BRAZILIAN CENTER FOR RESEARCH IN PHYSICS- CBPF / MCT www.cbpf.br

CBPF - National Institute/MCT

Rio de Janeiro

Jacques Danon

Magnetic and Structural Transitions in the New Fe-pnictide Superconductors

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Dalber Sanchez-Candela, UFF

HFI-NQI 2010, 12-17 September 2010 CERN, Geneva

Collaborators

Samples grown - Physical characterization

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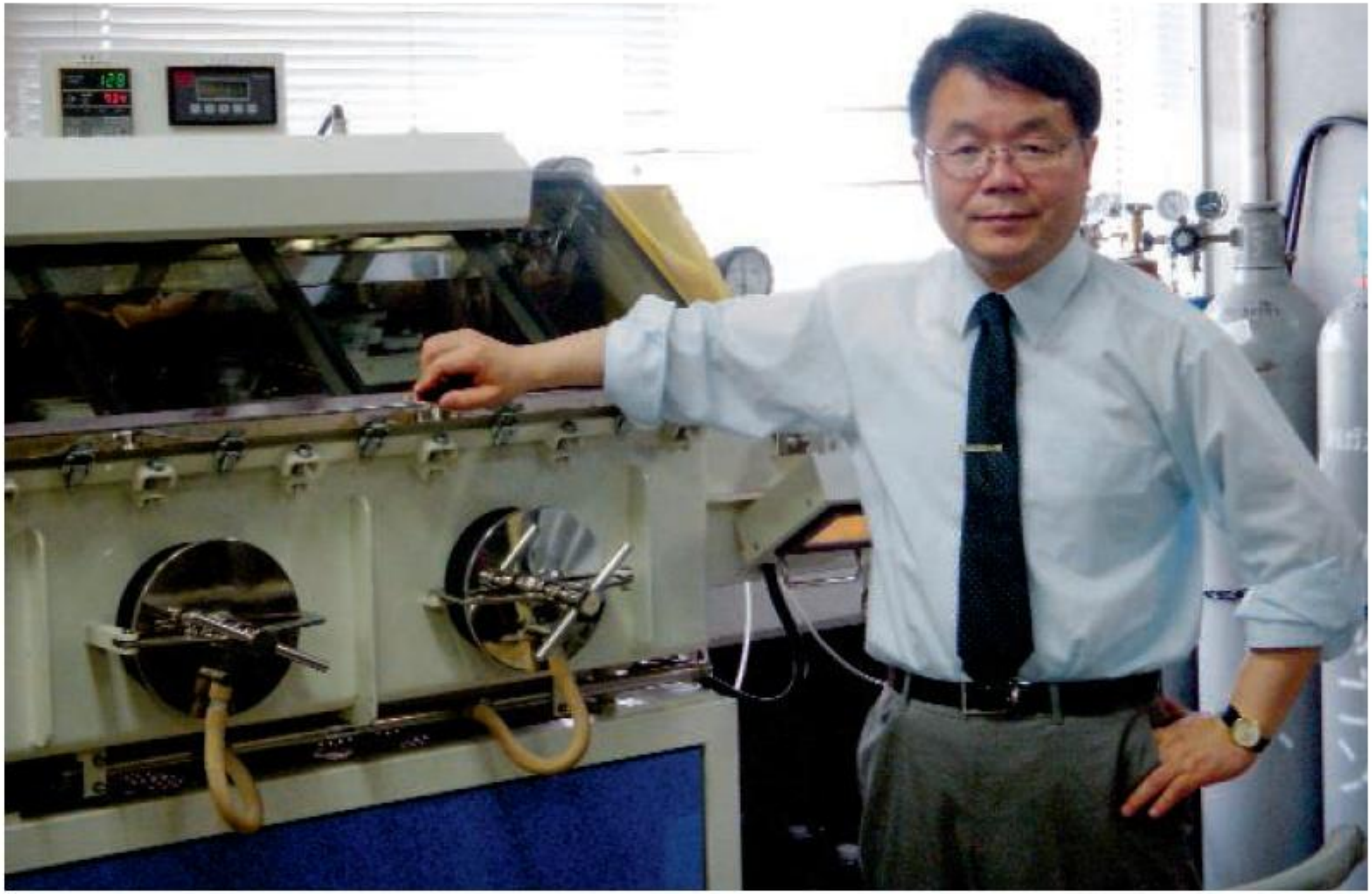
CIAM Collaboration Program (NSF/CNPq)



Discoverer. Hideo Hosono, a materials scientist at the Tokyo Institute of Technology, cooked the first of the new superconductors that have captivated researchers the world over.

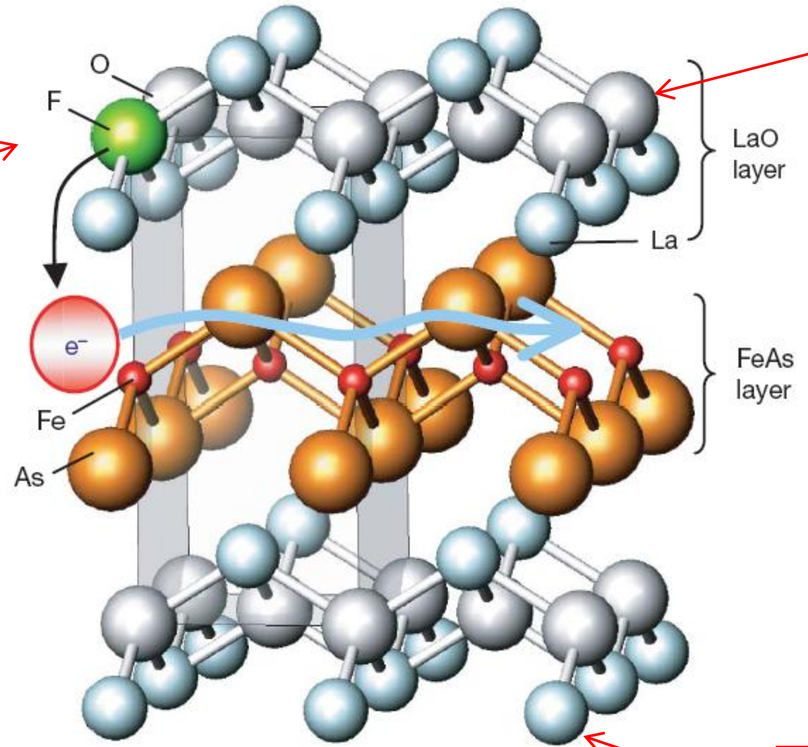
Hideo Hosono started with Iron-Based Layered SC: LaOFeP (Tc=6K) to $SmFeAsO_{1-x}$ (Tc= 55 K)

Iron-based layered compound LnOFeAs "1111 phase"

The discovery of superconductivity in fluorine doped LaFeAsO superconductor with T_C = 26 K* has generated enormous interests in the community of superconductivity

F-substitution
$LaFeAsO_{1-x}F_x$

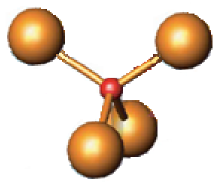
*Y. Kamihara, *et al*, J. Am. Chem. Soc. 130, 3296 (2008).



O-deficiency
$LnFeAsO_{1-y}$

T_c=55 K in $SmFeAsO_{1-\delta}$

Z.A. Ren et al. Europhys. Lett. **83** 17002 (2008)



Ln substitution ($Gd^{3+} \rightarrow Th^{4+}$)

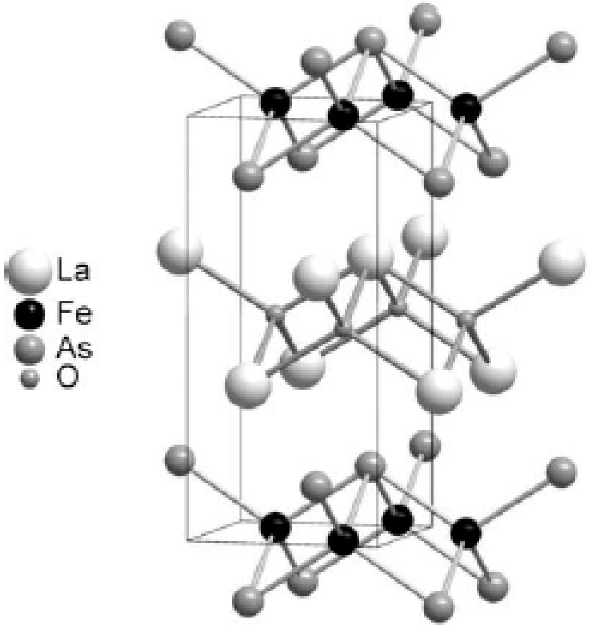
T_c up to 56K for (Gd,Th)FeAsO

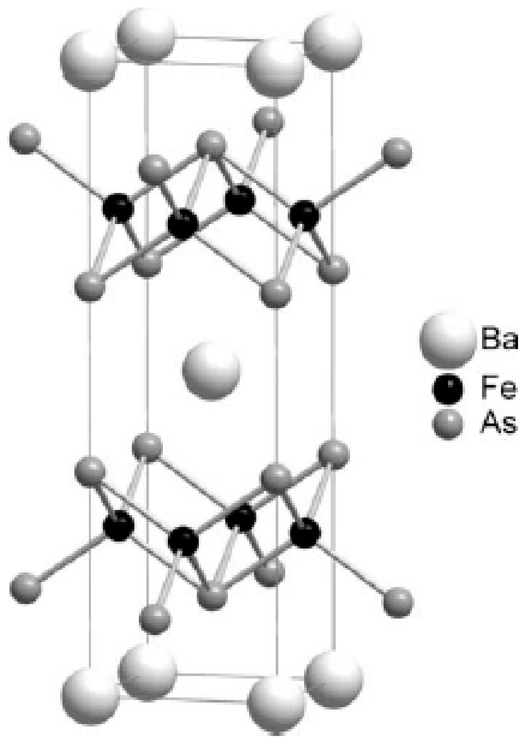
Cao Wang *et al EPL* **83 67006** (2008)

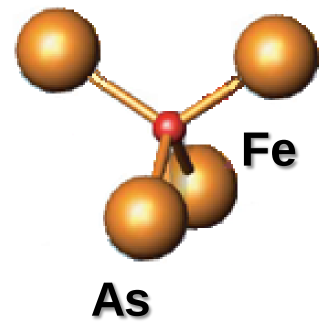
LaOFeAs -> (Tc = 26 K) SC under
- doping with F^-
- oxygen deficiency

**Hiroki Takahashi, Nature 453, 376 (2008)

Iron-arsenide- 1111 and 122 Phases







Fe has tetrahedral coordination and is formally divalent

1111 ROFeAs
R=La, …….., Gd

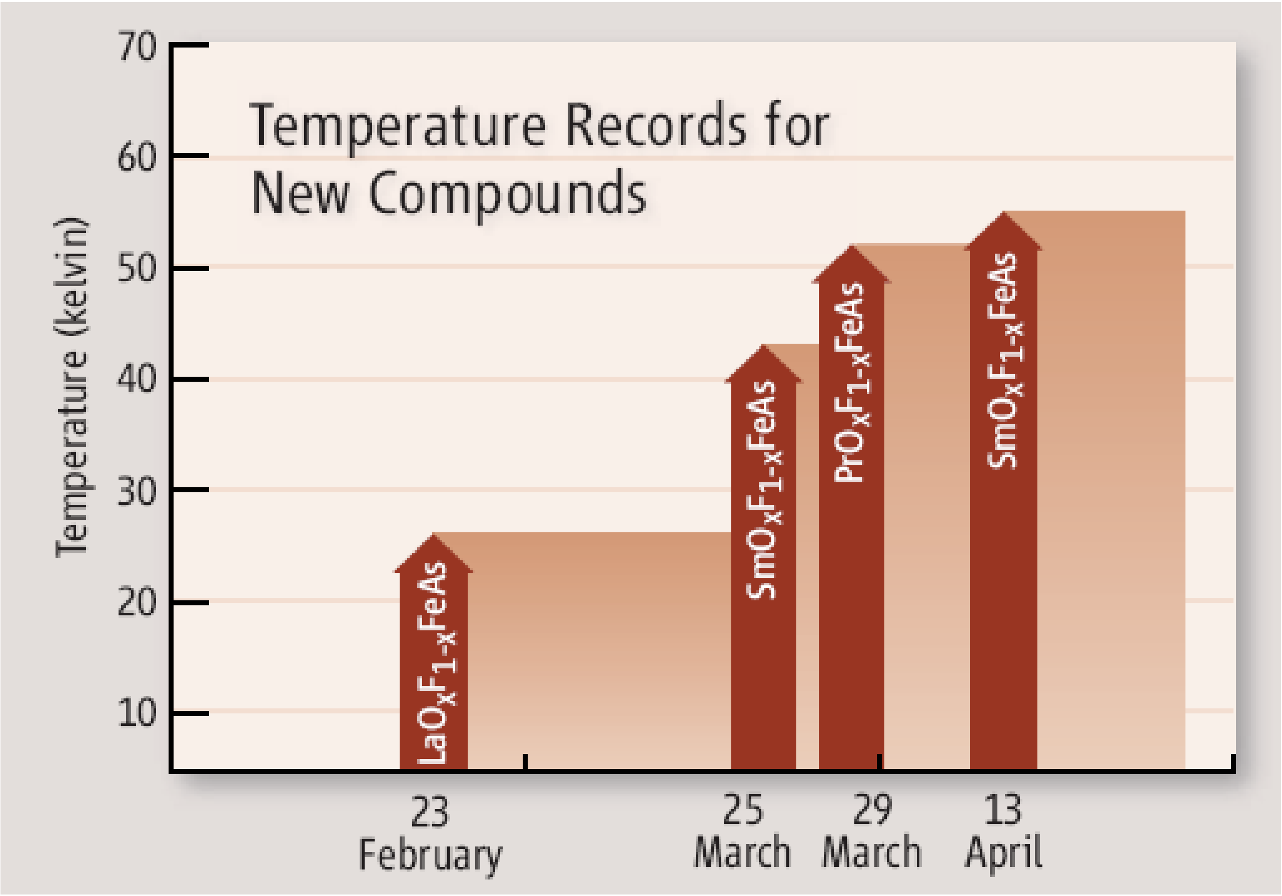
122 AFe_2As_2
A=Ba, Ca Sr, Eu

The crystal is composed of a stack of alternating REO or A and FeAs layers.

*D. Johrendt and R. Pottgen, Angew. Chem. Int. Ed. 47, 4782 (2008).

