# Theoretical investigation of magnetic exchange interactions in dilute magnetic semiconductor quantum dots induced by defects

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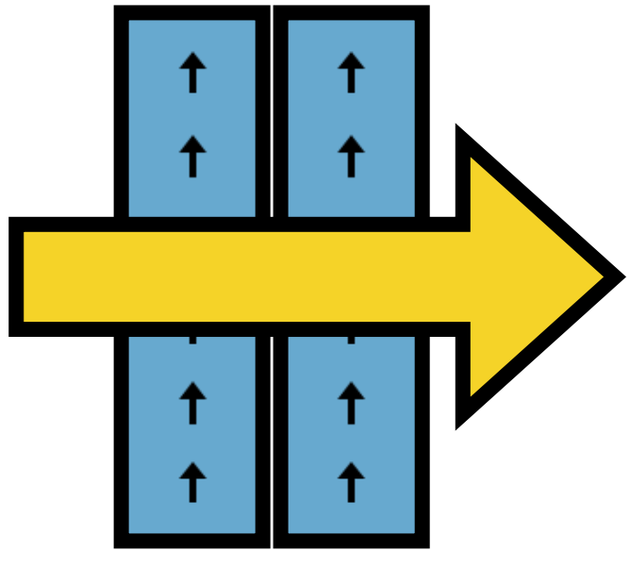
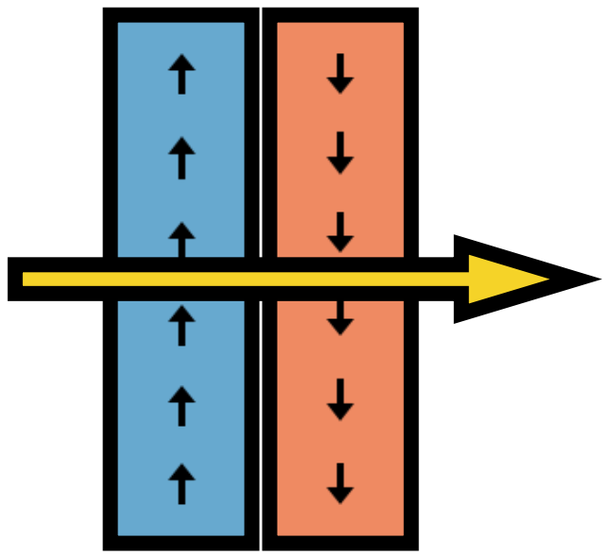
Tuesday, December 15, 11:20am

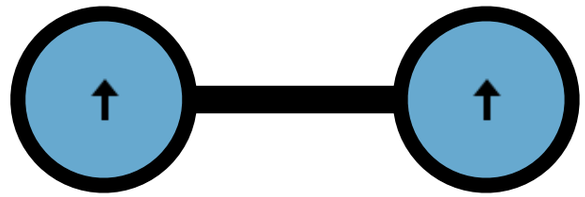
# Controlling magnetism at the nanoscale is important.

low resistance

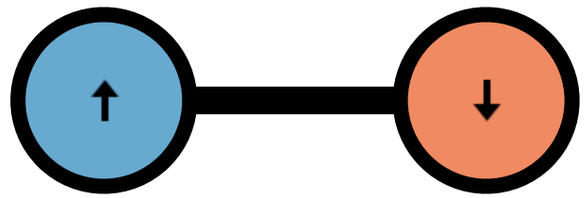


high resistance



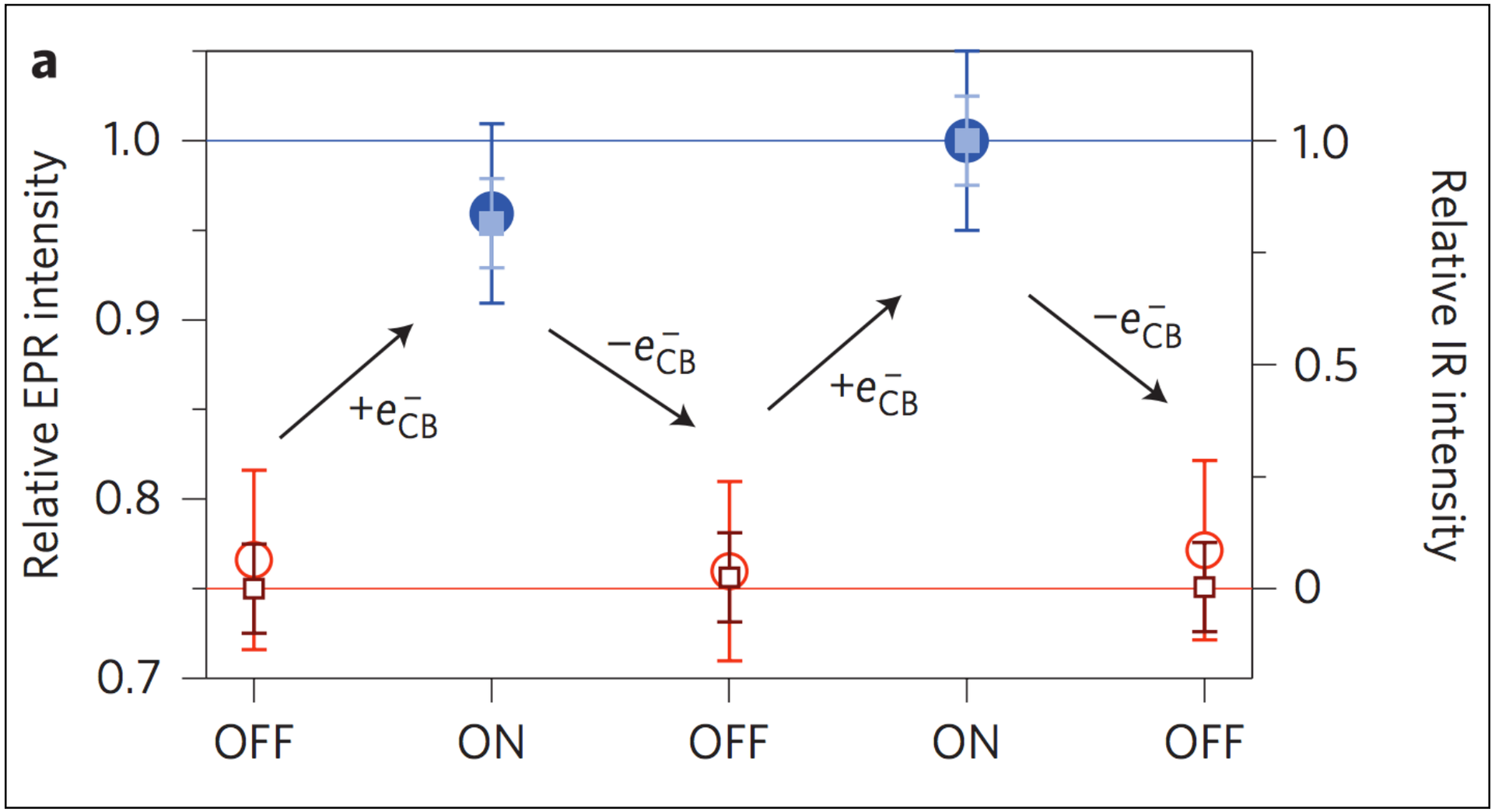


ultimate miniaturization:
ferromagnetic quantum dots linked
by semiconducting polymer



