



## Research article

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# Macroscopic QED for quantum nanophotonics: emitter-centered modes as a minimal basis for multiemitter problems

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**Abstract:** We present an overview of the framework of macroscopic quantum electrodynamics from a quantum nanophotonics perspective. Particularly, we focus our attention on three aspects of the theory that are crucial for the description of quantum optical phenomena in nanophotonic structures. First, we review the light–matter interaction Hamiltonian itself, with special emphasis on its gauge independence and the minimal and multipolar coupling schemes. Second, we discuss the treatment of the external pumping of quantum optical systems by classical electromagnetic fields. Third, we introduce an exact, complete, and minimal basis for the field quantization in multiemitter configurations, which is based on the so-called emitter-centered modes. Finally, we illustrate this quantization approach in a particular hybrid metallodielectric geometry: two quantum emitters placed in the vicinity of a dimer of Ag nanospheres embedded in a SiN microdisk.

**Keywords:** emitter-centered modes; hybrid cavities; macroscopic quantum electrodynamics; quantum nanophotonics.

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## 1 Introduction

In principle, quantum electrodynamics (QED) provides an “exact” approach for treating electromagnetic (EM) fields, charged particles, and their interactions, within a full quantum field theory where both matter and light are second quantized (i.e., both photons and matter particles can be created and annihilated). However, this approach is not very useful for the treatment of many effects of interest in fields such as (nano)photonics and quantum optics, which take place at “low” energies (essentially, below the rest mass energy of electrons), where matter constituents are stable and neither created nor destroyed, and additionally, there are often “macroscopic” structures such as mirrors, photonic crystals, metallic nanoparticles etc. involved. Owing to the large number of material particles (on the order of the Avogadro constant,  $\approx 6 \times 10^{23}$ ), it then becomes unthinkable to treat the electrons and nuclei in these structures individually. At the same time, a sufficiently accurate description of these structures is usually given by the macroscopic Maxwell equations, in which the material response is described by the constitutive relations of macroscopic electromagnetism. In many situations, it is then desired to describe the interactions between light and matter in a setup where there are one or a few microscopic “quantum emitters” (such as atoms, molecules, quantum dots, etc.) and additionally a “macroscopic” material structure whose linear response determines the local modes of the EM field interacting with the quantum emitter(s).

The quantization of the EM field in such arbitrary material environments, i.e., the construction of a second quantized basis for the medium-assisted EM field that takes into account the presence of the “macroscopic” material structure, is a longstanding problem in QED. The most immediate strategy is to calculate the (classical) EM modes of a structure and to quantize them by normalizing their stored energy to that of a single photon at the mode frequency,  $\hbar\omega$  [1]. However, an important point to remember here is that, even for lossless materials, EM modes always

form a continuum in frequency, i.e., there exist modes at any positive frequency  $\omega$ . Consequently, there are in general no truly bound EM modes, and what is normally thought of as an isolated “cavity mode” is more correctly described as a resonance embedded in the continuum, i.e., a quasi-bound state that decays over time through emission of radiation. An interesting exception here are guided modes in systems with translational invariance (i.e., where momentum in one or more dimensions is conserved), as modes lying outside the light cone  $\omega = ck$  then do not couple to free-space radiation [2]. An additional exception is given by “bound states in the continuum” [3], which arise owing to destructive interference between different resonances.

As a further obstacle to a straightforward quantization strategy as described above, the response functions describing material structures are necessarily dissipative owing to causality (as encoded in, e.g., the Kramers–Kronig relations). When these losses cannot be neglected, quantization is complicated even further by the difficulty to define the energy density of the EM field inside the lossy material [4–6].

Given all the points above, it is not surprising that there are many different approaches to quantizing EM modes in lossy material systems [2, 7–15]. In the following, we give a concise overview of a particularly powerful formal approach that resolves these problems, called macroscopic QED [16–24]. While there are excellent reviews of this general framework available (e.g., [22, 23]), we focus on its application in the context of quantum nanophotonics and strong light–matter coupling. In particular, we discuss the implications and lessons that can be taken from this approach on gauge independence and, in particular, the role of the so-called dipole self-energy term in the light–matter interaction in the Power–Zienau–Woolley (PZW) gauge, which has been the subject of some recent controversy [25–38]. We then review in detail a somewhat nonstandard formulation of macroscopic QED that allows one to construct a minimal quantized basis for the EM field interacting with a collection of multiple quantum emitters. This approach was first introduced by Buhmann and Welsch [39] and subsequently rediscovered independently by several other groups [40–44]. The fact that this very useful approach has been reinvented by different researchers over the past decade or so partially motivates the current article, which intends to give a concise and accessible overview, and presents some explicit relations that have (to our knowledge) not been published before. We also note that with “minimal basis,” we are here referring to a minimal *complete* basis for the medium-

assisted EM field, i.e., this basis contains all the information about the material structure playing the role of the cavity or antenna, and no approximations are made in obtaining it. This then makes it appropriate to serve as a starting point either for numerical treatments [45, 46] or for deriving simpler models where, e.g., the full EM spectrum is described by a few lossy modes [47].

In the final part of the article, we then present an application of the formalism to a specific problem, a hybrid dielectric-plasmonic structure [14, 48, 49]. In particular, we consider a dimer of metallic nanospheres placed within a dielectric microdisk, a geometry that is similar to that considered by Doeleman et al. [50].

## 2 Theory

Macroscopic QED provides a recipe for quantizing the EM field in any geometry, including with lossy materials. One particularly appealing aspect is that the full information about the quantized EM field is finally encoded in the (classical) EM dyadic Green’s function  $\mathbf{G}(\mathbf{r}, \mathbf{r}', \omega)$ . While there are several ways to derive the general formulation (see, e.g., the review by Scheel and Buhmann [22] for a discussion of various approaches), a conceptually simple way to understand the framework is to represent the material structures through a collection of fictitious harmonic oscillators coupled to the free-space EM field (which itself corresponds to a collection of harmonic oscillators [51]). Formally diagonalizing this system of coupled harmonic oscillators leads to a form where the linear response of the medium can be expressed through the coupling between the material oscillators and the EM field. The end result is that the fully quantized medium-supported EM field is represented by an infinite set of bosonic modes defined at each point in space and each frequency and labeled with index  $\lambda$  (see below),  $\hat{\mathbf{f}}_\lambda(\mathbf{r}, \omega)$ , which act as sources for the EM field through the classical Green’s function. These modes are called “polaritonic” as they represent mixed light–matter excitations [17]. While this is a completely general approach for quantizing the EM field in arbitrary structures, it cannot be used “directly” in practice owing to the extremely large number of modes that describe the EM field (a vectorial-valued four-dimensional continuum). Most uses of macroscopic QED thus apply this formalism to derive expressions where the explicit operators  $\hat{\mathbf{f}}_\lambda(\mathbf{r}, \omega)$  have been eliminated, e.g., through adiabatic elimination, perturbation theory, or the use of Laplace transform techniques [22–24, 52–55]. In particular, macroscopic QED has

been widely used in the context of dispersion forces, which are responsible for, e.g., the Casimir effect and Casimir–Polder force [23, 24].

## 2.1 Minimal coupling

In the following, we represent a short overview of the general theory of light–matter interactions in the framework of macroscopic QED. Since full details can be found in the literature [22, 23], we do not attempt to make this a fully self-contained overview, but rather highlight and discuss some aspects that are not within the traditional focus of the theory, in particular in the context of quantum nanophotonics.

