field-induced thermal transport in BEC antiferromagnets



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Fig. 1. The crystal structure of EtMe₃Sb[Pd(dmit)₂]₂









Chinese Terracotta Warriors (479-221 BC)



H. Langhals & D. Bathelt, Angew. Chem. Int. Ed. 42, 5676 (2003)

Angew. Chem. Int. Ed. 41, 2483 (2002)

BEC: coupled dimers



• Ba-Cu silicate, Han Purple = $BaCuSi_2O_6$

 $\boldsymbol{H} = \boldsymbol{J} \boldsymbol{\Sigma} \boldsymbol{S}_{i} \boldsymbol{S}_{i'} + \boldsymbol{J}' \boldsymbol{\Sigma} \boldsymbol{S}_{i} \boldsymbol{S}_{j}$



dimer = antiferromagnetic pair of S=1/2





• array of coupled spin dimers



BEC: coupled dimers



BEC ... in a magnetic field

single spin dimer





coupled spin dimers











BEC, III: PhD



courtesy of Marcelo Jaime, Netsu Sokutei **37**, 26 (2010).



BEC, IV: DTN



KITP, 8-30-12

courtesy of Marcelo Jaime



DTN







DTN, I: model



KITP, 8-30-12

thermal conductivity, other BECs



C. C.

thermal conductivity experiments, DTN

thermal transport in BEC antiferromagnets vs magnetic field

<u>S=1 AF NiCl₂-4SC(NH₂)₂</u>







thermal conductivity experiments, DTN



quick analysis

- \square at H=0, gaps \rightarrow conductivity is due to phonons alone (small $T < < \Delta \sim 3K$)
- \square excess of the conductivity is due to spin excitations
- \square peaks are near H_{c1} and H_{c2} , κ/κ_0 increases as *T* decreases
- \square at larger *T*, suppression of κ in the BEC phase is due to phonon scattering on magnons





puzzles, I: "no asymmetry" problem





puzzles, I: "no asymmetry" problem





"no asymmetry" problem





specific heat





-- m_2 is "bare", $m_1 < m_2$ is renormalized by fluctuations

labels the eigenstates of $S_{\mathbf{r}}^z$ with the eigenvalues $\{-1, 0, 1\}$. The spin operators in this representation are:

$$S_{\mathbf{r}}^{z} = n_{\mathbf{r}\uparrow} - n_{\mathbf{r}\downarrow}, \ S_{\mathbf{r}}^{+} = \left(S_{\mathbf{r}}^{-}\right)^{\dagger} = \sqrt{2} \left(b_{\mathbf{r}\uparrow}^{\dagger}b_{\mathbf{r}0} + b_{\mathbf{r}0}^{\dagger}b_{\mathbf{r}\downarrow}\right), (2)$$

with $n_{\mathbf{r}m} = b^{\dagger}_{\mathbf{r}m} b_{\mathbf{r}m}$. We enforce the constraint by introducing spatially uniform Lagrange multiplier μ

$$\hat{\mathcal{H}} = \mathcal{H} + \mu \sum_{\mathbf{r}} \left(b_{\mathbf{r}\uparrow}^{\dagger} b_{\mathbf{r}\uparrow}^{\dagger} + b_{\mathbf{r}\downarrow}^{\dagger} b_{\mathbf{r}\downarrow}^{\dagger} + b_{\mathbf{r}0}^{\dagger} b_{\mathbf{r}0}^{\dagger} - 1 \right).$$
(3)

