Magnetic Order in $Fe_{1+y}Te$ Compounds

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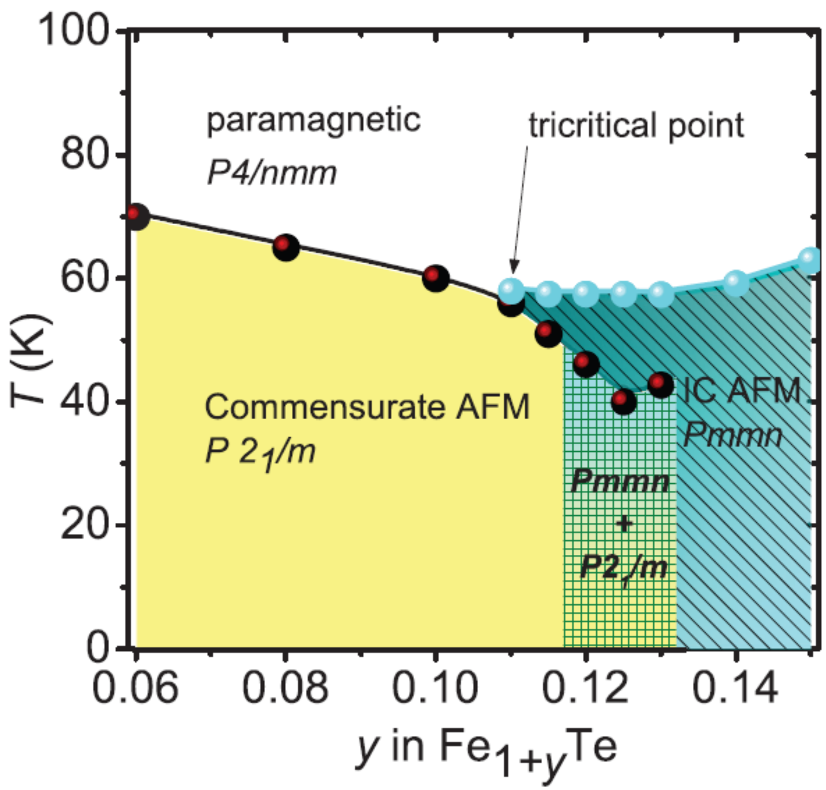
KITP, October 21 , 2014

Motivation

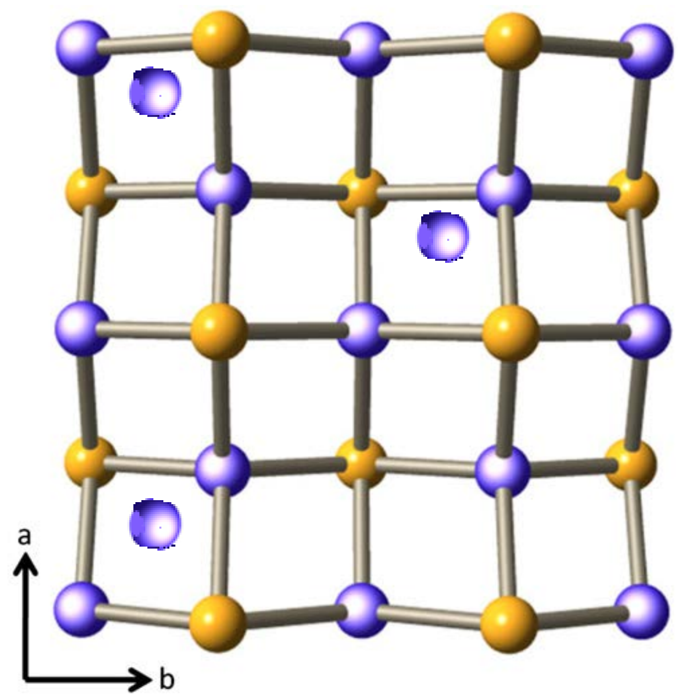
- The magnetism $Fe_{1+y}Te$ is still an open question:

($\pi/2$, $\pi/2$) double-stripy order for y<0.11

(q,q) incommensurate spiral order for y>0.11



Koz *et al*, Phys. Rev. B, 88, 094509 (2013)



Kawashima et. al., Physica B (2012)

Outline

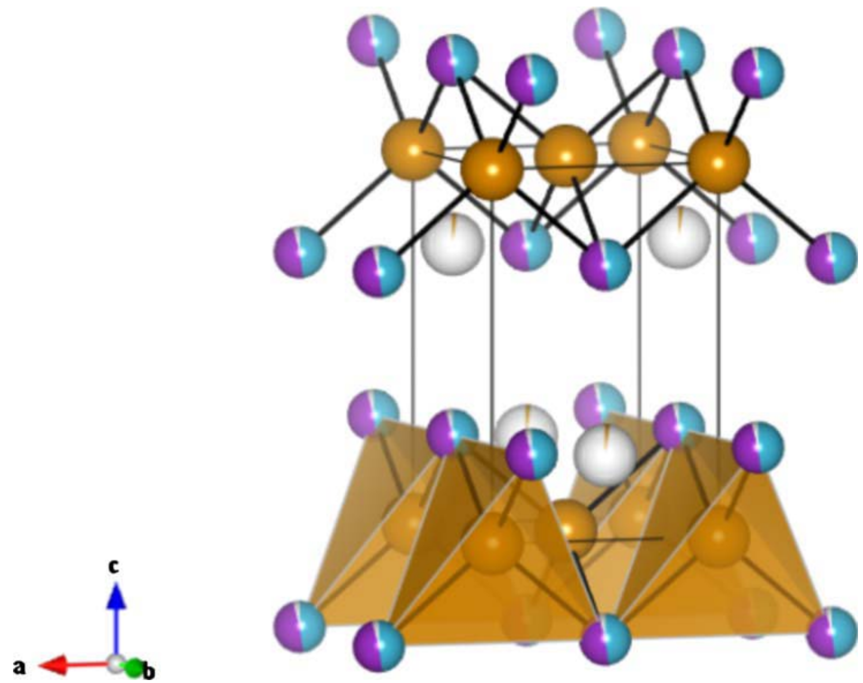
- **Introduction**
- **Double stripe in low-*y* $Fe_{1+y}Te$ compounds.**

  Classical vs Quantum approach.

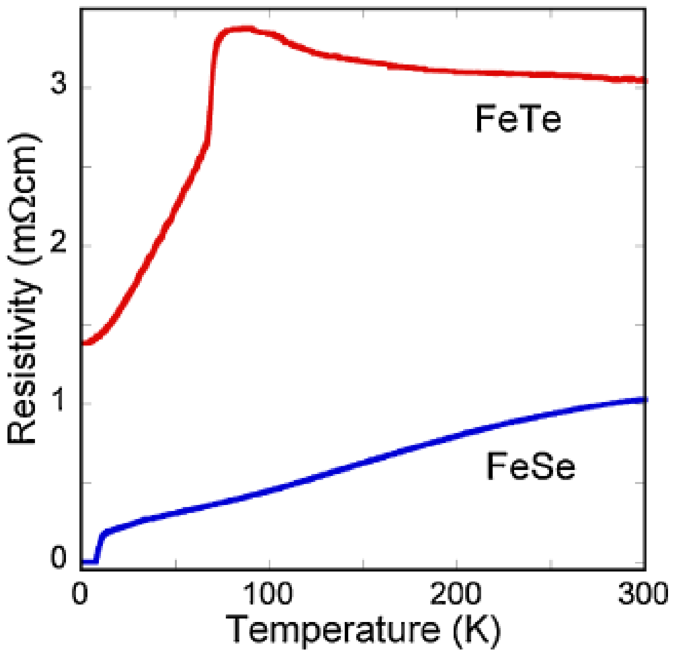
  Samuel Ducatman, Natalia Perkins, Andrey Chubukov , PRL 2012

- **Effects of Iron Excess – modified RKKY interaction causes an evolution of the magnetic structure.**

  Samuel Ducatman, Rafael Fernandes, Natalia Perkins, PRB 2014



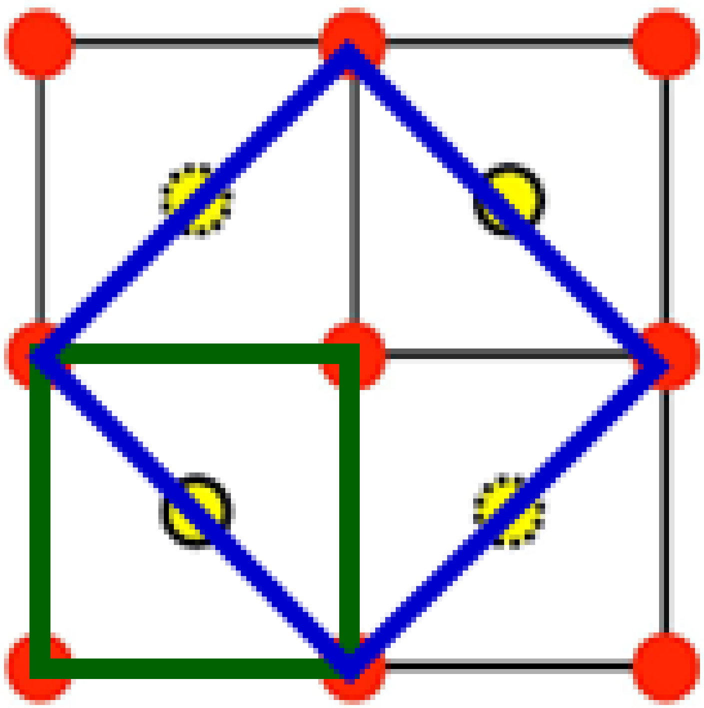
Y.Mizuguchi and Y. Takano (2010)



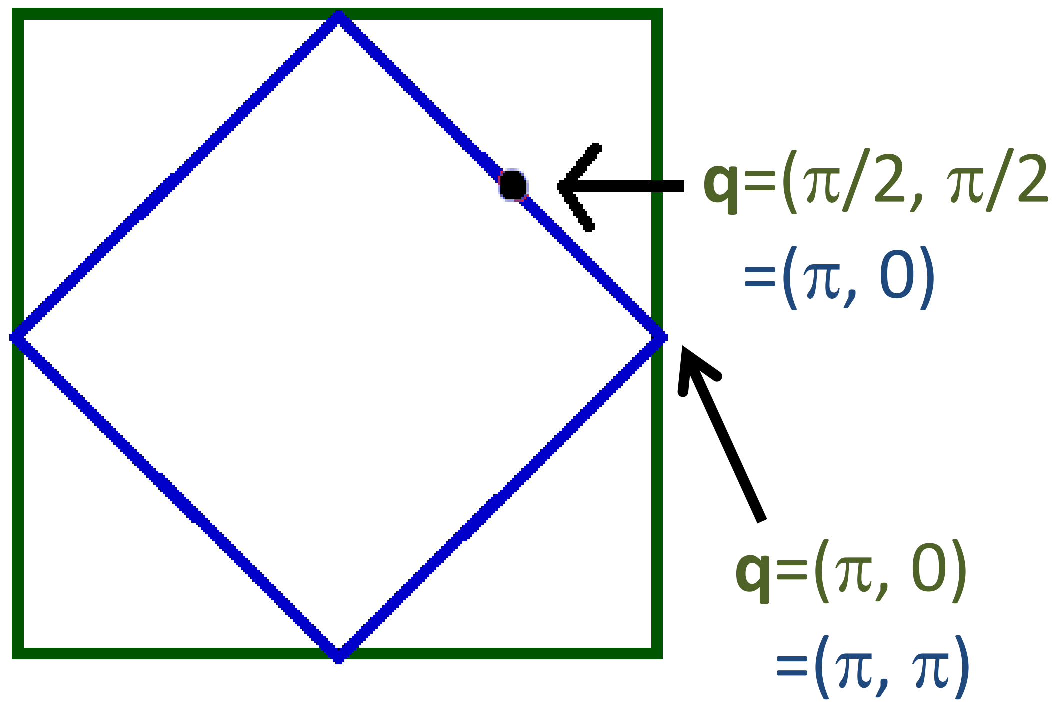
- $Fe_{1+y}Te$: the simplest structure composed of only Fe and Te layers
- Resistivity decreases with temperature (Poor Metal)
- Different q-vectors for "nesting" (π,0) or (0,π) and magnetic order (π/2,±π/2)

