Thermal diodes as an effective way on controlling heat flow in biological environments

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The pioneering work by Li et al., [1], introduced the idea of fabricating materials with spatial inhomogeneities, in order to produce thermal diodes. By using the simplest possible bead-spring model to describe the motion of the atoms in the medium, it is shown that bulk media with different mechanical properties are promising on the fabrication formation of simple thermal diodes. This central idea was analyzed even further by refining the initial model used, [2, 3], revealing the potential of this key idea. The present work revisits the study by Li et al. [1], to introduce a finite thickness in the interfacial area between the two different materials. Moreover, additional modifications in the Hamiltonian of Frenkel-Kontorova are made, in order to take into account potential asymmetries of the lattice substrate. In addition to that, the effective heat conductivity of the material, as a bulk, is determined. Concluding, scaling laws regarding the heat conductivity are revealed by using the machine learning framework.

I. INTRODUCTION

What drives our world is said to be the external forces caused by the environment we are surrounded, e.g. the sun or the core of the earth in our case. In particular, these external forces are responsible for the existence of life, due to the formation of the endless cycle called fluctuation-dissipation [4, 5]. That means the transfer of heat, mass and momentum transfer due to non-equilibrium processes are inherently connected to the most complex and yet less understood systems, the biological organisms.

In general, being able to control the transfer of energy in biological chemical pathways is crucial for understanding the response of biological systems in the molecular level. A great example is ATP hydrolysis [6], which is found responsible for powering molecular motors that participate in self-assembly processes of filaments [7]. Hence, by being able to accurately describe and control the heat flow within a medium is important for studying effectively those far-from equilibrium systems.

The advancement of nanotechnology led to the manufacturing of devices, called thermal diodes, which are able to precisely control the heat flow and consequently the temperature evolution in the bulk of a material [8]. Early theoretical studies have shown the potential and capabilities of such technology, by illustrating the precise control of the thermal flux [1, 3]. In particular, there is a directionality preference of heat flow, leading to an effective temperature 'switcher'. The very same idea can be used in biological environments, where all important chemical reactions or self-assembly processes are taking place in an effectively heterogeneous continuous phase.

Herein, we are interested to study the effects of triple-layered thermal diodes on the temperature profile. Our model lies on the same grounds as the one introduced by Li et al. [1], with the main difference that in the present study we consider an interfacial region of finite thickness. Additionally, substrates which are both symmetric and asymmetric are considered and their effect on the resulting macroscopic thermal conductivity is described.

II. PROBLEM FORMULATION

A. Frenkel-Kontorova Model

Frenkel-Kontorova model is one of the simplest models which is capable of describing classical particles coupled with their nearest neighbors. Additionally, the present model allows the connection with a periodic on-site potential. The interested reader on the derivation of this model should consult a series of papers by Frenkel and Kontrorova, [9–11]. According to the authors, this model is suitable to describe, in the simplest possible way, the structure and dynamics of a crystal lattice in the vicinity of a dislocation core. Herein, we are interesting in using this picture in systems where the functionality of biological molecules can be exploited for practical applications, e.g. ATP hydrolysis [6].

The basic assumptions of the model are the harmonic interatomic force between the particles and the consideration of a sinusoidal on-site potential. It was found that these simple characteristics are able to describe a broad spectrum of nonlinear, physically important phenomena, such as propagation of charge-density waves, [12], the dynamics of absorbed layers of atoms on crystal surfaces, [13], commensurable-incommensurable phase transitions, [14], domain walls in magnetically ordered structures, and more recently, the rectification of heat flux, [1, 15]. In the FK model each segment is described by the Hamiltonian

$$H = \sum_s \frac{p_s^2}{2m} + \frac{1}{2}k(x_s - x_{s+1} - a)^2 - V(x_s) \qquad (1)$$

In the present study two types of potentials will be considered. The first one is the same as the one used by Li et al., [1], which is the simpler form of the interparticle potential. That is expressed by the following relation

$$V(x_s) = \frac{V}{(2\pi)^2}\cos(2\pi x_s) \qquad (2)$$

The second version of the interatomic potential was derived by Remoissenet & Peyrard [16], which takes into

account the asymmetries in the considered substrate. As underlined by the authors, the present asymmetric substrate potential is capable of taking into account the correct soliton physics in a lattice, which are crucial for correctly predicting the heat transfer induced by chemical reaction in carbon nanotubes, [17]. The mathematical expression of the asymmetric potential is the following

$$V\left(x_s\right)=\frac{V}{(2\pi)^2}\frac{\left(1-\sigma^2\right)^2\left[1-\cos\left(2\pi x_s\right)\right]}{\left[1+\sigma^2+2\sigma\cos(\pi x_s)\right]^2} \tag{3}$$

In both expressions, V denotes the amplitude of the potential and σ is the shape parameter. For $\sigma = 0$ or $|\sigma| \to \infty$ the asymmetric potential degenerates to the simple expression used in [1].

