

## Thermal conductance of the Fermi-Pasta-Ulam chains: Atomic to mesoscopic transition

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We demonstrate that in the atomic-scale limit the thermal conductance  $\mathcal{K}$  of the Fermi-Pasta-Ulam (FPU) model and its variants strongly deviates from the mesoscopic behavior due to the relevance of contact resistance. As a result, atomic chains follow  $\log \mathcal{K} = \nu \log T$ , where the power-law coefficient  $\nu$  is exactly two times larger than the mesoscopic value. We smoothly interconnect the atomic and mesoscopic limits and demonstrate that this turnover behavior takes place in other nonlinear FPU-like models. Our results are significant for nanoscale applications, manifesting an atomic thermal conductance with temperature scaling superior to the mesoscopic limit.

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The Fermi-Pasta-Ulam (FPU) model [1] and its variants provide an ideal test bed for addressing fundamental issues in statistical mechanics such as equipartition of energy, the onset of chaos in nonlinear dynamical systems, and the validity of macroscopic laws in low dimensional systems [2]. In particular, the thermal conductivity of the FPU model has been extensively investigated, demonstrating a breakdown of the normal-diffusional Fourier's law dynamics [3]. The underlying question addressed in these studies has typically been whether a one-dimensional (1D) chain of oscillators with a specific force field can demonstrate equilibrium and dynamical properties characterizing macroscopic objects. Thus, simulations were mostly carried out using long chains of  $10^3$ – $10^4$  beads, targeting the mesoscopic-macroscopic limit.

Nonetheless, in recent years the thermal conduction properties of *nanoscale junctions* have been of fundamental and practical interest [4]. Here, the physical setup includes a nanoscale object, e.g., a nanotube [5] or an alkane molecule [6], coupled to two (or more) thermal contacts. As devices' size shrinks, an enormously important question is the applicability of the macroscopic dynamical laws at the nanoscale. Even in the framework of classical mechanics, contact effects and the constriction geometry may imply on the onset of new scaling rules. Furthermore, one of the major problems in molecular electronics is junction heating, limiting the device stability. Understating heat transport in molecular systems thus attracts considerable attention both experimentally [4,6–8] and theoretically [9–13]. Managing thermal transport across interfaces [14] is also important in microelectronic devices [4], biophysical applications [15], and in thermoelectric energy conversion devices [16].

Motivated by these challenges we focus here on the *atomic-scale steady-nonequilibrium FPU model* and its variants. As the effect of the contact (interface) and internal nonlinear interactions cannot be trivially separated, a complex new dynamics is revealed at different temperature domains for both weak and strong system-bath interaction strengths. Specifically, using numerical simulations we demonstrate that when the contact resistance controls the dynamics; e.g., for extremely short chains or at relatively low temperatures (yet above the harmonic-anharmonic transition), the conductance of FPU-type systems follows a  $\mathcal{K} \propto T^\nu$  power law,

where  $\nu=1/2$  for the  $\beta$ -FPU model. This behavior stands in a sharp contrast to the mesoscopic  $\beta$ -FPU limit where the value  $\nu=1/4$  is obtained, as expected from the phenomenological Debye theory. We justify our results within the effective-phonon theory [17,18] and further interconnect the atomic and mesoscopic limits, demonstrating a smooth turnover of the dynamics with increasing chain size and temperature.

We consider a 1D lattice of  $N$  atoms whose Hamiltonian reads as

$$H = \sum_{i=1}^N \frac{p_i^2}{2} + g_2 \Omega^2 V_2 + g_4 \beta V_4,$$

$$V_2 = \frac{1}{2} \sum_{i=0}^N (x_{i+1} - x_i)^2; \quad V_4 = \frac{1}{4} \sum_{i=0}^N (x_{i+1} - x_i)^4. \quad (1)$$

Here  $x_i$  is the displacement from the equilibrium position of the  $i$ th particle,  $x_0$  and  $x_{N+1}$  are the fixed boundaries, and  $p_i$  are the particles' momenta. In what follows we assume a homogeneous chain with masses  $m_i=1$  and a force constant  $\Omega_2$ .  $\beta$  measures the strength of nonlinear interactions, and  $g_s$  ( $s=2,4$ ) are Boolean variables taking the values of 0 or 1. In what follows we will consider three models: (i) a harmonic model ( $H_2$ ) with  $g_2=1$ ,  $g_4=0$ , (ii) a quartic model ( $H_4$ ) with  $g_2=0$ ,  $g_4=1$ , and (iii) the  $\beta$ -FPU model ( $H_F$ ) with  $g_2=1$ ,  $g_4=1$ . The quartic model is introduced here to facilitate identifying the dynamics of the FPU model at high temperatures.

