Friction force on slow charges moving over supported graphene

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Abstract
We provide a theoretical model that describes the dielectric coupling of a two-dimensional (2D) layer of graphene, represented by a polarization function in the random phase approximation, and a semi-infinite three-dimensional (3D) substrate, represented by a surface response function in a non-local formulation. We concentrate on the role of the dynamic response of the substrate for low-frequency excitations of the combined graphene–substrate system, which give rise to the stopping force on slowly moving charges above doped graphene. A comparison of the dielectric loss function with experimental high-resolution electron energy loss spectroscopy (HREELS) data for graphene on a SiC substrate is used to estimate the effects of damping rate and the local field correction in graphene, as well as to reveal the importance of phonon excitations in an insulating substrate. While the local field correction and linearly dispersing damping rate did not yield any important effects compared to the constant damping rate in graphene, a strong signature of the hybridization between graphene’s $\pi$ plasmon and the substrate’s phonon is found in both the HREELS spectra and the stopping force. A friction coefficient that is calculated for slow charges moving above graphene on a metallic substrate shows an interplay between the low-energy single-particle excitations in both systems.

1. Introduction

After the initial wave of intense studies of the electronic and transport properties of graphene [1, 2], the focus of research seems to be turning to graphene’s interactions with a dielectric environment [2, 3]. While it was recognized early on that the presence of charged impurities in the material surrounding graphene is a likely cause of its peculiar minimum conductivity [4], much attention has also been paid to the dielectric screening of these impurities by either their host material or various high-kappa dielectrics [5]. Besides an obvious interest in SiO$_2$ as the most common substrate for graphene that is produced by the exfoliating technique [6, 7], interactions of epitaxially grown graphene on a SiC substrate have also attracted considerable attention [8, 9]. Most recently, several reports have appeared studying the properties of graphene on metallic substrates [10, 11], as well as graphene’s interaction with organic solvents and ionic solutions [12, 13].

In view of these developments and possible extensions of the recent electron energy loss spectroscopy (EELS) studies of free-standing graphene structures [14, 15] that would include the presence of a substrate, it is of interest to study the interaction of graphene with externally moving charged particles in a dielectric environment. Although it was recently shown that a dielectric environment can exert strong effects on the plasmon excitations of carbon nanotubes due to coupling of their $\sigma + \pi$ and $\pi$ plasmons with high-frequency modes in the nearby material [16], in this paper we limit ourselves to the effects of a substrate on slow moving external charges such that only the low-energy excitations of graphene’s $\pi$ electron bands occur. From an applied point of view, this problem is highly relevant to a recent experiment that used a high-resolution EELS (HREELS) technique to probe the momentum-resolved low-frequency plasmon excitations of doped graphene grown on a SiC substrate [17]. Moreover, interactions with slow heavy particles are relevant to studies of intercalation of alkali-metal atoms [18], friction forces on migrating atoms and molecules moving near graphene [19, 20], and monitoring of ion flow in an aqueous solution adjacent to carbon nanostructures [21].

From a theoretical point of view, the interaction of graphene with slow external charges can be described by the recently developed dielectric function in the random phase approximation (RPA), in which graphene’s $\pi$ electron bands are treated in the approximation of linearized electron energy dispersion, giving rise to the picture of massless Dirac
fermions (MDF) in two dimensions [22, 23]. This theoretical model captures all of the physical processes relevant to such interactions, including graphene’s two-dimensional (2D) \( \pi \) plasmon mode and both the intra-band and inter-band single-particle excitations (SPEs). Furthermore, the MDF-RPA dielectric function for graphene takes into account the strong dependence of these excitation modes on the level of doping in graphene, which often results from charge transfers between graphene and the substrate [6, 17, 11] or may be induced by a gate potential. However, all RPA dielectric functions suffer two major shortcomings: they neglect the local field effects (LFE) due to electron correlations, and they have an inherent assumption of an infinite lifetime of electron excitations. Only one of these shortcomings can be qualitatively corrected in the MDF-RPA dielectric function for graphene at a time, e.g. by using either the Hubbard approximation (HA) for the LFE [2] or by introducing a finite decay rate, \( \gamma \), using Mermin’s procedure [24, 25].

With respect to graphene’s coupling with a substrate, we note that few theoretical studies have gone beyond treating the substrate in a static mode and most assume a vanishing graphene–substrate gap, with the exception of recent studies of the remote scattering of charge carriers in graphene on surface phonon modes of a strongly polar substrate [26–28]. Therefore, in this work we first provide a detailed description of graphene’s coupling with a substrate by combining the LFE or Mermin-corrected MDF-RPA dielectric function for graphene with the surface response function for a substrate, which is described by a bulk dielectric function that is allowed to depend on both the frequency and the three-dimensional (3D) wavenumber. We note that the dependence of the substrate’s dielectric function on the wavenumber is often neglected, giving rise to the local approximation for its surface response function, which is appropriate for describing cases such as the non-dispersing Fuchs–Klieser surface phonons in strongly polar substrates [27, 28]. However, when the substrate’s bulk dielectric function depends strongly on the 3D wavenumber, such as in the case of a metal, one has to resort to the so-called specular reflection model (SRM) [29] in order to obtain a surface response function that takes into account the spatial dispersion of the substrate’s excitation modes, and hence gives rise to a non-local description of the substrate.

