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Heat conduction in a chain of colliding particles with a stiff repulsive potential

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One-dimensional billiards, i.e., a chain of colliding particles with equal masses, is a well-known example of a completely integrable system. Billiards with different particle masses is generically not integrable, but it still exhibits divergence of a heat conduction coefficient (HCC) in the thermodynamic limit. Traditional billiards models imply instantaneous (zero-time) collisions between the particles. We relax this condition of instantaneous impact and consider heat transport in a chain of stiff colliding particles with the power-law potential of the nearest-neighbor interaction. The instantaneous collisions correspond to the limit of infinite power in the interaction potential; for finite powers, the interactions take nonzero time. This modification of the model leads to a profound physical consequence—the probability of multiple (in particular triple) -particle collisions becomes nonzero. Contrary to the integrable billiards of equal particles, the modified model exhibits saturation of the heat conduction coefficient for a large system size. Moreover, the identification of scattering events with triple-particle collisions leads to a simple definition of the characteristic mean free path and a kinetic description of heat transport. This approach allows us to predict both the temperature and density dependencies for the HCC limit values. The latter dependence is quite counterintuitive—the HCC is inversely proportional to the particle density in the chain. Both predictions are confirmed by direct numerical simulations.

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A microscopic description of heat conduction in dielectrics remains an open and elusive problem despite a rather long history [1–4] and intensive research efforts over the past two decades [5–12]. One of the most intriguing questions involves the convergence of the heat conduction coefficient (HCC) in the thermodynamic limit [5,11,12]. A common understanding achieved as a result of these efforts suggests that in lattices with low-order polynomial nonlinearity (for instance, the famous Fermi-Pasta-Ulam lattice [5]), the behavior of HCC strongly depends on dimensionality. Namely, in one-dimensional lattices it diverges in the thermodynamic limit as L^{δ}, $0.3 \leqslant \delta \leqslant 0.4$, where L is the size (or number of particles) in the system. For two-dimensional models, the HCC is believed to behave as $\ln(L)$ [11,13], and for three dimensions, finally, it converges [14]. This common understanding is supported by solid theoretical arguments based on a combination of different approaches [10,15–18]. These approaches provide somewhat different estimations for the divergence exponent α (within the range of measured values for different model potentials), but in general this part of the picture seems self-consistent.

At the same time, it has long been claimed that in some one-dimensional chains, for instance in the chain of rotators [19,20], the HCC converges in the thermodynamic limit despite the momentum conservation. More recent results of this sort, namely the HCC convergence in a Lennard-Jones (LJ) chain, were reported in [21]. In this paper, similar convergent behavior has been claimed also for the α-β FPU chain and attributed to the asymmetry of the interaction potential. This latter claim for the α-β FPU has been disproved in [22]; the LJ chain has not been addressed there.

From a physical point of view, the low-order polynomial nonlinearity of the FPU and similar models arises as Taylor truncation of the complete interaction potential. The possibility of such truncation, "self-evident" at least for low temperatures, seems, however, problematic in the thermodynamic limit; recall that the latter corresponds to the infinite size of the system and infinite time. Any realistic physical potential of interaction should tend to zero as the interacting atoms are separated by a large distance—in other words, it should allow dissociation, as in the LJ chain. The polynomial truncation definitely fails to describe this feature and yields instead an unphysical infinite attraction force at large distances. In a realistic system, the dissociation or formation of abnormally long links between particles has an exponentially small but nonzero probability even at low temperatures. The polynomial truncation precludes this phenomenon. Such long links can presumably scatter phonons quite efficiently, and thus they could modify the HCC convergence properties. Further results on the HCC convergence in many one-dimensional models with the possibility of dissociation were reported in [23,24]. The HCC convergence in systems of LJ particles and particles with an elastic shell has been observed in a number of additional studies [25].