Tunnel Magnetoresistance
(TMR)

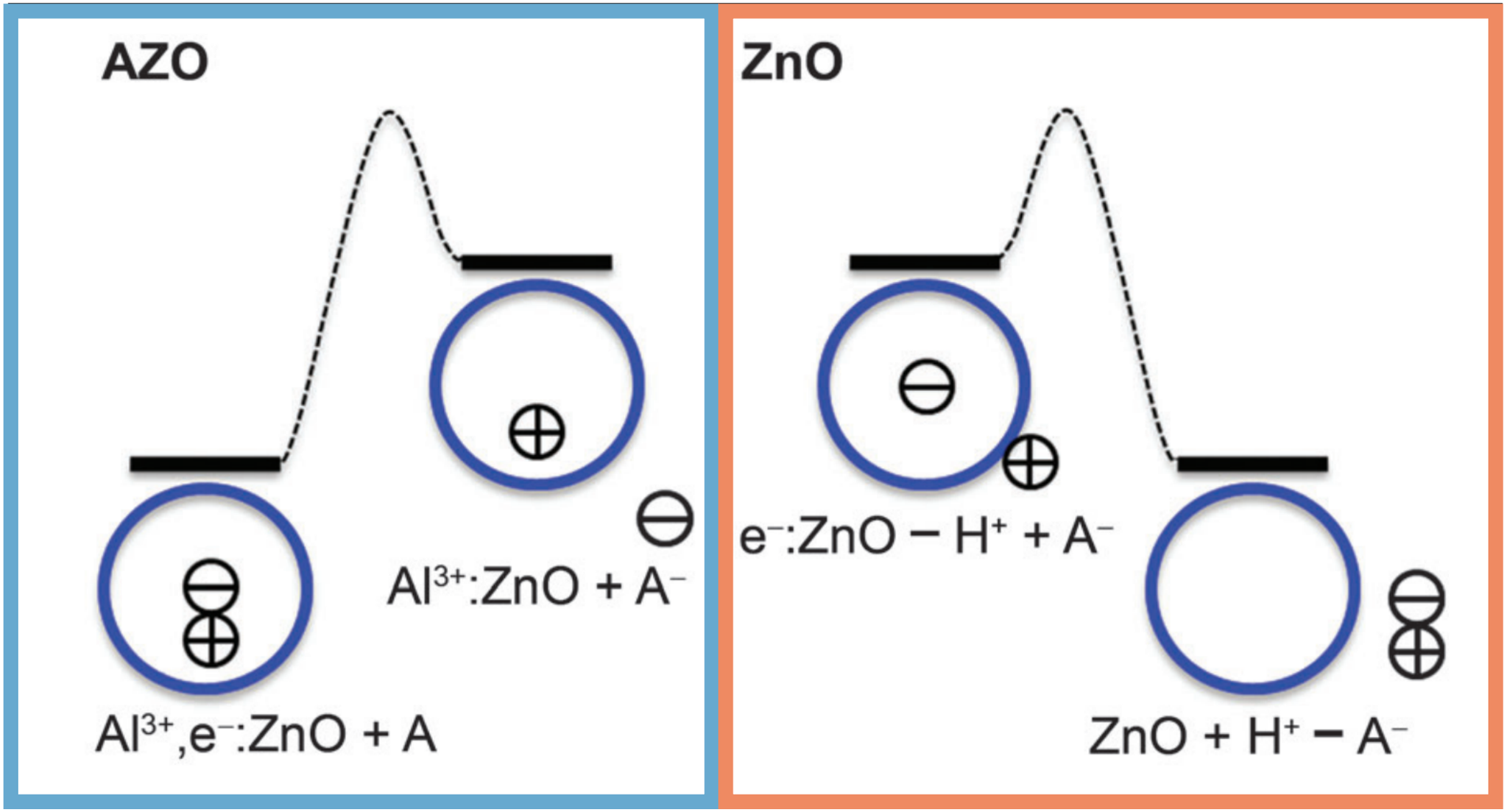
# **$Mn^{2+}$:ZnO** Nanocrystals (NC)
## Adding charge carriers can induce magnetic response




Oschenbein, *et al.* Nature Nanotechnology 4, 681 - 687 (2009)

