An introduction to the spin-Peierls compound $CuGeO_3$

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* introduction to $CuGeO_3$
  - experiments: phase diagram
  - model Hamiltonian: parameters

* theory: static and dynamic phonons
  - Jordan-Wigner transf./bosonization
  - Cross-Fisher approach
  - $CuGeO_3$: soft-phonon problem
  - $CuGeO_3$: magnetoelastic parameters

$CuGeO_3$

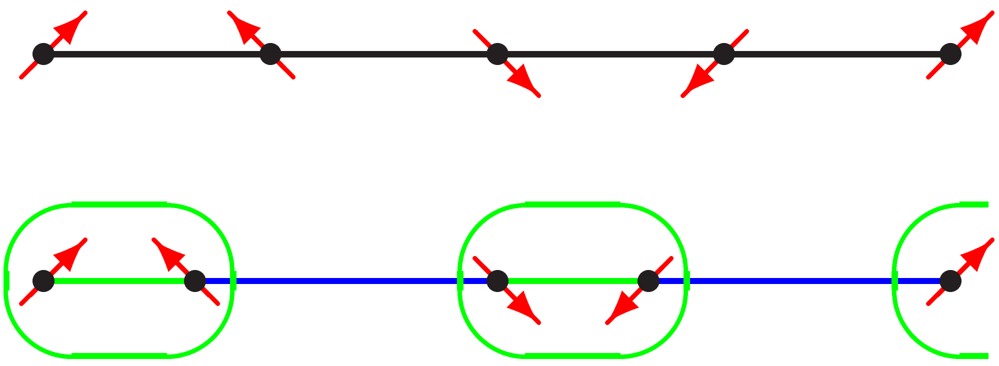
- $\ast$ first inorg. spin-Peierls compound
- $\ast$ coexistence: spin-Peierls + AF
- $\ast$ frustrating magn. interactions

spin-Peierls

- $\ast$ structural phase-transiton
  (magnetoelastic coupling)
- $\ast$ spin-1/2 moments $\rightarrow$ quasi-1D AF
  spin-excitations $\rightarrow$ continuum ($T > T_{SP}$)
  $\rightarrow$ energy gap ($T < T_{SP}$)

$T > T_{SP}$

$T < T_{SP}$



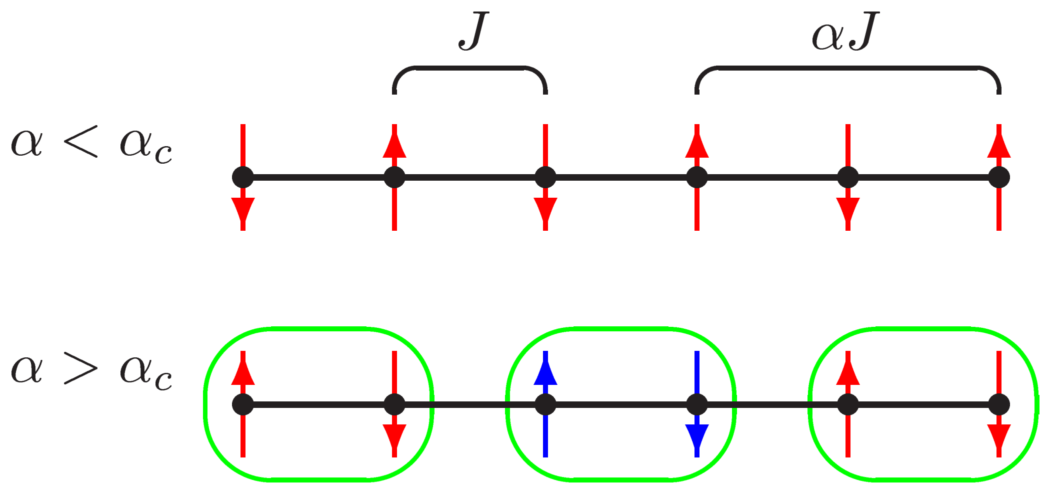
Hamiltonian

$(T > T_{SP})$

$$\boxed{H = J \sum_n \left[\, \mathbf{S}_n \cdot \mathbf{S}_{n+1} + \alpha\, \mathbf{S}_n \cdot \mathbf{S}_{n+2} \,\right]}$$

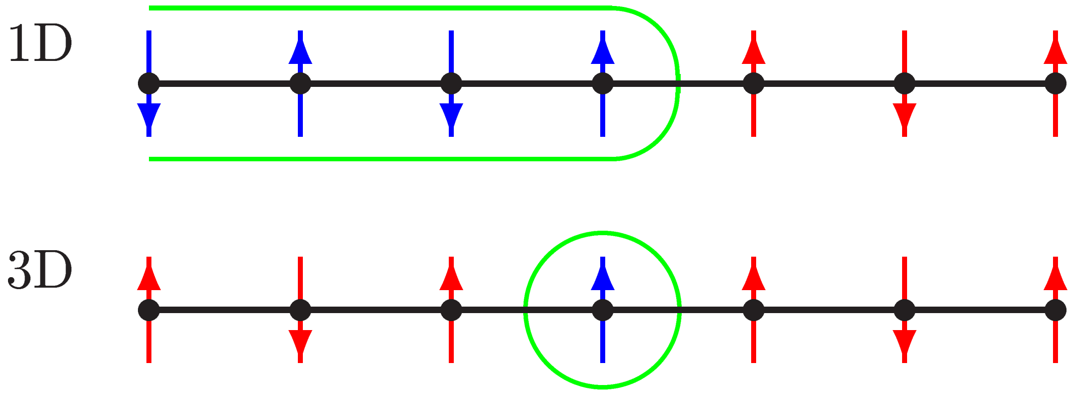
* $J \sim 156\,\mathrm{K}, \;\; \alpha \sim 0.24 - 0.35$

* $\alpha > \alpha_c \sim 0.24$: energy-gap $\Delta > 0$

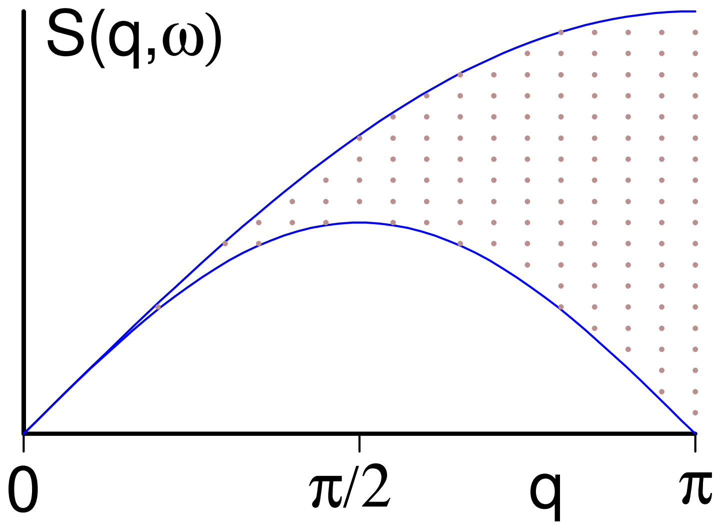


spinon vs. magnon

(1D vs. 3D)



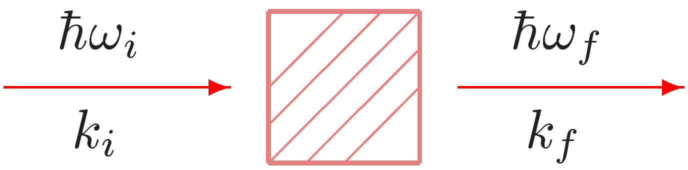
2-spinons-continuum



* $v_s \sim \frac{\pi}{2} J(1-1.12\alpha)$

[Fledderjohann and Gros, EPL '97]

