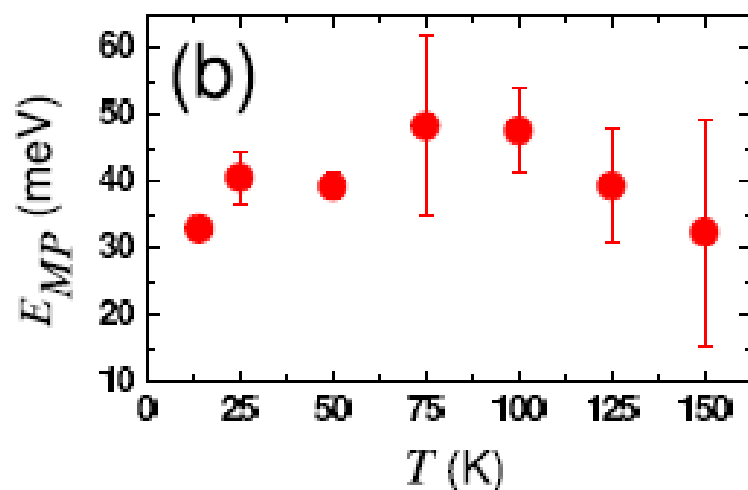
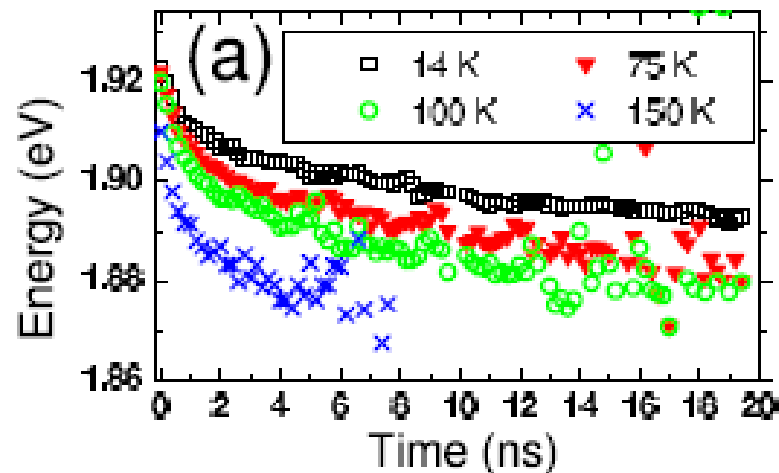


$t>0$



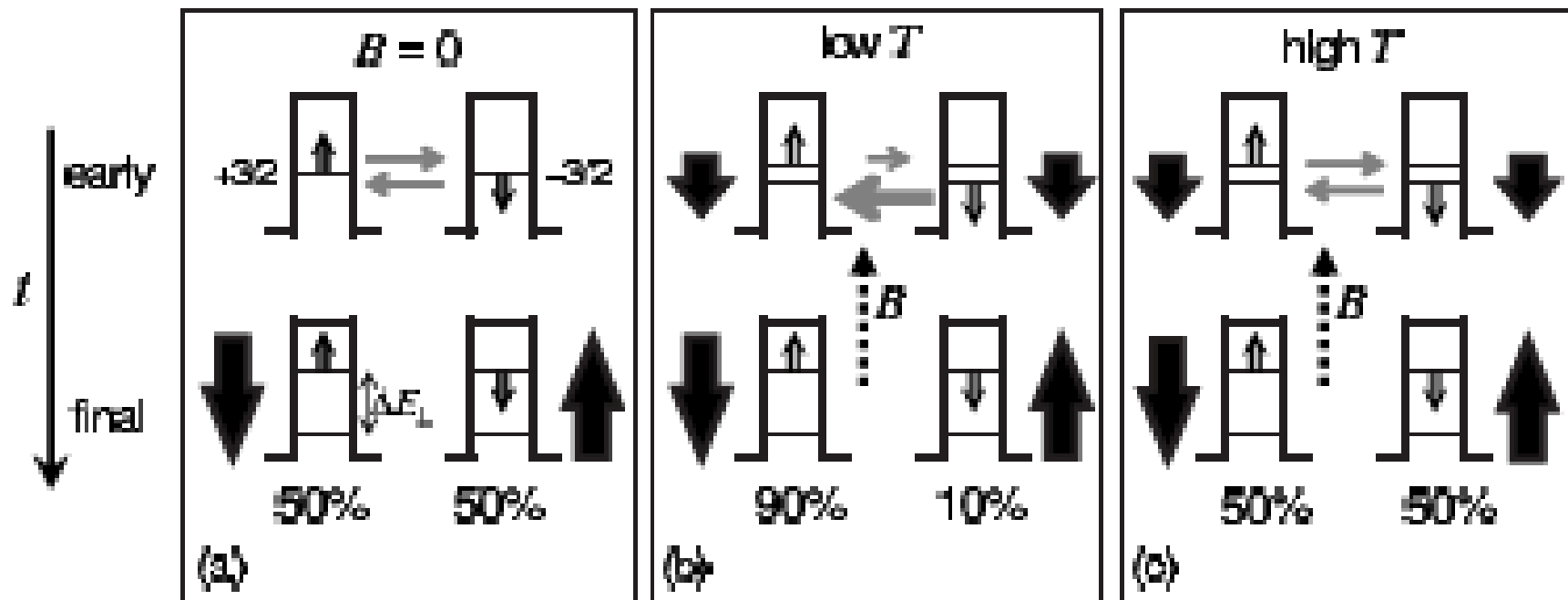
PL spectra as a function of time.

Formation of magnetic polaron (MP).

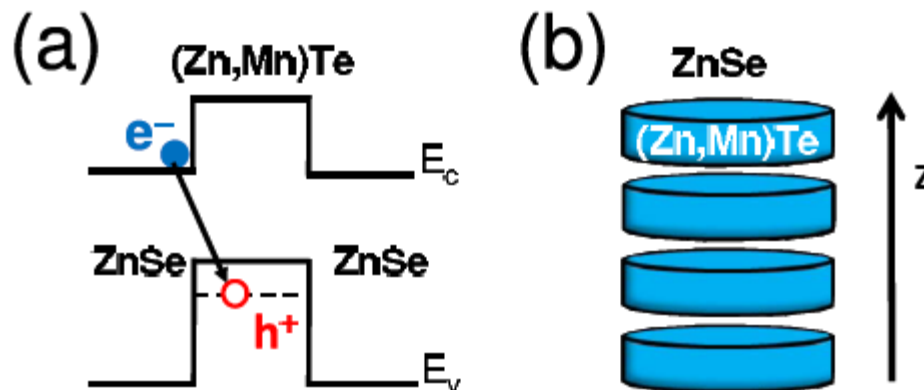


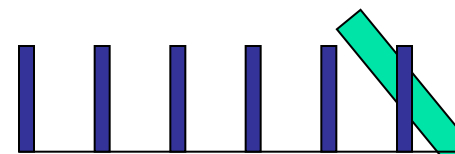
$$E_{MP} = \mu_0^{-1} (J_{ex}/2g\mu_B N_0)^2 \eta(E_{MP}/k_B T) \Omega_{eff}^{-1} \chi(T)$$

- (a) Time dependence of peak PL energy for T=14 to 150K.
 (b) Temperature dependence of the magnetic-polaron formation energy.