The first step in the application of macroscopic QED is the separation of all matter present in the system to be treated into two distinct groups: one is the macroscopic structure (e.g., a cavity, plasmonic nanoantenna, photonic crystal, ...) that will be described through the constitutive relations of electromagnetism, while the other are the microscopic objects (atoms, molecules, quantum dots, ...) that are described as a collection of charged particles. This separation constitutes the basic approximation inherent in the approach and relies on the assumptions that macroscopic electromagnetism is valid for the material structure (the medium) and its interaction with the charged particles. While this is often an excellent approximation, some care has to be taken for separations in the subnanometer range, where the atomic structure of the material can have a significant influence [56–61]. One significant advantage of this approach is that the microscopic objects are governed by the “standard” Hamiltonian of charged particles interacting through the Coulomb force. They can thus be represented using standard approximations, e.g., using the methods of atomic and molecular physics and quantum chemistry to obtain few-level approximations, or also of solid-state physics to obtain effective descriptions of their band structure, although care has to be taken with gauge invariance when such approximations are performed [62].

For simplicity, we assume that the medium response is local and isotropic in space, such that it can be encoded in the position- and frequency-dependent scalar relative permittivity  $\epsilon(\mathbf{r}, \omega)$  and relative permeability  $\mu(\mathbf{r}, \omega)$  that describe the local matter polarization and magnetization induced by external EM fields<sup>1</sup>. The extension of the quantization scheme to nonlocal response functions can be

found in reference [22]. We directly give the Hamiltonian  $\mathcal{H} = \mathcal{H}_A + \mathcal{H}_F + \mathcal{H}_{AF}$  within the minimal coupling scheme [22, 23]

$$\mathcal{H}_A = \sum_{\alpha} \frac{\hat{\mathbf{p}}_{\alpha}^2}{2m_{\alpha}} + \sum_{\alpha \beta} \sum_{\alpha \neq \beta} \frac{q_{\alpha} q_{\beta}}{4\pi\epsilon_0 |\hat{\mathbf{r}}_{\alpha} - \hat{\mathbf{r}}_{\beta}|}, \quad (1a)$$

$$\mathcal{H}_F = \sum_{\lambda} \int_0^{\infty} d\omega \int d^3\mathbf{r} \hbar \omega \hat{\mathbf{f}}_{\lambda}^{\dagger}(\mathbf{r}, \omega) \hat{\mathbf{f}}_{\lambda}(\mathbf{r}, \omega), \quad (1b)$$

$$\mathcal{H}_{AF} = \sum_{\alpha} \left[ q_{\alpha} \hat{\phi}(\hat{\mathbf{r}}_{\alpha}) - \frac{q_{\alpha}}{m_{\alpha}} \hat{\mathbf{p}}_{\alpha} \cdot \hat{\mathbf{A}}(\hat{\mathbf{r}}_{\alpha}) + \frac{q_{\alpha}^2}{2m_{\alpha}} \hat{\mathbf{A}}^2(\hat{\mathbf{r}}_{\alpha}) \right]. \quad (1c)$$

Here, the “atomic” Hamiltonian  $\mathcal{H}_A$  describes a (nonrelativistic) collection of point particles with position and momentum operators  $\hat{\mathbf{r}}_{\alpha}$  and  $\hat{\mathbf{p}}_{\alpha}$  and charges and masses  $q_{\alpha}$  and  $m_{\alpha}$ . The field Hamiltonian  $\mathcal{H}_F$  is expressed in terms of the bosonic operators  $\hat{\mathbf{f}}_{\lambda}(\mathbf{r}, \omega)$  discussed above, which obey the commutation relations

$$\left[ \hat{\mathbf{f}}_{\lambda}(\mathbf{r}, \omega), \hat{\mathbf{f}}_{\lambda'}^{\dagger}(\mathbf{r}', \omega') \right] = \delta_{\lambda\lambda'} \boldsymbol{\delta}(\mathbf{r} - \mathbf{r}') \delta(\omega - \omega'), \quad (2a)$$

$$\left[ \hat{\mathbf{f}}_{\lambda}(\mathbf{r}, \omega), \hat{\mathbf{f}}_{\lambda'}(\mathbf{r}', \omega') \right] = \left[ \hat{\mathbf{f}}_{\lambda}^{\dagger}(\mathbf{r}, \omega), \hat{\mathbf{f}}_{\lambda'}^{\dagger}(\mathbf{r}', \omega') \right] = \mathbf{0}, \quad (2b)$$

where  $\boldsymbol{\delta}(\mathbf{r} - \mathbf{r}') = \delta(\mathbf{r} - \mathbf{r}') \mathbf{1}$ , and  $\mathbf{1}$  and  $\mathbf{0}$  are the Cartesian ( $3 \times 3$ ) identity and zero tensors, respectively. The index  $\lambda \in \{e, m\}$  labels the electric and magnetic contributions, with the magnetic contribution disappearing if  $\mu(\mathbf{r}, \omega) = 1$  everywhere in space. The particle-field interaction Hamiltonian  $\mathcal{H}_{AF}$  contains the interaction of the charges both with the electrostatic potential  $\hat{\phi}(\mathbf{r})$  and the vector potential  $\hat{\mathbf{A}}(\mathbf{r})$ , both of which can be expressed through the fundamental operators  $\hat{\mathbf{f}}_{\lambda}(\mathbf{r}, \omega)$  [22, 23]. Note that in the above, we have neglected magnetic interactions owing to particle spin. We explicitly point out that although we work in Coulomb gauge,  $\nabla \cdot \hat{\mathbf{A}}(\mathbf{r}) = 0$ , the electrostatic potential  $\hat{\phi}(\mathbf{r})$  is in general nonzero owing to the presence of the macroscopic material structure, which also implies that the name “ $\mathbf{p} \cdot \mathbf{A}$ -gauge,” which is sometimes employed for the minimal coupling scheme, is misleading in the presence of material bodies.

The light–matter interaction Hamiltonian can be simplified in the long-wavelength or dipole approximation, i.e., if we assume that the charged particles are sufficiently close to each other compared to the spatial scale of local field variations that a lowest order approximation of the positions of the charges relative to their center of mass position  $\hat{\mathbf{r}}_i$  is valid. For an overall neutral collection of charges, this leads to

$$\mathcal{H}_{AF} \approx -\hat{\mathbf{d}} \cdot \hat{\mathbf{E}}^{\parallel}(\hat{\mathbf{r}}_i) - \sum_{\alpha} \frac{q_{\alpha}}{m_{\alpha}} \hat{\mathbf{p}}_{\alpha} \cdot \hat{\mathbf{A}}(\hat{\mathbf{r}}_i) + \sum_{\alpha} \frac{q_{\alpha}^2}{2m_{\alpha}} \hat{\mathbf{A}}^2(\hat{\mathbf{r}}_i), \quad (3)$$

<sup>1</sup> Note that since the response functions are considered time independent, effects due to the motion of the structure, such as in cavity optomechanics [63], cannot be treated without further modifications.

