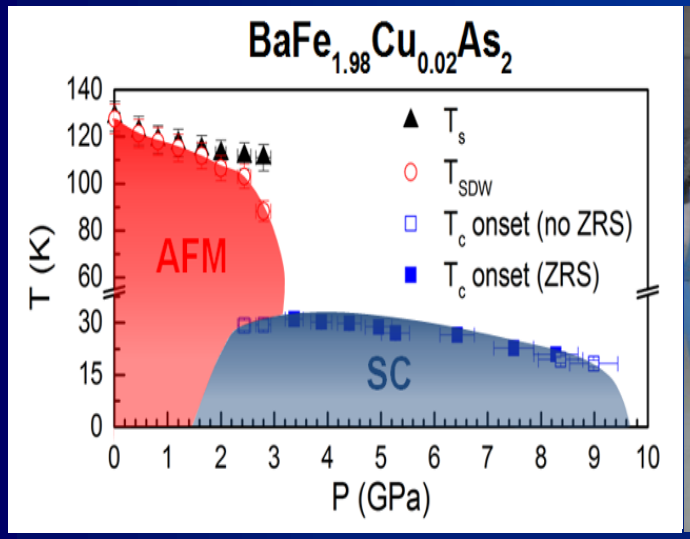
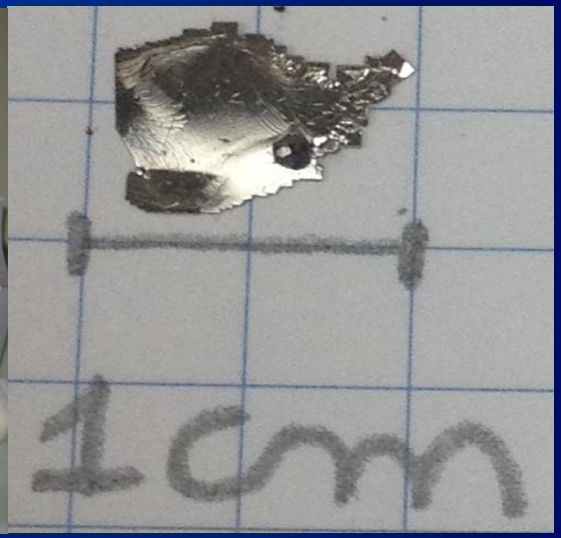
ICTP SAIFR | International Centre for Theoretical Physics
South American Institute for Fundamental Research

unesp
UNIVERSIDADE ESTADUAL PAULISTA "JÚLIO DE MESQUITA FILHO"

Workshop on Next Generation Quantum Materials

Combined external pressure and chemical substitution studies on $BaFe_2As_2$ single crystals





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GPOMS

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Outline

I. Motivation – Fe-based SC

II. In-flux crystals growth

III. The role of local distortions in the Fe-site

V. Actual doping x orbital differentiation of the Fe 3d bands?

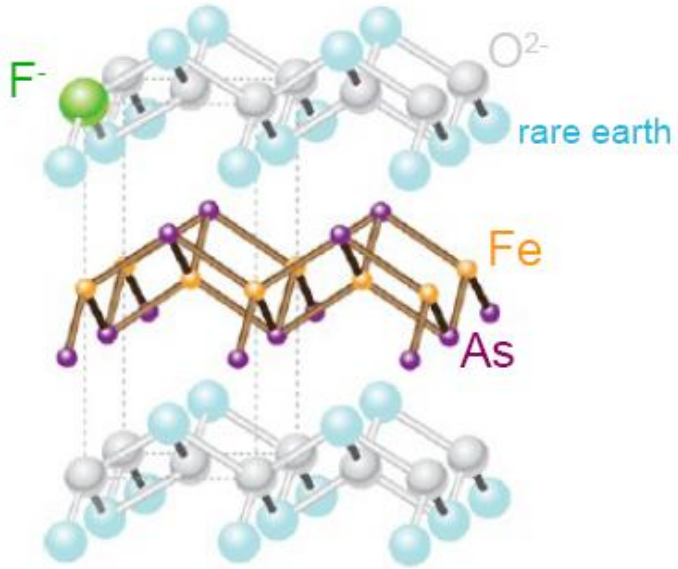
VI. Combined pressure and chemical substitution experiments

VII. The particular case of Cu-substitution in $BaFe_2As_2$

VIII. Final Remarks

Motivation

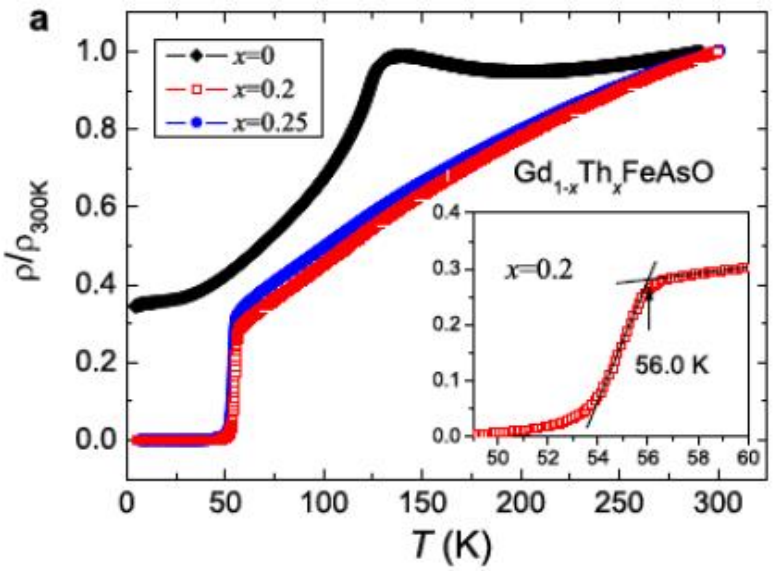
- Initial Discovery: $LaFeAsO_{1-x}F_x$ (Kamihara et al., 2008, T_c = 26 K) – T_c can get as high as ~ 56 K for R = Gd.



Initial discovery: T_c = 28 K in

F-doped LaOFeAs

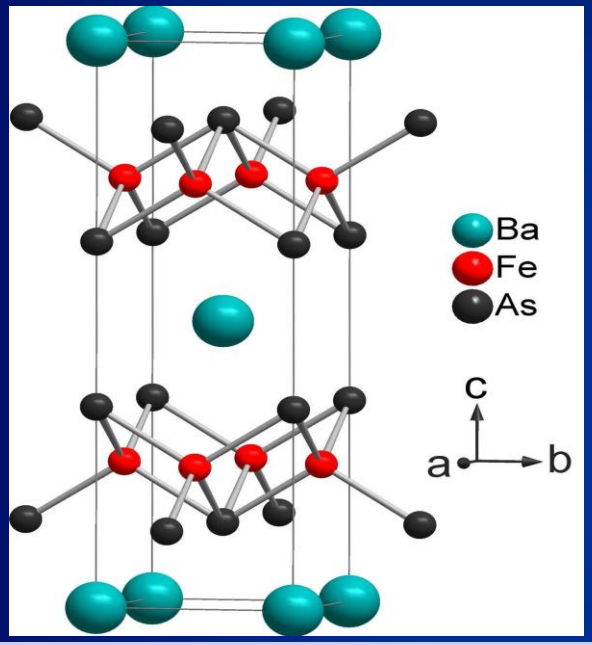
Y. Kamihara, T. Watanabe, M. Hirano, and H. Hosono, J. Am. Chem. Soc. **130**, 3296 (2008).

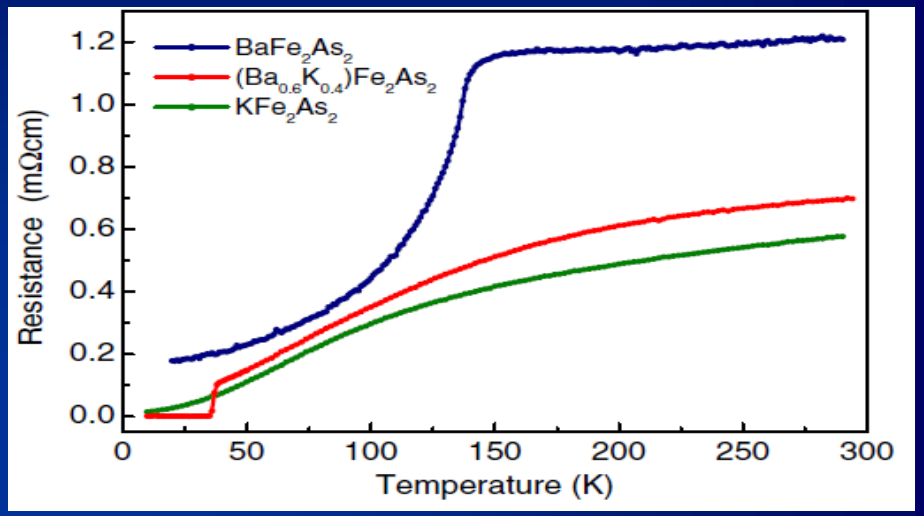


C. Wang *et al.*, Europhys. Lett. **83**, 67006 (2008).

Motivation

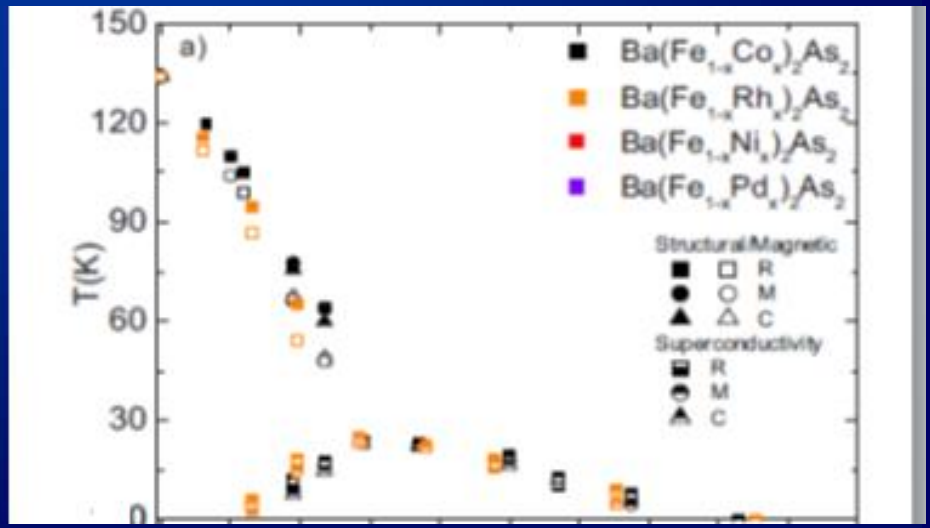
Discovery of Oxygen free intermetallic "relatives": $Ba_{1-x}K_xFe_2As_2$ (Rotter et al., 2008, $T_c \sim 40$ K).





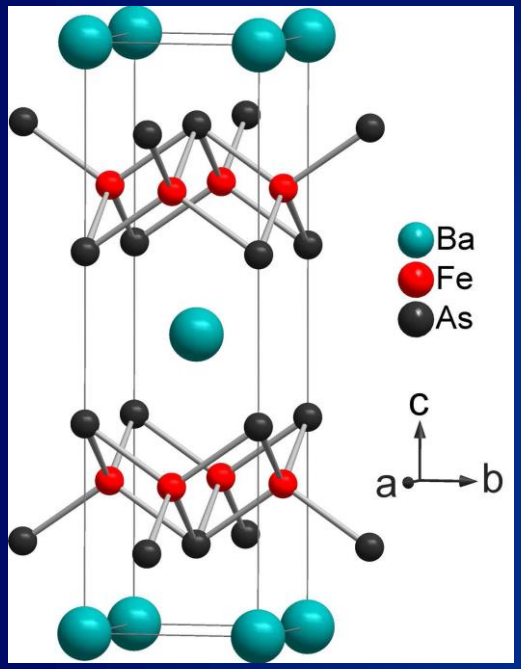
M. Rotter *et al.*, Phys. Rev. Lett. **101**, 107006 (2008)

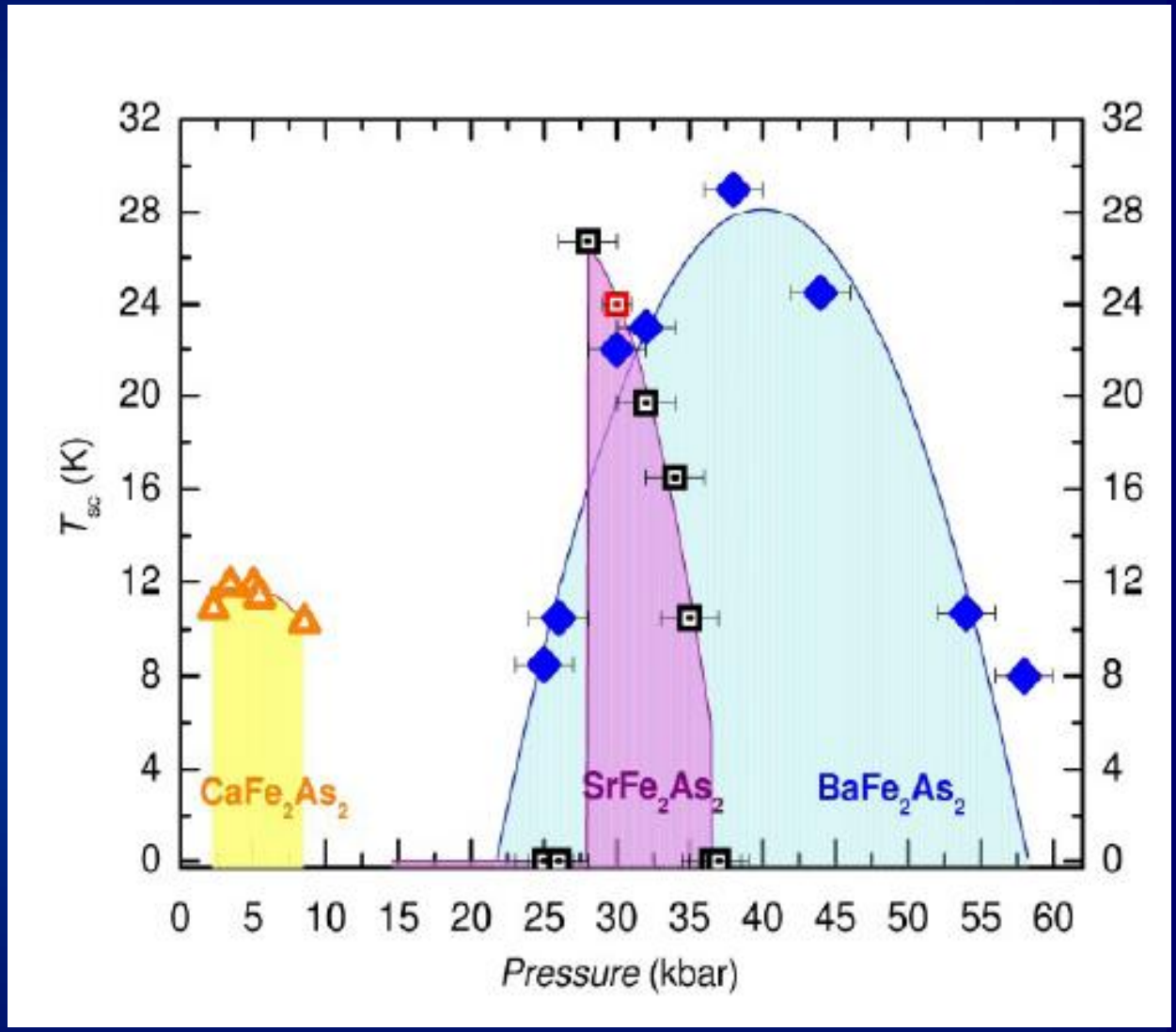
Two classes of FeAs-based materials (oxides and intermetallic compounds) with comparable Tcs and phase diagrams!!



N. Ni *et al.*, Phys. Rev. B **80**, 024511 (2009)

Pressure induced SC – similar to HFS

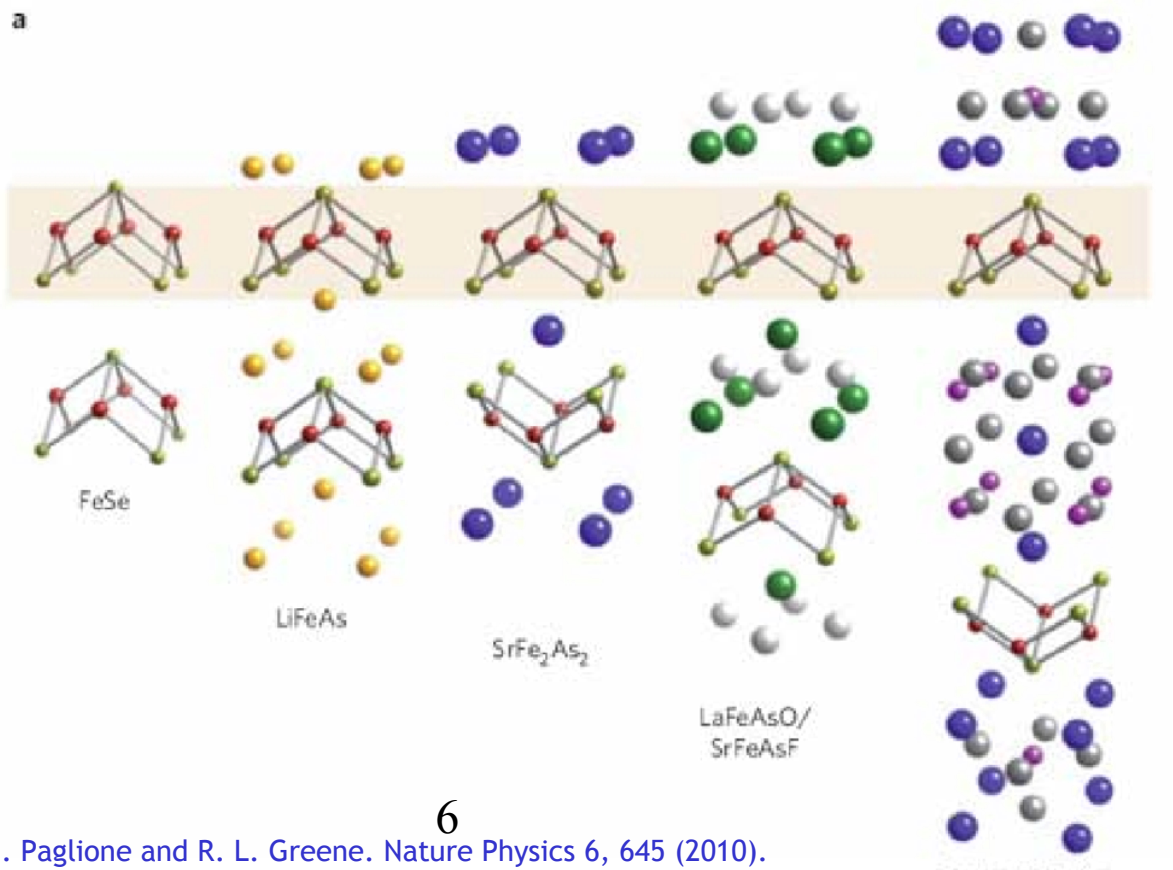


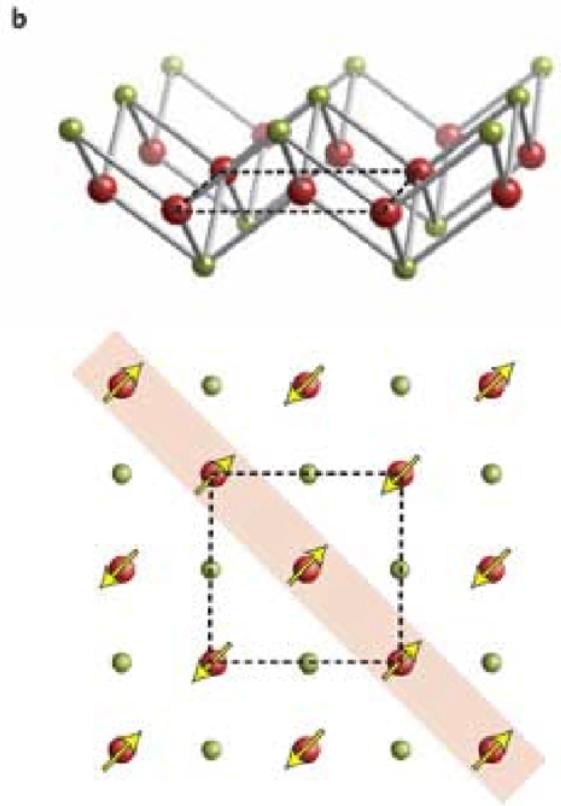


• M. S. Torikachvili, S.L. Bud´ko, N. Ni, and P.C. Canfield, Phys. Rev. Lett. **101**, 057006 (2008);
• ibidem Phys. Rev. B **78**, 104527 (2008).
• P. Alireza *et al.*, J. Phys.: Condens. Matter **21**, 012208 (2009).
• T. Park *et al.*, J. Phys..: Condens. Matter **20**, 322204 (2008).
•H. Fukasawa *et al.*, condmat/0808.0718.
• A. Mani *et al.*, cond-mat/0903.4236.
•E. Colombier, S.L. Bud´ko, N. Ni, and P.C. Canfield, condmat/0904.4488.