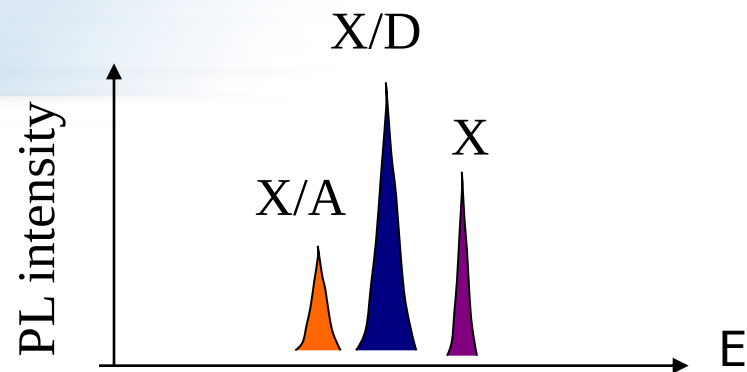
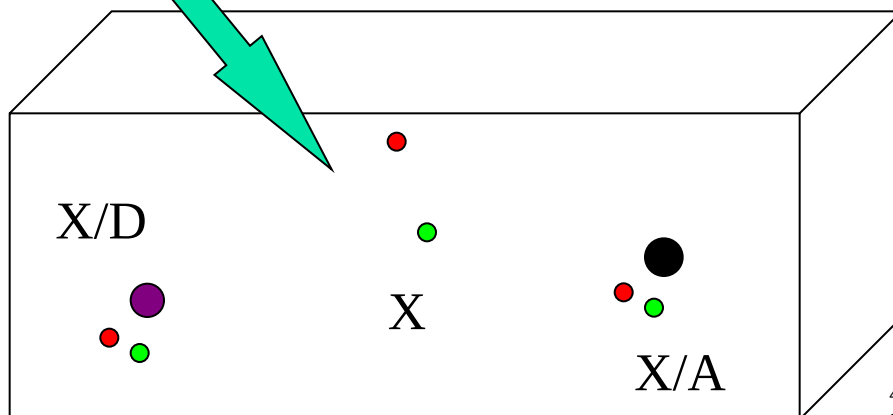


Schematic illustration of the magnetic-polaron formation.



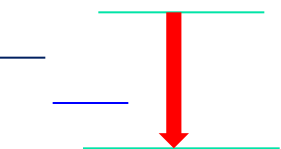


300 ps, 405 nm
2.5 MHz



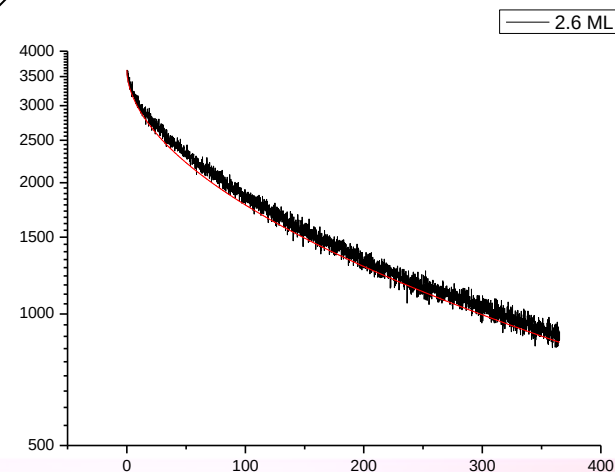
Phonon replica
DA pairs

Decay rate $dn/dt = -n/\tau$
 $n = n_0 \exp(-t/\tau)$
 $\ln(I/I_0) = -t/\tau$ to find lifetime



Unfortunately, it's not linear.

$\ln(I/I_0)$



Decay time



Stretched exponential relaxation (SER) -the problem that is 150 years old-

In 1847, R. Kohlrausch found that the decay of the residual charge on a glass Leyden jar was described by a stretched exponential function $\exp[-(t/\tau)^\beta]$, and he showed that this function could be derived by assuming that the decay rate was not constant, but decreased with time as $t^{\beta-1}$.

Reference:

1847 *Ann. Phys., Lpz.* **12** 393

Decay rate

$$dn/dt = -n/\tau$$

$$n = n_0 \exp(-t/\tau)$$

$$\ln(I/I_0) = -t/\tau$$

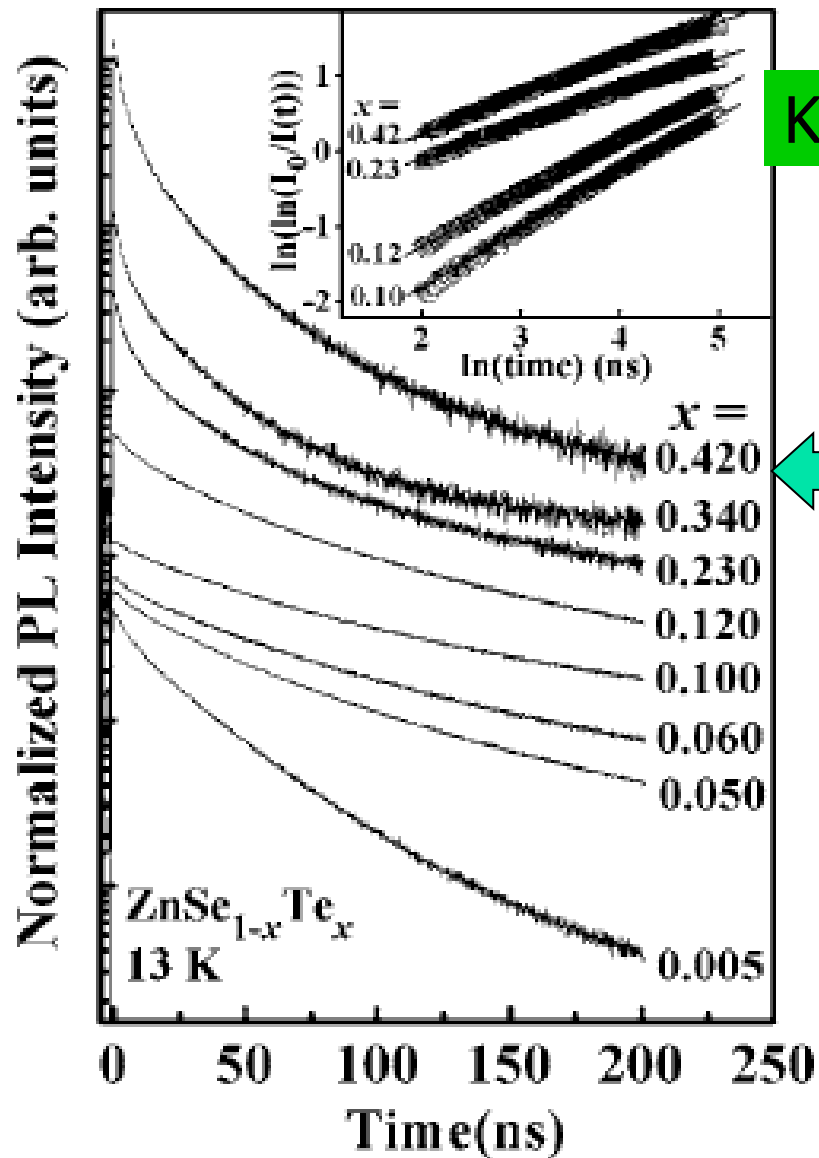
What is the origin SER?



