Dipolar quantum electrodynamics theory of the three-dimensional electron gas

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We provide a description of the interaction between light and a three-dimensional electron gas in the framework of the Power-Zienau-Wooly (PZW) formulation of the quantum electrodynamics. The existence of the collective plasmon and plasmon-polariton modes of the gas appears as a consequence of the general properties of the PZW Hamiltonian. Our description enables a fully microscopic theory of the surface plasmon-polariton excitations, based on a quantum Hamiltonian that is free from any additional hypothesis regarding the material dispersion. This theory can therefore be applied to study the quantum optical properties of plasmon and surface plasmon waves.

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I. INTRODUCTION

The quantum description of the interaction between the electromagnetic field with a solid state system can be devised into two major approaches. The first approach, inherent to quantum optics, is very general and can be traced back to the seminal Hopfield paper [1]. In this framework, the matter degrees of freedom are treated as elementary bosonic excitations, allowing to define a harmonic polarization field. The coupling between the polarization and electromagnetic fields yields mixed light-matter excitations, polaritons, which fully describe the propagation of the radiation inside the material medium. Using this approach, Hopfield recovered the resonant dielectric function which corresponds to excitons in crystals and the corresponding dispersion relation of electromagnetic waves [1]. The polariton Hopfield model became the basis for subsequent quantization schemes of the medium-assisted electromagnetic field in absorbing and dispersive dielectrics [2–4]. In particular, a recent generalization of the polariton model by Suttorp and Wubs [5] justified the very general modern schemes of phenomenological quantization, based on the Green function formalism [6,7].

The second approach is based on the many-body solid-state physics, where one rather starts with the particular system in consideration, and computes explicitly the interaction between the particles and the electromagnetic field. Accent is put on the complexity of the many-body problem where interactions among particles are fundamentally mediated by Coulomb forces [8]. In that case, the coupling with the radiation field is most often treated as a perturbation, in the linear response formalism [9], which provides the dielectric response of the ensemble of interacting particles. One of the most studied such systems is the electron gas [9,10], which has a dielectric response described by the Linhard formula [11,12]. The long-wavelength limit of the Linhard dielectric tensor is on the basis of the description of many electromagnetic problems involving the electron gas, including the surface plasmon polaritons that are at the heart of modern research in nanophotonics [13–15].

Recent years have seen the emergence of experimental systems where both solid state and quantum optics concepts become pertinent for the adequate description of the underlying physics. One such system is the quantum-confined electron gas in ultrastrong interaction with a microcavity in the mid- and far-infrared frequency domains [16–18]. In this system, the interaction with photons is mediated through the collective plasmonic excitations of the gas [19]. As these excitations are able to gather the oscillator strength of many electronic transitions, the overall light-matter interaction strength increases with the density of the electron gas. Eventually, at very high densities, the system is pushed into the ultrastrong light-matter coupling regime [18], where new quantum-optical phenomena are predicted [20]. Another example is the magneto-plasmon excitations of a 2D electron gas, coupled to split-ring resonators [21]. In all the aforementioned systems, the light-matter coupling strength becomes comparable with the energy of the collective excitations. At that level, quantum optics and many-body physics became intricate, and it is therefore interesting to provide a quantum formalism able to capture both aspects.

Recently, we developed such formalism for the case of the intersubband excitations of the 2D electron gas interacting with a microcavity, in the long-wavelength limit [22]. This formalism is based on the description of the electromagnetic field and its coupling with matter in the Power-Zineau-Wooley (PZW) representation [23,24]. In this framework, the radiation is described by a purely transverse displacement field $\mathbf{D}(\mathbf{r})$, while the electron gas enters through a polarization field density $\mathbf{P}(\mathbf{r})$. The full electromagnetic Hamiltonian contains not only the interaction between the particles and the radiated field through a linear coupling term $-\mathbf{D}(\mathbf{r})\cdot\mathbf{P}(\mathbf{r})$, but also the particle-particle interactions in a quadratic term $\mathbf{P}^2(\mathbf{r})$. The partial diagonalization of the matter degrees of freedom involving the $\mathbf{P}^2$ term allows to express the polarization density $\mathbf{P}$ in terms of quantum bosonic operators that describe the collective excitations of the gas, displaying clearly the link between the electromagnetic Hamiltonian with the many-body aspects of the system.

The PZW or the multipolar Hamiltonian is widely used in atomic-like systems [6,7,25]. It that case it allows to provide a simplified version of the electromagnetic interaction relying on the fact that the spatial extension of the atoms and molecules is typically very small as compared to the wavelength of radiation. However, the $\mathbf{P}^2$ term and the corresponding local fields effects are usually neglected [7,26,27]. In this paper, we stress the connection of this term with the many-body collective states of the system, as previously underlined with intersubband excitations [22]. This connection is formulated

in a general manner for a homogeneous (but not necessarily isotropic) system of interacting bosonic (dipolar) excitations. We show that we can define a general transverse dielectric tensor of the system, which accounts for the local field effects that induce the collective excitations, and also describes the propagation of electromagnetic waves in the material. This dielectric tensor can therefore be regarded as the electromagnetic generalization of the result of Linhard [11] formulated for the longitudinal Coulomb interaction, and widely used in solid state physics [9].

This approach is then applied to the case of the 3D electron gas. We derive the polarization field $\mathbf{P}(\mathbf{r})$ of the gas solely from the kinetic part of the electronic Hamiltonian, which turns out to be equivalent to the bosonisation of the 3D gas in the random-phase approximation (RPA) [12,28]. In the case where the gas interacts with a continuum of homogeneous waves, our approach is confirmed by recovering the Linhard formula of the dielectric function.

Most importantly, this frameworks allows us to formulate a fully quantum theory of the surface plasmon polaritons (SPPs) [15], providing thus a new tool for the quantum description of near-field optical phenomena. Indeed, so far, the SPP have been quantized in a phenomenological way, starting from the classical solutions of the Maxwell's equations on an interface with an electron gas [29–31]. Our formalism allows for a "bottom-up" approach [32], where the SSP is obtained by a self-consistent diagonalization of the first-principles microscopic Hamiltonian, which takes into account the geometrical constraints of the problem. This theory is free from the necessity to introduce a phenomenological electromagnetic energy density in the presence of material resonances [31]. Furthermore, the surface-plasmon polaritons are described by bosonic quantum operators, that are a linear superposition of matter and light fields. The corresponding Hopfield coefficients are explicitly provided, which allows to approach the SPPs excitations from the viewpoint of quantum optics [20,33]. Our theory thus sets convenient and rigorous ground for the description of quantum processes with SPPs, such as the spontaneous emission of SPPs as illustrated in Sec. III C.

This paper is organized as follows. In Sec. II, we study a general Hopfield-like model in the PZW representation, which describes a collection of elementary bosonic excitations coupled with light and interacting with each other through dipole-dipole forces. General results on the dielectric tensor of the system, and its connection with the collective modes are discussed in Sec. II B. In Sec. III, we derive the PZW Hamiltonian of the 3D elecron gas and we show how the corresponding plasmon modes can be deduced from it. The surface plasmon mode is discussed in Sec. III C. In Sec. III D, we discuss the connection between the PZW Hamiltonian and the minimal coupling Hamiltonian which is commonly used in the literature. Technical information is gathered in the appendixes.

II. PZW DESCRIPTION OF THE HOPFIELD DIELECTRIC

A. PZW Hamiltonian

We first consider the case of a bosonic polarization field coupled with the electromagnetic field, which corresponds to the original Hopfield model [1]. Our aim is to provide the formulation of this problem in the framework of the PZW representation. The material medium is considered as translational invariant but not necessary isotropic. In the PZW representation, the electromagnetic field is described in terms of the transverse field operators $\hat{\mathbf{D}}(\mathbf{r})$ (displacement field), $\hat{\mathbf{H}}(\mathbf{r})$ (magnetic field), and $\hat{\mathbf{A}}(\mathbf{r})$ (vector potential) expanded on a plane-wave basis with the help of creation $a^{\dagger}_{\mathbf{Q},\boldsymbol{\epsilon}_\mathbf{Q}}$ and annihilation $a_{\mathbf{Q},\boldsymbol{\epsilon}_\mathbf{Q}}$ operators [25]:

$$\hat{\mathbf{D}}(\mathbf{r}) = \sum_{\mathbf{Q},\boldsymbol{\epsilon}_\mathbf{Q}} i D_\mathbf{Q} \boldsymbol{\epsilon}_\mathbf{Q} e^{i\mathbf{Q}\mathbf{r}} \left(a_{\mathbf{Q},\boldsymbol{\epsilon}_\mathbf{Q}} - a^{\dagger}_{-\mathbf{Q},\boldsymbol{\epsilon}_\mathbf{Q}}\right), \tag{1}$$

$$\hat{\mathbf{H}}(\mathbf{r}) = \sum_{\mathbf{Q},\boldsymbol{\epsilon}_\mathbf{Q}} i \frac{D_\mathbf{Q}}{c\varepsilon_0} (\mathbf{Q} \wedge \boldsymbol{\epsilon}_\mathbf{Q}) e^{i\mathbf{Q}\mathbf{r}} \left(a_{\mathbf{Q},\boldsymbol{\epsilon}_\mathbf{Q}} + a^{\dagger}_{-\mathbf{Q},\boldsymbol{\epsilon}_\mathbf{Q}}\right), \tag{2}$$

$$\hat{\mathbf{A}}(\mathbf{r}) = \sum_{\mathbf{Q},\boldsymbol{\epsilon}_\mathbf{Q}} A_\mathbf{Q} \boldsymbol{\epsilon}_\mathbf{Q} e^{i\mathbf{Q}\mathbf{r}} \left(a_{\mathbf{Q},\boldsymbol{\epsilon}_\mathbf{Q}} + a^{\dagger}_{-\mathbf{Q},\boldsymbol{\epsilon}_\mathbf{Q}}\right), \tag{3}$$

$$D_\mathbf{Q} = \sqrt{\frac{\varepsilon_0 \hbar \omega_{c\mathbf{Q}}}{2V}}, \quad A_\mathbf{Q} = \frac{D_\mathbf{Q}}{\varepsilon_0 \omega_{c\mathbf{Q}}}, \tag{4}$$

$$\left[a_{\mathbf{Q},\boldsymbol{\epsilon}_\mathbf{Q}}, a^{\dagger}_{\mathbf{Q}',\boldsymbol{\epsilon}_{\mathbf{Q}'}}\right] = \delta_{\mathbf{Q},\mathbf{Q}'} \delta_{\boldsymbol{\epsilon}_\mathbf{Q},\boldsymbol{\epsilon}_{\mathbf{Q}'}}. \tag{5}$$

Here, c is the speed of light, ε_0 is the electric constant, V is the quantization volume, $\omega_{c\mathbf{Q}}$ is the plane-wave frequency, and $\boldsymbol{\epsilon}_\mathbf{Q} = \boldsymbol{\epsilon}_{1\mathbf{Q}}, \boldsymbol{\epsilon}_{2\mathbf{Q}}$ are the two orthogonal polarization vectors of each homogeneous wave with a wave vector $\mathbf{Q}$:

$$\omega_{c\mathbf{Q}} = |\mathbf{Q}|c, \qquad \mathbf{Q} \cdot \boldsymbol{\epsilon}_{1,2\mathbf{Q}} = 0, \tag{6}$$

and the free photon Hamiltonian $\mathcal{H}_{\text{ph}}$ is

$$\mathcal{H}_{\text{ph}} = \sum_{\mathbf{Q},\boldsymbol{\epsilon}_\mathbf{Q}} \hbar \omega_{c\mathbf{Q}} \left(a^{\dagger}_{\mathbf{Q},\boldsymbol{\epsilon}_\mathbf{Q}} a_{\mathbf{Q},\boldsymbol{\epsilon}_\mathbf{Q}} + 1/2\right). \tag{7}$$

The second ingredient of the PZW representation is the polarization field operator $\hat{\mathbf{P}}(\mathbf{r})$ that describes the matter degrees of freedom. Generally, $\hat{\mathbf{P}}(\mathbf{r})$ is expressed as a function of the position of each particle [23,24,34]. This formulation is convenient for spatially localized systems, as it allows for a multipolar expansion of the electromagnetic interaction. In our case, we rather seek to express the polarization field as a function of bosonic excitation operators. Such expression would be closer to the solid state physics formalisms, where the material excitations are associated with nonlocal creation and annihilation operators acting on the particle states, rather than the microscopic positions of each particle.

We therefore suppose that the material medium is described by a bosonic Hamiltonian $\mathcal{H}_{\text{mat}}$ and a polarization field operator $\hat{\mathbf{P}}(\mathbf{r})$ that can also be expanded in the plane-wave basis:

$$\mathcal{H}_{\text{mat}} = \sum_{\alpha,\mathbf{Q}} \hbar \omega_{\alpha\mathbf{Q}} b^{\dagger}_{\mathbf{Q}\alpha} b_{\mathbf{Q}\alpha}, \tag{8}$$

$$\hat{\mathbf{P}}(\mathbf{r}) = \sum_{\alpha,\mathbf{Q}} \frac{\mathbf{d}_{\mathbf{Q}\alpha}}{V} e^{-i\mathbf{Q}\mathbf{r}} \left(b_{\mathbf{Q}\alpha} + b^{\dagger}_{-\mathbf{Q}\alpha}\right), \tag{9}$$

$$[b_{\mathbf{Q}\alpha}, b^{\dagger}_{\mathbf{Q}'\beta}] = \delta_{\mathbf{Q},\mathbf{Q}'} \delta_{\alpha,\beta}. \tag{10}$$

In the above expression, $\omega_{\alpha\mathbf{Q}}$ is a resonance of the material medium, which can be generally wave-vector-dependent (dispersive), and the symbol α labels the internal quantum transitions of the medium. The vectors $\mathbf{d}_{\mathbf{Q}\alpha}$ have therefore the meaning of microscopic dipole moments associated to the transition α. They satisfy the condition

$$\mathbf{d}_{\mathbf{Q}\alpha} = \mathbf{d}^*_{-\mathbf{Q}\alpha}, \tag{11}$$

which stems from the hermicity of the polarization field operator (9). Another important property is the commutation rule:

$$[\hat{P}_i(\mathbf{r}), \hat{P}_j(\mathbf{r}')] = 0, \tag{12}$$

which is an immediate corollary from Eq. (10). Here the Latin indexes denote vector components in a Cartesian basis. In that picture, the polarization field $\hat{\mathbf{P}}(\mathbf{r})$ is considered as an independent dynamical variable of the system, that depends only on the intrinsic characteristics of the material medium.

In the general case considered here, the expression of the polarization field from Eq. (9) has a meaning of a postulate. This expression will be justified in the end of this section through comparison between the PZW Hamiltonian and the minimal coupling Hamiltonian. Furthermore, expressions of the form of Eq. (9) can actually be derived for a particular solid state system. This will be illustrated for the case of the 3D electron gas in Sec. III.

The PZW Hamiltonian that describes the coupling between the medium and the electromagnetic field is then

$$\mathcal{H} = \mathcal{H}_{\rm ph} + \mathcal{H}_{\rm mat} - \frac{1}{\varepsilon_0}\int \hat{\mathbf{D}}\hat{\mathbf{P}} d^3\mathbf{r} + \frac{1}{2\varepsilon_0}\int \hat{\mathbf{P}}^2 d^3\mathbf{r}. \tag{13}$$

In the above Hamiltonian, the coupling with the magnetic field is absent. This is an approximation that will be discussed further. The light-matter interaction is contained in the third term on the right-hand side (r.h.s.) of Eq. (13), which expresses a linear coupling between the medium polarization $\hat{\mathbf{P}}$ and the displacement field $\hat{\mathbf{D}}$. Since the displacement field is transverse by construction, this term describes a purely retarded interaction. The fourth term in the r.h.s., which is square in the polarization operator $\hat{\mathbf{P}}$, describes the interaction between particles, that leads to collective matter excitations, as we will show in the next section.

The linear $\mathcal{H}_{\rm int}$ and quadratic $\mathcal{H}_{\rm P2}$ interaction parts are written explicitly:

$$\mathcal{H}_{\rm int} = -\frac{1}{\varepsilon_0}\int \hat{\mathbf{D}}\hat{\mathbf{P}} d^3\mathbf{r} = -i\sum_{\mathbf{Q},\boldsymbol{\epsilon}_\mathbf{Q},\alpha} \frac{D_\mathbf{Q}(\boldsymbol{\epsilon}_\mathbf{Q}\mathbf{d}_{\mathbf{Q}\alpha})}{\varepsilon_0} \times \left(a_{\mathbf{Q},\boldsymbol{\epsilon}_\mathbf{Q}} - a^\dagger_{-\mathbf{Q},\boldsymbol{\epsilon}_\mathbf{Q}}\right)(b_{-\mathbf{Q}\alpha} + b^\dagger_{\mathbf{Q}\alpha}), \tag{14}$$

$$\mathcal{H}_{\rm P2} = \frac{1}{2\varepsilon_0}\int \hat{\mathbf{P}}^2 d^3\mathbf{r} = \sum_{\mathbf{Q},\alpha,\beta} \frac{\mathbf{d}_{\mathbf{Q}\alpha}\mathbf{d}^*_{\mathbf{Q}\beta}}{2\varepsilon_0 V}(b_{\mathbf{Q}\alpha} + b^\dagger_{-\mathbf{Q}\alpha})(b_{-\mathbf{Q}\beta} + b^\dagger_{\mathbf{Q}\beta}). \tag{15}$$

For the following discussion, it will be convenient to split the polarization field into longitudinal and transverse parts. For this, we use the orthonormal basis $(\boldsymbol{\epsilon}_{1\mathbf{Q}}, \boldsymbol{\epsilon}_{2\mathbf{Q}}, \mathbf{Q}/Q)$ with $Q = |\mathbf{Q}|$:

$$\hat{\mathbf{P}} = \hat{\mathbf{P}}_t + \hat{\mathbf{P}}_l, \tag{16}$$

$$\hat{\mathbf{P}}_t = \sum_{\alpha,\mathbf{Q},\boldsymbol{\epsilon}_\mathbf{Q}} \frac{(\mathbf{d}_{\mathbf{Q}\alpha}\boldsymbol{\epsilon}_\mathbf{Q})\boldsymbol{\epsilon}_\mathbf{Q}}{V} e^{-i\mathbf{Q}\mathbf{r}}(b_{\mathbf{Q}\alpha} + b^\dagger_{-\mathbf{Q}\alpha}), \tag{17}$$

$$\hat{\mathbf{P}}_l = \sum_{\alpha,\mathbf{Q}} \frac{(\mathbf{d}_{\mathbf{Q}\alpha}\mathbf{Q})\mathbf{Q}}{V Q^2} e^{-i\mathbf{Q}\mathbf{r}}(b_{\mathbf{Q}\alpha} + b^\dagger_{-\mathbf{Q}\alpha}). \tag{18}$$

Respectively, the square polarization Hamiltonian is split into two distinct parts $\mathcal{H}_{\rm P2} = \mathcal{H}_{t\rm P2} + \mathcal{H}_{l\rm P2}$. The longitudinal part expresses the Coulomb interactions between particles. Indeed, introducing the charge density operator $\hat{\rho}$:

$$\hat{\rho} = \nabla\hat{\mathbf{P}} = \nabla\hat{\mathbf{P}}_l = -i\sum_{\mathbf{Q},\alpha,\beta} \frac{(\mathbf{Q}\mathbf{d}_{\mathbf{Q}\alpha})}{V}(b_{\mathbf{Q}\alpha} + b^\dagger_{-\mathbf{Q}\alpha})e^{-i\mathbf{Q}\mathbf{r}}, \tag{19}$$

we can recast the longitudinal part of the self-polarization $\mathcal{H}_{l\rm P2}$ in the form of a Coulomb potential:

$$\mathcal{H}_{\rm lP2} = \frac{1}{4\pi\varepsilon_0}\int \frac{\hat{\rho}(\mathbf{r})\hat{\rho}(\mathbf{r}')}{|\mathbf{r}-\mathbf{r}'|} d^3\mathbf{r} d^3\mathbf{r}'. \tag{20}$$

This expression is obtained by resuming over the plane-wave components of the Fourier expansion in Eq. (18) [23]. Furthermore, we can define a current density operator $\hat{\mathbf{j}}$ that is obtained as the time derivative of the polarization field $\hat{\mathbf{P}}$ through the formula:

$$\hat{\mathbf{j}} = \frac{\mathrm{d}\hat{\mathbf{P}}}{\mathrm{d}t} = \frac{i}{\hbar}[\mathcal{H}, \hat{\mathbf{P}}] = \frac{i}{\hbar}[\mathcal{H}_{\rm mat}, \hat{\mathbf{P}}]. \tag{21}$$

Indeed, thanks to the commutation rule (12) for the polarization operator only the matter part $\mathcal{H}_{\rm mat}$ in Eq. (13) contributes to the evolution of the polarization field. Taking the explicit expression of the current density:

$$\hat{\mathbf{j}}(\mathbf{r}) = i\sum_{\mathbf{Q},\alpha} \frac{\omega_{\alpha\mathbf{Q}}\mathbf{d}_{\mathbf{Q}\alpha}}{V}(b_{\mathbf{Q}\alpha} - b^\dagger_{-\mathbf{Q}\alpha})e^{-i\mathbf{Q}\mathbf{r}}, \tag{22}$$

it is easy to verify that the operators $\hat{\mathbf{j}}$ and $\hat{\rho}$ satisfy the continuity relation:

$$\frac{\mathrm{d}\hat{\rho}}{\mathrm{d}t} + \nabla\hat{\mathbf{j}} = 0. \tag{23}$$

Here, like in Eq. (22), the time derivative symbol "$\mathrm{d}/\mathrm{d}t$" has a meaning of the commutator with the total Hamiltonian of the system. This relation ensures the self-consistency of the bosonic model and the polarization field operator defined through Eq. (9).