where  $\hat{\mathbf{d}} = \sum_{\alpha} q_{\alpha} \hat{\mathbf{r}}_{\alpha}$  is the electric dipole operator of the collection of charges, while  $\hat{\mathbf{r}}_{\alpha}$  and  $\hat{\mathbf{p}}_{\alpha}$  are the position and momentum operators in the center-of-mass frame of the charge collection<sup>2</sup>. Equation (3) explicitly shows that in the presence of material bodies, the emitter-field interaction has two contributions, one from longitudinal (electrostatic) fields owing to the Coulomb interaction with charges in the macroscopic body and one from transverse fields (described by the vector potential). The relevant fields are given by

$$\hat{\mathbf{E}}^{\parallel}(\hat{\mathbf{r}}) = \sum_{\lambda} \int_0^{\infty} d\omega \int d^3\mathbf{r}'^{\parallel} \mathbf{G}_{\lambda}(\mathbf{r}, \mathbf{r}', \omega) \cdot \hat{\mathbf{f}}_{\lambda}(\mathbf{r}', \omega) + \text{H.c.}, \quad (4a)$$

$$\hat{\mathbf{A}}(\hat{\mathbf{r}}) = \sum_{\lambda} \int_0^{\infty} \frac{d\omega}{i\omega} \int d^3\mathbf{r}'^{\perp} \mathbf{G}_{\lambda}(\mathbf{r}, \mathbf{r}', \omega) \cdot \hat{\mathbf{f}}_{\lambda}(\mathbf{r}', \omega) + \text{H.c.}, \quad (4b)$$

where the longitudinal and transverse components of a tensor  $\mathbf{T}(\mathbf{r}, \mathbf{r}')$  are given by

$$\mathbb{I}^{\perp} \mathbf{T}(\mathbf{r}, \mathbf{r}') = \int d^3\mathbf{s} \delta^{\parallel/\perp}(\mathbf{r} - \mathbf{s}) \mathbf{T}(\mathbf{s}, \mathbf{r}'), \quad (5)$$

with  $\delta^{\parallel/\perp}(\mathbf{r} - \mathbf{s})$  the standard longitudinal or transverse delta function in 3D space. The functions  $\mathbf{G}_{\lambda}(\mathbf{r}, \mathbf{r}', \omega)$  are related to the dyadic Green's function  $\mathbf{G}(\mathbf{r}, \mathbf{r}', \omega)$  through

$$\mathbf{G}_e(\mathbf{r}, \mathbf{r}', \omega) = i \frac{\omega^2}{c^2} \sqrt{\frac{\hbar}{\pi \epsilon_0}} \text{Im} \epsilon(\mathbf{r}', \omega) \mathbf{G}(\mathbf{r}, \mathbf{r}', \omega), \quad (6a)$$

$$\mathbf{G}_m(\mathbf{r}, \mathbf{r}', \omega) = i \frac{\omega}{c} \sqrt{\frac{-\hbar}{\pi \epsilon_0}} \text{Im} \mu^{-1}(\mathbf{r}', \omega) [\nabla' \times \mathbf{G}(\mathbf{r}', \mathbf{r}, \omega)]^T. \quad (6b)$$

We note that in the derivation leading to the above expressions, it is assumed that  $\text{Im} \epsilon(\mathbf{r}, \omega) > 0$  and  $\text{Im} \mu(\mathbf{r}, \omega) > 0$  for all  $\mathbf{r}$ , i.e., that the materials are lossy everywhere in space. The limiting case of zero losses in some regions (e.g., in free space) is only taken at the very end of the calculation. We will see that in the reformulation in terms of emitter-centered modes, subsection 2.4,  $\mathbf{G}_{\lambda}(\mathbf{r}, \mathbf{r}', \omega)$  disappears from the formalism relatively early, and only the “normal” Green's function  $\mathbf{G}(\mathbf{r}, \mathbf{r}', \omega)$  is needed (for which the limit is straightforward).

As mentioned above, the electrostatic contribution is not present in free space, and in the literature, it is often assumed that any abstract “cavity mode” corresponds to a purely transverse EM field. This is a good approximation

for emitters that are far enough away from the material, e.g., in “large” (typically dielectric) structures such as Fabry–Perot planar microcavities, photonic crystals, micropillar resonators, etc. [64], but can break down otherwise. In general, this happens for coupling to evanescent fields [65] and in particular when subwavelength confinement is used to generate extremely small effective mode volumes, such as in plasmonic [66, 67] or phonon-polaritonic systems [68, 69]. This observation is particularly relevant as such subwavelength confinement is the only possible strategy for obtaining large enough light–matter coupling strengths to approach the single-emitter strong coupling regime at room temperature [70–73]. For subwavelength separations, it is well known that the Green's function is dominated by longitudinal components, while transverse components can be neglected [74]. In this *quasistatic* approximation, we thus have  $\hat{\mathbf{A}} \approx 0$ , cf. Eq. (4), and the light–matter interactions are all due to electrostatic (or Coulomb) interactions, even within the minimal coupling scheme<sup>3</sup>. Conversely, owing to the strongly subwavelength field confinement, the long-wavelength or dipole approximation is not necessarily appropriate, and an accurate description requires either the direct use of the expression in terms of the electrostatic potential [75] or the inclusion of higher order terms in the interaction [76]. In this context, it should be noted that within an *ab initio* description (i.e., when the emitters are not treated as few-level systems), the term  $-\hat{\mathbf{d}} \cdot \hat{\mathbf{E}}^{\parallel}(\hat{\mathbf{r}}_i)$  within the dipole approximation can be problematic as, e.g., the interaction can become arbitrarily large when the computational box is too big, which in particular affects the more extended excited states, but even leads to the lack of a ground state in infinite space [28, 33]. For the electrostatic (longitudinal) interactions considered here, this has to be resolved by going beyond the dipole approximation, such that the potential is accurately represented within the whole computational box and in particular disappears at large distances to the material [35]. It is also important to keep in mind that in any case, the approximations inherent in macroscopic QED, i.e., that the emitter wave functions do not overlap with the material part, break down for large computational boxes for the emitter wave function.

<sup>2</sup> While we here assumed a single emitter (i.e., a collection of close-by charged particles), the extension of the above formula to multiple emitters is trivial. One important aspect to note is that the electrostatic Coulomb interaction between charges (second term in Eq. (1a)) is still present, i.e., there are direct instantaneous Coulomb interactions between the charges in different emitters.

<sup>3</sup> It could then be discussed what the field modes should be called in the limit when they contain negligible contributions from propagating photon modes. However, since *all* EM modes that are not just freely propagating photons will always have a somewhat mixed light–matter character, and since these modes always solve the macroscopic Maxwell equations, they are conventionally referred to as “light,” “EM,” or “photon” modes. It is thus important to remember that this does not imply that they are simply modes of the transverse EM field.

## 2.2 Multipolar coupling

We now discuss the PZW gauge transformation [77–79], which is used to switch from the minimal coupling scheme discussed up to now to the so-called multipolar coupling scheme, which will then in turn form the basis for the emitter-centered modes we discuss later. This scheme has several advantageous properties: it expresses all light–matter interactions through the fields  $\mathbf{E}$  and  $\mathbf{B}$  directly, without needing to distinguish between longitudinal and transverse fields and allows a systematic expansion of the field-emitter interactions in terms of multipole moments. Additionally, in the multiemitter case, it also removes direct Coulomb interactions between charges in different emitters, which instead become mediated through the EM fields. This property makes it easier to explicitly verify and guarantee that causality is not violated through faster-than-light interactions. We only point out and discuss some specific relevant results here and again refer the reader to the literature for full details [22, 23]. The PZW transformation is carried out by the unitary transformation operator

$$\hat{U} = \exp\left[\frac{i}{\hbar} \int d^3\mathbf{r} \sum_i \hat{\mathbf{P}}_i(\mathbf{r}) \cdot \hat{\mathbf{A}}(\mathbf{r})\right] \quad (7)$$

