# Bulk plasmaritons: Wave-mechanical and second-quantized theories

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Inspired by a recently established quantum theory for bulk and surface plasmons [Jung and Keller, Phys. Rev. A 103, 063501 (2021).], wave-mechanical and second-quantized quantum electrodynamic theories for bulk plasmaritons in a homogeneous jellium are presented. Starting from the "inner" structure of the transversely polarized classical plasmariton mode, it is argued that a plasmariton quasiparticle may be formed by an always attached pair consisting of a transverse (gauge) photon and a never observable transverse plasmon. A first-quantized plasmariton theory is established from a plasmariton Klein-Gordon equation. It is shown that the plasmariton can be perceived as a diamagnetically driven spin-1 boson quasiparticle. A Lagrangian-Hamiltonian formalism is used to extend the first-quantized plasmariton theory to the second-quantized level. The minimal coupling principle is used to change the free QED theory into a theory coupling the plasmariton to an electromagnetic gauge field. Using the microscopic transverse Lindhard (random phase approximation) dielectric function, it is shown that for small wave numbers, the classical Boltzmann equation part of the Lindhard dielectric function results in a hydrodynamic Brewster branch, the quantization of which follows that of a vectorial Klein-Gordon equation. For plasmariton wave numbers larger than the Fermi wave number, a pure electrostatic model (with no magnetic-field component) results in a harmonic-oscillator description of the plasmariton's polarization field. With inspiration and elements from the Power-Zinau-Wolley description, the Pauli-Fierz theory for particle-tied transverse photons, and the propagator theory of Keller for the spatial localization of transverse photons, a formalism in which the plasmariton quasiparticle is dressed by a cloud of transverse (long-wavelength) photons is developed. The quantized radiated part of the electromagnetic field is described by a free-field quantization of the transverse part of the displacement field ( $\mathbf{D}_{T}$ ). The total Hamiltonian is diagonalized, and the resulting dispersion relation obtained. The interaction between the transverse radiated and attached photons implies that a strongly localized and charged plasmariton quasiparticle with its tied photons vibrates as a simple harmonic oscillator in the "external" radiative transverse photon field.

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# I. INTRODUCTION

For over half a century, studies of the electromagnetic interaction between light and collective charged particle (electrons, ions) distributions in solid-state plasmas have played an increasingly important role in physical optics. An electromagnetic wave traveling through the plasma induces (microscopic) polarizations in the particle system, and the coupled field-particle modes are generally known as polariton modes. In the jellium approximation, where the ionic potential is smeared uniformly in space, the light-electron modes are sometimes called *plasmaritons* [1], a term we shall use in this paper. Following a period in which bulk plasmaritons were the main subject of study, investigations of surface plasmaritons have set the scene for many-faceted theoretical and experimental research [2-4]. In a bulk jellium, the plasmaritons are divergence-free [called transverse (T)] modes. Charged particles (e.g., electrons) can excite another collective plasma mode, called a *plasmon*. In a bulk jellium, the plasmon modes are rotational-free [called longitudinal (L)], and these modes

also exist as surface plasmons. In general, the surface modes are of a mixed plasmon-plasmariton type.

In the present article, we shall focus our attention on a theoretical study of bulk plasmaritons, and we will leave studies of surface plasmaritons for an upcoming paper. We do this because the connection between bulk and surface excitations from a theoretical point of view is less strong than often claimed in the literature. This is so since the surface region contains an extra source emitting evanescent modes, which in a covariant treatment comprises not only T-modes but also L-modes and scalar S-modes [5,6]. These L- and S-modes can be replaced by the sum of a gauge-mode (which can be eliminated by a gauge transformation within the Lorenz gauge) and a so-called near-field (NF) mode [7].

In recent years, studies aimed at technological applications of the physical optical plasmariton theory (bulk, surface, and interface) have attracted a great deal of attention, and terms such as "plasmonics" and "surface plasmonics" have been catchy designations.

In this work, we go in the opposite direction and focus on the basic foundation of plasmariton theory. We have three main goals: (i) To establish a quantum electrodynamic (QED) theory for plasmaritons that enables one to make use of the machinery of quantum optics, and thus investigate coherent,

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squeezed, and entangled states, and others for plasmaritons. (ii) To establish tight connections and analogies to the comprehensive literature dealing with single photons and massive quasiparticles surrounded by their tied photon clouds. (iii) To formulate and develop a formalism describing the plasmariton as a diamagnetically driven spin-1 particle.

The paper is organized as follows. In Sec. II, we revisit aspects of the classical plasmariton description, with an eve on the quantum theory to follow. Thus, in Secs. II A-II C, we discuss, respectively, the *T* and *L* dielectric functions [8], the eigenmode conditions, and the "inner" structure of the plasmariton dispersion relation. In Sec. III, we show how to formulate a first-quantized theory for a single plasmariton, in a manner closely related to the first-quantized theory for single photons [9–16]. We start by a Klein-Gordon equation inferred from the plasmariton dispersion in analogy with the setting up of the Klein-Gordon equation for a (massive) electron from its relativistic dispersion relation (Sec. III A). The dynamic evolution of the plasmariton wave function is governed by a Schrödinger-like first-order (in time) equation, much like the Landau-Peierls evolution equation for a photon [9] (Sec. III B). We proceed to describe the plasmariton as a diamagnetically spin-1 particle, and we also demonstrate how a plasmariton helicity concept can be introduced for the plasmariton (Sec. IV). The first-quantized plasmariton theory, which one may characterize as a plasmariton (PL) wave-mechanical (WM) theory (PLWM), is extended to the second-quantized (QED) level through a Lagrange-Hamiltonian approach for free plasmaritons in Sec. V. The inspiration for this line of thought has come from our previously established QED theory for bulk plasmons [17]. In Sec. VI, we develop the theory to include the plasmariton's interaction with an electromagnetic gauge field via the minimal coupling principle  $[\hat{p}_{\mu} => \hat{p}_{\mu} - Q\hat{A}_{\mu}, \hat{p}_{\mu} \text{ and } \hat{A}_{\mu}$  being the  $\mu$ th component of the plasmariton momentum and fourpotential field operators ( $\mu = 0-3$ ), and Q is the plasmariton charge] [6,18,19].

In Secs. VII and VIII, we study plasmaritons using the Lindhard transverse dielectric function [based on a randomphase-approximation (RPA) description] [8,20–22]. We show that this theory leads one to a QED theory with two types of modes (quanta) following, respectively, the Klein-Gordon equation [19] (Brewster-like branch) [2–4] and, at wave numbers exceeding the Fermi wave number, a simple harmonic-oscillator equation. In the oscillator-like regime, the Lindhard function is predominantly electrostatic. In the classical regime, where quantum interference effects are absent, we bring the Lindhard formalism in contact to the Boltzmann equation formalism [23-26] and then to the hydrodynamic theory for T-modes. The hydrodynamic approach can be derived from the Vlasov equation, i.e., the collisionless Boltzmann equation. The hydrodynamic theory often works quite well, particularly when extended to the density functional level [2,26].

Before the summary and outlook (Sec. X), we finish the general theory of plasmaritons with a Hamiltonian description based on a model where the transverse microscopic displacement field  $\mathbf{D}_T$  (multiplied by -1) plays the role of canonical field momentum (Secs. IX A and IX B). In this picture, the jellium oscillators appear with a tied *T*-photon cloud [a charged

jellium oscillator with renormalized plasma frequency ( $\omega_p \Rightarrow \sqrt{2}\omega_p$ )]. Among other important aspects, we prove that the transverse field tied to the oscillator interacts with the free  $\mathbf{D}_T$  field (Sec. IX C). This part of our theory establishes a link to the Pauli-Fierz theory [27,28] and the propagator gauge formalism developed previously to describe the spatial localization of quantized light emitted from a single-electron atom [29,30]. In Sec. IX D, we reach the final form for the total Hamiltonian operator, and in Sec. IX E, the Hamiltonian is diagonalized to obtain the dispersion relation for the plasmariton modes in the  $\mathbf{D}_T$ -description.

The interested reader may refer to Appendixes A and B for details on the transverse Lindhard dielectric function and the plasmariton oscillator surrounded by its *T*-photon cloud.

# II. THE CLASSICAL PLASMARITON DISPERSION RELATION REVISITED

# A. Lindhard dielectric functions at small wave numbers

In a bulk jellium system, the infinitesimal translational and rotational invariance dictates that the microscopic relative dielectric tensor in the wave vector(**q**)-frequency( $\omega$ ) domain,  $\boldsymbol{\varepsilon}(\mathbf{q}, \omega)$ , has the general dyadic form

$$\boldsymbol{\varepsilon}(\mathbf{q},\omega) = (\mathbf{U} - \hat{\mathbf{q}}\hat{\mathbf{q}})\varepsilon_T(q,\omega) + \hat{\mathbf{q}}\hat{\mathbf{q}}\varepsilon_L(q,\omega), \qquad (1)$$

where  $\hat{\mathbf{q}} = \mathbf{q}/q$ , and U is the 3 × 3 unit tensor. The transverse (*T*) dielectric function  $\varepsilon_T(q, \omega)$  and the longitudinal (*L*) dielectric function  $\varepsilon_L(q, \omega)$  depend on the magnitude of the wave vector  $|\mathbf{q}|$  only, and become identical in the long-wavelength limit, i.e.,

$$\varepsilon_T(q \to 0, \omega) = \varepsilon_L(q \to 0, \omega) \equiv \varepsilon(\omega).$$
 (2)

In the random phase approximation (RPA), also called the self-consistent field (SCF) approach, explicit expressions for  $\varepsilon_T(q, \omega)$  and  $\varepsilon_L(q, \omega)$  were given by Lindhard [20]. In the Lindhard theory, both single-particle and collective-mode excitations are included. In the present context, we focus on the collective excitations. These are obtained by an expansion of  $\varepsilon_T(q, \omega)$  and  $\varepsilon_L(q, \omega)$  to lowest order ( $\sim q^2$ ) in the wave vector. The inversion symmetry of the jellium implies that terms linear in the wave vector vanish.

For small q, one obtains (for  $\varepsilon_T$ ; see Appendix A 4)

$$\varepsilon_T(q,\omega) = 1 - \frac{\omega_p^2}{\omega^2 - D_T q^2}$$
(3)

and

$$\varepsilon_L(q,\omega) = 1 - \frac{\omega_p^2}{\omega^2 - D_L q^2},\tag{4}$$

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

$$D_T = \frac{1}{5}v_F^2,\tag{5}$$

$$D_L = \frac{3}{5} v_F^2,\tag{6}$$

are the so-called transverse and longitudinal diffusion coefficients. These are of the order of the electron Fermi velocity,  $v_F$ . The expressions in Eqs. (3) and (4) are derived most easily

from the classical Lindhard regime, identical to what may be obtained from a Boltzmann equation approach; see Sec. VII C and Appendix A 3. The results in Eqs. (3) and (4) are just those obtained also from a phenomenological hydrodynamic approach. While Eq. (4) is well-established [with  $D_L$  given by Eq. (6)], Eq. (3), which is of main interest in this paper, often is postulated [as an analogy to Eq. (4)] in macroscopic electrodynamics. However, the value  $D_T = (1/5)v_F^2$  for the diffusion coefficient follows exactly from the microscopic Boltzmann equation.

# **B.** Eigenmode conditions

In microscopic linear Maxwell-Lorentz electrodynamics, one introduces the relations

$$\mathbf{D}(\mathbf{q},\omega) \equiv \varepsilon_0 \mathbf{E}(\mathbf{q},\omega) + \mathbf{P}(\mathbf{q},\omega) = \varepsilon_0 \boldsymbol{\varepsilon}(\mathbf{q},\omega) \cdot \mathbf{E}(\mathbf{q},\omega), \quad (7)$$

where  $\mathbf{D}(\mathbf{q}, \omega)$ ,  $\mathbf{P}(\mathbf{q}, \omega)$ , and  $\mathbf{E}(\mathbf{q}, \omega)$  are the *microscopic* displacement field, polarization field, and the local electric field, respectively. Although the relations in Eq. (7) look like those well-known from macroscopic electrodynamics, they are different. Thus, the polarization field  $\mathbf{P}(\mathbf{q}, \omega)$  contains all multipole moments, not only the electric dipole moment (per unit volume). Microscopically, the electron current density given by

$$\mathbf{J}(\mathbf{q},\omega) = -i\omega \mathcal{P}(\mathbf{q},\omega) + i\mathbf{q} \times \mathcal{M}(\mathbf{q},\omega)$$
(8)

is a well-defined quantity, related to quantum mechanics (and QED). The division of  $\mathbf{J}(\mathbf{q}, \omega)$  into "polarization [ $\mathcal{P}(\mathbf{q}, \omega)$ ]" and "magnetization [ $\mathcal{M}(\mathbf{q}, \omega)$ ]" parts, to a certain extent, is arbitrary [31]. It is known that it is possible (by an appropriate transformation) to eliminate  $\mathcal{M}(\mathbf{q}, \omega)$  from the formalism, giving [31]

$$\mathbf{J}(\mathbf{q},\omega) = -i\omega \mathbf{P}(\mathbf{q},\omega). \tag{9}$$

It is the  $\mathbf{P}(\mathbf{q}, \omega)$ , related to the physical quantity  $\mathbf{J}(\mathbf{q}, \omega)$  in Eq. (9), which enters Eq. (7). Roughly speaking,  $\mathbf{P}(\mathbf{q}, \omega)$  contains also magnetic responses, and as a consequence  $\boldsymbol{\varepsilon}(\mathbf{q}, \omega)$  also describes magnetic effects for  $\mathbf{q} \neq \mathbf{0}$ . This formalism is well known from linear electrodynamics of BCS-superconductors, where it can account for the linear Meissner effect [32,33].

