Equilibrium long-ranged charge correlations at the interface between media coupled to the electromagnetic radiation

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We continue studying long-ranged quantum correlations of surface charge densities on the interface between two media of distinct dielectric functions which are in thermal equilibrium with the radiated electromagnetic field. Two regimes are considered: the nonretarded one with the speed of light c taken to be infinitely large and the retarded one with a finite value of c. The analysis is based on our results obtained by using fluctuational electrodynamics [L. Šamaj and B. Jancovici, Phys. Rev. E **78**, 051119 (2008)]. Using an integration method in the complex plane and the general analytical properties of dielectric functions in the frequency upper half plane, we derive explicit forms of prefactors to the long-range decay of the surface charge correlation functions for all possible media (conductor, dielectric, and vacuum) configurations. The main result is that the time-dependent quantum prefactor in the retarded regime takes its static classical form for any temperature.

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I. INTRODUCTION

In this paper, we continue studying long-ranged quantum correlations of surface charge densities on the interface between two distinct media, initiated in Refs. [1,2]. The model, formulated in the three-dimensional Cartesian space of coordinates (x,y,z), is inhomogeneous say along the first coordinate x (see Fig. 1). The two semi-infinite media with the frequency-dependent dielectric functions $\epsilon_1(\omega)$ and $\epsilon_2(\omega)$ are localized in the half spaces $x>0$ and $x<0$, respectively. Since there are difficulties in defining the frequency-dependent magnetic permeability $\mu(\omega)$ [3], we restrict ourselves to the case $\mu=1$ in both media. The interface is the plane $x=0$; a point on the interface is $\mathbf{R}=(0,y,z)$. The different electric properties of the media give rise to a surface charge density which must be understood as being the microscopic volume charge density integrated on some microscopic depth. It is related to the discontinuity of the x component of the electric field on the interface. Denoting by $\sigma(t,\mathbf{R})$ the surface charge density at time t and at a point $\mathbf{R}$, the (symmetrized) two-point correlation function, at times different by t, reads

$$S(t,\mathbf{R}) \equiv \tfrac{1}{2}\langle \sigma(t,\mathbf{R})\sigma(0,\mathbf{0}) + \sigma(0,\mathbf{0})\sigma(t,\mathbf{R})\rangle^{\mathrm{T}}, \quad (1)$$

where $\langle\cdots\rangle^{\mathrm{T}}$ represents a truncated statistical average at the inverse temperature β (we exclude the case of zero temperature, $\beta\to\infty$). We are interested in the behavior of the correlation function (1) at distances on the interface $R=|\mathbf{R}|$ large compared to the microscopic length scales (like the particle correlation function). The static case of zero time difference $t=0$ between the two distinct points is simpler than the one with $t\neq 0$ and so the two cases are treated separately.

The two media configuration studied so far was restricted to a conductor, localized say in the half space $x>0$ with the dielectric function $\epsilon_1(\omega)\equiv\epsilon(\omega)$, in contact with vacuum of the dielectric constant $\epsilon_2(\omega)=1$ (the vacuum part of the space is equivalent to a hard wall impenetrable to charged particles forming the conductor). In some theoretical studies, the dielectric function is approximated by a simple one-resonance Drude formula [4]

$$\epsilon(\omega) = 1 + \frac{\omega_p^2}{\omega_0^2 - \omega(\omega + i\eta)}, \quad (2)$$

where ω_p is the plasma frequency, η is the dissipation constant, and ω_0 is the oscillation frequency of harmonically bound charges: $\omega_0=0$ for conductors and $\omega_0\neq 0$ for dielectrics. In [1,2], we applied Eq. (2) to the jellium model (sometimes called the one-component plasma), i.e., a system of pointlike particles of charge e, mass m, and bulk number density n, immersed in a uniform neutralizing background of charge density $-en$. The dynamical properties of the jellium have a special feature: there is no viscous damping of the long-wavelength plasma oscillations for identically charged particles, so that $\eta\to 0^+$ in Eq. (2). The frequencies of nonretarded nondispersive long-wavelength collective modes, namely, ω_p of the bulk plasmons and ω_s of the surface plasmons, are given by

$$\omega_p = \left(\frac{4\pi n e^2}{m}\right)^{1/2}, \quad \omega_s = \frac{\omega_p}{\sqrt{2}}. \quad (3)$$

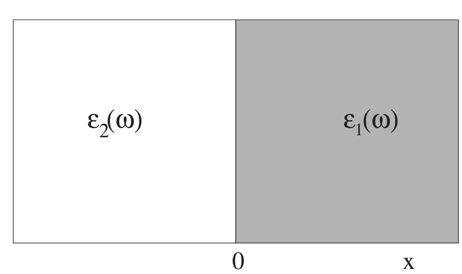

FIG. 1. Two semi-infinite media characterized by dielectric functions $\epsilon_1(\omega)$ and $\epsilon_2(\omega)$.

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For other conductors and dielectrics, we shall use only the general properties of $\epsilon(\omega)$, without restricting ourselves to the Drude model (2).

The problem of a conductor in contact with vacuum was studied in the past, with an increasing level of physical complexity and their range of validity. The problem can be treated as classical or quantum, in the nonretarded or retarded regime. In all cases, the asymptotic large-distance behavior of the surface charge correlation function (1) exhibits a long-ranged tail of type

$$\beta S(t,\mathbf{R}) \sim \frac{h(t)}{R^3}, \quad R \to \infty, \tag{4}$$

where the form of the prefactor function $h(t)$ depends on the model used. It is useful to introduce the Fourier transform

$$S(t,\mathbf{q}) = \int d^2R \exp(i\mathbf{q}\cdot\mathbf{R}) S(t,\mathbf{R}), \tag{5}$$

with $\mathbf{q}=(q_y,q_z)$ being a two-dimensional (2D) wave vector. Since, in the sense of distributions, the 2D Fourier transform of $1/R^3$ is $-2\pi q$, a result equivalent to Eq. (4) is that $\beta S(t,\mathbf{q})$ has a kink singularity at $\mathbf{q}=\mathbf{0}$, behaving like

$$\beta S(t,q) \sim -2\pi h(t) q, \quad q \to 0. \tag{6}$$

(i) *Classical nonretarded regime.* Let the conductor be modeled by a classical Coulomb fluid composed of charged particles with the instantaneous Coulomb interactions. By a microscopic analysis [5], the long-range decay of the static surface correlation function was found such that

$$h_{\rm cl}^{(\rm nr)}(0) = -\frac{1}{8\pi^2}, \tag{7}$$

where the subscript "cl" means "classical" and the superscript "nr" means "nonretarded" (i.e., considered without relativistic effects associated with the finiteness of the speed of light c). The same result has been obtained later [6] by simple macroscopic arguments based on a combination of the linear-response theory and the electrostatic method of images. Note the universal form of $h_{\rm cl}(0)$, independent of the composition of the Coulomb fluid.

