Bulk and surface plasmons: Wave-mechanical and second-quantized theories

Jesper Jung and Ole Keller*
Institute of Physics, Aalborg University, Skjernvej 4A, DK-9220 Aalborg Øst, Denmark

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In this paper we formulate the quantum theory for longitudinal (rotational free) collective electron modes, the so-called plasmons. Starting from the jellium dispersion relation, a first-quantized wave-mechanical theory is established. We show that plasmon quantum (quasi)particles are charged bosons described by complex Klein-Gordon fields satisfying a "relativistic" scalar Klein-Gordon equation in which the speed of light c is replaced by a velocity a on the order of the Fermi velocity. Based on a formally ($c \rightarrow a$) covariant description, the first-quantized theory is extended to the second-quantized level via a Lagrangian formalism. We show how the second-quantized theory enables the study of the plasmon quantum particles interaction with an electromagnetic gauge field via a modification of the free plasmon Lagrangian density by the minimal coupling principle. Utilizing the Weyl expansion, first- and second-quantized theories for surface-plasmon quantum particles are established in close analogy to those of the bulk plasmon quantum particle. This work opens up for the study of, e.g., squeezed, entangled, and coherent plasmon states in line with what has been studied theoretically and experimentally for photons for many years.

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I. INTRODUCTION

In solid-state plasma physics one studies those electrodynamics properties which in a first approximation can be described in terms of collective electron excitations [1]. In free-electron metals and semiconductors, and in BCS superconductors, the jellium model constitutes a good starting point for many theoretical analyses. In this model the electrons interact with one another in a uniformly smeared-out ionic background potential [2,3]. It is known that the collective electrodynamics properties of the electrons are well described in the framework of the random-phase-approximation (RPA) model. In this approach both single-particle (electron-hole pair formation) and collective excitations are included. At long wavelength only collective modes contribute to the microscopic dielectric response $\boldsymbol{\varepsilon}(\mathbf{q}, \omega)$ at wave vector $\mathbf{q}$ and cyclic frequency ω [4].

If we limit ourselves to the collective part of the excitation spectrum, quantum physics tells us that one would expect to be able to observe single-quantum phenomena. In the limit $\mathbf{q} \rightarrow \mathbf{0}$, the quantum energy $\hbar\omega_p$ (ω_p being the plasma frequency) has been observed by external excitation with electrons and light [1,4–8]. Also, plasmon dispersion relations $[\mathbf{q} = \mathbf{q}(\omega)]$ have been investigated in some detail. For finite q values it is important to distinguish between longitudinal (rotational-free) and transverse (divergence-free) plasmons, named L plasmons and T plasmons. The T plasmon can be resonantly coupled to photons, and the coupled system we call a plasmariton (to distinguish from the polariton, a resonantly coupled T-phonon-photon entity) [9].

In this work we study the quantum theory of the L plasmon, in the following just called the plasmon. In the absence of single-electron excitations, the plasmon plays the role as the basic quantum (quasi)particle. In turn, this suggests that a first-quantized description of the plasmon might be possible and fruitful.

In Sec. III A, we establish such a description starting from the plasmon dispersion relation. The plasmon is a boson, and it satisfies a "relativistic" scalar Klein-Gordon equation, in which the light velocity c is replaced by $a = \sqrt{3/5}v_F$, v_F being the Fermi velocity of the jellium. A comparison to the dispersion relation for a relativistic boson of mass M indicates that the plasmon mass is $\hbar\omega_p/a^2$, and its Compton wave number is ω_p/a. The "rest energy" of the plasmon $Ma^2 = \hbar\omega_p$ equals the self-field energy of the scalar field. The plasmon has a charge $Q = ne < 0$ (n: electron bulk density, e: electron charge), and its first-quantized formalism thus is described by a complex Klein-Gordon field [10].

A formally ($c \rightarrow a$) covariant description enables one to extend the first-quantized theory to the second-quantized level, via a Lagrangian formalism, as described in Sec. III B. Via the relevant Euler-Lagrange equation for the free plasmon, one regains the plasmon's Klein-Gordon equation, and find the canonical plasmon momenta for the plasmon field and its complex-conjugate partner. The integral of the free-plasmon Hamiltonian density finally gives one the plasmon energy and momentum quanta $\hbar[(aq)^2 + \omega_p^2]^{1/2}$ and $\hbar\mathbf{q}$, respectively.

In Sec. III C, the plasmon's interaction with an electromagnetic gauge field is established modifying the free-plasmon Lagrangian density by the minimum coupling principle, replacing the electron charge (e) by the plasmon charge ($Q = ne$). This principle also gives one the plasmon "four"-current density. We illustrate the basic principle by an examination of the plasmon's coupling to a prescribed scalar field.

*okeller@physics.aau.dk

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In Sec. IV, we establish the first- and second-quantized theories for surface plasmons confined to, and propagating along, a sharp and flat jellium-vacuum interface. Starting from a Weyl expansion [11,12] (Secs. IV A and IV B), the surface-plasmon quasiparticle quantum theory is established in close analogy to that of the bulk plasmon (Secs. IV C and IV E). The free-surface plasmons are *eigenmodes* of the jellium-vacuum half-space, and as such not directly observable. *Resonant excitation* of surface plasmons can be achieved using the external field from a moving electron, e.g., and observing characteristic of the *p*-polarized reflection [13–15].

In Sec. V, a screened propagator formalism [16] is used to analyze the surface-plasmon excitation by a prescribed external source. In particular, we obtain from the general *p*-polarized reflection coefficient (Sec. V B) the nonretarded reflection coefficient and the Ritchie dispersion relation (Sec. V C) [17,18]. We finish Sec. V with a detailed analysis of charged-particle excitation of surface plasmons in the case where the particle propagates in vacuum with constant velocity parallel to the surface (Sec. V D). In the Appendix, we summarize how the longitudinal Lindhard dielectric function leads to the hydrodynamic dielectric function for collective plasmon excitations, and how the domain of single-particle excitations is characterized from a kinematic point of view.

II. FRAMEWORK OF THE DESCRIPTION

A. Longitudinal Lindhard dielectric function

Over the years, powerful many-body techniques have been developed to describe the response of an electron gas to an external field [2,3]. In the jellium formalism the electron moves, as mentioned in the Introduction, in a smeared-out ionic background with constant density, and the electrons interact alone via nonretarded Coulomb forces, obeying also the Pauli principle, of course.

The perhaps simplest theoretical method is the self-consistent field (SCF) approach, also called RPA, for random-phase approximation [19–22]. The derivation of the RPA (SCF) approach can be based on a density matrix (Liouville equation) approach, or on Green functions and the ring diagram technique. In the SCF approach the individual electrons behave as free particles subjected to the average potential in the system. For this work the RPA model is sufficient because it is known to lead to quantitatively correct dispersion relations for collective bulk and surface excitations, as these manifest themselves through resonant interactions with externally impressed fields. As mentioned in the Introduction, this paper is devoted to a study of first- and second-quantized theories of longitudinal (L) collective excitations, the plasmons.

The longitudinal dielectric function $\varepsilon_L(\mathbf{q},\omega)$ is a microscopic scalar bulk quantity depending on the wave number ($\mathbf{q}$) and (cyclic) frequency (ω) of the excitation. It is defined via

$$\varepsilon_0\mathbf{E}_L(\mathbf{q},\omega)+\mathbf{P}_L(\mathbf{q},\omega)=\varepsilon_L(\mathbf{q},\omega)\mathbf{E}_L(\mathbf{q},\omega), \tag{1}$$

where $\mathbf{P}_L(\mathbf{q},\omega)=P_L(\mathbf{q},\omega)\boldsymbol{\kappa}$ is the microscopic longitudinal polarization of the jellium in the $\boldsymbol{\kappa}=\mathbf{q}/q$ direction, ε_0 is the vacuum permittivity, and $\mathbf{E}_L(\mathbf{q};\omega)=E_L(\mathbf{q},\omega)\boldsymbol{\kappa}$ is the longitudinal part of the local electric field, both at frequency ω. In RPA, $\varepsilon_L(\mathbf{q},\omega)$ is given by the Lindhard dielectric function [23]

$$\varepsilon_L(\mathbf{q};\omega)=1-\lim_{\eta\to 0}\left[\frac{e^2}{\varepsilon_0\Omega}\frac{1}{q^2}\sum_{\mathbf{k}}\frac{f_0(E_{\mathbf{k}+\mathbf{q}})-f_0(E_{\mathbf{k}})}{E_{\mathbf{k}+\mathbf{q}}-E_{\mathbf{k}}-\hbar\omega-i\hbar\eta}\right], \tag{2}$$

where $E_{\mathbf{k}}$ ($E_{\mathbf{k}+\mathbf{q}}$) is the free-electron energy belonging to the wave vector $\mathbf{k}$ ($\mathbf{k}+\mathbf{q}$), $f_0(E_{\mathbf{k}})$ [$f_0(E_{\mathbf{k}+\mathbf{q}})$] is the Fermi-Dirac distribution giving the occupation probability of state $\mathbf{k}$ ($\mathbf{k}+\mathbf{q}$), and Ω is the box volume for mode quantization. A deeper contour integral analysis shows that the collective and single-particle excitations are associated with the pole and branch-cut structure of the longitudinal Lindhard dielectric function as this structure appears in a Green function approach (e.g., see Sec. V). The Green function method is of particular value (importance) for studies of surface plasmons and their excitation (Secs. IV and V). With a somewhat limited scope, sufficient for presenting the framework of this paper one may derive the bulk plasmon dispersion relation by a long-wavelength expansion of $\varepsilon_L(q,\omega)$ (to lowest order $\sim q^2$). The well-known calculation, briefly summarized in the Appendix, gives the result

$$\varepsilon_L(q,\omega)=1-\frac{\omega_p^2}{\omega^2-Dq^2}, \tag{3}$$

where $\omega_p=[ne^2/(m\varepsilon_0)]^{1/2}$ is the plasma frequency (electron density: n, electron mass: m). The quantity

$$D=\tfrac{3}{5}v_F^2, \tag{4}$$

where v_F is the electron Fermi velocity, will be of utmost importance in Secs. III and IV since $D^{1/2}$ will turn out to be a common characteristic phase velocity of the bulk and surface plasmons in our quantum theories.

