Supplementary Material to the article "Thermopower of graphene under strong electron-hole scattering"

I. COLLISION INTEGRALS IN THE HYDRODYNAMIC REGIME AND DERIVATION OF THE EULER EQUATIONS

In the hydrodynamic regime the dominance of intra-carrier collisions forces the corresponding integrals to vanish, $\mathcal{C}_{e\text{-}e,\,h\text{-}h} = 0$. Owing to the conservation of particle number, momentum, and energy in intra-species collisions, this is achieved by the shifted Fermi–Dirac distribution. For electrons,

$$f_e^{hd} = \left[1 + \exp\left(\frac{\mathcal{E}_e - \mathbf{p}\cdot\delta\mathbf{u}_e - \mu_e}{k_BT}\right)\right]^{-1}. \tag{S1}$$

At small drift velocities, $|\mathbf{p}\cdot\delta\mathbf{u}_{e,h}| \ll k_BT$, it expands as $f_e^{hd} \approx f_e^0 + \delta f_e^{hd}$ with δf_e^{hd} given by Eq. (2) of the main text. Under this hydrodynamic distribution the remaining collision integrals can be linearized around equilibrium f_e^0 with respect to the drift velocities:

$$\mathcal{C}_{e\text{-}ext} = -\delta\mathbf{u}_e\cdot\mathbf{S}_{ext}[f_e^0], \qquad \mathcal{C}_{e\text{-}h} = -\left(\delta\mathbf{u}_e - \delta\mathbf{u}_h\right)\cdot\mathbf{S}_{e\text{-}h}[f_e^0, f_h^0], \tag{S2}$$

with the coefficients

$$\mathbf{S}_{ext}(\mathbf{p}) = \frac{f_e^0\left(1-f_e^0\right)}{k_BT}\,\frac{\mathbf{p}}{\tau_p(\mathcal{E})}, \qquad \mathbf{S}_{e\text{-}h}(\mathbf{p}) = \frac{g}{k_BT}\int W(q)\, f_e^0(\mathbf{p})\, f_h^0(\mathbf{p}_1)\left[1-f_e^0(\mathbf{p}-\mathbf{q})\right]\left[1-f_h^0(\mathbf{p}_1+\mathbf{q})\right]\mathbf{q}\,\frac{d^2\mathbf{q}\,d^2\mathbf{p}_1}{(2\pi\hbar)^4}, \tag{S3}$$

and symmetrically for holes. Here $W(q)$ is the golden-rule probability of screened Coulomb scattering, including the chiral overlap factors and the energy-conservation δ-function [1], and $\tau_p(\mathcal{E})$ is the relaxation time of the external channel, whose appearance in $\mathbf{S}_{ext}$ is established below. Both coefficients are positive and carry the same structure – the thermal blocking factors $f_{e,h}^0(1-f_{e,h}^0)/k_BT$ multiplied by the transferred momentum and the scattering rate.

The parameters $\delta\mathbf{u}_{e,h}$ are determined by the first moments of the kinetic equations (1) of the main text: multiplying by $\mathbf{p}$ and integrating over $d\Gamma_p = g\,d^2\mathbf{p}/(2\pi\hbar)^2$ turns the left-hand side into the driving force $\frac{1}{2}c_e\nabla T - en_e\nabla V$, with $\frac{1}{2}c_eT = \frac{3}{2}n_e\overline{\mathcal{E}_e} - n_e\mu_e$, and the moments of the integrals (S2) into the friction forces $-\rho_e/\tau_{e\text{-}ext}\cdot\delta\mathbf{u}_e$ and $-\rho_e/\tau_{e\text{-}h}^{tr}\cdot(\delta\mathbf{u}_e - \delta\mathbf{u}_h)$, where the times are defined as

$$\frac{\rho_e}{\tau_{e\text{-}ext}} = \frac{1}{2}\int \mathbf{p}\cdot\mathbf{S}_{ext}\,d\Gamma_p, \qquad \frac{\rho_e}{\tau_{e\text{-}h}^{tr}} = \frac{1}{2}\int \mathbf{p}\cdot\mathbf{S}_{e\text{-}h}\,d\Gamma_p, \tag{S4}$$