(ii) *Quantum nonretarded regime.* The extension of the static result (7) to a quantum Coulomb fluid, modeled by the jellium, was accomplished in Ref. [7]. The absence of damping was crucial in the treatment using long-wavelength collective modes, the bulk, and surface plasmons with frequencies given in Eq. (3). The Maxwell equations, obeyed by the plasmons, were considered in the nonretarded (nonrelativistic) regime with the speed of light c taken to be infinitely large, $c=\infty$, ignoring in this way magnetic forces acting on the charged particles. The obtained time-dependent result has the nonuniversal form [7–9]

$$h_{\rm qu}^{(\rm nr)}(t) = -\frac{1}{8\pi^2}[2g(\omega_s)\cos(\omega_s t) - g(\omega_p)\cos(\omega_p t)], \tag{8a}$$

$$g(\omega) = \frac{\beta\hbar\omega}{2}\coth\left(\frac{\beta\hbar\omega}{2}\right), \tag{8b}$$

where the subscript "qu" means "quantum." According to the correspondence principle, a quantum system admits the classical statistical description in the high-temperature limit $\beta\hbar\to 0$. In this limit, the function $g(\omega)=1$ for any ω and the quantum formula (8a) reduces to the classical nonretarded one

$$h_{\rm cl}^{(\rm nr)}(t) = -\frac{1}{8\pi^2}[2\cos(\omega_s t) - \cos(\omega_p t)]. \tag{9}$$

For $t=0$, we recover the classical static formula (7).

(iii) *Quantum retarded regime.* In the previous paper [1], we studied the surface charge correlations taking into account retardation (c is assumed finite) and the quantum nature of both the jellium and the radiated electromagnetic (EM) field, which are in thermal equilibrium. In other words, the quantum particles are fully coupled to both electric and magnetic parts of the radiated EM field. By using Rytov's fluctuational electrodynamics [3,10,15], we showed that there are two regions of distances R on the interface: the intermediate one given by the inequalities $\lambda_{\rm ph} \ll R \ll c/\omega_p$ ($\lambda_{\rm ph} \propto \beta\hbar c$ stands for the thermal de Broglie wavelength of photon and c/ω_p is the wavelength of electromagnetic waves emitted by charge oscillations at frequency ω_p), where the nonretarded results (8a) and (8b) apply, and the strictly asymptotic one given by the inequality $c/\omega_p \ll R$, where a retarded result applies. After long calculations in [1], a bit shortened through the alternative method of [2], a very simple form of the retarded result was found,

$$h_{\rm qu}^{(\rm r)}(t) = -\frac{1}{8\pi^2}. \tag{10}$$

Here, the superscript "r" means "retarded." We see that, for any temperature and time t, the inclusion of retardation effects causes the prefactor function $h(t)$ to take its universal static classical form (7), independent of $\hbar$ and c. Formula (10) does not change in the classical limit $\beta\hbar\to 0$, i.e.,

$$h_{\rm cl}^{(\rm r)}(t) = -\frac{1}{8\pi^2}. \tag{11}$$

The presence or the absence of magnetic fields is important in two-point classical statistical averages, taken at two different times [11]. Therefore, it is not surprising that the classical retarded formula (11) and the classical nonretarded one (9) do not coincide with one another for nonzero time differences, $h_{\rm cl}^{(\rm r)}(t) \neq h_{\rm cl}^{(\rm nr)}(t)$ for $t\neq 0$. On the other hand, for $t=0$ we have

$$h_{\rm cl}^{(\rm r)}(0) = h_{\rm cl}^{(\rm nr)}(0). \tag{12}$$

This equality is in agreement with the Bohr-van Leeuwen theorem [12,13] about an effective elimination of magnetic degrees of freedom from statistical averages (with zero time differences among the fixed points in the coordinate part of the configuration space) of classical systems; for a detailed treatment of this subject, see Ref. [14].

In the previous papers [1,2], we used the fact, special to the jellium model, that there is no damping for small wave numbers. The question whether the crucial formula (10) is still valid for a conductor with dissipation was left as an open problem. There are still many other unsolved physical situations that deserve attention. What happens in the case of a general dielectric in contact with vacuum? Filling the vacuum region by a material medium, other types of contacts are possible, such as conductor-conductor, dielectric-dielectric, and conductor-dielectric. Another kind of problem is the algebraic complicacy connected with the derivation of result (10) for the jellium with the relatively simple form of the dielectric function. Does there exist a simple method for evaluating the large-distance asymptotics of the surface charge density correlation function that is applicable to the jellium as well as to other more complicated systems? All questions asked are answered in the present paper.

The generalization of the formalism to contacts between all kinds of materials, defined by their dielectric functions, has already been done in Ref. [1]. The true problem is the mathematical handling of the final (nonretarded or retarded) formula for the Fourier transform $\beta S(t,q)$, written as an integral over real frequencies, to deduce the small-q behavior (6). Here, we accomplish the task first by extending the integration over real frequencies to a contour integration in the complex frequency plane and then using integration techniques in the complex plane together with the known analytical properties of dielectric functions in the frequency upper half plane. In short, the time-dependent retarded results maintain the simplicity of the jellium formula (10) and involve only dielectric functions of media in contact at zero frequency. The nonretarded results are complicated and available only, in general, as infinite series over Matsubara frequencies.

The paper is organized as follows. Section II is a generalization of the classical static result (7) for conductor to a dielectric. The result will serve us as a check of more general calculations. Section III summarizes briefly the general analytical properties of dielectric functions in the complex frequency upper half plane, which are necessary for the derivation of our basic results. Section IV applies the present method for the calculation of $\beta S(0,\mathbf{q})$. Section V does the same for $\beta S(t,\mathbf{q})$. Section VI is a conclusion.

