Hydrodynamic signatures in thermal transport in devices based on 2D materials: an ab initio study

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We investigate the features arising from hydrodynamic effects in graphene and phosphorene devices with finite heat sources, using ab initio calculations and solving the phonon Boltzmann transport equation through energy-based deviational Monte Carlo methods. We explain the mechanisms that create those hydrodynamic features, showing that boundary scattering and geometry are determinant factors, and that the length scales at which they can appear depend solely on the ability of intrinsic scattering to randomize the heat flux. We relate this latter point to the non-local lengths and mean free paths, additionally providing an insight into how the scattering operator must be treated to obtain a proper description of the hydrodynamic behavior.

Keywords: Phonon, hydrodynamics, Boltzmann Transport Equation

I. INTRODUCTION

Over the last decades, the continuous development of micro- and nanofabrication techniques has allowed for higher integration levels in electronic devices and more efficient thermoelectric systems, as well as better thermal insulators for improved energy savings [1–5]. A proper understanding of the thermal transport in those systems becomes essential to optimize their operation since, if not carefully managed, heat can severely hinder their efficiency and/or durability [6–9].

Classically, heat transport has been modeled with Fourier’s law [10]. However, Fourier’s law is known to break down in small systems and short timescales, with several examples available: for instance, it has been observed that Fourier’s law fails to properly reproduce several thermoreflectance [11,13] and thermal grating [14,15] experiments even with some, sometimes physically questionable, modifications [12]. Such erroneous descriptions have their origin in the shortcomings of Fourier’s law, i.e., the infinite velocity of heat propagation [16–18] and the lack of non-local effects [15–18]. The inclusion of these two effects into the theory makes heat acquire characteristics of a viscous fluid, which are describable via additional hydrodynamic (Navier-Stokes-like) terms [15,18,19]. Two clear signatures of hydrodynamic behavior are the formation of heat flux vortices and the appearance of non-monotonic temperature profiles in non-trivial sample geometries. Such hydrodynamic features have also been predicted to appear in the context of electronic transport for graphene [20], and recent works have also predicted the existence of thermal vortices in nanoporous silicon [21] and graphene-based devices [22,23], though the used methodology was, in these cases, based on fitted parameters or models instead of a full microscopic ab-initio-based description.

In the case of semiconductors, where phonons are the main heat carriers, it is possible to obtain an accurate description of thermal transport, including non-Fourier features, by solving the Peierls-Boltzmann transport equation (PBTE) with the appropriate boundary conditions [24–25]. Indeed, if supplied with first-principles data, this latter equation can even model devices based on novel materials for which other simpler descriptions are lacking. Despite its advantages in terms of precision and transferability, the solution of the PBTE, for devices and/or structures of interest for heat management or thermoelectric applications, is rather complex and expensive. Moreover, the overall complexity of the PBTE makes it hard to obtain an intuitive and clear physical picture of heat transport phenomena [19,26].

In this context, beyond-Fourier mesoscopic models (i.e., the ones that not only describe the classical diffusive regime of Fourier’s law, but other important transport regimes in the nanoscale, such as the ballistic and hydrodynamic ones) are essential for a fast, simple, and physically intuitive description of thermal transport at the micro/nanoscale [26]. Among all available mesoscopic equations, the hydrodynamic equation stands out as a promising candidate since it can be directly derived from the BTE, so that a microscopic physical description of its variables might be obtained, enabling their calculation with ab initio methods for accurate simulations [18,27].

In this work, we study hydrodynamic signatures in graphene and phosphorene-based nanodevices at room temperature using first principles by solving the PBTE via energy-based deviational Monte Carlo (MC) techniques. We provide insight into the mechanisms originating those features, and complement our results with solutions of the hydrodynamic equation for a better understanding of some size effects as well as studying signatures below the MC statistical noise.

