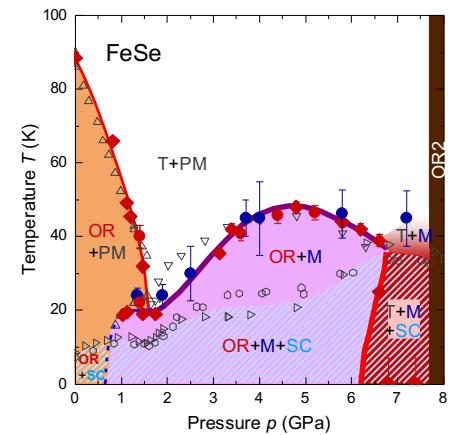
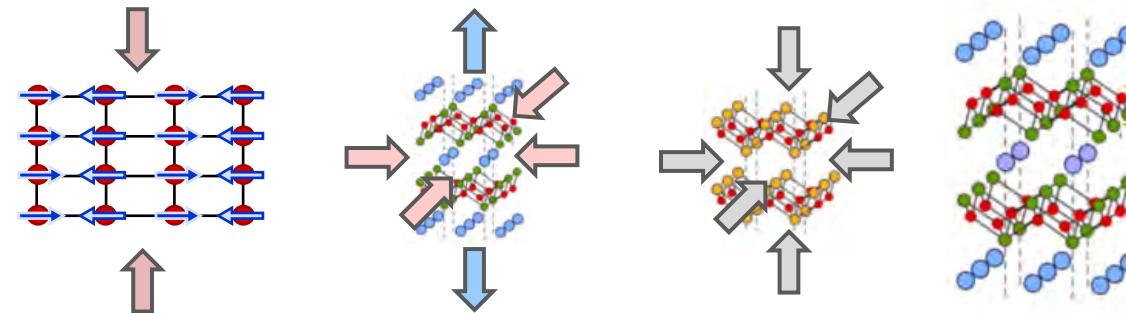
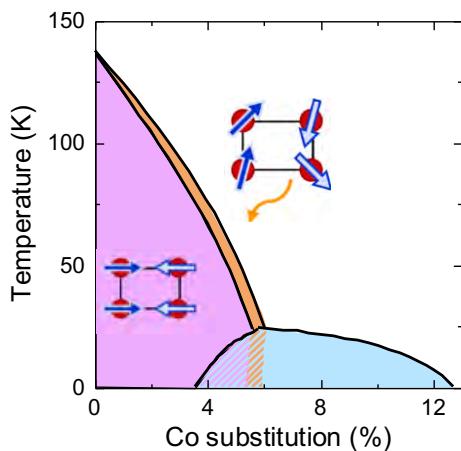


# Tuning magnetism in iron-based superconductors

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# Thanks to

Liran Wang

Frédéric Hardy

Thomas Wolf

Hilbert von Löhneysen

Christoph Meingast

**IFP, Karlsruhe Institute of Technology**

Jörg Schmalian

**Karlsruhe Institute of Technology**

Rafael Fernandes

**University of Minnesota, Minneapolis**

William Meier

Li Xiang

Elena Gati

Valentin Taufour

Sergey Bud'ko

Paul Canfield

**Ames Laboratory, Iowa State University**

Karunakar Kothapalli

Aashish Sapkota

Pinaki Das

Ben Ueland

Andreas Kreyssig

Alan Goldman

Qing-Ping Ding

Paul Wiecki

Yuji Furukawa

Herman Suderow

**Univ. Autonoma, Madrid**

**IOWA STATE  
UNIVERSITY**



## Funding:

*Helmholtz Association: Young Investigator Group, VH-NG 1242*

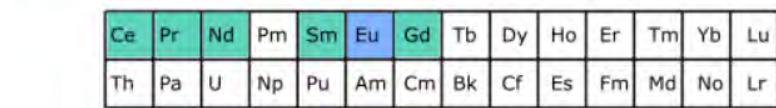
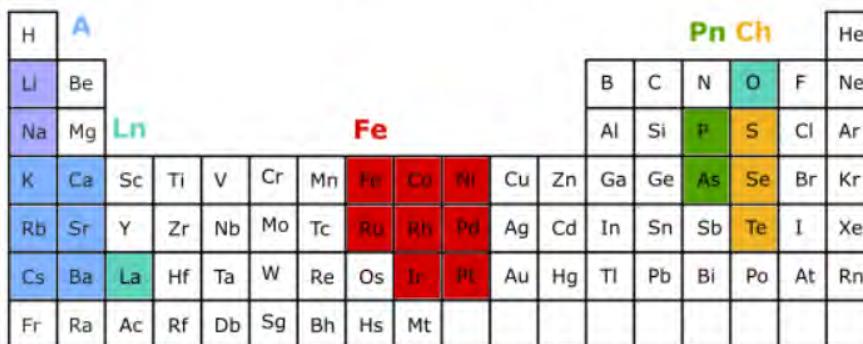
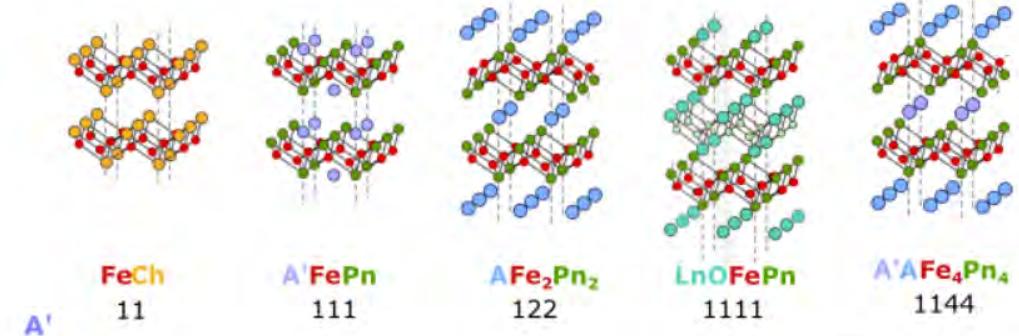
*Ames Laboratory, US DOE, under contract No. DE-AC02-07CH11358*



**HELMHOLTZ**  
RESEARCH FOR GRAND CHALLENGES

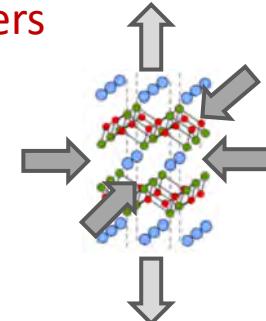
# Materials, methods and phenomena

## Iron-based materials

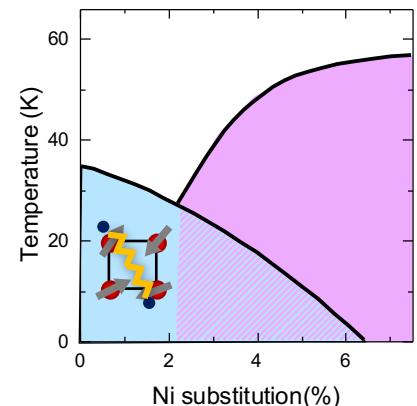
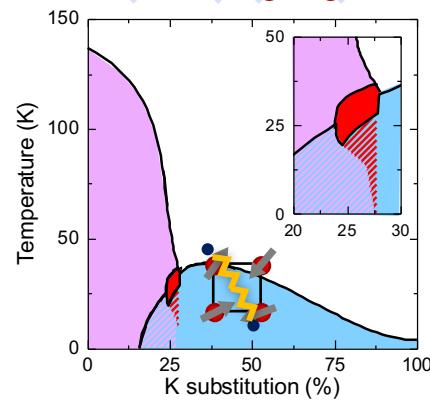
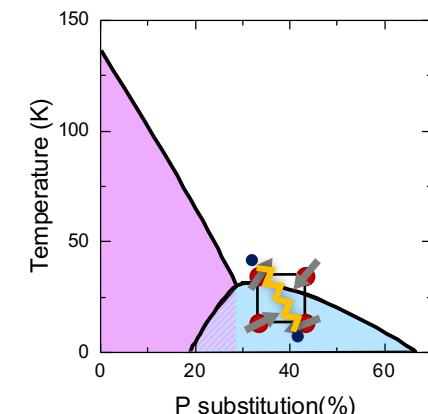
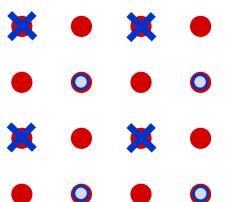
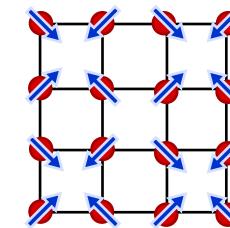
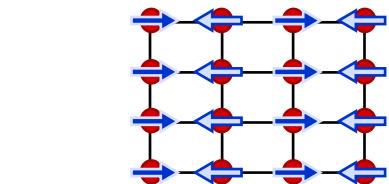


A. Böhmer, A. Kreyssig,  
Phys. Unserer Zeit **48**, 70 (2017)

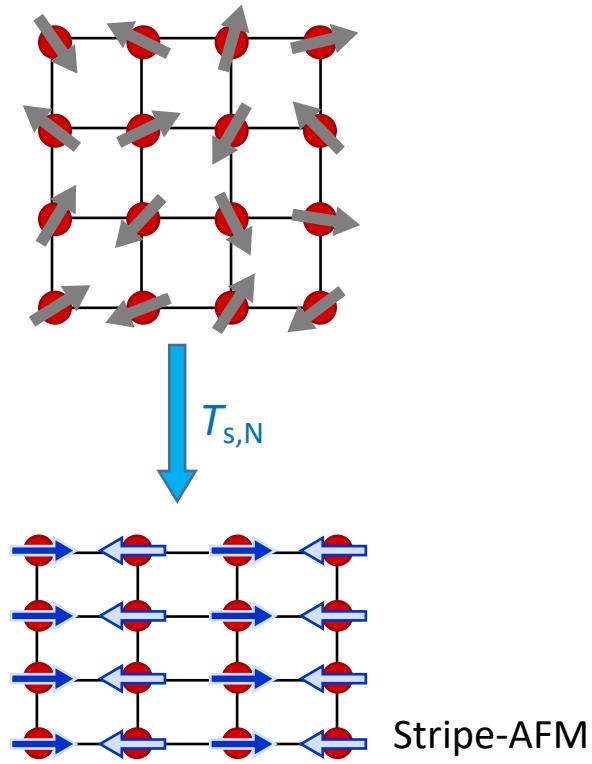
## Tuning parameters



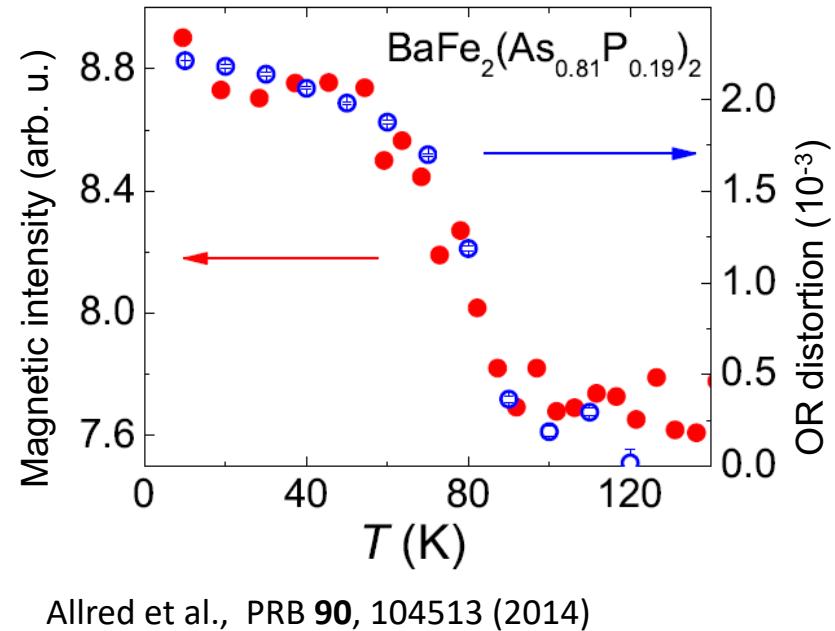
## Phases



# Magnetoelastic coupling

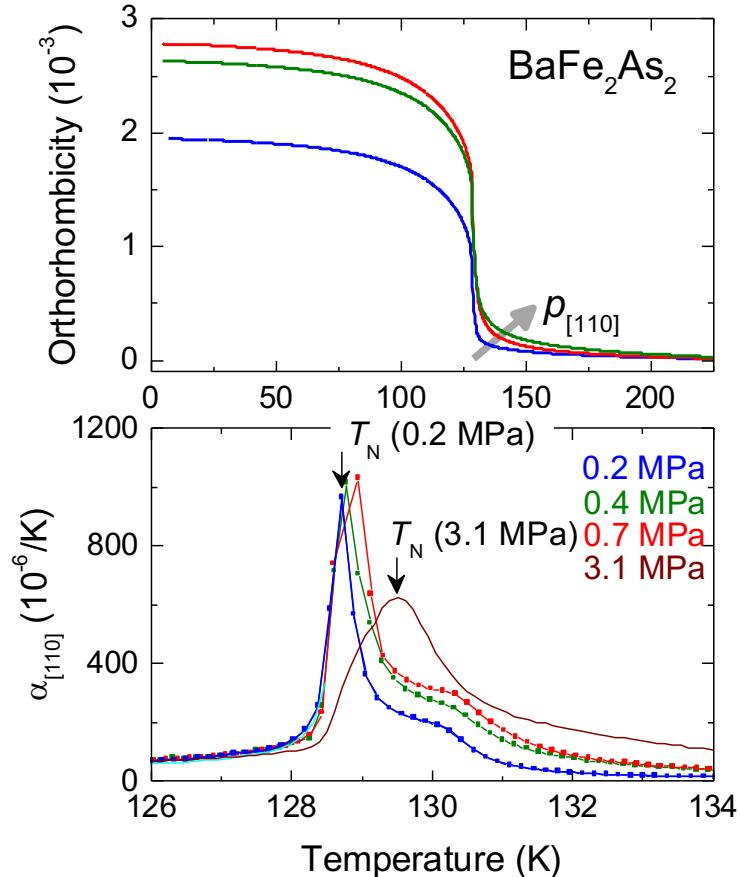
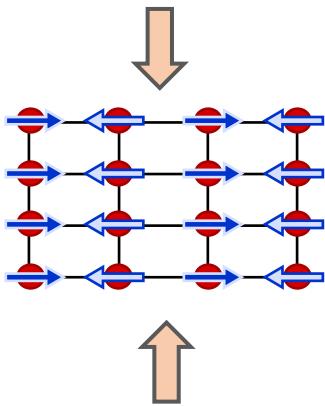


Stripe-type antiferromagnetism couples directly to structural distortion.  
Facilitates elastic tuning of magnetic order.

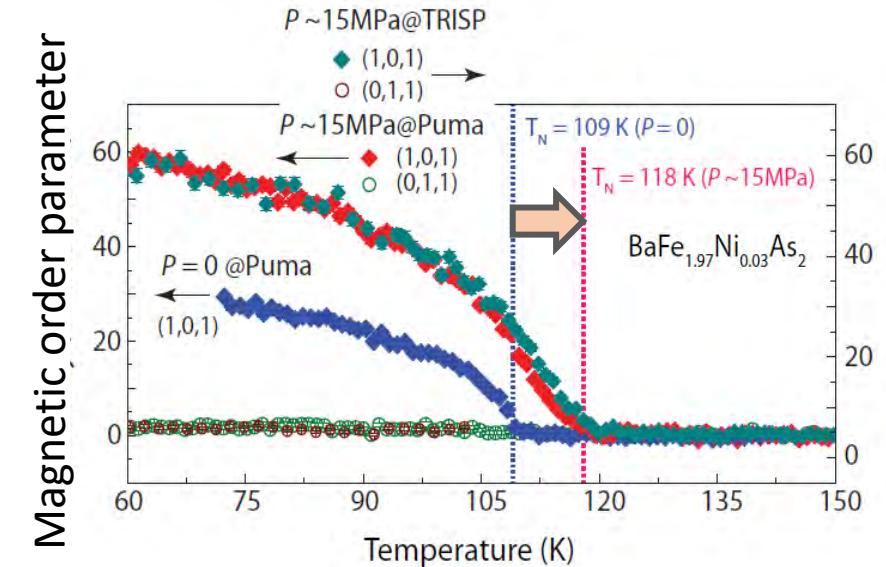


Allred et al., PRB **90**, 104513 (2014)

# Tune magnetic transition via uniaxial stress



Böhmer et al., unpublished

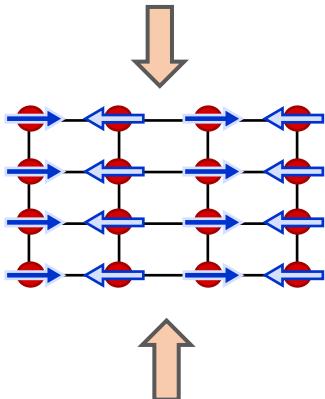


Lu et al., PRB **93**, 134519 (2016)

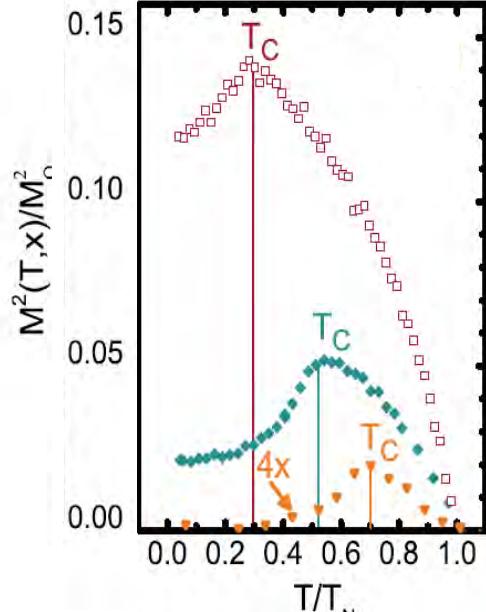


Tuning with  
uniaxial pressure

# Effect on superconductivity

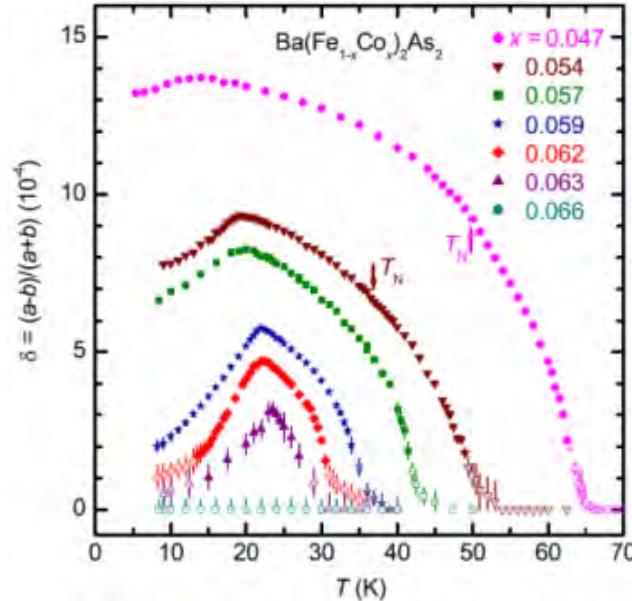


Ordered magnetic moment



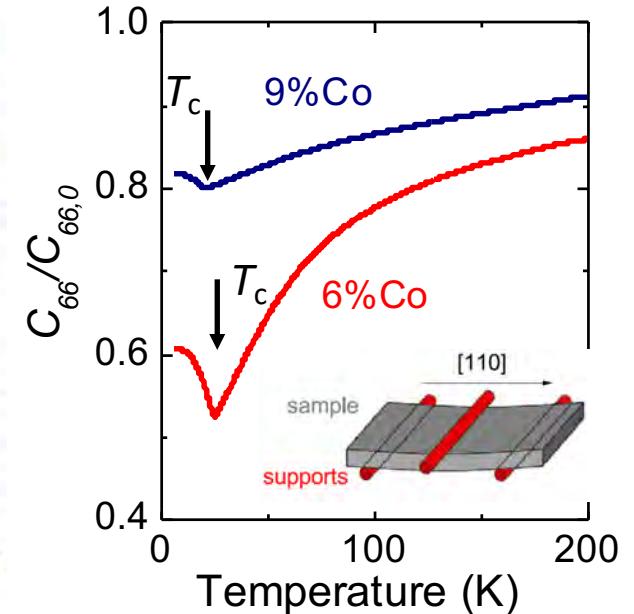
Fernandes et al., PRB 2010

Orthorhombic distortion



Nandi et al., PRL 104, 057006 (2010)

Elastic shear modulus



Böhmer et al., PRL 112, 047001 (2014)

- Competition between magnetism and superconductivity for the same electrons
- Results in a huge effect of superconductivity on lattice constants and elastic modulus
- $T_{sc}$  highly sensitive to uniaxial deformation:

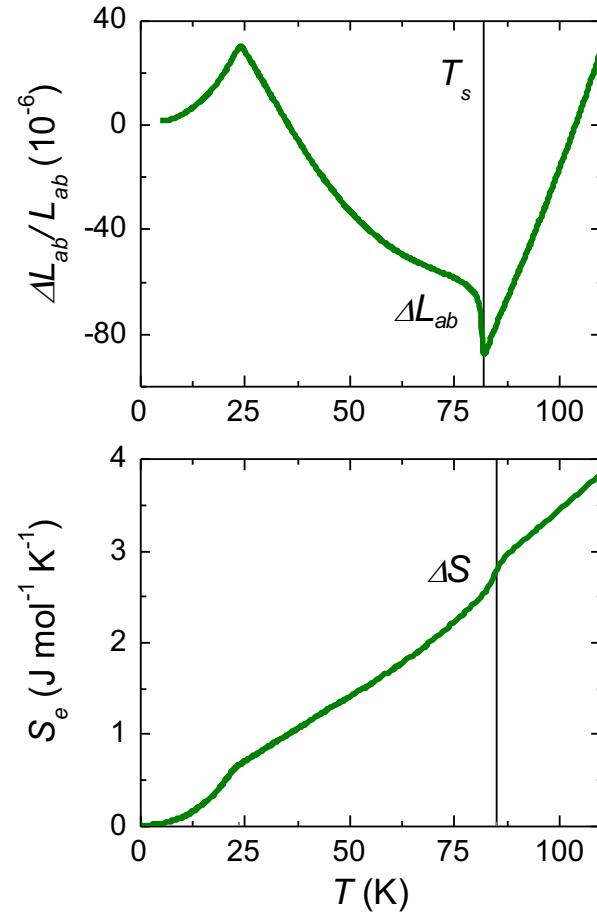
$$\frac{dT_{sc}}{d\varepsilon_a} - \frac{dT_{sc}}{d\varepsilon_b} \sim 9 \text{ K}/10^{-3} \text{ in } \text{Ba(Fe}_{0.955}\text{Co}_{0.045})\text{As}_2 \text{ with } T_{sc}=14 \text{ K}$$

For tetragonal samples, see:  
Malinowski et al., arxiv: 1911.03390

# What will anisotropic (symmetry-conserving) pressure do?

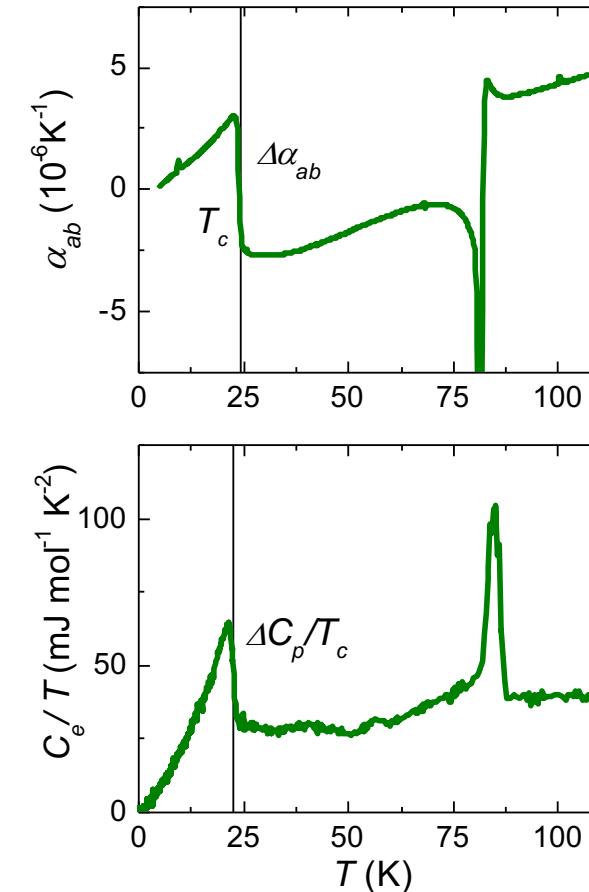
Clausius-Clapeyron  
(1st order transitions)

$$\frac{dT}{dp_i} = V_m \frac{\Delta L_i/L_i}{\Delta S}$$

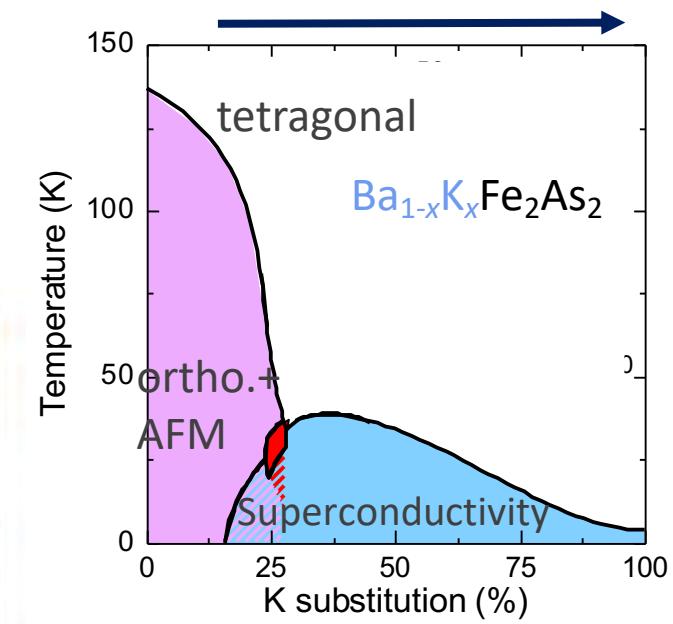
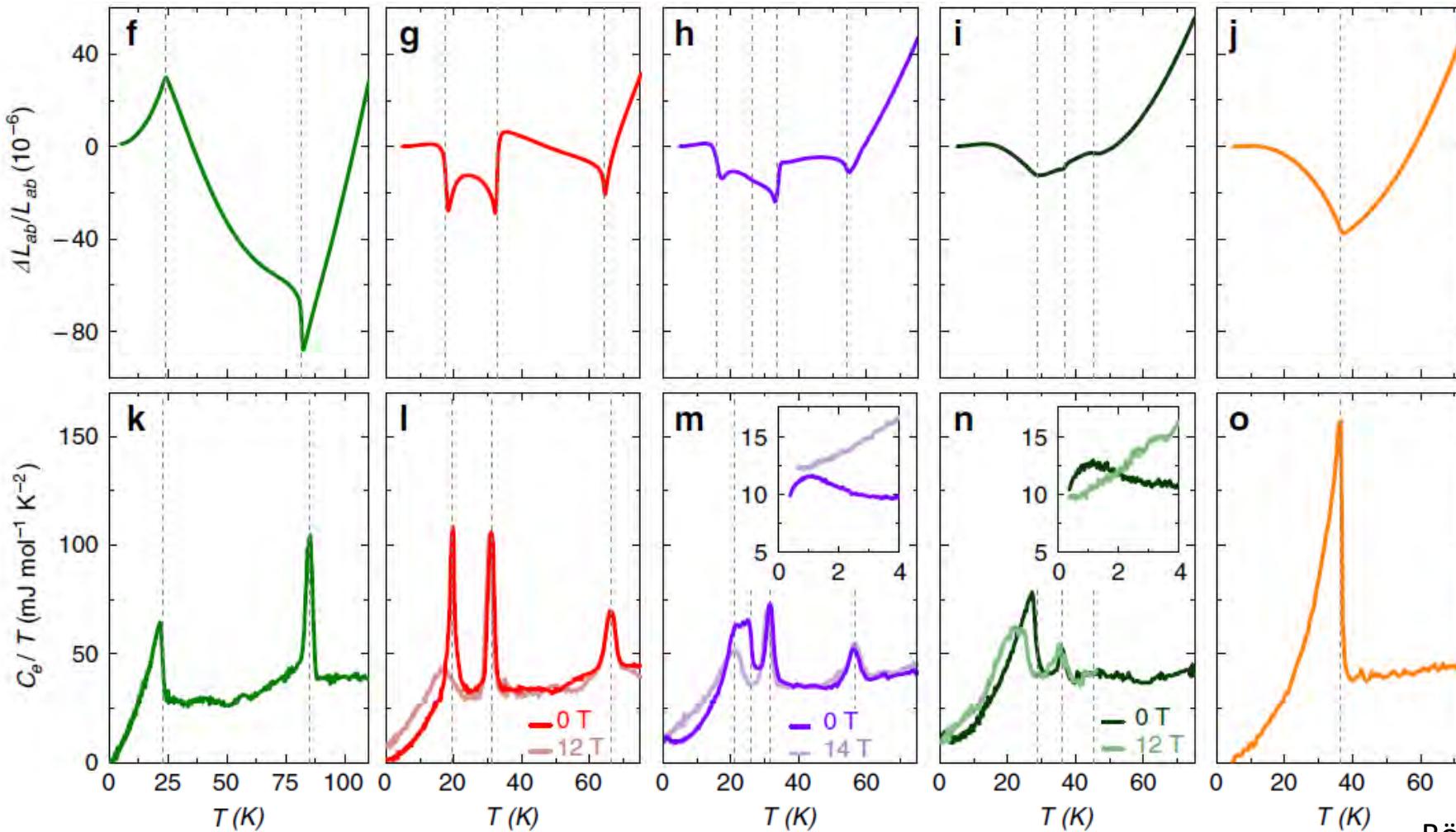


