On the Violations of Local Equilibrium and Linear Response

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(October 28, 2018)

We study how local equilibrium, and linear response predictions of transport coefficients are violated as systems move far from equilibrium. This is done by studying heat flow in classical lattice models with and without bulk transport behavior, in 1–3 dimensions. We see that linear response and local equilibrium assumptions break down at the same rate. The equation of state is also found to develop non-local corrections in the steady state. We quantify the breakdown through the analysis of both microscopic and macroscopic observables, which are found to display non-trivial size dependence.

In studies of non-equilibrium systems, local equilibrium is an assumption which is essential to allowing the use of statistical mechanics and equilibrium or non-equilibrium thermodynamics \cite{2}. Without local equilibrium, even the definition of temperature is not unique \cite{3}, and it becomes unclear how to define simple transport processes. Local equilibrium is typically justified through conditions argued to be necessary or sufficient. This might involve checking for Onsager reciprocity \cite{3}, enforcing upper limits on local fluctuations of temperature (density,...) \cite{4}, verifying that the equation of state holds locally \cite{4}, and so forth \cite{4}. However, these conditions and inequalities do not offer any quantitative guidance into how local equilibrium breaks down. Further, linear response has not been tested in conjunction with the breakdown of local equilibrium, which is an important consideration if one questions whether or not higher order corrections to Fourier’s Law are consistent. Previous studies have observed the breaking of local equilibrium \cite{1,8} as well as deviations from linear response \cite{10,11}. However few such cases are known, and the quantitative behavior of physical observables when local equilibrium is broken has not been studied previously. In \cite{8}, the breakdown of local equilibrium was observed in the X–Y model and the Lorentz gas under thermal gradients, which was attributed to the infinite number of local conservation laws in the dynamics. Our results for non–integrable models will show that the integrability of the systems are not necessary for the deviations from local equilibrium to occur. In this letter we establish a quantitative guide to the rate at which concepts like local equilibrium and linear response become violated in systems which are subject to thermal gradients. We study lattice models in $d = 1 – 3$ spatial dimensions, including the Fermi-Pasta-Ulam $\beta$ model, which does not have a bulk transport limit in $d = 1$. We also test how the equation of state is modified and its relation to expectations from irreversible thermodynamics.

How should physical observables behave away from local equilibrium? A natural idea is that a physical observable $A$ will deviate from its value in local equilibrium as we move further away from equilibrium. In our case, a temperature gradient $\nabla T$ provides a natural measure of how far we are from equilibrium. Since the intrinsic physical properties should not depend on which side of the box is at a higher temperature, the deviation $\delta A$ from its local equilibrium value is expected to behave as

\begin{equation}
\delta A = C_A \left( \frac{\nabla T}{T} \right)^2 + C_A' \left( \frac{\nabla T}{T} \right)^4 + \ldots
\end{equation}

While seemingly natural, such an analytic expansion is not trivial; in sheared fluids, transport coefficients have been seen to display non-analytic dependences on the shear rate, which have not been entirely clarified \cite{11}. The coefficients $C_A, C_A', \ldots$ are in principle dependent on $T$, and $L$, the size of the lattice in the direction of the gradient. If the relation is completely local, we expect that they will be independent of $L$. We shall find that the situation is more subtle.

We study two systems which display qualitatively different transport behavior. Their Hamiltonians are

\begin{equation}
H = \frac{1}{2} \sum_{\mathbf{r}} \left[ p^2 + (\nabla \phi)^2 + V \right].
\end{equation}

where $V = \beta(\nabla \phi)^4 / 2$ for the FPU–$\beta$ model and $V = \phi^4 / 2$ for the $\phi^4$ model. The $\phi^4$ model has a well defined bulk limit for the thermal conductivity in $d = 1 – 3$ \cite{10} whereas the FPU model has $L$ dependent thermal conductivity in $d = 1$ \cite{12,13}. We will use $\beta = 1$ without loss of generality. Here $\mathbf{r}$ runs over all sites in the lattice $(x = 1, ..., L, y, z = 1, ..., N_L)$, and the lattice derivative has components $\nabla_k \phi \equiv \phi_{\mathbf{r} + e_k} - \phi_{\mathbf{r}}$ ($e_k$ is the unit lattice vector in the $k$-th direction). For $d = 2, 3$ we take $N_L = 3 – 20$ sites in the transverse directions with periodic boundary conditions. We thermostat the endpoints $L = 0, N + 1$ dynamically at temperatures $T_1$ and $T_2$, as discussed in \cite{13,10}.
Once we determine $C_A$ for an observable $A$, it is possible to compute its non-equilibrium spatial distribution function. When $\kappa(T)$ behaves as a power law in the temperature range of interest, denoted $\kappa = cT^{-\gamma}$, the temperature profile is $T(x) = T_1(1 - (T_2/T_1)^{1-\gamma})(x/L)^{1/(1-\gamma)}$ [10]. Such a power law behavior for $\kappa(T)$ has been shown to hold for the $\phi^4$ model in $d = 1 - 3$ and also in most temperature regions in the $d = 1$ FPU $\beta$ model, including the region we work with here [13]. The agreement for the predicted profile is shown in Fig. 1(a). Using Fourier’s law $J = -\kappa \nabla T$, Eq. (1) and $T(x)$, we derive to leading order:

