Heat Conduction in Low Dimensions: From Fermi-Pasta-Ulam Chains to Single-Walled Nanotubes

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Heat conduction in 1-dimensional anharmonic systems is anomalous in the sense that the conductivity \( \kappa \) scales with a positive power of the system size, \( \kappa \sim L^\alpha \). In two dimensions, previous simulations and theoretical arguments gave a logarithmic divergence. For rectangular systems of size \( L_\parallel \times L_\perp \) there should be a cross-over from the 2-d to the 1-d behaviour as the aspect ratio \( r = L_\parallel / L_\perp \) increases from \( r = 1 \) to \( r \gg 1 \). When taking periodic boundary conditions in the transverse direction, this should be of direct relevance for the heat conduction in single-walled carbon nanotubes. In particular, one expects that \( \kappa \) nanotubes of diameter \( R \) should conduct heat better than a single nanotube of the same length and of radius \( kR \). We study this cross-over numerically by simulating the Fermi-Pasta-Ulam model. Apart from giving a precise estimate of the exponent \( \alpha \), our most intriguing results are that the divergence does not seem to be logarithmic in \( d = 2 \) but also power-like, and that the cross-over does not happen at a fixed aspect ratio. Instead, it happens at \( r = r^* \) with \( r^* \to \infty \) for \( L \to \infty \).

After years of intense studies it is now clear that heat conduction in typical anharmonic systems is anomalous in low dimensions [1]. In particular, in 1-d systems like the Fermi-Pasta-Ulam (FPU) \( \beta \)-model [2], the diatomic Toda chain [3] or in 1-d hard-particle gases with alternating masses \([3,4,5,6]\) the heat conductivity \( \kappa \), defined by \( J(x) = \kappa \nabla T(x) \), scales as

\[
\kappa \sim L^\alpha
\]  

where \( L \) is the size of the system. Notice that we have assumed here that \( T(x) \) exists, i.e. that local thermal equilibrium (LTE) is established in the limit \( L \to \infty \), which is non-trivial [7] but seems to be true for these systems.

The main reason for this anomalous behaviour is the fact that the free path length of phonons with long wave lengths \( \lambda \) diverges for \( \lambda \to \infty \). Thus soft modes propagate nearly ballistically. In higher dimensions this is also true, but due to the enlarged phase space for other modes it is restricted to such a small region that \( \kappa \) remains finite. In 1 dimension there are just not enough other modes with which soft phonons could interact to make \( \kappa \) finite. This argument requires of course that there is an acoustic phonon branch, i.e. that soft (Goldstone) modes exist because translation invariance is not broken. Thus this argument does not apply to charge conduction where the electrons move in an external potential braking translation invariance.

For 2-d systems, simulations of FPU lattices of rather modest sizes [8] indicated a logarithmic divergence \( \kappa \sim \ln L \). This was also supported by mode coupling theory. Indeed, a logarithmic divergence should not be too surprising in view of the logarithmic divergence of transport coefficients in 2-d hydrodynamics [9] due to long time tails. In [8] it was also observed that the cross-over from the 1-d to the 2-d scaling in rectangular systems happens at surprisingly large aspect ratios. Consider a system of size \( L_\parallel \times L_\perp \) where the length \( L_\parallel \) is parallel to the heat flow. The aspect ratio is defined as \( r = L_\parallel / L_\perp \). For the sizes studied in [8], the cross-over happened at \( r \approx 10 \), but no detailed study was made.

The main aim of the present paper is to present detailed simulations of the FPU \( \beta \)-model on much larger lattices than in [8], in order to study in detail the asymptotic behaviour of 2-d systems and of the cross-over. But before doing so, let us point out that this study is not entirely academic. It is of immediate experimental and maybe even of technological importance.

The lattices which we shall use have periodic boundary conditions in the transverse direction. Therefore, we actually study the heat conduction through (single-walled) tubes. By far the most important nanotubes are those made of carbon [10]. Their heat conduction is dominated by phonons. They can be made with very few lattice defects, such that their conductivity is basically controlled by phonon-phonon interactions as in the FPU model. Indeed, this conductivity has been both calculated [11] and measured [12], and is found to be very large. It was thus suggested that carbon nanotubes should be the ideal material to carry away Joule heat in the next generation of integrated circuits which will use nanoscale structures and for which cooling will be a major problem. Since most molecular dynamics simulations and measurements were done for fixed tube lengths, no length dependence of \( \kappa \) was seen in them. The only exception is Ref. [13] where a clear length dependence of \( \kappa \) was seen for the narrowest tubes, but not for wider ones.

In all simulations of carbon nanotubes the realistic Tersoff-Brenner potential [14] was used. If, as we claim, the size dependence of \( \kappa \) is universal, its main features (and in particular the exponent \( \alpha \) in Eq. \((1)\)) should be independent of the potential. There will be of course unknown scale factors when transferring our results from FPU systems to carbon tubes, nevertheless we should be able to use our results directly for an experimentally accessible system.

