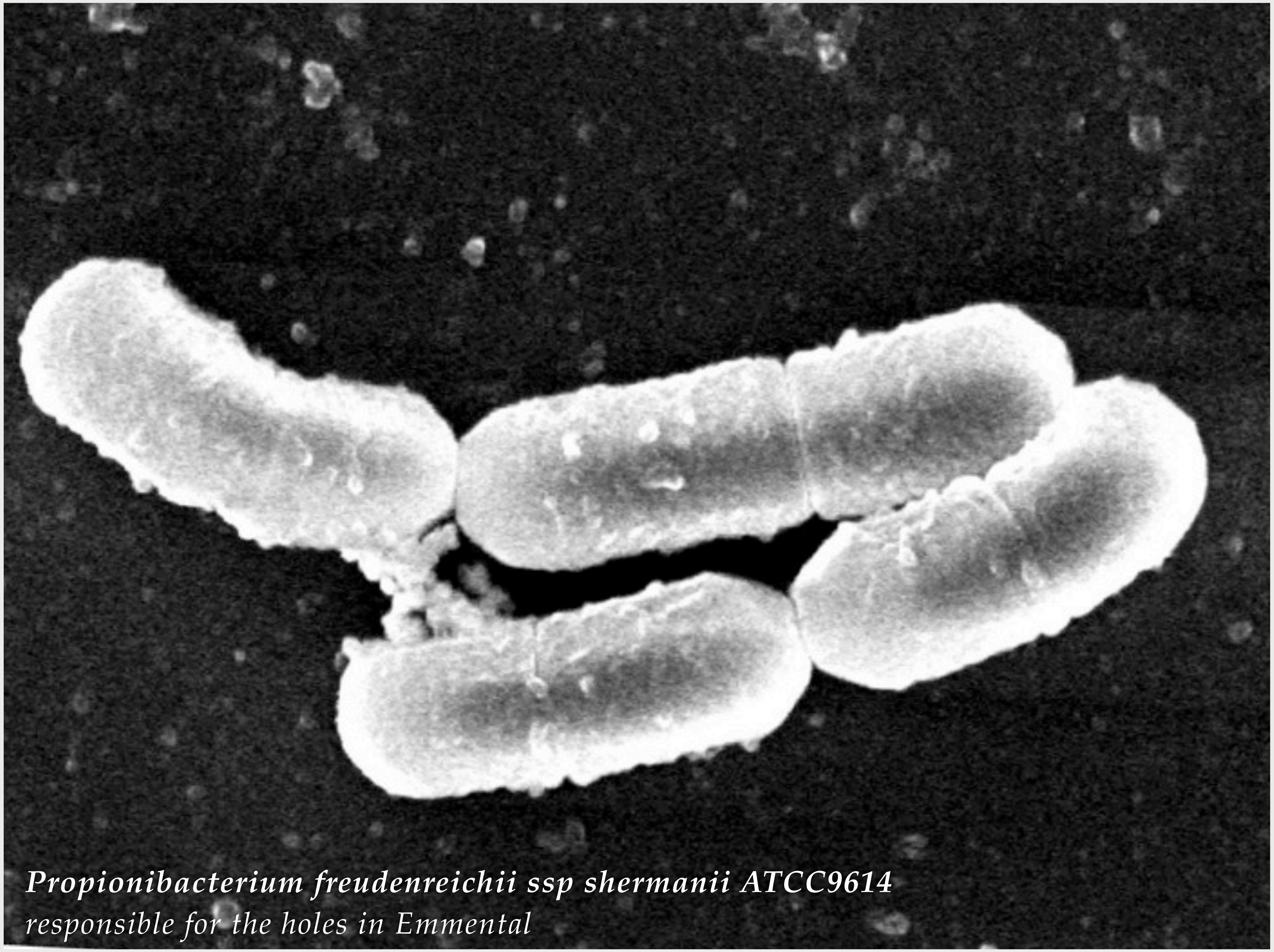


Propionibacterium freudenreichii ssp shermanii ATCC9614
responsible for the holes in Emmental

Impurity effects in Highly Frustrated Diamond-Lattice Antiferromagnets

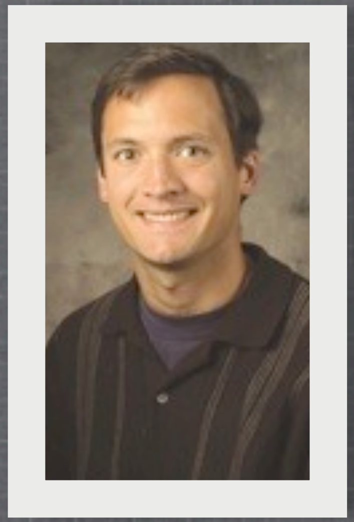
Lucile Savary

then: ÉCOLE NORMALE SUPÉRIEURE DE LYON

now: UCSB

Boston, February 28th, 2012

COLLABORATORS



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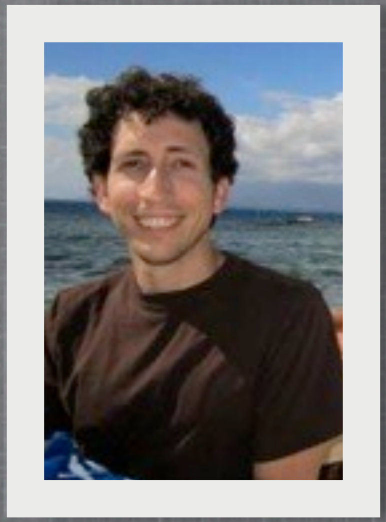

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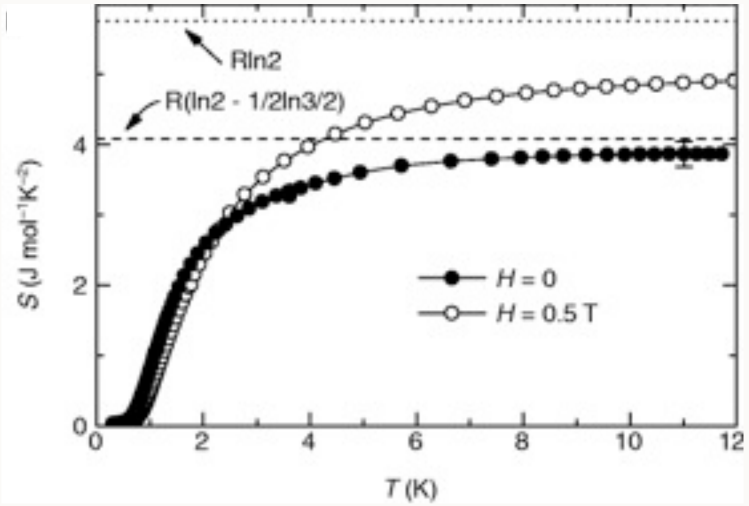
Physical Review B **84**, 064438 (2011)

UNFRUSTRATED CLASSICAL MAGNETS

- Weak/dilute disorder [Imry-Ma 1975, Harris 1974]
  - Random fields: strong effects, but not common
  - Random bonds: weak effects, except at phase transitions

FRUSTRATED MAGNETS

- Defined by degeneracy



entropy in spin ice $Dy_2Ti_2O_7$

- Effects:
  - enhanced thermal/ quantum fluctuations
  - sensitivity to weak perturbations

Ramirez *et al.* 1999

ORDER OR DISORDER?

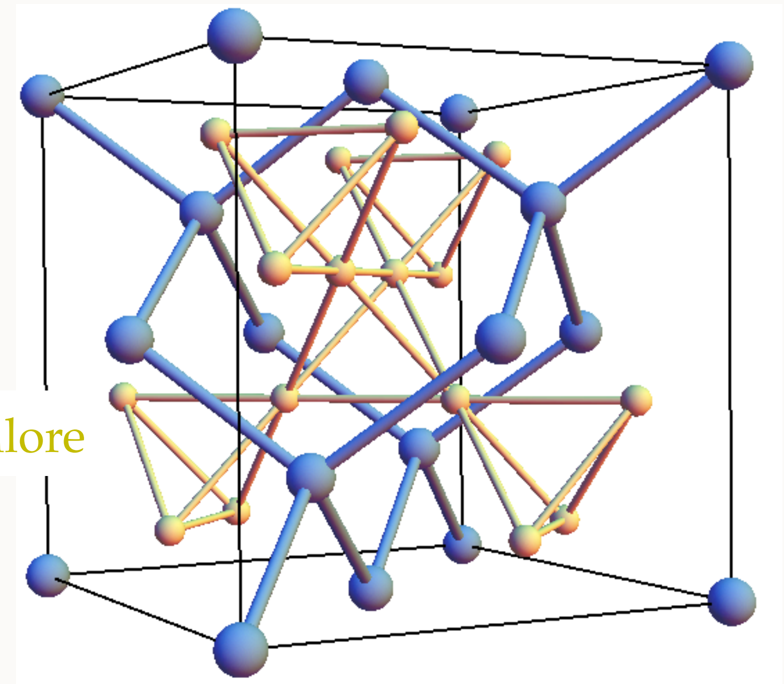
- Issue: Do impurities lead to order or disorder?
- Answer: It depends upon the nature of the frustration/degeneracy
  - Henley (1987): finite degeneracy => **order** (non-collinear)
  - Saunders+Chalker (2007): extensive degeneracy => **disorder** (spin glass)
  - This talk: sub-extensive degeneracy => **order**
    - How do we figure out which order?
    - When does this fail?

OUTLINE

- Spinel context
- Single impurities
- Local or global?
- Results
- Comparison with experiments

