# Electromagnetic vacuum of complex media. II. Lamb shift and total vacuum energy

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We study the relation between the electromagnetic vacuum energy of a random medium and its optical response. The medium is modeled by a collection of molecular electric dipoles. First, we evaluate the contribution of statistical fluctuations to the average total vacuum energy, which is made out of the integration of the variations of the Lamb shift of the individual dipoles with respect to the coupling constant. While the Lamb shift is a function of the electrical susceptibility only, the vacuum energy is generally not. Second, we compare several approximations to the computation of the vacuum energy. In particular, we make clear why the bulk energy of an effective medium does not account for the total vacuum energy of a molecular dielectric. Consequently, the Lamb shift does not derive from the effective medium bulk energy except at leading order in the molecular density. The local field factors provide natural cutoffs for the spectrum of the total vacuum energy at a wavelength of the order of the correlation length. Third, we investigate to what extent the shift of the resonant frequency of the dielectric constant may be attributed to the binding energy of a dielectric. In particular, in the effective medium approximation we have found an equivalence between the energy of longitudinal long-wavelength modes and the Lorentz-Lorenz shift. Nonetheless, we conclude that the dielectric constant resonance shift is insufficient to estimate the binding energy of molecular clusters.

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## I. INTRODUCTION

It has long since been recognized that the Casimir effects, the van der Waals (vdW) forces, and the Lamb shifts share a common origin [1–3]. It is customary to ascribe the vdW forces and the Lamb shifts to the short-range interactions between the microscopic constituents of dielectric media. In contrast, the Casimir effects are attributed to the long-range interactions between macroscopic objects. Also, some authors refer to the energy of discrete modes as Casimir energy while they term bulk energy that of the continuous spectrum [4–6]. In this paper we deal with a translation-invariant molecular dielectric made of atomic electric dipoles. Therefore, aside from possible geometrical resonances in clusters, the spectrum of electromagnetic (EM) fluctuations is continuous and the above distinctions are unnecessary.

We aim to compute the EM energy of the ground state of both the individual dipole constituents and the dielectric as a whole. Due to the mutual interactions among the dipoles, both ground-state energy levels happen to be shifted with respect to certain values considered as "bare." In the case of the self-energy of the individual dipoles that shift is referred to as the van der Waals shift [7–9], single-molecule Lamb shift, or Lamb shift in short [10,11]. In the case of the energy of the dielectric as a whole, that shift is referred to as binding energy [12–14]. It has been also recognized that the latter can be interpreted, when computed out of the vacuum fluctuations of the EM field with respect to the zero-point EM energy, as the EM vacuum energy [15–18]. Throughout this paper we refer to it both ways.

On the other hand, both the Lamb shift and the binding energy of the dielectric manifest themselves in the shifts of the resonant frequencies of the individual dipole polarizabilities and of the dielectric constant, respectively. While the correspondence is well understood in the case of individual dipoles [8,19], the relation is not that obvious in the case of the dielectric constant. In particular, the dielectric constant is defined in the long-wavelength limit of the effective medium theory. For that reason, the energy associated with the resonance shift of the dielectric constant is not expected to be the actual binding energy. It is referred to as collective Lamb shift after Friedberg *et al.* [11,20–22]. It is one of the purposes of this article to investigate the relation between the binding energy and the dielectric constant.

The energy shifts we are interested in are those due to the EM dipole-dipole interactions and to the interactions of each dipole alone with bare radiation. In order to identify these shifts we must first identify the bare bound-state levels prior to the coupling of the dipoles to each other and to bare radiation. These are the single-atom bound states which are obtained formally by solving the corresponding nonrelativistic Schrödinger equation with an electron-nucleus electrostatic interaction potential in the Born-Oppenheimer (BO) approximation, and the molecular clusters which are bounded by short-ranged forces other than the long-ranged dipole ones. Examples of the latter forces are those derived from the short-ranged part of the Lennard-Jones potential and the steric forces of the molecules in liquid crystals [23]. The addition of the energies of these bare bound states together with the energy of the bare radiative field in free space amount to a (divergent) zero-point constant value to be regularized.

The energies of the bare atomic levels are parameterized by the bare resonant frequencies of the atomic transitions, which account for the electrostatic interactions of the electrons and the nucleus of each atom alone in the BO approximation [24]. The renormalization of the bare resonance of each dipole is due to both its interaction with the bare radiative field and to its interaction with the rest of the dipoles. The former gives rise to the free-space Lamb shift [7,16,25]. It is common to all the dipoles and additive. The latter interaction is mediated by the multiple scattering of virtual photons and gives rise to

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an additional self-energy that we term scattering Lamb shift. In the literature the energy shift which experiences a foreign dipole due to its interaction with a host medium is often referred to as vdW shift (cf. [7–9]). Throughout this paper we stick to the term Lamb shift as used by Fleischhauer [10] to refer to the total self-energy gained by host single dipoles since the free-space and the scattering Lamb shifts enter the renormalized polarizability on the same footing. Physically, the scattering Lamb shift is the energy supplied (or released) in the removal of one of the dipoles out of the dielectric. The vdW forces and torques acting upon each dipole derive from it [19,26]. Its integration is a nonadditive binding energy which involves a number of dipoles and collective degrees of freedom (d.o.f.). The nonadditive character of the scattering Lamb shift can be better explained in terms of the decay of excited atoms. When one of the dipoles is in an excited state, its resonant frequency is shifted as a result of a "resonant" Lamb shift which contains a scattering contribution [7,8,19]. Therefore, part of the energy released through single dipole emission corresponds to the resonant scattering Lamb shift which involves multiple dipole interactions. The calculations in Refs. [8,19,27] have shown that the resonant Lamb shift of individual dipoles and the shifts in their resonant frequencies are equivalent at second-order quantum perturbation theory. On the contrary, when several neighboring dipoles are excited and decay over a time, the excess of energy released is not the sum of the individual Lamb shifts of every atom. That sum is corrected by an amount related to the collective Lamb shift [20], which accounts, at least partially, for some mutual dipole-dipole interactions. It is worth noting that Friedberg *et al.* have identified the collective Lamb shift with the shift on the resonant frequency of the dielectric constant in a homogeneous dielectric. However, we show later that this identification does not apply to the actual binding energy of the dielectric but in the electrostatic effective medium approximation.

When performing ensemble averages, two approximations are taken in this article as considering dipole-dipole interactions. First, we assume quenched disorder, which is the analog to the BO approximation in molecular dynamics. Further comments on this approximation are made in the next section. Second, we assume that the spatial correlation functions do not depend on the dipole-dipole long-ranged interactions but only on short-ranged ones. This is a common approximation in the literature (see, e.g., Refs. [12,28–30]). However, it becomes inadequate in situations in which short- and long-ranged forces compete in the molecular structure. For instance, this is the case of the coagulation process in colloids [31].

On the other hand, according to Schwinger's source theory, the vacuum energy can be computed out of the integration of the variations of an effective action for the EM field with respect to some effective coupling to the dielectric medium [17,18,32]. Since the variations are taken to be adiabatic, the vacuum energy depends on only the initial and final properties of the dielectric. However, the result depends on both the action varied and the parameter of the variations. In this respect, it is our purpose to explain why computations of the vacuum energy based on the variations of the effective medium action with respect to the effective dielectric constant are not suitable to compute either the total binding energy or the scattering Lamb shift.

On the other hand, it is conjectured that there must exist some relation between the spectrum of optical modes and the Lamb shift of individual dipoles and the binding energy of a molecular dielectric [4,16,17,33–35]. To this respect, Feynman [33], Power [16], and Milonni [34] have proved that the free-space Lamb shift can be computed from the variation of the vacuum energy due to the presence of a single atomic dipole. That result has been extended by Schaden, Spruch, and Zhou [35] to the case of a uniform distribution of nonmutually interacting dipoles. In Ref. [4] Milonni, Schaden, and Spruch extrapolated those approaches to the computation of the Lamb shift from variations on the effective medium bulk energy of a molecular dielectric due to a change in the refractive index. However, that result was not totally conclusive since near-field effects and local field factors (LFFs) were neglected in the calculation. In fact, the findings in [27] suggest that that result does not hold when proper account of LFFs and inherent correlations is taken.

The paper is organized as follows. In Sec. II we define fundamental concepts and review the basic formulas obtained in Ref. [27]. In Sec. III we explain the role of statistical fluctuations in the computation of the vacuum energy of a random medium. In Sec. IV we analyze the expressions for the vacuum energy in the quasicrystalline and bulk effective medium approximations. In the latter case, we explain why that approximation is not suitable to study the Lamb shift. In Sec. V we compute the Lamb shift and the vacuum energy density up to order two in the molecular density for a hard-sphere model. The results are compared in Sec. VI with those of the bulk effective medium approximation. Section VII deals with the EM binding energy of a molecular dielectric in the effective medium approximation. We analyze previous approaches [13,27] and comment on the possibility of finding signatures of the vacuum energy on the spectrum of the dielectric constant. The conclusions are summarized in Sec. VIII.

Regarding notation, we label three-spatial-component vectors with arrows and three-by-three tensors with overlines. We denote the Fourier-transform of functionals with $\vec{q},\omega$-dependent arguments instead of the $\vec{r},t$-dependent arguments of their space-time representation. Quantum operators are denoted with bold letters and hats on top.

## II. GROUND STATES, POLARIZABILITIES, AND PROPAGATORS

A generic statistically homogeneous molecular dielectric is made of $N$ equivalent point electric dipoles in a volume $\mathcal{V}$, with a typical correlation length $\xi$, such that, in the limit $\mathcal{V} \gg V \gg \xi^3$, the average numerical density is uniform, $V^{-1}\int_V d^3r\rho(\vec{r}) = \rho = N/\mathcal{V}$. For the sake of simplicity we consider the canonical ensemble at temperature $T$ with $\mathbf{Z}_N(T) = \int \prod_{i=1}^N d^3R^i \exp\left[-U(\vec{R}^1,\ldots,\vec{R}^N)/k_BT\right]$ the $N$-body canonical partition function. $U$ is the microscopical energy density, which is considered a function of the external d.o.f. of the dipoles only. That is, of the center-of-mass positions and velocities, $\vec{R}_i,\vec{v}_i$, $i = 1,\ldots,N$. Eventually, in the limit $k_BT \ll m_iv_i^2, U^{\max}$, $m_i$ being the scatterer mass and $U^{\max}$ the potential barrier from which steric forces derive, we can neglect the scatterer kinetic energy and assume that $U$ is a short-ranged potential. This is the BO-quenched disorder

approximation. In addition, this way we can ignore Doppler effects and guarantee that the correlation functions do not depend on the long-ranged dipole-dipole interaction in good approximation [12,29]. The dielectric constant and the rest of the effective optical parameters are stochastic functions which admit cluster expansions. These are series of $n$-scattering terms computed out of the convolution of $n$-body spatial correlation functions with single-particle polarizabilities and electric field propagators (see below and [30]). Further, we work for simplicity in the zero-temperature limit, $T = 0$. In addition to the aforementioned inequalities, it demands $k_B T \ll \hbar\omega_{AB}$, with $\hbar\omega_{AB}$ the typical excitation energy of the atomic dipoles, so that all the dipoles remain in their ground state.

The EM radiation is intended as a reservoir of an infinite number of d.o.f. while the internal d.o.f. of the dipoles constitute small systems. The coupling between the radiation reservoir and the dipoles is weak so that ordinary time-dependent perturbative calculations can be carried out. In the following we give some definitions and explain the notation.

### A. EM vacuum and atomic ground states

Let us consider an ensemble of $N$ indistinguishable two-level atomic dipoles. Prior to coupling each atom to the bare radiative field and to the rest of the dipoles, we denote the translation-invariant bare EM vacuum at $T = 0$ by $|\Omega_0\rangle$ and the bare ground and excited atomic states, common to all the dipoles, by $|A_b^i\rangle$ and $|B_b^i\rangle$, $i = 1, \ldots, N$, respectively. Dipoles and EM fluctuations are uncoupled and uncorrelated. The divergent zero-point energy of $|\Omega_0\rangle$ is that of the fluctuations of the bare radiative field. The energy of the bare atomic levels accounts for the bare Coulomb interaction between the nucleus (static in the BO approximation) and the electrons [24], in which case two stationary atomic states are assumed to arise with an energy difference $\hbar\omega_{AB}^b$.

Next, consider the dipoles infinitely separated from each other or otherwise a unique dipole in free space, say the one with position vector $\vec{R}^i$, and turn on the free-space interaction Hamiltonian,

$$\hat{H}_{\mathrm{int},0}^i(t) = -e\hat{\vec{\mathbf{r}}}_i(t)\cdot\hat{\vec{\mathbf{E}}}_\perp(\vec{R}^i,t). \tag{1}$$

Here, $e$ is the electronic charge, $e\hat{\vec{\mathbf{r}}}_i(t)$ is the dipole moment operator, and $\hat{\vec{\mathbf{E}}}_\perp(\vec{R}^i,t)$ is the transverse electric field operator upon the $i$th dipole in the Heissenberg representation. Because the Coulombian interaction has been considered integrated out in the bare atomic bound state, only the transverse component of $\hat{\vec{\mathbf{E}}}$ enters $\hat{H}_{\mathrm{int},0}^i$. In this case the EM vacuum gets polarized locally only due to the dipole fluctuations. Reciprocally, the atomic states get renormalized by the bare transverse EM fluctuations and radiation reaction. They are denoted by $|A_0^i\rangle$, $|B_0^i\rangle$, and are common to all the dipoles. As a result, the free electrons' masses get renormalized and the resonant frequency is shifted to some value $\omega_{AB}^0$ [2,36] to be computed later.

Further, when the $N$ dipoles are brought together to form a specific fixed configuration, say $m$, with position vectors $\{\vec{R}_m^i\}$, $i = 1, \ldots, N$, and dipole moment density operator $-e\hat{\vec{\mathbf{r}}}(\vec{r},t) = \sum_{i=1}^N -e\hat{\vec{\mathbf{r}}}_i(t)\delta^{(3)}(\vec{r} - \vec{R}^i)$, the EM vacuum gets polarized by the local and nonlocal dipole fluctuations and the atomic states get renormalized by both the local interaction of each dipole with the bare EM fluctuations and the nonlocal mutual interactions between the fluctuating dipoles. While the former interactions include only bare transverse EM modes, the latter involve both longitudinal and transverse "dressed" EM modes. Alternatively, it can be interpreted that each atomic state gets renormalized by the local coupling of the corresponding dipole moment to the polarized fluctuations of the EM field, that is, polaritons. For this reason, in contrast to the case of a single dipole in free space, the interaction Hamiltonian in the dipole approximation must contain both the transverse and the longitudinal components of the electric field,

$$\hat{H}_{\mathrm{int}}(t) = -\int d^3r\, e\hat{\vec{\mathbf{r}}}(\vec{r},t)\cdot\hat{\vec{\mathbf{E}}}(\vec{r},t). \tag{2}$$

However, one must bear in mind that divergences coming from considering bare longitudinal modes are regularized in the definition of $\omega_{AB}^b$ (cf. [37]). In this case the polarized vacuum is denoted by $|\Omega_m\rangle$, which is not translation invariant, and the renormalized atomic states are denoted by $|A_m^i\rangle$, $|B_m^i\rangle$, $i = 1, \ldots, N$, being all different in general. The states $|\Omega_m\rangle, |A_m^i\rangle, |B_m^i\rangle$ will be defined later on by their physical content in a way analogous to that in Refs. [27,38]. That is, by the fluctuations on them of the EM field and dipole moment operators, respectively.

Finally, consider a dielectric as a random medium described by a statistical ensemble of dipole configurations. In our approximation, those configurations depend only on external d.o.f. and are independent of the atomic states. Now, on top of the quantum fluctuations that the dipoles and the EM field induce on each other, there are the stochastic fluctuations induced by the random distribution of scatterers. However, generally stochastic fluctuations do not enter the problem in the same fashion as quantum fluctuations. While the correlation time of the latter, $\tau$, satisfies $\tau \ll \omega_{AB}^{-1}$, where $\hbar\omega_{AB}$ is the typical energy transferred in the internal processes, so that quantum fluctuations drive the dynamics of the atoms, the correlation time of the ensemble of configurations is much longer. The latter is also necessary to assume quenched disorder. Thus, stochastic fluctuations enter only as performing ensemble averages and the average energies are thermodynamical potentials (see also note 3). Let $P_m^T$ be the probability density of the $m$th dipole configuration at temperature $T$,

$$P_m^T \prod_{i=1}^N d^3R_m^i = \mathbf{Z}_N^{-1}(T)\prod_{i=1}^N d^3R_m^i \times \exp\left[-U\left(\vec{R}_m^1,\ldots,\vec{R}_m^N\right)/k_BT\right], \tag{3}$$

and $P_m^T|_{\vec{R}_m^i=\vec{r}}$ the conditional probability density with a dipole fixed at $\vec{r}$, say the $i$th,

$$P_m^T\big|_{\vec{R}_m^i=\vec{r}}\prod_{j=1}^N d^3R_m^j = \frac{\mathbf{Z}_N(T)}{\mathbf{Z}_{N-1}|_{\vec{R}^i=\vec{r}}(T)}\delta^{(3)}\left(\vec{r}-\vec{R}_m^i\right)P_m^T\prod_{j=1}^N d^3R_m^j, \tag{4}$$

with $\mathbf{Z}_{N-1}|_{\vec{R}^i=\vec{r}}(T)$ the conditional $(N-1)$-body partition function,

$$\mathbf{Z}_{N-1}|_{\vec{R}^i=\vec{r}}(T) = \int P_m^T \prod_{j=1}^{N} d^3R_m^j \delta^{(3)}(\vec{r}-\vec{R}_m^i). \tag{5}$$

The statistical average of the expectation value of any local operator $\hat{\mathcal{O}}$ in the zero-temperature limit, $\mathcal{O}_m^i = \langle A_m^i, \Omega_m|\hat{\mathcal{O}}|\Omega_m, A_m^i\rangle$, reads

$$\langle\mathcal{O}\rangle_{\text{avg}}^{T=0}(\vec{r}) = \int P_m^{T=0}\big|_{\vec{R}_m^i=\vec{r}} \prod_{j=1}^{N} d^3R_m^j \mathcal{O}_m^i. \tag{6}$$

The choice of $i$ is actually irrelevant since the dipoles are statistically equivalent and so must be any average observable. Alternatively, we can express symbolically $\langle\mathcal{O}\rangle_{\text{avg}}^{T=0}$ as the expectation value of $\hat{\mathcal{O}}$ in the stochastic polarized EM vacuum and renormalized atomic ground state, $|\Omega_{\text{avg}}, A_{\text{avg}}\rangle$. Formally, this can be written in terms of the stationary reduced local (i.e., at each dipole location) density matrix of the system,

$$\begin{aligned}\hat{\rho}^{T=0}(\vec{r}) &= \int P_m^{T=0}\big|_{\vec{R}_m^i=\vec{r}} \prod_{j=1}^{N} d^3R_m^j |\Omega_m\rangle\langle\Omega_m| \otimes |A_m^i\rangle\langle A_m^i| \\ &= |\Omega_{\text{avg}}\rangle\langle\Omega_{\text{avg}}| \otimes |A_{\text{avg}}\rangle\langle A_{\text{avg}}|\big|_{\vec{r}},\end{aligned} \tag{7}$$

in which the integration amounts to taking the partial trace over the statistical mixture of dipole configuration states. The averaged expectation value of $\hat{\mathcal{O}}$ at $T=0$ is

$$\begin{aligned}\langle\mathcal{O}\rangle_{\text{avg}}^{T=0}(\vec{r}) &= \int P_m^{T=0}\big|_{\vec{R}^i=\vec{r}} \prod_{j=1}^{N} d^3R_m^j \langle\Omega^m, A_m^i|\hat{\mathcal{O}}|A_m^i, \Omega^m\rangle \\ &= \langle\Omega_{\text{avg}}, A_{\text{avg}}|\hat{\mathcal{O}}|A_{\text{avg}}, \Omega_{\text{avg}}\rangle|_{\vec{r}} = \text{Tr}\{\hat{\rho}^{T=0}(\vec{r})\cdot\hat{\mathcal{O}}\},\end{aligned}$$

where $|A_m^i, \Omega^m\rangle$ above depends implicitly on $\{\vec{R}_m^j\}, j=1, \ldots, N$. If $\hat{\mathcal{O}}$ is a time-dependent interaction which couples weakly the dipoles to the EM field, by writing $\hat{\rho}$ as a direct product of EM and atomic states we assume that the EM field behaves as a stationary reservoir with respect to the weak interaction and that the time correlation between the reservoir and the dipoles is negligible considering the dynamics of each dipole (cf. Chapter IV of Ref. [39]). Also, in a statistically homogeneous dielectric the spatial correlations functions are translation invariant and so are the stochastic states and average expectation values. Hence, it suffices to compute them at a single point.

