separating flour from bran

how to optimally estimate transport coefficients from short equilibrium molecular-dynamics simulations

Stefano Baroni
Scuola Internazionale Superiore di Studi Avanzati
Trieste — Italy

virtual talk given at the IPAM workshop on L*arge-Scale Certified Numerical Methods in Quantum Mechanics*, May 6, 2022

the linear-response theory of transport

$$\boldsymbol{J} = \sigma \boldsymbol{F}$$

the linear-response theory of transport

$$\boldsymbol{J} = \sigma \boldsymbol{F}$$

charge transport

$$\boldsymbol{J}_{\mathcal{Q}} = \sum_{I} q_I \boldsymbol{V}_I$$

$$\boldsymbol{F}_{\mathcal{Q}} = -\nabla \phi$$

$\sigma =$ electric conductivity

the linear-response theory of transport

$$\boldsymbol{J} = \sigma \boldsymbol{F}$$

charge transport

$$\boldsymbol{J}_{\mathcal{Q}} = \sum_I q_I \boldsymbol{V}_I$$

$$\boldsymbol{F}_{\mathcal{Q}} = -\nabla \phi$$

$\sigma =$ electric conductivity

energy transport

$$\boldsymbol{J}_{\mathcal{E}} = \sum_I e_I \boldsymbol{V}_I + \frac{1}{2} \sum_{I \neq J} (\boldsymbol{V}_I \cdot \boldsymbol{F}_{IJ})(\boldsymbol{R}_I - \boldsymbol{R}_J)$$

$$\boldsymbol{F}_{\mathcal{E}} = -\nabla T$$

$\sigma =$ thermal conductivity

the linear-response theory of transport

$$\boldsymbol{J} = \sigma \boldsymbol{F}$$

charge transport

$$\boldsymbol{J}_{\mathcal{Q}} = \sum_I q_I \boldsymbol{V}_I$$

$$\boldsymbol{F}_{\mathcal{Q}} = -\nabla \phi$$

σ = electric conductivity

energy transport

$$\boldsymbol{J}_{\mathcal{E}} = \sum_I e_I \boldsymbol{V}_I + \frac{1}{2} \sum_{I \neq J} (\boldsymbol{V}_I \cdot \boldsymbol{F}_{IJ})(\boldsymbol{R}_I - \boldsymbol{R}_J)$$

$$\boldsymbol{F}_{\mathcal{E}} = -\nabla T$$

σ = thermal conductivity

$\sum_I e_I = E$

$-\frac{\partial e_I}{\partial \boldsymbol{R}_J}$

the linear-response theory of transport

$$\boldsymbol{J} = \sigma \boldsymbol{F}$$

charge transport

$$\boldsymbol{J}_{\mathcal{Q}} = \sum_I q_I \boldsymbol{V}_I$$

$$\boldsymbol{F}_{\mathcal{Q}} = -\nabla\phi$$

σ = electric conductivity

energy transport

$$\boldsymbol{J}_{\mathcal{E}} = \sum_I e_I \boldsymbol{V}_I + \frac{1}{2}\sum_{I\neq J}(\boldsymbol{V}_I \cdot \boldsymbol{F}_{IJ})(\boldsymbol{R}_I - \boldsymbol{R}_J)$$

$$\boldsymbol{F}_{\mathcal{E}} = -\nabla T$$

σ = thermal conductivity

$$\sigma \propto \int_0^\infty \langle \boldsymbol{J}(t)\boldsymbol{J}(0)\rangle dt$$

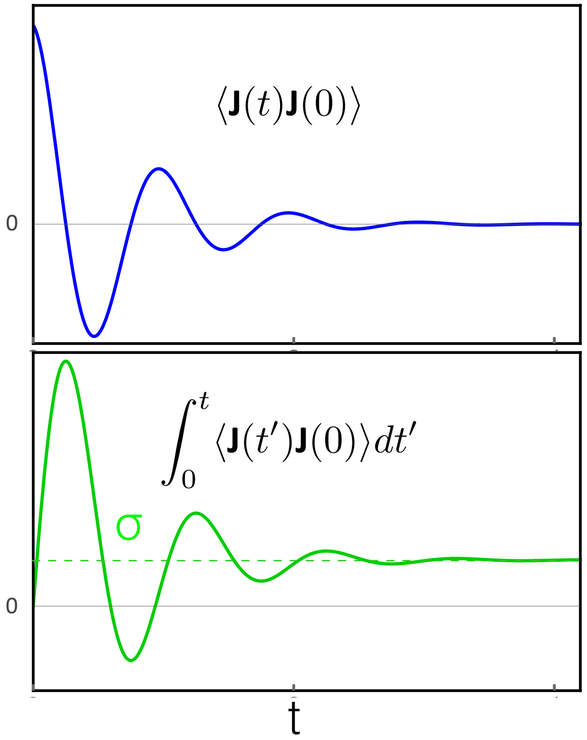
Green-Kubo

the linear-response theory of transport

$$\boldsymbol{J} = \lambda \boldsymbol{F}$$

Green-Kubo

$$\sigma \propto \underbrace{\int_0^\infty \langle \boldsymbol{J}(t)\boldsymbol{J}(0)\rangle dt}_{\langle \boldsymbol{J}^2\rangle \tau}$$

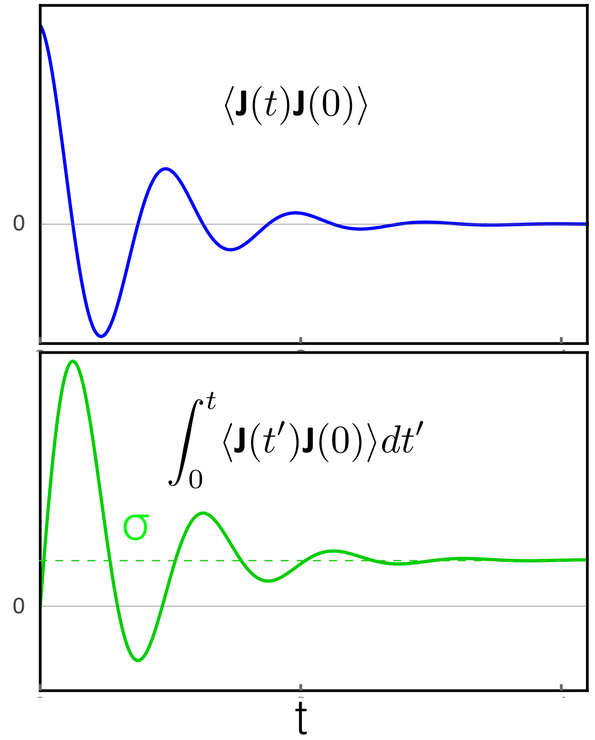


the linear-response theory of transport

$$\boldsymbol{J} = \lambda \boldsymbol{F}$$

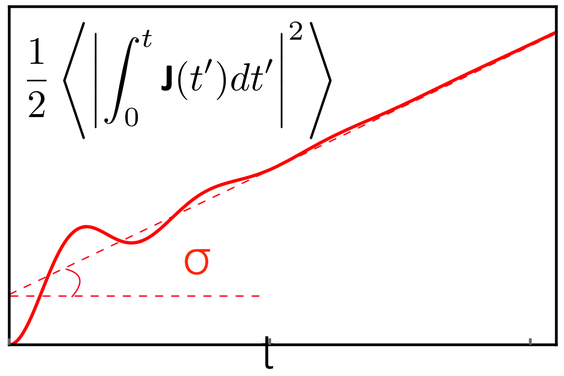
Green-Kubo

$$\sigma \propto \underbrace{\int_0^\infty \langle \boldsymbol{J}(t)\boldsymbol{J}(0)\rangle dt}_{\langle \boldsymbol{J}^2\rangle \tau}$$



Einstein-Helfand

$$\sigma \propto \lim_{t\to\infty} \frac{1}{2t} \mathrm{var}\left[\int_0^t \boldsymbol{J}(t')dt'\right]$$



hurdles toward an ab initio Green-Kubo theory

$$\boldsymbol{J}_{\mathcal{E}} = \sum_I e_I \boldsymbol{V}_I + \frac{1}{2}\sum_{I\neq J}(\boldsymbol{V}_I \cdot \boldsymbol{F}_{IJ})(\boldsymbol{R}_I - \boldsymbol{R}_J)$$

PRL **104,** 208501 (2010) PHYSICAL REVIEW LETTERS week ending 21 MAY 2010

Thermal Conductivity of Periclase (MgO) from First Principles

Stephen Stackhouse*
Department of Geological Sciences, University of Michigan, Ann Arbor, Michigan, 48109-1005, USA

Lars Stixrude†
Department of Earth Sciences, University College London, Gower Street, London WC1E 6BT, United Kingdom

Bijaya B. Karki‡
Department of Computer Science, Louisiana State University, Baton Rouge, Louisiana 70803, USA
and Department of Geology and Geophysics, Louisiana State University, Baton Rouge, Louisiana 70803, USA

sensitive to the form of the potential. The widely used Green-Kubo relation [14] does not serve our purposes, because in first-principles calculations it is impossible to uniquely decompose the total energy into individual contributions from each atom.

hurdles toward an ab initio Green-Kubo theory

$$\boldsymbol{J}_{\mathcal{E}} = \sum_{I} e_I \boldsymbol{V}_I + \frac{1}{2} \sum_{I \neq J} (\boldsymbol{V}_I \cdot \boldsymbol{F}_{IJ})(\boldsymbol{R}_I - \boldsymbol{R}_J)$$

PRL **104,** 208501 (2010) PHYSICAL REVIEW LETTERS week ending 21 MAY 2010

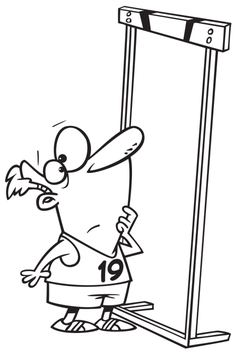
Thermal Conductivity of Periclase (MgO) from First Principles

Stephen Stackhouse*
Department of Geological Sciences, University of Michigan, Ann Arbor, Michigan, 48109-1005, USA

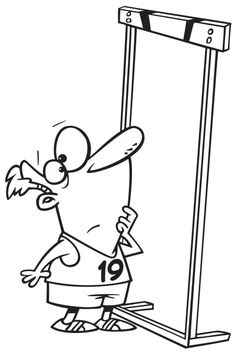
Lars Stixrude†
Department of Earth Sciences, University College London, Gower Street, London WC1E 6BT, United Kingdom

Bijaya B. Karki‡
Department of Computer Science, Louisiana State University, Baton Rouge, Louisiana 70803, USA
and Department of Geology and Geophysics, Louisiana State University, Baton Rouge, Louisiana 70803, USA

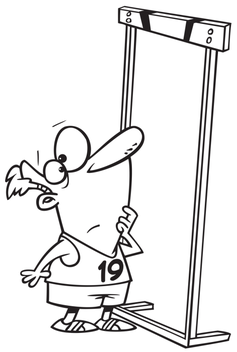
sensitive to the form of the potential. The widely used Green-Kubo relation [14] does not serve our purposes, because in first-principles calculations it is impossible to uniquely decompose the total energy into individual contributions from each atom.



how come?



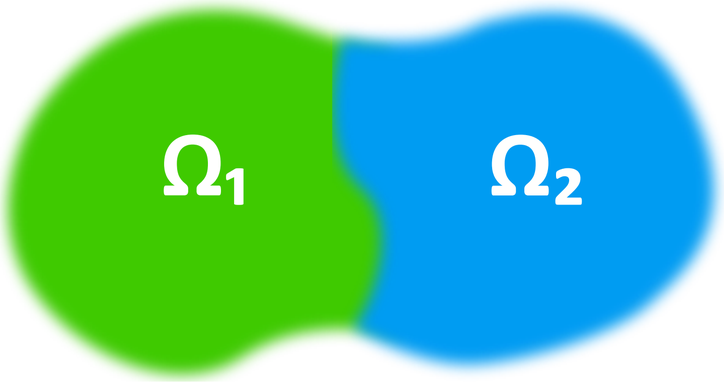
how come?



how is it that a formally exact theory of
the electronic ground state cannot predict
all measurable adiabatic properties?

gauge invariance of transport coefficients

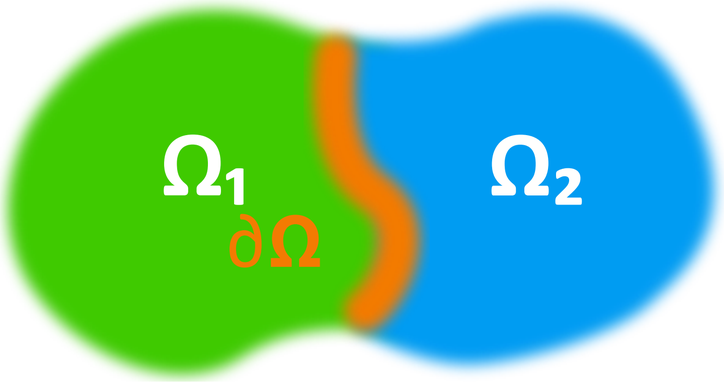
energy is extensive



$$E[\Omega_1 \cup \Omega_2] = E[\Omega_1] + E[\Omega_2]$$

gauge invariance of transport coefficients

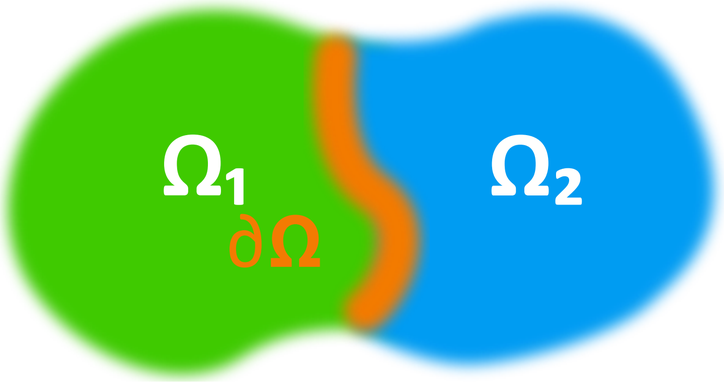
energy is extensive



$$E[\Omega_1 \cup \Omega_2] = E[\Omega_1] + E[\Omega_2] + W[\partial\Omega]$$

gauge invariance of transport coefficients

energy is extensive



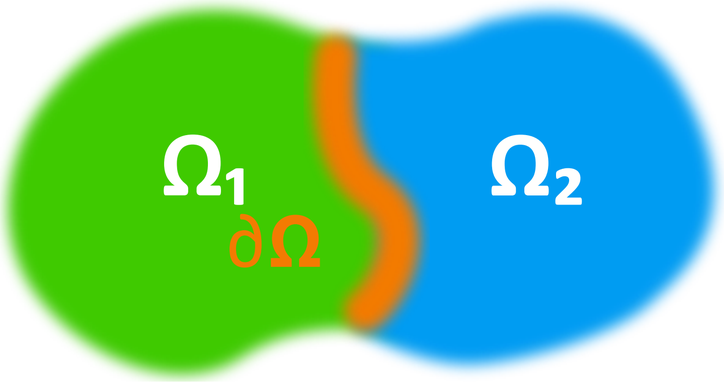
$$\mathrm{E}[\Omega_1 \cup \Omega_2] = \mathrm{E}[\Omega_1] + \mathrm{E}[\Omega_2] + \mathrm{W}[\partial\Omega]$$

