Green-Kubo formula for heat conduction in open systems

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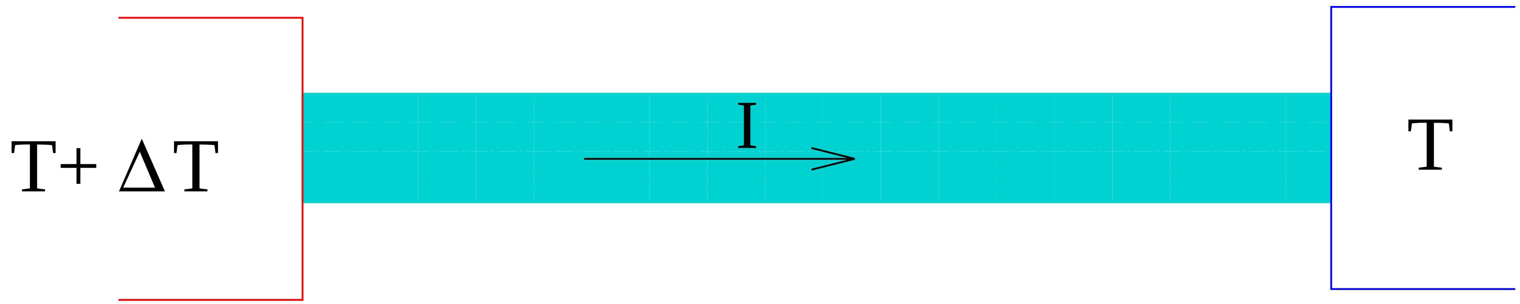
22^{nd} April, 2010.

Response to small temperature difference



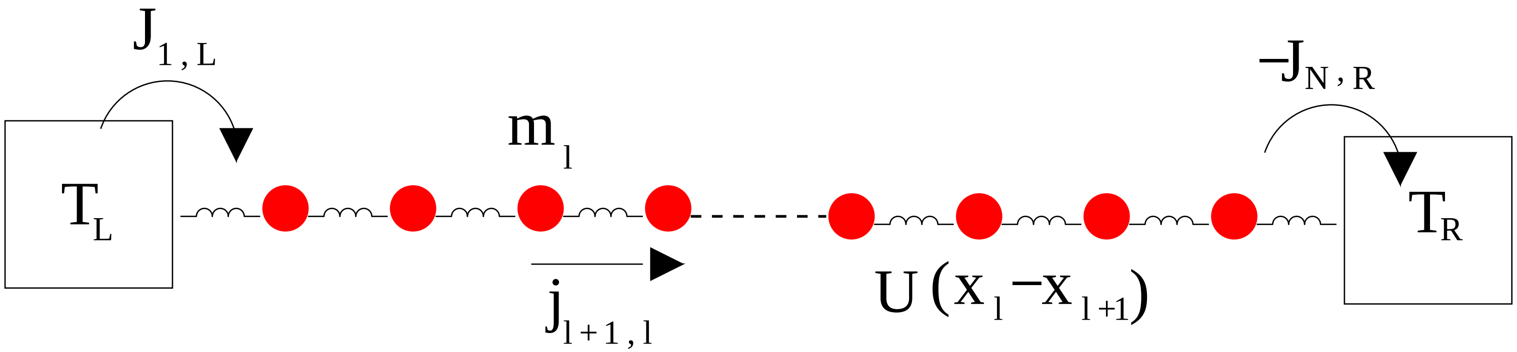
$$I = G\Delta T$$

G is the conductance of the system. For macroscopic systems it is useful to define the conductivity $\kappa = \frac{GL}{A} = \frac{IL}{A\Delta T}$.

(L = length of system, A =cross-sectional area.)

κ expected to be an intrinsic property of the system and is given by linear response theory:
Response of a system to small perturbations applied on the system is related to the equilibrium time correlation function.

