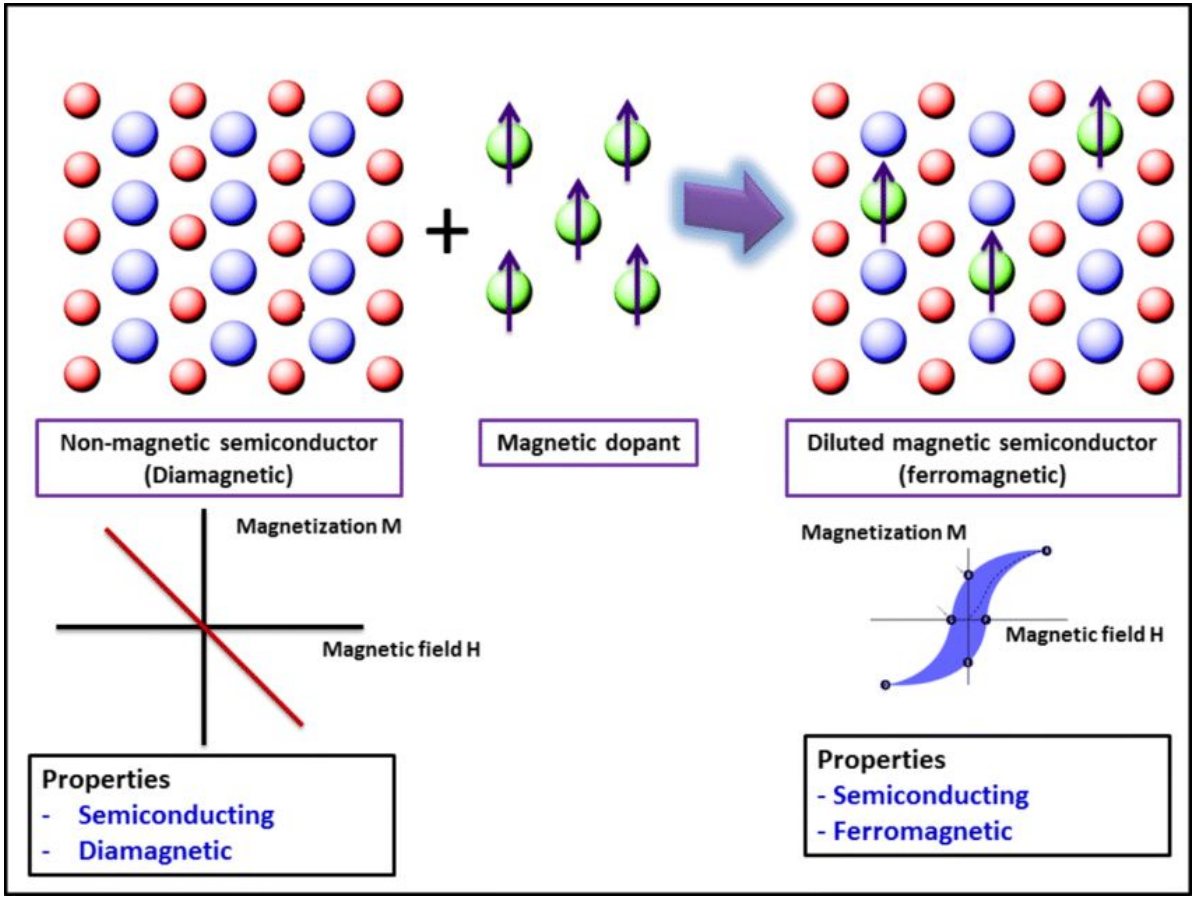
Mn-doped monolayer MoS2: An atomically thin dilute magnetic semiconductor

Ben Safvati

References:

- **A. Ramasubramaniam and D. Naveh, Phys. Rev. B, 2013, 87, 195201**
- **Caleb's Paper: Quantum Imaging of Single-Atom Spin-Splitting in a Monolayer Semiconductor**