The paper is structured as follows: after briefly discussing hydrodynamics in Sec. II, we detail the applied methodology for the heat fluxes and thermal profiles calculation in Sec. III and we present the bulk properties, cumulative curves and a discussion of the expected hydrodynamic features in Sec. IV. The results for devices are provided in Sec. V, both for thinner (Sec. VA) and wider ballistic devices (Sec. VB). Our summary and conclusions are given in Sec. VI.
II. HYDRODYNAMICS AND 2D MATERIALS: THE ROLE OF NORMAL AND UMKLAPP PROCESSES

The steady-state hydrodynamics equation of heat transport is given by:

$$\mathbf{J} = -\kappa \nabla T + \ell^2 \nabla^2 \mathbf{J}$$  \hspace{1cm} (1)

where \( \mathbf{J} \) is the heat flux, \( \kappa \) is the thermal conductivity tensor, \( T \) is the temperature and \( \ell \) is a non-local length tensor, which is related to the distance in which phonon distribution can conserve its inertia even under the effect of intrinsic scattering. One of the earliest derivations of this hydrodynamic equation for heat transport was by Guyer and Krumhansl (GK) [28]. In their work, they derived it by expanding the phonon distribution on eigenvectors of the normal collision operator under the condition that normal (\( N \)) scattering is dominant over umklapp (\( U \)) and extrinsic (\( E \)) scattering. Indeed, within the GK framework, hydrodynamic features are only possible when \( N \) processes dominate over all other kinds of scattering [19, 22, 29, 30].

A common approach to the scattering operator when solving the PBTE is the so-called relaxation time approximation (RTA), where it is assumed that each phonon mode relaxes to equilibrium independently. Despite its simplicity, such an approach has been observed to properly describe thermal properties for prototypical semiconductors like silicon. However, since in the RTA all scattering processes are deemed resistive, the RTA is expected to fail for cases in which most of the processes are not directly resistive but indirectly so through population redistribution, which has been usually associated to \( N \gg U, E \) [28, 29]. Thus, in 2D materials, in which the RTA has been observed to provide a poor description of thermal properties [29, 31], hydrodynamic features should be stronger than for other materials [22, 29].

It is worth noting that the role of \( U \) and \( N \) processes in phonon hydrodynamics is more nuanced than the widespread belief, rooted in the GK derivation, that \( N \) processes must dominate [29, 30]. That is a sufficient but not necessary condition, since the hydrodynamic regime is possible even in the case where \( N \) and \( U \) scattering are comparable or when \( E \) dominates over intrinsic scattering [19, 23, 27]. In fact, by projecting the PBTE over energy and quasimomentum, and by expanding the phonon distribution on macroscopic variables like the flux, Sendra et al. [27] have derived a general hydrodynamic formalism that does not rely on the \( N \) dominance or on the classification of three-phonon processes into \( N \) and \( U \), which is subject to the specific choice of the primitive cell [32, 33]. This generalizes the hydrodynamic regime to materials where both resistive and non-resistive processes are comparable—i.e.: out of the range of applicability of the RTA—, as in the case of diamond or 2D materials [19], but also to materials like room-temperature silicon in which intrinsic resistive scattering is the dominant mechanism [27].

Taking into consideration everything mentioned here, we have selected graphene and phosphorene for our study. The former was chosen because, in addition to it being the prototypical 2D material, several works [20, 22] have already predicted current vortices in both electronic and thermal transport. On the other hand, the latter was chosen in view of its excellent electronic properties [34], which have positioned it as a principal actor in the survival of Moore’s law down to atomic sizes [35], thus making a proper description of heat transport, including hydrodynamics, essential for the developing of phosphorene-based devices.

III. METHODOLOGY

Previous works have predicted electrical current vortices and negative nonlocal resistance in 2D materials (graphene) when finite sources are injecting heat/electrons [20, 22, 23]. A sketch of those geometries, henceforth called Levitov configurations, is depicted in Fig. 1. The dimensions of the studied Levitov configurations were selected to investigate all possible regimes; to that end, a mean free path (\( \lambda_\alpha \)) and a non-local length (\( \ell \)) have been computed to provide an approximate idea of the limiting sizes of each transport regime. The mean free path is defined as

$$\lambda_\alpha \equiv ([\kappa^{\text{SG}}]^{-1} \kappa^{\text{RTA}})_{\alpha\alpha}. \hspace{1cm} (2)$$

