Evidence for spinon localization in the heat transport of the spin-$\frac{1}{2}$ ladder compound $(C_5H_{12}N)_2CuBr_4$

A. V. Sologubenko,[1] T. Lorenz,[1,*] J. A. Mydosh,[1,†] B. Thielemann,[2] H. M. Rønnow,[3] Ch. Rüegg,[4] and K. W. Krämer[5]
[1]*II. Physikalisches Institut, Universität zu Köln, Zülpicher Str. 77, 50937 Köln, Germany*
[2]*Laboratory for Neutron Scattering, ETH Zürich & Paul Scherrer Institute, 5232 Villigen PSI, Switzerland*
[3]*Laboratory for Quantum Magnetism, École Polytechnique Fédérale de Lausanne (EPFL), 1015 Lausanne, Switzerland*
[4]*London Centre for Nanotechnology and Department of Physics and Astronomy, University College London, London WC1E 6BT, United Kingdom*
[5]*Department of Chemistry and Biochemistry, University of Bern, Freiestrasse 3, 3012 Bern, Switzerland*
(Received 27 November 2009; published 29 December 2009)

We present experiments on the magnetic-field-dependent thermal transport in the spin-$\frac{1}{2}$ ladder system $(C_5H_{12}N)_2CuBr_4$. The thermal conductivity $\kappa(B)$ is only weakly affected by the field-induced transitions between the gapless Luttinger-liquid state realized for $B_{c1}<B<B_{c2}$ and the gapped states, suggesting the absence of a direct contribution of the spin excitations to the heat transport. We observe, however, that the thermal conductivity is strongly suppressed by the magnetic field deeply within the Luttinger-liquid state. These surprising observations are discussed in terms of localization of spinons within finite ladder segments and spinon-phonon umklapp scattering of the predominantly phononic heat transport.

DOI: 10.1103/PhysRevB.80.220411 PACS number(s): 75.40.Gb, 05.60.Gg, 75.47.−m

Studies of the heat transport in one-dimensional (1D) spin systems are of strong current interest.[1–3] From the theoretical side, there is consensus that the intrinsic spin-mediated heat transport of integrable spin models is ballistic, while the situation in nonintegrable spin models is less clear. Experimentally, such studies were stimulated by the observation of a strong anisotropy of the thermal conductivity $\kappa(T)$ in the spin-$\frac{1}{2}$ ladder compound $(Sr,Ca,La)_{14}Cu_{24}O_{41}$, which has been explained by a large spin contribution κ_s along the ladder direction.[4–6] In order to relate model calculations to experimental data, theory has to incorporate the coupling between spin excitations and the underlying lattice, while experimentally it is necessary to separate κ_s from the measured total κ. Here, studies of the magnetic-field dependent $\kappa(B,T)$ can provide much more information than just the zero field $\kappa(T)$ since strong enough magnetic fields change the spin excitation spectra and cause transitions between different quantum phases. Yet, the above-mentioned $(Sr,Ca,La)_{14}Cu_{24}O_{41}$ is unsuitable for such studies due to the strong exchange interaction ($\sim$2000 K) in the ladders that require magnetic fields far above typical laboratory magnets.

Piperidinium copper bromide $(C_5H_{12}N)_2CuBr_4$ (Ref. 7) is one of the best spin-$\frac{1}{2}$ ladder compounds with weak intraladder exchange. It has a monoclinic structure (space group $P2_1/c$) with the ladders running along the a axis. The rung exchange $J_\perp \simeq 13$ K is about four times larger than the leg exchange $J_\parallel \simeq 3.6$ K.[8–14] The zero-field spin excitation gap $\Delta \simeq 9.5$ K decreases in an external magnetic field, so that between $B_{c1} \simeq 7.1$ T and the full saturation field $B_{c2} \simeq 14.5$ T [for $B \parallel a$, where $g=2.06$ (Ref. 7)] the spin excitations are gapless and the system is in the Luttinger-liquid (LL) state. The LL state extends down to the temperature of a 3D magnetic ordering transition at $T_N \leq 110$ mK, suggesting an interladder coupling $J' \sim 27$ mK$\ll J_\perp$, $J_\parallel$.[10,12,13]

In this Rapid Communication, we present measurements of the thermal conductivity of $(C_5H_{12}N)_2CuBr_4$ between 0.3 and 10 K in magnetic fields up to 17 T. Surprisingly, no sign of spin-mediated heat transport is observed, which is interpreted by spinon localization. The heat transport is dominated by phonons being strongly scattered by spin excitations in the vicinity of the commensurate wave vector.