$$\frac{\delta A}{A} = C_A \left( \frac{J}{\kappa T} \right)^2 = C_A \left( \frac{1}{a + bx} \right)^2$$

(3)

where $a = T_1^{1-\gamma}c/J$, $b = \gamma - 1$, and $J$ is the heat flow. So knowing $C_A$, we can also predict the spatial variation of the non-equilibrium observable $A$. This brings to light an interesting relation to coarse graining. Coarse graining in $x$ over regions of length $\ell$, with $L > \ell > \lambda$, where $\lambda$ is the mean free path of the excitations, will provide no significant improvement towards recovering local equilibrium, since (up to the sign of $C_A$), the functional form of (3) is positive definite. (This will be evident in Fig. 1(b).)

One might also be inclined to expand $\phi^4$ in powers of $(\nabla^a T)/T$. However, in the region where the temperature dependence of the thermal conductivity can be described by a power law, $\kappa = cT^{-\gamma}$, one can show, using Fourier’s law, that $\nabla^a T = a_n \times (\nabla^a T)^n$, where $a_n$ is a temperature independent constant. Strictly speaking, Fourier’s law holds only close to local equilibrium, but as we shall see later, the deviations from it is of order $(\nabla^a T)^2$ so that the difference is a higher order correction in the expansion in $(\nabla^a T)$. Local Equilibrium: With the local temperature given by $T_k = \langle p_k^2 \rangle$, a natural measure for the deviations from local equilibrium is the deviation of the momentum distribution from the Maxwellian distribution. The cumulants $\langle \langle p_k^2 \rangle \rangle = \langle \langle p_k^2 \rangle \rangle - 3\langle \langle p_k^2 \rangle \rangle^2$, $\langle \langle p_k^2 \rangle \rangle = \langle \langle p_k^2 \rangle \rangle - 15\langle \langle p_k^2 \rangle \rangle^2 + 30\langle \langle p_k^2 \rangle \rangle^3$, and so on, normalized by the local temperature, $\langle \langle p^n \rangle \rangle/T_n^{n/2}$, provide a quantitative measure on how far we are from local equilibrium. In local equilibrium, $\langle \langle p^n \rangle \rangle = 0$ $(n > 2)$. Consider first how systems typically behave under thermal gradients. In Fig. 1, the local temperature and $\langle \langle p^2 \rangle \rangle/T^2$ are plotted against the position in both $\phi^4$ and FPU systems. When the system is not too far from equilibrium, the temperature profile can be understood by locally applying Fourier’s law $J = -\kappa(T) \nabla T$, where the heat flow $J$ is constant [10,14,15]. Since $\kappa$ depends on $T$, $\nabla T$ will also depend on $T$ and hence the temperature profile becomes curved for increasing boundary temperature differences. We note that the temperature profile for the $\phi^4$ theory is visibly more curved under the same boundary conditions, even though the FPU is further from local equilibrium (Fig. 1(b)).

In Fig. 1(b), we see that the $\langle \langle p^4 \rangle \rangle$ cumulants are non-zero inside the system, so that we are no longer in local equilibrium. Contrary to naive intuition, the steepest gradient does not lead to the system being furthest from local equilibrium. In fact, the converse is true — the system is furthest from equilibrium in the flattest region.

