Green-Kubo formula for open systems

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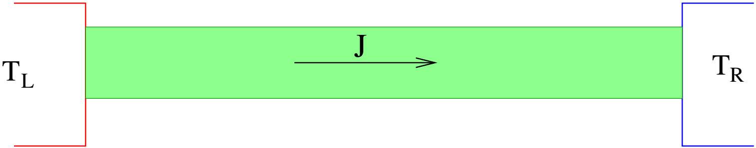
Outline

- Introduction to the problem.
  - Green-Kubo formula for thermal conductivity.
  - Anomalous heat conduction in low-dimensional systems.
  - Conductance for open systems: Landauer formula, Fluctuation theorem.
- Exact linear response formula for conductance.
- Frequency dependent conductance.
- Summary.

Classical treatment.

Response to small temperature difference



For small $\Delta T = T_L - T_R$ and system size L :

$$\text{Fourier's law implies}: \quad j \sim \kappa \frac{\Delta T}{L}$$

The thermal conductivity κ is expected to be an intrinsic material property.

Computing κ

$$\kappa = \frac{j\,L}{\Delta T}$$

Computing κ is difficult since this is a **nonequilibrium** problem.

In equilibrium we know that

$$Prob[\mathbf{x},\mathbf{v}] \quad = \quad P_0(\mathbf{x},\mathbf{v}) = \frac{e^{-\beta H(\mathbf{x},\mathbf{v})}}{Z}$$

Hence we can find the expectation value of any physical observable, say $A(\mathbf{x},\mathbf{v})$. Thus

$$\langle A \rangle = \int d\mathbf{x} \int d\mathbf{v}\; A(\mathbf{x},\mathbf{v})\; P_0(\mathbf{x},\mathbf{v})\;.$$

For a system with one end at T and the other end at $T+\Delta T$, in general, one does not know $Prob(\mathbf{x},\mathbf{v})$ even in the nonequilibrium steady state. Hence difficult to find $j = \langle j(\mathbf{x},\mathbf{v})\rangle$.

Green-Kubo linear response theory

For small deviations from equilibrium caused by small changes of Hamiltonian, one can do perturbation theory.

- Start with equilibrium phase space distribution $P_0(\mathbf{x}, \mathbf{v})$ corresponding to Hamiltonian H. In this case $\langle J \rangle = 0$.
- Add perturbation (e.g electric field):

$$H_T = H + Ve^{st} \qquad V = -e \sum \phi(x_i)$$

- Solve eqn. of motion $\frac{\partial P}{\partial t} = -\{P, H_T\}$ to linear order in V to find $P_{neq} = P_{eq} + \delta P$.
- Calculate $\langle j \rangle = \int d\mathbf{x} \int d\mathbf{v}\, j(\mathbf{x}, \mathbf{v})\, \delta P$. This gives a formula for the conductivity in terms of *equilibrium* current autocorrelation functions.

Green-Kubo formula for thermal conductivity

$$\kappa = \lim_{\tau \to \infty} \lim_{L \to \infty} \frac{1}{k_B T^2 L^d} \int_0^\tau dt \, \langle \, J(t) \, J(0) \, \rangle \, ,$$

where $J(t) = \int j(\mathbf{x}, t) d\mathbf{x}$, and $j(\mathbf{x}, t)$ is the heat flux density in x -direction. L is the linear size of the system.

However.....

- For heat conduction derivation is more difficult since current is response to boundary driving forces which cannot be included in the Hamiltonian.
- Requires assumption of local thermal equilibrium. Luttinger's mechanical derivation: assumes local thermal equilibium and relation between responses to applied field and temperature gradient.
- Limit of infinite system size necessary. Order of limits important.

Heat conduction in finite systems

Clearly the usual Green-Kubo formula cannot be directly used for:
(i) small systems (e.g single molecules) and
(ii) low-dimensional systems with anomalous transport.