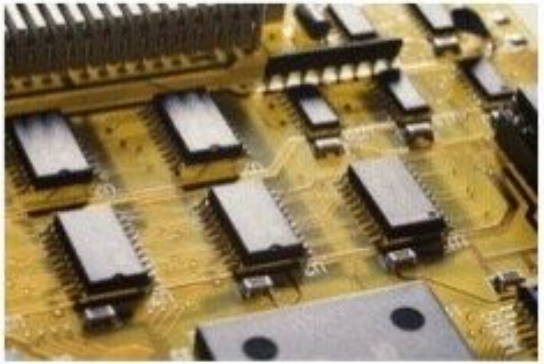
Introduction: Dilute Magnetic Semiconductors



Introduction: DMSs for Spintronics

Nearly incompatible technologies in present-day computers:

semiconductors: processing



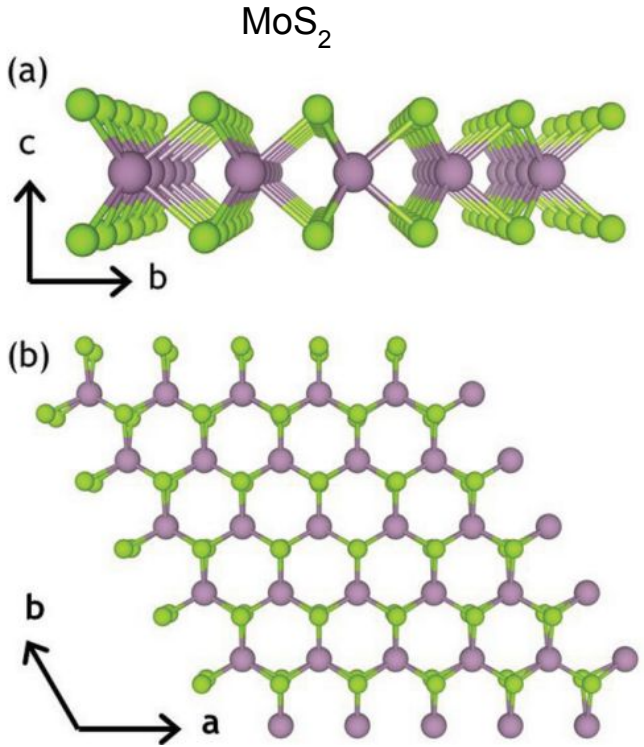
ferromagnets: data storage

ferromagnetic semiconductors: **integration on a single chip?**

single-chip computers for embedded applications:
cell phones, intelligent appliances, security

Transition Metal Dichalcogenide DMSs

- Existing monolayer exfoliation technology (nanoelectronics)
- Direct band gap ~2 eV suitable for semiconductor technology (shrinking transistors)
- Idea: substitute Mo^{4+} with Mn^{4+}, extra 3d valence electron acts to break spin degeneracy, emergent magnetism.



Outline of Paper

- Use Density Functional Theory (DFT) to calculate magnetic ground state properties and exchange coupling constants.
- Map DFT parameters to Heisenberg model, use statistical methods (Monte Carlo) to approximate thermal properties of the material.
- Conclusions: room temp. Ferromagnetism emerges from short-ranged double-exchange interactions in Mn/MoS_2 for dopant concentrations around 10-15%.