Fe-based superconductors

- Common Structural parameter: $Fe(As,Se)_4$ tetrahedral
- Common Electronic parameter: SDW magnetic instability





J. Paglione and R. L. Greene. Nature Physics 6, 645 (2010).

Open Questions!

1) What is main microscopic tuning parameter responsible for driving SDW to SC ?

2) Are the structural and SDW phase transitions first or second order?

3) Do these two transitions happen simultaneously/independently? Do they have direct influence on the emergence of the SC phase?

4) What are the universal emergent phenomena in the Fe-based SC regarding the pairing mechanism and the SC gap symmetry?

5) What is the real potential for application of the existing Fe-based superconductors?

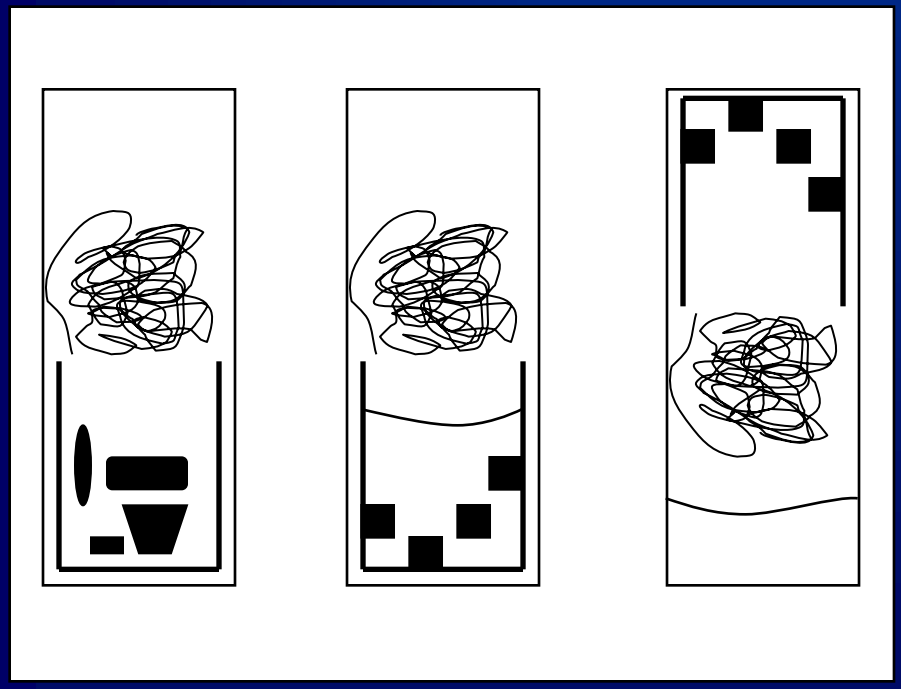
Flux Growth - Introduction

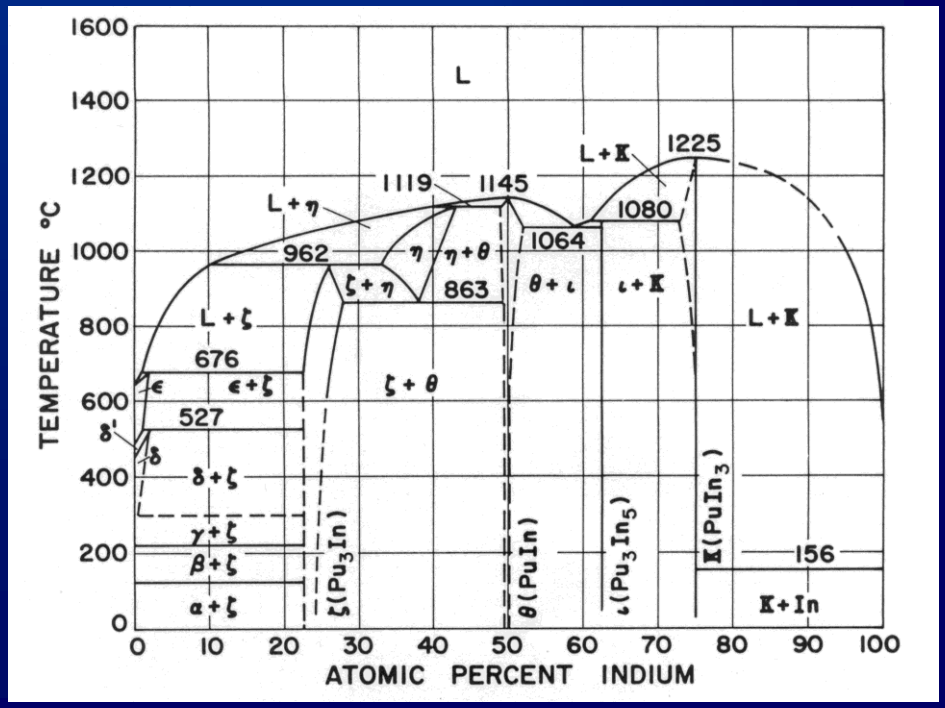
Growth of single crystals from low-melting solvents

- Simple hardware/inert atmosphere
- Low temperature / short time process
- Self-cleaning / well-faceted morphologies

Fisk & Remeika (1989)

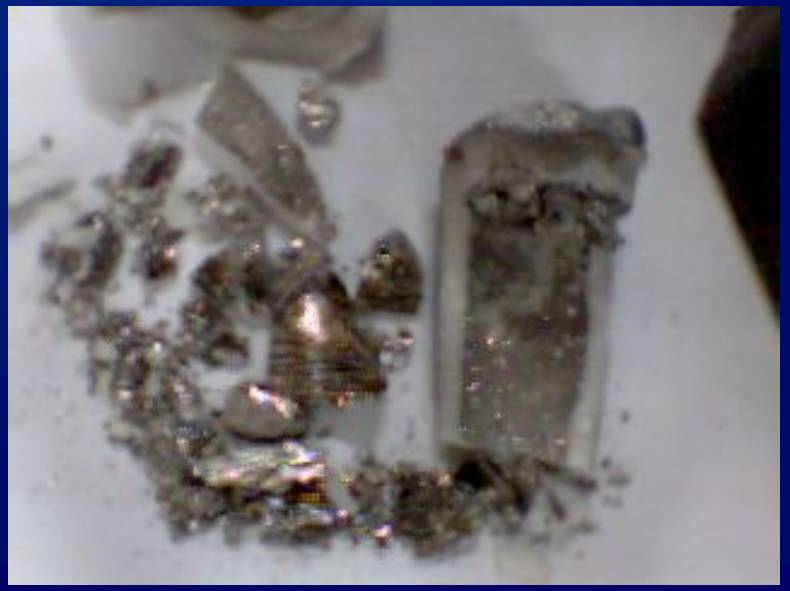
Canfield & Fisk (1992)



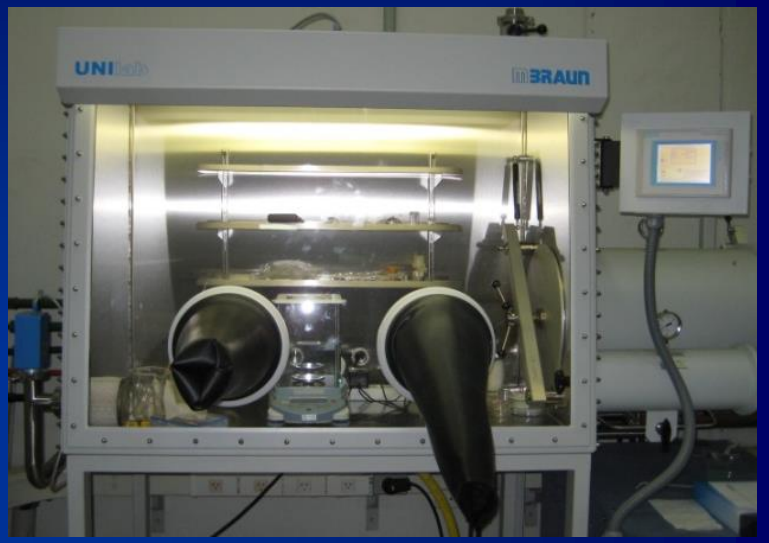


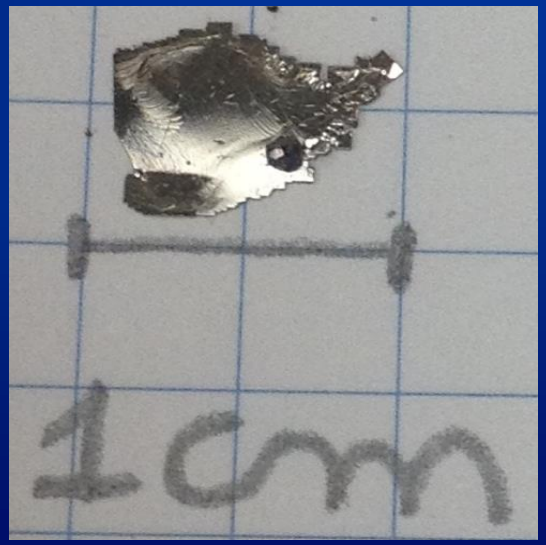
Metallic Flux Growth

Metallic Flux Growth

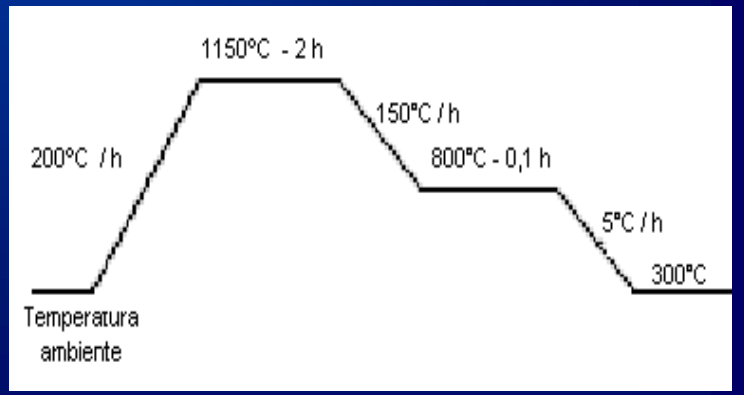


In-flux samples growth at GPOMS - Campinas

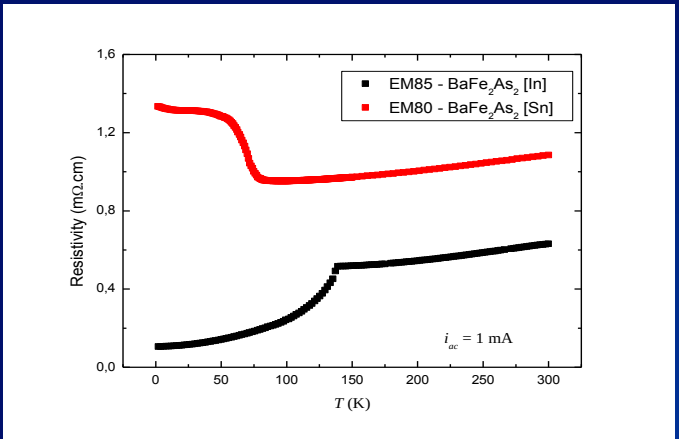


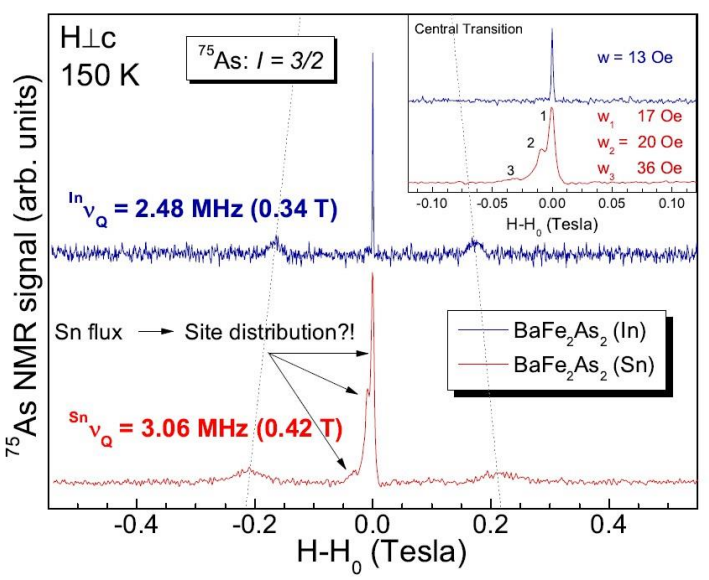


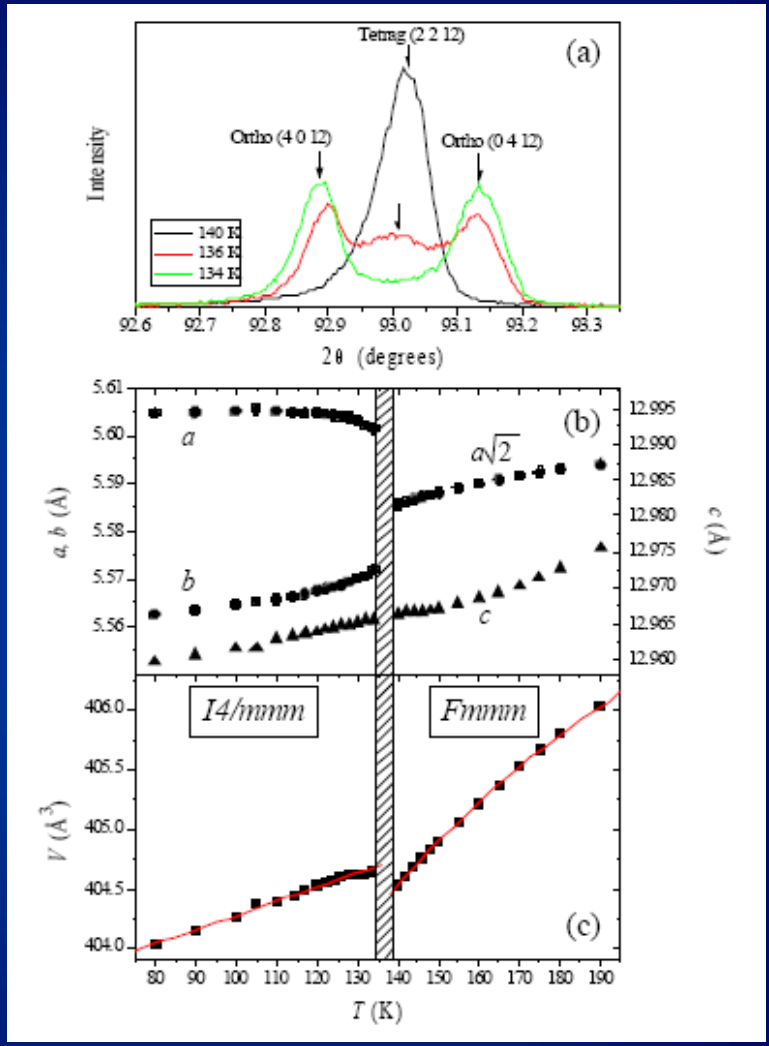
1 Ba+ 2 Fe + 2 As + 25 In



Comparison $BaFe_2As_2$: In- e Sn







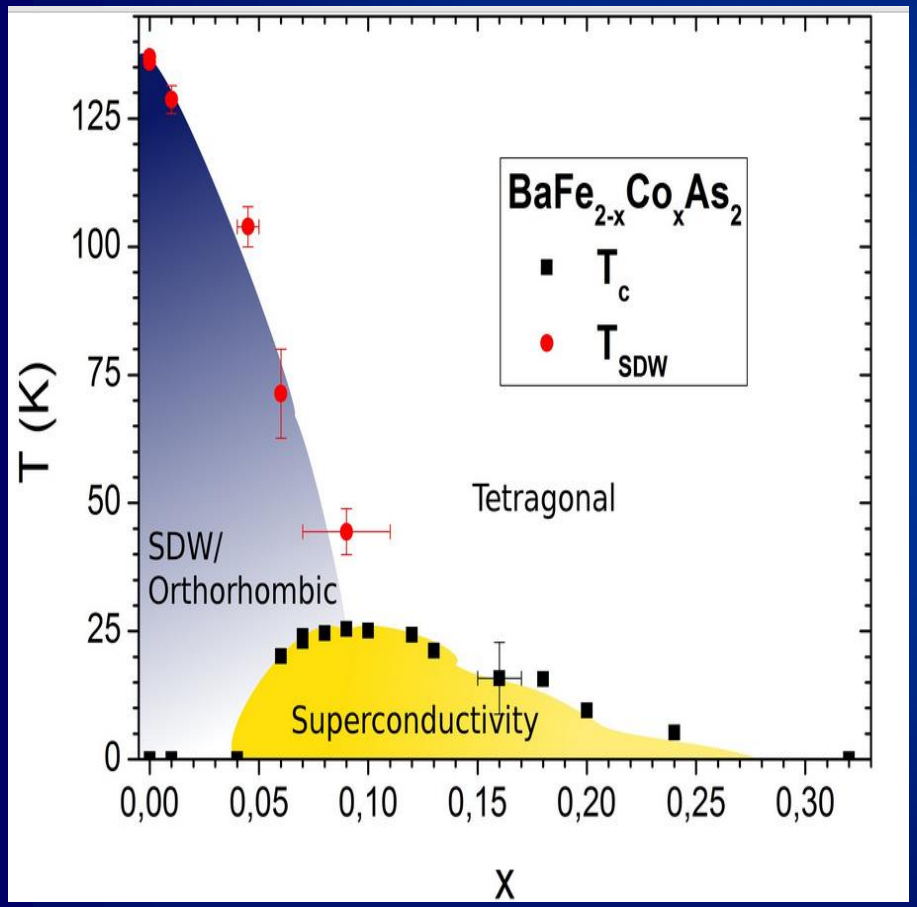
R. R. Urbano... P. G. Pagliuso *et al* ,*Phys. Rev. Lett.* **105** 107001 (2010).

