

Thermal conductivity of a classical one dimensional spin-phonon system

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We investigate the thermal conductivity κ of the classical Heisenberg spin chain coupled to a variety of phonon systems using a Green-Kubo approach as well as coupling to heat reservoirs. The decay of energy current correlations is power law or exponential, depending on the type of (an)harmonicity of the phonon system and the presence of a substrate. In particular, we find that coupling the spin system to displacements with an acoustic type harmonic inter-site potential leads to a diverging κ . Adding a substrate, harmonic or periodic such as sine-Gordon, drives the system to diffusive behavior. With anharmonic potential without a substrate the generic transport is diffusive even for the integrable Toda potential, while for the FPU chain is ballistic.

Recent experiments[1, 2] 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 (undoped or hole doped) as well as spin-1 compounds have been studied. The observed highly anisotropic thermal conductivity, that is 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. This observation falls in line with a proposal of unconventional transport in 1D integrable systems[3].

In general, the energy transport in magnetic materials has a phonon (lattice) and a magnetic contribution. The thermal conductivity is limited by phonon-phonon (anharmonicity), spin-spin and spin-phonon scattering. In the materials discussed above, described by the 1D spin-1/2 Heisenberg Hamiltonian (assuming that there is no disorder), in the absence of spin-spin scattering, the thermal conductivity is only limited by phonon-phonon and spin-phonon scattering. Clearly there is experimental and theoretical interest in understanding the thermal conductivity in this singular case[4, 5, 6]. However, the generic case of 1D magnetic thermal transport for higher or even classical spin (large S limit) is a benchmark that is also of experimental interest, e.g. for $S = 1, 3/2, 5/2$ compounds[2].

In this work we focus on the classical Heisenberg model coupled to lattice displacements. As both systems are classical we can resort to large scale, state of the art numerical simulations. This is in contrast to the quantum problem which is numerically intractable because of the unbounded phonon spectrum and thus limited to perturbative or semi-phenomenological analysis.

There is a great deal of work done on the thermal conductivity of 1D classical lattices[7] where a variety of behaviors (ballistic vs. diffusive) was established depending on the anharmonicity of the lattice and presence of a sub-

strate. For the classical Heisenberg model it seems that the thermal transport is diffusive[8, 9].

In the following, we present a series of simulations exploring the role of the phonon bath transport properties to the total thermal conductivity. In particular we find that the coupling to acoustic phonons leads to a diverging κ . This is in contrast to conclusions for the $S = 1/2$ quantum chain[4, 6] based on the memory function approach. This difference is surprising because we would expect that the quantum spin chain (with ballistic transport) would be more robust to coupling to phonons than the classical one.

We consider the prototype classical Heisenberg spin chain Hamiltonian coupled to lattice degrees of freedom carrying the spins,

$$H = \sum_n J_{n,n+1} (\mathbf{S}_n \cdot \mathbf{S}_{n+1}) + \frac{1}{2} M (dQ_n/dt)^2 + M v_0^2 \{V([Q_{n+1} - Q_n]/l) + U(Q_n/l)\} \quad (1)$$

where, $\mathbf{S}_n = S \mathbf{e}_n$ (\mathbf{e}_n a unit vector), Q_n is the displacement of the n th atom with mass M from the equilibrium position nl , l is the lattice spacing, v_0 is the velocity of longitudinal sound, V (U) the dimensionless interatomic (substrate) potential ($V(0) = V'(0) = 0$, $V''(0) = 1$, $U(0) = U'(0) = 0$, $U''(0) = k \geq 0$). The exchange constant $J_{n,n+1}$ depends on the distance between the n th and $(n+1)$ th atoms as $J_{n,n+1} = J[1 - \chi(Q_{n+1} - Q_n)/l]$, $J > 0$ with χ the dimensionless spin-phonon coupling.