Ehrenfest  
(2nd order transitions)

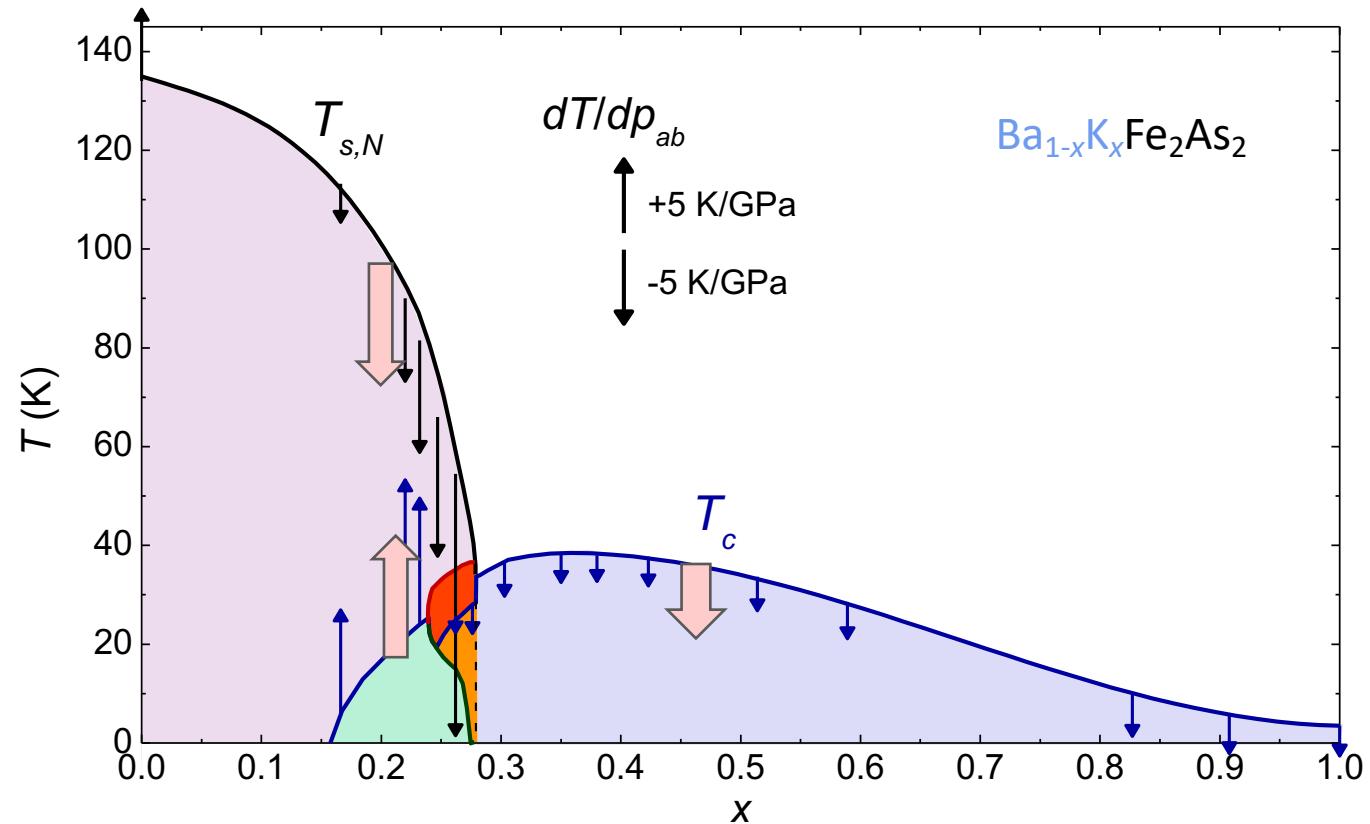
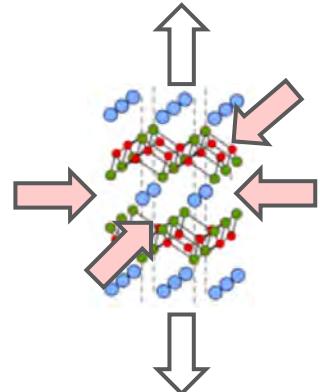
$$\frac{dT}{dp_i} = V_m \frac{\Delta \alpha_i}{\Delta C_p/T_c}$$



# Pressure derivatives in hole-doped $\text{Ba}_{1-x}\text{K}_x\text{Fe}_2\text{As}_2$

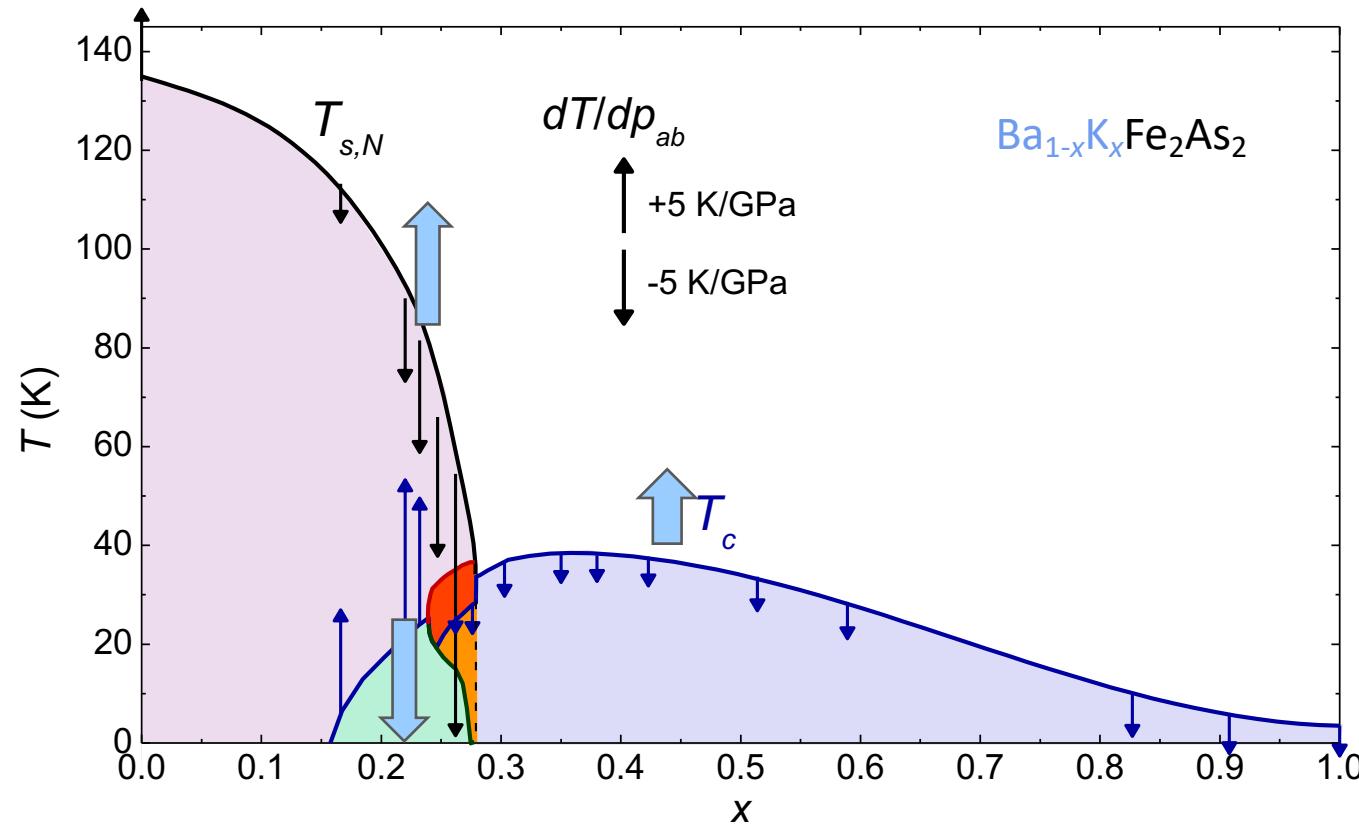


# Effect of biaxial in-plane pressure

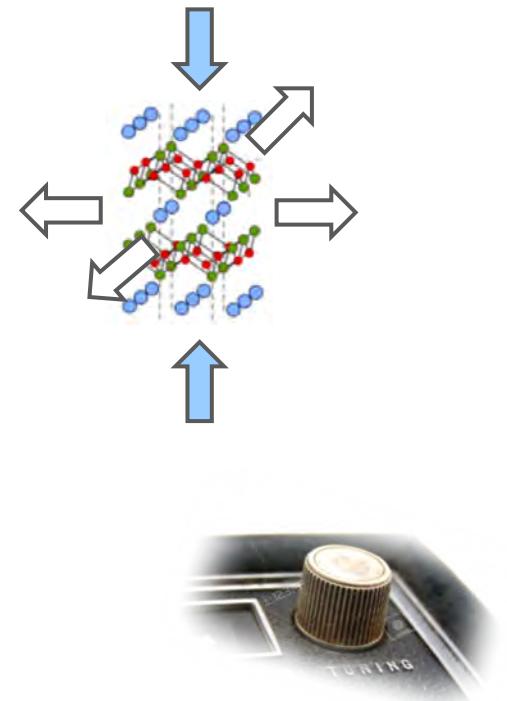


Opposite pressure derivatives of  $T_{\text{AFM}}$  and  $T_{\text{sc}}$ : one phase is stabilized at the expense of the other

# Effect of *c*-axis pressure



Opposite sign of pressure derivatives  $dT/d\sigma_{ab}$  and  $dT/d\sigma_c$

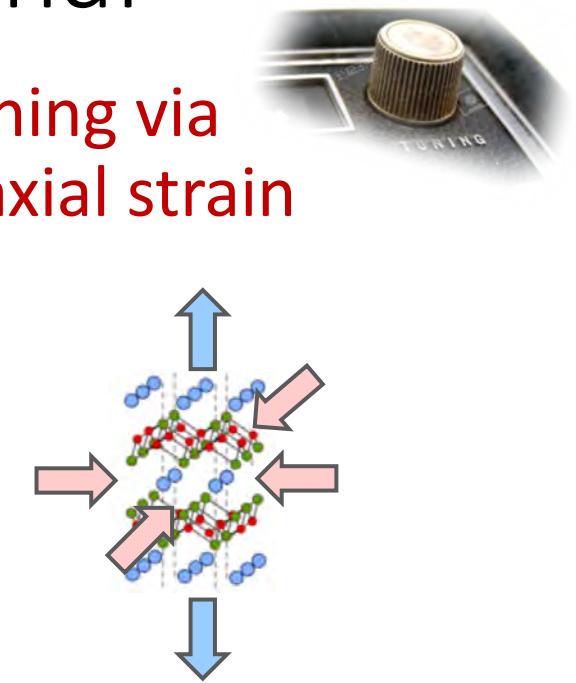
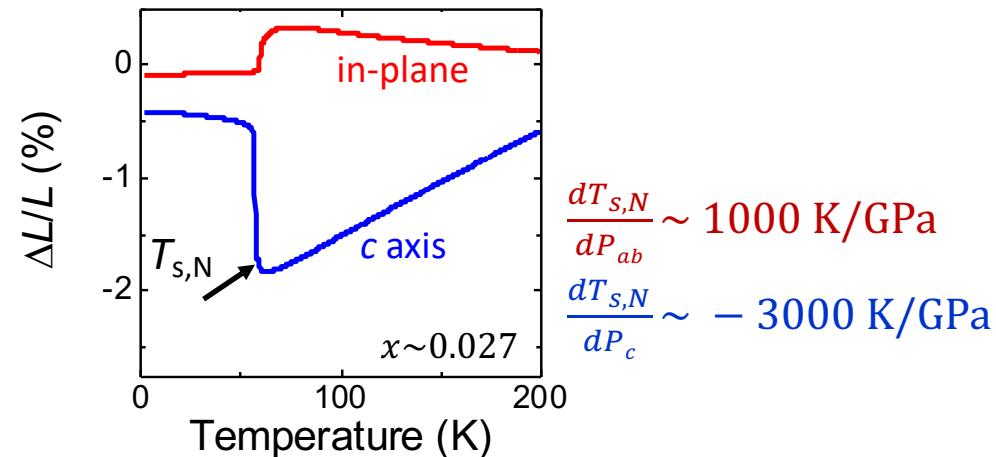
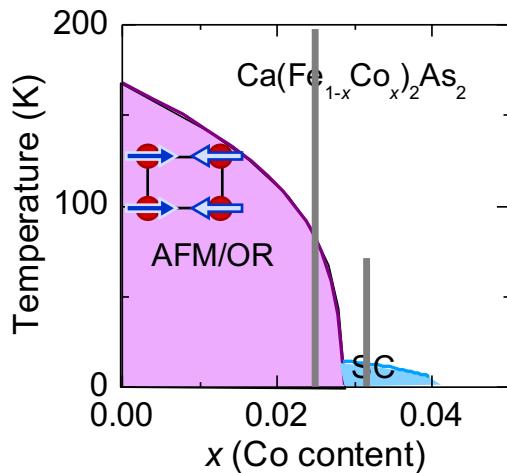


The “best” structural tuning is via the  $c/a$ -ratio

See: Meingast et al., PRL 2012  
Böhmer, PhD thesis

# Uniaxial pressure derivatives from thermal expansivity of $\text{Ca}(\text{Fe},\text{Co})_2\text{As}_2$

Tuning via  
biaxial strain



$\text{Ca}(\text{Fe},\text{Co})_2\text{As}_2$  is incredibly  
sensitive to the  $c/a$  ratio!

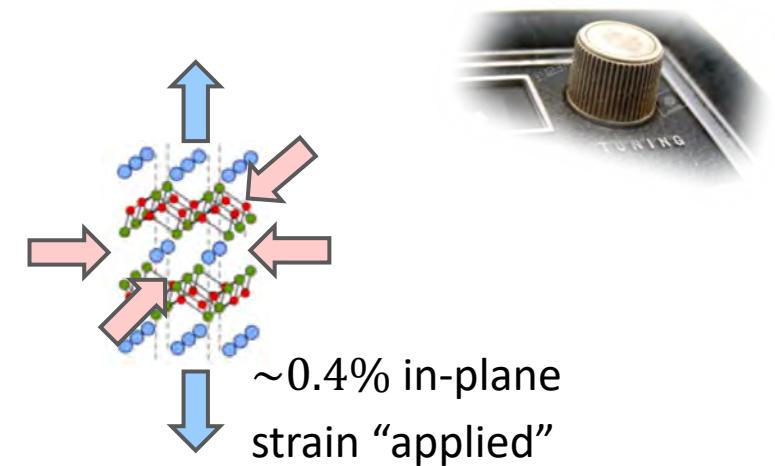
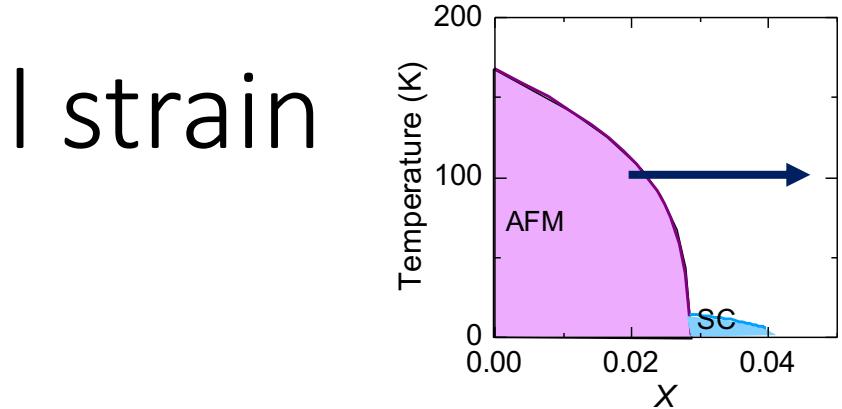
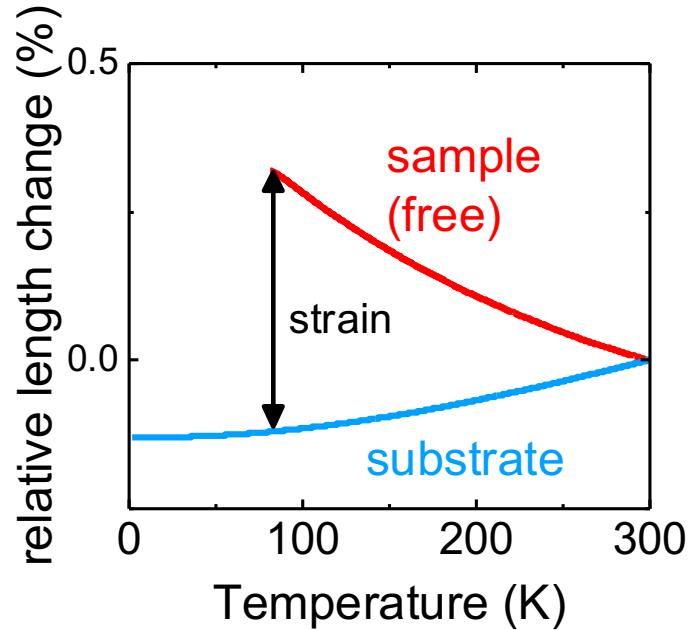
$$\frac{dT_c}{dP_{ab}} \sim 70 \text{ K/GPa}$$

$$\frac{dT_c}{dP_c} \sim -200 \text{ K/GPa}$$

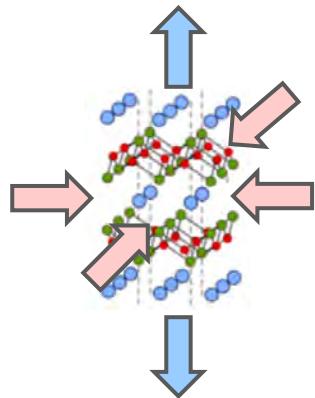
# Tune $\text{Ca}(\text{Fe}_{1-x}\text{Co}_x)_2\text{As}_2$ via biaxial strain



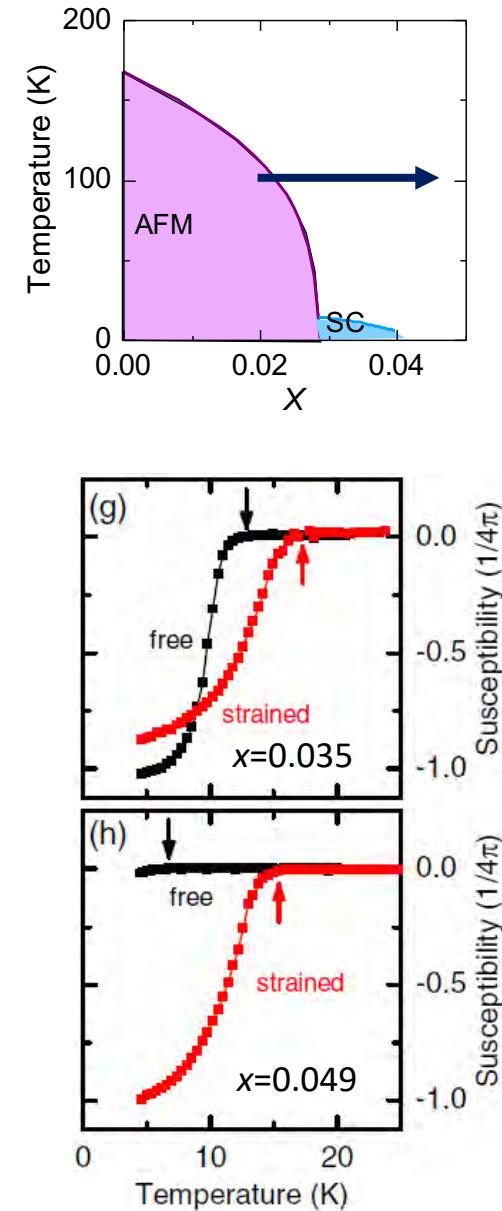
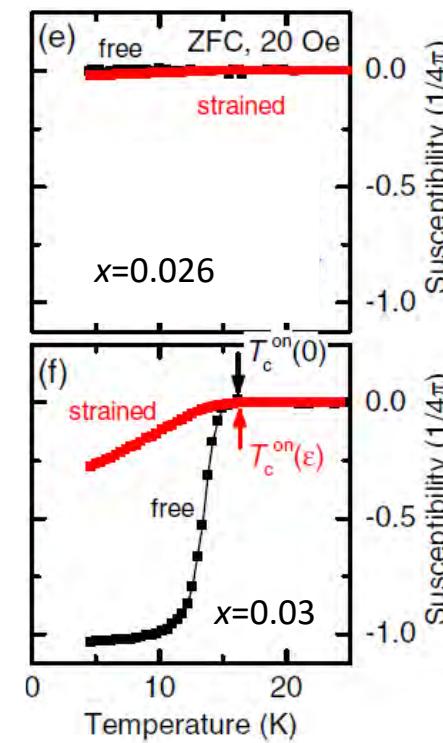
Sample fixed to a glass substrate



# Tune $\text{Ca}(\text{Fe}_{1-x}\text{Co}_x)_2\text{As}_2$ via biaxial strain

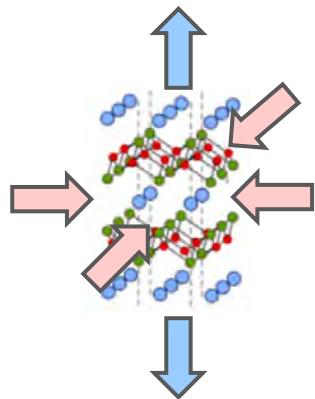


diamagnetic shielding

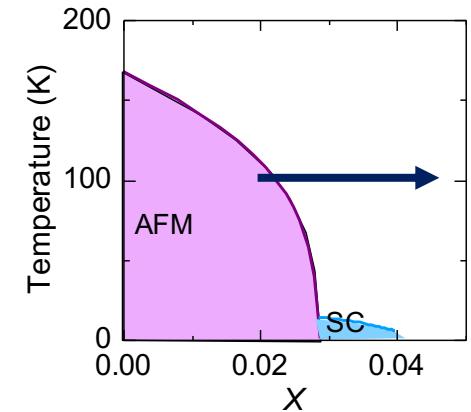
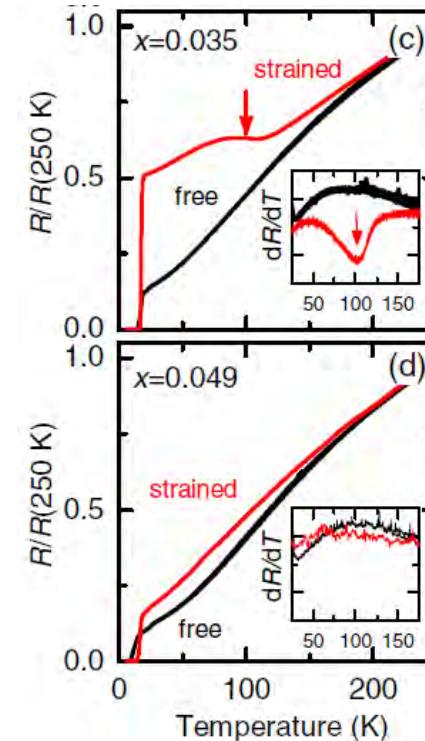
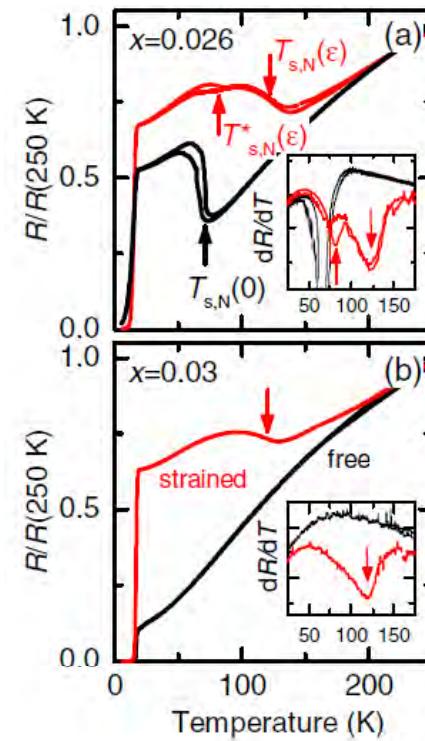