Such a formulation of the graphene–substrate interaction allows for the possibility of studying the hybridization of collective modes in the two systems [16], which is illustrated here for the example of plasmon modes in graphene and surface phonon modes in a SiC substrate corresponding to the recent HREELS experiment [17]. Moreover, the graphene–substrate dielectric coupling is expected to affect the energy losses of a slow external charge moving parallel to graphene. For example, it has been experimentally confirmed that slow ions can lose their kinetic energy during grazing scattering from an insulating surface due to excitations of optical phonons in the substrate [30], and it would therefore be interesting to explore how such a process would change in the presence of graphene and the possibility of simultaneous excitations of graphene’s \( \pi \) plasmon and its intra-band SPEs. Moreover, techniques such as HREELS can be used to probe the hybridization of the substrate’s phonon modes with graphene’s \( \pi \) plasmon under controlled doping [26, 28], and further explore the role of the nearby SPE continuum as a region in which such hybridized modes can undergo Landau damping. Metallic substrates, on the other hand, have attracted attention only recently as a possibly interesting dielectric environment for graphene [10, 11]. It has been recognized for many years that metal surfaces can exert strong dissipative and conservative (image) forces on moving external charges, especially in the grazing scattering geometry [31]. In particular, low-energy SPEs in a metal have been shown to give rise to strong friction on particles moving slowly over metal surfaces [20, 32, 33]. It is therefore tempting to explore how this concept of friction would be affected by the presence of graphene on a metal substrate, resulting in an overlap of the metal’s SPEs and graphene’s intra-band SPEs. Similar effects have been explored using a non-local description for a Cu overlayer on an Si substrate [34].

Consequently, we believe that studying both the loss function of the graphene–substrate system and the stopping force on an external moving charge can be used to explore the effects of the substrate’s dynamic response. There are many model parameters that may be relevant to the description of such processes. In our previous work [35], we have concentrated on amending the MDF-RPA dielectric function for graphene with a constant damping rate, \( \gamma \), through Mermin’s procedure, as well as on analyzing the effects of the equilibrium charge carrier density in graphene, \( n \), and the graphene–substrate gap, \( h \), for a substrate treated in the static mode by simply assigning a dielectric constant to it. In this work, we include two further refinements to the MDF-RPA dielectric function for graphene by considering a dispersing damping rate and the LFE correction, and we refine our description of the substrate by including its dynamic and non-local properties within the SRM, which gives rise to an effective surface dielectric function for the substrate that depends on both frequency and wavenumber.

With regards to specific applications, we first test these model improvements by comparing the loss function of the graphene–substrate system with the recent HREELS experimental spectra on epitaxial graphene [17] and find that the inclusion of either the linearly dispersing damping or the LFE correction gives no improvements over a similar comparison in our previous work [35]. However, we find that the inclusion of a transverse optical (TO) phonon in the SiC substrate both improves the agreement at long wavelengths and reveals a strong hybridization between graphene’s plasmon and the substrate’s phonon. While such hybridization could not be found in our previous work since the substrate was treated in the static mode [35], the finding of hybridization in this work enables us to predict an avoided crossing between the plasmon and phonon dispersion curves that seems to have been corroborated in a recent unpublished experiment by Liu and Willis [36].

Our second application of this improved model is concerned with the stopping force on slow charges moving over graphene when the substrate dynamics are described non-locally. While we find that the effects of both linearly
dispersing damping and the LFC corrections to graphene’s dielectric function are small compared to the effects of constant damping $\gamma$ and the doping density of graphene $n$, we find that the presence of the TO phonon in a SiC substrate and the low-energy SPEs in an Al substrate exert large effects on the stopping force. We note that while optical phonons in a polar substrate are only weakly dispersive, making the local description of such a substrate well founded, it is absolutely necessary to use a non-local approach for describing the low-energy SPEs in a metallic substrate. As far as we are aware, a non-local treatment of a substrate has not been applied before in the context of graphene–substrate interactions. In all our applications, we reduce the parameter space of our model by considering only two equilibrium charge densities: $n = 0$, characterizing intrinsic or undoped graphene, and $n = n_0$, where $n_0 = 10^{13}$ cm$^{-2}$, as a value very close to graphene’s charge carrier density for both the SiC [17] and Al [11] substrates. We also take $h = 1$ Å as a reasonable estimate for the graphene–substrate gap [11, 35].

