Teaching Nanomagnets New Tricks

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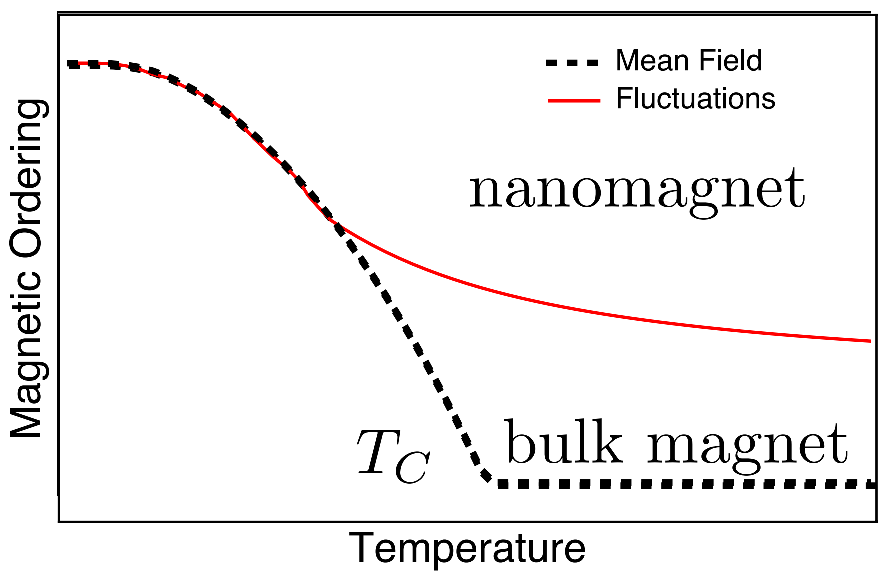
A. Petukhov, South Dakota School Mines & Technology
P. Stano, RIKEN

Mean Field Theory: Friend and Foe in Nanostructures

simple, inexpensive, but often misleading



no phase transitions

Nanomagnetism in Quantum Dots?

Dilute Magnetic Semiconductors: (II,Mn)VI, (III,Mn)V,…
carrier-mediated magnetism, controlling magnetism by changing number of carriers (photo-excitations, gating,…)

T. Dietl and H. Ohno, Rev. Mod. Phys. 86, 187 (2014)

Differences as Compared to Bulk?

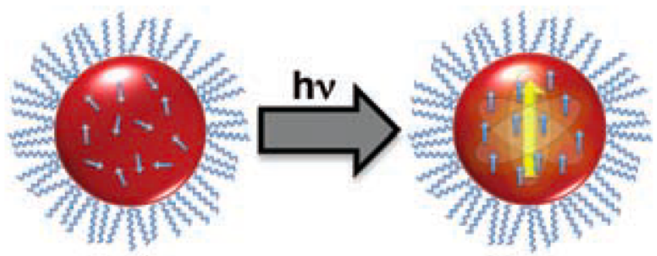
- Controlling Magnetism at Fixed Number of Carriers
- Magnetic Order Enhanced by Heating
- Unexpected Magnetism in Closed-Shell Systems
- Small Systems: Mean-Field Theory?

Magnetism in Quantum Dots

epitaxial or colloidal QDs

Experiments:

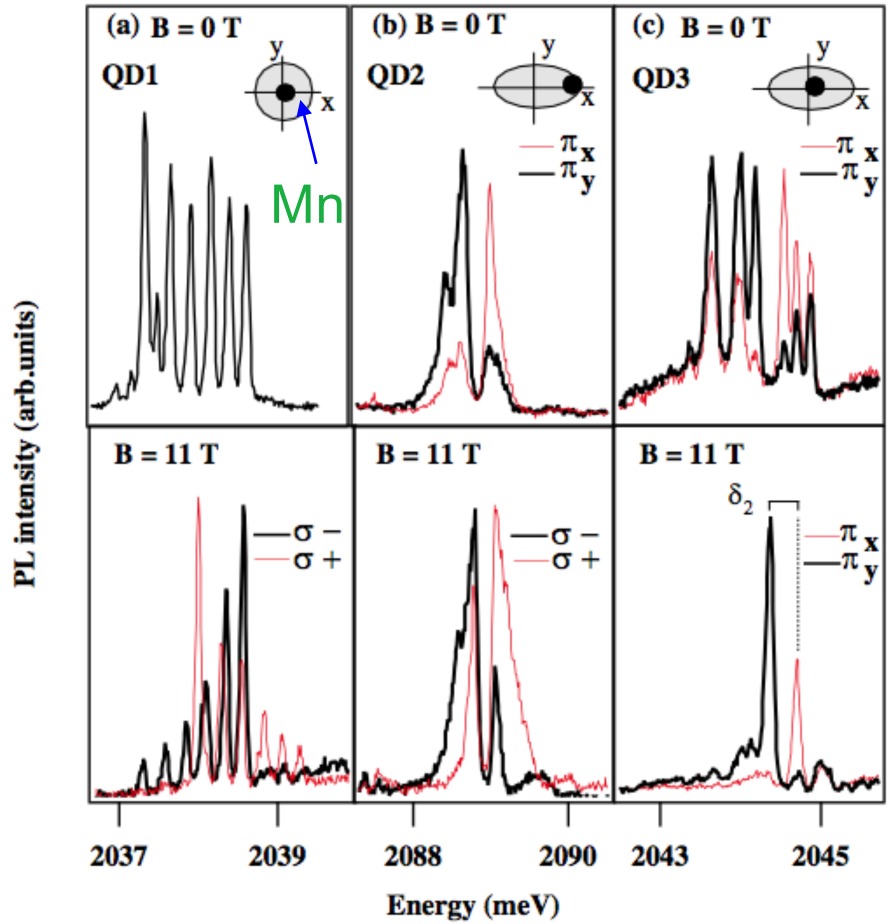
Many or 1 Mn in QDs



Large B_{eff} ~ 100 T

R. Beaulac et al., Science 325, 973 (2009)

Photoluminescence (PL) (Cd,Mn)Te/ZnTe QDs



T=5 K

Early Theories:

J. Fernandez-Rossier, L.Brey, PRL 93 (2004)

A. O. Govorov, PRB 72 (2005)

Al. L. Efros et al., PRL 87 (2001)

fingerprint for Mn location & QD confinement

Y. Leger et al., PRL 95, 047403 (2005) @ Grenoble

Model: *N* electrons, N_m Mn-impurities (spin *S*)

(II,Mn)VI Mn-isolectronic with group-II elements

carriers controlled by chemical doping or gate electrodes

$$H = H_e + H_{Mn} - J_{ex} \underset{i,I}{S} s \cdot S \, d(r_i - R_I)$$

e-Mn interaction $S=5/2$

$$H_e = \underset{i}{S} \left[\frac{-1}{2m^*} \alpha^2 + U_{QD}(r_i) + V_{ee}(r_i) \right]$$

$$U_{QD}(r) = V(x,y) + \frac{1}{2} m^* W^2 z^2$$

"weaker" harmonic in-plane (x,y) confinement

$$V_{ee}(r_i) = \frac{e^2}{e} \underset{j}{S} \frac{1}{|r_i - r_j|}$$

e-e interaction

$$H_{Mn} = \underset{I,I'}{S} J_{I,I'} \, S . S$$

AFM Mn-Mn interaction

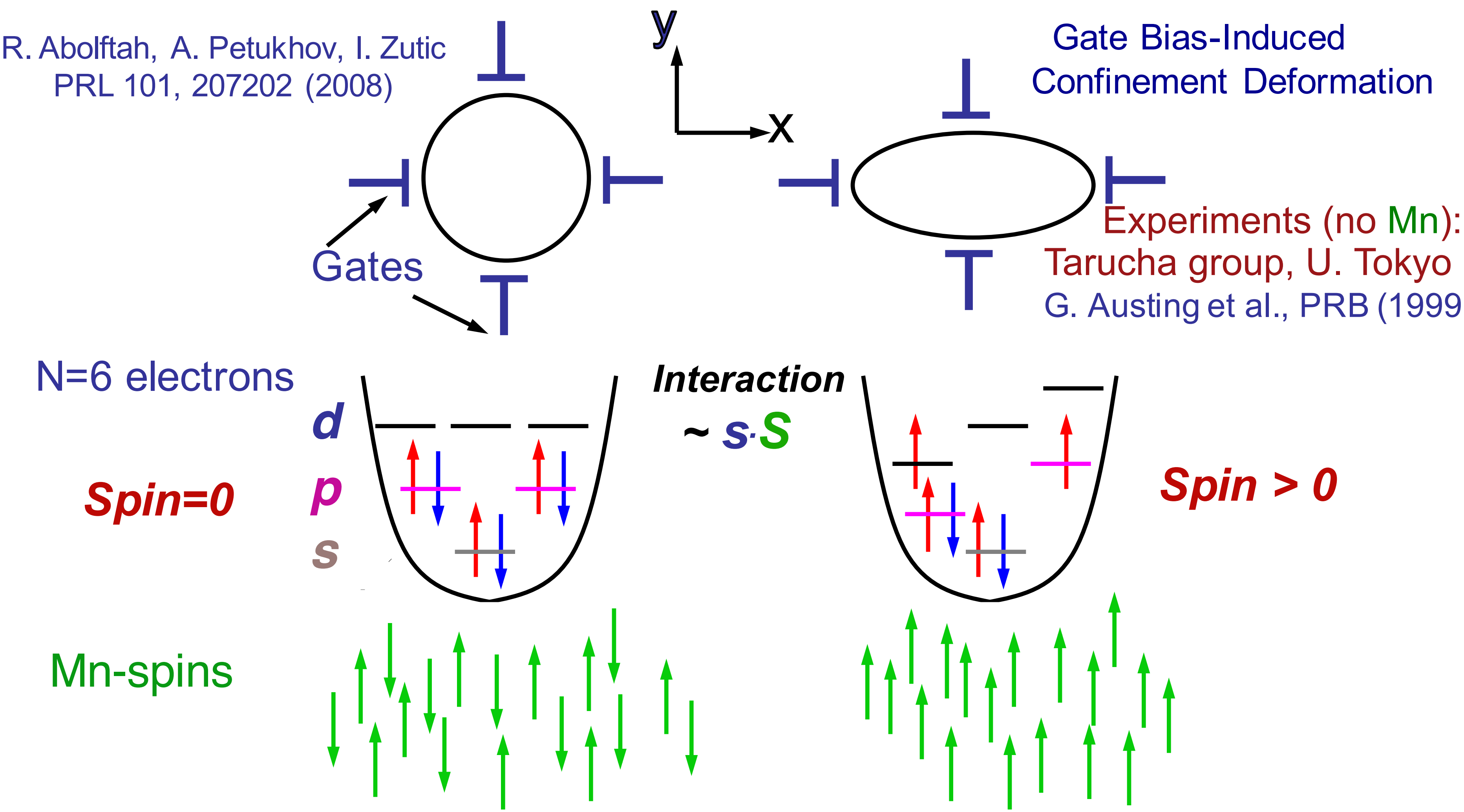
$$N_m = 10 \Rightarrow 6^{10} \approx 6 \times 10^7$$