The equations of motion for the magnetic and elastic subsystems,

$$\frac{d}{dt} \mathbf{S}_n = -\mathbf{S}_n \times \frac{\partial H}{\partial \mathbf{S}_n}, \quad M \frac{d^2}{dt^2} Q_n = -\frac{\partial H}{\partial Q_n}, \quad (2)$$

written in dimensionless form are,

$$\dot{\mathbf{e}}_n = c_t [(1 - \chi \rho_{n-1}) \mathbf{e}_{n-1} \times \mathbf{e}_n - (1 - \chi \rho_n) \mathbf{e}_n \times \mathbf{e}_{n+1}], \quad (3)$$

$$\ddot{u}_n = c_e \chi (f_{n-1} - f_n) + V'(\rho_n) - V'(\rho_{n-1}) - U'(u_n), \quad (4)$$

where the dot denotes differentiation with respect to the dimensionless time $\tau = v_0 t/l$, $\rho_n = u_{n+1} - u_n$, $u_n = Q_n/l$ is the dimensionless displacement and $f_n = (\mathbf{e}_n \cdot \mathbf{e}_{n+1})$. Here we have introduced, the coefficient $c_t = JS/l/v_0$ as the ratio of characteristic elastic (l/v_0) and magnetic ($1/JS$) times and $c_e = JS^2/Mv_0^2$ as the ratio of characteristic magnetic (JS^2) and elastic (Mv_0^2) energies. In the following, we take JS^2 as the unit of energy.

We are going to consider three types of inter-cite potentials, (i) the harmonic potential $V(\rho) = \rho^2/2$, (ii) the FPU potential $V(\rho) = \rho^2/2 + \rho^4/4$, (iii) the Toda potential $V(\rho) = \exp(-\rho) + \rho - 1$ and two type of on-cite substrates, (iv) the harmonic $U(u) = ku^2/2$ and (v) the anharmonic sine-Gordon, $U(u) = 1 - \cos(2\pi u)$.

The framework for studying the thermal conductivity is the linear response Green-Kubo formalism where the thermal conductivity κ at temperature T is given by the energy current J autocorrelation, $C(\tau) = \lim_{N \rightarrow \infty} C_N(\tau)$, $C_N(\tau) = \langle J(\tau)J(0) \rangle / NT^2$,

$$\kappa = \lim_{\tau \rightarrow \infty} \int_0^\tau C(s) ds. \quad (5)$$

N is the number of particles in a chain with periodic boundary conditions, $J(\tau) = \sum_{n=1}^N j_n(\tau)$ is the total energy current, with the local one j_n obtained from the continuity equation for the energy; $j_n = \{g_{n-1} + g_n + (\dot{u}_{n-1} + \dot{u}_n)[c_e \chi(\mathbf{e}_{n-1} \cdot \mathbf{e}_n) - V'(\rho_n)]\}/2$ and $g_n = c_t c_e (1 - \chi \rho_{n-1})(1 - \chi \rho_n)(\mathbf{e}_{n-1} \times \mathbf{e}_n \cdot \mathbf{e}_{n+1})$. For $\chi = 0$ we recover the usual expressions for the isolate spin and lattice energy current. The averaging $\langle \cdot \rangle$ is performed over thermalized states of the chain.

To thermalize the system we use Langevin dynamics with the a white Gaussina noise thermostat coupled either to the lattice displacements only or to the spin and lattice displacements[9]. We verified that both approaches give practically the same results for thermodynamic (energy, specific heat) as well as dynamic quantities. Following thermalization, the integration of equations of motion (3,4) gives $C(\tau)$ up to times $\tau \sim 1000$ for $N = 4000$ spin/atoms in a chain with periodic boundary conditions – large enough to practically eliminate finite size effects. As $C(\tau)$ depends significantly on the concrete realization of the thermalized chain, we perform an average over about $10^4 - 10^5$ independent initial realizations to improve statistics. In all simulations we consider the parameters $c_t = 1$ and $c_e = 10$ that facilitate the convergence of the simulations but are also fairly realistic as the magnetic energy scale is often larger than the phonon scale.