What is the physics behind the Kohlrausch's stretched exponential law ?

Although the Kohlrausch law closely fits donor-acceptor pair luminescence decay, Kuskovsky *et al.* concluded that it has no fundamental significance. (PRL 80, 2412)

However, Phillips, who reviewed the decay dynamics of numerous material systems, summarized a few microscopic models based on the Kohlrausch law and concluded that this law is **nature's best kept secret.** (Rep. Prog. Phys. **59**, 1133)



Kohlrausch's stretched exponential law

Non-mono-exponential

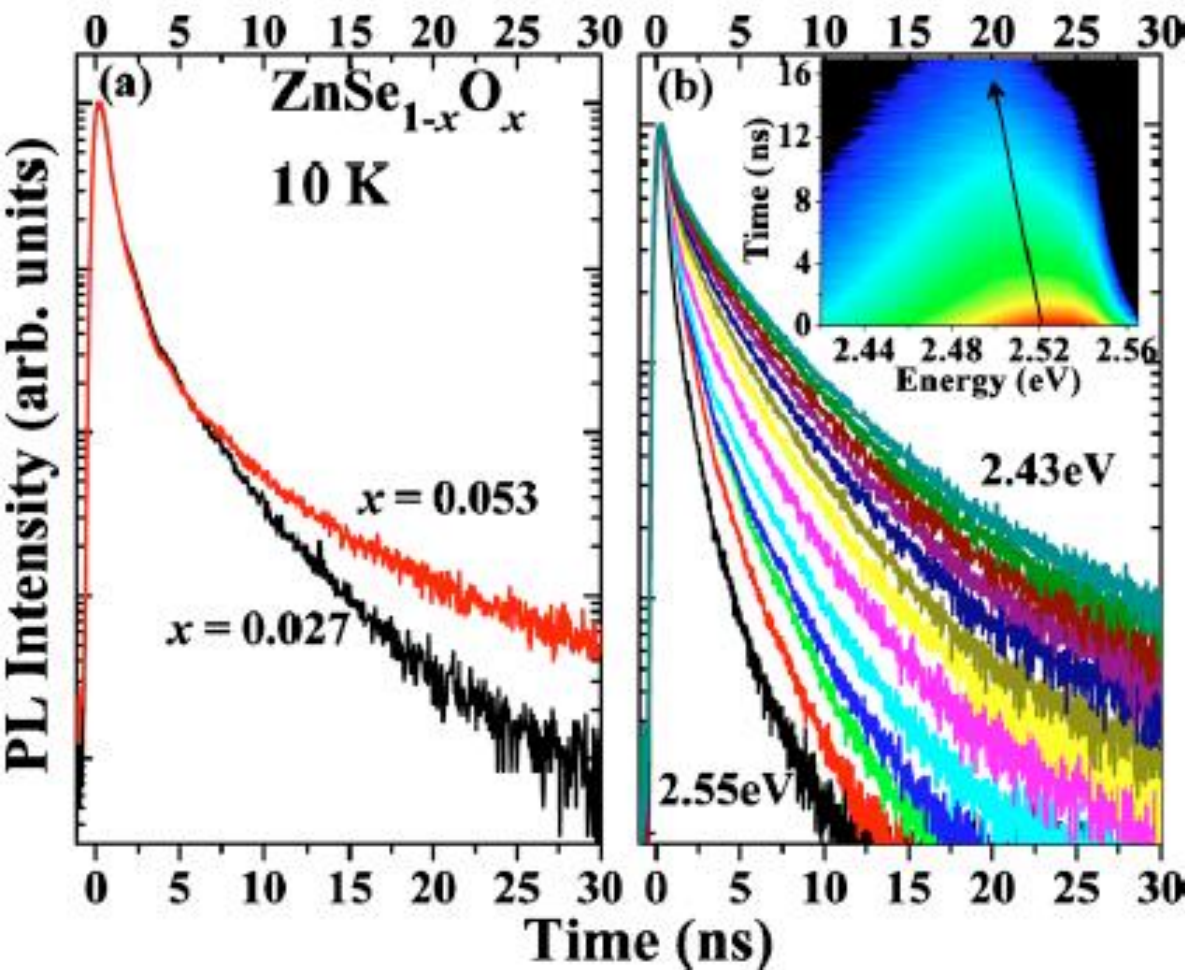
$$I(t) = I_0 \cdot e^{-(t/\tau)^\beta}$$

τ : decay time

β : stretched exponent

What is the origin of non-mono-exponential photoluminescence decay?

Carrier dynamics in isoelectronic $\text{ZnSe}_{1-x}\text{O}_x$ semiconductors



Non-mono-exponential

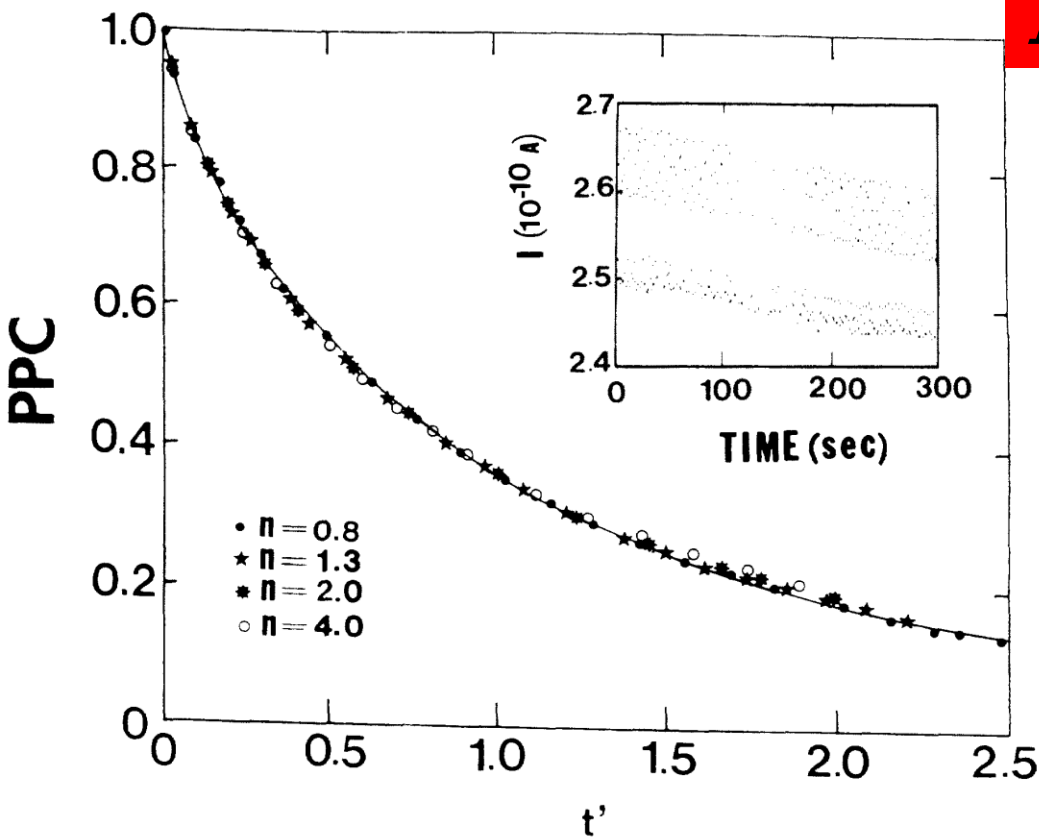
$$I(t) = I_0 \cdot e^{-(t/\tau)^\beta}$$