Let us now bring into play the microscopic Maxwell equation

$$i\mathbf{q} \times \mathbf{B}(\mathbf{q},\omega) = \mu_0 \mathbf{J}(\mathbf{q},\omega) - \frac{i\omega}{c^2} \mathbf{E}(\mathbf{q},\omega),$$
 (10)

written in the  $(\mathbf{q}, \omega)$ -representation of  $\nabla \times \mathbf{B}(\mathbf{r}, t) = \mu_0 \mathbf{J}(\mathbf{r}, t) + c^{-2} \partial \mathbf{E}(\mathbf{r}, t) / \partial t$ . With the help of Eqs. (7) and (9), Eq. (10) can be written as  $(\mu_0 \varepsilon_0 = c^{-2})$ 

$$-\frac{c^2}{\omega}\mathbf{q}\times\mathbf{B}(\mathbf{q},\omega)=\boldsymbol{\varepsilon}(\mathbf{q},\omega)\cdot\mathbf{E}(\mathbf{q},\omega),\qquad(11)$$

and hence in the jellium case

$$-\frac{c^2}{\omega} \mathbf{q} \times \mathbf{B}(\mathbf{q}, \omega) = \varepsilon_L(q, \omega) \cdot \mathbf{E}_L(\mathbf{q}, \omega) + \varepsilon_T(q, \omega) \cdot \mathbf{E}_T(\mathbf{q}, \omega), \quad (12)$$

where  $\mathbf{E}_T(\mathbf{q}, \omega) = (\mathbf{U} - \hat{\mathbf{q}}\hat{\mathbf{q}}) \cdot \mathbf{E}(\mathbf{q}, \omega)$  and  $\mathbf{E}_L(\mathbf{q}, \omega) = \hat{\mathbf{q}}\hat{\mathbf{q}} \cdot \mathbf{E}(\mathbf{q}, \omega)$  are the transverse and longitudinal parts of the

local electric field. With the help of the Maxwell equation  $\mathbf{B}(\mathbf{q}, \omega) = \omega^{-1}\mathbf{q} \times \mathbf{E}(\mathbf{q}, \omega) \quad [\nabla \times \mathbf{E}(\mathbf{r}, t) = -\partial \mathbf{B}(\mathbf{r}, t)/\partial t],$  the left side of Eq. (12) can be rewritten as

$$\frac{c^2}{\omega} \mathbf{q} \times \mathbf{B}(\mathbf{q}, \omega) = \left(\frac{cq}{\omega}\right)^2 (\mathbf{U} - \hat{\mathbf{q}}\hat{\mathbf{q}}) \cdot \mathbf{E}(\mathbf{q}, \omega)$$
$$= \left(\frac{cq}{\omega}\right)^2 \mathbf{E}_T(\mathbf{q}, \omega). \tag{13}$$

Thus, one obtains

$$\varepsilon_L(q,\omega)\mathbf{E}_L(\mathbf{q},\omega) + \left[\varepsilon_T(q,\omega) - \left(\frac{cq}{\omega}\right)^2\right]\mathbf{E}_T(\mathbf{q},\omega) = \mathbf{0}.$$
(14)

To satisfy Eq. (14), its *L*- and *T*-parts separately must be zero. This gives [for  $\mathbf{E}_L(\mathbf{q}, \omega) \neq \mathbf{0}$ ] the bulk plasmon dispersion relation

$$\varepsilon_L(q,\omega) = 0,\tag{15}$$

and [for  $\mathbf{E}_T(\mathbf{q}, \omega) \neq \mathbf{0}$ ] the bulk plasmariton dispersion relation

$$\varepsilon_T(q,\omega) = \left(\frac{cq}{\omega}\right)^2.$$
 (16)

These microscopic dispersion relations have been known for many years. In many (macroscopic) classical studies, the often small wave-number dispersion in  $\varepsilon_T(\mathbf{q}, \omega)$  is neglected; cf. Eq. (2).

# C. The "inner" structure of the bulk plasmariton dispersion relation

In view of the quantum theory for plasmaritons, which will be established in Secs. III–VI, it is worthwhile to reflect on, and compare, the structure of Eqs. (15) and (16). Using the hydrodynamic expression for  $\varepsilon_L(q, \omega)$  [Eq. (4)], the (squared) plasmon dispersion relation takes the form

 $\omega^2 = (a_L q)^2 + \omega_p^2$ 

$$a_L = D_L^{1/2} = \sqrt{\frac{3}{5}} v_F.$$
(18)

(17)

In one of our recently published papers [17], a wavemechanical theory for bulk plasmons was established starting from Eq. (17). On the basis of the Lagrangian formalism, this theory afterward was extended to the QED level. Note that only (two) parameters relating solely to the jellium, viz.,  $a_L$ and  $\omega_p$ , appear in Eq. (17) (both parameters are functions of the electron density). At a deeper level, this circumstance is related to the fact that the electric *L*-field can be eliminated in favor of the electron position coordinates in the Schrödinger equation, and thus the density becomes a factor. In density functional theory, which is exact only for  $\mathbf{E}_T(\mathbf{r}, t) = \mathbf{0}$ , the (many-body) Schrödinger equation can be rewritten as a functional of the in general inhomogeneous electron density  $n(\mathbf{r})$ [34,35].

It is tempting, *but generally wrong*, to develop the quantum theory of plasmaritons from an equation analogous to Eq. (15), viz.,

$$\varepsilon_T(q,\omega) = 0, \tag{19}$$

an equation giving the hydrodynamic [Eq. (3)] dispersion relation (in squared form)

$$\omega^2 = (a_T q)^2 + \omega_p^2, \qquad (20)$$

where

$$a_T = D_T^{1/2} = \sqrt{\frac{1}{5}} v_F.$$
 (21)

As discussed in Appendix A 2, the dispersion relation is the correct one obtained from the Lindhard formula at large wave numbers  $q \gg k_F = mv_F/\hbar$ ,  $k_F$  being the electron Fermi wave number. For  $q/k_F \gg 1$ , magnetic effects can be neglected  $(c \rightarrow \infty, \text{effectively})$  and the dispersion relation becomes the "electrostatic" one given by Eq. (19).

However, the correct quantum theory for the plasmaritons involves attached photons; this is obvious from Eq. (12) in a sense. From the correct dispersion relation for T-modes [Eq. (16)], the following result is obtained from Eq. (3):

$$\omega^2 = (cq)^2 + \omega_p^2. \tag{22}$$

The dispersion relation in Eq. (22) is the starting point for our quantum theory for the bulk plasmaritons.

The mixed *T*-plasmon/photon character of the dispersion relation in Eq. (22) appears explicitly from the fact that jellium  $(\omega_p)$  and photon (c) parameters appear in Eq. (22). We have added the words *attached photon* and not just photon, because it is impossible to split the plasmariton into a free *T*-photon and a *T*-plasmon. Attached transverse photons appear in general fields of physics at long wavelengths. The transverse field tied to a classical particle is conveniently described in terms of the Pauli-Fierz representation [27]. See a graphical illustration of the "inner" plasmariton structure and the quasiparticles tied photons in Fig. 1.

# III. FIRST-QUANTIZED THEORY OF BULK PLASMARITONS

In this section, we establish and discuss a first-quantized theory of bulk plasmaritons. This quantum-mechanical theory of a new single-particle plasmariton field is based on a wave equation of the Klein-Gordon type. The vectorial wave function can be divided into two species composed of orthogonal polarization states in the wave-vector representation. The positive-frequency part of the plasmariton wave function satisfies a first-order (in time) differential equation of the Riemann-Silberstein-Oppenheimer type used extensively in photon wave mechanics by Bialynicki [13] and others [9-12,14-16].

# A. Plasmariton Klein-Gordon equation

Although parts of the quantum-mechanical theory of the plasmariton can be established technically in analogy to that of the plasmon [17], the underlying physical interpretation is qualitatively different, and it offers a new perspective on the divergence-free ( $\sim$  transverse) collective modes.



FIG. 1. Schematic illustration of the "inner" bulk plasmariton structure. (a) Light-line in red (straight line), *T*-plasmon in blue (lower curved line), plasmariton in green (middle curved line), and the renormalized in violet,  $\tilde{\omega}_p = \sqrt{2}\omega_p$  (upper curved line) dispersion relations. (b) A localized plasmariton quasiparticle (big circle) with its "internal" boson particles: *T*-photon, red (left) and *T*-plasmon, blue (right). (c) Plasmariton quasiparticle with its tied *T*-photon cloud in violet (gray shaded); see the renormalization analysis in Sec. IX.

A quantum-mechanical wave equation for the plasmariton is obtained from the dispersion relation in Eq. (22) via the usual prescription  $-i\omega \Rightarrow \partial/\partial t$  and  $i\mathbf{q} \Rightarrow \nabla$ . This gives the following Klein-Gordon-like wave equation for the plasmariton wave function  $\Phi(\mathbf{r}, t)$ :

$$\left(\nabla^2 - \frac{1}{c^2} \frac{\partial^2}{\partial t^2}\right) \mathbf{\Phi}(\mathbf{r}, t) - Q_C^2 \mathbf{\Phi}(\mathbf{r}, t) = \mathbf{0}, \qquad (23)$$

where a plasmariton Compton number is defined as

$$Q_C = \frac{\omega_p}{c}.$$
 (24)

Due to the fact that the transverse dynamics in the jellium has two independent polarization degrees of freedom, the plasmariton wave function is a vectorial quantity, satisfying in space-time the transversality condition

$$\nabla \cdot \mathbf{\Phi}(\mathbf{r}, t) = \mathbf{0}. \tag{25}$$

The introduction of the wave function  $\Phi(\mathbf{r}, t)$  brings into focus the fact that the plasmariton inevitably is a *single quantity*, not a coupled *T*-plasmon–photon state, as classical electromagnetic theory suggests. The plasmariton cannot in any manner experimentally be separated into *T*-plasmon and photon parts. Seen in this perspective, the plasmariton in a sense has a status similar to the tied (attached) field picture appearing for single electrons in the long-wavelength Pauli-Fierz theory [27] and in the spatial delocalization problem related to single-photon emission from an atom [36]. Sometimes it is convenient to divide  $\Phi(\mathbf{r}, t)$  into two polarization species related to a summation over the wave-vector spectrum of the plasmariton wave function. Thus, let us write the wave function as a Fourier integral, viz.,

$$\mathbf{\Phi}(\mathbf{r},t) = \sum_{i=1,2} \int_{-\infty}^{\infty} \Phi_i(\mathbf{q},\omega) \hat{\mathbf{e}}_i(\hat{\mathbf{q}}) e^{i(\mathbf{q}\cdot\mathbf{r}-\omega t)} \frac{d^3q}{(2\pi)^3} \frac{d\omega}{2\pi}.$$
 (26)

The pair of generally complex unit vectors  $\hat{\mathbf{e}}_i$  (*i* = 1, 2) satisfy the conditions

$$\hat{\mathbf{q}} \cdot \hat{\mathbf{e}}_i(\hat{\mathbf{q}}) = \mathbf{0}, \quad \hat{\mathbf{e}}_i^*(\hat{\mathbf{q}}) \cdot \hat{\mathbf{e}}_j(\hat{\mathbf{q}}) = \delta_{ij}, \quad (27)$$

where  $\hat{\mathbf{q}} = \mathbf{q}/q$  is a unit vector in the wave-vector (**q**) direction,  $\delta_{ij}$  is the Kronecker symbol, and i, j = 1, 2. The decomposition in Eq. (26) obviously satisfies Eq. (25). In the space-time domain, the two plasmariton wave-function species are

$$\Phi_{i}(\mathbf{r},t) = (2\pi)^{-4} \int_{-\infty}^{\infty} \Phi_{i}(\mathbf{q},\omega) \hat{\mathbf{e}}_{i}(\hat{\mathbf{q}}) e^{i(\mathbf{q}\cdot\mathbf{r}-\omega t)} d^{3}q d\omega,$$
  

$$i = 1, 2.$$
(28)

For the free plasmariton field, the two wave-function species develop independently in time.

The two species each satisfy the relativistic energymomentum relation

$$\hbar\omega = +[(c\hbar q)^2 + (Mc^2)^2]^{1/2},$$
(29)

where

$$M = \frac{\hbar\omega_p}{c^2} \tag{30}$$

is the plasmariton "rest mass." Defining covariant and contravariant derivatives with metric signature (1, -1, -1, -1), i.e.,

$$\{\partial_{\mu}\} \equiv \left(\frac{1}{c}\frac{\partial}{\partial t}, \boldsymbol{\nabla}\right), \ \{\partial^{\mu}\} \equiv \left(\frac{1}{c}\frac{\partial}{\partial t}, -\boldsymbol{\nabla}\right), \quad (31)$$

the vectorial plasmariton wave equation reads

$$\left(\partial_{\mu}\partial^{\mu} + Q_{C}^{2}\right)\boldsymbol{\Phi}(\mathbf{r},t) = \mathbf{0}.$$
(32)

## B. Dynamical evolution equation for plasmaritons

Let us consider the analytical (positive-frequency) part,  $\Phi^{(+)}(\mathbf{r}, t)$ , of the plasmariton wave function,  $\Phi^{(+)}(\mathbf{r}, t)$ . This part, given by

$$\mathbf{\Phi}^{(+)}(\mathbf{r},t) = \frac{1}{2\pi} \int_{-\infty}^{\infty} \theta(\omega) \mathbf{\Phi}(\mathbf{r};\omega) e^{-i\omega t} d\omega, \qquad (33)$$

where  $\theta(\omega)$  is the Heaviside unit step function, also satisfies the Klein-Gordon equation, i.e.,

$$\left(\nabla^2 - \frac{1}{c^2} \frac{\partial^2}{\partial t^2}\right) \mathbf{\Phi}^{(+)}(\mathbf{r}, t) - Q_C^2 \mathbf{\Phi}^{(+)}(\mathbf{r}, t) = \mathbf{0}.$$
 (34)

In the  $(\mathbf{q}, \omega)$ -domain, Eq. (34) multiplied by  $-c^2$  takes the form

$$\left[c^{2}\left(q^{2}+Q_{C}^{2}\right)-\omega^{2}\right]\boldsymbol{\Phi}^{(+)}(\mathbf{q},\omega)=\mathbf{0}.$$
(35)

We now rewrite Eq. (35) as

$$[c\sqrt{q^{2}+Q_{C}^{2}}+\omega][c\sqrt{q^{2}+Q_{C}^{2}}-\omega]\Phi^{(+)}(\mathbf{q},\omega)=\mathbf{0}.$$
 (36)

Since  $\Phi^{(+)}(\mathbf{q}, \omega) = \theta(\omega)\Phi(\mathbf{q}, \omega) = 0$  for  $\omega < 0$ , Eq. (36) is identical to

$$\left[c\sqrt{q^2+Q_C^2}-\omega\right]\Phi^{(+)}(\mathbf{q},\omega)=\mathbf{0}.$$
(37)

In the  $(\mathbf{q}; t)$ -domain, the analytical part of the plasmariton wave function hence satisfies the dynamical (first-order in time) evolution equation

$$i\hbar\frac{\partial}{\partial t}\mathbf{\Phi}^{(+)}(\mathbf{q};t) = \hat{H}(q)\mathbf{\Phi}^{(+)}(\mathbf{q};t), \qquad (38)$$

where

$$\hat{H}(q) = c\hbar\sqrt{q^2 + Q_C^2} \tag{39}$$

is the Hamiltonian (operator); cf. Eq. (29). If one defines a nonlocal operator  $(Q_C^2 - \nabla^2)^{1/2}$  via its action in Fourier space, i.e.,

$$\sqrt{\mathcal{Q}_C^2 - \nabla^2} \mathbf{F}(\mathbf{r}) \equiv \int_{-\infty}^{\infty} \sqrt{\mathcal{Q}_C^2 + q^2} \mathbf{F}(\mathbf{q}) e^{i\mathbf{q}\cdot\mathbf{r}} \frac{d^3q}{(2\pi)^3}, \quad (40)$$

the plasmariton evolution equation can be written in the Schrödinger-like form in space-time, viz.,

$$i\hbar\frac{\partial}{\partial t}\mathbf{\Phi}^{(+)}(\mathbf{r},t) = c\hbar\sqrt{Q_C^2 - \nabla^2}\mathbf{\Phi}^{(+)}(\mathbf{r},t).$$
(41)

The quantity  $\sqrt{-\nabla^2} (Q_C = 0)$  was introduced by Landau and Peierls in 1930 [9] in one of the first attempts to formulate a wave-mechanical theory for photons; see Ref. [5] and references therein.