Let us now examine this model in the minimal-coupling representation, which was used for the original Hopfield model [1]. The image of the Hamiltonian (13) in this representation is obtained by applying the unitary transformation [23]:

$$T = \exp\left(-\frac{i}{\hbar}\int \hat{\mathbf{A}}\hat{\mathbf{P}} d^3\mathbf{r}\right). \tag{24}$$

Since the vector potential in Eq. (3) is transverse (Coulomb gauge), the above transformation affects only the transverse

fields. Its action can be computed explicitly with the use of the Baker-Hausdor expansion:

$$e^{i\hat{B}}\mathcal{H}e^{-i\hat{B}} = \mathcal{H} + i[\hat{B},\mathcal{H}] + \frac{i^2}{2!}[\hat{B},[\hat{B},\mathcal{H}]] + \cdots . \quad (25)$$

Thanks to the commutation rule (12) the quadratic part of the Hamiltonian (13) is unaffected by the transformation

$$T^{+}\mathcal{H}_{\mathrm{P2}}T = \mathcal{H}_{\mathrm{P2}}. \quad (26)$$

Using the bosonic commutation rules for the photonic [Eq. (5)] and polarization operators [Eq. (10)], as well as Eq. (25), we obtain

$$T^{+}\mathcal{H}_{\mathrm{ph}}T = \mathcal{H}_{\mathrm{ph}} - \mathcal{H}_{\mathrm{int}} + \mathcal{H}_{\mathrm{tP2}}, \quad (27)$$

$$T^{+}\mathcal{H}_{\mathrm{int}}T = \mathcal{H}_{\mathrm{int}} - 2\mathcal{H}_{\mathrm{tP2}}. \quad (28)$$

In Eq. (27), the second term corresponds to the first-order commutator of the Baker-Hausdor expansion from Eq. (25), and the third term to the second-order commutator. All higher-order terms vanish because of the bosonic commutation rules [Eqs. (5) and (10)], which reduce successively the commutators into c numbers. This is a general feature of the bosonic model, which renders the unitary transformation (24) exact at the second order of the expansion (25).

Combining Eqs. (26), (27), and (28), we obtain

$$T^{+}(\mathcal{H}_{\mathrm{ph}} + \mathcal{H}_{\mathrm{int}} + \mathcal{H}_{\mathrm{P2}})T = \mathcal{H}_{\mathrm{ph}} + \mathcal{H}_{\mathrm{P2}} - \mathcal{H}_{\mathrm{tP2}}. \quad (29)$$

The combined transformation of the photon, interaction, and square polarization Hamiltonian therefore provides the longitudinal part $\mathcal{H}_{\mathrm{lP2}}$ defined in Eq. (20). We see that the effect of the inverse PZW transformation defined in Eq. (24) is to subtract the transverse part from the total polarization self-energy.

Applying the transformation (24) to the remaining matter part $\mathcal{H}_{\mathrm{mat}}$ of the total Hamiltonian, we obtain

$$T^{+}\mathcal{H}_{\mathrm{mat}}T = \mathcal{H}_{\mathrm{mat}} + \mathcal{H}'_{\mathrm{int}} + \mathcal{H}_{\mathrm{A2}}. \quad (30)$$

Here, $\mathcal{H}'_{\mathrm{int}}$ is the light-matter interaction Hamiltonian expressed in the minimal-coupling representation:

$$\mathcal{H}'_{\mathrm{int}} = i\sum_{\mathbf{Q},\boldsymbol{\epsilon}_{\mathbf{Q}},\alpha} A_{\mathbf{Q}}\omega_{\alpha\mathbf{Q}}(\mathbf{d}_{\mathbf{Q}\alpha}\boldsymbol{\epsilon}_{\mathbf{Q}}) \times \left(a_{\mathbf{Q},\boldsymbol{\epsilon}_{\mathbf{Q}}} + a^{\dagger}_{-\mathbf{Q},\boldsymbol{\epsilon}_{\mathbf{Q}}}\right)\left(b_{-\mathbf{Q}\alpha} - b^{\dagger}_{\mathbf{Q}\alpha}\right). \quad (31)$$

Using the expression for the current operator (22), we recognize (31) to be

$$\mathcal{H}'_{\mathrm{int}} = \int \hat{\mathbf{j}}\hat{\mathbf{A}}d^3\mathbf{r}. \quad (32)$$

Finally, the remaining term of the Hamiltonian $\mathcal{H}_{\mathrm{A2}}$ is expressed as

$$\mathcal{H}_{\mathrm{A2}} = \sum_{\alpha,\mathbf{Q},\boldsymbol{\epsilon}_{\mathbf{Q}},\boldsymbol{\epsilon}'_{\mathbf{Q}}} \frac{\omega_{\alpha\mathbf{Q}}(\mathbf{d}_{\mathbf{Q}\alpha}\boldsymbol{\epsilon}_{\mathbf{Q}})(\mathbf{d}^{*}_{\mathbf{Q}\alpha}\boldsymbol{\epsilon}'_{\mathbf{Q}})}{\hbar} A^2_{\mathbf{Q}} \times \left(a_{\mathbf{Q},\boldsymbol{\epsilon}_{\mathbf{Q}}} + a^{\dagger}_{-\mathbf{Q},\boldsymbol{\epsilon}_{\mathbf{Q}}}\right)\left(a_{-\mathbf{Q},\boldsymbol{\epsilon}'_{\mathbf{Q}}} + a^{\dagger}_{\mathbf{Q},\boldsymbol{\epsilon}'_{\mathbf{Q}}}\right). \quad (33)$$

This is the part of the minimal-coupling Hamiltonian that is square in the vector potential $\mathbf{A}$. The image of the Hamiltonian (13) in the minimal-coupling representation is therefore

$$T^{+}\mathcal{H}T = \mathcal{H}_{\mathrm{ph}} + \mathcal{H}_{\mathrm{mat}} + \mathcal{H}'_{\mathrm{int}} + \mathcal{H}_{\mathrm{A2}} + \mathcal{H}_{\mathrm{lP2}}. \quad (34)$$

The first four terms in the above equation can be identified with the Hopfield polariton model [1], while the last term is a Coulomb potential describing dipole-dipole coupling between the matter excitations. The minimal-coupling representation of the Hamiltonian (13) confirms the fact that, provided that the polarization field operator $\hat{\mathbf{P}}$ of a particular solid state system can be expressed as a sum of elementary bosonic excitations, one can derive both the interaction of the system with light, as well as the particle-particle interactions within the system.

Let us now discuss the underlying approximations of this approach. The first obvious approximation is that the magnetic properties of matter are neglected. Indeed, in the general PZW Hamiltonian, along with the polarization operator $\hat{\mathbf{P}}$, the matter is also described with a magnetization operator $\hat{\mathbf{M}}$ [23]. The magnetization operator is linearly coupled to the magnetic field $\hat{\mathbf{H}}$ of the electromagnetic waves propagating inside the material. It also contributes to the total current in Eq. (21) through a term $\nabla \times \hat{\mathbf{M}}$. In this work, we neglect the magnetization operator $\hat{\mathbf{M}}$, thus neglecting the dynamic corrections of the magnetic field on the particle trajectories. Intuitively, this approximation considers that the particles move along spatially bounded straight segments, such as those of localized oscillating dipoles. This situation also covers static effects, such as the case where constant homogeneous magnetic field is applied on the system, and the particle trajectories are Landau-quantized circular orbits. In that case, the effects of the magnetic field can be absorbed in the kinetic part of the matter Hamiltonian, while interaction with light is dominated by an electric dipole type term [35]. Indeed, the circular movement of a charged particle can always be considered as a superposition of two oscillating dipoles in two orthogonal directions, and this picture could eventually be used to construct a polarization filed of the type (9) that couples to $\hat{\mathbf{D}}$. As mentioned above, the exclusion of $\hat{\mathbf{M}}$ from the Hamiltonian is equivalent to neglect the dynamic effects of the time-oscillating magnetic component of the electromagnetic wave interacting with the system, such as the magnetic-moment-allowed quantum transitions.

A second, more fundamental approximation underlying in the expression of the polarization field (9) is the assumption that matter is described by bosonic operators. Indeed, the elementary constituents of the matter, such as electrons and nuclei, are fermionic particles, and therefore the polarization density $\hat{\mathbf{P}}$ is necessary expressed from anticommuting creation and annihilation operators. It is precisely the concepts developed in solid state physics [12] that allow to justify how polarization fields of the form of Eq. (9) can be assigned to a dense ensemble of interacting fermionic particles. This will be illustrated in Sec. III where we show that the RPA applied to the single-particle kinetic energy electronic Hamiltonian of the 3D electron gas allows to construct a bosonized polarization field. The implementation of this polarization field with the PZW Hamiltonian in the form of Eq. (13) then allows to derive the elementary excitations of the gas. Such excitations are either collective, involving all particles of the system at once, or electron-hole single-particle excitations. In the next paragraph, we will show that the emergence of collective excitations

in the system is actually a general property of the PZW Hamiltonian (13).

B. Dielectric tensor and collective modes

1. Definition of the dielectric tensor

The major convenience of the PZW representation is that the matter degrees of freedom contained in the polarization field $\hat{\mathbf{P}}$ appear as dynamical variables that are independent from the radiation degrees of freedom (fields $\hat{\mathbf{D}}$ and $\hat{\mathbf{H}}$). We can therefore regroup all the matter degrees of freedom into a separate Hamiltonian:

$$\mathcal{H}_{\rm mat}+\mathcal{H}_{\rm P2}=\sum_{\alpha,\mathbf{Q}}\hbar\omega_{\alpha\mathbf{Q}}b^{\dagger}_{\mathbf{Q}\alpha}b_{\mathbf{Q}\alpha}+\sum_{\mathbf{Q},\alpha,\beta}\frac{\mathbf{d}_{\mathbf{Q}\alpha}\mathbf{d}^{*}_{\mathbf{Q}\beta}}{2\varepsilon_0 V}(b_{\mathbf{Q}\alpha}+b^{\dagger}_{-\mathbf{Q}\alpha})(b_{-\mathbf{Q}\beta}+b^{\dagger}_{\mathbf{Q}\beta}). \tag{35}$$

This quadratic Hamiltonian can be diagonalized through a Fano-Bogoliubov procedure [36] to yield the new coupled modes of the system. The details of this procedure are provided in Appendix A, and in this paragraph we summarize the main results. In Appendix A, we show that we can associate to the Hamiltonian (35) the following spatial tensor:

$$\bar{\bar{\epsilon}}(\omega,\mathbf{Q})=\bar{\bar{I}}-\sum_{\alpha}\frac{2\omega_{\alpha\mathbf{Q}}(\mathbf{d}^{*}_{\mathbf{Q}\alpha}\otimes\mathbf{d}_{\mathbf{Q}\alpha})}{\varepsilon_0 V\hbar\left(\omega^2-\omega^2_{\alpha\mathbf{Q}}\right)}. \tag{36}$$

Here, the symbol "$\otimes$" denotes a tensor product, and $\bar{\bar{I}}$ is the spatial unit tensor. We will show further that (36) can be interpreted as the dielectric tensor of the medium. At the current stage, the utility of this tensor is that the zeros of its determinant $\| \bar{\bar{\epsilon}}(\omega,\mathbf{Q}) \|$ provide the eigenvalues $\Omega_{i\mathbf{Q}}$ of the matter Hamiltonian (35):

$$\| \bar{\bar{\epsilon}}(\Omega_{i\mathbf{Q}},\mathbf{Q}) \|=0. \tag{37}$$

These eigenvalues correspond to the new coupled modes of the system induced by the particle-particle interactions described by the quadratic part $\mathcal{H}_{\rm P2}$ of the matter Hamiltonian. Equation (37) provides implicitly their dispersion relation $\Omega_{i\mathbf{Q}}=\Omega_i(\mathbf{Q})$. The index i now labels the coupled modes with a total number equal to that of the initial excitations labeled by the index α. The definition (36) implies that the tensor $\bar{\bar{\epsilon}}(\omega,\mathbf{Q})$ is Hermitian, and therefore all the eigenvalues $\Omega_{i\mathbf{Q}}$ are real. Furthermore, the hermiticity implies that we can find an orthogonal basis $\mathbf{n}=(\mathbf{n}_1,\mathbf{n}_2,\mathbf{n}_3)$ in space where the tensor (36) is diagonal. Let us introduce the following characteristic frequencies $W_{\alpha\mathbf{nQ}}$ through the equation

$$W^2_{\alpha\mathbf{nQ}}=\frac{2\omega_{\alpha\mathbf{Q}}|\mathbf{d}_{\mathbf{Q}\alpha}\mathbf{n}|^2}{\varepsilon_0 V\hbar}. \tag{38}$$

Then the diagonal form of (36) is explicitly written as

$$\bar{\bar{\epsilon}}(\omega,\mathbf{Q})=\bar{\bar{I}}-\sum_{\alpha\mathbf{n}}\frac{W^2_{\alpha\mathbf{nQ}}}{\omega^2-\omega^2_{\alpha\mathbf{Q}}}(\mathbf{n}\otimes\mathbf{n}). \tag{39}$$

The squares $W^2_{\alpha\mathbf{nQ}}$ of the characteristic frequencies clearly weight the contribution of each material resonance ω_α into the eigenvalue tensor (36). The structure of Eq. (38) is very similar to the expression of the oscillator strength $f_{21}=2m\omega_{21}d^2_{21}/\hbar$ of a two-level system with a transition frequency ω_{21} and a dipole moment d_{21} [25]. Indeed, the quantities $W_{\alpha\mathbf{nQ}}$ can be used to re-express the light-matter coupling Hamiltonian $\mathcal{H}_{\rm int}$ from Eq. (14):

$$\mathcal{H}_{\rm int}=i\sum_{\mathbf{Q},\boldsymbol{\epsilon}_\mathbf{Q},\alpha,\mathbf{n}}\frac{\hbar W_{\alpha\mathbf{nQ}}}{2}\sqrt{\frac{\omega_{c\mathbf{Q}}}{\omega_{\alpha\mathbf{Q}}}}(\boldsymbol{\epsilon}_\mathbf{Q}\mathbf{n})\times\left(a^{\dagger}_{\mathbf{Q},\boldsymbol{\epsilon}_\mathbf{Q}}-a_{-\mathbf{Q},\boldsymbol{\epsilon}_\mathbf{Q}}\right)(b^{\dagger}_{-\mathbf{Q}\alpha}+b_{\mathbf{Q}\alpha}). \tag{40}$$

To obtain this expression, we have used the decomposition of the light polarization vectors $\boldsymbol{\epsilon}_\mathbf{Q}=\sum_\mathbf{n}(\boldsymbol{\epsilon}_\mathbf{Q}\mathbf{n})\mathbf{n}$ into the orthonormal basis $\mathbf{n}$. This form of the Hamiltonian comforts our interpretation of $W_{\alpha\mathbf{nQ}}$ as light-matter coupling strengths; therefore we shall refer to their squares $W^2_{\alpha\mathbf{nQ}}$ defined in Eq. (38) as "oscillator strengths."

Together with the tensor (37) an important role is played by its inverse $\bar{\bar{\epsilon}}(\omega,\mathbf{Q})^{-1}$, which, as shown in Appendix A, can be expressed in the basis $\mathbf{n}=(\mathbf{n}_1,\mathbf{n}_2,\mathbf{n}_3)$ as

$$\bar{\bar{\epsilon}}(\omega,\mathbf{Q})^{-1}=\bar{\bar{I}}+\sum_{i\mathbf{n}}\frac{R^2_{i\mathbf{nQ}}}{\omega^2-\Omega^2_{i\mathbf{Q}}}(\mathbf{n}\otimes\mathbf{n}). \tag{41}$$

Here, the coefficients $R^2_{i\mathbf{nQ}}$ are provided by

$$R^2_{i\mathbf{nQ}}=1/\sum_{\alpha}\frac{W^2_{\alpha\mathbf{nQ}}}{\left(\omega^2_{\alpha\mathbf{Q}}-\Omega^2_{i\mathbf{Q}}\right)^2}. \tag{42}$$

These coefficients allow to express the new bosonic operators $\Pi^{\dagger}_{\mathbf{Q}ni}$ that diagonalize the Hamiltonian (35):

$$\mathcal{H}_{\rm mat}+\mathcal{H}_{\rm P2}=\sum_{i\mathbf{Q}\mathbf{n}}\hbar\Omega_{i\mathbf{Q}}\Pi^{\dagger}_{\mathbf{Q}ni}\Pi_{\mathbf{Q}ni}, \tag{43}$$

$$\Pi^{\dagger}_{\mathbf{Q}ni}=\frac{R_{i\mathbf{nQ}}}{\sqrt{2\varepsilon_0 V\hbar\Omega_{i\mathbf{Q}}}}\sum_{\alpha}(\mathbf{n}\cdot\mathbf{d}^{*}_{\mathbf{Q}\alpha})\times\left[\frac{b^{\dagger}_{\mathbf{Q}\alpha}}{\Omega_{i\mathbf{Q}}-\omega_{\alpha\mathbf{Q}}}+\frac{b_{-\mathbf{Q}\alpha}}{\Omega_{i\mathbf{Q}}+\omega_{\alpha\mathbf{Q}}}\right]. \tag{44}$$

Making use of these operators, the expression of the light-matter coupling Hamiltonian $\mathcal{H}_{\rm int}$ becomes

$$\mathcal{H}_{\rm int}=i\sum_{\mathbf{Q},\boldsymbol{\epsilon}_\mathbf{Q},i,\mathbf{n}}\frac{\hbar R_{i\mathbf{nQ}}}{2}\sqrt{\frac{\omega_{c\mathbf{Q}}}{\Omega_{i\mathbf{Q}}}}(\boldsymbol{\epsilon}_\mathbf{Q}\mathbf{n})\times\left(a^{\dagger}_{\mathbf{Q},\boldsymbol{\epsilon}_\mathbf{Q}}-a_{-\mathbf{Q},\boldsymbol{\epsilon}_\mathbf{Q}}\right)(\Pi^{\dagger}_{-\mathbf{Q}ni}+\Pi_{\mathbf{Q}ni}). \tag{45}$$

This form of the interaction Hamiltonian indicates that the coefficients $R_{i\mathbf{nQ}}$ play the role of light-matter coupling strengths for the new normal boson modes induced from the quadratic self-interaction Hamiltonian $\mathcal{H}_{\rm P2}$. Independently from the interaction with light, the coefficients $R_{i\mathbf{nQ}}$ take into account the local field effects that both contribute to the emergence of collective excitations [37], and also screen the external electromagnetic field applied to the system. With this respect we call the squares $R^2_{i\mathbf{nQ}}$ "collective oscillator strengths."

