Halevi’s extension of the Euler-Drude model for plasmonic systems

Gino Wegner,¹,² Dan-Nha Huynh,¹ N. Asger Mortensen,³,⁴ Francesco Intravaia,¹ and Kurt Busch¹,⁵

¹Humboldt-Universität zu Berlin, Institut für Physik, AG Theoretische Optik & Photonik, 12489 Berlin, Germany
²Friedrich-Schiller-University Jena, Institute of Condensed Matter Theory and Optics Max-Wien-Platz 1, 07743 Jena, Germany
³Center for Nano Optics, University of Southern Denmark, Campusvej 55, DK-5230 Odense M, Denmark
⁴Danish Institute for Advanced Study, University of Southern Denmark, Campusvej 55, DK-5230 Odense M, Denmark
⁵Max-Born-Institut, 12489 Berlin, Germany

(Dated: September 13, 2022)

The nonlocal response of plasmonic materials and nanostructures is usually described within a hydrodynamic approach which is based on the Euler-Drude equation. In this work, we reconsider this approach within linear response theory and employ Halevi’s extension to this standard hydrodynamic model. After discussing the impact of this improved model, which we term the Halevi model, on the propagation of longitudinal volume modes, we accordingly extend the Mie-Ruppin theory. Specifically, we derive the dispersion relation of cylindrical surface plasmons. This reveals a nonlocal, collisional damping term which is related to earlier phenomenological considerations of limited-mean-free-path effects and influences both, peak width and amplitude of corresponding resonances in the extinction spectrum. In addition, we transfer the Halevi model into the time-domain thereby revealing a novel, diffusive contribution to the current which shares certain similarities with Cattaneo-type currents and analyze the resulting hybrid, diffusive-wave-like motion. Further, we discuss the relation of the Halevi model to other approaches commonly used in the literature. Finally, we demonstrate how to implement the Halevi model into the Discontinuous-Galerkin Time-Domain finite-element Maxwell solver and are able to identify an oscillatory contribution to the diffusive current. The Halevi model thus captures a number of relevant features beyond the standard hydrodynamic model. Contrary to other extensions of the standard hydrodynamic model, its use in time-domain Maxwell solvers is straightforward – especially due its affinity to a class of descriptions that allow for a clear distinction between bulk and surface response. This is of particular importance for applications in nano-plasmonics where nano-gap structures and other nano-scale features have to be modeled efficiently and accurately.

Keywords: hydrodynamic, elasticity, diffusion, plasmonics, viscoelasticity, surface electrodynamics

I. INTRODUCTION

Over the last decades, the preparation of complex metallic nanostructures has experienced tremendous progress due to advances in material quality and nanostructuring techniques. Examples are dimers of spheres [1] and cylindrical wires [2], mixed wire-sphere systems [3], and bowtie antennas. The latter are connected [4] or unconnected [5], tip-to-tip nano-triangles. All such dimers (and many more conceivable structures) feature nano-gaps that give rise to strong field enhancements that can be exploited for numerous applications which include (but are certainly not limited to) sensing via surface- and tip-enhanced Raman scattering and surface-enhanced infrared absorption spectroscopy as well as various wave mixing processes. The performance of such devices relies on the plasmonic properties of the conduction electrons at optical frequencies and they can be tailored via the choice of material as well as via size and geometry [6] of the system’s constituents as well as on their surface preparation [7,8].

The efficient modeling of plasmonic nanostructures on both, the interpretative and predictive level requires appropriate material models that are able to capture the relevant physics on the involved time- and length-scales and are amenable to a performant implementation in numerical scheme. Owing to the dispersive nature of the plasmonic material response, the latter aspect is particularly important for time-domain simulations of the Maxwell equations. One particular semiclassical model, which satisfies these requirements is often referred to as the hydrodynamic [9,12] or hydrodynamic Drude model [13,15] whose details we describe in section II. On the one hand, it captures certain quantum effects which become relevant in the nanometer regime and, on the other hand, its coarseness allows for a language of just a few degrees of freedom. Therefore, when being incorporated into a Maxwell solver, it facilitates the treatment of scattering phenomena of nano-scale features that modulate a much larger structure. In practise, this multi-scale aspect is accompanied by a moderate usage of computational resources.

An illustrative phenomenon which is well-described by the original hydrodynamic model is the size-dependent shift of plasmonic resonances for small metallic particles [16,19]. Further, through the addition of diffusive dynamics the size-dependent broadening of these resonances is qualitatively accounted for within the so-called Generalized Nonlocal Optical Response (GNOR) model [18,19]. In the GNOR model, the

* E-mail address: wegner@physik.hu-berlin.de
standard hydrodynamic current is supplemented by a diffusive current that follows Fick’s first law. Furthermore, comparing the GNOR model to Halevi’s extension of the hydrodynamic model (which is based on the Boltzmann-Mermin approach) facilitates the derivation of a quantum mechanical diffusion constant.

In this work, we will reconsider the plasmonic response of metallic structures by direct application of Halevi’s extension of the hydrodynamic model which we refer to as the Halevi model. As we go along, we also focus on various extensions to the original hydrodynamic model and elaborate on the difficulties of a one-to-one identification with the GNOR model. While the Halevi as well as the GNOR model initially describe the dynamics of electrons in the bulk, we determine further response properties near the surface of a finite scatterer. Specifically, we consider nano-scale wires as prototypical structures. In fact, nano-wires are rather popular and convenient for experimental studies as they can be fabricated with excellent quality via membrane- or template-based synthesis with diameters down to about 5 nm [20]. Possible nano-scale devices, that utilize plasmonic nano-wires include waveguides [21, 22], nano-antennas [6], and sensors [3].

Under normal illumination, analytical expressions for the electromagnetic fields in and around infinitely-extended straight plasmonic nano-wires described by the hydrodynamic Drude model have been obtained with [23] and without [16] the quasi-static approximation. Here, the incident plane wave’s electric field is polarized perpendicular to the nanowire axis [16, 23]. In particular, we aim at extending the work of Ref. [16] to the Halevi model.

The manuscript is organized as follows. Starting with Sec. II, we briefly review the basics of the hydrodynamic Drude model and elaborate on the extension derived by Halevi. Further, we place this description into the framework of the viscoelastic model [24] in order to deepen the conceptual understanding and provide a route for future extensions. In the following Sec. III, we adapt the theory developed in Ref. [16] to the Halevi model, provide a corresponding justification, and elaborate on the implications. In particular, we discuss the Halevi model’s influence on mode propagation and derive the dispersion relation of surface plasmon polaritons within the wire. The latter reveals a nonlocal damping term that is connected to an earlier phenomenological proposition on limited-mean-free-path effects and resonance broadening. In the subsequent Sec. IV we develop the time-domain formulation of the Halevi model. We explicitly show, that the Halevi extension to the hydrodynamic model introduces a novel current, that is conceptually closely related to the Cattaneo-current [25] which models classical diffusion processes with finite propagation velocity. Next, in Sec. V we briefly review the GNOR-model to allow for a detailed comparison with the Halevi model, specifically regarding the respective diffusive paradigms. We will show that, despite certain similarities, there are marked differences between the two models which prohibit a one-to-one mapping. In Sec. VI we then proceed to numerical simulations of the scattering setup of Sec. III using the time-domain formulation of the Halevi model and analyze the influence of the Halevi extension on the propagation of the electric field and the mode profiles. Finally, in Sec. VII we summarize our findings and provide an outlook for future studies.

II. JUSTIFICATION OF THE HALEVI MODEL

It appears, that Felix Bloch proposed and discussed the first treatment of electron dynamics by means of a hydrodynamic model [26] (see also Refs. [19, 27, 29] for further discussions) Back in 1933, as demonstrated in Ref. [30], such an approach presented an analytically amenable means to estimate the stopping power associated with the response of certain atoms. Since then, the model has been rederived and extended following different paradigms. Usually the connection to conservation equations is pointed out and their specific forms are related to an equation of state. The latter may be deduced from (quantum) statistics [28, 31, 33]. A special case is the derivation of equations of motion from an energy principle. Within such an approach, the equation of state follows from the choice of internal energy functional [17, 27, 30, 34, 35]. Another strategy considers already existing, semi-classical models and tries to asymptotically identify their response functions with those which are motivated by continuum theories [24, 26, 35, 30]. In the present work, we follow the latter approach.

The continuum assumption lies at the heart of hydrodynamic models for describing the conduction electrons in metals. This means that a mesoscopic perspective is adapted where the electrons form a charged fluid such that a given fluid element is (i) much larger than the actual constituents of the fluid and their mean separation and (ii) much smaller than the volume occupied by the fluid [37]. The ionic background is treated as a rigid, motionless continuum – the rigid jellium [38] – and restores overall charge neutrality. The thus introduced electron continuum may dynamically change shape and volume.

The associated dynamical quantities are the moments of a distribution function that describes the microscopic electron dynamics. Quite generally, this distribution function $f(r, p, t)$ represents the probability of finding at time $t$ a representative electron with its microscopic momentum $p$ in an infinitesimal volume element surrounding the position $r$. This effective one-particle distribution function is obtained from the distribution function of the full $N$-body-electron-system by integrating over the positions and microscopic momenta of the remaining $N - 1$ particles. Due to the Coulomb interaction between the electrons, as well as their interaction with the
The dynamics of a \(m\)-particle-distribution contains terms that couple to the \((m + 1)\)-particle distribution where \(0 < m < N\). This coupled set of \(N - 1\) equations is referred to as the Bogoliubov–Born–Green–Kirkwood–Yvon hierarchy [31]. Breaking this hierarchy then facilitates the implementation of the above-mentioned method of moments. Among the truncated descriptions of one-particle distributions, a suitable equation of motion, from which the hydrodynamic equations can be deduced is the Boltzmann equation [40, 41] with an electron-ion collision integral that may be expressed following Mermin’s recipe for a charge-conserving, single-relaxation-time correction [42] and the electron Coulomb interaction being treated by a mean-field approximation. Upon expanding the distribution function of the Boltzmann-Mermin model into the moments of the microscopic momentum another hierarchy of evolution equations is obtained where successive-order moments become coupled and the individual equations of motion represent conservation laws. A closed set of equations of motion can be obtained through further approximations that relate a given moment to lower-order moments only – specifically, this procedure is connected to additional approximations to the chosen coupling term of the distributions functions.

