Let us in the following focus on the important case of quantum systems. Therefore also lead to a change in how we understand experimental results. A change in description can for theory, but it is also the basis of how we interpret modeling of quantum systems is not only important [1]. Evaluating and improving our theoretical description then with stringent tests of our theoretical description [1]. Comparing spectroscopic measurements with theoretical predictions provides us many possible combinations of perturbations and monitored aspects. Comparing the spectroscopic measurements with theoretical predictions provides us then with stringent tests of our theoretical description [1]. Evaluating and improving our theoretical modeling of quantum systems is not only important for theory, but it is also the basis of how we interpret experimental results. A change in description can therefore also lead to a change in how we understand quantum systems. Let us in the following focus on the important case of the electromagnetic response of a quantum system to an electromagnetic perturbation, e.g., measuring the absorption or emission spectrum of a molecule, an atom or solid. One of the most fundamental assumption in our current theoretical modeling of real systems is then that the (transversal) light field is treated as decoupled from the quantum system [1]. This assumption leads to two important simplifications. First, the quantum system under investigation consists only of charged particles (electrons and nuclei) and it is only the longitudinal Coulomb interaction among the particles and their renormalized masses (bare and electromagnetic [2]) that remain in the description of the photon field. One of the consequences then is that our approximate theoretical modeling cannot account for finite lifetimes of excited atomic states [2]. Second, the electromagnetic perturbation is external, i.e., the probe is not a dynamical variable of our modeling. We therefore do not have direct access to changes in the electromagnetic field and possible retardation, polarization and local-field effects. For instance, the laser pulse that perturbs a molecule is modeled as a fixed classical field that is not changed even though it interacts with the molecule and a charge current is induced that acts back on the radiation field. Although these simplifications are justified in most cases when modeling spectroscopic experiments in, e.g., quantum chemistry and material science, in the recent years tremendous experimental advances in the field of strongly-coupled light-matter systems [3,4] have uncovered many situations where the fundamental assumption

I. INTRODUCTION

Spectroscopic techniques are central to modern quantum physics [1]. Measuring the reaction of a microscopic system, such as an atom, a molecule or a solid-state system to some perturbation allows us to deduce important information about that system. We can use many different perturbations, ranging from electromagnetic radiation, particles to even mechanical deformation and heat. At the same time we then monitor similar aspects of the physical system, e.g., the radiation emitted from an atom after a perturbation with a laser pulse. This allows us many possible combinations of perturbations and monitored aspects. Comparing the spectroscopic measurements with theoretical predictions provides us then with stringent tests of our theoretical description [1]. Evaluating and improving our theoretical modeling of quantum systems is not only important for theory, but it is also the basis of how we interpret experimental results. A change in description can therefore also lead to a change in how we understand quantum systems.

We introduce linear-response theory for non-relativistic quantum-electrodynamics in the long wavelength limit, which allows us to treat correlated excited-state phenomena of matter-photon systems from first principles. By using quantum-electrodynamical density-functional theory we can reformulate the resulting fully coupled photon-matter response equations as a pseudo-eigenvalue problem. This provides a direct extension of the conventional matter-only response theory. Our approach can be solved numerically very efficiently and existing ab-initio density-functional response implementations can be easily extended to take the full photon-matter response into account. We highlight how the coupling between light and matter changes the usual response functions and introduces new types of response functions that measure the matter-photon subsystem responses. We show how correlating light and matter changes the Maxwell equations and highlight how the spectra of real systems are changed upon strongly coupling to the photon field. A key feature of treating the combined matter-photon response is that natural lifetimes of excitations become directly accessible from first principles and no artificial broadening of spectra is required anymore. By introducing a straightforward extension of the random-phase approximation for the coupled matter-photon problem, we are able to present the first ab-initio spectra for a real molecular system that is coupled to the quantized electromagnetic field.

Light-Matter Response Functions in Quantum-Electrodynamical Density-Functional Theory: Modifications of Spectra and of the Maxwell Equations

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of decoupling light and matter is no longer valid. This is the case, for instance, in polariton chemistry \cite{5}, strong light-matter coupling in solid-state physics \cite{6}, biological systems \cite{7} or nanoplasmonics \cite{8, 9} and Floquet engineering \cite{10}. As a result of mixing matter and photon degrees of freedom novel effects emerge such as changes in chemical pathways \cite{11, 13} ground-state electroluminescence \cite{14}, cavity-controlled chemistry for molecular ensembles \cite{15, 16}, or optomechanical coupling in optical cavities \cite{17} and new topological phases of matter \cite{18}. Due to the inherent complexity of such coupled fermion-boson problems, the theoretical treatment is usually restricted to simplified effective models that heavily rely on input parameters. However, a recent generalization of density-functional theory (DFT) and its time-dependent generalization TDDFT to coupled matter-photon systems \cite{19–22} overcomes this restriction and allows for an ab-initio description of real systems strongly coupled to photons. This so-called quantum-electrodynamic density-functional theory (QEDFT) reformulates quantum electrodynamics (QED) in terms of the exact quantum-fluid description for coupled fermion-boson systems \cite{23}. In this way efficient and accurate approximations to treat many-particle systems coupled to the photon field can be devised \cite{24, 25}, which allow us to go beyond the assumption of decoupling light and matter when describing real systems. This also challenges our conception of light and matter as distinct entities \cite{26} and first results for real systems \cite{27} highlight that what we perceive as the ground state of a molecular system is always defined only with respect to the surrounding photon vacuum \cite{28}. While QEDFT has now been successfully applied to real systems in equilibrium, the changes in the ground-state observables are known to be relatively small, i.e., it is Lamb-shift physics. Instead of ground-state properties, what is used experimentally to quantify the strong-coupling regime is the so-called Rabi splitting where the emergence of hybrid light-matter states lead to a split of spectral lines \cite{28}. Furthermore, strong coupling can lead to changes in lifetimes of excited states and induce super- and sub-radiant behavior in complex quantum many-body systems. The changes are therefore most profound in spectroscopic quantities, where polaritonic states lead to large changes in, e.g., the absorption spectra \cite{1}. Further, the response of the hybrid quantum system depends strongly on whether we use classical or quantum light \cite{1}. This allows for novel types of spectroscopic techniques that make specific use of the quantum nature of light \cite{1, 29}. In this work, we present how these changes of well-known spectroscopic quantities like the absorption spectrum can be efficiently and accurately determined within the framework of QEDFT. We further show how novel spectroscopic quantities emerge that are based on the quantum nature of light and matter alike. This leads, besides others, to a change of the Maxwell equations in matter. We demonstrate these different aspects first for a simple model system. With this model system we highlight the range of validity of a random-phase-type approximation for the matter-photon coupling, which we then apply to present the first ab-initio-type calculation of spectral properties for a real three-dimensional hybrid light-matter system, a benzene molecule placed in a high-Q optical cavity. Besides the well-known Rabi-splitting, we demonstrate how lifetimes of excited states become accessible from first-principles and how the mixed matter-photon character of resonances can be determined. These results highlight that an ab-initio description of matter and light allows to properly access so far elusive properties of real systems and provides a new perspective on well-established concepts in quantum chemistry and material science.

This paper is structured as follows: In Sec. \textbf{II} we first provide a brief introduction into QED in the non-relativistic and long wave-length limit, before deriving the four possible combinations of linear-response functions of hybrid light-matter systems. We then discuss the QEDFT reformulation of coupled matter-photon systems, introduce the according Kohn-Sham (KS) construction and linear-response formalism. Next we discuss the influence of the hybrid light-matter states on observables, e.g., how the Maxwell’s equation change due to the coupling between photon and quantum matter. We further present a random-phase approximation for the coupling to the photons that allows us to perform linear-response calculations for coupled photon-matter systems. In Sec. \textbf{III} we exemplify those concepts for the generalized Rabi model and assess the quality of the random-phase approximation and compare it to the exact and the rotating-wave approximation. Next, in Sec. \textbf{IV} we then apply the random-phase approximation to calculate the absorption spectra of a benzene molecule in an optical cavity and discuss the emerging polaritonic branches and lifetimes. Finally, in Sec. \textbf{V} we present a summary of the results and provide an outlook of novel physical situations that are now possible to treat within QEDFT.

\section{Light-Matter Interaction in the Long Wave-Length Limit}

Our fundamental description of how the charged constituents of atoms, molecules and solid-state systems, i.e., electrons and positively charged nuclei, interact is based on QED \cite{1, 2, 30, 31}. In this theory the interaction is mediated via the exchange of photons. Adopting the Coulomb gauge for the photon field allows us to single out the longitudinal interaction among the particles which gives rise to the well-known Coulomb interaction and leaves the photon field purely transversal. Assuming then that the kinetic energies of the nuclei and electrons are relatively small, allows us to take the non-relativistic limit for the matter subsystem of the
coupled photon-matter Hamiltonian, which gives rise to the so-called Pauli-Fierz Hamiltonian [1 2 21] of non-relativistic QED. In a next step one then usually assumes that the combined matter-photon system is in its ground state such that the transversal charge currents are small and that the coupling to the (transversal) photon field is very weak. Besides the Coulomb interaction it is then only the physical mass of the charged constituents (bare plus electromagnetic mass [2]) that is a reminder of the photon field in the usual many-body Schrödinger Hamiltonian. In this work, however, we will not disregard the transversal photon field. This becomes necessary, for instance, when we consider the system in a high-Q optical cavity or near some nanoplasmonic structure [3 4]. In such cases the properties of the many-body system are changed, e.g., the excitation energies and lifetimes are modified. This happens because certain modes of the photon vacuum field are enhanced which can lead to a strong coupling of light with matter. We will keep this modes explicitly in our description of the combined light-matter system. Since the wavelength of these modes are usually much larger than the extend of the matter subsystem, we can then adopt the long-wavelength or dipole approximation in the length-gauge [32] which leads (in SI units) to [20 21 21]

\[
\begin{align*}
\hat{H}(t) = & \sum_{i=1}^{N} \left( -\frac{k^2}{2m_e} \nabla_i^2 + v(r_i,t) \right) + \frac{e^2}{4\pi\epsilon_0} \sum_{i>j=1}^{N} \frac{1}{r_{ij}} \\
& + \sum_{\alpha=1}^{M} \frac{1}{2} \left[ \hat{p}_{\alpha}^2 + \omega_\alpha^2 \left( \hat{q}_\alpha - \frac{\lambda_\alpha}{\omega_\alpha} R \right) \right] + j_\alpha(t) \frac{\hat{q}_\alpha}{\omega_\alpha} \delta v(r) 
\end{align*}
\]

(1)

Here we consider the nuclei merely externally, i.e., we assume them clamped, such that they give rise to attractive nuclear potentials (but the inclusion of the nuclei as quantum particles is straightforward) and we only keep the N electrons with mass \(m_e\) of the quantum system explicitly. We restrict ourselves to arbitrarily many but a finite number \(M\) of modes \(\alpha \equiv (k, s)\) with \(s\) being the two transversal polarization directions that are perpendicular to the direction of propagation \(k\). The frequency \(\omega_\alpha\) and polarization \(\epsilon_\alpha\) that enters in \(\lambda_\alpha = \epsilon_\alpha \lambda_\alpha\) with \(\lambda_\alpha = S_k(r)/\sqrt{c_0}\) and mode function \(S_k(r)\) define these electromagnetic modes. \(S_k(r)\) is normalized and has the unit \(1/\sqrt{V}\) with the volume \(V\). These modes couple via the displacement coordinate \(\hat{q}_\alpha = \sqrt{2\omega_\alpha}(\hat{a}_\alpha + \hat{a}_\alpha^\dagger)\) strongly to the total dipole moment \(R = \sum_{i=1}^{N} e_i r_i\). The \(q_\alpha\) corresponds to the mode \(\alpha\) of the displacement field \(D_\alpha = \epsilon_\alpha \omega_\alpha \lambda_\alpha \hat{q}_\alpha\) [32]. Further, the conjugate momentum of the displacement coordinate is given by \(p_\alpha = -i\sqrt{\frac{\hbar}{2\omega_\alpha}}(\hat{a}_\alpha - \hat{a}_\alpha^\dagger)\). Besides a time-dependent external potential \(v(r,t) = v_0(r) + \delta v(r,t)\), where \(v_0(r)\) describes the attractive potentials of the nuclei and \(\delta v(r,t)\) a classical external probe field that couples to the electronic subsystem, we also have an external perturbation \(j_\alpha(t) = j_{\alpha,0} + \delta j_\alpha(t)\) that acts directly on the mode \(\alpha\) of the photon subsystem. Here \(j_{\alpha,0}(t)\) is connected to a classical external charge current that acts as a source for the inhomogeneous Maxwell equation

\[
(\frac{1}{c^2} \partial_t^2 - \nabla^2)A(r,t) = \mu_0 \epsilon_0 j(r,t).
\]

(2)

Formally, however, due to the length-gauge transformations, the \(j_{\alpha}(t)\) corresponds to the time-derivative of this (mode-resolved) classical external charge current [20 21]. Physically the static part \(j_{\alpha,0}\) merely polarizes the vacuum of the photon field and leads to a static electric field [22 25]. The time-dependent part \(\delta j_\alpha(t)\) then generates real photons in the mode \(\alpha\).

### A. Linear Response in the Length Gauge

With the above Hamiltonian in length gauge we can then in principle solve for a given initial state of the coupled matter-photon system \(\Psi_0(r_1\sigma_1,...,r_N\sigma_N,q_1,...,q_M)\) the corresponding time-dependent Schrödinger equation (TDSE)

\[
\frac{i\hbar}{\partial t} \Psi(r_1\sigma_1,...,t) = \hat{H}(t)\Psi(r_1\sigma_1,...,t),
\]

(3)

where the \(\sigma\) correspond to the spin degrees-of-freedom. However, instead of trying to solve for the infeasible time-dependent many-body wave function, we restrict ourselves to weak perturbations \(\delta \hat{v}(r,t)\) and \(\delta j_{\alpha}(t)\) and assume that our system is in the ground state of the coupled matter-photon system at \(t = 0\). In this case first-order time-dependent perturbation theory can be used (see App. [A] for details) to approximate the dynamics of the coupled matter-photon system. This framework gives us access to linear spectroscopy, e.g., the absorption spectrum of a molecule. Traditionally, if we made a decoupling of light and matter, i.e., we would assume \(\Psi_0(r_1\sigma_1,...,r_N\sigma_N,q_1,...,q_M) \approx \psi_0(r_1\sigma_1,...,r_N\sigma_N) \otimes \phi_0(q_1,...,q_M)\), we would only consider the matter subsystem \(\psi\) (the photonic part \(\varphi\) would be completely disregarded) and would investigate the classical dipole field that the electrons induced due to a classical external perturbation \(\delta \hat{v}(r,t)\). To determine this we would only consider the linear response of the density operator \(\hat{n}(r) = \sum_{\alpha=1}^N \delta (r - r_{\alpha})\) which would be given by the usual density-density response function in terms of the electronic wave function only. Here, since we do not assume this decoupling, the density-density response is taken with respect to the combined ground-state wave function \(\Psi_0\) and is consequently different to the traditional density-density response. Further, since we can also perturb the photon field in the cavity by \(\delta j_{\alpha}(t)\) which will subsequently induce density fluctuations, the density response gets a further contribution
leading to
\[
\delta n(\mathbf{r}, t) = \int dt' \int d\mathbf{r}' \chi_n^a(\mathbf{r}, \mathbf{r}', t) \delta v(\mathbf{r}', t') + \sum_{\alpha=1}^M \int dt' \chi_n^{a}\alpha(\mathbf{r}, t') \delta j_\alpha(t').
\] (4)

Here \(\chi_n^a(\mathbf{r}, \mathbf{r}', t)\) corresponds to the density-density response but with respect to the coupled light-matter ground state and \(\chi_n^{a}\alpha(\mathbf{r}, t)\) corresponds to the response induced by changing the photon field (see App. A for the exact definitions). The response function \(\chi_n^a(\mathbf{r}, \mathbf{r}', t)\) act definitions). The response function \(\chi_n^a(\mathbf{r}, \mathbf{r}', t)\) is not equal to \(\chi_n^a(\mathbf{r}, \mathbf{r}', t)\) except for the position independent case of the Rabi model. [33]. The entire linear-response in non-relativistic QED for the density and photon coordinate can be written in matrix form as

\[
\begin{pmatrix}
\delta n \\
\delta q_1 \\
\delta q_2 \\
\vdots \\
\delta q_M \\
\end{pmatrix} =
\begin{pmatrix}
\chi_n^1 & \chi_n^1 & \chi_n^2 & \cdots & \chi_n^M \\
\chi_n^1 & \chi_n^2 & \chi_n^2 & \cdots & \chi_n^M \\
\vdots & \vdots & \vdots & \cdots & \vdots \\
\chi_n^1 & \chi_n^2 & \chi_n^2 & \cdots & \chi_n^M \\
\end{pmatrix}
\begin{pmatrix}
\delta v \\
\delta j_1 \\
\delta j_2 \\
\vdots \\
\delta j_M \\
\end{pmatrix}
\] (6)

where we imply integration over time and space when appropriate. In this form we clearly see that the density response of the coupled matter-photon system depends on whether we use a classical field \(\delta v(\mathbf{r}, t)\), photons, which are created by \(\delta j_\alpha(t)\), or combinations thereof for the perturbation. Furthermore, we can also decide not to consider the classical response of the coupled matter-photon system due to \(\delta n(\mathbf{r}, t)\), but rather directly monitor the quantized modes of the photon field \(\delta q_\alpha(t)\). This response yet again depends on whether we choose to use a classical field \(\delta v(\mathbf{r}, t)\) that induces photons in mode \(\alpha\) or whether we directly generate those photons by an external current \(\delta j_\alpha(t)\). And we also see that the different modes are coupled, i.e., that photons interact. Similarly as charged particles interact via coupling to photons, also photons interact via coupling to the charged particles. Keeping the coupling to the photon field explicitly therefore, on the one hand, changes the standard spectroscopic observables, and on the other hand also allows for many more spectroscopic observables than in the standard matter-only theory as will be discussed in Sec. [ITC]