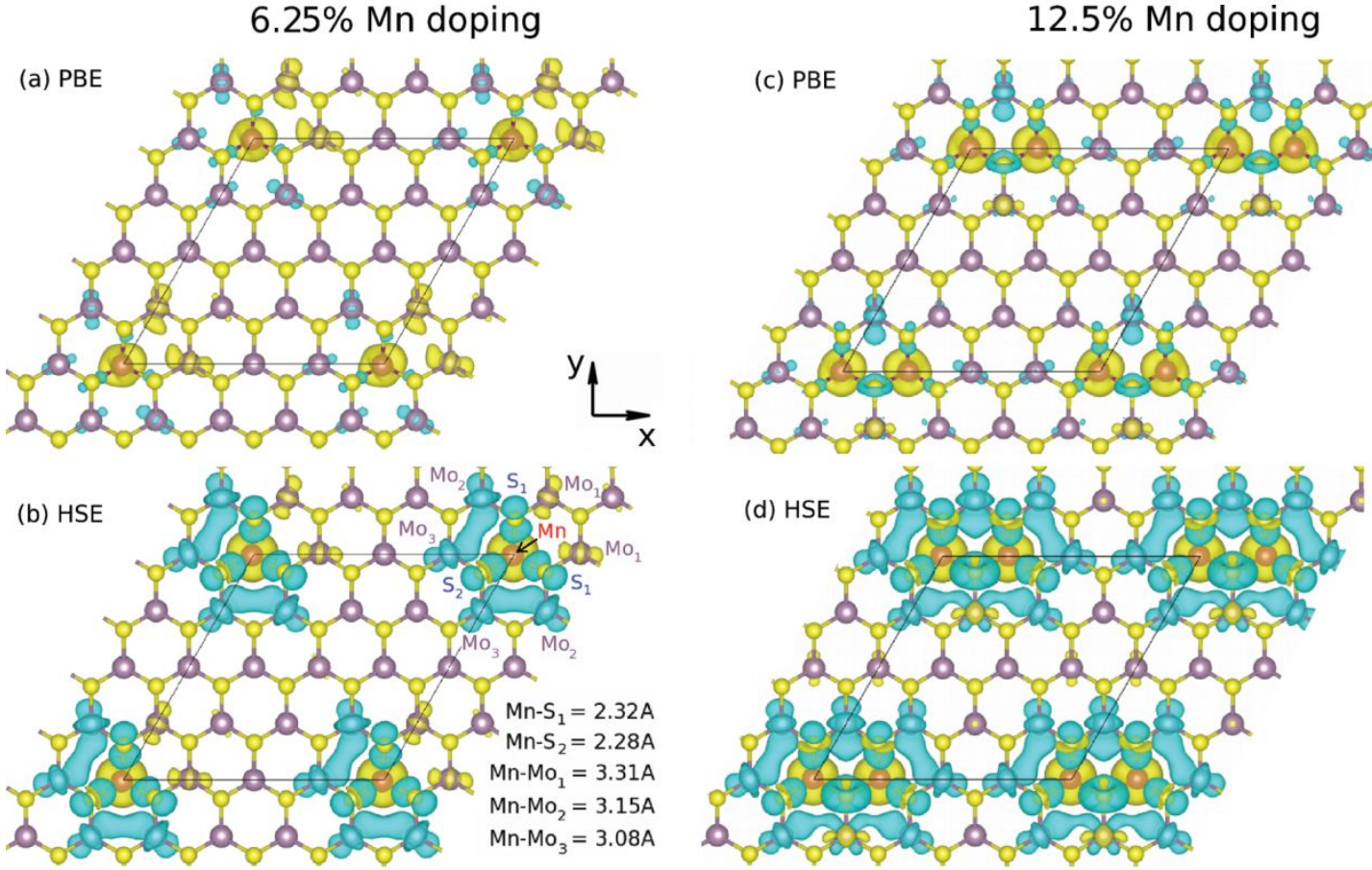
Predictions of DFT

- Basic idea: calculate many-body ground state electronic density from effective single-electron theory, only depends on material crystal structure.

- Comparison of standard model (PBE) with hybrid DFT (HSE).
  - PBE has been shown to predict excessive delocalization of d orbital wavefunctions, leads to inaccurate interaction parameters.
  - HSE corrects for self-interaction errors in standard DFT, improvement of DFT predictions.

