Anomalous Thermal Conductivity of Frustrated Heisenberg Spin Chains and Ladders

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We study the thermal transport properties of several quantum-spin chains and ladders. We find indications for a diverging thermal conductivity at finite temperatures for the models examined. The temperature at which the nondiverging prefactor $\kappa^{(\mathrm{th})}(T)$ peaks is, in general, substantially lower than the temperature at which the corresponding specific heat $c_V(T)$ is maximal. We show that this result of the microscopic approach leads to a substantial reduction for estimates of the magnetic mean-free path λ extracted by analyzing recent experiments, as compared to similar analyses by phenomenological theories.

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Introduction.—The nature of thermal transport in systems with reduced dimensions is a long-standing problem and has been studied intensively in classical [1] systems either by direct numerical out-of-equilibrium simulations [2] or by investigation of soliton-soliton scattering processes [3]. There has been, on the other hand, considerably less progress for quantum systems. Huber, in one of the firsts works on the subject [4], evaluated the thermal conductivity $\kappa(T)$ for the Heisenberg chain with an equation-of-motion approximation and found a *finite* $\kappa(T)$, a result which is, by now, known to be wrong. It has been shown recently [5] that the energy-current operator commutes with the Hamiltonian for the spin-1/2 Heisenberg chain. The thermal conductivity is consequently *infinite* for this model. The intriguing question, "Under which circumstances does an interacting quantum system show an infinite thermal conductivity?", is until now completely open, the analogous question for classical systems being intensively studied [1].

A second motivation to study $\kappa(T)$ for quantum-spin systems comes from experiment. An anomalous large magnetic contribution to κ has been observed [6] for the hole-doped spin-ladder system $Sr_{14-x}Ca_xCu_{24}O_{41}$. For $Ca_9La_5Cu_{24}O_{41}$, which has no holes in the ladders, an even larger thermal conductivity has been measured [7] raising the possibility of ballistic magnetic transport limited only by residual spin-phonon and impurity scattering. There is, however, until now no microscopic calculation for the thermal conductivity of spin ladders.

In this Letter, we present a finite-size analysis of the thermal conductance in Heisenberg $J_1 - J_2$ chains and ladders suggesting ballistic thermal transport in these two families of spin models. Following this result, we propose a possible scenario that would account for a number of anomalies found in the thermal conductivity in spin systems. We apply these ideas to the case of $Ca_9La_5Cu_{24}O_{41}$, obtaining good agreement with recent experimental measurements.

Models.—We consider quasi-one-dimensional systems for which the Hamiltonian takes the form $H = \sum_{x=1}^{L} H_x$, where L is the number of units along the chain. We will consider two models, the isotropic Heisenberg chain with dimerized nearest and homogeneous next-nearest neighbor exchange couplings,

$$H_x^{(\mathrm{ch})} = J\{[1 + \delta(-1)^x]\mathbf{S}_x \cdot \mathbf{S}_{x+1} + \alpha\, \mathbf{S}_x \cdot \mathbf{S}_{x+2}\},$$

and the two-leg Heisenberg ladder:

$$H_x^{(\mathrm{lad})} = J_{\|}(\mathbf{S}_{1,x} \cdot \mathbf{S}_{1,x+1} + \mathbf{S}_{2,x} \cdot \mathbf{S}_{2,x+1}) + J_{\perp}\mathbf{S}_{1,x} \cdot \mathbf{S}_{2,x}.$$

$H^{(\mathrm{ch})}$ has been proposed to model the magnetic properties of the spin-Peierls compound $CuGeO_3$. The nonfrustrated dimerized Heisenberg chain ($\alpha = 0$) models the magnetic behavior [8] of $(VO)_2P_2O_7$. $Ca_9La_5Cu_{24}O_{41}$ contains doped spin chains and undoped spin ladders [7], described by $H^{(\mathrm{lad})}$.

