Workshop on correlated electronic structure and spin dynamics
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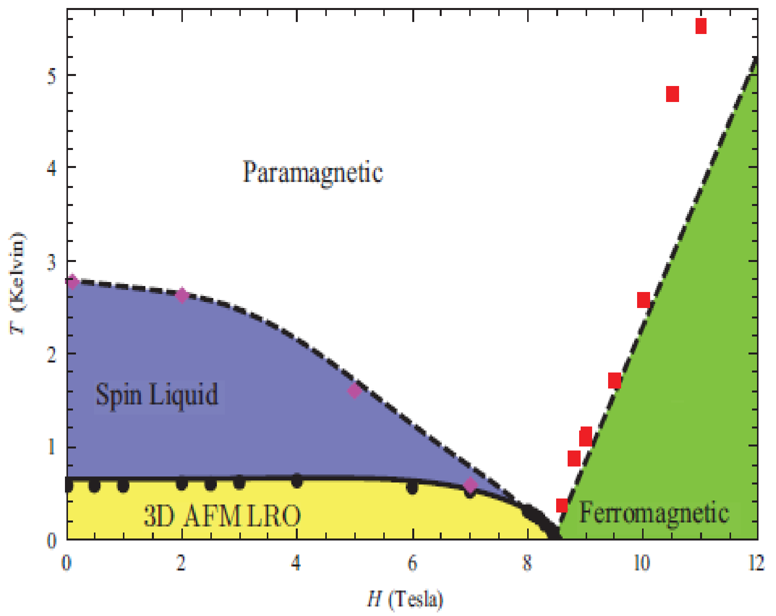
with A. Kreisel, S. Streib, T. Herfurth, M. Peter (theory)
B. Wolf, M. Lang (experiment)

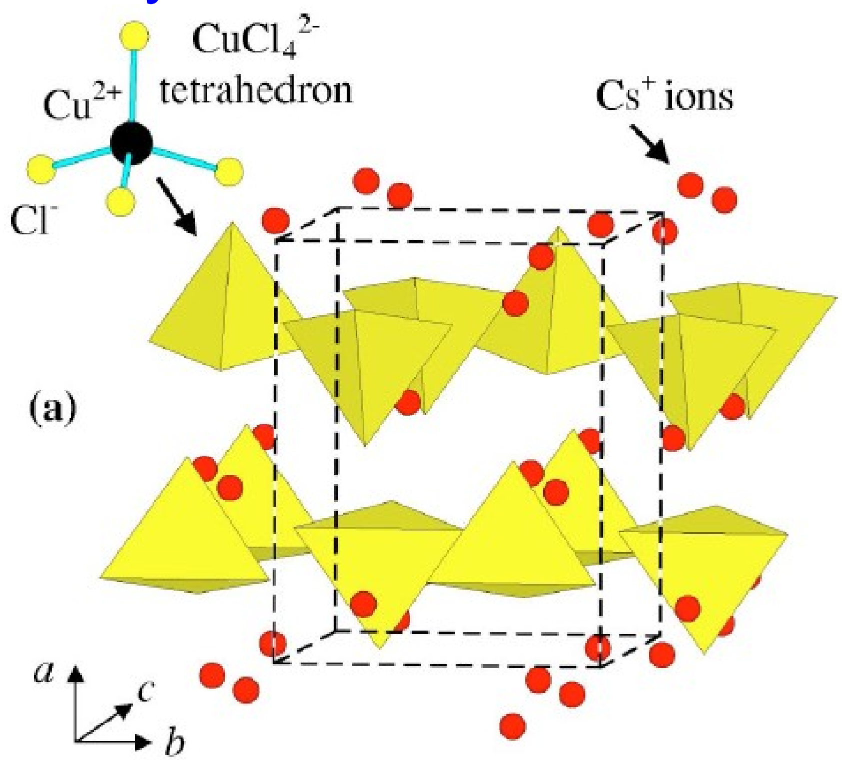
Magnetic excitations and ultrasound in the triangular lattice antiferromagnet Cs_2CuCl_4

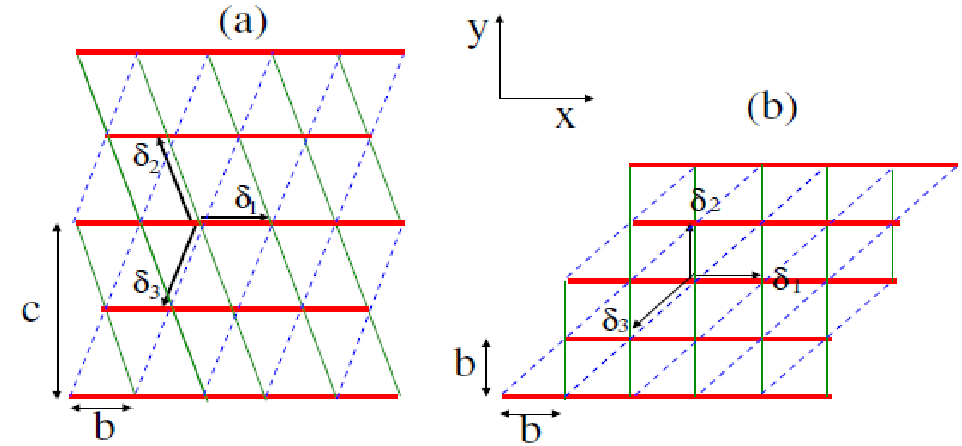
1. Ordered phase (cone-state):
   Spin-wave theory and ultrasound
2. Ultrasound in spin liquid phase
3. Vicinity of quantum critical point



Spin model for Cs_2CuCl_4

Crystal structure (from Coldea et al, 2003)





$$J_{ij} \equiv J(\boldsymbol{R}_i - \boldsymbol{R}_j) \qquad J_1 = J = 4.34\,\mathrm{K}$$

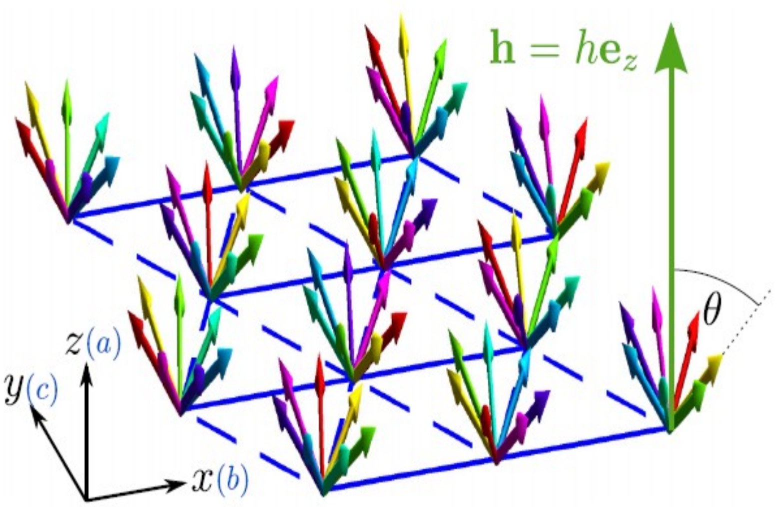
$$J_2 = J_3 = J' = 1.49\,\mathrm{K}$$

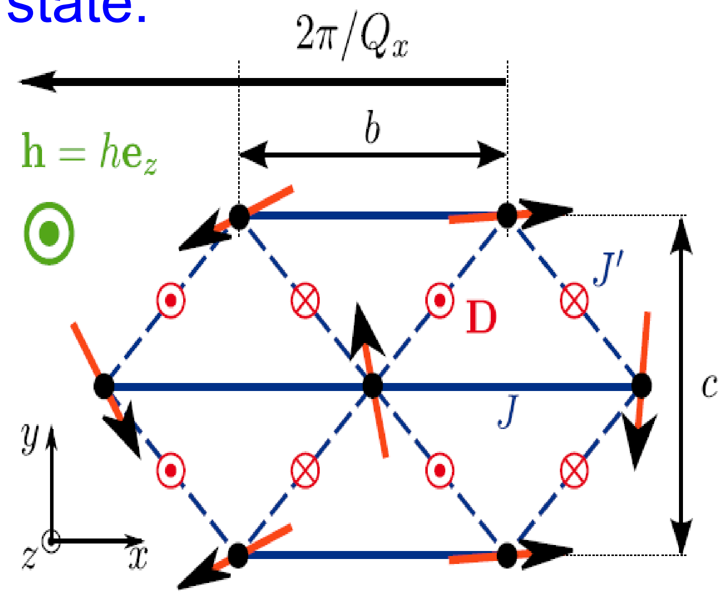
$$\mathcal{H} = \frac{1}{2}\sum_{ij}\left[J_{ij}\boldsymbol{S}_i \cdot \boldsymbol{S}_j + \boldsymbol{D}_{ij}\cdot(\boldsymbol{S}_i \times \boldsymbol{S}_j)\right] - h\sum_i S_i^z,$$

