Naval Research Laboratory

Superconductivity, magnetism and spin fluctuations in Fe-based superconductors

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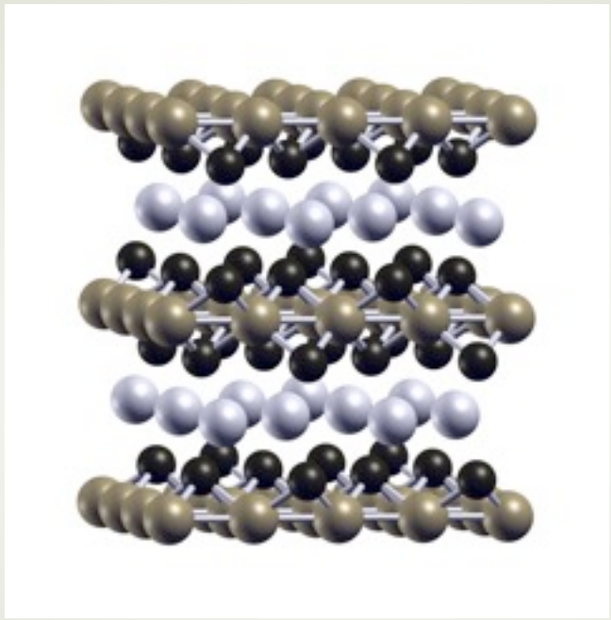


Density Functional Theory

Fundamental Theorems of DFT:

(both due to Hohenberg & Kohn)

i) The ground state is a unique *functional* of the density.

$$E[\rho(\vec{r})] = F[\rho(\vec{r})] + \int V_{ext}(\vec{r})\rho(\vec{r})$$

universal specific to system

ii) The density that minimizes the total energy is the ground state density.

$$\frac{\delta E[\rho(\vec{r})]}{\delta \rho(\vec{r})} = 0$$

Given the external potential as an input, the ground state density of the system can be obtained by minimizing the total energy functional.

Density Functional Theory

$$E[\rho(\vec{r})] = E_{ii}[\rho(\vec{r})] + \underbrace{E_{ei}[\rho(\vec{r})]}_{\int V_{Ext}(\vec{r})\rho(\vec{r})d\vec{r}} + \underbrace{E_{ee}[\rho(\vec{r})]}_{E_{Kin}[\rho(\vec{r})]+E_{Pot}[\rho(\vec{r})]}$$

constant

$$\rho(\vec{r}) = \sum_i \psi_i^*(\vec{r})\psi_i(\vec{r})$$

Kohn-Sham approximation

$$E_{Kin}[\rho(\vec{r})] = \nabla^2\psi(\vec{r}) + \quad ?$$

$$E_{Pot}[\rho(\vec{r})] = E_H[\rho(\vec{r})] + \underbrace{E_{EX}[\rho(\vec{r})]}_{?} + \underbrace{E_{Corr}[\rho(\vec{r})]}_{?}$$

$$\int \frac{\rho(\vec{r})\rho(\vec{r}')}{|\vec{r}-\vec{r}'|}d\vec{r}\,d\vec{r}'$$

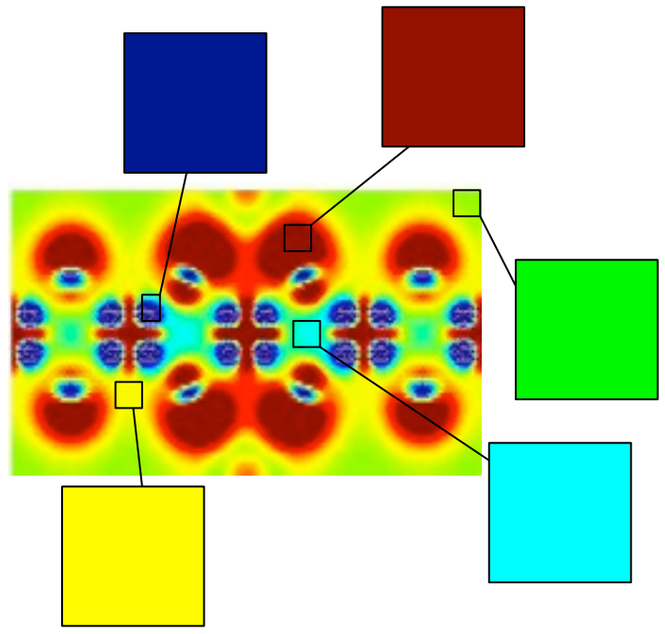
$$E[\rho(\vec{r})] = \int V_{Ext}(\vec{r})\rho(\vec{r})d\vec{r} + \nabla^2\psi(\vec{r}) + \int \frac{\rho(\vec{r})\rho(\vec{r}')}{|\vec{r}-\vec{r}'|}d\vec{r}\,d\vec{r} + \boxed{E_{XC}[\rho(\vec{r})]}$$

Density Functional Theory

LOCAL DENSITY APPROXIMATION (LDA)

$$E_{XC}[\rho(\vec{r})] = \int \rho(\vec{r})\varepsilon_{XC}(\rho)d\vec{r}$$

The XC potential at every point is taken to be that of a homogenous electron gas with a uniform density corresponding to the density of the point in question.



Other (better?) XC potentials:

GGA (Generalized Gradient Approximation)
WDA (Weighted Density Approximation)
B3LYP (Becke's 3rd Lee-Yang-Parr)
HSE06 (Heyd-Scuseria-Ernzerhof)

DFT in practice

Self-Consistent Formulation:

Guess an input density: $\rho_0(\vec{r})$

Write out the Hamiltonian: $H[\rho_0(\vec{r})]$

Solve for the wavefunctions: $\psi_i(\vec{r})$

Create output density: $\rho'(\vec{r}) = \sum \psi_i^* \psi_i$

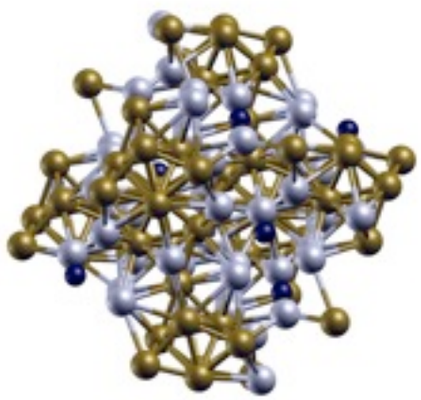
repeat

mix old and new ρ

Compare: $\int (\rho_0(\vec{r}) - \rho'(\vec{r}))d\vec{r}$

Large?

Small? → DONE!



Constituent Atoms
Approximate Structure

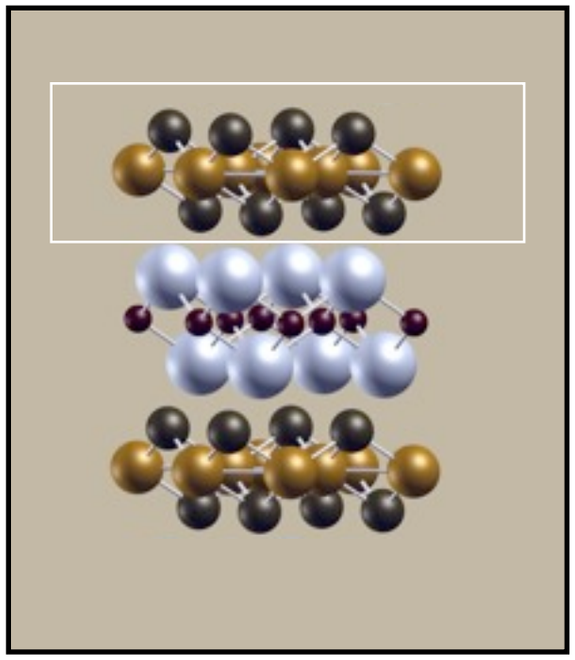
DFT

Total Energy
Magnetization
Forces
Sincle particle energy levels
(band structure)

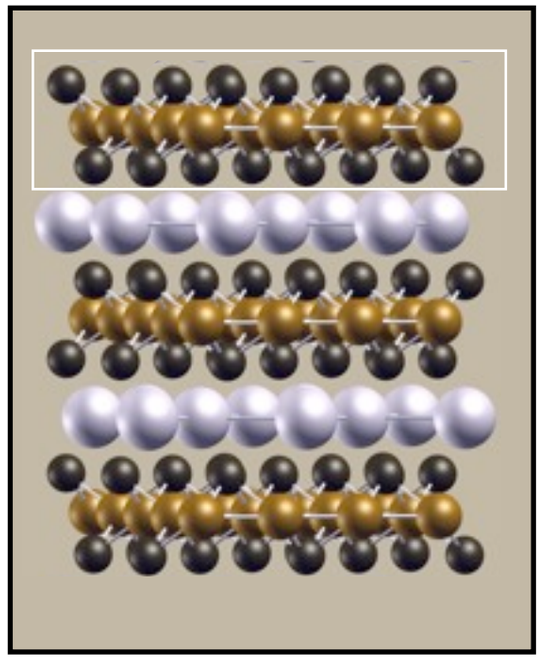
Fe-based Superconductors

Parent compounds:

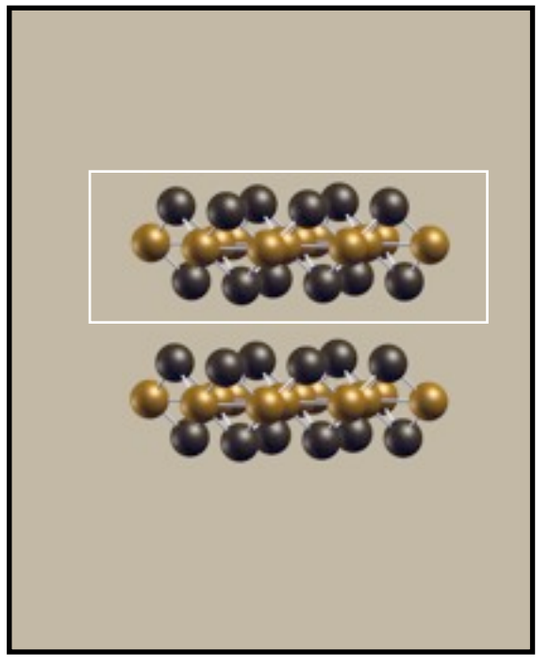
1111 ($LaOFeAs$)

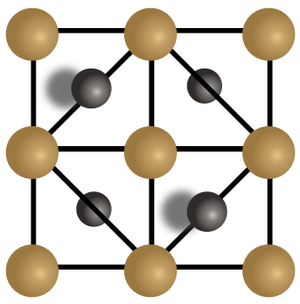


122 ($BaFe_2As_2$)



11 ($FeTe$)





Also:
LiFeAs, SrFeAsF
(111)