B. Adjacent jellium-vacuum half-spaces: Surface plasmons

The quantum physical theory we shall develop for surface plasmons in Sec. IV is based on the perhaps simplest model for a spatially nonlocal jellium occupying a half-space. Thus, it will be assumed that the electron barrier towards the vacuum half-space is infinitely high, and that an incoming plane-wave jellium wave function when scattered specularly from the surface barrier does not interfere with its reflected part. In this so-called semiclassical infinite barrier (SCIB) model [24,25], the field-unperturbed electron density is constant up to the surface, where it drops to zero in a sharp step. Spill-out effects are thus neglected (due to the infinite barrier height), as are spill-in effects (stemming from electron interference in reflection processes).

C. Bulk and surface propagating eigenmode conditions

The first- and second-quantized theories for bulk plasmons are established starting from the eigenmode condition

$$\varepsilon_L(q,\omega)=0, \tag{5}$$

given the relevant dispersion relation $\omega=\omega(q)$. The eigenmode condition for surface plasmons, given by

$$\varepsilon_L(q_\parallel,\omega)=-1, \tag{6}$$

gives the dispersion relation for modes running along the surface plane. The wave vector of such modes along the surface is $\mathbf{q}_{\parallel}$, and they decay exponentially perpendicular to the surface plane, both in the jellium and in vacuum. The derivation of Eq. (6) contains some in-depth analyses (see Sec. IV). The conditions in Eqs. (5) and (6) lead to a uniform quantum description of bulk and surface plasmons.

III. BULK PLASMONS

In this section we establish and discuss the first- and second-quantized theory of bulk plasmons. Starting from the plasmon dispersion relation a wave equation is introduced for a new *single-particle (plasmon) field*. The plasmon is a boson particle, and in the first-quantization it satisfies a Klein-Gordon type of wave equation. A rigorous Lagrangian formalism is set up for the plasmon field, and the related Hamiltonian is quantized in the usual manner for a boson field. The plasmon's interaction with an external field is established using a minimal coupling principle.

A. First-quantized theory

1. Dispersion relation, wave equation

Like for other particles (photons, electrons, plasmaritons, magnon particles, etc.) the first-quantized theory describes a *single plasmon* (in a sense a quasiparticle) on the basis of a *quantum mechanical* formalism. In the collective regime one no longer has the electron as the basic entity. Of course, the roles of the individual electrons manifest themselves by giving rise to a damping of the plasmon field. This damping must not be included in a normal mode analysis leading to the second-quantized field formalism. The coupling of the plasmon particle to individual electrons can be studied using a diagrammatic approach (with absorption, emission, and scattering processes). It is also clear that the jellium formalism can be extended to account for ion lattice periodicity, generalizing the Lindhard formalism in a well-known manner [4]. Such a generalization allows one to investigate individual plasmon-phonon scattering processes, etc.

When the expression in Eq. (3) is inserted in the eigenmode condition in Eq. (5), one obtains the (squared) dispersion relation

$$\omega^2 = (aq)^2 + \omega_p^2, \tag{7}$$

where

$$a = D^{1/2} = \sqrt{\frac{3}{5}}v_f \tag{8}$$

is a characteristic phase velocity, of the order of the Fermi velocity. The following analysis holds also if a refined model changes the prefactor $\sqrt{3/5}$ a bit. These refined models account for the exchange and correlation hole around an electron. In the simplest extensions this amounts to a slight change in the prefactor. A detailed discussion can be found in Ref. [3]. The important point is that a is a constant of the order of the Fermi velocity. From the dispersion relation a quantum mechanical wave equation for the plasmon is obtained via the usual prescription $-i\omega \Rightarrow \partial/\partial t$ and $i\mathbf{q} \Rightarrow \nabla$. Thus, the plasmon wave function $\phi(\mathbf{r}, t)$ satisfies a Klein-Gordon type of wave equation, viz.,

$$\left(\nabla^2 - \frac{1}{a^2}\frac{\partial^2}{\partial t^2}\right)\phi(\mathbf{r}, t) - Q_C^2\phi(\mathbf{r}, t) = 0, \tag{9}$$

where a Compton wave number is defined as

$$Q_C \equiv \frac{\omega_p}{a}. \tag{10}$$

The formal similarity of Eq. (9) to the relativistic Klein-Gordon equation describing the dynamics of a boson particle is striking. The relativistic boson satisfies the famous dispersion relation

$$\omega^2 = (cq)^2 + \left(\frac{m_0c^2}{\hbar}\right)^2, \tag{11}$$

where c is the vacuum speed of light, and m_0 is the particle's rest mass. The plasmon wave equation is of course not a relativistic equation, alone for the reason that it is founded on a (special) type of solution of the Scrödinger equation.

Let us write the plasmon energy-momentum relation in the "relativistic" form

$$\hbar\omega = +[(a\hbar q)^2 + (Ma^2)^2]^{1/2}, \tag{12}$$

where

$$M \equiv \frac{\hbar\omega_p}{a^2} \tag{13}$$

is the plasmon "rest mass." In a sense, the plasmon mass concept is quite illuminating. Thus,

$$E_0 \equiv Ma^2 = \hbar\omega_p \tag{14}$$

plays the role of a self-field energy of the scalar field. This is so because the wave propagation vanishes in the long-wavelength limit ($q \to 0$). The Einstein–de Broglie–type relation between particle and wave properties in the plasmon case is for the energy $E = \hbar\omega$ and the momentum $\mathbf{p} = \hbar\mathbf{q}$, where $\mathbf{q} = q\boldsymbol{\kappa}$ is the plasmon wave vector in the direction $\boldsymbol{\kappa}$. The plasmon kinetic energy $a\hbar q$ has the "usual" form, replacing c by a. The characteristic length scale in scattering processes is the plasmon Compton wavelength

$$\Lambda_C = \frac{h}{Ma}. \tag{15}$$

2. Plasmon "four-current" density: Charge conservation

For what follows it is convenient to make use of a "formally covariant" notation. Thus, we define the "covariant" and "contravariant" derivatives

$$\left\{\frac{\partial}{\partial x^\mu}\right\} \equiv \{\partial_\mu\} \equiv \left(\frac{1}{a}\frac{\partial}{\partial t}, \nabla\right), \tag{16}$$

$$\left\{\frac{\partial}{\partial x_\mu}\right\} \equiv \{\partial^\mu\} \equiv \left(\frac{1}{a}\frac{\partial}{\partial t}, -\nabla\right), \tag{17}$$

using the "metric" signature $(1, -1, -1, -1)$. Above, we have put relevant notational names between quotation marks because the various quantities are *not* relativistically covariant. In the remaining part of our article we shall leave out the quotation marks for simplicity.

In the covariant notation the plasmon wave equation reads as

$$\left(\partial_\mu \partial^\mu + Q_C^2\right)\phi(\mathbf{r}, t) = 0. \tag{18}$$

Since the plasmon is charged (global charge = charge of all jellium electrons), we know from analogy to the relativistic Klein-Gordon equation that $\phi(\mathbf{r}, t)$ is *not* a probability amplitude, but a charge probability amplitude [9,26]. [If $\phi(\mathbf{r}, t)$ in the relativistic case describes the dynamics of negatively charged bosons, the *independent* complex-conjugate charge probability amplitude $\phi^*(\mathbf{r}, t)$ relates to the positively charged antiparticles, or vice versa.] In the jellium case there is of course no antiplasmon. However, if one extends the theory to semiconductor plasmas (InSb, GaAs, ...), a longitudinal collective state of holes in the valence band takes the role of an antiplasmon. A coupling between the plasmon and antiplasmon can be made by interaction with light, e.g., in the same manner a relativistic boson and its amplitude can be coupled via γ radiation. It is obvious also that single-particle wave packets can be formed, subjected to analogous spatial confinement limitations as for other bosons, like the photon.

The components of the (unnormalized) plasmon four-current density

$$J^\mu = i(\phi^* \partial^\mu \phi - \phi \partial^\mu \phi^*),\ \mu = 0\text{–}3 \tag{19}$$

satisfy the equation of continuity

$$\partial_\mu J^\mu = 0, \tag{20}$$

as one may realize with the help of the Klein-Gordon equation in Eq. (18). The charge of the plasmon ($Q = ne < 0$; n the jellium electron density) is given by

$$Q = i \int_V (\phi^* \partial_0 \phi - \phi \partial_0 \phi^*) d^3 r, \tag{21}$$

where V is the box volume of the spatial mode quantization.

B. Second-quantized theory

1. Lagrangian formalism for free plasmons: Hamilton density

Taking as a starting point the plasmon Lagrangian density (this Lagrangian density follows from the well-known relativistic Klein-Gordon density [10] replacing covariant derivatives by our formal covariant derivatives)

$$\mathcal{L}_P = (\partial^\mu \phi)(\partial_\mu \phi^*) - Q_C^2 \phi \phi^* \tag{22}$$

for a free particle (subscript P for particle or plasmon), the Euler-Lagrange equation for ϕ^*, viz.,

$$\partial_\mu \left[\frac{\partial \mathcal{L}_P}{\partial(\partial_\mu \phi^*)} \right] - \frac{\partial \mathcal{L}_P}{\partial \phi^*} = 0, \tag{23}$$

immediately gives the Klein-Gordon equation in Eq. (18). The second quantization of the plasmon field is based on the plasma Hamiltonian density. From the canonical momenta π_ϕ and π_{ϕ^*} for ϕ and ϕ^*, i.e.,

$$\pi_\phi \equiv \frac{\partial \mathcal{L}_P}{\partial(\partial_0 \phi^*)} = \partial^0 \phi, \tag{24}$$

$$\pi_\phi^* \equiv \frac{\partial \mathcal{L}_P}{\partial(\partial_0 \phi)} = \partial^0 \phi^*, \tag{25}$$

one obtains for the Hamiltonian density $\mathcal{H}_P$, given by

$$\mathcal{H}_P = \pi_\phi \dot{\phi}^* + \pi_{\phi^*} \dot{\phi} - \mathcal{L}_P \tag{26}$$

(with the abbreviation $\dot{\phi} \equiv \partial_0 \phi [= \partial^0 \phi]$), the explicit expression

$$\mathcal{H}_P = (\partial^0 \phi)(\partial_0 \phi^*) + (\nabla \phi) \cdot (\nabla \phi^*) + Q_C^2 \phi \phi^* \tag{27}$$

since $\pi_\phi \dot{\phi} = \pi_{\phi^*} \dot{\phi} = (\partial_0 \phi)(\partial_0 \phi^*)$. The total Hamiltonian (H_P) for the plasmon is obtained by integration of $\mathcal{H}_P$ over V:

$$H_P = \int_V \mathcal{H}_P(\mathbf{r}, t) d^3 r. \tag{28}$$

2. Plasmon particle operators for energy and momentum and their eigenvalues

The form given for the plasmon $\mathcal{H}_P$ clearly allows a quantization procedure as for other massive boson fields. The quantization becomes particularly easy if one rewrites the expression for H_P in the alternative form

$$\begin{aligned} H_P &= \int_V \left[(\partial^0 \phi)(\partial_0 \phi^*) - \phi^* \left(\nabla^2 \phi - Q_C^2 \phi\right)\right] d^3 r \\ &= \int_V [(\partial_0 \phi)(\partial^0 \phi^*) - \phi^* \partial_0 \partial^0 \phi] d^3 r. \end{aligned} \tag{29}$$

