

***Ab initio* study of Coulomb drag driven electron-hole bifluidity in doped graphene**Dwaipayan Paul^{✉,*}, Elena Trukhan,[†] and Nakib H. Protik^{✉,‡}*Institut für Physik and CSMB, Humboldt-Universität zu Berlin, Berlin, Germany*

(Received 15 December 2025; revised 8 June 2026; accepted 23 June 2026; published 20 July 2026)

Motivated by the notion that a preponderance of Coulomb interactions might lead to hydrodynamics, we carry out an *ab initio* calculation of the charge-carrier transport properties of the electron-hole gas of doped graphene. We include both the phonon and Coulomb interactions within a momentum and band-resolved Boltzmann transport formalism. We find that under suitable conditions, the strong Coulomb drag effect induces phenomena like negative conductivity and joint electron-hole hydrodynamics (bifluidity) in the system. We also identify the exclusive electron or hole hydrodynamics. We find that there is a strong violation of the Wiedemann-Franz law in the low-doped regimes. Our work elucidates the roles of the microscopic scattering mechanisms that drive the hydrodynamic phenomena.

DOI: [10.1103/q4mx-9tmn](https://doi.org/10.1103/q4mx-9tmn)

Introduction. The hydrodynamic transport has been a long-standing topic of curiosity in the condensed matter and the broader materials science communities. It is a low-dissipation mode of transport, characterized by a large number of scattering events that, however, are (quasi)momentum conserving. In such a nonequilibrium state, a system of particles drifts together with a characteristic global velocity, akin to how water flows through a pipe. This mode of transport lies between the low-temperature ballistic and the high-temperature diffusive regimes, and has been rather difficult to observe and control in 3D systems, usually due to the inevitable presence of impurity interactions that tend to destroy momentum. Two-dimensional (2D) materials, on the other hand, allow a way to introduce charge carriers via electrostatic gating, i.e., without the need to introduce dopants. As such, these systems are well suited for studying hydrodynamics.

Now, graphene [1], since its discovery, has been a fertile ground for studying the properties of 2D materials. This is partly because it is easy and inexpensive to obtain, and for its superior charge-carrier tunability. It can sustain high charge conduction states [2,3]. And, being a semimetal, it simultaneously hosts both an electron and a hole subsystem. Pertinent to this work is the fact that the charge-carrier hydrodynamics in this material has been predicted in several experimental and theoretical studies [4–11]. Previous works [4,7,11] established the formation of strongly interacting Dirac fluid at the charge-neutrality point (CNP), which breaks the Fermi-liquid picture and exhibits a strong violation of the Wiedemann-Franz law

[4,12,13]. Adding to the complexity, there is also the formation of local impurities called charge puddles [14,15], which provide a strong scattering channel for the charge carriers at low temperatures. Our focus here is on doped graphene closer to room temperature where we can safely ignore charge puddle effects [16,17]. Furthermore, the existence of hydrodynamics was predicted to be not only in the Dirac fluid regime, but also in the Fermi-liquid regime [17]. The latter is the focus of our work.

From a theoretical point of view, it is interesting to ask what types of scattering processes dictate the charge conductivity of graphene. Interactions of the charge carriers with phonons? With the sample boundary? Interactions between the charge carriers via Coulomb interactions? Since graphene hosts both an electron and a hole gas, it is important to ask whether the alleged hydrodynamic state is only in one or the other subsystem? Or is it of a more exotic kind where both the electron and hole subsystems share a joint global drift velocity, forming what is called an electron-hole bifluid?

So far, answers to such questions necessarily had to employ semiempirical transport theories [7,17,18] that contain simplified assumptions and tunable parameters. In fact, current publicly available versions of state of the art *ab initio* Boltzmann electronic transport equation (BTE) solvers [19–22] do not provide the functionality to include the interparticle Coulomb interactions in the collision integral. We are aware of one *ab initio* study on doped silicon for which, however, the software is not publicly available [23]. In that work, the authors include both the electron-electron and electron-plasmon interactions, albeit through a relaxation-time approximation (RTA) solution of the BTE. Capturing the hydrodynamic transport physics, however, necessarily requires the full solution of the BTE, or, at least, going beyond the RTA, in order to fully capture the effect of the momentum feedback loop between the two interacting subsystems. To fill the existing methodological and knowledge gaps, we implement the necessary computational tools in the `elphbolt` [21] code to enable an *ab initio* study of the role of the Coulomb interactions

