Carrier induced ferromagnetism in II-IV-V_2 Mn-doped chalcopyrites

Paul Kent
University of Tennessee

Thomas Schulthess
ORNL

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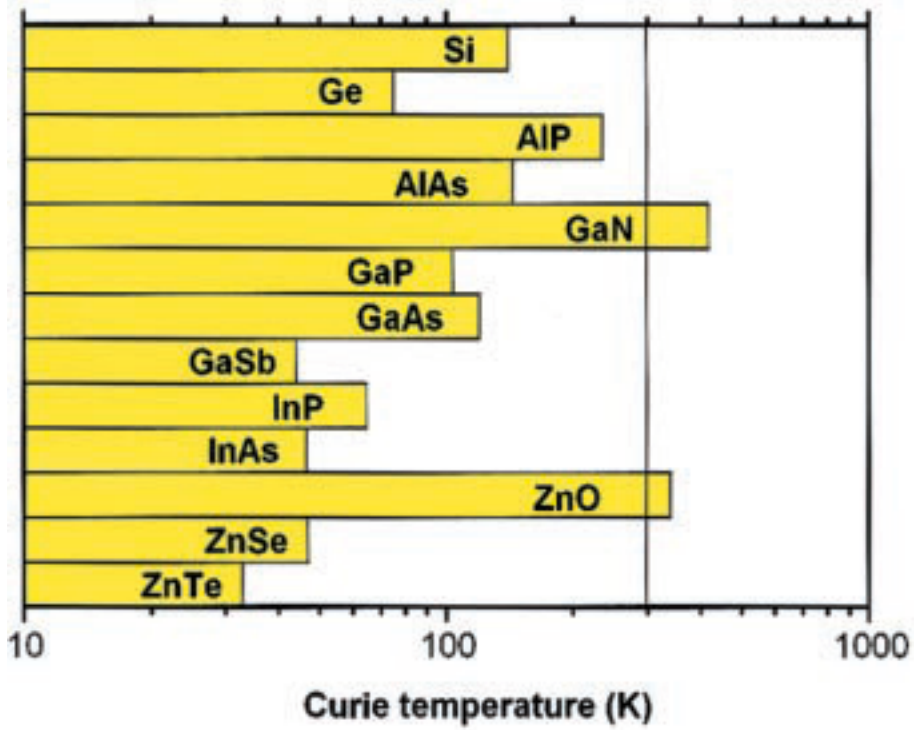
Problem: Low Mn solubility in III-Vs

