Simultaneous Charge Ordering and Spin Dimerization: Theory of the Spin-SAF Transition

Gennady Y. Chitov
Laurentian University
Sudbury, ON, Canada

Collaborators:
Claudius Gros, *U. of Frankfurt (Germany)*
Jesse Leeson, *Laurentian U. (Canada)*

Outline:

- ***Motivation***
- ***Spin-Pseudospin Model (XX, XXX) and Methods***
- ***Phase Diagram***
- ***Dimerized XXX Chain***
- ***BCS Ratio***
- ***More Applications of Spin-SAF Theory to*** NaV_2O_5
- ***Conclusions***

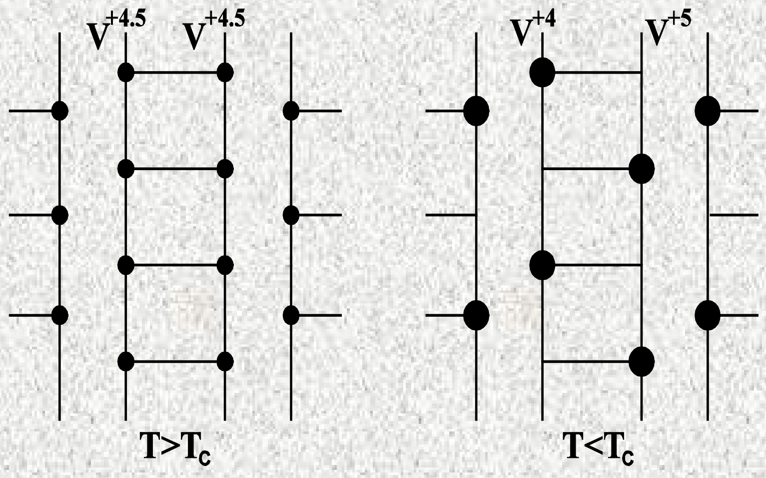
Phase Transition in NaV_2O_5: Experiment

- 1/4-filled ladder compound:
  Smolinski, et al, 1998

- Spin Gap: $T_c \approx 34K,\ \Delta_{sg} \approx 106K$
  Isobe & Ueda, 1996

- 2D charge order:
  van Smaalen, et al, 2002
  Grenier, et al, 2002
  Chitov & Gros, 2004 (SAF)

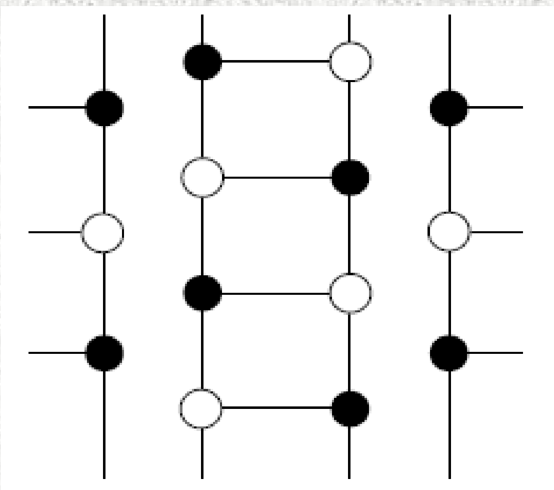
- Phase transition:
  Thermal, close to 2D Ising universality class
  Ravy, et al, 1999
  Gaulin, et al, 2000
  Fagot-Revurat, et al, 2000

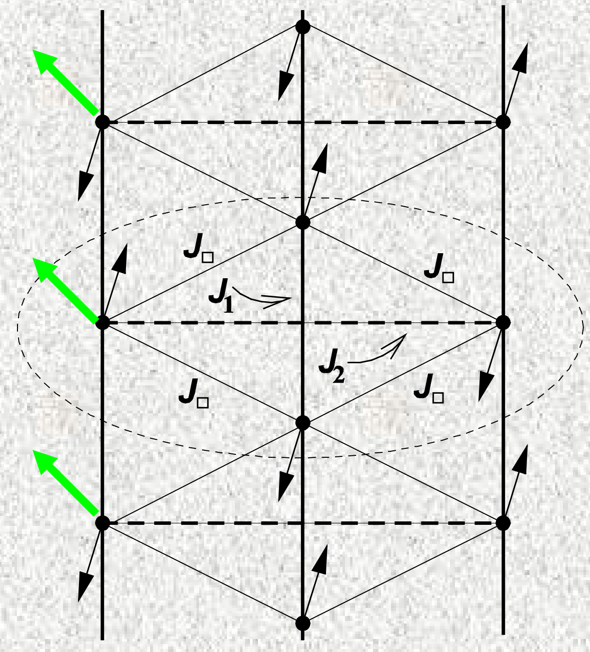


Mechanism:

spin-Peierls - NO
Charge+Spin - ??