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on the charge conduction properties of real materials. Using this developed tool, we find that the electron-hole plasma in doped graphene can sustain three types of hydrodynamic states, one of which is the electron-hole bifluid. We find that these hydrodynamic phenomena are driven by the Coulomb interactions between the charge carriers, and that the standard phonon-limited carrier transport theory fails to capture the essential physics, both quantitatively and qualitatively. We further observe a strong Coulomb drag induced negative conductivity, corroborating recent measurements reported in Ref. [24]. Last, we observe a strong violation of the Wiedemann-Franz (WF) law close to the bifluid state.

Results and discussion. We operate within the linear response regime where the system is driven slightly out of equilibrium by an external electric field $\mathbf{E}$ or a temperature gradient ∇T . The state of a particle is tagged by a band index n_1 and wave vector $\mathbf{k}_1$, which we write together as 1 . The nonequilibrium distribution of a state is denoted by f_1 , which deviates from the equilibrium, Fermi-Dirac counterpart f_1^0 , by a small amount. This small deviation from equilibrium is written in the following form:

$$\delta f_1 = -f_1^0(1 - f_1^0)\beta(\nabla T \cdot \mathbf{I}_1 + \mathbf{E} \cdot \mathbf{J}_1), \quad (1)$$

where $\mathbf{I}_1$ and $\mathbf{J}_1$ are the response functions to the corresponding applied field and β is the inverse temperature energy. BTEs can be written in terms of these response functions, and the solutions then give access to the various transport coefficients, expressions for which have previously been given in Ref. [21].

In this work, we focus on capturing the drag effect between electrons and holes, which is the bidirectional flow of momentum between these two systems of charge carriers under nonequilibrium conditions. Since this effect is mediated by the Coulomb interaction, it also goes by the name Coulomb drag in the literature [25]. (Generally, the Coulomb drag is defined as the drag between the charge-carrier systems living on macroscopically separated materials. Graphene can be thought of as a special case where the two systems of charge carriers—electron and holes—coexist in the same material sample.) Now, in order to compute this effect within an *ab initio* framework, we extend the `elphbolt` code to include a screened Coulomb collision integral. This collision integral has been derived from first principles by Sanborn in Ref. [26] for the case of doped, 3D semiconductors, and here we simply adapt it for our 2D case. In this approximation, the exchange-interaction term is ignored. We also ignore the local field effects. In our notation, the linearized collision integral is given by

$$\begin{aligned} I_1^{\text{Coul}}[\mathbf{J}] = & \frac{4\pi}{\hbar f_1^0(1 - f_1^0)A^2} \sum_{23} \sum_{\mathbf{G}} |U_3 U_1^\dagger|^2 \left| \frac{V_{\mathbf{q}}^{2\text{D},\infty}(\mathbf{G})}{\epsilon_{\mathbf{q}}^{\text{TF}}(\mathbf{G})} \right|^2 \\ & \times \delta(E_1 + E_2 - E_3 - E_4) f_1^0 f_2^0 (1 - f_3^0) \\ & \times (1 - f_4^0)(\mathbf{J}_1 + \mathbf{J}_2 - \mathbf{J}_3 - \mathbf{J}_4) \cdot \mathbf{E}, \end{aligned} \quad (2)$$

where $\hbar$ is the reduced Planck's constant, E is the energy, $\mathbf{q} \equiv \mathbf{k}_1 - \mathbf{k}_3$, $\hbar\omega \equiv E_1 - E_3$, A is the area of the crystal, $V_{\mathbf{q}}^{2\text{D},\infty}$ is the 2D bare Coulomb interaction [27] screened by a high-frequency dielectric, and $U_1^\dagger$ is the diagonalizer of the Wannier interpolated Hamiltonian in state 1. Note that the

wave vector of state 4 is determined by those of the remaining three. We choose the value of ϵ^∞ to be the out-of-plane dielectric of hBN (4.1) [28] to model the experimental conditions of the work in Ref. [24]. $\epsilon_{\mathbf{q}}^{\text{TF}}$ is the Thomas-Fermi dielectric, given by