# of configurations

(Mean Field needed for Mn)

R. Abolftah, A. Petukhov, I. Zutic, PRL 101, 207202 (2008)
R. Abolftah, P. Hawrylak, I. Zutic, PRL 98, 207203 (2007)

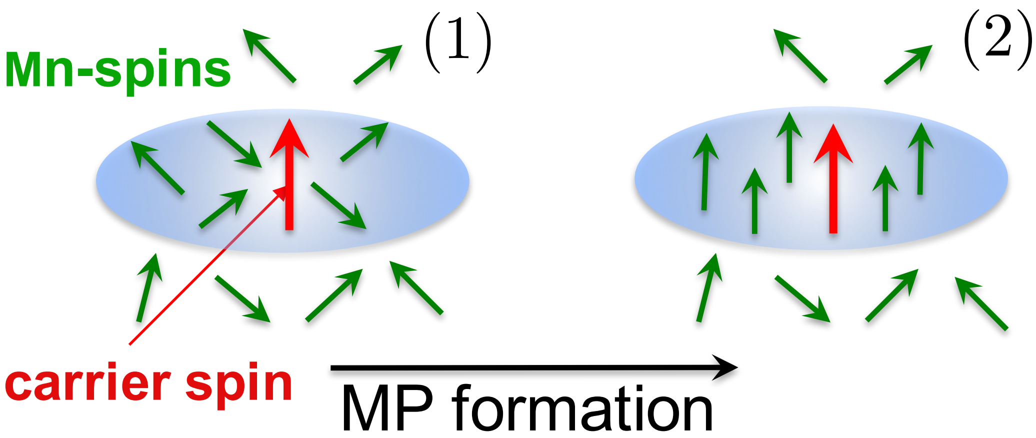
“Piezomagnetism” in QDs?



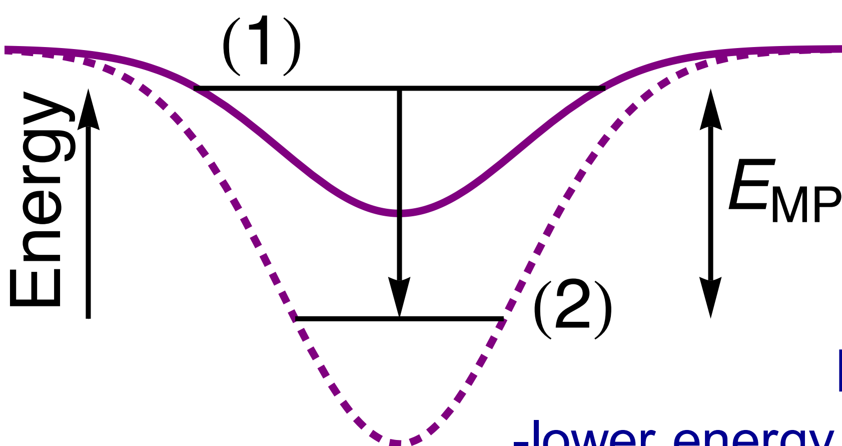
Zero Mn-Magnetization ***Large Mn-Magnetization!***

Controlling Magnetism at **Fixed** Number of Carriers

Magnetic Polarons (MPs)



known for 50+ years in the bulk
but still not fully understood
deGennes, Phys. Rev. 1960



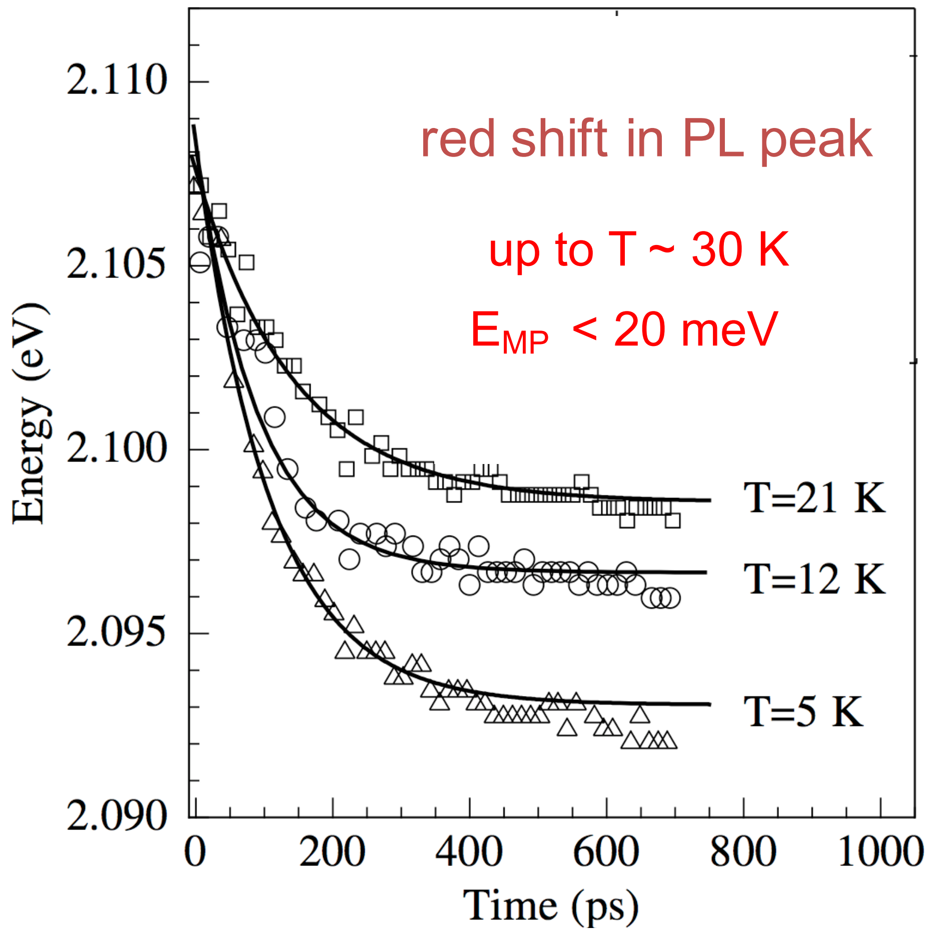
- **binding energy of MP**

MP formation
-lower energy -> Red Shift in the optical data

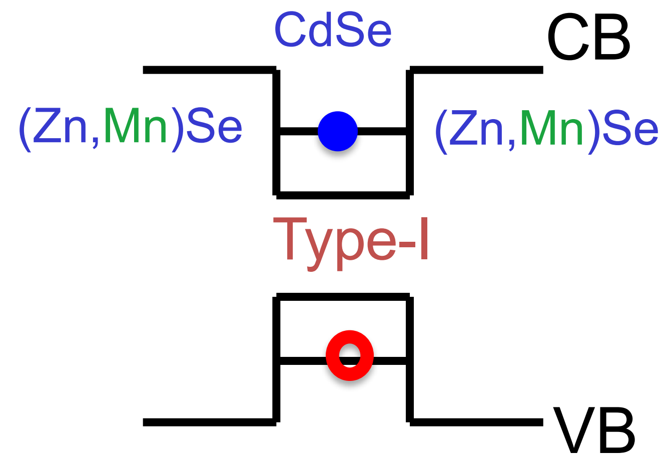
Reduced Dimensionality (Confinement) – Higher MP Stability

Magnetic Polaron Formation (Type-I QDs)

almost all experiments in Type-I QDs



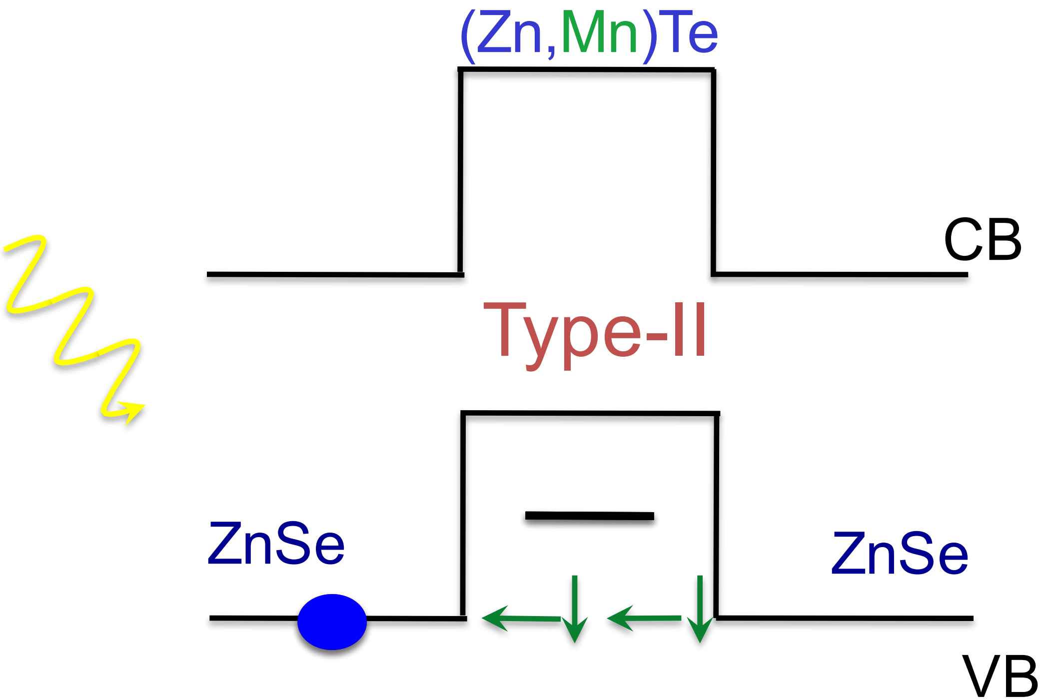
J. Seufert et al., *Phys. Rev. Lett.* **88**, 027402 (2002)



