

Magnetyzm i nadprzewodnictwo w domieszkowanym EuFe₂As₂

Zbigniew Bukowski

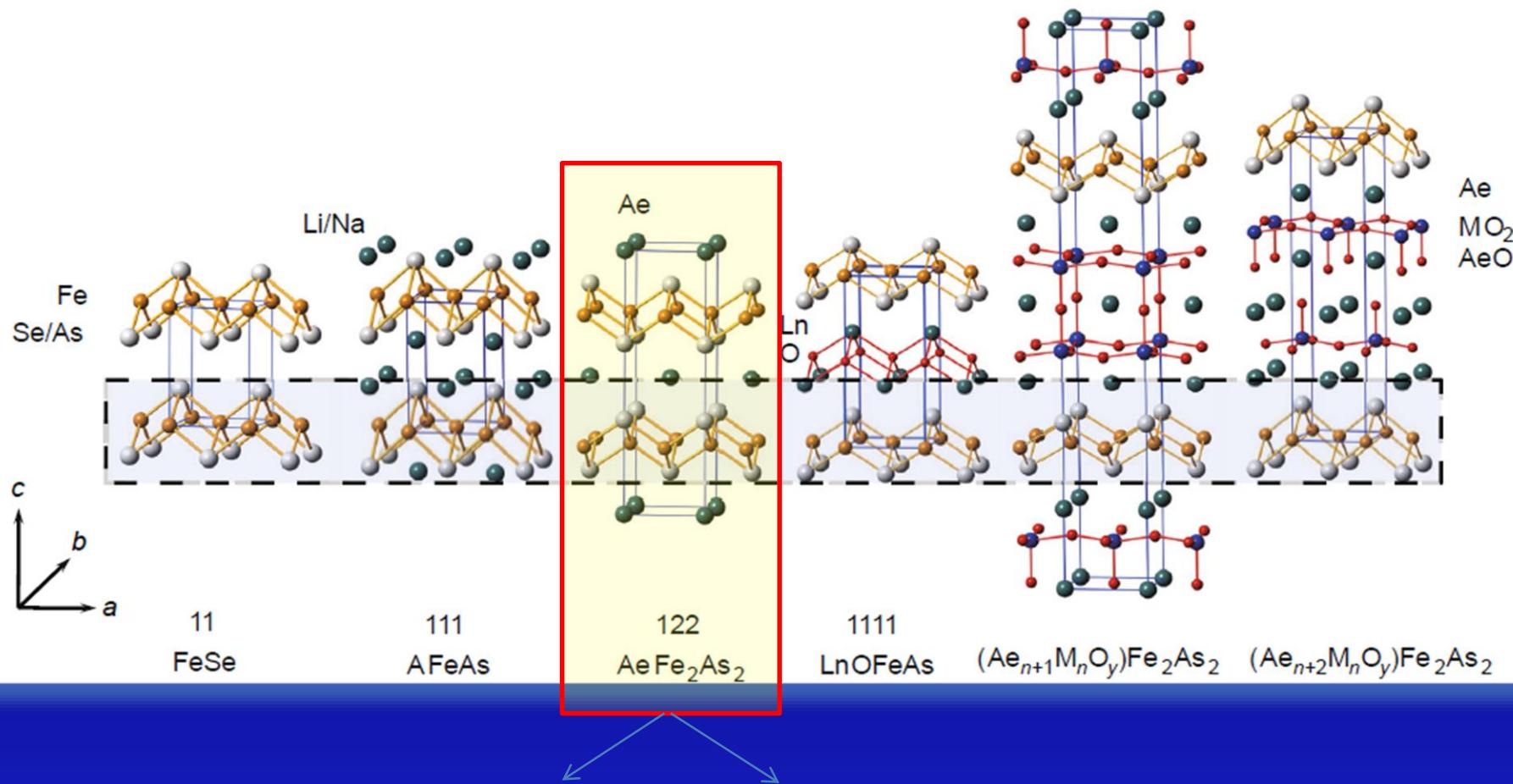
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Wrocław, ul. Okólna 2*



Plan:

1. Podstawowe właściwości EuFe_2As_2
2. Wzrost monokryształów z metalicznych topników
3. Diagram fazowy EuFe_2As_2
 - wpływ pola magnetycznego
 - wpływ ciśnienia
4. Podstawienia chemiczne w EuFe_2As_2
 - domieszkowanie dziurowe – K, Na
 - domieszkowanie elektronowe – La
 - podstawienia izowalencyjne – Ca, Sr, Ba
 - podstawienia izowalencyjne – P
 - podstawienia metalami przejściowymi – Co, Ni, Ir, Ru, Rh...
5. Nadprzewodnictwo i magnetyzm w $\text{EuFe}_{2-x}\text{Ni}_x\text{As}_2$ - wybrane przykłady
6. Spontaniczne worteksy
7. Poszukiwanie nadprzewodnictwa w $\text{EuFe}_{2-x}\text{Ni}_x\text{As}_2$

Crystal structure of iron-based superconductors



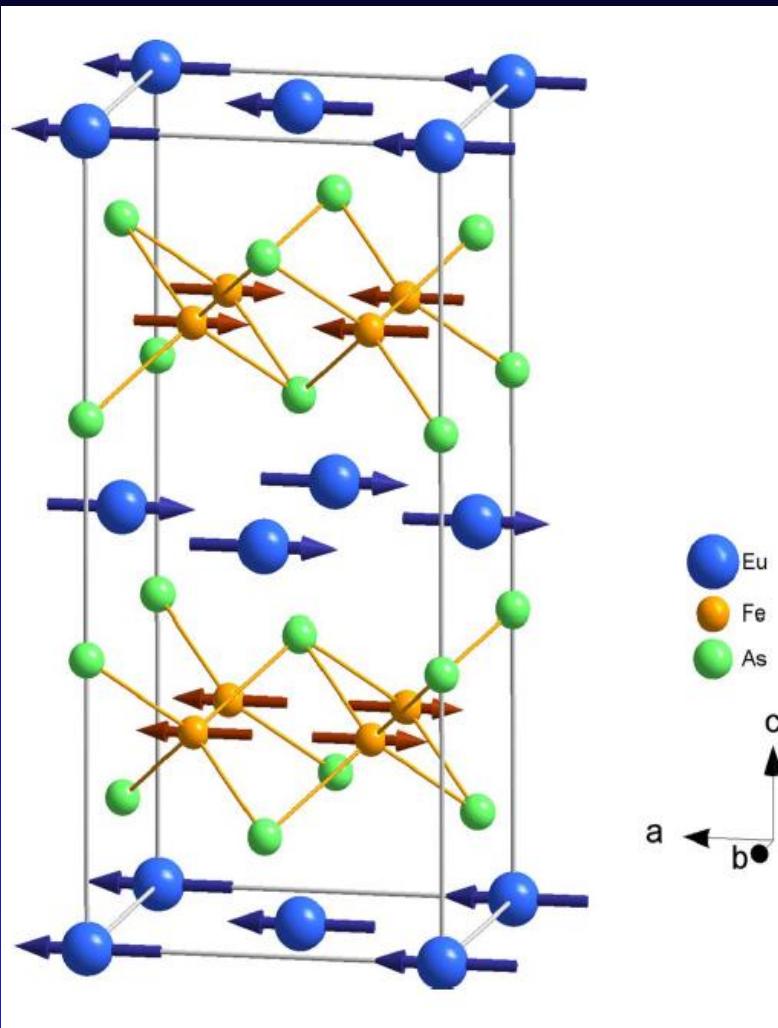
nonsuperconducting
parent compounds

BaFe_2As_2
 SrFe_2As_2
 CaFe_2As_2
 EuFe_2As_2

KFe_2As_2
 RbFe_2As_2
 CsFe_2As_2

low-Tc superconductors

Magnetic structure of EuFe₂As₂



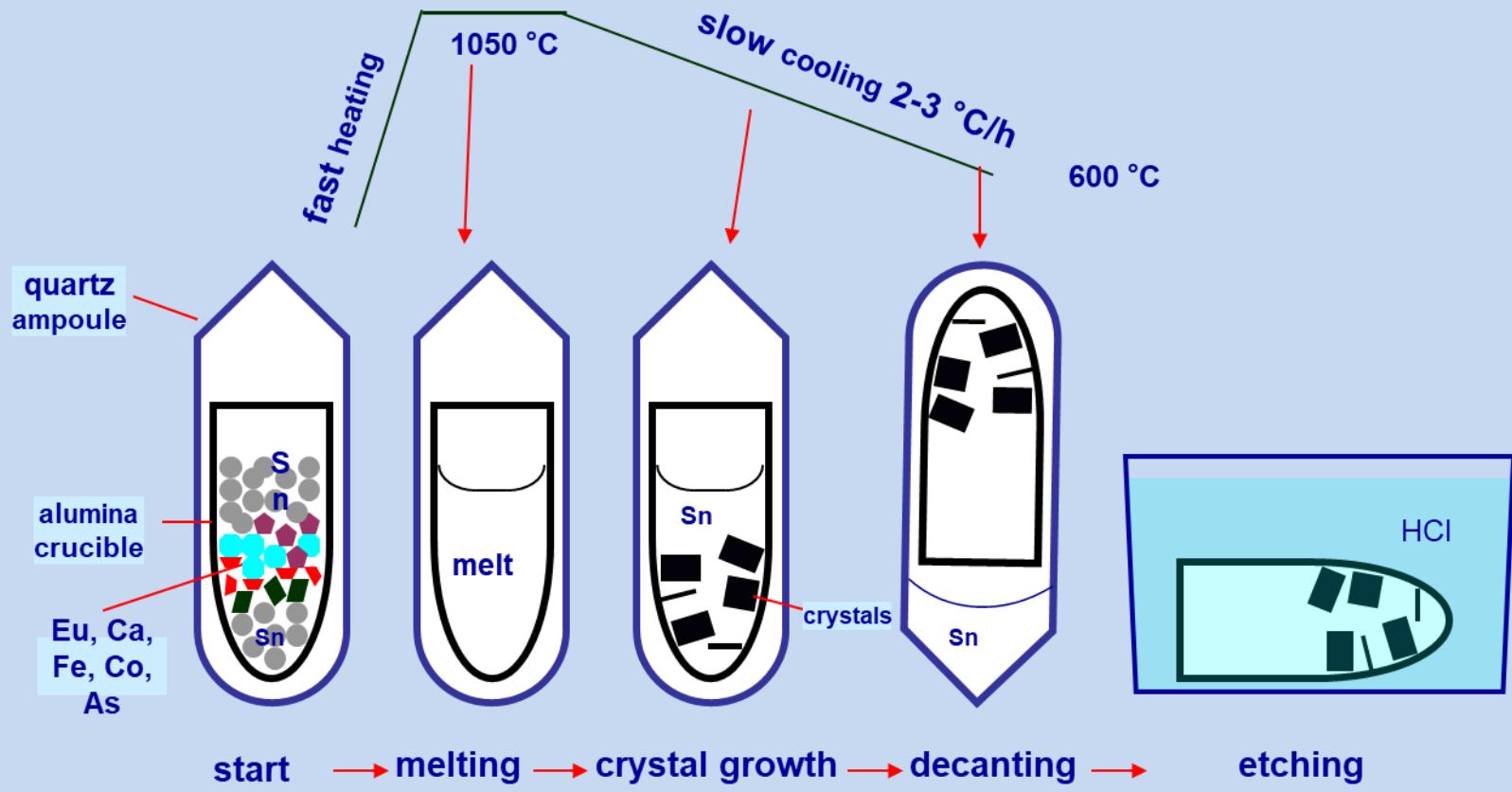
Two magnetic sublattices

Fe²⁺ 3d itinerant electrons
Spin Density Wave
Fe saturation moment of 0.988 μ_B
aligned along the long *a* axis.
 $T_{SDW} = 190$ K

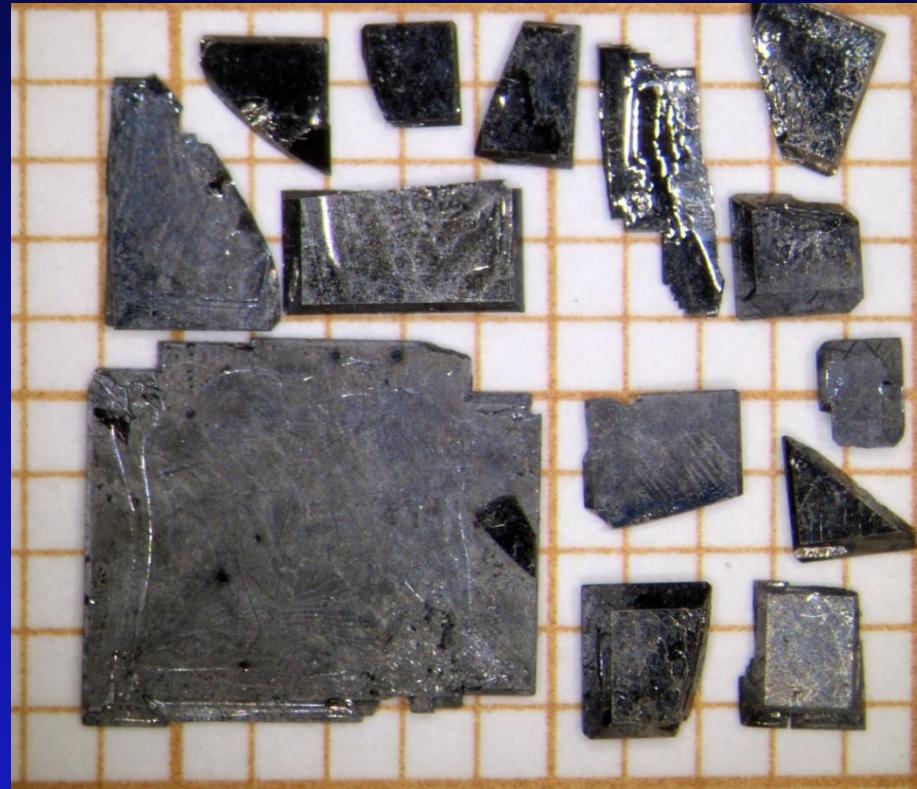
localised Eu²⁺ 4f electrons,
spin $S=7/2$ $\mu_{eff}= 7.94 \mu_B$
RKKY A-type antiferromagnet
 $T_N=19$ K

Growth of single crystals from Sn flux

Single crystals of doped EuFe_2As_2 with a size up to few millimeters were grown from Sn flux. Starting components: Eu,(Ca), Fe,(Co), As, and Sn were placed in alumina crucibles and sealed in silica tubes under reduced pressure of Ar. The ampoules were heated to 1050°C and slowly cooled down to 600°C , when liquid tin was decanted.



