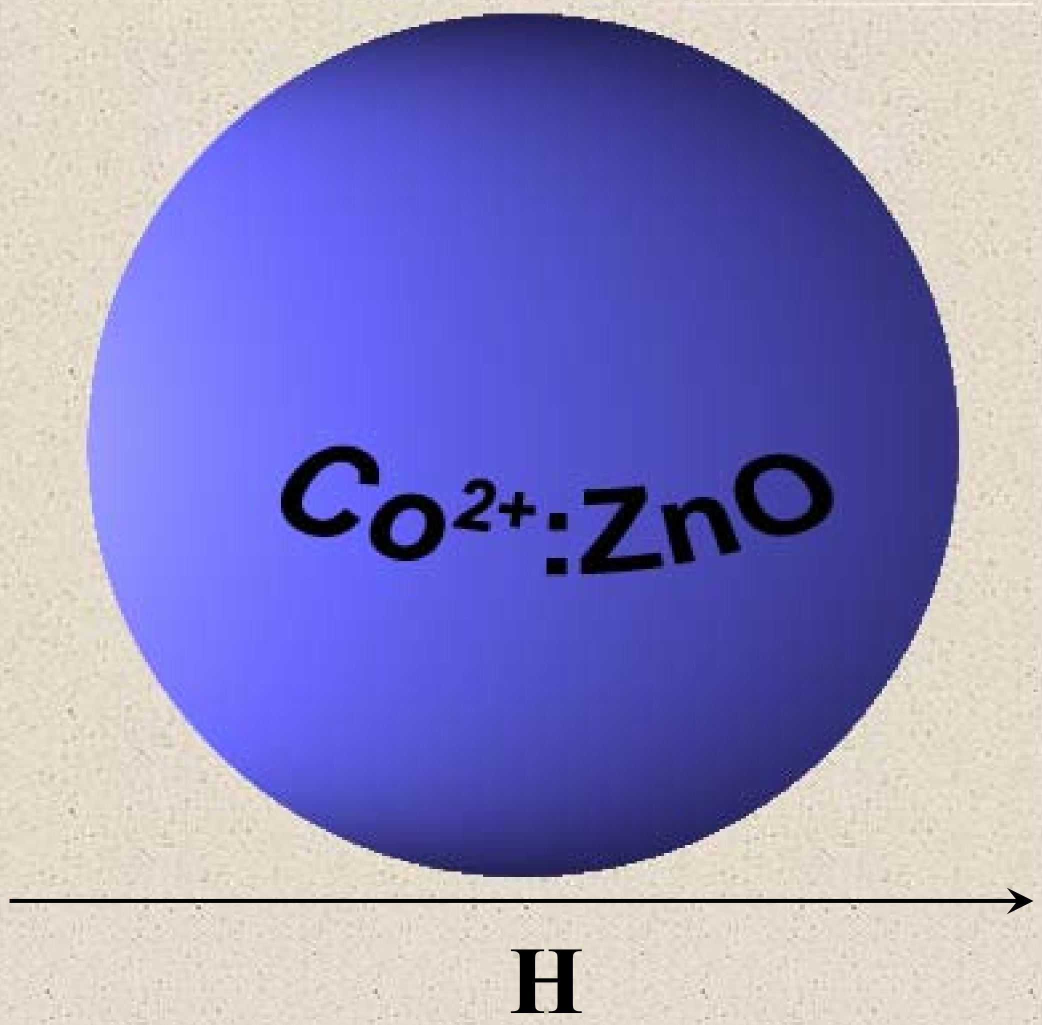
Exchange Interactions in Magnetic ZnO Quantum Dots