Problems (low temperature effect):

- carriers (excitons) quickly recombine
- small overlap of Mn & carriers

Magnetic Polaron Formation (Type-II QDs)



Ising Exchange Splitting (Holes):

$$\Delta_{\mathrm{Max}} = x_{\mathrm{Mn}} N_0 \beta S$$

$$N_0 \beta = 1.05 \text{ eV}$$

x_{Mn}~ 5-10% Mn, ~100 Mn

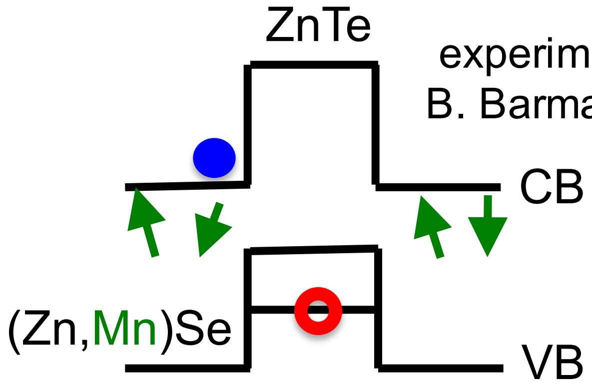
Desirable Properties:

- Large overlap between Mn and holes
- Small overlap between electrons and holes (long recombination times)

experiments at U. Buffalo
samples: W.-C. Chou (NTCU)

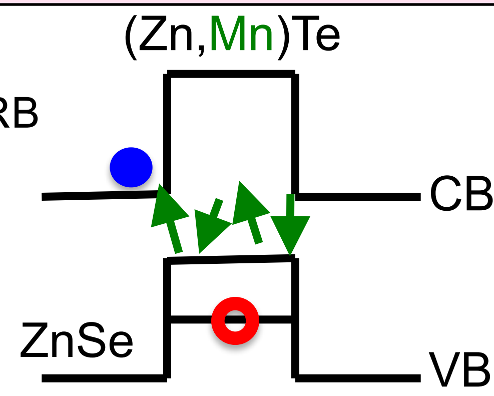
I. R Sellers et al.,
PRB 82, 195320 (2010)

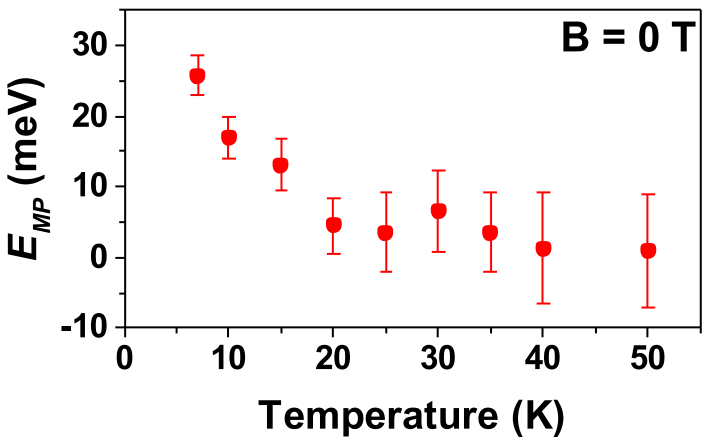
Unconventional Magnetic Polarons ?

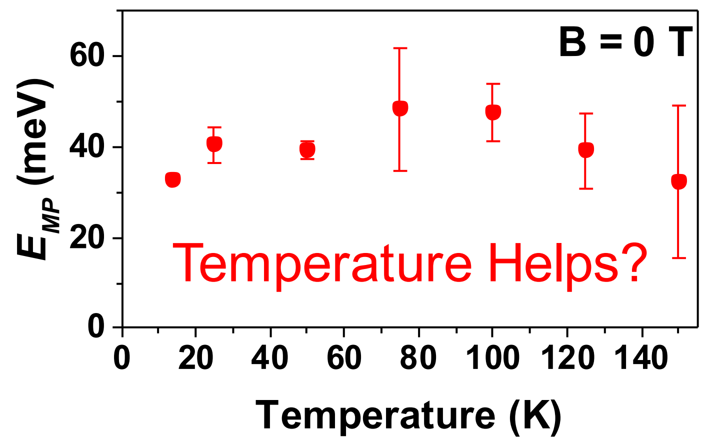


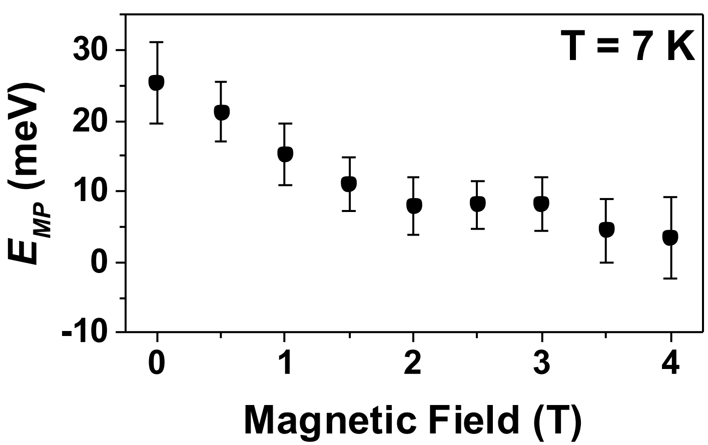
experiments @ Buffalo
B. Barman et al., to appear in PRB

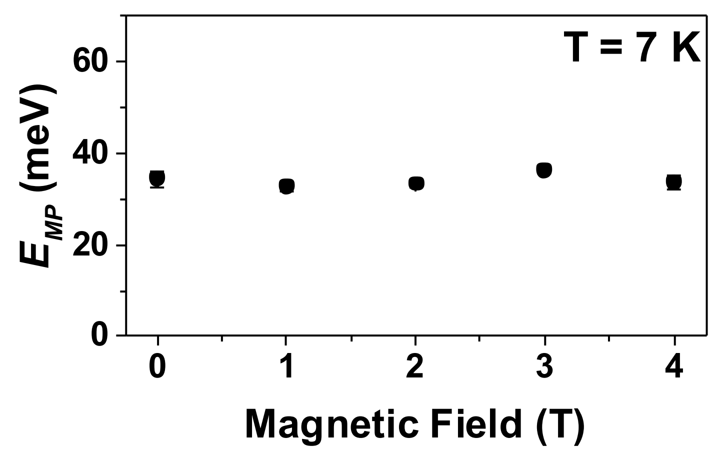
Type-II





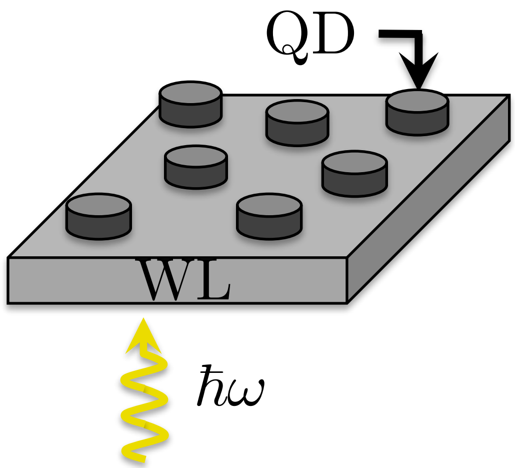


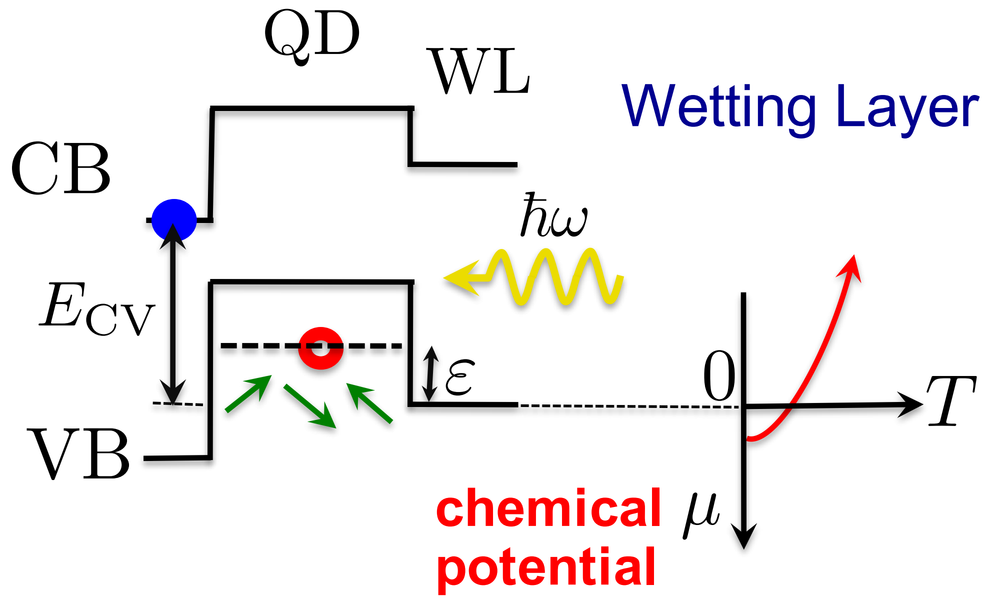




Magnetic Order Enhanced by Heating ?

