Anomalous Heat Diffusion

Peter Hänggi

1. V. Zarbudaev, S. Denisov and P. Hänggi, *Perturbation spreading in many-particle systems: A random walk approach*, **PRL 106, 180061(2011)**; *ibid,* **PRL 109, 069903(2012)**.

2. S. Liu, P. Hänggi, N. Li, J. Ren, and B. Li, *Anomalous heat diffusion*, **PRL 112, 040601 (2014).**

3. S. Liu, J. Liu, P. Hänggi, C. Wu, and B. Li, *Triggering waves in Nonlinear Lattices: Quest for Anharmonic Phonons and Corresponding Mean Free Paths,* **PRB 90, 174304 (2014).**

Collaborators

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PRL 112, 040601 (2014)

Fourier's law

Fourier's law of heat conduction

$$J = -\kappa \nabla T(x)$$

κ – thermal conductivity of the material.

Using Fourier's law and the energy conservation equation

$$\frac{\partial \epsilon}{\partial t} + \nabla J = 0$$

gives the heat DIFFUSION equation:

$$\frac{\partial T}{\partial t} = \frac{\kappa}{c} \nabla^2 T$$

Thus Fourier's law implies diffusive heat transfer.

Theories of heat transport

- Kinetic theory
- Peierl's-Boltzmann theory
- Green-Kubo linear response theory
- Landauer theory – This is an open systems approach especially useful for mesoscopic systems.
    - Nonequilibrium Green's function formalism
    - Langevin equations and Green's function formalism

Proving Fourier's law

Proving Fourier's law from first principles (Newton's equations of motion) is a difficult open problem in theoretical physics.

Review article:
Fourier's law: A challenge for theorists
(Bonetto, Rey-Bellet, Lebowitz) (2000).

It seems there is no problem in modern physics for which there are on record as many false starts, and as many theories which overlook some essential feature , as in the problem of the thermal conductivity of nonconducting crystals.
R. Peierls (1961)

Results so far

All the standard theories of transport involve uncontrolled approximations and do not provide a “proof” of Fourier’s law.

Direct studies (simulations and exact results) in one and two dimensional systems find that Fourier’s law is in fact not valid. The thermal conductivity is not an intrinsic material property.

For anharmonic systems without disorder , κ diverges with system size L as:

$$\begin{aligned} \kappa &\sim L^{\alpha} \quad 1D \\ &\sim L^{\alpha'}, \quad \log L \qquad 2D \\ &\sim L^{0} \quad 3D \end{aligned}$$

A.D, Advances in Physics, vol. 57 (2008).

Lack of Fourier Law in Low Dimension

- Harmonic model: $H = \sum_{l=1}^{N} \frac{p_l^2}{2m} + \sum_{l=1}^{N-1} \frac{1}{2} k(x_l - x_{l+1})^2$

  RLL model (1D Langevin baths at the ends)

  $$J = \frac{k k_B}{2\gamma} \left[1 + \frac{\nu}{2} - \frac{\nu}{2}\sqrt{1 + \frac{4}{\nu}} \right] (T_1 - T_N)$$

  $$\kappa = \frac{j}{\Delta T/N} \propto N$$

  Not intensive !!!

❑ Rieder, Lebowitz, Lieb, ***J. Math. Phys.*** 8, 1073 (1967)

$$F_{1,N} = \eta_{1,N} - \gamma v_{1,N}$$

$$\langle \eta_{1,N}(t) \eta_{1,N}(0) \rangle = 2\gamma k_B T_{1,N} \delta(t)$$

$$\nu = \frac{mk}{\gamma^2}$$

$\alpha = 1$	***Ballistic***

- With disorder: (1D)

  $$\kappa = \frac{j}{\Delta T/N} \sim N^{\alpha}$$

  free boundaries: $\alpha = 1/2$

  fixed boundaries: $\alpha = -1/2$

❑ Dhar, ***Phys. Rev. Lett.*** 86, 5882 (2001)

❑ Roy and Dhar, ***Phys. Rev. E*** 78, 051112 (2008)

$0 < \alpha < 1$	***Superdiffusive***
$\alpha < 0$	***Subdiffusive***

- With anharmonicity:

  $$\kappa = \frac{j}{\Delta T/N} \sim N^{\alpha}$$ **for 1D systems**

  $$\kappa = \frac{j}{\Delta T/N} \sim \log N$$ **for 2D systems**

❑ Lepri, ***Phys. Rev. Lett.*** 78, 1896 (1997)

❑ Yang *et al*, ***Phys. Rev. E*** 74, 062101 (2006)

Normal Heat Transport

Fourier Law

$$\boldsymbol{j} = -\kappa \boldsymbol{\nabla} T$$

$\boldsymbol{j}$: heat flux through an unit surface per unit time

$\boldsymbol{\nabla} T$: gradient of temperature

κ : thermal conductivity

an intensive property

Heat **(Diffusion)** equation

$$\frac{\partial \epsilon}{\partial t} = \frac{\kappa}{c} \Delta\epsilon = D_E \Delta\epsilon$$

ϵ : local energy density

c : volumetric specific heat

D_E: thermal diffusivity

$$\frac{\partial \epsilon}{\partial t} + \nabla \cdot \boldsymbol{j} = 0 \qquad \epsilon = cT$$

Normal Diffusion

Heat equation $\quad \dfrac{\partial \epsilon}{\partial t} = D_E\, \Delta\epsilon\ ;\ \ x \in \mathbf{R};\ t \in (0, \infty)$

Initial condition:

$$\epsilon(x,0) = \delta(x)$$

Solution (Green function):

$$\Phi(x,t) = \frac{1}{\sqrt{4\pi D_E t}} \exp\left(-\frac{x^2}{4D_E t}\right)$$

General initial condition:

$$\epsilon(x,0) = \eta(x)$$

($\eta(x)$, not necessarily positive)

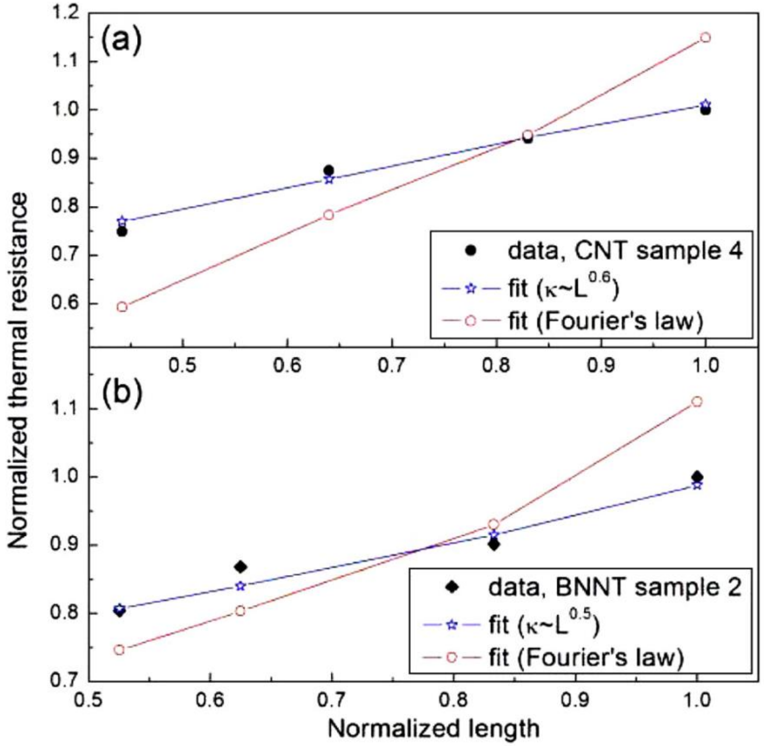
Solution:

$$\epsilon(x,t) = \int \Phi(x - x', t)\eta(x')dx'$$

$$\langle x^2(t)\rangle = \frac{\int x^2 \epsilon(x,t)dx}{\int \epsilon(x,t)dx} = 2Dt + \frac{\int x^2 \eta(x)dx}{\int \eta(x)dx}$$

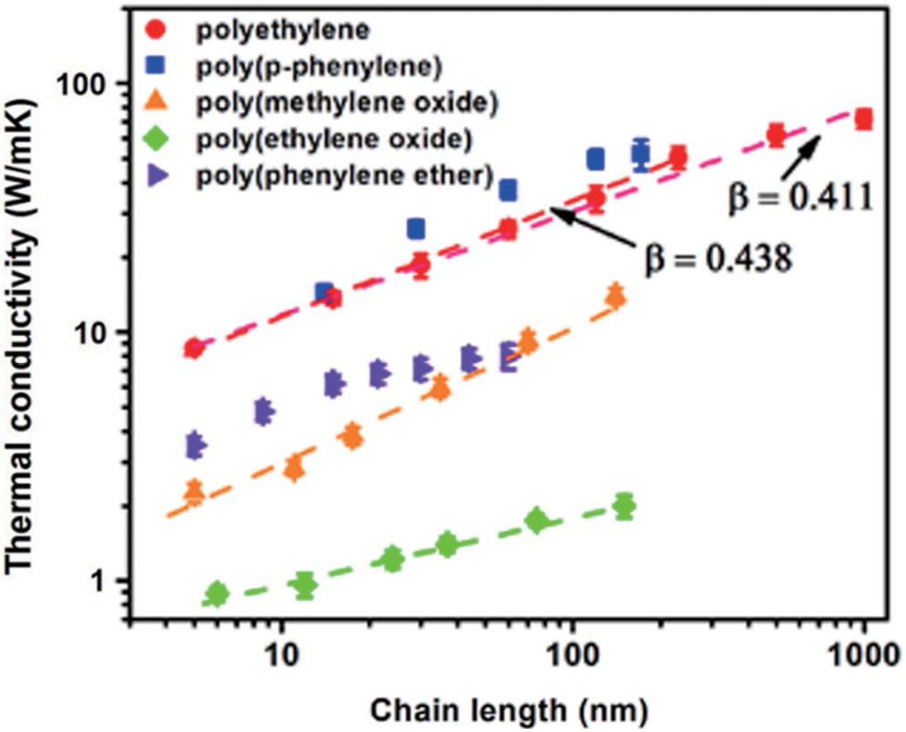
Low Dimensional Materials

- Chang *et al*, ***Phys. Rev. Lett.*** 101, 075903 (2008)



