Iron Based Superconductors: Lessons from Raman Spectroscopy

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Outline

Part I

1. Intro to Fe based superconductors
2. Signatures of structural transition
3. Raman as a probe of orbital correlations
4. Critical slowdown of orbital fluctuations

Part II

1. Family of iron(Fe)-based superconductors with only electron pockets: $A_xFe_{2-y}Se_2$, A= K,Rb,Cs
2. Pairing symmetry: “s”, “d”,”s+-” and “s+i d”
3. Raman in $A_xFe_{2-y}Se_2$, A= K,Rb,Cs

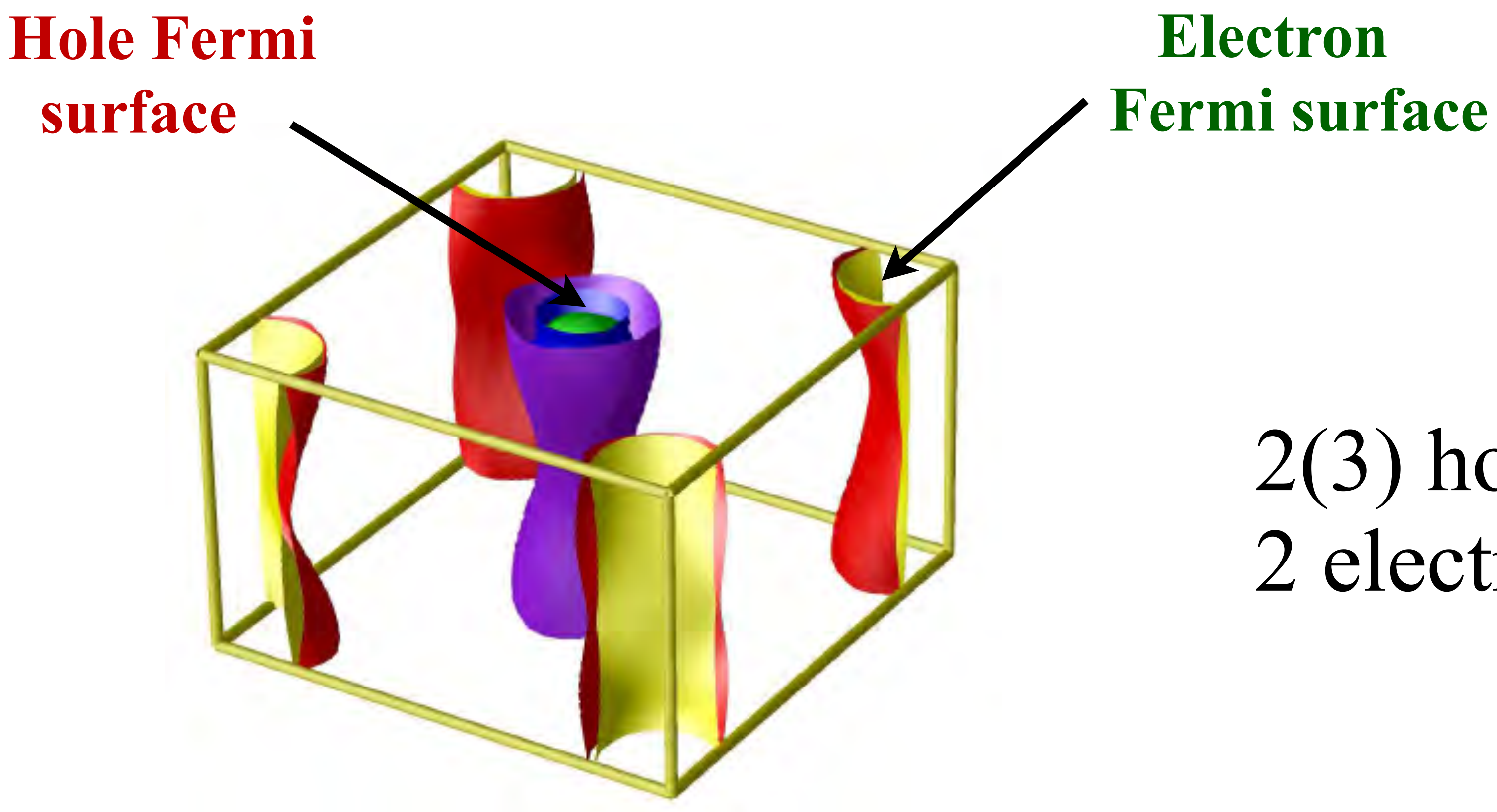
I. Band structure

1. Multi-band multi-orbital metals

2. Quasi-two-dimensional materials

3. Conduction bands:
   partially filled *dxz dyz (dxy)* orbitals

Band structure(cont)



2(3) hole pockets at $(0, 0)$
2 electron pockets at (π, π)

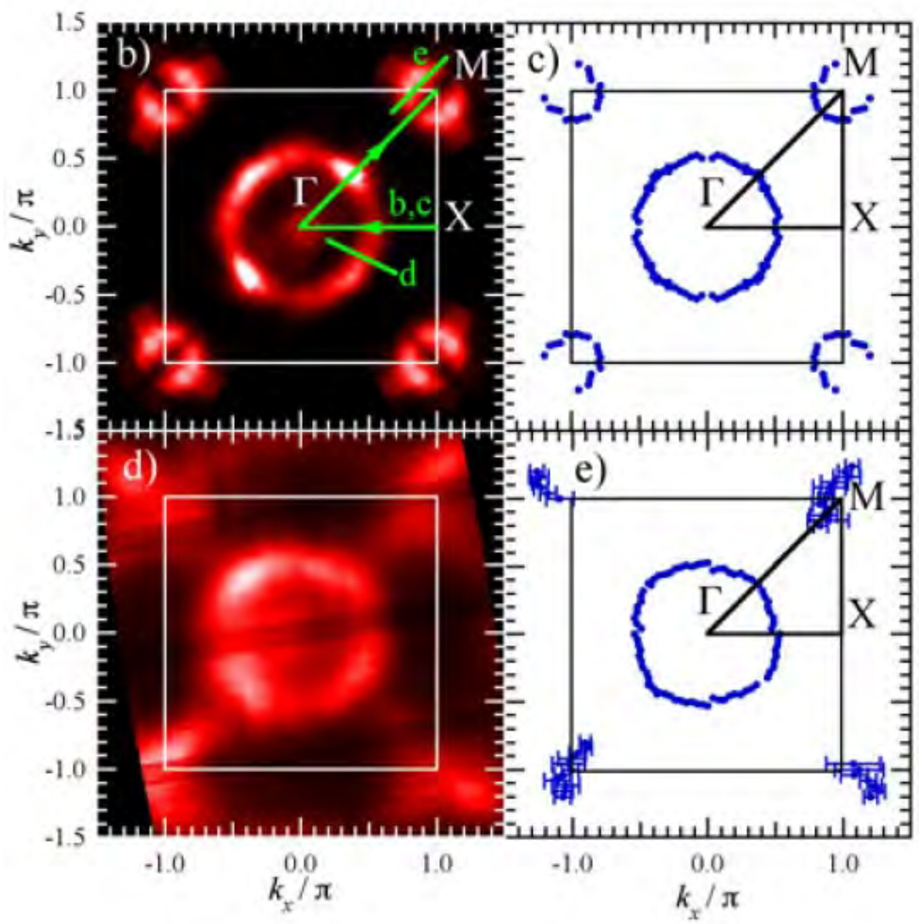
Lebegue, Mazin et al, Singh & Du, Cvetkovic & Tesanovic...

Band structure (cont)

ARPES

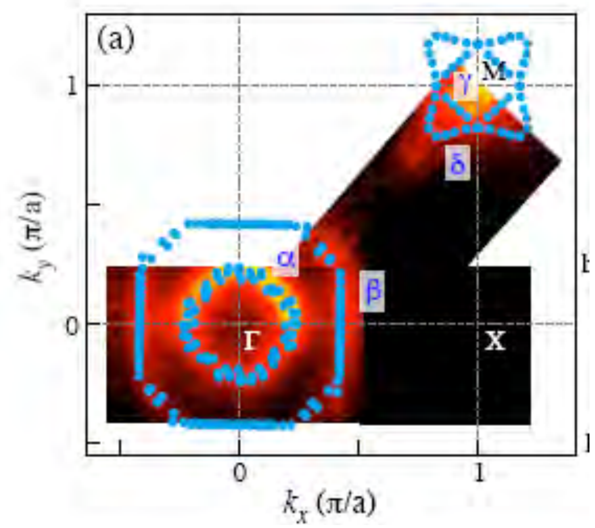
$NdFeAs(O_{1-x}F_x)$ (x=0.1)

A. Kaminski et al.



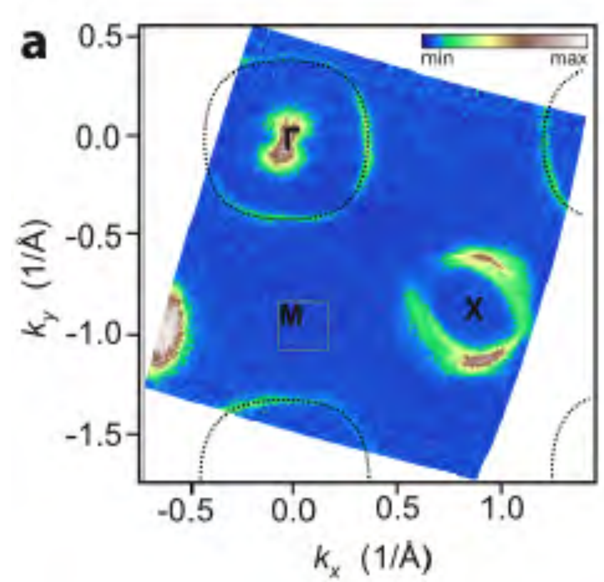
$Ba_{06}K_{04}Fe_2As_2$

H. Ding et al.



LiFeAs

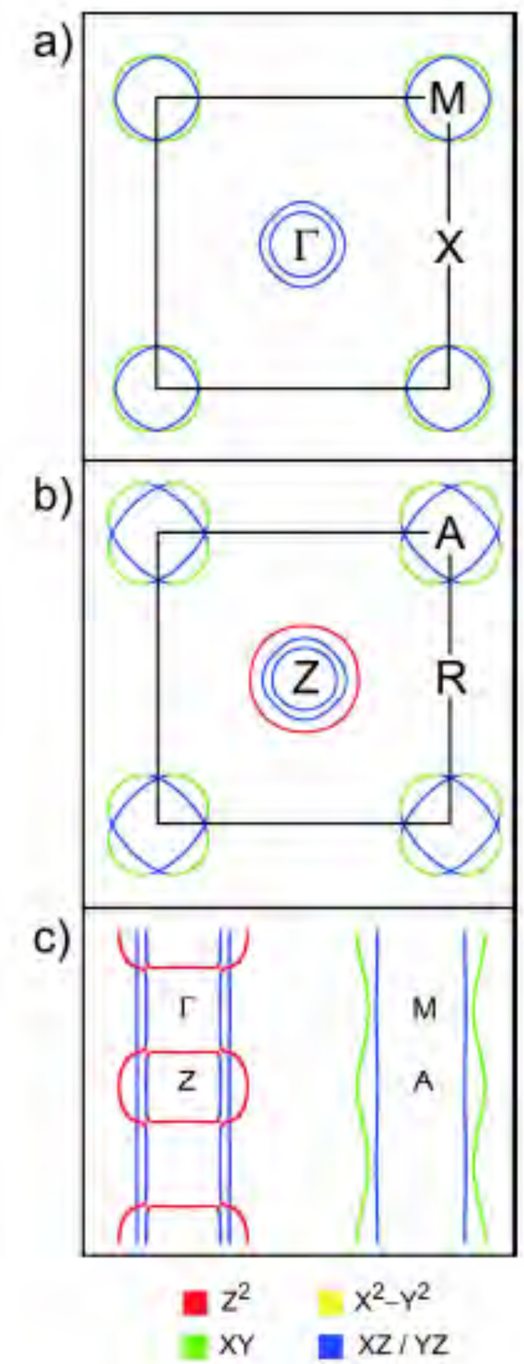
A. Kordvuk et al



dHVa

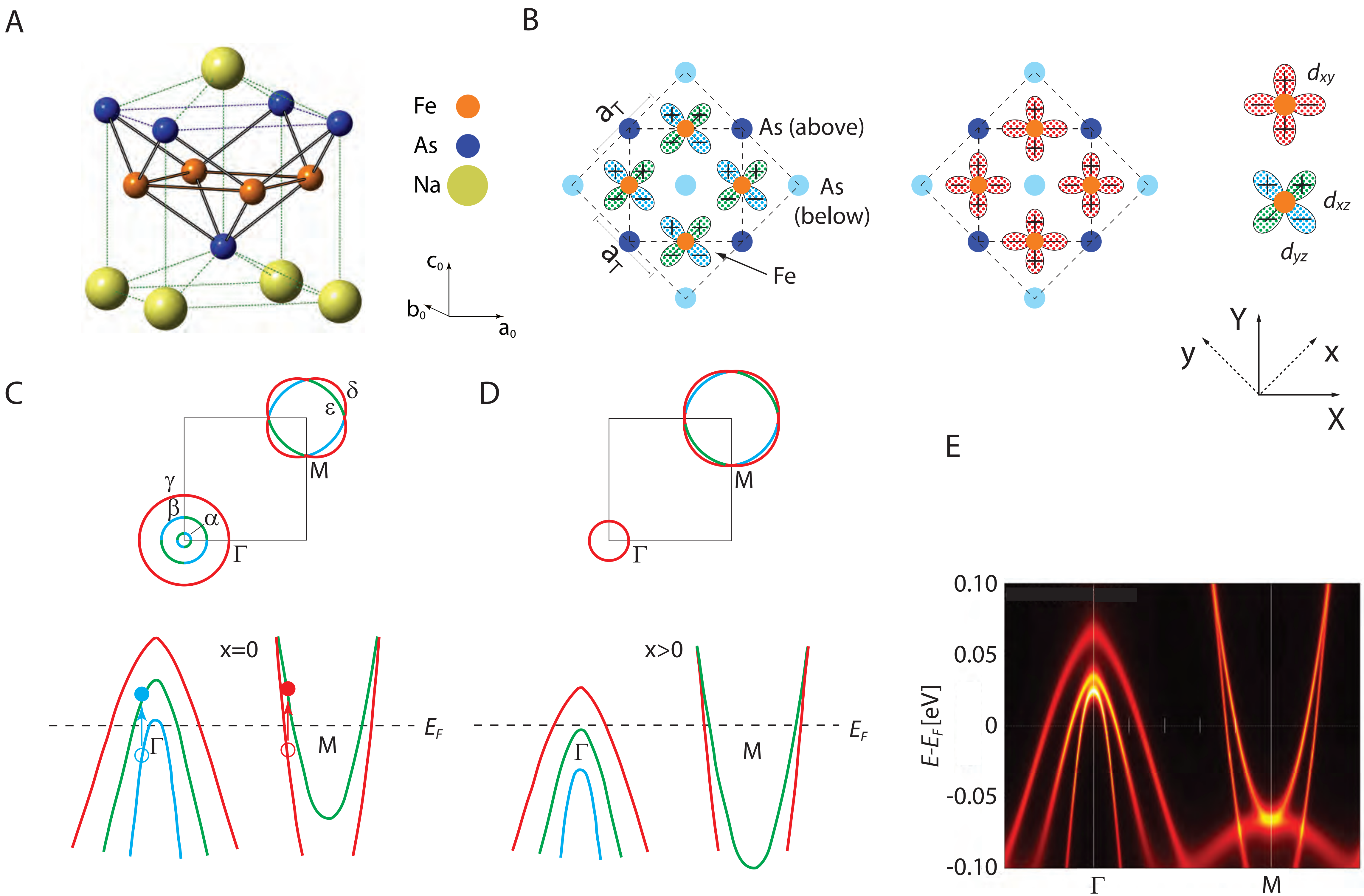
LaFeOP

A. Coldea et al,



Hole pockets near (0,0)
Electron pockets near (π,π)

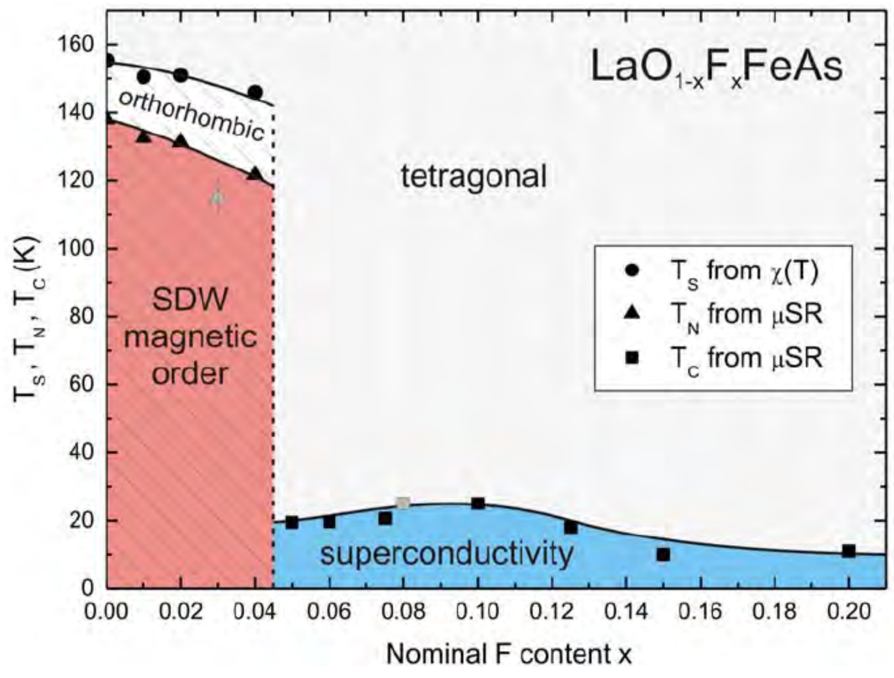
$NaFe_{1-x}Co_xAs$



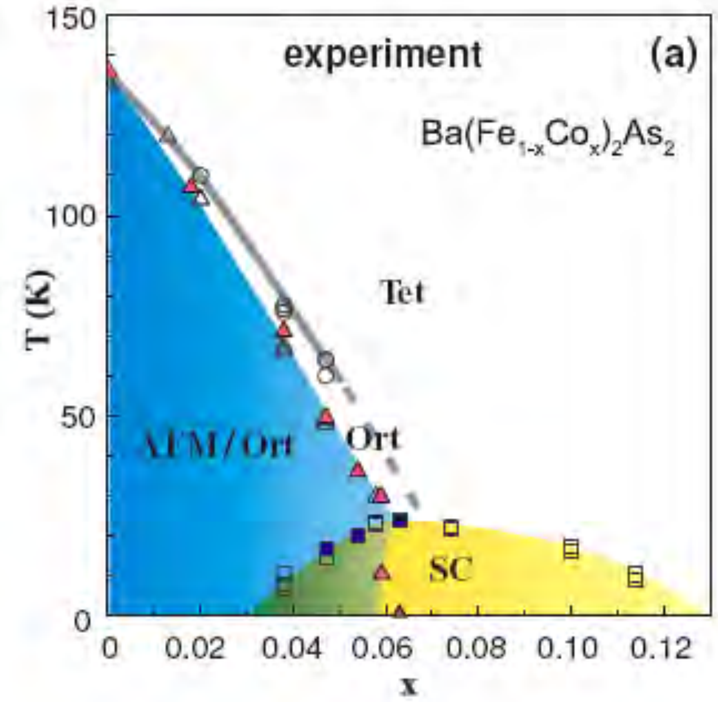
arXiv:1410.6456

V. K. Thorsmølle, MK, Z. P. Yin, Chenglin Zhang, S. V. Carr, Pengcheng Dai, G. Blumberg

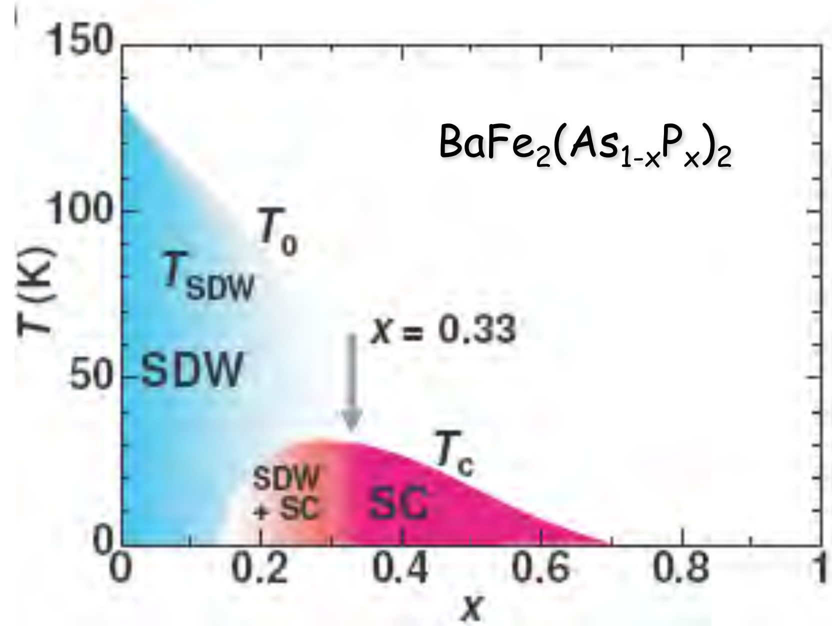
II. Phase Diagram



Luetkens et al



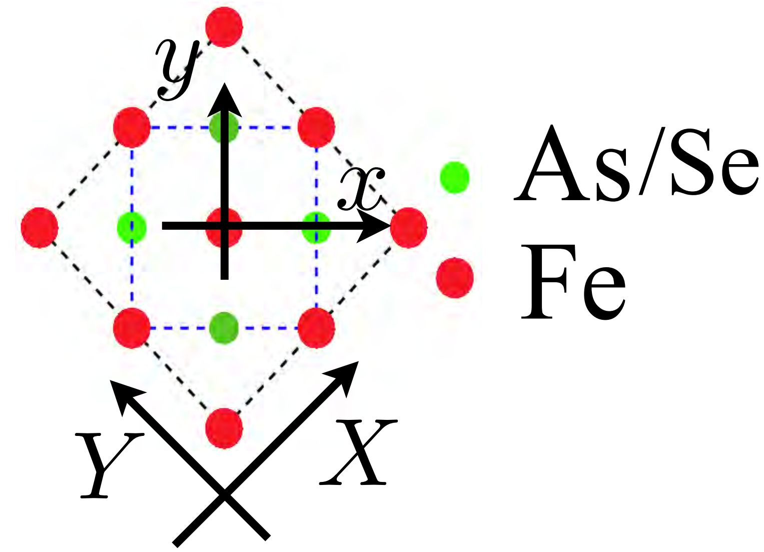
Fernandes et al

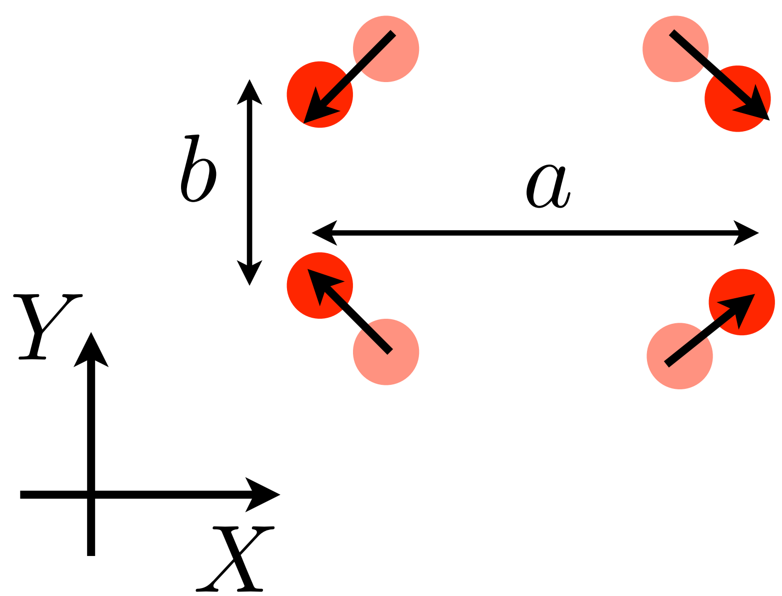


Matsuda et al

Structural transition

Orthorhombic distortion





$a \neq b$ anisotropy

Discrete $C_4 \rightarrow C_2$ symmetry breaking

$$\epsilon_6 = \frac{\partial u_X}{\partial X} - \frac{\partial u_Y}{\partial Y} \neq 0$$

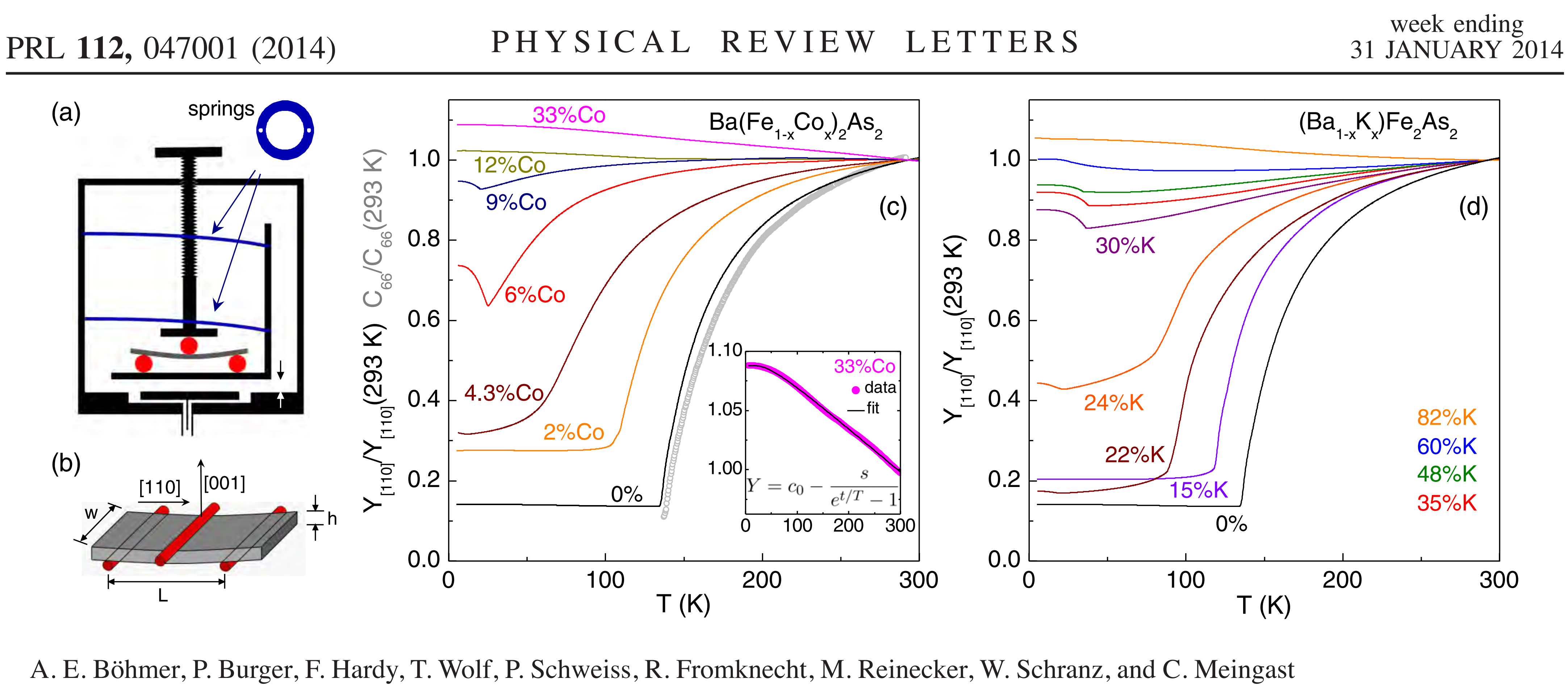
Order Parameter

Structural transition: lattice softening

Shear *stress* → $\sigma_6 = C_{66}\epsilon_6$ ← Shear *strain* (orthorhombic distortion)