At the same time, a recent treatise on the nonequilibrium hydrodynamics of anharmonic chains [26] pointed to an important difference between the aforementioned model of rotators and the models similar to the FPU or LJ chains. The difference is a number of conservation laws; for the chain of rotators, only the total momentum and energy are conserved. In addition, in FPU, LJ, and similar chains, a total length of the system is conserved. This additional conservation law obviously does not depend on the possibility of dissociation. This qualitative difference is believed to lead to a difference in the HCC convergence properties [26]. From this point of view, in the thermodynamic limit, all nonintegrable chains with the three conservation laws mentioned above should behave in a qualitatively similar manner and thus have the divergent HCC. From this viewpoint, the observed convergence in the LJ chain,

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a chain of semielastic particles, and similar dissociating chains may be interpreted as the finite-size effect. Such “finite-size” saturation of the HCC with resumed growth for larger system sizes has been demonstrated in α-β FPU [22] and in a chain of rigid particles with alternating masses [27]. To the best of our knowledge, no “resumed” growth of the HCC in LJ or similar models has been reported so far. At the same time, one should admit that any numerical simulation in general cannot prove (or disprove) the HCC convergence in the thermodynamic limit for any model. To be on the safe side, we refer to the observed phenomenon as saturation of the HCC for a certain large scale of the system, without an explicit claim of convergence. In the LJ and similar models with dissociation, this saturation occurs at a scale of 10^4–10^5 particles. For typical interatomic distances, such a specimen will have a length of order 1–10 μm.

The HCC saturation in the LJ chain and the chain of semielastic particles have one more important common feature. The HCC behavior in the saturation regime can be interpreted in terms of simple kinetic theory [24]. For the chain of semielastic particles, one can predict the dependence of the HCC on the temperature and other system parameters. Similar estimations (to a lesser extent) are available also for the LJ chain. This simple kinetics seems related to the observed exponential decay of the autocorrelation of the heat flux in the saturation regime.

The claim of HCC saturation in dissociating chains has a profound counterexample, or even a group of counterexamples [12]. One-dimensional (1D) billiards of perfectly rigid colliding particles with equal masses has obviously divergent heat conductivity. Moreover, this model is completely integrable and therefore unable to form even a linear temperature profile when attached to thermostats. For the point 1D billiards, this integrability is preserved even in the presence of an on-site potential [28]. Other billiards models are not integrable, but they also exhibit divergent heat conductivity [29].

The current paper addresses this group of counterexamples. In traditional billiards models, the collisions between the particles are instantaneous, i.e., they take zero time. Such behavior requires an infinitely large interaction force. So, the potential of interaction between such particles includes a vertical potential wall. Such instantaneous collisions are apparently unphysical, since a repulsive core of any realistic interatomic potential grows rapidly, but with a finite rate at nonzero distances. Therefore, the realistic interparticle collision will take some finite, maybe very small, but nonzero time. We claim that this peculiarity leads to a drastic change in the transport properties of the 1D chain, since it makes the probability of triple collisions nonzero. In the case of equal particle masses, the double collisions, even with finite interaction time, do not violate integrability. The reason is that, as a result of momentum and energy conservation, the colliding particles with equal mass just exchange their momenta, similarly to the instantaneous collisions. The triple collisions, however, do violate the integrability. We are going to demonstrate that they also bring about HCC saturation in the 1D case. So, similarly to the case of the FPU-type chains, correction of the unphysical features of the interaction potential may lead to a significant modification of the heat transport properties, at least at the saturation mesoscale.

To demonstrate that, we consider a chain of one-dimensional particles with the following purely repulsive potential of the nearest-neighbor interaction:

$$\begin{aligned} &V(r)=0 \quad \text{for } r>D,\\ &V(r)=V_l(r)=K|D-r|^{\alpha} \quad \text{for } r\leqslant D. \end{aligned} \tag{1}$$

Here, $\alpha\geqslant 2$ is the parameter that governs the growth of the repulsive force, r is the distance between the centers of neighbor particles, and D is the size of the rod. The case $\alpha=2$ corresponds to the semielastic particles considered in [24]; $\alpha=5/2$ corresponds to the case of Hertzian contact. Without restricting generality, we further use the nondimensional parameters $D=1$ and $K=1$. Then, the limit $\alpha\to\infty$ corresponds to the case of a perfect instantaneous elastic collision as $r=1$.

To avoid numerical problems related to the nonanalyticity of potential (1) at $r=1$, it is substituted in the simulations by a smoothed potential function,

$$V_h(r)=2^{-\alpha}[\sqrt{\rho^2+hf(\rho)}-\rho]^{\alpha},\quad \rho=r-1, \tag{2}$$

where the function $f(\rho)=1/(1+5\rho^2)^6$, and the parameter $h>0$. In the limit $h\to 0$, the smoothed potential (2) tends to the nonanalytic potential (1).