Critical Temperatures

22K $LaO_{0.9}F_{0.1}FeAs$

27K $FeSe$

38K $Ba_{0.6}K_{0.4}Fe_2As_2$

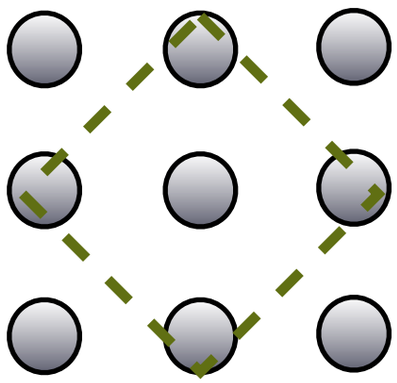
55K $SmFeAsO_{0.9}F_{0.1}$

56K $Sr_{0.5}Sm_{0.5}FeAsF$

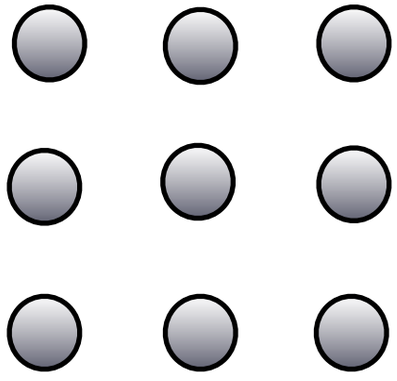
Two phase transitions

I) Structural Transition

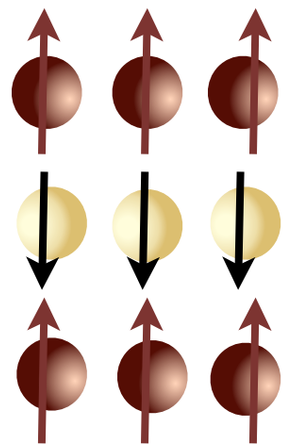
122/1111

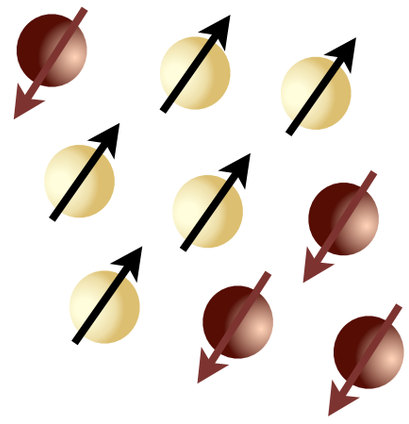


FeTe



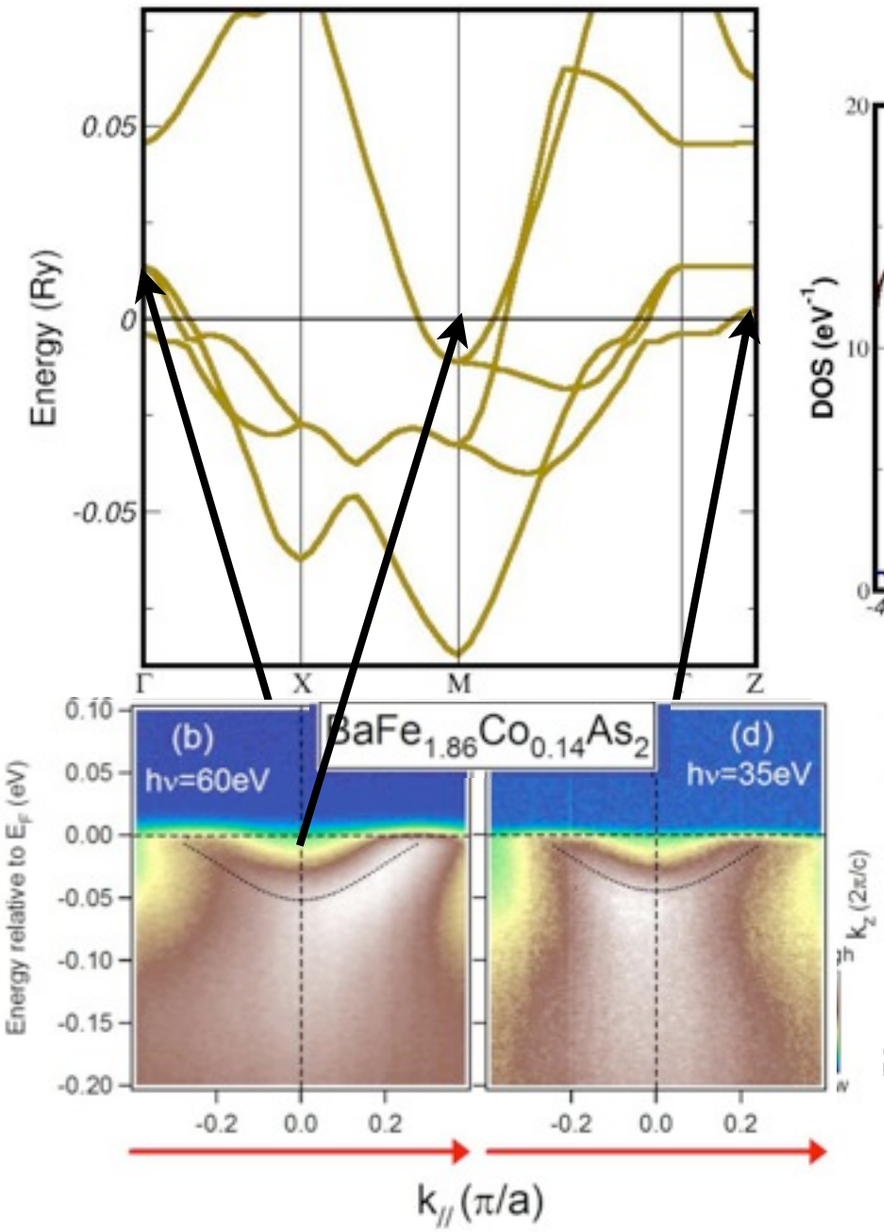
II) Magnetic Transition



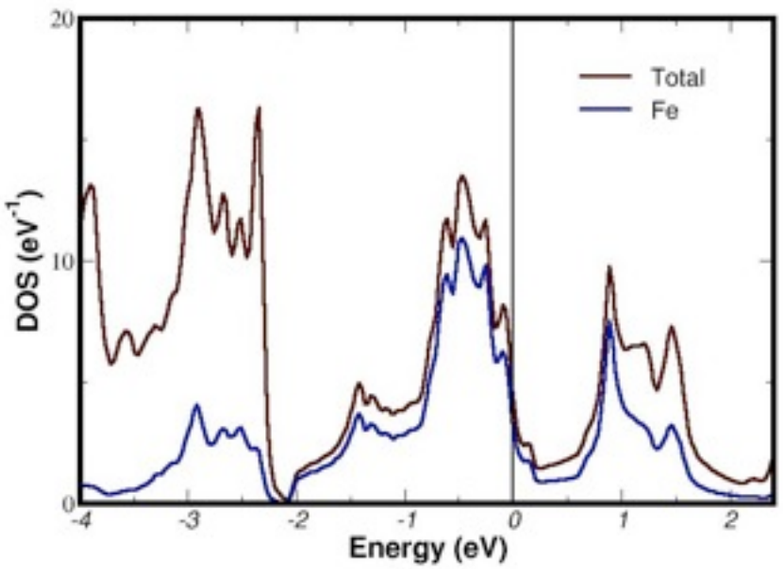


Transitions are simultaneous for FeTe and 122's, but structural transition is first in 1111's

Non-magnetic electronic structure



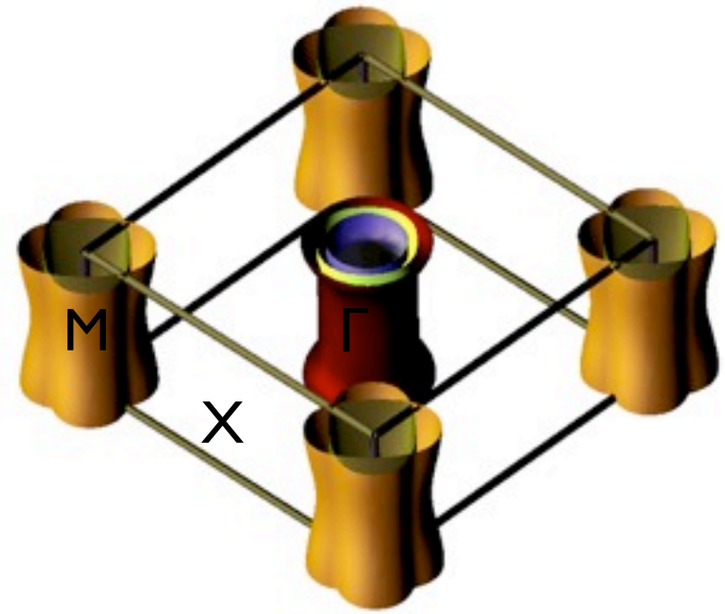
W. Malaeb *et al.* J. Phys. Soc. Jpn. **78**, 123706 (2009)



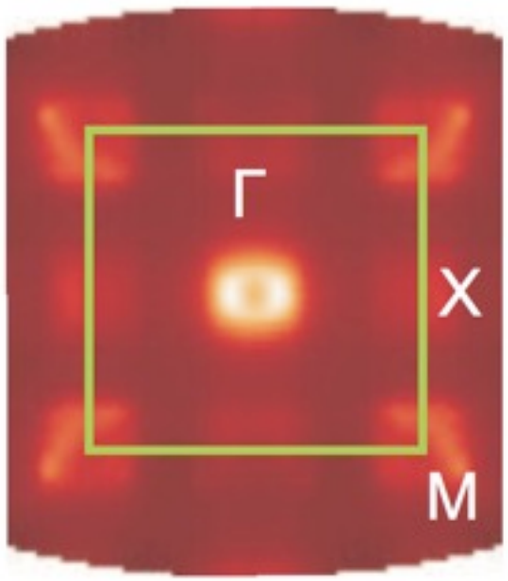
FSs (1 hole, 1

antly Fe *d*

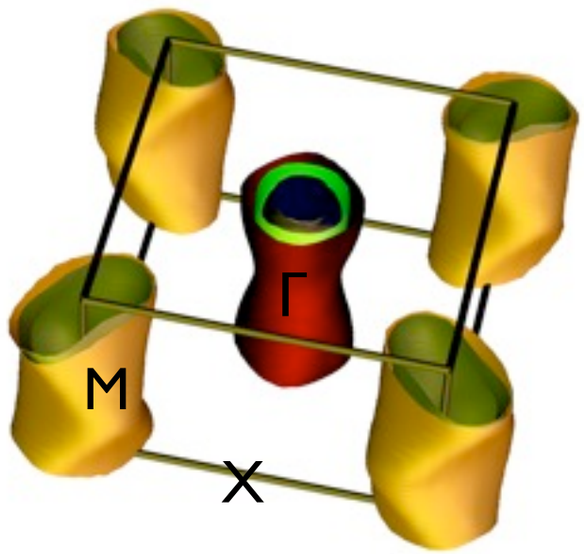
LaFeAsO



Y. Zia *et al.* PRL **103**, 037002 (2009)



$BaFe_2As_2$

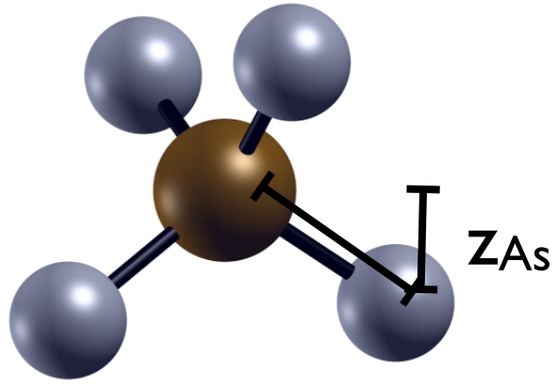


For superconducting compounds, electronic structure matches experiment well

Non-magnetic physical structure

- Relaxation of non-magnetic system (even in GGA) results in a vastly underestimated Fe-As distance

	non-magnetic	spin-polarized	experiment
Fe-As	2.32 Å	2.41 Å	2.40 Å
$\|\|_{As}$	1.97 Å	1.99 Å	1.98 Å
z_{As}	1.22 Å	1.35 Å	1.36 Å



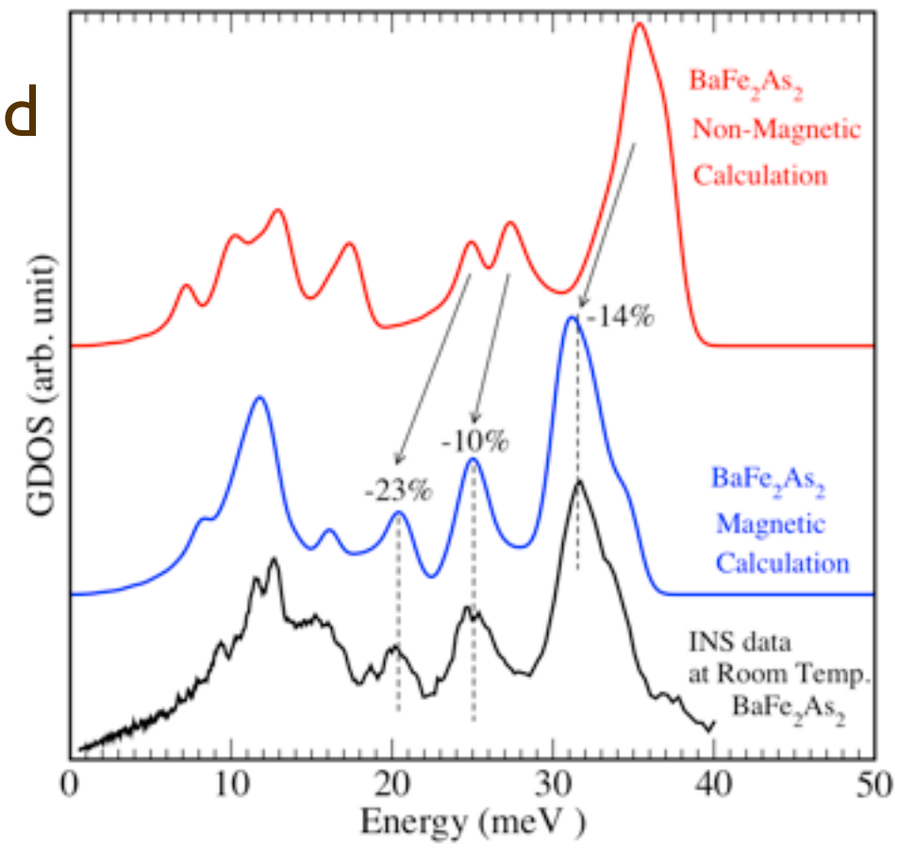
Spin-polarized relaxation is correct to within .8%

- Non-magnetic phonon spectrum is strongly shifted from experiment

Spin-polarized spectrum is vastly improved

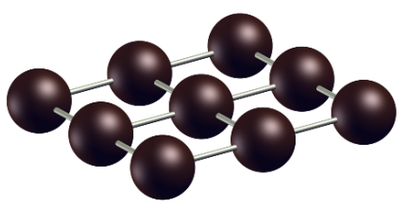
T.Yildirim arXiv: 0902.3462

Z.P.Yin *et al.* PRL **101**, 047001 (2008)

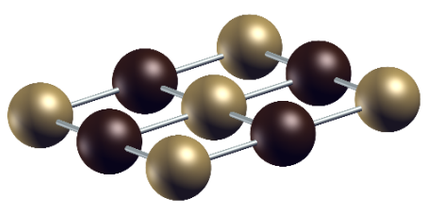


Structural distortion requires magnetism

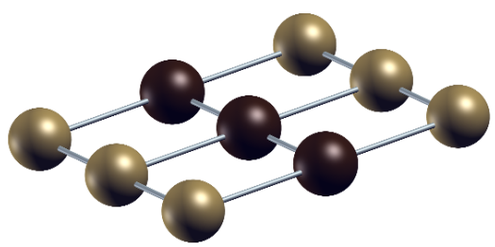
- Structural distortion not reproduced in non-magnetic case (or *wrong* magnetic case)



Non-magnetic



Checkerboard



Stripe

Spin-polarized calculations reproduce (predict!) distortion to within .6% of observed values

Calculations indicate that magnetism is a condition for distortion, not a result of distortion

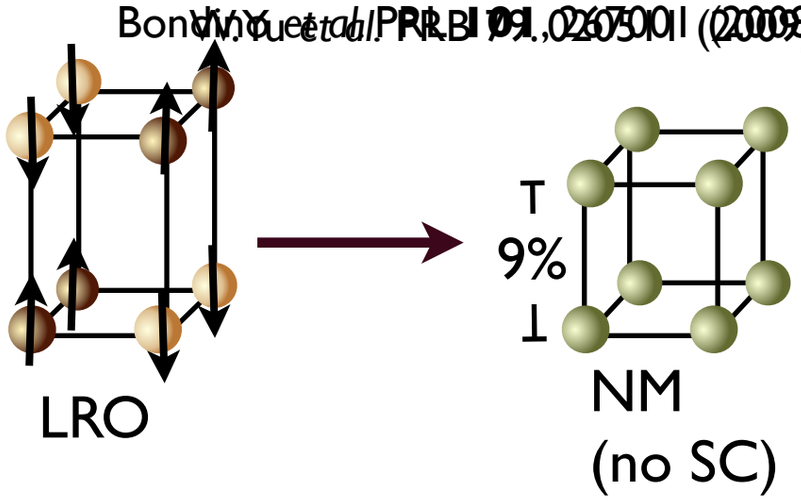
DFT vastly overstimates local magnetic moment → fluctuations are present

Conclusion: compounds are magnetic locally, though not ordered, even above the structural transition temperature

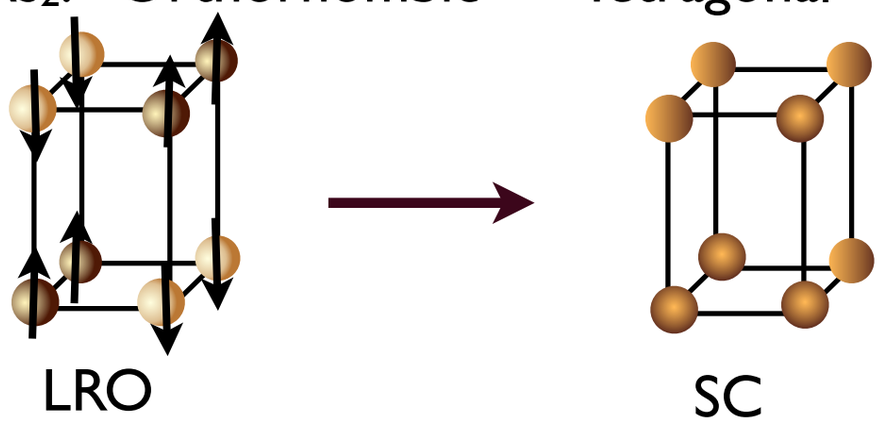
Magnetism exists below critical temperature

Where magnetism has definitively been killed, so has superconductivity
Signature of spin polarization in doped samples from core-level spectroscopy

$CaFe_2As_2$: Orthorhombic → Collapsed Tetragonal



$BaFe_2As_2$: Orthorhombic → Tetragonal



Alireza *et al.*, J. Phys.: Condens. Matter **21**, 012208 (2009).