Single crystals grown from Sn flux

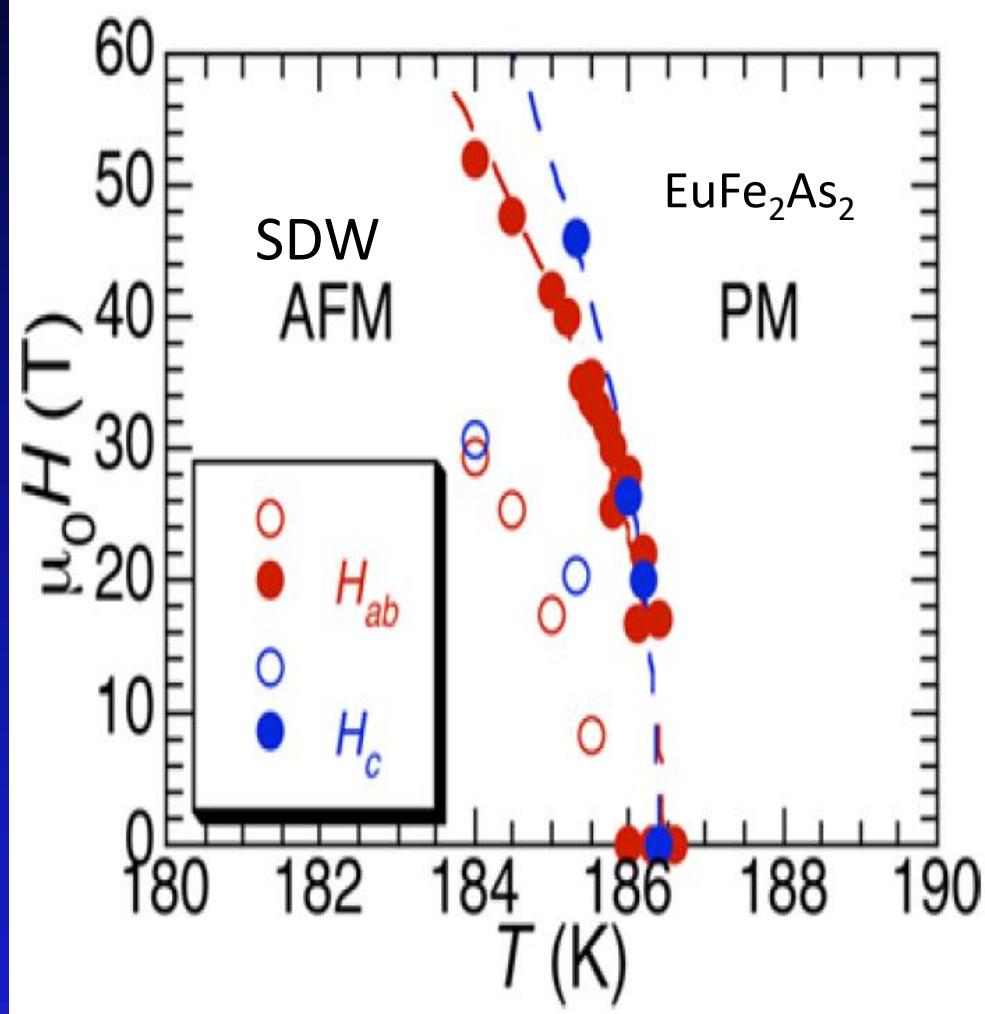


EuFe_2As_2

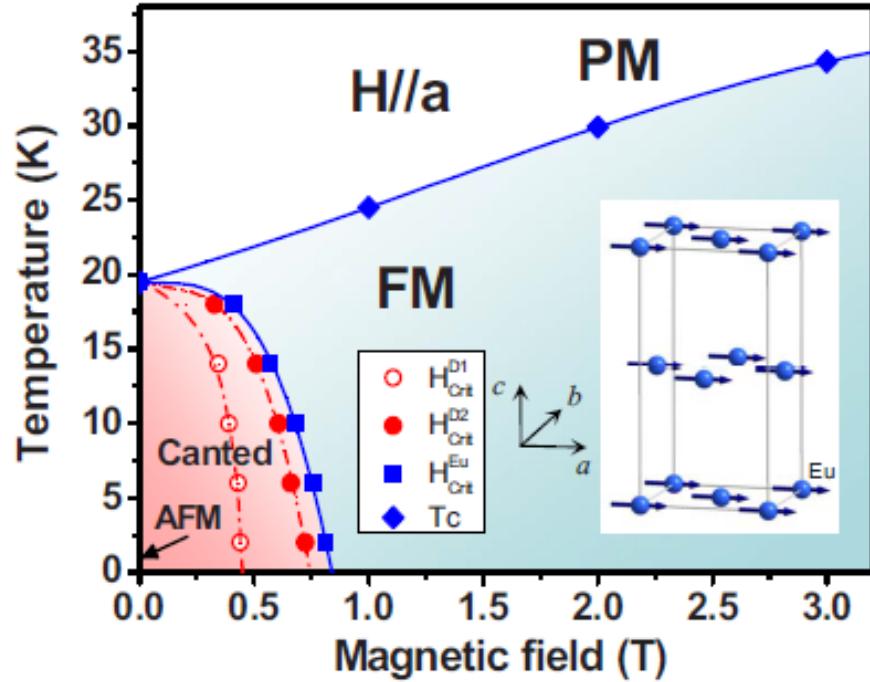
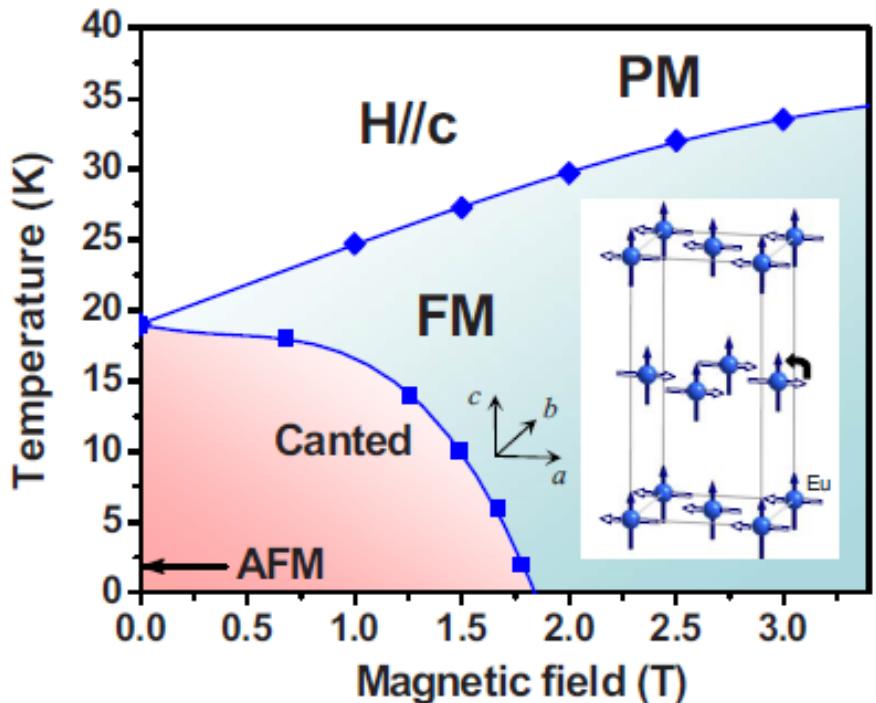
Effect of magnetic field on SDW ordering

Tokunaga et al. J. Low Temp. Phys. 159 (2010) 601

Simple extrapolation suggests that an **extremely high field (>500 T) is needed to suppress the AFM state** at low temperatures.



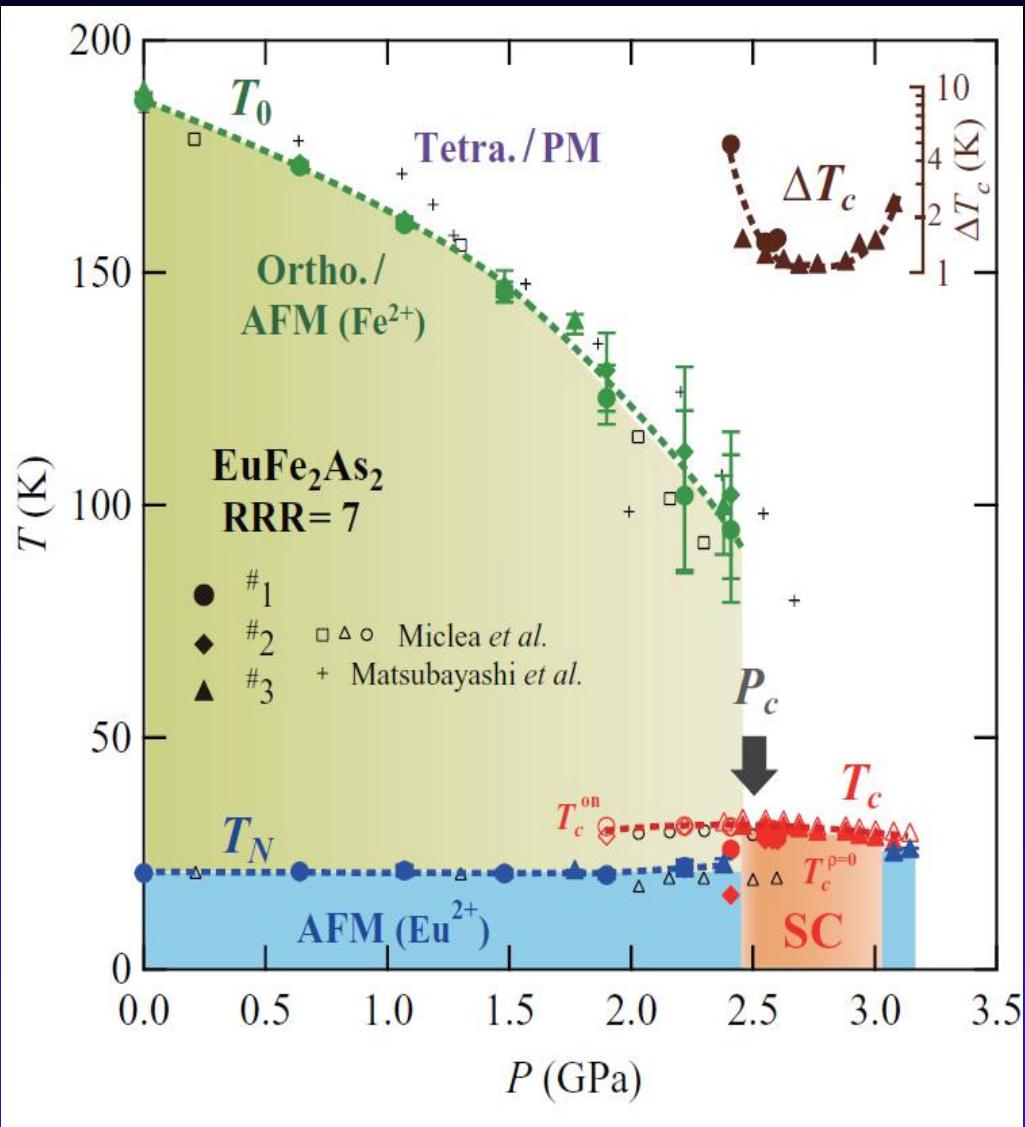
Effect of magnetic field on magnetic order in EuFe_2As_2



- spin canting
- metamagnetic transitions
- field induced ferromagnetism

Xiao et al. PRB 81, 220406R (2010)

Effect of pressure

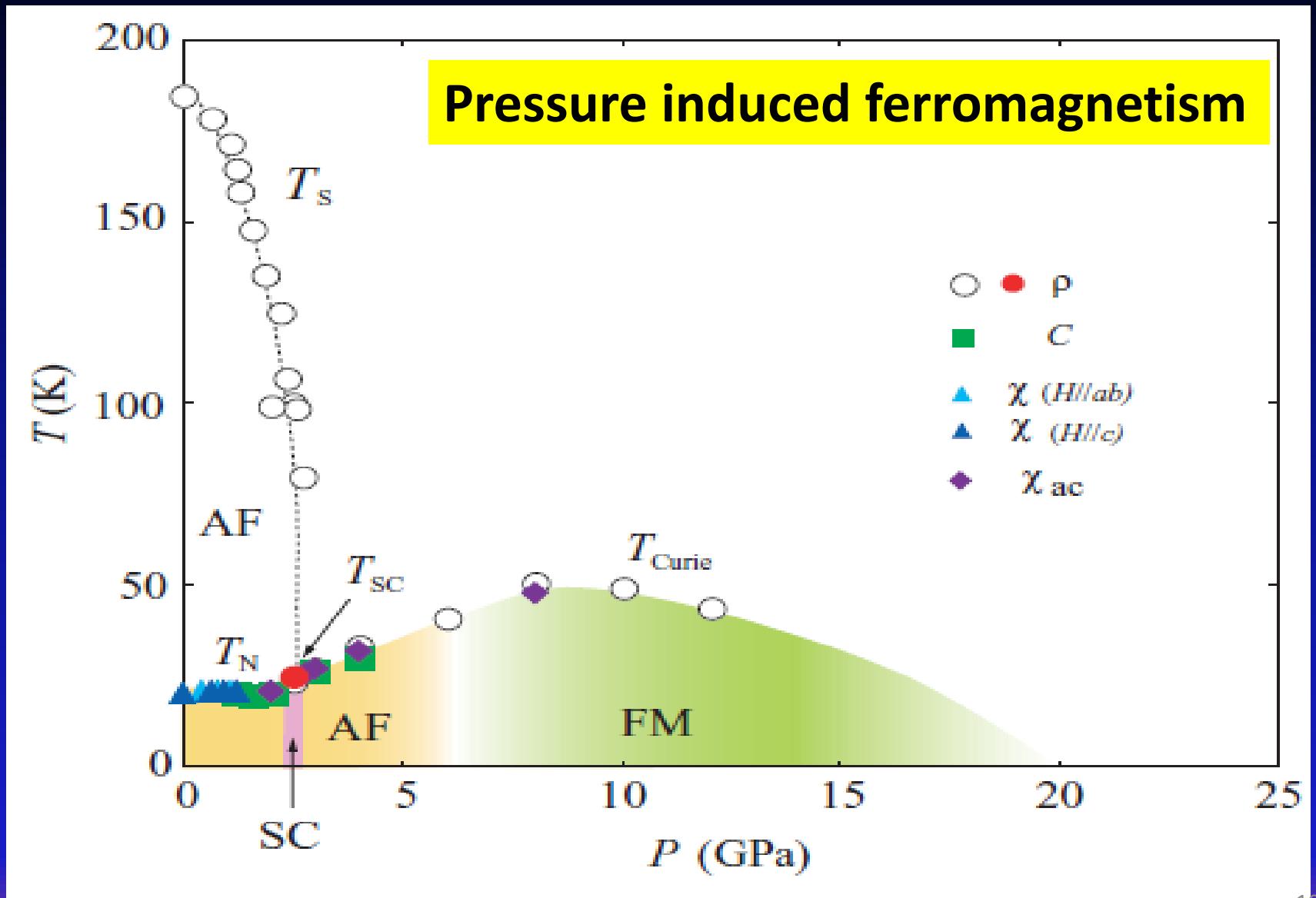


Pressure-suppressed SDW order

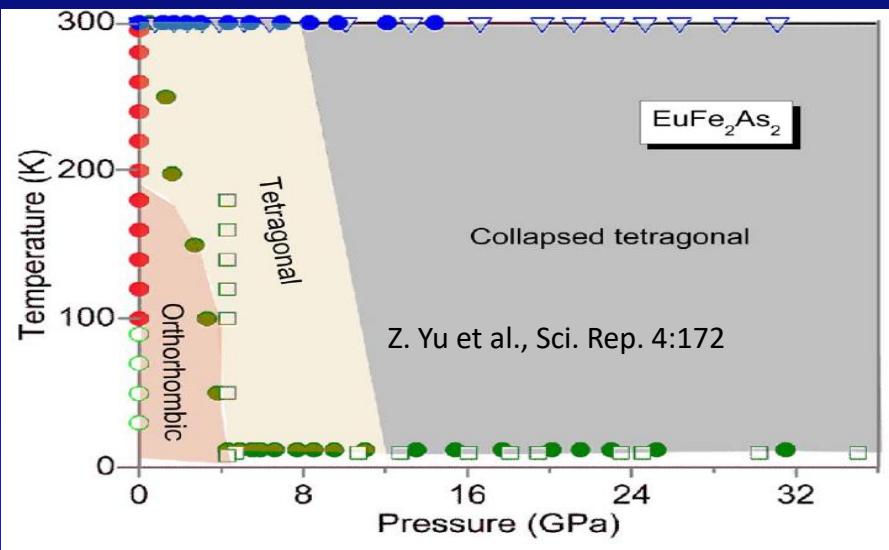
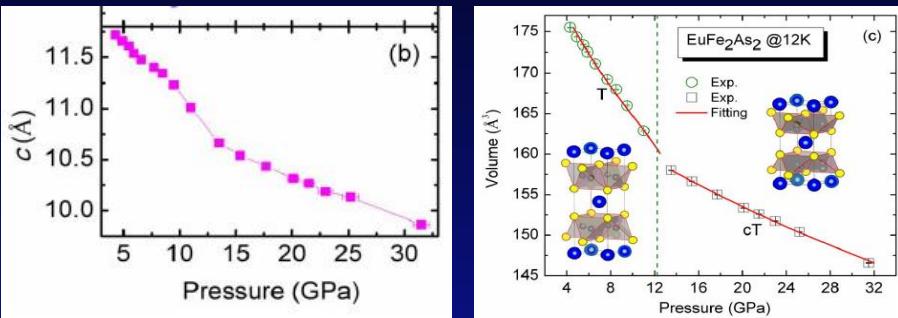
Persistent Eu^{2+} magnetic order

Pressure-induced superconductivity

Effect of pressure

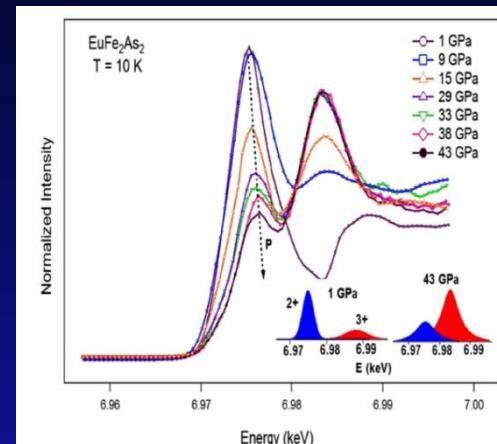


Effect of pressure on EuFe₂As₂ crystal structure



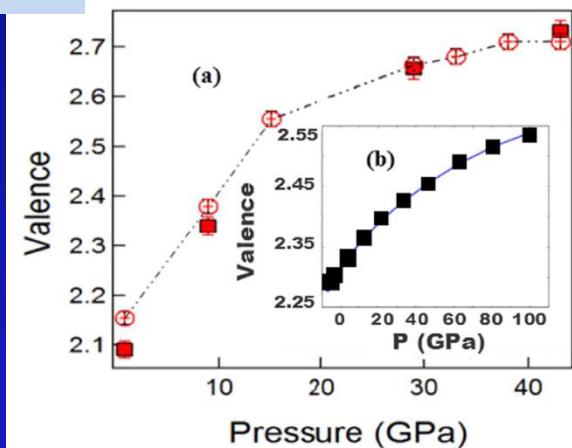
Pressure induced tetragonal- "collapsed tetragonal" phase transition