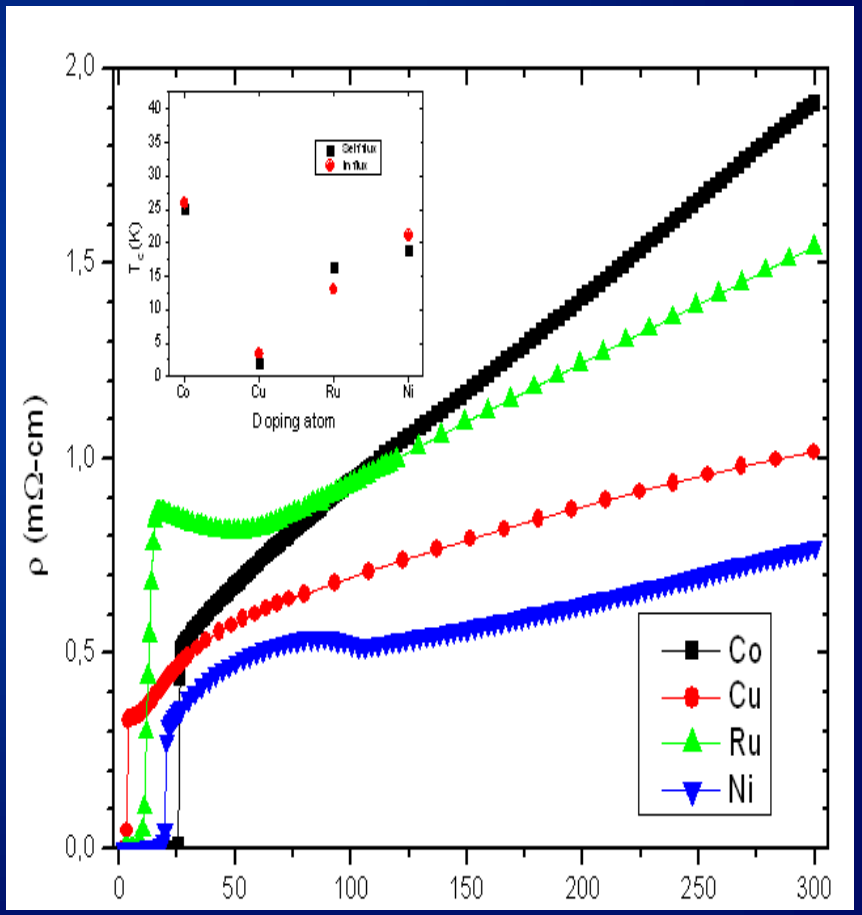
T. M . Garitezi…P. G. Pagliuso *et al* 2013 *Braz. J. Phys.* 43, 223-229 (2013).

In-flux grown samples are free of In – inclusion

Results – In flux grown samples

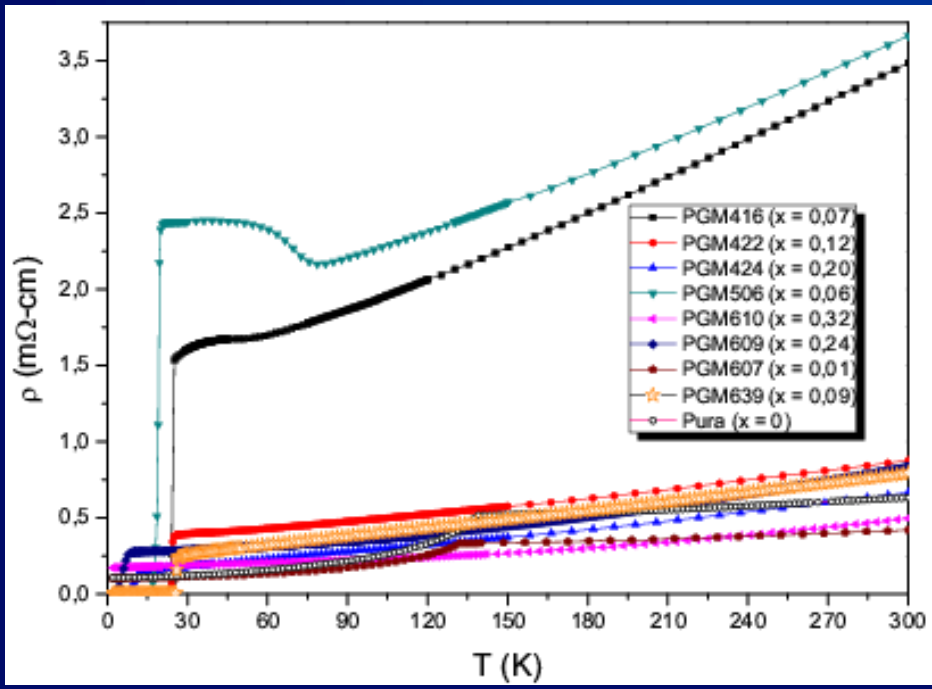
- Phase Diagram and Tcs for OPD samples

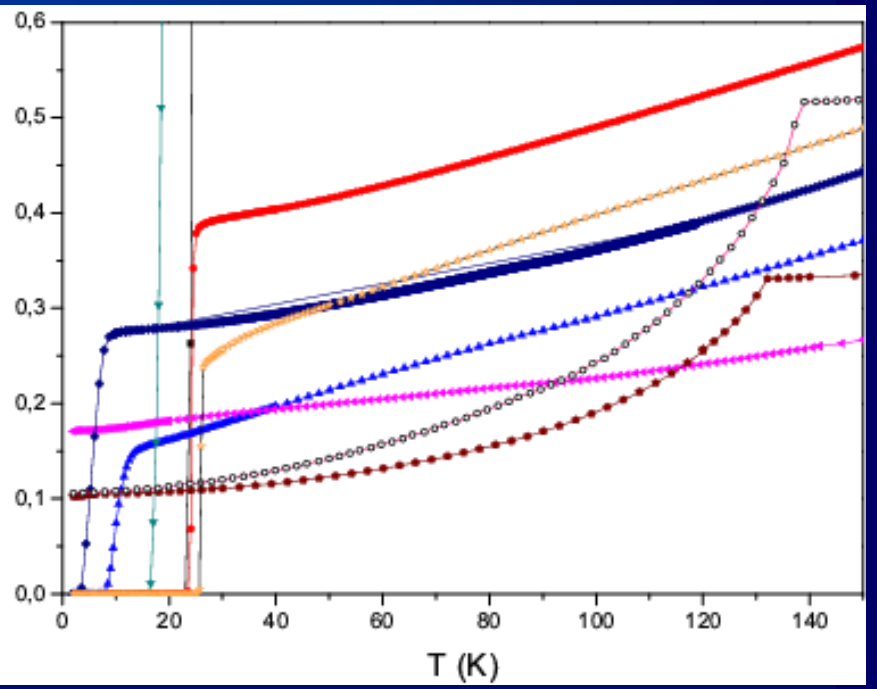




Results – In-flux grown samples

- Resistividade DC
  - Doping evolution
  - Residual Resistivity : $0.1 \leq \rho \leq 0.7$ mΩ-cm, similar and/or smaller than self-flux samples (PRB **82,** 220504 e 064501 (2010)), but no need of As excess!

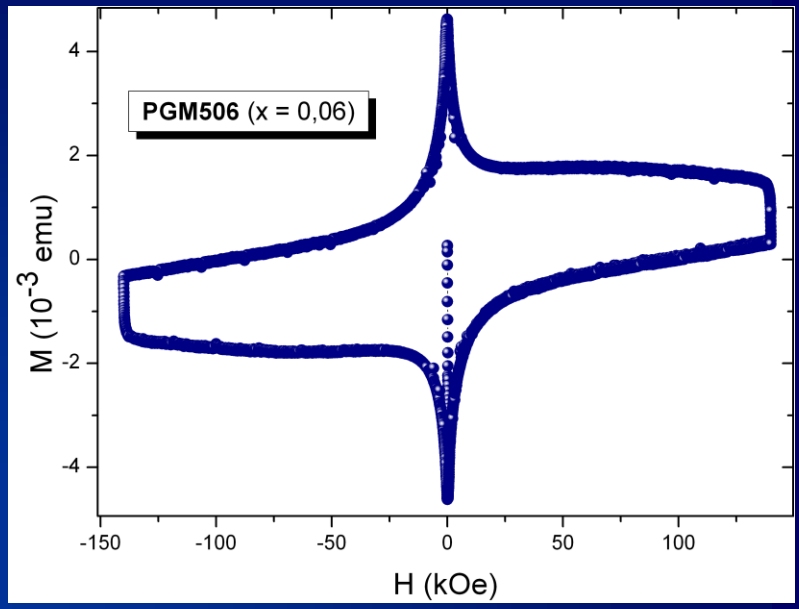




Results – In-flux grown samples

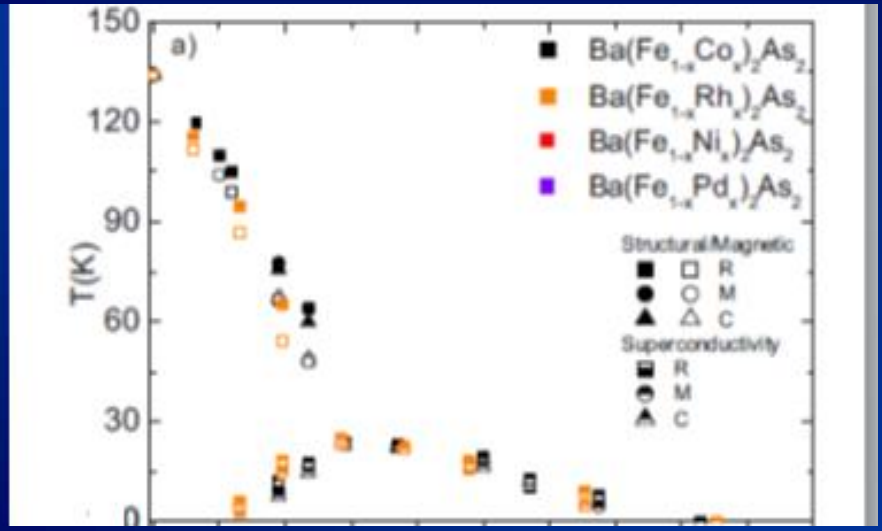
- J_c's similar or smaller than self-flux samples ($J_c \sim 10^6$ A/cm²) (arXiv:0911.5582v1; PRB **80**, 174517 (2009); Appl. Phys. Lett. **94**, 062511 (2009); arXiv:1011.5721v1)

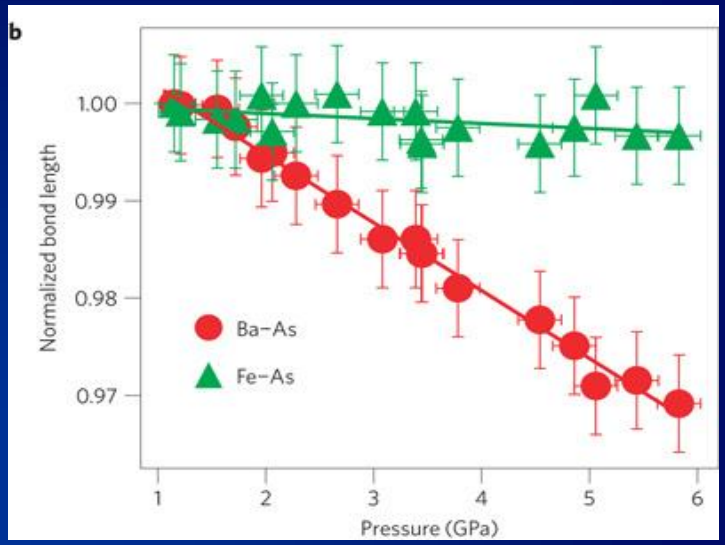
- High quality samples!



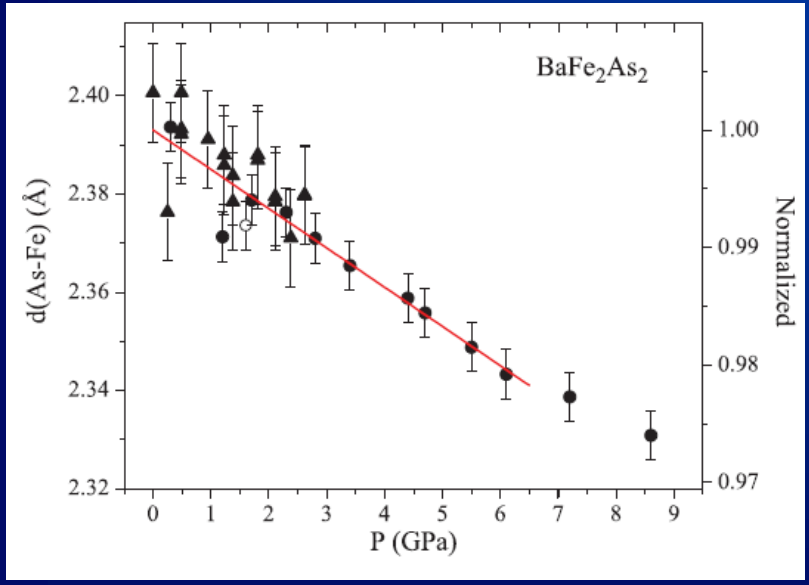
Samples	J_c (2 K, H = 0) (A/cm²)	H_{c1} (Oe)	~λ (nm)
PGM505 (x = 0,16)	$2{,}9\ 10^6$	~7500	---
PGM506 (x = 0,06)	$7{,}5\ 10^4 – 6{,}2\ 10^5$	~240	165
PGM507 (x = 0,18)	$5{,}8\ 10^3$	~360	---
PGM424 (x = 0,20)	---	~250	145
PGM416 (x = 0,07)	$1{,}1\ 10^4$	~2000	48

Results – EXAFS – Decrease of d(Fe-As)





Kimber S.A.J. et al., Nat. Mat. **8**, 471 (2009)

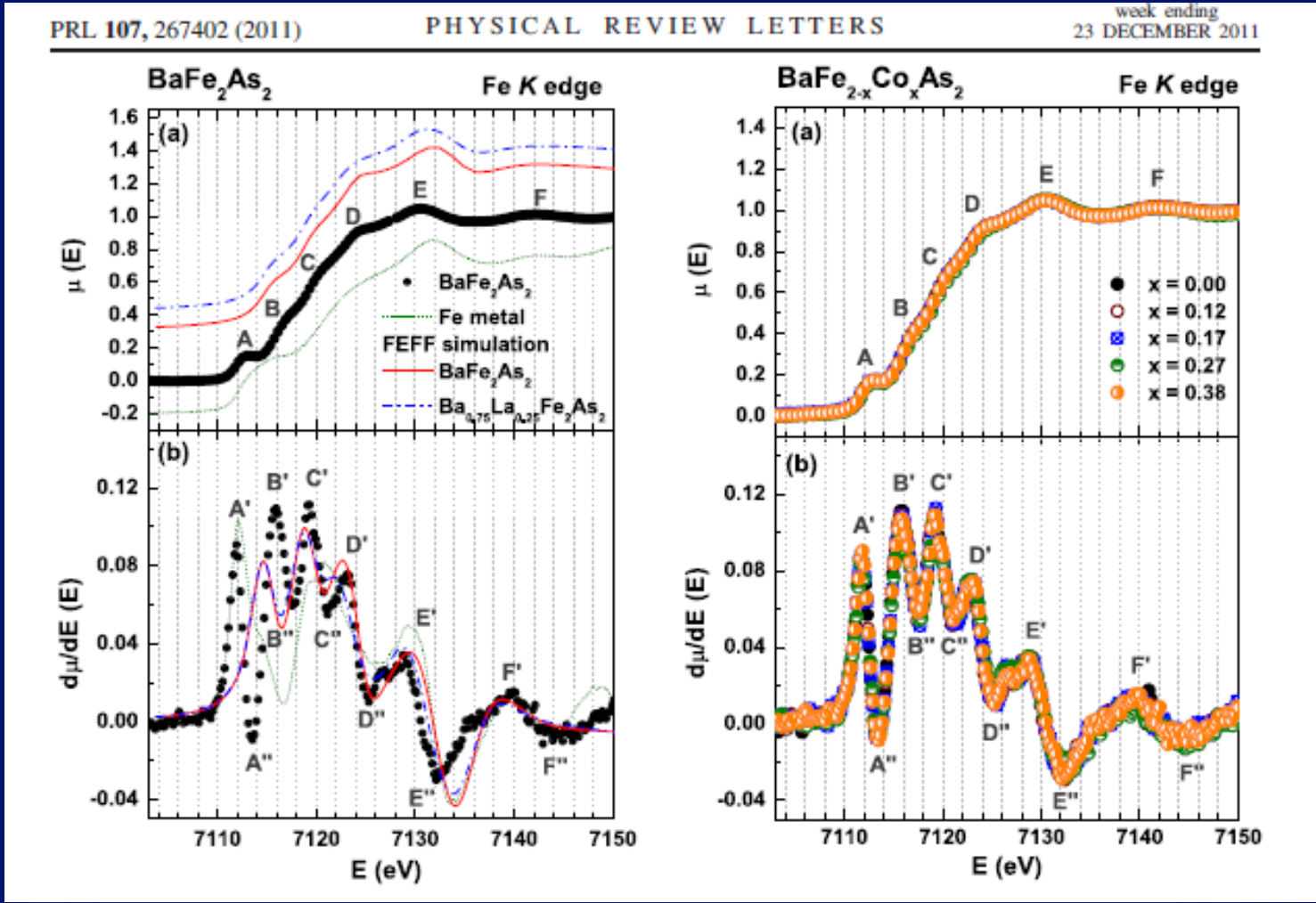


E. Granado..,P. G. Pagliuso et al. PRB **83**, 184508 (2011)

TABLE I. Refined As-M (M = Fe, Co) distances and Debye-Waller factors obtained from the fits of x-ray absorption fine structure data at the As K edge at ambient pressure. Errors given in parentheses are statistical only, and are defined as the standard deviation of the results obtained from repeated measurements under identical conditions.

