Fluctuational electrodynamics for nonlinear materials in and out of thermal equilibrium

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We develop fluctuational electrodynamics for media with nonlinear optical response in and out of thermal equilibrium. Starting from the stochastic nonlinear Helmholtz equation and using the fluctuation dissipation theorem, we obtain perturbatively a deterministic nonlinear Helmholtz equation for the average field, the physical linear response, as well as the fluctuations and Rytov currents. We show that the effects of nonlinear optics, in or out of thermal equilibrium, can be taken into account with an effective dielectric function. We discuss the heat radiation of a planar, nonlinear surface, showing that Kirchhoff's law must be applied carefully. We find that the spectral emissivity of a nonlinear nanosphere can in principle be negative, implying the possibility of heat-flow reversal for specific frequencies.

I. INTRODUCTION

Fluctuational electrodynamics (FE) has been instrumental in describing physical phenomena that involve electromagnetic noise, such as the Casimir effect and heat transfer [1–6]. By using the fluctuation dissipation theorem (FDT), FE connects the properties of the involved objects (such as reflection coefficients) with the fluctuations of charges and fields in or out of equilibrium. This has led to a wealth of theoretical results that have been tested extensively in experiments [7–15]. The FE theory has historically focused on systems that respond purely linearly to the electric field. This simplifies the already formidable problem, because fluctuations can be treated separately from other parts of the field due to the superposition principle. As a result, however, many interesting phenomena classified under nonlinear optics have not been taken into account. These include, for example, frequency mixing, the optical Kerr effect, as well as Raman and Brillouin effects [16,17].

The concept of fluctuations in systems with a nonlinear response has been investigated for more than 50 years [18,19]. However, research regarding fluctuation phenomena for optically nonlinear systems appears limited. The noise polarization has been discussed in the context of nonlinear macroscopic quantum electrodynamics [20,21]. Equilibrium Casimir forces have been studied from a field-theoretical perspective [22], focusing on the situation of a nonlinear material immersed between two bodies, while van der Waals forces for objects with nonlinear polarizability were analyzed in Refs. [23–25]. A generalization of the framework of FE to nonlinear materials was performed in Ref. [26], where it was found that proximity between nonlinear objects can change their effective linear properties and gives rise to a qualitatively different Casimir force. Also, nonequilibrium cases have been studied, such as heat radiation of a single nonlinear optical cavity using classical Langevin equations [27,28]. However, there seems to be no literature available on theoretical approaches to out-of-equilibrium phenomena in nonlinear optics, which are based on the vector Helmholtz equation.

In this paper, we extend the framework of Ref. [26], first providing a more extensive discussion and derivation of the theory for equilibrium processes. We then develop a framework for out-of-equilibrium scenarios. Starting from the nonlinear stochastic Helmholtz equation, we derive the fluctuations of the electromagnetic field and the corresponding Rytov source fluctuations, which are in agreement with the FDT. Based on a local equilibrium assumption, we then derive the fluctuations for bodies at different temperatures and the corresponding heat radiation and transfer formulas.

We find that Casimir forces as well as heat radiation and transfer can be rationalized in terms of an effective dielectric function (i.e., an effective linear-response function), which is to replace the dielectric function in well-known formulas for linear materials. We determine the properties of the effective dielectric function and demonstrate that it depends on the shape of an object and the position of other objects. For nonequilibrium scenarios, the dielectric function also depends on the *temperatures* of objects. We carefully discuss in which scenarios the effective dielectric function can be computed and in which cases it contains a divergence from the Green's function at coinciding points.

Regarding the heat radiation of a body with nonlinear dielectric properties, we discuss the applicability of Kirchhoff's law of radiation as well as the (im)possibility to exceed the blackbody limit of a planar surface. Regarding the radiation of a nonlinear nanosphere, we show that, in principle, the heat can flow from the colder sphere to a warmer environment in some frequency range.

The paper is organized as follows. In Sec. II we introduce the stochastic nonlinear Helmholtz equation. We determine the equilibrium fluctuations as well as the effective dielectric function and calculate it numerically for single and parallel plate geometries. In Sec. III we allow objects to have different temperatures and derive formulas for heat radiation and

transfer, which are exemplified for the case of a plate and a nanosphere.

II. NONLINEAR FLUCTUATIONAL ELECTRODYNAMICS IN EQUILIBRIUM

A. Stochastic nonlinear Helmholtz equation

The macroscopic Maxwell's equations describe the dynamics of the electric ($\mathbf{E}$) and magnetic ($\mathbf{B}$) fields in matter via the polarization ($\mathbf{P}$) and magnetization ($\mathbf{M}$) fields. In this article, we consider nonmagnetic materials ($\mathbf{M} = \mathbf{0}$). Maxwell's equations can then be cast in the form of the well-known Helmholtz equation (also known as the wave equation), which in frequency space is given as

$$\mathbf{\nabla} \times \mathbf{\nabla} \times \mathbf{E} - (1 + i\zeta)\frac{\omega^2}{c^2}\mathbf{E} - \frac{1}{\varepsilon_0}\frac{\omega^2}{c^2}\mathbf{P} = i\mu_0\omega\mathbf{J}. \quad (1)$$

$\mathbf{E} = \mathbf{E}(\mathbf{r};\omega)$ is the Fourier component of the electric field at position $\mathbf{r}$ and frequency ω, with c the speed of light and ε_0 the vacuum permittivity (we use SI units). An infinitesimal absorption $i\zeta$ is added to take into account vacuum fluctuations. On the right-hand side of Eq. (1) are the external sources (currents and static charges) $\mathbf{J}$ [29], which in our case will include the stochastic source for thermal and quantum noise, but also a perturbing source to measure the response of the system. In the following, we will consider systems without external charges or currents.

The polarization field $\mathbf{P}$ is a functional of the electric field $\mathbf{E}$, known as a constitutive relation. Since most materials respond dominantly linearly (unless the electric field is very high), the polarization is conventionally given in powers of $\mathbf{E}$. We will consider here materials with a spatially local response, in which case the polarization at a particular point depends on the electric field at that same point. A generalization to nonlocal materials is in principle possible. The polarization vector field is thus

$$\mathbf{P} = \varepsilon_0 \frac{c^2}{\omega^2}(\mathbb{V}\mathbf{E} + \mathcal{M}[\mathbf{E} \otimes \mathbf{E}] + \mathcal{N}[\mathbf{E} \otimes \mathbf{E} \otimes \mathbf{E}] + \cdots), \quad (2)$$

where the dots represent higher-order terms in $\mathbf{E}$. In this paper, we will neglect terms beyond third order.

Using summation over repeated tensor indices (as used throughout this paper), the linear term in Eq. (2) is given by

$$(\mathbb{V}\mathbf{E})_i = \frac{\omega^2}{c^2}\chi_{ij}^{(1)}(-\omega,\omega)E_j(\omega). \quad (3)$$

In this paper, we refer to $\mathbb{V}$ as the (electromagnetic) potential operator [30,31]. It is also common to use the dielectric function (permittivity) tensor ε or the linear susceptibility $\chi^{(1)} = \varepsilon - 1$, which is very useful for a consistent notation of higher orders, as we see below. For the same reason, we have also kept two frequency arguments (for outgoing and incoming waves, which is a standard notation in nonlinear optics), the first of which is typically omitted in the linear case because only waves of the same frequency interact. The susceptibility depends on a spatial coordinate, being zero in vacuum and typically finite and homogeneous inside objects.

The second-order term in Eq. (2) reads

$$\mathcal{M}[\mathbf{E} \otimes \mathbf{E}]_i(\omega) = \frac{\omega^2}{c^2}\int d\omega_1 d\omega_2 \delta(\omega - \omega_\sigma) \\ \times \chi_{ijk}^{(2)}(-\omega_\sigma,\omega_1,\omega_2)E_j(\omega_1)E_k(\omega_2). \quad (4)$$

The second-order susceptibility $\chi^{(2)}$ carries formally three frequency arguments. The δ function with $\omega_\sigma = \omega_1 + \omega_2$ reflects the fact that the time-domain response depends only on time differences, i.e., that the susceptibilities are constant in time. The assumption of spatial locality implies that it depends on a single spatial coordinate, so that the two fields in Eq. (4) are evaluated at the same position in space. We also introduced the dyadic product "$\otimes$," so that the argument of $\mathcal{M}$ is a second-rank spatial tensor. The third-order term is then a natural extension,

$$\mathcal{N}[\mathbf{E} \otimes \mathbf{E} \otimes \mathbf{E}]_i(\omega) \\ = \frac{\omega^2}{c^2}\int d\omega_1 d\omega_2 d\omega_3 \delta(\omega - \omega_\sigma) \\ \times \chi_{ijkl}^{(3)}(-\omega_\sigma,\omega_1,\omega_2,\omega_3)E_j(\omega_1)E_k(\omega_2)E_l(\omega_3). \quad (5)$$

Note that we have here $\omega_\sigma = \omega_1 + \omega_2 + \omega_3$. Due to intrinsic symmetries in $\chi^{(2)}$ and $\chi^{(3)}$ [17], the operators $\mathcal{M}$ and $\mathcal{N}$ are commutative in their operands. This means $\mathcal{M}[\mathbf{A} \otimes \mathbf{B}] = \mathcal{M}[\mathbf{B} \otimes \mathbf{A}]$ and the same applies for permutations in $\mathcal{N}[\mathbf{A} \otimes \mathbf{B} \otimes \mathbf{C}]$.

Introducing the free Helmholtz operator $\mathbb{H}_0 = \mathbf{\nabla} \times \mathbf{\nabla} \times -(1 + i\zeta)\frac{\omega^2}{c^2}\mathbb{I}$ and using Eq. (2), the stochastic nonlinear Helmholtz equation can be written as

$$(\mathbb{H}_0 - \mathbb{V})\mathbf{E} - \mathcal{M}[\mathbf{E} \otimes \mathbf{E}] - \mathcal{N}[\mathbf{E} \otimes \mathbf{E} \otimes \mathbf{E}] \\ = \mathbf{F} + \mathbb{H}_0\mathbf{E}_{\text{in}}, \quad (6)$$

where we replaced the generic source term by $i\mu_0\omega\mathbf{J} = \mathbf{F} + \mathbb{H}_0\mathbf{E}_{\text{in}}$. $\mathbf{F}$ is the stochastic source of thermal and quantum noise, whereas $\mathbf{E}_{\text{in}}$ denotes a deterministic probing field. Since we consider a system without external charges or currents, we must have $\langle \mathbf{F} \rangle = \mathbf{0}$.

