

Carrier induced ferromagnetism in II-IV-V₂ Mn-doped chalcopyrites

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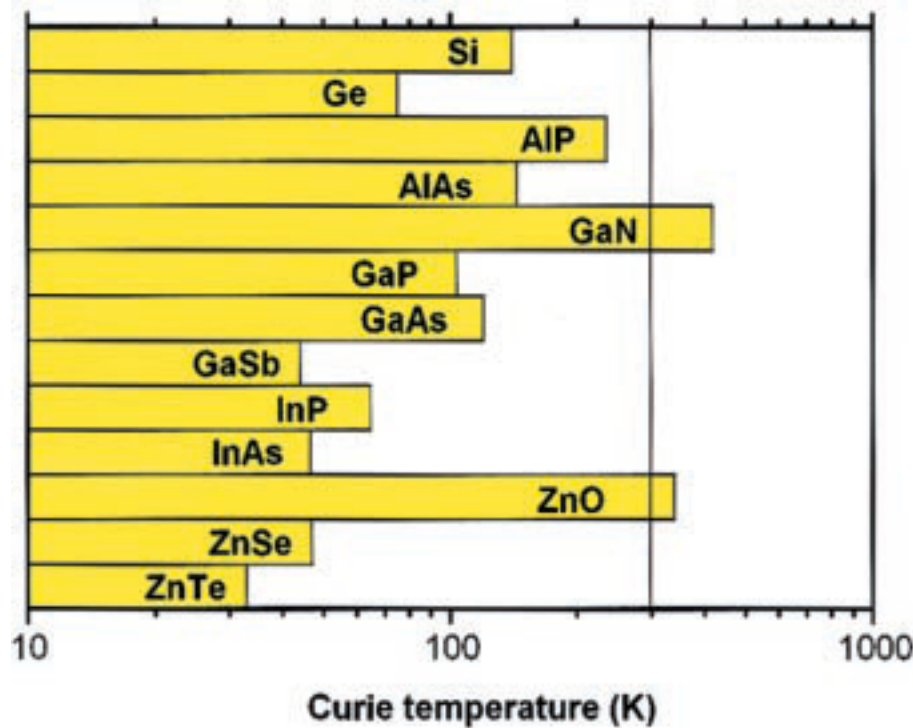
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Problem: Low Mn solubility in III-Vs



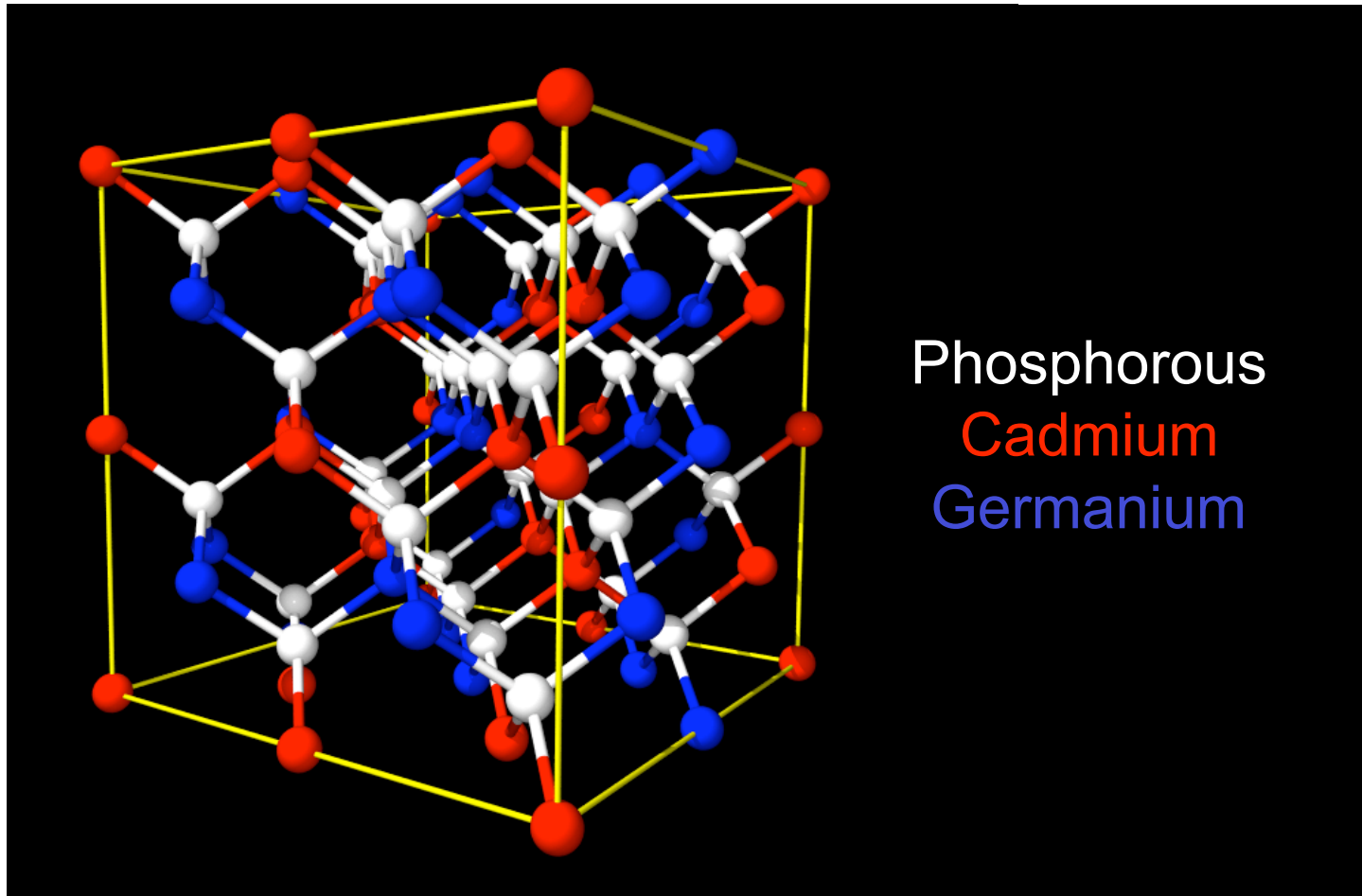
T. Dietl Science **287** 1019 (2000)

