Magnon-Phonon Interactions: elastic constants & ultrasonic attenuation rate in Cs₂CuCl₄

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Cs₂CuCl₄ as frustrated antiferromagnet



- classical spins: energy $E_0^{cl} = N \frac{S^2}{2} \left[s_{\vartheta}^2 J_{\mathsf{k}=0} + c_{\vartheta}^2 J_{\mathsf{Q}}^D \right] - NShs_{\vartheta}$
- classical ground state: spiral minimization with respect to *v* and Q

$$NSc_{\vartheta} \left[Ss_{\vartheta} \left(J_0^D - J_{\mathbf{Q}}^D \right) - h \right] = 0$$
$$\nabla_{\mathbf{k}} \left(J_{\mathbf{k}} - iD_{\mathbf{k}} \right)_{\mathbf{k} = \mathbf{Q}} = 0$$

 $s_{\vartheta} = \sin \vartheta = h/h_c \quad c_{\vartheta} = \cos \vartheta$ $J_{\mathsf{k}} = 2J\cos(k_x b) + 4J'\cos(k_x b/2)\cos(k_y c/2)$ $D_{\mathsf{k}} = -4iD\sin(k_x b/2)\cos(k_y c/2)$

projection of ground state to plane: "Cone state"

 $2\pi/Q_x$

b

 $J_{\mathbf{k}}^{D} = J_{\mathbf{k}} - iD_{\mathbf{k}}$

Veillette et al. '05



 magnon-magnon interactions here: in non-diagonal basis

$$\hat{H}_{3} = \frac{1}{\sqrt{N}} \sum_{\mathbf{k}_{1}\mathbf{k}_{2}\mathbf{k}_{3}} \delta_{\mathbf{k}_{1}+\mathbf{k}_{2}+\mathbf{k}_{3},0} \Big[\frac{1}{2!} \Gamma_{3}^{b^{\dagger}b^{\dagger}b}(\mathbf{k}_{1},\mathbf{k}_{2};\mathbf{k}_{3}) b_{-\mathbf{k}_{1}}^{\dagger} b_{-\mathbf{k}_{2}}^{\dagger} b_{\mathbf{k}_{3}} \\ \frac{1}{2!} \Gamma_{3}^{b^{\dagger}bb}(\mathbf{k}_{1};\mathbf{k}_{2},\mathbf{k}_{3}) b_{-\mathbf{k}_{1}}^{\dagger} b_{\mathbf{k}_{2}} b_{\mathbf{k}_{3}} \Big]$$

$$\Gamma_3^{b^{\dagger}b^{\dagger}b}(\mathsf{k}_1,\mathsf{k}_2;\mathsf{k}_3) = -c_{\vartheta}s_{\vartheta}\frac{\sqrt{2S}}{i}\frac{h_c}{S} + \mathcal{O}(\mathsf{k}^2)$$

constant term for finite magnetic field (strong interactions at long wavelengths)



 $s_{\vartheta} = \sin \vartheta = h/h_c$ $c_{\vartheta} = \cos \vartheta$



Hermitian parametrization

 $\hat{H}_{2\text{mag}} = \frac{1}{2} \sum_{\mathbf{k}} \left\{ \hat{\Pi}_{-\mathbf{k}} \hat{\Pi}_{\mathbf{k}} + \epsilon_{\mathbf{k}}^2 \hat{\Phi}_{-\mathbf{k}} \hat{\Phi}_{\mathbf{k}} \right.$

$$b_{\mathbf{k}} = \sqrt{\frac{\Delta_{\mathbf{k}}}{2}} \hat{\Phi}_{\mathbf{k}} + \frac{i}{\sqrt{2\Delta_{\mathbf{k}}}} \hat{\Pi}_{\mathbf{k}} \qquad \begin{bmatrix} \hat{\Phi}_{\mathbf{k}}, \hat{\Pi}_{\mathbf{k}'} \end{bmatrix} = i \delta_{\mathbf{k}, -\mathbf{k}'}$$
$$\Delta_{\mathbf{k}} = A_{\mathbf{k}}^{+} - B_{\mathbf{k}}$$