We mark a few crucial differences between nonlinear and linear Helmholtz equations. First, the different frequency components of $\mathbf{E}$ are coupled through Eqs. (4) and (5). This means fluctuations of all frequencies influence the scattering of the electric field of any particular frequency. This is a manifestation of the absence of the superposition principle. As a consequence, different frequency components cannot be simply added, and in general Eq. (6) needs to be solved self-consistently. Our approach is to notice that the nonlinear terms are small for most realistic materials and therefore approach the problem perturbatively, giving results to leading order in $\chi^{(2)}$ and $\chi^{(3)}$.

It is useful to separate the electric field into a mean part and fluctuations as $\mathbf{E} = \overline{\mathbf{E}} + \delta\mathbf{E}$, where $\overline{\mathbf{E}} = \langle \mathbf{E} \rangle$ is a shorthand notation for the average field. In the linear case ($\mathcal{M} = \mathcal{N} = 0$) one obtains two independent equations for $\delta\mathbf{E}$ and $\overline{\mathbf{E}}$,

$$(\mathbb{H}_0 - \mathbb{V})\delta\mathbf{E} = \mathbf{F}, \quad (7)$$

$$(\mathbb{H}_0 - \mathbb{V})\overline{\mathbf{E}} = \mathbb{H}_0\mathbf{E}_{\text{in}}. \quad (8)$$

In the next two subsections, we will derive equations for the average field and for the fluctuations in the nonlinear case. We

will show that the behavior of the average field will depend on the strength of the fluctuations.

B. Average field, effective potential, and linear response

A linear or nonlinear scattering experiment typically detects the noise-averaged field $\overline{\mathbf{E}}$, and its equation of motion shall be derived here. As mentioned, due to the absence of the superposition principle, the noise in Eq. (6) has nontrivial consequences for the average field. This may be seen explicitly by substituting $\mathbf{E} = \overline{\mathbf{E}} + \delta\mathbf{E}$ into Eq. (6) and taking the average. Using the commutative properties of $\mathcal{M}$ and $\mathcal{N}$ together with the fact that by definition $\langle\delta\mathbf{E}\rangle = 0$, we obtain a nonlinear Helmholtz equation for the mean electric field,

$$(\mathbb{H}_0 - \mathbb{V})\overline{\mathbf{E}} - 3\mathcal{N}[\langle\delta\mathbf{E}\otimes\delta\mathbf{E}\rangle\overline{\mathbf{E}}]$$
$$- \mathcal{M}[\overline{\mathbf{E}}\otimes\overline{\mathbf{E}}] - \mathcal{N}[\overline{\mathbf{E}}\otimes\overline{\mathbf{E}}\otimes\overline{\mathbf{E}}]$$
$$= \mathbb{H}_0\mathbf{E}_{\text{in}} + \mathcal{M}[\langle\delta\mathbf{E}\otimes\delta\mathbf{E}\rangle] + \mathcal{N}[\langle\delta\mathbf{E}\otimes\delta\mathbf{E}\otimes\delta\mathbf{E}\rangle]. \quad (9)$$

In stationary systems, the time-domain correlators can only depend on time differences [32]. This means that in frequency space the fluctuations are δ-correlated with $\langle\delta\mathbf{E}(\omega)\otimes\delta\mathbf{E}(\omega')\rangle = \delta(\omega+\omega')\langle\delta\mathbf{E}\otimes\delta\mathbf{E}\rangle_\omega$. Therefore, we can consider the second term of the first line to be a linear and local operator $\mathcal{N}[\langle\delta\mathbf{E}\otimes\delta\mathbf{E}\rangle\cdot](\mathbf{r},\omega)$ acting on $\overline{\mathbf{E}}(\mathbf{r},\omega)$. This operator can be written explicitly,

$$\mathcal{N}[\langle\delta\mathbf{E}\otimes\delta\mathbf{E}\rangle\cdot]_{ij}(\mathbf{r};\omega)$$
$$= \frac{\omega^2}{c^2}\int d\omega'\,\chi_{ijkl}^{(3)}(\mathbf{r};-\omega,\omega,\omega',-\omega')\langle\delta E_k(\mathbf{r})\delta E_l(\mathbf{r})\rangle_{\omega'}, \quad (10)$$

where we give the spatial argument to emphasize spatial locality. Since it is linear and local, like $\mathbb{V}$ in Eq. (3), we may interpret it as an additional potential, resulting in the effective, fluctuation-dependent potential

$$\tilde{\mathbb{V}} = \mathbb{V} + 3\mathcal{N}[\langle\delta\mathbf{E}\otimes\delta\mathbf{E}\rangle\cdot]. \quad (11)$$

Equivalently, we can define an effective dielectric function corresponding to the effective potential $\tilde{\mathbb{V}}$ as

$$\tilde{\varepsilon}_{ij}(\mathbf{r};\omega) = \varepsilon_{ij}(\mathbf{r};\omega) + \int d\omega'\,N_{ij}(\mathbf{r};\omega,\omega'), \quad (12)$$

$$N_{ij}(\mathbf{r};\omega,\omega') = 3\chi_{ijkl}^{(3)}(\mathbf{r};-\omega,\omega,\omega',-\omega')$$
$$\times \langle\delta E_k(\mathbf{r})\delta E_l(\mathbf{r})\rangle_{\omega'}. \quad (13)$$

Moving on to the last line of Eq. (9), we see that in addition to the probing source, two fluctuation-induced source terms appear. The source from $\mathcal{M}$ can be written explicitly as

$$\mathcal{M}[\langle\delta\mathbf{E}\otimes\delta\mathbf{E}\rangle]_i(\mathbf{r};\omega)$$
$$= \delta(\omega)\frac{\omega^2}{c^2}\int d\omega'\,\chi_{ijk}^{(2)}(\mathbf{r};0,\omega',-\omega')\langle\delta E_j(\mathbf{r})\delta E_k(\mathbf{r})\rangle_{\omega'} = 0. \quad (14)$$

This term vanishes because $\delta(\omega)\omega^2 = 0$, and the integral over ω' is independent of ω. This term is thus not present in the Helmholtz equation. The third-order source term, $\mathcal{N}[\langle\delta\mathbf{E}\otimes\delta\mathbf{E}\otimes\delta\mathbf{E}\rangle]$, cannot be eliminated generally for any ω. Since the dynamics follows a nonlinear equation, the

fluctuations $\delta\mathbf{E}$ are generally non-Gaussian. It is therefore not obvious how to evaluate the three-point correlator.

Starting from here, we will only keep the *leading-order* terms in $\chi^{(2)}$ and $\chi^{(3)}$, neglecting terms of order $O([\chi^{(2)}]^2)$, $O(\chi^{(2)}\chi^{(3)})$, $O([\chi^{(3)}]^2)$, and beyond. This simplification is justified from the observation that $\chi^{(3)}$ (and $\chi^{(2)}$) are typically small in reality and we aim to calculate their leading influence on effects such as the Casimir force and heat transfer. This implies that the fields inside the $\mathcal{M}$ and the $\mathcal{N}$ operator in Eq. (9) can be found from linear theory. Most importantly, the fluctuations $\delta\mathbf{E}$ in this linear system (with $\chi^{(2)} = \chi^{(3)} = 0$) are Gaussian. Three-point (or any odd number) correlations vanish, so that the last term of Eq. (9) is zero.

With these considerations, we finally arrive at the nonlinear Helmholtz equation for the average field $\overline{\mathbf{E}}$ (valid for $\omega \neq 0$),

$$(\mathbb{H}_0 - \tilde{\mathbb{V}})\overline{\mathbf{E}} - \mathcal{M}[\overline{\mathbf{E}}\otimes\overline{\mathbf{E}}] - \mathcal{N}[\overline{\mathbf{E}}\otimes\overline{\mathbf{E}}\otimes\overline{\mathbf{E}}] = \mathbb{H}_0\mathbf{E}_{\text{in}}. \quad (15)$$

We recover the same structure as in Eq. (6), only without the noise term (making it deterministic) and a modified, renormalized, linear term. This equation determines the result of both linear and nonlinear scattering experiments. Furthermore, we note that $\chi^{(2)}$ and $\chi^{(3)}$ (entering $\mathcal{M}$ and $\mathcal{N}$) are not renormalized by the noise in this order. That would appear by inclusion of $\chi^{(5)}$ and so on.

To make even clearer the meaning of the effective potential (and the effective dielectric function), we explicitly compute the result of a linear-response scattering experiment. To that end, we interpret $\mathbf{E}_{\text{in}}$ as the incident field in such an experiment and find the scattered one linear in it, thereby defining the linear-response function $\tilde{\mathbb{G}}$,

$$\tilde{\mathbb{G}} = \left.\frac{\delta\overline{\mathbf{E}}}{\delta(\mathbb{H}_0\mathbf{E}_{\text{in}})}\right|_{\mathbf{E}_{\text{in}}\to 0} = \left.\frac{\delta\overline{\mathbf{E}}}{\delta\mathbf{E}_{\text{in}}}\right|_{\mathbf{E}_{\text{in}}\to 0}\mathbb{G}_0, \quad (16)$$

with the free Green's function $\mathbb{G}_0 = \mathbb{H}_0^{-1}$ [29]. $\tilde{\mathbb{G}}$ follows directly from Eq. (15) as [26]

$$\tilde{\mathbb{G}} = (\mathbb{H}_0 - \tilde{\mathbb{V}})^{-1}. \quad (17)$$

The linear response is indeed given by the effective potential $\tilde{\mathbb{V}}$ or, equivalently, by the effective dielectric function $\tilde{\varepsilon}$, which depends on the fluctuations through Eq. (13).

Notably, for $\chi^{(3)} = 0$, we recover the well-known linear-response function. In that case, Eq. (17) reduces to the Green's function $\mathbb{G} = (\mathbb{H}_0 - \mathbb{V})^{-1}$ of the system [30]. We will see in the following that it is the effective potential that will give rise to the effects of $\chi^{(3)}$ on Casimir forces and heat transfer. Analogously to Eq. (16), higher-order derivatives can be used to infer the nonlinear susceptibilities, but they will not be relevant for the remainder of the paper, because they play no role in fluctuational effects.