In our FPU simulations we used a square lattice with sites indexed by integer vectors \( \mathbf{n} = (i,j) \) with \( 1 \leq i \leq L_\parallel \) and \( 1 \leq j \leq L_\perp \). For the
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FIG. 1: Temperature profiles for lattices with $N_x = 128$ and $N_y = 1, 3, \text{and } 32$. For these simulations we used a smaller step size $dt = 0.025$.

$N_x$ and $1 \leq j \leq N_y$. We use periodic b.c. in $j$. The lattice constant, the particle mass, and the $\beta$ parameter are all set to unity. Thus $N_x = L_y$ and $N_y = L_x$ and, apart from the heat baths at $i = 1$ and $i = N_x$, the hamiltonian is

$$H = \frac{1}{2} \sum_n p_n^2 + \sum_{n,m > 1} \left[ \frac{1}{2}(q_n - q_m)^2 + \frac{1}{4}(q_n - q_m)^4 \right].$$

Notice that identically the same form (with $N_y = 1$) can be used also for 1-d chains. To measure a heat flux we keep the left boundary ($i = 1$) at a lower temperature $T_h = 10$, and the right boundary ($i = N_x$) at $T_c = 6$. These are fairly large in order to obtain fast equilibration (similar temperatures were used in [3]). These thermostats are implemented as Nosé-Hoover heat baths with response times $\Theta = 1$. Thus the equations of motion for the particles in the boundaries are modified to

$$\dot{q}_n = -\frac{\partial H}{\partial q_n} - \xi_n p_n \quad \text{with} \quad \dot{\xi}_n = \frac{p_n^2}{T} - 1. \quad (3)$$

Since our overall accuracy was mainly limited by statistical fluctuations and not by integration errors, we used mainly a simple leap-frog integrator [5] with a large step size $dt = 0.05$. Test runs with smaller $dt$ were made to verify the results (see below). Total integration times were typically between $2 \times 10^6$ and $2 \times 10^7$ units.

Although there exist more sophisticated symplectic integrators for Nosé-Hoover baths [13], and although the standard form given in Eq. [8] is not symplectic, the straightforward leap-frog was also used for them. Apart from simplicity and robustness, our main reason was that LTE should be strongly violated at the boundaries anyhow, due to the large temperature gradients [8] at the boundaries (see also Fig.1). If LTE would hold everywhere, and $\kappa$ would depend only on $T$ and on the system size, the constancy of the heat flow throughout the sample would imply a nearly linear temperature profile (since $\kappa$ depends smoothly on $T$). This is obviously not the case. A more direct check of LTE is obtained by estimating the normalized kurtosis (fourth cumulant) of the momentum distribution,

$$Q_i = \frac{N_y \langle \sum_{j=1}^{N_y} p_{i,j}^4 \rangle}{\langle \sum_{j=1}^{N_y} p_{i,j}^2 \rangle^2} - 3. \quad (4)$$

In Fig.2 we show these cumulants for length $N_x = 128$ and different widths. All three curves show that indeed $|Q_i| \ll 1$, verifying approximate LTE (for larger $N_x$ we find even smaller $Q_i$, as we should expect from the fact that temperature gradients decrease with $N_x$). The detailed curves depend strongly on $N_y$, but in all cases the kurtosis is largest near the boundaries. Thus data obtained from the boundary regions should not be used for the analysis anyhow, and integration errors in these regions should not matter too much.

Like the kurtosis, local temperatures and heat fluxes were indeed not calculated as functions of the spatial positions $(x, y)$ but for fixed particles: The temperature of the $i$-th layer is simply $T_i = N_y^{-1} \langle \sum_{j=1}^{N_y} p_{i,j}^2 \rangle$, while the average heat flux through any horizontal bond connecting the layers $i$ and $i + 1$ is

$$J_i = (2N_y)^{-1} \left\{ \sum_{j=1}^{N_y} (\dot{q}_{i,j} + \dot{q}_{(i+1,j)}) F_{(i,j)} \right\}, \quad (5)$$

where $F_{(i,j)}$ is the force acting between particles $(i, j)$ and $(i + 1, j)$. We checked that indeed $J_i$ is independent of $i$, within the statistical errors.

Conductivities obtained by dividing the flux by the globally averaged temperature gradient,

$$\kappa_{\text{global}} = \frac{\sum_{i=1}^{N_x} J_i}{(T_h - T_c)}, \quad (6)$$

are shown in Fig. 3, while conductivities obtained by using the gradient averaged only over the central half of the
FIG. 3: Conductivities defined by Eq.(6) for systems with fixed lengths $N_x = 32, 64, 128, \ldots 8192$ against $N_y$ (for $N_x = 8192$ there is one single point at $N_y = 1$). Statistical and integration errors are less than 2 percent.

FIG. 4: Conductivities defined by Eq.(7) from the same runs as the data in Fig.3.

FIG. 5: Same data as in Fig. 4, but plotted against $N_x$ and with data for the same $N_y$ connected by lines.