Superconductivity can be suppressed or induced

# Tune $\text{Ca}(\text{Fe}_{1-x}\text{Co}_x)_2\text{As}_2$ via biaxial strain

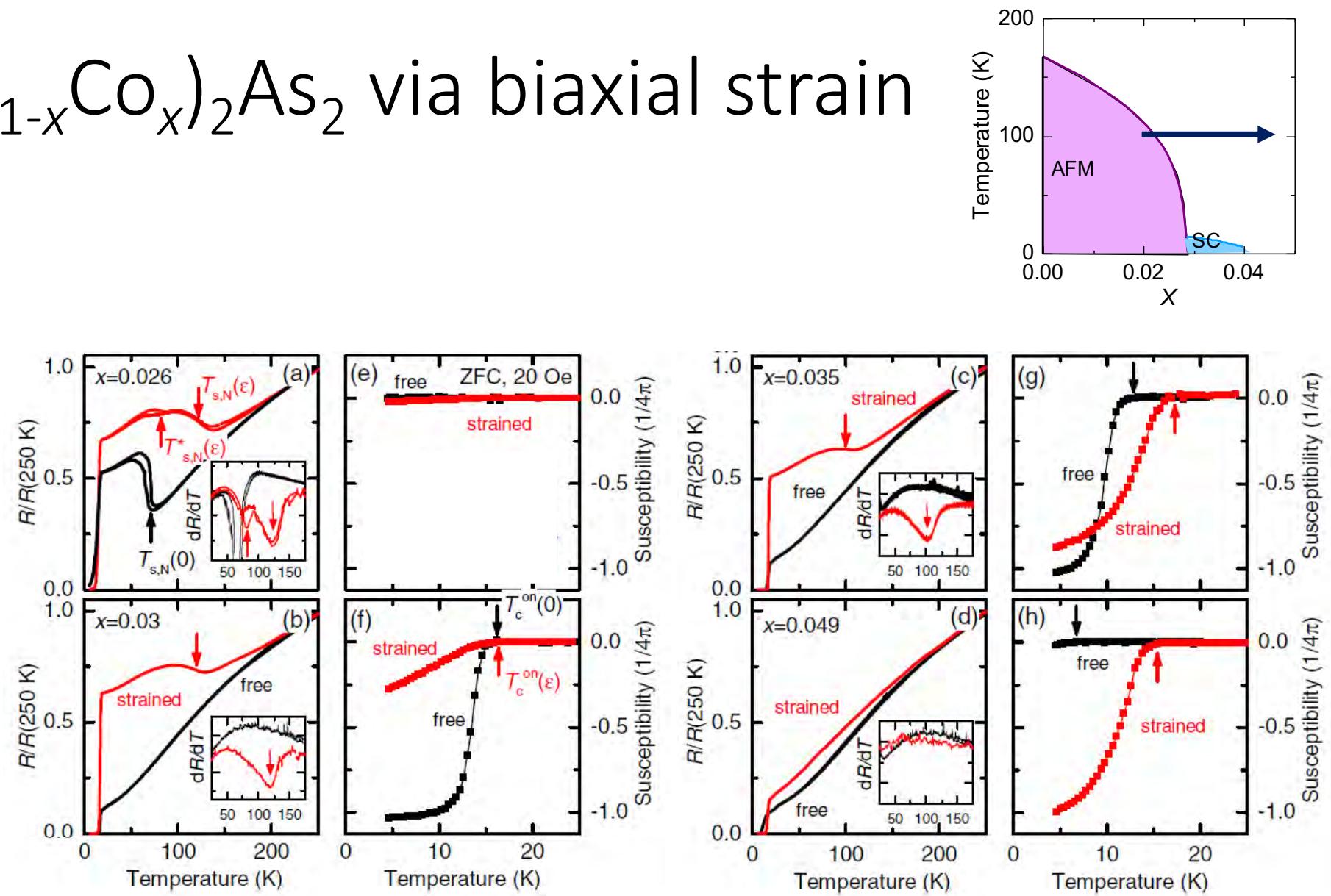
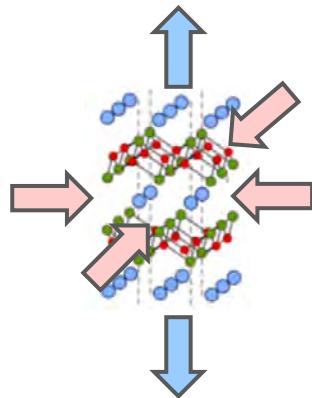


electrical resistance

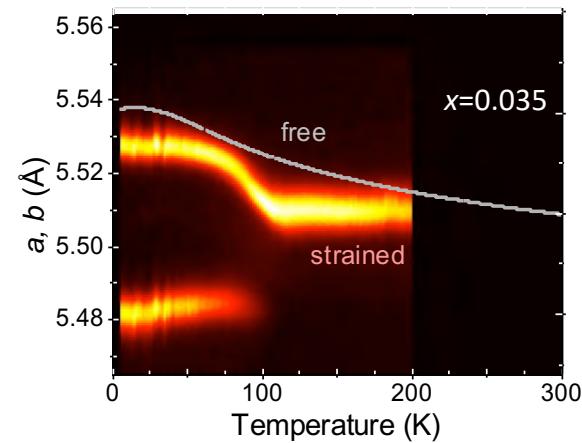
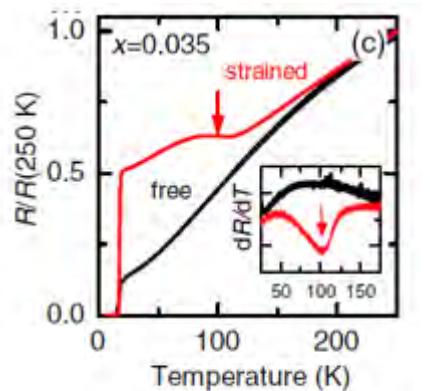
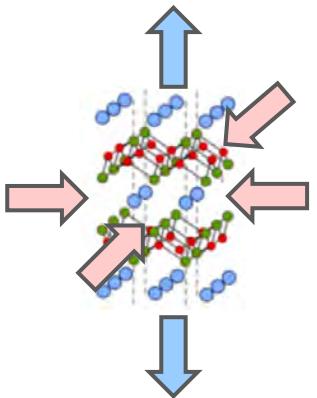


High-temperature transition shifted or induced

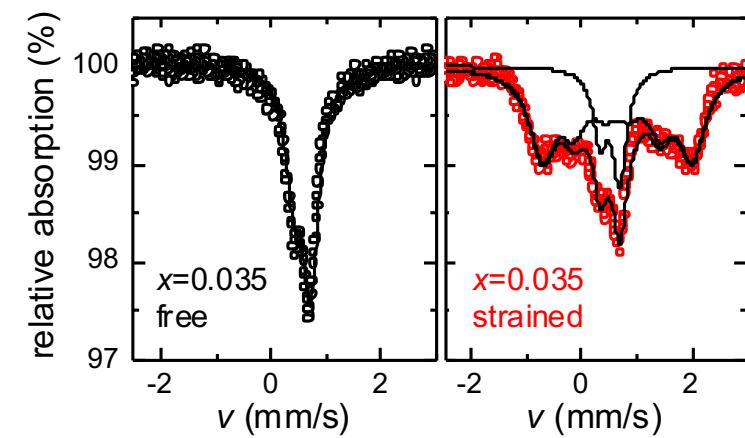
# Tune $\text{Ca}(\text{Fe}_{1-x}\text{Co}_x)_2\text{As}_2$ via biaxial strain



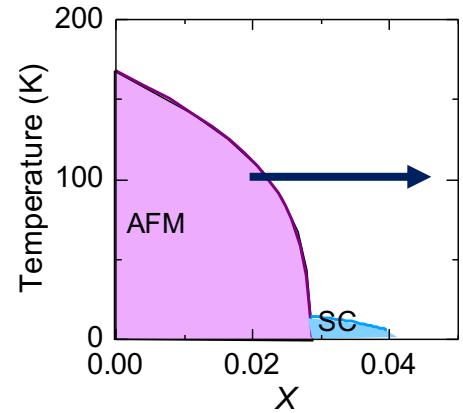
# The strain-induced transition



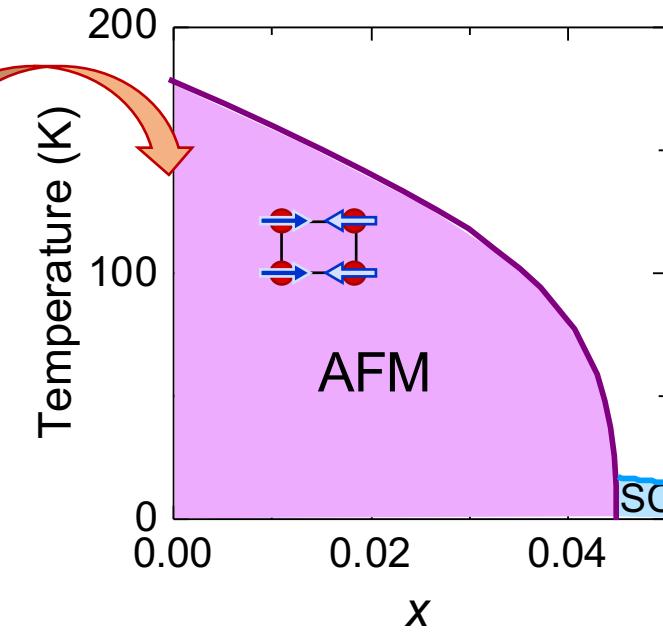
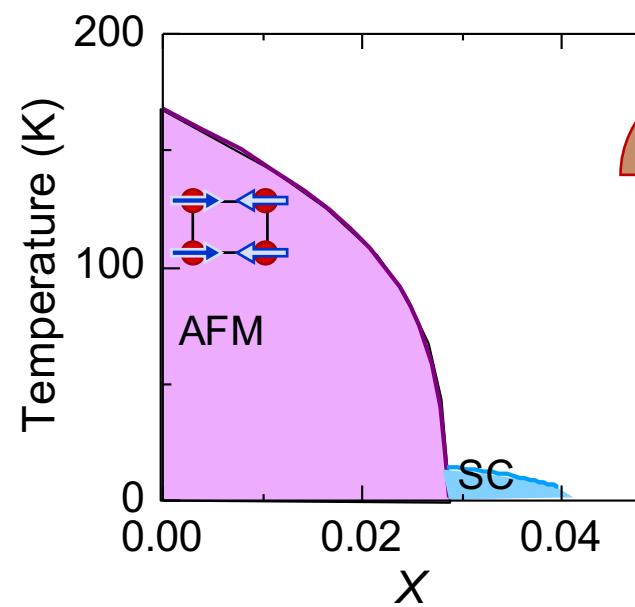
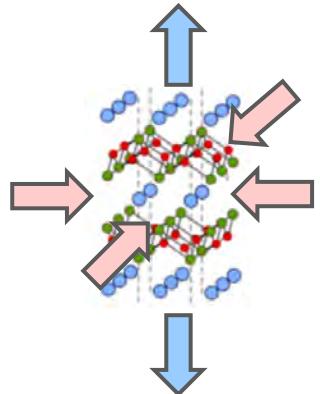
Strain induced  
structural transition



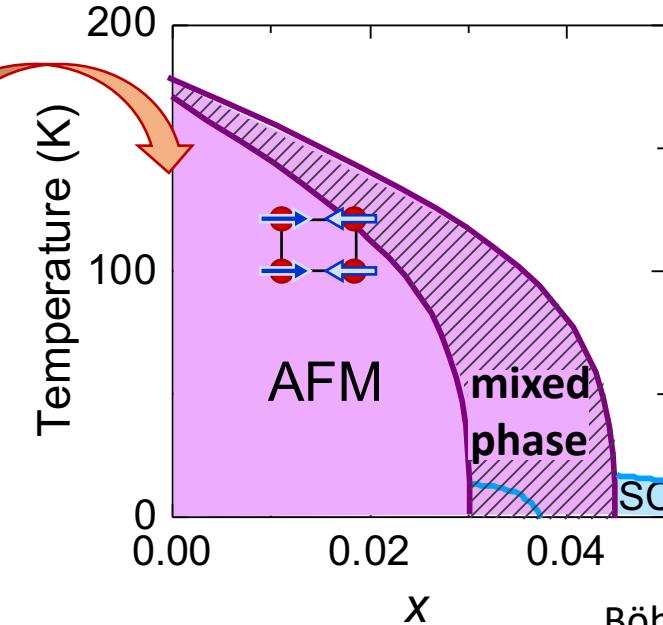
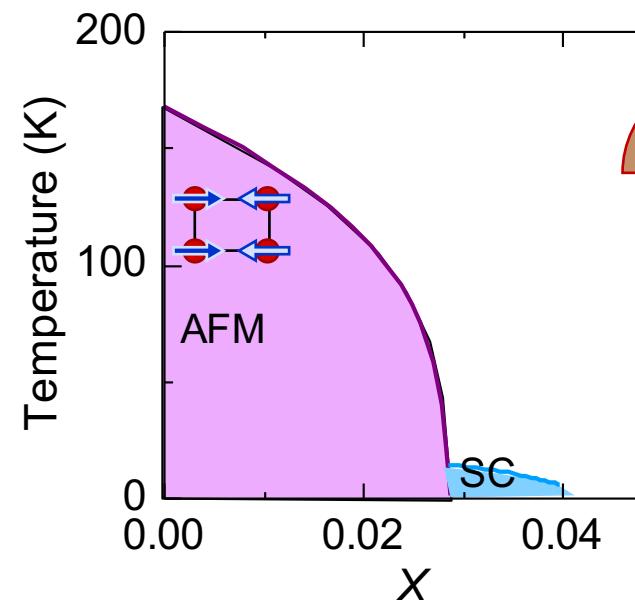
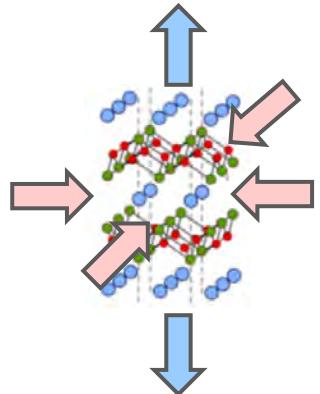
Strain induced  
magnetic order



# Manipulation of the phase diagram



# Manipulation of the phase diagram

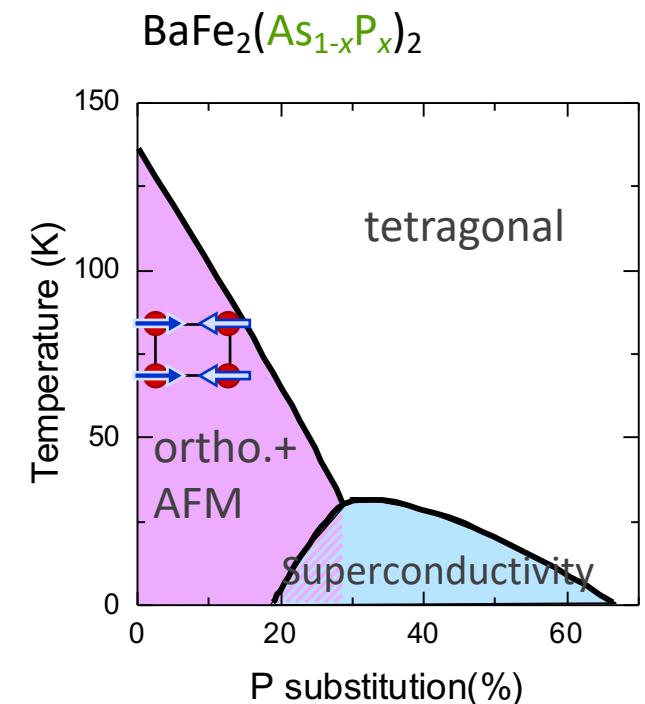
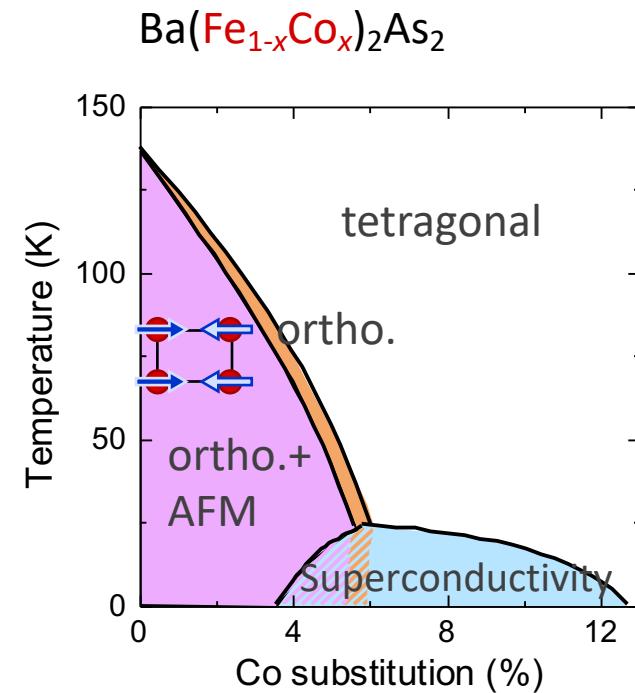
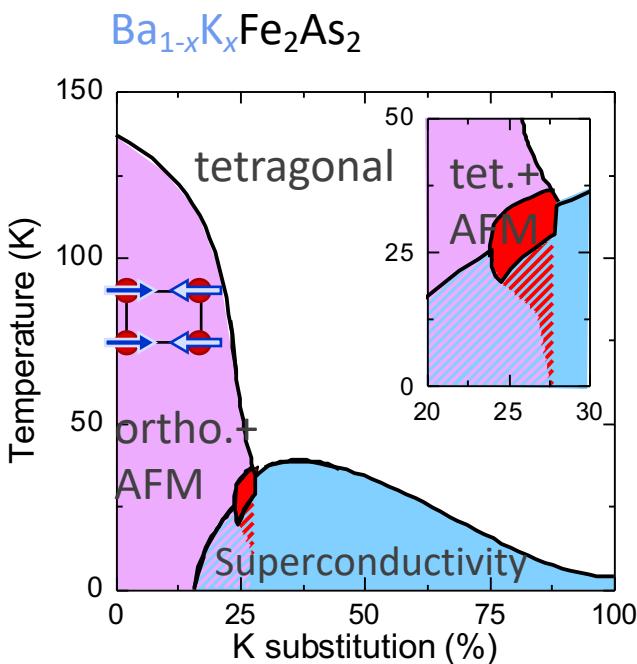
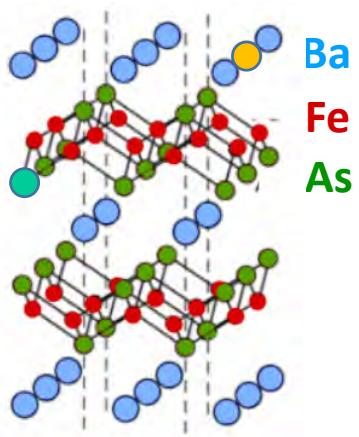


Consequence of fixed *strain* not stress

Böhmer et al., PRL **118**,  
107002 (2017)  
See: Fente et al., PRB **97**,  
014505 (2018)

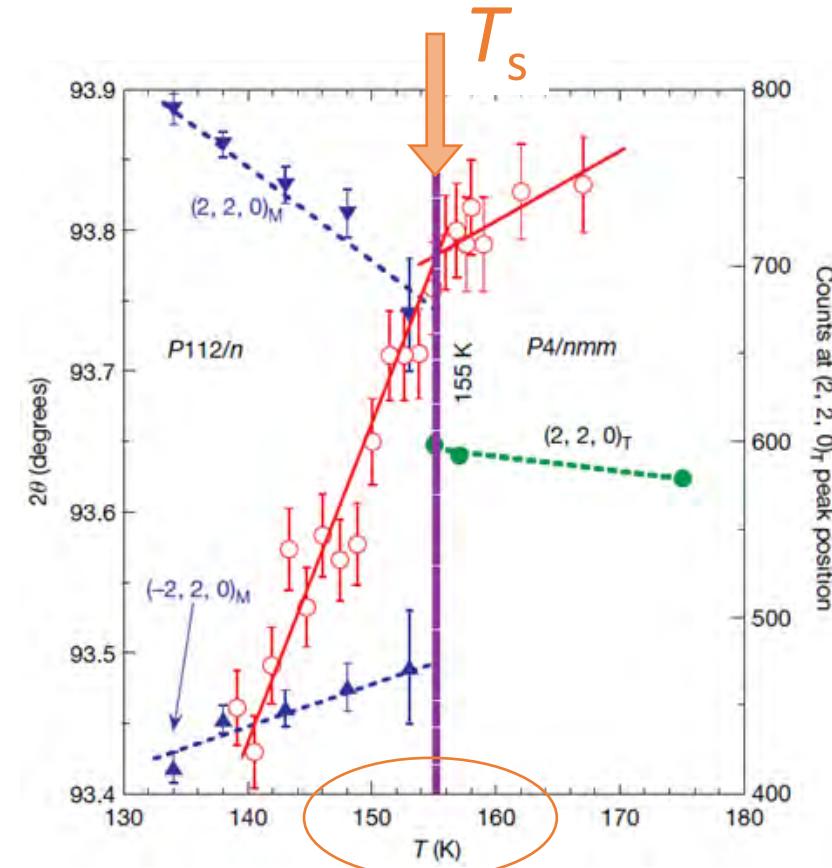
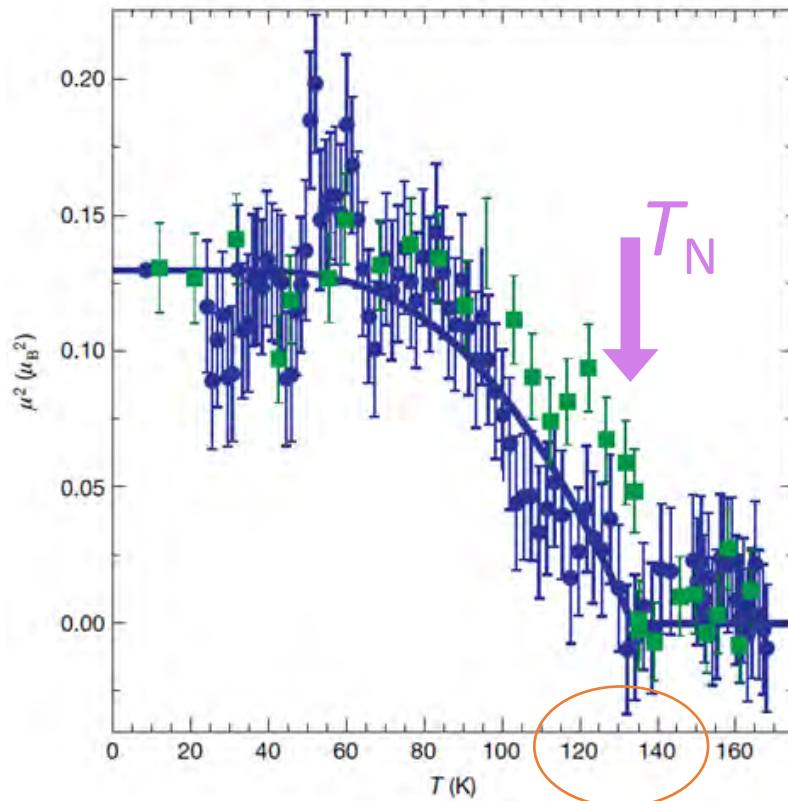
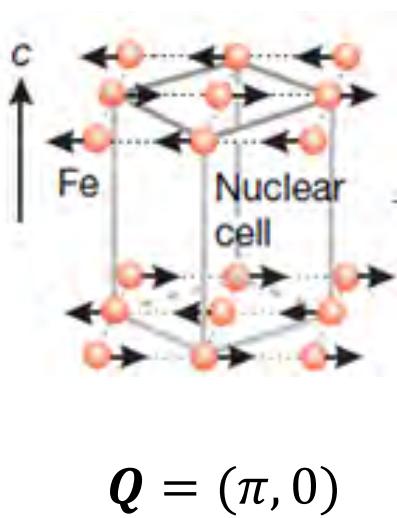
# Phase diagram variants

Tuning via chemical composition



Which magnetic phases can occur in these systems?  
How can they be tuned/stabilized?  
How do they couple to the crystal lattice?

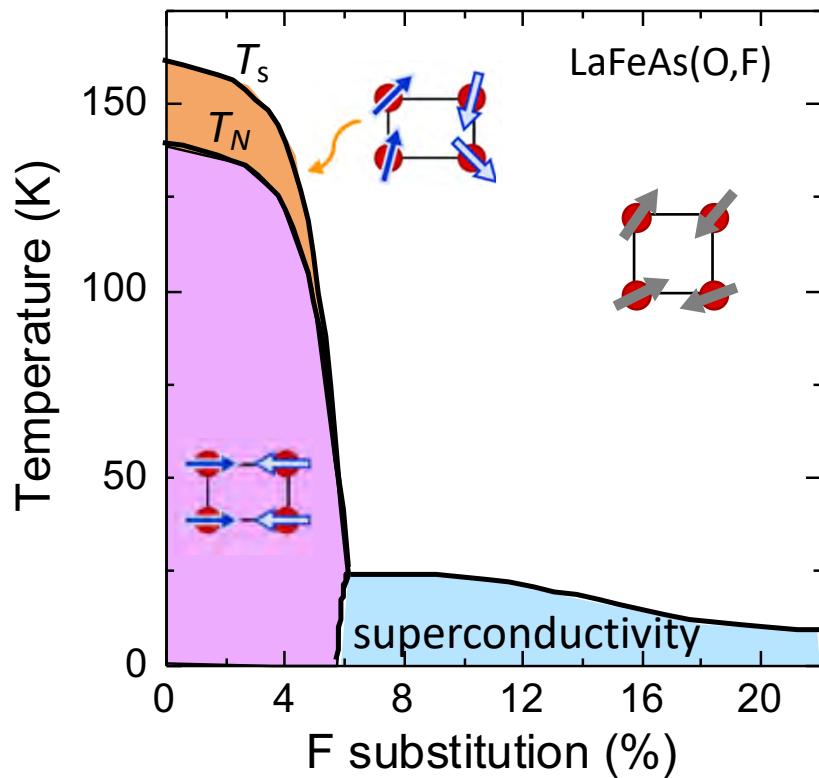
# A surprise in the very beginning...