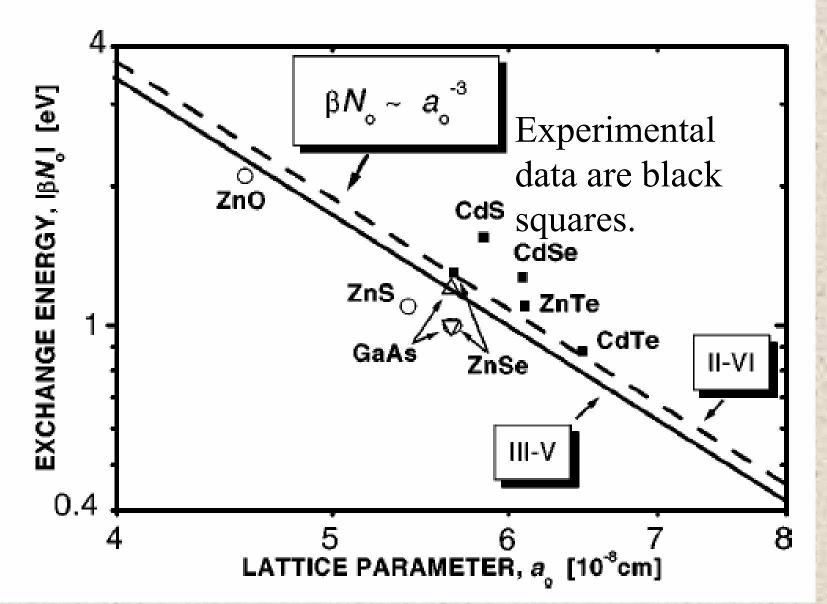


One Model for Ferromagnetism

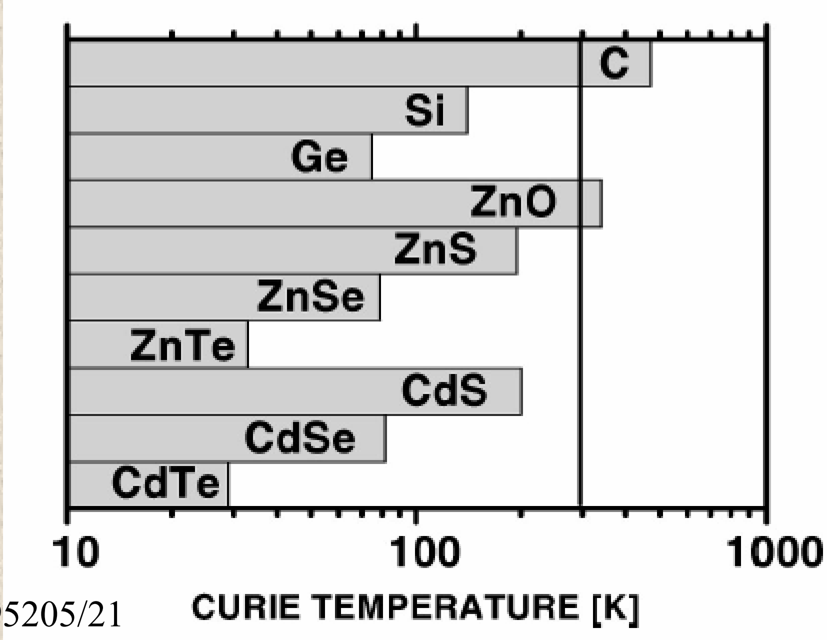
This model assumes β is a constant and that only N_0 changes between semiconductors. It does not take into account changes in the LMCT transition.

$$T_C \propto x N_0 S(S+1)\beta^2$$

$$N_0\beta \approx -\frac{1}{S}\left[\frac{\left\langle \Psi_{VB} \middle| \hat{H}_{pd} \middle| \Psi_{t_2} \right\rangle^2}{E_{LMCT}}\right]$$



Experimental data are black squares.