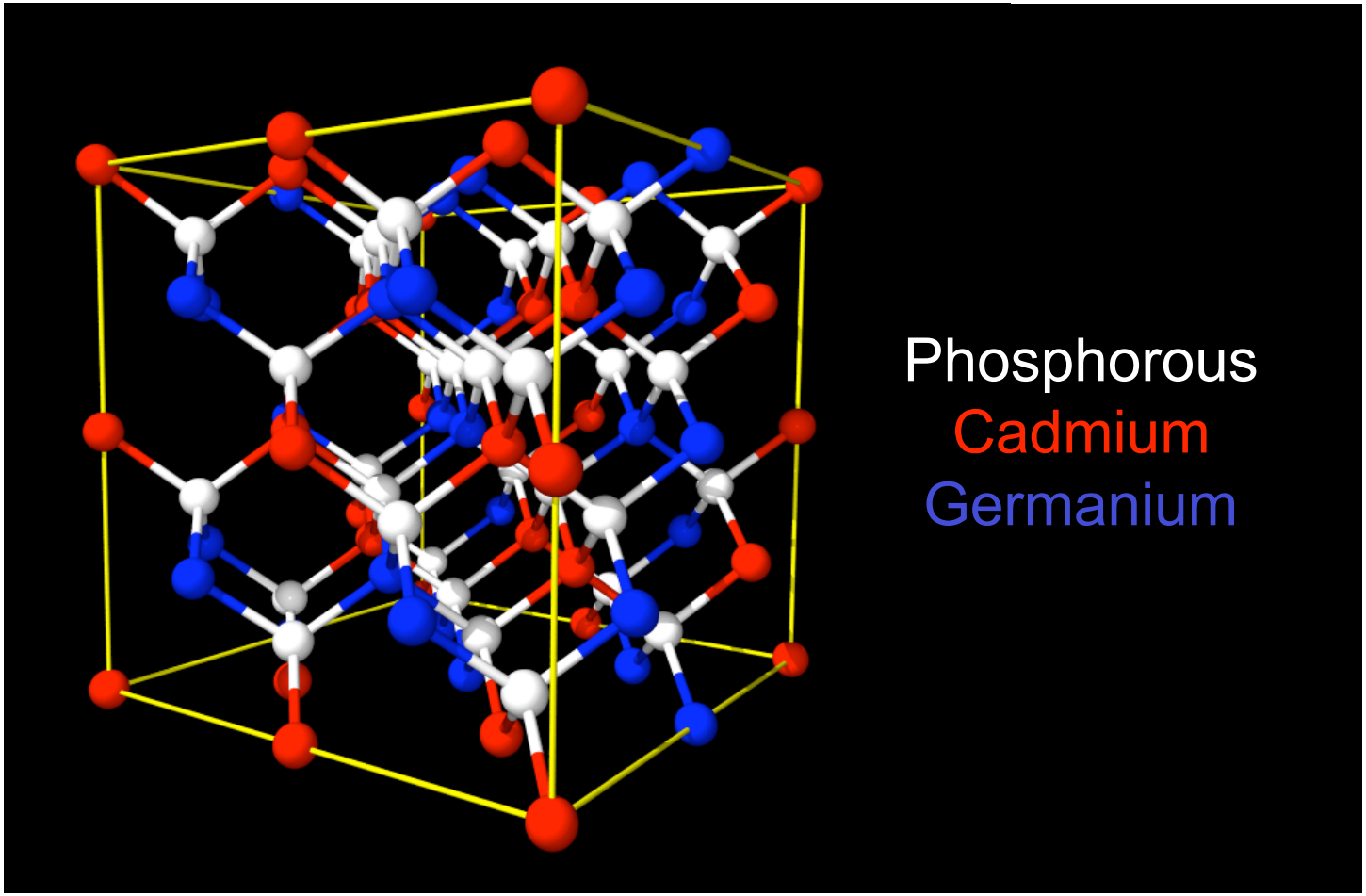


T. Dietl Science **287** 1019 (2000)
5% Mn 3.5×10^{20} holes cm^{-3}

Smaller lattice constants, higher T_c

- Low solubility; Mn^{2+} poorly suited to group III site in III-Vs
- Mn introduces holes and spins in III-Vs
- Look for III-V compatible materials with higher Mn solubility

II-IV-V_2 chalcopyrites (e.g. CdGeP2)



II-IV-V_2 Structure: III-V Zinc-Blende structure "on average"
Motivation: Mn^{2+} readily substitutes on group II site

Experimental situation

High T_cs have been observed, but the location, distribution of Mn, and nature of magnetism is uncertain

$Cd_{1-x}Mn_xGeP_2$: T_c>RT x=variable Medvedkin JJAP **39** 949 (2000)

$Zn_{1-x}Mn_xGeP_2$: T_c~300K x=0.2 Medvedkin JCG **236** 609 (2002)

$Zn_{1-x}Mn_xGeSiN_2$: T_c>200K x~0.05 Pearton JAP **92** 2047 (2002)

$Zn_{1-x}Mn_xSnAs_2$: T_c=329K x=0.01 Choi JJAP **39** 949 (2000)

Why are chalcopyrites ferromagnetic?

Medvedkin proposed: Hole-creating defects result in ferromagnetism (Medvedkin JJAP **39** L949 (2000))

Theory:

Antiferromagnetism (AFM) dominates in bulk, like II-VIs. Dopable (Zhao & Freeman PRB **65** 094415 (2002))

Carriers promote ferromagnetism(FM); anti-site defects are energetically prefered under certain growth conditions (Mahadevan & Zunger PRL **88** 047205 (2002))

Question: Which chalcopyrites have high T_c?

