In this Comment, we would like to address a series of papers by H.-Y. Deng on surface plasmons at metal-dielectric interfaces. The first one appeared in 2015 on the arXiv (with co-workers K. Wakabayashi and C.-H. Lam) and was finally published in Phys. Rev. B under the title “Universal self-amplification channel for surface plasma waves” [Ref.1]. A paper claiming the same phenomenon to occur in a metallic film appeared in the same year[2], followed by an alternative argument for the instability of surface plasma waves based on energy conservation[3]. In New J. Phys. (2019), Deng has also addressed the question why this prediction went unnoticed in the literature[4]. In other arXiv posts[5–7] the instability of surface plasmons is also mentioned, while the scope is widened, e.g., to energy electron loss spectroscopy.

The discovery of the surface plasmon dates back to 1957 when Ritchie[8] formulated a general model for the energy loss spectrum of charged particles passing through a thin metal foil, a topic that had attracted numerous experimental investigations. Ritchie could successfully describe collective and individual excitations of the metallic electrons, that are accompanied by oscillating electric fields. Next to the already known concept of volume plasmons introduced by Pines and Bohn[9,10] Ritchie found an additional loss peak below the plasma frequency which is proper to a bounded metal and was later called the surface plasmon. In the 1970s the theoretical description of (surface) plasmons moved from classical grounds to incorporate quantum aspects of the electron response[11,12] using, e.g., the jellium model to describe the metal and applying tools of the ideal Fermi gas[13,14]. Theoretical and experimental progress has led to a variety of applications in the now sprawling field of plasmonics, for example nano-scale light sources as the surface plasmon nanolaser[15] or spaser[16,17] that may be useful as a biosensor, in microscopy, optical computing and information storage. A big challenge to real-life plasmonic devices are the large losses in metals. Dissipation channels are provided by collisions of conduction electrons, by Landau damping and interband absorption. This can be mitigated by the introduction of amplifying media (see, e.g., Refs. [16] and [18]). In contrast to these proposals, Deng has suggested that there exist an “intrinsic channel of amplification” that would involve the ballistic motion of carriers reflected from the metal surface[19,20]. This has triggered the present work. We consider a geometry similar to Deng’s, consisting of a metal-vacuum interface that is planar on the macroscopic scale, the metallic body being essentially infinitely thick. The conduction electrons are described by the jellium model (i.e., ignoring the structure of the crystal backbone).

To make the paper self-contained, we recall in Sec. [1] the main ideas of Deng’s model. We analyze his statement that a non-vanishing normal current must exist at the surface to warrant the existence of surface plasmons. A key issue is how to deal with charge conservation and the spatial structure of the charge distribution. In Sec. [2] we present in detail the semiclassical model that is used for the electronic response, based on the Boltzmann equation in the relaxation time approximation and supplemented by boundary conditions. We review the history of scattering models (partially specular, partially diffuse) and compare to Deng’s results. A detailed estimate of the surface plasmon loss rate, based on the energy balance argument of Ref. [3], is computed in Sec. [3]. We find indeed, similar to Deng, that the scattering of electrons at the metal surface produces one term corresponding to amplification. The surface plasmon is, however, overall lossy when all terms are taken into account. A surface contribution to the energy balance crucial to Deng’s analysis is argued to be questionable.

Keywords: surface plasmon, surface electrodynamics
In Sec. [IV] we show how a macroscopic electrodynamic model can be embedded into an approach based on excess interfacial fields developed by Bedeaux and Vlieger (BV). This clarifies the issue whether charge conservation provides sufficient information to calculate the dispersion relation. We also recall that the usual hydrodynamic model with a vanishing normal surface current does indeed allow for surface plasmons. At this stage, a mathematical and physical discussion on how to perform the local limit is given. In Sec. [V] we supplement the macroscopic descriptions of Deng and of BV by a surface electrodynamics approach due to García-Moliner and Flores (GF) [19, 20], designed with an apparently similar scope to Deng’s recent work [4]. For the response in the bulk metal, one could take here any conductivity with or without spatial dispersion. Following GF, we display the surface plasmon dispersion for a hydrodynamic model. Sec. [VI] summarizes our conclusions about the validity of Deng’s proposal.

I. A CHARGE-CENTERED FORMULATION OF SURFACE ELECTRODYNAMICS

A. Geometry: metallic half-space

The geometry considered is that of a metal and a dielectric (typically vacuum) occupying the half-spaces \( z \geq 0 \) and \( z < 0 \), respectively. The metal surface is macroscopically located at \( z = 0 \) and infinitely extended in the \( xy \)-plane. It appears flat on the scale of the plasmon wavelength \( 2\pi/k \) where \( k \) is the wave vector parallel to the surface. The description of Refs. [1] and [4] is macroscopic in the sense that the microscopic details in the surface region (its width \( d_s \) is typically a few lattice constants) are not resolved: we work in the limit \( kd_s \to 0 \). The electric current density in the metal can then be written as

\[
\mathbf{j}(x, t) = \mathbf{J}(x, t) \Theta(z),
\]

where \( \mathbf{J} \) is the current inside the metal at position \( x = (x, y, z)^T \) and \( \Theta \) the Heaviside step function that represents the rapid change in the surface region.

Deng’s formulation of the surface plasmon problem is focused on the dynamics of the charge and current densities, while the electric field is eliminated in a self-consistent way by using Coulomb’s law. His formulation of charge conservation [3] takes the form

\[
\text{Deng: } (\partial_t + \tau^{-1}) \rho + \nabla \cdot \mathbf{j} = 0,
\]

where \( 1/\tau \) is the collision rate of conduction electrons. Because of the collision term, Eq. (2) does not locally conserve charge. Actually, it follows from a naive application of the relaxation time approximation to the Boltzmann equation (see Sec. [II] below). The failure of not conserving charge locally should be discussed carefully. In the history of metal optics, this problem appeared several times in different guise – a brief summary is given in Subsec. [I D]. We may ignore it for the moment, knowing that most of Deng’s actual estimates are taken in the collisionless limit \( \tau \to \infty \) anyway.

Inserting Eq. (1) for the current density, one gets for \( z \geq 0 \)

\[
\text{Deng: } (\partial_t + \tau^{-1}) \rho + \Theta(z) \nabla \cdot \mathbf{j} = -\Theta'(z) J_z(x_0),
\]

where \( x_0 = (x, y, 0) \) is a position in the surface and \( \Theta'(z) = \delta(z) \) is localized in the surface region. The term \( -\Theta'(z) J_z(x_0) \) on the right hand side is described by Deng [1] after Eq. (24): “Physically, the right hand side of Eq. (3) means that charges must pile up on the surface if they do not come to a halt before they reach it.” He considers this term to be crucial for the existence of a surface plasmon which is claimed to exist only if the normal component \( J_z(x_0) = \lim_{z \to 0} J_z(x) \) of the current density (called “surface current” in the following) does not vanish. Otherwise, “the surface would be completely severed from the rest of the metal” [3] and only volume plasmons could be excited [3].

According to an anecdote told by Plummer et al. [11], a discussion between Ritchie and Gabor brought up the insight that it is actually the electric field normal to the surface that must be nonzero to generate a surface plasmon. This condition does not imply that the surface current be nonzero: examples are provided by various formulations of a non-local current response (spatial dispersion) that go back historically to the anomalous skin effect (see Secs. [II] and [IV]).

On the microscopic scale, the current smoothly changes between the bulk values in both media, vacuum and metal in our case. But on a macroscopic scale, information about the surface region gets lost. One key question that we would like to clarify here is what are the length scales a given “macroscopic” model actually tries to resolve (or not). Different cases disagree among each other in terms of the values (zero or not) of surface current and charge. In this spirit, the localized term \( \Theta'(z) \) on the right of Eq. (3) is the result of not resolving the surface region in which the electron density drops to zero.

To illustrate this point, consider the Drude model for the response of the metallic electrons to the electric field. We shall treat the system in linear response and assume that all fields are proportional to \( \exp(i(kx - \omega t)) \). Only a \( z \)-dependence remains. We also work on spatial scales where electromagnetic retardation can be neglected. The electric field, for example, can then be generated by a potential \( \phi(z) \) and has nonzero components \( E_x = -ik\phi \) and \( E_z = -\partial_x \phi \).

In the Drude model, the current density \( \mathbf{J}(z) = \sigma(\omega) \mathbf{E}(z) \) depends only on the local electric field at the same position in the metal (\( z \geq 0 \)). By taking the integral of Eq. (2) or (3) over a thin layer centered around \( z = 0 \) whose thickness eventually shrinks to zero, we get

\[
-\ii \omega \rho_s + J_z(0^+) = 0
\]
This links the time derivative of the charge $\rho$, in the surface layer and the normal component of the current density, evaluated in the metal just outside the layer. (On the vacuum side, $j_z(0^-) = 0$, of course.) Note that we have adopted here the canonical formulation of the continuity equation, dropping the relaxation time $\tau$ from Eq. (2). By Coulomb’s law, the surface charge generates an electric potential (cgs units)

$$\phi(z) = 2\pi \rho_z \frac{e^{-k|z|}}{k}$$  

(5)

whose decay length $1/k$ is set by the periodic variation of all fields parallel to the surface. According to the Drude model, the surface current in Eq. (4) takes the form

$$J_z(0^+) = k\sigma(\omega)\phi(0) = k\frac{i\omega_p^2}{4\pi(\omega + i/\tau)}\phi(0)$$  

(6)

where $\omega_p$ is the plasma frequency and $\tau$ the relaxation time of the electric current (average scattering time of electrons). The three equations (4)–(6) yield, provided that $\rho_z \neq 0$, the dispersion equation $\omega(\omega) = \omega_p^2/2$ with $\omega = \omega + i/\tau$. Its solution is

$$\omega(k) = \sqrt{\frac{\omega_p^2}{2} - \frac{1}{4\tau^2} - \frac{i}{2\tau}}$$  

(Drude model)  

(7)

This simple calculation provides a starting point to compare with Deng’s results regarding several points.

The imaginary part of the surface plasmon frequency satisfies $-1/\tau < \Im \omega(k) < 0$: the surface plasmon is damped. For the auxiliary complex frequency $\tilde{\omega}$, one gets $\Im \tilde{\omega} \geq 0$, as claimed by Deng. He works with a relaxation term in the continuity equation [see Eq. (2)] and finds a local dispersion equation in the form $\omega^2 = \omega_p^2/2$, giving a damping twice as large. While in Eq. (7), $\Im \tilde{\omega}$ depends on the relaxation rate $1/\tau$, the text in Ref. 1 around Eq. (8), claims the contrary.

For typical metals, we have $\omega_p \gg 1/\tau$ so that the frequency $\omega_s = \omega_p/\sqrt{2}$ sets the long-wavelength limit of the surface plasmon dispersion (real part) at a metal–vacuum interface. Several authors have shown, both on general grounds and for particular models, that this remains true beyond the local (Drude) approximation. Deng claims in Refs. 1 and 3 that the long-wavelength limit $\omega(k \to 0)$ should depend on model parameters [see Eq. (14) below].

The calculation assumes that the charge density is nonzero only in the thin surface layer $z \sim 0$, otherwise the electric potential (5) must be modified. In the local approximation, the electric field component $E_z$ changes sign when $z = 0$ is crossed. This may have led to an erroneous result that can be inferred from Ref. 3 [the paragraph after Eq. (11)]: there $J_z(0^+)$ appears with a sign opposite to Eq. (6).

The passage to the local limit and the handling of charge conservation in general, will be a key point of our discussion in what follows. For example, we analyze in Sec. IV B 1 how the results of the hydrodynamic model recover those of the Drude approach. In Appendix A 1 we give a discussion of integral representations for the electric field, in particular how they behave in the limit $z \downarrow 0$.

B. Plasmon dispersion relation as an eigenvalue problem

We come back to Deng’s formulation of the surface plasmon where a non-local relation between current and field (spatially dispersive conductivity) is adopted. This motivates the introduction of an integral operator $\hat{H}$ that relates the current divergence and the charge according to

$$(-i\omega) \nabla \cdot J(z) = \hat{H} \rho(z) = \int dz' \hat{H}(z, z') \rho(z'),$$  

(8)

where again $\omega = \omega + i/\tau$. As an intermediate step, the operator $\hat{H}$ involves solving the Poisson equation to get the electric potential for a given charge density. For the conductivity, a semiclassical kinetic theory based on the Boltzmann equation in the relaxation time approximation similar to earlier work by Reuter and Sondheimer and Wagner, for example, is taken. This calculation is simplified by representing the charge density as a cosine transform:

$$\rho(z) = \frac{2}{\pi} \int_0^\infty dq \rho_q \cos(qz).$$  

(9)

In addition, Deng focuses on that part of the operator $\hat{H}$ that describes the excitations of an infinite system, neglecting terms depending on scattering at the surface. (For a discussion of this approximation, refer to the paragraph after Eq. (16).) Its (double) cosine transform may then be given by

$$\hat{H}(q, q') \approx \Omega^2(k, q) \delta(q - q'),$$  

(10)

where $\Omega(k, q)$ is related to the bulk dispersion relation. Using this result in the cosine transform of Eq. (3) and using Eq. (9), one finds that the charge density is given by

$$\rho_q = \frac{i\omega J_z(0)}{\Omega^2(k, q) - \tilde{\omega}^2},$$  

(11)

where $J_z(0)$ is the amplitude of the surface current. (If the correct continuity equation is used, the denominator contains the product $\omega\tilde{\omega}$ rather than $\tilde{\omega}^2$.) If one sets $J_z(0) = 0$ and seeks a solution with $\rho_q \neq 0$, then Eq. (11) yields the dispersion relation for bulk excitations, $\omega^2 - \Omega^2(k, q) = 0$ (neglecting, as mentioned, the influence of boundary conditions on the bulk plasmon spectral density.)