Here, instead of conductivity, one is interested in the actual current through the system or its conductance defined by:

$$G = \frac{J}{\Delta T} .$$

- Fourier's law is not generally valid in low-dimensional systems . κ depends on system size L.
- Necessary and sufficient conditions for validity of Fourier's law ?
  Role of anharmonicity, disorder and dimensionality.

Lepri, Livi, Politi, Phys. Rep. (2003) .
Dhar , Adv. Phys. (2008) .

One-dimensional systems with nonintegrable interactions

Momentum conserving system: FPU - model

$$H = \sum_{l=1}^{N} \frac{p_l^2}{2m} + \sum_{l=1}^{N+1} \left[k_2 \frac{(q_l - q_{l-1})^2}{2} + k_3 \frac{(q_l - q_{l-1})^3}{3} + \lambda \frac{(q_l - q_{l-1})^4}{4} \right] .$$

Momentum non-conserving system: ϕ^4 - model

$$H = \sum_{l=1}^{N} \left[\frac{p_l^2}{2m} + k_o \frac{q_l^2}{2} \right] + \sum_{l=1,N+1} k_2 \frac{(q_l - q_{l-1})^2}{2} + \sum_{l=1}^{N} \lambda \frac{q_l^4}{4} .$$

- Momentum conserving: $\kappa \sim L^{1/3}$
- Momentum nonconserving (pinned case): $\kappa \sim L^0$

Disorder + Anharmonicity: $\kappa \sim L^{\alpha}$. Computing α is a difficult problem.
Diverging conductivity leads to a problem in use of the Green-Kubo formula.

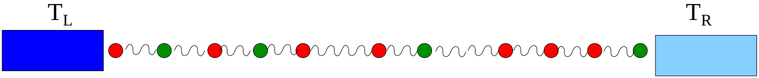
Conductance of finite systems: earlier results

- Landauer-like formula for thermal conductance. Can be derived using:
  (i) nonequilibrium Green's function (Keldysh formalism) ,
  (i) Langevin equations of motion approach,
  (iii) Green-Kubo formula by including infinite reservoirs.

  Most useful for non-interacting (e.g. harmonic) systems.

- Steady state fluctuation theorem of Gallavotti-Cohen implies a Green-Kubo-like formula. Valid for finite OPEN systems. No general proof exists.

Landauer-like formula for thermal conductance



Exact expression for nonequilibrium heat current ["Landauer-like" formula for phonons.]
In classical case:

$$G = \frac{I}{\Delta T} = \frac{k_B}{2\pi} \int_0^\infty d\omega \mathcal{T}(\omega) \ ,$$

where $\mathcal{T}(\omega)$ is the phonon transmission function.

[Casher and Lebowitz (1971), Rubin and Greer (1971), Dhar and Roy (2006)].

In quantum case:

$$I = \frac{1}{2\pi} \int_0^\infty d\omega \ \hbar\omega \mathcal{T}(\omega) \ [f(\ \omega, T_L) - f(\omega, T_R)\ ] \ .$$

Steady state fluctuation theorem

Cohen-Gallavotti SSFT: Let $\Delta\beta = 1/T_R - 1/T_L$. Consider rate of entropy production over time τ.

$$Q = \int_0^\tau dt\, j(t)\ , \qquad s = (\Delta\beta)\frac{Q}{\tau}$$
$$\frac{P(s)}{P(-s)} = \exp[s\tau] \quad \tau \to \infty$$

Let $Z(\lambda) = \langle \exp[-\lambda Q] \rangle \sim \exp[g(\lambda)\tau]$.

