Efficient Monte Carlo Electron Transport Formalism for Highly Nanostructured Materials

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INTRODUCTION

Nanomaterials and nanostructures are typical in many technologies. One category of benefiting from nanostructuring is thermoelectric (TE) materials [1]. New-generation TE materials are typically highly nanostructured, with the nano-features spanning from macro- to nano-scale (including boundaries, potential barriers, pores, nanoinclusions, atomic defects, second-phasing, etc.) (Fig. 1a, 2a). To theoretically evaluate carrier transport in these cases, a common method is the solution of Boltzmann transport equation (BTE) stochastically using the Monte Carlo (MC) method. However, existing MC methods have many limitations in this setting, the most severe being the huge challenge to gather statistics from two opposite going fluxes under linear response (Fig. 1b) [2]. To make things worse, as geometry complexity increases the simulation time increases exponentially (Fig. 2b). Here we develop an efficient MC transport algorithm tuned specifically for complex nanostructures under linear response.

SIMULATION FORMALISM

Our novel method uses a single-injected flux from the left of the domain only, which traverses through the channel with consecutive free-flight and scattering events (Fig. 3), and stochastically provides the typical flight time (ToF). The ToF is used for the flux calculation (eq. 1) and then the transport distribution function (TDF) (eq. 2), which then evaluates conductivity (eq. 3) as [2].

$$F(E) = \frac{1}{< ToF(E) >} \tag{1}$$

$$\Xi(E) = F(E) \times DOS \tag{2}$$

$$\sigma = q^2 \int_E \Xi(E).\left(-\frac{\partial f}{\partial E}\right).dE \tag{3}$$

Compared to common MC algorithms, our method has the following differences: (i) we employ a mean-free-path approach (mfp), where the carriers scatter definitively after every mfp, rather than using self-scattering events, thus avoiding the large number of scattering events for low velocity, low energy carriers (Fig. 3). (ii) We map the particle flux from MC to the analytical TDF under acoustic phonon scattering conditions (ADP), thus avoiding the need for super-electron charges and peculiarities in simulating a 2D vs. a 3D channel. Under ADP the TDF is linear, and that is what we capture in our MC flux as well (Fig. 4, 5). The mapping is done using a single constant *C* (Fig. 6, inset), which maps the conductivities and the TDF. (iii) We use only one flux and mimic the flux differences and applied fields with the *df/dE* term in BTE, thus avoiding the difficulty in subtracting two opposite fluxes (Fig. 4, red line). Note that this is particularly useful for the Seebeck coefficient where not only the difference of left/right going fluxes needs to be resolved, but simultaneously the ones that flow above/below the Fermi level (Fig. 5, red line).

We have computed the conductivity in Fig. 6 for a pristine channel, a channel with pores, a channel with grain boundaries, and a channel with both nano-features. The latter result is as predicted from Matthiessen's rule using the first two nanostructured channel simulations. We estimated at least an order of magnitude computational savings compared to the current methods.

CONCLUSION

We developed an efficient MC algorithm by overcoming limitations of existing MC methods, tuned specifically for the electronic transport in complex nanostructures under linear response.

REFERENCES

[1] D. Beretta, et. al. *Thermoelectrics: From history, a window to the future*, Mat. Sci. and Eng.: Reports. **138**, 100501 (2019).

[2] P. Priyadarshi and N. Neophytou, Computationally efficient Monte Carlo electron transport algorithm for nanostructured thermoelectric material configurations, *Journal of Applied Physics*. **133**, 054301 (2023).

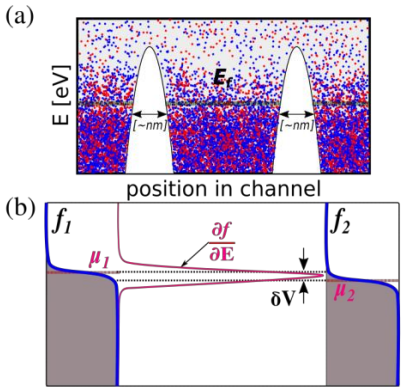

Fig. 1. (a) MC ensemble particles in a nanostructured domain. (b) Energy derivative of the Fermi distribution, which mimics by a small applied potential δV in low-field transport conditions.