B. Physical System

In contrast to previous works [1, 15], three distinct regions will be considered. This model results in aneight parameter model, i.e. the lattice spring parameters (k_L, k_{int}, k_R), the potential amplitudes (V_L, V_{int}, V_R) and the applied temperatures (T_L, T_R). It is important to note that the temperature values are normalized with respect to the ambient temperature T_o.

The two ends of the lattice model exist in reservoirs with constant temperature, T_L and T_R, respectively. It is wise to define the ratio $\Delta = \frac{T_L - T_R}{2T_o}$ as an important parameter for our system. Additionally, the potentials and the spring constants of the right and left regions are related with each other as follows

$$\begin{aligned} V_R &= \lambda V_L \\ k_R &= \lambda k_L \end{aligned} \tag{4}$$

The properties of the interfacial area are considered to be the mean average of the two regions, as the material there is assumed to be a well-mixed region between the two different materials. Hence, $k_{int} = \frac{\lambda+1}{2}k_L$ and $V_{int} = \frac{\lambda+1}{2}V_L$. Additionally, both local temperature and local heat flux on the position of particle n are defined as

$$\begin{aligned} T_n &= m\left\langle \dot{x}_n^2 \right\rangle \\ J_n &= k\left\langle \dot{x}_n(x_n - x_{n-1})\right\rangle \end{aligned} \tag{5}$$

It is interesting to note that these quantities are able to be acquired using a Markov-Chain Monte Carlo method, in order to find all the possible non-trivial non-equilibrium configurations, based on the bath temperatures used at each end. The set of equation of motion based on the Hamiltonian formulation is the following

$$\begin{pmatrix} \dot{x}_s \\ \dot{p}_s \end{pmatrix} = \begin{pmatrix} \frac{p_s}{m} \\ k_r(x_{s+1} - 2x_s - x_{s-1}) - \frac{dV_r}{dx_s} \end{pmatrix} \tag{6}$$

From eq. (18) it is obvious that FK model produces the classical definition of velocity $\dot{x}_s$, while the classical Newton's equation of motion obeys the following form

$$m\ddot{x}_s = k_r(x_{s+1} - 2x_s - x_{s-1}) - \frac{dV_r}{dx_s} \tag{7}$$

Without loss of generality, the position of the first particle in the lattice, x_o, is considered to be at 0. For simplicity, the mass of each particle, m, and the FK parameter, a, are set to be $m = 1$ and $a = 0$, respectively. In addition to that, in order to solve eq. (19), it is preferable to consider the velocity of each particle as an additional variable, $u_s = \dot{x}_s$. The initial equilibrium state assumes stationary particles and thus $u_s(0) = 0\,\forall s \in (1, 2, \dots)$.

The boundary conditions on the lattice are implemented by considering Langevin heat baths. More specifically the end nodes of the particles mesh are descriibed by the following equations

$$\begin{aligned} m\ddot{x}_1 &= k_r(x_2 - 2x_1) - \frac{dV_r}{dx_s}(x_1) - \zeta\dot{x}_1 + g_L(t) \\ m\ddot{x}_N &= k_r(x_{N-1} - 2x_N) - \frac{dV_r}{dx_s}(x_N) - \zeta\dot{x}_N + g_R(t) \end{aligned} \tag{8}$$

where $\zeta = 1$ and $g_L(t), g_R(t)$ are the white noise terms. Based on the fluctuation-dissipation theorem, [18], the variance of $g_L(t)$ and $g_R(t)$ should satisfy the following relations

$$\begin{aligned} \langle g_L(t)g_L(t')\rangle &= 2\zeta k_B T_L \delta(t - t') \\ \langle g_R(t)g_R(t')\rangle &= 2\zeta k_B T_R \delta(t - t') \end{aligned} \tag{9}$$

The solution of these equations is performed by using a stochastic version of the 4^{th} order Runge-Kutta as described in [19, 20]. The high non-linear character of the Stochastic ODE system requires time integration step of the order $O(10^{-4})$. A typical non-equilibrium steady state is attained after $O(10^6)$ dimensionless time units.