$$\epsilon_{\mathbf{q}}^{\text{TF}}(\mathbf{G}) = 1 + \frac{e^2\beta}{2\epsilon_0 A} \sum_{\mathbf{l}} \frac{f_{\mathbf{l}}^0(1 - f_{\mathbf{l}}^0)}{|\mathbf{q} + \mathbf{G}|}, \quad (3)$$

where e is the magnitude of the electronic charge and ϵ_0 is the permittivity of free space. The corresponding expression for the temperature gradient field case is found through the replacement ($\mathbf{J} \rightarrow \mathbf{I}$, $\mathbf{E} \rightarrow \nabla T$).

The Coulomb collision integral is added to the electron-phonon counterpart [21] assuming Matthiesen's rule, i.e., that the scattering channels are independent. The final charge-carrier BTE is fully solved to self-consistency using an iterative approach. That is, our approach is beyond any type of RTA. Doing so is crucial in order to accurately capture any hydrodynamics. Details of our *ab initio* BTE solution method have been given before in Ref. [21] and are not reproduced here. But, briefly, this approach relies on the density functional+perturbation theory as implemented in the Quantum Espresso code [29,30], maximally localized Wannier functions [31], and the real-space, Wannier representations of the electron-phonon interaction matrix elements obtained by interfacing with the EPW code [32,33]. Details of the employed computational methods are given in Supplemental Material (SM) [34].

Since we compare to the measurements in Ref. [24], we include a boundary scattering term in all our calculations using a simple, phenomenological scattering rates expression: $|\mathbf{v}_1|/L$, where $\mathbf{v}_1$ is the charge-carrier group velocity and $L = 20 \mu\text{m}$ characterizes the sample size. This expression is added to the rest of the RTA scattering rates using, again, Matthiesen's rule. We ignore any in-scattering corrections from this scattering channel.

In this work, we choose the majority carrier concentrations to range between 1.6×10^{11} and $1.7 \times 10^{12} \text{ cm}^{-2}$. Getting any closer to the CNP is not possible at the moment due to the extreme computational complexity; see SM [34]. In any case, the application of the semiclassical BTE is invalid near the CNP, as mentioned in the Introduction.

In Fig. 1, we present our calculated room-temperature resistivity as a function of the difference of the electron and hole concentrations, $n_e - n_h$. We include a comparison to recent 300 K measurements from Ref. [24] (green line). We find that without the inclusion of the Coulomb interactions (empty disks), the calculated resistivity is in poor agreement with the measurements (green line vs green empty disks) over the entire range of concentrations considered. The theory including the Coulomb interactions gives quantitatively good agreement (green line vs green solid disks) in the moderate to high concentration range. For low concentrations, however, the Coulomb enabled theory overestimates the resistivity. This could be due to a couple of reasons. First, the lattice constant used in our theoretical modeling might not match the one of the sample in Ref. [24] since the graphene flake there is sandwiched between hBN substrates. We take into account the role of hBN only through an extra screening factor, as mentioned

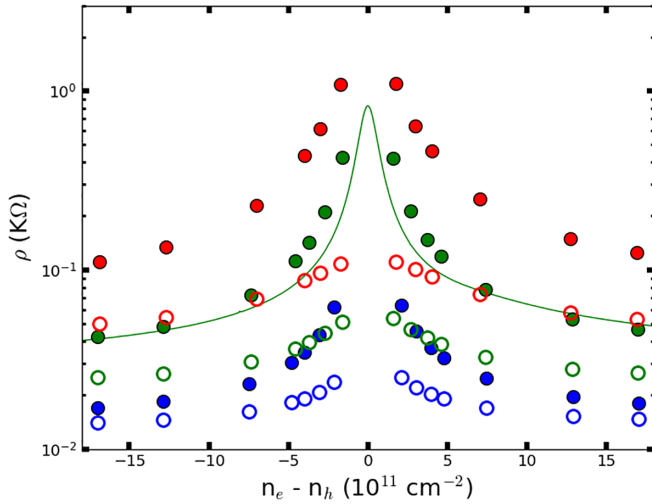


FIG. 1. Resistivity vs charge-carrier concentration at various temperatures. Positive (negative) sector of the horizontal axis corresponds to majority electron (hole) concentration. Solid green line gives the 300 K measurements from Ref. [24] (data obtained from the primary author via private communication and converted to resistivity units). Calculated values are presented in disks for three temperatures: 200 K (blue), 300 K (green), and 400 K (red). Solid (empty) symbols correspond to the theory that includes (excludes) the Coulomb interactions.