De la Cruz et al., Nature 453, 899 (2008)

- Magnetic transition is expected to result in a structural transition
- But the structural transition occurs at a higher temperature,  $T_s > T_{AFM}$

# Sequence of magnetic and structural transitions



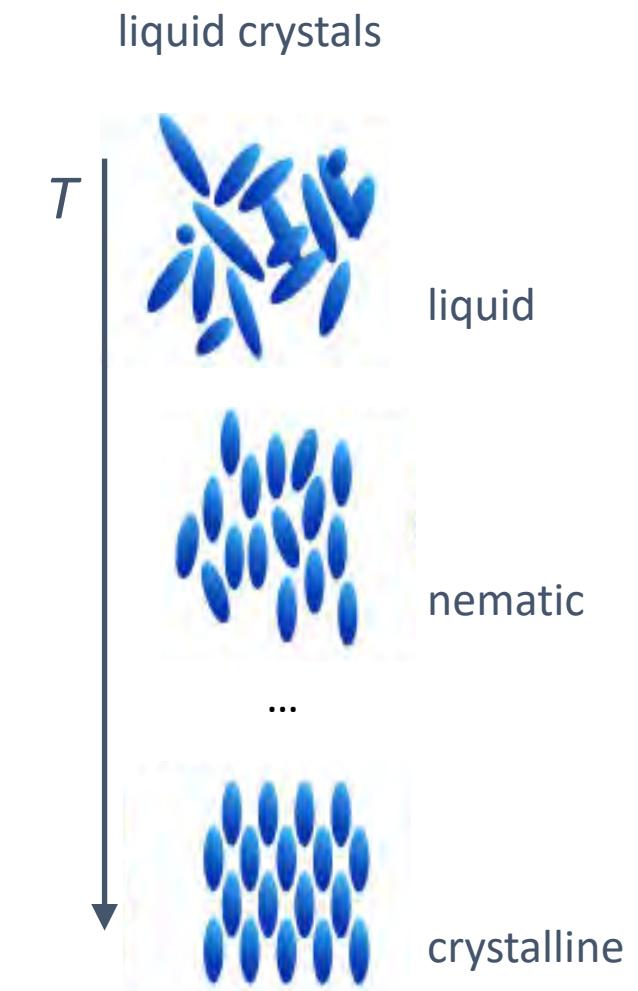
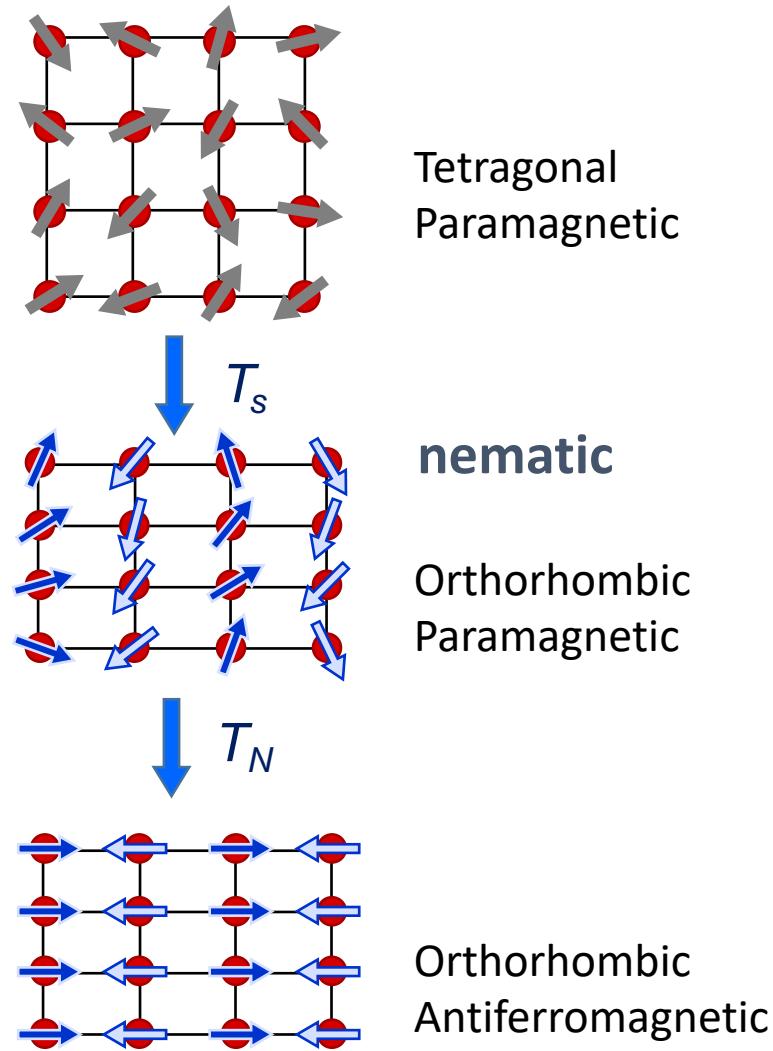
Luetkens et al., Nat. Mat. 8, 305 (2009)

Ni et al., PRB 78, 214515 (2008)

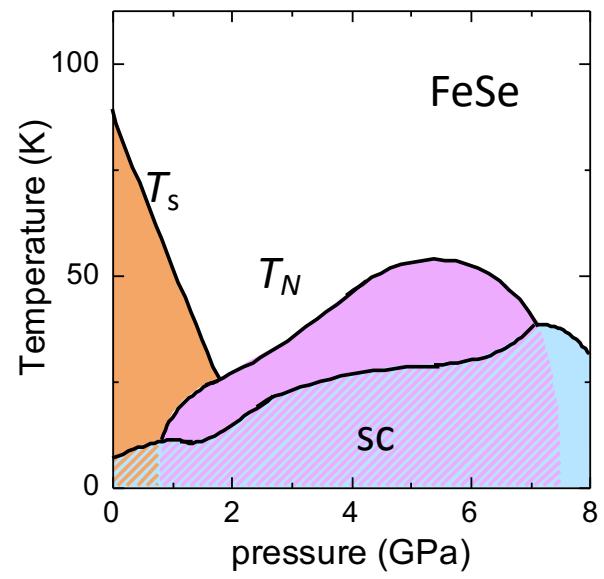
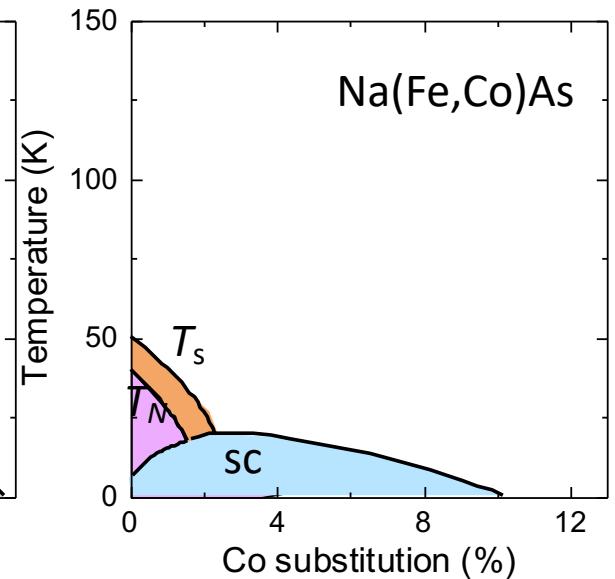
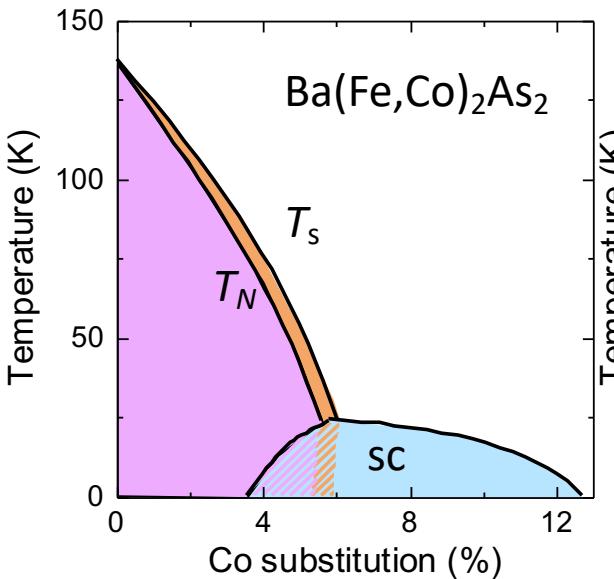
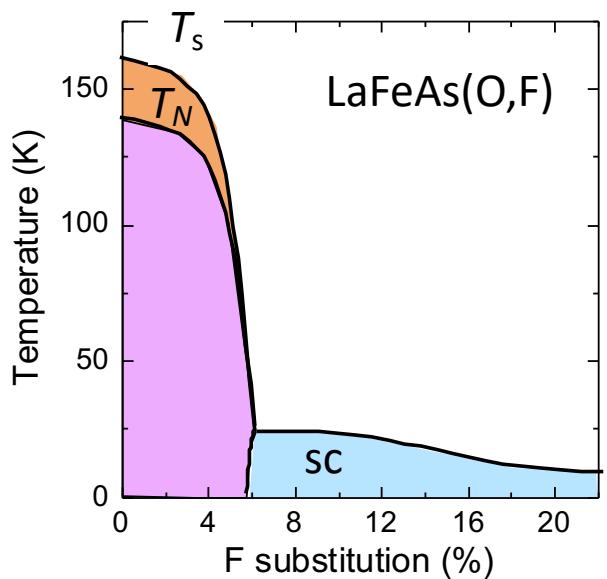
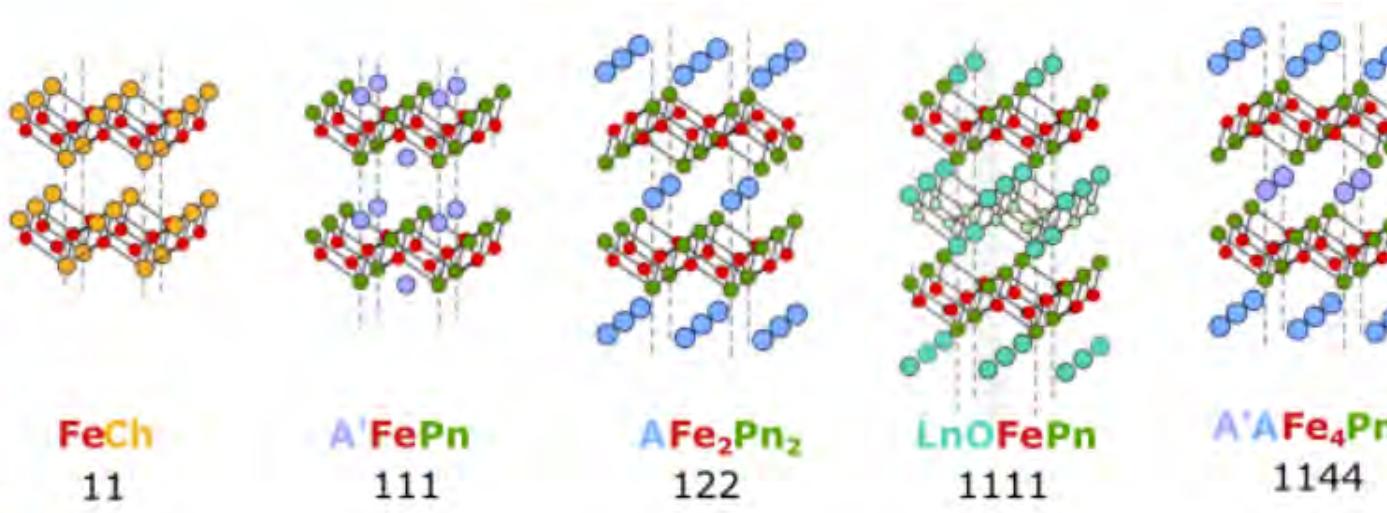
Chu et al., PRB 79, 014506 (2009)

Lester et al., PRB 79, 144523 (2009)

...



# Nematic phase is very common



# Special nature of magnetism in iron-based materials

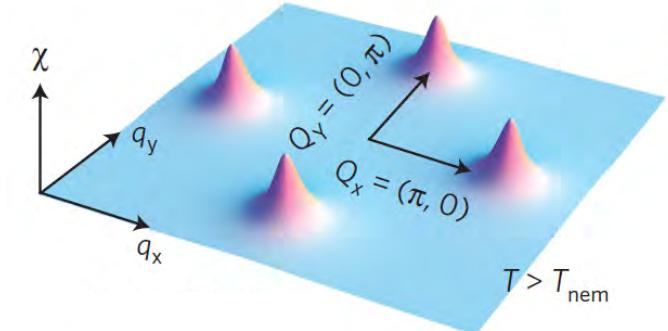
- Antiferromagnetic fluctuations characterized by a pair of wavevectors:

$$\mathbf{Q}_x = (\pi, 0), \mathbf{Q}_y = (0, \pi)$$

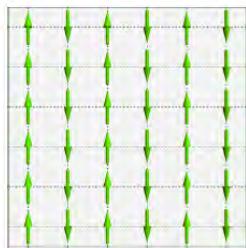
- These are symmetry-equivalent for the tetragonal system

SDW can be described as:

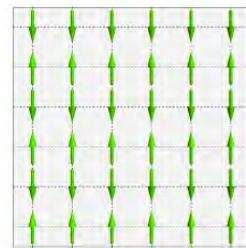
$$\mathbf{M}(\mathbf{r}) = \mathbf{M}_1 \cos(\mathbf{Q}_x \mathbf{r}) + \mathbf{M}_2 \cos(\mathbf{Q}_y \mathbf{r})$$



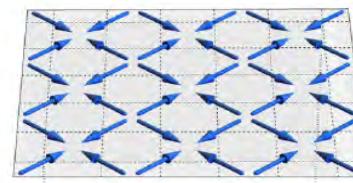
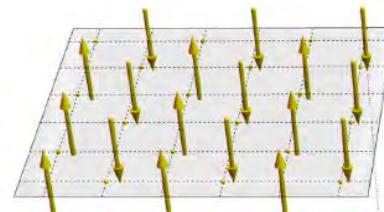
Various types of commensurate magnetic order are possible from these wavevectors



Stripe-type AFM



“C4” magnetic order



Spin-vortex crystal

Lorenzana et al., Phys. Rev. Lett. 101, 186402 (2008)  
Eremin, Chubukov, Phys. Rev. B 81, 024511 (2010)

Fernandes, Kivelson, and Berg, PRB 93, 014511 (2016)  
Fernandes, Orth, Schmalian ARCMP 10, 133 (2019)

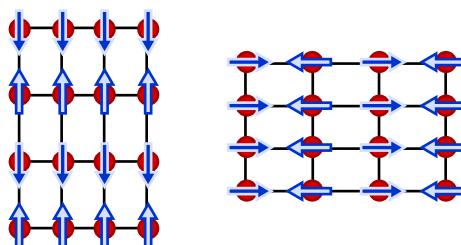
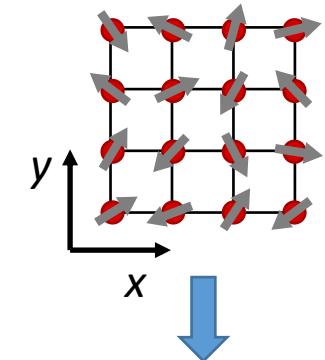
Note: Leave aside materials that feature magnetic fluctuations with other wavevectors such as  $\text{Fe}_{1+y}\text{Te}$ ,  $\text{K}_x\text{Fe}_{2-y}\text{Se}_2$

# Option 1: stripe-type spin-density wave (SSDW)

$$\mathbf{M}(\mathbf{r}) = \mathbf{M}_1 \cos(\mathbf{Q}_X \cdot \mathbf{r}) + \mathbf{M}_2 \cos(\mathbf{Q}_Y \cdot \mathbf{r}), \mathbf{Q}_X = (\pi, 0), \mathbf{Q}_Y = (0, \pi)$$

with

$$\mathbf{M}_1 = \mathbf{0} \text{ or } \mathbf{M}_2 = \mathbf{0}$$



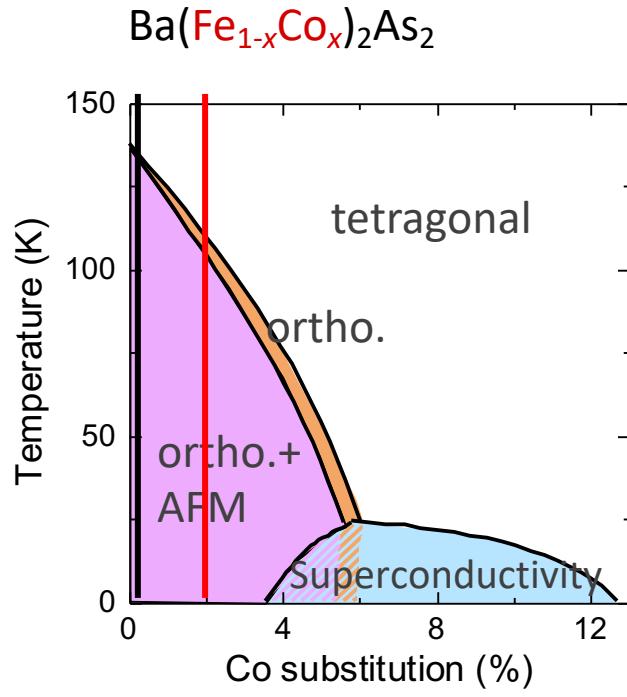
$$\mathbf{M}_1 = 0$$

$$\mathbf{M}_2 = 0$$

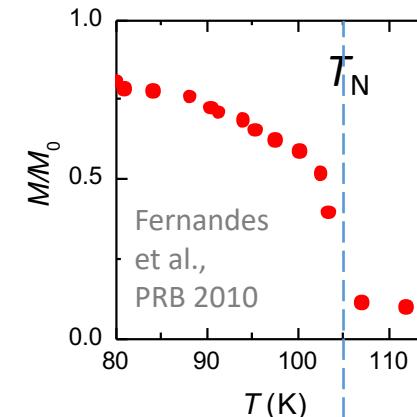
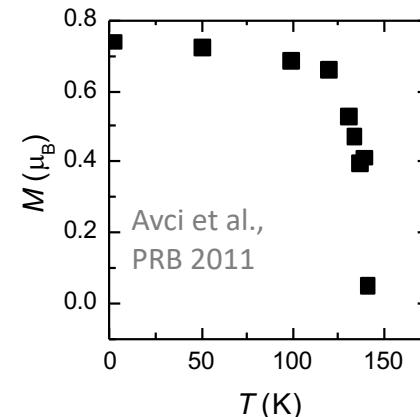
Intertwined order:

- New order Ising-type parameter (*nematic*)  
$$\varphi = \mathbf{M}_1^2 - \mathbf{M}_2^2$$
- Reduced rotational symmetry couples to lattice distortion: tetragonal-to-orthorhombic
- $\varphi$  “gets its own life” from magnetic fluctuations and induces nematic transition with  $T_{\text{nem}} \geq T_{AFM}$

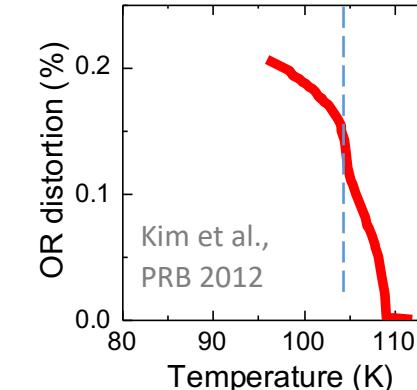
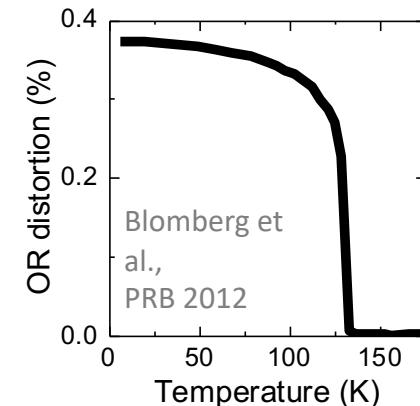
# Coupled but separated nematic order emerges



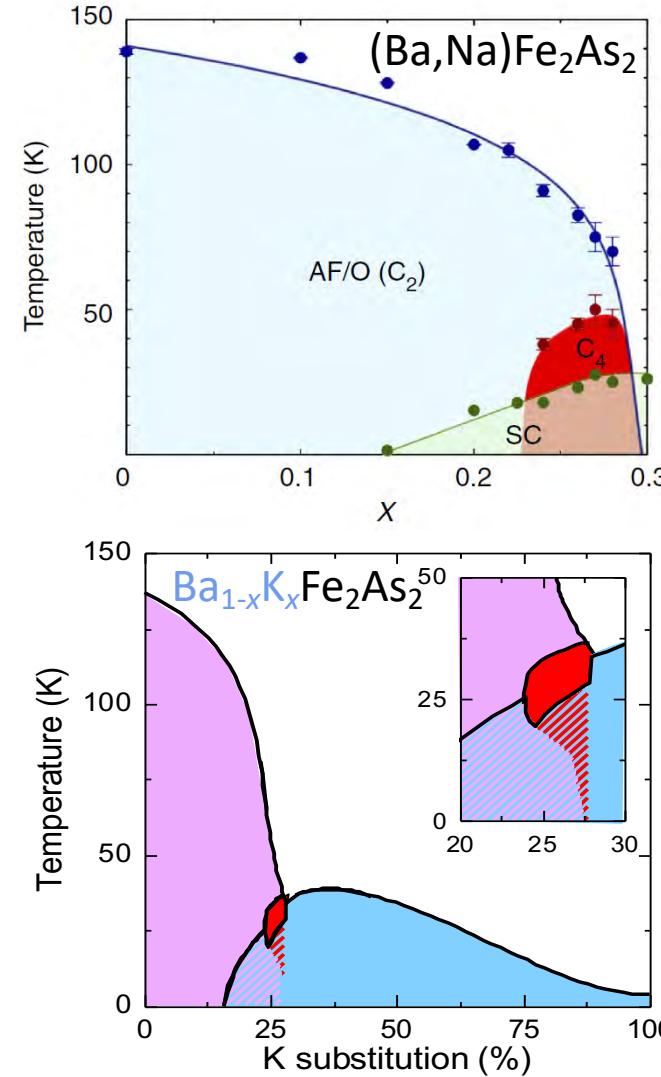
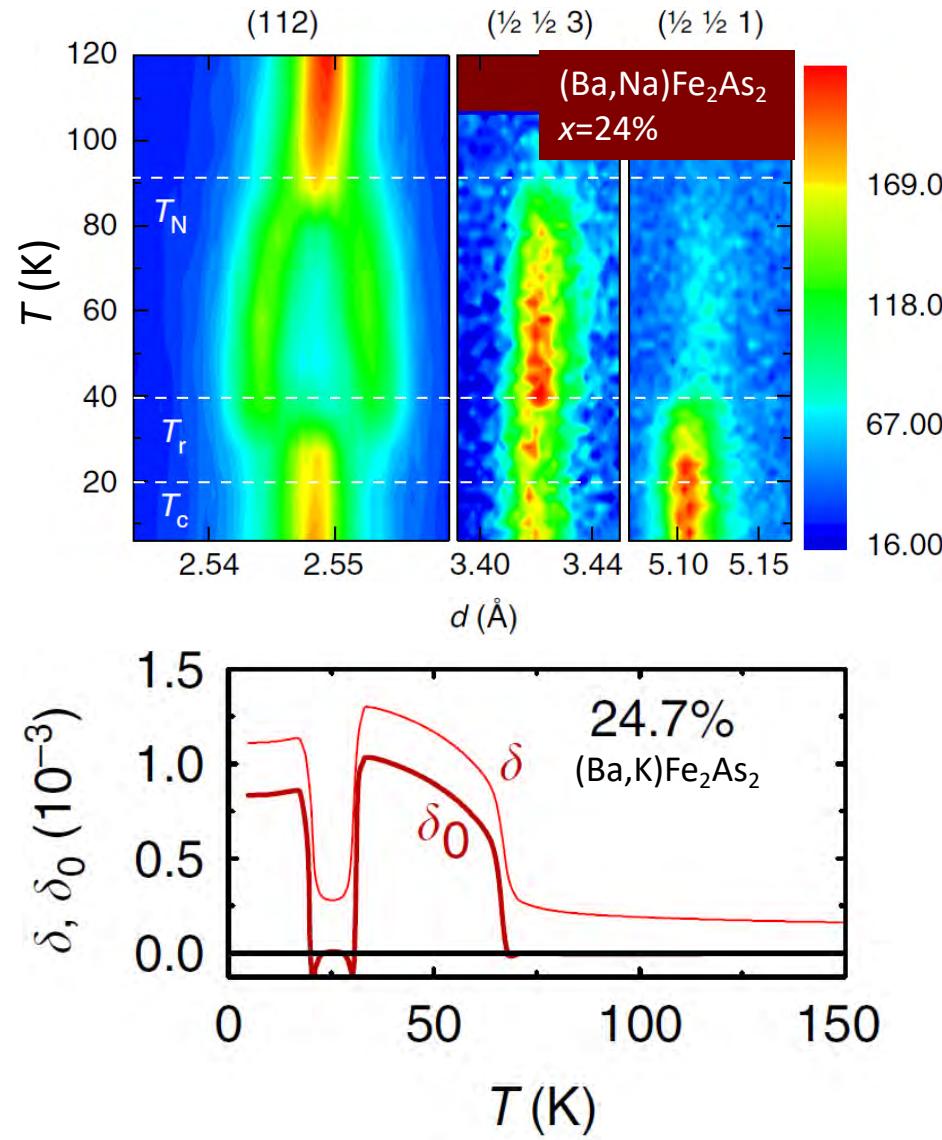
Magnetic order parameter



Nematic order parameter



# A surprisingly tetragonal magnetic phase



Avci et al., Nat. Comm. **5**,  
3845 (2014)

Böhmer et al., Nature  
Comm. **6**, 7911, (2015)

Discovery of a “new phase”:  
Hassinger et al., PRB **86**,  
1405052 (2012)

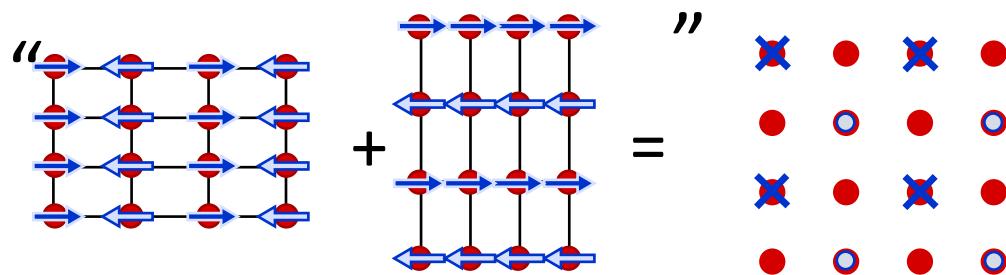
# Option 2: spin-charge density wave, “C4” phase

$$\mathbf{M}(\mathbf{r}) = \mathbf{M}_1 \cos(\mathbf{Q}_X \cdot \mathbf{r}) + \mathbf{M}_2 \cos(\mathbf{Q}_Y \cdot \mathbf{r}), \mathbf{Q}_X = (\pi, 0), \mathbf{Q}_Y = (0, \pi)$$

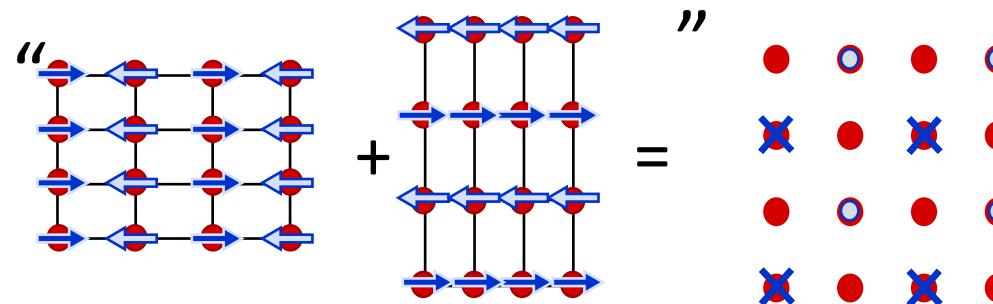
with

$$\mathbf{M}_1 = +\mathbf{M}_2 \text{ or } \mathbf{M}_1 = -\mathbf{M}_2$$

Choice:  $\mathbf{M}_1 = +\mathbf{M}_2$



Choice:  $\mathbf{M}_1 = -\mathbf{M}_2$



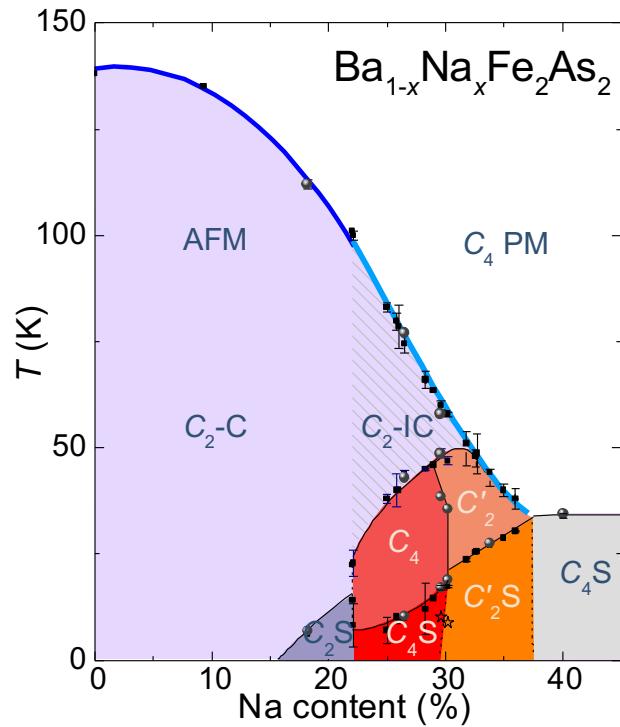
Intertwined order

- Reduced translational symmetry
- Scalar order parameter:  
$$\varphi = \mathbf{M}_1 \cdot \mathbf{M}_2$$
- Couples to charge density! i.e., SCDW

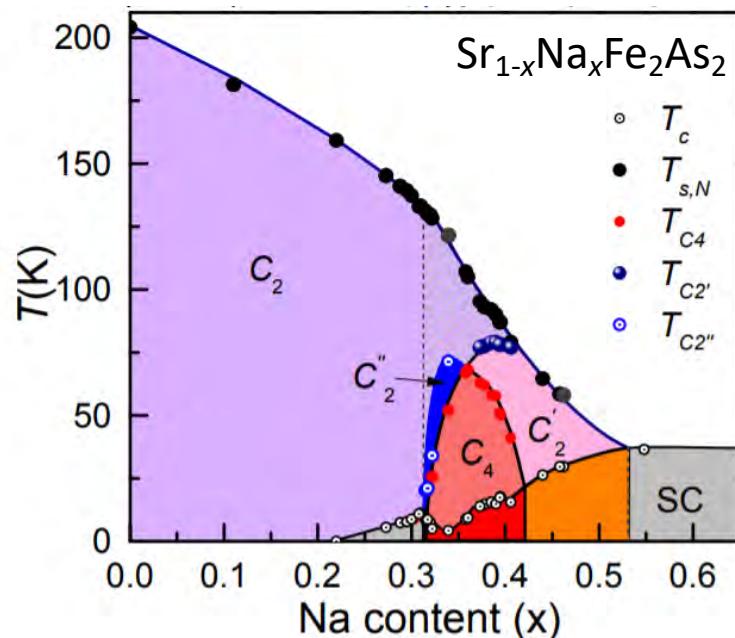
See: Yi et al., Phys. Rev. Lett. **121**, 127001 (2018)

“Spectral Evidence for Emergent Order in  $\text{Ba}_{1-x}\text{Na}_x\text{Fe}_2\text{As}_2$ ”

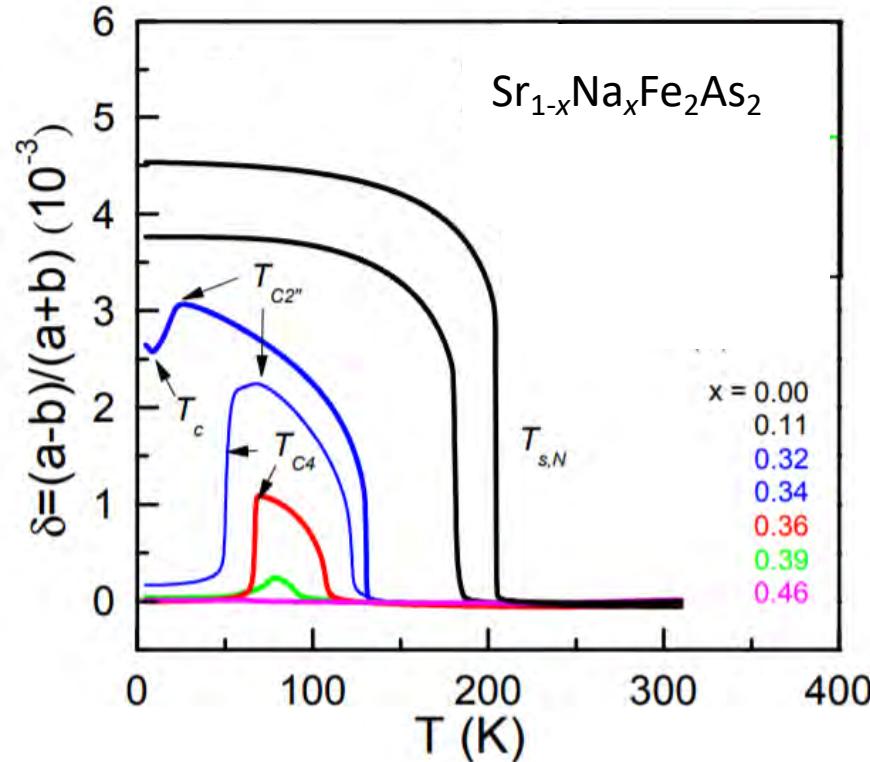
# It can be much more complex



$C_2'$  and  $C_2''$  phases: precise nature is unclear  
Textured phases?