The lowest energy state in the $H < H_{c1}$ paramagnetic regime is $b_{\mathbf{r}0}^{\dagger}|0\rangle$ and the ground state corresponds to a non-zero expectation value of the $S^z = 0$ boson: $b_{\mathbf{r}0}^{\dagger} = b_{\mathbf{r}0} = s$. By using the spin representation (2) with the mean-field value for $b_0^{(\dagger)}$ and neglecting higher-order terms in powers of $b_{\uparrow(\downarrow)}^{(\dagger)}$, we obtain the Hamiltonian in the harmonic approximation

$$\hat{\mathcal{H}} = E_0 + \sum_{\mathbf{k},\sigma} \left[A_{\mathbf{k}\sigma} \hat{b}^{\dagger}_{\mathbf{k}\sigma} \hat{b}_{\mathbf{k}\sigma} + \frac{B_{\mathbf{k}}}{2} \left(\hat{b}^{\dagger}_{\mathbf{k}\sigma} \hat{b}^{\dagger}_{-\mathbf{k}\bar{\sigma}} + \text{H.c.} \right) \right], (4)$$

with $A_{\mathbf{k}\sigma} = (\mu + s^2 \epsilon_{\mathbf{k}} - h_{\sigma})$ and $B_{\mathbf{k}} = s^2 \epsilon_{\mathbf{k}}$, where $E_0 = N(\mu - D)(s^2 - 1)$ is the bare ground-state energy, N is the number of sites, $\sigma = \{\uparrow, \downarrow\}, h_{\sigma} = \pm h, \bar{\sigma} = -\sigma, \hat{b}_{\mathbf{k}\sigma}^{(\dagger)}$ are the Fourier transformed bosonic operators, and $\epsilon_{\mathbf{k}} = 2 \sum_{\nu} J_{\nu} \cos k_{\nu}$. The anomalous terms indicate that bosons with opposite S^z are created and annihilated in the ground state. These are the quantum fluctuations that lead to renormalization of the quasiparticle dispersion relation. The Hamiltonian (4) is diagonalized by the Bogolyubov transformation

Κ

$$\hat{b}_{\mathbf{h}} = u_{\mathbf{h}}\beta_{\mathbf{h}} + v_{\mathbf{h}}\beta^{\dagger}, \qquad (5)$$

 $H \ge H_{c2}$ spins are fully polarized and the spectrum can be computed exactly. Since there are no quantum fluctuations for $H \ge H_{c2}$, the exact value of h_{c2} is $h_{c2} = g\mu_B H_{c2} =$ $D - 2\epsilon_{\mathbf{Q}}$, while the unrenormalized excitation spectrum is $\widetilde{\omega}_{\mathbf{k}}^{>} \equiv \epsilon_{\mathbf{k}} - \epsilon_{\mathbf{Q}} + h - h_{c2}$, which also has a minimum at \mathbf{Q} with the gap $\Delta^{>} = h - h_{c2}$. Since only the excitations near $\mathbf{k} = \mathbf{Q}$ are important at low temperatures, we define the mass tensors for $H < H_{c1}$ and $H > H_{c2}$ as:

$$\frac{1}{m_{\nu\nu}^*} = \frac{\partial^2 \widetilde{\omega}_{\mathbf{k}}^<}{\partial k_{\nu}^2} \bigg|_{\mathbf{k}=\mathbf{Q}}, \qquad \frac{1}{m_{\nu\nu}} = \frac{\partial^2 \widetilde{\omega}_{\mathbf{k}}^>}{\partial k_{\nu}^2} \bigg|_{\mathbf{k}=\mathbf{Q}}.$$
 (9)

Then the mass renormalization factor is given by

$$\frac{m_{\nu\nu}}{m_{\nu\nu}^*} = s^2 \frac{\mu}{\omega_{\mathbf{Q}}^0} \approx \frac{H_{c2}}{4H_{c1}} \cdot \left(1 + \sqrt{1 + \frac{8H_{c1}^2}{H_{c2}^2}}\right).$$
(10)

For the parameters of the Hamiltonian from Ref. [8], we obtain $m_{\nu\nu}/m_{\nu\nu}^* \simeq 3.2$. Such a large difference of masses must readily demonstrate itself in the strong asymmetry of the C_v vs H curves near H_{c1} and H_{c2} as well as in the slopes of the $m_2 \over C_v m_1} \approx \frac{H_{c2}}{2H_{c1}}$ for $H_{c1} \ll H_{c2}$ ere ted