C. Equilibrium fluctuations and Rytov currents

In the previous subsections, we have determined Eq. (6) for the fluctuating field $\mathbf{E}$ and Eq. (15) for its mean $\overline{\mathbf{E}}$. However, in the first case we need to know the noise source $\mathbf{F}$ and in the second case the correlations of the fluctuations $\delta\mathbf{E}$ in Eq. (11). Because these equations are linked by $\mathbf{E} = \overline{\mathbf{E}} + \delta\mathbf{E}$ and therefore $\langle\mathbf{E}\otimes\mathbf{E}\rangle = \overline{\mathbf{E}}\otimes\overline{\mathbf{E}} + \langle\delta\mathbf{E}\otimes\delta\mathbf{E}\rangle$, we will now

determine $\delta\mathbf{E}$ by use of the FDT. We will then find the correlations of $\mathbf{F}$ and connect them to Rytov theory [3,33].

The equilibrium fluctuations $\langle\delta\mathbf{E}\otimes\delta\mathbf{E}\rangle^{\mathrm{eq}}$ are related to the linear-response function $\tilde{\mathbb{G}}$ defined by Eq. (16) through the FDT [32], given explicitly as

$$\langle\delta\mathbf{E}_\omega\otimes\delta\mathbf{E}_{\omega'}^*\rangle^{\mathrm{eq}}=\delta(\omega-\omega')\frac{b(\omega)}{2i}\tilde{\mathbb{G}}_{\mathrm{AH}}^{\mathrm{eq}}(\omega),\qquad(18)$$

$$b(\omega)=\frac{\hbar}{\pi\varepsilon_0}\frac{\omega^2}{c^2}\frac{1}{1-\exp\left(-\frac{\hbar\omega}{k_{\mathrm{B}}T}\right)},\qquad(19)$$

with the reduced Planck constant $\hbar$ and thermal energy $k_{\mathrm{B}}T$. $\tilde{\mathbb{G}}^{\mathrm{eq}}=(\mathbb{H}_0-\tilde{\mathbb{V}}^{\mathrm{eq}})^{-1}$ is the linear response, given by Eq. (17) in equilibrium (denoted by the superscript "eq," also used for averages $\langle\cdots\rangle^{\mathrm{eq}}$). The subscript denotes the anti-Hermitian part of an operator, $\mathbb{O}_{\mathrm{AH}}=\mathbb{O}-\mathbb{O}^\dagger$. Note that for symmetric operators, $\frac{1}{2i}\mathbb{O}_{\mathrm{AH}}=\mathrm{Im}\,\mathbb{O}$. We use this slightly more involved form of the FDT to avoid restriction to cases with symmetric responses. In the rest of the work, we will only include the frequency dependence of operators explicitly if necessary to avoid ambiguity (e.g., when there is more than one frequency involved).

Unlike in linear systems, the linear response $\tilde{\mathbb{G}}$ as well as the effective potential $\tilde{\mathbb{V}}$ in Eq. (11) depend on the correlations of $\delta\mathbf{E}$ and are in general different in equilibrium as compared to the nonequilibrium situation considered in Sec. III.

The effective potential or dielectric function in equilibrium is given by Eq. (11) or (13) with the equilibrium correlator on the right-hand side. This will be discussed in Sec. II D below. Equation (18) gives a rigid and well-known relation between two different measurable quantities, and it is the heart of our analysis. The correlator $\langle\delta\mathbf{E}\otimes\delta\mathbf{E}\rangle^{\mathrm{eq}}$ determines the Casimir force while the linear response $\tilde{\mathbb{G}}^{\mathrm{eq}}$ describes optical scattering experiments.

Using the fluctuations obtained by the FDT, we can also determine the correlations of the noise sources $\mathbf{F}$. For $\overline{\mathbf{E}}=\mathbf{E}_{\mathrm{in}}=\mathbf{0}$, we have from Eq. (6) (we include here a finite $\chi^{(2)}$ to demonstrate its consequences)

$$\mathbf{F}=(\mathbb{H}_0-\mathbb{V})\delta\mathbf{E}-\mathcal{M}[\delta\mathbf{E}\otimes\delta\mathbf{E}]-\mathcal{N}[\delta\mathbf{E}\otimes\delta\mathbf{E}\otimes\delta\mathbf{E}].\tag{20}$$

As before, we note a subtlety at zero frequency for finite $\chi^{(2)}(0,\omega',-\omega')$. This implies a contradiction of Eq. (20) with the fundamental assumption $\langle\mathbf{F}\rangle=0$, however only at $\omega=0$.

Keeping only terms up to leading order in $\chi^{(2)}$ and $\chi^{(3)}$, the equilibrium correlator of $\mathbf{F}$ follows directly from Eq. (20),

$$\begin{aligned}\langle\mathbf{F}_\omega\otimes\mathbf{F}_{\omega'}^*\rangle^{\mathrm{eq}}={}&(\mathbb{H}_0-\mathbb{V})_\omega\langle\delta\mathbf{E}_\omega\otimes\delta\mathbf{E}_{\omega'}^*\rangle^{\mathrm{eq}}(\mathbb{H}_0-\mathbb{V})_{\omega'}^\dagger\\&-(\mathbb{H}_0-\mathbb{V})_\omega\langle\delta\mathbf{E}_\omega\otimes\mathcal{N}[\delta\mathbf{E}\otimes\delta\mathbf{E}\otimes\delta\mathbf{E}]_{\omega'}^*\rangle^{\mathrm{eq}}\\&-\langle\mathcal{N}[\delta\mathbf{E}\otimes\delta\mathbf{E}\otimes\delta\mathbf{E}]_\omega\otimes\delta\mathbf{E}_{\omega'}^*\rangle^{\mathrm{eq}}(\mathbb{H}_0-\mathbb{V})_{\omega'}^\dagger.\end{aligned}\tag{21}$$

We can further assume Gaussianity of the fields in the last two terms, because they carry already an explicit factor of $\chi^{(3)}$ (and are thus to be taken from the linear system). Specifically, by using Isserlis' theorem (also known as Wick's theorem or the Furutsu-Novikov formula) and the commutation properties of

$\mathcal{N}$, we can write

$$\begin{aligned}\langle\mathcal{N}[\delta\mathbf{E}\otimes\delta\mathbf{E}\otimes\delta\mathbf{E}]\otimes\delta\mathbf{E}^*\rangle^{\mathrm{eq}}\\=3\mathcal{N}[\langle\delta\mathbf{E}\otimes\delta\mathbf{E}\rangle^{\mathrm{eq}}\cdot]\langle\delta\mathbf{E}\otimes\delta\mathbf{E}^*\rangle^{\mathrm{eq}}.\end{aligned}\tag{22}$$

We note the appearance of the same operator as in Eq. (11), which may be written in terms of $\tilde{\mathbb{V}}$ or, equivalently, $\tilde{\mathbb{G}}$. We therefore find

$$\langle\mathbf{F}_\omega\otimes\mathbf{F}_{\omega'}^*\rangle^{\mathrm{eq}}=\left[(\tilde{\mathbb{G}}_\omega^{-1})\langle\delta\mathbf{E}_\omega\otimes\delta\mathbf{E}_{\omega'}^*\rangle(\tilde{\mathbb{G}}_{\omega'}^{-1})^\dagger\right]^{\mathrm{eq}}.\tag{23}$$

Using Eqs. (17) and (18), we can further write this as

$$\langle\mathbf{F}_\omega\otimes\mathbf{F}_{\omega'}^*\rangle^{\mathrm{eq}}=-\delta(\omega-\omega')\frac{b(\omega)}{2i}(\mathbb{H}_0-\tilde{\mathbb{V}}^{\mathrm{eq}})_{\mathrm{AH}},\tag{24}$$

which agrees with the relation for Rytov currents for linear systems [3,33]. As noted in Ref. [26], the noise sources (the Rytov currents) are related to the effective potential $\tilde{\mathbb{V}}^{\mathrm{eq}}$ in the same manner as they are related to the bare potential $\mathbb{V}$ in linear systems. This confirms the interpretation of $\tilde{\mathbb{V}}^{\mathrm{eq}}$ as the linear-response function, as it appears in a fluctuation dissipation theorem with the noise in Eq. (24). Equations (18) and (24) are thus two versions of the fluctuation dissipation theorem [32]. Note that the imaginary part of $\mathbb{H}_0$ in Eq. (24) accounts for the fluctuations in the absence of any objects (due to the dust [34]).

We have thus demonstrated the consistency of FDT and Rytov theory in the nonlinear case. It is interesting to note that the Rytov currents are uncorrelated in space, as they are in linear systems, because the effective potential is local in space. The potential and the noise are, however, nonlocal in the sense that their value at one position depends on the properties of the system at all other points in space. For example, the effective potential of a point inside an object depends on the shape of the object or on the presence of surrounding objects. It means that Eq. (24) is an implicit equation for $\langle\mathbf{F}\otimes\mathbf{F}^*\rangle^{\mathrm{eq}}$, just as Eq. (18) is an implicit equation for $\langle\delta\mathbf{E}\otimes\delta\mathbf{E}^*\rangle^{\mathrm{eq}}$.

We note that $\mathrm{Im}\,\tilde{\mathbb{V}}^{\mathrm{eq}}$ in Eq. (24) must be positive, as can already be seen by its connection to an autocorrelation function. This property is, however, hard to show explicitly without posing additional constraints on $\chi^{(3)}$.

D. The effective dielectric function in equilibrium

In previous sections, we showed how the effective potential $\tilde{\mathbb{V}}$ can be used to take into account nonlinear effects in equilibrium. The most important quantity is the linear-response function, equivalently expressed by the potential $\tilde{\mathbb{V}}^{\mathrm{eq}}$, the dielectric function $\tilde{\varepsilon}_{ij}^{\mathrm{eq}}$, or $\tilde{\mathbb{G}}^{\mathrm{eq}}$, because it governs the fluctuations. We will thus investigate the linear response in more detail with simple examples in this section.