From Eq. (45), it is clear that the knowledge of $R_{i\mathbf{nQ}}$ alone is not sufficient to quantify the coupling of a particular normal

mode with light. The other contribution in the light-matter coupling strength is provided by the factors $(\boldsymbol{\epsilon}_{\mathbf{Q}}\mathbf{n})$ in Eq. (45). These factors correspond to a selection rule for the collective normal modes, expressing the fact that light couples only with the transverse part of the material polarization. Let us define the transverse unit tensor through

$$\bar{\bar{I}}_{\mathrm{t}} = \bar{\bar{I}} - \frac{\mathbf{Q}\otimes\mathbf{Q}}{Q^2}. \tag{46}$$

Respectively, the transverse part of the inverse characteristic tensor is

$$\bar{\bar{\epsilon}}(\omega,\mathbf{Q})_{\mathrm{t}}^{-1} = \bar{\bar{I}}_{\mathrm{t}}\bar{\bar{\epsilon}}(\omega,\mathbf{Q})^{-1}\bar{\bar{I}}_{\mathrm{t}}. \tag{47}$$

The PZW Hamiltonian (13) is now expressed as a function of the new collective modes, and the only interaction that remains is the coupling with light. By proceeding with the complete diagonalization, as detailed in Appendix A, we obtain the eigenvalue determinant which provides the frequencies of the new light-matter coupled polariton modes:

$$\left\| \left(\omega^2/\omega_{c\mathbf{Q}}^2\right)\bar{\bar{I}}_{\mathrm{t}} - \bar{\bar{\epsilon}}(\omega,\mathbf{Q})_{\mathrm{t}}^{-1} \right\| = 0. \tag{48}$$

This equation is identical with the dispersion relation for an electromagnetic wave propagating in a homogeneous and anisotropic medium with a dielectric tensor $\bar{\bar{\epsilon}}(\omega,\mathbf{Q})_{\mathrm{t}}$ [38]. We shall therefore refer to the tensor $\bar{\bar{\epsilon}}(\omega,\mathbf{Q})$ as the "dielectric" tensor of the homogeneous anisotropic medium. Indeed, the tensor (36) encapsulates all the information about the dielectric properties of the medium described by a polarization field in the form of Eq. (9). In particular, its transverse part describes the propagation of electromagnetic waves in the medium in the sense of the microscopic Maxwell's equations.

Along with the transverse part, we can also define a longitudinal (scalar) part of the dielectric tensor (36):

$$\epsilon_{\mathrm{l}}(\omega,\mathbf{Q}) = \frac{1}{Q^2}\mathbf{Q}\cdot\bar{\bar{\epsilon}}(\omega,\mathbf{Q})\cdot\mathbf{Q}, \tag{49}$$

$$\epsilon_{\mathrm{l}}(\omega,\mathbf{Q}) = 1 - \sum_{\alpha} V_{\alpha\alpha}\frac{4\hbar\omega_{\alpha\mathbf{Q}}}{\hbar^2\left(\omega^2-\omega_{\alpha\mathbf{Q}}^2\right)}, \tag{50}$$

$$V_{\alpha\alpha} = \frac{(\mathbf{Q}\mathbf{d}_{\mathbf{Q}\alpha})(\mathbf{Q}\mathbf{d}_{\mathbf{Q}\alpha}^*)}{2\varepsilon_0 V Q^2}. \tag{51}$$

The coefficients $V_{\alpha\alpha}$ are proportional to the Fourier components of self-interaction terms contained in the longitudinal Coulomb potential (20). We can recognize in these expressions the definition of the Linhard dielectric function [28]. This dielectric function is often used to obtain the longitudinal collective modes of the system, driven by the Coulomb potential, through the equation $\epsilon_{\mathrm{l}}(\omega,\mathbf{Q}) = 0$ [39]. In the formalism developed here, this equation appears as a particular case of Eq. (37). Indeed, the coupled boson modes provided by Eq. (37) can be, in general, nor purely transverse or purely longitudinal. Respectively, the basis of eigenvectors $\mathbf{n} = (\mathbf{n}_1,\mathbf{n}_2,\mathbf{n}_3)$ is not necessarily related to the basis $(\mathbf{Q},\boldsymbol{\varepsilon}_{1\mathbf{Q}},\boldsymbol{\varepsilon}_{2\mathbf{Q}})$ which allows the separation of the polarization into transverse and longitudinal components, as in Eqs. (16) and (17). With this respect, the dielectric tensor introduced by Eq. (36) and, respectively, Eq. (37) can be considered as a generalization of the Linhard formula. Note that this results can not be recovered in the framework of the linear response theory, where the material polarization is induced by an external perturbation. As the perturbation is usually purely transverse (light field) or purely longitudinal (probe charge), then the resulting collective excitations are imposed to be respectively transverse or longitudinal.

However, the classification into transverse and longitudinal modes becomes exact in the case where two of the eigenvectors $\mathbf{n} = (\mathbf{n}_1,\mathbf{n}_2,\mathbf{n}_3)$ belong to the transverse subspace spanned by $(\boldsymbol{\varepsilon}_{1\mathbf{Q}},\boldsymbol{\varepsilon}_{2\mathbf{Q}})$. In particular, this is the case of an isotropic medium, where the dielectric tensor $\bar{\bar{\epsilon}}(\omega,\mathbf{Q})$ [Eq. (36)] is scalar-diagonal, and any orthogonal basis can be used as an eigen-basis $\mathbf{n}$. In that case, if we use the basis $(\mathbf{Q},\boldsymbol{\varepsilon}_{1\mathbf{Q}},\boldsymbol{\varepsilon}_{2\mathbf{Q}})$, the distinction between transverse and longitudinal eigenvalues becomes pertinent.

2. *Properties of the dielectric tensor*

The dielectric tensor (36) has a simple analytical structure, which allows to infer the general properties of the coupled-boson excitations which arise from the dipole-dipole interactions contained in the Hamiltonian $\mathcal{H}_{\mathrm{P2}}$. The first property is the conservation of the total oscillator strength between the initial excitations, described by the operators $b_{\mathbf{Q}\alpha}$ and the coupled modes described by $\Pi_{\mathbf{Q}ni}$, that is expressed with the sum rule:

$$\sum_{\alpha} W_{\alpha\mathbf{n}\mathbf{Q}}^2 = \sum_{i} R_{i\mathbf{n}\mathbf{Q}}^2. \tag{52}$$

To demonstrate this property, let us consider a particular main axis $\mathbf{n}$ and take the asymptotic limit of the corresponding component of the inverse dielectric tensor $\varepsilon_{nn}^{-1}(\omega,\mathbf{Q})$ as $\omega\to\infty$ [Eq. (41)]:

$$\varepsilon_{nn}^{-1}(\omega\to\infty,\mathbf{Q}) = 1 - \frac{1}{\omega^2}\sum_{i} R_{i\mathbf{n}\mathbf{Q}}^2 + o\left(\frac{1}{\omega^2}\right). \tag{53}$$

The same asymptotic limit can be obtained from the direct definition in Eq. (39):

$$\begin{aligned}\varepsilon_{nn}(\omega\to\infty,\mathbf{Q})^{-1} &= \left[1 + \frac{1}{\omega^2}\sum_{\alpha} W_{\alpha\mathbf{n}\mathbf{Q}}^2 + o\left(\frac{1}{\omega^2}\right)\right]^{-1} \\ &= 1 - \frac{1}{\omega^2}\sum_{\alpha} W_{\alpha\mathbf{n}\mathbf{Q}}^2 + o\left(\frac{1}{\omega^2}\right).\end{aligned} \tag{54}$$

Identifying the leading terms of the asymptotic expansions in Eqs. (53) and (54), we obtain the desired result stated in Eq. (52).

The second property is the redistribution of the oscillator strength of the system and the emergence of a single collective state that collects almost all the oscillator strength contained in the sum of Eq. (52) [18]. To illustrate this property, let us consider a system with three distinct transition frequencies, $\alpha = 1,2,3$, along a particular main axis $\mathbf{n}$ with $\omega_1 < \omega_2 < \omega_3$. We consider that all these transitions have an identical coupling with light:

$$W_{\alpha\mathbf{n}\mathbf{Q}}^2 = \tfrac{1}{3}\omega_0^2 = \mathrm{const}, \quad \alpha = 1,2,3. \tag{55}$$

For the purpose of this illustration, we take $\omega_1 = 0.5\omega_0$, $\omega_1 = 0.7\omega_0$ and $\omega_3 = \omega_0$. For simplicity, the dispersion

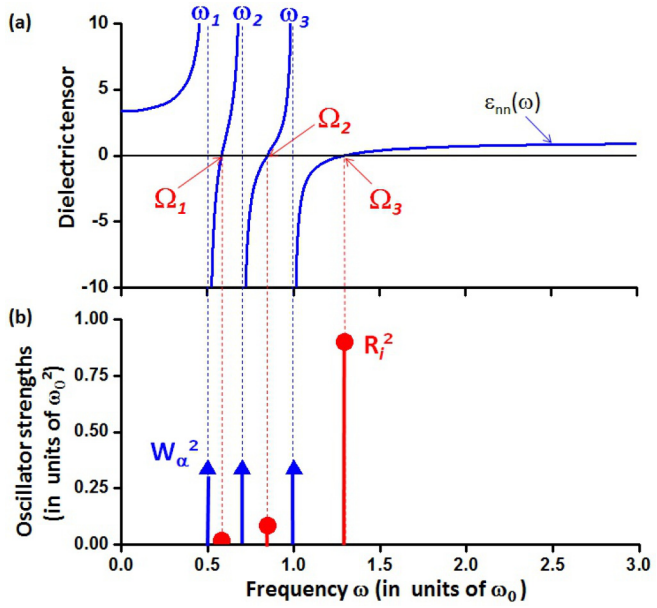

FIG. 1. (Color online) (a) Plot of the dielectric tensor component $\varepsilon_{nn}(\omega)$ with the transition frequencies ω_α and oscillator strengths W_α^2 as indicated in the text [Eq. (55)]. The eignenfrequencies of the normal modes Ω_i are obtained as the zeros of $\varepsilon_{nn}(\omega)$ [Eq. (37)]. (b) The corresponding oscillator strengths of the collective modes R_i^2 (circles) and the original oscillator strengths W_α^2 (triangles).

(wave-vector dependence) is not considered, and we drop the index $\mathbf{Q}$. The resulting dielectric tensor component $\varepsilon_{nn}(\omega)$ is plotted in Fig. 1(a) as a function of the frequency, indicating the zeros that correspond to the new eigenvalues Ω_i of the system. In Fig. 1(b), we have represented the initial oscillator strengths $W^2_{\alpha\mathbf{nQ}}$ together with the collective oscillator strengths $R^2_{i\mathbf{n}}$ of the coupled modes as provided by Eq. (42). From this figure we see that the first two coupled eigenstates Ω_1 and Ω_2 have very low oscillator strengths, while the highest frequency state Ω_3 has a much increased oscillator strength $R^2_{3\mathbf{n}}$, equal to 90% of the total oscillator strength ω_0^2. If the corresponding light transition is allowed ($\mathbf{n}.\boldsymbol{\varepsilon}_\mathbf{Q} \neq 0$), then this highest frequency coupled mode will be the only bright state of the system, while all other modes will remain dark.

This property is very general. Let us take a particular main direction $\mathbf{n}$ with N matter excitations $\omega_{\alpha\mathbf{Q}}$, considered nondegenerate for simplicity. According to the algebraic structure of the dielectric tensor, we can deduce that there will be N new normal modes with frequencies $\Omega_{i\mathbf{Q}}$ ($i = 1, \ldots, N$) that obey the following inequalities:

$$\omega_{1\mathbf{Q}} < \Omega_{1\mathbf{Q}} < \omega_{2\mathbf{Q}} < \Omega_{2\mathbf{Q}} < \ldots \omega_{N\mathbf{Q}} < \Omega_{N\mathbf{Q}}. \tag{56}$$

The highest frequency collective mode $\Omega_{N\mathbf{Q}}$ therefore satisfies

$$\Omega^2_{N\mathbf{Q}} - \omega^2_{1\mathbf{Q}} < \Omega^2_{N\mathbf{Q}} - \omega^2_{\alpha\mathbf{Q}} < \Omega^2_{N\mathbf{Q}} - \omega^2_{N\mathbf{Q}}. \tag{57}$$

Using the fact that $\Omega_{N\mathbf{Q}}$ obeys Eq. (37) and exploiting the explicit form of the dielectric tensor (39), we obtain the framing inequalities for the highest frequency eigenvalue:

$$\omega^2_{1\mathbf{Q}} + \sum_\alpha W^2_{\alpha\mathbf{nQ}} < \Omega^2_{N\mathbf{Q}} < \omega^2_{N\mathbf{Q}} + \sum_\alpha W^2_{\alpha\mathbf{nQ}}. \tag{58}$$

According to the first set of inequalities (56) all frequencies $\Omega_{i<N\mathbf{Q}}$ but the last remain in the vicinity of the original frequencies $\omega_{\alpha\mathbf{Q}}$. This is also seen from Fig. 1(a). Remarkably, the second set of inequalities (58) shows that the total oscillator strength of the system, defined in Eq. (52), appears as a lower bound to the highest frequency eigenstate $\Omega_{N\mathbf{Q}}$. In the situation where $\sum_\alpha W^2_{\alpha\mathbf{nQ}} \gg \omega^2_{N\mathbf{Q}}$ the value of $\Omega_{N\mathbf{Q}}$ becomes significantly larger than the original frequencies. As a consequence, its collective oscillator strength $R_{N\mathbf{nQ}}$ becomes very large. This can be inferred from Eq. (42), as in the limit $\sum_\alpha W^2_{\alpha n} \gg \omega^2_{N\mathbf{Q}}$, we can neglect the bare frequencies $\omega_{\alpha\mathbf{Q}}$ in the denominator with respect to $\Omega_{N\mathbf{Q}}$. Then using Eqs. (58) and (42), we obtain:

$$\Omega^2_{N\mathbf{Q}} \approx \sum_\alpha W^2_{\alpha\mathbf{nQ}}, \tag{59}$$

$$R^2_{N\mathbf{nQ}} \approx \sum_\alpha W^2_{\alpha\mathbf{nQ}}. \tag{60}$$

As the total oscillator strength is preserved according to Eq. (52), the last equality, Eq. (60), means that all other coupled states become dark. This can be also seen from Eq. (42). Indeed, when $i < N$, the corresponding solution $\Omega_{i<N\mathbf{Q}}$ is close to one of the initial frequencies $\omega_{\alpha\mathbf{Q}}$ and the denominator in Eq. (42) acquires a very large value, resulting in vanishing value of the corresponding oscillator strength, $R^2_{i<N\mathbf{Q}} \to 0$.

The dominant contribution to the inverse dielectric tensor is then the highest frequency solution:

$$\varepsilon(\omega,\mathbf{Q})^{-1}_{nn} \approx 1 + \frac{\Omega^2_{N\mathbf{Q}}}{\omega^2 - \Omega^2_{N\mathbf{Q}}}. \tag{61}$$

In the case where the selection rule ($\mathbf{n}.\boldsymbol{\varepsilon}_\mathbf{Q} \neq 0$) is satisfied, then the optical properties of the system are governed by this single bright collective excitation. In solid state physics, this situation can be achieved when the initial spectrum of the system (transition frequencies $\omega_{\alpha\mathbf{Q}}$) covers a finite frequency band, and if the number of particles is large, so that the overall oscillator strength $\sum_\alpha W^2_{\alpha n}$ is high. Recently, similar states have been observed with intersubband excitations of a bidimensional electron gas in highly doped quantum wells [18]. In the next part, we will show that the density plasma waves in a 3D electron gas, and in particular the surface plasmon polartion can also be described in that framework. We will also show that in this case the coefficients $W_{\alpha\mathbf{nQ}}$ and $R_{i\mathbf{nQ}}$ have a meaning of plasma frequencies.

III. APPLICATION TO THE 3D ELECTRON GAS

In order to continue our exploration of the PZW quantum electromagnetic Hamiltonian and to illustrate its close relationship with the solid state physics, we now apply it to a particular system: the three-dimensional electron gas. From the conceptional point of view, it is interesting to show that the PZW representation is not restricted only to the case of spatially bounded systems, such as atoms an molecules, but can also be conveniently applied to systems where electrons are completely delocalized in one or more directions of space. Furthermore, we show that it is possible to construct the polarization field of the system, in the form of Eq. (9), solely from the kinetic part of the electronic Hamiltonian

$\mathcal{H}_e$ and the corresponding single-particle wave functions that are free-running electronic waves. The combination of this polarization field, together with the electromagnetic PZW Hamiltonian from Eq. (13) then allows to deduce both the interaction between the electrons and their coupling with the free space modes of the electromagnetic field.

This approach is conditioned by an approximation, which enables us to associate a bosonic polarization field to the ensemble of electrons, that are fundamentally fermionic particles. This approximation is nothing else but the RPA bosonisation of the gas [12]. The dielectric tensor of the system, as obtained from Eq. (36) then coincides with the one provided by Linhard formula [11], and the corresponding coupled states are either the collective plasmonic excitation of the 3D gas, or the Stoner continuum of single-particle excitations. On this basis, we provide a fully microscopic theory of the surface plasmon-polarion modes hosted by an interface between the 3D gas and a homogeneous dielectric. This description of the plasmon-polariton modes can then be used to study their quantum-vacuum properties [20,33]. In the final section of this part, we compare the PZW Hamiltonian of the 3D gas with the minimal-coupling Hamiltonian through an inverse unitary transform, on the grounds of the general results from Sec. II.

A. RPA bosonization and polarization operator

The kinetic energy Hamiltonian for a nonrelativistic 3D free electron gas is

$$\mathcal{H}_e = \sum_{\mathbf{k}} E_{\mathbf{k}} c_{\mathbf{k}}^\dagger c_{\mathbf{k}}, \tag{62}$$

$$E_{\mathbf{k}} = \hbar\omega_{\mathbf{k}} = \frac{\hbar^2 \mathbf{k}^2}{2m}. \tag{63}$$

Here, $\mathbf{k}$ denotes the electronic wave vectors and $E_{\mathbf{k}}$ is the corresponding kinetic energy, m is the electron mass, and $c_{\mathbf{k}}^\dagger$ and $c_{\mathbf{k}}$ are the fermionic creation and annihilation operators of a state with a wave vector $\mathbf{k}$. The spatial wave functions of these states are the plane waves:

$$\varphi_{\mathbf{k}}(\mathbf{r}) = \frac{1}{\sqrt{V}} e^{i\mathbf{k}\mathbf{r}} \tag{64}$$

with V the quantization volume. We do not consider phenomena related to the spin, therefore we do not introduce a specific notation for it, however, each wave-vector state with momentum $\hbar\mathbf{k}$ is considered as two fold degenerate.