Mapping onto the Spin-Pseudospin Model





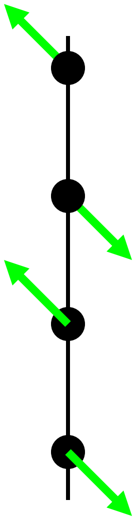
Left/Right (Charge) ⟺ Pseudospin (Ising) Up/Down

Spin ⟺ Spin

K.I. Kugel and D.I. Khomskii, *Usp. Fiz. Nauk* **136**, 621 (1982) [*Sov. Phys. Usp.* **25**, 231 (1982)].

M.V. Mostovoy and D.I. Khomskii, *Solid State Commun.* **113**, 159 (1999).
D. Sa and C. Gros, *Eur. Phys. J.* **B18**, 421 (2000).

Spin Sector: Heisenberg Chains

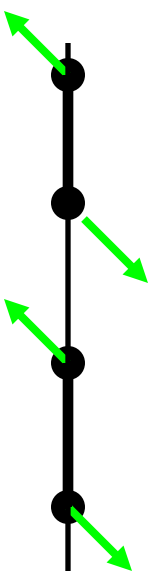


$$\mathcal{H} = J \sum_n \mathbf{S}_n \mathbf{S}_{n+1}$$

Heisenberg Chain:

Gapless State (Luttinger Liquid Universality Class)

No Magnetic LRO



$$\mathcal{H} = J \sum_n [1 + (-)^n \delta] \mathbf{S}_n \mathbf{S}_{n+1}$$

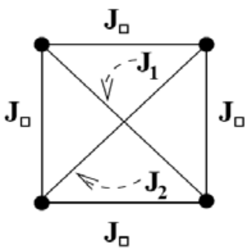
Dimerized Heisenberg Chain:

Gapped GS

No Magnetic LRO

T. Giamarchi, *Quantum Physics in One Dimension* (Oxford University Press, Oxford, 2004).

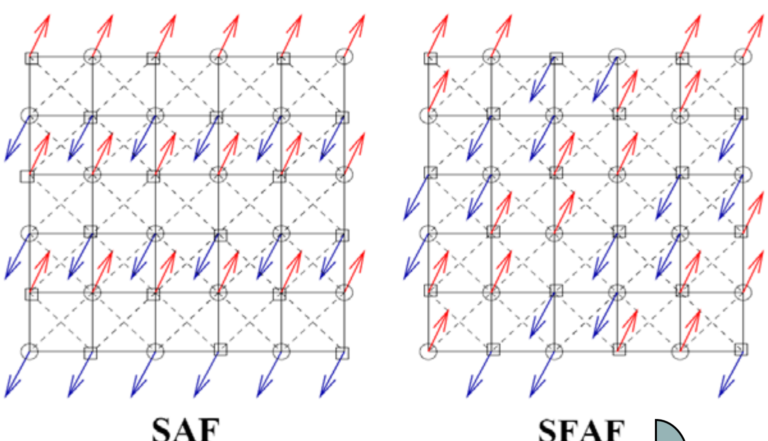
Charge Sector: 2D (nn+nnn) Ising Model



$$H = \frac{1}{2}\sum_{\langle \mathbf{i},\mathbf{j}\rangle} J_\square \mathcal{T}^x_{\mathbf{i}} \mathcal{T}^x_{\mathbf{j}} + \frac{1}{2}\sum_{\langle\langle \mathbf{k},\mathbf{l}\rangle\rangle} J_{\mathbf{kl}} \mathcal{T}^x_{\mathbf{k}} \mathcal{T}^x_{\mathbf{l}}$$

Ground-State Phase Diagram

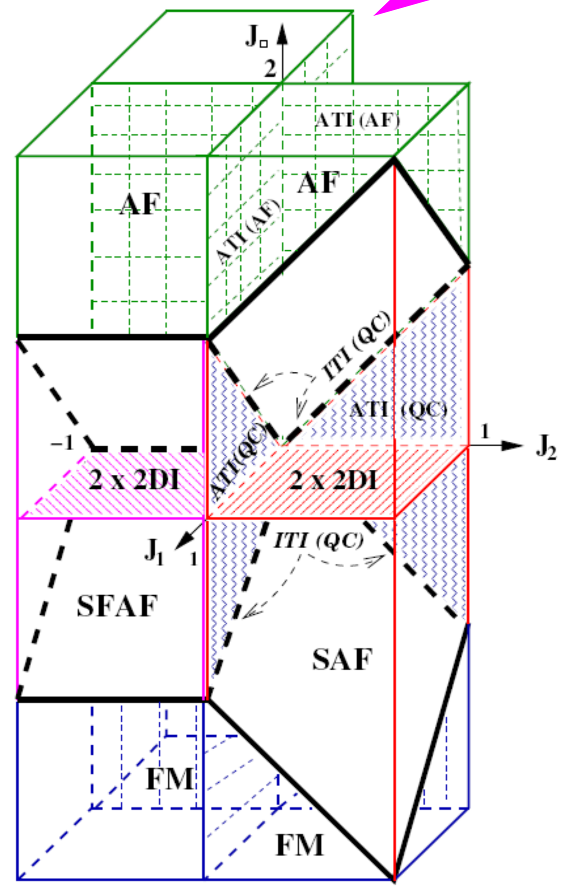
Frustrations!!