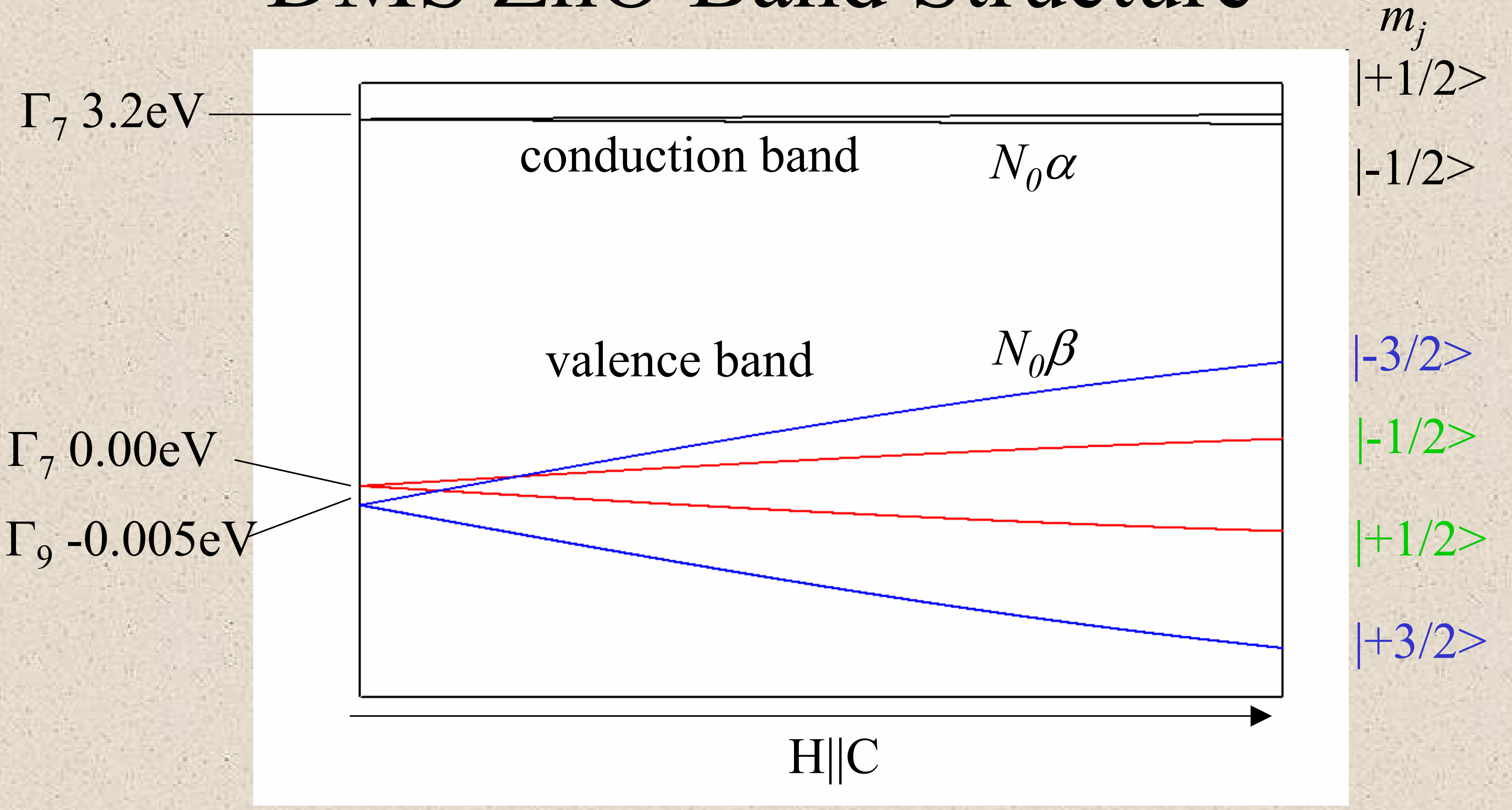


Dietl, T.; Ohno, H.; Matsukura, F., *Phys. Rev. B* **2001,** *63*, 195205/1-195205/21

