Нейтронные исследования железо-содержащих сверхпроводников

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From 2008: **Fe-based superconductors**

(pnictides and chalcogenides)

Cuprates are not an exceptional case in high-Tc

“Copper age” transformed to “Iron age”

huge amount of data

hope to understand the phenomenon of HTSC

Are higher Tc’s possible?

Composition, structure, phase diagrams, properties.

Spin dynamics: doped vs undoped, local vs itinerant.

Magnetic resonance.

Similar or not to cuprates?

Pnictides: binary compounds with a *pnictogen* element

V-th column in the Periodic Table: Pn = N, P, As, Sb, Bi.
typically Pn^{3-} anions

FeAs compound itself is a (heli)magnetic metal with Fe^{3+} cations

Pnictide-*based* superconductors:
(formally) *divalent* ion Fe^{2+}

General formulae:

X-FePn

with Fe^{2+}, Pn^{3-} and $X=X^{+1}$

Superconductors with Pn = P, As, Sb

while the highest Tc's with As

X^{+1}-$Fe^{2+}Pn^{3-}$

Historically first: ***LaOFeAs*** (26K, by H.Hosono, Japan, 2008)
looking for a transparent magnetic semiconductors for screens…
(CuS first, then FeP with 5K)

1111: X = **Re**O **Re=**La, Ce… or SrF, CaF

SC: doping ($O_{1-x}F_x$) or ($Re'_{1-x}Re''_x$) magnetic AF order in parent compounds
maximal Tc>50K with Nd, Gd, Sm (56.3K in $Gd_{0.8}Th_{0.2}OFeAs$)

then ***$BaFe_2As_2$*** (by D.Johrendt, Germany)

122: X = (**Me**)/2, **A**lkali **H**alides, **Me** = Ca, Ba, Sr
SC: doping with Alkali metals ($Me_{1-x}A_x$),
also at the Fe-site ($Fe_{1-x}Co_x$…) and even with an isovalent pnictogen ($As_{1-x}P_x$…)
maximal Tc=38K in $Ba_{0.6}K_{0.4}Fe_2As_2$

LiFeAs, NaFeAs (S.Clarke, England; Ch.Jin, China)

111: X = **A**lkali metal, **A**=Li, Na
maximal Tc ~ 18K (stoichiometric compounds)
known since 40 years

Chalcogenides (Ch): S, Se, Te

11: X = 0, **$Fe^{2+}(Ch)^{2-}$** (by M.-K. Wu, Taiwan)

FeTe - non-SC, FeSe: Tc=8K, max Tc ~12-14K near compositions $FeSe_{0.5}Te_{0.5}$
(particularity: a few % “extra” Fe are required in order to stabilize
crystal structure of the SC phase: $Fe_{1+y}Te_xSe_{1-x}$)

Tc increases under pressure (at a few GPa up to 20-40K depending on composition)

Attempts to dope with Alkali metals have resulted in a new family:
$A_{0.8}(FeSe)_2$ with A = K, Cs (J.Guo, China, PRB2010)

Formal Fe valence far from +2?

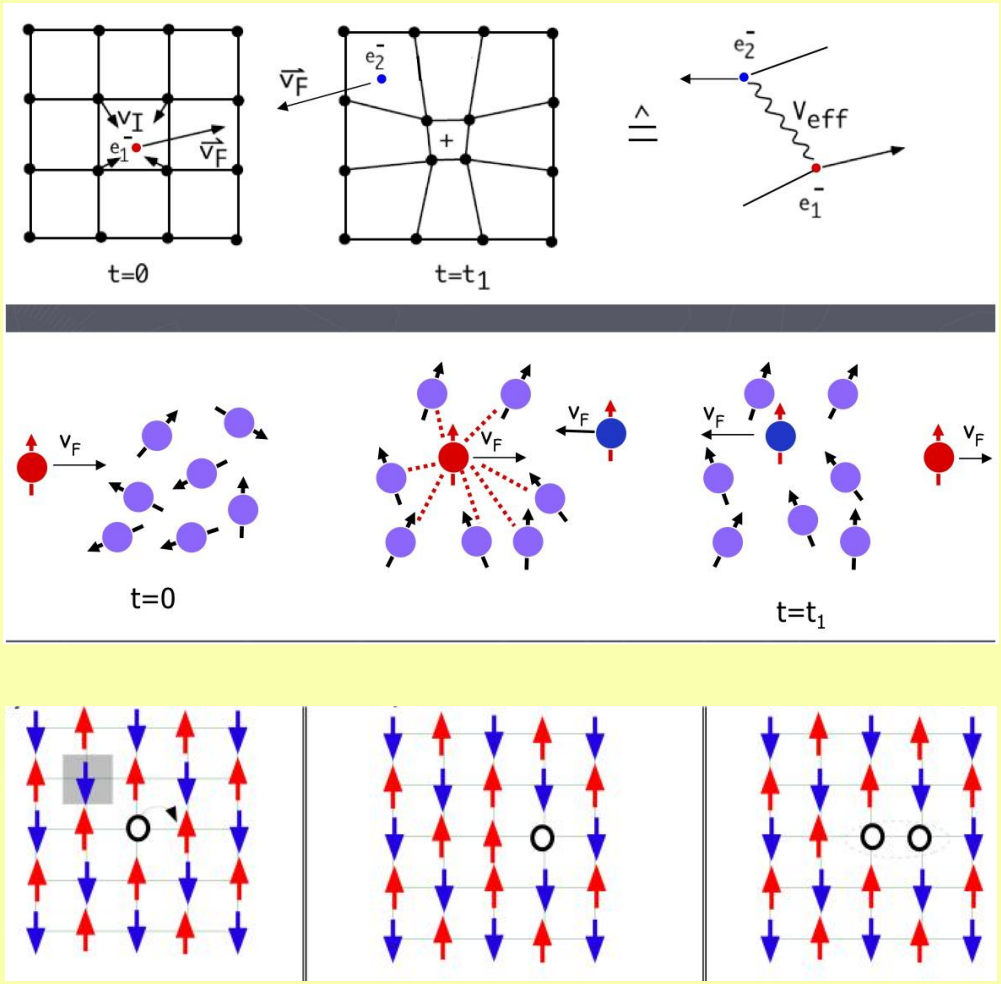
Correct composition: $A_{0.8}Fe_{1.6}Se_2$ or $A_2Fe_4Se_5$ (245-compounds) with Fe^{2+}
A=Rb, also (RbTl), (KTl)
Tc ~ 30K

FeSe layers with Fe-vacancies (ordered)

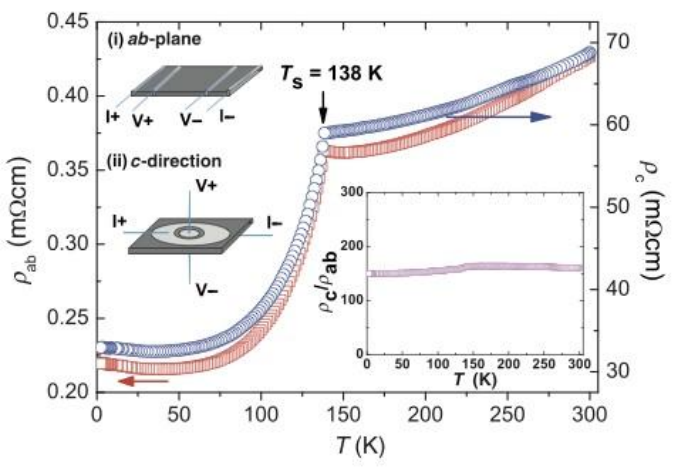
Unconventional superconductors

Early phonon DOS measurements in LaOFeAs
with the calculated $\alpha^2F(\omega)$ have shown *insufficient el-ph coupling*

Non-phonon mediated: electron fluctuations (Berk-Schrieffer, 1961)

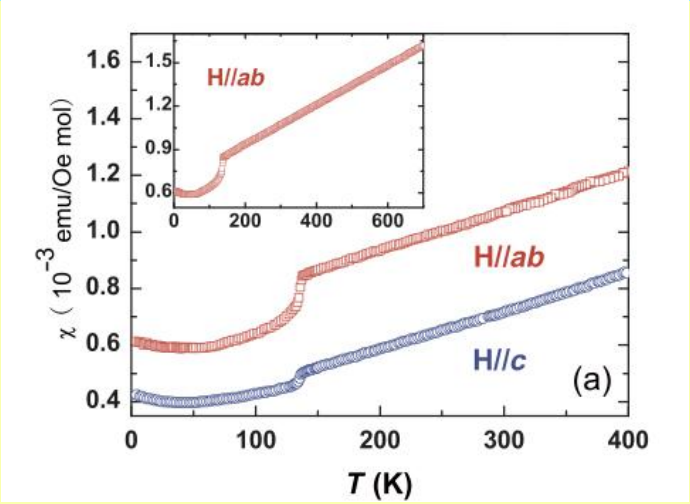


Strongly anisotropic compounds



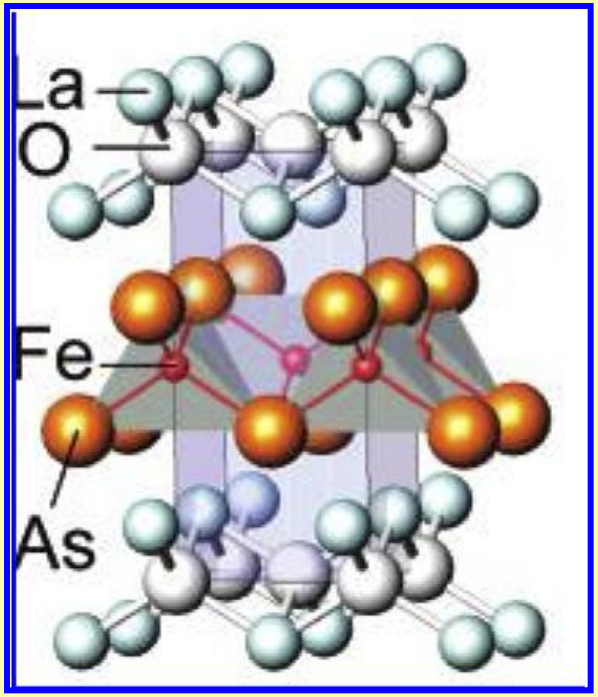
$BaFe_2As_2$

Wang *et al*, PRL 2009



Structural transition to a lower symmetry phase

magnetic long-range order



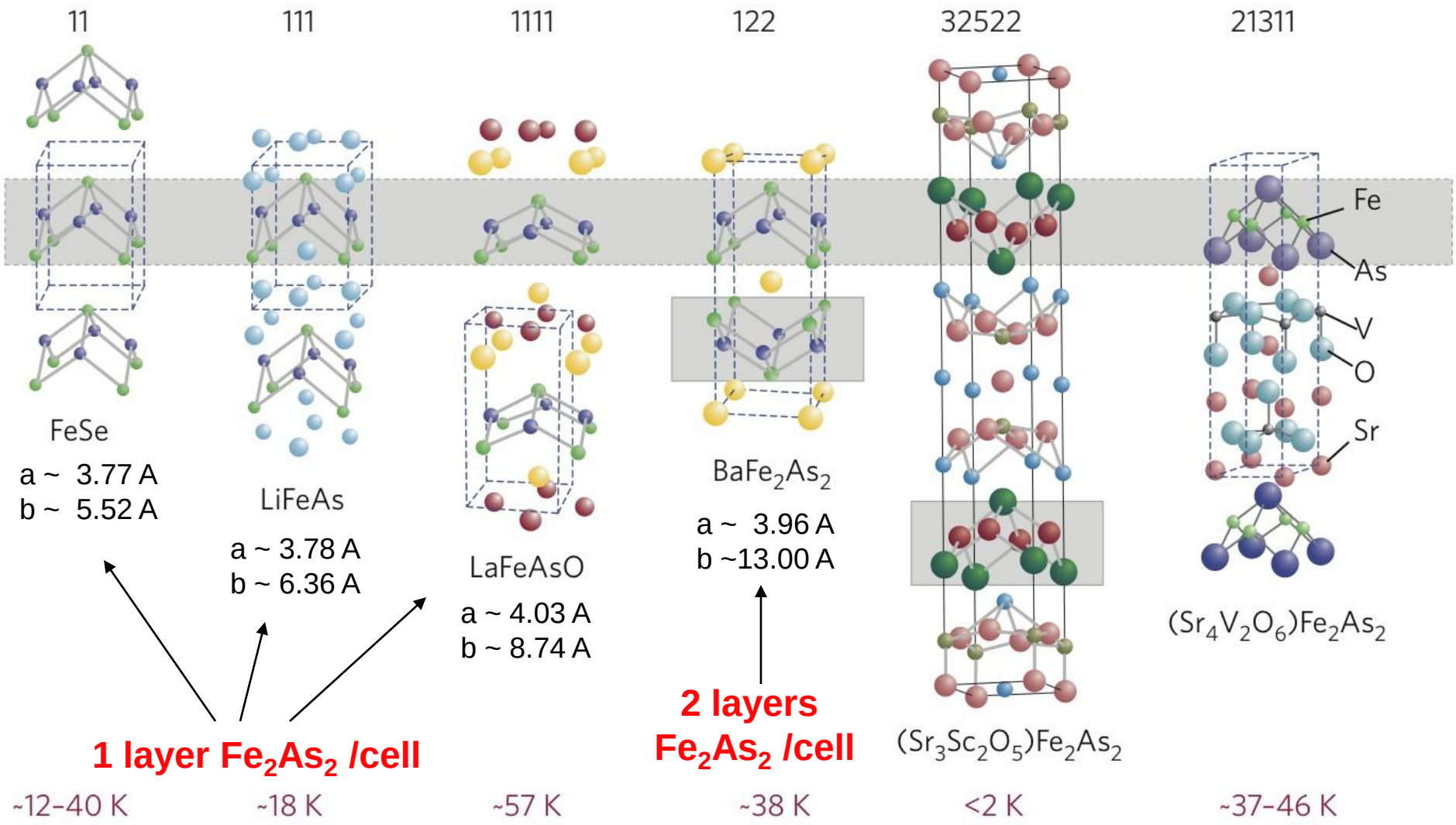
LaOFeAs

Kamihara *et al*

JACS 2008

Layered crystal structure but with a *tetrahedral* coordination, *not planar* as in cuprates: a more 3-D atomic arrangement

Crystal Structures



C.W.Chu Nature Physics 2009, v.5, p.787-789

Similarity to cuprates:
layered structures, magnetically active layers separated by non-magnetic ones
several structure types, each with some maximal Tc.
However, apparently nothing like double-layers such as in YBCO.