A-SITE SPINELS

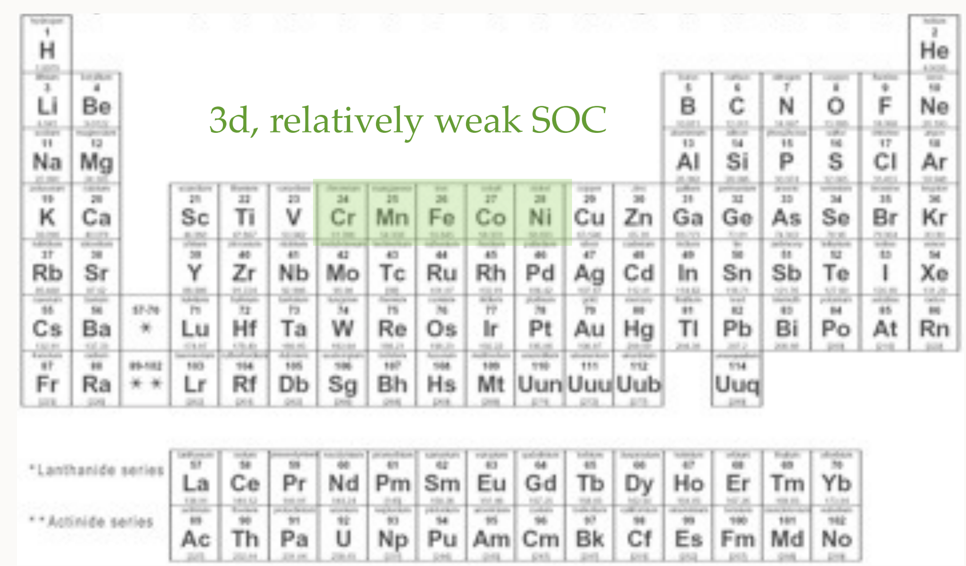
- spinels AB_2X_4
  magnetic

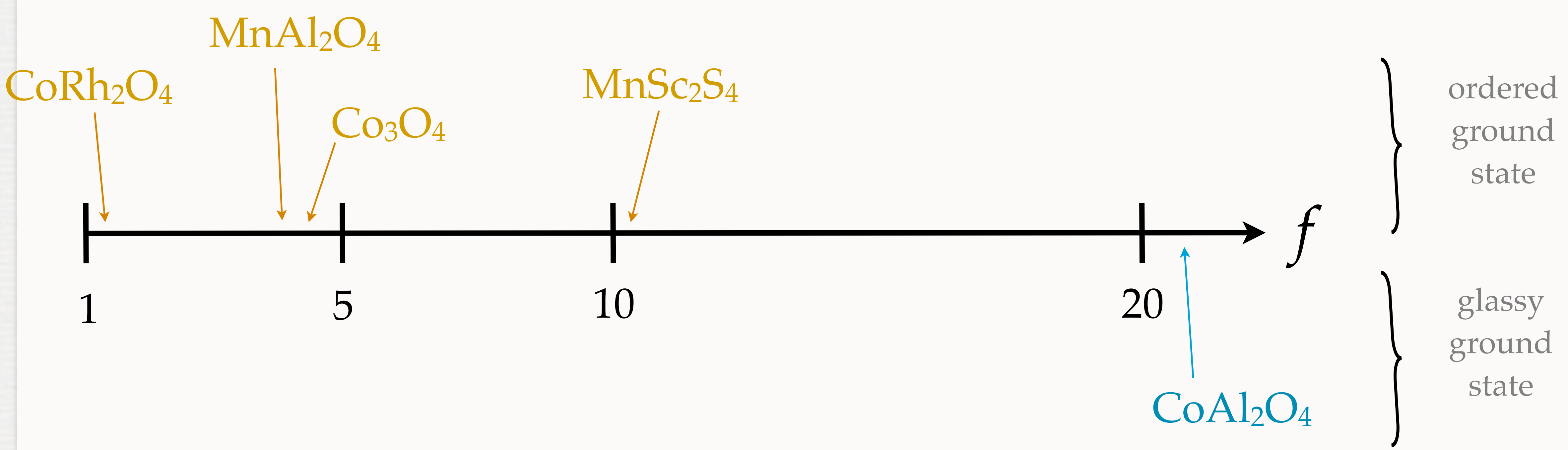


Roth 1964

A-SITE SPINELS

$$f = \frac{|\Theta_{CW}|}{T_N}$$

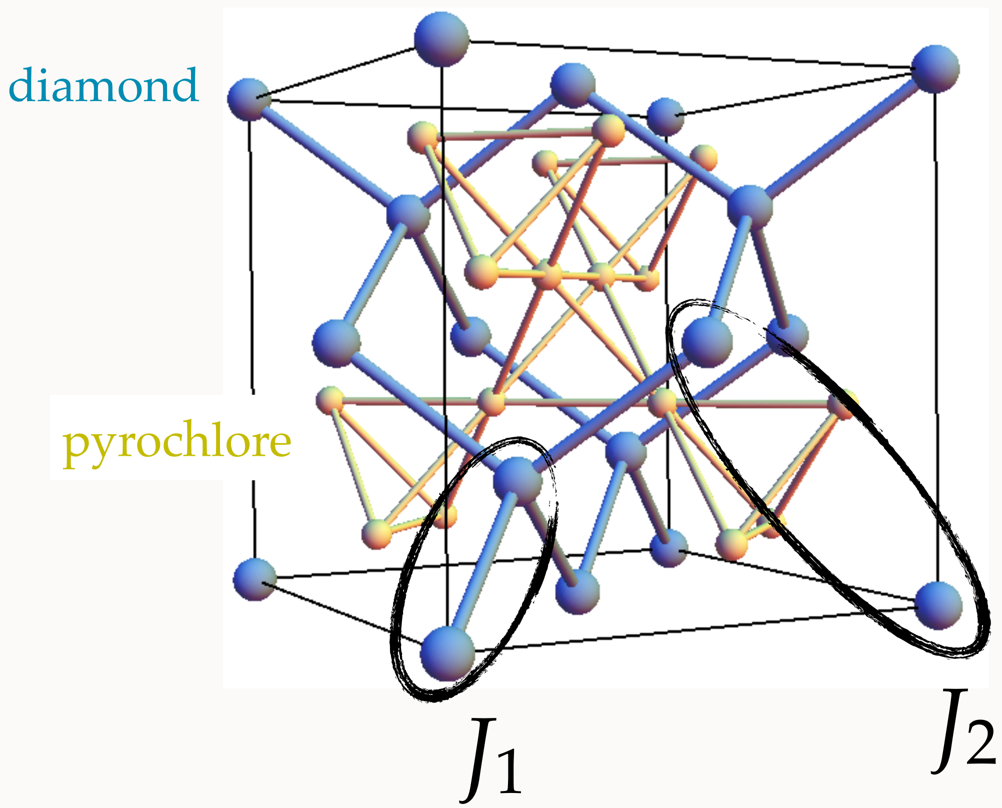




Fritsch *et al.* 1992, Tristan *et al.* 2005, Suzuki *et al.* 2006

A-SITE SPINELS

- spinels AB_2X_4
  magnetic

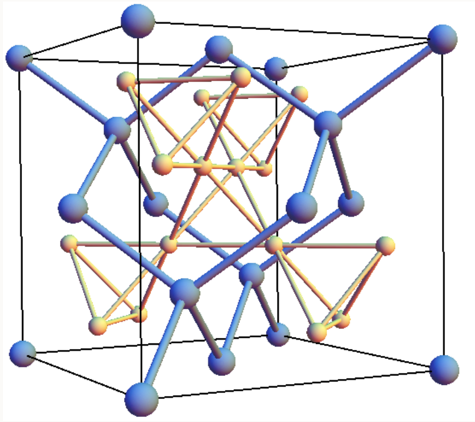


- diamond is bipartite
  - not frustrated
- second and third neighbor exchange not necessarily small
  - exchange paths A-X-B-X-B comparable

Roth 1964

FRUSTRATION: MINIMAL MODEL

$$H = J_1 \sum_{\langle i,j \rangle} \mathbf{S}_i \cdot \mathbf{S}_j + J_2 \sum_{\langle\langle i,j \rangle\rangle} \mathbf{S}_i \cdot \mathbf{S}_j \qquad J_2 > 0$$

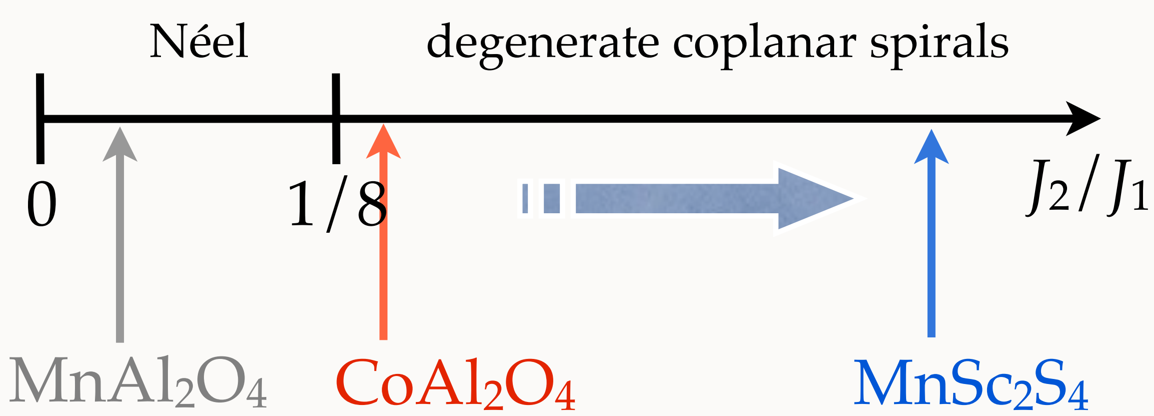


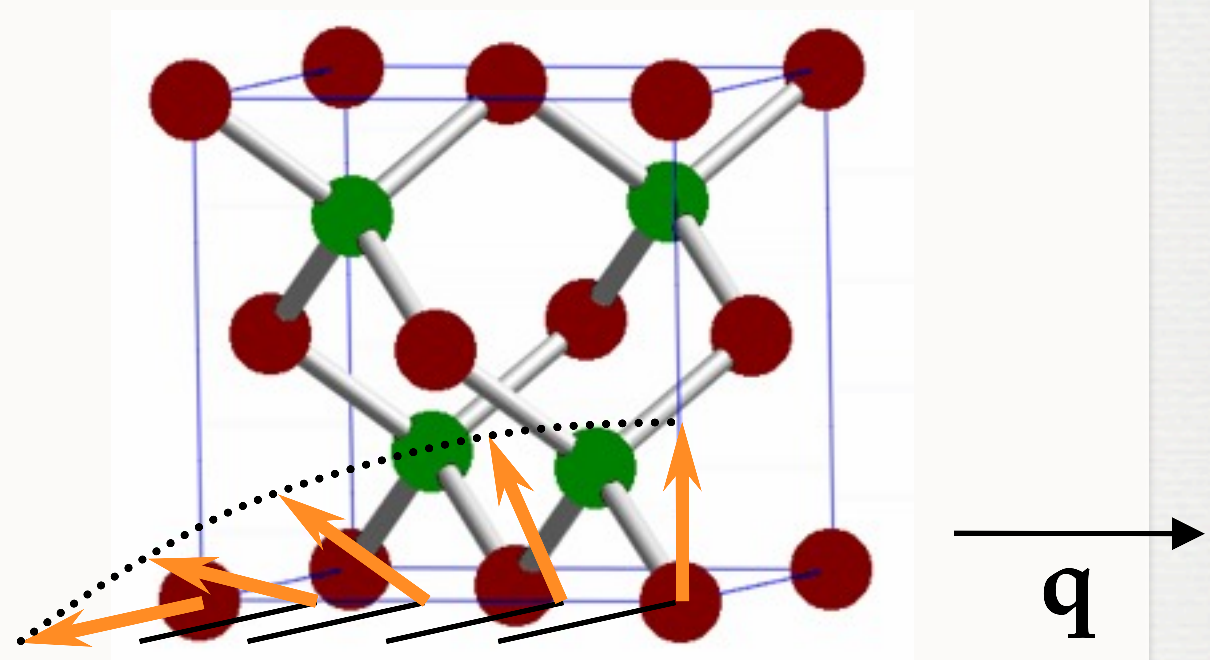
- $J_2 \longrightarrow 0$: diamond NN => Néel
- $J_1 \longrightarrow 0$: FCC NN => independent planes of spins

Bergman, Alicea, Gull, Trebst, Balents (2007)

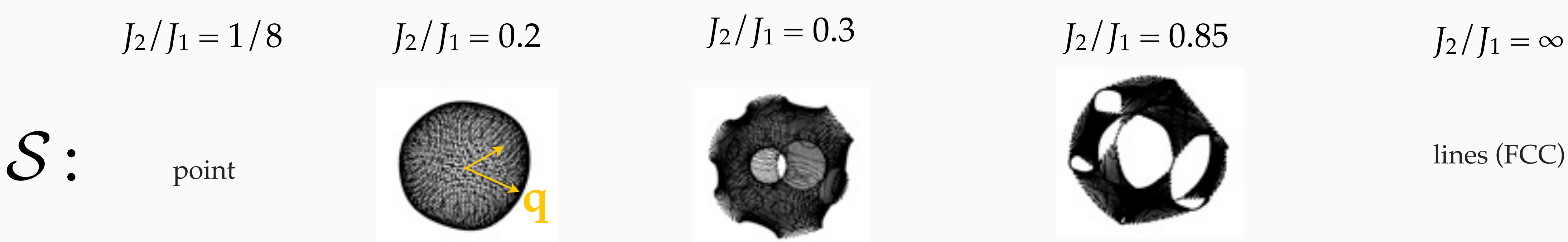
GROUND STATE EVOLUTION

- J_1-J_2 phase diagram





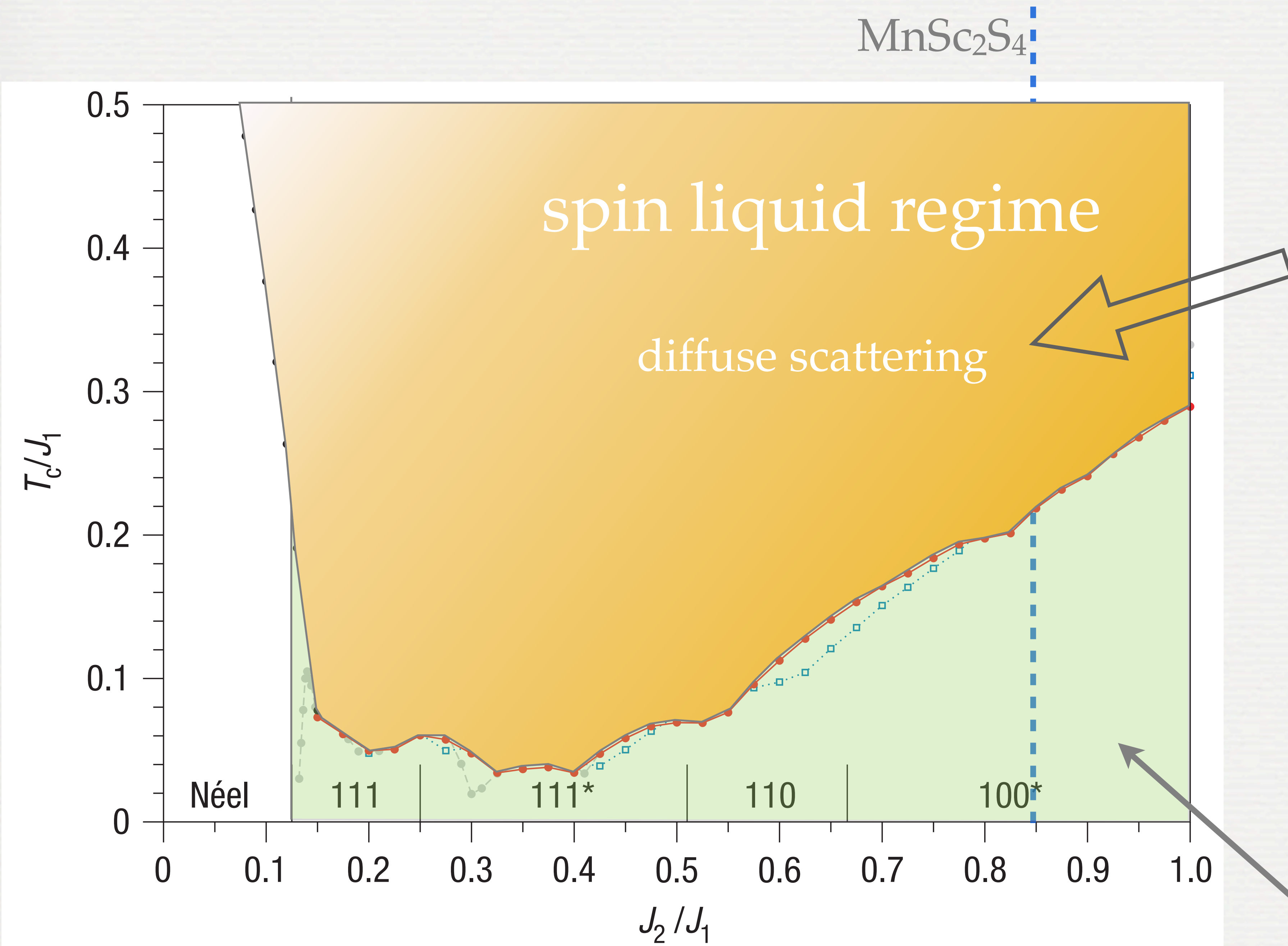
- example of degeneracy surfaces (reciprocal space)