1 vs 2 Fe Unit cell

- Unit Cell



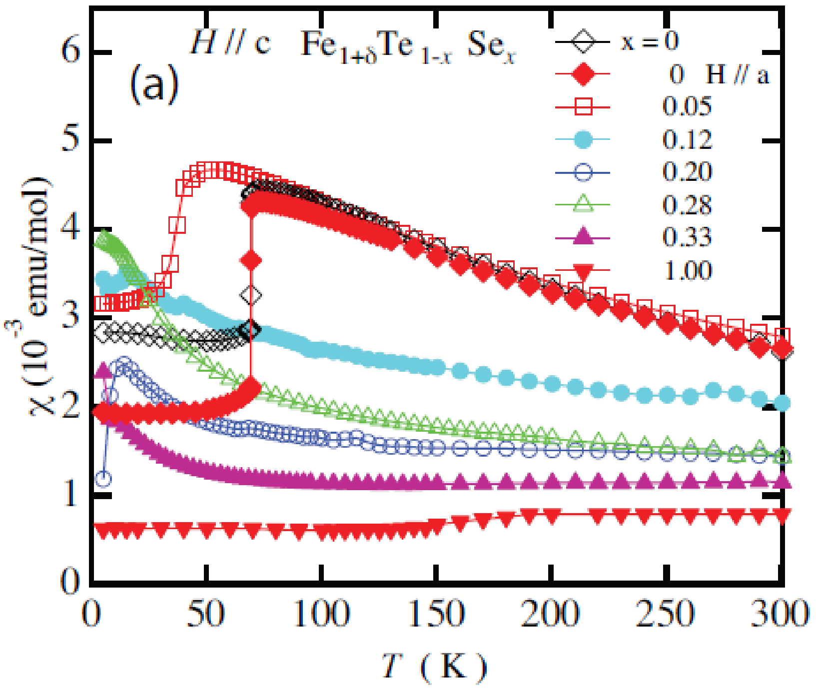
- Brillouin Zone



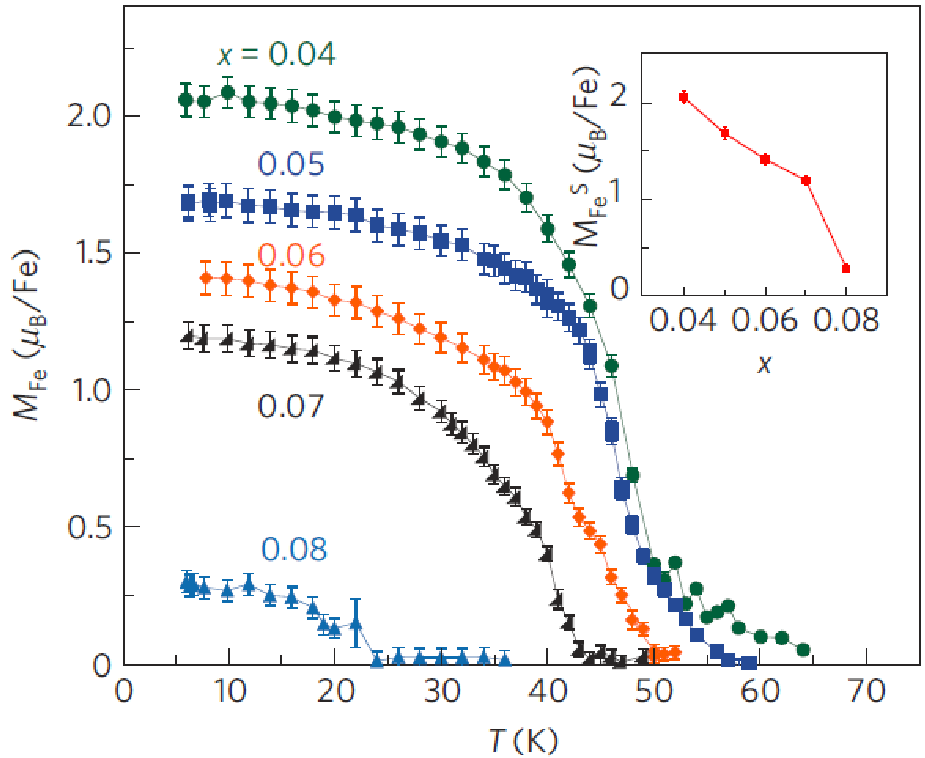
Most experimental results are presented in the folded BZ (2 Fe unit cell). We use the unfolded BZ (1 Fe unit cell).

Evidence for Local Magnetic Order

- Susceptibility shows Curie-Weiss T-dependence
- Ordered moment about $2.5\mu_B$

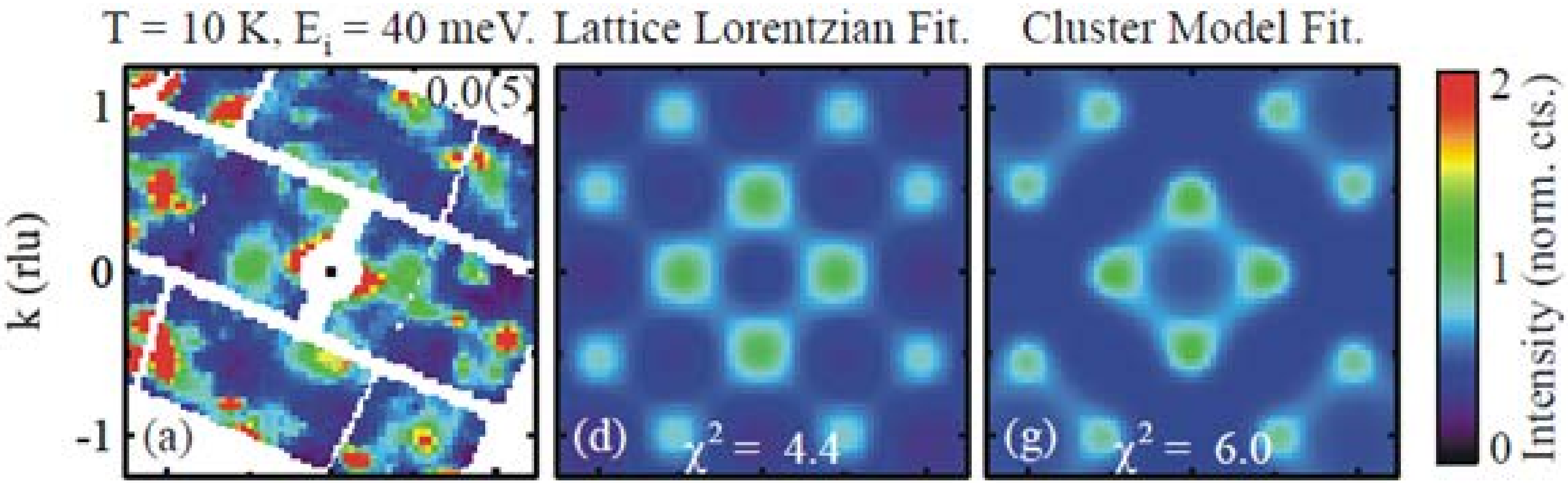


J. Yang et al, J. Phys. Soc. Jpn. **79**, 074704 (2010).



T.J. Liu et al., Nature Mater. 9, 718 (2010)

Magnetic order in FeTe has momenta $\pm(\pi/2, \pm\pi/2)$. However, this does not uniquely determine spin configuration as a generic $\pm(\pi/2, \pm\pi/2)$ order is a superposition of two different Q−vectors: $(\pi/2, -\pi/2)$ and $(\pi/2, \pi/2)$.



Zaliznyak et. al., PRL **107**, 216403 (2011)

Double stripe in low-*y* $Fe_{1+y}Te$ compounds.

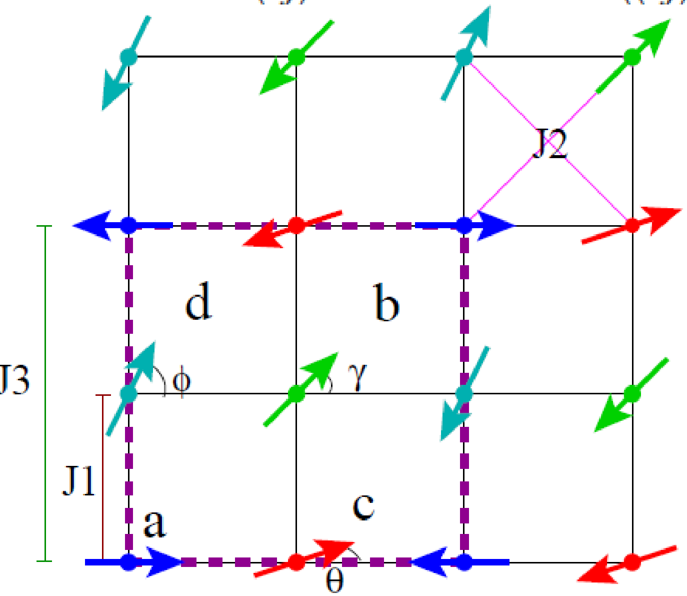
Classical vs Quantum approach.

Samuel Ducatman, Natalia Perkins, Andrey Chubukov , PRL 2012

Minimal model and classical ground state

Heisenberg J_1- J_2- J_3 Model

$$H = J_1 \sum_{\langle ij \rangle} \vec{S}_i \cdot \vec{S}_j + J_2 \sum_{\langle\langle ij \rangle\rangle} \vec{S}_i \cdot \vec{S}_j + J_3 \sum_{\langle\langle\langle ij \rangle\rangle\rangle} \vec{S}_i \cdot \vec{S}_j$$



R. Yu, *et. al* (2011);
P. Sindzingre,*et. al* (2010);
J. Reuther, *et al.*(2011)

J_3 > J_2/2>>J_1 (F. Ma, *et.al* , PRL 2009)

In this limit, the classical ground state is a spiral with the pitch vector **Q** =(±*q*,±*q*)

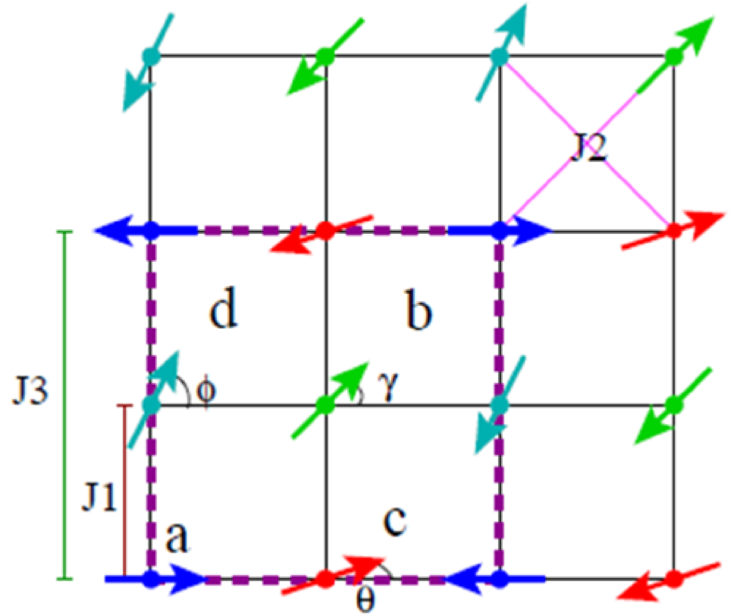
$$E_{cl} = -(2J_3 + \frac{J_1^2}{2J_2 + 4J_3})NS^2$$