$$\stackrel{?}{=} \mathcal{E}[\Omega_1] + \mathcal{E}[\Omega_2]$$

$$\mathcal{E}[\Omega] = \int_\Omega e(\mathbf{r})\, d\mathbf{r}$$

gauge invariance of transport coefficients

energy is extensive



$$\mathrm{E}[\Omega_1 \cup \Omega_2] = \mathrm{E}[\Omega_1] + \mathrm{E}[\Omega_2] + \mathrm{W}[\partial\Omega]$$

$$\stackrel{?}{=} \mathcal{E}[\Omega_1] + \mathcal{E}[\Omega_2]$$

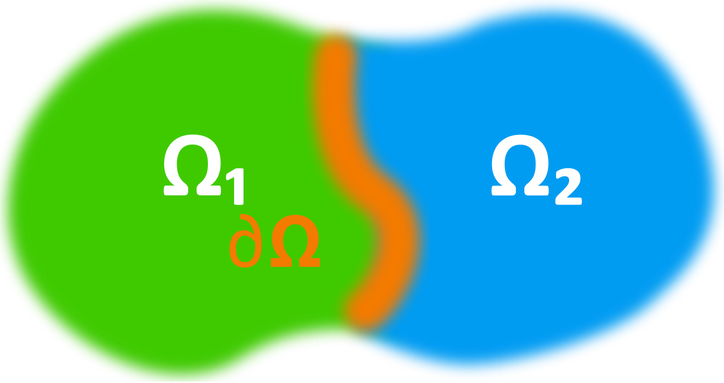
$$\mathcal{E}[\Omega] = \int_\Omega e(\mathbf{r})\, d\mathbf{r}$$

thermodynamic invariance

$$\mathcal{E}'[\Omega] = \mathcal{E}[\Omega] + \mathcal{O}[\partial\Omega]$$

gauge invariance of transport coefficients

energy is extensive



$$\mathrm{E}[\Omega_1 \cup \Omega_2] = \mathrm{E}[\Omega_1] + \mathrm{E}[\Omega_2] + \mathrm{W}[\partial\Omega]$$

$$\stackrel{?}{=} \mathcal{E}[\Omega_1] + \mathcal{E}[\Omega_2]$$

$$\mathcal{E}[\Omega] = \int_\Omega e(\mathbf{r})\, d\mathbf{r}$$

thermodynamic invariance

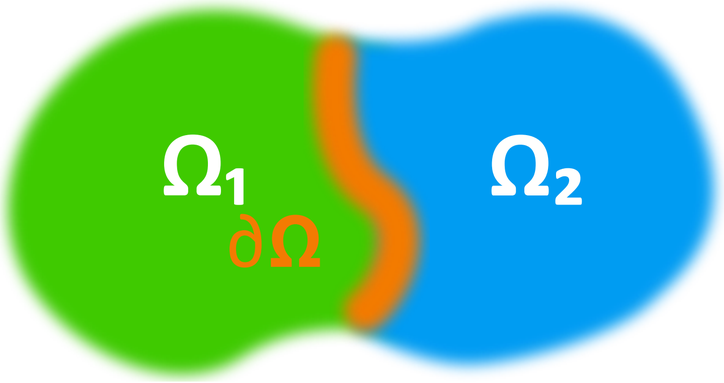
$$\mathcal{E}'[\Omega] = \mathcal{E}[\Omega] + \mathcal{O}[\partial\Omega]$$

gauge invariance

$$e'(\mathbf{r}) = e(\mathbf{r}) - \nabla \cdot \mathbf{p}(\mathbf{r})$$

gauge invariance of transport coefficients

energy is extensive



$$\mathrm{E}[\Omega_1 \cup \Omega_2] = \mathrm{E}[\Omega_1] + \mathrm{E}[\Omega_2] + \mathrm{W}[\partial\Omega]$$

$$\stackrel{?}{=} \mathcal{E}[\Omega_1] + \mathcal{E}[\Omega_2]$$

$$\mathcal{E}[\Omega] = \int_\Omega e(\mathbf{r})\, d\mathbf{r}$$

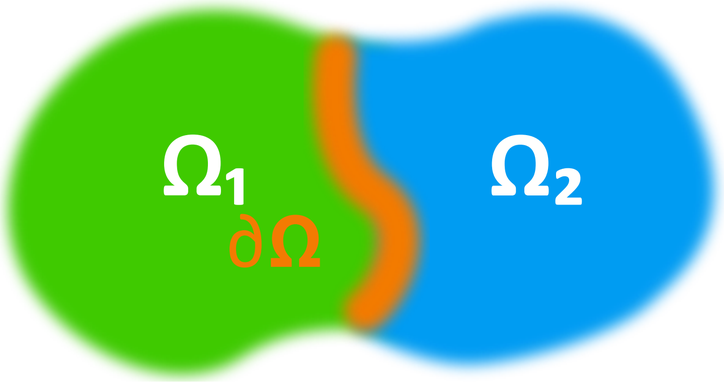
thermodynamic invariance $\quad \mathcal{E}'[\Omega] = \mathcal{E}[\Omega] + \mathcal{O}[\partial\Omega]$

gauge invariance $\quad e'(\mathbf{r}) = e(\mathbf{r}) - \nabla \cdot \mathbf{p}(\mathbf{r})$

energy conservation $\quad \dot{e}(\mathbf{r}, t) = -\nabla \cdot \mathbf{j}(\mathbf{r}, t)$

gauge invariance of transport coefficients

energy is extensive



$$\mathrm{E}[\Omega_1 \cup \Omega_2] = \mathrm{E}[\Omega_1] + \mathrm{E}[\Omega_2] + \mathrm{W}[\partial\Omega]$$

$$\stackrel{?}{=} \mathcal{E}[\Omega_1] + \mathcal{E}[\Omega_2]$$

$$\mathcal{E}[\Omega] = \int_\Omega e(\mathbf{r})\, d\mathbf{r}$$

thermodynamic invariance

$$\mathcal{E}'[\Omega] = \mathcal{E}[\Omega] + \mathcal{O}[\partial\Omega]$$

gauge invariance

$$e'(\mathbf{r}) = e(\mathbf{r}) - \nabla \cdot \mathbf{p}(\mathbf{r})$$

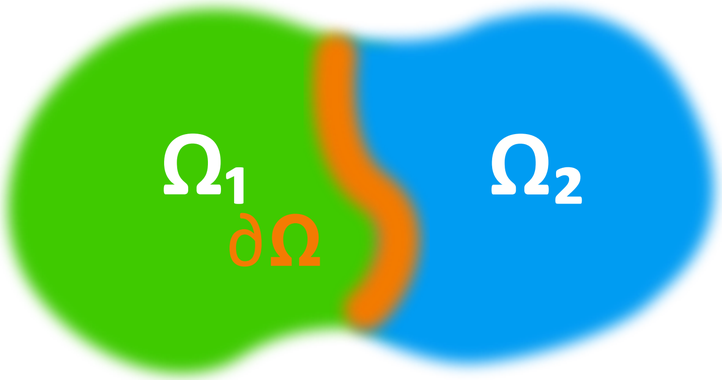
$$\mathbf{j}'(\mathbf{r}, t) = \mathbf{j}(\mathbf{r}, t) + \dot{\mathbf{p}}(\mathbf{r}, t)$$

energy conservation

$$\dot{e}(\mathbf{r}, t) = -\nabla \cdot \mathbf{j}(\mathbf{r}, t)$$

gauge invariance of transport coefficients

energy is extensive



$$\mathrm{E}[\Omega_1 \cup \Omega_2] = \mathrm{E}[\Omega_1] + \mathrm{E}[\Omega_2] + \mathrm{W}[\partial\Omega]$$

$$\stackrel{?}{=} \mathcal{E}[\Omega_1] + \mathcal{E}[\Omega_2]$$

$$\mathcal{E}[\Omega] = \int_\Omega e(\mathbf{r})\, d\mathbf{r}$$

$$\mathbf{J}(t) = \frac{1}{\Omega}\int \mathbf{j}(\mathbf{r}, t)\, d\mathbf{r}$$

$$\mathbf{P}(t) = \frac{1}{\Omega}\int \mathbf{p}(\mathbf{r}, t)\, d\mathbf{r}$$

thermodynamic invariance

$$\mathcal{E}'[\Omega] = \mathcal{E}[\Omega] + \mathcal{O}[\partial\Omega]$$

gauge invariance

$$e'(\mathbf{r}) = e(\mathbf{r}) - \nabla \cdot \mathbf{p}(\mathbf{r})$$

$$\mathbf{j}'(\mathbf{r}, t) = \mathbf{j}(\mathbf{r}, t) + \dot{\mathbf{p}}(\mathbf{r}, t)$$

$$\mathbf{J}'(t) = \mathbf{J}(t) + \dot{\mathbf{P}}(t)$$

energy conservation

$$\dot{e}(\mathbf{r}, t) = -\nabla \cdot \mathbf{j}(\mathbf{r}, t)$$

gauge invariance of transport coefficients

$$\mathbf{J}' = \mathbf{J} + \dot{\mathbf{P}}$$

gauge invariance of transport coefficients

$$\mathbf{J}' = \mathbf{J} + \dot{\mathbf{P}}$$

$$\lambda \sim \frac{1}{2t}\mathrm{var}\big[\mathbf{D}(t)\big] \qquad \mathbf{D}(t) = \int_0^t \mathbf{J}(t')dt'$$

gauge invariance of transport coefficients

$$\mathbf{J}' = \mathbf{J} + \dot{\mathbf{P}}$$

$$\lambda \sim \frac{1}{2t}\mathrm{var}\big[\mathbf{D}(t)\big] \qquad \mathbf{D}(t) = \int_0^t \mathbf{J}(t')dt'$$

$$\mathbf{D}'(t) = \mathbf{D}(t) + \mathbf{P}(t) - \mathbf{P}(0)$$

gauge invariance of transport coefficients

$$\mathbf{J}' = \mathbf{J} + \dot{\mathbf{P}}$$

$$\lambda \sim \frac{1}{2t}\mathrm{var}\left[\mathbf{D}(t)\right] \qquad \mathbf{D}(t) = \int_0^t \mathbf{J}(t')dt'$$

$$\mathbf{D}'(t) = \mathbf{D}(t) + \mathbf{P}(t) - \mathbf{P}(0)$$

$$\mathrm{var}\left[\mathbf{D}'(t)\right] = \mathrm{var}\left[\mathbf{D}(t)\right] + \mathrm{var}\left[\Delta\mathbf{P}(t)\right] + 2\mathrm{cov}\left[\mathbf{D}(t) \cdot \Delta\mathbf{P}(t)\right]$$

gauge invariance of transport coefficients

$$\mathbf{J}' = \mathbf{J} + \dot{\mathbf{P}}$$

$$\lambda \sim \frac{1}{2t}\mathrm{var}\left[\mathbf{D}(t)\right] \qquad \mathbf{D}(t) = \int_0^t \mathbf{J}(t')dt'$$

$$\mathbf{D}'(t) = \mathbf{D}(t) + \mathbf{P}(t) - \mathbf{P}(0)$$

$$\mathrm{var}\left[\mathbf{D}'(t)\right] = \underbrace{\mathrm{var}\left[\mathbf{D}(t)\right]}_{\mathcal{O}(t)} + \cancel{\underbrace{\mathrm{var}\left[\Delta\mathbf{P}(t)\right]}_{\mathcal{O}(1)}} + \cancel{\underbrace{2\mathrm{cov}\left[\mathbf{D}(t)\cdot\Delta\mathbf{P}(t)\right]}_{\mathcal{O}(t^{\frac{1}{2}})}}$$

gauge invariance of transport coefficients

any two conserved densities that differ by the divergence of a (bounded) vector field are physically equivalent

the corresponding conserved fluxes differ by a total time derivative, and the transport coefficients coincide

nature physics
ARTICLES
PUBLISHED ONLINE: 19 OCTOBER 2015 | DOI: 10.1038/NPHYS3509

Microscopic theory and quantum simulation of atomic heat transport

Aris Marcolongo[1], Paolo Umari[2] and Stefano Baroni[1]*

gauge invariance of heat transport

PRL **104,** 208501 (2010) PHYSICAL REVIEW LETTERS week ending 21 MAY 2010

Thermal Conductivity of Periclase (MgO) from First Principles

Stephen Stackhouse*
Department of Geological Sciences, University of Michigan, Ann Arbor, Michigan, 48109-1005, USA

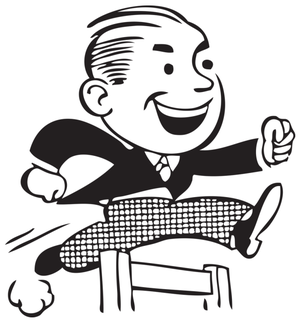
Lars Stixrude†
Department of Earth Sciences, University College London, Gower Street, London WC1E 6BT, United Kingdom

Bijaya B. Karki‡
Department of Computer Science, Louisiana State University, Baton Rouge, Louisiana 70803, USA
and Department of Geology and Geophysics, Louisiana State University, Baton Rouge, Louisiana 70803, USA

sensitive to the form of the potential. The widely used Green-Kubo relation [14] does not serve our purposes, because in first-principles calculations it is impossible to uniquely decompose the total energy into individual contributions from each atom.

solution:

choose *any* local representation of the energy that integrates to the correct value and whose correlations decay at large distance — the conductivity computed from the resulting current will be *independent* of the chosen representation.