where we have explicitly grouped the charges into several emitters, i.e., distinct (nonoverlapping) collections of charges, labeled with index  $i$ . The polarization operator  $\hat{\mathbf{P}}_i(\mathbf{r})$  of emitter  $i$  is usually defined as

$$\hat{\mathbf{P}}_i(\mathbf{r}) = \sum_{\alpha \in i} q_\alpha \hat{\mathbf{r}}_\alpha \int_0^1 d\sigma \delta(\mathbf{r} - \hat{\mathbf{r}}_i - \sigma \hat{\mathbf{r}}_\alpha), \quad (8)$$

where  $\hat{\mathbf{r}}_i$  is the center-of-mass position operator of emitter  $i$  and  $\hat{\mathbf{r}}_\alpha = \hat{\mathbf{r}}_\alpha - \hat{\mathbf{r}}_i$  is the relative position operator of charge  $\alpha$  belonging to emitter  $i$ . We note that there is considerable freedom in choosing a definition for  $\hat{\mathbf{P}}_i(\mathbf{r})$  as only its longitudinal component is physical. There are subtle issues associated with this choice, in particular, owing to the assumption of a point charge model and the lack of a UV cutoff leading to singular expressions. These issues are not specific to macroscopic QED, but general to the PZW transformation and have been discussed in detail in the literature [29, 80].

Since Eq. (7) describes a unitary transformation, physical results are unaffected in principle, although the convergence behavior with respect to different approximations can be quite different [26, 36]. Applying the transformation gives the new operators  $\hat{O}' = \hat{U} \hat{O} \hat{U}^\dagger$ . Expressing the Hamiltonian Eq. (1) in terms of these new operators then gives the multipolar coupling form. The

effect can be summarized by noting that the operators  $\hat{\mathbf{A}}$  and  $\hat{\mathbf{r}}_\alpha$  are unchanged, while their canonically conjugate momenta  $\hat{\boldsymbol{\Pi}}$  and  $\hat{\mathbf{p}}_\alpha$  are not. In light of the discussion of longitudinal (electrostatic) versus transverse interactions, it is interesting to point out that the electrostatic potential  $\hat{\phi}(\mathbf{r})$  is also unaffected, i.e., in the quasistatic limit, the PZW transformation has no effect on the emitter-field interaction and discussions of gauge dependence for this specific case become somewhat irrelevant. However, both the bare-emitter and the bare-field Hamiltonian are changed, as  $\hat{\mathbf{f}}_\lambda(\mathbf{r}, \omega)' \neq \hat{\mathbf{f}}_\lambda(\mathbf{r}, \omega)$ , i.e., the separation into emitter and field variables is different than in the minimal coupling scheme. We here directly show the multipolar coupling Hamiltonian after additionally neglecting interactions containing the magnetic field. This leads to

$$\mathcal{H}_A = \sum_i \left[ \frac{\hat{\mathbf{p}}_\alpha^2}{\sum_{\alpha \in i} 2m_\alpha} + \frac{1}{2\epsilon_0} \int d^3\mathbf{r} \hat{\mathbf{P}}_i^2(\mathbf{r}) \right], \quad (9a)$$

$$\mathcal{H}_F = \sum_\lambda \int_0^\infty d\omega \int d^3\mathbf{r} \hbar \omega \hat{\mathbf{f}}_\lambda^\dagger(\mathbf{r}, \omega) \hat{\mathbf{f}}_\lambda(\mathbf{r}, \omega), \quad (9b)$$

$$\mathcal{H}_{AF} = -\sum_i \int d^3\mathbf{r} \hat{\mathbf{P}}_i(\mathbf{r}) \cdot \hat{\mathbf{E}}(\mathbf{r}), \quad (9c)$$

where all operators are their PZW-transformed (primed) versions, but we have not included explicit primes for simplicity. In the long-wavelength limit, Eq. (9c) becomes simply

$$\mathcal{H}_{AF} = -\sum_i \hat{\mathbf{d}}_i \cdot \hat{\mathbf{E}}(\hat{\mathbf{r}}_i), \quad (10)$$

i.e., all field-emitter interactions are expressed through the dipolar coupling term, with the electric field operator given explicitly by

$$\hat{\mathbf{E}}(\mathbf{r}) = \sum_\lambda \int_0^\infty d\omega \int d^3\mathbf{r}' \mathbf{G}_\lambda(\mathbf{r}, \mathbf{r}', \omega) \cdot \hat{\mathbf{f}}_\lambda(\mathbf{r}', \omega) + \text{H.c.} \quad (11)$$

We note that the form of the bare-emitter Hamiltonian  $\mathcal{H}_A = \sum_i H_i$  [Eq. (9a)] under multipolar coupling is changed compared to the minimal coupling picture and in particular can be rewritten as

$$H_i = \sum_{\alpha \in i} \frac{\hat{\mathbf{p}}_\alpha^2}{2m_\alpha} + \sum_{\alpha, \beta \in i} \frac{q_\alpha q_\beta}{8\pi\epsilon_0 |\mathbf{r}_\alpha - \mathbf{r}_\beta|} + \frac{1}{2\epsilon_0} \int d^3\mathbf{r} \left[ \hat{\mathbf{P}}_i^\dagger(\mathbf{r}) \right]^2, \quad (12)$$

which makes explicit the fact that the emitter Hamiltonian in the multipolar gauge is equivalent to the emitter Hamiltonian in minimal coupling plus a term containing the transverse polarization only. In order to arrive at this form, we have used that the Coulomb interaction can be rewritten as an integral over the longitudinal polarization. The transverse part of the polarization in Eq. (12) corresponds to the

so-called dipole self-energy term [28]. When a single- or few-mode approximation of the EM field is performed *before* doing the PZW transformation, this term depends on the square of the mode-emitter coupling strength, but not on any photonic operator. However, when all modes of the EM field are included, as implicitly done here and as motivated by the fact that the term is not mode selective (it does not depend on any EM field operator), it is seen directly that this term becomes completely independent of any characteristics of the surrounding material structure, i.e., it cannot be modified by changing the environment that the emitter is located in. Instead, the bare-emitter Hamiltonian in the multipolar approach is simply slightly different than under minimal coupling. This raises the question whether in a few-mode approximation, such a term should be included in simulations of strongly coupled light–matter systems, i.e., whether the few-mode approximation should be performed before the PZW transformation or after [38]. Including the term improves some mathematical properties of the dipole approximation, in particular in large computational boxes and/or for very large coupling strengths [28, 33, 81]. However, it should also be remembered here that in realistic cavities capable of reaching few-emitter strong coupling, the dominant interaction term is due to longitudinal fields, for which this term does not exist (see discussion in subsection 2.1). Furthermore, it should be mentioned that a similar term can arise if the environment-mediated electrostatic interactions are taken into account explicitly instead of through the quantized modes, and one (or some) of the EM modes is additionally treated explicitly. The action of these modes on the emitters then has to be subtracted from the electrostatic interaction to avoid double counting them [27].

