High Temperature Thermal Conductivity of Two-Leg Spin-1/2 Ladders

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Based on numerical simulations, a study of the high temperature, finite frequency, thermal conductivity $\kappa(\omega)$ of spin-1/2 ladders is presented. The exact diagonalization and a novel Lanczos technique are employed. The conductivity spectra, analyzed as a function of rung coupling, point to a nondiverging dc limit but to an unconventional low frequency behavior. The results are discussed in perspective with recent experiments indicating a significant magnetic contribution to the energy transport in quasi-one-dimensional compounds.

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Introduction.—Recent experiments [1–5] convincingly promoted magnetic excitations as a very efficient mechanism for energy transport in quasi-one-dimensional materials. In particular, spin-1/2 Heisenberg chain and ladder compounds (undoped or hole doped) have been studied. In both systems, the observed highly anisotropic thermal conductivity (comparable in magnitude to that of metallic systems) was attributed to magnetic transport.

Theoretically, it has been noticed that in the one-dimensional (1D) spin-1/2 Heisenberg model the energy current commutes with the Hamiltonian and thus the thermal conductivity is ballistic at all temperatures [6–8]. This observation falls in line with a proposal of unconventional transport in 1D integrable systems [8]. Besides this rigorous result, the experimental developments motivated theoretical studies of the thermal conductivity in other (e.g., frustrated, gapped) spin chain and ladder Hamiltonians based on numerical simulations [9–11] or low energy effective theories [12,13]. These works focused on the thermal Drude weight D_{th} as a criterion of ballistic transport [8,14,15], and they led to an active discussion of whether nonintegrable systems also show ideal thermal conductivity.

The key to obtaining reliable information by numerical simulations is the study of large enough systems that allow analysis of the scaling with lattice size. In this work, we study the thermal conductivity within linear response theory, first, at high temperatures where numerical simulations on finite systems are most reliable (the characteristic thermal scattering length should be smaller than the size of the lattice) and, second, by using a novel technique based on the Lanczos method and the microcanonical ensemble (MCLM [16]). This method gives access to fairly larger systems than those studied until now by the exact diagonalization (ED) method.

The main issues of actual experimental and theoretical interest that we address are (i) whether the thermal transport is ballistic or diffusive, (ii) how the interchain coupling affects the ideal thermal transport of the decoupled (integrable) Heisenberg spin-1/2 chains, and (iii) the order of magnitude of the magnetic contribution to the thermal conductivity as a function of exchange couplings and, in particular, how it compares with experimental values in ladder compounds.

Hamiltonian and method.—The two-leg ladder Hamiltonian is given by the $q=0$ component of H_q, the Fourier transform of the local energy density, which we define as

$$H_q = J\sum_{l=1,L}\Big\{e^{iq(l+1/2)}(\mathbf{S}_{1,l+1}\cdot\mathbf{S}_{1,l}+\mathbf{S}_{2,l+1}\cdot\mathbf{S}_{2,l}) + \frac{J_\perp}{J}e^{iql}\mathbf{S}_{1,l}\cdot\mathbf{S}_{2,l}\Big\}. \tag{1}$$

$\mathbf{S}_l$ are spin-1/2 operators at site l and, in the following, we consider systems with periodic boundary conditions; we take $J=1$ as the unit of energy ($\hbar = k_B = 1$) and unit lattice constants.

To study the thermal conductivity, we define the energy current operator j^E using the continuity equation for H_q [17,18]. Extracting the long-wavelength transport limit, $\partial H_q/\partial t \sim -iqj^E$ for $q\to 0$, we obtain

$$j^E = J^2 i\sum_{l=1,L}\{\mathbf{S}_{1,l-1}\cdot(\mathbf{S}_{1,l}\times\mathbf{S}_{1,l+1})+\mathbf{S}_{2,l-1}\cdot(\mathbf{S}_{2,l}\times\mathbf{S}_{2,l+1})\} + \frac{J_\perp}{2J}\{\mathbf{S}_{1,l-1}\cdot(\mathbf{S}_{1,l}\times\mathbf{S}_{2,l})+\mathbf{S}_{2,l-1}\cdot(\mathbf{S}_{2,l}\times\mathbf{S}_{1,l})+\mathbf{S}_{2,l}\cdot(\mathbf{S}_{1,l}\times\mathbf{S}_{1,l+1})+\mathbf{S}_{1,l}\cdot(\mathbf{S}_{2,l}\times\mathbf{S}_{2,l+1})\}. \tag{2}$$