Method.—Our goal is to make a connection between the microscopic transport properties of the low dimensional models presented above and the experimental measurements of thermal conductivity mentioned in the introduction.

The thermal conductivity is defined as the response of the energy-current density $\mathbf{j}_x^E$ to a thermal gradient $\mathbf{j}_x^E = -\kappa\nabla T$, and it has units of $[\kappa] = \frac{W}{\mathrm{K\,m}^{d-2}}$, where d is the dimension ($d = 3$ for experiments). The specific form of the energy-current associated with a given Hamiltonian is determined by the continuity equation for the energy density, $\dot{H}_x + \nabla \cdot j_x^E = 0$, which leads via $\nabla \cdot j_x^E = (j_{x+1}^E - j_x^E)/c$ to [9]

$$[H_x, H_{x+1}] + [H_x, H_{x+2}] + [H_{x-1}, H_{x+1}] = \frac{i\hbar}{c}\, j_x^E,$$

where c is the lattice constant along the chain direction.

In the absence of applied magnetic fields, spin inversion symmetry holds, and the Kubo formula for $\kappa = \lim_{\omega\to 0} \kappa(\omega)$ reduces to [4]

$$\kappa = \lim_{\omega\to 0} \frac{\beta^2}{\mathrm{L}} \int_0^\infty dt e^{t(i\omega - s)} \langle J^E(t) J^E \rangle, \qquad (1)$$

where $J^E = \sum_x \mathbf{j}_x^E$ is the total energy current, $\beta = 1/(k_B T)$ is the inverse temperature, and $s \to 0$ the usual convergence factor.

We will examine here the possibility of infinite intrinsic thermal conductivity. For such a system heat transport is ballistic and energy is transported without dissipation. As an example, we consider the XXZ chain ($H^{(\mathrm{ch})}$ with $\alpha = 0 = \delta$ and a spin anisotropy). The total thermal current commutes with the Hamiltonian in this case [5] and the current-current correlation function is therefore time independent. Equation (1) reduces then to the static expectation value $\kappa(\omega = 0) = \beta^2\langle(J^E)^2\rangle/(Ls)$, a quantity which can be evaluated by the Bethe ansatz [10].

In general, we have $[J^E, H] \neq 0$, but the thermal transport will be ballistic if

$$\kappa^{(\mathrm{th})} = \frac{\beta^2}{ZL} \sum_{m,n;E_n=E_m} e^{-\beta E_m} |\langle m|J^E|n\rangle|^2 \qquad (2)$$

is finite in the thermodynamic limit $L \to \infty$. The thermal conductivity $\kappa = \kappa^{(\mathrm{th})}/s$ then diverges for $s \to 0$; as it does for the XXZ chain.

In reality, spin Hamiltonians such as $H^{(\mathrm{ch})}$ and $H^{(\mathrm{lad})}$ are coupled to an external environment, e.g., to phonons or impurities. Here we consider the case where this coupling is small. This coupling will then result in a finite *external* lifetime $\tau = 1/s$ for the eigenstates of the spin model [which is assumed to be energy independent in Eq. (1)]. Using the relation $\lambda = v_s \tau$ in between the *external* mean-free path λ, the spin-wave velocity v_s, and τ, we propose

$$\kappa = \kappa^{(\mathrm{th})}\, \tau = \kappa^{(\mathrm{th})} \, \frac{\lambda}{v_s}, \qquad (3)$$

to hold for the thermal conductivity κ.

Let us discuss the relation of Eq. (3) to the usual phenomenological formula [11] (here in one dimension)

$$\kappa^{(\mathrm{ph})} = c_V\, v_s \lambda, \qquad (4)$$

where c_V is the specific heat. When applied in order to analyze experimental data, the microscopic Eq. (3) will, in general, yield different values for the magnetic mean-free path λ. On the other hand, we might expect Eq. (4) to hold at low temperature for the Heisenberg chain in the gapless phase, when the Luttinger-liquid quasiparticles are well-defined and a Boltzmann approach is justified. Consequently, we expect $\kappa^{(\mathrm{th})}\, \lambda/v_s \equiv c_V v_s \lambda$ in this case, i.e., we expect $\kappa^{(\mathrm{th})}/c_V \equiv v_s^2$ in the limit $T \to 0$. All three quantities in this equation ($\kappa^{(\mathrm{th})}$, c_V, and v_s) can be computed by Bethe ansatz for the XXZ chain. One finds that this equation is exact [10] in the limit $T \to 0$.

