Universality of anomalous one-dimensional heat conductivity

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In one and two dimensions, transport coefficients may diverge in the thermodynamic limit due to long-time correlation of the corresponding currents. The effective asymptotic behavior is addressed with reference to the problem of heat transport in one-dimensional crystals, modeled by chains of classical nonlinear oscillators. Extensive accurate equilibrium and nonequilibrium numerical simulations confirm that the finite-size thermal conductivity diverges with system size L as $\kappa \propto L^{\alpha}$. However, the exponent α deviates systematically from the theoretical prediction $\alpha = 1/3$ proposed in a recent paper [O. Narayan and S. Ramaswamy, Phys. Rev. Lett. **89**, 200601 (2002)].

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Strong spatial constraints can significantly alter transport properties. The ultimate reason is that the response to external forces depends on statistical fluctuations which, in turn, crucially depend on the system dimensionality d. A relevant example is the anomalous behavior of heat conductivity for $d \leqslant 2$. After the publication of the first convincing numerical evidence of a diverging thermal conductivity in anharmonic chains [1], this issue attracted a renovated interest within the theoretical community. A fairly complete overview is given in Ref. [2], where the effects of lattice dimensionality on the breakdown of Fourier's law are discussed as well. Anomalous behavior means both a nonintegrable algebraic decay of equilibrium correlations of the heat current $J(t)$ (the Green-Kubo integrand) at large times $t \to \infty$ and a divergence of the finite-size conductivity $\kappa(L)$ in the $L \to \infty$ limit. This is very much reminiscent of the problem of long-time tails in fluids [3] where, in low spatial dimension, transport coefficients may *not exist at all*, thus implying a breakdown of the phenomenological constitutive laws of hydrodynamics. In one dimension (1D) one finds

$$\kappa(L) \propto L^{\alpha}, \quad \langle J(t)J(0)\rangle \propto t^{-(1+\delta)}, \tag{1}$$

where $\alpha > 0$, $-1 < \delta < 0$, and $\langle\ \rangle$ is the equilibrium average. For small applied gradients, linear-response theory allows establishing a connection between the two exponents. By assuming that $\kappa(L)$ can be estimated by cutting off the integral in the Green-Kubo formula at the "transit time" L/v (v being some propagation velocity of excitations), one obtains $\kappa \propto L^{-\delta}$—i.e., $\alpha = -\delta$.

Determining the asymptotic dependence of heat conductivity is not only important for assessing the universality of this phenomenon, but may be also relevant for predicting transport properties of real materials. For instance, recent molecular dynamics results obtained with phenomenological carbon potentials indicate an unusually high conductivity of single-walled nanotubes [4]: a power-law divergence with the tube length has been observed with an exponent very close to the one obtained in simple 1D models [5].

The analysis of several models [2] clarified that anomalous conductivity should occur generically whenever momentum is conserved. For lattice models, this amounts to requiring that at least one acoustic phonon branch be present in the harmonic limit. The only known exception is the coupled-rotor model, where normal transport [6] is believed to arise as a consequence of the boundedness of the potential.

It is thus natural to argue about the universality of the exponent α. On the one hand, there exist two theoretical predictions—namely, $\alpha = 2/5$, which follows from self-consistent mode-coupling theory [7,8], and $\alpha = 1/3$, obtained by Narayan and Ramaswamy [9] by performing a renormalization-group calculation on the stochastic hydrodynamic equations for a 1D fluid. On the other hand, the available numerical data for α range from 0.25 to 0.44. The most convincing confirmation of the 1/3 value has been obtained by simulating a one-dimensional gas of hard-point particles with alternating masses [10,11] and random-collision models [12]. In the former case, a careful determination of the scaling exponent is, however, hindered by the presence of large finite-size corrections that are still sizable for $\mathcal{O}(10^4)$ particles. As a matter of fact, other authors [13] report significantly smaller estimates ($\alpha \simeq 0.25$). This anomaly is possibly due to the lack of microscopic chaos in that model [11]. The results obtained for models of 1D crystals are more controversial, but consistently larger than 1/3. For instance, in the case of the Fermi-Pasta-Ulam (FPU) chain, the best estimate so far is $\alpha \simeq 0.37$ [8,14].