	$T = 2$ K	$T = 30$ K	$T = 298$ K
$BaFe_2As_2$			
d(As-Fe) (Å)	2.3915(12)	2.3914(7)	2.3985(14)
σ^2 (Å^2)	0.00266(12)	0.00250 (7)	0.00465(11)
$Ba[Fe_{0.937}Co_{0.063}]_2As_2$			
d[As-(Fe,Co)] (Å)	2.3833(12)	2.3838(9)	2.3951(12)
σ^2 (Å^2)	0.00262(12)	0.00268(9)	0.00466(9)
$Ba_{0.85}K_{0.15}Fe_2As_2$			
d(As-Fe) (Å)	2.3865(15)	2.3900(12)	2.3955(9)
σ^2 (Å^2)	0.00242(15)	0.00248(12)	0.00466(7)

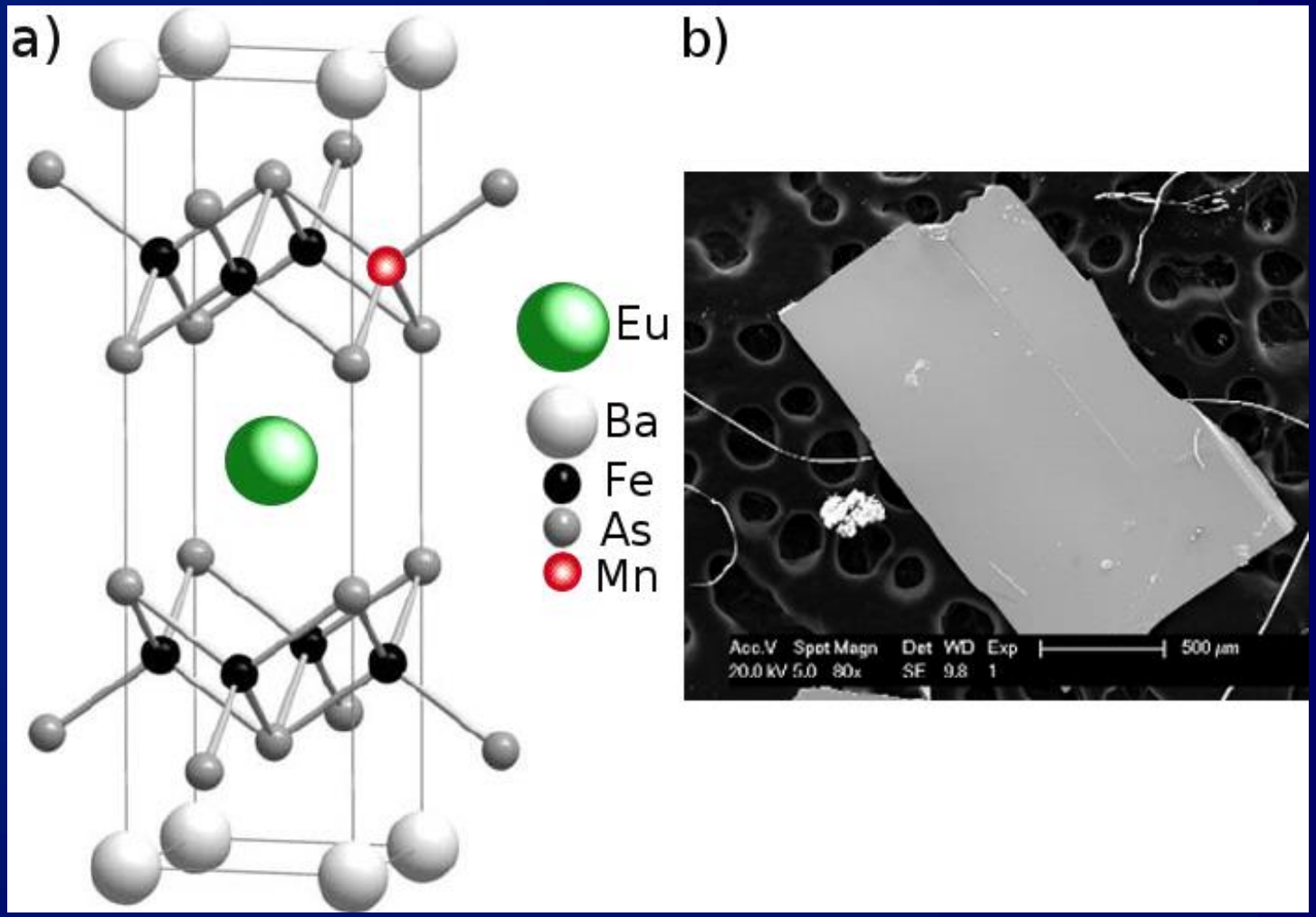
Results – XANES on In-flux grown samples



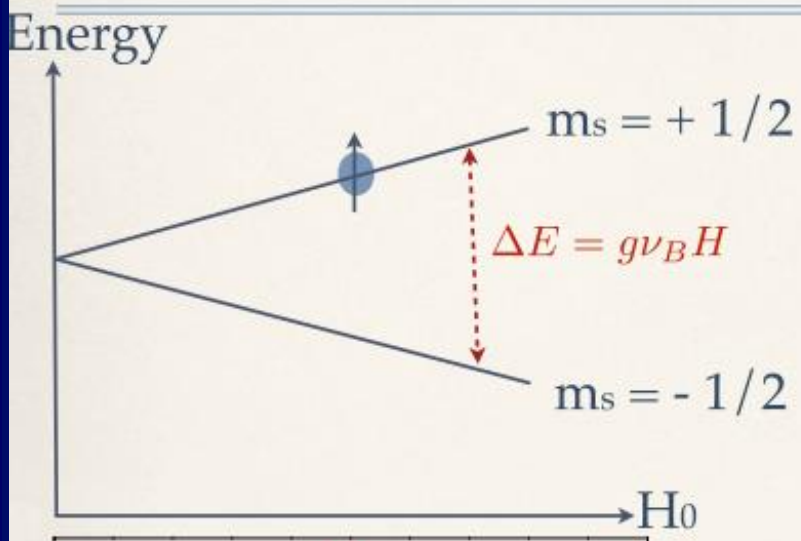
E. M. Bittar … P.G. Pagliuso et al. PRL **107**, 267402 (2011).

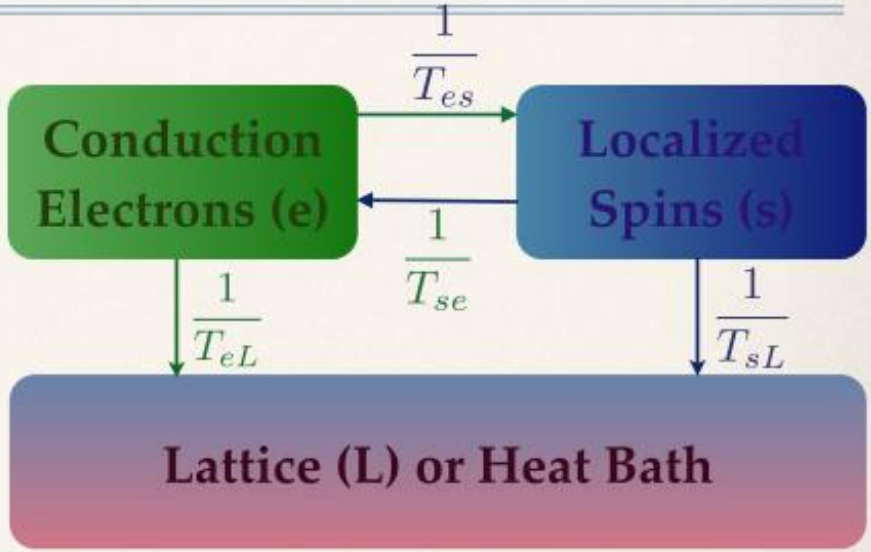
Fe K absorption edge is completely unaltered by Co-substitution!

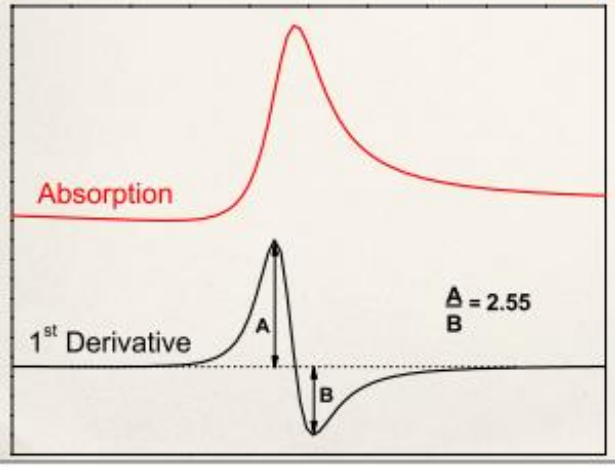
Site specific ESR on Fe-based Intermetallic Compounds

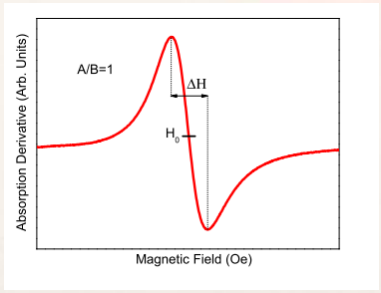


Electron spin resonance (ESR): Relaxation Mechanism in Metals









$H_o \rightarrow$ g-value or IT

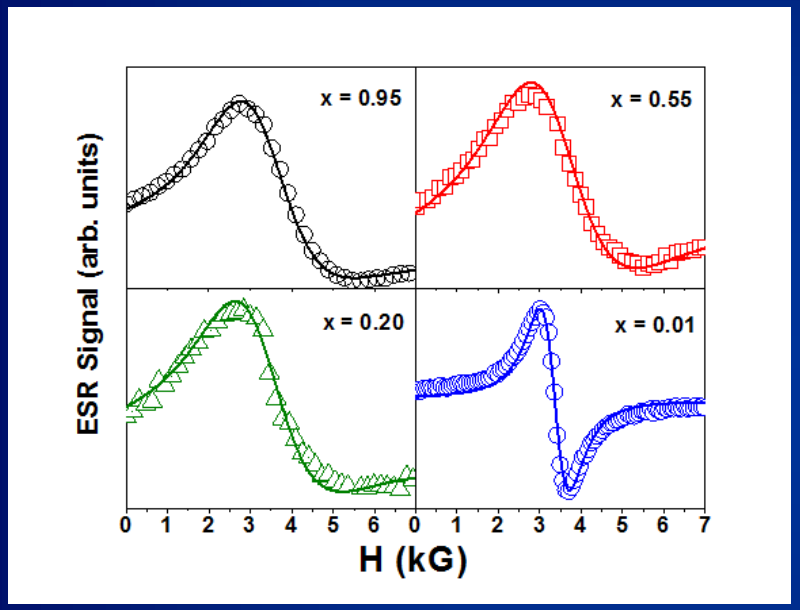
$\Delta H \rightarrow$ relaxation and inhomogeneity

$I \rightarrow \chi$ (T)

J. Korringa. Physica 16, 601 (1950).

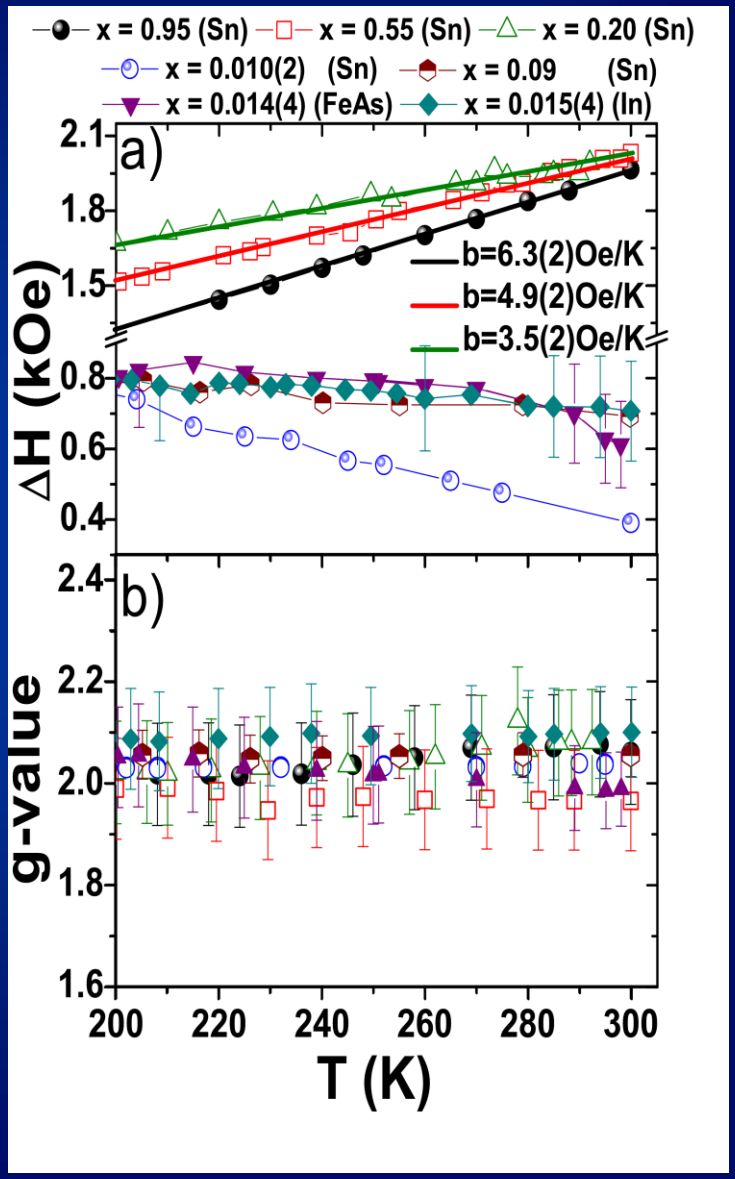
Results – ESR – $Ba_{1-x}Eu_xFe_2As_2$

- Decreasing Korringa rate.
- T and x independent g-factor.



$$\Delta g = J_{fs}(\mathbf{0})\ \frac{\eta\,(E_F)}{1-\alpha}$$

$$\frac{d\,(\Delta H)}{dT} = \frac{\pi k}{g\mu_B} < J_{fs}^2(\mathbf{q}) > \eta^2\,(E_F)\ \frac{K(\alpha)}{(1-\alpha)^2}$$



P. F. S. Rosa…P.G. Pagliuso et al. PHYSICAL REVIEW B **86**, 165131 (2012)

ESR Results

q-dependence of the exchange interaction

$b_{calculated} > b_{measured}$

$$\Delta g = J_{fs}(\mathbf{0})\frac{N(E_F)}{1-\alpha}$$

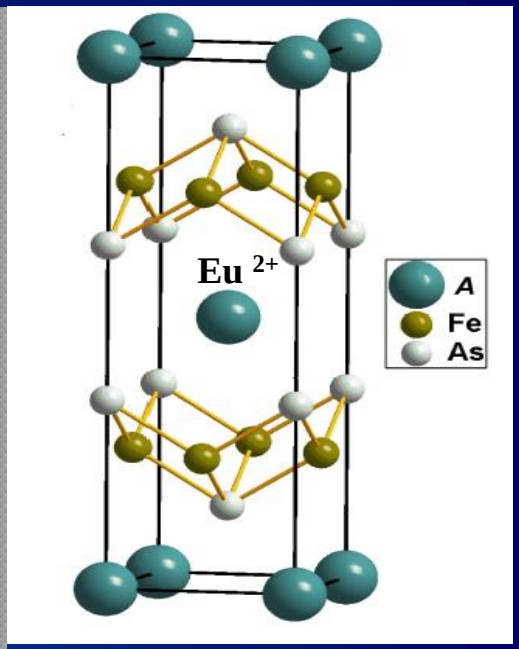
Stoner enhancement factor

$$\frac{d(\Delta H)}{dT} = \frac{\pi k}{g\mu_B}\langle J_{fs}^2(\mathbf{q})\rangle N(E_F)^2 \frac{K(\alpha)}{(1-\alpha)^2}$$

Decreasing Korringa rate (b) → Decreasing $\langle J_{fs}(\mathbf{q})\rangle$

- 3d Fe bands are becoming more anisotropic (less s-like) and are, in average, further away from the Eu sites.

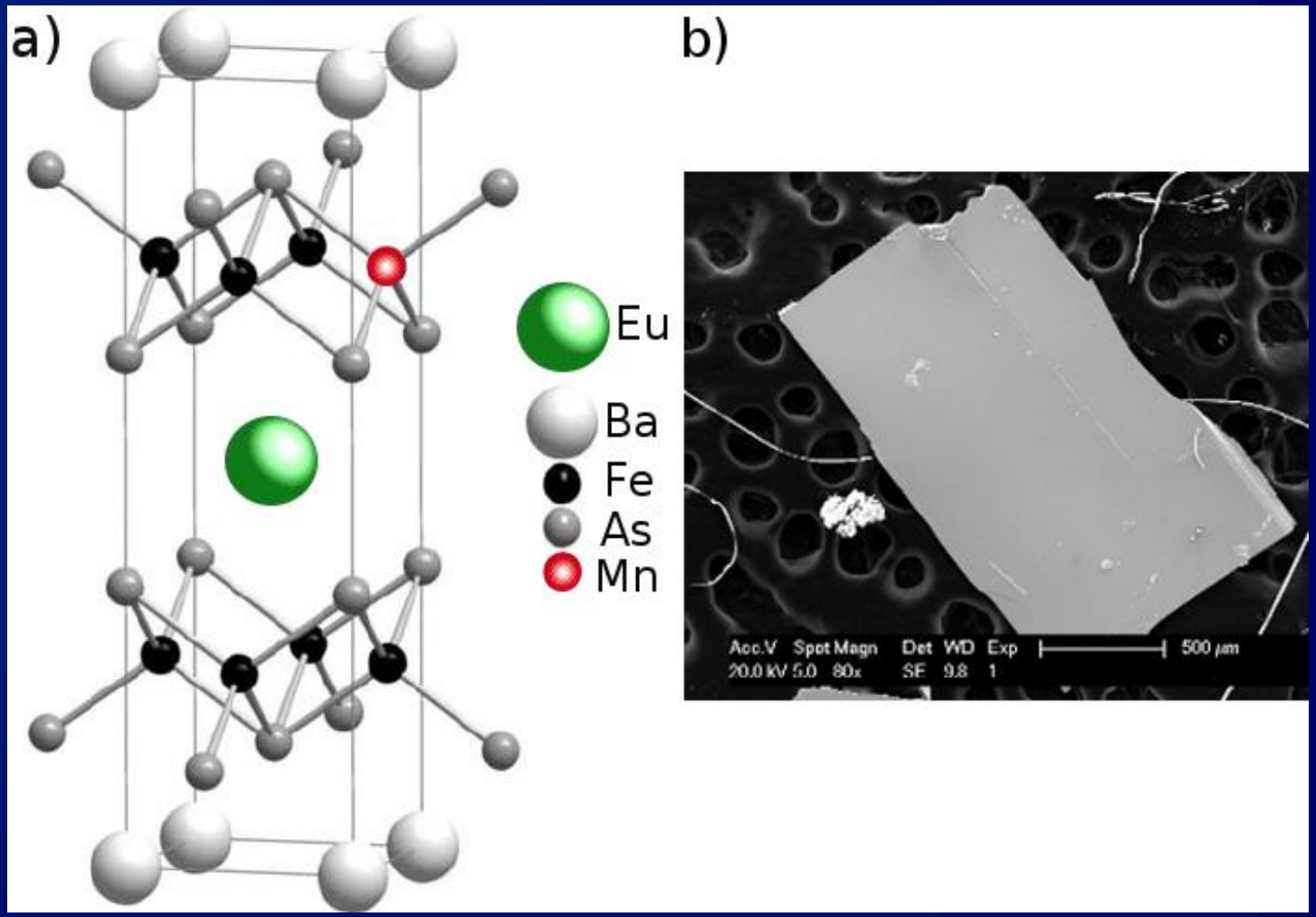
Wednesday, April 24, 2013



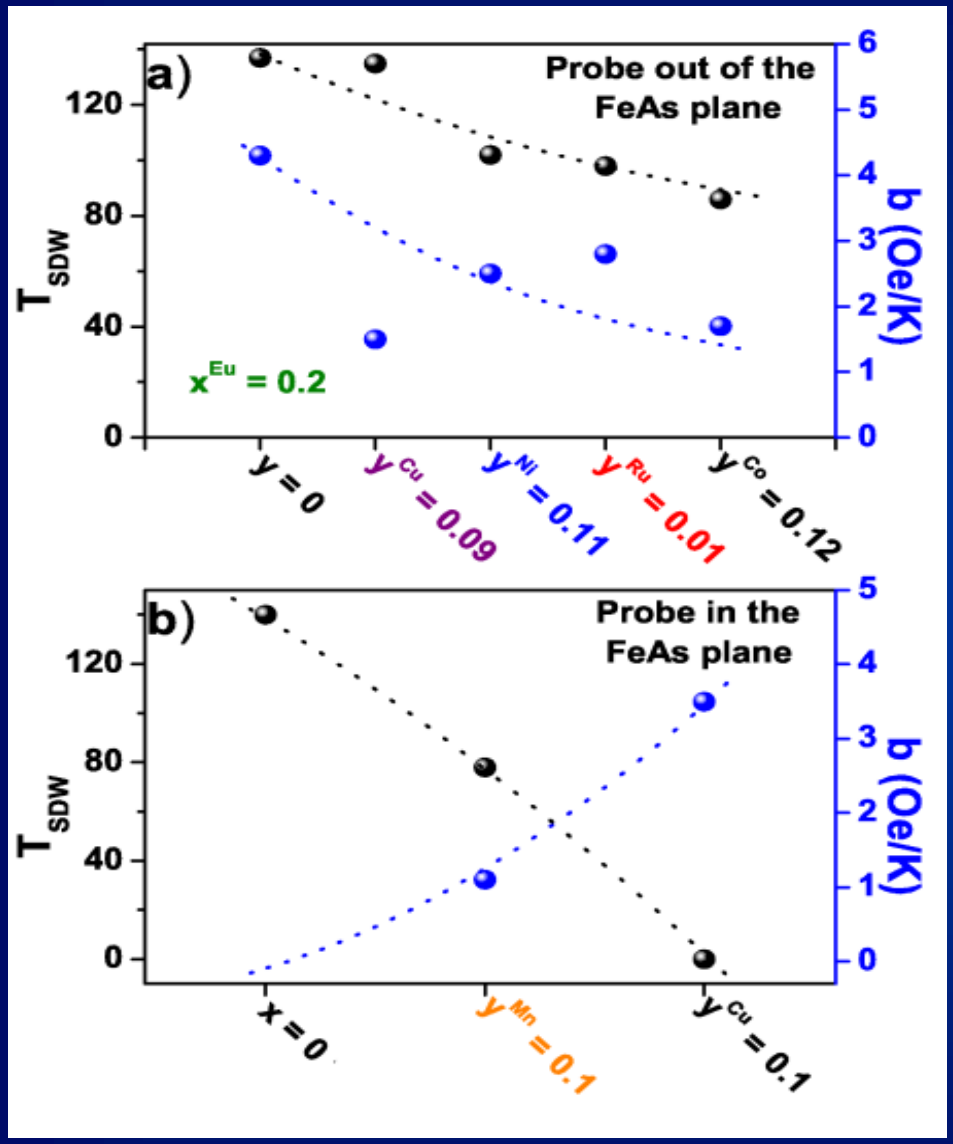
P. F. S. Rosa…PG. Pagliuso et al. PHYSICAL REVIEW B **86**, 165131 (2012)