Fan & Wu, 1969
NB: Fe-pnictides

Super-AntiFerromagnetic

4x4



Chitov & Gros, 2005

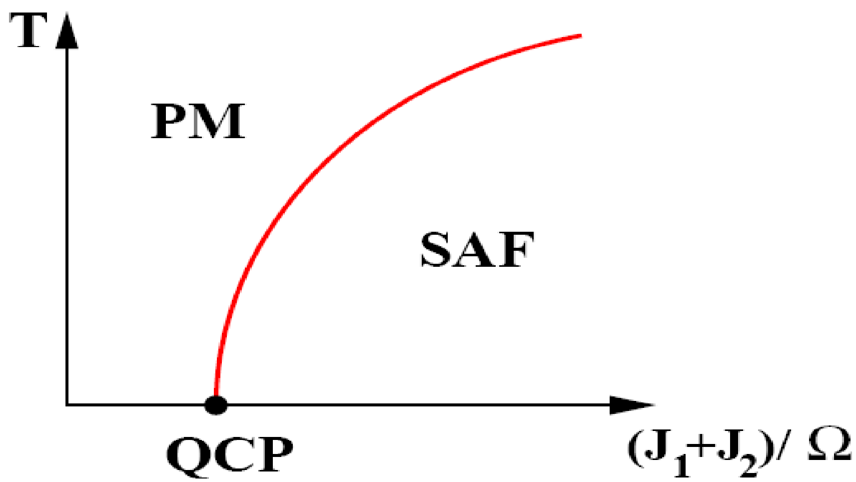
Charge Sector (Getting Worse): 2D (nn+nnn) Ising Model + Transverse Field

- Transverse (Quantum) Ising Model:

$$H = \frac{1}{2} \sum_{\mathrm{nn,nnn}} J_{\mathbf{kl}} \mathcal{T}_{\mathbf{k}}^{x} \mathcal{T}_{\mathbf{l}}^{x} - \Omega \sum_{\mathbf{i}} \mathcal{T}_{\mathbf{i}}^{z}$$

- Special Case: $J_{\square}^{2} < 4 J_1 J_2 , \ (J_1 , J_2) > 0$

2 Phases (PM/SAF) with a Quantum Critical Point (QCP)



Coupled Spin-Pseudospin Model and Approximation

$$\mathcal{H}_{\rm IMTF} = -\Omega \sum_{\mathbf{k}} \mathcal{T}^z_{\mathbf{k}} + \frac{1}{2} \sum_{nn,nnn} J_{\mathbf{k},\mathbf{l}} \mathcal{T}^x_{\mathbf{k}} \mathcal{T}^x_{\mathbf{l}}$$

Transverse (Quantum) Ising Model Hamiltonian

$$\mathcal{H} = \mathcal{H}_{\rm IMTF} + \sum_{m,n} \mathbf{S}_{mn} \mathbf{S}_{m,n+1} \left[J + \lambda \mathcal{T}^z_{mn} \mathcal{T}^z_{m,n+1} + \varepsilon \left(\mathcal{T}^x_{m+1,n+1} - \mathcal{T}^x_{m-1,n} \right) \right]$$