CONDENSED MATTER PHYSICS

New Superconductors Propel Chinese Physicists to Forefront



In 2008

More than 700 papers

Increase of T_C of RE(O,F)FeAs superconductor after announcement by Hosono on Feb., 2008, made by Chinese physicists including PI Wang and LE Wen

Catalogue of Fe-based Superconductors

Fe-1111

LnFePO Tc~5-6 K (Tc~8.8K at Pressure ~0.8 GPa)

$LnFeAsO_{1-x}F_x$

$LaFeAsO_{1-x}F_x$	x=0.11, Tc~26K (Tc~43K at P~4GPa)
$CeFeAsO_{1-x}F_x$	x=0.16, Tc~41K
$SmFeAsO_{1-x}F_x$	x=0.10, Tc~43K
$NdFeAsO_{1-x}F_x$	x=0.11, Tc~52K
$PrFeAsO_{1-x}F_x$	x=0.11, Tc~52K
$TbFeAsO_{1-x}F_x$	Tc~46K (x=0.1, 0.2)
$DyFeAsO_{1-x}F_x$	Tc~45K (x=0.1, 0.2)

$LnFeAsO_{1-x}$

$LaFeAsO_{1-x}$	31.2
$CeFeAsO_{1-x}$	46.5
$PrFeAsO_{1-x}$	51.3
$NdFeAsO_{1-x}$	53.5
$SmFeAsO_{1-x}$	55.0
$GdFeAsO_{1-x}$	53e
$TbFeAsO_{1-x}$	52
$DyFeAsO_{1-x}$	52

(Ln,RE)FeAs

$(La_{1-x}Sr_x)FeAsO$ Tc~ 25K(x=0.13), hole-doping

$Gd_{1-x}Th_x)FeAsO$ Tc~56K (x=0.2), electron-doping

Ca(Sr)FeAsF

$Ca(Fe_{1-x}Co_x)AsF$ Tc~22K (x=0.10) [$Ni^{2+}(3d^7) \rightarrow Fe^{2+}(3d^6)$]

$Ca(Fe_{1-x}Ni_x)AsF$ Tc~12K (x=0.05) [$Ni^{2+}(3d^8) \rightarrow Fe^{2+}(3d^6)$]

$(Sr_{1-x}La_x)FeAsF$ Tc~32K (x=0.4)

Fe-122

$CaFe_2As_2$ Tc~27K (Pressure~3.0 GPa)

$(Ba_{1-x}K_x)Fe_2As_2$ Tc~38K (x=0.4)

$(Sr_{1-x}Na_x)Fe_2As_2$ Tc~37K (x=0.4)

$Ba(Fe_{1-x}Co_x)_2As_2$ Tc~22K (x=0.1)

Fe -111

$Li_{1-x}FeAs$ Tc~18K

Fe-42622

Sc 42622
V 42622

Fe-11

$FeSe_{1-x}$ Tc~8K (x=0.88), Tc~27 K (P~ 1.48 GPa)

$FeSe_{1-x}Te_x$ Tc~15K

$FeTe_{1-x}S_x$ Tc~10K (x=0.2), non-toxic

$RFeAsO_{1-x}F_x$,	AFe2As2,	$Sr_4A_2O_6Fe_2As_2$
Fe-1111	**Fe- 122**	**Fe - 42622**

Catalogue of Fe-based Superconductors

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LnFePO Tc~5-6 K (Tc~8.8K at Pressure ~0.8 GPa)

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$PrFeAsO_{1-x}F_x$ x=0.11, Tc~52K
$TbFeAsO_{1-x}F_x$ Tc~46K (x=0.1, 0.2)
$DyFeAsO_{1-x}F_x$ Tc~45K (x=0.1, 0.2)

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(Ln,RE)FeAs

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Fe-122

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Fe -111

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Sc 42622
V 42622

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$FeSe_{1-x}Te_x$ Tc~15K
$FeTe_{1-x}S_x$ Tc~10K (x=0.2), non-toxic

$RFeAsO_{1-x}F_x$,	AFe2As2,	$Sr_4A_2O_6Fe_2As_2$
Fe-1111	Fe- 122	Fe - 42622

EXPERIMENTAL INFORMATION

Ceramic polycrystalline specimens of $RFeAsO_{1-x}F_x$ (R=Nd, Ce) were synthesized by G F Cheng and N. L. Wang at Institute of Physics, Chinese Academy of Sciences, Beijing

Single crystals of AFe2As2 (A=Ca,Ba, K) were synthesized at Ames National Lab., Ames, USA and N. L. Wang

$Sr_4A_2O_6Fe_2As_2$ (A=Sc,V) Oxypnictides polycrystalline specimens were provided by Hai-Hu **Wen,** from National Lab. Superconductivity, Beijing

μSR performed by Uemura group in TRIUMF on some samples with participation of Julian Munevar and Dalber Sanchez

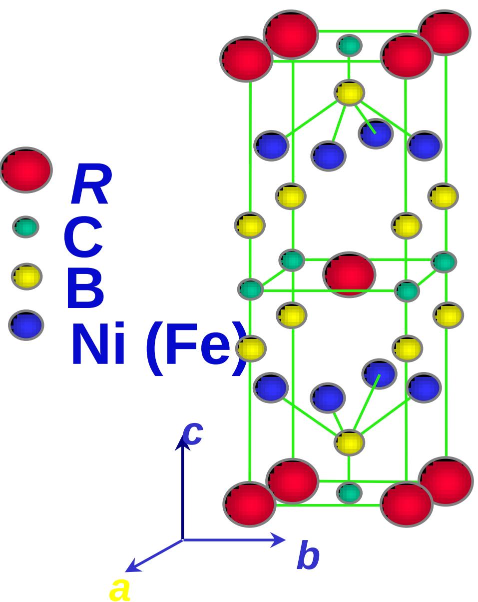
Mössbauer experiments were performed at CBPF, Rio de Janeiro with an Oxford cryostat in sinusoidal mode with transmission geometry, with $^{57}Co:Rh$ source and sample at the same T

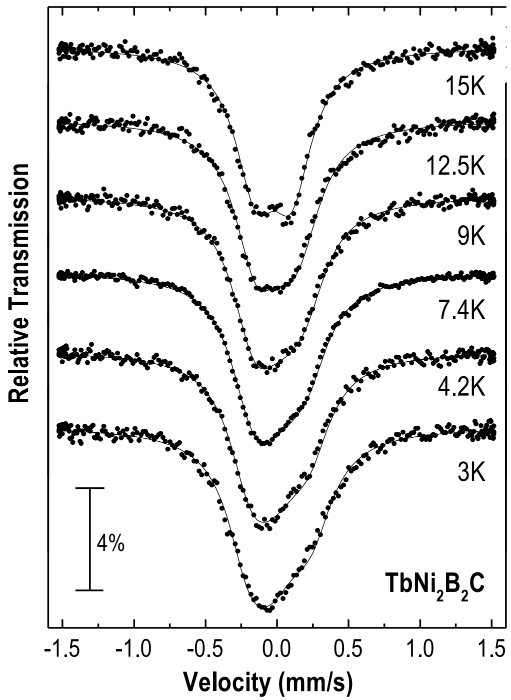
Previous Studies

RNi_2B_2C

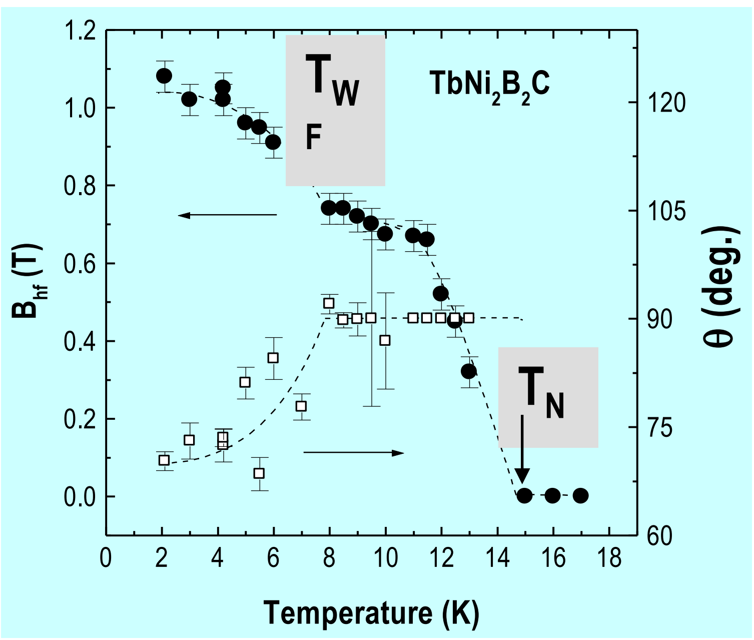
Mössbauer Spectroscopy in RNi_2B_2C and RNiBC with ^{57}Fe at Ni site

- **information about local symmetry and magnetic order**
- **Bhf at the ^{57}Fe nucleus, due to non collinear AF spin structure of the RE moments, acts as a pair-breaking field at the Ni site**





TbNi2B2C

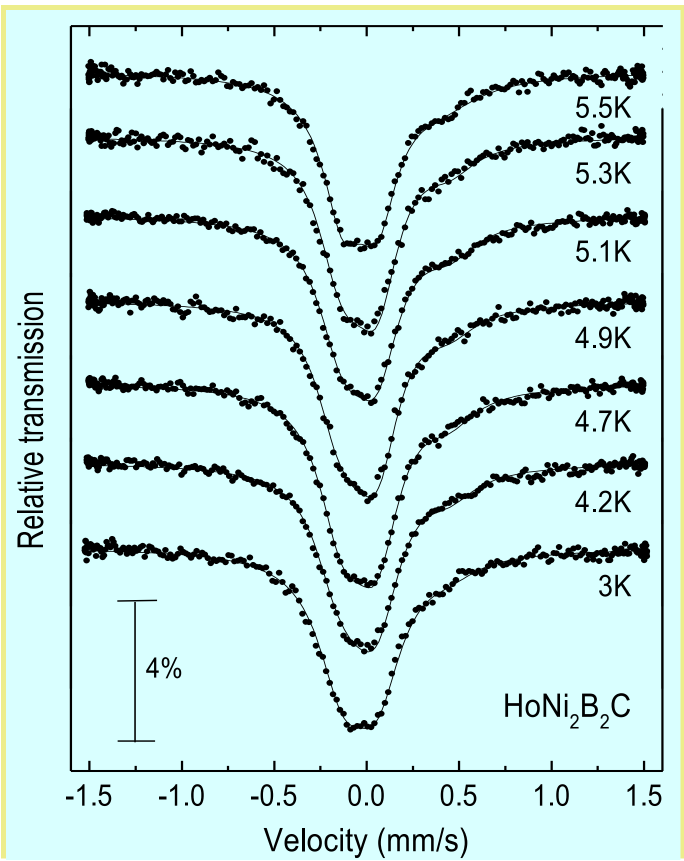


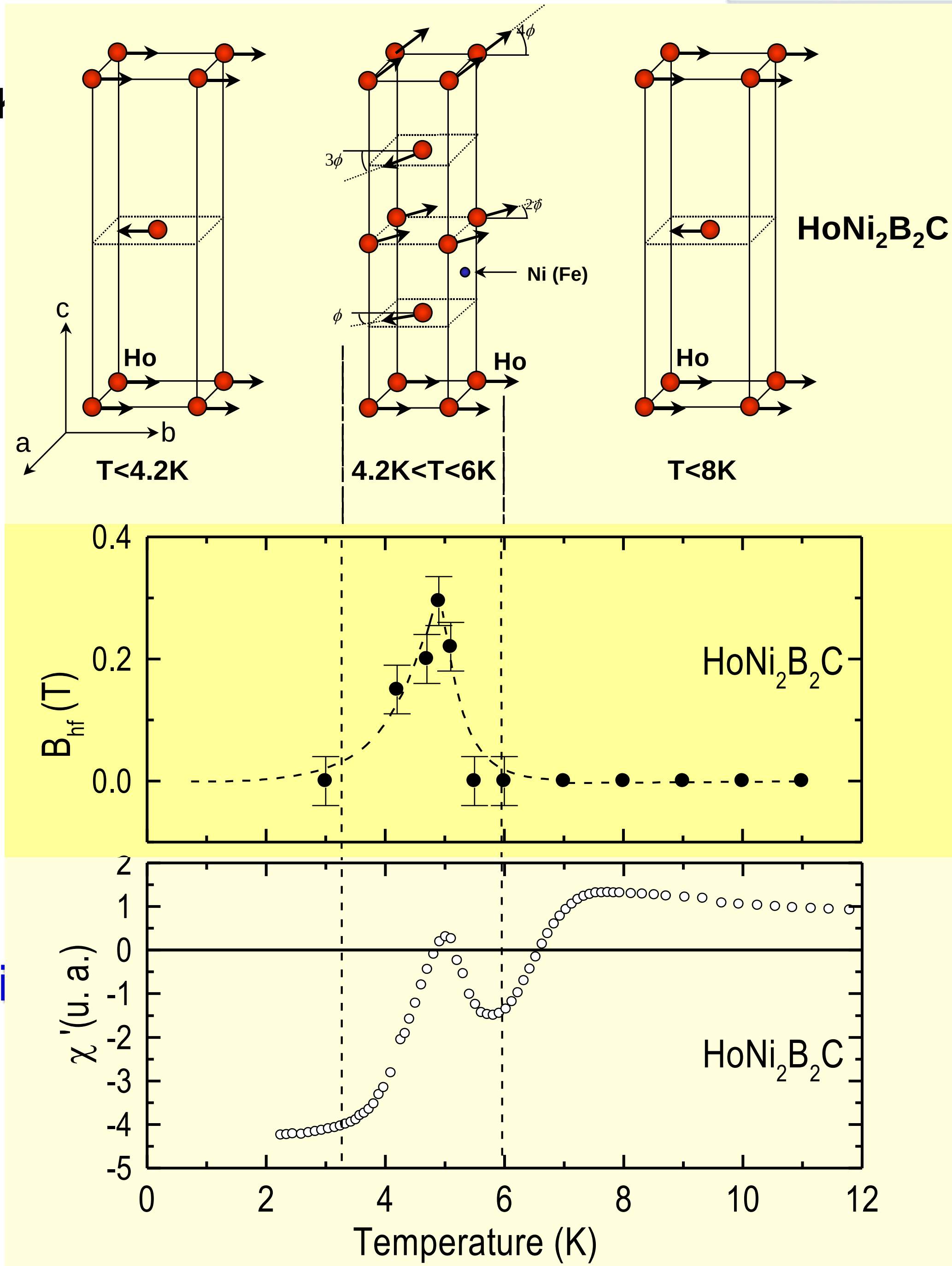
✓ **Magnetism is due exclusively to R magnetic moments:**

AF, FM, WFM, and SDW determined by coupling of R layers

Superconductivity and Magnetism in $HoNi_2B_2C$

Reentrant behavior and incommensurate modulated magnetic structure for $4.6 \leq T \leq 6$ K

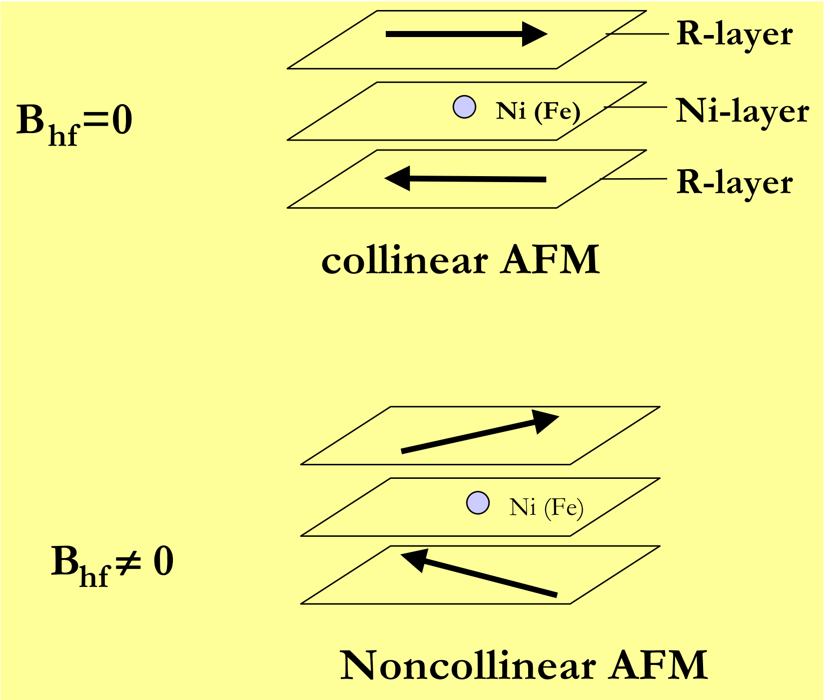




Evidence of pair-breaking field at the Ni

D. R. Sánchez, H. Micklitz, M. B. Fontes, S. L. Bud'ko and E. Baggio Saitovitch, Phys. Rev. Lett. 76, 507 (1996)

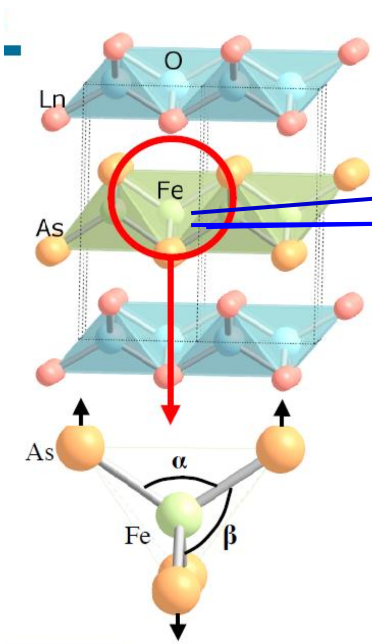
va

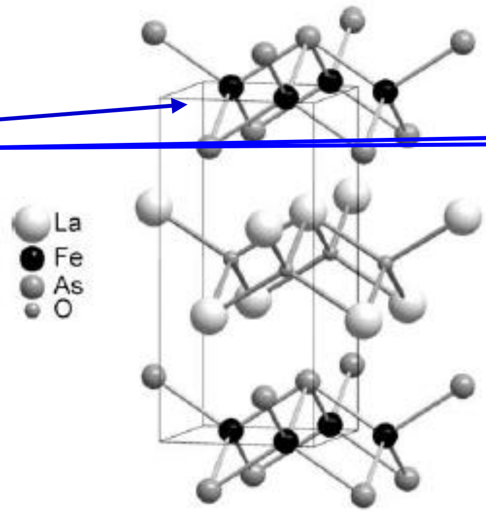


$$B_{hf} = B_{thf} = \sum_i \alpha_i \vec{s}_i$$

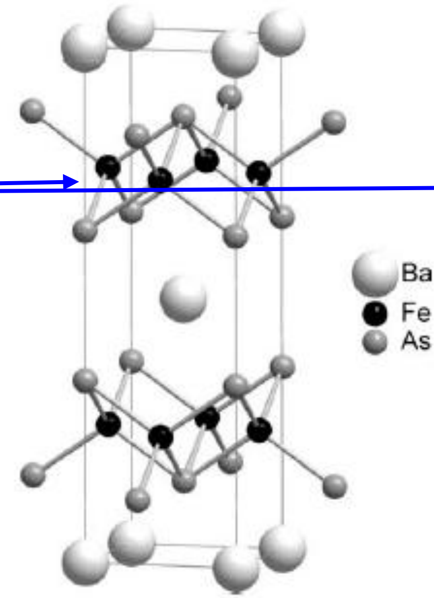
No **Bhf field was observed at any temperature below T_N for the AFM *superconductors* $ErNi_2B_2C$ and $DyNi_2B_2C$**