The form of the first member of Eq. (29) is obtained integrating the term $(\nabla \phi) \cdot (\nabla \phi^*)$ by parts (dropping the surface terms). The second member follows by utilizing the Klein-Gordon equation. The plasmon Hamilton operator $\hat{H}_P$ is obtained by upgrading the fields ϕ and ϕ^* to operator status, i.e., $\phi \Rightarrow \hat{\phi}$ and $\phi^* \Rightarrow \hat{\phi}^\dagger$. If one hereafter expands these operators in a complete monochromatic plane-wave set, one obtains

$$\hat{\phi}(\mathbf{r}, t) = \sum_n [\hat{a}_n \phi_n^{(+)}(\mathbf{r}, t) + \hat{b}_n^\dagger \phi_{-n}^{(-)}(\mathbf{r}, t)], \tag{30}$$

$$\hat{\phi}^\dagger(\mathbf{r}, t) = \sum_n \{\hat{a}_n^\dagger [\phi_n^{(+)}(\mathbf{r}, t)]^* + \hat{b}_n [\phi_{-n}^{(-)}(\mathbf{r}, t)]^*\}, \tag{31}$$

where the normalized (running) plasmon mode functions are

$$\phi_n^{(\pm)}(\mathbf{r}, t) = \left(\frac{\hbar a}{2 E_n V}\right)^{1/2} \exp\left[i\left(\mathbf{q}_n \cdot \mathbf{r} \mp \frac{E_n}{\hbar} t\right)\right] \tag{32}$$

with energies

$$E_n = \hbar \omega_n = \hbar a \left[q_n^2 + Q_C^2\right]^{1/2}. \tag{33}$$

We have retained the antiplasmon (hole) part for later applications to semiconductor plasmon theory. The plasmon (a operators) and antiplasmon (b operators) annihilation ($\hat{a}_n, \hat{b}_n$) and creation ($\hat{a}_n^\dagger, \hat{b}_n^\dagger$) operators satisfy the boson commutation relations

$$[\hat{a}_n, \hat{a}_m^\dagger] = [\hat{b}_n, \hat{b}_m^\dagger] = \delta_{nm}, \tag{34}$$

all other commutators being zero. By inserting the mode expansion in Eqs. (30) and (31) into Eq. (29) and using the commutation rules one obtains

$$\hat{H}_P = \sum_n \hbar \omega_n [\hat{a}_n^\dagger \hat{a}_n + \hat{b}_n^\dagger \hat{b}_n + 1]. \tag{35}$$

Leaving out the antiplasmon part (jellium system) the plasmon energy and momentum operators are

$$\hat{H}_P = \sum_{\mathbf{q}} \hbar\left[(aq)^2 + \omega_p^2\right]^{1/2}\left(\hat{N}(\mathbf{q}) + \frac{1}{2}\right) \tag{36}$$

and

$$\hat{\mathbf{P}}_P = \sum_{\mathbf{q}} \hbar\mathbf{q}\hat{N}(\mathbf{q}), \tag{37}$$

where

$$\hat{N}(\mathbf{q}) = \hat{a}^\dagger(\mathbf{q})\hat{a}(\mathbf{q}) \tag{38}$$

is the number operator for mode $\mathbf{q}$. In the number state $|n_{\mathbf{q}}\rangle$ (for a single mode $\mathbf{q}$) we thus obtain the eigenvalue equations

$$\hat{H}_P(\mathbf{q})\,|n_{\mathbf{q}}\rangle = \hbar\left[(aq_n)^2 + \omega_p^2\right]^{1/2}\left[n_{\mathbf{q}} + \frac{1}{2}\right]|n_{\mathbf{q}}\rangle\,, \tag{39}$$

$$\hat{\mathbf{P}}_P(\mathbf{q})\,|n_{\mathbf{q}}\rangle = \hbar\mathbf{q}_n\,|n_{\mathbf{q}}\rangle\,, \tag{40}$$

where $n_{\mathbf{q}}$ is the eigenvalue of the number operator. The expressions for the plane-wave plasmon eigenvalues, which in our paper have been derived in a rigorous manner, may perhaps have been guessed. However, our formalism opens the doorway for constructing and studying coherent states, squeezed states, entangled states, single-plasmon wave-packet states, two-plasmon states, and more.

C. Plasmon interaction with electromagnetic gauge fields

1. *Minimal coupling principle for the plasmon: Interaction Lagrangian*

In order to calculate the plasmon's interaction with an electromagnetic field, described by the four-potential $\{A_\mu\} = (A_0, \mathbf{A})$, we shall make use of minimal coupling, replacing the charge (e) by the plasmon charge ($Q = ne$). The potential of the electromagnetic field is a *relativistic four-vector* in the fundamental sense. Thus,

$$\{A^\mu\} = \left\{A^0 = \frac{\Phi}{c}, \mathbf{A}\right\}, \tag{41}$$

where Φ is the scalar potential. The minimal coupling substitution we give the plasmon form [27]

$$\partial_\mu \Rightarrow \partial_\mu - \frac{iQ}{\hbar}A_\mu, \tag{42}$$

where it must be remembered that $\{\partial_\mu\}$ is the plasmon covariant derivative [Eq. (16)]. In our second article on the plasmariton, which always is a coupled system of transverse (T) plasma displacements and an electromagnetic field, all covariant and (contravariant) derivatives will be genuine relativistic four-vectors: the plasmariton propagates with the vacuum speed of light (as phase velocity).

The minimal coupling substitution in Eq. (42) changes the Lagrangian density in Eq. (22) to $\mathcal{L}_P + \mathcal{L}_I$:

$$\mathcal{L}_P + \mathcal{L}_I = \left(\partial^\mu\phi + \frac{iQ}{\hbar}A^\mu\phi\right)\left(\partial_\mu\phi^* - \frac{iQ}{\hbar}A_\mu\phi^*\right) - Q_C^2\phi\phi^*, \tag{43}$$

which shows that the interaction Lagrangian density ($\mathcal{L}_I$) is given by

$$\mathcal{L}_I = \frac{iQ}{\hbar}[(A^\mu\phi)(\partial_\mu\phi^*) - (\partial^\mu\phi)(A_\mu\phi^*)] + \left(\frac{Q}{\hbar}\right)^2 A^\mu A_\mu\phi\phi^*. \tag{44}$$

The last term of $\mathcal{L}_I$ describes a nonlinear coupling between the plasmon and the electromagnetic field. The total Lagrangian density ($\mathcal{L}$), which we do not need in this work, is obtained by adding the free-field electromagnetic Lagrangian density

$$\mathcal{L}_F = \frac{\varepsilon_0}{2}\left[\left(\frac{\partial\mathbf{A}}{\partial t}\right)^2 - c^2(\nabla\times\mathbf{A})^2\right], \tag{45}$$

that is,

$$\mathcal{L} = \mathcal{L}_P + \mathcal{L}_F + \mathcal{L}_I. \tag{46}$$

2. *Plasmon current density: Interaction Hamiltonian*

In the presence of an electromagnetic field the minimal coupling principle leads to a plasmon current density

$$J^\mu = i\left[\phi^*\left(\partial^\mu\phi + i\frac{Q}{\hbar}A^\mu\phi\right) - \phi\left(\partial^\mu\phi^* - i\frac{Q}{\hbar}A^\mu\phi^*\right)\right],\ \mu = 0\text{–}3 \tag{47}$$

which we divide into "paramagnetic" $[J^\mu_{\text{para}}(A^\mu = 0)]$ and "diamagnetic" $[J^\mu_{\text{dia}}(A^\mu)]$ parts, viz.,

$$J^\mu_{\text{para}}(A^\mu = 0) = i(\phi^*\partial^\mu\phi - \phi\partial^\mu\phi^*),\ \mu = 0\text{–}3 \tag{48}$$

and

$$J^\mu_{\text{dia}}(A^\mu) = -\frac{2Q}{\hbar}A^\mu\phi\phi^*, \quad \mu = 0\text{–}3. \tag{49}$$

The *form* of the paramagnetic part is the same as that of Eq. (19), but ϕ must now be calculated from an inhomogeneous plasmon Klein-Gordon equation; see Sec. III C 3.

With coupling present the canonical momenta become

$$\pi_\phi = \frac{\partial(\mathcal{L}_P + \mathcal{L}_I)}{\partial(\partial_0\phi^*)} = \partial^0\phi + \frac{iQ}{\hbar}A^0\phi, \tag{50}$$

$$\pi_{\phi^*} = \frac{\partial(\mathcal{L}_P + \mathcal{L}_I)}{\partial(\partial_0\phi)} = \partial^0\phi^* - \frac{iQ}{\hbar}A^0\phi^*. \tag{51}$$

From these the sum of the plasmon ($\mathcal{H}_P$) and interaction ($\mathcal{H}_I$) Hamiltonian densities

$$\mathcal{H}_P + \mathcal{H}_I = \pi_\phi\dot{\phi}^* + \pi_{\phi^*}\dot{\phi} - \mathcal{L}_P - \mathcal{L}_I \tag{52}$$

is obtained:

$$\mathcal{H}_P + \mathcal{H}_I = (\partial^0\phi)(\partial_0\phi^*) + (\nabla\phi)\cdot(\nabla\phi^*) + Q_C^2\phi\phi^* + \frac{iQ}{\hbar}A^0(\phi\dot{\phi}^* - \phi^*\dot{\phi}) + \frac{iQ}{\hbar}[(\partial^\mu\phi)(A_\mu\phi^*) - (\partial^\mu\phi^*)(A^\mu\phi)] - \left(\frac{Q}{\hbar}\right)^2 A_\mu A^\mu\phi\phi^*, \tag{53}$$

and then finally

$$\mathcal{H}_P + \mathcal{H}_I = (\partial^0\phi)(\partial_0\phi^*) + (\nabla\phi)\cdot(\nabla\phi^*) + Q_C^2\phi\phi^* + \frac{iQ}{\hbar}[(\mathbf{A}\phi)\cdot(\nabla\phi^*) + (\mathbf{A}\phi^*)\cdot(\nabla\phi)] - \left(\frac{Q}{\hbar}\right)^2 A_\mu A^\mu \phi\phi^*. \quad (54)$$

The reader may check that the "standard" form of the interaction Hamiltonian density [28], viz., $-J^\mu A_\mu$ deviates from the one in Eq. (54) by a term $i(Q/\hbar)[\phi^*\partial^0\phi - \phi\partial^0\phi^*]A_0$.