Experiment: Normalized thermal resistance vs. normalized sample length for CNT and BNNT

- Liu and Yang, ***Phys. Rev. B*** 86, 104307 (2012)



Simulation: Single extended polymer chains

Divergent and Ultrahigh Thermal Conductivity in Millimeter-Long Nanotubes

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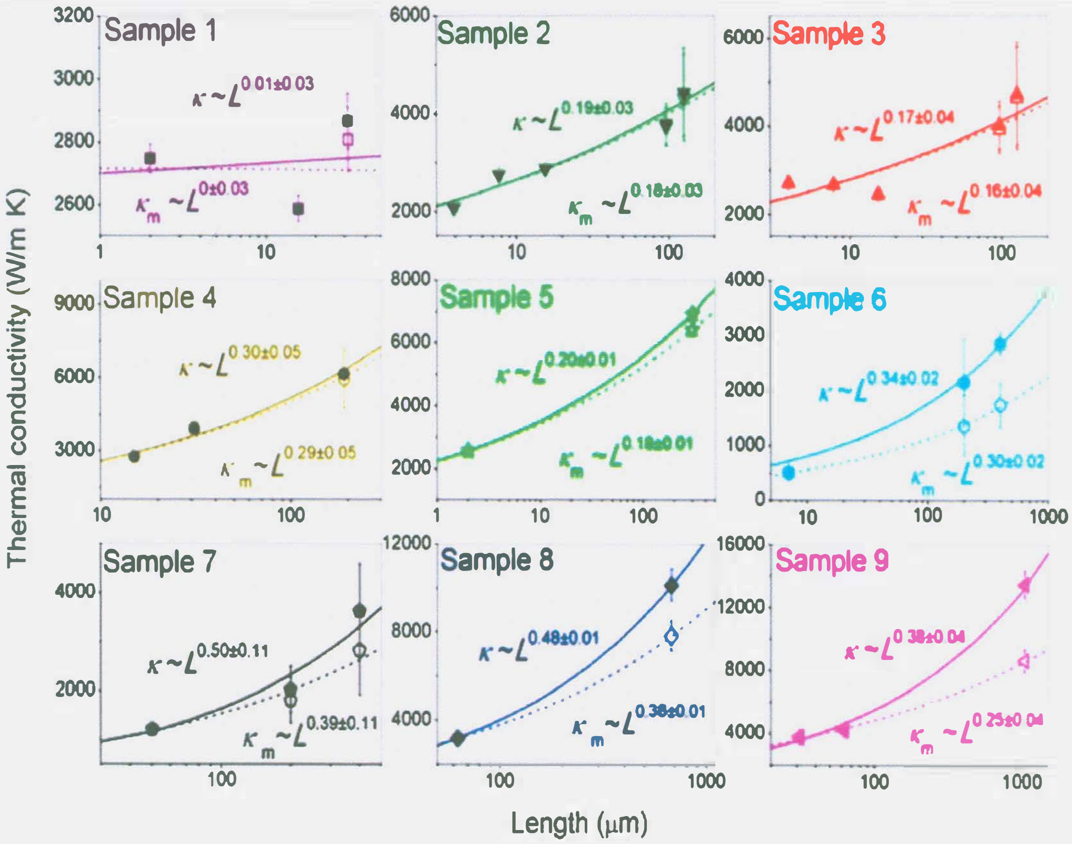


FIG. 3. κ vs L relations for nine different CNTs. Both measured κ_m's (open symbols) and corrected κ's (solid symbols, after incorporating radiation heat loss from the surface of CNTs) are shown for each sample. The measured κ_m's and corrected κ's are almost identical for $L < 100\ \mu$m. For the longest CNT investigated ($L = 1.039$ mm), the measured κ_m and the corrected κ reach 8640 and 13 300 W/mK, respectively. The fits (by parametrizing $\kappa \sim L^{\alpha}$) to the corrected κ's and measured κ_m's are shown by solid cu ves and dashed curves, respectively.

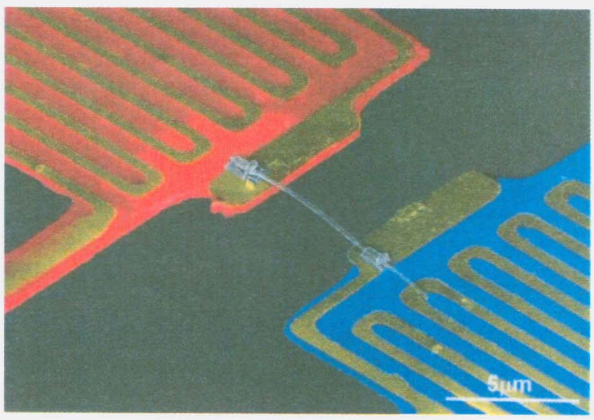


Fig. 8.5 A false colored SEM image of a thermal conductivity test fixture consisting of suspended heater and sensor pads with a SiGe nanowire anchored on it

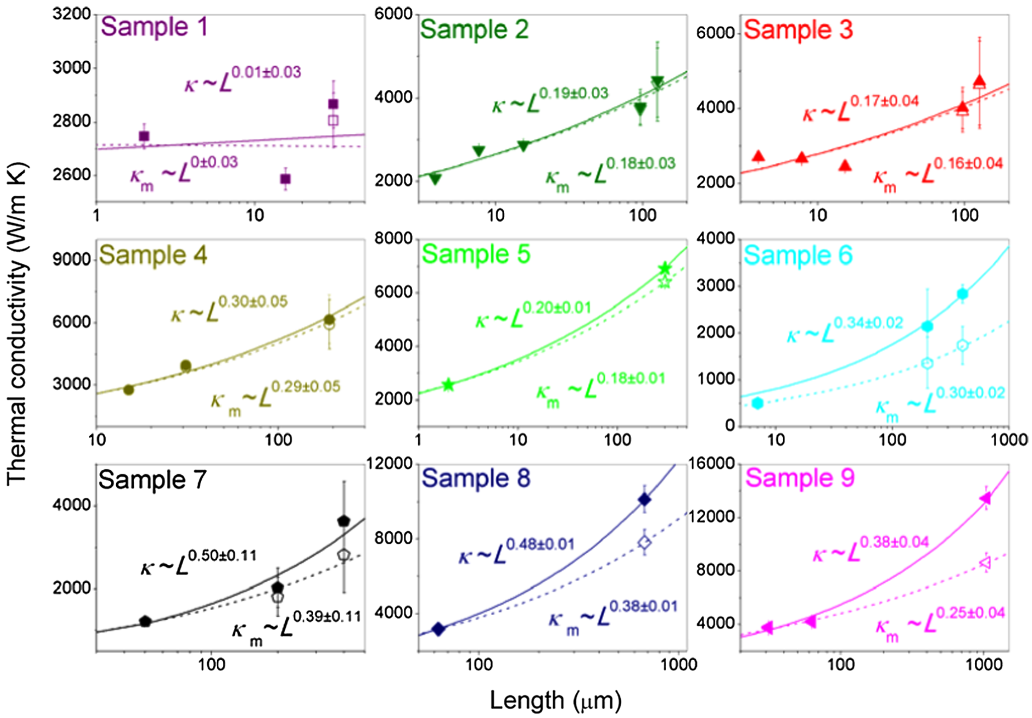


FIG. 3. κ vs L relations for nine different CNTs. Both measured κ_m's (open symbols) and corrected κ's (solid symbols, after incorporating radiation heat loss from the surface of CNTs) are shown for each sample. The measured κ_m's and corrected κ's are almost identical for $L < 100\ \mu$m. For the longest CNT investigated ($L = 1.039$ mm), the measured κ_m and the corrected κ reach 8640 and 13 300 W/mK, respectively. The fits (by parametrizing $\kappa \sim L^{\alpha}$) to the corrected κ's and measured κ_m's are shown by solid curves and dashed curves, respectively.

have $L^{1-\alpha} \gg K_s/K_c$ in Eq. (4) and the effect of contact thermal resistance vanishes when $L \gg 1\ \mu$m. Additionally, the effect of contact thermal resistance should be limited; for example, $K_s/K_c > 5$ would indicate that the intrinsic κ of a 1 μm-long CNT is larger than 18 000 W/m K, violating quantum mechanical constraints for a CNT [28,29]. Further analyses using Eq. (4) suggest that $0.17 < \alpha < 0.43$ and $K_s/K_c < 0.3$ yield good fits to the experimental data [26]. Figure 4 also shows a controlled experiment on a SiN_x beam displaying the expected diffusive thermal conduction, demonstrating the validities of our measurements and analyses. Therefore, we conclude that the experimentally observed divergent behavior of κ originates from the intrinsic properties of the ultralong CNTs, but not from artifacts of contact thermal resistance.

Because naturally abundant ethanol vapor was used as the synthetic source, isotopic impurities (98.9% ^{12}C and 1.1% ^{13}C) are expected in the investigated CNTs. In addition, impurities and defects are unavoidable for the ultralong CNTs. Furthermore, TEM images reveal a thin layer (~2 nm) of amorphous carbon covering some parts of the CNTs [26]. Surprisingly, the pronounced power-law divergence of κ emerges regardless of these structural imperfections and external perturbations. The result is consistent with 1D disordered models that show robust anomalous thermal conduction phenomena against defects or disorders [5]. But it disagrees with the prediction that the divergent behavior of κ would disappear when defects are introduced in CNTs [9,16]. We thus demonstrate that the divergence of κ persists for much longer distances than theoretically anticipated [9,10,16]. Our results also resolve the decade-long debate of whether the κ of a CNT would continue to diverge or saturate for $L > 1\ \mu$m [11–17]. The finding indicates that the wave properties of heat can be transmitted for much longer distances than previously thought, and it highlights the important contributions of long-wavelength phonons in low-dimensional systems.