$$\boldsymbol{D}_{ij} = D_{ij}\hat{\boldsymbol{z}} \qquad D(\pm\boldsymbol{\delta}_2) = D(\pm\boldsymbol{\delta}_3) = \mp D \qquad D = 0.23\mathrm{K}$$

1. Spin-waves and ultrasound in the cone-state

spin configuration in classical ground state:





$$\hat{\boldsymbol{m}}_i = \sin\,\theta[\cos(\boldsymbol{Q}\cdot\boldsymbol{R}_i)\hat{\boldsymbol{x}} + \sin(\boldsymbol{Q}\cdot\boldsymbol{R}_i)\hat{\boldsymbol{y}}] + \cos\,\theta\hat{\boldsymbol{z}}$$

classical ground stat energy:

$$\mathcal{H}_0 = \frac{S^2}{2}\sum_{ij} J^{\parallel}_{ij} - S\sum_i \boldsymbol{h}\cdot\hat{\boldsymbol{m}}_i$$

$$J^{\parallel}_{ij} = J_{ij}\hat{\boldsymbol{m}}_i\cdot\hat{\boldsymbol{m}}_j + \boldsymbol{D}_{ij}\cdot(\hat{\boldsymbol{m}}_i\times\hat{\boldsymbol{m}}_j)$$

$$\cos\,\theta = h/h_c,$$

$$\cos\left(\frac{Q_x b}{2}\right) = -\frac{J'}{2J} - \frac{D}{2J}\cot\left(\frac{Q_x b}{2}\right)$$

$$h_c = S\big(J^D_0 - J^D_Q\big) = S(J_0 - J_Q + i\,D_Q)$$

spin-wave dispersion

Holstein-Primakoff-transformation:

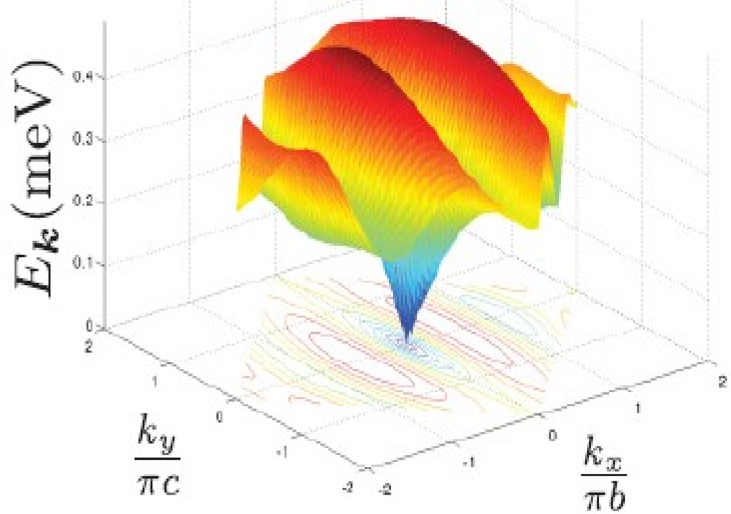
$$S_i^{\parallel} = S - n_i,$$

$$S_i^{+} = \sqrt{2S}\sqrt{1-\frac{n_i}{2S}}b_i$$

$$S_i^{-} = \sqrt{2S}b_i^{\dagger}\sqrt{1-\frac{n_i}{2S}}$$

$$\mathcal{H} = \mathcal{H}_0 + \sum_{n=2}^{\infty}\mathcal{H}_n \qquad \mathcal{H}_n \propto S^{2-n/2}$$

$h = 0.1h_c$



Spin-wave dispersion:

$$\mathcal{H}_2 = \sum_k \left\{ A_k b_k^{\dagger} b_k - \frac{B_k}{2}[b_k^{\dagger} b_{-k}^{\dagger} + b_{-k} b_k] \right\}$$

$$\mathcal{H}_2 = \sum_k \left[E_k \beta_k^{\dagger}\beta_k + \frac{\epsilon_k - A_k^{+}}{2} \right]$$

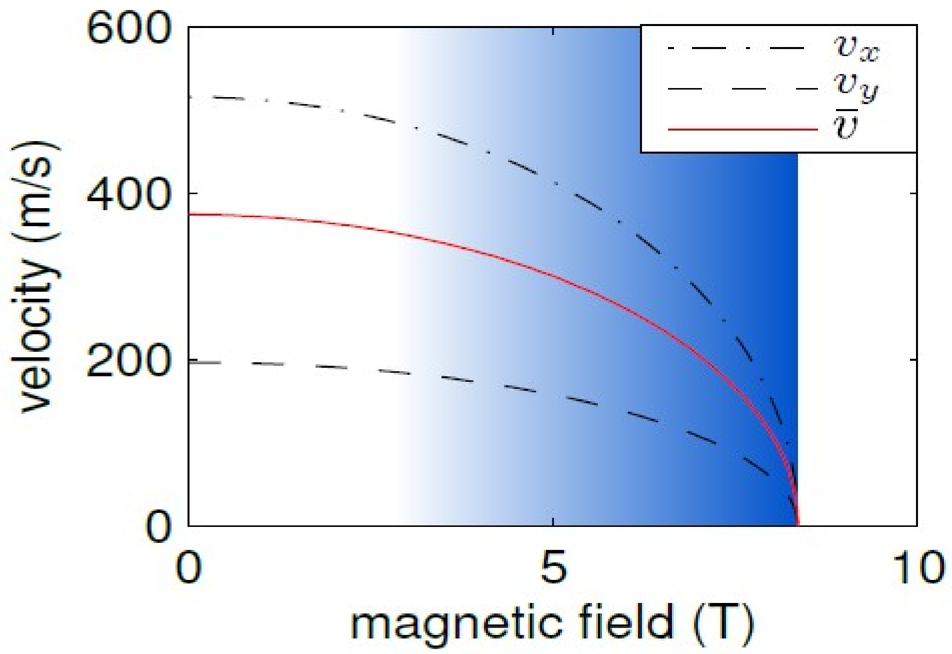
$$E_k = \epsilon_k + A_k^{-} \qquad A_k = A_k^{+} + A_k^{-}$$

$$\epsilon_k = \sqrt{(A_k^{+})^2 - B_k^2}.$$

Goldstone mode and spin-wave velocities

$$E_{\boldsymbol{k}} = v(\hat{\boldsymbol{k}})|\boldsymbol{k}| + \mathcal{O}(\boldsymbol{k}^3)$$

$$v(\hat{\boldsymbol{k}}) = \sqrt{v_x^2 \hat{k}_x^2 + v_y^2 \hat{k}_y^2},$$



How about interactions between spin-waves?