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However, with the only exception of Ref. [11], all numerical investigations limit themselves to fitting the scaling behavior in a suitable range, without determining possible finite-size corrections, so that none of them can be fully trusted. In view of the general relevance of establishing the existence of one or more universality classes, in the present paper we present far more accurate simulations which allow determining the effective exponents for different lengths and frequencies. We anticipate that α is definitely larger than 1/3 in a 1D crystal model and possibly in agreement with the mode-coupling prediction.

We consider an array of N pointlike identical atoms ordered along a line. The position of the nth atom is denoted with x_n, while its mass is fixed, without loss of generality, equal to unity. By further assuming that interactions are restricted to nearest-neighbor pairs, the equations of motion are written as

$$\ddot{x}_n = -F_n + F_{n-1}, \quad F_n = -V'(x_{n+1} - x_n), \tag{2}$$

where $V'(z)$ is shorthand notation for the first derivative of the interparticle potential V with respect to z. The microscopic expression of the heat current is

$$J = \sum_n \left[\frac{1}{2}(x_{n+1} - x_n)(\dot{x}_{n+1} + \dot{x}_n) F_n + \dot{x}_n h_n \right], \tag{3}$$

where h_n is a suitably defined local energy [2]. For small oscillations (compared to the lattice spacing $b = L/N$), the second term can be neglected and $x_n - x_{n-1} \simeq b$, so that Eq. (3) can be approximated by

$$J \simeq \frac{b}{2} \sum_n (\dot{x}_{n+1} + \dot{x}_n) F_n . \tag{4}$$

The customary way to evaluate the thermal conductivity κ is through the Green-Kubo formula

$$\kappa_{GK} = \frac{1}{k_B T^2} \lim_{t\to\infty} \int_0^t d\tau \lim_{L\to\infty} L^{-1} \langle J(\tau) J(0) \rangle . \tag{5}$$

A crucial, sometimes overlooked [15], point is that such formulas are formally identical for different statistical ensembles, but the definition of J differs, because of ''systematic'' contributions associated with other conservation laws that must be subtracted out [16]. For instance, expression (4) is correct in the microcanonical ensemble with zero total momentum, while in the canonical ensemble (for large N) it is

$$J = \frac{b}{2} \sum_n (\dot{x}_{n+1} + \dot{x}_n) F_n - b v_0 \left\langle \sum_n F_n \right\rangle , \tag{6}$$

v_0 being the center-of-mass velocity. This choice ensures that the autocorrelation of J vanishes for $t \to \infty$.

With reference to Eq. (1), the possibly anomalous behavior can be analyzed by computing the power spectrum of the heat current J. Since we are interested in the long-wavelength and small-frequency behavior, it is convenient to consider a highly nonlinear model in the hope that the asymptotic regime sets in over shorter time and space scales. Moreover, it is advisable to work with a computationally simple expression of the force. The best compromise we have found is the quartic Fermi-Pasta-Ulam potential

$$V(z) = \frac{1}{4}(z - a)^4 . \tag{7}$$

Indeed, after the change of coordinates $x_n = u_n + na$ [17], the physical distance a disappears from the equations of motion for u_n. This model has no free parameters: since the potential expression is homogeneous, the dynamics is invariant under coordinate rescaling, so that the energy per particle e can be set, without loss of generality, equal to 1.

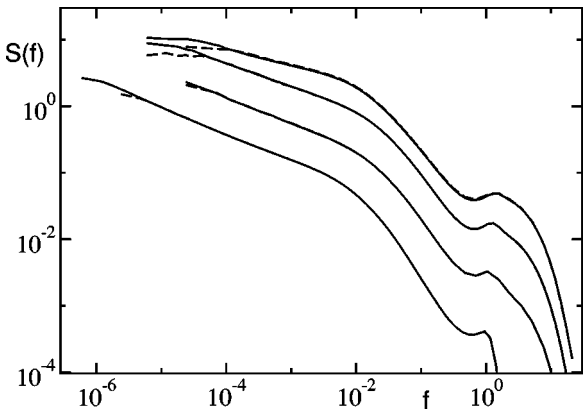

FIG. 1. Power spectra of the flux J as defined in Eq. (4). The lowermost curves refer to the purely quartic FPU model (7) with $N = 2048$ (solid line) and 1024 (dashed line). The upper curves correspond to the repulsive FPU model—Eq. (10)—for $a = 2$, 2.3, and 2.5 (from top to bottom) and $N = 1024$ (solid line) or 512 (dashed line). All microcanonical simulations are performed for the same energy density $e = 1$ with time step $h = 0.05$ for 10^6–10^7 steps. For clarity, the curves have been arbitrarily shifted along the vertical axis.