### 2.3 External (classical) fields

Adapting an argument by Sánchez-Barquilla et al. [45], here we show that macroscopic QED also enables a straightforward treatment of external incoming EM fields, in particular for the experimentally most relevant case of a classical laser pulse. Assuming that the incoming laser field at the initial time  $t = 0$  has not yet interacted with the emitters (i.e., it describes a pulse localized in space in a region far away from the emitters), it can simply be described by a product of coherent states of the EM modes for the initial wave function,  $|\psi(0)\rangle = \prod_n |\alpha_n(0)\rangle = \prod_n e^{\alpha_n(0)a_n^\dagger - \alpha_n^*(0)a_n} |0\rangle$ , where the index  $n$  here runs over all indices of the EM basis  $(\lambda, \mathbf{r}, \omega)$ , and the  $\alpha_n(0)$  correspond to the classical amplitudes obtained when expressing the laser pulse in the basis defined by

these modes. In order to avoid the explicit propagation of this classical field within a quantum calculation, the classical and the quantum field can be split in the Hamiltonian using a time-dependent displacement operator [82]  $T(t) = e^{\sum_n \alpha_n^*(t)a_n - \alpha_n(t)a_n^\dagger}$ , where  $\alpha_n(t) = \alpha_n(0)e^{-i\omega_n t}$ . Applying this transformation to the wavefunction,  $|\psi'\rangle = T(t)|\psi\rangle$ , adds an (time-dependent) energy shift that does not affect the dynamics and splits the electric field term in Eq. (10) into a classical and a quantum part,  $\widehat{\mathbf{E}}(\mathbf{r})' = \widehat{\mathbf{E}}(\mathbf{r}) + \mathbf{E}_{\text{cl}}(\mathbf{r}, t)$ .

We note that the above properties imply that within this framework, the action of any incoming laser pulse on the *full* emitter-cavity system can be described purely by the action of the medium-supported classical electric field on the emitters, with no additional explicit driving of any EM modes. This is different to, e.g., standard input–output theory, where the EM field is split into modes inside the cavity and free-space modes outside, and external driving thus affects the cavity modes<sup>4</sup>.

Importantly,  $\mathbf{E}_{\text{cl}}(\mathbf{r}, t)$  is the field obtained at the position of the emitter upon propagation of the external laser pulse through the material structure, i.e., it contains any field enhancement and temporal distortion. Since the Hamiltonian expressed by the operators  $\widehat{\mathbf{f}}_\lambda(\mathbf{r}, \omega)$  by construction solves Maxwell’s equations in the presence of the material structure,  $\mathbf{E}_{\text{cl}}(\mathbf{r}, t)$  can be obtained by simply solving Maxwell’s equations using any classical EM solver without ever expressing the pulse in the basis of the modes  $\widehat{\mathbf{f}}_\lambda(\mathbf{r}, \omega)$ . It is important to remember that EM field observables are also transformed according to

$$\langle \psi | O | \psi \rangle = \langle \psi' | T(t) O T^\dagger(t) | \psi' \rangle, \quad (13)$$

such that, e.g.,  $\langle \psi | a_n | \psi \rangle = \langle \psi' | a_n + \alpha_n(t) | \psi' \rangle$ . This takes into account that the “quantum” field generated by the laser-emitter interaction interferes with the classical pulse propagating through the structure and ensures a correct description of absorption, coherent scattering, and similar effects.

### 2.4 Emitter-centered modes

Following references [39–44], we now look for a linear transformation of the bosonic modes  $\widehat{\mathbf{f}}_\lambda(\mathbf{r}, \omega)$  at each frequency in such a way that in the new basis, only a minimal

<sup>4</sup> It should be noted that in principle, both macroscopic QED and input–output theory can provide for a complete description of the system. In fact, a few-mode description suitable for treatment within input–output theory or master equation formalisms can be obtained from macroscopic QED by, essentially, “reversing” Fano diagonalization and reexpressing the EM modes through a finite number of discrete modes coupled to exterior continua [15, 47].

number of EM modes couples to the emitters. To this end, we start with the macroscopic QED Hamiltonian within the multipolar approach Eq. (9), with the emitter-field interaction treated within the long-wavelength approximation Eq. (10). For simplicity of notation, we assume that the dipole operator of each emitter only couples to a single-field polarization,  $\hat{\mathbf{d}}_i = \hat{\mu}_i \mathbf{n}_i$ . Alternatively, the sum over  $i$  could simply be extended to include up to three separate orientations per emitter. Our goal can then be achieved by defining *emitter-centered* or *bright* (from the emitter perspective) EM modes  $\hat{B}_i(\omega)$  associated with each emitter  $i$ :

$$\hat{B}_i(\omega) = \sum_{\lambda} \int d^3\mathbf{r} \boldsymbol{\beta}_{i,\lambda}(\mathbf{r}, \omega) \cdot \hat{\mathbf{f}}_{\lambda}(\mathbf{r}, \omega) \quad (14a)$$

$$\boldsymbol{\beta}_{i,\lambda}(\mathbf{r}, \omega) = \frac{\mathbf{n}_i \cdot \mathbf{G}_{\lambda}(\mathbf{r}_i, \mathbf{r}, \omega)}{G_i(\omega)}, \quad (14b)$$

where  $G_i(\omega)$  is a normalization factor. Using Eq. (2), the commutation relations of the operators  $\hat{B}_i(\omega)$  reduce to overlap integrals of their components,  $[\hat{B}_i(\omega), \hat{B}_j^{\dagger}(\omega')] = S_{ij}(\omega)\delta(\omega - \omega')$ , with

$$\begin{aligned} S_{ij}(\omega) &= \sum_{\lambda} \int d^3\mathbf{r} \boldsymbol{\beta}_{i,\lambda}^*(\mathbf{r}, \omega) \cdot \boldsymbol{\beta}_{j,\lambda}(\mathbf{r}, \omega) \\ &= \frac{\hbar\omega^2}{\pi\epsilon_0 c^2} \frac{\mathbf{n}_i \cdot \text{Im} \mathbf{G}(\mathbf{r}_i, \mathbf{r}_j, \omega) \cdot \mathbf{n}_j}{G_i(\omega)G_j(\omega)}, \end{aligned} \quad (15)$$

such that the overlap matrix  $S_{ij}(\omega)$  at each frequency is real and symmetric (since  $\mathbf{G}(\mathbf{r}, \mathbf{r}', \omega) = \mathbf{G}^T(\mathbf{r}', \mathbf{r}, \omega)$ ). In the above derivation, we have used the Green's function identity

$$\sum_{\lambda} \int d^3\mathbf{s} \mathbf{G}_{\lambda}(\mathbf{r}, \mathbf{s}, \omega) \cdot \mathbf{G}_{\lambda}^{*T}(\mathbf{r}', \mathbf{s}, \omega) = \frac{\hbar\omega^2}{\pi\epsilon_0 c^2} \text{Im} \mathbf{G}(\mathbf{r}, \mathbf{r}', \omega) \quad (16)$$

to obtain a compact result [22, 23]. The normalization factor  $G_i(\omega)$  is obtained by requiring that  $S_{ii}(\omega) = 1$ , so

$$G_i(\omega) = \sqrt{\frac{\hbar\omega^2}{\pi\epsilon_0 c^2} \mathbf{n}_i \cdot \text{Im} \mathbf{G}(\mathbf{r}_i, \mathbf{r}_i, \omega) \cdot \mathbf{n}_i}. \quad (17)$$

We note that the coupling strength  $G_i(\omega)$  of the emitter-centered mode  $\hat{B}_i(\omega)$  to emitter  $i$  is directly related to the EM spectral density  $J_i(\omega) = [\mu G_i(\omega)/\hbar]^2$  for transition dipole moment  $\mu$  [83]. In the regime of weak coupling, i.e., when the EM environment can be approximated as a Markovian bath, the spontaneous emission rate at an emitter frequency  $\omega_e$  is then given by  $2\pi J_i(\omega_e)$ .