Coupled Model

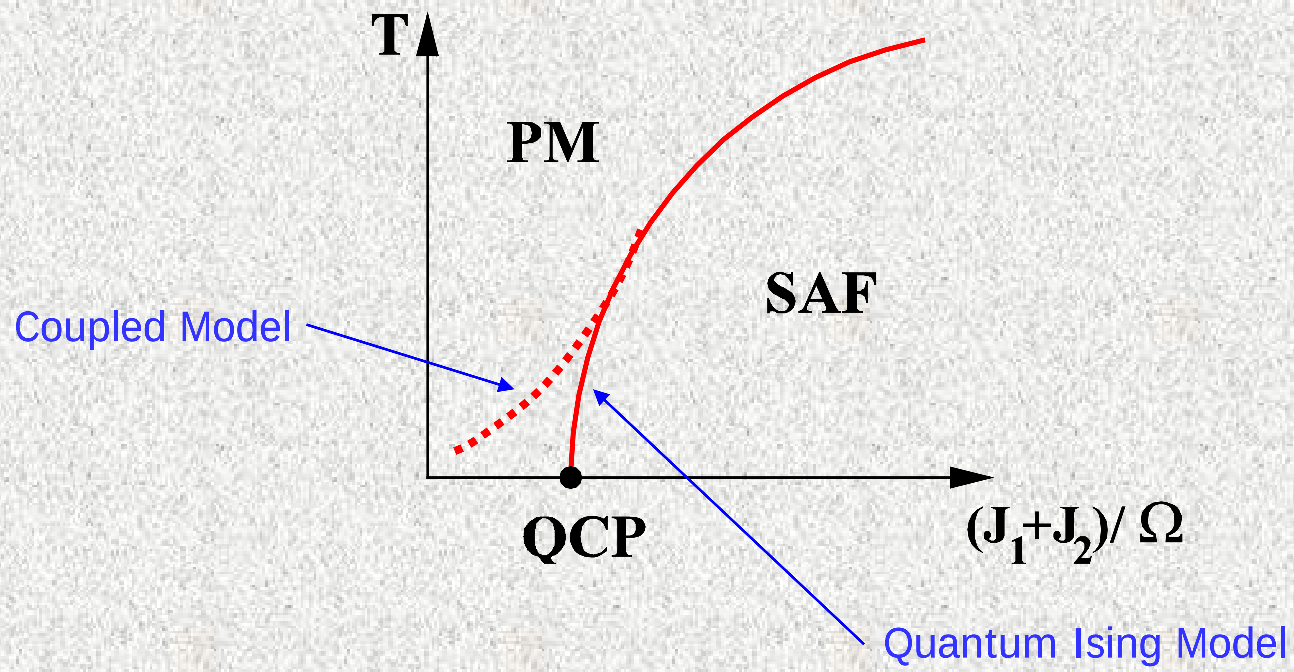
Creates Dimerization in the SAF phase

Approximation:

Ising Sector -- Mean-Field

Spin Sector (XX, XXX) -- Exactly

Spin-Pseudospin (Coupled) Model: Phase Diagram

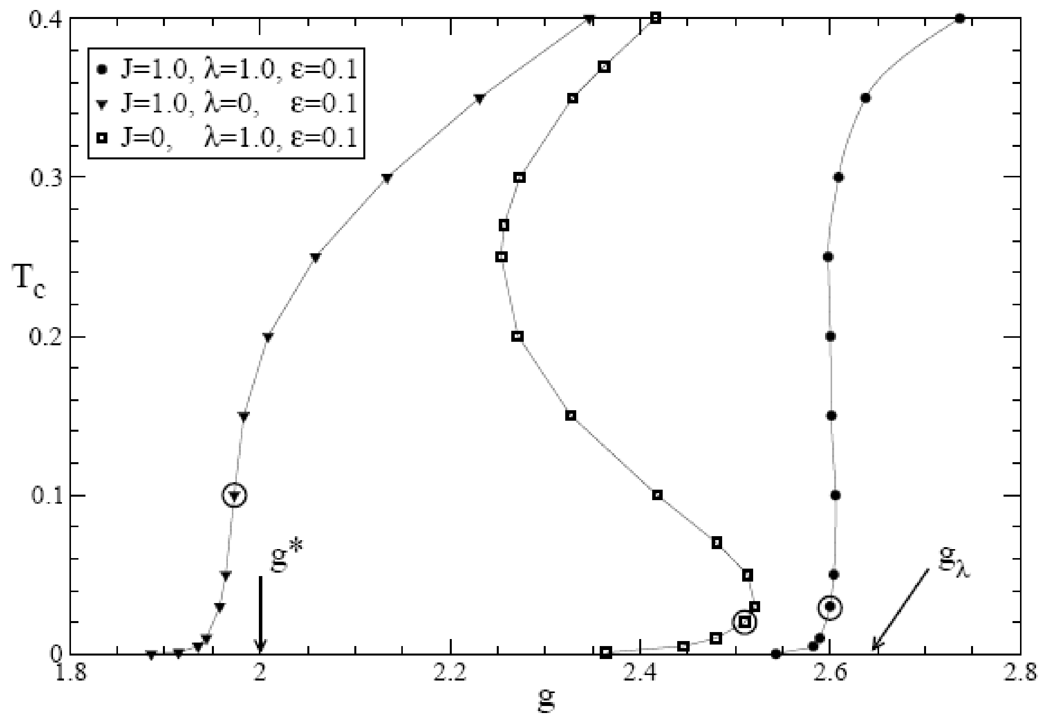


- **Coupled Model always orders via** *spin-SAF transition*
- **QCP is destroyed**
- **Ordered (spin-SAF) Phase:**
  SAF charge/Ising LRO +
  Spin Gap/Dimerization

Coupled (XY) Model: Phase Diagram (detailed)

- $\mathcal{H} = -\sum_{m,n} \mathcal{T}^z_{mn} + \frac{1}{2}\sum_{m,n} g_{\sharp} \mathcal{T}^x_{mn}\mathcal{T}^x_{m,n+1}$
  $+\sum_{m,n} \mathbf{S}_{mn}\mathbf{S}_{m,n+1}\left[J + \lambda \mathcal{T}^z_{mn}\mathcal{T}^z_{m,n+1} + \varepsilon\left(\mathcal{T}^x_{m+1,n+1} - \mathcal{T}^x_{m-1,n}\right)\right]$