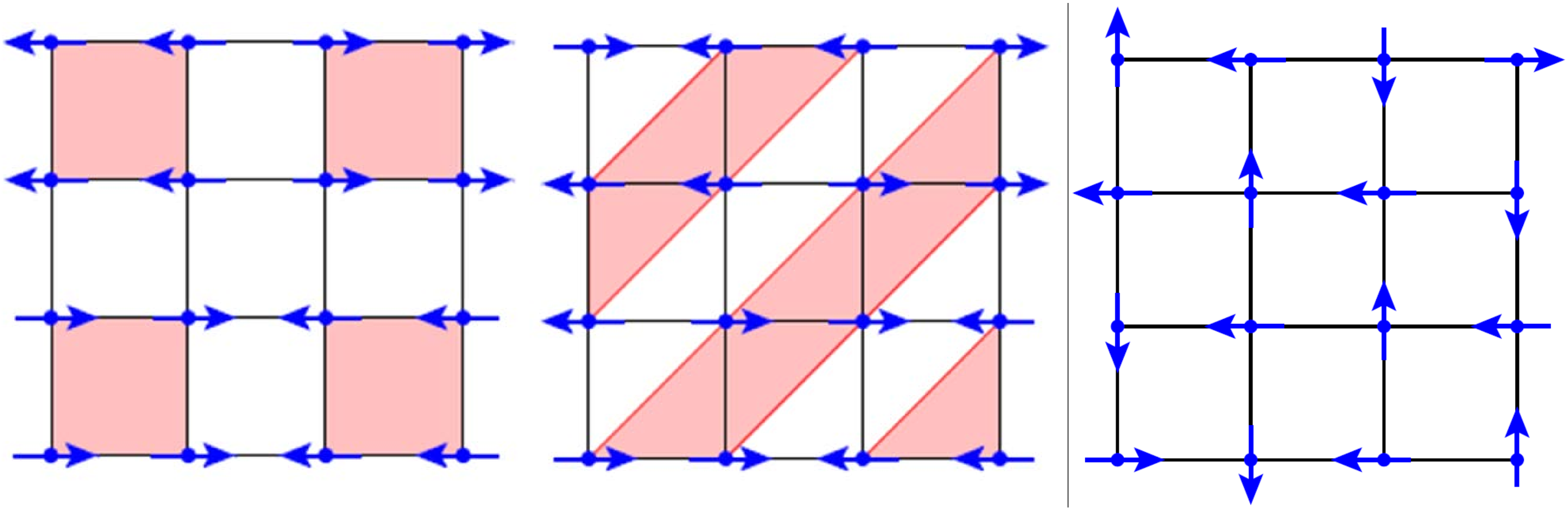
$$q = \arccos(\frac{-J_1}{2J_2+4J_3})$$

An infinite number of

q=(±π/2, ± π/2) states, all degenerate.

$E_{cl}=-2J_3NS^2$





DFT calculation: energy difference between double stripe and spiral is 0.06 meV.

How to stabilize **q**=(π/2, π/2) states and to remove the degeneracy between them?

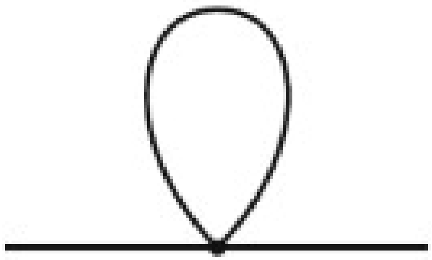
Classically:

Biquadratic term due to magnetoelastic coupling (or from a purely electronic basis)

$$H = \sum_{ij} [J_{ij} \mathbf{S_i} \cdot \mathbf{S_j} - K_{ij} (\mathbf{S_i} \cdot \mathbf{S_j})^2]$$

Quantum Mechanically:

Quantum fluctuations due to interacting spin waves

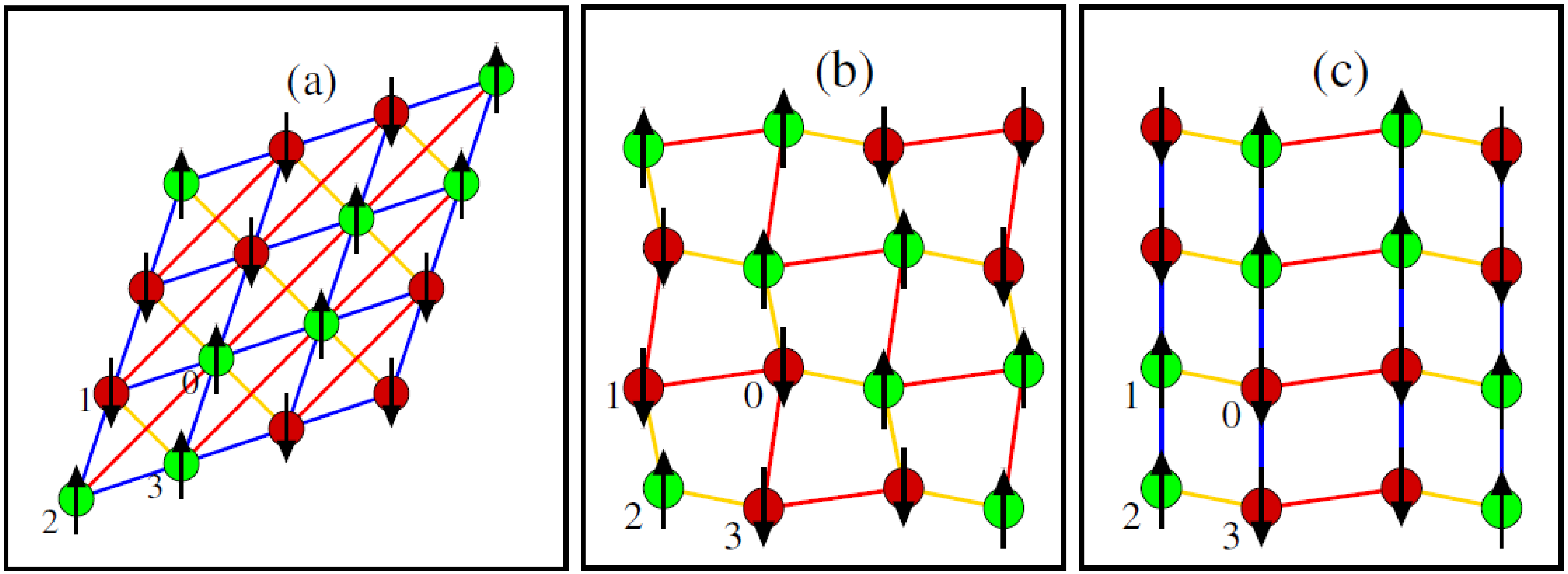


- Both mechanisms stabilize collinear structures and remove degeneracy

Magnetoelastic Couplings

Three primary lattice distortions associated with **q**=(π/2π/2) order

u_{xy} u_5 u_6/u_7



u_{xy} gives anisotropic J_2 but also biquadratic coupling along diagonals

u_5, u_6, u_7 gives anisotropic J_1 but also biquadratic coupling along sides and the ring exchange.

Magnetoelastic Hamiltonian

$$H = H_{\mathrm{M}} + H_{\mathrm{ME}} + H_{\mathrm{Elastic}}$$

$$H_{\mathrm{Elastic}} = \frac{c_{66}}{2} u_{\mathrm{xy}}^2 + \frac{\mathbf{\Omega}_1}{2}\mathbf{u}_5^2 + \frac{\mathbf{\Omega}_2}{2}(\mathbf{u}_6^2 + \mathbf{u}_7^2)$$

$$\begin{aligned} H_{\mathrm{ME}} &= g_1(\mathbf{S}_c \cdot \mathbf{S}_d - \mathbf{S}_a \cdot \mathbf{S}_b)u_{\mathrm{xy}} \\ &+ g_2[(\mathbf{S}_a \cdot \mathbf{S_c} - \mathbf{S_b} \cdot \mathbf{S}_d)\mathbf{u}_5^x + (\mathbf{S}_a \cdot \mathbf{S_d} - \mathbf{S_b} \cdot \mathbf{S}_c)\mathbf{u}_5^y] \\ &+ g_3[(\mathbf{S}_a \cdot \mathbf{S_c} + \mathbf{S_b} \cdot \mathbf{S}_d)\mathbf{u}_6^x + (\mathbf{S}_a \cdot \mathbf{S_d} + \mathbf{S_b} \cdot \mathbf{S}_c)\mathbf{u}_7^y] \end{aligned}$$

Integrating out u_{xy}, u_5, u_6, and u_7, we get effective biquadratic and ring exchange terms

$$\sum_{<ijkl>} K_{ijkl}(\mathbf{S_i} \cdot \mathbf{S_j})(\mathbf{S_k} \cdot \mathbf{S_l})$$

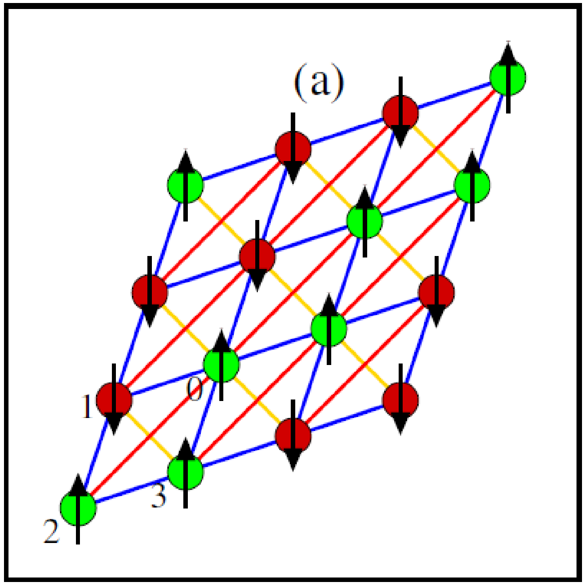
Dominant term: biquadratic coupling along diagonal due to u_{xy} distortions

I.Paul (2010)

How to stabilize **q**=(π/2, π/2) states and to remove the degeneracy between them?

Classically:

K_2 ,biquadratic coupling along diagonal, lowers the energy of bicollinear stripe



How to stabilize **q**=(π/2, π/2) states and to remove the degeneracy between them?

Quantum mechanically:

$J_1 = 0$ ➡ $J_2 - J_3$ model

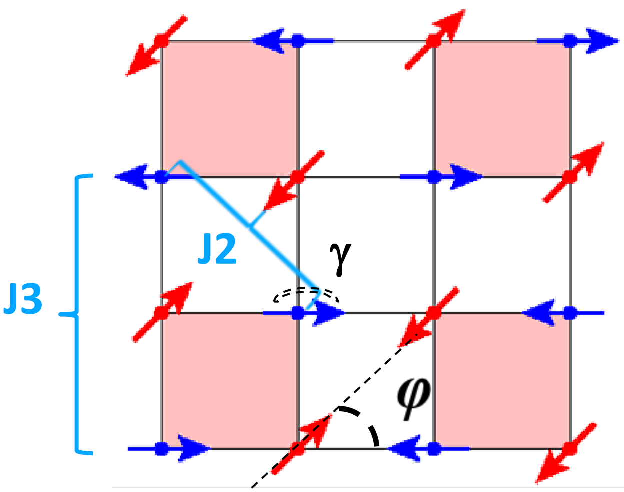
"$J_1 - J_2$"

J_2 plays the role of "J_1"

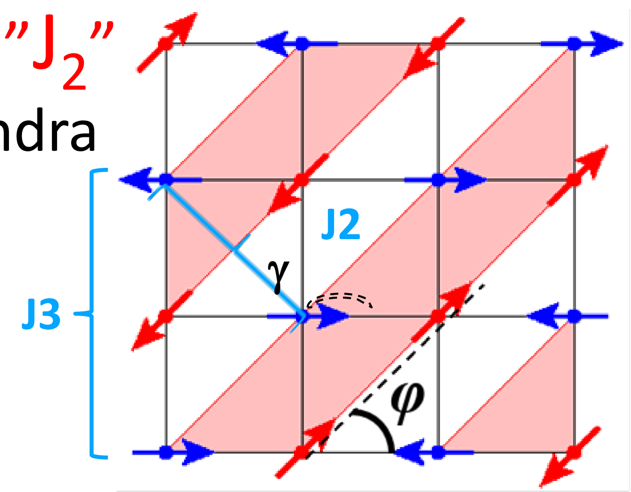
J_3 plays the role of "J_2"

Coleman, Larkin, Chandra

Generalized Plaquette



Generalized Bicollinear



For $J_3 > J_2/2$, quantum fluctuations select stripe configuration for each sublattice: the angle γ is locked at $\gamma = 0$ or $\gamma = \pi$, and the angle θ is locked to $\theta = \phi$ or $\theta = \phi + \pi$.

Order by Disorder!

