THERMAL RESPONSE OF SILICON CRYSTAL TO PICO-FEMTOSECOND HEAT PULSE BY MOLECULAR DYNAMICS

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INTRODUCTION

Lasers flashing pico to femtosecond pulses are now current laboratory facilities and clock periods of today's electronic microdevices reach a few hundredths of picosecond. The involved heat conduction regimes are clearly out of equilibrium and have generated much interest for four decades as emphasized in literature [1]. The basic reformulation of Fourier's Law consists in adding an inertia term which allows to derive the well known hyperbolic heat conduction equation:

$$\tau \frac{\partial^2 T}{\partial t^2} + \frac{\partial T}{\partial t} = \alpha \, \Delta T \qquad (0)$$

where α represents the thermal diffusivity and τ the heat flux relaxation time. Eq. (0) however leads to a paradox to entropy principle and solving Boltzmann Equation seems inevitable [2,3]. A simpler approach was nevertheless recently proposed based on the heat flux decomposition into ballistic and diffusive contributions [5].

We report a direct way to derive the heat flux response to an ultra-short temperature pulse generated in bulk Si crystals. While classical approach implies insensitivity of initial heat flux to pulse duration, we predict an linear dependence between both quantities. We investigate the relevance of this result based on molecular dynamics (MD) simulations performed in Si crystal showing accordance with predicted heat fluxes.

METHOD

Linear response theory defines the thermal susceptibility as proportional to the autocorrelation function of heat flux fluctuations at equilibrium $\langle q_0(0)q_0(t)\rangle$ [4,5]. The heat flux $\langle q(t)\rangle$ generated by a thermal time-dependent excitation e(t) can then be expressed as:

$$\left\langle q(t) \right\rangle = \frac{V}{3.k_B T^2} \left\langle q_0(0) q_0(t) \right\rangle \otimes e(t) \qquad (0)$$

Where V is the sample volume, k_B the Boltzmann constant and T the equilibrium temperature. A simple but relevant modeling of the autocorrelation function is a decaying exponential with characteristic time equals to phonon relaxation time τ. This approximation leads to the following non-dimensioned heat flux response:

$$\left\langle q(t)\right\rangle^{*} = e^{-\frac{t}{\tau}}.e^{-\frac{\beta^2}{4\tau^2}}\beta \qquad (2)$$

β being the pulse duration.

We show how to retrieve this behavior from molecular dynamics simulations. In MD computations, all the particle trajectories (positions and velocities $\mathbf{r}_i$ and $\mathbf{v}_i$) for a given system with a reasonably small number of atoms are calculated, which allows the derivation of the thermodynamic quantities of the system. The heat flux expression is derived from different formalisms (mechanical, spectral…), leading all to the same expression for the computed two and three-body potentials:

$$\mathbf{q}(t) = \frac{1}{V}\left[\sum_{i=1}^{N}\sum_{j=1,j\neq i}^{N}\left(\frac{1}{2}\mathbf{r}_{ij}.\left(\mathbf{F}_{ij}.\mathbf{v}_i\right)+\frac{1}{6}\sum_{k=1,k\neq i,j}^{N}\left(\mathbf{r}_{ij}+\mathbf{r}_{ik}\right).\left(\mathbf{F}_{ijk}.\mathbf{v}_i\right)\right)\right], \qquad (3)$$

where $\mathbf{F}_{ij}$ and $\mathbf{F}_{ijk}$ are the two and three-body forces and V is the system volume. Eq. (3) only includes positions, velocities and forces that are directly computed in basic MD simulations. Direct calculation of Eq. (0) can therefore be performed. The autocorrelation function was computed as indicated in Fig. (1) and numerical convolution allowed to obtain the thermal response to heat pulses with different durations.

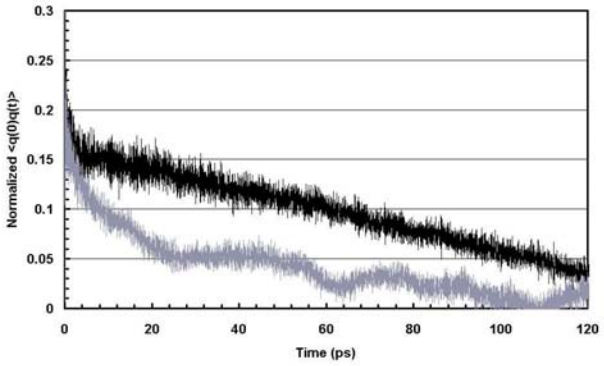

Figure 1: Heat flux autocorrelation function in a Si crystal at 200K (black line) and 500K (grey line)

CONCLUSION

In conclusion, we have show that the response to pico-femto laser pulses implies disagreements with classical predictions. Depending on the excitation duration, the ability of the medium to conduct heat can be reduced by two orders of magnitude. MD appears as a very unique method providing information on the crystal behaviour when femtoscale thermal perturbations are applied.

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