We perform a traditional numerical simulation of equilibrium heat transport in a one-dimensional model, and we consider a segment of length L parallel to the x axis. $N=p(L-1)+1$ particles are packed along this segment, where p $(0<p<1)$ stands for the packing “density” of the chain. Fixed boundary conditions are imposed on both ends of the chain, i.e., $x_1\equiv 0$, $x_N\equiv(N-1)a$, where $a=1/p$ stands for the period of the unperturbed chain. Fixed boundaries enforce the density conservation. The particles $1<n<N$ are then restricted to move in the x direction. The Hamiltonian of the chain in this case is expressed as

$$\mathcal{H}=\sum_{n=2}^{N-1}\frac{1}{2}{x_n'}^2+\sum_{n=1}^{N-1}V(x_{n+1}-x_n). \tag{3}$$

Here $\{x_n\}_{n=1}^N$ are the coordinates of the rod centers.

To model the heat transfer along the chain under consideration, stochastic Langevin thermostats are used. The left end $(x<L_0=10)$ of the chain is attached to the Langevin thermostat with temperature T_+, and the right end of the chain with the same length $[x>(N-1)a-L_0]$ is attached to the thermostat with temperature T_-. We adopt $T_\pm=(1\pm 0.05)T$, where T is the average temperature of the chain. The corresponding equations of motion have the following form:

$$\begin{aligned} &x_n''=-\partial\mathcal{H}/\partial x_n-\gamma x_n'+\xi_n^+ \quad \text{if } x_n<L_0,\\ &x_n''=-\partial\mathcal{H}/\partial x_n \quad \text{if } L_0\leqslant x_n\leqslant(N-1)a-L_0,\\ &x_n''=-\partial\mathcal{H}/\partial x_n-\gamma x_n'+\xi_n^- \quad \text{if } x_n>(N-1)a-L_0, \end{aligned} \tag{4}$$

where $\gamma=0.1$ is a damping coefficient, $\xi_n^\pm$ is Gaussian white noise, which models the interaction with the thermostats, and it is normalized as $\langle\xi_n^\pm(\tau)\rangle=0$, $\langle\xi_n^+(\tau_1)\xi_k^-(\tau_2)\rangle=0$, $\langle\xi_n^\pm(\tau_1)\xi_k^\pm(\tau_2)\rangle=2\gamma T_\pm\delta_{nk}\delta(\tau_2-\tau_1)$.

The system of Eqs. (4) with initial conditions $\mathbf{X}(0)=\{x_n(0)=(n-1)a,x_n'(0)=0\}_{n=1}^N$ was integrated numerically using the velocity Verlet method [30]. Then, after some initial transient, a stationary heat flux J and the stationary local temperature distribution $T(x)$ are achieved.

The total heat flux J was measured as the average work produced by the thermostats over unit time. For this purpose, at each step of numerical integration $\Delta\tau$ new coordinates of the particles were calculated without taking into account the interaction with thermostats $\mathbf{X}_0(\tau+\Delta\tau)$, and then the same coordinates were calculated for a chain interacting with the thermostats, denoted as $\mathbf{X}(\tau+\Delta\tau)$. We define E_+ as the energy of the leftmost segment of the chain, which consists of particles with coordinates $x_n < L/2$, and we define E_- as the energy of the rightmost segment, where the particles have coordinates $x_n > L/2$. Then the work done by the external forces in the time interval $[\tau,\tau+\Delta\tau]$ is expressed as

$$j_\pm = \{E_\pm[\mathbf{X}(\tau+\Delta\tau)] - E_\pm[\mathbf{X}_0(\tau+\Delta\tau)]\}/\Delta\tau. \qquad (5)$$

By taking the time average $J_\pm = \langle j_\pm\rangle_\tau$, we obtain the average value of the energy flux-out from the left "hot" thermostat and the average value of the energy flux-in into the right "cold" thermostat. The value of the energy flux along the chain is $J = J_+ = -J_-$. The accuracy of this balance is considered one of the criteria for the validity of the numerical procedure.

The local heat flux, i.e., the energy flow from particle n to the neighboring particle $n+1$, is defined as $J_n = \langle j_n\rangle_\tau$, where

$$j_n = (x_{n+1}-x_n)(x'_{n+1}+x'_n)F(x_{n+1}-x_n)/2 + x'_n h_n,$$

the function $F(r) = -dV(r)/dr$, and the energy density distribution along the chain is

$$h_n = \left[{x'_n}^2 + V(x_n - x_{n-1}) + V(x_{n+1}-x_n)\right]/2$$

(see [11]).