# $Al^{3+}$-doping ZnO:

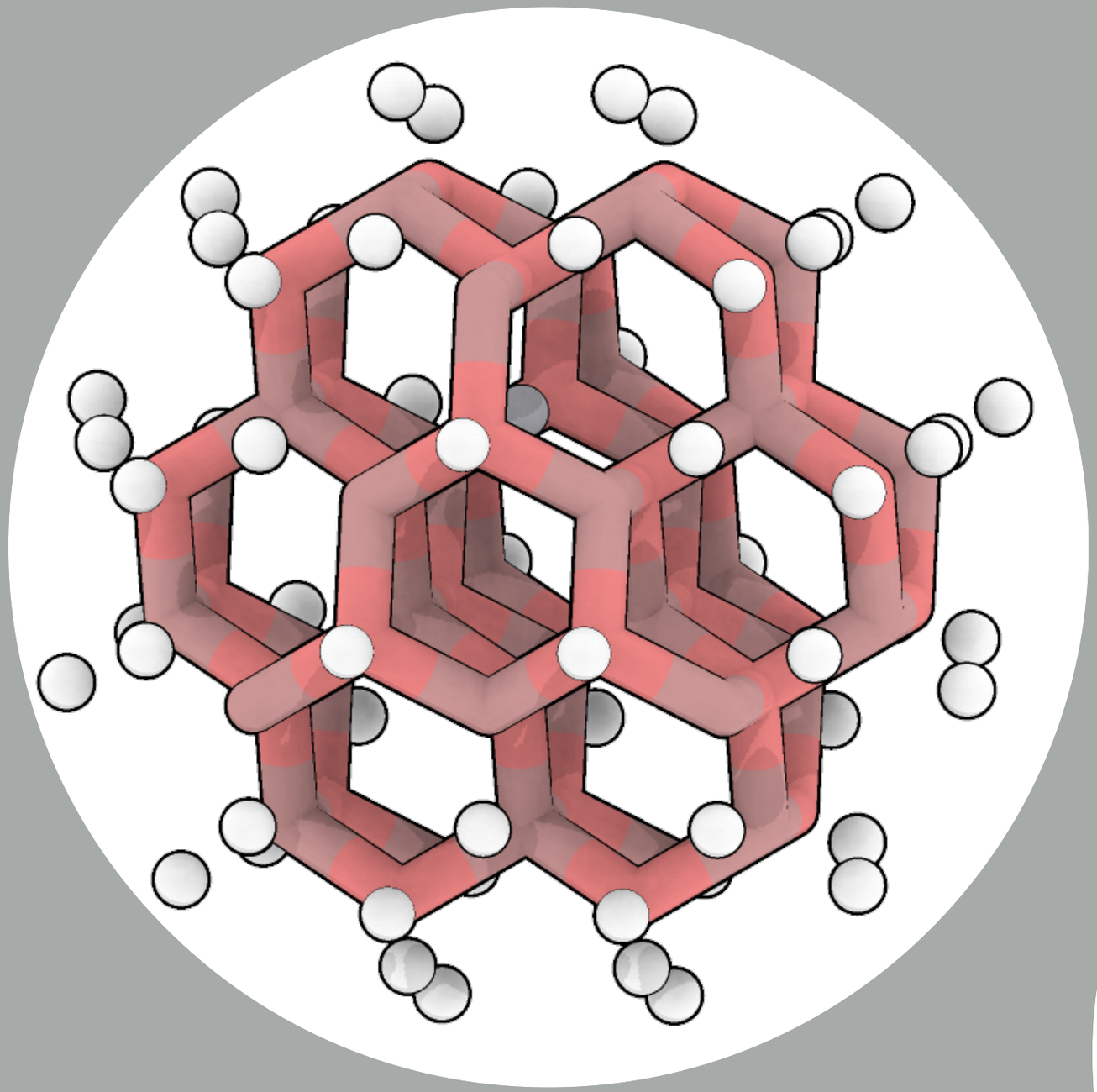
a stable way to add electrons to a ZnO nanocrystal.




Schimpf, *et al.* Chem. Commun. 48, 9352-9354 (2012)

Can we create permanent ferromagnetic QDs by co-doping (Al,Mn)-ZnO?