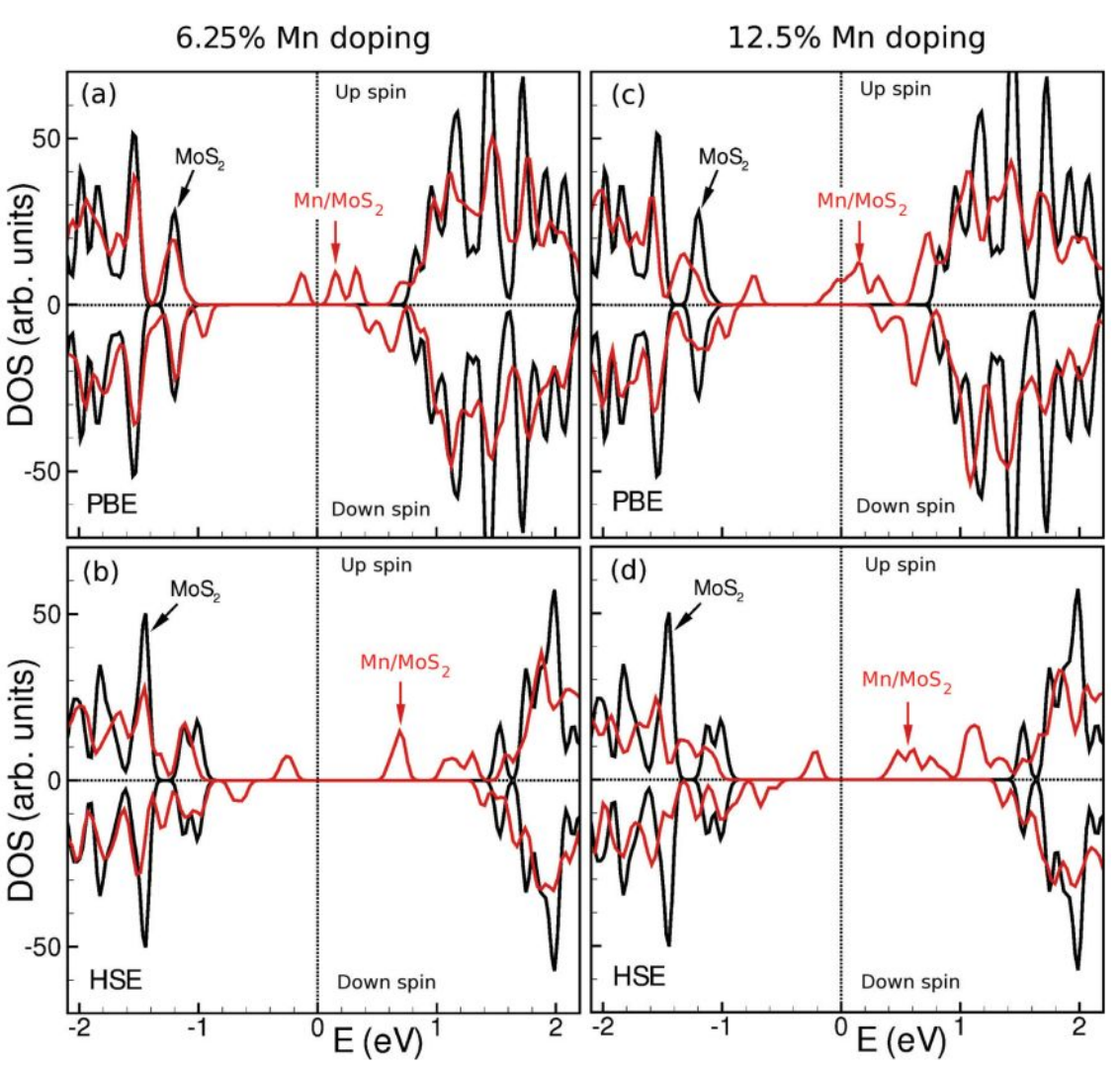
- HSE predictions for MoS_2 band gap agree more closely with experiment.