the factor $\frac{1}{2}$ follows from the tensor identity $\int p_iS_j\,d\Gamma_p = \frac{1}{2}\delta_{ij}\int\mathbf{p}\cdot\mathbf{S}\,d\Gamma_p$ of an isotropic system. Momentum conservation in inter-carrier collisions gives $\int\mathbf{p}\,\mathcal{C}_{e\text{-}h}\,d\Gamma_p = -\int\mathbf{p}\,\mathcal{C}_{h\text{-}e}\,d\Gamma_p$, binding the inter-carrier times together as $\rho_e/\tau_{e\text{-}h}^{tr} = \rho_h/\tau_{h\text{-}e}^{tr}$ (in fact, Newton's third law for the e and h fluids). This completes the derivation of Eq. (4) of the main text.

A. External channel

An arbitrary perturbation can be decomposed over energy shells ($p = \mathcal{E}/v_F = \text{const}$) and angular harmonics on each shell, $\delta f(p,\varphi) = \sum_l a_l(\mathcal{E})e^{il\varphi}$. Scattering off static impurities is elastic, and scattering off acoustic phonons is quasi-elastic (owing to sound velocity $s \ll v_F$), so the linearized external operator $\mathcal{C}_{e\text{-}ext}[\delta f]$ does not couple different energy shells. This fact together with the isotropy of both disorder and phonon systems (so the scattering probability $W = W(\mathcal{E},\theta)$ with $\theta = \varphi - \varphi'$) leads to the diagonalization of the external collision operator by the angular harmonics:

$$\mathcal{C}_{e\text{-}ext}\left[a_l(\mathcal{E})\,e^{il\varphi}\right] = -\frac{a_l(\mathcal{E})\,e^{il\varphi}}{\tau_l(\mathcal{E})}, \qquad \frac{1}{\tau_l(\mathcal{E})} = \int W(\mathcal{E},\theta)\,(1-\cos l\theta)\,d\Gamma_{p'}. \tag{S5}$$

The hydrodynamic correction δf_e^{hd} is a drift harmonic ($l=1$), so it is an eigenfunction of $\mathcal{C}_{e\text{-}ext}[\delta f]$ with the eigenvalue given by the scattering rate, $\tau_{l=1}^{-1} \equiv \tau_p^{-1}(\mathcal{E}) = \int W(1-\cos\theta)\,d\Gamma_{p'}$. Therefore, the relaxation time form of the linearized external collision integral is not an approximation, but an exact form under the hydrodynamic perturbation:

$$\mathcal{C}_{e\text{-}ext} = -\frac{\delta f_e^{hd}}{\tau_p(\mathcal{E})}. \tag{S6}$$

This reproduces the coefficient $\mathbf{S}_{ext}$ of (S3) with $-\partial f_e^0/\partial\mathcal{E} = f_e^0(1-f_e^0)/k_BT$. For acoustic phonons $\tau_p^{-1}(\mathcal{E}) = D^2 k_B T\mathcal{E}/(4\rho_s s^2\hbar^3 v_F^2)$ [2], so the expression (S4) reduces to a thermal average,

$$\frac{1}{\tau_{e\text{-}ext}} = \frac{D^2 k_B T}{3\rho_s s^2 v_F^2 \hbar^3}\cdot\frac{\overline{\mathcal{E}_e^2}}{\overline{\mathcal{E}_e}}, \tag{S7}$$

with D standing for the deformation potential, ρ_s for the sheet mass density of graphene, and s for the sound velocity.