**Is Al-doped MnZnO fundamentally any different than electronically doped MnZnO?**

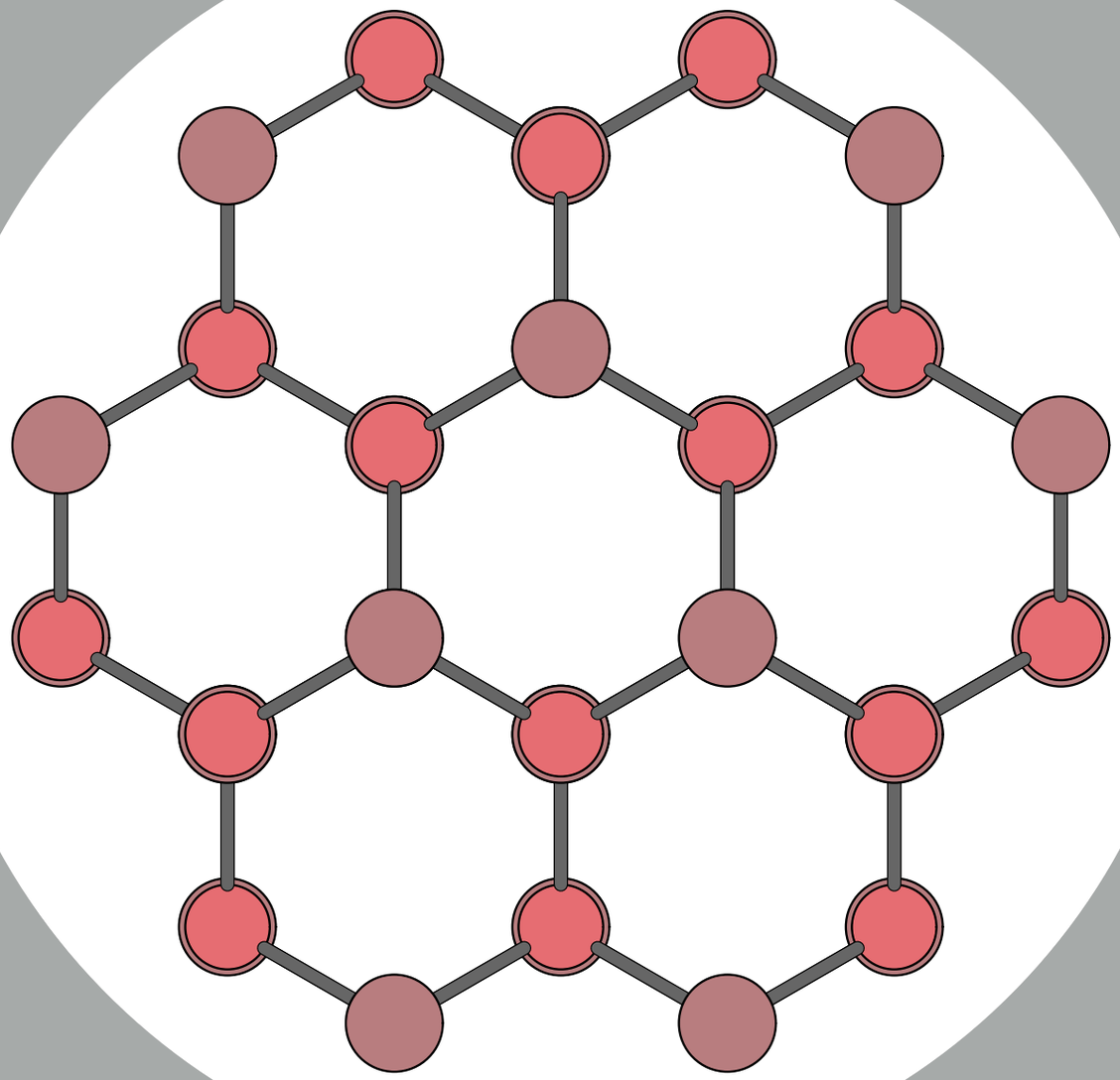


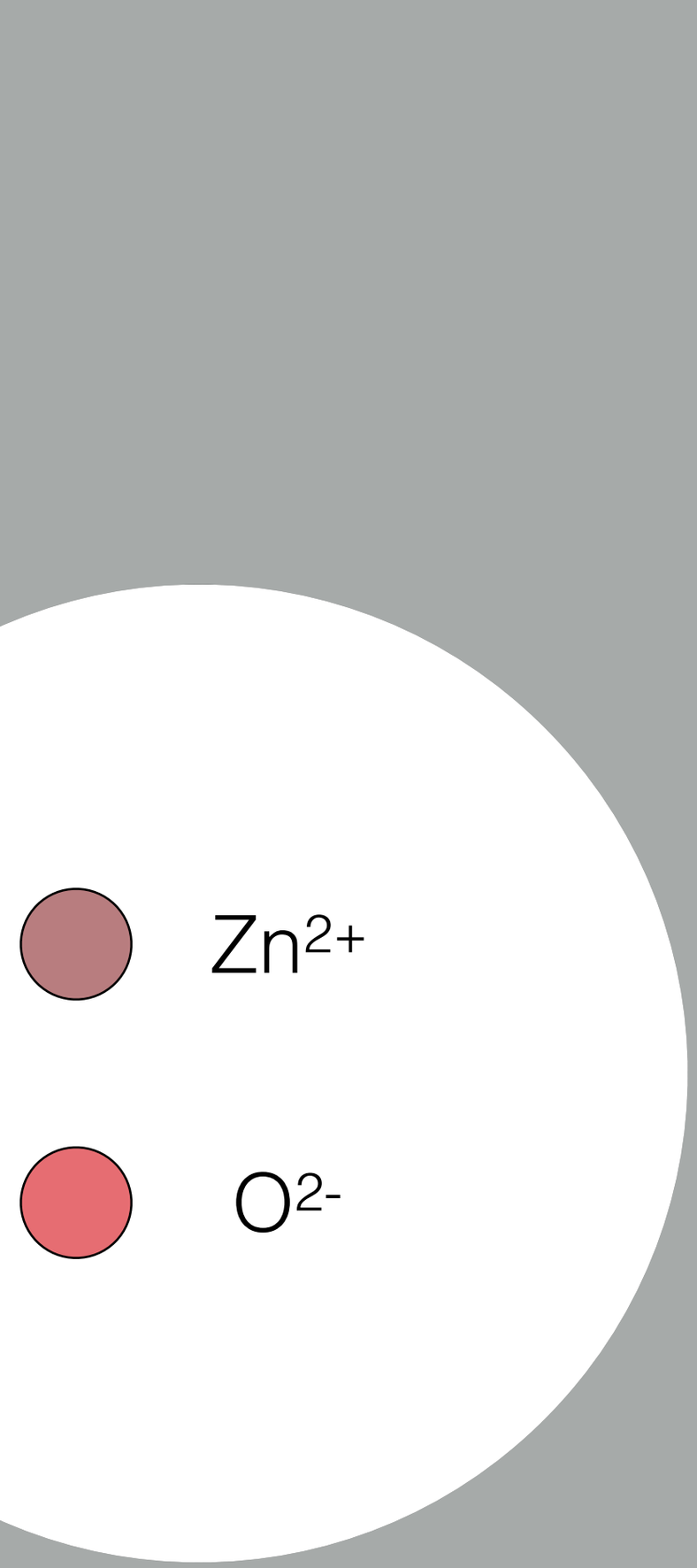
model ZnO

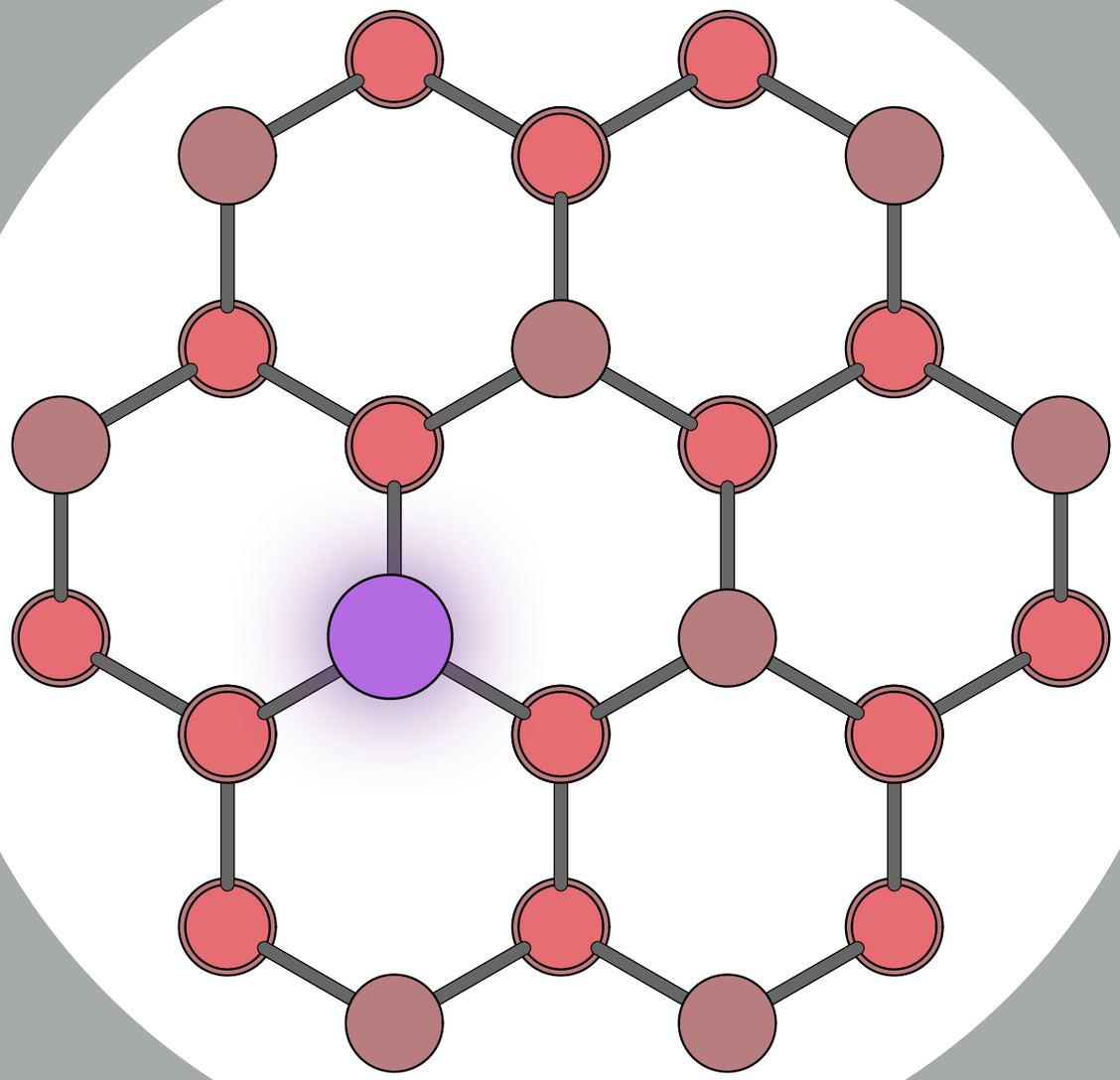
1.3 nm diameter

PBE1PBE/LANL2DZ

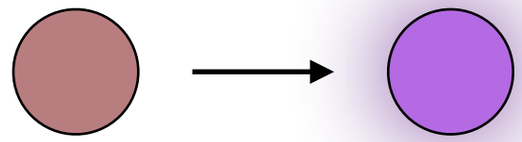
pseudohydrogen capping



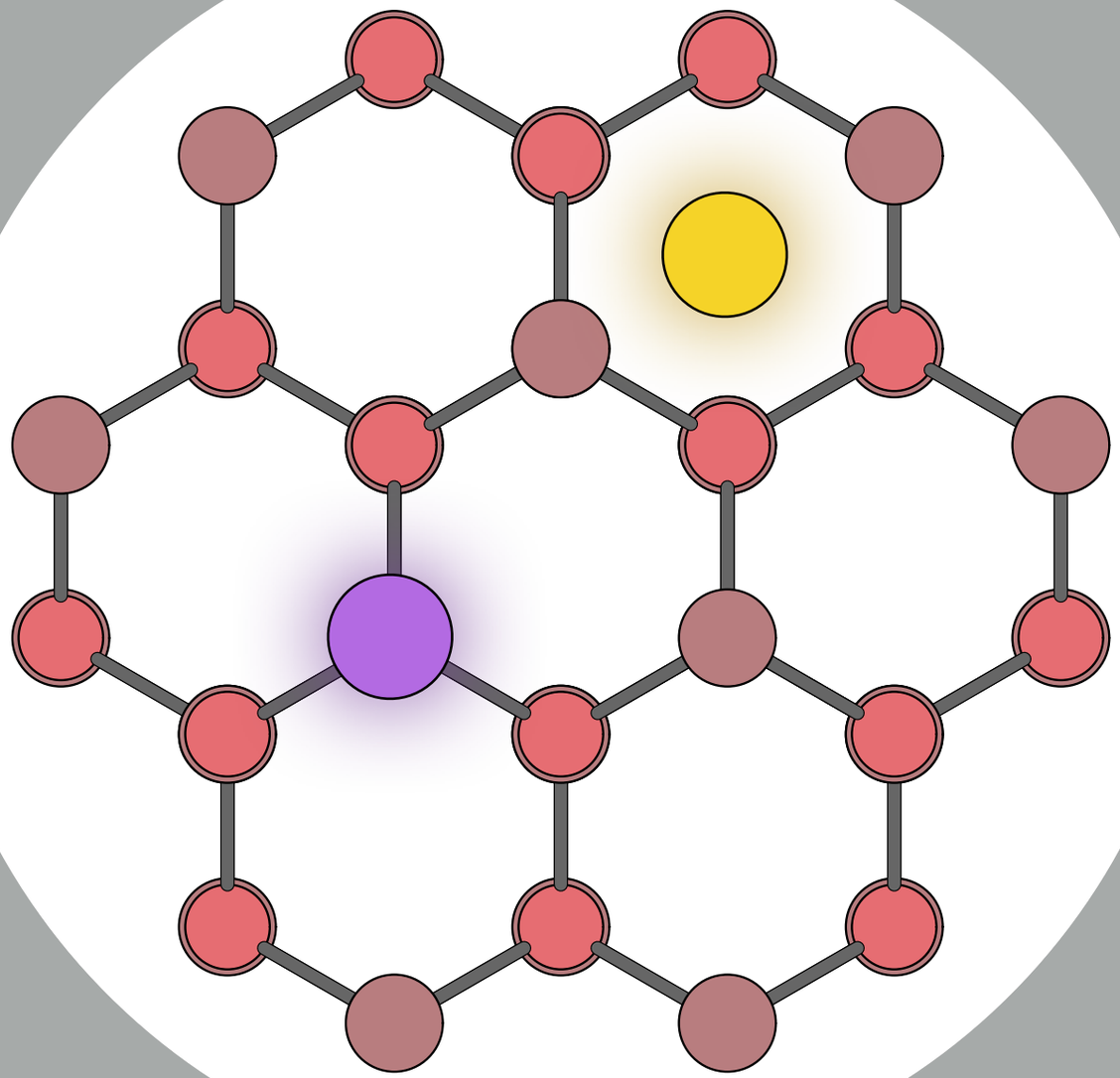

$Zn^{2+}$
$O^{2-}$



Replace $Zn^{2+}$ with $Mn^{2+}$



S = 5/2



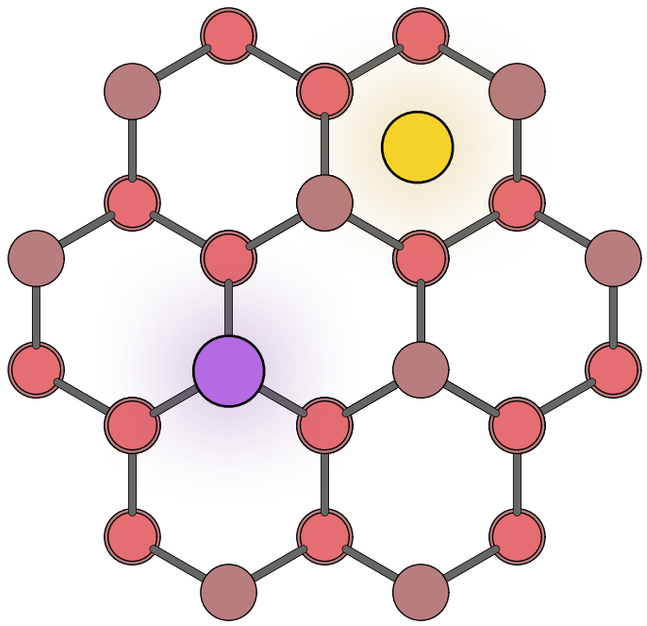
Add charge carrier

electron

hole

S = 1/2

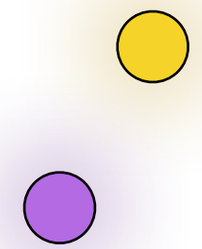
$\hat{H}_{exact}$



unparameterized;