Long range magnetic order is detrimental to superconductivity

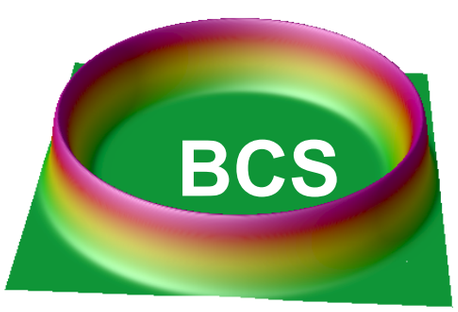
Spin fluctuations provide the pairing mechanism

Magnetism, but not LRO, is a defining characteristic of Fe-based SC compounds.

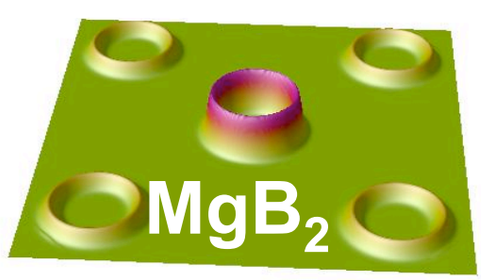
Spin fluctuations, superconductivity and fermiology

When phonons mediate SC, phonons connect FS pieces with the same gap sign.

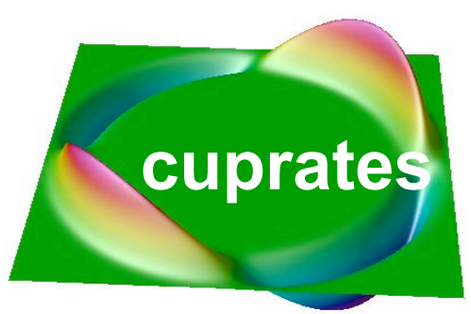
When spin fluctuations mediate SC, the magnon wave vector must connect FS pieces that are oppositely gapped.



s-wave

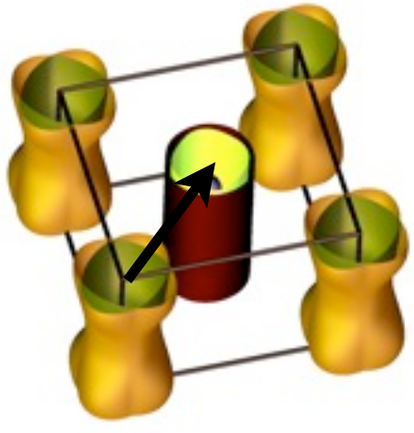


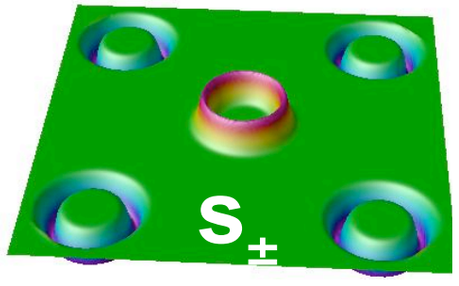
multiple s-wave



d-wave

In the Fe-based superconductors, the spin fluctuations are generated by FS nesting:





multiple s-wave, with changing sign

↓

S+/-

The particular nesting FS topology is essential for this kind of SC

Magnetism is the key

To understand the structure, LRO, and superconductivity and we must understand the magnetism.

- How does magnetic state arise?
- How is it suppressed by doping?
- How is it suppressed by pressure?

Nature of the magnetic interaction

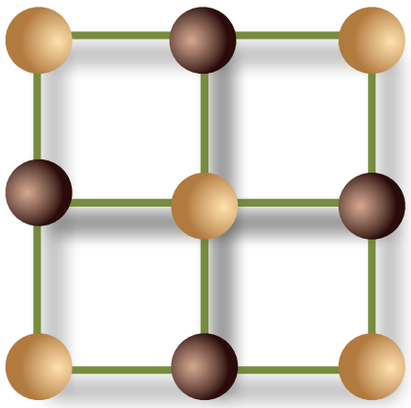
- Superexchange?
- Nesting-driven itinerant magnetism?

(neither!)

What does DFT get right/wrong?

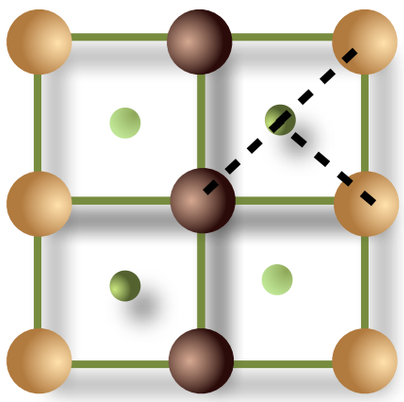
Energetics in the superexchange picture

Checkerboard



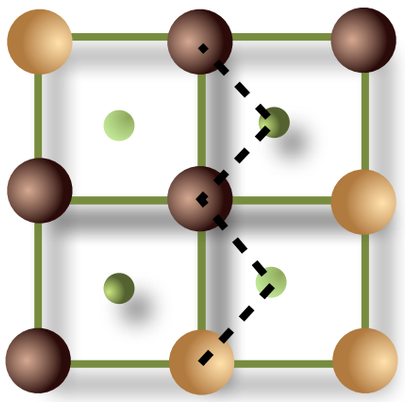
$-2J_1+2J_2+2J_3$

Stripe (SDW)



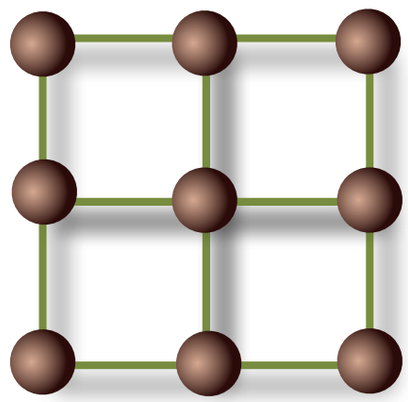
$-2J_2+2J_3$

Doublestripe



$-2J_3$

FM



$2J_1+2J_2+2J_3$

	$BaFe_2As_2$	FeTe
FM	unstable	-139
CB	-16	-118
Stripe	-64	-168
Dblstripe	-7	-194

Stable stripe order:
(over CB)
$J_2 > J_1/2$

Stable double stripe order:
(over stripe)
$J_3 > J_2/2$

Stable FM order:
(over CB)
negative J_1 or J_2

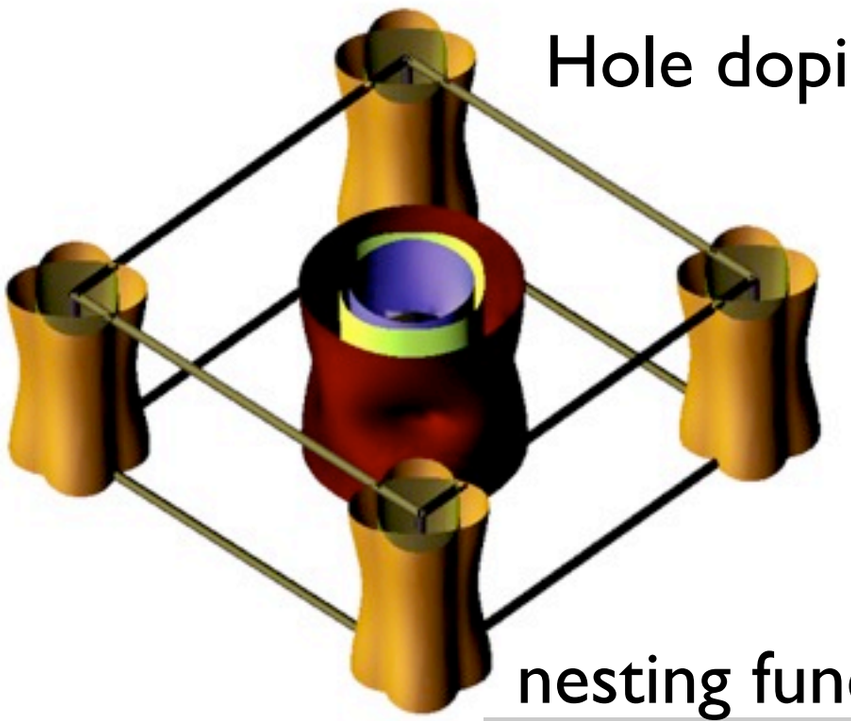
No consistent set of J's possible

μ itself varies strongly with pattern

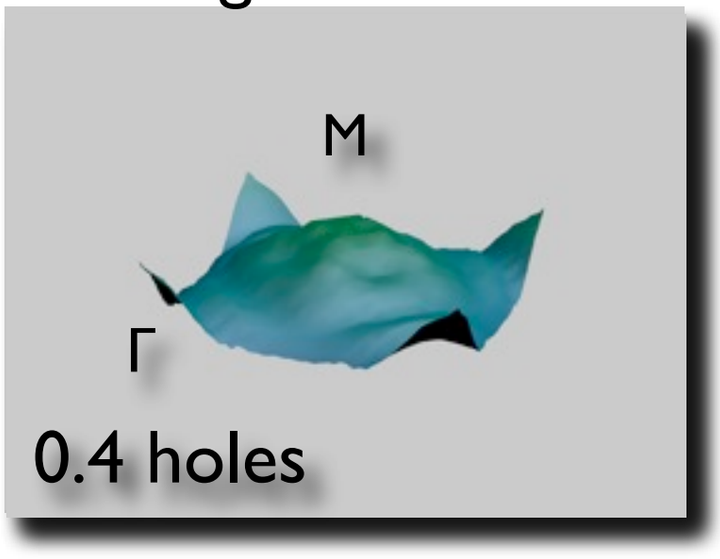
Not typical of localized moments !

Nesting of Fermi surfaces

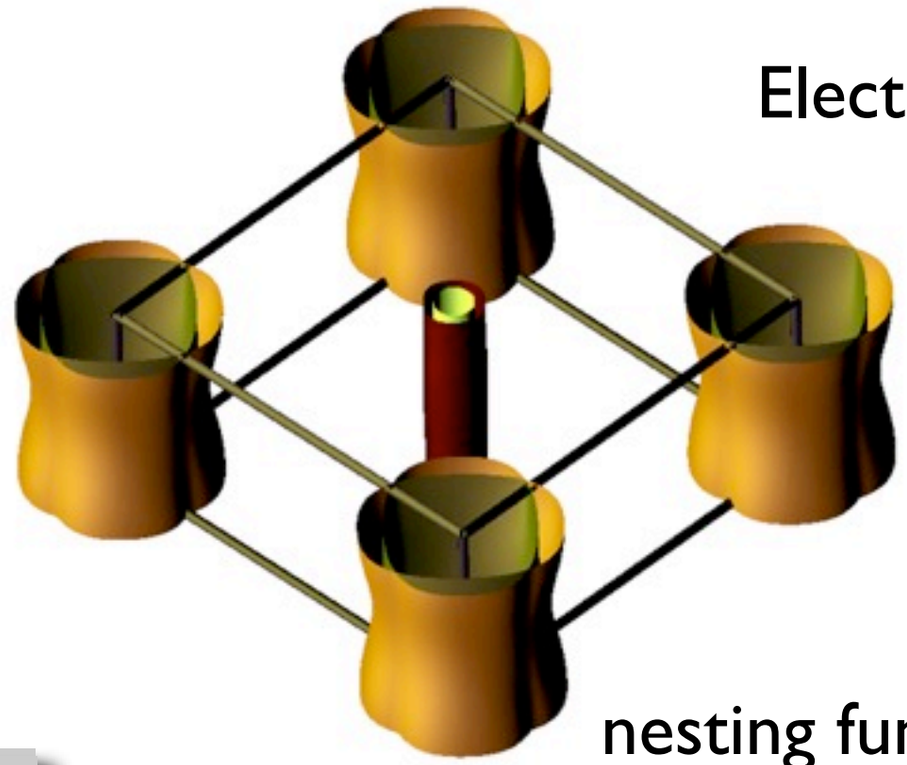
Hole doping



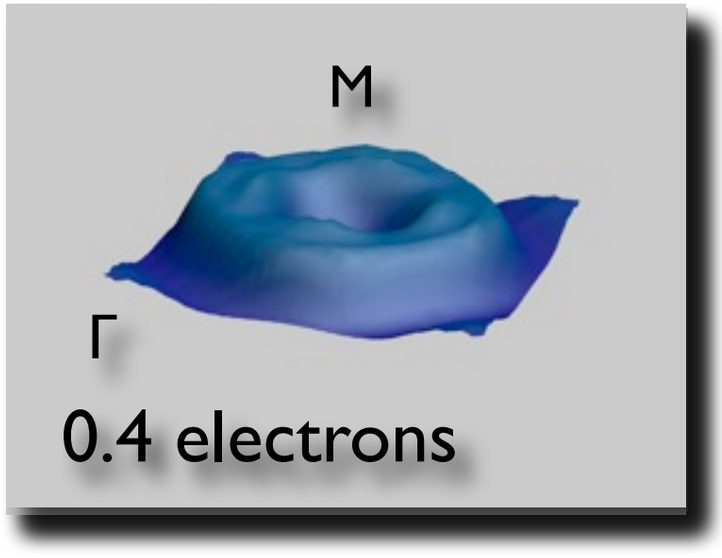
nesting function



Electron doping

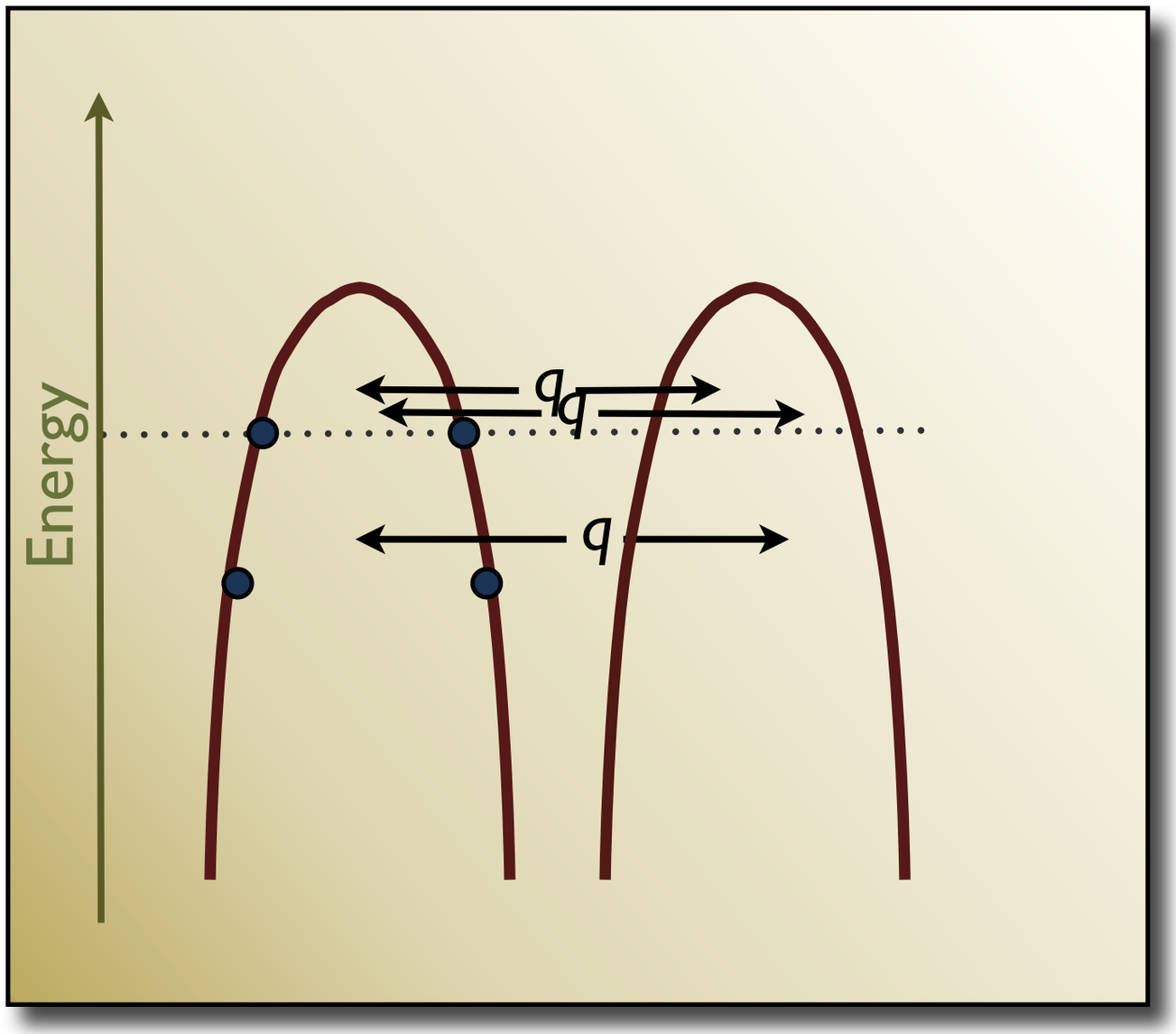


nesting function



Change in relative FS sizes destroys peak (but not weight!) of nesting function

Are nesting and magnetism related?