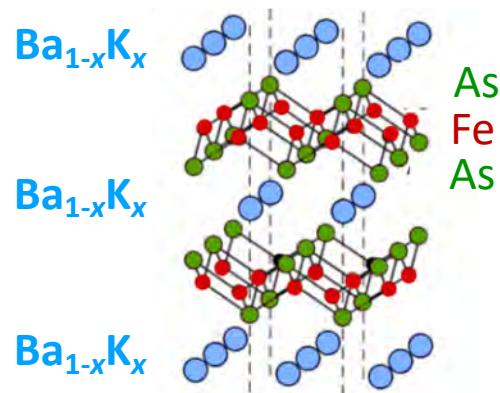


Orthorhombic distortion is a sensitive measure!

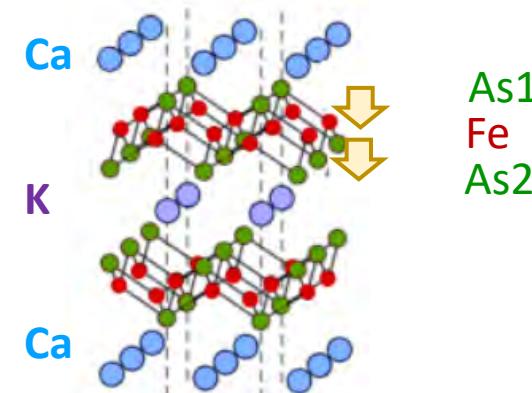


L. Wang et al., PRB 93, 014514 (2016)  
L. Wang et al., JPSJ 88, 104710 (2019)  
See: Morten Christensen, Brian Andersen, Rafael Fernandes et al.

# $\text{CaK}(\text{Fe},\text{Ni})_4\text{As}_4$ : a 1144 material



VS.



I4/mmm

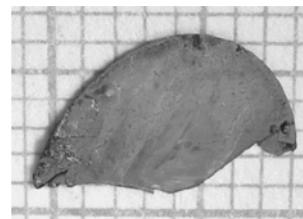


P4/mmm

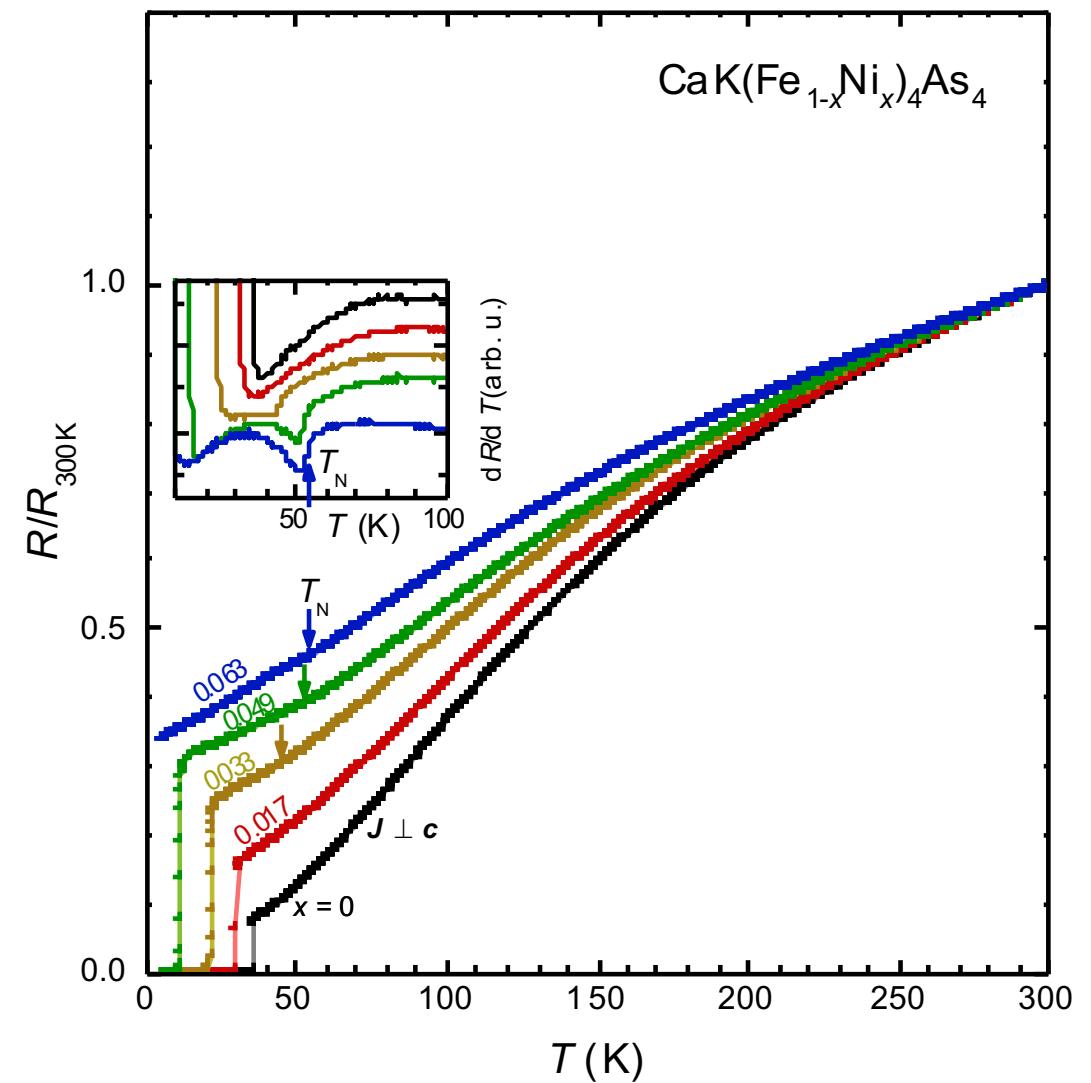
Iyo et al., *J. Am. Chem. Soc.* 138, 3410 (2016)

Meier et al., *Phys. Rev. B* 94, 064501 (2016)

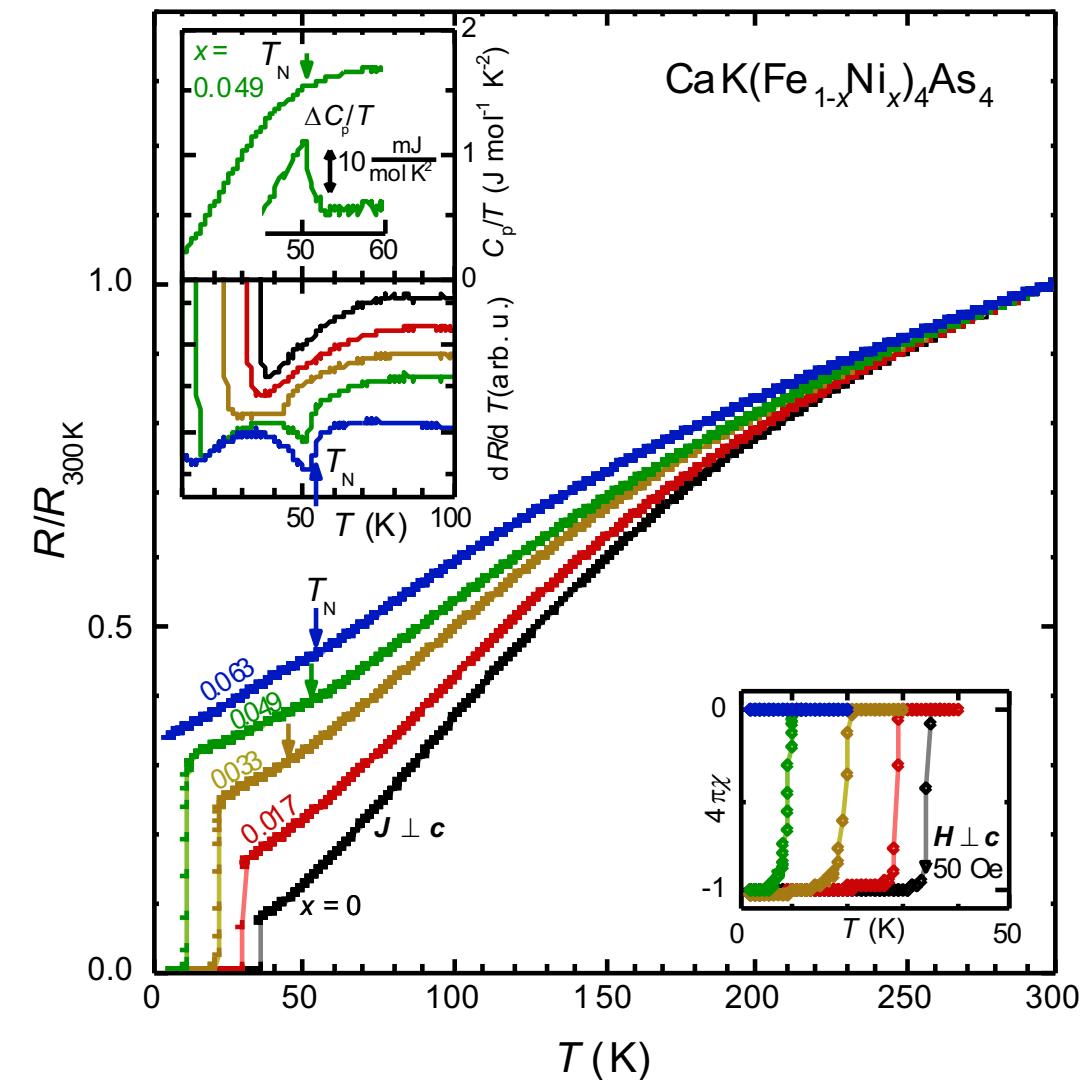
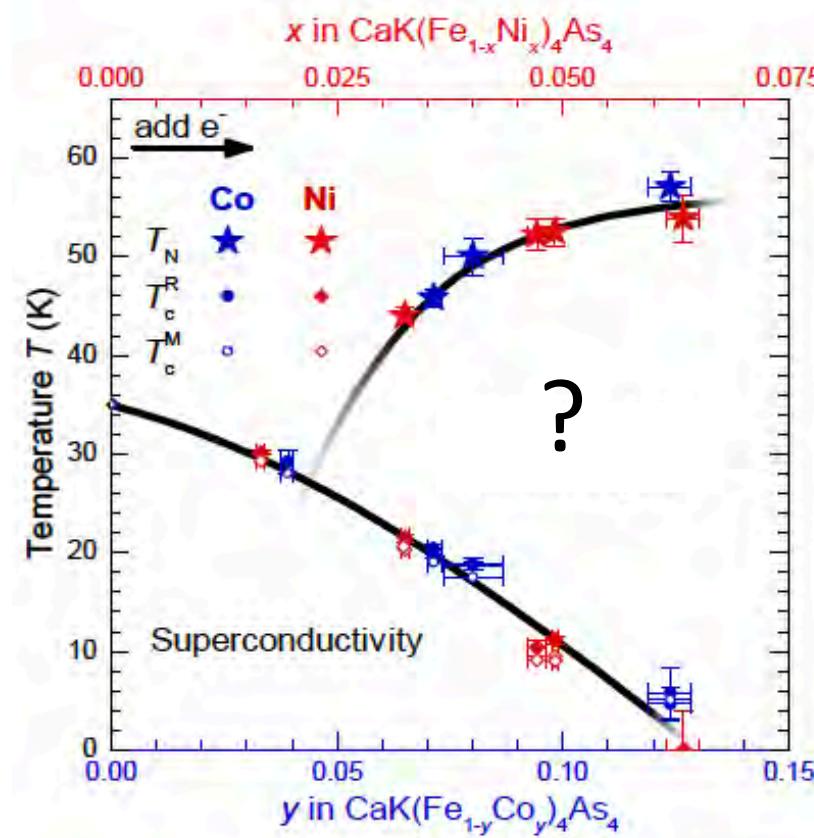
Meier et al., *Phys. Rev. Materials* 1, 013401 (2017)



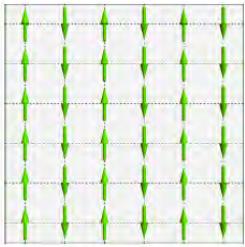
# $\text{CaK(Fe,Ni)}_4\text{As}_4$ : second-order phase transition



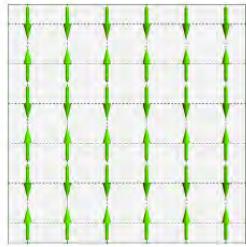
# $\text{CaK}(\text{Fe},\text{Ni})_4\text{As}_4$ : second-order phase transition



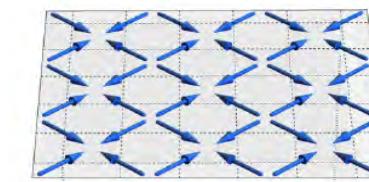
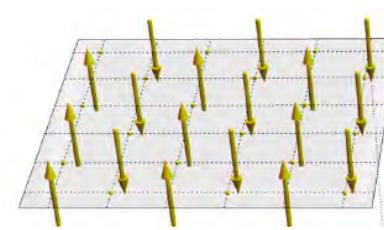
# Various types of commensurate magnetic order are possible



Stripe-type AFM



“C4” magnetic order



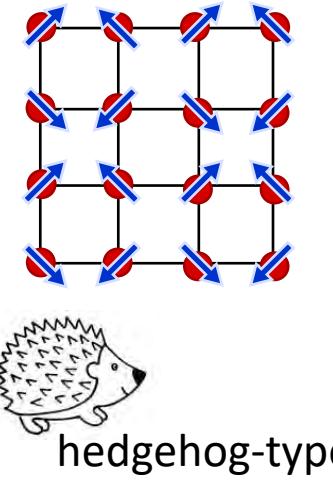
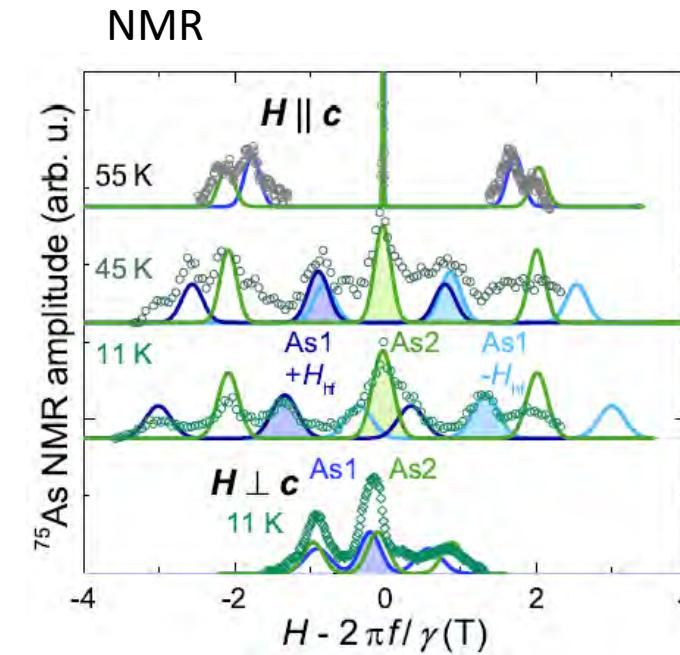
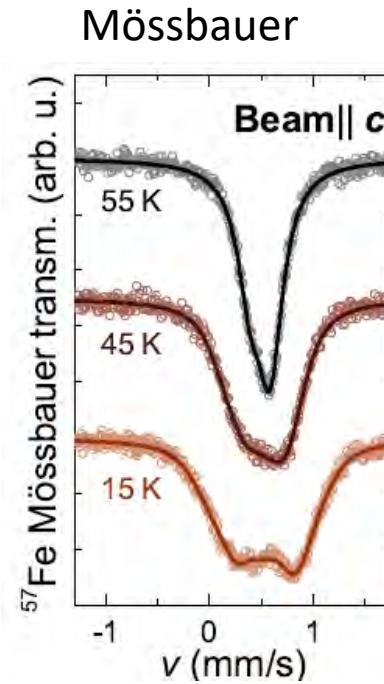
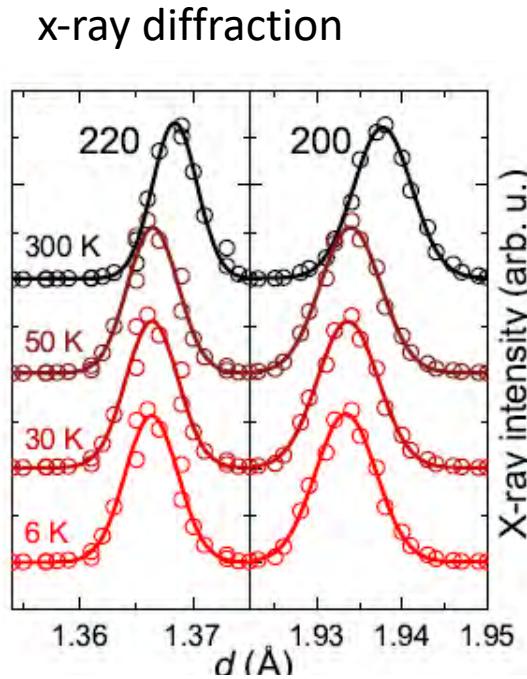
Spin-vortex crystal

Lorenzana et al., Phys. Rev. Lett. 101, 186402 (2008)  
Eremin, Chubukov, Phys. Rev. B 81, 024511 (2010)

Fernandes, Kivelson, and Berg, PRB **93**, 014511 (2016)  
Fernandes, Orth, Schmalian ARCMP 10, 133 (2019)

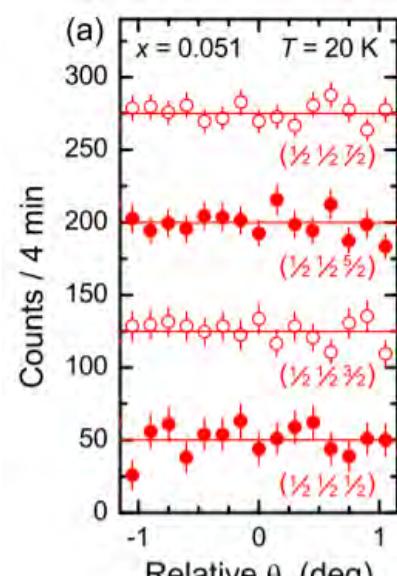
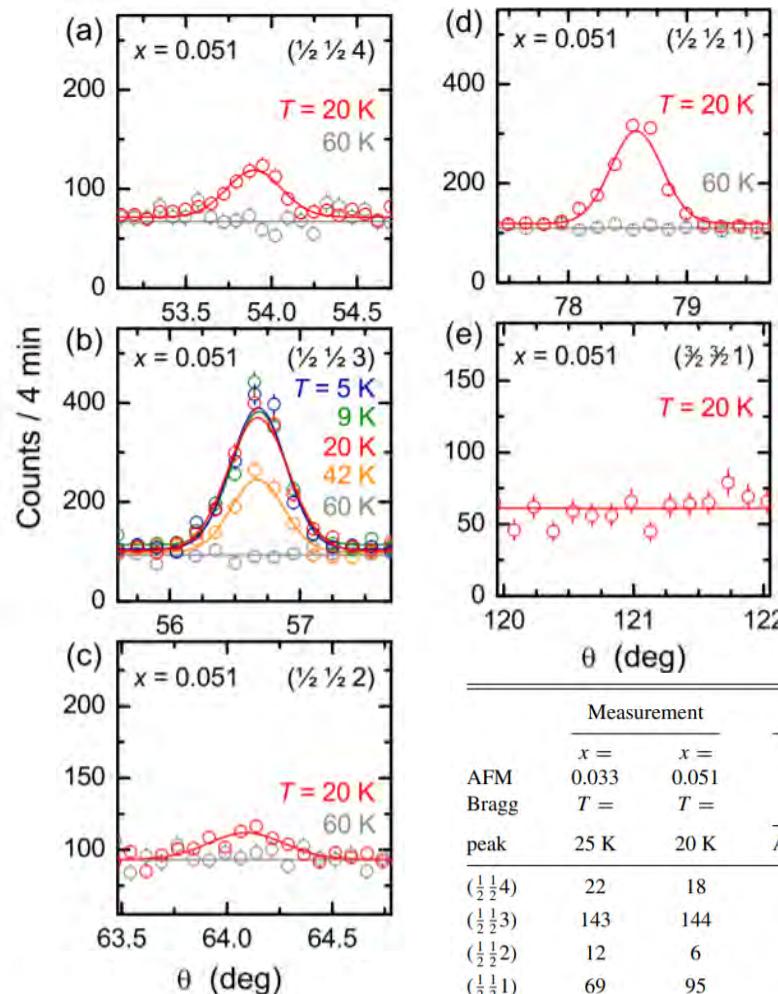
# Spin-vortex crystal magnetic order identified in $\text{CaK}(\text{Fe},\text{Ni})_4\text{As}_4$

- Tetragonal and magnetically ordered -> not nematic
- Mössbauer spectra indicate a magnetic moment on all iron sites -> not the SCDW
- NMR spectra provide strong evidence for “option 3”: spin-vortex crystal structure

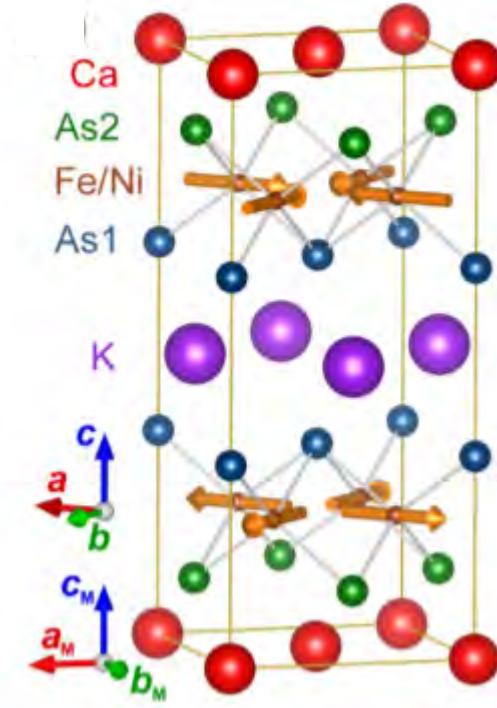


hedgehog-type

# Confirmation from neutron diffraction



Measurement			Calculation										
	$x =$	$x =$	Hedgehog SVC in $(ab)$ plane: $\mu_i \parallel \tau_i$		Loops SVC in $(ab)$ plane: $\mu_i \perp \tau_i$		SCDW $\mu_i \parallel c$						
	AFM	Bragg	$T =$	$T =$	peak	25 K	20 K	AFM along $c$	FM along $c$	AFM along $c$	FM along $c$	AFM along $c$	FM along $c$
( $\frac{1}{2} \frac{1}{2} 4$ )	22	18				19	80	26	107	13	54		
( $\frac{1}{2} \frac{1}{2} 3$ )	143	144				130	17	207	26	155	20		
( $\frac{1}{2} \frac{1}{2} 2$ )	12	6				9	164	21	384	24	441		
( $\frac{1}{2} \frac{1}{2} 1$ )	69	95				99	1	634	8	1071	14		
( $\frac{3}{2} \frac{3}{2} 1$ )	<2	<2				1	0.1	32	0.4	63	1		



A. Kreyssig et al., PRB 97,  
224521 (2018)

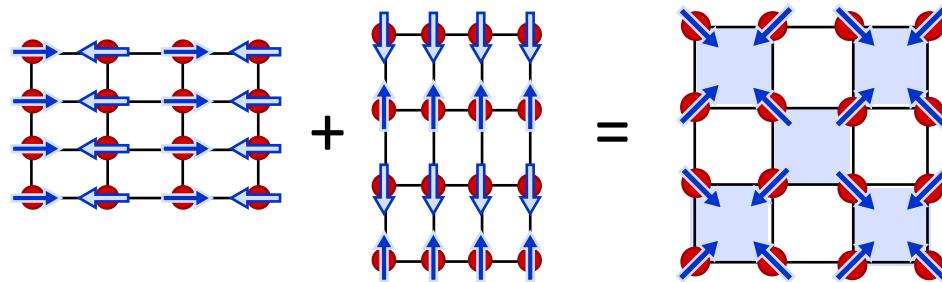
# Option 3: The spin-vortex crystal (SVC)

$$\mathbf{M}(\mathbf{r}) = \mathbf{M}_1 \cos(Q_X \mathbf{r}) + \mathbf{M}_2 \cos(Q_Y \mathbf{r}), Q_X = (\pi, 0), Q_Y = (0, \pi)$$

with

$$\mathbf{M}_1 \perp \mathbf{M}_2$$

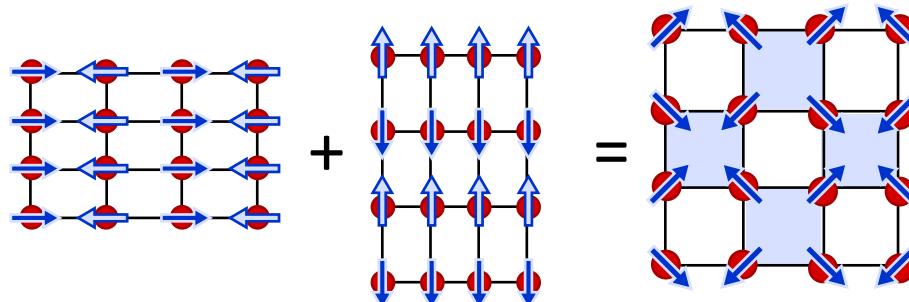
Choice  $\mathbf{M}_2$  “to the right” of  $\mathbf{M}_1$