all interactions

Map DFT to
spin Hamiltonian

$\hat{H}_{HDVV}$



$-2J\hat{S}_i\hat{S}_j$

only spin
interactions

# Map DFT to spin Hamiltonian

$H_{\text{eff}} =$ Energy difference of DFT spin configurations

Plug in spin of
local spin subsystem

$$H_{\text{eff}} = J[S_i(S_i+1) + S_j(S_j+1) - S_{i+j}(S_{i+j}+1)]$$

Solve for $J$

*more details at end

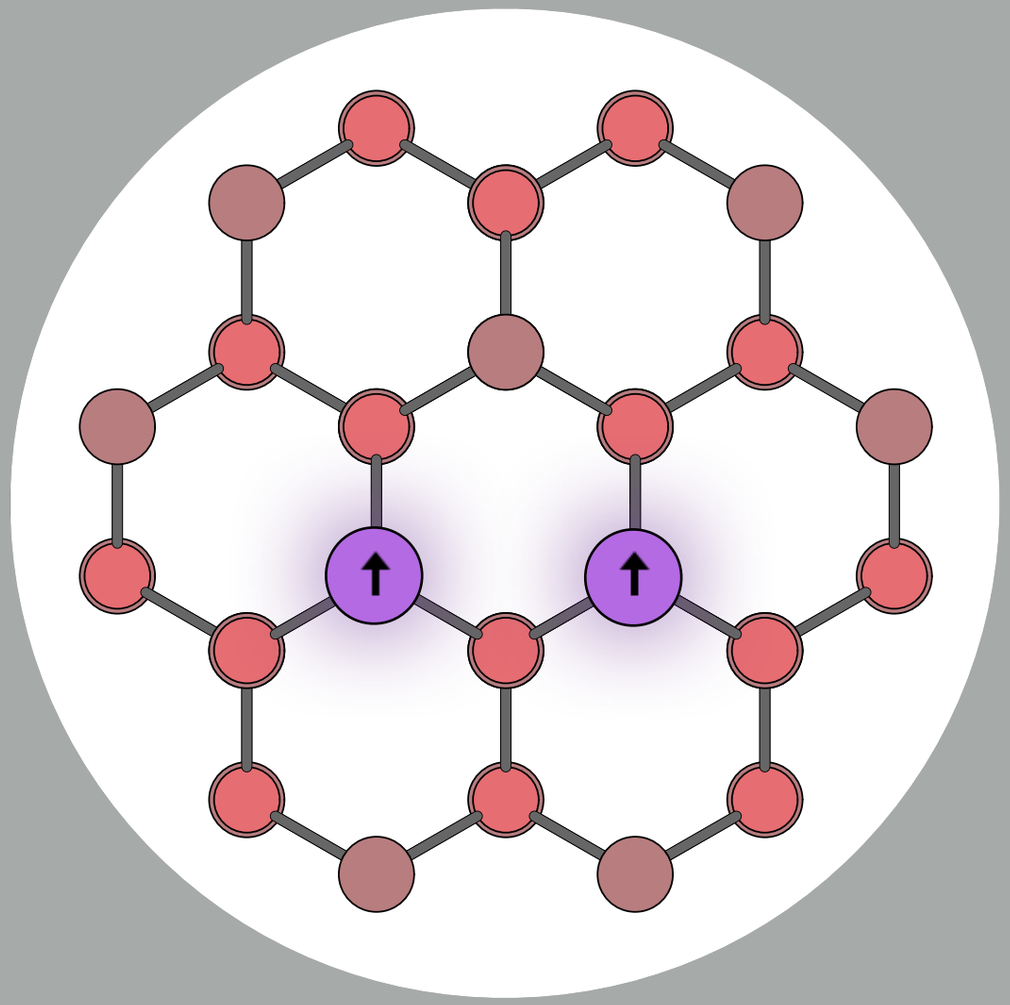
**Two-center spin interactions**:
charge-doped MnZnO quantum dots

$Mn^{2+}$ electron hole

| | | | |
|---|---|---|---|
| $h^+$ | ↑ ↓ | $J_{\text{p-d}} = -152\ \text{meV}$ | anti-ferromagnetic (AFM) |