Spin Sector =
Free Spinless Fermions



$$T_c \approx \begin{cases} \frac{g}{4}, & g \gg g_\lambda \\ \frac{\mathbb{A}\tilde{J}}{2}\exp\left[-\frac{\pi \tilde{J}}{4\varepsilon^2}(g_\lambda - g)\right], & \text{BCS regime} \end{cases}$$

BCS ratio

$$\frac{\overset{\circ}{\Delta}_{\mathrm{SG}}}{T_c} = \frac{\pi}{e^{\gamma}} \approx 1.76, \quad \text{BCS regime}$$

Chitov & Gros, 2004-2005

Coupled Model: XXX (Heisenberg) Spin Sector

$$\mathcal{H} = \mathcal{H}_{\rm IMTF} + \sum_{m,n} \mathbf{S}_{mn}\mathbf{S}_{m,n+1}\left[J + \varepsilon\left(\mathcal{T}^x_{m+1,n+1} - \mathcal{T}^x_{m-1,n}\right)\right]$$

Spin Sector → **Dimerized XXX Chains**

$$\mathcal{H}_{\rm XXX} = J\sum_n [1 + (-)^n\delta]\mathbf{S}_n\mathbf{S}_{n+1}$$

↕

Interacting Spinless Fermions

↓

Bosonization

Dimerized XXX Heisenberg Chain

$$\mathcal{H}_{\rm XXX} = J\sum_n [1+(-)^n\delta]\mathbf{S}_n\mathbf{S}_{n+1}$$

Sine-Gordon

$$v^{-1}H_{\rm sG} = \frac{1}{2}\int dx\left(\Pi^2 + (\partial_x\phi)^2\right) + 2\mu\int dx\cos\sqrt{2\pi}\phi\ ,$$

$v = \frac{\pi}{2}J$

$$\mu = \frac{A_\epsilon}{\pi}\delta$$

$$A_\epsilon = \frac{3}{\pi^2}\left(\frac{\pi}{2}\right)^{\frac{1}{4}}$$

??? Orignac, 2004

Marginal Term $\propto \cos\sqrt{8\pi}\phi$

Plus !!

Log-corrections

Dimerized XXX Heisenberg Chain = Sine-Gordon

$$f_s(T,\delta) = -Jt_\circ - \frac{1}{3}\frac{T^2}{J} - \frac{1}{2}\frac{J^2 a_\circ}{T}\delta^2$$

$$a_\circ \equiv \frac{1}{4}\left(\frac{\Gamma(1/4)}{\Gamma(3/4)}\right)^2 A_\epsilon^2 \equiv \vartheta_1 A_\epsilon^2$$

$$T_c = \vartheta_1 A_\epsilon^2 \frac{\varepsilon^2}{\mathcal{J}_c - \mathcal{J}}$$

$$f_s(0,\Delta_\circ) = -Jt_\circ - \frac{1}{2\pi\sqrt{3}}\frac{\Delta_\circ^2}{J}$$

Al. Zamolodchikov, 1995

$$\Delta_\circ = J\sqrt{\pi}\left(\frac{\Gamma(3/4)}{\Gamma(1/4)}\right)^{2/3}\frac{\Gamma(1/6)}{\Gamma(2/3)}A_\epsilon^{2/3}\delta^{2/3}$$

$$\Delta_\circ = \vartheta_2 A_\epsilon^2 \frac{\varepsilon^2}{\mathcal{J}_c - \mathcal{J}} \ , \quad T = 0$$

BCS Ratio:

$$\frac{\Delta_\circ}{T_c} = \frac{\vartheta_2}{\vartheta_1} = 6\sqrt{3}\frac{(\Gamma(1/3))^9}{(\Gamma(1/4))^8} = 2.47\ldots$$

Orignac, 2004

Chitov et al, 2009, unpub

Dimerized XXX Heisenberg Chain *(Continued)*

$$f_s \approx -Jt_\circ - \alpha J \frac{\delta^{4/3}}{\ln \frac{\delta_\circ}{\delta}}, \quad T = 0$$

Specific Free Energy

$$\Delta_{\mathrm{SG}} = \sqrt{\alpha_S} J \frac{\delta^{2/3}}{\ln^{\frac{1}{2}} \frac{\delta_S}{\delta}}, \quad T = 0$$

Spin Gap

$$\eta_n \propto \frac{\partial f_s}{\partial \delta}$$

Dimerization Susceptibility

$$\eta_n \approx \frac{4\alpha}{3} \frac{1}{\delta^{2/3} \ln \frac{\delta_\circ}{\delta}}, \quad T = 0$$