Raman scattering



- CuGeO$_3$: k_i, $\mathrm{k}_f \parallel \hat{c}$ (magn. Raman)
- $\Delta k = k_f - k_i \ll \pi/c, \quad \Delta k \sim 0$
- $\Delta E = \omega_f - \omega_i$: 1 phonon
  : 2 magnons (4 spinons)

magn. Raman

$(T > T_{SP})$

$$\boxed{H_R = g \sum_n \mathbf{S}_n \cdot \mathbf{S}_{n+1} \quad \rightarrow \quad [H_R, H] \sim \alpha}$$

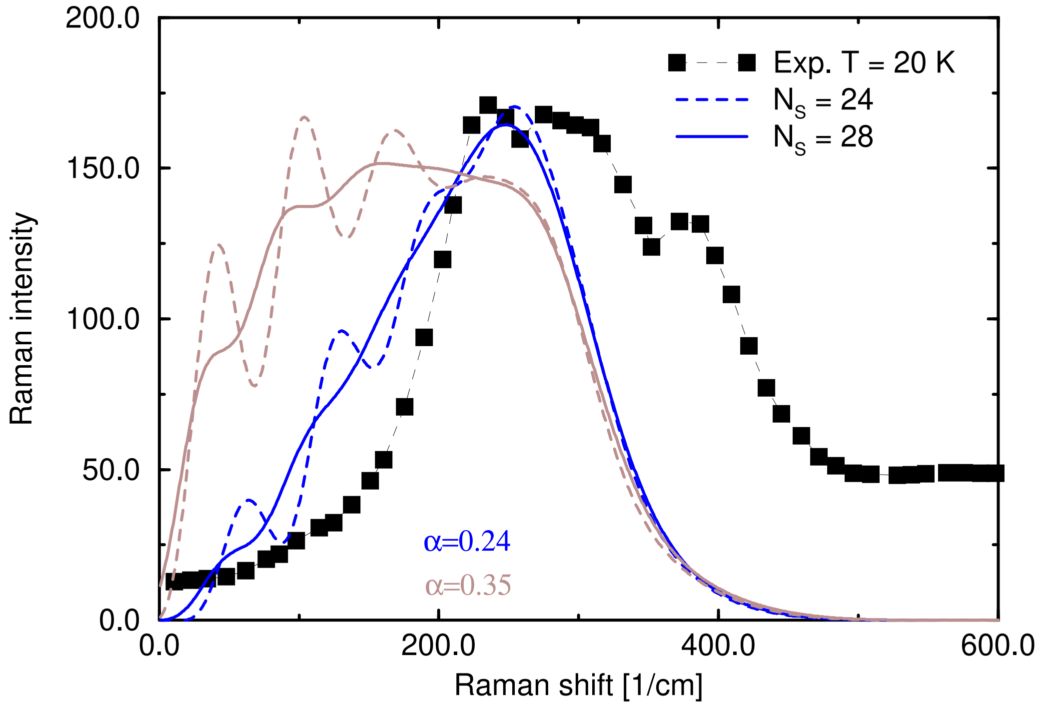
- exact diagonalization
- molecular-field theory (Jordan-Wigner)

exact diagonalization

$$I_R(\omega) = \frac{-1}{\pi} Im \langle \psi_0 | H_R \frac{1}{\omega - (H - E_0)} H_R | \psi_0 \rangle$$

$*$ chain length $N_s = 16, 24, 28$

[Gros et al., PRB '97]



$*$ $T > T_{SP}$: 4-spinon continuum

$*$ $\rightarrow$ $\alpha \lesssim \alpha_c = 0.24$

(susceptibility: $\alpha \approx 0.35$)