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-V₂ chalcopyrites (e.g. CdGeP₂)



II-IV-V₂ Structure: III-V Zinc-Blende structure “on average”

Motivation: Mn²⁺ readily substitutes on group II site

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 preferred under certain growth conditions (Mahadevan & Zunger PRL **88** 047205 (2002))

Experimental situation

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

$\text{Cd}_{1-x}\text{Mn}_x\text{GeP}_2$: $T_c > RT$ $x = \text{variable}$ Medvedkin JJAP **39** 949 (2000)

$\text{Zn}_{1-x}\text{Mn}_x\text{GeP}_2$: $T_c \sim 300\text{K}$ $x = 0.2$ Medvedkin JCG **236** 609 (2002)

$\text{Zn}_{1-x}\text{Mn}_x\text{GeSiN}_2$: $T_c > 200\text{K}$ $x \sim 0.05$ Pearton JAP **92** 2047 (2002)

$\text{Zn}_{1-x}\text{Mn}_x\text{SnAs}_2$: $T_c = 329\text{K}$ $x = 0.01$ Choi JJAP **39** 949 (2000)

Question: Which chalcopyrites have high T_c ?

Exchange interaction calculations

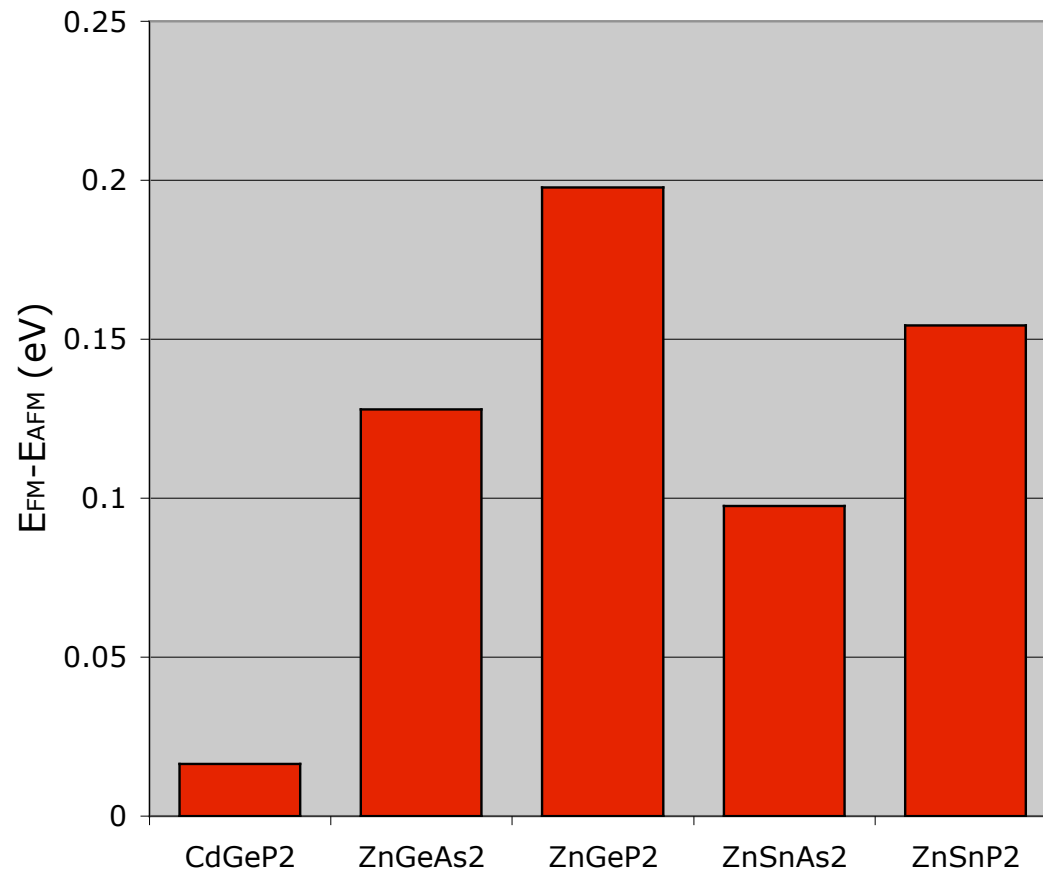
Guide to T_c : Exchange interaction between Mn pairs calculated via energy difference in FM and AFM configurations.

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

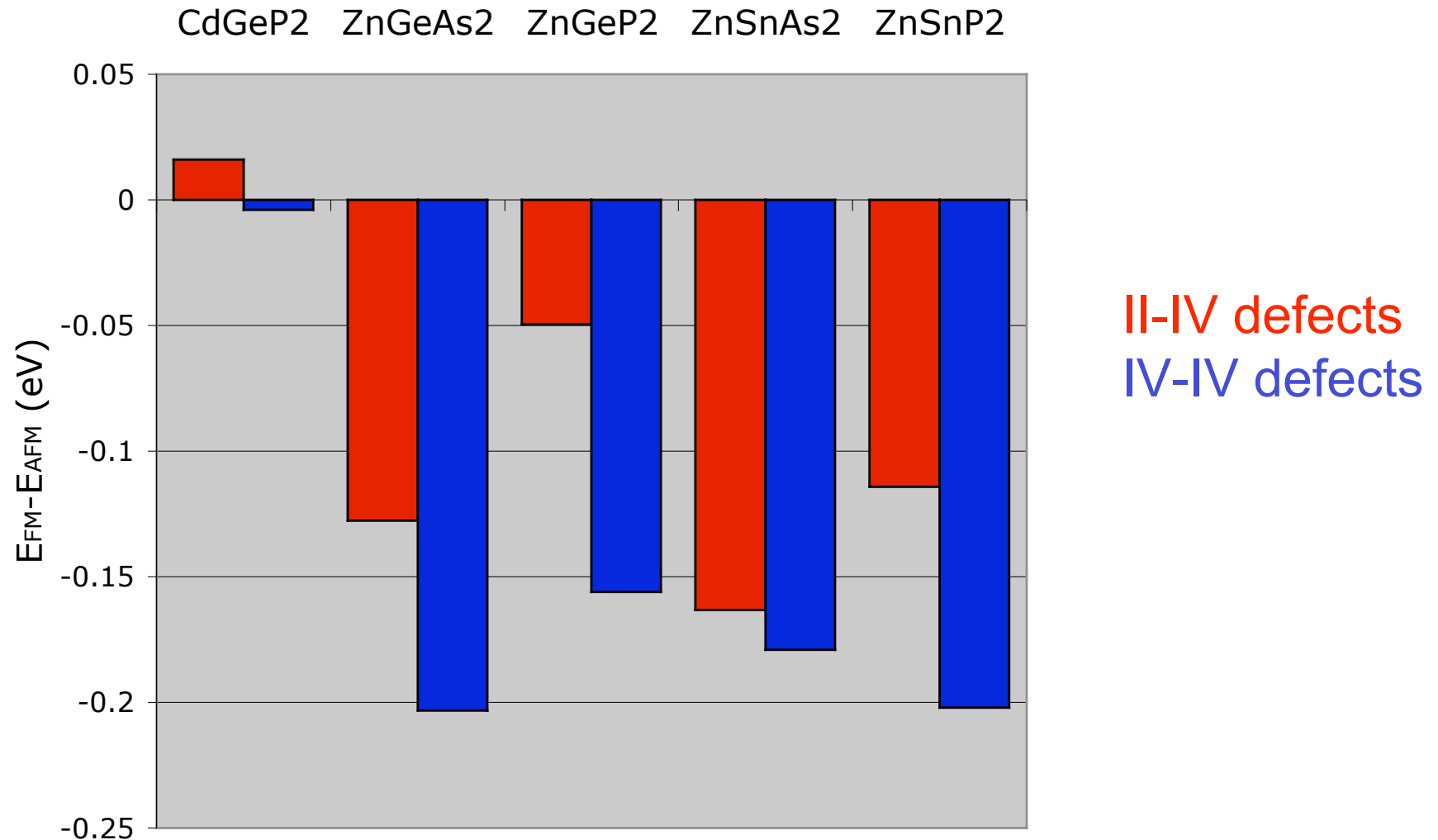
Density functional calculations (PW91 GGA, PAW)
64 atom supercells. d electrons in valence.

