On-line Supplemental Material for
Phonon thermal conductivity by non-equilibrium molecular dynamics using non-locality

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This supplement contains blah blah.

I. HEAT EXCHANGE ALGORITHM

The Müller-Plathe recipe is: find the hottest atom in the cold ($\ell = N_S/2$) slab, and the coldest atom in the hot ($\ell = 0$) slab. Interchange their velocities. Energy and momentum are both conserved, and the system is driven from equilibrium in a way that must be monitored. Cao and Li[1] among others, have suggested modified algorithms of this type. Furtado, Abreu, and Tavares (FAT) devised a gentler and more versatile method. An earlier version of the present paper, which was was posted on the arXiv[2], derived the same procedure. We call it the FAT algorithm, because Furtado *et al.* discovered it simultaneously and published it first.

In our computation, driving is done by spatially periodic injection and simultaneous removal of heat. Examine a slab ℓ (with $0 \leq \ell < N_S/4$), and its conjugate slab $N_S/2 - \ell$. The former is "hotter" than the latter because $\dot{e}(\ell) > 0 > \dot{e}(N_S/2 - \ell) = -\dot{e}(\ell)$. Find the coldest atom (meaning least kinetic energy) of all atoms of mass m_i in the hotter slab, denoting its velocity as $\vec{v}_C$. Find the hottest atom of the same mass in the colder slab, denoting its velocity $\vec{v}_H$. Given the large fluctuations of the Maxwell-Boltzmann ensemble, it is certain that $v_H^2 > v_C^2$. Choose an appropriate velocity $\vec{w}$ and add it to the velocity $\vec{v}_C$ and subtract it from $\vec{v}_H$:

$$\vec{v}_H \to \vec{v}_H - \vec{w}$$
$$\vec{v}_C \to \vec{v}_C + \vec{w} \tag{1}$$

The same operation should be done for the pair of slabs $-\ell$ and $\ell - N_S/2$. All slabs can be done simultaneously, or different random times can be used for different slabs.

There are three criteria for an appropriate $\vec{w}$, which uniquely fix the desired choice. (i) The cold atom's kinetic energy should increase by Δ, an energy that can be specified in advance as $(\dot{e}\Omega_S\tau)\cos(2\pi\ell/N_S)$, where τ is the average time interval between random interventions, and Ω_S is the volume of a slab. (ii) The hot atom's kinetic energy should decrease by Δ. Then both momentum and energy are conserved. Heating has the desired form $\dot{e}\cos(2\pi\ell/N_S)$ with $\dot{e}$ chosen not too different

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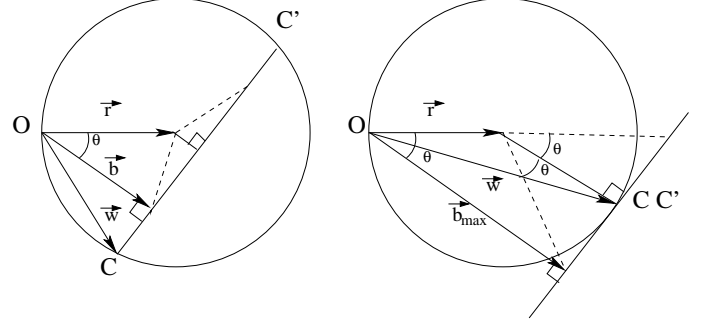[b)] Electronic mail: yerong.li@outlook.com

FIG. 1. Geometric construction for finding the smoothest velocity change $\vec{w}$ of the coldest atoms (with velocity $\vec{v}_C$ in the hotter region and the hottest atoms (with velocity $\vec{v}_H$ in the colder region. Both figures represent a sphere of radius $\vec{r} = (\vec{v}_C - \vec{v}_H)/2$. The plane perpendicular to $\vec{b}$, intersects the sphere in a circle, which is the locus of solutions $\vec{w}$ obeying energy and momentum conservation rules. The points C and C' (in the $\vec{r}$–$\vec{b}$ plane) give the solutions with least and greatest impulse. The right-hand version shows the largest vector $\vec{b}$ which allows solutions for $\vec{w}$. The minimum and maximum impulse solutions have merged to a point.

from $k_B T/\Omega_S\tau$. Trial calculations should test for the best choices of $\dot{e}$ and τ. (iii) There is still a one-dimensional family of vectors $\vec{w}$; from these, choose the smallest $|\vec{w}|$, which gives the least impulse to the affected atoms. Except for the sinusoidal spatial variation of Δ, these are exactly the criteria chosen by Furtado *et al.*. Their implementation of these criteria is also exactly like ours. We present our derivation here for the convenience of the reader.