Infrared divergence due to spin-wave interactions

(A. Kreisel, M. Peter, P.K. PRB 2014)

- use real-field parametrization of Holstein-Primakoff bosons:

$$b_{\boldsymbol{k}} = \frac{1}{\sqrt{2}}[\Phi_{\boldsymbol{k}} + i\Pi_{\boldsymbol{k}}] \, , \quad b^{\dagger}_{\boldsymbol{k}} = \frac{1}{\sqrt{2}}[\Phi_{-\boldsymbol{k}} - i\Pi_{-\boldsymbol{k}}] \qquad [\Phi_{\boldsymbol{k}}, \Pi_{\boldsymbol{k}}] = i\delta_{\boldsymbol{k},-\boldsymbol{k}'}$$

$$\mathcal{H}_3 = \frac{1}{\sqrt{N}} \sum_{\boldsymbol{k}_1\boldsymbol{k}_2\boldsymbol{k}_3} \delta_{\boldsymbol{k}_1+\boldsymbol{k}_2+\boldsymbol{k}_3,0} \Big[\frac{1}{3!}\Gamma^{\Phi\Phi\Phi}(\boldsymbol{k}_1,\boldsymbol{k}_2,\boldsymbol{k}_3)\Phi_{\boldsymbol{k}_1}\Phi_{\boldsymbol{k}_2}\Phi_{\boldsymbol{k}_3} + \frac{1}{3!}\Gamma^{\Pi\Pi\Pi}(\boldsymbol{k}_1,\boldsymbol{k}_2,\boldsymbol{k}_3)\Pi_{\boldsymbol{k}_1}\Pi_{\boldsymbol{k}_2}\Pi_{\boldsymbol{k}_3}$$
$$+ \frac{1}{2!}\Gamma^{\Phi\Phi\Pi}(\boldsymbol{k}_1,\boldsymbol{k}_2;\boldsymbol{k}_3)\Phi_{\boldsymbol{k}_1}\Phi_{\boldsymbol{k}_2}\Pi_{\boldsymbol{k}_3} + \frac{1}{2!}\Gamma^{\Pi\Pi\Phi}(\boldsymbol{k}_1,\boldsymbol{k}_2;\boldsymbol{k}_3)\Pi_{\boldsymbol{k}_1}\Pi_{\boldsymbol{k}_2}\Phi_{\boldsymbol{k}_3} \Big]$$

$$\Gamma^{\Phi\Phi\Pi}(\boldsymbol{k}_1,\boldsymbol{k}_2;\boldsymbol{k}_3) \approx \sin\theta\cos\theta\frac{h_c}{\sqrt{S}}.$$

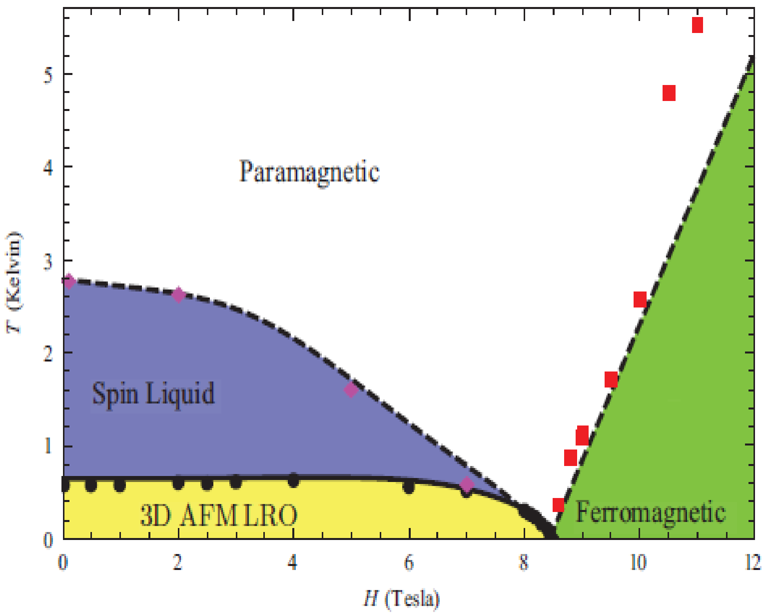
- coupling between transverse and longitudinal fluctuations leads to infrared divergencies even at T=0

- mapping to condensed phase of interacting Bose gas

- singular scattering continuum in longitudinal spin structure factor

Ultrasound experiments in Cs_2CuCl_4

- measurement of velocity and damping of acoustic phonons
- indirect probe of spin excitations (spin-lattice interactions)
- need different methods in different regimes:



- magnetically ordered: spin-wave theory
  (Kreisel, PK, Cong, Wolf, Lang, PRB 2011)

- spin liquid: Majorana-mean-field theory,
  Jordan-Wigner fermions
  (Herfurth et al PRB 2013; Streib et al, PRB 2015)

- quantum critical: hard-core bosons
  functional RG
  (Streib et al, in preparation)

Spin-phonon coupling from magneto-striction

$$H_{\rm spin}^{\rm pho} = \frac{1}{2}\sum_{ij}\left[J_{ij}\boldsymbol{S}_i\cdot\boldsymbol{S}_j + \boldsymbol{D}_{ij}\cdot(\boldsymbol{S}_i\times\boldsymbol{S}_j)\right] - \sum_i \boldsymbol{h}\cdot\boldsymbol{S}_i$$

exchange couplings depend on phonon coordinates:

$$J_{ij} = J(\boldsymbol{R}_{ij}) + (\boldsymbol{X}_{ij}\cdot\nabla_{\boldsymbol{r}})\, J(\boldsymbol{r})\big|_{\boldsymbol{r}=\boldsymbol{R}_{ij}} + \frac{1}{2}(\boldsymbol{X}_{ij}\cdot\nabla_{\boldsymbol{r}})^2\, J(\boldsymbol{r})\big|_{\boldsymbol{r}=\boldsymbol{R}_{ij}} + \cdots$$

$$\boldsymbol{X}_{ij} = \boldsymbol{X}_i - \boldsymbol{X}_j \qquad \boldsymbol{X}_i = \frac{1}{\sqrt{N}}\sum_{\boldsymbol{k}} e^{i\boldsymbol{k}\cdot\boldsymbol{R}_i}\boldsymbol{X}_{\boldsymbol{k}},$$

$$\boldsymbol{X}_{\boldsymbol{k}} = \sum_\lambda X_{\boldsymbol{k}\lambda}\boldsymbol{e}_{\boldsymbol{k}\lambda} \qquad X_{\boldsymbol{k}\lambda} = \frac{1}{\sqrt{2M\omega_{\boldsymbol{k}\lambda}}}\left(a_{\boldsymbol{k}\lambda} + a^\dagger_{-\boldsymbol{k}\lambda}\right)$$

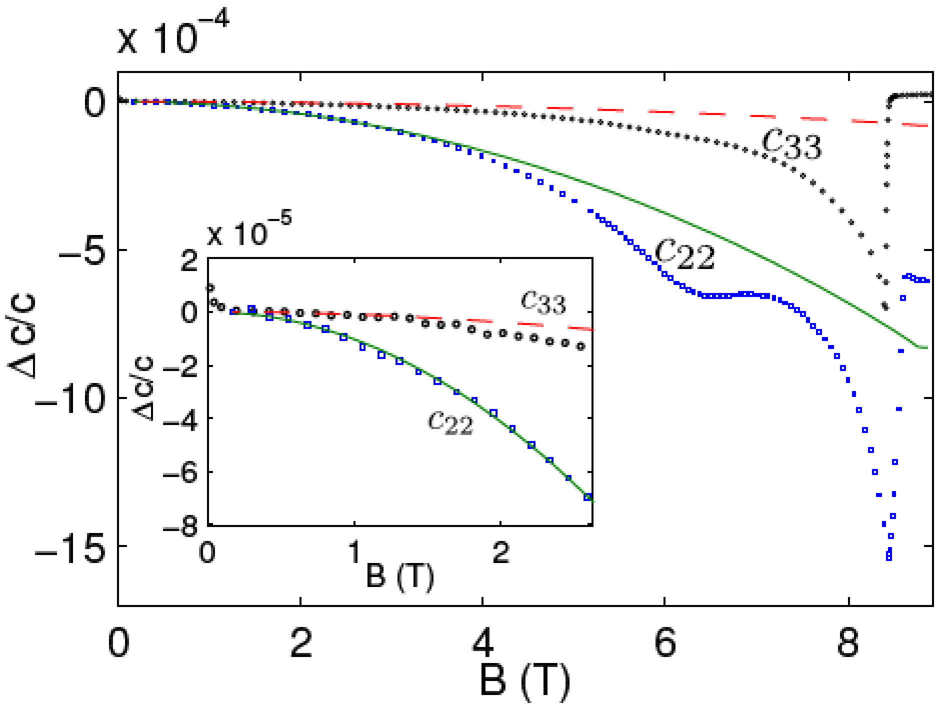
$$H^{\rm pho} = \sum_{\boldsymbol{k}\lambda}\omega_{\boldsymbol{k}\lambda}\left(a^\dagger_{\boldsymbol{k}\lambda}a_{\boldsymbol{k}\lambda} + \frac{1}{2}\right) \qquad \omega_{\boldsymbol{k}\lambda} = c_\lambda(\hat{\boldsymbol{k}})|\boldsymbol{k}|$$

renormalized sound velocities in cone-state

- for ultrasound: how is phonon propagator renormalized by magnons?