3. Inhomogeneous plasmon Klein-Gordon equation

It is often claimed that plasmons do not couple to the (transverse) photon field. This is certainly only correct in special cases, as we shall realize in Sec. V, where the plasmon's coupling to the field from an externally impressed prescribed current source is analyzed. The coupling also appears in a manifest manner from the Euler-Lagrange equation

$$\partial_\mu\left[\frac{\partial(\mathcal{L}_P + \mathcal{L}_I)}{\partial(\partial_\mu\phi^*)}\right] - \frac{\partial(\mathcal{L}_P + \mathcal{L}_I)}{\partial\phi^*} = 0, \quad (55)$$

from which one obtains the following inhomogeneous Klein-Gordon equation for the scalar field:

$$\left(\partial_\mu\partial^\mu + Q_C^2\right)\phi + \frac{iQ}{\hbar}[\partial_\mu(A^\mu\phi) + A_\mu(\partial^\mu\phi)] - \left(\frac{Q}{\hbar}\right)^2 A_\mu A^\mu\phi = 0. \quad (56)$$

Let us close this subsection by a brief account of a most simple example, viz., the interaction of the plasmon with a space-time constant scalar potential Φ. Equation (56) now is simplified to

$$\left(\partial_\mu\partial^\mu + Q_C^2\right)\phi + \frac{2iQ\Phi}{\hbar c}\frac{\partial\phi}{\partial t} + \left(\frac{Q\Phi}{\hbar}\right)^2\phi = 0 \quad (57)$$

since $\partial_0\phi = \partial^0\phi$ [metric signature $(1, -1, -1, -1)$]. From Eq. (57) one obtains a dispersion relation

$$\omega = \frac{a^2}{\hbar c}Q\Phi + \left\{(aq)^2 + \omega_p^2 + \left(\frac{aQ\Phi}{\hbar}\right)^2\left[1 + \left(\frac{a}{c}\right)^2\right]\right\}^{1/2}. \quad (58)$$

Since $a/c \ll 1$, the dispersion relation may ($\sim$ always) be reduced to

$$\omega \simeq \left[(aq)^2 + \omega_p^2 + \left(\frac{aQ\Phi}{\hbar}\right)^2\right]^{1/2}. \quad (59)$$

The term $(aQ\Phi/\hbar)^2$ originates in the nonlinear part of the coupling, and this displaces the plasma frequency to

$$\omega_p(\Phi) = \omega_p(\Phi = 0)\left[1 + \left(\frac{Q\Phi}{\hbar Q_C}\right)^2\right]^{1/2}. \quad (60)$$

One may conclude that it is a simultaneous two-photon scalar photon interaction with the plasmon which is responsible for the upwards displacement $[\omega_p(\Phi) > \omega_p(\Phi = 0)]$ of the plasma frequency. In Sec. IV E 2 we derive a closely related dispersion relation for surface plasmons.

IV. SURFACE PLASMONS

A. Weyl expansion of the bulk wave function

We begin our study of the first-quantized theory of surface plasmons by establishing a Weyl expansion of the bulk plasmon wave function in the space-frequency domain $\phi(\mathbf{r};\omega)$. The Weyl expansion we obtain in a sense represents a generalization of the well-known Weyl expansion for electromagnetic fields in vacuum [12]. The expansion derived below holds for relativistic boson fields if one makes the replacements $a \to c$ and $\omega_p \to m_0c^2/\hbar$.

It appears from the dispersion relation in Eq. (7) that $\phi(\mathbf{r};\omega)$ satisfies the Helmholtz equation

$$\left(\nabla^2 + \kappa_L^2\right)\phi(\mathbf{r};\omega) = 0, \quad (61)$$

where

$$\kappa_L = \frac{1}{a}\left(\omega^2 - \omega_p^2\right)^{1/2}. \quad (62)$$

Let us then assume that in any plane $z =$ constant the field may be represented by a two-dimensional (2D) Fourier integral of the form

$$\phi(\mathbf{r};\omega) = \int_{-\infty}^{\infty}\phi(\mathbf{q}_\parallel, \omega; z)e^{i\mathbf{q}_\parallel\cdot\mathbf{r}_\parallel}d^2q_\parallel, \quad (63)$$

where $\mathbf{q}_\parallel = (q_{\parallel,x}, q_{\parallel,y}, 0)$ and $\mathbf{r}_\parallel = (x, y, 0)$. It must be remembered that the collective wave number from a physical point of view must be limited to small wave numbers to avoid single-particle excitations [2,23]. The domain of single-particle excitations is briefly discussed in the Appendix. By inserting Eq. (63) into Eq. (61) one has

$$\int_{-\infty}^{\infty}\left(\kappa_L^2 - q_\parallel^2 + \frac{\partial^2}{\partial z^2}\right)\phi(\mathbf{q}_\parallel, \omega; z)e^{i\mathbf{q}_\parallel\cdot\mathbf{r}_\parallel}d^2q_\parallel = 0. \quad (64)$$

Since Eq. (64) must hold for all values of $\mathbf{r}_\parallel$, it appears that $\phi(\mathbf{r}_\parallel, z)$ satisfies the second-order differential equation

$$\left(\frac{\partial^2}{\partial z^2} + w^2\right)\phi(\mathbf{q}_\parallel, \omega, z) = 0, \quad (65)$$

where

$$w = \begin{cases} +\left(\kappa_L^2 - q_\parallel^2\right)^{1/2}, & q_\parallel \leqslant \kappa_L \\ +i\left(q_\parallel - \kappa_L^2\right)^{1/2}, & q_\parallel > \kappa_L. \end{cases} \quad (66)$$

The general solution to Eq. (65) thus is

$$\phi(\mathbf{q}_\parallel, \omega; z) = A(\mathbf{q}_\parallel, \omega)e^{iwz} + B(\mathbf{q}_\parallel, \omega)e^{-iwz}. \quad (67)$$

This in turn means that the bulk plasmon wave function has an expansion (over a chosen $q_\parallel$ plane)

$$\phi(\mathbf{r};\omega) = \int_{-\infty}^{\infty}A(\mathbf{q}_\parallel, \omega)e^{i(\mathbf{q}_\parallel\cdot\mathbf{r}_\parallel + wz)}d^2q_\parallel + \int_{-\infty}^{\infty}B(\mathbf{q}_\parallel, \omega)e^{i(\mathbf{q}_\parallel\cdot\mathbf{r}_\parallel - wz)}d^2q_\parallel. \quad (68)$$

The general solution for $\phi(\mathbf{r};\omega)$ thus consists of a superposition of three-dimensional (3D) *propagating plane waves* ($q_\parallel \leqslant \kappa_L$) and so-called inhomogeneous plane modes propagating along the given $\mathbf{q}_\parallel$ directions and *evanescent* (exponentially decaying) in the direction perpendicular to the $\mathbf{q}_\parallel$ plane ($q_\parallel > \kappa_L$). The expansion in Eq. (68), with ω given by Eq. (66), is the renowned Weyl expansion. By combining Eqs. (62) and (66) (for $q_\parallel > \kappa_L$) it follows that the one-dimensional (1D) evanescent components must satisfy the condition

$$(aq_\parallel)^2 + \omega_p^2 - \omega^2 > 0. \quad (69)$$

As we shall see in Sec. IV B, the surface plasmons are related to the 1D evanescent modes. Modes which are evanescent for all $|\mathbf{q}_\parallel|$, hence must have frequencies below ω_p. Wave-packet surface-plasmon wave functions can be constructed by relevant $\mathbf{q}_\parallel$ superposition of 1D evanescent modes. The theoretical limitation on how well localized a given wave packet can be is determined by the cutoff condition for the evanescent modes [inequality in Eq. (69)].

B. Surface plasmons on a sharp and flat jellium-vacuum interface

In the so-called semiclassical infinite-barrier model (SCIB model) it is assumed that the individual jellium electrons hiding the surface are (i) specularly reflected, that (ii) quantum interference between the incoming and reflected parts of the electron can be neglected, and that (iii) the sharp smeared-out ion barrier is infinitely high. This model has it limitations in that selvedge effects [Friedel-type spill out and spill in (for infinitely high barrier)] are omitted [24,29]. A Green function approach allows a study of selvedge effects for surface plasmons and surface plasmaritons [30]. In the SCIB model the relevant spatially nonlocal microscopic conductivity tensor $\boldsymbol{\sigma}(z, z'; \mathbf{q}_\parallel, \omega)$ is known [25] to have elements (leaving out the reference to $\mathbf{q}_\parallel$ and ω)

$$\sigma_{ij}(z, z') = \Theta(z)\Theta(z')\left[\sigma_{ij}^\infty(z - z') + \xi_j\sigma_{ij}^\infty(z + z')\right], \quad (70)$$

where $\xi_j = 1$ for $j = x, y$ and $\xi_j = -1$ for $j = z$. Θ denotes the Heaviside unit step function. Above, we assume that the surface is placed at the $z = 0$ plane. $\boldsymbol{\sigma}^\infty$ is the microscopic conductivity tensor of an infinitely extended jellium. $\boldsymbol{\sigma}^\infty$ contains both longitudinal (L) (relevant for bulk plasmons)and transverse (T) parts. As we shall see in Sec. V, both parts are of importance in resonance excitation of surface plasmons. The implications of the text accompany Fig. 1. As discussed in the text to Fig. 1 the SCIB model makes the jellium behave like an infinite medium with a sheet source at $z = 0$.