Numerics.—The computation of $\kappa^{(\mathrm{th})}(T)$ demands the whole spectrum of the Hamiltonian, restricting the maximum lattice size and a careful finite-size analysis is required. In general, one finds, e.g., for the XXZ model for which the exact Bethe ansatz result is known [10], that the $\kappa^{(\mathrm{th})}$ in chains with odd (even) number of sites chains is an upper (lower) bound to the exact result. The value of $\kappa^{(\mathrm{th})}(T)$ decreases (increases) for chains with an odd (even) number of sites when the lattice is enlarged. This observation has led us to consider the average,

$$\kappa^{(\mathrm{th})}(L_{\mathrm{eff}}) = \frac{L_0 \kappa^{(\mathrm{th})}(L_0) + L_1 \kappa^{(\mathrm{th})}(L_1)}{L_0 + L_1} \qquad (5)$$

of finite-size data, where $L_0 = 2i$, $L_1 = 2i + 1$, and $L_{\mathrm{eff}} = (L_0 + L_1)/2$. We find that $\kappa^{(\mathrm{th})}(L_{\mathrm{eff}})$ converges to the thermodynamic limit somewhat faster than taking the limit for and even (or odd) number of sites only, see Fig. 1. This average technique is reminiscent of the boundary-condition integration technique [12] for exact diagonalization studies.

The situation is similar when boundary conditions are changed from periodic to antiperiodic and for the ladders when the number of rungs changes from odd to even. We also find that (5) produces excellent results when applied to the specific heat, as illustrated in Fig. 2, where we compare the exact diagonalization data for ladders with quantum Monte Carlo (QMC) results [13].

Results—After these technical remarks, we discuss now the exact diagonalization results for the temperature dependence of $\kappa^{(\mathrm{th})}$. The general features in all the models studied are as follows: (i) a finite value of $\kappa^{(\mathrm{th})}$ at any finite temperature that does not vanish when the extrapolation to infinite lattice size is taken; (ii) a single maximum $\kappa^{(\mathrm{th})}_{\mathrm{max}}$ at an intermediate temperature $T_{\mathrm{max}}(\kappa)$ smaller than the maximum in the specific heat $T_{\mathrm{max}}(C_v)$; (iii) the high temperature regime follows, in general, the law $C(L)/T^2$, as expected from Eq. (2).

The results for $H^{(\mathrm{lad})}$ are presented for $J_\perp = 2J_\parallel$ in Fig. 2. We notice that the results for the specific heat are already converged nicely, albeit the small effective chain length used. We believe that the results for $\kappa^{(\mathrm{th})}$ shown in