# IV. PLASMARITON AS A DIAMAGNETICALLY DRIVEN SPIN-1 PARTICLE

Up to this point, we have considered the plasmaritons as quasiparticles built from T-plasmons with attached photons. In this section, the perspective is turned around in that we regard the plasmariton as a transverse photon inevitably subject to diamagnetic coupling to matter (here jellium). Seeing the plasmariton in different perspectives helps one to link the phenomenon to other fields of electromagnetics.

It is known that the diamagnetic effect is universal in electrodynamics. Thus, independent of the manner in which charged particles are coupled electronically (here in jellium via Coulomb forces), the effect *only* depends on the (quantummechanical) particle density,  $n(\mathbf{r})$ , in the field (~ transverse photon)-unperturbed state. Subjected to a self-consistently determined transverse vector potential,  $\mathbf{A}_T(\mathbf{r}, t)$ , a diamagnetic microscopic current density,

$$\mathbf{J}(\mathbf{r},t) = -\frac{n(\mathbf{r})e^2}{m}\mathbf{A}_T(\mathbf{r},t),$$
(42)

is induced in matter (charged particles assumed here to be electrons). The perhaps most prominent example of a diamagnetic phenomenon is the Meissner effect in superconductors [32,33]. Above the superconducting transition temperature ( $T_C$ ), the induced current density consists of paramagnetic and diamagnetic parts (as in all solids). The paramagnetic part is structure (e.g., ~ band structure) -dependent, and superconductors are unique in the sense that the paramagnetic part tends to vanish below  $T_C$ . In a two-fluid picture, only the diamagnetic part exists for the superconducting fluid part. Note that the expression in Eq. (42) is gauge-invariant, since  $A_T$  is. The transverse vector potential satisfies the microscopic wave equation

$$\left(\nabla^2 - \frac{1}{c^2} \frac{\partial^2}{\partial t^2}\right) \mathbf{A}_T(\mathbf{r}, t) = -\mu_0 \mathbf{J}_T(\mathbf{r}, t), \qquad (43)$$

where  $\mathbf{J}_T(\mathbf{r}, t)$  (in the diamagnetic case) is the divergence-free (transverse) part of  $\mathbf{J}_T(\mathbf{r}, t)$  in Eq. (42). For a nonuniform collection of electrons, Eq. (42) couples the transverse (*T*) and longitudinal (*L*) dynamics. However, for a uniform jellium (density: *n*), where  $\mathbf{J}_T(\mathbf{r}, t) = \mathbf{J}(\mathbf{r}, t)$ , the *T*-and *L*-dynamics decouple, and  $\mathbf{A}_T(\mathbf{r}, t)$  satisfies the inhomogeneous wave equation

$$\left(\nabla^2 - \frac{1}{c^2} \frac{\partial^2}{\partial t^2}\right) \mathbf{A}_T(\mathbf{r}, t) - \frac{\mu_0 n e^2}{m} \mathbf{A}_T(\mathbf{r}, t) = \mathbf{0}.$$
 (44)

In photon wave mechanics,  $\mathbf{A}_T(\mathbf{r}, t)$  (properly normalized) may represent the wave function of a single photon [13], and Eq. (44) then is the wave equation for a free  $\mathbf{A}_T$ -photon driven by a diamagnetic current density distribution [ $\mathbf{J}_T = \mathbf{J}$  in Eq. (42)].

Since  $\mu_0 n e^2/m = (\omega_p/c)^2 = Q_C^2$ , a comparison of Eqs. (23) and (44) shows that the plasmariton can be conceived as a *T*-photon unavoidably coupled diamagnetically to jellium. Seen in this perspective, the formalisms used in photon wave mechanics can be applied to plasmariton wave mechanics. The methods of photon wave mechanics in structural vacuum [36], an extension of the Riemann-Silberstein-Oppenheimer photon energy wave function formalism [10–16], appears particularly attractive [36].

One can introduce a plasmariton spin-1 operator in the description in the following manner. Let us first rewrite the curl of the plasmariton wave function as

$$\nabla \times \mathbf{\Phi} = i^{-1} (\boldsymbol{\sigma} \cdot \nabla) \mathbf{\Phi}, \tag{45}$$

where  $\boldsymbol{\sigma} = (\sigma_1, \sigma_2, \sigma_3)$  is the well-known dimensionless Cartesian spin-1 operator, the elements of which are the matrices

$$(\sigma_i)_{jk} = i^{-1} \varepsilon_{ijk}, \tag{46}$$

 $\varepsilon_{ijk}$  being the Levi-Civita symbol. Now, since  $\nabla \cdot \Phi = 0$  [Eq. (25)], one obtains

$$\nabla^2 \mathbf{\Phi} = -\nabla \times (\nabla \times \mathbf{\Phi}) = (\boldsymbol{\sigma} \cdot \nabla)^2 \mathbf{\Phi}, \qquad (47)$$

a relation that with the help of the momentum operator

$$\hat{\mathbf{p}} = \frac{\hbar}{i} \nabla \tag{48}$$

can be given in the form

$$-\hbar^2 \nabla^2 \mathbf{\Phi} = (\boldsymbol{\sigma} \cdot \hat{\mathbf{p}})^2 \mathbf{\Phi}.$$
 (49)

In view of this result, the Klein-Gordon equation for the plasmariton [Eq. (23)] can be recast in the quantum-mechanical form

$$\left[\left(i\hbar\frac{\partial}{\partial t}\right)^2 + (c\hat{\mathbf{p}}\cdot\boldsymbol{\sigma})^2\right]\boldsymbol{\Phi}(\mathbf{r},t) = (Mc^2)^2\boldsymbol{\Phi}(\mathbf{r},t), \quad (50)$$

utilizing that  $c\hbar Q_C = \hbar\omega_p \equiv Mc^2$ , where *M* is the plasmariton "rest mass." The reader may notice that

$$\hat{h} \equiv \hat{p}^{-1}(\hat{\mathbf{p}} \cdot \boldsymbol{\sigma}) \tag{51}$$

is the plasmariton helicity operator (above in the **r**-representation).

# V. SECOND-QUANTIZED THEORY OF BULK PLASMARITONS

#### A. Lagrange-Hamilton formalism for free plasmaritons

Let us return to Eq. (26) and write it in the compact form

$$\mathbf{\Phi}(\mathbf{r},t) = \sum_{i=1,2} \mathbf{\Phi}_i(\mathbf{r},t), \tag{52}$$

where  $\Phi_i(\mathbf{r}, t)$  is the *i*th polarization species. The relativistic energy-momentum relation [Eq. (29)] simplifies the scalar Fourier amplitude,  $\Phi_i(\mathbf{q}, \omega)$ , to the form

$$\Phi_i(\mathbf{q},\omega) = 2\pi \Phi_i(\mathbf{q})\delta\left\{\omega - \left[(cq)^2 + \omega_p^2\right]^{1/2}\right\}$$
(53)

where  $\delta$ {· · · } is the Dirac delta function, so that

$$\Phi_{i}(\mathbf{r},t) = \int_{-\infty}^{\infty} \Phi_{i}(\mathbf{q}) \mathbf{e}_{i}(\hat{\mathbf{q}}) \exp(i\{\mathbf{q}\cdot\mathbf{r} - [(cq)^{2} + \omega_{p}]^{1/2}t\}) \frac{d^{3}q}{(2\pi)^{3}}.$$
 (54)

From the last member of Eq. (27) it follows that the two polarization species satisfy the generalized orthogonality condition

$$\int_{-\infty}^{\infty} \mathbf{\Phi}_1^*(\mathbf{r}, t) \cdot \mathbf{\Phi}_2(\mathbf{r}, t) d^3 r = 0.$$
 (55)

Since global (observable) plasmariton-field operators involve integrations over the entire space, the orthogonality condition implies that independent Lagrange-Hamilton formalisms can be established for each of the two free species.

In the following, we use the generic term  $\Psi(\mathbf{r}, t)$  for the two wave-function species  $\Phi_1(\mathbf{r}, t)$  and  $\Phi_2(\mathbf{r}, t)$ . We now resolve  $\Psi(\mathbf{r}, t)$  into Cartesian components, i.e.,

$$\Psi(\mathbf{r},t) = \sum_{i=1-3} \varepsilon_i \Psi_i(\mathbf{r},t), \qquad (56)$$

where  $\{\boldsymbol{\varepsilon}_i\}$ , i = 1-3, is a set of unit vectors  $(\boldsymbol{\varepsilon}_1 \times \boldsymbol{\varepsilon}_2 = \boldsymbol{\varepsilon}_3)$ . A related particle (*P*) Lagrangian density

$$\mathcal{L}_P = \sum_i \mathcal{L}_{Pi} = \sum_i \left[ (\partial^\mu \Psi_i) (\partial_\mu \Psi_i^*) - Q_C^2 \Psi_i \Psi_i^* \right]$$
(57)

inserted into the Euler-Lagrange equations

$$\partial_{\mu} \left[ \frac{\partial \mathcal{L}_{P}}{\partial (\partial_{\mu} \Psi_{i}^{*})} \right] - \frac{\partial \mathcal{L}_{P}}{\partial \Psi_{i}^{*}} = 0$$
(58)

leads to the Klein-Gordon equations for the three components of the free plasmariton wave function,

$$\left(\partial_{\mu}\partial^{\mu} + Q_{C}^{2}\right)\Psi_{i}(\mathbf{r},t) = 0, \quad i = 1, 2, 3.$$
 (59)

By means of Eqs. (56) and (57), the Klein-Gordon equation for  $\Psi(\mathbf{r}, t)$  then becomes

$$\left(\partial_{\mu}\partial^{\mu} + Q_{C}^{2}\right)\Psi(\mathbf{r},t) = \mathbf{0}.$$
(60)

To obtain the associated free plasmariton Hamiltonian density, one needs the six canonical momenta (i = 1-3)

$$\Pi_{\Psi_i} \equiv \frac{\partial \mathcal{L}_P}{\partial (\partial_0 \Psi_i^*)} = \partial^0 \Psi_i, \tag{61}$$

$$\Pi_{\Psi_i^*} \equiv \frac{\partial \mathcal{L}_P}{\partial (\partial_0 \Psi_i)} = \partial^0 \Psi_i^*.$$
(62)

In turn, with

$$\Pi_{\Psi_i}(\partial_0 \Psi_i^*) = \Pi_{\Psi_i^*}(\partial_0 \Psi_i) = (\partial^0 \Psi_i)(\partial_0 \Psi_i^*), \qquad (63)$$

the Hamiltonian density,

$$\mathcal{H}_P = \sum_{i=1-3} [\Pi_{\Psi_i}(\partial_0 \Psi_i^*) + \Pi_{\Psi_i^*}(\partial_0 \Psi_i) - \mathcal{L}_{Pi}], \qquad (64)$$

can be calculated. Written in "oscillator" form, one obtains

$$\mathcal{H}_P = \sum_{i=1-3} \left[ (\partial^0 \Psi_i) (\partial_0 \Psi_i^*) + (\nabla \Psi_i) \cdot (\nabla \Psi_i^*) + Q_C^2 \Psi_i \Psi_i^* \right].$$
(65)

The Hamiltonian density is extended to the operator level with the associations

$$\Psi_i \Rightarrow \hat{\Psi}_i, \quad \Psi_i^* \Rightarrow \hat{\Psi}_i^{\dagger}, \quad i = 1, 2, 3.$$
 (66)

From the integral

$$\hat{H}_P = \int_V \hat{\mathcal{H}}_P d^3 r \tag{67}$$

(here over a quantization volume V), the Hamilton operator related to the generic  $\Psi(\mathbf{r}, t)$  plasmariton field can be obtained following the standard procedure [18]. Thus  $(\hat{H}_P \Rightarrow \hat{H}_P^{\Psi})$ ,

$$\hat{H}_{p}^{\Psi} = \sum_{\mathbf{q}} \hbar \left[ (cq)^{2} + \omega_{p}^{2} \right]^{1/2} \left[ \hat{N}(\mathbf{q}) + \frac{1}{2} \right], \quad (68)$$

where

$$\hat{N}(\mathbf{q}) = \hat{a}^{\dagger}(\mathbf{q})\hat{a}(\mathbf{q}) \tag{69}$$

is the number operator belonging to the wave-vector  $\mathbf{q}$ ,  $\hat{a}(\mathbf{q})$  [ $\hat{a}^{\dagger}(\mathbf{q})$ ] being the annihilation (creation) operator for plasmariton quanta.

Our final result for the free bulk plasmariton Hamilton operator belonging to the sum of the two polarization species (index here denoted by s) hence is

$$\hat{H}_{P} = \sum_{\mathbf{q},s} \hbar \left[ (cq)^{2} + \omega_{p}^{2} \right]^{1/2} \left[ \hat{N}_{s}(\mathbf{q}) + \frac{1}{2} \right], \quad (70)$$

where

$$\hat{N}_s(\mathbf{q}) = \hat{a}_s^{\dagger}(\mathbf{q})\hat{a}_s(\mathbf{q}). \tag{71}$$

The free plasmariton momentum operator is given by

$$\hat{\mathbf{P}}_{P} = \sum_{\mathbf{q},s} \hbar \mathbf{q} \hat{N}_{s}(\mathbf{q}).$$
(72)

## VI. PLASMARITON INTERACTION WITH AN ELECTROMAGNETIC GAUGE FIELD

## A. Self-consistent four-potential in a sea of plasmaritons

At this point we may consider the jellium as an assembly of plasmaritons, remembering that we alone are interested in long-wavelength collective excitations with transverse polarizations. The plasmaritons are charged quasiparticles that interact mutually via electromagnetic couplings. Although the transverse vector potential in the plasma,  $\mathbf{A}_T(\mathbf{r}, t)$ , satisfies a diamagnetically driven Klein-Gordon-type wave equation [Eq. (44)], which is form-identical to that of the plasmariton wave function  $\Phi(\mathbf{r}, t)$  [Eq. (23)], the *only* "external" electromagnetic property of the plasmariton is its charge, Q = ne. As we shall realize in Sec. IX, a useful quantum physical picture of the plasmariton sees this as a transverse displacement field ( $\mathbf{D}_T$ ) coupled to a charged polarization oscillator resonating at a dressed plasma frequency ( $\tilde{\omega}_p = \sqrt{2}\omega_p$ ).