One dimensional lattice Hamiltonian: Langevin reservoirs



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

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

Definitions

$$f_l = -\frac{\partial H}{\partial x_l}$$

$$\langle \eta^L(t)\eta^L(t')\rangle = 2\gamma^L k_B T_L \delta(t-t')$$

$$\langle \eta^R(t)\eta^R(t')\rangle = 2\gamma^R k_B T_R \delta(t-t')$$

Local energy: $\epsilon_l = \frac{m_l v_l^2}{2} + V(x_l) + \frac{1}{2}[U(x_l - x_{l-1}) + U(x_{l+1} - x_l)]$.
Continuity equation:

$$\frac{d\epsilon_l}{dt} + j_{l+1,l} - j_{l,l-1} = j_{1,L}\,\delta_{l,1} + j_{N,R}\,\delta_{l,N}$$

$$\text{where } j_{l+1,l} = \frac{1}{2}(v_l + v_{l+1}) f_{l+1,l}$$

$$\text{and } j_{1,L}(t) = -\gamma^L v_1^2(t) + \eta^L(t) v_1(t)$$

$$j_{N,R}(t) = -\gamma^R v_N^2(t) + \eta^R(t) v_N(t)$$

$$f_{l+1,l} = \partial_{x_{l+1}} U(|x_l - x_{l+1}|)$$

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

Green Kubo formula for thermal conductivity

For systems with Hamiltonian dynamics, the response to small applied fields is related to equilibrium correlation function.

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

N is the size of the system.

- For non-mechanical disturbances there is no external Hamiltonian.
- Derivations are not rigorous : assumes
  - Local thermal equilibrium (LTE).(Visscher, McLennan, Martin)
  - Luttinger's mechanical derivation: Fictitious "gravitational field" .
- Limit of infinite system size necessary. Order of limits important.

Heat conduction in finite system

Hence the usual Green-Kubo formula cannot be directly used for small systems and systems with anomalous transport.

Conductance of finite systems: earlier results.

- Include infinite reservoirs connected to finite systems. Get results identical to Landauer and nonequilibrium Green's function approach. Proved for non-interacting (e.g. harmonic) systems. (Allen and Ford -1968)
- Steady state fluctuation theorem implies the Green-Kubo formula. Valid for finite OPEN systems. Proved for anharmonic lattices connected to Markovian stochastic baths.

Fokker-Planck equation

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

where $\hat{L}^H(\mathbf{x},\mathbf{v}) = -\sum_l [\, v_l\, \partial/\partial x_l + (f_l/m_l)\, \partial/\partial v_l \,]$

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

Let $\quad T = \frac{1}{2}(T_L + T_R)$

and $\quad \Delta T = (T_L - T_R)$

Fokker-Planck equation

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

$$\text{where} \quad \hat{L}(\mathbf{x},\mathbf{v}) = \hat{L}^{H} + \frac{\gamma^L}{m_1}\frac{\partial}{\partial v_1}\left(v_1 + \frac{k_B T}{m_1}\frac{\partial}{\partial v_1}\right)$$

$$+ \frac{\gamma^R}{m_N}\frac{\partial}{\partial v_N}\left(v_N + \frac{k_B T}{m_N}\frac{\partial}{\partial v_N}\right)$$

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

At time $t = -\infty$ the system was in thermal equilibrium:
$P_0 = \frac{e^{-\beta H}}{Z}$.
Solve Fokker-Planck equation perturbatively upto order ΔT.

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

Solution of Fokker-Planck equation

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

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

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

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

Note that

$$J_{fp}(\mathbf{v}) = (\Delta\beta \ P_0)^{-1}\hat{L}^{\Delta T}P_0 = [(\Delta\beta \ P)^{-1}\frac{\partial P}{\partial t}]_{P=P_0}$$

Finding $\langle J_{\Delta T} \rangle$

The expectation value of the total current is given by

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

J_{fp} does not have any obvious physical interpretation.

We need an expression in terms of $\langle J(t) J(0) \rangle$.

Outline of our derivation

- Wrote equation for phase space distribution $P(\mathbf{x}, \mathbf{v}, t)$. Find $\mathcal{O}(\Delta T)$ correction to $P_0 = \frac{e^{-\beta H}}{Z}$. Expressed $\langle J \rangle_{\Delta T}$ in terms of $\langle J(t) J_{fp}(0) \rangle$. J_{fp} is specified current operator.

