Coherence of phonons reveals the wave-particle duality of heat conduction

Benoit Latour
Division of Engineering and Applied Science, California Institute of Technology, Pasadena, California 91125, USA and Laboratoire EM2C, CNRS, CentraleSupélec, Université Paris-Saclay, Grande Voie des Vignes, 92295 Châtenay-Malabry, France

Yann Chalopin*
Laboratoire EM2C, CNRS, CentraleSupélec, Université Paris-Saclay, Grande Voie des Vignes, 92295 Châtenay-Malabry, France

RELATION BETWEEN THE CROSS-SPECTRAL DENSITY FUNCTION AND THE SPECTRAL ENERGY DENSITY

In this first section, we have stated that the spatial Fourier transform of $T(\omega,\vec{R})$ corresponds the kinetic energy of the phonon mode $(\omega,\vec{k})$. $\vec{\dot{q}}(\omega,\vec{k},b)$ is the spatial Fourier transform of $\vec{v}(\omega,\vec{R}_i^0,b)$:

$$\vec{\dot{q}}(\omega,\vec{k},b)=\frac{1}{\sqrt{V}}\sum_{\vec{R}_i^0}\vec{v}(\omega,\vec{R}_i^0,b)e^{i\vec{k}\cdot\vec{R}_i^0} \tag{1}$$

and reciprocally

$$\vec{v}(\omega,\vec{R}_i^0,b)=\frac{1}{\sqrt{V}}\sum_{\vec{k}}\vec{\dot{q}}(\omega,\vec{k},b)e^{-i\vec{k}\cdot\vec{R}_i^0}, \tag{2}$$

with V the volume of the system.

Using Lattice Dynamics theory[1], we can find the eigenmodes of the system, $\vec{e}(\vec{k},b,\nu)$, which correspond to the mode defined by a wave vector $\vec{k}$ and a branch ν of the atom b in the unit cell. The normal mode coordinate $\dot{q}(\omega,\vec{k},\nu)$ allows to decouple all the eigenmodes of the system.

$$\dot{q}(\omega,\vec{k},\nu)=\sum_b\sqrt{m_b}\,\vec{e}^*(\vec{k},b,\nu)\cdot\vec{\dot{q}}(\omega,\vec{k},b). \tag{3}$$

The orthonormality of the eigenvectors easily leads to the following relation:

$$\vec{\dot{q}}(\omega,\vec{k},b)=\sum_\nu\frac{1}{\sqrt{m_b}}\,\dot{q}(\omega,\vec{k},\nu)\vec{e}(\vec{k},b,\nu). \tag{4}$$

Using Equations 2 and 4, the cross-spectral density function becomes

$$\begin{aligned}T(\omega,\vec{R})&=\frac{1}{2}\sum_b m_b\left\langle\frac{1}{V}\sum_{\vec{k},\vec{k}'}\vec{\dot{q}}^*(\omega,\vec{k},b)e^{i\vec{k}\cdot\vec{R}_i^0}\vec{\dot{q}}(\omega,\vec{k}',b)e^{-i\vec{k}'\cdot(\vec{R}^0+\vec{R})}\right\rangle_{\vec{R}^0}\\&=\frac{1}{2}\sum_b m_b\left\langle\frac{1}{V}\sum_{\vec{k},\vec{k}'}\sum_{\nu,\nu'}\frac{1}{m_b}\dot{q}^*(\omega,\vec{k},\nu)\vec{e}^*(\vec{k},b,\nu)e^{i\vec{k}\cdot\vec{R}_i^0}\dot{q}(\omega,\vec{k}',\nu')\vec{e}(\vec{k}',b,\nu')e^{-i\vec{k}'\cdot(\vec{R}^0+\vec{R})}\right\rangle_{\vec{R}^0}\\&=\frac{1}{2}\frac{1}{V}\sum_{\vec{k},\vec{k}'}\sum_{\nu,\nu'}\dot{q}^*(\omega,\vec{k},\nu)\dot{q}(\omega,\vec{k}',\nu')\left(\sum_b\vec{e}^*(\vec{k},b,\nu)\vec{e}(\vec{k}',b,\nu')\right)\left\langle e^{i(\vec{k}-\vec{k}')\cdot\vec{R}_i^0}\right\rangle_{\vec{R}^0}e^{-i\vec{k}'\cdot\vec{R}}.\end{aligned} \tag{5}$$

As $\left\langle e^{i(\vec{k}-\vec{k}')\cdot\vec{R}_i^0}\right\rangle_{\vec{R}^0}=\delta_{\vec{k}\vec{k}'}V$, considering the orthonormality of the eigenvectors for a given wave vector, Equation 5 becomes

$$\begin{aligned}T(\omega,\vec{R})&=\frac{1}{2}\sum_{\vec{k}}\sum_\nu\sum_{\nu'}\dot{q}^*(\omega,\vec{k},\nu)\dot{q}(\omega,\vec{k},\nu')\delta_{\nu\nu'}e^{-i\vec{k}\cdot\vec{R}}\\&=\sum_{\vec{k}}\left(\sum_\nu\frac{1}{2}|\dot{q}(\omega,\vec{k},\nu)|^2\right)e^{-i\vec{k}\cdot\vec{R}}.\end{aligned} \tag{6}$$