III. RESULTS AND DISCUSSION

A. Harmonic Lattice Validation

In order to validate the developed algorithm, the simple harmonic lattice was examined first. Due to the usage of Langevin Heat bath at both ends of the lattice chain, a linear dependence of the heat flux J vs. the temperature difference, ΔT at both ends is expected. Fig. 1 illustrates the heat flux, J as a function of ΔT. It is obvious that with decreasing the temperature difference, J decreases linearly, while for $\Delta T = 0$, J is approximately zero. This result was expected, as the Harmonic lattice is the root for the derivation of the Fourier's Law from the statistical viewpoint.

In relation to the Langevin heat bath, the Nose-Hoover model (NH) has also been proposed as a potential thermostat. Nevertheless, recent studies revealed that the NH model is non-ergodic producing artifacts in the physical phenomena observed, [21], e.g. non-monotonic behavior on J vs. ΔT for harmonic lattices.

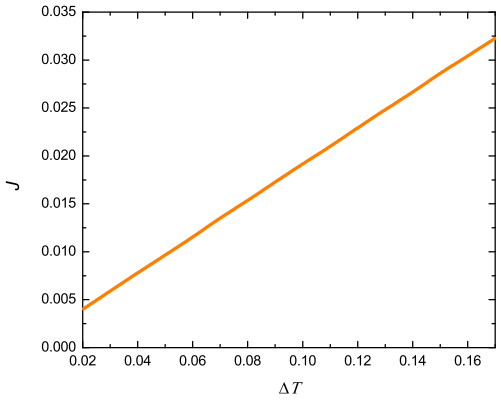

FIG. 1. Heat Flux vs. Temperature Difference between the two lattice ends

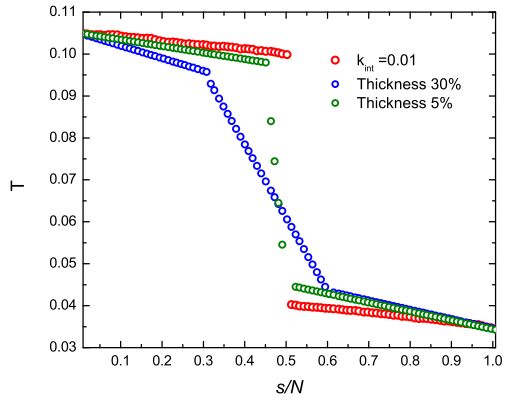

FIG. 2. Spatial profiles of Temperature with respect the normalized lattice length

B. On the discontinuous behavior of the Temperature in the lattice domain

As shown by Li et al. [1], the existence of different material properties in series is crucial in the establishment of a discontinuous field temperature along the mesh points. This is interesting because from continuum mechanics it is well know that between two different materials, only the fluxes can show a discontinuous behavior across an interface, while temperature, as a field variable, is always continuous, [22, 23]. This phenomenon, described and extensively studied by Li et al. and others, [1, 24, 25], is a manifestation of the thermal rectification phenomenon. Its mechanism though lies on the existence of an intermediate material between the two different lattices considered, which changes abruptly the dynamics of the chain. The authors considered only one particle point in the lattice, which as shown, it contributed tremendously in the existence of a discontinuous behavior in the temperature profile. This discontinuity was found to depend on the elastic constant, k_{int}, of this 'interfacial' material. More specifically, low values of k_{int} in relation with the bulk lattices induced larger spatial 'jumps' in temperature. With $k_{int} \to k_R$, the temperature profile was smoothed out, leading to a less pronounced discontinuity. Fig. 2 illustrates the validation of the results by Li et al. for $k_{int} = 0.01$. Also, T_L is considered to be larger than T_R. It is important to mention that all parameters are the same, while the interparticle potential is considered to be that of the simplest sinusoidal form, $\sigma = 0$. From fig. 2 it is obvious that the present algorithm developed to resolve the lattice dynamics with the Langevin heat bath is capable of reproducing existing literature results.

