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# Thermal conductivity of a classical one-dimensional Heisenberg spin model

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We investigate the thermal conductivity of classical spin chains through a Green-Kubo approach using both Monte Carlo and Langevin simulations as well as by direct numerical determination of the microscopic heat transfer processes. All three approaches show that magnetic heat conductivity is normal for all finite temperatures and magnetic fields. In ferromagnets, heat conduction grows linearly with the magnetic field while in antiferromagnets this holds only at high fields. At low fields a quadratic decrease leading to a minimum is attributed to spin mode velocity suppression.

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The macroscopic Fourier law states that under steady state the heat current passing through a system is proportional to the thermal gradient across it. The microscopic foundation of this law in Hamiltonian systems has been actively pursued for many decades and, while it is clear that thermal conductivity is infinite in linear systems due to absence of scattering, this anomaly persists in some quasi-one-dimensional nonlinear systems while others are normal.[1] Heat is transfered through vibrational modes in purely phononic systems but recent experiments have shown that magnetic heat currents are also possible in antiferromagnetic and ferromagnetic materials.[2,3] There is great interest in this magnetic mode of transport as it is extremely efficient, particularly at high temperatures, because (i) it is often characterized by an exchange constant of the order of $eV$ and thus it is comparable to that of metals, (ii) it was shown[4] that quantum, spin-$1/2$ one dimensional systems show ballistic heat transport at all temperatures, the energy current even commutes with the $S=1/2$ Heisenberg Hamiltonian.[5] The insulating nature of the magnetic materials thus promotes them as obvious candidates for technological applications, e.g., for carrying away heat in electronic devices.

In the present work we analyze pure magnetic heat transfer processes in one-dimensional classical spin models and investigate both the nature of the resulting thermal conduction as well as some peculiar properties that appear in the presence of an external magnetic field. They can be considered as the large spin $S$ limit of quantum systems [e.g., TMMC ($(CH_3)_4NMnCl_3$), DMMC ($(CH_3)_2NH_2MnCl_3$) with $S=5/2$] and thus they are directly experimentally relevant.

We consider a one-dimensional regular chain of classical rotors $\vec{S}_n=S_0\mathbf{e}_n$, where $\mathbf{e_n}$ is a unit vector at site $n$ and $S_0$ the dimensional spin part. The Hamiltonian for the classical Heisenberg chain is given by

$$H=\sum_n J_0(\vec{S}_n\cdot\vec{S}_{n+1})-\left(\mu\vec{B}\cdot\sum_n\vec{S}_n\right). \qquad (1)$$

The first sum represents the nearest neighboring spin interaction with exchange coupling constant $J_0\neq 0$. The second term denotes the coupling of the spins to an applied magnetic field, where $\vec{B}=B_0\mathbf{b}$, $B_0$ is the strength of the field, $\mathbf{b}$ is the unit dimensionless vector, and $\mu$ is the absolute value of the magnetic moment of the spin. Using the classical dynamics

$$\frac{d}{dt}\vec{S}_n=-\vec{S}_n\times\frac{\partial H}{\partial\vec{S}_n}, \qquad (2)$$

the equations of motion in dimensionless form become

$$\frac{d}{d\tau}\mathbf{e}_n=-c_0\mathbf{e}_n\times(\mathbf{e}_{n-1}+\mathbf{e}_{n+1}-b\mathbf{b}), \qquad (3)$$

where the coefficients $c_0=J_0/|J_0|=\pm 1$, $b=\mu B_0/|J_0|S_0$, and time $\tau=t/t_0$, $t_0=1/|J_0|S_0$. For $J_0>0$ (antiferromagnetic chain) $c_0=1$ while for $J_0<0$ (ferromagnetic chain) $c_0=-1$. We use hereafter dimensionless variables and since the model is isotropic, we may choose for the magnetic field the $z$ direction, i.e., $\mathbf{b}=\mathbf{e}_z=(0,0,1)$.