| $e^-$ | ↑ ↑ | $J_{\text{s-sd}} = +11\ \text{meV}$ | ferromagnetic (FM) |
| $Al^{3+}$ | ↑ ↑ | $J_{\text{s-sd}} = +9\ \text{meV}$ | ferromagnetic (FM) |

$$J > 0 \rightarrow \text{FM}, \quad J < 0 \rightarrow \text{AFM}$$

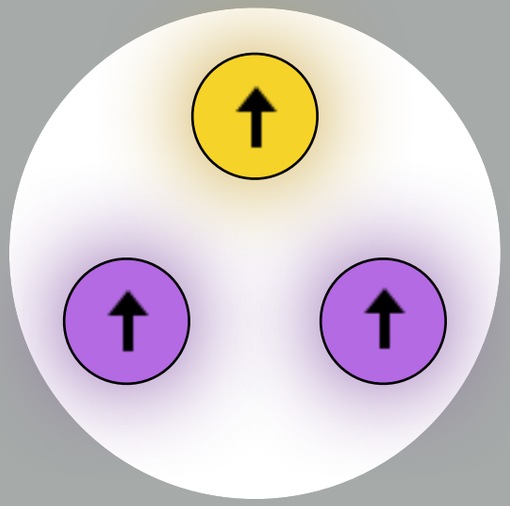
Nearest neighbor $Mn^{2+}$ — $Mn^{2+}$
anti-ferromagnetic (AFM)
super exchange (SE) interactions



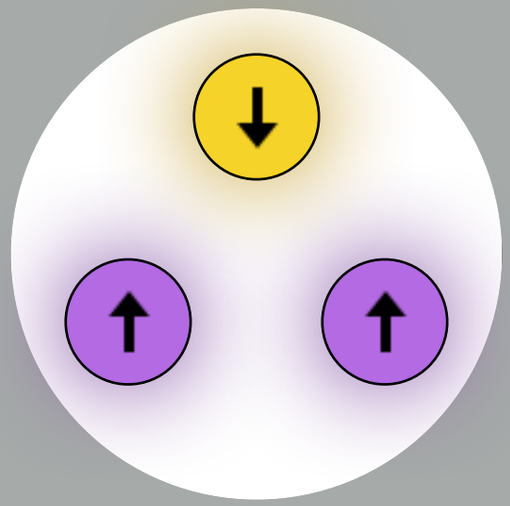
$$J_{\mathrm{SE}} = -2.4\ \mathrm{meV}$$

Subsequent interaction with charge carrier
leads to spin frustrated interactions

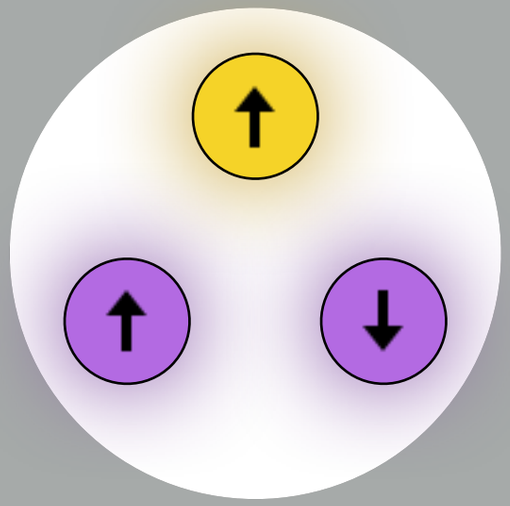
Gives rise to three possible spin configurations