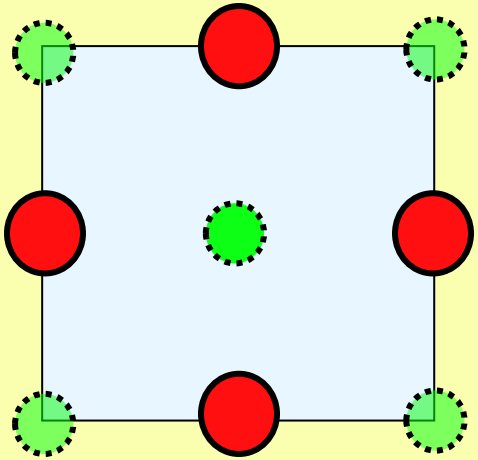
Magnetically active planes in Pnictides and Cuprates

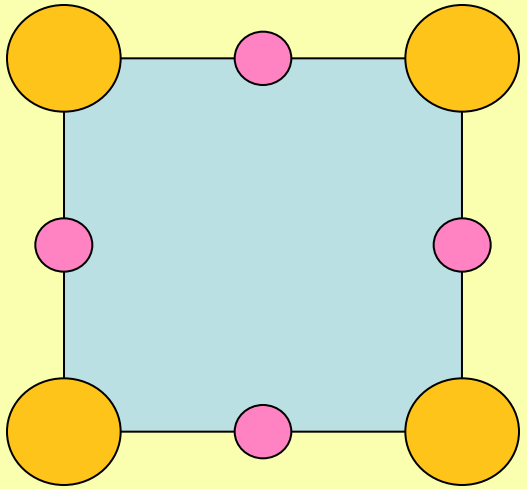
Fe_2As_2: Fe-Fe = 2.70 A

CuO_2: Cu-Cu = 3.85 A

Metal iron (bcc):
a = 2.866 A
Fe-Fe = 2.480 A

Magnetic moment
Fe = 2.2 μ_B





Fe (z = 0)
As (z≠0)

Cu (z = 0)
O (z = 0)

Parent compounds: magnetically ordered in these layers

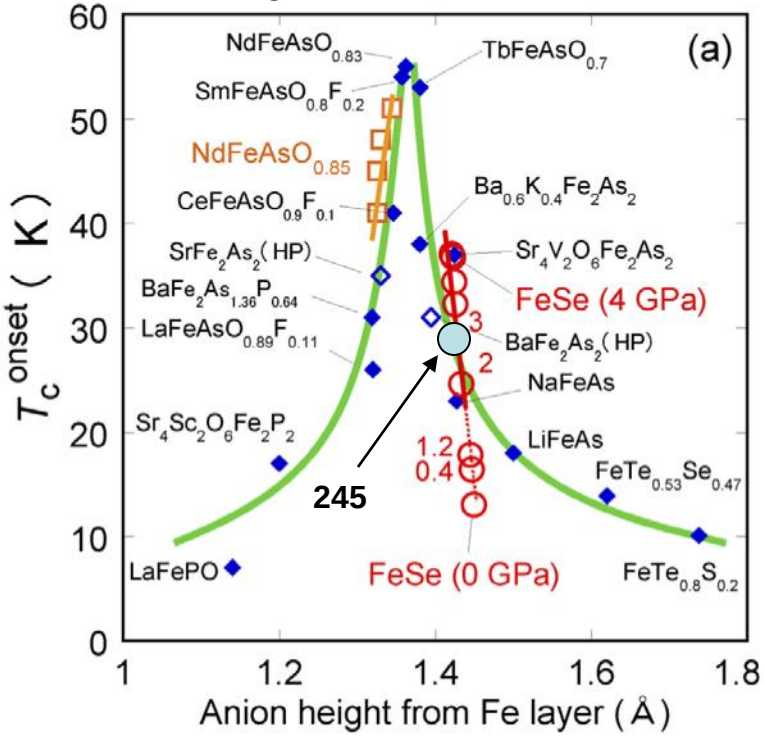
Cuprates: Mott insulators while **Iron-based: (bad) metals**

Spin Fe^{2+} (d^6): S=2
Magnetic moment Fe = 0.1 - 3.4 μ_B

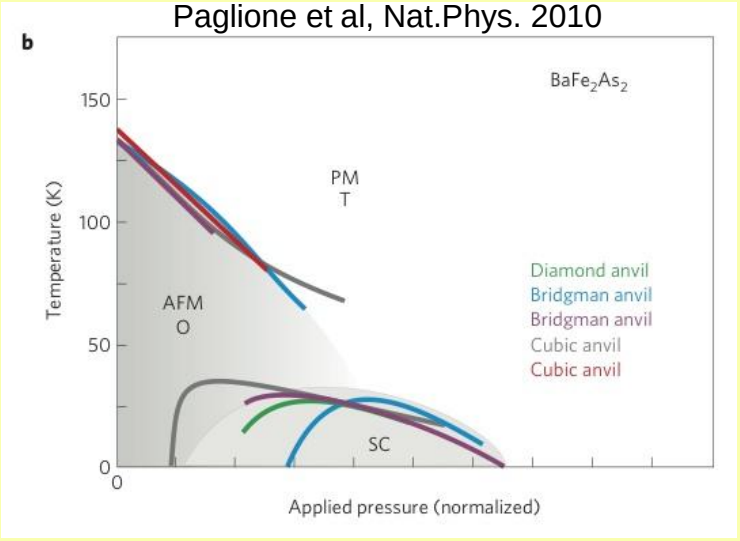
Spin Cu: S=1/2
Magnetic moment Cu ~ 0.5 μ_B

Crystal Chemistry and Superconductivity

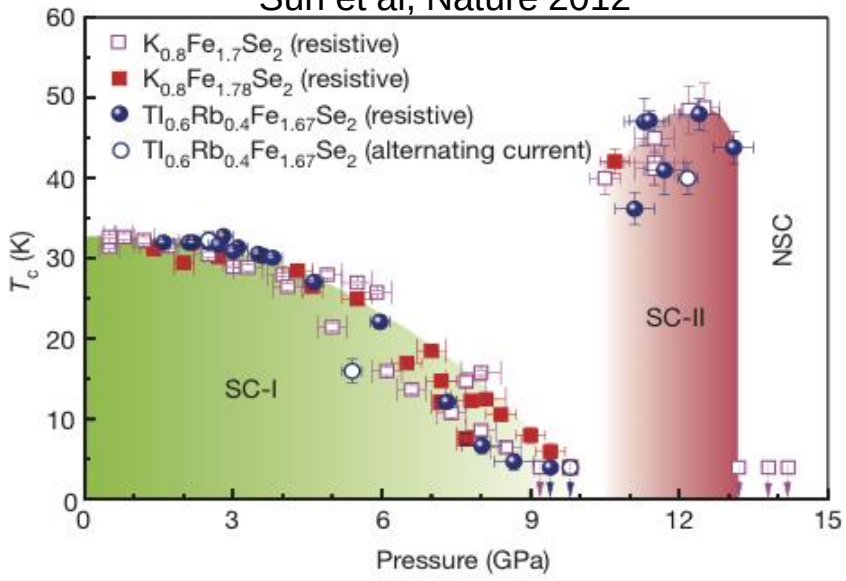
Mizuguchi et al, arXiv-2010



Paglione et al, Nat.Phys. 2010



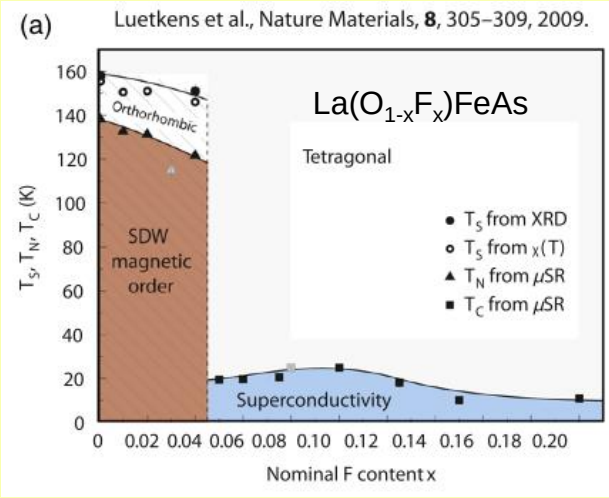
Sun et al, Nature 2012

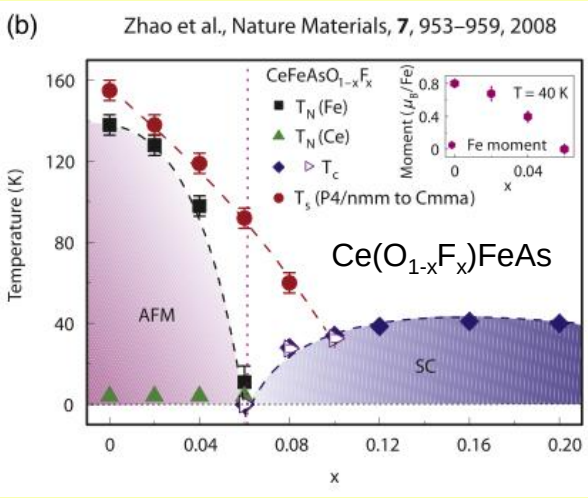


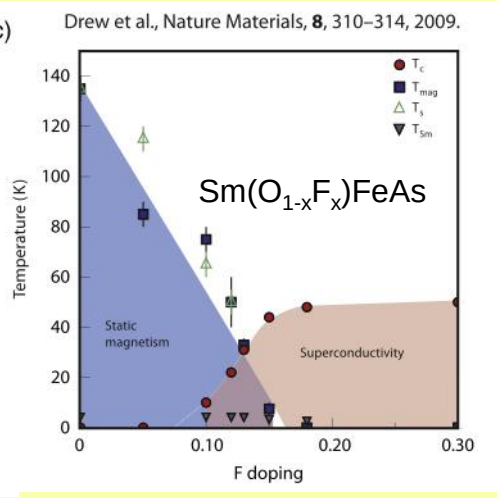
245: possible new path to higher Tc?

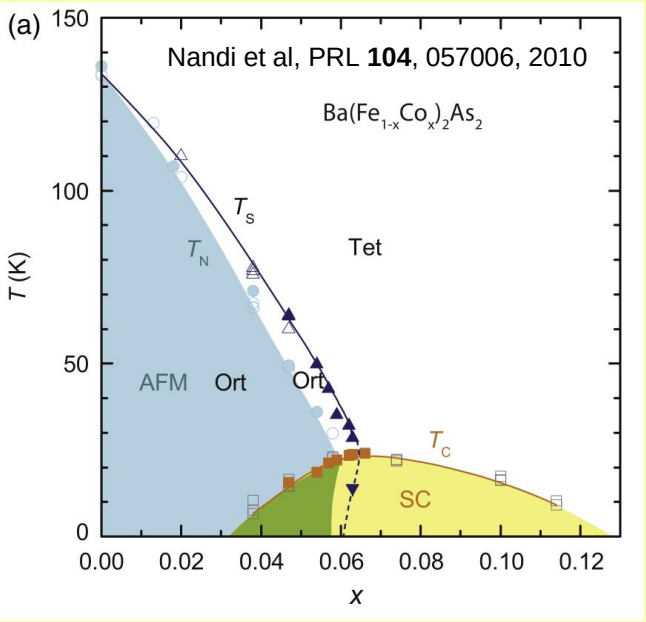
Phase Diagrams: Temperature-Doping

La, Ce, Sm 1111: O-F substituted





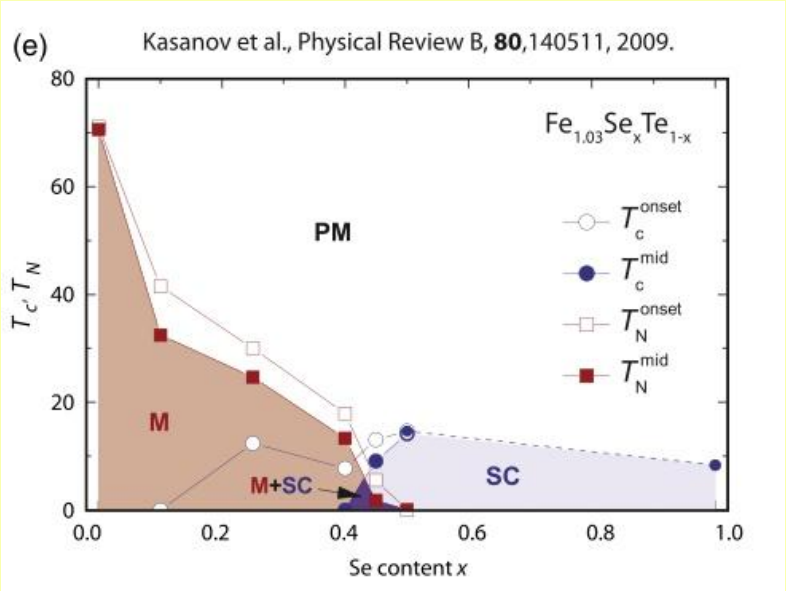




Similarity with cuprates:

undoped compounds with a long-range magnetic ordering and a structural transition

with increased doping the long-range magnetic order is suppressed and the SC state arises