### B. Response functions and fluctuations

Our goal is in the first place to give an expression for the Lamb shift of the atomic ground states, $\mathcal{E}^{LSh}$. Second, we use that expression to compute the total binding energy of the ground state of the dielectric at zero temperature. Because the latter can be equally intended as a variation of the zero-point energy of the EM vacuum due to the presence of the dielectric—in the same spirit as the interpretation of Power [16], Feynman [33], and Milonni [34] for the case of a single atom in free space—we refer to it as total vacuum energy density, $\mathcal{F}^V$. We will reinforce this interpretation giving an expression for $\mathcal{F}^V$ in terms of the source field propagator, in accordance to Schwinger's formalism.

In order to compute the Lamb shifts and the total vacuum energy it is necessary to know the expressions for the two-time quadratic correlation functions of the EM field and dipole operators in their renormalized vacuum and ground states, respectively (cf. Chapter IV of Ref. [39] and Ref. [40]). Generically, once the interaction Hamiltonian is turned on, ordinary second-order time-dependent perturbation theory [41] yields an energy shift in the ground state of each dipole, say the $i$th one with position vector $\vec{R}^i$, which corresponds to the generalized Lamb shift,

$$\begin{aligned}\mathcal{E}_{\kappa,i}^{LSh} &= \hbar^{-1}\text{Tr}\left\{\sum_\gamma \left|\langle\gamma, B_\kappa^i|e\hat{\mathbf{r}}_i^S\cdot\hat{\mathbf{E}}^S(\vec{R}^i)|A_\kappa^i, \Omega_\kappa\rangle\right|^2\right\} \\ &\quad\times \text{P}\left[\frac{1}{\omega_\gamma+\omega_{AB}^{\kappa,i}}\right] \\ &= \hbar^{-1}\text{Tr}\left\{\sum_\gamma |\langle\gamma|\hat{\mathbf{E}}^S(\vec{R}^i)|\Omega_\kappa\rangle|^2\cdot\left|\langle A_\kappa^i|e\hat{\mathbf{r}}_i^S|B_\kappa^i\rangle\right|^2\right\} \\ &\quad\times \text{P}\left[\frac{1}{\omega_\gamma+\omega_{AB}^{\kappa,i}}\right].\end{aligned} \tag{8}$$

In this formula P stands for principal value, $\{|\gamma\rangle\}$ is the set of intermediate excited EM states of energy $\hbar\omega_\gamma$, $\omega_{AB}^{\kappa,i}$ is the transition frequency of the $i$th atom, the script $S$ stands for the time-independent operators in the Schrödinger representation, and the script $\kappa$ can take the values $0, m, avg$, corresponding to a single dipole in free space, the $m$th configuration of fixed dipoles, and a statistical ensemble of dipole configurations, respectively. Using an appropriate identity for $\text{P}[\frac{1}{\omega_\gamma+\omega_{AB}^{\kappa,i}}]$, it is shown in Chapter IV of Ref. [39] and Ref. [40] that the above expression can be written in terms of quadratic correlators as

$$\mathcal{E}_{\kappa,i}^{LSh} = -(4\hbar)^{-1}\text{Tr}\left(\int_{-\infty}^{\infty} d\omega \text{Re}\left\{\langle\Omega_\kappa|\hat{\tilde{\mathbf{E}}}(\vec{R}^i;\omega)\otimes\hat{\tilde{\mathbf{E}}}^\dagger(\vec{R}^i;\omega)|\Omega_\kappa\rangle\int dt\,\exp[i\omega t]i\Theta(t)\langle A_\kappa^i|[e\hat{\mathbf{r}}_i(0), e\hat{\mathbf{r}}_i(-t)]|A_\kappa^i\rangle\right\}\right) \tag{9}$$

$$-(4\hbar)^{-1}\text{Tr}\left(\int_{-\infty}^{\infty} d\omega \text{Re}\left\{\langle A_\kappa^i|e\hat{\tilde{\mathbf{r}}}_i(\omega)\otimes e\hat{\tilde{\mathbf{r}}}_i^\dagger(\omega)|A_\kappa^i\rangle\int dt\,\exp[i\omega t]i\Theta(t)\langle\Omega_\kappa|[\hat{\tilde{\mathbf{E}}}(\vec{R}^i,0), \hat{\tilde{\mathbf{E}}}(\vec{R}^i,-t)]|\Omega_\kappa\rangle\right\}\right), \tag{10}$$

where Eq. (9) is the energy shift associated to the polarization of the dipole due to the vacuum field fluctuations while Eq. (10) is the energy shift due to the back-reaction of the polarized EM vacuum on the dipole. Therefore, the problem of computing $\mathcal{E}_{\kappa,i}^{LSh}$ reduces to calculate the equal point two-time quadratic correlation functions of the EM field and dipole

operators in the corresponding EM vacuum and atomic ground state, respectively [42]. In turn, the symmetric correlation function relates to the imaginary part of the linear response function via the fluctuation-dissipation theorem (FDT),

$$\begin{aligned}&\langle\Omega_\kappa|\hat{\vec{\mathbf{E}}}(\vec{R}^i;\omega)\otimes\hat{\vec{\mathbf{E}}}^\dagger(\vec{R}^i;\omega)|\Omega_\kappa\rangle\\&\quad=-\pi^{-1}\mathrm{Im}\Big\{\int dt\,\exp[i\omega t]i\Theta(t)\\&\qquad\times\langle\Omega_\kappa|[\hat{\vec{\mathbf{E}}}(\vec{R}^i,0),\hat{\vec{\mathbf{E}}}(\vec{R}^i,-t)]|\Omega_\kappa\rangle\Big\},\end{aligned}\tag{11}$$

$$\begin{aligned}&\big\langle A^i_\kappa\big|e\hat{\mathbf{r}}_i(\omega)\otimes e\hat{\mathbf{r}}^\dagger_i(\omega)\big|A^i_\kappa\big\rangle\\&\quad=-\pi^{-1}\mathrm{Im}\Big\{\int dt\,\exp[i\omega t]i\Theta(t)\\&\qquad\times\big\langle A^i_\kappa\big|[e\hat{\mathbf{r}}_i(0),e\hat{\mathbf{r}}_i(-t)]\big|A^i_\kappa\big\rangle\Big\},\end{aligned}\tag{12}$$

and the whole problem reduces to computing the local linear response functions, that is, the polarizability of the dipole in its ground state and the Green's function of the electric field, which can be calculated classically. In the following we elaborate on the physical meaning of the response functions.

#### *1. Bare polarizability and field propagator*

It is implicit in our above reasoning that only the bare electrostatic interaction between the electrons and the nucleus in each atom and the electrostatic electron self-interaction are treated nonperturbatively, whereas the rest of interactions are treated perturbatively in the dipole approximation. Therefore, all the functions, energies, and states defined as bare have a nonperturbative origin; while all those termed renormalized in subsequent sections are computed perturbatively. Because the nonperturbative calculations assume the bare radiative field and the intermolecular couplings to be turned off, the response functions and states from them derived are formally correct but do not posses any physical realization.

The equal-point two-time commutator of the dipole operator for an isolated two-level atom, say the $i$th one, in its bare ground state, $|A^i_b\rangle$, prior to the coupling to the radiation reservoir and at zero temperature reads

$$\begin{aligned}&i(\epsilon_0\hbar)^{-1}\Theta(t)\big\langle A^i_b\big|[e\hat{\mathbf{r}}_i(0),e\hat{\mathbf{r}}_i(-t)]\big|A^i_b\big\rangle\\&\quad=i(\epsilon_0\hbar)^{-1}\Theta(t)\big\langle A^i_b\big|e\hat{\mathbf{r}}^S_i\big|B^i_b\big\rangle\big\langle B^i_b\big|e\hat{\mathbf{r}}^S_i\big|A^i_b\big\rangle\\&\qquad\times\big[\exp\big(i\omega^{b,i}_{AB}t\big)-\exp\big(-i\omega^{b,i}_{AB}t\big)\big].\end{aligned}\tag{13}$$

The time-variable Fourier transform of the above equation is termed the bare dipole polarizability, $\bar{\alpha}_b(\omega)$; for brevity we have omitted the location dependence,

$$\begin{aligned}&\bar{\alpha}_b(\omega)\\&\quad\equiv2(\epsilon_0\hbar)^{-1}\big\langle A^i_b\big|e\hat{\mathbf{r}}^S_i\big|B^i_b\big\rangle\big\langle B^i_b\big|e\hat{\mathbf{r}}^S_i\big|A^i_b\big\rangle\omega^{b,i}_{AB}\big[\big(\omega^{b,i}_{AB}\big)^2-\omega^2\big]^{-1}\\&\quad=2(\epsilon_0\hbar)^{-1}\omega^{b,i}_{AB}\big[\big(\omega^{b,i}_{AB}\big)^2-\omega^2\big]^{-1}\vec{\mu}\otimes\vec{\mu},\quad\text{any } i,\end{aligned}\tag{14}$$

where $\vec{\mu}=\langle A^i_b|e\hat{\mathbf{r}}^S_i|B^i_b\rangle$ is the unique dipole-transition matrix element. $\bar{\alpha}_b(\omega)$ is the response function of the dipole to a total monochromatic field acting upon $\vec{R}^i$,[1]

$$\vec{p}^{\,\omega}(\vec{R}_i)=\epsilon_0\bar{\alpha}_b(\omega)\cdot\vec{E}^\omega_{\mathrm{tot}}(\vec{R}^i).\tag{15}$$

For isotropic dipoles with a single oscillator (we omit the dipole index),

$$\begin{aligned}\bar{\alpha}_b(\omega)\equiv\alpha_b\bar{\mathbb{I}}&=\frac{2}{3}(\epsilon_0\hbar)^{-1}\omega^b_{AB}\mu^2\big[\big(\omega^b_{AB}\big)^2-\omega^2\big]^{-1}\\&=\frac{e^2}{3\epsilon_0m^b_e}\big[\big(\omega^b_{AB}\big)^2-\omega^2\big]^{-1},\end{aligned}\tag{16}$$

where $m^b_e$ is the free-electron mass formally renormalized by its electrostatic self-interaction in free space [36]. However, since the effects of the EM vacuum fluctuations and self-polarization have not yet been incorporated, it does not satisfy the optical theorem.

On the other hand, the time-variable Fourier transform of the electric field commutator in the EM vacuum of free space, $i\epsilon_0\hbar^{-1}\Theta(t)\langle\Omega_0|[\hat{\vec{\mathbf{E}}}(\vec{r},0),\hat{\vec{\mathbf{E}}}(\vec{r}',-t)]|\Omega_0\rangle$, is $(\omega/c)^2$ times the Green's function of Maxwell's equation in free space, $\bar{G}^{(0)}(\vec{r}-\vec{r}';\omega)$,

$$\left[\frac{\omega^2}{c^2}\bar{\mathbb{I}}-\vec{\nabla}\times\vec{\nabla}\times\right]\bar{G}^{(0)}(\vec{r}-\vec{r}';\omega)=\delta^{(3)}(\vec{r}-\vec{r}')\bar{\mathbb{I}}.\tag{17}$$

$\bar{G}^{(0)}(\vec{r}-\vec{r}';\omega)$ is made of two contributions. These are an electrostatic one,

$$\bar{G}^{(0)}_{\mathrm{stat.}}(\vec{r};\omega)=\mathrm{P}\left[\frac{1}{k^2}\vec{\nabla}\otimes\vec{\nabla}\right]\left(\frac{-1}{4\pi\,r}\right)+k^{-2}\,\bar{\mathbb{L}}\delta^{(3)}(\vec{r}),\tag{18}$$

$k=\omega/c$, and a radiative field,

$$\bar{G}^{(0)}_{\mathrm{rad.}}(\vec{r};\omega)=\frac{e^{i\,kr}}{-4\pi\,r}\bar{\mathbb{I}}+\mathrm{P}\left[\frac{1}{k^2}\vec{\nabla}\otimes\vec{\nabla}\right]\frac{e^{i\,kr}-1}{-4\pi\,r},\tag{19}$$

where the $\delta$ function in Eq. (18) must be intended in the sense of a distribution. The source tensor $\bar{\mathbb{L}}$ takes account of the geometry of the exclusion volume around each dipole source. It satisfies $\mathrm{Tr}\{\bar{\mathbb{L}}\}=1$ and, for a spherical volume, $\bar{\mathbb{L}}=1/3\bar{\mathbb{I}}$ [43]. $\bar{G}^{(0)}(\vec{r}-\vec{r}';\omega)$ is the response function of the EM field to a unique point dipole oscillating at frequency $\omega$ at $\vec{r}'$ in free space, $\vec{p}^{\,\omega}(\vec{r}')$, $\vec{E}^\omega(\vec{r})=k^2\epsilon_0^{-1}\bar{G}^{(0)}(\vec{r}-\vec{r}';\omega)\cdot\vec{p}^{\,\omega}(\vec{r}')$. In momentum space the radiative component is totally transverse with respect to the propagation direction while the electrostatic one is fully longitudinal,

$$\bar{G}^{(0)}(\vec{q};\omega)=G^{(0)}_\perp(q)\bar{\mathrm{P}}_\perp(\hat{q})+G^{(0)}_\parallel(q)\bar{\mathrm{P}}_\parallel(\hat{q}),\tag{20}$$

with

$$G^{(0)}_\perp(q)=\frac{1}{k^2-q^2},\quad G^{(0)}_\parallel(q)=\frac{1}{k^2},\tag{21}$$

and $\bar{\mathrm{P}}_\perp(\hat{q})=\bar{\mathbb{I}}-\hat{q}\otimes\hat{q}$, $\bar{\mathrm{P}}_\parallel(\hat{q})=\hat{q}\otimes\hat{q}$ being the transverse and longitudinal projectors respectively, with $\hat{q}$ the unitary vector in the direction of propagation. Hereafter and for the sake of brevity we drop the explicit $\omega$ and/or $q$ dependence from the functional arguments unless necessary.

[1]The nomenclature used in this article varies slightly with respect to that in Ref. [27]. In particular, $\alpha_b$ was denoted there as $\alpha'$. Also, what was referred to as polarization propagator in Ref. [27] is referred to here as EM field propagator in order to avoid confusion.

#### 2. A single dipole in free space: Bare radiative corrections

Next, we incorporate the coupling to bare radiation perturbatively following the renormalization scheme of [27,37,44,45]. When the $i$th dipole, isolated, couples to radiation via the Hamiltonian of Eq. (1), the radiation reaction renormalizes the atomic states to $|A_0^i\rangle,|B_0^i\rangle$ and, correspondingly, the single-particle polarizability,

$$\bar{\alpha}(\omega) \equiv \bar{\alpha}_0(1 + ik^2\mathrm{Tr}\{\bar{\alpha}_0 \cdot \mathrm{Im}[\bar{G}^{(0)}(\vec{r},\vec{r};\omega)]\})^{-1}. \quad (22)$$

In the above expression, the real part of $\bar{G}^{(0)}(\vec{r},\vec{r};\omega)$ is absorbed in the 0 polarizability, $\alpha_0 \equiv \frac{e^2}{3\epsilon_0 m_e}[(\omega_{AB}^0)^2 - \omega^2]^{-1}$. In doing so, the divergent real part of $\bar{G}^{(0)}_{\mathrm{stat.}}(\vec{r},\vec{r};\omega)$ is implicitly regularized in $\alpha_b$ [36,37], while the real part of $\bar{G}^{(0)}_{\mathrm{rad.}}(\vec{r},\vec{r};\omega)$ renormalizes the free-electron mass up to $m_e$ and the free-space resonant frequency up to $\omega_{AB}^0$ [2]. The computation of these shifts is outlined later. Hereafter we denote $\omega_{AB}^0$ simply by $\omega_0$. $\bar{\alpha}(\omega)$ does satisfy the optical theorem with $\mathrm{Tr}\{\mathrm{Im}[\bar{G}^{(0)}(\vec{r},\vec{r};\omega)]\} = -k/2\pi$. It is the response function of a unique dipole in free space to an incident monochromatic probe field acting, say, upon $\vec{R}^i$,

$$\vec{p}^{\,\omega}(\vec{R}^i) = \epsilon_0\bar{\alpha}(\omega)\cdot\vec{E}^{\omega}_{\mathrm{inc}}(\vec{R}^i). \quad (23)$$

#### 3. A specific dipole configuration

Next, when all the dipoles are brought together, it was shown in Ref. [27] how a classical diagrammatic renormalization procedure leads to renormalized values for the single particle polarizability and the EM field propagator. In the first place, let us consider a fixed configuration of dipoles with label $m$. The polarization propagator reads

$$\bar{\Pi}^{\omega}_m(\vec{r},\vec{r}') = \sum_{i,j=0}^{N} \bar{\pi}^{\omega}_m\big(\vec{R}^i_m,\vec{R}^j_m\big)\,\delta^{(3)}\big(\vec{r}-\vec{R}^i_m\big)\delta^{(3)}\big(\vec{r}'-\vec{R}^j_m\big), \quad (24)$$

where $\bar{\pi}^{\omega}_m(\vec{R}^i_m,\vec{R}^j_m)$ is

$$\begin{aligned}\bar{\pi}^{\omega}_m&\big(\vec{R}^i_m,\vec{R}^j_m\big)\\ &= \int dt e^{i\omega t} i(\epsilon_0\hbar)^{-1}\Theta(t)\big\langle A^i_m\big|[e\hat{\vec{\mathbf{r}}}_i(0),e\hat{\vec{\mathbf{r}}}_j(-t)]\big|A^j_m\big\rangle\\ &= \big[\alpha_0^{-1}\bar{\mathbb{I}}\delta_{ij} + k^2\bar{G}^{(0)'}\big(\vec{R}^i_m,\vec{R}^j_m\big)\big]^{-1}. \end{aligned} \quad (25)$$

$\bar{G}^{(0)'}(\vec{R}^i_m,\vec{R}^j_m) \equiv \bar{G}^{(0)}(\vec{R}^i_m,\vec{R}^j_m) - \mathrm{Re}\{\bar{G}^{(0)}(\vec{R}^i_m,\vec{R}^j_m)\}\delta_{ij}$ is defined to take account of the regularization of the real divergence in $\bar{G}^{(0)}(\vec{R}^i_m,\vec{R}^i_m)$. The inversion in Eq. (25) must be intended with respect to the particle indices $i,j$. In particular, the local term proportional to $\delta_{ij}$ is defined as the $i$th renormalized polarizability, $\bar{\tilde{\alpha}}^i_m(\omega)$. It is made of an infinite series of multiple-scattering diagrams which start and end at the $i$th dipole. $\bar{\pi}^{\omega}_m(\vec{R}^i_m,\vec{R}^j_m)$ is the (nonlocal) response function of the dielectric to a generic monochromatic external electric field,

$$\vec{p}^{\,\omega}\big(\vec{R}^i_m\big) = \epsilon_0\sum_j^N \bar{\pi}^{\omega}_m\big(\vec{R}^i_m,\vec{R}^j_m\big)\cdot\vec{E}^{\omega}_{\mathrm{ext}}\big(\vec{R}^j_m\big). \quad (26)$$

The relation of $\bar{\pi}^{\omega}_m$ with the $\mathfrak{t}$ matrix defined in the so-called coupled dipole method (CDM) [46] is $\bar{\pi}^{\omega}_m = -k^{-2}\bar{\mathfrak{t}}^{\omega}_m$.