Intertwined order

- Reduced translational symmetry
- Chiral order parameter:  
 $\varphi = \mathbf{M}_1 \times \mathbf{M}_2$
- “Spin-vorticity-density wave”

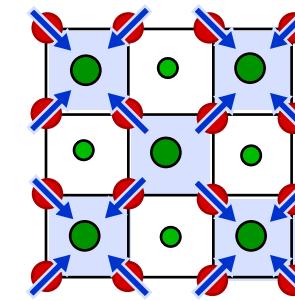
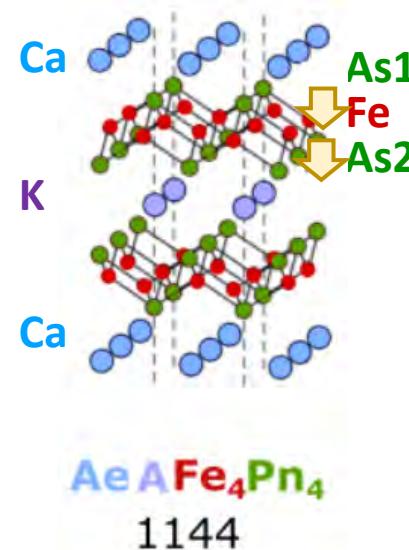
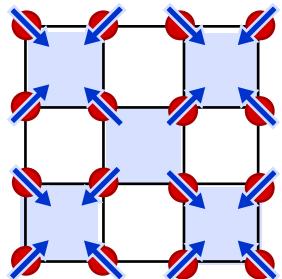
Choice  $\mathbf{M}_2$  “to the left” of  $\mathbf{M}_1$



# Why realized in 1144-type material?

## Intertwined order

- Reduced translational symmetry
- Chiral order parameter:  
$$\varphi = \mathbf{M}_1 \times \mathbf{M}_2$$
- “Spin-vorticity-density wave”



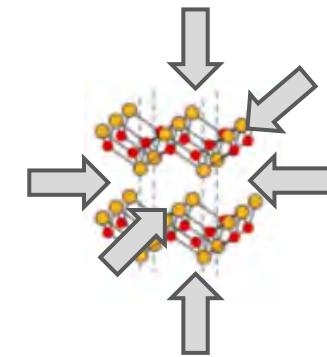
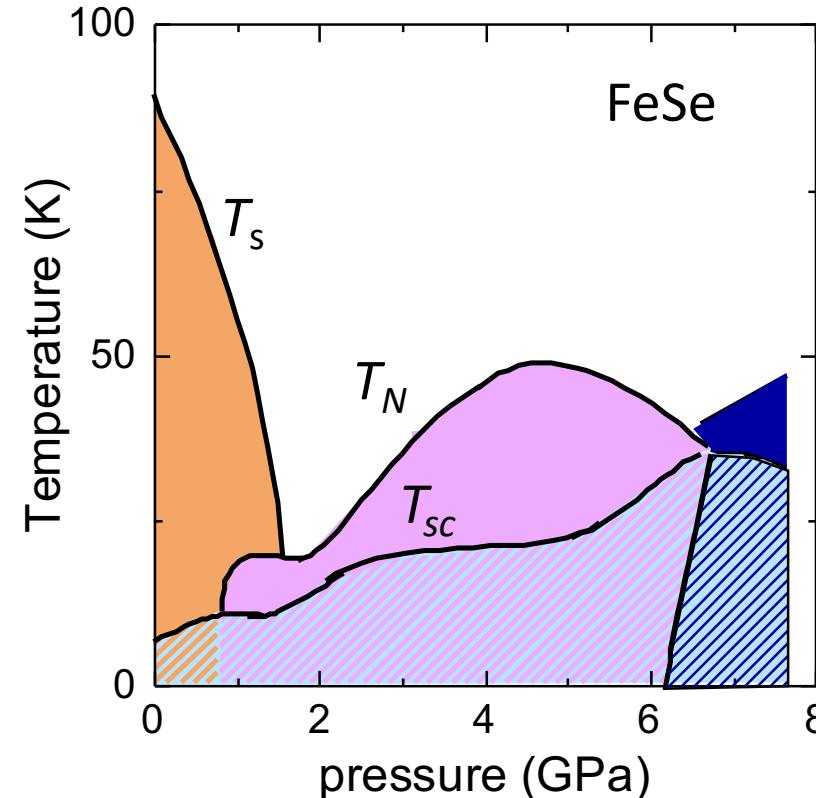
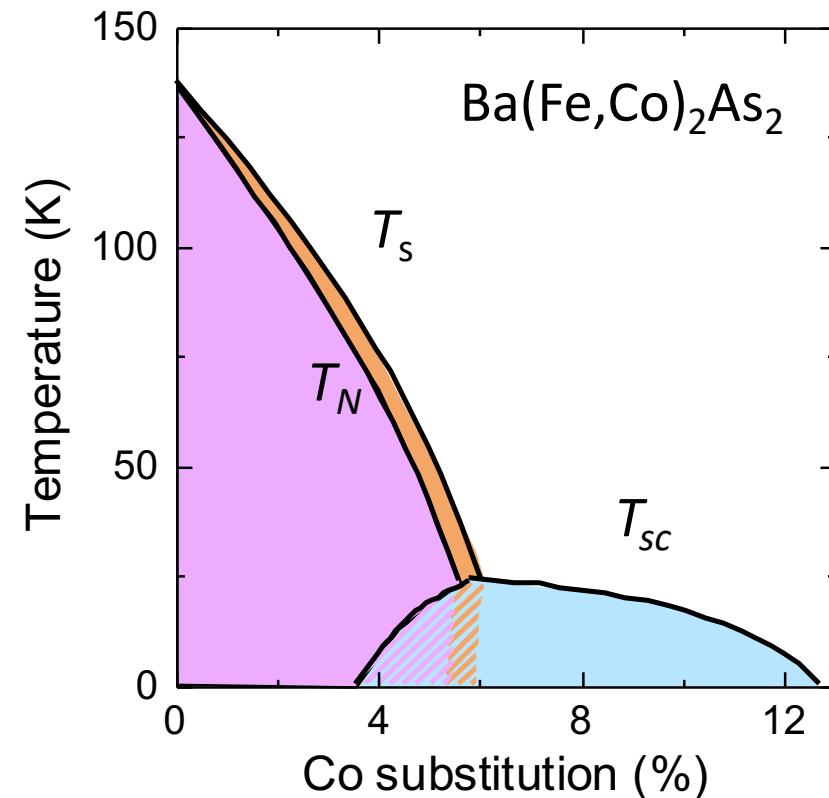
● As1  
● As2



“Tuning” with the internal crystal structure

# FeSe: a mysterious compound

Tuning with hydrostatic pressure



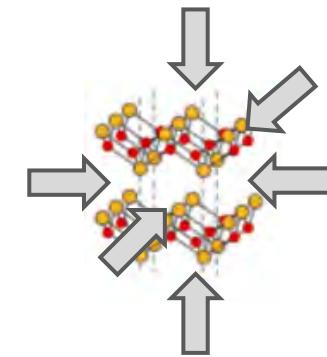
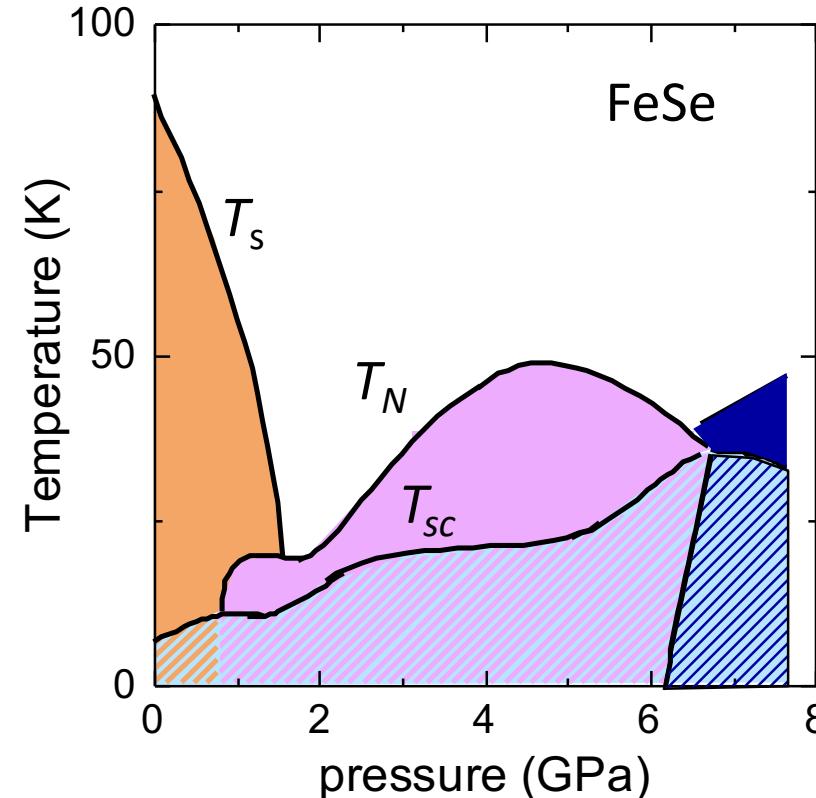
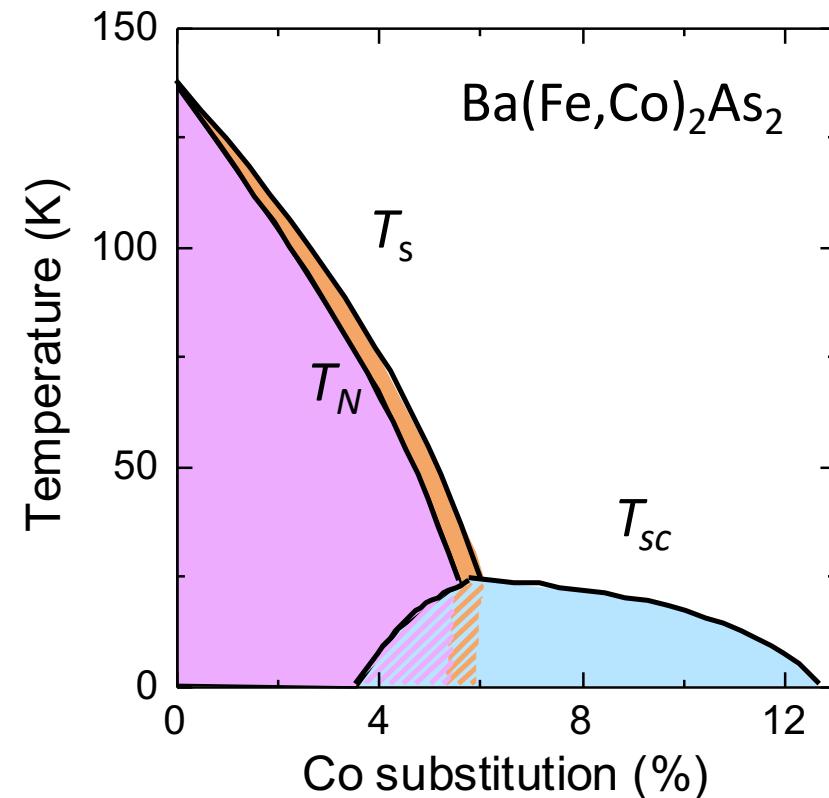
Sun et al., Nat Commun (2015)  
Miyoshi et al., JPSJ (2014)

Böhmer\*, Kothapalli\* *et al.*,  
PRB 100, 064515 (2019)

- Can FeSe and 122-type materials be compared? – many ambient-pressure properties are similar
- What is the relation between nematicity and magnetism and superconductivity in FeSe under pressure?

# FeSe: a mysterious compound

Tuning with hydrostatic pressure

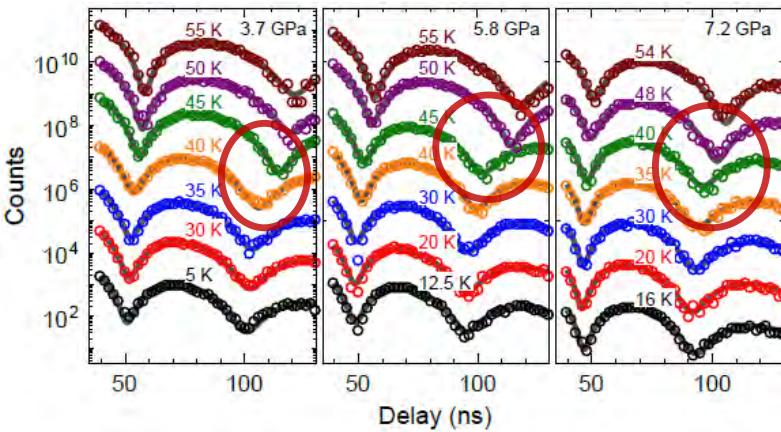
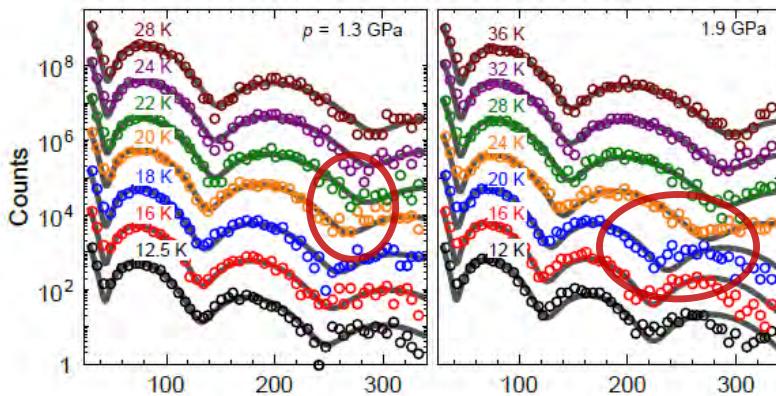
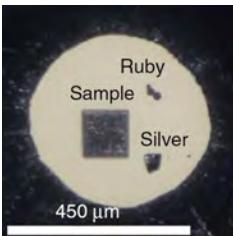


Sun et al., Nat Commun (2015)  
Miyoshi et al., JPSJ (2014)

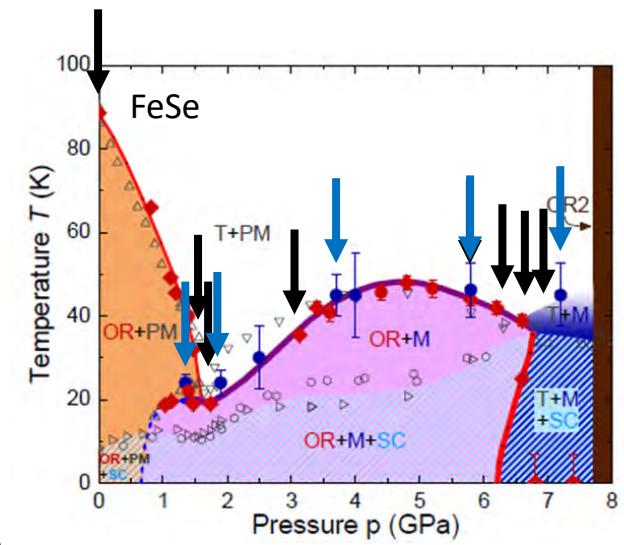
Böhmer\*, Kothapalli\* *et al.*,  
PRB 100, 064515 (2019)

- Can FeSe and 122-type materials be compared? – many ambient-pressure properties are similar
- What is the relation between nematicity and magnetism and superconductivity in FeSe under pressure?

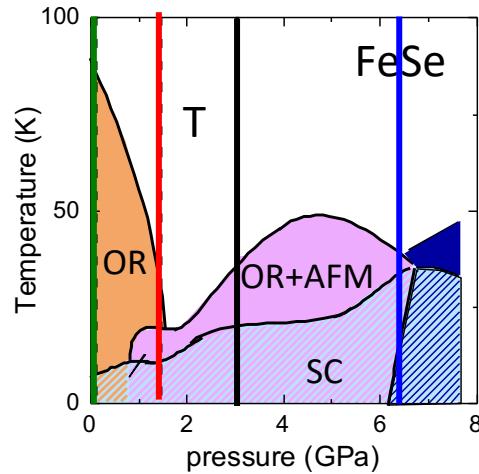
# Lattice parameters and magnetic order in FeSe under pressure



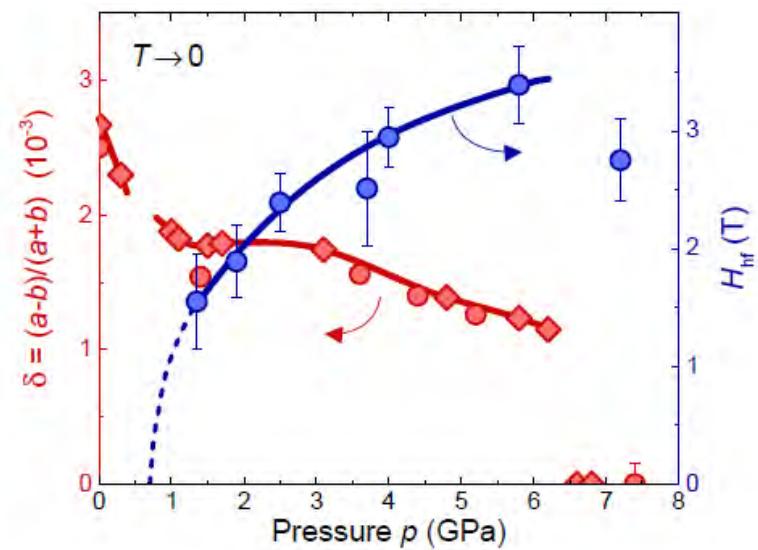
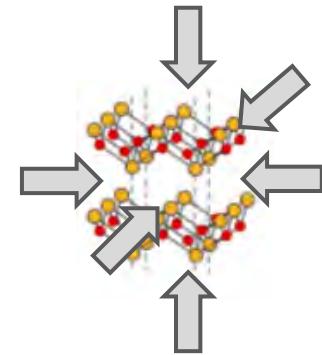
Kothapalli\*, Böhmer\* *et al.*, Nat. Comm. **7**, 12728 (2016)  
Böhmer\*, Kothapalli\* *et al.*, PRB **100**, 064515 (2019)



# Evolution of orthorhombic distortion



- Compatible with a dome of stripe-type magnetism
- A tetragonal magnetic phase at high pressure
- Order parameter coupling and phase diagram topology is a consequence of symmetry, not mechanism

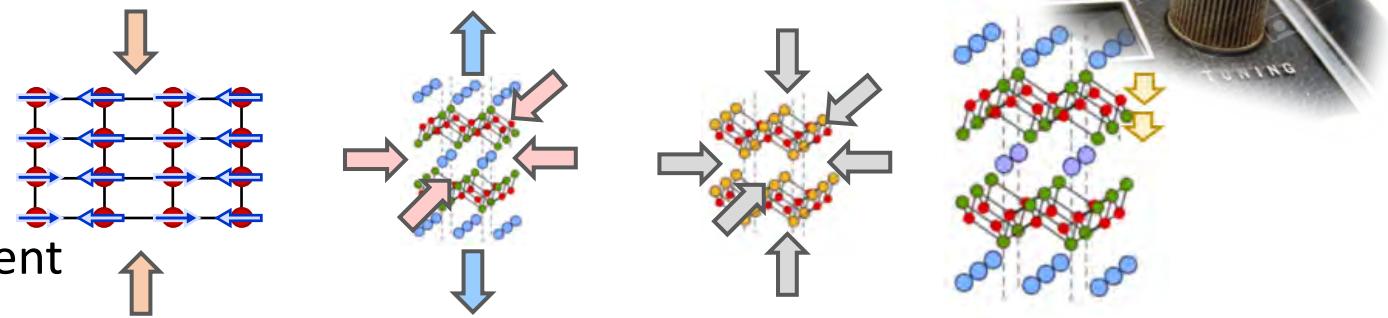


# Conclusions

Diverse magnetic structures from a two-component magnetic order parameter

Interesting intertwined orders emerge

A well-characterized material class that is highly sensitive to elastic tuning



## Nematicity/nematic susceptibility:

Anna Böhmer and Christoph Meingast, Comptes Rendus Physique **17**, 90, (2016) [review]

## Spin-vortex crystal in CaK(Fe,Ni)<sub>4</sub>As<sub>4</sub>:

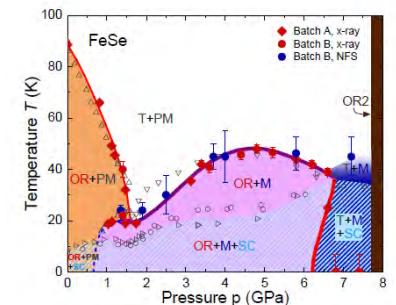
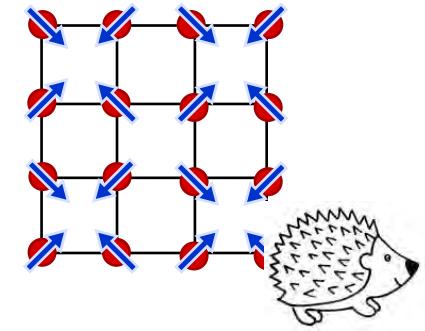
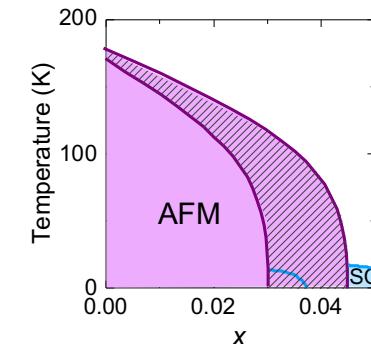
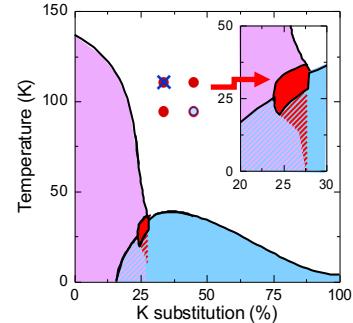
W. Meier et al., npj Quantum Materials **3**, 5 (2018)

## Phase interplay in FeSe:

Anna Böhmer and Andreas Kreisel, Journal of Physics: Condensed Matter, **30**, 023001 (2017) [topical review]

## Iron-based materials, broad audience:

Anna Böhmer and Andreas Kreyssig, Physik in unserer Zeit **48**, 70 (2017) [in German]

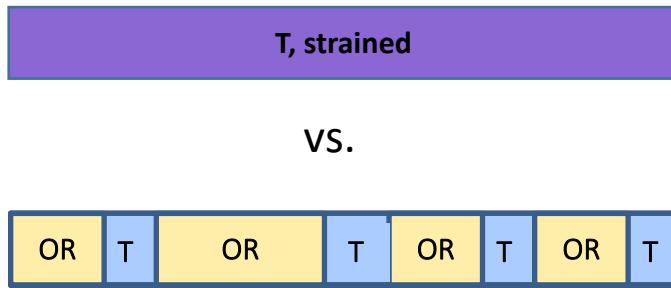




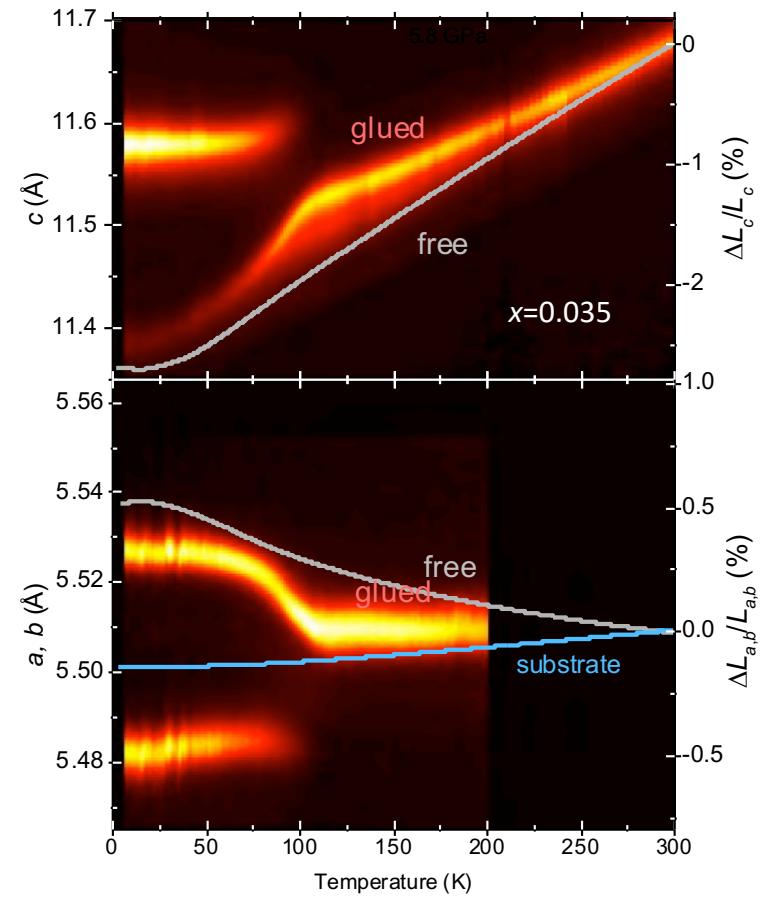
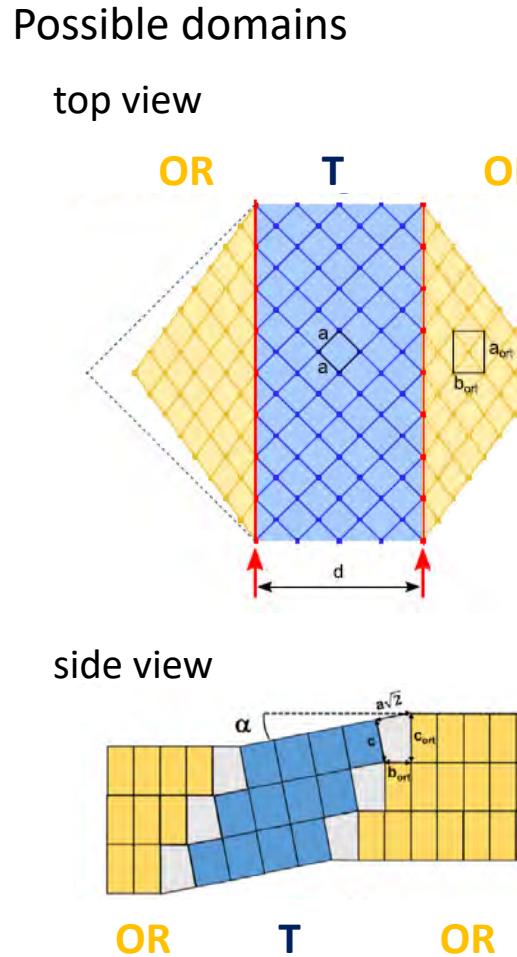
# Consequences of imposing strain *not* stress



$$L_{\text{substrate}} = L_{\text{sample}}$$

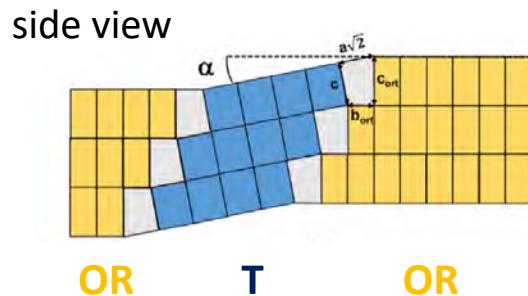


T=tetragonal, paramagnetic  
OR=orthorhombic, stripe-SDW

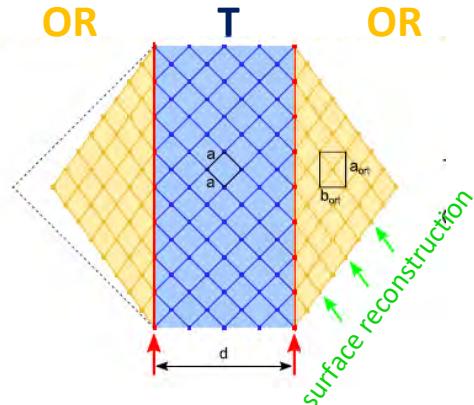


# Observation of unusual domains in strained $\text{Ca}(\text{Fe}_{1-x}\text{Co}_x)_2\text{As}_2$

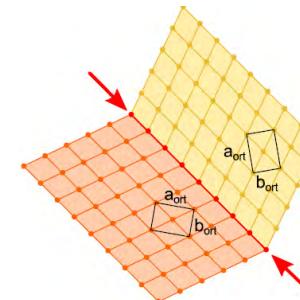
Fente, Correa-Orellana, Böhmer,..., Suderow  
PRB **97**, 014505 (2018)