The standard hydrodynamic model considers the first- and second-order moments, charge- and momentum density, and the corresponding equations of motion ensure charge conservation and momentum balance. In the resulting evolution equations, the momentum density is often replaced by the charge current density. The omission of higher-order moments and the corresponding equations of motions is tied to the absence of heat conduction [32] and to a specific form of the stress tensor (a.k.a. momentum current density). The latter is given by a pressure that can solely be expressed in terms of the charge density which, e.g., may be derived from the Thomas-Fermi theory of a degenerate electron gas [30].

The conservation of the current density \(J\) (or, equivalently, momentum conservation) is given by the linearized Euler equation of classical fluid dynamics [37] and reads

\[
\partial_t J(r, t) = \epsilon_0 \omega^2 \mathbf{E}(r, t) - \beta_{TF}^2 \nabla \rho(r, t). \tag{2}
\]

Here, we would like to note, that the strength of the Thomas-Fermi-pressure term scales with the parameter \(\beta_{TF} = v_F/\sqrt{3}\) and, thus with the Fermi velocity \(v_F\). It therefore inherits constraints of the Pauli exclusion principle [31]. In fact, the parameter \(\beta_{TF}\) may be viewed as the velocity of sound in the electron continuum [43] and, accordingly, characterizes density or pressure waves [18] which may build up in the electron continuum.

The total current is defined via the center-of-mass motion which arises from the total electric field \(\mathbf{E}(r, t)\) induced by the motion of the electron density relative to the fixed jellium background and any externally applied electric fields. In this treatment, we neglect the effects of magnetic fields (see Ref. [44] for the treatment of magneto-optic effects). This classical treatment of the electric field excludes quantum-mechanical effects such as the exchange interactions. An early work, based on Bloch’s hydrodynamic model, that derives resonances of a metallic scatterer without collisions but with an approximate form of electronic exchange-interactions, has been presented by Jensen in Ref. [45].

Since Bloch’s model focussed more on the electrons within the individual atoms/ions and less on the relative position of the latter, we expect deviations to his theory when considering more realistic metallic structures. As a first correction, the Drude term \(-\gamma J\) is added to the r.h.s. of Eq. (2) yielding the Euler-Drude model. This new term describes the hinderance of electronic motion due to aperiodicities in the ionic lattice that may come from defects in the crystal lattice or from lattice vibrations (phonons). It leads to collisions quantified with a rate \(\gamma\) which diminish electron momentum [24] and is part of the first-order moment of the single-relaxation-time approximation, discussed above.

As Bloch points out, the Thomas-Fermi model has been applied to the static behaviour of the electron continuum. From Eq. (2) we, thus, expect a quasi-static description. But such a treatment fails for typical metals at optical frequencies. As a remedy, the very same equation may be used by replacing the low-frequency value \(\beta_{TF}\) with the high-frequency value \(\beta_{IF} = \sqrt{3}/v_F\) [9, 17–19, 26, 28].

Naturally, the question arises which characteristic velocity should be used, in general [26, 29]. A model within the continuum approach that interpolates between the two limits has been provided by Halevi [26]. Specifically, Halevi considers the Euler-Drude model in \((r, \omega)\)-space and introduces
a frequency-dependent characteristic velocity $\beta_H(\omega)$. Consequently, Halevi’s extension to the standard hydrodynamic model reads

$$-i\omega J(r, \omega) = \epsilon_0 \omega_p^2 E(r, \omega) - \beta_H(\omega)^2 \nabla \rho(r, \omega) - \gamma J(r, \omega).$$

(3)

The longitudinal dielectric function with wavenumber $k$ and frequency $\omega$ that is derived from Eqs. (1) and (3) is subsequently expanded up to second order in the parameter $k v_F / \omega$. A comparison with a similar expansion within the Boltzmann-Mermin model is then performed. Halevi’s approach, therefore, allows to isolate possible extensions of the Euler-Drude model without introducing higher orders of spatial nonlocality, i.e., avoiding contributions that, e.g., scale with the second or higher powers of the Laplacian $\nabla^2$, or $\nabla(\nabla \cdot \rho)$, or other combinations of vectorial differential operators. Further, Halevi’s approach guarantees local charge conservation due to Mermin’s relaxation time approximation [22]. However, we would like to stress that the high-frequency, collisionless limit of the Boltzmann-Mermin model, i.e., the Vlasov model [28]. itself does not introduce any exchange-correlation effects in terms of pure electron-electron interaction [36].

Upon incorporating in $\beta_H(\omega)$ the dispersion introduced by the asymptotic expansion of the Boltzmann-Mermin model and comparing with the Bloch hydrodynamic model, Halevi obtains [26]

$$\beta_H(\omega)^2 = \frac{3}{\omega + i \gamma} v_F^2.$$

(4)

The cross-over regime is characterized by the Drude collision rate $\gamma$. Indeed, Eq. (4) interpolates between the low-frequency velocity $\beta(\omega \ll \gamma) = \beta_{TF}$ in the collision-dominated limit and the high-frequency velocity $\beta(\omega \gg \gamma) = \beta_{HF}$ in the field-dominated limit [26]. We refer to the system of equations [1] and [2] in the respective limits the low- and high-frequency Euler-Drude model. For typical metals $\omega_p$ lies in the visible or ultraviolet, such that $\gamma^2 \ll \omega_p^2$ and, therefore, the high-frequency value $\beta_{HF}$ is preferred at optical frequencies $\omega \sim \omega_p$. For intermediate frequencies, $\beta_H$ exhibits dispersion which Halevi relates to a phase mismatch between pressure and density fluctuations [26].

In order to deepen the physical understanding of the transition from $\beta_{TF}$ to $\beta_{HF}$, we recall, as pointed out in Ref. [36] that the Halevi model may be viewed as the longitudinal projection of the viscoelastic model derived by Conti and Vignale. While Halevi’s viewpoint focusses more on a formal correspondence to fluid dynamics, Conti and Vignale [24] emphasize elastic contributions that are well-known from the elastic properties of solids. The latter dynamics introduces both, an elastic shear and a compressibility of the electron continuum. The latter remains constant over the frequency range $\hbar \omega \ll E_F$. In contrast, the hydrodynamic limit of the viscoelastic model is determined via the same compressibility augmented with a kinematic viscosity. The hydrodynamic bulk viscosity actually vanishes. This is similar to a hypothesis made by G.G. Stokes about the total pressure which is assumed to be independent of the temporal change of the fluid density during a uniform dilatation [46].

The range of applicability of the viscoelastic model is set by the contraints [24, 38]

$$k \ll 2k_F, \quad \omega \gg k v_F, \quad \omega, \gamma \ll E_F/h.$$  

(5)

Here, the first, third and fourth inequalities facilitate the interpretation of the underlying dynamics in terms of the Boltzmann equation with collisions, where the momentum relaxes according to Mermin’s recipe on the timescale $2 \pi / \gamma$. The second inequality implies the transition to the viscoelastic paradigm confined by the hydrodynamic and elastic limit [38].

The formal equivalence of the latter continuum limits stems from the momentum conservation equation which can be rephrased in terms of the current density according to

$$-\omega(\omega + i \gamma) J(r, \omega) = -i \omega \omega_p^2 \epsilon_0 E(r, \omega) + \left[ \frac{\beta^2 - \frac{4}{3} i \omega \eta}{} \right] \nabla (\nabla \cdot J(r, \omega)) + i \omega \eta \nabla \times \nabla \times J(r, \omega).$$

(6)

Here, in the high-frequency limit, the center-of-mass velocity in an infinitesimal volume around a given position originates from the temporal change of the displacement of an infinitesimal charge element due to compression and/or elastic shear [24].

When comparing this result with the longitudinal and transverse response of the Boltzmann-Mermin model up to the second order in the wavevector, it turns out that the velocity $\beta$ is just the Thomas-Fermi velocity and that the parameter $\eta(\omega)$ provides an interpolation between the velocity of elastic shear waves $\beta_{sh}$ and kinematic viscosity $\eta(\omega)$ according to

$$-i \omega \eta(\omega) = \beta_{sh}^2(\omega) - i \omega \eta(\omega).$$

(7)

Here, the kinematic viscosity $\eta$ and the elastic shear velocity $\beta_{sh}$ are real quantities given by

$$\beta_{sh}^2(\omega) = \frac{\omega^2}{\omega^2 + \gamma^2} \frac{v_F^2}{3},$$

$$\eta(\omega) = \frac{\gamma^2}{\omega^2 + \gamma^2} \frac{v_F^2}{5 \gamma}.$$  

(8)

At this point, we would like to note that, similar to compression, also the shear is constrained by Pauli’s exclusion principle as manifest by the appearance of the Fermi velocity in $\beta_{sh}$ and $\eta$. Apart from that, the material parameters are independent of position. Thus, within the bulk longitudinal (transverse) waves remain longitudinal (transverse). However, analogous to the theory of elasticity, at a material interface both
polarisations mix \cite{47}. This represents a key property for plasmonic nano-particles, where the evanescent waves of surface plasmons can thus couple to bulk plasmons. For instance, transverse polarized radiation impinging onto a cylinder may excite bulk plasmons \cite{16}.

Considering that longitudinal quantities exhibit a vanishing curl, we can deduce the connection between the viscoelastic parameters and the Halevi velocity by combining Eq. (6) with the continuity equation (see Eq. (16) of \cite{36}) to yield

$$\beta_H(\omega) = \beta^2_{HF} - \frac{4}{3} i \omega \tilde{\eta}(\omega). \quad (10)$$

Accordingly, the fact that the Halevi velocity $\beta_H$ is a complex quantity stems from the interpolation between the fluid-like and solid-like response which, in turn, is encoded in the asymptotic expansion of the Boltzmann-Mermin model. Conceptually, the additional dispersion (as compared to the Bloch model) originates completely in viscoelasticity \cite{36}.

More precisely, within this perspective, the low-frequency, collision-dominated limit is viewed as the hydrodynamic regime and the high-frequency, collision-less limit is identified with the elastic regime \cite{24}. Since, we have for typical metals that $\omega_p \gg \gamma$, the plasmonic response of metallic structures has to be regarded as being predominantly elastic.