Next, let us compute the EM field propagator which yields the self-polarization corrections on $\bar{\tilde{\alpha}}^i_m(\omega)$, $\bar{\mathfrak{g}}_m(\vec{r},\vec{R}^i_m;\omega)$. It is given by an equation similar to that of Maxwell's in free space,

$$\left[\frac{\omega^2}{c^2}e^{\omega}_{m,i}(\vec{r})\bar{\mathbb{I}} - \vec{\nabla}\times\vec{\nabla}\times\right]\bar{\mathfrak{g}}_m\big(\vec{r},\vec{R}^i_m;\omega\big) = \delta^{(3)}\big(\vec{r}-\vec{R}^i_m\big)\bar{\mathbb{I}}, \quad (27)$$

with $\tilde{e}^{\omega}_m(\vec{r}) = 1 + \alpha_0\sum_{j=1,N}\delta^{(3)}(\vec{r}-\vec{R}^j_m)$ and $e^{\omega}_{m,i}(\vec{r}) = \tilde{e}^{\omega}_m(\vec{r}) - \alpha_0\delta^{(3)}(\vec{r}-\vec{R}^i_m)$. The source fixed at the position vector of the $i$th scatterer is removed from the permittivity function on the left-hand side of the equation. The self-polarization propagator is computed out of $\bar{\mathfrak{g}}_m$, making the source and the emitter coincide. It was calculated in Ref. [27] in function of the polarization propagator,

$$\bar{\mathfrak{g}}_m\big(\vec{R}^i_m,\vec{R}^i_m;\omega\big) = \sum_{j=0}^{N}\bar{G}^{(0)}\big(\vec{R}^i_m-\vec{R}^j_m\big)\cdot\bar{\pi}^{\omega}_m\big(\vec{R}^j_m,\vec{R}^i_m\big)\big[\bar{\tilde{\alpha}}^i_m\big]^{-1}, \quad (28)$$

and by consistency,

$$\bar{\tilde{\alpha}}^i_m = \bar{\alpha}_0\big[1 + k^2\mathrm{Tr}\big\{\bar{\mathfrak{g}}'_m\big(\vec{R}^i_m,\vec{R}^i_m;\omega\big)\cdot\bar{\alpha}_0\big\}\big]^{-1}, \quad (29)$$

where again $\bar{\mathfrak{g}}'_m(\vec{R}^i_m,\vec{R}^i_m;\omega) \equiv \bar{\mathfrak{g}}_m(\vec{R}^i_m,\vec{R}^i_m;\omega) - \mathrm{Re}\{\bar{G}^{(0)}(\vec{R}^i_m,\vec{R}^i_m;\omega)\}$.

Applying reciprocity, $\bar{\mathfrak{g}}_m(\vec{R}^i_m,\vec{r}';\omega)$ yields the incident field which reaches a host dipole at $\vec{R}^i_m$, whose source is a nonpolarizable external monochromatic dipole at any point $\vec{r}'$, $\vec{\mu}^{\omega}_{\mathrm{ext}}$,

$$\vec{E}^{\omega}_{\mathrm{inc}}\big(\vec{R}^i_m\big) = k^2\epsilon_0^{-1}\bar{\mathfrak{g}}_m\big(\vec{R}^i_m,\vec{r}';\omega\big)\cdot\vec{\mu}^{\omega}_{\mathrm{ext}}. \quad (30)$$

Note that since the host dipole at $\vec{R}^i_m$ is polarizable, the total field at $\vec{R}^i_m$ contains an additional contribution from the self-polarization (sp) field. As a result, we have

$$\begin{aligned}\vec{E}^{\omega}_{\mathrm{tot}}\big(\vec{R}^i_m\big) &\equiv \vec{E}^{\omega}_{\mathrm{inc}}\big(\vec{R}^i_m\big) + \vec{E}^{\omega}_{\mathrm{sp}}\big(\vec{R}^i_m\big)\\ &= k^2\epsilon_0^{-1}\bar{\alpha}_0^{-1}\cdot\bar{\tilde{\alpha}}^i_m\cdot\bar{\mathfrak{g}}_m\big(\vec{R}^i_m,\vec{r}';\omega\big)\cdot\vec{\mu}^{\omega}_{\mathrm{ext}}. \end{aligned} \quad (31)$$

From the above equation we can identify a new propagator which accounts for both the two-point, two-time commutator of the EM field in vacuum and the self-polarization field fluctuations which dress up the renormalized polarizability of the emitter or source dipole,

$$\bar{\tilde{\mathfrak{g}}}_m\big(\vec{R}^i_m,\vec{r}';\omega\big) \equiv \big[\bar{\mathbb{I}} + k^2\bar{\mathfrak{g}}'_m\big(\vec{R}^i_m,\vec{R}^i_m\big)\cdot\bar{\alpha}_0\big]^{-1}\cdot\bar{\mathfrak{g}}_m\big(\vec{R}^i_m,\vec{r}'\big), \quad (32)$$

such that

$$\vec{E}^{\omega}_{\mathrm{tot}}\big(\vec{R}^i_m\big) = k^2\epsilon_0^{-1}\bar{\tilde{\mathfrak{g}}}_m\big(\vec{R}^i_m,\vec{r}';\omega\big)\cdot\vec{\mu}^{\omega}_{\mathrm{ext}}. \quad (33)$$

$\bar{\tilde{\mathfrak{g}}}_m$ is the Green's function of an equation like Eq. (27) but replacing $e^{\omega}_{m,i}(\vec{r})$ there with the total dielectric function $\tilde{e}^{\omega}_m(\vec{r})$.

#### 4. *Random medium*

Finally, let us consider a statistical ensemble of configurations of statistically equivalent isotropic dipoles. By simply taking the statistical average on the precedent equations,

$$\bar{\mathcal{G}}(\vec{r},\vec{r}';\omega) = \int P_m^{T=0}\big|_{\vec{R}_m^i=\vec{r}} \prod_{j=1}^{N} d^3R_m^j \bar{\mathfrak{g}}_m\big(\vec{R}_m^i,\vec{r}';\omega\big), \quad (34)$$

or using diagrammatical techniques otherwise in momentum space (cf. [27]), we end up with

$$\begin{aligned} & i\frac{\epsilon_0}{k^2\hbar}\int dt d^3r \exp\left[i(\omega t+\vec{q}\cdot\vec{r})\right]\Theta(t) \\ & \quad \times\langle\Omega_{\rm avg}|[\hat{\vec{\mathbf{E}}}(\vec{0},0),\hat{\vec{\mathbf{E}}}(\vec{r},-t)]|\Omega_{\rm avg}\rangle \\ & = \mathcal{G}_\perp(q;\omega)(\bar{\mathbb{I}}-\hat{q}\otimes\hat{q})+\mathcal{G}_\parallel(q;\omega)\,\hat{q}\otimes\hat{q}, \end{aligned}$$

where

$$\begin{aligned} \mathcal{G}_\perp(q;\omega) &= \frac{\chi_\perp(q;\omega)}{\rho\tilde{\alpha}}G_\perp(q;\omega) = \frac{\chi_\perp(q;\omega)/(\rho\tilde{\alpha})}{k^2[1+\chi_\perp(q;\omega)]-q^2}, \\ \mathcal{G}_\parallel(q;\omega) &= \frac{\chi_\parallel(q;\omega)}{\rho\tilde{\alpha}}\,G_\parallel(q;\omega) = \frac{1}{\rho\tilde{\alpha}}\,\frac{\chi_\parallel(q;\omega)}{k^2[1+\chi_\parallel(q;\omega)]}, \end{aligned} \quad (35)$$

and $G_{\perp,\parallel}(q;\omega)$ are the transverse and longitudinal components of the Dyson (bulk) propagators readily identifiable from the second equalities. The stochastic renormalized polarizability, $\tilde{\alpha}$, is defined later. $\chi_{\perp,\parallel}(q;\omega)$ are the transverse and longitudinal components of the electrical susceptibility. Diagrammatically, $\bar{\chi}$ is made of a series of one-particle-irreducible (1PI) scattering processes like those in Figs. 1(b) and 1(c). The relationship of proportionality between the self-polarization and Dyson's propagator in $q$ space allows us to define the local field factors, $\mathcal{L}_{\perp,\parallel}(q)=\frac{\chi_{\perp,\parallel}(q)}{\rho\tilde{\alpha}}$, such that $\mathcal{G}_{\perp,\parallel}=\mathcal{L}_{\perp,\parallel}G_{\perp,\parallel}$. Alternatively, Eq. (35) can be written also as

$$\mathcal{G}_{\perp,\parallel}(q) = \frac{1}{k^2\rho\tilde{\alpha}}\left[1-\frac{G_{\perp,\parallel}}{G^{(0)}_{\perp,\parallel}}\right]. \quad (36)$$

Equivalently, in terms of the stochastic polarization propagator,

$$\begin{aligned} \bar{\Pi}^\omega(\vec{r},\vec{r}^{\,'}) = \int & \frac{\mathbf{Z}_N(0)}{\mathbf{Z}_{N-2}\big|^{\vec{R}^j=\vec{r}'}_{\vec{R}^i=\vec{r}}(0)} P_m^{T=0}\prod_{l=1}^{N} d^3R_m^l\,\bar{\pi}_m^\omega\big(\vec{R}_m^i,\vec{R}_m^j\big) \\ & \times\delta^{(3)}\big(\vec{r}-\vec{R}_m^i\big)\delta^{(3)}\big(\vec{r}^{\,'}-\vec{R}_m^j\big), \end{aligned} \quad (37)$$

with

$$\mathbf{Z}_{N-2}\big|^{\vec{R}^j=\vec{r}'}_{\vec{R}^i=\vec{r}}(T) = \int P_m^T\prod_{l=1}^{N} d^3R_m^l\,\delta^{(3)}\big(\vec{r}-\vec{R}_m^i\big)\delta^{(3)}\big(\vec{r}^{\,'}-\vec{R}_m^j\big), \quad (38)$$

we may write

$$\mathcal{G}_{\perp,\parallel}(q;\omega) = (\rho\tilde{\alpha})^{-1}G^{(0)}_{\perp,\parallel}(q;\omega)\Pi^\omega_{\perp,\parallel}(q). \quad (39)$$

For convenience we define a scalar potential in terms of the trace of the self-polarization propagator, $\phi(\omega)=\phi^{(0)}(\omega)+\phi^{sc}(\omega)$, where,

$$\phi^{(0)}(\omega) \equiv \frac{\omega^2}{c^2}i\,\mathrm{Im}\left\{2\int\frac{d^3q}{(2\pi)^3}G^{(0)}_\perp\right\} = \frac{-\omega^3}{2\pi c^3}i, \quad (40)$$

$$\begin{aligned} \phi^{sc}(\omega) &\equiv \frac{\omega^2}{c^2}\int\frac{d^3q}{(2\pi)^3}[2(\mathcal{G}_\perp-G^{(0)}_\perp)+(\mathcal{G}_\parallel-G^{(0)}_\parallel)] \\ &\equiv 2\phi^{sc}_\perp+\phi^{sc}_\parallel. \end{aligned} \quad (41)$$

For further convenience, we have distinguished the contribution of transverse from that of longitudinal modes in $\phi^{sc}$. The superscript *sc* stands for *scattering* since $\phi^{sc}(\omega)$ is the part of $\phi$ which involves multiple scattering processes. In function of $\phi$, $\alpha$ and the renormalized stochastic polarizability appearing in Eqs. (35) and (39) read

$$\alpha = \alpha_0(1+\alpha_0\phi^{(0)})^{-1}, \quad (42)$$

$$\tilde{\alpha} = \alpha_0(1+\alpha_0\phi)^{-1} = \alpha(1+\alpha\phi^{sc})^{-1}, \quad (43)$$

all being isotropic, $\bar{\alpha}_0=\alpha_0\bar{\mathbb{I}}$, $\bar{\alpha}=\alpha\bar{\mathbb{I}}$, $\bar{\tilde{\alpha}}=\tilde{\alpha}\bar{\mathbb{I}}$. When a single dipole is excited by a monochromatic external field, the emitted power is given by $W^\omega=\frac{\omega\epsilon_0}{2}|\vec{E}^\omega_{\rm ext}|^2\mathrm{Im}\{\tilde{\alpha}\}$, in agreement with

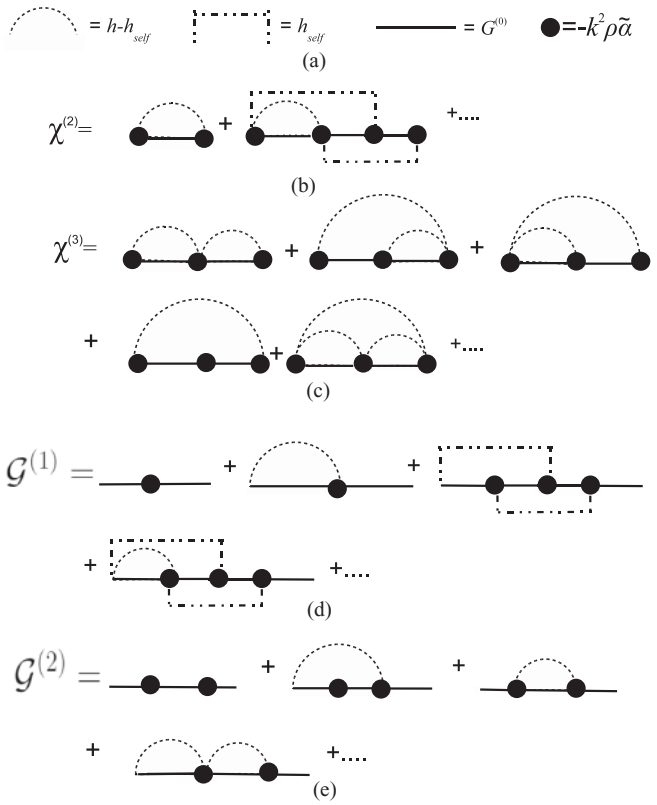


FIG. 1. (a) Diagrammatic representation of Feynman's rules. Only two-point correlation functions are considered for simplicity. The self-correlation function, $h_{\rm self}(\vec{r})=\rho^{-1}\delta^{(3)}(\vec{r})$, appears separated from the rest. (b), (c) Examples of 1PI diagrams which amount to $\chi^{(2)}$ and $\chi^{(3)}$, respectively. The lowest-order recurrent scattering diagram, $\chi^{(2,2)}$, is included in the series of $\chi^{(2)}$. (d), (e) Examples of order $\rho$ and order $\rho^2$ multiple-scattering diagrams which amount to $\mathcal{G}^{(1)}$ and $\mathcal{G}^{(2)}$, respectively. The third and fourth diagrams in (d), which amount to $\mathcal{G}^{(1,2)}$, are entangled recurrent scattering diagrams. In them, the self-correlation functions which affect the first and second scatterers cannot be factored out.

the optical theorem. By parameterizing $\alpha$ and $\tilde{\alpha}$ as Lorentzian polarizabilities, formulas were found in Ref. [27] for the decay rate, $\Gamma$, and frequency shifts. In particular, in free space, $\Gamma_0 = \frac{k_0^3\mu^2}{3\pi\epsilon_0\hbar}$.

As for the case of a specific dipole configuration it is possible to define a propagator which accounts for both the two-point, two-time commutator of the EM field in the stochastic vacuum and the self-polarization field fluctuations which dress up the renormalized stochastic polarizability,

$$\bar{\bar{\mathcal{G}}}(\vec{r},\vec{r}';\omega) \equiv \bar{\mathcal{G}}(\vec{r},\vec{r}';\omega)(1+\alpha_0\phi)^{-1}. \tag{44}$$

The total averaged electric field which acts upon a host dipole at $\vec{r}$ and whose source is an external monochromatic dipole sited at $\vec{r}'$ reads

$$\left\langle \vec{E}^{\omega}_{\rm tot}(\vec{r})\right\rangle_{\rm avg} = k^2\epsilon_0^{-1}\bar{\bar{\mathcal{G}}}(\vec{r},\vec{r}';\omega)\cdot\vec{\mu}^{\omega}_{\rm ext}. \tag{45}$$

For the specific computations in a random medium we use a renormalization scheme similar to that of Felderhof and Cichocki [45]. It consists of two complementary steps which reflect the mutual polarization of the dipoles and the EM reservoir. In the first one, the polarizabilities are renormalized by an infinite number of self-polarization cycles as outlined above. Diagrammatically, those processes amount to recurrent scattering terms in which the initial and final scatterers coincide. Note that those diagrams [cf. Fig. 1(d)] may contain entangled intermediate recurrent scattering processes signaled by self-correlation functions. In the second step, the rest of 1PI diagrams which are not accounted for in the renormalization of the polarizability are added up in the electrical susceptibility. Again, these diagrams may contain also entangled intermediate recurrent scattering processes [cf. Figs. 1(b) and 1(c)]. Consistency between both steps is guaranteed by demanding that the scatterers in all those diagrams be renormalized. Bearing in mind that unentangled recurrent scattering process are all accounted for in the definition of $\tilde{\alpha}$, the electrical susceptibility components, $\chi_{\perp,\parallel}(q;\omega)$, adjust to cluster expansions of the form

$$\chi_{\perp,\parallel}(q;\omega) = \sum_{n=1}\chi^{(n)}_{\perp,\parallel}(q;\omega) = \sum_{n=1,m=0}^{\infty} X^{(n,m)}_{\perp,\parallel}(q;\omega)\rho^n\tilde{\alpha}^{n+m}. \tag{46}$$

The functions $X^{(n,m)}_{\perp,\parallel}(q;\omega)$ incorporate the spatial dispersion due to the 1PI spatial correlations within clusters of $n$ scatterers in which all the self-correlation functions appear entangled. The index $m$ stands for the total multiplicity of entangled intermediate recurrent scattering events. The same kind of decomposition is applicable to $\mathcal{G}_{\perp,\parallel}$ and $\phi^{sc}$ (cf. Fig. 1).

## III. THE LAMB SHIFT AND THE TOTAL VACUUM ENERGY

### A. Lamb shift and Lamb energy in free space

Equipped with the formulas for the response functions, we are now able to compute all the physical quantities. We start with the Lamb shift in free space. Applying the FDT on Eqs. (9) and (10) with $\kappa = 0$ and substituting there Eqs. (40) and (42) we obtain the free-space Lamb shift[2] [7],

$$\mathcal{E}_0^{LSh} = \frac{\hbar}{2\pi}\int_0^{\infty} d\omega\,{\rm Im}\{\alpha_0(1+\alpha_0\phi^{(0)})^{-1}\phi^{(0)}\}. \tag{47}$$

From Eq. (47) we define the density of states contributing to the free-space shift, $\mathcal{N}_0^{LSh}(\omega) = {\rm Im}\{\frac{\rho}{2\pi}\alpha_0\phi^{(0)}(1+\alpha_0\phi^{(0)})^{-1}\}$.

Next, using the Feynman-Pauli theorem (cf. pp. 295–297 of [47]), the free-space vacuum energy is the energy gained by the system atoms-reservoir as the interaction Hamiltonian is turned on adiabatically neglecting dipole mutual couplings. This is parameterized by varying the coupling constant squared from zero to its actual value $e^2$. Since $\alpha_0$ is quadratic in $e$, we can write

$$\begin{aligned}\mathcal{F}_0^V &= \int_0^{\infty}\hbar d\omega\int_0^{\alpha_0}\frac{\delta\alpha_0'}{\alpha_0'}\mathcal{N}_0^{LSh} = \frac{\rho\hbar}{2\pi}\int_0^{\infty}d\omega{\rm Im}\{\ln[1+\alpha_0\phi]\}\\ &= -\frac{\rho\hbar}{2\pi}\int_0^{\infty}d\omega\,{\rm Im}\{\ln[\alpha/\alpha_0]\},\end{aligned} \tag{48}$$

a result that was first obtained by Agarwal [48]. The free-space energies are additive and so we refer to $\mathcal{F}_0^V$ also as free-space Lamb energy density, $\mathcal{F}_0^L$.