The direct way to determine the thermal conductance  $\mathcal{K}$  of a 1D chain is to couple the left and right ends of the system with two thermal baths at temperatures  $T_L$  and  $T_R$ , respectively [3]. In our simulations we use Langevin thermostats, with the motivation to simulate real experiments on the nanoscale, where a small molecule connects to macroscopic solids [6,8]. In such systems contact resistance is an unavoidable issue. The chain's particles obey the following equations of motion:

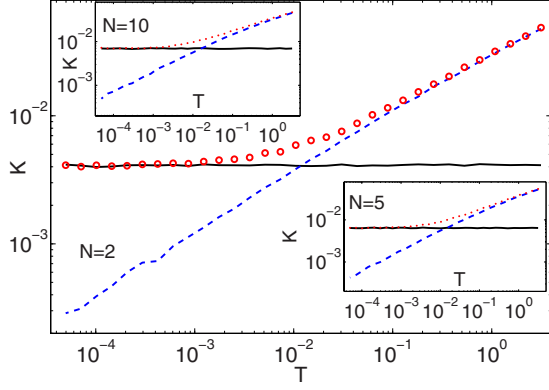


FIG. 1. (Color online) Thermal conductance of atomic FPU chains (circles or dots), a quartic chain (dashed line), and an harmonic chain (full line) as a function of the average temperature  $T = (T_L + T_R)/2$ .  $\gamma_L = \gamma_R = 0.6$ ,  $\Omega^2 = 0.01$ , and  $\beta = 0.01/4$ . The main plot displays data for  $N=2$ . The lower (upper) panel displays data for  $N=5$  ( $N=10$ ).

$$\ddot{x}_i = -\frac{\partial H}{\partial x_i} - (\gamma_R \dot{x}_i - F_R) \delta_{i,N} - (\gamma_L \dot{x}_i - F_L) \delta_{i,1}. \quad (2)$$

where  $\gamma_{L,R}$  reflects the coupling strength between the system and the  $L,R$  bath, and the Gaussian thermal white noises obey the fluctuation-dissipation relation ( $k_B=1$ ),  $\langle F_{L,R}(t) \rangle = 0$ ,  $\langle F_{L,R}(t) F_{L,R}(0) \rangle = 2\gamma_{L,R} T_{L,R} \delta(t)$ . The long-time heat flux can be calculated between every two sites as the force exerted by the  $i$ th particle on the  $i+1$  member,  $J_i = -\langle \dot{x}_i [g_2 \Omega^2 (x_{i+1} - x_i) + g_4 \beta (x_{i+1} - x_i)^3] \rangle$  where the average reflects the time and ensemble average, performed after the system has reached steady state. The thermal conductance is defined as  $\mathcal{K} = \frac{1}{\Delta T} \sum_{i=1}^{N-1} J_i / (N-1)$ , where  $\Delta T = (T_L - T_R) \rightarrow 0$ .