Hasselmann *et al.* '06 Kreisel *et al.* '08

- $+iA_{\mathbf{k}}^{-}(\hat{\Phi}_{-\mathbf{k}}\hat{\Pi}_{\mathbf{k}}+\hat{\Phi}_{\mathbf{k}}\hat{\Pi}_{-\mathbf{k}})-A_{\mathbf{k}}^{+}\Big\}$
- sort longitudinal and transversal fluctuations

Extension of model



• 1/S expansion of coupling term

$$\hat{H}_{spin}^{pho} = \hat{H}_{spin} + \hat{H}_{spin}^{1pho} + \hat{H}_{spin}^{2pho} + \dots$$

 $\hat{H}_{spin}^{npho} = \frac{1}{2} \sum_{ij} U_{ij}^{(n)} S_i \cdot S_j$
 $U_{ij}^{(1)} = (X_{ij} \cdot \nabla_r) J(r)|_{r=R_{ij}} \equiv X_{ij} \cdot J_{ij}^{(1)} \qquad J_{-k}^{(1)} = -J_k^{(1)} = (J_k^{(1)})^*$
 $U_{ij}^{(2)} = \frac{1}{2} (X_{ij} \cdot \nabla_r)^2 J(r)|_{r=R_{ij}} \equiv \frac{1}{2} X_{ij}^{T} J_{ij}^{(2)} X_{ij} \qquad J_{-k}^{(2)} = J_k^{(2)}$

Phonon shift

$$\hat{H}_{0}^{2\text{pho}} = \frac{M}{2} \sum_{\mathbf{k}\lambda} \Sigma_{0}^{\text{pho}}(\mathbf{k},\lambda) \hat{X}_{-\mathbf{k}\lambda} \hat{X}_{\mathbf{k}\lambda}$$
$$\Sigma_{0}^{\text{pho}}(\mathbf{k},\lambda) = \frac{S^{2}}{M} \mathbf{e}_{\mathbf{k}\lambda}^{\dagger} \left[s_{\vartheta}^{2} \left(\mathbf{J}_{0}^{(2)} - \mathbf{J}_{\mathbf{k}}^{(2)} \right) + c_{\vartheta}^{2} \mathbf{J}_{\mathbf{Q},\mathbf{k}}^{(2+)} \right] \mathbf{e}_{\mathbf{k}\lambda}$$

Hybridization (coupled magnoelastic waves) *Ĥ*^{1pho}_{1mag} = ∑_k Γ^{Xb}_k ⋅ (X -_kb_k + X k[†]_k) *Γ*^{Xb}_k = ⁱ/₄(2S)^{3/2}c_θ [J⁽¹⁺⁾_{k,Q} + s_θJ⁽¹⁻⁾_{k,Q}]

Hermitian parametrization (sorts relevant degrees of freedom) *Ĥ*^{1pho}₁ = ¹/₄ ∑ {Γ^{XΦ}_k ⋅ (X k^Φ_k + X k^Φ_k)

$$\frac{1}{1} \sum_{k=1}^{1} \frac{1}{2} \sum_{k=1}^{1} \left\{ \Gamma_{k}^{X\Phi} \cdot \left(\mathbf{X}_{-k} \hat{\Phi}_{k} + \mathbf{X}_{k} \hat{\Phi}_{-k} \right) + \Gamma_{k}^{X\Pi} \cdot \left(\mathbf{X}_{-k} \hat{\Pi}_{k} - \mathbf{X}_{k} \hat{\Pi}_{-k} \right) \right\}$$

