

Drag of electrons in graphene by substrate surface polar phonons

S. V. Koniakhin^{1,*} and A. V. Nalitov^{2,†}

¹Ioffe Physical-Technical Institute of the Russian Academy of Sciences, 194021 St. Petersburg, Russia
and St. Petersburg Academic University, Khlopina 8/3, 194021 St. Petersburg, Russia

²School of Physics and Astronomy, University of Southampton, Southampton SO17 1BJ, United Kingdom
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It is known that electron scattering by surface polar phonons (SPPs) of the substrate reduces their mobility in supported graphene. However, there is no experimental evidence for the contribution of the drag of electrons by SPPs to thermoelectric phenomena in graphene: graphene thermopower exhibits good agreement with Mott's law, which means that the diffusion contribution to the thermopower is dominant in a wide range of carrier densities and temperatures. Here we develop a complete theory of drag of electrons in graphene by SPPs. By solving the Boltzmann transport equation for electrons scattered by SPPs we derive SPP-drag contribution to the thermopower in graphene. Compared to diffusion thermopower, obtained values appear to be one order of magnitude lower for various substrates. This can be explained by the low occupation number of the SPPs and short mean free path of such phonons stemming from their small group velocity. We conclude that experiments on thermopower in graphene can be treated within the framework of Mott's law.

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I. INTRODUCTION

During the past decade there have been intense studies of the mechanical [1,2], electronic [3–5], optical [6–8], thermal [9–11], and magnetic properties of graphene. Among them, the experimental [12–16] and theoretical [17–24] investigation of thermoelectricity plays a significant role. An intriguing thermoelectric phenomenon that contributes to the thermopower in graphene, as well as in metals [25], carbon nanocomposites [26], and graphite [27–29], is the effect of phonon drag [22].

Suspending graphene sheets at a distance of hundreds of nanometers from the substrate provides a possibility to investigate the native properties of graphene. However, gating the graphene devices and controlling carrier density requires close contact between the graphene sheet and the substrate. Therefore various aspects of graphene-substrate interaction are currently under active research [30–33]. The substrate reduces the mobility of carriers in graphene due to electron scattering on surface charged impurities [5,34], surface corrugations [35–37], and atomic steps [38]. Molecular dynamics simulations show that the van der Waals interaction between the graphene [39] or nanotubes [40] and the substrate significantly reduces the relaxation time of intrinsic phonons. Reference [41] shows the importance of the interaction between graphene and the dielectric environment in the context of the Coulomb-drag effect.

In recent studies it was shown that scattering by surface polar phonons (SPPs) in substrates such as SiO₂ and SiC reduce electron mobility in graphene [42–45] and carbon nanotubes [46,47]. However, current experimental data on the thermoelectric properties of supported graphene [12–15] show that the behavior of thermopower in graphene coincides with Mott's law that describes the diffusion contribution.

Therefore it is important to elucidate why the contribution from drag by substrate phonons has not been robustly detected yet.

In this paper we consider a monolayer graphene with linear electron dispersion law $\varepsilon_{\mathbf{k}} = \hbar v_F k$, and degenerate electron gas obeying Fermi statistics. Fermi energy is related to the carrier density by $\varepsilon_F = \hbar v_F k_F = \hbar v_F \sqrt{\pi n}$. It is useful to introduce the dimensionless electron wave vector $\tilde{k} = ka_0/\pi$, where $a_0 = 2.46 \text{ \AA}$ is the graphene lattice constant. The tilde is used to denote other quantities, normalized in the same manner.

II. THEORY

Surface polar phonons of the substrate generate an electric field at significant distances from the substrate (see Fig. 1 from Ref. [46]). The phonon-induced field penetrates the graphene at the surface of the substrate and provides the probability for an electron in graphene to be scattered by remote substrate phonons. Electron scattering by SPPs is not the dominant mechanism of electron mobility reduction in relatively thick semiconductor layers in metal-oxide-semiconductor field-effect transistors, but plays a significant role for graphene and carbon nanotubes.