B. Maxwell-Kohn-Sham linear-response theory

The problem of this general framework in practice is that already in the simplified matter-only theory we usually cannot determine the exact response functions of a many-body system. The reason is that the many-body wave functions, which we use to define the response functions (see App. A), are difficult, if not impossible to determine beyond simple model systems. So in practice we need a different approach that avoids the many-body wave functions. Several approaches exist that instead of wave functions employ reduced quantities [34–39]. The workhorse of these many-body methods is DFT and its time-dependent formulation TDDFT [37–39]. Both theories have been extended to general coupled matter-photon systems within the framework of QED [19–22]. The basic idea of density-functional methods is to reformulate the original Hamiltonian equation given in Eq. (3) in terms of an equivalent and exact quantum-fluid problem [40]. Similar to classical physics, where one usually uses continuum mechanics and the corresponding Navier-Stokes equations to describe many interacting particles, in density-functional methods one uses densities and currents to determine the many-body system. For instance, since the Hamiltonian problem defined by Eq. (4) for a given initial state \(\Psi_0\) provides a bijective mapping between the internal pairs \((n(\mathbf{r}, t), q_\alpha(t))\) and the physical wave functions \(\Psi(\mathbf{r}, t)\), we can express all observables as functionals of the density and displacement coordinate expectation values \(\langle n(\mathbf{r}, t), q_\alpha(t)\rangle\), i.e., we can label all solutions of the TDSE uniquely by their density and displacement coordinate expectation values \(\langle n(\mathbf{r}, t), q_\alpha(t)\rangle\), we can label all observables as functionals of the internal pair \(\Psi([n, q_\alpha], t)\), and the corresponding Navier-Stokes equations to determine the many-body system. This allows us to solve instead of the TDSE equivalently a non-linear fluid equation for the charge density \(n(\mathbf{r}, t)\) coupled non-linearly to the mode-resolved inhomogeneous Maxwell equation [19–22]. While these equations are in principle easy to handle numerically, we do not know the forms of all the different terms explicitly in terms of \(\langle n(\mathbf{r}, t), q_\alpha(t)\rangle\). To find accurate approximations one then employs the Kohn-Sham (KS) scheme, where we model the unknown terms by a numerically easy to handle auxiliary system in terms of wave functions. The simplest approach is to use non-interacting fermions and bosons which lead to a similar set of equations, which are however uncoupled. Enforcing that both give the same density and displacement field dynamics gives rise to mean-field exchange-correlation (Mxc) potentials and currents [33–35]. Formally this Mxc potential and current is defined as the difference of the potential/current that generate a prescribed internal pair in the auxiliary non-interacting and uncoupled system \((\psi_{\text{v}}([n], \mathbf{r}, t), j_\alpha([q_\alpha], t))\) and the potential/current that generates the same pair in the physical system defined by Eq. (1) which we denote by \((\psi([n, q_\alpha], \mathbf{r}, t), j_\alpha([n, q_\alpha], t))\),
i.e.,

\[ v_{\text{Mxc}}([n, q_\alpha], \mathbf{r}, t) = v_s([n], \mathbf{r}, t) - v([n, q_\alpha], \mathbf{r}, t), \]  

\[ j_{\alpha,M}([n], t) = j_\alpha'([q_\alpha], t) - j_\alpha([n, q_\alpha], t) \]

\[ = -\lambda_\alpha \cdot \mathbf{R}(t) \omega^2. \]

In the time-dependent case we only have a mean-field contribution to the Mxc current \([20, 22]\) where the total dipole moment is written as \( \mathbf{R}(t) = \int d^3 \mathbf{r} n(\mathbf{r}, t) \). Further, we have ignored the so-called initial-state dependence because we assume (for notational simplicity) in the following that we always start from a ground state \([41, 42]\) of the matter-photon coupled system. In this way we can recast the coupled Maxwell-quantum-fluid equations in terms of coupled non-linear Maxwell-Kohn-Sham (MKS) equations for auxiliary electronic orbitals, which sum to the total density \( \sum_i |\varphi_i(\mathbf{r}, t)|^2 = n(\mathbf{r}, t) \), and the displacement fields \( q_\alpha(t) \), i.e.,

\[ i\hbar \frac{\partial}{\partial t} \varphi_i(\mathbf{r}, t) = \left[ -\frac{\hbar^2}{2m_e} \nabla^2 + v_{\text{KS}}([v, n, q_\alpha], \mathbf{r}, t) \right] \varphi_i(\mathbf{r}, t), \]

\[ \left( \frac{\partial^2}{\partial t^2} + \omega^2 \right) q_\alpha(t) = -j_\alpha(t) + \omega_\alpha \lambda_\alpha \cdot \mathbf{R}(t). \]

Here we use the self-consistent KS potential \( v_{\text{KS}}([v, n, q_\alpha], \mathbf{r}, t) = v(\mathbf{r}, t) + v_{\text{Mxc}}([n, q_\alpha], \mathbf{r}, t) \) that needs to depend on the fixed physical potential \( v(\mathbf{r}, t) \) \([41]\), and instead of the full bosonic KS equation for the modes \( \alpha \) we just provide the Heisenberg equation for the displacement field. Although the auxiliary bosonic wave functions might be useful for further approximations it is only \( q_\alpha(t) \) that is physically relevant and thus we get away with merely coupled classical harmonic oscillators, i.e., the mode resolved inhomogeneous Maxwell equation. It is then useful to divide the Mxc potential into the usual Hartee-exchange-correlation (Hxc) potential that we know from electronic TDDFT and a correction term that we call photon-exchange-correlation (pxc), i.e.,

\[ v_{\text{Mxc}}([n, q_\alpha], \mathbf{r}, t) = v_{\text{Hxc}}([n, \mathbf{r}, t] + v_{\text{pxc}}([n, q_\alpha], \mathbf{r}, t). \]

Clearly, the correction term \( v_{\text{pxc}} \) will vanish if we take the coupling \( |\lambda_\alpha| \) to zero and recover the purely electronic case. Since by construction the MKS system reproduces the exact dynamics, we also recover the exact linear-response of the interacting coupled system. We can express this with the help of the Mxc kernels defined by the functional derivatives of the Mxc quantities

\[ f_{\text{Mxc}}^n(\mathbf{r}, \mathbf{r}', t) = \frac{\delta v_{\text{Mxc}}(\mathbf{r}, t)}{\delta n(\mathbf{r}', t)}, \quad f_{\text{Mxc}}^{n_\alpha}(\mathbf{r}, \mathbf{r}', t) = \frac{\delta v_{\text{Mxc}}(\mathbf{r}, t)}{\delta q_\alpha(t)}, \]

\[ g^{n_\alpha}_M(t, \mathbf{r}', t') = \frac{\delta j_{\alpha,M}(t)}{\delta n(\mathbf{r}', t)}, \quad g^{n_\alpha_\beta}_M(t, \mathbf{r}', t') = \frac{\delta j_{\alpha_\beta,M}(t)}{\delta q_{\alpha}(t)} = 0. \]

and use the corresponding definitions for the Hxc kernel (that only for the variation with respect to \( n \) has a non-zero contribution) and the pxc kernels. We note that since \( j_{\alpha,M}(t) = \omega_\alpha \lambda_\alpha \int d\mathbf{r} \cdot \mathbf{n}(\mathbf{r}, t) \), we explicitly have

\[ g^{n_\alpha}_M(t - t', \mathbf{r}) = -\delta(t - t') \omega_\alpha \lambda_\alpha \cdot \mathbf{r}. \]

Via these kernels we find with \( \chi^{n,n}_n(\mathbf{r}, \mathbf{r}', t) \) and \( \chi^{n_\alpha,n_\beta}_n(\mathbf{r}, \mathbf{r}', t) \), where \( \chi^{n_\alpha,n_\beta}_n(\mathbf{r}, \mathbf{r}', t) \equiv 0 \) for \( \alpha \neq \alpha' \), the uncoupled and non-interacting response functions (see App. \( B \)) that

\[ \chi^{n_\alpha}_n(\mathbf{r}, \mathbf{r}', t) = \chi^{n_\alpha}_n(\mathbf{r}, \mathbf{r}', t) + \int d\mathbf{r} d\tau \chi^{n_\alpha}_n(\mathbf{r}, \mathbf{x} \tau) \left( \int d\tau' d\mathbf{r}' \chi^{n_\alpha}_n(\mathbf{x} \tau', \mathbf{r} \tau') \right) \]

\[ + \sum_\alpha \int d\tau' f^{n_\alpha}_M(\mathbf{x} \tau, \mathbf{r} \tau') \chi^{n_\alpha}_n(\mathbf{r}, \mathbf{r}' t'), \]

\[ \chi^{n_\alpha}_n(\mathbf{r}, \mathbf{r}', t) = \chi^{n_\alpha}_n(\mathbf{r}, \mathbf{r}', t) + \sum_\beta \int d\tau d\tau' d\mathbf{x} \chi^{n_\alpha}_n(\mathbf{x} \tau, \mathbf{r}') g^{n_\beta}_M(\tau, \mathbf{x} \mathbf{r}') \chi^{n_\beta}_n(\mathbf{r}', t'). \]
\[
\chi_{n}^{\alpha}(r, t, t') = \int dr d\mathbf{x} \chi_{n, \alpha}^{\alpha}(\mathbf{r}, \mathbf{x} r)(\int d\tau' d\mathbf{y} f_{\text{Mxc}}^{n}(\mathbf{x} r, \mathbf{y} r') \chi_{n}^{\alpha}(\mathbf{y} r', t') + \sum_{\alpha} \int d\tau' d\mathbf{y} f_{\text{Mxc}}^{n, \alpha}(\mathbf{x} r, \mathbf{y} r') \chi_{n}^{\alpha}(\mathbf{y} r', t')) ,
\]
\[
\chi_{n}^{\alpha}(t, r, t') = \sum_{\beta} \int d\tau d\mathbf{r} d\mathbf{y} \chi_{\beta, \alpha}^{\alpha}(t, \tau) g_{\alpha}^{\beta}(\tau, \mathbf{y} r') \chi_{n}^{\alpha}(\mathbf{y} r', t').
\]

Here we employed the formal connection between response functions and functional derivatives \(\chi_{n}^{\alpha}(\mathbf{r}, t, t') = \delta n(\mathbf{r}, t)/\delta v(\mathbf{r}', t')\) as well as \(\chi_{n}^{\alpha}(t, t') = \delta q_{\alpha}(t)/\delta j_{\alpha}'(t')\) and accordingly for the auxiliary system (see App. A for details). The Mxc kernels correct the unphysical responses of the auxiliary system to match the linear response of the interacting and coupled problem. So in practice, instead of the full wave function, what we need are approximations to the unknown Mxc kernels. Later we will provide such approximations, show how accurate they perform for a model system and then apply them to real systems. If we decouple light and matter, i.e., \(\Psi_{0} \simeq \psi_{0} \otimes \varphi_{0}\), and disregard the photon part \(\varphi_{0}\) (as is usually done in many-body physics), we recover the response function of Eq. \([15]\) with \(f_{\text{Mxc}}^{n, \alpha} \equiv 0\). The response function, which is calculated with the bare matter initial state \(\psi_{0}\), then obeys the usual Dyson-type equation relating the noninteracting and interacting response in TDDFT \([43]\) with \(v_{\text{Mxc}}([n, q_{\alpha}], \mathbf{r}, t) \rightarrow v_{\text{Hxc}}([n], \mathbf{r}, t)\).

C. Observables in coupled matter-photon response theory

As is obvious from the above discussion, the usual response functions will change and novel response functions are introduced if we keep the matter-photon coupling explicitly. Besides this, we also have new observables that become accessible with the quantum nature of light and well-known observables are changed. We can, for instance, by mixing the perturbations \(\delta v(\mathbf{r}, t)\) and \(\delta j_{\alpha}(t)\) get a different classical induced field. Usually, assuming matter and light decoupled, we determine the induced transversal electric field in Coulomb gauge \(\mathbf{E}(\mathbf{r}, t) = -\frac{1}{c^{2}} \partial_{t} \mathbf{A}(\mathbf{r}, t)\) \([44]\) from the inhomogeneous Maxwell equation in vacuum of Eq. \([2]\)

\[
\left[ \frac{1}{c^{2}} \frac{\partial^{2}}{\partial t^{2}} - \nabla^{2} \right] \mathbf{E}(\mathbf{r}, t) = -\mu_{0} \frac{\partial}{\partial t} \mathbf{J}(\mathbf{r}, t).
\]

Here the inhomogeneity is given by a classical (transversal) current (determined from the perturbed but decoupled matter system) that is connected to the polarization by \(\frac{\partial}{\partial t} \mathbf{J}(\mathbf{r}, t) = \frac{\partial^{2}}{\partial t^{2}} \mathbf{P}(\mathbf{r}, t)\). We can perform a mode-expansion of the electric field and the polarization in terms of eigenmodes, i.e.,

\[
\mathbf{E}(\mathbf{r}, t) = - \sum_{\alpha=1}^{M} \lambda_{\alpha}(\mathbf{r}) E_{\alpha}(t)
\]

\[
\mathbf{P}(\mathbf{r}, t) = \epsilon_{0} \sum_{\alpha=1}^{M} \lambda_{\alpha}(\mathbf{r}) \int d\mathbf{r}' \lambda_{\alpha}(\mathbf{r}) \cdot \mathbf{r} n(\mathbf{r}, t)
\]

Then, in dipole approximation, if we evaluate the quantities at the center of charge, i.e. \(\lambda_{\alpha}(\mathbf{r}) \rightarrow \lambda_{\alpha}\), we find for the electric field amplitude \(E_{\alpha}(t)\)

\[
\left[ \frac{\partial^{2}}{\partial t^{2}} + \omega_{\alpha}^{2} \right] E_{\alpha}(t) = -\frac{\partial^{2}}{\partial t^{2}} \lambda_{\alpha} \cdot \mathbf{R}(t).
\]

Using this kind of approach we can connect \(\delta n(\mathbf{r}, t)\) to the induced vacuum electric field \(\mathbf{E}(\mathbf{r}, t)\). In a final step, to avoid solving the above mode-resolved Maxwell equations, one often even ignores the spatial dependence of the induced field and merely uses \(E_{\alpha}(t) = -\lambda_{\alpha} \cdot \mathbf{R}(t)\). However, we also immediately see that due to a change in \(\chi_{n}^{\alpha}\) as well as due to the new term \(\chi_{n}^{\alpha} / \delta v\), the left-hand side of the approximate induced field can change. Furthermore, the fulfillment of the linear-response kernel in Eq. \([\alpha]\) shows that we get a feedback from the induced photon field onto the matter. Such intrinsic back-reaction effects are very important for large systems, as is well known from solid-state physics, where the bare (vacuum) electric field as determined by Eq. \([22]\) does not agree with the measured spectrum. One needs to include the self-consistent polarization of the system that counter-acts the external perturbing field which leads to the macroscopic Maxwell equations in matter. This can be done in linear response by solving self-consistently a Maxwell equation with the matter response as input \([45, 48]\). The same is true if we consider the vacuum electric field induced by a change in the photon modes, i.e., Eq. \([\alpha]\). In this case we have a back-reaction of the different modes onto each other that changes the density response. But the coupling to the photon modes now also allows to look directly at the quantized field and we do not need to employ the approximate Eq. \([22]\). Instead we can directly look at the Maxwell equations in matter of the photonic displacement field of Eq. \([10]\). Here the electric field takes into account the self-consistent polarization by construction, i.e., \([49]\)

\[
\dot{\mathbf{E}} = \sum_{\alpha=1}^{M} \lambda_{\alpha} \omega_{\alpha} \left( \dot{q}_{\alpha} - \frac{\dot{\lambda}_{\alpha}}{\omega_{\alpha}} \mathbf{R} \right).
\]
The response of the displacement coordinate in first order then becomes in the coupled matter-photon case

\[
\left( \frac{\partial^2}{\partial t^2} + \omega_n^2 \right) \delta q_n(t) = -\frac{\delta j_\alpha(t)}{\omega_n} + \omega_n \lambda_\alpha \int dr \delta n(r, t). \tag{24}
\]