### B. Lamb shift and total vacuum energy of a fixed configuration

Let us consider the $m$th configuration of dipoles. Following analogous steps to those for $\mathcal{E}_0^{LSh}$ and $\mathcal{F}_0^V$, we obtain instead,

$$\begin{aligned}\mathcal{E}_m^{LSh,i} &= \frac{\hbar}{2\pi}\int_0^{\infty}d\omega\,{\rm Im}\left\{{\rm Tr}\left[\bar{\tilde{\alpha}}_m^i\cdot\bar{\mathfrak{g}}_m'\left(\vec{R}_m^i,\vec{R}_m^i\right)\right]\right\}\\ &= \frac{\hbar}{2\pi}\int_0^{\infty}d\omega\sum_{j=1}^{N}{\rm Im}\left\{{\rm Tr}\left[\bar{G}^{(0)\prime}\left(\vec{R}_m^i,\vec{R}_m^j\right)\cdot\bar{\pi}_m^{\omega}\left(\vec{R}_m^j,\vec{R}_m^i\right)\right]\right\},\end{aligned} \tag{49}$$

where the trace operation applies over spatial tensor indices only. The first expression after the equality symbol was obtained by Buhmann *et al.* [8,19] using a fully quantum-mechanical formalism. Making the identification

$$\mathcal{N}_m^{LSh,i} = \mathcal{V}^{-1}\sum_{j=1}^{N}{\rm Im}\left\{{\rm Tr}\left[\bar{G}^{(0)\prime}\left(\vec{R}_m^i,\vec{R}_m^j\right)\cdot\bar{\pi}_m^{\omega}\left(\vec{R}_m^j,\vec{R}_m^i\right)\right]\right\}, \tag{50}$$

with the trace again intended over spatial indices only, the total vacuum energy density is

$$\begin{aligned}\mathcal{F}_m^V &= \sum_{i=1}^{N}\int_0^{\infty}\hbar d\omega\int_0^{\alpha_0}\frac{\delta\alpha_0'}{\alpha_0'}\mathcal{N}_m^{LSh,i}\\ &= \frac{\hbar}{2\pi\mathcal{V}}\int_0^{\infty}d\omega{\rm Im}\left({\rm Tr}\left\{\ln\left[\bar{\mathbb{I}}\delta_{ij}+k^2\bar{\alpha}_0\cdot\bar{G}^{(0)\prime}\left(\vec{R}_m^i,\vec{R}_m^j\right)\right]\right\}\right),\end{aligned} \tag{51}$$

[2]We emphasize that in the computation of any local physical observable which is a combination of dipole and EM field operators the inherent divergence in ${\rm Re}\{\bar{G}^{(0)}(\vec{R}_m^i,\vec{R}_m^i;\omega)\}$ must be intended as regularized in both the free-electron mass and the free-space resonant frequency of the oscillator. Hence, the use of regularized "primed" field propagators.

where the trace operation applies both over dipole indices $(i,j)$ and spatial tensor components. This result was first obtained by Renne [49] making the sum over normal modes and then by Agarwal [48] using the FDT in an approach very similar to ours and more recently by Emig *et al*. [50] using a variant of the Schwinger's source theory. Its expansion in multiple-scattering interactions is

$$\begin{aligned}\mathcal{F}_m^V &= \mathcal{F}_0^L + \frac{\hbar}{2\pi\mathcal{V}}\int_0^\infty d\omega \mathrm{Im}\left(\sum_{n=1}^\infty \frac{(-1)^{n+1}}{n}\mathrm{Tr}\{[k^2\bar{\alpha}\cdot\bar{G}^{(0)''}(\vec{R}_m^i,\vec{R}_m^j)]^n\}\right)\\ &= \mathcal{F}_0^L + \frac{\hbar}{2\pi\mathcal{V}}\int_0^\infty d\omega \mathrm{Im}\left\{\frac{-1}{2}\mathrm{Tr}\left[(k^2\bar{\alpha})^2\sum_{i\neq j}\bar{G}^{(0)}(\vec{R}_m^j,\vec{R}_m^i)\cdot\bar{G}^{(0)}(\vec{R}_m^i,\vec{R}_m^j)\right]\right.\\ &\quad\left. + \frac{1}{3}\mathrm{Tr}\left[(k^2\bar{\alpha})^3\sum_{i\neq j\neq l\neq i}\bar{G}^{(0)}(\vec{R}_m^i,\vec{R}_m^j)\cdot\bar{G}^{(0)}(\vec{R}_m^j,\vec{R}_m^l)\cdot\bar{G}^{(0)}(\vec{R}_m^l,\vec{R}_m^i)\right] + \cdots\right\},\end{aligned}$$

where

$$\bar{G}^{(0)''}(\vec{R}_m^i,\vec{R}_m^j) = \bar{G}^{(0)}(\vec{R}_m^i,\vec{R}_m^j) - \bar{G}^{(0)}(\vec{R}_m^i,\vec{R}_m^j)\delta_{ij}. \tag{52}$$

In order to get a deeper insight into the physical interpretation of the above formula, we can write it in three equivalent manners,

$$\mathcal{F}_m^V = -\frac{\hbar}{2\pi\mathcal{V}}\int_0^\infty d\omega \mathrm{Im}\big(\mathrm{Tr}\{\ln[\bar{\pi}_m^\omega(\vec{R}_m^j,\vec{R}_m^i)/\alpha_0]\}\big) \tag{53}$$

$$= -\frac{\hbar}{2\pi\mathcal{V}}\int_0^\infty d\omega \mathrm{Im}\big[\mathrm{Tr}\big(\ln\{\bar{\mathfrak{g}}_m(\vec{R}_m^i,\vec{R}_m^j)\cdot[\bar{G}^{(0)}]^{-1}(\vec{R}_m^i,\vec{R}_m^j)\}\big)\big] \tag{54}$$

$$= -\frac{\hbar}{2\pi\mathcal{V}}\int_0^\infty d\omega \mathrm{Im}\big[\mathrm{Tr}\big(\ln\{\bar{\tilde{\alpha}}_m^i\cdot\bar{\mathfrak{g}}_m(\vec{R}_m^i,\vec{R}_m^j)\cdot\bar{\alpha}_0^{-1}\cdot[\bar{G}^{(0)}]^{-1}(\vec{R}_m^i,\vec{R}_m^j)\}\big)\big], \tag{55}$$

which shows that $\mathcal{F}_m^V$ can be expressed as a function of atomic d.o.f. only Eq. (53), EM d.o.f. only Eq. (54), or as a combination of both Eq. (55). In particular, the expression in Eq. (54) in terms of the source EM field propagator resembles Schwinger's approach [32]. In either case, the normalization by the energy of free-space fluctuations amounts to the substraction of the zero-point EM energy and bare atomic bonding energy.

### C. Average Lamb shift and total vacuum energy of a random medium

Finally, let us compute the average Lamb shift and the average vacuum energy in a random medium, $\mathcal{E}_{\mathrm{avg}}^{LSh}$, $\mathcal{F}_{\mathrm{avg}}^V$. The Lamb shift and vacuum energy so computed must be intended, generally, as thermodynamic quantities. As a matter of fact, stochastic or thermodynamic quantities may be computed in application of the ergodic theorem, which states that statistical ensemble averages are equivalent to time averages.[3] In principle, since some stochastic observables can be calculated out of the optical response functions of the dielectric—$\bar{\chi}^\omega$, $\tilde{\alpha}(\omega)$, $\bar{\Pi}^\omega$, $\bar{G}^\omega$, $\bar{\mathcal{G}}^\omega$, it may seem possible to give a closed formula for $\mathcal{F}_{\mathrm{avg}}^V$ which depends only on the electrical susceptibility and the renormalized polarizability. The computation is, however, far more complicated since the ensemble average over $\mathcal{F}_m^V$ involves highly nonlinear terms in those functions. This can be expressed as

$$\langle\mathrm{Tr}(\ln\{\bar{\tilde{\mathfrak{g}}}\cdot[\bar{G}^{(0)}]^{-1}\})\rangle_{\mathrm{avg}} \neq \mathrm{Tr}(\ln\{\langle\bar{\tilde{\mathfrak{g}}}\cdot[\bar{G}^{(0)}]^{-1}\rangle_{\mathrm{avg}}\}). \tag{56}$$

In the first place, the ensemble average of Eq. (49) is the average Lamb shift,

$$\begin{aligned}\mathcal{E}_{\mathrm{avg}}^{LSh} &= \frac{\hbar}{2\pi\rho}\int_0^\infty d\omega k^2 \mathrm{Im}\left\{\int\frac{d^3q}{(2\pi)^3}[2G_\perp^{(0)}\Pi_\perp + G_\parallel^{(0)}\Pi_\parallel]\right\}\\ &= \frac{\hbar}{2\pi}\int_0^\infty d\omega k^2 \mathrm{Im}\left\{\int\frac{d^3q}{(2\pi)^3}\tilde{\alpha}[2\mathcal{G}_\perp^{'} + \mathcal{G}_\parallel^{'}]\right\} \qquad (57)\\ &= \frac{\hbar}{2\pi}\int_0^\infty d\omega\,\mathrm{Im}\{\tilde{\alpha}\phi\}, \qquad (58)\end{aligned}$$

where we can identify the average density of states, $\mathcal{N}_{\mathrm{avg}}^{LSh} = \rho\mathrm{Im}\{\frac{\tilde{\alpha}}{2\pi}\phi\}$. As noted by Bullough in Ref. [29], the integration of the equation analogous to that of Eq. (51) for $\mathcal{F}_{\mathrm{avg}}^V$,

$$\mathcal{F}_{\mathrm{avg}}^V = \int_0^\infty \hbar d\omega \int_0^{\alpha_0}\frac{\delta\alpha_0^{'}}{\alpha_0^{'}}\mathcal{N}_{\mathrm{avg}}^{LSh}, \tag{59}$$

needs of the knowledge of the functional dependence of $\bar{\chi}$ on the polarizability $\alpha_0$. Thus, $\mathcal{E}_{\mathrm{avg}}^{LSh}$ can be expressed in closed

[3] In principle, in some situations it could be possible to include the randomness of the dipole configurations in the microscopical (not averaged) internal dynamics of the dipoles as considering the computation of the microscopical (not averaged nor thermodynamic) total vacuum energy. For this to be the case the correlation time of the density fluctuations should be much less than the internal dynamical scale, $\sim 2\pi/\omega_0$. However, under these circumstances the quenched disorder approximation would not be valid anymore and Doppler and nonadiabatic effects should be considered.

form as function of $\bar{\chi}$ using Eq. (35),

$$\mathcal{E}_{\rm avg}^{LSh} = \frac{\hbar}{2\pi\rho}\int_0^\infty d\omega k^2 \mathrm{Im}\Big(\int \frac{d^3q}{(2\pi)^3}\{2\chi_\perp G_\perp^{(0)} \times [1+k^2 G_\perp^{(0)}\chi_\perp]^{-1} + \chi_\parallel G_\parallel^{(0)}[1+k^2 G_\parallel^{(0)}\chi_\parallel]^{-1}\}\Big). \tag{60}$$

It is worth stressing that although the computations of $\mathcal{E}_m^{LSh,i}$ in Eq. (49) and $\mathcal{E}_{\rm avg}^{LSh}$ in Eq. (60) are based on the second-order perturbative formula of Eq. (8), the fact that the response functions of dipoles and EM fields there are the renormalized ones makes the results nonperturbative. Hence, for a fixed configuration of dipoles $\mathcal{E}_m^{LSh,i}$ was first obtained by Agarwal [48] by nonperturbative means and it can be verified that the application of the nonpertubative treatment of Buhmann *et al.* [19] to a random medium would yield Eq. (60).

From Eq. (60) we define transverse and longitudinal density of states per unit of momentum volume, $\mathcal{N}_{\rm avg}^{LSh}|_{\perp,\parallel}$,

$$\mathcal{N}_{\rm avg}^{LSh}|_{\perp,\parallel} = \mathrm{Im}\{(2)_\perp k^2 \chi_{\perp,\parallel}^{\omega}(q) G_{\perp,\parallel}^{(0)}[1+k^2 G_{\perp,\parallel}^{(0)}\chi_{\perp,\parallel}^{\omega}]^{-1}\}, \tag{61}$$

where the prefactor $(2)_\perp$ applies only to transverse modes, so that

$$\mathcal{E}_{\rm avg}^{LSh} = \int_0^\infty \hbar d\omega \int \frac{d^3q}{(2\pi)^3}\big[\mathcal{N}_{\rm avg}^{LSh}\big|_\perp + \mathcal{N}_{\rm avg}^{LSh}\big|_\parallel\big]/\rho. \tag{62}$$

However, except for some specific models, $\mathcal{F}_{\rm avg}^V$ cannot be given in closed form. A way to go around this problem is to explode the cluster decomposition outlined previously for $\bar{\chi}$, $\bar{G}$, and $\phi$. First, by taking the ensemble average over the many-body expansion of Eq. (52) we can obtain a series for $\mathcal{F}_{\rm avg}^V$ in multiple scattering terms. Note that this is not yet a series in $\rho$ since, at a given order $n$ of Eq. (52), there are processes with $2,3,\ldots,n$ different indices. When performing upon them the ensemble average, they contribute with terms of order $\rho\alpha\phi^{(1)},\ldots,\rho\alpha\phi^{(n-1)}$ respectively. Equal index terms in Eq. (52) involve self-correlation functions $\delta^{(3)}(\vec{R}_a - \vec{R}_b)$ when performing the ensemble average.

Further, some realistic approximations can be carried out in order to obtain an expansion in powers of $\rho$. In the first place, self-polarization corrections in the renormalized polarizabilities which enter any diagram in $\phi^{(n)}$ can be disregarded in good approximation. Upon integration in $\omega$ both in Eq. (58) and in the average of Eq. (52), they yield terms of order $\sim k_0 r_e \sim \Gamma_0/\omega_0 \ll 1$ smaller than the computations involving bare polarizabilities, $r_e$ being the electron radius and $\Gamma_0$ being the free-space decay rate of an oscillator. Second, entangled intermediate recurrent scattering events can be ignored since they provide terms of orders $[(k_0\xi)^{-3}\Gamma_0/\omega_0]^{sm} \ll 1$, $m \geqslant 2$, $s = 1,2,3,\ldots,n$, smaller than the nonrecurrent, nonretarded near-field terms and terms of orders $[(k_0\xi)^{-1}\Gamma_0/\omega_0]^{sm} \ll 1$, $m \geqslant 2$, $s = 1,2,3,\ldots,n$, smaller than the nonrecurrent, retarded radiative term at any given order $n$ in $\rho$, with $\xi$ being the typical spatial correlation length, $s$ the number of scatterers repeated, and $m$ the recurrent multiplicity (cf. computation of $\phi^{(1)}$ in Sec. V and Appendix A). With these conditions provided, we can simplify Eq. (58) and the average of Eq. (52), respectively, as

$$\begin{aligned}\mathcal{E}_{\rm avg}^{LSh} &= \mathcal{E}_0^{LSh} + \mathcal{E}_{sc}^{LSh} \\ &\simeq \mathcal{E}_0^{LSh} + \frac{\hbar}{2\pi}\int_0^\infty d\omega\ \mathrm{Im}\left\{\int \frac{d^3q}{(2\pi)^3}\alpha_0 \sum_{m=1}^\infty \phi_{\alpha_0}^{(m,0)}\right\} \\ &\equiv \mathcal{E}_{\rm avg}^{LSh}\big|_{\alpha_0},\end{aligned} \tag{63}$$

$$\begin{aligned}\mathcal{F}_{\rm avg}^{V} &\simeq \mathcal{F}_0^{L} + \frac{\hbar}{2\pi}\int_0^\infty d\omega \mathrm{Im}\left\{\sum_{m=1}^\infty \frac{1}{m+1}\rho\alpha_0\phi_{\alpha_0}^{(m,0)}\right\} \\ &\equiv \mathcal{F}_{\rm avg}^{V}\big|_{\alpha_0},\end{aligned} \tag{64}$$

where the scattering Lamb shift, $\mathcal{E}_{sc}^{LSh}$, has been isolated explicitly and $\phi_{\alpha_0}^{(m,0)}(\omega)$ stands for the $m$-body term of the expansion of $\phi$ in which no entangled intermediate recurrent scattering events appear (norec.) and polarizabilites are taken as bare.

Last, we can write Eqs. (63) and (64) as functions of the cluster expansion for $\bar{\chi}$. This way we obtain a formula for $\mathcal{F}_{\rm avg}^V|_{\alpha_0}$ which depends explicitly on (the cluster expansion of) $\bar{\chi}$, order by order in $\rho$. As for Eq. (60), we use Eq. (35) and the precedent approximation to write

$$\begin{aligned}\rho\alpha_0\phi_{\alpha_0}^{(\rm norec.)} &= k^2\int\frac{d^3q}{(2\pi)^3}\big\{2\chi_{\perp,\alpha_0}^{\rm norec.}G_\perp^{(0)}\big[1+k^2G_\perp^{(0)}\chi_{\perp,\alpha_0}^{\rm norec.}\big]^{-1} \\ &\quad + \chi_{\parallel,\alpha_0}^{\rm norec.}G_\parallel^{(0)}\big[1+k^2G_\parallel^{(0)}\chi_{\parallel,\alpha_0}^{\rm norec.}\big]^{-1}\big\} \qquad (65) \\ &= \int\frac{d^3q}{(2\pi)^3}\sum_{m=1}(-1)^{m+1}\big\{2\big[k^2\chi_{\perp,\alpha_0}^{\rm norec.}G_\perp^{(0)}\big]^m \\ &\quad + \big[k^2\chi_{\parallel,\alpha_0}^{\rm norec.}G_\parallel^{(0)}\big]^m\big\}, \qquad (66)\end{aligned}$$

where $\chi_{\perp,\parallel,\alpha_0}^{\rm norec.}$ are the transverse and longitudinal components of the susceptibility with bare polarizabilities and no entangled recurrent scattering processes. Using the cluster expansion of Eq. (46) we obtain

$$\begin{aligned}\mathcal{E}_{\rm avg}^{LSh}\big|_{\alpha_0} &= \mathcal{E}_0^{LSh} + \frac{\hbar}{2\pi\rho}\int_0^\infty d\omega \mathrm{Im}\Big\{\int\frac{d^3q}{(2\pi)^3}2\big[k^2\chi_{\perp,\alpha_0}^{(2,0)}G_\perp^{(0)} \\ &\quad - \big(k^2\chi_{\perp,\alpha_0}^{(1,0)}G_\perp^{(0)}\big)^2 + \big(k^2\chi_{\perp,\alpha_0}^{(1,0)}G_\perp^{(0)}\big)^3 \\ &\quad + k^2G_\perp^{(0)}\chi_{\perp,\alpha_0}^{(3,0)} - 2\chi_{\perp,\alpha_0}^{(1,0)}\chi_{\perp,\alpha_0}^{(2,0)}(k^2G_\perp^{(0)})^2 + \cdots\big] \\ &\quad + \big[k^2\chi_{\parallel,\alpha_0}^{(2,0)}G_\parallel^{(0)} - \big(k^2\chi_{\parallel,\alpha_0}^{(1,0)}G_\parallel^{(0)}\big)^2 + \big(k^2\chi_{\parallel,\alpha_0}^{(1,0)}G_\parallel^{(0)}\big)^3 \\ &\quad + k^2G_\parallel^{(0)}\chi_{\parallel,\alpha_0}^{(3,0)} - 2\chi_{\parallel,\alpha_0}^{(1,0)}\chi_{\parallel,\alpha_0}^{(2,0)}(k^2G_\parallel^{(0)})^2 + \cdots\big]\Big\},\end{aligned} \tag{67}$$

$$\begin{aligned}\mathcal{F}_{\rm avg}^{V}\big|_{\alpha_0} &= \mathcal{F}_0^{L} + \frac{\hbar}{2\pi}\int_0^\infty d\omega \mathrm{Im}\Big(\int\frac{d^3q}{(2\pi)^3}2\Big\{\frac{1}{2}\big[k^2\chi_{\perp,\alpha_0}^{(2,0)}G_\perp^{(0)} \\ &\quad - \big(k^2\chi_{\perp,\alpha_0}^{(1,0)}G_\perp^{(0)}\big)^2\big] + \frac{1}{3}\big[\big(k^2\chi_{\perp,\alpha_0}^{(1,0)}G_\perp^{(0)}\big)^3 \\ &\quad + k^2G_\perp^{(0)}\chi_{\perp,\alpha_0}^{(3,0)} - 2\chi_{\perp,\alpha_0}^{(1,0)}\chi_{\perp,\alpha_0}^{(2,0)}(k^2G_\perp^{(0)})^2\big] + \cdots\Big\} \\ &\quad + \Big\{\frac{1}{2}\big[k^2\chi_{\parallel,\alpha_0}^{(2,0)}G_\parallel^{(0)} - \big(k^2\chi_{\parallel,\alpha_0}^{(1,0)}G_\parallel^{(0)}\big)^2\big]\end{aligned}$$

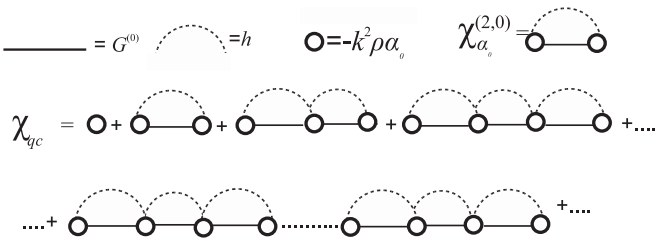

FIG. 2. Diagrammatic representation of Eq. (70).

$$+\frac{1}{3}\Big[\big(k^2\chi^{(1,0)}_{\parallel,\alpha_0}G^{(0)}_{\parallel}\big)^3+k^2G^{(0)}_{\parallel}\chi^{(3,0)}_{\parallel,\alpha_0} -2\chi^{(1,0)}_{\parallel,\alpha_0}\chi^{(2,0)}_{\parallel,\alpha_0}(k^2G^{(0)}_{\parallel})^2\Big]+\cdots\Big\}\Big), \tag{68}$$

which, in turn, yields an expansion in powers of $\rho e^2$ like that of Mclachlan [51]. It is plain that the problem in integrating Eq. (59) has been shifted to that of knowing $\chi_{\perp,\parallel}(q;\omega)$ as a power series of $\rho$ or, otherwise, knowing all order spatial correlation functions.