II. STATIC CORRELATIONS FOR CLASSICAL DIELECTRICS

It has been known for a long time [5] that the classical surface charge correlations on a conductor, made of particles interacting through the Coulomb law and bounded by a plane wall, are long ranged. We prefer to use the macroscopic language [6]. If $\mathbf{R}$ and $\mathbf{R}'$ are two points on the wall, the classical correlation, for distances $|\mathbf{R}-\mathbf{R}'|$ large compared to the microscopic scale, behaves like

$$\beta S(\mathbf{R}-\mathbf{R}') \equiv \beta\langle\sigma(\mathbf{R})\sigma(\mathbf{R}')\rangle \sim -\frac{1}{8\pi^2|\mathbf{R}-\mathbf{R}'|^3} \tag{13}$$

[we assume that the conductor is uncharged, $\langle\sigma(\mathbf{R})\rangle=0$].

A generalization of Eq. (13) for the case of a dielectric of static dielectric constant $\epsilon(0)=\epsilon_0$ bounded by a plane can be found by the same method that has been used in [6]. $\sigma(\mathbf{R})$ is related to the discontinuity of the x component of the electric field on the wall. We call $E_x^{\text{out(in)}}(\mathbf{R})$ the limit of that field component as $\mathbf{R}$ is zero from the outside (inside) of the dielectric. The correlation of the surface charge densities is

$$\langle\sigma(\mathbf{R})\sigma(\mathbf{R}')\rangle = \frac{1}{(4\pi)^2}\langle[E_x^{\text{in}}(\mathbf{R})-E_x^{\text{out}}(\mathbf{R})][E_x^{\text{in}}(\mathbf{R}') - E_x^{\text{out}}(\mathbf{R}')]\rangle. \tag{14}$$

If a test infinitesimal charge is introduced at $\mathbf{r}=(x>0,0,0)$ in the dielectric, the electric potential *created by the dielectric* at a point $\mathbf{r}'=(x'>0,y',z')$ in the dielectric is given by the method of images as [4]

$$\langle\phi(\mathbf{r}')\rangle_q = \frac{1}{\epsilon_0}\left[\frac{q}{|\mathbf{r}'-\mathbf{r}|} - \frac{q(1-\epsilon_0)}{(1+\epsilon_0)|\mathbf{r}'-\mathbf{r}^*|}\right] - \frac{q}{|\mathbf{r}'-\mathbf{r}|}, \tag{15}$$

where $\mathbf{r}^*=(-x,0,0)$ is the image of $\mathbf{r}$. The last term in Eq. (15) is the potential created by q, which should not be included in the potential created by the dielectric. The linear-response theory relates response (15) to the unperturbed correlation function between the additional Hamiltonian $q\phi(\mathbf{r})$ and $\phi(\mathbf{r}')$, giving

$$\langle\phi(\mathbf{r}')\rangle_q = -\beta q\langle\phi(\mathbf{r})\phi(\mathbf{r}')\rangle. \tag{16}$$

Since the x component of the electric field is $E_x(\mathbf{r}) = -(\partial/\partial x)\phi(\mathbf{r})$, using Eqs. (15) and (16) we can obtain the correlation of the x component of the electrical fields inside the dielectric at the wall as

$$\beta\langle E_x^{\text{in}}(\mathbf{R})E_x^{\text{in}}(\mathbf{R}')\rangle = \left(\frac{2}{1+\epsilon_0} - \frac{2}{\epsilon_0} + 1\right)\frac{1}{|\mathbf{R}-\mathbf{R}'|^3}. \tag{17}$$

Similar calculations give the correlation outside the dielectric

$$\beta\langle E_x^{\text{out}}(\mathbf{R})E_x^{\text{out}}(\mathbf{R}')\rangle = \frac{1-\epsilon_0}{1+\epsilon_0}\frac{1}{|\mathbf{R}-\mathbf{R}'|^3} \tag{18}$$

and the cross correlation

$$\beta\langle E_x^{\text{in}}(\mathbf{R})E_x^{\text{out}}(\mathbf{R}')\rangle = -\frac{1-\epsilon_0}{1+\epsilon_0}\frac{1}{|\mathbf{R}-\mathbf{R}'|^3}. \tag{19}$$

Using Eqs. (17)–(19) in Eq. (14), we obtain

$$\beta\langle\sigma(\mathbf{R})\sigma(\mathbf{R}')\rangle \sim -\frac{1}{8\pi^2}\left(\frac{1}{\epsilon_0} + 1 - \frac{4}{1+\epsilon_0}\right)\frac{1}{|\mathbf{R}-\mathbf{R}'|^3}, \tag{20}$$

which is the wanted generalization for a dielectric. With regard to definition (4), we have the nonuniversal result

$$h_{\text{cl}}(0) = -\frac{1}{8\pi^2}\left(\frac{1}{\epsilon_0} + 1 - \frac{4}{1+\epsilon_0}\right). \tag{21}$$

The value of $\epsilon(0)$ is infinite for any kind of conductor and we retrieve Eq. (13) from Eq. (20), or Eq. (7) from Eq. (21). This fact explains the universality of the static h_{cl} for conductors.

III. ANALYTICAL PROPERTIES OF DIELECTRIC FUNCTIONS

Analytical properties of dielectric functions are described in many textbooks [3,4,15]. They apply to an arbitrary dielectric function of real materials, including the idealized Drude formula (2). We shall mention only those properties that are important in the derivation of our basic results; the proofs of theorems are given in the above textbooks. The vacuum case $\epsilon(\omega)=1$ is excluded from the discussion.

Due to the causal relation between the displacement **D** and the electric field **E**, the dielectric function of every medium can be expressed as

$$\epsilon(\omega) = 1 + \int_0^\infty d\tau\, e^{i\omega\tau} G(\tau), \tag{22}$$

where the function $G(\tau)$ is finite for all values of τ, including zero. In particular, $G(\tau)$ tends to zero as $\tau\to\infty$ for dielectrics and it tends to $4\pi\sigma$ (σ is the conductivity) as $\tau\to\infty$ for conductors. Relation (22) has several important consequences.

Let us first consider the frequency ω to be purely real. It follows from Eq. (22) that $\epsilon^*(\omega)=\epsilon(-\omega)$. Denoting $\epsilon(\omega)=\epsilon'(\omega)+i\epsilon''(\omega)$, where both the real $\epsilon'(\omega)$ and the imaginary $\epsilon''(\omega)$ parts are real numbers, we thus have

$$\epsilon'(\omega) = \epsilon'(-\omega), \quad \epsilon''(\omega) = -\epsilon''(-\omega). \tag{23}$$

For any real material medium with absorption, it holds

$$\epsilon''(\omega) > 0(<0) \quad \text{for} \quad \omega > 0(<0). \tag{24}$$

The sign of $\epsilon'(\omega)$ is not subjected to any physical restriction.