Nesting function

$$\chi''(\vec{q},\omega)/\omega = \sum_{\vec{k}} \delta(\varepsilon_F)\delta(\varepsilon_{\vec{k}} - \varepsilon_{\vec{k}+\vec{q}}) \qquad \omega \to 0$$

Only Fermi surface points contribute

Real part of susceptibility

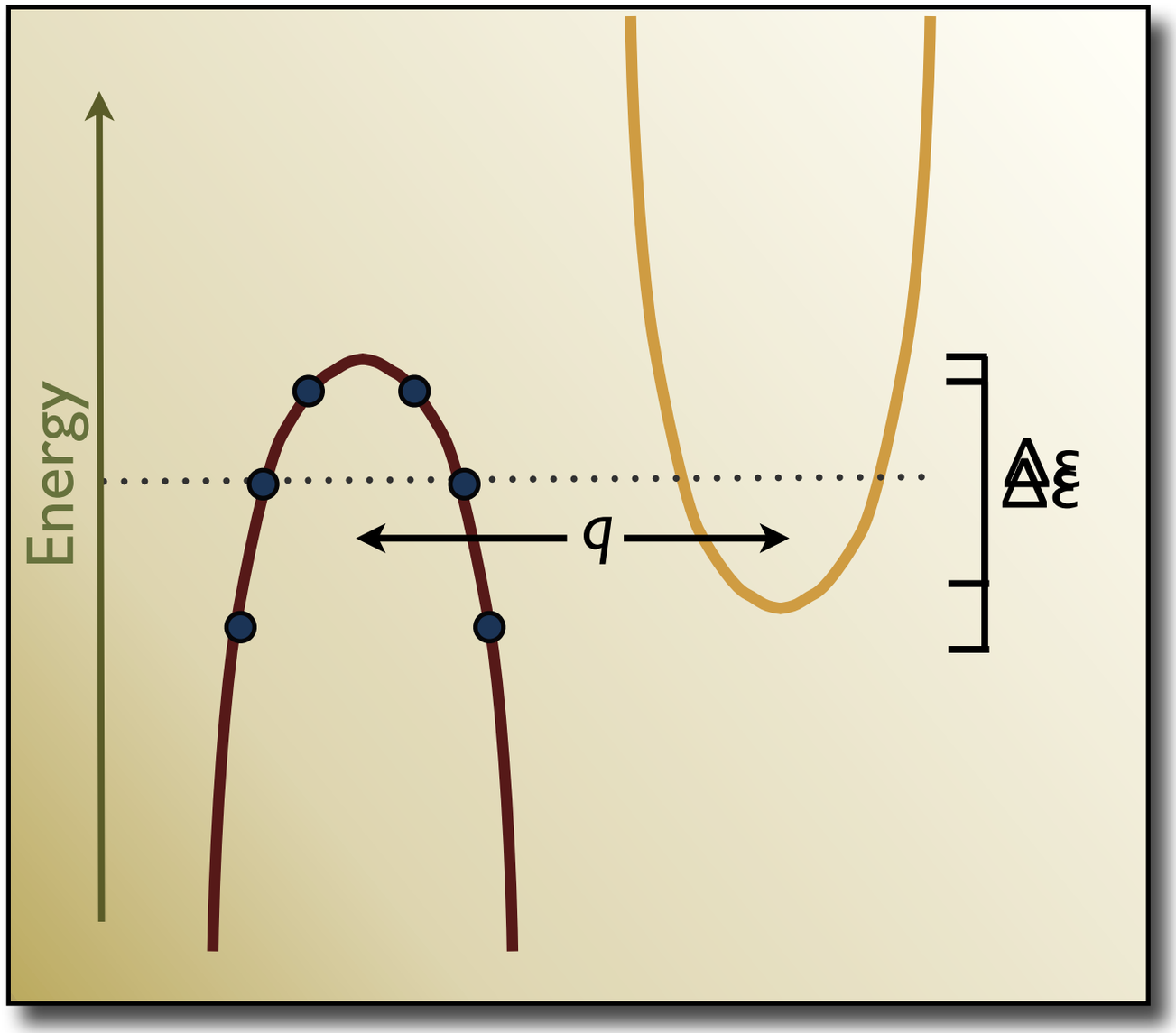
$$\chi'(\vec{q},0) = \sum_{\vec{k}} \frac{f(\varepsilon_{\vec{k}}) - f(\varepsilon_{\vec{k}+\vec{q}})}{\varepsilon_{\vec{k}} - \varepsilon_{\vec{k}+\vec{q}}}$$

Gathers from above and below E_F

Nesting function can have a maximum at a different (sometimes *very* different) place in q-space than real part of χ

The *real part* is responsible for magnetic instability

How does doping change nesting/magnetic properties?



Nesting function
(Spin fluctuations)

$$\chi''(\vec{q},\omega)/\omega = \sum_{\vec{k}} \delta(\varepsilon_F)\delta(\varepsilon_{\vec{k}} - \varepsilon_{\vec{k}+\vec{q}}) \qquad \omega \to 0$$

Real part of susceptibility

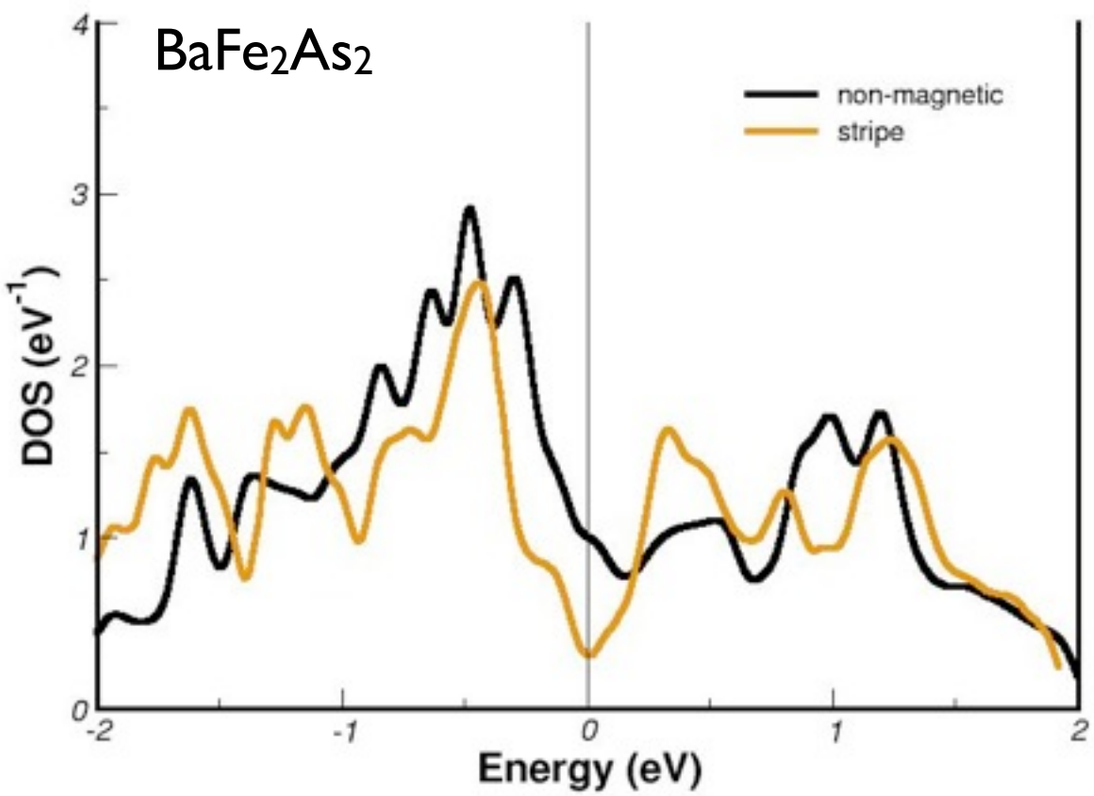
(Magnetic instability)

$$\chi'(\vec{q},0) = \sum_{\vec{k}} \frac{f(\varepsilon_{\vec{k}}) - f(\varepsilon_{\vec{k}+\vec{q}})}{\varepsilon_{\vec{k}} - \varepsilon_{\vec{k}+\vec{q}}}$$

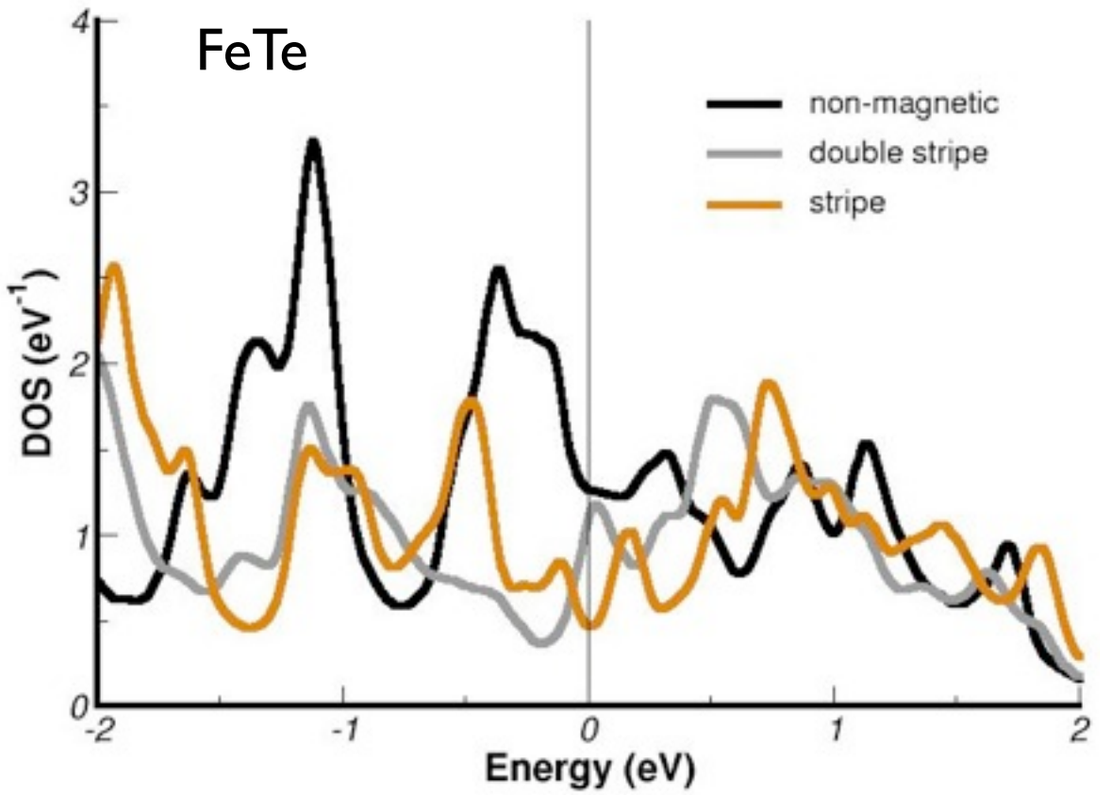
When separate surfaces are hole/electron, nesting and Reχ peaks tend to coincide - but each changes differently with doping

Nesting, especially FS cuts, is not a good predictor of tendency to LRO

Energy gain away from the Fermi energy



Ground state: stripe

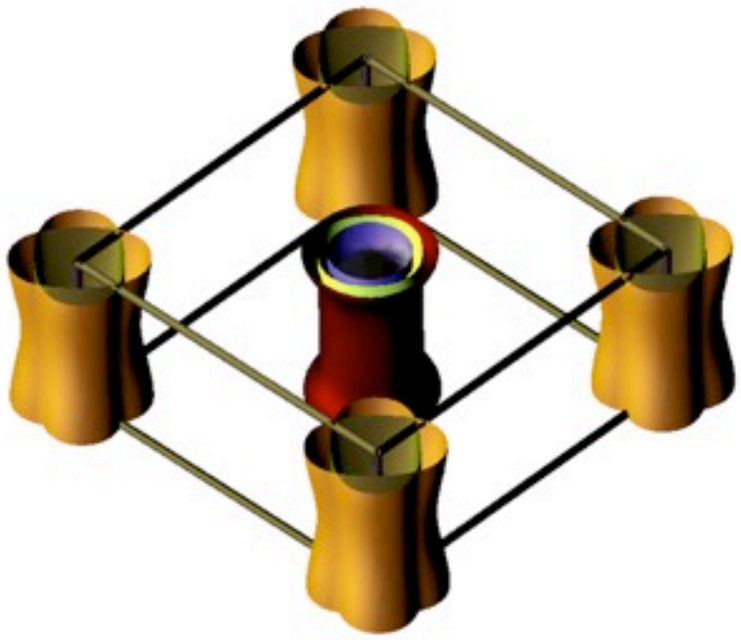


Ground state: double stripe
(stripe also very stable)

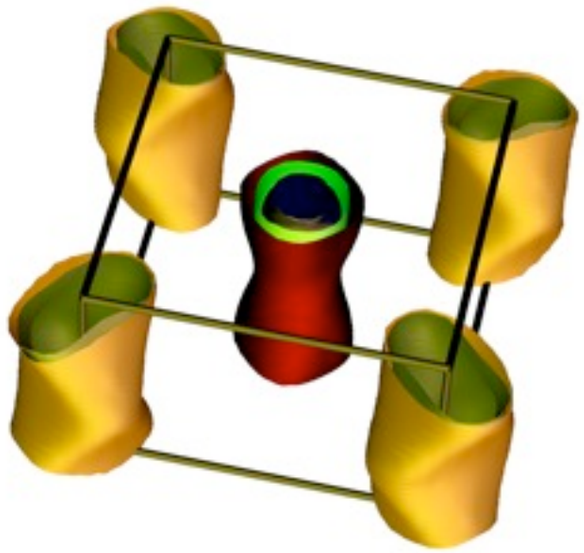
Ground state of both structure types is achieved through lowering of one-electron energies over a wide range

How does FeTe differ electronically from other structural types?