After outlining the basic theory in the following section, we present results comparing our model with the HREELS experiment of Liu et al [17]. We then consider the effects of various model parameters on the stopping force for free graphene, followed by examples of the stopping force for graphene on an SiC substrate and the friction coefficient for graphene on an Al substrate. A brief discussion of these results is provided in the concluding section. We use Gaussian electrostatic units throughout this paper unless otherwise stated.

2. Theory

We use a Cartesian coordinate system with coordinates $(r, z)$, where $r = (x, y)$ and assume that graphene is placed in the $z = 0$ plane. A semi-infinite substrate with a bulk dielectric function $\epsilon_{\text{sub}}(k)$ is assumed to occupy the region $z \leq -h$ underneath graphene, while the region $z > -h$ is assumed to be a vacuum or air [37]. The 3D wavevector describing the excitation modes in the substrate can be decomposed as $k = (q, k_z)$, where $q = (k_x, k_y)$ are the components parallel to the substrate surface and $k_z$ is the component perpendicular to the substrate surface. We further assume that an external charge with density $\rho_{\text{ext}}(r, z, t)$ moves along a classical trajectory localized to the region $z > 0$ above graphene. Following [37], we can express the induced potential $\Phi_{\text{ind}}(r, z, t)$ in the region above graphene by using the 2D Fourier transform with respect to the surface components $(r \rightarrow q)$ and with respect to time $(t \rightarrow \omega)$ as

$$\Phi_{\text{ind}}(q, z, \omega) = \left[ \frac{1}{\epsilon(q, \omega)} - 1 \right] \frac{2\pi}{q} S(q, \omega) e^{-qz},$$

(1)

where $\epsilon(q, \omega)$ is the dielectric function of the combined graphene–substrate system and

$$S(q, \omega) = \int_{-\infty}^{\infty} \mathrm{d}t \, e^{i\omega t} \int d^2r \, e^{-iqr} \int_0^{\infty} dz \, e^{-qz} \rho_{\text{ext}}(r, z, t),$$

(2)

is the structure factor of the external charge distribution. The dielectric function of the system can be written as

$$\epsilon(q, \omega) = \epsilon_{bg}(q, \omega) + V_C(q) \Pi(q, \omega),$$

(3)

where $\epsilon_{bg}(q, \omega)$ is the effective background dielectric function of the substrate, $V_C(q) = 2\pi e^2 / q$ is the Fourier transform of the 2D Coulomb interaction, and $\Pi(q, \omega)$ is the polarization function for free graphene.

Assuming that the substrate is isotropic so that its dielectric function only depends on the magnitude of the wavevector, $k = \sqrt{q^2 + k_z^2}$, one can use the SRM to handle the electrostatic boundary condition on the substrate’s surface [29]. Combining the steps of the derivation outlined in [38] with those in [37], we obtain

$$\epsilon_{bg}(q, \omega) = \left[ 1 - \frac{\epsilon_s(q, \omega) - 1}{\epsilon_s(q, \omega) + 1} e^{-2qh} \right]^{-1},$$

(4)

where $\epsilon_s(q, \omega)$ is the effective surface dielectric function of the substrate, defined by

$$\epsilon_s(k, \omega) = \left[ q \pi \int_{-\infty}^{\infty} \frac{dk_z}{k^2 \epsilon_{\text{sub}}(k, \omega)} \right]^{-1}.$$  

(5)

The expression $(\epsilon_s - 1) / (\epsilon_s + 1)$ in equation (4) represents a non-local surface response function for a substrate that exhibits strong dispersion, but in cases where the dispersion in the substrate can be neglected so that $\epsilon_{\text{sub}}(k, \omega) \approx \epsilon_{\text{sub}}(\omega)$, one recovers the local approximation $\epsilon_s(q, \omega) \approx \epsilon_{\text{sub}}(\omega)$.

As an example of a non-local surface dielectric function, one may consider the low-frequency expansion of Lindhard’s dielectric function [20, 32, 33],

$$\epsilon_{\text{sub}}(k, \omega) \approx 1 + \frac{K_{TF}^2}{k^2} + \frac{\pi \omega}{k V_F} H(2K_F - k),$$

(6)

where $K_{TF} = \sqrt{2}\Omega_p / V_F$ is the Thomas–Fermi (TF) inverse screening length of a 3D electron gas having a volume density $N$, $\Omega_p = \sqrt{4\pi\epsilon^2 e^2 N / m_e}$ is its bulk plasma frequency, $K_F = m_e V_F / (\pi \sqrt{2} N)^{1/3}$ is its Fermi wavenumber, and $m_e$ is the mass of an electron. This dielectric function describes the low-energy SPEs of a metal subject to the cut-off $k < 2K_F$, as implied by the Heaviside function $H(2K_F - k)$ in equation (6). We note that equation (6) has often been used in equation (5) to model various surface processes on metals [20, 32, 33].