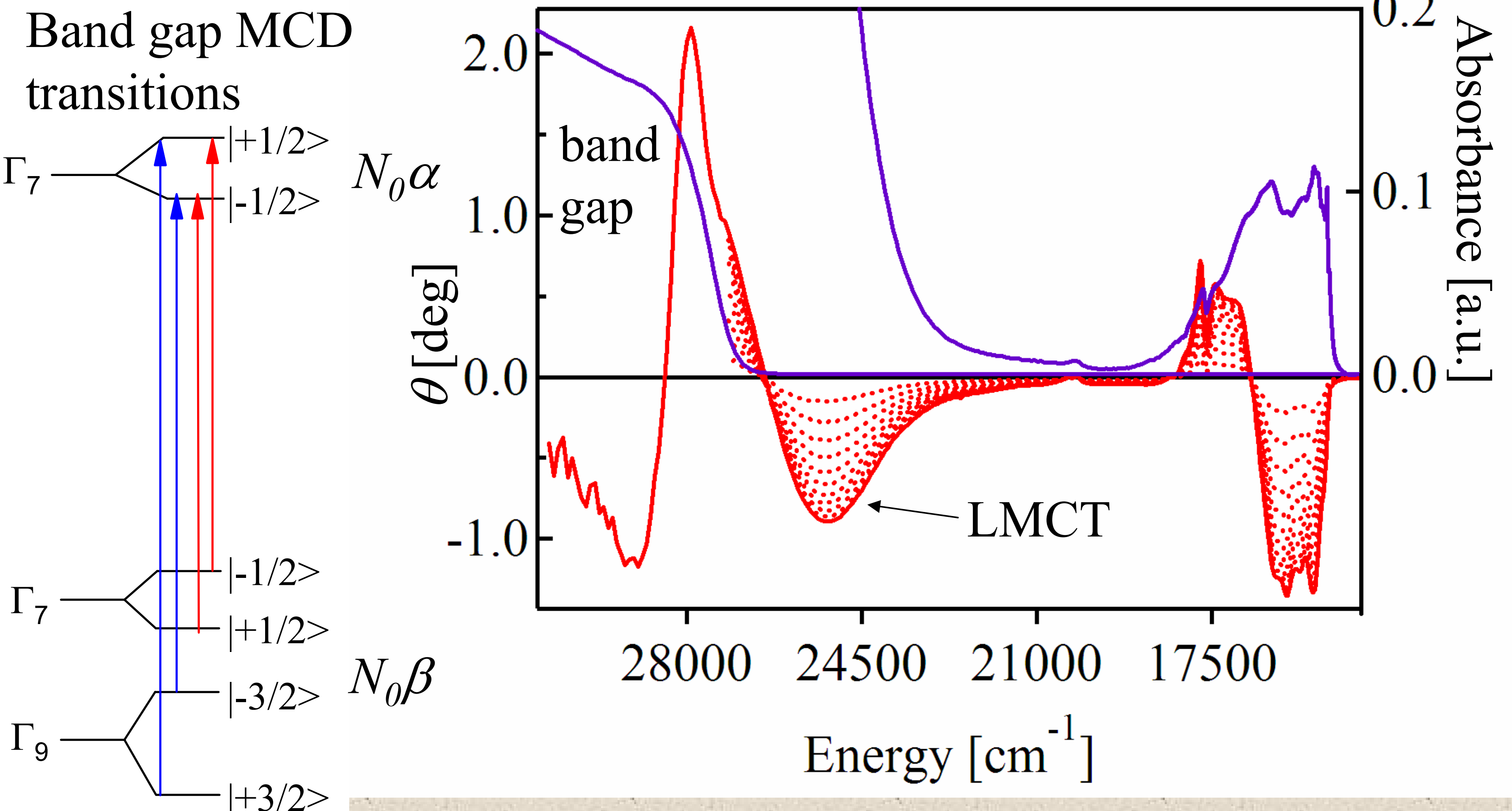
Exchange Between the Dopants and Band Electrons

- External magnetic fields magnetize the ground state of substitutional dopant ions in ZnO.
- In the ground state the ZnO DMSs band electrons are paired, and are confined to the valence band.
- The band electrons of paramagnetic DMS-ZnO are only magnetized in the excited state.

The One Electron Picture of the DMS ZnO Band Structure



Magnetization of Co^{2+}:ZnO Quantum Dots



Relationship between CT and Ferromagnetism.