Model:





Wetting Layer (WL)

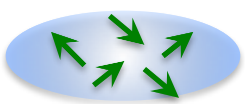
Photo-created holes in the WL which is a particle reservoir with quasi-Fermi Distribution of 2D hole gas

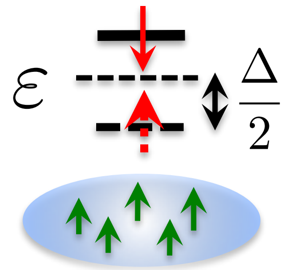
A heavy hole gets trapped in the QD and interacts with Mn-spins (Ising Interaction due to strong spin-orbit coupling)

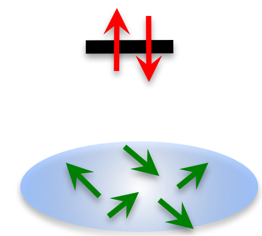
J. Pientka, R. Oszwaldowski, A.G. Petukhov, J. E. Han, and I. Zutic
PRB 86, 161403 (R) (2012)

QD States and Free Energy

Zero Occupancy	***Single Occupancy***	***Double Occupancy***
$E_1 = 0$	$E_{2,3} = \varepsilon \pm \frac{\Delta}{2}$	$E_4 = 2\varepsilon + U$







e, U Confinement, Coulomb Energy

Thermally Changing Occupancy – Alters Mn-Ordering

MP Gibbs Free Energy (Hole, Mn):

$$G_{\rm sys}(\xi) = F_h(\xi) + G_{\rm Mn}(\xi)$$

$$F_h(\xi) = F_0(\xi) + F_1(\xi) + F_2(\xi)$$

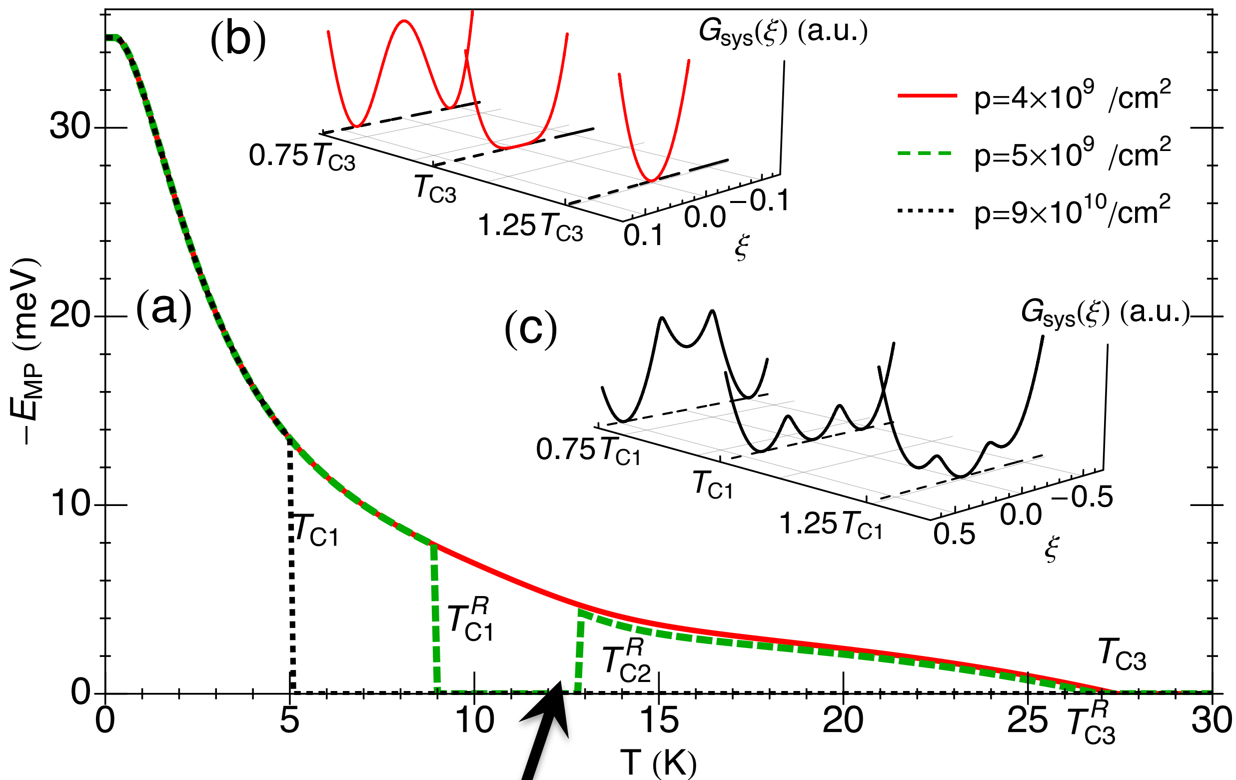
$$\xi = \frac{\Delta}{\Delta_{\rm max}}$$

$$\Delta_{\rm Max} = x_{\rm Mn} N_0 \beta S$$

J. Pientka et al., PRB 86, 161403 (R) (2012)

Mean-Field Description

- Minimize $G_{\rm sys}(\xi)$ to obtain $\xi_{\rm MF}$ (the most probable ξ)
- Calculate the average exchange energy to obtain $E_{\rm MP}$



$p = \text{hole densities}$

Multiple T_C

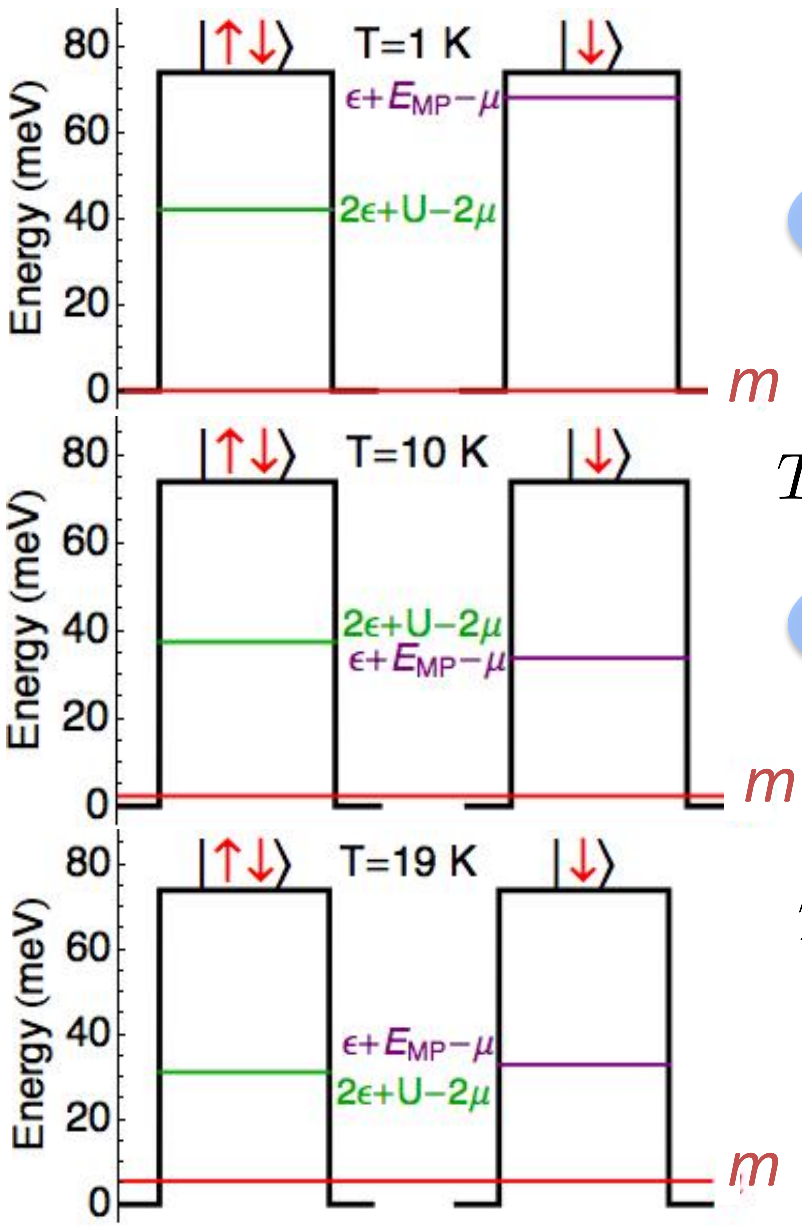
Phase Transitions
(b) 2nd (c) 1st Order

Reentrant MP Formation

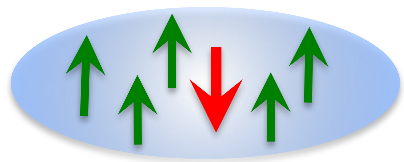
green curve: magnetic order enhanced by temperature