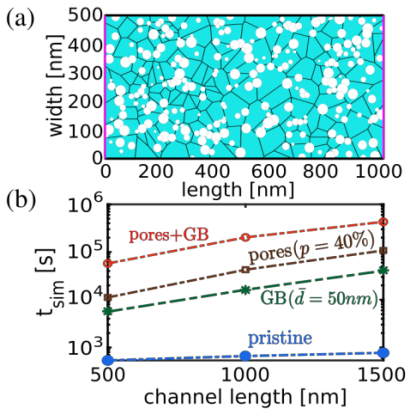

Fig. 2. (a) Complex nanostructured domain populated with grain boundaries and nanoporous features. (b) Simulation time versus device channel length of various nano-featured 2D domains.

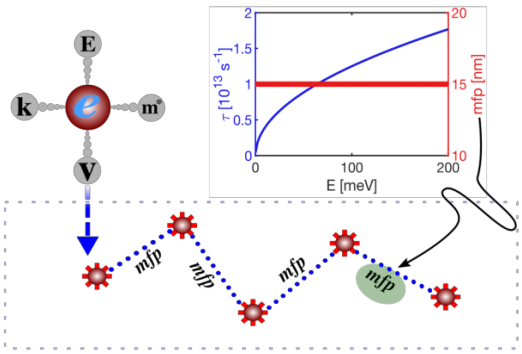

Fig. 3. Developed MC algorithm: need electron parameters, their mean free path, which is constant under ADP scattering.

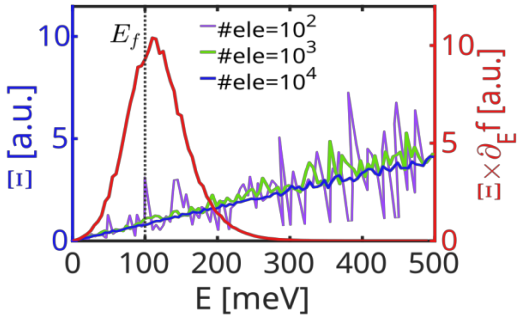

Fig. 4. Transport distribution function, Ξ(E), calculated stochastically using the MC algorithm. The red line (right axis) shows the product of Ξ(E) and the differential Fermi function with respect to energy ∂_Ef (the case of 10^4 electrons).

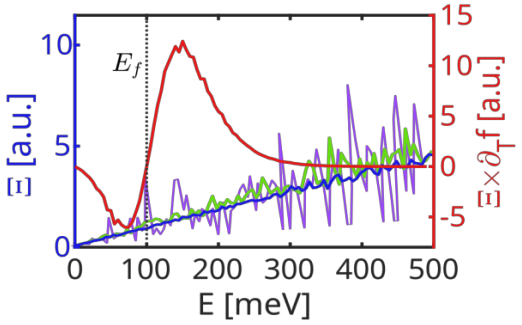

Fig. 5. Transport distribution function using the MC algorithm. The red line (right axis) shows the product of Ξ(E) and the differential Fermi function with respect to temperature ∂_Tf (shown for the case of 10^4 electrons).

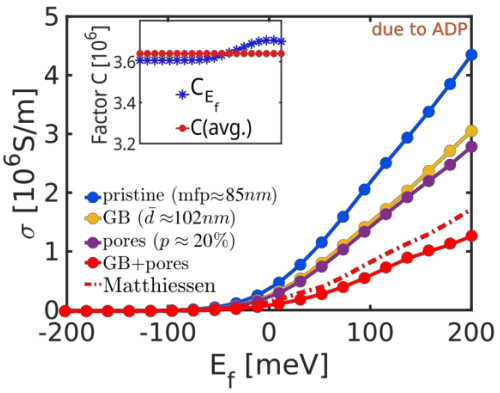

Fig. 6. Electrical conductivity calculated from the MC algorithm in nanostructured material domains populated with grain boundaries, pores, and the combination of both versus the Fermi energy. The red-dotted line shows the calculated conductivity using Matthiessen's rule. Inset: mapping of the analytical to the simulated conductivity.