The C_p was measured in single crystals of DTN grown from aqueous solutions of thiourea and nickel chloride, with magnetic field applied along the crystalline *c*-axis. The experimental C_p vs *H* was obtained using an AC technique [17], while sweeping the magnetic field in a ³He fridge furbished with a 17 T superconducting magnet system at the National High Magnetic Field Laboratory (NHMFL) and the Los Alamos National Laboratory. We also used the standard thermal relaxation method to obtain C_v vs *T* with a dilution re-

impurity scattering



$$\ell_{\rm imp}^{-1} = \frac{1}{|\mathbf{v}_{\mathbf{k}}|\tau_{\mathbf{k}}} = \frac{n_i}{2\pi} m^2 |V_{\rm imp}|^2$$

-- ratio of thermal conductivities:

$$\frac{\kappa(H_{c2})}{\kappa(H_{c1})} = \left(\frac{m_2}{m_1}\right) \cdot \frac{\ell_2}{\ell_1}$$

$$\mathfrak{D}_{\mathfrak{S}} \cdot \mathfrak{G}^{\mathfrak{L}}$$



- -- however, V_{imp} is also renormalized by fluctuations (!)
- -- it is **most natural** if renormalization of V_{imp} is the **same** as for $1/m_{r}$,

then $\ell_1 = \ell_2$ and $\frac{\kappa_2}{\kappa_1} = \frac{m_2}{m_1}$ χ_{spin} χ_{spin} ℓ_1 ℓ_2 ℓ_2 ℓ_2

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which impurity: bond/site, weak/strong?

-- ratio of thermal conductivities:

$$\frac{\kappa(H_{c2})}{\kappa(H_{c1})} = \left(\frac{m_2}{m_1}\right) \cdot \frac{\ell_2}{\ell_1}$$

$$\underbrace{\mathbb{D}_{oS} \cdot \mathcal{G}_{z}^{2}}$$



-- impurities: modulations of *D* and *J* ($V_{imp} = \delta D$ and $V_{imp} = \delta J$), weak and strong

"chemistry

- -- which is the leading effect?
- -- DTN=clean material \rightarrow structural distortions, *D* is the leading term \rightarrow weak δD
- -- "direct" check: all but one, same renormalization of V_{imp} as for 1/m





site-disorder

-- thermal conductivities:

$$\frac{\kappa(H_{c2})}{\kappa(H_{c1})} = \left(\frac{m_2}{m_1}\right) \cdot \frac{\ell_2}{\ell_1}$$

-- δD at H_{c1} :

$$\mathcal{H}_{\rm imp}^D = \delta D \sum_{\sigma, \mathbf{k}, \mathbf{k}'} e^{i\mathbf{R}_{\ell}(\mathbf{k} - \mathbf{k}')} \left(u_{\mathbf{k}} u_{\mathbf{k}'} + v_{\mathbf{k}} v_{\mathbf{k}'} \right) \beta_{\mathbf{k}\sigma}^{\dagger} \beta_{\mathbf{k}'\sigma}$$

-- different set of "coherence factors" than in m

$$\frac{\kappa(H_{c2})}{\kappa(H_{c1})} = (u_{\mathbf{Q}} + v_{\mathbf{Q}})^2 \cdot \frac{\left(u_{\mathbf{Q}}^2 + v_{\mathbf{Q}}^2\right)^2}{\left(u_{\mathbf{Q}} + v_{\mathbf{Q}}\right)^4}$$

$$\Rightarrow \frac{\kappa(H_{c2})}{\kappa(H_{c1})} \approx \left(\frac{m_2}{m_1}\right) \cdot \frac{1}{4} \left(1 + \left(\frac{m_1}{m_2}\right)^2\right)^2$$