correlation functions from molecular dynamics

$$\lambda \propto \int_0^\infty \langle \boldsymbol{J}(t)\boldsymbol{J}(0)\rangle dt$$

$$\boldsymbol{J}(t) = \boldsymbol{J}(\Gamma_t)$$

$$\Gamma_t = \{q_t, p_t\}$$

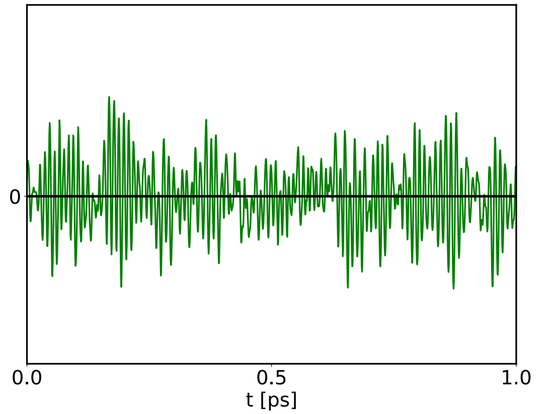
$$\dot{q} = \frac{\partial H}{\partial p}$$

$$\dot{p} = -\frac{\partial H}{\partial q}$$

correlation functions from molecular dynamics

$$\lambda \propto \int_0^\infty \langle \boldsymbol{J}(t)\boldsymbol{J}(0)\rangle dt$$

$$\boldsymbol{J}(t) = \boldsymbol{J}(\Gamma_t)$$



$$\Gamma_t = \{q_t, p_t\}$$

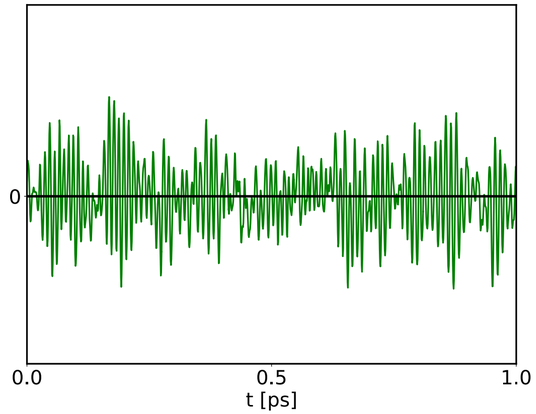
$$\dot{q} = \frac{\partial H}{\partial p}$$

$$\dot{p} = -\frac{\partial H}{\partial q}$$

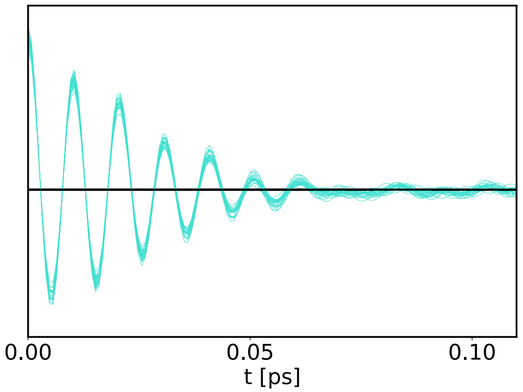
correlation functions from molecular dynamics

$$\lambda \propto \int_0^\infty \langle \boldsymbol{J}(t)\boldsymbol{J}(0)\rangle dt$$

$$\boldsymbol{J}(t) = \boldsymbol{J}(\Gamma_t)$$



$$\langle \boldsymbol{J}(t)\boldsymbol{J}(0)\rangle = \frac{1}{T-t}\int_0^{T-t} \boldsymbol{J}(t+t')\boldsymbol{J}(t')dt'$$



$$\Gamma_t = \{q_t, p_t\}$$

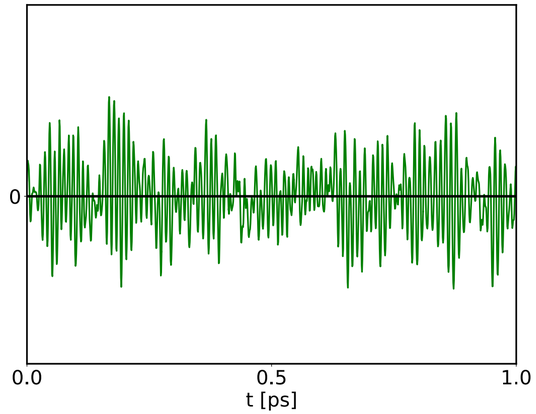
$$\dot{q} = \frac{\partial H}{\partial p}$$

$$\dot{p} = -\frac{\partial H}{\partial q}$$

correlation functions from molecular dynamics

$$\lambda \propto \int_0^\infty \langle \boldsymbol{J}(t)\boldsymbol{J}(0)\rangle dt$$

$$\boldsymbol{J}(t) = \boldsymbol{J}(\Gamma_t)$$

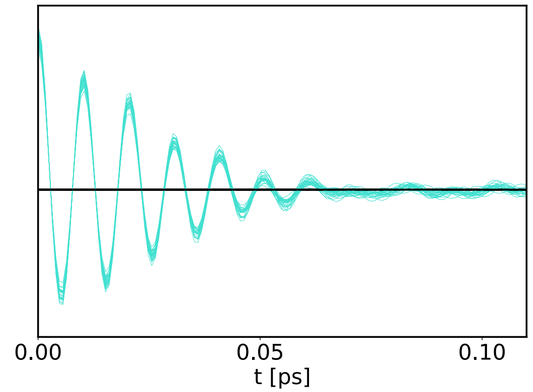


$$\Gamma_t = \{q_t, p_t\}$$

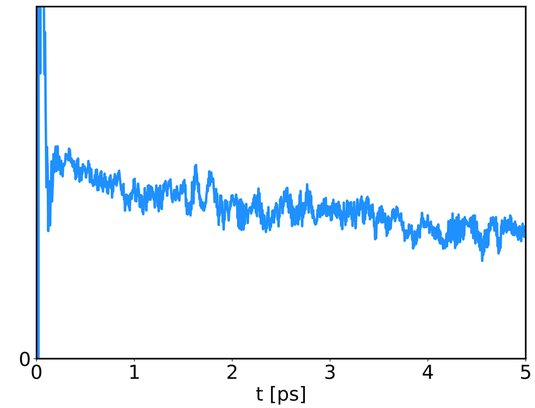
$$\dot{q} = \frac{\partial H}{\partial p}$$

$$\dot{p} = -\frac{\partial H}{\partial q}$$

$$\langle \boldsymbol{J}(t)\boldsymbol{J}(0)\rangle = \frac{1}{T-t}\int_0^{T-t} \boldsymbol{J}(t+t')\boldsymbol{J}(t')dt'$$



$$\sigma(t) = \int_0^t \langle \boldsymbol{J}(t')\boldsymbol{J}(0)\rangle dt'$$



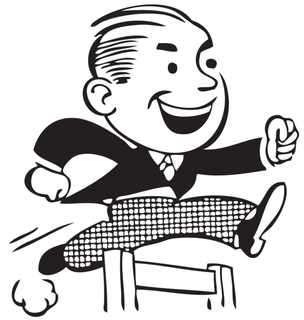
hurdles toward an ab initio Green-Kubo theory

PRL **104,** 208501 (2010) PHYSICAL REVIEW LETTERS week ending 21 MAY 2010

Thermal Conductivity of Periclase (MgO) from First Principles

Stephen Stackhouse, Lars Stixrude, and Bijaya B. Karki

sensitive to the form of the potential. The widely used Green-Kubo relation [14] does not serve our purposes, because in first-principles calculations it is impossible to uniquely decompose the total energy into individual contributions from each atom.

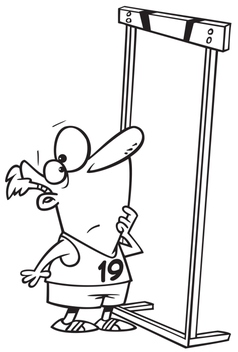


PRL **118,** 175901 (2017) PHYSICAL REVIEW LETTERS week ending 28 APRIL 2017

***Ab Initio* Green-Kubo Approach for the Thermal Conductivity of Solids**

Christian Carbogno, Rampi Ramprasad, and Matthias Scheffler

ulations: Because of the limited time scales accessible in aiMD runs, thermodynamic fluctuations dominate the HFACF, which in turn prevents a reliable and numerically stable assessment of the thermal conductivity via Eq. (2).



spectral analysis

$$\boldsymbol{J} = \int_V \boldsymbol{j}(\boldsymbol{r})\, d\boldsymbol{r}$$

$$= \sum_i \int_{V_i} \boldsymbol{j}(\boldsymbol{r})\, d\boldsymbol{r}$$

spectral analysis

$$\boldsymbol{J} = \int_V \boldsymbol{j}(\boldsymbol{r}) d\boldsymbol{r}$$
$$= \sum_i \int_{V_i} \boldsymbol{j}(\boldsymbol{r}) d\boldsymbol{r}$$

if $\langle \boldsymbol{j}(\boldsymbol{r}) \boldsymbol{j}(\boldsymbol{r}') \rangle$ is short-range, $\int_{V_i} \boldsymbol{j}(\boldsymbol{r}) d\boldsymbol{r}$ and $\int_{V_j} \boldsymbol{j}(\boldsymbol{r}) d\boldsymbol{r}$ for $i \neq j$ are independent stochastic variables and, by the central-limit theorem,

$\boldsymbol{J}(t)$ is a Gaussian process

spectral analysis

$$\boldsymbol{J} = \int_V \boldsymbol{j}(\boldsymbol{r})d\boldsymbol{r}$$
$$= \sum_i \int_{V_i} \boldsymbol{j}(\boldsymbol{r})d\boldsymbol{r}$$

if $\langle \boldsymbol{j}(\boldsymbol{r})\boldsymbol{j}(\boldsymbol{r}')\rangle$ is short-range, $\int_{V_i} \boldsymbol{j}(\boldsymbol{r})d\boldsymbol{r}$ and $\int_{V_j} \boldsymbol{j}(\boldsymbol{r})d\boldsymbol{r}$ for $i \neq j$ are independent stochastic variables and, by the central-limit theorem,

$\boldsymbol{J}(t)$ is a Gaussian process

$\tilde{\boldsymbol{J}}_T(\omega) = \int_0^T \boldsymbol{J}(t)\mathrm{e}^{i\omega t}dt$ is Gaussian as well

spectral analysis

$$\boldsymbol{J} = \int_V \boldsymbol{j}(\boldsymbol{r}) d\boldsymbol{r}$$

$$= \sum_i \int_{V_i} \boldsymbol{j}(\boldsymbol{r}) d\boldsymbol{r}$$

if $\langle \boldsymbol{j}(\boldsymbol{r}) \boldsymbol{j}(\boldsymbol{r}') \rangle$ is short-range, $\int_{V_i} \boldsymbol{j}(\boldsymbol{r}) d\boldsymbol{r}$ and $\int_{V_j} \boldsymbol{j}(\boldsymbol{r}) d\boldsymbol{r}$ for $i \neq j$ are independent stochastic variables and, by the central-limit theorem,

$\boldsymbol{J}(t)$ is a Gaussian process

$\tilde{\boldsymbol{J}}_T(\omega) = \int_0^T \boldsymbol{J}(t) \mathrm{e}^{i\omega t} dt$ is Gaussian as well

stationarity implies:

$$\langle \tilde{\boldsymbol{J}}_T(\omega) \tilde{\boldsymbol{J}}_T(-\omega') \rangle \sim \frac{1}{T} \quad \text{for} \quad \omega \neq \omega'$$

spectral analysis

$$\lambda = \int_0^\infty \langle \boldsymbol{J}(t)\boldsymbol{J}(0)\rangle dt \qquad S(\omega)$$

$$= \frac{1}{2}\int_{-\infty}^{\infty} \langle \boldsymbol{J}(t)\boldsymbol{J}(0)\rangle e^{i\omega t} dt \Big|_{\omega=0}$$

$$= \frac{1}{2} S(0)$$

spectral analysis

$$\lambda = \int_0^\infty \langle \boldsymbol{J}(t)\boldsymbol{J}(0)\rangle dt \qquad S(\omega)$$

$$= \frac{1}{2}\int_{-\infty}^{\infty} \langle \boldsymbol{J}(t)\boldsymbol{J}(0)\rangle e^{i\omega t} dt \Big|_{\omega=0}$$

$$= \frac{1}{2} S(0)$$

Wiener-Kintchine theorem

$$S(\omega) = \lim_{T\to\infty} \frac{1}{T}\left\langle \left|\tilde{J}_T(\omega)\right|^2 \right\rangle$$

$$\tilde{\boldsymbol{J}}_T(\omega) = \int_0^T \boldsymbol{J}(t) e^{i\omega t} dt$$

spectral analysis

$$\lambda = \int_0^\infty \langle \boldsymbol{J}(t)\boldsymbol{J}(0)\rangle dt \qquad S(\omega)$$

$$= \frac{1}{2}\int_{-\infty}^{\infty} \langle \boldsymbol{J}(t)\boldsymbol{J}(0)\rangle e^{i\omega t} dt \Big|_{\omega=0}$$

$$= \frac{1}{2} S(0)$$

Wiener-Kintchine theorem

$$S(\omega) = \lim_{T\to\infty} \frac{1}{T}\left\langle \left|\tilde{J}_T(\omega)\right|^2 \right\rangle$$

$$\tilde{\boldsymbol{J}}_T(\omega) = \int_0^T \boldsymbol{J}(t) e^{i\omega t} dt$$

$$\tilde{\boldsymbol{J}}_T(\omega_k) \sim \mathcal{CN}(0, TS(\omega_k) \times I)$$

spectral analysis

$$\lambda = \int_0^\infty \langle \boldsymbol{J}(t)\boldsymbol{J}(0)\rangle dt \qquad S(\omega)$$

$$= \frac{1}{2}\int_{-\infty}^{\infty} \langle \boldsymbol{J}(t)\boldsymbol{J}(0)\rangle \mathrm{e}^{i\omega t} dt \Big|_{\omega=0}$$

$$= \frac{1}{2} S(0)$$

Wiener-Kintchine theorem

$$S(\omega) = \lim_{T\to\infty} \frac{1}{T}\left\langle \left|\tilde{J}_T(\omega)\right|^2 \right\rangle$$

$$\tilde{\boldsymbol{J}}_T(\omega) = \int_0^T \boldsymbol{J}(t)\mathrm{e}^{i\omega t} dt$$

$$\tilde{\boldsymbol{J}}_T(\omega_k) \sim \mathcal{CN}(0, TS(\omega_k) \times I)$$

$$\hat{S}_k \doteq \frac{1}{T}\left|\tilde{J}_T(\omega_k)\right|^2$$

$$\sim \frac{1}{2} S(\omega_k)\hat{\chi}^2_2$$

sample spectrum
aka "*periodogram*"

$$\doteq S(\omega_k)\hat{\xi}_k$$

spectral analysis

$$\lambda = \int_0^\infty \langle \boldsymbol{J}(t)\boldsymbol{J}(0)\rangle dt \qquad S(\omega)$$

$$= \frac{1}{2}\int_{-\infty}^{\infty} \langle \boldsymbol{J}(t)\boldsymbol{J}(0)\rangle \mathrm{e}^{i\omega t} dt \Big|_{\omega=0}$$

$$= \frac{1}{2} S(0)$$

Wiener-Kintchine theorem

$$S(\omega) = \lim_{T\to\infty} \frac{1}{T}\left\langle \left|\tilde{J}_T(\omega)\right|^2 \right\rangle$$

$$\tilde{\boldsymbol{J}}_T(\omega) = \int_0^T \boldsymbol{J}(t)\mathrm{e}^{i\omega t} dt$$