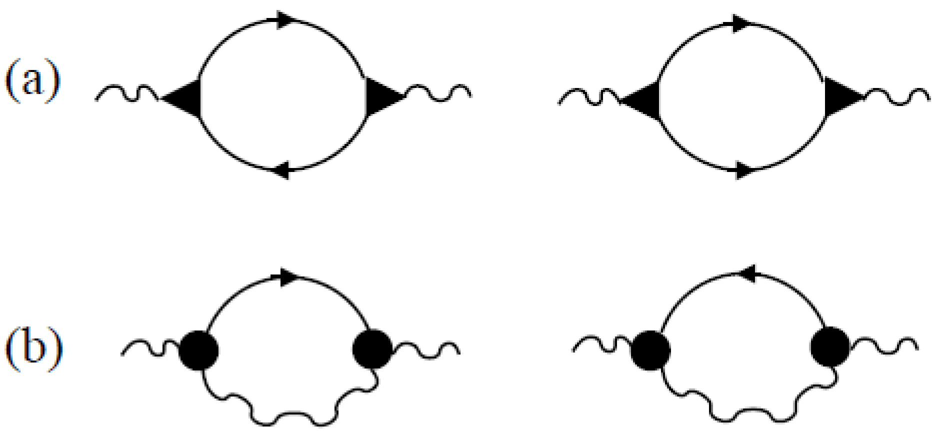
$$G^{\mathrm{pho}}(K\lambda) = \frac{M}{T}\langle X_{-K\lambda}X_{K\lambda}\rangle = \frac{1}{\omega^2 + \omega_{\boldsymbol{k}\lambda}^2 + \Sigma^{\mathrm{pho}}(K\lambda)}$$

- real part of phonon self-energy to second order in gradients gives shift in sound velocity → elastic constants



ultrasound attenuation in cone state

- imaginary part of phonon self-energy gives damping



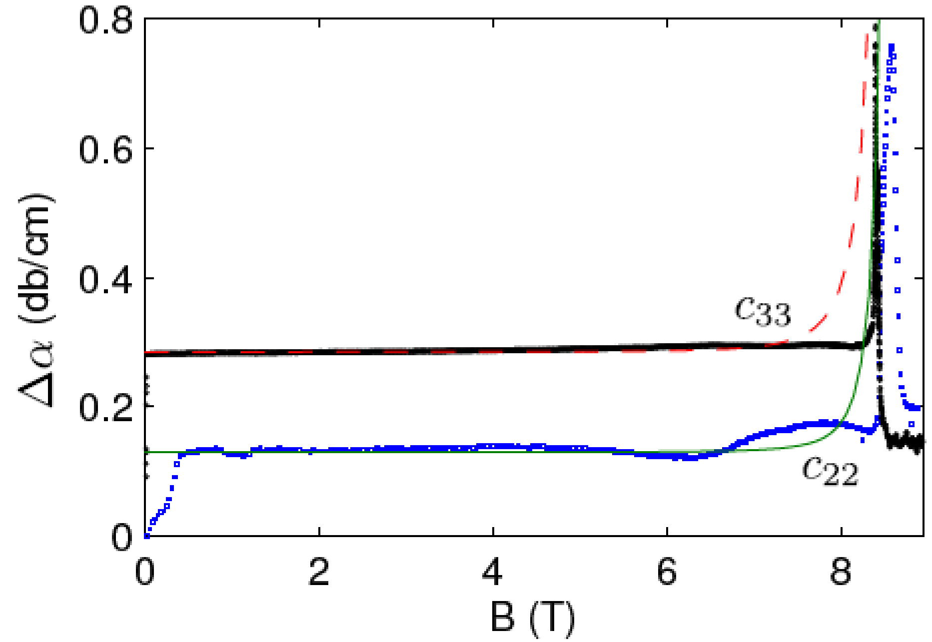


FIG. 7. (Color online) Experimental results for the relative ultrasonic attenuation $\Delta\alpha$ in Cs_2CuCl_4 of the longitudinal c_{22}-phonon mode (squares) and the c_{33}-mode (circles) taken at $T = 52$ mK (c_{22}-mode) and $T = 48$ mK (c_{33}-mode). The

2. Ultrasound in spin-liquid phase: dimensional reduction

- dimensional reduction (Balents, Nature 2010)
  spin liquid phase is quasi-one dimensional even for J'/J =1/3
- confirmed by Majorana-mean field theory
  (Herfurth, Streib, PK, PRB 2013)
- S=1/2 operators can be expressed in terms of 3 Majorana fermions

$$S_i^x = -i\eta_i^y \eta_i^z, \quad S_i^y = -i\eta_i^z \eta_i^x, \quad S_i^z = -i\eta_i^x \eta_i^y \qquad \eta_i^\alpha \eta_j^\beta + \eta_j^\beta \eta_i^\alpha = \delta_{ij}\delta^{\alpha\beta}$$

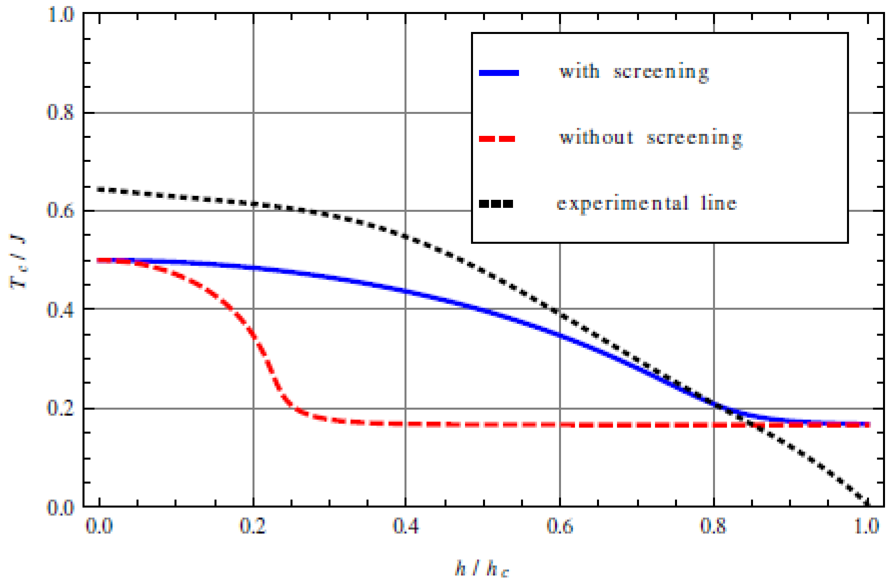
$$\mathcal{H} = \frac{1}{4}\sum_{ij}\sum_{\alpha\neq\beta} J_{ij} C_{ij}^\alpha C_{ij}^\beta + ih\sum_i \eta_i^x \eta_i^y \qquad C_{ij}^\alpha = \eta_i^\alpha \eta_j^\alpha$$