Structures of Fe-As: RFeAO , AFe_2As_2 and $Sr_4A_2O_6Fe_2As_2$



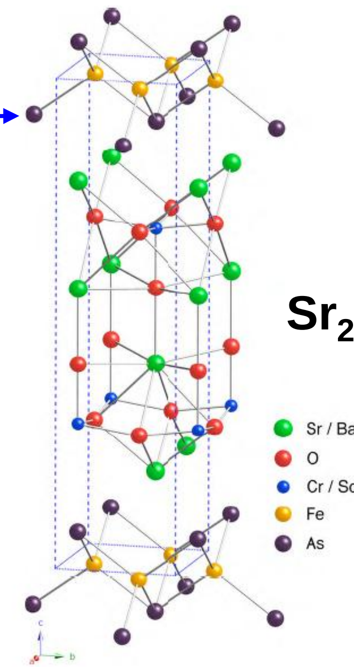


LaOFeAs - 111



$BaFe_2As_2$- 122

$Sr_2(V/Cr)O_3FeAs$



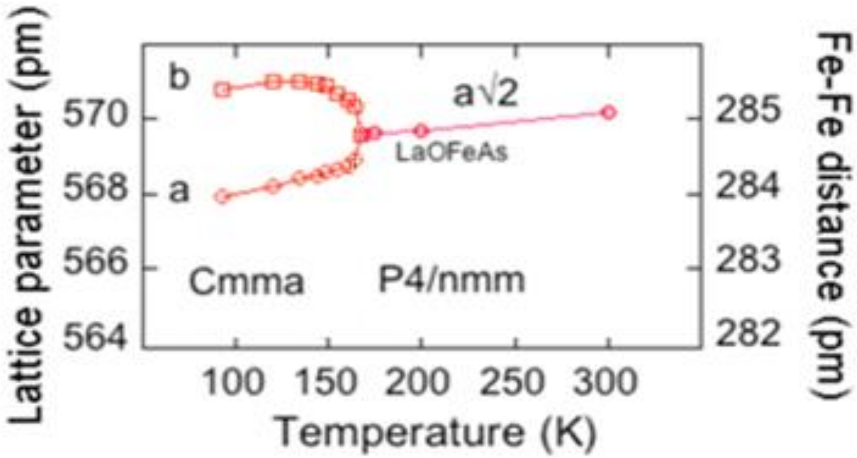
Sr_2AO_3

Structure:	Tetragonal *P4/nmm*	Tetragonal *I4/mmm*	Tetragonal *P4/nmm*
Structural transition:	~ 155K	~ 140K	*NONE*
Magnetic ordering:	~ 140K	~ 140K	36 K
Superconductor:	with F dop & O deficiency	withr K, Ca, Co doping	A=V

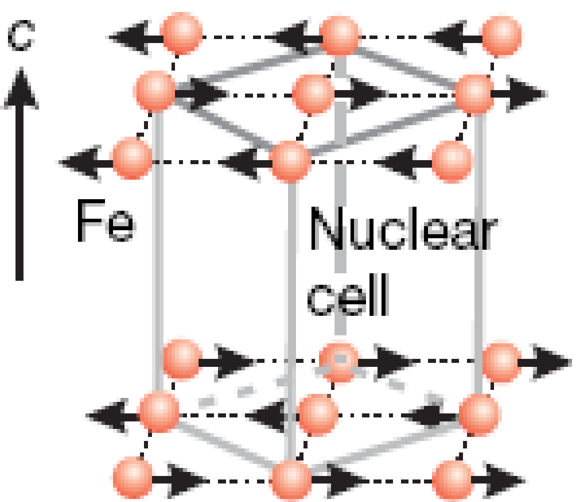
Local structure at Fe site is similar different Fe moments and 3d Fe contribution in the density of states at E_F level

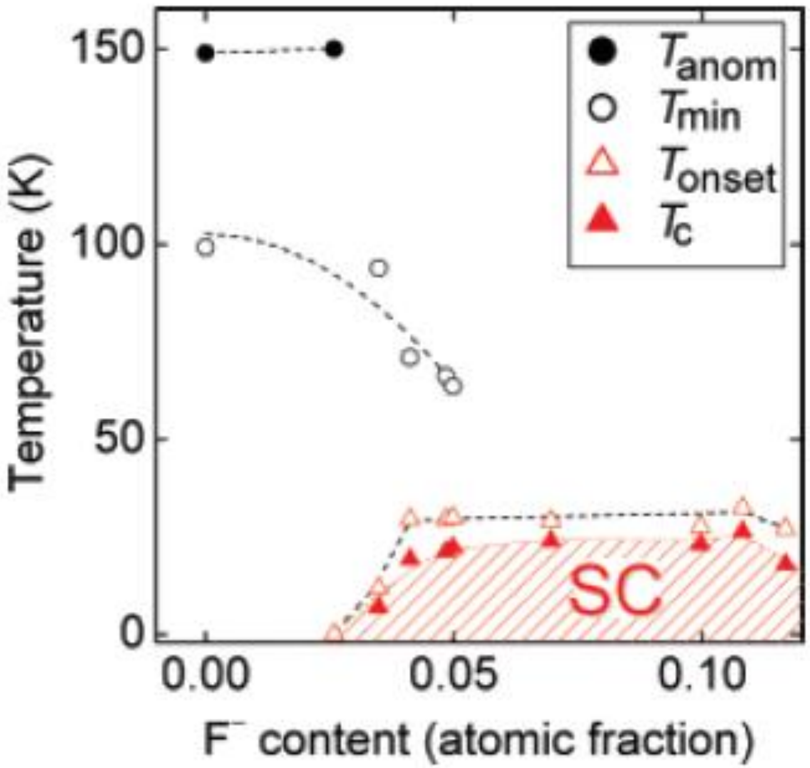
LaOFeAs

LaOFeAs has structural distortion below ~150 K



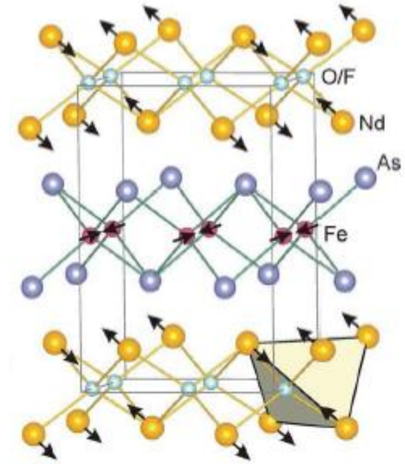
LaOFeAs has long range **SDW-type AF order** at ~134 K with $\mu_{Fe}=0.36\mu_B$





NdOFeAs and CeOFeAs

	NdOFeAs	CeOFeAs
T_S (P/4nmm-Cmma)	~ 150K	~158K
$T_{N\ (Fe)}$	~ 141K	~ 140 K
μ_{Fe}	~ 0.32 μB	~ 0.6 μB
$T_{N(R)}$	~ 2K	~ 4K

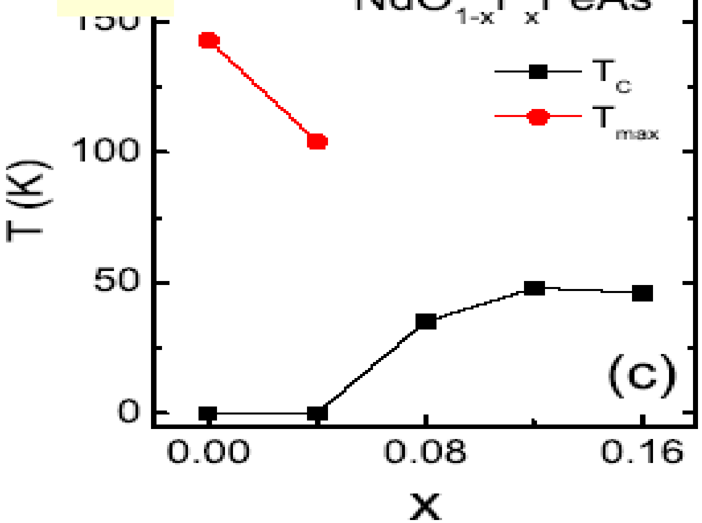


Fratini M, Supercond. Sci. Technol. 21, 2008. Ying Chen, Phy. Rev. B 78, 2008. Y. Qiu, Phys. Rev. Lett. 101, 2008.

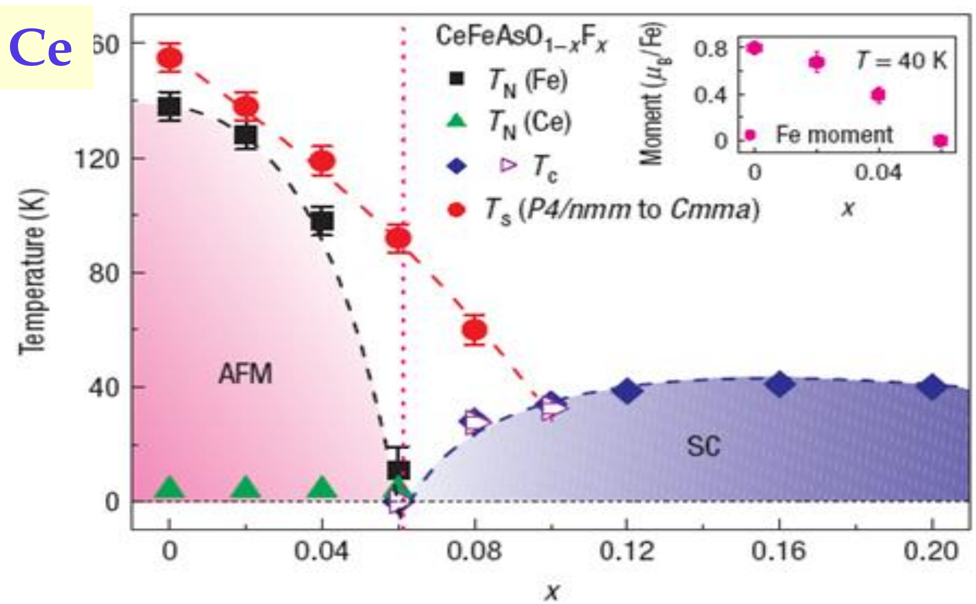
$NdFeAsO_{1-x}F_x$ and $CeFeAsO_{1-x}F_x$

Electron-doping by F^- substituting for O^{2-} increases T_c to about 50 K.

Nd

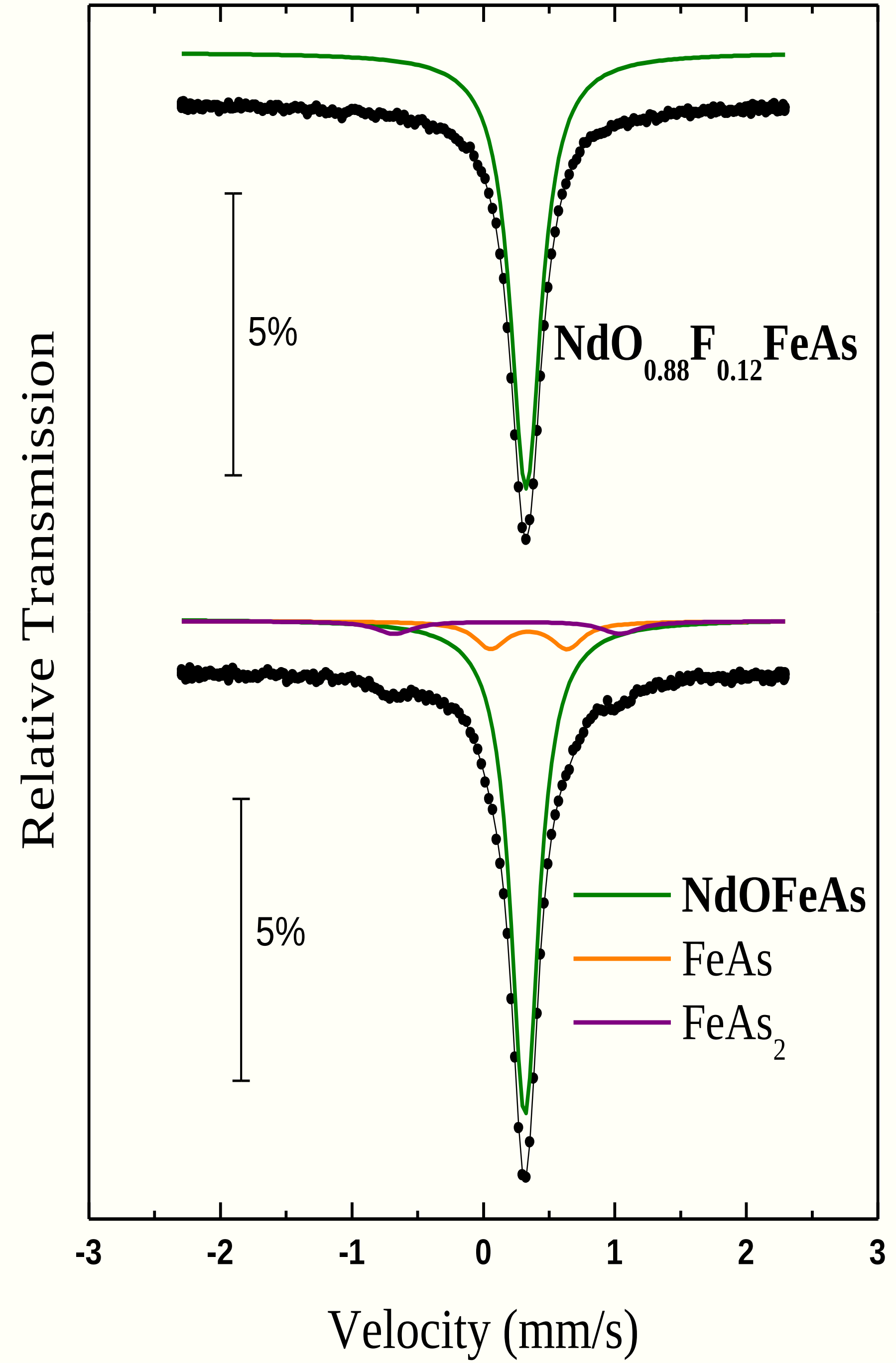


Ce



G. F. Chen, Phys. Rev. Lett. **100**, 247002 (2008) and Chin. Phys. Lett. 25, 2235 (2008). Jun Zhao, Nature Materials 7, 953 (2008)