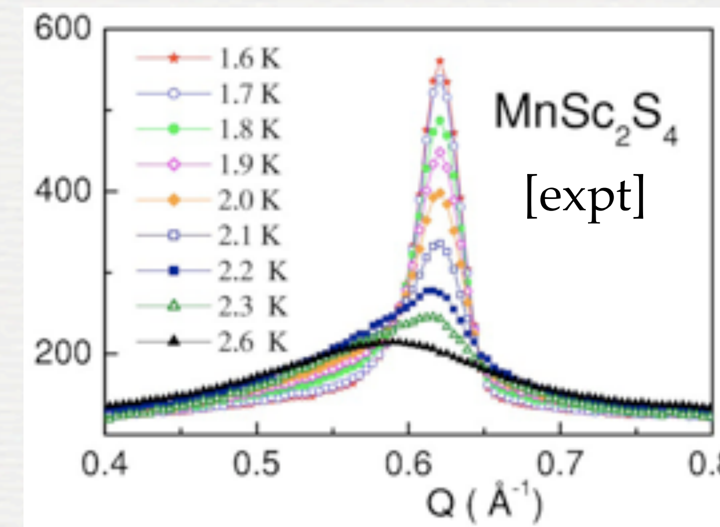


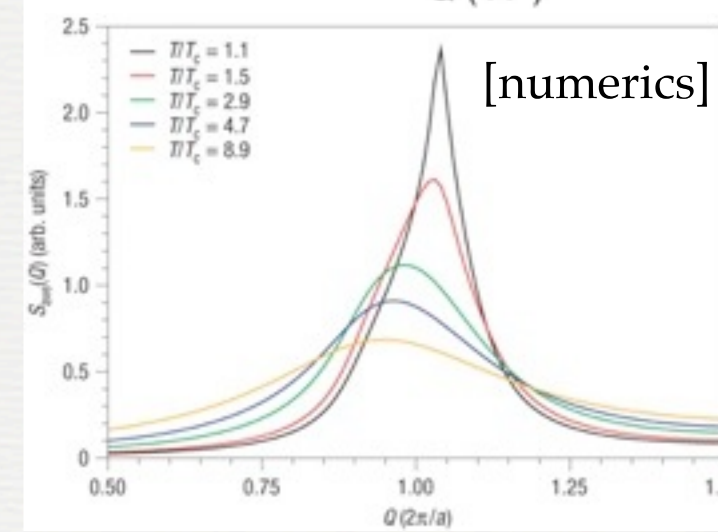
"accidental" 2D degeneracy: weak interactions will break it at $T = 0$

Fritsch *et al.* 1992, Tristan *et al.* 2005, Suzuki *et al.* 2006, Krimmel *et al.* 2006, Bergman *et al.* 2007

PHASE DIAGRAM (MONTE CARLO)







powder-averaged structure factor in a "spiral spin liquid"

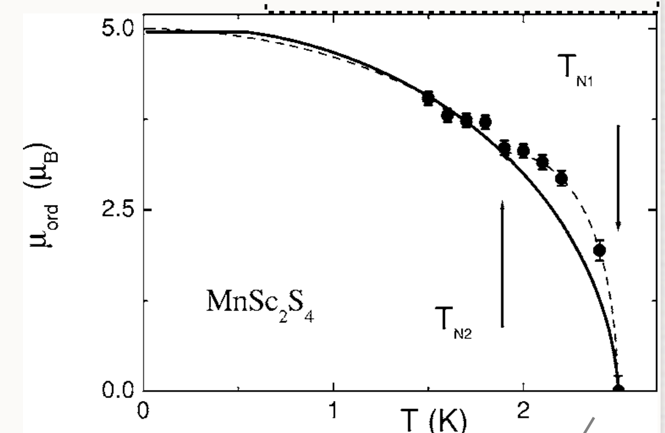
order by disorder

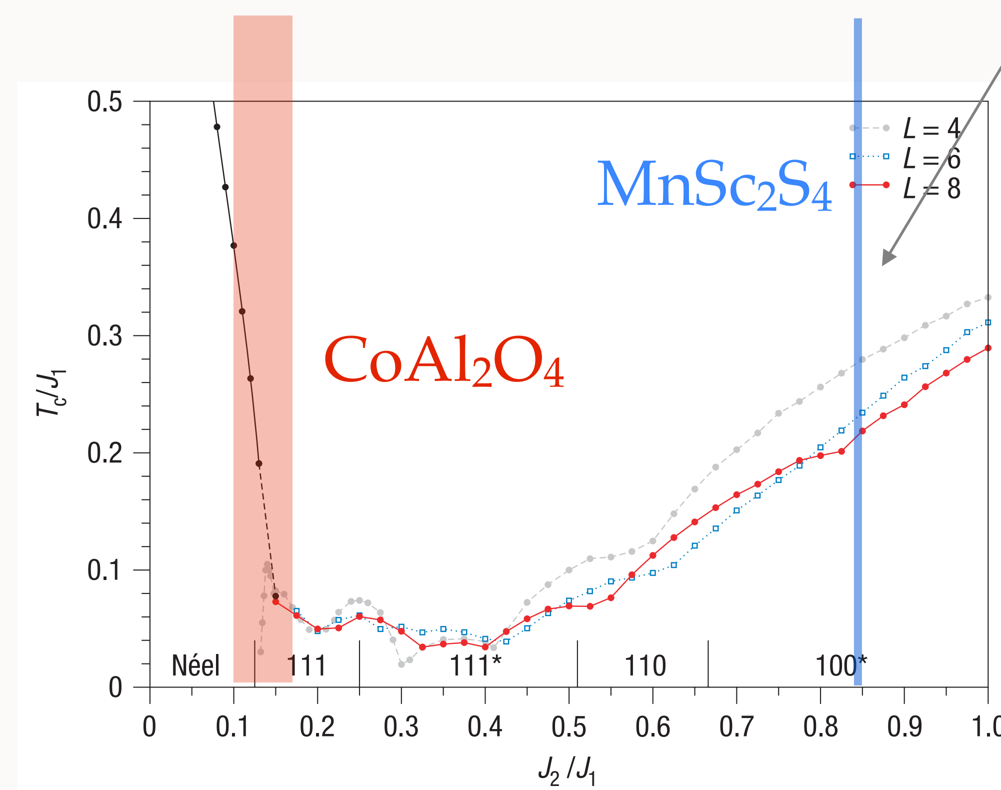
Krimmel *et al.* 2006, Bergman, Alicea, Gull, Trebst, Balents 2007

CONTRASTING BEHAVIORS: CAN IMPURITIES HELP?

- Why is $MnSc_2S_4$ ordered while $CoAl_2O_4$ is not? (chemists say quality is similar?)
- Is the contrasting behavior in these two "similar" materials consistent with a single theory for impurities?

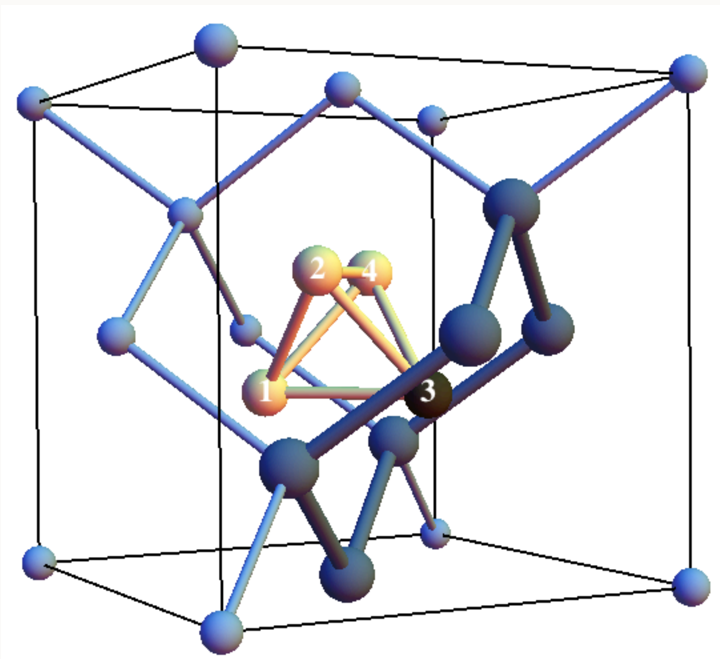
Krimmel *et al.* (2006)





Bergman, Alicea, Gull, Trebst, Balents (2007)

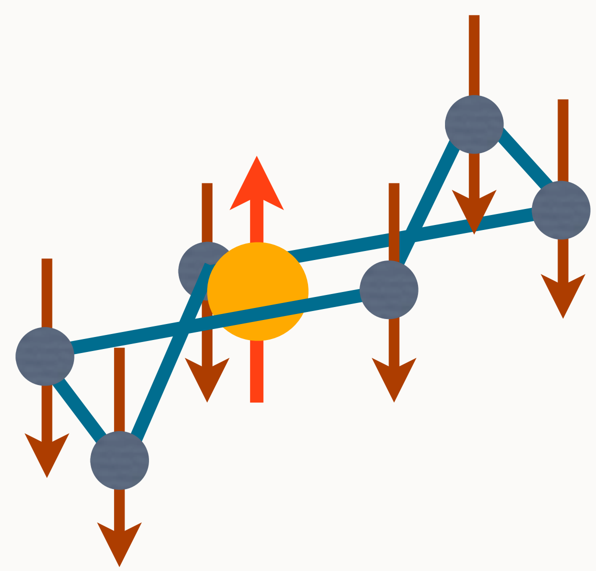
EXTRA B SPIN



impurity "type", $a = 1, .., 4$

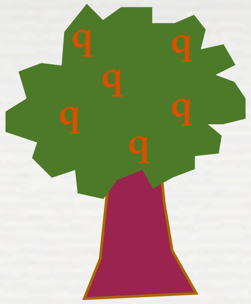
$$H^a_{imp} = J_{imp} \sum_{\langle a,i \rangle} \mathbf{S}_a \cdot \mathbf{S}_i$$