For $0\leqslant b<4$ the antiferromagnetic chain has a bistable ground state at $\mathbf{e}_n=\{(-1)^n\cos\phi\cos\theta,(-1)^n\sin\phi\cos\theta,\sin\theta\}$, $n=0,\pm 1,\pm 2,\ldots$, where the azimuthal angle with the field axis has arbitrary value $\phi\in[0,2\pi)$, but the polar angle $\theta=\theta_b$ $(0\leqslant\theta_b\leqslant\pi/2)$ depends on the value of the magnetic field $b\geqslant 0$, viz., $\theta_b=\min_{\theta\in[0,\pi/2]}\{-\cos^2\theta+\sin^2\theta-b\sin\theta\}$. For $b<4$ we have $\sin(\theta_b)=b/4$ while when $b\geqslant 4$ it has the solution $\theta_b=\pi/2$ and the antiferromagnetic chain will have only one ground state at $\mathbf{e}_n=\mathbf{b}=(0,0,1)$. The ferromagnetic chain has this ground state for all $b>0$.

In order to find the dispersion law of the spin modes we linearize the equations of motion by introducing appropriate small vector variables that move with the classical spin vectors $\mathbf{e_n}$; straightforward calculations lead to the dispersion law for antiferromagnets,

$$\omega(q)=2\sqrt{(1-\cos q)(1-\gamma\cos q)},\quad 0\leqslant b\leqslant 4, \qquad (4)$$

$$\omega(q)=b-2(1+\cos q),\quad b>4, \qquad (5)$$

where $\gamma=b^2/8-1$. We observe that in the regime $4/\sqrt{3}\leqslant b\leqslant 4$ the acoustic branch has a maximum for $q$ values not in the zone edge [see Fig. 1(a)] and as a result, the group velocity is zero for these values. One observes that for a field

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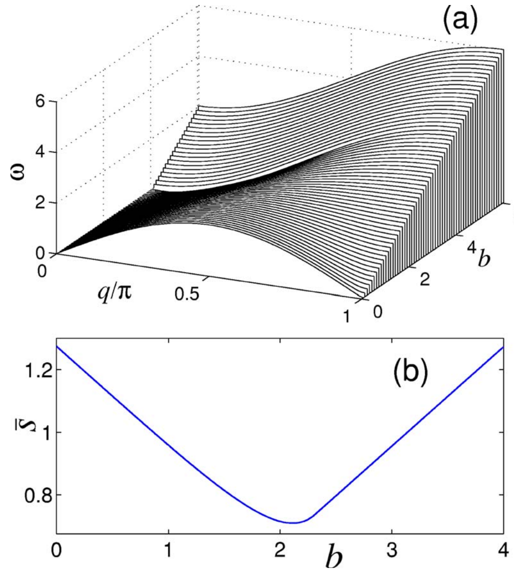


FIG. 1. (Color online) Dispersion curve $\omega(q)$ (a) and average value of envelope velocity $\bar{s}$ (b) as a function of external field $b$ for the antiferromagnetic spin chain.

$b \approx 2.1$ in the interval $q \in [\pi/2, \pi]$, the frequency $\omega(q)$ is practically independent of the wave number $q$ and the group velocity $s(q) = d\omega(q)/dq$ is very small with an average $\bar{s} = (1/\pi)\int_0^{\pi} |s(q)| dq$ that has a minimum as seen in Fig. 1(b). We thus anticipate that for field values $b \approx 2$ spin wave energy transport will be supressed. In the ferromagnetic chains on the other hand, the spectrum (5) has a fixed frequency width $[b, b+4]$ while showing a shift that depends linearly in the field.