Intensity of LMCT transtion

$$\psi_A^{'} = \sqrt{1-c^2}\,(\psi_A) - c\,\psi_D$$

metal t_2 orbital — VB Oxo

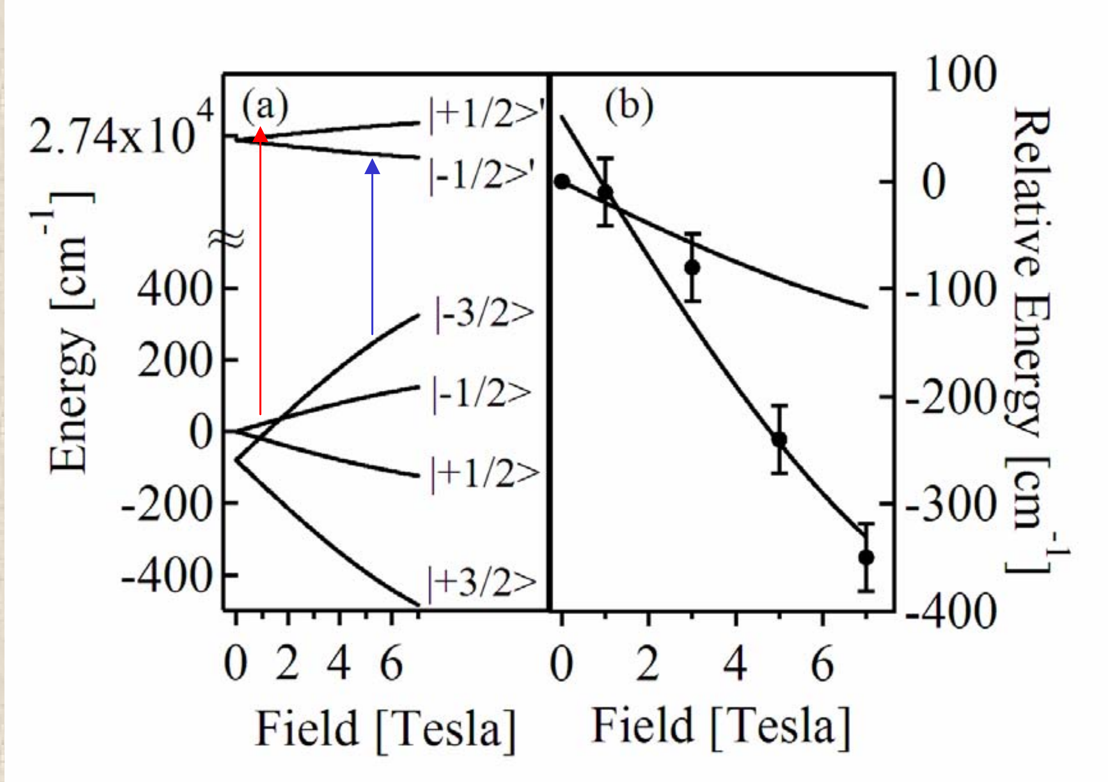
$$c \approx \frac{-\langle \psi_A | H | \psi_D \rangle}{E_A - E_D}$$

$$N_0\beta \approx -\frac{1}{S}\left[\frac{\langle \Psi_{VB} | \hat{H}_{pd} | \Psi_{t_2} \rangle^2}{E_{LMCT}}\right]$$

Exchange between the VB and the TM^{2+}

Calculation of $N_0\beta$ for Co^{2+}:ZnO Nanocrystals

$N_0\beta$ = -2.2(±0.3)eV from these experimental data. The predicted value is ~ -5.0 eV based on the model presented previously.