Exchange interaction calculations

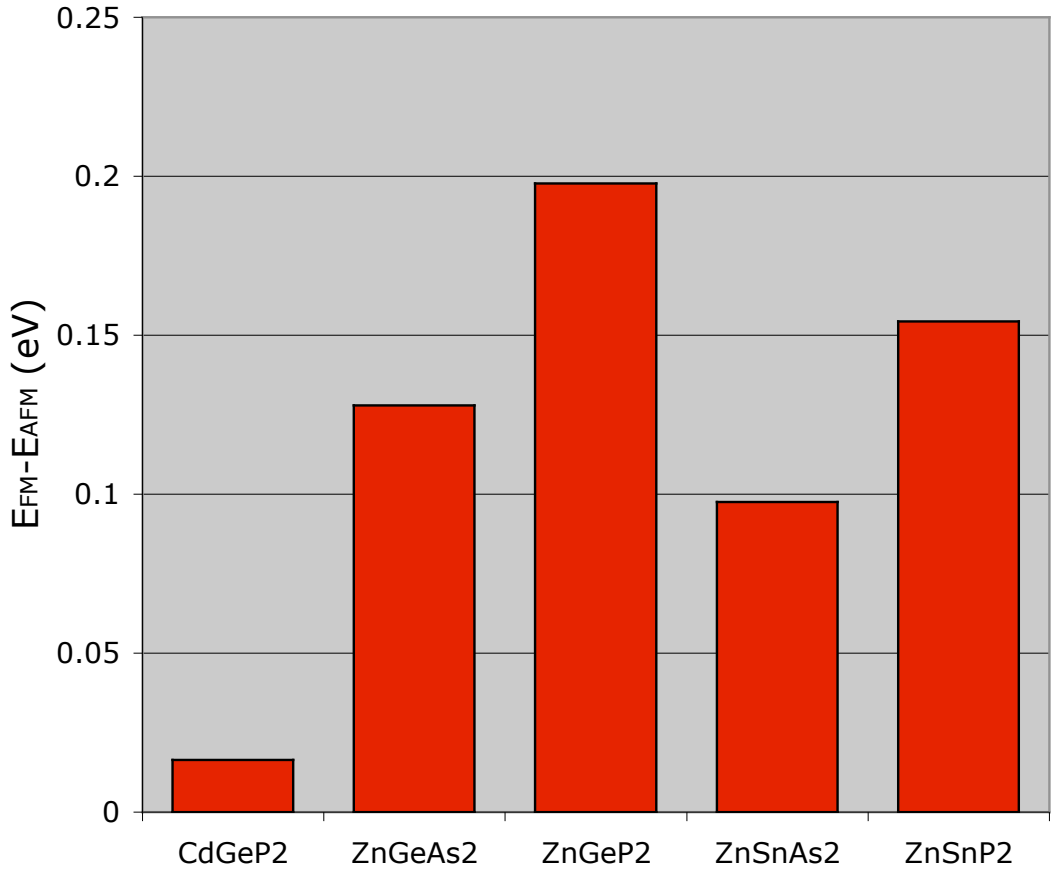
Guide to T_c: Exchange interaction between Mn pairs calculated via energy difference in FM and AFM configurations.

Three Mn pairs calculated:

1. II-II e.g. Mn on nearest-neighbor Zn sites
2. II-IV e.g. Mn on nearest-neighbor Zn/Ge sites
3. IV-IV e.g. Mn on nearest-neighbor Ge sites