Thermal equilibrium requires all local fluxes to be equal to the total heat flux multiplied by the chain period, $J_n = aJ$. The fulfillment of this requirement may be considered a criterion for the stationary regime of heat transport.

The local temperature distribution of the chain is calculated from the kinetic energy of the rod. We divide the line segment L, which consists of N particles, into unit-length cells $[i-1,i]$, $i = 1,\ldots,L$, and we define the following quantities: the average number of particles in the ith cell is $\bar{n}_i$, and the average kinetic energy in the cell is $\bar{E}_i$. Then the temperature of the cell is defined as $T(i) = 2\bar{E}_i/\bar{n}_i$. This latter definition is caused by a classical consideration of the heat transport in the current work. In quantum simulations, the definition of temperature causes major problems since the system sometimes does not thermalize under its own dynamics, even if it is attached to the thermostats [31,32].

Between the thermostats we observe a linear temperature gradient $T(n)$ and constant thermal flux J. So, the heat conduction coefficient of the free fragment of the chain between the thermostats (of length $L-20$) can be estimated as follows:

$$\kappa(L) = J[T(11) - T(L-10)]/(L-20). \qquad (6)$$

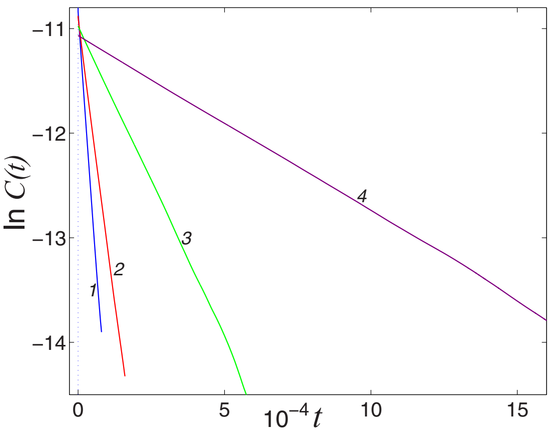

FIG. 1. Exponential decay of the autocorrelation function $C(t)$ for the chain of length $N = 10\,000$ for temperature $T = 0.033$ and density $p = 0.25$. Curves 1, 2, 3, and 4 correspond to $\alpha = 2$, 2.5, 4, and 6, respectively.

A well-known alternative way to evaluate the heat conduction coefficient is based on the well-known Green-Kubo formula

$$\kappa = \lim_{\tau\to\infty}\lim_{N\to\infty}\frac{1}{NT^2}\int_0^\tau C(s)ds, \qquad (7)$$

where $C(s) = \langle J_{\rm tot}(t)J_{\rm tot}(t-s)\rangle_t$ is the autocorrelation function of the total heat flux in the chain with periodic boundary conditions $J_{\rm tot}(t) = \sum_{n=1}^N j_n(t)$.

To compute the autocorrelation function $C(t)$, we consider a cyclic chain consisting of $N = 10^4$ particles. Initially all the particles in this chain are coupled to the Langevin thermostat with temperature T. After achieving thermal equilibrium, the system is detached from the thermostat, and the Hamiltonian dynamics is simulated. To improve the accuracy, the results were averaged over 10^4 realizations of the initial thermal distribution.

Numerical simulation of the thermalized cyclic chain of the particles had demonstrated that the autocorrelation function of the heat flux $C(t)$ decreases exponentially as $t\to\infty$; see Fig. 1. Consequently, the integral in the Green-Kubo formula (7) converges, yielding a finite value for the HCC in the chain. Direct numerical simulation of the heat transport between the thermostats also yields saturation of $\kappa(L)$ for large values of L; see Fig. 2. Both methods of simulation yield similar values of the HCC in the saturation regime.