- Use continuity equation to relate $\langle J(0) J(t) \rangle$ and $\langle J(0) J_b(t) \rangle$. $J_b(t)$ is an instanteneous current operator depending on heat flux from baths .

- Relate $\langle J(0) J_b(t) \rangle$ to $\langle J(0) J_{fp}(t) \rangle$ and then, using detailed balance to $\langle J(t) J_{fp}(0) \rangle$.

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

Definition:

$$J_b(t) = \frac{1}{2}(j_{1,L} - j_{N,R})$$
$$= \frac{1}{2}[-\gamma^L v_1^2(t) + \eta^L(t)v_1(t) - (-\gamma^R v_N^2(t) + \eta^R(t)v_N(t))]\ .$$

We prove:

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

Recall:

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

Relating $\langle J J_{fp} \rangle$ to $\langle J J_b \rangle$

Proof:

$$\langle J(0)\eta^L(t)v_1(t)\rangle = \langle J(0)\eta^R(t)v_N(t)\rangle = 0 \ .$$

This is proved using Novikov's theorem.

Also $\langle JT \rangle = 0$. Thus $\langle J(0)J_b(t)\rangle = \langle J(0)J_{fp}(t)\rangle$.

But we need $J(t)J_{fp}(0)$. Use detailed balance

Novikov's theorem:

$$< \eta_i(t)H[\eta] > \ = \ \sum_j \int \langle \eta_i(t)\eta_j(t')\rangle \left\langle \frac{\delta H[\eta]}{\delta \eta_j(t')} \right\rangle dt' \ ,$$

where $\delta H[\eta]/\delta\eta_j(t')$ represents a functional derivative of $H[\eta]$ with respect to η.

Relating $\langle J J_{fp} \rangle$ to $\langle J J_b \rangle$

Let $W(q,t|q',0)$ denote the transition probability from $q' = (\mathbf{x}', \mathbf{v}')$ to $q = (\mathbf{x}, \mathbf{v})$ in time t.

$$
\begin{aligned}
&\langle J_{fp}(t) J(0) \rangle \\
&= \int dq dq' J_{fp}(\mathbf{v}) J(\mathbf{x}', \mathbf{v}') P_0(\mathbf{x}', \mathbf{v}') W(\mathbf{x}, \mathbf{v}, t|\mathbf{x}', \mathbf{v}', 0) \\
&= \int dq dq' J_{fp}(\mathbf{v}) J(\mathbf{x}', \mathbf{v}') P_0(\mathbf{x}', -\mathbf{v}') W(\mathbf{x}', -\mathbf{v}', t|\mathbf{x}, -\mathbf{v}, 0) \\
&= \int dq dq' J_{fp}(-\mathbf{v}) J(\mathbf{x}', -\mathbf{v}') P_0(\mathbf{x}', \mathbf{v}') W(\mathbf{x}', \mathbf{v}', t|\mathbf{x}, \mathbf{v}, 0) \\
&= -\int dq dq' J_{fp}(\mathbf{v}) J(\mathbf{x}', \mathbf{v}') P_0(\mathbf{x}', \mathbf{v}') W(\mathbf{x}', \mathbf{v}', t|\mathbf{x}, \mathbf{v}, 0) \\
&= -\langle J(t) J_{fp}(0) \rangle.
\end{aligned}
$$

used: (i)detailed balance principle
(ii)J is odd in velocities, J_{fp} is even.

Relating $\langle J J_b \rangle$ to $\langle J J \rangle$

Final step of the proof:

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

Define $D_l(t) = \sum_{k=1}^{l} \epsilon_k - \sum_{k=l+1}^{N} \epsilon_k$ for $l = 1, 2, ...N-1$. From continuity equation $dD_l/dt = -2j_{l+1,l}(t) + 2J_b(t)$. Define $A(t) = \sum_{l=1}^{N-1} D_l$. Then

$$2J(t) - 2(N-1)J_b(t) = -\frac{dA}{dt}$$

Multiplying $J(0)$, taking $\langle ... \rangle$ and integrating from $t=0$ to ∞ gives above result. Use: $\langle A(0) J(0) \rangle = 0$ and $\langle A(\infty) J(0) \rangle = 0$.