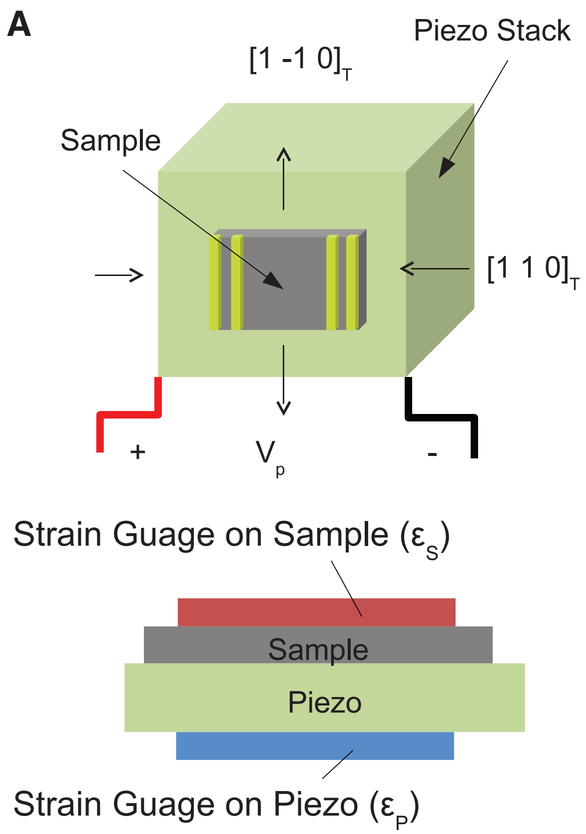
↓

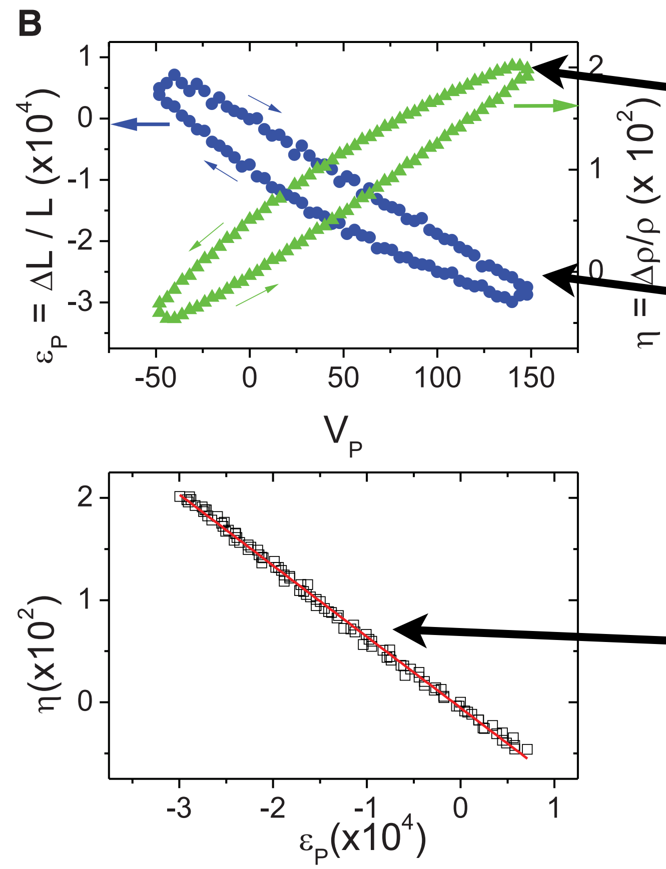
Elastic shear modulus



Towards transition: $C_{66} \rightarrow 0$

Structural transition: electrons vs lattice

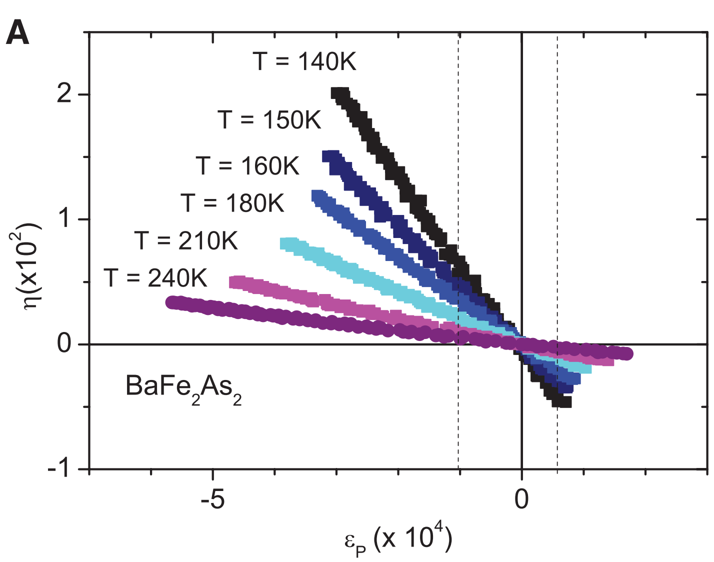


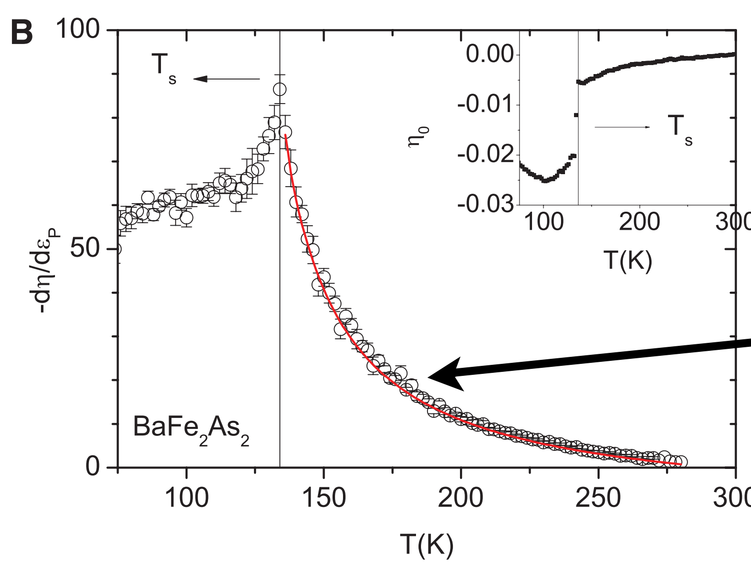


Resistivity anisotropy

Fractional length change

Resistivity is larger (smaller) along short (long) direction



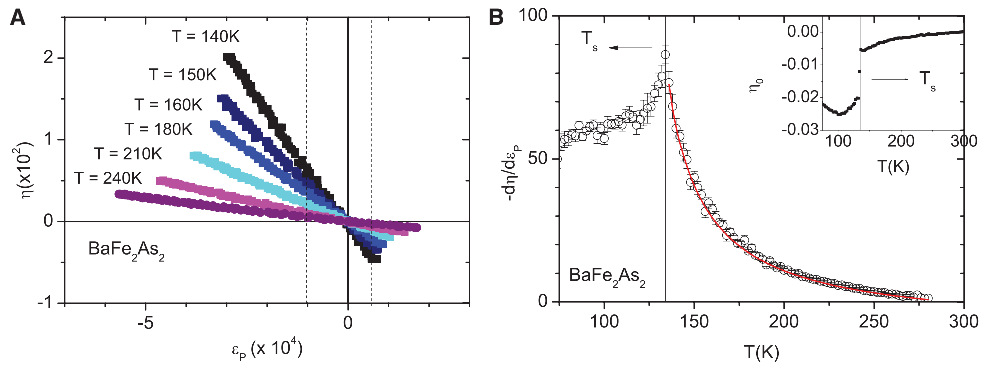


Divergent anisotropy susceptibility!

$$\bar{T}_s < T_s$$

116K < 138K

Jiun-Haw Chu *et al.* *Science* **337**, 710 (2012)



$$\bar{T}_s < T_s$$

116K < 138K

$$F[\phi, \epsilon_6] = \frac{1}{2}\chi_\phi^{-1}\phi^2 - \lambda\epsilon_6\phi + \frac{1}{2}C_{66,0}\epsilon_6^2 + \frac{B}{4}\phi^4$$

Strained controlled: $\epsilon_6 \to 0$

$$F[\phi, \epsilon_6 = 0] = \frac{1}{2}\chi_\phi^{-1}\phi^2 + \frac{B}{4}\phi^4 \quad \Rightarrow \quad \chi_\phi^{-1} = a(T - \bar{T}_s)$$

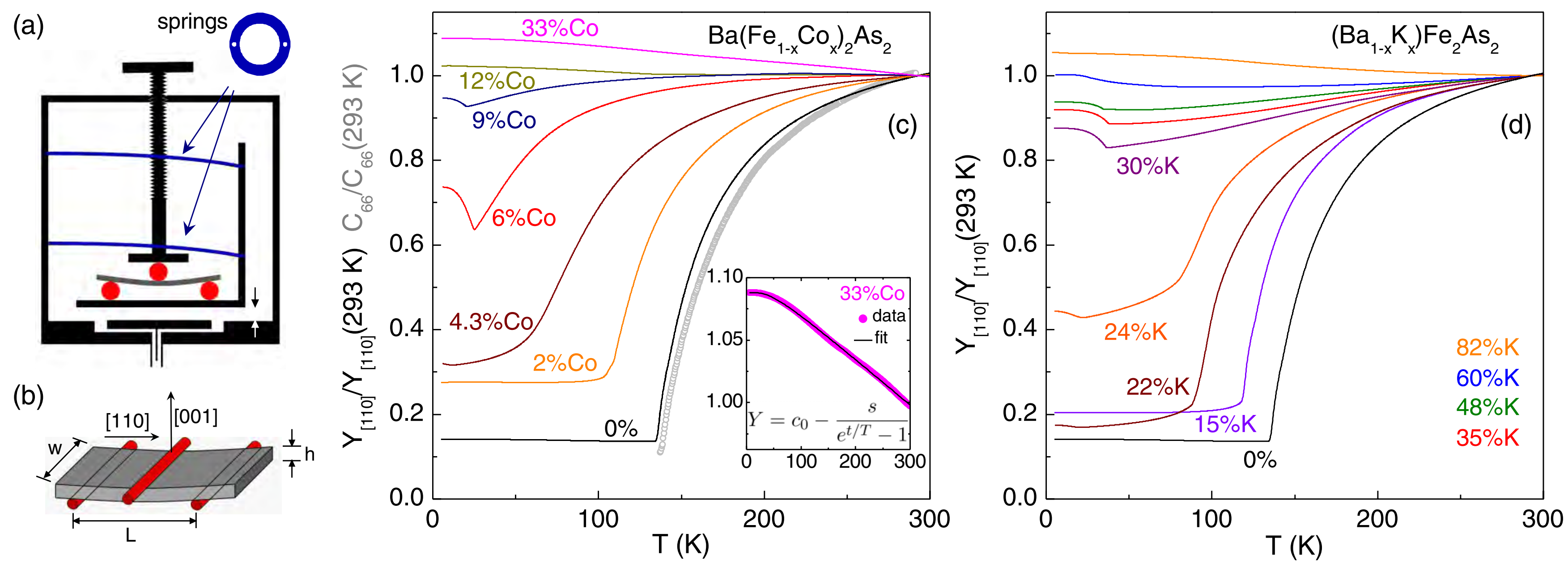
Strained free:

$$\frac{\partial F[\phi, \epsilon_6]}{\partial \epsilon_6} = 0 \quad \Rightarrow \quad \epsilon_6 = \frac{\lambda\phi}{C_{66,0}}$$

$$\Rightarrow \quad F = \frac{1}{2}a(T - T_s)\phi^2 + \frac{B}{4}\phi^4$$

$$\boxed{T_s = \bar{T}_s + \frac{\lambda^2}{aC_{66,0}}}$$

PRL **112,** 047001 (2014) PHYSICAL REVIEW LETTERS week ending 31 JANUARY 2014



$$C_{66} = \frac{d^2 F}{d\epsilon_6^2}$$

$$\frac{\partial F}{\partial \phi}[\phi(\epsilon), \epsilon] = 0$$

$$\frac{\partial F}{\partial \phi} = 0 \;\Rightarrow\; \frac{dF}{d\epsilon} = \frac{\partial F}{\partial \epsilon} \;\Rightarrow\; \frac{d^2 F}{d\epsilon^2} = \frac{\partial^2 F}{\partial \epsilon^2} + \frac{\partial^2 F}{\partial \phi \partial \epsilon}\frac{d\phi}{d\epsilon}$$

$$\frac{\partial F}{\partial \phi} = 0 \;\Rightarrow\; \frac{d\phi}{d\epsilon} = -\frac{\partial^2 F/(\partial \phi \partial \epsilon)}{\partial^2 F/\partial \phi^2}$$

Phenomenology (cont):

$$F[\phi, \epsilon_6] = \frac{1}{2}\chi_\phi^{-1}\phi^2 - \lambda\epsilon_6\phi + \frac{1}{2}C_{66,0}\epsilon_6^2 + \frac{B}{4}\phi^4$$

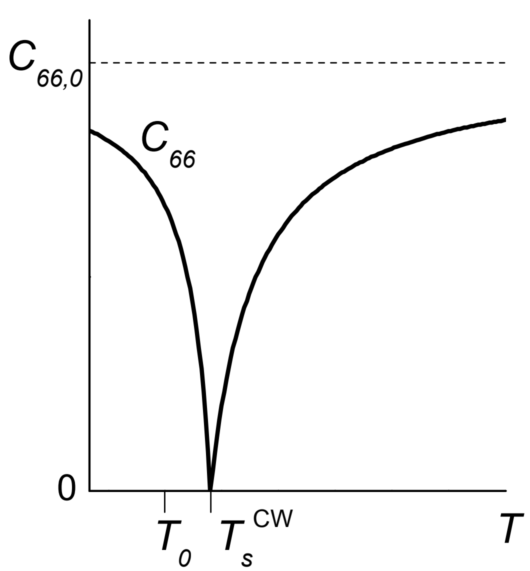
$$C_{66} = \frac{d^2F}{d\epsilon_6^2}$$

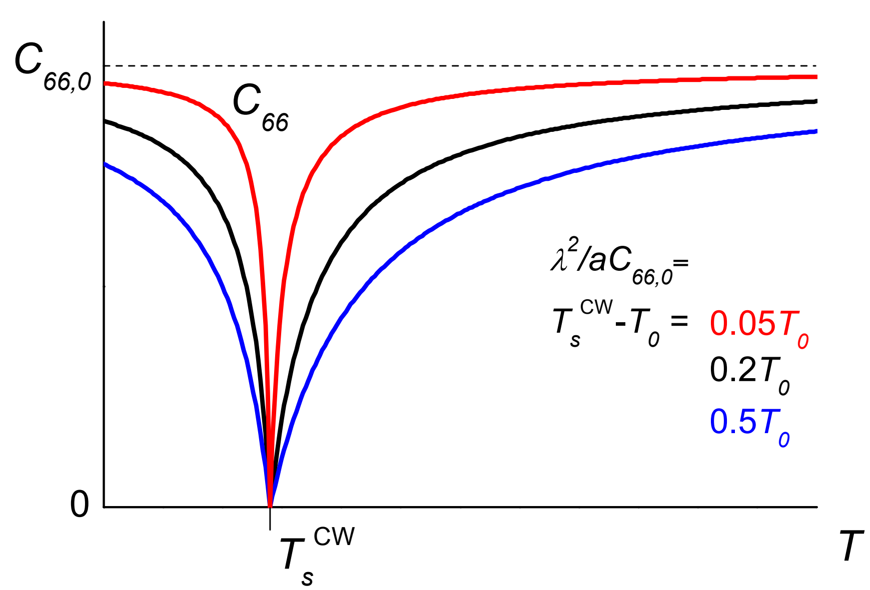
$$\chi_\phi^{-1} = a(T - \bar{T}_s)$$

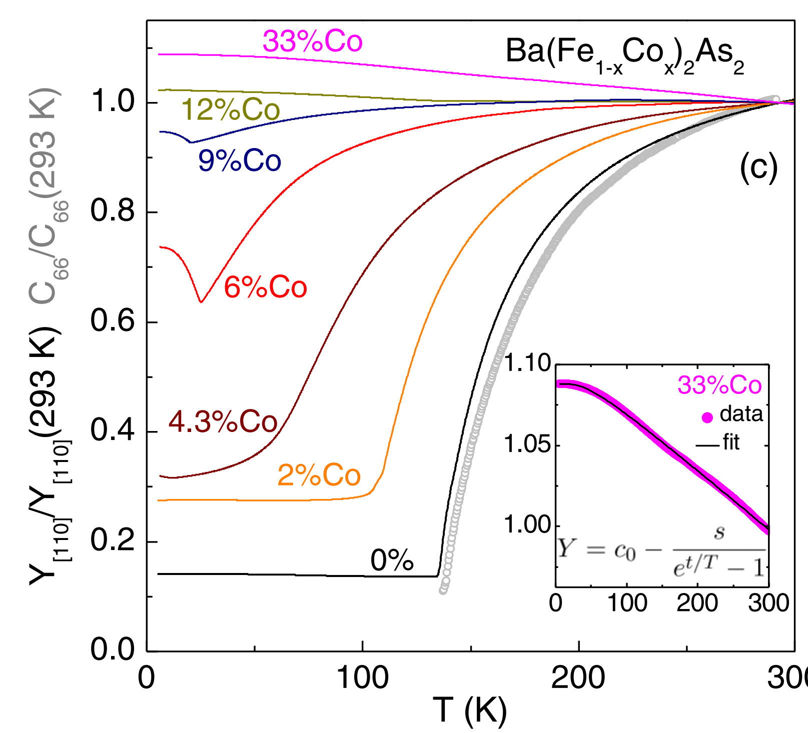
$$C_{66} = C_{66,0} + \frac{\lambda^2}{\chi_\phi^{-1} + 3B\phi^2}$$

$$C_{66} = C_{66,0}\frac{T - T_s}{T - \bar{T}_s}$$

Curie-Weiss







Anna E. Böhmer, Christoph Meingast, arXiv:1505.05120

Shear modulus, resistivity: static induced anisotropy

↓

Weak dynamic probe of ϕ is needed

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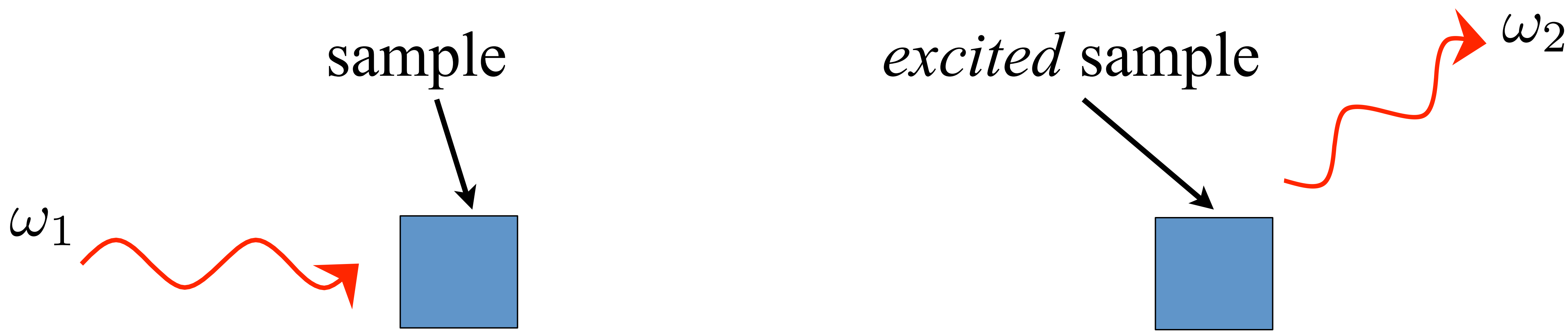
Raman spectroscopy

Raman: inelastic light scattering

Initial state | Final state



Measure:

$$\frac{d\sigma}{d\Omega d\omega} = \frac{\text{Flux of photons scattered into } d\Omega \text{ with } \omega < \omega_2 < \omega + d\omega}{\text{Flux of incident photons}}$$

$$\omega_2 - \omega_1 = \text{Excitation energy} = \text{Raman shift}$$

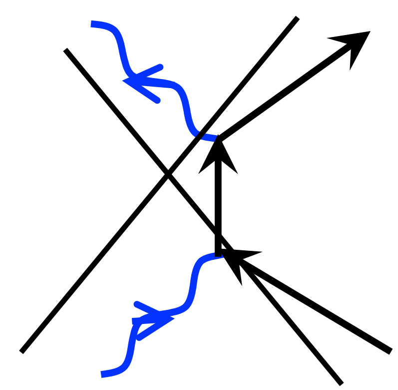
The case of electron plasma

P.M. Platzmann,
Phys. Rev. **139**, A379 (1965)

$$H = \sum_i \frac{1}{2m}\left[\boldsymbol{p}_i - \frac{e}{c}\boldsymbol{A}(\boldsymbol{r}_i, t)\right]^2 + H_{int}$$

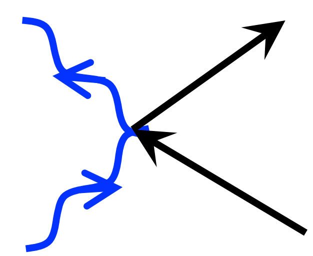
$$H = H_0 + V_1 + V_2$$

$$V_1 = -\frac{e}{mc}\sum_i \boldsymbol{p}_i \cdot \boldsymbol{A}(\boldsymbol{r}_i, t)$$



Stay off direct resonances:
Energy denominators!

$$V_2 = \frac{e^2}{2mc^2}\sum_i \boldsymbol{A}^2(\boldsymbol{r}_i, t)$$



$$r_0 = \frac{e^2}{mc^2}$$

The case of electron plasma (cont)

$$\hat{\rho}(\boldsymbol{r}) = \sum_i \delta(\boldsymbol{r} - \boldsymbol{r}_i)$$

$$V_2 = \frac{r_0}{2} \int d^d r \boldsymbol{A}^2(\boldsymbol{r}, t) \hat{\rho}(\boldsymbol{r})$$

Field Quantization: $A(\boldsymbol{r}, t) \rightarrow \hat{A}(\boldsymbol{r}, t)$

Photons: $\hbar\omega_k = c\hbar k$

Field Quantization(cont)

$$H_{EM} = \frac{1}{8\pi}\int d^3r(\boldsymbol{E}^2 + \boldsymbol{B}^2) = \sum_{\boldsymbol{k},\lambda} \hbar\omega_k \left(a^\dagger_{\boldsymbol{k}\lambda} a_{\boldsymbol{k}\lambda} + 1/2\right)$$

$$[a_{\boldsymbol{k}\lambda}, a^\dagger_{\boldsymbol{k}\lambda}] = 1$$

$$\hat{A}_\alpha(\boldsymbol{r},t) = \sum_{\boldsymbol{k}\lambda}\left(\frac{2\pi\hbar^2 c}{V\omega_k}\right)^{1/2} \boldsymbol{e}^\lambda_\alpha e^{i\boldsymbol{k}\boldsymbol{r}}\left(a_{\boldsymbol{k}\lambda}e^{-i\omega_k t} + a^\dagger_{-\boldsymbol{k}\lambda}e^{i\omega_k t}\right)$$

↓ One photon in a volume V

Initial state

$$|\psi_i\rangle = |i\rangle|\boldsymbol{k}_1\lambda_1\rangle$$

sample photons

$$\epsilon_i = E_i + \hbar\omega_{k_1}$$

Final state

$$|\psi_f\rangle = |f\rangle|\boldsymbol{k}_2\lambda_2\rangle$$

sample photons

$$\epsilon_f = E_f + \hbar\omega_{k_2}$$

Golden Rule: $$\Gamma_{f\leftarrow i} = \frac{2\pi}{\hbar}|\langle\psi_i|V_2|\psi_f\rangle|^2\delta(\epsilon_i - \epsilon_f)$$

Scattered flux: $$\frac{V}{(2\pi)^3}\sum_f k_2^2\, dk_2\, d\Omega\, \Gamma_{f\leftarrow i}$$

Incident flux: $$\frac{c}{V}$$

$$\rho_{\boldsymbol{q}} = \int d^3 r e^{-i\boldsymbol{q}\boldsymbol{r}} \hat{\rho}(\boldsymbol{r})$$

$$V_2 = \frac{r_0}{2} \frac{1}{V} \sum_{\boldsymbol{k}\boldsymbol{k}'} \sum_{\lambda\lambda'} \hat{\rho}_{\boldsymbol{k}+\boldsymbol{k}'} \frac{2\pi\hbar c^2}{\sqrt{\omega_k \omega_{k'}}} \sum_{\alpha} \boldsymbol{e}^{\alpha}_{k\lambda} \boldsymbol{e}^{\alpha}_{k'\lambda'} (a_{k\lambda} + a^{\dagger}_{-k\lambda})(a_{k'\lambda'} + a^{\dagger}_{-k'\lambda'})$$

Photon part of $\langle \psi_i, \boldsymbol{k}_1 \lambda | V_2 | \psi_f, \boldsymbol{k}_1 \rangle$ ⇒

$\boldsymbol{k}' = \boldsymbol{k}_1,\ \lambda' = \lambda_1$ $\qquad$ $\boldsymbol{k} = -\boldsymbol{k}_2, \lambda = \lambda_2$

$\boldsymbol{k}' = -\boldsymbol{k}_2, \lambda = \lambda_2$ $\qquad$ $\boldsymbol{k}' = \boldsymbol{k}_1, \lambda = \lambda_1$

P.M. Platzmann,
Phys. Rev. **139**, A379 (1965)

$$\Gamma_{f\leftarrow i} = \frac{1}{V^2}\frac{2\pi}{\hbar} r_0^2 \frac{(2\pi\hbar c^2)^2}{\omega_{k1}\omega_{k2}} |\langle f|\hat{\rho}_{\boldsymbol{k}_1-\boldsymbol{k}_2}|i\rangle|^2 \left|\sum_\alpha \boldsymbol{e}^\alpha_{\lambda_1}\boldsymbol{e}^\alpha_{\lambda_2}\right|^2 \delta(E_f - E_i - (\omega_2-\omega_1))$$

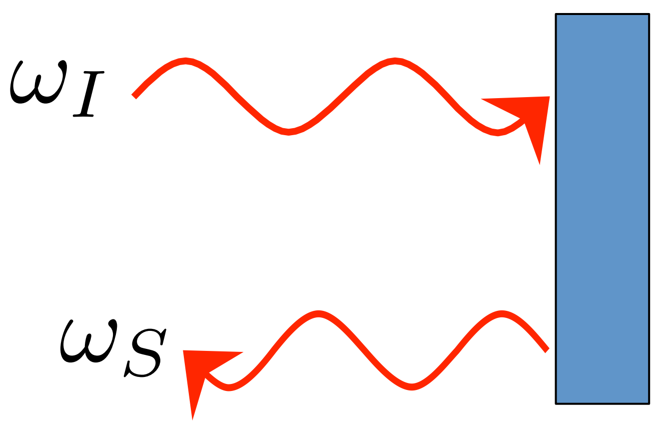
↓

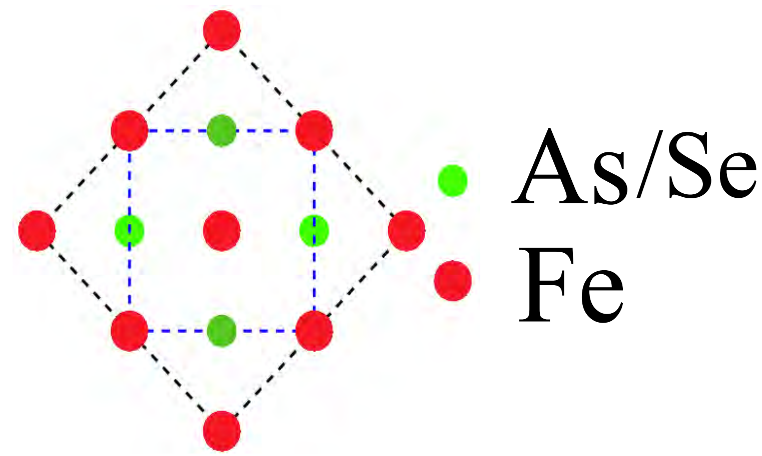
$$\frac{d\sigma}{d\Omega d\hbar\omega_2} = r_0^2 \frac{\omega_2}{\omega_1} \left|\sum_\alpha \boldsymbol{e}^\alpha_{\lambda_1}\boldsymbol{e}^\alpha_{\lambda_2}\right|^2 \underbrace{\sum_f |\langle f|\hat{\rho}_{\boldsymbol{k}-\boldsymbol{k}'}|i\rangle|^2 \delta(E_f - E_i - (\omega_2-\omega_1))}_{\text{Dynamical Structure Factor}}$$

$$\frac{d\sigma}{d\Omega d\hbar\omega_2} = \frac{r_0^2}{2\pi}\frac{\omega_2}{\omega_1}\frac{1}{1-e^{-\omega/T}}\chi''(\boldsymbol{k}_1-\boldsymbol{k}_2, \omega_2-\omega_1)$$

$$\chi(\boldsymbol{q},\omega) = -i\int_0^\infty dt e^{i\omega t}[\rho_{-\boldsymbol{q}}(t), \rho_{\boldsymbol{q}}(0)]$$ Raman susceptibility

Experiment: Quasi-backscattering geometry





Layered materials: $\boldsymbol{q} = 0$ $\rho_{\boldsymbol{q}}(t) = \rho_{\boldsymbol{q}}(0)$ $\chi(0, \omega \neq 0) = 0$

No Raman !?!?....

Raman coupling is different in multi-band/multi-orbital materials!

Shear modulus, resistivity: static induced anisotropy

↓

Weak dynamic probe of ϕ is needed

↓

Raman spectroscopy

↓

m^{-1} too symmetric to couple to anything less trivial than density

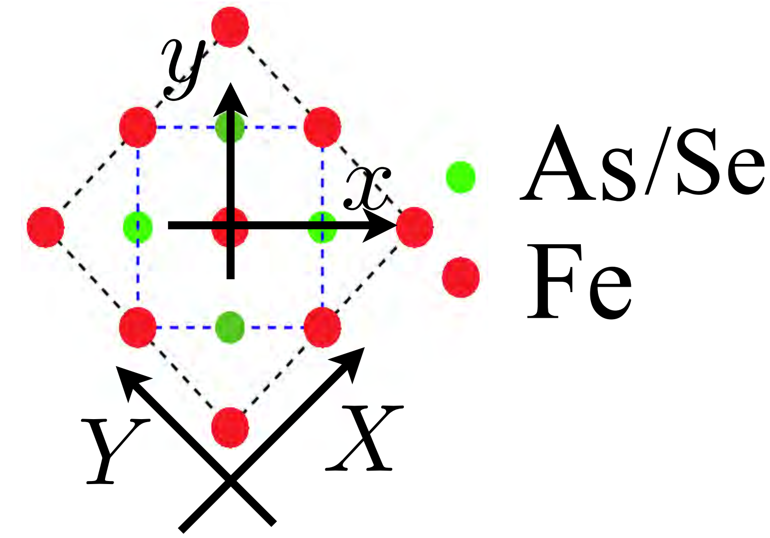
↓

What is the symmetry of ϕ? Does the lattice help?

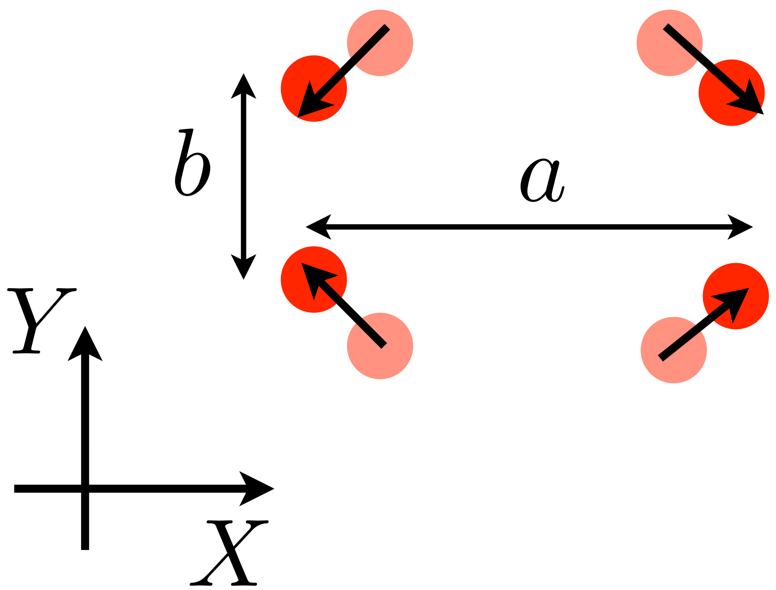
↓

$F \sim -\lambda \epsilon_6 \phi$ scalar → Symmetry of ϵ_6 ?