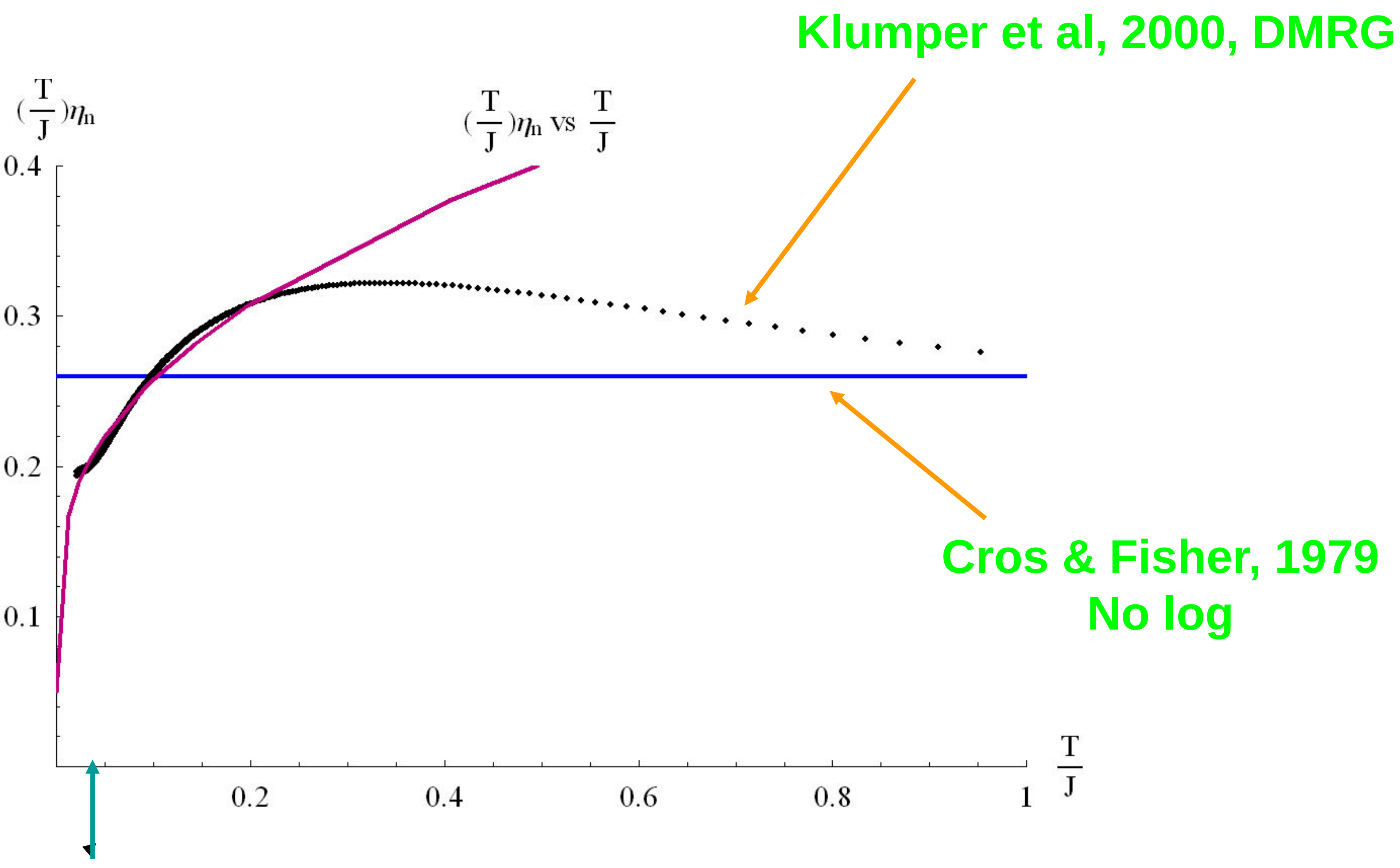
Problem:
“Prefactors and Cutoffs”

$$\eta_n \approx \frac{a_\circ J}{T} \frac{1}{\ln^{3/2} \frac{CJ}{T}}, \quad \delta = 0$$

Refs: Black & Emery, 1981; Affleck, et al, 1989

Dimerization Susceptibility

$$\eta_n \approx \frac{a_\circ J}{T} \frac{1}{\ln^{3/2} \frac{CJ}{T}}, \qquad \delta = 0$$