The response of the density is then found with help of the response functions Eqs. (15)-(17). In the case of an external classical field \( \delta v(r, t) \) we then find the induced field by (suppressing detailed dependencies with \( \int dr \rightarrow \int \) and \( \int dr \sum_\alpha \rightarrow \int \))

\[
\left( \frac{\partial^2}{\partial t^2} + \omega_n^2 \right) \delta q_n(t) = \omega_n \lambda_\alpha \cdot \int r \chi^{n}_{n,s} \delta v + \omega_n \lambda_\alpha \cdot \int r \chi^{n}_{n,s} f^{\alpha}_{\text{Mxc}} \chi^{n}_{n,s} \delta q_n + \sum_\alpha r \chi^{n}_{n,s} f^{\alpha}_{\text{Mxc}} \delta q_\alpha. \tag{25}
\]

Here the first term on the right-hand side corresponds to the non-interacting matter-response. However, due to the electron-electron interaction we need to take into account also the self-polarization of interacting matter (second term). Finally, the third term describes the matter-mediated photon-photon response. Let us consider what the terms on the right-hand side mean physically. First of all, in a matter-only theory the self-consistent solution of the Maxwell equations together with the response of the bare matter-system would correspond approximately to the first two terms on the right-hand side. The photon-photon interaction would not be captured in such an approximate approach. Secondly, to highlight the physical content of the different terms we can make the mean-field contributions due to

\[
v_M(rt) = \sum_\alpha \left( \int dr \lambda_\alpha \cdot r n(rt) - \omega_n q_\alpha(t) \right) \lambda_\alpha \cdot r \tag{26}
\]

explicit

\[
\left( \frac{\partial^2}{\partial t^2} + \omega_n^2 \right) \delta q_n(t) = \omega_n \lambda_\alpha \cdot \int r \chi^{n}_{n,s} \delta v + \omega_n \lambda_\alpha \cdot \int r \chi^{n}_{n,s} f^{\alpha}_{\text{Mxc}} \chi^{n}_{n,s} \delta q_n + \sum_\alpha \left( \lambda_\alpha \cdot r_\alpha \right)^2 \chi^{n}_{n,s} \delta v
\]

\[
- \omega_n \lambda_\alpha \cdot \int r \chi^{n}_{n,s} \left( \omega_\alpha \lambda_\alpha \cdot r_\alpha \right) \delta q_\alpha + \omega_n \lambda_\alpha \cdot \int r \chi^{n}_{n,s} f^{\alpha}_{\text{Mxc}} \chi^{n}_{n,s} \delta v + \omega_n \lambda_\alpha \cdot \sum_\alpha r \chi^{n}_{n,s} f^{\alpha}_{\text{Mxc}} \delta q_\alpha. \tag{27}
\]

The second term on the right-hand side then corresponds to the random-phase approximation (RPA) approximation to the instantaneous matter-matter polarization. Here a new term that corresponds to the dipole self-energy induced by the coupling to the photons arises. The third term on the right-hand side is the RPA approximation to the dipole-dipole mediated photon interaction. The rest are xc contributions that arise due to more complicated interactions among the electrons and photons. The last term effectively describe photon-photon interactions mediated by matter. If we only keep the mean-field contributions of the coupled problem we will denote the resulting approximation in the following as photon RPA (pRPA) to distinguish it from the bare RPA of only the Coulomb interaction. We see how the Maxwell equations in matter change for bound charges, i.e., fields due to the polarization of matter, only. A new term, the photon-photon interaction, appears. For free charges, i.e., due to an external charge current \( \delta j_\alpha(t) \), we see similar changes. Clearly, if we would not have a coupling to matter, then there would be no induced density change and we just find the vacuum Maxwell equations coupled to an external current for the electric field. In other terms, the displacement field trivially corresponds to the electric field. However, with the coupling we find the following terms upon perturbing with \( \delta j_\alpha(t) \)

\[
\left( \frac{\partial^2}{\partial t^2} + \omega_n^2 \right) \delta q_n(t) = -\frac{\delta j_\alpha(t)}{\omega_n} + \omega_n \lambda_\alpha \cdot \int r \chi^{n}_{n,s} f^{\alpha}_{\text{Mxc}} \chi^{n}_{n,s} \delta j_\alpha + \omega_n \lambda_\alpha \cdot \sum_\alpha r \chi^{n}_{n,s} f^{\alpha}_{\text{Mxc}} \delta q_\alpha. \tag{28}
\]
Here the second term on the right hand side describes the full polarization of the quantized medium due to the photon field and the third term highlights the matter-mediated photon-photon interaction. Making again the mean-field explicit leads to

\[
\frac{\partial^2}{\partial t^2} + \omega_n^2 \delta q_\alpha(t) = -\frac{\delta j_\alpha(t)}{\omega_n^2} + \omega_n \lambda_\alpha \cdot \int r \chi_{n,s}^n \left[ \frac{e^2}{4\pi\varepsilon_0 |r' - r|^2} + \sum_{\alpha'} (\lambda_{\alpha'} \cdot r')^2 \right] \chi_{q_\alpha}^n \delta j_\alpha - \omega_n \lambda_\alpha \cdot \sum_{\alpha'} \int r \chi_{n,s}^n (\omega_{\alpha'} \lambda_{\alpha'} \cdot r') \delta q_{\alpha'}.
\]

If we ignore the xc contributions to the matter-photon and photon-photon response we get the pRPA approximation to the Maxwell equation in matter. In this pRPA form we clearly see how the Maxwell equation becomes non-linear because of the feedback between light and matter. Such non-linearities of the Maxwell equations are investigated in great detail in high-energy physics in the context of strong-field QED [50]. In this case the strong fields lead to particle creation and thus a matter-mediated photon-photon interaction. In our case, we do not need these high energies because we consider the photon-photon interaction due to condensed matter in form of atoms, molecules or solids and use, e.g., a cavity to enhance the coupling.

Besides the above highlighted changes in, e.g., the Maxwell equations, novel observables become accessible. For instance, one can monitor the response of the matter system due to a perturbation of the photonic subsystem by an external current. This allows to investigate directly the cross-correlation between the matter and the photon subsystem induced by \( \chi_{n,s}^n \). Also note, that this cross-correlation observable allows to distinguish between the response due to a purely classical field \( \delta \nu \) or due to a quantized field, since \( \delta j_\alpha(t) \) generates photons that then perturb the correlated matter-photon system. A further important observable that can be captured in this approach is the intrinsic lifetimes of excited states, which is not accessible in standard matter-only quantum mechanics. Let us briefly explain what we mean by this. In standard quantum mechanics we find besides the ground state also other eigenstates, i.e., excited states. Hereby an eigenstate is a square-integrable eigenfunction of a self-adjoint, usually unbounded Hamiltonian. If we excite a matter system from its ground state into such an excited state, it will remain in this state as long as we do not perturb it. In quantum mechanics we then also have generalized eigenstates, so-called scattering states, which are not square-integrable and that constitute the continuous spectrum of such a Hamiltonian [51]. The simplest example is the free electron Hamiltonian \( T = \sum_{i=1}^N -\frac{\hbar^2}{2m_e} \nabla_i^2 \) which in infinite space has a purely continuous spectrum consisting of non-normalizable plane-waves [52]. The physical interpretation of such scattering states - as already the name indicates - is that particles propagate to infinity and do not stay bound anywhere. Thus exciting a matter system from its ground state into such a generalized eigenstate corresponds to the physical process of ionization. Ionization, however, is something completely different than the process of spontaneous emission. That is, if we put an atom or molecule into an “excited state”, even without a further perturbation it will relax to the ground state by emitting radiation. The time the system stays in this “excited state” before emitting a photon is called the lifetime. The process of spontaneous clearly cannot be captured by standard quantum mechanics where matter and light are decoupled. Non-relativistic QED, however, does capture this process [2] by coupling the matter system to the quantized electromagnetic field which consists of infinitely many harmonic oscillators. In this way the excited states of the bare matter system turn into resonances and the ground state (usually) remains the only eigenstate of the combined matter-photon system. While formally these resonances are indeed scattering states of the combined matter-photon system, it is only the photonic part that shows a scattering behavior, i.e., a photon leaves the vicinity of the matter subsystem. The matter subsystem just relaxes to the only stable state, its ground state [2]. In linear response such relaxation processes express themselves as finite line-widths of excitations, where the linewidth can be associated with the lifetimes of different resonances. In our slightly simplified treatment based on Eq. (1) we only consider finitely many photon modes, and hence we do not have genuine resonances. However, by including enough modes we sample the influence of the vacuum and instead of one sharp transition peak (which numerically is usually artificially broadened) we get many that approximate the resonance. In this way linear-response theory for non-relativistic QED in the long wave-length limit can determine lifetimes of real systems.

D. Linear-response theory as a pseudo eigenvalue problem

After having investigated the changes in observables and new types of responses we need to find an efficient way to solve these linear-response equations in terms of the MKS system. First, performing a Fourier transfor-
mation from time $t$ to frequency space $\omega$ and using the 
Hxc and pxc kernels we find

$$\chi_n = \chi_{n,s} + \sum_{\alpha} f_{\alpha} g_n \chi_{n,\alpha},$$

(30)

$$\chi_n^{Q_\alpha} = \sum_{\beta} g_{n,\beta} \chi_{n,\beta},$$

(31)

$$\chi_n^{Q_\alpha} = \sum_{\beta} f_{\alpha} g_n \chi_{n,\beta},$$

(32)

$$\chi_n^{Q_\alpha,\alpha} = \chi_{n,\alpha} + \sum_{\beta} g_{n,\beta} \chi_{n,\beta},$$

(33)

The Eqs. (30)-(33) are coupled with respect to the 
external perturbations. The perturbation with respect to 
the external potential $\delta V(r)$ results in a coupled 
set of response functions $\{\chi_n^{Q_\alpha}(r,\omega)\}$ and 
for the current $\delta j_{\alpha}(t)$ gives the coupled set 
$\{\chi_n^{Q_\alpha}(t,\omega), \chi_n^{Q_\alpha}(t,\omega)\}$.

$$\begin{pmatrix}
K(\Omega) & M(\Omega) \\
K^*(\Omega) & M^*(\Omega)
\end{pmatrix}
\begin{pmatrix}
X_1(\Omega_q) \\
Y_1(\Omega_q)
\end{pmatrix}
= \begin{pmatrix}
1 & 0 & 0 & 0 \\
0 & -1 & 0 & 0
\end{pmatrix}
\begin{pmatrix}
X_1(\Omega_q) \\
Y_1(\Omega_q)
\end{pmatrix},
$$

(38)

where $\Omega_q$ refers to the many-body excitation energies.
The quantity $L_{\alpha i,\beta j}(\Omega_q) = \delta_{ij} \delta_{\alpha \beta} (\epsilon_{\alpha} - \epsilon_{\beta}) + K_{\alpha i,\beta j}(\Omega_q)$ 
contains the difference of two Kohn-Sham energies $\epsilon_{\alpha}$ and 
$\epsilon_{\beta}$, where $i$ refers to occupied orbitals and the index $a$ to 
unoccupied orbitals. The quantity $K$ is given by

$$K_{\alpha i,\beta j}(\Omega_q) = \int \left\{ \right. \delta_{ij} \epsilon_{\alpha} \phi_{\alpha}(r) \phi_{\beta}(r) f_{\alpha} f_{\beta} \left. \right\},$$

(39)

In comparison to the standard linear response formulation of TDDFT, we also have now two new coupling functions 
appearing, $M$ and $N$ that couple the matter block 
with the photon block.

$M_{\alpha,\alpha}(\Omega_q) = \int \left\{ \right. \delta_{ij} \epsilon_{\alpha} \phi_{\alpha}(r) \phi_{\beta}(r) f_{\alpha} f_{\beta} \left. \right\},$

(40)

$N_{\alpha,\alpha} = \frac{1}{2\omega_{\alpha}^2} \int \delta_{ij} \epsilon_{\alpha} \phi_{\alpha}(r) g_{M,M}(r),$

(41)

We emphasize here that the exact coupling matrix $N_{\alpha,\alpha}$ 
has no frequency dependence since the exact kernel for 
Eq. (41) is equivalent to just the mean-field kernel of 
the photon modes. Given the exact kernels, the nonlin-
ear pseudo eigenvalue problem in Eq. (38) allows to 
compute the exact excitation energies of the coupled matter-
photon system. Of course, in practice, approximations 
functions have to be solved in a self-consistent way to 
obtain the exact interacting response functions. The re-
sponse functions of Eqs. (39) and (40) can be expressed in 
frequency space through a Fourier transform that yields

$$\delta n_{\alpha}(r,\omega) = \int d\omega' \chi_n^{Q_\alpha}(r,\omega') \delta V(r'),$$

(34)

$$\delta q_{\alpha,\beta}(r,\omega) = \int d\omega' \chi_n^{Q_\alpha,\beta}(r,\omega') \delta V(r'),$$

(35)

$$\delta n_{\beta}(r,\omega) = \sum_{\alpha} \chi_n^{Q_\alpha,\beta}(r,\omega) \delta j_{\alpha}(\omega),$$

(36)

$$\delta q_{\alpha,\beta}(\omega) = \sum_{\alpha'} \chi_n^{Q_\alpha,\beta}(\omega) \delta j_{\alpha'}(\omega).$$

(37)

A perturbation from the external potential $V(r,\omega)$ indu-
ces the responses $\delta n_{\alpha}(r,\omega)$ and $\delta q_{\alpha,\beta}(\omega)$. Making a 
substitution of Eqs. (39) and (40) into the density and 
displacement field response due to an external potential $V(r,\omega)$ yields after some algebra the following eigenvalue problem (for a detailed derivation, we refer the reader to 
App. C)
given explicitly by
\begin{equation}
U_{qq'} = \delta_{qq'}\omega_q^2 + 2\sqrt{\omega_q\omega_{q'}}K_{qq'}(\Omega_q),
\end{equation}
\begin{equation}
V_{qa} = 2\sqrt{\omega_aM_a(\Omega_a)\omega_\alpha},
\end{equation}
\begin{equation}
W_{aq} = 2\sqrt{\omega_a2M_a(\Omega_q)\omega_q}.
\end{equation}
The index \( q = (a, i) \) describes transitions from electronic occupied \((i)\) to unoccupied states \((a)\) and thus difference of Kohn-Sham energies is given by \( \omega_q = \epsilon_a - \epsilon_i \). With \( \alpha \) we denote the photon modes. The eigenvectors \( \mathbf{E}_1 \) and \( \mathbf{P}_1 \) can be used to compute oscillator strengths of the coupled matter-photon system (see App. D). In the decoupling limit of light-matter interaction, Eq. (42) reduces to the well-known Casida equation \( U\mathbf{E}_1 = \Omega^2\mathbf{E}_1 \) [54]. So far we did not solve anything but have just rewritten the problem in terms of unknown Mxc kernels that correct the uncoupled and non-interacting auxiliary response functions. To actually solve this problem we need to provide approximations to these unknown quantities. Here it can becomes advantageous to have divided the full Mxc kernels in Hxc and pxc terms, such that we can use well-established approximations from electronic TDDFT for the Hxc and specifically developed approximations for the pxc terms (see Sec. III for more details). In the following we will not pursue this approach, but instead choose a simple yet consistent approximation for all the parts of the Mxc kernels. We will employ the above introduced pRPA approximation, which is a straightforward generalization of the standard RPA of electronic-structure theory. We therefore set all the xc parts to zero and only keep the mean-field parts, i.e.,
\begin{equation}
f_{\mu}^a(\mathbf{r}, \mathbf{r}') = \frac{\epsilon_a^2}{4\pi\epsilon_0 |\mathbf{r} - \mathbf{r}'|}, \quad f_{\mu}^a(\mathbf{r}) = - \sum_{\alpha} (\lambda_{\alpha} \cdot \mathbf{r} )^2, (46)
\end{equation}
\begin{equation}
f_{p}^a(\mathbf{r}) = - \sum_{\alpha} \omega_{\alpha} \lambda_{\alpha} \cdot \mathbf{r}, \quad g_{M}^a(\mathbf{r}) = - \omega_{\alpha} \lambda_{\alpha} \cdot \mathbf{r}. (47)
\end{equation}
We note that on the pRPA level the matter-photon coupling mediated via \( g_{M}^a \) is exact. The influence of the discarded photon-matter xc contributions \( f_{xc}^a \) and \( f_{xc}^f \) will be highlighted in the next section. In the pRPA approximation all the frequency dependence that we suppressed at times for brevity now genuinely vanishes (an adiabatic approximation) which allows us to express \( M \) and \( N \) of Eqs. (40) and (41) as
\begin{equation}
M_{\alpha,\alpha, a} = - \omega_{\alpha} \int d\mathbf{r}\varphi_i(\mathbf{r})\varphi^*_a(\mathbf{r})\lambda_{\alpha} \cdot \mathbf{r}, (48)
\end{equation}
\begin{equation}
N_{\alpha, \alpha, a} = - \frac{1}{2} \int d\mathbf{r}\varphi^*_i(\mathbf{r})\varphi_a(\mathbf{r})\lambda_{\alpha} \cdot \mathbf{r}. (49)
\end{equation}
Next we want to connect to the standard matter-only linear-response framework. In defining the oscillator strength for the density-density response function, we make use of the relationship between the polarizability tensor and susceptibility. The first-order dipole polarizability is given by
\begin{equation}
\delta \mathbf{R}(t) = \int d\mathbf{r} \, d\mathbf{r} \, \delta n(\mathbf{r}, t), (50)
\end{equation}
and in frequency space \( \mathbf{R}(\omega) = \frac{\epsilon_0}{\omega} (\omega) \mathbf{E}(\omega) \). The dynamic polarizability tensor can then be written as
\begin{equation}
\frac{\epsilon_0}{\omega} (\omega) = \int d\mathbf{r} \frac{\delta n(\mathbf{r}, \omega)}{\delta E(\omega)}. (51)
\end{equation}
where we use the external potential \( v(\mathbf{r}, \omega) = \mathbf{r} \cdot \mathbf{E}(\omega) \).