From the local energy

$$h_n = \frac{1}{2} c_0 [(\mathbf{e}_{n-1} \cdot \mathbf{e}_n) + (\mathbf{e}_n \cdot \mathbf{e}_{n+1})] - b(\mathbf{b} \cdot \mathbf{e}_n), \tag{6}$$

and the continuity equation, $\dot{h}_n = j_n - j_{n+1}$, we obtain using (3) the local energy flux $j_n$,

$$j_n = \frac{1}{2} c_0 [(\mathbf{e}_{n-2} \times \mathbf{e}_{n-1} \cdot \mathbf{e}_n) + (\mathbf{e}_{n-1} \times \mathbf{e}_n \cdot \mathbf{e}_{n+1})] - b(\mathbf{e}_{n-1} \times \mathbf{e}_n \cdot \mathbf{b}). \tag{7}$$

The thermal conductivity $\kappa$ at temperature $T$ is obtained through the numerical evaluation of a current-current correlation function and the Green-Kubo formula,[6]

$$\kappa = \lim_{\tau \to \infty} \int_0^{\tau} \lim_{N \to \infty} \frac{1}{NT^2} \langle J(s) J(0) \rangle ds, \tag{8}$$

where $N$ is the number of spins in a chain with periodic boundary conditions, $J(s) = \sum_{n=1}^{N} j_n(s)$, and the averaging $\langle \cdot \rangle$ is performed over all thermalized states of the chain.

In order to evaluate the thermal conductivity coefficient through the use of the Green-Kubo formula we calculate numerically the current-current correlation function $C(\tau) = \langle J(\tau) J(0) \rangle / N$ using two independent means, viz., a Monte Carlo ensemble and a dynamic Langevin method. In the former, we use the Metropolis algorithm to generate an ensemble of thermalized initial states, follow the time evolution of this ensemble through the numerical solution of the Hamiltonian equations of motion (3), and evaluate ensemble-averaged quantities, such as $C(\tau)$, the system energy, specific heat, etc. In the latter Langevin method we solve numerically the stochastic version of the Landau-Lifshitz-Gilbert (LLG) equation for magnetic systems:[7,8]

$$(1+\alpha^2) \frac{d}{d\tau} \mathbf{e}_n = \mathbf{e}_n \times \left( \xi_n - \frac{\partial \mathcal{H}}{\partial \mathbf{e}_n} \right) - \alpha \mathbf{e}_n \times \left[ \mathbf{e}_n \times \left( \xi_n - \frac{\partial \mathcal{H}}{\partial \mathbf{e}_n} \right) \right]. \tag{9}$$

In Eq. (9), $\alpha$ is a damping parameter and the white Gaussian noise variables $\xi_n$ represent the thermostat at temperature $T$:

$$\langle \xi_n(\tau) \rangle = 0, \quad \langle \xi_n(\tau_1) \xi_k(\tau_2) \rangle = 2\alpha T \delta_{nk} \delta(\tau_1 - \tau_2). \tag{10}$$

We obtain the thermalized state after time integration of the stochastic equations (9) using an arbitrary set of random initial conditions. The question regarding the finiteness of the heat conductivity in the classical 1D Heisenberg model reduces then to the issue of convergence of the integral $\int_0^{\infty} C(\tau) d\tau$, where $C(\tau)$ is obtained independently through either the ensemble or Langevin methods.

Extensive numerics using both methods demonstrates that they give compatible thermalized states that are practically identical with almost identical energies and very similar specific heats. There is a small difference in the zero temperature behavior of the specific heat that is attributed to the singular zero noise limit of the Langevin method. We account for this by introducing a renormalized noise correlation coefficient that adjusts the zero noise limit in the Langevin results to those of the Monte Carlo method.

The third method we use is direct, viz., proceeds through the numerical simulation of the microscopic heat transfer process. This is performed by embedding the spin chain of size $N$ between two Langevin thermostats, simulated by two spin chains as well. The one on the left-hand side (lhs) ($n \leq 0$) has size $N_-$ and temperature $T_+$ while the other on the right-hand side (rhs) ($n > N$) has size $N_+$ and is at temperature $T_-(T_+ > T_-)$. While the equations of motion for the basic spin system are the Hamiltonian ones (3), the equations for the spin thermostat chains follow the stochastic LLG equations (9) with the same damping $\alpha = 1/\tau_\alpha$ and different noises $\xi_n^{\pm}$ so that the proper temperatures are reached in the edge segments. This system of $N_+ + N + N_-$ equations is integrated numerically for $\alpha = 0.1$, $N_\pm = 40$, $N = 10, 20, 40, 80, 160, 320, 640$ and initial conditions corresponding to the ground state of the chain. After time $\tau \sim 10^5$ the end particles achieve thermal equilibrium with the thermostat and stationary heat flux forms. Subsequently we calculate the time-averaged spin energies $E_n = \langle e_n(\tau) \rangle_\tau$ by