τ : decay time

β : stretched exponent

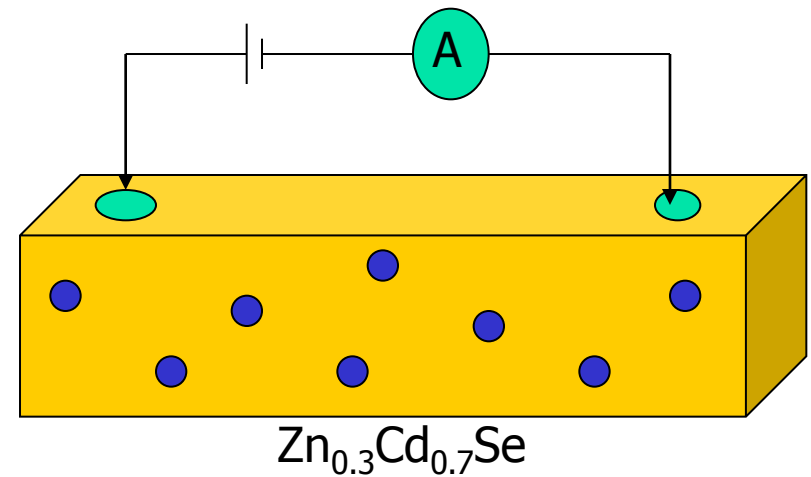
What is the origin of non-mono-exponential photoluminescence decay?

Percolation transition of persistent photoconductivity in II-VI mixed crystals, H.X. Jiang and J.Y. Lin, PRL 64, 2547 (1990)



$$\tau' = t/\tau, \beta = 0.77$$

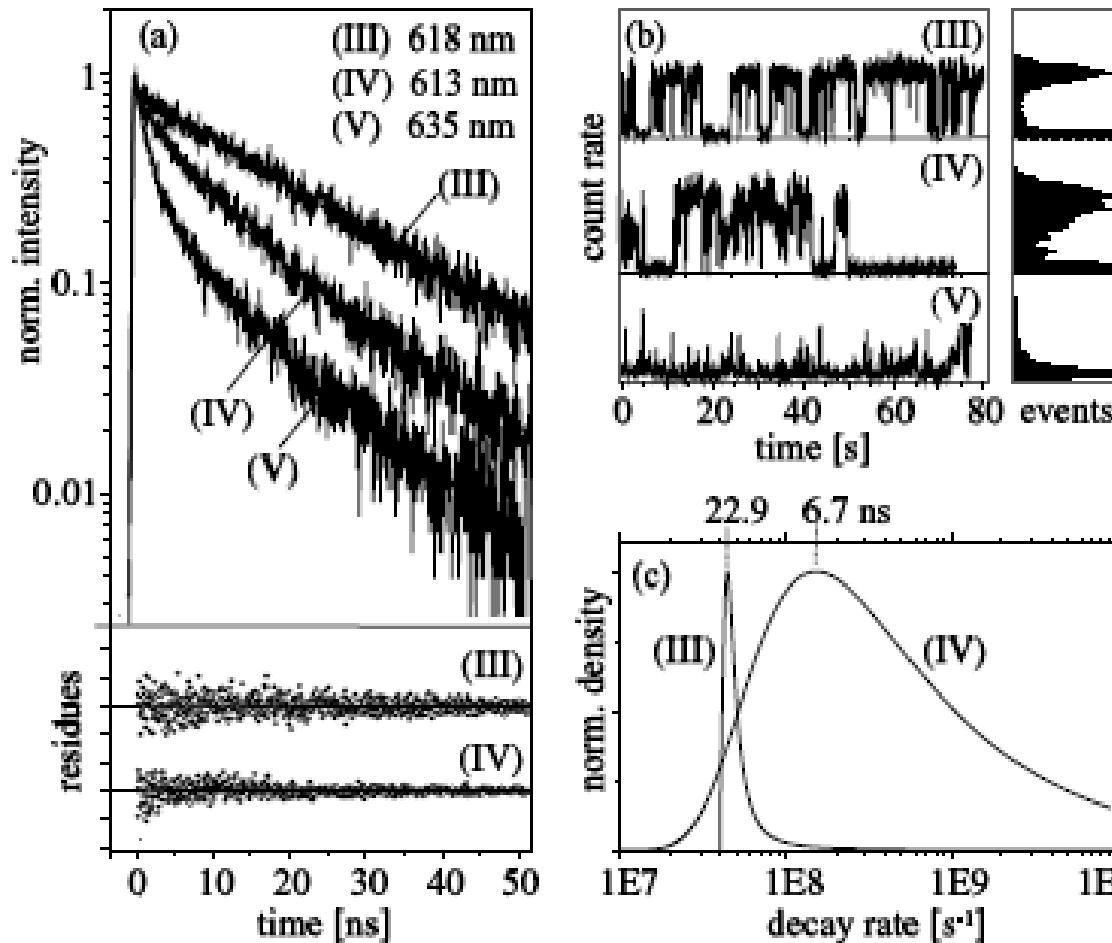
$$I_{ppc}(t) = I_{ppc}(0) \exp [(-t/\tau)^\beta]$$



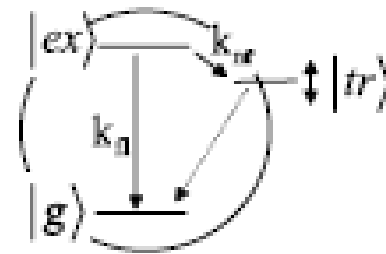
● Random local potential fluctuation

Fluorescence Decay Time of Single Semiconductor Nanocrystals, G. Schlegel et al., PRL 88, 137401 (2002)

CdSe/ZnS core shell quantum dots



the fluorescence decay time is fluctuating during the investigation leading to a multiexponential decay even for a single nanocrystal