Structural transition



Orthorhombic distortion



$a \neq b$ anisotropy

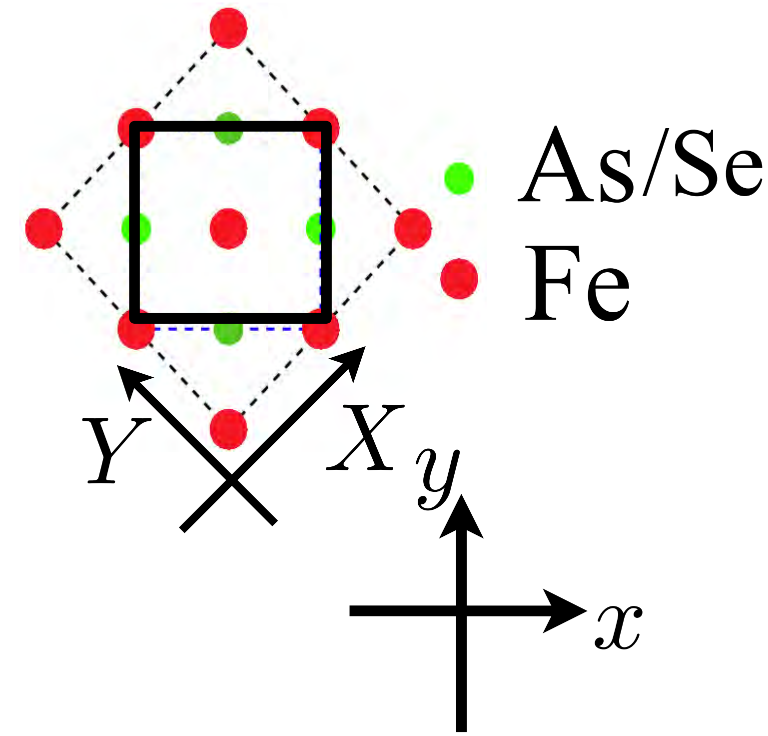
Discrete $C_4 \rightarrow C_2$ symmetry breaking

$$\epsilon_6 = \frac{\partial u_X}{\partial X} - \frac{\partial u_Y}{\partial Y} \neq 0$$

Order Parameter

Structural transition: order parameter symmetry

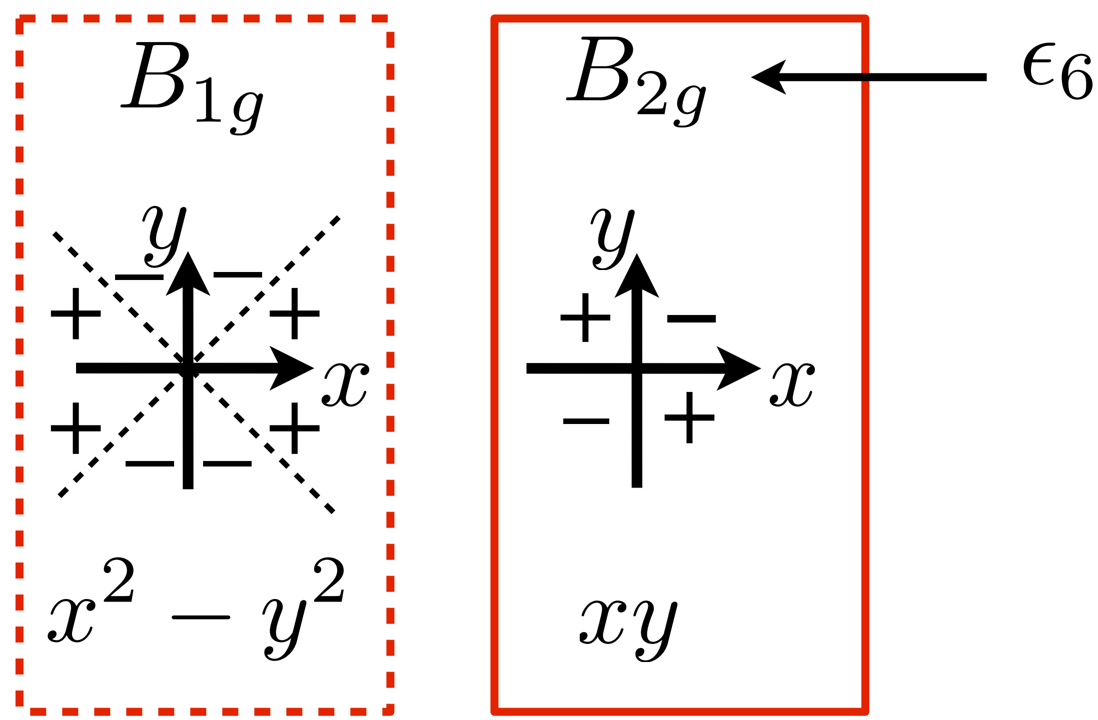
$$\epsilon_6 = \frac{\partial u_X}{\partial X} - \frac{\partial u_Y}{\partial Y} \sim X^2 - Y^2 = 2xy$$



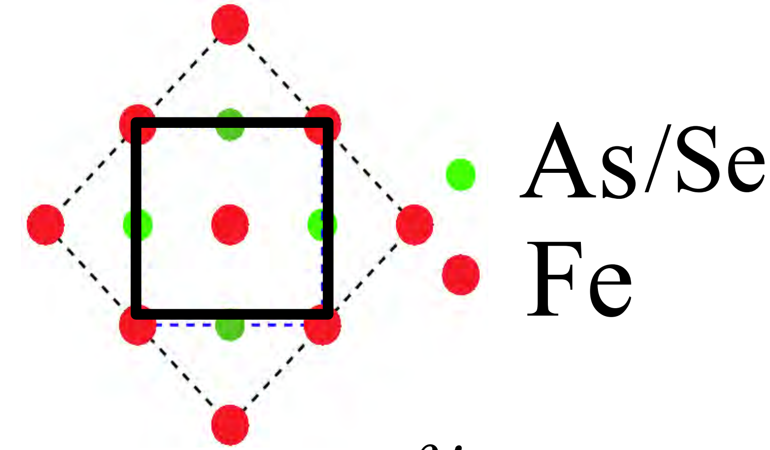
$$D_{4h} = D_4 \otimes I = 8 * 2 = 16$$

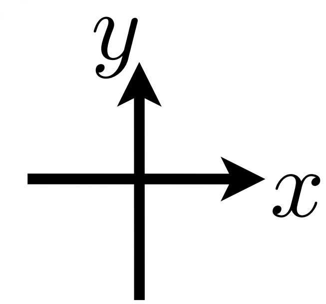
A_{1g}

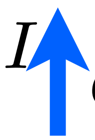
Fully symmetric (scalar)

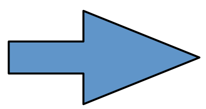


ϵ_6 and $\phi \sim xy = B_{2g}$



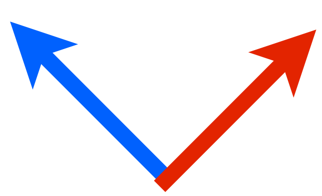


Raman? $\boldsymbol{e}^I$  $\boldsymbol{e}^S$ 

Tensor  $e^I_\alpha e^S_\beta$

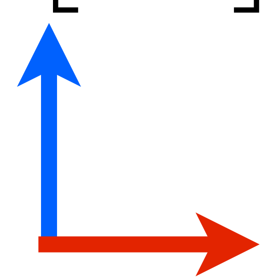
B_{1g}

$$\begin{bmatrix} 1 & 0 \\ 0 & -1 \end{bmatrix}$$



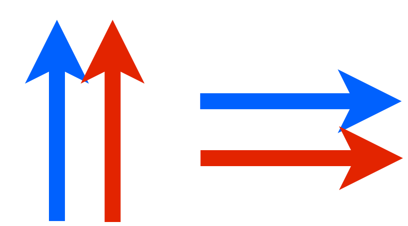
$B_{2g} \sim xy$

$$\begin{bmatrix} 0 & 1 \\ 1 & 0 \end{bmatrix}$$

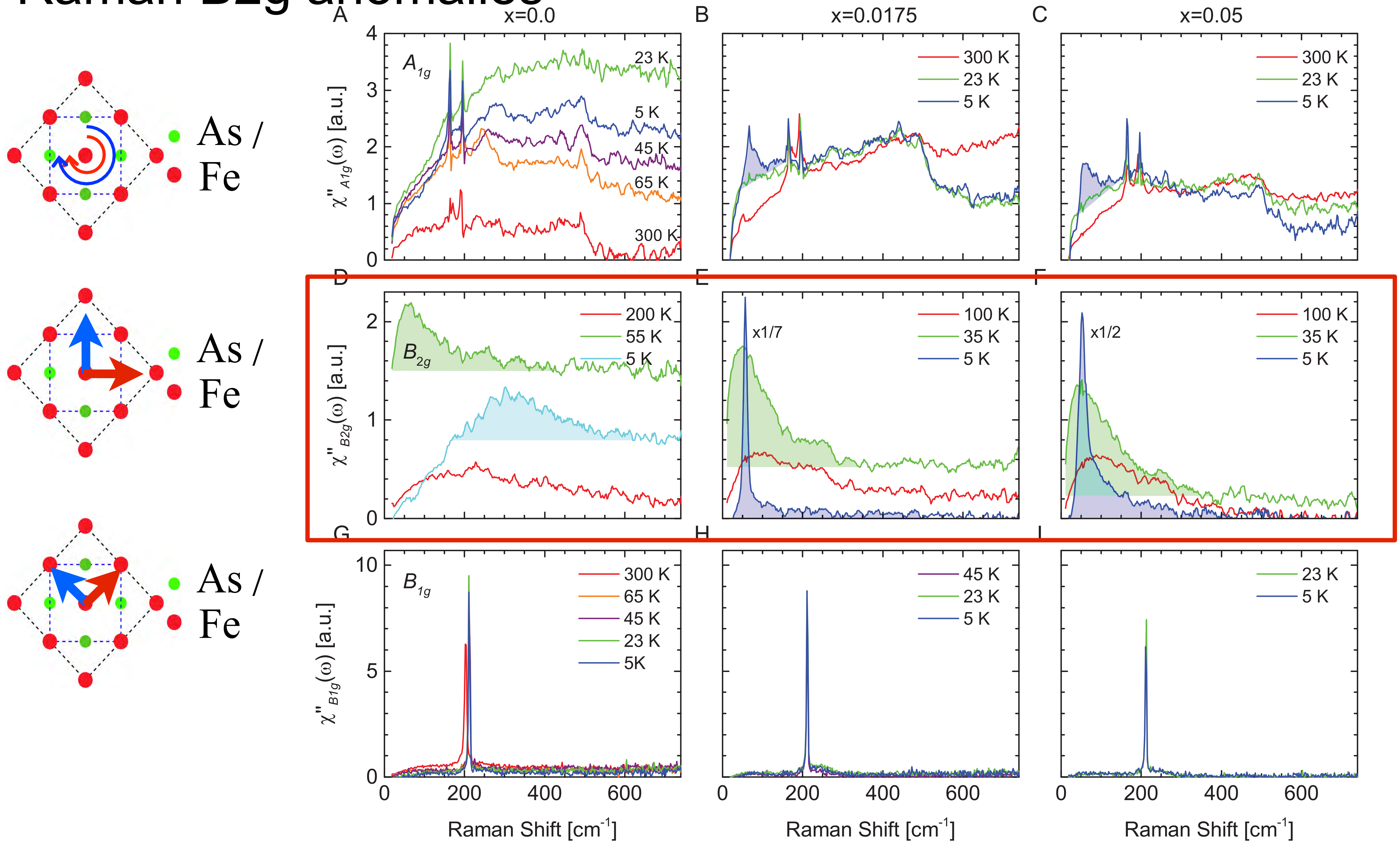


A_{1g}

$$\begin{bmatrix} 1 & 0 \\ 0 & 1 \end{bmatrix}$$



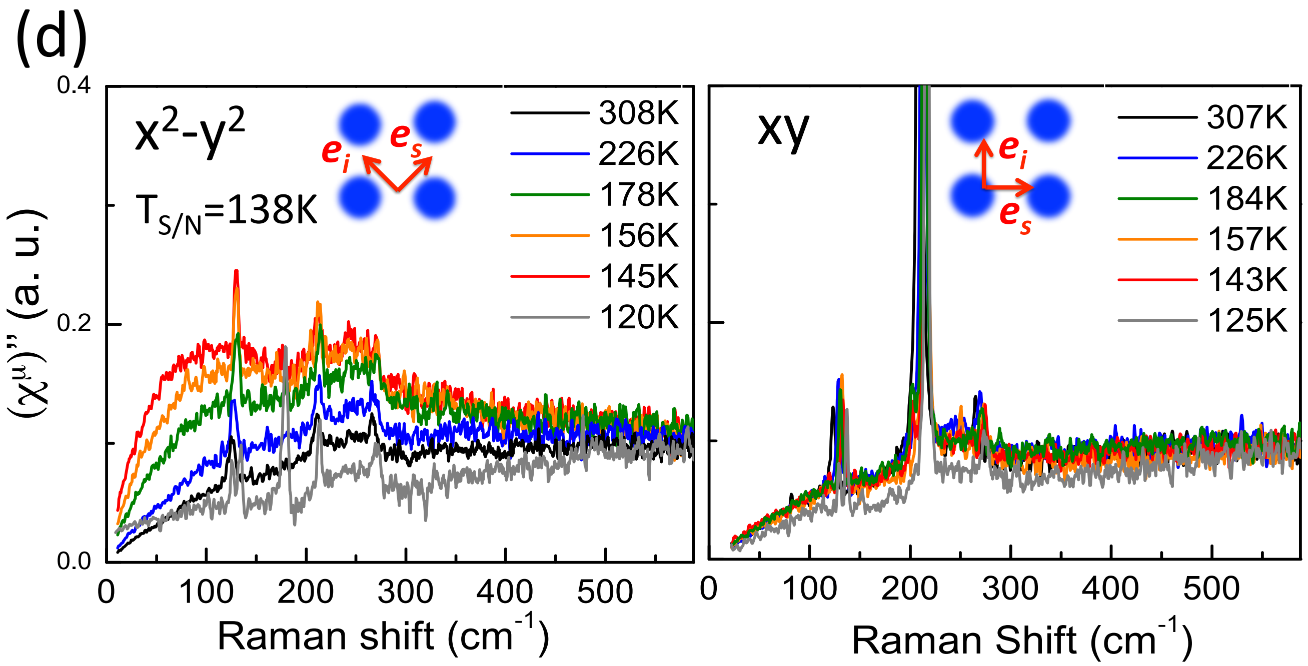
Raman B2g anomalies



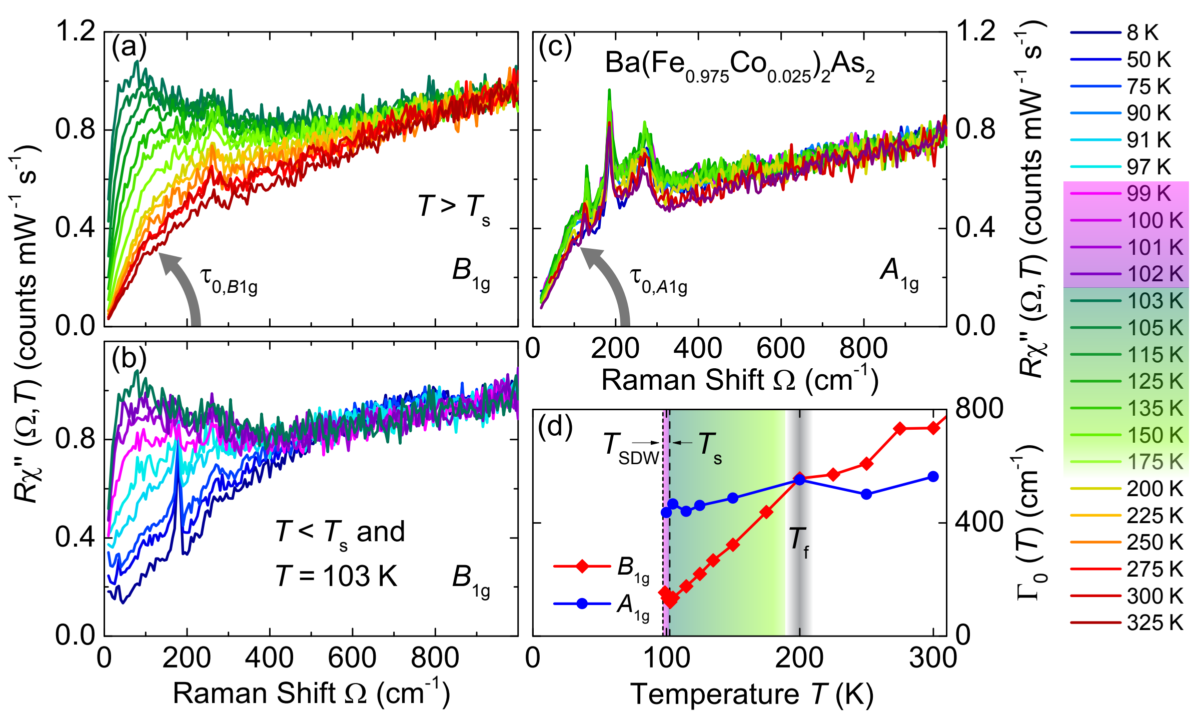
arXiv:1410.6456

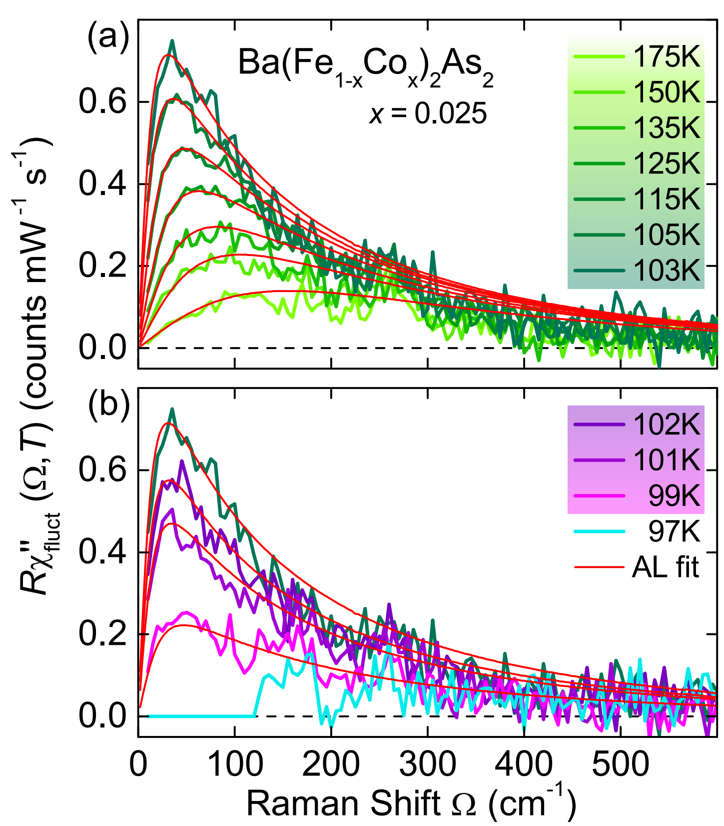
Critical Charge Fluctuations in Iron Pnictide Superconductors

V. K. Thorsmølle, MK, Z. P. Yin, Chenglin Zhang, S. V. Carr, Pengcheng Dai, G. Blumberg



Y. Gallais, R. M. Fernandes, I. Paul, L. Chauviere, Y.-X. Yang, M.-A. Measson, M. Cazayous, A. Sacuto, D. Colson, A. Forget, Phys. Rev. Lett. 111, 267001 (2013)



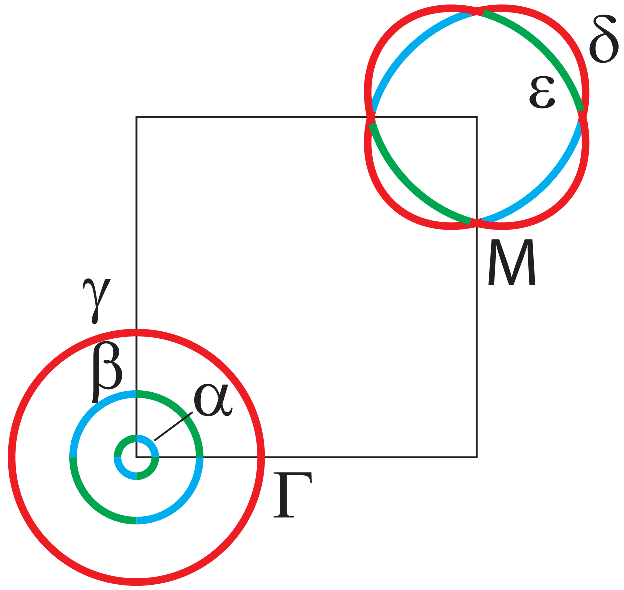


arXiv:1507.06116 Florian Kretzschmar, Thomas Böhm, Una Karahasanović, Bernhard Muschler, Andreas Baum, Daniel Jost, Joerg Schmalian, Sergio Caprara, Marco Grilli, Carlo Di Castro, James G. Analytis, Jiun-Haw Chu, Ian Randal Fisher, Rudi Hackl

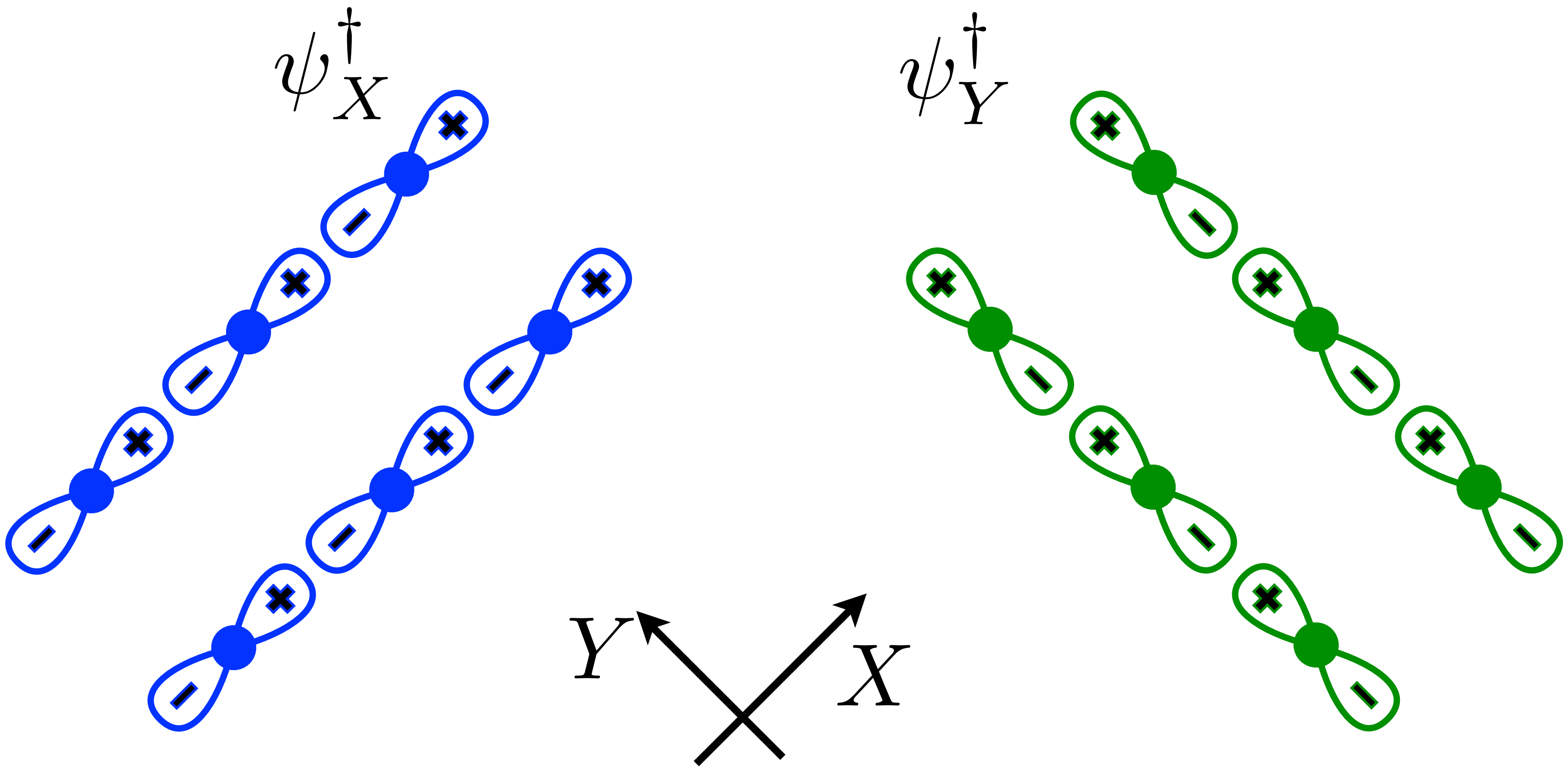
What does Raman see?

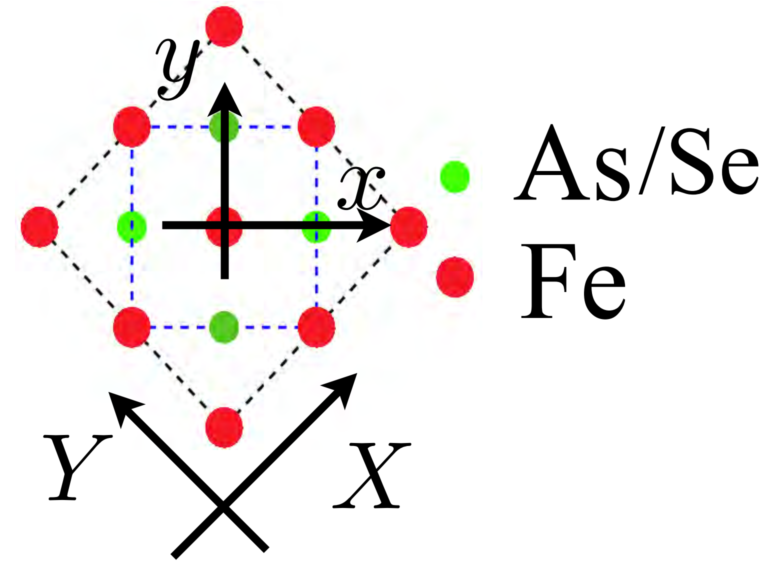
Examine band structure

Focus on Γ point $\alpha\beta$ bands



At Γ double degeneracy (no spin-orbit)

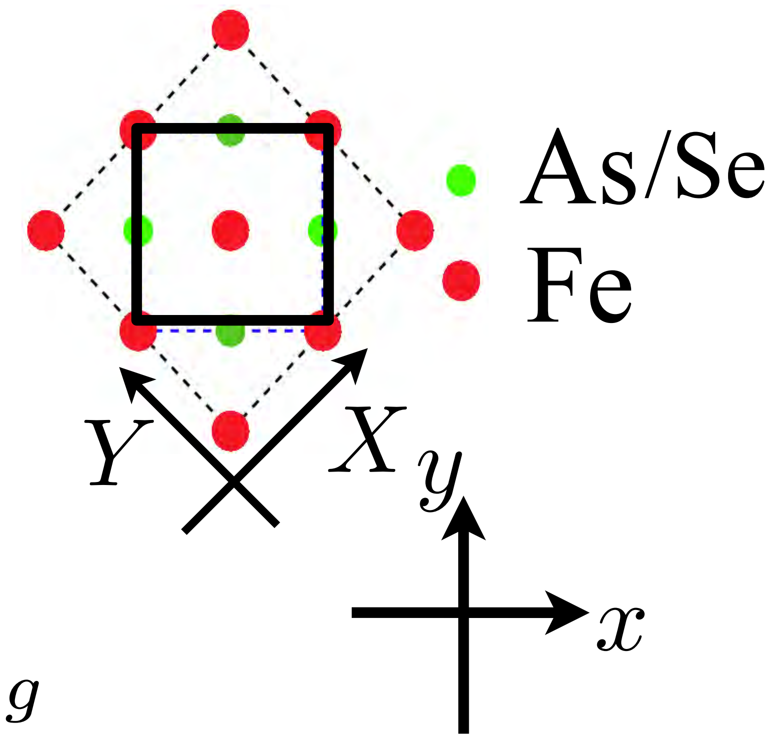




Iron only lattice

Slightly off Γ point

Method of invariants (Luttinger) $H \in A_{1g}$

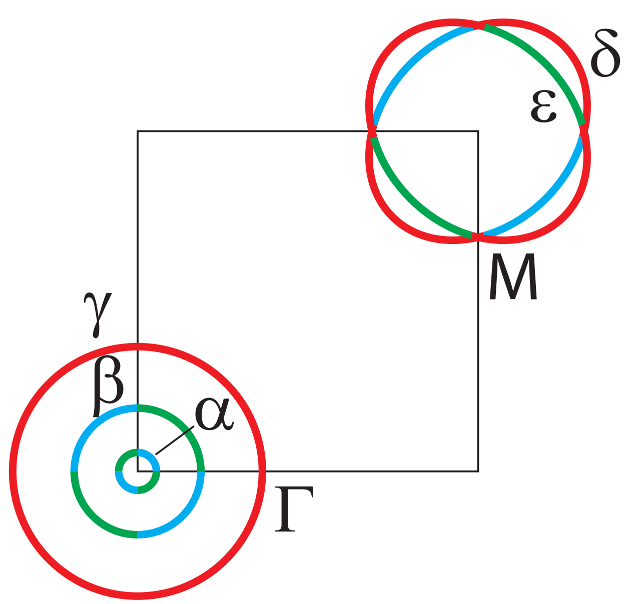


Symmetry allowed terms:

$$\sim (k_x^2 + k_y^2)(\psi_X^\dagger \psi_X + \psi_Y^\dagger \psi_Y) \qquad A_{1g} * A_{1g}$$

$$\sim (k_x^2 - k_y^2)(\psi_X^\dagger \psi_Y + \psi_Y^\dagger \psi_X) \qquad B_{1g} * B_{1g}$$

$$\sim k_x k_y (\psi_X^\dagger \psi_X - \psi_Y^\dagger \psi_Y) \qquad B_{2g} * B_{2g}$$

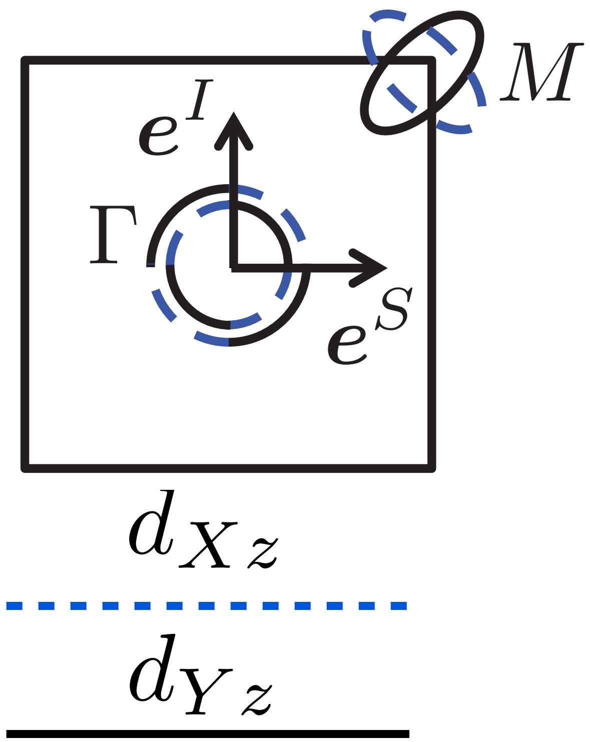


Holes Γ

$$\mathcal{H}_{\boldsymbol{k}}^{\Gamma} = \begin{bmatrix} \epsilon_\Gamma + \frac{k^2}{2m_\Gamma} + a k_x k_y & \frac{c}{2}(k_x^2 - k_y^2) \\ \frac{c}{2}(k_x^2 - k_y^2) & \epsilon_\Gamma + \frac{k^2}{2m_\Gamma} - a k_x k_y \end{bmatrix}$$

V. Cvetkovic and O. Vafek, Phys. Rev. B **88**, 134510 (2013)

What does B2g Raman see?