Since the overlap matrix  $S_{ij}(\omega)$  of the modes associated with emitters  $i$  and  $j$  is determined by the imaginary part of the Green's function between the two emitter positions, it follows that the modes  $\hat{B}_i(\omega)$ , or equivalently, the

coefficient functions  $\boldsymbol{\beta}_i(\mathbf{r}, \omega)$ , are not orthogonal in general. This can be resolved by performing an explicit orthogonalization, which is possible as long as the modes are linearly independent. When this is not the case, linearly dependent modes can be dropped from the basis until a minimal set is reached [42]. In the following, we thus assume linear independence. We can then define new orthonormal modes  $\hat{C}_i(\omega)$  as a linear superposition of the original modes (and vice versa)

$$\hat{C}_i(\omega) = \sum_{j=1}^N V_{ij}(\omega) \hat{B}_j(\omega), \quad (18a)$$

$$\boldsymbol{\chi}_{i,\lambda}(\mathbf{r}, \omega) = \sum_{j=1}^N V_{ij}(\omega) \boldsymbol{\beta}_{j,\lambda}(\mathbf{r}, \omega), \quad (18b)$$

which also implies that  $\hat{B}_i(\omega) = \sum_{j=1}^N W_{ij}(\omega) \hat{C}_j(\omega)$ , where  $\mathbf{W}(\omega) = \mathbf{V}(\omega)^{-1}$ . In the above, the transformation matrix  $\mathbf{V}(\omega)$  is chosen such that  $[\hat{C}_i(\omega), \hat{C}_j^{\dagger}(\omega')] = \delta_{ij}\delta(\omega - \omega')$ , which implies  $\mathbf{V}(\omega)\mathbf{S}(\omega)\mathbf{V}^{\dagger}(\omega) = \mathbb{1}$ . The coefficient matrices  $\mathbf{V}(\omega)$  can be chosen in various ways, corresponding to different unitary transformations of the same orthonormal basis. We mention two common approaches here. One consists in performing a Cholesky decomposition of the overlap matrix,  $\mathbf{S}(\omega) = \mathbf{L}(\omega)\mathbf{L}^T(\omega)$ , with  $\mathbf{V}(\omega) = \mathbf{L}(\omega)^{-1}$ , where  $\mathbf{L}(\omega)$  and  $\mathbf{L}(\omega)^{-1}$  are lower triangular matrices. This is the result obtained from Gram–Schmidt orthogonalization, with the advantage that  $\hat{C}_i(\omega)$  only involves  $\hat{B}_j(\omega)$  with  $j \leq i$  and vice versa, such that emitter  $i$  only couples to the first  $i$  photon continua. Another possibility is given by Löwdin orthogonalization, with  $\mathbf{V}(\omega) = \mathbf{S}(\omega)^{-1/2}$ , where the matrix power is defined as  $\mathbf{S}^{\alpha} = \mathbf{U}\boldsymbol{\Lambda}^{\alpha}\mathbf{U}^{\dagger}$ , with  $\mathbf{U}(\boldsymbol{\Lambda})$  a unitary (diagonal) matrix containing the eigenvectors (eigenvalues) of  $\mathbf{S}$ . This approach maximizes the overlap  $[\hat{C}_i(\omega), \hat{B}_i^{\dagger}(\omega)]$  while ensuring orthogonality and can thus be seen as the “minimal” correction required to obtain an orthonormal basis.

Using the orthonormal set of operators  $\hat{C}_i(\omega)$ , which are themselves linear superpositions of the operators  $\hat{\mathbf{f}}_{\lambda}(\mathbf{r}, \omega)$ , we can perform a unitary transformation (separately for each frequency  $\omega$ ) of the  $\hat{\mathbf{f}}_{\lambda}(\mathbf{r}, \omega)$  into a basis spanned by the emitter-centered (or bright) EM modes and an infinite number of “dark” modes  $\hat{D}_i(\omega)$  that span the orthogonal subspace and do not couple to the emitters, such that

$$\hat{\mathbf{f}}_{\lambda}(\mathbf{r}, \omega) = \sum_{i=1}^N \boldsymbol{\chi}_{i,\lambda}^*(\mathbf{r}, \omega) \hat{C}_i(\omega) + \sum_j \mathbf{d}_{j,\lambda}^*(\mathbf{r}, \omega) \hat{D}_j(\omega). \quad (19)$$

where  $\int d^3\mathbf{r} \mathbf{d}_{j,\lambda}^*(\mathbf{r}, \omega) \cdot \boldsymbol{\chi}_i(\mathbf{r}, \omega) = 0$ . The Hamiltonian can then be written as

$$\mathcal{H} = \int_0^\infty d\omega \left[ \sum_{i=1}^N \hbar\omega \hat{C}_i^\dagger(\omega) \hat{C}_i(\omega) + \sum_j \hbar\omega \hat{D}_j^\dagger(\omega) \hat{D}_j(\omega) - \sum_{i,j=1}^N \hat{\mu}_i(g_{ij}(\omega) \hat{C}_j(\omega) + \text{H.c.}) \right] + \sum_{i=1}^N \hat{H}_i \quad (20)$$

where  $g_{ij}(\omega) = G_i(\omega)W_{ij}(\omega)$ . We note here that  $\mathbf{V}(\omega)$  and thus  $\mathbf{W}(\omega)$  and  $g_{ij}(\omega)$  can always be chosen real owing to the reality of  $S_{ij}(\omega)$ , but we here treat the general case with possibly complex coefficients. Since the dark modes are decoupled from the rest of the system, they do not affect the dynamics and can be dropped, giving

$$\mathcal{H} = \sum_{i=1}^N \hat{H}_i + \int_0^\infty d\omega \left[ \sum_{i=1}^N \hbar\omega \hat{C}_i^\dagger(\omega) \hat{C}_i(\omega) - \sum_{i,j=1}^N \hat{\mu}_i(g_{ij}(\omega) \hat{C}_j(\omega) + \text{H.c.}) \right]. \quad (21)$$

We mention for completeness that if the dark modes are initially excited, including them might be necessary to fully describe the state of the system. We have now explicitly constructed a Hamiltonian with only  $N$  independent EM modes  $\hat{C}_i(\omega)$  for each frequency  $\omega$  [39–44]. This Hamiltonian corresponds to a set of  $N$  emitters coupled to  $N$  continua and can be treated using, e.g., a wide variety of methods developed in the context of open quantum systems [84–87].