The last step towards surface plasmon modes is to express the surface current by another integral operator

$$i\omega J_z(0) = \int_0^\infty dq \frac{G(K, \omega)}{K^2 + q^2} \rho_q,$$  

(12)
where $\mathbf{K} = (k, 0, q)$. If we insert $\rho_q$ under the integral and simplify both sides by $J_z(0) \neq 0$, we get

$$\int_0^\infty \frac{dq}{k^2 + q^2} \frac{G(K, \omega)}{\Omega^2(k, q) - \omega^2} = 1. \quad (13)$$

Deng solves this equation numerically. The results can be written as a complex dispersion relation for surface plasmons,

$$\omega = \omega_s(k) + 1(\gamma_0(k) - 1/\tau), \quad (14)$$

where Fig. 1 from Ref. 11 gives the following typical values: for $k \approx 0.07 \omega_p/v_F$, the surface plasmon frequency is $\omega_s(k) \sim 0.9 \omega_p$ and its imaginary part $\gamma_0(k) \sim 0.1 \ldots 0.2 \omega_p$, a positive value highlighting the instability. (Recall that for metals like gold and silver, one has $\omega_p \sim 100/\tau$.) Here $\omega_p$ is the metal’s plasma frequency and $v_F$ the Fermi velocity. In Ref. 3 the approximation $\gamma_0(k) \approx \omega_p(0.16 - 0.066 p) - 0.25 kv_F$ is found, where $0 \leq p < 1$ is a parameter describing the fraction of electrons that show specular reflection at the metal surface.

It is remarkable that these numbers deviate strongly from earlier work. The long-wavelength limit of the real part $\omega_s(k) \to \omega_p/\sqrt{2}$ (for a metal-vacuum interface) is well established, follows from the matching of macroscopic fields in the local approximation (dielectric function $\varepsilon(\omega) = -1$) and is consistently recovered in models including spatial dispersion.

The imaginary part of the dispersion relation is even more surprising: it is the key claim of Deng’s paper,11 that the ballistic motion of electrons, after reflection from the surface, provides an amplification channel that may overtake the loss rate $1/\tau$ in Eq. (14). One may raise the question why in that case the Fermi sea of filled electronic levels should become unstable since it is constructed as the stage of lowest energy for a fixed charge density.

C. Approximations within Deng’s description

From Eqs. (6, 12) we learn that the kernels $\mathcal{H}$ and $G$ can be determined from the current density which is itself given by

$$J_{\mu}(z) = \sum_{\nu} \int dz' \sigma_{\mu\nu}(z, z') E_\nu(z') \quad (15)$$

within linear response theory. Here, $\mu, \nu \in \{x, y, z\}$ label Cartesian components. Exploiting the translation symmetry of our surface problem, the conductivity tensor $\sigma_{\mu\nu}(z, z')$ depends on two positions in addition to wavevector $k$ and frequency $\omega$ (spatial and temporal dispersion). The integration over the variable $z'$ translates for example the ballistic motion of electrons on the scale of the mean free path $v_F \tau$. The dependence of $\sigma$ on two positions $z, z'$ (rather than their difference) describes the breaking of translational symmetry by the boundary conditions. We emphasize that $\sigma(z, z')$ is the conductivity tensor of the metallic half-space and includes surface scattering. It can be generally decomposed into

$$\sigma(z, z') = \sigma_b(z - z') + \sigma_s(z, z'), \quad (16)$$

where $\sigma_b$ is the bulk conductivity and $\sigma_s$ embodies surface effects. Deng passes this decomposition on to the operators $\mathcal{H}$ and $G$.

There are two assumptions that lead to the diagonalization of $\mathcal{H}$ in Eq. (10): First, Deng assumes that the current in the metallic bulk is mainly determined by $\sigma_b$ and neglects the contribution of $\sigma_s$ which is expected to be localized at the surface. We discuss in Sec. V an approach which takes $\sigma_s$ into account. Second, the cosine transform that is used to represent the charge density in Eq. (9), makes boundary terms vanish that involve the derivative $\partial_z \rho(0)$. This is not true, however, for a charge distribution that behaves like $\rho(0) \exp(-\kappa z)$, for example, whose cosine transform is simply

$$\rho_q = \frac{\rho(0) \kappa}{q^2 + \kappa^2}. \quad (17)$$

We show in Secs. IVB-IVC that this $\rho_q$’s hydrodynamic model with the finite gradient $\partial_z \rho(0)$ appears naturally because of spatial dispersion; it can be conveniently represented within a Fourier expansion of $\rho(z)$. In this model, a surface plasmon is found although the surface current vanishes, $J_z(0) = 0$. It provides a counter-example to Deng’s interpretation of Eq. (11) that surface plasmons with a finite charge density should necessarily have a nonzero surface current.

The decomposition of the kernel $G$ in Eq. (12) into bulk and surface parts $G_b + G_s$ follows from Eqs. (15, 16), evaluating the current at $z = 0$. Deng identifies the surface part $G_s$ with translation symmetry breaking and finds it to play a key role for the amplification of the surface plasmon. In all explicit applications except when using the Boltzmann equation (semiclassical model, SCM), he approximates the bulk conductivity $\sigma_b$ by its local Drude form for the calculation of $G_b$.

In contrast to Deng’s proposition, the specular reflection model does, in fact, include translation symmetry breaking. This can be seen by writing the integral (15) in the form

$$J_{\mu}(z) = \int_0^\infty dz' \sum_{\nu} \sigma_{b, \mu\nu}(z - z') E_\nu(z') \quad (18)$$

$$+ \int_0^\infty dz' \left[ \sigma_{b, \mu x}(z + z') E_x(z') - \sigma_{b, \mu z}(z + z') E_z(z') \right].$$

The first term alone would be the result of the so-called dielectric approximation,23 where the integration range in Eq. (15) is restricted to $z' \geq 0$ and $\sigma$ is replaced by its bulk version. This approximation is also used by Deng22 when he computes the approximate form $\mathcal{H}_0$ to get Eq. (11). (This can be seen from the lower integration limit of the $z'$-integral written after his Eq. (6).) The second term describes the specular scattering
of charges accelerated towards the surface. Following Ritchie and Marusak, it can be viewed as the response of the bulk conductivity to the electric field extended by mirror symmetry to $z' < 0$

$$E(-z') = ME(z' > 0),$$

(19)

where $M = \text{diag}(1, 1, -1)$ is the mirror reflection at the surface $z = 0$. Such an extended field has been called “pseudo-field” in the work of García-Moliner and Flores, and the sign flip of its normal component corresponds to a “fictitious” charge sheet at $z = 0$. From Eq. (18), the decomposition of the conductivity can be read off, and obviously leads, in the limit $z \to 0$, to a finite value for $G_M$, i.e., translation symmetry breaking is present.

The approximations outlined here illustrate that there are implicit additional boundary conditions behind the approach of Deng which seems difficult to be considered universal. For the boundary conditions used in Deng’s version of the semiclassical model, see Sec. II.

### D. Charge conservation

Another reason for the deviation of Deng’s results from the literature may be found in the way charge conservation is handled. It was already pointed out that Eq. (2) does not conserve charge in the metal bulk because of the scattering rate $1/\tau$. This problem arises from the relaxation time approximation to the Boltzmann equation that guarantees charge conservation only at the global level. As suggested by Mermin, this can be improved by specifying that the collision term relaxes the system’s distribution function to its local equilibrium value. In Sec. II we show how this correction can be used to recover the actual form of the continuity equation. Another way to enforce local charge conservation is to use the Boltzmann equation only to determine the current density and to solve for $\rho$ from the continuity equation (24). This strategy was followed, for example, in Ref. [25]. Deng follows a similar strategy [see end of §3 in Ref. [25]], although he uses the continuity equation (2) with the relaxation term. Other, more recent examples of implementing conservation laws in the Boltzmann equation can be found in Refs. [30] and [31]. Atwal and Ashcroft, for example, compute hydrodynamic approximations to the bulk plasmon dispersion relation.

The treatment of the near-surface charge density according to Eq. (3) reveals another inconsistency. It should contain two types of charges: surface charges with area density $\rho_s$ that are (on the macroscopic scale) localised at the surface $z = 0$, and a smooth charge density $\rho_b(z)$ in the bulk $z > 0$ (see Sec. IV.A for details). Indeed, the distribution $\Theta'(z)$ on the rhs must have a surface charge as its pendant on the lhs; the divergence $\nabla \cdot J$ being non-singular by Eq. (1). By splitting the continuity equation into bulk and surface parts, one gets

$$\partial_t \rho_b + \nabla \cdot J_b = 0,$$

$$\partial_t \rho_s + \nabla_\parallel \cdot J_s = -J_{b,z}(x_0),$$

(20)

rather than Eq. (3), with a surface coordinate $x_0$. The localized current density $J_b$ is by consistency parallel to the surface, hence only the parallel part $\nabla_\parallel$ of the gradient. The second equation illustrates that a nonzero current $J_{b,z}(x_0)$ signals a transfer from bulk to surface charge. If we adopt a model with $J_b = 0$, the second line of Eq. (20) yields Eq. (4) used above.

In the literature, this problem is handled in different ways. In the local approximation, there is only a surface charge which is generated by a jump in the Ohmic current $\sigma E$. Models that treat the electron dynamics in more detail lead in general to a non-local current-field relation [as in Eq. (15)]. The majority of authors simply exclude any surface charge and use the bulk component $\rho_b$ in the first line of Eq. (20) to model a charge density localized within the sub-surface region on some spatial scale that is actually resolved in the model. Typical candidates for this scale are the Thomas-Debye length $v_F/\omega_p$ and the mean free path $v_F \tau$. Consistency with Eqs. (20) then requires the surface current $J_s(0)$ to vanish. Contrary to Deng’s claims, this does not exclude the existence of a surface plasmon mode as we recapitulate in several examples throughout this paper.

Alternative approaches keep both bulk and surface charge, but are then in need of a model for the evolution of the surface charge that cannot increase indefinitely. We review recent examples in Sec. IV within the framework of Eq. (20). An explicit splitting of the charge into bulk and surface parts is not manifest in Deng’s papers. It appears in several places, however, that he has in mind a charge sheet localized at the surface. The limiting value $\rho_s = \lim_{q \to \infty} \rho_b$ of the cosine transform obviously provides its amplitude. If this is the only relevant charge, the electric field behaves like $e^{-kz}$, but Deng notes that this can only be used “outside the layer of surface charges” [after Eq. (30) and (33) of Ref. [3]]. In Ref. [4] he points out for a metal-dielectric interface where currents on both sides are different, that charges may “pile up” in the interface region (which is not resolved on the macroscopic scale). Deng calls this a “capacitive effect”, having probably in mind the Coulomb energy in this region of high charge density. Apart from that remark, no special treatment is applied to secure that charge be conserved during the transfer between bulk and surface. (In particular, a charge trap ensues if no desorption from the interface region is implemented in the model.)

Finally, Deng uses in the semiclassical model (SCM), based on the Boltzmann equation, a boundary condition for the distribution function that combines specular and diffuse scattering with probabilities $\rho$ and $1 - \rho$, respectively. We review this approach in the following section. It may suffice to say that the consistency of this treatment with respect to charge con-
servation has been discussed in the literature because it generates a nonzero surface current that is excluded unless one allows for a genuine surface charge. Zaremba, for example, suggested that the fraction $1 - \rho$ of diffusely scattered electrons should not simply disappear from the current balance, but be described by a different velocity distribution.

We mention in Sec. II B a general formulation in terms of a boundary scattering kernel put forward in Ref. 35. The same criticism of an unphysical surface current has been formulated against the dielectric approximation mentioned after Eq. (18), see for example Ref. 36.

II. SEMICLASSICAL (BOLTZMANN) MODEL FOR ELECTRON DYNAMICS

A Boltzmann description for the response of metallic conduction electrons has been used intensively in the past, starting with the anomalous skin effect where also a surface/interface problem had to be solved. We review in this section a solution to the Boltzmann equation at an interface, within the relaxation time approximation and implementing local charge conservation. We recall in particular the handling of electron scattering at the metal surface.

In this section, we consider the case that the electronic density contains only a “bulk” part in the sense of Eq. (20). An alternative model system where volume electrons can be trapped in the surface region and where a stationary state is defined by the balance between trapping of incoming electrons and desorption back into the bulk, will be treated in Sec. IV. Another approach to the surface plasmon dispersion relation that is not based on the Boltzmann equation, is presented in Sec. V.

A. The volume problem

The Boltzmann equation determines the distribution function for the metal electrons in phase space $f(v, x, t)$. The relaxation time approximation replaces the collision term by $(f_0 - f)/\tau$ where $f_0$ is the equilibrium (Fermi-Dirac) distribution that depends only on the electron energy. Integrating over all velocities, one gets Eq. (2), understood as describing bulk charges only, which does not locally conserve the charge. Warren and Ferrell and Mermin showed that this can be repaired with a relaxation term $(f_{\text{loc}} - f)/\tau$ where $f_{\text{loc}}$ differs from $f_0$ by a shift in the Fermi energy such that the local electron density is $n_0 + \delta n(x, t)$. Using the scaling law for free electrons, $\epsilon_F \sim n_0^{2/3}$, one gets

$$f_{\text{loc}}(v, x, t) = f_0 - f_0' \frac{2\epsilon_F}{3n_0} \delta n(x, t)$$  \hspace{1cm} \text{(21)}$$
to first order in $\delta n$. Here $\epsilon_F$ is the equilibrium Fermi energy and $f_0'$ the energy derivative of the Fermi-Dirac distribution.