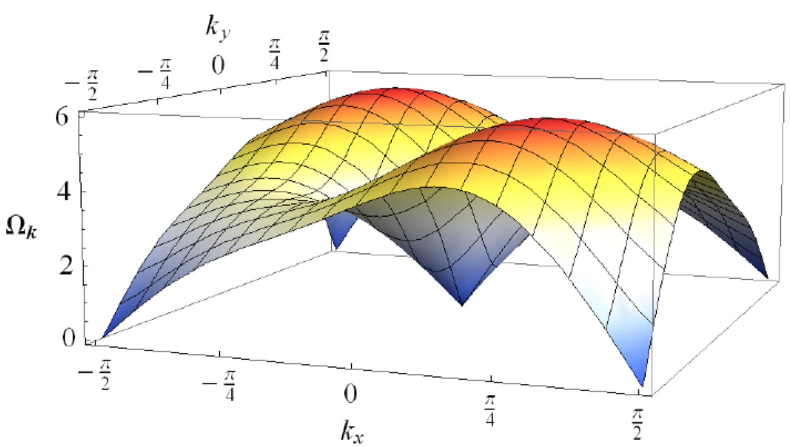
J_1 = 0: Spin-wave excitations

$$H_{sw} = S(\Omega_{\alpha\mathbf{k}}\alpha^\dagger_{\mathbf{k}}\alpha_{\mathbf{k}} + \Omega_{\beta\mathbf{k}}\beta^\dagger_{\mathbf{k}}\beta_{\mathbf{k}})$$

Even sites (↑ $\alpha^\dagger_{\mathbf{k}}$) Odd sites (↑ $\beta^\dagger_{\mathbf{k}}$)

$$\Omega_{\mathbf{k}} = S(A^2_{\mathbf{k}} - B^2_{\mathbf{k}})^{1/2},\ \ A_{\mathbf{k}} = 4J_3 + 2J_2\cos(k_x + k_y),$$
$$B_{\mathbf{k}} = 2J_2(\cos 2k_x + \cos 2k_y) + 2J_2\cos(k_x - k_y).$$

- Linear Spin Wave (LSW) Theory: two spectrums, one for even sites and one for odd sites



Nodes at ±(π/2,±π/2), but some of them are accidental

1/S Corrections

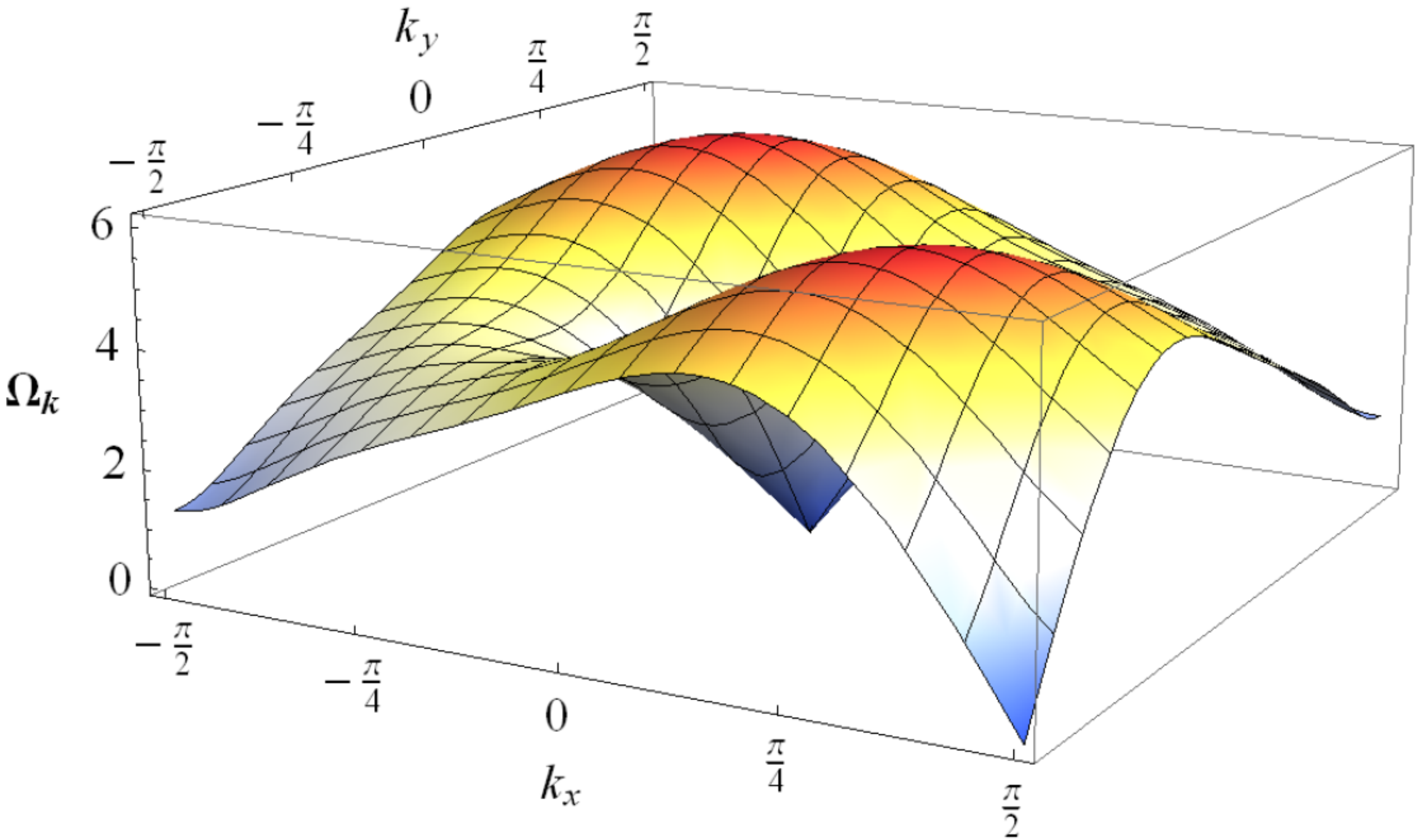
$$H_4 = \sum_{<ij>} J_{ij}[-\frac{1}{2}a_i^\dagger a_i a_j^\dagger a_j + \ldots]$$ ⇨

Performing Hartree-Fock

$$H_4 = \sum_{<ij>} J_{ij}[-\frac{1}{2}a_i^\dagger a_i < a_j^\dagger a_j > -\frac{1}{2}a_i a_j < a_i^\dagger a_j^\dagger > + \ldots]$$

- Gaps open at “accidental zeroes”

1/S Corrections



Small J_1

- J_1 provides a coupling between two sublattices
- J_1 is introduced peturbatively, and only leads to a strong renormoralization in the spectra near the Goldstone modes.

Two cases:

1) The excitations have Goldstone modes at the same q vectors — case for diagonal double stripe (bicollinear)
2) The Goldstone modes have different q-vectors — case for orthogonal double stripe (plaquette)

Spectrum of Bicollinear State

$$\Omega^{\alpha}_{\tilde{\mathbf{k}}} = \Omega^{\beta}_{\tilde{\mathbf{k}}}$$

$$H_2 = \frac{S}{2}\sum_{\mathbf{k}}[\omega_{\mathbf{k}}(\alpha^{\dagger}_{\mathbf{k}}\alpha_{\mathbf{k}} + \alpha_{-\mathbf{k}}\alpha^{\dagger}_{-\mathbf{k}} + \beta^{\dagger}_{\mathbf{k}}\beta_{\mathbf{k}} + \beta_{-\mathbf{k}}\beta^{\dagger}_{-\mathbf{k}})$$
$$+\Delta_{\mathbf{k}}\ (-\alpha^{\dagger}_{\mathbf{k}}\beta_{\mathbf{k}} - \alpha_{-\mathbf{k}}\beta^{\dagger}_{-\mathbf{k}} + \alpha^{\dagger}_{-\mathbf{k}}\beta^{\dagger}_{\mathbf{k}} + \alpha_{\mathbf{k}}\beta_{-\mathbf{k}})$$
$$+\Delta^{*}_{\mathbf{k}}\ (-\alpha^{\dagger}_{-\mathbf{k}}\beta_{-\mathbf{k}} - \alpha_{\mathbf{k}}\beta^{\dagger}_{\mathbf{k}} + \alpha^{\dagger}_{\mathbf{k}}\beta^{\dagger}_{-\mathbf{k}} + \alpha_{-\mathbf{k}}\beta_{\mathbf{k}})$$

$$H_2 = \frac{S}{2}\sum_{k}[\epsilon_k + 2\tilde{\omega}_{1_k}\tilde{\alpha}^{\dagger}_k\tilde{\alpha}_k + 2\tilde{\omega}_{2_k}\tilde{\beta}^{\dagger}_k\tilde{\beta}_k]$$

$$\tilde{\omega}^2_{1,2_{\mathbf{k}}} = \omega^2_{\mathbf{k}} \pm 2\sqrt{\omega^2_{\mathbf{k}}|\Delta_{\mathbf{k}}|^2 - 4|Re[\Delta_{\mathbf{k}}]Im[\Delta_{\mathbf{k}}]|^2}$$

$$\tilde{\omega}_{1,2}(\frac{\pi}{2} + \tilde{k}, \frac{-\pi}{2} - \tilde{k}) = 4\sqrt{\pm\sqrt{J_1^2\tilde{k}^2((2+J_2)^2 - J_1^2\cos^2\theta)}}$$

Interacting Goldstone bosons with $\Delta_k \sim J_1$

Instability in spectrum near **q**=(π/2, π/2), grows as

where $\tilde{k}$ is distance from Goldstone point $\sqrt{\tilde{k}}$

Spectrum of Plaquette

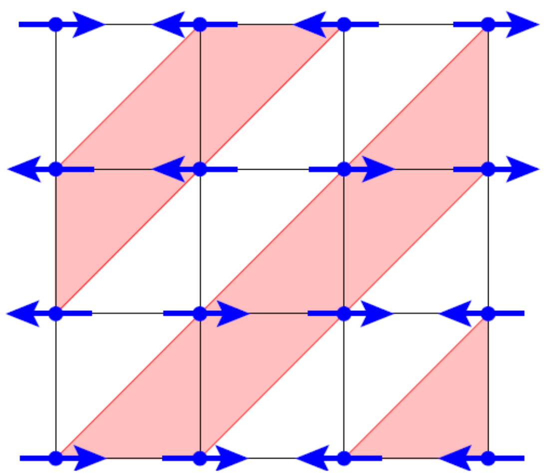
$$E_{1,2}^2 = \frac{1}{2}\Big((\Omega_{\tilde{k}}^{\alpha})^2 + (\Omega_{\tilde{k}}^{\beta})^2 \pm \sqrt{((\Omega_{\tilde{k}}^{\alpha})^2 - (\Omega_{\tilde{k}}^{\beta})^2)^2 + 16(\Delta_{\tilde{\mathbf{k}}}^{QDS})^2 \Omega_{\tilde{k}}^{\alpha} . \Omega_{\tilde{k}}^{\beta}} \Big)$$

One solution is gapped to order 1/S, the other is linear in $\tilde{k}$ with the stiffness which differs from its value at $J_1 = 0$ by $O(J_1 S/J_3)$.