$NdFeAsO_{0.88}F_{0.12}$ and NdFeAsO



Room temperature MS

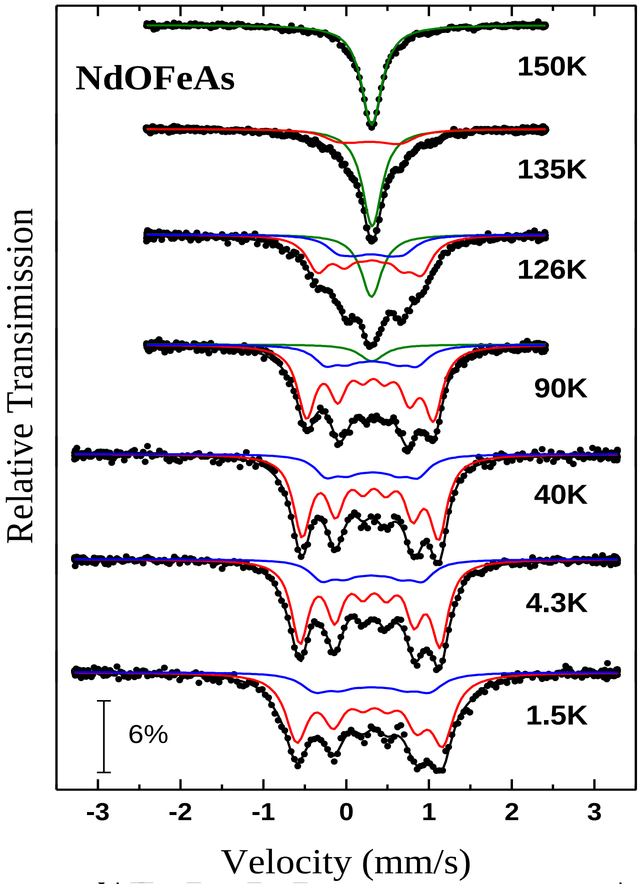
RT $NdFeAsO_{0.88}F_{0.12}$ spectrum: unique phase, similar to that found for LaOFeAs*

ΔE_Q=0.02(2) *mm/s*
IS=*0.437(1) mm/s,*

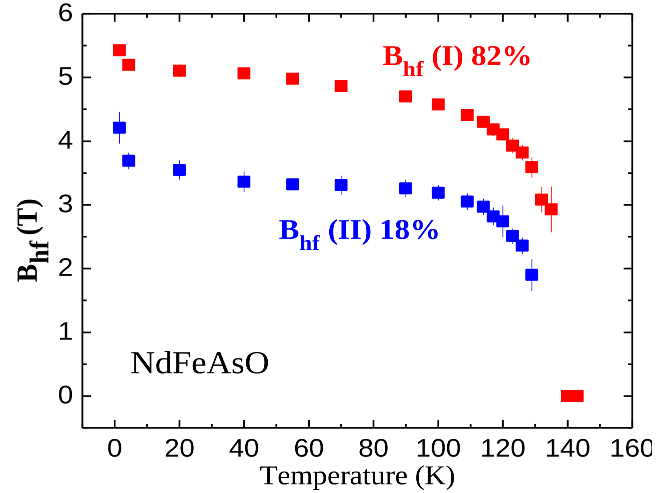
RT NdFeAsO: small impurities, **FeAs** (~ 8%) and **$FeAs_2$** (~5%).
The main component (doublet) is attributed to Fe in NdFeAsO phase and their hyperfine parameters are almost the same as for $NdFeAsO_{0.88}F_{0.12}$

*H.-H. Klauss et al., Phys. Rev. Lett **101,** 077005 (2008).

Results for NdFeAsO

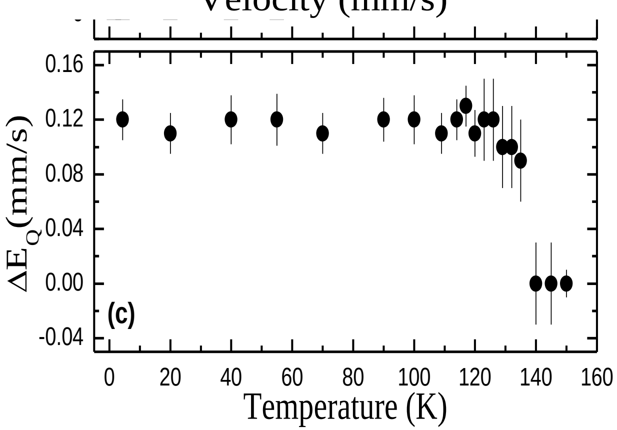


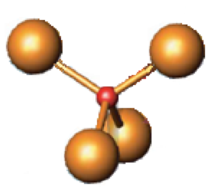
The magnetic transition at ~ 140K is due to the magnetic ordering of Fe moments.



CM1 : θ(I) = 90 ± 5º, commensurate AF structure with the Fe spins lying in the (*a*,*b*)

CM2: θ(II) ≈ 55º, all angles in the range 0≤ θ ≤90º occur with equal probability





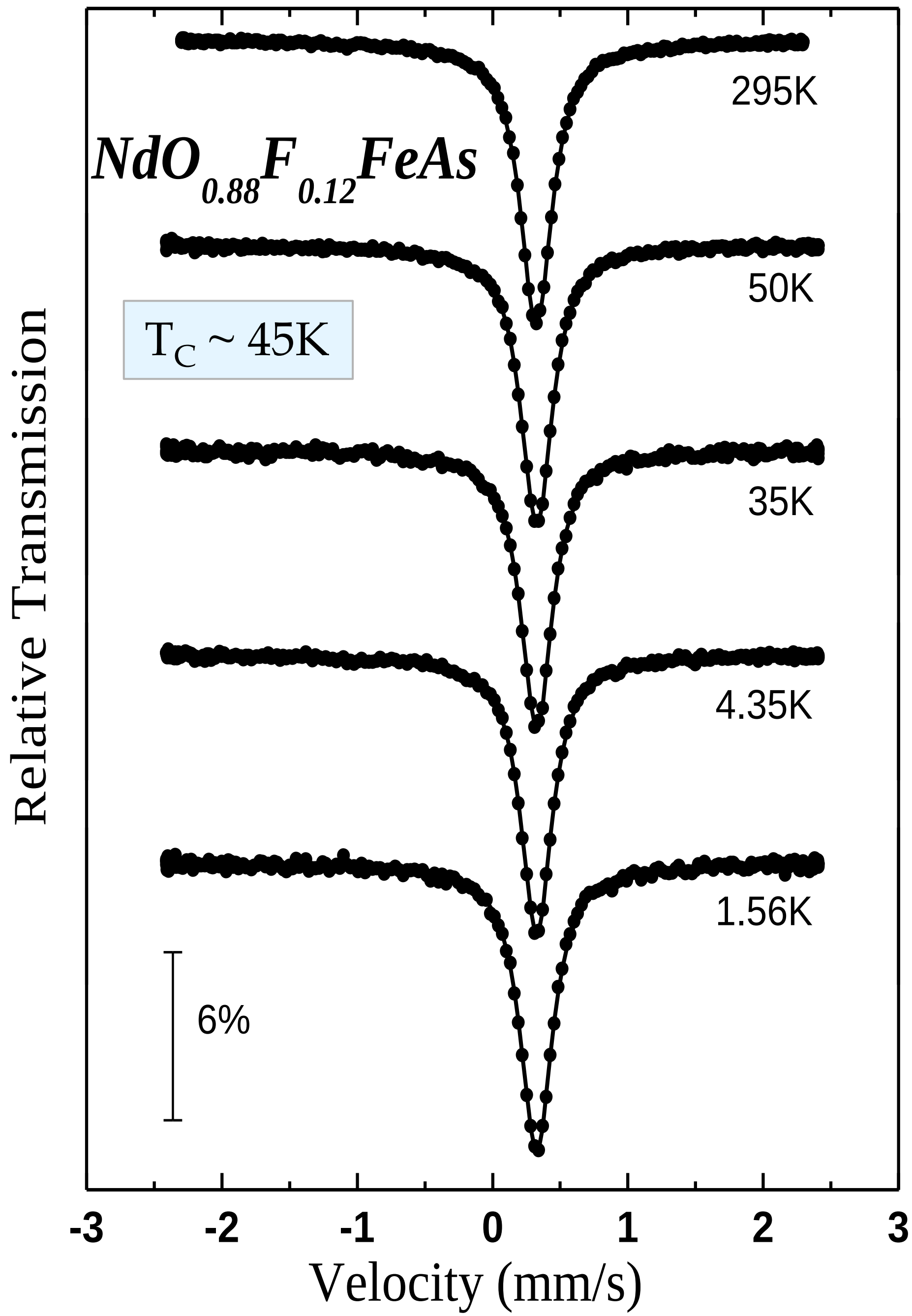
(ΔE_Q → measure of the As Tetrahedra)

Below ~2K an additional increase of B_{hf} : transferred B_{hf} at Fe due to collinear AF order of **Nd moments.**

μSR results* support our model used to analyze our Mössbauer spectra.

*A. Aczel, et al, Phys. Rev. B78, 214503 (2008)

Results for SC $NdFeAsO_{0.88}F_{0.12}$



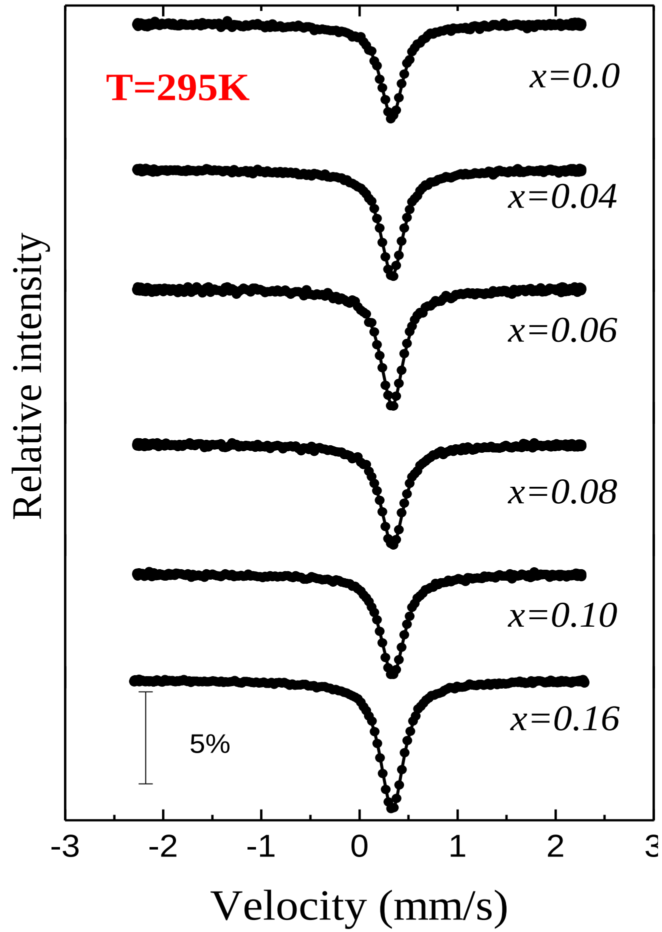
Low temperature MS

No hyperfine magnetic field was observed at ^{57}Fe nucleus [$B_{hf}(1.5K) \leq 0.1T$] of Fe in superconducting $NdFeAsO_{0.88}F_{0.12}$ at any temperature (down to 1.5K)

Absence of magnetism in this compound

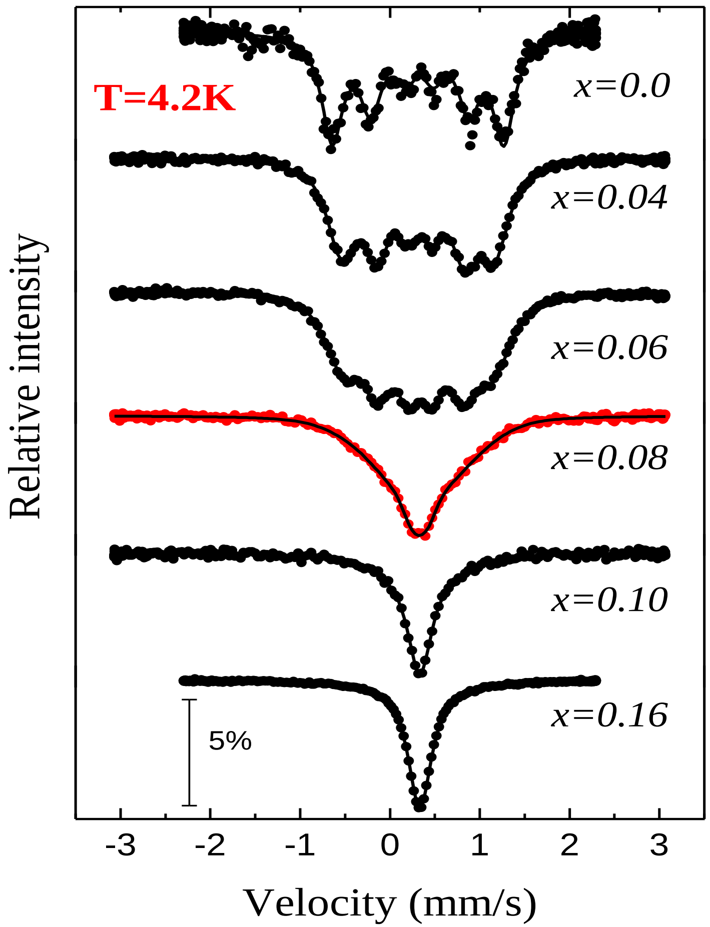
Results for in $CeFeAsO_{1-x}F_x$

Room temperature MS



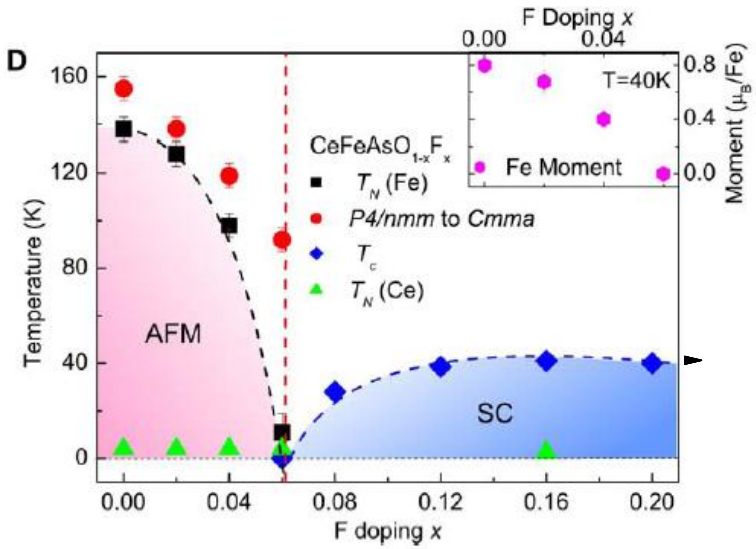
All spectra were fitted with single lines

4.2K MS



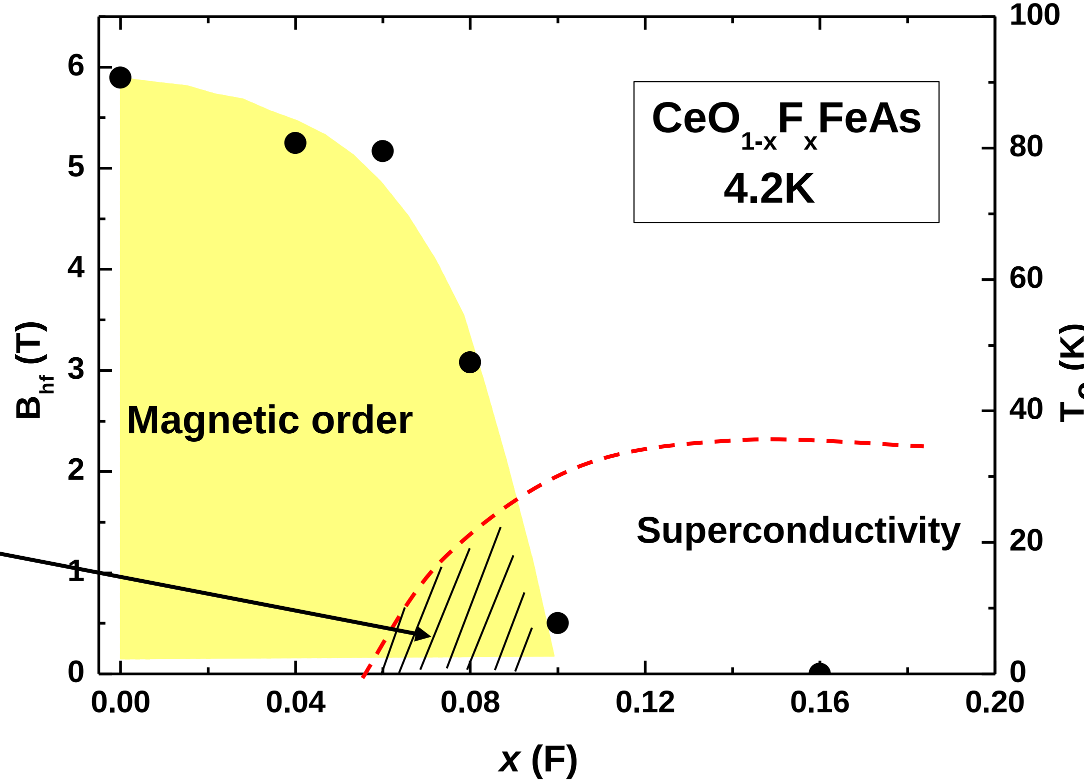
The Bhf field at Fe nucleus decreases with F content and disappears for x=0.16.
Magnetism suppressed by F doping.