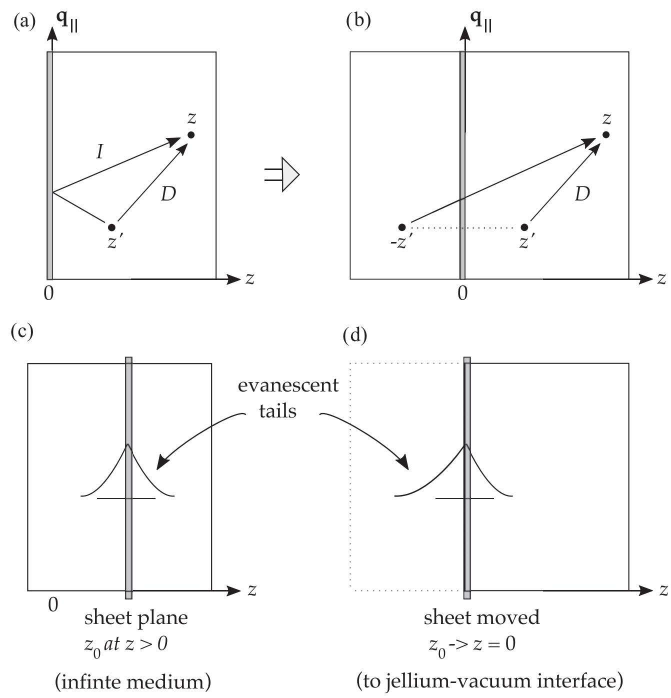

FIG. 1. Schematic illustrations of the SCIB model and the Weyl expansion used in the establishment of the surface-plasmon dispersion relation. In (a) it is shown how the radiation from a source point at a distance z' from the surface is connected via direct (D) and indirect (I) field propagation to an observation point at z. The neglect of quantum interference in electron reflection from the surface allows one to consider the semi-infinite jellium as in infinitely extended medium in which [as shown in (b)] the indirect field propagation appears to originate from the mirror point at (depth) $-z'$. For the equivalent infinite jellium the Weyl expansion of the bulk plasmon field $\phi(\mathbf{q}_\parallel, \omega, z)$ from a sheet source located at depth z_0 is shown in (c). As indicated, the field decays exponentially with distance from z_0 (the decay constant is the same for $z < z_0$ and $z > z_0$). To describe the surface-plasmon field, the sheet is moved from z_0 to the interface at $z = 0$ (d). The decay constant now is smaller for $z < 0$ than for $z > 0$ [cf. Eqs. (62) and (65)].

C. Dispersion relation

The surface-plasmon dispersion relation now can be deduced as follows: The bulk plasmon dispersion relation originates in the nonretarded Coulomb interaction in the *entire* space. At long wavelength ($q \to 0$) the classical energy in the jellium mode is proportional to ω_p^2. For a half-space ($z > 0$), one thus expects a jellium energy proportional to $\omega_p^2/2$ in the framework of the SCIB model (cf. Fig. 1). In consequence, we must have a (squared) dispersion relation for surface plasmons of the form

$$\omega^2 = (aq_\parallel)^2 + \omega_{pS}^2, \quad (71)$$

where

$$\omega_{pS} = \frac{\omega_p}{\sqrt{2}} \quad (72)$$

is the (well-known) surface-plasmon frequency. A rigorous derivation of Eq. (71) can be found in Ref. [13], Chap. 1. Equation (71) is identical to Eq. (6). Note that we have reached this dispersion relation (essentially) on the basis of the SCIB model applied to longitudinal collective electron dynamics in bulk jellium. As we shall see in Sec. V, more "complicated" (in our point of view) surface-plasmon dispersion relations have been suggested in the literature [13,17,18]. All these are established from resonance studies, not giving a quite clear picture of the basics, and not of the "simple oscillator" type, a direct starting point for the first- and second-quantized theories for surface plasmons (at sharp jellium and vacuum interfaces). The boson character of the surface plasmons is obvious from the form in Eq. (71). We have in Eq. (71) used the same $a \sim v_F$ as for bulk plasmons (consistent with the SCIB model). Refined models may (and probably will) change the prefactor $(3/5)^{1/2}$ [in Eq. (8)] slightly.

D. First-quantized theory

At this point, it is clear that a first-quantized theory for surface plasmons can be established in analogy that given for bulk plasmons in Sec. III A. The wave function of the surface (S) plasmon, denoted from now on as $\phi_S(\mathbf{r}_\parallel, t)$, satisfies the 2D Klein-Gordon equation (prescription: $i\mathbf{q}_\parallel \Rightarrow \nabla_\parallel$)

$$\left(\nabla_\parallel^2 - \frac{1}{a^2}\frac{\partial^2}{\partial t^2}\right)\phi_S(\mathbf{r}_\parallel, t) - Q_{CS}^2\phi_S(\mathbf{r}_\parallel, t) = 0, \tag{73}$$

where

$$Q_{CS} \equiv \frac{\omega_{pS}}{a} \tag{74}$$

is a Compton wave number for surface plasmons, and $\nabla_\parallel = (\frac{\partial}{\partial x}, \frac{\partial}{\partial y}, 0)$ in Cartesian coordinates. With a surface-plasmon "rest mass"

$$M_S = \frac{\hbar\omega_{pS}}{a^2}, \tag{75}$$

one readily obtains the "relativistic" energy-momentum relation

$$\hbar\omega = +[(a\hbar q_\parallel)^2 + (M_S a^2)^2]^{1/2}. \tag{76}$$

The plasmon self-field energy now is $E_{0S} = M_S a^2 = \hbar\omega_{pS}$.

In a formally 3D covariant notation where

$$\{\partial_\mu\} = \left(\frac{1}{a}\frac{\partial}{\partial t}, \nabla_\parallel\right) [\equiv \partial_{\parallel,\mu}], \tag{77}$$

etc., the surface-plasmon wave equation is written as

$$(\partial_\mu\partial^\mu + Q_{CS}^2)\phi_S(\mathbf{r}_\parallel, t) = 0. \tag{78}$$

In the specular reflection mode, Eq. (70) implies that

$$J_z(z \to 0^+; q_\parallel, \omega) = 0, \quad \frac{d}{dz}\mathbf{J}_\parallel(z \to 0^+; q_\parallel, \omega) = 0. \tag{79}$$

The results in Eq. (79) follow from the spatially nonlocal constitutive equation between field and current density; see, e.g., Refs. [16,22,25]. In turn this means that the spatial part of the contravariant surface current density

$$J^\mu = i(\phi_S^*\partial^\mu\phi_S - \phi_S\partial^\mu\phi^*), \quad \mu = 0\text{–}2 \tag{80}$$

lies in the $z = 0$ plan. The wave equation in Eq. (73) implies that the equation of continuity $\partial_\mu J^\mu = 0$ is satisfied in the surface-plasmon case. The charge of the surface plasmon is $Q_S = n_S e (< 0)$, where n_S is the surface electron density. To obtain n_S a model for the surface region is needed. Basically, a complicated integral equation study aiming at a self-consistent determination of the microscopic electromagnetic field in the selvedge region is needed. Only approximate solutions can be obtained, and in many applications simpler models have been employed (see Ref. [30], and references therein). At low frequencies density functional theory can be used to get the charge and potential distributions in the selvedge region [31–33].

E. Second-quantized theory

1. Free surface plasmons

Starting from the surface-plasmon Lagrangian density ($\mu = 0\text{–}2$)

$$\mathcal{L}_{pS} = (\partial^\mu\phi_S)(\partial_\mu\phi_S^*) - Q_{CS}^2\phi_S\phi_S^*, \tag{81}$$

one may repeat all the steps carried out in Sec. III B, and if one leaves out the anti-surface-plasmon part, the surface plasmon's energy and momentum operators take the following form in plane-wave ($\mathbf{q}_\parallel$) mode quantization:

$$\hat{H}_{pS} = \sum_{\mathbf{q}_\parallel}\left\{\hbar\left[(aq_\parallel)^2 + \omega_{pS}^2\right]^{1/2}\left(\hat{N}_S(\mathbf{q}_\parallel) + \frac{1}{2}\right)\right\}, \tag{82}$$

$$\hat{\mathbf{P}}_{pS} = \sum_{\mathbf{q}_\parallel}\hbar\mathbf{q}_\parallel\hat{N}_S(\mathbf{q}_\parallel), \tag{83}$$

where

$$\hat{N}_S(\mathbf{q}_\parallel) = \hat{a}^\dagger(\mathbf{q}_\parallel)\hat{a}(\mathbf{q}_\parallel) \tag{84}$$

is the surface-plasmon number operator. The surface annihilation and creation operators satisfy boson commutation relations, that is,

$$[\hat{a}(\mathbf{q}_{\parallel,n}), \hat{a}^\dagger(\mathbf{q}_{\parallel,m})] = \delta_{nm}, \tag{85}$$

all other commutators being zero.

2. Surface-plasmon interaction with electromagnetic gauge fields

In the minimal coupling scheme the charge of the surface plasmon Q_S enters the plasmon Lagrangian ($\mathcal{L}_{pS}$) formalism. With the substitution

$$\partial_\mu \Rightarrow \partial_\mu - \frac{iQ_S}{\hbar}A_\mu, \quad \mu = 0\text{–}2 \tag{86}$$

the Lagrangian including the interaction with the potential field is

$$\mathcal{L}_{pS} + \mathcal{L}_{IS} = \left(\partial^\mu\phi_S + \frac{iQ_S}{\hbar}A^\mu\phi_S\right)\left(\partial_\mu\phi_S^* - \frac{iQ_S}{\hbar}A_\mu\phi_S^*\right) - Q_{CS}^2\phi_S\phi_S^*. \tag{87}$$

From the Euler-Lagrange equation

$$\partial_\mu\left[\frac{\partial(\mathcal{L}_{pS} + \mathcal{L}_{IS})}{\partial(\partial_\mu\phi_S^*)}\right] - \frac{\partial(\mathcal{L}_{pS} + \mathcal{L}_{IS})}{\partial\phi_S^*} = 0, \tag{88}$$

the inhomogeneous surface-plasmon Klein-Gordon equation is obtained:

$$(\partial_\mu\partial^\mu + Q_{CS}^2)\phi_S = \frac{iQ_S}{\hbar}[\partial_\mu(A^\mu\phi_S) + A_\mu(\partial^\mu\phi_S)] - \left(\frac{Q_S}{\hbar}\right)^2 A_\mu A^\mu\phi_S = 0. \tag{89}$$

A number of specific cases can be derived from Eq. (89). For instance, for coupling to a constant scalar potential (Φ), one ends up with a dispersion relation of the form given by Eq. (59), with the replacement $Q \to Q_S$, $q \to q_\parallel$, and $\omega_p \to \omega_{pS}$. Hence, with the abbreviation

$$x = \frac{aQ_S\Phi}{\hbar\omega_{pS}}, \tag{90}$$

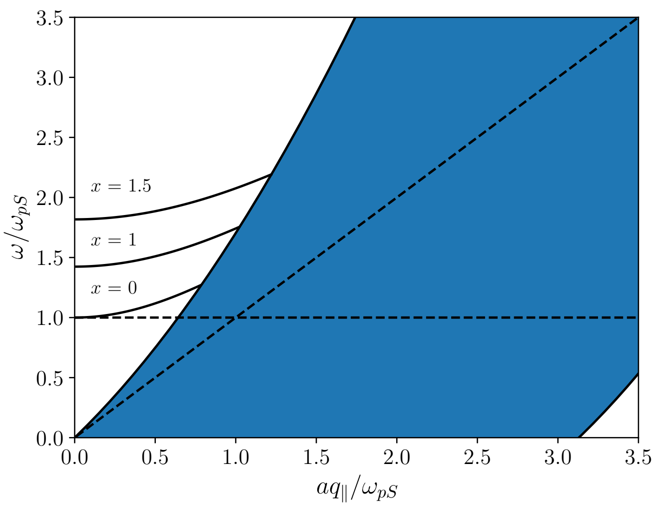

FIG. 2. The surface-plasmon dispersion relation in Eq. (91) plotted for three different values of $x = aQ_S\Phi/(\hbar\omega_{pS})$. The blue region is the domain of single-particle excitations (see the Appendix for a brief summary). The surface plasmons are strongly damped when single-particle excitations occur. The dotted lines have slope a/ω_p and 0.

one has

$$\frac{\omega}{\omega_{pS}} = \left[\left(\frac{aq_\parallel}{\omega_{pS}}\right) + 1 + x^2\right]^{1/2}. \tag{91}$$

A plot of the dispersion relations in Eq. (91) for different x values is shown in Fig. 2.