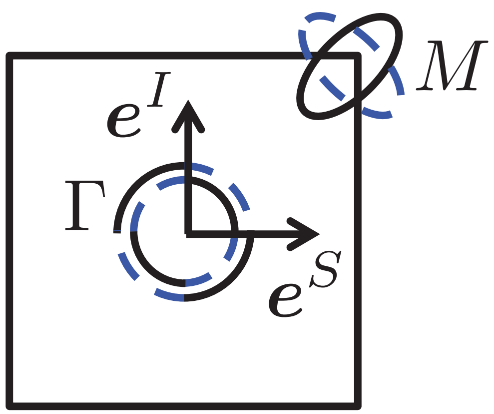
$$\mathcal{H}_{\boldsymbol{k}}^{\Gamma} = \begin{bmatrix} \epsilon_\Gamma + \frac{k^2}{2m_\Gamma} + a k_x k_y & \frac{c}{2}(k_x^2 - k_y^2) \\ \frac{c}{2}(k_x^2 - k_y^2) & \epsilon_\Gamma + \frac{k^2}{2m_\Gamma} - a k_x k_y \end{bmatrix}$$

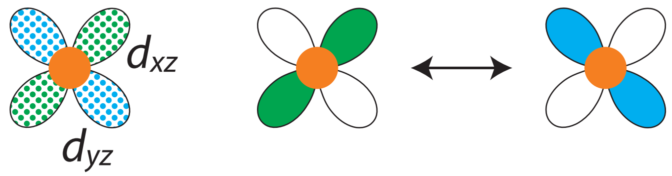
$$\boldsymbol{k} \to \boldsymbol{k} + \boldsymbol{A} \quad \Rightarrow \quad r_{i,j}^{\Gamma(M)} = \sum_{\lambda\lambda'} e_\lambda^I e_{\lambda'}^S \frac{\partial^2 \mathcal{H}_{ij}^{\Gamma(M)}}{\partial k_\lambda \partial k_{\lambda'}} .$$

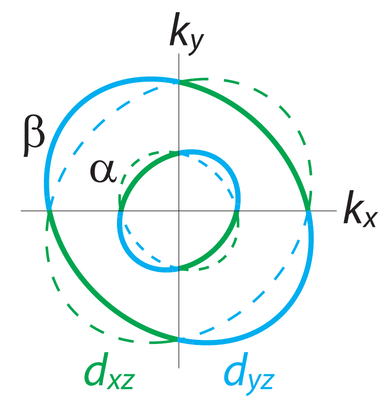
$$r^\Gamma = a \begin{bmatrix} 1 & 0 \\ 0 & -1 \end{bmatrix} \quad \Leftrightarrow \quad \hat{r}^\Gamma \propto \psi_X^\dagger \psi_X - \psi_Y^\dagger \psi_Y$$

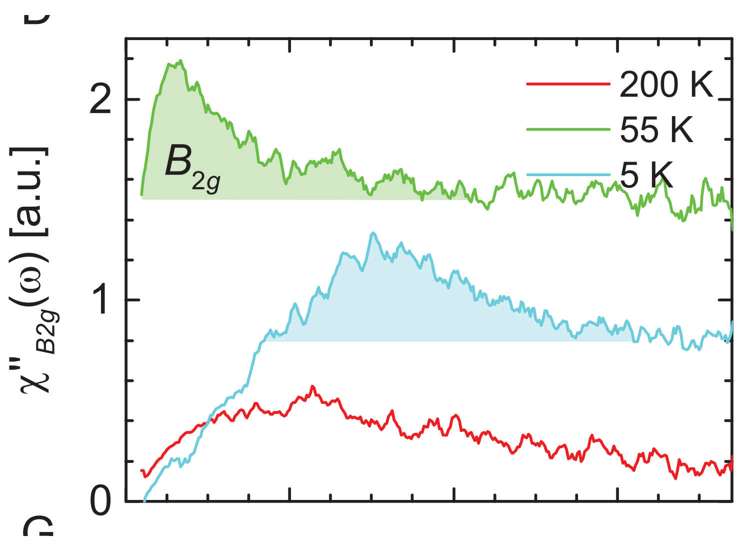
B2g Raman (cont)

$$r^{\Gamma} = a \begin{bmatrix} 1 & 0 \\ 0 & -1 \end{bmatrix} \Longleftrightarrow \hat{r}^{\Gamma} \propto \psi_X^\dagger \psi_X - \psi_Y^\dagger \psi_Y = n_{Xz} - n_{Yz}$$







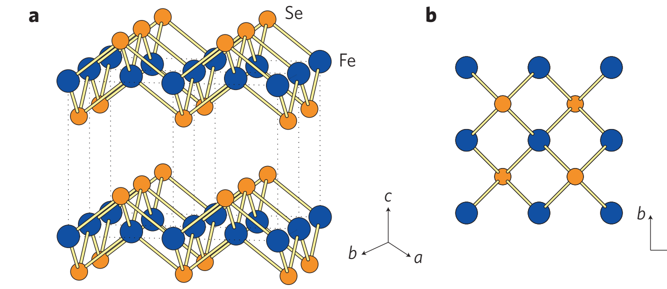


Tendency to orbital ordering

nature materials

ARTICLES

PUBLISHED ONLINE: 10 NOVEMBER 2014 | DOI: 10.1038/NMAT4138

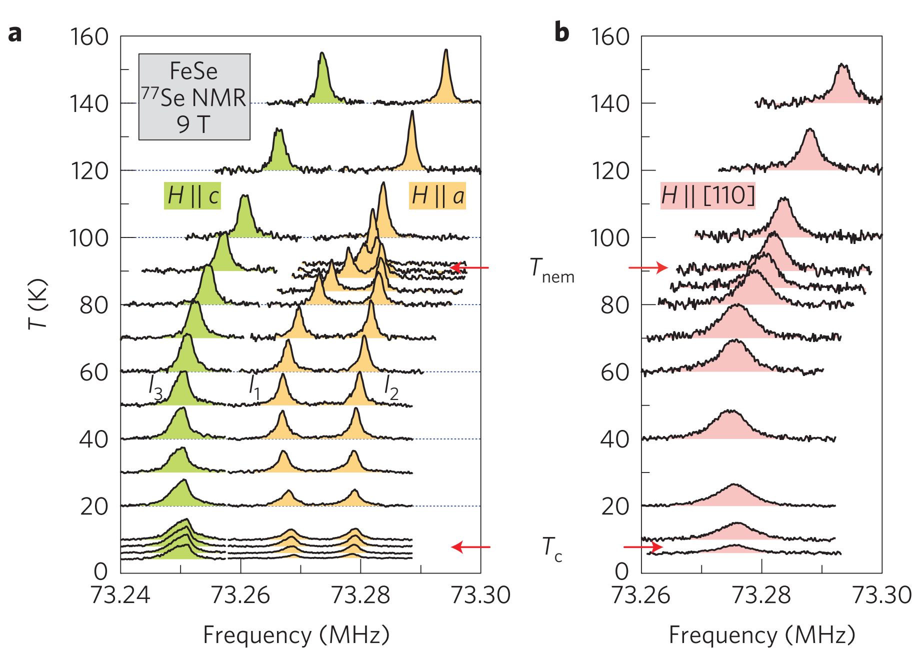


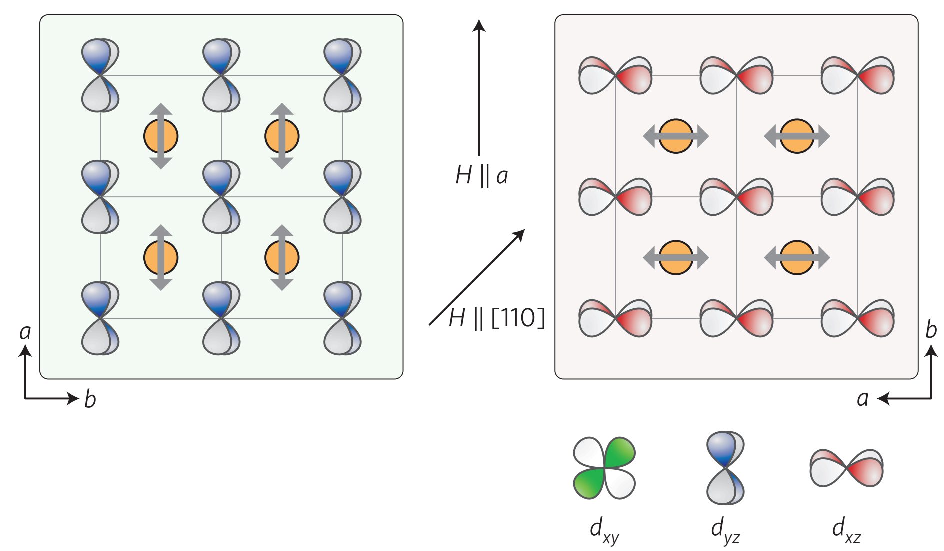
Orbital-driven nematicity in FeSe

S-H. Baek[1]*, D. V. Efremov[1], J. M. Ok[2], J. S. Kim[2], Jeroen van den Brink[1,3] and B. Büchner[1,3]

A fundamental and unconventional characteristic of superconductivity in iron-based materials is that it occurs in the vicinity of two other instabilities. In addition to a tendency towards magnetic order, these Fe-based systems have a propensity for nematic ordering: a lowering of the rotational symmetry while time-reversal invariance is preserved. Setting the stage for superconductivity, it is heavily debated whether the nematic symmetry breaking is driven by lattice, orbital or spin degrees of freedom. Here, we report a very clear splitting of NMR resonance lines in FeSe at $T_{nem} = 91$ K, far above the superconducting T_c of 9.3 K. The splitting occurs for magnetic fields perpendicular to the Fe planes and has the temperature dependence of a Landau-type order parameter. Spin-lattice relaxation rates are not affected at T_{nem}, which unequivocally establishes orbital degrees of freedom as driving the nematic order. We demonstrate that superconductivity competes with the emerging nematicity.

$$\frac{1}{T_1 T} \sim \sum_{\mathbf{q}} |\gamma_n A(\mathbf{q})|^2 \frac{\mathrm{Im}\,\chi(\mathbf{q}, f)}{f}$$

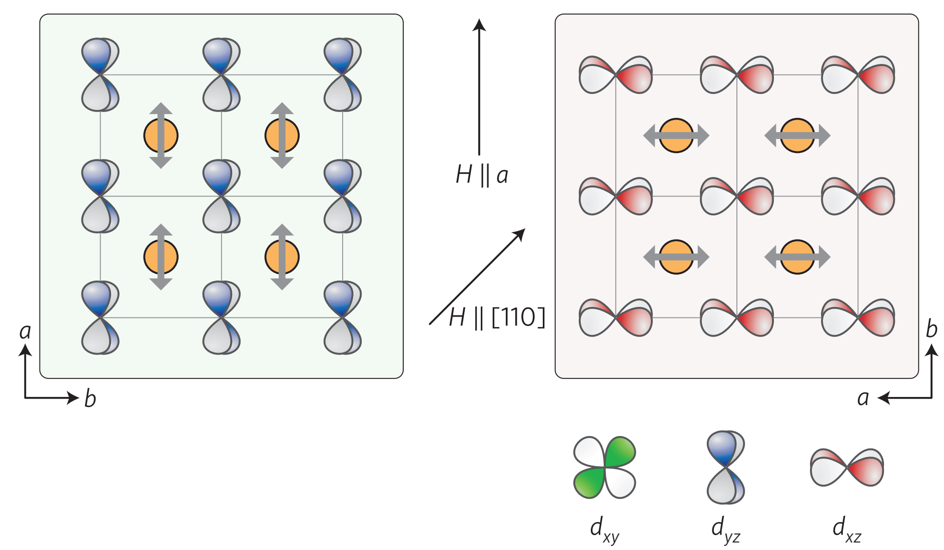




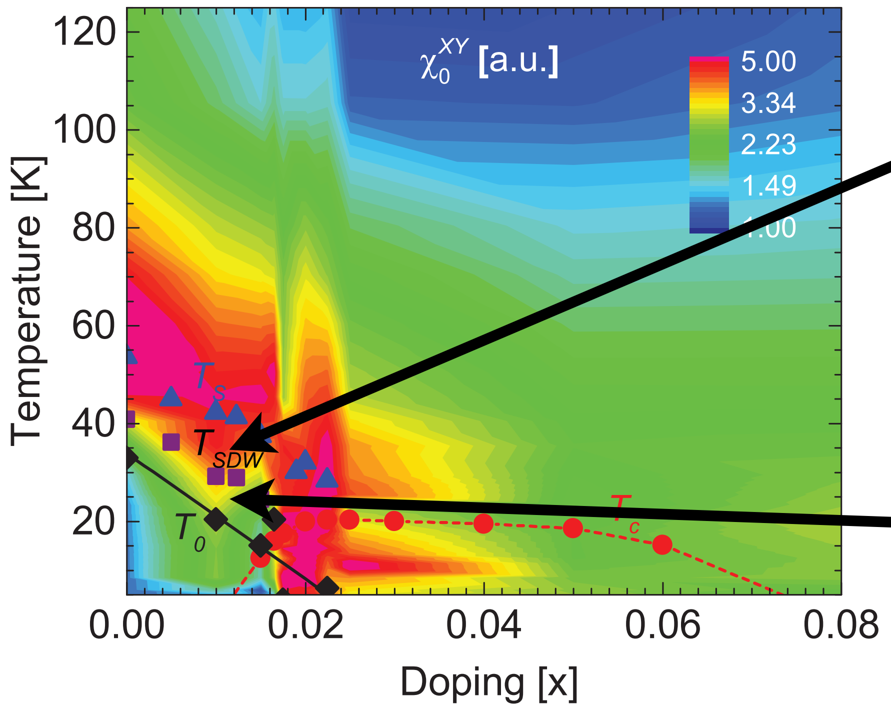
Orbital order: C4 to C2 discrete symmetry breaking

Ising nematic transition: difference in orbital occupation

$$n_{Xz} - n_{Yz} \neq 0$$



Alternative: *Spin* Ising Nematic Transition

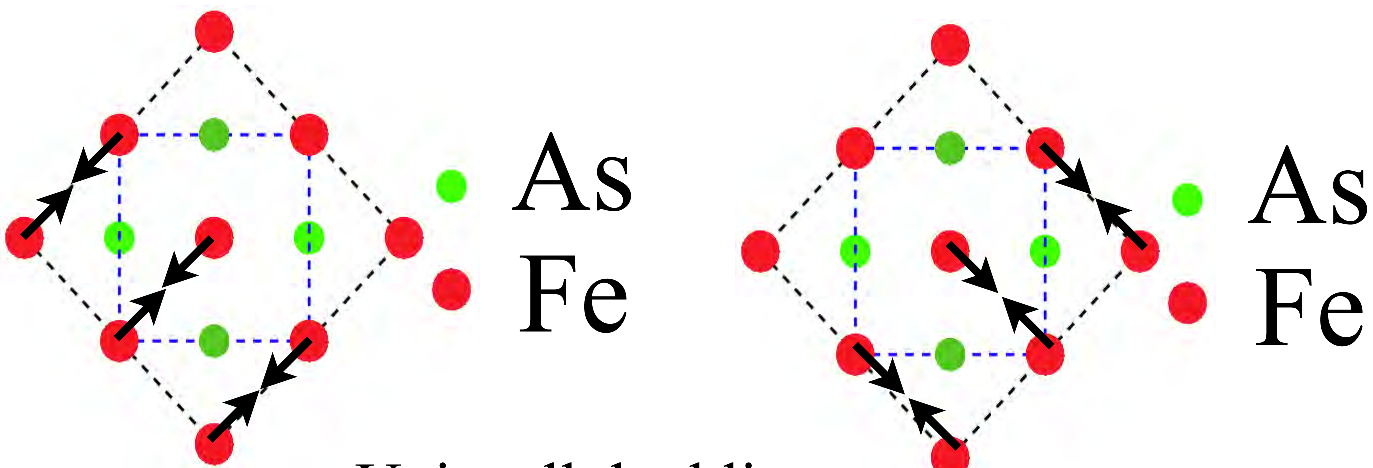


$$T_{SDW} < T < T_S$$

$$\langle \boldsymbol{\Delta}_X^2 \rangle - \langle \boldsymbol{\Delta}_Y^2 \rangle \neq 0$$

Stripe magnetizations $\langle \boldsymbol{\Delta}_{X,Y} \rangle = 0$

$$T < T_{SDW}$$



Unit cell doubling

R. M. Fernandes, A. V. Chubukov, J. Knolle, I. Eremin, and J. Schmalian, Phys. Rev. B, **85**, 024534 (2012).

nature physics

REVIEW ARTICLE

PUBLISHED ONLINE: 31 JANUARY 2014 | DOI: 10.1038/NPHYS2877

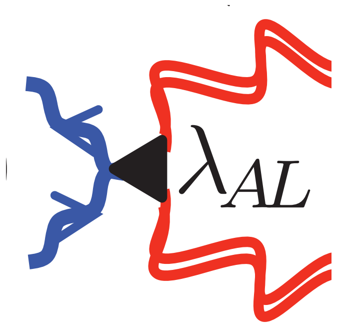
What drives nematic order in iron-based superconductors?

R. M. Fernandes[1]*, A. V. Chubukov[2]* and J. Schmalian[3]*

Although the existence of nematic order in iron-based superconductors is now a well-established experimental fact, its origin remains controversial. Nematic order breaks the discrete lattice rotational symmetry by making the *x* and *y* directions in the iron plane non-equivalent. This can happen because of a regular structural transition or as the result of an electronically driven instability — in particular, orbital order or spin-driven Ising-nematic order. The latter is a magnetic state that breaks rotational symmetry but preserves time-reversal symmetry. Symmetry dictates that the development of one of these orders immediately induces the other two, making the origin of nematicity a physics realization of the 'chicken and egg problem'. In this Review, we argue that the evidence strongly points to an electronic mechanism of nematicity, placing nematic order in the class of correlation-driven electronic instabilities, like superconductivity and density-wave transitions. We discuss different microscopic models for nematicity and link them to the properties of the magnetic and superconducting states, providing a unified perspective on the phase diagram of the iron pnictides.

2 electronic alternatives are not independent

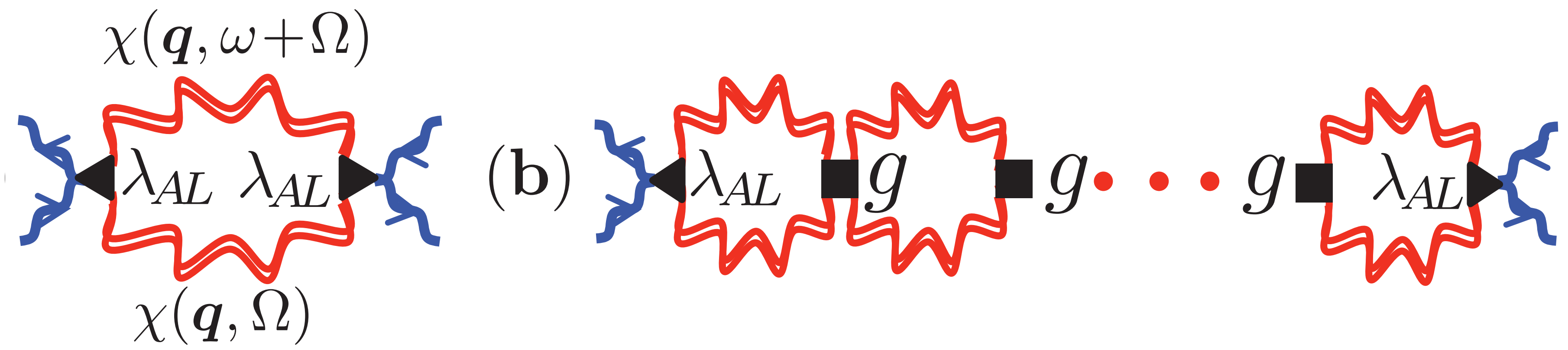
$$n_{Xz} - n_{Yz}$$



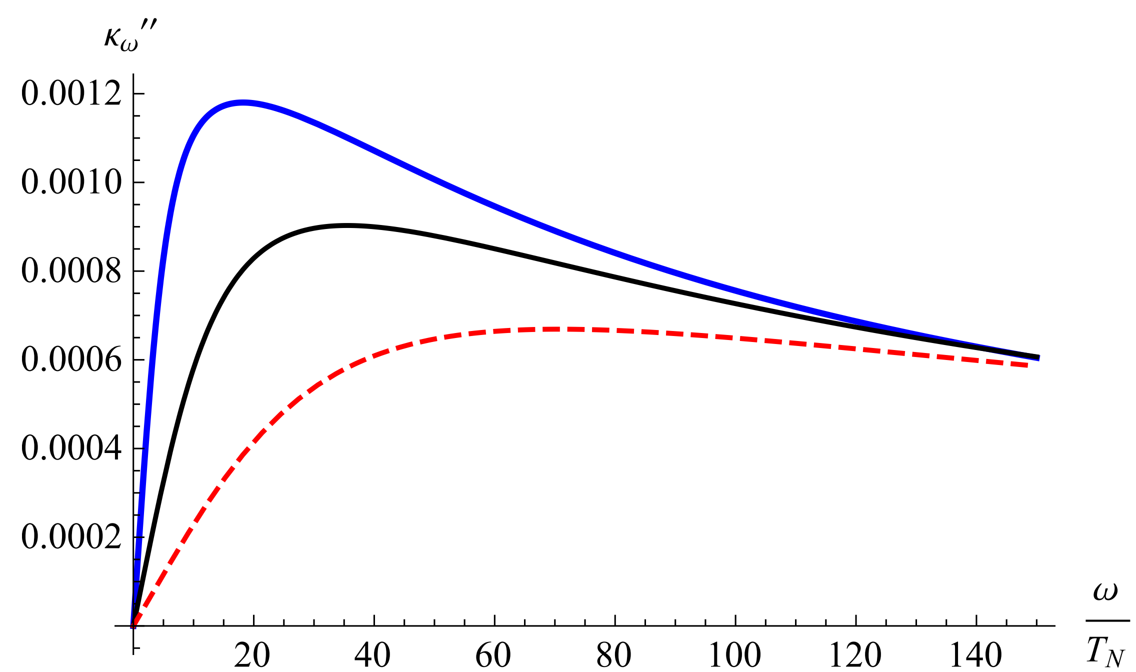
$$|\Delta_X|^2 - |\Delta_Y|^2$$

Raman correlations in magnetic scenario

$$S_{\text{eff}}\left[\mathbf{\Delta}_X, \mathbf{\Delta}_Y\right] = r_0\left(\mathbf{\Delta}_X^2 + \mathbf{\Delta}_Y^2\right) + \frac{u}{2}\left(\mathbf{\Delta}_X^2 + \mathbf{\Delta}_Y^2\right)^2 - \frac{g}{2}\left(\mathbf{\Delta}_X^2 - \mathbf{\Delta}_Y^2\right)^2 + v\left(\mathbf{\Delta}_X \cdot \mathbf{\Delta}_Y\right)^2$$



Quasi-Elastic Scattering
The case of critical slow-down



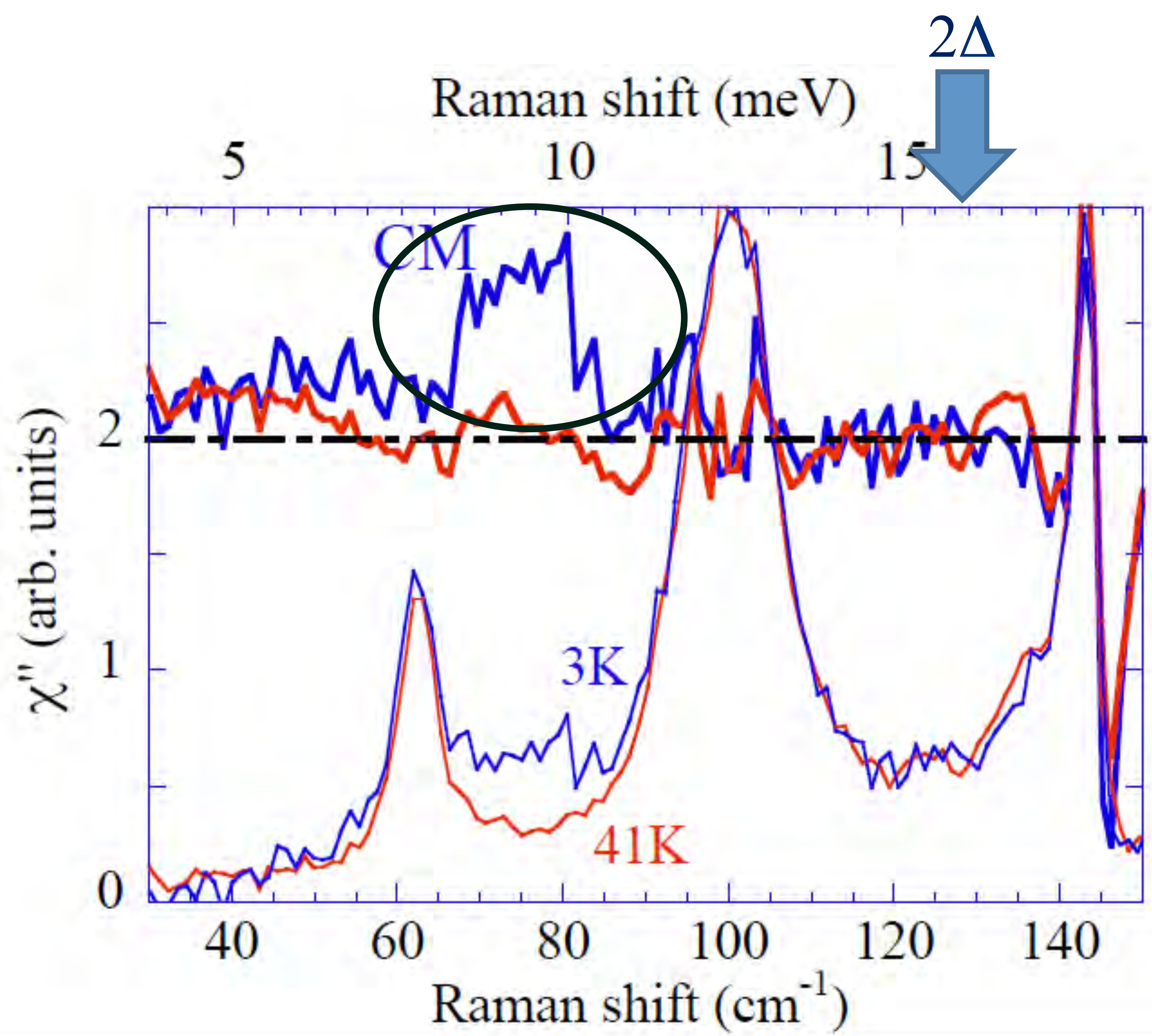
MK and A. Levchenko, Phys.Rev. B **91**, 235119 (2015)

arXiv:1504.06841
Una Karahasanovic, Florian Kretzschmar, Thomas Boehm, Rudi Hackl, Indranil Paul, Yann Gallais, Joerg Schmalian

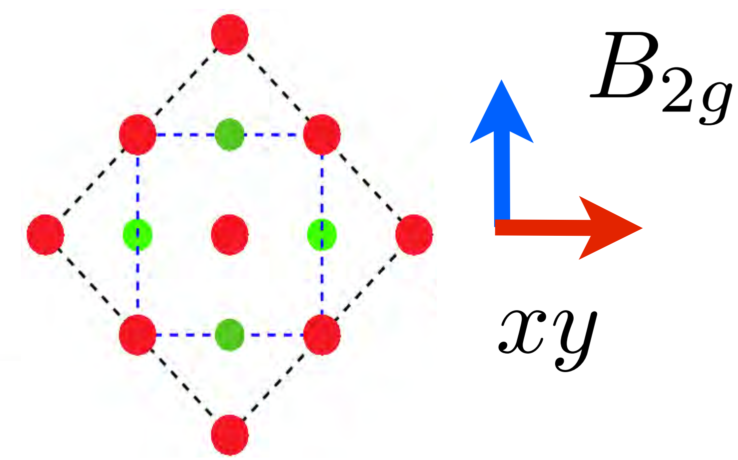
Part II

Raman in Iron Selenides

A. Ignatov, A. Kumar, P. Lubik, R. H. Yuan, W. T. Guo, N. L. Wang, K. Rabe, and G. Blumberg,
Phys. Rev. B **86**, 134107 (2012)



$K_{0.75}Fe_{1.75}Se_2$



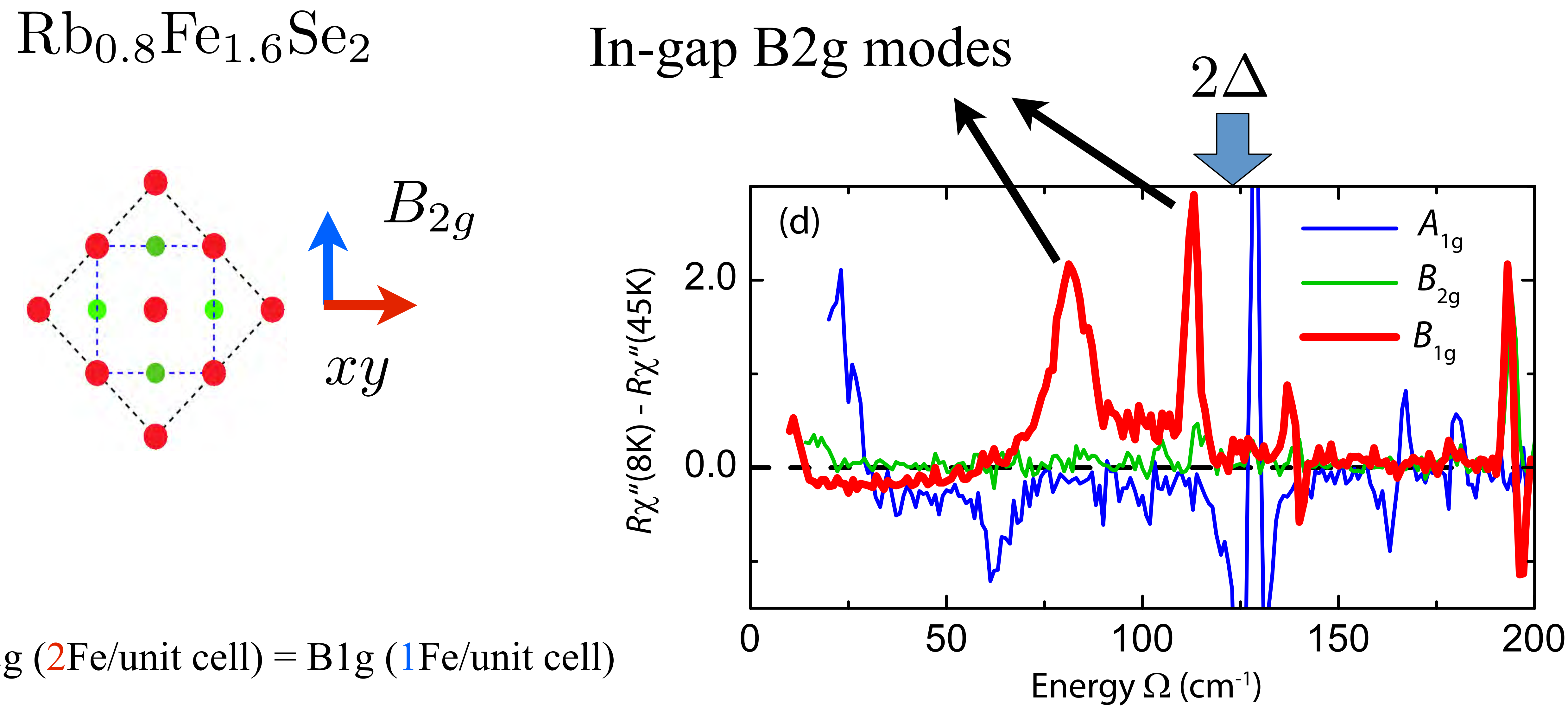
ARPES:

$\Delta \approx 8\text{meV}$

M.Xu,Q.Q.Ge,R.Peng,Z.R.Ye,J.Jiang, F.Chen,X.P. Shen, B. P. Xie, Y. Zhang, A. F. Wang, X. F. Wang, X. H. Chen, and D. L. Feng,
Phys. Rev. B **85**, 220504 (2012)

Raman-Scattering Detection of Nearly Degenerate *s*-Wave and *d*-Wave Pairing Channels in Iron-Based $Ba_{0.6}K_{0.4}Fe_2As_2$ and $Rb_{0.8}Fe_{1.6}Se_2$ Superconductors

F. Kretzschmar,[1] B. Muschler,[1] T. Böhm,[1] A. Baum,[1] R. Hackl,[1] Hai-Hu Wen,[2] V. Tsurkan,[3,4] J. Deisenhofer,[3] and A. Loidl[3]



B2g (2Fe/unit cell) = B1g (1Fe/unit cell)

Raman: In-gap B2g (d-wave) excitation

↓

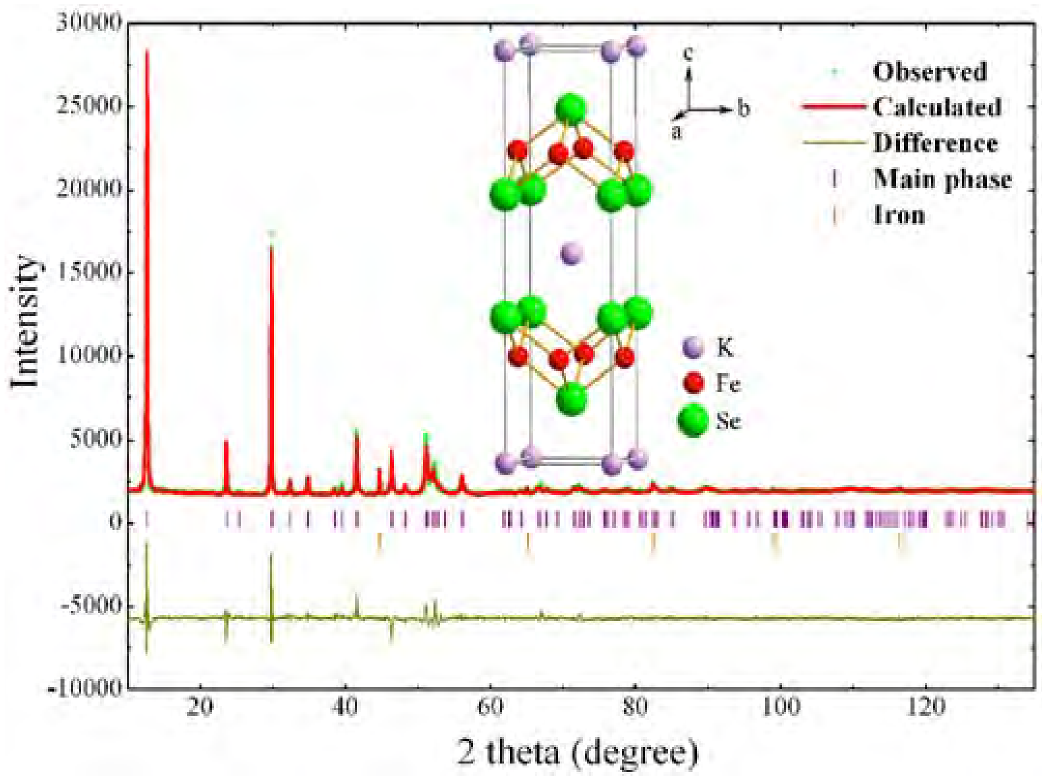
What about ground state ?