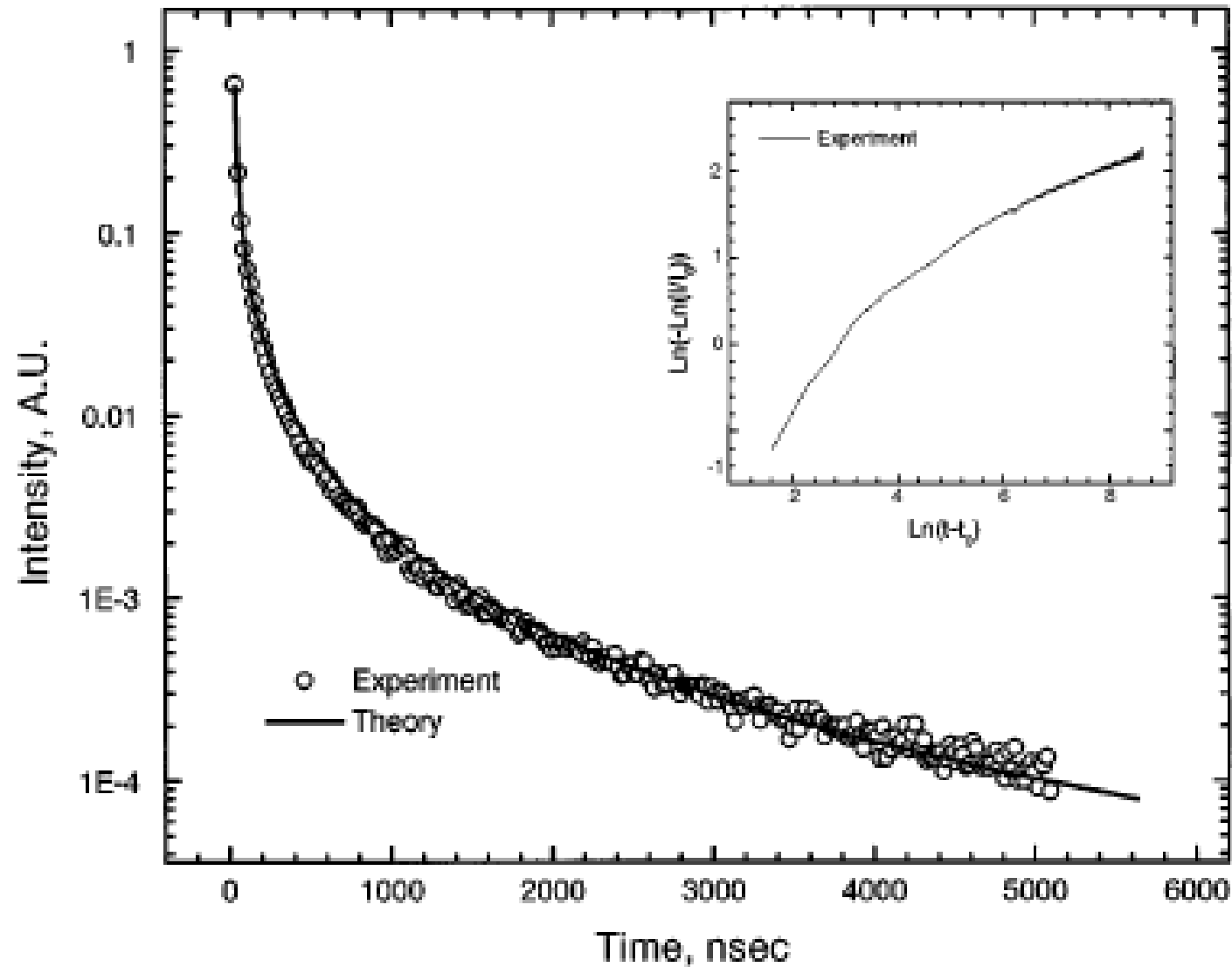


fluctuating nonradiative decay channels leading to variable dynamic quenching processes of the excited state

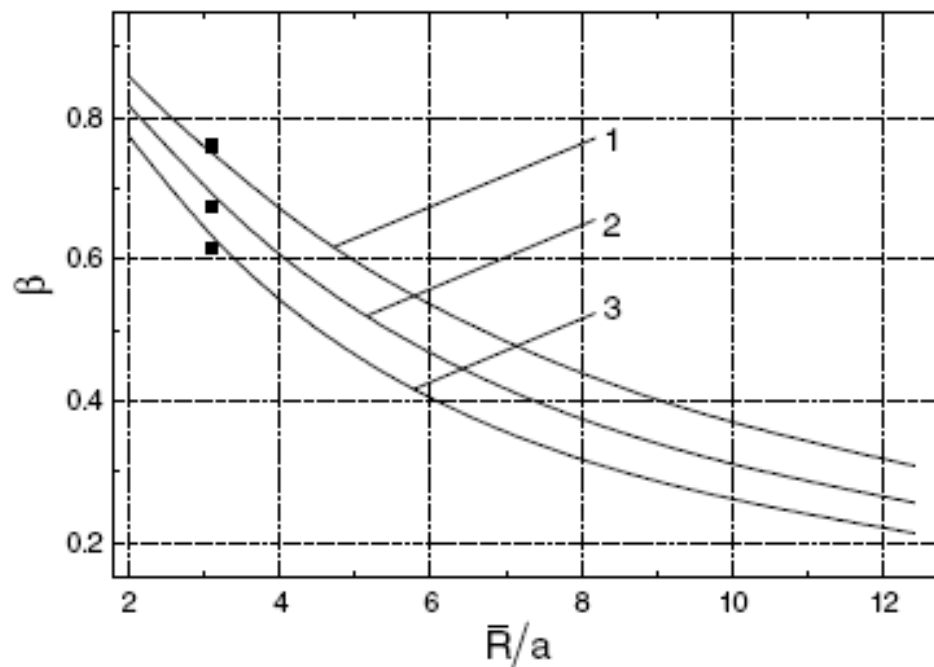
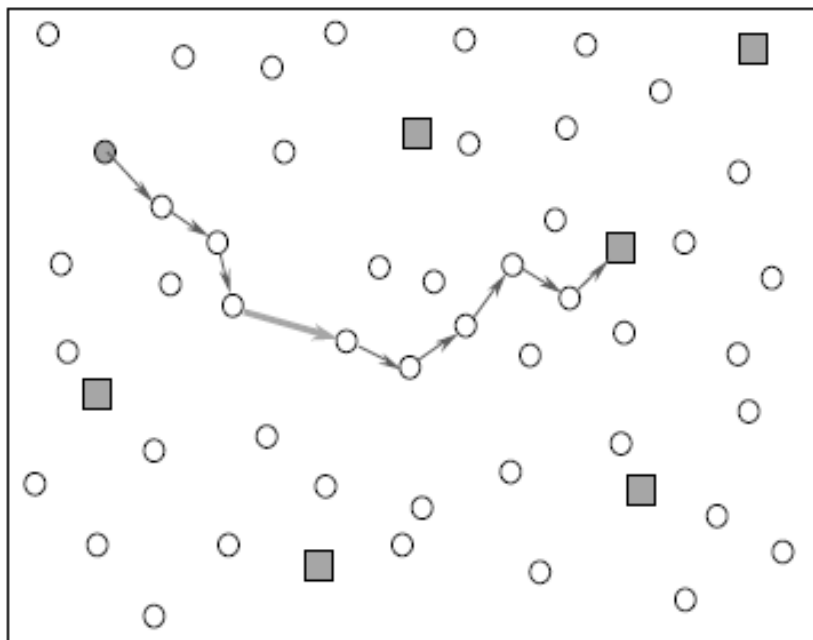
$$\beta_{III}=0.97, \beta_{IV}=0.66$$

$$I(t) = I_0 \cdot e^{-(t/\tau)^\beta}$$

Decay Dynamics in Disordered Systems: Application to Heavily Doped Semiconductors (**ZnSe:N**), PRL 80, 2413, (1998)