Within linear response theory [19], the real part of the thermal conductivity at frequency ω is given by

$$\kappa(\omega) = 2\pi D_{\text{th}}\delta(\omega) + \kappa_{\text{reg}}(\omega), \tag{3}$$

with the regular part,

$$\kappa_{\text{reg}}(\omega>0) = \frac{\beta}{\omega L}\tanh\Big(\frac{\beta\omega}{2}\Big)\Im i\int_0^{+\infty}dt\, e^{izt}\langle\{j^E(t), j^E\}\rangle, \tag{4}$$

($\beta = 1/T$, $z = \omega + i\eta$) and the thermal Drude weight,

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$$D_{\rm th} = \frac{\beta^2}{2L} \sum_{\substack{n,m \\ \epsilon_n=\epsilon_m}} p_n |\langle n|j^E|m\rangle|^2. \quad (5)$$

$|n\rangle$ (ϵ_n) are the eigenstates (eigenvalues) and p_n the Boltzmann weights. For $\beta \to 0$ we obtain the sum rule,

$$\int_{-\infty}^{+\infty} d\omega\, \kappa(\omega) = \frac{\pi\beta^2}{L}\langle j^{E2}\rangle = I, \quad (6)$$

which suggests the analysis of $2\pi D_{\rm th}/I$ in order to estimate the contribution of ballistic transport.

In the following, we employ the ED method to study $D_{\rm th}$ and $\kappa(\omega)$ for systems up to $L = 9$ rungs. For larger systems, up to $L = 14$, we use the MCLM method. In both cases we implement the translational symmetry that provides independent k Hamiltonian subspaces. In the MCLM method [16], we replace the thermal average in (4) by the expectation value over a single state $|\lambda\rangle$ of energy λ that equals the canonical ensemble value of the energy at the desired temperature [20], $\lambda = \langle H\rangle$. In most cases, we present results for $\beta \to 0$, where $\lambda \sim 0$ due to the symmetric spectrum of the Hamiltonian. Otherwise, we determine λ by extrapolation of thermal energies evaluated using ED results.

The state $|\lambda\rangle$ is constructed by employing a first Lanczos procedure of about 1000 steps using as "effective Hamiltonian" the operator $K = (H - \lambda)^2$; in practice, for large systems with dense spectra, the procedure cannot fully converge. $|\lambda\rangle$, the ground state of K in the constructed Lanczos subspace, is characterized by a distribution over the eigenstates $|n\rangle$ of variance in energy of $O(0.01)$ that imposes a maximum ω resolution to the spectra. Then, starting from $j^E|\lambda\rangle$, a second Lanczos procedure of about 4000 steps provides $\kappa(\omega)$ using the continued fraction technique [21] (typically $\eta \sim 0.01$). Thus, we effectively evaluate $\kappa(\omega)$ by

$$\kappa(\omega) \mapsto \frac{\beta}{\omega L}\tanh\left(\frac{\beta\omega}{2}\right)(-\Im)\left(\langle\lambda|j^E\frac{1}{z-(H-\lambda)}j^E|\lambda\rangle + \langle\lambda|j^E\frac{1}{z+(H-\lambda)}j^E|\lambda\rangle\right). \quad (7)$$

Note that, as the Lanczos procedures do not fully converge, an eventual $\delta(\omega)$-peak contribution appears as a broadened weight at very low frequencies [16].

Thermal conductivity.—The first step in characterizing $\kappa(\omega)$ is the evaluation of the Drude weight at finite T. If $D_{\rm th}$ is finite then the transport is ballistic; if it vanishes, the transport is normal provided the $\kappa_{\rm dc} = \kappa(\omega \to 0)$ limit exists. For a finite system $D_{\rm th}$ is always nonzero, so it is crucial to examine its scaling as a function of size. Exact results obtained by the ED method are shown in Fig. 1. In the high temperature limit $D_{\rm th}/\beta^2$ rapidly decreases, seemingly exponentially fast, with system size (for each series of even or odd L). It already represents only a couple of percent of the sum rule for $L = 9$ rungs (inset). Of course, the size of the studied lattices is rather limited, but a vanishing Drude weight in the $\beta \to 0$ limit is corroborated by the $\kappa(\omega)$ spectra shown below.