earlier. Second, our Coulomb collision integral includes only the direct interaction term. Our theoretical predictions might be improved through the inclusion of the exchange term, since the direct and exchange terms have opposite signs. We leave this for a future work. Now, the transport coefficients presented in Fig. 1, being band-summed quantities, hide certain crucial failures of the Coulomb-free theory. And, in order to observe these failures of the phonon-limited transport to capture important microscopic physics, we next look at the valence- and conduction-band contributions separately.

In Fig. 2, we give the spectral conductivities at 200 K for a high (thick lines) and a low (thin lines) hole-doped case. The Dirac point is placed at the zero of the horizontal axis. Note that the conduction-band contribution is plotted with a scaling factor of $10\times$. For the Coulomb-free case (dashed lines), both the valence and the conduction bands give positive contributions to the conductivity, as one would expect from a classical Drude picture. In stark contrast, the activation of the Coulomb interactions (solid lines) leads to a significantly lower positive contribution from the valence band, along with a *negative* contribution from the conduction band. A completely analogous effect happens for the electron-doped case, where the inclusion of the Coulomb interactions causes the holes to give a negative contribution to the conductivity; see SM [34]. There, we also show that this effect persists even at room temperature. What we described so far is the evidence for an extreme Coulomb drag effect between the electron and hole systems. The essential physics we observe here is that the majority carriers transfer momentum to the minority ones, causing them to develop a current in a direction opposite to what is dictated by the coupling to the external electric field. On the other hand, the minority carriers also drag the majority carriers in the opposite direction, causing them to give a lower,

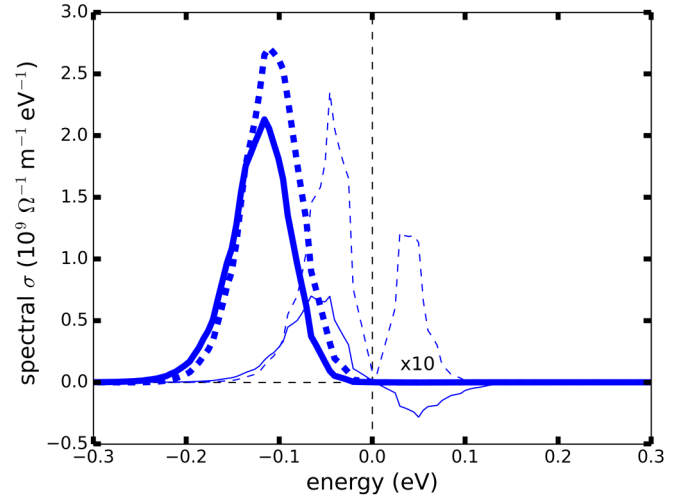


FIG. 2. The 200 K spectral conductivity from the valence and conduction bands for hole doped cases. Thick (thin) lines are for a majority hole concentration of 1.3×10^{12} (2×10^{11}) cm^{-2} . Solid (dashed) lines denote the theory including (excluding) the Coulomb interactions. The Dirac point is set at zero energy. The contribution from the conduction band (positive energy) is displayed with a factor of 10 enlargement.

albeit still positive, contribution. This extreme Coulomb drag effect becomes more prominent the closer one gets to the CNP. Note that this phenomenon has been experimentally observed in Ref. [24].

It is now interesting to inquire whether the mode of this charge transport is hydrodynamic. This question is motivated by the prevailing wisdom that the Coulomb scatterings between the charge carriers contain a large proportion of the momentum conserving (Normal) type processes. Now, it is well-known that in 2D systems the decay of the screened Coulomb potential goes as r^{-1} for small distance r as opposed to exponentially (Yukawa type) as in 3D, making the former a long-range interaction [35]. However, having a long-range Coulomb interaction alone is not a sufficient condition for the hydrodynamic behavior. The scattering events must also be predominantly of the Normal type. In such a case, the nonequilibrium distribution function takes the form of a shifted Fermi distribution, the shift amount being characterized by a global drift velocity. We investigate this next.