Reentrant MP Formation

Note: Hole Energy increases downwards



$T < T^R_{C1}$

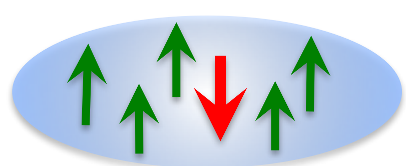


low-*T* single occupancy & Mn-alignment

$T^R_{C1} \leq T < T^R_{C2}$

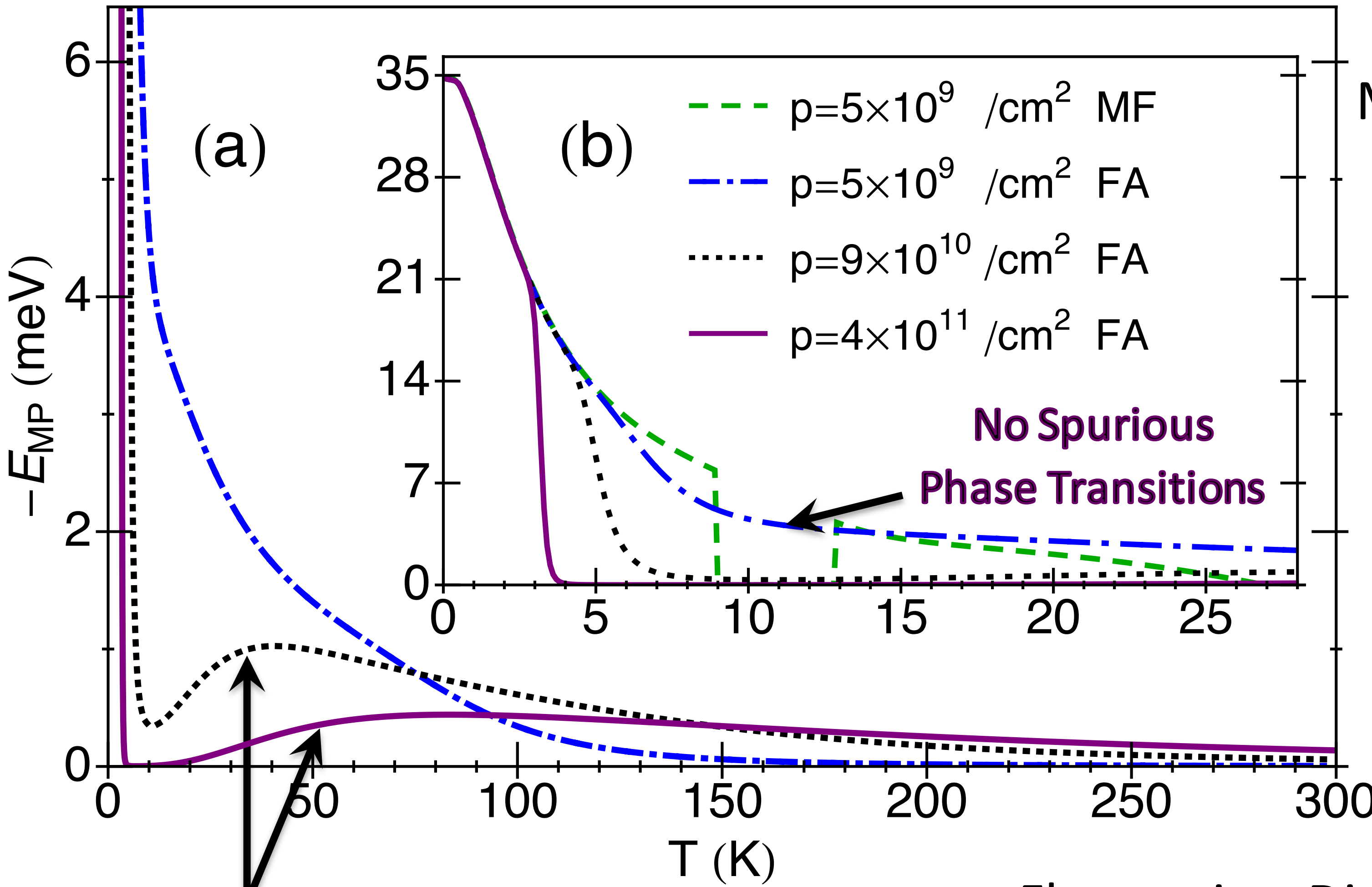
1st Transition due to decrease in E_{MP}

$T^R_{C2} \leq T < T^R_{C3}$

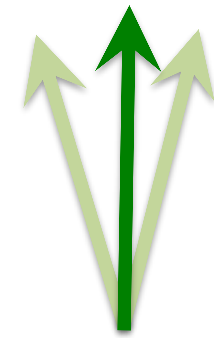


2nd Transition due to decrease in *m*

Fluctuations Approach (FA)



Magnetization Fluctuations



Reentrant MP Formation

beyond Mean-Field

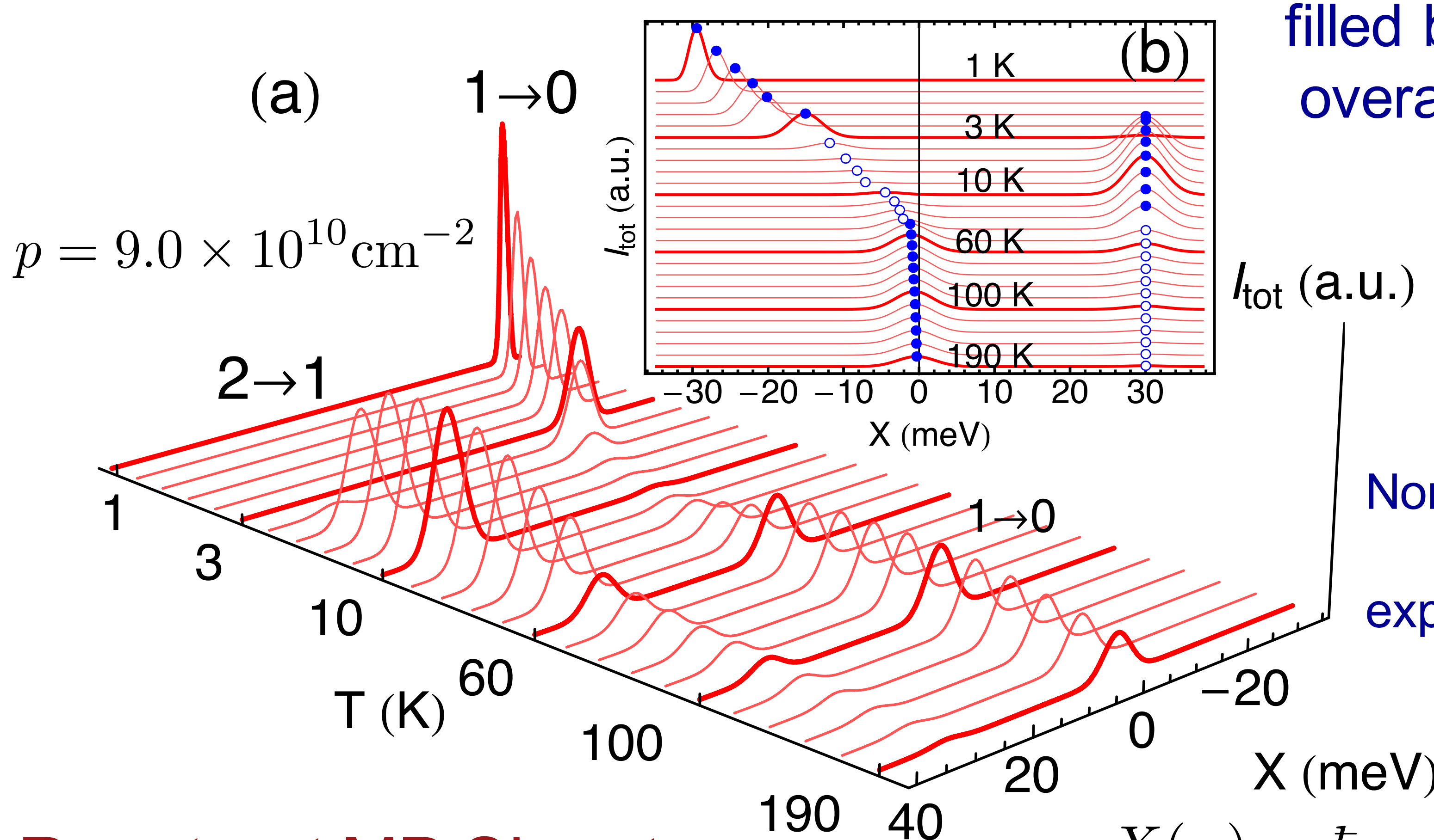
Fluctuation-Dissipation Tm.

$$\chi = \frac{\mu_o}{\Omega}\frac{\langle M^2\rangle - \langle M\rangle^2}{k_B T}$$

J. Pientka et al., PRB 86, 161403 (R) (2012)

Experimental Probe: PL Spectrum

PL Spectrum: $I_{total}(w) = I_{1\to 0}(w) + I_{2\to 1}(w)$



filled blue circles overall PL peak

Nonmonotonic $E_{MP}(T)$ exp. @ U. Buffalo

$$X(\omega) = \hbar\omega - E_{\mathrm{CV}} - \varepsilon$$

Reentrant MP Signature

nomonotonic T-dependence for I_{total} peak position (red & blue shifts) and for I_{total} peak intensity, consistent with nonmonotonic $E_{MP}(T)$

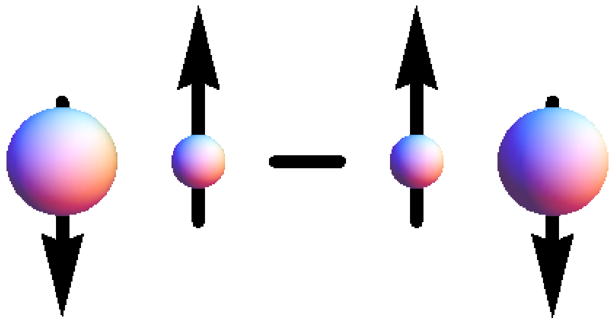
Magnetism in Closed-Shell QDs?