Two samples of $(C_5H_{12}N)_2CuBr_4$ of approximate dimensions $5\times 1.7\times 0.8$ mm^3 were cut from two crystals of the same batch with the longest dimension either along the a axis (along the ladders) or along the c^* axis. The thermal conductivity was measured using the standard uniaxial heat-flow method, where the temperature difference was produced by a heater attached to one end of the sample and monitored using a matched pair of RuO_2 thermometers. The heat flow was directed along the longest direction of each sample and the magnetic field was parallel to a.

The zero-field thermal conductivities $\kappa^\parallel, \kappa^\perp$ for both directions of the heat flow are shown in Fig. 1(a). Their behavior is typical for a phononic heat transport, approaching $\kappa \propto T^3$ at lowest temperature. The field dependencies of the thermal conductivity normalized to its zero-field value at several constant temperatures are presented in Fig. 1(b). The most pronounced features of the $\kappa(B)$ curves are two minima located in the middle of the field interval between B_{c1} and B_{c2} symmetrically with respect to $B_{\text{middle}}=(B_{c1}+B_{c2})/2 \simeq 10.8$ T. With decreasing temperature, the two minima become deeper and move closer to B_{middle}. It is noteworthy that the critical fields B_{c1} and B_{c2} are not marked by a distinct feature of $\kappa(B)$. As displayed in the inset of Fig. 1(b), $\kappa^\parallel(B)$ and $\kappa^\perp(B)$ show the same behavior.

The total heat conductivity of a magnetic insulator comprises the phonon and the spin contributions

$$\kappa = \kappa_s + \kappa_{\text{ph}}, \tag{1}$$

where each contribution is given, as a first approximation, by a product of the specific heat C_i, the average velocity v_i, and the mean free path ℓ_i of phonons (i=ph) and spin excitations

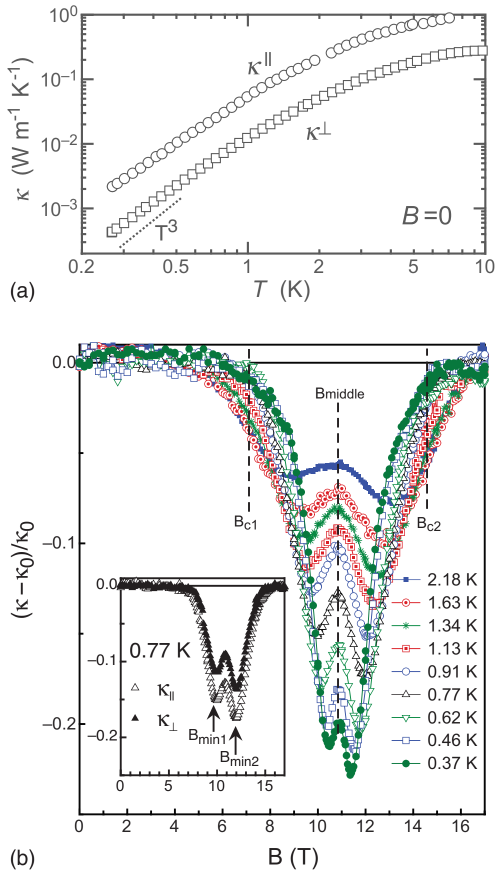

FIG. 1. (Color online) (a) Zero-field thermal conductivity of $(C_5H_{12}N)_2CuBr_4$ parallel and perpendicular to the ladder direction as a function of temperature. (b) Relative change in the thermal conductivity parallel to the ladders as a function of magnetic field at constant temperatures. Inset: data for $T=0.77$ K parallel and perpendicular to the ladders.