## IV. APPROXIMATIONS TO THE TOTAL VACUUM ENERGY OF A RANDOM MEDIUM

### A. The quasicrystalline approximation

Previous works (cf. [13,27,29]) have reported closed formulas for $\mathcal{F}^V_{\rm avg}$ as functions of $\chi_{\perp,\parallel}$. However, they are model dependent. Nonetheless, they are useful for estimating orders of magnitude. In particular, that in Ref. [27] corresponds to the so-called quasicrystalline (qc) approximation. According to this approximation the only relevant correlation function is the two-body one, $h(r)$, and self-correlations are ignored. In this approximation, the series of $\bar{\chi}(q)$ becomes geometrical and the only quantity to be computed is $\bar{\chi}^{(2,0)}(q)$,

$$\chi^{(2,0)}_{p,\alpha_0}(q)=\frac{-k^2\alpha_0^2\rho^2}{(1+\delta^p_{\perp})}\int d^3r e^{i\vec{q}\cdot\vec{r}}h(r)\mathrm{Tr}\{\bar{G}^{(0)}(\vec{r})\cdot\bar{\mathrm{P}}_p(\hat{q})\}, \tag{69}$$

with $p=\perp,\ \parallel$. The geometrical series in Fig. 2 is

$$\chi^{\rm qc}_{\perp,\parallel}(q;\omega)=\frac{\rho\alpha_0}{1-\chi^{(2,0)}_{\perp,\parallel,\alpha_0}(q;\omega)/\rho\alpha_0}, \tag{70}$$

and so

$$\mathcal{G}^{\rm qc}_{\perp,\parallel}(q,\omega)=G^{(0)}_{\perp,\parallel}\big[1+\rho\alpha_0\big(k^2G^{(0)}_{\perp,\parallel}-\chi^{(2,0)}_{\perp,\parallel,\alpha_0}/\rho^2\alpha_0^2\big)\big]^{-1}, \tag{71}$$

and

$$\mathcal{F}^V_{\rm qc}=\frac{-\hbar}{2\pi}\int_0^\infty d\omega\mathrm{Im}\Big(\int\frac{d^3q}{(2\pi)^3} \times\ln\big\{\big[\mathcal{G}^{\rm qc}_{\perp,\alpha_0}\big]^2\mathcal{G}^{\rm qc}_{\parallel,\alpha_0}[G^{(0)}_{\perp}]^{-2}[G^{(0)}_{\perp}]^{-1}\big\}\Big) \tag{72}$$

$$=\frac{\hbar}{2\pi}\int_0^\infty d\omega\mathrm{Im}\Big(\int\frac{d^3q}{(2\pi)^3} \times\Big\{2\ln\big[1+\rho\alpha_0\big(k^2G^{(0)}_{\perp}-\chi^{(2,0)}_{\perp,\alpha_0}/\rho^2\alpha_0^2\big)\big] +\ln\big[1+\rho\alpha_0\big(k^2G^{(0)}_{\parallel}-\chi^{(2,0)}_{\parallel,\alpha_0}/\rho^2\alpha_0^2\big)\big]\Big\}\Big), \tag{73}$$

where by ignoring self-polarization it holds that $\tilde{\mathcal{G}}^{\rm qc}_{\perp,\parallel}=\mathcal{G}^{\rm qc}_{\perp,\parallel}$. It is because of the aforementioned analogy between the formulas for a specific configuration of dipoles (cf. a cubic lattice [13]) and those of the qc approximation that the inequality expressed in Eq. (56) turns into an equality in this case. It is also possible to write Eq. (73) as

$$\mathcal{F}^V_{\rm qc}=-\hbar\mathrm{Im}\left\{\int_0^\infty\frac{d\omega}{2\pi}\int\frac{d^3q}{(2\pi)^3}2\ln[\chi^{\rm qc}_{\perp}G^{\rm qc}_{\perp}]+\ln[\chi^{\rm qc}_{\parallel}G^{\rm qc}_{\parallel}]\right\} \tag{74}$$

$$+\hbar\mathrm{Im}\left(\int_0^\infty\frac{d\omega}{2\pi}\int\frac{d^3q}{(2\pi)^3}2\ln\{[G^{(0)}_{\perp}]\}+\ln\{[G^{(0)}_{\parallel}]\}\right) \tag{75}$$

$$+3\hbar\mathrm{Im}\left\{\int_0^\infty\frac{d\omega}{2\pi}\int\frac{d^3q}{(2\pi)^3}\ln[\alpha_0]\right\}. \tag{76}$$

Making the identification $\int\frac{d^3q}{(2\pi)^3}=\rho$ in Eq. (76)[4] we read that the bare atomic bonding energy and the EM zero-point energy enter as substraction terms. This justifies the interpretation of $\mathcal{F}^V_{\rm qc}$ in Ref. [13]. That is, $\mathcal{F}^V_{\rm qc}$ takes account of the zero-point energy of bare EM modes in Eq. (75) and the atomic bonding energy of Eq. (76) and substitutes them with the binding energy of the coupled system of Eq. (74).

Further, by considering the continuum limit of the effective medium approximation, $q\xi\ll 1$, we will see that $\mathcal{F}^V_{\rm qc}(q\xi\ll 1)$ can be written as a function of the refractive index. We will discuss the accuracy of this approximation.

Although the expression obtained in Ref. [27] for $\mathcal{F}^V_{\rm avg}$ coincides with that of Eq. (72), the derivation there was erroneous and hence its validity restricts to the qc approximation. The reasoning followed in Ref. [27] was that the average energy could be obtained by extending appropriately Schwinger's approach on a bulk effective medium [18] to a molecular dielectric. Thus, the steps followed by Schwinger *et al.* [52] and Schwinger in Refs. [17,18] were mimicked in Ref. [27] but for the fact that the effective bulk propagator was replaced by the averaged source field propagator of a molecular dielectric. The reason for doing this was the constatation in Ref. [27] that the effective bulk propagator used in Refs. [17,18,52] is not the one which enters the formula for the Lamb shift in Eq. (57). Accidentally, the approach in Ref. [27] turns the inequality of Eq. (56) into an equality, which is generally incorrect.

### B. The Schwinger bulk energy of an effective medium

In Ref. [52], Schwinger, de Raad, and Milton applied Schwinger's (Sch.) source theory [32] to the computation of the vacuum energy of a piecewise homogeneous effective medium suitable for the further computation of the Casimir forces between macroscopic effective dielectrics. To this aim they postulated an effective EM action with an effective interaction Hamiltonian,

$$H^{\rm eff}_{\rm int}=-\int d^3r\,\vec{P}_{\rm eff}(\vec{r},t)\cdot\vec{E}^{\perp}_{\rm eff}(\vec{r},t). \tag{77}$$

[4]This equivalence can be proved passing to the continuum from a lattice of dipoles whose cell volume is $\rho^{-1}$. It acts as a regulator.

In this Hamiltonian the effective electric field $\vec{E}^{\perp}_{\rm eff}$ is the transverse macroscopic "bulk" field which relates to the effective polarization density, $\vec{P}_{\rm eff}$, through the transverse effective Dyson propagator, $G^{\rm eff}_{\perp}(q;\omega)=(\epsilon^{\omega}_{\rm eff}k^2-q^2)^{-1}$,

$$\vec{E}^{\perp}_{\rm eff}(\vec{r};\omega)=k^2\epsilon_0^{-1}\int d^3r'\bar{G}^{\rm eff}_{\perp}(\vec{r},\vec{r}';\omega)\cdot\vec{P}^{\omega}_{\rm eff}(\vec{r}'). \tag{78}$$

In $G^{\rm eff}_{\perp}$, $\epsilon^{\omega}_{\rm eff}=1+\chi^{\omega}_{\rm eff}$ is the effective dielectric constant and, adapting our nomenclature, $G^{\rm eff}_{\perp}$ characterizes the EM field fluctuations of the "Schwinger vacuum," $|\Omega_{\rm Sch.}\rangle$. The corresponding energy shift reads

$$\mathcal{E}_{\rm Sch.}=-{\rm Re}\left\{\int d^3r\,\langle\Omega_{\rm Sch.}|\hat{\vec{\mathbf{P}}}_{\rm eff}(\vec{r},t)\cdot\hat{\vec{\mathbf{E}}}^{\perp}_{\rm eff}(\vec{r},t)|\Omega_{\rm Sch.}\rangle\right\}, \tag{79}$$

and varying the action of the effective medium, Schwinger *et al*. [52] inferred the identification of the effective polarization source quadratic fluctuations,

$$\int dt\,\exp[i\omega t]i(\epsilon_0\hbar)^{-1}\Theta(t)\langle\Omega_{\rm Sch.}|\hat{\vec{\mathbf{P}}}_{\rm eff}(\vec{r},0)\otimes\hat{\vec{\mathbf{P}}}_{\rm eff}(\vec{r}',t)|\Omega_{\rm Sch.}\rangle$$
$$=\delta\bar{\chi}^{\omega}_{\rm eff}\delta^{(3)}(\vec{r}-\vec{r}').$$

Inserting the above expression together with Eq. (78) into Eq. (79) we get [17]

$$\begin{aligned}\delta_{\chi'}\mathcal{E}_{\rm Sch.}&=\frac{\hbar}{2\pi\rho}\int_0^{\infty}d\omega k^2{\rm Im}\left\{\int\frac{d^3q}{(2\pi)^3}2\delta\chi'^{\omega}_{\rm eff}G^{\rm eff}_{\perp}\right\}\\&=\frac{\hbar}{2\pi\rho}\int_0^{\infty}d\omega k^2{\rm Im}\left\{\int\frac{d^3q}{(2\pi)^3}2\delta\chi'^{\omega}_{\rm eff}G^{(0)}_{\perp}\right.\\&\quad\left.\times\left[1+k^2G^{(0)}_{\perp}\chi'^{\omega}_{\rm eff}\right]^{-1}\right\}.\end{aligned} \tag{80}$$

The functional integration immediately yields

$$\mathcal{F}^{V}_{\rm Sch.}=\frac{-\hbar}{2\pi}\int_0^{\infty}d\omega{\rm Im}\left(\int\frac{d^3q}{(2\pi)^3}\ln\left\{[G^{\rm eff}_{\perp}]^2[G^{(0)}_{\perp}]^{-2}\right\}\right), \tag{81}$$

which is a function of the transverse effective Dyson propagator only. Direct comparison of Eq. (81) and Eqs. (74)–(76), with $\chi^{\rm qc}_{\perp,\parallel}$ considered $\chi_{\rm eff}$ in the continuous limit, reveals that $\mathcal{F}^{V}_{\rm qc}$ includes contributions disregarded in the computation of [52]. In particular, no longitudinal modes and no LFFs are present in Eq. (81). Nonetheless, the equations of [52] are sufficient for the treatment of problems in which the internal properties of the objects are integrated in the definition of their effective permittivities and remain unaffected during the dynamical phenomena under study. This is the case of the Casimir forces between macroscopic dielectrics separated by macroscopic distances in which the relevant variations in energy are those with respect to that of the macroscopic objects infinitely far apart. To this situation corresponds the Lifshitz problem [1,12] that the authors address in Ref. [52], in which only retarded modes matter. A similar reasoning leads to the derivation of the retarded vdW forces from radiative modes only [6].

#### *1. Relation with the microscopic approach*

In order to understand the link with the microscopic computation we first notice the similarity between Eqs. (60) and (81) for the microscopic computation of $\mathcal{E}^{LSh}_{\rm avg}$. Ignoring the fact that Schwinger neglected the contribution of longitudinal modes, Eq. (81) can be written also as

$$\mathcal{F}^{V}_{\rm Sch.}=\int\hbar d\omega\int\frac{d^3q}{(2\pi)^3}\int_0^{\chi^{\omega}_{\rm eff}}\frac{\delta\chi'^{\omega}_{\rm eff}}{\chi'^{\omega}_{\rm eff}}\mathcal{N}^{LSh}_{\rm avg}|^{\rm eff}_{\perp}, \tag{82}$$

where $\mathcal{N}^{LSh}_{\rm avg}|^{\rm eff}_{\perp}$ is the average density of states restricted to transverse modes in the continuum. This implies that, effectively, in the approach of Schwinger *et al*. the microscopical interaction Hamiltonian of Eq. (2) with dipole moment operator $e\hat{\mathbf{r}}$ and coupling constant $e$ is substituted with the effective Hamiltonian of Eq. (77), which couples the transverse effective EM field to an effective polarization density operator $\hat{\vec{\mathbf{P}}}_{\rm eff}$ with some effective coupling constant proportional to $\chi^{1/2}_{\rm eff}$. Because $\chi_{\rm eff}$ is made of the integration of dipole-dipole interactions, the energy of those interactions is disregarded in the approach of Schwinger *et al*.[5] This means that the Schwinger *et al*. approach is not appropriate to study phenomena driven by the variation of the energies of internal d.o.f.. This is the case of Lamb shifts in cold atomic clusters, atomic vapors [21], and artificial solids [53]; and vdW forces and phase transitions in liquid crystals [54] and colloidal suspensions [31] in which the relevant variations in energy are those with respect to that of the molecular constituents infinitely far apart (see also [55]).[6] Nonetheless, the reason why $\mathcal{E}^{LSh}_{0}$ can be computed out of $\mathcal{F}^{V}_{\rm Sch.}$ [35] is that, at leading order in $\rho$, $\bar{\chi}(\vec{r})\simeq\rho\alpha_0\bar{\mathbb{I}}\delta^{(3)}(\vec{r})$ and so $\mathcal{F}^{V}_{0}=\mathcal{F}^{V}_{\rm Sch.}$ at $O(\rho)$.

The original suggestion of Schwinger to explain sonoluminescence represents a sort of intermediate problem [17]. Basically, when a bubble embedded in water collapses, molecules of water fill in the void and he conjectured that the light emitted in this process carries the excess of EM vacuum energy stored in the void with respect to the energy of the homogenous aqueous medium. Evidently the molecules of water which fill the void form part of the initial aqueous medium surrounding the bubble. Therefore, leaving aside the validity of further dynamical approximations (cf. [56]), the total volume filled by the water is greater after the collapse, the density of water is less and so the dielectric constant must vary, at least locally, in the region within and around the primordial bubble. As a consequence, it is reasonable to think that, if the time scale of light emission is less than the typical homogenization time, the molecules of water within the volume originally occupied by the bubble will have different spatial disposition to that of the homogenous surrounding medium. Therefore, their internal energy will be different to that of the homogeneous phase and the problem involves not only a variation of energy between macroscopic objects (aqueous media with and without a bubble) but also

[5]Analogously, in assuming the bare resonant frequency $\omega^{b}_{AB}$ as a constant value, the variations of the electrostatic binding energy between the electrons and the nucleus of each atom is also disregarded in our approach.

[6]Note that this does not imply by any means that the generic Schwinger's source theory be inapplicable to these problems as shown explicitly for the case of a specific configuration of dipoles Eq. (54) (cf. Ref. [50]).

a difference in the internal energy of the molecules which were initially in a homogenous disposition and fill in the void afterward in a different arrangement. On the contrary, if the homogenization is reached prior to emission and, in good approximation, the permittivity of the filled bubble is identical to that of the surrounding medium, Schwinger's approach might be a good approximation.

#### 2. *Beyond the bulk effective medium approximation*

The result of Schwinger in Ref. [17] yields only the energy associated to radiative modes which depends solely on the effective refractive index, $n(\omega) = \sqrt{1+\chi^{\omega}_{\rm eff}}$, as

$$\mathcal{F}^{V}_{\rm Sch.} \simeq \frac{\hbar}{6\pi^2 c^3}{\rm Re}\left\{\int_0^\infty d\omega\, \omega^3[1-n^3]\right\}, \tag{83}$$

and the Schwinger energy shift is

$$\mathcal{E}_{\rm Sch.} \simeq \frac{-\hbar}{4\pi^2 c^3 \rho}{\rm Re}\left\{\int_0^\infty d\omega\, \omega^3 n \chi^{\omega}_{\rm eff}\right\}. \tag{84}$$

In Sec. V B we compare in detail the above results with the actual microscopical calculation. Schwinger argued [17] that these results in the effective medium approximation could be extended to incorporate spatial dispersion in the electrical susceptibility. In doing so we obtain an extended (ext) Schwinger transverse bulk energy, $\mathcal{F}^{V}_{\rm Sch.}|^{\rm ext}$. However, even in this case the discrepancy between the actual $\mathcal{F}^{V}_{\rm avg}$ and $\mathcal{F}^{V}_{\rm Sch.}|^{\rm ext}$ appears already at order $\rho^2$. For the sake of simplicity we restrict ourselves to the approximations used in Eq. (68) and find

$$\begin{aligned}\mathcal{F}^{V}_{\rm avg}\big|_{\alpha_0} \simeq{}& \mathcal{F}^{V}_{\rm avg}\big|^{\parallel}_{\alpha_0} + \mathcal{F}^{V}_{\rm Sch.}\big|^{\rm ext}_{\alpha_0} + \frac{\hbar}{2\pi}\int_0^\infty d\omega \\ &\times {\rm Im}\left\{\int \frac{d^3q}{(2\pi)^3} 2\left[-\frac{1}{2}k^2\chi^{(2,0)}_{\perp,\alpha_0}G^{(0)}_{\perp}\right.\right. \\ &-\frac{2}{3}k^2 G^{(0)}_{\perp}\chi^{(3,0)}_{\perp,\alpha_0} - \frac{2}{3}\chi^{(1,0)}_{\perp,\alpha_0}\chi^{(2,0)}_{\perp,\alpha_0}(k^2G^{(0)}_{\perp})^2 \\ &\left.\left.-\left(k^2\chi^{(1,0)}_{\perp,\alpha_0}G^{(0)}_{\perp}\right)^3+\cdots\right]\right\},\end{aligned} \tag{85}$$

where $\mathcal{F}^{V}_{\rm avg}|^{\parallel}_{\alpha_0}$ contains the longitudinal modes whose contribution includes radiative energy (cf. computation of $\mathcal{F}^{V}_{\mathcal{O}(\rho^2)}$ in Sec. V A).