Unlike electrical conductivity of materials that can vary by more than 27 orders of magnitude from insulators to metals,

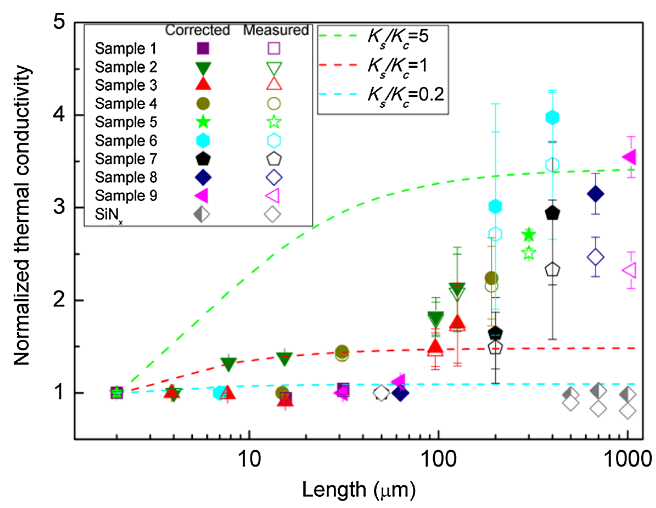


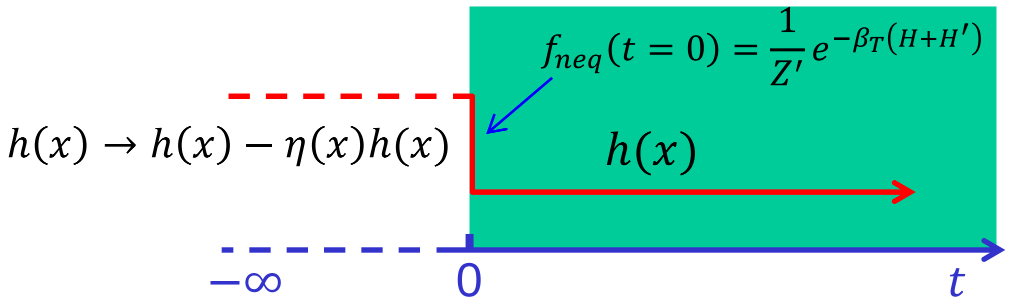
FIG. 4. Normalized κ vs L for the investigated samples. Here the corrected κ's (solid symbols) and measured κ_m's (open symbols) are normalized, respectively, by those of each sample's shortest L. The effects of contact thermal resistance from small ($K_s/K_c = 0.2$) to large ($K_s/K_c = 5$) are calculated using Eq. (4) (with $\alpha = 0$), demonstrating that the observed divergent of κ or κ_m cannot be attributed to contact thermal resistances adding to a diffusive thermal conductor. A controlled experiment on a SiN_x beam shows the expected normal thermal conduction.

Open Problems

- Analytical results in anomalous heat diffusion are only available for CTRW models
- General theory for heat diffusion, especially in *solids*, is unknown

- The general features of (anomalous) heat diffusion beyond the phenomenological Fourier law and exists there a generalized heat equation?
- The fundamental connection between (anomalous) heat diffusion and (anomalous) heat conduction.

Energy diffusion



- 1D homogeneous system $H = \int h(x) dx$

- equilibrium for system H: $f_{eq} = e^{-\beta_T H}/Z$

1. nonequilibrium initial condition at $t = 0$;

$$f_{neq}(t=0) = \frac{1}{Z'} e^{-\beta_T (H+H')}$$

$$H' = -\int \eta(x) h(x) dx , \qquad Z' = \int e^{-\beta_T (H+H')} d\Gamma$$

$\eta(x)$ depicts the initial energy distribution.

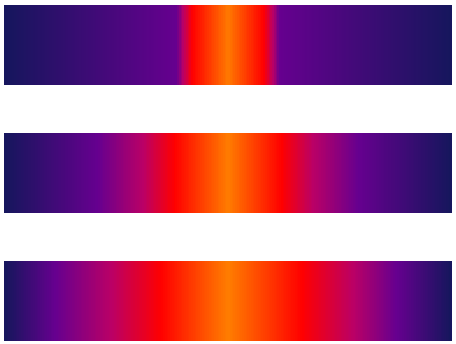
$$h(x) = \sum \delta(x - x_n) h_n$$

Excess Energy Density

2. Evolve according to Liouvillian dynamics

$$\partial_t f_{neq}(t) = L f_{neq} = \left\{H, f_{neq}\right\} \quad (\boldsymbol{t > 0})$$

f_{neq}: *phase space density*



3. Calculate the excess energy density in space

$$\delta\langle h(x,t)\rangle_{neq} = \langle h(x,t)\rangle_{neq} - \langle h(x)\rangle_{eq}$$

$$= \int h(x) f_{neq}(t)\, d\Gamma - \int h(x) f_{eq}\, d\Gamma$$

Preparation of Initial Condition

- The initial condition can be prepared by applying at infinite past the perturbation $h(x) \to [h(x) - \eta(x)h(x)]$, with the perturbation suddenly switched off at $t = 0$.

- If a concept of local temperature exist, it enters the result via the preparation of initial condition

$$\eta(x) = \delta T(x)/T$$

In such a case, the Hamiltonian density $h(x)$ couples formally to the conjugated thermodynamic affinity $\delta T(x)/T$ at $t = 0$

- Note: the concept of a time-dependent local equilibrium temperature $T(x,t)$ is NOT needed in the diffusion process.

Linear Response Result

- We calculate $\delta\langle h(x,t)\rangle_{neq}$ to the first order of $\eta(x)$

$$\delta\langle h(x,t)\rangle_{neq} = \frac{1}{k_B T}\int \langle \Delta h(x,t)\Delta h(x',0)\rangle_{eq}\eta(x')dx'$$

$$= \frac{1}{k_B T}\int C_{hh}(x-x',t)\eta(x')dx'$$

For heat equation $\epsilon(x,t) = \int \Phi(x-x',t)\eta(x')dx'$

- The conservation of total excess energy

$$\delta E(t) = \int \delta\langle h(x,t)\rangle_{neq}dx = const = cT\int \eta(x)dx$$

- The normalized excess energy density (**NOT** a probability density)

$$\rho_E(x,t) = \frac{\delta\langle h(x,t)\rangle_{neq}}{\delta E(t)} = \frac{1}{N}\int C_{hh}(x-x',t)\eta(x')dx'$$

Normalization constant $N = k_B T^2 c\int \eta(x)dx$

$\rho_E(x,t)$ can be negative!

The Mean Square Deviation (MSD)

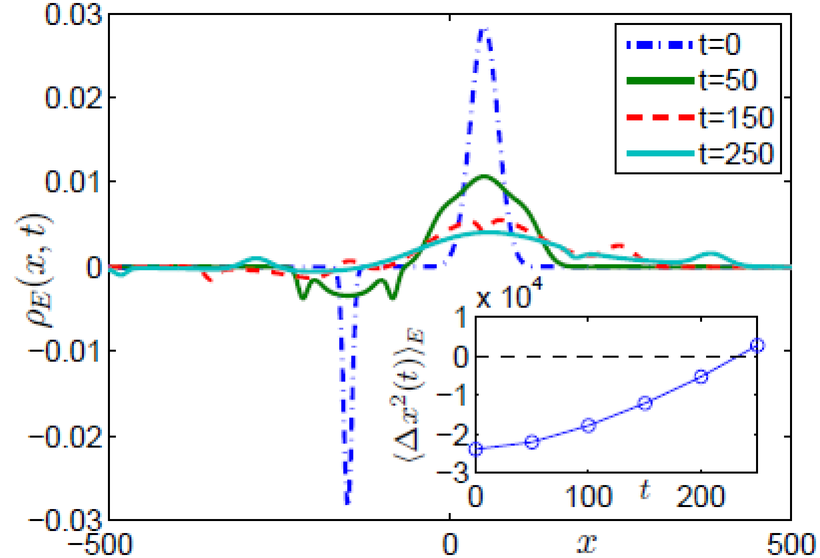
➢ Define the mean square deviation of energy diffusion as

$$\langle \Delta x^2(t) \rangle_E = \int (x - \langle x \rangle_E)^2 \rho_E(x,t) dx = \langle x^2(t) \rangle_E - \langle x \rangle_E^2.$$

$\langle x \rangle_E = \int x \rho_E(x,t) dx$ is a constant for homogeneous systems

- It is the variance for the excess energy distribution rather than the $\langle [x(t) - x(t_0)]^2 \rangle$ as for particle diffusion

- $\langle \Delta x^2(t) \rangle_E$ can be negative



For FPU model

The Evolution of the MSD

Acceleration:

$$\frac{d^2\langle\Delta x^2(t)\rangle_E}{dt^2} = \frac{1}{N}\iint x^2 \frac{\partial^2 C_{hh}(x-x',t)}{\partial t^2}\eta(x')dxdx'$$

$$= \frac{1}{N}\iint x^2 \frac{\partial^2 C_{jj}(x-x',t)}{\partial x^2}\eta(x')dxdx'$$

$$= \frac{2C_{JJ}(t)}{k_B T^2 c}$$

$$\frac{\partial^2 C_{hh}(x,t)}{\partial t^2} = \frac{\partial^2 C_{jj}(x,t)}{\partial x^2}$$