The electron transition rate arising from the scattering by SPPs is given by [43]

$$W_{\mathbf{k} \rightarrow \mathbf{k} + \mathbf{q}}^{SPP} = A_{\mathbf{k}, \mathbf{q}} \frac{4\pi^2 e^2 F^2}{Sq} \exp(-2qz_0), \quad (1)$$

where \mathbf{k} and \mathbf{q} are electron and phonon wave vectors, respectively, e is the electron charge, S is a surface of graphene sheet, and $z_0 \approx 3.5 \text{ \AA}$ is the van der Waals distance between the graphene sheet and the substrate. Multiplier $A_{\mathbf{k}, \mathbf{q}} = \frac{1}{2}[1 + \cos(\theta_{\mathbf{k} + \mathbf{q}} - \theta_{\mathbf{k}})]$ arises from the chiral nature of carriers in graphene. The electric field magnitude and consequently the scattering rate W overall are defined by

*kon@mail.ioffe.ru

†a.nalitov@soton.ac.uk

Fröhlich coupling

$$F^2 = \frac{\hbar\omega_{ph}}{2\pi} \left(\frac{1}{\varepsilon_\infty + \varepsilon_{env}} - \frac{1}{\varepsilon_0 + \varepsilon_{env}} \right), \quad (2)$$

where ε_0 and ε_∞ are low- and high-frequency dielectric constants of the substrate, respectively, and ε_{env} is the environment dielectric constant. Following Ref. [43] we assume the latter to be equal to 1. As can be seen from the exponential multiplier

$$\begin{aligned} \left(\frac{\partial}{\partial t} f(k) \right)_{ph} &= -\frac{2\pi}{\hbar} \frac{S}{4\pi^2} \int d\mathbf{q} \{ W_{k \rightarrow k+q} N_{ph}(q) f^{(0)}(\varepsilon_k) [1 - f^{(0)}(\varepsilon_{k+q})] \delta(\varepsilon_k - \varepsilon_{k+q} - \hbar\omega_{ph}) \\ &\quad + W_{k \rightarrow k-q} f^{(0)}[N_{ph}(q) + 1] f^{(0)}(\varepsilon_k) [1 - f^{(0)}(\varepsilon_{k-q})] \delta(\varepsilon_k - \varepsilon_{k-q} + \hbar\omega_{ph}) - W_{k-q \rightarrow k} N_{ph}(q) f^{(0)}(\varepsilon_{k-q}) \\ &\quad \times [1 - f^{(0)}(\varepsilon_k)] \delta(\varepsilon_k - \varepsilon_{k-q} + \hbar\omega_{ph}) - W_{k+q \rightarrow k} [N_{ph}(q) + 1] f^{(0)}(\varepsilon_{k+q}) [1 - f^{(0)}(\varepsilon_k)] \delta(\varepsilon_k - \varepsilon_{k+q} - \hbar\omega_{ph}) \}, \end{aligned} \quad (3)$$

where $f^{(0)}(\varepsilon_k)$ is the equilibrium Fermi distribution function.

In the presence of a temperature gradient phonon distribution function $N_{ph}(q)$ can be written as

$$N_{ph}(\mathbf{q}) = N_{ph}^{(0)}(q) + N_{ph}^{(1)}(\mathbf{q}). \quad (4)$$

In the equation above, $N_{ph}^{(0)}(q)$ is the Bose equilibrium distribution function for phonons and $N_{ph}^{(1)}(\mathbf{q})$ is a small correction due to the temperature gradient, which can be written in the relaxation time approximation as

$$N_{ph}^{(1)}(\mathbf{q}) = \tau_{ph}(q) \mathbf{v}(\mathbf{q}) \vec{\nabla} T \frac{\partial N_{ph}^{(0)}(q)}{\partial T}, \quad (5)$$

where $\mathbf{v}(\mathbf{q}) = \nabla_{\mathbf{q}} \omega_{ph}$ is a group velocity of SPPs. The dispersion law of such phonons can be written as

$$\omega_{ph}(q) = \omega_0(1 - \beta \tilde{q}^2), \quad (6)$$

where $\tilde{q} = qa_0/\pi$ is the phonon wave vector normalized to the graphene lattice constant. Approximating optical phonon dispersion curves of many semiconductors and dielectrics allows one to estimate $\beta \sim 0.1$.

We assume that the SPP lifetime τ_{ph} is independent on the phonon wave vector q . Multiple studies devoted to molecular dynamics simulations [49–52] and Raman experiments [53–58] indicate an optical phonon lifetime of about dozens of picoseconds in various solids, and we set $\tau_{ph} = 10$ ps. The SPP group velocity is linear in q , therefore $\tau_{ph} v(q) = L_0 \tilde{q}$, where $L_0 = 2\tau_{ph}\omega_{ph}\beta \frac{a_0}{\pi}$. Taking $\tau_{ph} = 10$ ps, $\hbar\omega_{ph} = 0.1$ eV, and $\beta = 0.1$ yields $L_0 = 25$ nm.