Figure 1 presents the thermal conductance of relatively short chains with  $N=2, 5, 10$ , for the harmonic, quartic, and the FPU models in the strong friction limit using  $\Omega=0.1$ ,  $\gamma = \gamma_{L,R} = 0.6$ ,  $\beta = 0.0025$ , and  $T_L = 0.8T_R$ . The following observations can be made: (i) disregarding anharmonic interactions, the conductance does not depend on temperature, (ii) for  $N=2$  the quartic model shows a power-law dependence,  $\mathcal{K} \sim T^{1/2}$ , (iii) the FPU model smoothly interconnects the harmonic and quartic results, with a crossover temperature around  $10^{-2}$ , and (iv) for long chains the power-law coefficients at low and high temperatures are distinct. The upper inset ( $N=10$ ) demonstrates this effect, to be discussed in more details in Fig. 2. We can explain these results as follows. For a 1D harmonic chain it can be analytically shown [19] that the average stationary thermal flux is given by

$$J = \frac{\Omega^2}{2\gamma} \phi(1) \Delta T, \quad (3)$$

where  $\phi(j) = \frac{\sinh(N-j)\alpha}{\sinh N\alpha}$  and  $e^{-\alpha} = 1 + z/2 - \sqrt{z + z^2/4}$ ,  $z = \Omega^2/\gamma^2$ . In the strong-coupling limit,  $z \rightarrow 0$ , and we obtain  $\alpha \sim \sqrt{z}$ , resulting in  $\phi(1) \sim 1/2$  for  $N=2$  and  $\phi(1) \sim 1$  for  $N \gg 1$ . Thus, the strong-coupling harmonic conductance reduces to

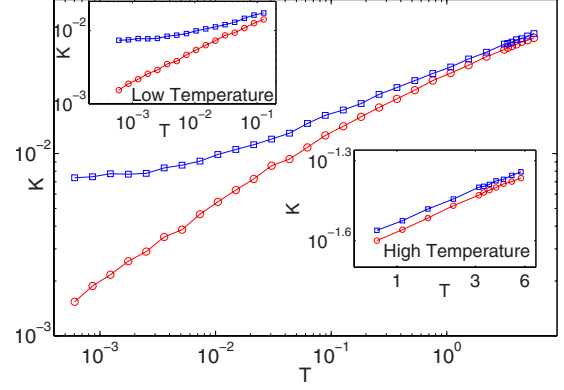


FIG. 2. (Color online) Power-law behavior at low and high temperatures.  $\gamma_L = \gamma_R = 0.6$ ,  $\Omega^2 = 0.01$ ,  $\beta = 0.01/4$ , and  $N=20$ , an FPU chain ( $\square$ ), a quartic chain ( $\circ$ ). The upper and lower panels zoom on data in the low and high-temperature limit, respectively.

$$\mathcal{K}_{N=2} = \frac{\Omega^2}{4\gamma}; \quad \mathcal{K}_{N \rightarrow \infty} = \frac{\Omega^2}{2\gamma}. \quad (4)$$

In the opposite weak-coupling limit ( $z \rightarrow \infty$ ) we get  $\alpha \sim \ln z$ ,  $\phi(1) \sim 1/z$ , and  $\mathcal{K} = \gamma/2$  irrespective of chain size. These harmonic expressions essentially reflect the contact contribution, as the harmonic object perfectly conducts. In the present case, Eq. (3) predicts the conductance values 0.0041, 0.0064, and 0.0070 for the  $N=2, 5, 10$  chains (respectively). Our simulation data perfectly agree with these numbers. At low temperatures, since the dominant contribution to the transport comes from the quadratic term, the FPU model obeys Eq. (3), alike a pure harmonic system.

We turn next to the low-temperature quartic model. Here a power-law dynamics is observed with  $\mathcal{K} \sim T^{1/2}$  ( $N=2$ ). In the high-temperature regime ( $T > 10^{-2}$ ) both FPU and quartic models yield  $\mathcal{K} \sim T^{0.4}$ . These observations stand in sharp contrast to other results observing a  $T^{1/4}$  dependency [20–22]. Note however that these works were concerned with the thermal conductivity of periodic or long anharmonic chains, where contact effects were negligible or nonexistent. In contrast, in short atomic systems contact resistance plays a crucial role [3,8,14]. We consider its influence next.