Shell structure in Fermionic systems: atoms, nuclei, QDs,…
Closed-shell systems, like noble gases, known for stability and 0 total spin

Wigner Theorem (Lieb & Mattis, Phys. Rev. '62)
The ground state of any nonmagnetic 2-electron system is a spin singlet

It seems no magnetic ordering in closed-shell QDs doped with Mn

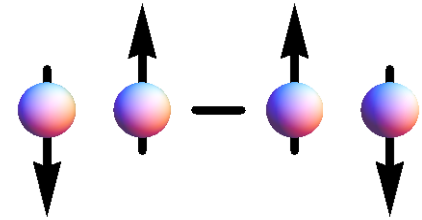
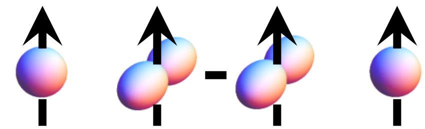
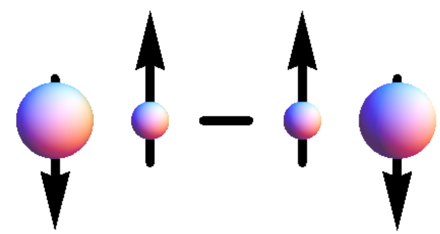
But: Pseudosinglet (0 total spin) can have lower energy!



2 different Bohr radii

R. Oszwaldowski, I. Zutic, A. G. Petukhov, PRL 106, 177201 (2011)

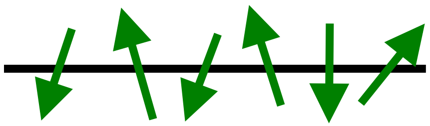
Schematic Description of Two-Fermion States

state	orbital	spin	wavefunction
Singlet (S)	symmetric	antisymmetric	
Triplet (T)	antisymmetric	symmetric	
Pseudosinglet (PS)	not separable		

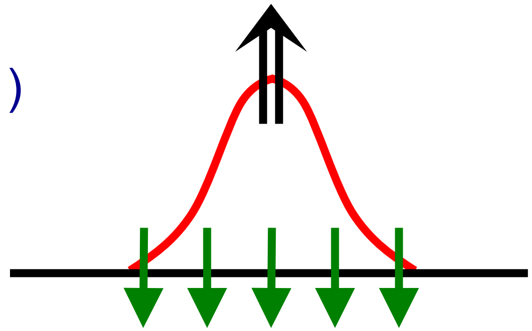
R. Oszwaldowski, I. Zutic, A. G. Petukhov, PRL 106, 177201 (2011)

Proposal for Magnetic Bipolarons

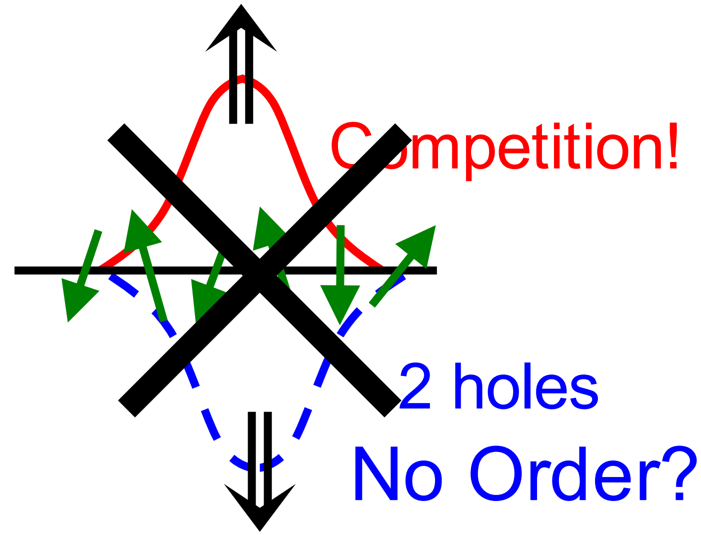
R. Oszwaldowski et al.,
PRL 106, 177201 (2011)



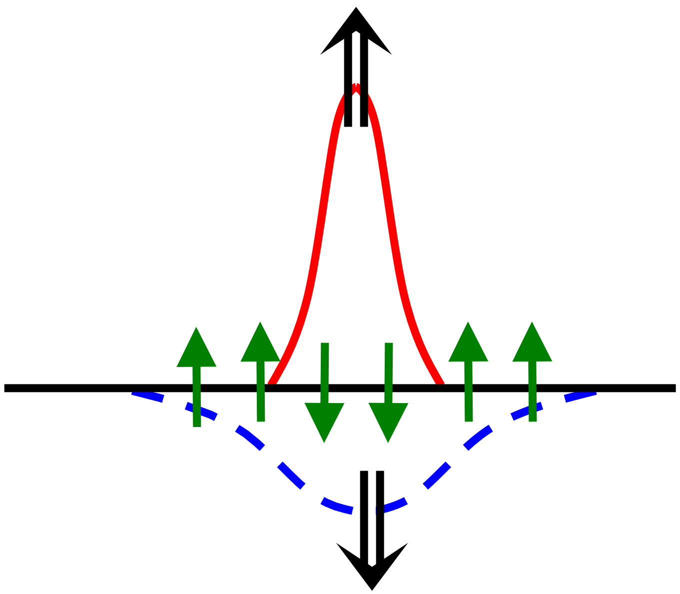
no MP (0 holes)



MP (1 hole)



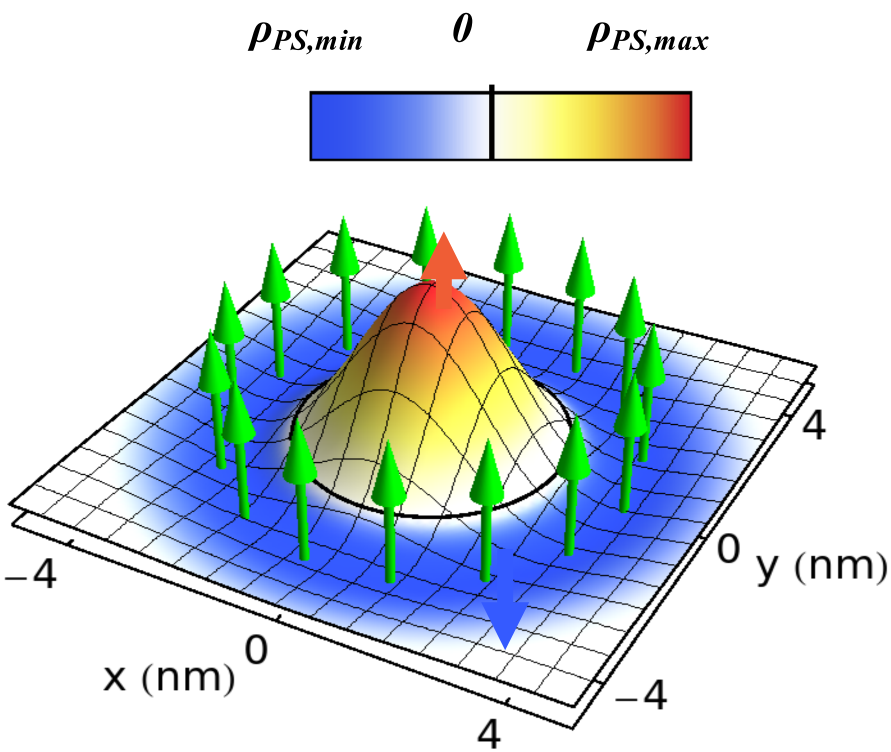
R. Oszwaldowski et al.,
PRB 86, 201408(R) (2012)



"Core-Halo"

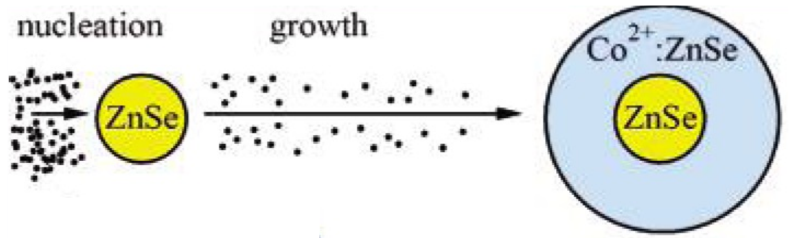
Spontaneous
Symmetry Breaking !
PseudoSinglet

Spin Corral



Experimental realization?

- **self-assembled (Zn,Mn)Se/CdSe**
- **colloidal QDs with radial segregation**



N. S. Norberg et al., JACS 40 13195 (2006)

"Ferromagnetic" alignment in a closed-shell system

- **STM to assemble Mn atoms 1 by 1:**

C. F. Hirjibehedin et al., Science 312, 1021 (2006)
D. Kitchen et al., Nature 442, 436 (2006)

2-carrier QD (analog of He)

R. Oszwaldowski, I. Zutic, A. G. Petukhov, PRL 106, 177201 (2011)

Wigner Crystallization & Wigner Molecules

correlations, "Coulomb/Kinetic Energy" ~ r_s

correlation driven quantum phase transition

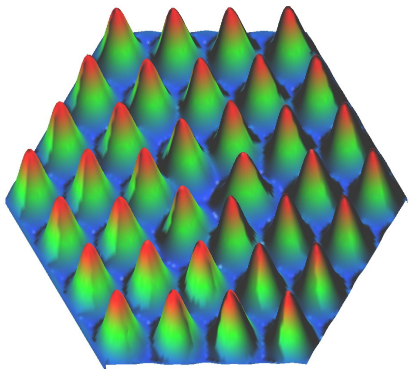
elusive in 3D, density too low (r_s >100)

Wigner Crystal observed in QDs (r_s ~ 1)