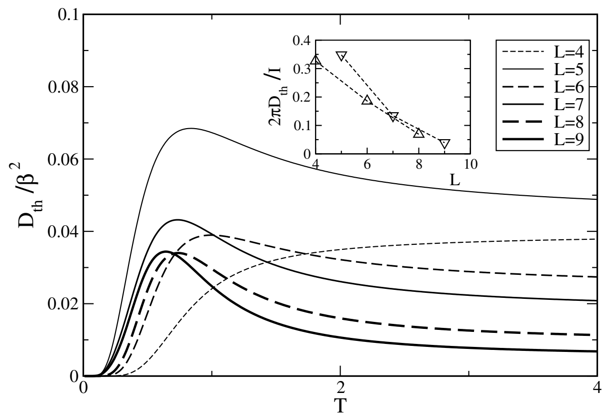

FIG. 1. Thermal Drude weight for $J_\perp = 1$ as a function of temperature for systems with L rungs (ED evaluation). Inset: Normalized $D_{\rm th}$ for $\beta \to 0$ as a function of system size.

At lower temperatures, we find a nonmonotonic scaling for even-rung systems. Note, however, that the change of scaling behavior of $D_{\rm th}$, from decreasing with increasing L at high T to the opposite at low T, shifts to a lower temperature as we consider larger systems (see also [11]). We can then argue that over the whole temperature range the Drude weight scales to zero as $L \to \infty$ (in agreement with [10]). It would be unexpected if there is a transition to a finite $D_{\rm th}$ below some critical temperature.

Next, we discuss the thermal conductivity as a function of ω and L in an attempt to determine whether it represents normal transport (diffusive behavior exemplified by a Lorentzian form), unconventional (e.g., power law at low frequencies), or some other behavior that still implies a finite dc conductivity. Furthermore, we study the dependence on $J_\perp$ in order to find out the effect of interchain coupling on the ballistic transport of decoupled chains. For $J_\perp = 0$, only the $2\pi\delta(\omega)D_{\rm th}$ contribution exists as $[j^E, H] = 0$. In Fig. 2, we show a series of spectra for different L, $J_\perp = 1$, and $\beta \to 0$. To illustrate the applicability of the MCLM method, we compare a spectrum from a canonical ensemble ED study for $L = 8$ (weights smoothed with Lorentzians) with one obtained by the MCLM averaged over all k symmetry subspaces (in both cases $\eta = 0.05$). We find that the agreement is fair and the broadened Drude $\delta(\omega)$ peak is reproduced, the apparent value at $\omega = 0$ depending on η. We also show the regular part from the ED evaluation indicating that the presence of the Drude weight, which is fairly large for $L = 8$, is limited to $\omega \lesssim 0.3$.

Next, in order to examine the finite size scaling of $\kappa(\omega)$, we present spectra for larger systems, $L = 10, 12, 14$ ($\eta = 0.01$). We notice that the statistical fluctuations drastically decrease with increasing L, so that for $L = 14$ it is sufficient to consider only one k subspace ($k = 0$ is shown, the curves being practically indistinguishable for the other k subspaces). The reason is that the

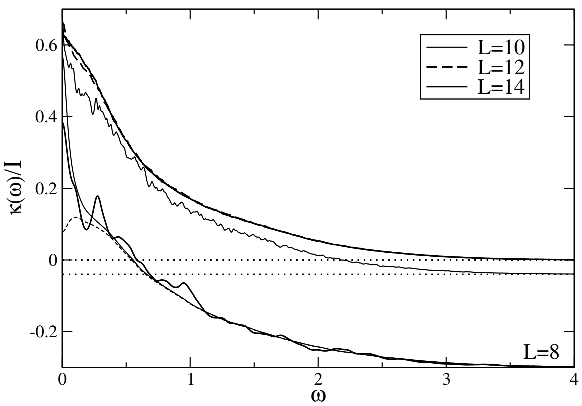

FIG. 2. Upper curves: Normalized thermal conductivity for different L by the MCLM method (curves displaced for clarity). Lower curves for $L = 8$: thick line, MCLM; thin line, $\kappa(\omega)/I$ by ED; thin-dashed line, $\kappa_{\rm reg}(\omega)/I$ by ED.