If ω is complex, $\omega=\omega'+i\omega''$, representation (22) tells us that $\epsilon(\omega)$ is an analytical function of ω in the upper half plane $\omega''>0$. Apart from a possible pole at $\omega=0$ (for conductors), the analyticity extends also to the real ω axis. $\epsilon(\omega)$ has no zeros in the upper half plane. We notice that the analyticity properties of $\epsilon(\omega)$ can also be derived from the Kramers-Kronig relations combined to the positivity of $\epsilon''(\omega)$ for $\omega>0$.

The function $\epsilon(\omega)$ does not take real values at any finite point in the upper half plane, except on the imaginary axis. We can deduce from Eq. (22) that for any complex ω it holds $\epsilon^*(\omega)=\epsilon(-\omega^*)$. For purely imaginary $\omega=i\omega''$, indeed we find

$$\epsilon(i\omega'') = \epsilon^*(i\omega'') \Rightarrow \text{Im}\ \epsilon(\omega) = 0 \quad \text{for} \quad \omega = i\omega''. \tag{25}$$

Moreover, on the imaginary axis, $\epsilon(\omega)$ decreases monotonically from $\epsilon_0>1$ (for dielectrics) or from ∞ (for conductors) at $\omega=i0$ to 1 at $\omega=i\infty$.

With regard to symmetries (23) for a real ω, the expansion of the dielectric function around the origin $\omega=0$ reads, for a conductor with conductivity $\sigma>0$,

$$\epsilon(\omega) = \frac{4\pi\sigma i}{\omega} + a + O(\omega), \tag{26}$$

where the sign of the constant a is not restricted; for a dielectric medium of static dielectric constant ϵ_0,

$$\epsilon(\omega) = \epsilon_0 + ia\omega + O(\omega^2), \quad a > 0, \tag{27}$$

where the positive sign of the constant a is fixed by the physical requirements (24).

In the limit $|\omega|\to\infty$, a Taylor series expansion of $G(\tau)$ around $\tau=0^+$ in Eq. (22) implies that, for both conductors and dielectrics,

$$\text{Re}[\epsilon(\omega) - 1] = O\left(\frac{1}{\omega^2}\right), \tag{28a}$$

$$\text{Im}\ \epsilon(\omega) = O\left(\frac{1}{\omega^3}\right). \tag{28b}$$

IV. STATIC SURFACE CHARGE CORRELATIONS

In the general case of the plane contact between two media of dielectric functions $\epsilon_1(\omega)$ and $\epsilon_2(\omega)$ pictured in Fig. 1, the quantum formula for the Fourier transform of the surface charge correlation function (5) in the long-wavelength limit $q\to 0$ was derived in Ref. [1]. Its static $t=0$ version can be re-expressed as

$$\beta S_{\text{qu}}(0,q) = \int_{-\infty}^{\infty} \frac{d\omega}{\omega} \text{Im}\ f(\omega), \tag{29}$$

where the form of the function $f(\omega)$ depends on the considered (retarded or nonretarded) regime. In the retarded case, $f(\omega)\equiv f_{\text{qu}}^{(\text{r})}(\omega)$ is given by two equivalent representations

$$\begin{aligned} f_{\text{qu}}^{(\text{r})}(\omega) &= \frac{q^2}{4\pi^2} g(\omega) \frac{1}{\kappa_1(\omega)\epsilon_2(\omega) + \kappa_2(\omega)\epsilon_1(\omega)} \frac{[\epsilon_1(\omega) - \epsilon_2(\omega)]^2}{\epsilon_1(\omega)\epsilon_2(\omega)} \\ &= \frac{q^2}{4\pi^2} g(\omega) \frac{\kappa_1(\omega)\epsilon_2(\omega) - \kappa_2(\omega)\epsilon_1(\omega)}{q^2[\epsilon_1(\omega) + \epsilon_2(\omega)] - \omega^2\epsilon_1(\omega)\epsilon_2(\omega)/c^2} \\ &\quad \times \left(\frac{1}{\epsilon_1(\omega)} - \frac{1}{\epsilon_2(\omega)}\right), \end{aligned} \tag{30}$$

where $g(\omega)$ is defined in Eq. (8b) and the (complex) inverse lengths κ_1 and κ_2, one for each of the half-space regions, are given by

$$\kappa_{1,2}^2(\omega) = q^2 - \frac{\omega^2}{c^2}\epsilon_{1,2}(\omega), \quad \text{Re}\ \kappa_{1,2}(\omega) > 0. \tag{31}$$

In the nonretarded case, $f(\omega)\equiv f_{\text{qu}}^{(\text{nr})}(\omega)$ is obtained from Eq. (30) by setting the speed of light $c\to\infty$. Since $\kappa_1=\kappa_2=q$ in this limit, we get

$$f_{\text{qu}}^{(\text{nr})}(\omega) = \frac{q}{4\pi^2} g(\omega) \left(\frac{1}{\epsilon_1(\omega)} + \frac{1}{\epsilon_2(\omega)} - \frac{4}{\epsilon_1(\omega) + \epsilon_2(\omega)}\right). \tag{32}$$

The derivation procedures outlined below are applicable to both retarded and nonretarded regimes and so, whenever possible, we use the simplified notation $f(\omega)$ to cover both functions $f_{\text{qu}}^{(\text{r})}(\omega)$ and $f_{\text{qu}}^{(\text{nr})}(\omega)$.

For a real frequency ω, the symmetry relation $\epsilon^*(\omega)=\epsilon(-\omega)$ implies that $\kappa^*(\omega)=\kappa(-\omega)$. Consequently, $f^*(\omega)=f(-\omega)$, i.e.,

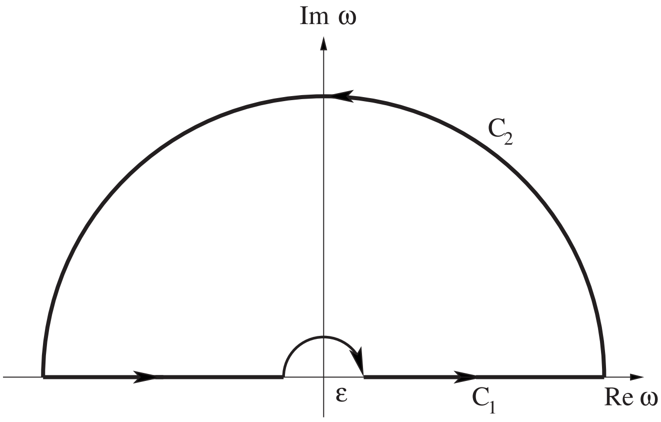

FIG. 2. The contour in the complex frequency plane for $t=0$.