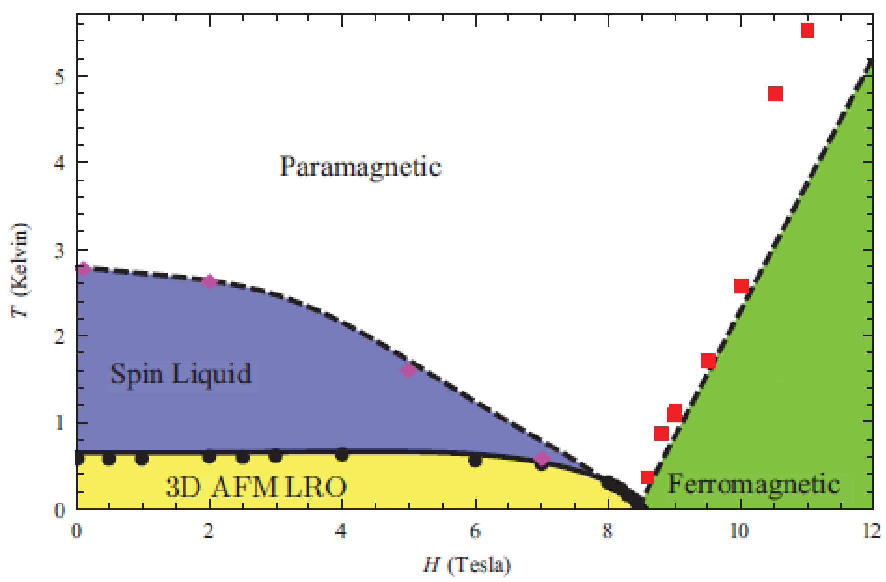
- Mean-field decoupling:

$$\boldsymbol{S}_i \cdot \boldsymbol{S}_j = \frac{1}{2}\sum_{\alpha\neq\beta} \eta_i^\alpha \eta_j^\alpha \eta_i^\beta \eta_j^\beta \to \frac{1}{2}\sum_{\alpha\neq\beta}\left[C_{ij}^\alpha \langle C_{ij}^\beta\rangle + \langle C_{ij}^\alpha\rangle C_{ij}^\beta - \langle C_{ij}^\alpha\rangle\langle C_{ij}^\beta\rangle\right]$$
$$- \left[\eta_i^x \eta_i^y \langle \eta_j^x \eta_j^y\rangle + \langle \eta_i^x \eta_i^y\rangle \eta_j^x \eta_j^y - \langle \eta_i^x \eta_i^y\rangle\langle \eta_j^x \eta_j^y\rangle\right]$$

Majorana MFT

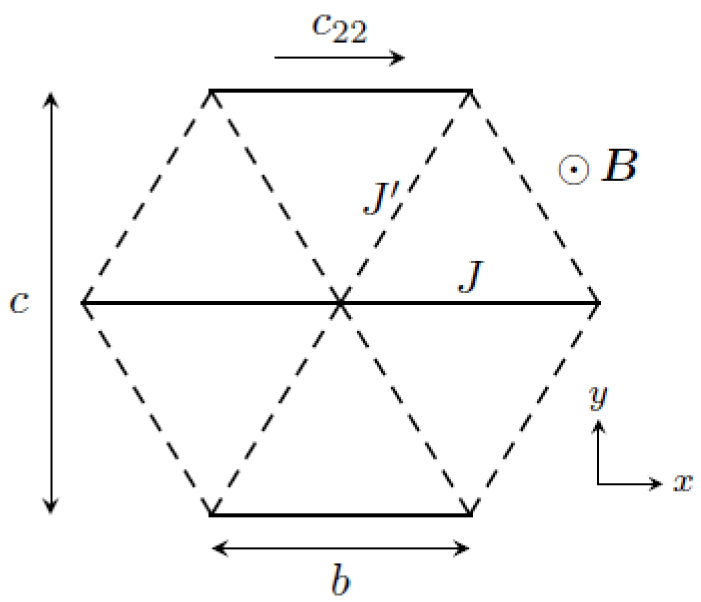
- in mean-field state Majorana fermions propagate only in 1d
- explains shape of crossover temperature spin-liquid to paramagnet





Jordan-Wigner mean-field theory for spin liquid phase

(Streib et al, PRB 2015)



- Hypothesis: since spin-liquid state is 1d, ultrasound experiments probing sound along b-axis can be explained within 1d Heisenberg chain + phonons

$$\mathcal{H} = \sum_n J_n \left[\boldsymbol{S}_n \cdot \boldsymbol{S}_{n+1} - 1/4 \right] - h \sum_n S_n^z + \mathcal{H}_2^p,$$

$$\mathcal{H}_2^p = \sum_q \left[\frac{P_{-q} P_q}{2M} + \frac{M}{2} \omega_q^2 X_{-q} X_q \right]$$

$$J_n \approx J + J^{(1)} (X_{n+1} - X_n) + \frac{J^{(2)}}{2} (X_{n+1} - X_n)^2$$

Jordan-Wigner transformation and mean-field decoupling

- Jordan-Wigner transformation: spins in terms of spinless fermions

$$S_n^+ = (S_n^-)^\dagger = c_n^\dagger (-1)^n e^{i\pi \sum_{j<n} c_j^\dagger c_j} \qquad S_n^z = c_n^\dagger c_n - 1/2$$

$$\mathcal{H} = -\frac{1}{2}\sum_n J_n (c_n^\dagger c_{n+1} + c_{n+1}^\dagger c_n + c_n^\dagger c_n + c_{n+1}^\dagger c_{n+1})$$
$$+ \sum_n J_n c_n^\dagger c_n c_{n+1}^\dagger c_{n+1} - h \sum_n c_n^\dagger c_n + Nh/2 + \mathcal{H}_2^p$$

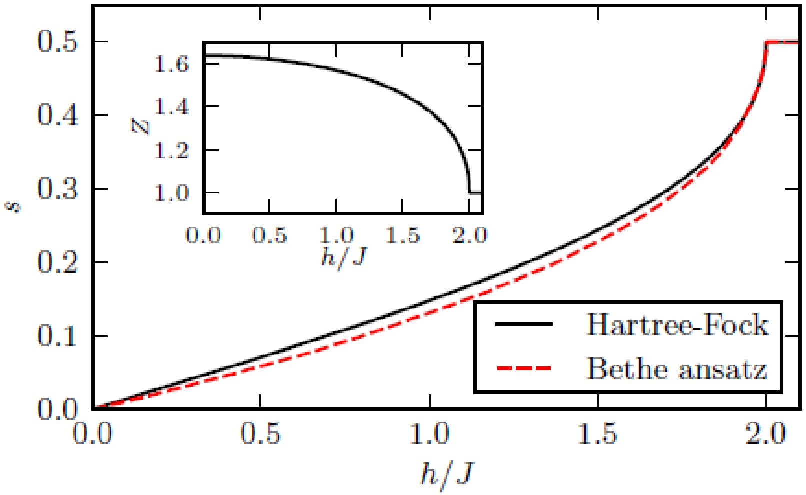
- mean-field decoupling of interaction:

$$c_n^\dagger c_n c_{n+1}^\dagger c_{n+1} \approx \rho(c_{n+1}^\dagger c_{n+1} + c_n^\dagger c_n) - \rho^2 - \tau(c_n^\dagger c_{n+1} + c_{n+1}^\dagger c_n) + \tau^2$$

$$\rho = \langle c_n^\dagger c_n \rangle \qquad \tau = \langle c_n^\dagger c_{n+1} \rangle$$