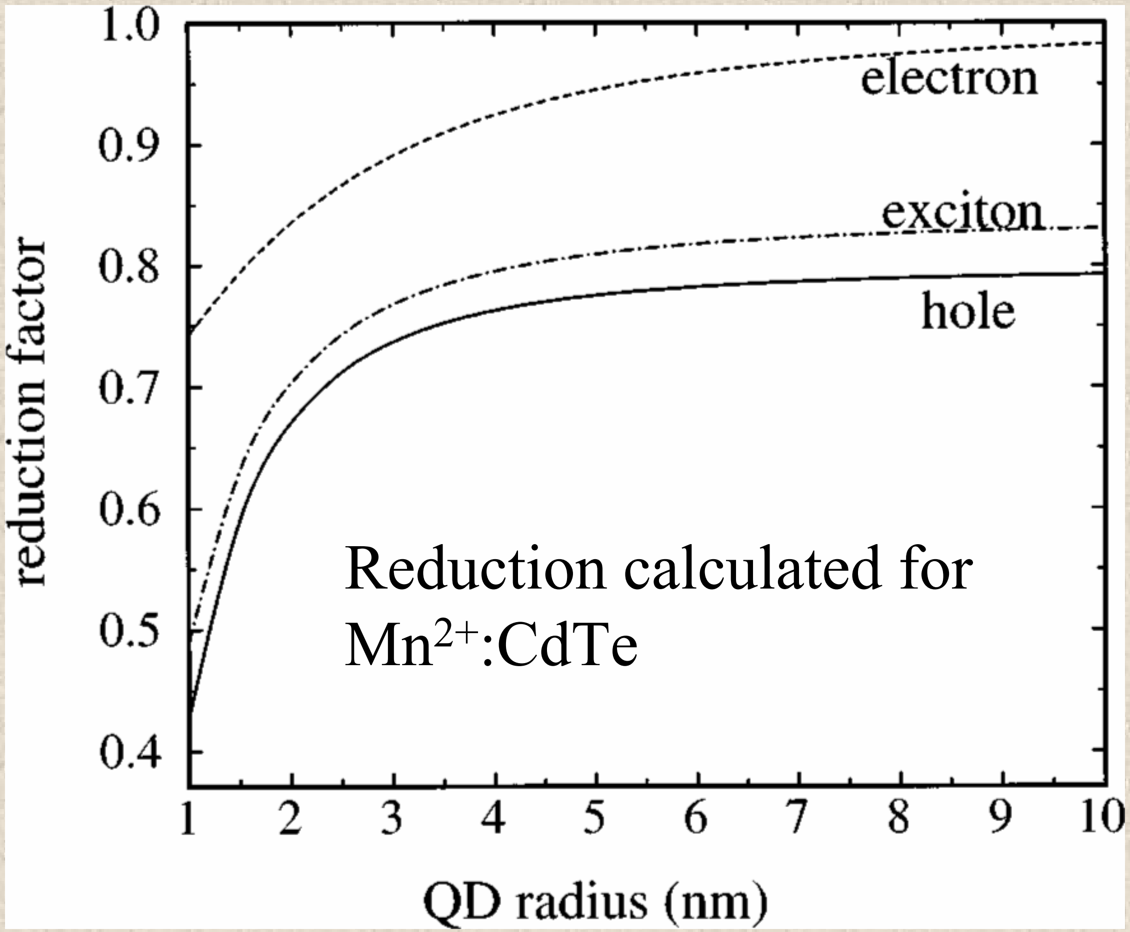


Experimental values of $N_0\beta$ in eV

	Co^{2+}	Mn^{2+}
CdTe	-2.33	-0.83
CdSe	-2.12	-1.30