SSFT implies the symmetry relation: $g(\lambda) = g(\Delta\beta - \lambda)$.
Expanding both sides and comparing coefficients gives:

$$G = \lim_{\Delta T \to 0} \frac{\langle j \rangle_{\Delta T}}{\Delta T} = \frac{1}{k_B T^2} \int_0^\infty \langle j(0) j(t) \rangle_T\ .$$

(Gallavotti, Lebowitz-Spohn, Andrieux-Gaspard):

New results

For $J = \int dx j(x)$:

$$\lim_{\Delta T \to 0} \frac{\langle J \rangle_{\Delta T}}{\Delta T} = \frac{1}{k_B T^2 L} \int_0^\infty \langle J(0) J(t) \rangle_T \, .$$

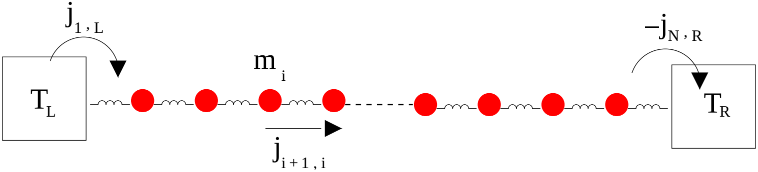
- Correlation is evaluated not with Hamiltonian dynamics but with the dynamics of the open system. Heat bath dynamics either deterministic or stochastic.
- No need of infinite size limit.

In this work:

- A general derivation of the open-system conductance formula, derived by perturbation theory, and without invoking the fluctuation theorem.
- Generalization of the result to the case of current response to time-dependent temperature perturbations.

Outline of the derivation

- Hamiltonian system in contact with heat baths: stochastic Markovian dynamics.
- Write Fokker-Planck equation for phase space distribution $P(\mathbf{x}, \mathbf{v}, t)$. Set $P = P_0 + p$ and solve for p perturbatively to $\mathcal{O}(\Delta T)$. [$P_0 = e^{-\beta H}/Z$].
- Use p to compute $\langle J \rangle_{\Delta T}$. This gives the response in terms of the correlator $\langle J(0) J_{fp}(t) \rangle$ where J_{fp} is a boundary current operator.
- Relate $\langle J(0) J_{fp}(t) \rangle$ to $\langle J(0) J_b(t) \rangle$ where J_b is an instantaneous boundary current operator.
- Use continuity equations to relate $\langle J(0) J_b(t) \rangle$ to $\langle J(0) J(t) \rangle$.



$$H = \sum_{l=1}^{N} \left[\frac{m_l v_l^2}{2} + V(x_l) \right] + \sum_{l=1}^{N-1} U(x_l - x_{l+1})$$

$$\begin{aligned} m_1 \dot{v}_1 &= f_1 - \gamma^L v_1 + \eta^L \\ m_l \dot{v}_l &= f_l \qquad\qquad l = 2, 3, ...N-1 \\ m_N \dot{v}_N &= f_N - \gamma^R v_N + \eta^R \ . \end{aligned}$$

$$\begin{aligned} f_l &= -\partial H / \partial x_l \ , \\ \langle \eta^L(t) \eta^L(t') \rangle &= 2\gamma^L k_B T_L \delta(t - t') \ , \ \ \langle \eta_R(t) \eta^R(t') \rangle = 2\gamma_R k_B T_R \delta(t - t') \ . \end{aligned}$$

Definitions

Local energy: $\epsilon_l(t)$

Continuity equation :

$$\partial\epsilon_l(t)/\partial t = j_{l,l-1} - j_{l+1,l} + \delta_{l,1} j_{1,L} + \delta_{l,N} j_{N,R} \ .$$

This defines the local current operators:

$$\begin{aligned} j_{l,l-1} &= \frac{1}{2}\,(v_{l-1} + v_l)\; f_{l,l-1} \\ j_{1,L} &= v_1\;(-\gamma_L v_1 + \eta_L) \\ j_{N,R} &= v_N\;(-\gamma_R v_N + \eta_R) \end{aligned}$$