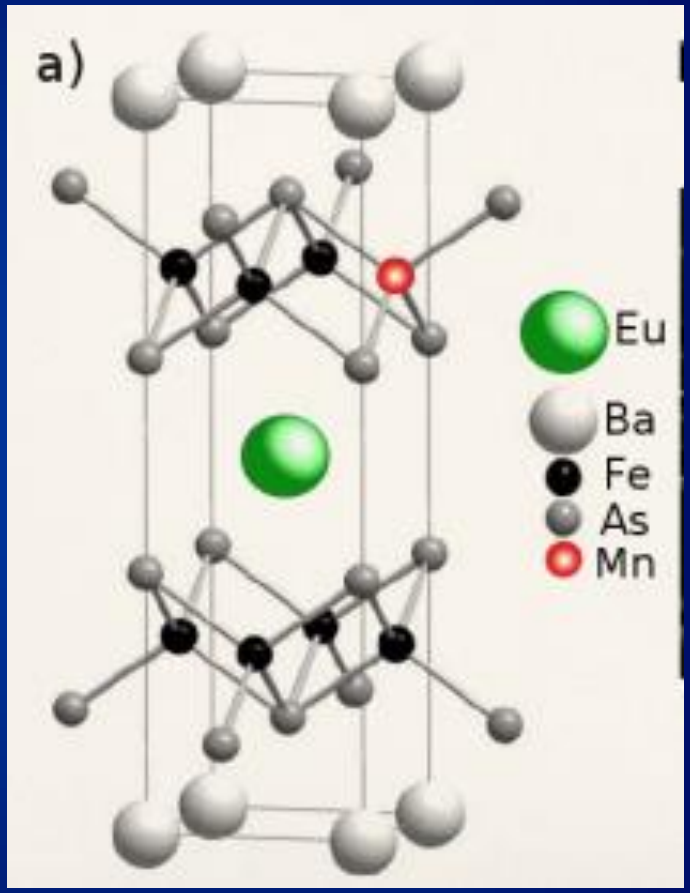
P. F. S. Rosa…PG Pagliuso et al. Nature-Scientific Reports 4, 6543 (2014) ibid Nature-Scientific Reports 4, 6252 (2014)

Site specific ESR on Fe-based Intermetallic Compounds



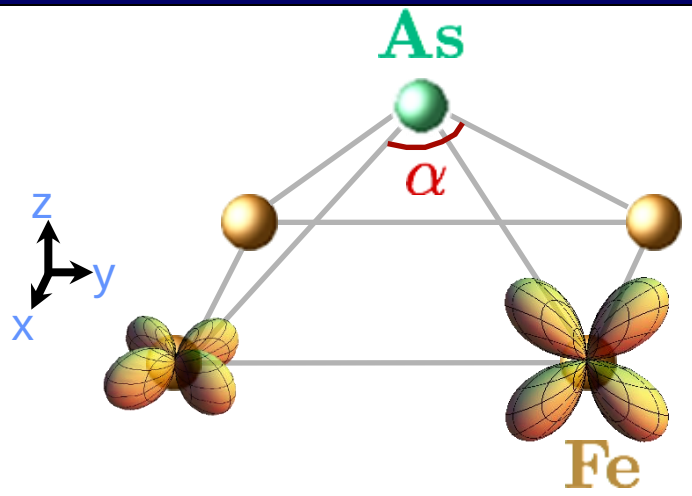
ESR – Results – Site Specific

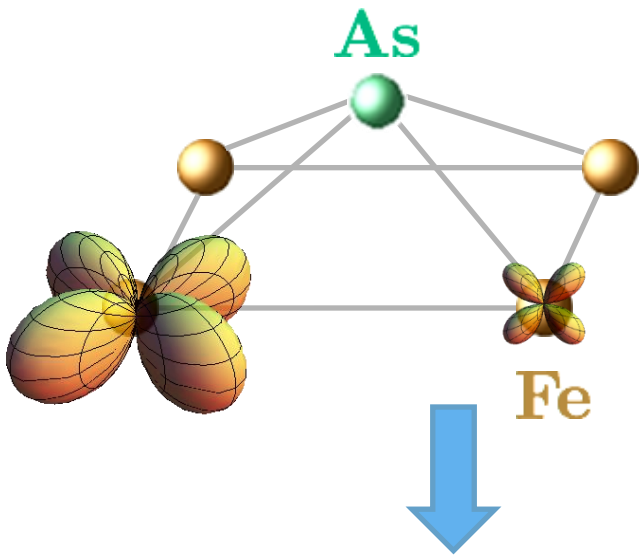




P. F. S. Rosa…PG. Pagliuso et al. PHYSICAL REVIEW B **86**, 165131 (2012)

P. F. S. Rosa…PG Pagliuso et al. Nature-Scientific Reports 4, 6543 (2014) ibid Nature-Scientific Reports 4, 6252 (2014)





Smaller $d_{Fe\text{-}As}$ with chemical substitution, hydrostatic pressure and magnetic field in the plane!

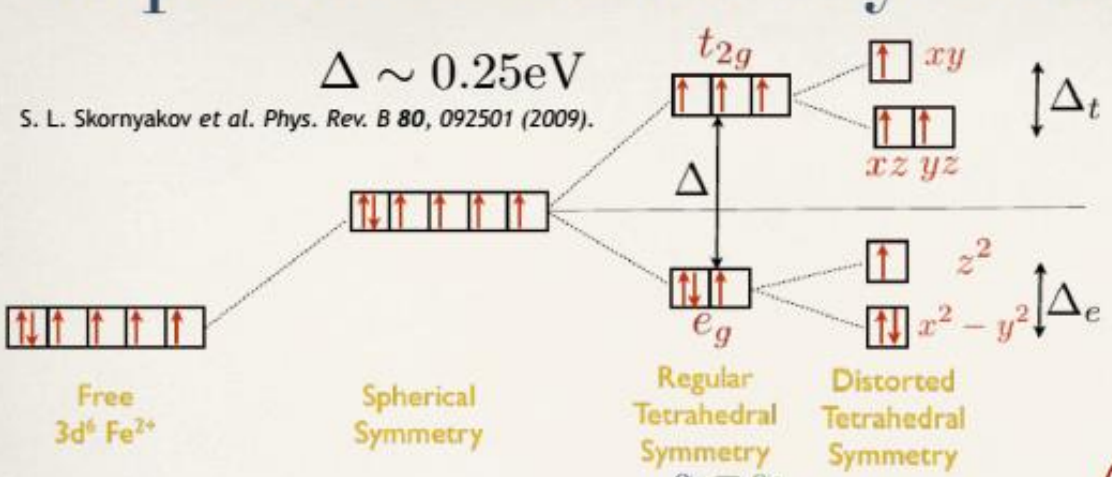
xy occupation increases

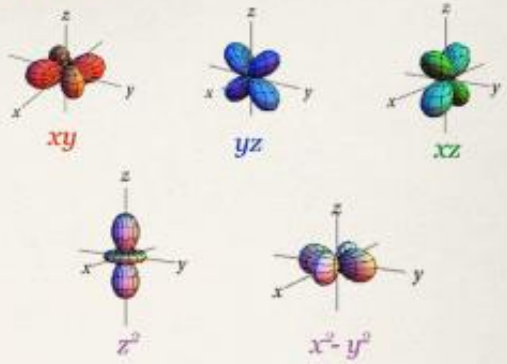
SDW Phase vanishes

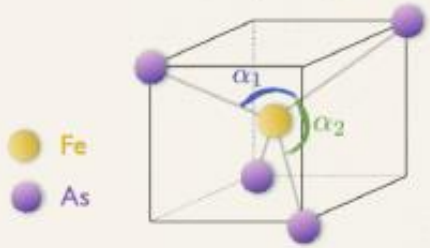
Importance of the Crystal Field

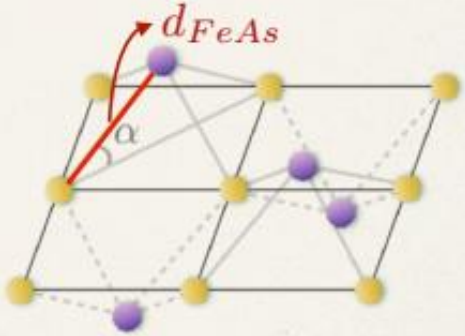
$\Delta \sim 0.25\text{eV}$

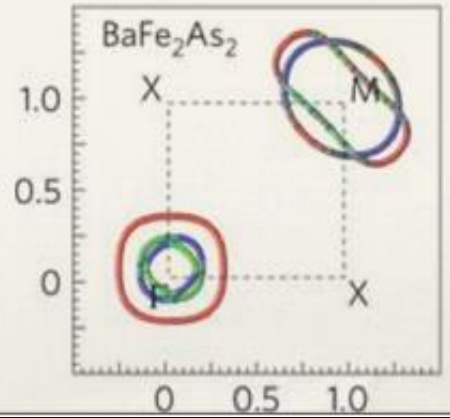
S. L. Skornyakov *et al. Phys. Rev. B* **80**, 092501 (2009).



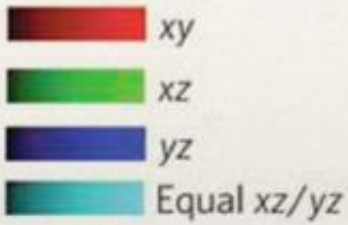




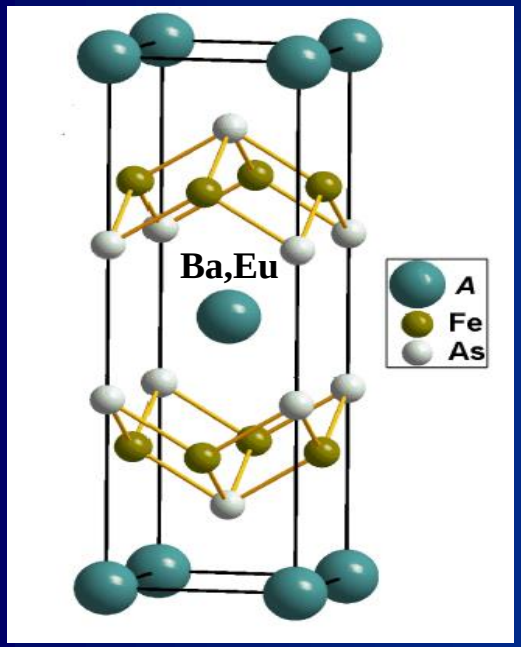




Z. P. Yin *et al. Nature Materials*, **10** 932-935 (2011).



Quantum Oscillations Results



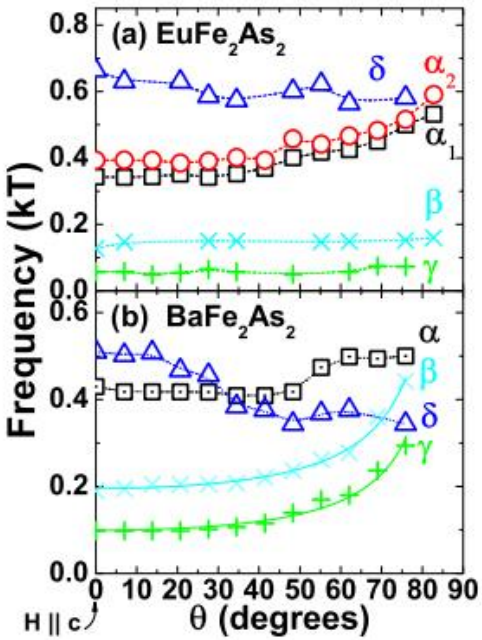


FIG. 4. (Color online) Angular field dependence (anisotropy) of the observed QO frequencies of (a) $EuFe_2As_2$ (Eu122) and (b) $BaFe_2As_2$ (Ba122).

The calculated effective masses m^*_{DFT} also display agreement with the experimental data, as shown in Table I. We note that an ideal quantitative agreement of effective masses is often

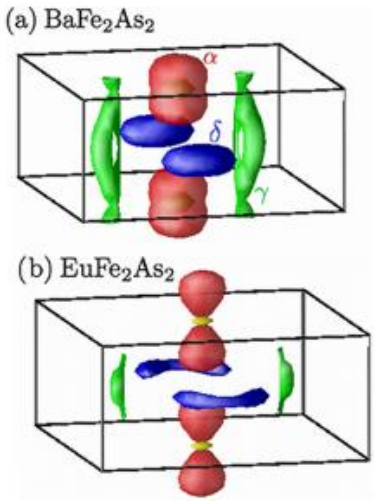


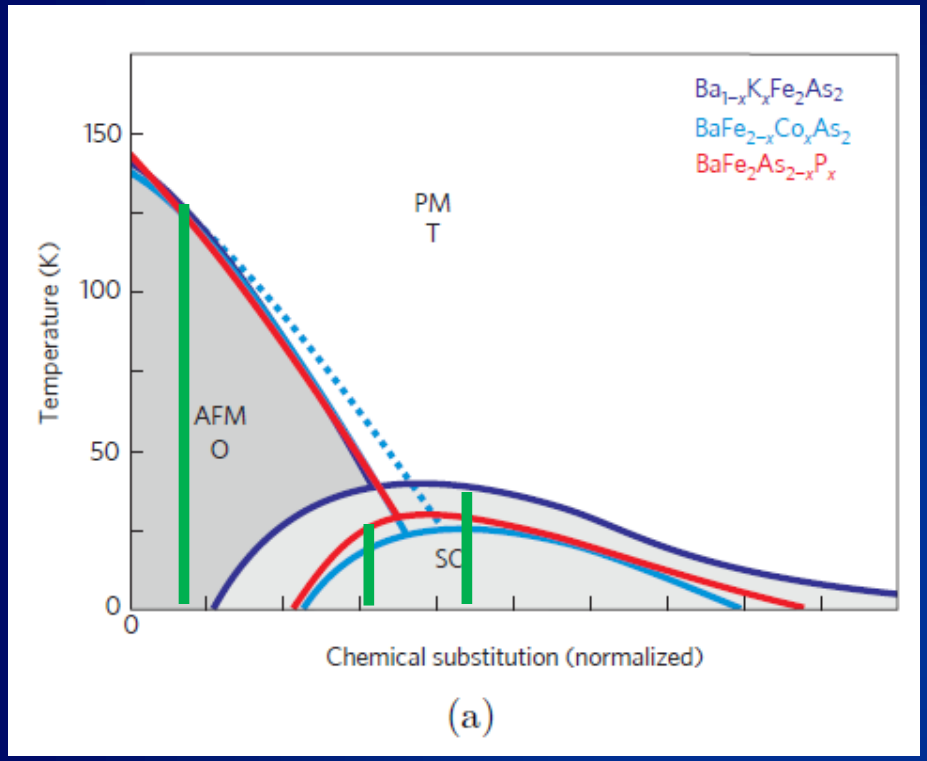
FIG. 5. (Color online) The shifted Fermi surfaces of (a) $BaFe_2As_2$ and (b) $EuFe_2As_2$ compounds in the magnetic phase. We identify the large hole sheet (red) as the α pocket, the crescent electron sheet (blue) as the δ pocket, and the tubelike electron pocket (green) as the γ pocket.

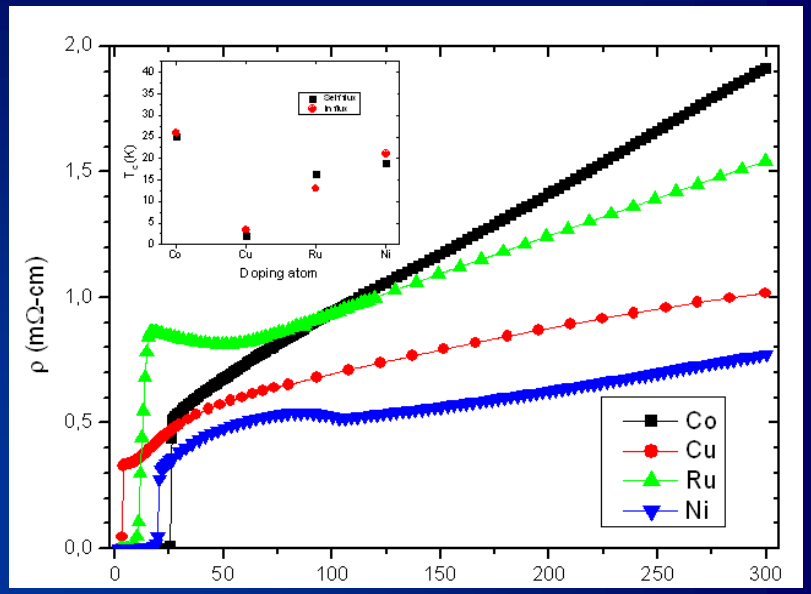
195146-4

- Eu122 displays a much more isotropic and three-dimensional-like FS when compared with Ba122.
- Results suggest an anisotropic contribution of the Fe 3d orbitals to the FS in Ba122. We speculate that this orbital differentiation may be responsible for the suppression of the SDW phase in the FeAs-based compounds.