Expanding the distribution function to first order in the electric field, $f = f_0 + f_1 + \ldots$, the modified Boltzmann equation yields

$$[\partial_t + \tau^{-1} + v \cdot \nabla] f_1(v, x, t) = -f_0' \left[ \epsilon v \cdot E(x, t) + \frac{2\epsilon_F}{3n_0 \tau} \delta n(x, t) \right], \hspace{1cm} \text{(22)}$$

where $\epsilon$ is the electron charge. When both sides are integrated over velocity space, we get

$$[\partial_t + \tau^{-1}] \rho(x, t) + \nabla \cdot J(x, t) = \frac{\epsilon}{\tau} \delta n(x, t),$$ \hspace{1cm} \text{(23)}$$
where the charge $\rho$ and current $J$ densities are the zeroth and first moment of the perturbed velocity distribution $f_1$. If we now take $\rho = e\delta n$, the terms involving $1/\tau$ cancel and local charge conservation follows:

$$\partial_t \rho(x, t) + \nabla \cdot J(x, t) = 0,$$ \hspace{1cm} \text{(24)}$$
rather than Eq. (3). There is no localized source term here on the rhs (and the boundary condition for the surface current is $J_z(x_0) = 0$), unless one introduces a surface component in the charge density, as in Eq. (20).

For the surface plasmon problem, the distribution function inherits the dependence $\exp[\alpha(kz - \omega t)]$ of the electric field, and Eq. (22) yields

$$[\partial_z + \eta] f_1(v, z) = -f_0' \frac{\epsilon v \cdot E(z)}{v_z} \left[ \epsilon v \cdot E(z) + \frac{2\epsilon_F}{3n_0 \tau} \eta z \right].$$ \hspace{1cm} \text{(25)}$$
Here we have defined the (complex) inverse length $\eta = (1/\tau - \omega + ikv_z)/v_z$. To simplify the following calculations, we drop the Mermin correction (the term with $\delta n$ on the rhs). It was also not taken into account by Deng.

B. The surface problem

We now discuss how the solutions to Eq. (25) involve the boundary conditions at $z = 0$. The general solution is

$$f_1(v, z) = C(v) e^{-\eta z} - f_0' \frac{\epsilon}{v_z} \int_0^z dz' \epsilon v \cdot E(z') e^{\eta(z' - z)},$$ \hspace{1cm} \text{(26)}$$
where the function $C(v)$ must be determined from the asymptotic behaviour of $f_1$. This is a standard procedure that proceeds by considering separately the cases $v_z \leq 0$. For electrons moving towards the surface, $v_z < 0$, causality requires that their contribution can only depend on the electric field along their path in the past, $z' > z$. Requiring that $f_1(v, z)$ vanishes for $z \to \infty$, we get

$$v_z < 0: \quad C(v) = f_0' \frac{\epsilon}{v_z} \int_0^\infty dz' \epsilon v \cdot E(z') e^{\eta z'}. \hspace{1cm} \text{(27)}$$
Deng insists that the convergence of the integral in Eq. (27) is only ensured when Re $\eta < 0$. Allowing for a complex frequency $\omega$, this yields for $v_z < 0$ the condition Im $\omega + 1/\tau > 0$. 

\[\]
There are no contradictions in the specular case $p = 1$ where Eqs. (28,32) coincide and lead to $J_\parallel(0) = 0$ (by mirror symmetry with respect to the plane $z = 0$). He comments on this specular reflection model in integral

$$v'_z < 0 : \quad \int_{v'_z > 0} d^3v' S(v' \rightarrow v) = 1 . \quad (30)$$

A partially diffuse surface can be described by splitting $S$ into a specularly reflecting part with weight $p$, proportional to $\delta(v - v'_c)$, and a diffuse part proportional to $\cos \theta$ where $\theta$ is the angle of the final velocity $v$ relative to the surface normal (the $z$-axis\(^25\). By taking the cosine to a higher power, the diffuse scattering would be preferentially along the surface normal\(^25\). In both cases, there is no memory left of the direction of the incident velocity. This approximate scattering model describes a surface which is rough on the scale of the Fermi wavelength $\lambda_F$ both in rms height and correlation length. Since $\lambda_F$ is much smaller than the plasmon wavelength, the surface can still be viewed as smooth from a macroscopic perspective. It is thus legitimate to assume translation invariance so that $S$ does not depend on the in-plane coordinates $x, y$. Assuming elastic scattering and keeping in mind the normalization (30), we arrive at

$$v'^2 S(v' \rightarrow v) = \delta(v - v') \times$$

$$\left[ p \delta(\phi - \phi') \delta(\cos \theta + \cos \theta') + \frac{1}{\pi} \frac{p}{\cos \theta} \right] , \quad (31)$$

where $\theta, \ldots \phi'$ are spherical coordinates relative to the surface normal. Inserting this into Eq. (29), we obtain the boundary condition

$$v_z > 0 : \quad C(v) = p C(v_\perp) + (1 - p) A(v) \quad (32)$$

with

$$A(v) = \frac{1}{\pi} \int_{\cos \theta' < 0} d\Omega' \left( -\cos \theta' \right) C(v, \theta', \phi') , \quad (33)$$

where $d\Omega' = \sin \theta' d\theta' d\phi'$ and $C$ is given by Eq. (27). This is a simple additional boundary condition (ABC) to be used in conjunction with the Boltzmann equation when there is no accumulation of charges at the surface. The additional term $(1 - p)A(v)$ in Eq. (32) describes the diffuse scattering; it takes care of balancing the currents of incoming and outgoing charges.\(^{33}\) The procedure leading to this ABC illustrates also that the response of a surface is a problem on its own: it cannot be solved from information about the bulk behaviour alone. It is only conceivable within microscopic models that no ABC is needed: in that case, the behaviour of the electron wave functions in the surface region embodies the (additional) boundary condition. This would be the viewpoint of density functional theory\(^{33}\) for example.

One may ask why Deng did not run into contradictions in the specular case $p = 1$ where Eqs. (28,32) coincide and lead to $J_\parallel(0) = 0$ (by mirror symmetry with respect to the plane $z = 0$). He comments on this specular reflection model in...
Appendix B of Ref. 4, where the nature of the fictitious charge sheet is analyzed, which we have alluded to after Eq. (19). The existence of surface plasmons is connected to a divergence of the fictitious charge amplitude. Deng manages to transform this condition into the dispersion relation of Eq. (13) by suitably identifying the bulk dielectric function, although the division by \( J_s(0) = 0 \) is illegitimate.

C. Approximate solution

We now determine the actual distribution function for a surface plasmon problem. The equations determined so far are still lacking a specific form for the electric field. We compute its potential \( \phi \) by solving Poisson’s equation \( \Delta \phi = -4\pi \rho \). Since we are interested in a solution with zero surface current, only volume charge is present. Keeping in mind that in our geometry \( \nabla = (ik, 0, \partial_z) \) and using the method of Green’s functions, we arrive at

\[
\phi(z) = \frac{2\pi}{k} \int_{-\infty}^{\infty} dq' e^{-k|z-z'|} \rho(q') \tag{34}
\]

within the metal half space. Rather than the cosine transform used by Deng, we use the Fourier expansion

\[
\rho(z) = \int_{-\infty}^{\infty} dq \ e^{iqz} \tilde{\rho}(q) \tag{35}
\]

for the charge density \( \rho \). It turns out that the charge density varies on length scales \( v_F/\omega_p \) much shorter than the plasmon wavelength \( 1/k \). Therefore, Deng introduces the approximation of a charge sheet insofar as it appears under the integral \( [\text{34}] \). Using the form \( \rho(z) \approx \rho_0 \delta(z - d) \) with \( d > 0 \) slightly located in the metal, we obtain the fields

\[
\phi(z) = \frac{2\pi \rho_0}{k_0} \ e^{-k|z-d|} \tag{36}
\]

\[
\frac{E_x(z)}{E_z(z)} = -2i\rho_0 \int dq \ \frac{\bar{k}}{\bar{k}^2 + \omega^2} \ e^{i\bar{k}(z-d)} = 2\pi \rho_0 \left( \frac{-i}{\text{sgn}(z-d)} \right) e^{-k|z-d|}, \tag{37}
\]

where \( \text{sgn} \) is the sign function. The \( z \)-component of this expression is discontinuous at \( z = d \). By allowing for \( d > 0 \), we can control carefully how to interchange the \( q \)-integral with the \( z \)-integral of Eq. (26): according to the residue theorem, the position relative to the charge layer at \( z = d \) decides where to close the integration path in the complex \( q \)-plane. Deng takes \( d = 0 \) and seems to ignore this difficulty.

The following calculations are straightforward: we put the electric field \( [37] \) into the solution \( [26] \) of the Boltzmann equation, use the boundary conditions \( [32] \) and \( [33] \) and integrate separately over the surface region \( (0 < z < d) \) and the bulk of the metal \( (z > d) \). Eventually we take the limit \( d \to 0 \). We use this scheme in Sec. III to compute the power loss of the surface plasmon. One finds that the contributions from the region \( 0 < z < d \) vanish when the limit \( d \to 0 \) is taken. We therefore present in the following the results in this limit only.

The procedure generates the following result for the boundary term \( C(v) \) [Eq. (27)]:

\[
v_z < 0 : \quad C(v) = -\frac{2\pi \rho_0 f_0'}{k} F_0(k, \bar{\omega}, v) \tag{38}
\]

\[
F_0(k, \bar{\omega}, v) = \frac{k \cdot v}{\bar{\omega} - k \cdot v}, \tag{39}
\]

where \( k = (k, 0, ik) \) and \( \bar{\omega} = \omega + i/\tau \). The distribution function itself comes as a formidable sum of three terms as follows

\[
\begin{align*}
\frac{f_0(v, z)}{2\pi \rho_0 f_0'} &= -\frac{e^{-kz}}{k} F_0(k, \bar{\omega}, v), \quad (40a) \\
\frac{f_{s,F}(v, z)}{2\pi \rho_0 f_0'} &= \Theta(v_z) \frac{e^{-qz}}{k} \left[ F_0(k, \bar{\omega}, v) - p F_0(k, \bar{\omega}, v_z) \right], \quad (40b) \\
\frac{f_{s,Z}(v, z)}{2\pi \rho_0 f_0'} &\approx \Theta(v_z) e^{-qz/(1-p)} \frac{2v}{\bar{\omega}} \left( \frac{i}{3} + \frac{k v}{8 \bar{\omega}} \right), \quad (40c)
\end{align*}
\]

where \( \eta = -i(\bar{\omega} - kv_z)/v_z \), as defined after Eq. (25). Here, \( f_0 \) represents the volume solution in an infinitely extended metal to a field with momentum \( k \), \( f_{s,F} \) describes the specularly scattered fraction in the Fuchs model [Eq. (28)], and \( f_{s,Z} \) takes into account diffuse scattering within Zaremba’s model [second term of Eq. (32)]. In evaluating this last term, we have taken the two leading terms for small \( kv_F/\bar{\omega} \).

D. Comparison to Deng

Expressions of similar complexity are also found in Refs. 4 and 6 although a Fourier/cosine integral over \( \rho_0 \) is performed in the last step. In concrete evaluations, the same simplification as here is applied, see for example from Ref. 3 “using \( E_z(z) \approx 2\pi \rho_0 e^{-kz} \) outside the layer of surface charges” [i.e. for \( z > d \to 0 \) in our notation, see Eq. (37)]. Our calculation takes advantage of evaluating this integral with the residue theorem first and thus avoids convergence problems at large \( q \). To ensure convergence at large \( q \), Deng indeed has to introduce a cutoff \( q_c \sim \omega_p/\omega_F \).

From the boundary conditions to the Boltzmann equation stated above, we may thus expect that the terms \( f_0 + f_{s,F} \) correspond essentially with Deng’s expressions, while \( f_{s,Z} \) is a correction that takes care of charge conservation at the partially diffuse surface. In Appendix A we check that this is indeed the case, and the results, according to Deng’s notation, are displayed in Table 1. It is remarkable to which extent the splitting into “bulk” and “surface” is ambiguous. In the following section, we evaluate the amplification (or loss) rate of the surface plasmon using the energy balance argument developed in Ref. 3.
We emphasize that the surface current is the power into real and imaginary parts, decomposing the frequency into real and imaginary parts. Decomposing the fields into real and imaginary parts, one gets the surface area, one gets the surface current. The amplification rate of the surface plasmon is determined by Deng by a power balance equation that is derived from charge conservation. Eq. (3) is multiplied by \( \phi(x, t) \) and integrated over \( x \). We find two differences with respect to Deng's argument. First, the collision term \( \rho/\tau \) on the lhs should be suppressed, as charge must be conserved locally. Second, when the “\( \nabla \cdot J \) term” is integrated by parts, we get

\[
\int d^3x \phi(\nabla \cdot J) = -\int dA \phi(x_0) J_z(x_0) + \int d^3x \Phi \cdot J. \tag{41}
\]

The first term on the rhs is a surface integral at \( z = 0 \) (with coordinates \( x_0 \)), while the other boundary, deep in the bulk, does not contribute, of course. This boundary term cancels exactly the integral over the singular term \( -\Theta'(z) J_z(x_0) \) on the rhs of Eq. (3):

\[
-\int d^3x \Theta'(z) \phi(x) J_z(x_0) = -\int dA \phi(x_0) J_z(x_0). \tag{42}
\]

We emphasize that the surface current \( J_z(x_0) \) drops out at this stage from the energy balance which takes the form

\[
\int d^3x \phi \partial_t \rho + \int d^3x \Phi \cdot J = 0. \tag{43}
\]

To proceed, we use the exponential \( x- \) and \( t- \) dependence of all fields to re-write the term \( \phi \partial_t \rho \) as the derivative of an electromagnetic energy. Factoring out the surface area, one gets the balance equation \( \partial_t \mathcal{E} + \mathcal{P} = 0 \), where \( \mathcal{E} \) and \( \mathcal{P} \) are the electromagnetic potential energy and power per unit area. Decomposing the frequency into real and imaginary parts, \( \omega = \omega_s + i \gamma \), we get

\[
\gamma = -\frac{\mathcal{P}}{2\mathcal{E}}. \tag{44}
\]

with energy and power per unit area, averaged over one oscillation cycle, given by

\[
\mathcal{E} = \frac{1}{4} \int dz \Re \left[ \phi^*(z) \rho(z) \right] e^{2\gamma t}, \tag{45a}
\]

\[
\mathcal{P} = \frac{1}{2} \int dz \Re \left[ \Phi^*(z) \cdot J(z) \right] e^{2\gamma t}. \tag{45b}
\]

A positive \( \gamma \) would signal an instability where energy from the electronic system is converted into a surface plasmon oscillation.