Electrons in graphene are scattered by phonons with q of the order of $k_F \ll \pi/a_0$ and for actual values of Fermi energy $\hbar\omega_{ph}\beta \tilde{k}_F^2 \ll k_B T$. Thus the occupation number of phonons does not depend significantly on the magnitude of the phonon wave vector q and the quantity

$$\frac{\partial N_{ph}^{(0)}(q)}{\partial T} = k_B \frac{\hbar\omega_{ph} e^{\hbar\omega_{ph}/k_B T}}{(e^{\hbar\omega_{ph}/k_B T} - 1)^2 (k_B T)^2} \quad (7)$$

is considered below to be constant.

When $N_{ph}^{(0)}$ is substituted into the right-hand part of (3), the latter vanishes due to energy conservation law entering

in (1), q^{-1} is a characteristic distance at which the electric field decays outside the substrate. In Table II of Ref. [43] the energies of surface polar phonons and values of Fröhlich coupling strength for various substrate materials are listed. For a variety of substrates $\hbar\omega_{ph} \sim 0.1$ eV and the F^2 value is close to 0.5 meV.

The SPP collision integral, entering the Boltzmann transport equation on the electron distribution function $f(\varepsilon_{\mathbf{k}})$, can be written as [48]

the delta functions. After replacing \mathbf{q} with $-\mathbf{q}$ in the second and the third terms in the curly braces in (3), substituting first the correction for phonon distribution (5), one obtains the following correction for the electron distribution function within the framework of relaxation time approximation:

$$f^{(1)}(\mathbf{k}) = -\frac{2\pi\tau(k)}{v_F} \frac{e^2 F^2}{\hbar^2} L_0 \frac{\partial N_{ph}^{(0)}(q)}{\partial T} \left(\frac{\mathbf{k}}{k} \vec{\nabla} T \right) I_1(\tilde{k}), \quad (8)$$

where $\tau(k)$ is the electron transport time, governed by different momentum relaxation processes, such as scattering on Coulomb and point defects, as well as intrinsic acoustic and optical phonons and SPPs of the substrate. $I_1(\tilde{k})$ in the expression above is the dimensionless integral over phonon wave vector:

$$\begin{aligned} I_1(\tilde{k}) &= \int_0^1 d\tilde{q} d\theta \tilde{q} A_{\mathbf{k},\mathbf{q}} \exp^{-2z_0 \tilde{q}(\pi/a_0)} \cos(\theta) \\ &\quad \times [f^{(0)}(\tilde{k}) - f^{(0)}(|\tilde{\mathbf{k}} + \tilde{\mathbf{q}}|)] \\ &\quad \times [\delta(\tilde{k} + \tilde{\omega}_{ph} - |\tilde{\mathbf{k}} + \tilde{\mathbf{q}}|) - \delta(\tilde{k} - \tilde{\omega}_{ph} - |\tilde{\mathbf{k}} + \tilde{\mathbf{q}}|)], \end{aligned} \quad (9)$$

where $\tilde{\omega}_{ph} = \frac{\omega_{ph} a_0}{v_F \pi}$, $\tilde{\mathbf{k}} = \frac{\mathbf{k} a_0}{\pi}$, and $f^{(0)}(\tilde{k}) = \{1 + \exp[(\pi \hbar v_F \tilde{k}/a_0) - \varepsilon_F]/k_B T\}^{-1}$. Figure 1(a) shows a typical profile of the integral $I_1(\tilde{k})$ as a function of the electron wave vector amplitude for various Fermi energies.

In the case of nearly elastic scattering of electrons by acoustic phonons the expression in the first set of square brackets in (9) yields $\pm \frac{\partial f^{(0)}(\varepsilon_k)}{\partial \varepsilon} \hbar\omega_{ph}(q) \approx \pm \hat{\delta}(\varepsilon_k - \varepsilon_F) \hbar\omega_{ph}(q)$, where $\hat{\delta}$ is a delta function with a broadening of the order of temperature.