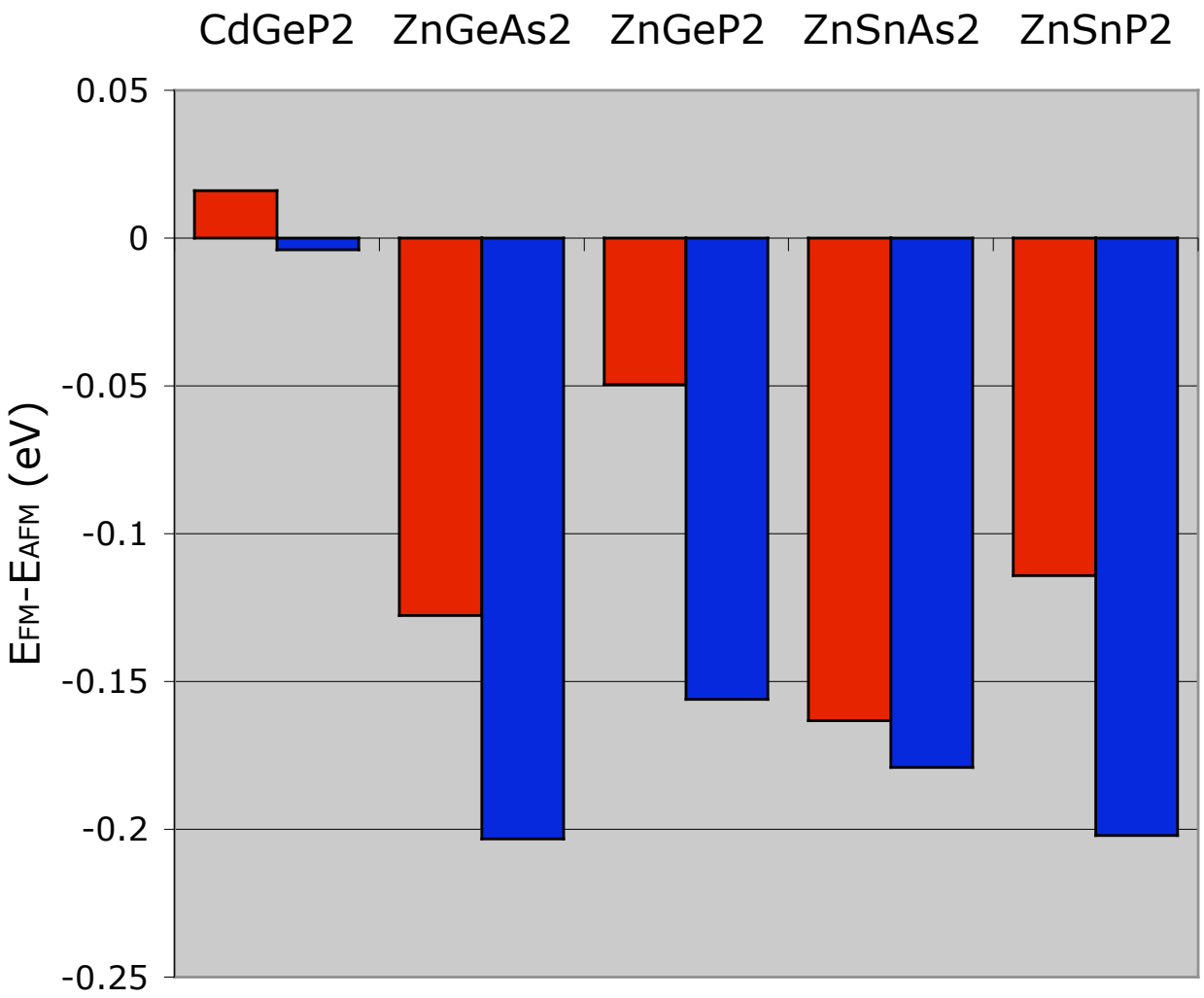
Density functional calculations (PW91 GGA, PAW)
64 atom supercells. d electrons in valence.