CeFeAsO$_{1-x}$F$_x$ phase diagram



Neutron studies
[13] Jun Zhao, et al.,
Nature Materials 7, 953 - 959 (2008).

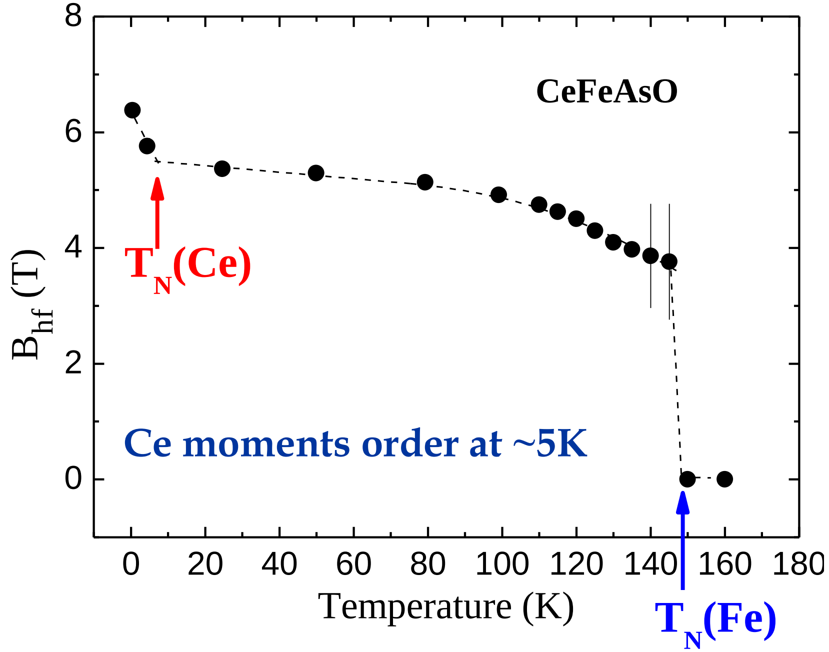
Mössbauer results shown a region of coexistence of superconductivity and magnetism (phase separation).



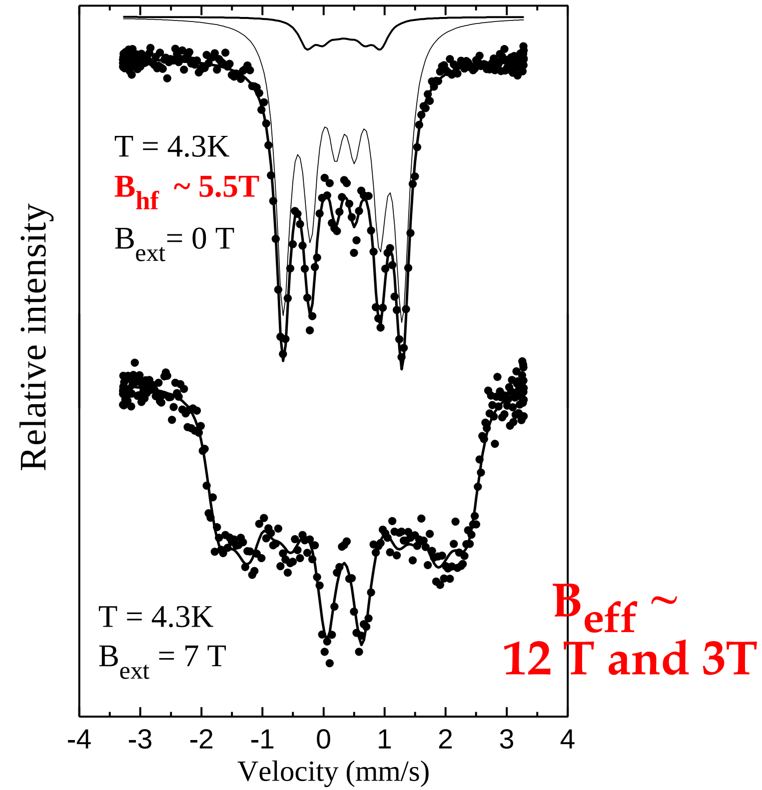
For CeOFeAs $\mu_{Fe} \approx 0.39\ \mu_B$

Results for CeFeAsO

MS with different T



CeFeAsO under applied external field



Fe spins ordering at ~145K lying in the (*a*,*b*) plane

MS support the AFM ordering of Fe spins

A fraction of Fe spins tend to align parallel (**B_{eff} ~ 12 T**) and the remaining ones antiparallel (**B_{eff} ~ 3 T**) to the external field

$$B_{eff} = B_{ext} + B_{hf}$$

The Bhf in **NdFeAsO** and **CeFeAsO** can be associated to SDW AFM order of Fe spins below ~140 and ~145K, as seen by neutrons

The AFM ordering of Nd and Ce spins seen by neutrons at ~2K and ~5K generate additional Bhf

***No* Bhf was observed at ^{57}Fe nucleus in $NdFeAsO_{0.88}F_{0.12}$ and $CeFeAsO_{0.84}F_{0.16}$ down 2K**

Doping with F suppresses both the magnetic order and structural distortion in favor of superconductivity

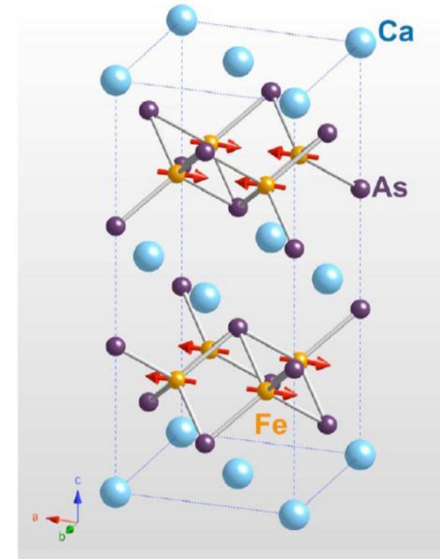
Fe magnetic moment estimated to be ~0.35 μ_B in NdFeAsO and ~0.39 μ_B in CeFeAsO. Similar to neutron data

Coexistence (phase separation) of SC and magnetism has been observed in $CeFeAsO_{1-x}F_x$ for $0.05 \leq x \leq 0.11$.

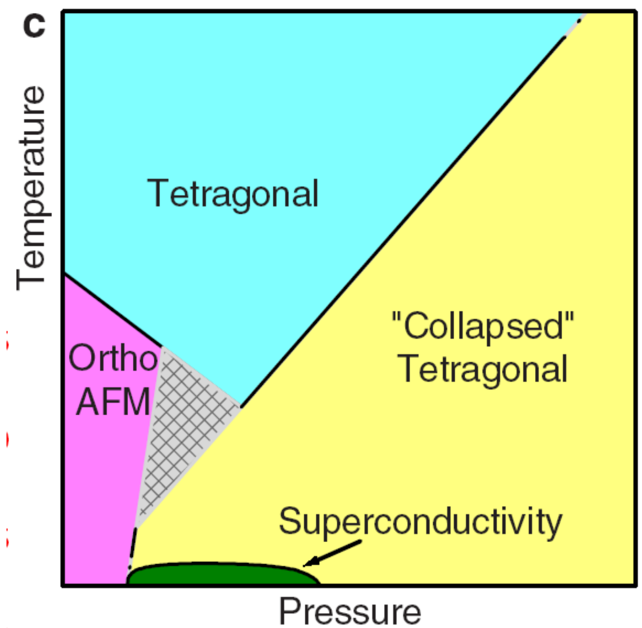
Ternary iron-arsenide $CaFe_2As_2$

- Structural transition (Tetr to orthorhombic) at ~ 170K

- Order of the Fe moments, in a commensurate AF structure.



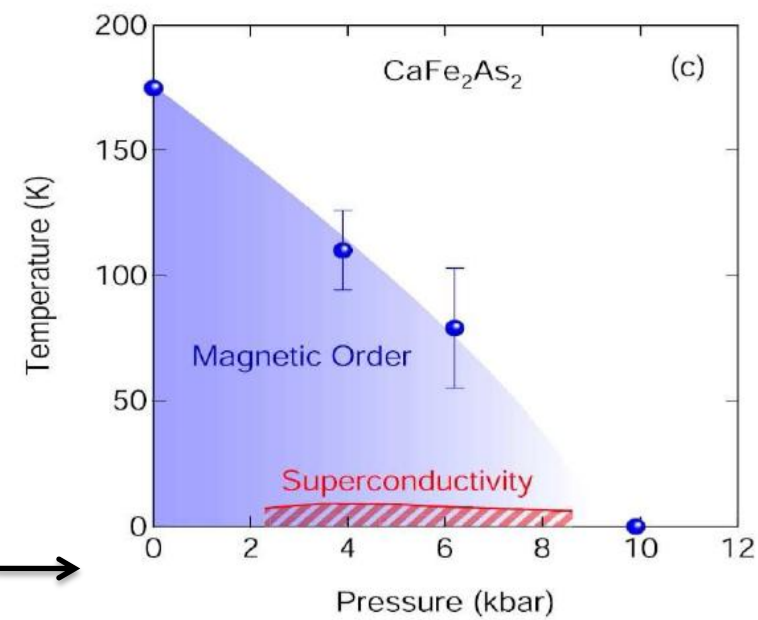
A.I. Goldman et al., Phys. Rev. B 78, 100506R (2008)



*Pressure Induced Superconductivity in $CaFe_2As_2$.

A. Kreyssig et al., Phys. Rev. B 78, 184517 (2008)
Milton S. Torikachvili et al., PRL 101, 057006 (2008)

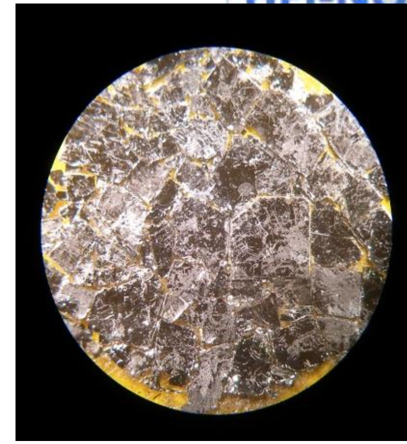
μSR →



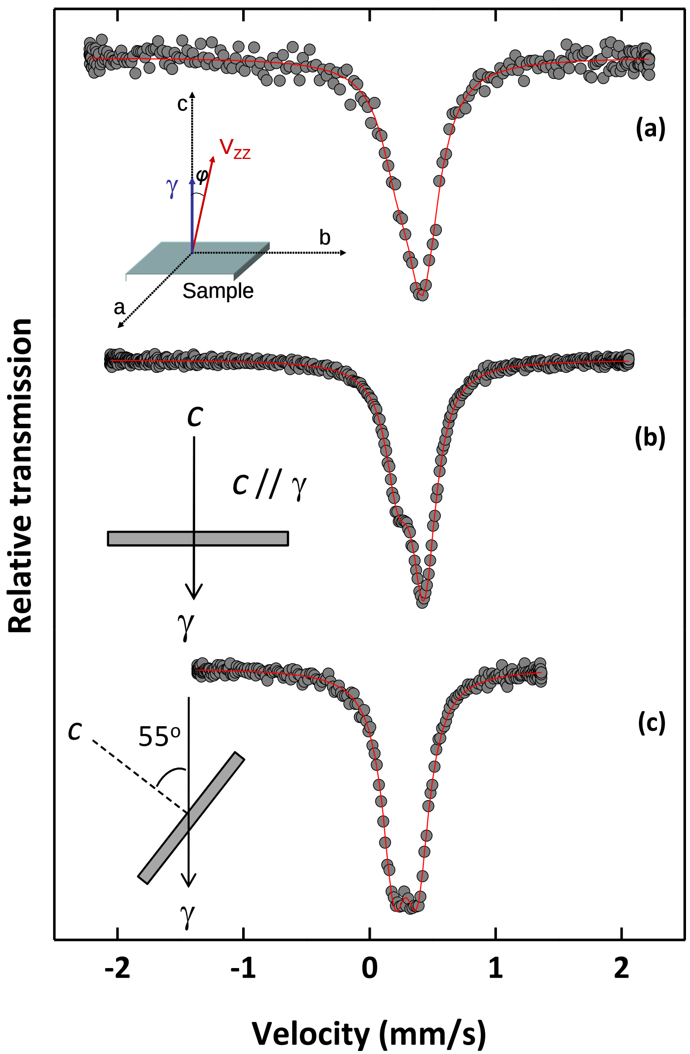
T. Goko, Uemura, Sanchez, Saitovitch et al. Phys. Rev. B78

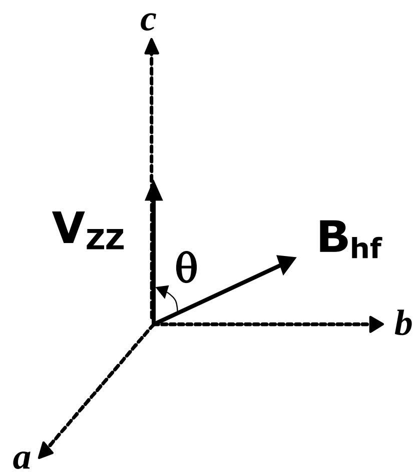
$$\frac{I_{\frac{3}{2}}(\varphi)}{I_{\frac{1}{2}}(\varphi)} = \frac{1+\cos^2\varphi}{\frac{2}{3}+\sin^2\varphi}$$

$CaFe_2As_2$ single crystal



Mariella

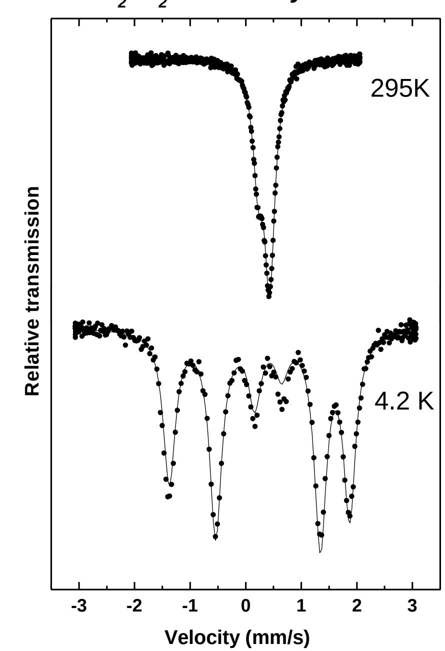




ΔE_Q=0.207 mm/s
IS=0.44 mm/s
Γ=0.26 mm/s
$\varphi \approx 10^o$

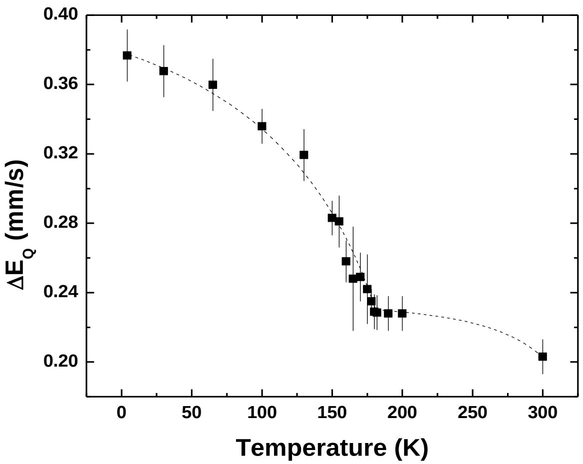
Only one site at low T

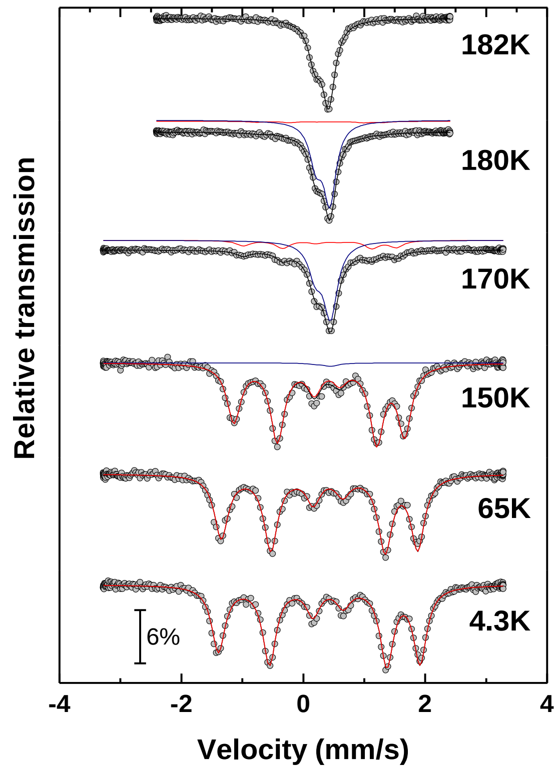
ΔE_Q=0.339 mm/s
B_{hf}= 9.95T
Angle between V_{zz} and B_{hf}: $\theta \approx 85^o \pm 5^o$