Writing Eqs. (12) and (13) using the FDT in Eq. (18) gives the effective dielectric function in equilibrium,

$$\tilde{\varepsilon}_{ij}^{\mathrm{eq}}(\mathbf{r};\omega)=\varepsilon_{ij}(\mathbf{r};\omega)+\int d\omega'\,N_{ij}^{\mathrm{eq}}(\mathbf{r};\omega,\omega'),\tag{25}$$

$$\begin{aligned}N_{ij}^{\mathrm{eq}}(\mathbf{r};\omega,\omega')=3\chi_{ijkl}^{(3)}(\mathbf{r};-\omega,\omega,\omega',-\omega')\\\times\frac{b(\omega')}{2i}\mathbb{G}_{\mathrm{AH}}(\mathbf{r},\mathbf{r};\omega')_{kl}^{\mathrm{eq}}.\end{aligned}\tag{26}$$

Note that we used the Green's function $\mathbb{G}=(\mathbb{H}_0-\mathbb{V})^{-1}$ instead of the linear response (with a tilde) as in Eq. (18).

This is correct to leading order in $\chi^{(3)}$. $\mathbb{G}$ is known exactly for several geometries, so that these equations are closed.

We note that the imaginary part of the Green's function, as appearing in Eq. (26), generally diverges at coinciding points in absorbing media, which is a well-known property [29] and a recurrent problem of perturbative expansions in field theories [35]. There have been suggestions on how to circumvent this divergence, e.g., by introducing a rigid sphere approximation of the δ function [36] appearing in the Green's function, which appears very similar to an ultraviolet cutoff often introduced in classical field theory.

This problem can also be mitigated in some cases. For example, when computing the Casimir force in Ref. [26], we noted that the nontrivial distance dependence of the force is not sensitive to the divergence, as it cancels out when comparing two different distances. It is thus important to carefully investigate which experimental quantities are insensitive to the mentioned divergence and can thus be predicted. In the remainder of the paper, we will point to this issue for any shown example and reflect on it in Sec. IV. We also comment that using a purely real ε omits the divergence in any circumstance.

In the interest of simplifying the calculation and interpretation of specific examples, especially in view of the complicated tensorial structure of $\chi^{(3)}_{ijkl}$, we consider a highly symmetric material. First, we assume that the bare dielectric function is isotropic, such that $\varepsilon_{ij} = \delta_{ij}\varepsilon$. Regarding $\chi^{(3)}_{ijkl}$, it is known that for centrosymmetric materials, the third-order susceptibility can be written as [17]

$$\chi^{(3)}_{ijkl} = \chi^{(3)}_{1122}\delta_{ij}\delta_{kl} + \chi^{(3)}_{1212}\delta_{ik}\delta_{jl} + \chi^{(3)}_{1221}\delta_{il}\delta_{jk}. \quad (27)$$

Further simplifying, we only keep the first term from Eq. (27), such that we use

$$\chi^{(3)}_{ijkl} = \chi^{(3)}\delta_{ij}\delta_{kl}. \quad (28)$$

With these simplifications, the resulting effective dielectric function is isotropic,

$$\tilde{\varepsilon}^{\mathrm{eq}}_{ij}(\mathbf{r};\omega) = \delta_{ij}\left[\varepsilon(\mathbf{r};\omega) + \int d\omega' N^{\mathrm{eq}}(\mathbf{r};\omega,\omega')\right], \quad (29)$$

$$N^{\mathrm{eq}}(\mathbf{r};\omega,\omega') = 3\chi^{(3)}(\mathbf{r};-\omega,\omega,\omega',-\omega')$$
$$\times\, b(\omega')\mathrm{Im}\mathbb{G}(\mathbf{r},\mathbf{r};\omega')^{\mathrm{eq}}_{kk}. \quad (30)$$

We will consider the examples of a single plate and two parallel plates using Ref. [37]. It gives $\mathbb{G}$ in a plane-wave basis for arbitrary parallel layered structures, which contains the cases of a single semi-infinite plate (two layers: vacuum-plate) and two parallel semi-infinite plates (three layers: plate-vacuum-plate).

We start with a single plate. As mentioned above, the imaginary part of the Green's function at coinciding points [$\mathrm{Im}\tilde{\mathbb{G}}(\mathbf{r},\mathbf{r};\omega')$, as appearing in Eq. (30)] is infinite inside absorbing materials, so the effective dielectric function cannot be computed without further (microscopic) information in general. Subtracting from it the solution of an unbound (bulk) system with the same dielectric function heals the divergence, except for points that are very close to the plate's surface. In the case of a single semi-infinite plate, we therefore restrict ourselves to a real bare $\varepsilon = 4$, for which Eq. (30) can be numerically evaluated. The result, which is nevertheless insightful,
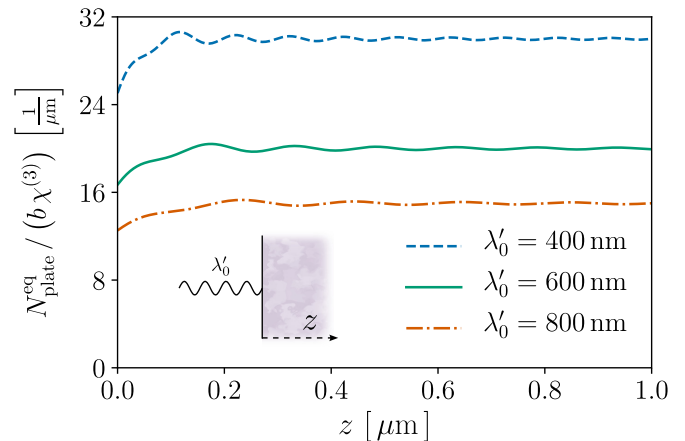

FIG. 1. Spatial dependence of the effective dielectric function inside a single isotropic nonabsorbing plate ($\varepsilon = 4$). z is the distance from the surface and $\lambda'_0 = 2\pi\frac{c}{\omega'}$ is the wavelength corresponding to ω' in vacuum. Note that while $N^{\mathrm{eq}}_{\mathrm{plate}}(\mathbf{r};\omega,\omega')$ depends also on ω, the plotted quantity is independent of ω due to division with $\chi^{(3)}(\mathbf{r};-\omega,\omega,\omega',-\omega')$.

is shown in Fig. 1. The effective dielectric function is inhomogeneous even though all material parameters ($\chi^{(1)}$ and $\chi^{(3)}$) are homogeneous. We thus see explicitly the aforementioned property: The effective dielectric function at a certain position depends on the shape of the object. Specifically, there is an interference pattern of half of the wavelength of the primed frequency, which corresponds to a single reflection from the surface, while far away from the surface the bulk value is approached. Note, however, that since in Eq. (29) we integrate over all ω', this interference pattern appears in the effective dielectric function only if $\chi^{(3)}(-\omega,\omega,\omega',-\omega')$ has a sharp resonance peak in ω'.

The case of two identical parallel surfaces at distance d provides additional insights. Here, we may compute the difference of the effective dielectric function between the cases of the second plate present or absent. Mathematically, we thus subtract from $N^{\mathrm{eq}}_{\mathrm{double}}$ the result of the isolated plate, $N^{\mathrm{eq}}_{\mathrm{plate}}$, and obtain a finite result, because the divergence in $\mathbb{G}$ cancels. This important observation, which was also used in Ref. [26] to compute the Casimir force, contains a very important physical statement: While it is difficult to predict the response of a single object, it is possible to predict the response of two objects, given that the responses of the individual objects are known.

The numerical results for identical plates are shown in Figs. 2 (without absorption) and 3 (with absorption). Not unlike in the single-plate case, an interference pattern arises due to reflections from the second plate (notice the phase shift at different separations). In the nonabsorbing case, these persist throughout the material, while they are limited by the skin depth in absorbing materials. As expected, at large separations d we recover the single-plate result.

These graphs have a very direct connection to the Casimir force between two parallel nonlinear plates of Ref. [26]. As mentioned before, the Casimir force is now found by using the well-known Lifshitz formula [2] for linear materials, but replacing the bare ε by the (inhomogeneous) effective one. Recall that in Ref. [26], we found that the force displays a
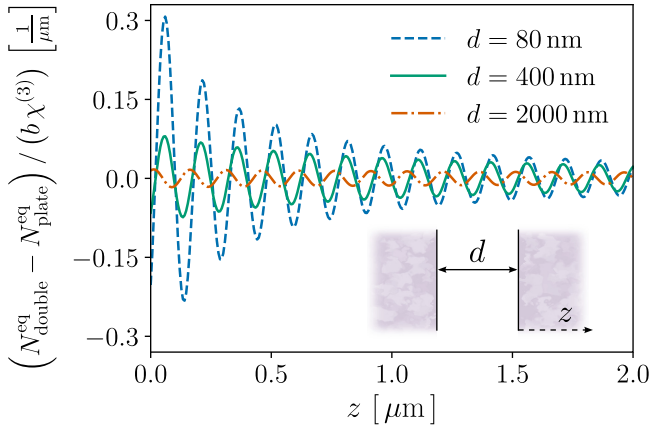
FIG. 2. The effective dielectric function inside one of two nonabsorbing parallel plates ($\varepsilon = 4$), where z is the distance from the surface and d is the separation between the plates. $\lambda'_0 = 2\pi \frac{c}{\omega'} = 600$ nm is the corresponding wavelength in vacuum.

different power law as a function of d for close separations. For the quantum limit the power law changes as $d^{-4} \to d^{-8}$, and for the thermal limit $d^{-3} \to d^{-6}$. This may now be understood in terms of Figs. 2 and 3, because the effective dielectric function changes with d, yielding an additional d dependence in the Casimir force.

III. NONEQUILIBRIUM: HEAT RADIATION

A. Nonequilibrium Rytov currents and field correlations

In Sec. II we developed FE for nonlinear materials in equilibrium, from which inhomogeneous dielectric functions and the Casimir effect for nonlinear objects can be found. In equilibrium, the theory is well grounded by the FDT, relating the linear response and field fluctuations directly by Eq. (18). We now aim to address the out-of-equilibrium scenario of N objects held at different temperatures T_n ($n = 1, \ldots, N$), with an environment at temperature T_0, and compute heat radiation and heat transfer for these objects.