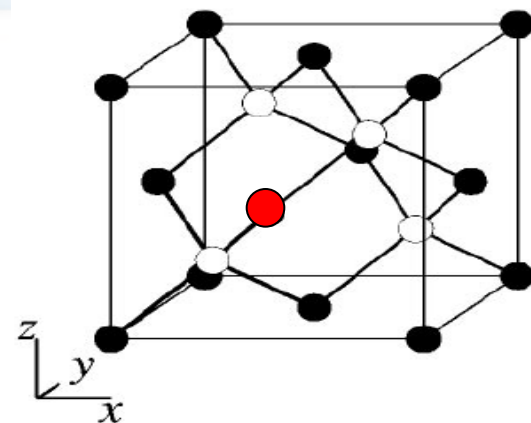
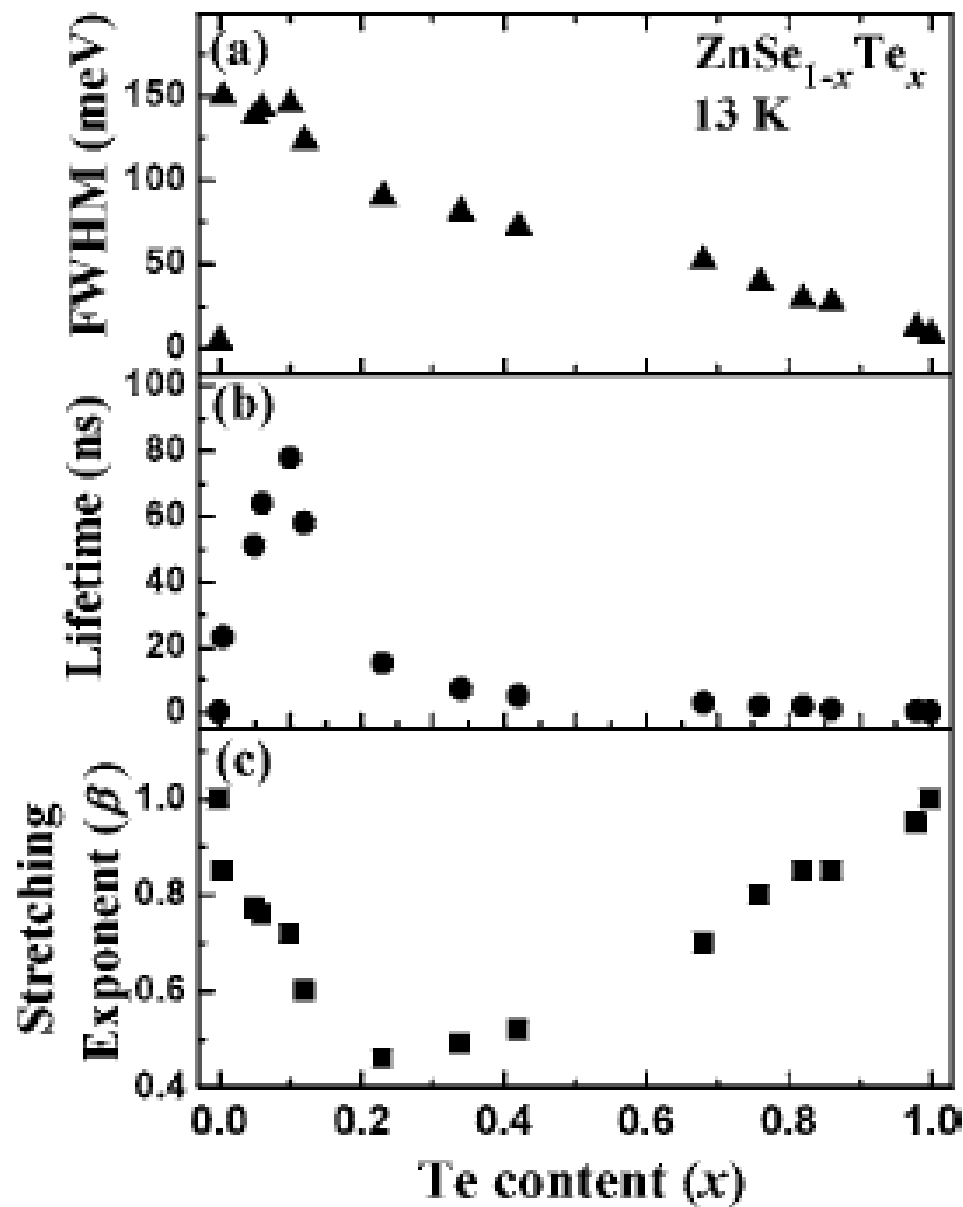


Origin of Stretched Exponential Relaxation for Hopping-Transport Models



$$I(t) = I_0 \cdot e^{-(t/\tau)^\beta}$$

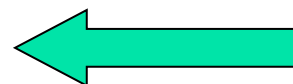
Dependence of the stretching index on $R=a$; the curves 1, 2, and 3 are plotted for $N_0/N_T = 40, 20$, and 10 .



● Te ● Se ○ Zn

Magic number for long-range Coulomb force

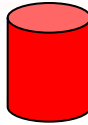
(Rep. Prog. Phys. **59**, 1133)



$\beta=3/7$?



electron

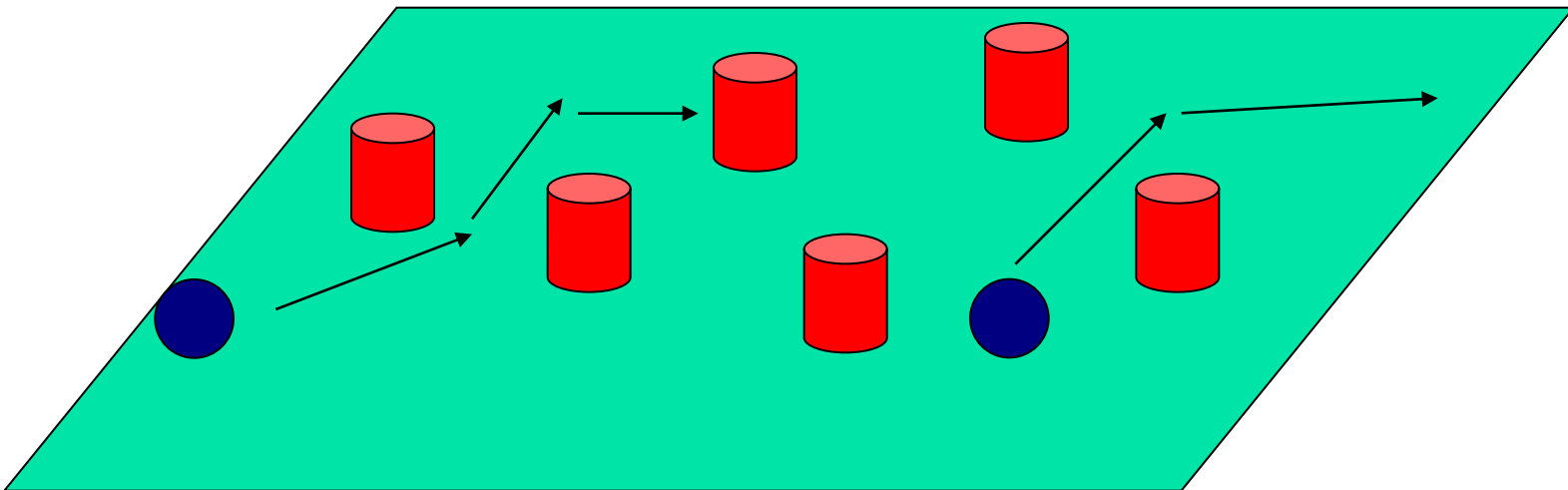


ZnMnTe quantum dot (hole)