III. EXAMPLES FOR THE COUPLED MATTER-PHOTON RESPONSE

Although we are considering a well-known problem, we have taken a slightly different perspective on the response of real many-body systems. Therefore many ideas and concepts we have used to connect to standard linear-response theory are less well-known. We therefore want - before we show results for a real system - to take a step back and discuss this new perspective in more detail for a simple yet non-trivial model system. Since it is not the interaction among the particles but between charged particles and photons that led to a change in perspective, we focus in the following on a simple model of one particle coupled to photons. We employ a slight generalization of the Rabi model [55, 56], which is the standard model of quantum optics. The model Hamiltonian we consider is given by (in this section we switch for simplicity to atomic units)
\begin{equation}
\hat{H}_R(t) = \frac{\omega_0}{2} \hat{a}^\dagger \hat{a} + \omega_\lambda \hat{a}^\dagger \hat{a} + \lambda \hat{a} \hat{q} + j(t) \hat{q} + v(t) \hat{\sigma}_x. (55)
\end{equation}
where $\omega_0$ is the transition frequency between the ground state $|g\rangle$ and excited state $|e\rangle$ and $\hat{\sigma}_z$ as well as $\hat{\sigma}_x$ are the usual Pauli matrices. We only keep one photon mode with frequency $\omega_c$ and use the usual photon creation and annihilation operators to represent the harmonic oscillator of this mode. By further compressing the notation, we then describe the coupling between matter and light by a coupling strength $\lambda$ and the adopted displacement coordinate $\hat{q} = (\hat{a} + \hat{a}^\dagger)$. Finally, we couple the matter system to a classical external perturbation $v(t)$ and the photon system to a classical external current $j(t)$ (a pictorial representation of the coupled system is given in Fig. 1). We note for consistency with respect to other works [21, 24, 25] that in the above Rabi model we can perform a unitary transformation that allows us to exchange $\hat{\sigma}_x$ and $\hat{\sigma}_z$. Both forms of the extended Rabi model are therefore equivalent. We further note that with respect to the full non-relativistic QED problem in the long wave-length approximation of Eq. (11) the Rabi model does not include the dipole self-energy term proportional to $(\hat{\mathbf{R}} \cdot \hat{\mathbf{r}})^2$. This is because the analogous term in this model is just a constant energy shift, i.e., it is proportional to $\sigma_x^2 = 1$. For more levels this is no longer the case [24] and this term has to be taken into account, else the resulting eigenstates do not have a proper continuum limit [34]. The responses that we want to consider in the following are those observables that couple to the external perturbations. In our case this is $\sigma_x(t) = \langle \Psi(t)|\hat{\sigma}_x|\Psi(t)\rangle$ (in essence the atomic dipole) and the displacement field $q(t) = \langle \Psi(t)|\hat{q}|\Psi(t)\rangle$. The response of these observables $\langle \delta \sigma_x(t), \delta q(t) \rangle$ to perturbations by the external pair $(\delta v(t), \delta j(t))$ can be written similarly as Eq. (6) in the collective form

$$
\begin{bmatrix}
\delta \sigma_x(t) \\
\delta q(t)
\end{bmatrix} = \int dt' \begin{bmatrix}
\chi_{xg}^\sigma(t,t') \\
\chi_{xq}^\sigma(t,t')
\end{bmatrix} \begin{bmatrix}
\delta v(t') \\
\delta j(t')
\end{bmatrix}.
$$

(56)

Again we find besides the usual matter-matter response $\chi_{xg}^\sigma$, also matter-photon responses $\chi_{xq}^\sigma$ and $\chi_{xq}^\sigma$, respectively, as well as a photon-photon response function $\chi_{qq}^\sigma$. Next, in analogy to Sec. II we reformulate the coupled matter-photon problem in form of a MKS auxiliary problem. Using by now well-established results of QEDFT for the extended Rabi model systems [21, 24] we can introduce two effective fields

$$
v_{\text{Mxc}}([\sigma_x, q]; t) = v_s([\sigma_x, q]; t) - v([\sigma_x, q]; t),
$$

(57)

$$
j_M([\sigma_x]; t) = j_s(q; t) - j([\sigma_x]; t),
$$

(58)

that force the auxiliary uncoupled, yet non-linear MKS system to generate the same dynamics of the internal pair $(\sigma_x(t), q(t))$ as the corresponding coupled reference system. For an uncoupled initial MKS state $|\Phi_0\rangle = |\psi_0\rangle \otimes |\phi_0\rangle$ that provides the same initial conditions for the internal pair as the physical initial state [21, 24], we then have to solve self-consistently

$$
\begin{aligned}
\frac{i}{\hbar} & \frac{\partial}{\partial t} |\psi(t)\rangle = \begin{bmatrix}
\frac{\omega_0}{2} & \delta z + (v(t) + v_{\text{Mxc}}([\sigma_x, q]; t)) \delta x \\
(\omega_c^2 + \omega_e^2) & -2\omega_c j(t) - 2\omega_c \lambda \sigma_x(t).
\end{bmatrix} |\psi(t)\rangle,
\end{aligned}
$$

(59)

Here we determine the internal matter observable from $\sigma_x(t) = \langle \psi(t)|\hat{\sigma}_x|\psi(t)\rangle$. Since the photon subsystem is merely a shifted harmonic oscillator we get away with only solving the classical harmonic oscillator equation coupled to the dipole of the matter subsystem. We can then express the coupled response functions of Eq. (56) in analogy to Eqs. (15)-(18) by the uncoupled auxiliary response functions $\chi_{xg}^\sigma, \chi_{xg}^\sigma, \chi_{xg}^\sigma, \chi_{xq}^\sigma, \chi_{xq}^\sigma, \chi_{xq}^\sigma, \chi_{qq}^\sigma, \chi_{qq}^\sigma, \chi_{qq}^\sigma$ and $\chi_{qq}^\sigma$ as

$$
\chi_{xg}^\sigma(t, t') = \chi_{xg}^\sigma(t, t') + \int d\tau \int d\tau' \chi_{xg}^\sigma(t, \tau) f_{\text{Mxc}}(\tau, \tau') \chi_{xg}^\sigma(\tau', t')
$$

(60)

$$
\chi_{xq}^\sigma(t, t') = \int d\tau \int d\tau' \chi_{xq}^\sigma(t, \tau) f_{\text{Mxc}}(\tau, \tau') \chi_{xq}^\sigma(\tau', t'),
$$

(62)

$$
\chi_{qg}^\sigma(t, t') = \int d\tau \int d\tau' \chi_{qg}^\sigma(t, \tau) g_{\text{Mxc}}(\tau, \tau') \chi_{qg}^\sigma(\tau', t'),
$$

(63)

$$
\chi_{qg}^\sigma(t, t') = \int d\tau \int d\tau' \chi_{qg}^\sigma(t, \tau) g_{\text{Mxc}}(\tau, \tau') \chi_{qg}^\sigma(\tau', t'),
$$

(64)

The only real difference is in the Rabi case we do not have a longitudinal interaction and therefore the Mxc contributions come solely from the matter-photon coupling, i.e., $f_{\text{Mxc}}^\sigma = f_{\text{ps}}^\sigma$ and $f_{\text{Mxc}}^q = f_{\text{pse}}^q$. This allows us to study exclusively the influence of these new terms and how approximations of them perform.

A. Matter-photon correlation effect in Maxwell equations

Let us follow the previous general section and briefly consider the influence of the matter-photon coupling
on the Maxwell equations in this model system, i.e., Eq. (60). The inhomogeneous Maxwell equation here accounts for the back-reaction of the matter on the field through the atomic dipole operator $\sigma_x([v, j]; t)$. If we, for instance, perturb the two-level system directly via $\delta v(t)$, the response of the Maxwell equation expressed in terms of the uncoupled problem with the help of Eq. (62) becomes

$$(\partial_t^2 + \omega_c^2) \delta q(t) = -2\lambda \omega_c \int dt' \chi_{\sigma_x, s}^e(t, t') \delta v(t') - 2\lambda \omega_c \int \int dt' \int dt'' \chi_{\sigma_x, s}^e(t, \tau) f_{\text{Mxc}}^q(t, \tau, \tau') \delta q(\tau').$$

Having no coupling, i.e., the Mxc terms are zero, merely recovers the usual inhomogeneous Maxwell equation for a classical external current. The matter system evolves according to the perturbation and we can determine its induced Maxwell field without any back-reaction. The second term describes the matter polarization due to the induced field and leads to an effective self-interaction of the two-level system. If there would be more than one particle this would induce an effective matter-matter interaction as well. The third term then accounts for the field polarization and induces an effective self-interaction in the mode of the light field. That is, the coupling to matter leads to a photon-photon interaction. This can be made more explicit by separating the mean-field contribution $v_M(t) = \lambda q(t)$ and rewriting the above equation as

$$(\partial_t^2 + \omega_c^2) \delta q(t) = -2\omega_c \delta j(t) - 2\lambda^2 \omega_c \int dt \chi_{\sigma_x, s}^e(t, \tau) \delta q(\tau) - 2\lambda \omega_c \int dt \int dt' \chi_{\sigma_x, s}^e(t, \tau) f_{\text{Mxc}}^q(\tau, \tau') \delta q(\tau').$$

The third term on the right-hand side is then the pRPA form of photon-photon response. Similar terms also appear for a perturbation induced by an external current $\delta j(t)$ which can be rewritten with the help of Eq. (63) and the mean-field made explicit as

$$(\partial_t^2 + \omega_c^2) \delta q(t) = -2\omega_c \delta j(t) - 2\lambda^2 \omega_c \int dt \chi_{\sigma_x, s}^e(t, \tau) \delta q(\tau) - 2\lambda \omega_c \int dt \int dt' \chi_{\sigma_x, s}^e(t, \tau) f_{\text{Mxc}}^q(\tau, \tau') \delta q(\tau').$$

Here we used that $f_{\text{Mxc}}^q(\tau, \tau') = \lambda \delta(\tau - \tau')$. As is most obvious in the pRPA limit, for both types of perturbations lead to the same resonance conditions, i.e., peaks in the responses. They are connected to the combined eigenstates of the matter-photon system. However, the detailed response can differ strongly. That these resonance conditions that we get from the pRPA are indeed connected to the coupled eigenstates we will show next.

**B. Application of the pseudo-eigenvalue problem**

As a preparatory step we first rewrite the linear-response problem of the extended Rabi model in terms of the previously introduced pseudo-eigenvalue problem of Eq. (42). In the two-level one-mode case we consider here, this reduces to

$$\left( \begin{array}{cc} U(\Omega_q) & V(\Omega_q) \\ W(\Omega_q) & \omega_c^2 \end{array} \right) \left( \begin{array}{c} E_1 \\ P_1 \end{array} \right) = \Omega_q^2 \left( \begin{array}{c} E_1 \\ P_1 \end{array} \right).$$

Where the matrices in the model system reduce to functions of $\Omega_q$ as $U = \omega_c^0 + 2\omega_c K(\Omega_q)$, $V(\Omega_q) = 2\omega_c^{1/2} M(\Omega_q)^{1/2} N^{1/2} \omega_c^{1/2}$, and $W(\Omega_q) = 2\omega_c^{1/2} N^{1/2} M(\Omega_q)^{1/2} \omega_c^{1/2}$. The coupling functions are...
given explicitly using Eqs. (25), (41) as
\[
K(\Omega_q) = f_\alpha^\sigma + f_\sigma^\tau_x(\Omega_q),
\]
\[
M(\Omega_q) = f_\sigma^\tau_z + f_\tau_x^\sigma(\Omega_q),
\]
\[
N = g_{\alpha\sigma}^\tau,
\]
where the Kohn-Sham states is the dipole matrix element \( \varphi_\alpha \varphi_\sigma^* = \langle g | \hat{\sigma}_x | c \rangle = 1 \). The Mxc kernels can be defined using the inverse of the auxiliary and interacting response functions (see also Eqs (37), (38) in the appendix) and are given in frequency space by
\[
f_{\alpha\sigma}^\tau_z(\omega) = (\chi_{\sigma\sigma}^\tau(\omega))^{-1} - (\chi_{\alpha\sigma}^\tau(\omega))^{-1},
\]
\[
f_{\sigma\tau_z}^\alpha(\omega) = - (\chi_{\alpha\sigma}^\tau(\omega))^{-1}.
\]
Here \( (\chi_{\sigma\sigma}^\tau(\omega))^{-1} \) and \( (\chi_{\alpha\sigma}^\tau(\omega))^{-1} \) are the inverses of the uncoupled response function of the electronic subsystem, the fully coupled response function of the electronic dipole and of the displacement field of the Rabi model, respectively. With these quantities we then determine spectroscopic observables such as the photoabsorption cross section. To determine this cross section we first note that the linear polarizability \( \alpha(\omega) \) induced by the external potential \( v(\omega) \) is related to the “dipole-dipole” response function as \( \alpha(\omega) = -\chi^\sigma_x(\omega) \). Using Eq. (54), we can determine the photoabsorption cross section of the Rabi model (see Fig. 2 a displayed in dotted-red for the numerically exact case).
\[
\sigma(\omega) = -4\pi\omega \Im \chi_{\sigma\sigma}^\tau(\omega).
\]

Analogously, we define a linear “field polarizability” \( \beta(\omega) \) due to polarizing the photon mode by an external current. In the same way, we relate the field polarizability to the response function of the photon mode as \( \beta(\omega) = -\chi_{\tau\tau}^\sigma(\omega) \) and then determine a photonic spectrum from (see Fig. 2 b displayed in dotted-red for the numerically exact case).
\[
\sigma(\omega) = -4\pi\omega \Im \chi_{\tau\tau}^\sigma(\omega).
\]

Finally, we consider mixed spectroscopic observables where we perturb one subsystem and then consider the response in the other. We analogously employ \( \chi^\sigma_x(\omega) \) and \( \chi^\tau_x(\omega) \) in Eqs. (71) and (72) respectively, to determine a “mixed polarizability”. If we plot this mixed spectrum (see Fig. 2 c displayed in dotted-red for the numerically exact case), we find that we have positive and negative peaks. Indeed, this highlights that excitations due to external perturbations can be exchanged between subsystems, i.e., energy absorbed in the electronic subsystem can excite the photonic subsystem and vice versa. Next, we want to employ the pRPA approximation to the extended Rabi model and try to solve it analytically. The pRPA is equivalent to using the mean-field approximation in the coupled equations, i.e., approximating the electron-photon coupling term as \( \hat{\sigma}_x \hat{q} \approx \langle \hat{\sigma}_x \rangle \hat{q} + (\hat{q}) \hat{\sigma}_x \). This corresponds then to a coupled Schrödinger-Maxwell treatment of the coupled matter-photon problem [1]. In the MKS equations this leads to approximating the full \( \phi_{\mathrm{Mxc}} \) by the mean-field potential \( v_{\mathrm{M}} = \lambda q. \) In the case of the pseudo-eigenvalue problem this amounts to approximating \( K = f_\alpha^\sigma = 0, M = f_\sigma^\tau = \lambda \) and \( N = g_{\alpha\sigma}^\tau = \lambda \). Consequently we have
\[
U = \omega_0^2, \quad V = W = 2\lambda\sqrt{\omega_0\omega_c}, \quad \omega_0^2 = \omega_c^2.
\]
The resulting nonlinear eigenvalue equation yields the excitation frequencies
\[
\Omega_1^2 (-) = \frac{1}{2} (\omega_0^2 + \omega_c^2) - \frac{1}{2} \sqrt{(\omega_0^2 - \omega_c^2)^2 + 16\lambda^2\omega_0\omega_c,}
\]
\[
\Omega_1^2 (+) = \frac{1}{2} (\omega_0^2 + \omega_c^2) + \frac{1}{2} \sqrt{(\omega_0^2 - \omega_c^2)^2 + 16\lambda^2\omega_0\omega_c},
\]
and the corresponding normalized eigenvectors can be given in closed form as
\[
E_1 = \left( -\sin \theta \quad \cos \theta \right), \quad \text{and} \quad P_1 = \left( \cos \theta \quad \sin \theta \right)
\]
with the mixing angle defined using
\[
\sin 2\theta = \frac{-4\lambda\sqrt{\omega_c\omega_c}}{\sqrt{(\omega_c^2 - \omega_0^2)^2 + 16\lambda^2\omega_0\omega_c}},
\]
\[
\cos 2\theta = \frac{(\omega_c^2 - \omega_0^2)}{\sqrt{(\omega_c^2 - \omega_0^2)^2 + 16\lambda^2\omega_0\omega_c}}.
\]