Antiferromagnetic interactions between II-II pairs



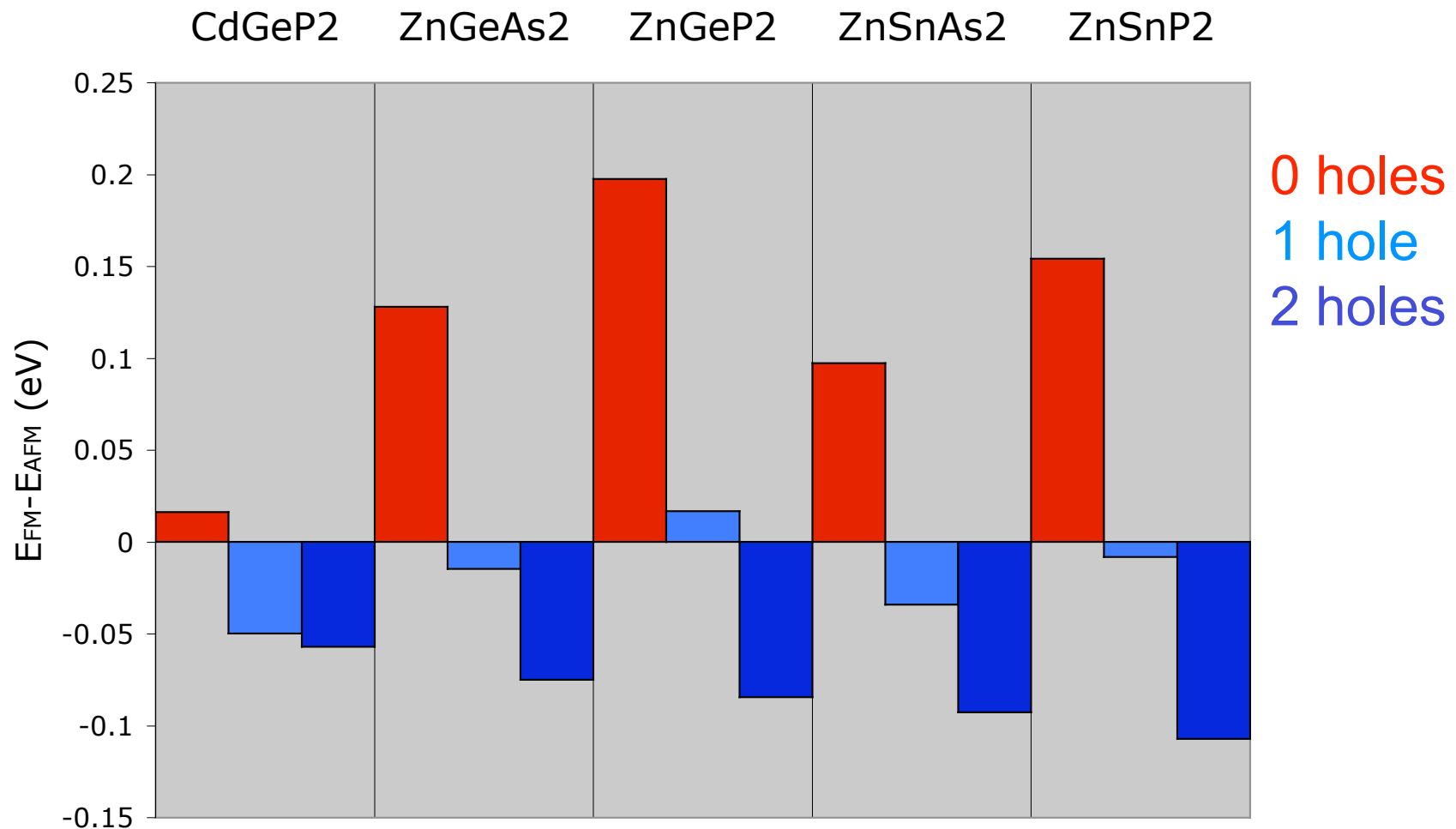
Antiferromagnetism dominates
Also confirmed for 25% Mn [001] superlattices