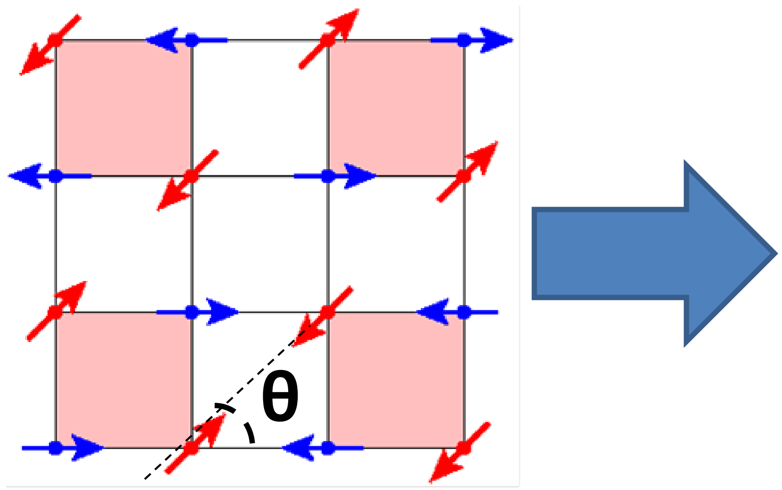
The Plaquette states are stable as long as $J_1 S/J_3$ is small. Largest energy renormalization for collinear plaquette

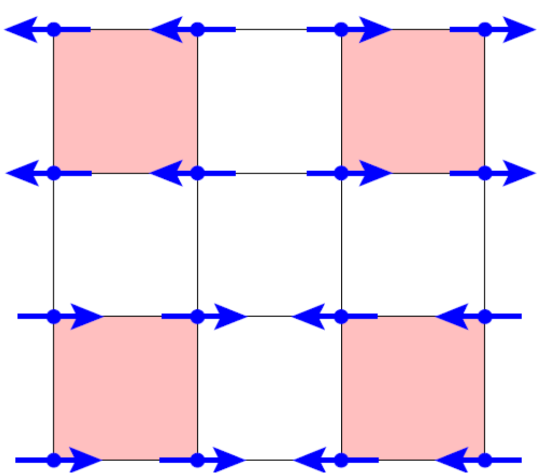
Results

For isotropic case, bicollinear structure unstable to quantum fluctuations. Lattice distortions probably stabilize this state



- Quantum fluctuations select plaquette order

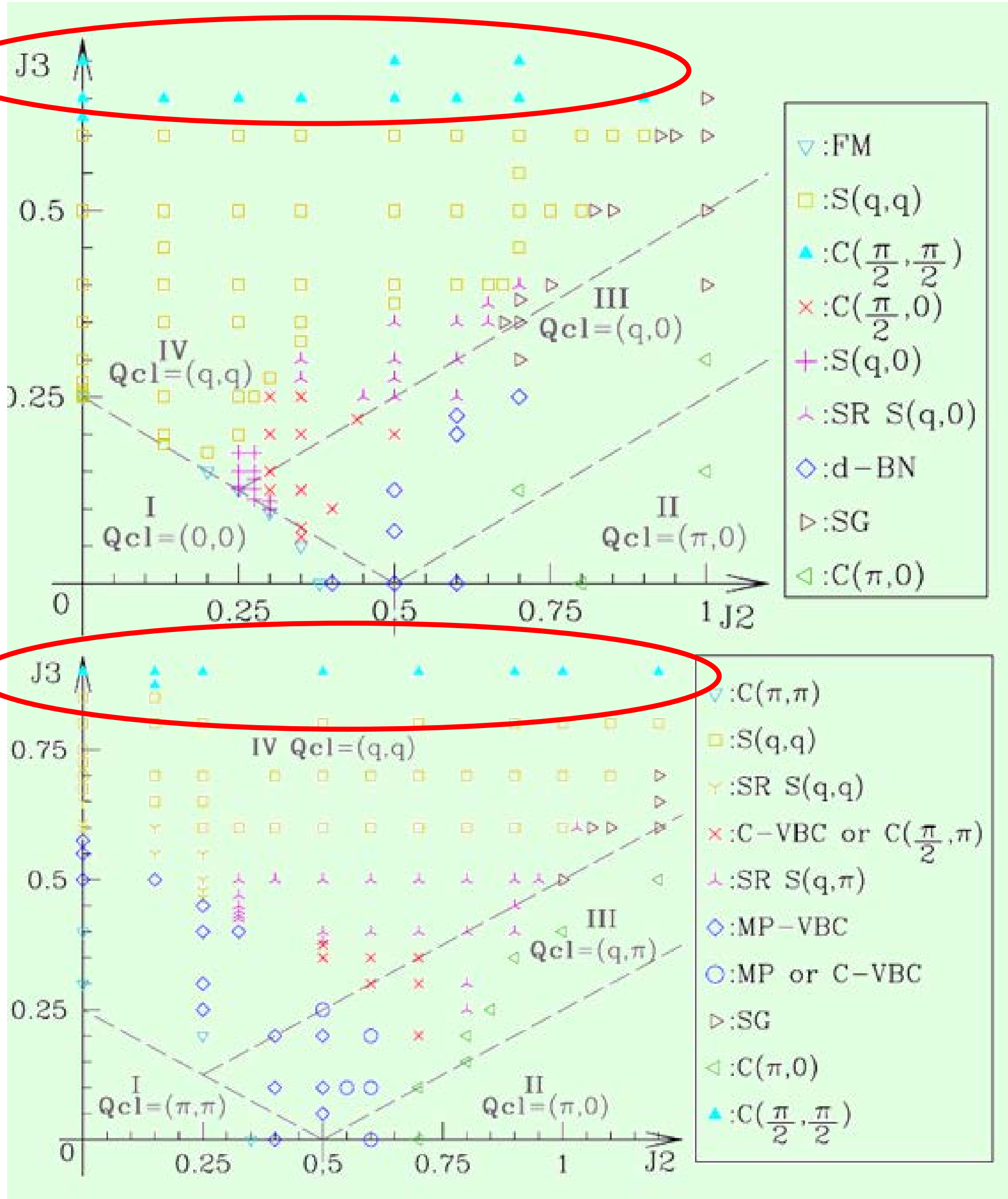




(π/2, π/2) order found in exact diagonalization

J_1=-1

J_1=1



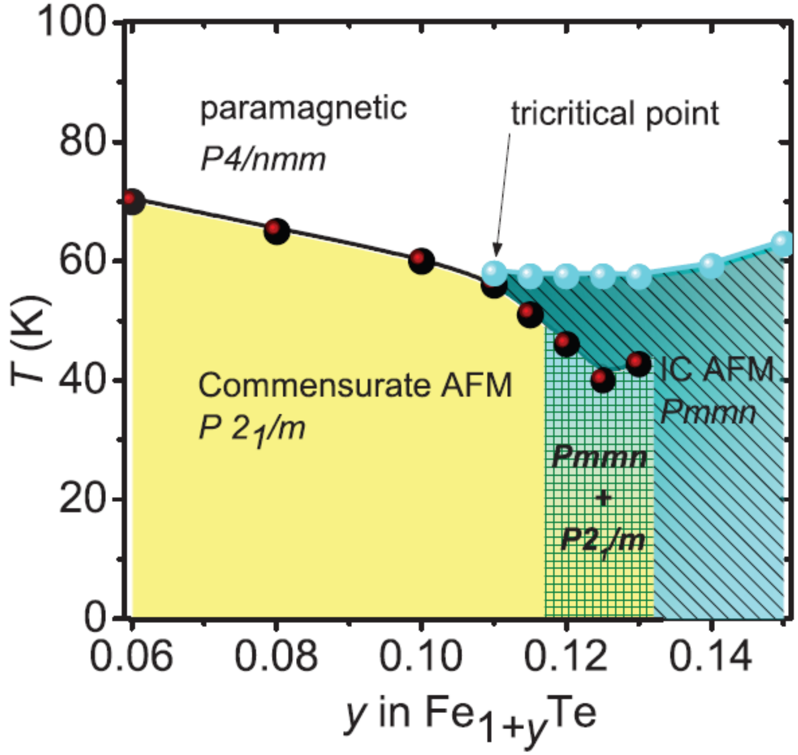
P. Sindzingre, N. Shannon, T.Momoi (2009)

- **Effects of Iron Excess – modified RKKY interaction causes an evolution of the magnetic structure.**

S. Ducatman, R. Fernandes, N. Perkins, PRB 2014

Magnetic Transitions in $Fe_{1+y}Te$

- Magnetic and structural lowering of symmetry coincide
- At y<0.11, 1st order PT from paramagnet to **q**=(π/2, π/2) state
- y>0.11, 2nd order PT to incommensurate spiral state with **q**=(π/2 – Δ, π/2 - Δ), magnetic order varies with T
- Δ locks at 0.02 for low T



Koz et al., PRB **88**, 094509 (2013)

Zaliznyak et al., PRB **85**, 085105 (2012)

How can we model Iron excess?

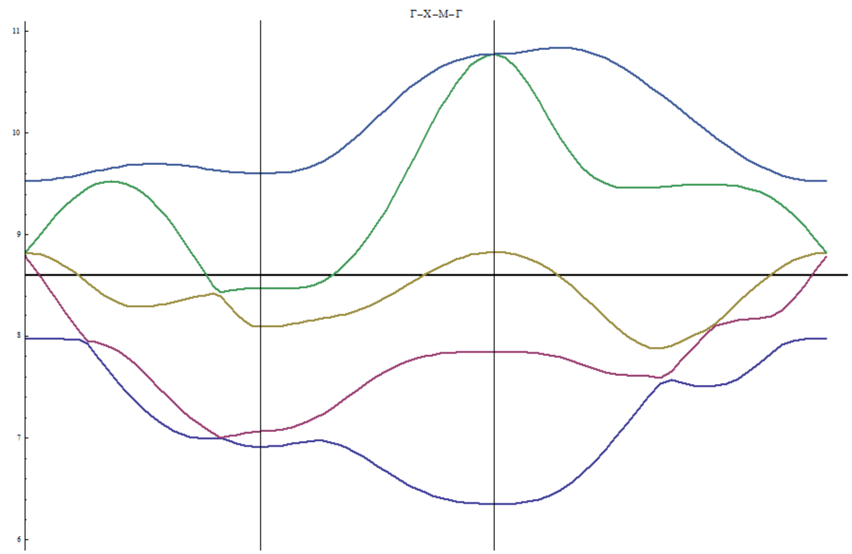
- Local spin model does not capture evolution from iron excess, itinerant model does not capture correct magnetic order
- Recent DMFT calculations of Lanata et al suggested Hund's coupling driven orbital selected localization at $T>T_N$.
- Consider hybrid model: Coexistence of local spins and itinerant electrons

Haule et al, New J. Phys. **11**, 025021 (2009)

Lanata et al, PRB **87**, 045122 (2013)

Localization of electrons

- FeTe: multi-orbital nature of degrees of freedom
- The x^2-y^2, $3z^2$–r^2 orbitals are almost localized due to narrow bandwidth and larger interactions. xy, yz, zx are still itinerant.
- We use the tight binding (TB) model of F. Wang et al, PRB **81**, 184512 (2010), and project out the x^2-y^2, $3z^2$–r^2 orbitals



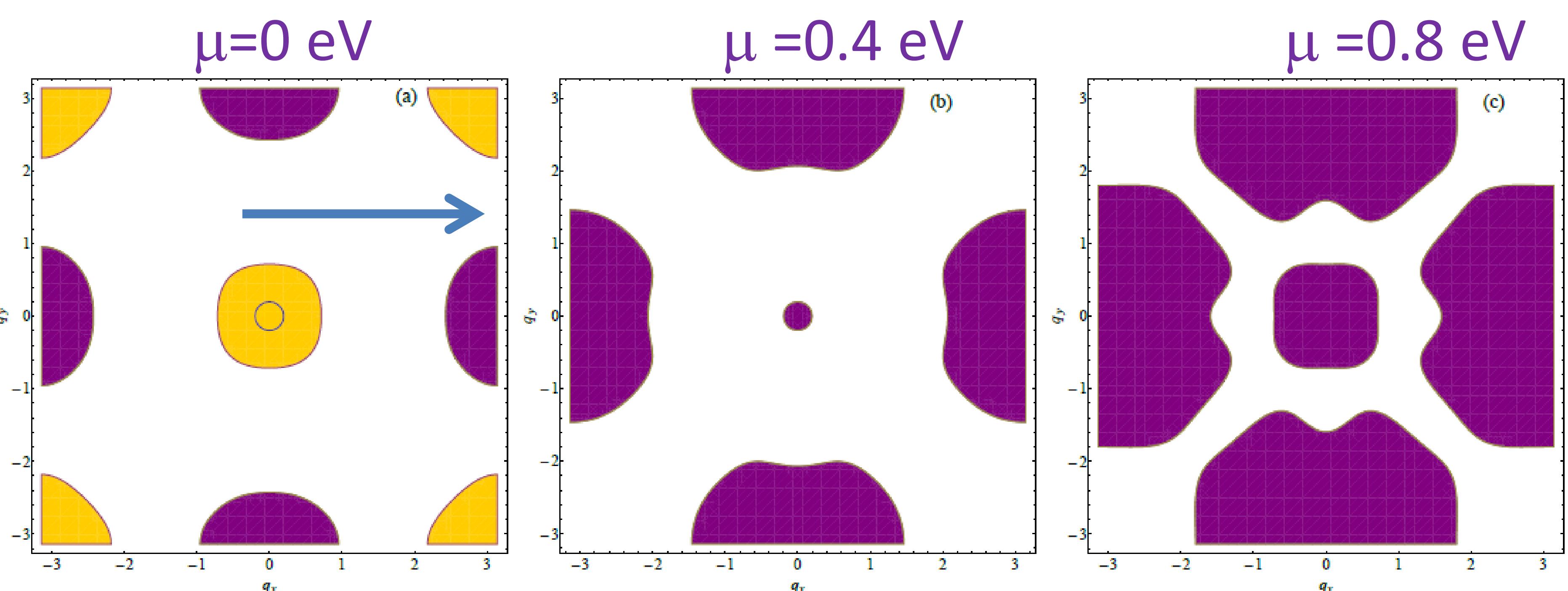
Fermi Surfaces
(Virtual crystal approximation)

Purple: electron Pockets

Yellow: hole Pockets

Nesting vector of (π,0), no perfect nesting.