Assuming that the total thermal resistance of an atomic-scale chain is given by the sum  $R_T = R_M + R_C$ , where  $R_M$  is the bulk molecular resistance and  $R_C$  is the contact resistance, one finds that the total conductance satisfies  $\mathcal{K}_T = \mathcal{K}_M \mathcal{K}_C / (\mathcal{K}_M + \mathcal{K}_C)$ , where  $\mathcal{K}_n = 1/R_n$ ;  $n=T, M, C$ . Since the molecular resistance increases with size (not necessarily linearly), we can generally assume that for short chains  $\mathcal{K}_T \sim \mathcal{K}_C$ , while for long systems  $\mathcal{K}_T \sim \mathcal{K}_M$ . Next we estimate separately the contact conductance  $\mathcal{K}_C$  and the molecular contribution  $\mathcal{K}_M$ .

It is well justified that a sort of Virial theorem holds for nonlinear potentials, resulting in normal-mode spectra which are pseudoharmonic for both the quartic potential and the FPU model [17], with the dispersion relation

$$\tilde{\omega}_k^2 = \alpha \Omega^2 \omega_k^2. \quad (5)$$

Here the integer  $k$  counts the normal modes,  $\omega_k = 2 \sin(k\pi/N)$ , and the renormalization factor satisfies

$$\alpha = g_2 + g_4 \frac{2\beta \langle V_4 \rangle_H}{\Omega^2 \langle V_2 \rangle_H}. \quad (6)$$

The average over the potential energy is taken with respect to either the  $H_2$ ,  $H_4$  or the  $H_F$  Hamiltonian. It is remarkable that the factor  $\alpha$  does not depend on the wave number  $k$ . The above relation is valid for systems at equilibrium. In our simulations only a small temperature bias is applied; thus we pose the ansatz that Eq. (5) still holds, and that the equipartition relation,  $T = \alpha \Omega^2 \omega_k^2 \langle q_k^2 \rangle_T$ , is valid close to equilibrium, where averages are taken with respect to the mean temperature  $T = (T_L + T_R)/2$ . Here  $q_k$ 's are the normal modes of a 1D harmonic chain. Our numerical results, validating Eq. (8) below, justify this conjecture [23]. Calculating the thermal averages in Eq. (6), in the high-temperature regime the dynamics of the FPU model follows the behavior of the quartic model, while at low  $T$  it coincides with the harmonic dynamics,

$$\alpha \sim \begin{cases} \frac{\beta}{\Omega^2} T^{1/2} & \text{high } T \\ 1 & \text{low } T. \end{cases} \quad (7)$$

Since for short chains boundary resistance dominates heat transfer, the conductance follows Eq. (4),  $\mathcal{K}_C \propto \alpha \Omega^2 / \gamma$ , corrected by renormalization factor (7). This leads to the following scaling law:

$$\mathcal{K}_C \propto \alpha \sim \begin{cases} T^{1/2} & \text{high } T \\ T^0 & \text{low } T. \end{cases} \quad (8)$$

This behavior is significant since a macroscopic-phenomenological theory provides a different scaling law for the FPU model and its variants: Within the Debye formula the *heat conductivity* can be written as  $\kappa = \sum_k c_k v_k \tau_k$ , where  $c_k$ ,  $v_k$ , and  $\tau_k$  are the specific heat, phonon velocity, and the phonon relaxation time, respectively. Based on this expression, the following relation can be derived [20,21],  $\kappa \propto \sqrt{\alpha} / \epsilon$ , where  $\alpha$  is the same factor as in Eq. (7), and  $\epsilon = \langle V_4 \rangle_H / \langle g_4 V_4 + g_2 V_2 \rangle_H$ . In the high-temperature limit this results in the following proportion, valid for both the quartic and the FPU models,  $\kappa \propto \sqrt{\alpha} T^{1/4}$ . Assuming that the temperature gradient is linear in our system (besides the contact drop [22]), see [24], we retrieve the bulk molecular conductance

$$\mathcal{K}_M \propto \sqrt{\alpha} \sim T^{1/4}. \quad (9)$$

We thus conclude that  $\mathcal{K}_C \propto T^\nu$  whereas  $\mathcal{K}_M \propto T^{\nu/2}$ . While in the low-temperature limit and for short chains we expect  $\mathcal{K}_C$  to dominate the overall conductance, in the opposite limit the bulk resistance dominates, and eventually the scaling  $\mathcal{K} \sim T^{1/4}$  emerges.