The energy shift criteria (i) and (ii) give equations $-2\vec{v}_H \cdot \vec{w} + w^2 = -2\Delta/m \equiv -\delta$, and $2\vec{v}_C \cdot \vec{w} + w^2 = +\delta$. Adding and subtracting these equations give

$$\delta = (\vec{v}_H + \vec{v}_C) \cdot \vec{w}$$
$$w^2 = (\vec{v}_H - \vec{v}_C) \cdot \vec{w}, \tag{2}$$

a linear and a quadratic equation for $\vec{w}$. These have a simple geometric interpretation shown in Fig. 1. The first equation restricts the projection of $\vec{w}$ along the vector $\vec{v}_H + \vec{v}_C$. Geometrically, this means that $\vec{w}$ lies on the plane (shown by C, C' in Fig. 1) perpendicular to the vector $\vec{b} \equiv \delta(\vec{v}_C + \vec{v}_H)/|\vec{v}_C + \vec{v}_H|^2$, where the origin of

$\vec{w}$ coincides with the origin of $\vec{b}$. The second equation restricts $\vec{w}$ to the surface of a sphere of radius $|\vec{v}_H - \vec{v}_C|/2$, centered at the end of the vector $\vec{r} \equiv (\vec{v}_H - \vec{v}_C)/2$, whose origin also coincides with the origin of $\vec{w}$. The sphere and the plane intersect on a circle, indicated by C, C' in Fig. 1. This circle is the one-dimensional family of solutions $\vec{w}$ satisfying Eqs.(2). It is also clear from the geometry that the shortest vector $\vec{w}$ (the one that satisfies criterion (iii)) is the one shown, from O to C. This lies in the same plane as the two known vectors $\vec{r}$ and $\vec{b}$ (also the same plane as $\vec{v}_H$ and $\vec{v}_C$). Therefore

$$\vec{w} = \alpha\vec{r} + \beta\vec{b}$$
$$\vec{r} = (\vec{v}_H - \vec{v}_C)/2$$
$$\vec{b} = \frac{\delta(\vec{v}_H + \vec{v}_C)}{|\vec{v}_H + \vec{v}_C|^2}. \tag{3}$$

These definitions allow Eqs.(2) to be written as

$$b^2 = \vec{w} \cdot \vec{b}$$
$$w^2 = 2\vec{r} \cdot \vec{w}. \tag{4}$$

The solution for $\vec{w}$ is

$$\beta = 1 - \alpha\frac{\vec{r} \cdot \vec{b}}{b^2}$$
$$\alpha = 1 - \sqrt{1 - X}$$
$$X = \frac{(b^2 - 2\vec{b} \cdot \vec{r})b^2}{b^2 r^2 - (\vec{b} \cdot \vec{r})^2} \tag{5}$$

To derive this, substitute Eq.(3) for $\vec{w}$ in terms of the unknown coefficients α and β into the Eqs.(2). The linear equation is used to find β in terms of α. Eliminating β in favor of α in the quadratic equation gives a quadratic equation for α. The appropriate solution is displayed in Eq.(5). An alternate version directly in terms of the velocities $\vec{v}_H$ and $\vec{v}_C$ is

$$X = (2\Delta/m)\frac{(2\Delta/m) - (v_H^2 - v_C^2)}{v_H^2 v_C^2 - (\vec{v}_H \cdot \vec{v}_C)^2} \tag{6}$$

$$\vec{w} = \frac{\alpha}{2}(\vec{v}_H - \vec{v}_C) + \left[\frac{2\Delta}{m} - \frac{\alpha}{2}(v_H^2 - v_C^2)\right]\frac{\vec{v}_H + \vec{v}_C}{|\vec{v}_H + \vec{v}_C|^2} \tag{7}$$

Notice that X in Eq.(6) has a non-negative denominator that becomes zero in an accidental event where $\vec{v}_H$ and $\vec{v}_C$ are parallel; X is then ill-defined, because no solution exists. An alternate pair of C and H atoms must be chosen. In the simulations reported in subsequent sections, we find that Δ should be chosen small, making the numerator of X in Eq.(6) negative. Thus both X and α are negative, contrary to the version shown in Fig. 1. This does not adversely affect anything.

There is a second solution, $\alpha = 1 + \sqrt{1 - X}$, corresponding to the maximum $|\vec{w}|$, designated as C' in Fig. 1. For $|\vec{b}| > b_{\max}$, there are no real solutions. This corresponds to $X > 1$. The condition for the two solutions to coincide is $X = 1$, which agrees with $b_{\max} = r(1 + \cos\theta)$, where θ is the angle between $\vec{b}$ and $\vec{r}$. This can be understood from the right hand part of Fig. 1, illustrating the case where the circle collapses to a point. For reasonable choices of the parameter $m\delta/2 = \Delta$, meaning values smaller than or similar to $k_B T/N_S$, solutions should always exist.