If we compare Eq. (45) with Eq. (8) of Ref. 3, we see that on the lhs, just \( \gamma \) rather than \( \gamma + 1/\tau \) appears, which is due to the collision term in question. Deng adds on the rhs the power (per area) due to the surface current

\[
\mathcal{P}_{sc} = \frac{1}{2} \Re \left[ \phi^*(0) J_z(0) \right] e^{2\gamma t} \tag{46}
\]

which should, as mentioned above, cancel with the boundary term from the partial integration [Eq. (41)]. As the surface power \( \mathcal{P}_{sc} \) plays a key role in the claimed surface plasmon instability of Ref 3 this cancellation is a crucial point. We discuss its contribution in Sec. III B where we add up the imaginary parts of the frequency.

The energy balance condition (44) provides a simple way to picture a potential instability of the surface plasmon. With \( \gamma > 0 \) and from Eq. (45b), it would feature a current density opposite to the electric field (and in phase with it) because \( \mathcal{E} \) will always be positive. Now, due to partial reflection at the inner surface, it is possible that a fraction of charges indeed flows “against the field”. Ritchie and Marusak phrase their analysis into the following, however: “The origin of surface plasmon damping in the present approximation [treating the electrons within the Boltzmann equation] lies in the fact that the surface, which is assumed infinitely massive, is able to absorb momentum. Thus an electron in the semi-infinite gas may collide with the surface, lose momentum to it, and then may be able to interact with a surface plasmon.” [Ref. 22 end of § 1] We detail in the following the different contributions to the power \( \mathcal{P} \).

| Type    | “\( F_{\pm} \)”                                                                 | “\( F_0 \)”                                                                 |
|---------|---------------------------------------------------------------------------------|--------------------------------------------------------------------------------|
| “bulk”  | \(-\frac{e^{-kz} F_0(\mathbf{k}, \omega, \mathbf{v})}{2(\Theta(v_c) e^{-\eta z} [F_0(\mathbf{k}, \omega, \mathbf{v}) - F_0(\mathbf{k}, \omega, \mathbf{v}_-)]) + e^{-kz} F_0(\mathbf{k}, \omega, \mathbf{v})}\) | \(+\frac{e^{-kz} F_0(\mathbf{k}, \omega, \mathbf{v})}{2(\Theta(v_c) e^{-\eta z} [F_0(\mathbf{k}, \omega, \mathbf{v}) - F_0(\mathbf{k}, \omega, \mathbf{v}_-)])} + e^{-kz} F_0(\mathbf{k}, \omega, \mathbf{v})\) |
| “surface” | \(+\Theta(v_c) e^{-\eta z} [F_0(\mathbf{k}, \omega, \mathbf{v}) - F_0(\mathbf{k}, \omega, \mathbf{v}_-)]\) | \(-\Theta(v_c) e^{-\eta z} [F_0(\mathbf{k}, \omega, \mathbf{v}) - p F_0(\mathbf{k}, \omega, \mathbf{v}_-)]\) |
| total   | \(-e^{-kz} F_0(\mathbf{k}, \omega, \mathbf{v}) + \Theta(v_c) e^{-\eta z} [F_0(\mathbf{k}, \omega, \mathbf{v}) - p F_0(\mathbf{k}, \omega, \mathbf{v}_-)]\) | \(+e^{-kz} F_0(\mathbf{k}, \omega, \mathbf{v}) + \Theta(v_c) e^{-\eta z} [F_0(\mathbf{k}, \omega, \mathbf{v}) - p F_0(\mathbf{k}, \omega, \mathbf{v}_-)]\) |

TABLE I. Contributions to the electronic distribution function according to the labels used in Refs. 3 and 4. The table rows correspond to Eqs. (A11), (A12) in Appendix A, the columns to the terms involving the functions \( F_0 \) and \( F_\pm \) defined in Eq. (39) and Eqs. (A13), (A14). We collect the results of the \( dq \)-integrations using the approximation \( \rho \approx \rho_c \). Evaluating the integrals with the residue theorem, the “\( F_\pm \)” terms turn into the “\( F_0 \)” term. A common factor \( 2\pi \epsilon \rho_c f_0^2/k \) is taken out.

III. SURFACE PLASMON AMPLIFICATION RATE

A. Electric energy balance

The amplification rate of the surface plasmon is determined by Deng by a power balance equation that is derived from charge conservation. Eq. (3) is multiplied by \( \phi(x, t) \) and integrated over \( x \). We find two differences with respect to Deng’s argument. First, the collision term \( \rho/\tau \) on the lhs should be suppressed, as charge must be conserved locally. Second, when the “\( \nabla \cdot J \)” term is integrated by parts, we get

\[
\int d^3x \phi(\nabla \cdot J) = -\int dA \phi(x_0) J_z(x_0) + \int d^3x \Phi \cdot J. \tag{41}
\]

The first term on the rhs is a surface integral at \( z = 0 \) (with coordinates \( x_0 \)), while the other boundary, deep in the bulk, does not contribute, of course. This boundary term cancels exactly the integral over the singular term \( -\Theta'(z) J_z(x_0) \) on the rhs of Eq. (3):

\[
-\int d^3x \Theta'(z) \phi(x) J_z(x_0) = -\int dA \phi(x_0) J_z(x_0). \tag{42}
\]

We emphasize that the surface current \( J_z(x_0) \) drops out at this stage from the energy balance which takes the form

\[
\int d^3x \phi \partial_t \rho + \int d^3x \Phi \cdot J = 0. \tag{43}
\]

To proceed, we use the exponential \( x- \) and \( t- \) dependence of all fields to re-write the term \( \phi \partial_t \rho \) as the derivative of an electromagnetic energy. Factoring out the surface area, one gets the balance equation \( \partial_t \mathcal{E} + \mathcal{P} = 0 \), where \( \mathcal{E} \) and \( \mathcal{P} \) are the electromagnetic potential energy and power per unit area. Decomposing the frequency into real and imaginary parts, \( \omega = \omega_s + i \gamma \), we get

\[
\gamma = -\frac{\mathcal{P}}{2\mathcal{E}}. \tag{44}
\]
B. Discussion of the amplification rate

From the potential (36) we get for the energy density [Eq. (45a)]
\[ \mathcal{E} = \frac{\pi |P_s|^2}{2k} e^{2\gamma t}. \] (47)

The contributions to the distribution function listed in Eq. (40) generate the current density \( J \) needed for the power \( P \) [Eq. (45b)]. To simplify the velocity integrals, we continue to expand consistently to the first order in \( kv_F/\omega_s \) and \( \gamma/\omega_s \) and take for the real part \( \omega_s = \omega_p/\sqrt{2} \), the well-known long-wavelength limit of the surface plasmon frequency.\(^{23,24}\)

Referring to the three parts of the distribution function [Eq. (40)], we get a self-consistent equation with three terms
\[ \gamma = \gamma_b + \gamma_{s,F} + \gamma_{s,Z}, \] (48)
namely
\[ \gamma_b = -\gamma - \frac{1}{\tau}, \] (49a)
\[ \gamma_{s,F} = -\frac{3}{4} kv_F \left( 3(1 - p) - \frac{1}{16} k v_F \right), \] (49b)
\[ \gamma_{s,Z} = -\frac{1}{3} - \frac{p}{3} kv_F. \] (49c)

The bulk contribution \( \gamma_b \) follows from the factor \( 1/\bar{\omega} \) in the distribution function (40). When inserted into Eq. (48), it effectively halves the amplification rate \( \gamma \). All contributions provide a damping of the surface plasmon, so that a detailed comparison to Deng’s result (3) is in order. This requires some care, since our calculation is organised in a different manner. In particular, the assignment of terms to “bulk” and “surface” is not unique (see Table I). Recall, for example, the first line in Eq. (13), which resembles a bulk contribution, although it acquires a surface character because the integral is cut off at the metal surface (dielectric approximation). We therefore aim at discussing the complete result(s) for the amplification rate. Its dependence on the plasmon momentum \( k \) may also guide our physical insight.

First of all, the terms depending on the relaxation rate \( 1/\tau \) cannot be compared in a meaningful way because in Ref. (3) the equation of continuity in the bulk system [Eq. (2)] contains an unphysical charge relaxation.

Second, the behaviour of the current at the surface is completely different. As mentioned after Eq. (41), the contribution from the jump in the current density, \( P_{sc} \) [Eq. (46)], drops out from the energy balance. In Deng’s papers, however, it plays a key role [see Ref. 3, Sec. 6]. He finds that the contributions to lowest order in \( kv_F/\omega_s \) cancel each other in the sum \( P + P_{sc} \). This is equivalent to removing the term \( \gamma_b \) in Eq. (45) and leads to amplification rates that come out twice as large as in our calculation.

Equipped with this rule, we can analyze the contribution to the amplification rate that Deng would reach from the power \( P \), had he ignored the surface current. From the last equation (not numbered) in Ref. 3, Sec. 5 (with \( p = 1 \)):
\[ \text{Deng, } P \text{ only: } \gamma = -\frac{1}{\tau} - \frac{3}{2} kv_F. \] (50)

The last term is twice as large as the first term of \( \gamma_{s,F} \), but has the same sign (plasmon damping). It is interpreted by Deng as the contribution of Landau damping because it arises from a pole in his \( q \)-integrals at \( q = i\eta \). (This condition is indeed equivalent to \( \omega = kv_F + kv_z \), where an electron moves in phase with the electric field.) This is consistent with our calculation, since the same term appears from a part of the distribution function that varies with \( e^{-\eta^2} \) [\( f_{s,F} \), see Eq. (40b)].

It is interesting to note that the term proportional to \( 1 - p \) in Eq. (49b) is positive. Since it is related to the fraction of charges that are not specularly reflected, it seems to confirm Deng’s picture that the symmetry breaking by the surface is essential for the amplification of the plasmon mode. This does not hold in our calculation, however, because one has to add the term (49c) to avoid having a charge sink at the surface. The sum of the two contributions is negative, \( 3/16 - 1/3 = -7/48 \), so that also the diffuse scattering provides an overall damping channel.

Third, let us try to get an idea what would change when the surface current contribution \( P_{sc} \) to the power had been kept. This may give a semi-quantitative estimate of the error incurred in Deng’s calculation. In our calculation, the surface model is constructed in such a way that the surface current \( J_z(0) = 0 \) (see discussion in Sec. II B). It appears by inspection that Deng’s solution for the distribution function only contains the terms called \( f_0 \) and \( f_{s,F} \) listed in Eq. (40); he uses indeed the Fuchs boundary condition (28) without further modifications. This means that we can use the correction term \( f_{s,Z} \) to estimate Deng’s result for the surface current (note the minus sign)
\[ \text{Deng: } J_z(0) = -N \int d^3 v e v_z f_{s,Z}(v, z = 0). \] (51)

Here, \( N = (m/2\pi\hbar)^3 \) is a scale factor for the velocity integral when the distribution function \( f_0 \) is taken as the Fermi-Dirac distribution. This calculation is fairly easy because the velocity dependence of \( f_{s,Z}(v, z = 0) \) is simple [see Eq. (40c)], and we get
\[ \text{Deng: } J_z(0) = \frac{1}{4} - p - \frac{\omega_p^2}{\omega} \left( \frac{3}{2} kv_F \right). \] (52)

The scaling with the diffuse scattering fraction \( 1 - p \) is as expected from the qualitative discussion in Sec. II B: it corresponds to the ‘missing charge’ that is not (speculatively) reflected. The corresponding power gets contributions from the \( 1/\bar{\omega} \) term and a term linear in \( kv_F \). Working out the ratio to the electrostatic energy, we find
\[ \text{Deng: } \gamma_{sc} = -\frac{1}{4} - p - \frac{\omega_p^2}{\omega} \left( \frac{3}{2} kv_F \right). \] (53)
This is also a damping contribution, in distinction to the claim around Eq. (42) from Ref. 3. If we added it to the three terms in Eq. (51), we would get a similar structure as in Deng’s Eq. (44) where the amplification rate $\gamma$ must be computed in a self-consistent way.

Let us finally illustrate the poor convergence at large $q$ of Deng’s amplification rate, Eq. (42) from Ref. 3. The term that is claimed to “dominate all the contributions from other parts of $P$ and $P_{\text{sc}}$” is given by [the scale factor $N$ was defined after Eq. (51)]

$$
\Gamma = \frac{1 - p}{\rho_s} \int_{-\infty}^{\infty} dq \rho_q \times \int_{v_z > 0} d^3v \left(1 - v_z^2\right) v_z \left(\frac{K \cdot v}{\omega} - \frac{K}{\omega} \cdot v\right), \tag{54}
$$

where $K = (k, 0, q)$ and $\rho_q$ is extended in an even way to $q < 0$. From the calculations displayed by Deng, it is clear that this term arises by subtracting its limit for small $Kv_F/\omega_s$. This leads, however, to a poor convergence of the $q$-integral, as can be easily seen by performing the $v$-integration first. The result of this (numerical) calculation is shown in Fig. 1: the Landau peak at $q \approx k_s = \omega_s/v_F$ is close to the cutoff momentum $q_c$ [the value $q_c = 1.5 k_s$ is taken from Ref. 1]. From the calculation of integrals like (54) with the residue theorem [see Appendix A2], we expect that the result is mainly imaginary [up to small corrections $O(1/\omega_s \tau)$]. Consistent with this is the visual impression in Fig. 1 that the areas under the real part of the integrand cancel.