The ground state of the Hamiltonian (62) is the Fermi sphere with radius equal to the Fermi wave vector k_F:

$$|F\rangle = \prod_{|\mathbf{k}|<k_F} c_{\mathbf{k}}^\dagger |0\rangle. \tag{65}$$

We are now seeking to identify the elementary dipolar excitations required for the construction the PZW Hamiltonian of the electron gas. For this, we consider the excited states with a momentum transfer $\hbar\mathbf{Q}$ across the Fermi sphere; these correspond to a hole with a momentum $\hbar|\mathbf{k}| < \hbar k_F$ and an electron with a momentum $\hbar|\mathbf{k}+\mathbf{Q}| > \hbar k_F$ [12]. These excited states are described by the action of an operator $d_{\mathbf{Qk}}^\dagger$ on the ground state $|F\rangle$ defined as

$$d_{\mathbf{Qk}}^\dagger = c_{\mathbf{k}+\mathbf{Q}}^\dagger c_{\mathbf{k}}, \quad \begin{cases} |\mathbf{k}| < k_F \\ |\mathbf{k}+\mathbf{Q}| > k_F \end{cases}. \tag{66}$$

It is well known from the RPA approach [12,39] that if we restrict the description of the system on the subspace $\mathcal{S}$ spanned by the fundamental state and the single-particle states defined above, $\mathcal{S} = \{|F\rangle, d_{\mathbf{Qk}}^\dagger |F\rangle\}$, then the operators defined in Eq. (66) obey bosonic commutation rules:

$$[d_{\mathbf{Qk}}, d_{\mathbf{Q'k'}}^\dagger]|_{\mathcal{S}} = \delta_{\mathbf{k},\mathbf{k'}}\delta_{\mathbf{Q},\mathbf{Q'}}. \tag{67}$$

We can therefore use the operators $d_{\mathbf{Qk}}^\dagger$ as elementary dipole excitation operators in order to apply the approach developed in the previous part. For this, we need to construct a polarization field operator $\hat{\mathbf{P}}(\mathbf{r})$, which is a linear combination of the elementary boson operators (66) and their Hermitian conjugates, like in Eq. (9). To this end, we first compute the electronic current that corresponds to the quantum transition from the fundamental to excited states of the system $\hat{\mathbf{j}}(\mathbf{r})$, and then we define $\hat{\mathbf{P}}(\mathbf{r})$ through the relation (21) [22]. In the absence of a static magnetic field applied to the gas, the expression of the current in the PZW representation lacks the diamagnetic contribution and is provided by

$$\hat{\mathbf{j}}(\mathbf{r}) = \frac{ie\hbar}{2m}[\hat{\Psi}^\dagger \nabla \hat{\Psi} - \nabla \hat{\Psi}^\dagger \hat{\Psi}]. \tag{68}$$

Here, $\hat{\Psi}(\mathbf{r})$ is the field operator constructed from the plane-wave electronic wave functions:

$$\hat{\Psi}(\mathbf{r}) = \sum_{\mathbf{k}} c_{\mathbf{k}} \varphi_{\mathbf{k}}(\mathbf{r}) = \sum_{\mathbf{k}} c_{\mathbf{k}} \frac{e^{i\mathbf{k}\mathbf{r}}}{\sqrt{V}}. \tag{69}$$

The expression of the current is therefore

$$\hat{\mathbf{j}}(\mathbf{r}) = \frac{\hbar e}{2mV} \sum_{\mathbf{k},\mathbf{Q}} (2\mathbf{k}+\mathbf{Q}) c_{\mathbf{k}+\mathbf{Q}}^\dagger c_{\mathbf{k}} e^{-i\mathbf{Q}\mathbf{r}}. \tag{70}$$

Let us first consider the $\mathbf{Q} = \mathbf{0}$ contribution in the above sum:

$$\hat{\mathbf{j}}_{\mathbf{Q}=\mathbf{0}} = \frac{\hbar e}{mV} \sum_{\mathbf{k}} \mathbf{k} c_{\mathbf{k}}^\dagger c_{\mathbf{k}}. \tag{71}$$

This contribution is proportional to the total electronic momentum, which is a conserved quantity. It commutes with the Hamiltonian (62) and can not contribute to the dynamic polarization density according to the definition (21). Therefore the contribution $\mathbf{Q} = \mathbf{0}$ is excluded from now on, although this will not appear explicitly in the notations.

To establish the polarization density through Eq. (21), we notice that the following commutation relation holds:

$$[\mathcal{H}_e, c_{\mathbf{k}+\mathbf{Q}}^\dagger c_{\mathbf{k}}] = \hbar\Delta\omega_{\mathbf{kQ}} c_{\mathbf{k}+\mathbf{Q}}^\dagger c_{\mathbf{k}}, \tag{72}$$

$$\Delta\omega_{\mathbf{kQ}} = \omega_{\mathbf{k}+\mathbf{Q}} - \omega_{\mathbf{k}} = \frac{\hbar}{2m}\mathbf{Q}(2\mathbf{k}+\mathbf{Q}). \tag{73}$$

Note that for the single particle excitations defined in Eq. (66), we have necessarily $\Delta\omega_{\mathbf{kQ}} > 0$. Then a possible definition for the polarization field, which satisfies Eq. (21) according to

Eq. (72) is

$$\hat{\mathbf{P}}(\mathbf{r}) = \frac{-ie}{V}\sum_{\mathbf{Q}} \hat{\mathbf{P}}^{\dagger}_{\mathbf{Q}} e^{-i\mathbf{Q}\mathbf{r}}, \tag{74}$$

where the vector operator $\hat{\mathbf{P}}^{\dagger}_{\mathbf{Q}}$ is defined as

$$\hat{\mathbf{P}}^{\dagger}_{\mathbf{Q}} = \sum_{\mathbf{k}} \boldsymbol{\beta}_{\mathbf{Q}\mathbf{k}} c^{\dagger}_{\mathbf{k}+\mathbf{Q}} c_{\mathbf{k}}, \tag{75}$$

$$\boldsymbol{\beta}_{\mathbf{Q}\mathbf{k}} = \frac{2\mathbf{k}+\mathbf{Q}}{\mathbf{Q}(2\mathbf{k}+\mathbf{Q})}. \tag{76}$$

The operator $\mathbf{P}^{\dagger}_{\mathbf{Q}}$ satisfies the relation

$$\hat{\mathbf{P}}^{\dagger}_{\mathbf{Q}} = -\hat{\mathbf{P}}_{-\mathbf{Q}}. \tag{77}$$

The polarization field in Eq. (75) is obtained from the most general properties of the free particle Hamiltonian (62). However, Eq. (75) can not be used yet as a definition of the polarization operator. Indeed, the commutation rule of Eq. (12) is not necessarily satisfied because of the fermionic commutation rules obeyed by the operators $c^{\dagger}_{\mathbf{k}}$ and $c_{\mathbf{k}}$. However, if we restrict the action of the polarization field operator [Eq. (75)] onto the subspace $\mathcal{S}$ of RPA states, defined in the paragraph after Eq. (66), we can obtain a self-consistent definition owe to the bosonic commutation rules in Eq. (67). This restriction can be obtained from Eq. (75) by splitting the sum over the wave vectors into $|\mathbf{k}| > k_F$ and $|\mathbf{k}| < k_F$ contributions. For the $|\mathbf{k}| > k_F$ states, we make the substitution $\mathbf{k} \to \mathbf{k}+\mathbf{Q}$ and we retain the sum only over the wave vectors that satisfy Eq. (66). As a result, we obtain the restricted version $\widetilde{\mathbf{P}}^{\dagger}_{\mathbf{Q}} = \hat{\mathbf{P}}^{\dagger}_{\mathbf{Q}}|_{\mathcal{S}}$ of the polarization operator on the RPA states:

$$\begin{aligned}\widetilde{\mathbf{P}}^{\dagger}_{\mathbf{Q}} &= \sum_{\mathbf{k}\in\mathcal{S}} \boldsymbol{\beta}_{\mathbf{Q}\mathbf{k}}(d^{\dagger}_{\mathbf{Q}\mathbf{k}} + d_{-\mathbf{Q}-\mathbf{k}}) \\ &= \sum_{\mathbf{k}} f_{\mathbf{k}}(1-f_{\mathbf{Q}+\mathbf{k}})\boldsymbol{\beta}_{\mathbf{Q}\mathbf{k}}(d^{\dagger}_{\mathbf{Q}\mathbf{k}} + d_{-\mathbf{Q}-\mathbf{k}}).\end{aligned} \tag{78}$$

Here, we defined the occupation number $f_{\mathbf{k}} = \langle F|c^{\dagger}_{\mathbf{k}}c_{\mathbf{k}}|F\rangle$ of the state $\mathbf{k}$. Thanks to this definition, we can conveniently extend the sum in the second line of Eq. (78) over all wave vectors $\mathbf{k}$.

Using the polarization field operator defined through Eqs. (74) and (78), we obtain the following PZW bosonized Hamiltonian of the electron gas interacting with the free electromagnetic waves:

$$\mathcal{H} = \mathcal{H}_{\mathrm{e}} + \mathcal{H}_{\mathrm{ph}} + \mathcal{H}_{\mathrm{int}} + \mathcal{H}_{\mathrm{P2}}, \tag{79}$$

$$\mathcal{H}_{\mathrm{int}} = -\sum_{\mathbf{Q},\boldsymbol{\epsilon}_{\mathbf{Q}}} \sqrt{\frac{e^2\hbar\omega_{c\mathbf{Q}}}{2\varepsilon_0 V}}(\boldsymbol{\epsilon}_{\mathbf{Q}}\widetilde{\mathbf{P}}^{\dagger}_{\mathbf{Q}})\left(a_{\mathbf{Q},\boldsymbol{\epsilon}_{\mathbf{Q}}} - a^{\dagger}_{-\mathbf{Q},\boldsymbol{\epsilon}_{\mathbf{Q}}}\right), \tag{80}$$

$$\mathcal{H}_{\mathrm{P2}} = \frac{e^2}{2\varepsilon_0 V}\sum_{\mathbf{Q}} \widetilde{\mathbf{P}}^{\dagger}_{\mathbf{Q}}\widetilde{\mathbf{P}}_{\mathbf{Q}}. \tag{81}$$

As explained in the previous part, the interaction between the electrons is contained in the square-polarization term $\mathcal{H}_{\mathrm{P2}}$. It is remarkable that the PZW approach allows the derivation of this term only from the knowledge of the free electron Hamiltonian $\mathcal{H}_{\mathrm{e}}$ [Eq. (62)]. We will see in Sec. III D how this term is related to the Coulomb interaction between the RPA states.

Note that because of the commutation rules (67) and (72) the restriction of the electronic Hamiltonian on the subspace $\mathcal{S}$ behaves as an effective bosonic Hamiltonian:

$$\mathcal{H}_{\mathrm{e}}|_{\mathcal{S}} \to \sum_{\mathbf{kQ}} \hbar\Delta\omega_{\mathbf{kQ}} d^{\dagger}_{\mathbf{Qk}} d_{\mathbf{Qk}}. \tag{82}$$

The PZW model of the electron gas described here therefore maps exactly in the general Hopfield-like model described in the previous part, which allows us to use directly the results stated there.

B. Linhard dielectric tensor and plasmon modes

If the PZW Hamiltonian theory exposed in the previous sections is correct, then the dielectric tensor provided by Eq. (36) should coincide with the Linhard dielectric tensor of the electron gas [11]. Respectively, the collective excitations of the system described in Sec. IIB2 should coincide with the well-known bulk plasmon and plasmon-polariton modes of the 3DEG [12].

Applying the definition of the dielectric tensor (36) to the Hamiltonian from Eqs. (79)–(81), we obtain

$$\begin{aligned}\bar{\bar{\epsilon}}(\omega,\mathbf{Q}) = \bar{\bar{I}} &- \frac{e^2}{\varepsilon_0 V\hbar} \\ &\times \sum_{\mathbf{k}} f_{\mathbf{k}}(1-f_{\mathbf{Q}+\mathbf{k}})\frac{2\Delta\omega_{\mathbf{kQ}}(\boldsymbol{\beta}_{\mathbf{Qk}}\otimes\boldsymbol{\beta}_{\mathbf{Qk}})}{(\omega^2-\Delta\omega^2_{\mathbf{kQ}})}.\end{aligned} \tag{83}$$

Let us consider the expression of this tensor in the basis $(\boldsymbol{\epsilon}_{1\mathbf{Q}},\boldsymbol{\epsilon}_{2\mathbf{Q}},\mathbf{Q}/Q)$. Since the electron gas is isotropic, for symmetry reasons, it is clear that the tensor is diagonal in this basis. The following expressions are obtained for the longitudinal and transverse components:

$$\begin{aligned}\epsilon(\omega,\mathbf{Q})_{\mathrm{l}} = 1 &- \frac{e^2}{\varepsilon_0 V\hbar Q^2} \\ &\times \sum_{\mathbf{k}} f_{\mathbf{k}}(1-f_{\mathbf{Q}+\mathbf{k}})\frac{2\Delta\omega_{\mathbf{kQ}}}{\omega^2-\Delta\omega^2_{\mathbf{kQ}}},\end{aligned} \tag{84}$$

$$\begin{aligned}\epsilon(\omega,\mathbf{Q})_{\mathrm{t}} = 1 &- \frac{e^2}{\varepsilon_0 V\hbar Q^2}, \\ &\times \sum_{\mathbf{k}} f_{\mathbf{k}}(1-f_{\mathbf{Q}+\mathbf{k}})\frac{2\Delta\omega_{\mathbf{kQ}}}{\omega^2-\Delta\omega^2_{\mathbf{kQ}}}F_{\perp}(\mathbf{k},\mathbf{Q}),\end{aligned} \tag{85}$$

$$F_{\perp}(\mathbf{k},\mathbf{Q}) = \frac{2\left[Q^2k^2-(\mathbf{kQ})^2\right]}{(2\mathbf{kQ}+Q^2)^2}. \tag{86}$$

Using the substitution $\mathbf{k}' = -\mathbf{k}-\mathbf{Q}$, the above expressions can be recast in a more familiar form:

$$\epsilon(\omega,\mathbf{Q})_{\mathrm{l}} = 1 - \frac{e^2}{\varepsilon_0 V\hbar Q^2}\sum_{\mathbf{k}} \frac{f_{\mathbf{k}}-f_{\mathbf{Q}+\mathbf{k}}}{\omega-\Delta\omega_{\mathbf{kQ}}}, \tag{87}$$

$$\epsilon(\omega,\mathbf{Q})_{\mathrm{t}} = 1 - \frac{e^2}{\varepsilon_0 V\hbar Q^2}\sum_{\mathbf{k}} \frac{f_{\mathbf{k}}-f_{\mathbf{Q}+\mathbf{k}}}{\omega-\Delta\omega_{\mathbf{kQ}}}F_{\perp}(\mathbf{k},\mathbf{Q}). \tag{88}$$

We can recognize in these equations the Linhard result for the RPA dielectric tensor of the electron gas [11]. Note

that previously this result has been obtained from a first-order perturbation theory, while now it appears as a closed solution of the electronic PZW Hamiltonian. The collective excitations of the system are provided, according to Eq. (37), as the zeros of the dielectric tensor. From the structure of the dielectric functions in Eqs. (84) and (85) as well as the results in Sec. IIB2, it is clear that most of the solutions of Eq. (37) will have frequencies that are very close to the bare transition frequencies $\Delta\omega_{\mathbf{kQ}}$ and vanishing effective oscillator strengths, $R^2 \to 0$. These solutions form the particle-hole continuum of the electron gas [39].

The collective modes with frequencies $\Omega \gg \Delta\omega_{\mathbf{kQ}}$ can be obtained in a closed form in the limit $\mathbf{Q} \to 0$ [12,39]. In that limit, Eqs. (84) and (85) acquire the form of Drude scalar dielectric constants:

$$\epsilon(\omega,\mathbf{Q})_{\mathrm{l}} = \epsilon(\omega,\mathbf{Q})_{\mathrm{t}} = 1 - \frac{W_P^2}{\omega^2}, \tag{89}$$

$$W_P^2 = \frac{e^2 N_e}{\varepsilon_0 m V}. \tag{90}$$

Here, N_e is the total number of electrons in the quantization volume V and W_P is the bulk plasma frequency of the electron gas [12]. It follows immediately that the inverse of the dielectric function is

$$\epsilon(\omega,\mathbf{Q})_{\mathrm{l}}^{-1} = \epsilon(\omega,\mathbf{Q})_{\mathrm{t}}^{-1} = 1 + \frac{W_P^2}{\omega^2 - W_P^2}. \tag{91}$$

The zeros of the Drude constant (89) provide the collective excitation of the gas as bulk plasmon modes with a frequency W_P, and comparing Eq. (91) with Eq. (61), we conclude that these states are the only ones that have nonvanishing effective oscillator strength in the sense of the R^2 coefficients introduced in the previous part. In this case, according to Eq. (91) the square of the plasma frequency W_P^2 plays the role of effective oscillator strength for the plasmon modes. The structure of the dielectric tensor indicates that there are two transverse plasmon modes, able to couple with light, and one dark longitudinal mode. Using Eqs. (48) and (91), we obtain the dispersion relation of the bulk plasmon-polaritons modes resulting from the coupling between the transverse plasmons and the electromagnetic waves, in the long-wavelength limit:

$$\omega^2\left(\omega^2 - W_P^2 - \omega_{c\mathbf{Q}}^2\right) = 0. \tag{92}$$

The nonzero frequency solution of this equation, $\omega_{\mathbf{Q}}^{+} = \sqrt{W_P^2 + \omega_{c\mathbf{Q}}^2}$, is twofold degenerate with one branch for each polarization vector $\boldsymbol{\epsilon}_{\mathbf{Q}}$. Since the typical light wave vectors $\mathbf{Q}$ are much smaller than the Fermi wave vector k_F for a dense gas, Eq. (92) is virtually always consistent with the long-wavelength approximation.

The PZW Hamiltonian in Eqs. (79)–(81) therefore allows to recover the well-known excitation spectrum of the electron gas, also recalled in Fig. 2(a). In Fig. 2(b), we have indicated the corresponding effective oscillator strengths, in analogy with Fig. 1, for a fixed value of the wave vector $\mathbf{Q} = \mathbf{Q}_0$.