In Fig. 2 we plot the deviations of the 4-th cumulant of the momentum for the $\phi^4$ theory and FPU model against $\nabla T/T$, which corresponds to taking $A = \langle p^4 \rangle$ in (4). We are interested in the physics away from the boundaries and we shall always measure the physical quantities locally, well inside the system, although boundary effects can be readily understood [13]. We find that $\nabla T/T$ provides a good measure of how far we are from equilibrium and in both models, the cumulants behave as
\[ \delta_{LE} = \frac{\langle \langle p^4 \rangle \rangle}{3T^2} = C_{LE} \left( \frac{\nabla T}{T} \right)^2, \]

where \( C^{\phi}_L E = 1.1(8)L^{0.9(2)} \) \((T=1)\) and \( C_{L E}^{FPU} = 4.3(4)L^{0.99(2)} \) \((T=8.8)\). Similar investigations at different \( T \) yields a weak \( T \) dependence for \( C_{LE} \) which is difficult to establish. These results are consistent with \( d > 1 \) in both models at the same temperatures. Using \( C^{\phi}_L E, C_{L E}^{FPU} \), we can predict the shape of \( \langle \langle p^4 \rangle \rangle/T^2 \): In Fig. 1(b), the non-equilibrium distribution \((\square)\) symbols is compared to simulation results (lines) agreeing nicely.

Here, a relatively simple picture emerges: as we move away from equilibrium by increasing the difference in the boundary temperatures, each point in the interior deviates from local equilibrium in a predictable manner, without any threshold. Away from equilibrium, local equilibrium is an approximation that is quite good for small gradients since the deviations from it only vary as \((\nabla T)^2\). Similar results hold for higher momentum cumulants.

The \( L \) dependence of \( C_A \) is quite intriguing. Naive argument suggests that since the gradients and the cumulants are local, the relation between them would not depend on \( L \), at least in the \( \phi^4 \) theory where there is a bulk limit. This turns out not to be the case. In principle, it is possible that the effect we see will disappear in the large \( L \), bulk limit. However, this seems implausible since we have excluded the region within the mean free path from the boundaries in the above results, using the properties of the model extracted in \([10,13]\).

**Linear Response:** Let us now investigate the validity of linear response theory. This has been discussed previously as one of the criteria for the breakdown of local equilibrium \([9]\), even though no deviation was seen there. A priori, it is not clear if the linear response law can be used as a criterion for the breakdown of local equilibrium, rather than as an indication of higher order equilibrium corrections \([16]\). Such an analysis assumes that even when linear response is broken, local equilibrium holds sufficiently well so that one can unambiguously define the temperature inside the system (or that we adopt a particular definition for \( T \)). The linear response prediction of the heat flow \( J \) is obtained by computing \( \kappa \) from applying Fourier’s law locally. This is denoted as \( J_{LR} \), and agrees with direct measurements of \( J \) in the near equilibrium limit \([10]\).

**FIG. 2.** \( \langle \langle p^4 \rangle \rangle/T^2 \) away from equilibrium for the \( \phi^4 \) theory \((d = 1, 2, 3, T = 1)\) and the FPU model \((d = 1, 2)\) for various lattice sizes on an arbitrary scale. The fits of the data to a behavior const. \( \times (\nabla T/T)^2 \) are plotted for each lattice size.
FIG. 3. The deviations from the linear response law as a function of $\nabla T/T$ for the FPU model for $T = 8.8$ (left) and the $\phi^4$ theory for $T = 1$ (right) for $d = 1$. The dashes represent linear response and the solid line shows the quadratic deviations from it of the form $\kappa\nabla T \left[ 1 + C_{LR}(\nabla T/T)^2 \right]$.

When the temperature gradient is small, linear response theory is applicable so that Fourier’s law is satisfied globally: $J_0 = -\kappa(T)(T_2 - T_1)/L$. As the gradient increases, curvature develops. When $\kappa \sim T^{-\gamma}$, we can integrate Fourier’s law to obtain the next leading order correction due to curvature in $T(x)$:

$$\frac{J_{LR} - J_0}{J_0} = \frac{\gamma(\gamma + 1)}{24} L^2 \left( \frac{\nabla T}{T} \right)^2 + \cdots. \quad (5)$$

This $L^2$ dependence simply indicates that Fourier’s law is satisfied locally rather than globally. As the gradient increases even further, the energy that can be pumped through the system becomes less than that predicted by linear response locally, which carries an error in itself. The relative deviation from the linear response result (as shown in Fig. 3) can be reasonably well explained by

$$\delta_{LR} = \frac{J - J_{LR}}{J_{LR}} = C_{LR} \left( \frac{\nabla T}{T} \right)^2, \quad (6)$$

where $C_{LR}^{\phi} = -4(3)L^{1,0}(2) (T = 1)$ and $C_{LR}^{FPU} = -6.6(8)L^{0,9}(1) (T = 8.8)$. For large gradients, Eq. (6) will naively give rise to decreasing current with increasing gradient. However, when the gradient is this large, the higher order terms in $(\nabla T/T)$ becomes as important. We do not know how the system behaves under such an extremely large gradient, but it would be natural to expect that the current will saturate. The quadratic behavior in (6) is also seen for both the $\phi^4$ theory and for the FPU model in $d = 1$ at other temperatures and also for the models in $d > 1$, even though the extraction of $C_{LR}$ involves larger errors in those cases.