Finally, $T(\omega,\vec{R})$ is the spatial Fourier transform of the kinetic energy $T(\omega,\vec{k})$ of the phonon mode $(\omega,\vec{k})$. As the spatial dependence of $T(\omega,\vec{R})$ is a attenuated wave, the spatial Fourier transform leads to a Lorentzian profile along the wave vector axis of $T(\omega,\vec{k})$. The imaginary part of the wave vector is extracted as the width of $T(\omega,\vec{k})$. As $l_c = 1/(2\mathrm{Im}k)$, we have checked that both estimators give the same results.

CONDITIONS WHERE THE MEAN FREE PATH AND THE SPATIAL COHERENCE LENGTH COINCIDE

The following calculation has been proposed in other research areas such as optics[2, 3]. We will derive it for thermal phonons. Let $f(\omega,k)=0$ be the dispersion relation of the considered material, where ω and k are both complex:

$$\omega = \omega_0 + \Omega + i\mathrm{Im}(\omega), \tag{7}$$

$$k = k_0 + \kappa + i\mathrm{Im}(k), \tag{8}$$

where (ω_0,k_0) stands for the eigenmodes of the dynamical matrix and f is smooth enough to be assumed of class $\mathbf{C^n}$. By making the assumption $|\Omega + i\mathrm{Im}(\omega)| \ll \omega_0$ and $|\kappa + i\mathrm{Im}(k)| \ll k_0$, a Taylor expansion at the first order of the dispersion relation leads to:

$$\begin{aligned} f(\omega,k) = f(\omega_0,k_0) &+ (\Omega + i\mathrm{Im}(\omega))\frac{\partial f}{\partial \omega}(\omega_0,k_0) \\ &+ (\kappa + i\mathrm{Im}(k))\frac{\partial f}{\partial k}(\omega_0,k_0). \end{aligned} \tag{9}$$

As ω and k are close to ω_0 and k_0, f also represents the dispersion relation in the harmonic case, so $f(\omega_0,k_0)=0$. Moreover, if Ω and κ are respectively assumed to be negligible compared to $\mathrm{Im}(\omega)$ and $\mathrm{Im}(k)$, Equation 9 becomes

$$0 = (i\mathrm{Im}(\omega))\frac{\partial f}{\partial \omega}(\omega_0,k_0) + (i\mathrm{Im}(k))\frac{\partial f}{\partial k}(\omega_0,k_0) \tag{10}$$

The group velocity $v_g(\omega_0,k_0)$ is defined as:

$$\begin{aligned} v_g(\omega_0,k_0) &= \frac{\partial \omega}{\partial k}(\omega_0,k_0) \\ &= -\frac{\partial f}{\partial k}(\omega_0,k_0)/\frac{\partial f}{\partial \omega}(\omega_0,k_0). \end{aligned} \tag{11}$$

Finally, Equation 10 leads to

$$v_g(\omega_0,k_0) = \frac{\partial \omega_0}{\partial k_0} = \frac{\mathrm{Im}(\omega)}{\mathrm{Im}(k)}. \tag{12}$$

$\mathrm{Im}(\omega)$ is related to the phonon relaxation time τ and $\mathrm{Im}(k)$ to the spatial coherence length l_c. Finally, we conclude that when $\Omega \ll \mathrm{Im}(\omega) \ll \omega_0$ and $\kappa \ll \mathrm{Im}(k) \ll k_0$, the mean free path Λ is equal to the spatial coherence length l_c.

LEGENDS FOR THE MOVIES

Movie wave_packet_A_ballistic_coherent:

Coherent ballistic transport in superlattices: the wave packet (wave packet A) spreading over tens of periods remains unaffected by the interface scattering.

Movie wave_packet_B_interface_scattering:

Interface scattering: the optical mode (wave packet B) exhibits a much smaller coherence length close to d_{SL}. Propagating from right to left (negative group velocity), it undergoes reflection and transmission at the interfaces.

Movie wave_packet_C_standing_mode:

Standing mode at the zone edge (wave packet C): with a coherence length much larger than d_{SL}, the simulation clearly reveals the behavior originating from the sum of two counter-propagative modes.

[1] J. M. Ziman, *Electrons and phonons: the theory of transport phenomena in solids*, Vol. 20 (Clarendon Press, 1960).

[2] A. Krokhin and P. Halevi, Physical Review B **53**, 1205 (1996).
[3] K. C. Huang, E. Lidorikis, X. Jiang, J. D. Joannopoulos, K. A. Nelson, P. Bienstman, and S. Fan, Physical Review B **69** (2004).