Let's at first discuss the simplest case of a lattice with only harmonic inter-site interaction $V(\rho) = \rho^2/2$ coupled to spins. No substrate is present ($U(u) = 0$) and thus the phonons are acoustic with linear low energy dispersion. As there is a zero mode with infinite lifetime due to the translational symmetry we take care to use thermalized

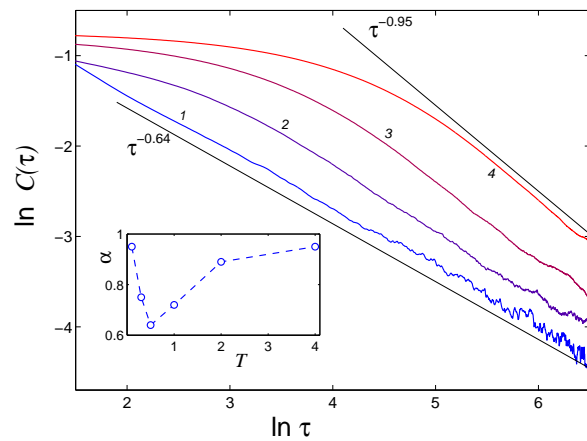


FIG. 1: The power decay of the autocorrelation function $C(\tau) \sim \tau^{-\alpha}$ in the spin-phonon chain with harmonic inter-cite potential (no substrate, $U(u) = 0$) for $\chi = 0.2$ and different temperatures: $T = 0.5, 1, 2, 4$ (curves 1, 2, 3, 4). In the inset, temperature dependence of power α .

states with zero center of mass motion $\sum_{n=1}^N \dot{u}_n(\tau) = 0$.

If χ was zero, then the thermal conductivity would have two independent contributions. The first, from the phonon system that is diverging (purely ballistic) as the lattice is harmonic and the energy current is a constant of motion. The second, from the spins, that is finite (diffusive) with the spin energy current correlations decaying exponentially in time[9]. Thus $C(\tau)$ would decay exponentially fast to a constant (due to the phonon part) and the total thermal conductivity would of course be divergent.

In the spin-phonon coupled system ($\chi \neq 0$), as is shown in Fig. 1, the simulations indicate that the decay of the total energy current autocorrelations becomes power law $C(\tau) \sim \tau^{-\alpha}$ with $\alpha < 1$. Thus the thermal conductivity κ of the coupled system – diffusive spin plus ballistic phonon transport – still diverges. In the inset, the temperature dependence of α is shown. It is nonmonotonic, with a minimum at $T \approx 0.5$, and $\alpha \rightarrow 1$ as $T \rightarrow 0, \infty$, implying a logarithmically diverging conductivity at these limits.

In order to analyze mechanisms for breaking the ballistic behavior, we will now study phonon systems with substrate or with anharmonic inter-site interaction. First, we consider the same harmonic inter-site potential but now in the presence of a substrate harmonic potential $U(u) = ku^2/2$ with $k = 0.25$. Thus the phonon system remains harmonic and the corresponding phonon thermal transport ballistic. The same discussion as above applies for the case $\chi = 0$. Now however, as the simulations indicate, again a power law decay of the energy current autocorrelations is found $C(\tau) \sim \tau^{-\alpha}$ but with a power $\alpha > 1$ which implies a finite total thermal conductivity. The temperature dependence of the spin-phonon

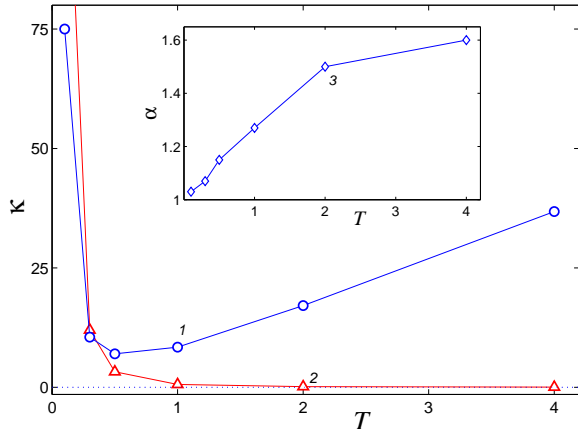


FIG. 2: Thermal conductivity κ as a function of temperature for spin system coupled to harmonic inter-site and substrate potential (curve 1). For reference, thermal conductivity κ_{sp} of uncoupled Heisenberg spin model (curve 2). In the inset, temperature dependence of α in $C(\tau) \sim \tau^{-\alpha}$ - straight lines are aid to the eye.