Final result

$$\langle J_{\Delta T} \rangle = \Delta\beta \int_0^\infty dt \langle J(t) J_{fp}(0) \rangle$$

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

Define Current: $\bar{j} = J/(N-1)$.

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

One dimensional fluid system coupled to Maxwell baths

Smooth interaction potential
Maxwell baths: End particles (1 and N) interact with baths: $\Pi(v) = m_1 \, \beta_L \, \theta(v) \, v \, \exp[-\beta_L m_1 v^2/2]$.
After a small time interval ϵ, the phase space density $P(\mathbf{x};\mathbf{v};t+\epsilon)$ is

$$\begin{aligned} P(\mathbf{x};\mathbf{v};t+\epsilon) &= \beta_L m_1 e^{-\frac{1}{2}\beta_L m_1 v_1^2} \int_0^\infty P(0,\mathbf{x}'-\mathbf{v}'\epsilon;-v_0,\mathbf{v}'-\mathbf{a}'\epsilon;t) v_0 dv_0 \\ & \qquad \text{for } x_1 < v_1\epsilon \\ &= \beta_R m_N e^{-\frac{1}{2}\beta_R m_N v_N^2} \int_0^\infty P(\mathbf{x}'-\mathbf{v}'\epsilon, L;\mathbf{v}'-\mathbf{a}'\epsilon, v_0;t) v_0 dv_0 \\ & \qquad \text{for } x_N > L + v_N\epsilon \\ & = P(\mathbf{x}-\mathbf{v}\epsilon, \mathbf{v}-\mathbf{a}\epsilon, t) \qquad \text{otherwise} \end{aligned}$$

and $J_{fp} = -\frac{1}{2}(\frac{1}{2}m_1 v_1^2 - k_B T) v_1 \delta(x_1)\theta(v_1)$
$-\frac{1}{2}(\frac{1}{2}m_N v_N^2 - k_B T) v_N \delta(x_N - L)\theta(-v_N)$.

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

$$
\begin{aligned}
j_b &= \frac{1}{2}(j_{1L} - j_{NR}) \\
j_{1,L} &= -\frac{1}{2}m_1 v_1 (v_{1,L}^2 - v_1^2)\,\delta(x_1)\theta(-v_1) \\
j_{N,R} &= \frac{1}{2}m_N v_N (v_{N,L}^2 - v_L^2)\,\delta(x_N - L)\theta(v_N)\,.
\end{aligned}
$$

Transition rate satisfies detailed balance:

$$
\begin{aligned}
W(\mathbf{x},\mathbf{v},t|\mathbf{x}_0,\mathbf{v}_0,t) &= \beta m_1 e^{-\frac{1}{2}m_1 v_1^2} v_1 \theta(x_1)\theta(x_{01})\theta(v_1)\theta(-v_{01}) \\
&= W(\mathbf{x}_0,-\mathbf{v}_0,t|\mathbf{x},-\mathbf{v},t)
\end{aligned}
$$

Finally we get:

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

Other systems

- Easy to generalize above proof to lattice models in arbitrary dimensions.
- Easy to generalize to fluid systems in higher dimensions.
- Also proved above result for Nose-Hoover baths and for an exponentially correlated stochastic bath.
- Nose-Hoover:

$$\dot{\zeta}_L = (\beta_L m_1 v_1^2 - 1)/\theta_L \quad \dot{\zeta}_R = (\beta_R m_N v_N^2 - 1)/\theta_R \ .$$

- Exponentially correlated bath:

$$\dot{y}_L = -y_L/(\nu^L \gamma^L) - \gamma^L v_1 + \eta^L \quad \dot{y}_R = -y_R/(\nu^R \gamma^R) - \gamma^R v_N + \eta^R \ . \quad (1)$$

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-Markovin 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. Equilibration in absence of bias 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.
- Quantum systems