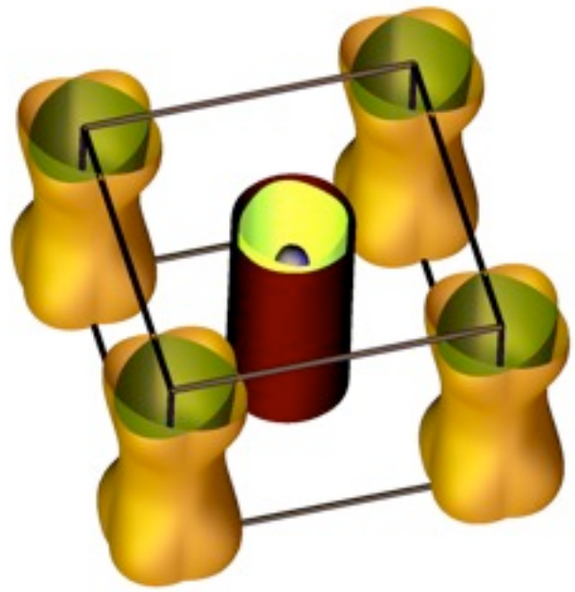
1111



122



FeTe



FeTe is isoelectronically doped (with Se) to achieve superconductivity

FeTe Fermi surface is extremely similar to LaFeAsO and $BaFe_2As_2$ surfaces.

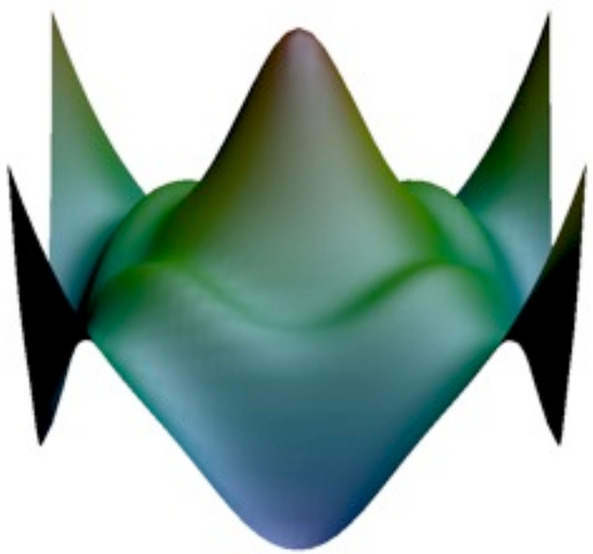
Nesting properties should also be very similar

Re χ_0 could differ (DOS away from E_F shows significant differences)

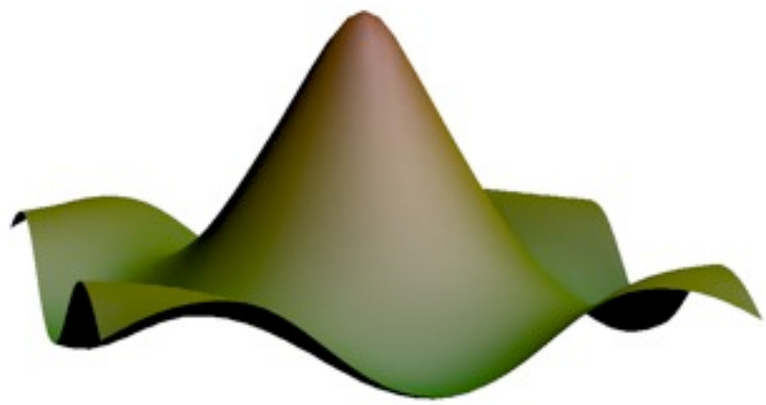
Are the calculable properties different in FeTe?

FeTe

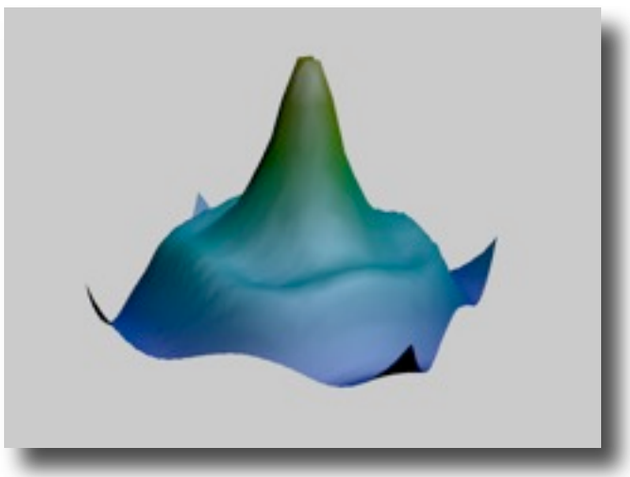
Nesting function

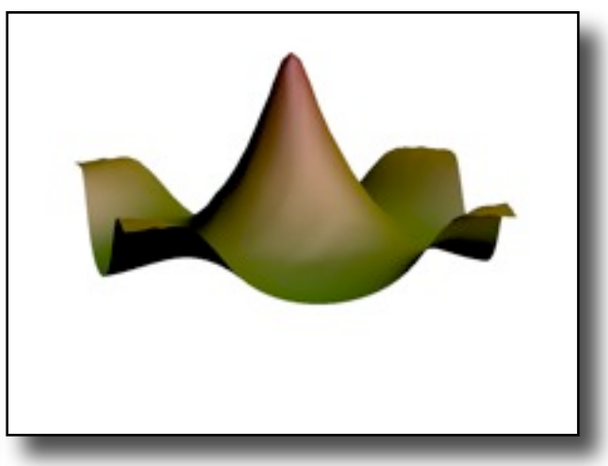


Real part of susceptibility

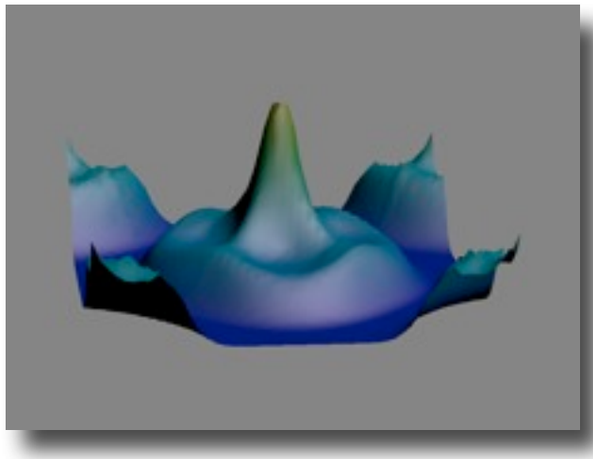


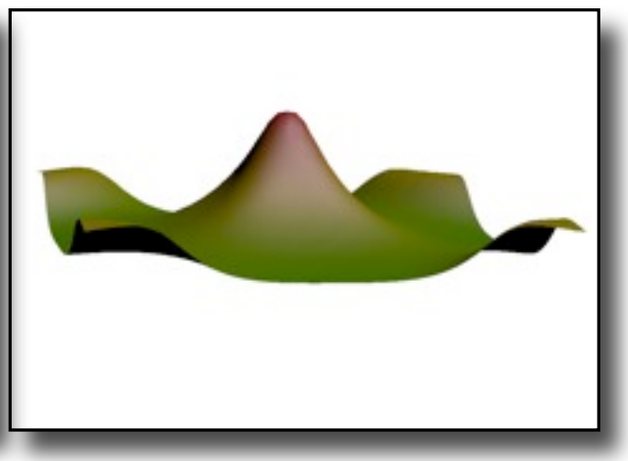
Comparison to 1111, 122 systems:





LaFeAsO





$BaFe_2As_2$

FeTe shows no very different nesting or tendency toward magnetic instability

Mechanism for magnetic LRO is not found in fermiology

Not conventional correlated local moments + superexchange

- Bandwidth is larger than crystal field and larger than Hubbard U
- FM state is essentially unstable, while AFM state is extremely stable

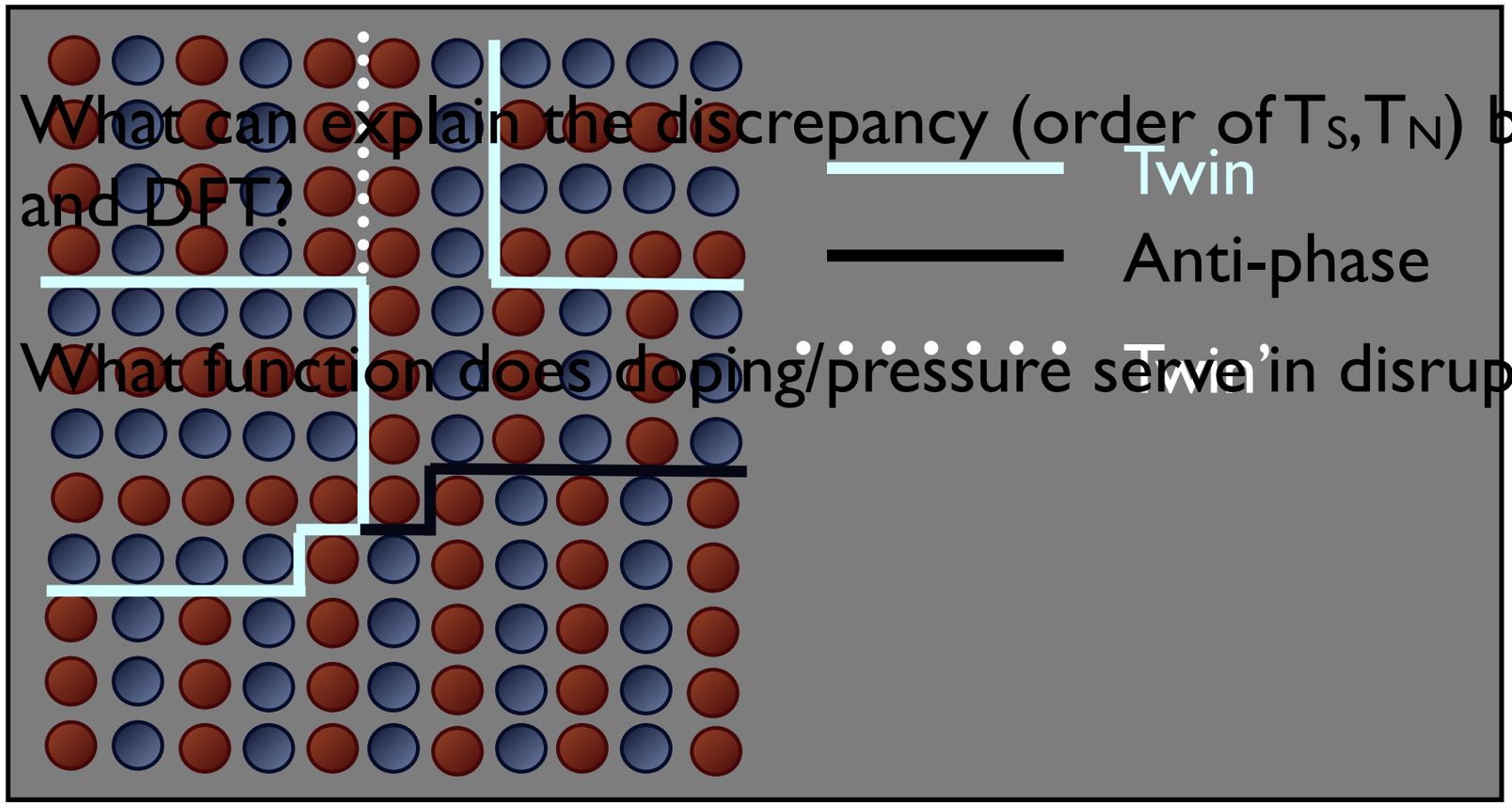
Not FS nesting-driven

- Non-magnetic state is stable against small, but not large AFM perturbations
- Large magnetization is incompatible with Fermi surface driven order

Dual character magnetism

- Local moment (large) formation driven by Hund's rule coupling
- Long range ordered imposed by "itinerant coupling" utilizing entire bandwidth

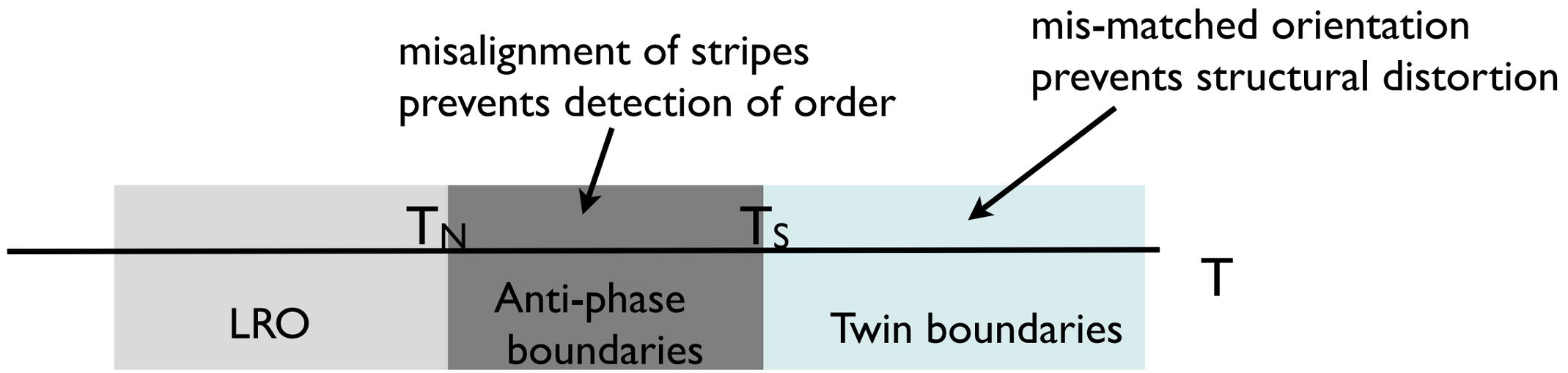
What does the fluctuating magnetism look like?



What can explain the discrepancy (order of T_S, T_N) between experiment and DFT?

What function does doping/pressure serve in disrupting LRO?