crucial interaction

• three particle interactions – one phonon two magnon scattering $\hat{H}_{2\text{mag}}^{1\text{pho}} = \frac{1}{\sqrt{N}} \sum_{\mathbf{k}\mathbf{k}'} \left[\Gamma_{\mathbf{k},\mathbf{k}'}^{b^{\dagger}b} \cdot \mathbf{X}_{\mathbf{k}-\mathbf{k}'} b_{\mathbf{k}}^{\dagger} b_{\mathbf{k}'} \right]$

$$+ \frac{1}{2!} \left(\Gamma_{\mathbf{k},\mathbf{k}'}^{b^{\dagger}b^{\dagger}} \cdot \mathbf{X}_{\mathbf{k}+\mathbf{k}'} b_{\mathbf{k}}^{\dagger} b_{\mathbf{k}'}^{\dagger} + \Gamma_{\mathbf{k},\mathbf{k}'}^{bb} \cdot \mathbf{X}_{-\mathbf{k}-\mathbf{k}'} b_{\mathbf{k}} b_{\mathbf{k}'} \right)$$

- two phonon one magnon scattering

$$\hat{H}_{1\text{mag}}^{\text{2pho}} = \frac{1}{2!\sqrt{N}} \sum_{\mathbf{k},\mathbf{k}'} \sum_{\lambda\lambda'} \Big[\Gamma_{\mathbf{k}\lambda,\mathbf{k}'\lambda'}^{XXb^{\dagger}} \hat{X}_{\mathbf{k}\lambda} \hat{X}_{\mathbf{k}'\lambda'} b_{\mathbf{k}+\mathbf{k}'}^{\dagger} \Big]$$

 $+ \Gamma^{XXb}_{\mathbf{k}\lambda,\mathbf{k}'\lambda'} \hat{X}_{-\mathbf{k}\lambda} \hat{X}_{-\mathbf{k}'\lambda'} b_{\mathbf{k}+\mathbf{k}'} \Big]$

- magnon shift performed in Hermitian parametrization
 - $$\begin{split} \Gamma_{\mathbf{k}}^{X\Pi} &\to 0 \\ \mathbf{b}_{\mathbf{k}} &= \tilde{\mathbf{b}}_{\mathbf{k}} + \lambda_{\mathbf{k}} \cdot \mathbf{X}_{\mathbf{k}} \end{split} \qquad \lambda_{\mathbf{k}} = \frac{i}{\sqrt{2\Delta_{\mathbf{k}}}} \Gamma_{\mathbf{k}}^{X\Pi} \end{split}$$
- renormalization of one-phonon two magnon interaction $\Gamma_{\mathbf{k},\mathbf{k}'}^{b^{\dagger}b^{\dagger}} \rightarrow \tilde{\Gamma}_{\mathbf{k},\mathbf{k}'}^{b^{\dagger}b^{\dagger}} = \Gamma_{3}^{b^{\dagger}bb}(-\mathbf{k} - \mathbf{k}';\mathbf{k},\mathbf{k}')\lambda_{\mathbf{k}+\mathbf{k}'}$ $\tilde{H}_{2\mathrm{mag}}^{1\mathrm{pho}} = \frac{1}{\sqrt{N}} \sum_{\mathbf{k}\mathbf{k}'} \left[\tilde{\Gamma}_{\mathbf{k},\mathbf{k}'}^{b^{\dagger}b} \cdot \mathbf{X}_{\mathbf{k}-\mathbf{k}'} \tilde{b}_{\mathbf{k}}^{\dagger} \tilde{b}_{\mathbf{k}'} + \frac{1}{2!} \left(\tilde{\Gamma}_{\mathbf{k},\mathbf{k}'}^{b^{\dagger}b^{\dagger}} \cdot \mathbf{X}_{\mathbf{k}+\mathbf{k}'} \tilde{b}_{\mathbf{k}}^{\dagger} \tilde{b}_{\mathbf{k}'}^{\dagger} + \tilde{\Gamma}_{\mathbf{k},\mathbf{k}'}^{bb} \cdot \mathbf{X}_{-\mathbf{k}-\mathbf{k}'} \tilde{b}_{\mathbf{k}} \tilde{b}_{\mathbf{k}'} \right) \right]$