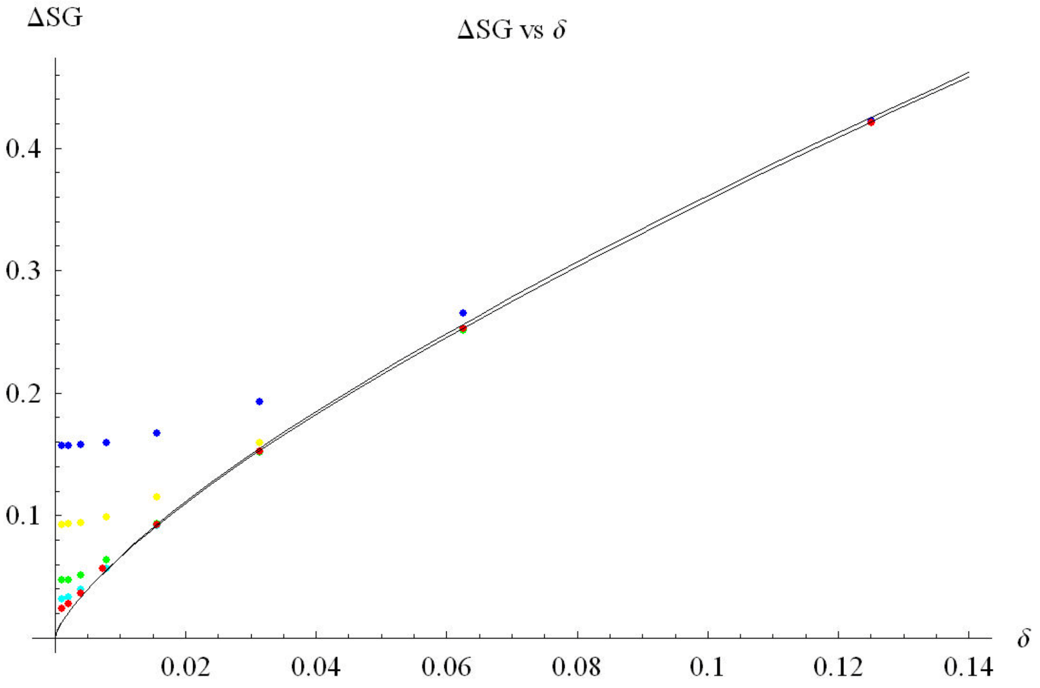


NB: ***NaV2O5: Tc/J =0.06***

Spin Gap

$$\Delta_{\mathrm{SG}} = \sqrt{\alpha_S} J \frac{\delta^{2/3}}{\ln^{\frac{1}{2}} \frac{\delta_S}{\delta}}, \quad T = 0$$

Papenbrock et al, 2003, DMRG



Spin Gap vs T_c : BCS Ratio

XY Dimerized Chain (Free Fermions):

$$\frac{\overset{\circ}{\Delta}_{SG}}{T_c} = \frac{\pi}{e^{\gamma}} \approx 1.76,$$

spin-Peierls, spin-SAF

XXX Dimerized Chain: Neglected Marginal (log) Terms

Orignac & Chitra, 2004, spin-Peierls Theory

$$\frac{\Delta(T=0)}{T_{SP}} \simeq 2.47$$

spin-Peierls, spin-SAF

XXX Dimerized Chain: Marginal (log) Terms Included

$$\frac{\overset{\circ}{\Delta}_{SG}}{T_c} \approx 2.44$$

spin-SAF
spin-Peierls (?)

BCS Ratio -- Experiments:

spin-Peierls: $(\mathrm{Organics}, \, CuGeO_3) \sim 2-4$ (Pouget, 2001)
spin-SAF: $(\mathrm{NaV_2O_5}) \sim 3.1$

Spin-SAF Theory: Further Application to NaV_2O_5

- Parameters:

$$g_\lambda = 4t_a,\ t_a = 0.35\,\mathrm{eV},\ \varepsilon \approx 0.4J_1 = 19.3\,\mathrm{meV}\ \text{(Gros \& Chitov, 2005)}$$
$$T_c = 34\,\mathrm{K} = 2.93\,\mathrm{meV}$$

$$g_\lambda - g \approx 0.132\,\mathrm{eV},\ \text{or}\ g/g_\lambda \approx 0.91$$

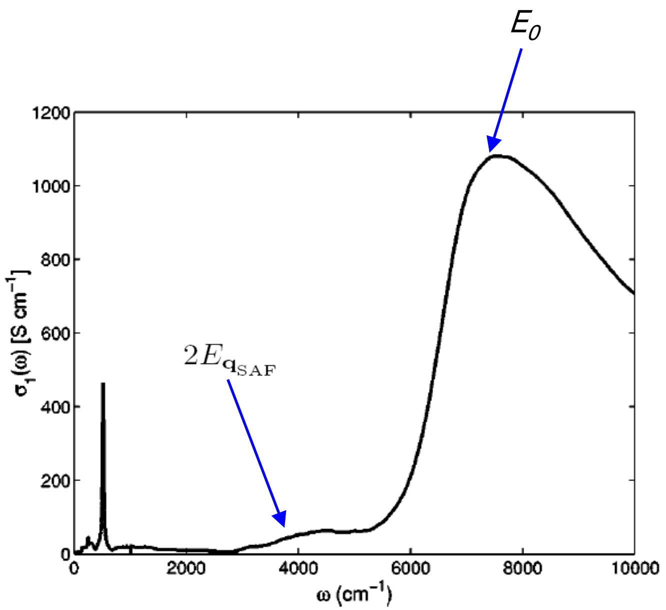
- Pseudospin (charge) excitations (theory):

$$E_{0/\mathbf{q}_{\mathrm{SAF}}} = 2t_a\sqrt{1 \pm g/g_\lambda}$$

$$E_0 \approx 0.97\,\mathrm{eV} \approx 7800\ cm^{-1}$$

$$E_{\mathbf{q}_{\mathrm{SAF}}} \approx 0.21\,\mathrm{eV} \approx 1700\ cm^{-1}$$