On the other hand, the local approximation may be suitable for cases such as a strongly polar insulating substrate characterized by non-dispersing TO phonon modes with frequencies $\omega_{\text{TOi}}$ and damping rates $\gamma_{\text{TOi}}$, in which case one may use a dielectric function of the form [26–28]

$$\epsilon_{\text{sub}}(\omega) = \epsilon_\infty + \sum_i \frac{f_i \omega_{\text{TOi}}}{\omega_{\text{TOi}} + \gamma_{\text{TOi}}},$$

(7)

where $\epsilon_\infty = \lim_{\omega \to \infty} \epsilon_{\text{sub}}(\omega)$ is the high-frequency dielectric constant of the substrate and the oscillator strengths, $f_i$, satisfy the relation $\sum_i f_i = \epsilon_0 - \epsilon_\infty$, where $\epsilon_0 = \epsilon_{\text{sub}}(0)$ is the static dielectric constant of the substrate [26, 27].
We note that in the literature on graphene it is common to assume a zero gap \((\hbar = 0)\) and to describe the substrate by only the static dielectric constant \([2, 22, 23]\), for which equation (4) gives an effective background dielectric constant \(\varepsilon_b(q, \omega) \rightarrow \varepsilon_{\text{eff}} = (\varepsilon_0 + 1)/2\). In this case, a simple description of the screening of electron–electron interactions in graphene can be quantified by the Wigner–Seitz radius, \(r_s = e^2/(\varepsilon_{\text{eff}}\hbar v_F)\) \([23]\), where \(v_F \approx c/300\) is the Fermi speed of graphene and \(c\) is the speed of light in vacuum. We note that the limiting case of free graphene \((\varepsilon_b(q, \omega) = 1)\) can be obtained by letting \(h \rightarrow \infty\) in equation (4) or by setting \(\varepsilon_0 = 1\) in the zero gap case with a static substrate, but the focus of this work is on the effects of substrate including a finite gap, non-local effects, and its dynamic response.

For the dielectric response of graphene in equation (3), we use the polarization function for graphene’s \(\pi\) electron excitations in the MDF-RPA \([22, 23]\), \(\Pi(q, \omega)\), as well as the corrections to the polarization function for the LFE and the finite lifetime of the excitation modes of charge carriers in graphene. The LFE are described in the Hubbard approximation (HA) by replacing the MDF-RPA polarization by \((1 - G(q))\Pi(q, \omega)\), where \(G(q) = (q/g_0)/\sqrt{q^2 + k_F^2}\) and \(g_0 = 4\) is the degeneracy factor of graphene’s \(\pi\) electrons \([2]\). The finite lifetime of the excitation modes of charge carriers in graphene is treated by introducing a finite damping rate, \(\gamma\), in the MDF-RPA polarization function through Mermin’s procedure \([24, 25]\), whereby one replaces \(\Pi(q, \omega)\) with

\[
\Pi_M(q, \omega, \gamma) = \frac{\Pi(q, \omega + i\gamma)}{1 - i\frac{\omega}{\omega + i\gamma}}\frac{1 - \Pi_0(q, \omega + i\gamma)}{\Pi_0(q, \omega)}, \tag{8}
\]

where the static limit of the RPA polarization, \(\Pi_0(q) = \lim_{\omega \rightarrow 0} \Pi(q, \omega)\), is given elsewhere \([22, 23]\).

In the case of a point charge \(Ze\) moving parallel to graphene with velocity \(v\) and at a fixed distance \(z_0 > 0\), we obtain \(\Sigma(q, \omega) = 2\pi Ze\delta(\omega - q \cdot v)\) from equation (2). Using this expression in the inverse Fourier transform of equation (1), we can evaluate the stopping force on the point charge \(Ze\) from the definition

\[
F_s = -Ze\nabla \cdot \Phi_{\text{ind}}^\omega(\mathbf{r}, z, t)|_{\mathbf{r} = \mathbf{r}, z = z_0}, \tag{9}
\]

where \(\nabla \cdot \Phi_{\text{ind}}\) giving \([37]\)

\[
F_s = \frac{2}{\pi} \frac{Z^2e^2}{v} \int_0^\infty dq \, q^2e^{-2qz_0} \times \int_0^{qz_0} d\omega \frac{\omega}{\sqrt{q^2v^2 - \omega^2}} \text{Im}\left[\frac{1}{\varepsilon(q, \omega)}\right]. \tag{10}
\]

We note that \(\text{Im}[1/\varepsilon(q, \omega)]\) represents the loss function of the system that, apart from a kinematic prefactor describing the scattering event, is directly accessible in the HREELS experiment \([17, 39]\).