Lagrangian functional integral

 Transform to Bogoliubov basis $\begin{pmatrix} b_{\mathsf{k}} \\ b_{-\mathsf{k}}^{\dagger} \end{pmatrix} = \begin{pmatrix} u_{\mathsf{k}} & -v_{\mathsf{k}} \\ -v_{\mathsf{k}} & u_{\mathsf{k}} \end{pmatrix} \begin{pmatrix} \beta_{\mathsf{k}} \\ \beta_{-\mathsf{k}}^{\dagger} \end{pmatrix}$ integrate out canonical momentum fields $\mathcal{Z} = \int \mathcal{D}[\mathsf{P}, \mathsf{X}, \beta, \overline{\beta}] e^{-S'[\mathsf{P}, \mathsf{X}, \beta, \overline{\beta}]} \sim \widetilde{\Gamma}^{\beta^{\dagger}\beta}$ $\bigwedge \tilde{\Gamma}^{\beta\beta} \qquad \bigwedge \tilde{\Gamma}^{XX\beta}$ $= \int \mathcal{D}[\mathbf{X}, \boldsymbol{\beta}, \bar{\boldsymbol{\beta}}] e^{-S[\mathbf{X}, \boldsymbol{\beta}, \bar{\boldsymbol{\beta}}]}$ $~~\tilde{\Gamma}^{\beta^\dagger\beta^\dagger}$
$$\begin{split} S[\mathsf{X} \ , \bar{\beta}, \beta] &= S^{2\mathrm{pho}}[\mathsf{X} \] + S_{2\mathrm{mag}}[\bar{\beta}, \beta] + S^{1\mathrm{pho}}_{1\mathrm{mag}}[\mathsf{X} \ , \bar{\beta}, \beta] \\ &+ S^{1\mathrm{pho}}_{2\mathrm{mag}}[\mathsf{X} \ , \bar{\beta}, \beta] + S_{3\mathrm{mag}}[\bar{\beta}, \beta] + \dots \end{split}$$

Many particle methods: Phonon renormalization



Shift of elastic constants

• classical spin background

$$\frac{(\Delta c_{\lambda})_{0}}{c_{\lambda}} = \sqrt{1 - \lim_{|\mathbf{k}| \to 0} \frac{\Sigma_{0}^{\text{pho}}(\mathbf{k}, \lambda)}{\omega_{\mathbf{k}\lambda}^{2}}} - 1$$

 magnon-phonon Hybridization (equivalent to diagonalization in Hamilton formulation)

$$S_1[\mathsf{X},\beta,\beta] = S^{2\text{pno}}[\mathsf{X}] + S_{2\text{mag}}[\beta,\beta] + S_{1\text{mag}}^{1\text{pno}}[\mathsf{X},\beta,\beta]$$

 $\frac{(\Delta c_{\lambda})_{1}}{c_{\lambda}} = \frac{S^{3}}{4} \left(\frac{v(\hat{\mathbf{k}})}{c_{\lambda}} \right) \left(\frac{h_{c}}{Mc_{\lambda}^{2}} \right) \left| s_{\vartheta} \mathbf{f}_{1}^{X\beta}(\hat{\mathbf{k}}) \cdot \mathbf{e}_{\mathbf{k}\lambda} - c_{\vartheta}^{2} \mathbf{f}_{2}^{X\beta}(\hat{\mathbf{k}}, \hat{\mathbf{k}}) \cdot \mathbf{e}_{\mathbf{k}\lambda} \right|^{2}$

$$e^{-S_{\rm eff}^{\rm 2pho}[\mathsf{X}]} = \int \mathcal{D}[\beta, \bar{\beta}] e^{-S_1[\mathsf{X}, \bar{\beta}, \beta]}$$

$$\frac{(\Delta c_{\lambda})_{1}}{c_{\lambda}} = \lim_{|\mathbf{k}| \to 0} \frac{\left|\Gamma_{\mathbf{k}}^{\mathbf{\lambda}\beta} \cdot \mathbf{e}_{\mathbf{k}\lambda}\right|^{2}}{2M\omega_{\mathbf{k}\lambda}^{3}}$$