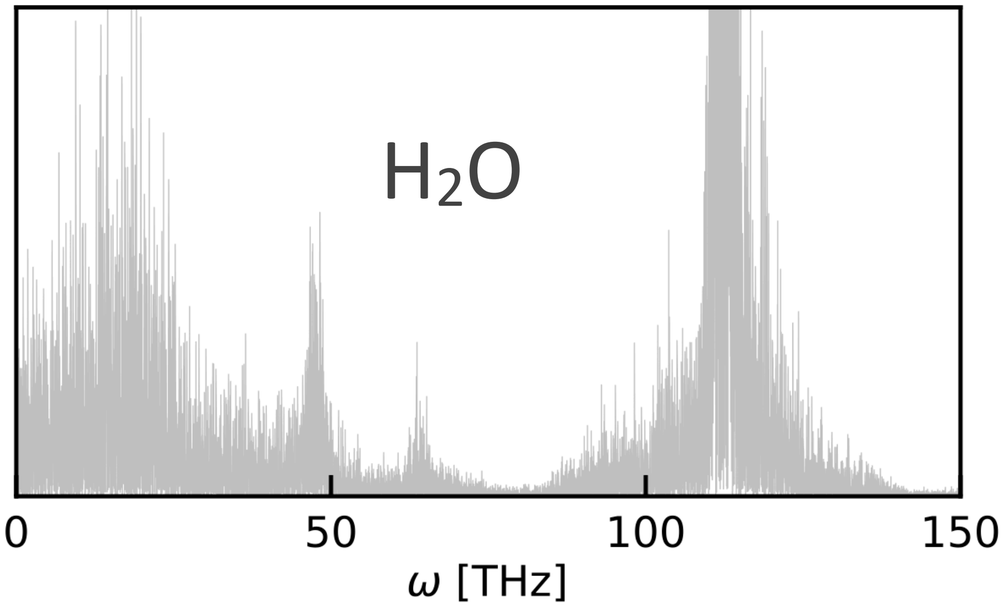
$$\tilde{\boldsymbol{J}}_T(\omega_k) \sim \mathcal{CN}(0, TS(\omega_k) \times I)$$

$$\hat{S}_k \doteq \frac{1}{T}\left|\tilde{J}_T(\omega_k)\right|^2$$

$$\sim \frac{1}{2} S(\omega_k)\hat{\chi}^2_2$$

sample spectrum
aka "*periodogram*"

$$\doteq S(\omega_k)\hat{\xi}_k$$



spectral analysis

$$\lambda = \int_0^\infty \langle \boldsymbol{J}(t)\boldsymbol{J}(0)\rangle dt \qquad S(\omega)$$

$$= \frac{1}{2}\int_{-\infty}^{\infty} \langle \boldsymbol{J}(t)\boldsymbol{J}(0)\rangle e^{i\omega t} dt \Big|_{\omega=0}$$

$$= \frac{1}{2} S(0)$$

Wiener-Kintchine theorem

$$S(\omega) = \lim_{T\to\infty} \frac{1}{T}\left\langle \left|\tilde{J}_T(\omega)\right|^2 \right\rangle$$

$$\tilde{\boldsymbol{J}}_T(\omega) = \int_0^T \boldsymbol{J}(t) e^{i\omega t} dt$$

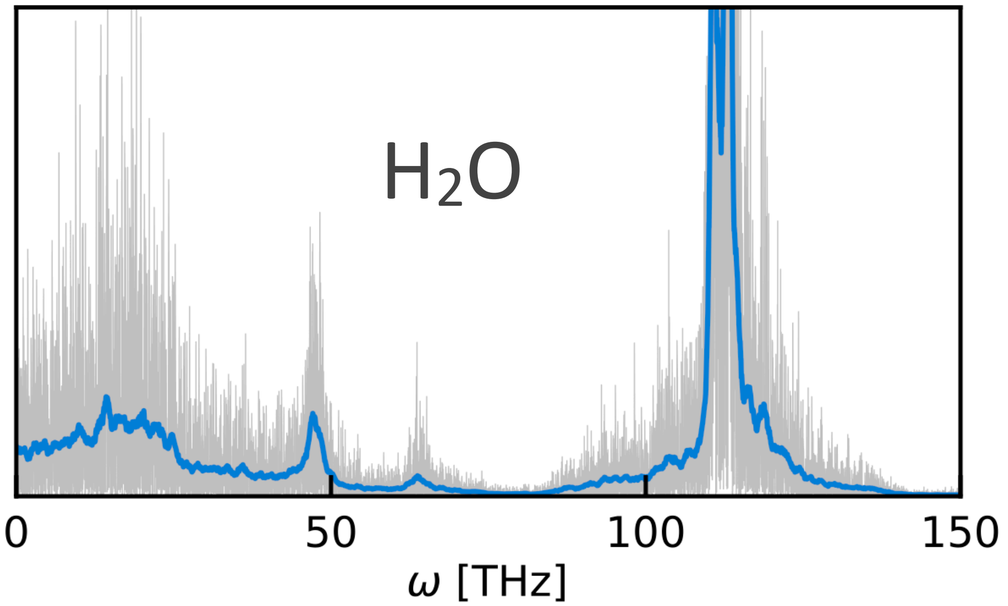
$$\tilde{\boldsymbol{J}}_T(\omega_k) \sim \mathcal{CN}(0, TS(\omega_k) \times I)$$

$$\hat{S}_k \doteq \frac{1}{T}\left|\tilde{J}_T(\omega_k)\right|^2$$

$$\sim \frac{1}{2} S(\omega_k)\hat{\chi}^2_2$$

sample spectrum
aka "*periodogram*"

$$\doteq S(\omega_k)\hat{\xi}_k$$



spectral analysis

$$\lambda = \int_0^\infty \langle \boldsymbol{J}(t)\boldsymbol{J}(0)\rangle dt \qquad S(\omega)$$

$$= \frac{1}{2}\int_{-\infty}^{\infty} \langle \boldsymbol{J}(t)\boldsymbol{J}(0)\rangle \mathrm{e}^{i\omega t} dt \Big|_{\omega=0}$$

$$= \frac{1}{2} S(0)$$

Wiener-Kintchine theorem

$$S(\omega) = \lim_{T\to\infty} \frac{1}{T}\left\langle \left|\tilde{J}_T(\omega)\right|^2 \right\rangle$$

$$\tilde{\boldsymbol{J}}_T(\omega) = \int_0^T \boldsymbol{J}(t)\mathrm{e}^{i\omega t} dt$$

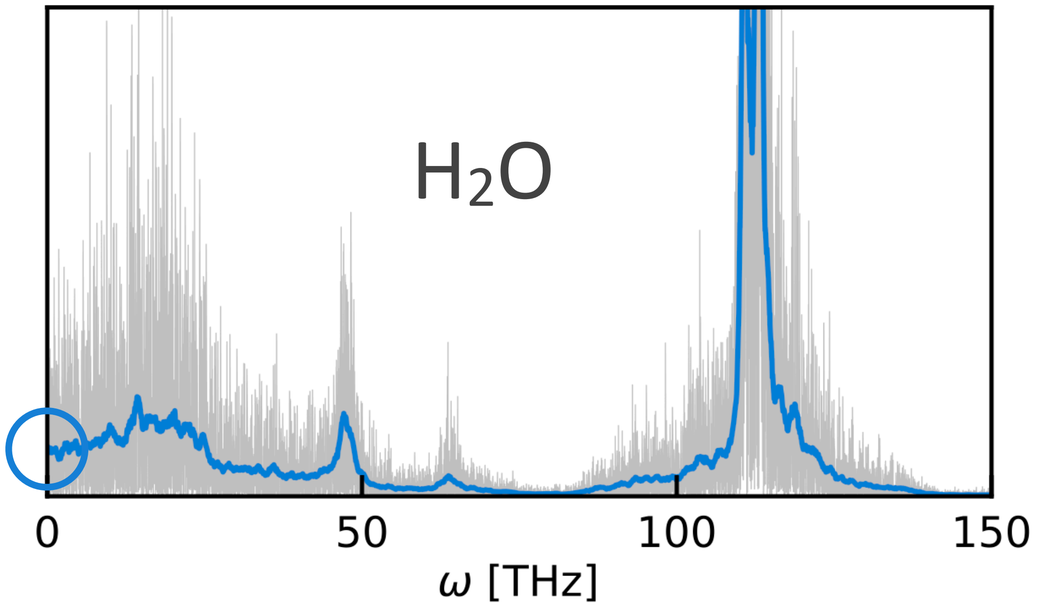
$$\tilde{\boldsymbol{J}}_T(\omega_k) \sim \mathcal{CN}(0, TS(\omega_k) \times I)$$

$$\hat{S}_k \doteq \frac{1}{T}\left|\tilde{J}_T(\omega_k)\right|^2$$

$$\sim \frac{1}{2} S(\omega_k)\hat{\chi}^2_2$$

$$\doteq S(\omega_k)\hat{\xi}_k$$

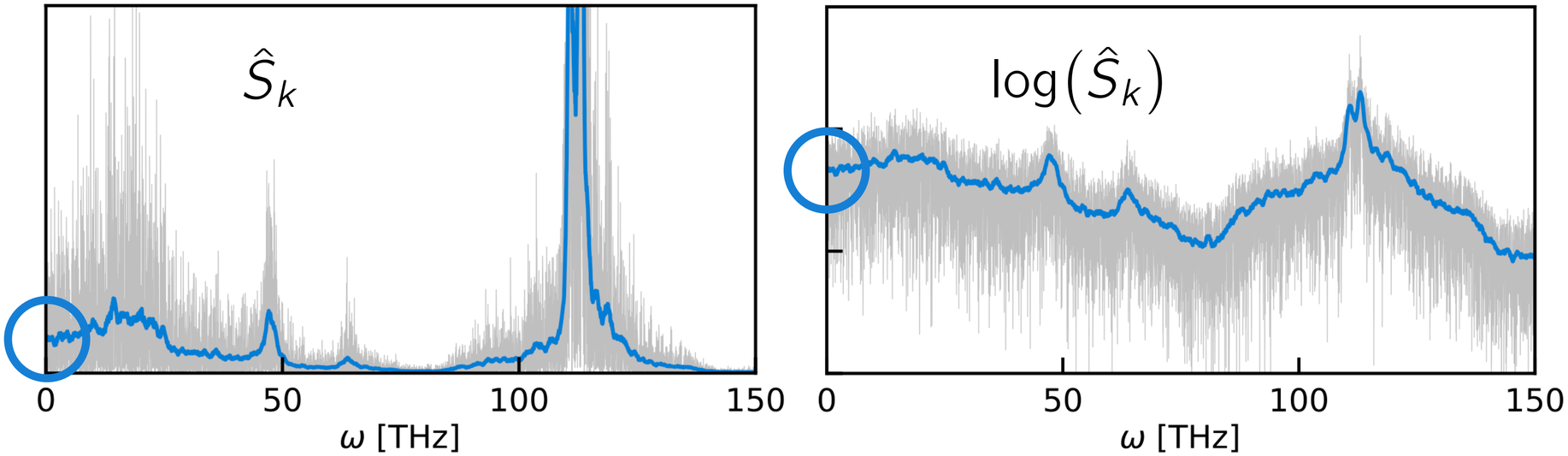
sample spectrum
aka "*periodogram*"



separating flour from bran

$$\hat{S}_k = S(\omega_k)\hat{\xi}_k$$

$$\log\left(\hat{S}_k\right) = \log\left(S(\omega_k)\right) + \log\left(\hat{\xi}_k\right)$$



separating flour from bran

$$\hat{S}_k = S(\omega_k)\hat{\xi}_k$$

$$\log(\hat{S}_k) = \log\big(S(\omega_k)\big) + \log(\hat{\xi}_k)$$

$$= \log\big(S(\omega_k)\big) + \lambda + \hat{\lambda}_k$$

$$\langle \hat{\lambda} \rangle = 0$$

$$\langle \hat{\lambda}^2 \rangle = \sigma^2$$

separating flour from bran

$$\hat{S}_k = S(\omega_k)\hat{\xi}_k$$

$$\log\left(\hat{S}_k\right) = \log\left(S(\omega_k)\right) + \log\left(\hat{\xi}_k\right)$$

$$= \log\left(S(\omega_k)\right) + \lambda + \hat{\lambda}_k$$

$$\langle\hat{\lambda}\rangle = 0$$

$$\langle\hat{\lambda}^2\rangle = \sigma^2$$

"cepstral coefficients"
(J.W. Tukey, 1963)

$$\hat{C}_n \doteq \frac{1}{N}\sum_{k=0}^{N-1} \log\left(\hat{S}_k\right) \mathrm{e}^{2\pi i \frac{kn}{N}}$$

$$= C_n + \lambda\delta_{n0} + \hat{w}_n$$

$$\hat{w} \sim \mathcal{N}\left(0, \frac{\sigma^2}{N}\right)$$

separating flour from bran

$$\hat{S}_k = S(\omega_k)\hat{\xi}_k$$

$$\log\left(\hat{S}_k\right) = \log\left(S(\omega_k)\right) + \log\left(\hat{\xi}_k\right)$$

$$= \log\left(S(\omega_k)\right) + \lambda + \hat{\lambda}_k$$

$$\langle\hat{\lambda}\rangle = 0$$

$$\langle\hat{\lambda}^2\rangle = \sigma^2$$

"cepstral coefficients"
(J.W. Tukey, 1963)

$$\hat{C}_n \doteq \frac{1}{N}\sum_{k=0}^{N-1} \log\left(\hat{S}_k\right) e^{2\pi i \frac{kn}{N}}$$

$$= C_n + \lambda\delta_{n0} + \hat{w}_n$$

$$\hat{w} \sim \mathcal{N}\left(0, \frac{\sigma^2}{N}\right)$$

$$\sum_{n=0}^{N-1} \hat{C}_n e^{2\pi i \frac{kn}{N}} = \log\left(S(\omega_k)\right) + \lambda + \hat{\lambda}_k$$

$$\sum_{n=-P^*+1}^{P^*-1} \hat{C}_n e^{2\pi i \frac{kn}{N}} = \log\left(S(\omega_k)\right) + \lambda + \widehat{W}_k$$

$$\widehat{W} \sim \mathcal{N}\left(0, \frac{4P^* - 2}{N}\sigma^2\right)$$

the cepstral cavobulary

The Quefrency Alanysis of Time Series for Echoes: Cepstrum, Pseudo-Autocovariance, Cross-Cepstrum and Saphe Cracking

Bruce P. Bogert, M. J. R. Healy,* John W. Tukey†
Bell Telephone Laboratories and Princeton University

Proceedings of the Symposium on Time Series Analysis (M. Rosenblatt, Ed) Chapter 15, 209-243. New York: Wiley, 1963.

the cepstral cavobulary

The Quefrency Alanysis of Time Series for Echoes: Cepstrum, Pseudo-Autocovariance, Cross-Cepstrum and Saphe Cracking

Bruce P. Bogert, M. J. R. Healy,* John W. Tukey†
Bell Telephone Laboratories and Princeton University

Proceedings of the Symposium on Time Series Analysis (M. Rosenblatt, Ed) Chapter 15, 209-243. New York: Wiley, 1963.

the cepstral cavobulary

The Quefrency Alanysis of Time Series for Echoes: Cepstrum, Pseudo-Autocovariance, Cross-Cepstrum and Saphe Cracking