B. Inter-carrier channel

No analogous reduction can be made for the electron-hole channel: these collisions exchange energy $\omega \sim k_BT$ between the species, so the linearized operator couples different energy shells and depends on both distribution functions. Its moment (S4), evaluated in the Born approximation with the dynamically screened (random phase approximation, RPA) Coulomb interaction, takes the standard fluctuation form of the mutual-drag coefficient (here we used natural units $\hbar = v_F = 1$) [3]:

$$\frac{\rho_e}{\tau_{e\text{-}h}^{tr}} = \frac{2}{\pi}\int\frac{d^2\mathbf{q}}{(2\pi)^2}\int\limits_{-\infty}^{+\infty} d\omega\,\frac{q^2}{4k_BT}\,\frac{V_0^2(q)}{|\varepsilon(q,\omega)|^2}\left[\operatorname{Im}\Pi_{cc}\operatorname{Im}\Pi_{vv} + \operatorname{Im}\Pi_{cv}^2\right] n_\omega\,(n_\omega+1)\,, \tag{S8}$$

where $V_0(q) = 2\pi e^2/(\kappa_0 q)$ is the bare Coulomb interaction with the background permittivity κ_0, $n_\omega = [e^{\omega/k_BT}-1]^{-1}$ is the Bose function, $\Pi_{\lambda\lambda'}(q,\omega)$ are the polarization operators for transitions between the conduction and valence bands $\lambda,\lambda' = \{c,v\}$, and $\varepsilon(q,\omega) = 1 + V_0(q)\sum_{\lambda\lambda'}\Pi_{\lambda\lambda'}$ is the RPA dielectric function of graphene. The term $\operatorname{Im}\Pi_{cc}\operatorname{Im}\Pi_{vv}$ describes direct Coulomb scattering of electrons off holes, and $\operatorname{Im}\Pi_{cv}^2$ – exchange scattering. The integral (S8) is evaluated numerically at each μ and, together with (S7), provides the μ-dependent scattering times used in all figures of this work.

II. TRANSPORT COEFFICIENTS AND RELATION TO THE GRTA

On the hydrodynamic (shifted Fermi-Dirac) distributions (S1), since $\delta\mathbf{u}_{e,h}$ are the drift velocities of e and h fluids, the charge and heat fluxes acquire simple forms: $\mathbf{j} = e(n_h\delta\mathbf{u}_h - n_e\delta\mathbf{u}_e)$ and $\mathbf{j}^Q = \frac{1}{2}T(c_h\delta\mathbf{u}_h + c_e\delta\mathbf{u}_e)$. The latter expression is a consequence of the relation between areal heat capacity and the thermodynamic averages already given before $\frac{1}{2}c_eT = \frac{3}{2}n_e\overline{\mathcal{E}_e} - n_e\mu_e$; it means that the areal entropy for each carrier evaluates to $s_{e,h} = \frac{1}{2}c_{e,h}$, as transported physical value for the heat flux is actually $s_{e,h}\cdot T$. Extracting $\delta\mathbf{u}_{e,h}$ from the Euler system in terms of ∇V and ∇T brings the fluxes to the linear forms $\mathbf{j} = -\sigma\nabla V - a\nabla T$ and $\mathbf{j}^Q = -\varkappa\nabla T + \Pi\mathbf{j}$, from which the phenomenologically defined thermopower $S = a/\sigma$ and thermal conductivity $\varkappa$ readily take the forms of Eqs. (5) and (10) of the main text.

In the GRTA the transport coefficients are obtained along the same route, with one structural difference. With the model integrals, defined in Eq. (11) of the main text, substituted into the kinetic equations, the corrections $\delta f_{e,h}$ are expressed through external fields ∇V, ∇T and the parameters $\delta\mathbf{u}_{e,h}$. These velocities in turn are bounded with the external fields by momentum conservation in intra-carrier collisions, $\int\mathbf{p}\,\mathcal{C}_{e\text{-}e,\,h\text{-}h}\,d\Gamma_p = 0$. The linear system resulting from these momentum conservations is structurally identical to the hydrodynamic Euler equations. However, $\delta f_{e,h}$ are no longer of the hydrodynamic form and $\delta\mathbf{u}_{e,h}$ are not drift velocities, so the fluxes must be evaluated directly from their definitions $\mathbf{j}_e^Q = \int(\mathcal{E}-\mu_e)\,\mathbf{v_p}\,\delta\mathbf{f_e}\,\mathbf{d\Gamma_p}$, $\mathbf{j}_e = -e\int\mathbf{v_p}\,\delta\mathbf{f_e}\,\mathbf{d\Gamma_p}$ (the hydrodynamic expressions for the fluxes appear as these definitions under $\delta f_{e,h} = \delta f_{e,h}^{hd}$).