Finally, for cases in which the loss function in equation (10) can be expanded to the first order in frequency as \(\text{Im}[1/\varepsilon(q, \omega)] \approx \omega F(q)\), one can define a friction coefficient \(\eta\) for a sufficiently slow particle moving parallel to graphene through the equation \(F_s \approx -\eta v\). In such cases, the friction coefficient is given by

\[
\eta = \frac{1}{2} Z^2e^2 \int_0^\infty dq \, q^2e^{-2qz_0} F(q). \tag{11}
\]

For example, this procedure has been performed using equations (5) and (6) to obtain the friction coefficient for slow particles on metal surfaces \([20, 32]\). A similar approach can also be used for graphene in the limit of vanishing damping rate, giving

\[
\Pi(q, \omega) \approx \Pi_0(q) + \frac{i\omega}{\pi \hbar v_F^2} H(2k_F - q)\sqrt{\left(\frac{2k_F}{q}\right)^2 - 1}. \tag{12}
\]

where \(k_F = \sqrt{\pi n}\) is graphene’s Fermi wavenumber. A different but analogous expression can be obtained from equation (8) in the case of a finite damping rate in graphene \([35]\).

3. Results

3.1. Comparison with HREELS experiment

It has been known for some time that the damping rate \(\gamma\) of elementary excitations in a many-electron system can strongly influence the friction forces on slow particles \([20, 32]\). However, the exact value of the damping rate is generally difficult to measure, and in the case of graphene is presently unknown. Nevertheless, \(\gamma\) is often used as an empirical parameter in the RPA response function modified by Mermin’s procedure \([24, 25, 35]\). Therefore, to obtain a reasonable estimate for the value of \(\gamma\) for graphene, we compare the experimental data of Liu \textit{et al} \([17]\) for the HREELS spectra of doped graphene on an SiC substrate with the loss function \(\text{Im}[-1/\varepsilon(q, \omega)]\) obtained from equation (3) by using the Mermin polarization function for graphene, which is given in equation (8). In addition to constant values of \(\gamma\), which were explored in our previous work \([35]\), we explore a damping rate with a linear dispersion of the form \(\gamma = v_\gamma q\), where \(v_\gamma\) is a constant speed. As a motivation for such a form of damping, we refer to equation (49) in \([40]\) which gives a scattering rate for charge carriers in graphene on a screened external charged impurity that is linear in the quasi-momentum to the leading order at low temperatures. The gap height \(h\) is also treated as an empirical parameter with a reasonable value of 1 Å. We note that our comparison with the HREELS experiment is only tentative because we do not include the low-frequency contributions from scattering kinematics or temperature effects \([39]\). Nevertheless, we expect that the loss function obtained from the Mermin polarization function with a suitable choice of \(\gamma\) will give the correct order of magnitude for spectral widths at frequencies that are not too low.

The effects of a SiC substrate on graphene are not as well understood as those of a SiO\(_2\) substrate, but without entering the current debate \([8, 9]\) we simply neglect any changes in graphene’s \(\pi\)-band structure that may result from a hybridization of its \(\pi\) orbitals with the substrate. In addition to treating the SiC substrate in the static mode with
dielectric constant $\varepsilon_0 = 9.7$, which was discussed in our previous work [35], we are able to reproduce some of the low-frequency features from the HREELS spectra [17] in a qualitative manner by including the dominant TO phonon mode of SiC in equation (7) with eigen-frequency $\omega_{TO} = 97$ meV [27], damping rate $\gamma_{TO} = 10$ meV [41], and high-frequency dielectric constant $\varepsilon_{\infty} = 6.5$ [27]. Also, by including the TO phonon in our comparison with the HREELS data, we gain some insight into the dynamic response of an insulating substrate prior to using it in calculations of the stopping force on a moving charge in section 3.2.

In figure 1, we display a comparison of the HREELS data [17] for wavenumbers ranging from 0.008 to 1.26 Å$^{-1}$ with the loss function obtained from the MDF-RPA polarization function with $\gamma = 0$ (solid lines) and the loss functions obtained from the Mermin polarization function with $\gamma = v_F k_0$ (dashed lines) and $\gamma = v_F q$ (dotted lines), where $k_0 = \sqrt{\pi n_0}$ and $n_0 = 1\times10^{13}$ cm$^{-2}$. We also show cases for which the SiC phonon is included in the model (thick lines) and for which the SiC substrate is treated in the static mode (thin lines). The equilibrium charge carrier density in graphene is set at $n = 1.9 \times 10^{13}$ cm$^{-2}$ (hence $k_F \approx 0.08$ Å$^{-1}$), giving the Fermi energy $E_F = h v_F k_F \approx 570$ meV to match experimental conditions [17]. Note that since the HREELS data is scaled to arbitrary units of intensity, the loss functions with finite $\gamma$ are scaled so that their maximum peak heights coincide with those from the experiment.