For the sake of completeness we mention that the approach of Abrikosov, Dzyaloshinskii, Gorkov, Lifshitz, and Piaevskii (ADGLP) [1,12] is based on a semiphenomenological prescription for the vacuum energy which accounts for the restriction of the EM fluctuations to those which contain a strictly local *polarization operator*, $\sim\chi_{\rm eff}\bar{\bar{\mathbb{I}}}\delta^{(3)}(\vec{r}-\vec{r}')$. Hence, its content is similar to that of Schwinger's in the effective medium approximation but for the fact that it includes the contribution of long-wavelength bulk longitudinal modes,

$$\begin{aligned}&\mathcal{F}_{\rm ADGLP} \\ &= -\hbar{\rm Im}\left\{\int_0^\infty \frac{d\omega}{2\pi}\int\frac{d^3q}{(2\pi)^3}\ln\left[\frac{(k^2-q^2)^2}{\epsilon_{\rm eff}(\epsilon_{\rm eff}k^2-q^2)^2}\right]\right\},\end{aligned} \tag{86}$$

where $\epsilon_{\rm eff}$ is the effective dielectric constant. A critical analysis of it has been carried out by Bullough in Ref. [57].

## V. THE LEADING-ORDER LAMB SHIFT AND VACUUM ENERGY IN A RANDOM MEDIUM

At leading order, the free-space Lamb shift and Lamb energy are additive. Also, at leading order in $\rho$, $\mathcal{F}^{L}_{0} \simeq \rho\mathcal{E}^{LSh}_{0}$. The actual computation of $\mathcal{F}^{L}_{0}$,$\mathcal{E}^{LSh}_{0}$ has been carried out by a number of authors [16,29,34,58],

$$\mathcal{F}^{L}_{0} = \hbar\rho{\rm Im}\left\{\int_0^\infty\frac{d\omega}{2\pi}\ln\left[1-i\frac{\omega^3}{2\pi c^3}\alpha_0\right]\right\} \tag{87}$$

$$\simeq \rho\mathcal{E}^{LSh}_{0,\alpha_0} = -\hbar\rho{\rm Im}\left\{\int_0^\infty\frac{d\omega}{(2\pi)^2}i\frac{\omega^3}{c^3}\alpha\right\} \tag{88}$$

$$\simeq -\hbar\rho{\rm Re}\left\{\int_0^\infty\frac{d\omega}{(2\pi)^2}\frac{\omega^3}{c^3}\alpha_0\right\}. \tag{89}$$

Upon subtraction of the divergent free-electron radiative self-energy [34], $\mathcal{E}_e = \frac{e^2\hbar}{\pi m_e c^3}\int d\omega\,\omega$, the above integral presents a UV divergence which needs to be regularized. In the nonrelativistic approximation, the "natural choice" for the UV cutoff $\Lambda$ is that of the Compton wavelength of the electron such that $\Lambda^e_C = 2\pi c\lambda^e_C = m_e c^2/\hbar$. For a single oscillator with $\mu^2 = \frac{e^2\hbar}{2m_e\omega_0}$,

$$\mathcal{F}^{L}_{0} \simeq \frac{\rho}{3\pi}\alpha_f\frac{\hbar\omega_0}{m_e c^2}\ln\left[\frac{m_e c^2}{\hbar\omega_0}\right]\hbar\omega_0 \tag{90}$$

$$\simeq \frac{\rho}{2\pi}\ln\left[\frac{m_e c^2}{\hbar\omega_0}\right]\hbar\Gamma_0, \tag{91}$$

where $r_e = \frac{e^2}{4\pi\epsilon_0 m_e c^2}$ is the electron radius and $\alpha_f = \frac{e^2}{4\pi\epsilon_0\hbar c}$ is the fine-structure constant. Equation (90) equals Bethe's result when expressing the atomic energy in terms of the velocity of a bounded electron [29,59].[7] Had we computed Eq. (87) and set the wavelength cutoff at the electron radius, $\Lambda^e_r = c/r_e$, we would have obtained the classical result of Dowling [60] by considering the singularity around this value. However, this is inconsistent with the nonrelativistic approximation. The need of a cutoff just reflects our lack of knowledge of both the internal structure of the dipoles and the manner the EM field couples to the internal d.o.f..

At higher orders the Lamb shift is nonadditive. At $\mathcal{O}(\rho)$ we have,

$$\mathcal{E}^{LSh}_{\mathcal{O}(\rho)} = \hbar\rho{\rm Im}\left\{\int_0^\infty\frac{d\omega}{2\pi}\,\alpha\phi^{(1)}_{\alpha}(\omega)\right\}, \tag{92}$$

where the subscript $\alpha$ means that the renromalized porlarizabilities in $\phi^{(1)}(\omega)$ must be replaced with free-space ones in order to keep the order $\rho$ of $\mathcal{E}^{LSh}_{\mathcal{O}(\rho)}$. At this order, the corresponding $\phi$ factors read from Fig. 1(d),

$$\phi^{(1)}_{\alpha\perp,\parallel} = \int\frac{d^3q}{(2\pi)^3}\{-\rho\alpha[G^{(0)}_{\perp,\parallel}]^2 + \chi^{(2)}_{\alpha\perp,\parallel}G^{(0)}_{\perp,\parallel}/(\rho\alpha)\}. \tag{93}$$

[7] Also, a similar expression to that in Eq. (90) was obtained by Welton [25] from the variation of the nonrelativistic Compton scattering cross section due to the position fluctuations of the electron.

From the above equation we can write, in function of LFFs,

$$\mathcal{E}^{LSh}_{\mathcal{O}(\rho)} = \hbar\rho \mathrm{Im}\Bigg( \int_0^\infty \frac{d\omega}{2\pi}\alpha^2 \int \frac{d^3q}{(2\pi)^3}\big\{\chi^{(2)}_{\alpha\parallel}(q)/(\rho\alpha)^2 - 1 + 2k^2 G^{(0)}_\perp(q)\big[\chi^{(2)}_{\alpha\perp}(q)/(\rho\alpha)^2 - k^2 G^{(0)}_\perp(q)\big]\big\}\Bigg). \quad (94)$$

This is to show how the LFFs enter the Lamb shift at order $\rho$. Related to this fact, the authors of [61] have computed the effect of LFFs on the vdW forces on a single dipole in an Onsager (real) cavity using Eq. (92).

At leading order in $\alpha_0$, it is plain from Eqs. (63) and (64) that $\mathcal{F}^V_{\mathcal{O}(\rho^2)}|_{\alpha_0} = \rho\mathcal{E}^{LSh}_{\mathcal{O}(\rho)}|_{\alpha_0}/2$. From those equations the same simple relation holds at higher orders, $\mathcal{F}^V_{\mathcal{O}(\rho^m)}|_{\alpha_0} = \rho\mathcal{E}^{LSh}_{\mathcal{O}(\rho^{m-1})}|_{\alpha_0}/m$. This allows for an expansion of both $\mathcal{F}^V_{\mathrm{avg}}|_{\alpha_0}$ and $\mathcal{E}^{LSh}_{\mathrm{avg}}|_{\alpha_0}$ in $m$-body terms of order $(e^2\rho)^m$. However, beyond the approximations used there, no simple relation exists since any given order $\rho^m$ contains higher powers of $e^2$ due to entangled recurrent scattering and additional self-polarization corrections.

### A. Computation in the hard-sphere model

Except in free space, the actual computation of $\mathcal{E}^{LSh}_{\mathrm{avg}}$ and $\mathcal{F}^V_{\mathrm{avg}}$ is model dependent. Nonetheless, generic results can be obtained within the simplest analytical model. Let $h(r-\xi)$ be the two-point correlation function, $\xi$ being the correlation length. Generally, $h(r-\xi)$ can be modeled by the addition of three terms,

$$h(r-\xi) \simeq h^{\mathrm{ex.}}(r-\xi) + \rho^{-1}\delta^{(3)}(\vec{r}) + h^{\mathrm{ovd}}(r-\xi). \quad (95)$$

In this equation, $h^{\mathrm{ex.}}(r-\xi)$ accounts for the exclusion volume around each dipole and its precise form depends on the interaction potential between pairs of scatterers. It tends to $-1$ for $r \lesssim \xi$ and to zero for $r \gtrsim \xi$. Usual forms are those of a Lennard-Jones potential and a hard-sphere potential. $h^{\mathrm{ovd}}(r-\xi)$ takes account of the overdensity of first neighbors around a given dipole. It might be relevant for high-ordered media. The three-dimensional $\delta$ function stands for the self-correlation. $h^{\mathrm{ex.}}$ and the self-correlation functions are inherent in any molecular dielectric.

In order to make contact with previous approaches we further demand the existence of an effective medium for the frequency range of interest. That implies $\zeta \equiv k\xi \ll 1$ for $\omega \leqslant \omega_0$. For the sake of simplicity we neglect in first approximation both the overdensity and the self-correlation terms in $h(r-\xi)$. We show *a posteriori* that the latter is plainly justified while the former implies slight modifications in numerical prefactors.

Without much loss of generality we take a hard-sphere (hs) exclusion volume correlation function, $h^{\mathrm{ex.}}(r-\xi) = h^{\mathrm{hs}}(r-\xi) = -\Theta(r-\xi)$, which derives from the potential $U(\vec{R}^1,\ldots,\vec{R}^N) \to \infty$ if $|\vec{R}^i - \vec{R}^j| \leqslant \xi$, $i \neq j$, and 0 otherwise. The numerical factors of the calculations which involve near-field modes depend on the precise profile of $h^{\mathrm{ex.}}(r-\xi)$. On the contrary, radiative propagating modes are model independent.

The computation of $\phi^{(1,0)}$ in spatial coordinates is easier than that of Eq. (93) in Fourier space,

$$\phi^{(1,0)}_{\alpha,hs} = \mathrm{Tr}\left\{\int d^3r\, \bar{G}^{(0)}(\vec{r})(-k^4\rho\alpha)\bar{G}^{(0)}(\vec{r})[1-\Theta(r-\xi)]\right\}$$
$$= \frac{-k^3}{2\pi}\rho\alpha e^{2i\zeta}\left[\frac{1}{\zeta^3} - \frac{2}{\zeta^2}i - \frac{1}{\zeta} + \frac{1}{2}i\right] \quad (96)$$
$$\simeq \frac{-k^3}{2\pi}\rho\alpha\left[\frac{1}{\zeta^3} + \frac{1}{\zeta} + \frac{7}{6}i - \zeta\right], \quad \zeta < 1. \quad (97)$$

The decomposition into transverse and longitudinal components is given in Appendix A. Inserting Eq. (96) into Eq. (92) and integrating in $\omega$ we obtain,

$$\mathcal{E}^{LSh}_{\mathcal{O}(\rho)}|^{\mathrm{norec.}}_{\mathrm{hs}} \simeq \frac{-\rho\mu^2}{12\epsilon_0}\frac{\Gamma_0}{\omega_0}\left\{\left(\zeta_0^{-3} - \zeta_0^{-1}\right) + \frac{14}{3\pi}(5/6 - \gamma_{\mathcal{E}} - \ln[2\zeta_0])\right\}, \quad (98)$$

where $\gamma_{\mathcal{E}}$ is the Euler constant and, as for the computation of $\mathcal{F}^L_0$, the cut of the integrand at $\Lambda^e_r$ has been neglected. For simplicity, the integral has been expanded in powers of $\zeta_0 = k_0\xi \ll 1$ up to order zero, the leading-order term being that of London's potential [62]. In contrast to $\mathcal{F}^L_0$, the oscillating factor $e^{2i\zeta}$ serves as a natural UV cutoff at $\Lambda \simeq c/2\xi$.[8]

In the last equations the superscripts (1,0) signal that recurrent scattering terms have not been included. As advanced in Sec. III C, the condition $\zeta_0^3 \gg \Gamma_0/\omega_0 \sim k_0 r_e$, implicit also in Eq. (98), suffices to guarantee that recurrent scattering can be neglected in good approximation. In Eq. (B3) of Appendix B we give the expression for the complete series of recurrent scattering diagrams which amount to $\phi^{(1)}_\alpha$. Its expansion in powers of $\alpha$ and further integration in $\omega$ yields a series of the form

$$\mathcal{E}^{LSh}_{\mathcal{O}(\rho)}|_{\mathrm{hs}} = \rho\epsilon_0^{-1}\mu^2\frac{\Gamma_0}{\omega_0}\sum_{m=0} f^{LSh}_m(\zeta_0)(\Gamma_0/\omega_0)^{2m}. \quad (99)$$

The functions $f^{LSh}_m(\zeta_0)$ contain both negative and positive powers of $\zeta_0$ together with terms proportional to $\ln[(2(m+1)\zeta_0]$. Each order $m$ presents a wavelength cutoff at $4\pi(m+1)\xi$. For $m=0$, $f^{LSh}_0(\zeta_0)$ is readily identifiable from Eq. (98). The leading order term of a generic function $f^{LSh}_m(\zeta_0)$ is of the order of $\zeta_0^{-3(2m+1)}$. Therefore, $[f^{LSh}_{m+1}(\Gamma_0/\omega_0)^{2(m+1)}]/[f^{LSh}_m(\Gamma_0/\omega_0)^{2m}] \sim (\frac{\Gamma_0/\omega_0}{\zeta_0^3})^2$ and the convergence of the series is guaranteed by the aforementioned inequality, $\zeta_0^3 \gg \Gamma_0/\omega_0$. At the same time, this makes the neglect of recurrent scattering terms a good approximation.

Because each order in recurrent scattering carries an additional factor $\alpha_0^2$, the integration of $\mathcal{E}^{LSh}_{\mathcal{O}(\rho)}|_{\mathrm{hs}}$ yields for

[8] Making use of the property $\int_0^\infty d\omega \mathrm{Im}\{\phi(\omega)\} = \int_0^\infty d\omega\phi(i\omega)$ [7,48], all the integrals in $\omega$ are performed with the change of variables $u = -i\omega$. Since our computation refers to the ground state, resonant terms are absent.

nonrecurrent and recurrent terms, respectively,

$$\mathcal{F}^V_{\mathcal{O}(\rho^2)}\big|^{\text{norec.}}_{\text{hs}} \simeq \frac{-\rho^2\mu^2}{24\epsilon_0}\frac{\Gamma_0}{\omega_0}\left\{\left(\zeta_0^{-3}-\zeta_0^{-1}\right)+\frac{14}{3\pi}(5/6-\gamma_\varepsilon - \ln[2\zeta_0])\right\}, \tag{100}$$

$$\mathcal{F}^V_{\mathcal{O}(\rho^2)}\big|_{\text{hs}} = \rho^2\epsilon_0^{-1}\mu^2\frac{\Gamma_0}{\omega_0}\sum_{m=0}\frac{1}{m+2}f_m^{LSh}(\zeta_0)(\Gamma_0/\omega_0)^{2m}. \tag{101}$$

The numerical constants in Eq. (98) depend on the specific profile of $h^{\text{ex.}}$. Nonetheless, the order and the sign of the terms are generic. Hence, the script $hs$ can be dropped from Eqs. (99) and (101) since those expansions are not constrained to any particular model. As a matter of fact, the addition to Eq. (96) of the terms in Eq. (A9) which account for the overdensity two-point correlation function, $h^{\text{ovd}}(r) = C\xi\delta^{(1)}(r-\xi)$, just modifies slightly the numerical prefactors of the terms in Eqs. (98) and (100), but neither their order nor their sign.

It is worth mentioning that, although at a given order $\rho^n$ the leading contribution comes from nonrecurrent terms, there are recurrent terms in orders $\rho^s, 0<s<n$, which are of order $(\rho\xi^3)^{-(n-s)} \geqslant 1$ greater than the leading order nonrecurrent term of the order $\rho^n$.

### B. Discrepancy between $\mathcal{F}^V_{\text{avg}}|_{\text{rad}}$ and $\mathcal{F}^V_{\text{Sch.}}$

We investigate to which extent the Schwinger vacuum energy of a bulk effective medium is a good approximation to the actual vacuum energy of radiative modes. We compare their relation up to order $\rho^3$. For the reasons given in the previous section recurrent scattering is negligible and the effective susceptibility is well approximated by that of a Maxwell-Garnett (MG) dielectric. An MG dielectric is characterized by the fact that the only relevant correlation function is that of an exclusion volume, $h^{\text{ex.}}(r-\xi)$. In the electrostatic-long-wavelength limit it is proven that the qc approximation is exact [63] [see Fig. 3] and the effective susceptibility is independent of the precise form of $h^{\text{ex.}}(r-\xi)$. In particular, we can use the results of the hs model without loss of generality. According to this, the electrical susceptibility is the sum of a geometrical series of ratio $\chi^{(2)}_{\text{MG}}/\rho\alpha = \rho\alpha/3$, in which only the bare longitudinal propagator and free-space polarizabilities enter. It yields

$$\chi_{\text{MG}} = \frac{\rho\alpha}{1-\chi^{(2)}_{\text{MG}}/\rho\alpha} = \frac{\rho\alpha}{1-\rho\alpha/3}, \tag{102}$$

and from here and neglecting further renormalization on $\alpha$ for the reasons given above, the effective refractive index is $n \simeq 1+\rho\alpha/2+(\rho\alpha)^2/24+\cdots$. It has been already seen that $\mathcal{F}^V_{\text{Sch.}}$ contains the free-space Lamb energy. At $\mathcal{O}(\rho^2)$, by inserting the series of $n$ in Eq. (83) we get

$$\mathcal{F}^V_{\text{Sch.}}\big|^{\mathcal{O}(\rho^2)} \simeq -\frac{7\hbar}{48\pi^2}\text{Re}\left\{\int_0^\infty d\omega\, k^3(\rho\alpha)^2\right\}. \tag{103}$$

Next we turn to the hs model for the microscopical computation. The $\zeta$-independent terms of $\phi^{(1,0)}_{\alpha,hs}$ in Eq. (97) amount to the energy of radiative propagating modes (rad) within $\mathcal{F}^V_{\mathcal{O}(\rho^2)}|_{\text{rad}}$ upon integration in $\omega$. We show in Appendix A that, out of them, $-i\frac{k^3}{2\pi}\frac{5}{6}\rho\alpha$ comes from the bulk transverse propagator in $2\phi^{(1,0)}_\perp$[9] [Eqs. (A1), (A3), and (A7)]. The remaining $-i\frac{k^3}{2\pi}\frac{\rho\alpha}{3}$ comes from the bare transverse propagator in the susceptibility function of $\phi^{(1,0)}_\parallel$ [Eqs. (A3) and (A8)]. Hence, we can write

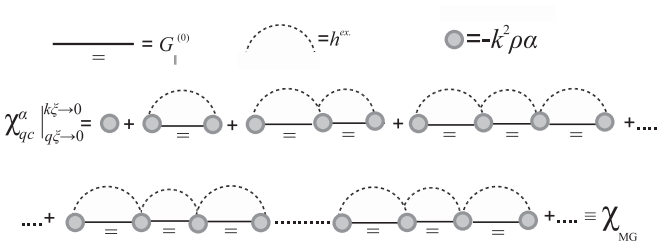


FIG. 3. Diagrammatic representation of the 1PI processes which amount to the MG susceptibility. Only the exclusion volume two-point correlation function, $h^{\text{ex.}}(r-\xi)$, is relevant and self-correlations are disregarded. In the electrostatic-long-wavelength limit, $k\xi, q\xi \to 0$, the qc approximation is exact.

$$\mathcal{F}^V_{\mathcal{O}(\rho^2)}\big|_{\text{rad}} \simeq -\frac{7\hbar}{48\pi^2}\text{Re}\left\{\int_0^\infty d\omega\, k^3(\rho\alpha)^2\right\}, \tag{104}$$

and so $\mathcal{F}^V_{\text{Sch.}}|^{\mathcal{O}(\rho^2)} \simeq \mathcal{F}^V_{\mathcal{O}(\rho^2)}|_{\text{rad}}$. However, this equality is accidental. To see this, it is necessary to express $\mathcal{F}^V_{\text{avg}}$ and $\mathcal{F}^V_{\text{Sch.}}$ in comparable terms. To this aim we use the expression of Eq. (64) for $\mathcal{F}^V_{\text{avg}}$ but with $\alpha$ instead of $\alpha_0$ there, and the extended version of $\mathcal{F}^V_{\text{Sch.}}$ which incorporates spatial dispersion [17], $\mathcal{F}^V_{\text{Sch.}}|^{\text{ext}}$. Inserting Eq. (36) into Eq. (81) for $\mathcal{F}^V_{\text{Sch.}}$ but with spatial dispersion we have

$$\mathcal{F}^V_{\text{Sch.}}|^{\text{ext}} = -2\hbar\text{Im}\left\{\int_0^\infty\frac{d\omega}{2\pi}\int\frac{d^3q}{(2\pi)^3} \times \ln\left[1-\rho\tilde{\alpha}\sum_{m=0}k^2\mathcal{G}^{(m)}_\perp(q)\right]\right\}, \tag{105}$$

Further, we expand the latter equation up to order $\rho^3$,[10]

$$\mathcal{F}^V_{\text{Sch.}}|^{\text{ext}} \simeq \mathcal{F}^{(1)}_V + \hbar\text{Im}\left(\int_0^\infty\frac{d\omega}{2\pi}\left\{\rho^2\left(2\alpha\phi^{(1)}_{\alpha\perp}/\rho + \alpha^2k^4\int\frac{d^3q}{(2\pi)^3}[G^{(0)}_\perp]^2 + i\alpha^2\frac{k^3}{2\pi}\phi^{(1)}_\alpha/\rho\right)\right. \tag{106}$$

[9] A comment is in order here to amend some erroneous interpretations in Ref. [27]. In the first place, the distinction between coherent and incoherent radiation carried out in Sec. IV of Ref. [27] is erroneous. While Eq. (54) of Ref. [27] is correct, its equivalence with Eqs. (46), (48) and (50) of Ref. [27], and with the addition of Eqs. (42) and (44) of Ref. [27] is not. Consequently, the conclusion that only one LFF appears in the expression of the coherent radiation is incorrect. The correct calculations will be published somewhere else [64].