 $k \rightarrow 0 \underline{k'}$

thermal conductivity vs H_c 's ratio







 -- unless H_{c1} << H_{c2}, κ₂ ≈ κ₁ for any BEC system at low enough T
 -- valid for the **dimer-based systems** as well: modulations of **intra-dimer** J lead to the same scattering as δD
 -- proposal: pressure experiment on DTN, will reduce κ₁





away from QCPs, dispersion, DOS





puzzles, III: minima



"effective" DoS is not that different between $\omega \sim k$ and $\omega \sim k^2$ regimes

✓ what are the minima in the BEC phase?✓ strongly field-dependent scattering?





puzzles, II: "peak migration" problem

 \square peaks/maxima in κ "migrate" **away** from H_c 's as T increases \rightarrow ??











unpublished

scatterings in the paramagnetic phase

 \square both b-b and impurity scattering are important for $\omega_{\mathbf{k}} = \Delta + k^2/2m$ band

$$\mathcal{H} = \sum_{\mathbf{k}} (\varepsilon_{\mathbf{k}} - \mu_0) b_{\mathbf{k}}^{\dagger} b_{\mathbf{k}} + g_0 \sum_{\mathbf{k}, 1, 2} b_{\mathbf{k}+2-1}^{\dagger} b_1^{\dagger} b_2 b_{\mathbf{k}}$$

$$\mathcal{H}_{\rm imp}^D = \delta D \sum_{\mathbf{i}} b_{\mathbf{i}}^{\dagger} b_{\mathbf{i}} = \delta D \sum_{\mathbf{k},\mathbf{k}'} e^{i\mathbf{R}_{\mathbf{i}}(\mathbf{k}-\mathbf{k}')} b_{\mathbf{k}}^{\dagger} b_{\mathbf{k}'}$$

 $\square impurity scattering: k \neq k'$

 \square <u>b-b scattering</u>: k + k' = p + p'







impurity scattering

 \boxdot impurity scattering does not depend on the gap value, only velocity of the boson

$$\frac{1}{\tau_{\mathbf{k}}^{\mathrm{imp}}} \propto n_i |V_i|^2 \, m \, |\mathbf{k}|$$





b-b scattering, on-shell





thermal conductivity, impurity only

$$\kappa \sim \int_0^\infty k^2 dk \cdot \frac{k^2}{m^2} \cdot \frac{\omega_{\mathbf{k}}^2}{T^2} \cdot \frac{e^{\omega_{\mathbf{k}}/T}}{(e^{\omega_{\mathbf{k}}/T} - 1)^2} \cdot \tau_{\mathbf{k}}$$
$$\omega_{\mathbf{k}} = \varepsilon_{\mathbf{k}} - \mu$$

☑ impurity only:☑ some T-dependence, change in shape





thermal conductivity, Goldilocks gap





☑ more heat carriers, but also more scatterers
 → "optimal" gap





what is "optimal" gap (optimal \$\Delta H\$)?
 when the impurity and b-b mean-free paths are equal \$\ell_{imp}\$ \$\approx\$ \$\ell_{bb}\$ (only b-b part knows about the gap \$\Delta=\Delta H\$)\$









✓ strongly field-dependent scattering?

kinematics and interactions



non-collinearity \rightarrow transverse-longitudinal coupling \rightarrow 3-boson terms

- three-boson terms are necessary for 1-in-2 and 2-in-1 decay/recombination processes
- "kinematic" conditions (*E* and *k* conservations) make it sufficient
- forbidden for the convex spectrum and outside of the symmetry-broken phase

two-particle continuum energy: $E^{(2)}_{\mathbf{k}}(\mathbf{q}) = \omega(\mathbf{q}) + \omega(\mathbf{k}-\mathbf{q})$



possible role of 3-boson processes ...







conclusions

- <u>clear</u> field-induced thermal current by spins in DTN
 - "no asymmetry" intriguing compensation of m, δD renorm.
 - "migrating peaks" interplay of imp. and b-b, "Goldilocks gap"
 - "minima" in the ordered state 3-boson decays
- experiments in other BEC's needed (clean and low T!!)

more to come