From figure 1, it can be seen that the best fit to the data can be obtained with the constant value $\gamma = v_F k_0$, while the best fit of the linear damping rate, $\gamma = v_F q$, tends to underestimate the widths of the experimental spectra at long wavelengths. Moreover, it appears that the inclusion of the substrate’s TO phonon hints at the experimentally observed non-dispersing plasmon at long wavelengths before it crosses into the inter-band SPEs continuum at $q \approx 0.07$ Å$^{-1}$ and becomes broadened from Landau damping. Interestingly, the plasmon peak is broader in the case with the substrate phonon than in the case of a static substrate. This is a consequence of the hybridization of the substrate phonon with damping rate $\gamma_{TO} = 10$ meV and graphene’s $\pi$ plasmon with a vanishing damping rate. Therefore, using the known value of $\gamma_{TO} = 10$ meV as a ‘marker’, one can surmise that the peaks shown in figure 1 with thick solid lines at long wavelengths are, in fact, the substrate phonon that is promoted due to an avoided crossing with the $\pi$ plasmon [16]. This idea will be discussed in more detail shortly.

Since the HREELS experiment [17] has been designed to determine the plasmon dispersion in graphene on an SiC substrate, in figure 2 we show the peak positions deduced from the experimental spectra and from the various theoretical models displayed in figure 1. In addition, we show the peak positions resulting from the polarization function for graphene
with the LFE correction, \( \Pi_{\text{LFE}}(q, \omega) \), in the \( \gamma = 0 \) case (dash-dot lines) [2]. While it is clear that the LFE correction is unable to reproduce the widths of the experimental spectra, figure 2 also demonstrates that its effects on the plasmon dispersion are hardly discernible from the case of the MDF-RPA polarization with \( \gamma = 0 \). Figure 2 further confirms that the case \( \gamma = v_F k_0 \) provides the best fit to the experimental plasmon dispersion. For the static substrate case (thin lines), one notices that the peak positions approach the value \( \omega = 0 \) as \( q \to 0 \) in a manner that depends on damping rate, so that a typical 2D plasmon with \( \omega \sim \sqrt{\gamma} \) is produced for \( \gamma = 0 \) and \( \gamma = v_F q \), while a constant \( \gamma \) appears to give rise to a quasi-acoustic relation \( \omega \sim q \) when \( q \to 0 \).

Finally, in all models of the plasmon peaks, the inclusion of the substrate phonon (thick lines) appears to capture the tendency of the experimental data to approach a finite frequency rather than vanish as \( q \to 0 \). A closer inspection shows that the frequency in question is near the value of 116 meV, which corresponds to the Fuchs–Kliever (FK) surface phonon at the frequency \( \omega_{\text{FK}} = \omega_{TO}(\sqrt{60} + 1)/(\epsilon_{\infty} + 1) \) [27, 41]. This behavior is commensurate with the concept of a phonon–plasmon hybridization [26], which is illustrated in figure 2 by two thick solid lines showing the zeros of the dielectric function for the MDF-RPA polarization function with \( \gamma = 0 \). These two lines display an avoided crossing near the point where the thin solid line (representing graphene’s \( \pi \) plasmon dispersion for a static substrate) would cross the FK phonon frequency \( \omega_{\text{FK}} \approx \omega_{TO} \approx 116 \) meV. One can see from figure 2 that the substrate phonon frequency starts at \( \omega_{\text{FK}} \) for \( q = 0 \) and is promoted to higher frequencies as \( q \) increases, following a dispersion curve similar to that of graphene’s \( \pi \) plasmon in the case of a static substrate. This promoted phonon dispersion enters the inter-band SPE continuum at \( \omega > v_F (2k_F - q) \) with \( \omega > v_F q \). On the other hand, graphene’s plasmon starts from the origin of the \((q, \omega)\) plane and initially follows the standard long wavelength plasmon dispersion \( \omega_p(q) = v_F \sqrt{2 \pi k_0 q^2} \) [22, 23, 37], but quickly levels to the TO phonon frequency \( \omega_{TO} = 97 \) meV as \( q \) increases. As a consequence of this avoided crossing [16], the plasmon branch is pushed into the intra-band SPEs continuum of graphene at \( \omega < v_F q \), where it broadens after crossing the point \( q_c \approx \omega_{TO}/v_F \approx 0.015 \) Å\(^{-1}\) from Landau damping. This avoided crossing appears to have been corroborated in figure 3 of [36], which shows dispersion curves from a subsequent HREELS experiment conducted by Liu and Willis using a smaller incident electron energy and a greater momentum resolution than in [17]. We finally note that this complicated scenario is depicted in figure 2 for \( n = 1.9 \times 10^{13} \) cm\(^{-2}\), which is the spontaneous doping density of graphene on an SiC substrate [17], but by changing graphene’s charge carrier density with a gate potential, for example, one can alter the picture of the plasmon–phonon hybridization quite significantly.