Let us assume that we excite the sea of plasmaritons by a prescribed external four-potential  $\{A_{\mu}^{ext}\}$ . The dynamics of the external field hence is determined by charged source particles outside the jellium system in consideration. Via its interaction with the plasmariton charges, the  $\{A_{\mu}^{ext}\}$ -field induces a field  $\{A_{\mu}^{ind}\}$  in the plasmariton sea. In a self-consistent manner, the plasmariton quasiparticle hence is driven effectively by a four-potential field

$$\{A_{\mu}\} = \{A_{\mu}^{\text{ext}}\} + \{A_{\mu}^{\text{ind}}\}.$$
 (73)

# B. Plasmaritonian interaction Lagrangian density

The covariant relativistic four-potential vector

$$\{A_{\mu}\} = \left(A_0 = \frac{U}{c}, \mathbf{A}\right),\tag{74}$$

where U and A are the scalar and vector potentials, respectively, and its contravariant partner [using here the "metric" signature (1, -1, -1, -1)] transfers via minimal coupling substitution for complex fields, with plasmariton charge Q, i.e.,

$$\partial_{\mu} \Rightarrow \partial_{\mu} - i \frac{Q}{\hbar} A_{\mu},$$
 (75)

$$\partial^{\mu} \Rightarrow \partial^{\mu} + i \frac{Q}{\hbar} A^{\mu},$$
 (76)

the Lagrangian density of the generic field  $\Psi(\mathbf{r}, t)$  [Eq. (57)] to  $\mathcal{L}_P + \mathcal{L}_I$ :

$$\mathcal{L}_{P} + \mathcal{L}_{I} = \sum_{i} (\mathcal{L}_{Pi} + \mathcal{L}_{Ii})$$

$$= \sum_{i} \left[ \left( \partial^{\mu} \Psi_{i} + i \frac{Q}{\hbar} A^{\mu} \Psi_{i} \right) \times \left( \partial_{\mu} \Psi_{i}^{*} - i \frac{Q}{\hbar} A_{\mu} \Psi_{i} * \right) - Q_{C}^{2} \Psi_{i} \Psi_{i}^{*} \right]. \quad (77)$$

The interaction Lagrangian density  $(\mathcal{L}_I)$  thus has the explicit form

$$\mathcal{L}_{I} = \sum_{i} \left\{ \frac{iQ}{\hbar} [(A^{\mu}\Psi_{i})(\partial_{\mu}\Psi_{i}^{*}) - (\partial^{\mu}\Psi_{i})(A_{\mu}\Psi_{i}^{*})] + \left(\frac{Q}{\hbar}\right)^{2} A^{\mu}A_{\mu}\Psi_{i}\Psi_{i}^{*} \right\}.$$
(78)

In compact notation one may write the expression for  $\mathcal{L}_{I}$  as follows:

$$\mathcal{L}_{I} = \frac{iQ}{\hbar} [(A^{\mu}\Psi) \cdot (\partial_{\mu}\Psi^{*}) - (\partial^{\mu}\Psi) \cdot (A_{\mu}\Psi^{*})] + \left(\frac{Q}{\hbar}\right)^{2} A^{\mu}A_{\mu}\Psi \cdot \Psi^{*},$$
(79)

remembering that the summation over upper and lower  $\mu$ -indices is kept implicit in the notation.

# C. Inhomogeneous plasmariton Klein-Gordon equation

From the three (i = 1-3) Euler-Lagrange equations written here in compact form as

$$\partial_{\mu} \left[ \frac{\partial (\mathcal{L}_{P} + \mathcal{L}_{I})}{\partial (\partial_{\mu} \Psi^{*})} \right] - \frac{\partial (\mathcal{L}_{P} + \mathcal{L}_{I})}{\partial \Psi^{*}} = \mathbf{0}$$
(80)

for the generic plasmariton field  $\Psi(\mathbf{r}, t) = \Phi_1(\mathbf{r}, t)$  or  $\Phi_2(\mathbf{r}, t)$ , one obtains the electromagnetically driven vectorial Klein-Gordon equation

$$\left(\partial_{\mu}\partial^{\mu} + Q_{C}^{2}\right)\Psi + \frac{\iota Q}{\hbar} \left[\partial_{\mu}(A^{\mu}\Psi) + A_{\mu}(\partial^{\mu}\Psi)\right] - \left(\frac{Q}{\hbar}\right)^{2} A_{\mu}A^{\mu}\Psi = \mathbf{0}.$$
 (81)

#### VII. PLASMARITONS WITH SPATIAL DISPERSION

## A. Hydrodynamic model

It is well known that a classical theory for wave propagation of bulk plasmons cannot be established unless one incorporates spatial nonlocality in the longitudinal dielectric function. In cases in which the solid (to a good approximation) can be considered as a translationally invariant medium (e.g., jellium), spatial nonlocality is commonly called spatial dispersion. In the wave-vector-frequency domain, many properties of bulk plasmons are well described in the framework of a hydrodynamic model, with  $\varepsilon_L(q, \omega)$  given by Eq. (4). In our recently developed wave-mechanical and second-quantized theories for bulk (and surface) plasmons, the hydrodynamic dispersion relation was taken as a starting point [17]. As we have seen in Secs. III–VI, a rigorous quantum theory for bulk plasmaritons can be developed without the inclusion of spatial dispersion. In fact, the overwhelming majority of classical plasmariton studies are carried out using a spatially local dielectric function. However, it is of theoretical importance to investigate the possibilities (and needs) for extending the quantum theory of plasmaritons to the nonlocal domain. The perhaps simplest spatially dispersive model is based on the hydrodynamic expression for  $\varepsilon_T(q, \omega)$ , given in Eq. (3).

As shown in Appendix A 3, the hydrodynamic expression for the transverse dielectric function [Eq. (3)] can be derived from the linearized Boltzmann equation. The designation "hydrodynamic" relates to the circumstance that all quantities are integrated over the electron velocity distribution, exemplified by the linearized (denoted here by subscript 1) current density

$$\mathbf{J}_{1}(\mathbf{r},t) = en \int_{-\infty}^{\infty} \mathbf{v} f_{1}(\mathbf{r},\mathbf{v},t) d^{3}v, \qquad (82)$$

whose  $(\mathbf{q}, \omega)$  representative appears in Eq. (9)'s classical small-q expression.

At this point, it is important to note that the frequency  $(\omega)$  and the wave number (q) are *independent* quantities in  $\varepsilon_T(q, \omega)$ . This implies that small q does not tell us anything about the  $\omega$ -values. In the hydrodynamic approach, branchcut contributions are associated with the singular points in  $\arctan(i/u)$  (see Appendix A 3), where

$$u = \frac{\omega}{qv_F} \tag{83}$$

in the collisionless limit (closed system) adopted herein. It follows from Eq. (A15) that the singular points appear at  $u = \pm 1$ . Physically, the branch cuts (two) describe the single-particle excitation spectrum. Including quantum interference effects, whose importance in the Lindhard theory is characterized by the parameter

$$z = \frac{q}{2k_F},\tag{84}$$

the singularities are located at  $u + v = \pm 1$  and  $u - v = \pm 1$ , cf. Eq. (A5). For  $z \to 0$  (the classical regime), the branch cuts extend from  $u = \pm \omega/(qV_F)$  to infinity  $(u \to \pm \infty)$ . The corner of the branch cut (for  $\omega > 0$ ) forms a straight line in the  $(\omega, q)$ -plane, viz.,

$$\omega = qv_F$$
, classical. (85)

Readers interested in a detailed analysis of the Lindhard dielectric functions may consult Ref. [8], where a comprehensive list of references to original works can also be found. In addition, Ref. [37] provides examples of Lindhard's theory extension to *nonlinear nonlocal* electrodynamics.

In the hydrodynamic model for charged gas plasma systems, the zeros of the denominators in Eqs. (3) and (4), viz.,

$$\omega = \begin{cases} \sqrt{\frac{1}{5}} v_F q, & T\text{-mode,} \\ \sqrt{\frac{3}{5}} v_F q, & L\text{-mode,} \end{cases}$$
(86)

characterize the onset of instabilities for the electron subsystem. In our jellium model, the ionic background is uniform and stationary. Such an approximation in general does not work in gas plasmas, where the (free) ions form a system that is essentially dynamic, yet is coupled strongly to the electron plasma in most cases.

Since  $(v_F/c)^2 \sim 10^{-4}$ , it turns out that the small-q dispersion relation for the plasmariton is given by (in squared form)

$$\omega^2 = (cq)^2 + \omega_p^2, \quad \omega > \omega_p.$$
(87)

This means that spatial dispersion is negligible. In the local regime, the dispersion relation in Eq. (87) is known as the Brewster branch [2].

In the classical theory of *surface plasmaritons*, an extra low-frequency dispersion relation, called the Fano branch, is present [2]. The surface plasmaritons mix in L- and T-dynamics. Inclusion of spatial dispersion in the L-dynamics (where it is of most importance) shows that the Fano branch is linear in the classical electrostatic limit presented herein. The Ritchie dispersion relation [2,17]

$$\sqrt{\frac{3}{5}} v_F q_{\parallel}^R = \omega \left[ 1 - \left( \frac{\omega_{pS}}{\omega} \right)^2 \right], \text{ classical}, \qquad (88)$$

where  $q_{\parallel}^{R}$  is the Ritchie (*R*) wave number parallel to the surface, and  $\omega_{pS} = \omega_p/\sqrt{2}$  is the surface *L*-plasmon resonance frequency in the limit  $q \rightarrow 0$ , is of central importance in electron scattering studies from surface plasmons. For  $\omega \gg \omega_{pS}$ , the Fano branch becomes linear, i.e.,

$$\omega^R = \sqrt{\frac{3}{5}} v_F q_{\parallel}^R. \tag{89}$$

The related quantum theory of the L-plasmon [17] was the springboard for our development of the bulk plasmariton quantum theory.

Although there is no Fano branch for bulk plasmaritons, it is interesting that the Ritchie relation in Eq. (89), and the branch-cut line in Eq. (85), are both linear, and with slopes  $\omega^R/q_{\parallel}^R = (3/5)^{1/2}v_F \sim v_F = \omega/q$  of the order of the Fermi velocity. We shall take up this aspect in our next paper on the quantum theory of surface plasmaritons.

Plots relating to the hydrodynamic model are shown in Fig. 2, using material parameters for doped free-electron-like *n*-InSb.

In the low-temperature limit ( $T \rightarrow 0$  K) and for  $\omega > \omega_p$ , the single-particle excitation domain occupies the region of the ( $\omega$ , q)-plane located between the two curves  $\hbar\omega =$  $(\hbar^2/m)(q^2/2 - qk_F)$  and  $\hbar\omega = (\hbar^2/m)(q^2/2 + qk_F)$ , where  $k_F$  is the Fermi wave number. These relations stem from the energy-momentum conversation related to scattering of an electron from state **k** to **k** + **q** by a photon of momentum  $\hbar$ **q**. For  $\hbar\omega < \varepsilon_F$ ,  $\varepsilon_F$  being the Fermi energy, the Pauli principle introduces a further restriction on the extension of the singleparticle excitation domain, as is well known [8]. Thus, the energy-momentum conservation can be expressed as

$$\frac{2m\omega}{\hbar} = q^2 + 2k_{\parallel}q,\tag{90}$$

where  $k_{\parallel} = \mathbf{k} \cdot \mathbf{q}/q$  is the component of the electron wave vector in the  $\mathbf{q}/q$  direction. The Pauli principle requires that the initial state must be a filled state ( $|\mathbf{k}| < k_F$ ) and the final

4.54.03.53.0  $3^{a_{2.5}}_{32.0}$ <sup>d</sup>3/0.04 6.02 3 1.50.00 1.0 $cq/\omega_p$ 0.50.02502 500 750 1000 4

FIG. 2. Illustration [in a normalized  $(q, \omega)$ -plane] of key elements of the classical Lindhard (Boltzmann) domain in relation to the bulk plasmariton theory: (i) Fully drawn thick black curve: Brewster branch, with asymptotic light line (broken curve). (ii) Green curve: unobservable *T*-plasmon. (iii) Violet curve (inset): linear branch-cut corner and (iv) in black (inset): The linear Fano branch part. (v) Gray toned domain: single-particle excitation region. Material parameters for *n*-InSb:  $m = 0.015m_0$  ( $m_0$ , free-electron mass) and  $n = 4 \times 10^{18}$  cm<sup>-3</sup>.

 $cq/\omega_p$ 

state an empty state  $(|\mathbf{k} + \mathbf{q}| > k_F)$ . Thus,

$$k_F^2 - \frac{2m\omega}{\hbar} \leqslant k_{\parallel}^2 + k_{\perp}^2 \leqslant k_F^2, \tag{91}$$

where  $k_{\perp}$  is the magnitude of the initial electron wave vector component perpendicular to **q**. The minimum value of  $k_{\perp}$ ,  $k_{\perp}^{\min}$ , is thus

$$k_{\perp}^{\min} = \left(k_F^2 - k_{\parallel}^2 - \frac{2m\omega}{\hbar}\right)^{1/2}.$$
 (92)

By eliminating  $k_{\parallel}^2$  from Eq. (92) using Eq. (90), and from the necessary condition  $k_{\perp}^{\min} \ge 0$ , it appears that the Pauli principle gives an extra boarder line curve

$$\frac{\hbar\omega}{\varepsilon_F} = 2\left(\frac{q}{k_F}\right) - \left(\frac{q}{k_F}\right)^2 \tag{93}$$

to the single-particle excitation below the plasma frequency; see Fig. 2.