High-spin
(HS)



ferromagnetic
(FM)

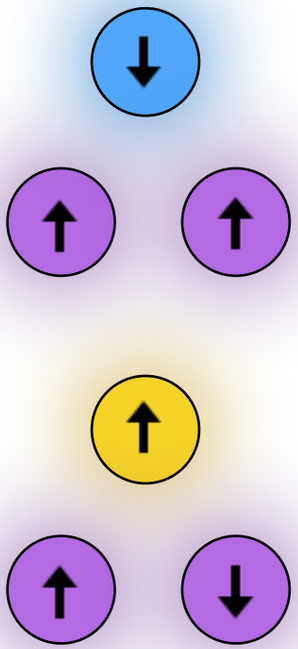
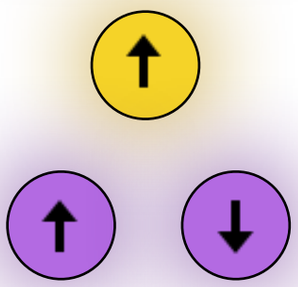


anti-ferromagnetic
(AFM)

$Mn^{2+}$ charge-carrier

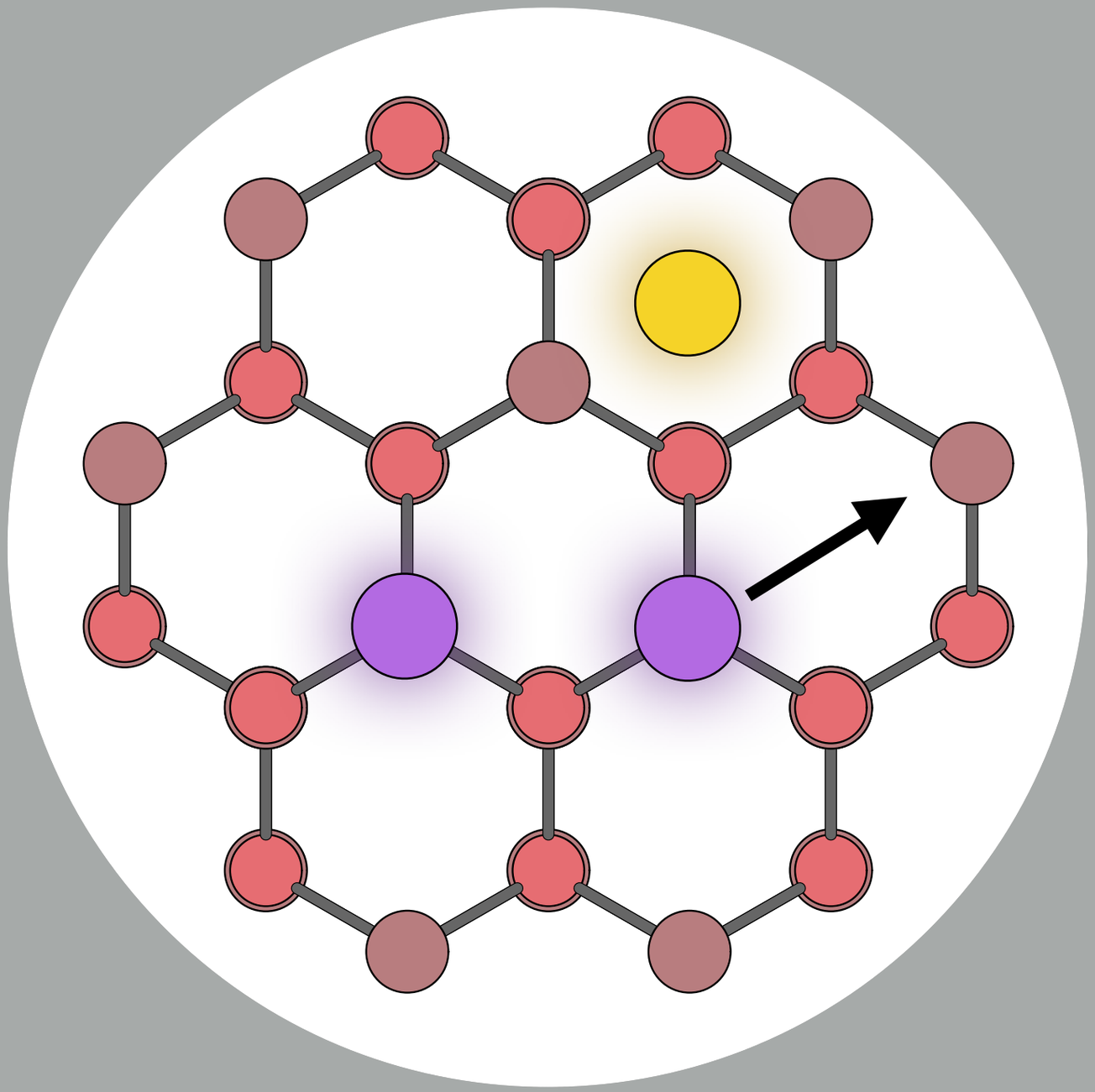
**Three-center spin interactions**:
charge-doped MnZnO quantum dots

$Mn^{2+}$ electron hole

| | | | |
|---|---|---|---|
| $h^+$ |  | $J_{\text{SE}} = -13$ meV<br>$J_{\text{p-d}} = -127$ meV | ferromagnetic (FM) |
| $e^-$ | | $J_{\text{SE}} = -2$ meV<br>$J_{\text{s-sd}} = +11$ meV | anti-ferromagnetic (AFM) |
| $Al^{3+}$ |  | $J_{\text{SE}} = -2$ meV<br>$J_{\text{s-sd}} = +9$ meV | anti-ferromagnetic (AFM) |