This can be checked by performing the $dq$-integral first, leading to the entry “$g_b$ | $F_{\pm}$” in Table 1, and then evaluating the $d^3v$-integral. We find that this term contributes to the damping rate the expression given in Eq. (53), multiplied by $-2$. The correction to the local approximation is of relative order $k v_F/\omega_s$, in contradiction to the claim in Ref. 3, Eq. (43). Taken alone, this term would be interpreted as amplification. It is, however, just one contribution. There are terms that are dropped in Deng’s calculation of the “surface power” $P_{\text{sc}}$, for example those that appear proportional to $\sin qz$ in an integral representation of the electric field [Eq. (A2)]. Although this term appears to vanish at $z = 0$, the $q$-integral may actually generate a function that is discontinuous at $z = 0$ and whose limiting value for $z \downarrow 0$ is nonzero (see the Appendix A1). This discussion is somewhat futile, since, recalling the argument of Sec. III A, the “surface power” $P_{\text{sc}}$ should not be counted at all in the energy balance.

To summarize this section: we have performed an estimation of the surface plasmon amplification rate along an energy balance scheme put forward by Deng. It has been found that a correct treatment of charge conservation in the metal bulk and at its surface leads to striking differences: a potentially amplifying channel related to non-specular scattering at the surface turns into damping when care is taken to avoid a charge sink at $z = 0$. The surface current $J_s(0)$ that gives the major contribution in Deng’s argument is actually absent from the energy balance if the power exchanged between charges and field is computed in an appropriate way. The final result for the imaginary part of the surface plasmon frequency is

$$
\gamma = -\frac{1}{2\tau} - \left[\frac{3}{8} - \frac{7}{96}(1 - p)\right] k v_F. \tag{55}
$$

In line with other publications using the semiclassical model, we obtain an overall damped surface plasmon. Incidentally, non-specular scattering increases the damping proportional to the fraction $1 - p$. We shall see a similar result in Sec. IV B but obtained within a different approach that does not need to solve the Boltzmann equation. Eq. (55) is also consistent with a result of Zaremba using the SCM as can be seen in his Table III: the damping increases progressively as the scattered electrons have a more and more isotropic distribution.

IV. HYDRODYNAMIC MODEL

In the following, we embed Deng’s macroscopic model into an approach devised by Bedeaux and Vlieger (BV) which describes the charge density by a two-type model, as discussed in Sec. I D. The model naturally accounts for the “capacitive effect” and illustrates why the continuity equation does not provide enough information to determine the dispersion relation. We then specialize to the hydrodynamic model (HDM), collect explicit formulas for field and current profiles and discuss two different ways to perform the local limit. This serves to clarify misconceptions of Deng about plasmons at specularly reflecting surfaces and about taking the local value of the surface current. This eventually leads to a natural interpretation why additional boundary conditions are absent in the

![FIG. 1. Integrand of Eq. (54) for the integration over q (we added the contributions from q and $-q$, improving the UV convergence). The singularity appears at $Kv_F = \omega_s$, with $K = \sqrt{k^2 + q^2}$, but it is smoothed by the imaginary part of $\omega$. The vertical lines illustrate UV cutoffs quoted in Refs. 1 and 4. We assumed a ratio $\rho_q/\rho_s \approx 1$, the momentum scale is $k_s = \omega_s/v_F$.](image)
local limit. Eventually, we will use a two-type charge model\[44\] to illustrate the role of a finite surface current.

A. Bulk and surface charges

In the BV approach, charge and current density are decomposed into

\[
\rho_{\text{tot}}(x) = \rho_s \delta(z) + \rho(x) \Theta(z) \tag{56a}
\]
\[
J_{\text{tot}}(x) = J_z \delta(z) + J(x) \Theta(z) \tag{56b}
\]

where \( \rho \) and \( J \) are restricted to the bulk metal \((z > 0)\), while the localized \( \rho_s \) as well as \( J_z \) are called ‘excess quantities’\[53\]. Analogous decompositions are applied for all fields. The excess quantities describe on a macroscopic scale the differences between the actual surface electrodynamics and the extrapolated bulk dynamics. If the excesses were absent, ‘a sharp transition from one bulk phase to the other’ would be described, leading to Fresnel surface\[32\].

BV show that the normal component of the excess current \( J_{n,z} \) does not contribute to the matching conditions at the surface. So, without loss of generality, \( J_z \) shall be directed along the surface. If we plug Eqs. (56) into the continuity equation\[24\], separate localized and extended distributions and use the dependence of all fields proportional to \( \exp[\pm ikx - \omega t] \), we get

\[
\begin{align*}
  z > 0 &: \quad 0 = -i\omega \rho(z) + ikJ_z(z) + \partial_z J_z(z) \tag{57a} \\
  z = 0 &: \quad 0 = -i\omega \rho_s + ikJ_{s,x} + J_z(0^+) \tag{57b}
\end{align*}
\]

which is the fixed-frequency representation of Eq. (20). We thus get a pair of continuity equations that are coupled by the bulk current \( J_z(0^+) \) extrapolated to the surface. This current thus describes the charge exchange between bulk and surface.

As mentioned earlier, Deng does not split the charge density into surface and bulk parts. The parallel surface current \( J_z \) is also absent in his model. This illustrates that already a certain “additional boundary condition” (ABC) has been applied: in his model, the electrons which accumulate at the surface are not allowed to move along it. Neglecting the parallel component of the surface excess current (see Ref.\[24\]) for an estimation of its impact on the surface plasmon dispersion), Eq. (57b) yields

\[
\omega \rho_s = J_z(0^+) \tag{58}
\]

as already used in Sec. 4.A Eq. (4). A similar splitting of the charge into bulk and surface components can also be spotted in Deng’s papers. After Eq.(18) in Ref.\[3\] for example, the charge density computed from the distribution function is identified as a bulk charge. The nonzero value of the surface current points to a surface charge component, but the two components are not manifestly separated in the cosine transform \( \rho_q \) of the charge density. Within the approximation \( \rho_q \approx \rho_s \), the bulk charge component would vanish.

The excess field formalism derived by BV illustrates that the continuity equation for the (total) charge density is not sufficient to determine the surface plasmon dispersion relation. In particular, Eq. (58) must be supplemented by a model (often called an ABC) of how the accumulated charge reacts back on the surface current, e.g., by a repulsive force (“capacitive effect”) or a desorption process (“charge trap”). We provide a simple example in Sec. IV D.

B. Explicit sub-surface profiles

In Secs. 4.2 & A of Ref\[4\] Deng discusses surface plasmons for the hydrodynamic electronic response. The latter is described by the (linearized) Euler equation of fluid dynamics which determines the current through

\[
z > 0 : \quad J(z) = \frac{\sigma}{1 - i\omega \tau} E(z) - \frac{v_0^2 \tau}{1 - i\omega \tau} \nabla \rho(z) \tag{59}
\]

The first term is the Drude conductivity (DC value \( \sigma \)), magnetic forces are neglected, since they are of second order in deviations from equilibrium, and the second one translates the pressure arising from gradients in the charge density, using a linearized equation of state. Its coefficient is proportional to the compressibility of the electron fluid, and \( v_0 = O(v_F) \) gives the speed of charge density waves in the bulk (longitudinal speed of sound). The relaxation rate for the current density \( 1/\tau \) differs a priori from the one for the distribution function, but we keep the same letter for simplicity.

The bulk density \( \rho \) can be determined by virtue of Eqs. (57a,59). Starting with an exponential Ansatz and using Coulomb’s law \( \vec{E} = 4\pi \rho \), one finds

\[
\rho(z) = \rho(0^+) e^{-\kappa z} \tag{60}
\]

The inverse (complex) length scale \( \kappa \) is given by

\[
\kappa^2 = k^2 + \frac{\omega_p^2 - \omega \omega_c}{v_0^2} \tag{61}
\]

where \( \omega_p^2 = 4\pi \sigma / \tau \) is the squared plasma frequency. Note the term \( \omega \omega_c \) which appears as \( \omega^2 \) in Eq. (A7) of Ref\[4\] because of the wrong formulation of charge conservation. Adopting the viewpoint that Eq. (60) is the bulk charge and allowing for a charge \( \rho_s \) localized at the surface, we find from Eqs. (54,60) the electrostatic potential

\[
\begin{align*}
  z \geq 0 &: \quad \phi(z) = \frac{2\pi}{k} \left[ \rho_s e^{-kz} + \rho(0^+) \left( e^{-\kappa z} + \frac{e^{-\kappa z} - e^{-kz}}{k - \kappa} \right) \right] \tag{62a} \\
  z \leq 0 &: \quad \phi(z) = \frac{2\pi}{k} \left( \rho_s + \rho(0^+) \right) e^{kz} \tag{62b}
\end{align*}
\]
From this and Eqs. (59), the normal current density follows as

\[ J_z(z) = \frac{1}{\omega} \left\{ \frac{\omega^2}{2} \left[ \rho_s e^{-kz} + \frac{\rho_0(0^+)}{k} \left( \frac{\kappa e^{-\kappa z}}{k + \kappa} + \frac{\kappa e^{-\kappa z} - k e^{-kz}}{k - \kappa} \right) \right] + \nu_0^2 \kappa \rho(0^+) e^{-\kappa z} \right\}. \]

(63)

These equations cannot be solved because one has to determine the ratio \( \rho_s / \rho(0^+) \) between the two types of charges. To proceed, we adopt a model for the surface charge \( \rho_s \) or the surface current \( J_z(0^+) \). The two are directly related because of the charge conservation law \( \rho \).

1. Usual hydrodynamic boundary condition

In the hydrodynamic model, the condition

\[ J_z(0^+) = 0 \]

(64)

can be interpreted as the impossibility of concentrating the electron fluid into a true surface charge with zero extension at the surface; indeed, it implies \( \rho_s = 0 \). Because of the charge gradient \( \partial_z \rho(0^+) \neq 0 \) in Eq. (59), this boundary condition does not lead to a vanishing surface field \( E_z(0^+) \) and therefore allows for a surface plasmon mode\( ^{23} \) as discussed by Ritchie and Gabor\( ^{13} \).

Deng claims in Appendix A of Ref.\( ^{4} \) that this surface plasmon mode is wrong, and we shall examine his arguments in parallel to the relevant equations. From Eqs. (63) \& (64), we find [Ref.\( ^{4} \) Eq. (A9)]

\[ -\frac{\omega_s^2}{\kappa + k} + \nu_0^2 \kappa = 0 \]

(65)

with \( \omega_s = \omega_0/\sqrt{2} \). We have assumed \( \rho(0^+) \neq 0 \). Using the definition \( \rho \), one gets the dispersion relation

\[ \omega(k) = \Omega(k) - \frac{i}{2\tau}, \]

(66a)

\[ \Omega^2(k) = \omega_s^2 - \frac{1}{4\tau^2} + kv_0 \sqrt{\omega_s^2 + \frac{k^2 v_0^2}{4}} + \frac{k^2 v_0^2}{2}, \]

\[ \Omega(k) \approx \omega_s - \frac{1}{8\omega_s^2 \tau^2} + \frac{i}{2} \nu_0 k. \]

(66b)

In the last expression, we have taken the small-\( k \) limit to confirm that \( \omega_s \) is the long-wavelength limit of the surface plasmon dispersion, as it must\( ^{32} \). We recognize that the small parameter of this expansion is \( kv_0/\omega_s \), which coincides with \( k/\kappa_s \) introduced by Deng\( ^{23} \).

Eqs. (66a) \& (66b) correspond to Eq. (A10) of Ref.\( ^{4} \) except that Deng obtains a damping twice as large as here. Still, he claims that this surface plasmon is "plainly false" because of its behaviour in the local limit, i.e., for \( kv_0/\omega_s \ll 1 \). His argument hinges on the limiting value of the integrated (bulk) charge density \( Q = \int dz \rho(z) \). This depends on two parameters. It is true, of course, that the spatial extent \( \sim 1/\kappa \) of \( \rho(z) \) shrinks to zero, as can be seen from Eq. (61). (We neglect, for the simplicity of the argument, the imaginary part of \( \omega \).

One thus gets \( Q = \rho(0^+)/\kappa \approx v_0 \rho(0^+)/\omega_s \) in the local limit. For a meaningful comparison, however, we have to express the boundary value \( \rho(0^+) \) by a quantity that is well-defined in this limit. One candidate is indeed the potential \( \phi(0) \): from Eq. (62) with \( \rho_s = 0 \), we find

\[ \rho(0^+) = \frac{k \phi(0)}{2\pi}(k + \kappa) \]

(67)

Pulling these two expressions together, we have

\[ Q = \int dz \rho(z) \approx \frac{k \phi(0)}{2\pi} \text{ for } \kappa \to \infty \]

(68)

This coincides with the jump in the normal electric field, which the potential approaches in the local limit the form \( \phi(z) \approx \phi(0) e^{-k|z|} \) from Eqs. (62). Deng’s argument that the integrated charge density vanishes in the local limit\( ^{4} \) is thus fallacious.

2. The local limit

We have seen that in the local limit, the charge density of the hydrodynamic model shrinks to a surface charge. How does the current density behave to avoid a conflict between the boundary condition \( \rho (0^+ ) \) and the charge conservation law \( \rho \)? The answer requires elements from boundary layer (or multiple scale) techniques\( ^{15} \) and work out how the bulk charge density \( \rho(z) \) apparently becomes localized when the local limit is taken.
Let us note first that when the limit \( \nu_0 \to 0 \) is taken in Eq. (59), the order of the differential equation is reduced, which changes qualitatively the number of boundary conditions. Since the Maxwell boundary conditions for the fields hold independently of the local limit, it is the ABC (64) that has to be discarded. The behavior of the current density is illustrated in Fig. 2 for different values of the parameter \( k \nu_0/\omega_s \). Note how the current profile degenerates into a jump at the surface as \( k \nu_0/\omega_s \to 0 \). In boundary layer theory, one introduces an “inner expansion” for \( 0 \leq z \lesssim \nu_0/\omega_s \) (marked by the vertical dashed line) and an “outer” one for \( z \sim 1/k \). The two length scales get widely different in the local limit.

The outer expansion describes the current on the macroscopic scale. We keep \( z > 0 \) fixed in Eq. (63) and take the limit \( \nu_0 \to 0 \) or equivalently \( \kappa \to \infty \), giving

\[
z > 0 : \lim_{\kappa \to \infty} J_z(z) = \frac{i\omega_p^2}{4\pi\Omega}k\phi(0)e^{-kz} . \tag{69}\]

In this calculation, we discard terms \( e^{-\kappa z} \to 0 \) and keep \( e^{-kz} \).