Ferromagnetic interaction between defects



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

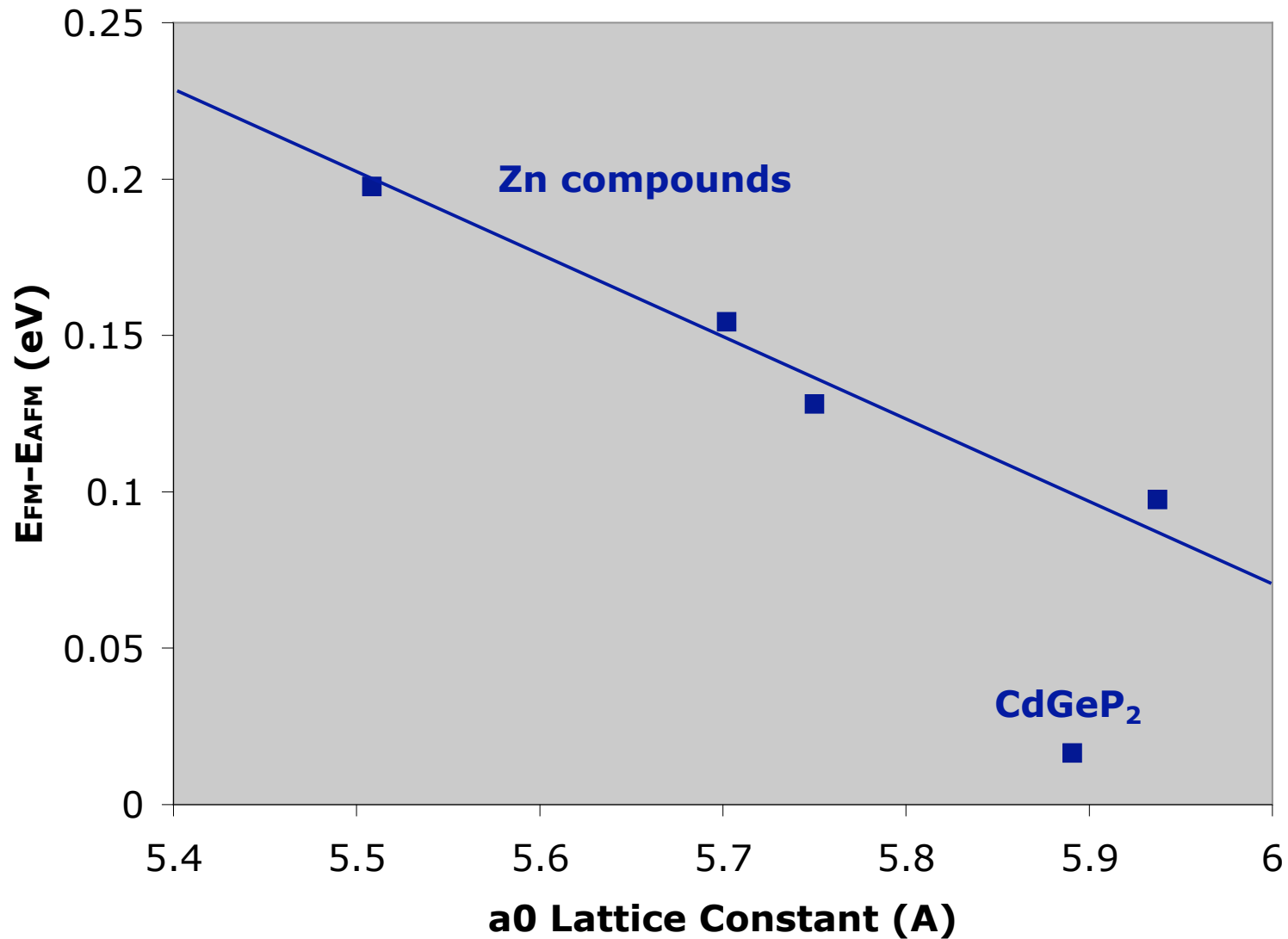
Hole doping promotes ferromagnetic interaction between II-II pairs