Total current: $J = \sum_{l=1,N-1} j_{l+1,l}$

Fokker-Planck equation

$$\frac{\partial P(\mathbf{x}, \mathbf{v}, t)}{\partial t} = \hat{L}^H P(\mathbf{x}, \mathbf{v}, t) + \hat{L}^B P(\mathbf{x}, \mathbf{v}, t) \; .$$

$$\hat{L}^H = -\sum_{l=1}^{N} \left(+ \frac{\partial}{\partial x_l} v_l + \frac{\partial}{\partial v_l} \frac{f_l}{m_l} \right)$$

$$\hat{L}^B = \frac{\gamma^L}{m_1} \frac{\partial}{\partial v_1} \left(v_1 + \frac{k_B T_L}{m_1} \frac{\partial}{\partial v_1} \right) + \frac{\gamma^R}{m_N} \frac{\partial}{\partial v_N} \left(v_N + \frac{k_B T_R}{m_N} \frac{\partial}{\partial v_N} \right) \; .$$

Let $T = \frac{1}{2}(T_L + T_R)$ and $\Delta T = (T_L - T_R)$.

Fokker-Planck equation

$$\frac{\partial P(\mathbf{x},\mathbf{v},t)}{\partial t} = \hat{L}P(\mathbf{x},\mathbf{v},t) + \hat{L}^{\Delta T}P(\mathbf{x},\mathbf{v},t)\ .$$

$$\hat{L} = \hat{L}^H + \frac{\gamma^L}{m_1}\frac{\partial}{\partial v_1}\left(v_1 + \frac{k_B T}{m_1}\frac{\partial}{\partial v_1}\right) + \frac{\gamma^R}{m_N}\frac{\partial}{\partial v_N}\left(v_N + \frac{k_B T}{m_N}\frac{\partial}{\partial v_N}\right)\ ,$$

$$\hat{L}^{\Delta T}(\mathbf{v}) = \frac{k_B \Delta T}{2}\left(\frac{\gamma^L}{m_1^2}\frac{\partial^2}{\partial v_1^2} - \frac{\gamma^R}{m_N^2}\frac{\partial^2}{\partial v_N^2}\right)\ .$$

At time $t = -\infty$ system is in equilibrium: $P = P_0 = \exp(-\beta H)/Z$.

Solve Fokker-Planck equation perturbatively upto order ΔT.

Solution of Fokker-Planck equation

Let $P(\mathbf{x},\mathbf{v},t) = P_0(\mathbf{x},\mathbf{v}) + p(\mathbf{x},\mathbf{v},t)$.

$$p(\mathbf{x},\mathbf{v},t) = \int_{-\infty}^{t} dt'\ e^{\hat{L}(t-t')}\ \hat{L}^{\Delta T}(\mathbf{v})\ P_0(\mathbf{x},\mathbf{v})$$
$$= \Delta\beta \int_{-\infty}^{t} dt'\ e^{\hat{L}(t-t')}\ J_{fp}(\mathbf{v})\ P_0(\mathbf{x},\mathbf{v})\ ,$$

with $J_{fp}(\mathbf{v}) = -\frac{\gamma^L}{2}\left[v_1^2 - \frac{k_B T}{m_1}\right] + \frac{\gamma^R}{2}\left[v_N^2 - \frac{k_B T}{m_N}\right]$.

Note that:

- $\langle j_{1,L}\rangle = \langle v_1(-\gamma_L v_1 + \eta_L)\rangle = -\gamma_L\ (v_1^2 - k_B T_L/m_1)$.
- $J_{fp} = (\Delta\beta\ P_0)^{-1}\hat{L}^{\Delta T} P_0 = \Delta\beta\left[\frac{1}{P}\frac{\partial P}{\partial t}\right]_{P=P_0}$.

Finding $\langle J \rangle_{\Delta T}$

The expectation value of total current is given by:

$$\begin{aligned}
\langle J \rangle_{\Delta T} &= \int d\mathbf{x} d\mathbf{v}\, J\, p(\mathbf{x}, \mathbf{v}) \\
&= \Delta\beta \int_0^\infty dt \int d\mathbf{x} d\mathbf{v}\, J\, e^{\hat{L}t}\, J_{fp}\, P_0 \\
&= \Delta\beta \int_0^\infty dt\, \langle J(t) J_{fp}(0) \rangle_T \\
&= -\Delta\beta \int_0^\infty dt\, \langle J(0) J_{fp}(t) \rangle_T \quad \text{(using detailed balance principle)}\ .
\end{aligned}$$

J_{fp} does not have any obvious physical interpretation.
We massage this to get an expression in terms of $\langle J(t) J(0) \rangle$.