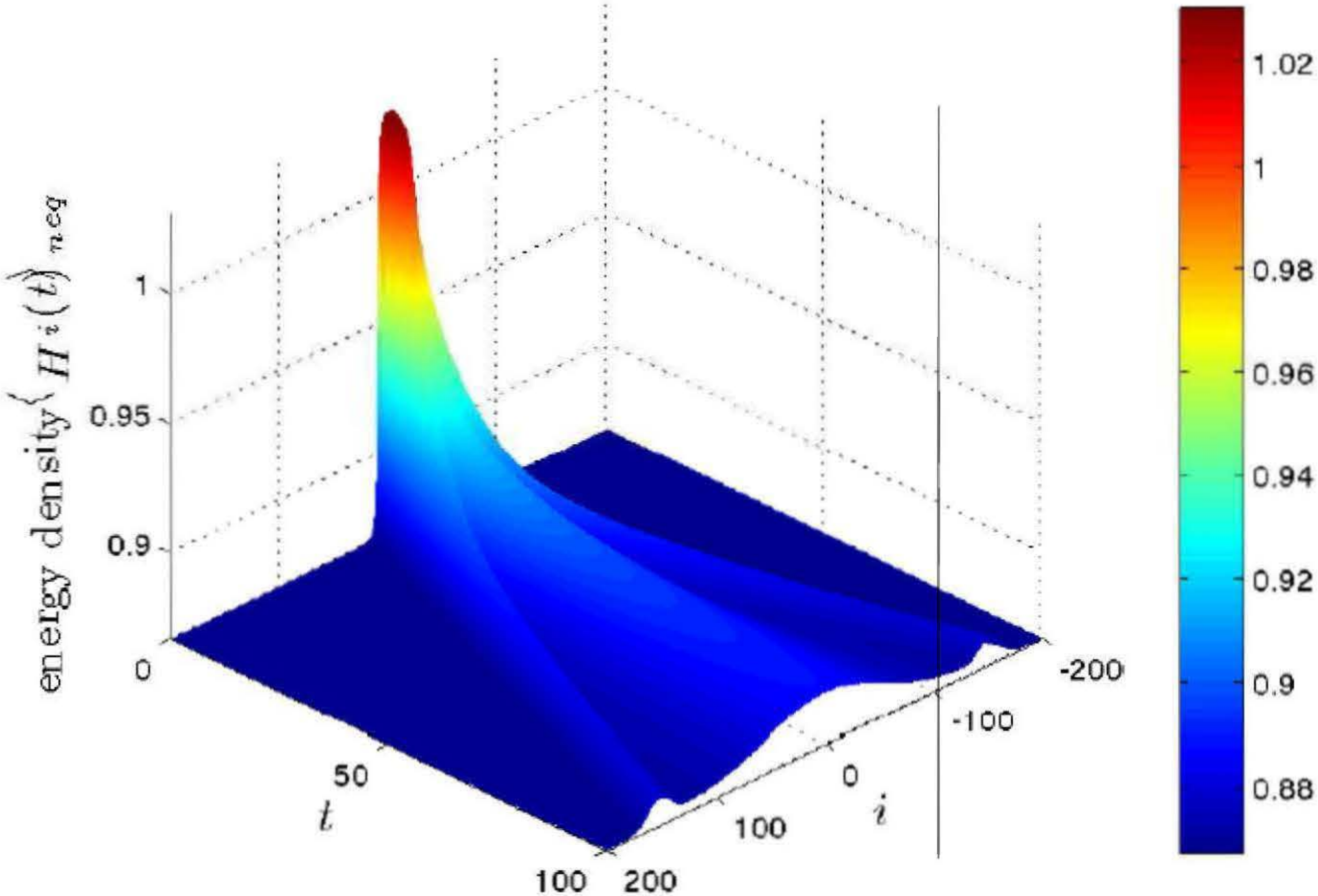
Heat flux auto-correlation function: $C_{JJ}(t) = \lim_{L\to\infty}\frac{1}{L}\langle J_L(t)J_L(0)\rangle_{eq} = \int C_{jj}(x,t)dx$

Volumetric heat capacity: c

Initial conditions:

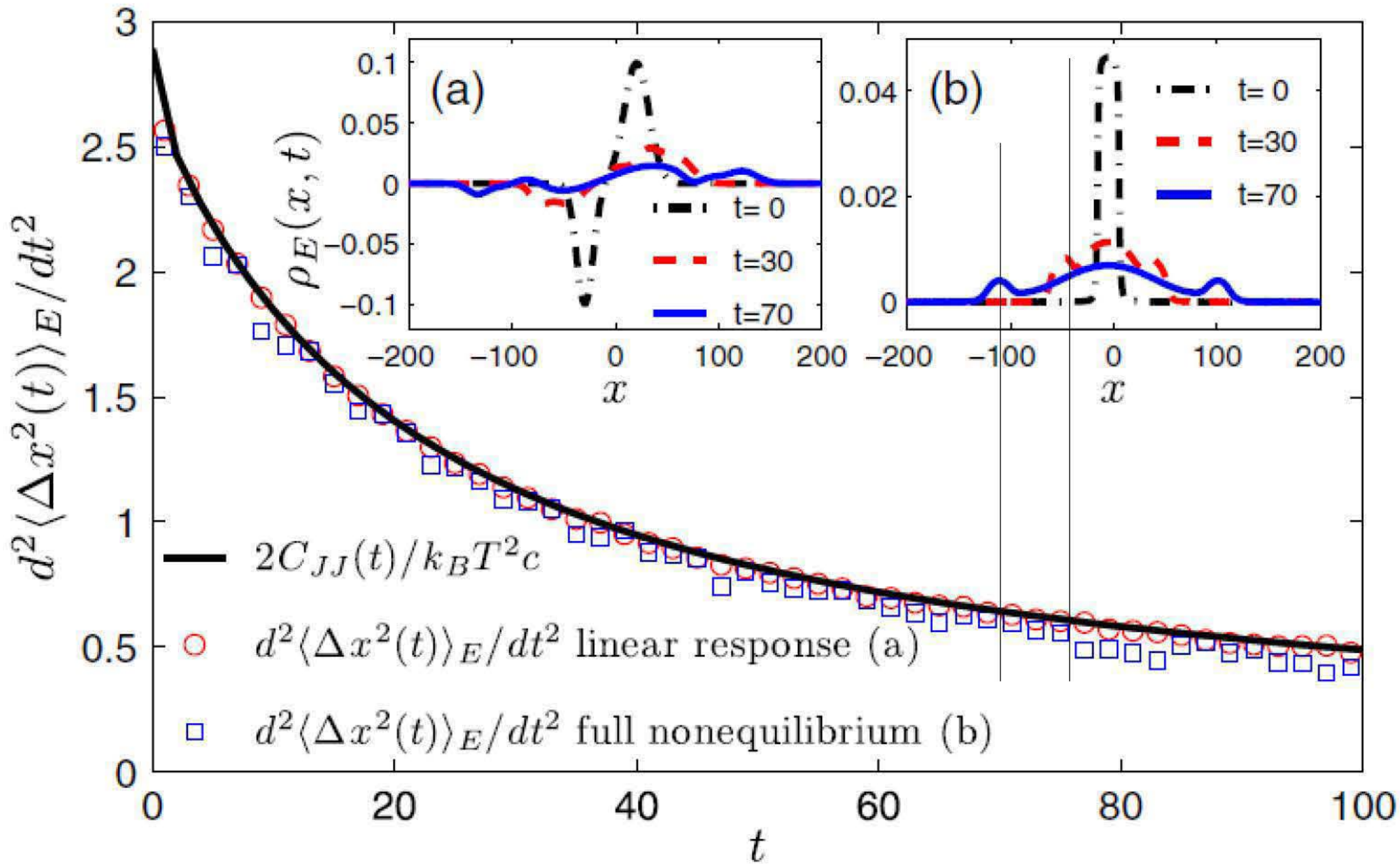
$\langle\Delta x^2(t=0)\rangle_E$ depends on initial energy profile $\eta(x)$; $\left.\frac{d\langle\Delta x^2(t)\rangle_E}{dt}\right|_{t=0} = 0$

Energy Diffusion



Time evolution of the nonequilibrium energy density for a manifest near equilibrium energy diffusion dynamics

Numerical Validation



Discussion

The Assumptions used in the derivation:

Ergodicity

- No nonstationary (i.e. aging) phenomena for long time correlations.
- Not applicable to all anomalous subdiffusive and superdiffusive energy diffusion processes that undergo aging, e.g. in many CTRW models. (however, those models lack a microscopic Hamiltonian basis)
- Applicable to ergodic anomalous diffusion dynamics stemming from a generalized Langevin equation, driving by fractional Brownian motion

The Helfand Moment

- Green-Kubo formula $$\kappa = \frac{1}{k_B T^2} \int_0^{\infty} C_{JJ}(t')dt'$$

- The Helfand relation for normal heat conduction (equivalent to Green-Kubo)

$$\kappa = \lim_{t\to\infty} \frac{1}{2k_B T^2 t} \lim_{L\to\infty} \frac{\langle [G(t) - G(0)]^2 \rangle_{eq}}{L} \equiv \lim_{t\to\infty} \frac{\langle \Delta\mathcal{G}^2(t) \rangle_{eq}}{2k_B T^2 t}$$

Helfand moment for heat conduction:

$$G_L(t) = \sum_i x_i \left(E_i - \langle E_i \rangle_{eq} \right) = \int xh(x,t)dx\,; \quad \frac{dG_L}{dt} = J_L = \int j(x,t)dx$$

- Helfand ***Phys. Rev.*** 119, 1 (1960)
- Viscardy, Servantie and Gaspard, ***J. Chem. Phys.*** 126, 184513 (2009)
- Gaspard and Gilbert, ***J. Stat. Mech.*** P11021 (2008)

The Helfand Moment

- It can be shown that

$$\frac{d^2\langle\Delta\mathcal{G}^2(t)\rangle_{eq}}{dt^2} = 2C_{JJ}(t) = k_B T^2 c \frac{d^2\langle\Delta x^2(t)\rangle_E}{dt^2}$$

- Initial condition: $\left.\frac{d\langle\Delta\mathcal{G}^2(t)\rangle_{eq}}{dt}\right|_{t=0} = 0$; $\langle\Delta\mathcal{G}^2(t=0)\rangle_{eq=0}$ (always positive)

- Helfand ***Phys. Rev.*** 119, 1 (1960)
- Viscardy, Servantie and Gaspard, ***J. Chem. Phys.*** 126, 184513 (2009)
- Gaspard and Gilbert, ***J. Stat. Mech.*** P11021 (2008)

Time-local Helfand Relation

$$\frac{d^2\langle \Delta x^2(t)\rangle_E}{dt^2} = \frac{2C_{JJ}(t)}{k_B T^2 c} \quad , \qquad \left.\frac{d\langle \Delta x^2(t)\rangle_E}{dt}\right|_{t=0} = 0$$

$$\Rightarrow \quad \frac{d\langle \Delta x^2(t)\rangle_E}{dt} = \frac{2}{k_B T^2 c}\int_0^t C_{JJ}(t')dt' = \frac{1}{k_B T^2 c}\frac{d\langle \Delta \mathcal{G}^2(t)\rangle_{eq}}{dt}$$