The advantage of our formalism is to be able to provide the underlying quantum picture of the collective plasmon modes. Using the results from Sec. II B and Appendix A, we can express the collective bosonic operators that describe the

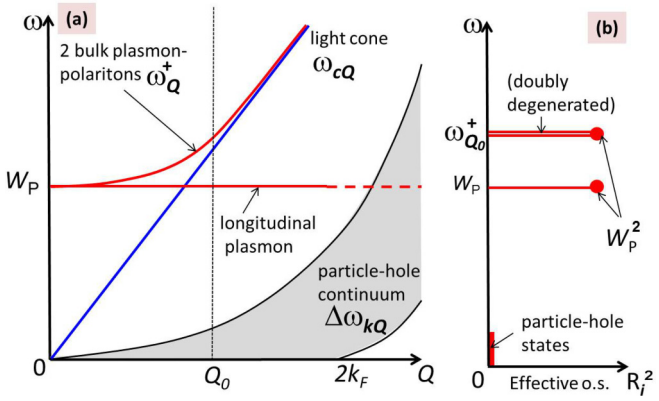

FIG. 2. (Color online) (a) Excitation diagram of the electron gas as provided by the PZW Hamiltonian described by Eqs. (79)–(81), in the long-wavelength limit. (b) The corresponding effective oscillator strengths, in therms of the R^2 parameters defined in the previous part. The bright states of the systems are two transverse plasmon-polaritons, while the longitudinal plasmon is dark by construction.

plasma oscillations of the electron gas as a coherent sum over the electron-hole excitations across the Fermi sphere, in the case where $W_P \gg \Delta\omega_{\mathbf{kQ}}$:

$$\Pi_{\mathbf{Qn}}^{\dagger} = \sum_{\mathbf{k}\in\mathcal{S}} \frac{-ie(\boldsymbol{\beta}_{\mathbf{Qk}}\cdot\mathbf{n})}{\sqrt{2\varepsilon_0\hbar W_P V}} \times\left[\left(d_{\mathbf{Qk}}^{\dagger} + d_{-\mathbf{Q}-\mathbf{k}}\right) + \frac{\Delta\omega_{\mathbf{kQ}}}{W_P}\left(d_{\mathbf{Qk}}^{\dagger} - d_{-\mathbf{Q}-\mathbf{k}}\right)\right]. \tag{93}$$

Here, $\mathbf{n}$ is one of the eigenvectors $(\mathbf{Q}/Q, \boldsymbol{\epsilon}_{1\mathbf{Q}}, \boldsymbol{\epsilon}_{2\mathbf{Q}})$. The first-order terms $\Delta\omega_{\mathbf{kQ}}/W_P$ in the expansion (93) are needed to preserve the bosonic commutation rules, $[\Pi_{\mathbf{Qn}}, \Pi_{\mathbf{Q'n}}^{\dagger}] = \delta_{\mathbf{QQ'}}$, yet these terms do not contribute to the polarization field of the plasmon modes. The latter can be expressed using the results of Appendix A:

$$\hat{\mathbf{P}}(\mathbf{r}) = \sqrt{\frac{\varepsilon_0\hbar W_P}{2V}} \sum_{\mathbf{Qn}} \mathbf{n}(\Pi_{\mathbf{Qn}}^{\dagger} + \Pi_{-\mathbf{Qn}}) e^{-i\mathbf{Qr}}. \tag{94}$$

This expression will be used in the next section for the derivation of the quantum theory of surface plasmon-polaritons. We can use Eq. (94) in order to obtain a simplified version of the electronic PZW Hamiltonian Eqs. (79)–(81), which is restrained only on the collective plasmon modes:

$$\mathcal{H} = \mathcal{H}_{\mathrm{e}}^{\mathrm{eff}} + \mathcal{H}_{\mathrm{ph}} + \mathcal{H}_{\mathrm{int}}^{\mathrm{eff}}, \tag{95}$$

$$\mathcal{H}_{\mathrm{e}}^{\mathrm{eff}} = \sum_{\mathbf{Qn}} \hbar W_P \Pi_{\mathbf{Qn}}^{\dagger}\Pi_{\mathbf{Qn}}, \tag{96}$$

$$\mathcal{H}_{\mathrm{int}}^{\mathrm{eff}} = -i \sum_{\mathbf{Q},\boldsymbol{\epsilon}_{\mathbf{Q}}} \frac{\hbar\sqrt{W_P\omega_{c\mathbf{Q}}}}{2} \times \left(a_{\mathbf{Q},\boldsymbol{\epsilon}_{\mathbf{Q}}} - a_{-\mathbf{Q},\boldsymbol{\epsilon}_{\mathbf{Q}}}^{\dagger}\right)\left(\Pi_{-\mathbf{Q}\boldsymbol{\epsilon}_{\mathbf{Q}}} + \Pi_{\mathbf{Q}\boldsymbol{\epsilon}_{\mathbf{Q}}}^{\dagger}\right). \tag{97}$$

For each polarization vector $\boldsymbol{\epsilon}_{\mathbf{Q}}$, this Hamiltonian describes two coupled quantum oscillators: the plasmon excitation at a frequency W_P and the electromagnetic wave at frequency $\omega_{c\mathbf{Q}}$. This Hamiltonian is studied in further details

in Appendix B, where, in particular, we provide the bosonic plasmon-polariton operators, describing the light-matter interaction for this system, in a closed form.

C. Surface plasmon-polariton

1. Dispersion relation

The usual approach to quantize the surface-plasmon polaritons starts by defining the normal modes of the system, which are the classical solutions of the Maxwell's equation for an interface between an electron gas and a dielectric, in the form of surface waves bound on the interface [29,30]. Then creation and annihilation operators are associated to the normal modes, following closely the quantization procedure of free electromagnetic waves [24]. The difficulty of this approach is the proper normalisation of the normal mode amplitudes, or in other words, the definition of a "quantum of plasmon." This normalization is essential, as it allows to access the probability amplitudes of elementary processes, such as the spontaneous and stimulated emission of surface plasmons, and the quantum optical properties of the plasmon waves [33]. One of the reasons why this normalization could become a difficult problem is the strong frequency dispersion, related to the plasma resonances of the gas [40]. Then one has to define a Hamiltonian of the matter-assisted electromagnetic field which takes into account the dispersion. For instance, a Hamiltonian based on the classical electromagnetic energy density for dispersive media [38] has been proposed [31]. However, this approach is phenomenological, rather than truly microscopic, as it relies on the classical dielectric function of the gas. Other difficulties arise from the fact that the surface plasmon-polariton is an evanescent wave. In that case, it is not completely clear whether the use of the standard commutation relations for the electromagnetic field, derived in the case of homogeneous plane waves, is well-justified [41,42], although many authors have assumed their validity.

Here, we show that we can obtain a quantum-mechanical description of the SPPs, based on the PZW representation developed in the previous part, that is free from the aforementioned difficulties. We benefit from the fact that the electromagnetic and the polarization part are well separate in the PZW Hamiltonian, which allows to define the electromagnetic density energy without invoking any additional hypothesis concerning the dispersion. For the description of the matter part we shall make use of the collective plasma operators defined in Eq. (93) and the corresponding polarization operator as provided by Eq. (94). We therefore build a "bottom-up" approach [32] that relies on the elementary microscopic excitations of the gas, and the system is supposed to be ideally conservative. Respectively, if one wants to include the dissipation of the system, one also has to adopt a microscopic approach. For instance, the dissipation can be introduced by coupling the longitudinal part of the electronic polarization with a bath of dark polarization modes, like in the seminal paper by Huttner and Barnet [2].

The geometry of the problem is depicted in Fig. 3(a). We consider an interface, perpendicular to the z axis, between two dielectric media with background dielectric constants ε_1 and ε_2. The medium with a constant ε_2 is hosting a three-dimensional electron gas. This is a slightly more general

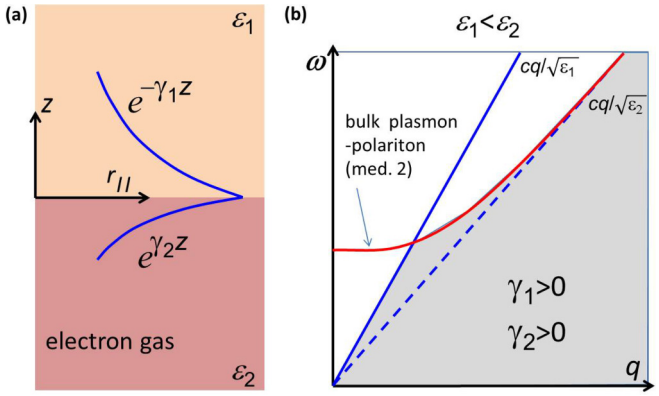

FIG. 3. (Color online) (a) An interface between two dielectric media with constants ε_1 and ε_2. The medium 2 is hosting an electron gas. A surface wave is propagating on the interface, with evanescent tails into each media. (b) The domain of allowed dispersion for the surface wave depicted in (a), for the case $\varepsilon_1 < \varepsilon_2$. The asymptotes $\omega = cq/\sqrt{\varepsilon_{1,2}}$ indicate the light cone for each medium.

situation than the one described in the previous section, and it requires a proper renormalization of the plasma frequency, as we shall see further.

The problem will be treated in a self-consistent way, in which we will postulate the existence of a surface mode propagating on the interface between the two media 1 and 2, and we will deduce its dispersion relation from the PZW Hamiltonian expressed with the collective operators from Eq. (93). As the interface at $z = 0$ breaks the translational invariance along the z axis, the three-dimensional wave vectors $\mathbf{Q}$ are split into planar $\mathbf{q}$ and perpendicular components on each side of the interface: $\mathbf{Q}_{1,2} = (\mathbf{q}, i\gamma_{1,2})$. Respectively, the position vector is split into in-plane and perpendicular components, $\mathbf{r} = (\mathbf{r}_{||}, z)$. The presence of an interface enables the existence of evanescent waves in both media 1 and 2. These waves decay exponentially on both sides of the interface, meaning that both γ_1 and γ_2 are real. We assume that the surface mode with a frequency $\omega_{\mathbf{q}}$ is constructed from such evanescent waves, therefore it satisfies the Helmholtz dispersion relation stemming from Eqs. (48) and (89) for each media:

$$\varepsilon_1 \frac{\omega_{\mathbf{q}}^2}{c^2} = \mathbf{q}^2 - \gamma_1^2, \tag{98}$$

$$\varepsilon_2 \frac{\omega_{\mathbf{q}}^2 - W_P^2}{c^2} = \mathbf{q}^2 - \gamma_2^2. \tag{99}$$

For convenience, we have redefined the plasma frequency in this part as $W_P^2 = N_e e^2/m\varepsilon_0\varepsilon_2 V$. The condition for existence of a surface wave confined to the interface is $\gamma_1^2 > 0$ and $\gamma_2^2 > 0$. For instance, for the case $\varepsilon_1 < \varepsilon_2$ this condition corresponds to the shaded region defined below the light cone for the media 1 and the bulk plasmon-polariton dispersion curve for the medium 2, as illustrated in Fig. 3(b). This case is most common for the interface between air and doped semiconductor.

The dispersion relation $\omega_{\mathbf{q}} = \omega_{\mathbf{q}}(\mathbf{q})$ of the surface wave is then obtained by assuming that it satisfies the geometrical constraints in Eqs. (98) and (99), being altogether an eigenvalue

solution of the PZW Hamiltonian. This Hamiltonian is written starting from the general expression from Eq. (95). The spatial integrals in the photonic part are expressed for a general TM (transverse magnetic)-polarized electromagnetic field:

$$\mathcal{H} = \mathcal{H}_{\rm ph} + \mathcal{H}_{\rm e}^{\rm eff} + \mathcal{H}_{\rm int}^{\rm eff}, \tag{100}$$

$$\mathcal{H}_{\rm ph} = \frac{1}{2\varepsilon_0}\int \mathbf{D}^2 \frac{1}{\varepsilon(z)} d^3\mathbf{r} + \frac{\mu_0}{2}\int \hat{H}_\perp^2 d^3\mathbf{r}, \tag{101}$$

$$\mathcal{H}_{\rm e}^{\rm eff} = \sum_{\mathbf{Qn}} \hbar W_P \Pi_{\mathbf{nQ}}^\dagger \Pi_{\mathbf{nQ}}, \tag{102}$$

$$\mathcal{H}_{\rm int}^{\rm eff} = -\frac{1}{2\varepsilon_2\varepsilon_0}\int \hat{\mathbf{D}}\hat{\mathbf{P}}\theta(-z) d^3\mathbf{r}. \tag{103}$$

Here, $\theta(z)$ is the Heaviside step function, and we assume that the integral in the interaction part $\mathcal{H}_{\rm int}^{\rm eff}$ [Eq. (103)] extends only in the medium 2, while the expression of $\hat{\mathbf{P}}$ is the one derived for an infinite medium in the long-wavelength approximation from Eq. (94). In Eq. (101), the piecewise dielectric function is defined as $\varepsilon(z) = \theta(z)\varepsilon_1 + \theta(-z)\varepsilon_2$. It is important to note that the expression of the electromagnetic energy density $\mathcal{H}_{\rm ph}$ is that of a free electromagnetic field, and therefore ε_2 is the dielectric constant of the medium 2 in the absence of the electron gas. Just like for the homogeneous problem treated in the previous part, the dispersive Drude-like dielectric function in the form of Eq. (89) will appear through the inclusion of the plasma and interaction contribution $\mathcal{H}_{\rm e}^{\rm eff}$ and $\mathcal{H}_{\rm int}^{\rm eff}$, and only after the full Hamiltonian of the system [Eq. (100)] has been diagonalized. In this approach, the photonic Hamiltonian $\mathcal{H}_{\rm ph}$ always describes a free electromagnetic field. This is an important feature of the PZW representation, that allows for an unambiguous definition of the photon energy density independently from the material dispersion.

The electromagnetic field components of the TM-polarized surface waves are

$$\hat{D}_z = A_0 \sum_{\mathbf{q}} iq\hat{D}_{\mathbf{q}} e^{i\mathbf{qr}_{||}}[\theta(z)e^{-\gamma_1 z} + \theta(-z)e^{\gamma_2 z}], \tag{104}$$

$$\hat{D}_{||} = A_0 \sum_{\mathbf{q}} \hat{D}_{\mathbf{q}} e^{i\mathbf{qr}_{||}}[\theta(z)\gamma_1 e^{-\gamma_1 z} - \theta(-z)\gamma_2 e^{\gamma_2 z}], \tag{105}$$

$$\hat{H}_\perp = A_0 \sum_{\mathbf{q}} \hat{H}_{\mathbf{q}} e^{i\mathbf{qr}_{||}}[\theta(z)e^{-\gamma_1 z} + \theta(-z)e^{\gamma_2 z}], \tag{106}$$

$$A_0 = \sqrt{\frac{4\varepsilon_0\hbar c}{S}}. \tag{107}$$

Here, S is the sample area, and the value of the normalization constant A_0 is chosen for further convenience. In the above equations, $\hat{\mathbf{D}}$ is purely traverse field, $\mathrm{div}\hat{\mathbf{D}} = 0$ as required by the PWZ picture. The electric and magnetic fields are described in terms of scalar intensity operators $\hat{D}_{\mathbf{q}}$ and $\hat{H}_{\mathbf{q}}$, rather than with creation and destruction operators, which is more convenient for evanescent waves. The hermiticity of the field operators requires that

$$\hat{D}_{\mathbf{q}}^\dagger = \hat{D}_{-\mathbf{q}}, \qquad \hat{H}_{\mathbf{q}}^\dagger = \hat{H}_{-\mathbf{q}}. \tag{108}$$

Using Eqs. (98), (99), (101) and, (104)–(107), we obtain the following expression of the photon Hamiltonian $\mathcal{H}_{\rm ph}$:

$$\mathcal{H}_{\rm ph} = \hbar c \sum_{\mathbf{q}} (\alpha_{\mathbf{q}} \hat{D}_{\mathbf{q}}\hat{D}_{-\mathbf{q}} + \beta_{\mathbf{q}}\hat{H}_{\mathbf{q}}\hat{H}_{-\mathbf{q}}), \tag{109}$$

$$\alpha_{\mathbf{q}} = \frac{q^2+\gamma_1^2}{\varepsilon_1\gamma_1} + \frac{q^2+\gamma_2^2}{\varepsilon_2\gamma_2}, \tag{110}$$

$$\beta_{\mathbf{q}} = \frac{\omega_{\mathbf{q}}^2}{c^2}\left(\frac{1}{\gamma_1} + \frac{1}{\gamma_2}\right). \tag{111}$$

To determine completely the fields, we also need to impose the following commutation relations on the scalar operators $\hat{D}_{\mathbf{q}}$ and $\hat{H}_{\mathbf{q}}$ [24]:

$$[\hat{D}_{\mathbf{q}}, \hat{D}_{\mathbf{q}'}] = 0, \qquad [\hat{H}_{\mathbf{q}}, \hat{H}_{\mathbf{q}'}] = 0, \tag{112}$$

$$[\hat{D}_{\mathbf{q}}, \hat{H}_{\mathbf{q}'}^\dagger] = [\hat{D}_{\mathbf{q}}, \hat{H}_{-\mathbf{q}'}] = iC_{\mathbf{q}}\delta_{\mathbf{q},\mathbf{q}'}. \tag{113}$$

These equations lead to the standard commutation relations for the annihilation and creation operators in the case of plane waves, once the latter have been defined through the quadrature operators $\hat{D}_{\mathbf{q}}$ and $\hat{H}_{\mathbf{q}}$. The coefficient $C_{\mathbf{q}}$ is determined from the requirement that the fields satisfy the forth Maxwell's equation (in the absence of external currents): $d\mathbf{D}/dt = \mathrm{rot}\mathbf{H}$. The latter can be expressed, for the quantum field operators, in a Hamiltonian form as

$$\frac{1}{i\hbar}[\hat{\mathbf{D}}, \mathcal{H}] = \frac{1}{i\hbar}[\hat{\mathbf{D}}, \mathcal{H}_{\rm ph}] = \mathrm{rot}\hat{\mathbf{H}}_\perp. \tag{114}$$

Indeed, thanks to the commutation relations (112) and (113), as well as the fact that the matter operators commute with the field operators in the PZW picture, only $\mathcal{H}_{\rm ph}$ contributes to the evolution of the electric field. Consequently, using the expressions of the field components and $\mathcal{H}_{\rm ph}$ the constant $C_{\mathbf{q}}$ is determined to be

$$C_{\mathbf{q}} = \frac{1}{2\frac{\omega_{\mathbf{q}}}{c}\left(\frac{1}{\gamma_1} + \frac{1}{\gamma_2}\right)}. \tag{115}$$

The electromagnetic fields are now fully determined, and the only free parameter that remains at this stage is the plasmon-polariton frequency $\omega_{\mathbf{q}}$. The next step is to explicit the interaction Hamiltonian $\mathcal{H}_{\rm int}^{\rm eff}$ by imposing the geometrical constraints of the problem. Using Eqs. (104), (105), and (94), we obtain

$$\begin{aligned} &-\frac{1}{2\varepsilon_2\varepsilon_0}\int \hat{\mathbf{D}}\hat{\mathbf{P}} d^3\mathbf{r} \\ &= -\frac{\hbar}{\varepsilon_2}\sqrt{\frac{2\varepsilon_2 W_P c}{L}} \sum_{\mathbf{Q}=(\mathbf{q},q_z)} \hat{D}_{\mathbf{q}} \\ &\quad \times \left[\frac{iq(\Pi_{\mathbf{Q},z}^\dagger + \Pi_{-\mathbf{Q},z}) - \gamma_2(\Pi_{\mathbf{Q},||}^\dagger + \Pi_{-\mathbf{Q},||})}{\gamma_2 - iq_z}\right]. \end{aligned} \tag{116}$$

To obtain the above expression, we split the three-dimensional wave vector $\mathbf{Q}$ in Eq. (94) into its in-plane and z component $\mathbf{Q} = (\mathbf{q}, q_z)$. The denominator $\gamma_2 - iq_z$ arises from the integral $\int e^{-iq_z z + \gamma_2 z}\theta(-z)dz$, and L is the quantization length of the electron gas in the z direction. Furthermore, we have

considered plasmon oscillations where $\mathbf{n}$ points either along the plane (subscript "||") or along the z axis (subscript "z").