II. DEBYE-RTA MODEL, ESPECIALLY $p=2$

Appendix A gives formulas in the Debye-RTA model. Here are details. Equation 13 of ref. 4 is

$$\kappa_{\mathrm{RTA}}(q) = \frac{k_B}{\Omega}\sum_Q\frac{v_{Qx}^2}{1/\tau_Q + iqv_{Qx}}. \tag{8}$$

This is the classical limit of the solution of the Boltzmann equation in the relaxation time approximation (RTA). Now make a Debye model, and model the scattering rate as $1/\tau_Q = (1/\tau_D)(Q/Q_D)^p$. Multiplying numerator and denominator by τ_D, and using $x = Q/Q_D$, $\mu = v_x/v = \cos\theta$, and $\lambda = qv$, this generates Eq.(A1),

$$\tilde{\kappa}_p(q) = \frac{9}{2}\kappa_0\int_0^1 dx x^2\int_{-1}^1 d\mu\frac{\mu^2}{x^p + i\lambda\mu}, \tag{9}$$

where $\kappa_0 = Nk_B v^2\tau/\Omega$. One way to perform the integrations is to do the μ-integral first.

$$\frac{\tilde{\kappa}_p(q)}{\kappa_0} = \frac{9}{\lambda}\int_0^1 dx x^2\left[\frac{x^p}{\lambda} - \left(\frac{x^p}{\lambda}\right)^2\tan^{-1}\left(\frac{\lambda}{x^p}\right)\right]. \tag{10}$$

The most important case is $p =$, and also the trickiest to integrate further. Using $u = x^2$, this can be written as

$$\frac{\tilde{\kappa}_p(q)}{\kappa_0} = \frac{9}{2\lambda^2}\int_0^1 du u^{3/2}\left[1 - \frac{u}{\lambda}\cot^{-1}\frac{u}{\lambda}\right]. \tag{11}$$

This uses $\tan^{-1}(1/x) = \cot^{-1}x$. The answer can be written as

$$\frac{\tilde{\kappa}_p(q)}{\kappa_0} = \frac{9}{7\lambda^2}[h(1) - h(0)] \tag{12}$$

where $h(u)$ is

$$h(u) = \frac{7}{2}\int du\left[u^{3/2} - \frac{u^{5/2}}{\lambda}\cot^{-1}\frac{u}{\lambda}\right]$$
$$= u^{5/2} + 2\lambda^2 u^{1/2} - \frac{u^{7/2}}{\lambda}\cot^{-1}\frac{u}{\lambda}$$
$$-\frac{\lambda^{5/2}}{\sqrt{2}}\left[\frac{1}{2}\log\left(\frac{u + \sqrt{2u\lambda} + \lambda}{u - \sqrt{2u\lambda} + \lambda}\right) + \tan^{-1}\left(\frac{\sqrt{2u\lambda}}{\lambda - u}\right)\right] \tag{13}$$

It is necessary to be careful in evaluating $h(1) - h(0)$. As we integrate u from 0 towards 1 (as in Eq.??), all

the terms in the integral, Eq.13 have smooth behavior except the last term, $\tan^{-1}[\sqrt{(2u\lambda)}/(\lambda - u)]$. Note that $0 < \lambda < 1$ since L is large. Therefore, in this term, the argument of the arctangent increases from zero to $+\infty$, where the arctangent equals $\pi/2$, then switches to $-\infty$, with the arctangent still equal to $\pi/2$, and finally increases toward 0 from the negative side, with the arctangent approaching π from the negative side. Therefore, the value of $\tan^{-1}[\sqrt{(2u\lambda)}/(\lambda - u)]$ at $u = 1$ is $\pi - \tan^{-1}[\sqrt{(2\lambda)}/(1 - \lambda)]$. The answer is thus

$$
\frac{7\lambda^2}{9\kappa_0}\tilde{\kappa}(q_{\min}) = 1 + 2\lambda^2 - \frac{\tan^{-1}\lambda}{\lambda}
$$
$$
-\frac{\lambda^{5/2}}{\sqrt{2}}\left[\frac{1}{2}\log\left(\frac{1 + \sqrt{2\lambda} + \lambda}{1 - \sqrt{2\lambda} + \lambda}\right) + \pi - \tan^{-1}\left(\frac{\sqrt{2\lambda}}{1 - \lambda}\right)\right]
$$
(14)

This does not look the same as Eq.(A4). Deal with this later!

This result confirms the conjecture of Refs. 3 and 4 that numerical result might be best extrapolated by plotting κ *versus* $\sqrt{(1/L)}$.

III. ACKNOWLEDGEMENTS

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