First, we have performed equilibrium microcanonical simulations by integrating Eqs. (2) (with periodic boundary conditions) with a fourth-order symplectic algorithm [18]. The power spectra $S(f)$ of J are reported in Fig. 1. The lowest curves are data for the quartic FPU model obtained by averaging over 30 000 random initial conditions. In order to further decrease statistical fluctuations, a binning of the data over contiguous frequency intervals has been performed as well.

The long-time tail (1) manifests itself as a power-law divergence f^{δ} in the low-f region. By comparing the results obtained for different numbers of particles, one can clearly see that finite-size corrections are negligible above a size-dependent frequency $f_c(N)$. By fitting the data in the scaling range $[f_c(N), f_s]$, where $f_s \simeq 10^{-3}$, we find $\delta = -0.39(6)$. These values are consistent with previous, less-accurate, findings for similar models, such as the standard FPU [8,14] and the diatomic Toda [10] chains, thus confirming the expectation that they all belong to the same universality class.

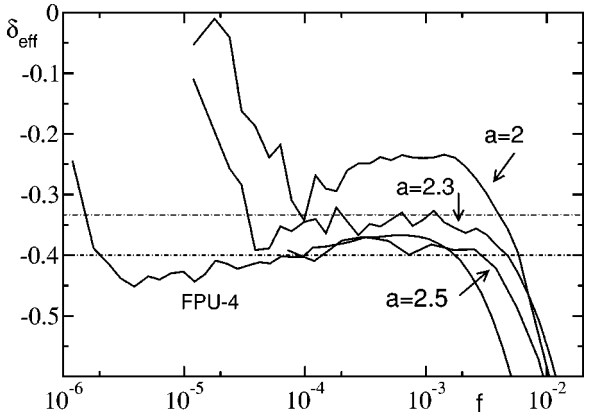

FIG. 2. The logarithmic derivative δ_{eff} of the energy-flux spectrum versus the frequency f for the pure FPU quartic potential—FPU-4 with $N=2048$—and for model (10) with $a=2$, $a=2.3$, and $a=2.5$ and $N=1024$, respectively. The two horizontal lines correspond to the theoretical predictions $-1/3$ and $-2/5$. The statistical error is on the order of the observed irregular fluctuations.

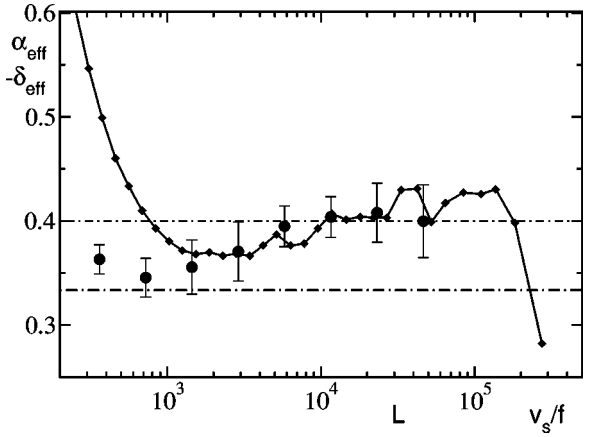

FIG. 3. Quartic FPU model: the effective exponent α_{eff} of the finite-size conductivity for $T_+=1.2, T_-=0.8$ (solid circles), compared with the results ($-\delta_{\text{eff}}$) of equilibrium simulations. The two horizontal lines correspond to the theoretical predictions 1/3 and 2/5.

In order to perform a more stringent test of the scaling behavior, we have determined the logarithmic derivative

$$\delta_{\text{eff}}(f)=\frac{d\ln S}{d\ln f} \tag{8}$$

for different frequencies. Since finite-size effects are responsible for the saturation of $S(f)$ when $f\to 0$, $f_c(N)$ can be identified (see Fig. 2) as the frequency below which δ_{eff} starts growing towards zero. Above f_c, the quality of our numerical data allows revealing a slow but systematic decrease of δ_{eff} upon decreasing f, which approaches -0.44, a value that is incompatible not only with the renormalization-group prediction of Ref. [9], but also with the result of mode-coupling theory [7,8]. Furthermore, convergence seems not fully achieved in the accessible frequency range.