$$\mathrm{Re}\, f(\omega) = \mathrm{Re}\, f(-\omega), \quad \mathrm{Im}\, f(\omega) = -\mathrm{Im}\, f(-\omega). \tag{33}$$

As $\omega \to 0$,

$$\mathrm{Im}\, f(0) = 0, \tag{34a}$$

$$\mathrm{Re}\, f(0) = \frac{q}{4\pi^2}\left(\frac{1}{\epsilon_1(0)} + \frac{1}{\epsilon_2(0)} - \frac{4}{\epsilon_1(0)+\epsilon_2(0)}\right). \tag{34b}$$

If ω is complex and $|\omega| \to \infty$, with the aid of the asymptotic relations (28a) and (28b) we find that

$$\lim_{|\omega|\to\infty} f(\omega) = 0. \tag{35}$$

It is complicated to calculate the correlation function directly from formula (29) by expressing the imaginary part of $f(\omega)$, then integrating over ω and finally taking the $q \to 0$ limit. We shall find the value of the integral of interest in another way, by using integration techniques in the complex plane and the analytical properties of dielectric functions, summarized in the previous section.

We can change slightly the path of integration, writing

$$\int_{C_1} \frac{d\omega}{\omega} f(\omega) = -i\pi f(0) + \mathcal{P}\int_{-\infty}^{\infty} \frac{d\omega}{\omega} f(\omega), \tag{36}$$

where C_1 is the path following the real axis, except it goes around the origin $\omega=0$ in a small semicircle in complex upper half plane whose radius ε tends to zero (Fig. 2). The first term on the right-hand side (rhs) of Eq. (36) is the contribution of the negatively oriented semicircle around the origin; $\mathcal{P}$ denotes the Cauchy principal value avoiding the origin,

$$\mathcal{P}\int_{-\infty}^{\infty} \frac{d\omega}{\omega} f(\omega) \equiv \lim_{\varepsilon\to 0}\left[\int_{-\infty}^{-\varepsilon} \frac{d\omega}{\omega} f(\omega) + \int_{\varepsilon}^{\infty} \frac{d\omega}{\omega} f(\omega)\right]. \tag{37}$$

It is easy to see from the symmetry relations (33) and equality (34a) that

$$\mathcal{P}\int_{-\infty}^{\infty} \frac{d\omega}{\omega} f(\omega) = i\int_{-\infty}^{\infty} \frac{d\omega}{\omega} \mathrm{Im}\, f(\omega). \tag{38}$$

We can close the path C_1 by a semicircle at infinity C_2 (see Fig. 2) along which the integral $\int_{C_2} d\omega\, f(\omega)/\omega$ is zero because of the asymptotic relation (35). Denoting the closed contour as C ($C = C_1 \cup C_2$) and applying the operation Im to both sides of relation (36), we arrive at

$$\int_{-\infty}^{\infty} \frac{d\omega}{\omega} \mathrm{Im}\, f(\omega) = \pi f(0) + \mathrm{Im} \oint_C \frac{d\omega}{\omega} f(\omega). \tag{39}$$

The integral over the contour C can be evaluated by using the residue theorem at poles $\{\omega_j\}$ of the function $f(\omega)$ in the ω upper half plane bounded by C,

$$\mathrm{Im} \oint_C \frac{d\omega}{\omega} f(\omega) = 2\pi \sum_j \frac{\mathrm{Res}(f,\omega_j)}{\omega_j}, \tag{40}$$

provided that $\mathrm{Res}(f,\omega_j)/\omega_j$ is real (which will be the case); Res denotes the residue. The static correlation function (29) is expressible as

$$\beta S_{\mathrm{qu}}(0,q) = \pi f(0) + 2\pi \sum_j \frac{\mathrm{Res}(f,\omega_j)}{\omega_j}, \tag{41}$$

where the value of $f(0)$ is real, given by Eq. (34b). The original algebraic task thus reduces to the problem of searching for all poles of the function $f(\omega)$ in the ω upper half plane.

Both the retarded (30) and the nonretarded (32) versions of the f function contain $g(\omega)$ defined in Eq. (8b). Since $g(\omega)$ can be expanded in ω as follows [16]:

$$g(\omega) = 1 + \sum_{j=1}^{\infty} \frac{2\omega^2}{\omega^2 + \xi_j^2}, \quad \xi_j = \frac{2\pi}{\beta\hbar} j, \tag{42}$$

it has in the upper half plane an infinite sequence of simple poles at the imaginary Matsubara frequencies

$$\omega_j = i\xi_j, \quad \mathrm{Res}(g,\omega_j) = \omega_j \quad (j = 1,2,\ldots). \tag{43}$$

The nonretarded f function (32) has no further poles in the upper half plane since the dielectric functions $\epsilon_1(\omega)$ and $\epsilon_2(\omega)$ do not take there real values at any finite point. The retarded f function (30) might have some further poles at points ω satisfying the equation

$$\omega^2 = (cq)^2\left(\frac{1}{\epsilon_1(\omega)} + \frac{1}{\epsilon_2(\omega)}\right). \tag{44}$$

Interestingly, in the case of the jellium in vacuum (with real dielectric functions), this is just the dispersion relation for the surface plasmons (polaritons); for a recent review, see [17]. We are interested in the long-wavelength limit $q \to 0$. If $q=0$, the only solution of Eq. (44) is $\omega=0$; this point is not inside the contour C due to the presence of the semicircle around $\omega=0$. Now, let us study how the solutions of Eq. (44) "glue off" from $\omega=0$ when q is infinitesimal, but not identically equal to zero. We need the small-ω expansion of $1/\epsilon(\omega)$. It follows from expansions (26) and (27) that for both conductors and dielectrics we can write

$$\frac{1}{\epsilon(\omega)} = \frac{1}{\epsilon(0)} - ib\omega + O(\omega^2), \quad b \geq 0, \tag{45}$$

where the material constant b, $b=1/(4\pi\sigma)$ for conductors and $b=a/\epsilon_0^2$ for dielectrics, is always positive, except for vacuum when $b=0$. For small q, Eq. (44) thus exhibits two solutions:

$$\omega_\pm \sim \pm cq\left(\frac{1}{\epsilon_1(0)} + \frac{1}{\epsilon_2(0)}\right)^{1/2} - i\frac{(cq)^2}{2}(b_1+b_2). \tag{46}$$