As was mentioned before, we hypothesize that the observed HCC saturation may be attributed to the triple collisions between the particles. To describe the transport process, it is convenient to define a set of quasiparticles associated with the momenta of individual particles [28]. The quasiparticles are not affected by the double collisions (the momenta just hop to the next particles), but they are scattered by the triple collisions. Therefore, at the phenomenological level, one can evaluate the HCC in terms of kinetic theory in the following way:

$$\kappa \sim c\lambda v \sim p\lambda v. \qquad (8)$$

Here v is the characteristic velocity of the quasiparticles, c is the heat capacity of the system, and λ is the mean free path of the quasiparticles. Scattering events are related to triple

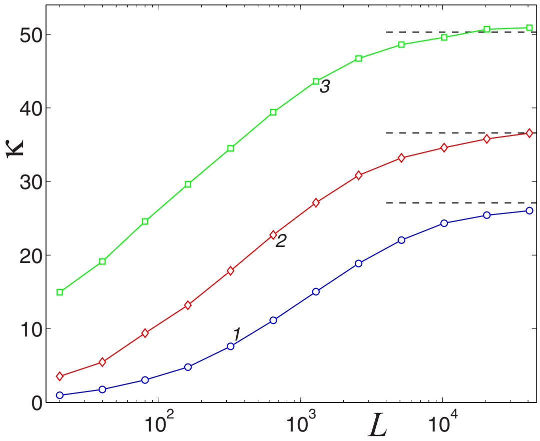

FIG. 2. Dependence of the heat conduction coefficient κ on the distance between thermostats L for temperatures $T = 0.001$ (curve 1), 0.01 (curve 2), 0.1 (curve 3), and fixed density. The system includes particles with diameter $d = 1$ and density $p = 0.5$ (the average distance between the particles $a = 1/p = 2$), where the power of the potential function is $\alpha = 5/2$. Straight dashed lines correspond to the results obtained from the Green-Kubo formula.

collisions, so the mean free path corresponds to the distance traveled by the quasiparticle between such triple collisions:

$$\lambda \sim 1/pP_{\text{tr}}, \tag{9}$$

where P_{tr} is the probability that a given collision is triple. This probability can be estimated as

$$P_{\text{tr}} \sim \tau_c/\tau_f. \tag{10}$$

Here τ_f is the time of flight between two successive collisions, and τ_c is the characteristic time of collision. One can also estimate $\tau_f \sim L/pv$ and therefore

$$\kappa \sim 1/p\tau_c. \tag{11}$$

Evaluation of the time of collision is simple due to the finite range of interaction. If the particles collide with relative velocity v_0 at infinity, then the integral of energy for the two-particle system reads

$$\tfrac{1}{2}\dot{x}^2 + x^\alpha = \tfrac{1}{2}v_0^2, \tag{12}$$

where $x(t)$ is the non-negative relative displacement of the particles. The relative velocity becomes zero at the distance $x_m = (v_0^2/2)^{1/\alpha}$. The time of collision is presented as

$$\begin{aligned}\tau_c &= 2\int_0^{x_m} \frac{dx}{\sqrt{v_0^2 - x^\alpha}}\\ &= 2^{1-1/\alpha} v_0^{2/\alpha-1} \int_0^1 \frac{d\xi}{\sqrt{1-\xi^\alpha}}, \quad \xi = x/x_m. \end{aligned}\tag{13}$$

Summarizing Eqs. (10), (11), and (13), and adopting $v_0 \sim T^{1/2}$, one obtains

$$\tau_c \sim v_0^{2/\alpha-1} \sim T^{1/\alpha-1/2} \Rightarrow \kappa \sim f(\alpha)p^{-1}T^{1/2-1/\alpha}. \tag{14}$$

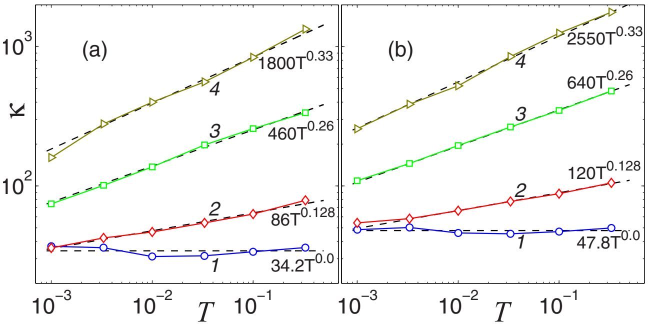

FIG. 3. Temperature dependence of the HCC for density (a) $p = 0.375$ and (b) $p = 0.25$. Curves 1, 2, 3, and 4 correspond to $\alpha = 2$, 2.5, 4, and 6, respectively. Straight dashed lines correspond to the power functions $\kappa = bT^\beta$.