- good agreement with Bethe-Ansatz (Bulaevski 1962)

$$s = \rho - 1/2 \qquad Z = 1 + 2\tau$$



ultrasound in spin-liquid phase

- strategy: perturbation theory for fermion-phonon interaction

$$\mathcal{H} = F_0 + \sum_k \xi_k c_k^\dagger c_k + \mathcal{H}_2^p + \delta\mathcal{H}_2^p + \mathcal{H}_3^{sp} + \mathcal{H}_4^{sp}$$

$$\delta\mathcal{H}_2^p = 2J^{(2)}(\tau^2 - \rho^2) \sum_q \sin^2(q/2) X_{-q} X_q$$

$$\mathcal{H}_3^{sp} = \frac{1}{\sqrt{N}} \sum_{k'kq} \delta^*_{k',k+q} \Gamma_3(k,q) c_{k'}^\dagger c_k X_q$$

$$\mathcal{H}_4^{sp} = \frac{1}{2N} \sum_{k'kq_1q_2} \delta^*_{k',k+q_1+q_2} \Gamma_4(k,q_1,q_2) c_{k'}^\dagger c_k X_{q_1} X_{q_2}$$

- phonon propagator: $[\omega^2 + \omega_q^2 + \Pi(q, i\omega)]^{-1}$

$$\Pi_2(q) = [J^{(2)}/M][\tau^2 - \rho^2] 4\sin^2(q/2) \qquad \Pi_4(q) = \frac{1}{MN} \sum_k f_k \Gamma_4(k,q,-q)$$

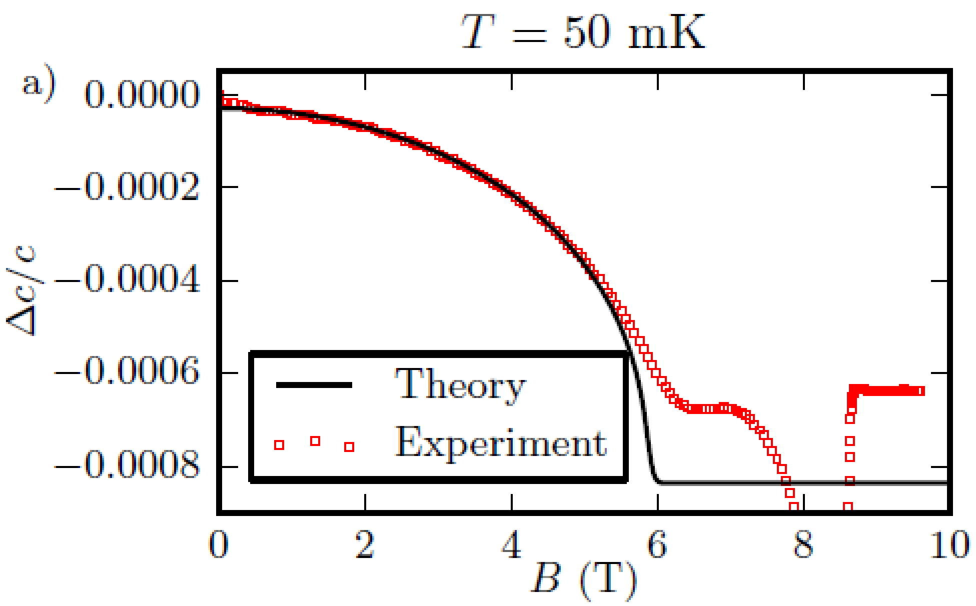
$$\Pi_3(q, i\omega) = \frac{1}{MN} \sum_k \frac{f_k - f_{k+q}}{\xi_k - \xi_{k+q} + i\omega} |\Gamma_3(k,q)|^2$$

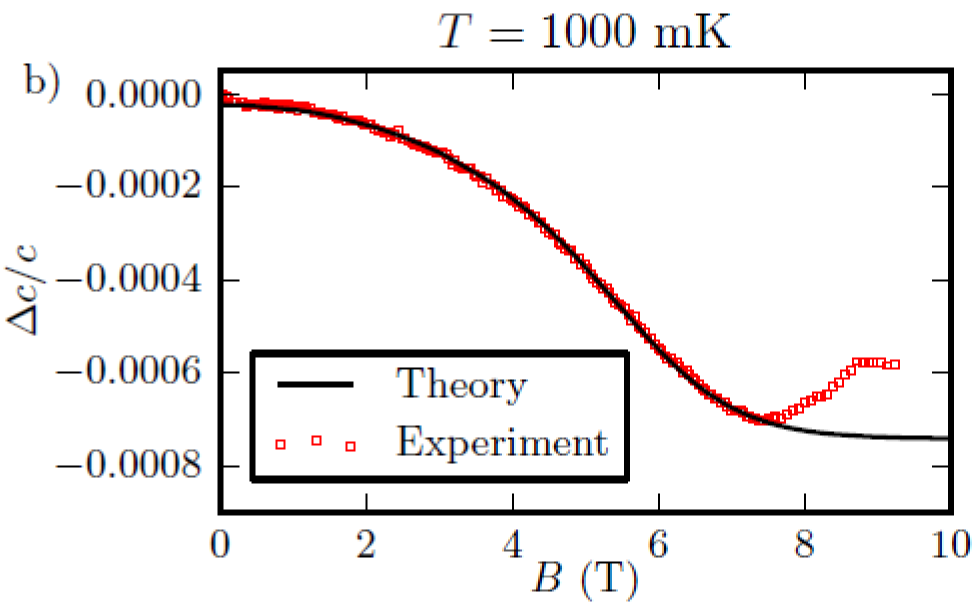
sound velocity in spin-liquid phase: theory+experiment

$$\tilde{\omega}_q = \omega_q + \frac{\mathrm{Re}\Pi(q, \omega_q + i0)}{2\omega_q} \qquad \tilde{c}/c = \lim_{q\to 0} \tilde{\omega}_q/\omega_q$$

ordered phase:

spin-liquid phase:





3. Vicinity of quantum critical point

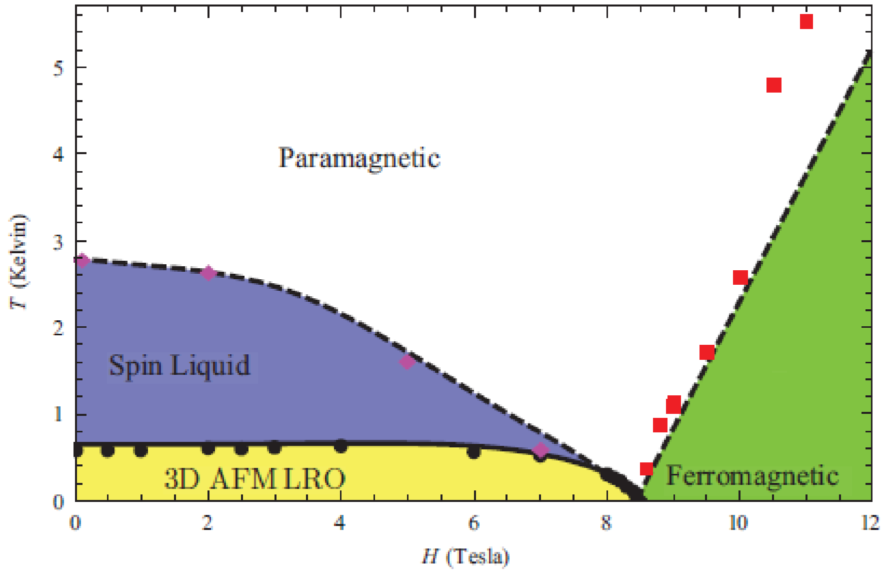
(with Simon Streib, unpublished)