We present next data for a longer chain of  $N=20$  (see Fig. 2). Focusing on the quartic model (circles), it is evident that the low- $T$  and the high- $T$  power-law coefficients deviate. Specifically, at low  $T$  the quartic model follows  $\mathcal{K} \sim T^{0.41}$  (upper panel) while at high  $T$ ,  $\mathcal{K} \sim T^{0.26}$  (lower panel). The high  $T$  values are characteristic for the FPU model as well ( $\square$ ). Furthermore, we can systematically extract the power-law coefficients as a function of chain size at different tem-

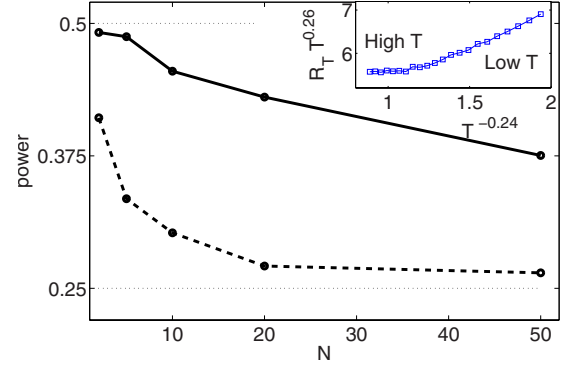


FIG. 3. (Color online) Power-law exponents as a function of chain size for high temperatures,  $T > 1$  (dashed line) and low temperatures  $T < 0.01$  (full). Data were obtained from the slope of log-log plots of conductance vs temperature for the quartic potential, parameters as in Figs. 1 and 2. Inset: the  $N=50$  data, marking the dominance of contact resistance for  $T < 0.7$ .

perature ranges, see Fig. 3. While at low  $T$  the power-law behavior is approximately fixed,  $\mathcal{K} \sim T^\nu$  with  $\nu \sim 0.4-0.5$ , at high  $T$  a clear transition to the mesoscopic result is obtained with an exponent  $\nu \sim 0.25$  for  $N > 20$  at  $T > 1$ . For convenience the slopes were estimated using the quartic-potential data.

We can further identify the critical temperature  $T_c$  where bulk conductance dominates contact effects. We conjecture that  $R_T \sim aT^{-1/2} + bT^{-1/4}$ , where the first (second) term reflects the contact (bulk) resistance, and  $a$  and  $b$  are ( $N$ -dependent) constants. A plot of  $R_T T^{1/4}$  vs  $T^{-1/4}$  should thus become flat above a critical temperature. For  $N=50$ , the inset of Fig. 3 demonstrates this turnover at the value 1.1, which translates into  $T_c \sim 0.7$ . We estimate that above this value the phononic mean-free path is significantly shorter than the molecular length. The coefficients  $a$  and  $b$  are the slope of the linear line and the asymptotic (constant) value, respectively.

The divergence of the bulk conductance from the contact conductance, given by scaling laws (8) and (9) for the  $\beta$ -FPU models, should take place in other models of confining potentials. In particular, we study the thermal conductance of the 2-6 potential,  $H = H_T + g_2 \Omega^2 V_2 + g_6 \beta V_6$ , with  $V_6 = \frac{1}{6} \sum_{i=0}^N (x_{i+1} - x_i)^6$  and  $g_6$  a Boolean variable. Here  $H_T$  is the kinetic energy and  $V_2$  is the harmonic term, same as in Eq. (2). In the high-temperature limit we obtain [17]  $\alpha \sim \langle V_6 \rangle_H / \langle V_2 \rangle_H = \frac{\sqrt{\pi}}{3\Gamma(5/6)} T^{2/3}$ ;  $\Gamma$  is the Gamma function. Short chains, controlled by the contact resistance, are expected to follow  $\mathcal{K} \propto \alpha \sim T^{2/3}$ , beyond the harmonic-anharmonic transition, while long chains should obey  $\mathcal{K} \propto \sqrt{\alpha} \sim T^{1/3}$ . Our numerical simulations have verified this behavior. Similarly, studying the  $\alpha$ -FPU model, we obtained a power-law behavior with  $\nu = 1/3$  for short chains, contrasting the mesoscopic  $\nu = 1/6$  value [21].