$(i=s)$. In addition, a term in Eq. (1) due to spin-phonon drag is expected.[15] In quasi-1D systems, both κ_s and spin-phonon drag are only essential along the chain (ladder) direction. From the almost absent anisotropy of $\kappa^{\parallel}(B)$ and $\kappa^{\perp}(B)$, we conclude that κ_s and the drag contribution are small.

The spin contribution $\kappa_s(T,B)$ can be analyzed using a mapping of the spin-$\frac{1}{2}$ ladder onto the effective spin-$\frac{1}{2}$ *XXZ* chain. Within this mapping, the ground-state singlet and the low-energy component of the triplet form an effective spin $\widetilde{S}=1/2$ on each rung of a ladder.[16–18] The effective Hamiltonian is

$$H_{XXZ} = J\sum_{i}^{N} (\widetilde{S}_x^i\widetilde{S}_x^{i+1} + \widetilde{S}_y^i\widetilde{S}_y^{i+1} + \delta\widetilde{S}_z^i\widetilde{S}_z^{i+1}) - g\mu_B\widetilde{B}\sum_{i}^{N}\widetilde{S}_z^i, \quad (2)$$

where δ is the anisotropy parameter. For a spin-$\frac{1}{2}$ ladder in the strong-coupling limit $(J_{\perp}/J_{\parallel}\gg 1)$, $\delta=1/2$, $J=J_{\parallel}$, and $\widetilde{B}=B-(J_{\perp}+J_{\parallel}/2)/g\mu_B$. Thus, $B=B_{\mathrm{middle}}$ corresponds to the zero effective field $\widetilde{B}=0$. The model Hamiltonian (2) describes very well many observed low-temperature features of $(C_5H_{12}N)_2CuBr_4$, as, e.g., the magnetization,[8] thermal-expansion,[9] NMR,[10] and specific-heat[12] measurements. Some discrepancies are mainly caused by the fact that the condition $J_{\perp}/J_{\parallel}\gg 1$ is only approximately satisfied.

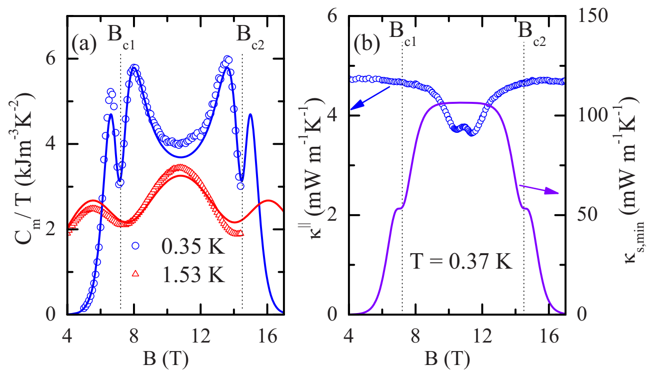

FIG. 2. (Color online) (a) B dependence of the magnetic specific heat of $(C_5H_{12}N)_2CuBr_4$. The symbols are experimental data (Ref. 12) and the lines are calculated within the MFT model (see text). (b) Comparison of the expected spin thermal conductivity $\kappa_{s,\mathrm{min}}$ (line) and the experimental total thermal conductivity $\kappa^{\parallel}$ along the ladders (symbols).

The heat transport in the spin-$\frac{1}{2}$ *XXZ* chain can be well described for $T<J/k_B$ within a relaxation-time approximation combined with a mean-field theory (MFT) approach via the Jordan-Wigner (JW) transformation. Equation (2) is mapped onto a system of interacting spinless fermions,[19,20] which occupy a cosine band

$$\varepsilon_k = -J(1+2\delta\Omega)\cos(ka) - g\mu_B\widetilde{B} + 2J\delta m. \quad (3)$$