Here \( \alpha \) indicates a Cartesian axis, \( \kappa^{\text{RTA}} \) is the RTA thermal conductivity tensor and \( \kappa^{\text{SG}} \) is the small-grain thermal conductivity defined as \( \kappa^{\text{SG}} = k_B T \sum_\sigma n_\sigma^0 (n_\sigma^0 + 1) \left( \frac{\omega_\sigma}{v_\sigma} \right)^2 v_\sigma \otimes v_\sigma \), where \( k_B \) is the Boltzmann constant, \( V_\text{uc} \) is the volume of the unit cell, \( \sigma \) is a phonon mode, \( n_\sigma^0 \) is the Bose-Einstein distribution for mode \( \sigma \), \( h \) is the reduced Planck constant, \( u_\sigma \) is the phonon frequency and \( v_\sigma \) is the phonon group velocity. In addition, the non-local length (\( \ell \)) within the RTA and for isotropic materials, \( \ell^{\text{iso}} \), can be calculated as [27]:

$$\ell^{\text{iso}} = \frac{1}{5} \left[ \int_{BZ} hq_\sigma v_\sigma^2 \tau_\sigma^2 \partial_T n_\sigma^0 d\sigma \right]. \hspace{1cm} (3)$$

where \( q_\sigma \) is the phonon quasimomentum, \( \tau_\sigma \) is the phonon lifetime and \( \partial_T n_\sigma^0 \) is the derivative with respect to temperature of the Bose-Einstein distribution. Additionally, we also provide the thermal conductivity as it is an essential quantity in the hydrodynamic equation. Cumulative quantities were also computed as a mean to obtain a deeper understanding of size effects in these configurations [25].

Heat flux and temperature profiles for several Levitov configurations are obtained through the PBTE solution. Owing to the high complexity of the Levitov geometry, we use an efficient, energy-based deviational Monte Carlo approach [36, 41] to solve the PBTE under the RTA and beyond (bRTA), implemented in BTE-Barna [42] to solve the PBTE.
FIG. 1: Sketch of a Levitov configuration with characteristic lengths $H$, $W_{\text{reservoir}}$ and $W_{\text{device}}$ indicated. The transport axis, armchair (AC) and zigzag (ZZ), for phosphorene case are given as reference.

Atomic positions, harmonic and anharmonic interatomic force constants (IFCs), needed to compute the basic phonon properties (i.e.: group velocities, lifetimes, frequencies) and the propagator \cite{40, 42}, required for the PBTE Monte Carlo solvers, were obtained from Refs. \cite{43} for graphene and \cite{44} for phosphorene. Harmonic IFCs were corrected to enforce crystal symmetry, translational invariance, and rotational invariance, necessary for a proper description of quadratic acoustic bands \cite{45}, the broadening parameter was fixed to 1 for energy conservation and the phosphorene and graphene layer thicknesses were set to 0.533 nm \cite{46} and 0.335 nm \cite{40} respectively. The phosphorene (graphene) phonon properties and the propagator were calculated on a $\Gamma$-centered $q$-mesh of $50 \times 50 \times 1$ ($80 \times 80 \times 1$) points, for which the thermal bulk conductivity is found to be converged—within 5% with respect to a higher quality mesh of $100 \times 100 \times 1$ ($110 \times 110 \times 1$) points—at 300 K. For both materials, the propagator was calculated using a time step of 0.25 ps and a reference temperature of 300 K.

Finally, to efficiently investigate some dimensional limits and other features as the existence of vortices with magnitudes well below statistical noise caused by the intrinsic scattering algorithms \cite{39, 42}, we solved Eq. \ref{eq:1} using finite elements package FEniCS \cite{47, 48}, with its parameters extracted from cumulative curves (see the fcnics.py script in the Supplemental Material \cite{49} for further details on those calculations).

IV. BULK PROPERTIES AND SIZE EFFECTS

In their work, Shang et al. \cite{22} proposed two different mechanisms as the origin of thermal vortices. For small systems, in the ballistic regime, the source would be the combination of elastic boundary scattering plus the phase of phonons but not hydrodynamics per se. On the other hand, for larger devices, they concluded that vorticity is due to $N$-scattering dominance together with boundary conditions (finite sources injecting the heat into the system).