Since $b_1+b_2>0$, the two poles $\omega_\pm$ move, as q increases from zero to a small positive number, from $\omega=0$ to the lower ω half plane, i.e., outside of the region enclosed by the C contour. We conclude that, in both retarded and nonretarded regimes, only the poles on the imaginary axis at the Matsubara frequencies (43) contribute to the static correlation function (41), which thus becomes expressible as follows:

$$\beta S_{\rm qu}(0,q) = \frac{q}{4\pi}\left(\frac{1}{\epsilon_1(0)} + \frac{1}{\epsilon_2(0)} - \frac{4}{\epsilon_1(0)+\epsilon_2(0)}\right) + F(0,q), \tag{47a}$$

$$F(0,q) = 2\pi\sum_{j=1}^{\infty}\frac{{\rm Res}(f,i\xi_j)}{i\xi_j}. \tag{47b}$$

The first term on the rhs of Eq. (47a) is independent of $\beta\hbar$ and c; the explicit form of the (static) function $F(0,q)$ depends on the considered (retarded or nonretarded) regime.

A. Retarded regime

In the retarded case (30), we have

$$F_{\rm qu}^{(\rm r)}(0,q) = \frac{q^2}{2\pi}\sum_{j=1}^{\infty}\frac{1}{\kappa_1(i\xi_j)\epsilon_2(i\xi_j)+\kappa_2(i\xi_j)\epsilon_1(i\xi_j)} \times\frac{[\epsilon_1(i\xi_j)-\epsilon_2(i\xi_j)]^2}{\epsilon_1(i\xi_j)\epsilon_2(i\xi_j)}. \tag{48}$$

We recall from Sec. III that the values of the dielectric functions $\epsilon_{1,2}(i\xi_j)$, and consequently of the inverse lengths $\kappa_{1,2}(i\xi_j)$ (31), are real. We are interested in the limit $q\to 0$ for which $\kappa_{1,2}(i\xi_j)=\xi_j\epsilon_{1,2}^{1/2}(i\xi_j)$. Since $\xi_j\propto j$ and, according to Eq. (28a), $\epsilon(i\xi_j)-1=O(1/j^2)$, the sum in Eq. (48) converges. This means that the function $F_{\rm qu}^{(\rm r)}(0,q)$, being of the order $O(q^2)$, becomes negligible in comparison with the first term in Eq. (47a) when $q\to 0$. In view of representation (6), we find the static prefactor associated with the asymptotic decay to be

$$h_{\rm qu}^{(\rm r)}(0) = -\frac{1}{8\pi^2}\left(\frac{1}{\epsilon_1(0)} + \frac{1}{\epsilon_2(0)} - \frac{4}{\epsilon_1(0)+\epsilon_2(0)}\right). \tag{49}$$

Since this expression does not depend on the temperature and $\hbar$, its classical $\beta\hbar\to 0$ limit is the same, i.e.,

$$h_{\rm cl}^{(\rm r)}(0) = -\frac{1}{8\pi^2}\left(\frac{1}{\epsilon_1(0)} + \frac{1}{\epsilon_2(0)} - \frac{4}{\epsilon_1(0)+\epsilon_2(0)}\right). \tag{50}$$

B. Nonretarded regime

In the nonretarded case (32), we have

$$F_{\rm qu}^{(\rm nr)}(0,q) = \frac{q}{2\pi}\sum_{j=1}^{\infty}\left(\frac{1}{\epsilon_1(i\xi_j)} + \frac{1}{\epsilon_2(i\xi_j)} - \frac{4}{\epsilon_1(i\xi_j)+\epsilon_2(i\xi_j)}\right). \tag{51}$$

It is evident from the asymptotic behavior $\epsilon(i\xi_j)-1=O(1/j^2)$ that the sum in Eq. (51) converges. The function $F_{\rm qu}^{(\rm nr)}(0,q)$ is of the order $O(q)$ and its contribution to $\beta S_{\rm qu}^{(\rm nr)}(0,q)$ in Eq. (47a) is nonzero in the limit $q\to 0$.

The explicit evaluation of the infinite sum over the Matsubara frequencies in Eq. (51) is, in general, very complicated. As a check of the presented formalism, we reconsider the previously studied case of the jellium in contact with vacuum, i.e.,

$$\epsilon_1(\omega) = 1 - \frac{\omega_p^2}{\omega^2}, \quad \epsilon_2(\omega) = 1. \tag{52}$$

Inserting these dielectric functions into Eq. (51) and using the analog of the summation formula (42),

$$\sum_{j=1}^{\infty}\frac{(\alpha/\pi)^2}{j^2+(\alpha/\pi)^2} = \frac{1}{2}(\alpha\coth\alpha - 1) \tag{53}$$

for $\alpha=\beta\hbar\omega_p/2$ and $\beta\hbar\omega_s/2$ ($\omega_s=\omega_p/\sqrt{2}$), we obtain

$$F_{\rm qu}^{(\rm nr)}(0,q) = \frac{q}{4\pi}[2g(\omega_s) - g(\omega_p) - 1] \tag{54}$$

with $g(\omega)$ defined in Eq. (8b). Adding to this function the first term $q/(4\pi)$ in Eq. (47a), we reproduce the $t=0$ case of the previous result (8a).

In the classical (high-temperature) limit $\beta\hbar\to 0$, each of the frequencies $\{\xi_j\}_{j=1}^{\infty}$ tends to infinity, the corresponding terms in the summation over j in Eq. (51) vanish, and so $F_{\rm qu}^{(\rm nr)}(0,q)\to 0$. We are left with only the contribution identical to the retarded classical result (50), i.e., $h_{\rm cl}^{(\rm nr)}(0)=h_{\rm cl}^{(\rm r)}(0)$, as it should be. For the configuration of a conductor, $\epsilon_1(0)=\infty$, in contact with vacuum, $\epsilon_2(0)=1$, we recover the universal result (7). For the configuration of a dielectric, $\epsilon_1(0)=\epsilon_0$, in contact with vacuum, $\epsilon_2(0)=1$, we recover our previous result (21). The classical static prefactor $h_{\rm cl}(0)$ to the $1/R^3$ asymptotic decay is nonzero for an arbitrary configuration of two distinct media, except for the special case of two conductors. In that special case, the asymptotic decay is of a short-ranged type.