Effect of pressure on Eu-ion valence in EuFe₂As₂



X-ray absorption spectra

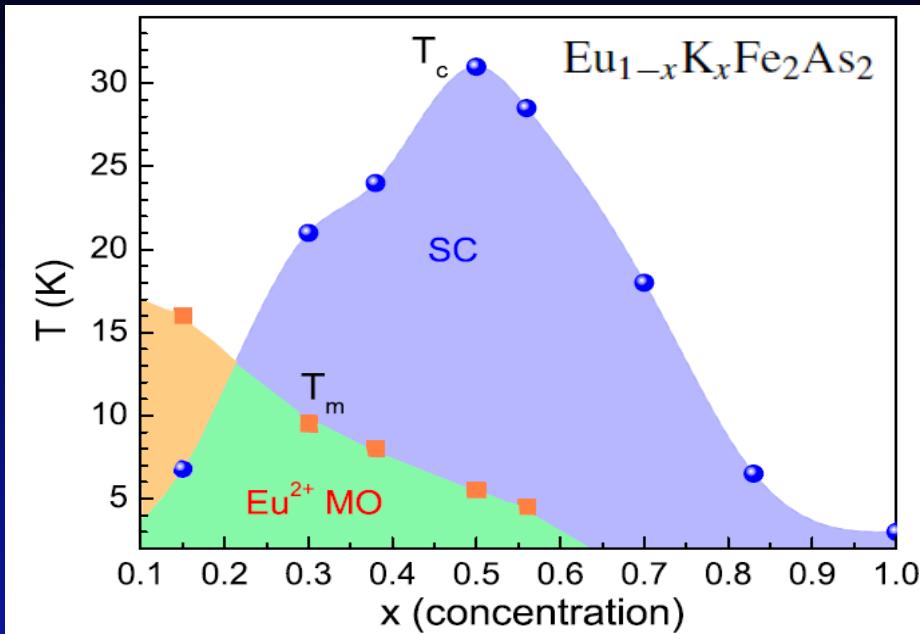
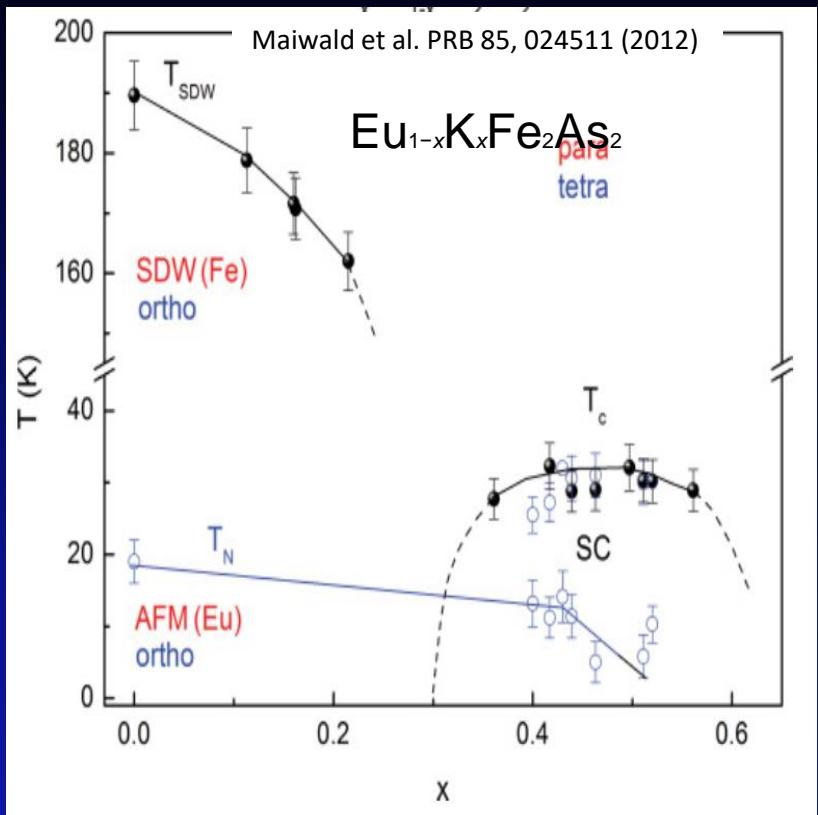
Kumar et al. Appl. Phys. Lett. 104, 042601 (2014)



Conversion of Eu²⁺ to Eu³⁺ under pressure

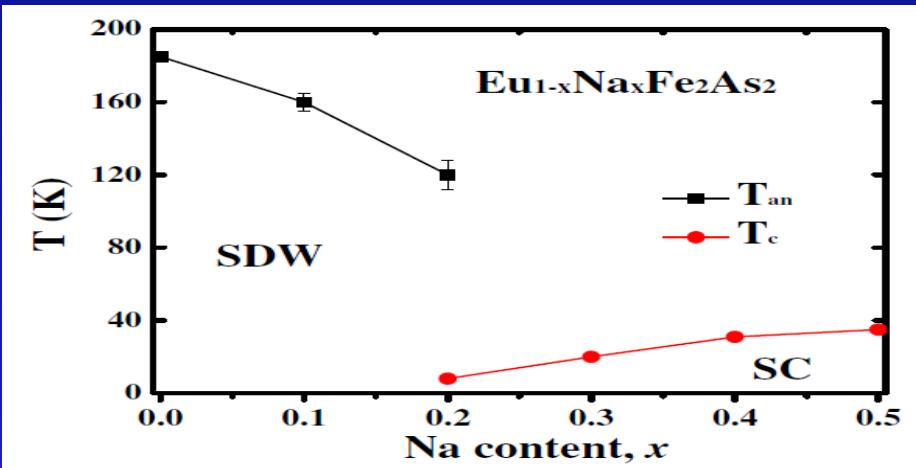
K, Na-substitution

Anupam et al. J. Phys.: Condens. Matter 23 (2011) 455702



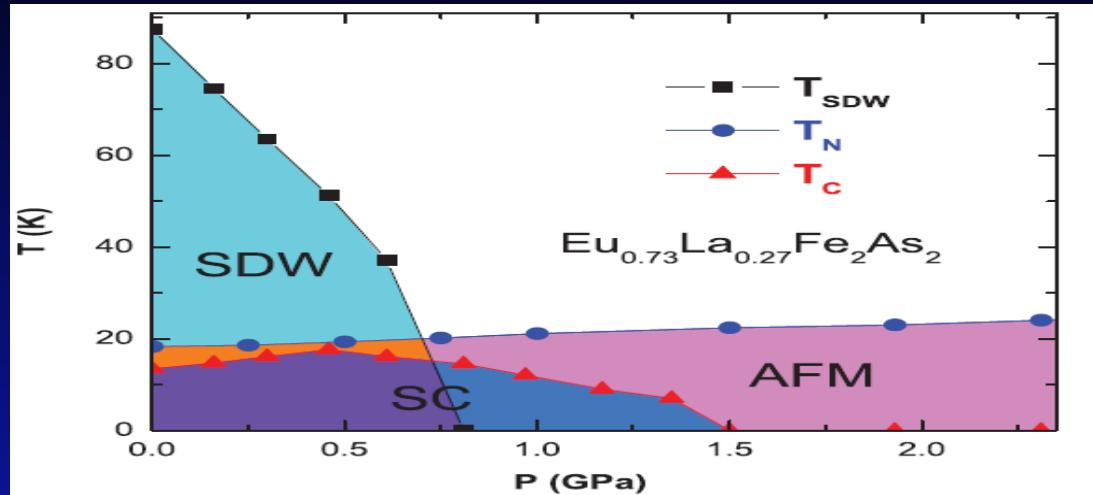
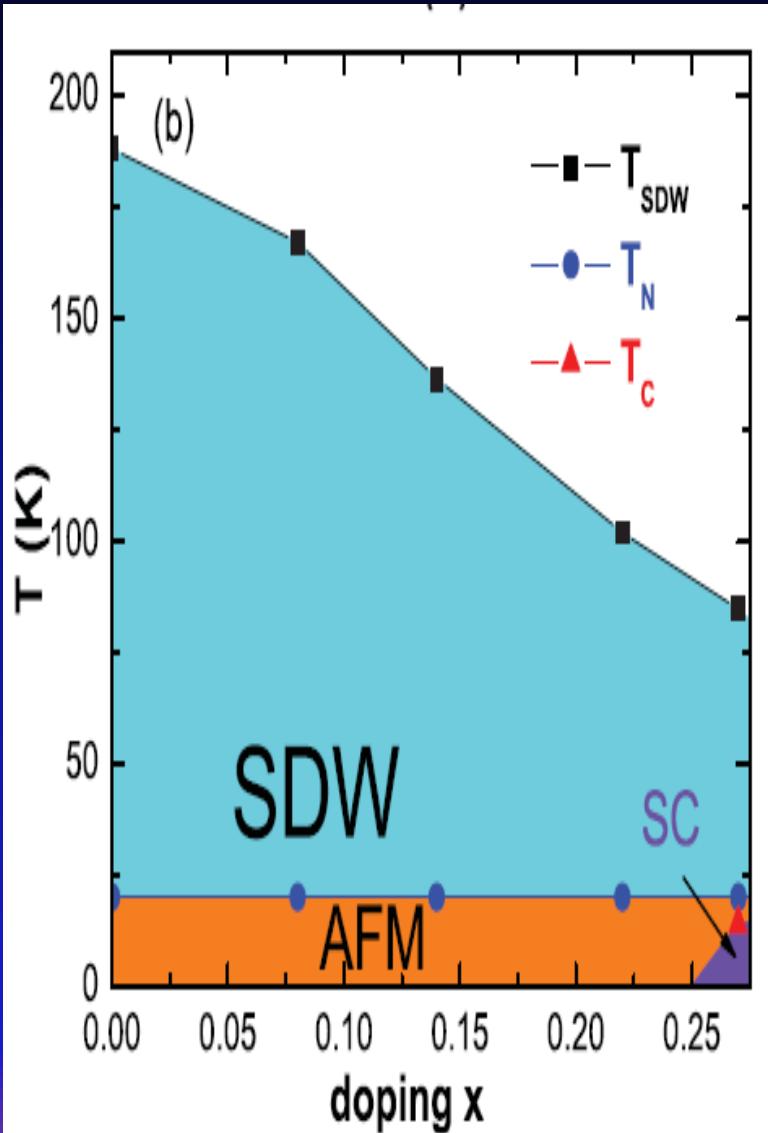
hole doping:

- SDW is suppressed
- Eu²⁺ AF order disappears
- appearance of superconductivity



La-substitution

M. Zhang et al., PRB 85, 092503 (2012)



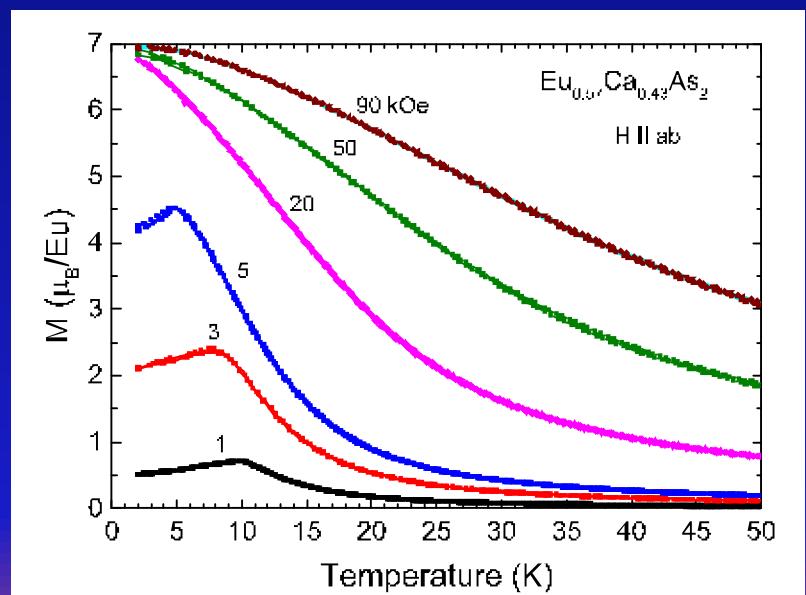
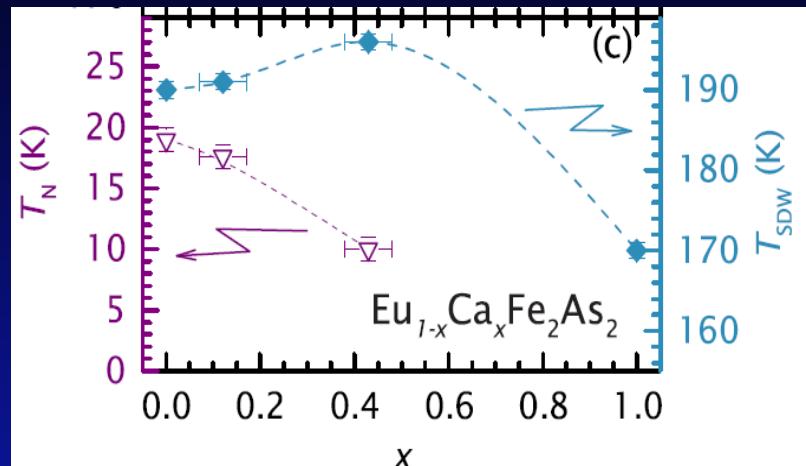
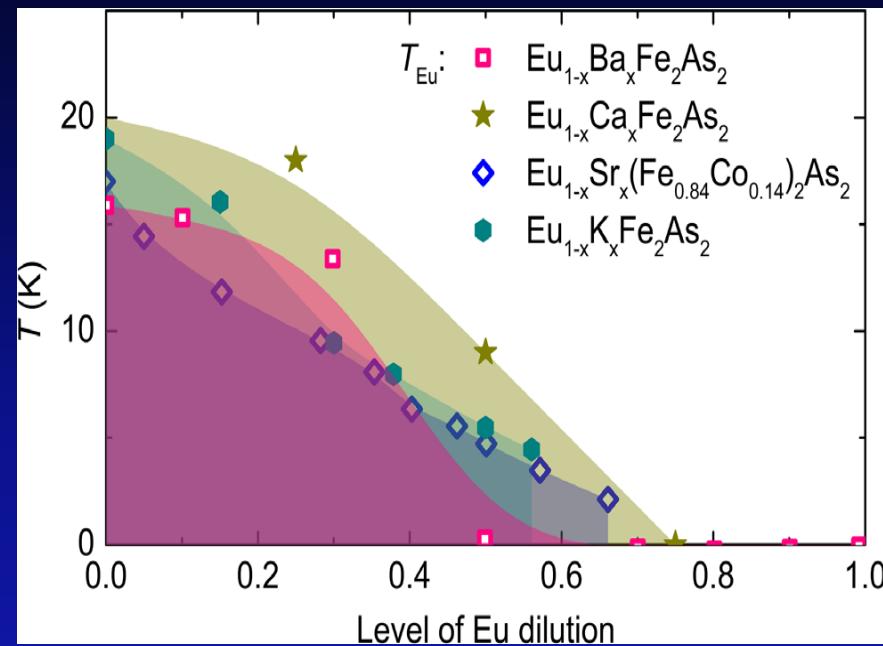
Electron doping:

- SDW suppression
- superconductivity

Dilution of Eu-sublattice with nonmagnetic ions

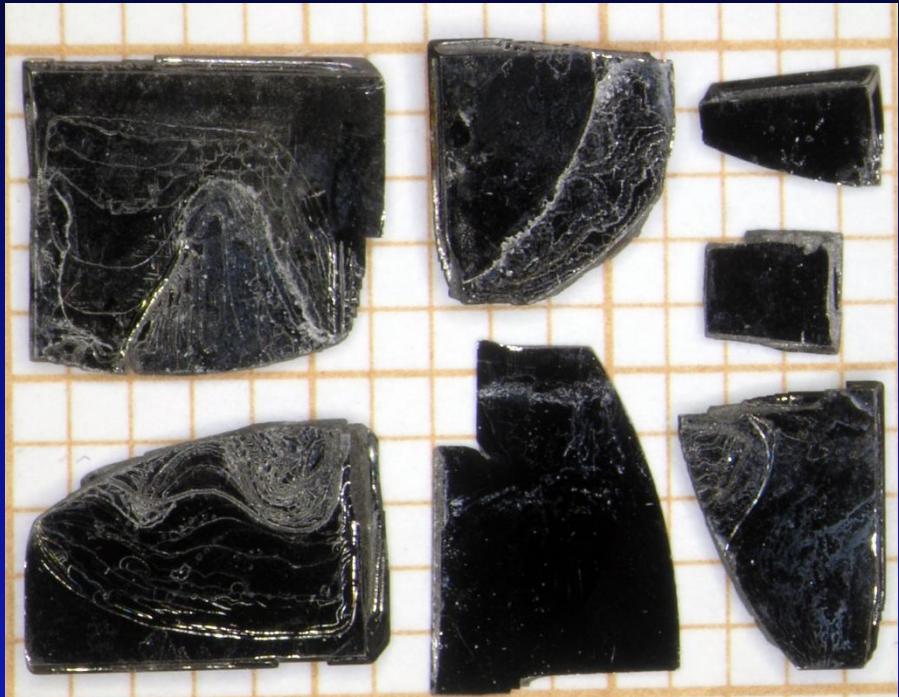
Zapf and Dressel, Rep. Prog. Phys. **80** (2017) 016501