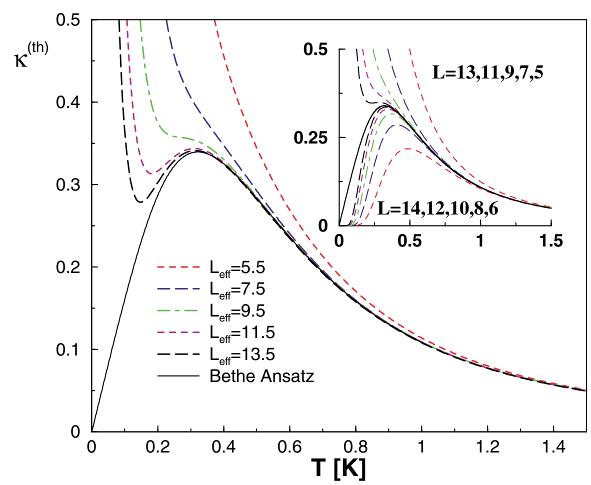

FIG. 1 (color online). $\kappa^{(\mathrm{th})}$, as defined by Eq. (2), for the Heisenberg chain as a function of temperature. Inset: Data for $L = 5$–14, in comparison with the Bethe ansatz result [10] (full line). Main panel: Results using Eq. (5), with $L_{\mathrm{eff}} = (L_0 + L_1)/2$. We note that the position of the maximum in $\kappa^{(\mathrm{th})}$ can be determined confidently with the averaging procedure.

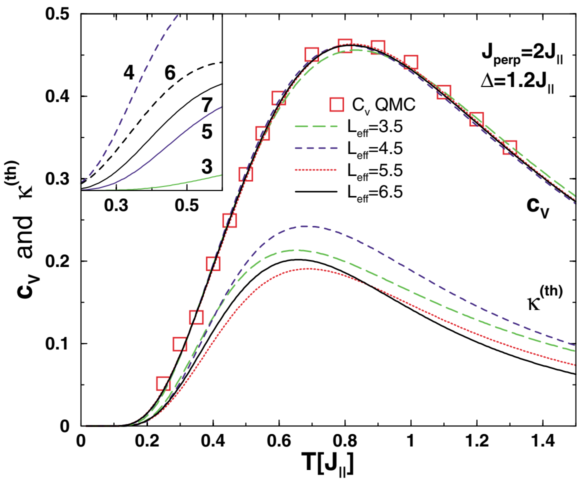

FIG. 2 (color online). Temperature dependence of $\kappa^{(\mathrm{th})}$ and the dimensionless specific heat for ladders with twisted boundary conditions and $J_\perp = 2J_\|$. Both magnitudes are computed using the average procedure (5) described in the text. The specific heat for a 2×100-ladder computed with QMC is plotted for comparison (squares). Inset $\kappa^{(\mathrm{th})}$ for $L = 3, 5, 7$ (from bottom up) and $L = 4, 6$ (from top down) in the low-temperature region.

Fig. 2 to be accurate to about 10%, enough to allow a detailed analysis of the experimental data which we will perform further below. In the inset of Fig. 2 we present a blowup for the data for $\kappa^{(\mathrm{th})}$ at low temperatures for $L = 3, \ldots, 7$. We note the systematic increase of $\kappa^{(\mathrm{th})}$ with increasing L for the odd values $L = 3, 5, 7$, indicating a finite value in the thermodynamic limit. The even values $L = 4, 6$ seem to constitute upper bounds to $\kappa^{(\mathrm{th})}$.

In Fig. 3, we present the results for $H^{(\mathrm{ch})}$, $\alpha = 0.35$ and $\delta = 0$. We have also studied the dimerized phase with

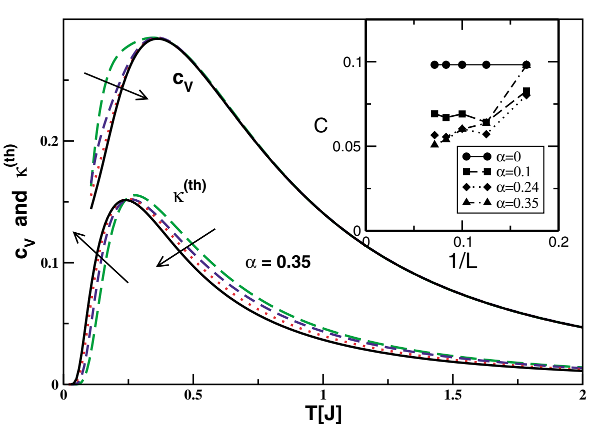

FIG. 3 (color online). $\kappa^{(\mathrm{th})}$ and dimensionless specific heat for the frustrated Heisenberg chain with $\alpha = 0.35$ and different lattice sizes $L = 8, 10, 12, 14$, the arrows indicate increasing L, compare [9]. Inset : Finite size analysis ($L = 6$–14) of the high temperature residue $C(L)$ defined as $\kappa^{(\mathrm{th})} = C(L)/T^2$ at $T \gg J$ for different values of α.