The sum over the spatial wave vectors q_z can be carried over in order to obtain more compact bosonic operators that integrate the spatial variations of the polarization field. This procedure is analogous to the one in the previous section, where we performed a sum over the electronic vectors $\mathbf{k}$ in order to define collective plasmonic operators $\Pi^{\dagger}_{\mathbf{Qn}}$ [Eq. (93)]. Let us define a new bosonic creation operator:

$$B^{\dagger}_{\mathbf{q}} = N_{\mathbf{q}} \sum_{q_z} \frac{iq\Pi^{\dagger}_{\mathbf{Q},z} - \gamma_2 \Pi^{\dagger}_{\mathbf{Q},||}}{\gamma_2 - iq_z}. \tag{117}$$

The constant $N_{\mathbf{q}}$ is obtained requiring that $[B_{\mathbf{q}}, B^{\dagger}_{\mathbf{q}'}] = \delta_{\mathbf{q},\mathbf{q}'}$, which provides the normalisation condition:

$$1 = N^2_{\mathbf{q}}\left(q^2 + \gamma_2^2\right) \sum_{q_z} \frac{1}{q_z^2 + \gamma_2^2}. \tag{118}$$

Converting the sum into integral, we obtain

$$\sum_{q_z} \frac{1}{q_z^2 + \gamma_2^2} = \frac{L}{2\pi} \int_{-\infty}^{\infty} \frac{dq_z}{q_z^2 + \gamma_2^2} = \frac{L}{2\gamma_2}.$$

Replacing the value of $N_{\mathbf{q}}$ in Eq. (117) and using the definitions of $B^{\dagger}_{\mathbf{q}}$ in the interaction Hamiltonian we obtain the following compact expression for $\mathcal{H}_{\text{int}}$:

$$\mathcal{H}_{\text{int}} = -\hbar \sum_{\mathbf{q}} \sqrt{\frac{W_P\left(q^2 + \gamma_2^2\right)c}{\varepsilon_2 \gamma_2}} \hat{D}_{\mathbf{q}}\left(B^{\dagger}_{\mathbf{q}} + B_{-\mathbf{q}}\right). \tag{119}$$

As expected, the quantization parameters S and L have disappeared from the final result. Since the definition of Eq. (117) is in fact a unitary transformation, and, furthermore, W_P does not have any $\mathbf{q}$ dependence (at least, in the long-wavelength limit, which is well satisfied for a dense gas where $q \ll k_F$), we can express the electronic plasma Hamiltonian as

$$\mathcal{H}^{\text{eff}}_{\text{e}} = \sum_{\mathbf{q}} \hbar W_P B^{\dagger}_{\mathbf{q}} B_{\mathbf{q}}. \tag{120}$$

This effective Hamiltonian now sums only over the spatial plasmonic modes that are coupled to the surface wave. Having expressed the total Hamiltonian in terms of the quantities $\hat{D}_{\mathbf{q}}$,$\hat{H}_{\mathbf{q}}$, $B^{\dagger}_{\mathbf{q}}$, and $B_{\mathbf{q}}$, we now seek to diagonalize the problem by introducing a new polaritonic operator,

$$Y_{\mathbf{q}} = m_{\mathbf{q}} B_{\mathbf{q}} + h_{\mathbf{q}} B^{\dagger}_{-\mathbf{q}} + z_{\mathbf{q}} \hat{D}_{\mathbf{q}} + t_{\mathbf{q}} \hat{H}_{\mathbf{q}}, \tag{121}$$

such as $[Y_{\mathbf{q}}, \mathcal{H}] = \hbar\omega_{\mathbf{q}} Y_{\mathbf{q}}$, where, as mentioned before, we require for self-consistency that $\omega_{\mathbf{q}}$ is the frequency of the surface wave. The corresponding Hopfield matrix is

$$\begin{bmatrix} W_P - \omega_{\mathbf{q}} & 0 & 0 & i\Omega_{\mathbf{q}} C_{\mathbf{q}} \\ 0 & -W_P - \omega_{\mathbf{q}} & 0 & i\Omega_{\mathbf{q}} C_{\mathbf{q}} \\ -\Omega_{\mathbf{q}} & \Omega_{\mathbf{q}} & -\omega_{\mathbf{q}} & -2i\alpha_{\mathbf{q}} C_{\mathbf{q}} c \\ 0 & 0 & 2i\beta_{\mathbf{q}} C_{\mathbf{q}} c & -\omega_{\mathbf{q}} \end{bmatrix}. \tag{122}$$

Here, we denoted by $\Omega_{\mathbf{q}}$ the "coupling strength" $\Omega_{\mathbf{q}} = \sqrt{W_P(q^2 + \gamma_2^2)c/\varepsilon_2\gamma_2}$. The characteristic equation of this matrix is

$$\left(\omega^2_{\mathbf{q}} - W_P^2\right)\left(\omega^2_{\mathbf{q}} - 4c^2\alpha_{\mathbf{q}}\beta_{\mathbf{q}}C^2_{\mathbf{q}}\right) - 4c\Omega^2_{\mathbf{q}} W_P \beta_{\mathbf{q}} C^2_{\mathbf{q}} = 0. \tag{123}$$

After some manipulations, Eq. (123) can be recast in the form

$$\frac{\omega^2_{\mathbf{q}}}{c^2}\left(\frac{1}{\gamma_1} + \frac{1}{\gamma_2}\right) = \frac{q^2 + \gamma_1^2}{\varepsilon_1 \gamma_1} + \frac{q^2 + \gamma_2^2}{\gamma_2} \frac{1}{\varepsilon_{2t}(\omega_{\mathbf{q}})}. \tag{124}$$

Here, we introduced the Drude-like dielectric constant of the electron gas in the medium 2:

$$\varepsilon_{2t}(\omega) = \varepsilon_2\left(1 - \frac{W_P^2}{\omega^2}\right). \tag{125}$$

Finally, eliminating $\mathbf{q}$ from Eq. (124) through the use of the constraints from Eqs. (98) and (99), we recover the SPP dispersion relation it its well known form [15]:

$$\frac{\gamma_1}{\varepsilon_1} + \frac{\gamma_2}{\varepsilon_{2t}(\omega_{\mathbf{q}})} = 0. \tag{126}$$

The fact that we are able to recover the dispersion relation of surface plasmons in the form of Eq. (126) shows that our quantum formalism is compatible with the classical boundary conditions for the electromagnetic field [15]. This is due to our initial hypothesis that the polarization field goes abruptly to zero at the interface between the media 2 and 1. A more refined model, that takes into account the quantum confinement of the electrons near the interface can also be used [43].

The microscopic description of the surface plasmon-polariton is completed with the specification of the Hopfield coefficients defined in Eq. (121). These coefficients have been provided in Appendix C, and, as we shall see in the next section, they will be useful for the derivation of the spontaneous emission rate of plasmons.

2. *Spontaneous emission of SPPs*

Once the quantum description of the SPP is available, it can be used to determine the probability amplitudes for elementary processes implying surface plasmons. This will be illustrated with the computation of the spontaneous emission of SPPs by an external point dipole source situated in the vicinity of the gas.

Let us consider for simplicity that $\varepsilon_1 = \varepsilon_2 = 1$, which corresponds to the interface between the air and a metal. The dispersion relation of the SPP in this case is provided in Fig. 4(a). Figure 4(a) also illustrates the layout of the problem: a point source with a dipole moment μ is placed in the vicinity of the electron gas, at a distance $z = z_0$ above the interface. Without loss of generality we shall consider that the dipole is oriented along the z axis. We can associate to this dipole source a polarization field [44]:

$$\mathbf{P}_{\mu}(\mathbf{r}) = \mu \mathbf{e}_z \delta(\mathbf{r} - z_0 \mathbf{e}_z). \tag{127}$$

Here, $\mathbf{e}_z$ is the unit vector of the z axis. We shall restrict to a semiclassical treatment where the polarization density from Eq. (127) is a c number instead of an operator. We thus avoid to be too specific about the emitting system. The PZW interaction Hamiltonian that describes the coupling between the dipole

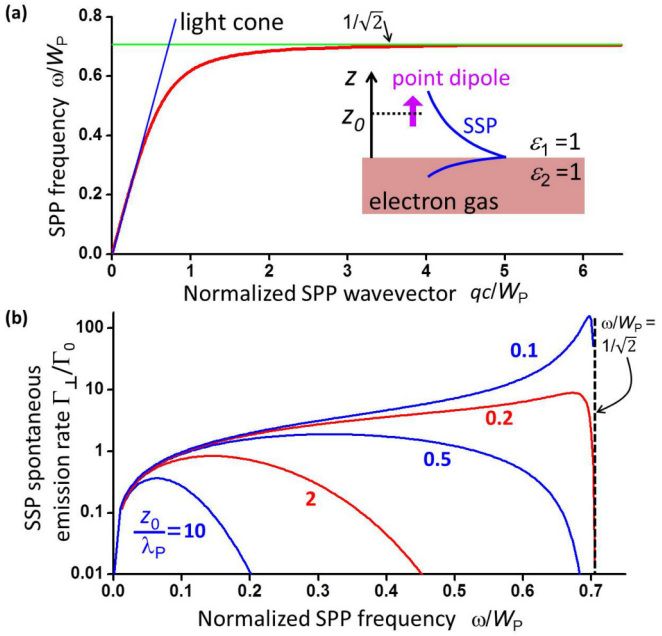

FIG. 4. (Color online) (a) Plasmon polariton dispersion from Eq. (126) for the case $\varepsilon_1 = \varepsilon_2 = 1$. The inset depicts a point dipole, oriented along the z axis, that radiates SPP modes. (b) Spontaneous emission rate of SPP normalized on the free space emission rate $\Gamma_0(\omega)$, as a function of the frequency, for different distances of the dipole with respect to the metal/air interface.

and the SSP displacement field in the medium 1 is then

$$\mathcal{H}_{\rm int}^{\mu-D} = \frac{1}{\varepsilon_0}\int \hat{\mathbf{D}}\mathbf{P}_\mu d^3\mathbf{r} = \frac{-i\mu A_0}{\varepsilon_0}\sum_{\mathbf{q}} q\hat{D}_{\mathbf{q}} e^{-\gamma_1 z_0}. \tag{128}$$

The PZW Hamiltonian also contains a square polarization part associated to Eq. (127), which induces shifts of the energy levels of the emitting system [44]. It can be treated correctly only on a quantum-mechanical basis [44], and therefore we shall ignore it as our emitter is treated in a semiclassical way.

The emission process of surface plasmons can be seen as the quantum transition $|G\rangle \to Y_{\mathbf{q}}^\dagger|G\rangle$ between the ground state of the electron gas $|G\rangle$ and the excited state with one surface plasmon $Y_{\mathbf{q}}^\dagger|G\rangle$, induced by the Hamiltonian $\mathcal{H}_{\rm int}^{\mu-D}$. Note that here $|G\rangle$ is the ground state of the total Hamiltonian in Eq. (100) and does not necessarily coincide with the Fermi sphere $|F\rangle$. The ground state satisfies

$$Y_{\mathbf{q}}|G\rangle = 0. \tag{129}$$

Note that eventually $|G\rangle$ could contain quantum correlations that arise from the coupling of the electron gas with light [20,33]. Using the Fermi's golden rule, we have the following expression for the spontaneous emission rate of surface plasmons at the frequency $\omega_{\mathbf{q}}$:

$$\Gamma_\perp = \frac{2\pi}{\hbar}\left|\langle G|\mathcal{H}_{\rm int}^{\mu-D}Y_{\mathbf{q}}^\dagger|G\rangle\right|^2 \rho_{\rm 2D}(\omega_{\mathbf{q}}). \tag{130}$$

Here, the subscript "$\perp$" refers to the orientation of the dipole, and $\rho_{\rm 2D}(\omega_{\mathbf{q}})$ is the density of states of two-dimensional surface waves:

$$\rho_{\rm 2D}(\omega_{\mathbf{q}}) = \frac{S}{\pi}\frac{qdq}{\hbar d\omega_{\mathbf{q}}}. \tag{131}$$

The probability amplitude for the emission process is provided by the matrix element:

$$\langle G|\mathcal{H}_{\rm int}^{\mu-D}Y_{\mathbf{q}}^\dagger|G\rangle = \frac{-i\mu A_0 q e^{-\gamma_1 z_0}}{\varepsilon_0}\langle G|\hat{D}_{\mathbf{q}}Y_{\mathbf{q}}^\dagger|G\rangle. \tag{132}$$

Using the fact that $\langle G|Y_{\mathbf{q}}^\dagger = (G_{\mathbf{q}}|F\rangle)^\dagger = 0$ we can express this ground-state matrix element as

$$\langle G|\hat{D}_{\mathbf{q}}Y_{\mathbf{q}}^\dagger|G\rangle = \langle G|[\hat{D}_{\mathbf{q}}, Y_{\mathbf{q}}^\dagger]|G\rangle = iC_{\mathbf{q}}t_{\mathbf{q}}^*. \tag{133}$$

Here, we used the definition from Eq. (121) and the commutation relation (113). Using the results from Appendix C, we can now evaluate the spontaneous emission rate from Eq. (130) in a closed form:

$$\Gamma_\perp(\omega_{\mathbf{q}}) = \frac{\mu^2c^2}{\hbar\varepsilon_0}\frac{q^3e^{-2\gamma_1 z_0}}{\omega_{\mathbf{q}}\left(\frac{1}{\gamma_1}+\frac{1}{\gamma_2}\right)}\frac{dq}{d\omega_{\mathbf{q}}} \times \frac{\left(W_P^2-\omega_{\mathbf{q}}^2\right)\left(W_P^2-2\omega_{\mathbf{q}}^2\right)}{\left(W_P^2-\omega_{\mathbf{q}}^2\right)\left(W_P^2-2\omega_{\mathbf{q}}^2\right)+W_P^2\left(q^2+\gamma_2^2\right)c^2}. \tag{134}$$

With the same procedure, one can obtain also the spontaneous emission for a dipole parallel to the interface:

$$\Gamma_\parallel(\omega_{\mathbf{q}}) = \frac{\gamma_1^2}{q^2}\Gamma_\perp(\omega_{\mathbf{q}}). \tag{135}$$

The ratio between the two emission rates is provided simply by the square of the ratio between the in-plane and perpendicular component of the displacement field. It is clear that the spontaneous emission of an arbitrarily oriented dipole source can be recovered as a linear combination of $\Gamma_\parallel$ and $\Gamma_\perp$.

In order to study these results numerically, we have normalized Eq. (134) on the spontaneous emission rate $\Gamma_0(\omega)$ in the free space:

$$\Gamma_0(\omega) = \frac{\mu^2\omega^3}{3\pi\varepsilon_0\hbar c^3}. \tag{136}$$

The dimensionless ratio $\Gamma_\perp/\Gamma_0$ is studied in Fig. 4(b) as a function of the frequency, in the band of allowed SSP frequencies $0 < \omega_{\mathbf{q}} < W_P/\sqrt{2}$. Different plots are provided as a function of the distance z_0, expressed as a fraction of the plasma wavelength $\lambda_P = 2\pi c/W_P$. Similar plots have been provided by Archambault *et al.* [31] on the basis of the classical electromagnetic Hamiltonian for dispersive media [38]. We observe that the emission of SPP is weak for dipoles far from the interface, as should be expected from the factor $e^{-2\gamma_1 z_0}$ in Eq. (134). Furthermore, as the frequency approaches the asymptotic value $W_P/\sqrt{2}$, the emission rate of SPP is strongly suppressed for distances $z_0 \geqslant 0.5\lambda_P$. For dipoles that are sufficiently close, $z_0 \leqslant 0.5\lambda_P$, the emission is actually enhanced as $\omega_{\mathbf{q}}$ approaches $W_P/\sqrt{2}$. This enhancement is due to the high density of states close to the saturation of the dispersion relation of the SPP at high wave vectors [45].

Quite astonishingly, the plots from Fig. 4(b) show that the emission of plasmons is strongly suppressed in the far-infrared

limit $\omega_{\mathbf{q}} \to 0$. This means that the SPP emission rate decays even faster with the frequency than the spontaneous emission in free space [Eq. (136)], which decays as $\sim\omega^3$. Indeed, using the dispersion relation from Eq. (126), we can show that $\gamma_1(\omega_{\mathbf{q}} \to 0) \sim \omega_{\mathbf{q}}^2/cW_P$. From this, we obtain the following asymptotic values for the emission rates:

$$\Gamma_\perp(\omega_{\mathbf{q}} \to 0) \sim \frac{\mu^2}{2\varepsilon_0\hbar W_P c^3}\omega_{\mathbf{q}}^4, \tag{137}$$

$$\Gamma_\parallel(\omega_{\mathbf{q}} \to 0) \sim \frac{\mu^2}{2\varepsilon_0\hbar W_P^3 c^3}\omega_{\mathbf{q}}^6. \tag{138}$$

The origin of this strong decay is the rapid increase of the SPP effective volume V_{eff}, that can be written roughly as $V_{\text{eff}} = S(1/\gamma_1 + 1/\gamma_2)$ [this can be justified, for instance, from Eq. (115)]. In the low-frequency limit, the confinement of the SPP is strongly reduced as $V_{\text{eff}} \sim ScW_P/\omega_{\mathbf{q}}^2$ and the plasmon quanta are diluted in an increasingly large volume. Respectively, the vacuum field amplitude of the SSP mode is strongly reduced.

The approach developed here can be readily extended to other geometries involving SPPs, such as thin film waveguides, double-metal waveguides [46], or localized surface plasmon polaritons [47]. For each specific case, we assign a polarization field of the form (94) only to the regions occupied by the electron gas. In each region of space, we can expand the electromagnetic fields into confined waves which satisfy the Helmholtz dispersion relation in each media. The photonic Hamiltonian is then constructed like the one of free transverse fields, without the inclusion of the material polarization. The Maxwell's equation (114) then provides the vacuum field normalisation of the quadrature operators $\hat{D}_{\mathbf{q}}$ and $\hat{H}_{\mathbf{q}}$ by taking into account the geometrical constraints of the problem. Note that, once the normalization of the field is set if needed, we can always construct ladder operators $a_{\mathbf{q}}$ and $a_{\mathbf{q}}^\dagger$ as linear combinations of $\hat{D}_{\mathbf{q}}$ and $\hat{H}_{\mathbf{q}}$, however, these will not have any straightforward physical interpretation, contrary to the final plasmon-polariton quantum operators in Eq. (121).

D. Discussion of the inverse PZW transformation

In this section, we discuss the correspondence between the PZW theory of the 3D electron gas developed above with the full Hamiltonian in the standard minimal-coupling representation. This correspondence will help us understand better the underlying approximations in the theory. The minimal-coupling Hamiltonian of the 3D gas can be expressed as

$$\mathcal{H}_{\text{st}} = \mathcal{H}_{\text{e}} + \mathcal{H}_{\text{ph}} + \int \hat{\mathbf{j}}\hat{\mathbf{A}}d^3\mathbf{r} + \hat{V}_{\text{Coul}} + \hat{\mathsf{H}}_{\text{A2}}, \tag{139}$$

$$\hat{\mathsf{H}}_{\text{A2}} = \frac{e^2}{2m}\int \hat{\Psi}^\dagger\hat{\Psi}\hat{\mathbf{A}}^2 d^3\mathbf{r}. \tag{140}$$

The fourth therm of Eq. (139) is the Coulomb potential of the jellium model [48]:

$$\hat{V}_{\text{Coul}} = \frac{e^2}{2\varepsilon_0 V}\sum_{\mathbf{Q}\neq\mathbf{0},\mathbf{k},\mathbf{k}'}\frac{1}{Q^2}c_{\mathbf{k}'-\mathbf{Q}}^\dagger c_{\mathbf{k}+\mathbf{Q}}^\dagger c_{\mathbf{k}}c_{\mathbf{k}'}. \tag{141}$$

The last term from Eq. (139), $\hat{\mathsf{H}}_{\text{A2}}$, that is expressed in Eq. (140), is the diamagnetic term that is quadratic in the vector potential. It can be expanded in the basis of electronic plane waves [Eq. (64)] as

$$\hat{\mathsf{H}}_{\text{A2}} = \sum_{\substack{\mathbf{k},\mathbf{Q},\mathbf{Q}'\\ \boldsymbol{\epsilon}_{\mathbf{Q}},\boldsymbol{\epsilon}'_{\mathbf{Q}'}}}\frac{\hbar e^2(\boldsymbol{\epsilon}_{\mathbf{Q}}\cdot\boldsymbol{\epsilon}'_{\mathbf{Q}'})}{4\varepsilon_0 Vm\sqrt{\omega_{c\mathbf{Q}}\omega_{c\mathbf{Q}'}}}c_{\mathbf{k}+\mathbf{Q}-\mathbf{Q}'}^\dagger c_{\mathbf{k}} \times \left(a_{\mathbf{Q},\boldsymbol{\epsilon}_{\mathbf{Q}}} + a_{-\mathbf{Q},\boldsymbol{\epsilon}_{\mathbf{Q}}}^\dagger\right)\left(a_{-\mathbf{Q}',\boldsymbol{\epsilon}'_{\mathbf{Q}'}} + a_{\mathbf{Q}',\boldsymbol{\epsilon}'_{\mathbf{Q}'}}^\dagger\right). \tag{142}$$

Note that in the standard representation $\hat{\mathsf{H}}_{\text{A2}}$ is an operators that acts both on the electronic and photonic degrees of freedom.