Here, k is the wave vector, m is the magnetization, and a is the distance between neighboring spins. The parameters Ω and m are determined from $\Omega=\frac{a}{\pi}\int_0^{\pi/a}\cos(ka)f_k dk$ and $m=-\frac{1}{2}+\frac{a}{\pi}\int_0^{\pi/a}f_k dk$, where $f_k=[\exp(\varepsilon_k/k_BT)+1]^{-1}$ is the Fermi distribution function. The magnetic field plays the role of the chemical potential for the JW fermions. The spin thermal conductivity is given by

$$\kappa_s = \frac{Na}{\pi}\int_0^{\pi/a}\frac{df_k}{dT}\varepsilon_k v_k \ell_{s,k} dk, \quad (4)$$

where N is the number of spins per unit volume, $v_k=\hbar^{-1}d\varepsilon_k/dk$ is the velocity, and $\ell_{s,k}$ is the mean free path of the spin excitations. The applicability of the MFT JW-fermion model is demonstrated by the example of the specific heat $C_s(T,B)$, which is given by the right-hand side of Eq. (4), omitting $v_k\ell_{s,k}$ from the integrand. In Fig. 2(a), the calculated values of $C_s(B)$ are compared with the experimental data of Ref. 12 for $T=0.35$ and 1.53 K.[21] The good agreement between the calculated and measured $C_s(B)$ is obvious. Similarly, magnetostriction and thermal expansion of $(C_5H_{12}N)_2CuBr_4$ are well described by the JW-fermion model.[9]

In order to compare the measured thermal conductivity with the expected spin thermal conductivity, information about the mean free path ℓ_s is required. At low temperatures, scattering by magnetic impurities and disorder is dominant. The scattering by phonons should be rather weak in the present case because the concentration of phonons is much lower than the concentration of spin excitations, as the bandwidth of spinons ($\sim J_{\parallel} \simeq 3.6$ K) is much smaller than the phonon bandwidth (of the order of the Debye temperature $\theta_D \simeq 10^2$ K). In previous studies of the heat transport in spin-$\frac{1}{2}$-ladder[22] and spin-1-chain compounds,[23] the impurity-limited mean free path is found to be (k,B,T) independent and of the order of the mean distance between impurities in the ladder (chain) direction. Our analysis of the low-temperature magnetization data of similarly grown $(C_5H_{12}N)_2CuBr_4$ crystals yields an average distance of about 0.16 μm between "defective" rungs, i.e., rungs that are not in the singlet state for low fields. The corresponding calculated minimum spin thermal conductivity $\kappa_{s,\min}(B)$ is shown for $T=0.37$ K in Fig. 2(b) and compared with the measured total $\kappa^{\parallel}$. Near the critical fields, where the gap closes, $\kappa_s(B)$ should show a characteristic two-step increase from a negligibly small value to a constant of the order of 100 mW m^{-1} K^{-1}. Thus, one would expect an increase of κ_s that exceeds the measured total $\kappa=\kappa_s+\kappa_{ph}$ by more than one order of magnitude. Within the experimental resolution (about 0.1 mW m^{-1} K^{-1} at $T=0.37$ K), our $\kappa(B)$ data do not show any indication of such steps near either B_{c1} or B_{c2}. The absence of the expected steps cannot be simply explained by a very short mean free path in the studied crystal because $\kappa_s<0.1$ mW m^{-1} K^{-1} would correspond to an unphysically small $\ell<1$ Å. Thus, the thermal conductivity in $(C_5H_{12}N)_2CuBr_4$ is dominated by the phononic contribution $\kappa\approx\kappa_{ph}$. This conclusion is true for all temperatures we studied.

The absence of κ_s most likely results from impurity scattering, but due to the extremely weak interladder coupling this scattering is not consistent with a constant mean free path given by the distance between impurities. As suggested in Ref. 23, disorder leads to a constant mean free path of spin excitations only in the presence of weak yet non-negligible interladder coupling. In particular, the condition