$\delta > 0$ and found similar behaviors. The maximum value of $\kappa^{(\mathrm{th})}$ is nearly size independent for $L = 8$–14. The raise of $\kappa^{(\mathrm{th})}$ with increasing system size L (as indicated by the arrows in Fig. 3) in the low-temperature regime, is consistent with the notion of a finite $\kappa^{(\mathrm{th})}$ in the thermodynamic limit.

The way finite-size chains approach the $L \to \infty$ limit is almost identical for frustrated chains and the exactly solvable $\alpha = 0$ case, as exemplified by the size dependence for the prefactor of the leading $1/T^2$ term presented in the inset of Fig. 3, showing the reliability of the finite-size analysis. We can conclude that the frustration produces a substantial drop in the *extrapolated* finite value of the prefactor.

Analysis.—Comparison with experimental results for the thermal conductivity $\kappa^{(\exp)}$ can be made using the dimensional analysis:

$$\kappa^{(\exp)} = \frac{k_B J}{\hbar c}\left(\frac{N_c c^3}{abc}\right)\left(\frac{\lambda}{c}\right)\left(\frac{Jc}{\hbar v_s}\right)\tilde{\kappa}^{(\mathrm{th})}, \tag{6}$$

where the quantities in the brackets are dimensionless. $\tilde{\kappa}^{(\mathrm{th})}$ is the dimensionless thermal conductance (2) which we will evaluate by exact diagonalization. a, b, and c are the lattice constants (the chains run along the c direction) and N_c is the number of chains per unit cell. Note that $(\frac{\lambda}{c})(\frac{Jc}{\hbar v_s}) = (\frac{J\tau}{\hbar})$ and (6) can be used to extract the lifetime τ (in units of the coupling constant J) directly by comparison with the experimental $\kappa^{(\exp)}$.

We now analyze the experimental data [7] for $Ca_9La_5Cu_{24}O_{41}$. A quantitative good description of the magnetic excitations in $La_9La_5Cu_{24}O_{41}$ can be obtained by $H^{(\mathrm{lad})}$ with the inclusion of an additional ring-exchange term [14] (a 15% correction) and $J_\perp \simeq J_\|$ [14,15]. Here we disregard the possible ring-exchange term and use $J \equiv J_\perp = J_\| = 832.6$ K, which we extracted by fitting the magnetic contribution [7] $\kappa^{(\exp)}(T)$ by $444.2/[1.0 + 0.093\exp(419.6/T) + (T/156.4)^2]$ (a very good fit at all temperatures, see inset of Fig. 4). The gap Δ for the isotropic Heisenberg ladder is [16] $\Delta = 0.504J_\perp$, which leads for $Ca_9La_5Cu_{24}O_{41}$ to $J = 419.6/0.504$ K $= 832.6$ K.

Using the appropriate lattice constants and $N_c = 14$ (rungs per unit cell [17]) we used Eqs. (5) and (6) for 2×6 and 2×7 ladders in order to extract the spin-environment relaxation time τ, see Fig. 4. We find the following for the lifetime τ (in units of the coupling constant J): $\frac{\tau(T)\hbar}{J} \simeq 7[\frac{725}{T} + (\frac{561}{T})^2]$. The assumption of a weak spin-environment coupling entering Eq. (3) is therefore justified in the experimental relevant temperature regime. The lifetime is, to give an example, 132 times larger than the coupling constant J at $T = 150$ K, leading to an external broadening of the energy levels of only $1/132 = 0.76\%$, in units of J.