Accordingly, it is advisable to look at thermal conductivity by means of nonequilibrium simulations too. It is sufficient to measure the heat flux in a system put in contact with two heat reservoirs operating at different temperatures T_+ and T_-. Several methods have been proposed based on both deterministic and stochastic algorithms [2]. Regardless of the actual thermostatting scheme, after a transient, an off-equilibrium stationary state sets in, with a net heat current flowing through the lattice. The finite-size thermal conductivity $\kappa(L)$ is then estimated as the ratio between the average flux $\overline{J}$ and the overall temperature gradient $(T_+-T_-)/L$. Notice that, by this definition, κ amounts to an effective transport coefficient including both boundary and bulk scattering mechanisms.

We have used the Nosé-Hoover thermostats described in detail in Ref. [2]. In order to fasten the convergence towards the stationary state, the initial conditions have been generated by thermostatting each particle to yield a linear temperature profile (see [1]). This method is very efficient, especially in long chains, when bulk thermalization may be significantly slow. The heat flux $\overline{J}$ has been obtained by integrating the equations over more than 10^6 time units and by further averaging over a set of about 30 initial conditions. Simulations of the quartic FPU model with chains of length up to 65 536 sites and free boundary conditions exhibit again a systematic increase of the effective exponent

$$\alpha_{\text{eff}}(L)=\frac{d\ln\kappa}{d\ln L}, \tag{9}$$

as can be seen from the solid circles in Fig. 3, although one can also observe that the four rightmost values are in very good agreement with the mode-coupling exponent.

In order to compare more closely equilibrium and nonequilibrium simulations, one can assume, following the argument exposed below Eq. (1), that the finite-size conductivity $\kappa(L)$ is determined by correlations up to time $\tau=L/v_s$, where v_s is the sound velocity. This means that the frequency f can be turned into a length $L=v_s/f$. It might be argued that the absence of a quadratic term in Eq. (7) prevents a straightforward definition of such a velocity in the $T=0$ limit; nevertheless, it has been shown [19] that an effective phonon dispersion relation at finite energy density can be evaluated for model (7), yielding $v_s=1.308$ at $e=1$. Using this value, we can ascertain that, at least for $N>1000$, there is an excellent agreement between the two approaches (see again Fig. 3).

The data presented so far rule out the value predicted in Ref. [9] for the model (7). On the other hand, such a prediction is consistent with numerical results for hard-core interactions [11,12]. In order to test for universality we thus tried to bridge the two classes of models by introducing a strong repulsive potential. This has also the merit of removing one of the main drawbacks of a potential like Eq. (7)—namely, that negative values of $x_{n+1}-x_n$ are allowed—i.e., that particles can formally cross each other if x_n is interpreted as their actual position. This unphysical feature may somehow be circumvented by introducing explicitly a physical distance. For this purpose, we have added to the FPU potential a repulsive term of the Lennard-Jones (LJ) form

$$V(z)=\frac{1}{4}(z-a)^4+\frac{1}{12}\frac{1}{z^{12}}-V_0, \tag{10}$$

where V_0 is a suitable constant needed to set the minimum of the energy equal to 0. The core repulsion introduces a further time scale—namely, that of mutual ''collisions'' between particles [20]. Upon increasing a, at fixed energy, the role of the repulsive term becomes negligible and model (10) reduces to the purely quartic FPU (7). For instance, in the region where $V(z)<e$ the LJ energy contribution can be as large as 0.57 for $a=2$ and $e=1$, but it is at most 0.028, when a is increased to 2.5. Upon decreasing a, the LJ term progressively affects the high-frequency spectral range. This is because core repulsion becomes more relevant close to the minimum.

In this context, one should, in principle, refer to the general heat-flux expression (3), which in the limit of pure hard points reduces to $\Sigma_n \dot{x}_n^3/2$. Nevertheless, in the parameter range investigated hereby, the spectra of this quantity never exceed 10% of the spectra of Eq. (4) in all frequency channels and, more importantly, hardly show any singular low-frequency behavior. We have therefore kept determining the power spectrum of the flux as defined in Eq. (4).