$$\bar{v}_s^{\perp}/a > \bar{v}_s^{\parallel}/d_{def} \quad (5)$$

should be satisfied, where $\bar{v}_s^{\perp}$ and $\bar{v}_s^{\parallel}$ are the characteristic velocities of, respectively, the spin excitations perpendicular and parallel to the ladder direction, and d_{def} is the average distance between the defects. When Eq. (5) is satisfied, an energy transfer between neighboring ladders is possible and $\ell_s\approx d_{def}$. Otherwise, strong backscattering by impurities combined with the low probability of interladder transfer may lead to a spinon localization and thus $\kappa_s\approx 0$. In our recent heat transport studies in the spin-chain compounds copper pyrazine dinitrate and NENP,[23,24] the left-hand side of the inequality (5) was an order of magnitude larger than the right-hand side for the temperature range where a large l_s has been identified. However, in the present case of $(C_5H_{12}N)_2CuBr_4$ for all investigated temperatures, the estimates show that $\bar{v}_s^{\perp}/a \le \bar{v}_s^{\parallel}/d_{def}$. Therefore, we conclude that the absence of a measurable spin contribution to the heat transport results from a localization of the spin excitations in finite ladder segments of $(C_5H_{12}N)_2CuBr_4$.

Theoretical efforts treating localization effects in 1D systems have focused on the electrical conductivity.[25] Only recently random-disorder-induced localization in the spin thermal conductivity of the spin-$\frac{1}{2}$ XXZ chain has been specifically addressed by numerical diagonalization of small-size systems up to 20 spins.[26] For finite T, these results suggest the possibility of spinon localization for the easy-plane case [$\delta=0$ in Eq. (3)]. Similar calculations, which would be applicable for our case ($\delta=1/2$ and much longer d_{def}), have not been published so far.

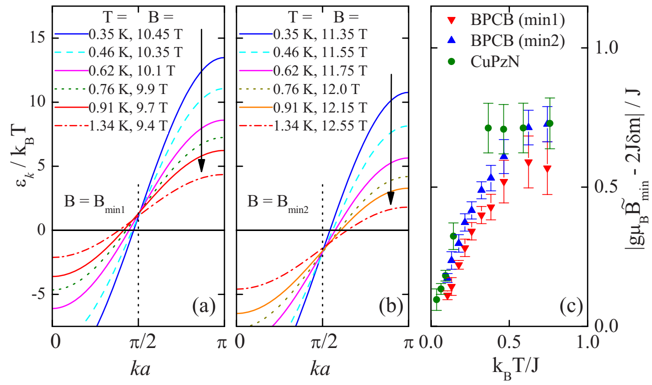

FIG. 3. (Color online) [(a) and (b)] Energy of the spin excitations normalized by k_BT as a function of the wave vector. The dispersion curves are calculated via Eq. (3) for T and B corresponding to the minima of $\kappa(B)$. The arrows indicate increasing T for $ka>\pi/2$. (c) Spin excitation energy at $ka=\pi/2$ for $B=B_{\min}$, calculated via Eq. (3), as a function of temperature for $(C_5H_{12}N)_2CuBr_4$ (BPCB) and copper pyrazine dinitrate (CuPzN). Both scales are normalized by the respective exchange couplings J (see text).

Now we turn to another salient feature of the measured $\kappa(B)$, namely, the two minima of $\kappa_{ph}(B)$ in the vicinity of B_{middle}. These minima are obviously caused by the scattering of phonons by spin excitations. In order to gain insight into what part of the spinon spectrum contributes to the scattering of phonons, one may use the dominant-phonon method. At low temperatures, when phonons are mainly scattered by sample boundaries (ℓ_{ph}=const), phonons with energy $\simeq 3.7k_BT$ provide most of the total thermal conductivity.[27] Thus, any additional phonon-scattering mechanism, which scatters phonons around a certain frequency ω_r, produces the strongest reduction of κ_{ph} at $T\simeq\hbar\omega_r/3.7k_B$. Hence, if particular spinons (in k space) are responsible for the minima of $\kappa(B_{\min},T)$, the energy of these spinons $\varepsilon_k(B_{\min},T)$ should scale linearly with temperature. Accordingly, we calculated the dispersion curves ε_k of the JW fermions for the (B,T) values of the lower ($B=B_{\min 1}$) and upper ($B=B_{\min 2}$) minima via Eq. (3) and normalized them by k_BT. The result is shown in Figs. 3(a) and 3(b). All calculated curves ε_k/k_BT practically meet at the commensurate point $ka=\pi/2$, suggesting that the spinons with $ka\simeq\pi/2$ are responsible for this additional phonon scattering.