$2E_{\mathbf{q}_{\mathrm{SAF}}} \approx 3400\ cm^{-1} \longmapsto$ broad peak near $4000\ cm^{-1}$
$\longmapsto$ 2-particle (pseudospin) excitations with $\mathbf{q}_{\mathrm{SAF}}$

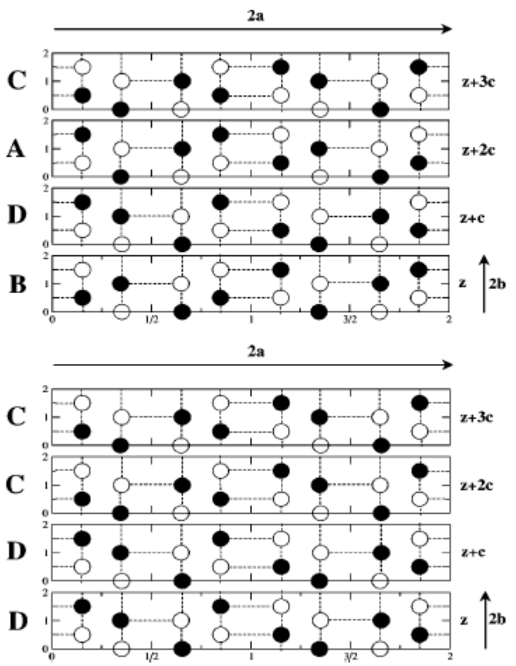


Optical Conductivity: Damascelli, et al, 2000.

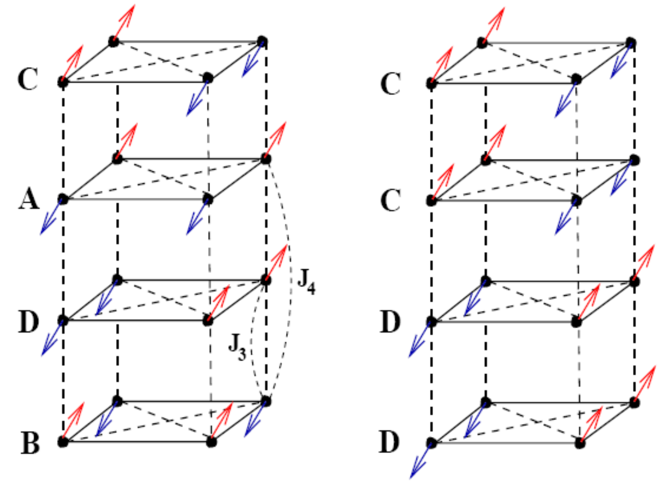
Stacking Charge Order in NaV_2O_5

- Charge order in NaV_2O_5 (X-ray): from Grenier, *et al*

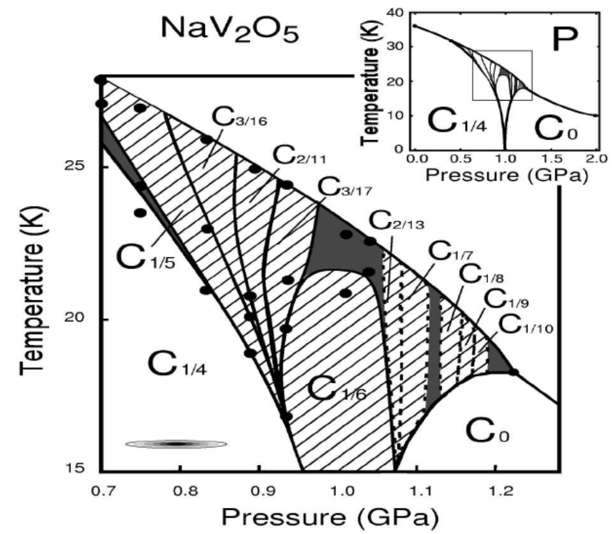
PHYSICAL REVIEW B **65** 180101(R)



- **Ground states in the nn and nnn axial Ising model**



- **Devil's staircase in NaV_2O_5: from Ohwata, et al PRL'01**



Summary & Discussion

1. Spin-pseudospin model is proposed and the theory of spin-SAF transition is developed.
   Spin-SAF phase = simultaneous Super-Anti-Ferroelectric (SAF) charge order + spin gap.

2. BCS ratio is calculated.
   Similarities between spin-SAF and spin-Peierls are emphasized.

3. Numerical parameters of the effective Hamiltonian
   Peaks in the optical conductivity, absence of the soft mode

4. Further progress:
   - Ising (pseudospin / charge) sector beyond mean-field (??)
   - Fate of QCP?