ZnTe	-3.03	-1.10

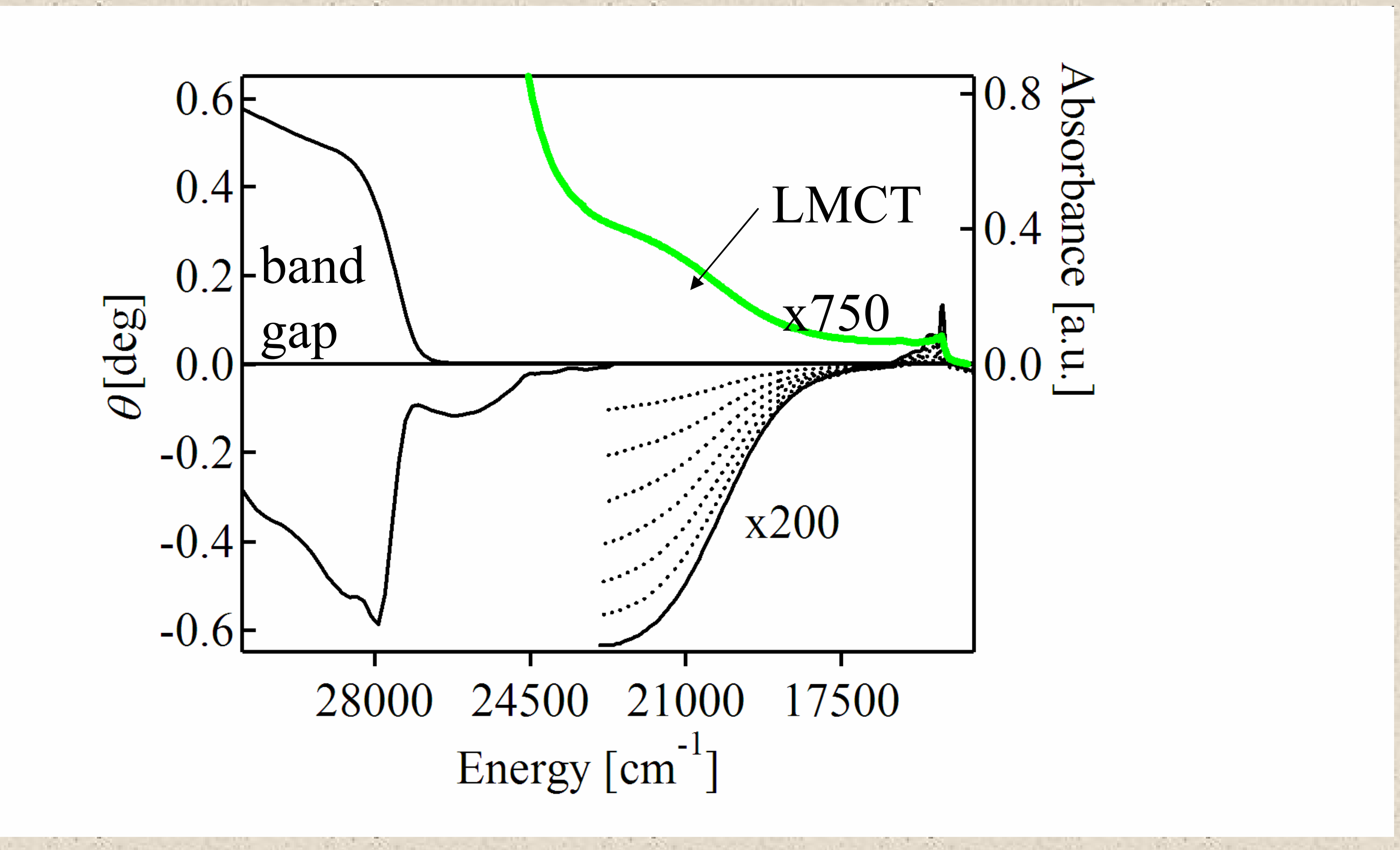
$$E_{CB}(m_j)=E_{CB}-m_jN_0\alpha x<S_z>$$
$$E_{VB}(m_j)=E_{VB}-1/3m_jN_0\beta x<S_z>$$