 $s_artheta = \sin artheta = h/h_c$ c

 $c_{artheta} = \cos artheta$

magnetic field dependence

Ultrasound Attenuation Rate



 $S_{2}[\mathsf{X} \ ,\bar{\beta},\beta] = S^{2\mathrm{pho}}[\mathsf{X} \] + S_{2\mathrm{mag}}[\bar{\beta},\beta] + \tilde{S}^{1\mathrm{pho}}_{2\mathrm{mag}}[\mathsf{X} \ ,\bar{\beta},\beta] + S^{2\mathrm{pho}}_{1\mathrm{mag}}[\mathsf{X} \ ,\bar{\beta},\beta]$

attenuation rate

$$\gamma_{\mathbf{k}\lambda} = -\frac{\mathrm{Im}\Sigma_{2}^{\mathrm{pho}}(\omega_{\mathbf{k}\lambda} + i0^{+}, \mathbf{k}, \lambda)}{2\omega_{\mathbf{k}\lambda}}$$



Ultrasound attenuation rate



Ultrasound attenuation rate

process (b)



$$\gamma_{\mathbf{k}\lambda}^{(\mathrm{b})} = \frac{\pi S^3}{4} \left(\frac{\mathbf{k}^2}{2M}\right) \left(\frac{\mathbf{k}^2}{V_{\mathrm{BZ}}}\right) \sum_{\lambda'} \left(\frac{h_c}{Mc_{\lambda'}^2}\right) \left(\frac{c_\lambda}{c_{\lambda'}}\right)^2 \int_0^{2\pi} d\varphi' \frac{u(\hat{\mathbf{k}},\varphi')}{c_{\lambda'}} |\mathbf{e}_{\mathbf{k}\lambda}^{\dagger} \mathbf{F}^{XX\beta}(\hat{\mathbf{k}},\varphi') \mathbf{e}_{\mathbf{k}'\lambda'}|^2 ,$$

higher power in v/c<<1

both contributions

$$\gamma_{\mathbf{k}\lambda} = \gamma^{(\mathrm{a})}_{\mathbf{k}\lambda} + \gamma^{(\mathrm{b})}_{\mathbf{k}\lambda} \propto \mathbf{k}^4$$

Comparison to experiments

model

 $J(x) = J(b)e^{-\kappa(x-b)/b}$ $J'(r) = J'(d)e^{-\kappa'(r-d)/d}$

- shift of ultrasound velocity for c₂₂ mode: fix parameters $|\kappa| \approx 15$ $|\kappa'| \approx 51$
- attenuation rate calculate from parameters

$$\gamma_{\mathbf{k}\lambda} \approx \frac{\pi^2}{64} \left(\frac{\mathbf{k}^2}{2M}\right) \left(\frac{S^2 c_\lambda^2 \mathbf{k}^2}{V_{\mathrm{BZ}} v_x v_y}\right) \frac{\left[\mathbf{f}_1^{X\beta}(\hat{\mathbf{k}}) \cdot \mathbf{e}_{\mathbf{k}\lambda}\right]^2}{(1 - h/h_c)^2}$$

P. T. Cong *et. al* (in preparation)





Summary

- ultrasonic technique: probe magnetic properties
- combine spin-wave approach for ordered "cone-state" with expansion in terms of lattice vibrations
- calculate renormalization of phonons using an effective action
- good description of phonon properties (sound velocity, damping) away from critical point



(b)

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 $^{1}B(T)$