Strong AFM preferences overcome by hole doping

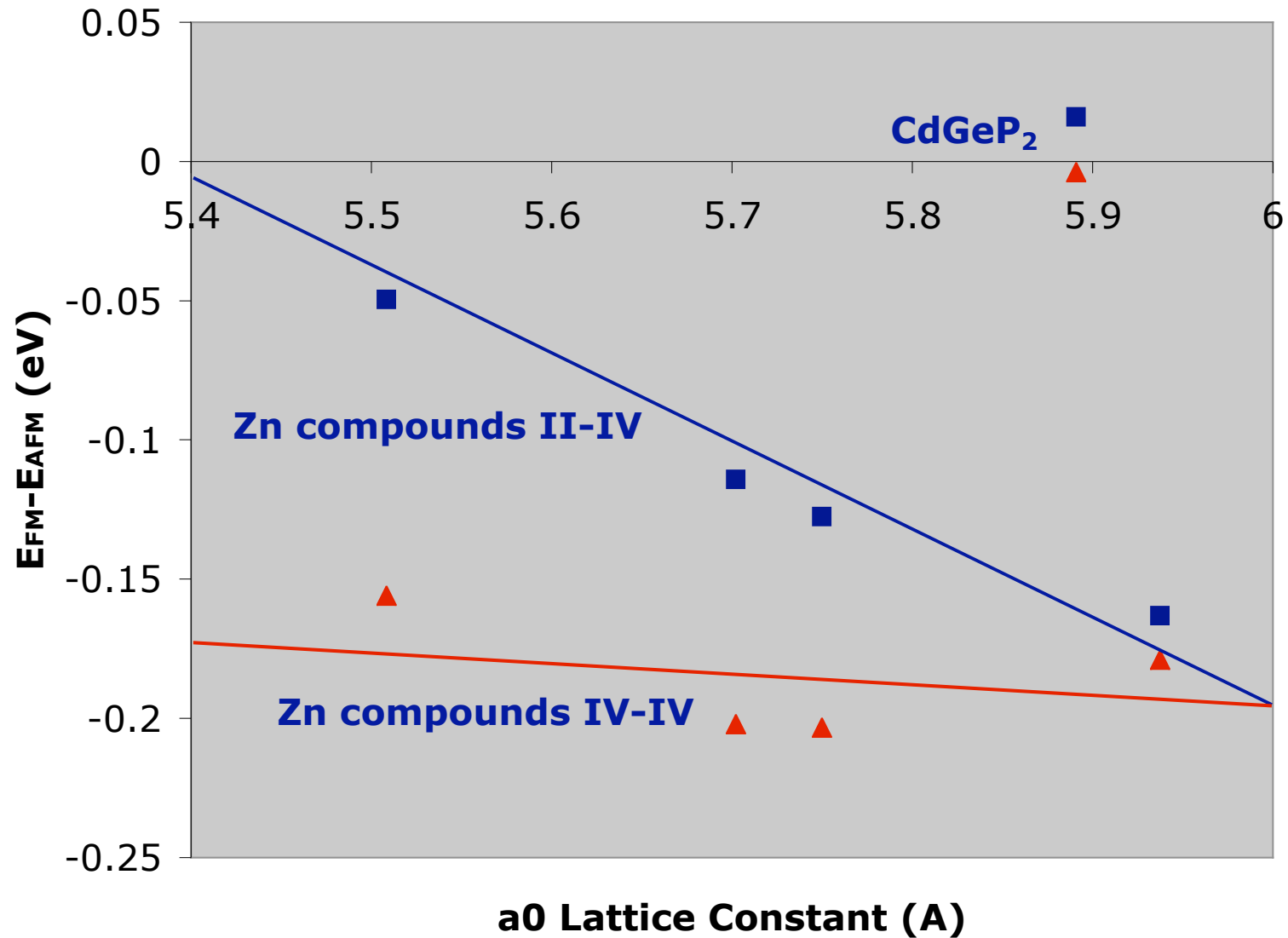
J vs lattice constant

No simple relation between lattice constant and ferromagnetism



J vs lattice constant

No simple relation between lattice constant and ferromagnetism



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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