In order to compare the standard Hamiltonian [Eqs. (139)–(142)] and the dipolar Hamiltonian [see Eqs. (79)–(81)], we proceed like in Section II A. We apply the unitary transformation from Eq. (24) on the PZW Hamiltonian, in order to obtain its image in the minimal coupling representations. This image Hamiltonian is then compared to the full Hamiltonian described in Eqs. (139)–(142). The generator of the transformation is

$$\int \hat{\mathbf{P}}\hat{\mathbf{A}}d^3r = -i\sum_{\mathbf{Q},\boldsymbol{\epsilon}_{\mathbf{Q}}} A_{\mathbf{Q}}(\boldsymbol{\epsilon}_{\mathbf{Q}}\widetilde{\mathbf{P}}_{\mathbf{Q}}^\dagger)\left(a_{\mathbf{Q},\boldsymbol{\epsilon}_{\mathbf{Q}}} + a_{-\mathbf{Q},\boldsymbol{\epsilon}_{\mathbf{Q}}}^\dagger\right). \tag{143}$$

Using Eq. (29) from Sec. II A, we obtain the following result for the longitudinal part of the square polarization part, which describes the interaction between particles:

$$T^+(\mathcal{H}_{\text{ph}} + \mathcal{H}_{\text{int}} + \mathcal{H}_{\text{P2}})T = \mathcal{H}_{\text{ph}} + \frac{e^2}{2\varepsilon_0 V}\sum_{\substack{\mathbf{Q}\neq\mathbf{0}\\ (\mathbf{k},\mathbf{k}')\in\mathcal{S}}}\frac{1}{Q^2}(d_{\mathbf{Q}\mathbf{k}} + d_{-\mathbf{Q}-\mathbf{k}}^\dagger)(d_{\mathbf{Q}\mathbf{k}'}^\dagger + d_{-\mathbf{Q}-\mathbf{k}'}). \tag{144}$$

(To obtain this equation, we have used the equality $\mathbf{Q}\boldsymbol{\beta}_{\mathbf{k}\mathbf{Q}} = 1$.) Using the definitions of the dipole operators (66) and lumping the $|\mathbf{k}| > k_F$ and $|\mathbf{k}| < k_F$ contributions together in the sum, we recast the expression for the longitudinal part (144) as

$$\frac{e^2}{2\varepsilon_0 V}\sum_{\substack{\mathbf{Q}\neq\mathbf{0},\mathbf{k},\mathbf{k}'\\ (\mathbf{k},\mathbf{k}')\in\mathcal{S}}}\frac{1}{Q^2}c_{\mathbf{k}'-\mathbf{Q}}^\dagger c_{\mathbf{k}+\mathbf{q}}^\dagger c_{\mathbf{k}}c_{\mathbf{k}'} + \frac{e^2}{2\varepsilon_0 V}\sum_{\mathbf{Q}\neq\mathbf{0},\mathbf{k}}\frac{c_{\mathbf{k}}^\dagger c_{\mathbf{k}}}{Q^2}. \tag{145}$$

This equation looks very familiar to the Coulomb potential (141), however, we should bear in mind that it is restricted only on the subspace $\mathcal{S}$ of RPA states defined in Sec. III A. The expression (144) is actually the dipole-dipole part of the Coulomb interaction [39], whereas the exchange-correlation part is excluded from the full Coulomb interaction. This is the reason why we called our approach "dipolar quantum electrodynamics." The second term of the above equation arises from the reordering of the operator product and represents an infinite self-energy of each electron [48]. This part also arises in the classical PZW transform {see Eq. (4.12) in Ref. [23]} and does not influence the physics of the system. Indeed, the corresponding operator commutes with the kinetic energy Hamiltonian $\mathcal{H}_{\text{e}}$ [Eq. (62)], and therefore it does not

contribute to the dynamic (oscillating) polarization field $\hat{\mathbf{P}}$ of the electron gas defined through Eq. (21).

Applying further the inverse PZW transform to the electronic Hamiltonian of Eq. (62), and using the exact commutation relations from Eq. (72) as well as the results from Sec. II A, we obtain

$$T^{+}\mathcal{H}_{e}T = \mathcal{H}_{e} + \int \hat{\mathbf{j}}\hat{\mathbf{A}}d^{3}\mathbf{r} + \mathcal{H}_{\mathrm{A2}}. \tag{146}$$

The second term is the linear coupling between the 3D gas and the electromagnetic field. Once again, we should bear in mind that here the current operator $\hat{\mathbf{j}}$ describes only transitions between the ground state of the gas $|F\rangle$ with the RPA excited states described in Eq. (66). For the square vector potential term, we obtain the following expression:

$$\mathcal{H}_{\mathrm{A2}} = \sum_{\mathbf{k}\in\mathcal{S},\mathbf{Q},\boldsymbol{\epsilon}_{\mathbf{Q}},\boldsymbol{\epsilon}'_{\mathbf{Q}}} \frac{\hbar e^{2}}{4\varepsilon_{0}Vm\omega_{c\mathbf{Q}}} \frac{4(\boldsymbol{\epsilon}_{\mathbf{Q}}\mathbf{k})(\boldsymbol{\epsilon}'_{\mathbf{Q}}\mathbf{k})}{\mathbf{Q}(2\mathbf{k}+\mathbf{Q})} \times \left(a_{\mathbf{Q},\boldsymbol{\epsilon}_{\mathbf{Q}}} + a^{\dagger}_{-\mathbf{Q},\boldsymbol{\epsilon}_{\mathbf{Q}}}\right)\left(a_{-\mathbf{Q},\boldsymbol{\epsilon}'_{\mathbf{Q}}} + a^{\dagger}_{\mathbf{Q},\boldsymbol{\epsilon}'_{\mathbf{Q}}}\right). \tag{147}$$

The sum over the wave vectors can be evaluated explicitly in the long wavelength limit $\mathbf{Q}\to\mathbf{0}$:

$$\mathcal{H}_{\mathrm{A2}} = \sum_{\mathbf{Q},\boldsymbol{\epsilon}_{\mathbf{Q}},\boldsymbol{\epsilon}'_{\mathbf{Q}}} \frac{\hbar e^{2}N_{\mathrm{e}}(\boldsymbol{\epsilon}_{\mathbf{Q}}\boldsymbol{\epsilon}'_{\mathbf{Q}})}{4\varepsilon_{0}Vm\omega_{c\mathbf{Q}}} \times \left(a_{\mathbf{Q},\boldsymbol{\epsilon}_{\mathbf{Q}}} + a^{\dagger}_{-\mathbf{Q},\boldsymbol{\epsilon}_{\mathbf{Q}}}\right)\left(a_{-\mathbf{Q},\boldsymbol{\epsilon}'_{\mathbf{Q}}} + a^{\dagger}_{\mathbf{Q},\boldsymbol{\epsilon}'_{\mathbf{Q}}}\right). \tag{148}$$

The prefactor after the sum symbol can be further expressed as $\hbar W_{P}^{2}/4\omega_{c\mathbf{Q}}$. Comparing this equation with the exact result from Eq. (142), we can identify the approximations of our PZW approach. First, we have considered only the processes that preserve the excitation wave vector $\mathbf{Q}=\mathbf{Q}'$. This approximation clearly neglects the dynamic action of the magnetic field, and the corresponding magnetisation field density, as discussed in Sec. II A. Second, Eq. (148) can be obtained from Eq. (142) by averaging over the fundamental state of the system, since the total number of electrons can be expressed as a ground-state average $N_{\mathrm{e}} = \langle F|\sum_{\mathbf{k}} c^{\dagger}_{\mathbf{k}}c_{\mathbf{k}}|F\rangle$. Formally, we can express these approximations as

$$\mathcal{H}_{\mathrm{A2}} \approx \langle F|\hat{\mathsf{H}}_{\mathrm{A2}}|F\rangle|_{\mathrm{LWL}}. \tag{149}$$

This approximation is valid if the system is weakly excited and the gas remains essentially in its ground state. Futhermore, the long-wavelength limit ("LWL") expresses the fact that the excitation wave vectors $\mathbf{Q}$ remain small as compared to the Fermi wave vector of the gas, k_{F}. These approximations are once again related to the RPA approach, which is valid in the case of a dense electron gas.

In our approach, the polarization field was constructed from excited states that involve only one electron. Presently, it is not clear whether a general expression of the polarization field $\hat{\mathbf{P}}$ of the gas can be constructed beyond the RPA approximation. It is clear that a general expression should involve any number of excited particles. Already, two-particle excitation of the gas, combined with coupling with an external system (phonons) gives rise to nontrivial phenomena, such as superconductivity [39]. Furthermore, the general PZW approach [23] requires that the polarization field operator $\hat{\mathbf{P}}$ is supplemented also with a magnetisation operator $\hat{\mathbf{M}}$. Both the fields $\hat{\mathbf{P}}$ and $\hat{\mathbf{M}}$ then will, in general, be expressed with momentum changing combinations of fermionic operators of the type $c^{\dagger}_{\mathbf{k}+\mathbf{Q}-\mathbf{Q}'}c_{\mathbf{k}}$. In that case, we can speculate that a self-consisted PZW picture beyond the RPA could be provided in a closed form only if the Baker-Hausdor expansion (25) is interrupted at some finite order.

IV. CONCLUSION

In conclusion, we applied the Power-Zineau-Wooley representation of the quantum electrodynamics in order to obtain an extension of the Hopfield polariton model [1] for the case of generally anisotropic and homogeneous medium, featuring frequency dispersion. The advantage of this approach is that the photonic part of the Hamiltonian coincides with the expression for the free electromagnetic field, while the dispersion arises from the matter part of the full Hamiltonian. The latter is expressed with the help of a polarization field operator $\hat{\mathbf{P}}(\mathbf{r})$ which appears as a linear combination of elementary bosonic operators, that are associated with the quantum transitions of the material. As a result, we have derived a general expression of the dielectric tensor of the material that both describes the propagation of electromagnetic waves within the media and also provides the collective excitations of the systems driven by dipole-dipole interactions. In particular, we demonstrated a very general property of the interacting system, which is the emergence of a single collective state that sums coherently the whole oscillator strength of the elementary excitations, while the remaining eigenstates of the PZW Hamultonian have essentially a single-particle behavior and vanishing coupling with light.

Our approach allows therefore to obtain the dielectric tensor of the medium from the microscopic quantum description of the system. We have successfully applied this program in the case of the 3D electron gas. In that case, a remarkable feature of the PZW representation is that the only input information required is the kinetic energy Hamiltonian $\mathcal{H}_{\mathrm{e}}$ of the gas, together with the corresponding basis of free electronic plane waves. The polarization field operator $\hat{\mathbf{P}}(\mathbf{r})$ is then constructed from the constitutive relation in Eq. (21), with the help of the current-density operator $\hat{\mathbf{j}}(\mathbf{r})$. For this we used a particular subset of the excitation spectrum of $\mathcal{H}_{\mathrm{e}}$, which is the ensemble of bosonized RPA electron-hole excitations of the gas. Once the polarization field is known, then the PZW Hamiltonian leads automatically to a self-consistent description of the electron-electron interactions at the dipole-dipole order, as well as the coupling of the gas with the electromagnetic field. The longitudinal plasmon and the transverse plasmon-polaritons then appear as the collective eigenstates of the PZW Hamiltonian with the maximum oscillator strength. Furthermore, they are described by bosonic operators, which are expressed as a coherent quantum superposition of elementary particle-hole excitations.

Based on this approach, we also provide a microscopic quantum theory for the surface plasmon polaritons, that is free from any ambiguities related to the definition of the electromagnetic energy density in a medium with dispersion. This theory can be implemented to a large variety

of geometries. Furthermore the surface-plasmon modes are described explicitly as light-matter coupled excitations, which are linear combinations of microscopic plasmon and photon operators, rather than quantized normal mode solutions of the macroscopic Maxwell's equations. As the expression of the SPP operators are provided in a closed form, this theory can used to describe the quantum-optical properties of the surface plasmons [33].

The focus for this work were systems with internal translational invariance, however the same approach can be applied also for the case of quantum-confined systems. For instance, the intersubband plasmonic excitations of highly doped quantum wells can be treated in a similar framework [18,22]. A generalized treatment of the quantum confined 2D electron gas, including both intersubband and intrasuband excitations will be presented in a subsequent paper.

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APPENDIX A: DIELECTRIC FUNCTION AND EXACT DIAGONALIZATION OF THE BOSONIC MODEL

1. Diagonalization of the matter part

In order to diagonalize the matter Hamiltonian from Eq. (35) through the Hopfield-Bogoliubov method, we introduce the new bosonic operators $\Pi_{\mathbf{Q}i}$ as the linear combinations of the old ones:

$$\Pi_{\mathbf{Q}i} = \sum_{\alpha} \left(m^i_{\alpha\mathbf{Q}} b_{\mathbf{Q}\alpha} + h^i_{\alpha\mathbf{Q}} b^\dagger_{-\mathbf{Q}\alpha} \right). \tag{A1}$$

These new operators satisfy

$$[\Pi_{\mathbf{Q}i}, \mathcal{H}_{\text{mat}} + \mathcal{H}_{\text{P2}}] = \hbar\Omega_{i\mathbf{Q}} \Pi_{\mathbf{Q}i}. \tag{A2}$$

Computing explicitly the above commutator, we have

$$\begin{aligned} &[\Pi_{\mathbf{Q}i}, \mathcal{H}_{\text{mat}} + \mathcal{H}_{\text{P2}}]/\hbar \\ &= \sum_{\alpha} \omega_{\alpha\mathbf{Q}} \left(m^i_{\alpha\mathbf{Q}} b_{\mathbf{Q}\alpha} - h^i_{\alpha\mathbf{Q}} b^\dagger_{-\mathbf{Q}\alpha} \right) \\ &\quad + \sum_{\alpha,\beta} \frac{\mathbf{d}_{\mathbf{Q}\alpha} \mathbf{d}^*_{\mathbf{Q}\beta}}{2\hbar\varepsilon_0 V} \left(m^i_{\beta\mathbf{Q}} - h^i_{\beta\mathbf{Q}} \right) \left(b_{\mathbf{Q}\alpha} + b^\dagger_{-\mathbf{Q}\alpha} \right) \\ &= \Omega_{i\mathbf{Q}} \sum_{\alpha} \left(m^i_{\alpha\mathbf{Q}} b_{\mathbf{Q}\alpha} + h^i_{\alpha\mathbf{Q}} b^\dagger_{-\mathbf{Q}\alpha} \right). \end{aligned} \tag{A3}$$

Let us introduce the vectors

$$\mathbf{H}_{\mathbf{Q}i} = \sum_{\alpha} \frac{\mathbf{d}^*_{\mathbf{Q}\alpha}}{2\hbar\varepsilon_0 V} \left(m^i_{\alpha\mathbf{Q}} - h^i_{\alpha\mathbf{Q}} \right). \tag{A4}$$

Then using the linear independence of the operators $b_{\mathbf{Q}\alpha}$ and $b^\dagger_{-\mathbf{Q}\alpha}$, we can express the Hopfield coefficients as

$$m^i_{\alpha\mathbf{Q}} = \frac{\mathbf{d}_{\mathbf{Q}\alpha} \mathbf{H}_{\mathbf{Q}i}}{\Omega_{i\mathbf{Q}} - \omega_{\alpha\mathbf{Q}}}, \tag{A5}$$

$$h^i_{\alpha\mathbf{Q}} = \frac{\mathbf{d}_{\mathbf{Q}\alpha} \mathbf{H}_{\mathbf{Q}i}}{\Omega_{i\mathbf{Q}} + \omega_{\alpha\mathbf{Q}}}. \tag{A6}$$

These relations allow to rewrite Eq. (A4) in the following vectorial form:

$$\mathbf{H}_{\mathbf{Q}i} = \sum_{\alpha} \frac{\mathbf{d}^*_{\mathbf{Q}\alpha} (\mathbf{d}_{\mathbf{Q}\alpha} \mathbf{H}_{\mathbf{Q}i})}{\hbar\varepsilon_0 V} \frac{2\omega_{\alpha\mathbf{Q}}}{\Omega^2_{i\mathbf{Q}} - \omega^2_{\alpha\mathbf{Q}}}. \tag{A7}$$

We recognize in the above equation the definition of the dielectric tensor in Eq. (36). Indeed, we can recast the above equation in the form:

$$\bar{\bar{\epsilon}}(\Omega_{i\mathbf{Q}}, \mathbf{Q}) \mathbf{H}_{\mathbf{Q}i} = \mathbf{0}. \tag{A8}$$

Since we are looking for nontrivial solutions for the vector $\mathbf{H}_{\mathbf{Q}i}$, this equation is equivalent to Eq. (37).

We must now express the light-matter coupling Hamiltonian in terms of the new operators $\Pi_{\mathbf{Q}i}$. Using the expressions of the Hopfield coefficients (A5) and (A6), we have

$$\Pi_{\mathbf{Q}i} + \Pi^\dagger_{-\mathbf{Q}i} = \sum_{\alpha} \frac{2\mathbf{d}_{\mathbf{Q}\alpha} \mathbf{H}_{\mathbf{Q}i}}{\Omega^2_{i\mathbf{Q}} - \omega^2_{\alpha\mathbf{Q}}} (b_{\mathbf{Q}\alpha} + b^\dagger_{-\mathbf{Q}\alpha}). \tag{A9}$$

We need to inverse Eq. (A9) in order to express the old bosonic operators as a function of the new ones. To carry out this task, we first study some of the properties of the dielectric tensor (36). As outlined in the main text, there is an orthogonal basis $\mathbf{n} = \mathbf{n}_1, \mathbf{n}_2, \mathbf{n}_3$, where this tensor is diagonal:

$$\bar{\bar{\epsilon}}(\omega, \mathbf{Q}) = \bar{\bar{I}} - \sum_{\mathbf{n}\alpha} \frac{|\mathbf{n}\mathbf{F}_{\mathbf{Q}\alpha}|^2}{\omega^2 - \omega^2_{\alpha\mathbf{Q}}} (\mathbf{n} \otimes \mathbf{n}), \tag{A10}$$

$$\mathbf{F}_{\mathbf{Q}\alpha} = \sqrt{\frac{2\omega_{\alpha\mathbf{Q}}}{\varepsilon_0 V \hbar}} \mathbf{d}_{\mathbf{Q}\alpha}. \tag{A11}$$

In this diagonal basis, the secular equation Eq. (37) is equivalent to three scalar equations, which correspond to each of the principal axis of the tensor, of the form

$$\bar{\bar{\epsilon}}_{nn}(\Omega_{i\mathbf{Q}}, \mathbf{Q}) = 0. \tag{A12}$$

Moreover, we can relabel each eigenvalue $\Omega_{i\mathbf{Q}} = \Omega_{i\mathbf{Q}n}$ with the particular axis $\mathbf{n}$ where the above equation is satisfied. Of course, a particular value $\Omega_{i\mathbf{Q}n}$ can be shared by several axes; however, it is clear that we can always chose the corresponding vector $\mathbf{H}_{\mathbf{Q}i}$ to point along one of the directions $\mathbf{n}$. This is evident in the case where the eigenvalue $\Omega_{i\mathbf{Q}n}$ is completely anisotropic, however, if this is not the case, we can always choose a rotation in the corresponding subspace of vectors $\mathbf{H}_{\mathbf{Q}i}$ so that $\mathbf{H}_{\mathbf{Q}i} = Cst \times \mathbf{n}$.

Now let us consider a particular axis $\mathbf{n}$. By construction we have

$$\bar{\bar{\epsilon}}_{nn}(\omega, \mathbf{Q}) = 1 - \sum_{\alpha} \frac{|\mathbf{n}\mathbf{F}_{\mathbf{Q}\alpha}|^2}{\omega^2 - \omega^2_{\alpha\mathbf{Q}}} = \frac{X(\omega^2)}{\prod_{\alpha} \left(\omega^2 - \omega^2_{\alpha\mathbf{Q}} \right)}. \tag{A13}$$

Here, we have regrouped all the denominators such that $\mathbf{n}\mathbf{F}_{\mathbf{Q}\alpha} \neq 0$. It is clear that $X(\omega^2)$ is a polynomial of the same order as the product $\prod_{\alpha} (\omega^2 - \omega^2_{\alpha\mathbf{Q}})$; moreover, since $\bar{\bar{\epsilon}}_{nn}(\omega \to \infty, \mathbf{Q}) \to 1$ and because of Eq. (A12), we have

$$X(\omega^2) = \prod_{i} \left(\omega^2 - \Omega^2_{i\mathbf{Q}n} \right). \tag{A14}$$

The number of the corresponding zeros $\Omega_{i\mathbf{Q}n}$ is exactly equal to the number of frequencies $\omega_{\alpha\mathbf{Q}}$ that contribute to

the denominator of Eq. (A13). We can therefore express the inverse of the dielectric function as

$$\bar{\bar{\epsilon}}_{nn}(\omega,\mathbf{Q})^{-1} = 1 + \sum_i \frac{R^2_{i\mathbf{Q}n}}{\omega^2 - \Omega^2_{i\mathbf{Q}n}}, \tag{A15}$$

which leads directly to Eq. (41). The expression for the coefficients of Eq. (42) are easily obtained as the residues of the rational function $\bar{\bar{\epsilon}}_{nn}(\omega)^{-1}$ at the poles $\Omega^2_{i\mathbf{Q}n}$.