thermal conductivity is shown in Fig. 2, along with the thermal conductivity of the spin system κ_{sp} alone. We note that, unlike the spin thermal conductivity that decreases basically as $1/T^2$ at high T due to the bounded spin spectrum, the total one increases linearly with T at high temperatures (note that the uncoupled phonon part diverges). Thus it seems that the presence of a substrate is necessary for obtaining a finite κ in the spin-phonon coupled system. The temperature dependence of α is shown in the inset indicating marginal, logarithmic convergence of the energy current autocorrelations as $T \rightarrow 0$ and a stronger one – albeit still power law – at high T . At low temperatures, although the phonon contribution is divergent, the total thermal conductivity is lowered compared to the spin part. Thus the phonons basically act only as scatterers to the spin excitations. Finally, note that as $c_e = 10$, the ratio of magnetic to lattice energies, the phonon system is highly excited at $T/J \sim 1$ ($T/Mv_0^2 \sim 10$).

The above results were found using the Green-Kubo approach. Direct simulation of heat transport in a finite chain of length N , with ends in contact to heat reservoirs at different temperatures, gives good agreement between the two methods. In particular, for the harmonic chain without substrate the heat conductivity coefficient diverges as $\kappa(N) \sim N^{0.34}$ for $N \rightarrow \infty$ and $T = 1$. For the

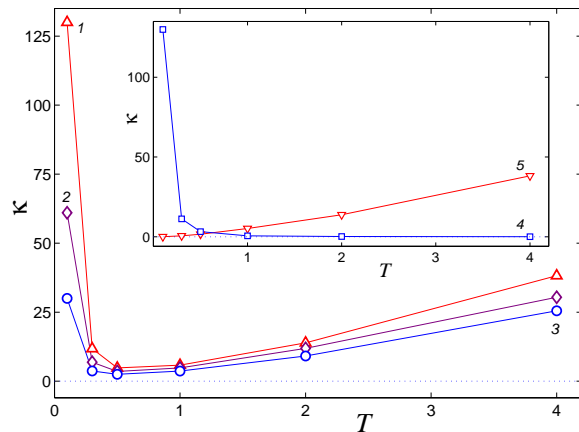


FIG. 3: Thermal conductivity κ as a function of temperature for harmonic inter-site and anharmonic (periodic) substrate potentials for coupling $\chi = 0, 0.1, 0.2$ (curve 1, 2, 3). The case $\chi = 0$ corresponds to the sum of uncoupled spin κ_{sp} and phonon κ_{ph} contributions. In the inset, temperature dependence of individual κ_{sp} and κ_{ph} (curve 4 and 5).

chain with harmonic substrate ($k = 0.25$) a finite limit exists, $\kappa = 8.57$, compared to the value $\kappa = 8.4$ obtained from (5).

To study the coupling of the diffusive spin system with a diffusive phonon one, we consider the on-site sine-Gordon potential $U(u) = 1 - \cos(2\pi u)$ with the same harmonic inter-site one $V(\rho) = \rho^2/2$. This anharmonic phonon system is known[10, 11] to show normal (diffusive) thermal conductivity. Thus for the uncoupled system $\chi = 0$, the autocorrelations of each component decay exponentially with time and the thermal conductivities simply add. For $\chi \neq 0$, now the simulations show that the energy current autocorrelation of the spin-phonon coupled system (as the individual ones) decays exponentially $C(\tau) \sim e^{-\gamma\tau}$, $\gamma > 0$, and the thermal conductivity is finite. In the inset of Fig. 3 we show the temperature dependence of the individual ones for $\chi = 0$ - κ_{sp} for the spin, κ_{ph} for the phonon subsystem. As we see κ_{ph} increases monotonically with T in the temperature region we are considering, while κ_{sp} decreases roughly as $1/T^2$. Actually we expect that at lower temperatures κ_{ph} increases again as the phonon system becomes asymptotically harmonic[12]. The thermal conductivity κ of the coupled system, Fig. 3, goes through a minimum at $T \approx 0.5$. It is interesting to note that the sum of individual component, κ for $\chi = 0$, is significantly larger than the actual κ for $\chi \neq 0$ indicating the importance of spin-phonon scattering in limiting the thermal conductivity. The change $\delta\kappa/\kappa$ is largest, about 50 – 80%, at low temperatures and decreases saturating to 20 – 30% at high temperatures.