This is the well-known local response according to Ohm’s law with an AC conductivity \( \sigma(\omega) = \omega_p^2/\bar{\sigma} \) (see Eq. (59)). This is shown as the dashed curve in Fig. 2. Since the outer expansion discards the boundary layer, it is not surprising that taking formally the limit \( z \to 0 \) in Eq. (69) gives a nonzero surface current. Its value conforms with charge conservation (58), considering the integrated charge density (68)…provided the frequency is fixed to \( \omega = \omega_0^2 \). In this way, the local calculation recovers the long-wavelength dispersion relation [including the losses spelled out in Eq. (66)]. A glance at Fig. 2 illustrates how the zero-surface-current ABC cannot be satisfied in the local limit.

To illustrate the characteristic behavior of the current within the boundary layer, we display the inner expansion. We work on the short length scale and take \( z \sim 1/\kappa \), where the hydrodynamic pressure (i.e., the density gradient) is significant, while the condition \( kz \ll 1 \) expresses the separation of length scales in the local limit. Using Eqs. (63) (67), we find

\[
z \sim 1/\kappa : \lim_{1/\kappa \to \infty} J_z(z) = k\phi(0)\sigma(\omega) \left(1 - e^{-\kappa z}\right) . \tag{70}\]

We recognize that this current suits the condition of a vanishing surface current and saturates to its local limit for \( 1/\kappa \ll z \ll 1/k \).

C. Critique of the hydrodynamic surface plasmon

In the two previous subsections, we have tried to argue how the surface plasmon of the usual hydrodynamic calculation connects smoothly with the local limit. We learned that the boundary condition (64) for the surface current does not conflict with the emergence of a surface mode, quite distinct from the claims in Refs. 1-4.

1. Dielectric function

The condition of a vanishing surface current is often interpreted as describing specular scattering at the surface 14. The specular scattering case \( p = 1 \) is special in the sense that a dispersion relation can be found with the help of a symmetry argument, using any dielectric function (conductivity) in the bulk metal 20,21,25. This relation reads

\[
1 = \frac{k}{\pi} \int \frac{dq}{K^2\epsilon_L(K,\omega)} , \tag{71}\]

where \( \epsilon_L(K,\omega) \) is the bulk dielectric function that only depends on the modulus \( K \) of the wave vector \( K = (k,0,q) \). The integral (71) can be worked out analytically as a contour integral by taking the (longitudinal) dielectric function of the hydrodynamical model

\[
\text{HDM: } \epsilon_L(K,\omega) = 1 - \frac{\omega_p^2}{\omega\omega - v_p^2K^2} . \tag{72}\]

The resulting sum over residues then yields the dispersion relation (65) found before by an elementary calculation.

We note that the dielectric function (72) corresponds to Eq. (59). This can be checked via the conductivity \( \sigma_L(K,\omega) \). (The subscript \( L \) is for “longitudinal”, as we are dealing with an electric field parallel to \( K \) in Fourier space.) Taking the spatial Fourier transform of (59) in a bulk medium and noting that \( K \cdot J = \omega p \) (charge conservation), one gets \( J = \sigma_L(K,\omega)E \) with

\[
\sigma_L(K,\omega) = \frac{i\omega\sigma/\tau}{\omega - v_p^2K^2} . \tag{73}\]

The standard relation \( \epsilon_L(K,\omega) = 1 + 4\pi i\sigma_L(K,\omega)/\omega \) then yields the hydrodynamic model (72). The pole of Eqs. (72) (73) at \( \omega \approx \nu_0 K \) is characteristic for the intrinsic sound waves in the electron gas. The zeros of the dielectric function (72) corresponds to bulk plasma waves that approach \( \omega_p \) for \( k \to 0 \) and disperse \( \sim \nu_0 K \) at large \( K \gg \omega_p/v_0 \). Replacing 1 by a background dielectric constant would be a way to incorporate the response of bound charges (e.g., d-electrons in gold).

Deng claims that there is a “non-equivalent approach” to the hydrodynamical model and uses for the dielectric function (4.2 and Footnote 1 in Ref. 4)

\[
\text{Deng: } \epsilon_L(K,\omega) = 1 - \frac{\Omega_0^2 + v_0^2K^2}{\omega} , \tag{74}\]

where \( \Omega_0 \) must be identified with the plasma frequency \( \omega_p \) from the small-\( K \) and high-frequency asymptotics. The condition \( \epsilon_L = 0 \) gives a similar dispersion relation for bulk plasma waves as above (with a different damping because of the square \( \omega^2 \) in the denominator), but sound-wave poles are absent. Note that the exact factors \( \omega \) in Eqs. (72) and (73) arise from charge conservation and the relation \( J = -i\omega P \) between the current density and the polarization field.
2. Breaking of translation symmetry

The fundamental equation for the surface plasmon dispersion in Refs. 10, 11 is Eq. (13) above, where the integral operators \( H \) and \( G \) introduced in Eqs. (5, 12) appear. What attracts attention is that for the bulk \( H \) operator, in most cases only the translation invariant part is taken, i.e., the kernel is approximated \( H(q,q') \sim \delta(q-q') \) [see Eq. (10) above]. Within the hydrodynamic formula \( G \) for the current density, we can identify the correction to this approximation. After a partial integration, one finds from the definition for the kernel \( H \):

\[
\int dq' \, H(q,q') \rho(q') = \int dz \cos(qz)(-i\bar{\omega})\nabla \cdot J
\]

\[
= v_0^2 \partial_z \rho(z^+)[0] + \int_0^\infty dz \cos(qz) \left( \omega_p^2 + v_0^2 K^2 \right) \rho(z)
\]

(75)

where we have set \( K^2 = k^2 + q^2 \). The integral on the rhs gives the cosine transform \( \rho_0 \) and represents the bulk (translation invariant) kernel \( H_\text{b}(k,q) = (\omega_p^2 + v_0^2 K^2) \delta(q-q') \). We shall argue in the following that the other term in \( H \) is nonzero, so that Deng’s statement that boundary terms are generally negligible compared to \( H_\text{b} \), should be treated with caution.

It is the derivative \( \partial_z \rho(0^+) \) that breaks translation invariance. It is set to zero by Deng because of the general form of the cosine transform \( \Theta \) [see, e.g., Ref. 4 before Eq. (A13)]. This is a subtle point, in particular when the local limit is considered. Indeed, the exponential charge density \( \rho_0 \) does have a nonzero derivative \( \partial_z \rho(0^+) = -\kappa \rho(0^+) \). In the local limit \( \kappa \rightarrow \infty \), this charge density provides an example of a “skew” representation of the \( \delta \)-function that is entirely localized in the region \( z \geq 0 \). Its cosine transform is well-defined for \( \kappa < \infty \) and provides the integral representation

\[
\rho(z) = \Theta(z) \frac{2Q \kappa^2}{\pi} \int_0^{\infty} dq \cos(qz) \frac{\omega_p^2}{\kappa^2 + q^2}
\]

(76)

Here, \( Q \) is the integrated charge (per area) defined in Eq. (68). The evaluation of \( \partial_z \rho \) requires an UV regularization of the integral. This can be performed by evaluating the integral as a contour integral along the entire real line in the complex \( q \)-plane, writing the integrand as \( q e^{iq}\bar{\omega}/(\kappa^2 + q^2) \), closing the contour with a circle at infinity in the upper half-plane and picking the residue at \( q = i\kappa \). If the limit \( \kappa \rightarrow \infty \) is taken first in Eq. (76), one gets a “symmetric” \( \delta \)-function and the prefactor \( \Theta(z) \) halves its weight to be consistent with the real-space representation \( \rho(z) \). Keeping \( \kappa \) finite, on the other hand, one gets a function \( \sim e^{-\kappa|z|} \) whose derivative at \( z = 0 \) is not zero, but shows a jump. This is in stark contrast to the naive analysis of the integrand around \( z = 0 \). (For more details on these integrations, see Appendix A11.)

We now evaluate the kernel \( G(k,q) \) that links the surface current \( J_z(0^+) \) to the charge density \( \rho_q \) [Eq. (12)]. From Eq. (59), we get

\[
i\bar{\omega} J_z(0^+) = \frac{\omega_p^2}{4\pi} \partial_z \phi(0) + v_0^2 \partial_z \rho(0^+)
\]

(77)

For the potential \( \phi(0) \) in the first term, there is no need to make the limit \( z \downarrow 0 \) explicit if we may assume that it is continuous across the boundary. (Only a “double layer” or perpendicular surface polarization would change this picture.) This term corresponds to the Drude model and would be taken by Deng as the translation-invariant part \( G_\text{b} \). One may wonder whether the second term with the derivative \( \partial_z \rho(0^+) \) breaks translation invariance: after all, the derivative \( \partial_z \rho \) appears in the same form anywhere in the bulk. This may have lead Deng to the statement [Ref. 3 Sec. 4.2 preceding Eq. (21)] that symmetry breaking is absent in the hydrodynamic model.

We would like to put forward the viewpoint that the symmetry breaking arises in hydrodynamics from the boundary condition \( J_z(0^+) = 0 \) itself. From this viewpoint, the hydrodynamic surface plasmon appears related to a charge distribution in the kernel of the non-trivial integral operator

\[
\text{HDM: } G(k,q) = -\frac{\omega_p^2}{\pi} \lim_{z \downarrow 0} (2q \sin(qz) - k)
\]

\[
- \frac{2v_0^2}{\pi} \lim_{z \downarrow 0} qK^2 \sin(qz)
\]

(78)

where the definition [12] of the integral operator \( G \) was applied. The first term corresponds to the Ohmic current response to the electric field, the second term to the charge density gradient. Note that the limit \( z \downarrow 0 \) must be taken as the last step [after the \( q \)-integration of Eq. (12)], since one may have to deal with singular charge distributions where values \( q \rightarrow \infty \) are significant. The local limit \( \rho(z) \rightarrow \rho_0 \delta(z) \) provides an example. Deng ignores the contribution \( 2q \sin(qz) \) to the first term and finds an electric field with the opposite sign. (It would apply at \( z \rightarrow 0^- \) rather than \( z \rightarrow 0^+ \) for a localized charge, see Eq. (77).) One may speculate whether such a sign change may be responsible for the surface plasmon amplification, when charges seem to “flow uphill”.

In view of this discussion, we may comment on the status of Eq. (13). It is fundamental to Deng’s analysis, see e.g., the evaluation in Ref. 4, where a splitting \( G = G_\text{b} + G_s \) is done and the imaginary part of \( G_s \) is responsible for amplification. The denominator \( \Omega^2(k,q) - \omega^2 \) in Eq. (13) arises from the bulk approximation to the kernel \( H \) as mentioned above, and misses the surface correction discussed above. If in the derivation one simplifies by \( J_z(0) \neq 0 \), then by charge conservation, a genuine surface charge \( \rho_s \) must be present, which is distinct, however, from the volume charge of which \( \rho_q \) is the cosine transform. We discuss such a “composite model” in the following section.
D. Composite charge model

To elaborate on the nature of the surface charge and current, we want to introduce a phenomenological boundary condition which has been derived by Horovitz and Henkel\(^\text{11}\) for this mesoscopic model. Solving the Boltzmann equation for the volume and (infinitely thin) surface region separately, but allowing for a collision term that mixes surface and bulk electrons, they obtain

\[
J_z(0^+) = \frac{\rho_s}{\tau_d} - \alpha v_0 \rho(0^+) ,
\]

(79)

where \(1/\tau_d\) and \(\alpha\) describe the desorption rate of surface electrons back into the bulk and a probability of trapping a bulk electron in the surface, respectively.\(^\text{11}\) The desorption process prevents the unphysical accumulation of charges at the surface which we have discussed in Sec.\(^\text{11D}\)

Eq. (79) provides the essential information on how the mixing of surface and bulk electrons takes place. This could not be accounted for by the continuity equation of the whole system itself, as we have argued after Eq. (58). If we had assumed a vanishing surface current, then Eq. (79) would describe the balance of trapping and desorption. But then also \(\rho_s\) would vanish by Eq. (58). Hence, to allow at least for a nonzero bulk charge, we would be forced to take \(\alpha = 0\) and mixing between the two charges would be completely absent.

In the following, we assume \(J_z(0^+) \neq 0\) which may be interpreted as a measure of non-specular scattering. Combined with charge conservation, we then get

\[
\rho_s = \frac{\alpha v_0 \tau_d}{1 - i \omega \tau_d} \rho(0^+) \quad (80)
\]

Using Eqs. (79) \(^\text{23}\) and (80) in Eqs. (58) \(\text{63}\) \(\text{63}\) we find the implicit dispersion relation

\[
0 = -\frac{\omega_s^2}{\kappa + \kappa} + \frac{v_0^2 \kappa}{\kappa} + \alpha v_0 \tau_d \left[ \omega_s^2 - \omega \left( \omega + \frac{i}{\tau} \right) \right] . \quad (81)
\]

The solution in the local limit \(v_0 \to 0\) is given by the well-known surface plasmon frequency\(\text{23}\)\(\text{23}\), i.e.

\[
v_0 \to 0 : \quad \omega \to \sqrt{\omega_s^2 - \frac{1}{4 \tau^2} - \frac{i}{2 \tau}} . \quad (82)
\]

In particular, trapping and desorption at the surface are irrelevant on the local scale – in contrast to Eq. (51) and Fig. A1(a) of Ref.\(\text{4}\) where the plasmon resonance depends on surface scattering.