V. EXCITATION BY EXTERNAL SOURCE: SCREENED PROPAGATOR FORMALISM

A. Jellium screened propagator

Having established the quantum theory of surface plasmons at a flat jellium-vacuum interface we address the question of how these plasmons are coupled to a prescribed electromagnetic field originating in an external current density distribution (source) $\mathbf{J}_0(\mathbf{r};\omega)$. Under the assumption that the source is located outside the jellium half-space the electric field in points $(\mathbf{r})$ in the vacuum half-space outside the source domain (V_0) can be written in the compact form

$$\mathbf{E}(\mathbf{r};\omega) = -i\mu_0\omega\int_{V_0}\mathbf{G}(\mathbf{r},\mathbf{r}';\omega)\cdot\mathbf{J}_0(\mathbf{r}';\omega)d^3r' \tag{92}$$

in the space-frequency domain. The central quantity in Eq. (92) is the tensorial electromagnetic propagator $\mathbf{G}(\mathbf{r},\mathbf{r}';\omega)$ [16,34]. This consists of two parts

$$\mathbf{G}(\mathbf{r},\mathbf{r}';\omega) = \mathbf{G}_0(\mathbf{r}-\mathbf{r}';\omega) + \mathbf{I}(\mathbf{r},\mathbf{r}';\omega), \tag{93}$$

where $\mathbf{G}_0(\mathbf{r}-\mathbf{r}';\omega)$ describes the *direct* field propagation between source $(\mathbf{r}')$ and observation $(\mathbf{r})$ points. As indicated, this part only depends on the difference $\mathbf{r}-\mathbf{r}'$ (vacuum is homogeneous) and $\mathbf{I}(\mathbf{r},\mathbf{r}';\omega)$ accounts for the *indirect* propagation from $\mathbf{r}'$ to $\mathbf{r}$ via reflection from the jellium surface as shown schematically in Fig. 3.

The propagator $\mathbf{G}(\mathbf{r},\mathbf{r}';\omega)$ is constructed in such a manner that Eq. (92) also can describe "external" sources located

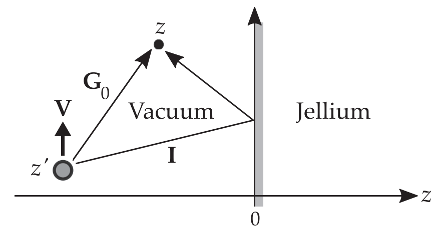

FIG. 3. A charged particle moving with a constant velocity $\mathbf{V}$ parallel to the vacuum-jellium interface can excite transverse (T) and longitudinal (L) surface plasmons via p-polarized field reflection from the surface. The field from the source plane z' reaches the plane of observation z through direct (propagator: $\mathbf{G}_0$) and indirect (propagator: $\mathbf{I}$) channels. The $\mathbf{I}$ channel contains information on the T- and L-surface plasmons.

inside the jellium, provided the dynamics of these (to a good approximation) develop independently of the plasma screening or can be described in a simple relaxation time approach. Examples may be a fast going charged particle (∼ion, electron, ...) or a (laser-driven) impurity atom (∼oscillating electric dipole). External sources inside the jellium can couple (strongly) to bulk plasmons.

In Eq. (92) we have considered the electromagnetic field as a classical quantity. However, it is possible to extend the formalism to a quantum mechanical (first-quantized) photon theory based on, e.g., the Riemann-Silberstein-Oppenheimer-Bialynicki energy wave function [35,36] driven by an external source [37]. The first-quantized theory can be extended to second quantization (low-energy QED). In a related problem, viz., the scattering from a Bethe hole in a thin jellium screen, first- and second-quantized theories have been established in recent years (see [38,39] and references therein). In these studies the (quantized) plasmon response was not considered explicitly.

It is known [16,34] that the indirect propagator has a Weyl-type expansion of the general form

$$\mathbf{I}(\mathbf{r},\mathbf{r}';\omega) = (2\pi)^{-2}\int_{-\infty}^{\infty}\mathbf{S}^{-1}\cdot\mathbf{I}(z+z';q_\parallel,\omega)\cdot\mathbf{S} \times \exp[i\mathbf{q}_\parallel\cdot(\mathbf{r}_\parallel-\mathbf{r}'_\parallel)]d^2q_\parallel, \tag{94}$$

where $\mathbf{S}$ is the rotation matrix (for rotations around the $q_{\parallel,z}$ axis) which takes $(q_{\parallel,x}, q_{\parallel,z}, 0)$ into $(q_\parallel, 0, 0)$.

B. *p*-polarized reflection

If the source is a pointlike atom radiating as an electric dipole, or a charged particle propagating along the surface, it is necessary to take into account the rotation of $\mathbf{I}(z+z';q_\parallel,\omega)$, integrating $\mathbf{S}^{-1}\cdot\mathbf{I}(z+z';q_\parallel,\omega)\cdot\mathbf{S}$ over the entire $q_\parallel$ plane. This integration gives contributions from both the propagating and evanescent parts of the wave-vector $(\mathbf{q}_\parallel)$ spectrum. A particularly interesting situation occurs if the external particle (say electron) propagates along the surface. Then, it is possible to excite a coherent combination of surfriding transverse and longitudinal collective modes. If the phase velocity (on a given direction) exceeds the phase velocity of the surface eigenmodes in this direction surface shock waves may be generated [40]. These are a combination of Cherenkov and Landau shock waves.

Limiting ourselves to incoming p-polarized monochromatic plane waves emitted (approximately) by an appropriate external source, the p-polarized amplitude reflection coefficient turns out to be given by [34]

$$r_p(q_\parallel,\omega)=\frac{q_\perp^0-\left(\frac{\omega}{c}\right)^2M(q_\parallel,\omega)}{q_\perp^0+\left(\frac{\omega}{c}\right)^2M(q_\parallel,\omega)},\qquad(95)$$

where $q_\perp^0=[(\omega/c)^2-q_\parallel^2]^{1/2}$, in a configuration where the polarization unit vector of the incoming field is $\mathbf{e}_i=(q_\perp^0,0,-q_\parallel)c/\omega$. The quantity $M(q_\parallel,\omega)$ is given by

$$M(q_\parallel,\omega)=\frac{2i}{\pi}\int_0^\infty\left[\frac{q_\perp^2}{N_T(q)}+\frac{q_\parallel^2}{N_L(q)}\right]\frac{\cos(q_\perp 0^+)}{q^2}dq_\perp\qquad(96)$$

in the notation $\lim_{z\to0^+}\int F(q_\perp,z)dq_\perp\equiv\int F(q_\perp,0^+)dq_\perp$. The information on the half-space jellium excitations is contained in

$$N_T(q)=\left(\frac{\omega}{c}\right)^2\varepsilon_T(q,\omega)-q^2\qquad(97)$$

and

$$N_L(q)=\left(\frac{\omega}{c}\right)^2\varepsilon_L(q,\omega).\qquad(98)$$

It thus appears that $r^p(q_\parallel,\omega)$ can be expressed in a form where integrals of the transverse $[\varepsilon_T(q,\omega)]$ and longitudinal $[\varepsilon_L(q,\omega)]$ bulk dielectric response functions appear as central quantities. The reason that it is bulk epsilons which appear is our use of the "simple" specular electron reflection model. As it stands, $r_p(q_\parallel,\omega)$ depends on single-particle as well as collective transverse and longitudinal excitations.

It is important to note that $r_p(q_\parallel,\omega)$ basically *always* depends on both the transverse and longitudinal electrodynamics of the jellium [cf. the expression for $M(q_\parallel,\omega)$]. Being interested in this work in the role of the collective eigenmodes as they may manifest themselves in $r_p(q_\parallel,\omega)$, the integral in $M(q_\parallel,\omega)$ is carried out as a contour integral in a complex $q_\perp$ plane. The (two) pole contributions associated to $N_T([q_\parallel^2+q_\perp^2]^{1/2},\omega)$ and $N_L([q_\parallel^2+q_\perp^2]^{1/2},\omega)$ relate to the transverse surface plasmariton (also called polariton) and the surface-plasmon eigenmodes of the jellium.

C. Coupling to surface plasmons: Ritchie dispersion relation

A detailed discussion of the p-polarized reflection coefficient in Eq. (95) is beyond the scope of this paper. If retardation effects are neglected ($c\to\infty$) the plasmariton contribution to $r_p(q_\parallel,\omega)$ vanishes, and one obtains

$$r_p(q_\parallel,\omega)=\frac{\varepsilon(\omega)-1-\frac{iq_\parallel\kappa_\perp^L}{\kappa_L^2}\varepsilon(\omega)\left[1-\varepsilon_L^{-1}(q_\parallel,\omega)\right]}{\varepsilon(\omega)+1+\frac{iq_\parallel\kappa_\perp^L}{\kappa_L^2}\varepsilon(\omega)\left[1-\varepsilon_L^{-1}(q_\parallel,\omega)\right]},\qquad(99)$$

where $\varepsilon(\omega)=1-(\omega_p/\omega)^2$ is the common expression for $\varepsilon_T(q\to0,\omega)=\varepsilon_L(q\to0,\omega)$ in the long-wavelength limit. Furthermore,

$$\kappa_L^2=\frac{1}{a^2}\left(\omega^2-\omega_p^2\right),\qquad(100)$$

$$\kappa_\perp^L=+\frac{i}{a}\left[\omega_p^2+(aq_\parallel)^2-\omega^2\right]^{1/2},\qquad(101)$$

and

$$\varepsilon_L^{-1}(q_\parallel,\omega)-1=\frac{\omega_p^2}{\omega^2-(aq_\parallel)^2-\omega_p^2}.\qquad(102)$$

At this point, the reader may notice that a quantity

$$\omega^2-(aq_\parallel)^2-\omega_p^2=\left[\omega^2-(aq_\parallel)^2-\omega_{pS}^2\right]-\omega_{pS}^2\qquad(103)$$

appears in Eqs. (101) and (102). The surface-plasmon dispersion relation [Eq. (71)] appears in Eq. (103), setting $[\ldots]=0$.