↓

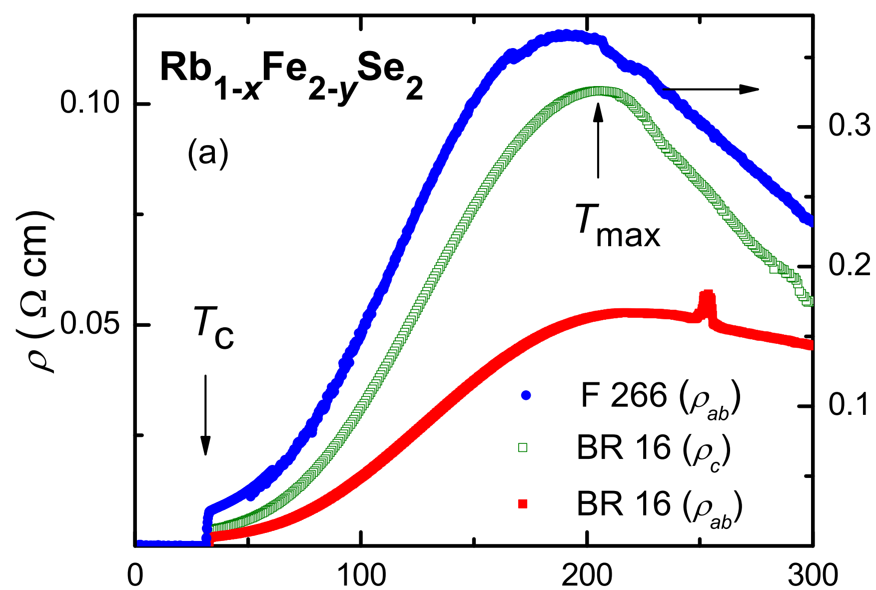
Take a closer look at selenides

$A_xFe_{2-y}Se_2$ (A=K,Rb,Cs)

$T_c \simeq 30 - 40\mathrm{K}$



$\mathbf{Rb_{1-x}Fe_{2-y}Se_2}$



V. Tsurkan, et.al. (2011)

$T_c \approx 32\mathrm{K}$

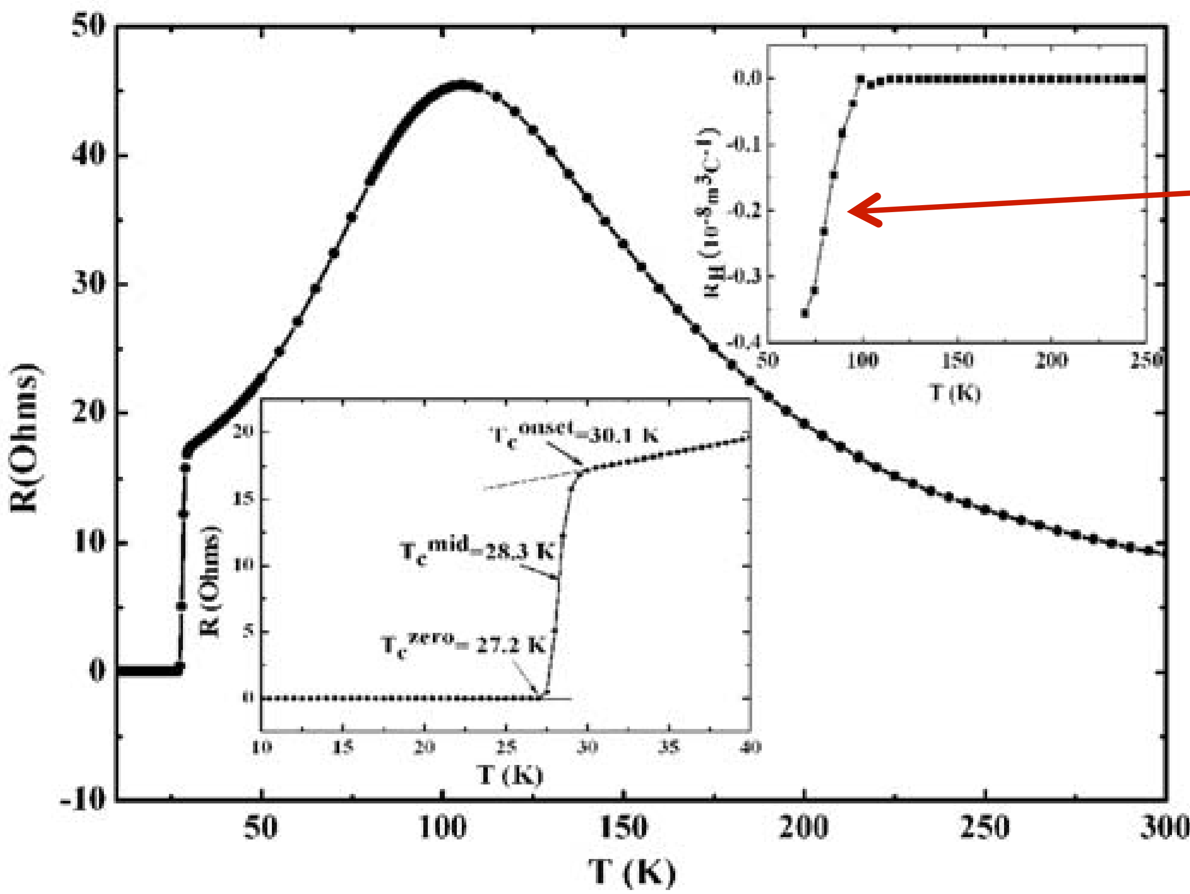
J. Guo et.al. $K_xFe_2Se_2$ $(0 \le x \le 1)$

Isostructural to $Ba_xFe_2As_2$ (122)

Intercalation of alkali metal A in between FeSe layers

“Strongly electron doped” systems, $A_xFe_{2-y}Se_2$ A= K,Rb,Cs

Alkali metal A donates electrons to FeSe layers



Negative Hall coefficient

$$R_H < 0$$

electrons

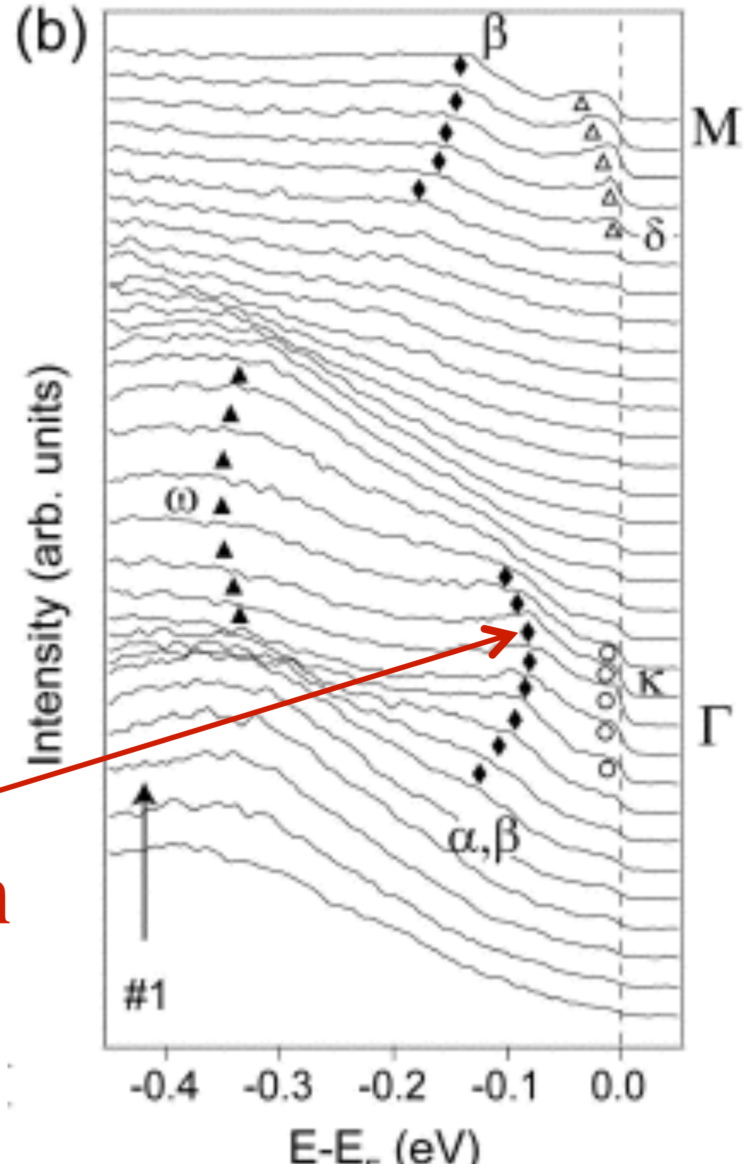
J. Guo et.al. $K_xFe_2Se_2$ $(0 \leq x \leq 1)$

"Strongly electron doped" systems, $A_xFe_{2-y}Se_2$ A= K,Rb,Cs

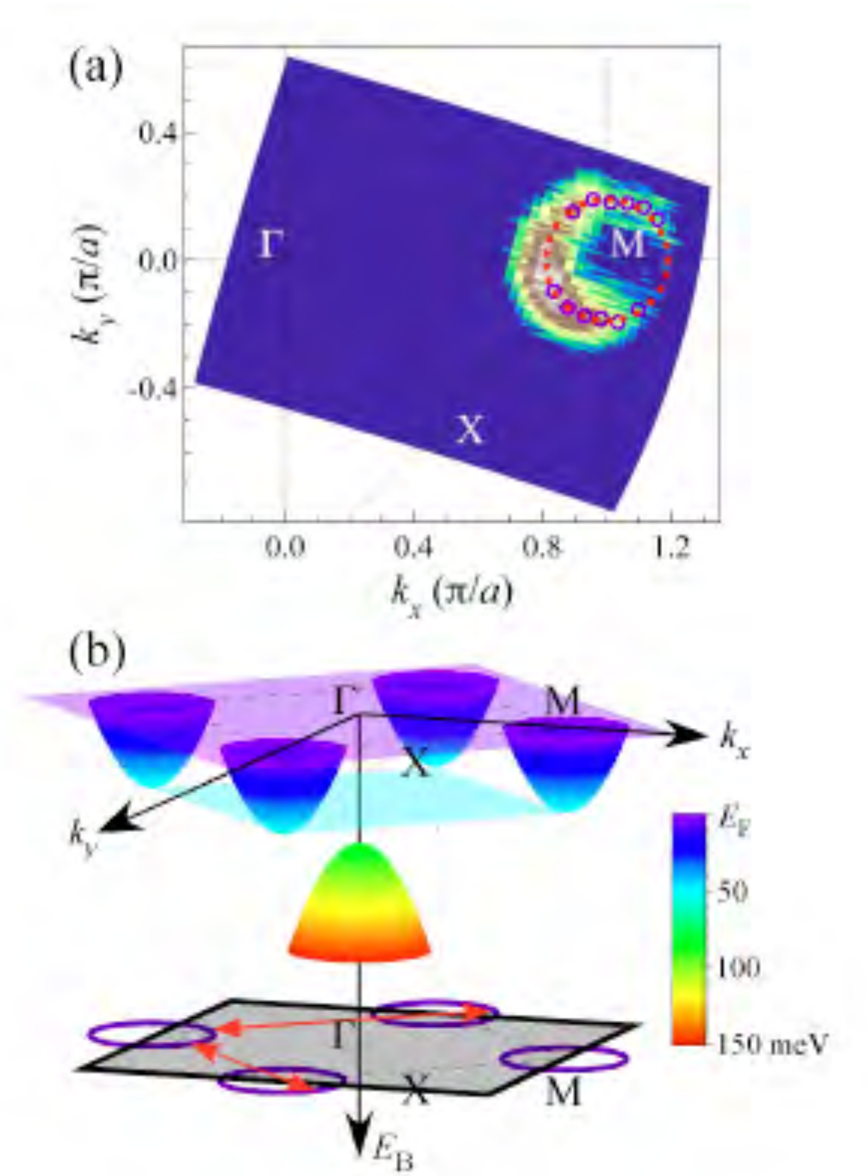
Tc ~40K

Only electron FSs are present

KFe_2Se_2



hole dispersion



Y. Zhang et al

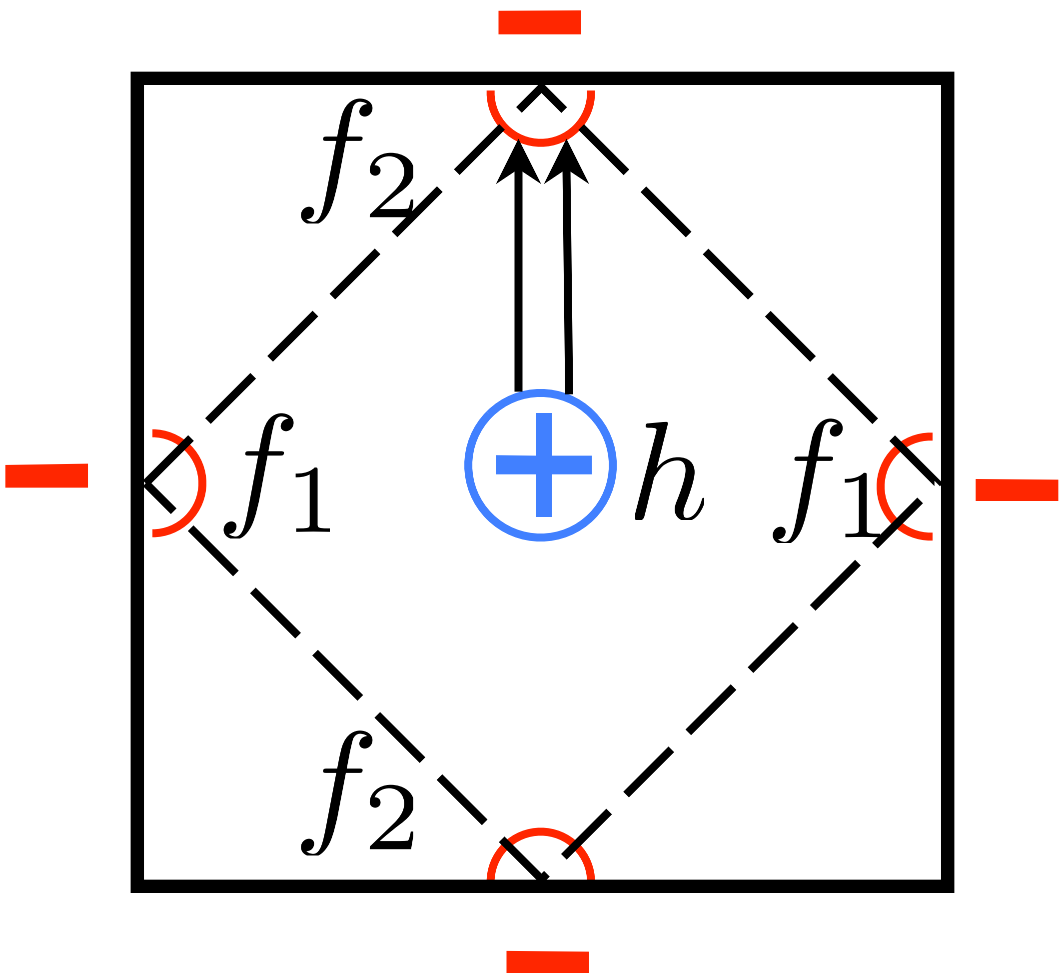
Hong Ding et al

What is the pairing *symmetry* ?

Pairing: order parameter *symmetry* ?

Look at pnictides with both electron and *hole* pockets

$$\begin{cases} \Delta_h + u\Delta_e \log\dfrac{\Lambda}{T_c} = 0 \\ \Delta_e + u\Delta_h \log\dfrac{\Lambda}{T_c} = 0 \end{cases}$$



$u > 0$ ⇒ $s^{\pm}$

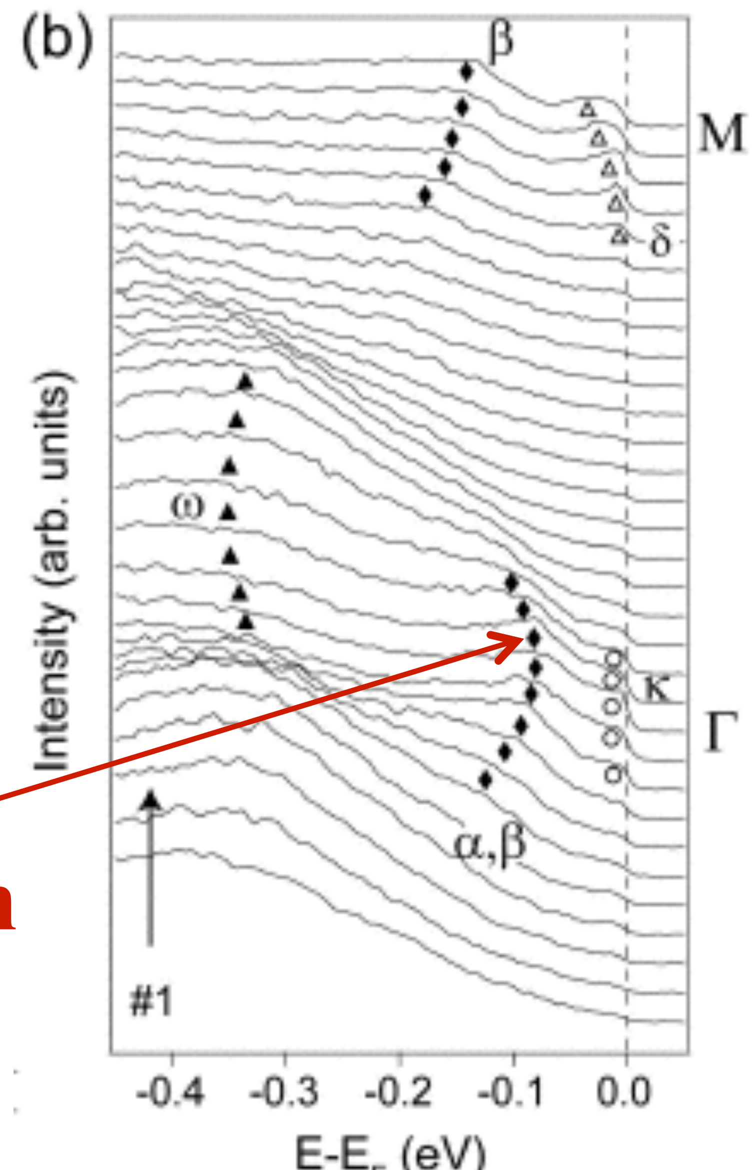
S ⇒ Invariant under *crystal* symmetry

"Strongly electron doped" systems, $A_xFe_{2-y}Se_2$ A= K,Rb,Cs

Tc ~40K

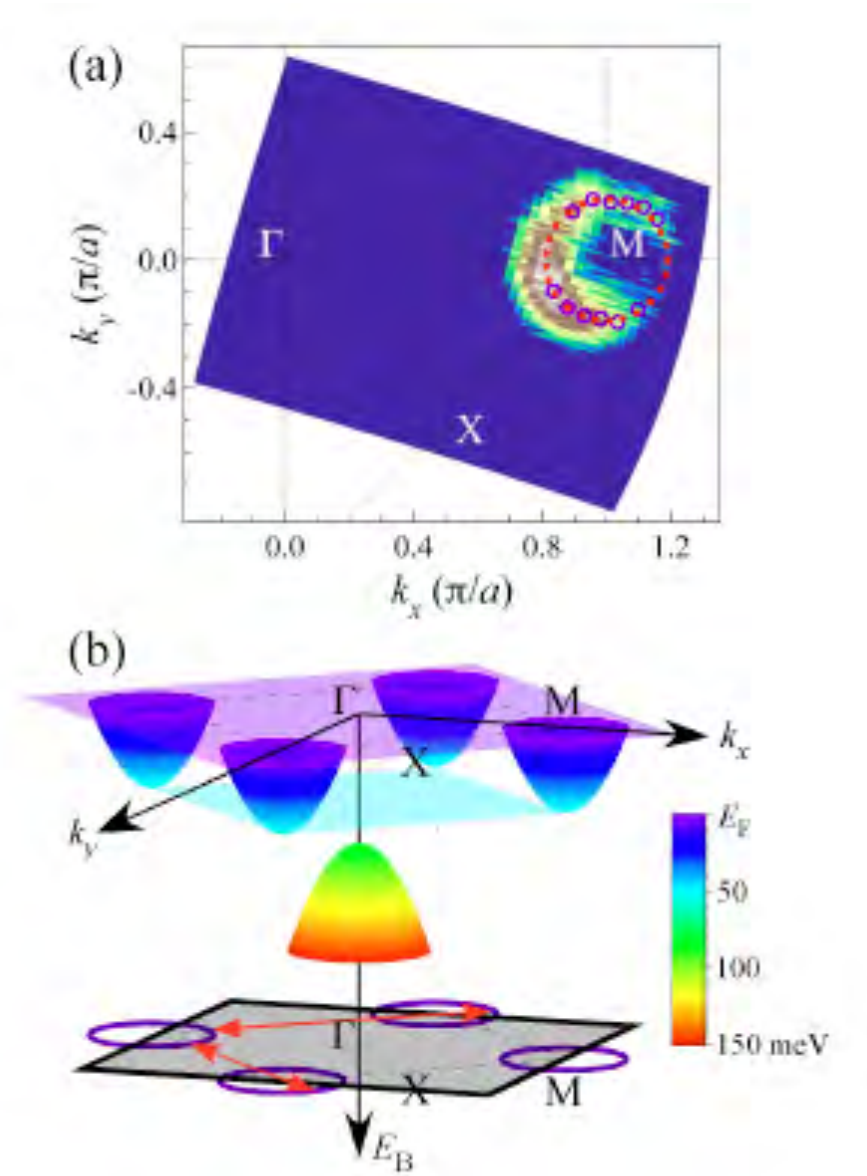
Only electron FSs are present

KFe_2Se_2



hole dispersion

Y. Zhang et al



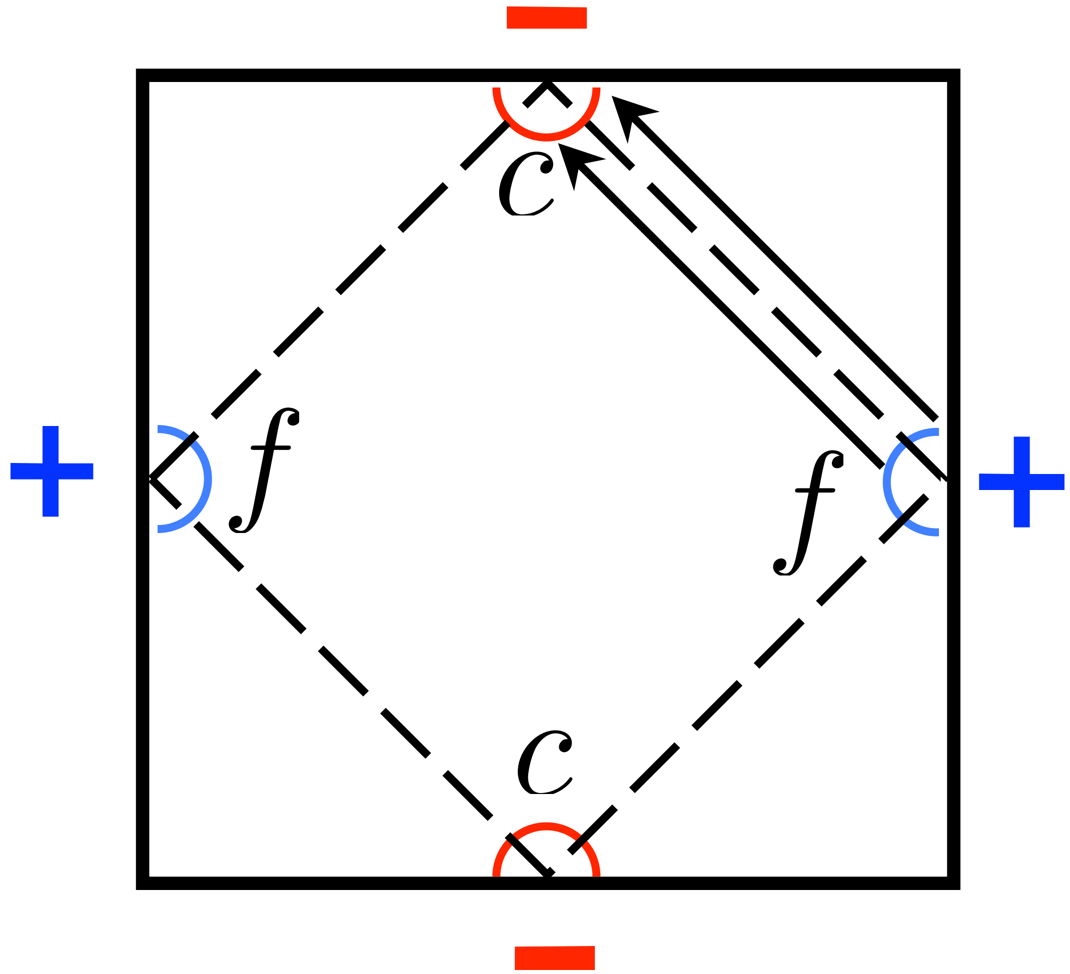
Hong Ding et al

Hole pockets are gapped –the driving force for s-wave SC is gone

Can this still work for AFe_2Se_2 ?

Nodeless d?

Different from pnictides with <u>*hole*</u> pockets!

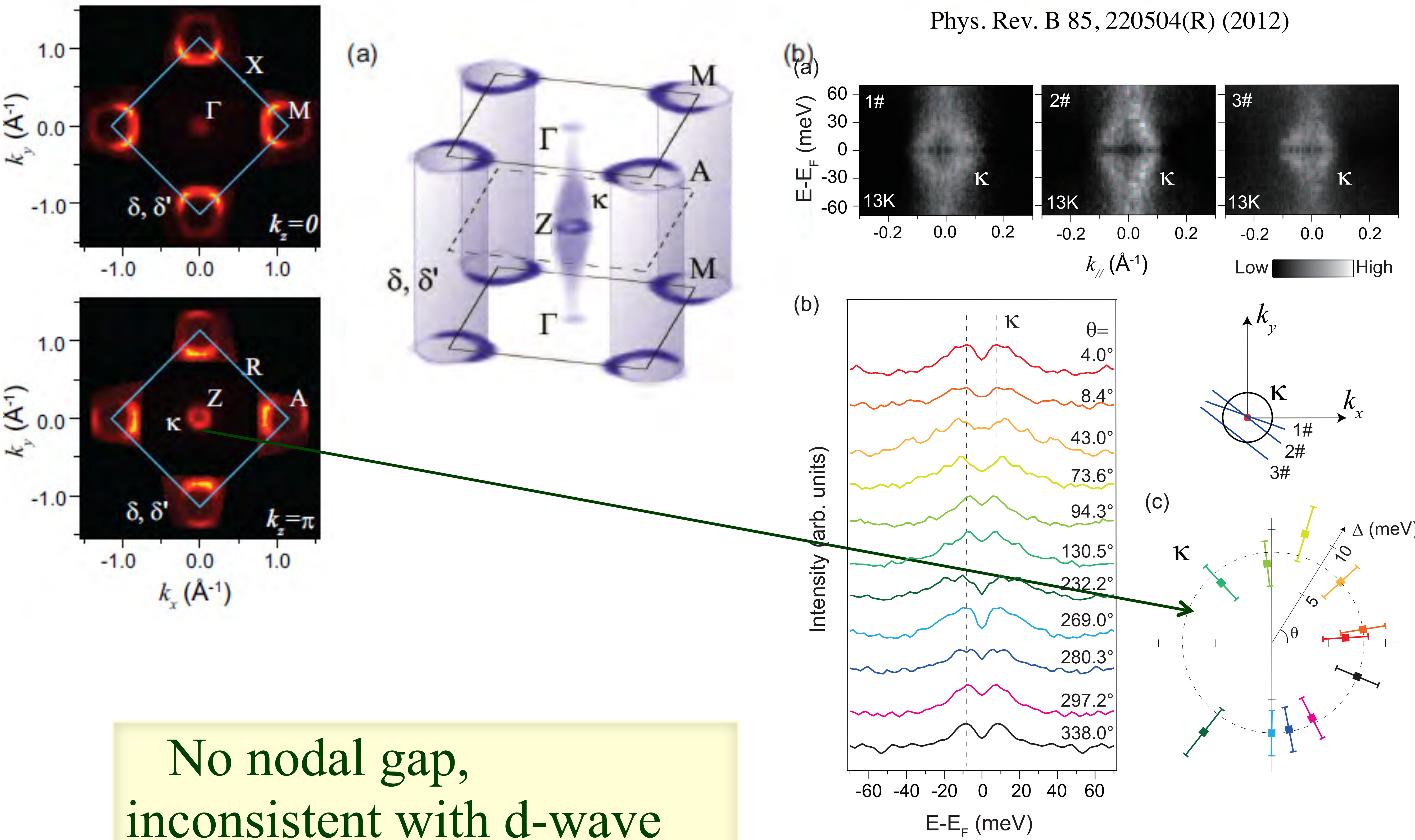


Experiment: ?