In this nonequilibrium case, certain assumptions are necessary to compute the fluctuations of the electric field, because
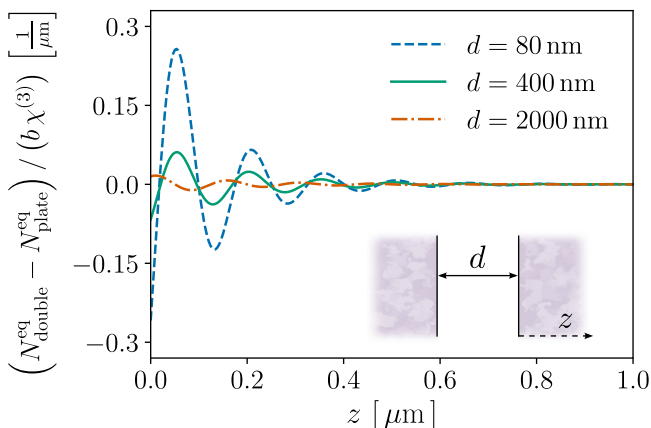

FIG. 3. Same quantity as in Fig. 2 for lossy plates with a dielectric constant $\varepsilon = 4 + i$ and vacuum wavelength $\lambda'_0 = 2\pi \frac{c}{\omega'} = 600$ nm.

the FDT is in general not valid. A useful approximation, which is also used for linear FE, is the assumption of local thermal equilibrium (LTE). In this case, the (nonoverlapping) objects are considered to be in thermal equilibrium at temperatures T_n.

To be able to assign different temperatures, we start by denoting the susceptibilities of order m ($m = 1, 3$) of object n as $\chi_n^{(m)}(\mathbf{r})$. These are nonzero only when $\mathbf{r}$ lies within object n. Note that $\chi^{(2)}$ has no influence on the following discussion. Since the objects are spatially separated from one another, the total susceptibilities can be found through summation as

$$\chi^{(m)}(\mathbf{r}) = \sum_{n=1}^{N} \chi_n^{(m)}(\mathbf{r}). \tag{31}$$

We may also write $\chi^{(m)}(\mathbf{r} \in V_n) = \chi_n^{(m)}(\mathbf{r})$, where V_n is the volume of the object. The same applies for the bare potential ($\mathbb{V} = \sum_{n=1}^{N} \mathbb{V}_n$) and the nonlinear operator ($\mathcal{N} = \sum_{n=1}^{N} \mathcal{N}_n$), as they follow from $\chi^{(1)}$ and $\chi^{(3)}$, respectively. The effective potential is then $\tilde{\mathbb{V}} = \sum_{n=1}^{N} \tilde{\mathbb{V}}_n$, where we have [see Eqs. (10) and (11)]

$$\tilde{\mathbb{V}}_n = \mathbb{V}_n + 3\mathcal{N}_n[\langle \delta\mathbf{E} \otimes \delta\mathbf{E} \rangle \cdot]. \tag{32}$$

The key point in implementing the LTE approximation within FE is to recognize that the equilibrium Rytov currents in Eq. (24) can be written as

$$\langle \mathbf{F}_\omega \otimes \mathbf{F}_{\omega'}^* \rangle^{\mathrm{eq}} = \delta(\omega - \omega') \sum_{n=0}^{N} \frac{b(\omega)}{2i} \left(\tilde{\mathbb{V}}_n^{\mathrm{eq}} \right)_{\mathrm{AH}}. \tag{33}$$

We have denoted $\mathbb{V}_0 = \tilde{\mathbb{V}}_0 = -\mathbb{H}_0$ as the vacuum potential with an infinitesimal imaginary part (absorption). This is known as the environment dust [34] and is needed to take into account radiation to and from the environment.

Since object n is considered to be in local equilibrium at temperature T_n, we can assign an index to the distributions $b(\omega)$,

$$b_n(\omega) = \frac{\hbar}{\pi \varepsilon_0} \frac{\omega^2}{c^2} \left[1 - \exp\left(-\frac{\hbar\omega}{k_\mathrm{B} T_n} \right) \right]^{-1}. \tag{34}$$

Recalling that index $n = 0$ denotes the environment with temperature T_0, we arrive at the nonequilibrium correlator of the Rytov currents,

$$\langle \mathbf{F}_\omega \otimes \mathbf{F}_{\omega'}^* \rangle = \delta(\omega - \omega') \sum_{n=0}^{N} \frac{b_n(\omega)}{2i} \left(\tilde{\mathbb{V}}_n^{\mathrm{eq}} \right)_{\mathrm{AH}}. \tag{35}$$

Note that the effective potential $\tilde{\mathbb{V}}$ depends on the field fluctuations $\langle \delta\mathbf{E} \otimes \delta\mathbf{E} \rangle$ and is thus different out of equilibrium compared to the corresponding equilibrium potential $\tilde{\mathbb{V}}^{\mathrm{eq}}$.

The correlations of the field fluctuations $\delta\mathbf{E}$ out of equilibrium can be found by following the same reasoning leading to Eq. (23) (with $\overline{\mathbf{E}} = \mathbf{0}$). Using $\mathbb{A}\mathbb{B}_{\mathrm{AH}}\mathbb{A}^\dagger = (\mathbb{A}\mathbb{B}\mathbb{A}^\dagger)_{\mathrm{AH}}$, we obtain

$$\langle \delta\mathbf{E}_\omega \otimes \delta\mathbf{E}_{\omega'}^* \rangle = \tilde{\mathbb{G}}_\omega \langle \mathbf{F}_\omega \otimes \mathbf{F}_{\omega'}^* \rangle \tilde{\mathbb{G}}_{\omega'}^\dagger$$

$$= \delta(\omega - \omega') \sum_{n=0}^{N} \frac{b_n(\omega)}{2i} (\tilde{\mathbb{G}} \tilde{\mathbb{V}}_n \tilde{\mathbb{G}}^\dagger)_{\mathrm{AH}}. \tag{36}$$

Recall that the linear-response operator is given by Eq. (17) as $\tilde{\mathbb{G}} = (\mathbb{H}_0 - \tilde{\mathbb{V}})^{-1}$ together with Eq. (32), so that Eq. (35) is

indeed physically meaningful, because the potential $\tilde{\mathbb{V}}$ is the physical linear response of the nonequilibrium system.

As in the equilibrium case, we have an implicit system of equations to determine the fluctuations and the effective potential. It can be solved perturbatively in $\chi^{(3)}$ and we will derive an explicit form for the effective dielectric function in the next subsection, from which the correlator $\langle \delta \mathbf{E} \otimes \delta \mathbf{E}^* \rangle$ can then be computed with Eq. (36).

B. The nonequilibrium effective dielectric function

From Eqs. (35) and (36) we can see that, as in the equilibrium case, the effects of nonlinear terms on fluctuations can be taken into account with the effective potential $\tilde{\mathbb{V}}$ or, equivalently, the effective dielectric function. The latter is obtained by substituting the correlator for the electric-field fluctuations of Eq. (36) into Eqs. (12) and (13), giving us

$$\tilde{\varepsilon}_{ij}(\mathbf{r};\omega) = \varepsilon_{ij}(\mathbf{r};\omega) + \int d\omega' N_{ij}(\mathbf{r};\omega,\omega'), \quad (37)$$

$$N_{ij}(\mathbf{r};\omega,\omega') = \sum_{m=0}^{N} 3\chi_{ijkl}^{(3)}(\mathbf{r};-\omega,\omega,\omega',-\omega')$$
$$\times \frac{b_m(\omega')}{2i}(\tilde{\mathbb{G}}\tilde{\mathbb{V}}_m\tilde{\mathbb{G}}^{\dagger})_{\text{AH}}(\mathbf{r},\mathbf{r};\omega')_{kl}. \quad (38)$$

These expressions reduce to the equilibrium cases $\tilde{\varepsilon}_{ij}^{\text{eq}}$ and N_{ij}^{eq} of Eqs. (25) and (26) if all temperatures are equal. This is because, by definition,

$$\sum_{m=0}^{\infty} \tilde{\mathbb{V}}_m = -\tilde{\mathbb{G}}^{-1}. \quad (39)$$

Note that the sum starts at 0, so that it contains also the famous environment dust [34]. If all temperatures are equal, then one recovers $\sum_m \frac{b(\omega')}{2i}(\tilde{\mathbb{G}}\tilde{\mathbb{V}}_m\tilde{\mathbb{G}}^{\dagger})_{\text{AH}} = \frac{b(\omega')}{2i}\tilde{\mathbb{G}}_{\text{AH}}$, as in the equilibrium expression.

It is instructive and useful to isolate the nonequilibrium contribution of the effective dielectric function. It is defined as

$$N^{\text{neq}} = N - N^{\text{eq}}, \quad (40)$$

where N^{eq} is the equilibrium limit corresponding to the temperature at the position where N is evaluated. For $\mathbf{r}$ located inside object n, it reads

$$N_{ij}^{\text{neq}}(\mathbf{r} \in V_n;\omega,\omega')$$
$$= 3\chi_{ijkl}^{(3)}(\mathbf{r};-\omega,\omega,\omega',-\omega') \sum_{m=0}^{N} \frac{b_m(\omega') - b_n(\omega')}{2i}$$
$$\times (\tilde{\mathbb{G}}\tilde{\mathbb{V}}_m\tilde{\mathbb{G}}^{\dagger})_{\text{AH}}(\mathbf{r},\mathbf{r};\omega')_{kl}. \quad (41)$$

This expression depends on the temperatures of all objects, because the nonlinear term couples the fluctuations in the different objects.

More precisely, in the above expression only objects with $T_m \neq T_n$ contribute, where T_n is the temperature at $\mathbf{r}$. This has an important implication regarding the mentioned divergence of $\mathbb{G}$ at coinciding points. Because $\tilde{\mathbb{V}}_m$ is only nonzero inside body m and $\mathbf{r}$ is inside body n, the two Green's functions in Eq. (41) connect points in *different* objects only (the sum

does not contain the term $m = n$). The expression for $\mathbb{G}$ evaluated at two different points is notably finite. We thus find that the deviation of the effective dielectric function from its equilibrium value is a quantity that can be predicted within this framework.

If we have only a single body in vacuum, then the above expression simplifies to

$$N_{ij,\text{single}}^{\text{neq}}(\mathbf{r};\omega,\omega') = 3\chi_{ijkl}^{(3)}(\mathbf{r};-\omega,\omega,\omega',-\omega')$$
$$\times [b_{\text{env}}(\omega') - b_{\text{obj}}(\omega')]$$
$$\times (\tilde{\mathbb{G}}\,\text{Im}[-\mathbb{G}_0^{-1}]\tilde{\mathbb{G}}^{\dagger})(\mathbf{r},\mathbf{r};\omega')_{kl}. \quad (42)$$

We see that if there is only a single body in vacuum, the effective dielectric function depends on the temperature of the environment, in stark contrast to linear materials.