We study next a weakly coupled junction,  $\gamma < \Omega$ . In this parameter range (3) predicts that the conductance of both harmonic and FPU-like systems (at low  $T$ ) should be constant, independent of temperature and size. Our numerical simulations, Fig. 4, confirm this expectation, as both models yield  $\mathcal{K} \sim \gamma/2 = 0.025$ . In the high-temperature limit small deviations from this value disclose contributions to energy

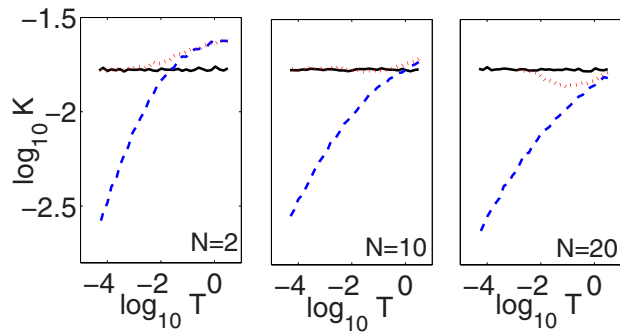


FIG. 4. (Color online) Conductance of the harmonic chain (full), quartic chain (dashed) and the  $\beta$ -FPU chain (dotted) in the weak-coupling limit,  $\Omega^2=0.01$ ,  $\gamma_L=\gamma_R=0.05$ , and  $\beta=0.01/4$ .

transfer beyond the effective-phonon theory [22]. It is also worth noting the intricate dynamics for  $N=20$ . As expected, with increasing temperature the FPU model nicely interpolates the harmonic to the quartic limit. However, quite interestingly, since the conductance of the anharmonic chain lies *below* the harmonic value, the FPU model demonstrates a decrease of  $\mathcal{K}$  with increasing  $T$  around the harmonic-anharmonic transition (see the right panel in Fig. 4).

As a purely classical treatment is adopted here, one may

question the importance of quantum effects, in particular for short chains at low  $T$ . A simple route for generalizing this study to the quantum regime would involve incorporating our simulations with quantum thermal baths, accounting for quantum statistics [25,26]. Nevertheless, quantum effects should be rudimentary here: the low-temperature dynamics of the FPU model is dominated by harmonic interactions. The interesting scaling revealed here is exposed at high temperatures, above the harmonic-anharmonic transition, see Fig. 1, where quantum effects were shown to be small [25].

In summary, we presented here results for the temperature scaling of the conductance of nonlinear chains with confining potentials. In the atomistic limit contact thermal resistance, controlling the junction conductance results in a dynamics significantly distinct from the mesoscopic limit where bulk conductance dominates. These results are significant fundamentally, demonstrating a new atomic limit with enhanced thermal conductance for the celebrated FPU model. From the practical point of view our results deliver an encouraging message for nanoscale applications, manifesting that the combination of contact effects and nonlinearity could be beneficial for thermal transport, leading to an enhanced conductivity in comparison to the macroscopic limit.

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- [23] The simulations were consistent for  $T_L=\eta T_R$ , for various  $\eta$  close to 1.
- [24] Note the distinction between the thermal conductance  $\mathcal{K}=J/\Delta T$  and the thermal conductivity  $\kappa=-J/\nabla T$ . We have numerically verified that at high enough  $T$ , the temperature gradient is constant along both FPU and quartic junctions,  $\nabla T \sim \Delta T/N$ , besides the finite jumps at the boundaries. Thus, the temperature scaling of  $\kappa$  and  $\mathcal{K}$  should be the same. This is also justified using the profile  $T(x)=T_L\{1-[1-(T_R/T_L)^{1-\gamma}]^{\frac{x}{N}}\}^{1/(1-\gamma)}$  valid for systems manifesting the power-law conductivity  $\kappa \sim T^{-\gamma}$  [22]. For  $\Delta T \ll T_L, T_R$  one can show that  $T(x) \sim T_L(1+\Delta T/T_L \frac{x}{N})$ ; thus the conductance and conductivity scale similarly with temperature.
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