However "details" may be very much different

Ba-122: Co-doped

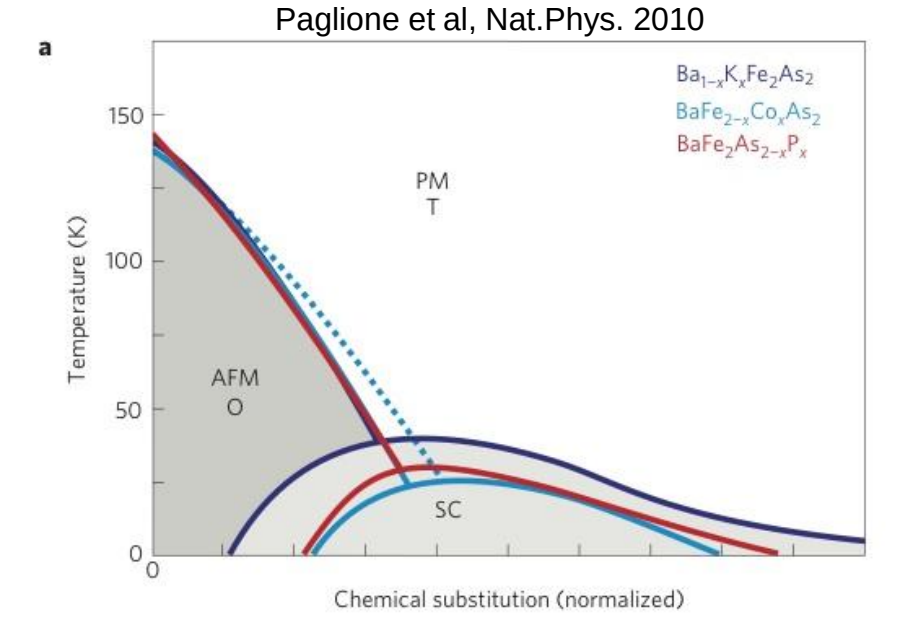
$Fe_{1+y}(Te_{1-x}Se_x)$

Fe-based: distinct from cuprates

Parent Fe-compounds are metals, not insulators contrary to cuprates.

3D character is more pronounced

Paglione et al, Nat.Phys. 2010



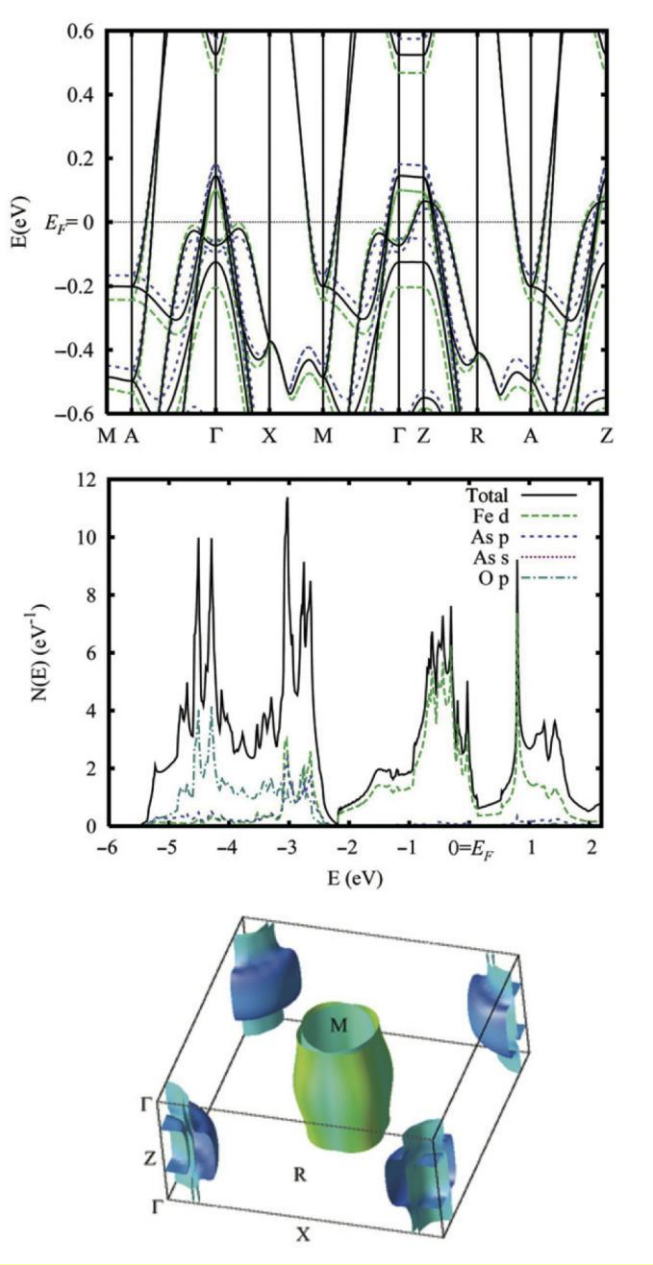
Doping: all is possible electron or hole, iso-valent and also at the Fe-sites

In cuprates: SC degrades rapidly if Cu is substituted

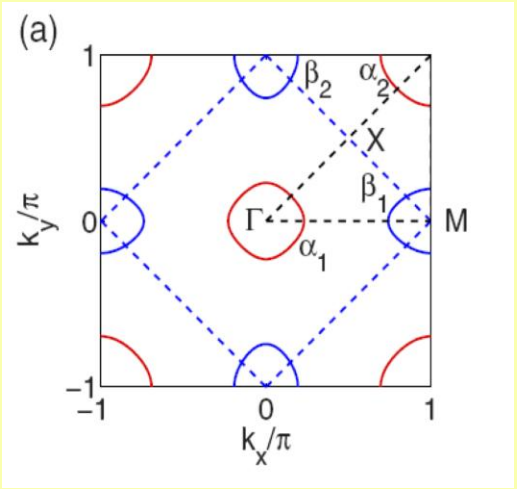
Larger variation of magnetic ordering temperatures and ordered moment than in cuprates.

Cuprates are more "similar" in one structural sub-family
Iron-based exhibit more diverse properties
Can the latter be indeed considered as a single family?

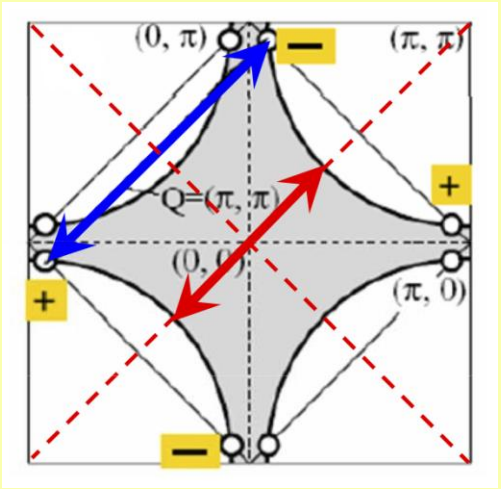
Electronic structure



LDA bands in LaOFeAs, Singh et al 2008



Fe



Cu

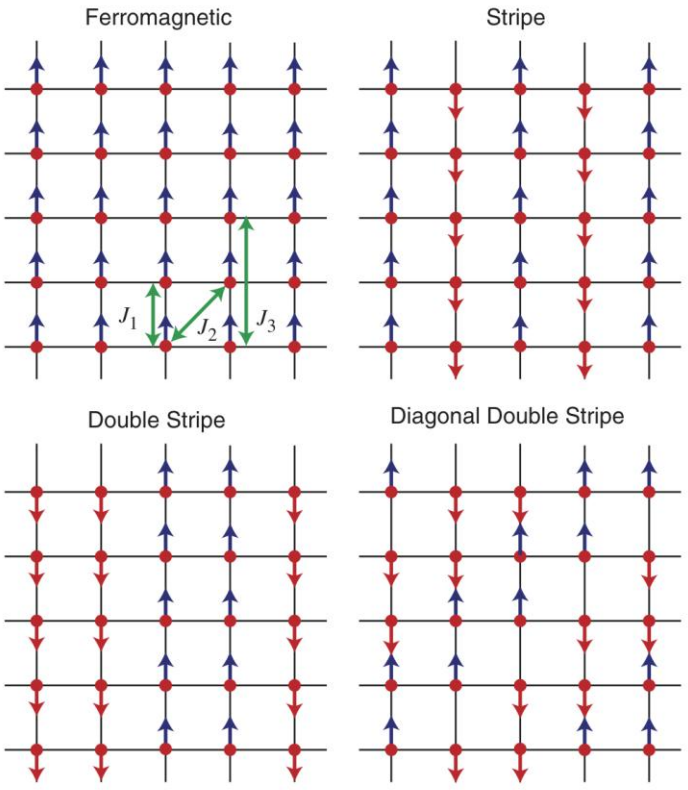
Several electron and hole bands in Fe-based mostly due to d-electrons of Fe contrary to one single band in cuprates

More complicated:

multi-band, multi-orbital, multi-gap

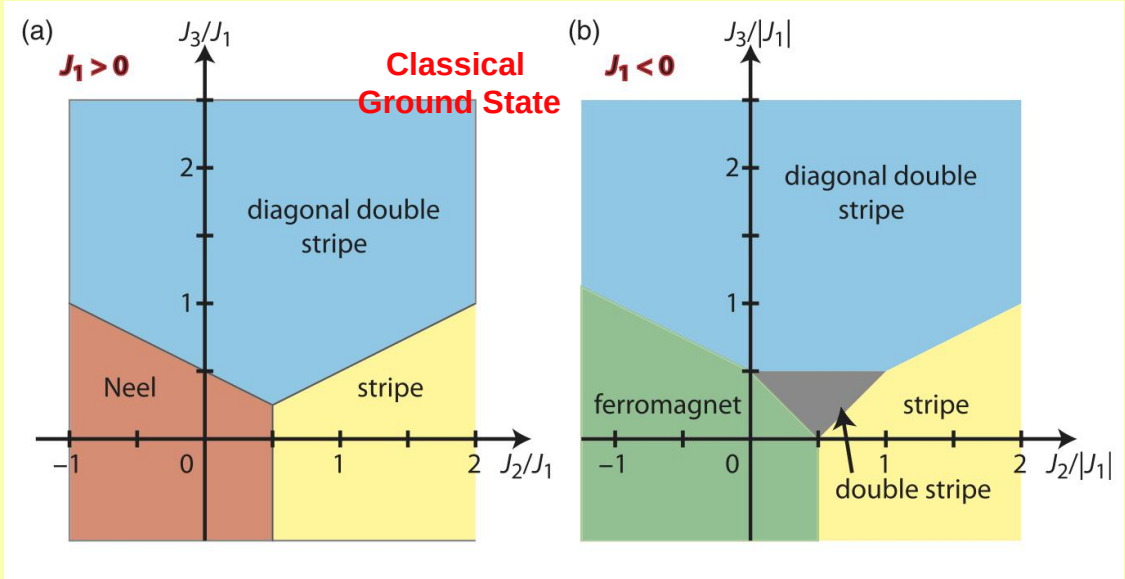
Magnetic Structures

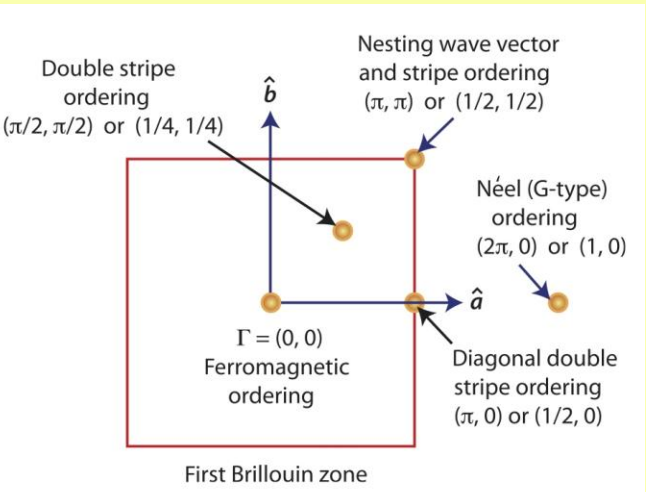
The simplest picture: local moments on Fe-sites, square plane

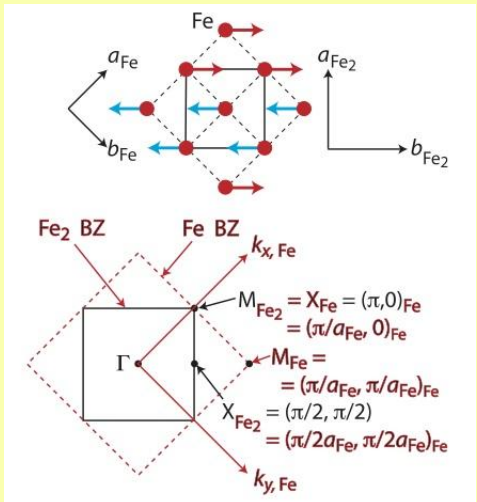


"3J" model ($J>0$ is AF coupling)

$$H = J_1 \Sigma \boldsymbol{S}_i \boldsymbol{S}_j + J_2 \Sigma \boldsymbol{S}_i S_k + J_3 \Sigma \boldsymbol{S}_i \boldsymbol{S}_m$$