Using these analytic results, we can then construct the photoabsorption cross section where we obtained one of the strengths of \( \alpha(\omega) \) using the eigenfunctions above as
\[
f_1 = 2 \left| r_\uparrow \right|^2 S^{1/2} E_1 \left| r_\uparrow \right|^2 = 2\omega_0 \sin^2 \theta
\]
where \( r_\uparrow = \langle g | \sigma_x | c \rangle \uparrow = 1 \) and \( S^{1/2} = \omega_0^{1/2} \). The resulting pRPA-approximated spectra are displayed in Fig. 2 in dashed-blue. We will discuss the results in a little more detail at the end of this section. Before we consider a slightly more advanced approximation based on the rotating-wave approximation (RWA). If we slightly simplify the full Rabi problem by approximating the full coupling as \( \hat{\sigma}_x \hat{q} \approx (\hat{\sigma}_x \hat{\alpha} + \hat{\sigma}_\hat{\alpha} \hat{\alpha}) \) we end up with the Jaynes-Cumming Hamiltonian [37] given as
\[
\hat{H}_{JC}(t) = \frac{\omega_0}{2} \hat{\sigma}_x + \omega_0 \hat{a}^{\dagger} \hat{a} + \lambda (\hat{\sigma}_x \hat{\alpha} + \hat{\sigma}_\hat{\alpha} \hat{\alpha}) + j(t) \hat{q} + v(t) \hat{\sigma}_x.
\]
Here we used \( \hat{\sigma}_\pm = (\hat{\sigma}_x \pm i\hat{\sigma}_y)/2 \). The above approximation is called the RWA because we ignore quickly oscillating terms and thus assume that the excitation of the matter subsystem can only destroy and the de-excitation only create a photon. This approximation is justified (with respect to the full wave function) if we are in the weak coupling regime, i.e., \( \lambda \ll \omega_c \), and near to resonance, i.e., \( \delta = \omega_0 - \omega_c \approx \omega_c \). The ground-state of the
Jaynes-Cummings model is the uncoupled tensor product of the matter ground-state and the photon ground-state with ground-state energy of $E_0 = -\omega_0/2$. The excited states of the Jaynes-Cummings Hamiltonian are known analytically and are given by (we only show the lowest lying excited states where a single photon is excited)

$$| -, 0 \rangle = -\sin \theta_0 |g\rangle |1\rangle + \cos \theta_0 |e\rangle |0\rangle, \quad (78)$$

$$| +, 0 \rangle = \cos \theta_0 |g\rangle |1\rangle + \sin \theta_0 |e\rangle |0\rangle, \quad (79)$$

where

$$\sin \theta_0 = \frac{2\lambda}{\sqrt{4\lambda^2 + (\Omega_0 - \delta)^2}}, \quad \cos \theta_0 = \frac{\Omega_0 - \delta}{\sqrt{4\lambda^2 + (\Omega_0 - \delta)^2}},$$

and $\Omega_0 = \sqrt{\delta^2 + 4\lambda^2}$. With these eigenstates we find the transition frequencies that correspond to the linear response from the ground state (only one photon absorbed or emitted) to be

$$\Omega_-(0) = \frac{1}{2} (\omega_c + \omega_0 - \Omega_0), \quad (80)$$

$$\Omega_+(0) = \frac{1}{2} (\omega_c + \omega_0 + \Omega_0). \quad (81)$$

So we already know where the RWA will generate the poles of the response function. Since we know analytically the eigenfunctions in the RWA, we can construct the RWA response functions analytically. Using the definitions of the Mxc kernels of Eq. (69) we can then analytically construct the RWA Mxc kernels. These kernels are frequency dependent, therefore the resulting Mxc approximation is non-adiabatic [39]. Substituting them into $M(\Omega_q)$ we recover the known poles $\Omega_q = \Omega_\pm(0)$ from Eq. (68). Further, we can then construct the different spectra associated with the RWA. We show them in Fig. 2 in full-orange.

Let us now analyze the obtained spectra and scrutinize the different approximations to the Mxc kernels. In Fig. 2a, b and c we see how the spectra change for an increasing coupling strength $\lambda$. Already for small coupling the originally degenerate states of the uncoupled matter-photon systems show an avoided crossing that leads to the splitting of the electronic state into an upper and lower polariton. Approximatively these states are given in terms of the RWA as $|+, 0\rangle$ and $|-, 0\rangle$. The difference in energy between the lower and upper polariton is called the Rabi splitting $\Omega_R$ and is used to indicate
the strength of the matter-photon coupling. In molecular experiments values of up to $\Omega_R/\omega_c \approx 0.25$ have been measured \cite{59,60}. Up to $\lambda = 0.1$ the different spectra for the exact (dotted-red), the pRPA (dashed-blue) as well as the RWA (full-orange) agree well. Already the mean-field treatment is enough to recover the quantized matter-photon responses, even for the coupled matter-photon spectra in Fig.\ref{fig:2}c. Consequently the pRPA seems a reasonable approximation for linear-response spectra even for strong coupling situations. Only upon increasing the coupling strength further and thus going into the ultra-strong coupling regime, first discrepancies appear.

For ultra-strong coupling (for $\lambda = 0.3$ the Rabi splitting is already of the order of 0.5) the approximations do not recover the exact results. Increasing further leads then to not only a disagreement in transition frequencies but also the weights of the transitions become increasingly different. Besides a simple check for the approximations to the Mxc kernels, the extended Rabi model also allows us to get some understanding of the novel response functions $\chi_{\sigma q}^{\sigma x}, \chi_{\sigma q}^{\sigma p}$ and $\chi_{\sigma q}^{\sigma q}$. The oscillator strength of the photonic spectrum (based on $\chi_{\sigma q}^{\sigma q}$) in Fig.\ref{fig:2}b provides us with a measure of how strong the displacement field (and with this also the electric field) reacts to an external classical charge current with frequency $\omega$. We see, for instance, for $\lambda = 0.01$ that the photon subsystem mainly absorbs on the lower polariton since the system is almost decoupled at this coupling strength and picks up the poles of the decoupled field. Similarly, the mixed spectrum (based on $\chi_{\sigma q}^{\sigma x}$ or $\chi_{\sigma q}^{\sigma p}$) in Fig.\ref{fig:2}c provides us with information of how strong one subsystem of the coupled system reacts upon perturbing the other one. The oscillator strength here is not necessarily positive. What is absorbed by one subsystem can be transferred to the other and is then emitted. We note at this point that the peaks in Fig.\ref{fig:2} are artificially broadened and in reality correspond to sharp transitions due to excited states with infinite lifetimes. How to get lifetimes will be discussed in the next section.

\section{IV. Coupled Matter-Photon Response: Real Systems}

In this section we finally apply the introduced formalism in pRPA approximation to a real system. We consider a benzene molecule in an optical cavity. In Fig.\ref{fig:3} we schematically depict the experimental setup for an absorption experiment under strong light-matter coupling. First we study the prototypical cavity QED set up where the molecule is strongly coupled to a single cavity mode of a high-Q cavity. In a second setup we lift the restriction of only one mode and instead couple the benzene molecule to many modes that sample the electromagnetic vacuum field without enhancing the coupling to a specific mode by hand. While the first setup provides us with the first ab-initio calculation for the spectrum of a real molecule in a high-Q cavity, the second setup allows us to determine the intrinsic lifetimes of excited states from first principles. These two examples highlight the novel possibilities and perspectives that the QEDFT framework provides.

\subsection{A. Numerical details}

We start by discussing the general setup before considering the specialized situations discussed above. We have implemented the linear-response eigenvalue equation into the real-space code OCTOPUS \cite{61,62}. The absorption spectrum of the Benzene molecule has been very successfully studied with TDDFT calculations \cite{60,62}. Small organic molecules and benzene in particular are rewarding systems to be studied with TDDFT, since the adiabatic approximation in concert with the local density approximation \cite{63,64} capture the occurring $\Pi-\Pi^*$ transition at around 7eV in the benzene molecule exceptionally well. This transition is a characteristic of carbon conjugate compounds \cite{63}. Regarding the specific setups of electronic structure calculations of the systems, we follow closely the setup of Ref. \cite{60}. Thus, we use a cylindrical real space grid of 8 Å length with the radius of 6 Å. For the benzene nuclear structure, we use the CC bond length of 1.396 Å, and CH bond length of 1.083 Å. We explicitly describe only the 30 valence electrons, while the core atoms are described by the LDA Troullier-Martins pseudopotentials \cite{65} and include 50 unoccupied states in the eigenvalue calculation. Solving the linear-response eigenvalue problem of Eq.\ref{eq:42} provides us with transition amplitudes, as well as the excitation energies. To obtain the spectra usually these peaks are broadened. To do so, we use the standard implementation of the octopus code that uses a Lorentzian broadening of the following form

$$
\Gamma(\omega, E_i) = \frac{1}{\pi} \frac{\Delta}{(\omega - E_i)^2 + \Delta^2}.
$$

**(FIG. 3. Schematic of absorption spectroscopy of optical cavities:** Benzene (C₆H₆) molecule and $\lambda_o$ denotes the polarization direction of the photon field.)
The actual absorption crossection is the obtained by

$$\sigma(\omega) = \sum_i f_i \Gamma(\omega, E_i),$$

(83)

where $f_i$ are the oscillator strength defined in Eq. 53.

### B. Strong light-matter coupling

![Absorption Spectra (Benzene)](image)

**FIG. 4.** Absorption crossection for benzene molecule in free space (black) and under strong light-matter coupling in an optical cavity to ultra-strong coupling (blue). The value for lambda is given in units of [eV$^{1/2}$/nm].

The first results we discuss are due to a coupling to a single mode of a high-Q cavity. Here we included a single cavity mode in resonance to the $\Pi-\Pi^*$ transition, i.e., $\omega_0 = 7.11$ eV. For the light-matter coupling strength $\lambda_0$, we choose five different values, i.e. $\lambda_0 = (0, 2.73, 5.45, 8.18, 10.91)$ eV$^{1/2}$/nm. In Fig. 4 we show the absorption spectra for these different values. We start by discussing the $\lambda_0 = 0$ case. The spectrum is shown in black. This spectrum corresponds to a calculation of the benzene molecule in free space and the spectrum is within the numerical capabilities identical to Ref. 60. We tune the electron-photon coupling strength $\lambda_0$ in Fig. 4. We find for increasing coupling strength the splitting of the HOMO-LUMO peak into two polaritonic branches. The lower polaritonic branch has higher intensity, compared to the upper polaritonic peak.

### C. Lifetimes of excitations from first principles

Next we consider how we can obtain lifetimes from QEDFT linear-response theory. We explicitly couple the molecule to a wide range of photon modes similar as in the spontaneous emission calculation of Ref. 28. While in Ref. 28, we have sampled the system with 200 photon modes, we choose here 3400 photon modes. The energies of the sampled photon modes cover rather densely a range from 4.7 meV, for the smallest energy up to 30.5 eV for the largest one. However, we do not sample the full three-dimensional mode space together with the two polarization possibilities per mode but rather consider a one-dimensional slice in mode space. This one-dimensional sampling of mode frequencies will change the actual three-dimensional lifetimes, but for demonstrating the possibilities of obtaining lifetimes this is sufficient. The results of this calculation are shown in Fig. 5. In Fig. 5a we show the full spectrum. In grey we plot the standard linear response calculation that is identical to the black spectrum shown in Fig. 4. This spectrum has been broadened by a Lorentzian function as given in Eq. (82). The electron-photon absorption function is plotted in blue. Since we have sampled the photon part densely, we do not need to artificially broaden the peaks anymore. Formulated differently, we can directly plot the oscillator strength and the excitation energies of the equation. Connecting to the eigenstates $E_1$ and $P_1$ of Eq. (72) we can measure the photonic and electronic nature of the individual poles of the response function...
The sum of both is normalized to one, i.e. \( \sigma_c + \sigma_p = 1 \). This quantity is color coded in Fig. 3 from blue (more photonic) to red (more electronic). In (b) we zoom to the II-II\(^+\) transition. Due to one-dimensional nature of the quantization volume, we find a broadening of the peak that is larger than it is for the case of a three-dimensional cavity due to the sampling of the electromagnetic vacuum. This is similar to changing the vacuum of the electromagnetic field. Accordingly the lifetimes of the electronic states are shorter if the electromagnetic field is confined to one dimension. In Fig. 5c we finally show the ab-initio peak of the \( \sigma - \sigma^\dagger \) transition. Since no broadening has been used in the coupled photon-matter response calculation, the lifetimes can be inferred directly from the width of the peaks.

V. SUMMARY AND OUTLOOK

In this work we have introduced linear-response theory for non-relativistic quantum-electrodynamics in the long wavelength limit. Compared to the conventional matter-only response approaches, we have highlighted how in the coupled matter-photon case the usual response functions change, how novel photon-photon and matter-photon response functions are introduced and how we can efficiently calculate these response functions in the framework of QEDFT. By investigating first a simple model system, we have shown how the spectrum of the matter subsystem is changed upon coupling to the photon field and also that the Maxwell equations of the photonic subsystem become modified. Further we have demonstrated the range of validity of a simple yet reliable approximation to the in general unknown mean-field exchange-correlation kernels. Using this approximation we have presented the first ab-initio calculations of the spectrum of a real system (benzene) coupled to the modes of the quantized electromagnetic field. In one example we have calculated the change upon strong coupling to a single mode of a high-Q cavity, which leads to a large Rabi splitting. In the second example we have calculated from first principles the natural linewidths of benzene coupled to a specific sampling of the vacuum field. These results demonstrate the versatility and possibilities of QEDFT, where light and matter are treated on equal quantized footing. In the context of strong light-matter coupling, e.g., in polaritonic chemistry, the presented linear-response formulation allows now to determine polaritically modified spectra from first principles. Together with ab-initio ground-state calculations [27] QEDFT now provides a workable first-principle description to analyze and predict photon-dressed chemistry and material sciences. Further, in the context of standard ab-initio theory, the linear-response formulation of QEDFT now allows the calculation of intrinsic lifetimes. For optical physics, the presented linear-response framework presents an interesting opportunity to study the modifications of the Maxwell equations in matter from first principles. Finally we want to highlight that although the QEDFT linear-response framework is new, its similarity to the usual matter-only linear-response formulation in terms of a pseudo-eigenvalue problem makes it very easy to include in already existing first-principle codes. This, together with the above discussed novel possibilities in different fields of physics, shows that there are many interesting cases that can be studied with the presented method.