- Comparing with the ordinary Helfand relation $\kappa = \frac{1}{2k_B T^2}\lim_{t\to\infty}\frac{\langle \Delta \mathcal{G}^2(t)\rangle_{eq}}{t}$ for normal transport, here we obtain a *time-local version*
- i.e. $\frac{d\langle \Delta x^2(t)\rangle_E}{dt}$ instead of $\frac{\langle \Delta x^2(t)\rangle_E}{t}$
- Is in suitable form to establish the connection between (anomalous) heat diffusion and (anomalous) heat conduction

Normal Energy Transport

$$\lim_{t\to\infty}\frac{\langle \Delta x^2(t)\rangle_E}{t} = 2D_E \quad \text{or} \quad \langle \Delta x^2(t)\rangle_E \sim 2D_E t$$

Green-Kubo Formula $\quad \kappa = \frac{1}{k_B T^2}\int_0^\infty C_{JJ}(t')dt' \Leftarrow \boxed{\frac{d^2\langle \Delta x^2(t)\rangle_E}{dt^2} = \frac{2C_{JJ}(t)}{k_B T^2 c}}$

$$\kappa = \frac{c}{2}\lim_{t\to\infty}\frac{d\langle \Delta x^2(t)\rangle_E}{dt} = cD_E$$

thermal conductivity= volumetric heat capacity × thermal diffusivity

Do not require $\langle \Delta x^2(t)\rangle_E = 2D_E t$

Do not require heat equation,

do not require local relation between j and ∇T

Superdiffusive Energy Transport

$$\langle \Delta x^2(t) \rangle_E \sim t^{\beta} \ (1 < \beta \leq 2)$$

$$\boxed{\frac{d^2 \langle \Delta x^2(t) \rangle_E}{dt^2} = \frac{2C_{JJ}(t)}{k_B T^2 c}}$$

$$\kappa = \frac{1}{k_B T^2} \lim_{t \to \infty} \int_0^t C_{JJ}(t') dt' = \frac{c}{2} \lim_{t \to \infty} \frac{d \langle \Delta x^2(t) \rangle_E}{dt} \qquad \text{diverges!}$$

If one is interested in the length dependence of thermal conductivity, the usual procedure is then to put a cut-off time $t \sim L/v_s$ in the upper limit of the integral and consider a length-dependent thermal conductivity.

❑ Lepri, Livi and Politi, ***Phys. Rep.*** 377, 1 (2003)

❑ Dhar, ***Adv. Phys.*** 57. 457 (2008)

$$\kappa(L) \sim \frac{1}{k_B T^2} \int_0^{\frac{L}{v_s}} C_{JJ}(t') dt' = \frac{c}{2} \frac{d \langle \Delta x^2(t) \rangle_E}{dt} \bigg|_{t = \frac{L}{v_s}} \sim \ L^{\beta - 1}$$

$$\kappa(L) \sim L^{\alpha}, \qquad \alpha = \beta - 1$$

Subdiffusive Energy Diffusion

$$\frac{d^2\langle \Delta x^2(t)\rangle_E}{dt^2} = \frac{2C_{JJ}(t)}{k_B T^2 c}$$

$$\langle \Delta x^2(t)\rangle_E \sim t^\beta \ (0 < \beta < 1)$$

$$\kappa = \frac{1}{k_B T^2} \lim_{t\to\infty} \int_0^t C_{JJ}(t')dt' = \frac{c}{2} \lim_{t\to\infty} \frac{d\langle \Delta x^2(t)\rangle_E}{dt} = 0,$$ thermal insulator

Following the same reasoning used in the study of heat conduction in open systems with Markovian-heat baths, but with the spectral densities for the heat baths suitably tailored in order to yield such subdiffusive heat diffusion, we still use the cut-off time to get

$$\kappa(L) \sim L^\alpha, \qquad \alpha = \beta - 1$$

Take Home Messages I

- we studied (anomalous) heat diffusion in absence of ergodicity breaking.

- The excess energy distribution in nonequilibrium energy diffusion is a convolution of the Green function and the initial energy profile. (*similar to the solution of heat equation*) The Green function is given by the canonical energy-energy correlation function.

- $$\frac{d^2\langle \Delta x^2(t)\rangle_E}{dt^2} = \frac{2C_{JJ}(t)}{k_B T^2 c}$$

Take Home Messages I continued

- Given the premise that anomalous stationary heat flux follows anomalous length dependent heat conductance $\kappa(L) \sim L^{\alpha}$, then MSD $\sim t^{\beta}$ implies $\alpha = \beta - 1$.

- The dynamical relations we derived applies for all times t, therefore it can be invoked as well for those intermediate cases where anomalous, length-dependent heat conductivity occurs over a finite size.

Lecture Notes in Physics 921

Stefano Lepri *Editor*

Thermal Transport in Low Dimensions

From Statistical Physics to Nanoscale Heat Transfer

Springer

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PAPER

1D momentum-conserving systems: the conundrum of anomalous versus normal heat transport

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Keywords: heat diffusion, momentum diffusion, viscosity

effective viscosity in the spirit defined above. In contrast, a Fourier-like behavior may become possible if the inherent momentum dynamics is more fluid-like, consequently possessing a finite effective momentum diffusivity. An appealing conjecture therefore is that it is the physics of momentum diffusion which rules whether heat transport occurs normal or anomalous. In short, we next test with different models the following hypothesis:

(i) *Heat transport in nonlinear 1D momentum-conserving Hamiltonian lattice systems occurs normal whenever the spread of the profile of the excess momentum density, upon subtracting a possibly present leading ballistic part, is normal.*

(ii) *The corollary being that heat transport occurs anomalous whenever this so adjusted, subleading momentum excess density spreads superdiffusive.*

If this hypothesis holds true it is expected to hold vice versa, i.e., with heat/momentum substituted by momentum/energy.

Colloquium:
Phononics: Manipulating heat flow with electronic analogs and beyond

N. Li, J. Ren, L. Wang, G. Zhang,
P. Hänggi, and B. W. Li

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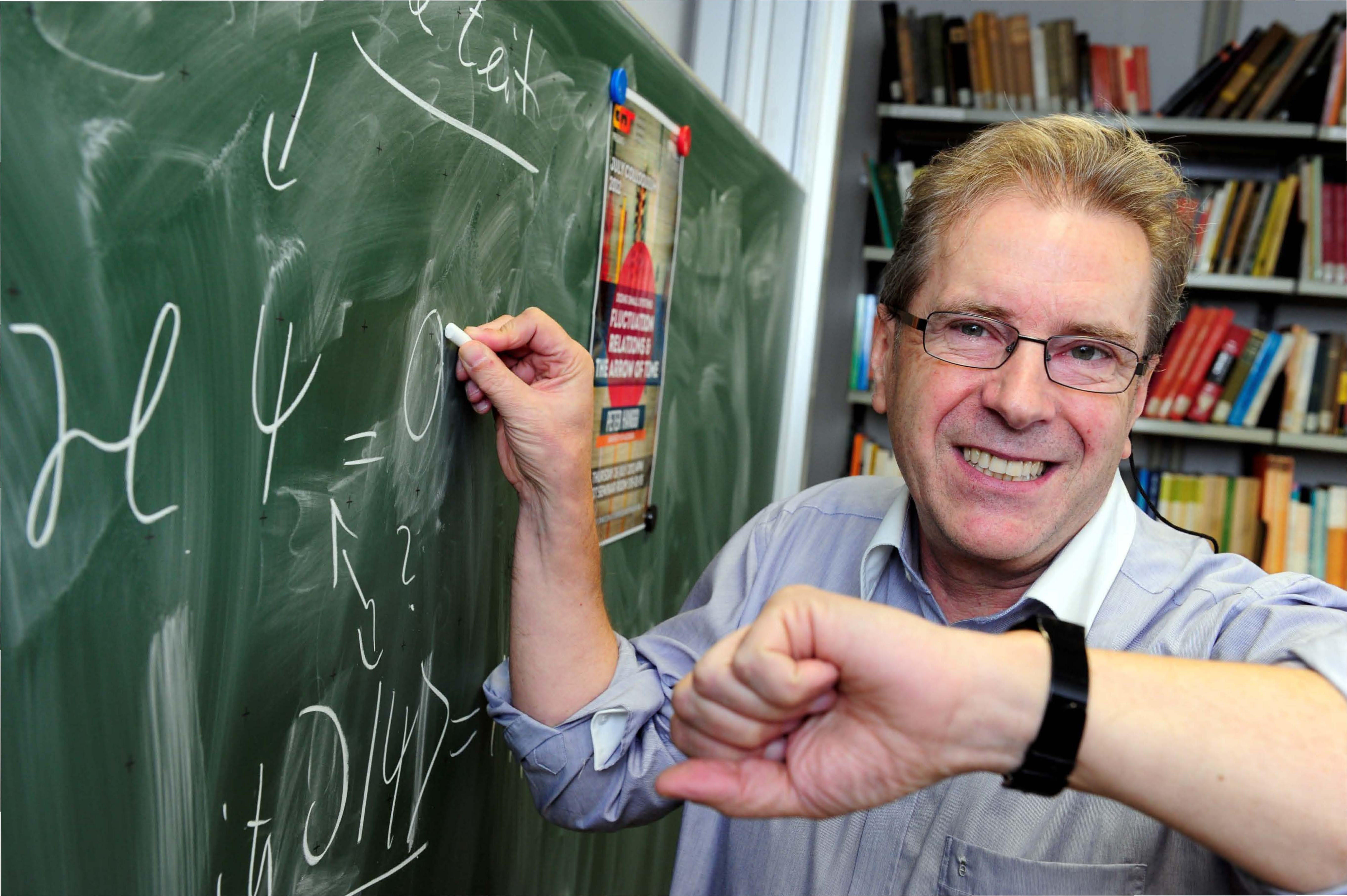
FLUCTUATION RELATIONS &
THE ARROW OF TIME

Triggering Waves in Nonlinear Lattices:

Quest for anharmonic phonons and corresponding Mean Free Paths (MFP)

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Phonon concept

- the phonon is one of the most important concepts in solid state physics.

  - eigenstates of lattice vibration, harmonic chain, superposition, wavelength, frequency, dispersion relation …
  - phonon-phonon interaction, relaxation time, mean free path …
  - sound, elastic phonon, acoustic phonons, optical phonons,…

Kinetic theory of heat conduction: Peierls-Boltzmann:

$$\kappa = \sum c_k v_k l_k$$

Open problems

- **Anharmonic lattices:** Equations of motion cannot be decoupled into eigenmodes

➢ **Then:** what is the "meaning" of a phonon ?!!