In contrast with the case of acoustic phonons, here $f^{(1)}$ is not linear in $\frac{\partial f^{(0)}}{\partial \varepsilon}$. However, $f^{(1)}$ differs from zero for wave vectors k close to the elastic circle of the radius k_F [see Fig. 1(a)]. Therefore, in (8) we can assume $\tau(k) = \tau(k_F) = \text{const}$ without significant loss of accuracy. Electron conductivity in graphene is directly related to the electron momentum relaxation time

as [4,43]

$$\sigma = \frac{e^2}{\hbar} \frac{v_F k_F \tau(k_F)}{\pi}. \quad (10)$$

The SPP driven current is given by integral

$$j_{ph} = \frac{\pi^2 e v_F}{a_0^2} \int d\tilde{\mathbf{k}} f^{(1)}(\tilde{\mathbf{k}}). \quad (11)$$

Substituting the electron distribution correction (8) and dividing the obtained expression by conductivity (10) yields the SPP-drag thermopower:

$$S_{SPP} = \frac{\pi^4 k_B}{e} \frac{F^2 e^2}{\varepsilon_F a_0 k_B T} \frac{L_0}{a_0} \frac{\hbar \omega_{ph}}{k_B T} \frac{e^{\hbar \omega_{ph}/k_B T}}{(e^{\hbar \omega_{ph}/k_B T} - 1)^2} I_2, \quad (12)$$

where

$$I_2 = \frac{1}{2} \int_0^1 \tilde{k} d\tilde{k} I_1(\tilde{k}). \quad (13)$$

As usual, the expression for the thermopower has a form of $\frac{k_B}{e} = 86 \mu\text{V K}^{-1}$ times a dimensionless factor, which depends on ε_F , T , etc.

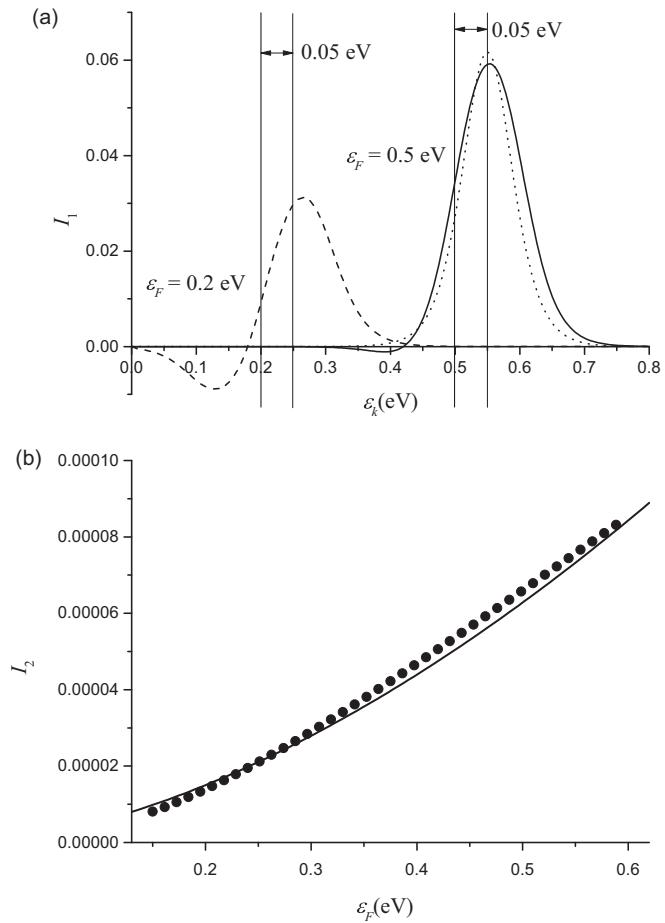


FIG. 1. (a) Dependence of $I_1(\tilde{k})$ magnitude on $\varepsilon_k = \hbar v_F \pi \tilde{k} / a_0$. SPP energy is 0.1 eV. The solid curve is for $\varepsilon_F = 0.5$ eV and the dashed curve is for $\varepsilon_F = 0.2$ eV. Interestingly, notice that for large Fermi energies $I_1(k) \approx \frac{\partial}{\partial \varepsilon} f^{(0)}(\varepsilon - \frac{1}{2} \hbar \omega_{ph})$, denoted by the dotted curve. (b) I_2 as a function of Fermi energy. $T = 300$ K, $\hbar \omega_{ph} = 0.1$ eV. Circles denote the results of numerical integration and the black curve shows approximation with (14).