Dynamic domains



Relationship to 'nematic' phase

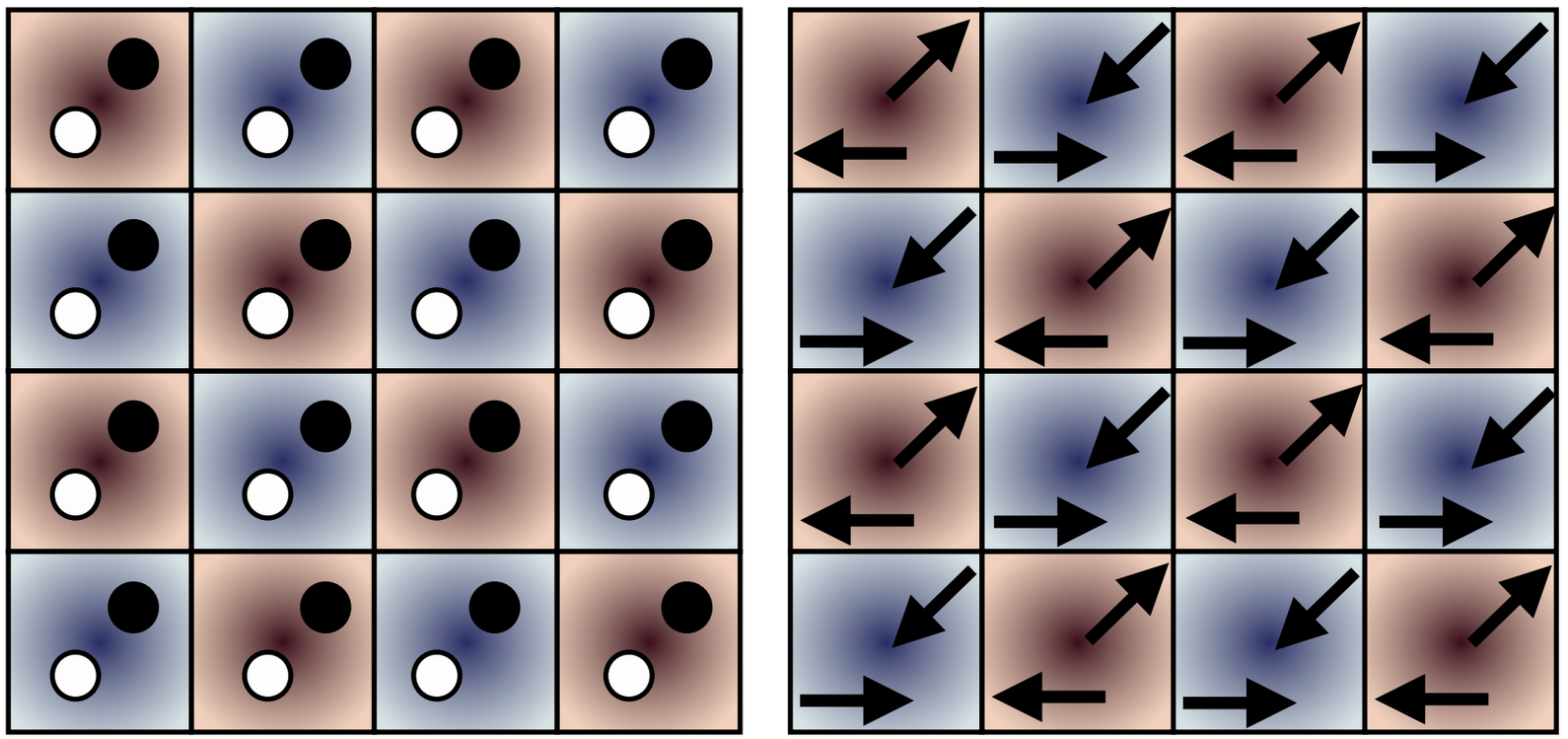
Fang *et al.* PRB **77**, 224509 (2008)

Xu *et al.* PRB **78**, 020501(2008)

Two sublattices, two independent Heisenberg order parameters :

$m_1 = \langle M_i^{\bullet} \rangle$

$m_2 = \langle M_i^{\circ} \rangle$



Mazin and Schmalian arXiv: 0901:4790

Coupling between two sublattices is an Ising variable:

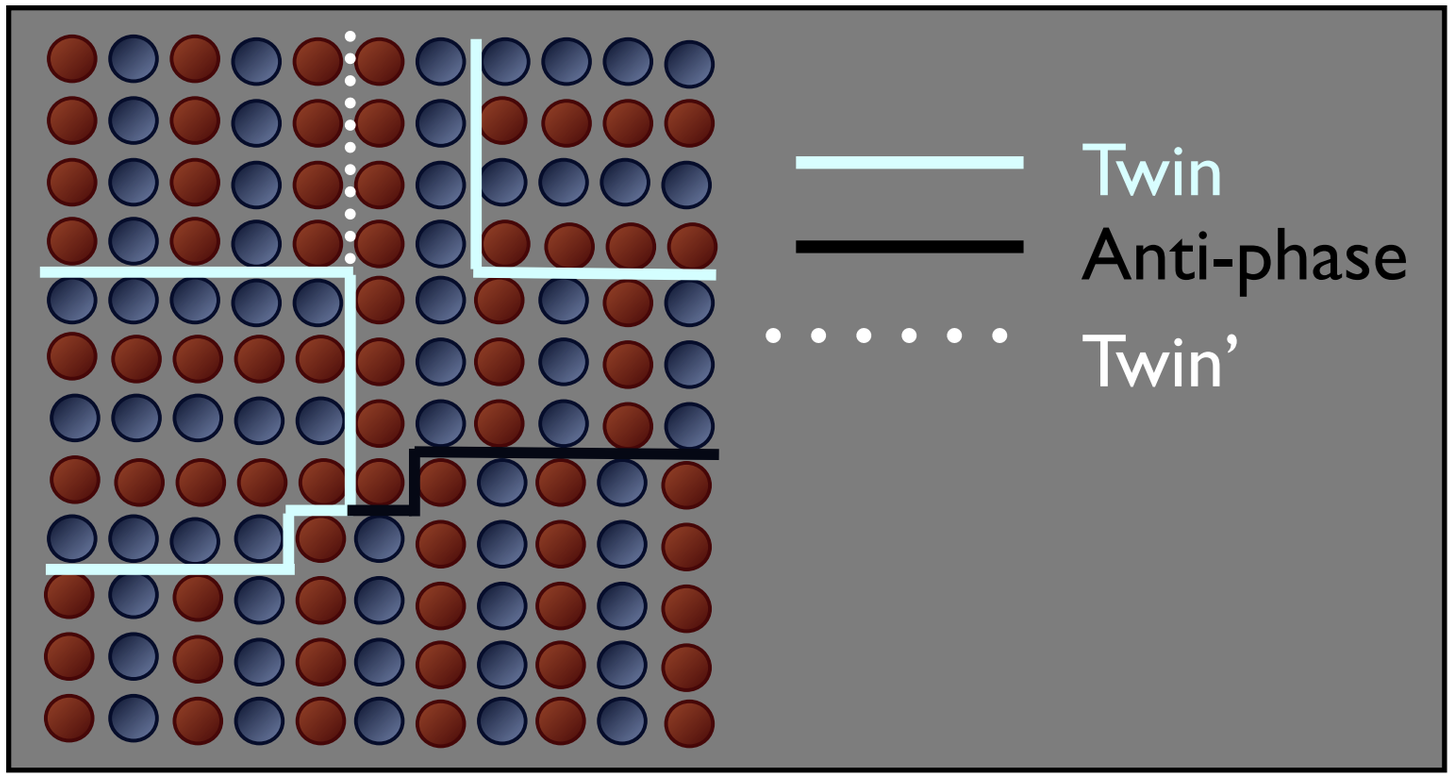
$\sigma = \langle M_i^{\bullet} \cdot M_i^{\circ} \rangle$ Spins inside unit cell must be coupled by bi-linear term

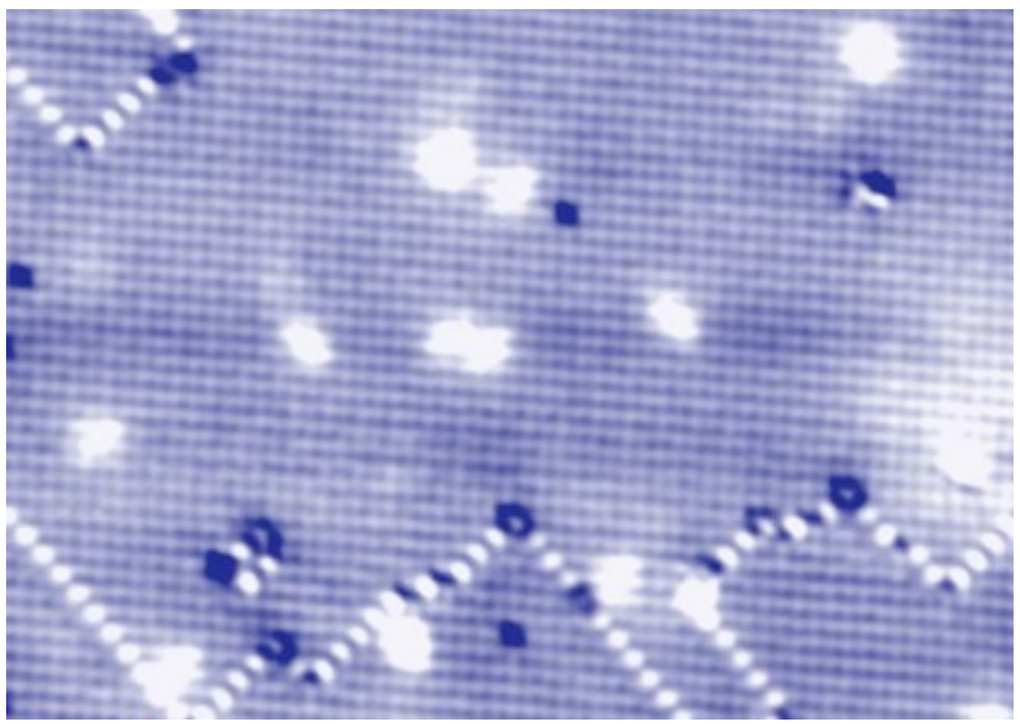
tendency to (π,π) ordering is only necessary element (J_1/J_2 model not essential)

Our domain model is a snapshot of region with finite σ, fluctuating m_1, m_2

Nematic phase

Magnetic domains





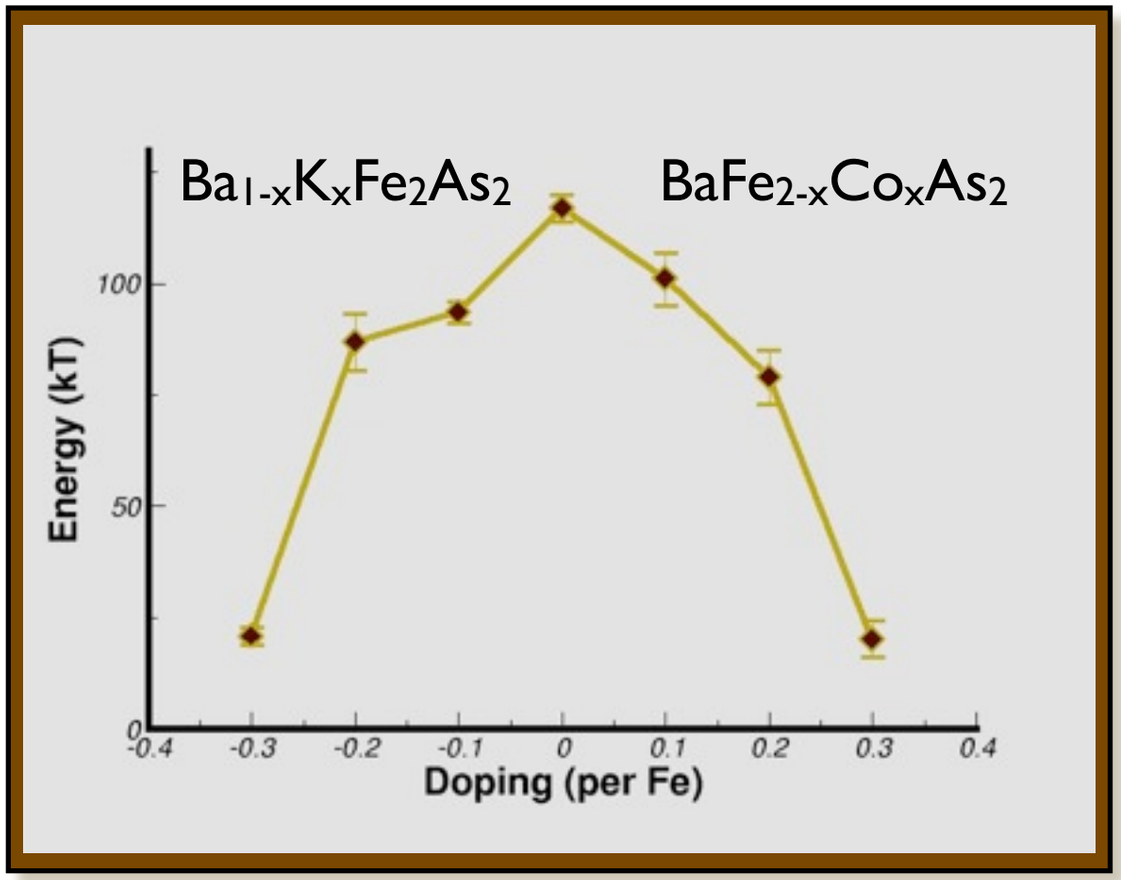
STM can see frozen anti-phase domains that match the prediction

Li *et al.* arXiv:1006.5907

How does doping kill long range order?