Relating $\langle JJ_{fp} \rangle$ to $\langle JJ_b \rangle$

Definition:

$$
\begin{aligned}
J_b(t) &= \frac{1}{2}(j_{1,L} - j_{N,R}) \\
&= \frac{1}{2}[-\gamma^L v_1^2(t) + \eta^L(t) v_1(t)\,] - \frac{1}{2}[-\gamma^R v_N^2(t) + \eta^R(t) v_N(t)]\,. \\
\text{Recall}: & \\
J_{fp}(\mathbf{v}) &= \frac{\gamma^L}{2}\left[-v_1^2 + \frac{k_B T}{m_1}\right] - \frac{\gamma^R}{2}\left[-v_N^2 + \frac{k_B T}{m_N}\right]\,.
\end{aligned}
$$

We can prove:

$$\langle J(0) J_b(t) \rangle = \langle J(0) J_{fp}(t) \rangle$$

Relating $\langle JJ_b \rangle$ to $\langle JJ \rangle$

Final step of proof:

$$\int_0^\infty dt \langle J(0) J_b(t) \rangle = \frac{1}{N-1} \int_0^\infty dt \langle J(t) J(0) \rangle \ .$$

Define $A(t) = \sum_{k=1}^{l} \epsilon_k - \sum_{k=l+1}^{N} \epsilon_k$ for any $l = 1, 2, ...N-1$.

From continuity equation $J_b - j_{l+1,l}(t) = \frac{1}{2} \frac{dA}{dt}$.

Multiplying by $J(0)$, taking $\langle ... \rangle$ and integrating from $t = 0$ to ∞ gives:

$$\int_0^\infty dt \langle \ J(0) \ J_b(t) \ \rangle = \int_0^\infty dt \langle \ J(0) \ j_{l+1,l}(t) \ \rangle \ .$$

Used: $\langle A(0) J(0) \rangle = 0$, $\langle A(\infty) J(0) \rangle = 0$.

Summing over l gives required result.

Final result

$$
\begin{aligned}
\langle J \rangle_{\Delta T} &= \Delta\beta \int_0^\infty dt \, \langle J(t) J_{fp}(0) \rangle && \text{Perturbation theory} \\
&= -\Delta\beta \int_0^\infty dt \, \langle J(0) J_{fp}(t) \rangle && \text{Detailedbalance} \\
&= -\Delta\beta \int_0^\infty dt \, \langle J(0) \, J_b(t) \rangle && \text{Novikov theorem} \\
&= \frac{1}{k_B T^2 (N-1)} \int_0^\infty dt \, \langle J(t) J(0) \rangle \, . && \text{current conservation}
\end{aligned}
$$

Equivalently....let $j = J/(N-1)$.

$$
G = \lim_{\Delta T \to 0} \frac{\langle j \rangle_{\Delta T}}{\Delta T} = \frac{1}{k_B T^2} \int_0^\infty dt \langle j(t) j(0) \rangle \, .
$$

Comments

- Easy to generalize above proof to lattice models in arbitrary dimensions.
- Result valid for fluid system coupled to Maxwell baths.
- Also proved above result for Nose-Hoover baths and for an exponentially correlated stochastic bath.
- For harmonic chain we recover Landauer-like formula in terms of transmission.

Generalization to finite frequencies

Consider the case where the temperature at the two ends are oscillating in time:

$$T_L(t) = T + \frac{\Delta T(t)}{2} \qquad T_R(t) = T - \frac{\Delta T(t)}{2} \quad .$$

How does the system respond ?