In Fig. 1(b) we have shown the dependence of I_2 on the Fermi energy in the graphene sample. Due to the fact that $I_1(k) \approx \frac{\partial}{\partial \varepsilon} f^{(0)}(\varepsilon - \frac{1}{2} \hbar \omega_{ph})$, for actual values of the Fermi energy, temperature and phonon frequency I_2 were approximated via the formula

$$I_2 \simeq A_0 \left(\frac{\varepsilon_F + \frac{1}{2} \hbar \omega_{ph}}{\varepsilon_0} \right) \left(\frac{\varepsilon_F}{\varepsilon_0} \right)^{0.7} \left(\frac{\hbar \omega_{ph}}{\varepsilon_0} \right)^{0.7}, \quad (14)$$

where $A_0 = 5 \times 10^{-6}$ and $\varepsilon_0 = 0.1$ eV, and sufficient accuracy was reached.

III. RESULTS AND DISCUSSION

Table I lists the values of the SPP-drag thermopower for graphene on various substrates. The values $\varepsilon_F = 0.5$ eV and $T = 300$ K were used. Values of phonon energies and magnitudes of Fröhlich coupling were adopted from [43].

For $\varepsilon_F = 0.5$ eV, $T = 300$ K, and phonon lifetime $\tau_{ph} = 10$ ps one has $S_{SPP} \approx 2 \mu\text{V K}^{-1}$ for graphene on the SiO₂ substrate, which is one order of magnitude lower than the diffusion contribution. One sees that the S_{SPP} is of the same order for other considered substrates.

The substitution of (14) into (12) allows us to conclude that the SPP thermopower moderately grows with Fermi energy in graphene. Due to the exponential growth with temperature of the SPP occupation number, the SPP-drag mechanism is only significant at relatively high temperatures. Figure 2 shows S_{SPP} as a function of the Fermi energy and the temperature. By different dependencies on temperature and Fermi energy the SPP contribution to the thermopower can be straightforwardly distinguished from the contributions of diffusion and the intrinsic phonon-drag mechanism.

To suggest the substrate with maximal SPP-drag effect it is important to analyze how S_{SPP} depends on the SPP energy. Equation (2) shows that Fröhlich coupling linearly grows with phonon energy and by averaging over dielectric constants of various materials one can write the following approximate relation: $F^2 = 0.005 \hbar \omega_{ph}$. Substituting this to (12) allows one to plot S_{SPP} as a function of the energy of the substrate phonon. Figure 3 shows that the phonon energy most favorable for increasing the SPP-drag thermopower is about 0.1 eV. The energies of SPPs in considered substrates are close to this value. This means that the maximal achievable value of S_{SPP} is reached for considered substrates.

It is interesting to compare coupling between electrons and intrinsic acoustic phonons with that for SPPs. The coupling of electrons with intrinsic acoustic phonons in graphene reads as [22,45]

$$W_{\mathbf{k} \rightarrow \mathbf{k} + \mathbf{q}}^{intrinsic} = A_{\mathbf{k}, \mathbf{q}} \frac{\hbar D^2 q}{\rho S v_s}, \quad (15)$$

TABLE I. SPP-drag thermopower for graphene on SiO₂, HfO₂, SiC, and h-BN. $\varepsilon_F = 0.5$ eV and $T = 300$ K. Values are given in $\mu\text{V K}^{-1}$.