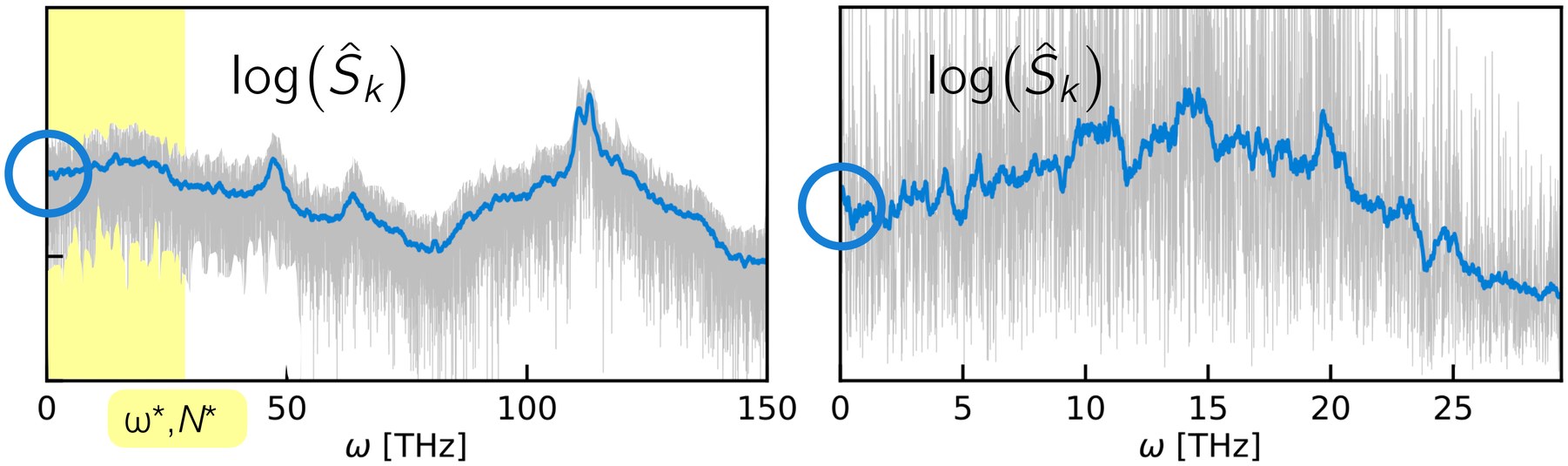
Bruce P. Bogert, M. J. R. Healy,* John W. Tukey†
Bell Telephone Laboratories and Princeton University

Proceedings of the Symposium on Time Series Analysis (M. Rosenblatt, Ed) Chapter 15, 209-243. New York: Wiley, 1963.

spectrum	.....	cepstrum
frequency	.....	quefrency
analysis	.....	alansys
period	.....	repiod
filtering	.....	liftering
phase	.....	saphe

separating flour from bran

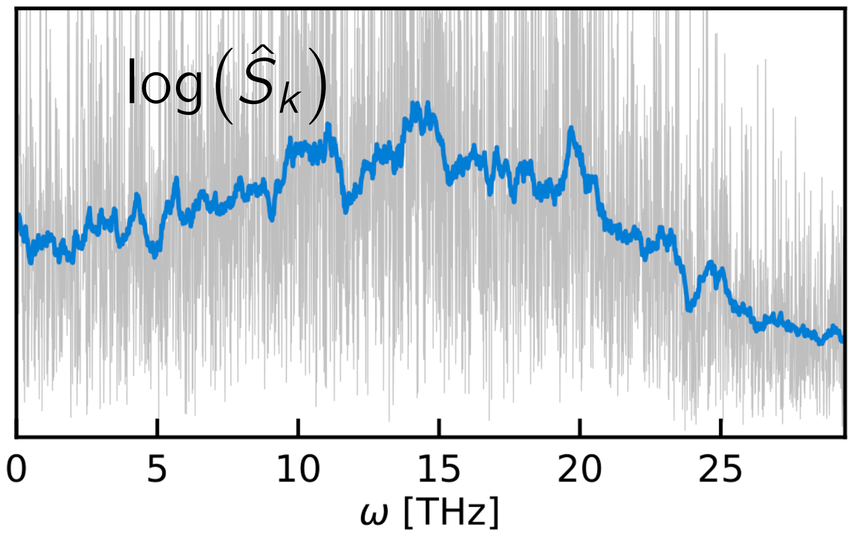
$$\hat{C}_n \doteq \frac{1}{N^*} \sum_{k=0}^{N^*-1} \log\left(\hat{S}_k\right) \mathrm{e}^{2\pi i \frac{kn}{N}}$$

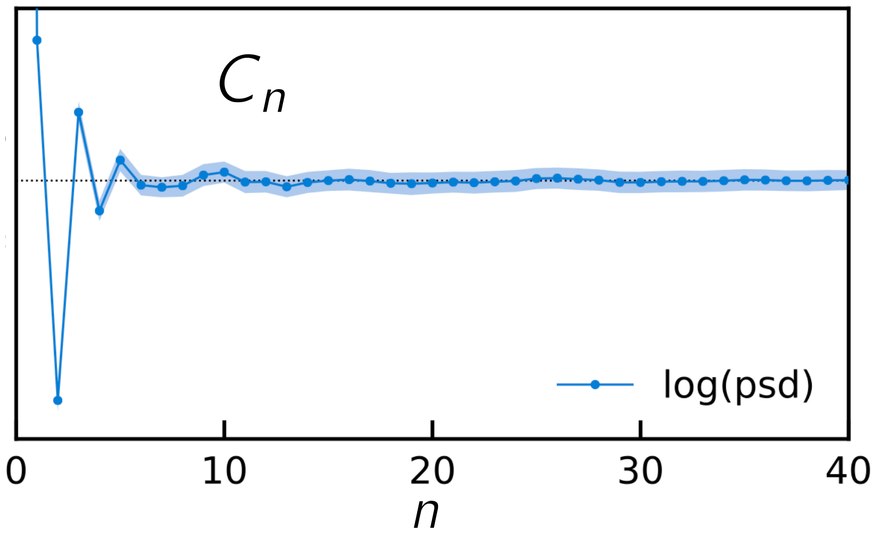


separating flour from bran

$$\hat{C}_n \doteq \frac{1}{N^*} \sum_{k=0}^{N^*-1} \log\left(\hat{S}_k\right) e^{2\pi i \frac{kn}{N}}$$

$$\sum_{n=0}^{N^*-1} \hat{C}_n e^{2\pi i \frac{nk}{N^*}} = \log\left(S(\omega_k)\right) + \lambda + \text{noise}$$



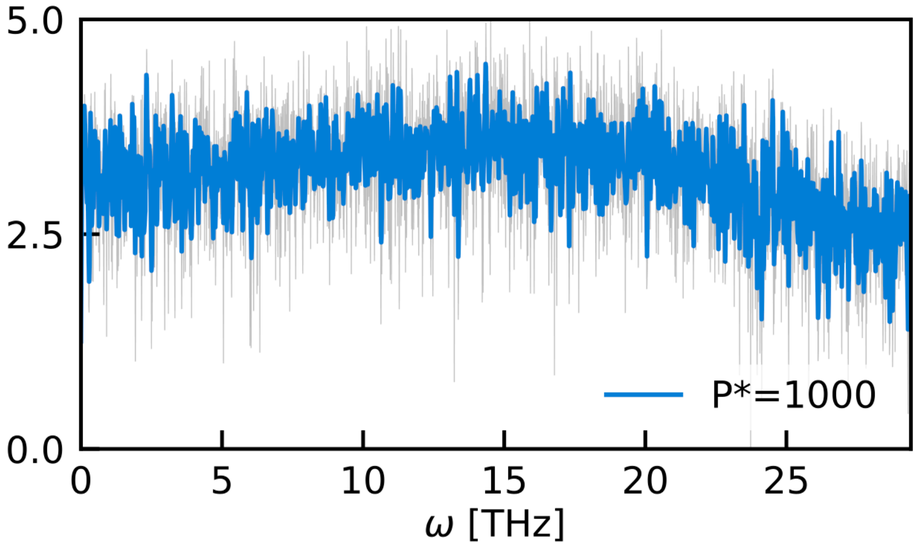


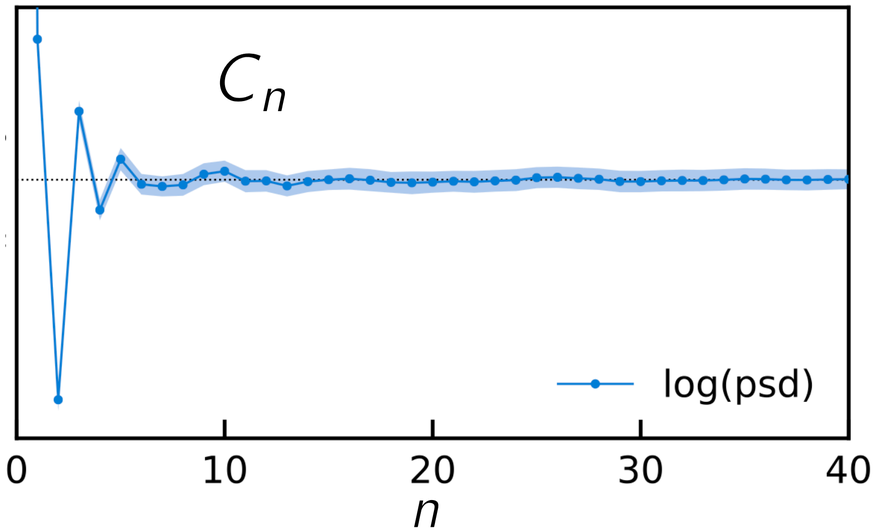
separating flour from bran

$$\hat{C}_n \doteq \frac{1}{N^*} \sum_{k=0}^{N^*-1} \log\left(\hat{S}_k\right) e^{2\pi i \frac{kn}{N}}$$

$$\sum_{n=0}^{N^*-1} \hat{C}_n e^{2\pi i \frac{nk}{N^*}} = \log\left(S(\omega_k)\right) + \lambda + \text{noise}$$

$$\sum_{n=-P^*+1}^{P^*-1} \hat{C}_n e^{2\pi i \frac{nk}{N^*}} = \log\left(S(\omega_k)\right) + \lambda + \text{less noise}$$



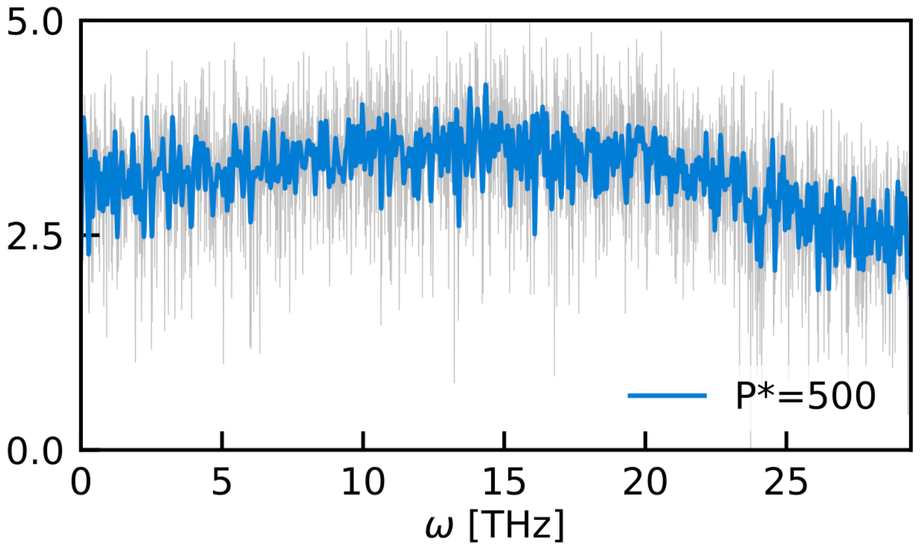


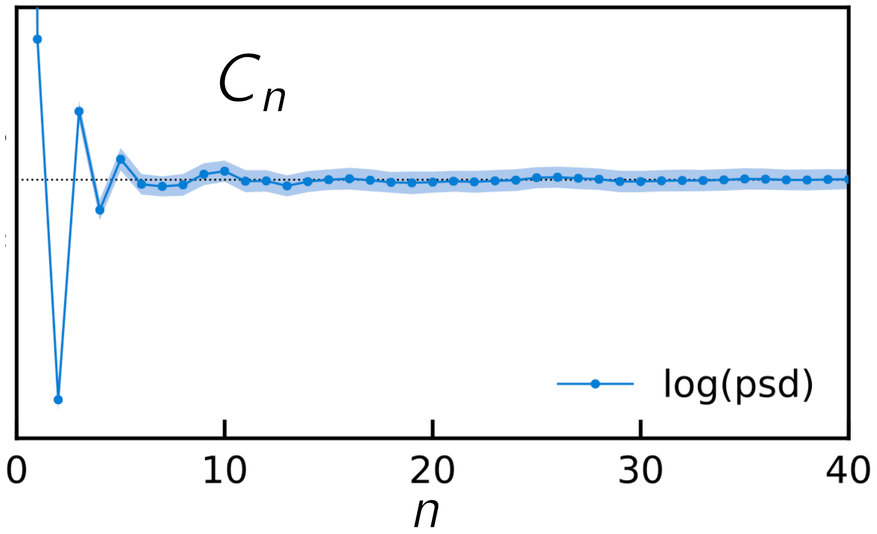
separating flour from bran

$$\hat{C}_n \doteq \frac{1}{N^*} \sum_{k=0}^{N^*-1} \log\left(\hat{S}_k\right) e^{2\pi i \frac{kn}{N}}$$

$$\sum_{n=0}^{N^*-1} \hat{C}_n e^{2\pi i \frac{nk}{N^*}} = \log\left(S(\omega_k)\right) + \lambda + \text{noise}$$

$$\sum_{n=-P^*+1}^{P^*-1} \hat{C}_n e^{2\pi i \frac{nk}{N^*}} = \log\left(S(\omega_k)\right) + \lambda + \text{less noise}$$