In Figs. 3(a)–3(c), we plot for the 200 K case the band-resolved, normalized deviation function Δ_1/β where $\Delta_1 \equiv \delta f_1/f_1^0/(1 - f_1^0)$. (This definition is the fermionic analog of the bosonic one given in Ref. [36].) We do this along a section of the $(\mathbf{K}-k_x)-\mathbf{K}-(\mathbf{K}+k_x)$ path of the Brillouin zone measured from the $\mathbf{K}$ point, for three different majority hole-doped cases. Here, k_x is proportional to the unit vector in the x direction. The electric field is set at 1 eV/nm/C, in both x and y directions. We observe that this function is strongly linear, either for one or both of the electron and hole subsystems, only when the Coulomb interactions are turned on (solid disks and solid lines). Note that the linearity of Δ_1 is the signature of the hydrodynamic transport regime. We notice that as we go from the high doped [Fig. 3(c), furthest from charge neutrality] toward low doped [Fig. 3(a), closest to charge neutrality],

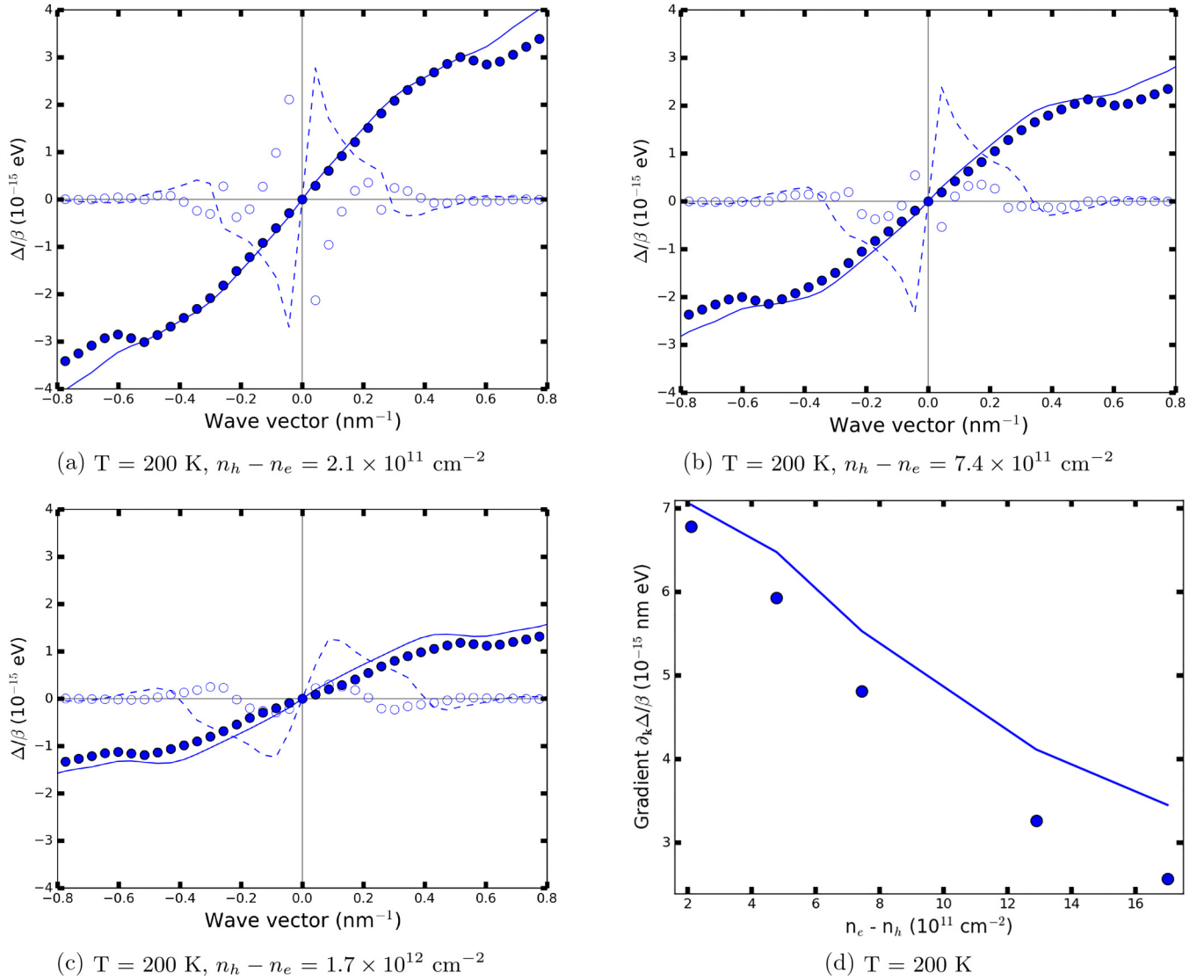


FIG. 3. (a)–(c) Normalized deviation function Δ/β vs magnitude of electron wave vector, measured from $\mathbf{K}$ point and along the $(\mathbf{K}+k_x)$ – $\mathbf{K}$ – $(\mathbf{K}+k_x)$ direction of the Brillouin zone. (d) Gradient $\partial_k \Delta$ near the $\mathbf{K}$ point for the valence and conduction bands. In all plots, lines represent the valence band and disks represent the conduction band. Solid lines and symbols are for the Coulomb enabled theory, whereas dashed lines and empty symbols are for the Coulomb-free theory.

both the valence (solid line) and conduction (solid disks) bands tend toward having a robustly linear Δ_1 . That is, by tuning the doping toward charge neutrality, both the electron and hole subsystems go hydrodynamic. On the other hand, as we move further away from the charge-neutrality condition, we retain robust hydrodynamics in the