To get an idea of the interplay between the device dimensions and its transport regime, we have calculated the bulk non-local length and mean free path for graphene and phosphorene at room temperature (see Table \ref{tab:1}). Additionally, we also provide the thermal conductivity because of its importance for heat hydrodynamics, with very good agreement with other theoretical calculations and experimental values. Although the non-local lengths and mean free paths offer a general idea of transport regime limits, they fail to provide insight on how system boundaries partially suppress the macroscopic thermal properties (i.e. $\kappa$ and $\ell$), as longer-mean-free-path phonons do not fully contribute to these macroscopic variables \cite{24}. The last is especially important as it modifies $\ell$, which is one of the quantities determining the transport regime. While it would be possible to obtain the effective macroscopic quantities for given geometries via averaging \cite{25}, the overall complexity of the Levitov configuration makes such a task unfeasible. On the other hand, even though, as noted by Li et al. \cite{25}, mean free path cumulative functions do not hold accurate predictive power, they are a useful tool to interpret size effects on transport variables. To that end, we provide the cumulative thermal conductivity and non-local length as a function of phonon mean free path (see Figs. \ref{fig:2} and \ref{fig:3}).

It should be noted that the theoretical formulas used to obtain the $\ell_{\text{iso}}^{\text{RTA}}$ values are derived under the assumption of isotropy and are therefore expected to be useful only as approximations in the phosphorene case. Regarding $\ell$ values beyond the RTA, we expect them to be higher overall as in the RTA all processes are incorrectly deemed as directly resistive \cite{27, 42}. Notwithstanding this, in the case of graphene the dominance over $\ell_{\text{iso}}^{\text{RTA}}$ of low-frequencyZA-modes in the neighborhood of $\Gamma$ with extremely large mean free paths (of the order of 4 nm)—to the point that when suppressed the $\ell_{\text{iso}}^{\text{RTA}}$ gets reduced from 53.9 $\mu$m to 43.5 $\mu$m—makes the qualitative prediction of the beyond-RTA-$\ell$ behavior much more complicated, as the proper description of scattering operator would highly affect those ZA-modes. This is not surprising, as the RTA is known to fail in predicting graphene’s thermal properties \cite{24}. For instance, in our case the RTA predicts the $\kappa$ to be only 27% of the fully converged value.

Regarding device dimensions, in the phosphorene case we expect hydrodynamic features to be observable out of the (quasi)ballistic regime, as the mean free path is
TABLE I: Calculated $\kappa$, $\ell$ and $\lambda$ for bulk graphene and bulk phosphorene at 300 K. The experimental values of $\kappa$ and values of other theoretical calculations are also provided.

|                | $\kappa_{\text{RTA}}$ ($\frac{W}{m\cdot K}$) | $\kappa_{\text{beyond RTA}}$ ($\frac{W}{m\cdot K}$) | $\kappa_{\text{ref}}$ ($\frac{W}{m\cdot K}$) | $\ell_{\text{RTA}}$ (nm) | $\lambda$ (nm) |
|----------------|---------------------------------------------|---------------------------------------------|---------------------------------------------|---------------------|----------------|
| **Graphene**   | 1.17 $\times$ 10\(^3\)                      | 4.20 $\times$ 10\(^3\)                      | (4.84-5.30)$\times$10\(^3\)               | 5.39 $\times$ 10\(^4\) | 169            |
|                |                                             |                                             |                                             |                     |                |
| **Phosphorene (AC)** | 20.8                                       | 27.5                                       | 23.9\(^c\)                                 | 27.7                | 20.2\(^c\)    |
|                |                                             |                                             |                                             |                     |                |
| **Phosphorene (ZZ)** | 57.8                                       | 82.9                                       | 82.1\(^c\)                                 | 42.3                | 33.5\(^c\)    |

\(^a\) Experimental $\kappa$ obtained from Ref. [50].
\(^b\) Experimental $\kappa$ obtained from Ref. [51].
\(^c\) First-principles calculated $\kappa$ obtained from Ref. [52]. $\kappa$ is rescaled to take into account differences in assumed thickness.
\(^d\) First-principles calculated $\kappa$ obtained from Ref. [53]. $\kappa$ is rescaled to take into account differences in assumed thickness.
\(^e\) First-principles calculated $\kappa$ obtained from Ref. [54].

\(^1\) this value corresponds to the $\ell_{\text{iso}}$ calculation without including low frequency ZA modes in the neighborhood of $\Gamma$ with $\lambda$ around 4.2 mm.