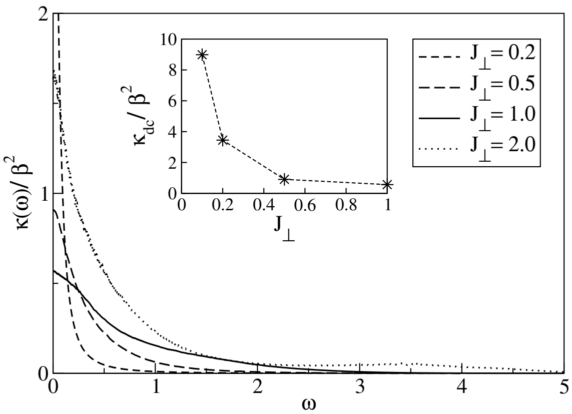

FIG. 3. Thermal conductivity for $L = 14$ rungs as a function of $J_\perp$ for $\beta \to 0$. Inset: dc limit.

dimension of the Hilbert space increases by a factor ~ 10 when increasing L by two rungs. For $L = 14$ it is $O(3 \times 10^6)$ states in each k subspace and so it represents a fairly dense spectrum at high energies (temperatures). Thus, in the following, we show spectra constructed with only the $k = 0$ subspace for $L = 14$ rungs.

Finally, we note that the spectra rapidly collapse to the same curve. Noticeable deviations for $L \geq 10$ are limited to the $\omega \sim 0$–0.3 range. The data apparently converge to a $\kappa(\omega)$ curve that (i) it is characterized by a finite $\kappa_{\rm dc} = \kappa(\omega \to 0)$ value, (ii) it approaches the $\omega \to 0$ limit with a finite slope, (iii) it shows a change of curvature at $\omega \sim 0.5$ for all L, and (iv) its overall ω dependence does not correspond to a Lorentzian or power law behavior; a minimal description of $\kappa(\omega)$ is obtained by an $e^{-|\omega|\tau}$ form [22].

Of course, the deduced $\omega \to 0$ behavior is rather tentative considering the limited size of the systems that we are able to study. A finite L imposes a cutoff on the lowest frequency behavior that can be reliably extracted. We cannot exclude the emergence of a more conventional form—with vanishing zero frequency slope—in the range $\omega \sim 0$–0.2. The apparent finite slope might also be due to the mixing, in the very low frequency spectrum, of a remnant $\delta(\omega)$-Drude contribution that we expect to disappear as $L \to \infty$ (see Fig. 1). In any case, a prominent Drude peak is not observed.

Next, in Fig. 3, we show a series of spectra for $L = 14$ as a function of interchain coupling $J_\perp$ in the $\beta \to 0$ limit (in all cases the contribution of the Drude weight is estimated to less than 10% of the sum rule). For $J_\perp < 1$, first, the dc conductivity (inset) scales as $\kappa_{\rm dc} \sim 1/J_\perp^2$; second, $\kappa(\omega)$ correctly tends to a $\delta(\omega)$ peak as $J_\perp \to 0$ signaling the ballistic transport of decoupled chains; third, the frequency range of finite weight extends up to $\omega \sim 4J_\perp$. For $J_\perp > 1$, a second weak peak appears at $\omega \sim 2J_\perp$ (corresponding to the energy of singlet-triplet rung excitations) and $\kappa_{\rm dc} \sim J_\perp$ as now $J_\perp$ becomes now the dominant energy scale. Notice that, for $J_\perp < 1$, the overall behavior of $\kappa(\omega)$ is what one would qualitatively expect by taking into account within a perturbative scheme the effect of interchain coupling on the ballistic transport of a noninteracting system. In this case, however, the "free" limit is the spin-1/2 Heisenberg Hamiltonian which implies that the effect of a perturbation on the ideal transport of an integrable system is described within a simple picture.

In Fig. 4, we show the frequency dependence of the thermal conductivity at lower temperatures. An overall similar behavior as for $\beta \to 0$ is found except that the rounding at $\omega \sim 0.5$ disappears for $T = 1$. The data are qualitatively described by two separate exponential forms, one in the frequency range $\omega \sim 0$–0.7 and another one above. No excessive enhancement of the dc conductivity is observed. The sum rule (6) is now satisfied to about 95% and the L dependence of the conductivity spectra remains within the statistical noise of the data. It is rather difficult to study even lower temperatures as the energy spectrum is very sparse at low energies, and thus the fluctuations significantly increase.