$$J_{imp} \gg J_1, J_2$$

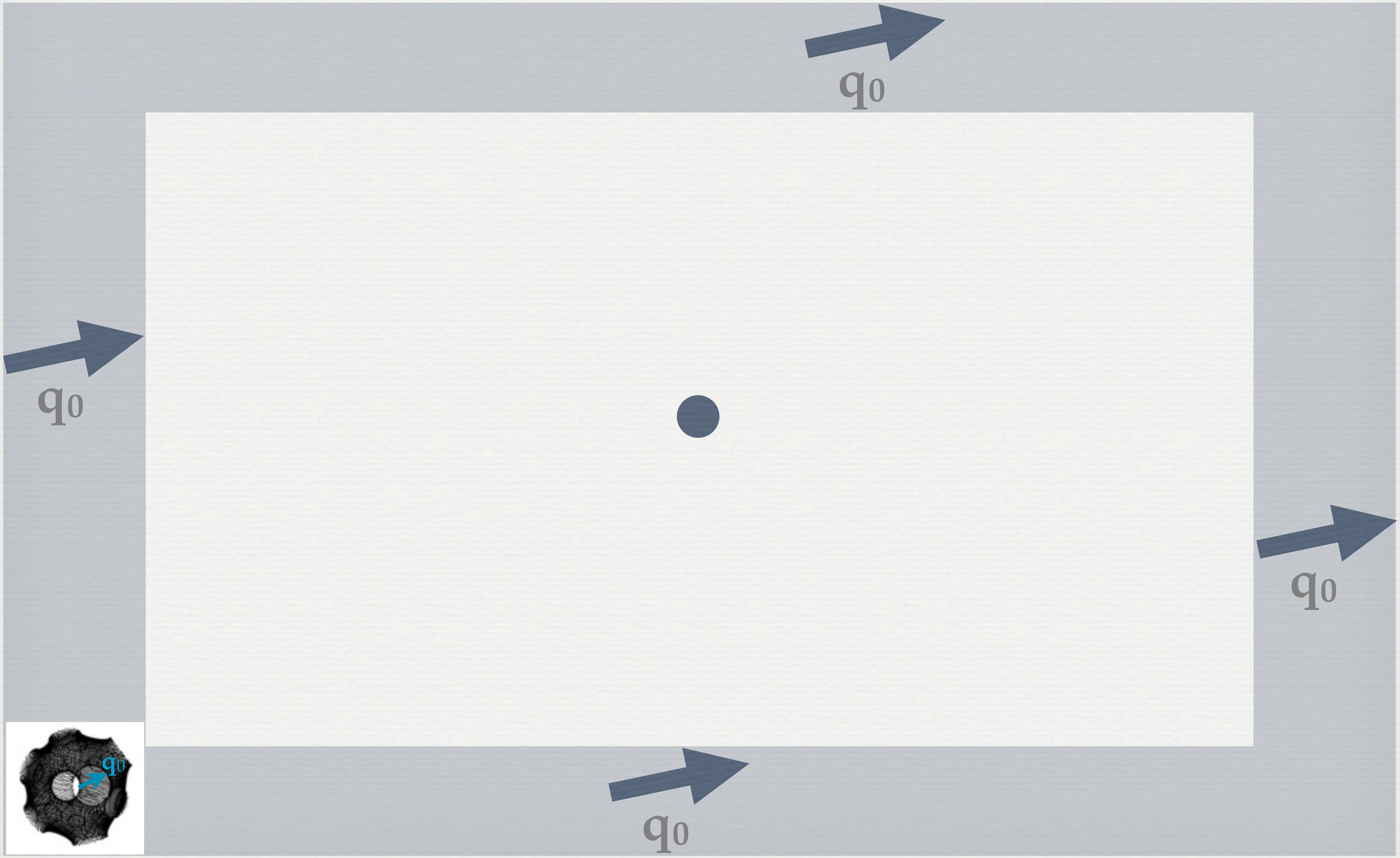


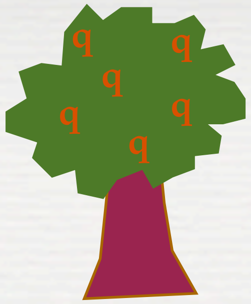
all NN spins aligned

- any kind of local impurity would do the job!
- expect surface degeneracy breaking



Q PICKING





Q PICKING

$\mathbf{q}_0$

$\mathbf{q}_0$

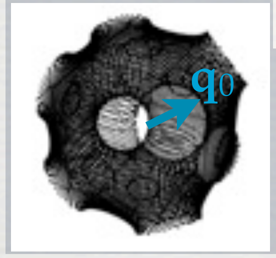
$$E_a(\mathbf{q}_0) = \text{energy}(\mathbf{q}_0; \text{with impurity}) - \underbrace{\text{energy}(\mathbf{q}_0; \text{without impurity})}_{}$$

test spiral wave vector

clean system's ground state energy
(energy of a ground state spiral)

$\mathbf{q}_0$

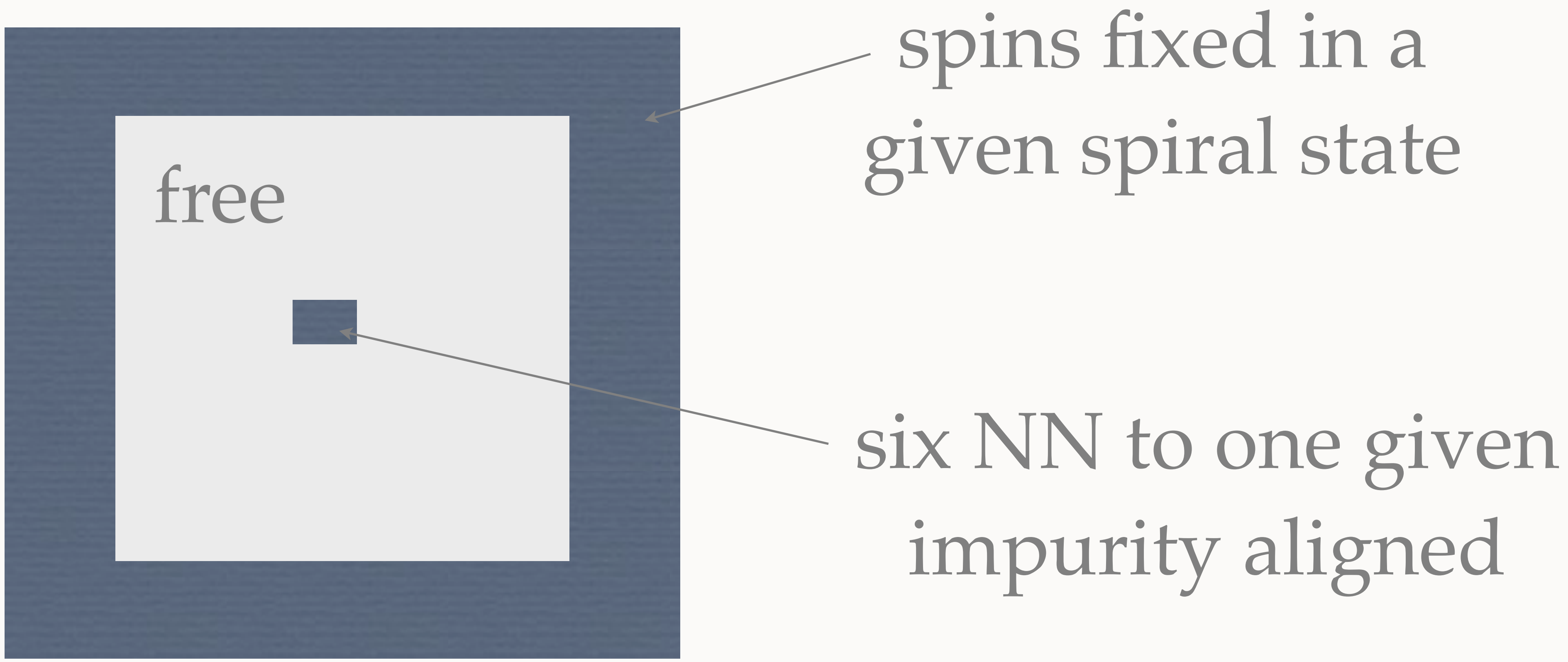
note: impurity energy cost can always be made $O(1)$



$\mathbf{q}_0$

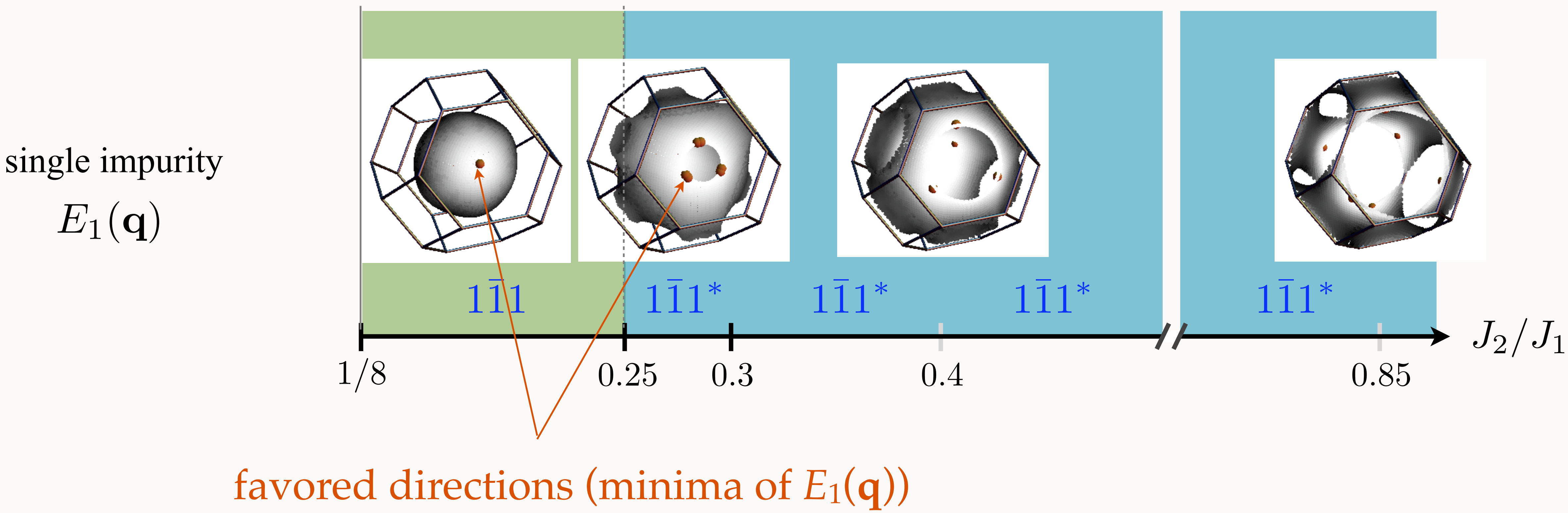
NUMERICS

- Classical Monte Carlo with



IMPURITY FAVORED DIRECTIONS

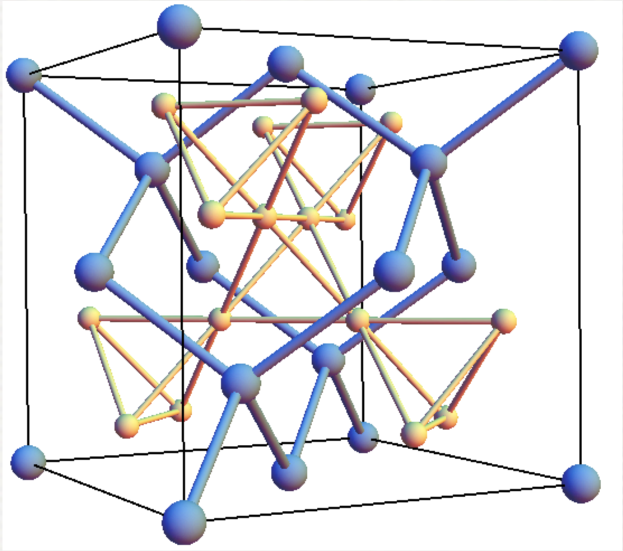
- *single* impurity "phase diagram"



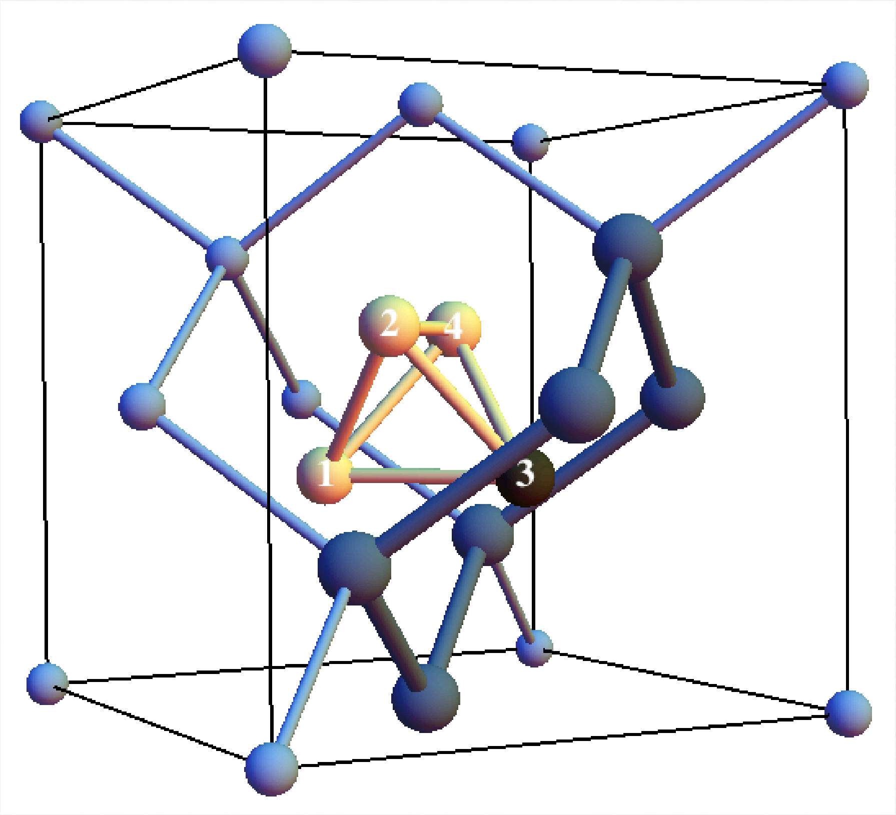
But what happens with
more than one impurity?