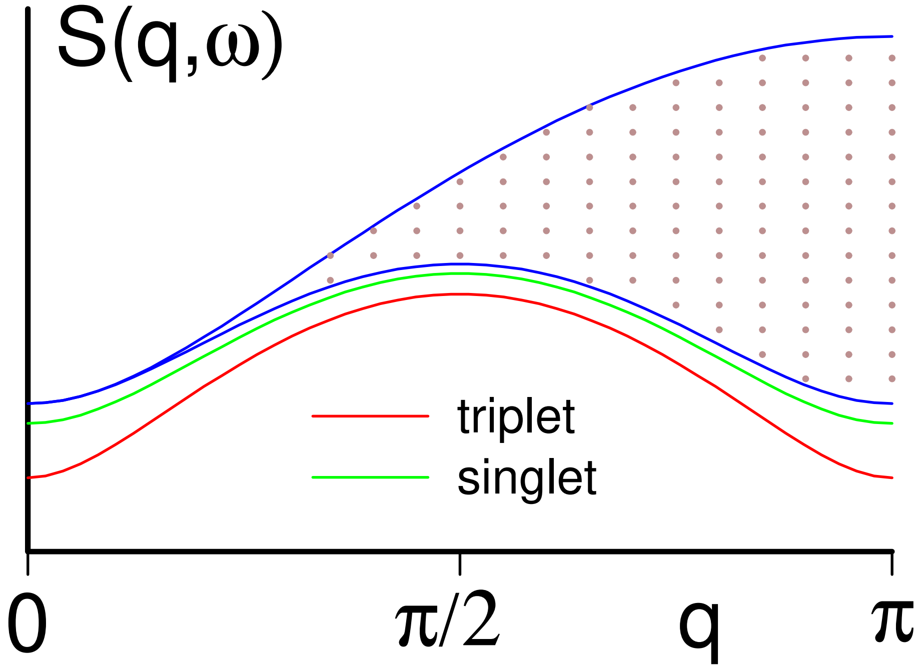
spin-Peierls phase

$$T < T_{SP}$$

$$H' = J\delta \sum_n (-1)^n \, \mathbf{S}_n \cdot \mathbf{S}_{n+1}$$

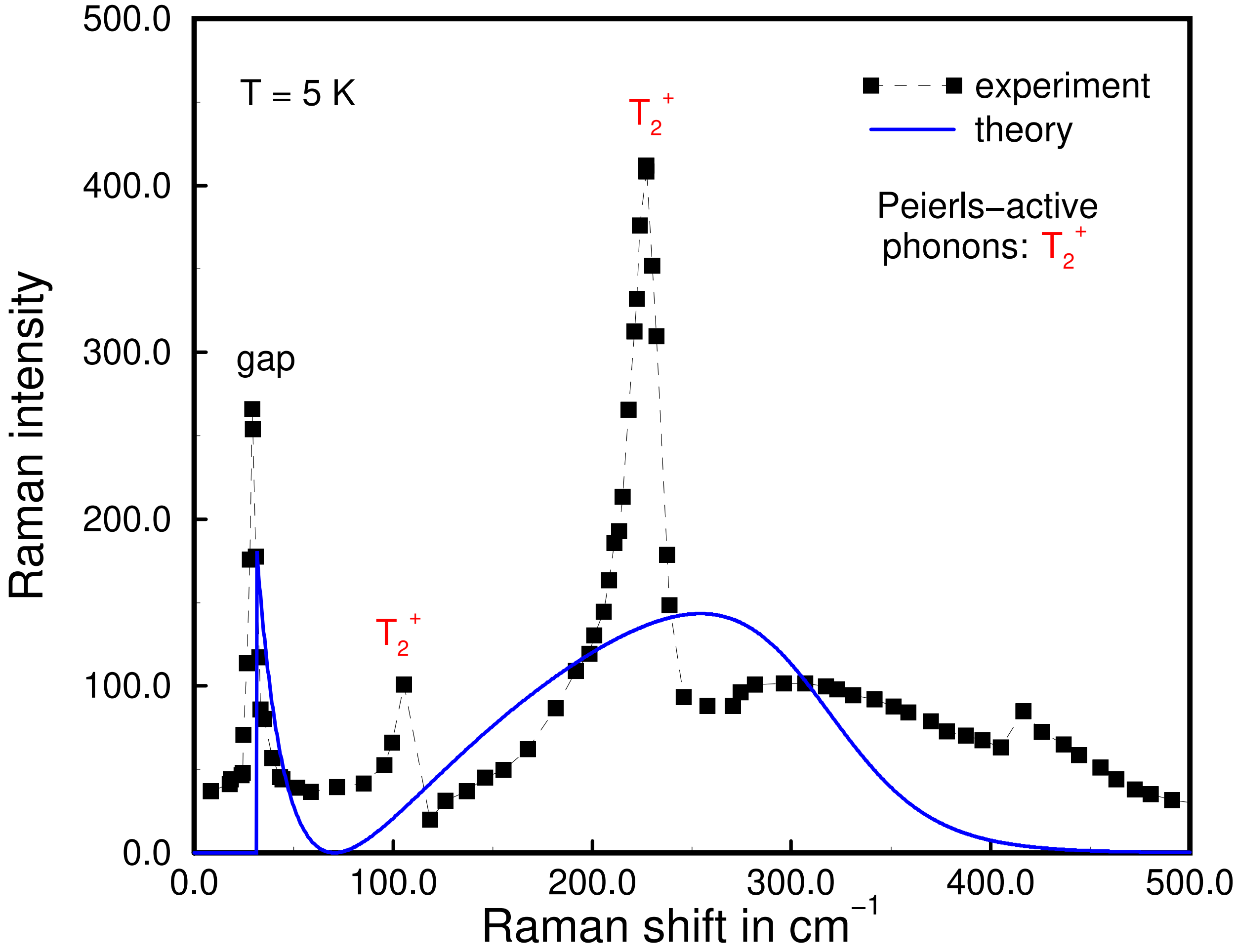
$$H'_R = \Gamma \sum_n (-1)^n \, \mathbf{S}_n \cdot \mathbf{S}_{n+1}$$

* quantum critical: $\Delta \sim \delta^{2/3}$
* bound states: triplet and singlet



Raman: $T < T_{SP}$

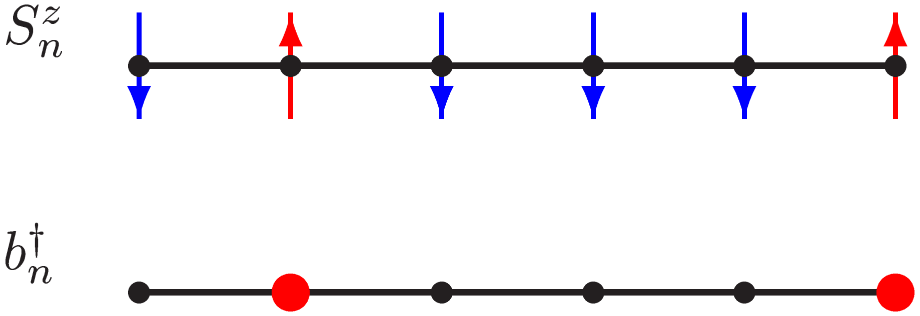
[Muthukumar et al., PRB '96]



- $\ast$ $\Delta(\text{Raman}) \lesssim 2\Delta(\text{neutrons})$
- $\ast$ theory: exact diagonalization
  + analytic fit
- $\ast$ dimerization of the Raman operator

hard-core bosons

$$S_n^+ = b_n^\dagger, \qquad S_n^- = b_n, \qquad S_n^z = b_n^\dagger b_n - \frac{1}{2}$$



* commutation relations

$$[S_n^\alpha, S_m^\beta]_- = i\delta_{n,m}\epsilon_{\alpha,\beta,\gamma} S_n^\gamma, \qquad (\alpha, \beta, \gamma = x, y, z)$$

$$[b_n, b_m^\dagger]_- = 0, \qquad [b_n, b_n^\dagger]_+ = 1$$

* exchange term

$$S_n^x S_{n+1}^x + S_n^y S_{n+1}^y = \frac{1}{2}\left[\, S_n^+ S_{n+1}^- + S_n^- S_{n+1}^+ \,\right]$$

exchange Hamiltonian

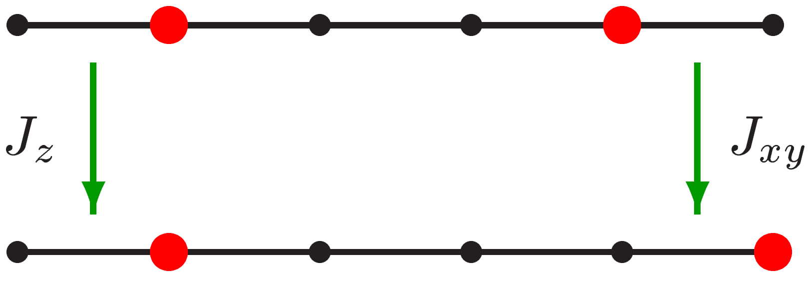
[hard-core boson repr.]

$$H = \sum_n \frac{J_{xy}}{2} \left[S_n^+ S_{n+1}^- + S_n^- S_{n+1}^+ \right] + \sum_n J_z \, S_n^z S_{n+1}^z$$

$$= \sum_n \frac{J_{xy}}{2} \left[b_n^\dagger b_{n+1} + b_n b_{n+1}^\dagger \right]$$

$$+ \sum_n J_z \left[\left(\rho_n - \frac{1}{2} \right) \left(\rho_{n+1} - \frac{1}{2} \right) \right]$$

* $\rho_n = b_n^\dagger b_n$



* $J_{xy} \mathrel{\widehat{=}}$ kinetic energy

Jordan-Wigner transformation

(JW)

$$c_n^\dagger = \mathrm{e}^{i\pi \sum_{m<n} \rho_m}\, b_n^\dagger, \qquad b_n^\dagger = \mathrm{e}^{i\pi \sum_{m<n} \rho_m}\, c_n^\dagger,$$

* $\rho_m = b_m^\dagger b_m = c_m^\dagger c_m$
* non-local transformation
* string $\rightarrow$ phase factor
  $\exp[i\pi \sum_{m<n} \rho_m] = \pm 1 \quad (\rho_m = 0, 1)$
* Fermi-commutation relations

$$\boxed{[c_n, c_m]_+ = 0, \qquad [c_n^\dagger, c_m]_+ = \delta_{n,m}}$$

exchange Hamiltonian

[JW]

$$H = \sum_n \frac{J_{xy}}{2} \left[c_n^\dagger c_{n+1} + c_n c_{n+1}^\dagger \right] + \sum_n J_z \left[\left(\rho_n - \frac{1}{2}\right) \left(\rho_{n+1} - \frac{1}{2}\right) \right]$$