The GRTA model integrals share the conservation laws and zero modes of the microscopic integrals (S2): $\mathcal{C}_{e\text{-}ext}$ vanishes only in full equilibrium ($\delta f_{e,h} = 0$), $\mathcal{C}_{e\text{-}e}$ with the shifted Fermi-Dirac (S1), and $\mathcal{C}_{e\text{-}h}$ on (S1) once $\delta\mathbf{u}_e = \delta\mathbf{u}_h$. Generally, the GRTA times are free parameters subject only to $\rho_e/\tau_{e\text{-}h}^{tr} = \rho_h/\tau_{h\text{-}e}^{tr}$. However, since the first moment of the model intra-carrier collision integrals reproduce the Euler system, evaluating $\tau_{e\text{-}ext}$ and $\tau_{e\text{-}h}^{tr}$ from (S4) is necessary for consistency between the two models. Thus, under the same calculation of these scattering times hydrodynamics is recovered as the limiting case of GRTA at $\tau_{e\text{-}e} \ll \tau_{e\text{-}h}, \tau_{e\text{-}ext}$, i.e., when $\delta f_{e,h} \to \delta f_{e,h}^{hd}$.

The remaining times $\tau_{e\text{-}e}$ and $\tau_{e\text{-}h}$ appear only in the GRTA framework. We estimate them as quasiparticle lifetimes due to intra- and inter-carrier scattering, given by the imaginary part of the on-shell RPA self-energy (here we set again $\hbar = v_F = 1$) [3–5]:

$$\frac{1}{\tau_{e\text{-}e,\,e\text{-}h}(p)} = 2\sum_{\mathbf{q},\,\lambda}\left\{\left[1-f^\lambda_{\mathbf{p}-\mathbf{q}}\right][n_\omega+1] + f^\lambda_{\mathbf{p}-\mathbf{q}}\,n_\omega\right\}\operatorname{Im}\Pi_{\lambda'\lambda}(q,\omega)\,\frac{V_0^2(q)\left|u^{+1\lambda}_{\mathbf{p},\mathbf{p}-\mathbf{q}}\right|^2}{|\varepsilon(q,\omega)|^2}\Bigg|_{\omega=p-\lambda|\mathbf{p}-\mathbf{q}|}, \tag{S9}$$

where $\lambda' = \lambda = +1$ for $\tau_{e\text{-}e}$, while $\lambda' = -1$ and $\lambda = \pm 1$ for $\tau_{e\text{-}h}$ ($+1$ for conduction and -1 for valence bands), and $|u^{\lambda\lambda'}_{\mathbf{p}_1\mathbf{p}_2}|^2 = (1 + \lambda\lambda' \cos\theta_{\mathbf{p}_1\mathbf{p}_2})/2$ are the chiral overlap factors. As the GRTA times are momentum-independent parameters, the rates (S9) are evaluated at the characteristic momentum $\bar{p}v_F = \max\{k_BT \ln(1 + e^{\mu_0/k_BT}), k_BT \ln 2\}$.

III. EFFECT OF ELECTRON–HOLE BAND ASYMMETRY

Beyond graphene, the enhancement of thermopower by electron–hole scattering should apply to two-dimensional materials with generic electron–hole spectra. HgCdTe-based two-dimensional semimetals [6, 7] have nearly linear yet electron–hole-asymmetric dispersions and can be approximately described as a Dirac fluid with unequal electron and hole Fermi velocities. The electron–hole symmetry of the graphene spectrum is itself only approximate [8]. To study the role of such asymmetry, we set $v_{e,h} = v_F(1 \pm \frac{1}{2}\,\delta v/v_F)$ and take $\delta v/v_F = 0.10$ as a conservative upper bound.