P. F. S. Rosa…PG. Pagliuso et al. PHYSICAL REVIEW B 90, 195146 (2014)

Combined external pressure and chemical substitution results





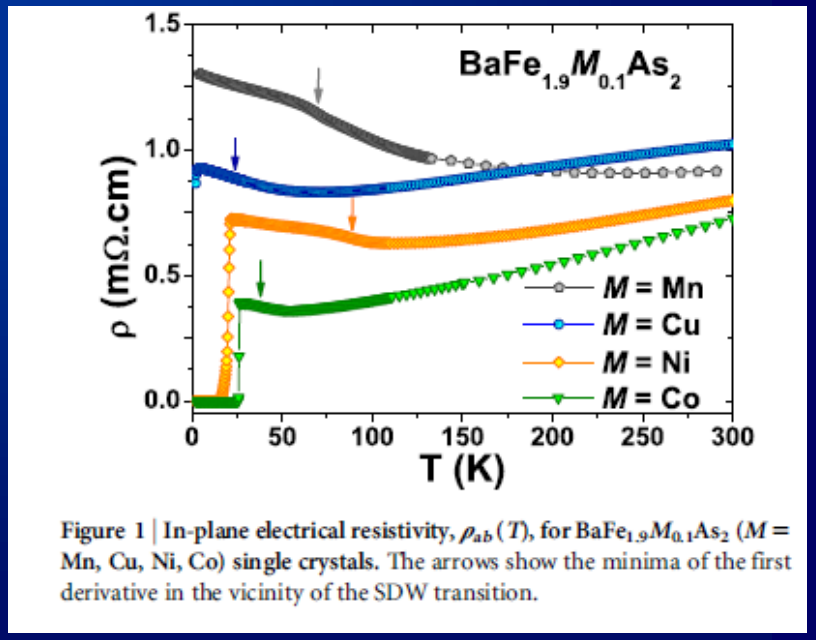


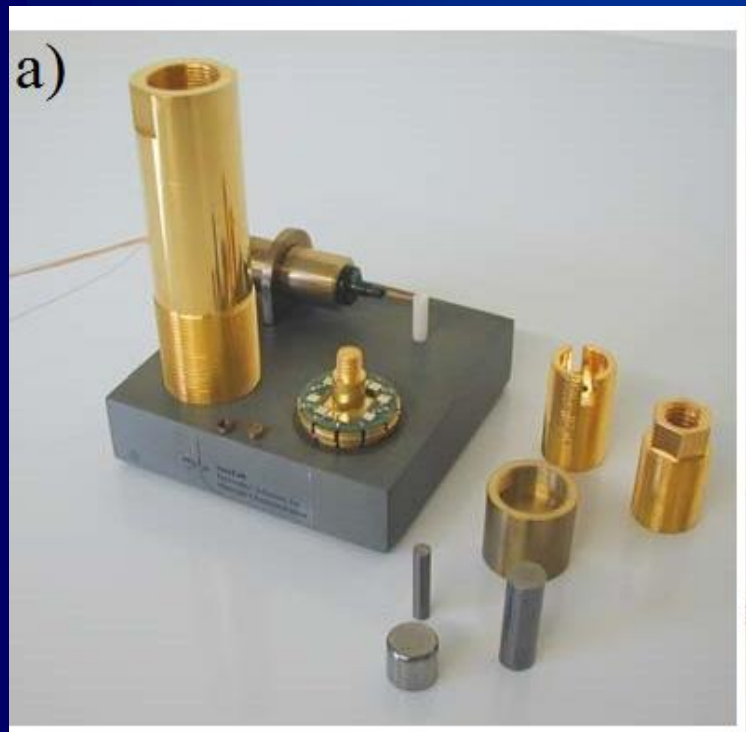
Figure 1 | In-plane electrical resistivity, $\rho_{ab}(T)$, for $BaFe_{1.9}M_{0.1}As_2$ (M = Mn, Cu, Ni, Co) single crystals. The arrows show the minima of the first derivative in the vicinity of the SDW transition.

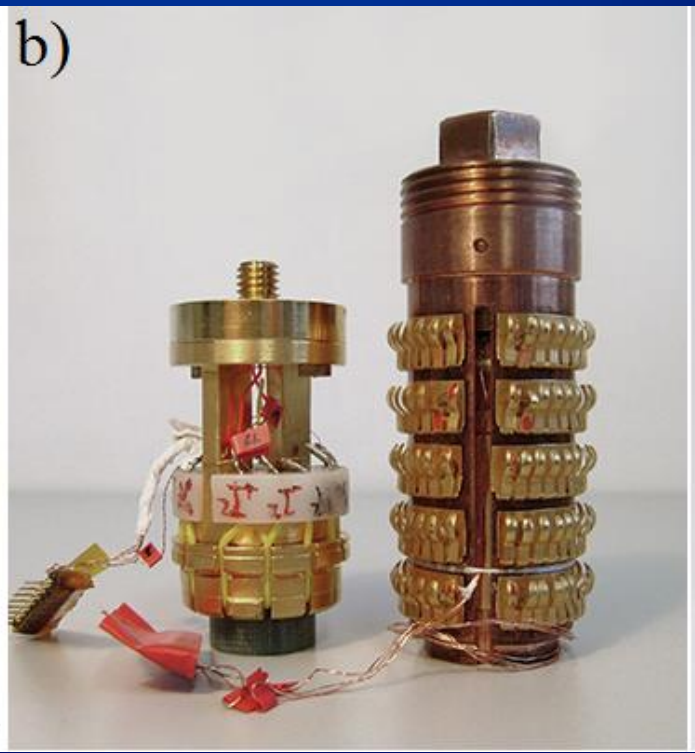
- Different P-dependences are expected as a function of x for substituted samples!

P. F. S. Rosa…PG. Pagliuso et al. PHYSICAL REVIEW B 90, 195146 (2014)

Experimental Details

- In-plane electrical resistivity under quasi-hydrostatic pressure:

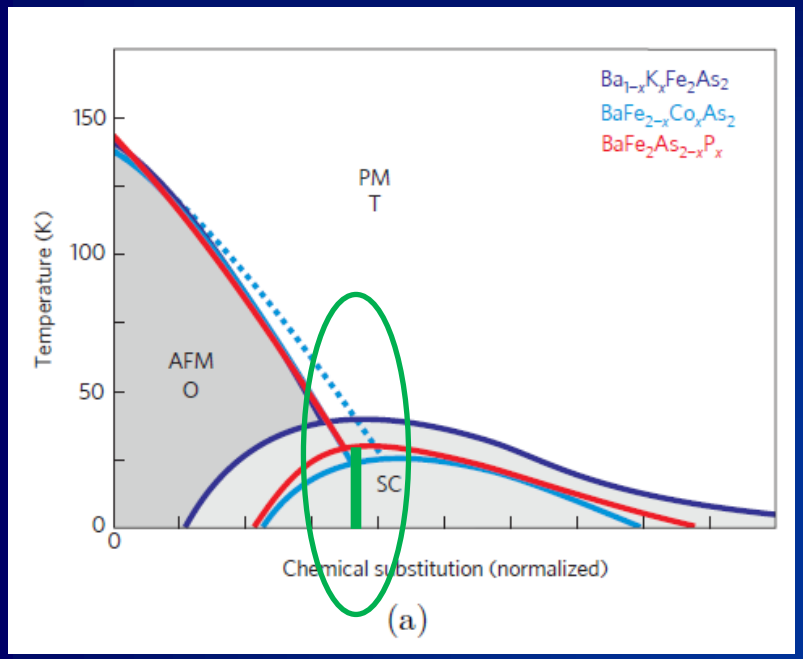


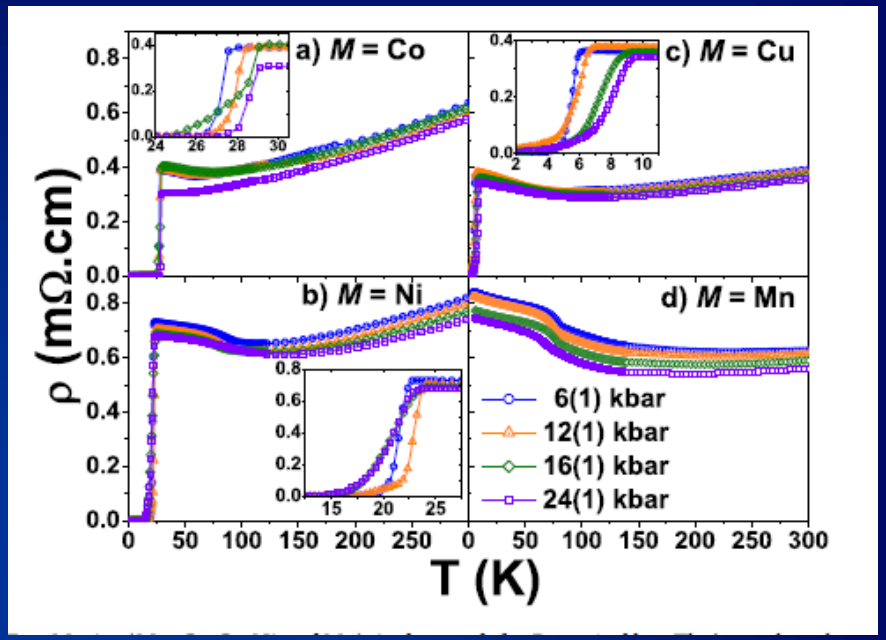


Extracted from Application Note on Pcell 15/30

Extracted from <http://www.dilatometer.info/>

Combined external pressure and chemical substitution results

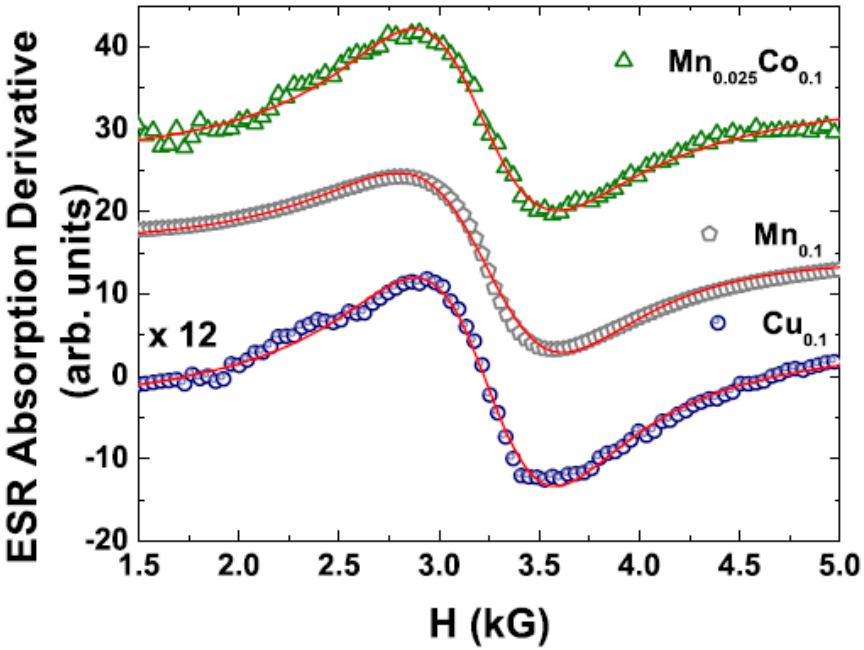




- Very weak (increasing) Tc-dependence as a function of P for M = Co or Ni for x near OPD region, as expected.
- Surprisingly, T_c increases more than a factor of two as a function of pressure for M = Cu.

P. F. S. Rosa…PG Pagliuso et al. Nature-Scientific Reports 4, 6252 (2014)

Combined external pressure and chemical substitution results



- Abrikosov-Gokov - IMPB

$$\left|\frac{\Delta T_c}{\Delta c}\right| = \frac{\pi^2}{8}\eta(E_F)\langle J^2(\mathbf{q})\rangle S(S+1),$$

- ESR Korringa rate.

$$\frac{d(\Delta H)}{dT} = \frac{\pi k}{g\mu_B} < J_{fs}^2(\mathbf{q}) > \eta^2(E_F)$$

Table I | Experimental and calculated parameters for $BaFe_{1-x}M_yAs_2$ (this work) and conventional SC (refs. [31, 37])

Sample	c (%)	g_{ESR}	$\lvert\Delta T_c^{exp}\rvert$ (K)	$T_{c,0}$ (K)	$\langle J^2(\mathbf{q})\rangle_{ESR}^{1/2}$ (meV)	$\langle J^2(\mathbf{q})\rangle_{AG}^{1/2}$ (meV)
$BaFe_{1.9}Cu_{0.1}As_2$	5	2.08(3)	22	26	1.2(5)	111(10)
$BaFe_{1.88}Mn_{0.12}As_2$	6	2.05(2)	≥26	26	0.7(5)	≥32(3)
$BaFe_{1.895}Co_{0.100}Mn_{0.005}As_2$	0.25	2.06(2)	10	26	0.8(5)	98(9)
$Lu_{1-x}Gd_xNi_2B_2C$	0.5	2.035(7)	≈0.3	15.9	10(4)	11(1)
$Y_{1-x}Gd_xNi_2B_2C$	2.1	2.03(3)	≈0.9	14.6	9(3)	10(1)
$La_{1-x}Gd_xSn_3$	0.4	2.010(10)	≈0.5	6.4	20(2)	≈20(2)

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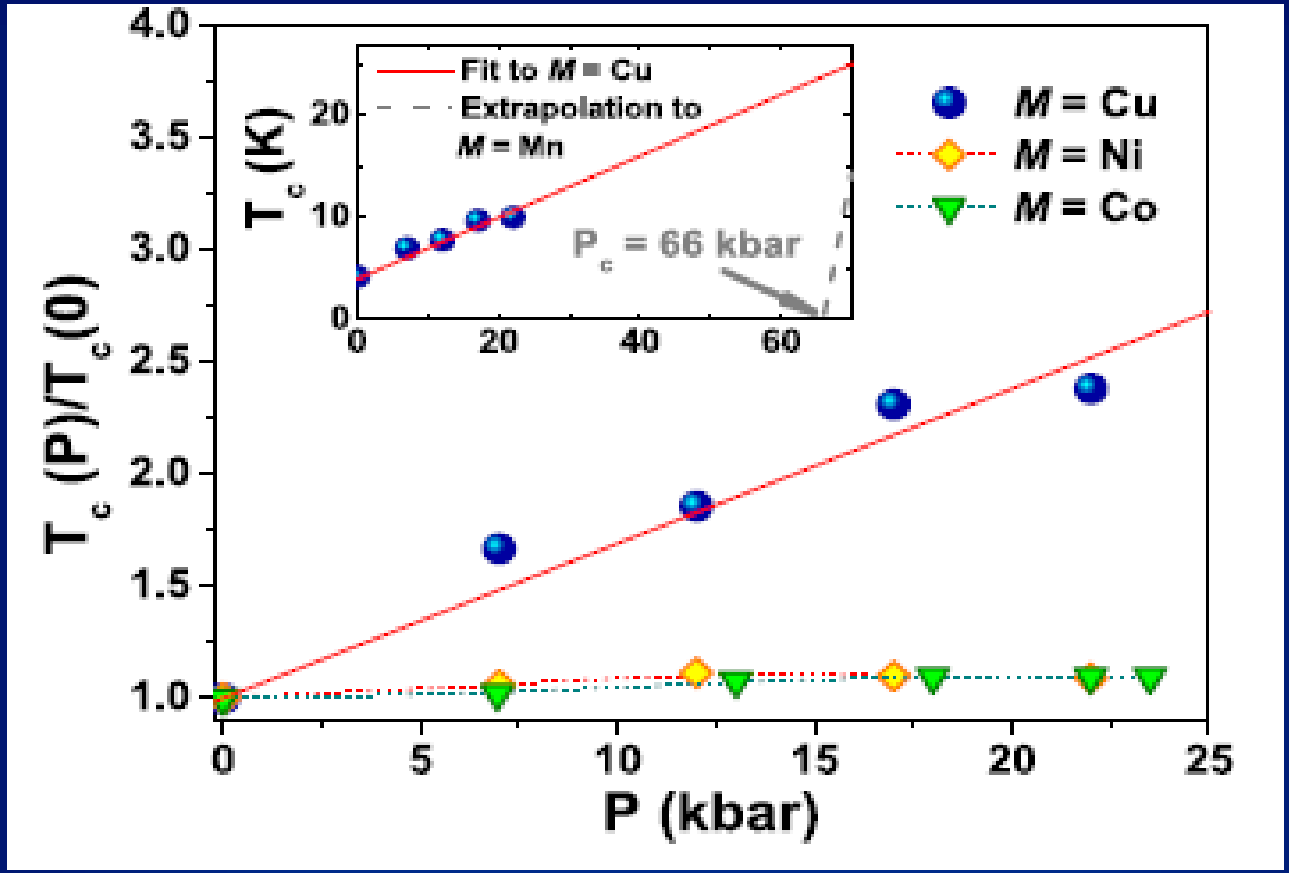
Combined external pressure and chemical substitution results



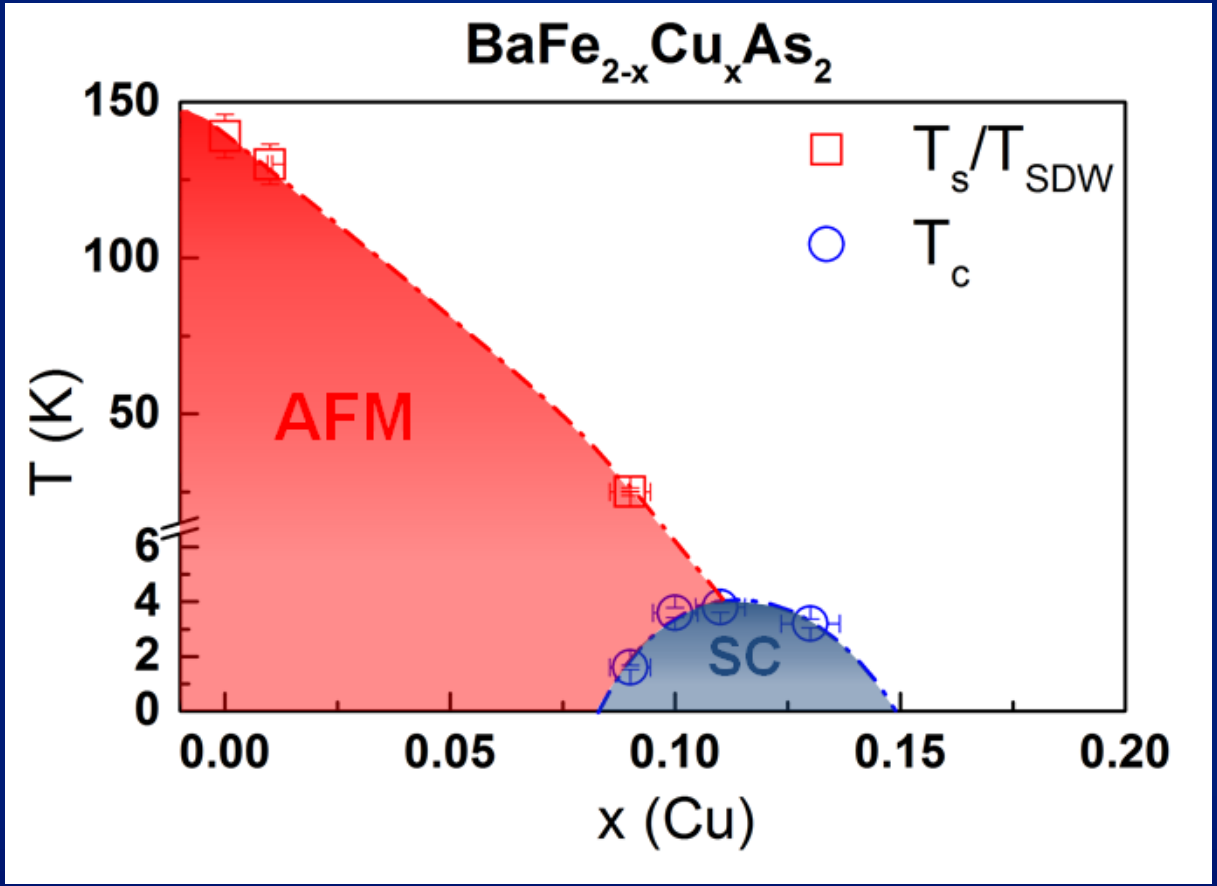
the SC domes. The linear fit for the M = Cu compound (solid line) was obtained from the phenomenological expression $\Delta T_c = S(S+1)(a - bP)$. Using the same expression and $S = 5/2$, we obtain the dashed line for the M = Mn compound.