From the extracted dependence of $\kappa_{\rm dc}$ on $J_\perp$, we can estimate an experimentally relevant—$\kappa_{\rm dc}^{\rm exp}$—magnetic contribution to the thermal conductivity at high temperatures. Reinserting units in the basic expression (4), $\kappa_{\rm dc}^{\rm exp}$ is given by

$$\kappa_{\rm dc}^{\rm exp} = \frac{k_B}{abc}\left(\frac{J^2 c}{\hbar}\right)^2\left(\frac{\hbar}{J}\right)\frac{1}{J^2}\kappa_{\rm dc}\left(\beta J, \frac{\hbar}{J}\omega\right). \quad (8)$$

a, b, c are lattice constants (c along the ladder axis) and energies are in degrees K. For characteristic lattice constants of $O(10\ \text{Å})$ and $J \sim O(1000\ \text{K})$, we obtain a thermal conductivity $O(2\ \text{W/mK})$ that scales as $(\beta J)^2$ and $(J/J_\perp)^2$ for $T \gg J$. If we assume an exponential increase of the conductivity (due to the freezing of Umklapp processes) with characteristic energy $J(= J_\perp)$ below a temperature of $O(J)$, we obtain a room temperature thermal conductivity of order 10–100 W/mK that is consistent with experiment. We should keep in mind in this estimation that it is notoriously difficult [23] to establish

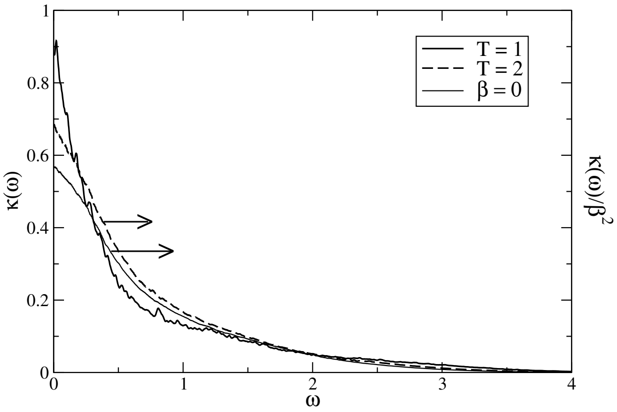

FIG. 4. Thermal conductivity for $L = 14$ at $T = 1$, $T = 2$ ($\beta = 0.5$), and for comparison at $\beta \to 0$.

the temperature dependence of the thermal conductivity for $T \leq J$.

We can then argue that the experimentally observed high values of the thermal conductivity [1,3] in ladder compounds are due to the large exchange coupling J and that they are limited by spin-spin as well as spin-phonon scattering. This is in contrast to quasi-1D materials described by the Heisenberg spin-1/2 Hamiltonian [2,5], where it is unambiguous that the spin-spin scattering is absent.

Discussion.—The presented numerical study indicates that, at least in the high temperature limit, the thermal conductivity in two-leg spin-1/2 ladders is characterized by a finite dc value and thus nonballistic transport. The low frequency spectra do not seem consistent with a Lorentzian form. At lower temperatures there is a change in behavior but not a dramatic enhancement of the dc conductivity or the appearance of a prominent Drude peak. Presently, with the available numerical methods, it is rather challenging to explore an eventual crossover to a quasiballistic regime at low temperatures.

Furthermore, this analysis exemplifies the effect of a nonintegrable interaction (interchain coupling) on the ballistic transport of an integrable system. The obtained perturbative result indicates that the presence in a Hamiltonian of an integrable case with diverging conductivity is signaled over a finite range in interaction parameter space.

Regarding experimental realizations, this study is more relevant to ladder compounds with J smaller than the room temperature, but it shows that materials with high thermal conductivities can be obtained by increasing the coupling J and/or reducing the interchain coupling $J_\perp$. It also shows that it would be interesting to explore the unusual frequency dependence, for instance, by light scattering. As to the observability of the exceptional transport in integrable systems, we expect the effect to be most remarkable at high temperatures in small exchange coupling J one-dimensional spin-1/2 Heisenberg systems.

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