$$\langle e_n(\tau) \rangle_\tau = \lim_{\tau \to \infty} \frac{1}{\tau} \int_0^{\tau} \{ c_0 [\mathbf{e}_n(s) \cdot \mathbf{e}_{n+1}(s)] - b[\mathbf{b} \cdot \mathbf{e}_n(s)] - c_1 \} ds \tag{11}$$

($c_1$ is the energy of the ground state) and the time-averaged heat flux

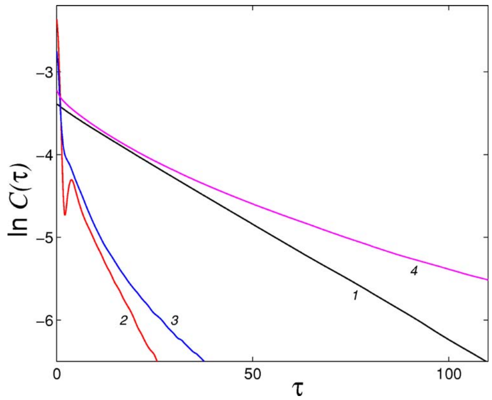


FIG. 2. (Color online) The exponential decay of the autocorrelation function $C(\tau)$ in the thermalized antiferromagnetic spin chain for external fields $b=0$ (curve 1), $b=1.5$ (curve 2), $b=2$ (curve 3), and $b=3$ (curve 4) and temperature $T=0.25$.

$$J_n = \langle j_n(\tau)\rangle_\tau = \lim_{\tau\to\infty} \frac{1}{\tau}\int_0^\tau j_n(s)ds. \quad (12)$$

We establish that in steady state the inner fragment of the chain $N_+ < n \leqslant N_+ + N$ has constant heat flux, i.e., ($J_n = J$) and the temperature has a linear profile. The heat conductivity coefficient is determined through the use of information concerning the inner fragment of the chain:

$$\kappa(N) = \kappa_e(N)(E_{N_+ +1} - E_{N_+ + N})/(T_+ - T_-), \quad (13)$$

where the renormalized coefficient of the heat conductivity

$$\kappa_e(N) = \frac{J(N)}{(E_{N_+ +1} - E_{N_+ +N})} = \frac{J(N)}{[E(T_+) - E(T_-)]}, \quad (14)$$

or, by fitting a linear function $T_n = a_1 n + a_2$, where $\kappa_e(N) = J/a_1$, to the inner fragment of the chain. The limiting value

$$k = \lim_{N\to\infty} \frac{E(T_+) - E(T_-)}{T_+ - T_-} k_e(N) = c(T)\bar{k}_e, \quad (15)$$

where $\bar{k}_e = \lim_{N\to\infty} k_e(N)$ and $c(T) = dE(T)/dT$ is the heat capacity corresponding to temperature $T = (T_+ + T_-)/2$ for thermal energy $E(T) = (E_+ + E_-)/2$. In the context of this third, direct method, the issue regarding the finiteness or not of the heat conductivity reduces to the existence of a finite limiting value for $k$ in Eq. (15).

Extensive numerical investigation of the classical spin model using both the Green-Kubo approach with either Monte Carlo or Langevin thermalization as well as the direct heat-flux method shows that the one component 1D spin chain has finite thermal conductivity at all finite temperatures (in agreement with previous infinite temperature studies[9,10]), in sharp contrast to the unconventional spin conductivity.[10] For all $T > 0$ and fields $b \geqslant 0$, both for ferromagnetic and antiferromagnetic interactions, the correlation function decays exponentially to zero at long times (Fig. 2) and, as a result, the integral in the Green-Kubo formula (8) is finite and the heat conductivity coefficient $\kappa$ has a finite value. These results are corroborated by the direct heat-flux method