Furthermore, one can obtain an explicit expression for the electric field operator based on the modes  $\hat{C}_i(\omega)$ , which to the best of our knowledge has not been presented previously in the literature. This is obtained by inserting Eq. (19) in Eq. (11), again dropping the dark modes  $\hat{D}_i(\omega)$  and again using the integral relation for Green's functions from Eq. (16), leading to

$$\hat{\mathbf{E}}^{(+)}(\mathbf{r}) = \sum_{i=1}^N \int_0^\infty d\omega \mathbf{E}_i(\mathbf{r}, \omega) \hat{C}_i(\omega) \quad (22a)$$

$$\mathbf{E}_i(\mathbf{r}, \omega) = \sum_{j=1}^N V_{ij}^*(\omega) \boldsymbol{\varepsilon}_j(\mathbf{r}, \omega) \quad (22b)$$

$$\boldsymbol{\varepsilon}_j(\mathbf{r}, \omega) = \frac{\hbar\omega^2}{\pi\epsilon_0 c^2} \frac{\text{Im} \mathbf{G}(\mathbf{r}, \mathbf{r}_j, \omega) \cdot \mathbf{n}_j}{G_j(\omega)} \quad (22c)$$

These relations show that we can form explicit photon modes in space at each frequency by using orthonormal superpositions of the emitter-centered EM modes  $\boldsymbol{\varepsilon}_j(\mathbf{r}, \omega)$ . We note that this also provides a formal construction for the emitter-centered EM modes, i.e., the modes created by the operators  $\hat{B}_i(\omega)$ , with a field profile corresponding to the imaginary part of the Green's function associated with that emitter. These modes are well behaved: they solve the source-free Maxwell equations, are real everywhere in

space, and do not diverge anywhere (here, it should be remembered that the real part of the Green's function diverges for  $\mathbf{r} = \mathbf{r}'$ , while the imaginary part does not). As a simple example, we can take a single  $z$ -oriented emitter at the origin in free space. The procedure used here then gives exactly the  $l=1, m=0$  spherical Bessel waves, i.e., the only modes that couple to the emitter when quantizing the field using spherical coordinates. Furthermore, it should be noted that these modes contain the absorption of the EM field in the material, which is encoded in the Green's function that solves the macroscopic Maxwell equations including losses. This can be understood by remembering that macroscopic QED can be seen as solving the dynamics of an infinite set of coupled harmonic oscillators corresponding to the EM modes and material excitations (including reservoir modes that describe dissipation), cf. subsection 2. Since the coupled system consists only of harmonic oscillators, this problem can be diagonalized without approximations (i.e., without using a master equation or similar formalisms), resulting in a description of the full system in terms of continua of oscillators. The electric field operator as described by Eq. (22) can then be seen as a projection of the full solution onto the field subspace [23]. The end effect of this is that although the Hamiltonian, Eq. (21), is Hermitian, dissipation in the material is fully included. Finally, we note that it can be verified easily that inserting Eq. (22) in Eq. (9) and simplifying leads exactly to Eq. (21).

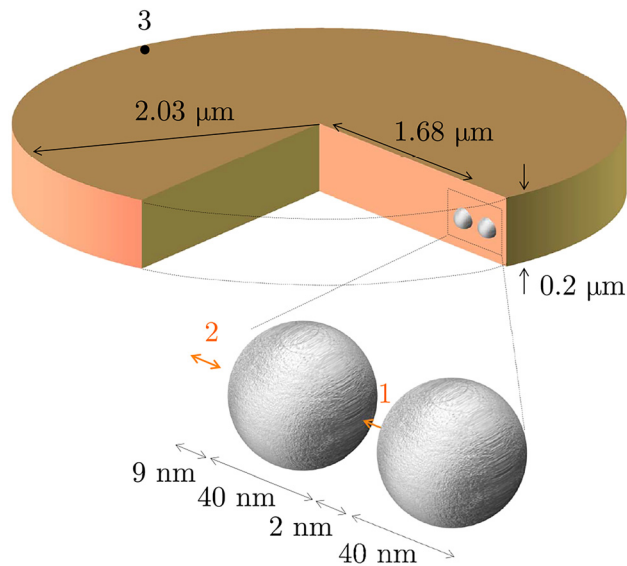
### 3 Example

As an example, we now treat a complex metallodielectric structure, as shown in Figure 1. It is composed of a dielectric microdisk resonator supporting whispering gallery modes, with a metallic sphere dimer antenna placed within. The SiN ( $\epsilon = 4$ ) disk is similar to that considered in Ref. [50], with a radius of  $2.03 \mu\text{m}$  and a height of  $0.2 \mu\text{m}$ . Two  $40\text{-nm}$ -diameter Ag (with permittivity taken from Ref. [88]) nanospheres separated by a  $2\text{-nm}$  gap are placed  $1.68 \mu\text{m}$  away from the disk axis. Two point-dipole emitters modeled as two-level systems and oriented along the dimer axis are placed in the central gap of the dimer antenna and just next to the antenna (labeled as points 1 and 2 in Figure 1). Their transition frequencies are chosen as  $\omega_{e,1} = \omega_{e,2} = 2 \text{ eV}$ , while the dipole transition moments are  $\mu_1 = 0.1 \text{ e nm}$  and  $\mu_2 = 3 \text{ e nm}$  (roughly corresponding to typical single organic molecules and  $J$ -aggregates, respectively [89]). As shown in the theory section, the emitter dynamics are then fully determined by the Green's

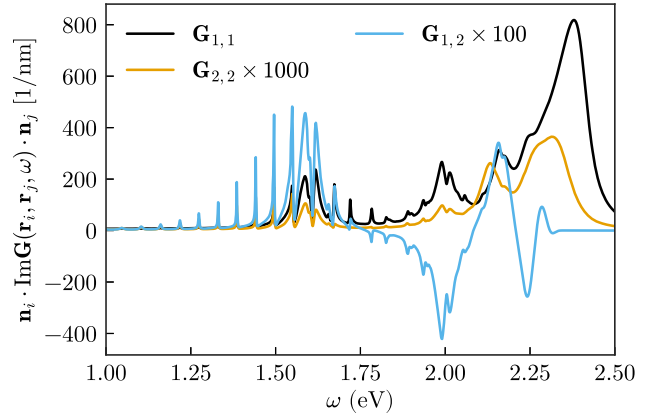


function between the emitter positions (as also found in multiple scattering approaches [54]). In Figure 2, we show the relevant values  $\mathbf{n}_i \cdot \text{Im} \mathbf{G}(\mathbf{r}_i, \mathbf{r}_j, \omega) \cdot \mathbf{n}_j$ , which clearly reveals the relatively sharp Mie resonances of the dielectric disk, hybridized with the short-range plasmonic modes of the metallic nanosphere dimer. In addition, it also shows that the coupling between the two emitters, i.e., the off-diagonal term with  $i = 1, j = 2$ , has a significant structure and changes sign several times within the frequency interval.

We now study the dynamics for the Wigner–Weisskopf problem of spontaneous emission of emitter 1, i.e., for the case where emitter 1 is initially in the excited state, while emitter 2 is in the ground state and the EM field is in its vacuum state, such that  $|\psi(t = 0)\rangle = \sigma_1^+|0\rangle$ , where  $|0\rangle$  is the global vacuum without any excitations and  $\sigma_1^+$  is a Pauli matrix acting on emitter 1. We also treat the light–matter coupling within the rotating wave approximation (RWA), i.e., we use  $\sum_{i,j=1}^N \mu_i(g_{ij}(\omega)\sigma_i^+\hat{C}_j(\omega) + \text{H.c.})$  as the light–matter interaction term. The number of excitations is then conserved, and the system can be solved easily within the single-excitation subspace by discretizing the photon continua in frequency [51]. The resulting emitter dynamics, i.e., the population of the excited states of the emitters, are shown in Figure 3. This reveals that the EM field created by emitter 1 due to spontaneous emission is partially reabsorbed by emitter 2. Comparison with the dynamics of emitter 1 when emitter 2 is not present (shown as a dashed



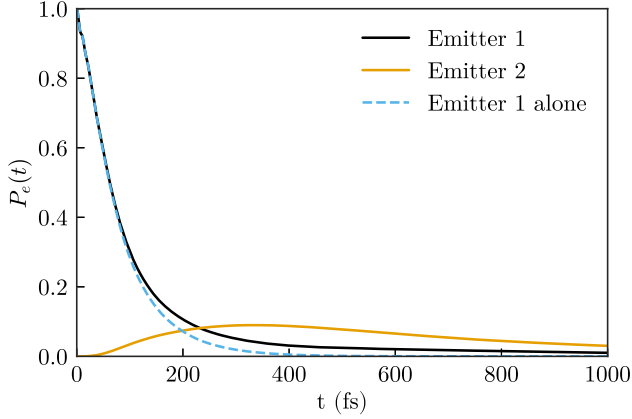
**Figure 1:** Sketch of the hybrid metallodielectric structure we treat: a dielectric microdisk resonator with a metallic dimer antenna placed on top. The positions of the two emitters are indicated by points 1 and 2, while the field evaluation point used later is indicated as point 3.