➢ dispersion relation ?

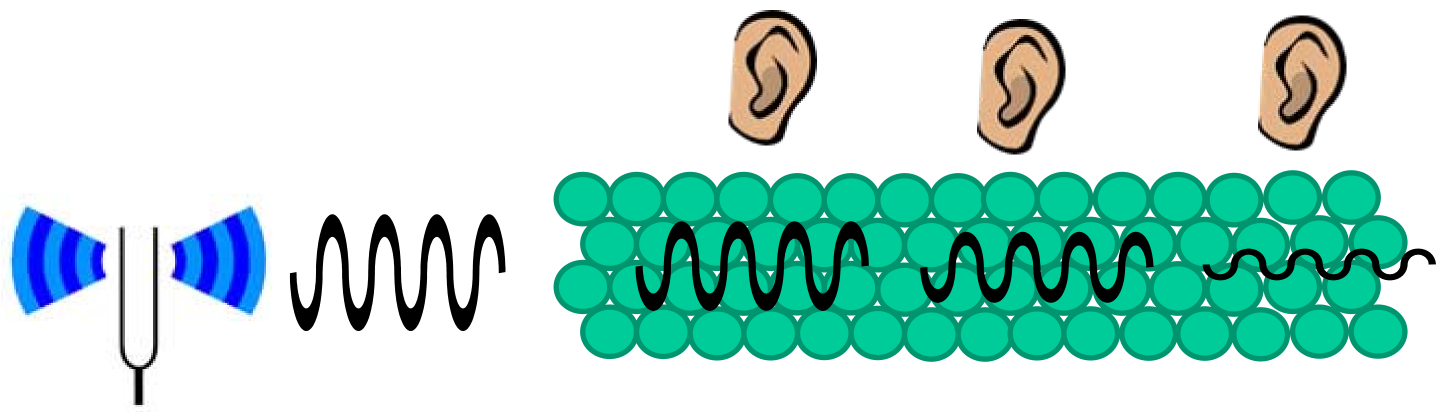
- **A priori**: a phonon mean free path and/or a phonon relaxation time cannot be justified for anharmonic lattices. →which "effective phonon" makes sense?

- Is a single-mode relaxation time approximation feasible → yielding MFP/relaxation time ?

➢ Anomalous heat conductivity: a MFP seemingly nonexisting/**divergent**

All these can be solved only if we can really "see" phonons

Tuning fork experiment



- Identify phonon by observing the propagating sound in the lattices
  - Is it wave-like? (wavelength, frequency …)
  - Does it decay exponentially? (mean free path)

$$v_n(t) = A(n)\cos(\omega t + kn) \quad A(n) = e^{-\frac{n}{\mathcal{L}}}$$

Triggering anharmonic phonons in anharmonic lattices

- Nonlinear equations of motion for anharmonic lattices are not analytically solvable. A different approach is needed.
- Nonlinear media may generate a multi-frequency response for a single-frequency input. However, if the driving force is small, we are in the **linear response regime** → the output will still be at the input single-frequency only.

Classical Linear Response Result

$$H_0 = \sum_{n=1}^{N} \left[\frac{p_n^2}{2} + V(x_{n+1} - x_n) + U(x_n) \right]$$

$$H_{tot} = H_0 + H_{ext} = H_0 - f_d(t) x_1$$

$$f_d(t) = f_1 \cos \omega t$$

Linear Response of the particle velocities

$$\begin{aligned} \langle v_n(t) \rangle_f &= \beta_T \int_{-\infty}^{t} ds \, \langle v_n(t-s) v_1(0) \rangle \, f_d(s) \\ &= f_1 \mathrm{Re}[\underline{\chi_n(\omega)} e^{i\omega t}] \end{aligned}$$

$$\chi_n(\omega) = \beta_T \int_0^{\infty} d\tau \, \langle v_n(\tau) v_1(0) \rangle \, e^{-i\omega\tau} \equiv |\chi_n(\omega)| e^{i\phi_n}$$

Therefore $\langle \boldsymbol{v_n(t)} \rangle_{\boldsymbol{f}} = \boldsymbol{f_1 \, |\chi_n(\omega)| \cos(\omega t + \phi_n)}$

Expectations for anharmonic phonons

$$\langle v_n(t)\rangle_f = f_1\,|\chi_n(\omega)|\cos(\omega t + \phi_n)$$

- ϕ_n is linearly dependent on n: $\phi_n = -k\,n + \phi_0$ so that a wavenumber can be defined for the excitation. The relation between k and ω give the dispersion relation

- $|\chi_n|$ exponentially decays with n: $|\chi_n| \sim e^{-k'n}$ so that an MFP l can be defined $l = 1/k'$
  l is frequency dependent

Models

$$H = \sum \frac{p_i^2}{2m} + \sum V(x_i - x_{i+1}) + \sum U(x_i)$$

FPU-β Model $V(x) = \frac{1}{2}x^2 + \frac{1}{4}x^4 \quad U(x) = 0$

FPU-$\alpha\beta$ Model $V(x) = \frac{1}{2}x^2 + \frac{1}{3}x^3 + \frac{1}{4}x^4 \quad U(x) = 0$

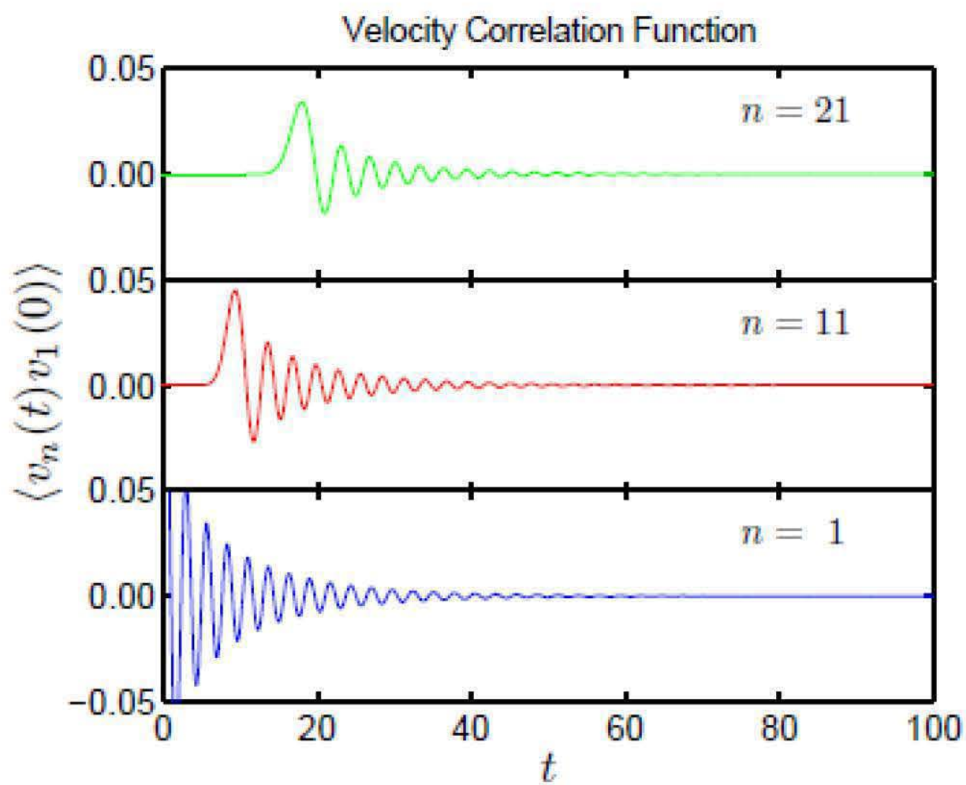
Phi4 Model $V(x) = \frac{1}{2}x^2 \quad U(x) = \frac{1}{4}x^4$

Numerical details

1. The v-v correlation $\langle v_n(t) v_1(0) \rangle$ is calculated for $n = 1, 2, \cdots, N$ and $t = 0, h, 2h, \cdots, t_m$

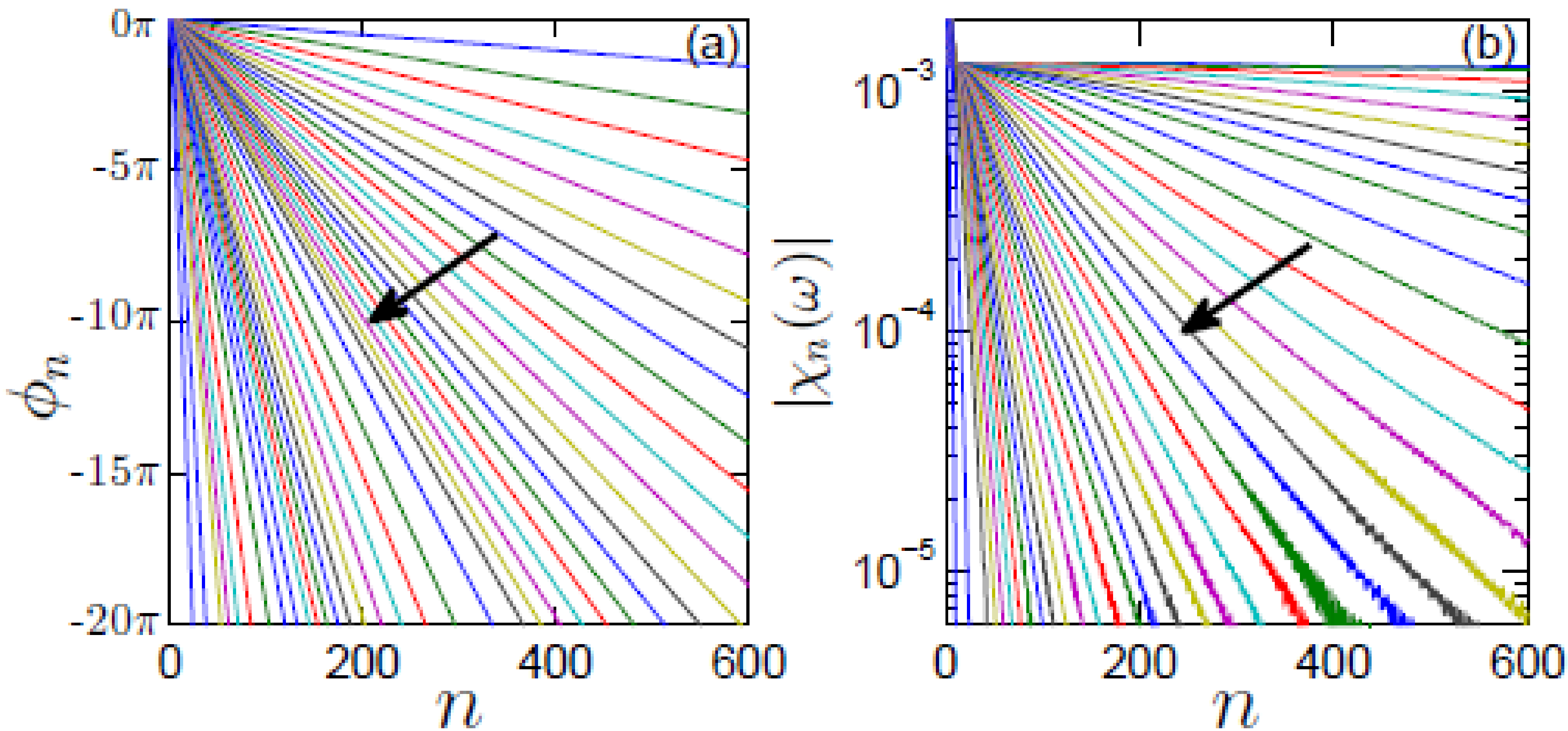
$$N = 2048$$
$$h = 0.02$$
$$t_m = 655.36$$