To estimate the effective mean-free path, we use $\lambda(T) = \bar{v}_s(T)\tau(T)$, where we have used for $\bar{v}_s$ the thermal

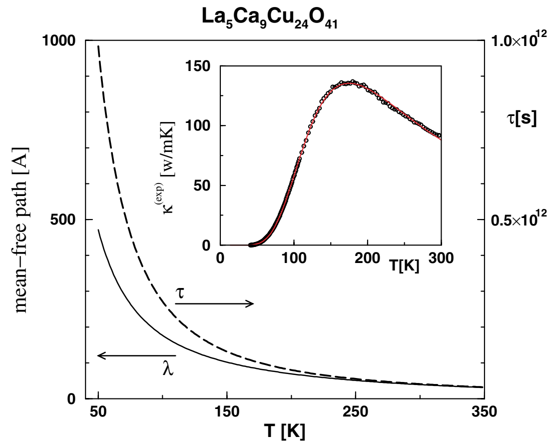

FIG. 4 (color online). $\tau(T)$ (right scale) and $\lambda(T)$ (left scale) for $Ca_9La_5Cu_{24}O_{41}$ extracted using Eq. (6). The experimental data by Hess *et al.* [7] is shown in the inset (filled circles) together with an analytic fit (solid line) discussed in the text.

expectation value of the magnon velocity within the independent-triplet model:

$$\bar{v}_s = \frac{1}{Z}\int dk\varepsilon'(k)n[\varepsilon(k)] = \frac{1}{Z}\int_{\Delta}^{\Delta_2} d\varepsilon n(\varepsilon),$$

where $Z = \int dkn[\varepsilon(k)]$ is the partition function and $n[\varepsilon(k)] = 3/(\exp[\beta\varepsilon(k)] + 3)$. We approximated the one-magnon dispersion $\varepsilon(k)$ by $2\,\varepsilon^2(k) = (\Delta^2 + \Delta_2^2) + (\Delta_2^2 - \Delta^2)\cos(k)$. For $J_\perp = J_\parallel$, we have [16] that $\Delta_2 \approx 3.8\Delta$. In the low-temperature limit, $\bar{v}_s(T) \approx \sqrt{T}$ holds and the mean-free path $\lambda(T)$ is therefore less divergent at low T than the relaxation-time $\tau(T)$ [see Fig. 4]. At very low temperature, we expect impurity scattering to become relevant and $\lambda(T)$ to plateau off.

Discussion.—Our results for the mean-free length $\lambda(T)$ for $Ca_9La_5Cu_{24}O_{41}$ obtained by the microscopic formula (3) are substantially smaller than the one obtained using (4). At $T = 100$ K, Hess *et al.* [7] estimated a large $\lambda \approx 3000$ Å, in contrast to our result $\lambda(100) \approx 176$ Å. This is, in a certain sense, surprising, since the effective λ obtained from (4) might be thought to contain additional spin-spin scattering. Physically, the reason for our smaller mean-free path stems from the fact that $\kappa^{(\mathrm{th})}(T)$ peaks at substantially smaller temperatures than $c_V(T)$, see Fig. 3. We note that all excitations contribute to c_V on an equal footing, but that magnetic excitations near the bottom of the one-magnon band seem to contribute dominantly to the thermal conductivity, leading to a substantial reduction of the temperature where $\kappa^{(\mathrm{th})}(T)$ is maximal, with respect to $c_V(T)$.

Conclusions.—We have presented a microscopic approach of thermal transport in quasi-one-dimensional spin models. For several models, we find indications for a diverging thermal conductivity. A finite thermal conductivity is obtained when couplings to external degrees of freedom are taken into account with the relaxation-time approximation. We have analyzed recent experiments for spin-ladder compounds and found only weak spin-environment coupling for the ladder compounds, which decreases fast with decreasing temperature. Our estimates for the mean-free length turned out to be substantially smaller than previous estimates using simple phenomenological formulas. These results highlight the importance of microscopic theories for transport in quasi-one-dimensional quantum-spin systems.

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