LOCAL OR GLOBAL?

AB_2X_4



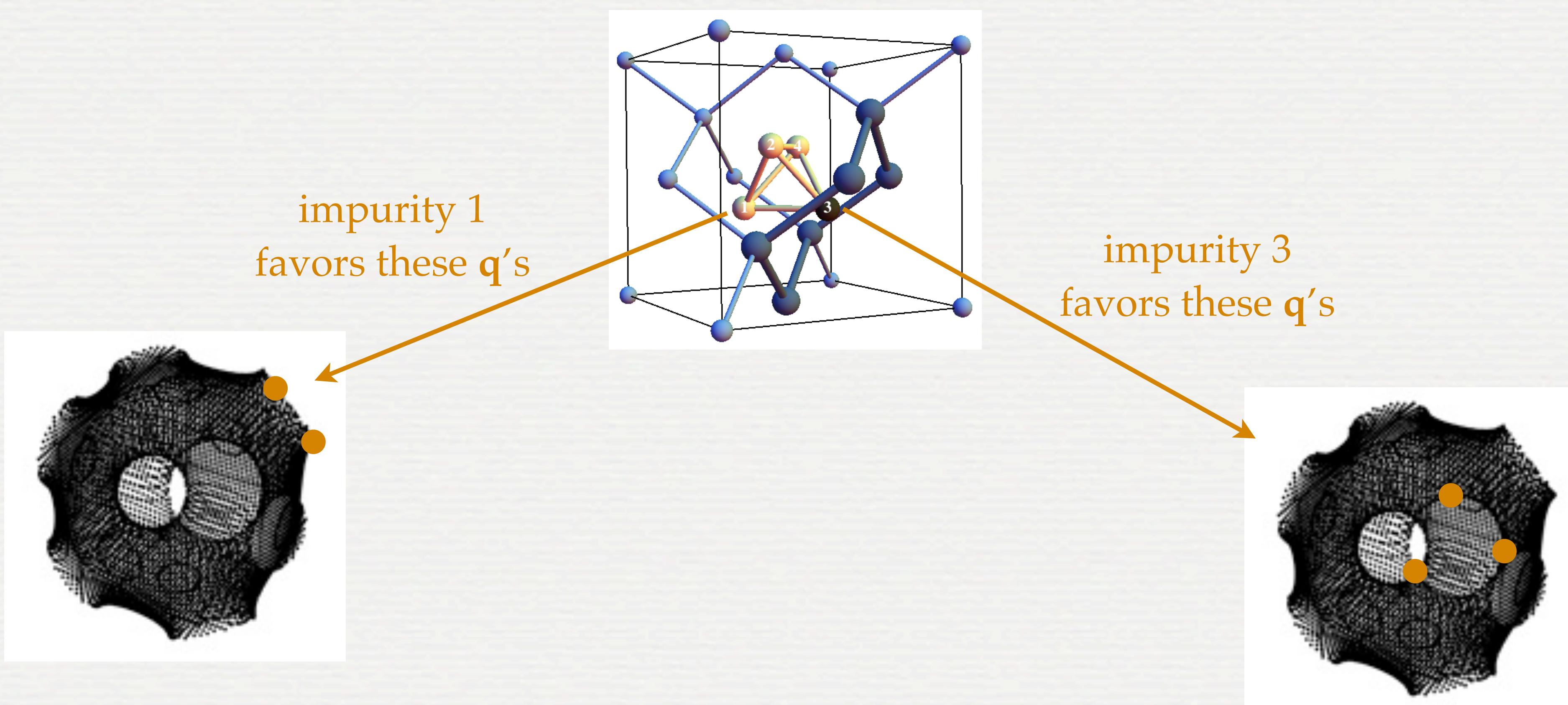
4 possibilities



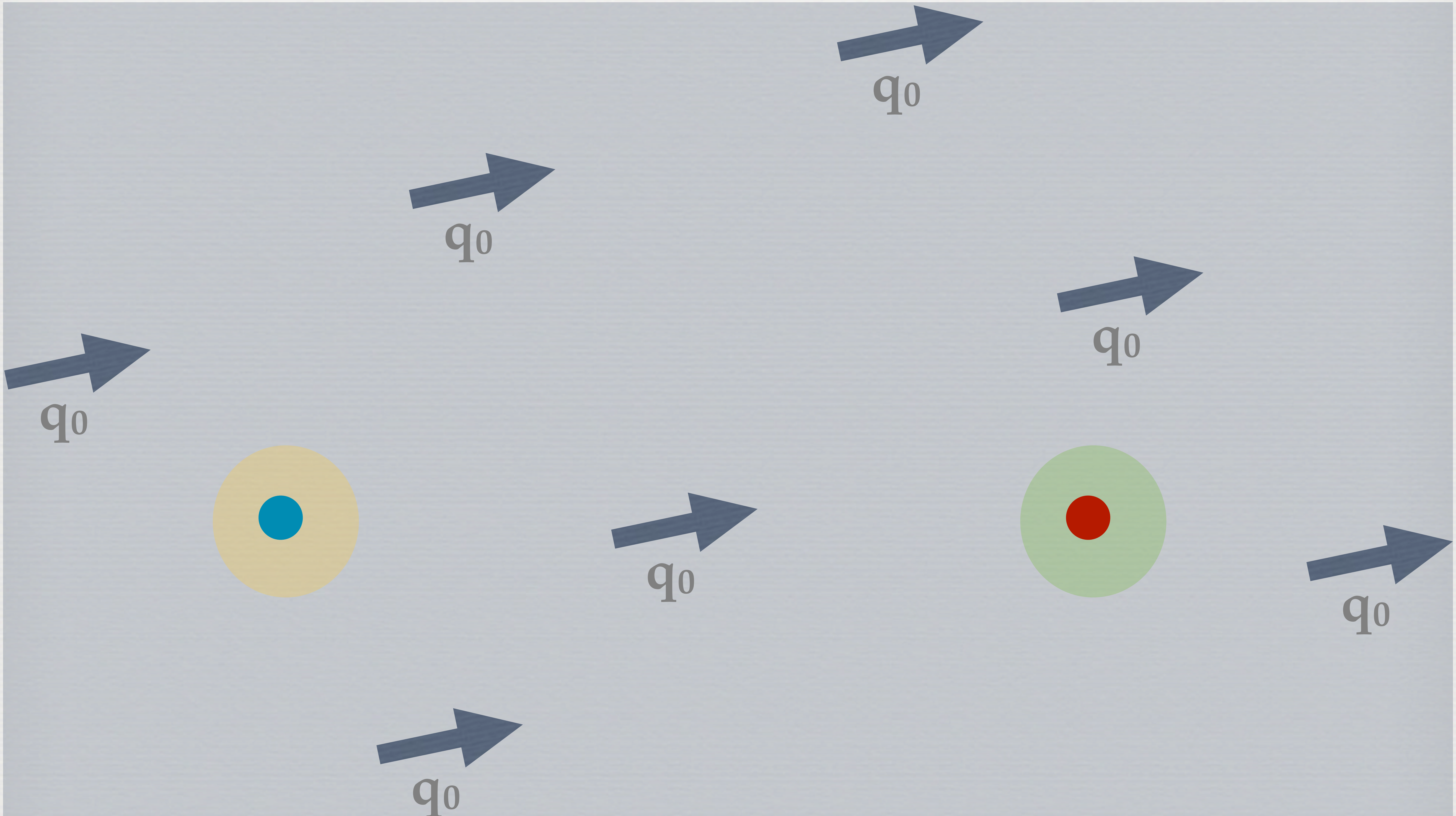
LOCAL OR GLOBAL?

impurities break the degeneracy
but favor *different* **q** vectors,
so what happens?

e.g.



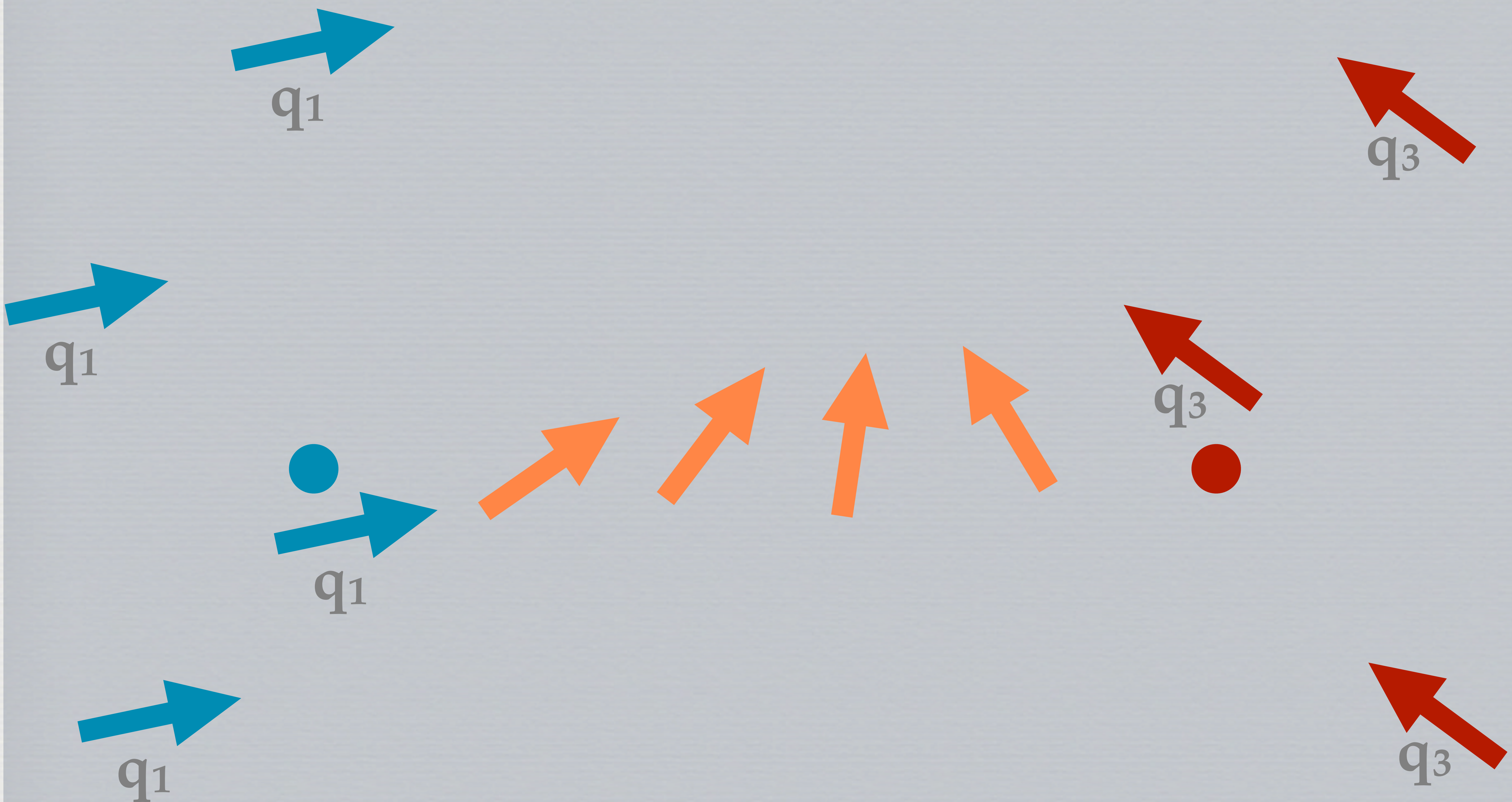
LOCAL OR GLOBAL?



q$_0$ is a compromise => larger energy cost near impurities

local

LOCAL OR GLOBAL?