2. Fourier transform to obtain $\chi_n(\omega)$



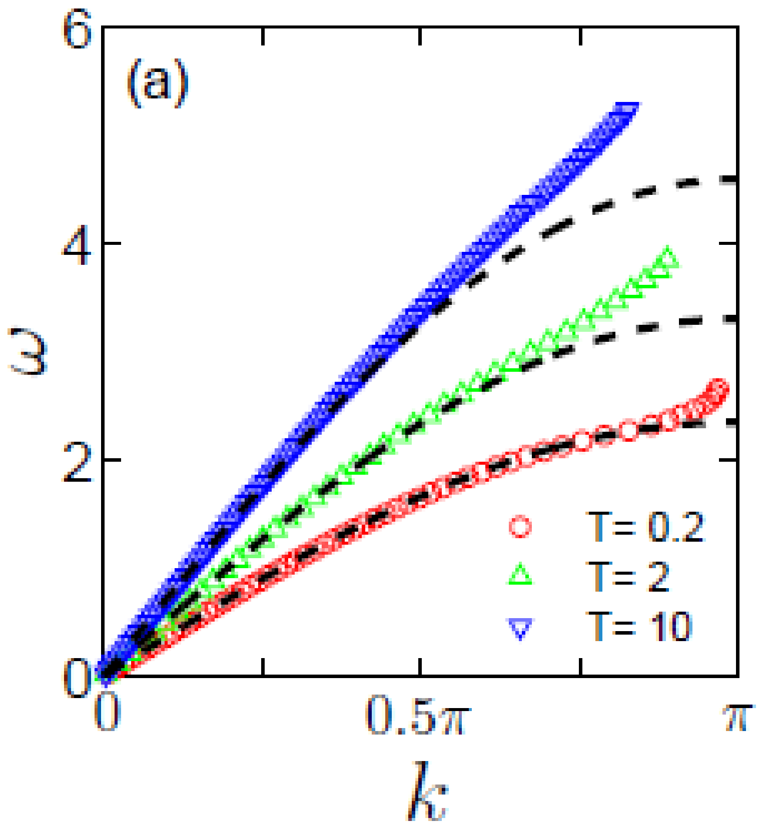
For FPU-β model at T=0.2.
Other models display similar oscillatory behavior

FPU-β model



- Perfect linear dependence of ϕ_n on n → **anharmonic phonon can be defined**
- Perfect exponential decay of $|\chi_n|$ → **MFP can be defined**

FPU-β Dispersion Relation



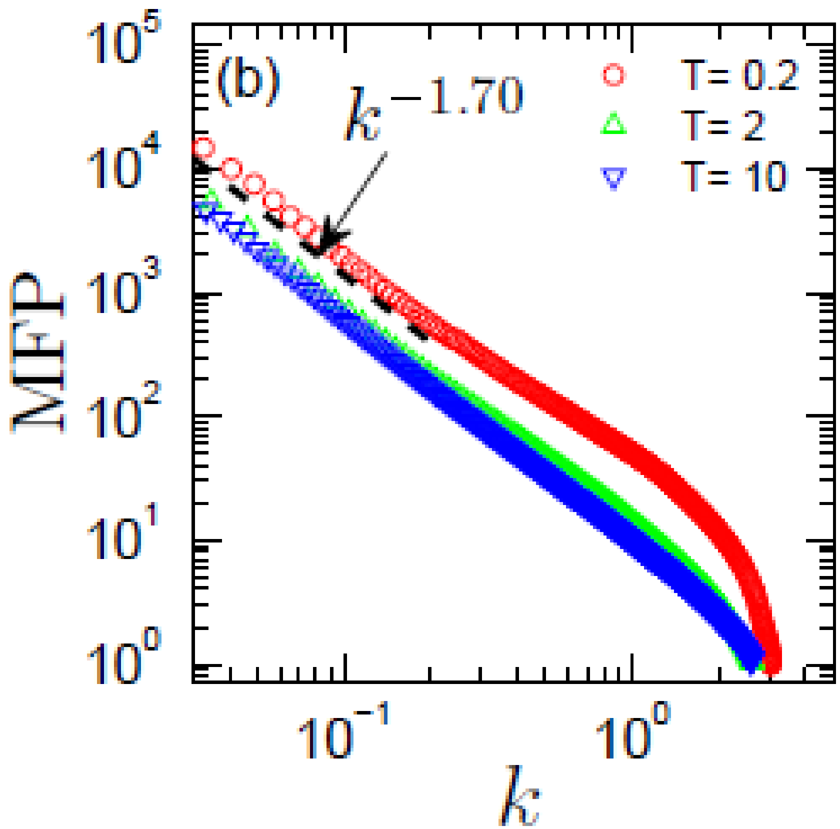
Dashed lines:
Effective phonon theory

$$\omega = 2\sqrt{\alpha_1}\sin\frac{k}{2}$$

$$\alpha_1 = 1 + \frac{\int x^4 \mathrm{e}^{-(x^2/2+x^4/4)/T}\,\mathrm{d}x}{\int x^2 \mathrm{e}^{-(x^2/2+x^4/4)/T}\,\mathrm{d}x} \approx 1.37$$

- N. Li, *EPL* **75,** 49 (2006)

FPU-β MFP



Dashed line: power law

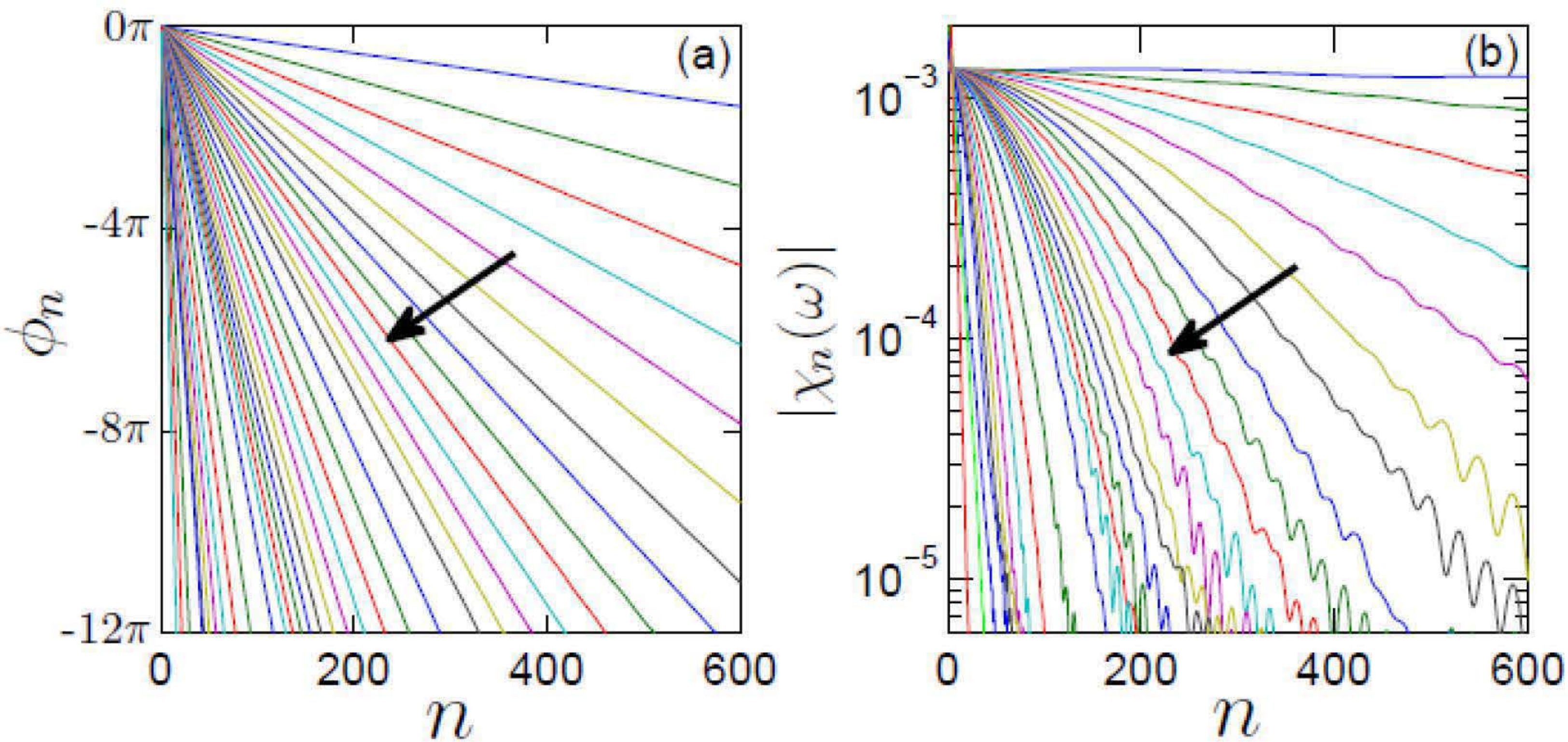
$$l(k) \sim k^{-\mu} \approx k^{-1.70}$$