Therefore, and contrary to what is sometimes stated in the literature, the velocity $\beta_{HF}$ is not entirely the result of the compressibility within the Thomas-Fermi model. Instead, $\beta_{HF}$ also features contributions from elastic shear.

Furthermore, the elastic response at short time scales ($2\pi/\omega \ll 2\pi/\gamma$) and the viscous response at longer time scales ($2\pi/\omega \gg 2\pi/\gamma$) are characteristic of highly viscous fluids \cite{28}, such as glycerin or resin \cite{48}. As proposed by Maxwell, the deformation of such fluids due to periodic, external forces induces internal shear stress, that is damped on a certain time scale $\tau$. As the period $2\pi/\omega$ of the external forces progresses from values much lower than $\tau$ to values much greater values than $\tau$, the elastic response changes from solid- to (viscous-)liquid-like characteristics \cite{48}. Since in the plasmonic case $\tau$ is roughly given by $2\pi/\gamma$, it is the rate of Drude collisions with the ionic background, that determines the extent of the solid-like response and, thus, of the continuum’s tendency to restore its equilibrium shape. In the elastic regime, the periodic deformation is so fast, that collisions cannot relax the internal stress within one period. Note, again, that the magnitude of the shear is determined by the Fermi velocity [Eqs. (1) and (9)], so that the internal stress is not only given by collisions but also tied to the Pauli principle. Since both paradigms are based on the continuum approximation we stick to the more general term *electron continuum*.

Interestingly, not only the longitudinal, but also the transverse dielectric function derived from Eq. (6) is formally equivalent to a hydrodynamic dielectric function with a characteristic, squared transverse velocity $\beta^2_T(\omega) = -i \omega \tilde{\eta}(\omega)$. In the elastic regime, we have $\beta_T = \beta_{sh}(\omega \gg \gamma) = v_F/\sqrt{5}$. We emphasize this fact, since, for instance, Garcia-Moliner and Flores \cite{49} have applied the hydrodynamic dielectric function for the longitudinal and the transverse response of a metallic half-space in the limit $\omega \sim \omega_p \gg \gamma$, with $\beta_L = \beta_{HF}$ and $\beta_T = \beta_{sh}(\omega \gg \gamma)$. Hence, the transverse and longitudinal dielectric function turn out to be high-frequency limits of the viscoelastic model – corrections towards intermediate frequencies are thus at hand.

Having completed the discussion of characterics of the Halevi model and obtaining a deeper understanding due to the viscoelastic interpretation, we now proceed, in the next section, to a first investigation of the model’s impact on light-matter interaction within an analytical test case.

### III. EXTENDED RUPPIN-MIE THEORY FOR A NONLOCAL, CYLINDRICAL SCATTERING PROBLEM

Analytical expressions for quantities that describe the light-matter interaction at metallic nanostructures are often tied to highly symmetric scatterers. An infinitely-extended circular cylinder with radius $a$, homogeneous along its rotational axis and placed within vacuum represents such an archetypal scatterer \cite{50}. It further represents a very good approximation to realistic cylinders with high aspect ratios \cite{2}.

Analytical formulas for the fields, given local and dispersive response within the metal, have been derived based on Mie-theory \cite{50} and have later been extended by Ruppin to allow for linear hydrodynamic response \cite{16}. In the latter work, the incident wave is injected perpendicular to the cylindrical axis and the electric field is polarized also orthogonal to this axis, thus leading to an effectively two-dimensional problem (cf. Fig. [1]). Specifically, this setup allows for bulk modes in the metal \cite{16}, as we have discussed in Sec. [II].

Following a discussion of Melnyk and Harrison \cite{51}, Ruppin demands the absence of an infinitesimally thick surface charge density, which ultimately leads to a continuity of the radial component of the electric field. Since this is tied to the absence of the normal current right below the surface, similar to the hardwall boundary condition \cite{17, 19, 35}, and the tangential current may, in principle, be finite in this region, this auxiliary boundary condition (ABC) is also known as slip boundary condition \cite{52}. Such a tangential surface current must not be mistaken with an infinitesimally thick surface current sheet, which would lead to a discontinuous, tangential magnetic field at the surface. In addition to the above, the continuity of the tangential electric field and normal magnetic flux field is enforced.

The transverse response is encoded by the Drude dielectric function given by

$$\epsilon_{\text{Drude}}(\omega) = 1 - \frac{\omega_p^2}{\omega(\omega + i\gamma)}, \quad (11)$$
FIG. 1. Sketch of the infinitely-extended cylindrical scatterer excited normally with a plane wave propagating along the $x$-direction and polarized normal to the rotational axis ($\hat{z}$). The electromagnetic problem effectively reduces to two dimensions.

which gives for the wavenumber of the internal transverse mode

$$k_L^2 = k_0^2 \frac{\epsilon_D(\omega)}{\epsilon_{BG}}.$$  \hspace{1cm} (12)

Here, $k_0 = \epsilon_{BG}\omega/c$ is the wave number of the incident wave with background dielectric function $\epsilon_{BG}$. In the following, we use vacuum as the background material and thus set $\epsilon_{BG} = 1$.

In addition, the longitudinal response is given by the (high-frequency) Euler-Drude dielectric function

$$\epsilon_L(k, \omega) = 1 - \frac{\omega_p^2}{\omega(\omega + \gamma)} - \beta_{HF}^2 k^2.$$  \hspace{1cm} (13)

The corresponding wavenumber of the internal longitudinal mode derives from the implicit equation

$$\epsilon_L(k_L, \omega) = 0.$$  \hspace{1cm} (14)

Completely analogous to standard Mie-theory, the incident, scattered and internal transverse portions of the electric field are expanded into solenoidal vector cylindrical harmonics. However, in the internal region an irrotational vector cylindrical harmonic is added to account for longitudinal waves. Solving the above-discussed set of boundary conditions for this Ansatz, Ruppin derives the multipole expansion coefficients of the scattered field as

$$s_n = \left[ \frac{c_n + D_n(k_T a)}{c_n + D_n(k_T a)} J_n(k_0 a) - \sqrt{\epsilon_T(\omega)J_n'(k_0 a)} \right] \left[ \frac{c_n + D_n(k_T a)}{c_n + D_n(k_T a)} H_n(k_0 a) - \sqrt{\epsilon_T(\omega)H_n'(k_0 a)} \right],$$  \hspace{1cm} (15)

where $D_n(x) = J'_n(x)/J_n(x)$.

Here, $J_n(x)$ and $H_n(x)$ are, respectively, the Bessel and outgoing Hankel functions of order $n$.

The nonlocal correction term $c_n$ is given by

$$c_n = \frac{n^2}{k_L a} \left[ D_n(k_L a) \right]^{-1} \frac{\epsilon_T(\omega) - 1}{k_0 a \sqrt{\epsilon_T(\omega)}}.$$  \hspace{1cm} (16)

This nonlocal correction originates from the presence of the longitudinal field and vanishes in the limit $\beta \to 0$, where the longitudinal nonlocality is not resolved. Here $n$ counts the multipole order. For the monopole, $n = 0$, we recall from Ref. 50 that

$$s_0 \propto (k_0 a)^4 \left( \sqrt{\epsilon_T(\omega)} - 1 \right),$$  \hspace{1cm} (17)

in a local material description with vacuum as the host material, and provided that optical frequencies and radii within the range of $a \lesssim 10$ nm are considered. Therefore, for a spatially local response, we do not expect a significant monopole contribution to the field. Further, the nonlocal coefficient $c_0$ vanishes and the same can be shown for the monopole coefficient of the internal, longitudinal field. In contrast, significant contributions are expected from the dipole ($n = 1$) as well as the quadrupole ($n = 2$), for which the nonlocal coefficients $c_1$ and $c_2$ cannot be neglected.

From the discussion in Sec. II we deduce that the Halevi model does not introduce any higher-order spatial derivatives as compared to the high-frequency Euler-Drude model. Otherwise, additional (spatial) boundary conditions would have to be introduced. We thus use the wavenumbers of the Halevi model within Eqs. (15) and (16).

As Ruppin points out, the longitudinal wavenumber describes the propagation of the longitudinal modes. Therefore, we will now determine its features within the Halevi model and deduce important differences compared to the high-frequency Euler(-Drude) model. To do so, we start with the remark, that Eqs. (4), (13) and (14) actually only provide the square of the wavenumber given by

$$k_L^2 = \frac{\omega(\omega + \gamma) - \omega_p^2}{\beta_H(\omega)^2},$$  \hspace{1cm} (18)

where we substituted $\beta_{HF} \to \beta_H(\omega)$. This disguises some of the wavenumber’s characteristics in the complex plane. We fill in the blanks by sticking to a passive system, thus enforcing a non-negative imaginary part. By choosing the principal branch of the complex square root, the real part must remain positive.

In Fig. 2 we depict the frequency dependence of the longitudinal wavevector for different material models of silver (see Tab. I for material parameters). From this, we can infer several features which distinguish the Halevi from the Euler(-Drude) model. Specifically, below the volume plasma frequency a kink occurs (cf. right inset of Fig. 2h). The actual frequency is derived as follows: To first order in $\gamma/\omega_p$, the real part of the squared wavenumber changes sign at $\omega_p$ - being negative for frequency values below $\omega_p$. At the same time, the imaginary part changes sign at the real frequency

$$\omega_0 = \omega_p \sqrt{\frac{4}{13} - \frac{5}{13} \frac{\gamma^2}{\omega_p^2}},$$  \hspace{1cm} (19)

as long as $0 < \gamma < \sqrt{4/5} \omega_p \approx 0.894 \omega_p$, where $\omega_0 \approx 0.555 \omega_p$ for typical metals (cf. Fig. 2f). Thus, the real part
of the longitudinal wavenumber, given by

$$\text{Re} \ k_L(\omega) = \sqrt{\frac{1}{2} \left( \text{Re} k_L^2(\omega) + \sqrt{[\text{Re} k_L^2(\omega)]^2 + [\text{Im} k_L^2(\omega)]^2} \right)},$$

should exhibit a root at $\omega_0$.