SiO ₂	HfO ₂	SiC	h-BN
1.7	1.1	1.1	0.6

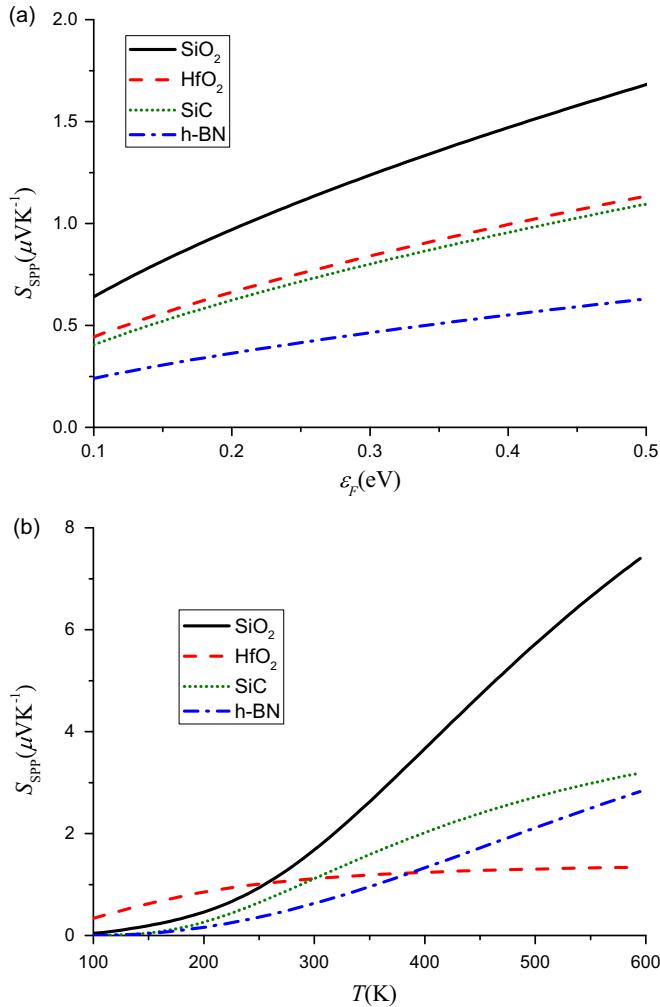


FIG. 2. SPP-drag thermopower in graphene on various substrates. Constant phonon lifetime τ_{ph} is assumed to be 10 ps. (a) Dependence of S_{SPP} on Fermi energy in graphene, $T = 300$ K. (b) Dependence of S_{SPP} on temperature for constant phonon lifetime, $\varepsilon_F = 0.5$ eV.

where v_s is the sound velocity in graphene, $D = 16$ eV is the deformation potential, and $\rho = 6.5 \times 10^{-8} \text{ g cm}^{-2}$ is the mass density of graphene. This expression is similar to the one for SPPs (1).

For the phonon wave vector q of the order of Fermi wave vector k_F corresponding to $\varepsilon_F = 0.1$ eV one can estimate $W^{\text{SPP}}/W^{\text{intrinsic}} \approx 600$ and for $\varepsilon_F = 0.5$ eV one obtains $W^{\text{SPP}}/W^{\text{intrinsic}} \approx 15$. This means that the electrostatic interaction of electrons with remote substrate phonons can be stronger than coupling with intrinsic phonons via deformation potential.

Nevertheless, the strong coupling between electrons in graphene and SPPs is not enough to overcome exponentially the low occupation number of SPPs and short mean free

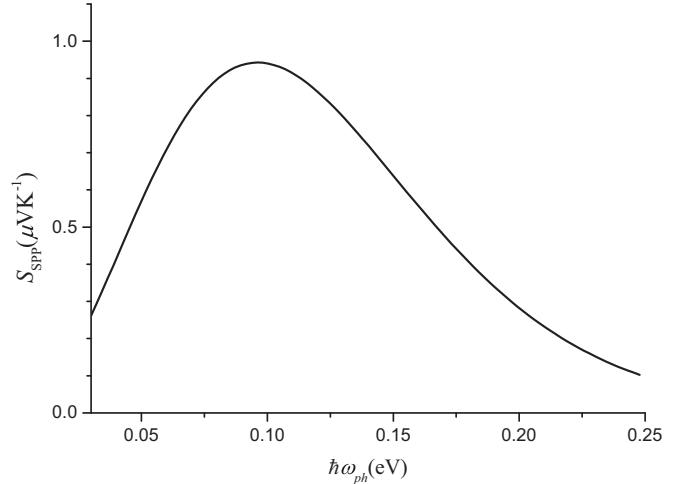


FIG. 3. SPP-drag thermopower as a function of phonon energy. $T = 300$ K, $\varepsilon_F = 0.5$ eV. Phonon lifetime $\tau_{ph} = 10$ ps.

path stemming from the low group velocity of phonons with $q \approx k_F$. We conclude that the contribution to thermopower from the drag of electrons by SPPs is weaker than the diffusion contribution, which explains the current experimental data on the thermopower in graphene [12–15], being in agreement with Mott's law.

The effect of phonon drag will potentially play a role for a substrate with a high SPP lifetime. Limitation of the phonon lifetime due to three-phonon processes will not allow dominance of SPP-drag contribution in graphene thermopower with high probability. Moreover, to obtain the predicted values of thermopower the interface between the graphene sheet and the substrate is to be thin and smooth. In the case of mechanically exfoliated graphene, the distance between the substrate and graphene sheet can be expected to be higher than the here adopted value. For epitaxial graphene, a dead buffer layer can also negatively affect the graphene properties [59].

The considered effect of SPP drag can be expected even for nonpolar substrates like diamond due to the polarizability of interatomic bonds [60], which is important for creating composite nanocarbon-based thermoelectric devices. However, one should expect that for nonpolar substrates the SPP-drag thermopower will be smaller than for the polar substrates considered in this paper.

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