C. Heat radiation and transfer

In Appendix, we show that the net heat radiated from a body can be written in terms of the fluctuation correlations [see Eq. (A6)], starting from the Poynting vector. By using the result obtained in Eq. (36), the net heat (including incoming and outgoing radiation) from object n in the presence of $N - 1$ other objects can be written as (derivation in Appendix)

$$H_n = -\frac{1}{4\mu_0} \sum_{m=0}^{N} \int \frac{d\omega}{2\pi} \frac{1}{\omega} [b_n(\omega) - b_m(\omega)]$$
$$\times \text{Tr}[(\tilde{\mathbb{V}}_n)_{\text{AH}}\tilde{\mathbb{G}}(\tilde{\mathbb{V}}_m)_{\text{AH}}\tilde{\mathbb{G}}^{\dagger}]. \quad (43)$$

We were not able to show that $\tilde{\mathbb{V}}$ and therefore $\tilde{\mathbb{G}}$ are generally symmetric (implying microreversibility [34]) in the considered nonequilibrium situation. This is why Eq. (43) is not symmetric in indices n and m. If $\tilde{\mathbb{V}}$ is symmetric, the slightly simpler Eq. (A12) follows (see Appendix), which is then symmetric in n and m like the corresponding formula for linear systems [31,38].

Equation (43) reiterates the statement that in order to calculate the heat radiation or transfer, all we need to know are the effective linear properties of the system—the effective nonequilibrium dielectric function or linear response. Recall that in the nonlinear case, the effective dielectric function depends on the geometry and temperature of the rest of the system in a nontrivial fashion as per Eqs. (37) and (38).

Equation (43), apart from the mentioned issue about symmetries, is similar in form to trace formulas obtained in Refs. [31,38–40]. Reference [31] writes it in terms of the scattering or T-operators,

$$\tilde{\mathbb{T}} = \mathbb{H}_0\tilde{\mathbb{G}}\tilde{\mathbb{V}}, \quad (44)$$

where the tilde again denotes the physical linear response. More precisely, formula Eq. (A12) in Appendix, the symmetric version of Eq. (43), is equivalent to the expressions of Refs. [31,41] when reduced to the linear system.

For a single body in vacuum (assuming symmetry of $\tilde{\mathbb{V}}$), the heat radiation takes the form that is reminiscent of the corresponding result for linear systems [31],

$$H = \frac{1}{\mu_0} \int \frac{d\omega}{2\pi} \frac{1}{\omega} [b_{\text{obj}}(\omega) - b_{\text{env}}(\omega)]$$
$$\times \text{Tr}(\text{Im}[\mathbb{G}_0]\text{Im}\tilde{\mathbb{T}} - \text{Im}[\mathbb{G}_0]\tilde{\mathbb{T}}\,\text{Im}[\mathbb{G}_0]\tilde{\mathbb{T}}^*). \quad (45)$$
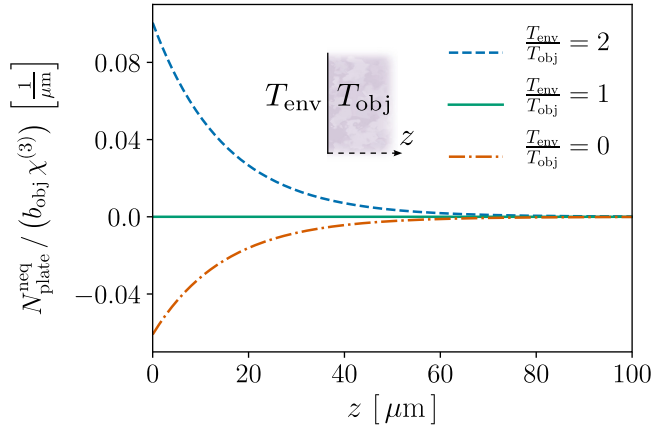
FIG. 4. The nonequilibrium effective dielectric function inside a semi-infinite plate, where z is the distance from the surface. $\varepsilon = 4 + i$, $\lambda_0' = 2\pi \frac{c}{\omega'} = 50 \ \mu$m, and $T_{\text{obj}} = \frac{\hbar \omega'}{k_B} \approx 287$ K were fixed while T_{env} was varied between zero and $2 T_{\text{obj}}$.

This equation follows from Eq. (A12) when reduced to a single body and substituting the identities (44) and $\tilde{\mathbb{G}} = \mathbb{G}_0 + \mathbb{G}_0 \tilde{\mathbb{T}} \mathbb{G}_0$.

D. Heat radiation of a semi-infinite plate: Kirchhoff's law and Planck's law

We proceed by computing the nonequilibrium part of the effective dielectric function for a single plate using Eq. (42). To simplify the following discussion, we consider again a highly symmetric material with $\varepsilon_{ij} = \varepsilon \delta_{ij}$ and $\chi_{ijkl}^{(3)} = \chi^{(3)} \delta_{ij} \delta_{kl}$. In that case, the effective dielectric function is diagonal, and we obtain from Eq. (42)

$$\bar{\varepsilon}_{ij}(\mathbf{r}; \omega) = \delta_{ij} \left[\varepsilon(\mathbf{r}; \omega) + \int d\omega' N(\mathbf{r}; \omega, \omega') \right], \quad (46)$$

$$N^{\text{neq}}(\mathbf{r} \in V_n; \omega, \omega') = 3 \chi^{(3)}(\mathbf{r}; -\omega, \omega, \omega', -\omega')$$

$$\times \sum_{m=0}^{N} [b_{\text{env}}(\omega') - b_{\text{obj}}(\omega')]$$

$$\times \left(\tilde{\mathbb{G}} \text{Im}[-\mathbb{G}_0^{-1}] \tilde{\mathbb{G}}^* \right)(\mathbf{r}, \mathbf{r}; \omega')_{kk}, \quad (47)$$

where $N = N^{\text{eq}} + N^{\text{neq}}$, with the equilibrium part given in Eq. (30).

The numerical results (using again the Green's function for planar systems from Ref. [37]) are shown in Fig. 4 for $\varepsilon = 4 + i$. The term $\tilde{\mathbb{G}} \text{Im}[-\mathbb{G}_0^{-1}] \tilde{\mathbb{G}}^*$ was evaluated using so-called environment dust [31,34], which is finite. We see that there is no interference pattern forming in $N_{\text{plate}}^{\text{neq}}$, which is in contrast to the equilibrium dielectric function. Deep inside the material, i.e., for large z, $N_{\text{plate}}^{\text{neq}}$ vanishes. For nonabsorbing media, $N_{\text{plate}}^{\text{neq}}$ is independent of z (not shown).

Notably, the nonequilibrium contribution changes sign with $\Delta T = T_{\text{obj}} - T_{\text{env}}$, as can be seen from Eq. (47) since b is monotonic in temperature. This implies that with a nonzero imaginary part of $\chi^{(3)}$ (of either sign), it is in principle possible to obtain $\text{Im}\bar{\varepsilon} < 0$, i.e., a medium with a negative absorption at the given frequency. This means that a probing

wave would experience gain in an otherwise passive system due to interactions with the nonequilibrium fluctuations. While there is no fundamental principle ruling this out (in contrast to the equilibrium case), it remains to be seen whether materials with suitable combinations of $\chi^{(1)}$ and $\chi^{(3)}$ exist to display any such phenomenon.

More importantly, Fig. 4 shows that the effective dielectric function (which determines heat radiation) of the plate depends on the temperature of the environment, not just the plate itself. This is a clear nonlinear effect (it is absent for linear materials), which can be measured experimentally. In the case of multiple bodies, the effective dielectric function also depends on the temperatures of other objects (in addition to their positions as in the equilibrium case).

This observation has implications for Kirchhoff's law of radiation. It states that, in equilibrium, the absorptivity and emissivity of a body are equal. It is thus a variant of the principle of detailed balance. Although it strictly only holds in equilibrium, it has been shown to be valid (and often used) for linear bodies out of equilibrium as well (see, e.g., the discussion in Ref. [31]). The analysis in Sec. III A, as well as Fig. 4, shows an interesting variant of this law: Because the nonequilibrium effective dielectric function can be used for computation of absorption as well as emission coefficients, Kirchoff's law stays indeed valid in the considered order of $\chi^{(3)}$. However, these coefficients depend on the temperature of the environment. This means that the experiments measuring the emission and absorption need to be performed in exactly the same conditions (same temperatures, surrounding bodies, etc.).

We noted in the previous subsection that we cannot prove the symmetry of the nonequilibrium potential $\tilde{\mathbb{V}}$ in general, mostly due to the lack of symmetries of $\chi_{ijkl}^{(3)}$. For the highly symmetric version used in Fig. 4, $\chi_{ijkl}^{(3)} = \chi^{(3)} \delta_{ij} \delta_{kl}$, $\tilde{\mathbb{V}}$ is symmetric. If $\tilde{\mathbb{V}}$ may turn out to be nonsymmetric in other cases, this would manifest a more dramatic breakdown of Kirchoff's law, because such nonsymmetric nonequilibrium $\tilde{\mathbb{V}}$ would explicitly break microreversibility. This has to be investigated in the future.

Noting the change of the dielectric function in Fig. 4, one may ask whether the plate in that figure may radiate stronger than a blackbody. This question is immediately answered from the observation that the radiation of the plate is given by known formulas (see, e.g., [31]), where the dielectric function ε needs to be replaced by the effective one of Fig. 4. While explicit computation of the corresponding Fresnel coefficients for spatially varying ε may be challenging, a general statement is nevertheless possible. The radiation of a planar surface, irrespective of the values of $\varepsilon(\omega)$, is bound by the radiation of a blackbody [see, e.g., Eq. (26) of Ref. [42] or Ref. [43]]. Therefore, for the radiation linear in $\chi^{(3)}$, we have

$$d\omega \frac{\mathcal{H}(\omega)}{A} \leqslant d\omega \frac{\hbar \omega^3}{4\pi^2 c^2} \left[\exp\left(\frac{\hbar \omega}{k_B T} \right) - 1 \right]^{-1}, \quad (48)$$

where the radiation of the plate per surface area A is $H/A = \int_0^\infty d\omega \, \mathcal{H}(\omega)/A$. The radiation of a planar surface thus obeys the fundamental bounds implied by Planck's law. We note, however, that Eq. (48) relies on the symmetry of $\tilde{\mathbb{V}}$; the

possibility of nonsymmetric $\tilde{\mathbb{V}}$ out of equilibrium must be investigated in the future.