q1 / **q**3 better close to impurities and small energy cost but over longer distances

global

REASON BY CONTRADICTION

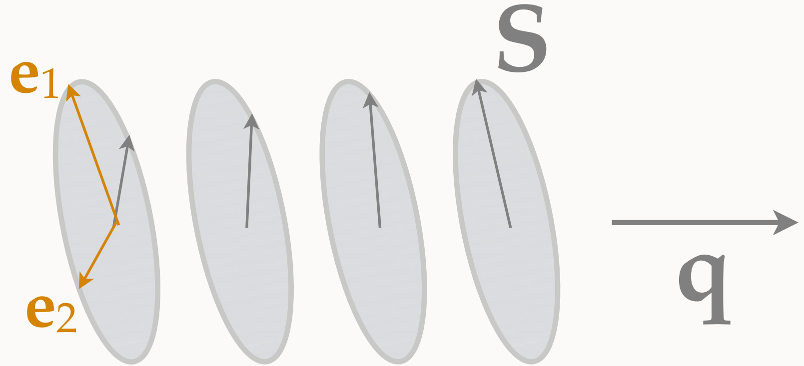
- calculate energy of smoothly deformed spirals
- show it is divergent
- deduce that deformations are local

"ORDER PARAMETER"

- spirals: $\mathbf{S}(\mathbf{r}) = \mathrm{Re}\left[\mathbf{d}(\mathbf{r})e^{i\mathbf{q}\cdot\mathbf{r}}\right]$

$$\mathbf{d} = \hat{\mathbf{e}}_1 + i\hat{\mathbf{e}}_2$$
$$\hat{\mathbf{e}}_1 \cdot \hat{\mathbf{e}}_2 = 0$$

- Landau-like expansion of energy density



  - given $(\mathbf{d}, \mathbf{q})$: spiral
  - redundancy, e.g.: $\mathbf{q} \to \mathbf{q} + \delta\mathbf{q}$
    $\mathbf{d} \to \mathbf{d}e^{-i\delta\mathbf{q}\cdot\mathbf{r} - i\delta\gamma}$
  - fix this "gauge": $\mathbf{S}(\mathbf{r}) = \mathrm{Re}\left[\mathbf{d}(\mathbf{r})e^{i\mathbf{q}_0\cdot\mathbf{r}}\right]$

all variations are encoded in $\mathbf{d}$

WEAKLY DEFORMED SPIRALS

$$\mathbf{S}(\mathbf{r}) = \mathrm{Re}\left[\mathbf{d}(\mathbf{r})e^{i\mathbf{q}_0\cdot\mathbf{r}}\right]$$

physical wavevector: $q^\mu = q_0^\mu + \frac{1}{2}\mathrm{Im}\left[\mathbf{d}^* \cdot \partial_\mu \mathbf{d}\right]$

$$\mathbf{d}(\mathbf{r}) = \mathbf{d}_0 + \delta\mathbf{d}(\mathbf{r})$$

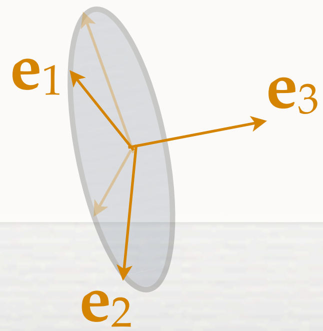
$$\hat{\mathbf{e}}_3 = -\frac{1}{2}\mathrm{Im}\left[\mathbf{d} \times \mathbf{d}^*\right]$$

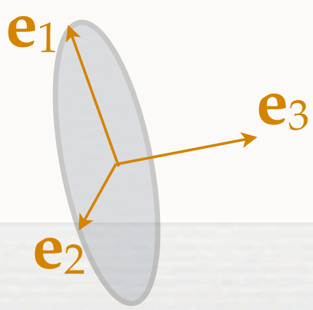
$$\delta\mathbf{d}(\mathbf{r}) = \underbrace{i\phi(\mathbf{r})}_{\text{rotates } \mathbf{d} \text{ within the same plane}}\mathbf{d}_0 + \underbrace{\psi(\mathbf{r})}_{\text{takes } \mathbf{S} \text{ to another plane}}\hat{\mathbf{e}}_3$$

$\phi \in \mathbb{R} \quad \psi \in \mathbb{C}$

rotates **d** within the same plane

takes **S** to another plane





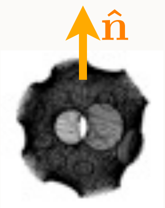
ENERGY DENSITY OF A WEAKLY DEFORMED SPIRAL

$$\mathbf{S}(\mathbf{r}) = \mathrm{Re}\left[\mathbf{d}(\mathbf{r})e^{i\mathbf{q}_0\cdot\mathbf{r}}\right] \qquad \delta\mathbf{d}(\mathbf{r}) = i\phi(\mathbf{r})\mathbf{d}_0 + \psi(\mathbf{r})\hat{\mathbf{e}}_3$$

- constraints:
  - undeformed spirals: zero energy
  - variation must cost zero energy when **q** stays on spiral surface
  - stability

$\hat{\mathbf{n}}$: unit vector perpendicular to the spiral surface

$$\nabla_\perp = \hat{\mathbf{n}}\cdot\boldsymbol{\nabla} \qquad \boldsymbol{\nabla}_\parallel = \boldsymbol{\nabla} - \hat{\mathbf{n}}\nabla_\perp$$



consequence of *curved* degeneracy surface for **q**

$$\mathcal{E} = \frac{c}{2}(\nabla_\perp\phi)^2 + c'\nabla_\perp\phi\nabla_\parallel^2\phi + \frac{c''}{2}(\nabla_\parallel^2\phi)^2 + d\nabla_\perp\psi^*\nabla_\perp\psi + d'\boldsymbol{\nabla}_\parallel\psi^*\cdot\boldsymbol{\nabla}_\parallel\psi$$

"stiffness" $\varkappa$

SPINEL V/S PYROCHLORE

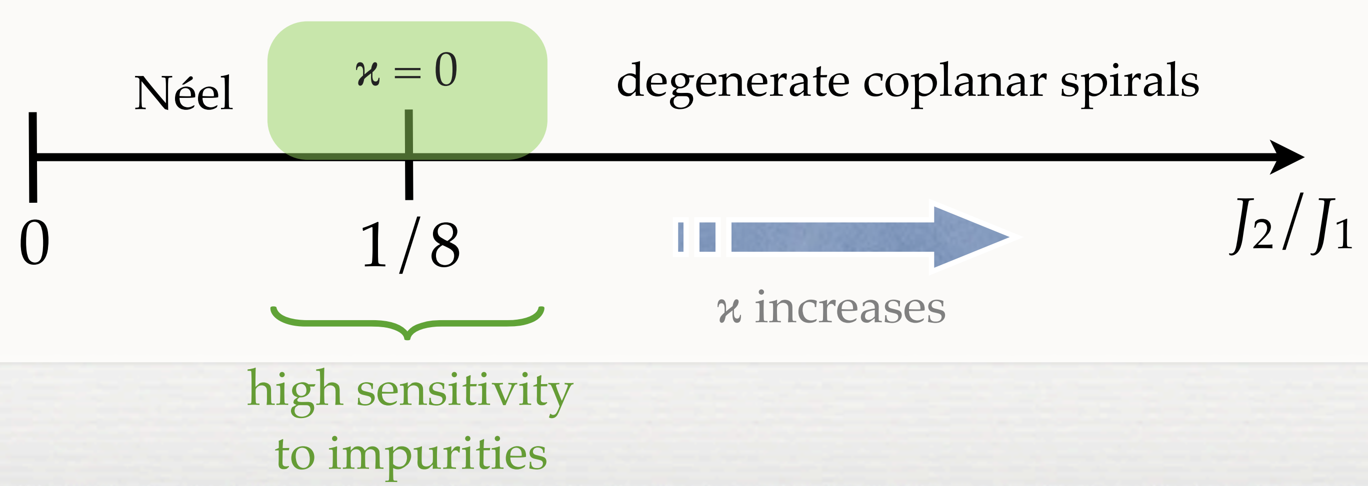
stiffness: measures the energy cost of an *infinitesimal* change of the spin state, deformed in a smooth fashion.

- Recall local *real space* degeneracy in pyrochlore

$$H \sim J \left(\sum_{\mu=0}^{3} \mathbf{S}^{\mu} \right)^2$$

local degeneracy => *no* stiffness

- Here *no real space* picture. Stiffness of **q** in reciprocal space. Stiffness varies along phase diagram.



SCALING

$$\mathcal{E} = \frac{c}{2}(\nabla_\perp \phi)^2 + c' \nabla_\perp \phi \nabla_\parallel^2 \phi + \frac{c''}{2}(\nabla_\parallel^2 \phi)^2 + d \nabla_\perp \psi^* \nabla_\perp \psi + d' \boldsymbol{\nabla}_\parallel \psi^* \cdot \boldsymbol{\nabla}_\parallel \psi$$

energy density of deformation

$$\sim \frac{(\delta\phi)^2}{L_\perp^2} \qquad \sim \frac{(\delta\phi)^2}{L_\perp L_\parallel^2} \qquad \sim \frac{(\delta\phi)^2}{L_\parallel^4}$$

isotropic

$$\sim \frac{|\delta\psi|^2}{L^2}$$

=> relaxation length anisotropy: $L_\perp \sim L_\parallel^2$

energy density scaling of deformation

$$\delta \mathbf{q} = \mathbf{q} - \mathbf{q}_0 = \boldsymbol{\nabla}\phi \quad => \quad \delta\phi^{\parallel} \sim L_\parallel \delta q_\parallel$$

integrate over deformation volume

$\Leftrightarrow \times L_\parallel^2 L_\perp$

$$E_{\text{deform}}(L) \sim (L_\parallel \delta q)^2 + |\delta\psi|^2 L$$

large scale (~ L) deformations of **q** : prohibitively costly (>> $O(1)$)

QED

consequence: impurities act independently of one another

NOTE: PHASE FLUCTUATION SUBTLETIES

$$\mathcal{E} = \underbrace{\frac{c}{2}(\nabla_\perp \phi)^2 + c' \nabla_\perp \phi \nabla_\parallel^2 \phi + \frac{c''}{2}(\nabla_\parallel^2 \phi)^2}_{\sim \frac{(\delta\phi)^2}{L_\parallel^4}} + d \nabla_\perp \psi^* \nabla_\perp \psi + d' \boldsymbol{\nabla}_\parallel \psi^* \cdot \boldsymbol{\nabla}_\parallel \psi$$

$$\delta\phi \sim \underbrace{(\delta q) L}_{\text{prohibited}} + \delta\phi_{\text{non}\,\delta q}$$

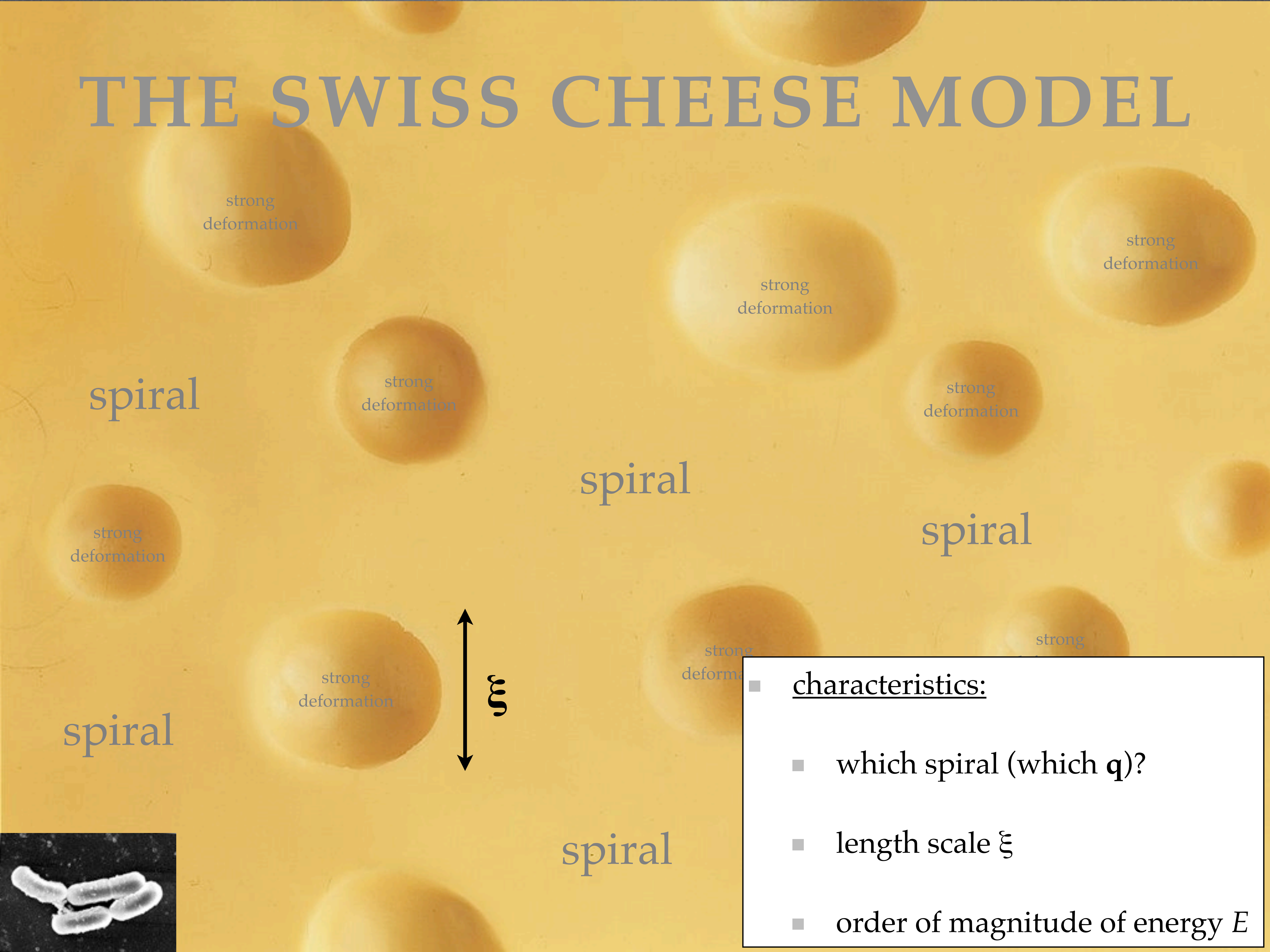
→ large scale fluctuations of ϕ are a priori allowed