### 3.2. Stopping force with a dynamic substrate

After the discussion on the low-frequency excitations in graphene on an insulating substrate with a prominent TO phonon, we now explore how the various model parameters of this system affect the stopping force on a point charge, defined in equation (10). Given the complexity of the system, we first analyze the case of free graphene by letting \( h \to \infty \).

In figure 3, we show the velocity dependence of the stopping force on a proton \((Z = 1)\) moving a distance \( z_0 = 10 \) Å away from graphene for the cases of intrinsic graphene and graphene with a reduced charge carrier density \( n = n_0 \). From figure 3, one can see that all models give qualitatively similar results at high speeds, but at low speeds there are significant differences in the behavior of the stopping force due to variations in the charge carrier density \( n \) and the damping rate \( \gamma \). In particular, a threshold velocity \( v = v_F \) exists for the cases of intrinsic graphene with \( \gamma = 0 \) due to the lack of intra-band SPEs and the presence of inter-band SPEs in the \( \omega > v_F q \) region of the \((q, \omega)\) plane [22, 23]. Furthermore, all cases with \( \gamma > 0 \) or \( n > 0 \) result in a linear dependence of the stopping force on the particle speed, which can be described using the concept of friction. The slopes of these curves do not appear to be affected significantly by the choice of a constant or linear \( \gamma \), but the slopes do depend significantly on the density, as discussed previously [35]. It also appears that the LFE correction to the MDF-RPA polarization function has a small effect on the stopping force for speeds in the range \( v < v_F \). Therefore, we may conclude that the major influences on the polarization of graphene by an external charge are determined by the equilibrium charge carrier density \( n \) and the damping rate \( \gamma \), which can be described by a constant value.
We now focus on analyzing the effects of the TO phonon in an SiC substrate on the stopping force of a proton moving a distance $z_0 = 10 \text{ Å}$ away from graphene with a graphene–substrate gap height of $h = 1 \text{ Å}$. Given our results for free graphene, in figure 4 we present the stopping force for the damping rates $\gamma = 0$ and $\gamma = \nu_F k_0$, as well as for the two charge carrier densities $n = 0$ and $n = n_0 = 10^{13} \text{ cm}^{-2}$. In all cases, the inclusion of the phonon (thick lines) results in a surprisingly large increase of the stopping force for speeds in the range $v > 2\nu_F$ when compared with the case of a static substrate (thin lines). This is due to the plasmon–phonon hybridization and the fact that as the speed increases, the integration range $0 < \omega < q_\text{FK}$ in equation (10) probes a larger portion of the promoted phonon dispersion curve shown in figure 2, which approaches a finite frequency rather than vanishing as $q \rightarrow 0$. However, we are more interested in the stopping force at low speeds ($v < \nu_F$) for which the phonon–plasmon hybridization produces some peculiar effects. We first analyze the $\gamma = 0$ case in figure 4(a). For intrinsic graphene, which does not support a plasmon mode, one can see from the inset that the inclusion of the substrate phonon gives rise to a small but non-vanishing stopping force in the subthreshold region due entirely to the non-dispersing FK phonon at frequency $\omega_{FK} \approx 116 \text{ meV}$. For the finite density $n = n_0$, the inset in figure 4(a) shows that the linearly increasing stopping force in the static substrate case is augmented by a contribution from the original $\pi$ plasmon, which is pushed into the intra-substrate SPE region due to an avoided crossing with the substrate phonon and traverses this region at the constant frequency $\omega_{TO} \approx 97 \text{ meV}$ without dispersion but with broadening from Landau damping. It is important to note that the inclusion of the TO phonon destroys the linear velocity dependence of the stopping force at low speeds, making the concept of friction inappropriate for the $\gamma = 0$ case. Moving to the case of $\gamma = \nu_F k_0$, which is shown in figure 4(b), one notices that the large peak in the magnitude of the stopping force has been broadened and suppressed. Furthermore, although the effects of the substrate phonon are somewhat softened at low speeds, they still result in a non-linear dependence of the stopping force on the velocity for both $n = 0$ and $n_0$.