Angular Momentum Reduction in Quantum Dots



Bhattacharjee, A.K. *Phys. Rev. B*. **1998,** *52,* 15660-15665

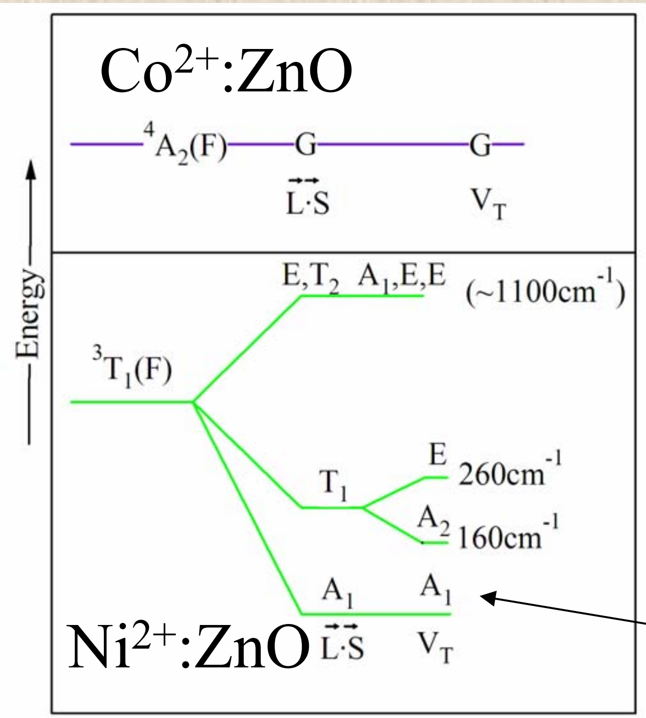
Ni^{2+}:ZnO Charge Transfer Transition



$$Ni^{2+} + h\nu \rightarrow Ni^{+} + h^{+}$$

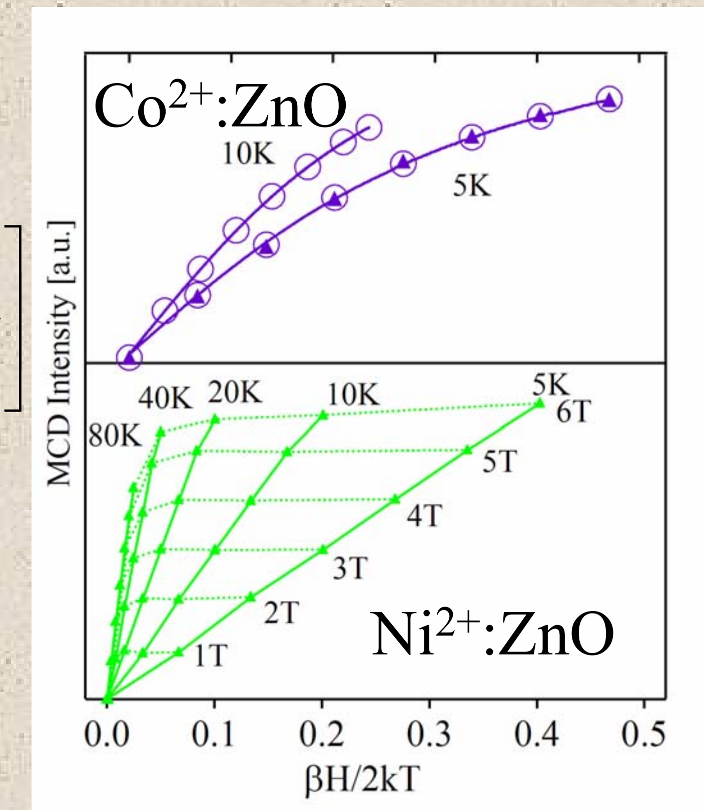
$N_0\beta$ and Ni^{2+}:ZnO

Experimental LF magnetization



$$N_0\beta \approx -\frac{1}{S}\left[\frac{\langle \Psi_{VB} | \hat{H}_{pd} | \Psi_{t_2} \rangle^2}{E_{LMCT}}\right]$$

Non-magnetic ground state for tetrahedral Ni^{2+}.



$N_0\beta$ should actually be larger (~-4.3 eV) for Ni^{2+} than $N_0\beta$ for Co^{2+}, but the magnetization of the nickel ground state is one order of magnitude less than cobalt due to the in state spin-orbit coupling found in tetrahedral nickel.

Conclusions

- MCD demonstrates that both the Co^{2+}:ZnO and Ni^{2+}:ZnO quantum dots are true DMS materials.
- $N_0\beta$ for Co^{2+}:ZnO and Ni^{2+}:ZnO nanocrystals was calculated using a combination of Zeeman measurements, absorption and MCD spectroscopy.
- The small value of $N_0\beta$ is interesting and possibly due to the reduction of angular momentum found in quantum confined structures.

Acknowledgments

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