Related to anomalous heat conduction

$$\kappa = 1 - \frac{1}{\mu} \approx 0.411$$

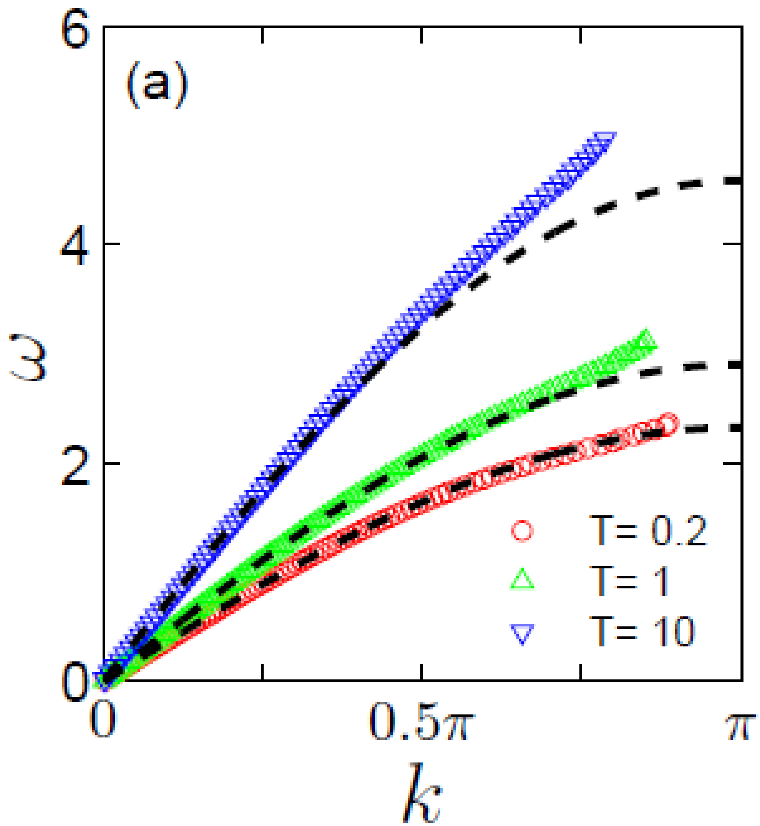
- A. Dhar, ***Adv Phys* 57** 457 (2008)
- A. Pereverzev ***Phys. Rev. E* 68** 056124 (2003)

FPU-$\alpha\beta$ model



- Perfect linear dependence of ϕ_n on n → **anharmonic phonon can be defined**
- non-exponential decay of $|\chi_n|$ → **MFP can not be well defined !!**

FPU-$\alpha\beta$: Dispersion Relation



No effective phonon theory exists for FPU-$\alpha\beta$ model

Dashed line:

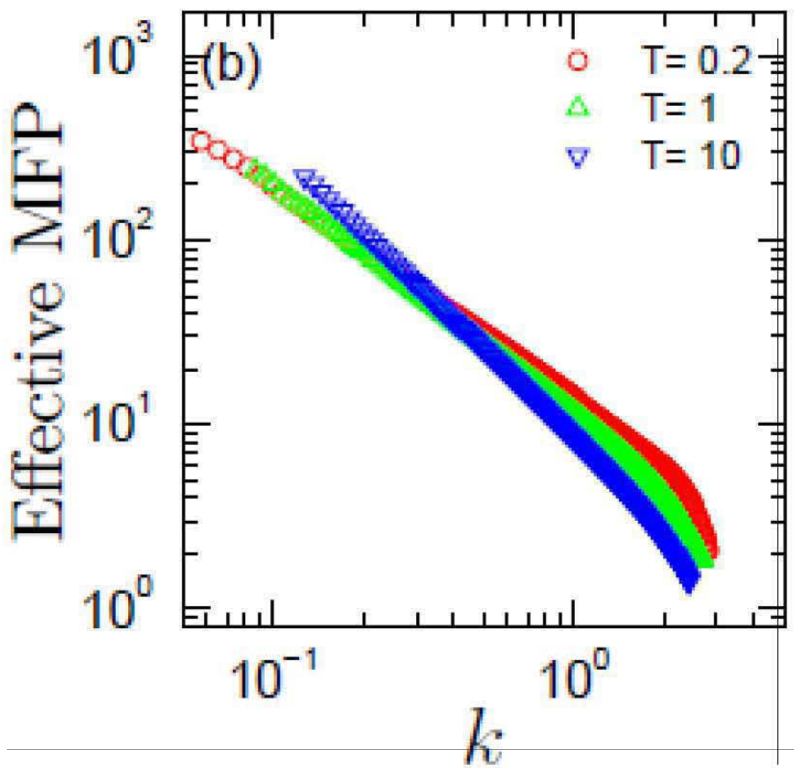
$$\omega = 2\alpha(T)\sin\frac{k}{2}$$

$$\alpha^2 = \frac{\frac{1}{2}\beta_T^{-2} + \langle V+px; V+px\rangle}{\beta_T\left(\langle x;x\rangle\langle V;V\rangle - \langle x;V\rangle^2\right) + \frac{1}{2}\beta_T^{-1}\langle x;x\rangle}$$

❑ H. Spohn, ***arXiv 1305.6412*** (2012)

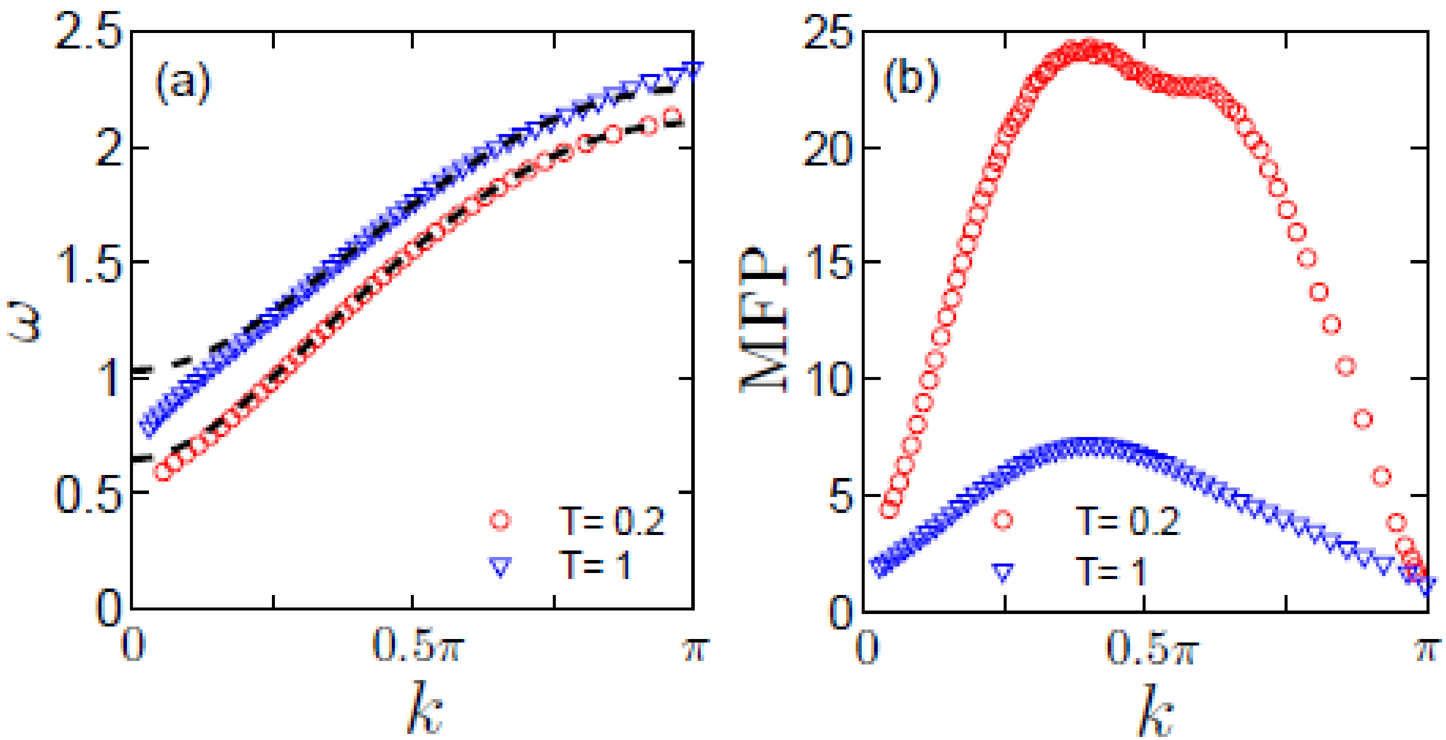
FPU-$\alpha\beta$: Effective MFP

$$\ell_{\rm eff}(\omega) := \int_1^\infty dn|\chi_n(\omega)|/|\chi_1(\omega)|$$



Phi4 model

Very similar to the FPU-β model: both the anhamronic phonon and MFP can be defined.



Dashed line: effective phonon theory $\omega = \sqrt{4\sin^2\frac{k}{2} + \sigma}$ with $\sigma = \sum_{i=1}^{N}\langle x_i^4\rangle / \sum_{i=1}^{N}\langle x_i^2\rangle$

Take home message II:

- A driving force method has been introduced to trigger waves in nonlinear lattices, which can assist to study the properties of (anharmonic) phonons in nonlinear lattices

- For a harmonic lattice the generated wave possesses the intrinsic phonon properties

- Linear response formula for the excited wave in anharmonic are derived, which enables fast calculation of the excited motions

- Numerical simulations have been performed on the FPU-β, FPU-$\alpha\beta$ lattice and Phi4 lattice.

A QUESTION ?