FIG. 2: Cumulative $\kappa_{\text{RTA}}$ for phosphorene (left) and graphene (right) with respect to mean free path at 300 K. Bulk values are provided as reference. These values have been computed in a denser $\mathbf{q}$-mesh, namely 300 $\times$ 300 $\times$ 1 (480 $\times$ 480 $\times$ 1) for phosphorene (graphene), through a cubic spline interpolation from the finer mesh to remove nonphysical artifacts.

a bit longer than the non-local length, the true value of which is expected to be larger than the one in Table I [42]. The latter suggests that phosphorene is a highly hydrodynamic material at the nanoscale, as the phonon distribution is capable of keeping its inertia even under the effect of significant intrinsic scattering. The case of graphene is somewhat more complex, as the major contribution to thermal variables, especially the non-local length, comes from the low-frequency ZA modes in the neighborhood of $\Gamma$ with extremely large mean free
paths. At the nanoscale, the contribution of these modes to thermal conductivity or non-local length becomes negligible because a large suppression factor due to boundary scattering—see Eq. (14) in Ref. 36—, thus lowering the non-local length to 43.5 nm. Hence for graphene nanodevices, since that non-local length is smaller than λ, one would only expect hydrodynamics within the (quasi)ballistic regime.

V. RESULTS

Before presenting the results of our simulations, we find it worth commenting on the hydrodynamic signatures we have analyzed. Although the hydrodynamic regime presents several characteristic features, based on previous results 20, 22, 23 we have focused our study on flux vortices and negative thermal resistance regions, namely regions in which the flux propagates in the same direction as the thermal gradient. None of those features can be easily described using Fourier’s law unless one relies on complex models allowing for a position-dependent non-diagonal and non-definite-positive κ-tensor. However, these features arise naturally in hydrodynamics (see Eq. 1) without using those complex and sometimes unphysical models, just unique and diagonal κ and ℓ tensors.

A. Quasiballistic devices

Here, we provide the thermal profile and heat flux for phosphorene and graphene-based Levitov configurations well inside quasiballistic regime (see Figs. 4 and 5), i.e. H < λ, for within and beyond the RTA.

All temperature profiles show a small linear variation of temperature between the reservoirs (i.e. x = 0), with abrupt changes near the reservoirs. Such a feature confirms that our devices are in the quasiballistic regime 54. This statement is further supported by heat flux profiles, in which the small differences in fluxes between the bRTA and RTA indicate the dominance of boundary scattering (as this mechanism is not dependent on the approach used to describe intrinsic scattering) thus devices are in the quasiballistic regime 29, 30.

Regarding the hydrodynamic signatures, we can observe two flux vortices for all devices. Moreover, by combining flux and thermal profiles, we observe zones of negative thermal resistance at both sides of the reservoirs (see hot (cold) regions at both sides of cold (hot) reservoirs with the heat flux going downwards in Figs. 4 and 5. Thus, all our (quasi)ballistic devices are in the hydrodynamic regime, even in the absence of N processes, in opposition to more classical hydrodynamic theories.

Finally, the observable differences between graphene and phosphorene, especially for heat fluxes, which are as expected of higher values in graphene, are easily understood by noting the higher group velocities and lower scattering rates of graphene when compared to the phosphorene ones. Moreover, the asymmetric vortices found in phosphorene have their root in the material anisotropy.

On top of that, and taking into account the role of boundary scattering on vortex formation pointed out by Shang et al. 22, we also provide the PBTE solution without intrinsic scattering (i.e. three-phonon and isotopic scattering), henceforth referred to as extrinsic (see Figs. 7).
From those results, it is clear that the boundary scattering is the only mechanism leading to vortex formation, as the other proposed contributing factor, the phase of phonons, is disregarded by the PBTE [55]. This last statement agrees with Zhang et al.’s observations [23]: if allowed by boundary scattering, i.e. system geometry, vortices will form, being protected from destruction by the absence or scarcity of intrinsic resistive scattering processes. Furthermore, phosphorene extrinsic results show a curious feature, two secondary vortices, which are found neither in the RTA nor in bRTA results. We need to stress that the flux magnitude of such secondary vortices is well below—\(\sim 10^4\) times smaller than the maximum, see Fig. 8—the error introduced by the scattering algorithms [39], so the observed flux in that area for the RTA and bRTA is nothing but noise-masked flux. To
FIG. 6: The extrinsic steady-state thermal profiles (top) and heat fluxes (bottom) for a ballistic phosphorene-based Levitov configuration with $W_{\text{reservoir}} = 25 \, \text{nm}$, $W_{\text{device}} = 160 \, \text{nm}$ and $H = 19.2 \, \text{nm}$, $T_{\text{HOT}} = 310 \, \text{K}$ and $T_{\text{COLD}} = 290 \, \text{K}$. The black bars at the colorbars of temperature profiles, indicate the highest and the lowest temperature found in device.