At lower frequencies, we observe a local maximum (see left inset of Fig. 2). Different real parts in the different models should, in principle, lead to spatial phase differences for a given frequency. Interestingly, we observe in Fig. 2, that the frequency of this local maximum initially increases with $\gamma$ until a value of roughly $0.2\omega_p$ is reached. Beyond this value, the frequency of the local maximum drops and vanishes at $\gamma = \sqrt{4/5}\omega_p$ along with the corresponding amplitude.

Regarding the imaginary part of the longitudinal wavenumber, we notice from Fig. 2 b, that a peak occurs at low frequencies. This is due to the small velocity of the low-frequency density waves, which, in the Halevi model, reaches the Thomas-Fermi value of $\beta_{TF} = v_F/\sqrt{3} < \beta_{HF}$. Increasing the ratio $\gamma/\omega_p$ leads to a broadening of the peak and for the plasma frequency of bulk silver, this peak extends into the infrared frequency range. Following Ruppin, this is tantamount to a faster decay of the longitudinal modes in this frequency range. Still looking at the silver curves, we deduce from the inset of Fig. 2 a, that for $\omega > \omega_p$, the decay is approximately given by the Drude damping due to the ionic background.

Now that we have determined a number of bulk features introduced by the Halevi model, we proceed to considering the surface resonances of nonlocal cylinders. In particular, we focus on the quasi-static approximation, i.e., where the incident wavelength and the skin depth are much larger than the cylinder radius. In this case, the phase of the plane wave is approximately constant across the cylinder’s cross section. Since we expect that the resonances are at typical wavelengths, i.e., of the order of a few 100nm, the quasi-static approximation is roughly valid for radii below a few 10nm. As derived in the Appendix the leading order contribution of the nonlocality manifests itself in the dispersion relation according to

$$\omega_n \approx \omega_{sp} - \frac{i\gamma}{2} + \frac{\beta_{HF} n}{\alpha} \left( 1 - \frac{2i\gamma}{9\omega_{sp}} \right),$$

for $n > 0$. The first three terms on the r.h.s. recover the result of the linearized Euler-Drude model regarding the nonlocal blueshift.

The imaginary part of the surface resonance frequency for...
a multipole of order \( n \) is given by

\[
\text{Im} \omega_n = -\frac{\gamma}{2} \left( 1 + \frac{2 \beta_{HF} n}{9 \omega_{sp} a} \right). \tag{22}
\]

It is proportional to the collision rate and negative, which conforms with our passivity requirement. In contrast to the well-known result for the high-frequency Euler-Drude model, the damping incorporates a nonlocal contribution. Such a feature has already been observed by Halevi in the dispersion relation of the volume plasmon \cite{26}. Halevi refers to this additional damping channel as 'collision-modified Landau damping'. While, the plasmonic frequencies lie deep within the elastic regime, the novel damping term vanishes for \( \gamma/\omega_{sp} = 0 \). Hence, the viscous damping, also proposed in Refs. \cite{56} and \cite{57} which contributes in the intermediate, viscoelastic regime, is essential for this damping channel to be present.

At this point, we wish to emphasize, that the calculations in the Appendix are based on the limiting case \( \nu_F n/\omega_{sp} a \ll 1 \) so that the nonlocal damping contribution is small compared to the Drude damping. In Tab. \ref{tab:1} we provide the relevant parameters for a number of metals that are used in plasmonics. From this, we infer that the nonlocal damping of the dipole for cylinder radius \( a = 1 \)\text{nm} only contributes with about a percent of the local Drude damping, with Copper having the largest value, so that the aforementioned assumption is well justified.

Analyzing the dissipation for frequencies close to \( \omega_{sp} \), Eq. \eqref{eq:22} appears to suggest a description of the system in terms of a Drude dielectric function where the Drude damping rate is amended with a term that scales with the inverse of the cylinder radius. Such an approach would allow to avoid the intricacies that come along with the nonlocal hydrodynamic description matched by a slip boundary condition. Nonetheless, while such an approach would, indeed, yield the same expression for the imaginary part of the dispersion relation, we abstain from such an identification as this would blur the clear conceptual distinction between bulk and surface dynamics established by the bulk dielectric function and the set of surface boundary conditions, respectively. Nevertheless, we would like to note that such an interpretation via a size-dependent damping rate is reminiscent of a proposition made in Refs. \cite{58} and \cite{59} which phenomenologically describes the measured extinction of small spherical silver nano-particles immersed in different host media by limited-mean-free path effects. In these works, the thus corrected damping rate was formally written as

\[
\gamma(a) = \gamma + A \frac{\nu_F}{a}, \tag{23}
\]

and utilized within a bulk Drude dielectric function. The value of the corresponding linewidth parameter \( A \) is then determined for different embedding scenarios by a fit to measured extinction data and the difference in values is motivated by a discussion of possible surface damping mechanisms. Within the Halevi model and when we assume that the cylinder is embedded in vacuum, we derive the linewidth parameter as

\[
A = \frac{n}{9} \frac{\beta_{HF}}{\nu_F} \frac{\gamma}{\omega_{sp}}. \tag{24}
\]

We note, that its structure is not only determined by the actual geometry and the choice of boundary condition but also
depends strongly on the actual bulk response. Within the high-frequency Euler-Drude model equipped with the same boundary condition, this novel effect would be completely absent. While in Refs. [58] and [59] the phenomenological surface damping has been used to describe the dipole mode for spherical scatterers only, we would like to point out that our analysis implies that a similar structure is to be expected for higher multipole modes, too. For results of the impact of leading-order nonlocality on higher-order surface (and bulk) plasmons excited in metal spheres, we refer the reader to Ref. [61]. Further, other scatterer geometries (for which numerical simulation might be required) will exhibit different values of the linewidth parameters, even for dipoles. Specifically, for silver spheres in vacuum a value of $A_{Ag} = 0.025$ for the dipole resonance has been determined experimentally [59]. A glance at Tab. [II] reveals, that the $A$-value for the dipolar resonance of silver nano-wires as derived from the Halevi model in conjunction with the slip boundary condition is about two orders of magnitude smaller. Owing to a larger value of $\gamma/\omega_{sp}$, the corresponding linewidth parameter for zinc nano-wires is about one order of magnitude larger. To complete our analysis of the nonlocal damping term, we briefly mention, that the application of the so-called GNOR model to metallic spheres with the very same ABC has also led to a motivation of the broken limited mean-free-path effects [15][18][62]. In Sec. [V] we will provide a comparison of the Halevi and this alternative model.

We conclude this section by stating that the additional damping should lead to a broadening of the quasi-static resonance peaks while, at the same time, decreasing the amplitude. This should be visible in the extinction efficiency defined as

$$Q_{ext} = -\frac{1}{2\alpha_{pl}|\omega|^2} \sum_{n=-\infty}^{\infty} \text{Re} s_n.$$  

(25)

In Fig. [3] we depict the extinction efficiency as a function of both, radius and frequency, for the Drude (panel a) and Halevi model (panel b). The results for the high-frequency Euler-Drude model are not shown because they are very similar to those of the Halevi model. We observe, that all models display a strong dipole and a weaker quadrupole surface plasmon mode. The peak of the latter branches off at about 6nm. For smaller radii, this mode is hardly discernable, since the right flank of the dipole mode is of a much larger magnitude. The nonlocality introduces bulk plasmons beyond the volume plasma frequency – a feature that is absent in the Drude model and is clearly visible in Fig. [3](d). For smaller radii, the bulk and surface resonances of the Halevi model are blue-shifted. In order to investigate in more detail the dipolar and quadrupolar surface resonances, we provide in Fig. [3](c) a corresponding blow-up, where the real part of Eq. (21) is displayed with dotted lines. The dipole follows this approximation quite well up to radii of about 4nm beyond which the approximation quickly tends to the retarded quadrupole resonance.

In fact, the retarded dipole falls below the local, electrostatic surface plasma frequency $\omega_p/\sqrt{2}$, which is not expected by Eq. (21). This turns out to be a shortcoming of the quasi-static approximation, which relies on the smallness of the parameter $\omega_{sp}/c$, an effect that would be pronounced for a surrounding medium with $\varepsilon_{HG} > 1$, e.g., glass. For larger radii, the concomitant retardation yields a redshift of the modes, which is already visible in the Drude model – otherwise the dipolar and quadrupolar mode would, in this model, be degenerate. The dispersion relation Eq. (21), applied to the Halevi model, further introduces the parameter $\beta_{HF}/\omega_{sp}$, leading to a small blue shift, which is larger for the quadrupole resonance. While the nonlocal blueshift decreases with increasing radius, the retardation-based redshift increases. Further, when considering both, retardation and the leading order in the longitudinal nonlocality, products of powers of these parameters may arise. In particular, the product of the parameters themselves is proportional to $\beta_{HF}/c \sim 10^{-2}$, such a correction is comparable to the nonlocal parameter. According to Fig. [3](c) the overall blueshift of the quadrupole still appears to surpass any redshifting contributions for radii of about $a \approx 10$nm. The retardation redshift has been phenomenologically described for small metallic spheres by employing a local material response within the framework of a self-consistent treatment of Mie theory [53].

Further, we would like to note that beyond radii of 5.5nm the right flank of the dipolar peak, which exhibits a growing asymmetry towards larger radii, increasingly overlaps with the region of bulk plasmons. It is, thus, conceivable that when deduced from numerical simulations, the field distributions of the latter become modulated by the surface plasmon mode.

In order to quantify the effect of nonlocal damping, we fit the peaks in both, the high-frequency Euler-Drude as well as the Halevi model with Lorentzians and deduce both, the width and amplitude as functions of the cylinder radius and depict the results in Fig. [3](e). The difference in widths relative to the value of the Euler-Drude model increases towards lower radii and the difference is always larger for the quadrupole mode. Both are expected from Eq. (22). Further, the increase in width is accompanied by a decrease in amplitude, which, again, turns out to be more pronounced for the quadrupole mode. We would like to note that the combined contributions due to retardation and nonlocality may be more pronounced for larger radii.