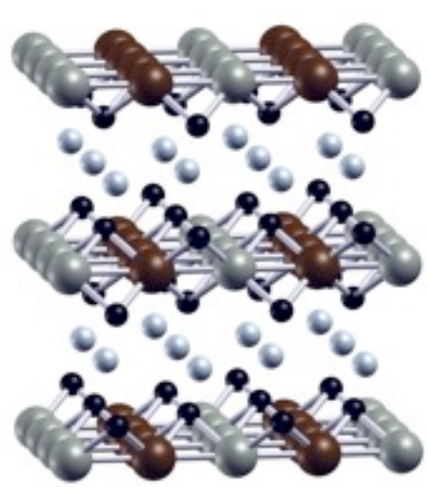
In 2D, spin fluctuations prevent LRO above T=0 (Mermin-Wagner)

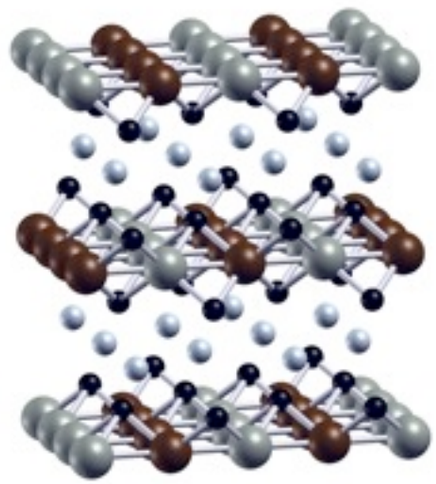
Degree of three-dimensionality could be key to ordering



DFT calculation of interplanar coupling:

$E_{FM} - E_{AFM}$

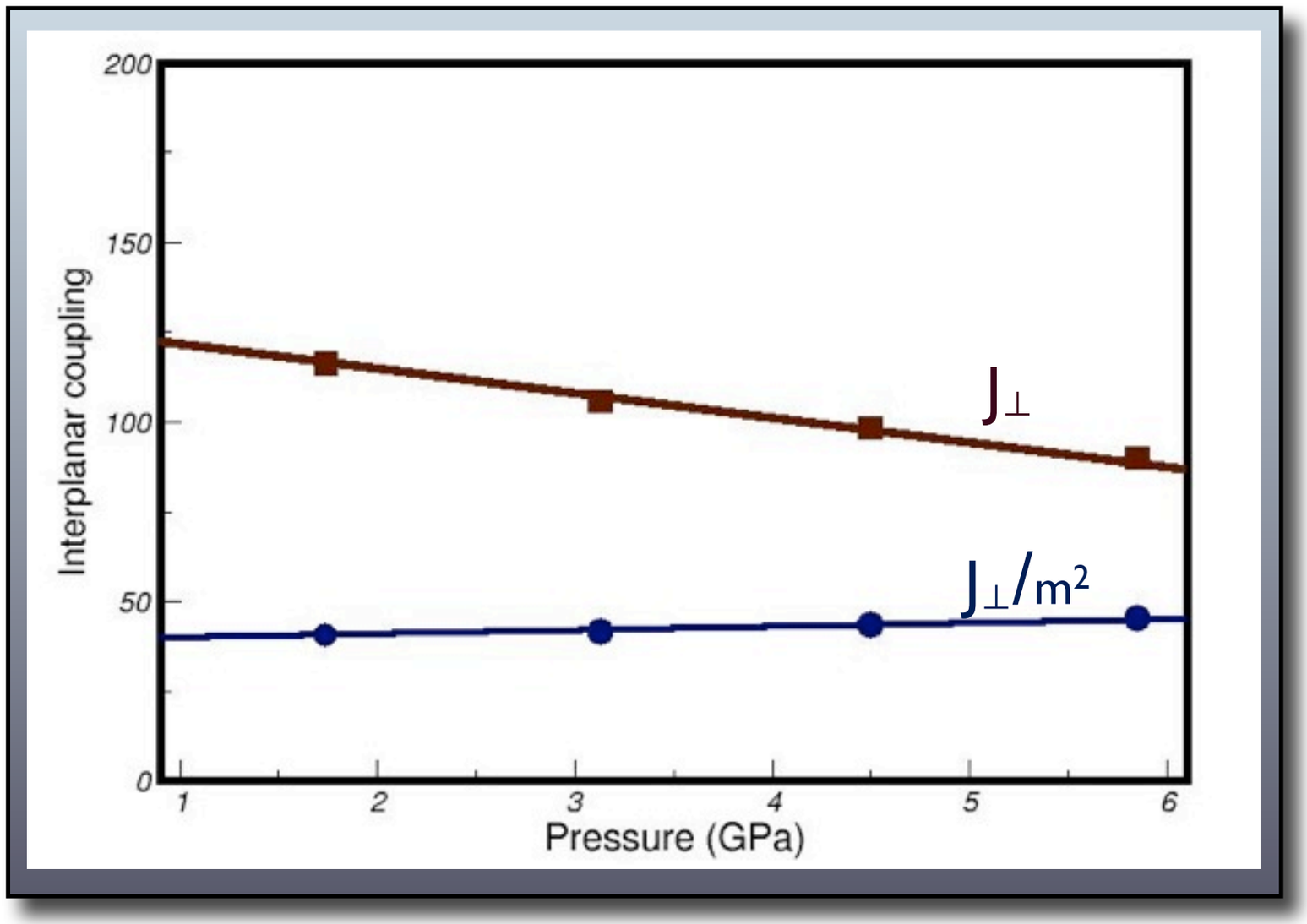




- Coupling between planes dies off with both hole and electron doping
- Energy scales with ordering temperature

Pressure dependency of interplanar coupling

Pressure decreases magnetic coupling between planes (increased SF)



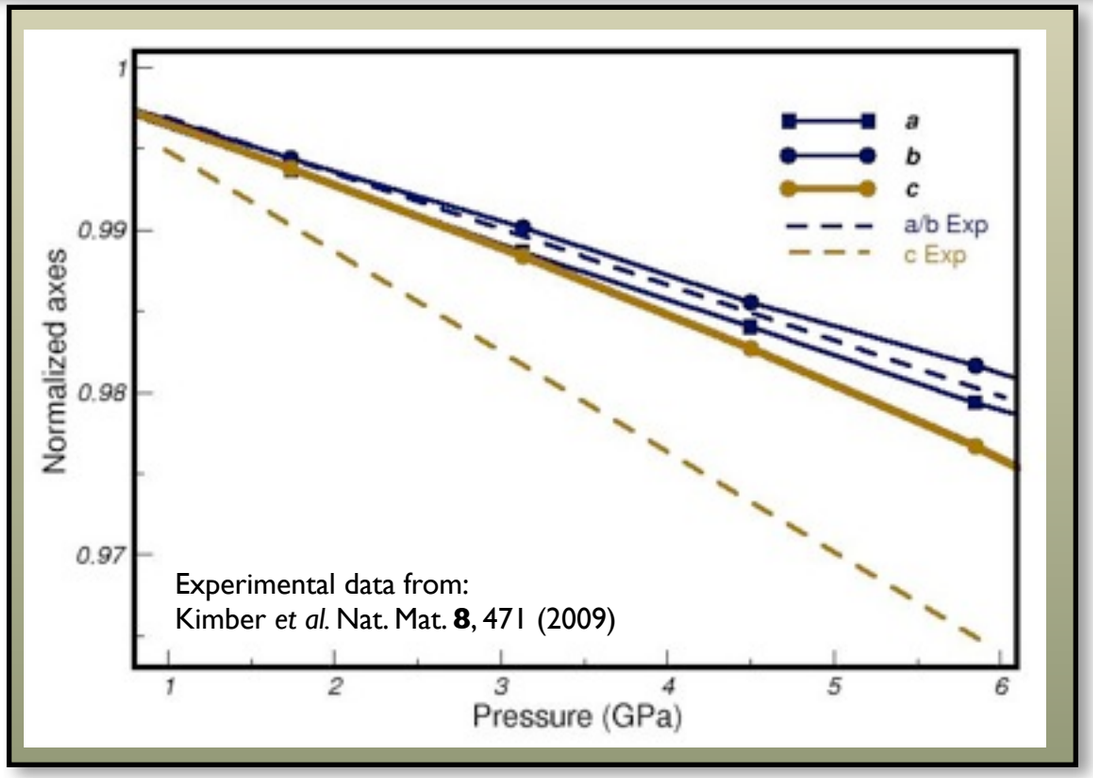
$J_\perp = E_{FM} - E_{AFM}$

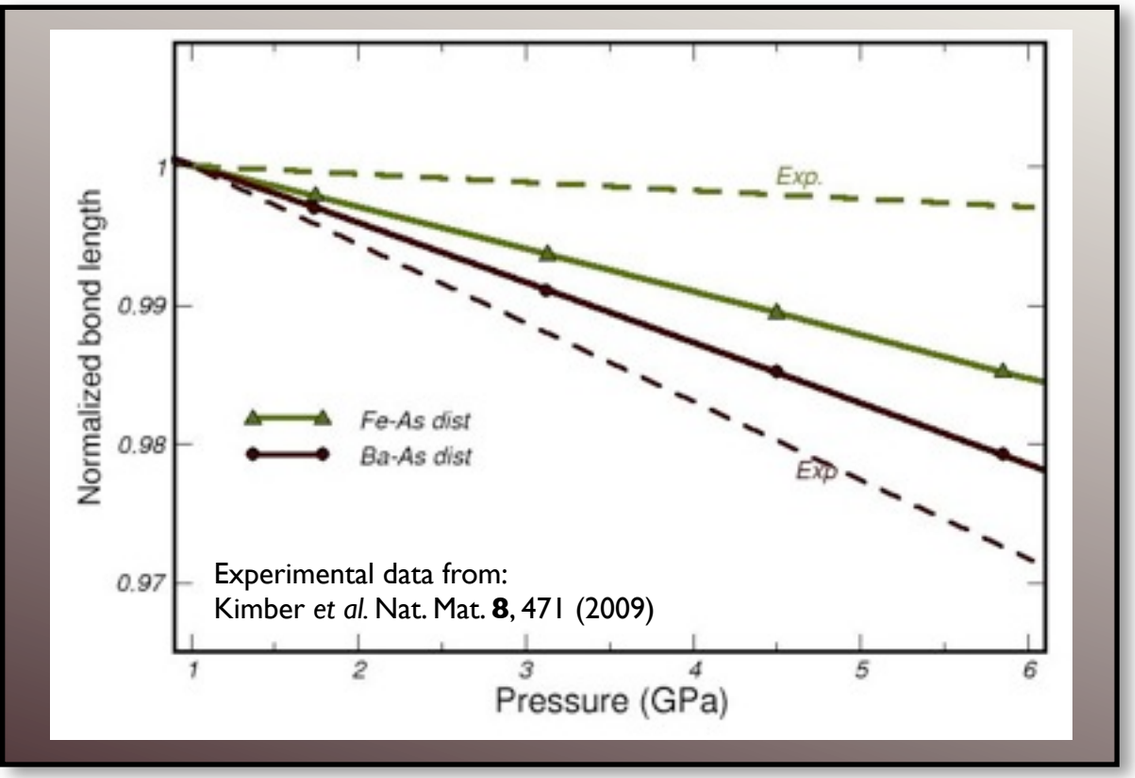
Interaction between planes increases, but overall decrease in $J_\perp$ stems from <u>decreased magnetic moment</u>

Pressure dependency of structural parameters

- *a,b* parameters reproduce experiment well
- *c* parameter considerably stiffer than experiment

Note that DFT system has LRO *throughout* pressure range; experiment loses LRO at ~3GPa





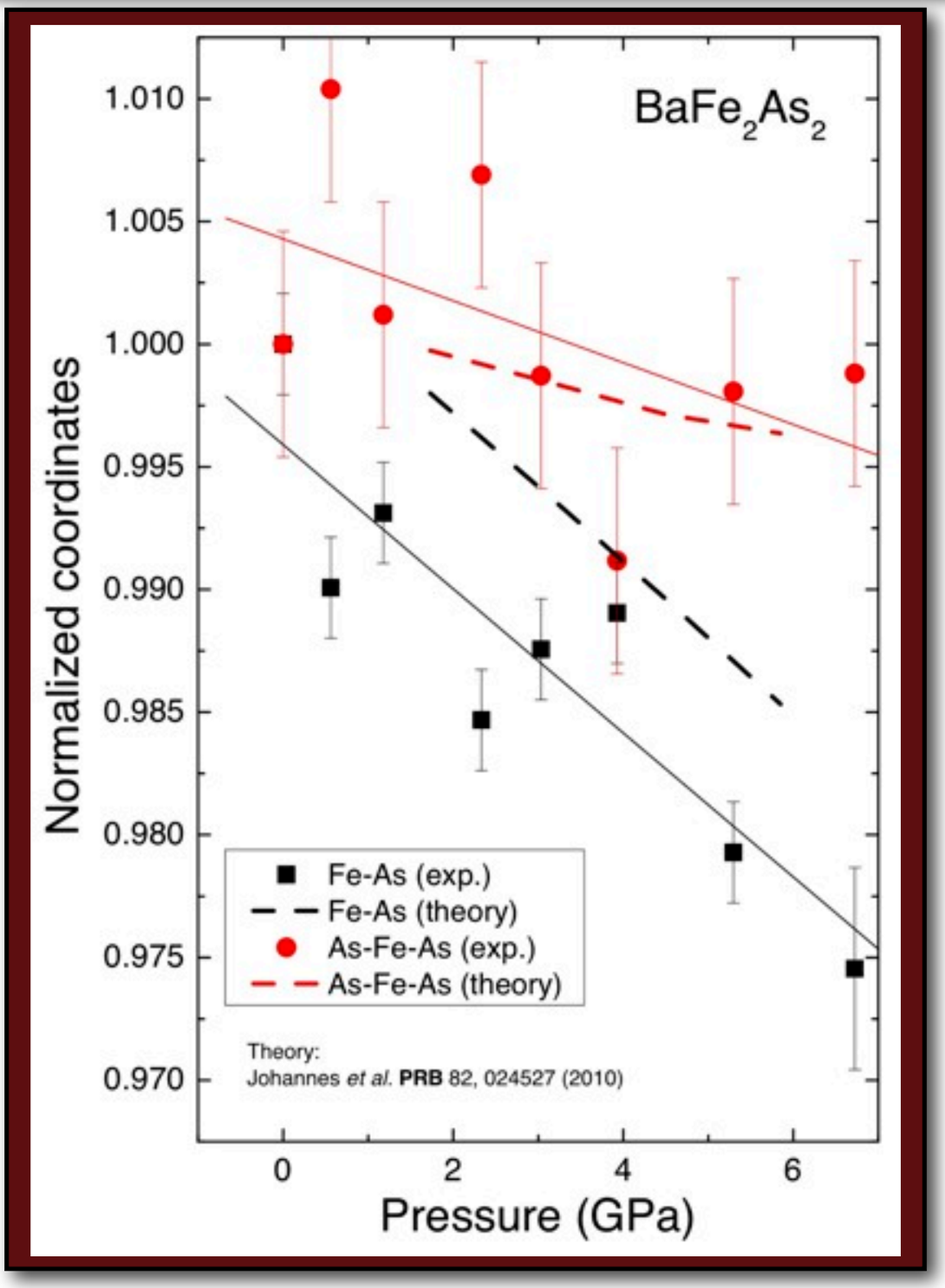
- DFT system accomodates pressure by shrinking Fe-As bond; experimental bond is extremely rigid

Since Fe-As bondlength is determined by μ_{Fe}, moment must not change with pressure

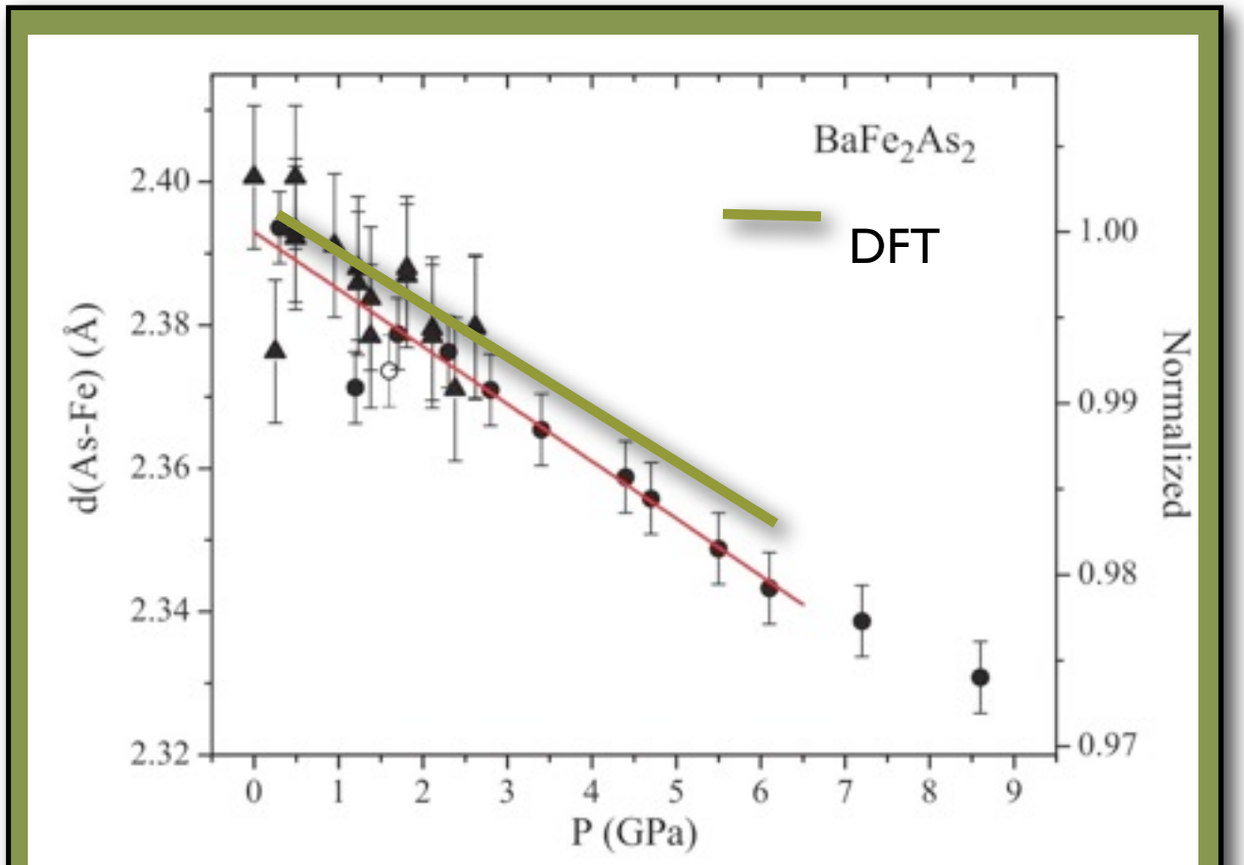
→ pressure increases spin fluctuations rather than decreasing moment, but DFT decreases moment

Residual local Coulomb correlations may be missed by DFT

But do DFT and experiment really disagree?