* magnetic field $(-B)$ $\widehat{=}$ chemical potential

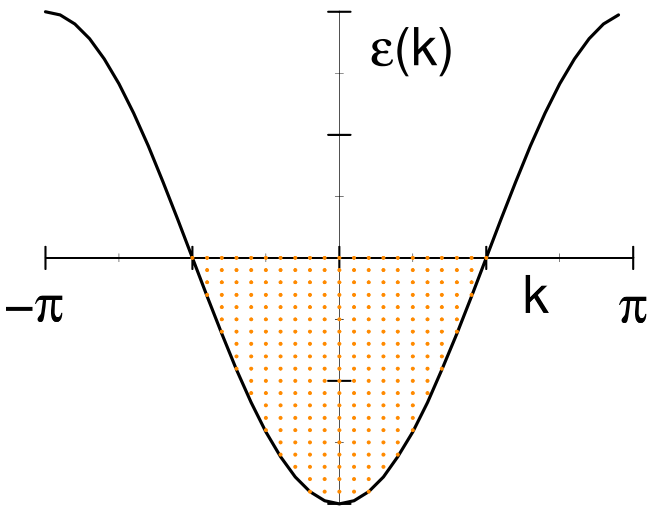
$$B\sum_n S^z = B\sum_n \left(\rho_n - \frac{1}{2}\right)$$

* unitary transf.: $J_{xy} \rightarrow -J_{xy}$
* dispersion relation $\epsilon_k = -J_{xy}\cos(k)$.

1D Fermions

$$H = \sum_k (\epsilon_k + B)\, c_k^\dagger c_k + H_{int}$$

* interaction $H_{int} \sim J_z$



* $B = 0$: half-filled
* $k_f = \pi/2$

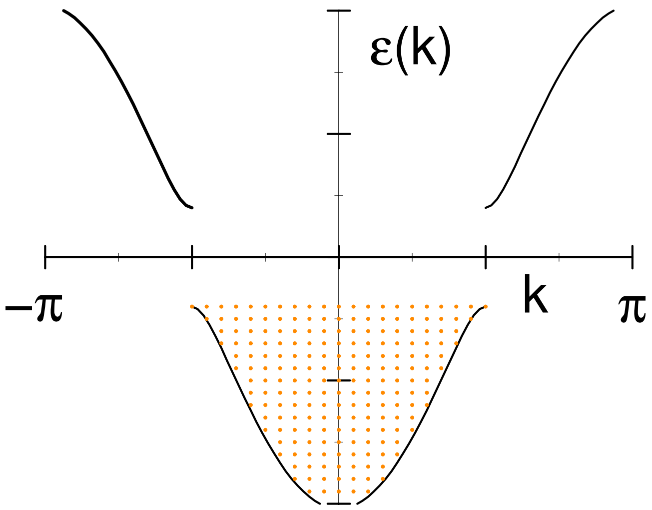
spin-Peierls phase

[static]

* dimerization of exchange term

$$\delta J \sum_n (-1)^n \, \mathbf{S}_n \cdot \mathbf{S}_{n+1} = 2i\delta J \sum_k \sin(k)\, c^\dagger_{k+\pi} c_k + H'_{int}$$

* gap Δ: $\quad \Delta/J \sim \begin{cases} \delta & (J_z = 0) \\ \delta^{2/3} & (J_z = J_{xy}) \end{cases}$



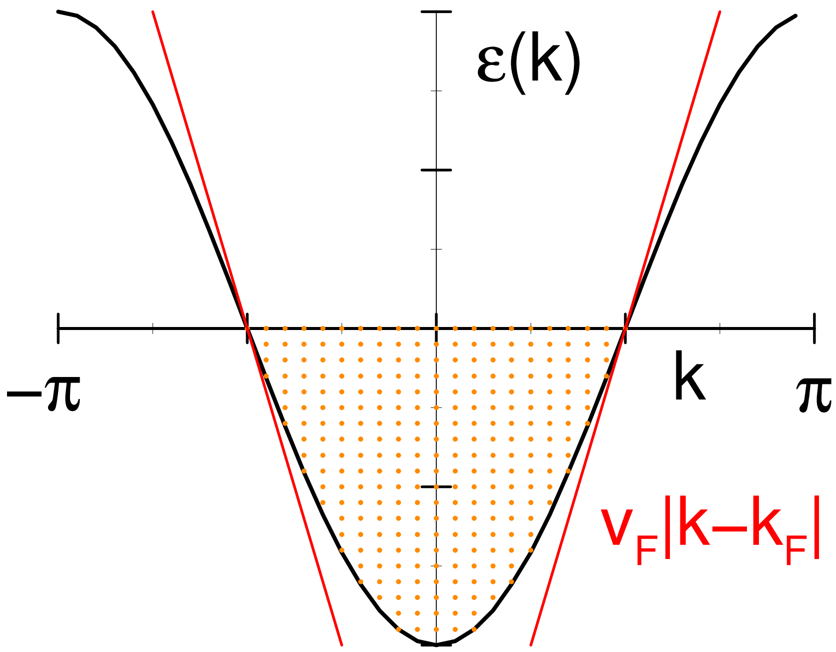
* $|B| < \Delta$: commensurable: $2k_f = \pi$
* $|B| > \Delta$: incommensurable: $2k_f < \pi$

$\rightarrow$ $\boxed{D \rightarrow I \text{ transition in } CuGeO_3}$

bosonization

$[\delta = 0]$

* linearization: $\epsilon(k) \approx v_F |k - k_F|$
* (bare) Fermi velocity: $v_F = J_{xy}$



* density operators: $\rho(q) = \sum_k c^\dagger_{k+q} c_k$
* commutation rel.: $[\rho(-q), \rho(q\prime)] = \frac{q}{2\pi}\delta_{q,q'}$

  $\rightarrow$ bosons for small q: $\boxed{b^\dagger_q = \sqrt{\frac{2\pi}{q}}\rho(q)}$

* Hamiltonian bilinear: $H \sim b^\dagger_q b_q, \; b_q b_{-q}$

  $\rightarrow$ solution: Bogoliubov transf.

  $\rightarrow$ $\boxed{\text{correlation functions}}$

spin-spin dynamical structure factor

$[J_z = J_{xy}]$

$$S^{(AF)}(q,\omega) = \sum_n \int d\omega\, \mathrm{e}^{i(\omega t - qn)} \langle \mathbf{S}_n(t) \cdot \mathbf{S}_0(0) \rangle$$

* bosonization

[Cross and Fischer, PRB '79]

$$\boxed{T\, S^{(AF)}(q,\omega) = -2d\, I_1\left(\frac{\omega - \Delta_q}{2\pi T}\right) I_1\left(\frac{\omega + \Delta_q}{2\pi T}\right)}$$