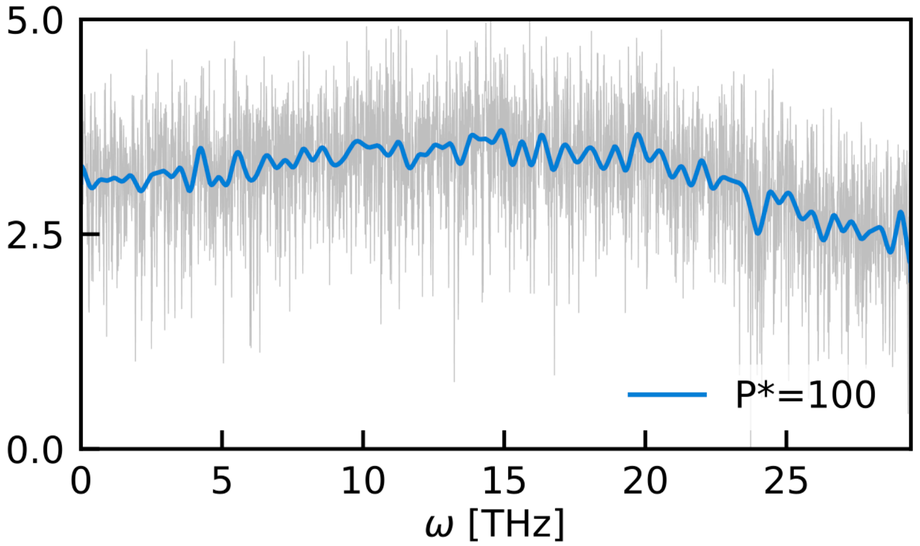


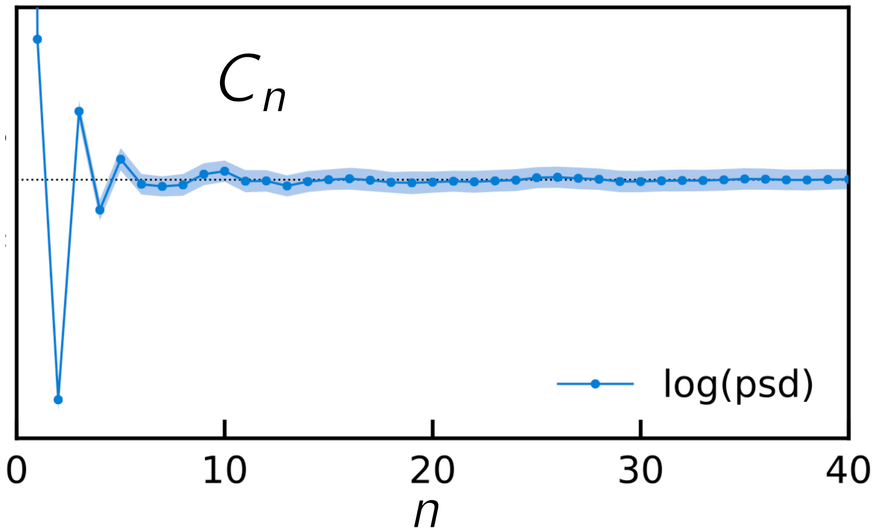
separating flour from bran

$$\hat{C}_n \doteq \frac{1}{N^*} \sum_{k=0}^{N^*-1} \log\left(\hat{S}_k\right) e^{2\pi i \frac{kn}{N}}$$

$$\sum_{n=0}^{N^*-1} \hat{C}_n e^{2\pi i \frac{nk}{N^*}} = \log\left(S(\omega_k)\right) + \lambda + \text{noise}$$

$$\sum_{n=-P^*+1}^{P^*-1} \hat{C}_n e^{2\pi i \frac{nk}{N^*}} = \log\left(S(\omega_k)\right) + \lambda + \text{less noise}$$



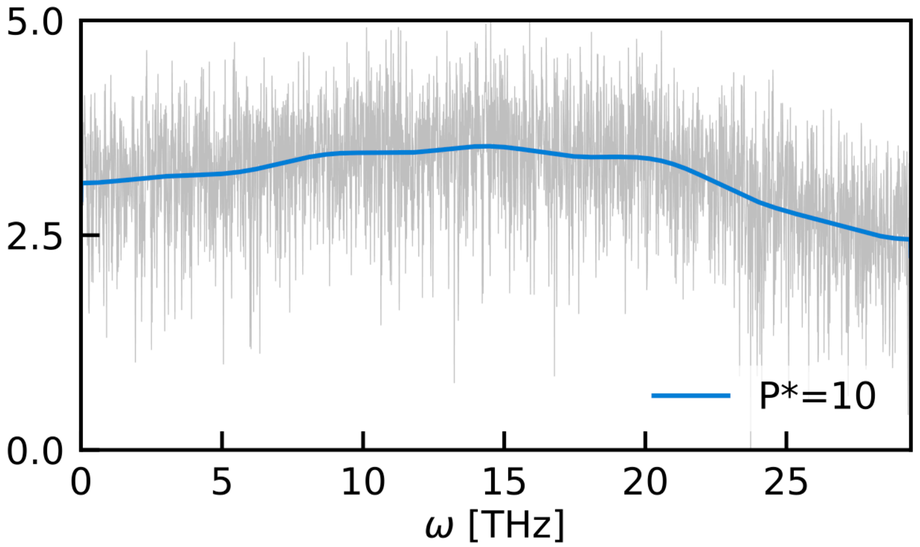


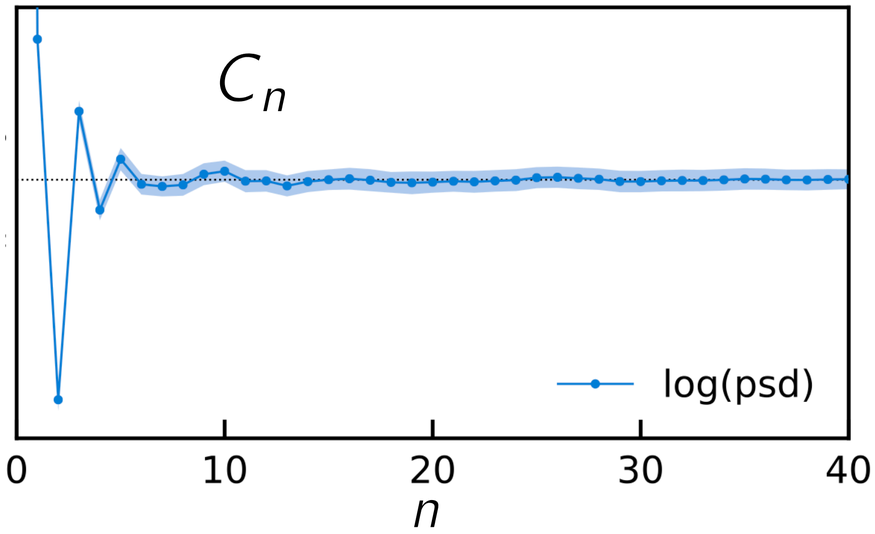
separating flour from bran

$$\hat{C}_n \doteq \frac{1}{N^*} \sum_{k=0}^{N^*-1} \log\left(\hat{S}_k\right) e^{2\pi i \frac{kn}{N}}$$

$$\sum_{n=0}^{N^*-1} \hat{C}_n e^{2\pi i \frac{nk}{N^*}} = \log\left(S(\omega_k)\right) + \lambda + \text{noise}$$

$$\sum_{n=-P^*+1}^{P^*-1} \hat{C}_n e^{2\pi i \frac{nk}{N^*}} = \log\left(S(\omega_k)\right) + \lambda + \text{less noise}$$





separating flour from bran

$$\sum_{n=-P^*+1}^{P^*-1} \hat{C}_n e^{2\pi i \frac{nk}{N^*}} = \log\big(S(\omega_k)\big) + \lambda + \text{less noise}$$

optimal number of coefficients, to be determined

separating flour from bran

$$\sum_{n=-P^*+1}^{P^*-1} \hat{C}_n e^{2\pi i \frac{nk}{N^*}} = \log\big(S(\omega_k)\big) + \lambda + \text{less noise}$$

optimal number of coefficients, to be determined

$$\log\big(S(0)\big) = -\lambda + C_0 + 2\sum_{n=1}^{P^*-1} C_n \pm \sigma\sqrt{\frac{4P^* - 2}{N^*}}$$

constants independent of the time series being sampled

determining the optimal number of cepstral coefficients

cepstral analysis amounts to assuming that the logarithm of the power spectrum can be modelled by a smooth Fourier series:

determining the optimal number of cepstral coefficients

cepstral analysis amounts to assuming that the logarithm of the power spectrum can be modelled by a smooth Fourier series:

Model: $\{P, C_0, C_1, \cdots C_{P-1}\}$; Data: $\{\hat{C}_0, \hat{C}_1, \cdots \hat{C}_{N/2}\}$.

determining the optimal number of cepstral coefficients

cepstral analysis amounts to assuming that the logarithm of the power spectrum can be modelled by a smooth Fourier series:

Model: $\{P, C_0, C_1, \cdots C_{P-1}\}$; Data: $\{\hat{C}_0, \hat{C}_1, \cdots \hat{C}_{N/2}\}$.
Optimal model, maximum of:

$$\mathcal{P}(\mathrm{M}|\mathrm{D}) = \frac{\mathcal{L}(\mathrm{D}|\mathrm{M})P(\mathrm{M})}{P(\mathrm{D})}$$

determining the optimal number of cepstral coefficients

cepstral analysis amounts to assuming that the logarithm of the power spectrum can be modelled by a smooth Fourier series:

Model: $\{P, C_0, C_1, \cdots C_{P-1}\}$; Data: $\{\hat{C}_0, \hat{C}_1, \cdots \hat{C}_{N/2}\}$.
Optimal model, maximum of:

$$\mathcal{P}(\mathrm{M}|\mathrm{D}) = \frac{\mathcal{L}(\mathrm{D}|\mathrm{M})P(\mathrm{M})}{P(\mathrm{D})}$$

$$-2\log(\mathcal{L}) \sim \frac{N}{\sigma^2}\left[\frac{1}{2}\left(C_0 + \lambda - \hat{C}_0\right)^2 + \sum_{n=1}^{P-1}\left(C_n - \hat{C}_n\right)^2 + \sum_{n=P}^{\frac{N}{2}}\hat{C}_n^2\right]$$

$$p(M) \propto \mathrm{e}^{-\alpha P}$$

determining the optimal number of cepstral coefficients

cepstral analysis amounts to assuming that the logarithm of the power spectrum can be modelled by a smooth Fourier series:

Model: $\{P, C_0, C_1, \cdots C_{P-1}\}$; Data: $\{\hat{C}_0, \hat{C}_1, \cdots \hat{C}_{N/2}\}$.
Optimal model, maximum of:

$$\mathcal{P}(\mathrm{M}|\mathrm{D}) = \frac{\mathcal{L}(\mathrm{D}|\mathrm{M})P(\mathrm{M})}{P(\mathrm{D})}$$

$$-2\log(\mathcal{L}) \sim \frac{N}{\sigma^2}\left[\frac{1}{2}\left(C_0 + \lambda - \hat{C}_0\right)^2 + \sum_{n=1}^{P-1}\left(C_n - \hat{C}_n\right)^2 + \sum_{n=P}^{\frac{N}{2}}\hat{C}_n^2\right]$$

$$p(M) \propto \mathrm{e}^{-\alpha P}$$

$$P^* = \underset{P}{\operatorname{argmin}}\left[\frac{N}{\sigma^2}\sum_{n=P}^{N/2} + \alpha P\right]$$

determining the optimal number of cepstral coefficients

cepstral analysis amounts to assuming that the logarithm of the power spectrum can be modelled by a smooth Fourier series:

Model: $\{P, C_0, C_1, \cdots C_{P-1}\}$; Data: $\{\hat{C}_0, \hat{C}_1, \cdots \hat{C}_{N/2}\}$.
Optimal model, maximum of:

$$\mathcal{P}(\mathrm{M}|\mathrm{D}) = \frac{\mathcal{L}(\mathrm{D}|\mathrm{M})P(\mathrm{M})}{P(\mathrm{D})}$$

$$-2\log(\mathcal{L}) \sim \frac{N}{\sigma^2}\left[\frac{1}{2}\left(C_0 + \lambda - \hat{C}_0\right)^2 + \sum_{n=1}^{P-1}\left(C_n - \hat{C}_n\right)^2 + \sum_{n=P}^{\frac{N}{2}}\hat{C}_n^2\right]$$

$$p(M) \propto \mathrm{e}^{-\alpha P}$$

$$P^* = \underset{P}{\operatorname{argmin}}\left[\frac{N}{\sigma^2}\sum_{n=P}^{N/2} + \alpha P\right]$$

AIC: $\alpha = 2$

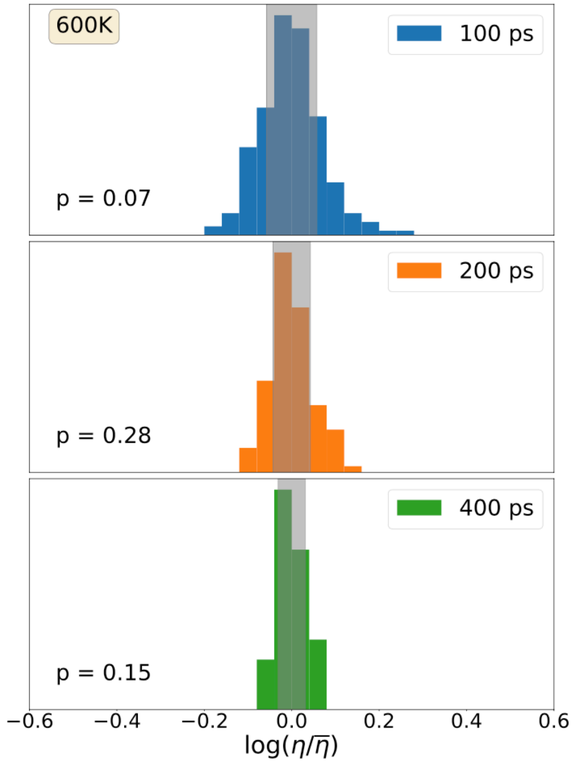
checking normality

$$\eta = \frac{V}{k_B T} \int_0^\infty \langle \sigma_s(t) \sigma_s(0) \rangle dt$$

checking normality

$$\eta = \frac{V}{k_B T} \int_0^\infty \langle \sigma_s(t)\sigma_s(0)\rangle dt$$

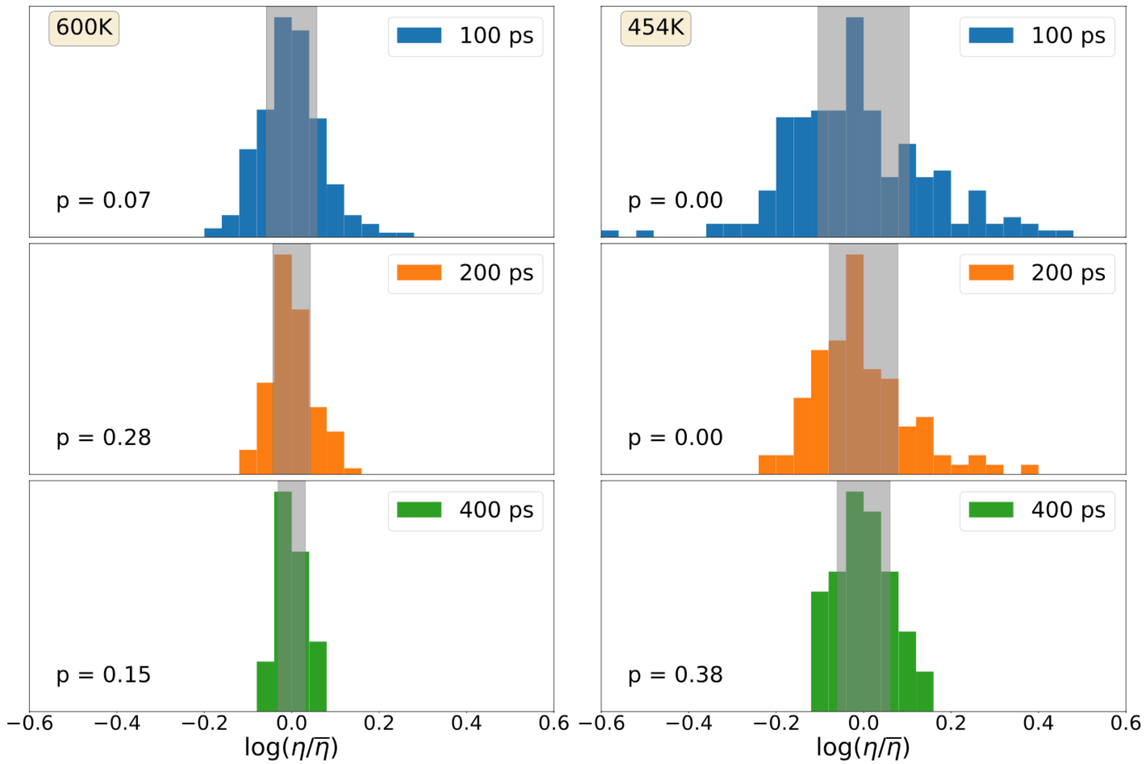
viscosity of water computed for different temperatures
and using trajectory segments of different lengths



checking normality

$$\eta = \frac{V}{k_B T} \int_0^\infty \langle \sigma_s(t)\sigma_s(0)\rangle dt$$