Equation (14) predicts two important features of the HCC in the considered model. First, it has a nontrivial temperature dependence. Second, quite surprisingly, it is inversely proportional to the concentration of the particles.

Predictions for the scaling exponents in simulations of Eq. (14) completely conform to the numerical results presented in Figs. 3 and 4. At the same time, the scaling function $f(\alpha)$ in (14) remains undetermined. We suggest that it is completely governed by the intricate dynamics of the three-particle collisions and thus depends solely on exponent α. To verify that, we plot the function $f(\alpha) = \kappa p T^{1/\alpha-1/2}$ versus exponent α.

Figure 5 presents a clear collapse of all available numerical data according to the above scaling function; the results suggest the power law $f \approx 1.12\alpha^{3.6}$. As expected, the HCC rapidly increases as a power function of α.

For all explored values of α, we observe the HCC saturation. Moreover, the observed scaling with concentration and temperature allows us to conclude that the observed saturation is caused by the triple-particle collisions. Therefore, modification of the billiards model and removal of unphysical instantaneous collisions lead to a drastic modification of the transport properties, namely the observed HCC saturation. As was mentioned above, it is not possible to claim convergence in the thermodynamic limit on the basis of numeric data for

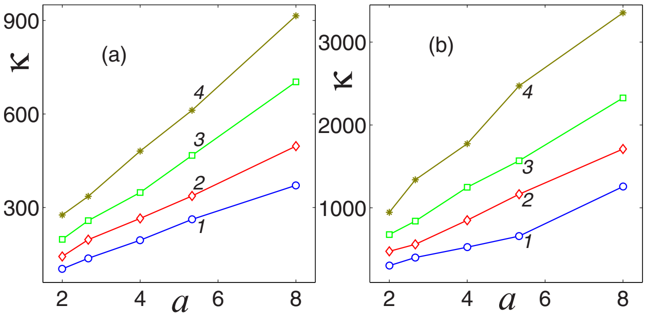

FIG. 4. Dependence of the HCC on inverse density $a = 1/p$ for fixed temperature and parameter (a) $\alpha = 4$ and (b) $\alpha = 6$. Curves 1, 2, 3, and 4 correspond to $T = 0.01$, 0.033, 0.1, and 0.33, respectively.

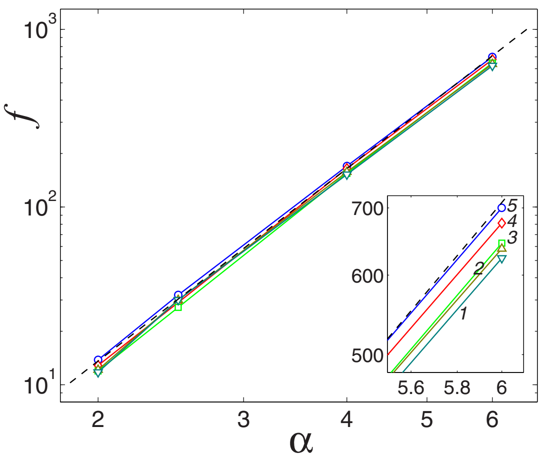

FIG. 5. Dependence of the scaling function $f(\alpha)$ on power α for all simulated values of temperatures and particle densities $p = 0.125$, 0.1875, 0.25, 0.375, and 0.5 (curves 1, 2, 3, 4, and 5, respectively, to guide the eye). The fine details of the data collapse are presented in the inset. The best linear fit (dashed line 6) corresponds to $f(\alpha) = 1.12\alpha^{3.6}$.

a finite system. Still, we do not observe any trend toward "resumed growth" of the HCC, similar to what was observed in the asymmetric FPU chain [22] and billiards with alternating masses [27]. Of course, it might happen that simulations of even longer chains would demonstrate such growth also in the considered chain of stiff colliding particles. However, contrary to the asymmetric FPU and alternating-mass billiards, the considered model also enables a clear and verifiable definition of the basic kinetic parameters. The chains with a possibility of dissociation possess a similar property [24]. Intuitively, as the mean free path can be defined, longer chains are expected to conform even better to the simple kinetic estimation of the heat conduction coefficient. Needless to say, this latter argument also does not prove anything, and further explorations are required to verify whether the considered model belongs to a universality class that is different from the FPU chain.

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