Self-sustaining solutions are obtained for $r_p(q_\parallel,\omega)\to\infty$, as it is clear from Eqs. (92) and (93). Setting the denominator of Eq. (99) to zero

$$\varepsilon(\omega)+1+\frac{iq_\parallel\kappa_\perp^L}{\kappa_L^2}\varepsilon(\omega)\left[1-\varepsilon_L^{-1}(q_\parallel,\omega)\right]=0,\qquad(104)$$

one obtains, utilizing Eqs. (100)–(102), and remembering the inequality in Eq. (69), the dispersion relation

$$aq_\parallel^R=\omega\left[1-\left(\frac{\omega_{pS}}{\omega}\right)^2\right].\qquad(105)$$

The result in Eq. (105) is the Ritchie (superscript R on $q_\parallel$) condition for longitudinal surface waves [17,18]. Three important comments should be made to Eq. (105): (i) As a self-sustaining solution the Ritchie dispersion relation is *not observable*, (ii) the Ritchie condition deviates from the surface-plasmon dispersion relation needed for the quantum theory (71), and (iii) the surface quantum condition in Eq. (71) is neither directly excited experimentally. The difference between $aq_\parallel^R$ and $aq_\parallel$ depends on frequency. Thus, with $\varepsilon_S(\omega)=1-(\omega_{pS}/\omega)^2$, one has

$$(aq_\parallel^R)^2=(aq_\parallel)^2+\varepsilon_S(\omega)\omega_{pS}^2.\qquad(106)$$

Near the surface-plasmon frequency the Ritchie and quantum dispersion relation approach each other.

D. Charged-particle excitation of surface plasmons

The long-range Coulomb field attached to a moving charged-point particle (e.g., an electron) can in an effective manner excite surface plasmons [1,3,4,15]. In the perhaps simplest case, from a theoretical point of view, the particle is assumed to propagate parallel to the surface with constant velocity $\mathbf{V}$ (see Fig. 3). A microscopic classical electrodynamic theory for this case has been given in Ref. [40]. Below we connect this theory to the quantum theory presented in this paper.

The nonretarded p-polarized amplitude reflection coefficient related to surface plasmons excited in a $\mathbf{q}_\parallel$ direction making an angle α ($0\leqslant\alpha<2\pi$) to $\mathbf{V}$ is obtained inserting

$$\omega=\mathbf{q}_\parallel\cdot\mathbf{V}=q_\parallel V\cos\alpha\qquad(107)$$

in Eq. (97) (cf. the analysis in Ref. [40]). In

$$r_p(q_\parallel,\omega)=r_p(q_\parallel,q_\parallel V\cos\alpha)\qquad(108)$$

one now has two independent parameters $q_\parallel$ and the product $V\cos\alpha$. To examine r_p in a $q_\parallel$-$V\cos\alpha$ plane, and also the Ritchie and quantum surface-plasmon dispersion relations, it

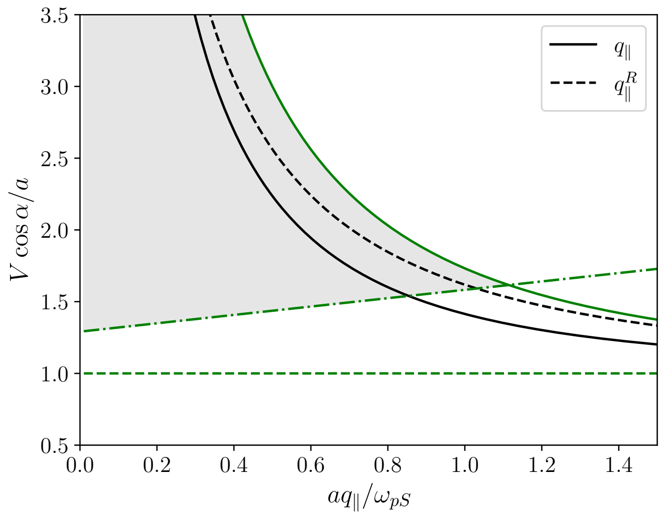

FIG. 4. The Ritchie ($q_\parallel^R$) and quantum ($q_\parallel$) dispersion relations shown on normalized form. As explained in the text the gray-toned domain is that of relevance for the collective surface excitations. This domain is bounded by the green curves representing Eqs. (110) (solid) and (113) (dashed-dotted). The shock-wave region is above the horizontal line $V\cos\alpha/a = 1$. The curves giving the transition to the single-particle excitation region have been drawn using data for doped n-InSb [electron density $n = 4\times10^{18}$ cm^{-3}, effective mass $m = 0.015m_0$ (m_0: free-electron mass)].

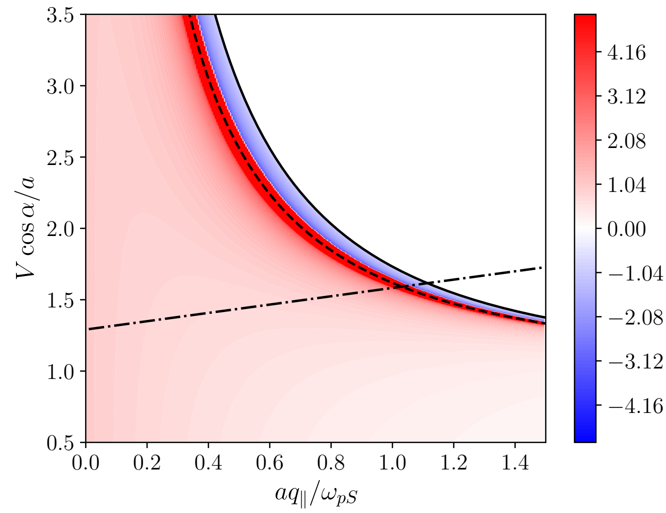

FIG. 5. The p-polarized amplitude reflection coefficient $r_p(q_\parallel,\omega) = r_p(q_\parallel, \omega = q_\parallel V\cos\alpha)$ as a function of the two independent quantities $q_\parallel$ and $V\cos\alpha$ (normalized to $aq_\parallel/\omega_{pS}$ and $V\cos\alpha/a$). Retardation effects are neglected ($c\to\infty$) in the plot. The fully drawn curve indicates the line between the $r_p = \mathrm{Re}(r_p)$ and $r_p = \mathrm{Im}(r_p)$ domains. The Ritchie dispersion relation is indicated as a dashed curve. The transition to the single-particle excitation region is given by the dashed-dotted curve, using n-InSb data (see Fig. 4 caption).

is convenient to introduce the dimensionless quantities

$$x = \frac{aq_\parallel}{\omega_{pS}}, \quad y = \frac{V\cos\alpha}{a}. \tag{109}$$

Let us consider first the dispersion relations. Written as $y = y(x)$ one obtains from Eq. (105) the following normalized Ritchie dispersion relation for $\omega = q_\parallel V\cos\alpha > 0$ ($-\pi/2 < \alpha < \pi/2$):

$$y = \frac{1}{2} + \left[\frac{1}{4} + x^{-2}\right]^{1/2}. \tag{110}$$

For the quantum dispersion relation [Eq. (71)] one gets

$$y = [1 + x^{-2}]^{1/2}. \tag{111}$$

Since the Cherenkov-Landau shock-wave condition $V\cos\alpha > a$ corresponds to $y > 1$ the Ritchie and the quantum dispersion relations are located entirely in the shock-wave domain. The dispersion relations are shown in Fig. 4. Which parts of the dispersion relations can be used? To answer this question, we know that the inequality in Eq. (69) must be satisfied. The border-line curve $[(aq_\parallel)^2 + \omega_p^2 - \omega^2 = 0]$ thus is given by

$$y = [1 + 2x^{-2}]^{1/2}. \tag{112}$$

Furthermore, it is necessary that we are outside the domain of single-particle excitations. As discussed in the Appendix one enters this domain at the kinematic condition

$$\hbar\omega = \frac{\hbar^2}{2m}(q_\parallel^2 + 2k_F q_\parallel). \tag{113}$$

By inserting $\omega = q_\parallel V\cos\alpha$, Eq. (113) can be rewritten in the form of a straight line

$$y = \frac{\hbar\omega_{pS}}{2ma^2}x + \frac{\hbar k_F}{ma}. \tag{114}$$

With $a = \sqrt{3/5}v_F = \sqrt{3/5}\hbar k_F/m$, and in terms of the Fermi energy, $\epsilon_F = (m/2)v_F^2$, the curve giving the border to the single-particle excitation domain can be written as

$$y = \frac{5}{12}\frac{\hbar\omega_{pS}}{\varepsilon_F}x + \left(\frac{5}{3}\right)^{1/2}. \tag{115}$$

This form is useful because of its "universality." It depends alone on the ratio between well-known (experimentally) quantities, viz., the long-wavelength surface-plasmon energy and the electron Fermi energy. In Fig. 4, the gray-toned area represents the domain in which the dispersion relations are of relevance.

In Fig. 5 we show the amplitude reflection coefficient r_p in the xy plane. When $y < [1 + 2x^{-2}]^{1/2}$ r_p is real, $r_p = \mathrm{Re}(r_p)$. In the forbidden region it is imaginary. The reader may notice that

$$r_p = \frac{\varepsilon(\omega) - 1}{\varepsilon(\omega) + 1} \tag{116}$$

on the border-line curve. The reason that $r_p \neq 0$ here associates to a deeper level of photon physics. Thus, if one neglects the L part of the dynamics in Eq. (95), one obtains the well-known expression [16,34]

$$r_p = \frac{q_\perp^0\varepsilon(\omega) - \kappa_\perp^T}{q_\perp^0\varepsilon(\omega) + \kappa_\perp^T}, \tag{117}$$

where $q_\perp^0 = [(\omega/c)^2 - q_\parallel^2]^{1/2}$ and $\kappa_\perp^T = [(\omega/c)^2\varepsilon(\omega) - q_\parallel^2]^{1/2}$ and $\varepsilon_T(\kappa_T, \omega) \approx \varepsilon(\omega)$ (long-wavelength value of the transverse dielectric function). When $c \to \infty$, Eq. (117) is reduced to Eq. (116). Thus, although retardation is neglected in Eq. (99), this equation (and the Ritchie dispersion relation) still contains a trace of the photon. This is so because the transverse photon dynamics has a *nonpropagating part* associated to the inability to localize a photon precisely in space-time [41]. It also appears from Fig. 5 that $\mathrm{Re}(r_p)$ changes sign near (but not exactly at) the Ritchie dispersion relation.