holes but begin to lose it in the electrons. Now, the most interesting result is that in the low-doped limit [Fig. 3(a)], the electron and hole subsystems tend toward a *shared* hydrodynamic state—the electron-hole bifluid. To quantify the last statement, we plot in Fig. 3(d) the gradient $\partial_k \Delta_1$ taken near the $\mathbf{K}$ point, along the same Brillouin-zone path. We see that the slopes for both the valence and the conduction bands approach a common value in the low-doping limit. This is most pronounced for the low-temperature case. A completely analogous situation occurs for the majority electron-doped cases, as shown in SM [34]. We also show there that with increasing temperature, this phenomenon becomes less pronounced. The bottom line is

that the electron-hole bifluidity in doped graphene manifests in the low-temperature and low-doping situations. The simpler theory that lacks the Coulomb interactions does not capture this essential physics (dashed lines and empty disks).

Finally, in Fig. 4, we plot the Lorentz number $\mathcal{L} = \kappa_{\text{el}}(\sigma T)^{-1}$ scaled by the universal (Sommerfeld) value $\mathcal{L}_0$ as a function of $n_e - n_h$, for three temperature points. Here, κ_{el} is the electronic thermal conductivity in the open-circuit condition and σ is the charge conductivity. The prevailing wisdom is that very close to the CNP in graphene at low temperatures, a violation of the WF law occurs due to the domination of the electron-hole interactions, which results in the thermal resistivity to vanish for a finite electric current [4]. Here, we find that when the Coulomb interactions are included (solid blue, green, and red lines), there is a strong violation of the WF law even in the electron- and hole-doped regimes. However, this violation increases as we approach the charge neutrality, consistent with the conclusion made in Ref. [4].