[10] The expansion of the logarithms in Eq. (105) yields a series of $q$ integrals whose integrands are products of powers of polarization propagators of the form $[\mathcal{G}^{(m)}_{\perp,\parallel}]^s$. Each factor $\mathcal{G}^{(m)}_{\perp,\parallel}$ is an $m$-scattering loop and the sum of the exponents, $s$, is the loop order; for example, $G^{(0)}_{\perp,\parallel}[\mathcal{G}^{(2)}_{\perp,\parallel}]^2\mathcal{G}^{(1)}_{\perp,\parallel}$ is a four-loop term made of one free-space, one single-scattering, and two double-scattering loops.

$$+\rho^3\Bigg[2\alpha\phi^{(2,0)}_{\alpha\perp}/\rho^2+\frac{2}{3}\alpha^3k^6\int\frac{d^3q}{(2\pi)^3}[G^{(0)}_\perp]^3 +2\alpha^2k^4\int\frac{d^3q}{(2\pi)^3}G^{(0)}_\perp\mathcal{G}^{(1,0)}_{\alpha\perp}/\rho+\mathcal{O}(\alpha^4)\Bigg]\Bigg\}\Bigg). \quad (107)$$

On the other hand, for the evaluation of the radiative modes of $\mathcal{F}^V_{\rm avg}$ we use the expressions for the MG $\phi$ factors and effective transverse propagator restricted to radiative modes [27],

$$\mathcal{G}^{\rm MG}_\perp(q)=\mathcal{L}_{\rm LL}G^{\rm eff}_{{\rm MG}\perp}(q),\quad \phi_{\rm MG}=-i\frac{k^3}{2\pi}\mathcal{L}^2_{\rm LL}n, \quad (108)$$

$$2\phi_{{\rm MG}\perp}=-i\frac{k^3}{2\pi}\mathcal{L}^2_{\rm LL}n,\quad \phi_{{\rm MG}\parallel}=\phi_{\rm MG}-2\phi_{{\rm MG}\perp}, \quad (109)$$

with $\mathcal{L}_{\rm LL}=\frac{\chi_{\rm MG}+3}{3}$ and $G^{\rm eff}_{{\rm MG}\perp}(q)=[(\chi_{\rm MG}+1)k^2-q^2]^{-1}$.

From Eq. (106) we have that the first term there contains only the transverse modes in $\mathcal{F}^V_{\mathcal{O}(\rho^2)}$. Its restriction to radiative modes amounts to $-\frac{5\hbar}{24\pi^2}{\rm Re}\{\int_0^\infty d\omega\, k^3(\rho\alpha)^2\}$. On the other hand, the second term of Eq. (106) amounts to $\frac{\hbar}{16\pi^2}{\rm Re}\{\int_0^\infty d\omega\, k^3(\rho\alpha)^2\}$. Adding up the last two quantities we obtain Eq. (103), which equals $\mathcal{F}^V_{\mathcal{O}(\rho^2)}|_{\rm rad}$, even though no longitudinal terms enter the Schwinger result. The reason for this equivalence is accidental, since the $\mathcal{O}(\rho^2)$ radiative term of the difference $\mathcal{F}^V_{\rm avg}|^\perp-\mathcal{F}^V_{\rm Sch.}|^{\rm ext}$ in Eq. (85) is equivalent, by reciprocity [27], to the longitudinal term in $\mathcal{F}^V_{\mathcal{O}(\rho^2)}|_{\rm rad}$ but with opposite sign. This relation is not model dependent. Note also that by incorporating spatial dispersion in $\mathcal{F}^V_{\rm Sch.}|^{\rm ext}$, the term $2\phi^{(1,0)}_\perp$ contains only one half of the nonpropagating term in Eq. (97) proportional to $\zeta_0^{-1}$. The rest of the near-field terms in $\mathcal{F}^V_{\mathcal{O}(\rho^2)}$ are disregarded in $\mathcal{F}^V_{\rm Sch.}|^{\rm ext}_{\mathcal{O}(\rho^2)}$.

Next we show that the accidental coincidence of $\mathcal{F}^V_{\rm Sch.}|^{\rm ext}_{\mathcal{O}(\rho^2)}$ and $\mathcal{F}^V_{\mathcal{O}(\rho^2)}|_{\rm rad}$ breaks down at $\mathcal{O}(\rho^3)$. Either by expanding the integrand of Eq. (83) or by direct integration of the $\mathcal{O}(\rho^3)$ terms in Eq. (107) we get

$$\mathcal{F}^V_{\rm Sch.}|^{\rm ext}_{\mathcal{O}(\rho^3)}\simeq-\frac{17\hbar}{288\pi^2}{\rm Re}\Bigg\{\int_0^\infty d\omega\, k^3(\rho\alpha)^3\Bigg\}, \quad (110)$$

while the insertion of Eq. (108) into Eq. (64) yields

$$\mathcal{F}^V_{\mathcal{O}(\rho^3)}\big|_{\rm rad}\simeq-\frac{17\hbar}{144\pi^2}{\rm Re}\Bigg\{\int_0^\infty d\omega\, k^3(\rho\alpha)^3\Bigg\}=2\mathcal{F}^V_{\rm Sch.}|^{\rm ext}_{\mathcal{O}(\rho^3)}. \quad (111)$$

## VI. THE LAMB SHIFT FROM SCHWINGER'S BULK EFFECTIVE MEDIUM VACUUM ENERGY

During the last decade studies on the effects of a dielectric environment on the enhancement and inhibition of decay rates and on the frequency shifts have revealed the relevance of LFFs and near-field contributions [27,44,65–67]. In these phenomena the microscopic structure matters, even as considering at the lowest order the effect of a dilute medium. The reason why the bulk effective medium approximation does not yield the correct result for the decay rate has been already discussed in Ref. [27]. In that approximation, unphysical divergences appear as considering dispersive media. It is our aim in this section to discuss why that approximation is not suitable either to study the Lamb shift.

To this respect, Schaden, Spruch, and Zhou (SSZ) [35] have computed the Lamb shift at leading order in $\rho$ from Eq. (83) of Schwinger's vacuum energy of a bulk effective medium. In turn, this is the free-space Lamb shift. For the reasons explained in Sec. IV B Schwinger's approach yields the correct result at leading order for $\chi_{\rm eff}\simeq\rho\alpha_0$. Nonetheless, the authors of [35] have applied a more general procedure consisting of computing the Lamb shift out of the variation of $\mathcal{F}^V_{\rm Sch.}$ with respect to "small" variations of a background refractive index. That is, adopting our nomenclature, they have used the variation of Eq. (80) together with the result of Eq. (84) and $\delta\chi_{\rm eff}=2n\Delta n$,

$$\mathcal{E}^{LSh}_{\rm SSZ}=\rho^{-1}\Delta_n\mathcal{F}^V_{\rm Sch.}=\frac{-\hbar}{4\pi^2c^3\rho}{\rm Re}\Bigg\{\int_0^\infty d\omega\,\omega^3n\Delta\chi_{\rm eff}\Bigg\}. \quad (112)$$

For a dilute medium, $n\simeq1$, $\Delta\chi_{\rm eff}=\rho\alpha_0$, and

$$\mathcal{E}^{LSh}_{\rm SSZ}=\frac{-\hbar}{4\pi^2c^3\rho}{\rm Re}\Bigg\{\int_0^\infty d\omega\,\omega^3\rho\alpha_0\Bigg\}, \quad (113)$$

which is nothing but the free-space Lamb shift. This is analogous to the computation carried out by Feynman, Power, and Milonni [16,33,34] in the limit $\rho\mathcal{V}\to1$ in which the medium consists of a single dipole in free space, $\mathcal{V}$ being the sample volume. More specifically, the authors of [35] have computed the difference of the free-space Lamb shift between two dielectric states, $I$ and $II$, with $\Delta\chi_{\rm eff}=\rho(\alpha_0^{II}-\alpha_0^I)$.

The computation of Milonni, Schaden, and Spruch (MSS) in Ref. [4] combines elements of [16,33–35]. That is, while in Ref. [4] the variation of the bulk energy density is calculated between two different states of molecular dielectrics as in Ref. [35], the difference between the states is given by the difference on the polarizability of only one of the dipoles as in Refs. [16,33,34]. Under the assumption that the dipole in question is randomly placed and the medium behaves as a continuum of refractive index $n\gg\rho\alpha_0^{I,II}/2$, the authors take $\Delta\chi_{\rm eff}=\rho(\alpha_0^{II}-\alpha_0^I)$ and, using Eq. (112), they get

$$\mathcal{E}^{LSh}_{\rm MSS}=\frac{-\hbar}{4\pi^2c^3\rho}{\rm Re}\Bigg\{\int_0^\infty d\omega\,\omega^3n\rho\big(\alpha_0^{II}-\alpha_0^I\big)\Bigg\}. \quad (114)$$

Likewise, for the Lamb shift due to the presence of the background medium (BGM), $\mathcal{E}^{LSh}_{\rm MSS}|^{I-II}_{\rm BGM}=-\frac{\hbar}{4\pi^2}{\rm Re}\{\int d\omega(\omega^3/c^3)(n-1)(\alpha_0^{II}-\alpha_0^I)\}$.

The authors of [4] had already warned that the derivation of the Lamb shift this way might need to be corrected by LFFs in highly ordered systems. We have proved in Sec. V B that this is indeed the case under any circumstance, no matter the degree of order. As was mentioned there, spatial dispersion and longitudinal modes are to be added to Schwinger's formula in order to obtain the correct result. Even if the host medium can be treated as a continuum, the microscopical calculation of the Lamb shift on the simplest geometries for the embedding of a dipole contains LFFs. At leading order, near-field electrostatic and other radiative factors enter the Lamb shift through the terms of $\phi^{(1)}$ in $\mathcal{E}^{LSh}_{\mathcal{O}(\rho)}$. For instance, for a small Onsager cavity (Ons.), those terms are the ones in Eq. (97) with $\xi$ being the

radius of the cavity and $k\xi < 1$ [10,61,66,67]. Thus, instead of $\mathcal{E}^{LSh}_{\rm MSS}|^{I-II}_{\rm BGM}$, we obtain at leading order in $(n-1)$[11]

$$\mathcal{E}^{LSh}_{\mathcal{O}(n-1)}|^{I-II}_{\rm Ons.} \simeq \frac{7}{3}\mathcal{E}^{LSh}_{\rm MSS}|^{I-II}_{\rm BGM} - \frac{\hbar}{2\pi^2}{\rm Im}\left\{\int d\omega(n-1) \times \left(\alpha_0^{II}-\alpha_0^{I}\right)\left[\frac{1}{\xi^3}+\frac{\omega^2}{c^2\xi}\right]\right\}. \quad (115)$$

Note that the Lamb shifts of Eqs. (114) and (115) are those referred to as vdW shifts in the literature [7–9] since they account for the interaction of a foreign dipole with a host background medium.

## VII. CONTINUUM APPROACH TO THE BINDING ENERGY OF AN EFFECTIVE MEDIUM

In this section we investigate the possibility of quantifying, at least partially, the binding energy through optical observations. This is motivated by the conjecture raised in the Introduction on the correspondence between the shift in the spectrum of the dielectric constant and the EM binding energy. Against this conjecture we have that there does not exist *a priori* a simple relation between the density of states for emission [27], $\mathcal{N}^{\rm emiss} = -2{\rm Im}\{\phi\}/(\pi\omega)$, and that for the vacuum energy, $\mathcal{N}^{V}_{\rm avg} = \frac{-2}{\omega}\int_0^{\alpha_0}\delta\alpha_0'\frac{\phi}{1+\alpha_0'\phi}$. In favor, we have already found that $\mathcal{E}^{LSh}_{\rm avg}$ can be expressed as a function of the electrical susceptibility only Eq. (60). Moreover, in the qc approximation $\mathcal{F}^{V}_{\rm qc}$ is a function of $\chi_{\perp,\parallel}(q)$ [Eqs. (72)–(76)]. Further, the qc approximation becomes exact in the long-wavelength limit of the effective medium theory of an MG dielectric. For this reason we concentrate on the MG model. Nonetheless, we must bear in mind that the MG model neglects recurrent scattering in $\chi_{\rm MG}$, which might be relevant, even in the long-wavelength limit, in highly dense media for which $\xi^3 \sim \rho^{-1}$ (cf. [45,68] and Appendix B).

### A. The Lorentz-Lorenz shift

The problem of the binding energy of an effective medium has been addressed by Bullough and Obada [13] in a molecular crystal and by ourselves in Ref. [27]. In the latter reference it has been found that in the long-wavelength limit $\mathcal{F}^{V}_{\rm qc}$ can be split into the energy of transverse bulk modes and that of LFFs and longitudinal bulk modes according to

$$\mathcal{F}^{V}_{\rm MG} = \mathcal{F}^{V}_{\rm Sch.} + \Delta\mathcal{F}^{V}_{\rm MG}. \quad (116)$$

Here, $\mathcal{F}^{V}_{\rm Sch.}$ is given in Eq. (83) and

$$\Delta\mathcal{F}^{V}_{\rm MG} = -\hbar\int_0^\infty\frac{d\omega}{2\pi}\int\frac{d^3q}{(2\pi)^3}{\rm Im}\left\{\ln\left[\frac{\chi^3_{\rm MG}[\alpha_0]}{(\rho\alpha_0)^3\epsilon_{\rm MG}[\alpha_0]}\right]\right\}, \quad (117)$$

where $\chi_{\rm MG}$ are functions of $\alpha_0$ in the qc approximation. An identical expression has been reported by Bullough and Obada [13], who have interpreted $\Delta\mathcal{F}^{V}_{\rm MG}$ as the electrostatic binding energy. However, the decomposition in Eq. (116), the identification of $\mathcal{F}^{V}_{\rm Sch.}$ with the radiative energy, and the identification of $\Delta\mathcal{F}^{V}_{\rm MG}$ with the electrostatic energy are all questionable.

Regarding the radiative energy, it was found in Sec. V B that not all the radiative energy is given by $\mathcal{F}^{V}_{\rm Sch.}$. In the first place, it was shown in Ref. [27] that $\mathcal{L}_\perp(q)$ induces radiation on the surrounding dipoles around an emitter, acting as a mediator of nonradiative energy transfer. For this to be the case, $\mathcal{L}_\perp(q)$ must appear coupled to $G_\perp(q)$ prior to integration in $q$. Clearly, the decomposition of Eq. (116) precludes this. On top of that, there is also energy in $\mathcal{F}^{V}_{\parallel,avg}$ carried by indirect radiation modes which are missing in $\mathcal{F}^{V}_{\rm Sch.}$. It comes from the transverse modes within $\chi_\parallel(q)$ which are neglected in the long-wavelength limit [e.g., the term $-i\frac{k^3}{2\pi}\frac{\rho\alpha}{3}$ mentioned before Eq. (104) belongs to indirect radiation].

Regarding the electrostatic energy, it is the EM vacuum energy obtained in the electrostatic limit, $c\to\infty$. Following [13], such a limit must be taken prior to the long-wavelength approximation, making radiative modes vanish. On the other hand, for $q\xi\to 0$ the transverse susceptibility equals the longitudinal one because only electrostatic modes enter $\chi_\perp(q\xi=0)$. However, as argued in the previous paragraph, $\mathcal{L}_\perp(q\xi=0)$ has a physical meaning when coupled to $G_\perp$ acting as an inductor of radiation. Therefore, if radiation is precluded for $c\to\infty$, $\mathcal{G}_\perp^{c\to\infty}$ does not contribute to the electrostatic energy and neither do the transverse LFFs. As a result, transverse LFFs should not enter the vacuum energy in the electrostatic-long-wavelength approximation despite their presence in Eq. (117) for $q\xi\to 0$. Thus, we find that the energy of electrostatic modes in the long-wavelength limit is

$$\mathcal{F}^{V}_{\rm eff}|_{\rm stat} = -\hbar\int_0^\infty\frac{d\omega}{2\pi}\int\frac{d^3q}{(2\pi)^3}{\rm Im}\left\{\ln\left[\frac{\chi_{\rm MG}[\alpha_0]}{\epsilon_{\rm MG}[\alpha_0]\rho\alpha_0}\right]\right\}, \quad (118)$$

which equals $\Delta\mathcal{F}^{V}_{\rm MG}$ but for the absence of the two transverse LFFs.