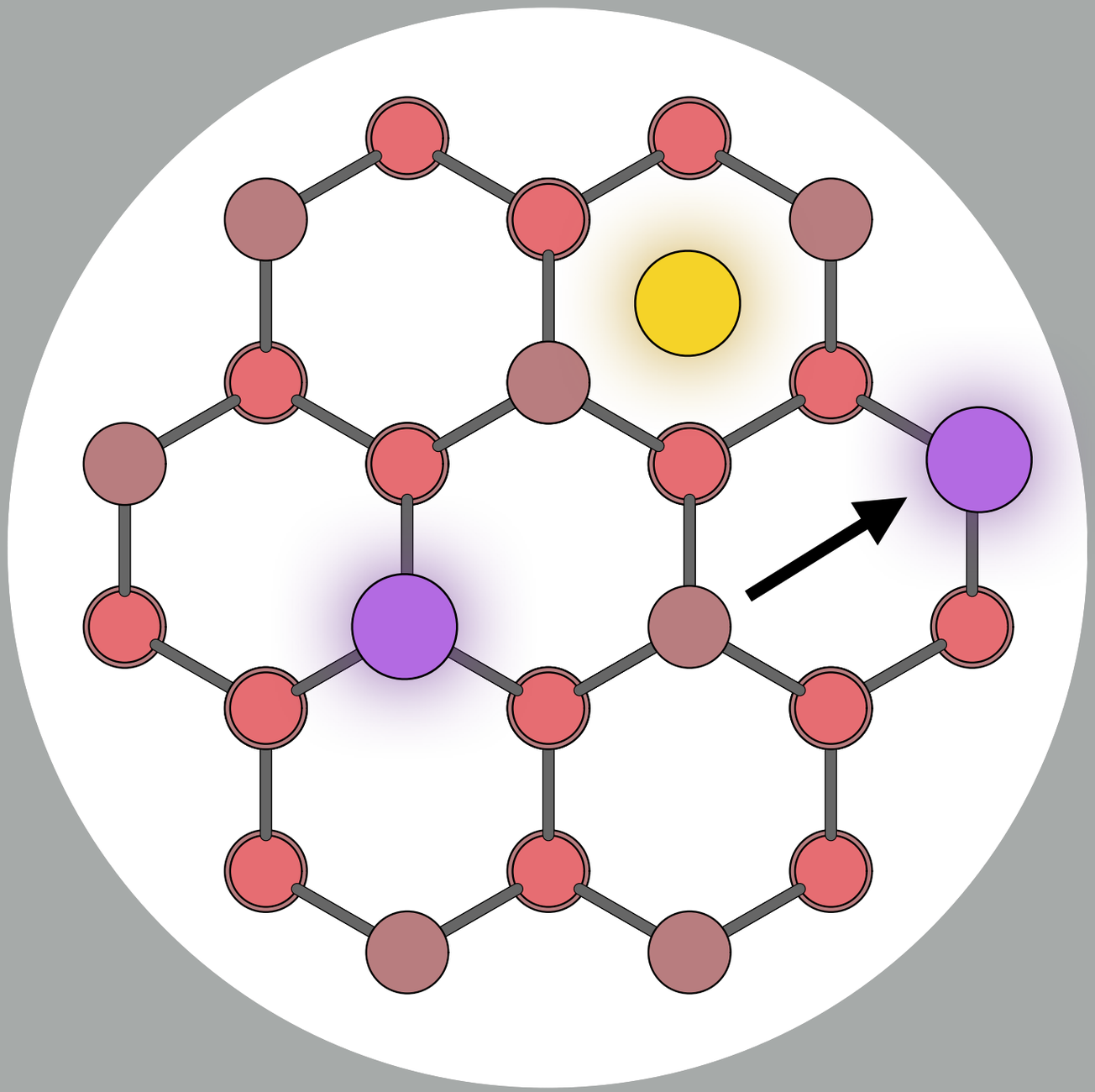
Spins compete, and FM favored when $5J_{\text{SE}} > J_{\text{carrier-Mn}}$

Of course, the interaction is highly position dependent.



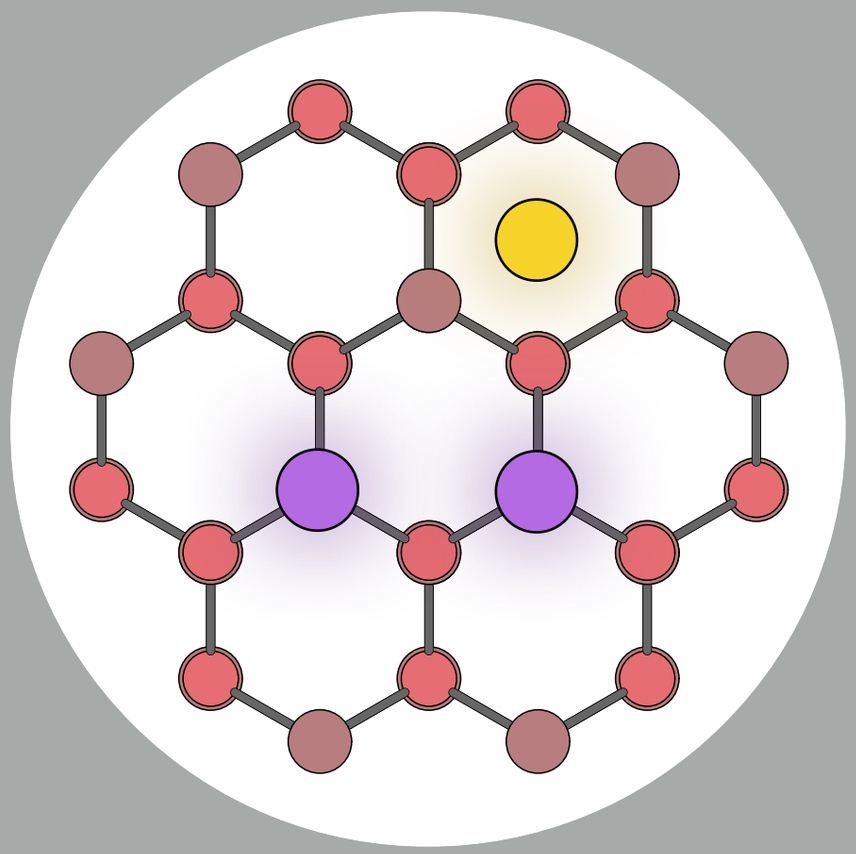
Split up Mn dimer, lose SE interactions, FM favored again.

Of course, the interaction is highly position dependent.



Split up Mn dimer, lose SE interactions, FM favored again.

# (Mn,Al) co-doped ZnO quantum dots



Aluminum-doping leads to stable charge carrier

Charge can mediate ferromagnetic spin coupling between distant Mn centers

Thank you!

# Map DFT to spin Hamiltonian

$$H_{\text{eff}} = \Delta E = E(\text{DFT spin config 1}) - E(\text{DFT spin config 2})$$

$$H_{\text{eff}} = -2J\hat{S}_i \cdot \hat{S}_j$$

$$-2\hat{S}_i \cdot \hat{S}_j = \hat{S}_i^2 + \hat{S}_j^2 - (\hat{S}_i + \hat{S}_j)^2$$ complete the square

$$\hat{S}_i^2|\psi\rangle = S_i(S_i+1)|\psi\rangle$$ act on spin eigenfunction

This gives for spin subsystems ( *i* ) and ( *j* ) :

$$H_{\text{eff}} = J[S_i(S_i+1) + S_j(S_j+1) - S_{i+j}(S_{i+j}+1)]$$