Within error, we see that the violation of linear response and local equilibrium are closely connected, occurring in the same manner:

$$\delta_{LE} \sim \delta_{LR}. \quad (7)$$

Local equilibrium and linear response have no threshold, and break down at the same rate.

Equation of State: The equation of state for these models are simple in equilibrium since there is only one independent variable, which can be taken to be $T$. (In the FPU model in $d = 1$, there is also a possibly weak dependence on $L$ in $P_{eq}$; unlike the $L$ dependence of $\kappa$, it is far less discernible.) We denote it $P(T)$ (or $E(T)$) where $P$ ($E$) is the pressure (energy density). We can measure them through the stress tensor, $P = P_{xx} = T^{11}$, $E = T^{00}$ [14]. In Fig. 4, where we plot the relative deviation of the pressure from its equilibrium value, $(P - P_{eq})/P_{eq}$, against $\nabla T/T$. We see that the equation of state develops new dependences of the form

$$P(T, \nabla T, L) = P_{eq}(T) \left[ 1 + C_P \left( \frac{\nabla T}{T} \right)^2 \right] \quad (8)$$

where $C_P^{\phi} = 1.5(1.2)L^{0,9}(2) (T = 1)$, $C_P^{FPU} = 4.1(6)L^{0.30(4)} (T = 8.8)$. The non-equilibrium equation of state, $P(T, \nabla T, L)$ develops a non-trivial size dependence in $C_P$, rendering it non-local. Similar analysis for energy density yields the coefficients $C_E^{\phi} = 0.5(3)L^{0,9}(2) (T = 1)$, $C_E^{FPU} = 1.7(7)L^{0.3}(1) (T = 8.8)$. The quadratic behavior as in (6) is seen also in both models at different $T$ for $d = 1 - 3$ but we do not have enough statistics to unambiguously extract $C_{P,E}$ in those cases. While Extended Irreversible Thermodynamics (EIT) predicts the quadratic dependence in [8] for particle gases and liquids, precise identification of the non-equilibrium definitions of the physical quantities between the two theories is necessary before quantitative comparisons can be made. EIT predicts a local behavior for $\delta P$ in contrast to our observations.
We have quantified the violations of local equilibrium and linear response in $\phi^4$ theory and the FPU $\beta$ model in $d = 1 - 3$ dimensions, and observed that they break down at the same rate, with similar order of magnitude. Both are found to vary with the thermal gradient as $(\nabla T/T)^2$, and there is no threshold for the violations, appearing immediately as one moves away from global equilibrium. Other physical quantities such as the pressure and the energy density were also found to behave in a similar manner. We found that using the coefficients $C_A$, we can predict the spatial dependence of non-equilibrium distributions of observables. As a consequence, coarse graining does not modify our conclusions.

Since the definition of $T$ is no longer unique when local equilibrium is broken, a question arises as to how the choice of non-equilibrium definition for $T$ affects the results. Expressions for non-equilibrium deviations of $J, P, E$ as in Eqs. (6), (8) will in general be affected covariantly; in particular, for a generic redefinition $T = T' + \nu (\nabla T/T)^2$, $C'_A = C_A + \nu (dA/dT)$. The local equilibrium violations seen in $\langle p^n \rangle / (pn)^{1/2}$ as in Eq. (4) are invariant under such redefinitions, up to the order we consider. The physics, of course, is invariant under any redefinition in temperature. We find that the momentum cumulants provide the most natural and also the most clear criterion for the violation of local equilibrium.

Certainly more questions remain: Importantly, we do not have an analytic understanding of how the coefficients $C_{LE}, C_{LR}, C_P, C_E$ are related to the parameters of the theories, including their $L$ dependence. Another question to consider is how other physical quantities behave away from local equilibrium. It would be interesting to further explore the consequences of non-locality in the equation of state, as well as use such models as a testing ground for non-equilibrium thermodynamics, since there is good control on the non-equilibrium steady state.

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\[ \text{FIG. 4. Pressure } P \text{ as a function of } \nabla T/T, \text{ away from equilibrium for } \phi^4 \text{ (top, } T = 1) \text{ and FPU (bottom, } T = 8.8). \text{ The fits of the data to a behavior const.} \times (\nabla T/T)^2 \text{ are plotted for each lattice size. } P \text{ is seen to increase away equilibrium in both models. (Vertical axis scaled as indicated.)} \]

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