IV. TRANSLATION INTO TIME DOMAIN

In section Sec. [III] our considerations have been carried out within the frequency domain. Further insight can be obtained from time-domain considerations. For this purpose, we consider Eqs. (5) and (4). After a few algebraic manipulations,
we find
\begin{equation}
(-i\omega + \gamma) \mathbf{J}(r, \omega) = \epsilon_0\omega_0^2 \mathbf{E}(r, \omega) - \beta_{HF}^2 \nabla \rho(r, \omega) + \frac{4\epsilon_0^2}{15} \frac{\gamma}{\gamma - i\omega} \nabla \rho(r, \omega). \tag{26}
\end{equation}

From our previous discussions, we recall that the Halevi model introduces additional dispersion in the nonlocal gradient term of the high-frequency Euler-Drude equation, which leads to the last term on the r.h.s. of Eq. (26). In order to incorporate dispersive response into a time-domain numerical framework, the technique of auxiliary differential equation (ADE) is often applied [63]. In our specific case, we equate the last term of Eq. (26) with a current \(J_D\) multiplied by the negative damping rate \(-\gamma\). Performing an inverse Fourier transform to the time-domain yields the ADE
\begin{equation}
\partial_t J_D(r, t) + \gamma J_D(r, t) = -\frac{4\epsilon_0^2}{15} \nabla \rho(r, t). \tag{27}
\end{equation}

which, apart from the temporal-derivative term on the l.h.s., is reminiscent of Fick’s first law, describing an ordinary diffusion current only. The identification with the Halevi velocity as well as the Gauss-Maxwell law and considering the longitudinal stress, encoded in \(\tilde{\eta}\), eventually allows for an identification of the additional dispersion with viscoelastic shear, encoded in \(\tilde{\eta}\). As apparent in Eq. (29) and \(\epsilon_0\omega_0\mathbf{E}(r, \omega) = -\beta_{HF}^2 \nabla \rho(r, \omega)\) of the respective quantity [64] defined by
\begin{equation}
\beta_{HF}^2 = 4\epsilon_0^2 \frac{\rho}{\gamma - i\omega}. \tag{28}
\end{equation}

The squared velocity \(v_D^2/5 = -i\omega\tilde{\eta}(\omega)\) corresponds to purely elastic waves and serves to restore the Thomas-Fermi velocity in the hydrodynamic limit. A comparison of Eq. (29) with the prefactors of the density-gradient terms in Eq. (26) eventually allows for an identification of the additional dispersion with viscoelastic shear, encoded in \(\tilde{\eta}\). As apparent in the second term on the r.h.s. of Eq. (6), \(\tilde{\eta}\) enters with a numerical prefactor of 4/3. Multiplying the latter with \(v_D^2/5\), the squared diffusive velocity \(v_D^2 = 4\epsilon_0^2/15\) is recovered.

While the dynamics of \(\mathbf{J}\) is coupled directly to that of \(J_D\), as apparent in Eq. (28), the dynamics of the novel current also depends on that of \(\mathbf{J}\) via the longitudinal stress \(\nabla \rho(r, t)\) emerging in Eq. (27). We can obtain insight into the modified dynamics, introduced via the auxiliary current, by inspecting the induced charge density. For this reason, we introduce the decomposition
\begin{equation}
\rho = \rho + \rho_D. \tag{30}
\end{equation}

Using Eq. (30) in the continuity equation, we obtain \(\nabla \cdot \mathbf{J}_D = -\partial_t \rho_D\) and rephrase Eq. (27) as
\begin{equation}
v_D^2 \Delta \rho = \partial_t^2 \rho_D + \gamma \partial_t \rho_D - v_D^2 \Delta \rho.D. \tag{31}
\end{equation}

Equating the r.h.s. to zero yields the Cattaneo equation [25].

Back in 1948, Cattaneo modified the ordinary diffusion current of a classical substance, given by Fick’s first law, by employing a relaxation-time approach. This enforces a finite propagation velocity of the classically diffusing quantity. Quantities that obey this type of equation perform a hybrid diffusive-wavelike propagation [47, 64]. Which propagation paradigm dominates, depends on the relative temporal change of the respective quantity [64] defined by
\begin{equation}
f_D = \frac{1}{2\pi} \left| \frac{\partial \rho_D}{\rho_D} \right|. \tag{32}
\end{equation}

in our case. Comparing this rate to \(\gamma\), the r.h.s. of Eq. (31) suggests wavelike transport in the collision-less regime \((f_D \gg \gamma)\) and diffusive transport in the collision-dominated regime. Accordingly, close to the collision-less regime, a small diffusive contribution obstructs a purely wave-like motion [47]. However, we would like to note that this hybrid propagation is modulated by the dynamics of \(\tilde{\eta}\), which, in general, cannot be treated as a source term.

Actually, both types of propagation are found for the total induced charge. In order to facilitate the discussion, we introduce the rate \(f\) similar to Eq. (32), but in terms of \(\rho\). Considering Eqs. (27) and (28) in conjunction with the Gauss-Maxwell law and the continuity equation, yields
\begin{equation}
-\omega_0^2 \rho = (\gamma \partial_t - \beta_{TF}^2 \nabla^2) \rho, \tag{33}
\end{equation}

in the hydrodynamic \((f, f_D \ll \gamma)\) and
\begin{equation}
-\omega_0^2 \rho = (\partial_t^2 - \beta_{HF}^2 \nabla^2) \rho, \tag{34}
\end{equation}

in the elastic regime \((f, f_D \gg \gamma)\). Accordingly, in these regimes the conduction electrons exhibit diffusive and wavelike transport, respectively, with a propagation velocity that changes from \(\beta_{TF}\) to \(\beta_{HF}\). This is linked to the insight, that the bare Euler-Drude model yields a Cattaneo-type differential equation for the totally induced charge.

However, we would like to note that the Cattaneo-type dynamics of the Euler-Drude model is further modulated by the Coulomb interaction which introduces a restoring force that keeps the electrons within the metal (see, for instance, the l.h.s. of Eqs. (33) and (34)). Incidentally, since the restoring term originates in the electric field that enters Eq. (26) and since screened dielectric functions are constructed as those functions which connect the current not only to some external but also to the internal field, we find
\begin{equation}
\epsilon_H(k, \omega) = 1 - \frac{\omega_0^2}{\omega(\omega + i\gamma) - \beta_{HF}^2 k^2 + \gamma v_D^2 k^2 / (\gamma - i\omega)}, \tag{35}
\end{equation}

where \(\epsilon_H(k, \omega)\) is the complex dielectric function of the metal, \(\omega_0\) is the cutoff frequency, and \(k\) is the wave vector. The factor \(1 - \epsilon_H(k, \omega)\) accounts for the screening of the electric field, which reduces the restoring force and thereby the propagation velocity.
Accordingly, the restoring term, proportional to $\omega_p^2$, only enters through the numerator of the second term on the r.h.s. of Eq. (35). In the hydrodynamic regime, the pole of this very term turns purely diffusive, i.e. $\omega = -i(\beta_H^2/\gamma)k^2$, and becomes purely wavelike in the elastic regime, i.e. $\omega = \beta_F^2 k^2$.

Finally, we would like to note that the quantum-mechanical nature of the conduction electrons enters the equations of motion solely via the characteristic velocities, which are proportional to the Fermi velocity. Apart from this, these equations are similar to those of classical material dynamics. This can be understood by reformulating the fourth condition of the viscoelastic model, (see Eq. (5)) to

$$\ell \sim v_F / \gamma \gg 1/k_F \sim \lambda_F. \quad (36)$$

As a result, the (bulk) mean-free-path $\ell$ is much larger than the Fermi wavelength $\lambda_F$ within the viscoelastic model. Hence, a representative electron conducts a random walk in which successive collisional events are independent of each other and quantum interference effects are absent [38].

V. COMPARISON WITH THE GNOR MODEL

In the previous Sec. IV we have shown, that the Halevi model extends the high-frequency Euler-Drude model via an additional current that shares some similarities with a diffusive current according to Fick’s first law. An extension of the Euler-Drude model due to diffusive dynamics is also a key element of the GNOR model [18]. Therefore, it is worthwhile to discuss similarities and differences of the GNOR and the Halevi model.

In Refs. [15] [18] [19] and [65] a generalized formal approach to nonlocality within linear response theory is discussed. Given a position $r$ within the electron continuum, as a central approximation, the influence of nonlocal processes is limited to a neighborhood with an extent that is much smaller than the length scale of variation of the electric field. Within the constitutive equations, this allows for a Taylor expansion of the field of the electron continuum where each term corresponds to a certain order of nonlocality. Given a homogeneous, isotropic medium with local response described by the Drude model, the leading-order-nonlocal current density is then given by

$$-i\omega J(r, \omega) = \epsilon_0 \omega_p^2 E(r, \omega) - \gamma J(r, \omega)$$

$$-\nu^2 \nabla [\epsilon_0 \nabla \cdot E(r, \omega)]. \quad (37)$$

The last term on the r.h.s. lumps together the effects of the considered order of nonlocality that extends the local Drude response.

Here, we would like to note that the differential operator in space is motivated by the Euler-Drude equation. As remarked in Ref. [19] the nonlocality of the latter only manifests itself in the longitudinal current response. Since it has been pointed out, that surface plasmons are affected not only by the longitudinal response [65], we add that the transverse nonlocality of the viscoelastic model, Eq. (6), serves as a plausible amendment.

Eventually, in order to construct the GNOR model, first the center-of-mass current $\rho_0 v$ is defined to fulfill the high-frequency Euler-Drude model, while the total current is defined by the sum of the latter and a diffusive current [18] [19] [65], such that

$$J(r, \omega) = \rho_0 v(r, \omega) - D\nabla \rho(r, \omega). \quad (38)$$

Accordingly, the diffusion is basically introduced by hand, following Fick’s first law [25] [65], given a diffusion constant $D$. In order to prohibit the build-up of charges, this constant has to be positive.

It can then be shown, that

$$\nu^2 = \beta_H^2 + D(\gamma - i\omega). \quad (39)$$

We would like to note that the r.h.s. of Eq. (39) provides the counterpart of Halevi’s interpolation formula defined in Eqs. (4) and (29). While both models include the high-frequency Euler-Drude model, which introduces a nonlocal process with real-valued length scale $\ell_\beta = \beta_H / \gamma$, both, the GNOR and the Halevi model, Eqs. (39) and (29), respectively, provide an extension of $\beta_H$ to complex-valued velocities.