(π,0) is not the Q-vector associated with magnetic order



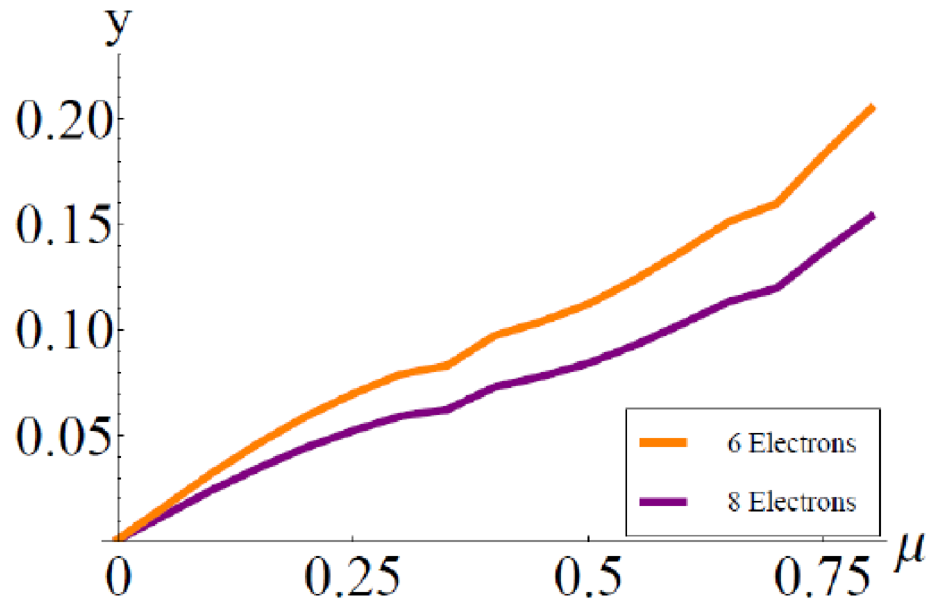
Increasing y

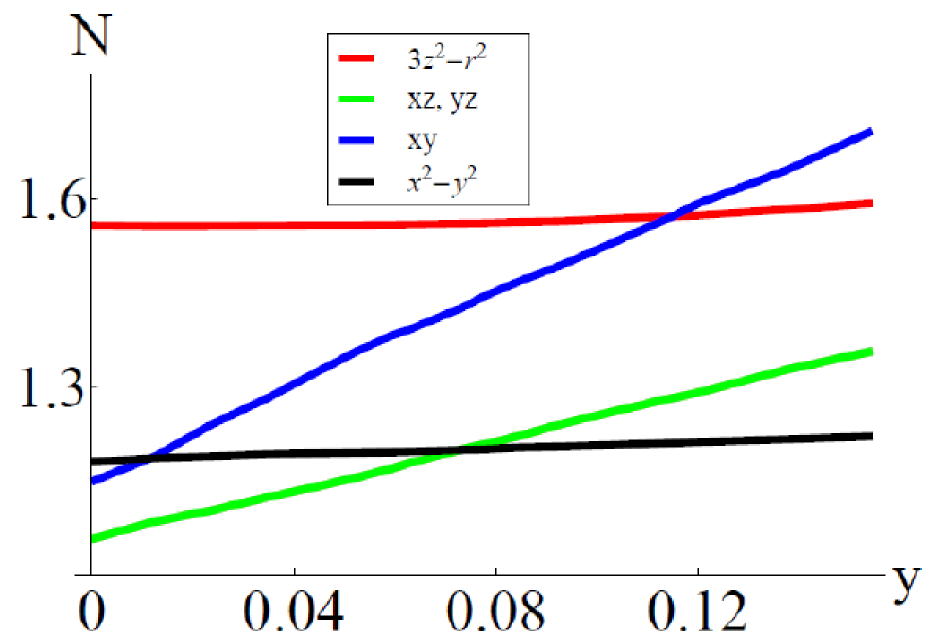
- y>0 increases the number of electrons. Excess iron donates 8 electrons per site.

Savrasov et al, PRL **103,** 067001 (2009)

P. Singh et al PRL **104**, 099701 (2010).

- We increase μ, calculate occupation number to find y.
- The extra electrons barely change the occupation of x^2-y^2, $3z^2-r^2$ orbitals





Hybrid Model: Local Moments in Multiband Correlated Electron sea

$$H = \boxed{H_{spin}} + H_{itinerant} + H_{coupling}$$

- Spin-Spin Interaction:

$$H_{spin} = \sum_{<ij>} J_{ij}\mathbf{S_i}\cdot\mathbf{S_j} + \sum_{<ijkl>} K_{ijkl}(\mathbf{S_i}\cdot\mathbf{S_j})(\mathbf{S_k}\cdot\mathbf{S_l})$$

- J_{ij}: J_1-J_2-J_3 superexchange couplings
- K_{ijkl}: Biquadratic and ring exchange terms arise from magnetoelastic effects
- S_j localized spins from electrons on $x^2 - y^2$, z^2 orbitals.

Assumption: S=1

Hybrid Model: Local Moments in Multiband Correlated Electron sea

$$H = H_{spin} + \boxed{H_{itinerant}} + H_{coupling}$$

- Effective 3 band Hubbard Model (after projection) with onsite Interactions

$$H_{itinerant} = H_0 + H_{int}$$

$$H_0 = \sum_{\mathbf{k},a,b,\sigma} \left(t^{ab}_{\mathbf{k}\sigma} c^{\dagger}_{\mathbf{k}a\sigma} c_{\mathbf{k}b\sigma} + h.c. \right)$$

$$H_{int} = \frac{1}{2} \sum_{i,ab\sigma\sigma'} (U_{ab} c^{\dagger}_{ia\sigma} c_{ia\sigma} c^{\dagger}_{ib\sigma'} c_{ib\sigma'} + J_{ab} c^{\dagger}_{ia\sigma} c_{ib\sigma'} c^{\dagger}_{ia\sigma'} c_{ib\sigma})$$

Hybrid Model: Local Moments in Multiband Correlated Electron sea

$$H = H_{spin} + H_{itinerant} + \boxed{H_{coupling}}$$

$$H_{coupling} = -J_H \sum_{j,a} \mathbf{S}_j \cdot \sigma_{ja}$$

- Coupling between local and itinerant moment arises from Hund's coupling
- S_j localized spins from electrons on $x^2 - y^2$, z^2 orbitals.

Assumption: S=1

- σ_{ja} itinerant electrons with orbital a=xy, yz, zx.

Derivation of an effective low energy theory

- Integrating out the itinerant electrons, we obtain additional long range spin-spin interactions

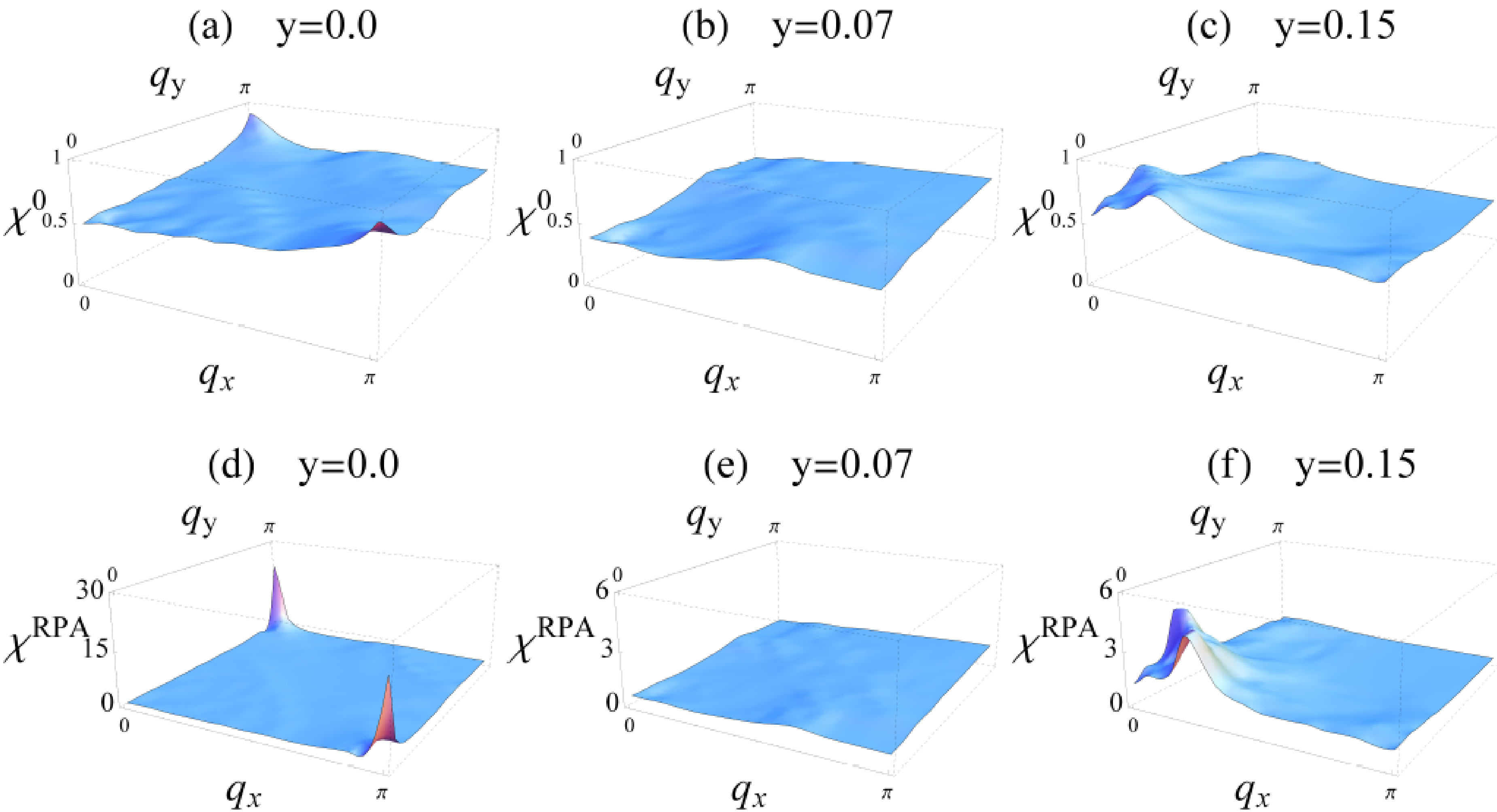
$$H_{RKKY} = \sum_{<ij>} J_{ij}^{RKKY} \mathbf{S}_i \mathbf{S}_j$$

$$J_{ij}^{RKKY}(R_i - R_j) = -J_H^2 \sum_{\mathbf{q}} e^{\imath(\mathbf{R}_i - \mathbf{R}_j)\mathbf{q}} \chi(\mathbf{q})$$