Antiferromagnetic interactions between II-II pairs



Antiferromagnetism dominates
Also confirmed for 25% Mn [001] superlattices

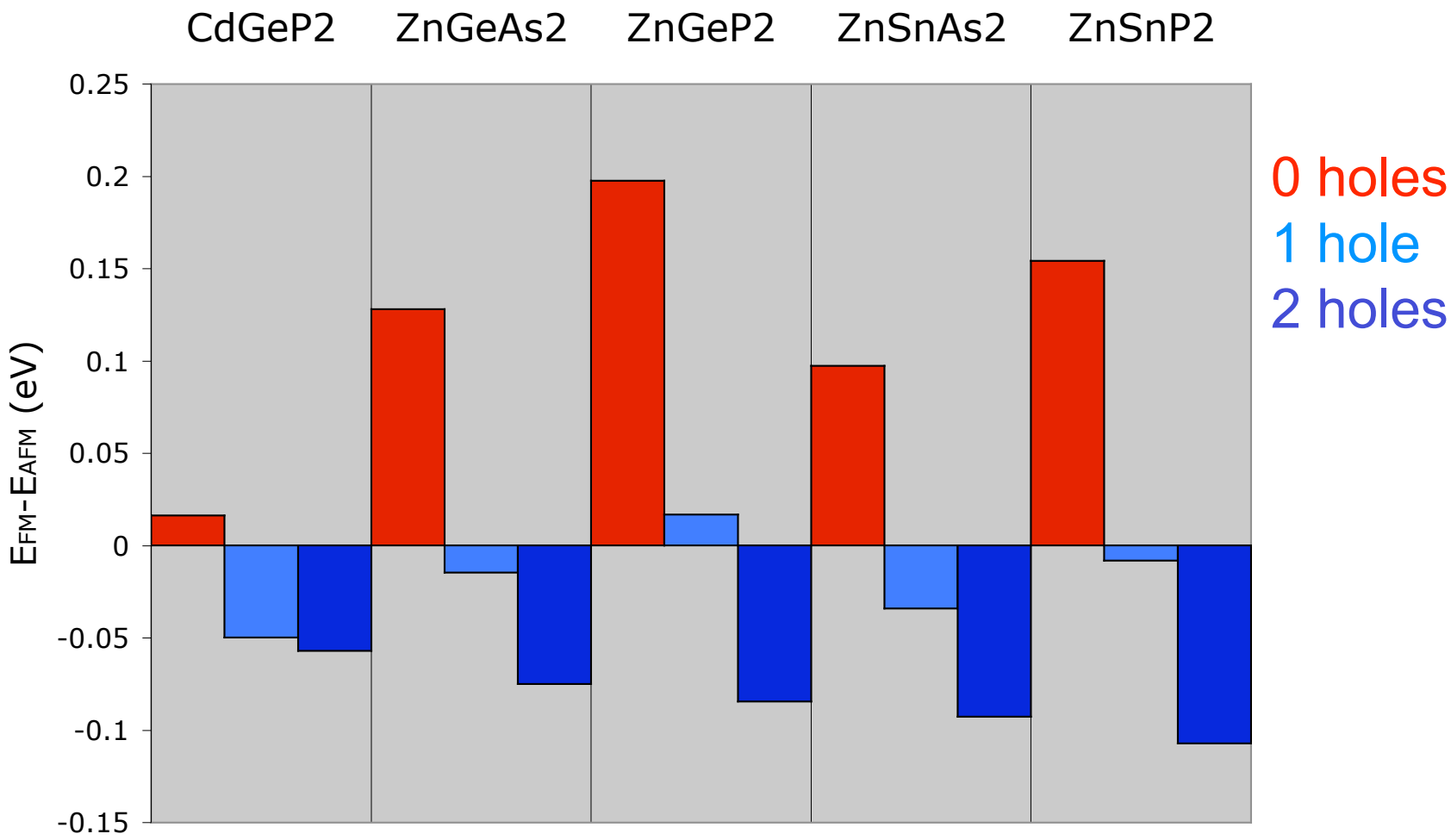
Ferromagnetic interaction between defects



II-IV defects
IV-IV defects

Predominant ferromagnetic interaction between “anti-site” defects. Largest in Zn-compounds.