In the original GNOR model, the diffusion constant is estimated via $D \sim v_F^2 / \gamma$ [18], yielding a diffusive length scale $\ell_D = \sqrt{D/\omega} \sim v_F / \sqrt{\omega_p}$, which we have evaluated at plasmonic frequencies. Therefore, considering typical metals this diffusive scale is much larger than $\ell_\beta \sim \sqrt{\gamma/\omega_p} \ell_D$. In contrast, within the Halevi model $\ell_\beta$ surpasses the diffusive length scale, the latter being given by $\ell_D^{Hal} \sim \sqrt{\gamma/\omega_p}(v_F/\omega_p) \sim \sqrt{\gamma/\omega_p} \ell_\beta$. The mismatch in the frequency dependence of each extension to $\beta_H$ has also been noted in Ref. [57].

Further, within the GNOR model, the additional current reads

$$\gamma J_{GNOR}(r, \omega) = [\nu^2 - \beta_H^2] \nabla \rho(r, \omega), \quad (40)$$

where we isolated the part proportional to $\beta_H$ stemming from the Euler-Drude model. Upon inserting Eq. (39) as well as the continuity equation, where the total current now obeys Eq. (38) per construction, we find

$$\gamma J_{GNOR}(r, \omega) = D\gamma \nabla \rho(r, \omega) - D\nabla [\nabla \cdot J(r, \omega)]. \quad (41)$$

The dynamics of the total current within the GNOR model then obeys

$$-i\omega J(r, \omega) = \epsilon_0 \omega_p^2 E(r, \omega) - \gamma(r, \omega) J(r, \omega)$$

$$- (\beta_H^2 + D\gamma) \nabla \rho(r, \omega) + D\nabla (\nabla \cdot J(r, \omega)). \quad (42)$$
As a result, we see that the first term on the r.h.s. of Eq. (41) adds to the density gradient term of the high-frequency Euler-Drude equation. Beyond that, the second term on the r.h.s. of Eq. (41) formally reproduces the longitudinal shear contribution of Eq. (6). The corresponding coefficient has to be positive, which is consistent with $D > 0$. If we then utilize the frequency-dependent (real-valued) diffusion constant deduced in Ref. [19] by equating the imaginary parts of Eqs. (39) and (4) given by $\frac{1}{\gamma} \sqrt{\frac{\gamma^2}{\omega^2 + \gamma^2}}$, we see that the density gradient term of Eq. (42) is equipped with the characteristic velocity $\beta_{HF}^2$ in the high-frequency regime and it is equipped with $\beta_{HF}^2 + v_D^2 > \beta_{TF}^2$ in the low-frequency regime. This results from the fact that $D(\omega \gg \gamma) \sim \gamma/\omega^2$ and $D(\omega \ll \gamma) \sim 1/\gamma$, which is not the case in the Halevi model.

Beyond that, by equating the real parts of Eqs. (39) and (4), we find the negative of the r.h.s. of Eq. (43) if we assume a real-valued diffusion constant.

Another difference between the GNOR and the Halevi model arises, when we derive the counterpart of Eq. (38) for the Halevi model. For this purpose, we consider Eq. (26) and define the total current as

$$J(r, \omega) = \rho_0 v(r, \omega) + f_H(r, \omega).$$  

(44)

Trying to restore the high-frequency Drude-Euler equation for $\rho_0 v$, we require the equivalence between $f_H(r, \omega)$ and the residual terms and obtain

$$f_H(r, \omega) = -\frac{v_D^2}{\gamma} \frac{\gamma^2}{(\omega + i\gamma)^2} \nabla \rho(r, \omega).$$  

(45)

This additional current is not simply defined by Fick’s first law. The difference originates in the way how the local equilibration is implemented in the Mermin approach. To yield equivalence with the Ansatz in Eq. (38), the diffusion constant does not only have to be frequency-dependent, but also complex-valued.

The particular choice of diffusive paradigm also influences the longitudinal wavenumber. Therefore, we added the results of the GNOR model to Figs. 2a and 2b, using the approximation $D \approx \frac{\beta_{HF}^2}{\gamma}$ that has been suggested in Ref. [18]. First, the real part exhibits a low-frequency local maximum, just as the Halevi model. However, there is no additional kink root below the plasma frequency. Further, beyond the plasma frequency the high-frequency Euler(-Drude)-model is not followed as closely. Considering the imaginary part, the amplitude of the low-frequency maximum is smaller than in the high-frequency Euler-Drude model as well as the Halevi model, owing to the larger velocity $\sqrt{\beta_{HF}^2 + D(\gamma)} > \beta_{HF}$, $\beta_{TF}$. This larger velocity is also tied to a larger blueshift for resonances that potentially occur within this frequency regime in nano-objects when considering a hardwall boundary condition. A similar observation has been made in Ref. [62]. Further, for frequencies beyond the volume plasma frequency, the GNOR model does not follow the high-frequency Euler-Drude model as close as does the Halevi model. Specifically, the imaginary part increases in the GNOR-model instead of decreasing in the Euler-Drude and the Halevi model.
However, owing to the different scaling with the ratio $\gamma/\omega$, the Halevi model is firmly rooted in bulk arguments. As a result, we would like to note that Ref. [52] only considers the Boltzmann equation without the Mermin correction while the latter has been mentioned in one of the original publications on the GNOR model in Ref. [19]. Actually, in Ref. [40], the Mermin correction has been linked to an additional, diffusive current. We stress this fact, since the Mermin-Ansatz for the single-relaxation-time approximation introduces an additional term which prohibits the occurrence of a charge sink in the continuity equation, as shown in Refs. [8] and [42].

Finally, we would like to note that Ref. [62] only considers the Boltzmann equation without the Mermin correction while the latter has been mentioned in one of the original publications on the GNOR model in Ref. [19]. Actually, in Ref. [40], the Mermin correction has been linked to an additional, diffusive current. We stress this fact, since the Mermin-Ansatz for the single-relaxation-time approximation introduces an additional term which prohibits the occurrence of a charge sink in the continuity equation, as shown in Refs. [8] and [42].

VI. TEMPORAL EVOLUTION OF FIELDS IN THE HALEVI MODEL

Eqs. (1), (27) and (28) together with the slip boundary condition comprise the complete system of equations for Halevi model of plasmonic materials.

In order to solve the Maxwell equations with this material model for an arbitrary geometry of a scatterer, we implement the time-domain version of the Halevi model via ADEs (see Sec. [IV]) into our home-made Discontinuous Galerkin Time-Domain (DGTD) approach [63] which is a finite-element method that is specifically designed to solve equations in conservation form and we have utilized the algorithm in nodal form developed by Hesthaven and Warburton [68] and typically employ third-order Lagrange polynomials as basis functions and confirm that fourth-order polynomials give the same results. Further, within the DGTD approach a numerical flux is introduced in order to couple adjacent elements. We use a pure upwind flux for the Maxwell equations [63] and employ for Eqs. (1) and (28) the Lax-Friedrichs flux [68]. Finally, we solve the ADE, Eq. (27), using a central flux. The resulting DGTD spatial discretization yields a set of ordinary

![Fig. 5. Illustration of the time evolution of the current components $J_{0,y}$ (upper row) and $J_{0,x}$ (lower row) of the auxiliary current [Eq. (27)] within the horizontal cross section of a silver cylinder (radius $a = 10$ nm, see Fig. 1 for the general scattering setup). Starting from the surface region an oscillatory contribution propagates towards the cylinder’s center and finally extends across the entire cross section.](image-url)
differential equations of first order in time which we solve via a 4th-order Low-Storage Runge-Kutta method with 14 stages \cite{69}. The Drude and Euler-Drude model have been implemented within DGTD and for details, we refer to Refs. \cite{63} and \cite{70}, respectively.

For our subsequent simulations, we consider a cylindrical silver wire of radius 10nm (see Tab. \ref{tab:material_params} for the material parameters) in vacuum. The wire is illuminated by an electromagnetic plane wave with wave vector and electric field normal to the cylinder axis, i.e. identical to the analytical case of Sec. \ref{sec:analytical} (see also Fig. \ref{fig:setup} for an illustration of the setup).

The exciting pulse is centered around $\omega_0 = 7.4\text{eV}$ (which is roughly 0.8 times the plasma frequency) and has a Gaussian envelope with a FWHM of $1.57\text{fs}$. This pulse is injected into the system using the TF/SF technique \cite{63}. Since the problem effectively reduces to two dimensions, the TF/SF contour is chosen to be a square with edge length 40nm centered on the cylinder axis. The pulse is launched from the left edge of the TF/SF contour so that the maximum at the TF/SF contour occurs at $t_0 = 4.67\text{fs}$. The scattered field box is bounded by the TF/SF contour and a centered square of edge length 440nm. To prevent unphysical back reflexions, we surround the entire computational domain with perfectly-matched layers and apply Silver-Müller boundary conditions at the outer boundary given by a centered square with edge length 1.04μm. The corresponding mesh is generated using Gmsh \cite{71}. In order to adequately resolve the cylindrical material interface, we utilize a minimal (maximal) insphere radius of 0.04nm (0.32nm) for the finite elements near this interface.

In Sec. \ref{sec:analytical} we have elaborated on the Halevi model as an extension of the high-frequency Euler-Drude model. Therefore, we now proceed to perform a direct comparison of the two models and focus on the relative difference in the field distributions. Incidentally, the relative difference at this time instance is reminiscent of the mode picture of hydrodynamic bulk plasmons \cite{70}. As such, the corresponding $y$-component possesses a mirror symmetry with respect to the $xz$-plane and is mapped onto itself by a rotation of 180 degree around the cylinder axis. The $x$-component, however, is mapped onto itself by a 90 degree rotation. These symmetries are also present at the other time instances. Nonetheless, the differences in the field distributions are rather weak.