Finally, we will consider anharmonic phonon systems without substrate. A classical example is the FPU chain[7] that has been extensively investigated and shows

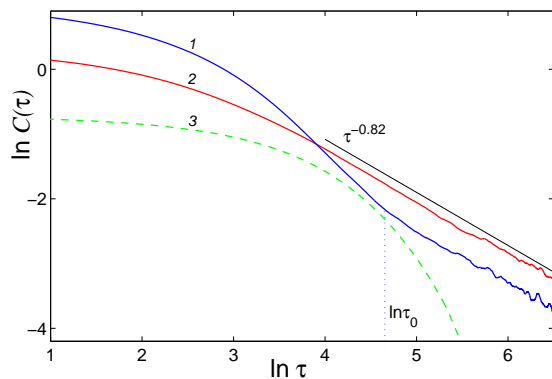


FIG. 4: The power decay of the autocorrelation function $C(\tau)$ in the spin-phonon chain with FPU inter-cite potential (no substrate, $U(u) = 0$) for $T = 0.2$ and $\chi = 0.2$ (curve 1). For comparison, $C(\tau)$ for the isolated ($\chi = 0$) FPU and spin chains (curves 2 and 3) also are given.

infinite conductivity. The autocorrelation function $C(\tau)$ tends to zero as a power law $\tau^{-\alpha}$ with $\alpha < 1$ that depends on temperature T . At high temperature $T = 10$, $\alpha = 0.63$ [13], while for $T = 0.2$, $\alpha = 0.82$ (see Fig. 4). The spin-phonon system turns out to also show ballistic behavior. As shown in Fig. 4, a difference to the FPU case is that here exist two time scales, $\tau < \tau_0$ where $C(\tau)$ decreases exponentially fast, turning to a power law for $\tau > \tau_0$. For $T = 0.2$, the switch time $\tau_0 \approx 90$ and increases as the temperature tends to zero, a result of the dominating spin transport. Note here, that the phonon FPU chain with harmonic substrate has finite conductivity[14] (power law decay of correlations with $\alpha > 1$) and of course the corresponding spin-phonon system has too.

Another classic anharmonic phonon system is the Toda chain, an integrable system with ballistic transport[15]. Our simulations (not shown) conclude that the spin-Toda chain has finite thermal conductivity with exponentially decaying correlations. The temperature dependence of κ is qualitatively similar to the one shown in Fig. 3. As the Toda chain with substrate potential (a nonintegrable system) becomes diffusive, we can speculate that the spin system acts as a substrate that breaks the integrability. Other spin-phonon examples with finite conductivity are the anharmonic Morse and Lenard-Jones potentials.

One surprising outcome for the composite spin-phonon system with a linear vibrational chain is that while conductivity is infinite when the phonon chain is purely acoustic, it becomes finite when there is a phonon gap. Numerics shows that in the high temperature regime thermalized spins act through the term $c_e \chi(f_{n-1} - f_n)$ of Eq. (4) as dynamic scatterers for phonons with a power spectrum similar but not identical to that of white noise. Specifically, we find that this effective

additive noise term has zero mean and is correlated with $\tau_c = 0.5$. In this high-temperature regime one may use the analysis of mode coupling theory relating the energy autocorrelation to the phonon group velocity $v_g(k) = dw(k)/dk$ and the effective local damping $\gamma(k)$ as $C(\tau) \sim \int_{-\pi}^{\pi} dk v^2(k) e^{-\gamma(k)\tau}$ [7]. Algebraic dependence of damping on the wavevector k , viz. $\gamma(k) \sim k^\delta$ results in algebraic decay in the purely acoustic chain as $C(\tau) \sim \tau^{-1/\delta}$ [7], while the presence of the gap results in dominant group velocity contribution at some characteristic wavevector $k \simeq k_0$ leading asymptotically to an exponential decay as $C(\tau) \sim e^{-\gamma(k_0)\tau}$. This heuristic picture is compatible with the numerical evidence presented above.

In conclusion we find that the thermal conductivity of a classical Heisenberg spin chain coupled to linear or nonlinear phonon chains with a substrate is finite. The same holds true when the spin system is coupled to anharmonic chains such as the integrable Toda lattice. However, when the phonon chains is either purely linear acoustic, or weakly nonlinear acoustic such as the FPU lattice conductivity becomes anomalous. Further theoretical work is needed for the understanding of these numerical observations.

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