## **B.** Electrostatic Lindhard model

The hydrodynamic plasmon and plasmariton models both have their roots in the classical Boltzmann transport equation, and as such the hydrodynamic theory cannot describe quantum interference effects. Quantum phenomena appear when the mode wave-vector magnitude (q) is comparable to (or larger than) the Fermi wave number ( $k_F$ ). In the Lindhard theory, quantum interference effects are included, thus the correctness of the hydrodynamic plasmariton dispersion relation must be determined from the Lindhard description of the transverse electrodynamics. The general Lindhard expression for  $\varepsilon_T(q, \omega)$  is given in Appendix A, where also a few of its





FIG. 3. Illustration [in a normalized  $(q, \omega)$ -plane] of the electrostatic Lindhard (essential quantum) domain in relation to the bulk plasmariton: (i) Fully drawn thick black curve: Electrostatic dispersion relation. (ii) Gray shaded domain: single-particle excitation domain. Material parameters for *n*-InSb:  $m = 0.015m_0$  ( $m_0$ , freeelectron mass) and  $n = 4 \times 10^{18}$  cm<sup>-3</sup>.  $\hbar \omega_p / \varepsilon_F = 0.99$ . The A  $\rightarrow$  B part of the electrostatic branch is an open window for undamped mode propagation (but a rigorous analysis requires that the changes coming from the replacement  $z \pm 1 \Rightarrow |z \pm u \pm 1|$  are included). The B  $\rightarrow$  C part is inside the single-particle domain, and the modes are strongly damped here. From C  $\rightarrow$  D (floating point) the waves are undamped. Beyond D, the quasiparticle quantum polarization oscillator (QO) model gives an accurate quantum description.

main properties are summarized. For large relative wave numbers  $(\geq q/k_F)$ , the dielectric *T*-function becomes essentially electrostatic. For a degenerate electron jellium, one obtains (see Appendix A)

$$\varepsilon_T(q,\omega) = 1 - \frac{3}{8} \left(\frac{\omega_p}{\omega}\right)^2 f(z), \text{ electrostatic,}$$
(94)

where, with  $z = q/(2k_F)$ ,

$$f(z) = z^{2} + 1 - \frac{[1 - z^{2}]^{2}}{2z} \ln \left| \frac{z + 1}{z - 1} \right|.$$
 (95)

In the electrostatic regime, there is no magnetic field present, and the dispersion relation is given by (see the plot in Fig. 3)

$$\varepsilon_T(q,\omega) = 0$$
, electrostatic, (96)

i.e., Eq. (19), which of course, as discussed in Sec. II C, is wrong when electromagnetic retardation effects are included. In explicit form, Eq. (96) can be given for  $z \gg 1$  as

$$\omega = \omega_p \left[ 1 - \frac{4}{5} \left( \frac{k_F}{q} \right)^2 \right]. \tag{97}$$

For large z, the Lindhard electrostatic dispersion relation approaches  $\omega_p$ . In this limit, the quantum theory is based on that of a "simple" harmonic oscillator.

#### C. Classical low-frequency Lindhard formula

The Boltzmann equation approach, used previously in linear and nonlinear response theory, is connected to the Lind-

hard theory for  $z \rightarrow 0$  (see Appendixes A 3 and A 5), and with  $u = \omega/(qv_F)$  one has

$$\varepsilon_T(q,\omega) = 1 - \frac{3}{2} \left(\frac{\omega_p}{\omega}\right)^2 f(u), \text{ classical,}$$
(98)

where

$$f(u) = 3u^{2} + 1 + u \ln \left| \frac{1 - u}{1 + u} \right|.$$
 (99)

For  $u \to \infty$ ,

$$f(u) = \frac{2}{3} + \frac{2}{15} \left(\frac{qv_F}{\omega}\right)^2 + O(q^4).$$
(100)

To order  $q^2$ , this results in a dispersion relation

$$cq(\omega) = \left[\frac{\omega^2 - \omega_p^2}{1 + \frac{1}{5} \left(\frac{\omega_p}{\omega}\right)^2 \left(\frac{v_F}{c}\right)^2}\right]^{1/2}.$$
 (101)

Here, we note that only for  $\omega > \omega_p$  is there a real solution for  $q(\omega)$ ; see also Fig. 2. Since  $(v_F/c)^2 \sim 10^{-4}$ , the last term is always negligible. The classical Lindhard model, therefore, reproduces the Brewster-branch dispersion relation, given in square form in Eq. (22). The Boltzmann theory (~ the classical Lindhard model) thus confirms that the transverse hydrodynamic theory is correct in the small-*q* collective mode region.

# VIII. REMARKS ON THE SECOND-QUANTIZED PLASMARITON THEORY WITH SPATIAL DISPERSION

#### A. Brewster branch

It has been realized that the inclusion of spatial dispersion in the dispersion relation of the Brewster branch is unimportant. The wave equation of the Brewster plasmariton hence is described by the Klein-Gordon theory presented in Sec. III A, and its extension to the QED level is given for the free plasmariton in Sec. V A, and for its interaction with an electromagnetic gauge field in Sec. VI A.

If one literally takes the words "Brewster plasmariton quasiparticle" as in classical theory, one would like to superimpose Brewster plane-wave modes to form a spatially well-localized quasiparticle. To have a stable quasiparticle propagating in space-time, one must avoid losses stemming from electron-hole pair excitations. These would result in an unstable Brewster plasmariton quasiparticle—a particle with a finite lifetime, so to speak. The Brewster dispersion relation reaches the single-particle border (*B*) at  $q = q_B$  determined by

$$\left[ (cq_B)^2 + \omega_p^2 \right]^{1/2} = \frac{\hbar}{m} \left( \frac{1}{2} q_B^2 - k_F q_B \right).$$
(102)

Vectorial free wave packets (more or less particlelike in the classical sense), here denoted by the generic name  $WP(\mathbf{r}, t)$ , hence are formed by superpositions of the type

$$\mathbf{WP}(\mathbf{r},t) = \int_0^{q_B} \mathbf{c}(\mathbf{q}) \exp\left(i\left\{\mathbf{q}\cdot\mathbf{r} - \left[(cq)^2 + \omega_p^2\right]t\right\}\right) \frac{dq^3}{(2\pi)^3},$$
(103)

where only the limit on the magnitude of  $\mathbf{q}$  is indicated in the limits for the 3D integration. Since

$$\nabla \cdot \mathbf{c}(\mathbf{q}) = 0, \tag{104}$$

the vectorial amplitude coefficients must satisfy the geometric transversality constraint

$$\hat{\mathbf{q}} \cdot \mathbf{c}(\mathbf{q}) = 0. \tag{105}$$

This constraint in itself puts a limit on the possibility for the spatial compression of a Brewster plasmariton quasiparticle. Thus, it is well known that one cannot, even if one lets  $q_B \rightarrow \infty$ , localize a transversely polarized wave packet better than to a size given by the transverse dyadic  $\delta$  function, given in spherical contraction by

$$\boldsymbol{\delta}_T(\mathbf{R}) = \frac{3\hat{\mathbf{R}}\hat{\mathbf{R}} - \mathbf{U}}{4\pi R^3}, \quad \mathbf{R} \neq \mathbf{0}$$
(106)

outside the center of confinement located at  $\mathbf{R} = \mathbf{0}$ . In Eq. (106), U is the 3 × 3 unit tensor, and  $\hat{\mathbf{R}} = \mathbf{R}/R$ . For the plasmariton confinement, spherical contraction has a privileged status, as for other types of vectorial boson particles, e.g., the transverse photon.

#### B. Quantum oscillator

Each of the high wave-number harmonic quantum oscillators, all belonging to the dispersion relation beyond the floating point D in Fig. 3, can for each frequency  $(\omega)$  form a localized oscillating single-plasmariton quasiparticle. As is well known from elementary electron dynamics, for example, a number state superposition can be constructed forming a coherent state, which during vibratory motion behaves much like a pseudoclassical particle in a harmonic potential. In the description presented in Sec. IX, where the transverse displacement field,  $\mathbf{D}_T$  (multiplied by -1), acts as canonical field momentum, the charged plasmariton oscillator behaves like a simple polarization oscillator with mass M = nm and resonance frequency  $\tilde{\omega}_p = \sqrt{2}\omega_p$ . The oscillator is surrounded by its tied T-photon cloud, and it oscillates in an "external" potential associated with the radiative part of the T-photon field. Further remarks on this point are given in Sec. X B.

# IX. PLASMARITON HAMILTONIAN WITH $-D_T$ AS CANONICAL FIELD MOMENTUM

# A. Charged jellium oscillator

It was realized in Sec. II that the microscopic polarization field, **P**, plays an important role in the plasmariton theory. In the long-wavelength limit ( $\mathbf{q} \rightarrow \mathbf{0}$ ), the time evolution of the jellium polarization is equivalent to that of a simple electrically charged harmonic oscillator with mass (M) and charge (Q) given by

$$M = nm, \quad Q = ne, \tag{107}$$

where n is the uniform electron density. A Lagrangian polarization density

$$\mathcal{L}_P = \frac{M}{2Q^2} \dot{\mathbf{P}}^2 - \frac{1}{2\varepsilon_0} \mathbf{P}^2$$
(108)

inserted into the Lagrange equation

$$\frac{d}{dt} \left( \frac{\partial \mathcal{L}_P}{\partial \dot{\mathbf{P}}} \right) - \frac{\partial \mathcal{L}_P}{\partial \mathbf{P}} = \mathbf{0}$$
(109)

leads to the correct oscillator equation for the polarization. In Eqs. (108) and (109),  $\dot{\mathbf{P}} \equiv d\mathbf{P}/dt$ , and Eq. (109) is a compact notation for the three Lagrange equations related to the Cartesian components of **P**. The vectorial quantity

$$\boldsymbol{\pi} \equiv \frac{\partial \mathcal{L}_P}{\partial \dot{\mathbf{P}}} = \frac{M}{Q^2} \dot{\mathbf{P}}$$
(110)

is the canonical momentum of the polarization field, thus  $\partial \mathcal{L}_P / \partial \mathbf{P} = (-1/\varepsilon_0) \mathbf{P}$ . Equation (109) gives the simple vectorial oscillator equation

$$\ddot{\mathbf{P}} + \omega_p^2 \mathbf{P} = 0, \tag{111}$$

where

$$\omega_p = \left(\frac{Q^2}{M\varepsilon_0}\right)^{1/2} = \left(\frac{ne^2}{m\varepsilon_0}\right)^{1/2} \tag{112}$$

is the plasma frequency, already introduced in Sec. II A. The Hamiltonian density of the polarization field, viz.,

$$\mathcal{H}_P \equiv \boldsymbol{\pi} \cdot \dot{\mathbf{P}} - \mathcal{L}_P, \tag{113}$$

thus takes the explicit oscillator form

$$\mathcal{H}_P = \frac{\boldsymbol{\pi} \cdot \boldsymbol{\pi}}{2MQ^2} + \frac{1}{2\varepsilon_0} \mathbf{P} \cdot \mathbf{P} = \frac{1}{Q^2} \left( \frac{\boldsymbol{\pi}^2}{2M} + \frac{1}{2}M\omega_p^2 \mathbf{P}^2 \right), \quad (114)$$

with "kinetic" and "potential" energies  $\pi^2/(2M)$  and  $(1/2)M\omega_p^2 \mathbf{P}^2$ , divided by  $Q^2$ , respectively.

# B. Lagrangian and Hamiltonian densities of the plasmariton field

In the following, we establish the Hamiltonian formalism for the plasmariton field following the standard procedure used for a coupled electron-photon system, remembering that we are interested only in the transversely polarized collective oscillations of the jellium. The total Lagrangian density

$$\mathcal{L} = \mathcal{L}_F + \mathcal{L}_P + \mathcal{L}_I \tag{115}$$

hence is the sum of a free electromagnetic field (F) part

$$\mathcal{L}_F = \frac{\varepsilon_0}{2} \{ [\dot{\mathbf{A}}_T(\mathbf{r}, t)]^2 - c^2 [\nabla \times \mathbf{A}_T(\mathbf{r}, t)]^2 \}, \qquad (116)$$

a plasma oscillator part, given by Eq. (108), and an interaction  $(\mathcal{L}_I)$  part. Usually one takes the interaction part in the form  $\mathbf{J} \cdot \mathbf{A}_T$ , but since one may add to  $\mathcal{L}_I$  a time derivative

$$\frac{d\mathcal{F}(\mathbf{r},t)}{dt} = -\frac{d}{dt}(\mathbf{P}\cdot\mathbf{A}_T) = -\mathbf{J}\cdot\mathbf{A}_T - \mathbf{P}\cdot\dot{\mathbf{A}}_T \qquad (117)$$

(remembering that  $\mathbf{J} = \dot{\mathbf{P}}$ ), without changing the physics, we make the equivalent choice

$$\mathcal{L}_I = -\mathbf{P} \cdot \dot{\mathbf{A}}_T. \tag{118}$$

In explicit form one hence has

$$\mathcal{L} = \frac{\varepsilon_0}{2} \left[ \dot{\mathbf{A}}_T^2 - c^2 (\mathbf{\nabla} \times \mathbf{A}_T)^2 \right] + \frac{M}{2Q^2} \dot{\mathbf{P}}^2 - \frac{1}{2\varepsilon_0} \mathbf{P}^2 - \mathbf{P} \cdot \dot{\mathbf{A}}_T.$$
(119)

Remembering that  $\mathbf{P} = \mathbf{P}_T$  for the transversely polarized polarization field, the momentum **M** conjugate to  $\mathbf{A}_T$  becomes

$$\mathbf{M} \equiv \frac{\partial \mathcal{L}}{\partial \dot{\mathbf{A}}_T} = \varepsilon_0 \dot{\mathbf{A}}_T - \mathbf{P}_T, \qquad (120)$$

and then

$$\mathbf{M} = -(\varepsilon_0 \mathbf{E}_T + \mathbf{P}_T) = -\mathbf{D}_T, \qquad (121)$$

where  $\mathbf{D}_T$  is the transverse part of the microscopic displacement field; cf. Eq. (7). In the plasmariton case,  $\mathbf{D}_T = \mathbf{D}$  of course. The canonical field is thus indirectly linked to the fact that the plasmariton is built from a *T*-photon and a *T*-plasmon (here without spatial dispersion included); cf. Fig. 1.