ZnSe buffer layer

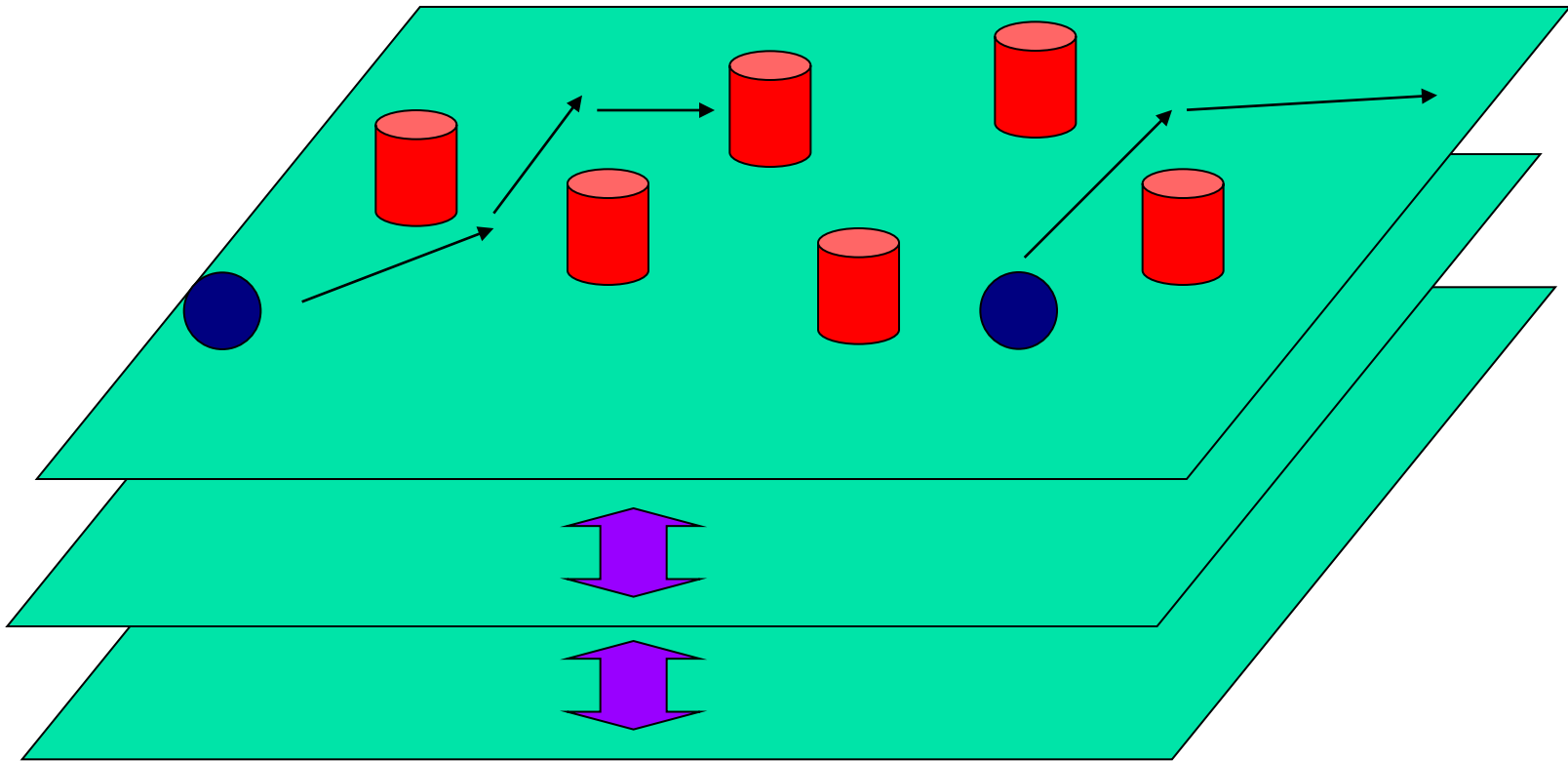
Two dimensional transport.



Does the Kohlrausch's stretched exponential law apply?

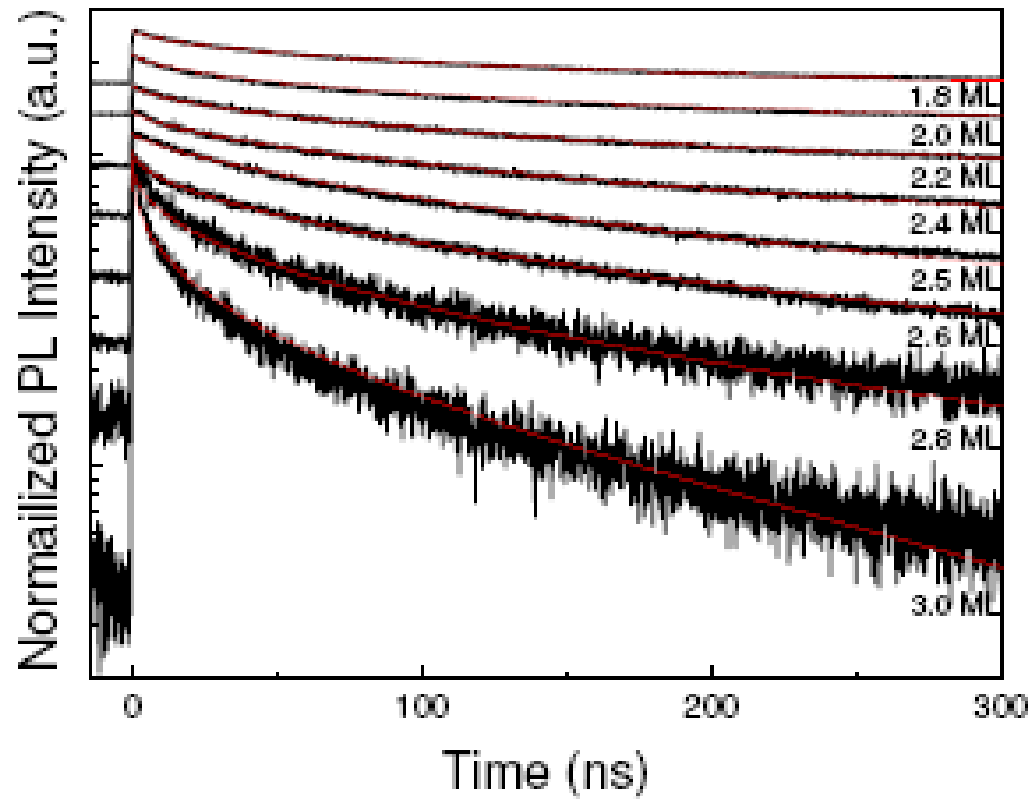
$$I(t) = I_0 \cdot e^{-(t/\tau)^\beta}$$

Multi-layered QDs: Two-three dimensional transport?