FIG. 7: The extrinsic steady-state thermal profiles (left) and heat fluxes (right) for ballistic graphene-based Levitov configuration with $W_{\text{reservoir}} = 37.5 \, \text{nm}$, $W_{\text{device}} = 375 \, \text{nm}$ and $H = 75 \, \text{nm}$, $T_{\text{HOT}} = 310 \, \text{K}$ and $T_{\text{COLD}} = 290 \, \text{K}$. The black bars at the colorbars of temperature profiles, indicate the highest and the lowest temperature found in device.

Check for the existence of those secondary vortices, we resorted to the hydrodynamic equation using the cumulative quantities for the given device dimension as equation parameters; the result is given in Fig. 9. Although there exist differences between this solution of the hydrodynamic equation and the MC simulations, those can be attributed to the facts that cumulative variables only capture the qualitative effects of size and that Eq. 1 does not capture ballistic effects present in the device. Moreover, we recall that the calculation of the $\ell$ values used here was conducted under the assumption of isotropy. Notwithstanding all these differences, this result together with extrinsic simulation strongly suggests the presence of additional vortices hidden by noise in Fig. 4.

We also provide evidence of the existence of ternary and higher order vortices by solving the hydrodynamic equation for a wider phosphorene device (see Fig. 10). Interestingly, we observe from those results that each vortex is approximately 100 times smaller in magnitude than the previous one (see Fig. 11) in line with MC sim-
length, the defining parameter of vorticity, for the RTA and bRTA were found in nanoribbons of sizes similar to those found in our devices. In view of such discrepancies, we investigated the existence of limiting cases, namely those cases in which a small difference in \( \ell \) might lead to different vorticity. In Fig. 13 we can see an example of such a limiting case for phosphorene, confirming the importance of a proper treatment of scattering even in the quasiballistic regime for a proper description of vortices. There, it can be observed that while in the RTA there are four different vortices, in the bRTA case there are still two different vortices. Moreover, those results also show vortex damping due to the lateral wall, demonstrating that a minimum distance between lateral walls and the reservoir ends is necessary for vortices to form [23]. This is not surprising, as in the limit of thermal reservoirs occupying all top and bottom edges one must recover a Poiseuille flow.

1. Vortex formation

To obtain a better understanding about how boundary scattering creates vortices, we look into how they are formed. For that purpose, we provide temporal snapshots showing the vortex formation within the bRTA framework, starting from a graphene-based Levitov configuration with \( W_{\text{reservoir}} = 37.5 \text{ nm} \), \( W_{\text{device}} = 375 \text{ nm} \) and \( H = 75 \text{ nm} \), \( T_{\text{HOT}} = 310 \text{ K} \) and \( T_{\text{COLD}} = 290 \text{ K} \), initially at 300 K in Fig. [14]. For a video of the time evolution of the heat flux and temperature see the flux_short.mp4 and temperature_short.mp4 videos in the Supplemental Material [49], respectively.

These results show that the formation of vortices begins at the sources, and how they advance with time towards the lateral edges until their closure. In further detail, the phonons arrive at the opposite vertical wall and slide laterally along it until they are stopped by boundary scattering, or collide with the lateral walls, closing the
FIG. 13: The RTA (left) and bRTA (right) steady-state heat fluxes for a limiting case phosphorene-based Levitov configuration with $W_{\text{reservoir}} = 25 \text{ nm}$, $W_{\text{device}} = 46 \text{ nm}$ and $H = 19.2 \text{ nm}$, $T_{\text{HOT}} = 310 \text{ K}$ and $T_{\text{COLD}} = 290 \text{ K}$.

vortex in both cases. In the former, we recall that walls act as phonon sources due to diffusive scattering, and thus they reemit a small part of those incident phonons, which again slide laterally along the edges until they are stopped by edge friction or the collision with the lateral wall forming in that way additional vortices.