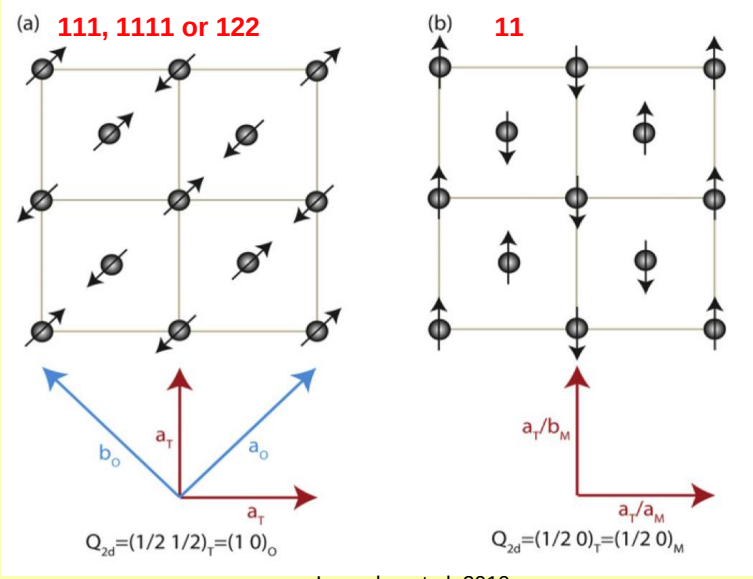




Magnetic Structures

by neutron scattering (also Mössbauer (^{57}Fe) and μSR)

Neutrons: done for 1111, 111 (on powders), 122 and 11, 245 (on single crystals)



Lumsden et al, 2010

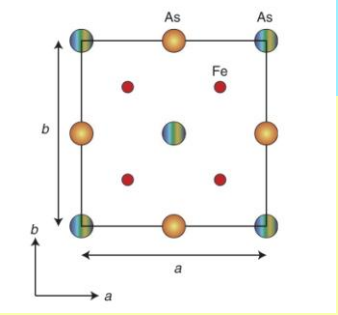
Compound	T_N(Fe)	Q_{magn}(tetra)	μ(Fe)	Ref.
LaOFeAs	137 K	(1/2 1/2 1/2)	0.36 μ_B	de la Cruz *et al*, Nature 2008
CeOFeAs	140 K	(1/2 1/2 0)	0.80 μ_B	Zhao *et al*, Nature Mat. 2008
PrOFeAs	136 K	(1/2 1/2 0)	0.35 μ_B	Kimber *et al*, PRB 2008
NdOFeAs	141 K	(1/2 1/2 1/2)	0.25 μ_B	Chen *et al*, PRB 2008
NdOFeAs	15 K	(1/2 1/2 0)	0.32 μ_B	Tian *et al*, PRB 2010
NaFeAs	37 K	(1/2 1/2 1/2)	0.09 μ_B	Li *et al*, PRB 2009
LiFeAs	no magnetic order observed			
$CaFe_2As_2$	173 K	(1/2 1/2 1)	0.80 μ_B	Goldman *et al*, PRB 2008
$SrFe_2As_2$	220 K	(1/2 1/2 1)	0.94 μ_B	Zhao *et al*, PRB 2008
$BaFe_2As_2$	143 K	(1/2 1/2 1)	0.87 μ_B	Huang *et al*, PRL 2008
$Fe_{1.068}Te$	67 K	(1/2 0 1/2)	2.25 μ_B	Li *et al*, PRB 2009
$Fe_{1.141}Te$	63 K	(0.38 0 1/2)	1.96 μ_B	Bao *et al*, PRB 2009
$A_2Fe_4Se_5$	470-560K	(2/5 1/5 1)	3.2-3.4 μ_B	Ye *et al*, PRL 2011

in-plane Fe-moments order characteristic to pnictides: 111, 1111 and 122 (left) and selenides: 11 (right), note out-of-plane moment direction in 245

Layers stacking along the *c*-axis can be F or AF adding a Q_z component equal to 0 or 1/2 (as in 11, 111, 1111 *P*-cells) or 1 (as in 122 or 245 bct *I*-cell)

Magnetic ordering of Rare-Earths, if present, happens at relatively low T and it does not change magnetic structure of Fe

Magnetic structures of the parent compounds survive at doping



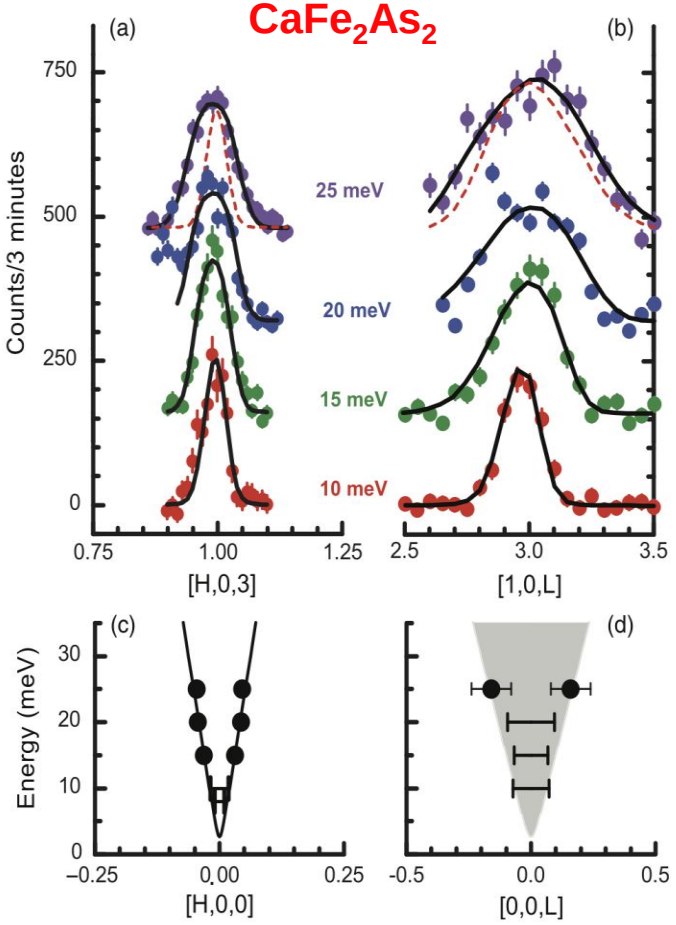
Magnetic Dynamics: parent compounds

122

as large single crystals for INS are available

Noticeable differences with respect to cuprates:
Parent compounds are magnetic metals,
Crystal structures are less 2D and
Magnetic coupling along c-axis is more important.

$CaFe_2As_2$



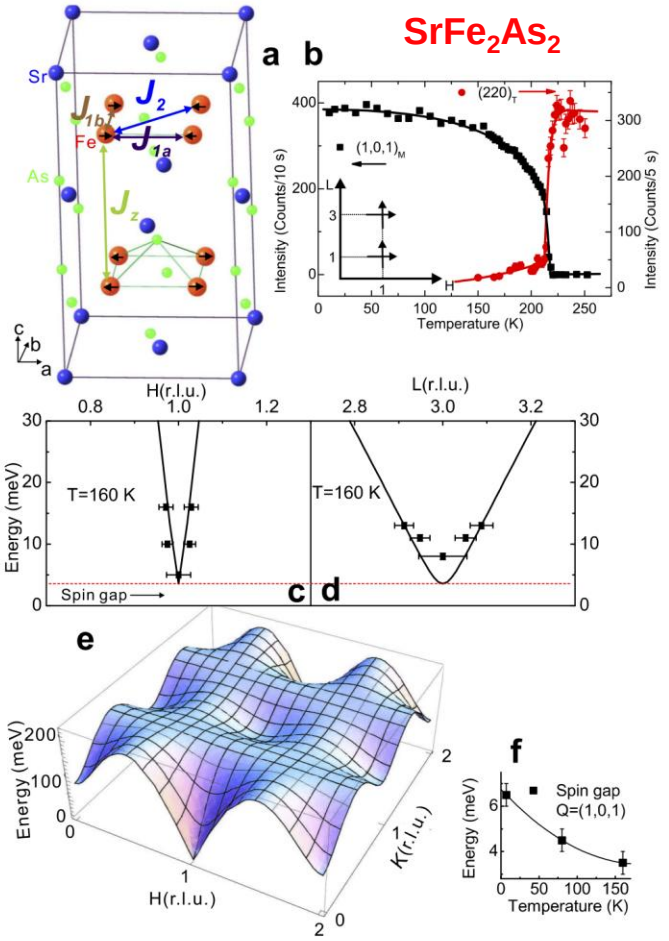
INS observations:

Magnon dispersion:
very steep ~200 meV
lower but similar to cuprates

Anisotropy:
in-plane and out of plane components are different
however, less pronounced than in 2D cuprates

Energy gap:
clearly present

$SrFe_2As_2$

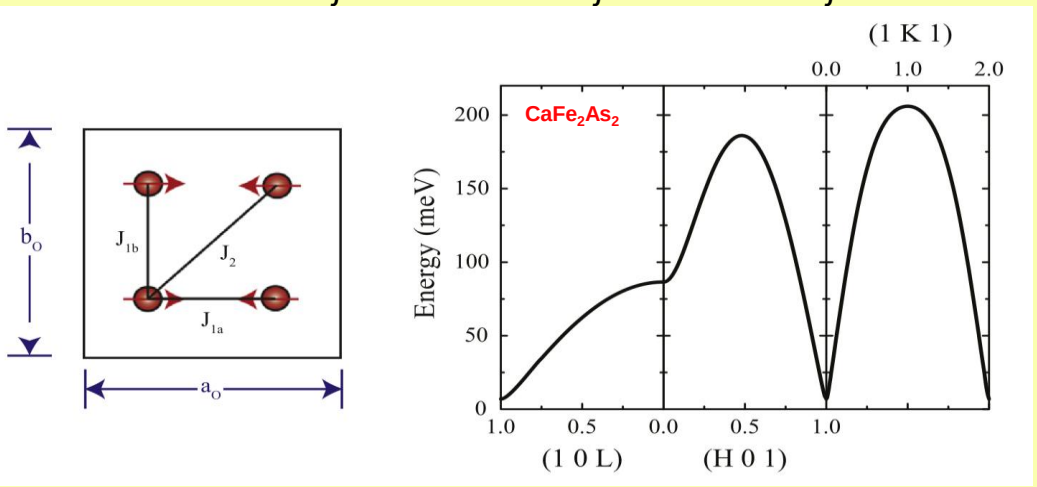


The presence of an energy gap is reported also on polycrystalline samples La-1111 and 11-$FeTe_{0.92}$
Magnetic signal emanating from the corresponding ordering wave vectors (1/2 1/2 0) and (1/2 0 1/2)

Magnetic Dynamics: parent compounds

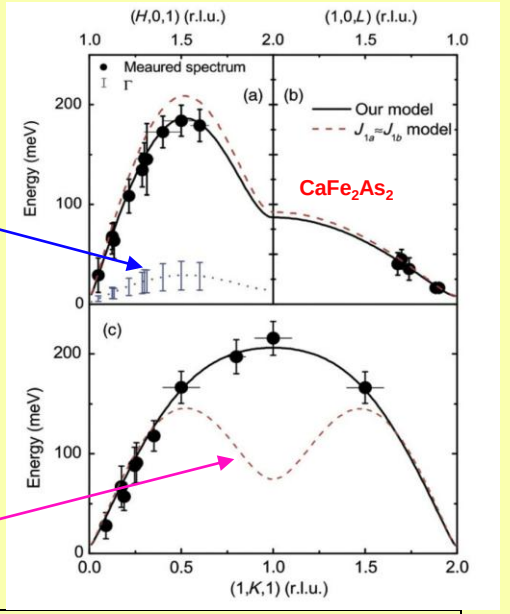
Local moment picture:

$$H = J_{1a}\Sigma \mathbf{S}_i\mathbf{S}_j + J_{1b}\Sigma \mathbf{S}_i S_j + J_2\Sigma \mathbf{S}_i\mathbf{S}_j + J_c\Sigma \mathbf{S}_i\mathbf{S}_j + D\Sigma(S_{xi})^2$$