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Appendix A: General linear response for coupled matter-photon system

The static and dynamical behavior of the coupled electron-photon systems is given by

\[
\hat{H}(t) = \hat{H}_0 + \hat{H}_{ext}(t).
\]

Here, the time-independent electron-photon Hamiltonian is

\[
\hat{H}_0 = \hat{T} + \hat{W}_{ee} + \frac{1}{2} \sum_{\alpha=1}^{M} \left[ p_\alpha^2 + \omega_\alpha^2 \left( \hat{q}_\alpha - \frac{\lambda_\alpha}{\omega_\alpha} \mathbf{R} \right)^2 \right] + \sum_{i=1}^{N} v_0(\mathbf{r}_i) + \sum_{\alpha=1}^{M} J_{\alpha 0} \hat{q}_\alpha,
\]

where kinetic energy operator is \( \hat{T} = -\frac{\hbar^2}{2m_\epsilon} \sum_{i=1}^{N} \nabla_\mathbf{r}_i^2 \), the Coulomb potential is \( \hat{W}_{ee} = \frac{e^2}{4\pi\epsilon_0} \sum_{i<j}^{N} \frac{1}{|\mathbf{r}_i-\mathbf{r}_j|} \) and time-dependent external perturbation is given by

\[
\hat{H}_{ext}(t) = \hat{V}_{ext}(t) + \hat{J}_{ext}(t).
\]

Here, the time-dependent external potential and current are

\[
\hat{V}_{ext}(t) = \sum_{i=1}^{N} v(\mathbf{r}_i, t), \quad \hat{J}_{ext}(t) = \sum_{\alpha} \frac{J_{\alpha 0}(t)}{\omega_\alpha} \hat{q}_\alpha.
\]

The state vector of the interacting electron-photon system is

\[
\Psi_I(t) = \hat{T}_0^{-1}(t) \Psi(t) = e^{i\hat{H}_0 t/\hbar} \Psi(t),
\]
where $\Psi_I(t)$ is the state vector in the interaction picture and $\Psi(t)$ is the state vector in the Schrödinger picture. The evolution of the interacting electron-photon system is described in the interaction picture as

$$i\hbar \frac{\partial}{\partial t} \Psi_I(t) = e^{i\hat{H}_0 t/\hbar} \hat{H}_{ext}(t) e^{-i\hat{H}_0 t/\hbar} \Psi_I(t). \quad (A5)$$

The equation is integrated to obtain the so-called mild form in the interaction picture given by

$$\Psi_I(t) = \Psi_0 - \frac{i}{\hbar} \int_{t_0}^t dt' \hat{H}_{ext,I}(t') \Psi_I(t'). \quad (A6)$$

The first-order solution is given as

$$\Psi(t) \simeq \hat{U}_0 \Psi_0 - \frac{i}{\hbar} \hat{U}_0(t) \int_{t_0}^t dt' \hat{H}_{ext,I}(t') \Psi_0. \quad (A7)$$

The change in the expectation value of an arbitrary observable $\hat{O}$ due to the external perturbation $\hat{H}_{ext}(t)$ is

$$\delta \langle \hat{O}(t) \rangle = \langle \Psi(t) | \hat{O} | \Psi(t) \rangle - \langle \Psi_0 | \hat{O} | \Psi_0 \rangle, \quad (A8)$$

such that the linear response is

$$\delta \langle \hat{O}(t) \rangle = -\frac{i}{\hbar} \int_{t_0}^t dt' \langle \Psi_0 | \left[ \hat{O}_I(t), \hat{H}_{ext,I}(t') \right] | \Psi_0 \rangle, \quad (A9)$$

where $\hat{O}_I(t)$ is the operator in the interaction picture. Now, the response of the electron density to $\hat{H}_{ext}(t)$ is

$$\delta n(r,t) = -\frac{i}{\hbar} \int_{t_0}^t dt' \int d^3r' \langle \Psi_0 | [\hat{n}_I(r,t), \hat{n}_I(r',t')] | \Psi_0 \rangle \delta v(r')$$

$$-\frac{i}{\hbar} \sum_{\alpha} \int_{t_0}^t dt' \langle \Psi_0 | [\hat{n}_I(r,t), \hat{q}_{\alpha,I}(t')] | \Psi_0 \rangle \frac{\delta j_{\alpha}(t')}{\omega_{\alpha}}.$$  

The response of the density is given as

$$\delta n(r,t) = \int_{t_0}^\infty dt' \int d^3r' \chi_n^0(r,t,r',t') \delta v(r')$$

$$+ \sum_{\alpha} \int_{t_0}^\infty dt' \chi_{\alpha,I}^0(r,t,t') \frac{\delta j_{\alpha}(t')}{\omega_{\alpha}},$$

where the response functions are

$$\chi_n^0(r,t,r',t') = -\frac{i}{\hbar} \Theta(t-t') \langle \Psi_0 | [\hat{n}_I(r,t), \hat{n}_I(r',t')] | \Psi_0 \rangle, \quad (A10)$$

$$\chi_{\alpha,I}^0(r,t,t') = -\frac{i}{\hbar} \Theta(t-t') \langle \Psi_0 | [\hat{n}_I(r,t), \hat{q}_{\alpha,I}(t')] | \Psi_0 \rangle. \quad (A11)$$

Similarly, the response of the photon coordinate $q_{\alpha}(t)$ to $\hat{H}_{ext}(t)$ is

$$\delta q_{\alpha}(t) = -\frac{i}{\hbar} \int_{t_0}^t dt' \langle \Psi_0 | [q_{\alpha,I}(t), \hat{V}_{ext,I}(t')] | \Psi_0 \rangle$$

$$-\frac{i}{\hbar} \int_{t_0}^t dt' \langle \Psi_0 | [q_{\alpha,I}(t), \hat{J}_{ext,I}(t')] | \Psi_0 \rangle.$$

Following similar steps as above, the response is given by

$$\delta q_{\alpha,I}(t,t') = \int_{t_0}^\infty dt' \int d^3r' \chi_{\alpha,I}^0(r,t,r',t') \delta v(r')$$

$$+ \sum_{\alpha'} \int_{t_0}^\infty dt' \chi_{\alpha',I}^0(r,t,t') \frac{\delta j_{\alpha'}(t')}{\omega_{\alpha'}},$$

where the response functions are

$$\chi_{\alpha,I}^0(r,t,r',t') = -\frac{i}{\hbar} \Theta(t-t') \langle \Psi_0 | [q_{\alpha,I}(t), \hat{v}(r',t')] | \Psi_0 \rangle, \quad (A12)$$

$$\chi_{\alpha',I}^0(r,t,t') = -\frac{i}{\hbar} \Theta(t-t') \langle \Psi_0 | [q_{\alpha,I}(t), \hat{q}_{\alpha',I}(t')] | \Psi_0 \rangle. \quad (A13)$$

Alternatively, the response functions of Eqs. (A10)-(A13) can be obtained using the functional dependence of the observables on the external pair $(v(r,t), j_{\alpha}(t))$. The wave function of Eq. (3) has a functional dependence $\Psi([v,j_{\alpha}];t)$ via the Hamiltonian Eq. (A1), i.e., $\hat{H}(t) = H([v,j_{\alpha}];t)$. Therefore, through the expectation of electron density and photon displacement coordinate, both have a functional dependence on the external pair as $n([v,j_{\alpha}];rt)$ and $q_{\alpha}([v,j_{\alpha}];t)$, respectively.

Considering the ground-state problem with external potential and current of $(v(0), j_{\alpha,0})$, we can perform a functional Taylor expansion of the density $n(rt)$ and photon coordinate $q_{\alpha}(t)$ to first-order as

$$\delta n([v,j_{\alpha}];rt) = \int d^3r' \chi_n^0(r,t,r',t') \delta v(r')$$

$$+ \sum_{\alpha} \int dt' \chi_{\alpha,I}^0(r,t,t') \delta j_{\alpha}(t'),$$

This reduces to the response of the electron density and photon coordinate given as

$$\delta n([v,j_{\alpha}];rt) = \int d^3r' \chi_n^0(r,t,r',t') \delta v(r')$$

$$+ \sum_{\alpha} \int dt' \chi_{\alpha,I}^0(r,t,t') \delta j_{\alpha}(t').$$
and
\[ \delta q_{\alpha}(\{v, j_{\alpha}; t\}) = \int dr' dt' \chi_{\alpha}^{\prime q_{\alpha}}(r, t'; r') \delta v(r') + \sum_{\alpha'} \int dt' \chi_{j_{\alpha'}(t')}^{\prime q_{\alpha}} \delta j_{\alpha'}(t'), \]
where we define the response functions of the above relation as
\[ \chi_{\alpha}^{\prime q_{\alpha}}(r, t'; r') = \frac{\delta q_{\alpha}(\{v, j_{\alpha}; t\}|r')}{\delta v(r')} \bigg|_{v_{0}(r), j_{\alpha,0}}, \quad (A14) \]
\[ \chi_{j_{\alpha}}^{\prime q_{\alpha}}(r, t', t') = \frac{\delta q_{\alpha}(\{v, j_{\alpha}; t\})}{\delta j_{\alpha}(t')} \bigg|_{v_{0}(r), j_{\alpha,0}}, \quad (A15) \]
\[ \chi_{\alpha}^{q_{\alpha}}(r, t'; t') = \frac{\delta q_{\alpha}(\{v, j_{\alpha}; t\})}{\delta v(t')} \bigg|_{v_{0}(r), j_{\alpha,0}}, \quad (A16) \]
\[ \chi_{j_{\alpha}}^{q_{\alpha}}(r, t, t') = \frac{\delta q_{\alpha}(\{v, j_{\alpha}; t\})}{\delta j_{\alpha}(t')} \bigg|_{v_{0}(r), j_{\alpha,0}}. \quad (A17) \]
These response functions defined in Eqs. (A10)-(A13) and Eqs. (A14)-(A17) are equivalent.

The response functions expressed in the so-called Lehmann representation is given by
\[ \chi_{\alpha}^{n}(r, r', \omega) = \frac{1}{\hbar} \lim_{\eta \to 0} \sum_{k} \left[ \frac{f_{k}(r) f_{k}^{\dagger}(r')}{\omega - \Omega_{k} + i\eta} - \frac{f_{k}(r') f_{k}^{\dagger}(r)}{\omega + \Omega_{k} + i\eta} \right], \]
\[ \chi_{\alpha}^{q}(r, \omega) = \frac{1}{\hbar} \lim_{\eta \to 0} \sum_{k} \left[ \frac{f_{k}(r) g_{\alpha,k}^{*}}{\omega - \Omega_{k} + i\eta} - \frac{g_{\alpha,k} f_{k}^{\dagger}(r)}{\omega + \Omega_{k} + i\eta} \right], \]
\[ \chi_{\alpha}^{\prime q}(r', \omega) = \frac{1}{\hbar} \lim_{\eta \to 0} \sum_{k} \left[ \frac{g_{\alpha,k} f_{k}^{\dagger}(r')}{\omega - \Omega_{k} + i\eta} - \frac{f_{k}(r') g_{\alpha,k}^{*}}{\omega + \Omega_{k} + i\eta} \right], \]
\[ \chi_{\alpha}^{q}(\omega) = \frac{1}{\hbar} \lim_{\eta \to 0} \sum_{k} \left[ \frac{g_{\alpha,k} g_{\alpha,k}^{*}}{\omega - \Omega_{k} + i\eta} - \frac{g_{\alpha,k}^{*} g_{\alpha,k}}{\omega + \Omega_{k} + i\eta} \right], \]
where \( f_{k}(r) = \langle \Psi_{0}|\hat{n}(r)|\Psi_{k}\rangle \) and \( g_{\alpha,k} = \langle \Psi_{0}|\hat{q}_{\alpha}|\Psi_{k}\rangle \) are the transition matrix elements and \( |\Psi_{0}\rangle \) is the correlated electron-photon ground state wave function. The excitation energies \( \Omega_{k} = (E_{k} - E_{0})/\hbar \) of the finite interacting system are the poles of the response functions of the unperturbed system.

**Appendix B: linear-response within QEDFT**

In this section, we present linear-response in QEDFT by employing the maps between interacting and non-interacting system, we express the interacting response functions in terms of two non-interacting response functions and exchange correlation kernels. The responses due to \( (v(r), j_{\alpha}(t)) \) are evaluated at the ground-state \( (v_{0}(r), j_{\alpha,0}) \) and will not be written explicitly.

The non-interacting subsystems moving in an effective potential and current \( (v_{s}(r), j_{\alpha}^{s}(t)) \) can be written as a time-dependent problem of the Schrödinger equation
\[ i\hbar \frac{\partial}{\partial t} \Phi(t) = \hat{H}(t) \Phi(t). \quad (B1) \]
Here, the non-interacting effective Hamiltonian \( \hat{H}(t) = \hat{H}_{s,0} + \hat{H}_{ext,s}(t) \) that is meant to reproduces the exact density and displacement field is given explicitly as
\[ \hat{H}_{s,0} = \hat{\Phi} + \hat{H}_{pl} + (v_{0}(r) + v_{0,Mxc}, [n, q_{\alpha}]) \hat{q}_{\alpha}, \]
and the time-dependent part is
\[ \hat{H}_{ext,s}(t) = (v(r) + v_{Mxc}, [n, q_{\alpha}]) \hat{q}_{\alpha}, \]
where \( \hat{H}_{pl} = \frac{1}{2} \sum_{\alpha=1}^{M} [\hat{p}_{\alpha}^{2} + \omega_{\alpha}^{2} \hat{q}_{\alpha}^{2}] \) is the oscillator for the photon mode and the mean-field xc potential and current are defined as
\[ v_{Mxc}, [n, q_{\alpha}]; r] := v_{s}([n]; r) - v([n], q_{\alpha}; r), \quad (B2) \]
\[ j_{\alpha,Mxc}, [n, q_{\alpha}]; t] := j_{\alpha,s}([q_{\alpha}; t] - j_{\alpha}([n, q_{\alpha}]; t), \quad (B3) \]
In the above definitions of \( v_{Mxc}, [n, q_{\alpha}]; r \) and \( j_{\alpha,Mxc}, [n, q_{\alpha}]; t \), the initial state dependence of the interacting \( \Psi_{0} \) and non-interacting \( \Phi_{0} \) system was dropped out. For completeness, the definition of \( j_{\alpha,Mxc}, [n, q_{\alpha}]; t \) accounts for a functional dependence on \( q_{\alpha} \) but this term can be calculated explicitly since it has no xc part as seen in Eq. (10). The simplified form of \( j_{\alpha,Mxc} \) is expressed in Eq. (8).

Through similar steps as in Eqs. (A5)-(A7), in first-order of the Schrödinger-Kohn-Sham equation
\[ \Phi(t) \simeq \hat{U}_{0} \Phi_{0} - \frac{i}{\hbar} \hat{U}_{0}(t) \int_{t_{0}}^{t} dt' \hat{H}_{ext,s},,\langle(t')|\Phi_{0} \rangle, \quad (B4) \]
where \( \hat{U}_{0} = e^{-iH_{s,0}t/\hbar} \). Next, the bijective mapping between the interacting and non-interacting system that yields the same density and photon coordinate is given as
\[ (v(r), j_{\alpha}(t)) \leftrightarrow^{1/\hat{q}_{\alpha}} (n(r), q_{\alpha}(t)) \leftrightarrow^{1/\hat{q}_{\alpha}} (v_{s}(r), j_{\alpha}^{s}(t)), \quad (B5) \]
which can be inverted as \( (v_{s}([v, j_{\alpha}]; r'), j_{\alpha}^{s}([v, j_{\alpha}]; t')). \)

The response of the electronic subsystem due to the perturbations with the external pair \( (v(r), j_{\alpha}(t)) \) is
\[ \delta v_{\alpha}(r) = -\frac{i}{\hbar} \int_{t_{0}}^{t} dt \int dx \langle \Phi_{0} | \hat{n}_{\alpha}(r, t') \hat{n}_{\alpha}(x, t') \rangle |\Phi_{0}\rangle \times \int dt' dt' \frac{\delta v_{\alpha}(\{v, j_{\alpha}; x\})}{\delta v(r')} \delta v_{\alpha}(r'), \]
\[ -\frac{i}{\hbar} \int_{t_{0}}^{t} dt \int dx \langle \Phi_{0} | \hat{n}_{\alpha}(r, t') \hat{n}_{\alpha}(x, t') \rangle |\Phi_{0}\rangle \times \sum_{\alpha} \int dt' \frac{\delta v_{\alpha}(\{v, j_{\alpha}; x\})}{\delta j_{\alpha}(t')} \delta j_{\alpha}(t'). \]

Where \( |\hat{n}_{\alpha}(r, t)|^{2} = 0 \) since both, electronic and photonic subsystems, are independent in the
non-interacting system. From Eq. (B3), we have \((v_n([n];rt), j_n([q];t))\) such that the above equation becomes

\[
\delta n(rt) = \int dt dx \chi_{n,s}^n(rt, x) \\
\times \int d\tau d\tau' d\tau'' \frac{\delta v_n([n]; \tau x) \delta n([v, j]_\alpha; \tau' y)}{\delta v(r')} \delta v(r') \\
+ \int dt dx \chi_{n,s}^n(rt, x) \\
\times \sum \int dt' d\tau' d\tau'' \frac{\delta v_n([n]; \tau x) \delta n([v, j]_\alpha; \tau' y)}{\delta j_\alpha(t')} \delta j_\alpha(t'), 
\]

(B6)

where \(\chi_{n,s}^n(rt, x) = \langle \Psi_0 | \hat{n}_I(rt) \hat{n}_I(x) | \Psi_0 \rangle \). For clarity, the above density response is \(\delta n(rt) = \delta n_u(rt) + \delta n_j(rt)\), where \((\delta n_u(rt), \delta n_j(rt))\) is the density response to the external pair \((v(rt), j_\alpha(t))\), respectively.