C. Ellenberger et al., PRL (2006)

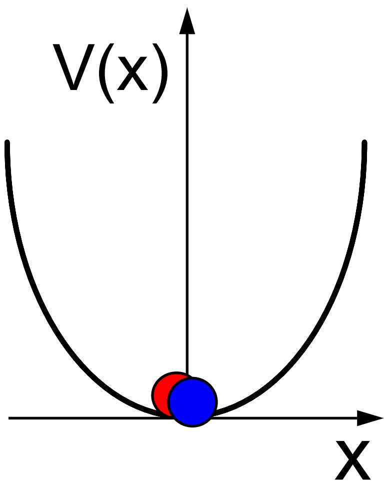
A. Singha et al., PRL 104 (2010)

E. Wigner, PR 46, 1002 (1934)

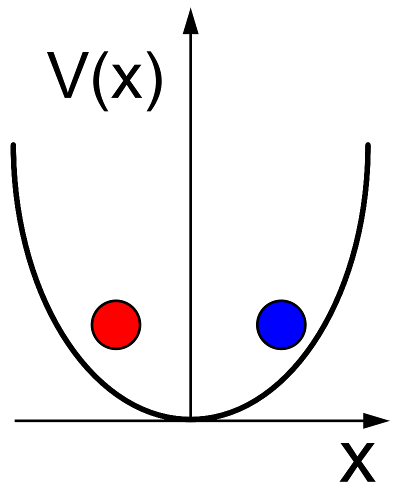


John Trail www.tcm.phy.cam.ac.uk

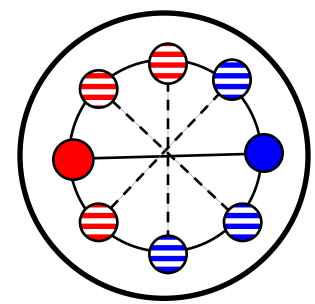
Wigner Molecules: 2 electron QDs, classical limit



no Coulomb repulsion

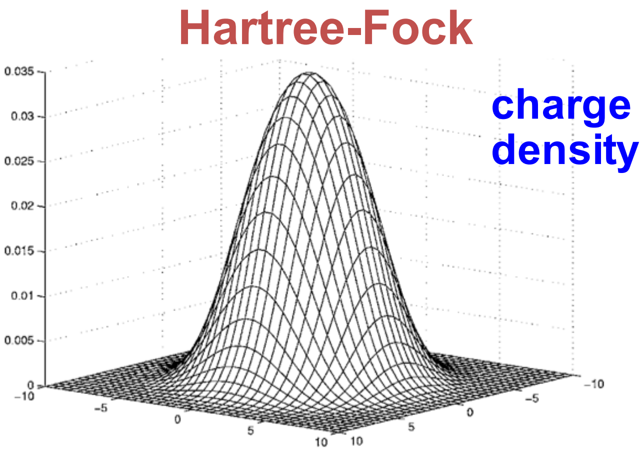


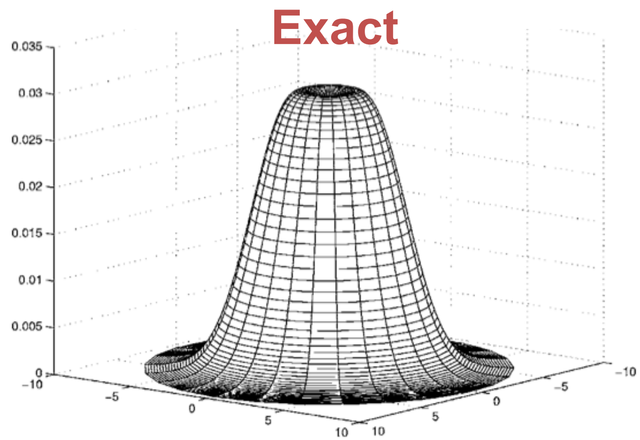
Top View

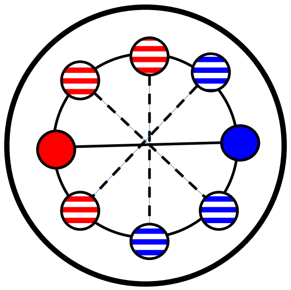


rotating "dimers"
Wigner Molecules

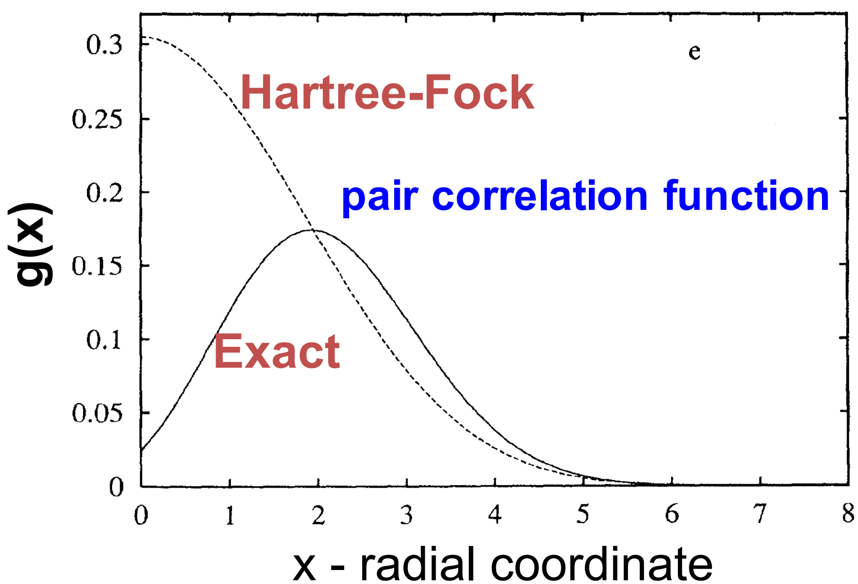
Identifying Wigner Molecules ?