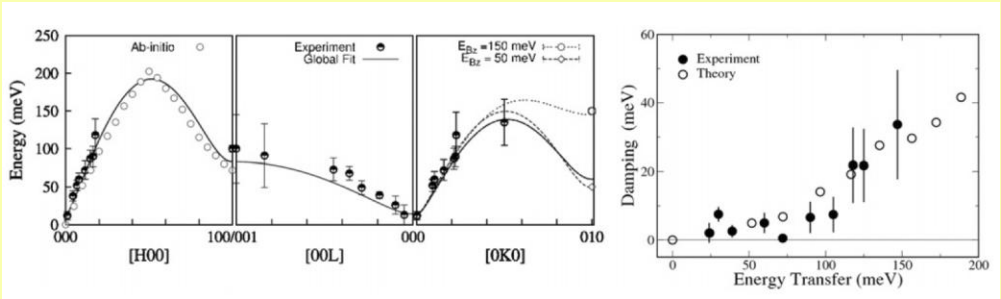


SW damping
Γ = 0.15 hω

$J_{1a} = J_{1b}$



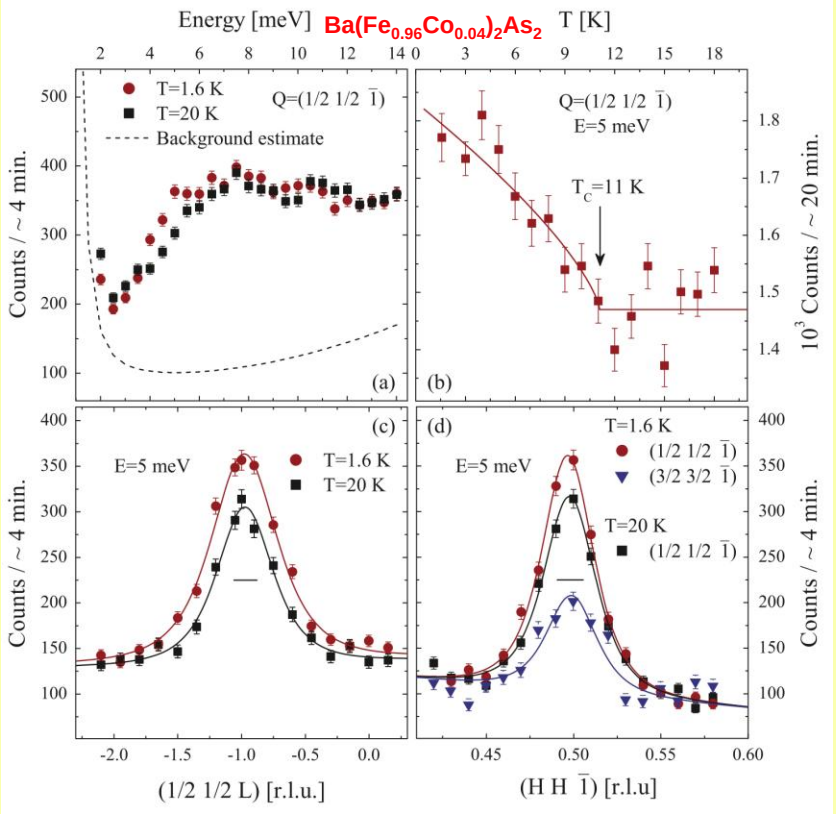
Compound	$S(J_{1a}+2J_2)$ meV	SJ_{1a} meV	SJ_{1b} meV	SJ_2 meV	SJ_c meV	Gap (SD) meV	$v_{//}$ meV*A	$v_{\perp}$ meV*A	$(v_{//})/(v_{\perp})$	Ref.
$CaFe_2As_2$	75				6.7	6.9 (0.07)	420	270	1.6	McQueeney et al, PRL 2008
$CaFe_2As_2$	93	31	13	31	4.5	- (0.06)	498	259	1.9	Diallo et al, PRL 2009
$CaFe_2As_2$	88	50	-6	19	5.3	0	516	243	2.1	Zhao *et al*, N.Phys. 2009
$SrFe_2As_2$	100				5	6.5 (0.18)	560	280	2.0	Zhao *et al*, PRL 2008
$BaFe_2As_2$	50				0.38 (0 at $T>T_N$)	9.8	280	57	4.9	Matan *et al*, PRB 2009

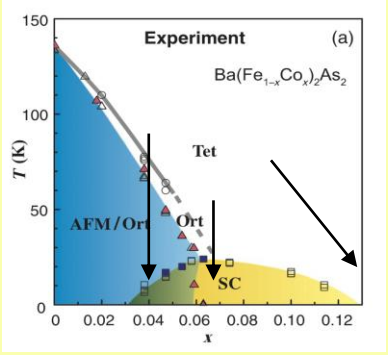


Landau damping rates from DFT (itinerant magnetism model) are consistent with the experimental observations

Itinerant or mixed local/itinerant are dominating over pure local moment picture

Magnetic Dynamics: doped compounds

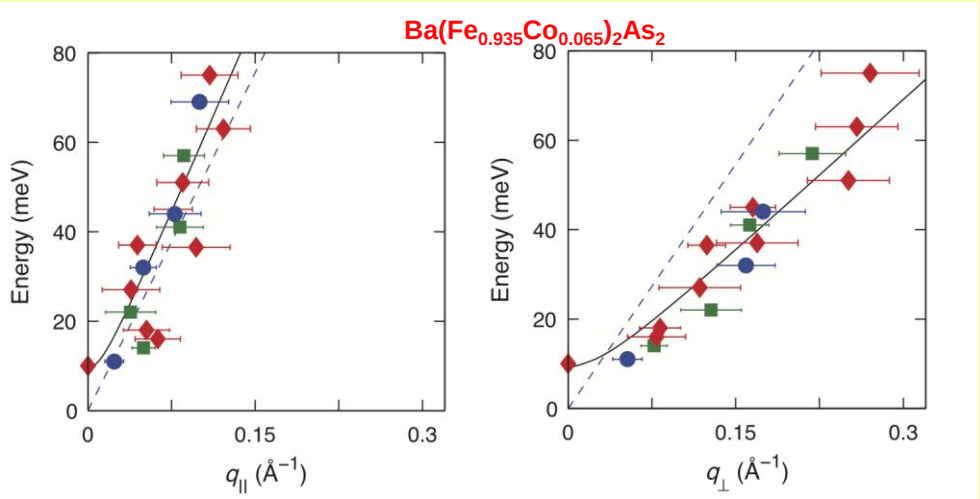




$Ba(Fe_{1-x}Co_x)_2As_2$
most studied by neutrons

Underdoped (x=0.04):
similar to x=0 apart from
lower T_N and the excitation frequencies
(the same anisotropy of ~4)
at Tc: an additional weak magnetic signal

Optimal doping (x=0.065-0.08):
strong enhancement of 2D character
anisotropy >100
asymmetry appears in the (a,b) plane
(Fe_2 layer)
at energies of ~10 meV
magnetic signal in SC state is enhanced



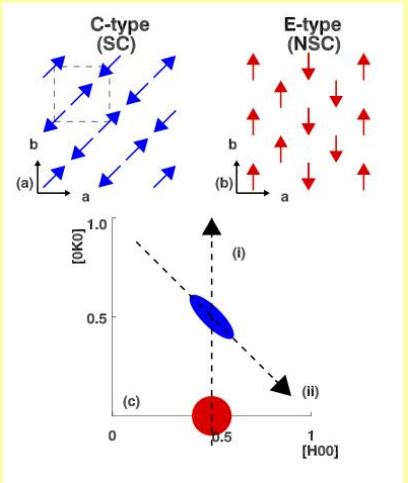
Overdoped:
magnetic signal decreases
$Ba(Fe_{1-x}Co_x)_2As_2$ and $La(O_{1-x}F_x)FeAs$ -
a signature of the vanishing hole pockets?
still to be investigated

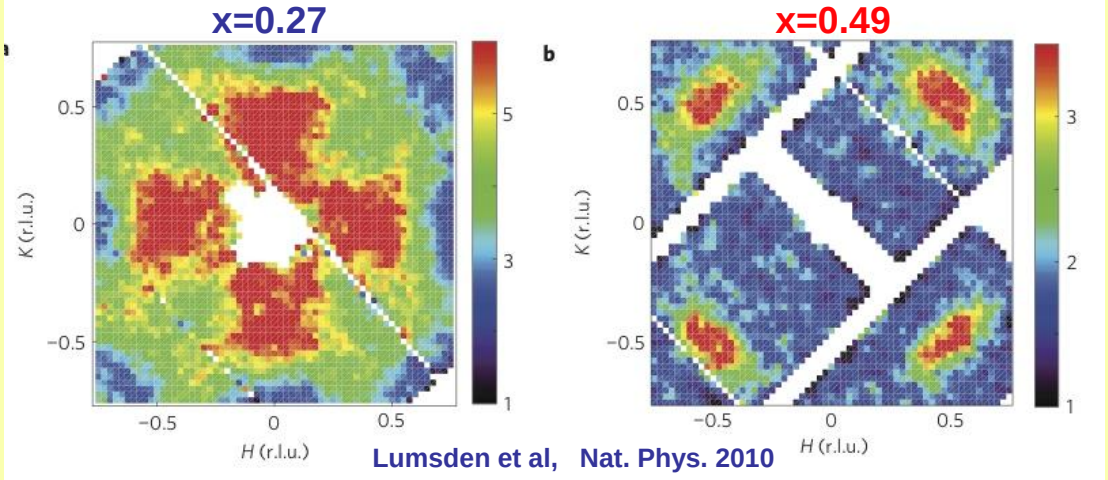
Magnetic Dynamics: Se-doped $Fe_{1+y}Te_{1-x}Se_x$

Change of the ordering vector: in the Se-doped (SC) compounds the excitations grow up at **(1/2 1/2 0)**, same as in all 111, 1111, 122 and different from (1/2 0 1/2) in $Fe_{1+y}Te$

Role of excess Fe: suppress SC, induce (1/2 0 1/2) order (long or short-range)

while for dynamics, and possibly SC, the common vector (1/2 1/2 0) is requested





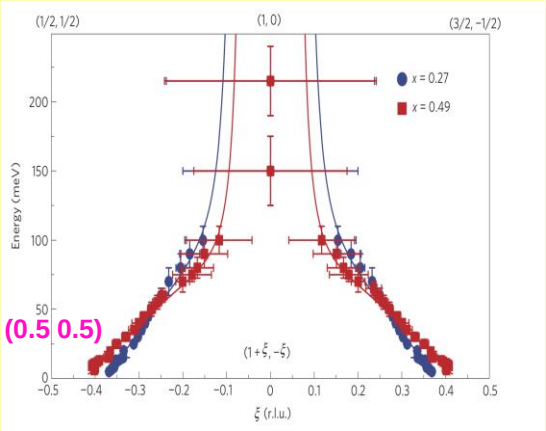
Lumsden et al, Nat. Phys. 2010

However both signals have the same origin as the same T-dependence (Chi, PRL 2011)

Incommensurate positions of the origin of SW dispersion.

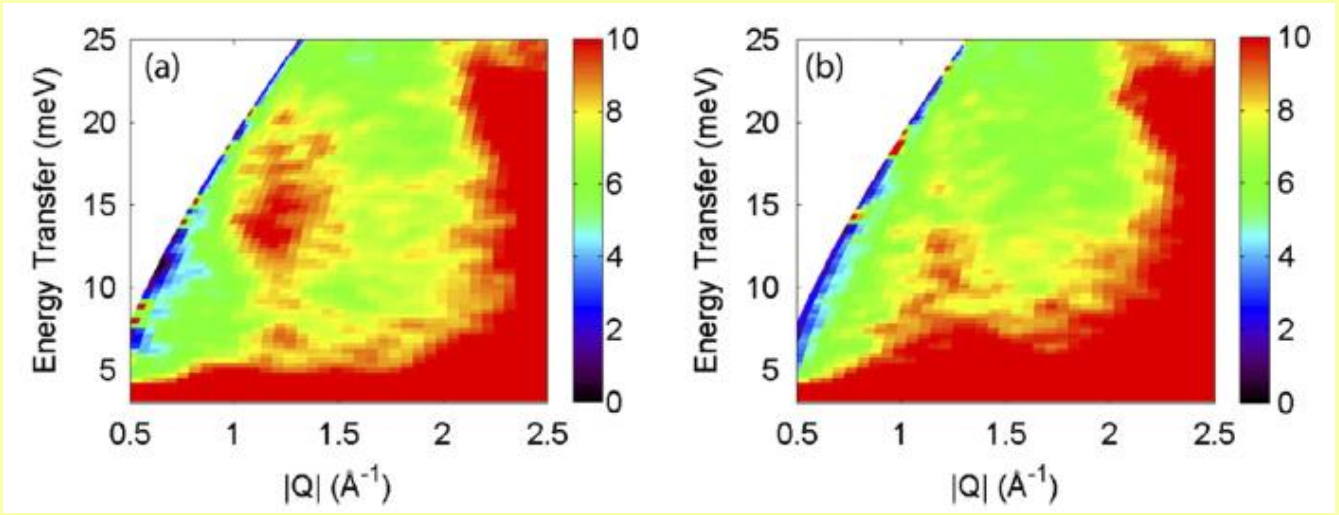
Incommensurability is doping-dependent: minimal at optimal doping (apparently near x=0.5)

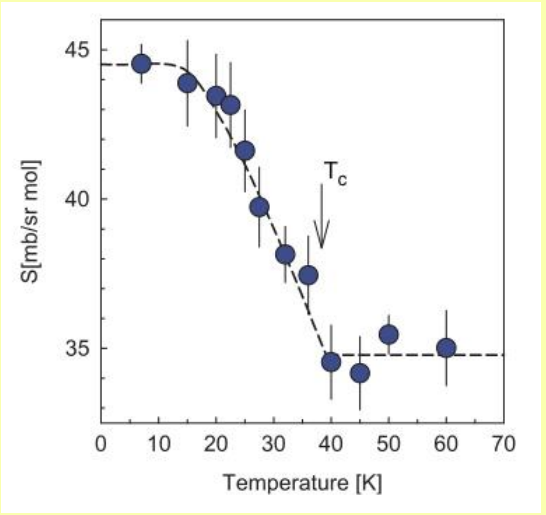
No Spin Wave cones: signature of itinerant interactions.



Magnetic Dynamics: Spin-Resonance excitation and SC

When entering the SC state, in particular close to optimal doping, a resonance excitation appears at Q_{2D}=(1/2 1/2), as in cuprates.