E. Radiation of a sphere: Negative radiation

We now turn to the radiation of a nanosphere. We start by evaluating the isotropic effective dielectric function in Eq. (46) using also the simplification $\chi_{ijkl}^{(3)} = \chi^{(3)}\delta_{ij}\delta_{kl}$ as in Sec. II D. In the limit where the radius is much smaller than the thermal wavelength $\lambda_T = \frac{\hbar c}{k_B T}$ and the skin depth $\delta = \frac{1}{\text{Im}\sqrt{\varepsilon\mu}}\frac{c}{\omega}$, the Green's function with one point outside and one point inside the sphere is given by $\mathbb{G} = \frac{3}{\varepsilon+2}\mathbb{G}_0$. Using $\text{Im}\mathbb{G}_0(\mathbf{r},\mathbf{r};\omega)_{ij} = \frac{1}{6\pi}\frac{\omega}{c}\delta_{ij}$, we have from Eq. (47)

$$N_{\text{sphere}}^{\text{neq}}(\omega,\omega') = -\frac{3}{2\pi}\frac{\omega'}{c}\chi^{(3)}(-\omega,\omega,\omega',-\omega')$$
$$\times \left|\frac{3}{\varepsilon(\omega')+2}\right|^2 [b_{\text{obj}}(\omega') - b_{\text{env}}(\omega')]. \quad (49)$$

This function is spatially constant inside the (pointlike) sphere. We may now use this dielectric function to compute the effective version of the polarizability,

$$\alpha \equiv \frac{\varepsilon-1}{\varepsilon+2}R^3, \quad (50)$$

which governs the radiation of small spheres [44,45]. By substituting the effective dielectric function into Eq. (50) and expanding in $N_{\text{sphere}}^{\text{neq}}$, we obtain

$$\tilde{\alpha}(\omega) = \tilde{\alpha}^{\text{eq}}\left[1 + \frac{3}{(\varepsilon-1)(\varepsilon+2)}\int d\omega' N_{\text{sphere}}^{\text{neq}}(\omega,\omega')\right], \quad (51)$$

where $\tilde{\alpha}^{\text{eq}}$ is the (effective) polarizability in equilibrium. The radiation of a sphere is then given by

$$H = 4\frac{\varepsilon_0}{\pi^2 c}\int d\omega\, \omega^2 [b_{\text{obj}}(\omega) - b_{\text{env}}(\omega)]\text{Im}\tilde{\alpha}(\omega). \quad (52)$$

$\text{Im}\tilde{\alpha}$, which is manifestly positive in equilibrium, may in principle be negative in the considered nonequilibrium situation, for suitable regimes regarding the sign of $(T_{\text{env}} - T_{\text{obj}})$ as well as $\text{Im}\chi^{(3)}(-\omega,\omega)$. As mentioned before, we see no fundamental reason that forbids such an occurrence, and it will be interesting to see whether it can exist in practice.

An instructive extreme case to consider is $\text{Im}\tilde{\varepsilon}^{\text{eq}} = \text{Im}\tilde{\alpha}^{\text{eq}} = 0$. This is a particle that does not absorb or emit energy in equilibrium, so that any absorption is only due to the finite $\text{Im}\chi^{(3)}$. Using Eqs. (49), (51), and (52), we then arrive at

$$H = -54\frac{\varepsilon_0}{\pi^3 c^3}\int d\omega \int d\omega' \omega^2 \omega' \text{Im}\chi^{(3)}(-\omega,\omega,\omega',-\omega')$$
$$\times \frac{[b_{\text{obj}}(\omega) - b_{\text{env}}(\omega)][b_{\text{obj}}(\omega') - b_{\text{env}}(\omega')]}{[\varepsilon(\omega)+2]^2[\varepsilon(\omega')+2]^2}. \quad (53)$$

The first observation regarding Eq. (53) is that the heat radiation of the sphere remains *unchanged* if the temperatures of the object and the environment are interchanged. Considering specifically $\text{Im}\chi^{(3)} < 0$, which is a typical observed case (see, e.g., Ref. [46] for metal-infused glasses), Eq. (53) yields $H > 0$. This means energy flowing away from the sphere for any combination of temperatures. For $T_{\text{env}} > T_{\text{obj}}$, this corresponds

to a flow of energy from a cold sphere to a hot environment. While this cannot be ruled out for a particular frequency, from thermodynamic considerations we expect that the total heat (after integration over all frequencies) flows from the hotter to the colder body.

IV. SUMMARY

In stochastic nonlinear optical systems, fluctuating fields and induced fields couple, which gives rise to a variety of phenomena that cannot be observed in purely linear systems. We show that fluctuation effects, such as the Casimir effect or heat radiation, can be described via known formulas, however using an effective dielectric function as input. This dielectric function depends on the shape of the objects, their relative position, and also on the temperatures of all objects in the system.

The divergence of the electromagnetic Green's function at coinciding points prevents a straight computation of the effective dielectric function on a macroscopic level for absorbing materials. It is nevertheless possible to circumvent this issue by considering *measurable* quantities. Using this principle, the dependence of the dielectric function on the distance between the objects is accessible theoretically. This is also true for the dependence of the dielectric function of one object on the temperatures of the other objects in a nonequilibrium scenario.

In addition to effects in equilibrium, we saw profound and thought-provoking implications in the case in which the temperatures of objects (and the environment) are different. We discussed the applicability of Kirchoff's law of radiation as well as the fundamental bounds of radiation of a planar surface. For a nanosphere out of equilibrium, we found that the spectral emission can surprisingly be negative in certain cases.

Overall, we saw that both equilibrium and nonequilibrium phenomena are intricately affected by nonlinear optical properties. By using the fluctuational electrodynamics framework, these results are also applicable for any geometry or materials. In the future, it may also be generalized to nonzero external fields, possibly allowing for even more control over the effect of the nonlinearities.

ACKNOWLEDGMENTS

APPENDIX: HEAT RADIATION AND TRANSFER FROM FLUCTUATIONAL ELECTRODYNAMICS

The total energy transmitted across a surface Σ_n surrounding object n is given by

$$H_n = \oint_{\Sigma_n} da\langle\mathbf{S}\rangle \cdot \mathbf{n}, \quad (A1)$$

where $\langle\mathbf{S}\rangle = \langle\mathbf{E}\times\mathbf{H}\rangle$ is the time average of the Poynting vector and $\mathbf{n}$ is a normal vector on Σ_n. The force on the object (called Casimir force in equilibrium) can be obtained from the same expression with the Poynting vector replaced by the Maxwell stress tensor $\sigma = \varepsilon_0(\langle\mathbf{E}\otimes\mathbf{E}\rangle - \frac{1}{2}\mathbf{E}^2) + \frac{1}{\mu_0}(\langle\mathbf{B}\otimes\mathbf{B}\rangle - \frac{1}{2}\mathbf{B}^2)$.

For stationary systems, the correlator $\langle \mathbf{E}(t) \otimes \mathbf{H}(t') \rangle$ depends only on time differences. We can therefore define a spectral density of the expectation value as

$$\langle \mathbf{E}(t) \otimes \mathbf{H}(t') \rangle = \int \frac{d\omega}{2\pi} e^{i\omega(t-t')} \langle \mathbf{E} \otimes \mathbf{H}^* \rangle_\omega, \qquad (A2)$$

where the integration is over positive and negative frequencies. Since $\langle \mathbf{E}(t) \otimes \mathbf{H}(t') \rangle$ is a real quantity, the real (imaginary) part of the spectrum is an even (odd) function of the frequency. Therefore, only the real part remains in the Poynting vector,

$$\langle \mathbf{S} \rangle = \int \frac{d\omega}{2\pi} \mathrm{Re} \langle \mathbf{E} \times \mathbf{H}^* \rangle_\omega. \qquad (A3)$$

Using the divergence theorem, we can rewrite Eq. (A1) as

$$H_n = \int_{V_n} dV \langle \mathbf{\nabla} \cdot \mathbf{S} \rangle$$
$$= \int \frac{d\omega}{2\pi} \int_{V_n} dV \, \mathrm{Re} \langle \mathbf{\nabla} \cdot (\mathbf{E}_\omega \times \mathbf{H}_\omega^*) \rangle. \qquad (A4)$$

With the Maxwell-Faraday equation $i\omega \mathbf{B}_\omega = \mathbf{\nabla} \times \mathbf{E}_\omega$, we can write for nonmagnetic materials ($\mu = 1$)

$$H_n = \frac{1}{\mu_0} \int \frac{d\omega}{2\pi} \frac{1}{\omega} \int_{V_n} dV \, \mathrm{Im} \langle \mathbf{E} \cdot (\mathbf{\nabla} \times \mathbf{\nabla} \times \mathbf{E})^* \rangle_\omega. \quad (A5)$$

By using the symmetric operator $\mathbb{G}_0^{-1} = \mathbb{H}_0 = \mathbf{\nabla} \times \mathbf{\nabla} \times -\frac{\omega^2}{c^2} \mathbb{I}$ and remembering that $\mathbf{A} \cdot \mathbf{B} = \mathbf{B} \cdot \mathbf{A}$, we get

$$H_n = -\frac{1}{\mu_0} \int \frac{d\omega}{2\pi} \frac{1}{\omega} \mathrm{Tr}_n \mathrm{Im} \big[\mathbb{G}_0^{-1} \langle \mathbf{E} \otimes \mathbf{E}^* \rangle_\omega \big], \qquad (A6)$$

where Tr_n denotes a (pseudo) trace, which is restricted to volume V_n. Here we see the imaginary part of the electric-field correlator.