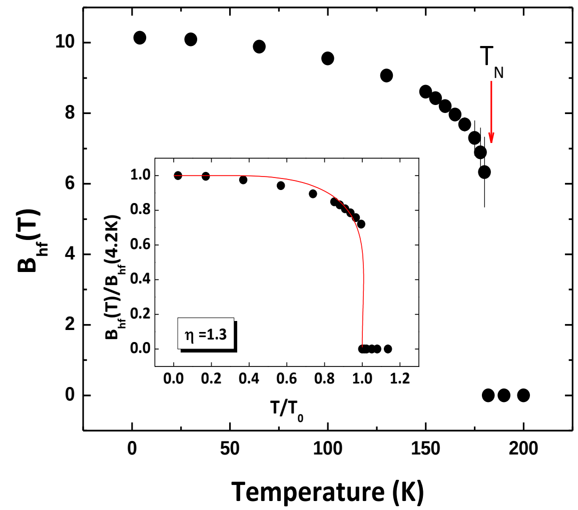


$\mu_{Fe} \approx 0.66\ \mu_B$ and lie in *a-b* plane
(agrees with neutron results)

$CaFe_2As_2$ (T)

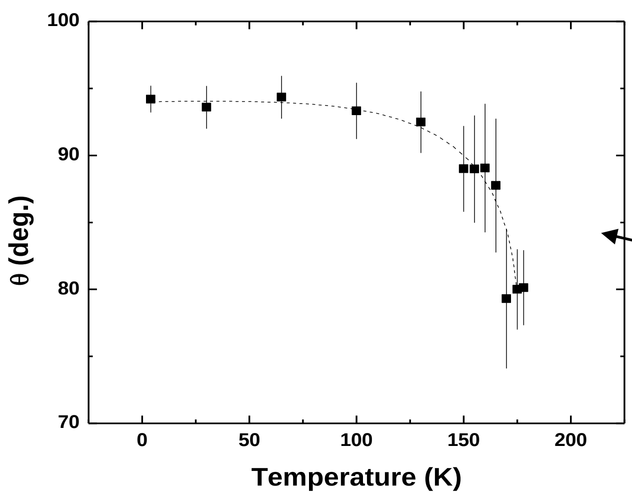






Structural transition

Jump in B_{hf}(T) at ~180K, typical for a first-order transition



commensurate AFM ordering with Fe spins lying in the (a,b) plane

V_{zz} in $CaFe_2As_2$ was found to be parallel to *c*.

Only one Fe site showing a first order transition at ~ 140K with the Fe spin lying in the (a,b) plane

Hole doped AFe_2As_2 (with K or Na) shows a phase separation at low temperature with a fraction of Fe ions in a paramagnetic state while remaining ones have a magnetic moment reduced related to undoped, order magnetically $T_O > T_c$.

This two phases are indistinguishable at room temperature

Same behavior is observed for Co doped $BaFe_2As_2$ with the difference that in this case the doping is being performed at the Fe site

In all the cases the doping distribution seems not to be homogeneous

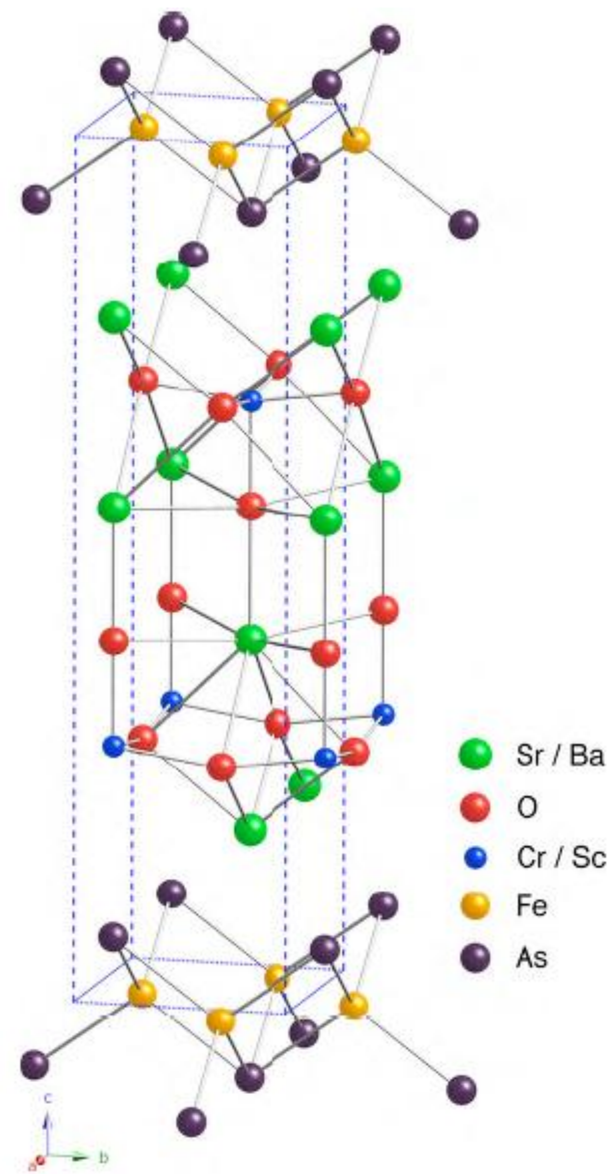
$Sr_4A_2O_6Fe_2As_2$ Iron Pnictides

- **(so-called 42622), with a perovskite layer between FeAs layers,**

  - **No SDW ordering**
  - **No structural transition**
  - **Large Fe-Fe interlayer distances (~15 Å)**

  - **T_C= 37 K for SrVOFeAs**
  - **0. 1 T transferred field to Fe in SrVOFeAs**

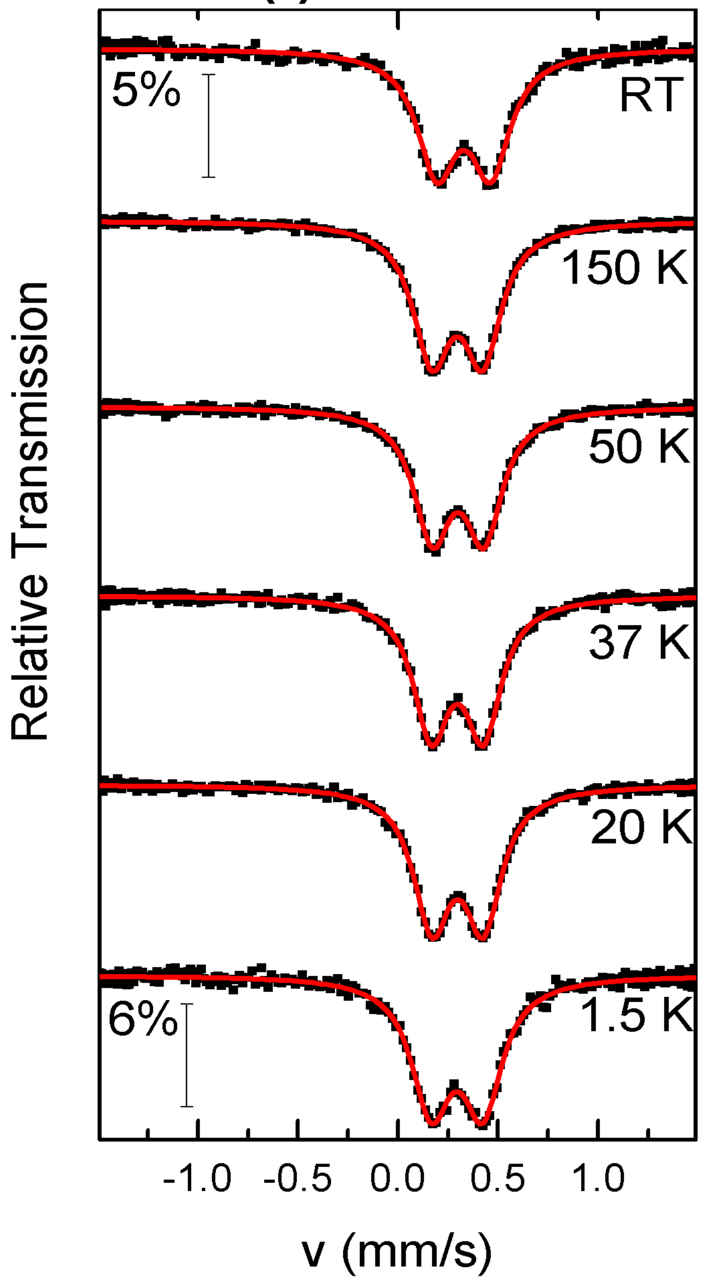
  - **No SDW in SrScOFeAs**

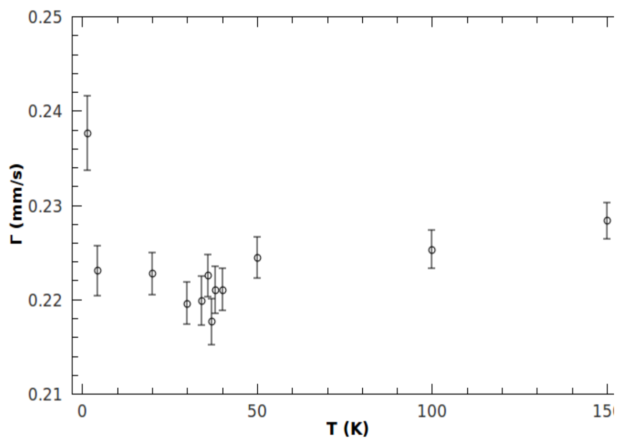


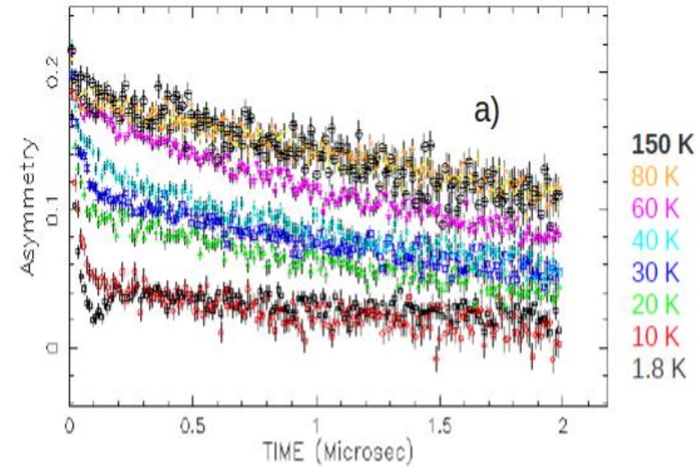
M. Tegel *et. Al, Zeitschrift fur anogarnische und allgemeine Chemie, V 635, 2242* (2009)
X. Zhu *et. Al,* Physical Review B 79, 220512(R), (2009)
Y. L. Xie *et al,* Europhysics Letters 86, 57007 (2009)