L. M. Tran et al. PRB **98**, 104412 (2018)

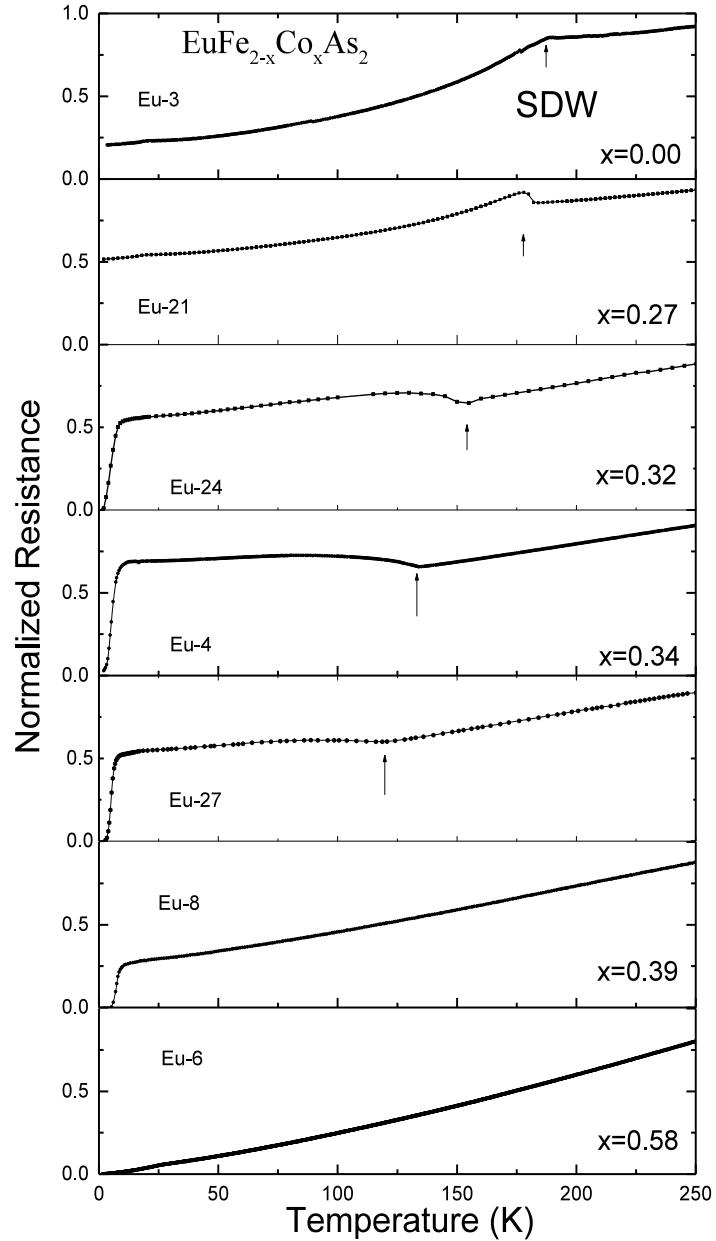


- Disappearance of magnetic order of Eu^{2+}
- SDW order remains intact

Co-substitution: $\text{EuFe}_{2-x}\text{Co}_x\text{As}_2$



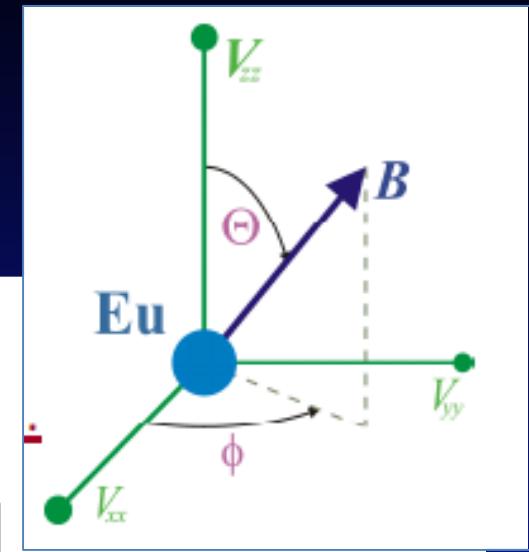
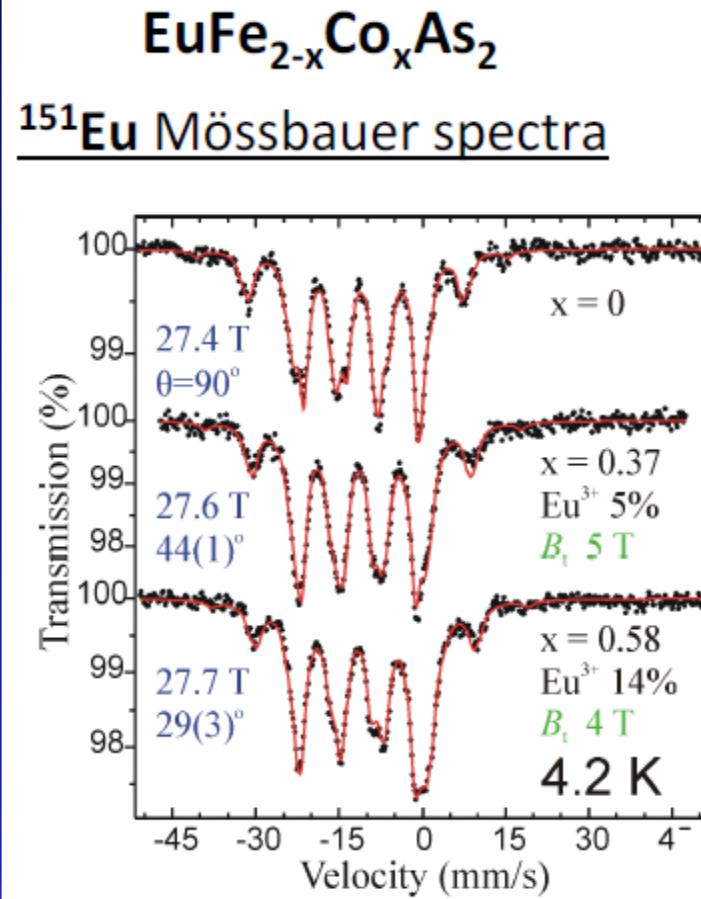
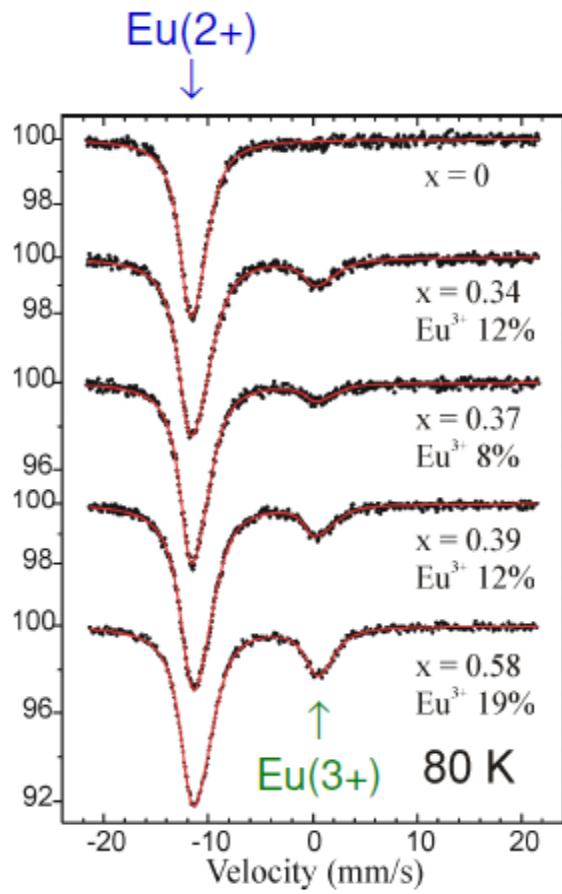
Single crystals of $\text{EuFe}_{2-x}\text{Co}_x\text{As}_2$
grown from Sn flux



Electrical resistivity

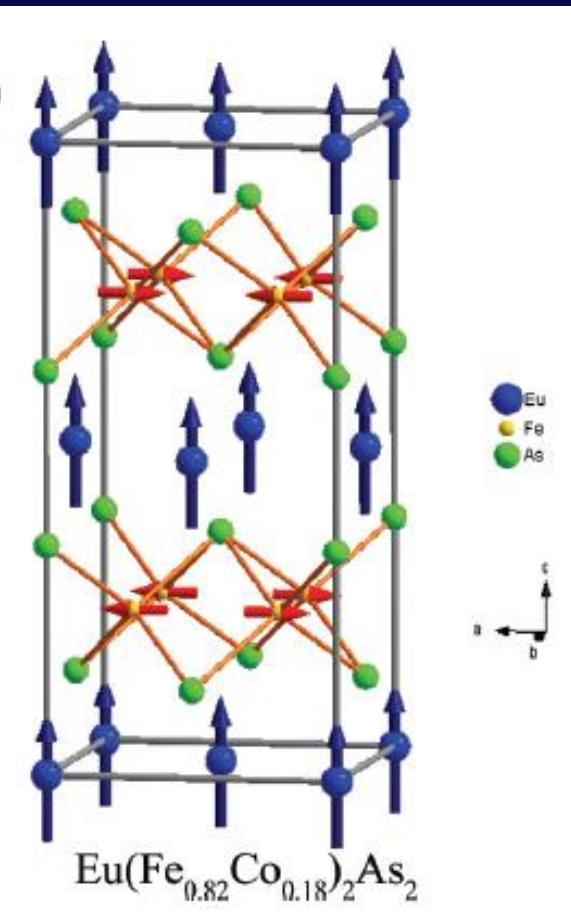
Mössbauer spectroscopy of $\text{EuFe}_{2-x}\text{Co}_x\text{As}_2$

A.Błachowski et al., Phys. Rev. B **84**, 174503 (2011)



Eu $^{2+}$ moments rotate from ab-plane toward c-axis direction

Magnetic structure of $\text{Eu}(\text{Fe}_{0.82}\text{Co}_{0.18})_2\text{As}_2$ (single-crystal neutron diffraction)



long-range ferromagnetic order of the Eu²⁺ moments along the *c* direction

$$T_C = 17 \text{ K}$$

no incommensurate magnetic reflections corresponding to the helical arrangement of the Eu²⁺ spins are observed

Antiferromagnetism of the Fe²⁺ moments still survives tetragonal-to-orthorhombic structural transition is observed transition temperatures of the Fe spin-density-wave (SDW) order and the structural phase transition are significantly suppressed to $T_{SDW} = 70 \text{ K}$ and $T_s = 90 \text{ K}$

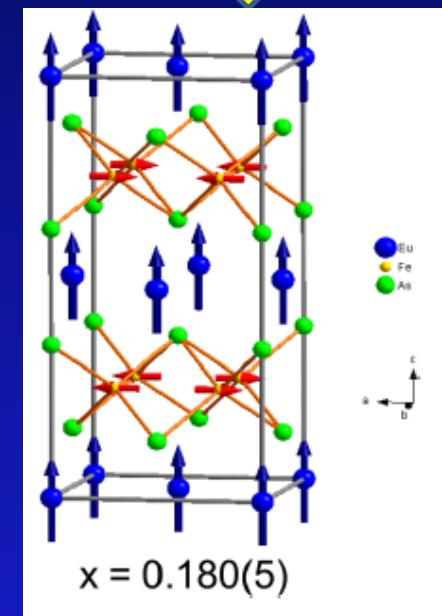
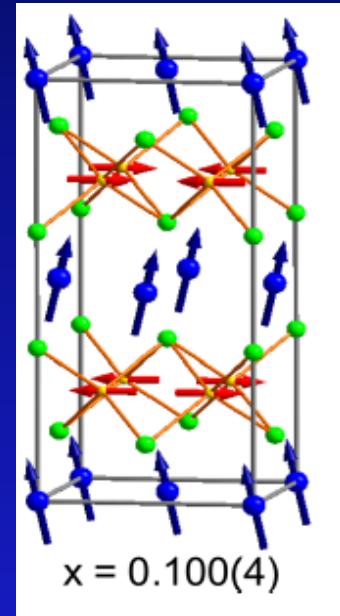
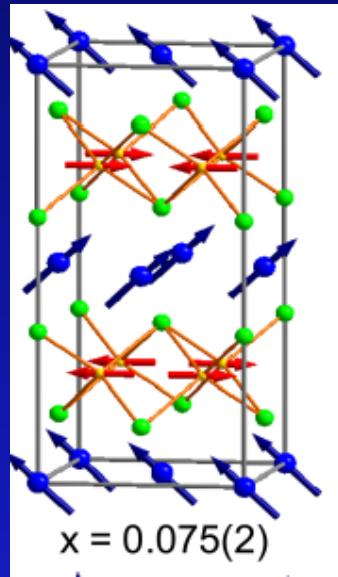
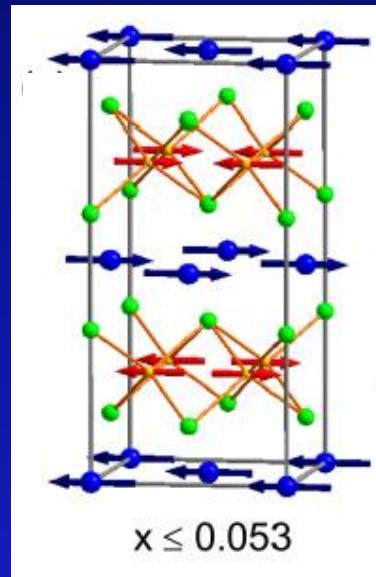
Superconducting $T_{SC} = 8 \text{ K}$

Effect of Co-doping on Eu²⁺ magnetic ordering in Eu(Fe_{1-x}Co_x)₂As₂ single crystals

Neutron diffraction

- ferromagnetic Eu²⁺ moment of $6.2\mu_B$ purely along the c direction
- Fe²⁺ moment is estimated to be $0.63(4)\mu_B$

W. T. Jin et al., Phys. Rev. B **94**, 184513 (2016)



Co concentration x →

A-type antiferromagnet



canted AF



ferromagnet

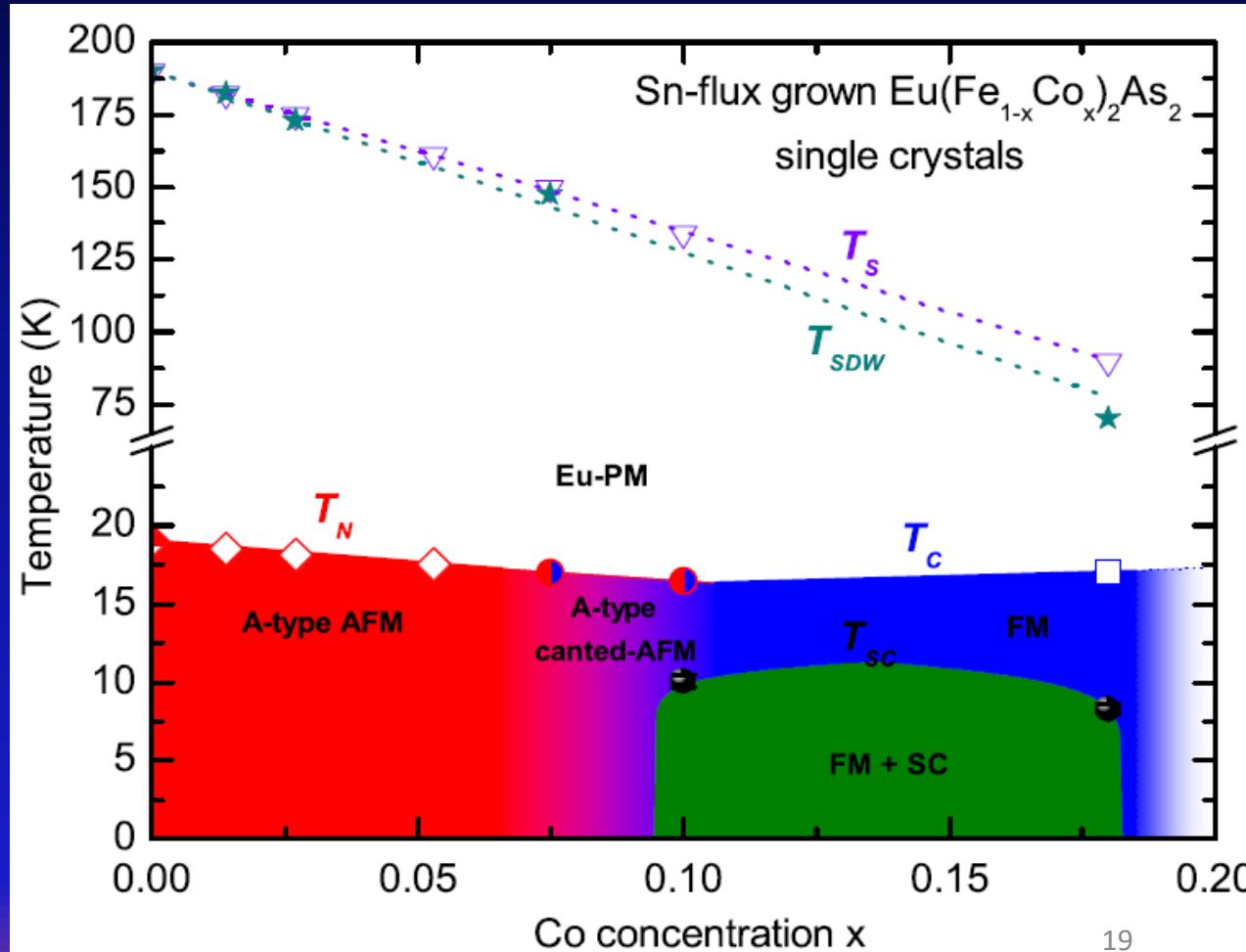
Magnetic phase diagram of $\text{Eu}(\text{Fe}_{1-x}\text{Co}_x)_2\text{As}_2$ (Sn-flux-grown single crystals)