We are interested in solving Eq. (81) for small \(k\) and expand in powers of \(k v_0/\omega_s\). For simplicity, bulk collisions are neglected (\(\tau \to \infty\)). Introducing the dimensionless quantities

\[
\tilde{\omega} = \frac{\omega}{\omega_s}, \quad \tilde{v}_0 = \frac{k v_0}{\omega_s}, \quad \tilde{\kappa} = \frac{\kappa v_0}{\omega_s} \quad \text{and} \quad \tilde{\tau}_d = \omega_s \tau_d ,
\]

(83)

we transform Eq. (81) into dimensionless form

\[
0 = -\frac{1}{\tilde{k} + \tilde{v}_0} + \tilde{\kappa} + \alpha \tilde{\tau}_d \frac{1 - \tilde{\omega}^2}{1 - i \tilde{\omega} \tilde{\tau}_d} . \quad (84)
\]

An expansion in powers of \(\tilde{v}_0\) gives the dispersion relation as

\[
\omega(k) = \omega_\text{s} + a k v_0 + b \frac{(k v_0)^2}{\omega_s} . \quad (85)
\]

Expanding \(\tilde{k}\) from Eq. (61) to second order in \(\tilde{v}_0\), and equating like powers of \(\tilde{v}_0\), we find to the second order

\[
\alpha = \frac{1 - i \alpha + \alpha/\tilde{\tau}_d + 1/\tilde{\tau}_d^2}{2 + 2 (\alpha + 1/\tilde{\tau}_d)^2} \quad (86a)
\]

\[
b = \frac{a (1 + i/\tilde{\tau}_d) (2 - 3a) + a (1 - i - 2a)}{1 + 2i \alpha + 1/\tilde{\tau}_d} . \quad (86b)
\]

These results are plotted in Fig. 3.

From Refs.\(\text{24, 47}\) and\(\text{48}\) we know that the linear dispersion of the real part of \(\omega(k)\) (Fig 3 upper set of curves) is proportional to the centroid of the charge density\(\text{29}\). It vanishes for a pure surface charge and increases with the ratio of volume to surface charge. From Eq. (80), this case corresponds to small \(\alpha\), consistent with the Figure. The damping (lower curves) is always regular (no amplification), and the maximum of its coefficient linear in \(k\) is achieved at \(\alpha = 1\) when we consider the limit \(\omega_s \tau_d \to \infty\) in Eq. (86a). This damping may be attributed to the charge trapping in the surface layer.

These results are consistent with Ref.\(\text{44}\) and obtained within a simpler calculation. One difference is that the model considered here cannot give a negative linear dispersion. This is related to the approximation that \(\rho_s\) represents a true surface charge and that one takes the position \(z = 0\) as a reference for the charge centroid.

We conclude that this hydrodynamic model of a composite charge distribution incorporates all elements of Deng’s...
The extended-medium approach: the surface breaks translation invariance, the nonzero surface current describes non-specular surface scattering (similar to Fuchs’ $p$-parameter), there is a surface plasmon mode with a well-defined local limit, the charge trapping at the surface reflects the “capacitive effects” highlighted in Ref. [4]—and still this model predicts a surface plasmon which is damped rather than unstable. The differences with respect to the dispersion found in the semiclassical approach (Boltzmann equation, Sec. [II]) can be attributed to the different dynamics involved in surface scattering: the Zaremba prescription in the semiclassical model gives an “instantaneous description” of non-specularly scattered charges, while here, the parameter $\tau_d$ plays the role of a mean dwell time.

V. EXTENDED-MEDIUM APPROACH

In this section, we review another macroscopic approach to the electromagnetic response of surfaces that incorporates surface roughness and has been introduced by García-Moliner and Flores (GF) [20]. We shall dub it the “pseudo-model” in the following. Its application in Ref. [19] deals with almost the same problem as Deng [4]. In particular, it is independent of the particular choice of the electron dynamics (bulk dielectric function). The construction is based on the fields rather than the charge or current density. Another difference is the restriction to a vanishing surface current (no physical surface charge).

We review the approach, discuss the nature of the corresponding surface plasmons and then propose connections to the specular reflection model and the dielectric approximation.

A. Physical half-spaces and fictitious stimuli

To describe the response of a metallic half-space, GF distinguish between two classes of paths that a charge can take in the medium to reach a given point [20]. As discussed in the Boltzmann theory (Sec. [II]), one class describes “direct propagation” and is only determined by the metal’s bulk properties, in particular it is translationally invariant. Apart from that, there are also paths that touch the surface and are scattered there. To model this, both the metal and the vacuum half-spaces are augmented by the other half-space. Of course, the new half-space is fictitious in nature. The extended media will be called ‘pseudo-media’. Now, to simulate surface effects, the actual perturbation at some point $\mathbf{x}' = (x, y, z > 0)$ will be mimicked by its mirror image at $\mathbf{x}'' = (x, y, -z)$. From there, a charge is assumed to propagate through the pseudo-medium towards $\mathbf{x}$ without further perturbation by the surface. Say we are concerned with the current $\mathbf{J}$ within the metal. Then, the constitutive relation is given by

$$\mathbf{J}(z) = \int_{z' > 0} dz' \left[ \sigma(\mathbf{x} - \mathbf{x}')\mathbf{E}(\mathbf{x}') + \sigma(\mathbf{x} - \mathbf{x}'')\mathbf{E}(\mathbf{x}'') \right].$$

(87)

Note that this formulation does not yet fix the value of the surface current. This requires first the determination of the field $\mathbf{E}$ in the entire pseudo-medium. If this field depends linearly on the physical field in the metal half-space, the second term in Eq. (87) can be identified with the symmetry-breaking conductivity $\sigma_s(z, z')$ introduced by Deng [Eq. (16)].

The extended-medium model is completed in three steps. First of all, GF devise a set of “fictitious stimuli”. These are currents and charges which are placed outside the respective real medium. They generate the field in the whole pseudometal. (Note that the approach is applied with electric and magnetic fields and allowing for externally incident fields.) If the field $\mathbf{E}(\mathbf{x}'')$ is constructed by a mirror symmetry, its normal component shows an (unphysical) jump at the surface which has to be compensated by introducing a fictitious surface charge density. (Note that in the local limit of the surface plasmon problem considered so far, this situation occurs with a real surface charge.) GF work, however, with the Fuchs $p$-parameter for non-specular scattering and use the condition

$$\phi^M(-z) = p\phi^M(z).$$

(88)

for the electric potential $\phi^M$ in the metallic pseudo-medium. The non-definite left-right symmetry of this potential (neither even nor odd) requires additional fictitious stimuli that GF take as a magnetic surface current and an electric surface dipole. For problems with an externally applied field, also volume charges are allowed for that can be understood as generating the external field. For the vacuum pseudo-medium, only a charge sheet is required.

The last condition fixes the surface current. Since GF try to avoid what they call an “unphysical charge accumulation”, they set the surface current to zero [19]

$$J_L(0^+) = 0.$$  

(89)

Only antisymmetric stimuli contribute to the surface current so that this condition fixes a relation between the magnetic surface current and the electric surface dipole. The latter can be understood as a particular choice of Zaremba’s correction to the Fuchs boundary condition, that yielded the function $(1 - p)A$ of Eqs. [32, 33].

B. Complex plasmon dispersion relation

To calculate the surface plasmon dispersion, GF consider a collisionless hydrodynamic model with longitudinal and transverse dielectric functions [see Eq. (72)]

$$\epsilon_\mu(K, \omega) = 1 - \frac{\omega_p^2}{\omega^2 - \beta_\mu^2 K^2} \quad \text{with} \quad \mu = L, T$$

(90)
Here, \( \beta_L \) and \( \beta_T \) are the corresponding sound velocities. Their values can be fixed by expanding Eq. (90) to second order in \( K\beta_\mu/\omega \) and comparing to the same expansion of the Lindhard dielectric functions:

\[
\beta_L^2 = \frac{3}{5} v_F^2 \quad \text{and} \quad \beta_T^2 = \frac{1}{3} v_F^2 .
\] (91)

The fields generated by the fictitious stimuli in the two pseudo-media are then constrained by the continuity relations of Maxwell’s equations, as well as Eqs. (88) (89). This determines all stimuli and yields, after expansion for small \( k\beta_\mu \ll \omega_L \), the dispersion relation

\[
\omega(k) = \omega_L + \frac{k\beta_L}{2} - i\frac{1}{4} k\beta_T \tag{92}
\]

The surface plasmon damping that appears here is attributed by GF to the mixing of surface and bulk modes\(^\text{28}\) somewhat similar to the coupling between surface and bulk charges in Sec. IV D. It is proportional to the diffusely scattering probability \(1 - p\). A damping arising from the bulk metal (for example due to Landau processes) would require a modification of the dielectric functions\(^\text{29}\).

A comparison to other results for the surface plasmon dispersion is shown in Table I. If we specialize the case of pure specular scattering at the surface (specular reflection model, \( p = 1 \)), GF find that all fictitious stimuli except the charge sheet vanish (as Deng also mentions in Appendix B of Ref. 4). The extended-medium construction of the fields in that case reduces to the derivation of Ritchie and Marusak\(^\text{28}\). It is worth recalling that the specular approximation can be made for any choice of bulk dielectric function, in contrast to the construction of Sec. 4.3 in Ref. 4.

C. The dielectric approximation

Heinrichs has introduced the so-called dielectric approximation (DA\(^\text{30}\)) to compute the surface plasmon dispersion. Using a sharp surface model, he introduces the constitutive relation for the displacement field

\[
z > 0 : \quad D(z) = \int_{z' > 0} dz' \epsilon(z - z', k, \omega) E(z') , \tag{93}
\]

where \( \epsilon \) is the bulk dielectric function. The presence of the surface is only taken into account by cutting the integral off at \( z' = 0 \). Heinrichs acknowledges the formal analogy of this constitutive relation to Reuter and Sondheimer’s treatment of the anomalous skin effect\(^\text{29}\) for diffuse reflection \( (p = 0) \), in particular in view of Eq. (88). This is an identification that he objects to, however, putting forward the different physical situations in the surface plasmon problem and the anomalous skin effect.

The dielectric approximation does have a problem with charge conservation, however, which has also been noted in Ref.\(^\text{36}\) for example. Indeed, if we compute the surface current from Eq. (93), we get in general a nonzero result (except for specific choices of the medium field \( E(z') \)), so that one should deal with a surface charge [Eq. (58), see also Eq. (40) of Ref.\(^\text{28}\)]. This is the reason why Heinrichs’ long-wave dispersion relation (see Table I) shows a different slope, if we compare to Eq. (92) of the GF pseudomodel. Heinrichs’ result hence also differs from Ritchie and Marusak’s specular reflection model (which by symmetry prevents a surface charge, see Eq. (66) and Eq. (26) of Ref.\(^\text{28}\)).

VI. CONCLUSION

We have tried in this paper to provide a transparent review of the historical work on the plasmon dispersion relation at a (sharp) metallic surface, in order to put the recent series of papers by Deng\(^\text{14}\) into perspective. Deng advocates a different viewpoint on the problem which is centered on the dynamics of charges and currents rather than the electromagnetic field. The behavior of electrons at the surface and in the sub-surface region plays a central role, both for the real part (linear dispersion) and the imaginary part (damping vs. instability) of the surface plasmon frequency. We have emphasized how to implement the conservation of charges in these processes, a basic task that does not seem to be fully addressed in Deng’s work. Already his starting point, the equation of continuity, contains an unphysical loss term attributed to “charge relaxation”. It turns out that a careful solution of the surface plasmon problem does not show any instability. There is a small fraction of electrons that “flow uphill” the electric potential at a diffusely scattering surface and give energy to the electromagnetic field. Their contribution is overwhelmed by other loss channels, however. Deng’s alternative reasoning based on energy conservation is shown to be technically flawed because the claimed contribution from the surface current (the electric current density extrapolated to the inner metallic surface) actually does not contribute. Throughout the calculations, we have tried to stay close to Deng’s approach. Another technical issue that we found is the nontrivial convergence of cosine-transformed fields at large momentum. It cannot be excluded that despite claims to the contrary, Deng’s results actually depend on the cutoff momentum.

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TABLE II. Comparison between surface plasmon dispersion relations for different models: local dielectric (LD), specular reflection (SR), dielectric approximation (DA), and the pseudo-model (GF). Approaches that may be used with any bulk dielectric function are evaluated using the hydrodynamic response \( [\omega] \). L/T – longitudinal/transverse fields with characteristic sound speeds \( \beta_{L,T} \); \( \omega_s \) – surface plasmon resonance in the long-wavelength limit; \( p \in [0,1] \) – fraction of specularly scattered electrons (Fuchs parameter).

| Model                  | LD | SR | DA | GF |
|------------------------|----|----|----|----|
| Symmetry breaking?     | no | yes| no | for \( p \neq 0 \) |
| L/T                    | L = T | L | L | L + T |
| Surface scattering     | not resolved | specular | diffuse | partially diffuse |
| Dispersion relation \( \omega(k) \) | \( \omega_s \) | \( \omega_s + \frac{1}{2} k \beta L \) | \( \omega_s + \frac{1}{2} k \left( \frac{1}{2} \beta L - i \beta T \right) \) |

**Appendix A: Checking the results of Deng**

1. Electric potential

Deng\(^{[3]}\) uses the cosine transform \( \{ \phi \} \) to represent the charge density. The resulting equations for the electric field are Eq. (9) in Ref.\(^{[3]}\) and Eqs. (17, 18) in Ref.\(^{[4]}\) (adapted to a vacuum/metal interface):

\[
E_x(z) = -i \int_0^\infty dq \frac{4k \rho_q}{K^2} (2 \cos qz - e^{-kz}) \tag{A1}
\]

and

\[
E_z(z) = \int_0^\infty dq \frac{4k \rho_q}{K^2} (2q \sin qz - k e^{-kz}) \tag{A2}
\]

with \( K^2 = k^2 + q^2 \). Deng evaluates the two terms in the parentheses under the integrals separately when solving the Boltzmann equation. We want to illustrate here that they are closely related. The key approximation is to assume that the charge density is well localized on the scale \( 1/k \) on which the potential varies ("charge sheet"). It is equivalent to replace \( \rho_q \to \rho_s \) and to pull it out of the integrals. As mentioned in the main text, this approximation is also used by Deng, it is only the organization of the calculation that differs.