- $BaCu_2As_2$ is a Pauli metal!

P. F. S. Rosa…PG Pagliuso et al. Nature-Scientific Reports 4, 6252 (2014)

Combined external pressure and chemical substitution results

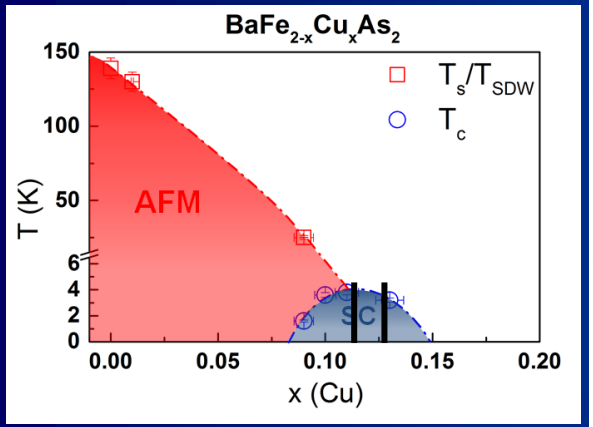
- Temperature-composition phase diagram:

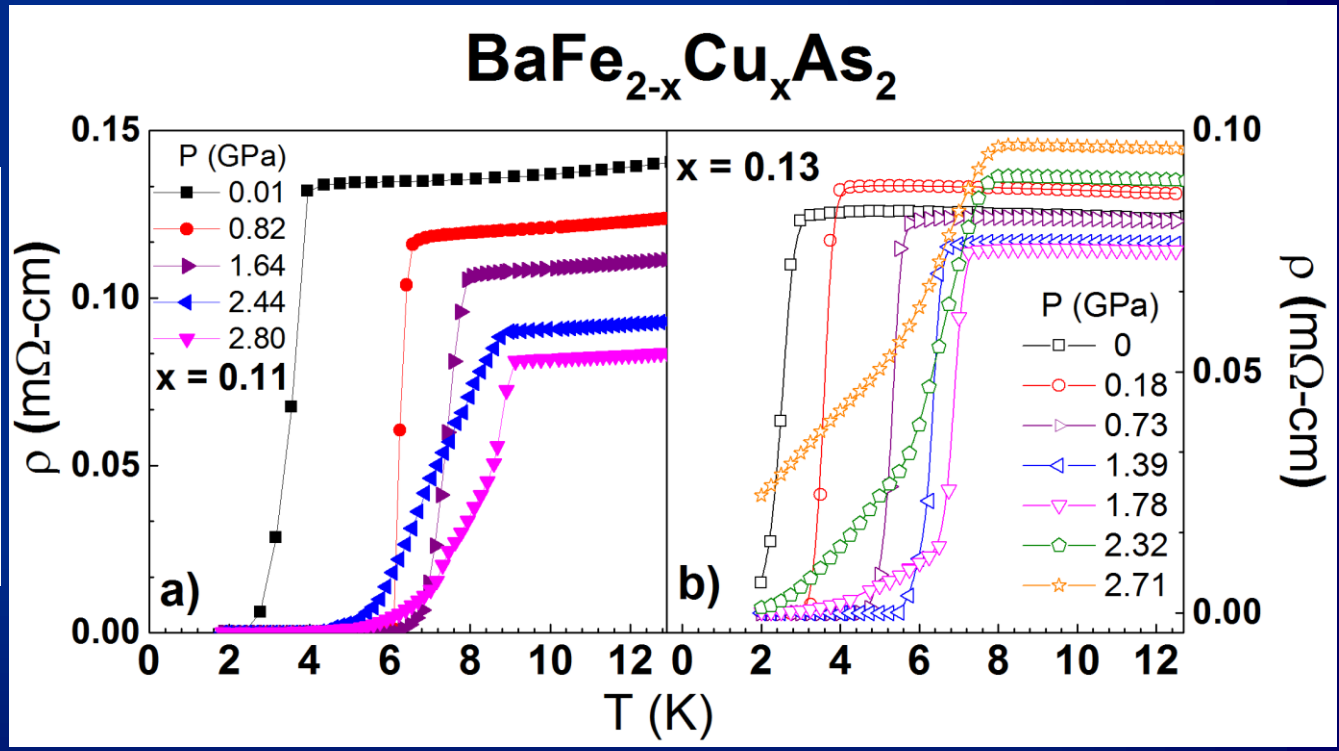


M M Piva …P.G. Pagliuso *et al J. Phys.: Condens. Matter* **27,** 145701 (2015)

Combined external pressure and chemical substitution results

- In-plane electrical resistivity at several pressures around the ODP limit:





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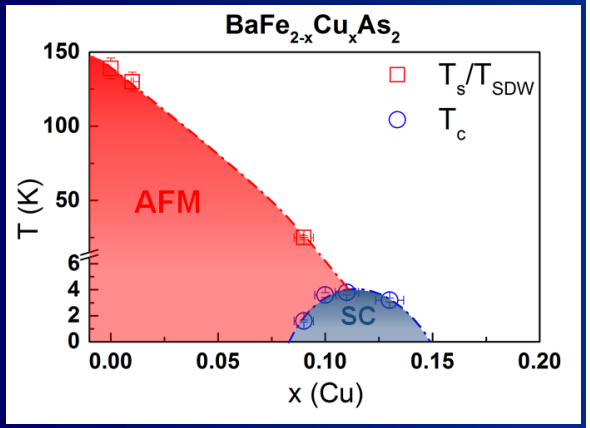
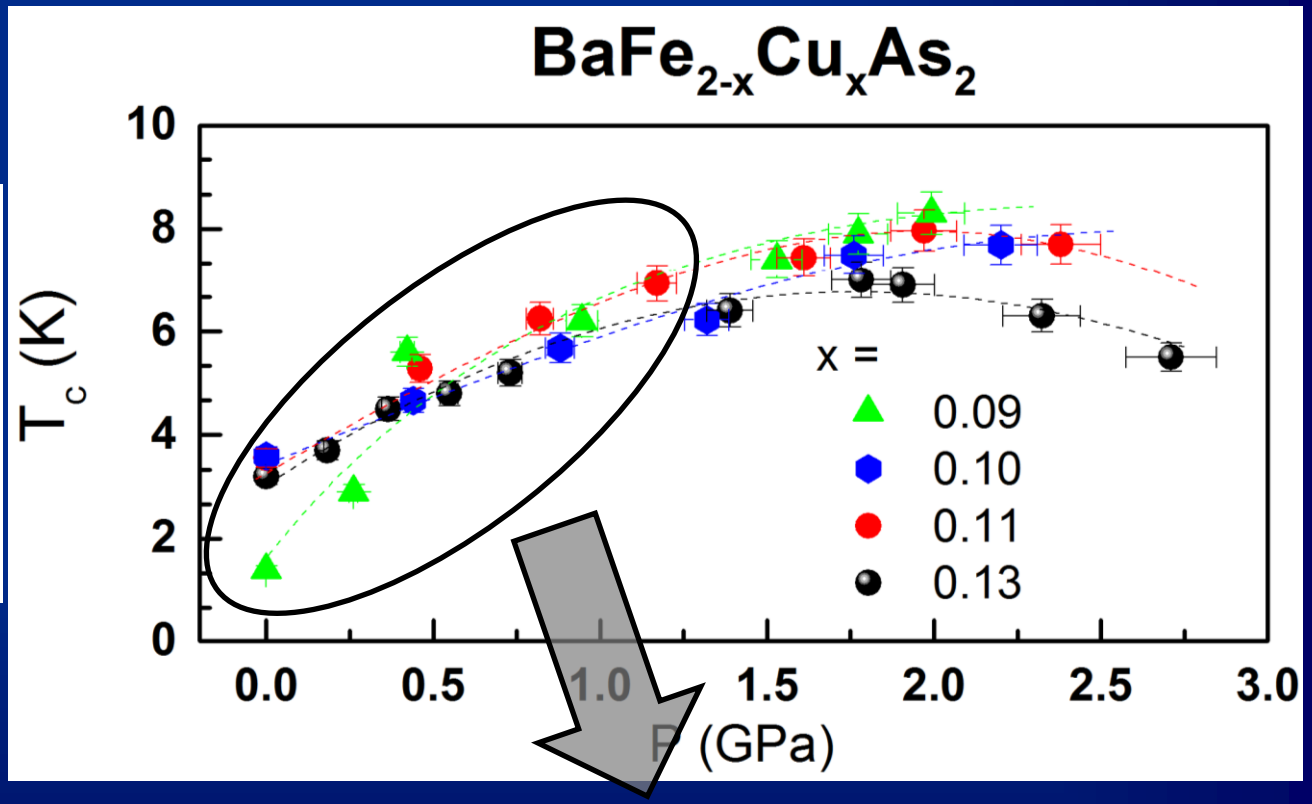
Combined external pressure and chemical substitution results

- Temperature-pressure phase diagram for Cu concentrations around the SC OPD:





Increase of T_c as a function of pressure

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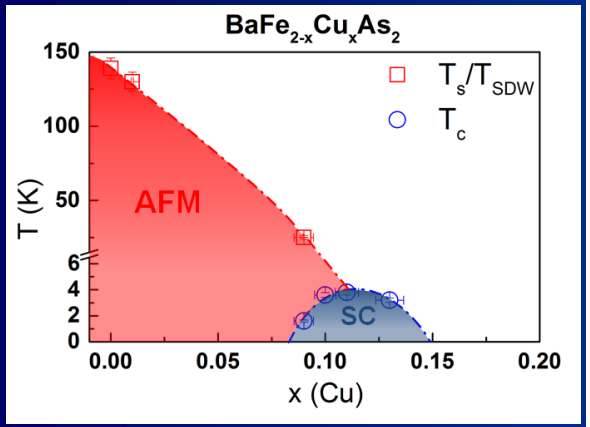
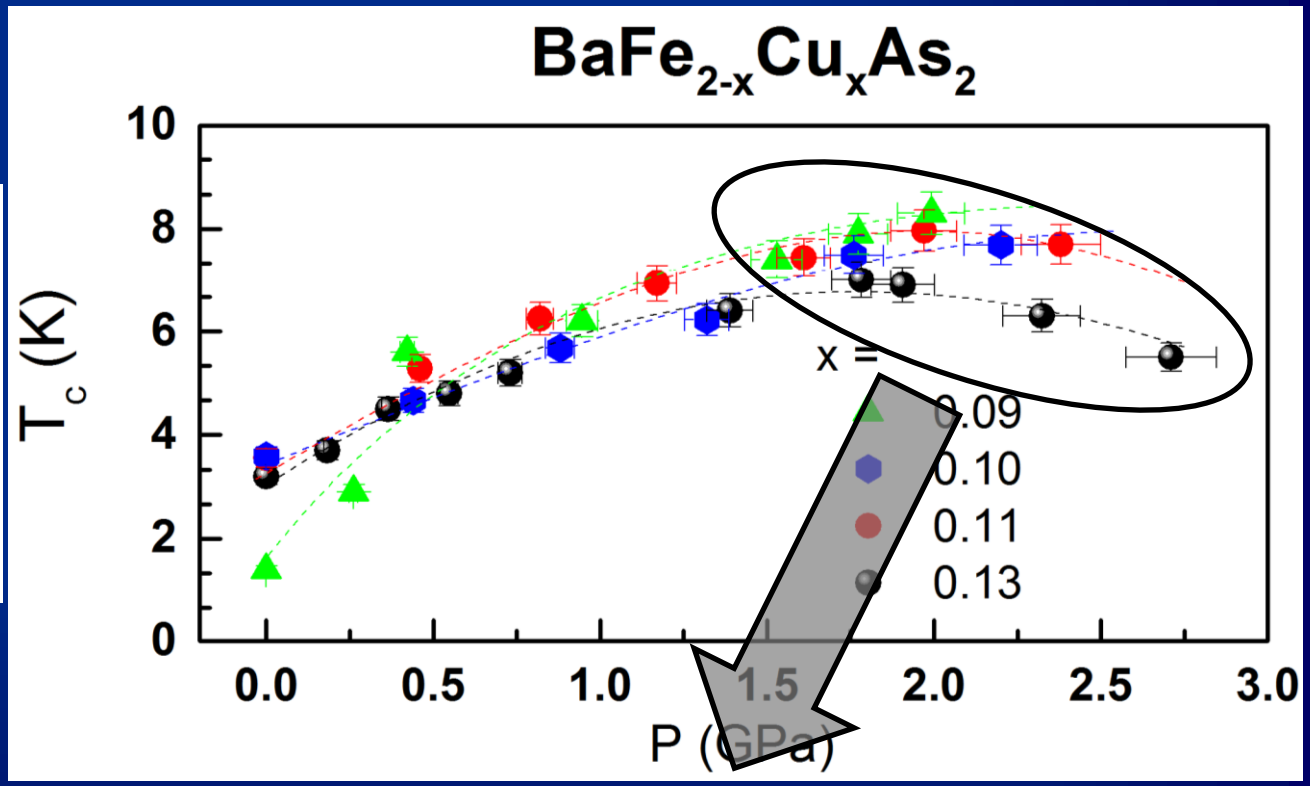
Combined external pressure and chemical substitution results

- Temperature-pressure phase diagram for almost all the Cu concentrations of the SC dome:





Tendency to saturation and decreasing of T_c

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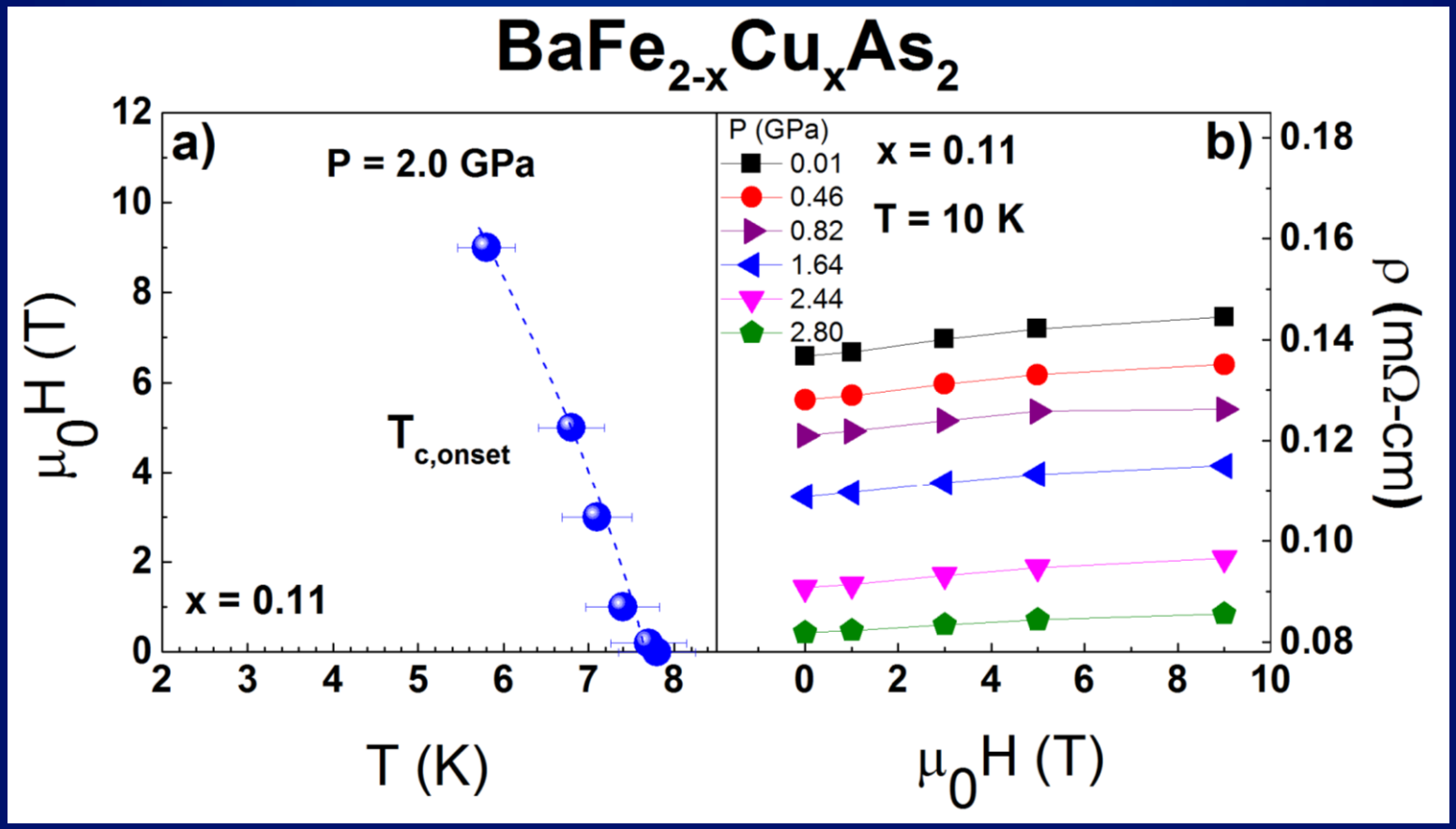
Combined external pressure and chemical substitution results

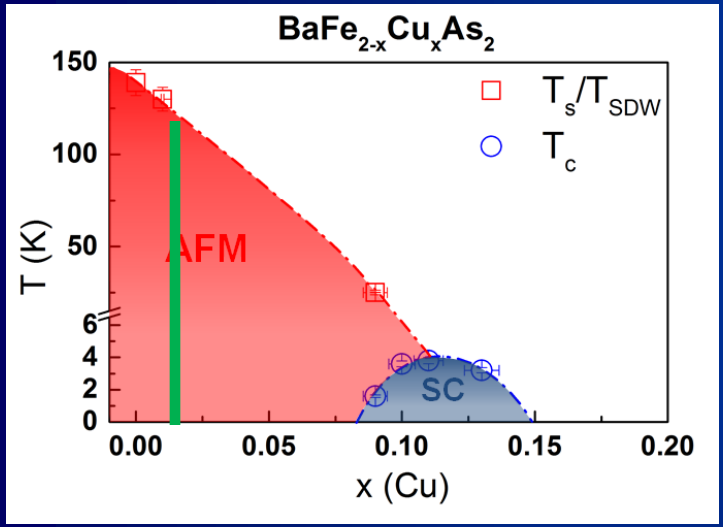
- Magnetic field dependence of T_c and magneto resistance for the OPD compound at several pressures:

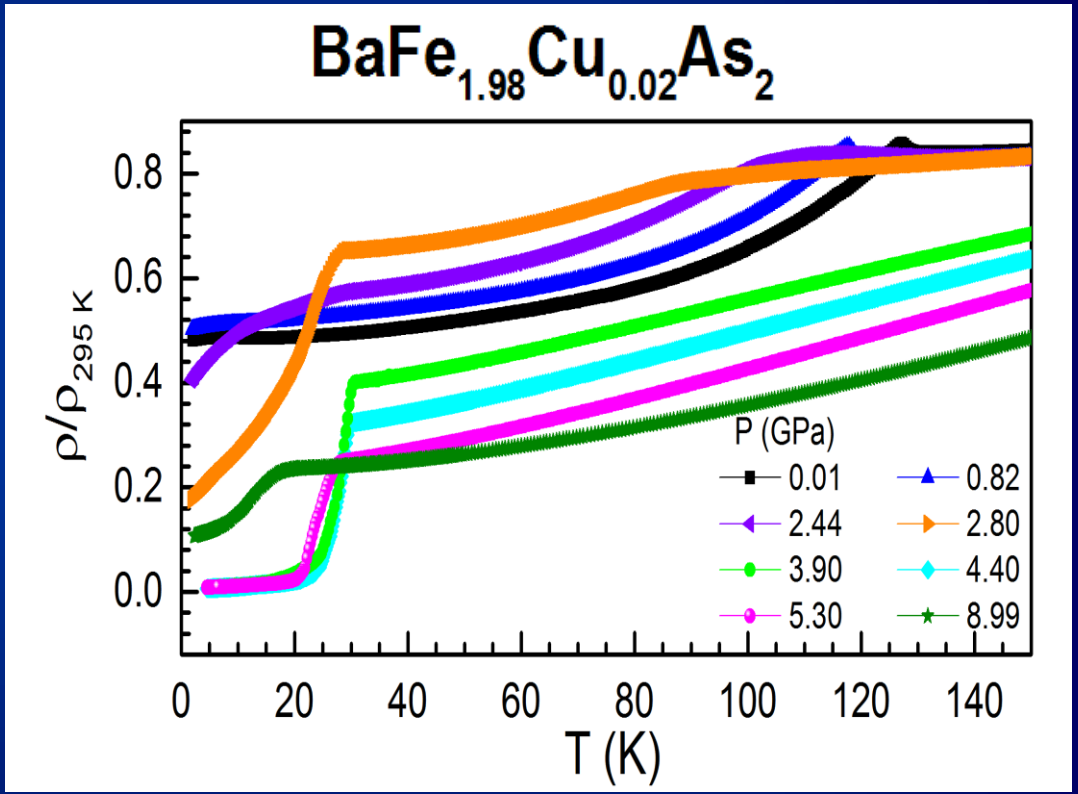


M M Piva …P.G. Pagliuso *et al J. Phys.: Condens. Matter* **27,** 145701 (2015)

Combined external pressure and chemical substitution results

- In-plane electrical resistivity for the under doped compound at several pressures:

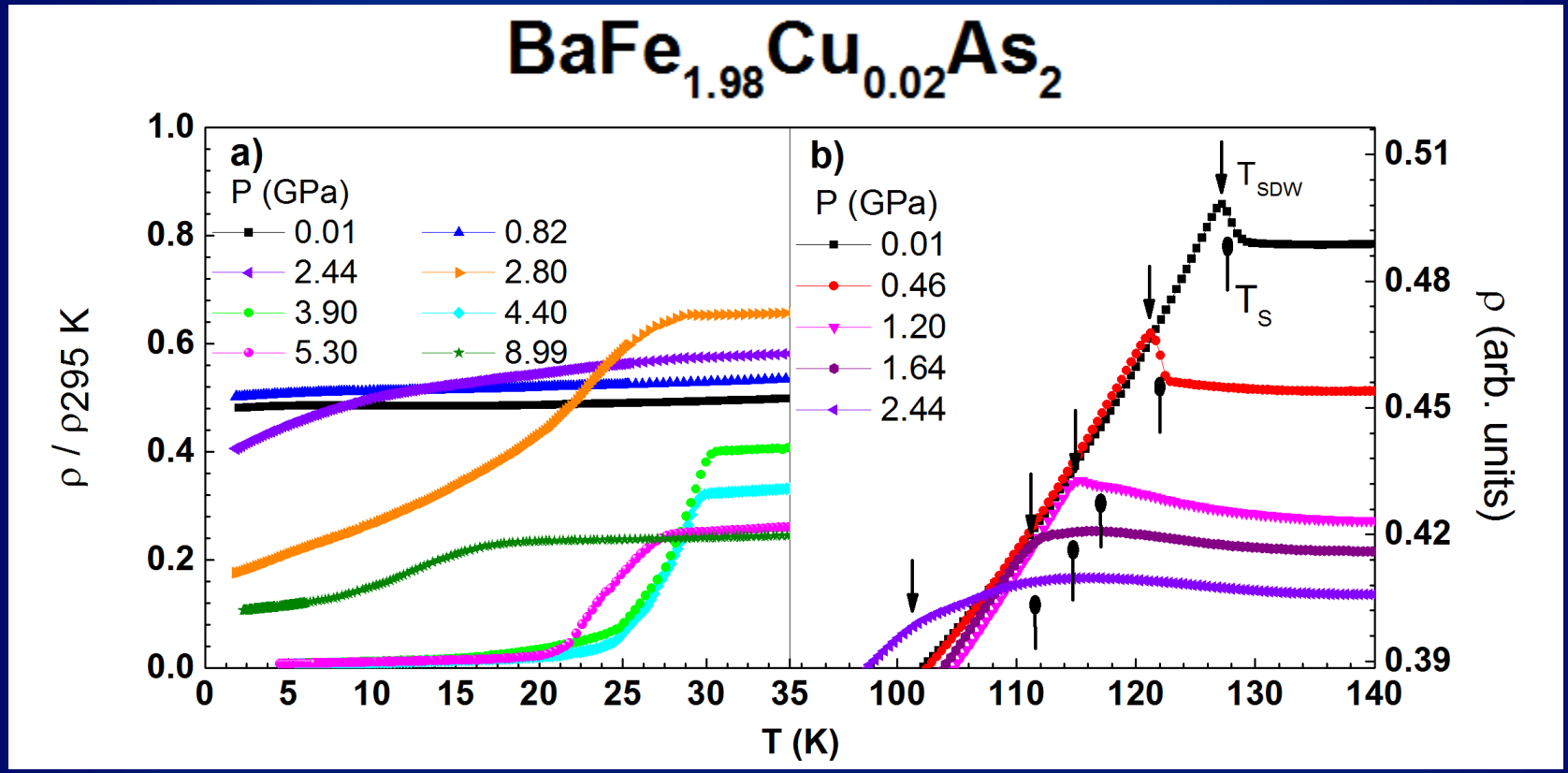




M M Piva …P.G. Pagliuso *et al J. Phys.: Condens. Matter* **27,** 145701 (2015)

Combined external pressure and chemical substitution results

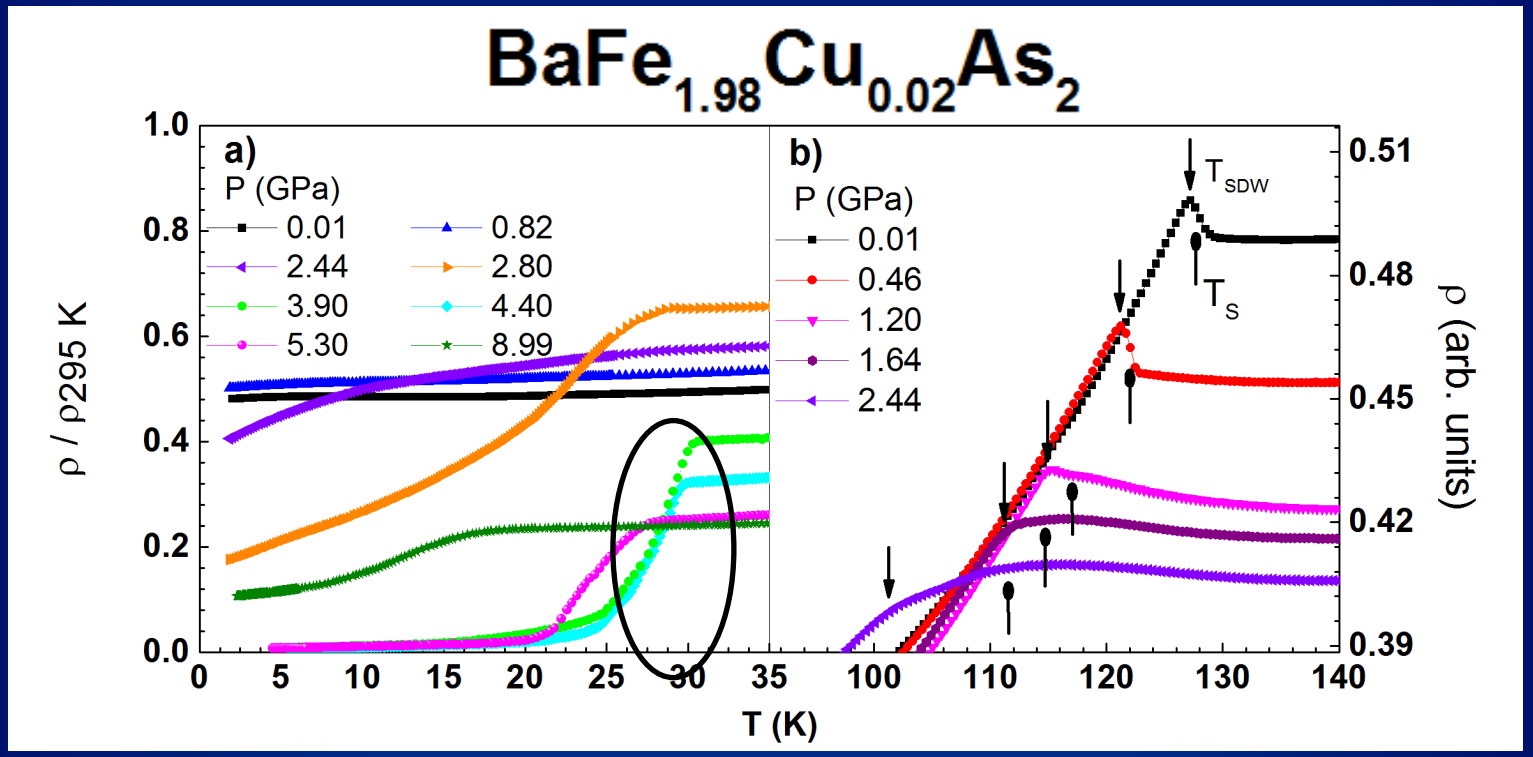
- Zoom in the transition temperatures:



M M Piva …P.G. Pagliuso *et al J. Phys.: Condens. Matter* **27,** 145701 (2015)

Combined external pressure and chemical substitution results

- Zoom in the transition temperatures:



This value of T_c is close to the maximum T_c achieved in the pure compound under pressure

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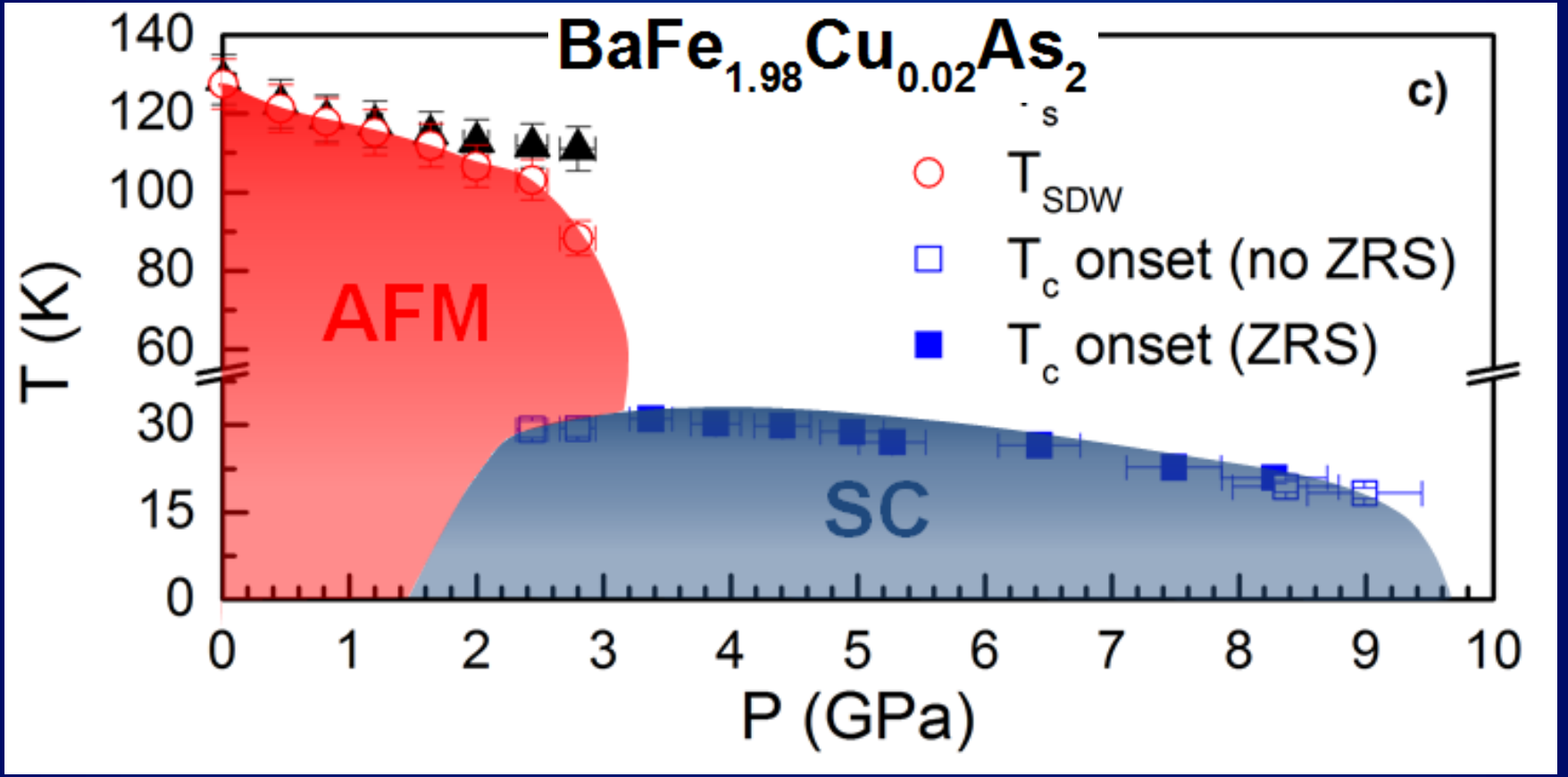
Combined external pressure and chemical substitution results

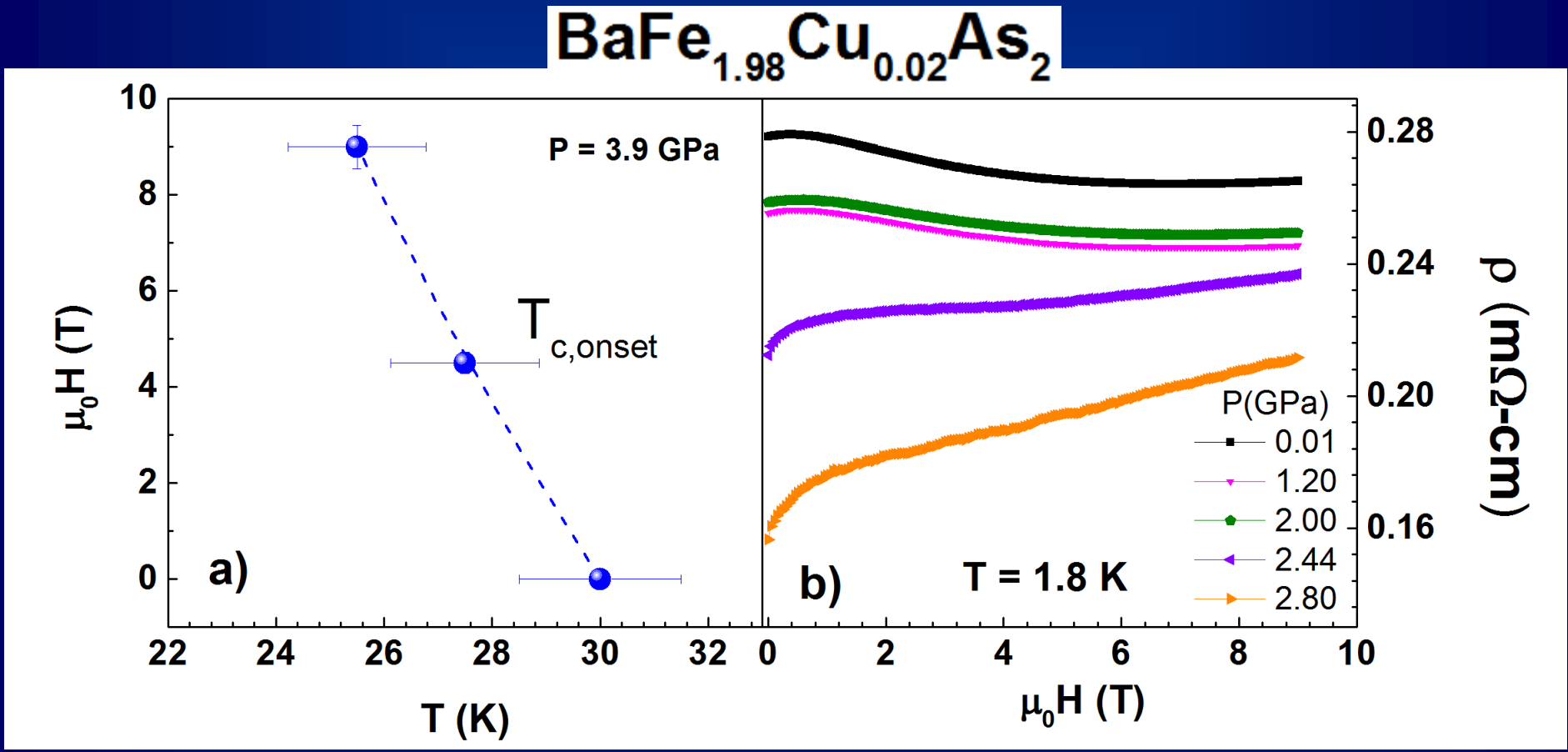
- Temperature-pressure phase diagram for the under doped sample:



M M Piva …P.G. Pagliuso *et al J. Phys.: Condens. Matter* **27,** 145701 (2015)

Combined external pressure and chemical substitution results

- Magnetic field dependence of T_c and magneto resistance for the under doped compound at several pressures:



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Combined external pressure and chemical substitution results

Positive MR → Fermi-liquid regime of normal metals

Negative MR

↓

Spin-dependent scattering mechanism

- Fe 3d electrons
- Cu^{2+} local moments

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Combined external pressure and chemical substitution results

Positive MR → Fermi-liquid regime of normal metals

Negative MR

↓

Spin-dependent scattering mechanism

- ~~Fe 3d electrons~~
- Cu^{2+} local moments

↓

Impurity magnetic pair-breaking effect

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Combined external pressure and chemical substitution results

Impurity magnetic pair-breaking effect

Applied pressure

Cu-substitution ($BaCu_2As_2$ is a Pauli paramagnet)

Screening of Cu^{2+} local moments

Consistent with the positive MR in OPD samples

Great enhancement of T_c

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Final Remarks

- Fe 3d bands orbital differentiation induced by local distortions of the dFe-As seeem to be most relevant tuning parameter to drive the SDW phase to SC in Ba112.

- Cu^{2+} and Mn^{2+} spins give rise to a non-concentional IPB mechanism in substituted Ba112 system

- Cu-substituted Ba112 show screening/suppresion of the Cu^{2+} spins as a function of pressure and Cu-concentration.

- Low-x Cu substitution samples showm the same pressure induced T_c around 30 K as the pure compound.

- Evolution of a negative MR to a positive one as a function of pressure or Cu substitution.

Acknowledgments

FAPESP

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CAPES

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CBPF

LNLS

Universidade Federal do ABC

NATIONAL HIGH MAGNETIC FIELD LABORATORY

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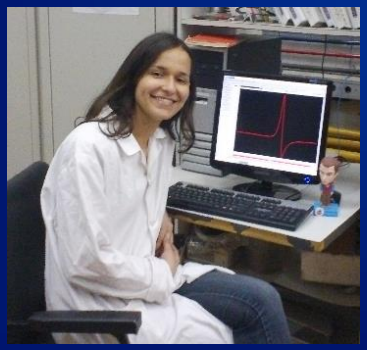

Dra. Priscila Rosa

Dr. Thales Garitezi

GPOMS no f : Gpoms Physics

“ FeAs” Team – Next Generation

GPOMS no f : Gpoms Physics

Thank you for your attention!