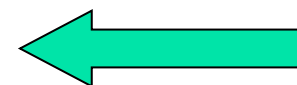
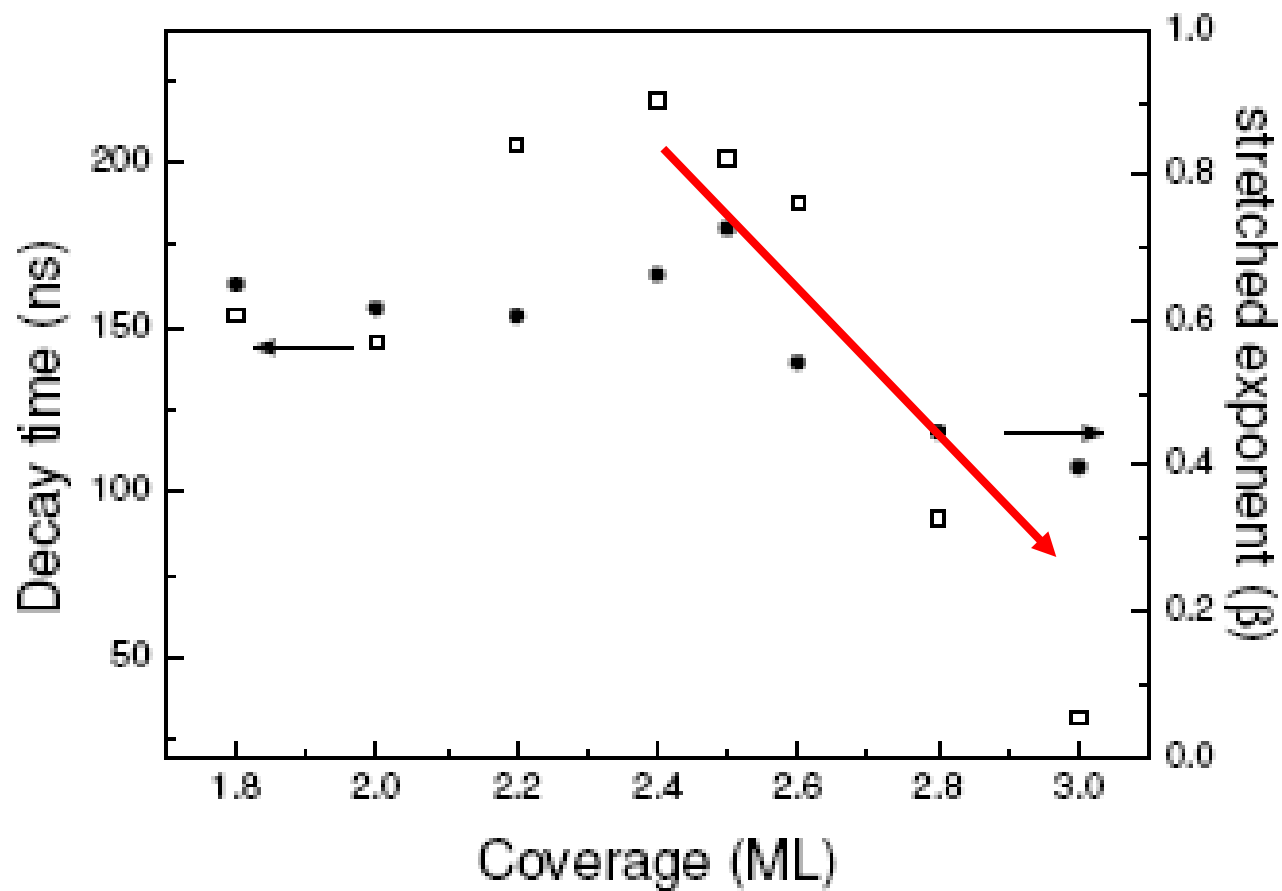
Does the Kohlrausch's stretched exponential law apply?

$$I(t) = I_0 \cdot e^{-(t/\tau)^\beta}$$



Kohlrausch's stretched exponential law

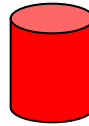
$$I(t) = I_0 \cdot e^{-(t/\tau)^\beta}$$



$\beta = 3/7$?



electron

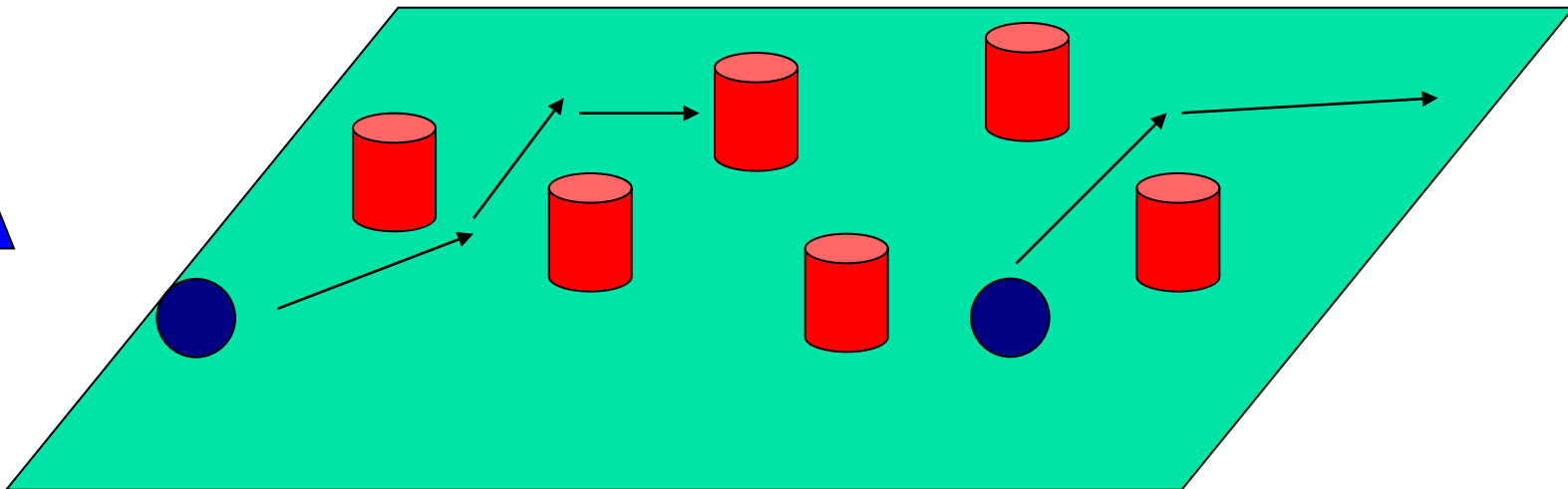
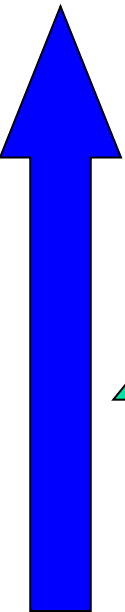


ZnMnTe quantum dot (hole)



ZnSe buffer layer

B



Two dimensional transport.



ZnMnTe/ZnSe MQDs 2.6 MLs