$Ba_{0.6}K_{0.4}Fe_2As_2$: Christianson et al, Nature 2008

1111: La(OF)FeAs (polycrystal);

122: (BaK)Fe_2As_2 (poly), Ba(FeCo)$_2As_2$, Ba(FeNi)$_2As_2$ (mono)

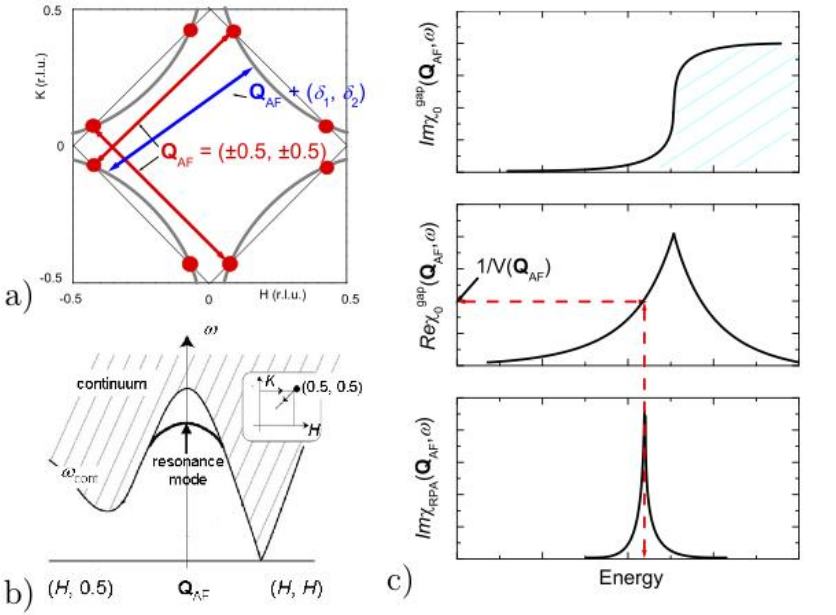
111: NaFeAs: not found so far (mono); LiFeAs possible? (poly)

11: Fe(TeSe) (mono)

245: $K_2Fe_4Se_5$ and $Rb_2Fe_4Se_5$ (mono)

Magnetic Dynamics: Spin-Resonance excitation and SC

Significance of the magnetic resonance with respect to superconductivity and, in particular, to the gap symmetry is outlined in itinerant models.



$$\chi_{\mathrm{RPA}}(\mathbf{q},\omega) = \frac{\chi_0^{\mathrm{gap}}(\mathbf{q},\omega)}{1 - V(\mathbf{q})\chi_0^{\mathrm{gap}}(\mathbf{q},\omega)}$$

$$Im\,\chi_{\mathrm{RPA}}(\mathbf{q},\omega) = \frac{Im\,\chi_0^{\mathrm{gap}}(\mathbf{q},\omega)}{[1 - V(\mathbf{q})\,Re\,\chi_0^{\mathrm{gap}}(\mathbf{q},\omega)]^2 + [V(\mathbf{q})\,Im\,\chi_0^{\mathrm{gap}}(\mathbf{q},\omega)]^2.}$$

Intensity ~ $[1- (\Delta_k/E_k)(\Delta_{k+Q}/E_{k+Q})]$

$\Delta(\mathbf{k}) = -\Delta(\mathbf{k}+\mathbf{Q}_{AF})$

$$\Delta_p = -\sum_{p'} \frac{V(p-p')\Delta_{p'}}{2E_{p'}}$$

Δ=const only possible with V<0 (phonons)

if V>0, the gap Δ(p) should change sign.

If only one band is present, then Δ should have nodes.

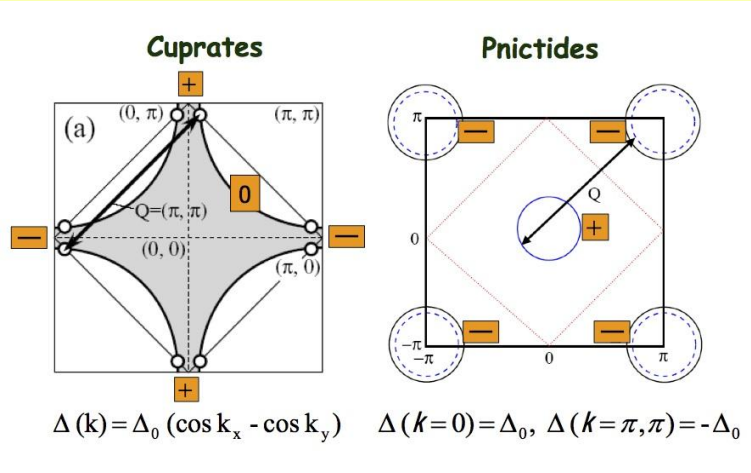
In a multi-band case Δ may change sign at different bands being const in absolute value.

Magnetic Dynamics: Spin-Resonance excitation and SC

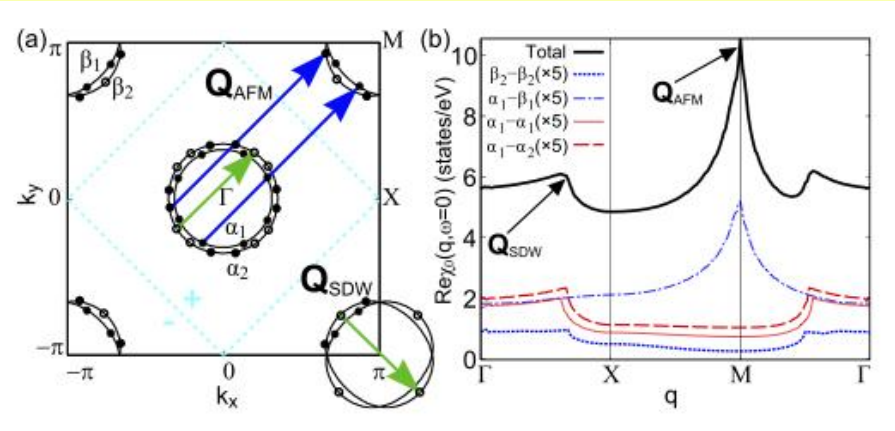
Origin: inter-band transitions between electron and hole pockets of the Fermi surface

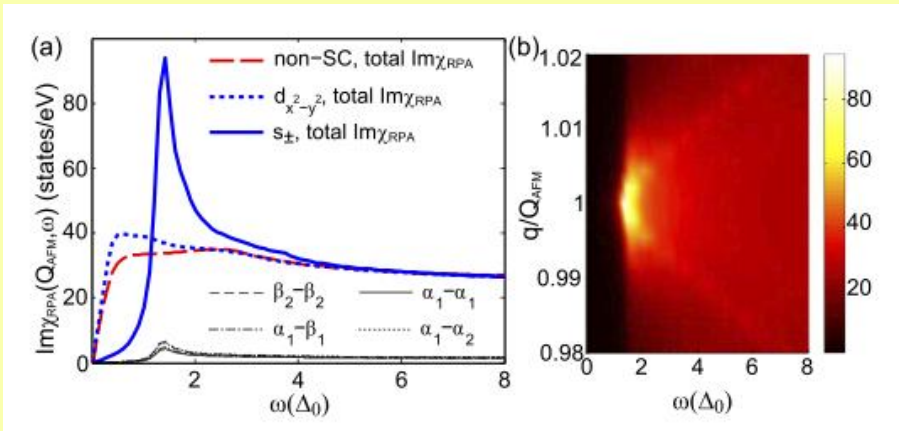
The pairing is **singlet** (Knight shift experiments) as in cuprates.

(in cuprates with a d-wave gap changing sign and a single band)



ARPES in Fe-based: **no nodes** in the gap function $\Delta(\mathbf{k})$: **s-wave** symmetry A way out - changing sign of $\Delta(\mathbf{k}) = -\Delta(\mathbf{k}+\mathbf{Q}_{AF})$ in different parts of BZ: $\mathbf{S}^{\pm}$



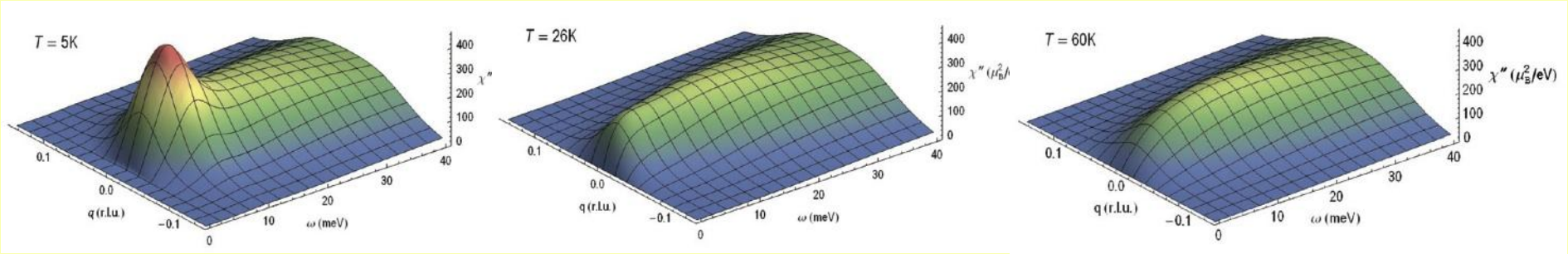


Korshunov & Eremin, PRB 2008

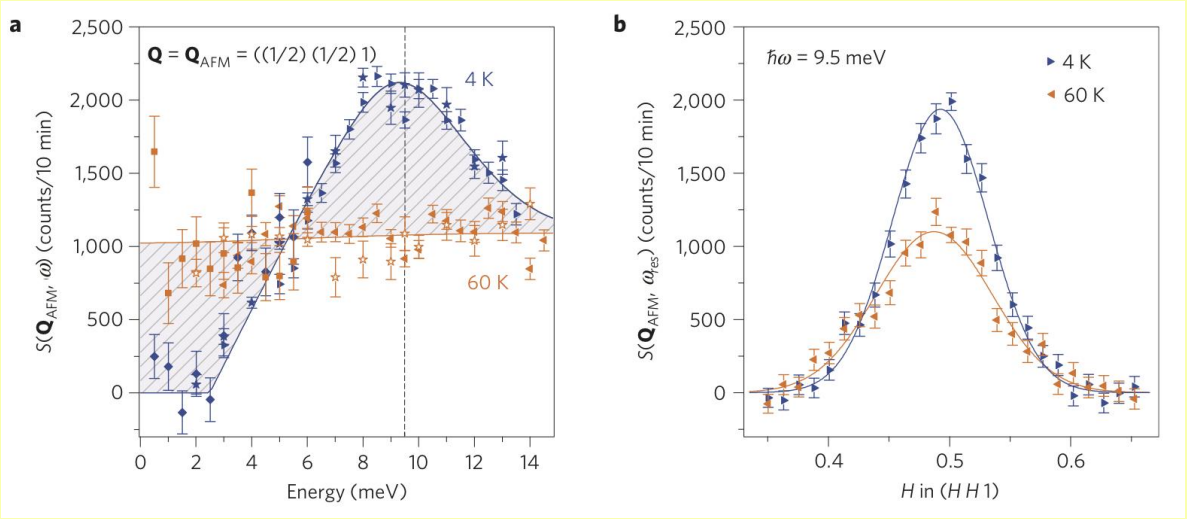
Magnetic Dynamics: Spin-Resonance excitation and SC

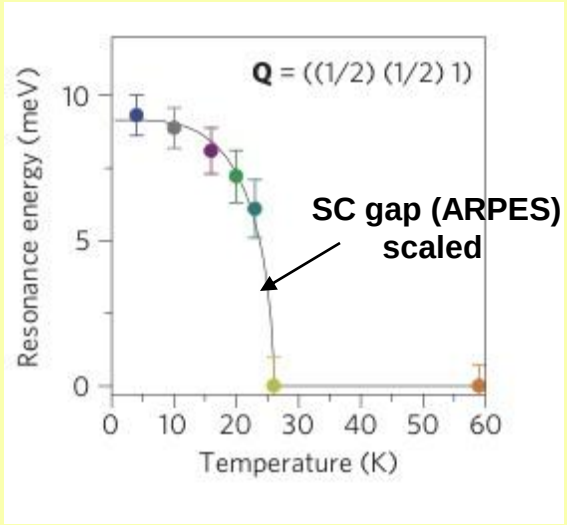
Temperature dependence

122: $Ba(FeCo)_2As_2$ $Q_{resonance} = Q_{ordering} = (1/2\ 1/2\ 1)$



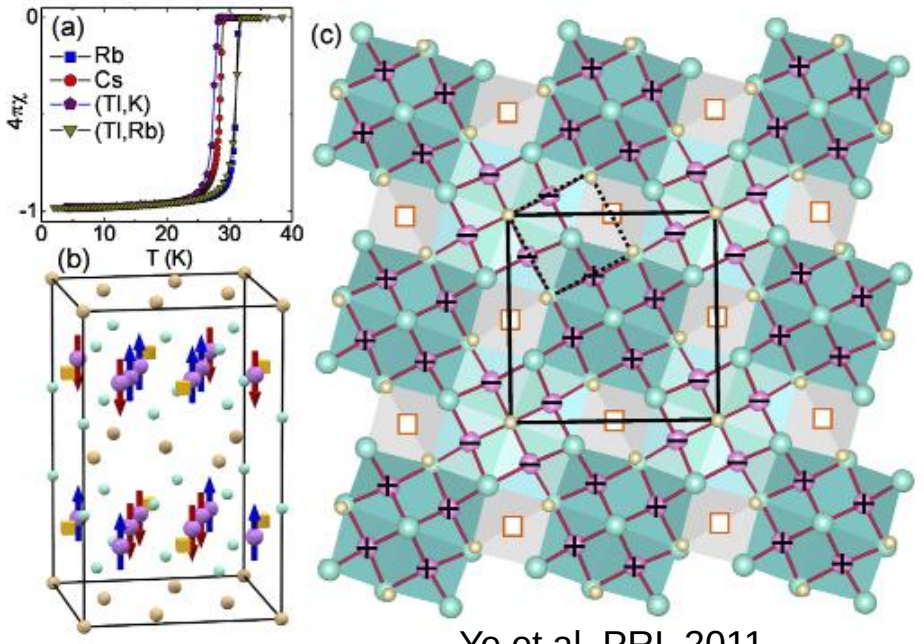
The resonance is built from the excitations already existing at T>Tc