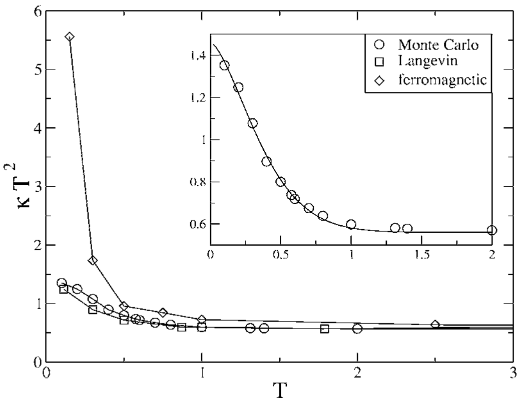


FIG. 3. The thermal conductivity coefficient $\kappa T^2$ as a function of temperature $T$ for an antiferromagnetic chain from the Green-Kubo formula using the Monte Carlo (circles) or Langevin (squares) thermalization method and ferromagnetic chain (diamonds) at zero field ($b=0$). Inset: the low temperature interpolation to a stretched exponential $\kappa T^2 = 0.56 + 0.9\exp(-3.8T^{1.6})$.

showing that a limiting value for large system sizes exists, viz., $c(T)\lim_{N\to\infty} k_e(N) = \kappa$ and is the same as the one obtained by the Green-Kubo method.

Having established both the compatibility of the results obtained via all three techniques used in this work and the finiteness of the thermal conductivity in the model under study, we now investigate its temperature and magnetic field dependence. Extracting the trivial $1/T^2$ prefactor in the Green-Kubo formula (8), we show in Fig. 3 $\kappa T^2$ as a function of temperature $T$ at zero field. In the high temperature limit (actually for $T \gtrsim 3$), this function is practically constant and we extract a $T\to\infty$ for the energy diffusion constant, $D_E = \kappa/c \simeq (0.58/T^2)/(1/3T^2) \simeq 1.74$ that is compatible with previous estimates and about a factor of two larger than the one obtained by the moment method.[11] In the inset we show the low temperature behavior that is well fitted by a stretched exponential law $\kappa T^2 = c_0 + c_1\exp(-c_3 T^a)$ with $a \simeq 1.6$, thus close to a Gaussian.

The field dependence of $\kappa(T=0.5)$ is summarized in Fig. 4 both for ferromagnetic and antiferromagnetic interactions; we observe that while in the former case the thermal conductivity increases linearly with the field, the dependence in the latter is much richer. The linear growth of $\kappa$ in the ferromagnetic case is related to the field-dependent shift of the linear dispersion relation; we have confirmed numerically that the nonlinear spectrum also shifts to higher frequencies with $b$. Furthermore, the spin velocity power spectrum[12] $p(\omega)$, $\int_0^\infty p(\omega)d\omega = \langle \dot{\mathbf{e}}_n \cdot \dot{\mathbf{e}}_n\rangle$, increases as spins tend to orient parallel to the field, leading to a decrease of the nonlinear terms contribution in the equations of spin motion. As a result, the heat conductivity grows linearly with $b$.

In the antiferromagnetic case we face a completely different situation. Heat conductivity increases linearly with $b$ only for large fields, i.e., for $b \geqslant 4$ (Fig. 4). In the interval $0 \leqslant b \leqslant 4$ we find a local minimum $\kappa_{min} = \min_{b\in[0,\infty)}\kappa$ for field values in the range $b_0 = 1.5$ to $b = 2$. In Fig. 4 we present the small field dependence of $\kappa/\kappa_0$, where $\kappa_0$ is the zero field heat conductivity for various temperatures. We observe that