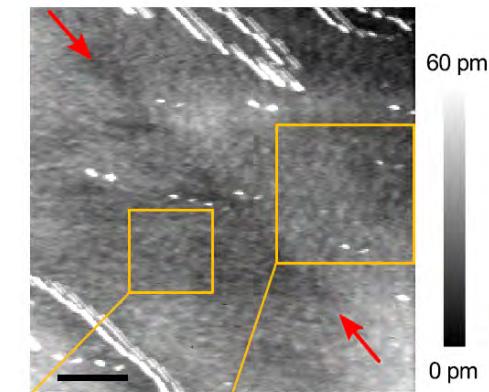
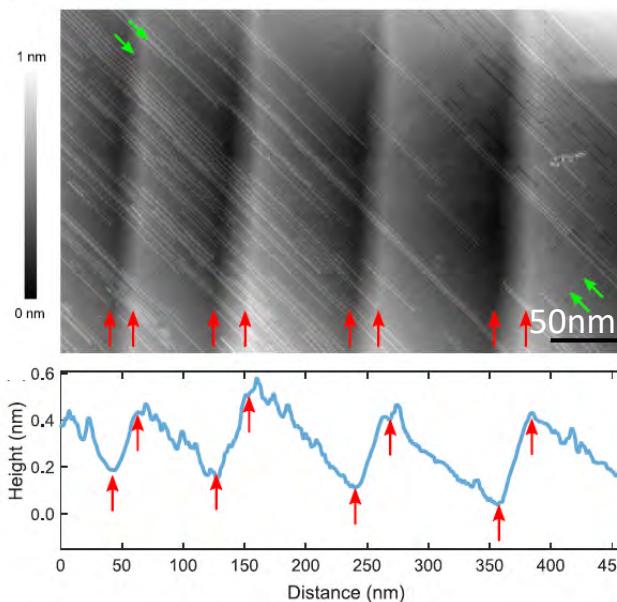
top view



Large domains are orthorhombic below  $\sim 70$  K

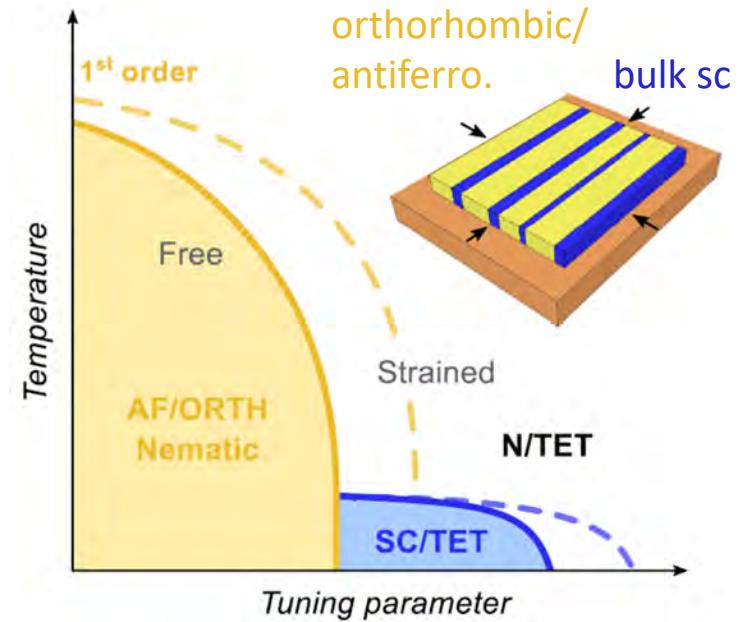
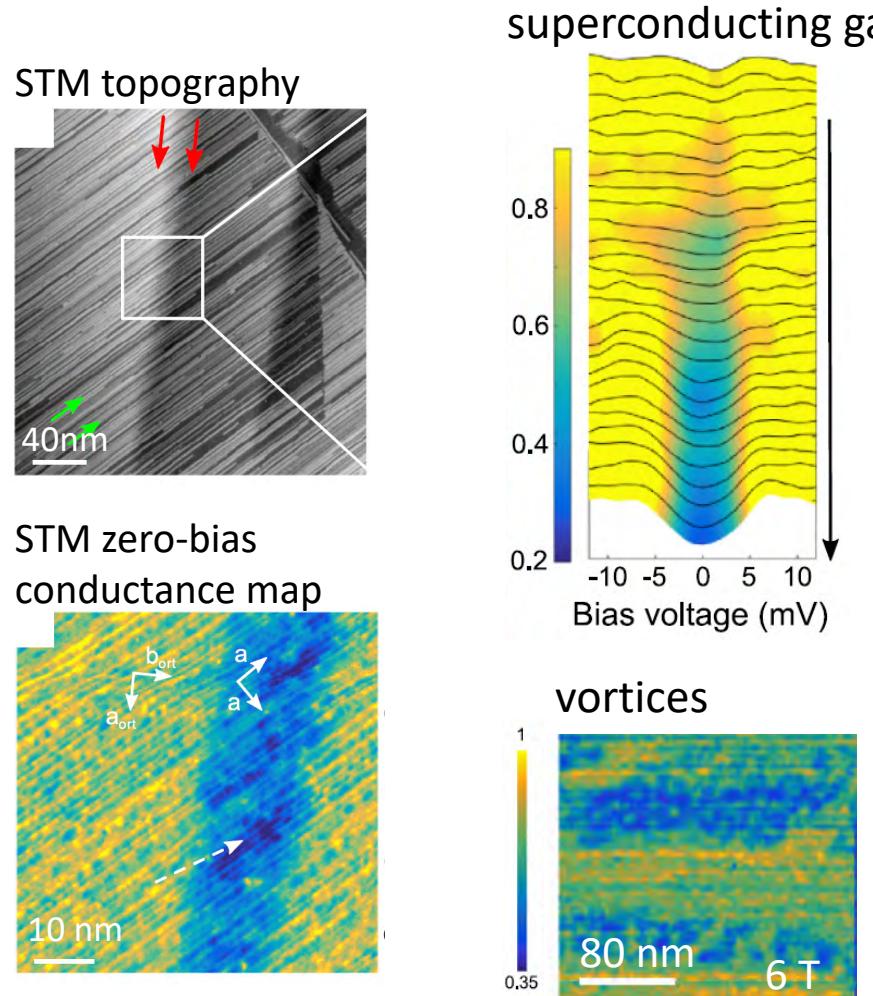


AFM topography



# Nanoscale superconducting domains

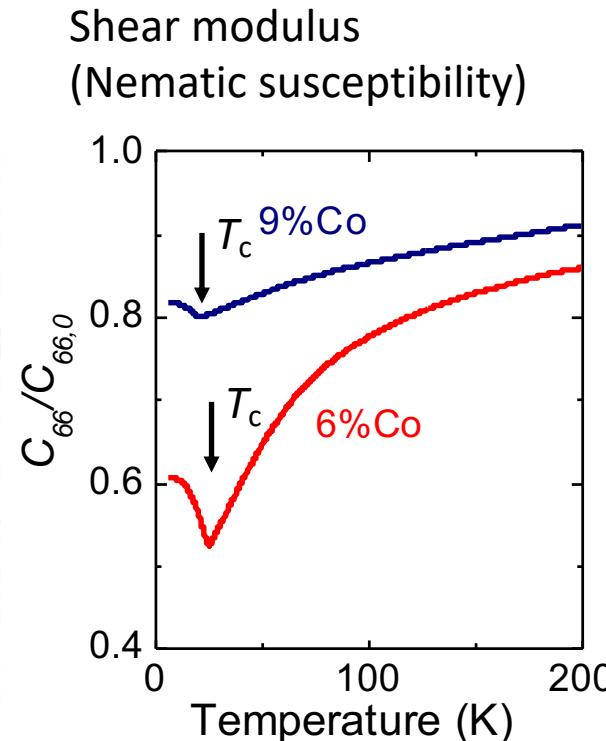
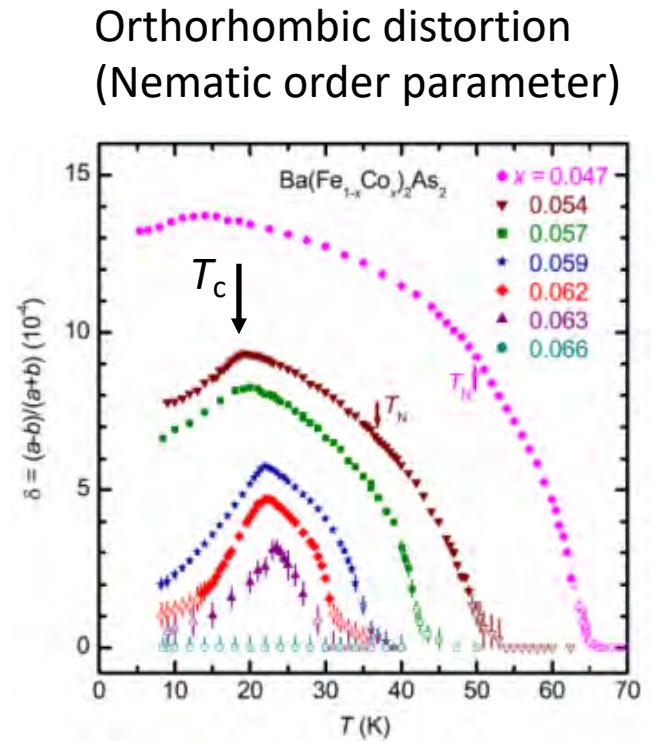
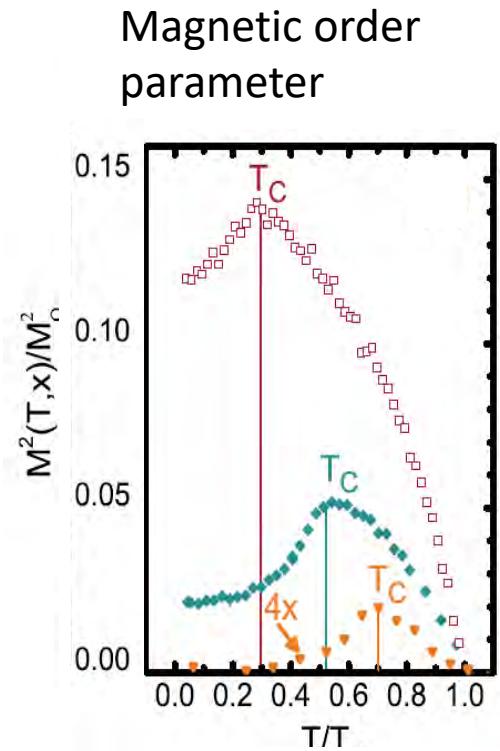
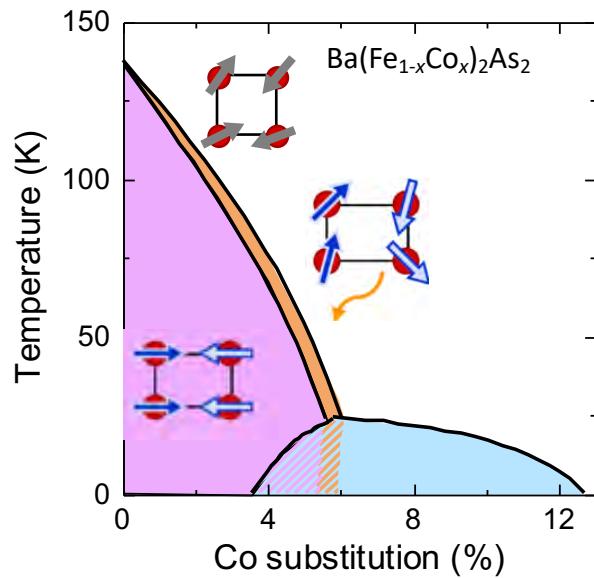
Fente, Correa-Orellana, Böhmer,..., Suderow  
PRB **97**, 014505 (2018)



A strain-induced nm-scale domain structure of alternating orthorhombic/antiferromagnetic and tetragonal/superconducting regions

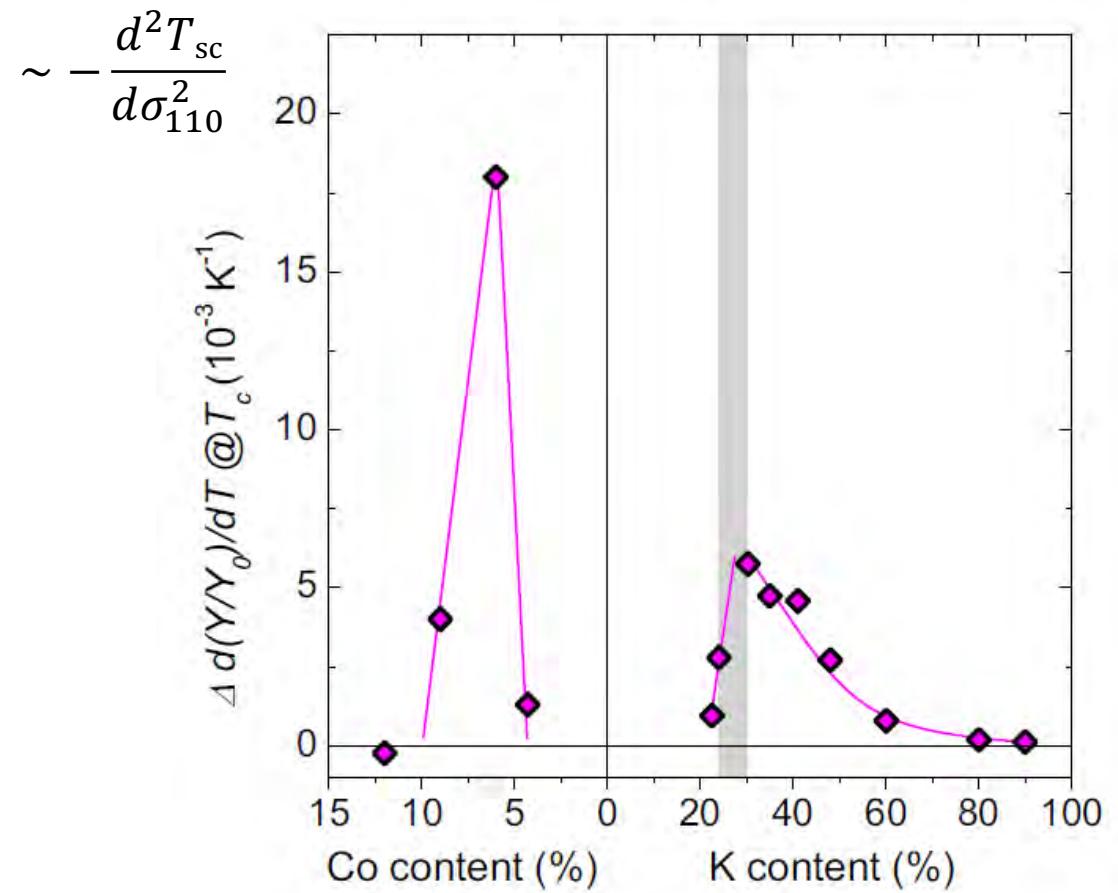


# Competing with superconductivity

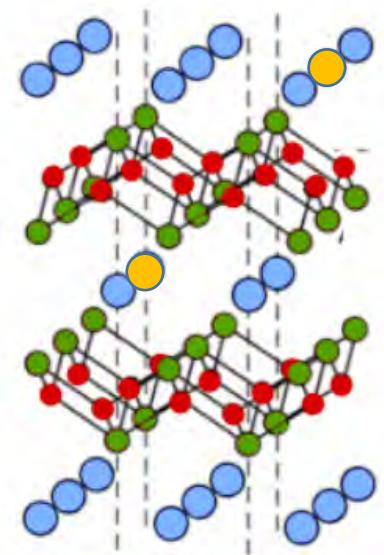


- Coexistence of superconductivity and magnetism/nematicity in a single phase
- Competition for the same electrons
- Results in a huge effect of superconductivity on lattice constants and elastic modulus

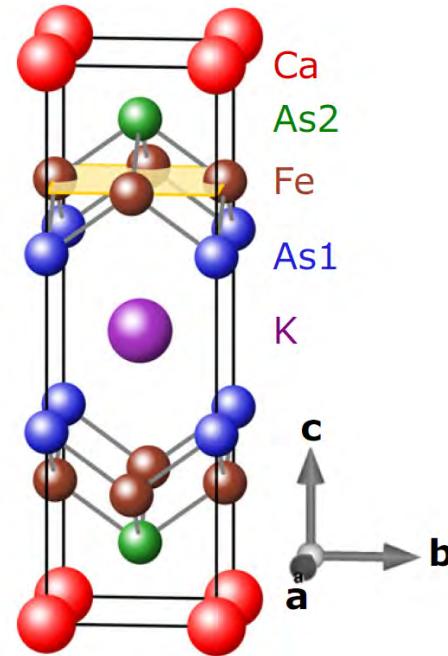
# Resulting tunability of $T_{sc}$



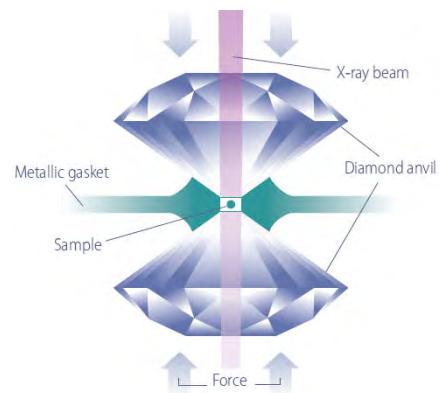
# Tuning via chemical composition



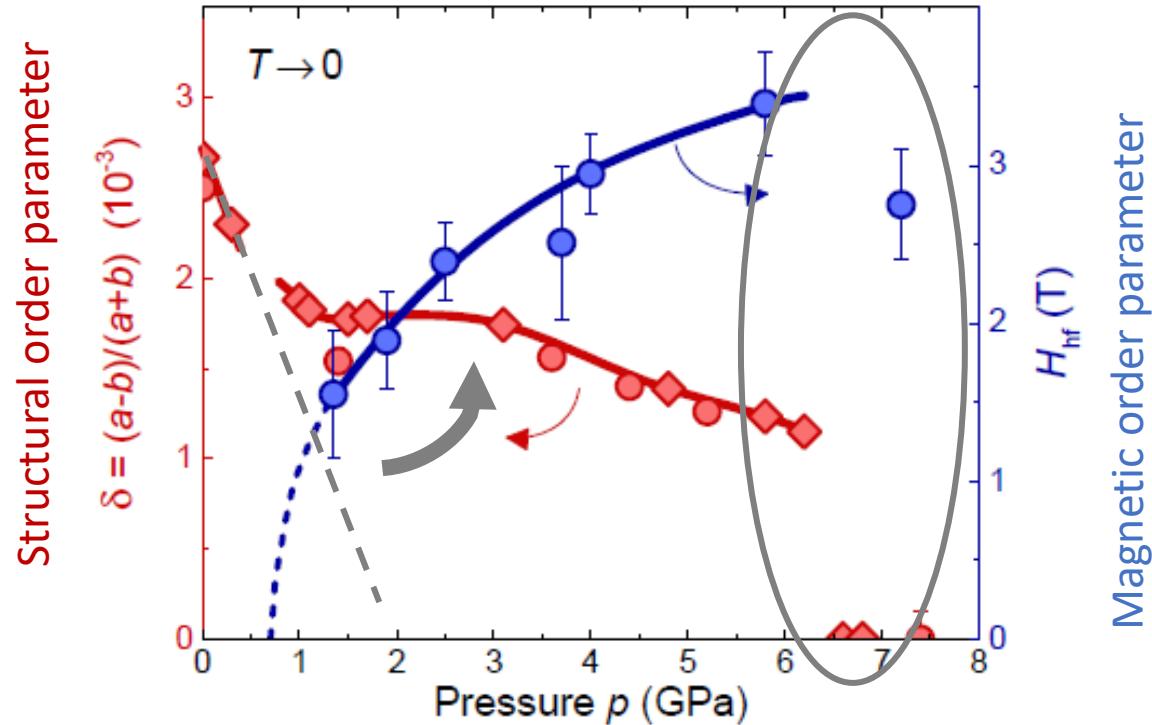
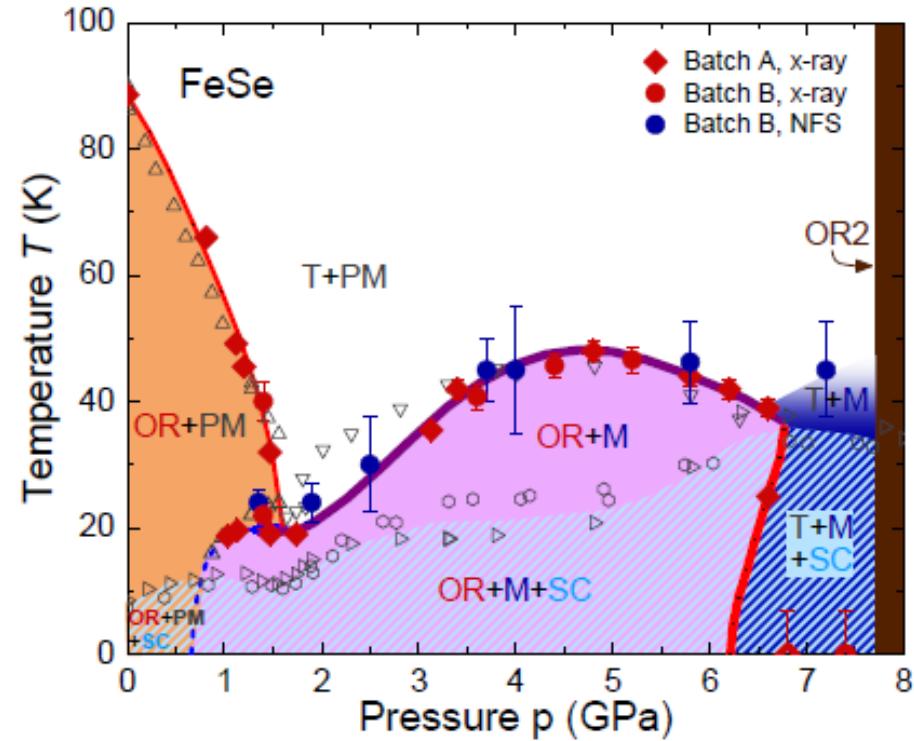
# “Tuning” via the internal crystal structure



# Tuning with hydrostatic pressure

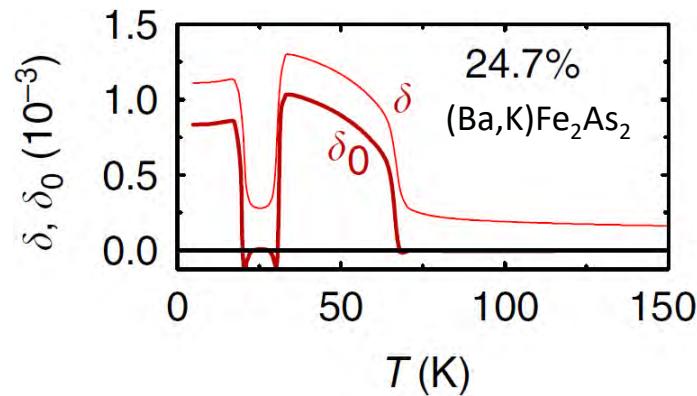
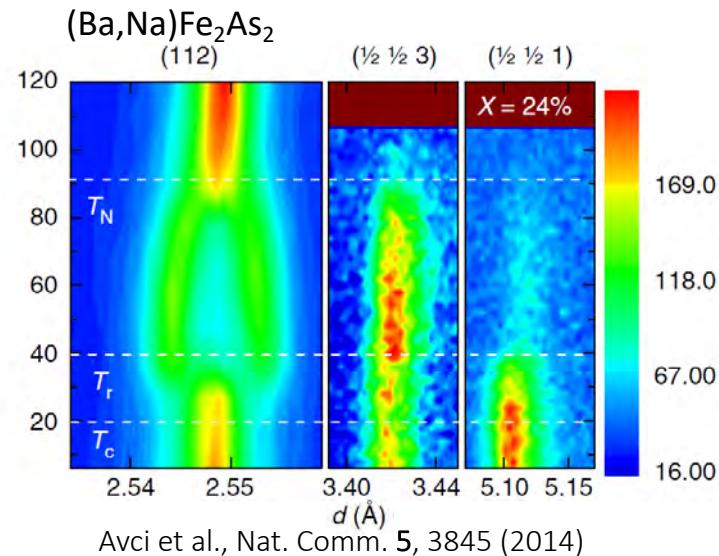


# Detailed phase diagram and order parameter evolution

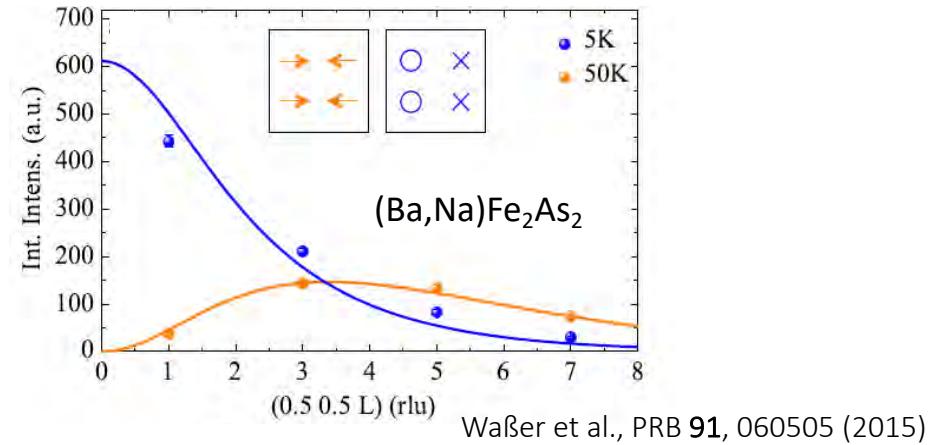


- Cooperative coupling of magnetic order and orthorhombic distortion
- Coupling breaks down at higher pressures

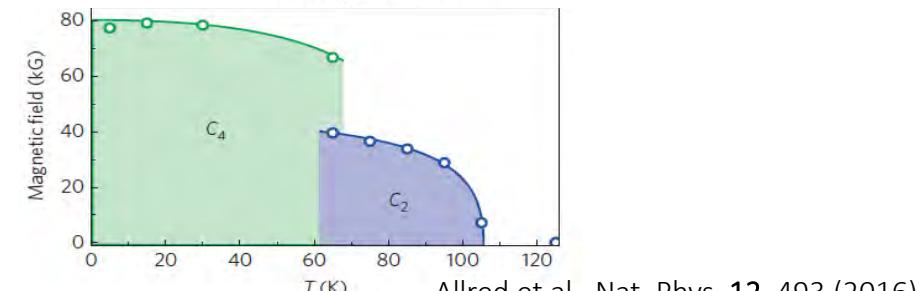
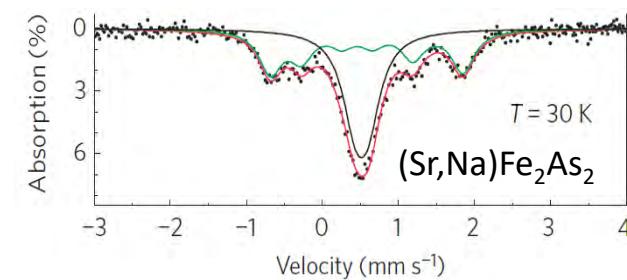
# Identification of spin-charge density wave order



Tetragonal!