The total Hamiltonian density

$$\mathcal{H} \equiv \mathbf{M} \cdot \dot{\mathbf{A}}_T + \boldsymbol{\pi} \cdot \dot{\mathbf{P}} - \mathcal{L}$$
(122)

can now be expressed as a function of "coordinates" (**P**,  $\mathbf{A}_T$ ) and conjugate "momenta" ( $\boldsymbol{\pi}, \mathbf{M}$ ) eliminating ( $\dot{\mathbf{P}}, \dot{\mathbf{A}}_T$ ) by means of Eqs. (110) and (120). The explicit result is given by

$$\mathcal{H} = \left\{ \frac{1}{2\varepsilon_0} \mathbf{M}^2 + \frac{\varepsilon_0 c^2}{2} (\mathbf{\nabla} \times \mathbf{A}_T)^2 \right\} + \frac{1}{Q^2} \left[ \frac{\pi^2}{2M} + \frac{1}{2} M \omega_p^2 \mathbf{P}^2 \right] + \frac{1}{2\varepsilon_0} \mathbf{P}^2 + \frac{1}{\varepsilon_0} \mathbf{M} \cdot \mathbf{P}. \quad (123)$$

## C. Structure of the Hamiltonian density

To emphasize the structure of Eq. (123) we have put in two extra sets of brackets,  $\{\cdots\}$  and  $[\cdots]$ . The expression in  $\{\cdots\}$  may be characterized as a quasifree transverse photon Hamiltonian density, where the word "quasi" refers to the fact that it is the **D**<sub>T</sub>-field, and *not* the **E**<sub>T</sub>-field, that enters the field-momentum term. The expression in  $[\cdots]$ , with prefactor  $Q^{-2}$ , is the Hamiltonian density of the charged jellium oscillator discussed in Sec. IX A. The term  $\mathbf{P}^2/(2\varepsilon_0)$ , which only depends on the polarization density variables, is to be grouped with the oscillator part. Since

$$\frac{1}{Q^2} \left[ \frac{\boldsymbol{\pi}^2}{2M} + \frac{1}{2} M \omega_p^2 \mathbf{P}^2 \right] + \frac{1}{2\varepsilon_0} \mathbf{P}^2$$
$$= \frac{1}{Q^2} \left[ \frac{\boldsymbol{\pi}^2}{2M} + \frac{1}{2} M \tilde{\omega}_p^2 \mathbf{P}^2 \right], \qquad (124)$$

where

$$\tilde{\omega}_p = \sqrt{2}\omega_p, \tag{125}$$

it appears that we still have oscillator dynamics, yet with a dressed resonance frequency. The term  $\mathbf{M} \cdot \mathbf{P}/\varepsilon_0$  in Eq. (123) is an interaction term between the quasifree photon and the dressed oscillator.

The reader may notice that the Hamiltonian density in Eq. (123) bears a formal similarity to the Power-Zienau-Woolley (PZW) Hamiltonian density, used to study the quantum electrodynamics of strongly localized particle distributions (atoms, molecules, etc.) in a multipole expansion scheme [38–41]. In the PZW Hamiltonian, **P** is the electric dipole moment per unit volume. In the present work, the microscopic **P** includes all multipole orders, cf. the discussion in Sec. II B. Also in a theoretical work by one of the authors (Keller), which in a propagator formalism describes



FIG. 4. Schematic illustration of how the tied *T*-photon principle can occur in different physical systems. All tied *T*-photon clouds in violet (gray shading). (a) Nonrelativistic Bethe cloud renormalization of the bare electron mass  $m_0$  to  $m = m_0 + \delta m$  ( $\delta m$  finite due to a high-frequency  $\omega$ -mode cutoff at the relativistic limit  $m_0c^2/\hbar$ ) [42]. (b) Tied *T*-photon cloud as it appears in Keller propagator theory describing the quantized light emission from a single-electron atom. The black circle (sphere) gives the trailing edge of the radiated field, propagating outward (as indicated by the arrows) with the vacuum speed of light. The best possible *T*-photon localization is determined by the tied *T*-photon cloud [29,30]. (c) *T*-photon cloud tied to a localized plasmariton quasiparticle (see also Fig. 1).

the spatial confinement of quantized light emitted from an atom, a "particle" term  $\mathbf{P}_T/(3\varepsilon_0)$  appears [29]. Physically this term relates to the transverse self-field  $\mathbf{E}_T^{SF}$  attached to the atom [ $\mathbf{E}_T^{SF} = -\mathbf{P}_T/(3\varepsilon_0)$ ]. A schematic illustration of the Bethe mass renormalization [42], Keller's propagator theory of spatial photon confinement [29,30], and the plasmariton quasiparticle with its tied *T*-photon are shown in Fig. 4.

## D. Quantization of integrated Hamiltonian density

The Hamiltonian density in Eq. (123) is extended to the operator level  $(\mathcal{H} \rightarrow \hat{\mathcal{H}})$  by replacing the vectors  $\mathbf{A}_T$ ,  $\mathbf{M}$ , and  $\mathbf{P}(=\mathbf{P}_T)$  by operators  $\hat{\mathbf{A}}_T$ ,  $\hat{\mathbf{M}}$ , and  $\hat{\mathbf{P}}$ . From the standard theory

$$\hat{\mathbf{A}}_{T}(\mathbf{r}) = \sum_{\mathbf{q},s} \left(\frac{\hbar}{2\varepsilon_{0}cqV}\right)^{1/2} \hat{\boldsymbol{\varepsilon}}_{s}(\hat{\mathbf{q}}) \left[\hat{a}_{\mathbf{q}s}e^{i\mathbf{q}\cdot\mathbf{r}} + \hat{a}_{\mathbf{q}s}^{\dagger}e^{-i\mathbf{q}\cdot\mathbf{r}}\right].$$
(126)

In Eq. (126) the transverse vector potential is expanded in plane-wave modes (wave vectors: **q**) over a quantization volume *V*. The two polarization states (s = 1, 2) belonging to a given  $\hat{\mathbf{q}} = \mathbf{q}/q$  direction are characterized here by the real orthogonal polarization vectors  $\boldsymbol{\epsilon}_s(\mathbf{q})$ , s = 1, 2. The mode annihilation ( $\hat{a}_{\mathbf{q},s}$ ) and creation ( $\hat{a}_{\mathbf{q}s}^{\dagger}$ ) operators obey the boson commutator relations

$$[\hat{a}_{\mathbf{q}s}, \hat{a}_{\mathbf{q}'s'}^{\mathsf{T}}] = \delta_{ss'} \delta_{\mathbf{q},\mathbf{q}'}, \qquad (127)$$

$$[\hat{a}_{\mathbf{q}s}, \hat{a}_{\mathbf{q}'s'}] = [\hat{a}_{\mathbf{q}s}^{\dagger}, \hat{a}_{\mathbf{q}'s'}^{\dagger}] = 0.$$
(128)

The quantization of the canonical *free* field momentum,  $\mathbf{M} = -\mathbf{D}_T = -i\varepsilon_0 \mathbf{E}_T$ , has the same plane-wave expansion as the electric field operator [18] multiplied by  $-i\varepsilon_0$ . Thus,

$$\hat{\mathbf{M}}(\mathbf{r}) = -i\varepsilon_0 \sum_{\mathbf{q},s} \left(\frac{\hbar cq}{2\varepsilon_0 V}\right)^{1/2} \boldsymbol{\varepsilon}_s(\hat{\mathbf{q}}) \left[\hat{a}_{\mathbf{q}s} e^{i\mathbf{q}\cdot\mathbf{r}} - \hat{a}_{\mathbf{q}s}^{\dagger} e^{-i\mathbf{q}\cdot\mathbf{r}}\right].$$
(129)

By integration of the field Hamiltonian density over the quantization volume, and by use of the relation

$$\frac{1}{V} \int_{V} e^{i(\mathbf{q}-\mathbf{q}')\cdot\mathbf{r}} d^{3}r = \delta_{\mathbf{q},\mathbf{q}'}, \qquad (130)$$

where  $\delta_{\mathbf{q},\mathbf{q}'}$  is the Kronecker delta, the field part of the Hamilton operator

$$\hat{H}_F = \int_V \hat{\mathcal{H}}_F d^3r \tag{131}$$

takes the expected "sum of oscillator" form [18]

$$\hat{H}_F = \sum_{\mathbf{q},s} \hbar c q \left( \hat{a}_{\mathbf{q}s}^{\dagger} \hat{a}_{\mathbf{q}s} + \frac{1}{2} \right).$$
(132)

For the free field, the zero-point energy plays a role, e.g., for the number states. Thus the expectation value of  $\mathbf{D}_T$  vanishes, whereas the expectation value of the intensity operator  $(\sim \mathbf{D}_T \cdot \mathbf{D}_T)$  fluctuates about its zero ensemble average. These fluctuations are nonzero even for the vacuum state.

The charged jellium oscillator model used in Sec. IX A relates to a spatially nondispersive dielectric function [Eq. (2)]. This implies that the scalar amplitude ( $P_0$ ) of a propagating polarization with wave vector (**q**) must be independent of **q**. As a consequence, the polarization operator can be expanded in plane-wave modes as follows:

$$\hat{\mathbf{P}}(\mathbf{r}) = \frac{P_0}{V^{1/2}} \sum_{\mathbf{q},s} \boldsymbol{\epsilon}_s(\hat{\mathbf{q}}) [\hat{b}_{\mathbf{q}s} e^{i\mathbf{q}\cdot\mathbf{r}} + \hat{b}_{\mathbf{q}s}^{\dagger} e^{-i\mathbf{q}\cdot\mathbf{r}}].$$
(133)

The polarization-field mode annihilation  $(\hat{b}_{qs})$  and creation  $(\hat{b}_{qs}^{\dagger})$  operators satisfy the boson commutator relations

$$[\hat{b}_{\mathbf{q}s}, \hat{b}^{\dagger}_{\mathbf{q}'s'}] = \delta_{ss'}\delta_{\mathbf{q},\mathbf{q}'}, \qquad (134)$$

$$[\hat{b}_{\mathbf{q}s}, \hat{b}_{\mathbf{q}'s'}] = [\hat{b}_{\mathbf{q}s}^{\dagger}, \hat{b}_{\mathbf{q}'s'}^{\dagger}] = 0.$$
(135)

The Hamilton operator of the polarization field,

$$\hat{H}_P = \int_V \mathcal{H}_P d^3 r, \qquad (136)$$

necessarily must attain the "sum of nondispersive oscillator" form, viz.,

$$\hat{H}_P = \hbar \tilde{\omega}_P \sum_{\mathbf{q},s} \left( \hat{b}_{\mathbf{q}s}^{\dagger} \hat{b}_{\mathbf{q}s} + \frac{1}{2} \right).$$
(137)

In Appendix B, we show that the scalar amplitude is given by

$$P_0 = (\varepsilon_0 \hbar \tilde{\omega}_p)^{1/2}.$$
 (138)

The interaction is given by the Hamilton operator

$$\hat{H}_{I} = \int_{V} \mathcal{H}_{I} d^{3}r = \frac{1}{\varepsilon_{0}} \int_{V} \hat{\mathbf{M}} \cdot \hat{\mathbf{P}} d^{3}r.$$
(139)

Introducing, tentatively, the abbreviation

$$K(q) = -iP_0 \left(\frac{\hbar cq}{2\varepsilon_0}\right)^{1/2},$$
(140)

one has

$$\hat{H}_{I} = \sum_{\mathbf{q},\mathbf{q}',s,s'} \frac{K(q)}{V} \int_{V} \boldsymbol{\varepsilon}_{s}(\hat{\mathbf{q}}) \cdot \boldsymbol{\varepsilon}_{s'}(\hat{\mathbf{q}}') [\hat{a}_{\mathbf{q}s}e^{i\mathbf{q}\cdot\mathbf{r}} - \hat{a}_{\mathbf{q}s}^{\dagger}e^{-i\mathbf{q}\cdot\mathbf{r}}] \\ \times [\hat{b}_{\mathbf{q}'s'}e^{i\mathbf{q}'\cdot\mathbf{r}} + \hat{b}_{\mathbf{q}'s'}^{\dagger}e^{-i\mathbf{q}'\cdot\mathbf{r}}] d^{3}r.$$
(141)

By utilizing Eq. (140) (for  $\mathbf{q}' = \mathbf{q}, -\mathbf{q}$ ), integration over the quantization volume results in

$$\hat{H}_{I} = \sum_{\mathbf{q},s,s'} K(q) [\boldsymbol{\varepsilon}_{s}(\hat{\mathbf{q}}) \cdot \boldsymbol{\varepsilon}_{s'}(-\hat{\mathbf{q}}) \hat{a}_{\mathbf{q}s} \hat{b}_{-\mathbf{q}s'} + \boldsymbol{\varepsilon}_{s}(\hat{\mathbf{q}}) \cdot \boldsymbol{\varepsilon}_{s'}(\hat{\mathbf{q}}) \hat{a}_{\mathbf{q}s} \hat{b}_{\mathbf{q}s'}^{\dagger} - \boldsymbol{\varepsilon}_{s}(\hat{\mathbf{q}}) \cdot \boldsymbol{\varepsilon}_{s'}(\hat{\mathbf{q}}) \hat{a}_{\mathbf{q}s}^{\dagger} \hat{b}_{\mathbf{q}s'} - \boldsymbol{\varepsilon}_{s}(\hat{\mathbf{q}}) \cdot \boldsymbol{\varepsilon}_{s}(-\hat{\mathbf{q}}) \hat{a}_{\mathbf{q}s}^{\dagger} \hat{b}_{-\mathbf{q}s'}^{\dagger}].$$
(142)

The orthogonality of the polarization unit vectors reduces Eq. (142) to

$$\hat{H}_{I} = iP_{0} \sum_{\mathbf{q},s} \left(\frac{\hbar cq}{2\varepsilon_{0}}\right)^{1/2} \times [\hat{a}_{\mathbf{q}s}^{\dagger}\hat{b}_{\mathbf{q}s} + \hat{a}_{\mathbf{q}s}^{\dagger}\hat{b}_{-\mathbf{q}s}^{\dagger} - \hat{a}_{\mathbf{q}s}\hat{b}_{-\mathbf{q}s} - \hat{a}_{\mathbf{q}s}\hat{b}_{\mathbf{q}s}^{\dagger}].$$
(143)

If one changes **q** to  $-\mathbf{q}$  in the summations over the terms containing  $\hat{b}_{-\mathbf{q}s}^{\dagger}$  and  $\hat{b}_{-\mathbf{q}s}$ , and uses that [see Eqs. (125) and (138)]

$$P_0\left(\frac{\hbar cq}{2\varepsilon_0}\right)^{1/2} = (\varepsilon_0 \hbar \tilde{\omega}_p)^{1/2} \left(\frac{\hbar cq}{2\varepsilon_0}\right)^{1/2} = \hbar (cq\omega_p)^{1/2}, \quad (144)$$

one finally obtains

$$\hat{H}_{I} = i\hbar \sum_{\mathbf{q},s} (cq\omega_{p})^{1/2}$$

$$\times [\hat{a}^{\dagger}_{\mathbf{q}s}\hat{b}_{\mathbf{q}s} + \hat{a}^{\dagger}_{-\mathbf{q}s}\hat{b}^{\dagger}_{\mathbf{q}s} - \hat{a}_{-\mathbf{q}s}\hat{b}_{\mathbf{q}s} - \hat{a}_{\mathbf{q}s}\hat{b}^{\dagger}_{\mathbf{q}s}]. \quad (145)$$

By gathering the results in Eqs. (132), (137), and (143), it appears that the total plasmariton Hamiltonian operator, expressed in terms of the boson annihilation and creation



FIG. 5. Schematic illustration of the  $D_T$ -photon  $\leftrightarrow$  dressed oscillator scattering and two-particle absorption/emission processes appearing in the interaction Hamiltonian operator [Eq. (146)]. Oscillator quanta (black), light quanta (gray). (a) Absorption (emission) of oscillator (light) quanta. (b) Opposite process to that in (a). (c) [(d)] Two-particle absorption (generation) processes.

operators, becomes

$$\begin{aligned} \hat{H} &= \sum_{\mathbf{q},s} \left\{ \hbar cq \left[ \hat{a}_{\mathbf{q}s}^{\dagger} \hat{a}_{\mathbf{q}s} + \frac{1}{2} \right] + \hbar \tilde{\omega}_{p} \left[ \hat{b}_{\mathbf{q}s}^{\dagger} \hat{b}_{\mathbf{q}s} + \frac{1}{2} \right] \\ &+ \frac{i\hbar}{\sqrt{2}} (cq \tilde{\omega}_{p})^{1/2} [\hat{a}_{\mathbf{q}s}^{\dagger} \hat{b}_{\mathbf{q}s} + \hat{a}_{-\mathbf{q}s}^{\dagger} \hat{b}_{\mathbf{q}s}^{\dagger} \\ &- \hat{a}_{-\mathbf{q}s} \hat{b}_{\mathbf{q}s} - \hat{a}_{\mathbf{q}s} \hat{b}_{\mathbf{q}s}^{\dagger} ] \right\}. \end{aligned}$$
(146)

The elementary scattering processes appearing in the interaction Hamiltonian operator are shown in the diagrams of Fig. 5.