The first term in Eq. \( (A1) \) is

\[
-i \int_0^\infty dq \frac{4k \rho_q}{K^2} 2 \cos qz = -i \int_0^\infty dq \frac{4k \rho_q}{q^2 + k^2} \cos qz, \tag{A3}
\]

using an even extension of \( \rho_q \) to \( q < 0 \) [see Ref.\(^{[3]}\) after Eq. (31)]. We apply the charge-sheet approximation and write the remaining integral as the real part of a contour integral

\[
-4ik \rho_s \text{Re} \int_{-\infty}^{\infty} dq \frac{e^{iqz}}{q^2 + k^2} = -4ik \rho_s \text{Re} \int_{-\infty}^{\infty} dq \frac{e^{-kz}}{2ik} = -4\pi i \rho_s e^{-kz}. \tag{A4}
\]

Assuming \( z > 0 \), the integration contour has been closed in the upper half-plane, picking the residue at \( q = ik \).

The other term in Eq. \( (A1) \) becomes an elementary integral for \( \rho_q \to \rho_s \), but could also be handled in the same way:

\[
i \rho_s \int_0^\infty dq \frac{4k}{q^2 + k^2} e^{-kz} = 2\pi i \rho_s e^{-kz}. \tag{A5}
\]

This cancels half of the cosine term in Eq. \( (A4) \), giving

\[
E_x(z) = -2\pi i \rho_s e^{-kz}. \tag{A6}
\]

This agrees with Eq. \( (37) \) for \( z > d \to 0 \).

For the evaluation of \( E_z \), one could re-use the previous result because it gives, up to a factor \(-ik\), the electric potential:

\[
z > 0 : \quad \phi(z) = \frac{2\pi \rho_s}{k} e^{-kz} \quad \tag{A7}
\]

in agreement with \( \{36\} \). Repeating the calculation with contour integrals is instructive, however, because it illustrates how the exponential \( e^{iqz} \) regularizes the integral in the UV:

\[
4\rho_s \int_{-\infty}^{\infty} dq \frac{2q \sin qz}{q^2 + k^2} = 4\rho_s \text{Im} \int_{-\infty}^{\infty} dq \frac{q e^{iqz}}{q^2 + k^2}. \tag{A8}
\]

Closing the contour for \( z > 0 \) in the upper half-plane and picking the pole at \( q = ik \), one gets

\[
4\rho_s \int_0^\infty dq \frac{2q \sin qz}{q^2 + k^2} = 4\pi \rho_s e^{-kz}. \tag{A9}
\]

This result illustrates the fallacies of taking the limit \( z \to 0 \) too early (under the integral) because of the poor UV convergence (see the discussion in Sec. IV C 2 for examples from Deng’s papers). One half of the expression \( (A9) \) is subtracted by the second term in Eq. \( (A2) \) so that we finally have

\[
E_z(z) = 2\pi \rho_s e^{-kz}. \tag{A10}
\]

in agreement with the potential \( (A7) \).

2. Distribution function

We re-calculate here the distribution function of the semi-classical model, using the formulas of Deng’s papers. The idea is similar to the preceding Appendix: by evaluating the \( q \)-integrations first, we get explicit results that avoid convergence problems at large \( q \). We use the notation \( \phi_{b,s} \) of Deng [Eqs. (19–22) in Ref.\(^{[3]}\) and Eqs. (32–35) in Ref.\(^{[4]}\) because we are going to see that the splitting into "bulk" and "surface" is
not unique. The function $F_+ (F_-)$ is even (odd) in the product variable $q v_z$, respectively. We evaluate the $dq$-integral in the charge-sheet approximation $\rho_q \rightarrow \rho_s$. The simplest case is

$$4 \rho_s \int_0^{\infty} dq \left( -F_0 \right) \frac{e^{-k z}}{q^2 + k^2} = -2 \pi \rho_s F_0 e^{-k z}.$$  

(A15)

The other integral in $g_0$ is extended to the entire real axis in the form

$$4 \rho_s \int_{-\infty}^{\infty} dq \left( F_+ e^{i q z} + F_- e^{-i q z} \right) \frac{1}{q^2 + k^2} = 4 \rho_s \int_{-\infty}^{\infty} dq \frac{e^{i q z}}{q^2 + k^2} \frac{K \cdot v}{\omega - K \cdot v}.$$  

(A16)

We look for poles in the upper half-plane and find $q = ik$ and $q v_z = \omega - k v_z = iv_z y$, provided $v_z > 0$ (assuming $\mathrm{Im} \, \omega > 0$). Working out the residues, Eq. (A16) turns into

$$= 4 \pi \rho_s \frac{e^{-k z}}{k} F_0 (k, \omega, v) - 4 \pi \rho_s \Theta(v_z) \left( \frac{2i \omega}{v_z (k^2 - \eta^2)} \right)^2 e^{-k z}.$$  

(A17)

Added to Eq. (A15), the first term changes the sign of the latter. Note the second term here whose structure is interpreted by Deng as describing electrons that move ballistically away from the surface ($v_z > 0$). In the calculation presented in Sec. II C where the $dq$-integral is performed first within the real-space representation of the electric field [see Eq. (37)],

this term is missing from the bulk distribution function $f_b$. Using the identity (A21), a similar term appears only in the surface distribution $f_s$ [see Eqs. (40)].

The first two terms of $g_s$ give

$$\rho_s \Theta(v_z) e^{-\eta z} \int_0^{\infty} dq \frac{4}{q^2 + k^2} \left( F_0 (k, \omega, v) - p F_0 (k, \omega, v_\downarrow) \right) = 2 \pi \rho_s \Theta(v_z) e^{-\eta z} \left( F_0 (k, \omega, v) - p F_0 (k, \omega, v_\downarrow) \right) (A18)$$

Finally, we evaluate the integral

$$-2 (1 - p) \rho_s \Theta(v_z) e^{-\eta z} \int_0^{\infty} dq \frac{4}{q^2 + k^2} F_+ (k, \omega, v) = - (1 - p) \rho_s \Theta(v_z) e^{-\eta z} \int_{-\infty}^{\infty} dq \frac{4}{q^2 + k^2} F_+ (k, \omega, v)$$  

(A19)

and make it convergent by inserting $e^{i q d}$ with $d \downarrow 0$. We close in the upper half-plane and find residues at $q = ik$ and $q = i \eta$ from the first summand in $F_+$ only (it would be the second one if we had used $e^{-i q d}$ and closed in the lower half-plane, giving the same result). We get for Eq. (A19)

$$= 2 \pi (1 - p) \rho_s \Theta(v_z) e^{-\eta z} \left( \frac{F_0 (k, \omega, v) + F_0 (k, \omega, v_\downarrow)}{k} + \frac{2i \omega}{v_z (k^2 - \eta^2)} \right)$$

(A20)

Simple algebra gives the identity

$$\frac{2i \omega}{v_z (k^2 - \eta^2)} = \frac{F_0 (k, \omega, v) - F_0 (k, \omega, v_\downarrow)}{k}$$  

(A21)

This leaves only the term $F_0 (k, \omega, v_\downarrow)$ in Eq. (A20).

Using the identity (A21) in Eq. (A17) and adding the results (A15) (A18) (A20), we eventually find

$$g_b + g_s = - \frac{e^{-k z}}{k} F_0 (k, \omega, v) + \Theta(v_z) e^{-\eta z} \left( \frac{2i \omega}{v_z (k^2 - \eta^2)} \right)^2$$  

(A22)

It is easy to check that the two lines in Eq. (A22) are identical to the first two lines in Eq. (40). The cancellations and simplifications in this calculation are truly remarkable. To better visualize the interrelations among the terms, we display them in Table I.

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1. H.-Y. Deng, K. Wakabayashi, and C.-H. Lam, Phys. Rev. B 95, 045428 (2017), arXiv:1511.07776 and 1701.01060
2. H.-Y. Deng, Phys. Rev. B 95, 125442 (2017), arXiv:1702.01570
3. H.-Y. Deng, J. Phys. Condens. Matter 29, 455002 (2017). arXiv:1606.06239
4. H.-Y. Deng, New J. Phys. 21, 043055 (2019) arXiv:1712.06101
5. H.-Y. Deng, “Beating plasmonic losses with an intrinsic channel of gain: the cases with $a_g$ and $a_l$,” (2017), arXiv:1706.03404
6. H.-Y. Deng, “A universal macroscopic theory of dynamical re-
...sponses for bounded systems,” (2018), arXiv:1806.08308
7 H.-Y. Deng, “On the electrical conductivity of metals with a rough surface.” (2020), arXiv:2001.08659
8 R. H. Ritchie, Phys. Rev. 106, 874 (1957).
9 D. Bohm and D. Pines, Phys. Rev. 82, 625 (1951)
10 D. Pines and D. Bohm, Phys. Rev. 85, 338 (1952)
11 W. Plummer, K.-D. Tsuei, and B.-O. Kim, Nucl. Instr. Meth. Phys. Res. B 96, 448 (1995).
12 S. Raimes, Rep. Progr. Phys. 20, 1 (1957).
13 G. Mukhopadhyay and S. Lundqvist, Physica Scr. 13, 19 (1975).
14 P. Apell, Phys. Scr. 17, 535 (1978).
15 L. Xu, F. Li, Y. Liu, F. Yao, and S. Liu, Appl. Sci. 9, 861 (2019).
16 D. J. Bergman and M. I. Stockman, Phys. Rev. Lett. 90, 027402 (2003).
17 E. I. Galanitza, R. Weingold, D. A. Nedosekin, M. Sarimollaoglu, J. Nolan, W. Harrington, A. S. Kuchyanov, R. G. Parkhomenko, F. Watanabe, Z. Nima, A. S. Biris, A. I. Plekhanov, M. I. Stockman, and V. P. Zharov, Nature Commun. 8, 15528 (2017).
18 A. Y. Smuk and N. M. Lawandy, Appl. Phys. B 84, 125 (2006).
19 F. Flores and F. García Moliner, J. Phys. (France) 38, 863 (1977).
20 F. García-Moliner and F. Flores, Introduction to the Theory of Solid Surfaces (Cambridge University Press: New York, 1979).
21 D. Wagner, Z. Naturf. A 21, 634 (1966).
22 R. Ritchie and A. Marusak, Surf. Sci. 4, 234 (1966).
23 P. J. Feibelman, Phys. Rev. B 3, 220 (1971).
24 F. Flores and F. García-Moliner, Solid State Commun. 11, 1295 (1972).
25 E. Zaremba, Phys. Rev. B 9, 1277 (1974).
26 G. E. H. Reuter and E. H. Sondheimer, Proc. Roy. Soc. A 195, 336 (1948).
27 G. Barton, Rep. Progr. Phys. 42, 963 (1979).
28 J. Heinrichs, Phys. Rev. B 7, 3487 (1973).
29 N. D. Mermin, Phys. Rev. B 1, 2362 (1970).
30 G. Röpke, A. Selchow, A. Werling, and H. Reinholz, Phys. Lett. A 260, 365 (1999).
31 G. S. Atwal and N. W. Ashcroft, Phys. Rev. B 65, 115109 (2002).
32 D. Bedeaux and J. Vlieger, Optical Properties of Surfaces (World Scientific, Singapore, 2004).
33 R. H. Ritchie, Progr. Theor. Phys. 29, 607 (1963).
34 J. M. Keller, R. Fuchs, and K. L. Kliuwer, Phys. Rev. B 12, 2012 (1975).
35 C. Cercignani and M. Lampis, Transport Theory Stat. Phys. 1, 101 (1971).
36 C. A. Mead, Phys. Rev. B 15, 519 (1977).
37 J. L. Warren and R. A. Ferrell, Phys. Rev. 117, 1252 (1960).
38 K. L. Kliuwer and R. Fuchs, Phys. Rev. 172, 607 (1968).
39 In the collisionless limit, τ → ∞, causality can be enforced as before, by solving Eq. (25) with the help electron trajectories arriving from the past at a given position. A complex frequency ω = Reω + i0 with an infinitesimal positive imaginary part does not signal an instability neither, because it is just a convenient tool to ensure an adiabatic switching-on of the electric field. The amplitude of the latter is taken proportional to exp(αt) with α → +0 so that the field vanishes in the remote past.
40 K. Fuchs, Proc. Cambr. Phil. Soc. 34, 100 (1938).
41 A. Liebsch, Electronic Excitations at Metal Surfaces, Physics of Solids and Liquids (Springer, New York, 1997).
42 The physical fields are taken as the real part of the complex ones. This explains the additional factor 1/2 in Eq. (45).
43 The integrals runs over the entire charge density which may be thought as being placed in a region 0 < z < d, see discussion after Eq. (55). Deng defines the energy and power densities by extracting a factor 1/2, see Eq. (3) in Ref. [3].
44 B. Horovitz and C. Henkel, Europhys. Lett. 97, 57010 (2012), in that paper, the direction of the z-axis is flipped, explaining the different sign in Eq. (79).
45 A. H. Nayfeh, Introduction to Perturbation Techniques (Wiley, New York, 1981).
46 This is not quite correct. Recall that the boundary condition Jz(0) = 0 has also been used in the Boltzmann solution of Sec. [11] using a Fuchs parameter 0 < p < 1.
47 J. Harris and A. Griffin, Phys. Lett. A 34, 51 (1971).
48 P. J. Feibelman, Phys. Rev. B 40, 2752 (1989).
49 The centroid is defined as ∫dz zρ(z); the origin being chosen such that the difference between the equilibrium charge ρ0(z) and a step function placed at z = 0 integrates to zero.
50 An obvious typo appears in Eq. (32) of Ref. [3] where the expression “2qF − sin(qz)” should be understood as 2iF_− sin(qz). In Eqs. (6, 7) and Eqs. (A6–A8) of Ref. [11]and in Eq. (12) of Ref. [2] a slightly different expression is given for the terms −2(1 − p)F_+ and −pF_0(k, ω, v_−) in Eq. (A12). In Ref. [2] we have taken the thick-film limit d → ∞. These may be typos.