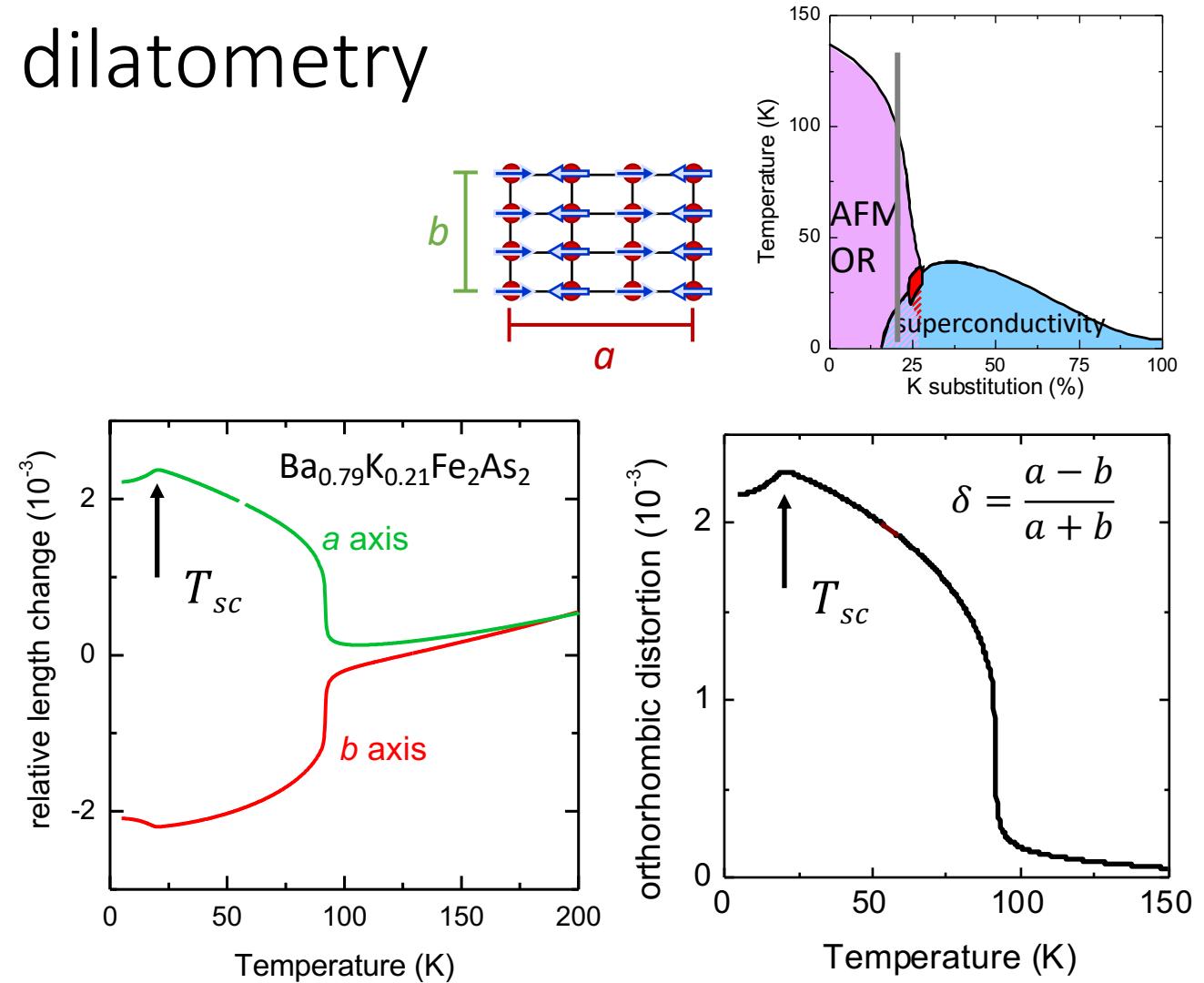
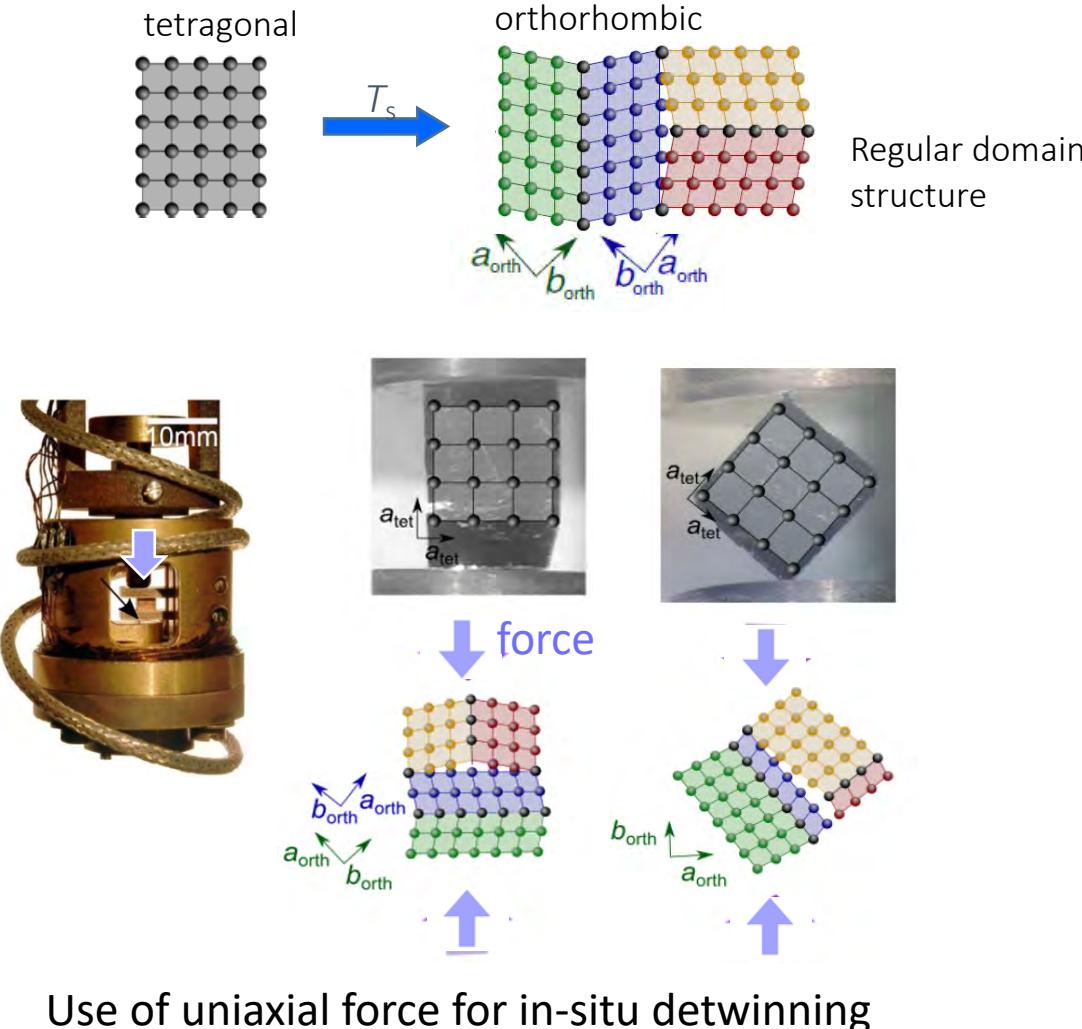


Out-of-plane moments!



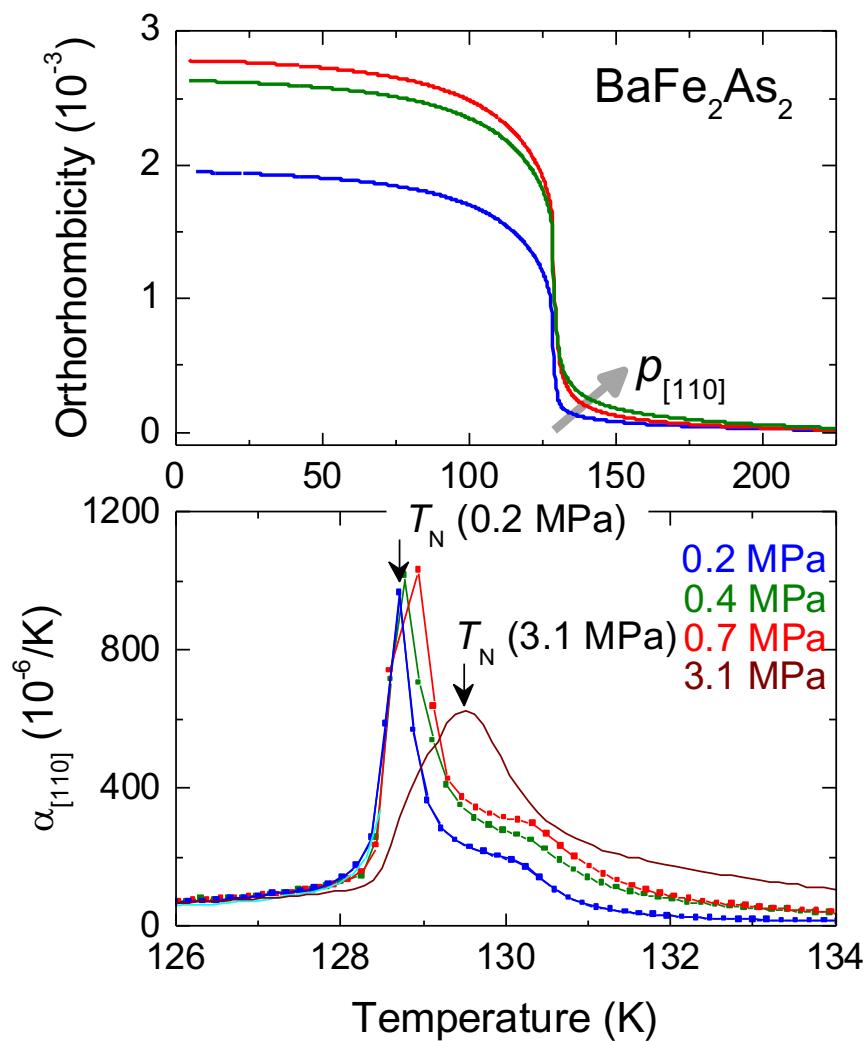
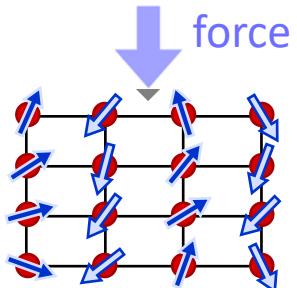
Two distinct magnetic Fe-sites!

# Orthorhombic distortion via dilatometry

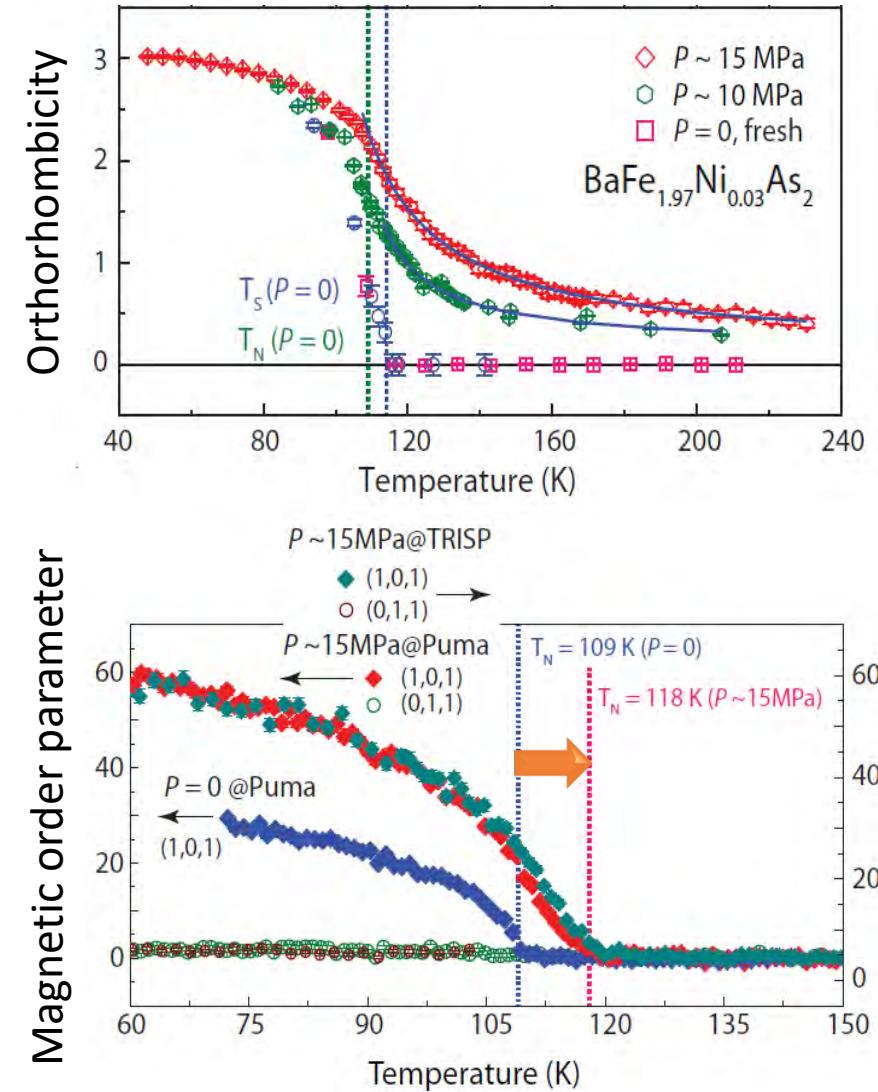


Orthorhombic distortion significantly decreases below  $T_{sc}$   
Competition between superconductivity and orthorhombicity/stripe-type magnetism.

# Tune stripe-AFM transition via “conjugate” uniaxial stress

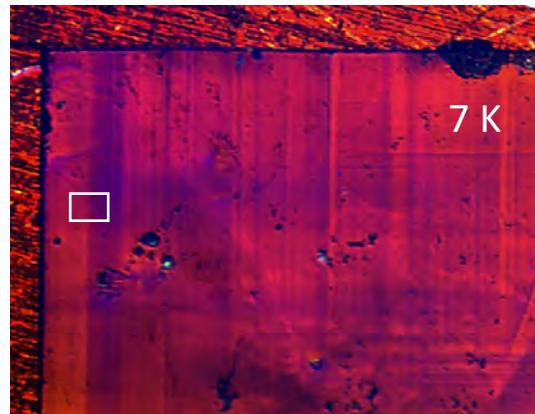
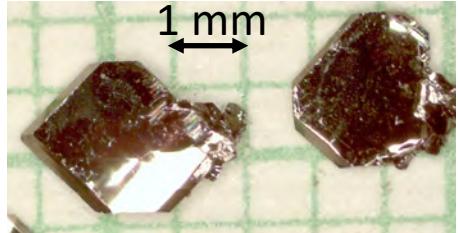


Böhmer et al., unpublished

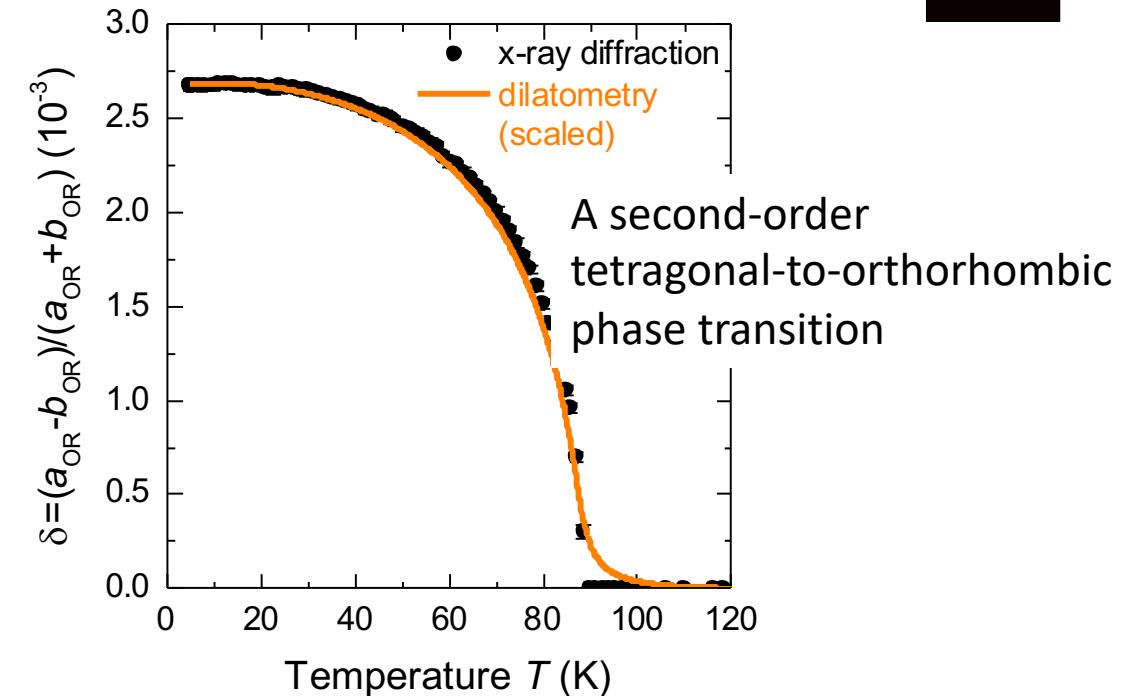
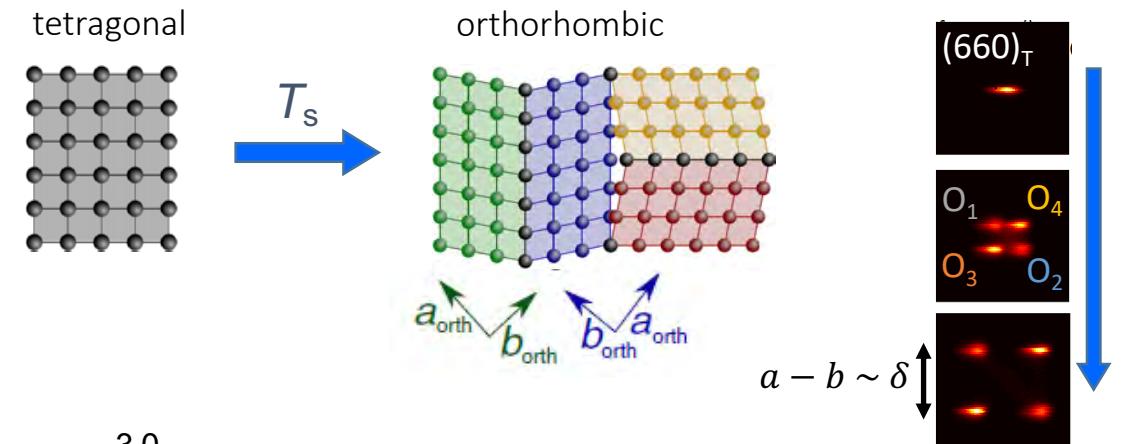
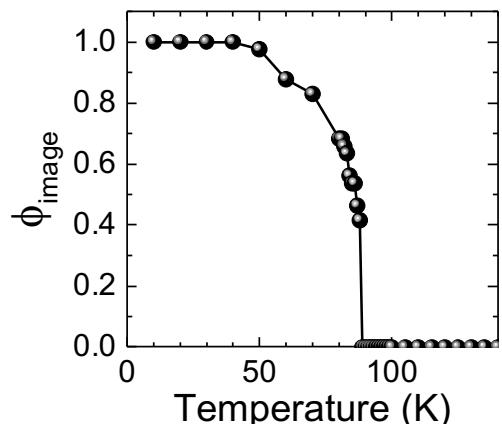


Lu et al., PRB 93, 134519 (2016)

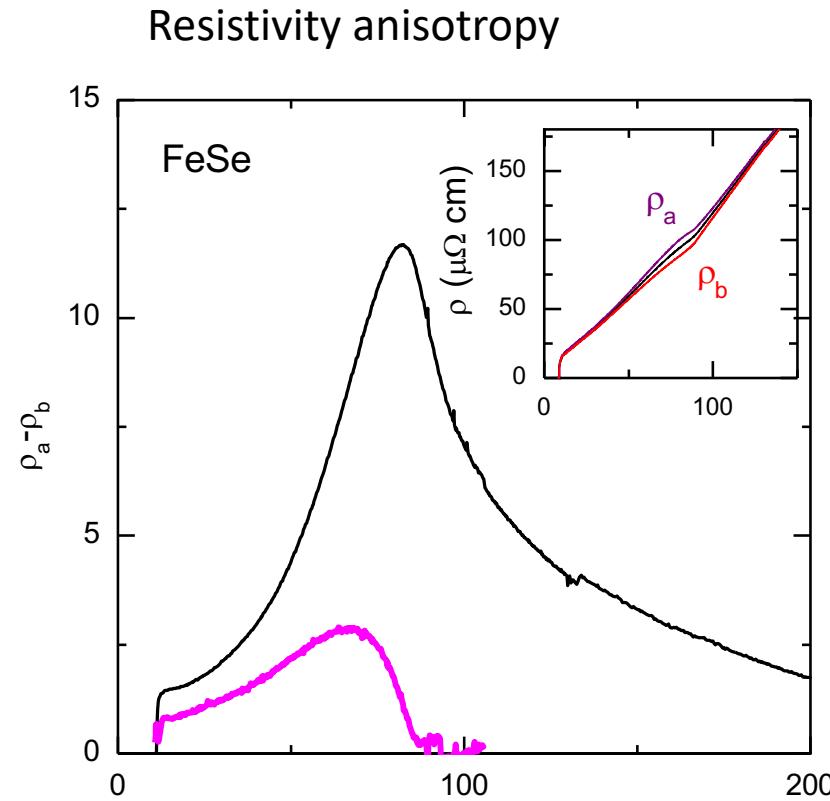
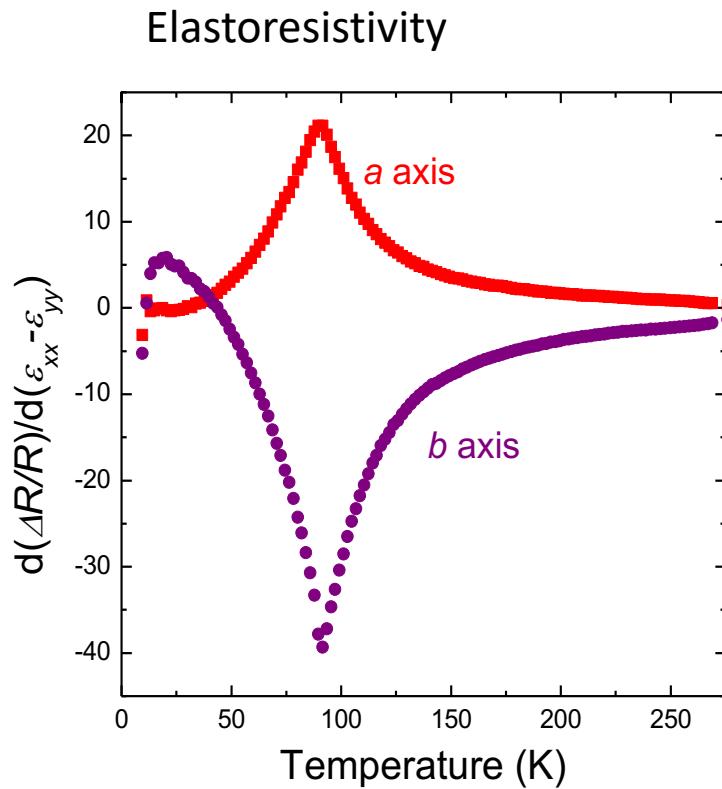
# Tetragonal-to-orthorhombic transition of FeSe



Color difference:

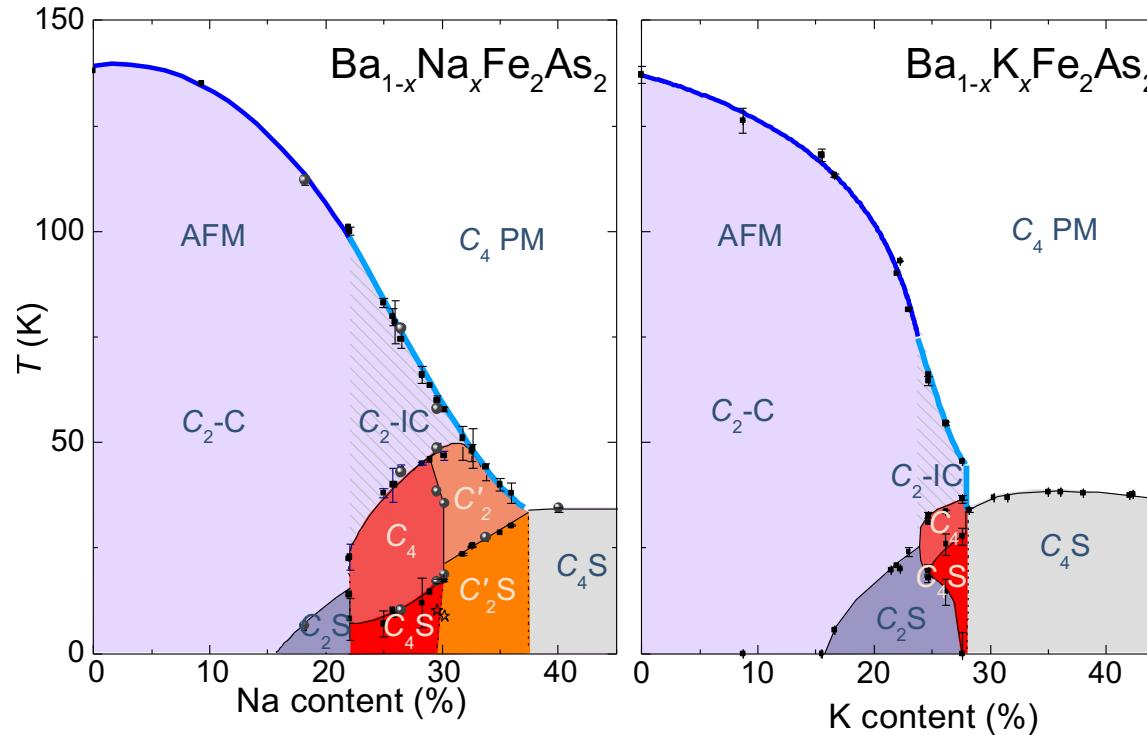


# Nematicity of FeSe

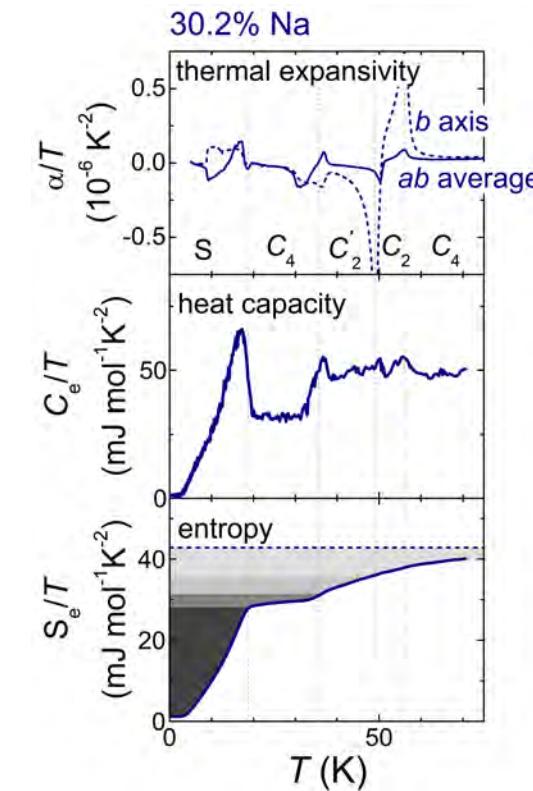


Tanatar, Böhmer, et al., Phys. Rev. Lett. **117**,  
127001 (2016).

# Unidentified phases: open questions



L. Wang et al., Phys. Rev. B 93, 014514 (2016)



- Incommensurate magnetic order?
- Different moment orientations?
- Different  $c$ -axis coupling?

# Tetragonal-to-orthorhombic transition of FeSe