$$C_\phi(\mathbf{r}) = \langle (\phi(\mathbf{r}') - \phi(\mathbf{r} - \mathbf{r}'))^2 \rangle \sim A|\mathbf{r}|^\alpha \qquad \text{for } |\mathbf{r}| \to \infty$$

$$\Rightarrow \quad C_{\mathbf{q}}(\mathbf{r}) = \langle (\mathbf{q}(\mathbf{r}') - \mathbf{q}(\mathbf{r} - \mathbf{r}'))^2 \rangle \sim \tilde{A}|\mathbf{r}|^{\alpha - 2}$$

expect $0 < \alpha < 2$

THE SWISS CHEESE MODEL

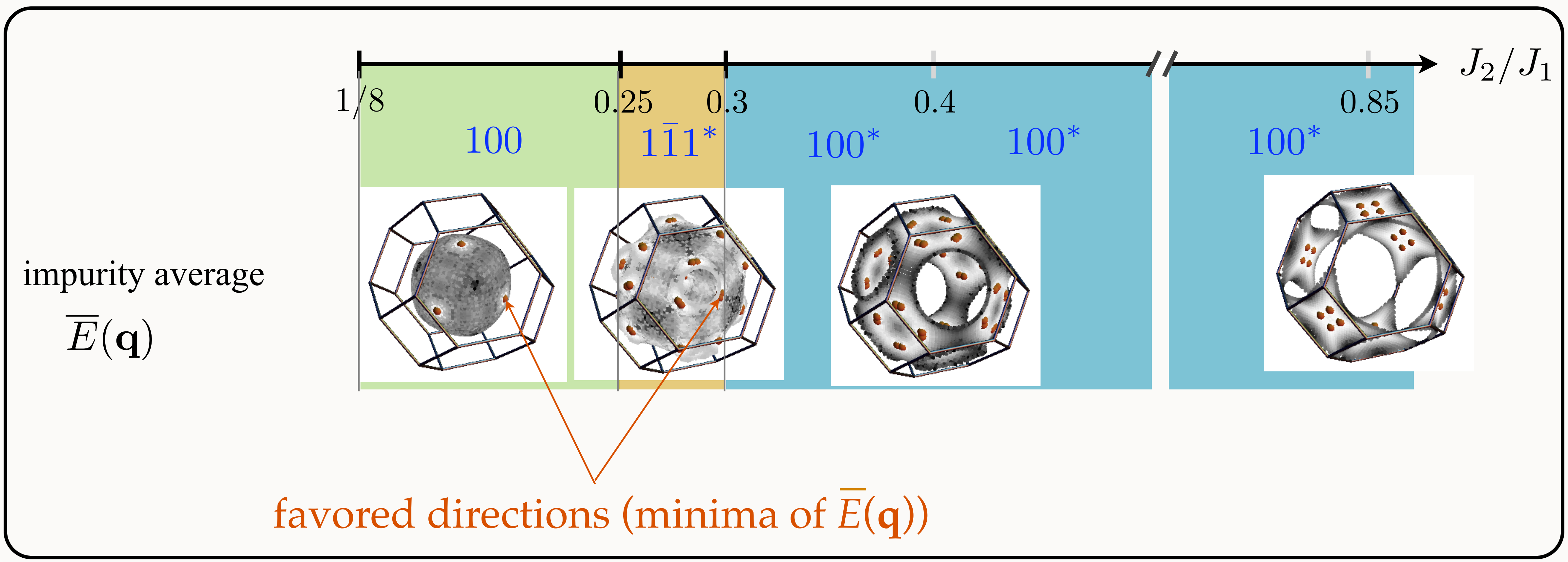


- <u>characteristics:</u>
  - which spiral (which **q**)?
  - length scale ξ
  - order of magnitude of energy *E*

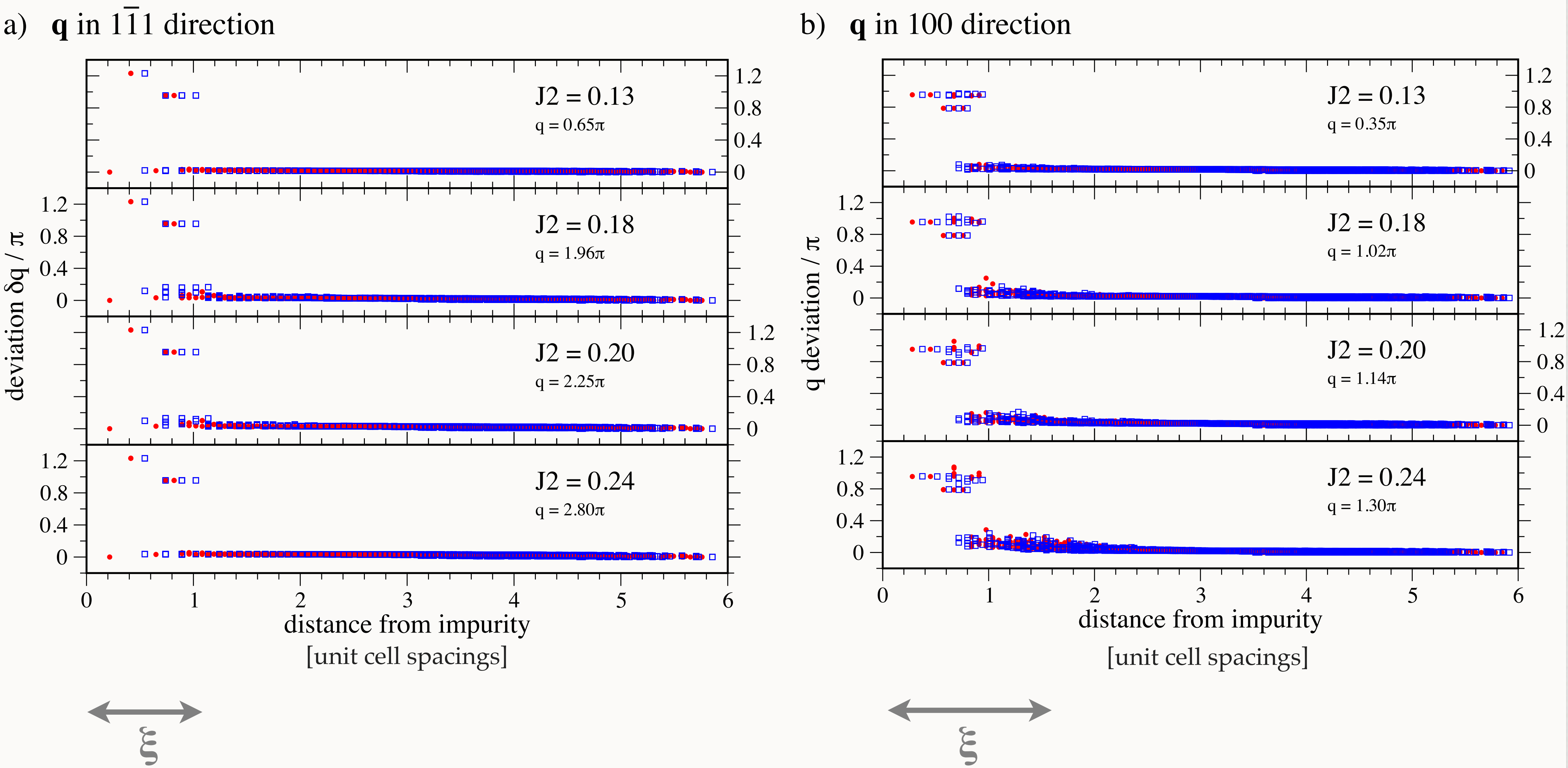
FAVORED DIRECTIONS

swiss cheese => $\overline{E}(\mathbf{q}) = \frac{1}{4} E_a(\mathbf{q})$

- impurity-induced order phase diagram:

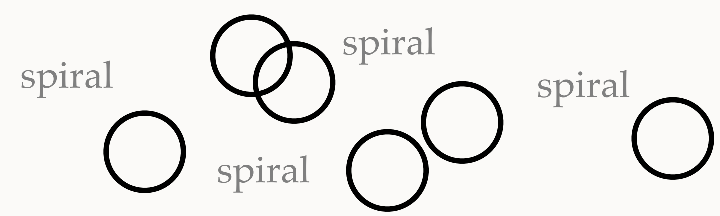


DECAY LENGTH ξ

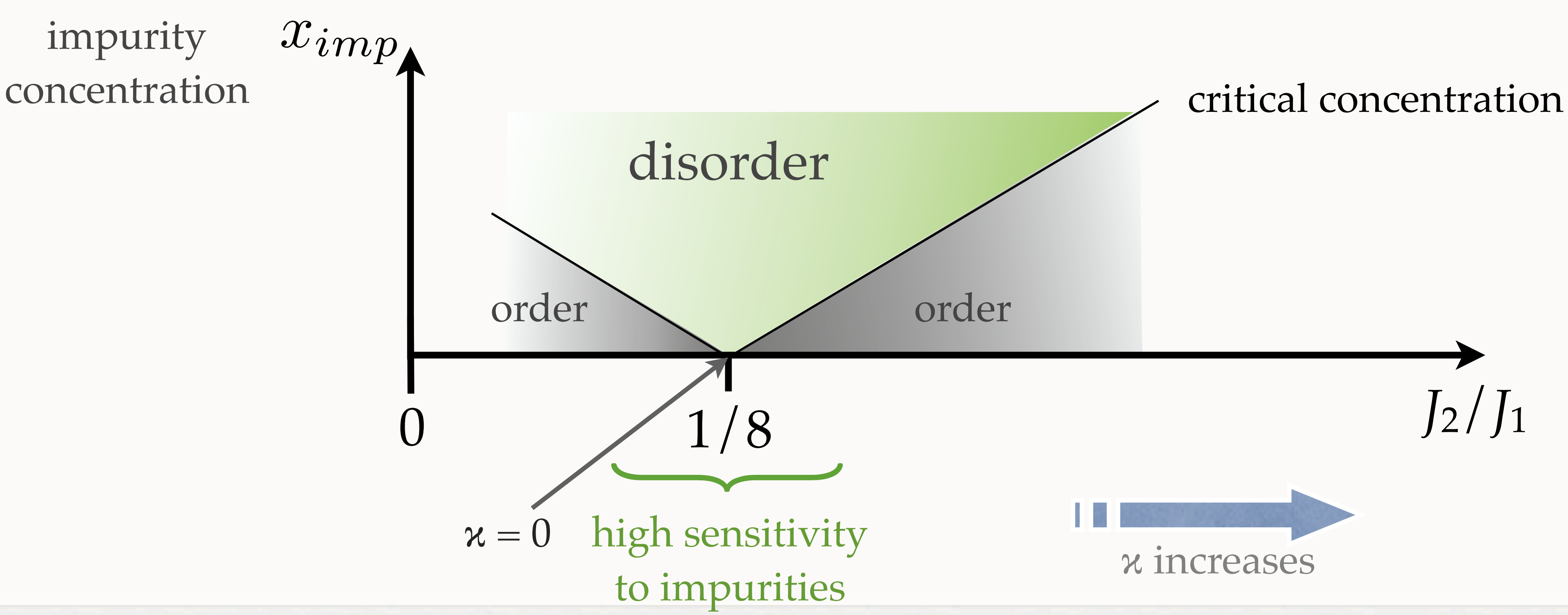


WHY DOES IT SOMETIMES BREAK DOWN?