Recent ARPES

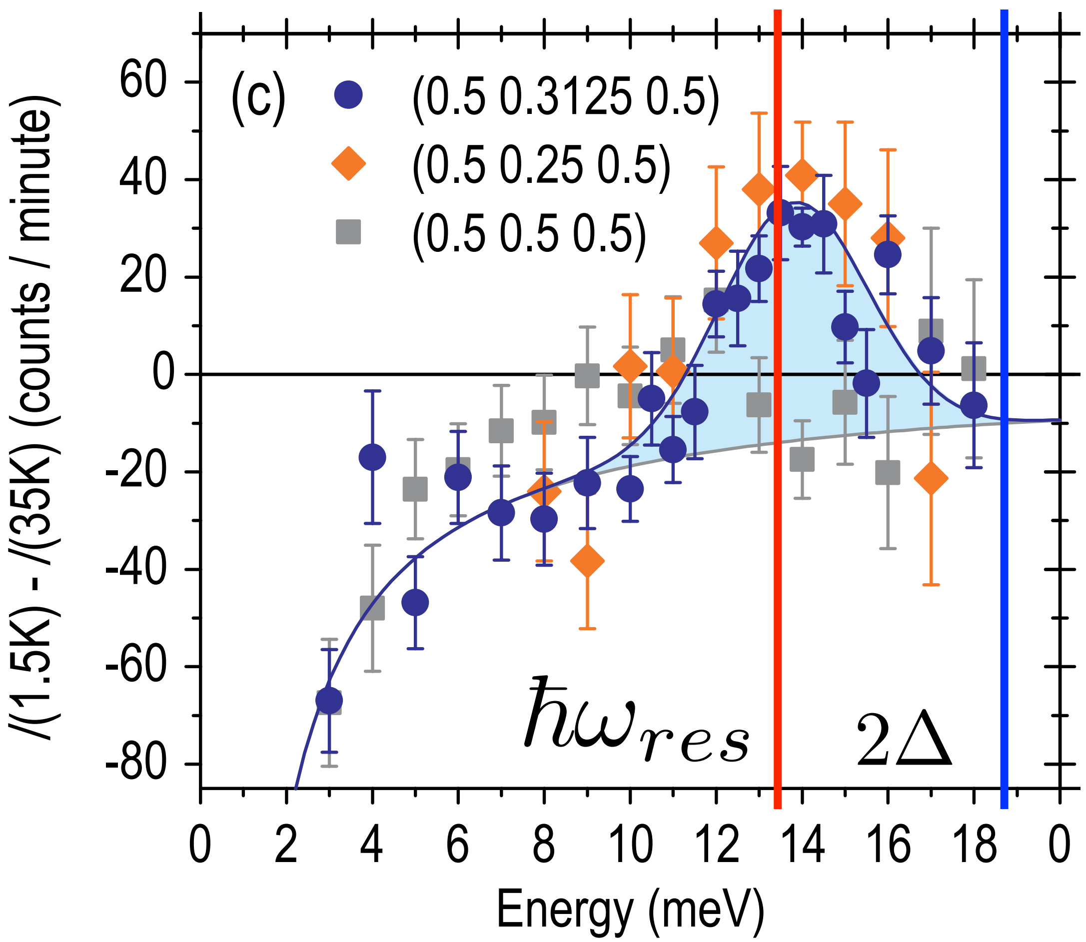
Feng's group

Phys. Rev. B 85, 220504(R) (2012)



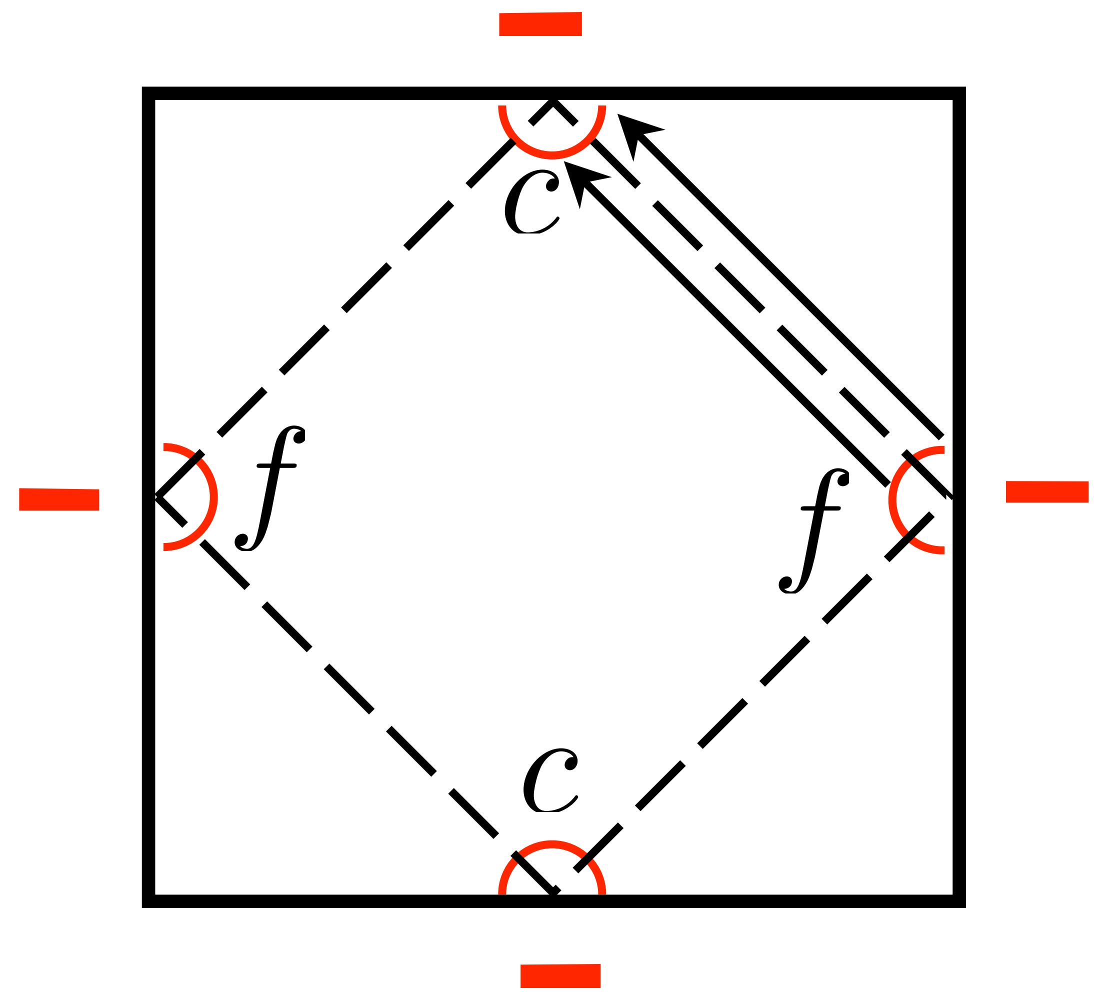
No nodal gap,
inconsistent with d-wave

What about regular s-wave?



Spin resonance below Tc

Inosov et al



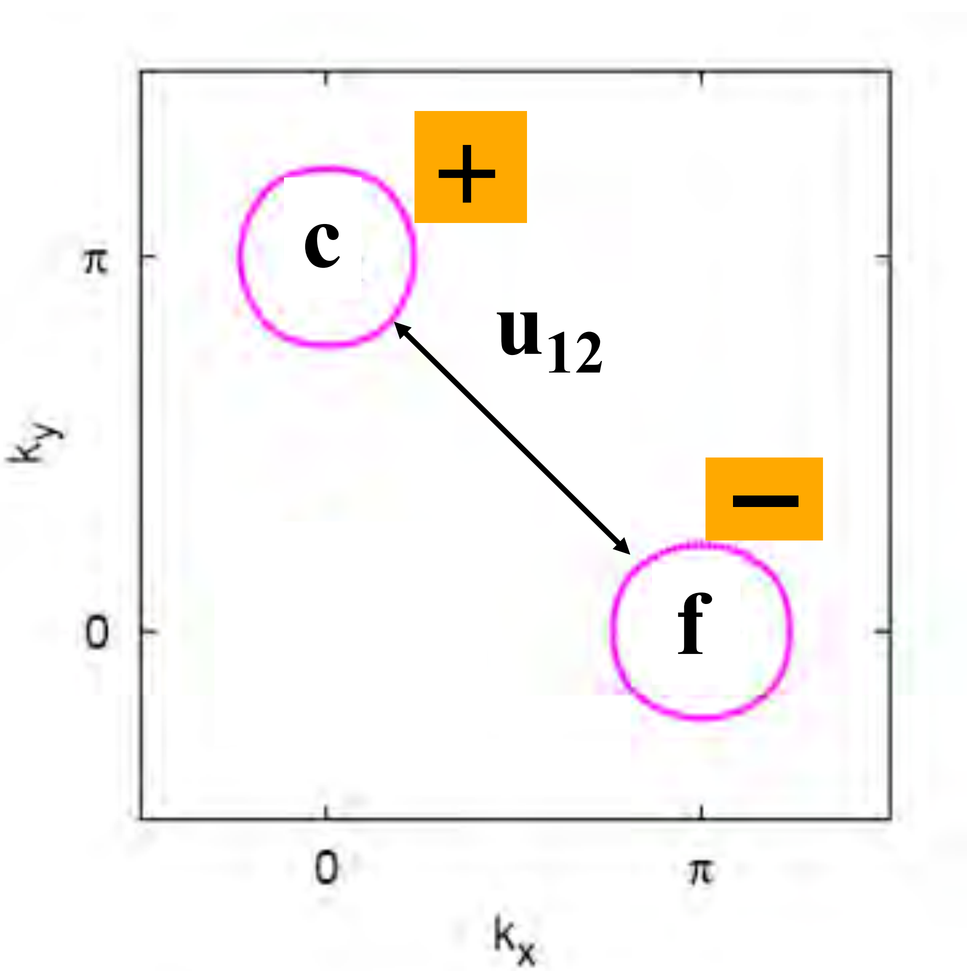
$$\frac{\hbar\omega_{res}}{2\Delta} \approx 0.7$$

Requires sign changing order parameter

There is a third possibility
“another s+-”

Consistent with both ARPES and neutrons

Let's go back to d-wave reasoning

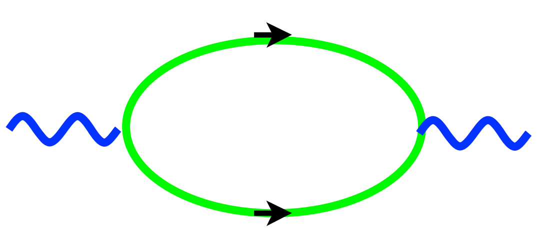


Suppose FSs are circles (must be identical circles)

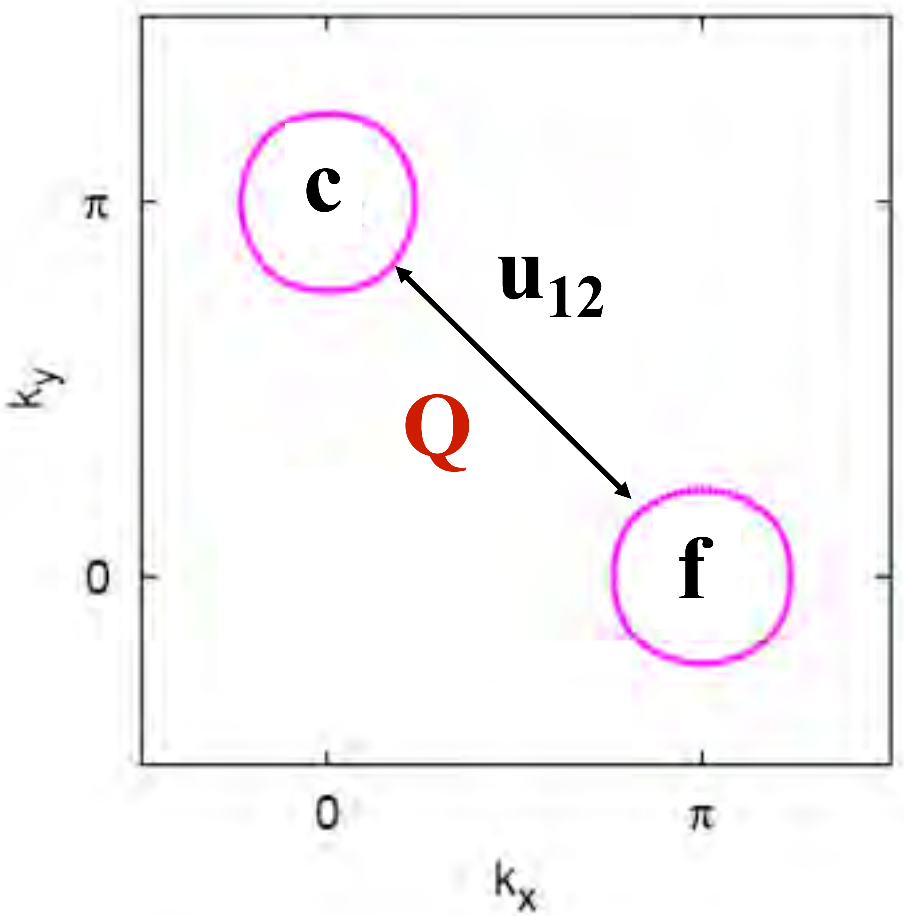
d-wave: $\Delta_c = -\Delta_f$

$$\Delta_c = \langle c^\dagger_\uparrow c^\dagger_\downarrow \rangle, \quad \Delta_f = \langle f^\dagger_\uparrow f^\dagger_\downarrow \rangle$$

$$T_c: \quad 1 = u_{12}\, \chi_{pp}(0)$$

$\chi_{pp}(0) =$  $= \iint_T \frac{d\omega\, d\varepsilon_k}{\omega^2 + \varepsilon_k^2} = \log \frac{E_F}{T}$ **Cooper logarithm**

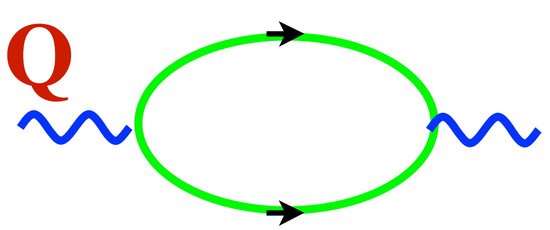
And what if we consider inter-pocket pairing ?



s-wave Singlet

$$\langle c^\dagger_\uparrow f^\dagger_\downarrow \rangle = \Delta = \langle f^\dagger_\uparrow c^\dagger_\downarrow \rangle$$

$$T_c: \quad 1 = u_{12}\, \chi_{pp}\,(Q)$$

$\chi_{pp}\,(Q) =$ 

$$= \iint_T \frac{d\omega\, d\varepsilon_k}{\omega^2 + \varepsilon_k^2} = \log \frac{E_F}{T}$$

Cooper logarithm

Exactly the same result as for plus-minus gap

d-wave and s-wave pairing states are completely degenerate for circular electron pockets

Electron Pockets

1) *not* circular 2) *hybridized*

↓

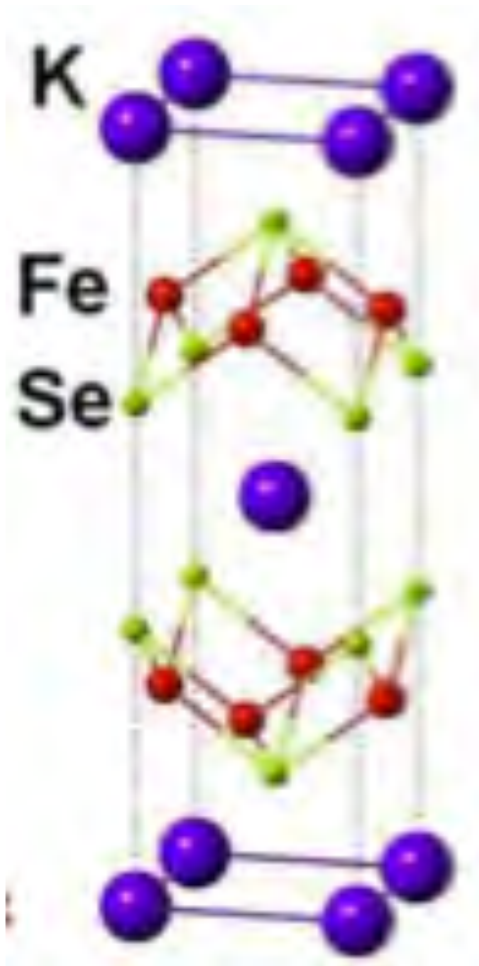
d-s degeneracy is lifted

↓

d-s competition → which way ?

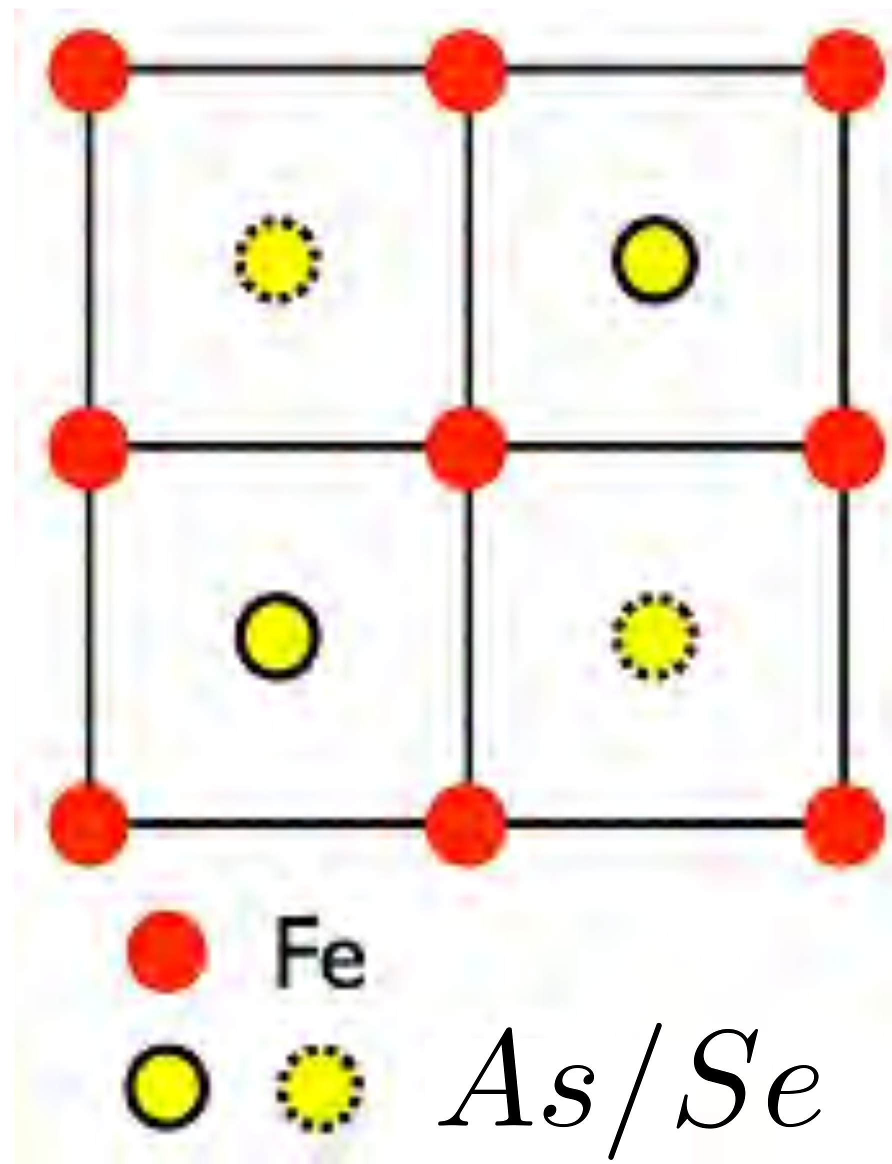
Hybridization

Folding: pnictogen/chalcogen is above or below Fe plane



Single FeSe layer

$\longrightarrow$

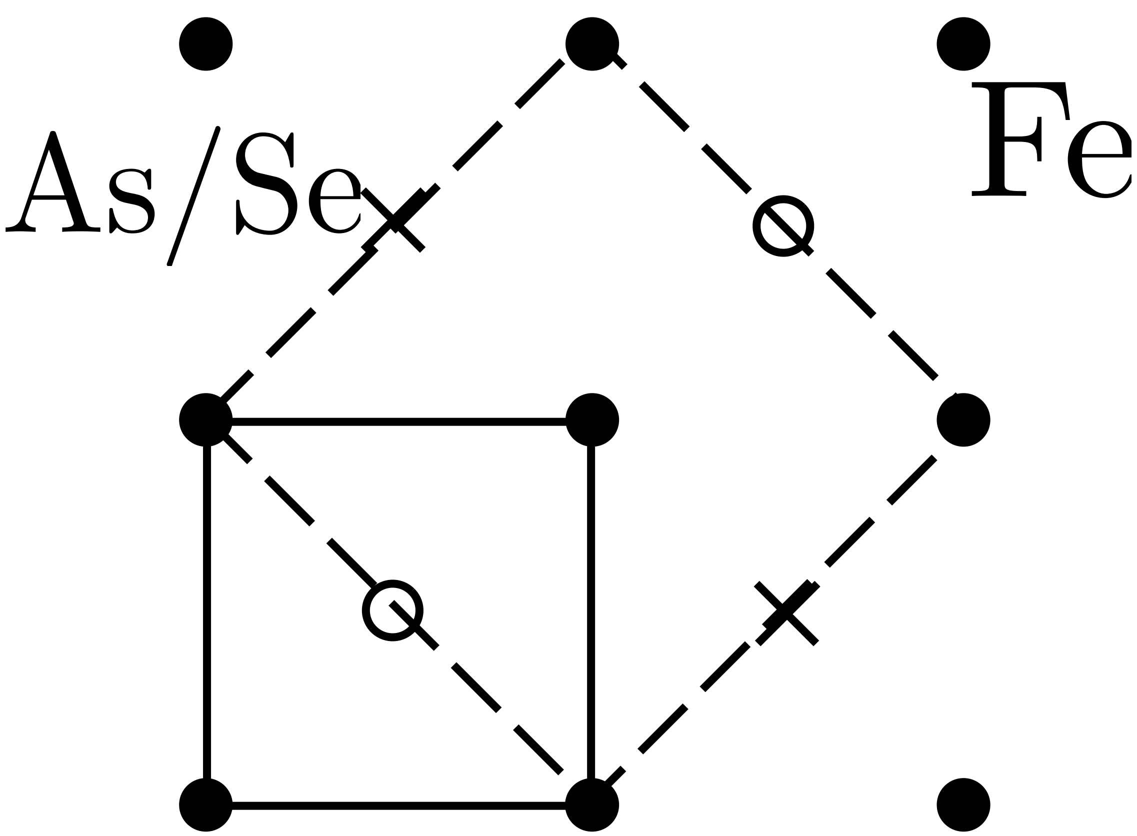


Two non-equivalent positions of Fe – unit cell has 2 Fe atoms

Hybridized pockets

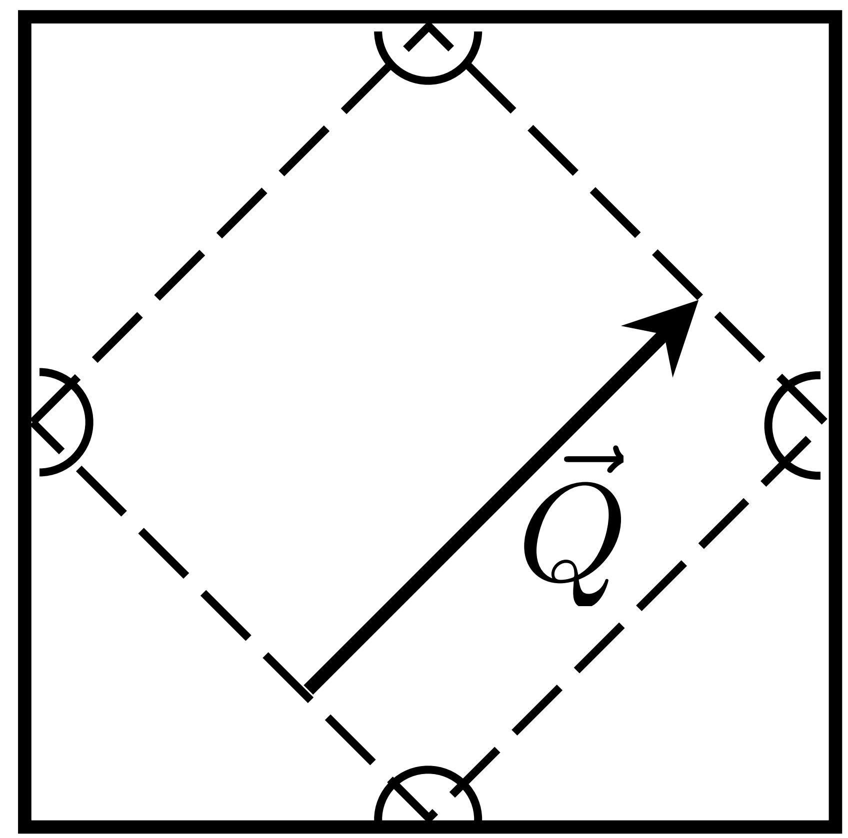
Single layer

Real space



Reciprocal lattice

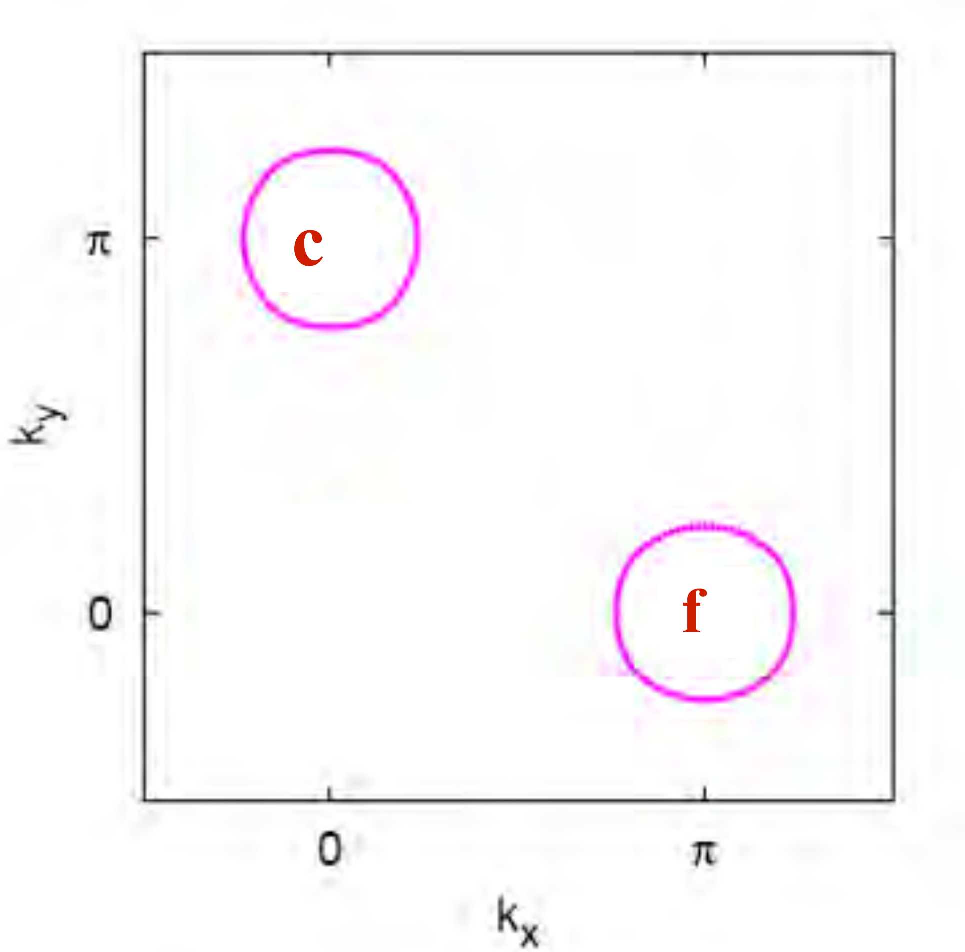
1FeBZ

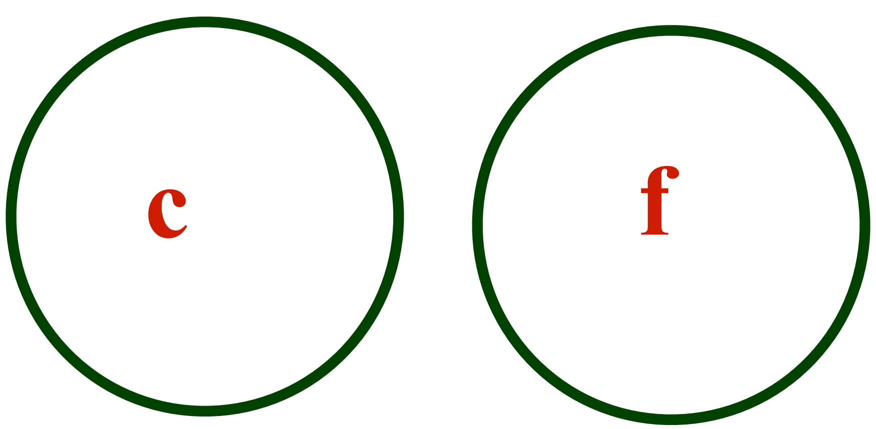


$$\vec{Q} = (\pi, \pi)$$

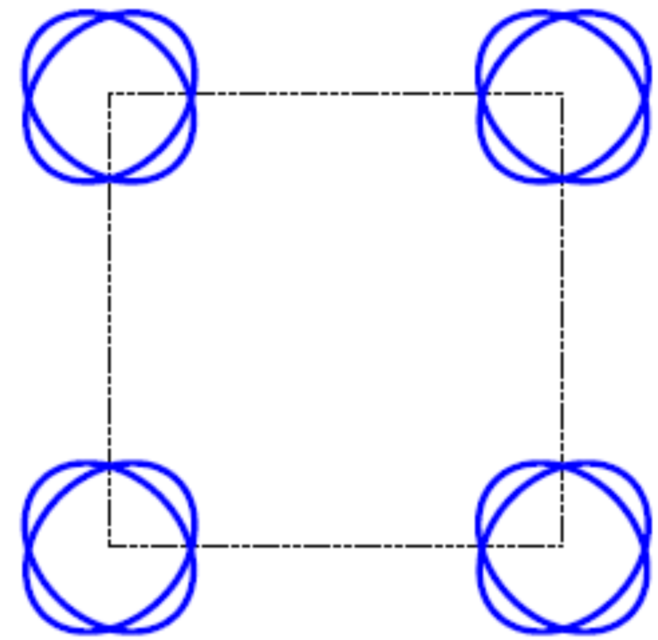
A simple picture of folding – a shift of the position of the FSs