#### E. Diagonalization of the Hamiltonian

In the displacement field quantization scheme, the eigenmodes and their dispersion relation are obtained by diagonalizing the Hamiltonian operator in Eq. (146). The form of the interaction term suggests that an annihilation operator ( $\hat{\alpha}_{qs}$ ) ansatz

$$\hat{\alpha}_{\mathbf{q}s} \equiv w\hat{a}_{\mathbf{q}s} + x\hat{b}_{\mathbf{q}s} + y\hat{a}^{\dagger}_{-\mathbf{q}s} + z\hat{b}^{\dagger}_{-\mathbf{q}s}, \qquad (147)$$

where w, x, y, and z are as yet unknown constants, will do the job. From the Heisenberg equation of motion for  $\hat{\alpha}_{qs}$ , the diagonalization is obtained from the condition

$$[\hat{\alpha}_{\mathbf{q}s}, \hbar^{-1}\hat{H}] = \omega_{\mathbf{q}s}\hat{\alpha}_{\mathbf{q}s}.$$
 (148)

The relation in Eq. (148) gives the values of the four constants. See also the landmark work of Hopfield on polaritons [43].

A few steps for the determination of the constants now follows. Thus, one obtains

$$[\hat{\alpha}_{\mathbf{q}s}, \hbar^{-1}\hat{H}_F] = cq(w\hat{a}_{\mathbf{q}s} - y\hat{a}_{-\mathbf{q}s}^{\dagger}), \qquad (149)$$

$$[\hat{\alpha}_{\mathbf{q}s}, \hbar^{-1}\hat{H}_P] = \tilde{\omega}_p(x\hat{b}_{\mathbf{q}s} - z\hat{b}_{-\mathbf{q}s}^{\dagger}), \qquad (150)$$

and

$$\begin{aligned} [\hat{\alpha}_{\mathbf{q}s}, \hbar^{-1}\hat{H}_{I}] \\ &= \frac{i}{\sqrt{2}} (cq\tilde{\omega}_{p})^{1/2} [w\hat{b}_{\mathbf{q}s} + x(\hat{a}_{-\mathbf{q}s}^{\dagger} - \hat{a}_{\mathbf{q}s}) + y\hat{b}_{\mathbf{q}s}]. \end{aligned} (151)$$

A combination of Eqs. (147)–(151) results in the set of homogeneous equations among the unknown constants:

$$\begin{pmatrix} cq - \omega & -A & 0 & 0 \\ A & \tilde{\omega}_p - \omega & A & 0 \\ 0 & A & -(cq + \omega) & 0 \\ 0 & 0 & 0 & -(\omega_p + \omega) \end{pmatrix} \begin{pmatrix} w \\ x \\ y \\ z \end{pmatrix}$$
$$= \begin{pmatrix} 0 \\ 0 \\ 0 \\ 0 \\ 0 \end{pmatrix}, \qquad (152)$$

with the abbreviation  $A = (i/\sqrt{2})(cq\tilde{\omega}_p)^{1/2}$ . Setting the determinant to zero, i.e.,

$$\begin{vmatrix} cq - \omega & -A & 0 & 0 \\ A & \tilde{\omega}_p - \omega & A & 0 \\ 0 & A & -(cq + \omega) & 0 \\ 0 & 0 & 0 & -(\omega_p + \omega) \end{vmatrix} = 0, \quad (153)$$

and remembering that only  $\omega > 0$  values are of interest, one obtains the following implicit form of the dispersion relation:

$$\omega^2 = \omega \tilde{\omega}_p + (cq)^2, \qquad (154)$$

with  $\omega_{qs} \equiv \omega$ . Although one cannot expect that the dispersion relation in the "dressed oscillator" picture is identical to the one in Eq. (22), they have in common the large-*q* linear form  $\omega = cq$ , and they both approach their relevant plasma frequencies  $\omega = \tilde{\omega}_p$  and  $\omega = \omega_p$  for  $q \to 0$ .

In quantum optics (of atoms and solids), it is often justified to neglect energy nonconserving processes in resonant fieldmatter interaction studies [28,31,44–46]. The approximation is known as the rotating-wave approximation (RWA). In the plasmariton analysis, these processes appear in the terms of the interaction Hamiltonian part of  $\hat{H}$  [Eq. (146)], which contain the operator products  $\hat{a}_{-qs}^{\dagger}\hat{b}_{qs}^{\dagger}$  [Fig. 5(d)] and  $\hat{a}_{-qs}\hat{b}_{qs}$ [Fig. 5(c)]. In the RWA, the y and z parts of the  $\hat{\alpha}_{qs}$  ansatz [Eq. (147)] are omitted. Setting the related 2 × 2 determinant,  $(cq - \omega)(\tilde{\omega}_p - \omega) + A^2$ , to zero, one obtains the RWA dispersion relation

$$\omega_{\text{RWA}} = \frac{1}{2} \{ \tilde{\omega}_p + cq + \left[ \tilde{\omega}_p^2 + (cq)^2 \right]^{1/2} \}.$$
 (155)

In Fig. 6, the difference between the general dispersion relation [obtained from Eq. (154)], i.e.,

$$\omega = \frac{1}{2} \{ \tilde{\omega}_p + \left[ \tilde{\omega}_p^2 + (2cq)^2 \right]^{1/2} \},$$
(156)

and its RWA form are plotted (in normalized form).

Knowledge of the dispersion relation  $\omega_{qs} = \omega_{qs}(q)$  allows one to obtain (up to a normalization constant) the eigenvector (w, x, y, z). For the present purpose, we do not need the explicit expression for the eigenvector. Since the charged oscillator model relates to a spatially nondispersive dielectric function, the dispersion relation only has a Brewster branch.



FIG. 6. Difference between the RWA  $[\omega_{RWA}(cq)]$  and the general  $[\omega(cq)]$  dispersion relations in a plot where frequencies and wave number (multiplied by c) are normalized to  $\tilde{\omega}_p = \sqrt{2}\omega_p$ .

## X. CONNEXUM

## A. Summary

In the present paper, wave-mechanical (first-quantized) and quantum electrodynamic (QED) theories of bulk plasmaritons in a homogenous jellium have been established. After having reviewed elements of the classical plasmariton dispersion relation, in particular the transverse (T) Lindhard dielectric function, the eigenmodes condition, and the "inner" structure of the plasmariton, a quasiparticle built from a T-photon and a T-plasmon, we turn our attention toward the formulation of a first-quantized theory for plasmaritons. Starting from the relativistic energy-momentum relation for the quasiparticle, a relation that leads to Klein-Gordon equations for each of the two plasmariton helicity species, we establish a dynamical (first-order in time) evolution equation for a single plasmariton. The analytical part of the plasmariton wave function satisfies a Schrödinger-like equation with a Hamiltonian resembling that used by Landau and Peirls [9] in 1930 in one of the first attempts to formulate a wave-mechanical description for photons. Seen in a somewhat different perspective, the plasmariton appears as a diamagnetically driven spin-1 particle. From a Lagrange-Hamilton formulation for free plasmaritons, we reach a QED theory for the plasmariton quasiparticle. From there, the fundamental minimal coupling principle allows us to describe the plasmariton's interaction with an electromagnetic gauge field (T-photon) in terms of an electromagnetically driven (inhomogeneous) Klein-Gordon equation.

In most cases, it is sufficient to be in possession of a spatially local plasmariton theory. Nevertheless, it is fundamentally important to understand in which manner the theory for bulk plasmaritons is affected in the presence of spatial dispersion. For this purpose, we use the Lindhard (random-phase-approximation) dielectric function for propagating transverse modes. The Lindhard formalism leads in the wave vector (**q**)-frequency ( $\omega$ ) domain to collective electron modes at low and high wave numbers (*q*). For wave numbers

much smaller than the electron Fermi wave number  $(k_F)$ , a classical Lindhard theory (without quantum interference phenomena) becomes identical to a theory established on the basis of the Boltzmann equation. The low-wave-vector collective plasmariton modes are obtained expanding the transverse dielectric function from q = 0 to order  $q^2$ . The often phenomenologically introduced hydrodynamic theory for transverse excitations follows rigorously from the microscopic theory, at least well known for longitudinal bulk plasmons. The hydrodynamical theory leads to a dispersion relation for bulk plasmaritons which has only one branch: a Brewster branch with real wave-number (q) modes only above the bulk plasma frequency at the q = 0 limit. The low-wavenumber plasmariton modes satisfy to an extreme degree of accuracy a Klein-Gordon wave equation, and the plasmariton quasiparticles emerge via the usual quantization scheme for a vectorial Klein-Gordon equation. At relative large wave numbers  $q/(2k_F) \ge 1$ , quantum effects dominate the Lindhard theory, and a density matrix formalism replaces the Boltzmann approach. For sufficiently large relative wave numbers, an electrostatic description can be used since magnetic effects become unimportant, and for  $q/k_F \gg 1$ , the quantization scheme is identical to that of "simple" nonpropagating oscillator modes.

The quasiparticle picture of an electron surrounded by a dynamic cloud of transverse photons is well-established in QED. In the atomic case, Bethe showed on a nonrelativistic basis (with photon modes cut off at the relativistic frequency  $mc^2/\hbar$ ) how the photon cloud renormalizes the electron mass. Qualitatively, the tied photon cloud is in Bethe's theory [42] related to two closed quantum processes: (i) an electron in state  $|a\rangle$  instantaneously emits and reabsorbs a T-photon, and (ii) the electron in state  $|a\rangle$  makes a transition to state  $|b\rangle$  by emitting a photon. Later the electron in state  $|b\rangle$  reabsorbs the photon and returns to state  $|a\rangle$ . These radiative processes lead to a dynamic renormalization of the electron mass, as discussed in detail in Ref. [28]. In this work, we have established a formally somewhat similar theory for the plasmariton quasiparticle on the basis of a Hamiltonian formalism in which the transverse microscopic displacement field  $\mathbf{D}_T$  (multiplied by minus one) plays the role of canonical field momentum. The total Hamiltonian density here is expressed in terms of "coordinates" (**P**, **A**<sub>T</sub>) and conjugate momenta ( $\pi = M\mathbf{P}/Q^2$ , **M** =  $-\mathbf{D}_T$ ) for the polarization and photon fields. The pair ( $\mathbf{P}, \boldsymbol{\pi}$ ) describes the charged plasmariton quasiparticle, with mass M = nm and charge Q = ne, n being the electron density in the bulk jellium. The T-photon cloud is stationary (tied) if only the long-wavelength T-photon mode is included in the analysis. The quasiparticle oscillator has a renormalized plasma frequency  $\tilde{\omega}_p = \sqrt{2}\omega_p$ ,  $\omega_p$  being the bulk plasma frequency at q = 0. The plasmariton quasiparticle part of the Hamiltonian now has an "extra" term  $[\mathbf{P}_T^2/(2\varepsilon_0)]$  relating to its self-field energy. In a propagator description of quantized light emitted from an atom, the limit for the spatial confinement of the *T*-photons is determined by a "particle" term  $\mathbf{P}_T(3\varepsilon_0)$ . Physically, this term relates to the transverse self-field  $\mathbf{E}_{SF} =$  $-\mathbf{P}_T/(3\varepsilon_0)$  attached to the atom.

The main body of our paper is finished with a description of the integrated Hamiltonian density in the  $D_T$ -formalism. An analytic diagonalization of the total Hamilton operator is

possible, and this results in an explicit and simple expression for the bulk plasmariton dispersion relation in the  $D_T$ -picture.

Different equivalent Hamiltonians can be used to study bulk plasmaritons in QED. In Sec. IX we used a Hamiltonian based on the  $D_T$ -formalism, because this Hamiltonian is particularly adequate for renormalization analyses, e.g., it is used to calculate the interaction between two localized systems of charges (atomic or molecular). In this case, the renormalized (PZW) interparticle self-field interaction plays a crucial role [18,38–40]. To obtain the transition to the classical plasmariton theory, it appears preferable to start from the Coulomb gauge, because the Maxwell equations among the field operators in this gauge are form-identical to the classical Maxwell equations. Although the analysis is beyond the scope of this work, the QED theory of coherent states with average plasmariton number much larger than 1 ultimately will link the Coulomb QED and classical dispersion relations.