Let us first check the case in which all temperatures are equal, where we expect no heat radiation. Taking Eq. (18) for the correlator, we get

$$H_n^{\mathrm{eq}} = \frac{1}{2\mu_0} \int \frac{d\omega}{2\pi} \frac{1}{\omega} b(\omega) \mathrm{Tr}_n \mathrm{Re} \big[\mathbb{G}_0^{-1} \tilde{\mathbb{G}}_{\mathrm{AH}} \big]. \qquad (A7)$$

With the identities $\tilde{\mathbb{G}} = \mathbb{G}_0 (\mathbb{I} + \tilde{\mathbb{V}} \tilde{\mathbb{G}}) = (\mathbb{I} + \tilde{\mathbb{G}} \tilde{\mathbb{V}}) \mathbb{G}_0$, we can obtain an expression with a full trace,

$$H_n^{\mathrm{eq}} = \frac{1}{2\mu_0} \int \frac{d\omega}{2\pi} \frac{b(\omega)}{\omega} \mathrm{Tr} \, \mathrm{Re} [(\tilde{\mathbb{V}}_n \tilde{\mathbb{G}})_{\mathrm{AH}}]. \qquad (A8)$$

Using the properties of the trace, $\mathrm{Tr} \mathbb{A}^{\mathrm{T}} = \mathrm{Tr} \mathbb{A}$ and $\mathrm{Tr}[\mathbb{A}\mathbb{B}] = \mathrm{Tr}[\mathbb{B}\mathbb{A}]$, we see that

$$H_n^{\mathrm{eq}} = -\frac{1}{\mu_0} \int \frac{d\omega}{2\pi} \frac{b(\omega)}{\omega} \mathrm{Tr} \, \mathrm{Im}[\mathrm{Im}(\tilde{\mathbb{V}}_n \tilde{\mathbb{G}})] = 0. \qquad (A9)$$

Therefore, we have shown for consistency that even with asymmetric response (no micro-reversibility) there is no heat transfer if the temperatures are equal.

Using Eq. (36) for the correlator with unequal temperatures, we obtain in a similar fashion to that mentioned above,

$$H_n = -\frac{1}{4\mu_0} \int \frac{d\omega}{2\pi} \frac{1}{\omega} b_n(\omega) \mathrm{Tr}[(\tilde{\mathbb{V}}_n)_{\mathrm{AH}} \tilde{\mathbb{G}}_{\mathrm{AH}}]$$
$$+ \frac{1}{4\mu_0} \sum_{m=0}^{N} \int \frac{d\omega}{2\pi} \frac{b_m(\omega)}{\omega} \mathrm{Tr}[(\tilde{\mathbb{V}}_n)_{\mathrm{AH}} \tilde{\mathbb{G}} (\tilde{\mathbb{V}}_m)_{\mathrm{AH}} \tilde{\mathbb{G}}^\dagger]. \qquad (A10)$$

For the final form, we subtract the contribution of a pseudosystem, where all bodies are at a temperature T_n (therefore giving no contribution to heat transfer), but with the response corresponding to the original out-of-equilibrium system,

$$H_n = -\frac{1}{4\mu_0} \sum_{m=0}^{N} \int \frac{d\omega}{2\pi} \frac{1}{\omega} [b_n(\omega) - b_m(\omega)]$$
$$\times \mathrm{Tr}[(\tilde{\mathbb{V}}_n)_{\mathrm{AH}} \tilde{\mathbb{G}} (\tilde{\mathbb{V}}_m)_{\mathrm{AH}} \tilde{\mathbb{G}}^\dagger]. \qquad (A11)$$

Note that the terms with $T_m = T_n$ (including $m = n$) do not contribute to heat radiation.

Furthermore, if $\tilde{\mathbb{V}}$ and therefore $\tilde{\mathbb{G}}$ are symmetric (implying microreversibility [34]), then we can further simplify Eq. (A11) as

$$H_n = \frac{1}{\mu_0} \sum_{m=0}^{N} \int \frac{d\omega}{2\pi} \frac{1}{\omega} [b_n(\omega) - b_m(\omega)]$$
$$\times \mathrm{Tr}(\mathrm{Im}[\tilde{\mathbb{V}}_n] \tilde{\mathbb{G}} \mathrm{Im}[\tilde{\mathbb{V}}_m] \tilde{\mathbb{G}}^*). \qquad (A12)$$

This is the final form of the net heat radiation from object n to the environment and other objects. Note that this equation has a similar structure to expressions in Refs. [31,38–40]. The imaginary parts of the potentials represent the sources and are connected by the Green's functions, representing transmission coefficients [39,40].

[1] H. B. Casimir, Proc. K. Ned. Akad. Wet. **51**, 793 (1948).

[2] E. M. Lifshitz, Sov. Phys. JETP **2**, 73 (1956).

[3] S. M. Rytov, Y. A. Kravtsov, and V. I. Tatarskii, *Principles of Statistical Radiophysics 3* (Springer, Berlin, 1989).

[4] D. Polder and M. Van Hove, Phys. Rev. B **4**, 3303 (1971).

[5] P. W. Milonni, *The Quantum Vacuum* (Academic, San Diego, 1994).

[6] M. Bordag, G. L. Klimchitskaya, U. Mohideen, and V. M. Mostepanenko, *Advances in the Casimir Effect* (Oxford University Press, Oxford, 2009).

[7] S. K. Lamoreaux, Phys. Rev. Lett. **78**, 5 (1997).

[8] U. Mohideen and A. Roy, Phys. Rev. Lett. **81**, 4549 (1998).

[9] G. Bressi, G. Carugno, R. Onofrio, and G. Ruoso, Phys. Rev. Lett. **88**, 041804 (2002).

[10] A. Kittel, W. Müller-Hirsch, J. Parisi, S.-A. Biehs, D. Reddig, and M. Holthaus, Phys. Rev. Lett. **95**, 224301 (2005).

[11] J. M. Obrecht, R. J. Wild, M. Antezza, L. P. Pitaevskii, S. Stringari, and E. A. Cornell, Phys. Rev. Lett. **98**, 063201 (2007).

[12] E. Rousseau, A. Siria, G. Jourdan, S. Volz, F. Comin, J. Chevrier, and J.-J. Greffet, Nat. Photon. **3**, 514 (2009).

[13] S. Shen, A. Narayanaswamy, and G. Chen, Nano Lett. **9**, 2909 (2009).

[14] R. S. Ottens, V. Quetschke, S. Wise, A. A. Alemi, R. Lundock, G. Mueller, D. H. Reitze, D. B. Tanner, and B. F. Whiting, Phys. Rev. Lett. **107**, 014301 (2011).

[15] K. Kim, B. Song, V. Fernández-Hurtado, W. Lee, W. Jeong, L. Cui, D. Thompson, J. Feist, M. T. H. Reid, F. J. García-Vidal, J. C. Cuevas, E. Meyhofer, and P. Reddy, Nature (London) **528**, 387 (2015).

[16] M. Schubert and B. Wilhelmi, *Nonlinear Optics and Quantum Electronics* (Wiley-Interscience, New York, 1986).

[17] R. W. Boyd, *Nonlinear Optics* (Elsevier, Amsterdam, 2008).

[18] N. G. van Kampen, in *Fluctuation Phenomena in Solids*, edited by R. E. Burgess (Academic, New York and London, 1965).

[19] R. L. Stratonovich, *Nonlinear Nonequilibrium Thermodynamics I* (Springer, Berlin, Heidelberg, 1992).

[20] P. D. Drummond, Phys. Rev. A **42**, 6845 (1990).

[21] S. Scheel and D.-G. Welsch, Phys. Rev. Lett. **96**, 073601 (2006).

[22] F. Kheirandish, E. Amooghorban, and M. Soltani, Phys. Rev. A **83**, 032507 (2011).

[23] D. Kysylychyn, V. Piatnytsia, and V. Lozovski, Phys. Rev. E **88**, 052403 (2013).

[24] K. Makhnovets and A. Kolezhuk, Materwiss. Werksttech. **47**, 222 (2016).

[25] H. Soo, D. S. Dean, and M. Krüger, Phys. Rev. E **95**, 012151 (2017).

[26] H. Soo and M. Krüger, Europhys. Lett. **115**, 41002 (2016).

[27] C. Khandekar, A. Pick, S. G. Johnson, and A. W. Rodriguez, Phys. Rev. B **91**, 115406 (2015).

[28] C. Khandekar and A. W. Rodriguez, Opt. Express **25**, 23164 (2017).

[29] J. D. Jackson, *Classical Electrodynamics* (Wiley, New York, 1998).

[30] S. J. Rahi, T. Emig, N. Graham, R. L. Jaffe, and M. Kardar, Phys. Rev. D **80**, 085021 (2009).

[31] M. Krüger, G. Bimonte, T. Emig, and M. Kardar, Phys. Rev. B **86**, 115423 (2012).

[32] R. Kubo, Rep. Prog. Phys. **29**, 255 (1966).

[33] M. L. Levin and S. Rytov, *Theory of Equilibrium Thermal Fluctuations in Electrodynamics* (Nauka, Moscow, 1967).

[34] W. Eckhardt, Phys. Rev. A **29**, 1991 (1984).

[35] M. Kardar, *Statistical Physics of Fields* (Cambridge University Press, Cambridge, 2007).

[36] R. Matloob, Phys. Rev. A **60**, 50 (1999).

[37] P. Johansson, Phys. Rev. B **83**, 195408 (2011).

[38] A. W. Rodriguez, M. T. Homer Reid, and S. G. Johnson, Phys. Rev. B **86**, 220302 (2012).

[39] P. Ben-Abdallah and K. Joulain, Phys. Rev. B **82**, 121419 (2010).

[40] S. A. Biehs, E. Rousseau, and J. J. Greffet, Phys. Rev. Lett. **105**, 234301 (2010).

[41] B. Müller, R. Incardone, M. Antezza, T. Emig, and M. Krüger, Phys. Rev. B **95**, 085413 (2017).

[42] V. A. Golyk, M. Krüger, and M. Kardar, Phys. Rev. E **85**, 046603 (2012).

[43] S.-A. Biehs and P. Ben-Abdallah, Phys. Rev. B **93**, 165405 (2016).

[44] L. Tsang, J. A. Kong, and K.-H. Ding, *Scattering of Electromagnetic Waves: Theories and Applications* (Wiley, New York, 2000).

[45] C. F. Bohren and D. R. Huffman, *Absorption and Scattering of Light by Small Particles* (Wiley, New York, 2008).

[46] Y. Hamanaka, A. Nakamura, N. Hayashi, and S. Omi, J. Opt. Soc. Am. B **20**, 1227 (2003).