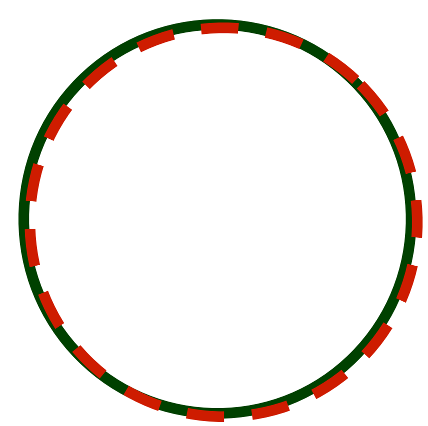
unfolded zone





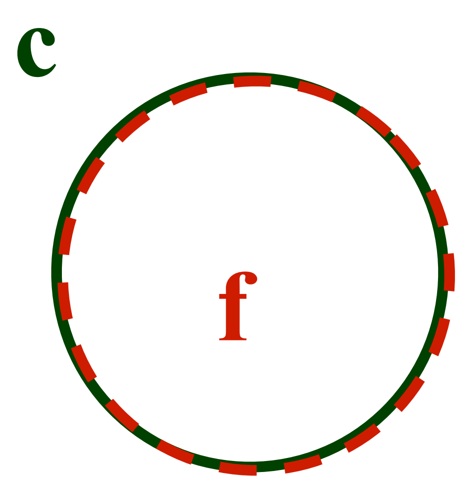
folded zone



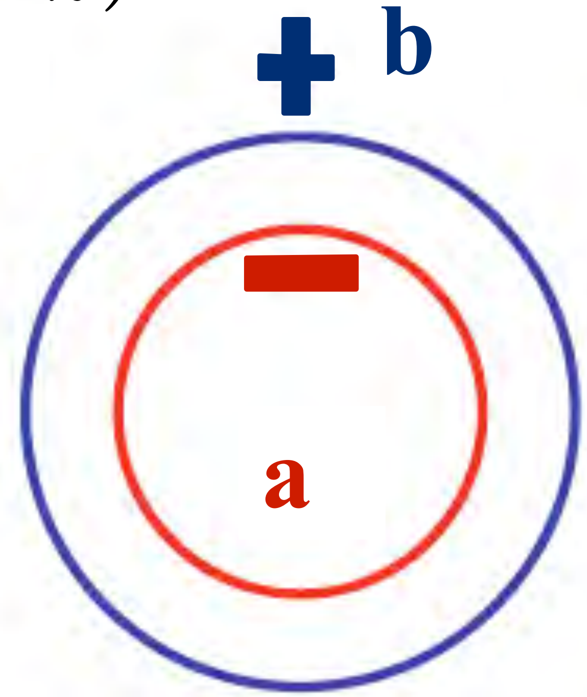


Inter-pocket and intra-pocket pairing are degenerate

$$H_{hybr} = \lambda(c_k^\dagger f_{k+Q} + f_{k+Q}^\dagger c_k)$$



no **hybridization**



s^{+-} gap

$$a = (c+f)/\sqrt{2}$$
$$b = (c-f)/\sqrt{2}$$

hybridization

$$\langle c_\uparrow^\dagger c_\downarrow^\dagger \rangle = \Delta = -\langle f_\uparrow^\dagger f_\downarrow^\dagger \rangle$$

d - wave

$$< a_\uparrow^+ a_\downarrow^+ > = \Delta = - < b_\uparrow^+ b_\downarrow^+ >$$

s - wave

$$\langle c_\uparrow^\dagger f_\downarrow^\dagger \rangle = \Delta = \langle f_\uparrow^\dagger c_\downarrow^\dagger \rangle$$

s - wave

$$< a_\uparrow^+ b_\downarrow^+ > = \Delta = < b_\uparrow^+ a_\downarrow^+ >$$

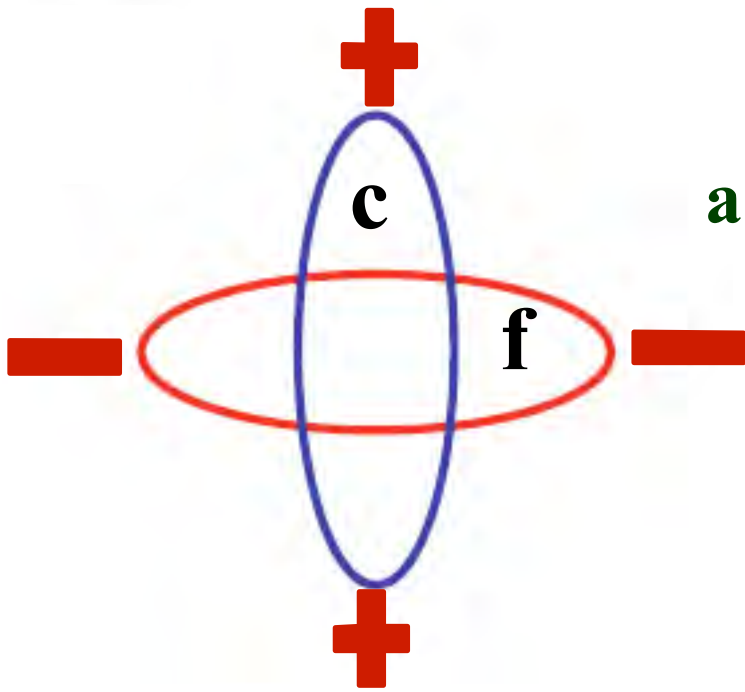
d - wave

s-wave is inter-pocket pairing in terms of original fermions

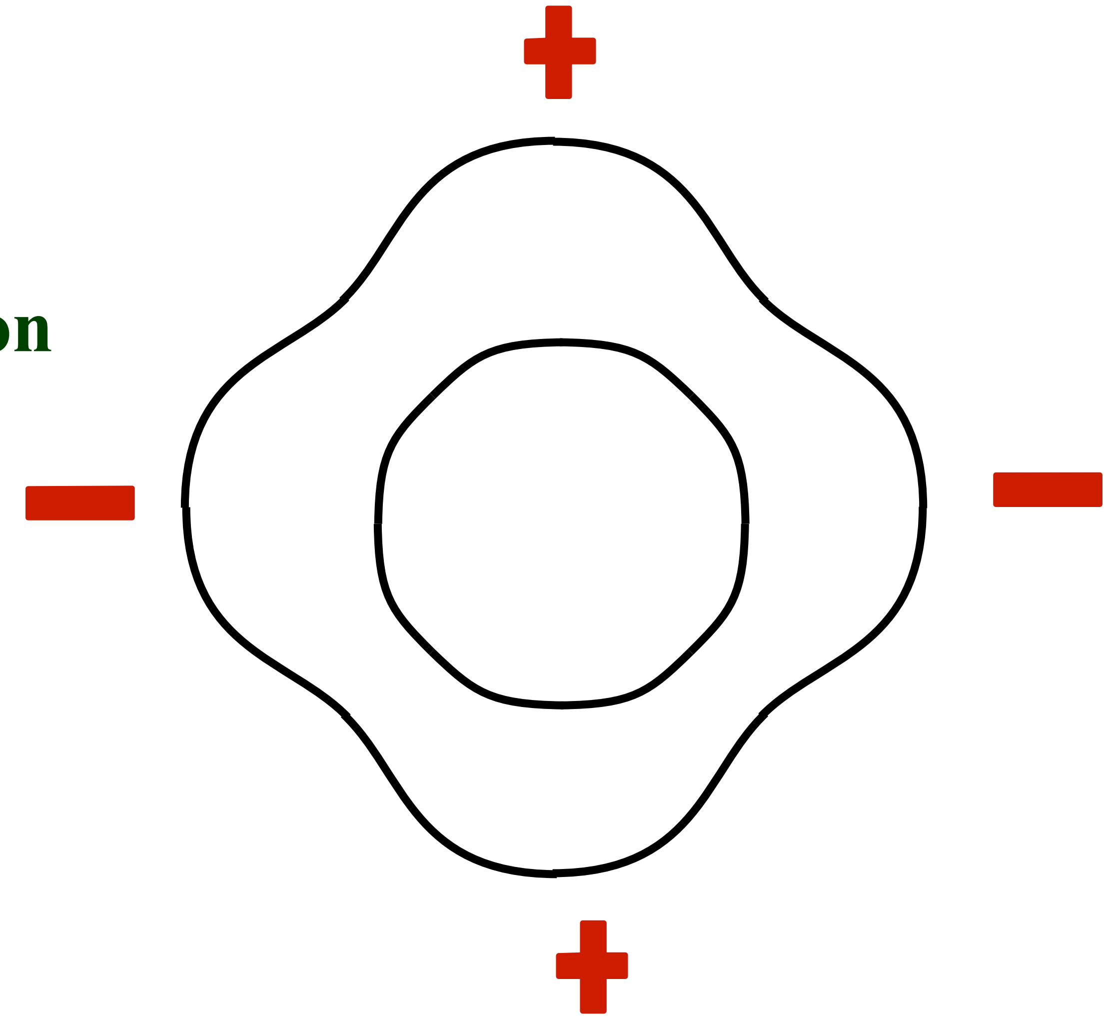
d-wave is inter-pocket pairing in terms of hybridized fermions

A situation is somewhat different when electron pockets are ellipses

no hybridization



add hybridization



d-wave is better

$$\langle c^\dagger_\uparrow c^\dagger_\downarrow \rangle = \Delta = -\langle f^\dagger_\uparrow f^\dagger_\downarrow \rangle$$

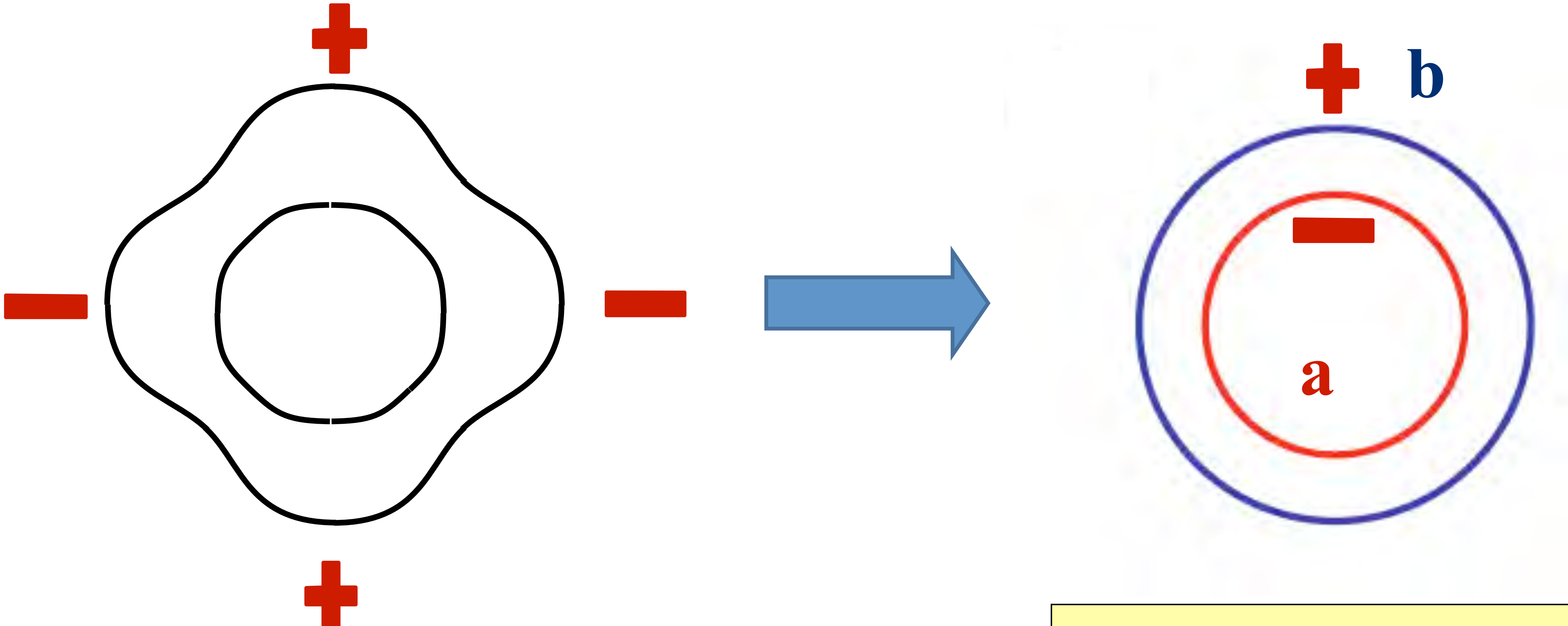
d - wave

$$\langle c^\dagger_\uparrow f^\dagger_\downarrow \rangle = \Delta = \langle f^\dagger_\uparrow c^\dagger_\downarrow \rangle$$

s - wave

for small hybridization, d-wave is still better. This is a conventional , d-wave

Let's increase hybridization further



Eventually s-wave wins

$$< a^{+}_{\uparrow} a^{+}_{\downarrow} > \; = \Delta = - < b^{+}_{\uparrow} b^{+}_{\downarrow} >$$

s - wave

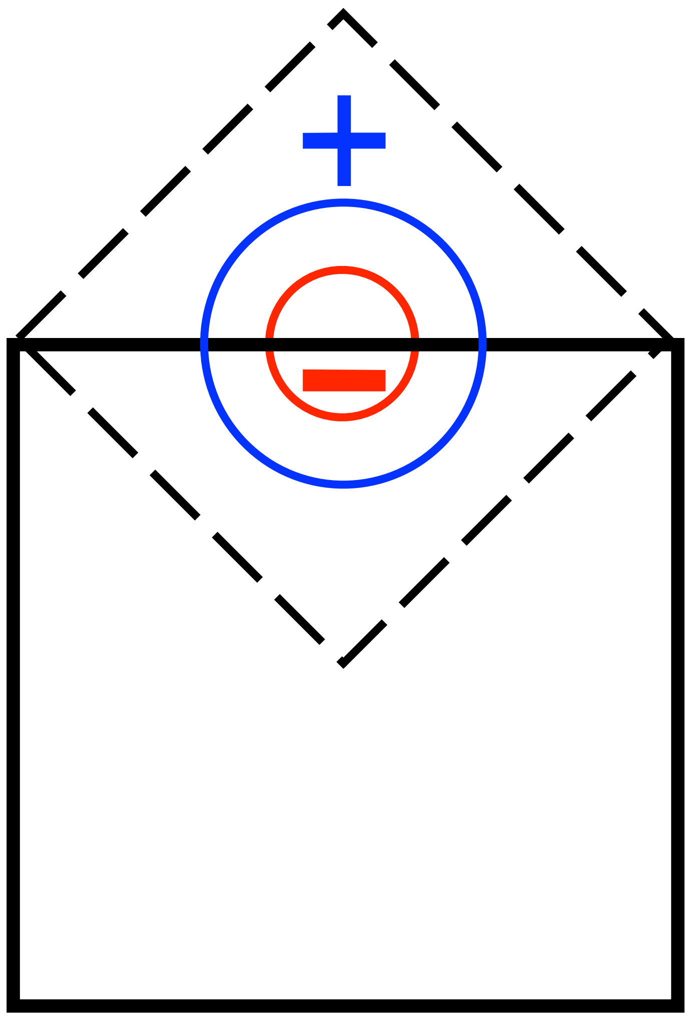
$$< a^{+}_{\uparrow} b^{+}_{\downarrow} > \; = \Delta = < b^{+}_{\uparrow} a^{+}_{\downarrow} >$$

d - wave

The effect of hybridization for different pocket configuration

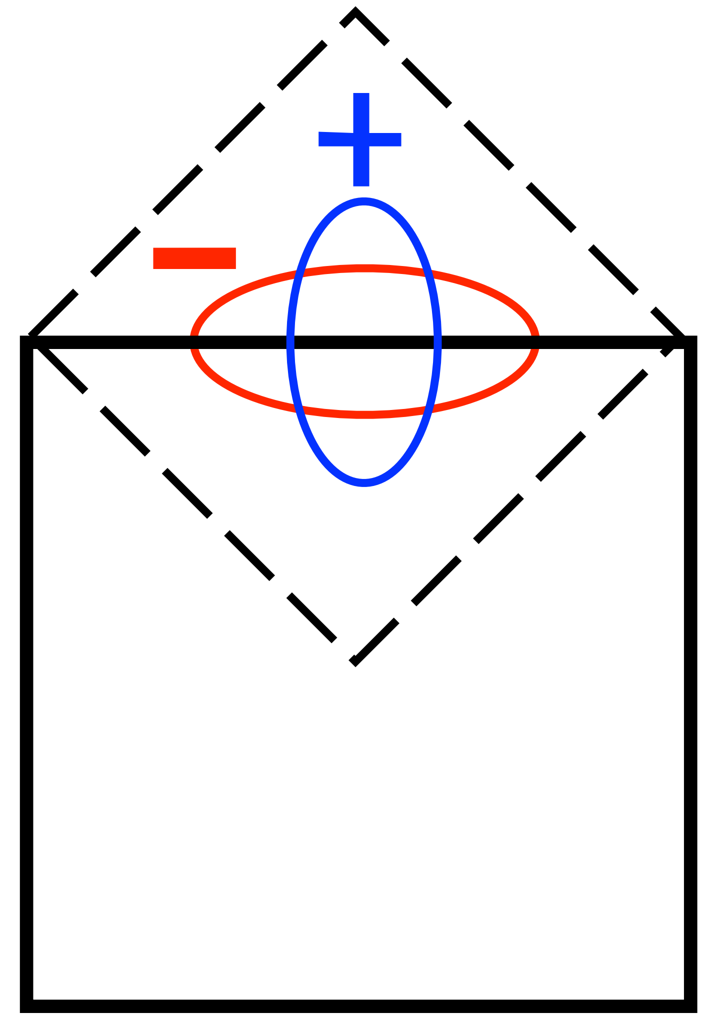
Circular pockets

$s^{\pm}$



Elliptical pockets

d



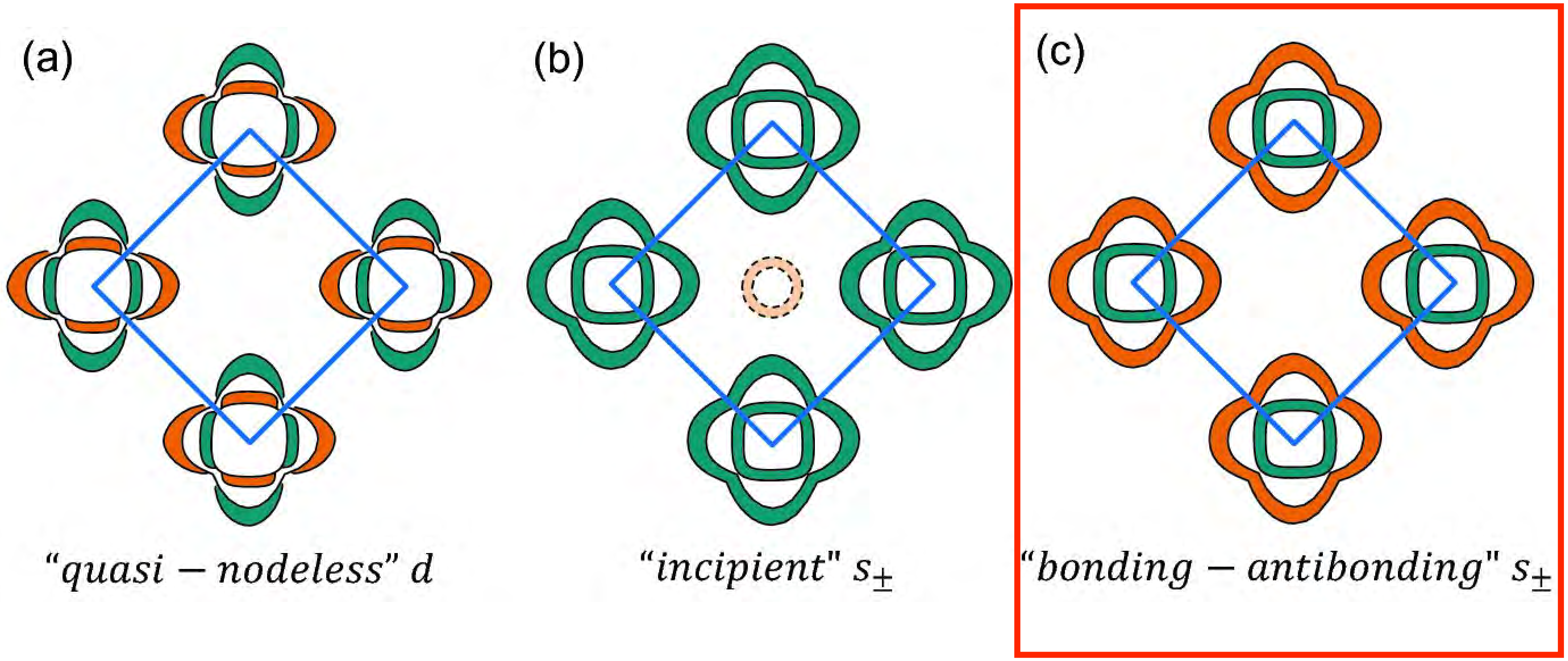
I. Mazin, PRB (2011)

Competition: hybridization vs. ellipticity

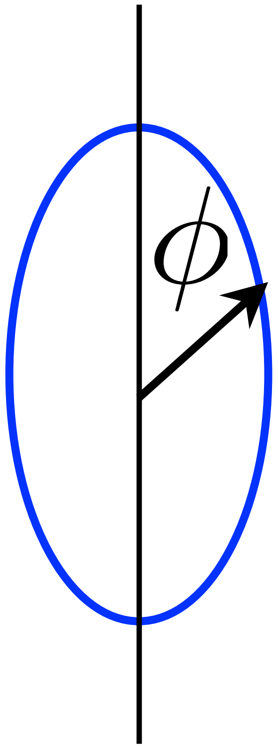
"Gap symmetry and structure of Fe-based superconductors"

P. J. Hirschfeld, M. M. Korshunov, I. I. Mazin

Rep. Prog. Phys. **74**, 124508 (2011)



Hybridization vs. ellipticity (cont)



$$v_F(\phi) = v_F(1 + a\cos 2\phi)$$

$$k_F(\phi) = k_F(1 + b\cos 2\phi)$$

Anisotropy parameter

$$\frac{\lambda}{v_F k_F}$$ → Dimensionless *hybridization*

} Small

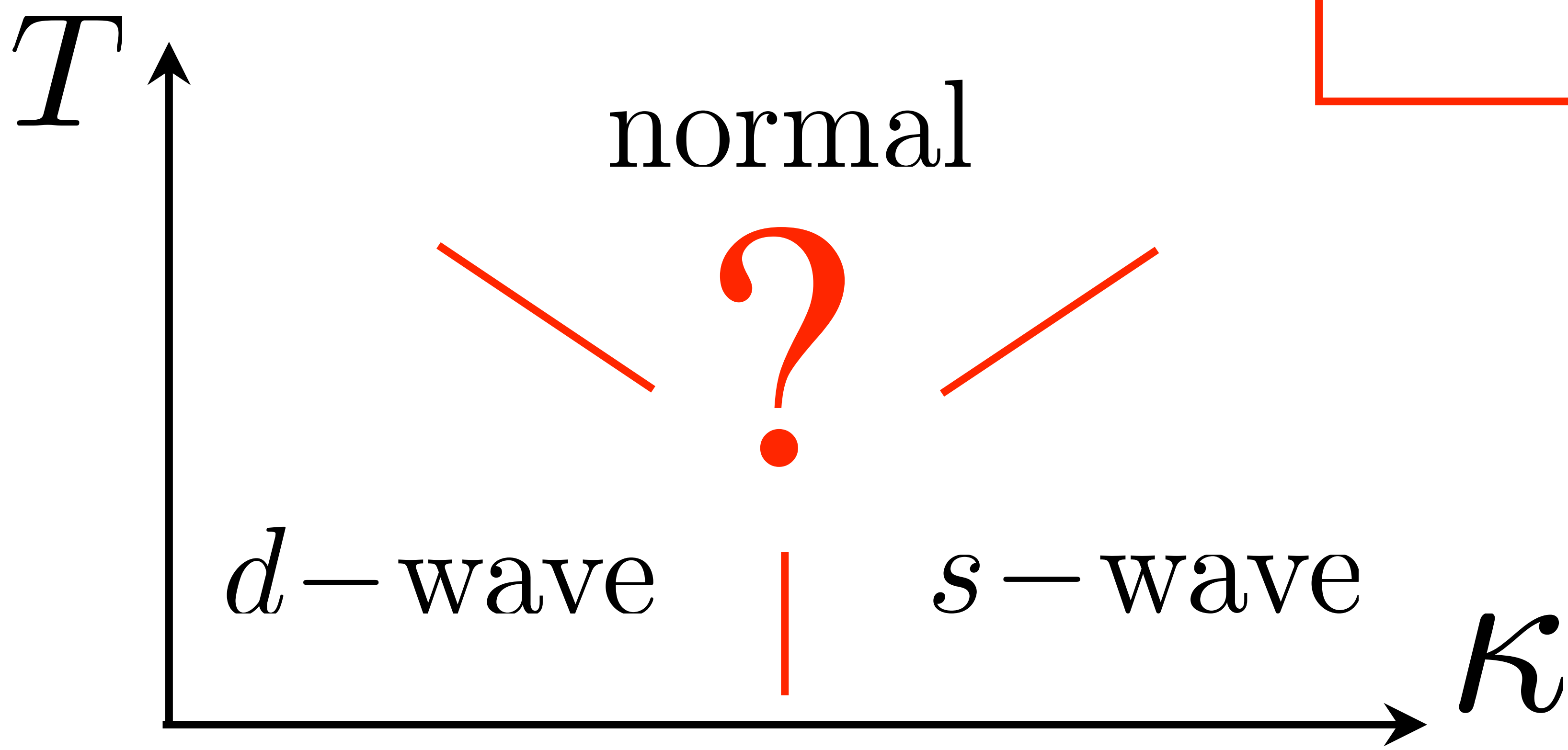
$$\kappa = \frac{\text{hybridization}}{\text{ellipticity}}$$

$$\kappa = \frac{\lambda}{v_F k_F |b|}$$ → Can be anything

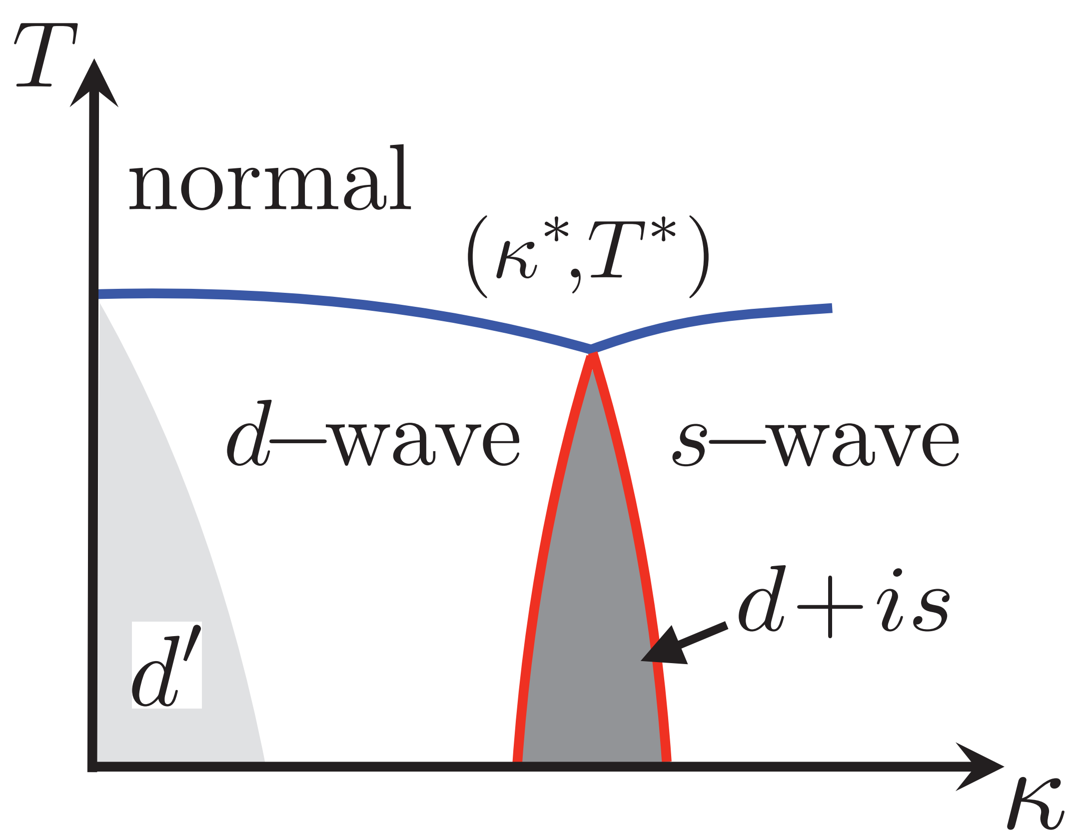
Results

$$\kappa = \frac{\lambda}{v_F k_F |b|}$$

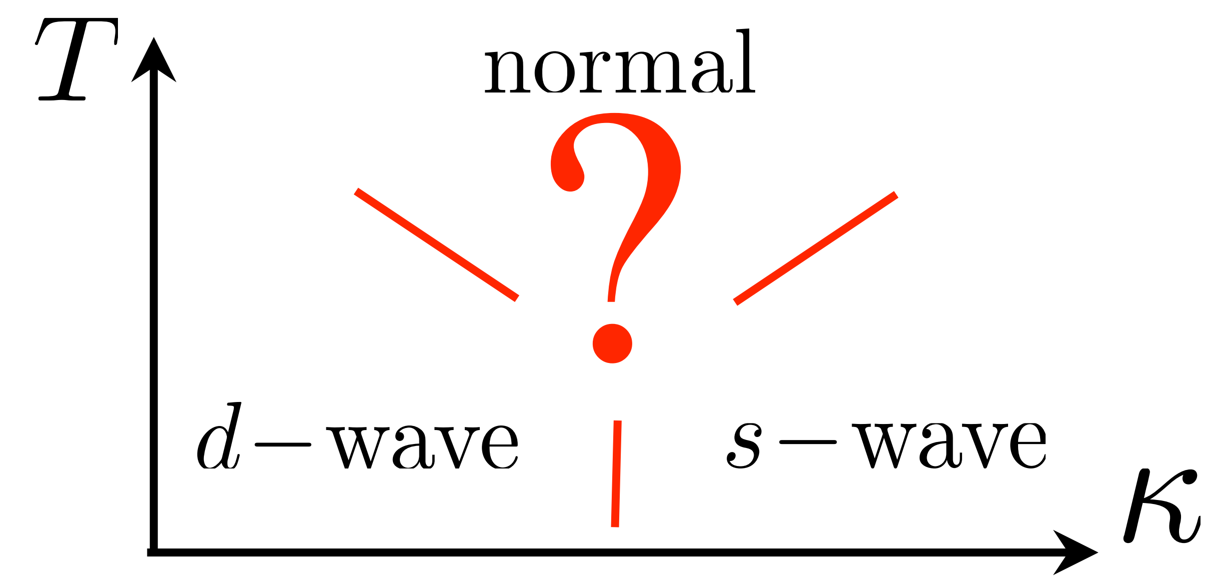
Question



$$T_c \ll \lambda (\approx 20\mathrm{meV})$$



Phase diagram



Ginzburg-Landau functional
<u>*two*</u> order parameters

Δ_s s-wave

Δ_d d-wave

~~$\Delta_d^* \Delta_s$~~ $\longleftarrow$ symmetry

$$F_{GL} = A_s|\Delta_s|^2 + A_d|\Delta_d|^2 + \frac{B_s}{2}|\Delta_s|^4 + \frac{B_d}{2}|\Delta_d|^4$$
$$+ C|\Delta_s|^2|\Delta_d|^2 + \frac{E}{2}[(\Delta_s\Delta_d^*)^2 + (\Delta_s^*\Delta_d)^2]$$