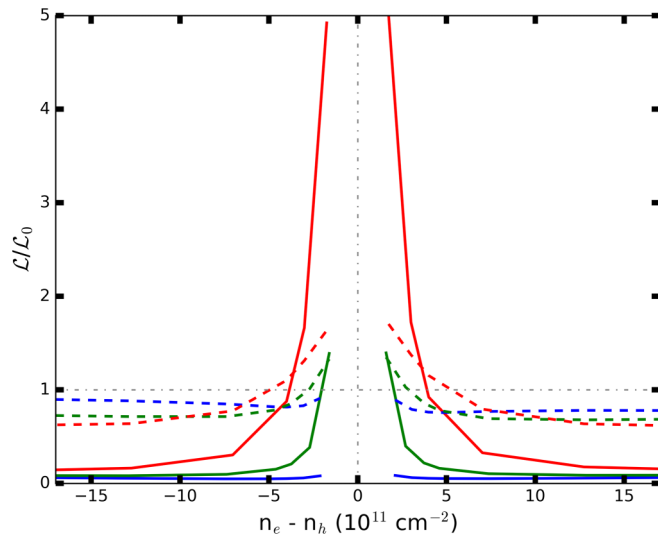


FIG. 4. Scaled Lorentz number vs carrier concentration. Blue, green, and red symbols are for 200, 300, and 400 K, respectively. Solid (broken) lines correspond to the theory including (excluding) the Coulomb interaction. The scaling is with respect to the universal value ($\mathcal{L}_0 \equiv 2.44 \times 10^{-8} \text{ V}^2 \text{ K}^{-2}$) according to the WF law.

This, however, does not correlate with the bifluidity, which we found to be a dominant effect at low temperatures, while here, the WF law violation increases at high temperatures. The theory that ignores the Coulomb interactions (dashed blue, green, and red lines) also shows a violation of the WF law, though much weaker than what is predicted by the better theory.

Summary and outlook. In this work we implement an *ab initio* method to include the Coulomb interactions among the charge carriers within the `elphbolt` Boltzmann transport codes suite. We use this tool to compute the charge-carrier transport properties of doped graphene. We find that the Coulomb interaction-enabled theory predicts three hydrodynamic transport regimes: (1) exclusive hole hydrodynamics in the high hole-doped case, (2) exclusive electron hydrodynamics in the high electron-doped case, and (3) coupled electron-hole bilfluidity in the weak-hole or electron-doped cases at low temperature. Our results corroborate the recent observations of negative conductivity in the extreme Coulomb drag regime [24]. We also find a strong violation of the WF law that is driven by the Coulomb interactions between electrons and holes. None of the above-mentioned phenomena is captured by the theory that includes the electron-phonon interactions, but ignores Coulomb.

Although the *ab initio* methodology described in this work is able to predict the strong Coulomb drag driven negative

conductivity and the electron-hole bilfluidity, we see several avenues for improvement and extensions that we wish to touch upon in future works. We briefly discuss them below.

(1) Exchange term: As mentioned earlier, our collision integral contains only the direct term. Since the direct and exchange terms come with opposite signs, we think that the inclusion of the latter will reduce calculated resistivity values at low doping levels, and result in theoretical predictions that are in better agreement with measurements.

(2) Dynamical screening: In this work, we employ the Thomas-Fermi screening model. One can, in fact, use a dynamical screening model within the random-phase approximation (RPA) [26]. The RPA includes the effect of the plasmons in the Coulomb kernel and, in principle, captures the screening better than the static Thomas-Fermi theory.

(3) Electron-phonon drag: In this work, we assume that the phonon system can be taken to remain in equilibrium. We do this for two reasons. First, we notice that the transport physics in the doping and temperature ranges considered here is entirely dictated by the Coulomb interactions, as is evidenced from the failure of the theory that ignores these. Second, computing the phonon thermal conductivity in graphene is a difficult and contentious matter. It has been argued that including the extremely computationally demanding four-phonon interactions is necessary to make decent lattice thermal conductivity predictions. That said, there might be other systems where the extremely expensive four-phonon interactions do not have to be considered. In such systems, it might be possible to carry out a coupled electron-phonon BTEs calculation including the Coulomb interactions. We note that in Ref. [37], the authors predicted a joint electron-phonon hydrodynamics in doped MoS_2 . An interesting research question would then be whether there exist materials that, under suitable conditions, exhibit both the electron-hole and electron-phonon hydrodynamics.

Acknowledgments. This work was funded by the Deutsche Forschungsgemeinschaft (DFG, German Research Foundation) through the Emmy Noether research grant (Grant No. 534386252). This work used computational resources provided by Noctua2 at the Paderborn Center for Parallel Computing (PC2), JUWELS at the Jülich Supercomputing Centre, and the CMS cluster of Humboldt-Universität zu Berlin. We thank Dr. Leonid A. Ponomarenko for kindly providing the resistivity measurement from Ref. [24].

Data availability. The theory results supporting the findings of this work are available at Ref. [38]. The `elphbolt` code used to carry out the calculations is free/libre and open source software.

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