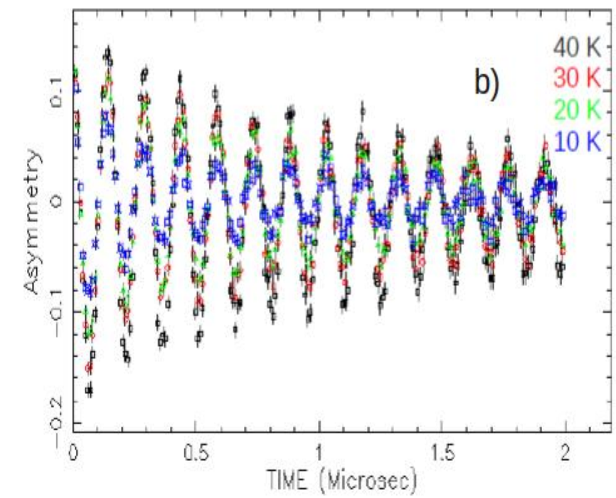
$Sr_4V_2O_6Fe_2As_2$

δ=0.297(2) mm/s ΔE_Q=-0.256(2) mm/s

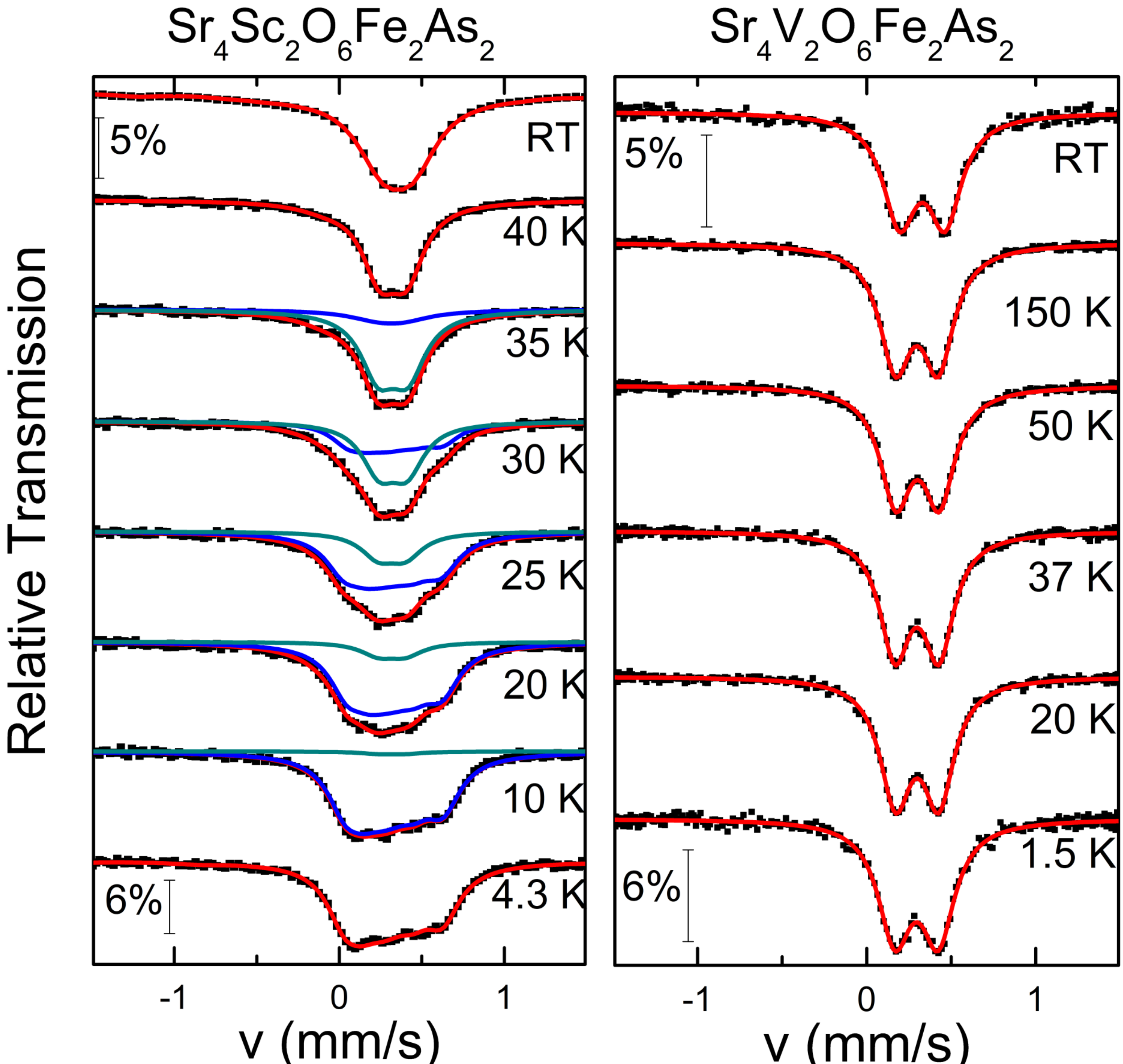






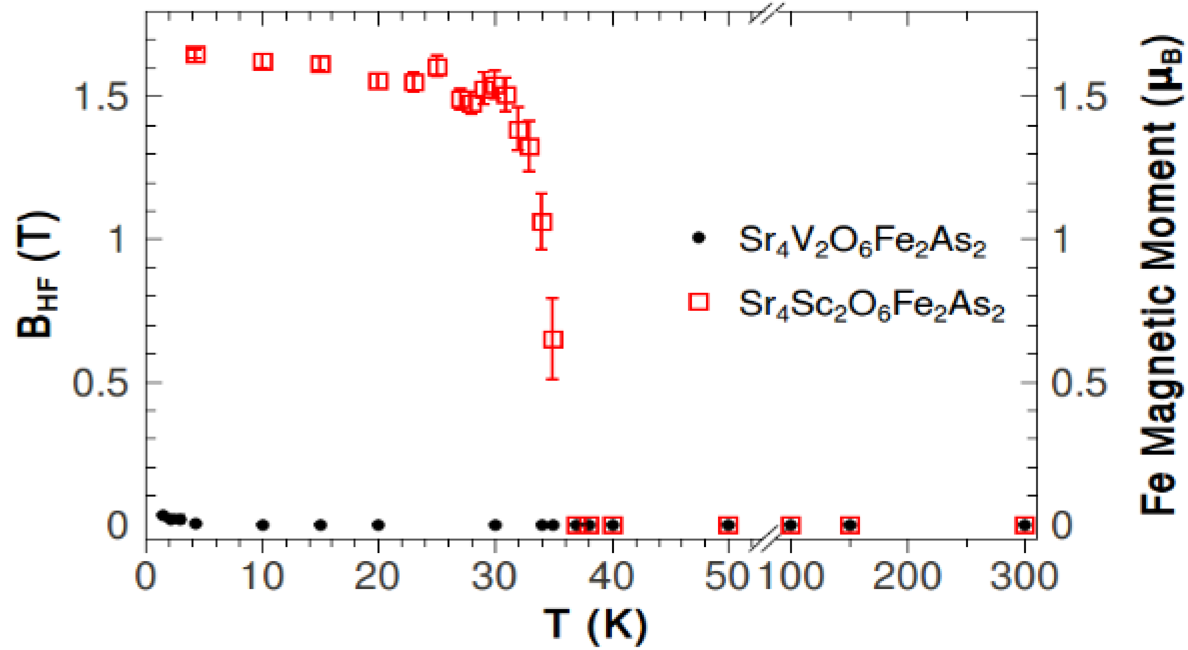


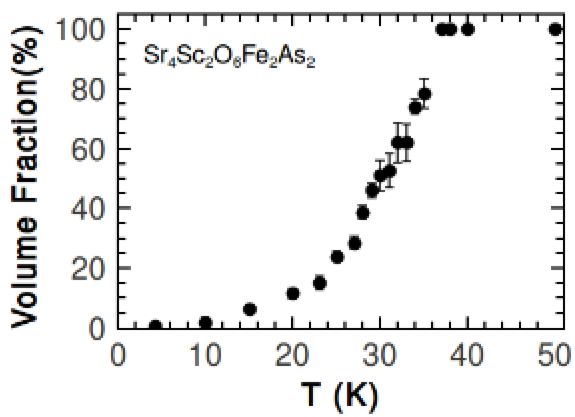
μSR show strong static magnetism below 100 K, increasing volume fraction for low temperature

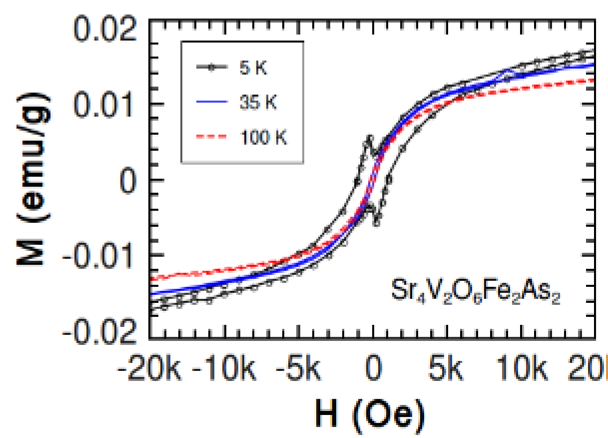


$Sr_4Sc_2O_6Fe_2As_2$
$Sr_4V_2O_6Fe_2As_2$
Relative Transmission
5%
RT
40 K
35 K
30 K
25 K
20 K
10 K
4.3 K
6%
5%
RT
150 K
50 K
37 K
20 K
1.5 K
6%
-1
0
1
v (mm/s)
-1
0
1
v (mm/s)

, Geneva


1.5
1
0.5
0
0
10
20
30
40
50
100
200
300
T (K)
B_{HF} (T)
Fe Magnetic Moment (μ_B)
$Sr_4V_2O_6Fe_2As_2$
$Sr_4Sc_2O_6Fe_2As_2$


$Sr_4Sc_2O_6Fe_2As_2$
Volume Fraction(%)
100
80
60
40
20
0
0
10
20
30
40
50
T (K)


5 K
35 K
100 K
$Sr_4V_2O_6Fe_2As_2$
M (emu/g)
0.02
0.01
0
-0.01
-0.02
-20k
-10k
-5k
0
5k
10k
20k
H (Oe)

- Possible coexistence of magnetic ordering in V atoms and superconductivity in FeAs layers in SrVOFeAs

- Magnetic ordering found for SrScOFeAs does not correspond to SDW or incommensurate-commensurate magnetic ordering

Perspectives for Mössbauer in the studies of Unconventional SC

Follow details at local level of magnetic transitions

Determine the structural phase transition and any special related feature

Thank you

FAPERJ, Pensa Rio, Infra estrutura , Cientista do Estado
PRONEX, CNPq (CIAM, DFG)
CAPES (PROBRAL)

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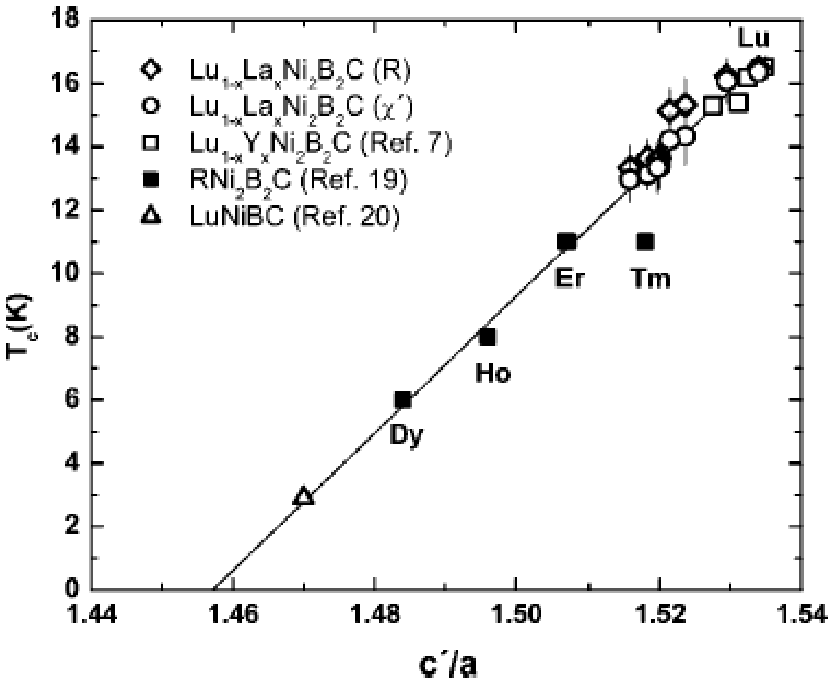
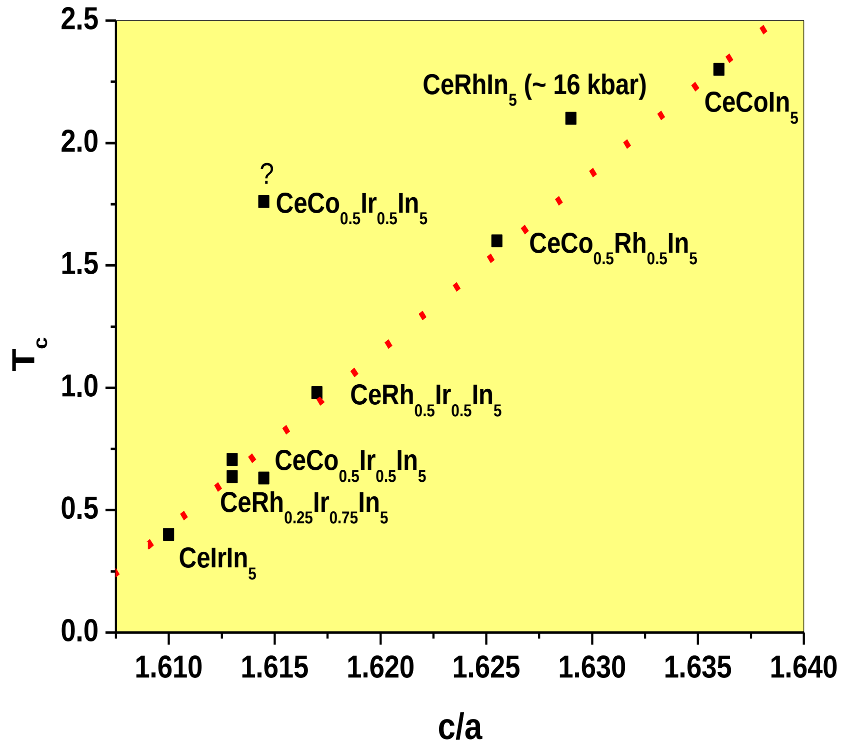
Tc versus c/a for RNi_nB_nC and for $CeMIn_5$

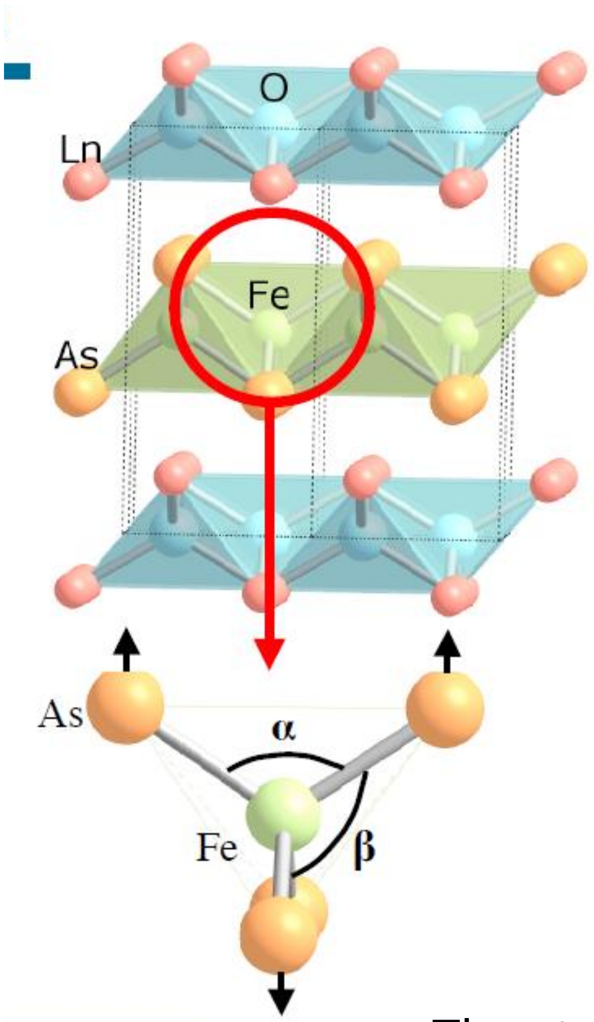




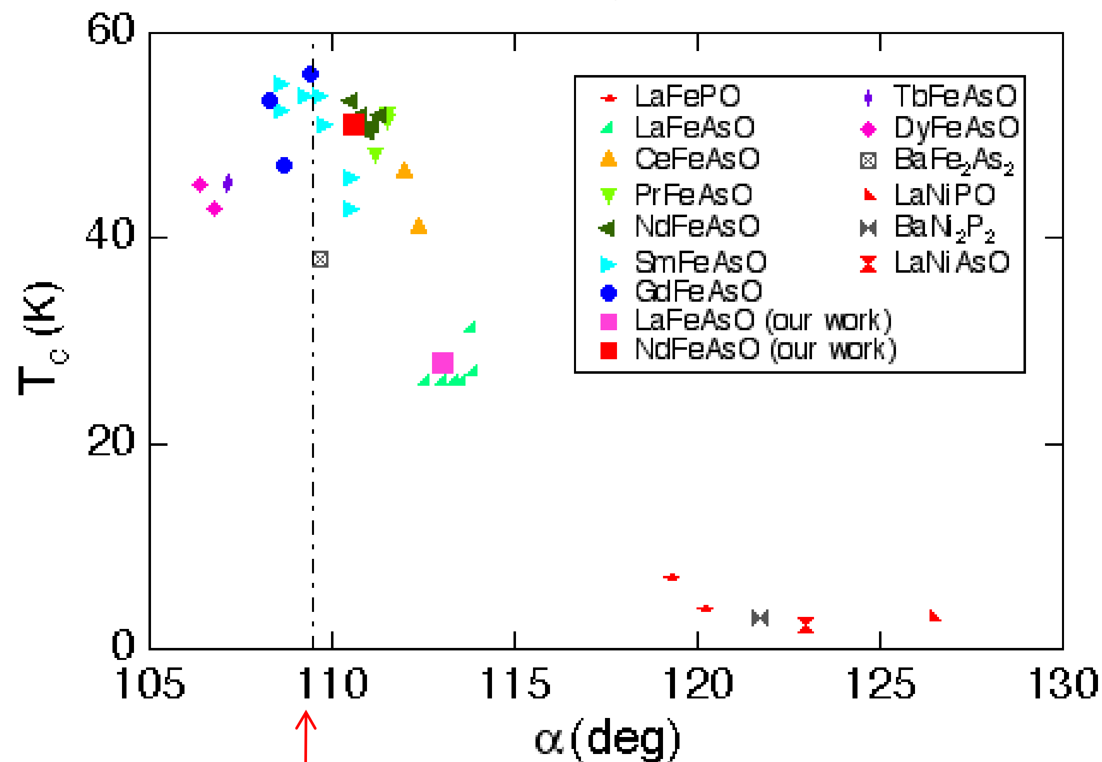
P. Pagliuso et al. Physica B 312–313 (2002) 129

E. Baggio-Saitovitch, D. R. Sánchez, H. Micklitz in *Rare Earth Transition Metal Borocarbides (Nitrides):*Kluwer Academic, Dordrecht, 2001,
D. R. Sánchez, E. M. Baggio-Saitovitch and H. Micklitz, P. R. B 71, 024509 (2005)