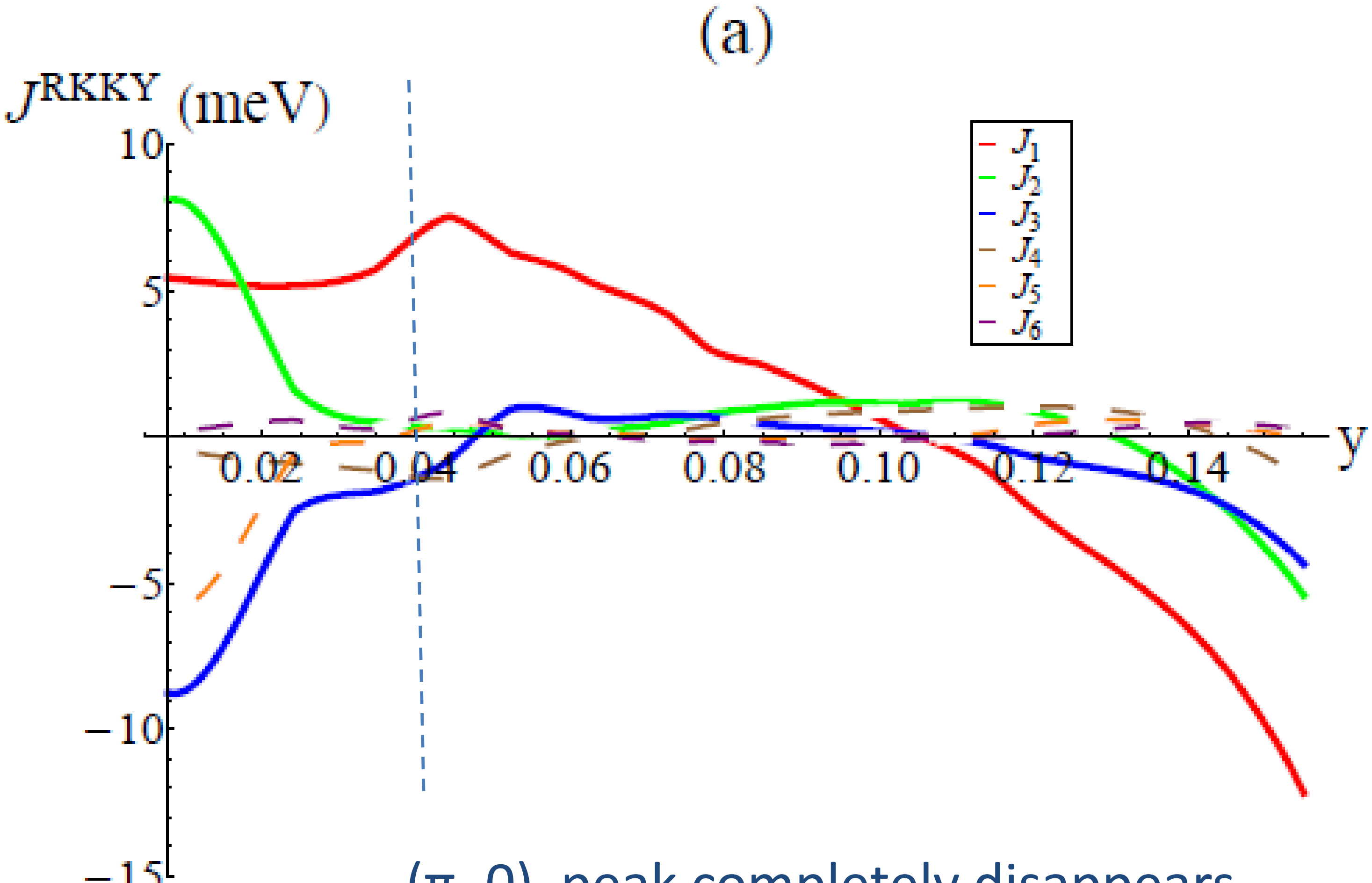
χ (q)- Pauli susceptibility computed using the tight-binding model

$$\chi_{aa'bb'}(\mathbf{q},\omega) = \chi^0_{aa'bb'}(\mathbf{q},\omega) + \chi^0_{aa'cc'}(\mathbf{q},\omega)\, U_{cc'dd'}\, \chi_{dd'bb'}(\mathbf{q},\omega)$$

Results (Bare and RPA Susceptiblity) $Fe_{1+y}Te$



J^{RKKY}



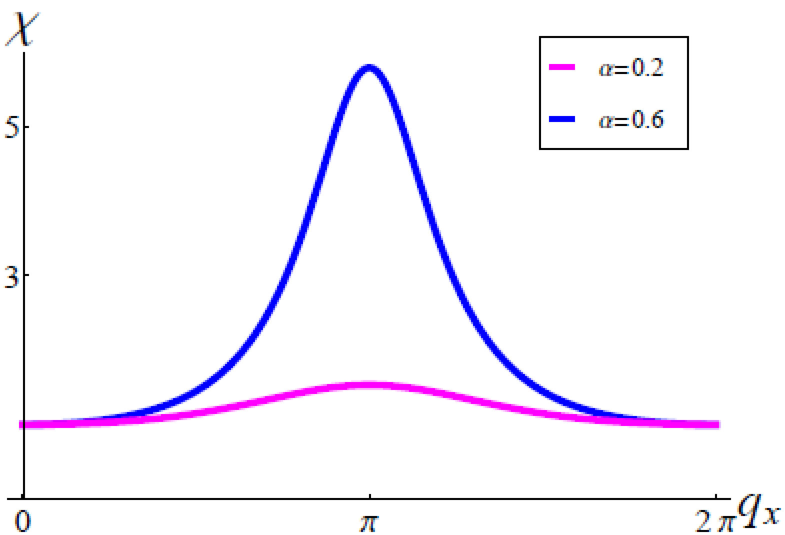
(π, 0) peak completely disappears

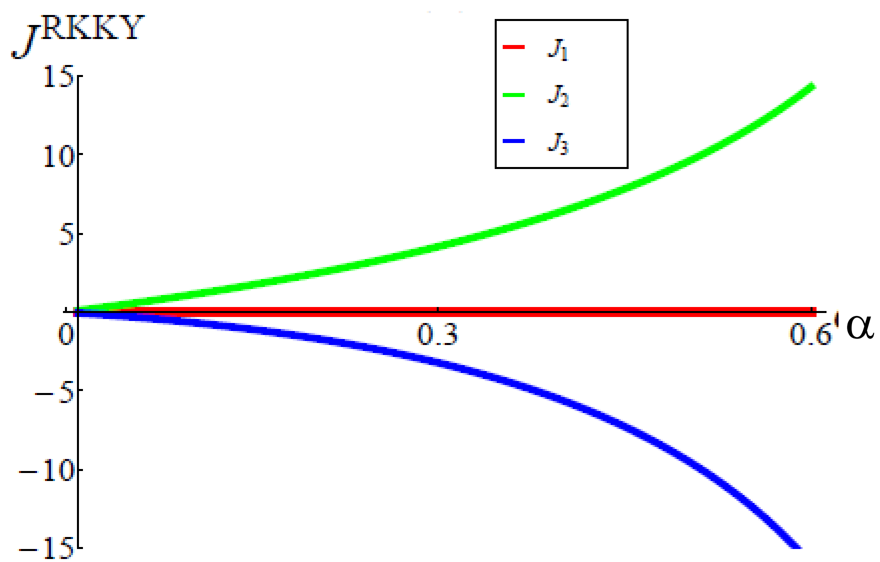
A toy model for χ for y<0.04

- Consider a phenomenological χ^{RPA} for y<0.04, where α is a parameter controlling the height of peak.

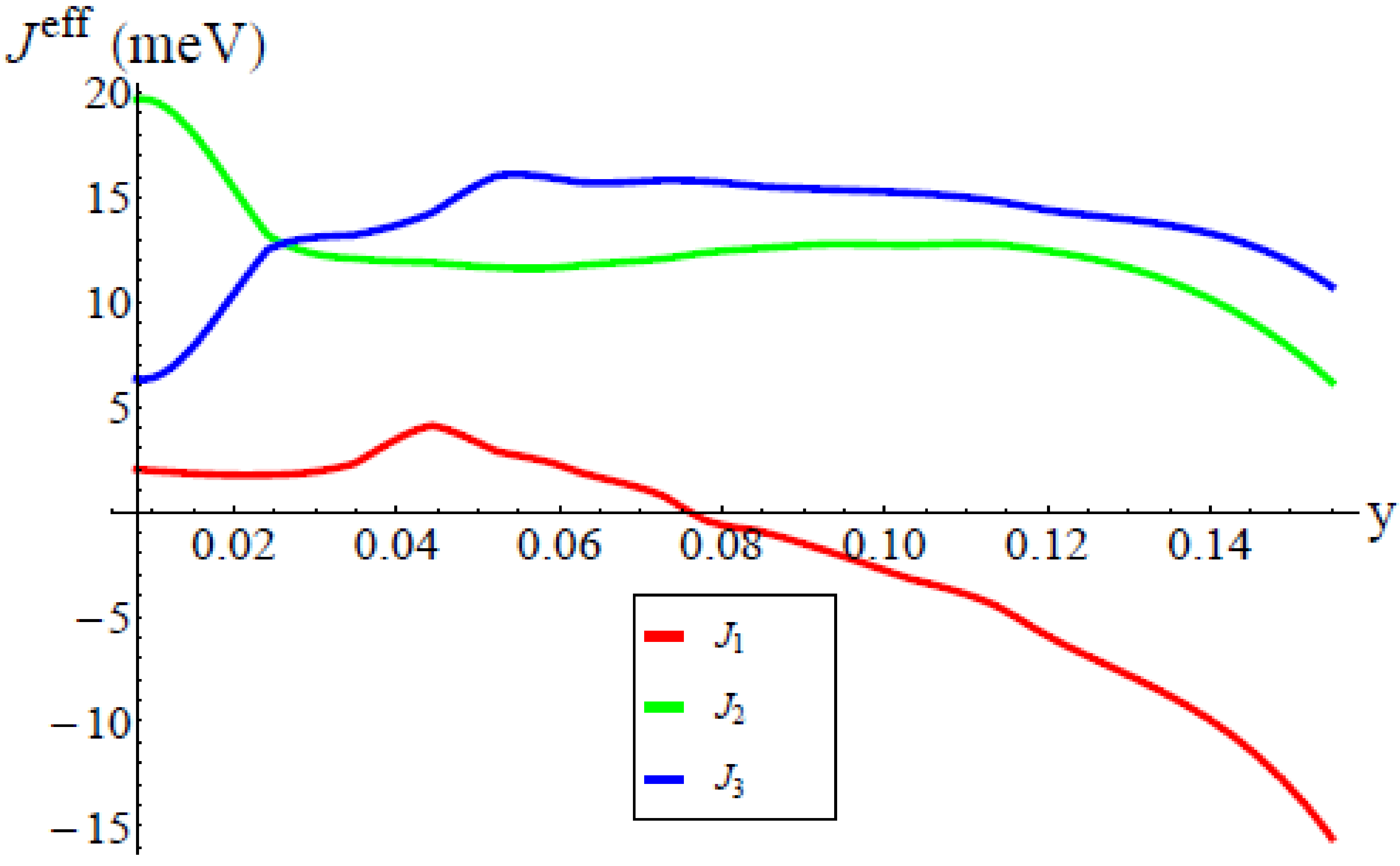
$$\chi^{-1}(\mathbf{q}) = \frac{1+\alpha\left[\cos q_x \cos q_y - \frac{1}{8}\left(\cos 2q_x + \cos 2q_y\right)\right]}{\chi_0\left(1+\frac{3}{4}\alpha\right)}$$

- With this model, we can calculate J^{RKKY} analytically.