- mapping to hard-core bosons at low densities

$$S_i^+ = \tilde{b}_i, \quad S_i^- = \tilde{b}_i^\dagger, \quad S_i^z = \frac{1}{2} - \hat{n}_i$$

$$\hat{n}_i = \tilde{b}_i^\dagger \tilde{b}_i = 0, 1$$

$$[\tilde{b}_i, \tilde{b}_j^\dagger] = \delta_{ij}(1 - 2\hat{n}_i)$$



$$\mathcal{H} = \frac{1}{2} \sum_{ij} [J_{ij} \boldsymbol{S}_i \cdot \boldsymbol{S}_j + \boldsymbol{D}_{ij} \cdot (\boldsymbol{S}_i \times \boldsymbol{S}_j)] - h \sum_i S_i^z$$

$$= \frac{N}{2} \left(\frac{J_0}{4} - h \right) + \sum_{\boldsymbol{k}} \left(\frac{J_{\boldsymbol{k}} + D_{\boldsymbol{k}}}{2} + h \right) \tilde{b}_{\boldsymbol{k}}^\dagger \tilde{b}_{\boldsymbol{k}} + \frac{1}{2N} \sum_{\boldsymbol{q}\boldsymbol{k}\boldsymbol{k}'} J_{\boldsymbol{q}} \tilde{b}_{\boldsymbol{k}+\boldsymbol{q}}^\dagger \tilde{b}_{\boldsymbol{k}'-\boldsymbol{q}}^\dagger \tilde{b}_{\boldsymbol{k}'} \tilde{b}_{\boldsymbol{k}};$$

from hard-core to soft-core bosons

- trade hard-core constraint for infinite on-site interaction (Matsubara+Matsuda 1956)

$$\mathcal{H}_{\text{projection}} = \frac{U}{2}\sum_i \hat{n}_i(\hat{n}_i - 1) \qquad U \to \infty \qquad [b_i, b_j^\dagger] = \delta_{ij}$$

$$\mathcal{H}_U = \frac{N}{2}\left(\frac{J_0}{4} - h\right) + \sum_{\boldsymbol{k}} (\epsilon_{\boldsymbol{k}} - \mu)\, b_{\boldsymbol{k}}^\dagger b_{\boldsymbol{k}} + \frac{1}{2N}\sum_{\boldsymbol{q}\boldsymbol{k}\boldsymbol{k}'} (J_{\boldsymbol{q}} + U) b_{\boldsymbol{k}+\boldsymbol{q}}^\dagger b_{\boldsymbol{k}'-\boldsymbol{q}}^\dagger b_{\boldsymbol{k}'} b_{\boldsymbol{k}};$$

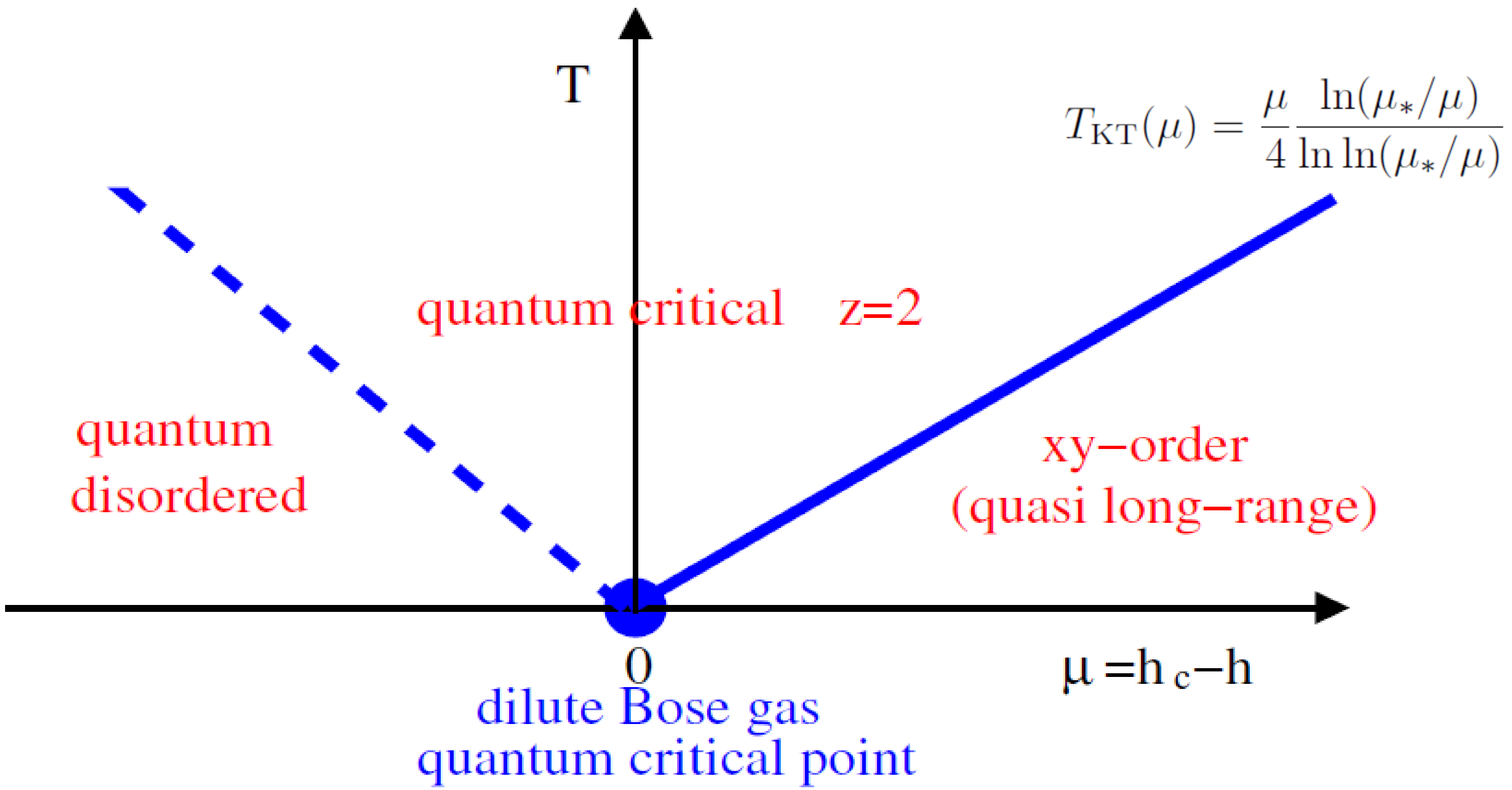
$$\epsilon_{\boldsymbol{k}} = \frac{J_{\boldsymbol{k}}^D - J_{\min}^D}{2} \qquad J_{\boldsymbol{k}}^D = J_{\boldsymbol{k}} + D_{\boldsymbol{k}} \qquad J_{\min}^D = \min_{\boldsymbol{k}}\left\{J_{\boldsymbol{k}}^D\right\} = J_{\boldsymbol{Q}}^D$$

$$\mu = \frac{J_0^D - J_{\min}^D}{2} - h \equiv h_c - h.$$

- hypothesis: softenting hard-core constraint does not change critical behavior (Kawashima, JPS Jpn 2004)

Phase diagram of dilute bosons in 2d

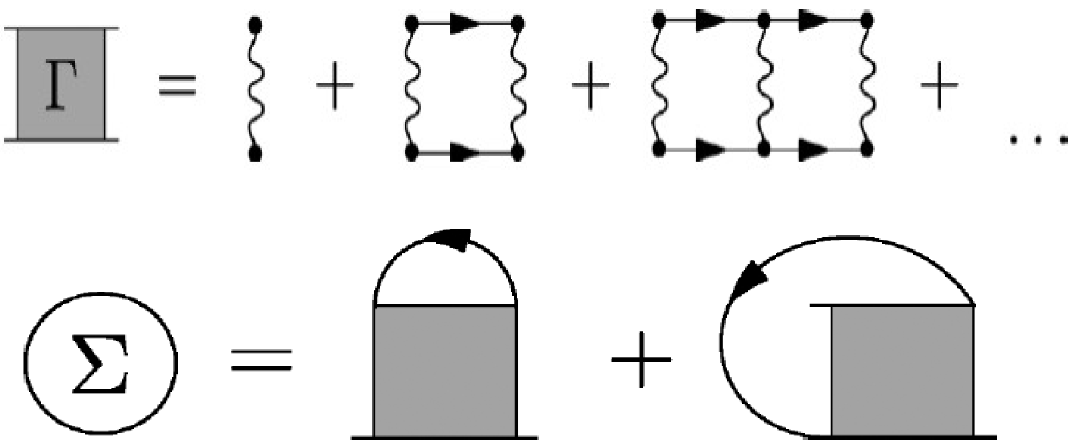
(Sachdev, Sentil, Shankar PRB 94; Straßel, P.K., Eggert, PRB 2015)