If we define the following vector:

$$\mathbf{R}_{i\mathbf{Q}} = \sum_{\mathbf{n}} R_{i\mathbf{Q}n}\mathbf{n}, \tag{A16}$$

we can express the inverse dielectric tensor in an basis-independent form:

$$\bar{\bar{\epsilon}}(\omega,\mathbf{Q})^{-1} = \bar{\bar{I}} + \sum_i \frac{\mathbf{R}_{i\mathbf{Q}} \otimes \mathbf{R}_{i\mathbf{Q}}}{\omega^2 - \Omega^2_{i\mathbf{Q}}}. \tag{A17}$$

Note that because of the denominator in Eq. (A13), we have $\bar{\bar{\epsilon}}_{nn}(\omega_{\alpha\mathbf{Q}})^{-1} = 0$, which allows to obtain a very useful relation:

$$1 = \sum_i \frac{R^2_{i\mathbf{Q}n}}{\Omega^2_{i\mathbf{Q}n} - \omega^2_{\alpha\mathbf{Q}}}. \tag{A18}$$

In the base-independent version, we have

$$\sum_i \frac{\mathbf{R}_{i\mathbf{Q}} \otimes \mathbf{R}_{i\mathbf{Q}}}{\Omega^2_{i\mathbf{Q}} - \omega^2_{\alpha\mathbf{Q}}} = \bar{\bar{I}}. \tag{A19}$$

The last equation will allow to invert Eq. (A9). To complete the inversion, we write

$$\mathbf{H}_{\mathbf{Q}in} = Cst \times R_{i\mathbf{Q}n}\mathbf{n}. \tag{A20}$$

In order to fix the value of the unknown constant, we use the normalization condition:

$$\sum_\alpha \left(\left|m^i_{\alpha\mathbf{Q}}\right|^2 - \left|h^i_{\alpha\mathbf{Q}}\right|^2\right) = 1, \tag{A21}$$

which leads to

$$Cst = \frac{1}{\sqrt{2\hbar\Omega_{i\mathbf{Q}n}\varepsilon_0 V}}. \tag{A22}$$

We now multiply Eq. (A9) by $R_{in}\mathbf{n}/\sqrt{\Omega_{i\mathbf{Q}n}}$ and sum over i and $\mathbf{n}$. Then we use Eq. (A19) to obtain

$$\sum_i \frac{\mathbf{R}_{i\mathbf{Q}}}{2\sqrt{\Omega_{i\mathbf{Q}}}}(\Pi_{\mathbf{Q}i} + \Pi^\dagger_{-\mathbf{Q}i}) = \frac{1}{\sqrt{2\hbar\varepsilon_0 V}} \sum_\alpha \mathbf{d}_{\mathbf{Q}\alpha}(b_{\mathbf{Q}\alpha} + b^\dagger_{-\mathbf{Q}\alpha}), \tag{A23}$$

which leads to Eq. (45).

2. Light-matter interaction

At this point, the full Hamiltonian of the system is

$$\begin{aligned}\mathcal{H} = &\sum_{i,\mathbf{Q}} \hbar\Omega_{i\mathbf{Q}}\Pi^\dagger_{\mathbf{Q}i}\Pi_{\mathbf{Q}i} + \mathcal{H}_{\mathrm{ph}} \\ &- i\sum_{\mathbf{Q},\boldsymbol{\epsilon}_\mathbf{Q},i} \frac{\hbar(\mathbf{R}_{i\mathbf{Q}}\boldsymbol{\epsilon}_\mathbf{Q})}{2}\sqrt{\frac{\omega_{c\mathbf{Q}}}{\Omega_{i\mathbf{Q}}}}\left(a_{\mathbf{Q},\boldsymbol{\epsilon}_\mathbf{Q}} - a^\dagger_{-\mathbf{Q},\boldsymbol{\epsilon}_\mathbf{Q}}\right) \\ &\times (\Pi_{-\mathbf{Q}i} + \Pi^\dagger_{\mathbf{Q}i}).\end{aligned} \tag{A24}$$

To diagonalize it, we introduce the polariton operators:

$$\begin{aligned}P_\mathbf{Q} = &\sum_{\boldsymbol{\epsilon}_\mathbf{Q}} \left(x_{\mathbf{Q}\boldsymbol{\epsilon}_\mathbf{Q}} a_{\mathbf{Q},\boldsymbol{\epsilon}_\mathbf{Q}} + y_{\mathbf{Q}\boldsymbol{\epsilon}_\mathbf{Q}} a^\dagger_{-\mathbf{Q},\boldsymbol{\epsilon}_\mathbf{Q}}\right) \\ &+ \sum_i (m_{i\mathbf{Q}}\Pi_{\mathbf{Q}i} + h_{i\mathbf{Q}}\Pi^\dagger_{-\mathbf{Q}i}).\end{aligned} \tag{A25}$$

The polariton dispersion is then provided by the equation:

$$[P_\mathbf{Q}, \mathcal{H}] = \hbar\omega P_\mathbf{Q}. \tag{A26}$$

The computation of this commutator is straightforward and we shall skip the details. For the computation, it appears that it is useful to introduce the quantities:

$$W_{i\mathbf{Q}\boldsymbol{\epsilon}_\mathbf{Q}} = i\frac{(\mathbf{R}_{i\mathbf{Q}}\boldsymbol{\epsilon}_\mathbf{Q})}{2}\sqrt{\frac{\omega_{c\mathbf{Q}}}{\Omega_{i\mathbf{Q}}}}, \tag{A27}$$

$$H_{\mathbf{Q}\boldsymbol{\epsilon}_\mathbf{Q}} = \sum_i W_{i\mathbf{Q}\boldsymbol{\epsilon}_\mathbf{Q}}(h_{i\mathbf{Q}} - m_{i\mathbf{Q}}), \tag{A28}$$

$$X_{i\mathbf{Q}} = \sum_{\boldsymbol{\epsilon}_\mathbf{Q}} W_{i\mathbf{Q}\boldsymbol{\epsilon}_\mathbf{Q}}\left(x_{\mathbf{Q}\boldsymbol{\epsilon}_\mathbf{Q}} + y_{\mathbf{Q}\boldsymbol{\epsilon}_\mathbf{Q}}\right). \tag{A29}$$

Using the linear independence of the bosonic operators, we have the following expressions for the Hopfield coefficients:

$$x_{\mathbf{Q}\boldsymbol{\epsilon}_\mathbf{Q}} = H_{\mathbf{Q}\boldsymbol{\epsilon}_\mathbf{Q}}/(\omega - \omega_{c\mathbf{Q}}), \tag{A30}$$

$$y_{\mathbf{Q}\boldsymbol{\epsilon}_\mathbf{Q}} = H_{\mathbf{Q}\boldsymbol{\epsilon}_\mathbf{Q}}/(\omega + \omega_{c\mathbf{Q}}), \tag{A31}$$

$$m_{i\mathbf{Q}} = X_{i\mathbf{Q}}/(\omega - \Omega_{i\mathbf{Q}}), \tag{A32}$$

$$h_{i\mathbf{Q}} = X_{i\mathbf{Q}}/(\omega + \Omega_{i\mathbf{Q}}). \tag{A33}$$

Eliminating the "X" quantities in favour of the "H" quantities, we obtain the following equation:

$$H_{\mathbf{Q}\boldsymbol{\epsilon}_\mathbf{Q}}\frac{\omega^2 - \omega^2_{c\mathbf{Q}}}{\omega^2_{c\mathbf{Q}}} = \sum_{\boldsymbol{\epsilon}'_\mathbf{Q}} \frac{\boldsymbol{\epsilon}'_\mathbf{Q}(\mathbf{R}_{i\mathbf{Q}} \otimes \mathbf{R}_{i\mathbf{Q}})\boldsymbol{\epsilon}_\mathbf{Q}}{\omega^2 - \Omega^2_{i\mathbf{Q}}} H_{\mathbf{Q}\boldsymbol{\epsilon}'_\mathbf{Q}}. \tag{A34}$$

Introducing the transverse projector $\bar{\bar{I}}_\mathrm{t}$ from Eq. (46) and the transverse vector:

$$\mathbf{H}_{\mathbf{Q}\mathrm{t}} = \sum_{\boldsymbol{\epsilon}_\mathbf{Q}} H_{\mathbf{Q}\boldsymbol{\epsilon}_\mathbf{Q}}\boldsymbol{\epsilon}_\mathbf{Q}, \tag{A35}$$

we can recast Eq. (A34) in the form:

$$\left[\frac{\omega^2}{\omega^2_{c\mathbf{Q}}}\bar{\bar{I}}_\mathrm{t} - \bar{\bar{\epsilon}}(\omega,\mathbf{Q})^{-1}_\mathrm{t}\right]\mathbf{H}_{\mathbf{Q}\mathrm{t}} = \mathbf{0}, \tag{A36}$$

which is equivalent to Eq. (48). Using the fact that $\bar{\bar{I}}_\mathrm{t}$ is the unit tensor in the transverse subspace spanned by the two polarization vectors $\boldsymbol{\epsilon}_\mathbf{Q}$, we can rearrange Eq. (48) into

$$\left\|\frac{\omega^2}{c^2}\bar{\bar{\epsilon}}(\omega,\mathbf{Q})_{\mathrm{t}ij} - \delta_{ij}\mathbf{Q}^2 + Q_i Q_j\right\| = 0. \tag{A37}$$

This is exactly the equation for propagation of electromagnetic waves in a homogeneous and anisotropic medium as introduced by Landau and Lifshitz [38].

APPENDIX B: PLASMA HAMILTONIAN OF THE 3DEG

Let us consider the plasmon Hamiltonian from Eq. (95). We retain only the transverse collective modes $\Pi_{\mathbf{nQ}}$ that are coupled with light, and furthermore, we fix the polarization vector $\boldsymbol{\epsilon}_{\mathbf{Q}}$ with $\mathbf{n} = \boldsymbol{\epsilon}_{\mathbf{Q}}$. We obtain the following Hamiltonian:

$$\mathcal{H} = \hbar W_P \sum_{\mathbf{Q}} \Pi^{\dagger}_{\mathbf{Q}} \Pi_{\mathbf{Q}} + \sum_{\mathbf{Q}} \hbar\omega_{c\mathbf{Q}} (a^{\dagger}_{\mathbf{Q}} a_{\mathbf{Q}} + 1/2) - i \sum_{\mathbf{Q}} \frac{\sqrt{W_P \omega_{c\mathbf{Q}}}}{2} (a_{\mathbf{Q}} - a^{\dagger}_{-\mathbf{Q}})(\Pi^{\dagger}_{\mathbf{Q}} + \Pi_{-\mathbf{Q}}). \quad \text{(B1)}$$

We skip the fixed polarization index for simplicity. This quadratic Hamiltonian is readily diagonalized using Hopfield-Bogoliubov procedure, like the one described in Appendix A, by introducing the polariton operators:

$$Y_{\mathbf{Q}} = x_{\mathbf{Q}} a_{\mathbf{Q}} + y_{\mathbf{Q}} a^{\dagger}_{-\mathbf{Q}} + m_{\mathbf{Q}} \Pi_{\mathbf{Q}} + h_{\mathbf{Q}} \Pi^{\dagger}_{-\mathbf{Q}}. \quad \text{(B2)}$$

This problem has two eigenvalue solutions [Eq. (92)]:

$$\omega^{+}_{\mathbf{Q}} = \sqrt{W_P^2 + \omega_{c\mathbf{Q}}^2}, \qquad \omega^{-}_{\mathbf{Q}} = 0. \quad \text{(B3)}$$

The solution $\omega^{-}_{\mathbf{Q}}$ is unphysical and must be eliminated. The Hopfield coefficient for the physical solution can be readily obtained in a closed form:

$$x^{+}_{\mathbf{Q}} = i \sqrt{\frac{\omega_{c\mathbf{Q}}}{4(\omega^{+}_{\mathbf{Q}})^3}} (\omega^{+}_{\mathbf{Q}} + \omega_{c\mathbf{Q}}), \quad \text{(B4)}$$

$$y^{+}_{\mathbf{Q}} = i \sqrt{\frac{\omega_{c\mathbf{Q}}}{4(\omega^{+}_{\mathbf{Q}})^3}} (\omega^{+}_{\mathbf{Q}} - \omega_{c\mathbf{Q}}), \quad \text{(B5)}$$

$$m^{+}_{\mathbf{Q}} = \sqrt{\frac{W_P}{4(\omega^{+}_{\mathbf{Q}})^3}} (\omega^{+}_{\mathbf{Q}} + W_P), \quad \text{(B6)}$$

$$h^{+}_{\mathbf{Q}} = -\sqrt{\frac{W_P}{4(\omega^{+}_{\mathbf{Q}})^3}} (\omega^{+}_{\mathbf{Q}} - W_P). \quad \text{(B7)}$$

It is interesting to express the displacement field $\hat{\mathbf{D}}$ and the polarization field $\hat{\mathbf{P}}$ as a function of the plasmon-polariton operators. For this, we make use of the following relation that follows from Eq. (B2) and (B4)–(B7):

$$(Y_{+,\mathbf{Q}} + Y^{\dagger}_{+,-\mathbf{Q}}) \sqrt{(\omega^{+}_{\mathbf{Q}})^3} = i \sqrt{\omega^3_{c\mathbf{Q}}} (a_{\mathbf{Q}} - a^{\dagger}_{-\mathbf{Q}}) + \sqrt{W_P^3} (\Pi_{\mathbf{Q}} + \Pi^{\dagger}_{-\mathbf{Q}}). \quad \text{(B8)}$$

This relation is however not sufficient to express the field as a function of the polariton operators. We need an additional relation that can be obtained from the zero frequency solution. For this, we set $\omega^{-}_{\mathbf{Q}}$ to be a vanishing, but nonzero value: $\omega^{-}_{\mathbf{Q}} = \epsilon \to 0^{+}$. We then obtain the following asymptotic expressions for the Hopfield coefficients:

$$x^{-}_{\mathbf{Q}} = \frac{-i}{2\sqrt{\epsilon}} \sqrt{\frac{\omega^2_{c\mathbf{Q}} W_P}{(\omega^{+}_{\mathbf{Q}})^2}} (1 + \epsilon/\omega_{c\mathbf{Q}}), \quad \text{(B9)}$$

$$y^{-}_{\mathbf{Q}} = \frac{i}{2\sqrt{\epsilon}} \sqrt{\frac{\omega^2_{c\mathbf{Q}} W_P}{(\omega^{+}_{\mathbf{Q}})^2}} (1 - \epsilon/\omega_{c\mathbf{Q}}), \quad \text{(B10)}$$

$$m^{-}_{\mathbf{Q}} = \frac{1}{2\sqrt{\epsilon}} \sqrt{\frac{\omega^2_{c\mathbf{Q}} W_P}{(\omega^{+}_{\mathbf{Q}})^2}} (1 + \epsilon/W_P), \quad \text{(B11)}$$

$$h^{-}_{\mathbf{Q}} = \frac{1}{2\sqrt{\epsilon}} \sqrt{\frac{\omega^2_{c\mathbf{Q}} W_P}{(\omega^{+}_{\mathbf{Q}})^2}} (1 - \epsilon/W_P). \quad \text{(B12)}$$

These allow to obtain the following relation:

$$(Y_{-,\mathbf{Q}} + Y^{\dagger}_{-,-\mathbf{Q}}) \sqrt{(\omega^{+}_{\mathbf{Q}})^2 \epsilon} = -i \sqrt{\omega^2_{c\mathbf{Q}} W_P} (a_{\mathbf{Q}} - a^{\dagger}_{-\mathbf{Q}}) + \sqrt{W_P^2 \omega_{c\mathbf{Q}}} (\Pi_{\mathbf{Q}} + \Pi^{\dagger}_{-\mathbf{Q}}). \quad \text{(B13)}$$

Inverting Eqs. (B8) and (B13) to express the old photon and polarization operators as a function of the new polariton operators, and taking the limit $\epsilon \to 0^{+}$, we obtain the following expressions for the displacement and polarization fields:

$$\hat{\mathbf{P}}(\mathbf{r}) = \hat{\mathbf{D}}(\mathbf{r}) = \sqrt{\frac{\varepsilon_0 \hbar (\omega^{+}_{\mathbf{Q}})^3}{2V}} \frac{\boldsymbol{\epsilon}_{\mathbf{Q}} e^{-i\mathbf{Q}\mathbf{r}}}{\omega_{c\mathbf{Q}} + W_P} \sum_{\mathbf{Qn}} (Y_{+,\mathbf{Q}} + Y^{\dagger}_{+,-\mathbf{Q}}). \quad \text{(B14)}$$

The perfect equality between the two fields stems from the presence of a zero frequency solution. Its physical meaning is that the electric field of the electromagnetic wave that propagates in the electron gas is perfectly screened by the field created by the oscillating charges. In particular, it can be shown that an oscillating dipole can not radiate such bulk plasmon-polaritons, as the dipole-field coupling term (matter-radiation interaction) and the dipole-polarization coupling (Coulomb interaction) term cancel each other. The situation is different for surface-plasmon polaritons, where $\hat{\mathbf{D}}(\mathbf{r})$ and $\hat{\mathbf{P}}(\mathbf{r})$ do not overlap perfectly in space, as $\hat{\mathbf{D}}(\mathbf{r})$ leaks partially to the medium 1 (Fig. 3). In that case, the spontaneous emission process becomes allowed.

APPENDIX C: HOPFIELD COEFFICIENTS FOR THE SPP

Using the Hopfield matrix for the SPP from Eq. (122), we obtain the following relations between the Hopfield coefficients introduced in Eq. (121):

$$m_{\mathbf{q}} = \frac{\Omega_{\mathbf{q}} C_{\mathbf{q}}}{W_P - \omega} z_{\mathbf{q}}, \quad \text{(C1)}$$

$$h_{\mathbf{q}} = -\frac{\Omega_{\mathbf{q}} C_{\mathbf{q}}}{W_P + \omega} z_{\mathbf{q}}, \quad \text{(C2)}$$

$$t_{\mathbf{q}} = i z_{\mathbf{q}}. \quad \text{(C3)}$$

The condition $[Y_{\mathbf{q}}, Y^{\dagger}_{\mathbf{q}'}] = \delta_{\mathbf{q},\mathbf{q}'}$ delivers the following link between the Hopfield coefficients:

$$|m_{\mathbf{q}}|^2 - |h_{\mathbf{q}}|^2 + 2|t_{\mathbf{q}}|^2 C_{\mathbf{q}} = 1. \tag{C4}$$

These equations allow to determine all the quantities introduced above up to phase factor. In particular, Eq. (C4) can be rewritten as

$$2C_{\mathbf{q}}|t_{\mathbf{q}}|^2 \left[1 + \frac{W_P^2}{\left(W_P^2 - \omega_{\mathbf{q}}^2\right)} \frac{\left(q^2 + \gamma_2^2\right)c^2}{\varepsilon_2(1 + \gamma_1/\gamma_2)}\right] = 1. \tag{C5}$$

Using this relation, together with the SPP dispersion relation from Eq. (126), we obtain the expression for the spontaneous emission rate in Eq. (134).

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