V. TIME-DEPENDENT CHARGE CORRELATIONS

For an arbitrary time difference $t\geq 0$ between two points on the interface, the quantum formula for the Fourier transform of the surface charge correlation function (5) reads [1]

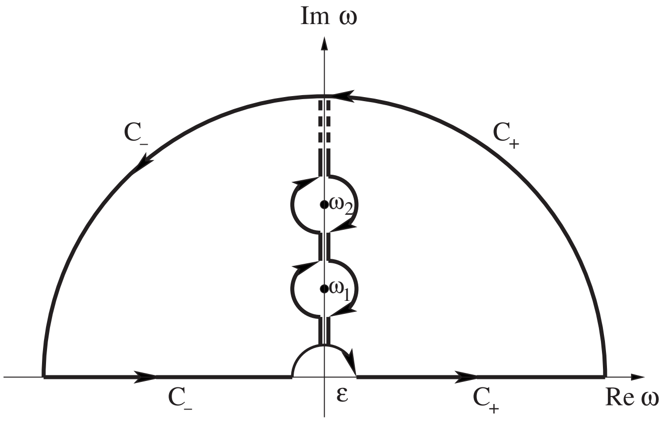

FIG. 3. Two contours in the complex frequency plane for $t\neq 0$; $\omega_j = i\xi_j$ $(j=1,2,\ldots)$ are the Matsubara frequencies.

$$\beta S_{\mathrm{qu}}(t,q) = \int_{-\infty}^{\infty} \frac{d\omega}{\omega} e^{-i\omega t}\, \mathrm{Im}\, f(\omega) \tag{55a}$$

$$= \int_{-\infty}^{\infty} \frac{d\omega}{\omega} \cos(\omega t) \mathrm{Im}\, f(\omega), \tag{55b}$$

where the retarded form of $f(\omega)$ is given in Eq. (30) and the nonretarded one is given in Eq. (32).

Substituting $f(\omega)$ by $e^{-i\omega t} f(\omega)$ we can follow, in principle, the procedure outlined between Eqs. (36) and (39) of the previous section to extend the integration over real frequencies to an integration over the contour C in Fig. 2. The trouble is that on the left-hand side (lhs) of Eq. (39) we end up with the integration over ω of $1/\omega$ multiplied by

$$\mathrm{Im}\, e^{-i\omega t} f(\omega) = \cos(\omega t) \mathrm{Im}\, f(\omega) + \sin(\omega t) \mathrm{Re}\, f(\omega). \tag{56}$$

Comparing this expression with representation (55b), we see that only the first term is needed. Within the C-contour formalism, we did not find a way how to get off the second (unwanted) term.

Our strategy is based on a transition from real times $t\geq 0$ to imaginary times. In order to keep the function $e^{-i\omega t} f(\omega)/\omega$ integrable over real ω, we make the substitutions $t\to -i\tau$ for $\omega>0$ and $t\to i\tau$ for $\omega<0$ $(\tau\geq 0)$, transforming in this way $e^{-i\omega t} f(\omega) \to e^{-|\omega|\tau} f(\omega)$. In the ω upper half plane, the contour C in Fig. 2 will be replaced with two contours C_+ and C_- in the quarter spaces Re $\omega>0$ and Re $\omega<0$, respectively (see Fig. 3). The contours are constructed in such a way that one avoids the singularities (simple poles) of $f(\omega)/\omega$: the one at $\omega=0$ and the infinite sequence of poles at the imaginary Matsubara frequencies $\{i\xi_j\}_{j=1}^{\infty}$. The contour C_+ is directed along the real axis from zero to ∞, except for an infinitesimal quarter circle around the origin $\omega=0$, continues by a quarter circle at infinity, and returns to the origin neighborhood along the imaginary axis, avoiding by infinitesimal semicircles the Matsubara frequencies $\{i\xi_j\}$. The contour C_- is directed along the real axis from $-\infty$ to zero, except for an infinitesimal quarter circle around the origin $\omega=0$, continues along the imaginary axis, avoiding by infinitesimal semicircles the Matsubara frequencies $\{i\xi_j\}$, and returns to the starting point by a quarter circle at infinity. In close analogy with the procedure outlined between Eqs. (36) and (39), we can derive the integral equality

$$\oint_{C_+} \frac{d\omega}{\omega} e^{-\omega\tau} f(\omega) + \oint_{C_-} \frac{d\omega}{\omega} e^{\omega\tau} f(\omega)$$
$$= -i\pi f(0) + \mathcal{P}\int_{-\infty}^{\infty} \frac{d\omega}{\omega} e^{-|\omega|\tau} f(\omega)$$
$$- i\pi \sum_{j=1}^{\infty} \frac{\mathrm{Res}(f,i\xi_j)}{i\xi_j} (e^{-i\xi_j\tau} + e^{i\xi_j\tau}). \tag{57}$$

Here, we have used two facts: the contributions coming from the straight-line fragments of the paths C_+ and C_- between two neighboring frequencies $(i\xi_j, i\xi_{j+1})$ cancel exactly with one another due to the opposite directions of the integrations and the contribution of the C_+ semicircle (C_- semicircle) around the pole $i\xi_j$ is equal to $-i\pi\, \mathrm{Res}(f,i\xi_j)/i\xi_j$ multiplied by the corresponding time-dependent factor $e^{-i\xi_j\tau}$ $(e^{i\xi_j\tau})$. The lhs of Eq. (57) is equal to zero since there are no poles of the integrated functions inside the contours C_+ and C_-. Since the ratio $\mathrm{Res}(f,i\xi_j)/i\xi_j$ is a real number, taking the imaginary part of Eq. (57) leads to

$$\int_{-\infty}^{\infty} \frac{d\omega}{\omega} e^{-|\omega|\tau}\, \mathrm{Im}\, f(\omega) = \pi f(0) + F(\tau,q), \tag{58a}$$

where

$$F(\tau,q) = 2\pi \sum_{j=1}^{\infty} \frac{\mathrm{Res}(f,i\xi_j)}{i\xi_j} \cos(\xi_j\tau). \tag{58b}$$

The function $F(\tau,q)$ represents the (imaginary) time generalization of the static $F(0,q)$ (47b). In the retarded case, the generalization of the static formula (48) reads

$$F_{\mathrm{qu}}^{(\mathrm{r})}(\tau,q) = \frac{q^2}{2\pi} \sum_{j=1}^{\infty} \frac{1}{\kappa_1(i\xi_j)\epsilon_2(i\xi_j) + \kappa_2(i\xi_j)\epsilon_1(i\xi_j)} \times \frac{[\epsilon_1(i\xi_j) - \epsilon_2(i\xi_j)]^2}{\epsilon_1(i\xi_j)\epsilon_2(i\xi_j)} \cos(\xi_j\tau). \tag{59}$$