Thus, such behavior supports the importance of top and bottom boundary scattering, as for larger devices vortices can start forming before a significant number of phonons can scatter at lateral borders. This latter statement is further confirmed by larger device simulations in which no phonon arrived at a lateral wall but the vortices are formed (see Fig. 15 for an animated version of the time evolution of the heat flux and temperature, see the flux_large.mp4 and temperature_larger.mp4 videos in the Supplemental Material [49], respectively).
FIG. 15: The bRTA heat flux at different times for a large graphene-based Levitov configuration with \( W_{\text{reservoir}} = 37.5 \text{ nm} \), \( W_{\text{device}} = 3.75 \mu \text{m} \) and \( H = 75 \text{ nm} \), \( T_{\text{HOT}} = 310 \text{ K} \) and \( T_{\text{COLD}} = 290 \text{ K} \), initially at 300 K. Only the central part of the device is depicted as most of the device is empty of phonons.

B. Non-ballistic devices

Here we provide results for less ballistic devices than those of Sec. VA, i.e. \( H \gtrsim \lambda \). As aforementioned, owing to the differences in the mean free path and non-local length between graphene and phosphorene, we expect both to show rather distinct vorticity.

1. Phosphorene

Devices out of the ballistic regime are in principle much more interesting in the case of phosphorene, as the non-local length is slightly larger than the mean free path, therefore allowing for hydrodynamic features outside the ballistic regime over a larger range than graphene. Moreover, based on the fit of \( \ell \) to Monte Carlo results found in Ref. [42], we expect its value to be larger than the calculated one, thus allowing for vortices and negative thermal resistances in even much wider devices than the ones suggested by the results in Table 1

Because of this, we have simulated three different heights: 50, 75, and 100 nm, the RTA and bRTA steady-state heat fluxes of which are shown in Figs. 10-18. These results are to be compared to the extrinsic-only solution for the same set of heights (see Figs. 19-21) to highlight the effect of intrinsic scattering on the steady-state properties. Additionally, we provide the bRTA thermal profiles for all heights in Figs. 22-24. In smaller devices, we observe that device geometry allows for the formation of two vortices (see Fig. 19), which are present even when intrinsic scattering is added. Interestingly, if we set aside the expected magnitude differences between all the treatments of the scattering operator, we can observe subtle differences in the vortex profiles. While within the extrinsic treatment we can see two clean vortices, the addition of intrinsic scattering through the RTA causes those vortices to start dissipating at the device’s corners. However, the proper treatment of the scattering leads to slightly different alterations, as such dissipation of vorticity is less pronounced, while the RTA incipient fracture of principal vortices into two becomes clearer in the bRTA case. Therefore, the full linearized scattering operator is necessary to obtain an appropriate description of vorticity.

The 75 and 100 nm results (see Figs. 17 and 18) further support this latter statement for non-ballistic devices. In both cases vorticity is geometrically permitted (see Figs. 20 and 21) but suppressed in the RTA case, which shows an almost perfect diffusive profile, especially for the 100 nm device. On the other hand, the correct de-
The description of the scattering operator allows the vortices to survive in both cases. This fact is nothing but a consequence of the RTA deeming all three-phonon processes as resistive, which translates into lower $\ell$ values. Indeed, we expect the flux direction to be qualitatively similar for devices much larger than $\ell$, as they tend to have diffusive profiles—in which the only difference between the RTA and bRTA is the flux magnitude due to the thermal conductivity differences.

Regarding thermal resistance, we can observe a low-magnitude negative resistance, when compared to ballistic devices, for the 50 device (Figs. 16 and 22), and even a lower one in the 75 nm one (Figs. 17 and 23). For the larger device (i.e. 100 nm, Figs. 18 and 24) no significant negative thermal resistance is observed, indicating (in line with the flux results) a weakening of hydrodynamic regime with increasing $H$. This is a consequence of the phonon distribution losing its inertia—i.e. preventing the effect of boundary scattering—while traveling due to intrinsic resistive scattering, up until the point that for larger distances ($H$) it is completely lost before arriving to the opposite side, and thus one arrives at the Fourier’s diffusive regime.