Next, let us make explicit calculations in the MG model with bare polarizabilities. According to that model, the electrical susceptibility is the sum of a geometrical series of ratio $\chi^{(2)}_{\rm MG}/\rho\alpha_0 = \rho\alpha_0/3$, in which only the bare longitudinal propagator enters. Neglecting self-polarization corrections in Eq. (102) we get,

$$\chi_{\rm MG}[\alpha_0] = \frac{\rho\frac{2\mu^2\omega_0}{3\hbar\epsilon_0}}{\omega_0^2-\omega^2-\rho\frac{2\mu^2\omega_0}{9\hbar\epsilon_0}}. \quad (119)$$

The question we aim to address is whether the shift of the resonant frequency of $\chi_{\rm MG}$ with respect to that of $\alpha_0$ has any counterpart in the binding energy of a molecular dielectric. In the particular case of an infinite MG dielectric that frequency shift is known as the Lorentz-Lorenz (LL) shift, which is

[11]The approximate expression of $\phi^{(1,0)}_{\alpha,\rm hs}$ for $\zeta<1$ in Eq. (97) has been used for simplicity since the exponential factor in the integrand of Eq. (96) suppresses frequencies greater than $c/(2\xi)$ and the singularities of the integrand locate around $\omega_0 \ll c/(2\xi)$. The $\xi$-dependent terms may be more relevant than the radiative ones if the host medium is highly dissipative within the frequency range of integration.

observable in optics experiments [21,69],[12]

$$\Delta\omega_{\rm LL} = \omega_0\left[\sqrt{1-\rho\frac{2\mu^2}{9\hbar\epsilon_0\omega_0}}-1\right] = -\rho\frac{\mu^2}{9\hbar\epsilon_0}+\mathcal{O}(\rho^2). \tag{120}$$

The associated energy shift is referred to as collective Lamb shift for obvious reasons [11,22]. The LL shift is clearly the result of the renormalization of $\chi_{\rm MG}$ in Eq. (119) by the electrostatic modes within $\chi^{(2)}_{\rm MG}$ and with all the dipoles in the ground state. Therefore, its energetic counterpart must find in the formula for $\mathcal{F}^V_{\rm eff}|_{\rm stat}$. Since no self-polarization corrections enter the integrand of Eq. (118) at all, it does not contain imaginary terms and we can write it as a sum over modes. That is, upon using the regularization $\int \frac{d^3q}{(2\pi)^3} = \rho$, the $\omega$ integral reduces to the sum of the poles minus the sum of the zeros of the factors within the logarithm. Using the formulas of an MG dielectric we obtain

$$\mathcal{F}^V_{\rm eff}\big|_{\rm stat} = \rho\frac{-\hbar\omega_0}{2}\left[\sqrt{1+\rho\frac{4\mu^2}{9\hbar\epsilon_0\omega_0}}-1\right]. \tag{121}$$

At leading order in $\rho$, $\mathcal{F}^V_{\rm eff}|_{\rm stat} \simeq \rho\hbar\Delta\omega_{\rm LL}$ holds. Because, by assumption, no electrostatic-long-wavelength modes renormalize the single-particle polarizability alone, we infer that $\mathcal{F}^V_{\rm eff}|_{\rm stat}$ is the binding energy of collective d.o.f. due to pairwise long-ranged electrostatic interactions, and so is the Lorentz-Lorenz shift. Had we started with the renormalized single-particle polarizabilities in Eq. (119), the resonant frequency $\omega_0$ would have been replaced by its renormalized value, which derives from the Lamb shift of the individual dipoles. Therefore, the collective Lamb shift of an effective medium shows up in addition to the Lamb shift of the individual dipoles although the former derives from the integration of the latter upon taking the long-wavelength limit. When sample finite size effects are considered, it has been computed by Friedberg *et al.* [20] and verified experimentally in Ref. [21] that the collective Lamb shift incorporates a surface term referred to as the cooperative Lamb shift, which accounts for the mutual dipole interactions mediated by the radiative modes reflected at the boundaries. The cooperative Lamb shift is of the same order as the LL shift, although it is not associated to the poles of the dielectric constant but to the transmition and reflection coefficients of the medium. To this respect, it seems more closely related to $\mathcal{F}^V_{\rm Sch.}$. However, by inserting Eq. (119) into that of $\mathcal{F}^V_{\rm Sch.}$ in Eq. (83) and cutting off the frequency integral at $\omega_{\rm max} = c\rho^{1/3}$ for consistency with the regularization of the momentum divergency in $\mathcal{F}^V_{\rm eff}|_{\rm stat}$, one obtains for the Schwinger bulk energy of an MG dielectric,

$$\mathcal{F}^{\rm MG}_{\rm Sch.} \simeq \frac{9}{2\pi^2}\frac{\omega_0\rho^{2/3}}{c}\hbar\Delta\omega_{\rm LL}. \tag{122}$$

This quantity is of the order of $\rho^{-1/3}\omega_0/c$ times that of $\mathcal{F}^V_{\rm eff}|_{\rm stat}$, which makes it negligible for dilute media where $c\rho^{1/3}/\omega_0 \gg 1$.

### B. Discussion on the effective medium approximation

Our microscopical approach together with the calculations of the previous section allow us to answer two of the questions posed by Bullough in Ref. [57]. The first one was whether the knowledge of the refractive index spectrum is sufficient to estimate the vacuum energy of long-wavelength fluctuations. Our results mean that neither the radiative nor the electrostatic energy can be correctly accounted for this way. The second question was concerned with the role of the Lorentz field in the continuum approach to the binding energy. We have proved, by strict isolation of the vacuum energy of long-wavelength longitudinal modes, that the Lorentz field gives rise to the LL shift and hence to $\mathcal{F}^V_{\rm eff}|_{\rm stat}$.

Regarding the radiative energy, we conclude from the previous sections that the knowledge of the spectrum of the refractive index is insufficient for the quantification of the total radiative energy. In the first place, part of the radiation is not accounted for in the (bulk) spectrum of $\mathcal{F}^V_{\rm Sch.}$. Second, the microscopical approach of Sec. V and Appendix A shows that the transverse propagators within the LFF terms amount to radiative modes which are disregarded in the long-wavelength limit of Eqs. (74)–(76).

Regarding the electrostatic energy, we first observe that the relation $\mathcal{F}^V_{\rm eff}|_{\rm stat} \simeq \rho\hbar\Delta\omega_{\rm LL}$ must not be interpreted as a quantitative estimate of the binding energy but as a result of consistency. That is, this equation means that if only the longitudinal long-wavelength modes which enter the renormalization of $\chi_{\rm MG}$ are considered in the computation of the binding energy of clusters, the equivalence must hold. On the contrary, it must not be interpreted as a reliable estimation of the electrostatic binding energy. To see this it suffices to verify from Eq. (100) that $\mathcal{F}^V_{\mathcal{O}(\rho^2)}|^{\rm norec.}_{\rm hs}$ does not contain terms of the order of Eq. (121). In particular, all the $\zeta_0$-independent terms in Eq. (100) have a radiative origin and are of the order of $\Gamma_0/\omega_0$ less than that of $\mathcal{F}^V_{\rm eff}|_{\rm stat}$. Moreover, since generally $\zeta_0^3 \gg \Gamma_0/\omega_0$, we have $\mathcal{F}^V_{\mathcal{O}(\rho^2)}|^{\rm norec.}_{\rm hs} \ll \mathcal{F}^V_{\rm eff}|_{\rm stat}$. We conclude that, contrarily to that suggested in Refs. [13,57], the continuum approximation is not even sufficient to estimate the orders of magnitude of electrostatic binding energies.[13] Therefore, while the Lamb shift of the individual dipole polarizabilities can be attributed to shifts in the internal energy levels, the same interpretation is not applicable to the collective Lamb shift with respect to the binding energy of the dielectric.

It is also remarkable that while the free-space Lamb shift can be computed out of $\mathcal{F}^V_{\rm Sch.}$ [16,33–35] and so involves the energy of long-wavelength transverse modes only, the collective Lamb shift of a homogeneous effective medium

[12]Despite of the fact that there is experimental evidence for $\Delta\omega_{\rm LL}$, it is also known that the MG model fails to provide accurate results close the resonance. The reason being that it does not incorporate the dominant recurrent scattering [45].

[13]The sum over modes of Eq. (117) yields $-\rho\hbar\omega_0[3-2\sqrt{3/(n_0^2+2)}-n_0\sqrt{3/(n_0^2+2)}]/2$ [13] upon use of the regularization $\frac{\int d^3q}{(2\pi)^3}=\rho$ and $n_0=\sqrt{1+\chi_{\rm MG}(\omega=0)}$. Equivalently, for a generic Lorentzian dielectric constant with field strength factor $f$, it yields $\rho f^2\hbar\omega_0/24$ [27].

accounts for the energy of long-wavelength longitudinal modes instead.

## VIII. SUMMARY

We have carried out a microscopical study of the EM vacuum energy of an isotropic and homogeneous molecular dielectric made of a random distribution of two-level atomic dipoles.

Before considering statistical averages, we have shown that for a specific configuration $m$, $\mathcal{F}^V_m$ can be expressed either as a function of the dipole fluctuations Eq. (53), as a function of the source EM field fluctuations Eq. (54), or as a combination of both Eq. (55).

When statistical averages are taken, the Lamb shift is a function of the electrical susceptibility only Eq. (60), $\chi_{\perp,\parallel}$, and hence can be computed out of optical observations. On the contrary, the total vacuum energy is not, and only a cluster expansion is possible Eq. (68). Only in the quasicrystalline approximation is it possible to give a closed expression for the vacuum energy in terms of $\chi^{\rm qc}_{\perp,\parallel}$ [Eqs. (74)–(76)].

Using a hard-sphere model we have discussed to what extent recurrent scattering terms contribute to the total vacuum energy. Except for the free-space Lamb energy, which needs a UV cutoff, no other divergences either in frequency or momentum space show up in the rest of the vacuum energy. In momentum space, LFFs kill the short distance divergences in the same manner they do in the spectrum of emission [27,67]. In frequency space, the UV divergences of retarded modes are exponentially suppressed by a natural cutoff at the wavelength of the order of the correlation length. On the contrary, both momentum and frequency divergences show up in the vacuum energy of a bulk effective medium which is a function of the effective Dyson propagator only and contains no LFFs. For this reason we interpret that the LFFs play the role of spectral functions with respect to the spectrum of bulk modes in the sense introduced by Ford [70].

Related to the last issue, the Schwinger approach to the vacuum energy of a bulk effective medium is, in general, insufficient to compute the Lamb shift out of variations of $\mathcal{F}^V_{\rm Sch.}$, which contradicts the result in Ref. [35] (Secs. V B and VI). $\mathcal{F}^V_{\rm Sch.}$ does not account for the total vacuum energy and contains artificial divergences due to the absence of LFFs. Nevertheless, $\mathcal{F}^V_{\rm Sch.}$ is sufficient to study the retarded Casimir forces between macroscopic dielectrics [52].

By evaluating the vacuum energy of the longitudinal long-wavelength modes of a Maxwell-Garnett dielectric we have obtained that $\mathcal{F}^V_{\rm eff}|_{\rm stat} \simeq \rho\hbar\Delta\omega_{\rm LL}$, which is readily identifiable with the collective Lamb shift in a homogeneous effective medium. This relation has been interpreted as a result of the consistency between the spectrum of $\chi_{\rm MG}$ and the energy of the modes involved in the construction of $\chi_{\rm MG}$. Nonetheless, it has been concluded that neither the spectrum of the refractive index nor the shift in the resonance of the dielectric constant are sufficient either to quantify the energy of radiative modes or to estimate the electrostatic binding energy of a molecular dielectric. In particular, the collective Lamb shift of the dielectric constant is not readily identifiable with a binding energy.


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## APPENDIX A: DECOMPOSITION OF $\phi^{(1,0)}$ AND COMPUTATION OF $\phi^{(1,0)}_{\rm ovd}$

We write Eq. (93) as a spatial space integral in order to show the radiative and electrostatic nature of the propagators involved. To this aim, we make use of the fact that $\bar{G}^{(0)}_{\rm rad.}(r)$ is totally transverse and $\bar{G}^{(0)}_{\rm stat.}(r)$ is totally longitudinal in Fourier space,

$$2\phi^{(1,0)}_{\alpha\perp} = -k^2\rho\alpha{\rm Tr}\Big\{\int d^3r\big[\bar{G}^{(0)}_{\rm rad.}(r)+\bar{G}^{(0)}_{\rm stat.}(r)\big]\cdot\bar{G}^{(0)}_{\rm rad.}(r)[1+h(r-\xi)]\Big\} = -k^2\rho\alpha\int d^3r$$

$$\times{\rm Tr}\big\{\bar{G}^{(0)}_{\rm rad.}(r)\cdot\bar{G}^{(0)}_{\rm rad.}(r) \tag{A1}$$

$$+\bar{G}^{(0)}_{\rm rad.}(r)\cdot\bar{G}^{(0)}_{\rm rad.}(r)h(r-\xi) \tag{A2}$$

$$+\bar{G}^{(0)}_{\rm stat.}(r)\cdot\bar{G}^{(0)}_{\rm rad.}(r)h(r-\xi)\big\}, \tag{A3}$$

$$\phi^{(1,0)}_{\alpha\parallel} = -k^2\rho\alpha{\rm Tr}\Big\{\int d^3r\big[\bar{G}^{(0)}_{\rm rad.}(r)+\bar{G}^{(0)}_{\rm stat.}(r)\big]\cdot\bar{G}^{(0)}_{\rm stat.}(r)[1+h(r-\xi)]\Big\} = -k^2\rho\alpha\int d^3r$$

$$\times{\rm Tr}\big\{\bar{G}^{(0)}_{\rm stat.}(r)\cdot\bar{G}^{(0)}_{\rm stat.}(r) \tag{A4}$$

$$+\bar{G}^{(0)}_{\rm stat.}(r)\cdot\bar{G}^{(0)}_{\rm stat.}(r)h(r-\xi) \tag{A5}$$

$$+\bar{G}^{(0)}_{\rm rad.}(r)\cdot\bar{G}^{(0)}_{\rm stat.}(r)h(r-\xi)\big\}. \tag{A6}$$

Note that it is the presence of $h(r-\xi)$ that makes the crossed terms nonvanishing.

Next, particularizing to the hs model with $h(r-\xi) = -\Theta(r-\xi)$,

$$2\phi^{(1,0)}_{\alpha\perp}\big|_{\rm hs} = \frac{ke^{i\zeta}}{4\pi\zeta^3}\rho\alpha[2-2i\zeta+e^{i\zeta}(-2+4i\zeta+2\zeta^2-i\zeta^3)]$$

$$\simeq \frac{-k}{2\pi}\rho\alpha\left[\frac{1}{2\zeta}+\frac{5}{6}i\right], \quad \zeta<1, \tag{A7}$$

$$\phi^{(1,0)}_{\alpha\parallel}\big|_{\rm hs} = \frac{-k}{2\pi}\rho\alpha e^{i\zeta}\left[\frac{1}{\zeta^3}-\frac{i}{\zeta^2}\right]$$

$$\simeq \frac{-k}{2\pi}\rho\alpha\left[\frac{1}{\zeta^3}+\frac{1}{2\zeta}+\frac{i}{3}\right], \quad \zeta<1, \tag{A8}$$

where $\zeta = k\xi$. The $\zeta$-independent terms correspond to the long-wavelength propagating modes. In Fourier space, they are given by the poles of the radiative propagators in Eqs. (A1), (A3), and (A6).

Next, we estimate the contribution of the two-point overdensity correlation function to $\phi^{(1,0)}$ in $\mathcal{E}^{LSh}_{\mathcal{O}(\rho)}$,$\mathcal{F}^V_{\mathcal{O}(\rho^2)}$. Let $h^{\rm ovd}(r-\xi) = \xi C\delta^{(1)}(r-\xi)$, where $C$ is a positive constant which accounts for the molecular coordination number. It

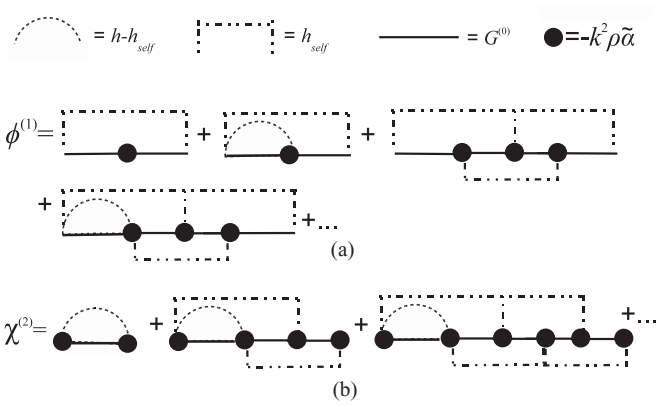


FIG. 4. Diagrammatic representation of the series of recurrent scattering diagrams which amount to (a) $\phi^{(1)}$ and (b) $\bar{\chi}^{(2)}$. In the latter series the first diagram corresponds to $\bar{\chi}^{(2,0)}$ while the remaining ones belong to $\bar{\chi}^{(2)}_{\rm rec}$.

suffices to integrate in angles to obtain

$$\phi^{(1,0)}_{\rm ovd} = \mathrm{Tr}\left\{\int d^3r\, \bar{G}^{(0)}(\vec{r})\cdot(-k^4\rho\tilde{\alpha})\bar{G}^{(0)}(\vec{r})\xi C\delta^{(1)}(r-\xi)\right\}$$
$$= \frac{-k^3}{2\pi}C\rho\tilde{\alpha}e^{2i\zeta}\left[\frac{3}{\zeta^3}-\frac{6}{\zeta^2}i-\frac{5}{\zeta}+2i+\zeta\right]. \qquad \text{(A9)}$$

## APPENDIX B: COMPUTATION OF $\phi^{(1)}_\alpha$ AND $\bar{\chi}^{(2)}_{\alpha\rm rec}$ INCLUDING RECURRENT SCATTERING

We proceed to sum up the infinite series of recurrent scattering diagrams which contribute to $\phi^{(1)}_\alpha$ in $\mathcal{E}^{LSh}_{\mathcal{O}(\rho)}$,$\mathcal{F}^{V}_{\mathcal{O}(\rho^2)}$. The series is pictured diagrammatically in Fig. 4(a). Replacing $\tilde{\alpha}$ with $\alpha$ the sum reads

$$\phi^{(1)}_\alpha = -k^4\rho\alpha\mathrm{Tr}\left\{\int d^3r\bar{G}^{(0)}(\vec{r})\right.$$
$$\left.\cdot\sum_{m=0}(k^2\alpha)^{2m}[\bar{G}^{(0)}(\vec{r})]^{2m}\cdot\bar{G}^{(0)}(\vec{r})[1+h(r-\xi)]\right\}, \qquad \text{(B1)}$$

where the scatterers are all taken bare to keep the order $\rho$ in the series. An analytical expression for this sum can be given. We follow the computation of [71] in which the authors decompose $\bar{G}^{(0)}(\vec{r})$ in transverse and longitudinal components with respect to the position vector, $\vec{r}$,

$$\bar{G}^{(0)}(\vec{r}) = P(r)[\bar{\mathbb{I}}-\hat{r}\otimes\hat{r}]+Q(r)\hat{r}\otimes\hat{r}, \qquad \text{(B2)}$$

with

$$P(r) = \frac{-e^{ikr}}{4\pi r}[1+i/(kr)-1/(kr)^2],$$
$$Q(r) = \frac{-e^{ikr}}{4\pi r}[-2i/(kr)+2/(kr)^2].$$

In terms of $P$, $Q$, Eq. (B1) reads

$$\phi^{(1)}_\alpha = -k^4\rho\alpha\int d^3r\left[\frac{2P^2}{1-(k^2\alpha P)^2}\right.$$
$$\left.+\frac{Q^2}{1-(k^2\alpha Q)^2}\right][1+h(r-\xi)]. \qquad \text{(B3)}$$

The integration in $r$ of Eq. (B1) and its further integration in $\omega$ in Eq. (92) require their expansion in powers of $\alpha$, hence, the series of Eq. (99).

In a similar fashion, the full series of recurrent scattering diagrams which amount to $\chi^{(2)}_{\rm rec\perp,\parallel}$ are those on the right-hand side of the equality of Fig. 4(b), except for the first one. Their sum yields

$$\bar{\chi}^{(2)}_{\alpha\rm rec}(\vec{r}) = -k^2(\rho\alpha)^2\bar{G}^{(0)}(\vec{r})\cdot\sum_{m=1}(-k^2\alpha)^{2m}$$
$$\times[\bar{G}^{(0)}(\vec{r})]^{2m}[1+h(r-\xi)]. \qquad \text{(B4)}$$

Using the decomposition of $\bar{G}^{(0)}$ in terms of $P$, $Q$, it can be written as

$$\bar{\chi}^{(2,2)}_\alpha(\vec{r}) = -k^2(\rho\alpha)^2\left[\frac{P}{1-(k^2\alpha P)^2}(\bar{\mathbb{I}}-\hat{r}\otimes\hat{r})\right.$$
$$\left.+\frac{Q}{1-(k^2\alpha Q)^2}\hat{r}\otimes\hat{r}\right][1+h(r-\xi)]. \qquad \text{(B5)}$$

At leading order in $(\alpha/\xi^3)^2$, the zero mode which modifies the original MG formula for the electrical susceptibility is

$$\chi^{(2)}_{\alpha\rm rec}|_{\perp,\parallel}(q\xi=0)\simeq\tfrac{1}{3}(\rho\tilde{\alpha})^2(\tilde{\alpha}/4\pi\xi^3)^2. \qquad \text{(B6)}$$


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