The equality of physical (observational) predictions in two different representations, j = 1, 2, basically is linked to the identity of the absolute square of the transition matrix elements,

$$\left| \left\langle \psi_{f}^{(1)} \right| \hat{U}^{(1)}(t_{f}, t_{i}) \left| \psi_{i}^{(1)} \right\rangle \right|^{2} = \left| \left\langle \psi_{f}^{(2)} \right| \hat{U}^{(2)}(t_{f}, t_{i}) \left| \psi_{i}^{(2)} \right\rangle \right|^{2},$$
(157)

where  $\hat{U}^{(j)}(t_f, t_i)$ , j = 1, 2, is the evolution operator relating the state vectors at the initial (*i*) and final (*f*) times, i.e.,

$$|\psi^{(j)}(t_f)\rangle = \hat{U}^{(j)}(t_f, t_i)|\psi^{(j)}(t_i)\rangle.$$
(158)

At the time of writing, we are not aware of any QED experiments on plasmaritons. Light scattering experiments appear as a promising possibility, and they have been carried out on plasmons in the long-wavelength limit; see, e.g., the landmark work of Patel and Slusher [1] and the papers on doped semiconductor plasmas [21]; see also Refs. [47,48] on light scattering by polaritons and in crystals as such.

# B. Outlook—Relativistic plasmariton quasiparticle: Inadequacy of the concept "field tied to a particle"

# 1. Electron

Simultaneously with the Pauli-Fierz theory, Kramers attempted to construct first a classical, then a quantum theory, in which the tied and radiated parts of the electromagnetic field are constantly distinguishable [49,50]. Kramers' idea of renormalization of the electron mass came from his uneasiness with the precise meaning of the mass concept in Dirac's radiation theory [51]. Kramers' struggle with the construction of a nonrelativistic theory culminated (and terminated) with Van Kampen's analysis [51]. It was Bethe's great insight to recognize that Kramers' ideas were the key to understanding the level shift observed by Retherford and Lamb. Bethe, in a nonrelativistic setting, came to the conclusion that the Lamb shift originates in the difference between the *infinite* self-energies of an electron bound in an atom and that of a free electron. His finally obtained renormalized electron mass gave an impressive agreement with the observed Lamb shift between the  $2s_{1/2}$  and  $2p_{1/2}$  states in hydrogen (experimental value: ~1.057 MHz; Bethe: ~1.040 MHz) [42].

To complete the "renormalization program," the electron must be described relativistically (in a way involving covariant variables). Finding a transformation that separates tied and radiated field parts becomes impossible. From a physical point of view, it is on the *probability amplitude* of the various processes and not the field itself that one has to separate the electromagnetic mass and the production of the field radiation. Thus, it is the probability amplitude between the initial state  $|\phi_i\rangle$  at an instant of time  $t_i$ , and the final state  $|\psi_f\rangle$ , viz. in the Schrödinger representation,

$$P \equiv \langle \psi_f | \hat{U}(t_f, t_i) | \psi_i \rangle, \qquad (159)$$

where  $\hat{U}(t_f, t_i)$  is the evolution operator between  $t_i$  and  $t_f$ , which is of physical relevance. The relativistic renormalization program is due to Schwinger, Dyson, Feynman, and Tomonaga, among others. A comprehensive review of the (history) of the relativistic theory is given in Ref. [52].

# 2. Plasmariton quasiparticle

Do relativistic considerations play a role for the plasmariton quasiparticle, born as a superposition of collective modes originating in *nonrelativistic* electron dynamics (Schrödinger equation)? At first glance, one would say "no." If that answer was correct, the "tied T-photon" concept would make perfect sense. But the general answer is "yes" even in a solid-state plasma. In our forthcoming article on surface plasmaritons, the selvedge region (surface profile region) plays a particularly important role, and at least in this region it is possible to accelerate the electrons to relativistic velocities in intense laser fields [53,54]. It is possible to compress the profile region as a hole, and generate also plasmariton modes. Among other effects, higher-harmonic generation appears. The issue as such belongs to nonlinear, nonlocal electron dynamics. In the wake of our forthcoming paper on surface plasmaritons, we will present a relativistic study of the plasmariton quasiparticle and the (inadequacy) of the "field tied to a quasiparticle" concept in the relativistic domain.

# APPENDIX A: TRANSVERSE DIELECTRIC FUNCTION

In this Appendix, a summary of some of the key formulas for the linear, microscopic, and transverse dielectric response theory is presented, with an eye to the particular manner in which the formalism is used in this paper (see Secs. II and VII).

## 1. Lindhard dielectric function

A quantum-mechanical calculation of the field-induced microscopic current density (**J**) can conveniently be obtained from a density matrix operator ( $\hat{\rho}$ ) calculation. Thus,

$$\mathbf{J} = \mathrm{Tr}\{\hat{\boldsymbol{\rho}}\mathbf{j}\},\tag{A1}$$

where  $\hat{\mathbf{j}}$  is the relevant electron current density operator. A many-body expression for the transverse microscopic conductivity function can be obtained by a linearization of Eq. (A1) [31]. In a homogeneous jellium, one obtains in the ( $\mathbf{q}, \omega$ )-domain

$$\mathbf{J}_T(\mathbf{q},\omega) = \sigma_T(q,\omega)\mathbf{E}_T(\mathbf{q},\omega). \tag{A2}$$

In the framework of the single-particle Lindhard (RPA) theory, the transverse dielectric function

$$\varepsilon_T(q,\omega) = 1 + \frac{i}{\varepsilon_0 \omega} \sigma_T(q,\omega)$$
 (A3)

is explicitly given by [20]

$$\varepsilon_T(q,\omega) = 1 - \frac{3}{8} \left(\frac{\omega_p}{\omega}\right)^2 f(u,z),$$
 (A4)

$$f(u,z) = z^{2} + 3u^{2} + 1 - \frac{1}{4z} \left\{ [1 - (z-u)^{2}]^{2} \ln \left| \frac{z-u+1}{z-u-1} \right| + [1 - (z+u)^{2}]^{2} \ln \left| \frac{z+u+1}{z+u-1} \right| \right\},$$
 (A5)

with  $u = \omega/(qv_F)$  and  $z = q/(2k_F)$ .

# 2. Electrostatic Lindhard dielectric function

If electromagnetic retardation effects can be neglected, one has u = 0, so that

$$f(u=0,z) \equiv f(z) = z^2 + 1 - \frac{[1-z^2]^2}{2z} \ln \left| \frac{z+1}{z-1} \right|.$$
 (A6)

The electrostatic approximation tends to give the dielectric function correctly when the quantum parameter tends towards infinity. The electrostatic dispersion relation  $\varepsilon_T(q, \omega) = 0$  reads

$$\omega = \sqrt{\frac{3}{8}} \omega_p f^{1/2}(z). \tag{A7}$$

For  $z \to \infty$ , one has asymptotically

$$f(z) = \frac{8}{3} \left[ 1 - \frac{1}{5z^2} + O(z^{-4}) \right],$$
 (A8)

resulting in  $\omega = \omega_p$  in the limit.

# 3. Classical Lindhard region: Boltzmann equation theory

Quantum interference effects are absent from the Lindhard dielectric function in the limit  $z \rightarrow 0$ . In this classical regime,

$$\varepsilon_T(q,\omega) = 1 - \frac{3}{8} \left(\frac{\omega_p}{\omega}\right)^2 f(u, z \to 0),$$
 (A9)

and hence

$$\varepsilon_T(q,\omega,z\to 0) \equiv 1 - \frac{3}{2} \left(\frac{\omega_p}{\omega}\right)^2 \left\{ u^2 - \frac{u(u^2-1)}{2} \ln\left|\frac{1-u}{1+u}\right| \right\}.$$
 (A10)

The classical dispersion relation, given in Eq. (16), can implicitly be expressed in terms of u as follows:

$$\left(\frac{c}{v_F}\right)^2 \frac{1}{u^2} = 1 - \frac{3}{8} \left(\frac{\omega_p}{\omega}\right)^2 f(u), \qquad (A11)$$

with the two important ratios  $c/v_F$  and  $\omega_p/\omega$ . Equation (A11) and its small *u* limit  $[f(u) \approx 1 + u^2]$  are discussed in Sec. VII A.

In the classical regime, the density matrix theory coincides with an approach based on the Boltzmann equation, which in the collisionless limit has the Vlasov form

$$\frac{\partial}{\partial t} f(\mathbf{r}, \mathbf{v}, t) + \mathbf{v} \cdot \nabla_{\mathbf{r}} f(\mathbf{r}, \mathbf{v}, t) + \frac{e}{m} (\mathbf{E} + \mathbf{v} \times \mathbf{B}) \cdot \nabla_{\mathbf{v}} f(\mathbf{r}, \mathbf{v}, t) = \mathbf{0}, \qquad (A12)$$

where the distribution function  $f(\mathbf{r}, \mathbf{v}, t)$  depends on the particle position ( $\mathbf{r}$ ) and velocity ( $\mathbf{v}$ ), and the time. Linearization ( $f = f_0 + f_1 + \cdots$ ) results in the following expression for the transverse dielectric function in the ( $\omega$ ,  $\mathbf{q}$ )-domain [20]:

$$\varepsilon_T(q,\omega) = 1 - \frac{3}{2} \left(\frac{\omega_p}{\omega}\right)^2 f(w),$$
 (A13)

where

$$f(w) = \frac{1}{w^2} \left[ \frac{1+w^2}{w} \arctan w - 1 \right],$$
 (A14)

with w = i/u. With the help of the relation

$$\arctan w = \frac{1}{2i} \ln \left( \frac{1+iw}{1-iw} \right),$$
 (A15)

one may show that  $\varepsilon_T(q, \omega)$  [Eqs. (A13) and (A14)] is identical to  $\varepsilon_T(q, \omega)$  [Eq. (A10)].

# 4. Dispersion relation at small wave numbers: Brewster branch

To obtain the classical dielectric function at long wavelengths, one starts from the Boltzmann transport equation and makes a series expansion of f(w) from w = 0. The limit  $w \to 0$  corresponds to  $q \to 0$ , and thus  $u = \omega/(qv_F) \to \infty$ , for fixed w. With the help of Eq. (A15), and a large u expansion of the ln function, one obtains  $(u^2 > 1)$ 

$$\arctan w = \frac{1}{2i} \ln \left( \frac{u-1}{u+1} \right) = -2 \sum_{k=1}^{\infty} \frac{1}{(2k-1)u^{2k-1}}.$$
(A16)

To second order in  $q^2$ , we then get from Eq. (A14) the explicit result

$$f(w) = \frac{2}{3} + \frac{2}{15} \left(\frac{qv_F}{\omega}\right)^2 + O(q^4).$$
(A17)

By inserting this result into Eq. (A13), the transverse dielectric function becomes

$$\varepsilon_T(q,\omega) = 1 - \left(\frac{\omega_p}{\omega}\right)^2 \left[1 + \frac{1}{5} \left(\frac{qv_F}{\omega}\right)^2\right] + O(q^4).$$
(A18)

The related squared dispersion relation [Eq. (16)] is therefore given by

$$(cq)^{2} = \frac{\omega^{2} - \omega_{p}^{2}}{1 + \frac{1}{5} \left(\frac{\omega_{p}}{\omega}\right)^{2} \left(\frac{v_{F}}{c}\right)^{2}}.$$
 (A19)

Solutions with real q-values obviously only exist for  $\omega > \omega_p$ , and since  $(v_F/c)^2 \sim 10^{-4}$ , one obtains to an extremely good approximation

$$(cq)^2 = \omega^2 - \omega_p^2. \tag{A20}$$

Hence, we have regained the Brewster branch [Eq. (22)] also in the presence of (weak) spatial dispersion. In a paper on the microscopic long-wavelength properties of plasmons (*L*-mode), Harris [25] started from the RPA theory, used the Wigner phase space distribution for scalar potentials [55] to reach the linearized Boltzmann equation involving the gradient of the scalar potential, and ended up with the microscopic form of the plasmon dispersion relation. In this work, the linearized Boltzmann equation related to the Lorentz field  $\mathbf{E} + \mathbf{v} \times \mathbf{B}$  [cf. Eq. (A12)] was obtained directly from the transverse classical Lindhard dielectric function, giving finally the *T*-mode dispersion relation (Brewster branch).

# 5. Note on branch cuts: Surf-riding resonance

The single-particle excitation spectrum is associated with the branch-cut structure of the Lindhard dielectric function, as is well known. These cuts give rise to the presence of *nonexponentially* decaying terms in the linear and nonlinear nonlocal electrodynamics. In a nonlinear context, the branch-cut contributions seem first to have been studied in connection with acousto-optic (~Brillouin) scattering [56].

Let us return to the transverse hydrodynamic dispersion relation, and assume, unrealistically for jellium (but possible in a charged gas plasma, for instance [57]), that

$$\omega = D_T^{1/2} q = \sqrt{\frac{1}{5}} v_F q. \tag{A21}$$

In the field of plasma physics this resonance is called a surf-riding resonance, because the (plasmariton) wave travels there [with the replacement  $(1/5)^{1/2}v_F \Rightarrow v_F$ ] in a stationary fashion synchronously with the Fermi velocity. In the Boltzmann approach, the corner of the branch cut which runs out to infinity is located at the surf-riding resonance [56].

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# APPENDIX B: DETERMINATION OF POLARIZATION-FIELD AMPLITUDE

Let us consider a simple harmonic one-dimensional oscillator with a scaled energy operator

$$\hat{H} = \frac{1}{Q^2} \left[ \frac{\hat{p}^2}{2M} + \frac{1}{2} M \tilde{\omega}_p^2 \hat{x}^2 \right].$$
 (B1)

The scaling factor  $Q^{-2}$  is introduced to make direct contact with Eq. (124). The scaling factor may be included in the momentum ( $\hat{p}$ ) and coordinate ( $\hat{x}$ ) operators, replacing these by new ones,  $\hat{p}/Q$  and  $\hat{x}/Q$ . The transformations

$$\hat{x} = Q \left(\frac{\hbar}{2M\tilde{\omega}_p}\right)^{1/2} (\hat{b}^{\dagger} + \hat{b}), \tag{B2}$$

$$\hat{p} = iQ \left(\frac{M\hbar\tilde{\omega}_p}{2}\right)^{1/2} (\hat{b}^{\dagger} - \hat{b}), \tag{B3}$$

where the operators  $\hat{b}$  and  $\hat{b}^{\dagger}$  satisfy the boson commutator relation

$$[\hat{b}, \hat{b}^{\dagger}] = 1.$$
 (B4)

By means of the transformation in Eqs. (B2) and (B3), one obtains

$$\hat{H} = \hbar \tilde{\omega}_p \left( \hat{b}^\dagger \hat{b} + \frac{1}{2} \right), \tag{B5}$$

a result that when generalized, i.e.,  $\hat{b}(\hat{b}^{\dagger}) \Rightarrow \hat{b}_{qs}(\hat{b}^{\dagger}_{qs})$ , leads to Eq. (137). From the classical connection between oscillator energy and amplitude, namely

$$\hbar\tilde{\omega}_p = \frac{M}{2Q^2}\tilde{\omega}_p^2 P_0^2, \tag{B6}$$

one obtains, with the help of Eq. (112), the result cited in Eq. (138).

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