This type of scattering seems not to be restricted to $(C_5H_{12}N)_2CuBr_4$. Minima of $\kappa(B)$ have also been observed in the spin-$\frac{1}{2}$ chain material copper pyrazine dinitrate.[24] Figure 3(c) compares the energies of the $ka=\pi/2$ JW fermions $\varepsilon_{\pi/2a}(B_{\min})=|-g\mu_B\widetilde{B}_{\min}+2J\delta m|$ [Eq. (3)], as a function of temperature for $(C_5H_{12}N)_2CuBr_4$ and for copper pyrazine dinitrate. Note that both axes have been normalized to J of the respective compound and that $\widetilde{B}\equiv B$ for the spin-$\frac{1}{2}$

chains. For both materials, we observe essentially the same behavior in the LL state, suggesting a common and universal origin of this additional phonon scattering, which is most likely associated with umklapp scattering. As discussed in Ref. 28, spinon umklapp scattering in the presence of weak disorder can lead to minima in the field dependence of the *spin* thermal conductivity $\kappa_s(B)$. However, the minima discussed in the present paper are observed in the *phonon* part of the heat conductivity κ_{ph}.

In conclusion, our measurements of the thermal conductivity in the two-leg spin-$\frac{1}{2}$ ladder compound $(C_5H_{12}N)_2CuBr_4$ provide clear evidence for the absence of spin-mediated heat transport. The most likely origin is a spinon localization in finite ladder segments, which is favored by the extremely weak interladder coupling, i.e., the high degree of one dimensionality, in $(C_5H_{12}N)_2CuBr_4$. The strongest effect observed in our experiments is a suppression of the phonon heat transport due to phonon-spinon scattering, which is exceptionally strong close to the commensurate filling of the spin excitation band.

We thank A. Rosch for many stimulating discussions and a critical reading of the paper. The contribution of H. R. Ott at the initial stage of the project is appreciated. We acknowledge useful discussions with C. Batista, M. Garst, P. Prelovšek, and E. Shimshoni. This work was supported by the Royal Society, by the Swiss National Science Foundation, and by the Deutsche Forschungsgemeinschaft through Sonderforschungsbereich 608.

*lorenz@ph2.uni-koeln.de

†Present address: Max Planck Institute for Chemical Physics of Solids, 01187 Dresden, Germany.

[1] F. Heidrich-Meisner, A. Honecker, and W. Brenig, Eur. Phys. J. Spec. Top. **151**, 135 (2007).

[2] A. V. Sologubenko, T. Lorenz, H. R. Ott, and A. Freimuth, J. Low Temp. Phys. **147**, 387 (2007).

[3] C. Hess, Eur. Phys. J. Spec. Top. **151**, 73 (2007).

[4] A. V. Sologubenko, K. Giannò, H. R. Ott, U. Ammerahl, and A. Revcolevschi, Phys. Rev. Lett. **84**, 2714 (2000).

[5] C. Hess, C. Baumann, U. Ammerahl, B. Büchner, F. Heidrich-Meisner, W. Brenig, and A. Revcolevschi, Phys. Rev. B **64**, 184305 (2001).

[6] K. Kudo, S. Ishikawa, T. Noji, T. Adachi, Y. Koike, K. Maki, S. Tsuji, and K. Kumagai, J. Phys. Soc. Jpn. **70**, 437 (2001).

[7] B. R. Patyal, B. L. Scott, and R. D. Willett, Phys. Rev. B **41**, 1657 (1990).

[8] B. C. Watson, V. N. Kotov, M. W. Meisel, D. W. Hall, G. E. Granroth, W. T. Montfrooij, S. E. Nagler, D. A. Jensen, R. Backov, M. A. Petruska, G. E. Fanucci, and D. R. Talham, Phys. Rev. Lett. **86**, 5168 (2001).