T_c *vs.* Ln relationship for $Ln\mathrm{FeAsO}_{1\text{-}y}$



Variation of T_c with BOND ANGLE

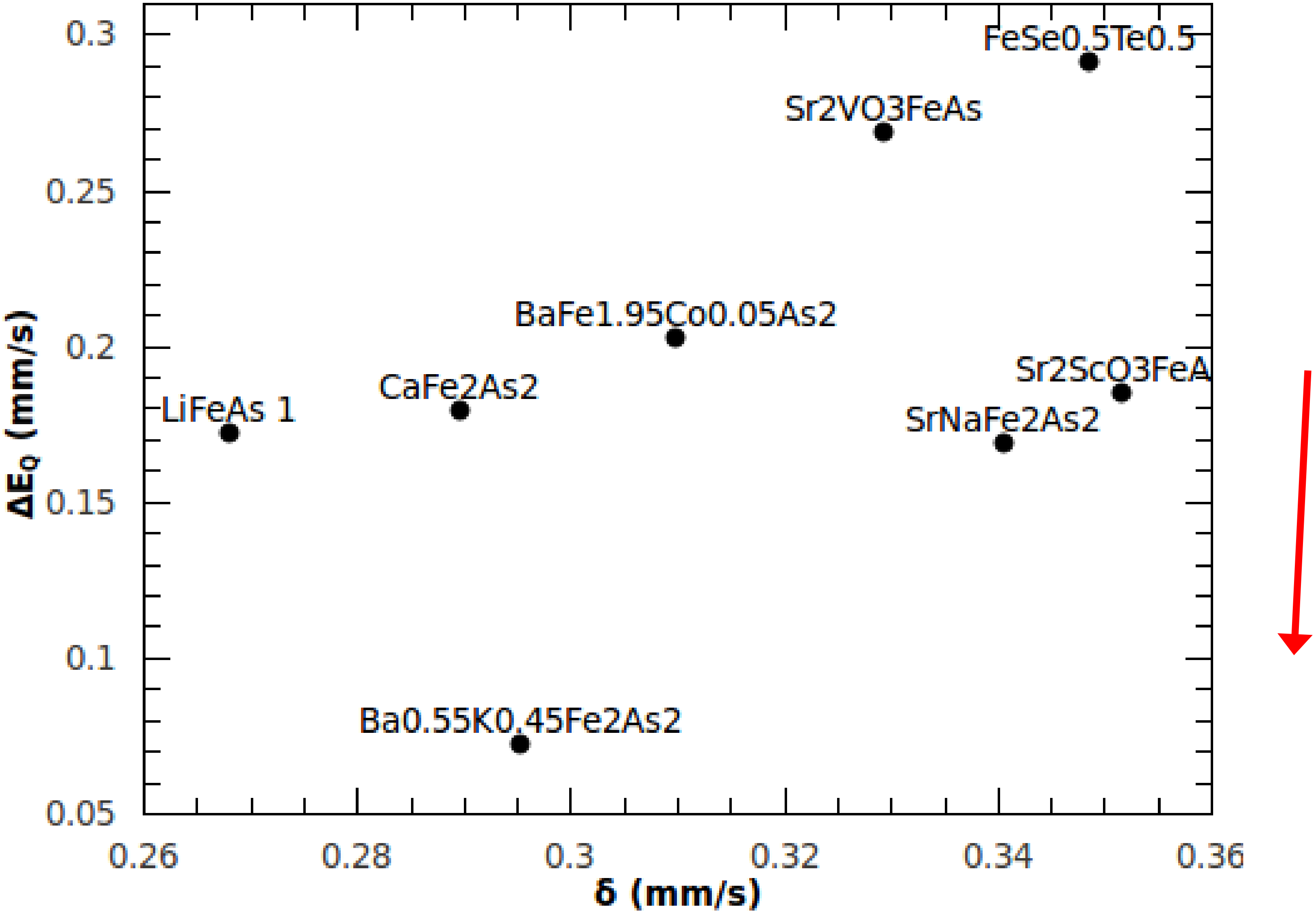


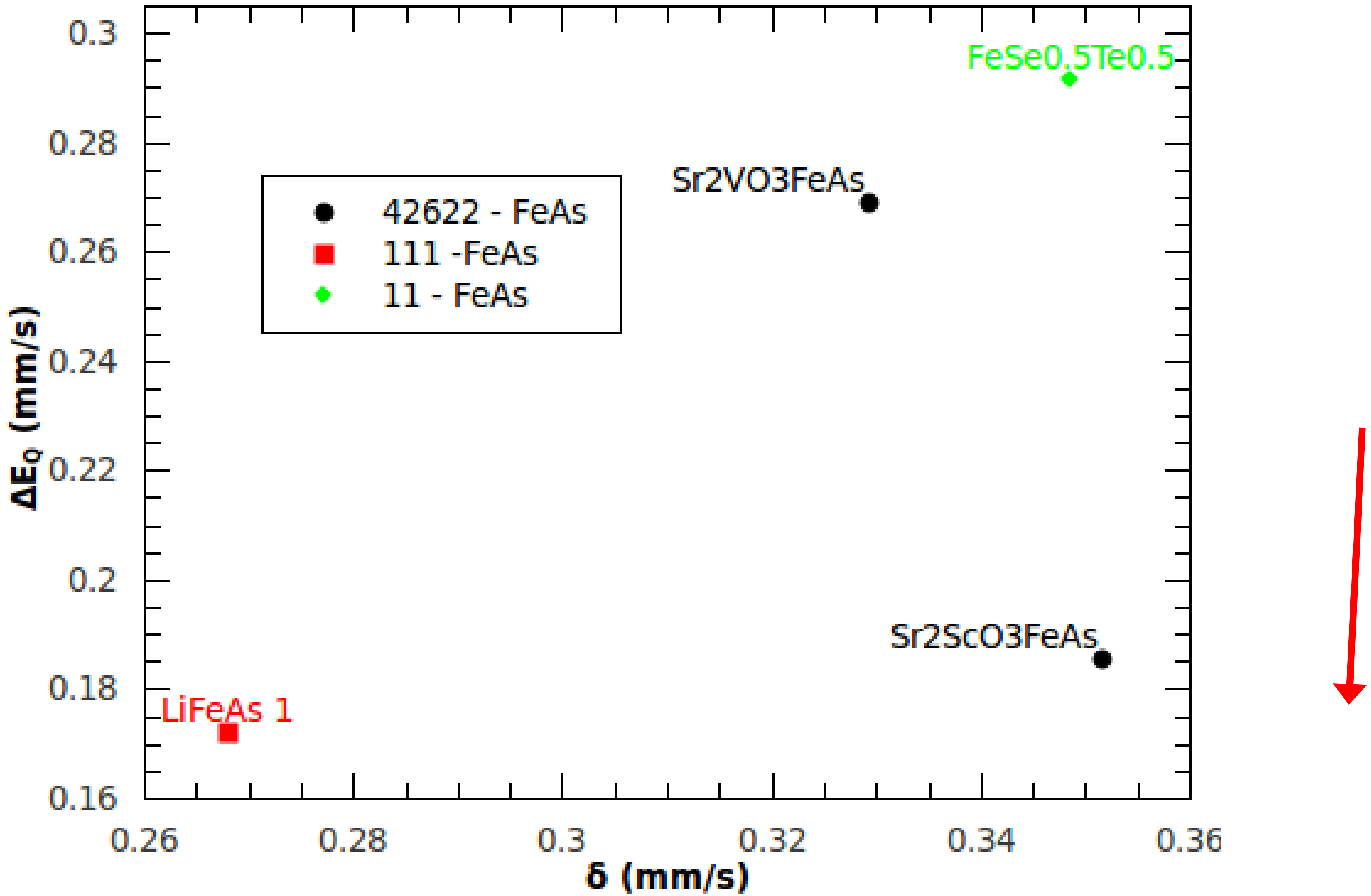
C. Lee *et al.*, JPSJ 77 (2008) 083704.

Ideal FeAs tetrahedral angle

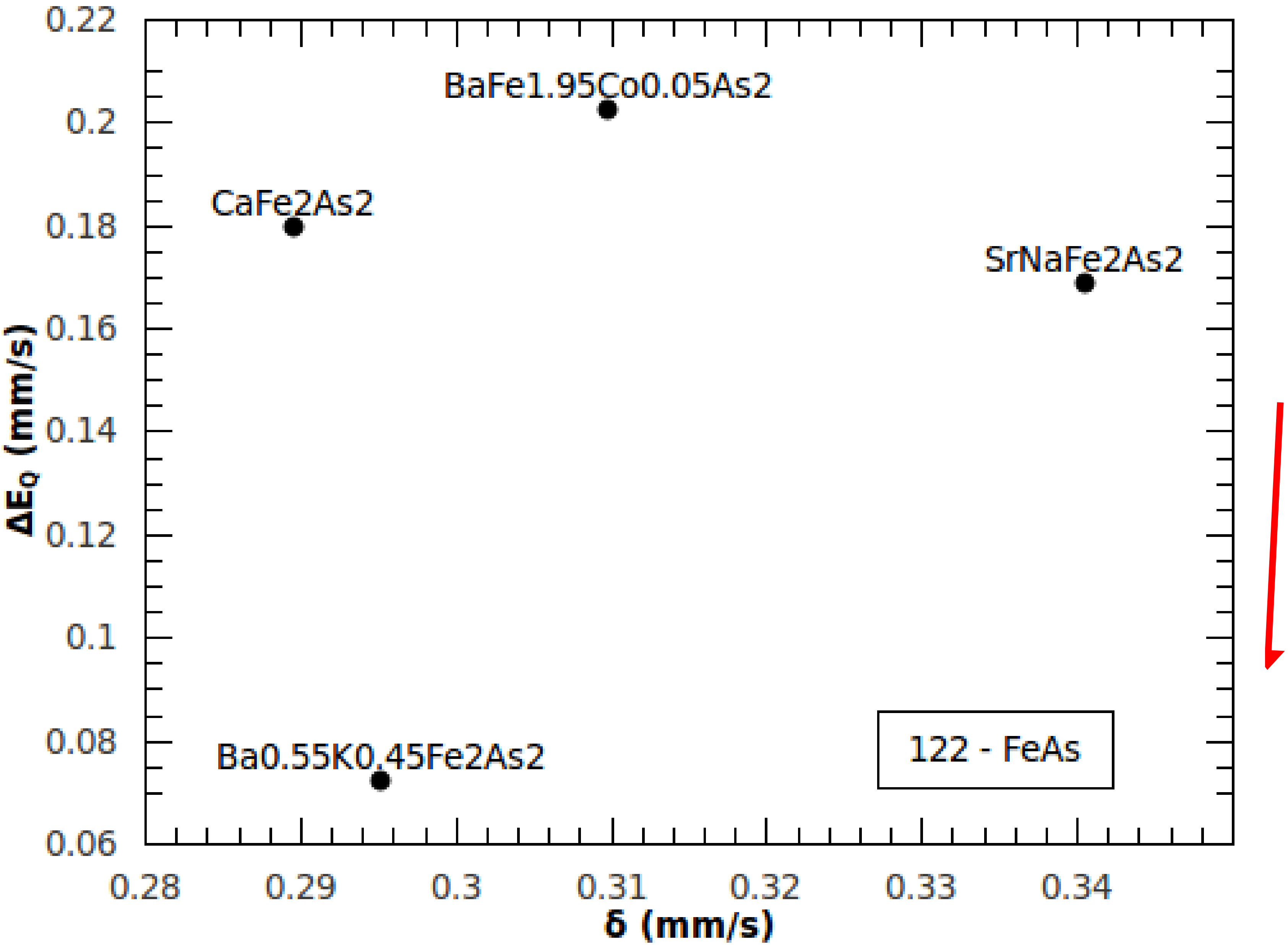
The structural distortion from the ideal FeAs tetrahedron is critical to the superconducting transition temperature

Fe is a Mössbauer probe

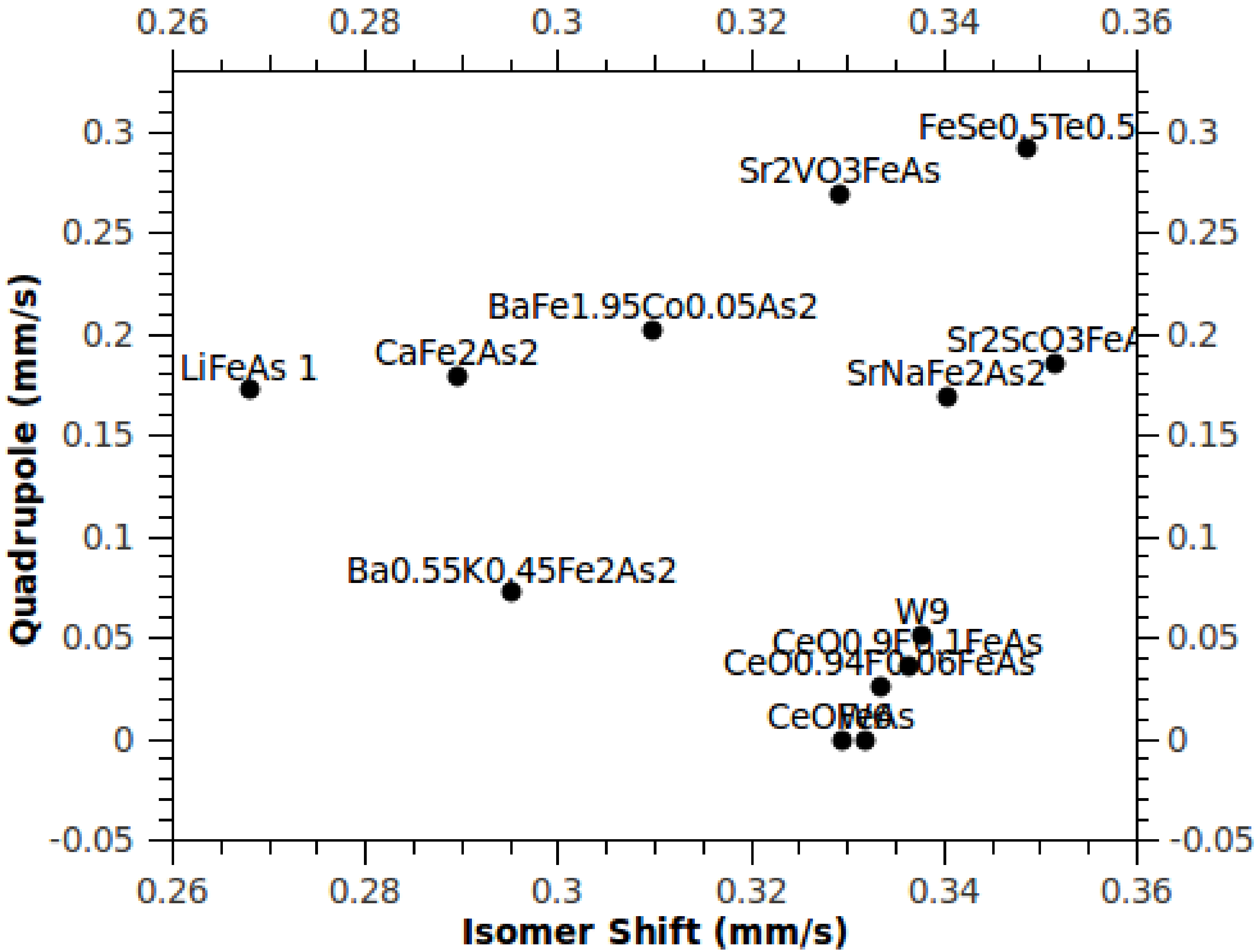

FeSe0.5Te0.5
Sr2VO3FeAs
BaFe1.95Co0.05As2
Sr2ScO3FeA
CaFe2As2
LiFeAs 1
SrNaFe2As2
Ba0.55K0.45Fe2As2
0.3
0.25
0.2
0.15
0.1
0.05
ΔEQ (mm/s)
0.26
0.28
0.3
0.32
0.34
0.36
δ (mm/s)



0.3
0.28
0.26
0.24
0.22
0.2
0.18
0.16
ΔE_Q (mm/s)
0.26
0.28
0.3
0.32
0.34
0.36
δ (mm/s)
42622 - FeAs
111 -FeAs
11 - FeAs
FeSe0.5Te0.5
Sr2VO3FeAs
Sr2ScO3FeAs
LiFeAs 1



0.22
0.2
0.18
0.16
0.14
0.12
0.1
0.08
0.06
ΔE_Q (mm/s)
BaFe1.95Co0.05As2
CaFe2As2
SrNaFe2As2
Ba0.55K0.45Fe2As2
122 - FeAs
0.28
0.29
0.3
0.31
0.32
0.33
0.34
δ (mm/s)



Quadrupole (mm/s)
Isomer Shift (mm/s)
FeSe0.5Te0.5
Sr2VO3FeAs
BaFe1.95Co0.05As2
CaFe2As2
LiFeAs 1
Sr2ScO3FeA
SrNaFe2As2
Ba0.55K0.45Fe2As2
W9
CeO0.9F0.1FeAs
CeO0.94F0.06FeAs
CeOFeAs
0.26
0.28
0.3
0.32
0.34
0.36
0.3
0.25
0.2
0.15
0.1
0.05
0
-0.05

$NdFeAsO_{0.88}F_{0.12}$

ΔE_Q=0.02(2) *mm/s*

IS=*0.437(1) mm/s,*

$Ba_{0.5}K_{0.5}Fe_2As_2$

Room temperature

ΔE_Q=0.08 mm/s

IS=0.37 mm/s

Γ=0.22 mm/s

$V_{zz} \parallel c$

$CaFe_2As_2$

ΔE_Q=0.207 mm/s

IS=0.44 mm/s

Γ=0.26 mm/s

$\varphi \approx 10^o$

$Sr_4Sc_2O_6Fe_2As_2$

IS=0.465(3) mm/s

EQ=0.186(2) mm/s

$Sr_4V_2O_6Fe_2As_2$

IS=0.411(3) mm/s

EQ=0.256(2) mm/s