VI. OUTLOOK

In a formal sense the first- and second-quantized theories of the plasmon have much in common with the corresponding theories for a (massive) photon. In consequence, the important first-quantized photon theory, based on the Oppenheimer-Riemann-Silberstein energy wave-function formalism, or on the transverse gauge photon approach, can be "copied" for the plasmon starting from the quantum field concept $\phi(\mathbf{r}, t)$. We consider it of great importance to be in possession of such a first-quantized formulation [below named "plasmon wave mechanics" (PlWM)] for two reasons: (i) In PlWM one focuses on the positive-frequency part $\phi^{(+)}(\mathbf{r}, t)$ of the Klein-Gordon field. This part satisfies [as for (massive) photons] a Schrödinger-type first-order differential equation in time, and this fact allows one to apply many of the techniques used in nonrelativistic wave mechanics in studies, e.g., of the scattering of plasmons from obstacles and holes in free-electron-like solids. A PlWM understanding of scattering of plasmons and plasmaritons (see below) appears to be of potential importance in the quantum technology (plasmonics) [31]. (ii) A number of difficult problems in plasmon QED, and in plasmon technology, can be difficult to handle and solve starting directly from QED. In such cases, PlWM offers an easier route to the QED level, in analogy with the procedure followed for the photon; see, e.g., Ref. [39]. We plan to exemplify point (ii) by a study of single-plasmon scattering from a mesoscopic hole in a solid-state plasma.

Much of the field-quantized development in quantum optics and QED can be paralleled with plasmons. We believe that it will be possible to introduce and analyze the physics of plasmon states which in the quantum optical sense are coherent, squeezed, etc., and to deal with interference and diffraction phenomena on the single- and few-plasmon level. A theory closely related to the one presented in this work for L plasmons can be established for T plasmons (plasmaritons), as we will show in a forthcoming paper. The T plasmon can be strongly coupled to a photon, and the T-plasmon-photon first-quantized field upon extension to the QED level appears as a collection of quasiparticles for which quantum optical states of different nonclassical forms can be constructed. Fundamental insight, completely hidden in the classical electromagnetic theory of plasmaritons, appears in the QED formulation. Thus, the photon attachment to the T plasmon turns out to have its roots in the dressing of the individual jellium electrons by transverse virtual photons. At long photon wavelength the phenomenon can be studied starting from the Pauli-Fierz representation of QED [42]. In a broader framework the attachment relates to our inability to localize transverse photons in space-time [43]. Both the L and T plasmons couple to the photon, and this may give rise to new strongly coupled triplet states (L plasmon + T plasmon + photon). It is suggested to name this new quasiparticle a *jellion*. The microscopic quantum theory of jellions will be developed in a forthcoming paper. The surface jellion dispersion relation has three branches. Without knowing the microscopic physics of the jellion, its classical electrodynamic three-branch dispersion relation has been identified a number of years ago [44].

APPENDIX: HYDRODYNAMIC DIELECTRIC FUNCTION: SINGLE-PARTICLE EXCITATION DOMAIN

In order to place the plasmon dispersion relation in its proper framework, we summarize the calculation which leads from the Lindhard dielectric function [Eq. (2)] to the so-called hydrodynamic expression for $\varepsilon_L(q, \omega)$, given in Eq. (3). We also briefly discuss the manner in which the domain of single-particle excitations is cut out from the (ω, q) plane [cf. Fig. 2 for the surface-plasmon $(\omega, q_\parallel)$ plane].

1. $\varepsilon_L(q, \omega)$ to order q^2

Let us consider in the Lindhard formula the sum

$$S(\mathbf{q}, \omega) \equiv \frac{1}{\Omega} \sum_{\mathbf{k}} \frac{f_0(E_{\mathbf{k}+\mathbf{q}}) - f_0(E_{\mathbf{k}})}{E_{\mathbf{k}+\mathbf{q}} - E_{\mathbf{k}} - \hbar\omega}, \tag{A1}$$

and let us notice that the summation over $\mathbf{k}$ values can be turned into an integral over $\mathbf{k}$ space in the usual manner, viz.,

$$\frac{1}{\Omega} \sum_{\mathbf{k}} (\dots) \Rightarrow 2 \int_{-\infty}^{\infty} (\dots) \frac{d^3k}{(2\pi)^3}, \tag{A2}$$

where the factor of 2 originates in the spin summation. Since we are interested in the plasmon eigenmodes, the $i\hbar\eta$ term in the denominator of Eq. (2) is left out (the term relates to plasmon damping effects). A substitution $\mathbf{k} \to \mathbf{k} - \mathbf{q}$ in the first part of the sum in Eq. (A1) gives

$$S(\mathbf{q}, \omega) = \frac{1}{\Omega} \sum_{\mathbf{k}} f_0(E_{\mathbf{k}}) \left[\frac{1}{E_{\mathbf{k}} - E_{\mathbf{k}-\mathbf{q}} - \hbar\omega} - \frac{1}{E_{\mathbf{k}+\mathbf{q}} - E_{\mathbf{k}} - \hbar\omega} \right]. \tag{A3}$$

Since

$$E_{\mathbf{k}} - E_{\mathbf{k}-\mathbf{q}} - \hbar\omega = \frac{\hbar^2}{m} \mathbf{k} \cdot \mathbf{q} - \hbar\omega - \frac{\hbar^2 q^2}{2m}, \tag{A4}$$

$$E_{\mathbf{k}+\mathbf{q}} - E_{\mathbf{k}} - \hbar\omega = \frac{\hbar^2}{m} \mathbf{k} \cdot \mathbf{q} - \hbar\omega + \frac{\hbar^2 q^2}{2m}, \tag{A5}$$

the expression for $S(\mathbf{q}, \omega)$, given as an integral over $\mathbf{k}$ space, is reduced to

$$S(\mathbf{q}, \omega) = \frac{\hbar^2 q^2}{m} \int_{-\infty}^{\infty} \frac{f_0(E_{\mathbf{k}})}{\left(\frac{\hbar^2}{m} \mathbf{k} \cdot \mathbf{q} - \hbar\omega\right)^2 - \left(\frac{\hbar^2 q^2}{2m}\right)^2} \frac{d^3k}{4\pi^3}. \tag{A6}$$

A Taylor expression of the integrand to order q^2 gives the following long-wavelength approximation:

$$S(\mathbf{q},\omega) \simeq \frac{q^2}{m\omega^2}\int_{-\infty}^{\infty} f_0(E_{\mathbf{k}}) \times \left[1+\frac{2\hbar}{m\omega}\mathbf{k}\cdot\mathbf{q}+3\left(\frac{\hbar\mathbf{k}\cdot\mathbf{q}}{m\omega}\right)^2\right]\frac{d^3k}{4\pi^3}. \quad \text{(A7)}$$

Using the low-temperature ($T \rightarrow 0$) approximation for $f_0(E_{\mathbf{k}})$, the integral is easily evaluated in spherical coordinates, noting that the integral part containing $\mathbf{k}\cdot\mathbf{q}$ vanishes on symmetry grounds. The result is

$$S(\mathbf{q},\omega) = \frac{nq^2}{m\omega^2}\left[1+\frac{3}{5}\left(\frac{\hbar k_F q}{m\omega}\right)^2\right], \quad \text{(A8)}$$

where k_F is the Fermi wave number, and $n = k_F^3/(3\pi^2)$ is the bulk electron density. Note that S is a function of $|\mathbf{q}|$ only. Insertion of the expression given in Eq. (A8) into the Lindhard formula [Eq. (2)], the longitudinal dielectric function becomes to order q^2

$$\varepsilon_L(q,\omega) = 1-\left(\frac{\omega_p}{\omega}\right)^2\left[1+\frac{3}{5}v_F^2\left(\frac{q}{\omega}\right)^2\right] \simeq 1-\frac{\omega_p^2}{\omega^2-\frac{3}{5}v_F^2q^2}. \quad \text{(A9)}$$

In the last step we have made use of the fact that the term containing the spatial dispersion is small in comparison to 1. The expression for $\varepsilon_L(q,\omega)$ in Eq. (A9) is the result cited in Eq. (3), with the abbreviation in Eq. (4).

2. Kinematics of single-particle excitations

The energy and momentum conservation in a process where a photon of energy $\hbar\omega$ knocks an electron from state $\mathbf{k}$ to $\mathbf{k}+\mathbf{q}$ is buried in the condition

$$\hbar\omega = \frac{\hbar^2}{2m}(\mathbf{k}+\mathbf{q})^2-\frac{\hbar^2k^2}{2m} = \frac{\hbar^2}{m}\left(\frac{q^2}{2}+\mathbf{k}\cdot\mathbf{q}\right), \quad \text{(A10)}$$

a condition which makes the associated denominator term in Eq. (A1) [or Eq. (2)] equal to zero: the energy-momentum conservation is a resonance condition for the integral in Eq. (A6). A single-particle excitation can occur only if the electron is transferred from a filled state inside the Fermi surface to an empty state outside. This criterion gives a *minimum-energy* change

$$\hbar\omega_{\min} = \begin{cases} 0, & q \leqslant 2k_F \\ \frac{\hbar^2}{m}\left(\frac{q^2}{2}-qk_F\right), & q > 2k_F. \end{cases} \quad \text{(A11)}$$

The *maximum* change in energy is obtained when $\mathbf{k} = \mathbf{k}_F \parallel \mathbf{q}$, that is,

$$\hbar\omega_{\max} = \frac{\hbar^2}{m}\left(\frac{q^2}{2}+qk_F\right), \quad q \geqslant 0. \quad \text{(A12)}$$

The ($\omega > 0$, $q > 0$) parts of the parabolas in Eqs. (A11) and (A12) are shown in Fig. 2 in the surface wave case $q \rightarrow q_\parallel$, with scalings ω/ω_{pS} and $aq_\parallel/\omega_{pS}$.

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