**Figure 2:** Green's function factor  $\mathbf{n}_i \cdot \text{Im} \mathbf{G}(\mathbf{r}_i, \mathbf{r}_j, \omega) \cdot \mathbf{n}_j$  connecting the two emitters in the geometry of Figure 1.

blue line in Figure 3) furthermore reveals that there is also significant transfer of the population back from emitter 2 to emitter 1. We note that while the system treated here is in the weak coupling regime and no Rabi oscillations are present, the method works equally well under strong light–matter coupling [45–47]. Within the RWA employed here, the emitter populations for long times go to zero. Without the RWA, the ground state of the system would be a dressed state with an energy shift corresponding exactly to the Casimir–Polder potential of the emitter in the structure (usually calculated within lowest order perturbation theory) [23]. In our example, the final state at long times would then be the new dressed ground state plus a wave packet of photons emitted into free space. Furthermore, since the number of excitations (in the uncoupled basis) is not conserved without the RWA, the initial state  $\sigma_1^+|0\rangle$  would actually have nonzero probability of emitting multiple photons. When the dressing is sufficiently large, the so-called ultrastrong coupling regime is entered [90]. We note for completeness that the theory presented here can treat this regime without problems. In fact, since ultrastrong coupling effects do not depend on (approximate) resonance between modes and levels, taking into account the full spectrum of EM modes is particularly important, and macroscopic QED is thus particularly well suited to treat such effects.

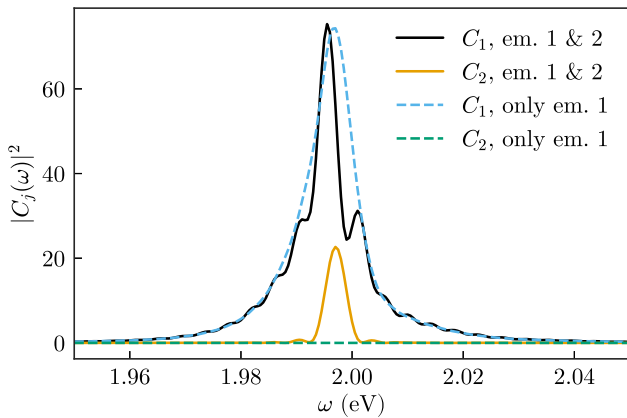
The direct access to the photonic modes in this approach provides interesting insight into, e.g., the photonic mode populations, which are shown in Figure 4 at the final time considered here,  $t = 1000$  fs. As mentioned above, there is some freedom in choosing the orthonormalized continuum modes  $C_j(\omega)$  as any linear superposition of modes at the same frequency is also an eigenmode of the EM field. We have here chosen the modes obtained through Gram–Schmidt orthogonalization,



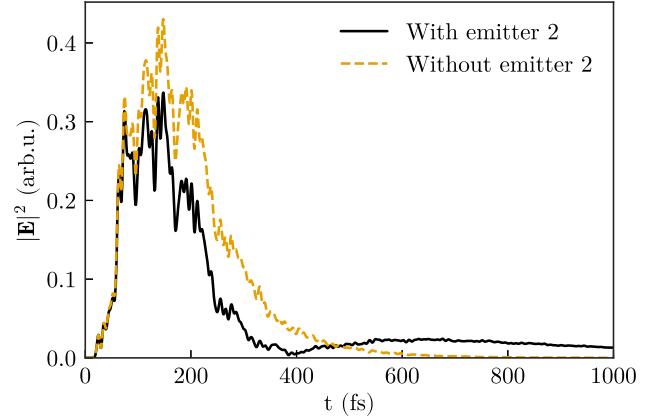
**Figure 3:** Population of emitters 1 (black line) and 2 (orange line) for the Wigner–Weisskopf problem, with emitter 1 initially excited. The blue dashed line shows the dynamics of emitter 1 if emitter 2 is not present.

which, as discussed above, have the advantage that emitter  $i$  only couples to the first  $i$  photon continua. In particular, emitter 1 only couples to a single continuum,  $\hat{C}_1(\omega)$ , while emitter 2 couples to the same continuum, and additionally to its “own” continuum  $\hat{C}_2(\omega)$ . This makes the comparison between the case of having both emitters present or only including emitter 1 quite direct, as can be observed in Figure 4. In particular, any population in the modes  $\hat{C}_2(\omega)$  must come from emitter 2, after it has in turn been excited by the photons emitted by emitter 1 into continuum 1.

Finally, we also evaluate the electric field in time at a third position (indicated as point 3 in Figure 1), as determined by Eq. (22). This is displayed in Figure 5 and shows a



**Figure 4:** Population  $\langle C_j^\dagger(\omega)C_j(\omega) \rangle$  of electromagnetic (EM) modes at time  $t = 1000$  fs in the presence of both emitters (solid black and orange lines) and in the presence of only emitter 1 (dashed blue and green lines). Gram–Schmidt orthogonalization has been used here, so that emitter 1 only couples to continuum 1 (i.e., populations for  $j = 2$  are identically zero when emitter 2 is not present), while emitter 2 couples to both continua.



**Figure 5:** Time-dependent electric field intensity  $|E|^2$  at position 3 in Figure 1, for the case when both emitters are present (black solid line) and when only emitter 1 is present (orange dashed line).

broad initial peak due to the fast initial decay of emitter 1 (filtered by propagation through the EM structure, with clear interference effects visible) and then a longer tail due to the longer-lived emission from both emitters, which is mostly due to emitter 2 (which is less strongly coupled to the EM field) and its backfeeding of the population to emitter 1.

## 4 Conclusions

In this article, we have presented a general overview of the application of the formalism of macroscopic QED in the context of quantum nanophotonics. Within this research field, it is often mandatory to describe from an *ab initio* perspective how a collection of quantum emitters interacts with a nanophotonic structure, which is usually accounted for by utilizing macroscopic Maxwell equations. Macroscopic QED then needs to combine tools taken from both quantum optics and classical electromagnetism. After the presentation of the general formalism and its approximations, we have reviewed in detail the steps to construct a minimal but complete basis set to analyze the interaction between an arbitrary dielectric structure and multiple quantum emitters. This minimal basis set is formed by the so-called emitter-centered modes, such that all the information regarding the EM environment is encoded into the EM dyadic Green’s function, which can be calculated using standard numerical tools capable of solving macroscopic Maxwell equations in complex nanophotonic structures. As a way of example and to show its full potential, in the final part of this article, we have applied this formalism to solve both the population dynamics and EM field generation associated with the coupling of two quantum emitters

with a hybrid plasmodielectric structure composed of a dielectric microdisk within which a metallic nanosphere dimer is immersed. We emphasize that this formalism can be used not only to provide exact solutions to problems in quantum nanophotonics but also to serve as a starting point for deriving simpler models and/or approximated numerical treatments.

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