Consider $\Delta T(t)) = \Delta T(\Omega) e^{i\Omega t}$.

The system will then eventually reach a time-dependent steady state where physical observables like temperature, current vary periodically with time. For current we expect, for small ΔT:

$$j_l(t) = G_l(\Omega)\, \Delta T(\Omega)\, e^{i\Omega t} \ .$$

Can use perturbation theory to find $G_l(\Omega)$.

Finite frequency conductance

Repeating the zero-frequency calculation gives:

$$\frac{\langle j_{l+1,l}(\Omega)\rangle}{\Delta T(\Omega)e^{i\Omega t}} = G_l(\Omega) = -\frac{1}{k_B T^2}\int_0^\infty \langle j_{l+1,l}(\tau)J_{fp}(0)\rangle e^{-i\Omega\tau}\,d\tau \ ,$$

In this case it is not possible to obtain a result in terms of $\langle j_{l+1,l}(t)j_{l+1,l}(0)\rangle$ or $\langle J(t)J(0)\rangle$.

Hence this cannot be related to the finite frequency response obtained from standard Green-Kubo theory.

[see B.S Shastry, Rep. Prog. Phys. (2008)].

Harmonic chain: Exact result

In this case the response function can be computed exactly and is given by:

$$G_l(\Omega) = \frac{1}{4\pi}\int_{-\infty}^{\infty} d\omega\ \omega\Big[\gamma_L\{\mathcal{G}^+_{l,1}(\omega-\Omega) - \mathcal{G}^+_{l+1,1}(\omega-\Omega)\}\ \{\mathcal{G}^+_{l,1}(-\omega) + \mathcal{G}^+_{l+1,1}(-\omega)\}$$
$$-\gamma_R\{\mathcal{G}^+_{l,N}(\omega-\Omega) - \mathcal{G}^+_{l+1,N}(\omega-\Omega)\}\ \{\mathcal{G}^+_{l,N}(-\omega) + \mathcal{G}^+_{l+1,N}(-\omega)\}\Big]$$

where

$$\mathcal{G}^+(\omega) = [-M\omega^2 + \Phi - \Sigma^+(\omega)]^{-1}$$

is the phonon Green's function and Σ^+ the self-energy correction due to baths. For DC case ($\Omega = 0$) this gives the Landauer result:

$$G = \frac{1}{2\pi}\int_0^{\infty} d\omega\ \mathcal{T}(\omega)$$

with $\mathcal{T} = 4\,\gamma_L\,\gamma_R\,\omega^2\,|\mathcal{G}^+_{1N}|^2$.

Discussion

- Exact Green-Kubo like expression for the linear response conductance in a system connected to heat baths.
- Results valid in arbitrary dimensions and sizes. Derived both for lattice and fluid models
- Various bath models have been considered. Markovian, non-Markovian and deterministic.
- Differences with the usual Green-Kubo formula:

  (i)No need to first take limit of infinite system size. Result valid for finite systems.
  (ii)Correlation function has to be evaluated *not* with Hamiltonian dynamics, but for an open system evolving with heat bath dynamics.

Discussion

- (iii) Assumption of local thermal equilibrium is not necessary.
- Likely that for systems with normal transport our formula will reduce to usual formula. Proof? For systems with anomalous transport (low dimensions), the present formula has to be used. Form of correlation functions very different. Boundary conditions important.
- Extension to finite frequency transport. Does not seem to be related to the standard Green-Kubo formula.
- Open problems
  - Extension of steady state fluctuation theorem.
  - Quantum systems

References:
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LAST DATE: December 15, 2010.

One dimensional disordered harmonic chain

- Almost all normal modes of the chain are localized and their amplitude at the boundaries is exponentially small (in L) leading to transmission decaying exponentially.
- Low frequency modes are extended and transmit energy.
- No Fourier's law: Strong boundary condition dependence. $\kappa \sim L^{1/2}, \quad L^{-1/2}$.
- Heat insulator in pinned case.