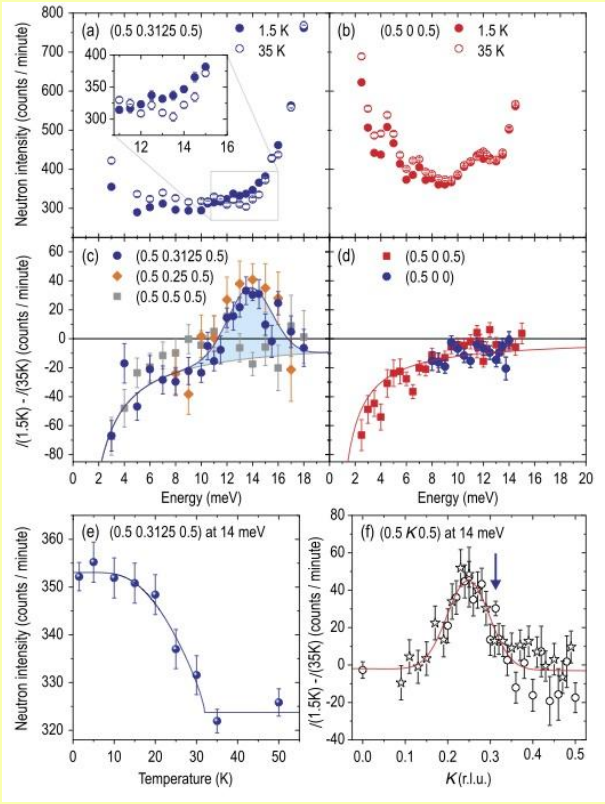


$Ba(Fe_{1.925}Co_{0.075})_2As_2$: Inosov et al, Nature Physics 2010

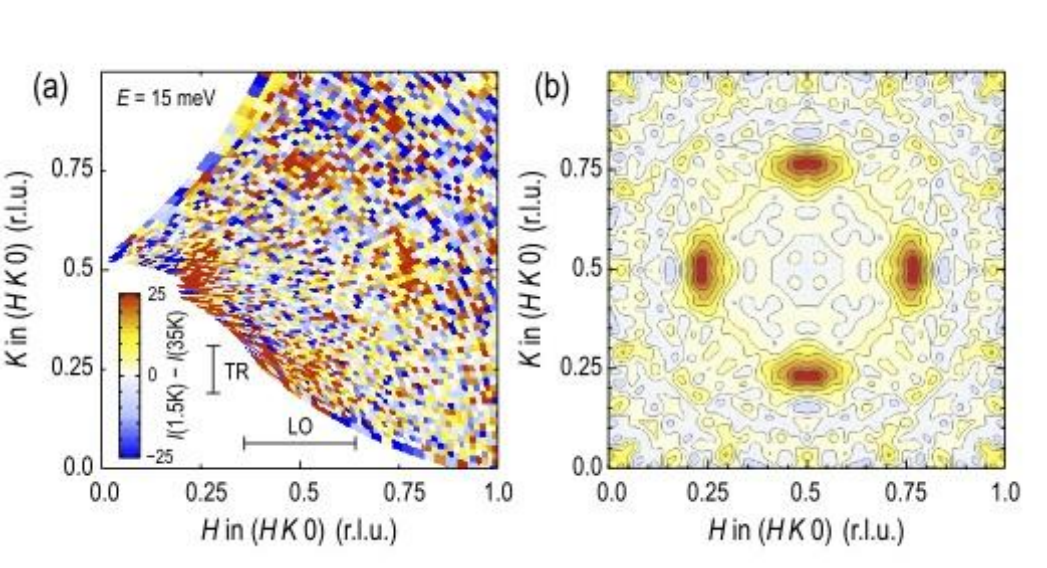
Resonance in $Rb_2Fe_4Se_5$

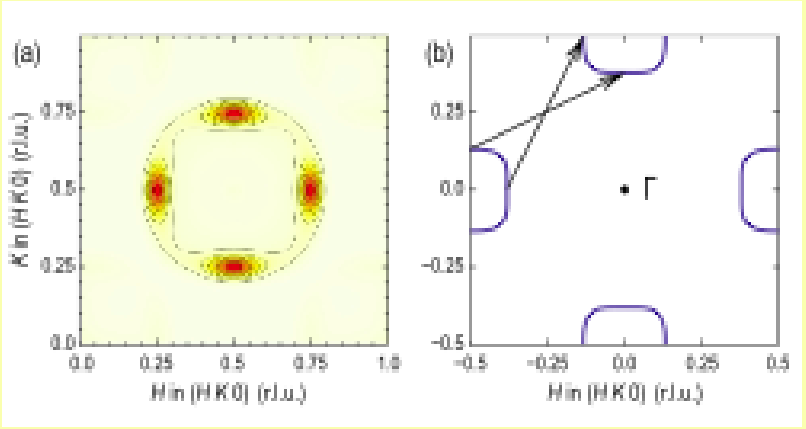


Ye et al, PRL 2011



Park et al, PRL 2011

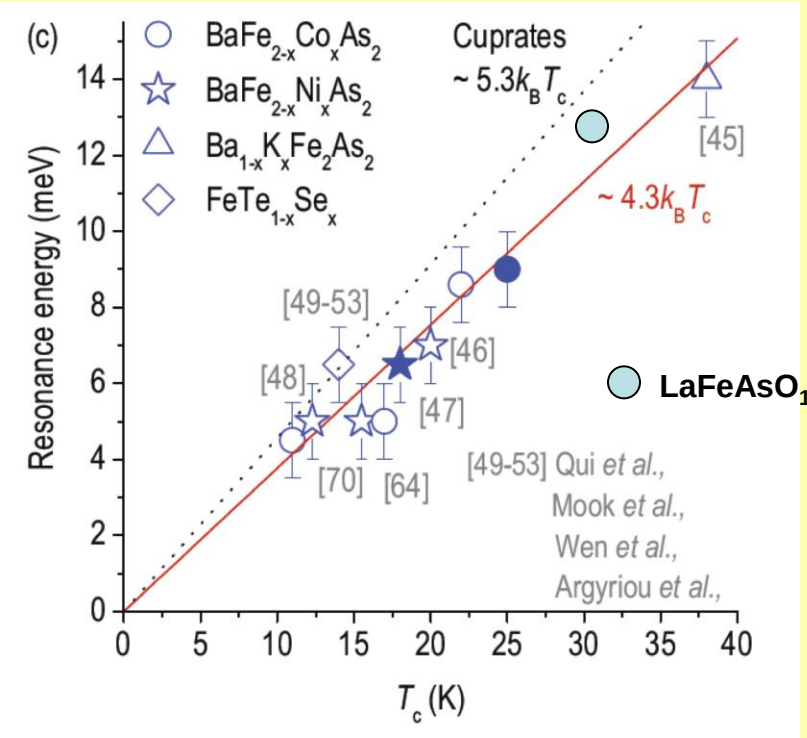




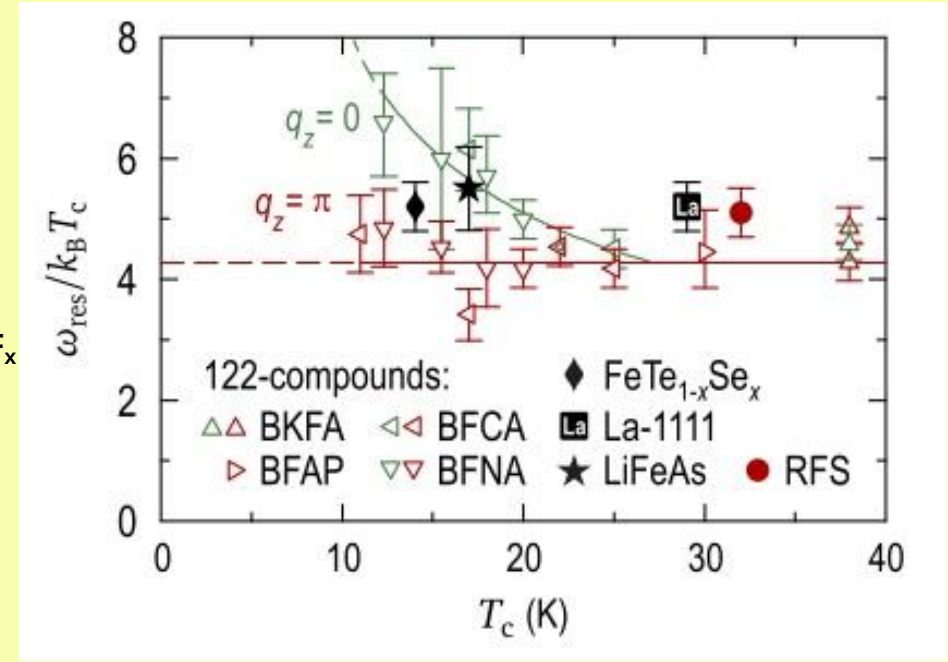
Friemel et al, submitted PRL 2012

Magnetic Dynamics: Spin-Resonance excitation and SC

Resonance *vs* Tc



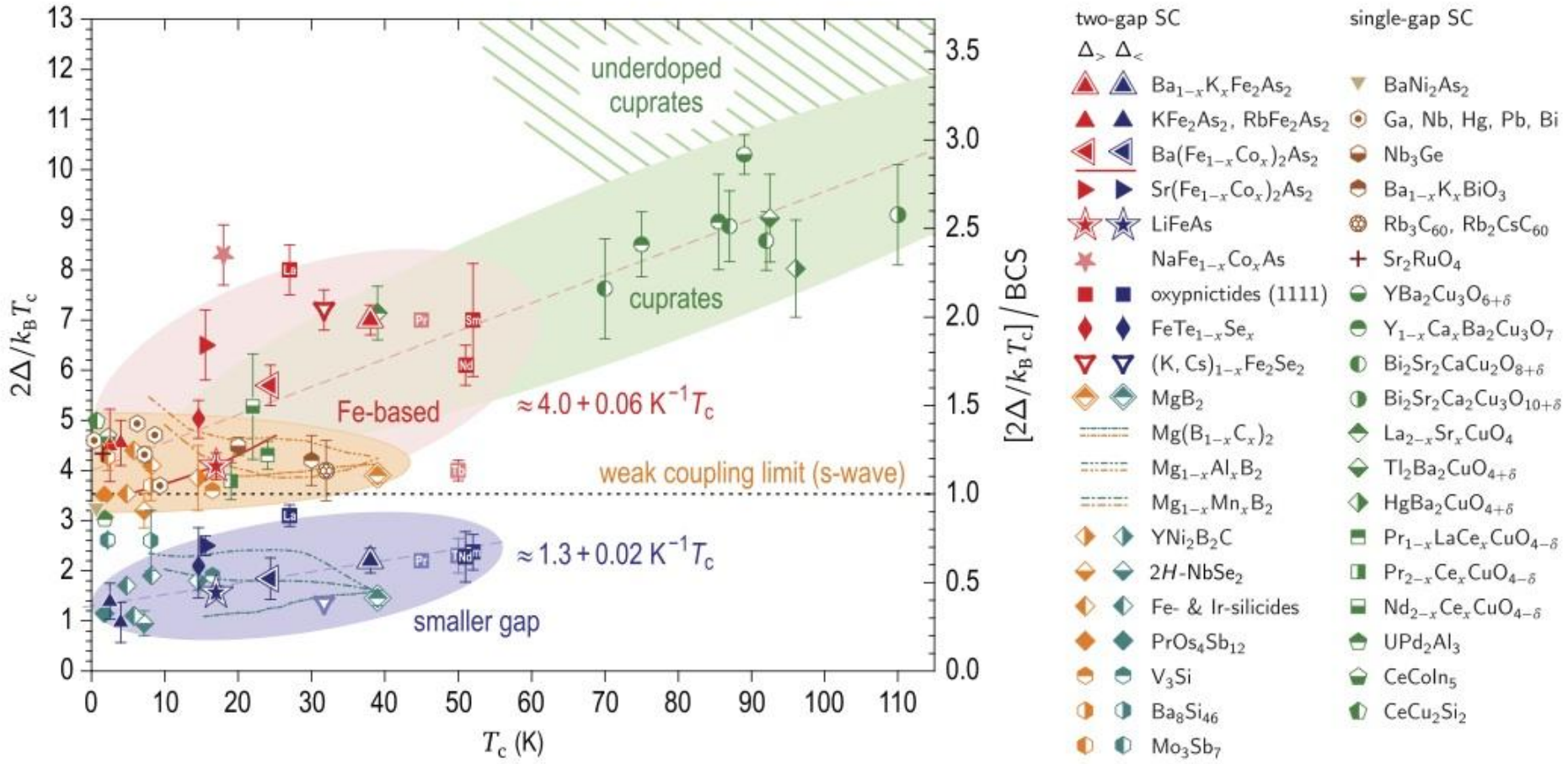
$LaFeAsO_{1-x}F_x$



$Ba(Fe_{1.925}Co_{0.075})_2As_2$: Park et al, PRB 2010

The resonance in pnictides is clearly linked to the superconducting state, similarly to cuprates.