In Sec. \ref{sec:results} we have shown, that the novel dispersion of the Halevi model (as compared to the high-frequency Euler-Drude model, Eq. \ref{eq:Euler-Drude}) translates into an additional current, that follows Eq. \ref{eq:current}. Accordingly, we expect, that the difference in the electric field is tied to the spatio-temporal evolution of this additional current. Consequently, in Fig. \ref{fig:results} we display the $x$- and $y$-component of the additional current $J_D$ at the same time instances where we depicted the field differences in Fig. \ref{fig:differences}. We observe that, at all time instances, both components concentrate the maximal amplitude at the surface of the cylinder. However, a small contribution is given by the spatially confined oscillations, which propagate in essentially the same way as the relative difference in the field components, also respecting the afore-discussed symmetry properties.

In order to measure, how fast the oscillations in the relative field difference propagate into the scatterer, we consider a cut through the cylinder center as indicated in Fig. \ref{fig:results}. Given the pulsed nature of our excitation, these oscillations build up and assume the form of wave packets – due to symmetry one in each half of the cut. In Fig. \ref{fig:results}(c) we provide an illustrative time frame. To highlight the spatial confinement, we determine the interpolative (upper and lower) envelope. Specifically, we sample the maxima and minima and perform corresponding cubic spline fits \cite{72}.

This approach works well for positions away from the surface but experiences some problems near the surface where the boundary conditions obstruct the formation of well-developed wave packets. From the global extrema of the upper and lower envelopes, we can deduce the movement of the wave packets which ultimately yields two estimates of the corresponding group velocity. For instance, we observe in Fig. \ref{fig:results}(c) that the extrema lie close to each other from about $t \approx 12.03\text{fs}$ (at which time a nice wave packet has build up) up to about $t \approx 23.05\text{fs}$ where the two wave packets have merged. From a linear regression, we obtain the velocities $v_{ue} = 0.00138c$ and $v_{le} = 0.00139c$ (where $c$ is the vacuum speed of light) for the upper and lower envelope, respectively. This is a rather interesting result, since these velocities are considerably smaller than all velocity scales that we have discussed so far. In fact, the values are closest to the elastic shear velocity $v_F/\sqrt{5}$.

Further, we note that within the pulse the spatial frequency experiences a chirp. Therefore, we provide in Fig. \ref{fig:results}(b) the
A solution of the spectrogram corresponds to about half the corresponding spectrogram. Although the minimal spatial resolution of this signal. While this provides an overview of the relevant contributions it lacks the desired spatial resolution. Therefore, we also provide in Fig. 4(b) the corresponding spectrogram. Although the minimal spatial resolution of the spectrogram corresponds to about 0.1 nm, the inverse scaling between spatial frequency and spatial resolution forces us to fix the spatial resolution to about 1 nm. Despite its coarseness, this resolution allows us to elaborate on the positive spatial chirp (towards the cylinder center) within each wave packet. Specifically, we find that the highest available frequencies are tied to the region, where the wave packets are located and, in this region, they increase towards the center.

We conclude our survey by calculating the modal distributions for the dipolar and quadrupolar resonance for both, the Halevi and the high-frequency Euler-Drude model from our time-domain simulations via an on-the-fly Fourier transform. In Fig. 7 we depict the real part of the electric field distributions at these resonances for the Halevi model and the difference of these field distributions to the results of the high-frequency Euler-Drude model. In addition, we normalize the difference to the largest value obtained within the Euler-Drude model at the respective frequency in order to bring out better...
the deviations between the results.

First, we observe that these modal patterns within the Halevi model are compatible with the symmetry classification of the Euler-Drude model [14]. Further, the field amplitudes in the Halevi model are enhanced. Specifically, for the dipole resonance we obtain an enhancement in the vicinity of the surface with a four-fold rotation symmetry for the $x$- and $y$-component of the electric field. For the quadrupolar resonance, the differences inside the scatterer are more inhomogeneous. Overall, the field enhancement is relatively small.

VII. CONCLUSION AND OUTLOOK

To summarize, we have analyzed the physical properties of the Halevi model for plasmonic materials. In particular, we have identified the Halevi model as an extension of the Euler-Drude model and have established its relation to the viscoelastic model by way of spectral as well as spatio-temporal investigations of different aspects of plasmonic light-matter interaction. We have further carried out an extension of the Mie-Ruppin theory of hydrodynamic cylinders to the Halevi model and determined the quasi-static dispersion relation of surface plasmons. Thereby, we have revealed a novel damping term which exibits formal similarities to the collision-modified Landau-damping of Halevi. In turn, this leads to a novel physical justification of an earlier phenomenological description of limited-mean-free path effects of dipole surface modes and we have shown how this novel damping term also affects higher-order surface modes. The shear-extension within the Halevi model eventually has lead to nonlocal surface damping. Further, the Halevi extension to the Euler-Drude model led to an increase in the relative difference of the width of surface dipolar and quadrupolar extinction peaks for cylinder radii in the range of $1$ to $10\text{nm}$, where, generally, the effect is larger for the quadrupolar resonance.

To complement the spectral investigations, we have employed the ADE technique to adopt the Halevi model for time-domain simulations. The additional dispersion of the latter yields a modification of the diffusion current of Fick-type. We have shown that the propagation of the induced charge, related to the current modifications, shares similarities with a hybrid, diffusive-wavelike paradigm as described by the Cat-taneo equation.

We have completed our analytical analysis by comparing the Halevi model to the GNOR model – the latter being another extension of the the Euler-Drude model that includes a diffusive contribution to the current without asymptotical comparison to a semiclassical model. The GNOR-model features several differences to the Halevi model that include a low-frequency, characteristic velocity exceeding the Thomas-Fermi value and the deviation from the Euler-Drude model for frequencies beyond the volume plasma frequency.

Further, we have shown that the latter model is connected to a difference in the scaling of the diffusive length scale at plasmonic frequencies by an additional factor $\omega_p/\gamma$ relative to the Halevi model. For typical metals and assuming a frequency-independent GNOR-diffusion constant this additional factor takes on values $\omega_p/\gamma \sim 10^2 \cdots 10^3$. As a result, while in the GNOR model the diffusive length exceeds the length scale of combined compression and shear, the opposite case is manifest Halevi model. Finally, we have shown that a direct comparison between the GNOR and the Halevi model at intermediate frequencies suggest that the GNOR-diffusion constant is complex-valued and frequency-dependent.

Next, by employing the time-domain formulation of the Halevi model, we have numerically determined the spatio-temporal evolution of the electric fields distributions in and around silver nano-wires. We have found that under pulsed excitations, the differences of the electric field components between the Halevi and the Euler-Drude model take on the form of wave pakets that build up at the sub-surface region, subsequently propagates towards the cylinder center from where they spread across the entire cross section of the cylinder and eventually fade away. This behavior correlates with a concomitant oscillatory contribution of the diffusive current. Upon analyzing the resulting wave paket envelopes, we have inferred an estimate of the corresponding group velocity which, in fact, has turned out to be smaller than all natural scales of the Halevi and viscoelastic model. Further, we have determined a positive chirp of the wave pakets spatial frequencies towards the center of the cylinder. In addition we have performed on-the-fly Fourier transforms of the temporal evolution of the field distribution and have determined the mode distributions of the dipolar and quadrupolar resonance. The Halevi model preserves the respective symmetries of the Euler-Drude model and – at least for monomers – the differences between both models are rather small.

Based on our results, we would like to provide a few comments regardin possible future routes. First of all, all our results rely on the hardwall boundary condition in the form of the so-called slip boundary condition. On the mesoscopic scale, this constraint could be lifted by introducing an infinitesimal charge sheet, e.g., via a variation of the composite-surface model of Ref. [75]. On the one hand, this would yield an approximate treatment of spill-out effects that are presently not contained within our model. On the other hand, it might be used to treat charge transfer effects at the interface to an embedding medium. This could phenomenologically for ‘chemical interface damping’, i.e., charge-transfer effects across the surface of a plasmonic nano-particle and its surrounding host material [58]. This could be further compared to the theory of Persson [60] that connects the latter to interactions with resonance states of adsorbates that form at a metal surface. In addition, a detailed discussion on the difference between slip- and no-slip boundary conditions would be highly desirable.
The latter cannot be enforced ad hoc, but has to be physically motivated with regard to the specific surface characteristics of the scatterer under consideration. A potential justification could be inferred by embedding the Halevi model into the viscoelastic model which, in turn, would provide a clear path to the introduction of transverse nonlocality. Specifically, the absence of a tangential surface current due to e.g. surface irregularities can affect the neighborhood of the surface due to shear, thus providing a physical motivation of the no-slip model. So far, the latter has been considered only for a half-space of metal yielding a nonlocal correction to the well-known s-polarized Fresnel formula. Further, an application of the full viscoelastic model to thin semiconductor films has been performed in Ref. [57] albeit with the slip boundary condition.

Finally, for the setup of a single nano-wire considered in our work, the relative differences in the widths and amplitudes of resonances between the Halevi and Euler-Drude model have turned out to be rather small. However, this does not always have to be the case. It is, therefore, very interesting to seek for physical setups in which changes to the optical, dielectric bulk properties (as encoded in the dielectric functions) are more pronounced. For instance, Kreibig and Fragstein have pointed out, that the extinction of metallic nanospheres is very sensitive to the changes of bulk quantities. Further, it might be interesting to investigate dimer and related structures, i.e., to consider the effect of the Halevi model on the electromagnetic field distribution in nano-gap systems. Recently, such systems have witnessed significant attention due to the potential of strongly modified light-matter interaction, for instance, with regards to strong coupling of emitters and/or enhanced nonlinear optical effects.

ACKNOWLEDGMENTS

G.W. and K.B. acknowledge funding by the German Research Foundation (DFG) in the framework of the Collaborative Research Center 1375 ‘Nonlinear Optics down to Atomic Scales (NOA)’ (project number 398816777). N. A. M. is a VILLUM Investigator supported by VILLUM FONDEN (Grant No. 16498). The authors wish to thank Matthias Plock for fruitful discussions.

Appendix: Quasi-static cylindrical surface plasmons

In this appendix, we calculate the quasi-static approximation for cylindrical surface plasmons within the Halevi model, Eq. (21).