W. T. Jin et al., PRB **94**, 184513 (2016)

suppression of SDW order

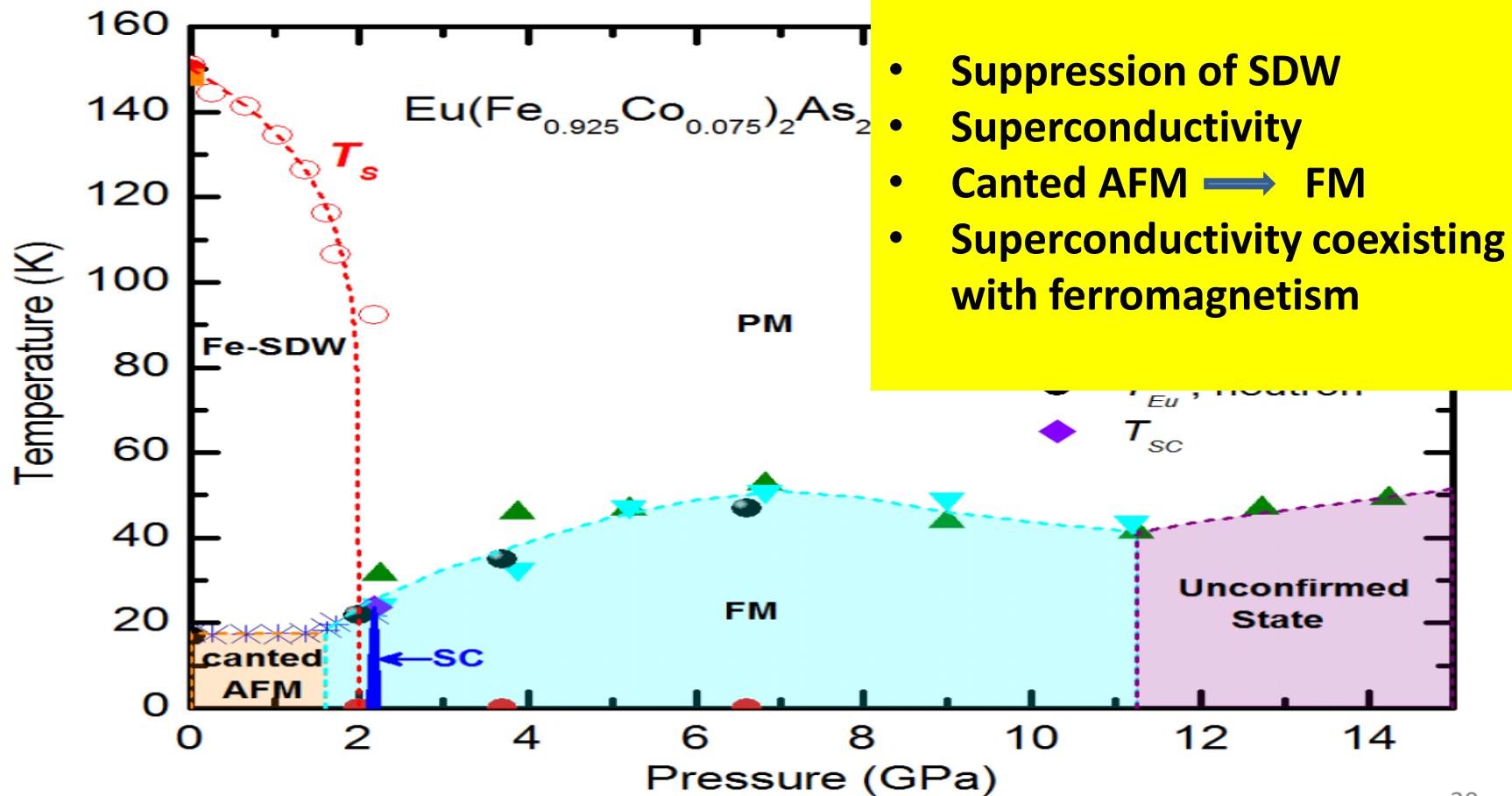
superconductivity competes
with Fe SDW
antiferromagnetic order

superconductivity coexists
with Eu ferromagnetism



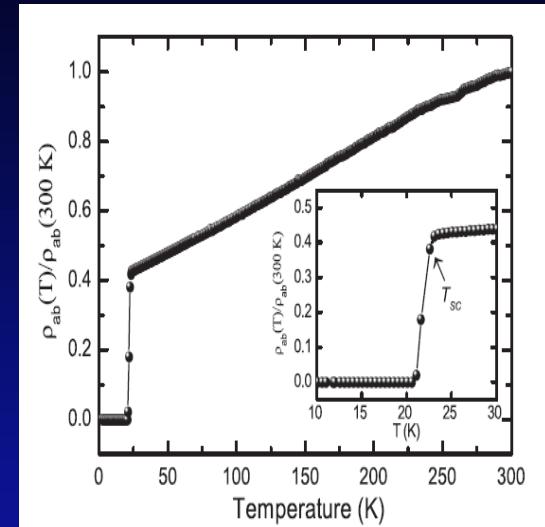
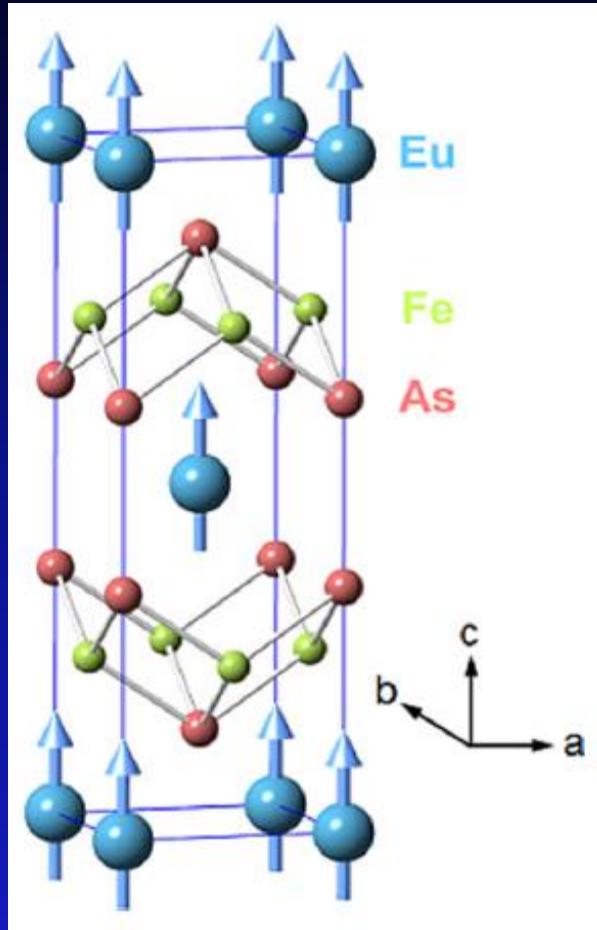
Hydrostatic pressure effects on the static magnetism in $\text{Eu}(\text{Fe}_{0.925}\text{Co}_{0.075})_2\text{As}_2$

W. T. Jin et al., Scientific Reports | 7: 3532 |



Ferromagnetic Eu(Fe_{0.86}Ir_{0.14})₂As₂

V. K. Anand et al., PRB **91** 094427 (2015)



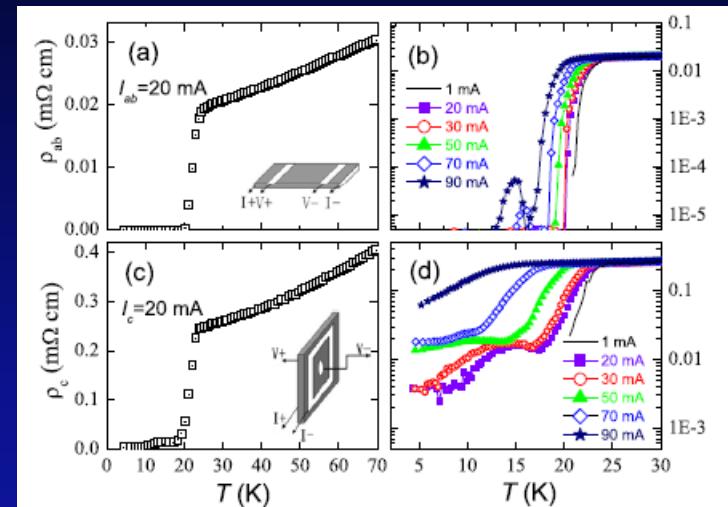
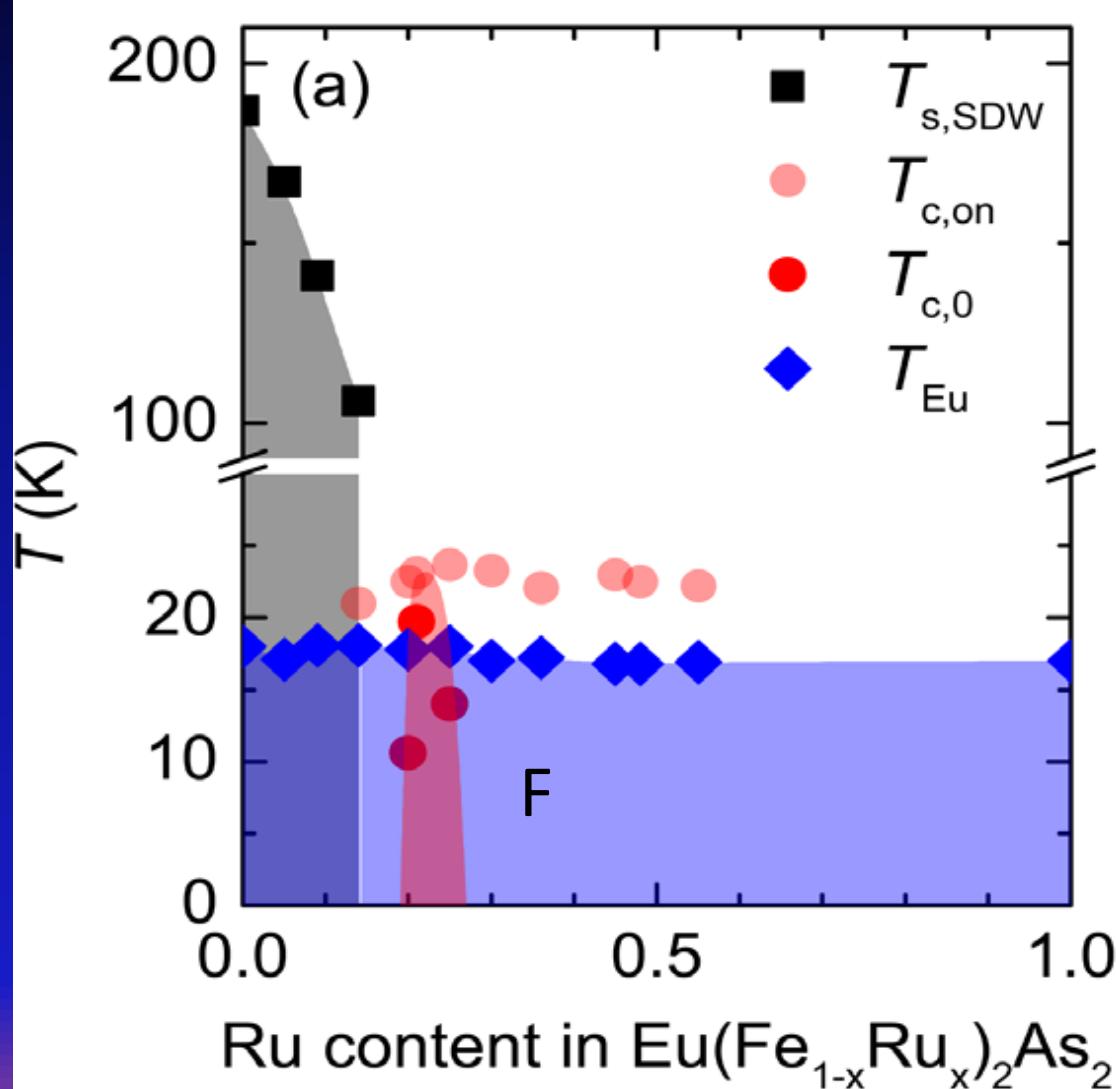
The body centered tetragonal chemical and magnetic unit cell (space group I4/mmm).

ferromagnetically coupled Eu moments are aligned along the c axis with a magnetic propagation wave vector $k = (0, 0, 0)$ and ordered moment of $6.29(5) \mu_B$ at 1.8 K.

$\text{Eu}(\text{Fe}_{0.75}\text{Ru}_{0.25})_2\text{As}_2$ ferromagnetic superconductor

Jiao et al., J. Phys.: Conf. Ser. 400 (2012) 022038

Jiao et al., EPL, 95 (2011) 67007

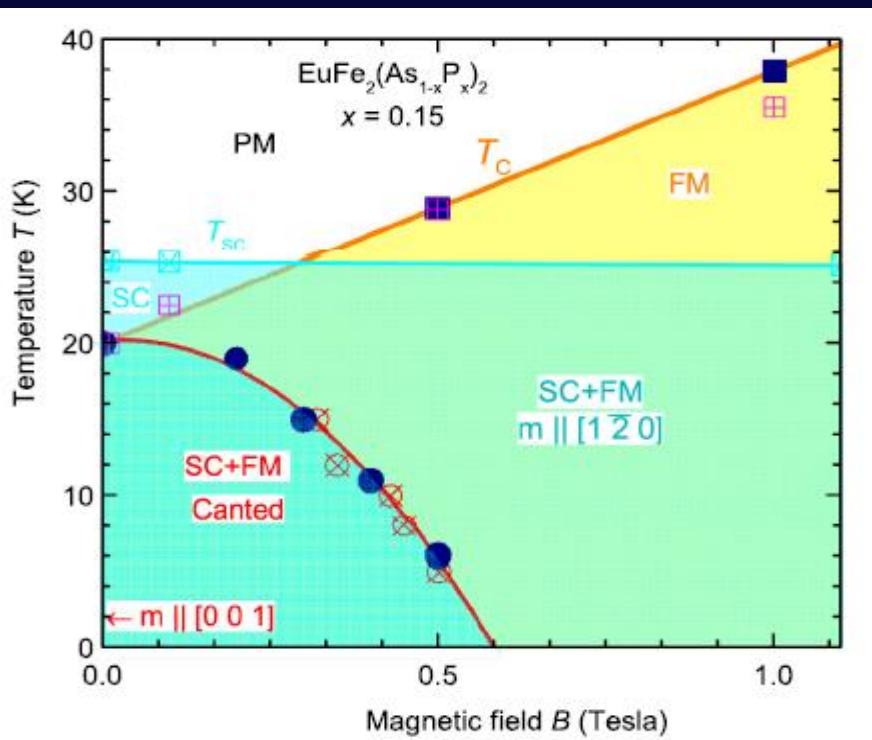


$$T_{\text{SC}} = 23 \text{ K}$$

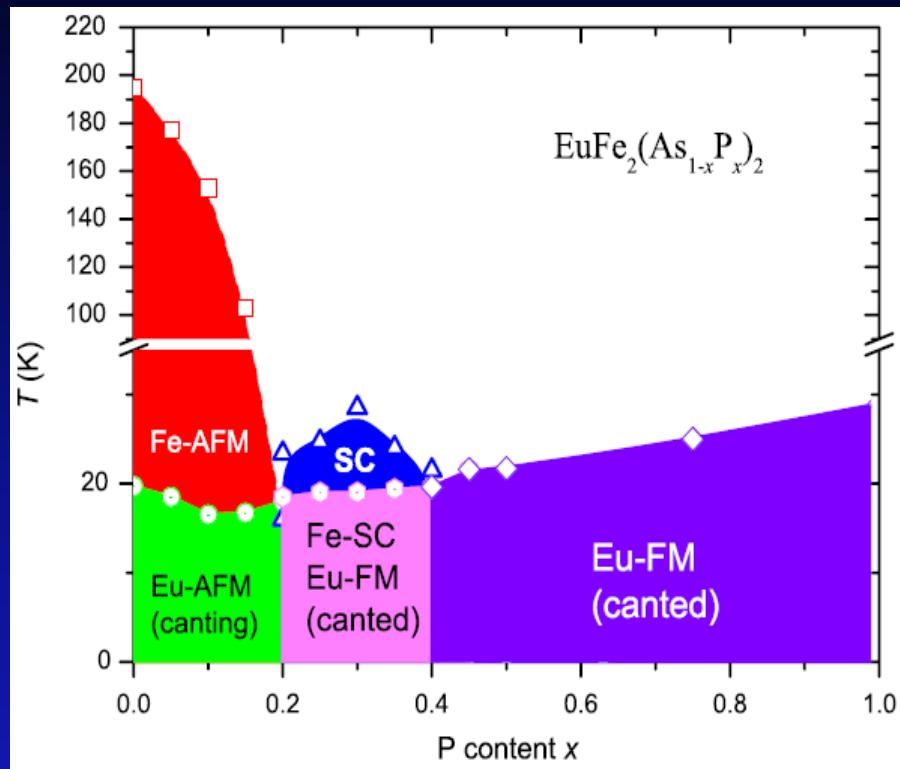
Mossbauer data indicate that the Eu^{2+} spins order ferromagnetically below 19.5 K with the moments tilted 20° from the c-axis.