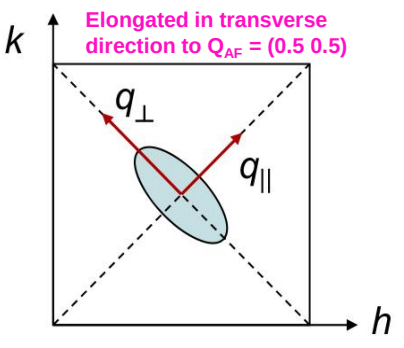
Interaction strength

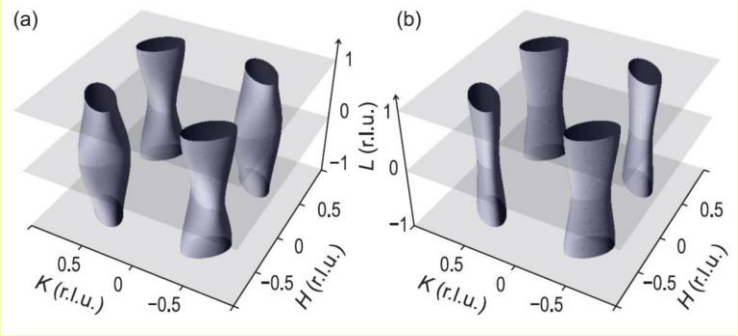


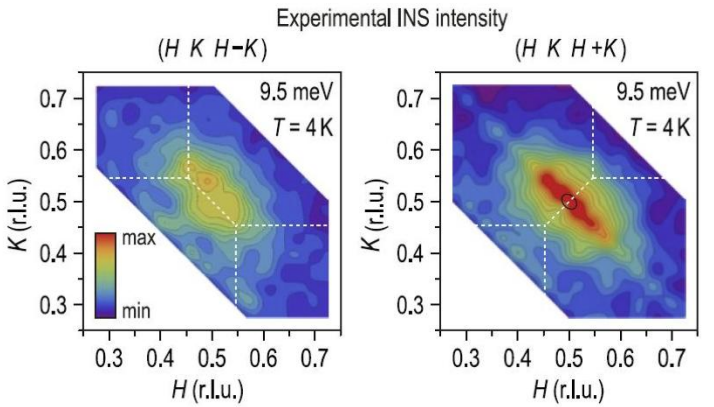
Iron-based are less similar inside their “family” than the other groups

Magnetic Dynamics: Spin-Resonance excitation and SC

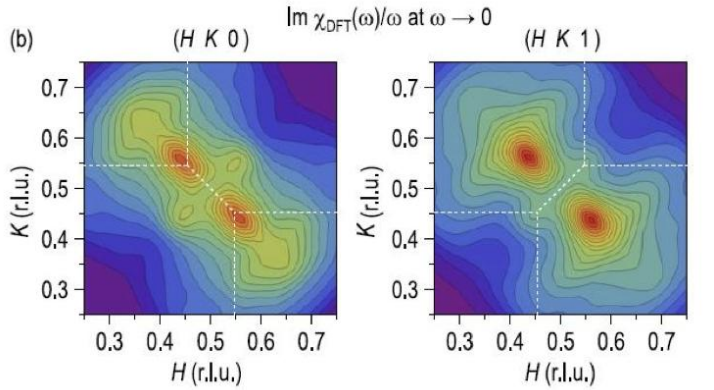
Magnetic response: crystal lattice symmetry or Fe only?

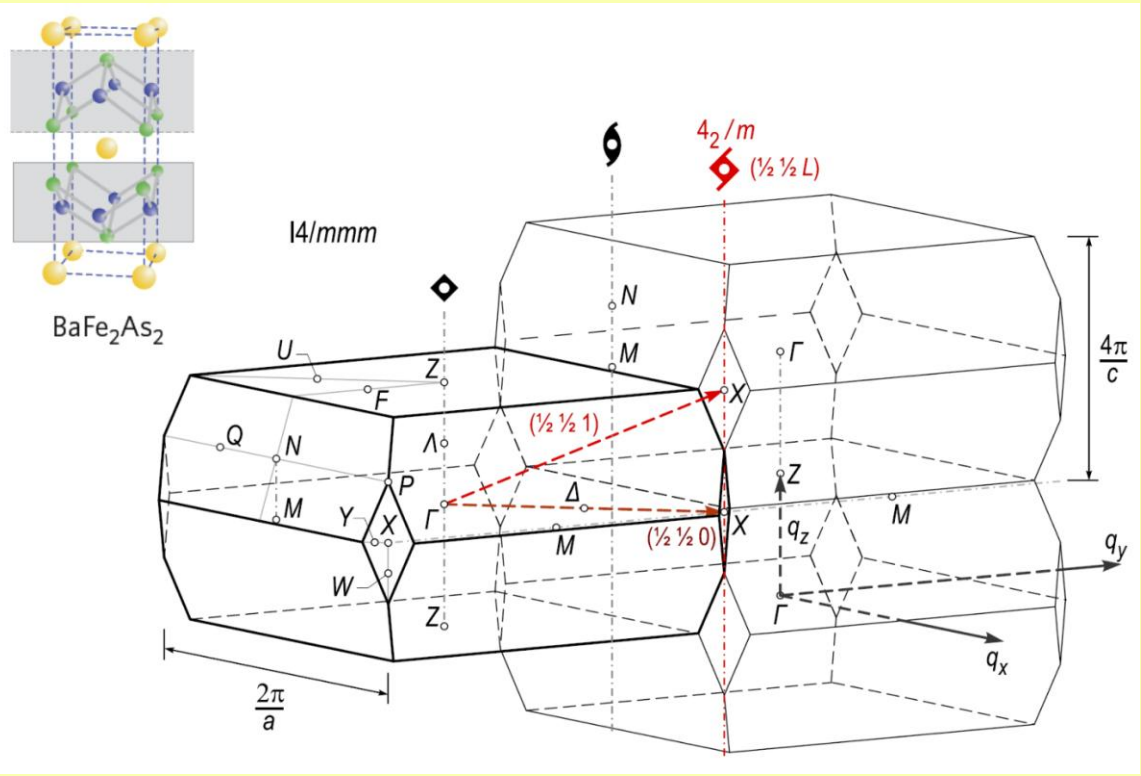






J. T. Park, D. S. Inosov *et al.*, PRB **82**, 134503 (2010)





Most important is the Fe-Fe arrangement

Conclusions

Itinerant picture seems to be more adequate for both doped and undoped compounds

Doping evolution of magnetic response needs more experimental research

S^{+-} symmetry gives a good basis for description of magnetic excitations in normal and superconducting phases

Fe-based superconductors exhibit more rich combination of properties than cuprates.

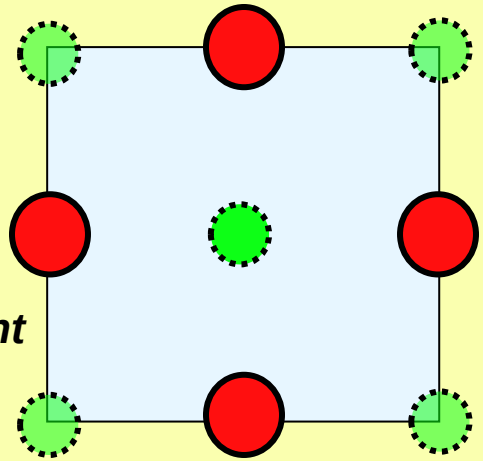
New interesting physics while perspectives of higher Tc than in cuprates are hardly brilliant

Magnetically active planes in Pnictides and Cuprates

Fe_2As_2: Fe-Fe = 2.70 A

Metal iron (bcc):
a = 2.866 A
Fe-Fe = 2.480 A

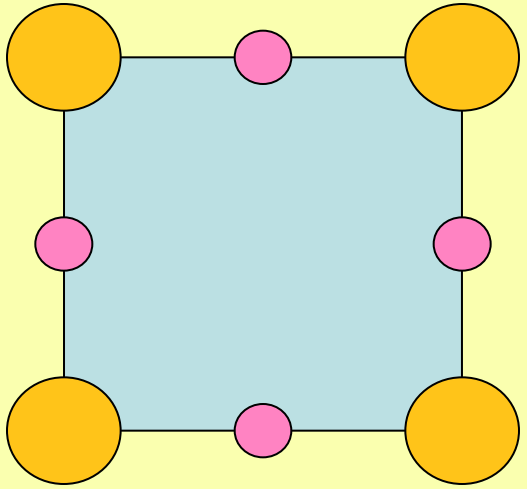
Magnetic moment
Fe = 2.2 μ_B



Fe (z = 0)
As (z≠0)

Spin Fe^{2+} (d^6): S=2
Magnetic moment Fe = ?

CuO_2: Cu-Cu = 3.85 A

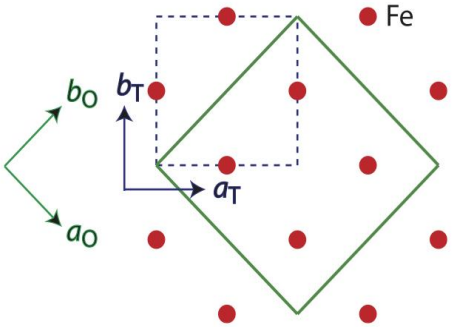


Cu (z = 0)
O (z = 0)

Spin Cu: S=1/2
Magnetic moment Cu ~ 0.5 μ_B

T: 1 x Fe_2

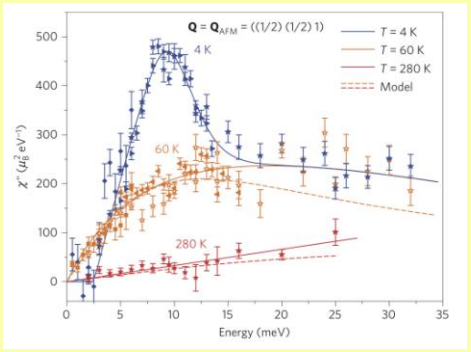
O: 2 x Fe_2

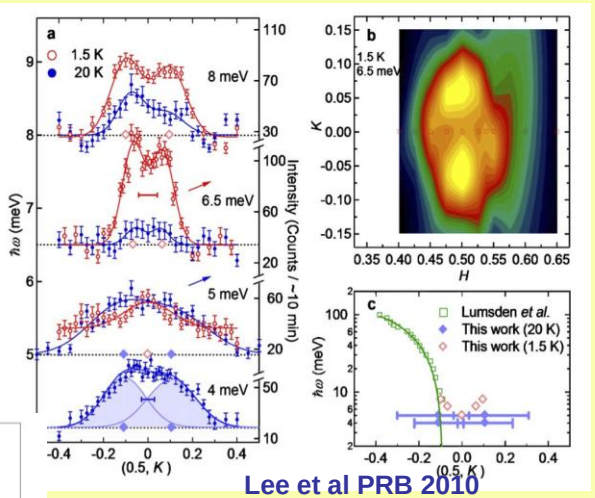


Lowering lattice symmetry prior to magnetic ordering: change of orientation of the crystallographic axes by 45 degrees
(there are also variants with no rotation)
Often keep "tetra" notation for Q-vectors

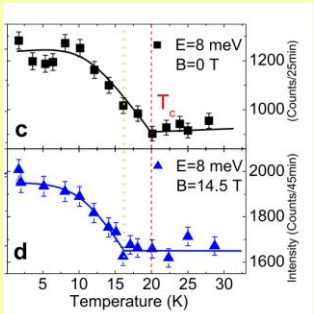
No Spin Wave cones: signature of itinerant interactions.
Incommensurability is doping-dependent: minimal at optimal doping (apparently near x=0.5)

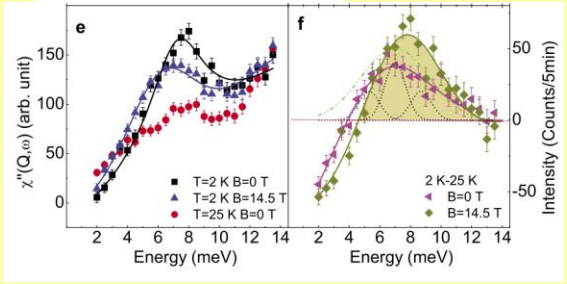
Magnetic field dependence



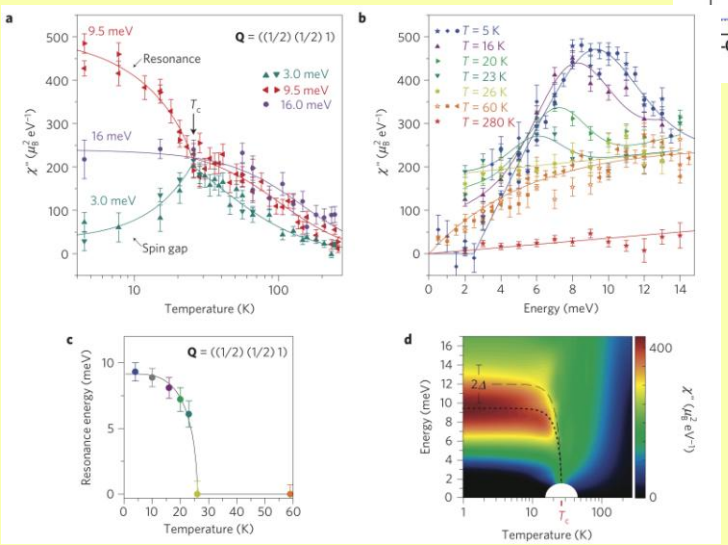


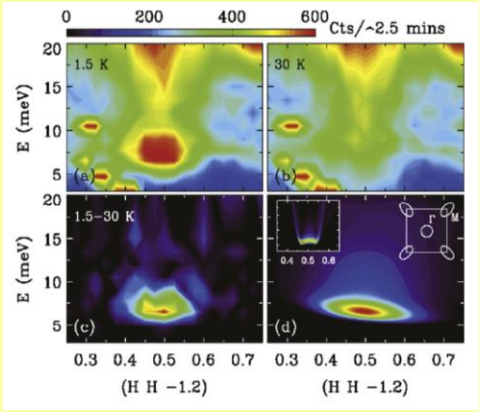
Lee et al PRB 2010





$Ba(Fe_{1.9}Nio_{0.1})_2As_2$: Zhao et al, PRB 2010





$FeTe_{0.6}Se_{0.4}$: Qiu et al, PRL 2009

Local *vs* Itinerant

If "very" local then for Fe^{2+} (d^6) S=2 and with g=2 $\mu(Fe) = 4\ \mu_B$

Moment reduction: itinerant effects are strong, magnetic ordering is a SDW transition, Fermi surface nesting effects are dominating. Stability of the commensurate Q_{magn} under doping (not-too-heavily doped La-1111 and Ba-122) is supported by band calculations (Yaresko *et al*, PRB 2009). But χ increase on heating?

Local moment frustrations and fluctuations in ordered phases is ruled out by low magnetic moment in the *paramagnetic* state of Ca-122 (Diallo *et al* PRB 2010).

It looks that the itinerant picture wins over the localized thus setting the pnictides at the lower-correlated side of the cuprates