**2. Graphene**

The graphene RTA and bRTA steady-state heat fluxes are depicted in Fig. 25 showing a clear diffusive (Fourier-like) profile in the RTA case. In contrast, some vorticity can be observed in the bRTA results, indicating a clear underestimation of non-local length—i.e. the ability of the distribution to keep its inertia—because of the RTA limitations. Therefore, it is indeed possible to have some small vorticity out of the ballistic regime. When incrementing the height, we observe that vortices cannot form even without intrinsic scattering (see Fig. 26) although some non-local effects appear in the flux, namely the curvature of the isolines near the corners of Fig. 26. However, such hydrodynamic effects are not present neither in the RTA nor bRTA (see Fig. 27) due to the intrinsic scattering effect (i.e. $\ell \ll H$), showing both a Fourier-like profile.

**VI. SUMMARY AND CONCLUSIONS**

In this work, we have presented *ab initio* calculations of hydrodynamic signatures in thermal transport for graphene and phosphorene-based devices using phonon properties obtained from first-principles to solve the phonon Boltzmann transport equation via energy-based deviational Monte Carlo methods. We find such signatures at room temperature in quasiballistic and non-
ballistic regimes, in contradiction with the predictions of classical hydrodynamic models, but in agreement with newer and more generalized models. In contrast to previous observations, we demonstrate that there is a shared mechanism among all transport regimes originating the hydrodynamic signatures, namely boundary scattering. Indeed, we show that in our devices boundary scattering at horizontal edges is the leading mechanism behind vortices, proving through the solution of the hydrodynamic equation that an increment in the horizontal dimension only leads to the formation of more convection cells instead of vortex (hydrodynamics) destruction. We have also illustrated the importance of the dimensions, especially the distance between sources and the opposite vertical walls (i.e. the ones causing the vortices), in the presence of hydrodynamic effects; concluding that those can exist even under the presence of significant intrinsic scattering (i.e. non-ballistic regime) as this cannot instantly randomize the flux but allows for certain inertia. Moreover, we have demonstrated the importance of making a proper description of the intrinsic scattering in...
the thermal vortex profiles, even well inside the ballistic regime, showing that the RTA can lead to an incorrect number of vortices or no vorticity at all, in opposition to the solution using the full linearized scattering operator.

Finally, based on our calculations, we point out that experimental observation of hydrodynamic signatures at room temperature would be favored by choosing a small graphene sample under 100 nm in height, a finite source and sink as narrow as possible and a device width at least three times larger than the source/sink widths.

FIG. 22: The bRTA steady-state temperature profile for phosphorene-based Levitov configuration with $W_{\text{reservoir}} = 25$ nm, $W_{\text{device}} = 160$ nm and $H = 50$ nm, $T_{\text{HOT}} = 310$ K and $T_{\text{COLD}} = 290$ K.

FIG. 23: The bRTA steady-state temperature profile for phosphorene-based Levitov configuration with $W_{\text{reservoir}} = 25$ nm, $W_{\text{device}} = 160$ nm and $H = 75$ nm, $T_{\text{HOT}} = 310$ K and $T_{\text{COLD}} = 290$ K.

FIG. 24: The bRTA steady-state temperature profile for phosphorene-based Levitov configuration with $W_{\text{reservoir}} = 25$ nm, $W_{\text{device}} = 160$ nm and $H = 100$ nm, $T_{\text{HOT}} = 310$ K and $T_{\text{COLD}} = 290$ K.

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FIG. 25: The RTA (right) and bRTA (left) steady-state heat fluxes (right) for graphene-based Levitov configuration with $W_{\text{reservoir}} = 37.5$ nm, $W_{\text{device}} = 375$ nm and $H = 170$ nm, $T_{\text{HOT}} = 310$ K and $T_{\text{COLD}} = 290$ K.

FIG. 26: The extrinsic steady-state heat fluxes (right) for graphene-based Levitov configuration with $W_{\text{reservoir}} = 37.5$ nm, $W_{\text{device}} = 375$ nm and $H = 340$ nm, $T_{\text{HOT}} = 310$ K and $T_{\text{COLD}} = 290$ K.

FIG. 27: The RTA (right) and bRTA (left) steady-state heat fluxes (right) for graphene-based Levitov configuration with $W_{\text{reservoir}} = 37.5$ nm, $W_{\text{device}} = 375$ nm and $H = 340$ nm, $T_{\text{HOT}} = 310$ K and $T_{\text{COLD}} = 290$ K.
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