- impurity concentration too high:
  - the "holes" overlap



  - critical concentration



NB: this is a sketch

vanishing stiffness: very high sensitivity to defects

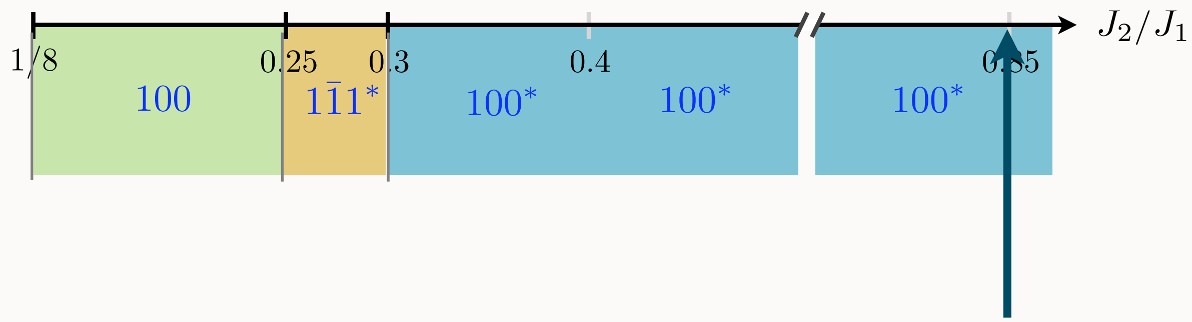
COMPARISON WITH EXPERIMENTS

WHAT WE COMPARE

- Do impurities matter at the order v/s disorder level?
- If order is what happens, is order-by-quenched-disorder the degeneracy-breaking mechanism?
- Interpretation of new experimental data on $CoAl_2O_4$

COMPARISON WITH EXPERIMENTS

- $MnSc_2S_4$



  - order in 110 direction, $J_2/J_1 \sim 0.85$

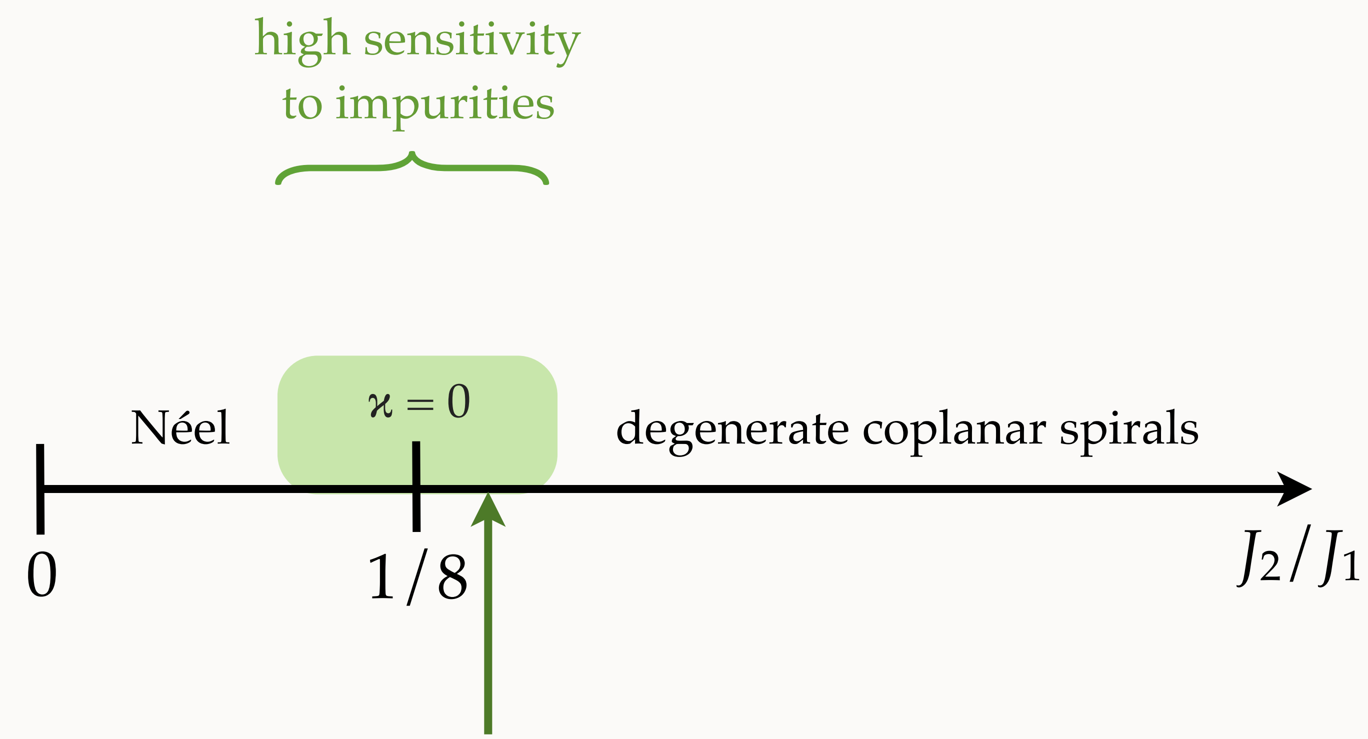
Krimmel *et al.* 2006

    - consistent with non-small stiffness $\varkappa$ ✓
    - direction not that of impurities (or that of a different type of impurities) ✗
    - also, J_3 is important, cf. Lee+Balents 2008

need parameters of other ordered materials

COMPARISON WITH EXPERIMENTS

- $CoAl_2O_4$



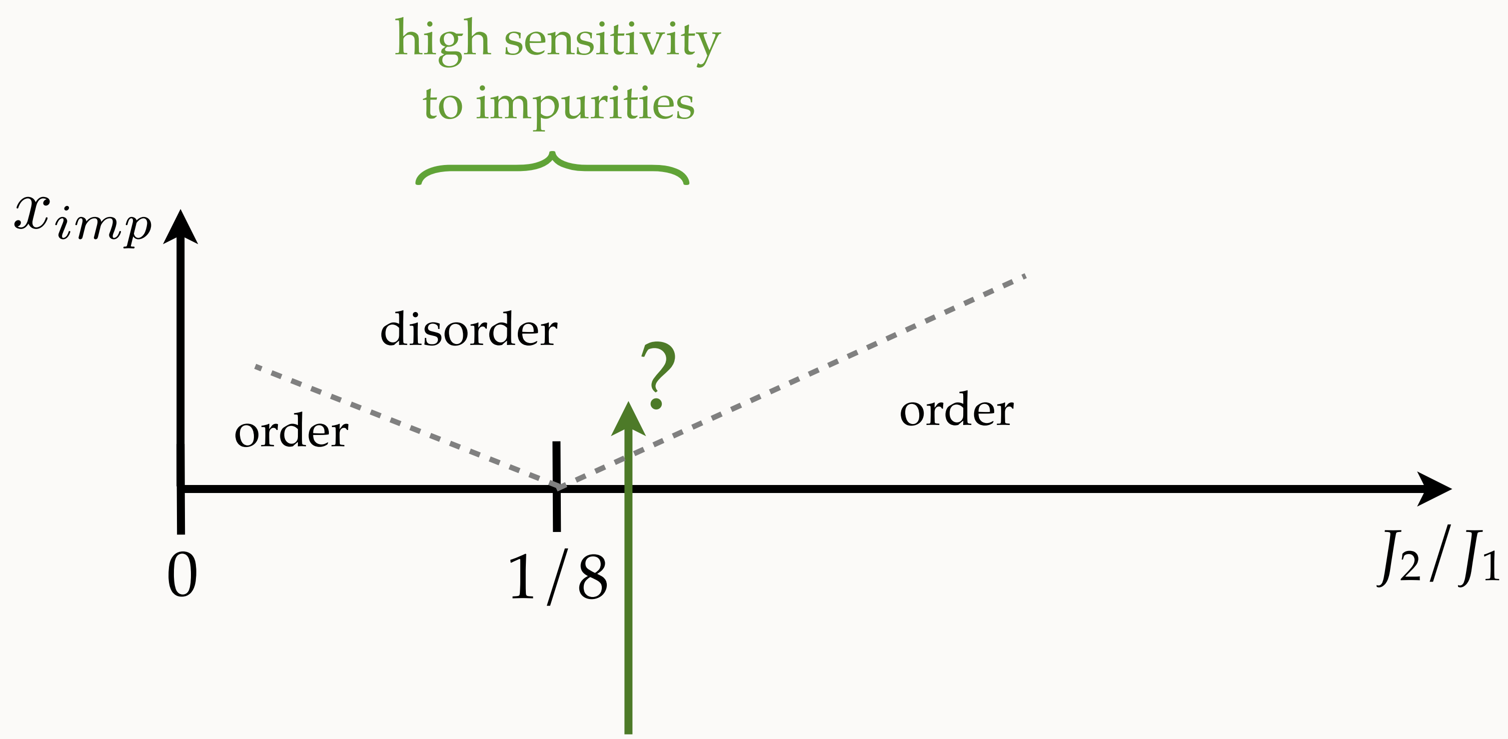
- glassy state, $J_2/J_1 \sim 0.17 \gtrsim 1/8$

Tristan *et al.* 2005, Krimmel *et al.* 2009

  - consistent with vanishing stiffness $\varkappa$ at $J_2/J_1 = 1/8$ ✓

COMPARISON WITH EXPERIMENTS

- $CoAl_2O_4$



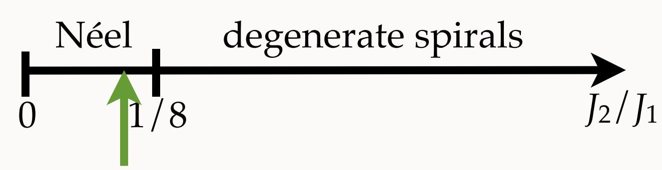
  - glassy state, $J_2/J_1 \sim 0.17 \gtrsim 1/8$

Tristan *et al.* 2005, Krimmel *et al.* 2009

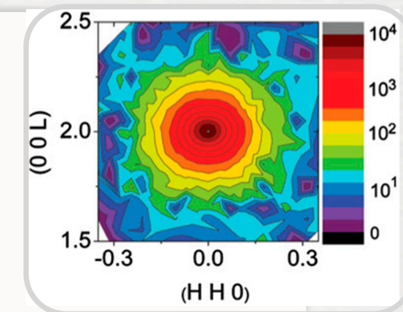
    - consistent with vanishing stiffness $\varkappa$ at $J_2/J_1 = 1/8$ ✓

NEW DATA ON $CoAl_2O_4$

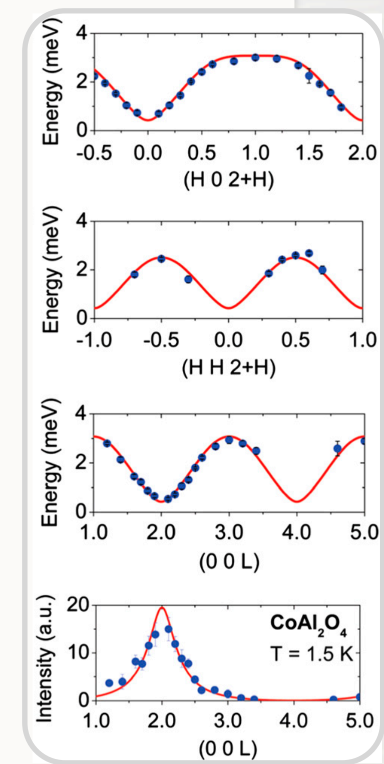
- No LRO, $T = 0$ correlation length ~ 10 r_{nn}
- spin waves -> $J_2/J_1 = 0.1$

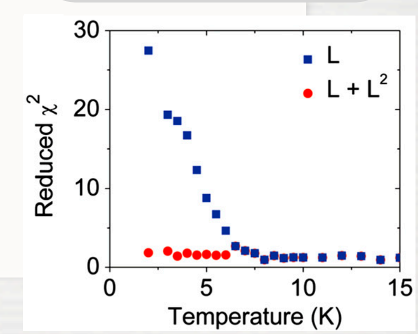


- Lineshape with large Lorentzian-squared component below T^* ~ 6.5K
    - random fields?
    - domains?
- Kinetics important?



correlations at $T = 3.5$K





MacDougall *et al.* 2011

SUMMARY AND PERSPECTIVES

- Summary:
    - very **general conclusions**
    - in general, impurities lead to **order**
    - cause of swiss cheese model: degeneracy manifold is a **curved surface**
    - physics of the swiss cheese model: **independent impurities** (+subtleties)
    - gives **criteria for sensitivity** to defects
    - allows to account for **different behaviors in single class of materials**
- Perspectives
    - compare with more materials or models
    - need **more materials close to Lifshitz point** to correlate glassiness with region of phase diagram
    - nature of glassy phase for stronger disorder/smaller stiffness?
    - quantum systems near Lifshitz point
    - ...

INTERNATIONAL *RESEARCH*

Thank you for your attention