In Sec. III, we have considered the scattering of a plane wave that propagates in vacuum and is normally incident onto an infinitely extended circular cylinder for the polarization perpendicular to the cylinder axis. Via Ruppin’s extension of the corresponding Mie solution (expansion into spherical harmonics), we have obtained the expansion coefficients for of the scattered field in Eq. (15). The surface resonances can be determined from the poles of expansion coefficients according to

\[ 0 = \left[ \epsilon_n + D_n(k_ar) \right] H_n(k_0a) - \sqrt{\epsilon_T(\omega)} H'_n(k_0a). \]  

(A.1)

As an illustration, we consider silver with material constants given by Tab. 4. From the definitions of the three wave numbers \( k_0, k_L, k_T \), we deduce for cylinders with radii \( a = 1\ldots10 \text{nm} \) that

\[ |k_0a|, |k_Ta| \lesssim 0.1 \quad \text{and} \quad 10 \lesssim |k_La|. \]  

(A.2)

Accordingly, we may utilize of Eq. (A.1) the Bessel functions’ asymptotic representations. We then find the implicit dispersion relation

\[ \epsilon_T(\omega) = -1 - \frac{i\gamma}{2k_1a} + O\left(\frac{\gamma^2}{\omega^{2sp}}\right). \]  

(A.3)

Here, we have selected the solution with a positive real part. Note, that this is also the local dispersion relation for surface plasmons at a planar interface between vacuum and a half-space filled with metal. Consequently, for normally incident and quasi-static electric fields polarized perpendicular to the cylinder axis, the geometry does not alter the local surface plasmons. This is a consequence of the quasi-static approximation. Otherwise, even the local solution would depend on the pole order \( n \) and radius \( a \).

Next, we seek a solution to Eq. (A.3), that includes nonlocal correction to Eq. (A.4) to leading order. Therefore, we use the Ansatz

\[ \omega_n = \omega_{sp} \left( 1 - \frac{i\gamma}{2\omega_{sp}} + f_n \right) \text{ with } |f_n| \ll 1. \]  

(A.5)

Considering the Taylor expansion in \( f_n \) of Eq. (11) around \( \omega = \omega_n \), we find

\[ \epsilon_T(\omega_n) = \epsilon_T(\omega_{sp}) + \epsilon'_T(\omega_{sp})\omega_{sp}f_n + O(f_n^2), \]  

(A.6)

where we have introduced \( \omega_{sp} = \omega_{sp} - i\gamma/2 \). To make further progress, we require the following Taylor expansions in \( \gamma/\omega_{sp} \)

\[ \epsilon_T(\omega_{sp}) = -1 + O\left(\frac{\gamma^2}{\omega_{sp}^2}\right) \]

\[ \epsilon'_T(\omega_{sp}) = \frac{4}{\omega_{sp}} + O\left(\frac{\gamma^2}{\omega_{sp}^2}\right). \]  

(A.7)
which yields
\[\epsilon_T(\omega_n) \approx -1 + 4 f_n.\]  
(A.8)

Equating this result with the r.h.s. of Eq. (A.3), we find for the nonlocal correction the implicit relation
\[f_n = \frac{in}{2\alpha k_L(\omega_n)}.\]  
(A.9)

 Keeping Eq. (A.18) in mind, we have to expand Eq. (A.4) into a manner analogous to Eqs. (A.6, A.7) and eventually arrive at
\[\beta_H^2(\omega_n) \approx \left(1 - \frac{A_i \gamma}{9 \omega_{sp}^2}\right) \beta_{HF}^2.\]  
(A.10)

Here, we have dropped the first-order term of the Taylor expansion because this is proportional to \((\gamma/\omega_{sp}) f_n\).

Upon inserting Eqs. (A.8, A.10) into Eq. (18) we obtain
\[\frac{1}{k_L(\omega_n)} \approx -\frac{i\beta_{HF}}{\omega_{sp}} \left[1 - \frac{2i \gamma}{9 \omega_{sp}^2} + f_n\right]\]  
(A.11)

which, when inserted into Eq. (A.9), gives
\[f_n = \frac{n\beta_{HF}}{2\omega_{sp}^2} \left(1 - \frac{2i \gamma}{9 \omega_{sp}^2}\right) \left[1 - \frac{n\beta_{HF}}{2\omega_{sp}^2}\right]^{-1}\]  
(A.12)

Indeed, since \(n \ll \omega_{sp}^2/\beta_{HF}\) and \(\gamma \ll \omega_{sp}\), we find \(|f_n| \ll 1\). To first order in \(n\beta_{HF}/\omega_{sp}^2\), the second term inside the square brackets can be neglected and this gives Eq. (21).

[1] Q. Zhang et al., Electron Energy-Loss Spectroscopy of Spatial Nonlocality and Quantum Tunneling Effects in the Bright and Dark Plasmon Modes of Gold Nanosphere Dimers, Adv. Quantum Technol. 1, 1800016 (2018).
[2] T. Itoh, Y. S. Yamamoto, Y. Kitahama, and J. Balachandran, One-dimensional plasmonic hotspot locations between silver nanowire dimers evaluated by surface-enhanced resonance Raman scattering, Phys. Rev. B 95, 115441 (2017).
[3] M. Rothe, Y. Zhao, J. Müllner, G. Kewes, C. T. Koch, Y. Lu, and O. Benson, Self-Assembly of Plasmonic Nanoantenna--Waveguide Structures for Subdiffractional Chiral Sensing, ACS Nano 15, 351 (2021).
[4] A. Wiener, H. Duan, M. Bosman, A. P. Horsfield, J. B. Pendry, J. K. W. Yang, S. A. Maier, and A. I. Fernández-Dominguez, Electron-Energy Loss Study of Nonlocal Effects in Connected Plasmonic Nanoantennas, ACS Nano 7, 6287 (2013).
[5] A. Sundaramurthy, K. B. Crozier, G. S. Kino, D. P. Fromm, P. J. Schuck, and W. E. Moerner, Field enhancement and gap-dependent resonance in a system of two opposing tip-to-tip Au nanoantangles, Phys. Rev. B 72, 165409 (2005).
[6] G. W. Bryant, F. J. García de Abajo, and J. Aizpurua, Mapping the Plasmon Resonances of Metallic Nanoantennas, Nano Lett. 8, 631 (2008).
[7] E. Zaremba, Surface-plasmon dispersion for diffuse boundary scattering, Phys. Rev. B 9, 1277 (1974).
[8] G. Wegner and C. Henkel, Remarks about surface plasmons and their stability, arXiv:2005.03716 [physics.optics] (2020).
[9] C. David and F. J. García de Abajo, Surface Plasmon Dependence on the Electron Density Profile at Metal Surfaces, ACS Nano 8, 9558 (2014).
[10] C. Ciraci, E. Poutrina, M. Scalora, and D. R. Smith, Origin of second-harmonic generation enhancement in optical split-ring resonators, Phys. Rev. B 85, 204103 (2012).
[11] C. Ciraci, E. Poutrina, M. Scalora, and D. R. Smith, Second-harmonic generation in metallic nanoparticles: Clarification of the role of the surface, Phys. Rev. B 86, 115451 (2012).
[12] D.-N. Huynh, M. Moeferdt, C. Matyssek, C. Wolff, and K. Busch, Ultrastark three-wave-mixing in plasmonic nanostructures, Appl. Phys. B 122, 139 (2016).
[13] K. R. Hiremath, L. Zschiedrich, and F. Schmidt, Numerical solution of nonlocal hydrodynamic Drude model for arbitrary shaped nano-plasmonic structures using Nédélec finite elements, Journal of Computational Physics 231, 5890 (2012).
[14] M. Moeferdt, T. Kiel, T. Sproll, F. Intravaia, and K. Busch, Plasmonic modes in nanowire dimers: A study based on the hydrodynamic Drude model including nonlocal and nonlinear effects, Phys. Rev. B 97, 075431 (2018).
[15] N. A. Mortensen, Mesoscopic electrodynamics at metal surfaces — From quantum-corrected hydrodynamics to microscopic surface-response formalism, Nanophotonics 10, 2563 (2021).
[16] R. Ruppin, Extinction properties of thin metallic nanowires, Opt. Commun. 190, 205 (2001).
[17] G. Toscano, J. Straubel, A. Kwiatkowski, C. Rockstuhl, F. Evans, X. H., M. N. A., and W. M, Resonance shifts and spill-out effects in self-consistent hydrodynamic nanoplasmonics, Nat. Commun. 6, 7132 (2015).
[18] N. A. Mortensen, S. Raza, M. Wubs, T. Søndergaard, and S. I. Bozhevolnyi, A generalized non-local optical response theory for plasmonic nanostructures, Nat. Commun. 5, 3809 (2014).
[19] S. Raza, S. I. Bozhevolnyi, M. Wubs, and N. A. Mortensen, Nonlocal optical response in metallic nanostructures, J. Condens. Matter Phys. 27, 183204 (2015).
[20] C. R. Martin, Nanomaterials: A Membrane-Based Synthetic Approach, Science 266, 1961 (1994).
[21] M. I. Stockman, Nanofocusing of Optical Energy in Tapered Plasmonic Waveguides, Phys. Rev. Lett. 93, 137404 (2004).
[22] R. M. Dickson and L. A. Lyon, Unidirectional Plasmon Propagation in Metallic Nanowires, The Journal of Physical Chemistry B 104, 6095 (2000).
[23] I. Villó-Pérez and N. R. Arista, Hydrodynamical model for bulk and surface plasmons in cylindrical wires, Surf. Sci. 603, 1 (2009).
[24] S. Conti and G. Vignale, Elasticity of an electron liquid, Phys.
 Particle Resonators: Numerical Analysis on the Basis of the Hydrodynamic Drude Model, J. Phys. Chem. B 120, 1163 (2016).

[71] C. Geuzaine and J.-F. Remacle, Gmsh: A 3-D finite element mesh generator with built-in pre- and post-processing facilities, Int. J. Numer. Meth. Engng. 79, 1309 (2009).

[72] Y. Yang, A Signal Theoretic Approach for Envelope Analysis of Real-Valued Signals, IEEE Access 5, 5623 (2017).

[73] B. Horovitz and C. Henkel, Surface plasmons at composite surfaces with diffusive charges, Europhys. Lett. 97, 57010 (2012).