* $d \approx 0.37, \quad \Delta_q = v_F |q - \pi|$
* $I_1(k) = \frac{1}{2\pi} \int_0^\infty dx\, \mathrm{e}^{ikx} (\sinh x)^{-1/2}$

quantum criticality

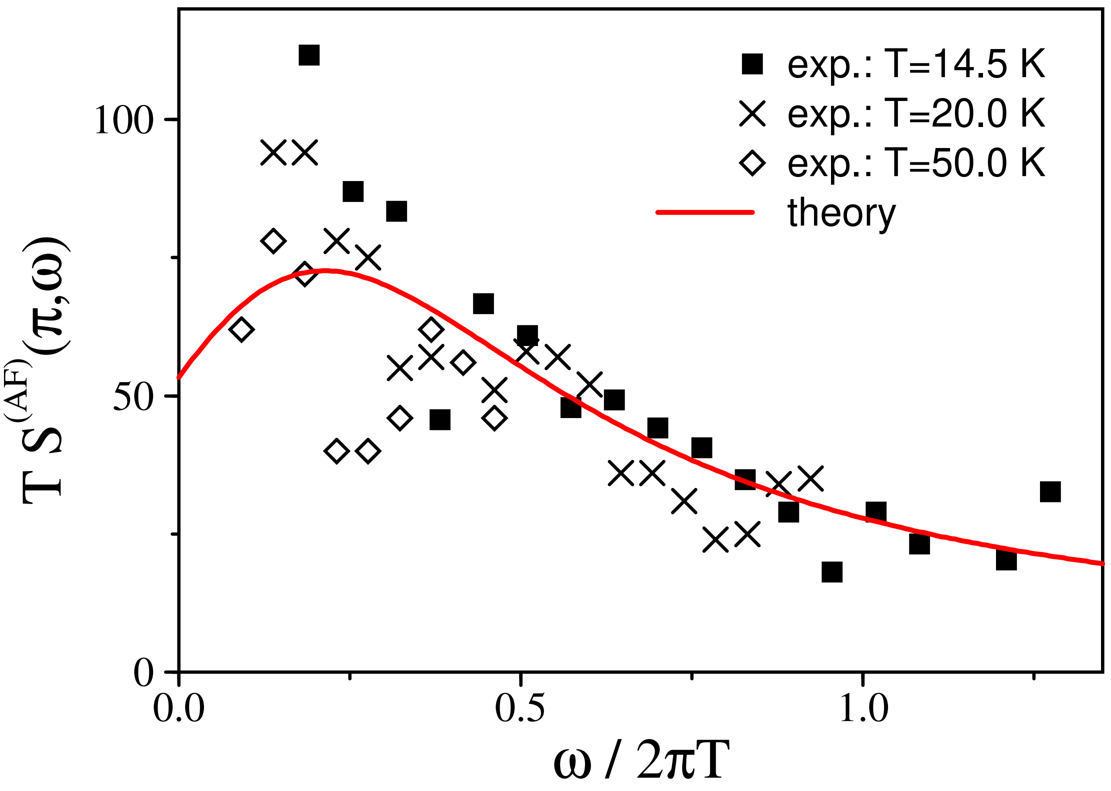
* $q = \pi, \quad \Delta_q = v_F |\pi - \pi| = 0$

$$\boxed{T\, S^{(AF)}(\pi,\omega) = -2d\, I_1^2\, (\omega/2\pi T)}$$

* independent of $v_F = \frac{\pi}{2} J$!

CuGeO$_3$: test of quantum criticality

[exp.: Pouget *et al.*, PRL '94]

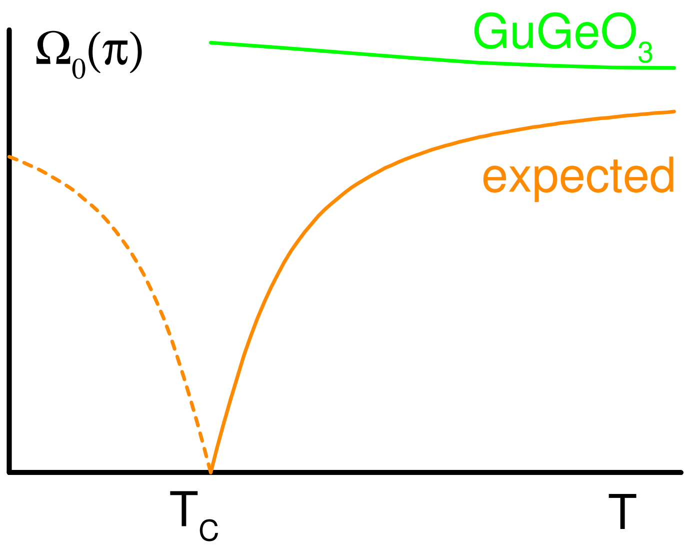


spin-Peierls transition

* 1D spin chains + magnetoelastic coupling
* four Peierls-aktive phonon modes: T_2^+
  [Braden *et al.*, PRL '98]
* $\Omega_0(\pi) = 3.2,\ 6.8,\ 11.1,\ 24.6\,\mathrm{THz}$
  $\approx\ J,\ 2J$
* $T < T_{SP}$: T_2^+ macroscopically occupied

the soft-phonon problem

$$\langle Q_q \rangle \sim \frac{1}{\sqrt{N}} \langle a^\dagger_{-q} + a_q \rangle$$



spin-phonon coupling

$$H_{ph} = \sum_q \Omega_0(q) \left(a_q^\dagger a_q + \frac{1}{2} \right)$$

$$H_{sp} = \sum_q \frac{g_q}{\sqrt{N}} \left(a_{-q}^\dagger + a_q \right) \sum_n e^{iqn} \, \mathbf{S}_n \cdot \mathbf{S}_{n+1}$$

* phonon creation/destruction: a_q, $a_q^\dagger$
* Jordan-Wigner $\to$ diagrams

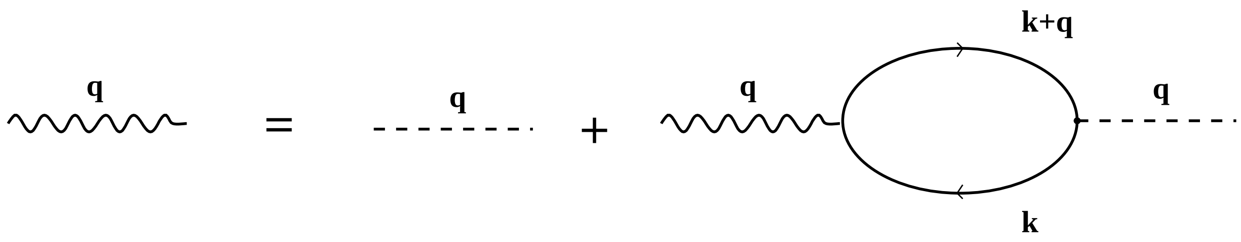
dynamics of the phonons

* unperturbed phonon propagator

$$D_q^{(0)}(\omega) = \frac{2\Omega_0(q)}{\omega^2 - \Omega_0^2(q)}$$

* RPA - approximation

$$\mathcal{D}_q(\omega) = \frac{D_q^{(0)}(\omega)}{1 - D_q^{(0)}(\omega) |g_q|^2 \chi_q(\omega)}$$



* $J_z = 0$: $\chi_q(\omega) = \frac{1}{N}\sum_k \frac{n_F(k)-n_F(k+q)}{\omega-(\epsilon_{k+1}-\epsilon_k)}$

* $J_z = J_{xy}$: $\chi_q(\omega) \rightarrow$ bosonization result

  [Cross and Fisher, PRB '79]

* $q = \pi \rightarrow$ quantum critical

$$\boxed{T\chi_q(\omega) = -0.74\, I_1^2 \left(\frac{\omega}{2\pi T}\right)}$$

collective modes

[zeros of the denominator]

* $1 - D_q^{(0)}(\omega)|g_q|^2\,\chi_q(\omega) = 0$

$$\frac{\omega^2 - \Omega_0^2}{2\Omega_0 g_\pi^2} = Re\chi_\pi(\omega) \approx \frac{-0.26}{T} + \frac{2.2}{T}\left(\frac{\omega}{2\pi T}\right)^2$$