Finally, we analyze the stopping force of a slow charge moving parallel to a layer of graphene on an Al surface. From a first-principles calculation, it has been shown that graphene’s $\pi$ electron band is not destroyed by a hybridization with aluminum, leaving its elementary excitation modes intact [11]. However, aluminum is close to an ideal metal with a volume density of quasi-free electrons given by $N_0 = 3/(4\pi R_e^3)$, where $R_e \approx 2.07a_B$ and $a_B$ is the Bohr radius, and supports a high-frequency bulk plasmon as well as a well-defined continuum of low-energy SPEs. Without addressing the problem of a hybridization of graphene’s $\pi$ plasmon and aluminium’s surface plasmon, which may be satisfactorily discussed in a local approach, we explore the interplay of the low-energy intra-band SPEs in graphene and the low-energy SPEs in aluminum, which can only be treated properly via a non-local approach, e.g. by a surface dielectric function in equation (5). Since a linear expansion of both the MDF-RPA polarization function for graphene, given in equation (12), and the surface dielectric function for aluminum, obtained from equations (5) and (6), is possible for low frequencies, we can apply the concept of the friction coefficient $\eta$ defined in equation (11). We note that the $q$ integral in equation (11) takes into account two wavenumber cut-offs: $q < 2k_F = 2\sqrt{\pi n}$ for graphene and $q < 2K_F \approx 3.5 \text{ Å}^{-1}$ for aluminum. Since typical charge carrier densities in graphene have $k_F \ll K_F$, it is interesting to see how significant graphene’s screening ability is in a combined system with a metal. Following [11], we take the equilibrium density of graphene on aluminum to be $n = n_0 = 10^{13} \text{ cm}^{-2}$, and the gap height to be $h = 1 \text{ Å}$.

In figure 5, we show the dependence of the friction coefficient on the distance $z_0$ for a proton moving above free graphene, a free aluminum surface (which is placed in the plane $z_0 = -1 \text{ Å}$), and a graphene on aluminum system. We also show the friction coefficient for a particle moving above free graphene with $n = 0$. Although we only consider the case of vanishing damping, we note that the effects of finite damping can be quite large in both systems [20, 32, 35].
explore the effects of the density any further, from the low-energy SPEs in aluminum. Although we do not one can see that graphene’s static screening reduces the friction contribution from graphene’s intra-band SPEs to the friction, system with intrinsic graphene, for which there is no direct range and dominates for \( z_0 > 4 \) Å. Furthermore, the friction coefficient of the combined system is suppressed when compared to both the case of free Al (at short distances) and the case of free graphene (at long distances). In the combined system with intrinsic graphene, for which there is no direct contribution from graphene’s intra-band SPEs to the friction, one can see that graphene’s static screening reduces the friction from low-energy SPEs in aluminum. Although we do not explore the effects of the density any further, from the \( n = 0 \) and \( n_0 \) cases one can surmise that the friction of a combined graphene on metal system can be controlled by changing the doping level of graphene externally.

4. Concluding remarks

We have provided a simple theoretical model that describes the dielectric coupling of graphene, represented by a 2D gas of massless Dirac’s fermions, and a semi-infinite 3D substrate, represented by a surface response function in a non-local formulation. The underlying assumption that the electronic band structures of these two systems are not altered by their proximity has allowed us to study the hybridization of their excitation modes due to Coulomb interaction. An emphasis was placed on the role of the dynamic response of a substrate in the low-frequency excitations of the combined graphene–substrate system, which give rise to the energy loss of slow moving charges above graphene. A comparison of the loss function for the graphene on an insulator system with a HREELS experiment was used as a motivation to explore methods of improving the polarization function for free graphene in the RPA, as well as to discuss hybridization between the \( \pi \) plasmon in graphene and the surface phonon in an SiC substrate, which enabled us to predict an avoided crossing between the dispersion curves of these two collective modes.

It was found that the inclusion of either the linearly dispersing damping rate or the local field correction in graphene’s polarization function does not improve comparison of the loss function with the HREELS experimental data. Further analysis showed that these two parameters also have small effects on the stopping force for a point charge moving parallel to free graphene when compared to the effects of a constant damping rate and the equilibrium density of charge carriers. These negative results likely indicate that both the linear dispersion of damping and the Hubbard approximation for the local field correction involve model assumptions that are too crude, leaving open the possibility for further refinements.

On the other hand, the stopping force for a charge moving over graphene supported by an SiC substrate, which was described by an empirical dielectric function that included an optical phonon excitation mode without dispersion, revealed quite strong effects coming from the hybridization of graphene’s plasmon and the substrate’s phonon for both low and high charge speeds. As another example, a friction coefficient was calculated for a slow charge moving above graphene on a metallic substrate by using Lindhard’s dielectric function to obtain a surface response function representing the low-energy single-particle excitations in aluminum in a non-local formulation. In the limit of vanishing damping rates for both graphene and aluminum, an interesting interplay was found between the low-energy single-particle excitations of both systems in the friction coefficient.

In all cases studied, the low-energy excitation modes in graphene were found to be strongly coupled with those in the substrate, giving rise to various forms of the stopping force on external charges that can be of interest to studies such as the chemical reactivity of graphene. Particularly intriguing is the possibility of affecting the dynamic response of the combined graphene–substrate system by changing the charge density in graphene by external means, thereby enabling a control of friction at the nano-scale.

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