Isovalent P-substitution EuFe₂(As_{1-x}P_x)₂

Nandi et al., PRB **89**, 014512 (2014)

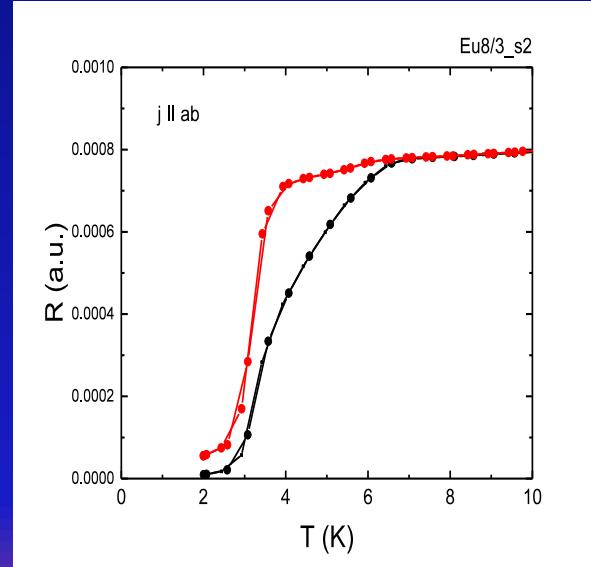
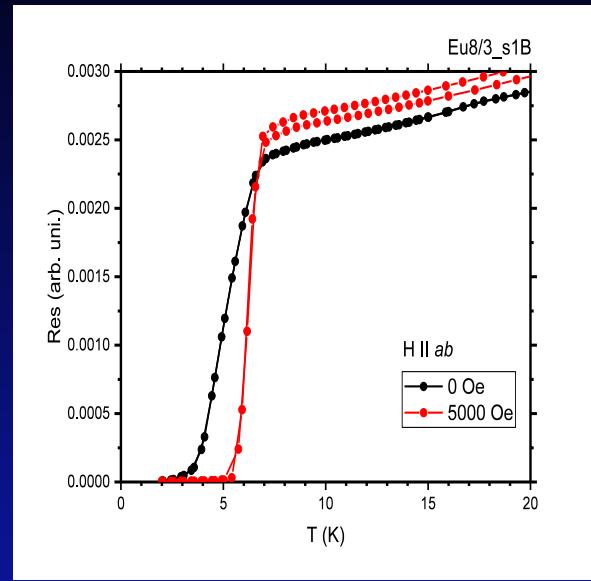
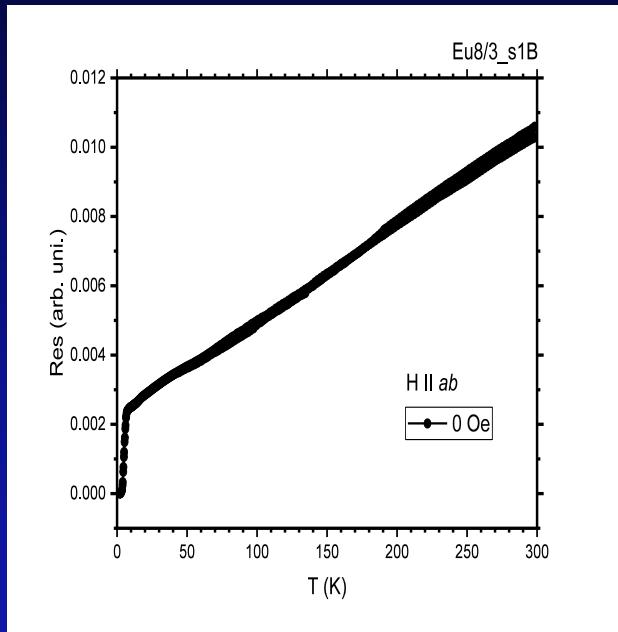


Cao et al. J. Phys.: Condens. Matter **23** (2011) 464204



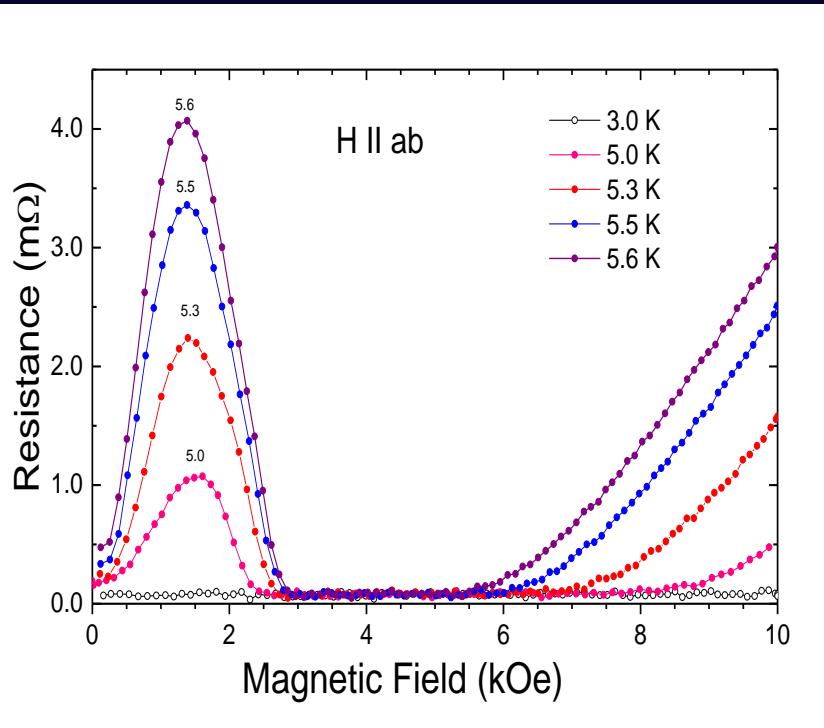
Superconductivity induced by partial substitution of P into As positions
 AFM \rightarrow Ferromagnetism
 Superconductivity coexists with ferromagnetism

Peculiar properties of Sn-flux-grown $\text{Eu}(\text{Fe}_{0.81}\text{Co}_{0.19})_2\text{As}_2$ single crystals

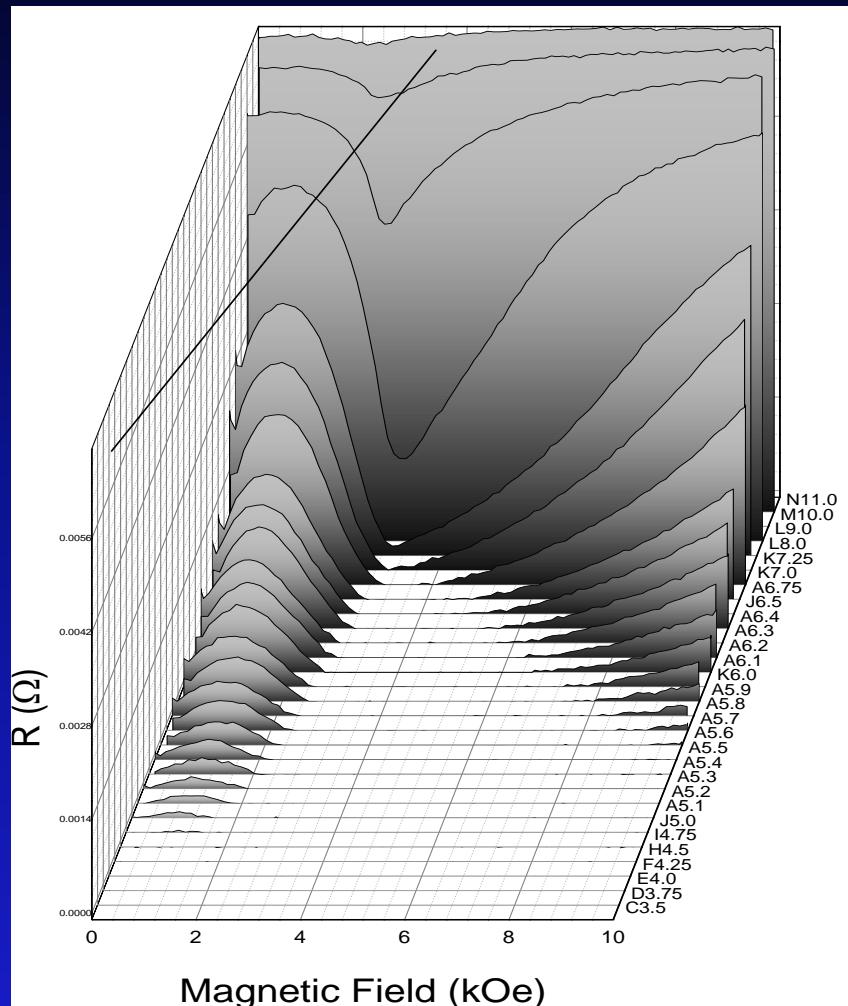


Magnetic field
enhancement of
superconductivity ?!

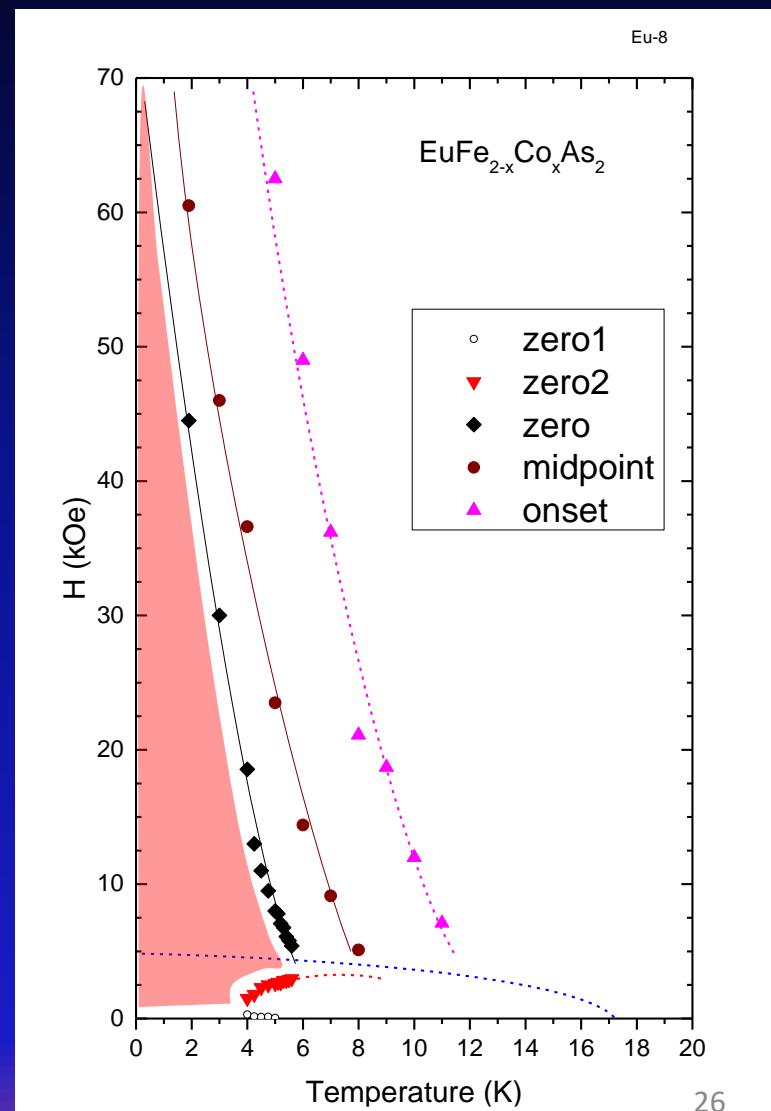
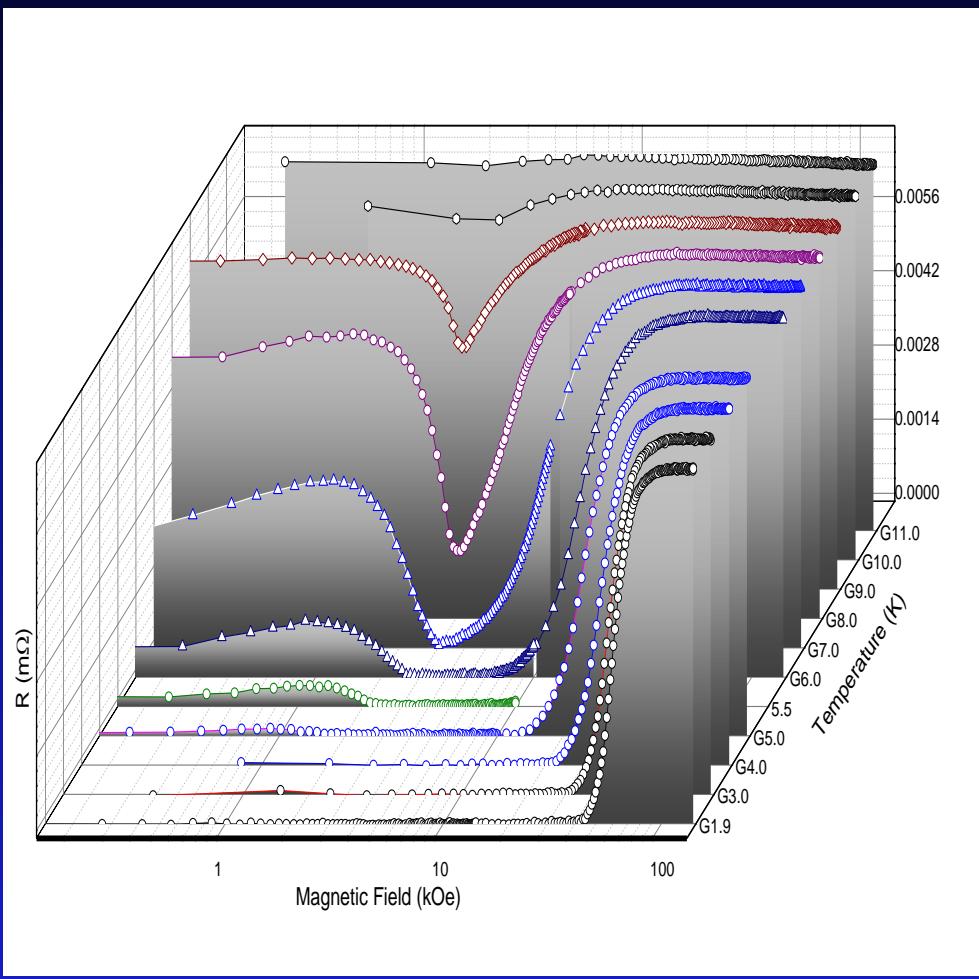
Peculiar properties of Sn-flux-grown $\text{Eu}(\text{Fe}_{0.81}\text{Co}_{0.19})_2\text{As}_2$ single crystals



