Thermal conductivity of a classical one-dimensional Heisenberg spin model

A. V. Savin, G. P. Tsironis, and X. Zotos

1Institute of Chemical Physics RAS, Kosygin Strasse 4, Moscow 119991, Russia
2Department of Physics, University of Crete and Foundation for Research and Technology-Hellas, P.O. Box 2208, 71003 Heraklion, Greece

(Rceived 24 August 2005; published 4 October 2005)

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.

DOI: 10.1103/PhysRevB.72.140402  PACS number(s): 75.10.Hk, 66.70.+f

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 transferred 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 shown4 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 ((CH₃)₄N_fmCl₃), DMMC ((CH₃)₂NH₂MnCl₃) with \( S=5/2 \)] and thus they are directly experimentally relevant.

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

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

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 \( \vec{B} = B_0 \vec{b} \), \( B_0 \) is the strength of the field, \( \vec{b} \) is the unit dimensionless vector, and \( \mu \) is the absolute value of the magnetic moment of the spin. Using the classical dynamics

\[
\frac{d}{dt} \vec{S}_n = -\vec{S}_n \times \frac{\partial H}{\partial \vec{S}_n}, \tag{2}
\]

the equations of motion in dimensionless form become

\[
\frac{d}{dt} \vec{e}_n = -c_0 \vec{e}_n \times (\vec{e}_{n+1} + \vec{e}_{n-1} - 2\vec{b}), \tag{3}
\]

where the coefficients \( c_0 = 1/J_0 |J_0| = \pm 1/2, -\mu B_0 |J_0| S_0 \), and \( \tau = t / t_0 \), \( t_0 = 1/J_0 |J_0| S_0 \). 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., \( \vec{b} = \vec{e}_z = (0, 0, 1) \).

For \( 0 < b < 4 \) the antiferromagnetic chain has a bistable ground state at \( \vec{e}_n = \{(-1)^n \cos \phi \cos \theta, (-1)^n \sin \phi \cos \theta, \sin \theta\} \), where the azimuthal angle with the field axis has arbitrary value \( \phi \in [0, 2\pi] \), but the polar angle \( \theta = \theta_b \) \((0 \leq \theta_b \leq \pi/2)\) depends on the value of the magnetic field \( b \).

For \( b > 4 \) we have \( \sin(\theta_b) = b/4 \) while when \( b \geq 4 \) it has the solution \( \theta_b = \pi/2 \) and the antiferromagnetic chain will have only one ground state at \( \vec{e}_n = \vec{b} = (0, 0, 1) \). The ferromagnetic chain has this ground state for all \( b \geq 0 \).

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 \( \vec{e}_n \); straightforward calculations lead to the dispersion law for antiferromagnets,

\[
\omega(b) = 2\sqrt{(1 - \cos q)(1 - \gamma \cos q)}, \quad 0 \leq b \leq 4, \tag{4}
\]

\[
\omega(b) = b - 2(1 + \cos q), \quad b > 4, \tag{5}
\]

where \( \gamma = b^2/4 - 1 \). We observe that in the regime \( 4/\sqrt{3} \lesssim b \leq 4 \) 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...
velocity of envelope velocity and the continuity equation, practically independent of the wave number of this function and the Green-Kubo formula,\textsuperscript{6} the antiferromagnetic spin chain.

We thus anticipate that for field values $H < H_{c1}$, the spectrum $\omega(q)$ is practically independent of the wave number $q$ and the group velocity $s(q) = \partial \omega(q) / \partial q$ is very small with an average $\bar{s} = \frac{1}{T} \int_0^T s(q) dq$ that has a minimum as seen in Fig. 1(b).

From the local energy

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

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

$$j_n = \frac{1}{2} c_0 ((\mathbf{e}_{n-2} \cdot \mathbf{e}_{n-1}) \cdot \mathbf{e}_n) + (\mathbf{e}_{n-1} \cdot \mathbf{e}_n) \cdot \mathbf{e}_{n+1})$$

$$- b (\mathbf{e}_{n-1} \cdot \mathbf{e}_n \cdot \mathbf{b}).$$

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

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

where $N$ is the number of spins in a chain with periodic boundary conditions, $J(s) = \sum_{n=1}^N \mathbf{j}(s)$, 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:\textsuperscript{7,8}

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

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

$$\langle \xi_n(\tau) \rangle = 0, \quad \langle \xi_n(\tau_1) \xi_n(\tau_2) \rangle = 2 \alpha T \delta_{\tau_1, \tau_2}.$$  

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) $(n \equiv 0)$ has size $N_c$ and temperature $T_c$ while the other on the right-hand side (rhs) $(n > N)$ has size $N_s$ and is at temperature $T_r$ ($T_r > T_c$). 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 $\alpha = 1/\tau_n$ and different noises $\xi_{n}^\infty$ so that the proper temperatures are reached in the edge segments. This system of $N_s + N + N_c$ equations is integrated numerically for $\alpha = 0.1$, $N_s = 40$, $N = 10, 20, 40, 80, 160, 320, 640$ 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 $E_n = \langle \mathbf{e}_n(\tau) \rangle$ by

$$\langle \mathbf{e}_n(\tau) \rangle = \frac{1}{\tau} \int_0^\tau \mathbf{c}_0 \langle \mathbf{e}_n(s) \cdot \mathbf{e}_{n+1}(s) \rangle - b \langle \mathbf{b} \cdot \mathbf{e}_n(s) \rangle - c_1 ds,$$

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

\begin{figure}[h]
\centering
\includegraphics[width=\textwidth]{dispersion}
\caption{(Color online) Dispersion curve $\omega(q)$ (a) and average value of envelope velocity $\bar{v}$ (b) as a function of external field $b$ for the antiferromagnetic spin chain.}
\end{figure}
THERMAL CONDUCTIVITY OF A CLASSICAL ONE-...
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 $b=2$ depends weakly on $b$ in this regime. Numerics show that in this field interval the power spectrum of spin velocities $\rho(\omega)$ 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 $0 \leq \tau \leq \tau_p$, where $\tau_p=2\pi/b$ is the period of spin waves with frequency $\omega=b$, the correlation function decays very fast. In the second time regime $\tau > \tau_p$ 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 $\rho(\omega)$ 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 $\kappa \sim c\tau$, 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 $\kappa(b)/\kappa(0) = \bar{s}^2(b)/\bar{s}^2(0)$ 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).

This work was supported by E.U. Grant No. MIRG-CT-2004-510543. One of us (A.V.S.) also thanks the Russian Federation of Basic Research (Award No. 04-02-17306) for partial financial support.