Using Eqs. (B2) and (B3), we define the mean-field xc kernels as:

\[
f_{M xc}^n([n, q]; rt, r't') = \frac{\delta v_n([n]; rt)}{\delta n(rt')} - \frac{\delta v([n, q]; rt)}{\delta n(rt')},
\]
(B7)

\[
f_{M xc}'^n([n, q]; rt, r') = \frac{\delta v([n, q]; rt)}{\delta q_\alpha(t')},
\]
(B8)

\[
g_{M xc}^n([n, q]; r, r't') = -\frac{\delta j_\alpha([n, q]; t)}{\delta n(rt')} \frac{\delta j_\alpha([n, q]; t)}{\delta q_\alpha(t')},
\]
(B9)

\[
g_{M xc}'^n([n, q]; r, r') = \frac{\delta j_\alpha([n, q]; t)}{\delta q_\alpha(t')},
\]
(B10)

where \(\frac{\delta v([n, q]; rt)}{\delta n(rt')} = 0 = \frac{\delta v([n, q]; rt)}{\delta q_\alpha(t')}\). These kernels are basically inverses of the interacting and non-interacting response functions.

From Eq. (B6), density response to \(\delta v\) can be written in terms of the density-density response function given by

\[
\chi_{n,s}^n(rt, r't') = \int dt dx \chi_{n,s}^n(rt, x) \\
\times \int d\tau d\tau' d\tau'' \frac{\delta v_n([n]; \tau x) \delta n([v, j]_\alpha; \tau' y)}{\delta v(r')} \delta v(r') \\
+ \int dt dx \chi_{n,s}^n(rt, x) \\
\times \int d\tau' d\tau'' \frac{\delta v([n, q]; \tau x) \delta n([v, j]_\alpha; \tau' y)}{\delta j_\alpha(t')} \delta j_\alpha(t').
\]

Making the following substitution in the above equation

\[
\int d\tau' d\tau'' \frac{\delta v([n, q]; \tau x) \delta n([v, j]_\alpha; \tau' y)}{\delta v(r')} \delta v(r') = \delta (x - r') \delta (\tau - t')
\]

where \(v([n, q]; \tau x) / \delta v(r') = \delta(x - r') \delta(\tau - t')\), we obtain the relation

\[
\chi_{n,s}^n(rt, r't') = \chi_{n,s}^n(rt, r't') + \int d\tau d\tau' d\tau'' \frac{\delta n([v, j]_\alpha; \tau' y)}{\delta j_\alpha(t')} \delta j_\alpha(t') \\
\times f_{M xc}^n(x, y) \chi_{n,s}^n(y, r't') \\
+ \sum_\alpha \int d\tau d\tau' \chi_{n,s}^n(rt, x) \\
\times f_{M xc}^n(x, y') \chi_{n,s}^n(y', r't'). 
\]

(B11)

Next, the density response to \(\delta j_\alpha\) in Eq. (B6) is expressed in terms of the response function as

\[
\chi_{\alpha}^n(rt, r't') = \int dt dx \chi_{\alpha}^n(rt, x) \\
\times \int d\tau d\tau' d\tau'' \frac{\delta n([v, j]_\alpha; \tau' y)}{\delta j_\alpha(t')} \delta j_\alpha(t') \\
\times f_{M xc}^n(x, y) \chi_{\alpha}^n(y, r't') \\
+ \sum_\alpha \int d\tau d\tau' \chi_{\alpha}^n(rt, x) \\
\times f_{M xc}^n(x, y') \chi_{\alpha}^n(y', r't').
\]

Using the relation (obtained from \(\delta v([n, q]; x) / \delta j_\alpha(t')\))

\[
\int d\tau d\tau' \frac{\delta v([n, q]; x) \delta n([v, j]_\alpha; y)}{\delta j_\alpha(t')} \frac{\delta n([v, j]_\alpha; y')}{\delta j_\alpha(t')} = -\sum_\alpha \int d\tau' \frac{\delta v([n, q]; x) \delta n([v, j]_\alpha; y')}{\delta j_\alpha(t')} \frac{\delta n([v, j]_\alpha; y)}{\delta j_\alpha(t')},
\]

the response function is given as

\[
\chi_{\alpha}^n(rt, r't') = \int d\tau d\tau' d\tau'' \frac{\delta n([v, j]_\alpha; x) \delta n([v, j]_\alpha; y)}{\delta j_\alpha(t')} \delta j_\alpha(t') \\
\times f_{M xc}^n(x, y) \chi_{\alpha}^n(y, r't') \\
+ \sum_\alpha \int d\tau d\tau' \chi_{\alpha}^n(rt, x) \chi_{\alpha}^n(x, y') \chi_{\alpha}^n(y', r't'). 
\]

(B12)

Similarly, the response to the photonic subsystem to linear perturbations from the external pair \((v(rt), j_\alpha(t))\) is

\[
\delta q_\alpha(t) = -\frac{i}{\hbar} \sum_\beta \int_{t_0}^t dt \langle \Psi_0 | [q_\beta, j_\beta(t)] | \Psi_0 \rangle \\
\times \int d\tau d\tau' \frac{\delta j_\beta([v, j]_\alpha; \tau)}{\delta v(r')} \delta v(r') \\
- \frac{i}{\hbar} \sum_\beta \int_{t_0}^t dt \langle \Psi_0 | [q_\beta, j_\beta(t)] | \Psi_0 \rangle \\
\times \int d\tau' d\tau'' \frac{\delta j_\beta([v, j]_\alpha; \tau)}{\delta j_\alpha(t')} \delta j_\alpha(t'),
\]

where \(\langle \hat{q}_\beta, j_\beta(t) \rangle = 0\) in the non-interacting system. By defining the non-interacting response function of the photon mode as \(\chi_{q_\beta}^n(t, \tau) = (-i/\hbar)\Theta(t - \tau)\langle \Psi_0 | [q_\beta, j_\beta(t)] | \Psi_0 \rangle\). Using Eq. (B3), we get

\[
\chi_{q_\beta}^n(t, \tau) = \chi_{q_\beta}^n(t, \tau') + \int \int \int d\tau d\tau' d\tau'' \frac{\delta n([v, j]_\alpha; \tau' y)}{\delta j_\alpha(t')} \delta j_\alpha(t') \\
\times f_{M xc}^n(x, y') \chi_{q_\beta}^n(y', \tau') \\
+ \sum_\alpha \int \int d\tau d\tau' \chi_{q_\beta}^n(rt, x) \chi_{q_\beta}^n(x, y') \\
\times f_{M xc}^n(x, y') \chi_{q_\beta}^n(y', \tau').
\]

(B13)
have \((v_i([n];rt),j^*_{\alpha}([q_{\alpha}];t))\), the response can be written as

\[
\delta q_{\alpha}(t) = \sum_{\beta} \int dt' \chi^{q_{\beta}}_{q_{\alpha},\beta}(t, t') \sum_{\beta'} \int dt'' \int dt''' \frac{\delta j_{\beta'\alpha}([n,q_{\alpha}];\tau)}{\delta q_{\beta'}(\tau')} \delta v(r'') \delta v(r''') \\
\times \frac{\delta j_{\beta}(n, q_{\alpha}; \tau)}{\delta q_{\beta}(\tau)} \delta q_{\beta'}([v, j_{\alpha}]; \tau') \delta v(r') \delta v(r''),
\]

The above response of the displacement field is \(\delta q_{\alpha}(t) = \delta q_{\alpha,v}(t) + \delta q_{\alpha,s}(t)\), where \(\delta q_{\alpha,v}(t)\), \(\delta q_{\alpha,s}(t)\) is the response to the external pair \((v(rt), j_{\alpha}(t))\), respectively.

From Eq. (B13), the field response to \(\delta v\) can be written in terms of the photon-density response function as

\[
\chi^{q_{\alpha},v}_{\alpha}(t, r', t'') = \sum_{\beta} \int dt' \chi^{q_{\alpha},v}_{q_{\beta},\alpha}(t, t') \sum_{\beta'} \int dt'' \int dt''' \frac{\delta j_{\beta'\alpha}([n,q_{\alpha}];\tau)}{\delta q_{\beta'}(\tau')} \delta v(r'') \delta v(r''') \\
\times \frac{\delta j_{\beta}(n, q_{\alpha}; \tau)}{\delta q_{\beta}(\tau)} \delta q_{\beta'}([v, j_{\alpha}]; \tau') \delta v(r') \delta v(r''),
\]

The above response of the displacement field is \(\delta q_{\alpha}(t) = \delta q_{\alpha,v}(t) + \delta q_{\alpha,s}(t)\), where \(\delta q_{\alpha,v}(t)\), \(\delta q_{\alpha,s}(t)\) is the response to the external pair \((v(rt), j_{\alpha}(t))\), respectively.

From Eq. (B13), the field response to \(\delta v\) can be written in terms of the photon-density response function as

\[
\chi^{q_{\alpha},v}_{\alpha}(t, r', t'') = \sum_{\beta} \int dt' \chi^{q_{\alpha},v}_{q_{\beta},\alpha}(t, t') \sum_{\beta'} \int dt'' \int dt''' \frac{\delta j_{\beta'\alpha}([n,q_{\alpha}];\tau)}{\delta q_{\beta'}(\tau')} \delta v(r'') \delta v(r''') \\
\times \frac{\delta j_{\beta}(n, q_{\alpha}; \tau)}{\delta q_{\beta}(\tau)} \delta q_{\beta'}([v, j_{\alpha}]; \tau') \delta v(r') \delta v(r''),
\]

Using the relation (obtained from \(\delta j_{\beta}(n, q_{\alpha}; \tau)/\delta v(r'')\))

\[
\sum_{\beta'} \int dt' \frac{\delta j_{\beta}(n, q_{\alpha}; \tau)}{\delta q_{\beta}(\tau')} \delta v(r') = - \int dt' \frac{\delta j_{\beta}(n, q_{\alpha}; \tau)}{\delta v(r')} \delta n(y') \delta v(r'),
\]

the response function is given as

\[
\chi^{q_{\alpha},v}_{\alpha}(t, r', t'') = \sum_{\beta} \int dt' \chi^{q_{\beta}}_{q_{\alpha},\beta}(t, t') \sum_{\beta'} \int dt'' \int dt''' \frac{\delta j_{\beta'}([n,q_{\alpha}];\tau)}{\delta q_{\beta'}(\tau')} \delta v(r'') \delta v(r''') \\
\times \frac{\delta j_{\beta}(n, q_{\alpha}; \tau)}{\delta q_{\beta}(\tau)} \delta q_{\beta'}([v, j_{\alpha}]; \tau') \delta v(r') \delta v(r''),
\]

where \(g^{n_{\beta}}_{Mxc} = g^{n_{\beta}}_{M}\) and \(g^{n_{\beta}}_{Mxc} = 0\) as determined from the equation of motion for the displacement field. Also, from Eq. (B13), field response to \(\delta j_{\alpha}\) can be written in terms of the photon-photon response function as

\[
\chi^{q_{\alpha},s}_{\alpha}(t, t'') = \sum_{\beta} \int dt' \chi^{q_{\beta}}_{q_{\alpha},\beta}(t, t') \sum_{\beta'} \int dt'' \int dt''' \frac{\delta j_{\beta'\alpha}([n,q_{\alpha}];\tau)}{\delta q_{\beta'}(\tau')} \delta v(r'') \delta v(r''') \\
\times \frac{\delta j_{\beta}(n, q_{\alpha}; \tau)}{\delta q_{\beta}(\tau)} \delta q_{\beta'}([v, j_{\alpha}]; \tau') \delta v(r') \delta v(r''),
\]

Making the following substitution (where \(j_{\beta}(n, q_{\alpha}; \tau)/j_{\alpha'}(t') = \delta(\tau - t')\delta_{\beta,\alpha'}\)) in the above equation

\[
\sum_{\beta} \int dt' \frac{\delta j_{\beta}([n,q_{\alpha}];\tau)}{\delta q_{\beta}(\tau')} \delta q_{\beta'}([v, j_{\alpha}]; \tau') \delta v(r') \delta v(r'') \\
\times \frac{\delta j_{\beta}(n, q_{\alpha}; \tau)}{\delta q_{\beta}(\tau)} \delta q_{\beta'}([v, j_{\alpha}]; \tau') \delta v(r') \delta v(r''),
\]

yields the photon-photon response function

\[
\chi^{q_{\alpha},s}_{\alpha}(t, t'') = \chi^{q_{\alpha}}_{q_{\alpha},\alpha}(t, t'') \\
+ \sum_{\beta} \int dt' \int dt'' \int dt''' \frac{\delta j_{\beta}([n,q_{\alpha}];\tau)}{\delta q_{\beta}(\tau')} \delta q_{\beta'}([v, j_{\alpha}]; \tau') \delta v(r') \delta v(r'') \\
\times \chi^{q_{\beta}}_{q_{\beta},\alpha}(\tau', x'; t''),
\]

where \(\chi^{q_{\alpha}}_{q_{\alpha},\alpha}(t, t'') = 0\).

**Appendix C: Matrix formulation of QEDFT response equations**

Through a Fourier transform of Eqs. (B11)-(B12) and Eqs. (B14)-(B15) and making a substitution into Eqs. (34)-(37), we express the responses in the following form:

\[
\delta n_{\alpha}(r, \omega) = \sum_{i,a} \left[ \varphi_{\alpha}(r) \varphi^*_i(r) P^{(1)}_{ai}(\omega) + \varphi_i(r) \varphi^*_\alpha(r) P^{(1)}_{ia}(\omega) \right],
\]

\[
\delta n_{\alpha}(r, \omega) = \sum_{i,a} \left[ \varphi_{\alpha}(r) \varphi^*_i(r) P^{(2)}_{ai}(\omega) + \varphi_i(r) \varphi^*_\alpha(r) P^{(2)}_{ia}(\omega) \right],
\]

\[
\delta q_{\alpha,s}(\omega) = L^{(1)}_{\alpha,-}(\omega) + L^{(1)}_{\alpha,+}(\omega),
\]

\[
\delta q_{\alpha,s}(\omega) = L^{(2)}_{\alpha,-}(\omega) + L^{(2)}_{\alpha,+}(\omega),
\]

where we used the Lehmann spectral representation of \(\chi^{q_{\alpha},s}_{\alpha}(r', \omega)\) and \(\chi^{q_{\alpha},s}_{\alpha,\alpha}(\omega)\) and we define \(P^{(\nu)}_{\alpha,-}, P^{(\nu)}_{\alpha,+}\) and \(L^{(\nu)}_{\alpha,-}\) and \(L^{(\nu)}_{\alpha,+}\) with \(\nu = 1, 2\) as

\[
[w - \omega_{i\alpha}] P^{(1)}_{ai}(\omega) = \int dr \varphi_{\alpha}(r) \varphi^*_i(r) \delta v_{s,1}(r, \omega),
\]

\[
[w + \omega_{i\alpha}] P^{(1)}_{ia}(\omega) = - \int dr \varphi_{\alpha}(r) \varphi^*_i(r) \delta v_{s,1}(r, \omega),
\]

\[
[w - \omega_{i\alpha}] P^{(2)}_{ai}(\omega) = \int dr \varphi_{\alpha}(r) \varphi^*_i(r) \delta v_{s,2}(r, \omega),
\]

\[
[w + \omega_{i\alpha}] P^{(2)}_{ia}(\omega) = - \int dr \varphi_{\alpha}(r) \varphi^*_i(r) \delta v_{s,2}(r, \omega),
\]

\[
[w - \omega] L^{(1)}_{\alpha,-}(\omega) = \frac{1}{2\omega^2_{\alpha}} \delta j_{\alpha,1}(\omega),
\]

\[
[w + \omega] L^{(1)}_{\alpha,+}(\omega) = - \frac{1}{2\omega^2_{\alpha}} \delta j_{\alpha,1}(\omega),
\]

\[
[w - \omega] L^{(2)}_{\alpha,-}(\omega) = \frac{1}{2\omega^2_{\alpha}} \delta j_{\alpha,2}(\omega),
\]

\[
[w + \omega] L^{(2)}_{\alpha,+}(\omega) = - \frac{1}{2\omega^2_{\alpha}} \delta j_{\alpha,2}(\omega),
\]
where $\omega_{ai} = (\epsilon_a - \epsilon_i)$ and the respective effective potentials and currents ($\delta v_{s,v}(r,\omega)$, $j_{s,v}^\alpha(\omega)$) as
\[
\delta v_{s,1}(r,\omega) = \delta v(r,\omega) + \int dr' f_{Mxc}^n(r, r', \omega) \delta n_e(r', \omega) + \sum_a f_{Mxc}^n(r, \omega) \delta q_{a,v}(\omega),
\]
\[
\delta v_{s,2}(r,\omega) = \int dr' f_{Mxc}^n(r, r', \omega) \delta n_j(r', \omega) + \sum_a f_{Mxc}^n(r, \omega) \delta q_{a,j,v}(\omega),
\]
\[
\delta j_{\alpha,1}^s(\omega) = \int dr' g_{Mxc}^n(r, r', \omega) \delta n_e(r', \omega),
\]
\[
\delta j_{\alpha,2}^s(\omega) = \delta j_a(\omega) + \int dr' g_{Mxc}^n(r, r', \omega) \delta n_j(r', \omega).
\]