Resistivity „peak” most likely corresponds to the flux flow effect

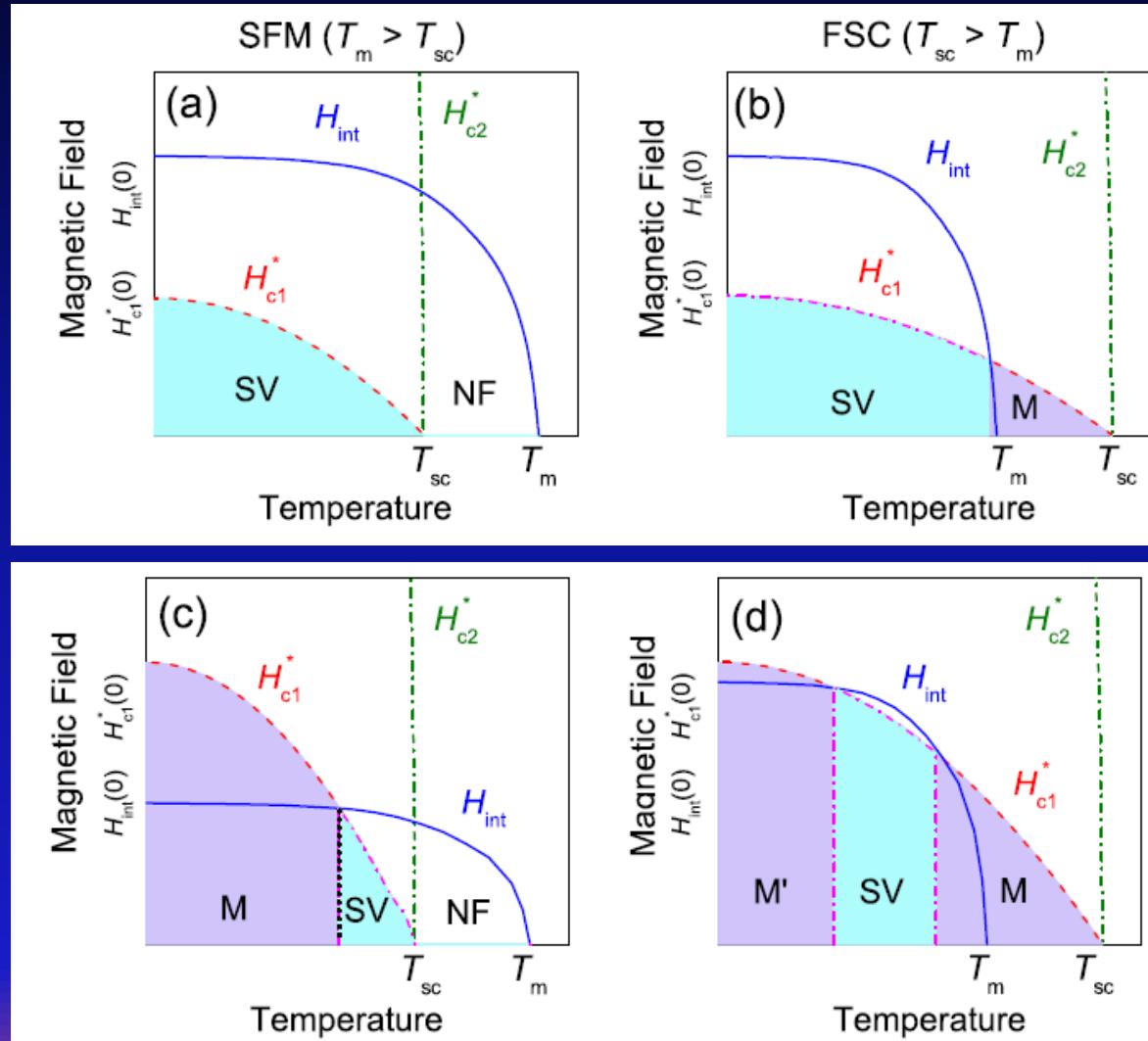


Peculiar properties of Sn-flux-grown $\text{Eu}(\text{Fe}_{0.81}\text{Co}_{0.19})_2\text{As}_2$ single crystals

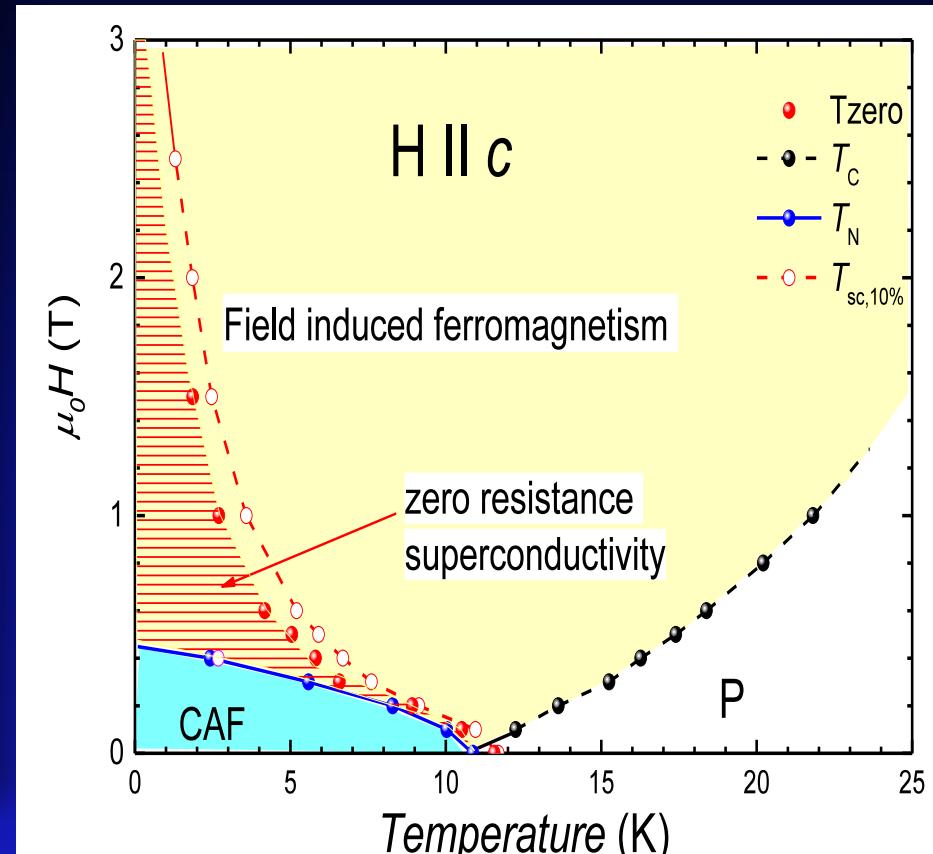
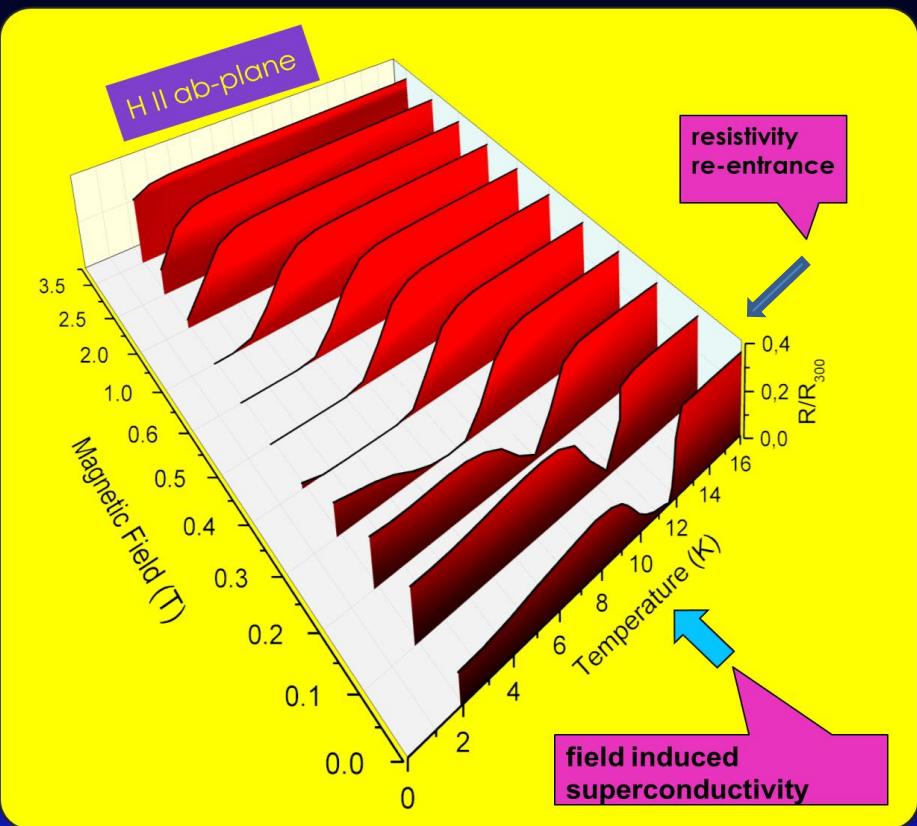


Spontaneous vortex state in ferromagnetic superconductor

W-H Jiao et al., npj Quantum Materials (2017) 2:50



Peculiar properties of Sn-flux-grown $\text{Eu}_{0.73}\text{Ca}_{0.27}(\text{Fe}_{0.87}\text{Co}_{0.13})_2\text{As}_2$ single crystals

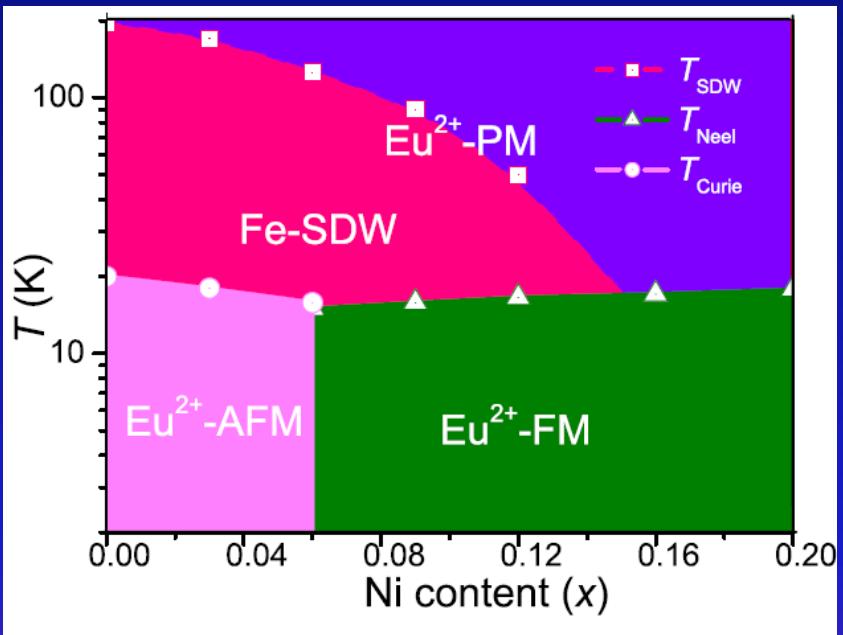


Zero-resistance superconductivity is suppressed in antiferromagnetic region and coexists with field induced ferromagnetism

Search for superconductivity in Ni-substituted EuFe_2As_2

I Nowik et al. *New Journal of Physics* **13** (2011) 023033

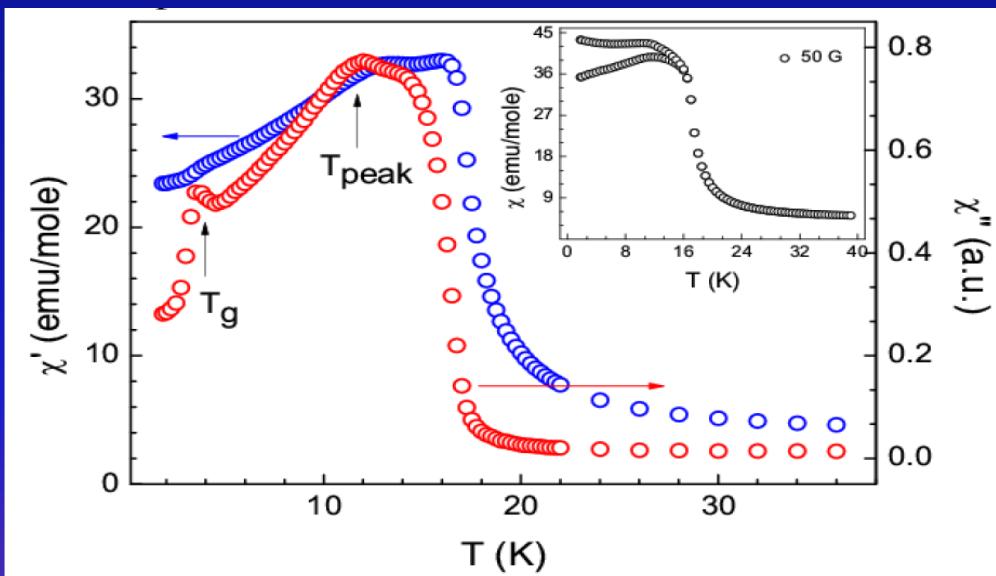
Polycrystalline material



Zhi Ren et al. *PRB* **79**, 094426(2009)

Mössbauer studies of $\text{Eu}(\text{Fe}_{0.9}\text{Ni}_{0.1})_2\text{As}_2$ and $\text{Eu}(\text{Fe}_{0.89}\text{Co}_{0.11})_2\text{As}_2$, in particular the Eu negative quadrupole interaction and the tilting of H_{eff} from the c -axis, are almost the same. This indicates a similar magnetic structure regardless of whether the system is normal conducting or SC

Anupam et al. *AIP Conf. Proc.* **1349**, 1293-1294 (2011)



Superconductivity not detected

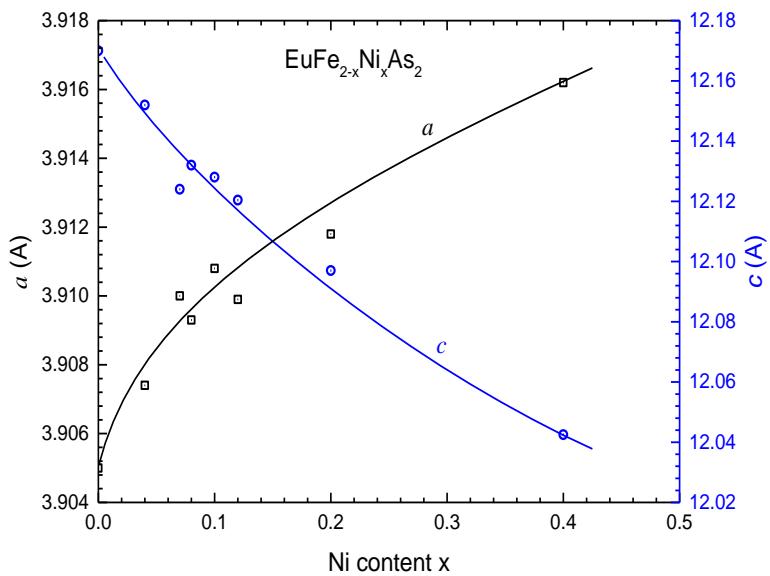
In $\text{EuFe}_{1.9}\text{Ni}_{0.1}\text{As}_2$ in addition to FM transition, two more transitions were observed. $T_g = 3.5$ K. The broad transition at $T_{\text{peak}} = 11.5$ K could be due to the transition from FM to AFM state. The transition at $T_g = 3.5$ K could be due to the spin glass ordering, which might arise due to the competition between FM and AFM ordering and hence leads to the spin freezing at T_g .