* T_{SP}: $\mathcal{D}_\pi(\omega = 0) \rightarrow \infty$

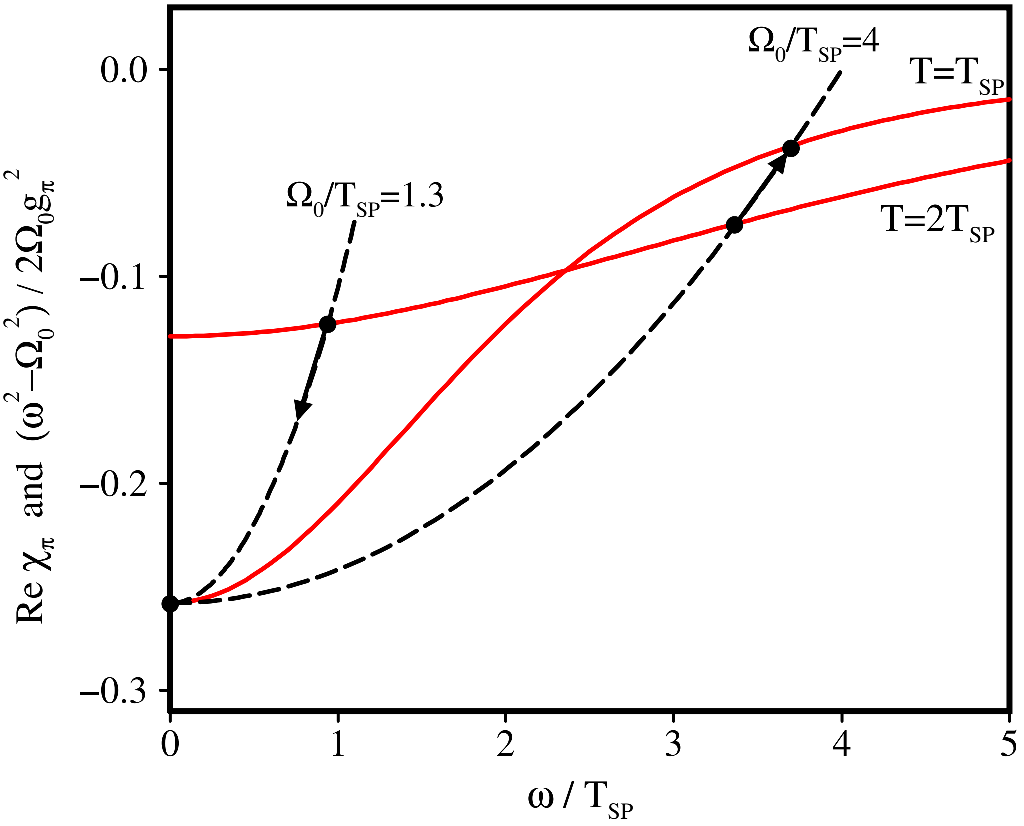
$$\boxed{T_{SP} = \frac{0.56\, g_\pi^2}{\Omega_0}}$$

* T_{SP} independent of J !

T-dependence of phonon modes

$\rightarrow \Omega_T(q)$

* graphic solution: $\frac{\omega^2-\Omega_0^2}{2\Omega_0 g_\pi^2} = Re\chi_\pi(\omega)$



* 'soft-phonon' regime: $\boxed{\Omega_0(\pi) < 2.2\,T_{SP}}$

$\rightarrow \Omega_T(\pi) \sim \sqrt{T - T_{SP}}$

* 'central-peak' regime: $\boxed{\Omega_0(\pi) > 2.2\,T_{SP}}$

$\rightarrow \Omega_T(\pi) \sim \Omega_0(\pi)$

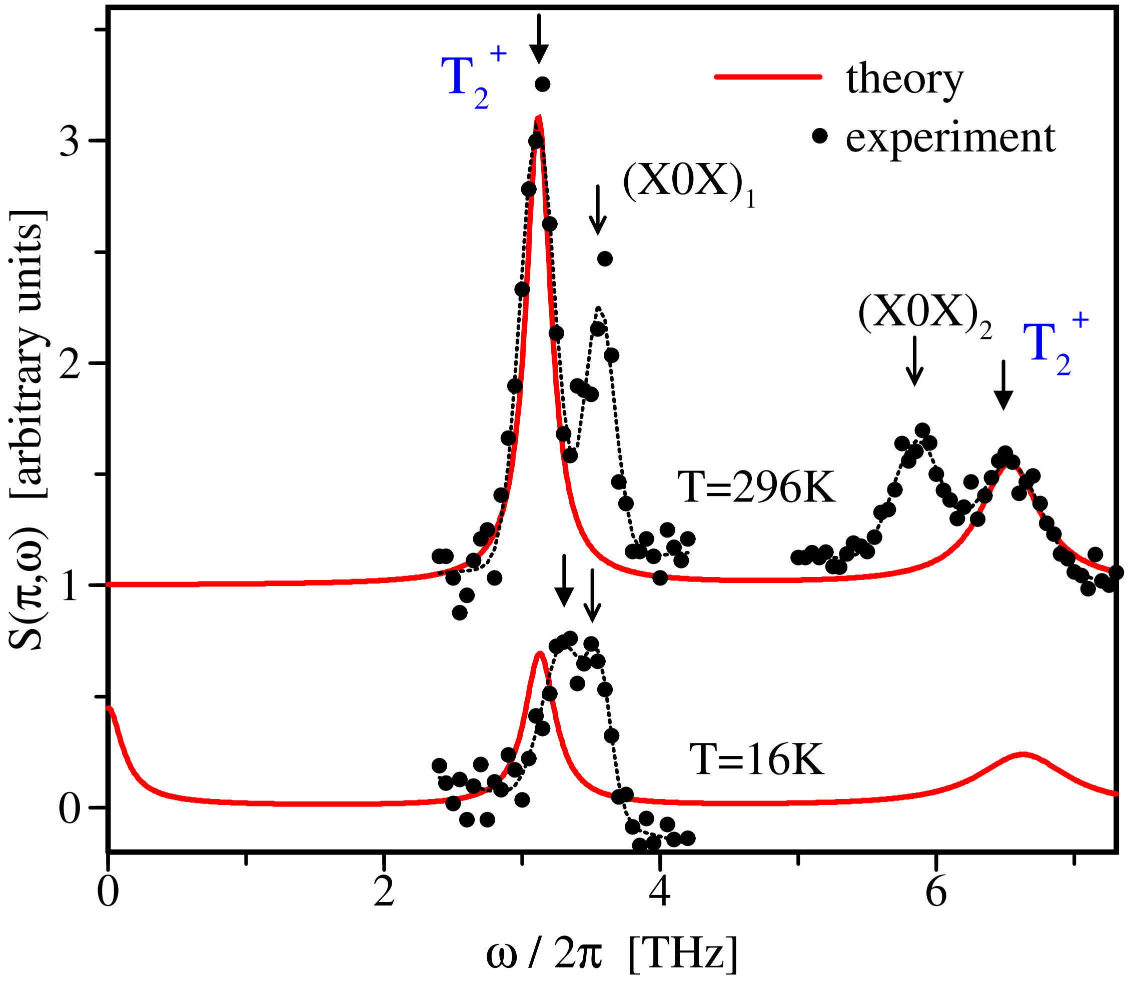
several phonon modes

$$T_{SP} = \frac{0.56\, g_1^2}{\Omega_1} + \frac{0.56\, g_2^2}{\Omega_2} + \dots$$