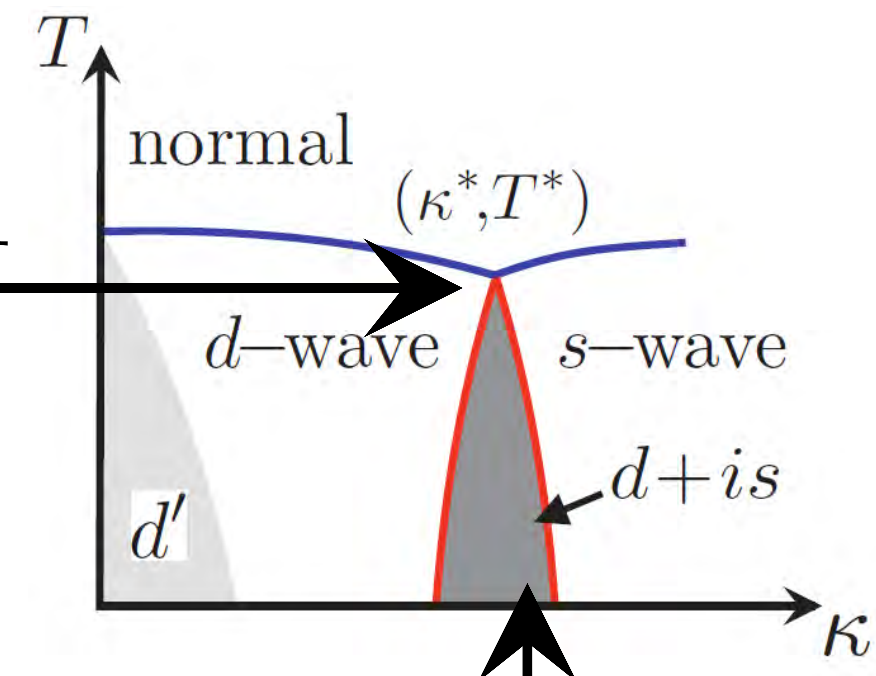
Ginzburg-Landau functional treatment (cont)

$$F_{GL} = A_s|\Delta_s|^2 + A_d|\Delta_d|^2 + \frac{B_s}{2}|\Delta_s|^4 + \frac{B_d}{2}|\Delta_d|^4 + C|\Delta_s|^2|\Delta_d|^2 + \frac{E}{2}[(\Delta_s\Delta_d^*)^2 + (\Delta_s^*\Delta_d)^2]$$

Tetracritical point:

$$A_s(\kappa^*, T^*) = A_d(\kappa^*, T^*)$$

$$\kappa^* \simeq 1$$



Time-reversal breaking

$$\lambda \gg T_c$$

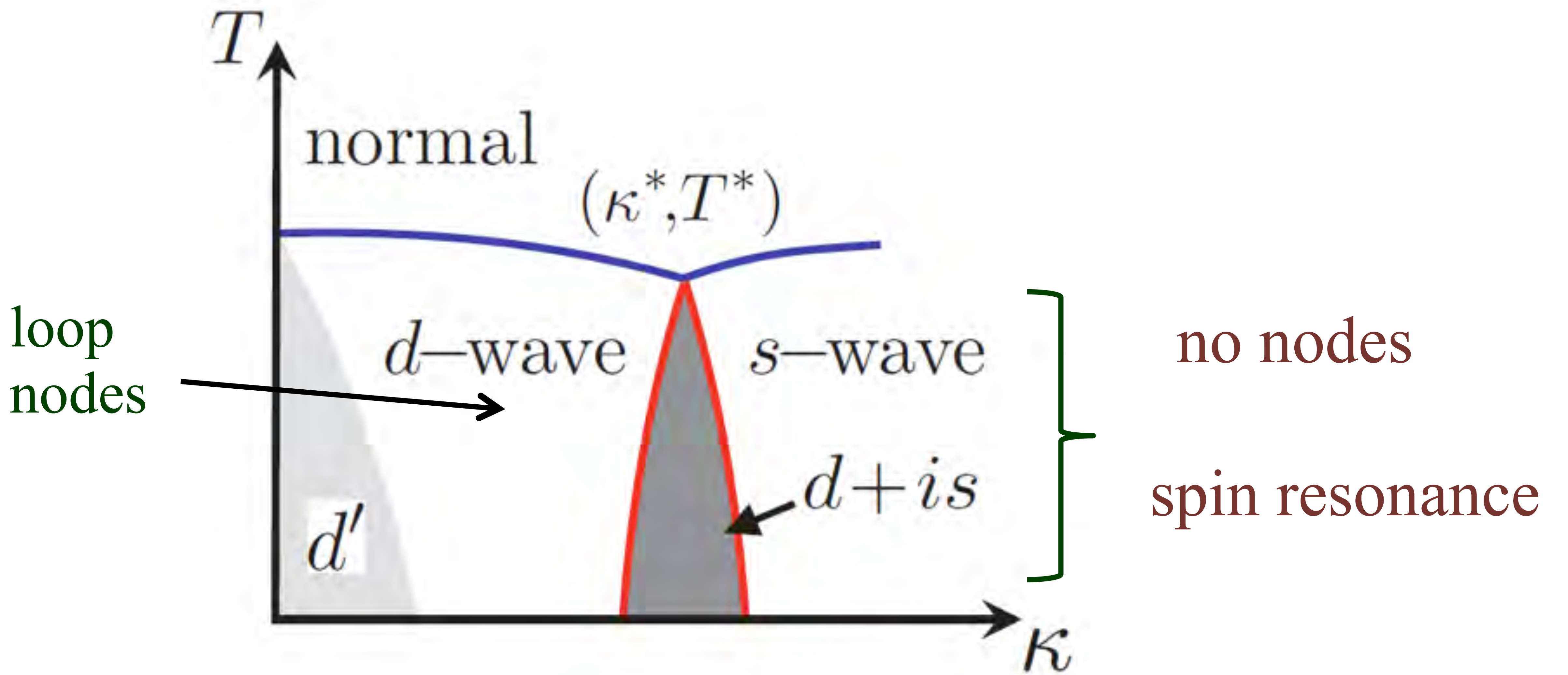
$$B_s = B_d = B = \frac{10}{3}E$$

$$C = 2E$$

B+E >C → $s + id$

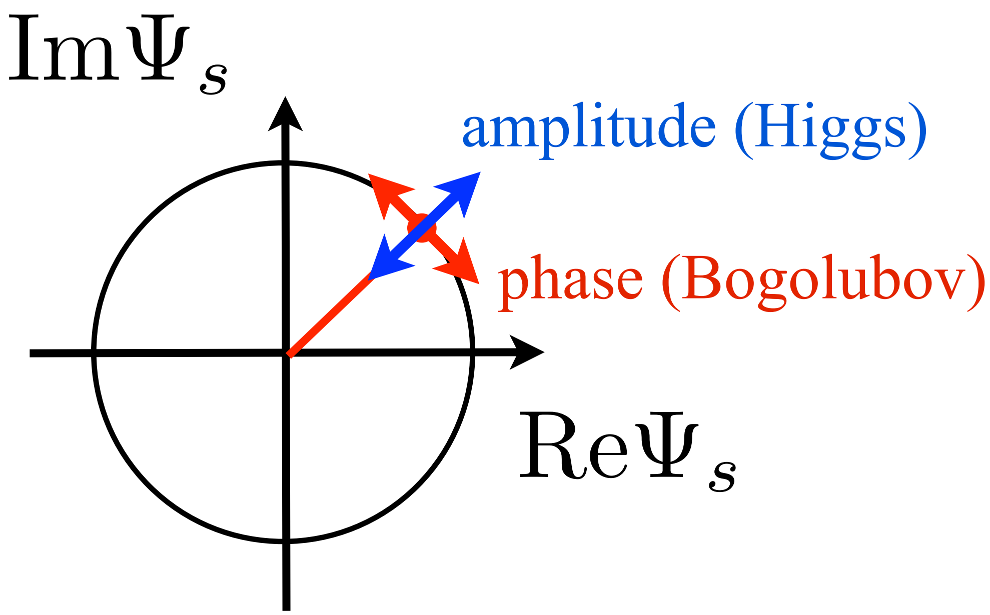
Conclusions:

The pairing between electron pockets MUST include inter-pocket pairing on equal footing with intra-pocket pairing



MK, A.V. Chubukov, Phys. Rev. Lett. **108**, 247003 (2012)

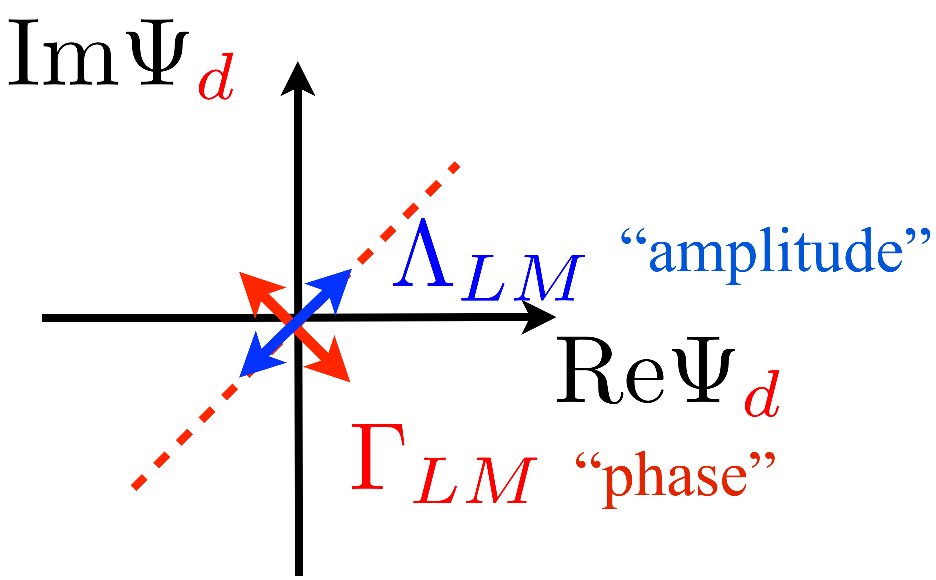
Bardasis-Schrieffer Modes (1961)



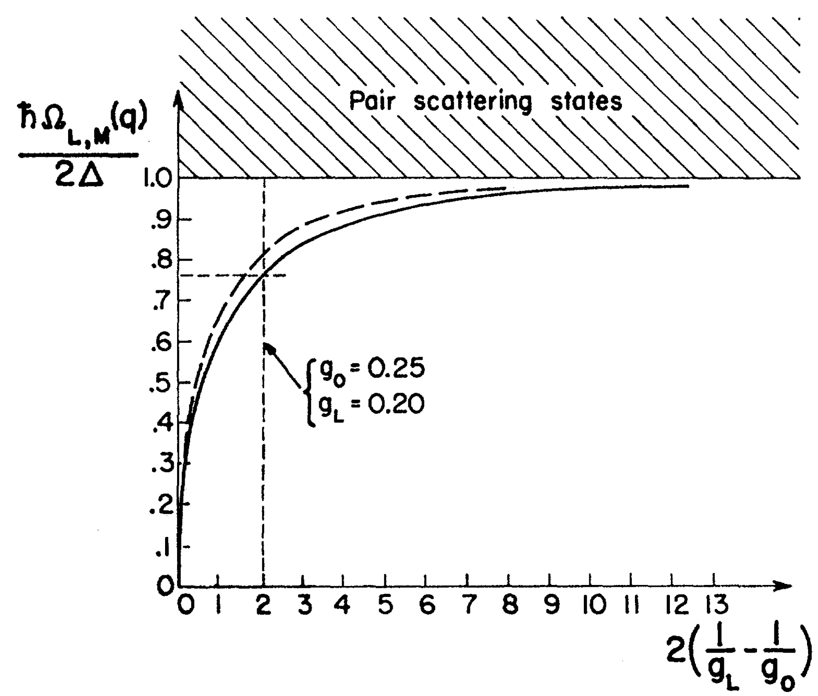
s-wave superconductor

$|\Psi_s| \neq 0$

Attraction in d-wave Cooper channel



1. In-gap "phase" mode
2. Damped decoupled "amplitude" mode

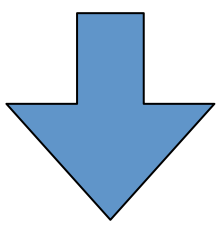


Particle-Hole Exciton

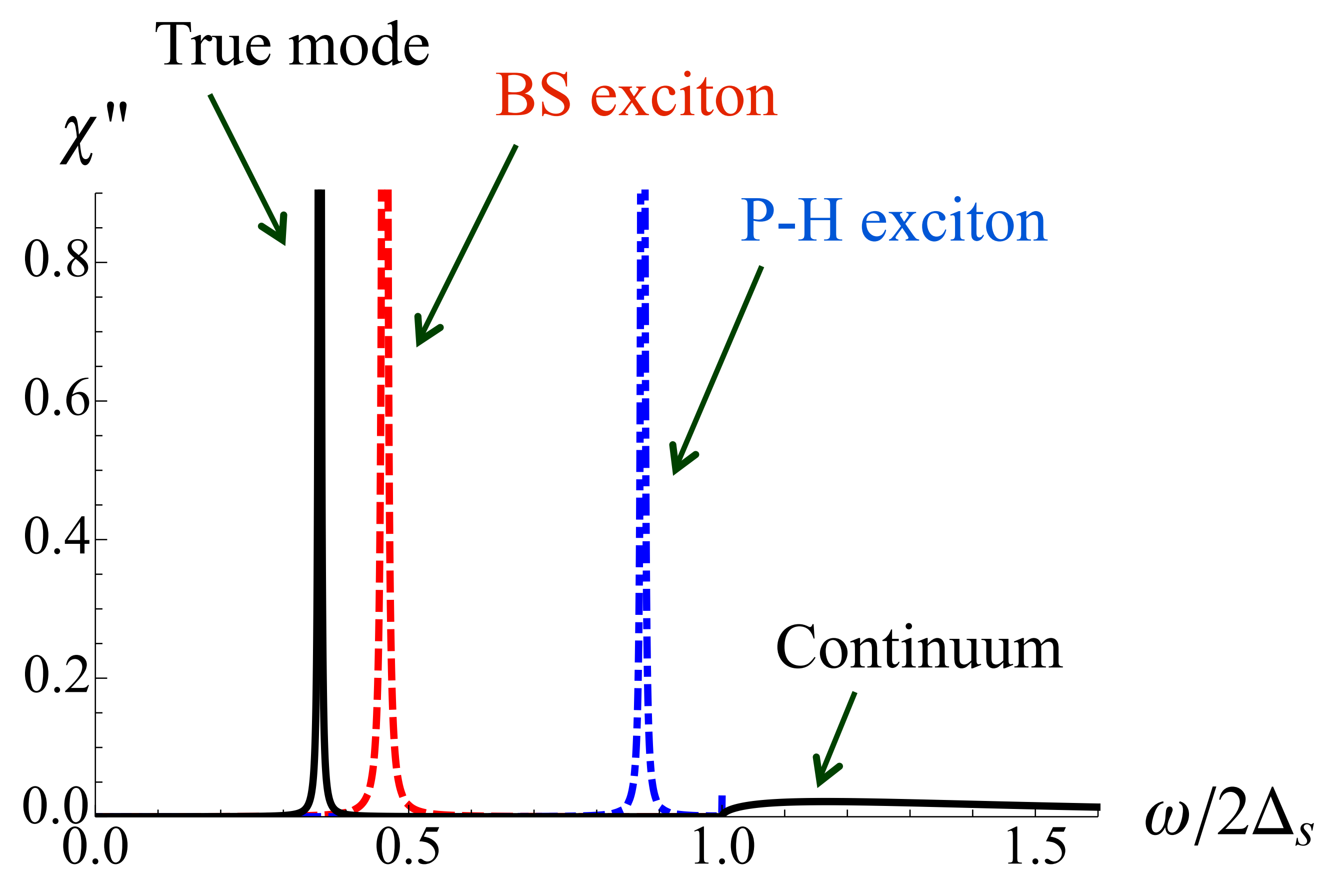
What if the P-H channel is attractive too?

Two excitons?? NO!

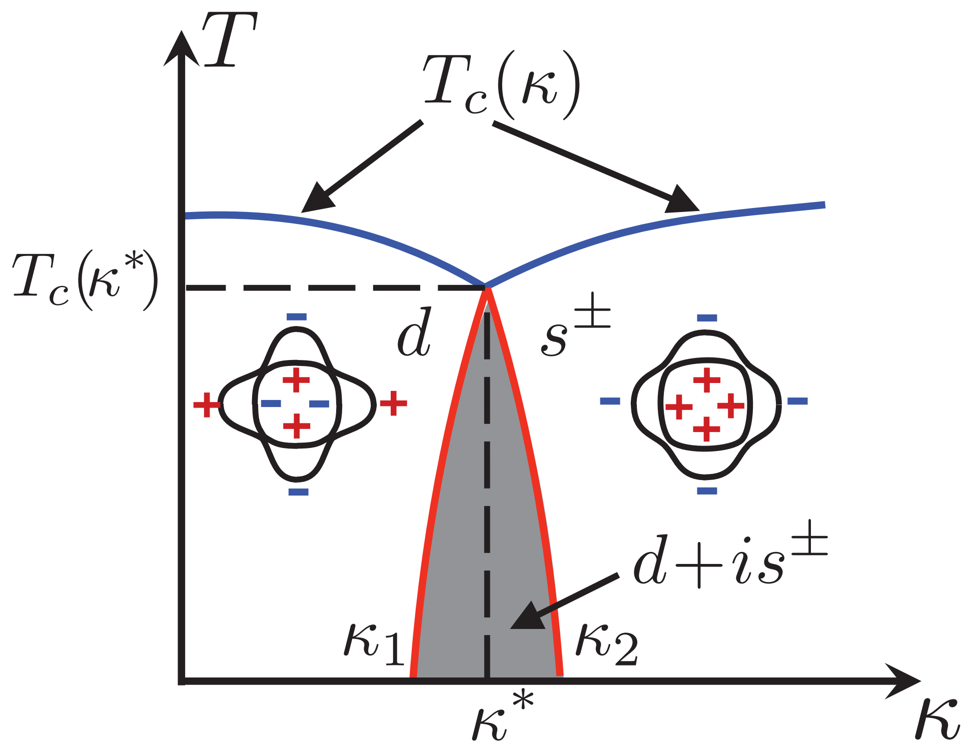
$$[\text{density}, \text{phase}]_- = i\hbar$$

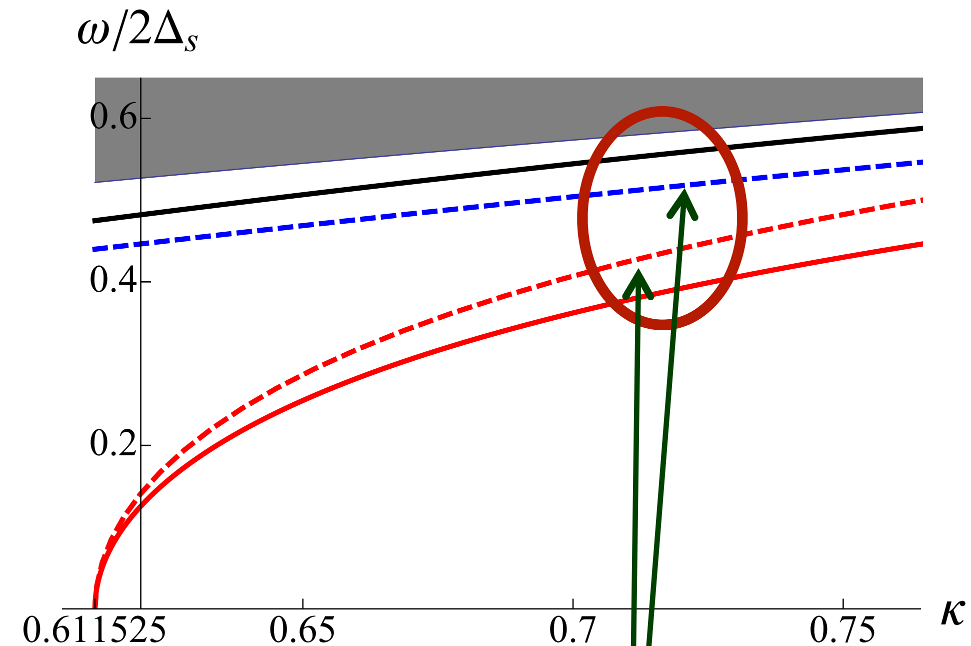


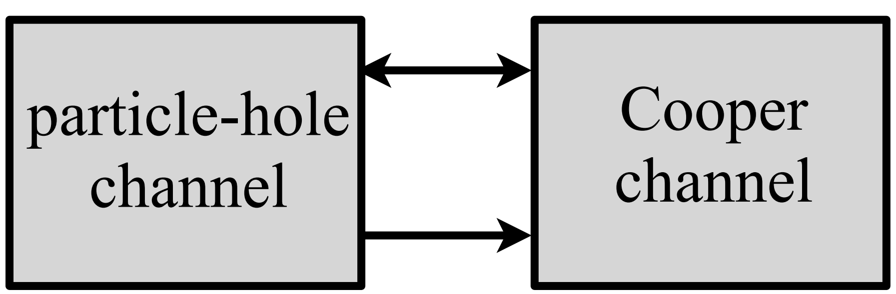
One mode



Raman in selenides: Results





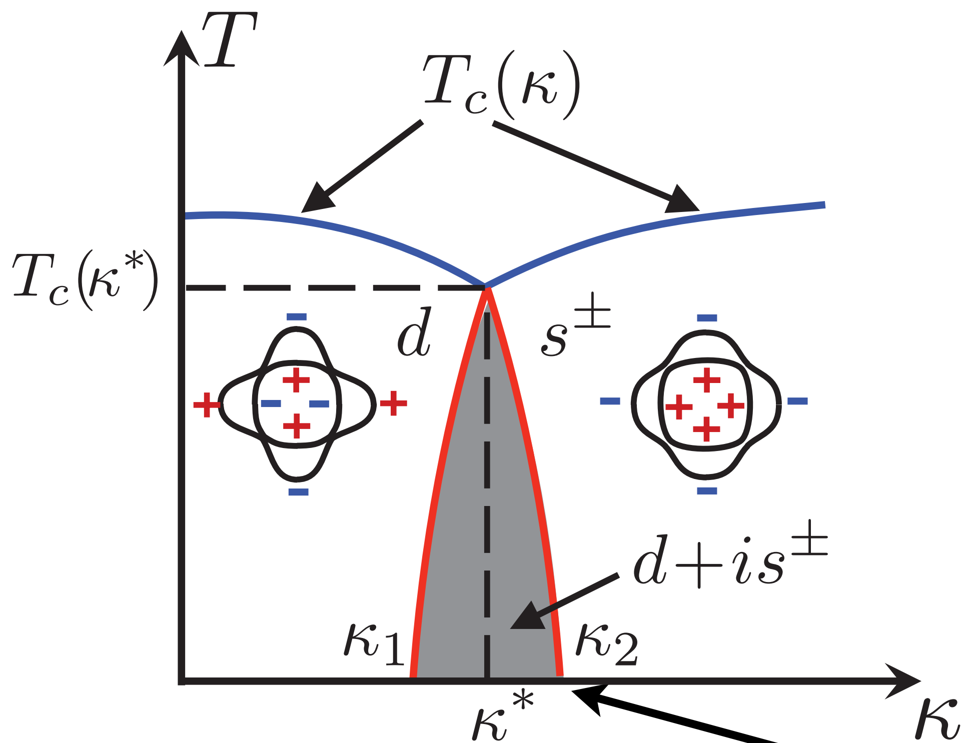


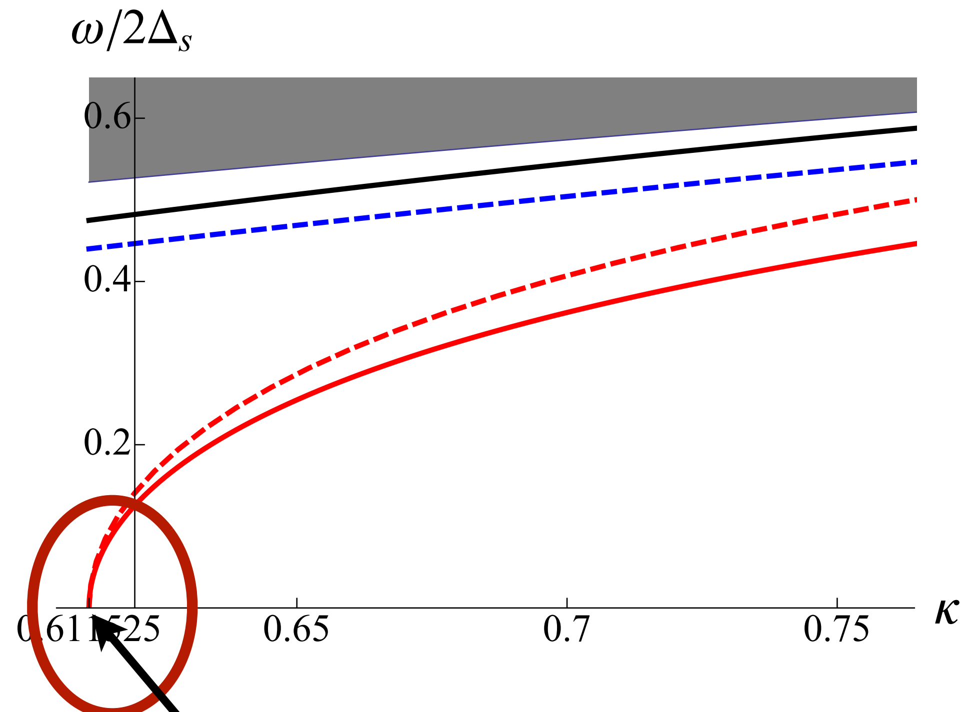
$\propto \Delta$ → No mixing → Two separate modes

Sign-Changing
Order Parameter

MK, A.V. Chubukov, G. Blumberg, PRB **89**, 245134 (2014)

Raman in selenides: Results (cont)





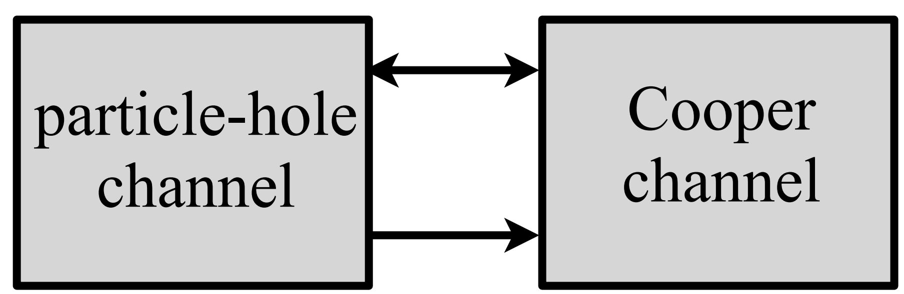
Decoupling at low frequencies

$[\text{density}, \text{phase}]_- = i\hbar$

$$[q, p]_- = i\hbar$$

$$L = p\dot{q} - H$$

$$~~p\dot{q}~~ \propto \omega$$

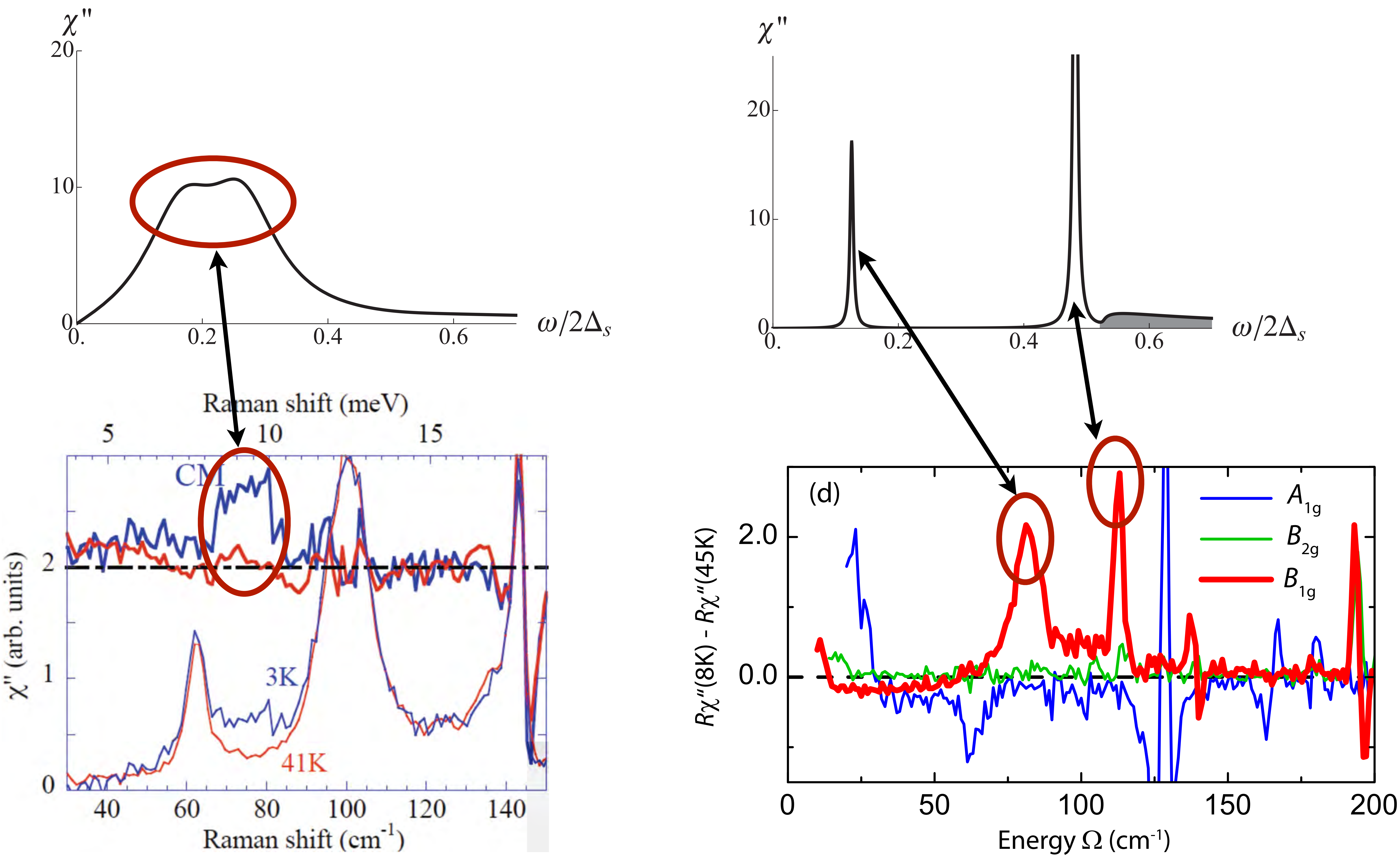


$\rightarrow 0$

$\omega \rightarrow 0$

MK, A.V. Chubukov, G. Blumberg, PRB **89**, 245134 (2014)

Raman in selenides: Results (cont)



MK, A.V. Chubukov, G. Blumberg, PRB **89**, 245134 (2014)

Conclusions II

1.Time reversal symmetry breaking → Raman active in-gap modes
2.Competing channels → sharp in-gap modes
3. Sign-changing gap → decoupling of Cooper and P-H channels