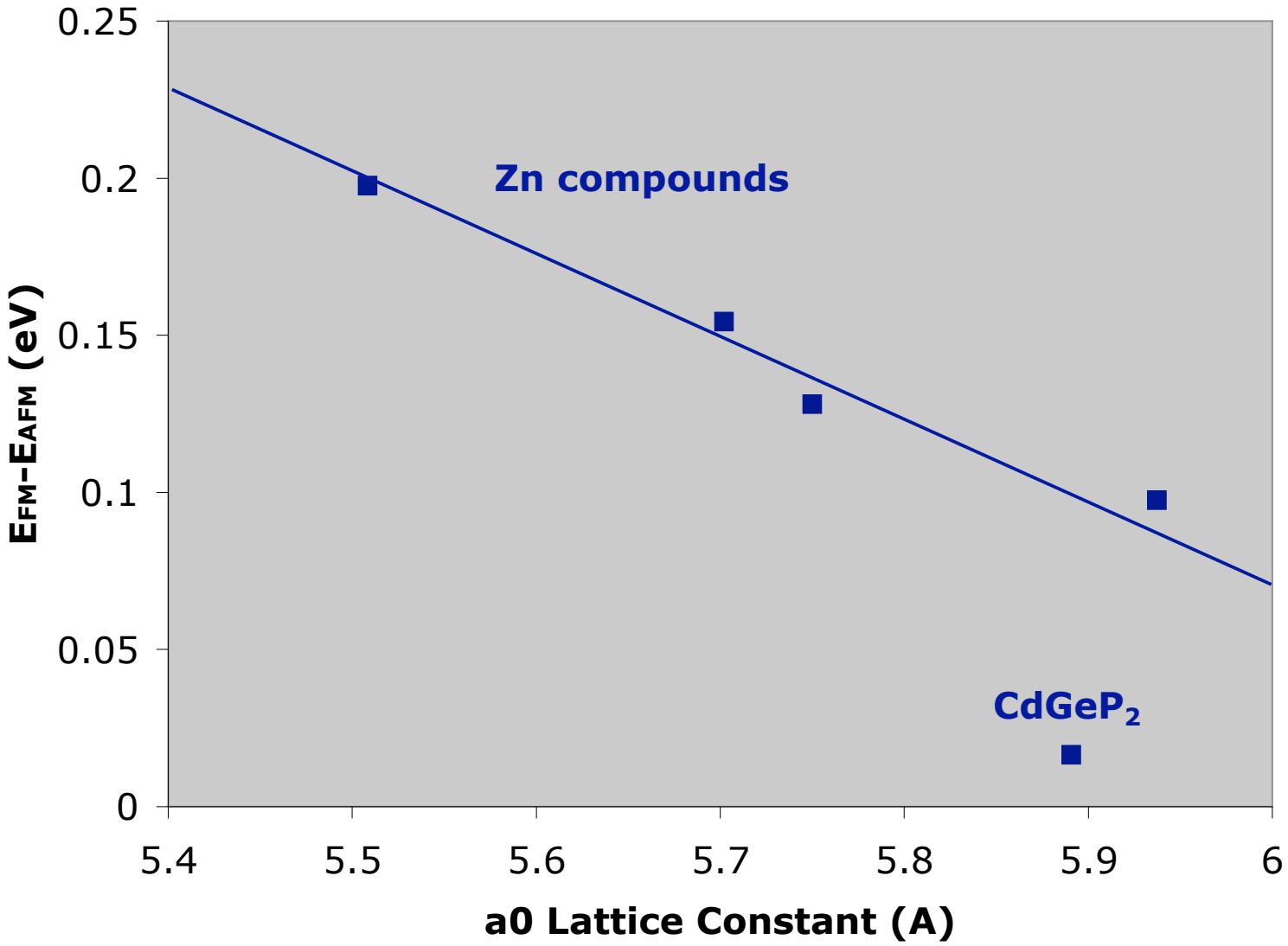
Hole doping promotes ferromagnetic interaction between II-II pairs



Strong AFM preferences overcome by hole doping

J vs lattice constant

No simple relation between lattice constant and ferromagetism



Conclusions

Doping or defects promote ferromagnetism

J (hence T_c) higher in Zn compounds than prototypical $CdGeP_2$

No simple relation between T_c (or J) and lattice constant,
as per simple models.

Many possibilities for influencing T_c (elements, doping, growth),
lattice matching to common substrates

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