$$J_{ij}^{eff} = J_{ij}^{Heis} + J_{ij}^{RKKY}$$

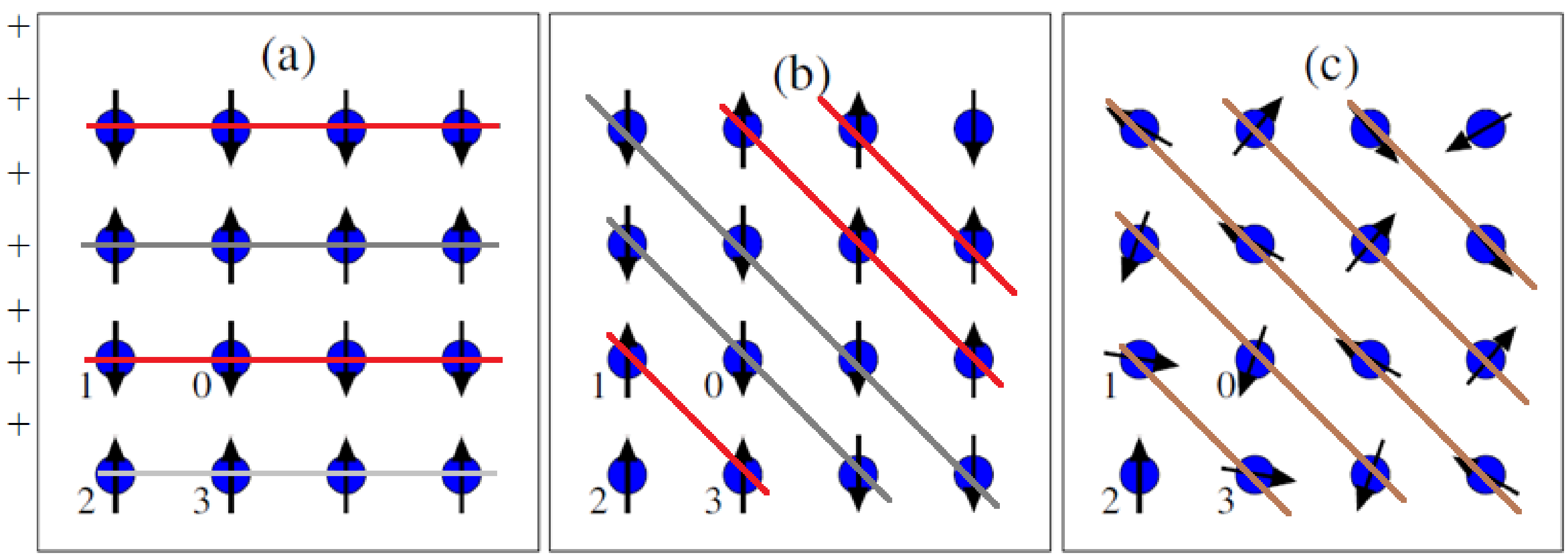


Ground State

$$H = \sum_{ij} J_{ij}^{eff} \mathbf{S_i} \cdot \mathbf{S_j} + \sum_{ijkl} K_{ijkl} (\mathbf{S_i} \cdot \mathbf{S_j})(\mathbf{S_k} \cdot \mathbf{S_l})$$

- We consider all possible 4-sublattice-single-q ground states. We find the ground state by the Hamiltonian minimizing over all variables
- Of all possible states, only three states appear for physical parameters in our phase diagram

$$E_{cl} = \frac{S^2}{4}\Big[J_1\Big(\cos\varphi_1 + \cos(\varphi_1 + 2q_x) + \cos(\varphi_3 - \varphi_2) + \cos(\varphi_3 - (\varphi_2 + 2q_x)) + \cos\varphi_3 + \cos(\varphi_3 + 2q_y)$$



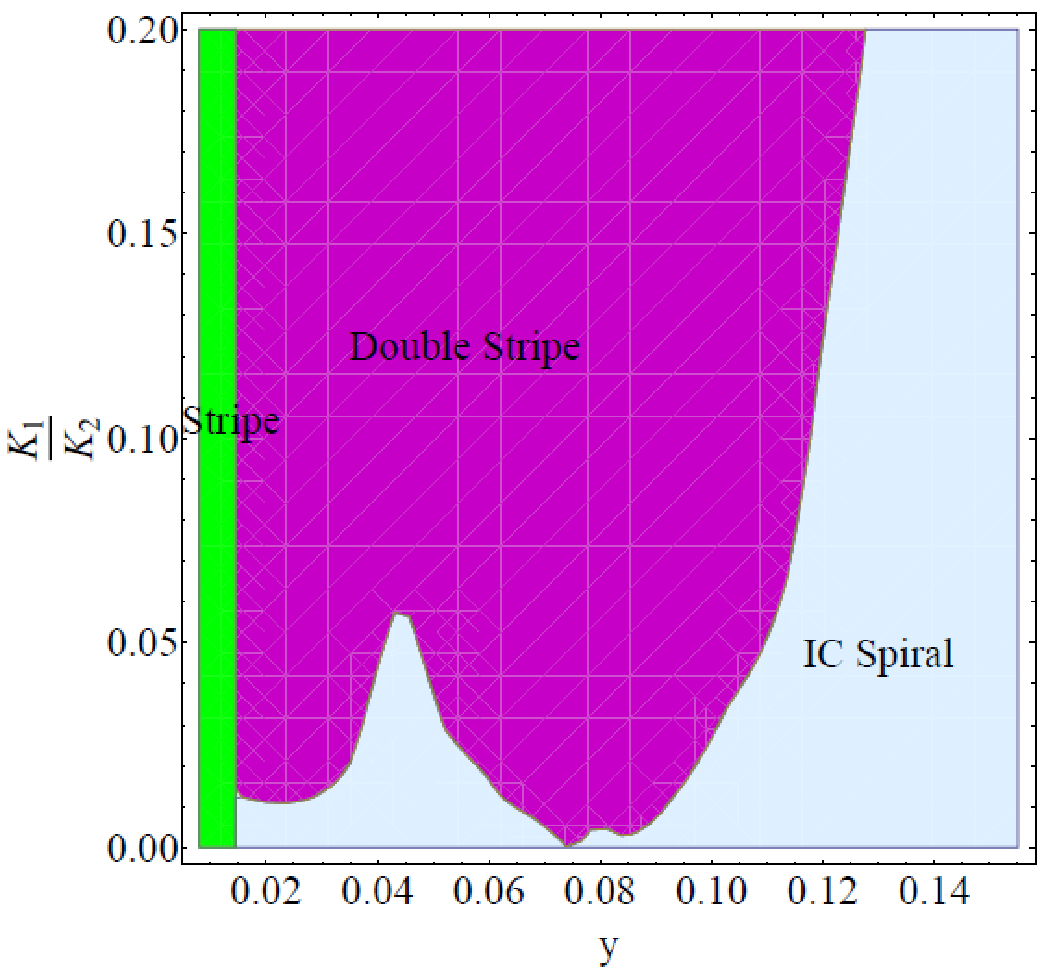
$2q_y)$

Phase Diagram

$$H = \sum_{ij} J_{ij}^{eff} \mathbf{S_i} \cdot \mathbf{S_j} + \sum_{ijkl} K_{ijkl} (\mathbf{S_i} \cdot \mathbf{S_j})(\mathbf{S_k} \cdot \mathbf{S_l})$$

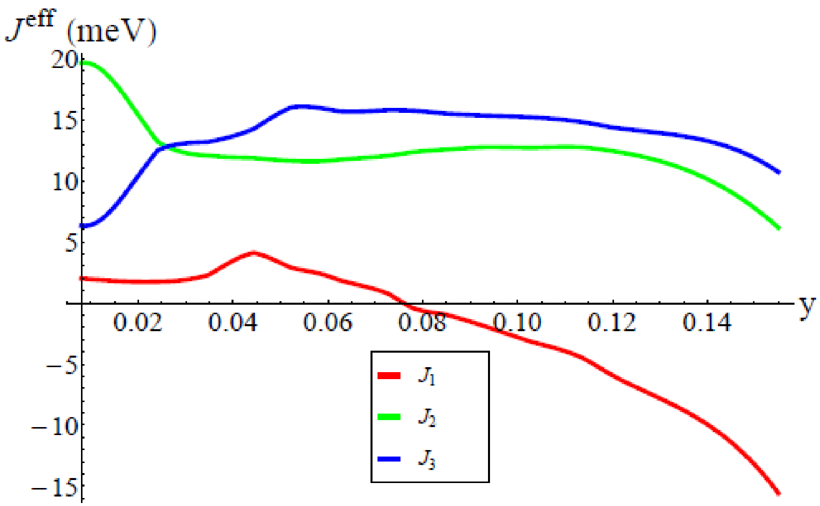
K_1 is the nearest neighbor biquadratic, K_2 is the value for both next-nearest neighbor biquadratic and ring exchange terms.

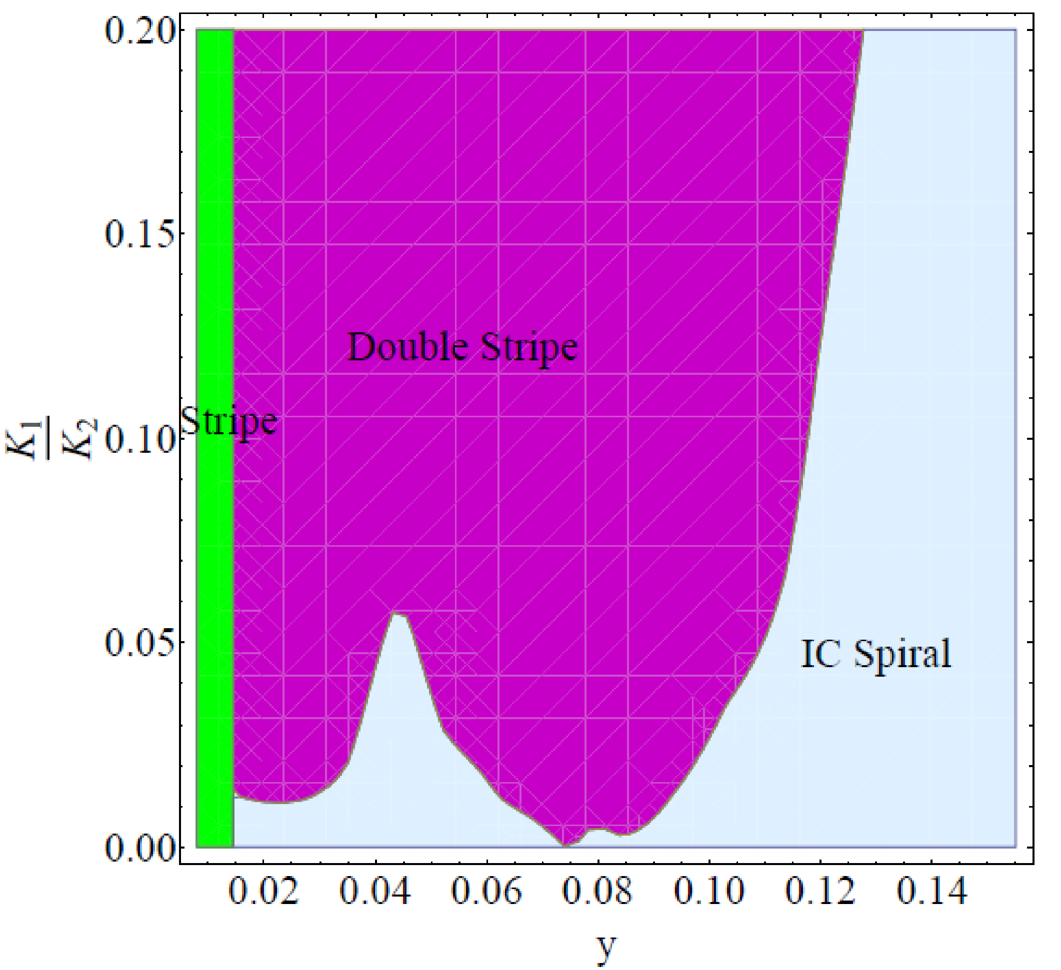
Here, fix K_2 = 3 meV



Phase Diagram

$$H = \sum_{ij} J_{ij}^{eff} \mathbf{S_i} \cdot \mathbf{S_j} + \sum_{ijkl} K_{ijkl} (\mathbf{S_i} \cdot \mathbf{S_j})(\mathbf{S_k} \cdot \mathbf{S_l})$$





Conclusion

- $Fe_{(1+y)}Te$ has features of both local magnetic moments and itinerant electrons.
- The **q**=(π /2, π/2) ground state (for y<0.11) can be obtained with a local model. It is not clear if it is possible to get it from the itinerant picture.
- Increasing y corresponds to electron doping.
- Integrating out itinerant electrons gives effective Heisenberg coupling, J_{ij}^{RKKY} (y).
- The hybrid model captures the evolution of magnetic order with increasing Fe excess.

Thank You!