As stated above, the Kohn-Sham scheme of QEDFT decouples the interacting system such that the responses are paired as $(\delta n_e(r,\omega), \delta q_{a,v}(\omega))$ due to $\delta v(r,\omega)$ and $(\delta n_j(r,\omega), \delta q_{a,j,v}(\omega))$ due to $\delta j_a(\omega)$. Therefore, substituting Eqs. (C13) and (C15) into Eqs. (C5) and (C6) and Eqs. (C9) and (C10) and after some simplification, we obtain
\[
\sum_{j,b} [\delta_{ab} \delta_{ij} (\omega_{ai} - \omega) + K_{ai,jb}(\omega)] P_{bj}^{(1)}(\omega) = -v_{ai}(\omega),
\]
\[
\sum_{j,b} [\delta_{ab} \delta_{ij} (\omega_{ai} + \omega) + K_{ia,jb}(\omega)] P_{bj}^{(1)}(\omega) = -v_{ia}(\omega),
\]
\[
[\omega - \omega_a] L_{\alpha,-}^{(2)}(\omega) + \sum_{j,b} [N_{\alpha,jb} P_{bj}^{(1)}(\omega) + N_{a,bj} P_{bj}^{(1)}(\omega)] = 0,
\]
\[
[\omega + \omega_a] L_{\alpha,+}^{(2)}(\omega) + \sum_{j,b} [N_{\alpha,jb} P_{bj}^{(1)}(\omega) + N_{a,bj} P_{bj}^{(1)}(\omega)] = 0.
\]

Also, substituting Eqs. (C14) and (C16) into Eqs. (C7) and (C8) and Eqs. (C11) and (C12) and after some simplification, we obtain
\[
\sum_{j,b} \delta_{ab} \delta_{ij} [((\omega_{ai} - \omega) + K_{ai,jb}(\omega)) P_{bj}^{(2)}(\omega) + K_{ai,jb}(\omega) P_{bj}^{(2)}(\omega) + \sum_{\alpha} M_{\alpha,jb}(\omega) \left[L_{\alpha,-}^{(2)}(\omega) + L_{\alpha,+}^{(2)}(\omega)\right]] = 0,
\]
\[
\sum_{j,b} \delta_{ab} \delta_{ij} [((\omega_{ai} + \omega) + K_{ia,jb}(\omega)) P_{ia}^{(2)}(\omega) + K_{ia,jb}(\omega) P_{ia}^{(2)}(\omega) + \sum_{\alpha} M_{\alpha,jb}(\omega) \left[L_{\alpha,-}^{(2)}(\omega) + L_{\alpha,+}^{(2)}(\omega)\right]] = 0,
\]
\[
[\omega - \omega_a] L_{\alpha,-}^{(2)}(\omega) + \sum_{j,b} [N_{\alpha,jb} P_{bj}^{(2)}(\omega) + N_{a,bj} P_{bj}^{(2)}(\omega)] = -\frac{1}{2\omega_a^2} \delta j_a(\omega),
\]
\[
[\omega + \omega_a] L_{\alpha,+}^{(2)}(\omega) + \sum_{j,b} [N_{\alpha,jb} P_{bj}^{(2)}(\omega) + N_{a,bj} P_{bj}^{(2)}(\omega)] = -\frac{1}{2\omega_a^2} \delta j_a(\omega),
\]

where we defined the coupling matrices
\[
K_{ai,jb}(\omega) = \iint dr dr' \phi_i(r) \phi_j^*(r) f_{Mxc}^n(r, y, \omega) \phi_b(y) \phi_j^*(y),
\]
\[
v_{ia}(\omega) = \int dr \phi_i^*(r) \delta v(r, \omega) \phi_a(r),
\]
\[
M_{\alpha,ai}(\omega) = \int dr \phi_i(r) \phi_a^*(r) f_{Mxc}^n(r, \omega),
\]
\[
N_{\alpha,ia} = \frac{1}{2\omega_a^2} \int dr \phi_i^*(r) \phi_a(r) g_{Mxc}^n(r, \omega),
\]
and
\[
\phi_i(r) = \phi_i(x, y), \quad \phi_a(r) = \phi_a(x, y).
\]
The coupling matrix $N_{\alpha,\alpha}$ has no frequency dependence since this is just the mean-field kernel of the photon modes. We now introduce the following abbreviations $L(\omega) = \delta_{ij}\delta_{jj}(\epsilon_a - \epsilon_i) + K_{\alpha,i,j}(\omega), \ K(\omega) = K_{\alpha,i,j}(\omega), \ M(\omega) = M_{\alpha,i,j}(\omega), \ N = N_{\alpha,\alpha}, \ X_1(\omega) = \ P^{(1)}(\omega), \ Y_1(\omega) = \ P^{(2)}(\omega), \ X_2(\omega) = \ P^{(3)}(\omega), \ Y_2(\omega) = \ P^{(4)}(\omega), \ A_1(\omega) = \ L^{(1)}_{\alpha,i,j}(\omega), \ B_1(\omega) = \ L^{(2)}_{\alpha,i,j}(\omega), \ A_2(\omega) = \ L^{(3)}_{\alpha,i,j}(\omega), \ B_2(\omega) = \ L^{(4)}_{\alpha,i,j}(\omega), \ V(\omega) = v_{\alpha,i}(\omega), \ J(\omega) = -\frac{1}{2}\omega^2\delta_{\alpha,j}(\omega)$.

Using these notations, we cast Eqs. (C.17)-(C.20) and Eqs. (C.21)-(C.24) into two matrix equations given by

\begin{equation}
\begin{pmatrix}
L(\omega) & K(\omega) & M(\omega) & M(\omega) \\
K^*(\omega) & L(\omega) & M^*(\omega) & M^*(\omega) \\
\alpha & \alpha & \omega & 0 \\
N & N^* & \alpha & 0
\end{pmatrix} + \omega \begin{pmatrix}
1 & 0 & 0 & 0 \\
0 & 1 & 0 & 0 \\
0 & 0 & -1 & 0 \\
0 & 0 & 0 & 1
\end{pmatrix} \begin{pmatrix}
X_1(\omega) \\
Y_1(\omega) \\
A_1(\omega) \\
B_1(\omega)
\end{pmatrix} = \begin{pmatrix}
V(\omega) \\
V^*(\omega) \\
0 \\
0
\end{pmatrix}
\end{equation}

Next, we argue that the right hand side of the above matrices remains finite as the frequency $\omega$ approaches the exact excitation frequencies $\omega \rightarrow \Omega_q$ of the interacting system while the density and displacement field responses on the left hand side have poles at the true excitation frequencies $\Omega_q$. This allows us to cast Eq. (C.29) and Eq. (C.30) into an eigenvalue problem.

\begin{equation}
\begin{pmatrix}
L(\Omega_q) & K(\Omega_q) & M(\Omega_q) & M(\Omega_q) \\
K^*(\Omega_q) & L(\Omega_q) & M^*(\Omega_q) & M^*(\Omega_q) \\
\alpha & \alpha & \omega & 0 \\
N & N^* & \omega & 0
\end{pmatrix} \begin{pmatrix}
X_1(\Omega_q) \\
Y_1(\Omega_q) \\
A_1(\Omega_q) \\
B_1(\Omega_q)
\end{pmatrix} = \Omega_q \begin{pmatrix}
1 & 0 & 0 & 0 \\
0 & -1 & 0 & 0 \\
0 & 0 & 1 & 0 \\
0 & 0 & 0 & -1
\end{pmatrix} \begin{pmatrix}
X_1(\Omega_q) \\
Y_1(\Omega_q) \\
A_1(\Omega_q) \\
B_1(\Omega_q)
\end{pmatrix}
\end{equation}

\begin{equation}
\begin{pmatrix}
L(\Omega_q) & K(\Omega_q) & M(\Omega_q) & M(\Omega_q) \\
K^*(\Omega_q) & L(\Omega_q) & M^*(\Omega_q) & M^*(\Omega_q) \\
\alpha & \alpha & \omega & 0 \\
N & N^* & \omega & 0
\end{pmatrix} \begin{pmatrix}
X_2(\Omega_q) \\
Y_2(\Omega_q) \\
A_2(\Omega_q) \\
B_2(\Omega_q)
\end{pmatrix} = \Omega_q \begin{pmatrix}
1 & 0 & 0 & 0 \\
0 & -1 & 0 & 0 \\
0 & 0 & 1 & 0 \\
0 & 0 & 0 & -1
\end{pmatrix} \begin{pmatrix}
X_2(\Omega_q) \\
Y_2(\Omega_q) \\
A_2(\Omega_q) \\
B_2(\Omega_q)
\end{pmatrix}
\end{equation}

It is convenient to cast Eqs. (C.31) and (C.32) into a Hermitian eigenvalue problem which is given by

\begin{equation}
\begin{pmatrix}
U & V \\
W & \omega^2
\end{pmatrix} \begin{pmatrix}
E_1 \\
P_1
\end{pmatrix} = \Omega_q^2 \begin{pmatrix}
E_1 \\
P_1
\end{pmatrix}
\end{equation}

\begin{equation}
\begin{pmatrix}
U & V \\
W & \omega^2
\end{pmatrix} \begin{pmatrix}
E_2 \\
P_2
\end{pmatrix} = \Omega_q^2 \begin{pmatrix}
E_2 \\
P_2
\end{pmatrix}
\end{equation}

where we assumed real-valued orbitals, i.e., $K = K^*$, $M = M^*$ and $N = N^*$, and the matrices are given by $U = (L - K)^{1/2}(L + K)(L - K)^{1/2}, \ V = 2(L - K)^{1/2}N^{1/2}\omega^{1/2}, \ W = 2\omega^{1/2}N^{1/2}M^{1/2}(L - K)^{1/2}$, and the eigenvectors are $E_1 = N^{1/2}(L - K)^{-1/2}(X_1 + Y_1)$ and $P_1 = M^{1/2}\omega_{\alpha}^{-1/2}(A_1 + B_1)$. The pseudo-eigenvalue problem of Eqs. (C.33) and (C.34) is the final form of QEDFT matrix equation for obtaining exact excitation frequencies and oscillator strengths.

### Appendix D: Oscillator Strengths

In this section, we derive the oscillator strengths resulting from the eigenvectors of the pseudo-eigenvalue problem of Eqs. (C.33) and (C.34). Multiplying out Eq. (C.29), we write the matrix equation in the form

\begin{equation}
(L + K)(X_1 + Y_1) + 2M(A_1 + B_1) - \omega(X_1 - Y_1) = -2v, \\
(L - K)(X_1 - Y_1) - \omega(X_1 + Y_1) = 0, \\
2N(X_1 + Y_1) + \omega\alpha(A_1 + B_1) - \omega(A_1 - B_1) = 0, \\
\omega\alpha(A_1 - B_1) - \omega(A_1 + B_1) = 0.
\end{equation}
From here on we set $S = (L - K)$, the above pair of equations now becomes

$$S(L + K)E_1 + 2SMP_1 - \omega^2E_1 = -2S\nu,$$
$$2\omega_\alpha NE_1 + \omega_\alpha^2P_1 - \omega^2P_1 = 0.$$ 

This can be written in matrix form as

$$\begin{bmatrix} 2S(L + K) & 2SM \omega_\alpha^2 \\ 2\omega_\alpha N & \omega_\alpha^2 \end{bmatrix} - \omega^2 \begin{bmatrix} 1 & 0 \\ 0 & 1 \end{bmatrix} \begin{bmatrix} E_1 \\ P_1 \end{bmatrix} = \begin{bmatrix} 2S\nu \\ 0 \end{bmatrix},$$

where $E_1 = X_1 + Y_1$ and $P_1 = A_1 + B_1$. We perform the same steps as above to make the nonlinear eigenvalue problem Hermitian and obtain

$$[C - \omega^2I]\begin{bmatrix} N^{1/2}s^{-1/2}E_1 \\ M^{1/2}s^{-1/2}P_1 \end{bmatrix} = -\begin{bmatrix} 2N^{1/2}s^{1/2}\nu \\ 0 \end{bmatrix},$$

where $C = \begin{bmatrix} U & V \\ W & \omega_\alpha^2 \end{bmatrix}$. We determine the vectors given as

$$E_1 = -2S^{1/2}[C - \omega^2I]^{-1}S^{1/2}\nu,$$
$$P_1 = -2\omega_\alpha^{1/2}M^{-1/2}[C - \omega^2I]^{-1}N^{1/2}s^{1/2}\nu.$$ 

When $Z_I$ is normalized, we can use the spectral expansion to get

$$[C - \omega^2I]^{-1} = \sum_I \frac{Z_I[Z_I^\dagger N^{1/2}s^{1/2}]}{(\Omega_I^2 - \omega^2)}.$$ 

where $Z_I = \begin{bmatrix} E_{1I} \\ P_{1I} \end{bmatrix}$.

1. **Oscillator strength for the photon-matter response function**

Next, we substitute the expression of the spectral expansion Eq.(D5) in Eq.(D4) and by substituting $P_1$ in Eq.(C2) yields

$$\deltaq_{\alpha, I}(\omega) = \sum_I \left\{ \frac{2\omega_\alpha^{1/2}M^{-1/2}Z_I[Z_I^\dagger N^{1/2}s^{1/2}]}{(\omega^2 - \Omega_I^2)} \right\} v(\omega).$$

The oscillator strength is given by

$$f_{np}^I = 2\omega_\alpha^{1/2}M^{-1/2}Z_I[Z_I^\dagger N^{1/2}s^{1/2}].$$ 

where $r_{\mu} = \int dr r_{\mu} \sum_{I, I} \Phi_{\mu}^{*}(r)$ and $\mu, \nu = 1, 2, 3$. It can be verified that $r_{\mu}^I S^{1/2}Z_I = 1/2 \langle \Psi_0 | r_{\mu} | \Psi_I \rangle$.

2. **Oscillator strength for the matter-photon response function**

Following similar steps as above with Eq.(C30) we obtain

$$E_2 = -2S^{1/2}N^{-1/2}[C - \omega^2I]^{-1}M^{1/2}\omega_\alpha^{1/2}J_\alpha,$$
$$P_2 = -2\omega_\alpha^{1/2}C^{-1/2}[C - \omega^2I]^{-1}\omega_\alpha^{1/2}J_\alpha.$$ 

where $J_\alpha(\omega) = \frac{1}{\omega_\alpha} J_\alpha(\omega)$. By substituting the spectral expansion Eq.(D5) in $E_2$ and further substituting in Eq.(C3) yields

$$\deltan_{\alpha}(r, \omega) = -2\sum_{i, I} \frac{\Phi_{ii} S^{1/2}N^{-1/2}Z_I[Z_I^\dagger M^{1/2}\omega_\alpha^{1/2}\Phi_{\alpha I}^*]}{(\Omega_I^2 - \omega^2)} J_\alpha(\omega),$$

Following a similar procedure as above, we have

$$\deltan_{\alpha}(r, \omega) = \sum_I \left\{ \frac{2\Phi S^{1/2}N^{-1/2}Z_I[Z_I^\dagger M^{1/2}\omega_\alpha^{1/2}\Phi^*]}{(\omega^2 - \Omega_I^2)} \right\} J_\alpha(\omega),$$

where $\Phi = \sum_{i, \alpha} \Phi_{ii}^*$ and the oscillator strength is given by

$$f_{np}^I = 2\Phi S^{1/2}N^{-1/2}Z_I[Z_I^\dagger M^{1/2}\omega_\alpha^{1/2}\Phi].$$

3. **Oscillator strength for the photon-photon response function**

We define a collective photon coordinate for the $\alpha$ modes $Q = \sum_{\alpha} q_{\alpha}$ (in analogy with $R = \sum_{i} r_{i}$). By directing perturbing the photon field through the photon coordinate with an external charge current $j_{\alpha}(\omega)/\omega_{\alpha}$, we induce a polarization of the field of mode $\alpha$ which we denote as $Q_{\alpha}(\omega) = \beta_{\alpha}(\omega)j_{\alpha}(\omega)/\omega_{\alpha}$. Where $\beta_{\alpha}(\omega)$ is the polarizability of field in the $\alpha$ mode. To first-order, the collective coordinate is given by

$$\delta Q(t) = \sum_{\alpha} \delta \alpha, t_{\alpha}(t).$$ 

The field polarizability in frequency space can be written as

$$\beta_{\alpha}(\omega) = \sum_{\alpha'} \delta \alpha, \alpha' \omega_{\alpha'}/\omega_{\alpha}.$$ 

By substituting Eq.(D8) in Eq.(C4) and using the spectral expansion yields

$$\beta_{\alpha}(\omega) = \sum_{I} \frac{2\omega_\alpha^{1/2}Z_I[Z_I^\dagger N^{1/2}s^{1/2}]}{(\Omega_I^2 - \omega^2)}.$$ 

Eq.(D3) is the field polarizability analogous to the atomic polarizability tensor of Eq.(52).
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