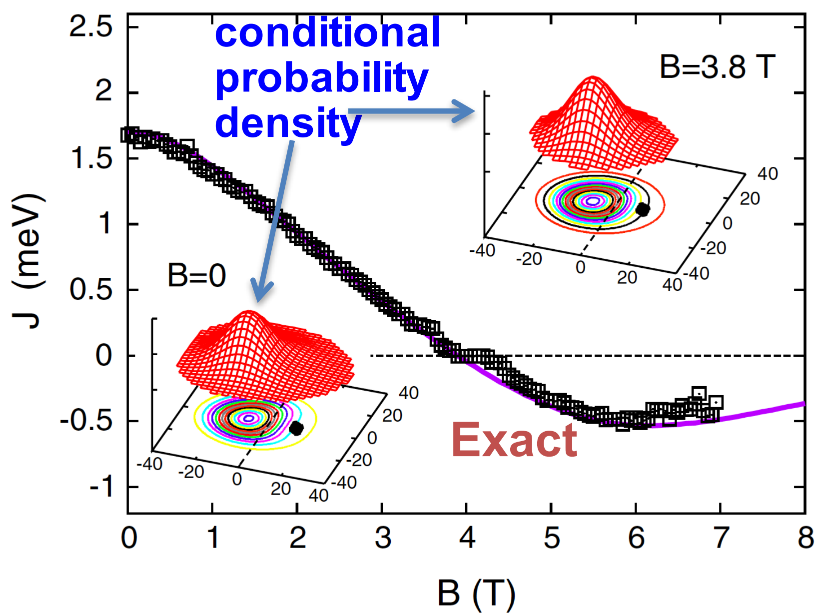




charge density not enough to identify Wigner molecules in nonmagnetic QDs

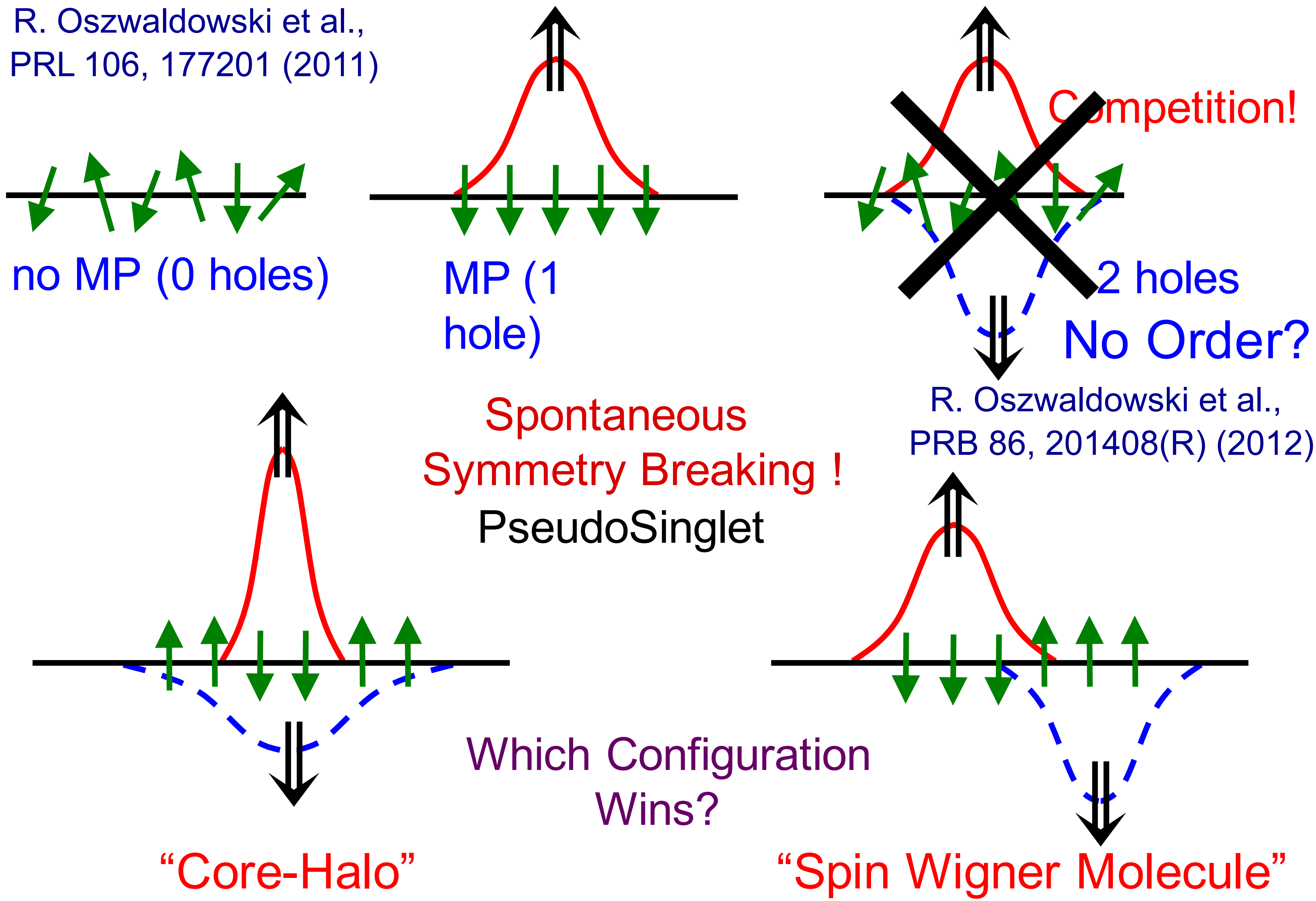


D. Pfannkuche et al., PRB (1993)



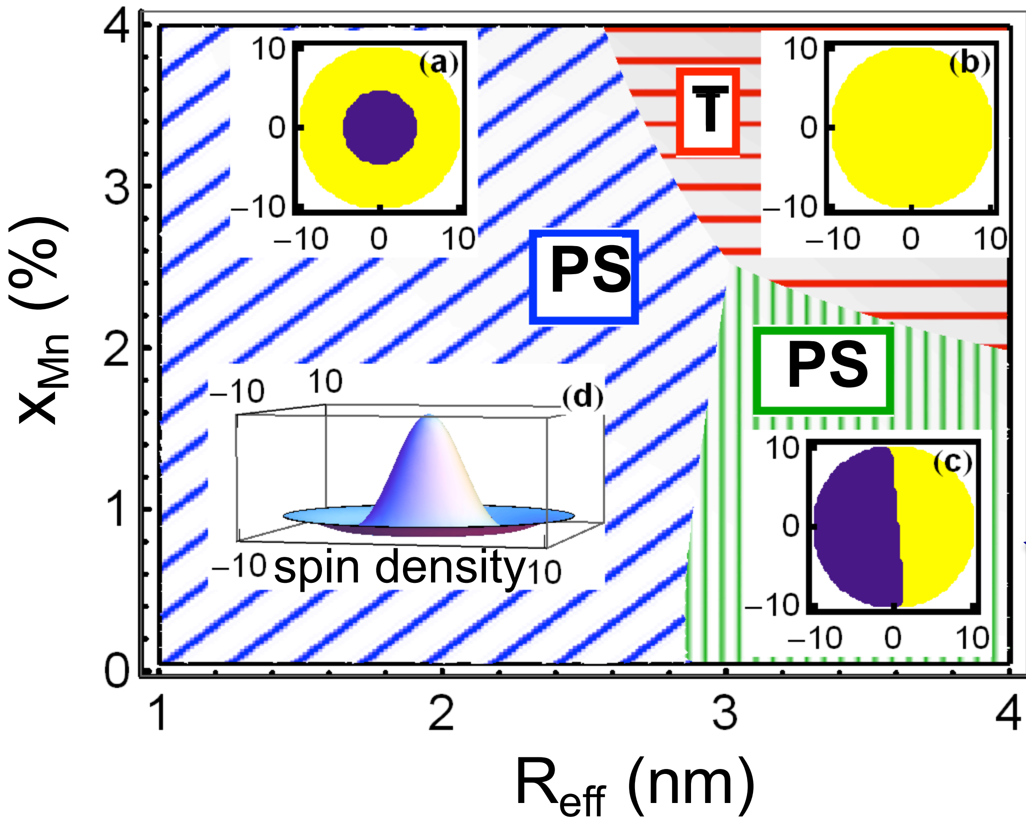
C. Ellenberger et al., PRL (2006)

Magnetic Bipolarons (Revisited)



Magnetic Patterns and Correlation Effects

2 hole Stability Diagram: "exact diagonalization"



Triplet (T) vs Pseudosinglet (PS)

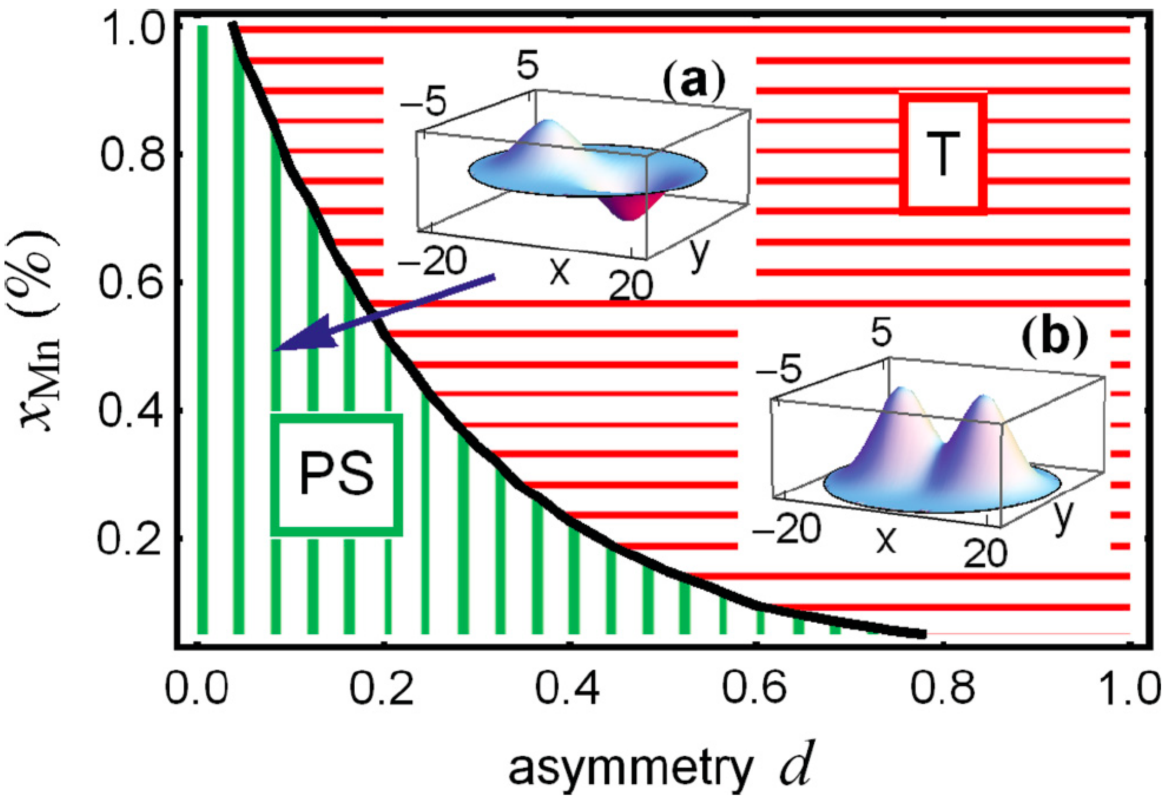
Violet Mn–up
Yellow Mn–down

spin-Wigner molecule
imprinting carrier correlations on **Mn** impurities

constant Mn–doping for $0 < r < R_{eff}$

R. Oszwaldowski, P. Stano, A. Petukhov, I. Zutic, PRB 86, 201408(R) (2012)

Robustness/Detection of Spin Wigner Molecules ?



confinement

$w_{x,y} = w_0/(1 \pm d)^{1/2}$

holes separated along the "softer" potential

Detection: Interband Photoluminescence

two emission lines:

2 to 1 (PseudoSinglet) and 1 to 0 (MP) QD occupancy

Conclusions & Future Directions

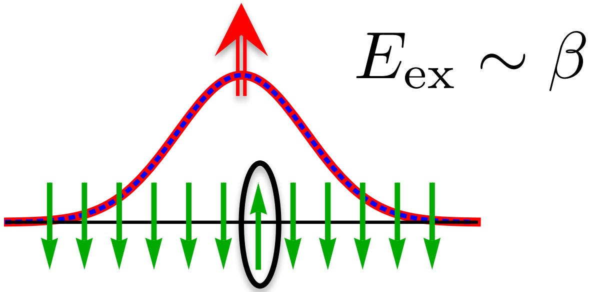
- Heat-Enhanced/Reentrant Magnetism
- Closed-Shell Magnetism
- Probing Correlation Effects With Mn-Impurities

Detecting Fluctuating Nanomagnetic Patterns ?

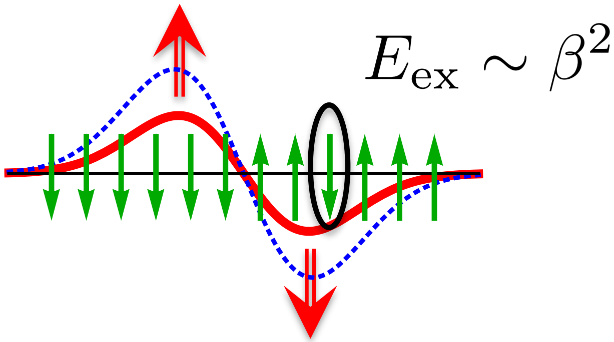
Spin-Polarized STM? Talk R. Wiesendanger   NV Centers? Talk N. Mizuochi
L. Molenkamp

- Beyond mean field: Fluctuation Approach, Monte Carlo,…

Non-interacting vs Interacting Problem



polaron: rigid hole-Mn coupling



bipolaron: hole in a polarized medium that mediates magnetic interaction