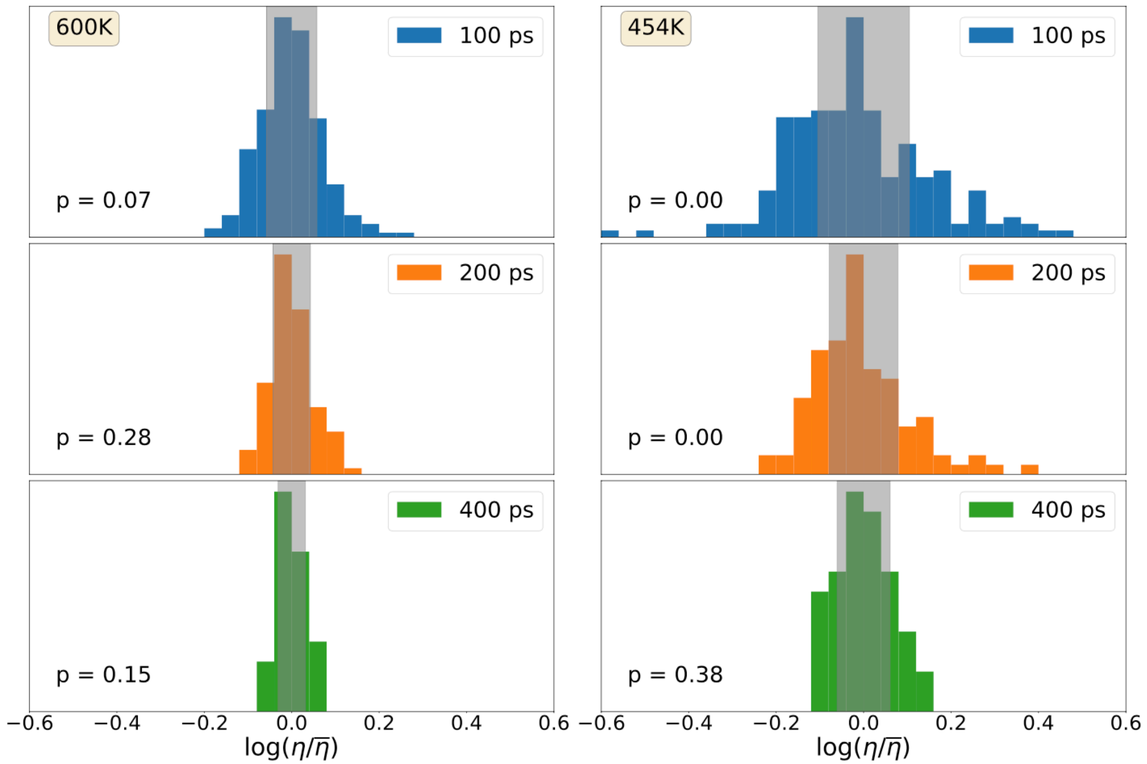
viscosity of water computed for different temperatures and using trajectory segments of different lengths

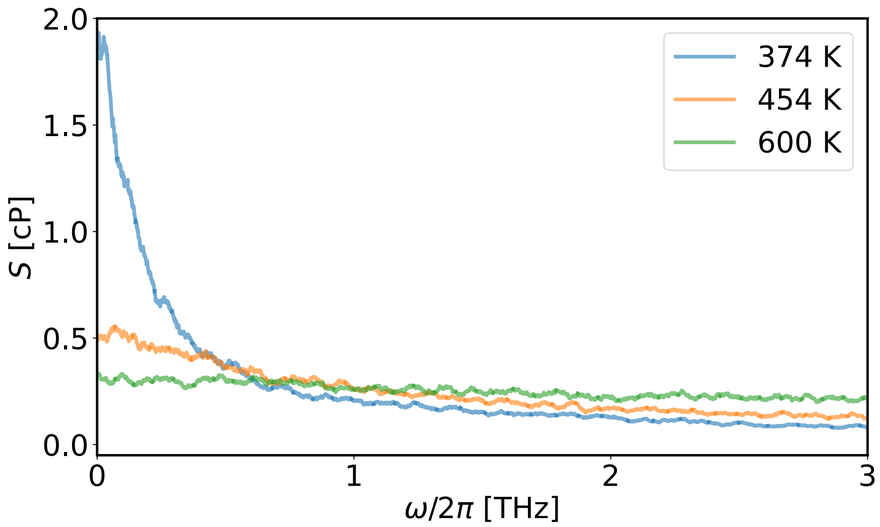


checking normality

$$\eta = \frac{V}{k_B T} \int_0^\infty \langle \sigma_s(t)\sigma_s(0)\rangle dt$$

viscosity of water computed for different temperatures and using trajectory segments of different lengths





multi-component systems

e.g. heat transport in a binary system: heat and mass/charge currents interact with each other

$$J_1 = \Lambda_{11} F_1 + \Lambda_{12} F_2$$
$$J_2 = \Lambda_{21} F_1 + \Lambda_{22} F_2$$

$$\Lambda_{ik} \propto \int_0^\infty \langle J_i(t) J_k(0) \rangle dt$$

multi-component systems

e.g. heat transport in a binary system: heat and mass/charge currents interact with each other

$$J_1 = \Lambda_{11} F_1 + \Lambda_{12} F_2$$
$$J_2 = \Lambda_{21} F_1 + \Lambda_{22} F_2$$

$$\Lambda_{ik} \propto \int_0^\infty \langle J_i(t) J_k(0) \rangle dt$$

$$J_1 = \kappa F_1$$
$$(J_2 = 0)$$

$$\kappa = \Lambda_{11} - \frac{\Lambda_{12}^2}{\Lambda_{22}}$$
$$= \frac{1}{(\Lambda^{-1})_{11}}$$

multi-component systems

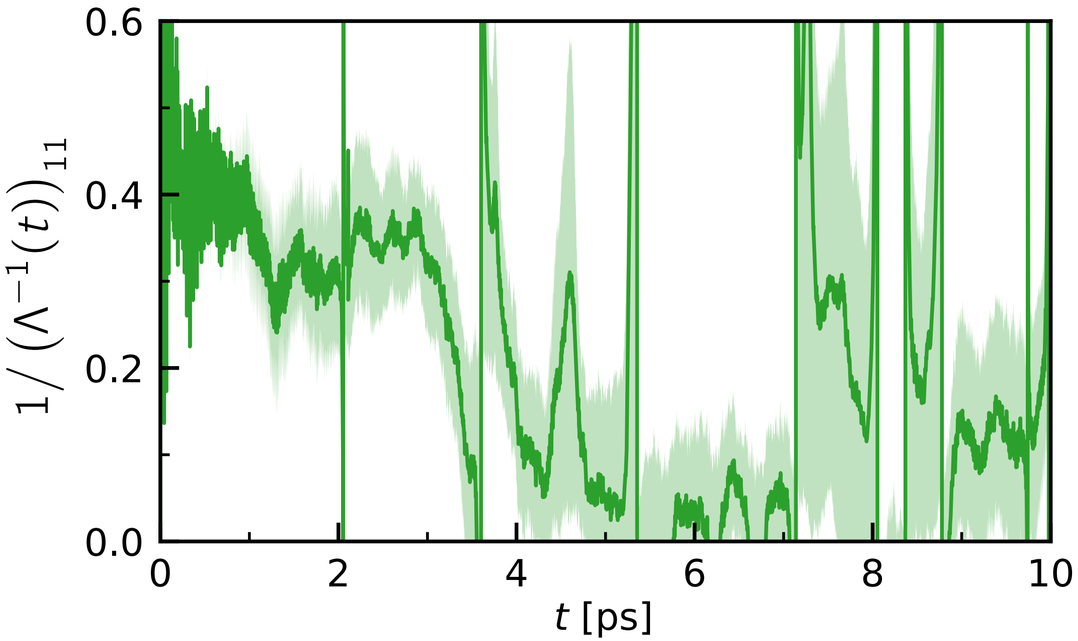
e.g. heat transport in a binary system: heat and mass/charge currents interact with each other

$$J_1 = \Lambda_{11} F_1 + \Lambda_{12} F_2$$
$$J_2 = \Lambda_{21} F_1 + \Lambda_{22} F_2$$

$$\Lambda_{ik} \propto \int_0^\infty \langle J_i(t) J_k(0) \rangle dt$$

$$J_1 = \kappa F_1$$
$$(J_2 = 0)$$

$$\kappa = \Lambda_{11} - \frac{\Lambda_{12}^2}{\Lambda_{22}}$$
$$= \frac{1}{(\Lambda^{-1})_{11}}$$



"whiskey"
50% water-ethanol solution

cepstral analysis of multi-component systems

$$\kappa = \Lambda_{11} - \frac{\Lambda_{12}^2}{\Lambda_{22}}$$
$$= \frac{1}{(\Lambda^{-1})_{11}}$$

Schur complement
of the 22 block in Λ

cepstral analysis of multi-component systems

$$\kappa = \Lambda_{11} - \frac{\Lambda_{12}^2}{\Lambda_{22}} = \frac{1}{(\Lambda^{-1})_{11}}$$

Schur complement of the 22 block in Λ

$$\Lambda_{ik} = \frac{1}{2} S_{ik}(0)$$

$$S_{ik}(\omega) = \int_{-\infty}^{\infty} \langle J_i(t) J(_k(0) \rangle \mathrm{e}^{i\omega t} dt = \lim_{T\to\infty} \left\langle \hat{S}_{T,ik}(\omega) \right\rangle$$

$$\hat{S}_{T,ik}(\omega) = \frac{1}{T} \tilde{J}_{T,i}(\omega) \tilde{J}_{T,k}(-\omega) \sim \mathcal{CW}\big(S_{ik}(\omega), 1, 2\big)$$

cepstral analysis of multi-component systems

$$\kappa = \Lambda_{11} - \frac{\Lambda_{12}^2}{\Lambda_{22}} = \frac{1}{(\Lambda^{-1})_{11}}$$

Schur complement of the 22 block in Λ

$$\Lambda_{ik} = \frac{1}{2} S_{ik}(0)$$

$$S_{ik}(\omega) = \int_{-\infty}^{\infty} \langle J_i(t) J(_k(0) \rangle \mathrm{e}^{i\omega t} dt = \lim_{T\to\infty} \left\langle \hat{S}_{T,ik}(\omega) \right\rangle$$

$$\hat{S}_{T,ik}(\omega) = \frac{1}{T} \tilde{J}_{T,i}(\omega) \tilde{J}_{T,k}(-\omega) \sim \mathcal{CW}\big(S_{ik}(\omega), 1, 2\big)$$

the Schur complement of a Wishart matrix, $\widehat{W}$, is a Wishart matrix whose expectation is the Schur complement $\left\langle \widehat{W} \right\rangle$

cepstral analysis of multi-component systems

$$\kappa = \Lambda_{11} - \frac{\Lambda_{12}^2}{\Lambda_{22}} = \frac{1}{(\Lambda^{-1})_{11}}$$

Schur complement of the 22 block in Λ

$$\Lambda_{ik} = \frac{1}{2} S_{ik}(0)$$

$$S_{ik}(\omega) = \int_{-\infty}^{\infty} \langle J_i(t) J(_k(0) \rangle \mathrm{e}^{i\omega t} dt = \lim_{T\to\infty} \left\langle \hat{S}_{T,ik}(\omega) \right\rangle$$

$$\hat{S}_{T,ik}(\omega) = \frac{1}{T} \tilde{J}_{T,i}(\omega) \tilde{J}_{T,k}(-\omega) \sim \mathcal{CW}\big(S_{ik}(\omega), 1, 2\big)$$

the Schur complement of a Wishart matrix, $\widehat{W}$, is a Wishart matrix whose expectation is the Schur complement $\left\langle \widehat{W} \right\rangle$

do cepstral analysis on the Schur complement!

cepstral analysis of multi-component systems

$$\kappa = \Lambda_{11} - \frac{\Lambda_{12}^2}{\Lambda_{22}} = \frac{1}{(\Lambda^{-1})_{11}}$$

Schur complement of the 22 block in Λ

$$\Lambda_{ik} = \frac{1}{2} S_{ik}(0)$$

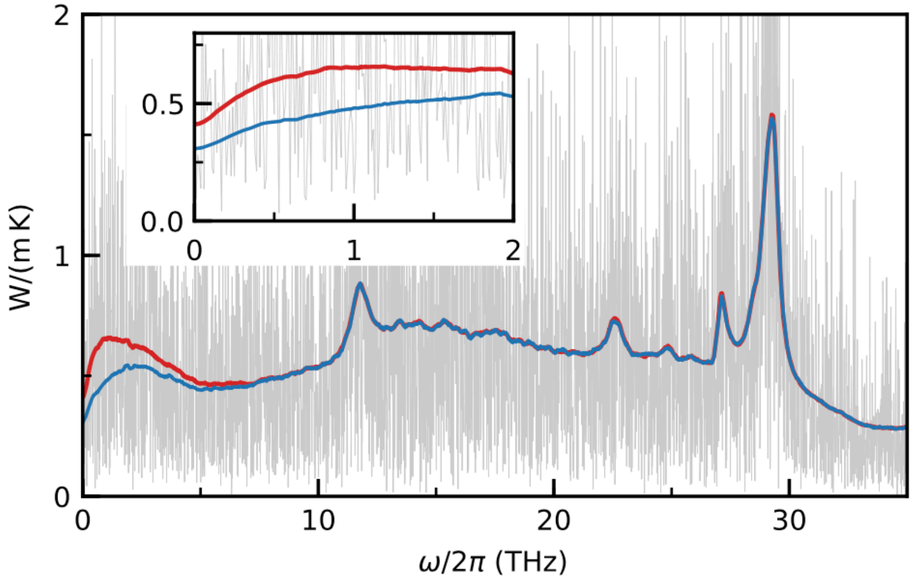
$$S_{ik}(\omega) = \int_{-\infty}^{\infty} \langle J_i(t) J(_k(0) \rangle e^{i\omega t} dt = \lim_{T\to\infty} \left\langle \hat{S}_{T,ik}(\omega) \right\rangle$$

$$\hat{S}_{T,ik}(\omega) = \frac{1}{T} \tilde{J}_{T,i}(\omega) \tilde{J}_{T,k}(-\omega) \sim \mathcal{CW}\big(S_{ik}(\omega), 1, 2\big)$$

the Schur complement of a Wishart matrix, $\widehat{W}$, is a Wishart matrix whose expectation is the Schur complement $\left\langle \widehat{W} \right\rangle$

do cepstral analysis on the Schur complement!