The derivation of Secs. I and II nowhere relies on band symmetry: the Euler equations and the transport coefficients, established in Eqs. (4), (5), (10) of the main text, hold for arbitrary dispersions of the two species, with only the constituent functions modified. At fixed μ/k_BT, the densities, heat capacities, and hydrodynamic densities scale as $n_{e,h}, c_{e,h}, \rho_{e,h} \propto v_{e,h}^{-2}$ and the external times follow from (S7) with $v_F \to v_{e,h}$. The friction coefficient $\rho_e/\tau^{tr}_{e\text{-}h}$ is kept at its symmetric-band value, as its $\sim \delta v/v_F$ corrections only weakly renormalize the rate ratio r. Then the constraint $\rho_e/\tau^{tr}_{e\text{-}h} = \rho_h/\tau^{tr}_{h\text{-}e}$ is enforced exactly with the asymmetric densities. The asymmetry leads to the shift of the charge neutrality point $n_e = n_h$ to $\mu^* \sim (\delta v/v_F)\, k_BT$, and the doping concentration $n = n_e - n_h$ is evaluated with the asymmetric dispersions.

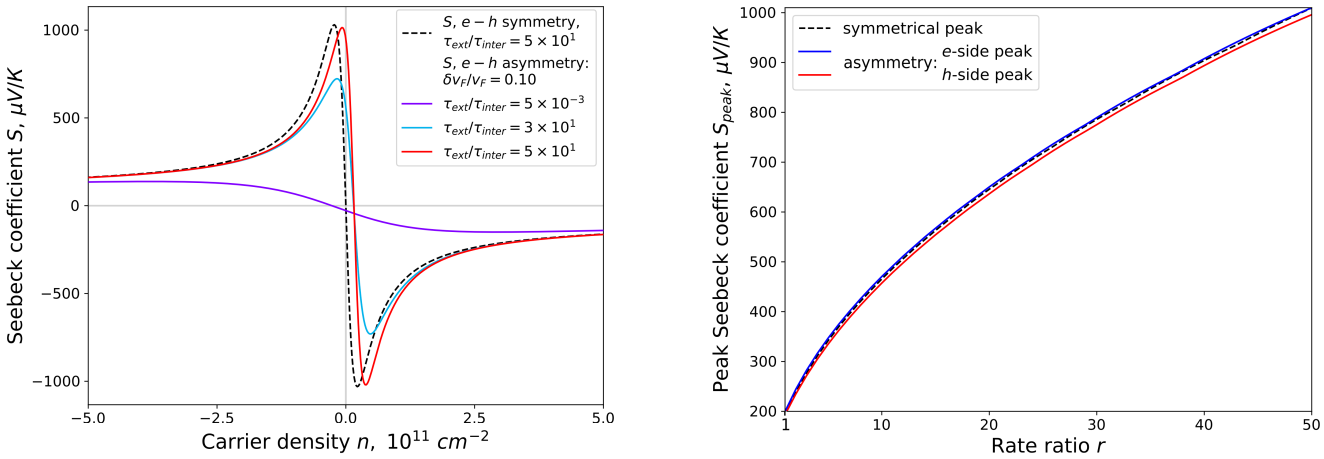

FIG. S1. Robustness of the thermopower enhancement to electron–hole band asymmetry ($\delta v_F/v_F = 0.10$, 300 K). (a) Hydrodynamic $S(n)$ for symmetric (dashed) and asymmetric bands at several rate ratios τ_{ext}/τ_{inter}, marked with different colors. (b) Electron- and hole-side peak values of S as a function of the rate ratio r for asymmetric bands, compared with the symmetric case (black dashed): the branches split by less than 2% and retain the $\sqrt{r}$ scaling.

Figure S1(a) shows the hydrodynamic $S(n)$ for symmetric and asymmetric bands. The asymmetry shifts the zero crossing of S away from charge neutrality, sets $S(n = 0)$ finite, and breaks the antisymmetry of the curve, while the electron- and hole-side peak values split by less than 2% and retain the $\sqrt{r}$ growth (Fig. S1(b)). The thermal conductivity behaves analogously. The GRTA-based transport coefficients experience the same robustness to asymmetry, as they are constituted of the same thermal averages. Ultimately, the substantial enhancement of thermopower by electron–hole scattering remains consistent for a realistic band asymmetry.

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