$*$ $CuGeO_3$: ‘central-peak’ regime

$$T_{SP} = 14.1\,\mathrm{K},\ \ \Omega_1 = 154\,\mathrm{K},\ \ \Omega_2 = 326\,\mathrm{K},\ \ \dots$$

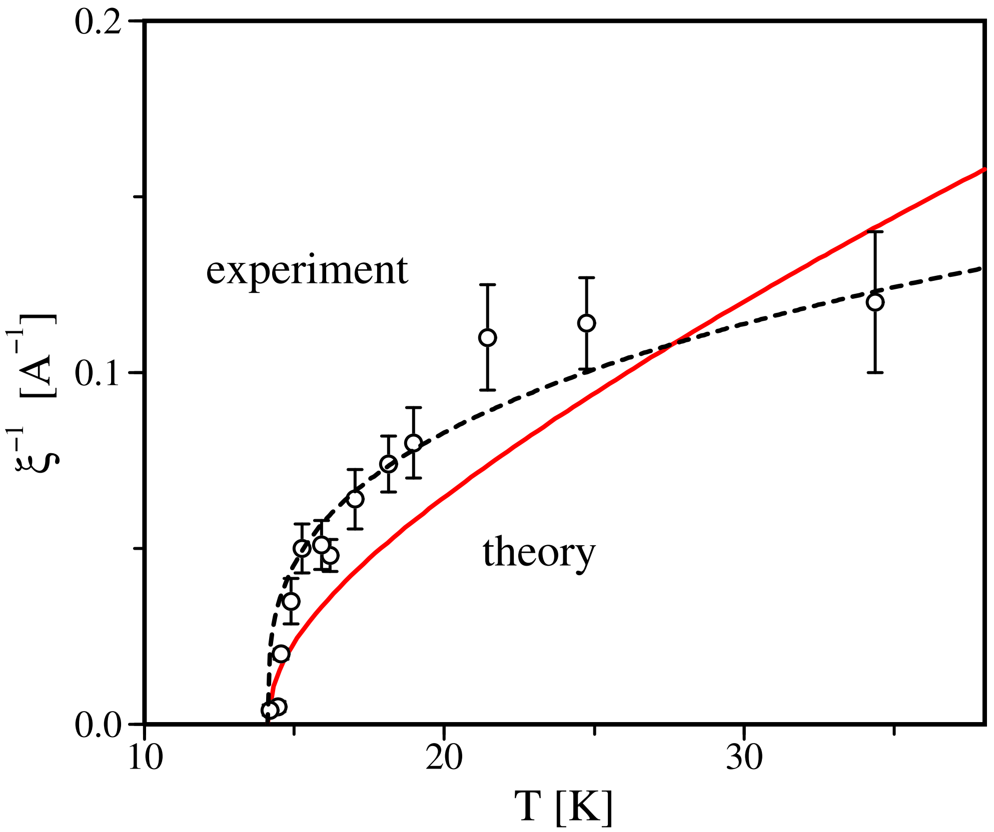
[Gros and Werner ‘98]



correlation length ξ

$$\lim_{z\to\infty}\int\frac{dq}{2\pi}\mathrm{e}^{iqz}Re\mathcal{D}_q(0)\;\sim\;\mathrm{e}^{i\pi z/c}\mathrm{e}^{-z/\xi},$$

* no further free parameters



determination of the coupling constants g_λ

* polarization-vectors $(\lambda = 1, \ldots, 4)$

[Braden *et al.*, PRL '98]

$$\vec{e}_\nu(\lambda), \;\; \nu = \mathrm{Cu, O(1), O(2), Ge}$$

* ionic displacements for $T < T_{SP}$ (RPA)

$$\boxed{\langle \vec{r}_\nu \rangle = \mathrm{const.} \sum_\lambda \frac{\vec{e}_\nu(\lambda)}{\sqrt{\hbar m_\nu}} \left(\Omega_\lambda\right)^{-3/2} g_\lambda}$$

* for $T < T_{SP}$ only (Exp.)

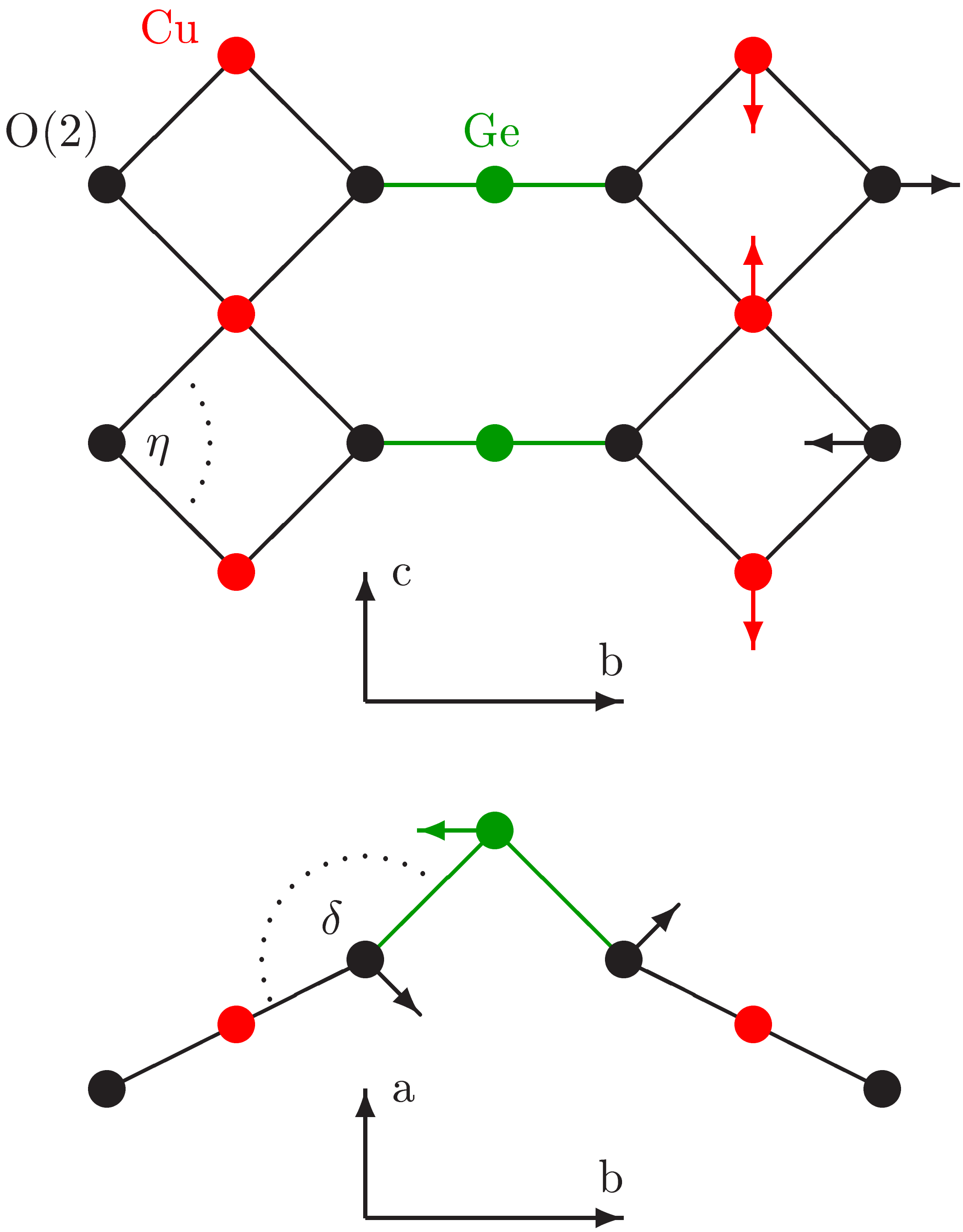
$$\langle r^z_{Cu} \rangle, \quad \langle r^x_{O2} \rangle, \quad \langle r^y_{O2} \rangle, \quad \langle r^y_{Ge} \rangle \quad \neq \quad 0$$

- 4×4 matrix $\rightarrow g_\lambda$
- $T_{SP} = 0.56 \sum_\lambda g_\lambda^2/\Omega_\lambda \rightarrow$ const.