The effect of the LJ term on the low-frequency behavior of $S(f)$ can be appreciated already for $a=2.3$. A direct fitting of the three upmost curves in Fig. 1 (in the available scaling ranges) yields δ decreasing from $-0.25(0)$ for $a=2.0$ up to $-0.37(8)$ for $a=2.5$. Having averaged the spectra over more than 5000 different samples, it is possible to investigate the convergence of each δ value through the effective exponent (8). Like in the quartic FPU model, one can see from Fig. 2 that δ_{eff} decreases upon decreasing frequency, even on the smaller available range. Although on the basis of numerics alone one cannot exclude that the asymptotic δ value depends on a, it is wiser to conjecture that the stronger the LJ term, the slower is the onset of the asymptotic regime.

Altogether our simulations do not confirm the claim contained in Ref. [9] that the hydrodynamic theory accounts for all 1D models. The exponent α is found to be definitely larger than the expected value 1/3 and certainly closer to the mode-coupling estimate 2/5. Anyhow, the systematic deviations shown in Fig. 2 make also the convergence to this latter value somehow questionable. In addition, we have also shown that the low-frequency power-law behavior is strongly influenced by the presence of a hard-core repulsion term: even small variations of the spatial scale associated with the equilibrium distance between interacting oscillators enhance finite-size effects and slow down convergence with respect to the purely anharmonic model. This scenario rather suggests that at least two different universality classes may exist, although their physical origin is up to now unclear.

A similar analysis should now be applied to the other models recently considered, in order to determine how much of the observed mutual fluctuations are the result of finite-size corrections. However, since we have basically reached the limit of our computing facilities letting a cluster of 48 PC's run for 2 months, it is also clear that more refined analytic estimates have to be worked out to shed light on this puzzling scenario.

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[1] S. Lepri, R. Livi, and A. Politi, Phys. Rev. Lett. **78**, 1896 (1997).
[2] S. Lepri, R. Livi, and A. Politi, Phys. Rep. **377**, 1 (2003).
[3] Y. Pomeau and R. Résibois, Phys. Rep. **63**, 19 (1975).
[4] S. Berber, Y. Kwon, and D. Tomanek, Phys. Rev. Lett. **84**, 4613 (2000).
[5] S. Maruyama, Physica B **323**, 193 (2002).
[6] C. Giardinà *et al.*, Phys. Rev. Lett. **84**, 2144 (2000); O.V. Gendelman and A.V. Savin, *ibid.* **84**, 2381 (2000).
[7] M.H. Ernst, Physica D **47**, 198 (1991).
[8] S. Lepri, R. Livi, and A. Politi, Europhys. Lett. **43**, 271 (1998).
[9] O. Narayan and S. Ramaswamy, Phys. Rev. Lett. **89**, 200601 (2002).
[10] T. Hatano, Phys. Rev. E **59**, R1 (1999).
[11] P. Grassberger, W. Nadler, and L. Yang, Phys. Rev. Lett. **89**, 180601 (2002).
[12] J.M. Deutsch and O. Narayan, Phys. Rev. E **68**, 010201 (2003).
[13] A. Dhar, Phys. Rev. Lett. **86**, 3554 (2001); G. Casati and T. Prosen, Phys. Rev. E **67**, 015203(R) (2003).
[14] P. Grassberger and L. Yang, e-print cond-mat/0204247.
[15] T. Prosen and D.K. Campbell, Phys. Rev. Lett. **84**, 2857 (2000).
[16] M.S. Green, Phys. Rev. **119**, 829 (1960).
[17] The average lattice spacing b does not need to coincide with the minimum a of V. This happens only if no pressure is exerted. As we do not expect pressure to play any role on the scaling behavior of physical observables, for simplicity we always fix $b=a$.
[18] R.I. Mclachlan and P. Atela, Nonlinearity **5**, 541 (1992).
[19] C. Alabiso and M. Casartelli, J. Phys. A **34**, 1223 (2001).
[20] In the absence of a hard core the very definition of collision is somehow arbitrary. We say that a collision occurs when $x_{n+1}-x_n$ is equal to the distance at which the LJ force term is 10 times larger than the FPU one.