Ni substitution in $\text{EuFe}_{2-x}\text{Ni}_x\text{As}_2$

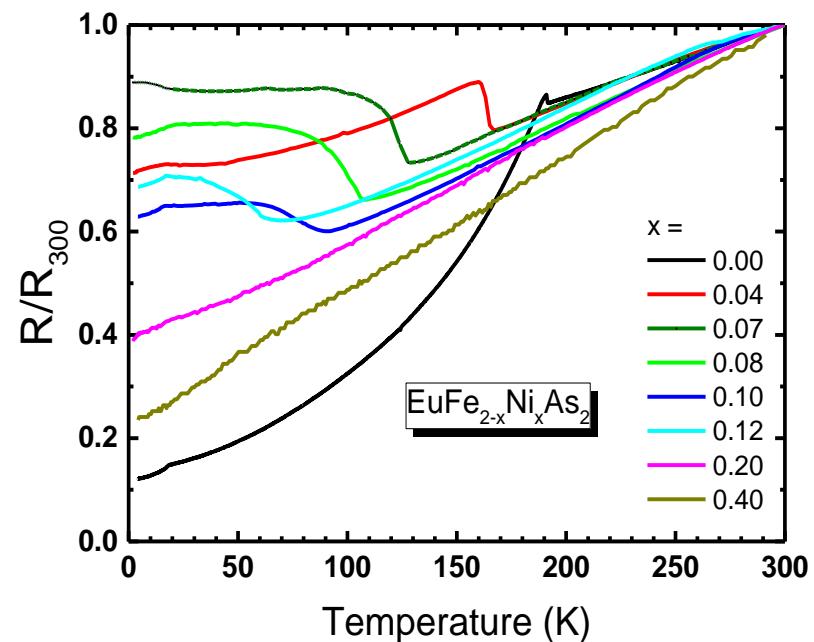


Single crystals of $\text{EuFe}_{2-x}\text{Ni}_x\text{As}_2$ grown from Sn flux
(up to $x=0.4$)
Chemical composition-determined from EDS data

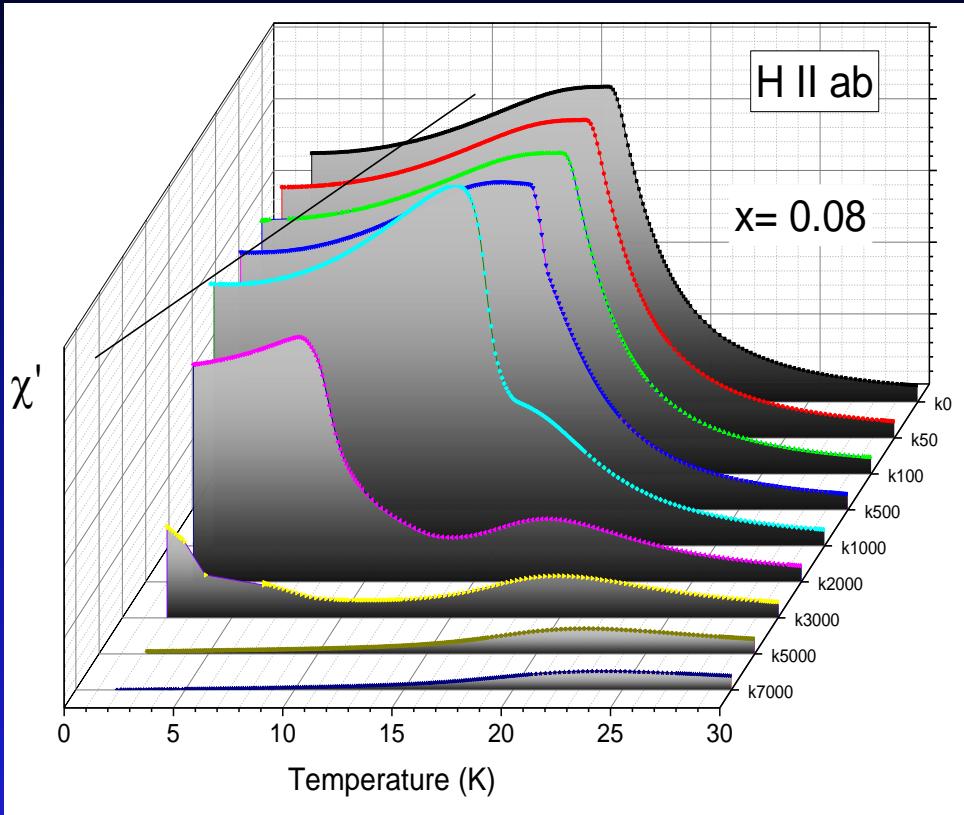
Lattice parameters



Electrical resistivity

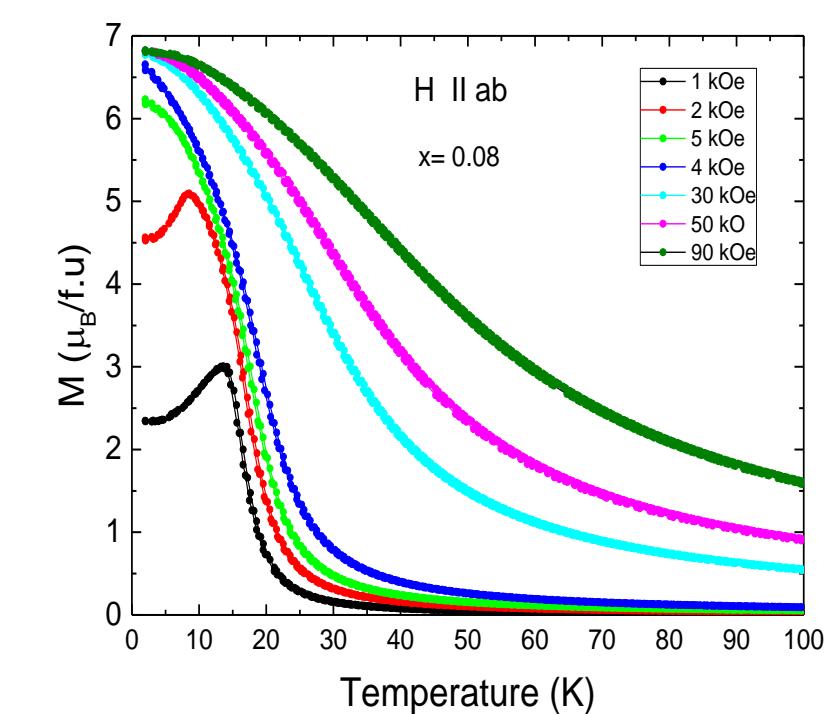


Magnetic properties of $\text{EuFe}_{1.92}\text{Ni}_{0.08}\text{As}_2$

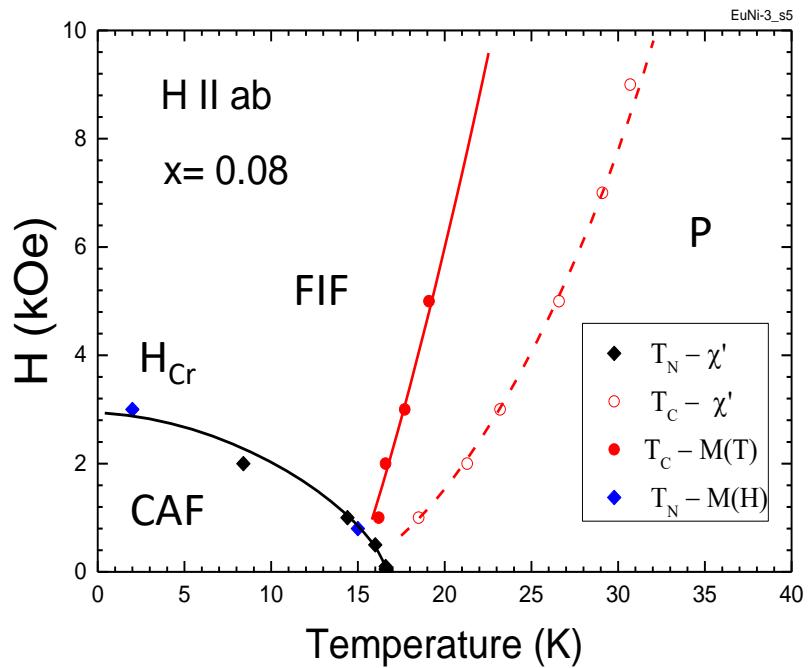
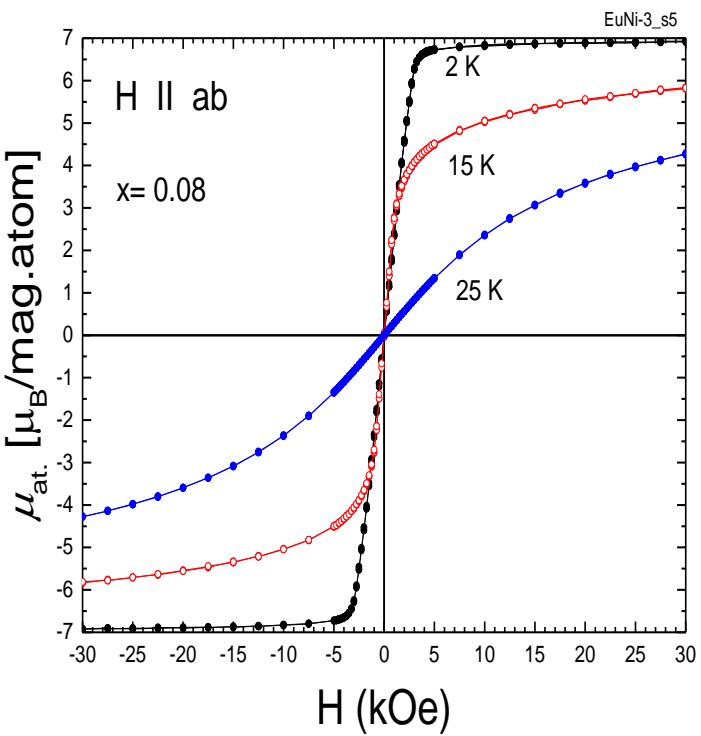


AC-susceptibility vs. Temperature in various magnetic fields

Magnetization vs. Temperature in various magnetic fields

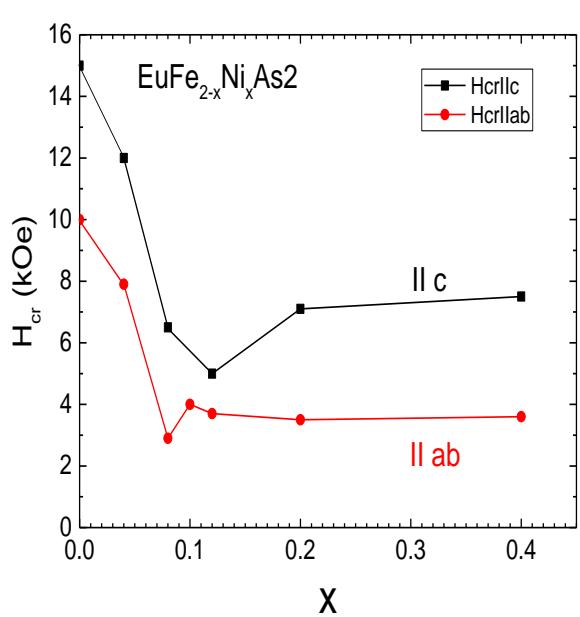


Magnetic properties of $\text{EuFe}_{1.92}\text{Ni}_{0.08}\text{As}_2$

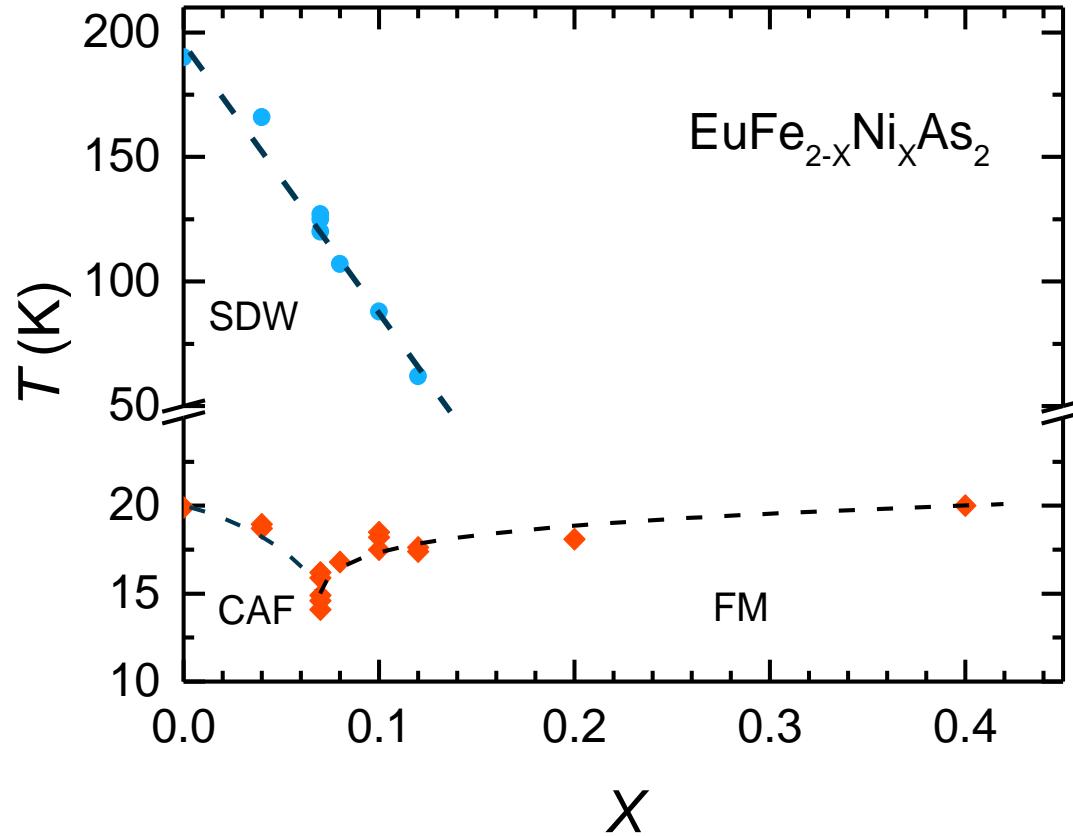


Field –dependent magnetization in various temperatures

Magnetic phase diagram of $\text{EuFe}_{2-x}\text{Ni}_x\text{As}_2$



Absence of superconductivity above 1.8 K

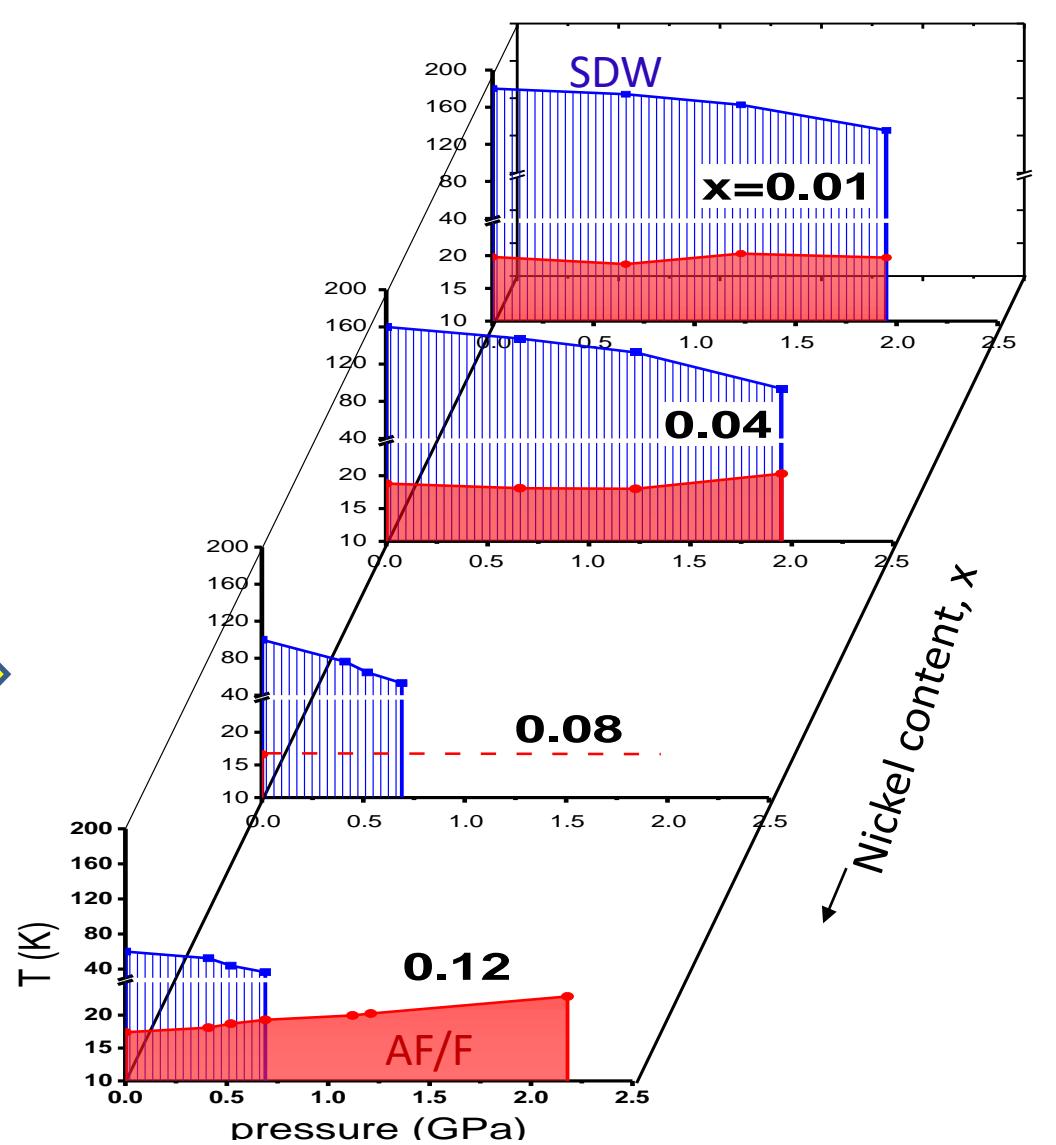


Magnetism of Eu in $\text{EuFe}_{2-x}\text{Ni}_x\text{As}_2$ is very similar to that in $\text{EuFe}_{2-x}\text{Co}_x\text{As}_2$ and seems to be not responsible for the absence of superconductivity.

Search for superconductivity in $\text{EuFe}_{2-x}\text{Ni}_x\text{As}_2$ under high pressure

Resistivity measured using piston-cylinder pressure cell

no evidence of superconductivity under pressure down to 2 K



Doped EuFe₂As₂

- Magnetic field easily aligns Eu²⁺ spins along the direction of the applied field (field induced ferromagnetism)
- Hydrostatic pressure, transition metal substitutions, and P substitution suppress SDW order, induce superconductivity and change magnetic order of Eu²⁺ moments from antiferro- to ferromagnetic
- Superconductivity coexists both with AF and F order of Eu²⁺ system

Coexistence of superconductivity and magnetism,
Zero-resistance as an effect of applied magnetic field,
High anisotropy,
Magnetic field sensitive electronic transport ,
Spontaneous superconducting vortices,
- potentially interesting for spintronics and other electronic applications

Collaboration:

Presented unpublished results obtained in fruitfull collaboration with:

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