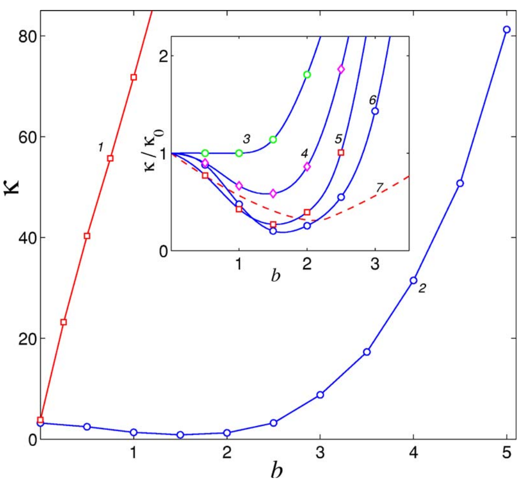


FIG. 4. (Color online) The dependence of thermal conductivity coefficient $\kappa$ on external field $b$ for ferro- (curve 1) and antiferromagnetic spin chain (curve 2) at temperature $T=0.5$) In the inset: the field dependence of relative value of heat conductivity $\kappa/\kappa_0$ on value $b$ ($\kappa_0$ is heat conductivity for $b=0$) for temperatures $T=2,1,0.5,0.25$ (curves 3, 4, 5, 6 resp.). Line 7 gives the dependence $\bar{s}^2(b)/\bar{s}^2(0)$.

$\kappa_{min}/\kappa_0$ decreases when $T$ becomes smaller, viz. the effect becomes more pronounced at smaller temperatures when nonlinearity in the interactions plays a more important role.

We noted earlier that the group velocity of linear antiferromagnetic modes becomes zero for $q$ values in the band while the linear dispersion relation for $b\leq 2$ depends weakly on $b$ in this regime. Numerics show that in this field interval the power spectrum of spin velocities $p(\omega)$ is also practically independent of $b$, and, furthermore, a strongly pronounced maximum appears in the field range $b=1.5-2$ at frequency $\omega_b=b$. The thermal conductivity minimum is a direct consequence of this heat transport mode velocity reduction. This picture is corroborated by the field dependence of the correlation function $C(\tau)$ (Fig. 2). While for $b=0$ and $b=3$, $C(\tau)$ decays exponentially with a practically constant slope, for $b=1.5$ and $b=2$ a distinct behavior appears with two distinct time scales. In the time regime $0\leq\tau\leq\tau_b$, where $\tau_b=2\pi/b$ is the period of spin waves with frequency $\omega=b$, the correlation function decays very fast. In the second time regime $\tau>\tau_b$ it tends exponentially to zero but with a substantially reduced rate. In this time regime there is a resonant suppression of mode transfer due to enhanced scattering at frequency $\omega=b$, provided that $b$ is in the range [1.5, 2].

When $b>2$ the spin velocity spectrum $p(\omega)$ shifts to higher $b$ values with the field, the frequency $\omega_b$ shifts to the right boundary of the frequency spectrum and the conductivity grows monotonically. Finally, for $b>4$ the frequency interval of linear spin waves $[b-4,b]$ shifts to the right of the value of external field $b$ and so does the spectrum leading to an increase of heat conductivity proportional to $b-4$.

In this work we have shown that both ferromagnetic and antiferromagnetic classical spin chains have finite heat conductivity for all values of temperature and external fields. The normal heat conductivity found is related to nonlinear spin wave interactions. While in the ferromagnetic system heat conduction is linear in the field, this dependence is true only at large fields in the antiferromagnetic case. Furthermore, in the latter case and at smaller fields a minimum in the conductivity appears that is a nonlinear manifestation of the suppression of propagation of spin modes. In a Drude-like picture of thermal conductivity, one may use the expression $\kappa\sim c\bar{s}^2\tau$, where $\tau$ is the collision time. In the small field regime, the antiferromagnetic chain specific heat is practically independent of the field while, to lowest order, the scattering time may be considered unaffected by it. The resulting quadratic decay of the heat conductivity $\kappa(b)/\kappa(0)\approx\bar{s}^2(b)/\bar{s}^2(0)$ as a function of the field shown in Fig. 4 (curve 7), and gives a very clear prediction regarding the low temperature magnetic field dependence of thermal conductivity in high spin quantum number antiferromagnets (see also a similar experimental observation in Ref. 13).

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