λ	1	2	3	4
Ω_λ [K]	154	326	548	1181
g_λ [K]	-15	58	-30	-12

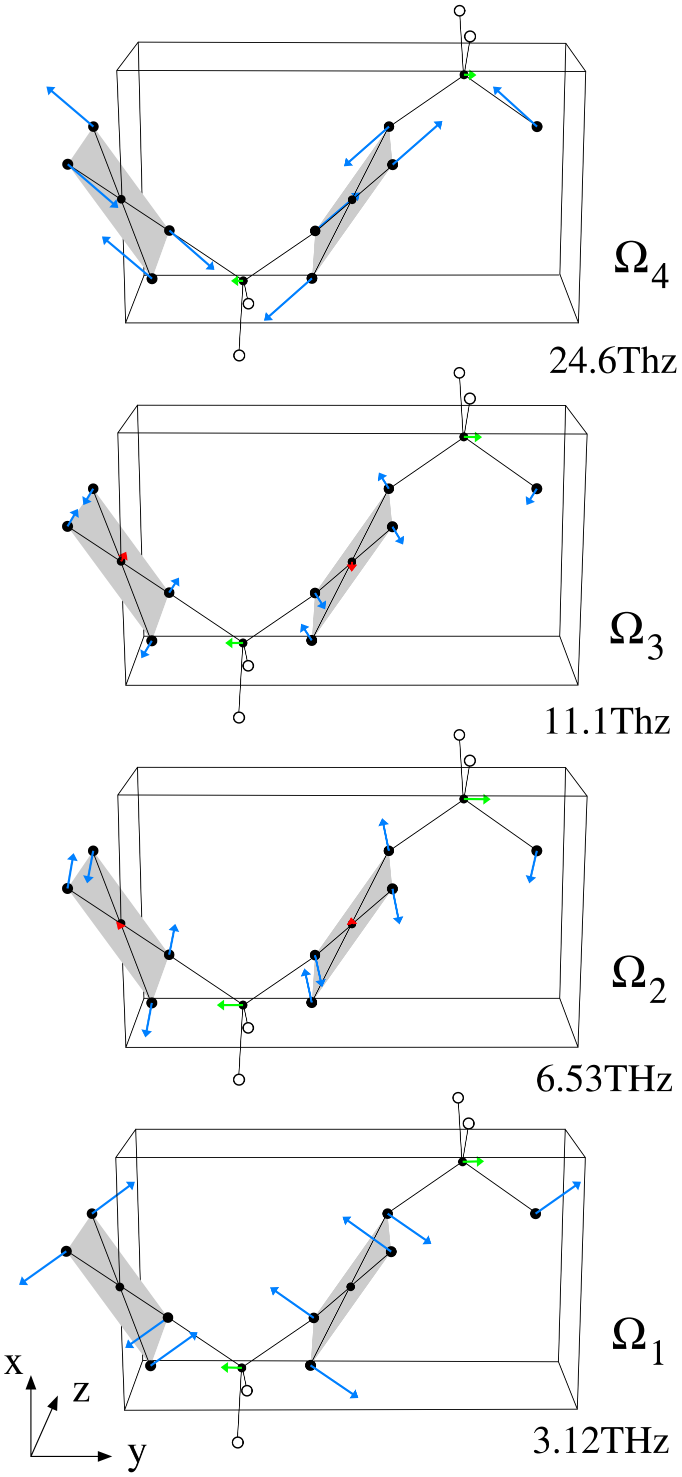
- $J = 156\,\mathrm{K}$

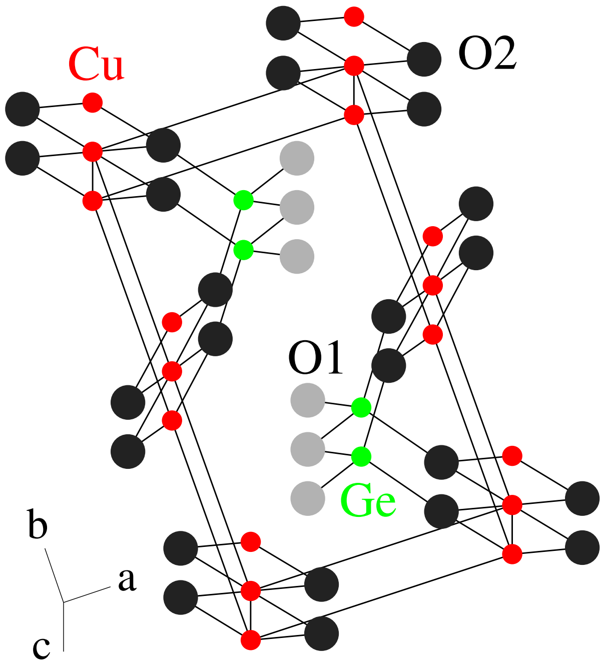
$CuGeO_3$



$CuGeO_3$

Eigen-modes





[Werner, Gros und Braden, '99]

magnetoelastic coupling

$$J\{\vec{r}_\nu\} = J + \sum_\nu \frac{\partial J}{\partial \vec{r}_\nu} \cdot \vec{r}_\nu$$

$*$ four parameters $(r^z_{Cu},\ r^y_{Ge},\ r^x_{O2},\ r^y_{O2})$

$$\begin{aligned} &\eta, \quad d_{Cu} = |\vec{r}_{O2} - \vec{r}_{Cu}|, \\ &\delta, \quad d_{Ge} = |\vec{r}_{O2} - \vec{r}_{Ge}| \end{aligned}$$

$*$ $(g_1, g_2, g_3, g_4) \leftrightarrow (g_\eta, g_\delta, g^d_{Cu}, g^d_{Ge})$

$$\boxed{\begin{array}{ccccccc} g_\eta & = & 15 & & g_\delta & = & 1.5 \\ g^d_{Cu} & = & 180 & & g^d_{Ge} & = & -96 \end{array}} \quad \begin{array}{l} [K/deg] \\ [K/\mathring{A}] \end{array}$$

$*$ pressure-dependence $(\vec{r}_\nu)$

[Bräuniger *et al.*, PRB '97]

- $-\frac{\partial \ln J}{\partial \rho} \approx 8.1\%$ per GPa
- exp. (susceptibility) $\rightarrow 5 - 11\%$

[Büchner *et al.*, PRL '96]

[Nishi *et al.*, PRB '95]

$*$ $J\delta \approx 24\,\mathrm{K}$

outlook – one-dimensional systems

* $CuGeO_3$ – magnetic excitations
  - Raman scattering
  - line shape of the ‘gap’-excitation

* $CuGeO_3$ – magnetoelastic coupling
  - ‘soft-phonon’ problem solved
  - microscopic coupling constants
  - transport: thermal conductivity

* NaV_2O_5: spin-Peierls?
  - 1/4-filled ladder compound

    [Smolinski *et al.*, PRL ‘98]
  - charge ordering, . . .

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