Predictions of DFT: Spin Density

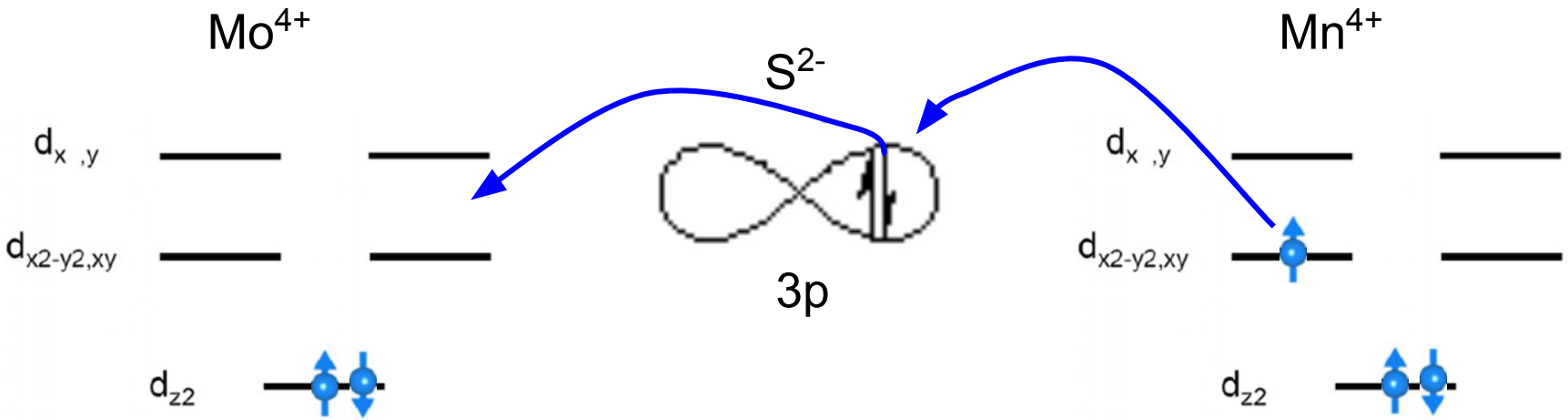


Predictions of DFT: Electronic Density of States

- Mn substitution breaks spin degeneracy, in-gap defect states emerge.
- The two DFT theories have relative shift in energies, leads to half-metallic behavior for PBE model.



Double-exchange Mechanism



Estimating Magnetic Interaction Parameters

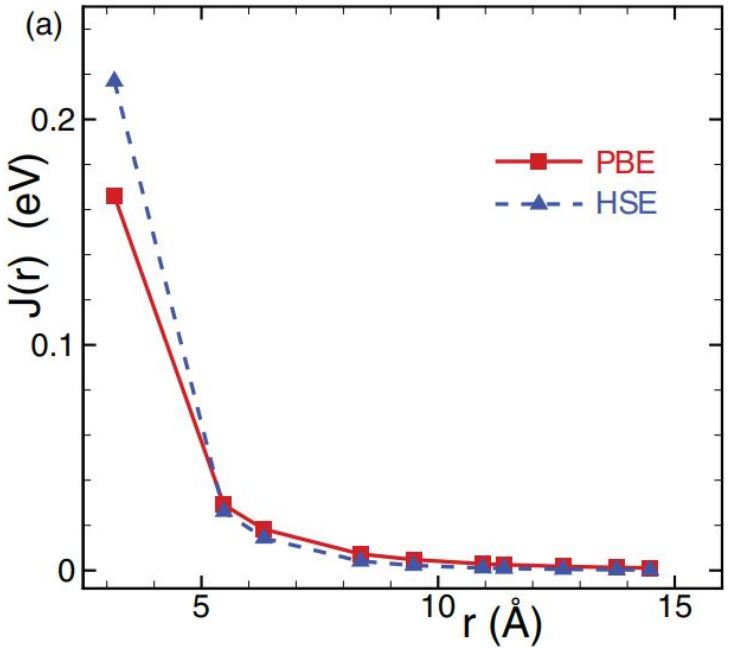
Parameterize interaction with DFT predictions

$$J(r) = \begin{cases} \frac{c}{r^3}\exp[-r/r_0], & \text{if} \quad r \leqslant r_c \\ 0, & \text{otherwise} \end{cases},$$

Supercell	Configuration	Δ_{AFM-FM} (eV) PBE	Δ_{AFM-FM} (eV) HSE	ΔE_{FM} (eV) PBE	ΔE_{FM} (eV) HSE
4×4	1^{st} n.n.	0.18	0.22	0.0	0.0
	2^{nd} n.n.	0.06	0.07	0.37	0.66
	3^{rd} n.n	0.03	−0.00	0.43	0.65
6×6	1^{st} n.n.	0.17			
8×8	1^{st} n.n.	0.17			

Heisenberg Model

$$H = -J\sum_{i,j}\vec{S}_i \cdot \vec{S}_j$$



Monte Carlo Simulation

- Mean-field theory inaccurately predicts magnetic ordering temperature, need to account for spatial disorder.
- Randomized simulation of Mn spins on triangular lattice, estimate percolation threshold.
- Critical temperature for ferromagnetism for Mn concentrations ~15%.