In the nonretarded case, the generalization of the static formula (51) reads

$$F_{\mathrm{qu}}^{(\mathrm{nr})}(t,q) = \frac{q}{2\pi} \sum_{j=1}^{\infty} \left(\frac{1}{\epsilon_1(i\xi_j)} + \frac{1}{\epsilon_2(i\xi_j)} - \frac{4}{\epsilon_1(i\xi_j) + \epsilon_2(i\xi_j)} \right) \cos(\xi_j\tau). \tag{60}$$

We have shown in Sec. IV that the series determining the functions in Eqs. (59) and (60) are convergent for $\tau=0$. The presence of the oscillating factor $\cos(\xi_j\tau)$ in the series for $\tau>0$ even improves their convergence property. Let us "pretend" that we have found the explicit form of the function $F(\tau,q)$ (58b); the type of the regime is irrelevant. To express the time-dependent correlation function (55b), we have to return from imaginary to real times by considering the substitution $\tau\to it$ in Eq. (58a). Since the integral on the lhs of

Eq. (58a) is finite for all complex τ with Re $\tau \geq 0$, there must exist a well-behaved analytical continuation $F(it,q)$ of the function $F(\tau,q)$. Consequently,

$$\beta S_{\rm qu}(t,q) = \frac{q}{4\pi}\left(\frac{1}{\epsilon_1(0)} + \frac{1}{\epsilon_2(0)} - \frac{4}{\epsilon_1(0)+\epsilon_2(0)}\right) + \text{Re } F(it,q). \tag{61}$$

For $t=0$, the operation Re on $F(0,q)$ become superfluous and we recover the static result (47a).

To check that the formalism works correctly, we reconsider the nonretarded regime for the contact between the jellium and vacuum. Inserting the corresponding dielectric constants (52) into Eq. (60) and using the summation formula [16]

$$\sum_{j=1}^{\infty} \frac{(\alpha/\pi)^2}{j^2+(\alpha/\pi)^2}\cos\left(j\frac{\pi}{\alpha}\omega\tau\right) = \frac{1}{2}\left[\alpha\frac{\cosh(\alpha-\omega\tau)}{\sinh \alpha} - 1\right], \tag{62}$$

we obtain

$$F_{\rm qu}^{\rm (nr)}(\tau,q) = \frac{q}{4\pi}\left(\beta\hbar\omega_s\frac{\cosh[(\beta\hbar\omega_s/2)-\omega_s\tau]}{\sinh(\beta\hbar\omega_s/2)} - \frac{\beta\hbar\omega_p}{2}\frac{\cosh[(\beta\hbar\omega_p/2)-\omega_p\tau]}{\sinh(\beta\hbar\omega_p/2)} - 1\right). \tag{63}$$

The analytical continuation of this function from τ to it is well defined. Using the relation $\text{Re}\cosh(\alpha - i\omega t) = \cosh \alpha \cos(\omega t)$ valid for real α and ωt, we get

$$\text{Re } F_{\rm qu}^{\rm (nr)}(it,q) = \frac{q}{4\pi}[2g(\omega_s)\cos(\omega_s t) - g(\omega_p)\cos(\omega_p t) - 1] \tag{64}$$

with $g(\omega)$ defined in Eq. (8b). Adding to this result the first term $q/(4\pi)$ in Eq. (61), we reproduce the previous time-dependent result (8a).

In the retarded case, the function $F_{\rm qu}^{\rm (r)}(\tau,q)$ in Eq. (59) is of the order $O(q^2)$. The same property holds for its analytical continuation $F_{\rm qu}^{\rm (r)}(it,q)$. Equation (61) then tells us that, in the limit $q\to 0$,

$$\beta S_{\rm qu}^{\rm (r)}(t,q) = \frac{q}{4\pi}\left(\frac{1}{\epsilon_1(0)} + \frac{1}{\epsilon_2(0)} - \frac{4}{\epsilon_1(0)+\epsilon_2(0)}\right). \tag{65}$$

In view of representation (6), the quantum time-dependent prefactor to the asymptotic decay takes, for any temperature, its static classical form

$$h_{\rm qu}^{\rm (r)}(t) = -\frac{1}{8\pi^2}\left(\frac{1}{\epsilon_1(0)} + \frac{1}{\epsilon_2(0)} - \frac{4}{\epsilon_1(0)+\epsilon_2(0)}\right). \tag{66}$$

This result holds for all possible media combinations if one takes $\epsilon(0)\to i\infty$ for conductors, $\epsilon(0)=\epsilon_0>1$ for dielectrics, and $\epsilon(0)=1$ for vacuum.

VI. CONCLUSION

Although the present work is rather technical, its main result (65) [or, equivalently, Eq. (66)] is of physical interest. It represents the generalization of the analogous result, obtained in Refs. [1,2] and valid exclusively for the special jellium model of conductors in contact with vacuum, to all possible media (conductor, dielectric, and vacuum) configurations. The derivation of the general result (65), based on the integration in the complex plane and on the known analytical properties of dielectric functions in the frequency upper half plane, is much simpler and more transparent than the one performed for the jellium in contact with vacuum [1,2].

There is still an open question: which *physical* reasons that the inclusion of retardation effects causes the asymptotic decay of time-dependent quantum surface charge correlations to take its static classical form independent of $\hbar$ and c? For the static quantum case $t=0$, a possible explanation might be that the surface charge correlation function depends on the only dimensionless quantity $\beta\hbar c/R$ constructed from universal constants and the distance. If this is the case, the large-distance asymptotics $R\to\infty$ is equivalent to the classical limit $\beta\hbar\to 0$. A verification whether this claim is true or not is left for the future. The case $t\neq 0$ will be still an open problem. Another explanation might be based on an assumption that at distances R much larger than the thermal photon wavelength $\beta\hbar c$ a decoupling between matter and radiation appears and, consequently, only the Coulomb instantaneous interaction intervenes in the asymptotic behavior. Analogous situations occur in the crossover $1/R^6\to 1/R^7$ for van der Waals interactions or in the thermal screening of transverse electromagnetic interactions [18]. It might be also possible to verify our results by using a fully microscopic approach.

We believe that Rytov's fluctuational electrodynamics and the techniques presented are applicable also to other important phenomena associated with the presence of a surface between distinct media. One of such problems can be the evaluation of the shape-dependent dielectric susceptibility tensor [3,19,20] for quantum Coulomb fluids, with and without retardation effects. Another interesting topic is the large-distance behavior of the current-current correlation function near an interface between two media.

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