FIG. 6: Conductivities defined by Eq.(7) for fixed aspect ratios.

lattice,

$$
\kappa_{\text{center}} = \sum_{i=1}^{N_x} \frac{J_i}{N_x(dT/dx)_{\text{center}}} \tag{7}
$$

with $(dT/dx)_{\text{center}} = 2(T_{3N_y/4} - T_{N_y/4})/N_x$, are shown in Figs. 4 and 5. There are small but significant differences between the two definitions of $\kappa$. In general, the plots for $\kappa_{\text{center}}$ show slightly more structures. We argue that they are more relevant, since they are less affected by boundary effects. We show the data for $\kappa_{\text{global}}$ also, because they are much less noisy.

From Figs. 3 and 4 we see that $\kappa$ saturates for large $N_y$. The saturation values increase roughly power-like with $N_x$. For intermediate $N_y$ there are very shallow minima, at $(N_y)_{\text{min}} \sim N_x^{0.35}$. A rough fit to the data for $N_y > 2$ is obtained with a scaling ansatz

$$
\kappa = N_x^{\alpha'} \phi(N_y/N_x^\beta), \quad \alpha' = 0.26, \beta = 0.35. \tag{8}
$$

with $\phi(x) \approx \text{const}$ for $x \gg 1$ and $\phi(x) \sim x^{(\alpha'-\alpha)/\beta}$ for $x \to 0$. Thus the cross-over happens at $r^* \sim N_x^{1-\beta} = N_x^{0.65}$. As seen from Fig. 5, both the 1-d behaviour ($r = \infty$) and the behaviour at $r \ll r^*$ are roughly power like, but with different powers. In between, $\kappa$ increases even faster with $N_x$ (for fixed $N_y$).

For fixed aspect ratios this leads to a decrease of $\kappa$ with $N_x$ for small $N_x$ ($r > r^*(N_x)$), and to a power increase for large $N_x$ (see Fig. 6). Indeed one sees definite deviations from a pure power law, even for large $N_x$. This means that $\kappa$ increases asymptotically less fast than suggested by a simple least square fit to the present data (which would give the exponent $\alpha' = 0.26$ used in Eq.(8)). Thus the true asymptotic exponent $\alpha(2d)$ for 2-d lattices is less than $\alpha'$. Any detailed extrapolation is of course uncertain, but our best estimate is $\alpha(2d) = 0.22 \pm 0.03$.

In order to see more precisely the behaviour of 1-d chains (where we have much higher statistics than for $N_y > 1$), and to see more clearly the errors made by the finite step size $dt$, we show $\kappa/N_x^{0.37}$ in Fig. 7. Indeed the
uncertainty in the definition of \( \kappa \) is much more important than the integration error. Although the latter is much larger than the statistical error (since we obtained very high statistics for \( N_y = 1 \)), it has hardly any effect on the scaling of \( \kappa \). While the curves for \( \kappa_{\text{global}} \) seem to flatten for large \( N_x \), those for \( \kappa_{\text{center}} \) seem to turn upward again for very large \( N_x \), after having bent downward for intermediate \( N_x \). Essentially the same behaviour was seen for the 1-d gas with alternating masses in [5]. Again any extrapolation is rather uncertain, our best estimate is \( \alpha(1d) = 0.37 \pm 0.01 \).

In summary, we have studied the heat conduction on rectangular 2-d FPU lattices with periodic lateral boundary conditions, including lattices with zero width (i.e. linear chains). We argued that the divergence of the conductivity with system size should be universal, so that our results should apply also, among others, to carbon nanotubes. One striking result is that we do not see the logarithmic increase of \( \kappa \) found in [3]. At least for system sizes up to five times the size of those studied in [3], the increase is power-like, with an exponent larger than 0.2. This increase flattens somewhat for even larger lattices, but our best asymptotic estimate is still around 0.2. For \( d = 1 \) our estimate of the exponent \( \alpha \) is slightly smaller than previous ones, but compatible with them.

Our other striking result is that the cross-over from 1-d to 2-d behaviour happens at an aspect ratio which diverges with system size. Thus it might be difficult to see the true 1-d behaviour in carbon nanotubes. But this does not mean that \( \kappa \) should not increase with their length, quite to the contrary: The increase at presently achievable tube lengths might even be faster than both the asymptotic 1-d and 2-d power laws, in view of the special shape of the cross-over. Since \( \kappa \) is decreasing with the width \( L_y \) for large aspect ratios, two tubes with small radius conduct better than one tube with twice the radius. Thus, if one wants to maximize the heat flux for a given amount of carbon and a given length of the tubes, one should make the tubes as narrow as possible.

Finally we would like to point out that the effect we discussed in this paper is closely related to the Casimir force. The latter deals with the change in energy of a cavity due to elimination of vacuum modes by the cavity walls. The enhanced conductivity of narrow strips compared to the bulk is due to the elimination of transverse phonons (i.e. phonons with \( p_y \neq 0 \)), the interactions with which otherwise would limit the free path length of soft longitudinal phonons which contribute to the energy transport.

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