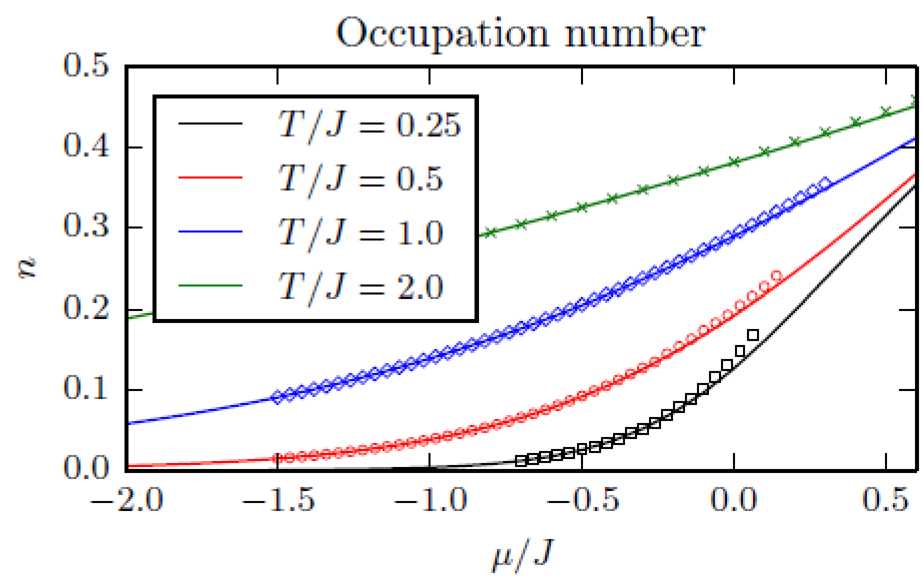
thermodynamics and correlation functions of hard core bosons

- quantum disordered+quantum critical regime at low densities: use self-consistent ladder approximation

- advantage: hard-core constraint can be implemented exactly (1d: Fauseweh, Stolze, Uhrig PRB 2014)



compare with exact results for 1d xy model:



- numerical solution in 2d: Streib+PK, soon to be published

beyond self-consistent ladder: FRG

- Hubbard-Stratonovich transformation in particle-particle channel:

$$S[a,\psi] = -\int_K (i\omega - \xi_{\boldsymbol{k}} + \mu)\bar{a}_K a_K + \frac{1}{2}\int_Q \int_K \int_{K'} J_{\boldsymbol{q}} \bar{a}_{K+Q}\bar{a}_{K'-Q} a_{K'} a_K$$
$$+ \int_P f_0^{-1}\bar{\psi}_P \psi_P + \frac{i}{2}\int_P [\bar{\psi}_P C_P + \bar{C}_P \psi_P]$$

$$f_0 = 2U \qquad C_P = \int_K a_K a_{P-K}$$

- introduce cutoffs (regulators) in Gaussian propagators:

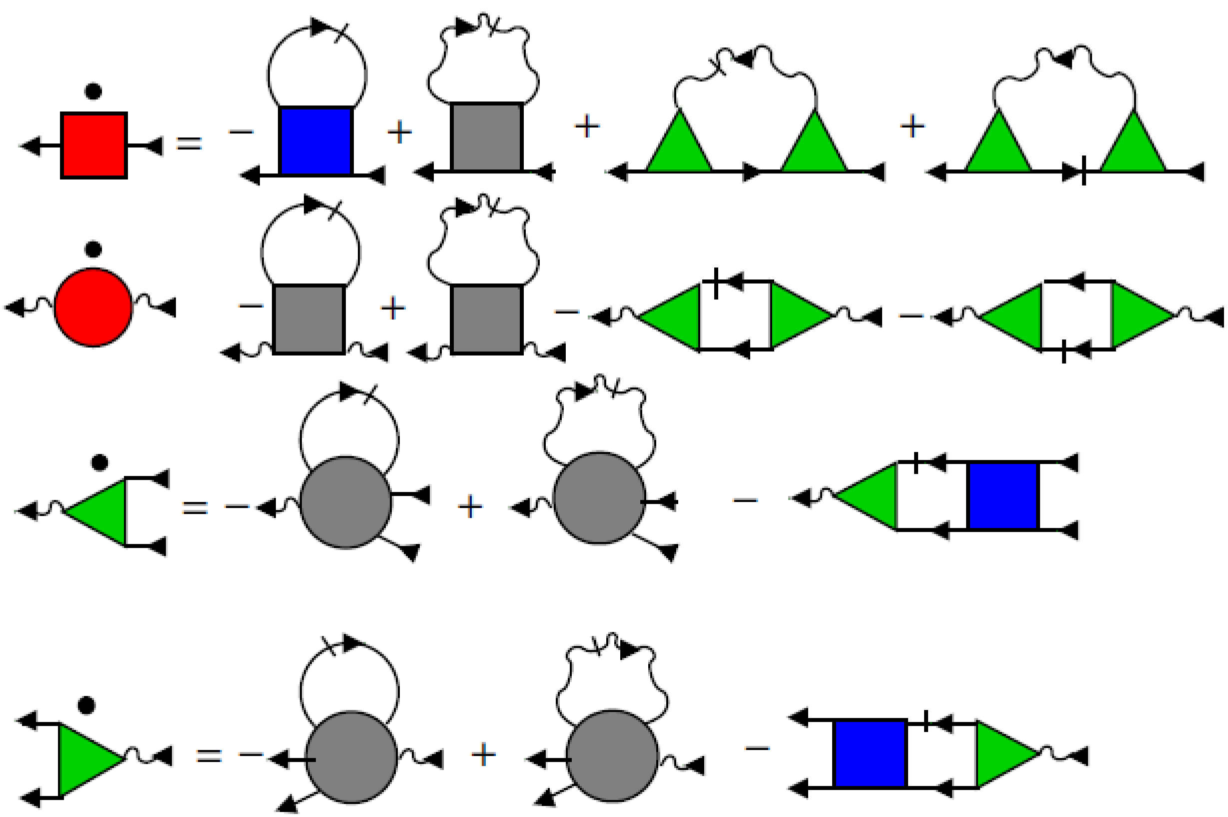
$$G_{0,\Lambda}^{-1}(K) = i\omega - \xi_{\boldsymbol{k}} + \mu - R_\Lambda^a(K)$$
$$F_{0,\Lambda}^{-1}(P) = f_0^{-1} + R_\Lambda^\psi(P)$$

- FRG: formally exact RG flow equations for irreducible vertices (review: PK, Bartosch, Schütz, Springer 2010)

FRG flow equations

$$\partial_\Lambda \Sigma_\Lambda(K) = -\int_{K'} \dot{G}_\Lambda(K')\Gamma_\Lambda^{\bar{a}\bar{a}aa}(K,K';K',K) \;+ \int_P \dot{F}_\Lambda(P)\Gamma_\Lambda^{\bar{a}a\bar{\psi}\psi}(K;K;P;P)$$

$$+\int_P \left[\dot{F}_\Lambda(P)G_\Lambda(P-K)+F_\Lambda(P)\dot{G}_\Lambda(P-K)\right]\times\Gamma_\Lambda^{\bar{a}\bar{a}\psi}(P-K,K;P)\Gamma_\Lambda^{aa\bar{\psi}}(P-K,K;P)$$



Summary+Conclusions:

Ultrasound in different parts of phase diagram of $\mathrm{Cs_2CuCl_4}$

•magneticlly ordered phase: good agreement with experiment and spin- wave theory

•spin-liquid phase: good agreement with experient and Jordan-Wigner theory of 1d Heisenberg chain

•close to quantum critical point:
use dilute bose gas formalism;
numerical implementation of self-consistent ladder approx
need RG methods to go beyond ladder approximation
ultrasound still to be calculated