whiskey



cepstral analysis of multi-component systems

$$\kappa = \Lambda_{11} - \frac{\Lambda_{12}^2}{\Lambda_{22}} = \frac{1}{(\Lambda^{-1})_{11}}$$

Schur complement of the 22 block in Λ

$$\Lambda_{ik} = \frac{1}{2} S_{ik}(0)$$

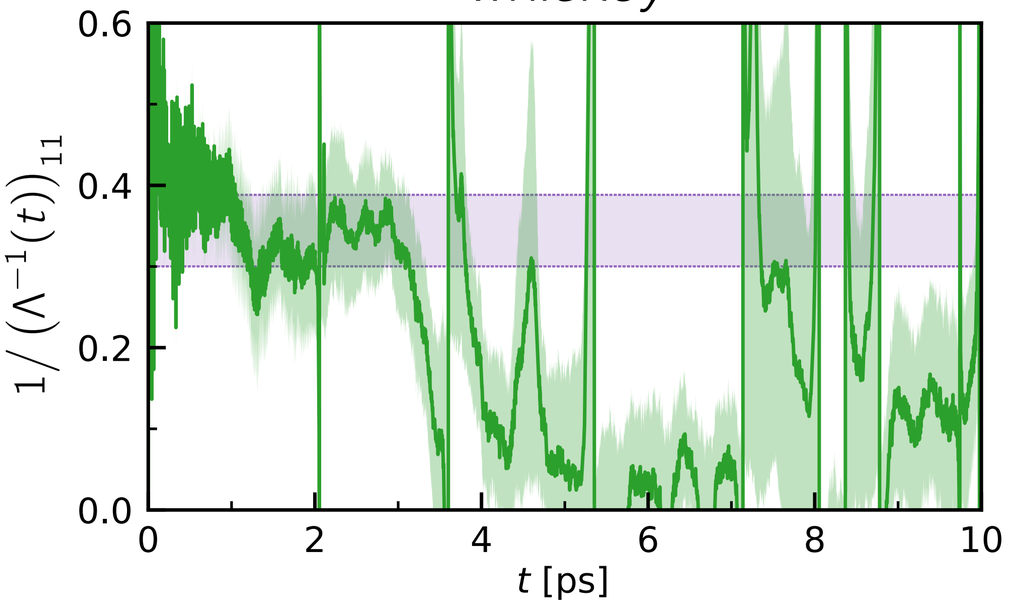
$$S_{ik}(\omega) = \int_{-\infty}^{\infty} \langle J_i(t) J(_k(0) \rangle \mathrm{e}^{i\omega t} dt = \lim_{T\to\infty} \left\langle \hat{S}_{T,ik}(\omega) \right\rangle$$

$$\hat{S}_{T,ik}(\omega) = \frac{1}{T} \tilde{J}_{T,i}(\omega) \tilde{J}_{T,k}(-\omega) \sim \mathcal{CW}\big(S_{ik}(\omega), 1, 2\big)$$

the Schur complement of a Wishart matrix, $\widehat{W}$, is a Wishart matrix whose expectation is the Schur complement $\left\langle \widehat{W} \right\rangle$

do cepstral analysis on the Schur complement!

whiskey



noise reduction in 1-component systems

$$\sigma = \int_0^\infty \langle J_1(t) J_1(0) \rangle dt$$

noise reduction in 1-component systems

$$\sigma = \int_0^\infty \langle J_1(t) J_1(0) \rangle dt$$

$$\int_0^\infty \langle J_2(t) J_2(0) \rangle dt = 0$$

noise reduction in 1-component systems

$$\sigma = \int_0^\infty \langle J_1(t) J_1(0) \rangle dt$$

$$\int_0^\infty \langle J_2(t) J_2(0) \rangle dt = 0$$

$$S_{11}(\omega) \sim 2\sigma + \mathcal{O}(\omega^2)$$
$$S_{22}(\omega) \sim \mathcal{O}(\omega^2)$$
$$S_{12}(\omega) \sim \mathcal{O}(\omega^2)$$

noise reduction in 1-component systems

$$\sigma = \int_0^\infty \langle J_1(t) J_1(0) \rangle dt$$

$$\int_0^\infty \langle J_2(t) J_2(0) \rangle dt = 0$$

$$S_{11}(\omega) \sim 2\sigma + \mathcal{O}(\omega^2)$$
$$S_{22}(\omega) \sim \mathcal{O}(\omega^2)$$
$$S_{12}(\omega) \sim \mathcal{O}(\omega^2)$$

$$S_{11}(\omega) - \frac{S_{12}(\omega)^2}{S_{22}(\omega)} \sim 2\sigma + \mathcal{O}(\omega^2) < S_{11}(\omega)$$

noise reduction in 1-component systems

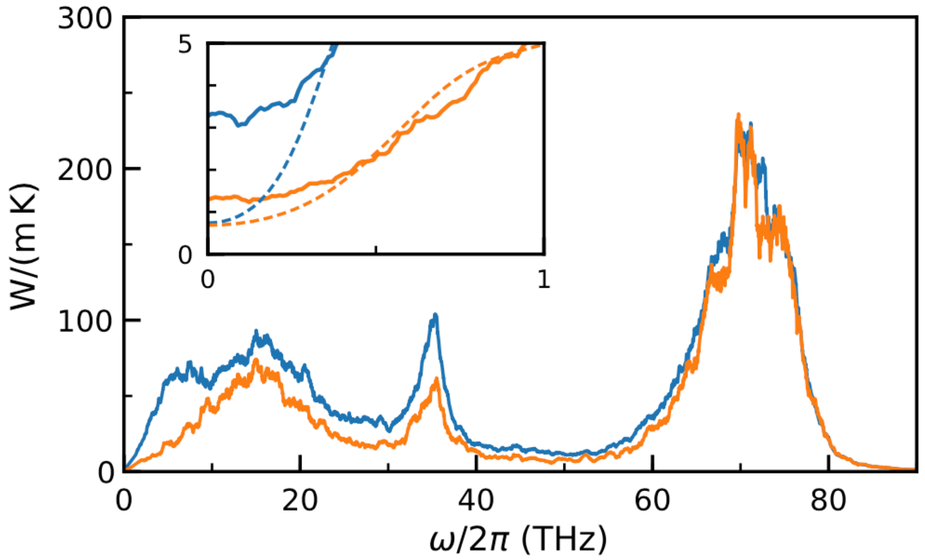
$$\sigma = \int_0^\infty \langle J_1(t) J_1(0) \rangle dt$$

$$\int_0^\infty \langle J_2(t) J_2(0) \rangle dt = 0$$

$$S_{11}(\omega) \sim 2\sigma + \mathcal{O}(\omega^2)$$
$$S_{22}(\omega) \sim \mathcal{O}(\omega^2)$$
$$S_{12}(\omega) \sim \mathcal{O}(\omega^2)$$

$$S_{11}(\omega) - \frac{S_{12}(\omega)^2}{S_{22}(\omega)} \sim 2\sigma + \mathcal{O}(\omega^2) < S_{11}(\omega)$$

heat transport in ab-initio water



J_{DFT},out of scale

$J_{DFT} + J_O$

$J_{DFT} + J_O + J_e$

——— running average

--------- cepstral analysis

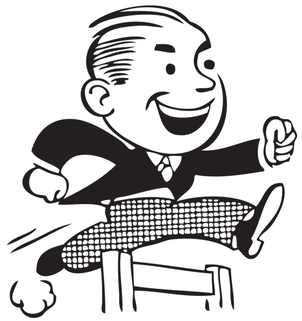
hurdles toward an ab initio Green-Kubo theory

PRL **104,** 208501 (2010) PHYSICAL REVIEW LETTERS week ending 21 MAY 2010

Thermal Conductivity of Periclase (MgO) from First Principles

Stephen Stackhouse, Lars Stixrude, and Bijaya B. Karki

sensitive to the form of the potential. The widely used Green-Kubo relation [14] does not serve our purposes, because in first-principles calculations it is impossible to uniquely decompose the total energy into individual contributions from each atom.

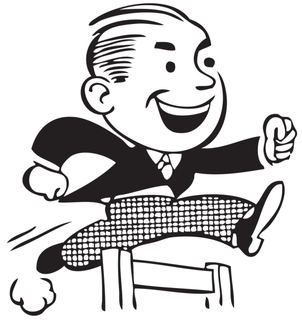


PRL **118,** 175901 (2017) PHYSICAL REVIEW LETTERS week ending 28 APRIL 2017

***Ab Initio* Green-Kubo Approach for the Thermal Conductivity of Solids**

Christian Carbogno, Rampi Ramprasad, and Matthias Scheffler

ulations: Because of the limited time scales accessible in aiMD runs, thermodynamic fluctuations dominate the HFACF, which in turn prevents a reliable and numerically stable assessment of the thermal conductivity via Eq. (2).



conclusions

conclusions

- a newly discovered gauge invariance principle for transport coefficients makes it possible to formulate a consistent density-functional or, more generally, quantum theory of adiabatic heat transport;

conclusions

- a newly discovered gauge invariance principle for transport coefficients makes it possible to formulate a consistent density-functional or, more generally, quantum theory of adiabatic heat transport;

- the straightforward application of the Green-Kubo theory of linear response to *ab initio* molecular dynamics is numerically challenging, in that it would require sampling trajectories of several nano-seconds;

conclusions

- a newly discovered gauge invariance principle for transport coefficients makes it possible to formulate a consistent density-functional or, more generally, quantum theory of adiabatic heat transport;
- the straightforward application of the Green-Kubo theory of linear response to *ab initio* molecular dynamics is numerically challenging, in that it would require sampling trajectories of several nano-seconds;
- cepstral analysis provides a rigorous and practical means of exploiting the information on the spectral properties of the current being sampled, to evaluate transport coefficients and their statistical uncertainties with MD simulations of the order of a few hundred ps;

conclusions

- a newly discovered gauge invariance principle for transport coefficients makes it possible to formulate a consistent density-functional or, more generally, quantum theory of adiabatic heat transport;
- the straightforward application of the Green-Kubo theory of linear response to *ab initio* molecular dynamics is numerically challenging, in that it would require sampling trajectories of several nano-seconds;
- cepstral analysis provides a rigorous and practical means of exploiting the information on the spectral properties of the current being sampled, to evaluate transport coefficients and their statistical uncertainties with MD simulations of the order of a few hundred ps;
- cepstral analysis naturally applies to multi-component systems, which would be simply intractable by a straightforward Green-Kubo approach;

conclusions

- a newly discovered gauge invariance principle for transport coefficients makes it possible to formulate a consistent density-functional or, more generally, quantum theory of adiabatic heat transport;
- the straightforward application of the Green-Kubo theory of linear response to *ab initio* molecular dynamics is numerically challenging, in that it would require sampling trajectories of several nano-seconds;
- cepstral analysis provides a rigorous and practical means of exploiting the information on the spectral properties of the current being sampled, to evaluate transport coefficients and their statistical uncertainties with MD simulations of the order of a few hundred ps;
- cepstral analysis naturally applies to multi-component systems, which would be simply intractable by a straightforward Green-Kubo approach;
- multi-component analysis allows one to considerably reduce the statistical noise even in 1-component systems;

conclusions

- a newly discovered gauge invariance principle for transport coefficients makes it possible to formulate a consistent density-functional or, more generally, quantum theory of adiabatic heat transport;
- the straightforward application of the Green-Kubo theory of linear response to *ab initio* molecular dynamics is numerically challenging, in that it would require sampling trajectories of several nano-seconds;
- cepstral analysis provides a rigorous and practical means of exploiting the information on the spectral properties of the current being sampled, to evaluate transport coefficients and their statistical uncertainties with MD simulations of the order of a few hundred ps;
- cepstral analysis naturally applies to multi-component systems, which would be simply intractable by a straightforward Green-Kubo approach;
- multi-component analysis allows one to considerably reduce the statistical noise even in 1-component systems;
- further work in model selection is desirable to improve the optimal estimate of the number of cepstral coefficients.

supported by:

QUANTUMESPRESSO

http://www.quantum-espresso.org

QUANTUM ESPRESSO FOUNDATION

http://foundation.quantum-espresso.org

http://www.max-centre.eu

thanks to:

Aris Marcolongo

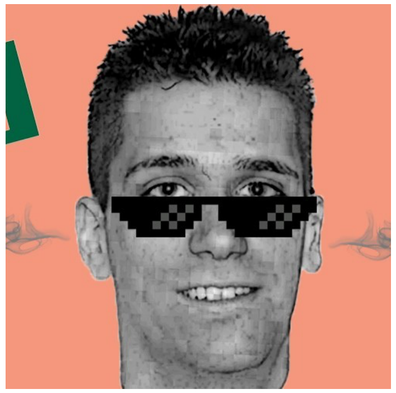
Cesare Malosso

Federico Grasselli

Loris Ercole

Riccardo Bertossa

Paolo Pegolo



Davide Tisi

nature physics

ARTICLES

PUBLISHED ONLINE: 19 OCTOBER 2015 | DOI: 10.1038/NPHYS3509

Microscopic theory and quantum simulation of atomic heat transport

Aris Marcolongo[1], Paolo Umari[2] and Stefano Baroni[1]*

SCIENTIFIC REPORTS

SCIENTIFIC REPORTS | 7: 15835 | DOI:10.1038/s41598-017-15843-2

Accurate thermal conductivities from optimally short molecular dynamics simulations

Loris Ercole[1], Aris Marcolongo[2] & Stefano Baroni[1]

PHYSICAL REVIEW LETTERS 122, 255901 (2019)

Theory and Numerical Simulation of Heat Transport in Multicomponent Systems

Riccardo Bertossa,[1] Federico Grasselli,[1] Loris Ercole,[1,*] and Stefano Baroni[1,2,†]

Heat Transport in Insulators from Ab Initio Green-Kubo Theory

35

Stefano Baroni, Riccardo Bertossa, Loris Ercole, Federico Grasselli, and Aris Marcolongo

© Springer Nature Switzerland AG 2020

W. Andreoni, S. Yip (eds.), *Handbook of Materials Modeling*,
https://doi.org/10.1007/978-3-319-44680-6_12

809

these slides soon available at
http://talks.baroni.me