Thesis of I. Efthimiopoulos, Stuttgart, 2010

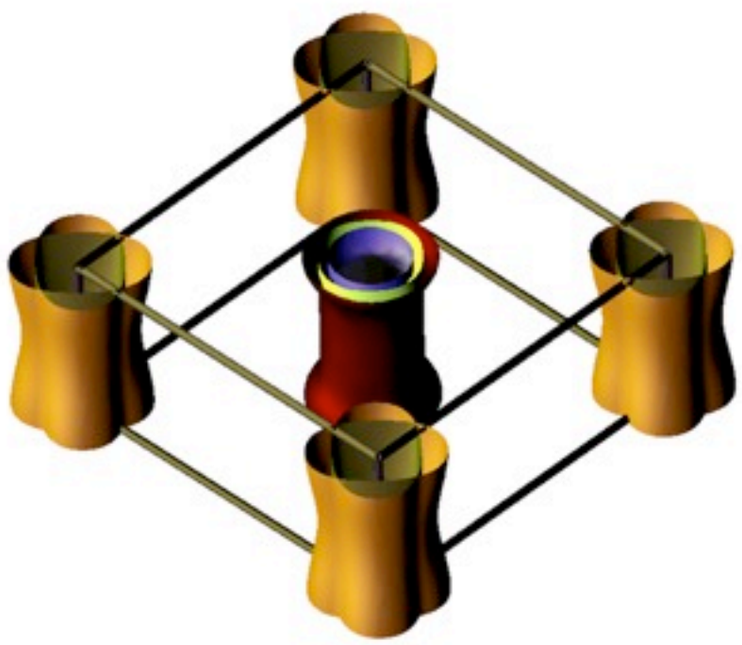


Granado *et al.* Phys. Rev. **B** 184508 (2011)

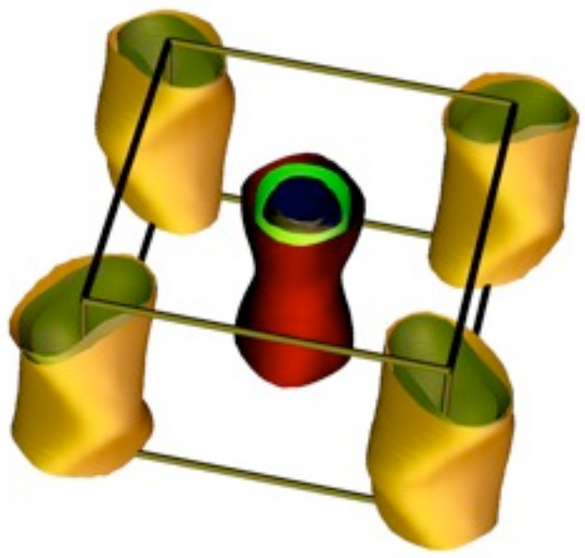
The size/evolution of the moment is crucial for the Fermiology and the interplanar coupling - that is, for both magnetism and superconductivity.

Non-magnetic Fermi surfaces

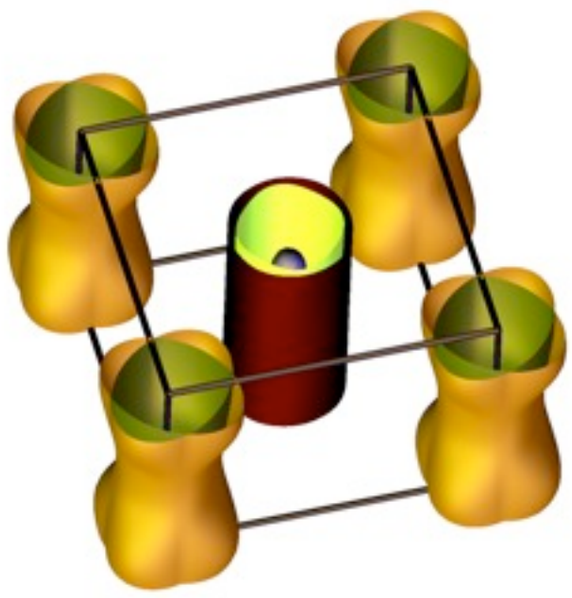
1111



122



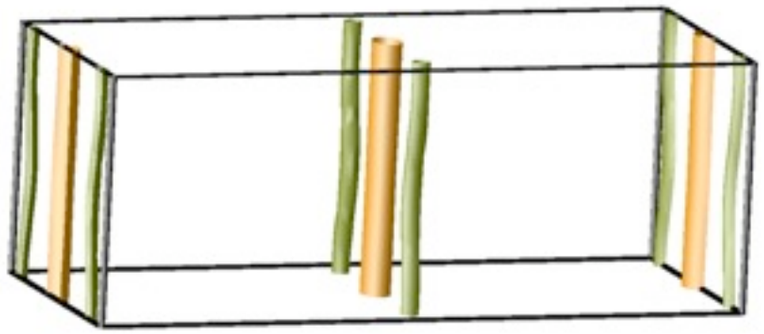
FeTe



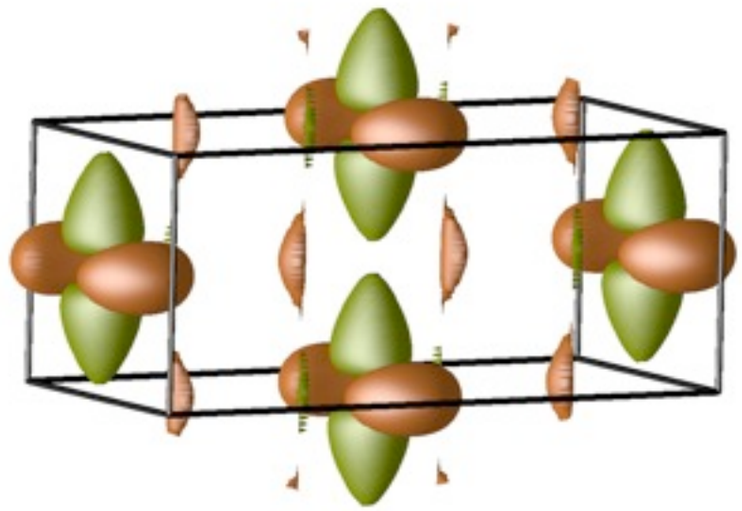
Magnetic Fermi surfaces

Extreme differences exist between the structural types in terms of magnetic Fermi surfaces

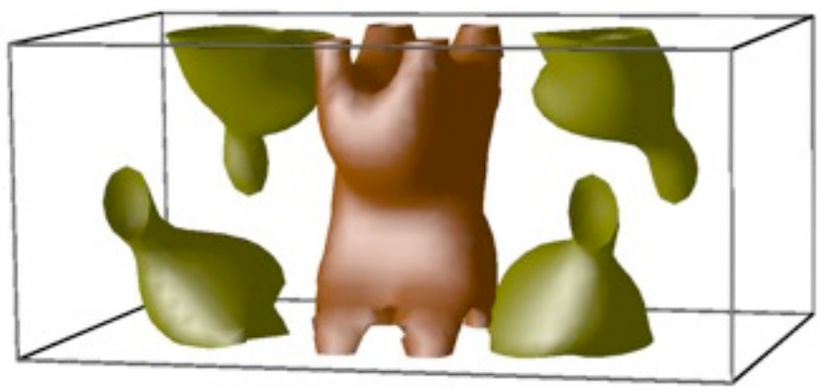
1111 (LaFeAsO)



122 ($BaFe_2As_2$)

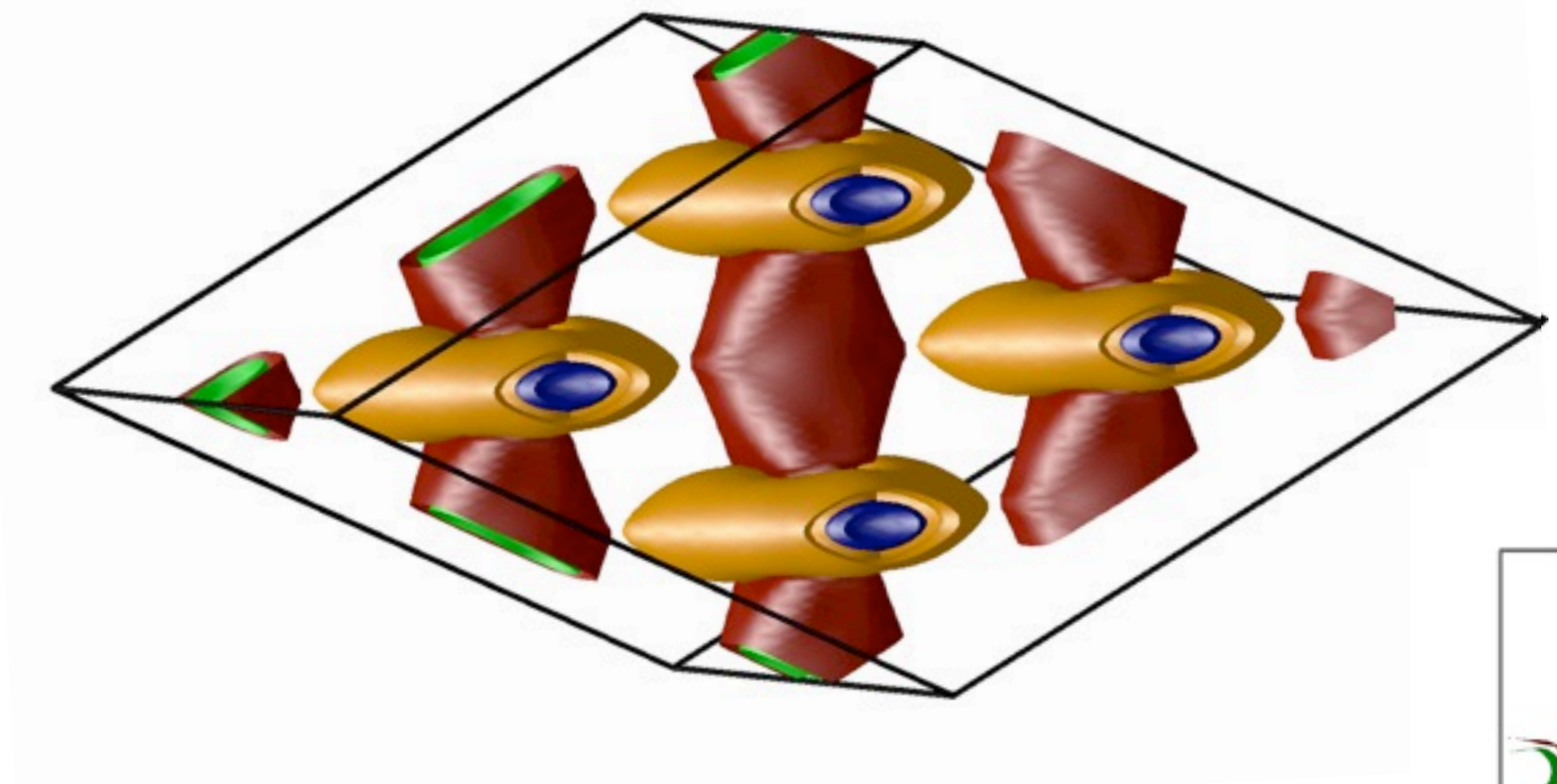


11 (FeTe)

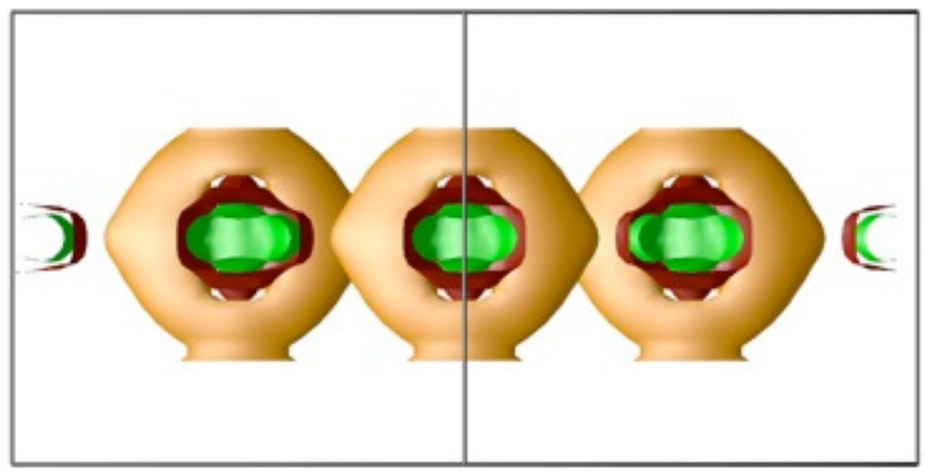


Pressure may affect different "types" differently

Evolution of Fermiology with pressure ?



interior surface



Pressure dramatically changes FS topology

- Notable 3-dimensionality
- No nesting between hole and electron surfaces

Caveat: it takes quite a bit more pressure to kill moment in DFT than in reality

Summary

- DFT calculations have been very successful in understanding and even predicting some properties of Fe-based superconductors

  Precise distortion pattern
  Ground state magnetic long range order

- DFT calculations miss some properties that may be crucial

  Size of the magnetic moment
  Evolution of structure with pressure ?

- Even DFT failures shed some light on underlying physics

  Fe-based superconductors are paramagnetic
  Structure and magnetism are inextricably linked