[9] T. Lorenz, O. Heyer, M. Garst, F. Anfuso, A. Rosch, C. Rüegg, and K. Krämer, Phys. Rev. Lett. **100**, 067208 (2008).

[10] M. Klanjšek, H. Mayaffre, C. Berthier, M. Horvatić, B. Chiari, O. Piovesana, P. Bouillot, C. Kollath, E. Orignac, R. Citro, and T. Giamarchi, Phys. Rev. Lett. **101**, 137207 (2008).

[11] F. Anfuso, M. Garst, A. Rosch, O. Heyer, T. Lorenz, C. Rüegg, and K. Krämer, Phys. Rev. B **77**, 235113 (2008).

[12] Ch. Rüegg, K. Kiefer, B. Thielemann, D. F. McMorrow, V. S. Zapf, B. Normand, M. B. Zvonarev, P. Bouillot, C. Kollath, T. Giamarchi, S. Capponi, D. Poilblanc, D. Biner, and K. W. Krämer, Phys. Rev. Lett. **101**, 247202 (2008).

[13] B. Thielemann, Ch. Rüegg, K. Kiefer, H. M. Rønnow, B. Normand, P. Bouillot, C. Kollath, E. Orignac, R. Citro, T. Giamarchi, A. M. Läuchli, D. Biner, K. W. Krämer, F. Wolff-Fabris, V. S. Zapf, M. Jaime, J. Stahn, N. B. Christensen, B. Grenier, D. F. McMorrow, and J. Mesot, Phys. Rev. B **79**, 020408(R) (2009).

[14] B. Thielemann, Ch. Rüegg, H. M. Rønnow, A. M. Läuchli, J.-S. Caux, B. Normand, D. Biner, K. W. Krämer, H.-U. Güdel, J. Stahn, K. Habicht, K. Kiefer, M. Boehm, D. F. McMorrow, and J. Mesot, Phys. Rev. Lett. **102**, 107204 (2009).

[15] E. Boulat, P. Mehta, N. Andrei, E. Shimshoni, and A. Rosch, Phys. Rev. B **76**, 214411 (2007).

[16] K. Totsuka, Phys. Rev. B **57**, 3454 (1998).

[17] F. Mila, Eur. Phys. J. B **6**, 201 (1998).

[18] G. Chaboussant, M. H. Julien, Y. Fagot-Revurat, M. Hanson, L. P. Levy, C. Berthier, M. Horvatic, and O. Piovesana, Eur. Phys. J. B **6**, 167 (1998).

[19] F. Heidrich-Meisner, A. Honecker, and W. Brenig, Phys. Rev. B **71**, 184415 (2005).

[20] F. Heidrich-Meisner, Ph. D. thesis, Technische Universität Braunschweig, 2005.

[21] The calculations were done with the parameters $g=2.06$, $J_{\parallel}=3.5$ K, $J_{\perp}=13.2$ K, $N=1.120\times10^{27}$ m^{-3} (number of rungs per unit volume), and $a=8.5$ Å.

[22] C. Hess, P. Ribeiro, B. Büchner, H. ElHaes, G. Roth, U. Ammerahl, and A. Revcolevschi, Phys. Rev. B **73**, 104407 (2006).

[23] A. V. Sologubenko, T. Lorenz, J. A. Mydosh, A. Rosch, K. C. Shortsleeves, and M. M. Turnbull, Phys. Rev. Lett. **100**, 137202 (2008).

[24] A. V. Sologubenko, K. Berggold, T. Lorenz, A. Rosch, E. Shimshoni, M. D. Phillips, and M. M. Turnbull, Phys. Rev. Lett. **98**, 107201 (2007).

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

[26] A. Karahalios, A. Metavitsiadis, X. Zotos, A. Gorczyca, and P. Prelovšek, Phys. Rev. B **79**, 024425 (2009).

[27] R. Berman, *Thermal Conduction in Solids* (Clarendon Press, Oxford, 1976).

[28] E. Shimshoni, D. Rasch, P. Jung, A. V. Sologubenko, and A. Rosch, Phys. Rev. B **79**, 064406 (2009).