The assumption of the existence of only one particle mesh point to describe the interfacial area, does not seem physically reasonable. Although, the fabrication of a thermal rectifying device which consists of multiple different layers of materials is possible in practice, it was shown recently by Cottrill & Strano, [26], that a phase change material is capable to produce similar thermal rectification results. Hence, going one step further than the work by Li et al. [1], a finite thickness interfacial area is considered to exist. This 'sandwich' area is assumed to have the average properties of both ends (van der Waals interface, [27]). This assumption is reasonable, as a finite thickness area exists on the interface of two different materials. The thickness of the area is infinitesimal from the continuum viewpoint, but it has been proved to play an important role on several phase transition phenomena on Li-ion batteries, [28]. Figure 2 depicts the results on the termpature profile along the considered medium. It is obvious that the existence of a finite thickness area with averaged properties eliminates the discontinuous behavior on temperature. This was expected as the material has a spatial region in which the thermal dynamics can diffuse, leading to the well-known result of discontinuous fluxes only. With decreasing thickness, the discontinuous behavior is recovered and the results are qualitatively similar with that of Li et al. [1].

C. Non-monotonic thermal conductivity effects

Cottrill & Strano, [26], highlighted the importance of the temperature dependence on thermal conductivity, κ. The authors showed that non-linear phenomena introduced by $\kappa(T)$ contributed significantly on the observation of thermal rectification. The use of the asymmetric interparticle potential introduced in Section 2.1, is capable on predicting the temperature dependence of κ. In order to predict the heat conductivity in the material the following formula is considered

$$\kappa = \frac{N_p J}{\Delta T} \tag{10}$$

The value of heat flux J is found upon reaching a non-equilibrium steady state in the system. Also, in the above equation N_p consists the total number of mesh points considered in the lattice. It is important to note that a homogeneous material is considered in the present section, in order to extract general relations for the thermal conductivity as a function of temperature.

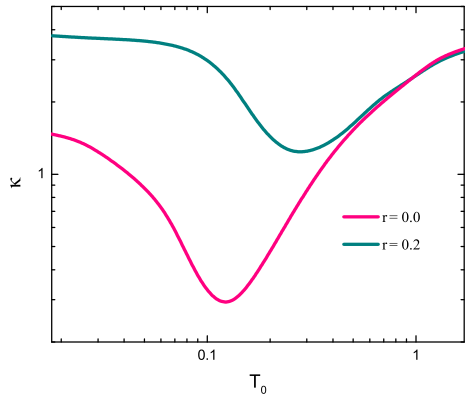

FIG. 3. Heat conductivity vs. supplied temperature

Fig. 3 shows the dependence of κ with respect to the input base temperature T_o. In the first case, the potential was assumed to obey a simple sinusoidal wave relation where $\sigma = 0$. On the second case, σ was considered to be equal to 0.2. From these results once can see the non-monotonic behavior of κ with increasing T. Irrespectively of the asymmetry considered in the potential of the lattice, the behavior is qualitatively the same for small T_o, while for values larger than 0.5 the results collapse onto a single curve. This behavior is completely different with that considered by Cottrill & Strano, [26], where their heat conductivity parameter had a hyperbolic tangential dependence. The physical mechanism which leads to exactly the same κ with increasing σ for large T is the following: with larger heat bath values, the both end mesh points attain larger kinetic energy, leading to increasing particle 'jingling'. Hence, for larger temperature inputs, the oscillation of all particles are in phase with each other due to the increased kinetic energy, leading to quantitatively similar dynamics. For $T_o > 0.5$ it is shown that κ scales as

$$\kappa \sim T^2 \tag{11}$$

while for $T_o \leq 0.5$ κ can be described by the following non-linear relation

$$\kappa \sim \tanh(AT^3) \tag{12}$$

These dependences were found by using the commercial package Eureqa$^{\text{TM}}$, [29], where machine learning is used in order to fit the data with a set of generalized functions.

IV. CONCLUSION

The present study accounted for the effects of lattice dynamics on both thermal rectification effect and the dependence of heat conductivity on temperature.

It was found that the finite thickness of the interface between two materials affects significantly the temperature profiles produced in the material. More specifically, the discontinuous behavior predicted by Li et al., [1], no longer exists for interface thickness larger than 5% of the chain length.

In addition to than, the consideration of asymmetric potential of the substrate effects significantly the heat conduction at low dimensionless temperatures. In particular, with increasing assymetry on the interparticle potential, the heat conduction is enhanced. For larger temperature though, the asymmetric effects in the lattice does not effect significantly the particle dynamics and thermal conductivity was found to be the same for different asymmetric numbers, σ. By using a commercial non-linear regression software, we were able to find the scalings which govern the behavior of κ with respect to the mean temperature of the heat bath.

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